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Laser Program **Annual Report ~1979**

Volume 3

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Laser Program **Annual Report - 1979**

Volume 3

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Advanced Quantum Electronics

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Introduction

The broad objectives of the LLL advanced laser effort are to identify, demonstrate, and assess laser driver systems for use in laser-fusion power reactors. A further aspect of this effort is to participate in program activities directed toward demonstration of the effectiveness of selected advanced laser systems for pellet implosions. In carrying out assessments of candidate laser systems over the past four years, we have assembled an integrated capability to conduct experimental, theoretical, and computational studies spanning the relevant aspects of fusion laser systems, including atomic and molecular collision physics, quantum electronic processes, and device and systems technologies.

The technical performance requirements placed on candidate laser systems (energy, power, wavelength efficiency, etc.) were derived initially from computer simulations of pellet implosions. These requirements are continually being refined as we obtain additional experimental pellet-implosion data and improved computer simulations. On the basis of these sources of guidance, our advanced laser driver effort has focused throughout its history on laser systems operating at relatively short wavelengths (0.25 to 2 μm). Current estimates of laser-driver performance requirements for fusion reactors include an efficiency in excess of five percent (at 250 nm), several megajoules of pulse energy, and pulse widths of 10 to 20 ns.

The programmatic and technical basis of our 1979 effort is a natural evolution of the previous year's activities, which were directed along two complementary paths:

- Research, development and further assessment of hybrid (angle multiplexing and Raman) pulse-compressor laser systems as a baseline activity.
- A renewed search for new laser media and pumping techniques capable of resulting in laser driver systems operating with efficiencies in excess of that projected for the baseline system (5%) and which are either less costly or less complex technologically, or both.

Our 1979 progress report begins with a section titled "Pulse-Compressor Laser Systems" where we outline technical activities and results in pulse compressor laser research and development. Topics include scaling of KrF excimer power amplifiers, scaling and limitations of Raman pulse-compression systems, experimental results on Raman and angle multiplexing using the Raman Amplifier Pumped by Intensified Excimer Radiation (RAPIER) pulse-compression laser system testbed, and a conceptual design of a MJ-class hybrid pulse-compressor system.

In the next section, "Advanced Concepts and Supporting Research," we present material for other potentially advanced laser candidates. First, preliminary analyses and conceptual designs for two new, advanced laser candidates are described. These are the free-electron laser (FEL) configured in a single-pass high-intensity (10^{12} W) geometry which is compatible with a fusion reactor vessel and a flashlamp-pumped high-average-power solid state laser based on vanadium-doped magnesium fluoride (V:MgF₂) as the active medium. Next, we present technical data that document our findings on Group VI laser media which have been studied by the ICF laser community during the past few years. We also outline alternative methods for producing population inversion in atomic iodine.

This is followed with a section on "Laser Theory and Design," wherein we describe developments in our laser theory and kinetics capabilities which are used to provide

technical perspective for efforts on both baseline and advanced concept laser systems. Subjects selected for review here include basic experimental results on the kinetics of rare-gas halides which contribute to computer modeling capabilities, new developments in methods for characterizing parasitic modes in high-gain laser media, and a series of theoretical developments treating kinetic processes important in rare-gas-halide laser media.

Our concluding discussion, "Advances in Atomic and Molecular Theory," provides a description of the development of several basic methods in computational quantum chemistry which broaden our ability to anticipate the microscopic behavior of electrically and optically excited gaseous pump and laser media relevant to fusion laser research and development.

Authors: W. F. Krupke, E. V. George, and L. D. Pleasance

Pulse Compressor Laser Systems

As set forth previously in the context of performance requirements for the driver system, UV excimer lasers command considerable interest. Several of the rare-gas-halide (RGH) lasers, such as KrF at 248 nm, are efficient and scalable to large energies. However, these lasers do not store appreciable energy and must be operated as power amplifiers. The pulse length for efficient operation is considerably longer than required for fusion applications. Thus, some technique to compress the laser system output pulse width must be identified and developed to exploit the otherwise attractive features of excimer laser systems.

One approach is to extract energy from an amplifier pumped with a long pulse consisting of a continuous train of several short pulses. These short pulses propagate sequentially through the amplifier at slightly different angles with respect to each other. Such angle-coded, time-sequenced pulses can then be applied simultaneously to a target by adding appropriate optical delays. Another technique for compressing a long output pulse from the excimer laser is backward-wave Raman scattering. The advantages, limitations, and status of this pulse compression technique will be discussed later in "Raman Compressors and Amplifiers." One can also employ a hybridization of both these concepts. That is, combine pulse stacking with Raman pulse compression in an effort to realize efficient, scalable laser systems with high overall compression factors. This minimizes the physics and technological limitations of each of the techniques when utilized separately.

Experimental and analytical studies on the hybrid pulse compressor are the main thrust of our

RAPIER program. Progress on this activity is discussed in "RAPIER Experimental Program." Finally, a point design for a MJ-class laser system that utilizes the hybrid Raman compressor pulse-stacking technique has been developed. A summary of the results of this system design is given at the end of this section.

KrF Amplifiers

Several factors determine the performance of large-scale KrF amplifiers. The output flux for a particular amplifier is dependent on the KrF production efficiency and lifetime, extraction efficiency, and power gain. The aperture and power output of large amplifiers are limited by amplified spontaneous emission and parasitic oscillations. The prediction of detailed laser performance requires a comprehensive integrated computer model. Such a code has been developed over the last few years at LLL. This section will describe some of the inputs to this code and presents results applied to large scale amplifier design.

Kinetics of Ar/Kr/F₂ Laser Mixtures. The basic kinetics of Ar/Kr/F₂ laser mixtures were well understood by 1978 and are described by Rokni, et al.¹ A schematic diagram of the kinetic chain leading to KrF and ultimately to Kr₂F is shown in Fig. 7-1. The links in the chain most important for the mixtures and pressures appropriate to electron-beam pumped lasers are drawn with heavy lines. The KrF molecule is formed primarily via the ionic recombination reaction,

for which theoretical rate coefficients are given by Flannery and Yang,² based on the theory of Bates and Flannery³ and refinements by Flannery.⁴ The

Electron beam

Secondary electrons

F^-

F_2

Ar

Kr

Kr^+

Kr_2^+

Ar^+

Ar_2^+

ArF^+

Ar_2F^+

$ArKr^+$

KrF^+

$ArKrF^+$

Kr_2F^+

Products

Reactions shown:

- $Secondary\ electrons + F_2 \rightarrow F^-$
- $Secondary\ electrons + Ar \rightarrow Ar^+$
- $Secondary\ electrons + Kr \rightarrow Kr^+$
- $Kr^+ + Kr \rightarrow Kr_2^+$
- $Ar^+ + 2 Ar \rightarrow Ar_2^+$
- $Ar_2^+ + F_2 \rightarrow ArF^+$
- $Ar_2^+ + Kr \rightarrow ArKr^+$
- $ArF^+ + 2 Ar \rightarrow Ar_2F^+$
- $ArKr^+ + F^- + Ar \rightarrow KrF^+$
- $ArF^+ + Kr \rightarrow KrF^+$
- $Ar_2F^+ + Kr \rightarrow KrF^+$
- $Kr_2^+ + F^- + Ar \rightarrow KrF^+$
- $KrF^+ + 2 Ar \rightarrow ArKrF^+$
- $ArKrF^+ + Kr \rightarrow Kr_2F^+$
- $Kr_2F^+ + F_2, RG \rightarrow Products$
- $KrF^+ + F_2 \rightarrow Products$
- $KrF^+ \rightarrow Products$ (wavy arrow)

$$\text{KrF} + \text{Ar} + \text{Kr} \rightarrow \text{Kr}_2\text{F} + \text{Ar}$$

7-3

Table 7-1. Rate constants for kinetic processes in rare-gas-halide mixtures.

Ar/F ₂ Kinetic rate coefficients		
	This work ^a	Literature ^a
Ar ₂ ⁺ + F ⁻ (+ Ar) → ArF* + Ar (+ Ar)	0.3 × FY ^b	FY ^b
ArF* + Ar + Ar → Ar ₂ F* + Ar	2.0 × 10 ⁻³¹	4.0 × 10 ⁻³¹
Ar ₂ F* + F ₂ → Products	4.0 × 10 ⁻¹⁰	2.1 × 10 ⁻¹⁰
Ar ₂ F* + Ar → Products	1.2 × 10 ⁻¹³	
Kr/F ₂ Kinetic rate coefficients		
	This work	Literature
Kr ⁺ + F ⁻ + Kr → KrF* + Kr	0.25 × FY ^b	FY ^b
Kr ₂ ⁺ + F ⁻ (+ Kr) → KrF* + Kr (+ Kr)	0.25 × FY ^b	FY ^b
KrF* + Kr + Kr → Kr ₂ F* + Kr	1.0 × 10 ⁻³¹	2.9 - 9.7 × 10 ⁻³¹
Kr ₂ F* + F ₂ → Products	1.5 × 10 ⁻¹⁰	4.3 × 10 ⁻¹⁰
Kr ₂ F* + Kr + Kr → Products	~1.0 × 10 ⁻³³	
Ar/Kr/F ₂ Kinetic rate coefficients		
	This work	Literature
Ar* + F ₂ → ArF* + F	4.0 × 10 ⁻¹⁰	4.0 - 10.0 × 10 ⁻¹⁰
ArF* + Kr → KrF* + Ar	3.0 × 10 ⁻¹⁰	1.6 × 10 ⁻⁹
Ar ₂ F* + Kr → KrF* + 2Ar	~6.0 × 10 ⁻¹¹	
KrF* + Ar + Ar → ArKrF* + Ar	4.0 × 10 ⁻³²	7.0 × 10 ⁻³²
ArKrF* + Kr → Kr ₂ F* + Ar	6.0 × 10 ⁻¹¹	
KrF* + Kr + Ar → Kr ₂ F* + Ar	8.0 × 10 ⁻³¹	6.5 × 10 ⁻³¹
Ar ₂ ⁺ + Kr → Kr ₂ ⁺ + 2Ar	7.5 × 10 ⁻¹⁰	6.0 - 7.5 × 10 ⁻¹⁰
Kr ₂ ⁺ + F ⁻ (+ Ar) → KrF* + Kr + Ar	0.7 × FY ^b	FY ^b
ArKr ⁺ + Kr → Kr ₂ ⁺ + Ar	1.0 × 10 ⁻¹⁰	3.2 × 10 ⁻¹⁰
ArKr ⁺ + F ⁻ (+ Ar) → KrF* (+ Ar)	0.5 × BF ^c	

^aUnits for rate constants are cm³/sec for two-body and cm⁶/sec for three-body processes.
^bFY, calculations of M.R. Flannery and T.P. Yang, APL, 33, 574 (1978).
^cBF, theory of D.R. Bates and M.R. Flannery, J. Phys. B 2, 184 (1969).

the afterglow of a short-pulse, low-current electron beam. (For further information, see "LAMP KrF Kinetics Experiments and Analysis.") These data have been used to derive a consistent set of rate coefficients for krypton fluoride kinetics. These rate coefficients and representative values from the literature are shown in Table 7-1. When used in the kinetic model, the rates of Morgan and Szoke give excellent agreement with their own experimental data and with the data of other experiments as well. Figure 7-2 shows the relative KrF fluorescent intensity in an experiment by Rokni, et al.¹ for Ar-Kr/F₂ mixtures containing 0.3% F₂ and 1% and 15% Kr over a pressure range of 0.25 to 4.0 atm. The

electron-beam current density is 10 A/cm². The calculations are normalized to the experiment at 1% Kr and 1 atm total pressure and, as can be seen, the agreement is excellent over the whole pressure range for both Kr fractions. These results give us reason to have confidence in the predictive capabilities of our models.

There has been much discussion recently of three more detailed problems in the understanding of KrF kinetics:

- The extent of mixing of the B-, C-, and D-states of KrF.
- The rate of electron-impact quenching of KrF.
- The effect of elevated gas temperature on laser performance.

Fig. 7-2. Comparison of kinetic model results with the experimental data of J. A. Mangano, J. H. Jacob, M. Rokni, and A. Hawryluk, *Appl. Phys. Lett.*, **31** 26 (1977).

As the laser operates on the B-X transition, it is of interest to know the mixing rate of the B- and C-states. The B- and C-ionic states of KrF lie in close proximity, the estimated separation being 0.08 eV; the D-state lies about 0.7 eV higher in energy.⁶ Calculations indicate that the B-state lies lowest in energy.^{6,7} However, there is experimental evidence to indicate the C-state may indeed lie lowest.^{8,9} To simulate the fully mixed case in a laser energy extraction calculation, one divides the KrF(B) stimulated-emission cross section by two. Experiments by Pummer and Hohla¹⁰ in a discharge at 2 atm indicate that the B-C mixing time is less than 1 ns and that the B-, C-, and D-collisional coupling time is greater than 20 ns. This indicates that the D-state is directly populated by chemical reaction; no energy is extracted from it. The electron-quenching rate for KrF will reduce the effective KrF lifetime at high-power deposition rates. With an electron-beam power deposition of 1 MW/cm³, the electron density is expected to be about 10¹⁵/cm³. Recent Born- and impact-parameter calculations by Hazi et al.¹¹ give an electron KrF(B) superelastic quenching-rate coefficient of 3×10^{-8} cm³/sec that is nearly independent of electron temperature. Jacob and Trainor¹² have extracted a total KrF deexcitation rate coefficient of 2.2×10^{-7} cm³/sec from an ex-

periment. However, this is not a direct measurement and is somewhat model-dependent.

It is expected that laser performance will be enhanced at modestly elevated gas temperatures.¹³ Our computer model indicates that, although the ionic recombination rate decreases as the gas temperature is increased, the rate of three body quenching of KrF will decrease even more for low pump rates. This results in a net improvement in performance. Additionally, the dimer ion absorption cross section is reduced at higher temperatures. It is expected that there will be experimental results on the effect of gas temperature on performance in the near future.¹⁴ There has also been evidence that the use of Ne as an additive enhances performance¹⁵ despite the strong absorption of Ne⁺₂ at 248 nm. More study of this needs to be made before anything definite can be stated.

In addition to the kinetic processes responsible for the creation and destruction of KrF, photoabsorption processes are important to the performance of the laser. The most important absorbers, and their cross sections, are shown in Table 7-2. These loss processes along with the kinetic processes described above will determine the optimum

Table 7-2. Cross sections for photoabsorption of KrF excimer radiation.

Species	Cross section (cm^2)
F_2	6.6×10^{-20}
F^*	5.6×10^{-18}
Ar^{**}	2.0×10^{-18}
Kr^{**}	1.9×10^{-18}
Ar_2^+	1.3×10^{-17}
Kr_2^+	1.1×10^{-18}
$\text{Kr}_2^{I,*}$ (saturable)	1.6×10^{-18}

operating conditions for the KrF laser in terms of Ar:Kr:F₂ fractions, total pressure, and electron-beam power deposition.

Amplified Spontaneous Emission and Parasitic Oscillations. The scaling of KrF laser media to large energies while maintaining high overall efficiency is determined by several factors:

- Amplified spontaneous emission (ASE) and control of parasitic oscillations.
- Extraction efficiency.
- Fluorine depletion.
- Electron-beam pump technology scalability.

In this section we discuss the energy and efficiency scaling of KrF amplifiers as determined by ASE, parasitic oscillations, extraction efficiency and fluorine depletion considerations. The constraints imposed by electron-beam pump technology limitations, although very important, are not considered.

As shown schematically in Fig. 7-3, several KrF amplifier extraction configurations are possible. In the conventional unidirectional or single-pass configuration, Fig. 7-3(a), the signal to be amplified is injected into one end of the amplifier. This signal is amplified as it passes through and exits the amplifier at the end opposite the input. The main disadvantage of the unidirectional amplifier is that the flux at the input aperture is relatively low and the gain is correspondingly high. This region of the amplifier is therefore susceptible to depumping by ASE and transverse parasitic oscillations that can seriously reduce amplifier efficiency. Of course, the gain at the input to the unidirectional amplifier can be decreased by increasing the intensity of the input field. However, as the input intensity is increased the power gain of the amplifier is reduced since in KrF nonsaturable losses limit the intensity of the

Fig. 7-3. Some possible KrF amplifier laser energy extraction configurations.

field for efficient extraction. The major limitations of the unidirectional amplifier can be overcome by using bidirectional, or two-pass, amplifier extraction configurations. These are illustrated in Fig. 7-3(b) and (c). In the bidirectional amplifier, the gain is more uniform since the input and output beams overlap. Next, we consider bidirectional amplifiers in more detail because of their superior gain control and scaling characteristics.

In general, when ASE effects are included, the extraction characteristics of bidirectional amplifiers are determined by the following dimensionless parameters:

- The net small-signal gain to nonsaturable loss coefficient ratio, g_0/γ_{NS} .
- The nonsaturable-loss coefficient-amplifier length product, $\gamma_{NS}L$.
- The input intensity normalized by the saturation intensity, I_i/I_s .
- The KrF molecule lifetime to radiative lifetime ratio, τ_1/τ_R .
- The amplifier radius-to-length, or aspect, ratio, a/L .

For further details, see "Analysis of Parasitic Oscillations" and "ASE in Bidirectional Amplifiers."

Once these parameters are specified, the extraction dynamics of the amplifier are fully determined. To include the effects of parasitic oscillations, other parameters, such as wall reflectivities, must be considered. Figures 7-4 and 7-5 illustrate the performance characteristics of bidirectional amplifiers for two representative values of the gain-to-loss

Fig. 7-4. Laser energy extraction characteristics of bidirectional amplifiers including ASE depumping. The basic laser parameter values are $g_0/\gamma_{NS} = 10$, $\gamma_{NS}L = 1$ and $\tau_L/\tau_R = 0.33$. The quantity, a/L , is the amplifier radius-to-length ratio. The ideal unidirectional amplifier is a single pass amplifier in which ASE depumping is not important, i.e., $a/L = 0$.

Fig. 7-5. Laser energy extraction characteristics of bidirectional amplifiers, including ASE depumping. The basic laser parameter values are $g_0/\gamma_{NS} = 20$, $\gamma_{NS}L = 1$ and $\tau_L/\tau_R = 0.33$. The quantity, a/L , is the amplifier radius-to-length ratio. The ideal unidirectional amplifier is a single pass amplifier in which ASE depumping is not important i.e., $a/L = 0$.

parameter, $g_0/\gamma_{NS} = 10$ and 20. The multiple curves in each case show the parametric dependence of amplifier performance on aspect ratio. As the aspect ratio is increased, the amplifier geometry evolves from a highly rectangular shape, $a/L = 0.05$, to an essentially cubic one, $a/L = 0.45$. At

higher aspect ratios, ASE depumping becomes more important. To overcome the serious energy losses due to ASE depumping for large aspect ratio amplifiers, the input intensity must exceed several tenths of a saturation flux. Figures 7-4 and 7-5 show that in large aperture devices performance

comparable to that of an ideal ($\text{ASE} = 0$) unidirectional amplifier can be achieved while maintaining relatively high extraction efficiency ($\eta_{\text{EXT}} \approx 0.4$ to 0.5) and power gain ($I_0/I_i \lesssim 10$). The severe penalty on extraction efficiency (roughly a factor of two) imposed by nonsaturable photoabsorption loss is

Fig. 7-6. Axial normalized loaded-gain, g/g_0 , distributions for bidirectional amplifiers including ASE depumping. The basic laser parameter values are $g_0/\gamma_{\text{NS}} = 16$, $\gamma_{\text{NS}}L = 1$, $\tau_i/\tau_R = 0.33$. The normalized input flux is $I_i/I_s = 0.3$. The significance of ASE depumping is illustrated here by varying the amplifier radius-to-length ratio, a/L .

also evident in Figs. 7-4 and 7-5. Operation at high g_0/γ_{NS} improves extraction efficiency and power gain.

In bidirectional amplifiers, the gain is loaded with near uniformity, $\Delta g/g \sim 0.25$, at relatively low input intensities, $I_i/I_s \lesssim 0.1$. See Fig. 7-6. The loaded gain reaches a maximum at the center of the amplifier when operated as shown in Fig. 7-3(b). When a mirror is used to fold the beam through the amplifier, Fig. 7-3(c), the loaded gain is greatest near this mirror. This latter configuration may be more susceptible to transverse parasitic oscillations that couple with the folding mirror. Figure 7-6 also shows that at an input intensity of approximately a third of a saturation flux, the gain is loaded quite heavily so that $g_{\text{max}}/g_0 \approx 0.25$. In addition, the loaded gain distribution is relatively insensitive to amplifier aspect ratio. The gain distribution is effectively controlled against ASE depumping by the amplified laser beam. In general, the extent to which the gain is loaded depends strongly on the magnitude of the input beam intensity. The maximum loaded gain, normalized to the small signal value as a function of normalized input intensity, is shown in Fig. 7-7 for two values of g_0/γ_{NS} . The curves are parametric in amplifier aspect ratio. At low input intensities, $I_i/I_s \gtrsim 0.1$, the gain is loaded

Fig. 7-7. Maximum loaded-gain characteristics of bidirectional amplifiers, including ASE depumping. The basic laser parameters are $\gamma_{\text{NS}}L = 1$ and $\tau_i/\tau_R = 0.33$. The small signal gain to nonsaturable loss ratio, g_0/γ_{NS} , is varied from (a) 10 to (b) 20. The amplifier radius-to-length ratio is denoted as a/L . The gain is loaded by increasing the input flux, I_i .

primarily by ASE depumping and, as shown in Figs. 7-4 and 7-5, the amplifier extraction efficiency is quite low. At input intensities in excess of a third of a saturation intensity, the maximum gain is drawn down to quite small values, i.e., $g_{\max}/g_0 \lesssim 0.25$. In this regime, ASE gain loading is much less important. Again better performance is achieved at the higher g_0/γ_{NS} value.

In addition to ASE depumping, KrF amplifier scaling is also limited by the onset of parasitic oscillations when the product of the loaded gain and dimensions of the amplifier exceed certain values. In stacker/compressor applications, large scale KrF amplifiers are temporally isolated from other optical train and amplifier elements. Under these cir-

cumstances parasitic onset conditions and the characteristics of specific parasitic modes depend on amplifier-geometry and amplifier-boundary reflectivities. A simple computer simulation (for further information, see "Analysis of Parasitic Oscillations") has been used to establish the threshold for parasitic oscillation in a rectangular, uniformly loaded, gain medium having two Lambertian, diffusely reflecting walls. The diffusely reflecting walls are assumed to be the electron-beam foils which separate the high vacuum electron-beam diode regions from the high pressure KrF gain medium. Although recent UV reflectivity measurements¹⁶ with the foil, Inconel 750X, indicate that foil reflectivities are non-Lambertian, the simple Lambertian

Fig. 7-8. Parasitic limits on KrF amplifier scaling. Fig. 7-8(a) shows the bidirectional amplifier model in which electron-beam foils have diffuse reflectivity. Figure 7-8(b) shows the dependence of the threshold diffuse reflectivity, R_{th} , for parasitic onset on amplifier loaded gain-length product, gL , for the bidirectional amplifier illustrated in Fig. 7-8(a). The curves are parametric in amplifier width-to-length, W/L , and height-to-length, H/L , ratios.

Fig. 7-9. Dependence of fluorine depletion time τ on electron-beam pump power density, P_{eb} . As shown in the figure, the time period τ is fullwidth at 80% of maximum g_0/γ_{NS} . The laser medium initial composition is 93.7% Ar, 6% Kr and 0.3% F_2 at a pressure of 1 atm. The fluorine burn out time τ corresponds to the useful laser medium time for the electron-beam pump power density shown.

model probably provides an optimistic upper bound on the threshold reflectivity. Figure 7-8 shows the threshold diffuse reflectivity for parasitic onset, R_{th} , as a function of amplifier-loaded gain-length product, $\bar{g}L$, for a bidirectional amplifier configuration with folding mirror. See Fig. 7-3(c). The curves in Fig. 7-8 are parametric in amplifier width-to-length, W/L , and height-to-length, H/L , ratios. The parasitic threshold for this amplifier model depends only on the laser-loaded gain-length value, the diffuse reflectivity of the walls, and the geometrical aspect ratios for the laser medium. The results presented in Fig. 7-8 show that the threshold diffuse

reflectivity is a very strong function of the amplifier-loaded gain parameter, $\bar{g}L$. For an amplifier-loaded gain parameter of $\bar{g}L = 3$, a diffuse wall reflectivity of less than 6% is required to prevent parasitic oscillation and subsequent depumping of the medium. At $\bar{g}L \approx 4$, the diffuse reflectivity must be less than 1%. Electron-beam foil reflectivity measurements¹⁶ indicate that Inconel 750X has a reflectivity of $<4\%$ at normal incidence for 238-nm light. The reflectivity is non-Lambertian and increases sharply at larger angles of incidence. Clearly, the constraints imposed to prevent onset of parasitic oscillations will place limits on KrF amplifier scaling. We will now explore these limitations by combining our

Fig. 7-10. Dependence of KrF amplifier medium parameters on electron-beam pump power density; P_{eb} and fluorine burn out time. The laser medium initial composition is 93.7% Ar, 6% Kr, 0.3% F_2 at a pressure of 1 atm. The fluorine burnout time corresponds to the useful laser medium time for the electron-beam pump power density as shown in Fig. 7-9. The quantity, η_{KrF} , is the KrF molecule production efficiency. The laser quantities g_0 , γ_{NS} and I_s are the small signal gain coefficient, nonsaturable loss coefficient, and saturation intensity, respectively. The laser medium parameters plotted are average values for the useful time interval τ .

bidirectional extraction analysis with results from our KrF medium kinetics code.

KrF Amplifier Scaling. A single, near-optimal, laser mixture is considered.

When a 1-atm mixture, composed of 93.7% Ar, 6% Kr and 0.3% F_2 is excited by a high energy electron beam whose volumetric power deposition rate, P_{eb} , is held constant, the laser medium parameters, g_0/γ_{NS} , γ_{NS} , and I_s vary with time. The gain-to-loss ratio varies most rapidly. The gain-to-loss ratio increases rapidly at first as the kinetics begin to form the KrF lasing molecule. However, as time passes, the F_2 present in the original mixture is depleted by attachment and dissociation kinetic processes. Consequently, the gain-to-loss ratio achieves a maximum and then declines. As the F_2 is consumed, the electron loss mechanism changes from dissociative attachment to dissociative recombination. Since the dissociative recombination rate is slower, the electron density rises. This changes both the saturation intensity and nonsaturable loss coefficient as the dominant kinetic processes and composition of the

medium change. The usefulness of the medium, from a laser standpoint, is determined by the time period over which g_0/γ_{NS} remains large. If we take this time period to be the fullwidth at 80% of maximum g_0/γ_{NS} and plot it as a function of P_{eb} , the electron-beam volumetric power deposition rate, we obtain the plot shown in Fig. 7-9. The dependence of the fluorine depletion or burnout time, τ , on P_{eb} is such that $P_{eb}\tau \approx 0.1 \text{ J/cm}^3$ for this mixture.

Fluorine burn out depends on the total energy deposited per unit volume in the medium. If the medium is pumped at a slower rate, it will last correspondingly longer. If we average the laser parameters over the time, τ , and plot them as a function of P_{eb} , we obtain Fig. 7-10. The quantity, η_{KrF} , is the KrF production efficiency. The KrF production efficiency is largest, almost 25%, at low pump rates, $P_{eb} \sim 100 \text{ kW/cm}^2$. It decreases, primarily due to electron quenching, as the electron-beam pump rate is increased to $P_{eb} \lesssim 1 \text{ MW/cm}^2$. Also, as the electron-beam pump rate is increased,

Fig. 7-11. Dependence of KrF bidirectional amplifier intrinsic efficiency on electron-beam pump power density, P_{eb} , corresponding fluorine burnout time, τ , and nonsaturable loss parameter, $\gamma_{NS}L$. The initial laser medium composition was 93.7% Ar, 6% Kr and 0.3% F_2 at a pressure of 1 atm. The amplifier was driven with an input signal intensity of $0.3 I_s$, where I_s is the appropriate saturation intensity, Fig. 7-10. The effect of ASE depumping was included, assuming that the amplifier output aperture, D , was constrained by parasitics to $gD = 2$.

the nonsaturable loss coefficient, γ_{NS} , and saturation intensity, I_s , increase. The average gain-to-loss, g_0/γ_{NS} , parameter passes through a maximum as the electron-beam pump rate is increased from 0.1 to ≥ 1 MW/cm². Initially, the gain-to-loss ratio increases, as the electron-beam pump power deposition is increased, because the gain increases in proportion to P_{eb} . However, as the electron-beam pump power density is increased further, electron density increases. Electron quenching of KrF ultimately reduces the rate of increase of the maximum small-signal gain compared to the nonsaturable loss, which also increases. Consequently, at high electron-beam pump rates, g_0/γ_{NS} declines as P_{eb} increases.

The performance characteristics of a bidirectional KrF amplifier that is pumped at a rate, P_{eb} , for a time period, τ , may be calculated by using Figs. 7-4, 7-5 and 7-10. For example, the intrinsic efficiency of a bidirectional KrF amplifier is calculated and then plotted in Fig. 7-11 for different values of P_{eb} with $\gamma_{NS}L$ as a parameter. The intrinsic efficiency is largest (9.5%) and nearly constant

Fig. 7-12. This figure shows the dependence of KrF bidirectional amplifier output energy on electron-beam pump power density, P_{eb} (corresponding fluorine burnout time, τ) and nonsaturable loss parameter $\gamma_{NS}L$. The output aperture was scaled by requiring gD to be equal to values of 1 and 2, as shown. The initial laser medium composition was 93.7% Ar, 6% Kr and 0.3% F_2 at a pressure of 1 atm. The amplifier was driven with an input signal intensity of $0.3 I_s$, where I_s is the appropriate saturation intensity, Fig. 7-10.

for pump rates in the 0.1- to 0.4-MW/cm² region which have F_2 burn out times in the 1.0- to 0.20- μ s range, respectively. As the length of the amplifier is increased, $\gamma_{NS}L$ increases and amplifier efficiency decreases as the extraction efficiency decreases due to increased photoabsorption losses. Let us now consider the energy scaling of KrF amplifiers over this pulse-length interval. In general, parasitic oscillations will limit the square output aperture dimension, D , of a rectangular amplifier module of length L , such that the loaded gain-aperture product, gD , is less than a certain value. Using this constraint on amplifier aperture, we can calculate the maximum KrF amplifier energy as a function of electron-beam pump power density that can be achieved for the mixture conditions under consideration. In Fig. 7-12, results are presented for

several values of $\bar{g}D$ and γ_{NSL} . This parasitic limit will depend on the boundary reflectivity characteristics of the amplifier and the mode structure of the parasitic wave. For a transverse parasitic wave, the onset limit for walls of reflectivity, R , is simply $\bar{g}D \approx \epsilon n (1/R)$. The limitations imposed by diffuse parasitic modes were discussed earlier. If parasitic oscillations limit $\bar{g}D$ in the range of 1 to 2, then, depending on the γ_{NSL} values, the maximum KrF amplifier energy per module will lie between the appropriate curves seen in Fig. 7-12. These results show that at high pump rates, $\sim 1 \text{ MW/cm}^2$, maximum module sizes, on the order of 10 kJ, are predicted within the constraints of efficiency and fluorine depletion set forth earlier. At lower pump rates, medium gains are proportionally lower. This means larger output apertures and, consequently, higher module energies are possible. For example, maximum module energies in excess of 100 kJ are possible for pump times of 400 ns and pump power densities of 300 kW/cm^2 . These limitations depend ultimately on the constraints on $\bar{g}D$. For example, for transverse parasitic waves when $\bar{g}D = 2$, onset occurs for a boundary specular reflectivity of 14%. For diffuse boundaries, Fig. 7-8, achievement of the maximum KrF amplifier energies plotted in Fig. 7-12, $\bar{g}D = 2$ and $\gamma_{NSL} = 1.0$ curve, requires diffuse reflectivities less than 5 to 8%, depending on P_{eb} . These constraints are not unreasonable when compared to the results of recent UV reflectivity measurements¹⁶ for electron-beam foils. However, the measurements indicate that electron-beam foils are neither purely diffuse nor specular reflectors and consequently more refined analysis of parasitic oscillations will be required in order to predict accurately KrF amplifier energy scaling.

Summarizing the current status of our KrF amplifier analysis, we find that bidirectional amplifier extraction configurations load medium gain with near uniformity while retaining good fluence gain (> 10). Nonsaturable losses force tradeoffs between intrinsic efficiency and energy per aperture. ASE limits extraction efficiency in large aspect ratio amplifiers ($a/L > 0.15$). Parasitic oscillations limit aperture scaling and require diffuse electron-beam foil reflectivities of less than a few percent (currently electron-beam foils have nondiffuse UV reflectivities $\leq 5\%$). Achievement of high amplifier energies ($> 100 \text{ kJ}$) within parasitic constraints requires long-pump times ($> 400 \text{ ns}$). If parasitic oscillations are controlled, KrF amplifiers appear scalable to high

energy ($\sim 100 \text{ kJ}$) and intrinsic efficiency ($\sim 8\%$) at long pulse lengths ($> 400 \text{ ns}$).

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Raman Compressors and Amplifiers

During the last year we have continued to refine our understanding of the second Stokes limits in Raman compressors in two key areas. The effects of the shape and timing of the pulses on the second Stokes gain have been examined in further detail and reported in the 1978 annual report.¹⁷ Also, we have studied ways of reducing or eliminating the second Stokes wave by using a variety of suppression mechanisms. As a result of these studies, we now believe that the performance limits published in the literature and in previous annual reports are quite conservative. It does appear possible, if the techniques presented here can be used to full advantage, to achieve compressions of a factor of approximately 20 at 70% extraction efficiency.

Pulse Shape, Timing, and End Effects. The critical parameter in improving the compressor performance is the second Stokes gain.^{18,19} Simply stated, the issue involves two questions:

- How much pump energy may be transferred to the Stokes pulse without generating excessive gain at the second Stokes frequency?
- What attributes of the compressor are most critical in our efforts to maximize this transferred energy?

To address these points, let us return briefly to the evaluation of the second Stokes gain, G_{2s} . We again consider the idealized case of a compressor using plane wave pulses that are spatially top-hat in shape.²⁰ The second Stokes wave builds from optical noise and experiences gain due to the Stokes wave, a gain which depends on both the pulse shape and the pulse distortion of the Stokes wave. The equation for the parasitic wave is

$$\left(\frac{\partial}{\partial z} + \frac{1}{c} \frac{\partial}{\partial t}\right) I_{2s} = \gamma_f I_s^2 \quad (1)$$

Here, the gain parameter, γ_f , is that appropriate for forward Raman scattering. On transforming from the variables (z, t) to the set (x', y) , where,

$$x' = t - n_{2s} z/c \quad (2)$$

and

$$y = t + n_p z/c \quad (3)$$

we find that

$$\partial I_{2s} / \partial y = \gamma_f c (n_p + n_{2s})^{-1} I_{2s} I_s \quad (4)$$

Now the Stokes intensity is obtained from the compressor analysis in terms of the Stokes time, x . In terms of x' , the Stokes time is

$$\begin{aligned} x &= t - n_s z/c \\ &= \frac{n_p + n_s}{n_p + n_{2s}} \left(x' + y \left[\frac{n_{2s} - n_s}{n_p + n_s} \right] \right) \\ &\equiv X(x', y) \end{aligned} \quad (5)$$

The difference between x and x' represents the difference in velocity of the Stokes and second Stokes waves. As the pulses traverse the cell, they move

slightly with respect to each other. In a nondispersive medium, of course, $x = x'$. In Eq. (4), the differential is to be taken holding x' constant, that is, at a particular position in the second Stokes pulse. At fixed x' , the argument, x , of the Stokes pulse varies with y . This indicates that the second Stokes wave experiences the Stokes intensity along a range of points in the Stokes pulse, as it moves relative to the Stokes pulse. Thus, from Eq. (4), the gain exponent experienced by the parasitic wave is

$$G_{2s}(x') = \int_a^b \gamma_f c (n_p + n_{2s})^{-1} I_s(X(x', y), y) dy \quad (6)$$

The integration limits are the times in the pump pulse at which the point, x , in the Stokes pulse enters and leaves the cell. Thus,

$$a = x' - 2n_{2s} z/c \quad (7)$$

and

$$b = x' + 2n_{2s} (L - z_1)/c \quad (8)$$

where z_1 is the distance into the cell at which the leading edges of the pump and Stokes waves meet.

This can be clarified by visualizing a specific example. Consider a Raman cell which is much longer than the required length $ct_p/2$. Further, let the Stokes pulse be very short, and timed so that it fully enters the cell before the pump has filled it. The cell length is such that the Stokes pulse propagates through the entire pump well before it reaches the end of the cell. Consequently, in this example, the Stokes pulse propagates through a length of Raman medium both before and after it enters the region of gain (defined by the pump pulse to be $1/2 ct_p$ in length).

The second Stokes wave experiences gain during the *entire* cell length, and not just over the gain region. Thus, in Eq. (6) the integration is not just over the pump pulse length, but includes the entire length of the cell. In time, the pump pulse length, or gain region, is $b - a = n_{2s} L/c$. Also, in the same example, let the pump be square and fill the region $0 \leq y \leq t_p$, and let the Stokes pulse be confined to the region $0 \leq x \leq t_s$. Thus, the leading edges of the pulses meet at $x = 0$ and $y = 0$, corresponding to the point $t = 0$ and $z = 0$. However, the medium extends back to the point, $z = -z_1$, and also forward to

the point, $z = L - z_1$. Thus, the pulses meet at a distance, z_1 , into the compressor cell. The second Stokes wave has already experienced gain along that portion of the cell. This is represented in the gain integral as a contribution from the region, $y < 0$, which represents times prior to the meeting of the leading edges, and where there is medium and Stokes energy but no pump. Finally, in the same example, note that a point in the second Stokes pulse, x' , behind the leading edge has traversed only a distance, $z_1 - cx'/n_{2s}$, by the time the leading edges meet. This means the contribution from the region, $y < 0$, is reduced by x' . This example also shows that the origin, $x = y = 0$, where the leading edges meet, is a useful reference point for discussing timing.

The pulse shape dependence of the second Stokes gain derives from two phenomena, dispersion and gain saturation. If the pump is shut off from the cell, then the Stokes wave propagates without gain and without pulse distortion due to gain saturation. Then, the gain at the second Stokes frequency becomes an integral over a finite portion of the Stokes wave,

$$G_{2s}^d(x') = \gamma \int_0^L dz I_s [x' + (n_{2s} - n_s)z/c] \quad (9)$$

This "smearing" of the gain can be effective in removing the undesirable effects of hot spots in the Stokes wave. If the Stokes intensity fluctuates with angular frequency, ω_c , then we may write it as follows,

$$I_s(x') = I_0 + I_r \sin(\omega_c x' + \phi_c) \quad (10)$$

where I_0 and I_r are slowly varying envelopes. If the envelopes vary but little in the time $(n_{2s} - n_s)L/c$, then the second Stokes gain is

$$G_{2s}^d(x') = \gamma_f I_0 L + \gamma_f I_r L \theta^{-1} [\cos(\theta + \phi_c) - \cos \phi_c] \quad (11)$$

where,

$$\theta = (n_{2s} - n_s) \omega_c L/c \quad (12)$$

If the angular frequency is large so that $\theta \gg 1$, the ripple on the gain is much less than the ripple on the Stokes intensity. Thus, dispersion quells the tendency of the hot spots in the Stokes wave to convert to the second Stokes wave.

On the other hand, if there is no dispersion, but the Stokes wave experiences gain, it will distort due

Fig. 7-13. Pulse shapes in a saturating amplifier. The pulse travels to the left.

to gain saturation as indicated in Fig. 7-13. Consider the second Stokes gain arising only from the gain region, $0 < y < t_p$. Then,

$$G_{2s}^g(x) = \int_0^{t_p} \gamma^2 c (n_p + n_{2s})^{-1} I_s(x, y) dy \quad (13)$$

At the point, x , the y -dependence of I_s , which is exponential in the small-signal regime, is strongly non-exponential in the saturated region. As an example, consider the point, $x = 0$, in Fig. 7-13. Initially, this point is considerably down from the peak of the Stokes pulse, but at the point, $y = 0.4$, it is close to the peak. However, for subsequent points it falls further away from the peak. For the point in the tail of the pulse, say at $x = 0.2$ in Fig. 7-13, the sequence is that of being initially near the peak of the Stokes pulse and subsequently falling away. On the other hand, at $x = 1.0$ the sequence is that of being initially far from the peak and approaching it closely as the pulse leaves the gain region. It is clear that the second Stokes gain is a maximum for those values of x that are close to the peak of the pulse in the highest Stokes fluence region, i.e., near the end of the gain region. Thus, as a function of x , the second Stokes gain tends to peak ahead of the peak of the Stokes input pulse. It also tends to peak to the rear of the peak of the Stokes output pulse.

Thus, the second Stokes gain is expected to show marked pulse-shape dependence. For a positively skewed Stokes input, the peak gain (as a

Fig. 7-14. Universal optimal output pulse shapes for the Raman compressor. The units are QS/t_s for intensity and $Q(x/t_s)$ for time.

function of x) will be larger than for a negatively skewed input, such as that used in Fig. 7-13. In last year's annual report,¹⁷ we described a calculation which showed how to optimize the pulse shapes for an ideal high-compression Raman cell. In this limit (high compression), the end corrections are ignored, as is dispersion. We have studied the optimal pulse shapes obtained this way in detail. Recall that the performance quotient, Q , of the compressor is defined by

$$Q = G_F/R \kappa_0, \quad (14)$$

where G_F is the maximum tolerable second Stokes gain, R is the forward-backward Raman gain ratio, and κ_0 is the nominal compression. The pump pulse is assumed to be flat and its length is t_p ns, whereas the Stokes pulse shape is assumed to be zero outside a time interval of t_s ns. That is, the Stokes pulse is confined to a region $0 \leq x \leq t_s$ but is otherwise unconstrained. Then, $\kappa_0 = t_p/t_s$ is the nominal compression. With the definition of a normalized time, t ,

$$t = Q(x/t_s), \quad (15)$$

it was shown that the optimal pulse shapes are functions of t and G alone, where G is the small-signal amplifier gain. Universal curves for the input and output optimal pulse shapes are given in Figs. 7-14 and 7-15 and the efficiency of extraction in Fig. 7-16. Each graph is parametric in G which takes the

Fig. 7-15. Universal optimal input pulse shapes for the Raman compressor. Same units as in Fig. 7-14.

range of values 2, 3, 4, ..., 10. The actual pulse shape for a particular value of Q and G is given by the curve corresponding to the value of G , taken between the limits $t = 0$ and $t = Q$. Thus, for low $Q \sim 1$, the output pulse shape drops approximately linearly from the leading edge, and the input rises linearly. However, for low Q , the efficiency also is low. (This is the high compression region.) On the other hand, for high Q the efficiency may be quite high. The output intensity drops rapidly away from the leading edge value and develops a long, essentially flat tail. The input intensity becomes close to an exponentially rising pulse. Thus, for moderate $Q \sim 1$, the output pulse shape is sufficiently skewed so that the actual compression is considerably larger than the nominal compression, κ_0 . The actual increase depends on how pulse length is defined for skewed pulses. However, it appears that a factor of 2 increase is not unreasonable.

If fluence is required, rather than intensity, then there is only a small increase in compression over the nominal value, κ_0 . Because the intensity tends to the value, $i = i_0 = QS/t_s$, beyond $t \sim 1$, there is always substantial energy in the tail of the pulse. Thus, there appears to be little advantage in optimization for fluence compression, although a factor of 2 increase in intensity compression seems plausible.

The distinction we draw between fluence and intensity compression is in a sense conceptual rather than physical. The definition of compression is different in the two cases, reflecting the fact that the

Fig. 7-16. Efficiency vs compression (or $1/Q$) for optimal pulse shapes. The actual compression is somewhat greater due to the skewed shape of the output pulse.

appropriate definition depends on the intended application. It is possible, therefore, that a particular choice of compression definition could be misleading in certain applications. Consequently, we prefer not to choose a particular definition, using instead the universal curves shown in Figs. 7-14, 7-15, and 7-16 and the definition of Q to characterize the compressor performance.

It is instructive to compare the optimal pulse results with previous results for Gaussian input Stokes pulses.^{17,20} The curves of efficiency versus $1/Q$ for the optimal pulse are given in Fig. 7-16. The corresponding curves for the Gaussian pulse are shown in Fig. 7-17. Note that Q for the Gaussian pulse is defined in terms of the full width at half maximum (FWHM) of the Stokes input, z . Thus, if

$$G_m = \max_{\{x\}} G_{25}(x) \quad (16)$$

the Q for the Gaussian pulse is

$$Q = G_m R(t_p, z) \quad (17)$$

The curves are remarkably similar, but the closeness is somewhat misleading. The output pulse shapes for the optimal case are strongly skewed and any reasonable definition of the pulse length gives a compression greater than κ_0 . For Gaussian input pulses, however, the output pulse length (FWHM) is generally about the same as that of the input.

Fig. 7-17. Efficiency vs compression (or $1/Q$) for Gaussian input pulses.

Note that a comparison of the two cases is not entirely unambiguous in that different pulse shapes are being compared. However, it appears that the optimal pulse shape is capable of improvement in compression up to a factor of ≈ 2 over the Gaussian input case.

The end corrections to the second Stokes gain itself are necessitated by the requirement that the cell be as long as, or longer than, the sum of the pump and Stokes pulse lengths. Their relative magnitude is therefore expected to be of the order of the ratio of the Stokes pulse length to the pump pulse length, or inversely proportional to the compression. Their impact on performance is given moderately well in terms of an equivalent, Q' . To determine the achievable compression at a given efficiency for optimal pulse shapes including the end corrections, first the curves of Fig. 7-16 are used to determine a Q . This gives a nominal compression κ_0 . The equivalent, Q' , is related to Q in a way which depends on the relative timing of the pulses. For best pulse shapes, optimally timed, it is given approximately by

$$Q' = Q + \gamma G_m G_{25} \quad (18)$$

This equivalent, Q' , is then used instead of Q to determine the compression κ_0 . Since $Q' > Q$, the end corrections always decrease the achievable compression at a given efficiency.

In the high efficiency region, this has a substantial impact on performance. For example, in a typical situation where $G = 5$ and $G_{25} \sim 20$, $Q'/Q \sim 1.25$ at high efficiency. Thus, for these parameters

the achievable compression at the given high efficiency is reduced by 25%. This effect is worse at low compressions. In optimization of the compressor efficiency, low compressions are required. However, at low compressions the cell length must be increased to permit the Stokes and pump pulses to overlap everywhere within the cell. Namely,

$$L = 1/2 \text{ ctp } (1 + 1/\kappa) . \quad (19)$$

Thus, the end correction is larger at lower compressions, and therefore the portion of the $G_{2\kappa}$ budget allocated to the gain region decreases as the compression is lowered. At the second Stokes limit, the efficiency for a given compression is therefore less than the high compression limit result (see Fig. 7-16) by an amount which increases with decreasing compression. Consequently, the achievement of very high efficiencies at low compressions, as indicated in Fig. 7-16, is not possible. These curves should turn over as $1/Q \sim 0$. The efficiency limits at low compression depend sensitively on the relative timing of the pulses, and on the pulse shapes. Unfortunately, in the region where the end correction is large, the optimization of the pulse shape cannot be carried out accurately by the specialized method used previously. Thus, the techniques developed here are inapplicable to the very high efficiency region. On the other hand, in the practical region $\kappa \lesssim 4$, the equivalent Q' approach described here is a good approximation.

Finally, we note that there are several implications for compressor performance:

- There appears to be some advantage to optimizing the Stokes pulse shape.
- End corrections may degrade the performance significantly.
- Consequently, low compression operation will not achieve the very high efficiencies seen in Fig. 7-16.

If high efficiency operation is to be achieved, it is clearly important to arrange the timing and cell length so that the end corrections are minimized.

Second Stokes Suppression Techniques. The methods considered for second Stokes suppression divide into two classes, depending on whether or not the method is capable of operating inside the Raman cell. The importance of this distinction between internal and external suppressors lies in their

potential for increased compression. External compressors can remove the parasitic wave only between cells, and can not affect the ultimate compression achievable within a cell. The compression achievable in a chain of cells is less than that in a single cell because second Stokes waves originating in one cell may propagate to the next. The total gain for the parasitic wave along the whole chain is necessarily larger than that for a single cell operating at the same efficiency and compression. Thus, in the absence of a suppressor, the achievable chain compression is less than the achievable single cell compression. Insofar as external suppressors are capable of resetting the parasitic input to zero at each stage, they allow the achievable chain compression to approach single cell performance. Thus, external methods are important in realizing single cell performance in a chain, but are incapable of improving the performance of the Raman cell itself.

Internal suppressors, on the other hand, are capable of attenuating the parasitic wave at any point within the cell. The total parasitic gain along the cell is the difference between the contribution of the Stokes wave, namely the G -integral, and the attenuation due to the suppressor. The G -integral increases monotonically with the compression, and thus, the introduction of a suppressor allows an increase in compression for the same total parasitic gain. A detailed analysis of the parasitic dynamics in the presence of the suppressors has been carried out. It shows that the increase in compression is indeed proportional to the attenuation. If $e^{-\gamma L}$ is the attenuation and G_F is the maximum tolerable gain, then, the compression increase under optimal conditions is

$$\kappa/\kappa_0 = 1 + \gamma L/G_F . \quad (20)$$

Since $G_F \sim 20$, this shows that very high attenuations are required to cause a significant increase in compression. We shall return later to the requirements imposed by near optimal conditions. These are in general hard to meet, and specific to the suppression mechanism.

The parasitic wave has two properties that distinguish it from the Stokes wave from which it must be separated. These are its frequency and angular divergence. The frequency separation is typically large enough that frequency-dependent properties

of materials can reasonably be considered. It is very difficult to envision an internal suppressor which selects the angularly bright signal. Thus, the external suppressors we have considered utilize frequency selection. These methods include the introduction of a selective absorber, dichroic mirror, the use of resonant Raman media, and chirped pulses. Other methods of frequency selection include the use of gratings and prisms to separate the parasitic wave from the Stokes wave, spatially, and the use of dispersive media to cause the Stokes and parasitic waves to separate longitudinally. Brightness selection is possible with the use of spatial filters and distance filtering. Clearly, the most important techniques to study for compression increase are the frequency selective internal suppressors.

Table 7-3 shows the methods considered to date and our present estimates of the achievable increase in compression. For external suppressors, this increase is set to the nominal value of 1.5, a number which represents the difference between chain and single cell performance. This should not be taken too literally since it depends on many factors, including the number of chain elements and the capability of shaping the Stokes pulse. For internal mechanisms, the potential for reaching high compression levels is clearly present; but, in practice, achievement of the conditions under which such potential is realized is difficult. Table 7-3 also shows the attenuation which can be realized, taking into account factors such as the materials available, technological constraints, and the degree to which optimal conditions are realizable. A few comments on these methods follow.

Absorbers. The absorber may be a vapor, distributed throughout the cell, or an ion in a solid host placed at discrete locations within the cell. Using an absorption cross section at 10^{-18} cm², the extinction length for vapor at 10 Torr is 30 mm, and, for solids doped with 1% ionic absorbers, it is 0.1 mm. There appears to be little difficulty in achieving attenuations ~100 with either method. In practice, however, materials which absorb the parasitic wave also absorb the pump and Stokes waves. In order for the efficiency not to suffer, there must be no attenuation of the pump and Stokes waves and high attenuation at the second Stokes frequency. In the case of solid hosts, reflection and

7-3. Techniques studied for second Stokes suppression.

Internal	Compression increase	Attenuation exponent
Absorbers	2-10	100
Dichroics	5	7
Atomic scatterers	2-10	N/A
Chirping	2-10	N/A
External		
Gratings	1.5	5
Prisms	1.5	5
Spatial filters	1.5	8
Distance filtering	1.5	3
Dispersion separation	1.5	~0

Fig. 7-18. Increase in compression for second Stokes suppression by an ideal absorber, as a function of the loss exponent. Very high attenuations are required to increase the achievable compression significantly.

scattering losses at the surface must also be eliminated. The analysis shows that the figure of merit of an absorber is the contrast ratio, r . This is the ratio of absorption cross sections,

$$r \sim \sigma_{2s}^A / (\sigma_s^A + \sigma_p^A) \quad (21)$$

Figure 7-18 shows the limit on the efficiency of the Raman cell as a function of performance quotient, $Q = RK/G_p$, typically $Q = \times 10$. Each curve is parametric in the attenuation, L , which

varies from $L = 0$ to $L = 120$ in steps of 20. These curves detail the linearity of κ in L . At any fixed efficiency and pulse shape, Eq. (1) is exactly satisfied. For imperfect absorbers, Fig. 7-19 shows the fractional loss in efficiency, $\delta\eta/\eta$, incurred by absorption at the pump and Stokes frequency as a function of r . These curves are also parametric in L and correspond to the curves of Fig. 7-18.

To a large extent, the fractional efficiency loss is independent of the efficiency itself, and in particular, for $\eta \sim 70\%$ this dependence is negligible. Clearly, to be useful, r must be in the range $r > 100$.

Several absorbers have been identified. For an XeCl-pumped CF_4 compressor, cadmium vapor has an atomic line at the parasitic frequency. Atomic vapors possess high contrast ratios, $r > 1000$, and are attractive candidates for absorbers. However, their utilization requires fine frequency control of the KrF laser and warm cells (200°C) to achieve the required vapor pressure. For KrF-pumped methane, cerium ions in barium fluoride absorb at 290 nm, but their contrast ratio is small, $r \sim 12$. We are also testing thallium-doped alkali halides for the KrF/ CF_4 application. Saturable absorbers such as SO_2 have also been suggested and are presently being studied.

Dichroic Mirrors and Transmission Gratings.

These separate elements are in almost every way equivalent to discrete absorbers. Again, the appropriate figure of merit is the contrast ratio,

$$r = L_{2S}/(L_S + L_P) \quad (22)$$

where L_s is the loss exponent $\ln(1 - T)$, at the Stokes frequency. Typically, the transmission is limited by surface scatter, coatings, and grating fidelity. Typical best values are $r \sim 50$. The performance of these suppressors is again given by Figs. 7-18 and 7-19. Unfortunately, the achievable loss, L_{2S} , is not great, only about -5 to -7 . To achieve significant compression increases, many elements must be introduced; the many surfaces in the beam then decrease beam quality. It is possible that this technology can achieve $L_{2S} \sim -12$ but -20 appears beyond reach. In summary, the compression achievable with these elements is limited mainly by the technology of their fabrication.

Fig. 7-19. The efficiency penalty associated with finite absorption of the pump and Stokes waves by the second Stokes absorber. Very high contrast ratios are needed to avoid a substantial loss due to this absorption.

Dispersive Scatterers. The use of a dispersive Raman medium in which the gain at the second Stokes frequency is less than that at the Stokes frequency, is equivalent to using a medium with a forward-backward ratio less than unity. The backward gain is evaluated at ω_s but the appropriate forward gain is evaluated at ω_{2S} . The G -integral is proportional to $R\kappa$; thus, any reduction in R allows a corresponding increase in κ . The compression increase is therefore

$$\kappa/\kappa_0 = \gamma(\omega_s)/\gamma(\omega_{2S}) \quad (23)$$

The use of atomic vapor Raman media takes advantage of this effect. To be useful, the candidate's strongest Raman line must have a high quantum efficiency. It must also possess a resonance near the pump frequency and none near the Stokes frequency. Additionally, it must have a vapor pressure high enough to give significant gain at realizable temperatures. For use with KrF or XeCl pumps, the best candidates appear to be TI,

In, Bi, Sb, and Pb, i.e., KrF-pumped Sb and Xe pumped In or Bi. However, the use of hot cells (800 to 1000°C) is required for all these candidates.

Chirping. This scheme for reducing the gain length for the second Stokes wave relies on chirping the frequencies of the pump and Stokes waves, and also on varying the Raman frequency of the medium along the cell.²¹ As the Stokes wave propagates through the cell, the locally generated parasitic wave has a frequency which at subsequent points in the cell becomes out of resonance with the new local parasitic frequency. The parasitic waves experience gain only so long as the frequencies match; thus, the gain length is reduced by the ratio of the total chirp-to-Raman line width. The chirp required is $\sim 10 \text{ cm}^{-1}$.

To achieve this chirp in the medium, a magnetic field of 1 to 10 T is required to split and shift the levels. This technique has been considered only in connection with atomic scatterers. Considerable technological analysis is required to assess the architecture of the required superconducting magnets surrounding a hot atomic cell. The cw power requirements to maintain this chirped cell are large and may be as much as 10^5 W . In summary, there appears to be no insurmountable obstacle to this technique, which can apparently reach an increase in compression by a factor of 10. However, many technological questions remain; no specific candidate system has been determined.

External Suppression. External methods include, as a subset, the internal methods which we have already discussed. These exclusively external methods include spatial filtering and the use of prisms or gratings to separate the waves spatially. Due to lack of a suitable material, no method of longitudinal separation using dispersion has been devised. Some exotic ideas such as coherent pulse propagation have been investigated. The leverage in external methods is not so great as for internal methods, and we shall spend less time on them. Table 7-3 summarizes the major conclusions concerning the attenuation.

The separation angle from prisms and gratings must obviously be greater than the angle of divergence of the parasitic wave. Since all surfaces contain microirregularities which scatter light, the separation angle should also be greater than the typical surface scatter angle. The contrast ratio, L_{25}/L_5 , should be very large since L_5 is limited by efficiency considerations. Perhaps the most effective

method is the spatial filter, modified by replacing each lens with a combination of a lens and a small-angle prism. The attenuation achievable in the resulting skew filter is potentially very high, except that the B-integral associated with the prisms may be quite large. In general highly dispersive materials suitable for the prism have large non-linear indices. Based on these considerations, we have concluded that the exclusively external techniques are not capable of providing attenuations much beyond e^{-5} to e^{-8} , even for the most optimistic estimates of the surface scatter and angular divergence of the parasitic wave. Since e^{-20} is required, multiple suppressors will be required between stages.

In conclusion, the most promising suppression techniques are the frequency selective internal methods which impact the single cell compression. Several exclusively external methods are available to give attenuation of about one-fourth the requirement. These do not improve compression, but merely prevent significant deterioration of chain performance from single cell performance. A judicious staging scenario paying careful attention to pulse shape effects, combined with spatial filters in the Stokes beam line and the best available internal methods in the cell, may be able to reach a compression increase of 1.5 to 3 over the single cell performance.

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RAPIER Experimental Program

The RAPIER system is a test bed for experiments on the technology and systems integration of KrF pulse stacker/Raman pulse compressor options for fusion. The system presently consists of a

KrF-discharge laser "front end" system operating at a nominal energy of 1 J. This system drives a 25- to 35-J, $10\text{-} \times 10\text{-} \times 50\text{-cm}$ amplifier which is referred to as the A-amplifier. This amplifier is pumped by two opposed, command-triggered electron beams using water Blumlein pulse-forming lines. A B-amplifier, capable of operating at energies of several hundred joules and driven by the A-amplifier, will complete the major components required for a test of an integrated Raman stacker-compressor system.

During 1979, we conducted experiments to study the laser performance of the A-amplifier as an oscillator and multipass pulse-stacked amplifier. Experiments using the discharge laser front end have concentrated on verification of the theoretical predictions of Raman amplifier performance. We have also studied techniques for producing the proper drive pulses for the A-amplifier and its Raman cell when the two systems are integrated in 1980. Pulse-power components of the A-amplifier are being upgraded for use in the construction of the B-amplifier. Laser triggering has been under active investigation as a technique for synchronizing the electrical systems of the components of the RAPIER system.

Raman Experiments. In last year's annual report,²² we described the front-end facility of the RAPIER laser system. In that report, we discussed the layout of the various pulsed discharge KrF lasers and methane cells to be used in backward wave Raman scattering experiments that critically test various aspects of the Raman pulse compression technique. That presentation also contained construction details for the individual lasers and the pulsed-power approach used to excite the gain media. Here, we present experimental results on backward wave Raman pulse compression using that facility.

The present investigation employs four UV-preionized KrF discharge gain media to produce a nominal 1 J of pump energy at 248 nm in a pulse length of about 35 ns and a line width 0.15 to 0.20 cm^{-1} . Such relatively high spectral purity is necessary to minimize the forward-to-backward Raman gain ratio.²³ Using three intracavity etalons to achieve the narrow line width, a master oscillator produces a 20 ns-long, 1-mJ pulse. This pulse injection locks a similarly sized ($0.5\text{-} \times 1\text{-} \times 100\text{-mm}$)

gain medium equipped with unstable resonator optics to generate a 150-mJ output. Details on a basically identical approach have been reported previously.²⁴ In turn, a fraction of this energy is used to injection lock a larger KrF-discharge volume. This volume also uses an unstable optical cavity and has a gain region of $1\text{-} \times 4\text{-cm}$ cross section by 1-m length. This source delivers up to about 1 J of laser energy when operated as an oscillator with a stable cavity (4% flat output coupler and 10-m maximum reflector at 248 nm). In practice, to reduce stressing the various components, the electrical excitation for this discharge is kept at reduced values corresponding to stable oscillator outputs $1/3$ to $1/2$ the maximum value. Finally, the 35-ns long, injection-locked pulse is amplified by a single pass through a second identical amplifier. This results in a total 248-nm, line-narrowed pump energy of about 1 J. The four KrF discharges use gas mixtures of 0.4% F_2 , 10% Kr, with the balance He, at total pressures of 1 to 1.5 atm. A master trigger unit fires the devices resulting in relative jitters among the discharges of less than a few ns.

Figure 7-20 illustrates the distribution of the KrF-pump laser intensity to a Stokes oscillator (that operates via stimulated forward Raman scattering), a backward Raman preamplifier, and a backward Raman amplifier. For the current investigations, the three stainless steel cells—capped with 20-mm thick, UV-grade quartz windows—were operated at about 380 psi methane pressure. The 1.5-in.-diam cells had the following temporal lengths: oscillator, 10.3 ns; preamplifier, 10.3 ns; amplifier 14.0 ns. As seen in Fig. 7-20, the preamplifier and amplifier are each equipped with pump delay mirrors. The Raman cell interiors, designated by solid circles, denote the intersection of the leading edges for the pump and Stokes wave. For the preamplifier, which is operated under small-signal Raman gain conditions, the pump delay option permits passive time-tailoring of the transmitted Stokes pulse. In general, a Stokes pulse with a slow rise time is desired for passage through the saturated amplifier. This prevents formation of an excessively steep leading edge that too rapidly reaches the production threshold of the second Stokes wave. Note that this parasitic process, generated by the back-traveling first Stokes wave, involves forward Raman scattering and, therefore, can have a relatively high cross section.^{22,23} The pair of pump delay mirrors associated with the

Fig. 7-20. Diagram of the experimental arrangement used in the backward-wave Raman scattering studies. The total KrF-pump laser intensity (solid line) is divided and directed to three methane-filled Raman cells. These cells serve (bottom to top) as the Stokes oscillator, backward Raman preamplifier, and amplifier. The Stokes oscillator uses stimulated forward Raman scattering to generate the 268-nm pulse, shown as a broken line. The two regions designated with solid circles indicate the intersection of the leading edges for the pump and the Stokes wave. Omitted for clarity are various focusing lenses and collimating telescopes.

saturated amplifier serve primarily as a convenience to fine tune the intersection location for the pump and Stokes pulses. Mirrors with partially transmitting dielectric coatings, as indicated in Fig. 7-20, allow recording of input-output beams for the two amplifiers. The pump and Stokes pulses transmitted by these mirrors were viewed by photodiodes equipped with apertures a few millimeters in diameter. The apertures were positioned to correspond with spatially smooth sections of the pump beams.

Representative waveforms for the pump and Stokes pulses exiting the saturated amplifier are shown in Fig. 7-21. The traces are recorded with S20-sensitive photodiodes, having response times of ≥ 1 ns, and Tektronix 519 oscilloscopes. In Fig. 7-21(a) the effect of a counterpropagating Stokes wave on reducing the transmitted pump intensity is clearly evident. Electronic integration of these signals indicates that the photon extraction efficiency is 70 to 75%,²⁵ while the energy extraction efficiency is lower by a factor 0.93 (248/268). Since the Stokes pulse is created in the local oscillator by stimulated forward Raman scattering, the back-traveling 268-nm wave begins with a relatively long duration, approximately equal to that of the 248-nm pump beam. Consequently, because of the relatively

Fig. 7-21. Photodiode traces of: (a) KrF pump laser and (b) 268-nm Stokes pulses exiting opposite ends of the methane-filled, backward Raman amplifier. In (a), the larger signal is recorded *without* the propagation of the back-traveling Stokes pulse; the smaller signal is recorded *with* the passage of the Stokes wave. For each of the conditions shown in (a), two traces are recorded to characterize the reproducibility of the data. In (b), a clarifying smooth line is drawn through the output Stokes pulse shapes. Electrical integration of the signals in (a) shows that the photon extraction efficiency is 70 to 75%. The output Stokes intensity is >3.5 times greater than the input pump intensity; the pulse compression ratio is ≈ 5 .

short lengths of the Raman amplifiers used in this study, all parts of the counterpropagating Stokes pulse do not sweep through the pump beam in a methane background. Therefore, final portions of the Stokes pulse see little, if any, Raman gain. Additionally, for the selected pump beam optical delay, the Stokes pulse that exits the small-signal preamplifier has a relatively slow rise time, compared to cases for which the pulse-edge intersections of the pump and Stokes pulses are positioned closer to the oscillator output. A detailed experimental study of pulse shape effects on saturated amplifier performance will not be part of this report. However, straightforward computer modeling of pulse propagation effects, as described below, underscores the utility of a slowly rising, backward first Stokes pulse. Such a pulse shape suppresses the onset of second Stokes generation and thereby maximizes the energy extraction from the pump beam. (For additional information, see "Second Stokes Suppression.") Furthermore, passive pulse-shaping techniques, as used in the present study or supplemented by electrooptic techniques, can help produce laser temporal shapes that are considered optimal for target irradiation in inertial confinement fusion studies.

The Stokes pulse emerging from the saturated backward Raman amplifier is depicted in Fig. 7-21(b). It has a pulse width about of 7 ns (FWHM), corresponding to a pulse compression ratio of five with reference to the 35-ns pump pulse. At the operating photon extraction efficiency level of 70 to 75%, the laser intensity enhancement of the counterpropagating Stokes wave is a factor of ≈ 3.5 , with respect to the pump pulse. Operation below the second Stokes limit was confirmed by the presence of signals at the 292-nm second Stokes wavelength which were a few percent of the first Stokes signal strength. Thus, in this work, we have substantially shortened a relatively long Stokes oscillator pulse by the combined effects of low trailing-edge Raman gain in the relatively short methane amplifiers and gain-steepening in the saturated amplifier. For the present investigation, we merely require that the resultant Stokes pulse width be less than the saturated amplifier length. For this condition, the behavior of the trailing part of the Stokes pulse is not critical to our evaluation of the Raman pulse compression technique. If necessary, the trailing signal can be eliminated with standard electrooptic techniques. Data were also obtained at the second

Stokes limit for photon-extraction efficiencies of 50% and 30%, corresponding to pulse compression ratios of about 10 and 25, respectively. In the latter case, about 35 mJ of first Stokes energy was obtained in a pulse width of ≈ 1.5 ns.

The experimental results were compared with the predictions of a computerized Frantz-Nodvik saturated amplifier model characterizing Raman pulse compressors.^{22,23} The main physics assumptions in this analysis include:

- Smooth plane waves.
- Slowly varying amplitude approximation.
- Exact resonance condition, i.e., $\nu_S = \nu_P - \nu_R$, where the labels are S = Stokes, P = pump, and R = Raman
- Steady-state scattering in which molecular transients are neglected.
- Length of the cell (L) and the timing of the pulses are close to the ideal case, $L = 1/2$ ctp.
- The Stokes-pulse length is short compared to the pump-pulse length, (t_p).

The final two conditions are barely satisfied for the lowest compression results. Computer solutions of the propagation equations for the Stokes and pump wave were obtained under these conditions. Representative results are shown in Fig. 7-22 wherein the photon extraction efficiency at the

Fig. 7-22. Photon extraction efficiency for a backward Raman methane amplifier as a function of the parameter, Rk/G_1 , at the generation limit for the second Stokes wave. Computations for three different input Stokes temporal pulse shapes are shown (solid lines) along with the experimental results and their associated uncertainties (boxes).

second Stokes production limit is plotted as a function of the parameter, $R\kappa/G_f$, where R = forward-to-backward Raman gain ratio, κ = the ratio of pump-pulse length to exiting Stokes-pulse width, and G_f = second Stokes gain. For the present investigation, the R value lies in the range 1.5 to 1.7. This value is determined mainly by the scatterer line width at the operating methane pressure (380 psi) and the line width of the 248-nm pump.^{22,23} G_f is set equal to 20, corresponding to a nominal superfluorescence limit above which the second Stokes wave is readily produced. The data, as typified by Figs. 7-21(a) and 7-21(b), permit determination of the photon extraction efficiencies and $R\kappa/G_f$ values for the three distinct cases studied. The experimental results, along with their associated uncertainties, are plotted as boxes in Fig. 7-22. Fairly good agreement exists with the model calculations and, in particular, with the predictions that consider a positively skewed Gaussian Stokes pulse incident on the saturated amplifier to characterize the data at the 50% and 30% photon extraction efficiencies. This representation is appropriate since no pump beam optical delay was used for the Raman preamplifier, resulting in forward-leaning Stokes input pulses to the amplifier. On the other hand, the measured extraction efficiency of 70 to 75% for the lowest compression case (which utilized the delay) appears to be somewhat less than the simulations indicate. Note that for this case a more appropriate Stokes input pulse shape is the negatively skewed Gaussian wave. Most probably, this discrepancy is due to the non-plane-wave nature of the pulses used in the experiments, the lack of exact alignment between the Stokes and pump beams in the saturated amplifier, and the limitations of the modeling due to finite amplifier cell length.

Currently, the front end of the RAPIER facility is being upgraded with several higher-energy KrF gain media and reconfigured pulsed-power circuitry. The upgrade also has provisions for greater timing compatibility with the A-amplifier. The combined front-end/A-amplifier subsystems will be used to conduct stacker-compressor experiments in the near future. Other front-end improvements presently underway include:

- Installation of a master system oscillator equipped with an extra-cavity Fabry-Perot filter to transmit a single longitudinal mode for subsequent amplification. This eliminates the temporal structure on the KrF pulses due to mode beating.

- Installation of a Pockels cell for the Stokes beam exiting the Raman preamplifier to allow for active pulse shaping. This supplements the passive pump delay technique already described.

In addition to the integrated experiments with the A-amplifier, the front-end facility will be used in future studies to:

- Perform detailed pulse shape comparisons between theory and experiment for backward Raman amplifiers. This will enable further critical testing of the model that is used to characterize the performance features of megajoule-class laser-fusion systems.

- Evaluate candidate second-Stokes suppressors.

- Evaluate the spatial averaging property inherent in the Raman pulse compression scheme which relaxes the requirements on the quality of the pump laser.

A-Amplifier Experiments. In last year's Annual Report,²² we described the pulsed-power design of the RAPIER electron beam pumped A-amplifier and some initial tests on the pulsed-power components. This year we have completed the assembly of the amplifier and studied its characteristics as a laser oscillator and amplifier.

A photograph of the A-amplifier is shown in Fig. 7-23. The laser cell and electron-beam diodes are housed in a lead shield at the center of the machine. Extending to either side are triaxial, positively charged Blumleins with their associated Marx banks located in the oil-filled rectangular tanks at the ends.

A cutaway view of one side of the system is shown in Fig. 7-24. The pressurized laser cell is separated from the evacuated diode chamber by a 13- μ m thick, Inconel 750-x foil supported by a ribbed support structure of 90% geometric transmission. The diode gap (13-mm wide) is formed between a hexagonal stainless steel honeycomb cathode and an aluminized Kapton^R foil attached to the back side of the ribs on the foil support structure. Honeycomb material was selected for this cold cathode in order to provide the gross field enhancement required for uniform illumination of the anode foil in the presence of electric field strengths of order 200 kV/cm. Electron pinhole images of this cathode indicate that an average of 3 to 4 emission sites are created in each hexagonal cell during the pulse. Illumination of radiochromic film by the electron beam and subsequent scanning of the film on

Fig. 7-23. The RAPIER amplifier showing the laser cell and electron-beam diodes housed in a lead shield at the center of the machine. Extending to either side are triaxial, positively charged Blumleins (blue cylinders) with their associated Marx banks located in oil-filled rectangular tanks at the ends.

Fig. 7-24. Cutaway sketch showing the internal parts of one side of the RAPIER A-amplifier.

Fig. 7-25. Electron-beam uniformity along the 500- and 100-mm sides of the A-amplifier pump beam. The optical density (OD) of radiochromic film is proportional to the beam current density. The flat portion of the beam has a current density of 120 A/cm^2 .

Fig. 7-26. Typical A-amplifier current and voltage performance diagnostics indicating a significant droop of the voltage waveform as a result of this method of prepulse control.

an optical densitometer result in the data shown in Fig. 7-25. This indicates that the requisite 10×50 -cm beam is created. Carbon slab calorimeters assembled in the laser cell have shown that 650-J electron beams are delivered by each of the opposed machines.

Referring again to Fig. 7-24, the electrical pulse is delivered to the diode by a positively charged triaxial Blumlein, where the two nested coaxial pulse-forming lines are charged in series. The outer line is charged through a metal rod (labeled "HV input") that penetrates from the rear to the middle cylinder; the inner line is charged through an inductor located at the front of the Blumlein. Water is used as the dielectric material in the Blumlein and is circulated from a reservoir where the water is de-ionized and filtered such that the resistivity is kept

$\leq 10 \text{ M}\Omega \cdot \text{cm}$. The output impedance of the line is 5Ω , matching the nominal impedance of the diode.

Since the charging inductor voltage drop appears across the diode during charging (an excursion from +35 kV to -20 kV at the cathode), certain precautions must be taken to avoid unwanted emission in the diode. This emission can result from the prepulse field and subsequent early closure of the diode. In particular, a stainless steel band is placed around the edge of the cathode to control field enhancement. In addition, the duration and amplitude of the prepulse field are controlled by limiting the charging time to $1 \mu\text{s}$ and putting the $1.5\text{-}\mu\text{H}$ charging inductor in parallel with a $100\text{-}\Omega$ copper sulfate resistor (not shown in Fig. 7-24). Typical current and voltage diagnostics are displayed in Fig. 7-26. Note that a significant "droop"

Fig. 7-27. Calculated contours of electron-beam energy deposition uniformity in a 2-atm laser mixture.

of the voltage waveform is a result of this method of prepulse control. Alternative schemes, involving somewhat increased complexity of the design, include the use of a prepulse switch or a system for balanced charging of the lines.

Upon charging the Blumlein with the four-stage Marx bank, the line is discharged by the firing of the annular output switch located at the rear of the Blumlein (see Fig. 7-24). This switch consists of two annular electrodes, separated by a 10-mm gap, and a disc-shaped trigger electrode at the midplane. The trigger electrode is resistively biased at one-half the charge voltage. The switch operates at 300 kV when pressurized to 0.79 MPa (114.5 psi) with SF_6 gas. The trigger pulse has an amplitude of 100 kV with a rise time of ~ 10 ns and is coupled to the trigger electrode through an oil-insulated ring capacitor. Details on the timing and triggering of the device appear below in a discussion of the injection-locked oscillator.

It was anticipated that the injection of electron beams from two sides into the nominal $10\text{-} \times \text{ } 10\text{-} \times \text{ } 50\text{-cm}$ A-amplifier gain medium would provide fairly uniform volumetric excitation. A calculation using the three dimensional Monte Carlo code SANDYL²⁶ was performed for the appropriate conditions. The results, shown in Fig. 7-27, indicate that energy is deposited over the $10\text{-} \times \text{ } 10\text{-mm}$ injection-locked, narrowband oscillator.²⁷ A confocal unstable resonator cavity was formed between

a 15-cm diameter meniscus mirror of 350-mm radius of curvature (concave surface reflectivity of 82% at 249 nm) and a 1.5-cm² feedback mirror with a 50-cm radius convex surface of 99% reflectivity. Fused silica Brewster windows were used intra-cavity to contain the laser mixture.

A narrowband (5 cm⁻¹) KrF-discharge laser source in an oscillator amplifier configuration was used to cause the A-amplifier to fire and to injection-lock the device. Approximately 10% of the 200-mJ output of the discharge laser was used to trigger a SF_6 spark gap by focusing the pulse through one electrode such that the laser-produced spark bridges the gap. This will be discussed further in a later section. The Marx banks on the A-amplifier were erected when the discharge laser excitation was initiated. During the period of Marx erection and charging of the Blumleins, coaxial cables leading to each of the trigger coupling capacitors on the Blumleins were charged to +200 kV. These cables were tied to a common spark gap which was activated by the laser-triggered spark gap. Thus, the output of the discharge laser was used to directly control the firing of the A-amplifier output switches. More recent versions of this system use direct laser triggering of the common spark gap.

The remainder of the discharge laser pulse was optically delayed by 100 ns and then injected through the coating on the meniscus mirror of the unstable resonator cavity. This allowed the injected pulse to fill the feedback mirror and expand to fill the aperture after one round trip.

Injection of a pulse with energy greater than 100 μJ at the beginning of the gain pulse in the cavity has been observed to have a dramatic effect on output characteristics of the system. The output spectrum, beam quality, pulse duration, and hence the output energy, all change due to the injection. With no injection flux in the laser cavity, the output builds up from the broadband spontaneous emission. In order to control the output bandwidth of the laser, the injected power must be greater than the spontaneous emission source radiated into the appropriate solid angle. A simplified model can be used to estimate the power required to control the output. The spontaneous emission source is taken to be the radiation from a slab of the gain medium located along the axis with a cross-sectional area equal to that of the scraper mirror and stretching over one gain length. The proper solid angle is that

Fig. 7-28. A-amplifier output spectrum as a function of different levels of injection. Input: 20 ns, 4 cm^{-1} ; output: 50 ns, 21 J.

substantiated by the scraper mirror across the length of the cavity. Such an estimate gives $\sim 4 \times 10^3 \text{ W}$ as the power of the spontaneous emission source which the injected signal must exceed. This is in agreement with the observed minimum power requirement. Figure 7-28 shows the spectrum of the A-amplifier output for different levels of injection.

Since the build-up time of the unstable resonator cavity is comparable to the duration of the gain, the injection decreases the build-up time and results in increased output pulse length (Fig. 7-29). Also, with sufficient injection, the far field pattern does not evolve from undirected spontaneous emission but starts out at nearly steady-state final value. The characteristic divergence of the output of this system was observed to be 100μ radians and is determined by the imperfections in the optical surfaces (Fig. 7-30). The maximum laser energy extracted under these conditions, as measured by a surface absorbing calorimeter, was 25 J in a 50-ns pulse. The energy deposited in the medium was measured by pressure rise techniques using a Kulite model HKM-375-25A pressure transducer mounted directly to the top of the laser cell. The volume of the cell, which was optically inaccessible, was taken into account in arriving at the appropriate value of deposited energy. The measured value of deposited energy agreed with the SANDYL code prediction to within 10%.

Upon varying the pressure from 1 to 2 atm in the laser, the intrinsic medium efficiency results

Fig. 7-29. Effect of injection on cavity buildup in the A-amplifier. Seeding pulse yields longer laser pulse duration.

shown in Fig. 7-31 were obtained. It must be noted that the extraction efficiency from a medium of this type is dependent upon pumping rate and cavity losses, including output coupling. The particular conditions encountered above (pumping rate:

Fig. 7-30. Photograph of the far-field pattern of the A-amplifier as an injection-locked, regenerative amplifier.

Fig. 7-31. Measured medium efficiency for the A-amplifier under conditions described in the text.

1 MW cm⁻²; meniscus mirror reflectivity: 82% to allow significant injected-pulse power for gain and line-width control; and cavity magnification: 7 for low feedback of 2.2% are not necessarily optimum for extraction efficiency. The low injection-locking thresholds suggest that cavity optimization can be performed without loss of gain control. This increases the extraction efficiency.

The output power of a quasi-cw laser, such as KrF, can be increased by extracting the available

energy from an amplifier in a series of consecutive angle-coded pulses. These pulses can then be propagated along different optical paths and meet simultaneously on a target. We shall refer to this technique here as pulse stacking. This approach has been proposed also for use on other laser systems, e.g., HF. In the case of RGH laser systems, it can be used in conjunction with backward Raman amplifiers for even greater power gains.

In order to demonstrate the feasibility of this approach, a 3-times stacking experiment was performed on the A-amplifier. The duration of gain in the A-amplifier is 60 ns and the energy was extracted in three 20-ns beams. When propagated far enough from the amplifier, the beams were completely separated in space and individually diagnosed. In order to reduce the input requirement, the amplifier was used in a three-pass configuration, as shown in Fig. 7-32. The lower 15-mm portion of the amplifier aperture is used as a preamplifier for the three input beams. The beams then strike a convex mirror and expand to fill the full aperture. A concave mirror recollimates the beam and returns it through the gain volume.

The experimental set-up for the stacking experiment is shown in Fig. 7-33. The oscillator pulse is generated by the same discharge laser system used for injection locking. The output of the oscillator triggers the common gap and causes the Blumleins to fire. After an appropriate optical delay, the output is split into three beams which are sent to the A-amplifier. The first beam goes directly into the amplifier while the second and third beams are delayed an additional 20 ns and 40 ns, respectively. At all times during the gain pulse, there is an input into the amplifier. The timing is adjusted by changing the optical time delay before the laser-triggered spark gap.

At the output of the A-amplifier, the beams come out with angular separation of 4 mrad. The optical setup causes each beam to start focusing. After 20 meters of propagation from the amplifier, each beam is reduced in size by a factor of two. The beam centers are separated from each other by more than twice the beam width. A burn pattern caused by these beams on black photographic paper shows completely separated beams (Fig. 7-34). The single-pass gain of the amplifier is e⁵. In the absence of an input, this means the three-pass gain is sufficient to give a superfluorescent output with higher beam divergence and quite different time dependence. An

Fig. 7-32. A three-pass, expanding-beam optical configuration used for stacking experiments.

input energy of 10 mJ per beam was sufficient to completely suppress the superfluorescence and to drive the amplifier into hard saturation. The output pulse shapes, as seen by one photodiode, show output pulses of the same duration as the input pulses. The shape, however, is modified somewhat by saturation.

The burn patterns, shown in Fig. 7-34, effectively reproduce the shape and position of the oscillator beams that propagate through the A-amplifier without turning on the gain. Looking at one of the output beams in the far field (Fig. 7-35), we see that here also the pattern, caused by distortion due to optical surfaces and use of spherical mirrors off-axis, has the same shape and position with and without the gain. This means that the excitation is uniformly distributed through the aperture and no undesirable index gradients are present.

The total energy of the three beams, including the known optical losses, was measured to be 20 J when the amplifier was operated at 2 atm. Considering the fact that the lower 15% of the aperture was obscured by the optics for the triple pass, this output is comparable to that obtained with the A-amplifier when operated as an injection-locked oscillator with an unstable resonator.

In conclusion, we have demonstrated that a KrF laser of high gain and moderate-sized aperture can be used efficiently both as an injection-locked oscillator with an unstable resonator and as a triple-pass amplifier for a pulse stacker. Such lasers generate pulses with controlled beam quality and line width. We shall now proceed to use the stacker output beams for a combined stacker-Raman compressor experiment.

Fig. 7-33. Layout for the three-pass, three-stack experiments on the A-amplifier.

Fig. 7-34. Output beams from the A-amplifier stacker experiment shown as they separate (on burned photographic paper).

B-Amplifier Line and Switch Design. The pulsed-power system for the RAPIER B-amplifier is intended not only to satisfy the specific input re-

quirements of the RGH laser medium but also to address several issues related to the construction of even larger laser systems. The overall approach we are using is an extension of the A-amplifier system.

Fig. 7-35. Photograph of the far-field intensity distribution of the A-amplifier as a three-pass stacker.

This involves a Marx generator-charged, water-cooled, pulse-forming line which drives a cold cathode electron-beam diode. We are making design improvements to permit more reliable operation at higher voltages, longer pulse lengths, and larger electron-beam areas. The width of the entire pulsed-power package is being minimized so that modules can be placed side-by-side to produce large-area electron beams.

The B-amplifier system will consist of six separate, though synchronized, pulse-power modules which produce six electron beams. Three beams will enter the laser medium from two opposing sides, as shown in Fig. 7-36. The characteristics of the B-amplifier system are summarized in Table 7-4, together with the corresponding characteristics of the A-amplifier system. The pulse-forming lines for each system are nearly the same diameter, though the B-amplifier lines will deliver more than twice the

power and nearly six times the energy of the A-amplifier lines. The entire system will deliver 17 times the energy of the A-amplifier and occupy little more than twice the floor space.

The increase in power output results from several improvements which permit operation at higher voltage. The output voltage is limited by breakdown in the water dielectric and along water-lucite interfaces of the pulse-forming line. Since these breakdown electric fields are time dependent, the line is most susceptible to breakdown during the charging of the line by the Marx generator. The highest field in the A-amplifier line occurs at the end of the intermediate conductor, as shown in Fig. 7-37(a). The peak field in this region exceeds the average field in the dielectric by a factor of 2. In the B-amplifier, we will control this field by inserting a shaped piece of plastic at the end of the line in place

Fig. 7-36. B-amplifier system.

Table 7-4. Pulsed power system characteristics.

	B	A
Voltage	450 keV	300 keV
Current	90 kA	60 kA
Impedance	5 Ω	5 Ω
Pulse length	150 ns	60 ns
Number of modules	6	2
Energy stored		
Marx		
Per module	15 kJ (available)	2 kJ
Total	90 kJ	4 kJ
Pulse forming line		
Per module	6 kJ	1.1 kJ
Total	36 kJ	2.2 kJ
Beam		
Per module	25 $\times$ 42 cm ²	10 $\times$ 50 cm ²
Per side	25 $\times$ 125 cm ²	10 $\times$ 50 cm ²
Output current density	70 A/cm ²	100 A/cm ²

of the metal ring, as shown in Fig. 7-37(b). The difference in dielectric constant between the water ($\epsilon = 80$) and the lucite ($\epsilon = 3$) causes the field to be distributed nearly parallel to the interface. This re-

duces the ratio of peak-to-average field to 1.1. The shape of the lucite reduces the field at the end of the conductor to a very low value in the water. Alignment of this feature with the line is not particularly critical. The electric field within the lucite (600 kV/cm) greatly exceeds the field in the water (100 kV/cm) but is well below the breakdown strength of the lucite (2 MV/cm). This approach substitutes an interface breakdown for a bulk breakdown but results in a significant improvement in performance.

We have made several changes in the triggered output switch to permit operation at higher voltage and to correct problems encountered in the previous design. The peak electric field in the water along the switch insulator surface is 1.5 times larger than the average field in the water for the A-amplifier design, shown in Fig. 7-38(a). By orienting this surface in the radial direction, as in Fig. 7-38(b), this ratio is reduced to 1.1. The changes in the switch electrodes, and the shape of the inside surfaces of the switch insulators, reduce the ratios of the peak-to-average fields along these surfaces from 1.6, in the previous

Fig. 7-37. Field grading at end of intermediate conductor.

Fig. 7-38. Blumlein output switch design.

Fig. 7-39. Blumlein charging circuits.

design, to 1.1, for the new switch. The electrode material will be changed from stainless steel to a copper-tungsten composite which has superior erosion characteristics. We have relocated the bolts that hold the switch together from outside the switch, where they were immersed in water, to inside the switch, where they are insulated with oil. The better match between the dielectric constant of the bolts and the insulating material should allow us to use mechanically better materials for the bolts. These changes should lead to more reliable operation at the higher operating voltage of the B-amplifier system.

The longer output pulse is obtained by increasing the physical length of the water-filled pulse-forming line. As a consequence of this change, we must alter the method of charging the line from the Marx generator. The equivalent circuit for the A-amplifier charging approach is shown in Fig. 7-39(a). For clarity we have separated the two transmission lines which form the Blumlein pulse-forming line. Energy flows from the Marx generator

through the primary charging inductor and into the outer line. Charge is transferred to the inner line through the secondary charging inductor. Temporal changes in the charging current produce a voltage drop across this inductor which also appears across the output of the machine. This charging prepulse voltage can lead to emission in the diode with disastrous consequences. The prepulse voltage can be reduced by lowering the secondary charging inductance; however, this inductance appears across the diode load during the output pulse and diverts energy from the diode. The lower the secondary charging inductance, the more energy diverted from the diode into the inductance. In the A-amplifier, we found a reasonable compromise between these conflicting requirements. However, both effects are exacerbated by longer pulse lengths. The charging time must be kept constant to maintain the breakdown strength of the dielectric, thus, more current must flow through the secondary inductance to charge the inner line. This increases the prepulse. The longer pulse length gives more time to divert energy from the diode. In the B-amplifier, we will use the balanced or symmetric charging circuit,

shown in Fig. 7-39(b). If the two sides of this current are identical, no prepulse is produced during the charging of the lines. The inductor to the right in Fig. 7-39(b) appears across the output pulse. However, it can be much larger than the secondary inductor in Fig. 7-39(a). Mutual inductance between the two charging inductors, produced by spatial coupling can improve performance even further.

The area of the electron beam produced by each module will be as large as is consistent with beam pinch. Beam pinch results from the deflection of individual beam electrons in the magnetic field produced by the beam current flow. If the electric field in the diode is parallel to flow of the electron beam, the deflection is inward and can be calculated analytically as $\sin \theta = d/r_L$, where θ is the angle between the electron trajectory and the normal-to-the-anode plane, d is the distance from the cathode to the anode, and r_L is the electron gyroradius evaluated at the output energy of the electrons. If we apply the analytic formula for a rectangular beam to the parameters of the B-amplifier, then, the maximum beam area is 200×420 mm. However, the analytic treatment ignores the compensating influence of transverse electric fields near the edges of the beam. We performed experiments in 1977 at 1 MeV and 300 A/cm^2 that demonstrated these fields to be effective in counteracting beam pinch, as in Fig. 7-40. It was demonstrated that these fields would permit the production of beams larger than indicated by the analytic model. We will verify this at the B-amplifier parameters early in development program.

Timing. Laser triggering of gas spark gaps can be a useful technique for low-jitter synchronization in pulsed-power systems.²⁸ Some designs in the literature use a laser focused on the surface of an electrode to form a plasma that acts as a trigger mechanism. Others depend on volume effects in the gas between the electrodes.

Spark-gap designs for very high electric fields with low inductance often use SF_6 gas because of its superior dielectric properties with dc fields. It is an interesting accident of nature that SF_6 also has a low breakdown threshold for ultraviolet irradiation. Figure 7-41 shows the gas breakdown threshold²⁹ for several common gases irradiated with 20-ns pulses from a 248-nm KrF laser. Gas breakdown is defined as a significant attenuation (about 10%) of

Fig. 7-40. Electron trajectories in diode with transverse electric fields.

Fig. 7-41. Breakdown thresholds for several common gases irradiated with 20-ns pulses of 248-nm radiation.

the energy transmitted through a focus in the gas, when observed in the far field. The intensity threshold for SF_6 is approximately the same when breakdown is defined as the appearance of a visible plasma in the focus. The breakdown intensity for

Fig. 7-42. Laser-triggered pulser used for delay and jitter studies.

SF_6 varies from $3 \times 10^9 \text{ W/cm}^2$ at 1 atm to $8 \times 10^9 \text{ W/cm}^2$ at 10 atm. This covers the pressure range of most interest for spark-gap switches.

We have studied the behavior of a KrF, laser-triggered, SF_6 gap with a low inductance pulser, as shown in Fig. 7-42. A 20-ns KrF laser pulse is focused through a 2.5-mm hole in one electrode of the gap using a lens of 200-mm focal length. The laser beam produces a spark with a length of several millimeters in the gap between the electrodes. It exits through a similar hole in the other gap electrode. This avoids electrode surface plasmas and ensures that all of the observed triggering is due to volume effects in the SF_6 . The gap spacing between the electrodes is 5 mm for the data presented here but the results are similar for a 10-mm gap. The spark gap discharges a 0.03- μF capacitor, charged to 60 to 120 kV dc through a 50- Ω copper sulfate resistor load and a current viewing resistor.

Detailed jitter and time delay measurements were made with the pulser charged to 80 kV dc and pressurized to a SF_6 pressure of 3 atm. This was 120% of the pressure at which spontaneous dc breakdown was observed for this charge voltage. The input laser pulse has a full width at half maximum of 20 ns, a 10 to 90% rise time of about 5 ns, and an energy of 7 mJ. Figure 7-43 shows twenty overlapping shots of the pulser output current rise with the gap triggered under these conditions. From

Fig. 7-43. Twenty overlapping output current pulses (2 ns/division). Conditions are as described in the text. Pulses are from the pulser shown in Fig. 7-42.

this we infer a jitter standard deviation of approximately 100 ps. The time delay between the 50% point of the laser input pulse and the 50% point of the current rise was about 7 ns.

The time delay to gap closure depends upon the position of the laser beam with respect to the hole in the electrode, as shown in Fig. 7-44. A spark near the edge of the hole produces minimum delay and jitter; the data presented above correspond to this

Fig. 7-44. Variation in delay as the laser breakdown channel is translated across the diameter of the laser input hole. Zero is at the center of the hole.

alignment. If the spark is centered on the 2.5-mm hole, the delay increases by about 3 ns and the jitter standard deviation increases from 100 to 300 ps. We suspect that the additional delay and jitter come from the time required to break down the gap between the edge of the hole in the electrodes and the ionized channel of the laser spark. A smaller laser input hole would, therefore, be preferable in applications requiring very low jitter, at the cost of a more critical alignment.

An increase in laser pulse energy from 7 to 50 mJ has no perceptible influence on delay or jitter when the spark is near the edge of the hole in the electrode. This causes a slight reduction in the larger delay and jitter observed when the spark is in the center of the hole. We have not made careful observations of switch performance with input energy less than 7 mJ. Jitter begins to increase at that level for sparks near the center of the input hole. It continues to increase as the input energy is further reduced. An increase in pressure above 120% of the self-break pressure increases both delay and jitter.

At 400% of the self-break pressure, the delay increases by about 5 ns; the jitter standard deviation increases to about 1 ns.

We have used a KrF laser-triggered switch to synchronize the firing of a KrF discharge laser to the A-amplifier. This was accomplished using the techniques discussed above. We feel this technique has great promise for reliable triggering of larger electron-beam pumped laser modules for fusion. This approach was used in the A-amplifier experiments described previously in this section. A schematic of the system is shown in Fig. 7-45. A fraction of the output of the probe laser is used to trigger a spark gap which generates an electrical pulse. This pulse in turn drives a "common" switch which generates two identical pulses suitable for triggering the main switch on each of the two Blumleins of the A-amplifier. The main portion of the probe beam is delayed sufficiently before being applied to the amplifier. This provides for the transit times in the Blumlein and triggering system. This

Fig. 7-45. Injection-locking setup for RAPIER A-amplifier.

system has worked extremely well in providing synchronization between the optical and electrical systems. Recently, the common switch has been laser-triggered directly with similar results.

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Large Energy Pulse Compression Systems

Rare-gas-halide excimers (KrF, XeF, XeCl) are efficient ultraviolet laser media. However, lasers that use these media are unable to store energy for longer than their upper laser-level lifetimes, typically ≤ 10 ns. The direct use of these lasers to generate the 10- to 20-ns pulses required for inertial confinement fusion would be inefficient and expensive. Use of these lasers is further complicated by the fact that nonsaturable photoabsorption losses within these laser media limit the generated output intensity of a power amplifier to values of approximately 10 to 20 MW/cm², at intrinsic medium efficiencies of 8 to 10%. Energies of interest for fusion can only be obtained for pulse lengths greater than a few hundred nanoseconds. Thus, some techniques of pulse compression and intensification with factors of order 10 are required, if these media are to be used in fusion applications.

We have studied several techniques for producing short pulses with RGH laser media. These include:

- Raman compression.
- Pulse stacking or angle multiplexing.
- Hybrids which combine both pulse stacking and Raman compression.

An important issue in pulse stackers is the cost and complexity of the optical system. In this section, we discuss a novel, pulse-stacking optical train that

Fig. 7-46. Schematic of pulse-stacking optical train. This system gives a compression ratio of 16, using a large-aperture KrF-laser amplifier.

may be used in pure stacking and hybrid compression systems. The number of large mirrors is minimized in this system.

We are currently analyzing this optical train, and other variations, to determine performance characteristics in pure stacking and hybrid stacker-compressor, fusion-laser systems. The optical train is shown schematically in Fig. 7-46. Telescopic beam-transfer optics control each beam cross section in a manner that simultaneously maximizes beam fluence and amplifier extraction efficiency. The system shown in Fig. 7-46 gives a compression ratio of 16, using a large-aperture, KrF-laser amplifier.

Compression ratios of 40 to 50 can be achieved by simply increasing the number of reflectors and suitably increasing the dimensions of the machine. To operate the system (Fig. 7-46), a short pulse from an appropriate master oscillator-preamplifier system is propagated onto an array of partially transparent mirrors. It is then divided into 16 equal

intensity pulses. These pulses are sequenced in time by reflection from another set of mirrors, as shown. For example, if the pulse length of the injected signal is 20 ns, the effective pulse length, after beam splitting and delay, will be 16 times larger, or 320 ns. The temporally sequenced pulses are reflected from a convex mirror and expanded to fill the amplifier aperture, as shown.

After passing through the amplifier once, each pulse encounters a large concave mirror that returns the pulse through the amplifier for further amplification and then directs it onto a second convex mirror. This mirror recollimates each beam and directs it to an array of plane reflectors. These mirrors are positioned such that, after reflection from the array, the beams are parallel to each other and all the pulses are temporally coincident. The final pulse length is once again 20 ns. However, the amplifier has been operating for 320 ns, and the output flux for each 20-ns amplifier pulse is about

Fig. 7-47. Optical error budgets for pulse-stacker system with a compression of 25. Individual beam divergence versus position for 4×5 array.

12 MW/cm². The total output fluence of the power amplifier is about 4 J/cm². Thus, if the aperture of the output array measured 2 m $\times$ 2 m, the output energy would be about 160 kJ.

The optical train for such a system would be contained within a compact 5- $\times$ 10- $\times$ 80-m space frame. Six such final amplifier configurations would produce almost 1 MJ of energy. The configuration of a single beam is shown in Fig. 7-46. By expanding and focusing each stacked beam, the optical flux is made nearly constant during the pulse-stacking process. This unique feature minimizes the size of each optical element, thereby reducing cost and system complexity. Furthermore, the spherical mirror tilt angles are chosen such that the wavefront distortions introduced by each tilted surface are corrected in the output beam. This allows a further reduction in the length of the pulse stacker without degrading the output beam wavefront.

Optical design calculations for the system shown in Fig. 7-46 indicate that astigmatism, defocus, and coma are the principal optical aberrations introduced by the optical train. As shown in Fig. 7-47, the maximum divergence caused by these aberrations is approximately 10 μrad . This value is comparable to contributions from refractive gas scattering by gas density fluctuations in amplifier and beam transport tubes. The angular beam divergence required for target irradiation is roughly 100 μrad . Therefore, optical aberrations appear to be manageable for this system. However, our analysis of laser pulse propagation in this stacker system indicates that scattering, diffraction, and

refraction processes will cause energy from one channel to enter into another channel. Since the channels are staged in time, this "crosstalk" energy appears as prepulse energy in several channels and can lead to destruction or alteration of target characteristics before the main laser pulse arrives. In Fig. 7-48 we plot the crosstalk scattering efficiency per solid angle, $d\eta/d\Omega$, as a function of the angular separation between channels for several crosstalk sources. Our analysis of this potential problem indicates that beam ripple (or edge diffraction), gas fluctuations, and microirregularity-scattering are potentially serious sources of channel crosstalk for stackers of the type illustrated in Fig. 7-46. For a 1-MJ laser pulse focused with a lens of focal length, $f_1 = 30$ m, onto a target of radius 10 mm, the prepulse energy on target is

$$\epsilon(J) \sim 0.35 \, d\eta/d\Omega \, (\text{sr}^{-1}) N_s \quad (24)$$

where N_s is the number of scattering channels or stacked pulses. Consequently, prepulse energies of 10 J, or greater, may occur in MJ-class systems of this type.

Suppression of laser prepulse energy can be achieved, in principle, by placing a saturable absorber at the output of the system. One generic class of saturable absorbers for ultraviolet lasers is based on photodissociation. From this class, ozone appears to be a suitable candidate for a KrF pulse-stacker system. Ozone is photodissociated by KrF laser light into $\text{O}(^1\Delta)$ and $\text{O}_2(^1\Delta)$. The absorption coefficient is large: approximately 0.17/cm/Torr. The

Fig. 7-48. Dependence of pulse-stacker, crosstalk, source-scattering efficiency on channel angular separation. The scattering efficiency, $d\eta/d\Omega$, at angle, θ , is the fraction of the total energy of a channel that appears in a second channel when separated by angle, θ , and centered on solid angle, $d\Omega$.

corresponding saturation fluence is also large: 166 mJ/cm^2 . Therefore, the fractional beam photoabsorption losses may be high unless the beam fluence is relatively high: $>5 \text{ J/cm}^2$. This places some stress on the damage and survival capabilities of optical elements. Reconstitution of the medium appears possible if O_3 and O_2 densities are approximately $10^{16}/\text{cm}^3$ and $10^{19}/\text{cm}^3$, respectively. The key unknowns for this particular saturable absorber system are the optical properties

of $\text{O}_2(^1\Delta)$ at high optical fluxes. When pulse-stacker systems are used in hybrid Raman compression schemes, the prepulse problem is essentially eliminated during the Raman compression process.

In summary, we have identified and analyzed a novel, pulse-stacking, optical train that shows promise in fusion laser systems employing pulse stacking or stacking and Raman compression. This novel system utilizes telescopic optical transfer

Fig. 7-49. Pulse compression efficiency plotted as a function of total system compression for three compression techniques.

Fig. 7-50. Fusion-driven, hybrid-stacker compressor design for a stacker-Raman compressor.

techniques to simultaneously maximize beam fluence and amplifier extraction efficiency. This, in turn, minimizes mirror surface area and cost.

Other designs have been identified which use regenerative extraction techniques to greatly simplify the input optical train. As shown in Fig. 7-46, stackers can achieve large and efficient compressions that yield high laser efficiencies ($>5\%$).

The potential advantages of stacking and compression can be combined to achieve large efficient compressions (Fig. 7-49) in a hybrid stacker-Raman compressor system.^{30,31} For example, a pulse stacking of 4 to 12 to create 50- to 100-ns KrF-laser pulses, combined with Raman compression of these pulses to generate the 10- to 20-ns target irradiation pulses, appears feasible. Relatively small (4 to 5) Raman compressions are used in order to achieve high compression efficiency ($\sim 80\%$) without use of second Stokes absorbers. In hybrid systems, two-pass extraction in the Raman cell can be used to achieve amplifications of 1000 at high efficiency (~ 70 to 80%) without the use of second Stokes suppression. This greatly simplifies the front end in the Raman compressor stage of this system. An example of such a hybrid system, combining pulse stacking and multipass Raman conversion, is shown in Fig. 7-50. A more detailed discussion of the performance characteristics of this system may be found in the next section.

At present, the most promising KrF-pulse compression systems are pure pulse stackers and hybrids. Both approaches appear to be capable of efficient ($>5\%$) and high-energy (>1 MJ) operation.

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Advanced Concepts and Supporting Research

At the present time, rare-gas-halide excimer lasers are the principal focus of the advanced laser program. We are continuing, however, a small ef-

fort to study less-developed laser candidates and techniques that may offer either higher efficiency, better target coupling, or less complexity than the rare-gas-halide stacker-compressor systems. For example, the free electron laser may offer high efficiency and high average power along with the elimination of expensive and vulnerable final-focusing optics. New solid-state systems, such as V^{2+} : MgF_2 , may offer efficient and inexpensive flashlamp pumping and simple optical designs. Broadbanding of fusion lasers, using stimulated rotational Raman scattering, may provide improved target coupling for present fusion lasers as well as possible applications in the future. These, and several other candidates, either new lasers or possible improvements in well known systems, are the subject of continuing small-scale research.

Free-Electron Laser for Fusion

We are continuing to search for advanced lasers that offer significant advantages over KrF-Raman compressor-stacker systems. One very different, but attractive, concept is based on the free-electron laser (FEL). This device directly converts a high-quality electron beam into coherent radiation. It has long been known that such conversion was possible,³² but only recently has it been shown that efficient, high-pulsed-power FEL amplifiers might be built. These new FEL amplifiers are theoretically capable of extracting a large portion of the electron-beam energy on a single pass. This eliminates the need for the electron-storage ring included in many other FEL designs. We are analyzing the FEL in light of these new developments so that we may define the major physics issues and determine the feasibility and technological requirements for utilizing a high-power FEL for fusion applications.

Our main efforts have been directed toward the construction of a numerical model of the FEL that allows us to rapidly design the amplifier and then accurately simulate its performance. As part of this program, we are also investigating the requirements for small-scale FEL experiments that provide the data necessary to design and build a fusion-class FEL amplifier.

Theory of Operation. The major components of an FEL system are a laser oscillator, an electron accelerator, an FEL amplifier (wiggler magnet), and an electron-beam dump for energy recovery. These

Fig. 7-51. Major components of a high-power free-electron laser (FEL) system.

are shown schematically in Fig. 7-51. The FEL can be thought of as a coherent electron decelerator. In contrast to the usual longitudinal accelerating fields found in linac microwave cavities, the FEL decelerating field is formed from a transverse static magnetic field (the wiggler field) and the amplified laser signal field. These two fields interact to form a moving potential well ("bucket") which influences the motion of the electron. If the electron can be kept in the potential well throughout the length of the FEL amplifier, the radiation field will grow and the electron will lose energy. This is accomplished by precisely adjusting the wiggler wavelength (λ_w) and magnetic field strength (B_w). The details of trapping and decelerating electrons in buckets have been treated elsewhere.³⁻³⁵ We will merely give the results of those studies which serve as a basis for our FEL simulations.

Equations (25) and (27) specify the motion of the electron:

$$\frac{d\gamma}{dz} = -\frac{e}{k_w} \frac{w}{k_w} \sin \psi, \quad (25)$$

$$\begin{aligned} \frac{d\psi}{dz} = & k_w - \frac{k_s}{2\gamma^2} \left[1 + \left(\frac{b_w}{k_w} \right)^2 \right. \\ & \left. + 2 \frac{e_s b_w}{k_w k_s} \cos \psi + \left(\frac{e_s}{k_s} \right)^2 \right] + \frac{d\phi}{dz}, \end{aligned} \quad (26)$$

$$\psi = \int (k_w + k_s) dz - \omega_s t + \phi. \quad (27)$$

Equations (28), (29), and (30) specify the behavior of the electromagnetic field:

$$E = E_s(z) \cos [k_s z - \omega_s t + \phi(z)] \quad (28)$$

$$\frac{dE_s}{dz} = \frac{Z_0 b_w}{\sqrt{2} k_w} J \left\langle \frac{\sin \psi}{\gamma} \right\rangle_{\text{all electrons}} - \alpha E_s \quad (29)$$

$$\frac{d\phi}{dz} = \frac{Z_0 b_w}{\sqrt{2} k_w} \frac{J}{E_s} \left\langle \frac{\cos \psi}{\gamma} \right\rangle_{\text{all electrons}} \quad (30)$$

All variables are defined in Table 7-5.

These equations describe the motion of any electron in the specified fields providing these fields do not undergo large amplitude changes during a wiggler period. A complete description of the FEL amplifier is obtained by solving Eqs. (25) and (26) for all electrons in the beam and simultaneously performing averages over all electrons in order to

Table 7-5. Notation, MKS units. Subscript r labels resonant particle.

A_{Λ}	= bucket area
B_w	= wiggler magnetic field
b_w	= $eB/mc\sqrt{2}$, reduced wiggler field (m^{-1})
c	= speed of light
E_s	= laser (signal) electric field
e_s	= $eE_s/\sqrt{2} mc^2$, reduced laser field (m^{-1})
e	= electron charge ($e > 0$)
I_0	= input laser flux
J	= electron beam current density
J_{Λ}	= bunched electron beam current density
k_s	= $2\pi/\lambda_s$
k_w	= $2\pi/\lambda_w$
m	= electron mass
P	= maximum energy spread of bunched electrons in units of mc^2
v	= electron velocity
z	= longitudinal coordinate
z_0	= impedance of free space, 377 Ω
α	= distributed electric field loss
γ	= $(1 - v^2/c^2)^{-1/2}$
$\Delta\gamma$	= energy spread of input beam in units of mc^2
ϕ	= electromagnetic phase
λ_w	= period of wiggler magnetic field
λ_s	= laser wavelength
ψ	= generalized electron coordinate

solve Eqs. (29) and (30). This is inconvenient if one wishes to quickly design several different FEL amplifiers; hence, a simplification is required.

It has been previously shown^{33,34} that a resonant-particle approximation to Eqs. (25), (26), and (27) allows one to treat the electron distribution as two distinct classes of electrons. The first group consists of those electrons whose energy and phase satisfy the following conditions:

$$|\gamma - \gamma_r| \leq P(\psi, \psi_r) = \left(\frac{e_s b_w}{k_w^2} \right)^{1/2} \quad (31)$$

$$(\cos \psi + \cos \psi_r - (\pi - \psi - \psi_r) \sin \psi_r)^{1/2}$$

where γ_r and ψ_r are the energy and phase of the electron that maintains a constant phase throughout the FEL amplifier, i.e., $d\psi_r/dZ = 0$. This electron is called the resonant electron. Equation (31) defines a region in phase space (γ, ψ) called a bucket and labeled Λ . We define A_{Λ} to be the area of the phase-space region delimited by Eq. (31).

$$A_{\Lambda} = 2 \int P(\psi, \psi_r) d\psi \quad (32)$$

The second group consists of those electrons not within the bucket. In the resonant approximation, this second group will, on the average, not contribute to the laser field growth and can be ignored. Thus, the laser gain is only proportional to the number of electrons within the bucket. Furthermore, all of these electrons will on the average undergo energy and phase changes similar to that of the resonant electron. When we assume that a nonresonant electron exhibits behavior similar to the resonant electron, we are ignoring the synchrotron motion of the nonresonant electron. Under these circumstances, Eqs. (25) and (26) must be solved only for a single resonant particle and the field Eqs. (29) and (30) need only include the contributions from the trapped electrons. Therefore, in this approximation the expressions for field growth and optical-phase change become:

$$\frac{dE_s}{dz} = \frac{Z_0 b_w}{\sqrt{2} k_w} \frac{J_{\Lambda}}{E_s} \frac{e \sin \psi_r \Delta}{\gamma_r} \alpha E_s \quad (33)$$

$$\frac{d\phi}{dz} = \frac{Z_0 b_w}{\sqrt{2} k_w} \frac{J_{\Lambda}}{E_s} \frac{e \cos \psi_r \Delta}{\gamma_r} \quad (34)$$

where J_{Λ} is the current density within the bucket. The averages over all electrons in Eqs. (29) and (30) have been replaced by averages over the electrons in the bucket. These are easily computed if we assume that the electron beam not only is bunched with energy γ_r and phase ψ_r , but that it has a uniform phase-space density. Equations (25) and (26), for the resonant particle only, and Eqs. (33) and (34), for the laser field, now form the basis for our FEL designs. Equations (25) and (26), for all electrons, and Eqs. (29) and (30), for the field, are the basis for our simulation.

The purpose of the FEL simulation is four-fold:

- To investigate the validity of the gain equation, Eq. (33), without requiring the linearization of Eqs. (25), (26), and (27) about the resonant electron.
- To study the initial bunching of the electrons and attempt to evaluate the trapping efficiency of several different wiggler designs.

- To determine if the electrons can be kept in the bucket for a period long enough to achieve substantial signal amplification.

- To determine the stability of the FEL amplifier against input fluctuations in electron-beam voltage, current, energy spread, laser-oscillator power, and wiggler irregularities.

The simulation has several limitations. Most importantly, it is one dimensional. It does not account for any transverse inhomogeneities in either the wiggler field or the laser field. The effects of transverse field variations are discussed elsewhere.³⁵ Space charge effects have also been ignored; however, we do not expect space charge considerations to change our results significantly.³⁶ Finally, we have considered only a monochromatic field; the disadvantages (and possible advantages) of a multicolor-laser field have been ignored in the present work.

Design. An FEL design specifies λ_w and b_w as a function of distance down the amplifier. A successfully designed FEL amplifier will continuously decelerate a large fraction of the electron beam. We base our designs on the assumption that if the resonant particle is decelerated, and simultaneously the bucket area remains large, a large fraction of the electron beam will be decelerated along with the resonant particle. This leads to substantial laser gain. This assumption is tested by the numerical simulation which is described later in this article in "FEL Amplifier Simulation." Therefore, our design problem is simplified to specifying λ_w and b_w in a manner which maintains $0 < \psi_r < \pi/2$ and keeps the bucket area large.

The design problem is defined by Eqs. (25), (32), (33), and (34). There are seven independent unknowns, $(\gamma_r, \psi_r, E_0, \phi_0, A_A, b_w, \lambda_w)$ in these four equations; therefore, in order to determine a design we must have two additional constraints. These constraints, along with a set of initial conditions, are at the discretion of the FEL builder. We will only consider a few of the many possible options. A sixth equation is selected from the following list:

$$\frac{d^2 b_w}{dz} \lambda_w = 0 \quad (35a)$$

$$\frac{db_w}{dz} b_w = 0 \quad (35b)$$

$$\frac{d}{dz} \left(\frac{b_w}{k_w} \right) = 0 \quad (35c)$$

while a seventh equation is chosen to be either

$$\frac{d\psi}{dz} = 0 \quad (36a)$$

or

$$\frac{d^2 A_A}{dz} = \text{constant} \quad (36b)$$

The growing bucket option, Eq. (36b), is only practical at high-current densities.^{33,37}

The initial values of γ_r , λ_w , λ_s , ψ_r , J were arbitrarily selected to best illustrate high-power FEL amplifier performance. The initial magnetic field was chosen so that $d\psi/dz = 0$ at $z = 0$ (Eq. 26). Beam quality was specified by the parameter,

$$\Delta\gamma = (\gamma_{\max} - \gamma_{\min})/2 \quad (37)$$

The initial laser intensity was chosen to make the maximum bucket height equal to $\Delta\gamma$, that is

$$e_s(z=0) = \frac{(k_w \Delta\gamma)^2}{2b_w |\cos \psi_r - (\pi/2 - \psi_r) \sin \psi_r|} \quad (38a)$$

If we assume $b_w/k_w \approx 1$, Eq. (38a) may be rewritten as

$$I_0(z=0) = \frac{3.16 \times 10^{10}}{\lambda_s (\mu\text{m})} \left(\frac{\Delta\gamma}{\gamma} \right)^4 \text{ W/cm}^2 \quad (38b)$$

Thus, a 0.25- μm FEL amplifier, pumped by an electron beam with a 2% energy spread ($\pm 1\%$), requires a laser input intensity of $3.16 \times 10^{10} \text{ W/cm}^2$.

$A_A(z=0)$ is now calculated from Eq. (32) and $\phi(z=0)$ is set to 0. We then estimate the trapped current by

$$J_{\Lambda} = J \frac{A_{\Lambda}(z)}{2\pi(2\Delta\gamma)} \quad (39)$$

with the additional restriction that $J_{\Lambda}(z) \leq J(0)$, for all z greater than zero.

In general, the bucket can be kept from shrinking (and electrons from being detrapped) only by maintaining high laser gain, i.e., high-current density. For example, we know that FEL performance is much improved if we start with

$$J_{\Lambda} > \frac{1.4 \times 10^6}{\gamma \lambda_s^2 (\mu\text{m})} \left(\frac{\Delta\gamma}{\gamma} \right)^4 \quad (40)$$

Unfortunately, these design equations assume that the electron beam is already bunched, while a realistic simulation must utilize an unbunched beam. This discrepancy has a detrimental effect on FEL performance. We allow for an initially unbunched beam by replacing $(\sin \psi)_{\Lambda}$ and $(\cos \psi)_{\Lambda}$ in Eqs. (33) and (34) with $\tau(\sin \psi)_{\Lambda}$ and $\tau(\cos \psi)_{\Lambda}$, where we arbitrarily choose

$$\tau = 1 - e^{[\gamma_r(z) - \gamma_r(0)]/\Delta\gamma} \quad (41)$$

This allows the bunch to form slowly, partially compensating for the initial absorption of laser power by electrons not found within the bucket. After the resonant particle has been decelerated by more than a few times the initial energy spread of the beam, the resonant particle description should be more accurate and we may permit the design process to proceed as outlined above. We have not attempted to optimize τ .

The design problem is now completely defined and we merely need to numerically integrate Eqs. (25), (26), (32), (33), (34), (35a, b, or c), and (36a or b) in order to calculate $\lambda_w(z)$ and $b_w(z)$ and estimate the FEL amplifier performance $[e_s(z)]$.

We have used the procedure described above to design a high-power 385- μm FEL amplifier which might serve as an FEL test facility. We chose $\gamma = 15$, $\Delta\gamma = 0.2$, $\lambda_w = 8.7$ cm, $\psi_r = 0.4$, and $J = 100$ A/cm². These parameters imply an estimated trapped-current density of 28 A/cm² and an input laser flux of 0.67 MW/cm². In this example, we

have chosen to keep λ_w and ψ_r constant (design options 35a and 36a) and have not included any losses. These parameters put the amplifier in the high-current density regime.

Figure 7-52(a) illustrates the magnetic field profile, which in conjunction with the constant 8.7-cm period wiggler, completely specifies the FEL amplifier. Figure 7-52(b) shows the planned

Fig. 7-52. Characteristics of a 385- μm FEL: a) magnetic-field profile, b) electron-energy loss and c) laser-intensity design.

Fig. 7-53. Well (bucket area) for a 385- μm FEL.

deceleration of the resonant electron from $\gamma = 15$ to $\gamma = 10.6$ over 16 meters. Figure 7-52(c) illustrates that the laser power is expected to grow from $0.67 \text{ MW}/\text{cm}^2$ to $60 \text{ MW}/\text{cm}^2$. Figure 7-53 shows that the bucket area is also expected to increase. This confirms that the device is operating in the high-current density regime. The design appears to meet the dual goals of resonant particle deceleration and bucket growth.

FEL Amplifier Simulation. Operation of the FEL amplifier is simulated by calculating the one-dimensional trajectories of 500 electrons in the previously designed wiggler. This allows us to determine if our design procedure is reasonable. Equations (25) and (26) are numerically integrated for all 500 electrons with a specified $b_w(z)$ and $\lambda_w(z)$. The field variables are calculated from Eqs. (29) and (30) with the averages taken over all 500 electrons.

Figure 7-54(a) shows the initial energy-phase space distribution of the electrons. They are uniformly distributed in phase between $-\pi$ and π , and in energy between $\gamma = 14.8$ and $\gamma = 15.2$. Thus, this particular simulation starts with an electron beam identical to that assumed in the design stages. Initial laser intensity is also set to the design value ($0.67 \text{ MW}/\text{cm}^2$). Figure 7-54(b) shows that after 1.6 m the electrons are beginning to bunch. Some of the electrons have been decelerated, while others are

actually accelerated. This implies that initially there can be either absorption or gain in an FEL amplifier depending on the initial energy of the electron beam. The solid line outlines the estimated bucket area. We expect that electrons within this region will be trapped and decelerated continuously. Figure 7-54(c) demonstrates that within 3.2 m the beam has been bunched in phase and that more electrons have been decelerated than accelerated. This implies net laser gain. Figures 7-54(d) and (e) illustrate further bunching and deceleration of the electrons within the bucket and indicate that the untrapped electrons do not appear to exchange additional energy with the fields. Finally, Fig. 7-54(f) illustrates the electron distribution after 16 m of interaction with the laser and wiggler fields. Almost all of the electrons initially caught in the bucket have been decelerated, while those that were not trapped did not exchange energy with the fields after approximately 2 m. Some detrapping began at about $\gamma = 13$ ($z = 9.5 \text{ m}$), presumably because the bucket was shrinking and the adiabatic approximation was no longer valid.

Counting the number of electrons in the bucket reveals that a higher percentage of the beam was trapped and decelerated than was estimated by design Eq. (40). This excess trapping is a general result of the high current density regime. It can be attributed to both the growing bucket and to the inclusion of the transition region of the amplifier where the design modifications expressed by Eq. (41) are important. Figure 7-55 shows that the simulation predicts a final laser intensity of almost $150 \text{ MW}/\text{cm}^2$. This is 2.5 times greater than specified by the design and is a consequence of the fact that the simulation shows much greater electron trapping than the design. The first meter of the amplifier shows negligible gain because this section serves only to bunch the electron beam. The dips in the laser intensity plot are caused by electrons revolving in phase space. The precise nature of these oscillations is dependent on the initial loading (energy and phase) of the electrons.

It is important to realize that this particular design worked very well in spite of the fact that both the trapped current and the laser field did not behave as planned. We have investigated this inherent stability further by altering the initial simulation conditions without changing the FEL amplifier

Fig. 7-54. Electron phase-space trajectories in a 385- μm FEL: a) $z = 0$, b) 1.6 m, c) 3.2 m, d) 5.6 m, e) 9.6 m, f) 16 m.

design. Figure 7-56 illustrates the fraction of electrons trapped after we have decelerated the resonant electron by 25% as a function of current density at three different initial conditions:

- Mean electron energy on resonance.
- Mean electron energy at 0.65% above

Fig. 7-55. Simulation results of laser intensity in 385- μm FEL, indicating a prediction of final laser intensity of almost 2.5 times that specified by the design.

resonance.

- Mean electron energy at 0.65% below resonance.

As shown by Fig. 7-56, high current densities appear to enable the FEL to withstand electron energy fluctuations and promote better trapping.

We have also used these procedures to design a 250-nm FEL fusion amplifier. The basic laser parameters are illustrated in Table 7-6. These parameters were obtained by using design options defined by Eqs. (35a) and (35b). The electron energy was loaded slightly below resonance in accordance with the results shown in Fig. 7-56.

We envision a fusion driver consisting of one 200-ns accelerator and 20 FEL wigglers built from permanent magnets. The 200-ns electron beam would be divided into twenty 10-ns beams. Appropriate delays would then be added to the electron beams before they enter the wigglers. This enables the resulting 20 laser beams (1 MJ) to be stacked simultaneously on target. The output laser intensity is of sufficient intensity and small enough in size (5 mm) so that we can direct the beam onto a reactor-grade pellet without final-focusing optics. This would provide a substantial simplification for the laser system.

Fig. 7-56. Percentage of electrons trapped after the resonant electron energy has decreased by 25% in a 385- μm FEL. Energy decrease is a function of current density at three initial conditions: (a) mean electron energy on resonance, (b) mean electron energy at 0.65% above resonance, (c) mean electron energy at 0.65% below resonance.

Table 7-6. FEL fusion driver characteristics.

Wavelength	250 nm
Electron energy	767 MeV
Electron current	20 kA
Electron energy spread	$\pm .3\%$
Normalized emittance (γE)	7.9×10^{-2} rad-cm
Wiggler period	13.5 cm
Wiggler magnetic field	3 kG (peak)
Wiggler length	120 m
Laser input intensity	9.7×10^9 W/cm ²
Laser output intensity	3.0×10^{13} W/cm ²
Electron beam to laser beam conversion efficiency	39%

This simplicity, coupled with reliability and a potential for system efficiency greater than 10%, make the FEL an interesting advance-laser candidate.

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Energy-Storage Solid-State Lasers—V:MgF₂

Solid-state laser media have many technical characteristics³⁸ which make them attractive^{39,40} for use in fusion-laser drivers:

- High energy-density storage (100 to 1000 J/l).
- Long storage lifetime (100 to 1000 μ s).
- Low small-signal gain coefficient (0.01 to 0.05 cm⁻¹).

These characteristics allow the possibility of designing laser systems with inexpensive, long-lived, and well developed power conditioning technology. They also permit optical extraction and alignment as well as control systems which are less complex

than those required in pulse-compressor laser systems. However, to exploit these potential technological and cost benefits in a fusion laser driver, the relatively low thermal-conductivity characteristic of dielectric solids (0.01 to 1.0 W/cm²/°C) must be accommodated in the laser design. In the "axial gradient laser" (AGL) design approach,⁴¹⁻⁴⁴ the solid state amplifier medium is configured into a parallel array of thin slabs. These are cooled by extracting heat through the slab faces by means of a flowing coolant. Although liquid coolants have been used in laser devices studied to date, gas cooling of a slab array, oriented at the Brewster angle, appears to be quite feasible in the fusion application.^{45,46} Other average-power solid-state laser design concepts have also been explored^{47,48} in recent years. Clearly, the average-power design issues can be greatly minimized at the outset, if a solid-state host medium can be found which exhibits both desirable laser kinetic properties and high-thermal conductivity as well. It is well known that laser glasses have relatively low-thermal conductivities⁴⁹ (0.01 W/cm²-°C), whereas dielectric crystals have relatively high-thermal conductivities⁵⁰ (0.3 W/cm²-°C). Considerably larger thermal conductivities can be obtained in crystals at reduced temperatures, whereas the thermal conductivities of glasses generally decrease somewhat from their room temperature values when the temperature is lowered. These simple considerations, supported by more complete and quantitative analyses, suggest that crystalline hosts are highly desirable for fusion applications. This assumes aperture scaling of the crystalline material is practical. Recent developments in the growth of large-diameter (50 cm) optical-quality single crystals⁵¹⁻⁵⁴ offer increased prospects for attaining crystals of the required size and quality. These developments have reinforced an exploratory assessment of crystalline solid-state laser media for high-average power-fusion lasers. In selecting various combinations of crystalline host, active impurity, and pump excitation source, we have set the laser system performance goals as follows:

- Efficiency: 10%.
- Pulse repetition rate: 5 Hz, or less.
- Cost per optical joule: \$100/J, at the megajoule level.

The additional requirement for low, nonlinear, refractive index directed our attention to fluoride crystalline hosts,⁵⁵ such as the alkaline earth

Fig. 7-57. Electronic structure (schematic) and spectra of vanadium-doped magnesium fluoride (V:MgF_2), showing absorption²¹ and emission⁴⁵ spectra.

fluorides. Spectroscopic requirements for broad-pump bands, a long-lived metastable state, and high quantum efficiency directed our attention to transition metal ions with d^3 or d^7 ground electronic configurations. These are well known^{56,57} for their long-lived, red-infrared fluorescence and broad-band excitation spectra in the visible spectral region. A search of the rather extensive literature on transition-metal-doped alkaline-earth fluorides⁵⁸⁻⁶² has led to the identification of several potentially viable systems. Among these, vanadium (V^{2+})-doped magnesium fluoride (MgF_2) is particularly interesting and has been carried through the preliminary stage of analysis, as summarized in this article.

Properties of the V:MgF_2 Laser Medium. Laser action at a wavelength of $1.12 \mu\text{m}$ in $\text{V}^{2+}:\text{MgF}_2$ was reported by Johnson et al.^{61,62} in 1966. The electronic structure of the V^{2+} active ion in the MgF_2

host medium is shown schematically in Fig. 7-57 together with absorption⁵⁸ and emission⁶² spectra. The single-coordinate potential energies of the electronic states of the V^{2+} ion, formed by the equivalent d^3 electrons of the ground configuration, are shown on the left. Fairly electronic electric-dipole transitions between these states are parity- (and in some cases also spin-) forbidden. However, weak phonon-assisted, electric-dipole transitions do occur between these states, as seen in the companion absorption and emission spectra. Peak absorption and emission transition cross sections lie in the range of 10^{-20} cm^2 . The $4T_2$ upper-laser level has a radiative lifetime of 2.3 ms⁶²; its potential energy minimum (zero-phonon state) is displaced significantly from that of the $4A_2$ ground state, resulting in a substantial Stokes shift in the fluorescence emission. The fluorescence emission terminates on a multiphonon level lying 1166 cm^{-1} above the $4A_2$ ground state potential minimum.

(This situation gives rise to the term “phonon-terminated” laser used to describe the V:MgF₂ laser.⁶³) The lifetime of the terminal phonon level is extremely short ($\ll 1$ ns), so that the system can be described as a four-level laser whose lower-laser level will *not* be “bottle-necked” for pulses of interest to us (< 1 ns). Because the gain profile is lifetime-broadened, and contains a high density of phonon states,⁶³ the system is expected to saturate essentially homogeneously with an estimated saturation fluence of $I'_{\text{sat}} = 22 \text{ J/cm}^2$.

The V:MgF₂ medium is particularly interesting because it appears that the V²⁺ metastable level can be excited with high efficiency using a conventional xenon flashlamp. The absorption spectrum of a V²⁺:MgF₂ crystal grown by Harshaw and the emission spectrum calculated for a pulsed-xenon flashlamp are shown in Fig. 7-58. The lamp spectrum was calculated⁶⁴ for a 1.5-cm-bore lamp with 200 Torr of xenon, radiating with a discharge current density of 1 kA/cm². Such conditions are consistent with long lamp life and high electrical-to-light conversion efficiency ($\sim 80\%$). The strongest and broadest V²⁺ absorption band, centered at 900 nm, arises from the $^4A_2 \rightarrow ^4T_2$ transition and is particularly well coupled to the intense line-emission of the xenon flashlamp. The VODEP code⁶⁵ has been used to estimate the fractional energy absorption in each of the V²⁺ absorption bands and the associated amount of lattice heating as the excited ions relax to the 4T_2 metastable level. This code generates the spectral radiation intensity emitted from the lamp and passes this radiation through a slab of V:MgF₂ material with a V²⁺ ion density of ρ , ion/cc and a physical thickness, l . The excitation efficiency of producing 4T_2 states is numerically calculated as a function of the optical-depth, ρl . The excitation efficiency is defined as the energy coupled into the 4T_2 state divided by the energy stored in the capacitor bank. Transport losses between flashlamp and laser medium are not included in this calculation.

The excitation efficiency of the 4T_2 metastable level of V²⁺:MgF₂ is shown in Fig. 7-59 for a 1.5-cm-bore xenon flashlamp with a discharge current of 1 kA/cm². The lamp spectrum is integrated from 0.25 to 1.1 μm and the resultant excitation efficiency is plotted as a function of the optical depth, ρl , of the V:MgF₂ medium. Note that the excitation efficiency varies rapidly for small values of optical depth, rising to an efficiency in the 20-to-30% range

Fig. 7-58. Absorbance spectrum of V²⁺: MgF₂ single crystal (Harshaw) and spectral emission intensity of 15-mm-bore xenon flashlamp with a discharge current density of 1 kA/cm². (Calculated using the GENEFF²⁷ xenon lamp model.)

Fig. 7-59. VODEP calculated excitation efficiency of V:MgF₂ as a function of V²⁺ ion density, ρ , and medium thickness, l . Pump source is a 15-mm-bore xenon flashlamp with a discharge current density of 1 kA/cm².

for ρl values in excess of 2.2×10^{20} ions/cm². Since physical slab thicknesses for multi-hertz, repetition-rated lasers will lie in the range of 2 to 4 cm, vanadium-ion-doping levels of 3 to 5×10^{20} ions/cc will be required for efficient laser pumping. VODEP calculations also show that the relative pumping efficiencies into the 250-to-450-nm, 450-to-700-nm, and 700-to-1100-nm pump bands are 21, 37, and 42%, respectively, and that the relative amounts of

Table 7-7. $V^{2+}:\text{MgF}_2$ spectroscopic²⁵ and optomechanical properties.

Spectroscopic		Optomechanical	
Pump cross-section, σ_p	1 E-20 cm ²	Density	3.18 g cm ⁻³
Upper level lifetime, $\tau(^4T_2)$	2.3 msec (77 °K)	Index of refraction	1.38
Lower level lifetime, $\tau(^4A_2)$	<1 nsec	Nonlinear index, n_2	3 E-14 esu
Laser cross-section, σ_l	8 E-21 cm ²	Thermal conductivity	0.3 W/cm ² K
Laser wavelength, λ_l	1.12 μm	Thermal expansion	11 E-6/°K
Saturation fluence	22 J cm ⁻²	Young's modulus	2.5 E7 psi
ESA cross-section, σ'	<3 E-21 cm ²	Fracture strength	7.6 E3 psi
		Damage threshold	>100 J/cm ²

lattice heating from each of the three pump bands are 37, 42, and 21%, respectively. The weak blue bands (250 to 400 nm) produce relatively little gain but disproportionally heat the lattice. Methods such as coating the xenon flashlamp with near-uv reflecting, visible-red-transmitting films^{66,67} to enhance long-wave emission, while preserving overall radiant efficiency, may prove quite useful in the $V:\text{MgF}_2$ laser.

Some of the important spectroscopic and optomechanical parameters for $V:\text{MgF}_2$ are summarized in Table 7-7. The laser spectroscopic parameters were determined by Johnson and Guggenheim⁶² from $V:\text{MgF}_2$ laser experiments performed at 77 K. The long radiative lifetime (2.3 ms), coupled with a large fluorescence emission linewidth (1500 cm⁻¹), results in a relatively low stimulated-emission cross section, $\sigma = 8 \times 10^{-21}$ cm². Given the nature of the gain profile, the gain is anticipated to saturate homogeneously with a saturation fluence of $\Gamma_{\text{sat}} = hc/\sigma = 22 \text{ J/cm}^2$ at low temperatures. The existence of an excited-state absorption process at the laser wavelength $\lambda_l = 1.12 \mu\text{m}$ has been characterized⁶² by a cross section 3×10^{-21} cm². The true cross section for this process is important in determining the limiting value of the specific thermal-loading factor. The MgF_2 host medium is characterized by an exceptionally low value of the nonlinear index of refraction,⁵⁵ $n_2 = 3 \times 10^{-14}$ esu. It has a moderately large fracture strength (7600 psi) and excellent resistance to optical damage. These properties are compatible with the relatively large saturation fluence (22 J/cm²) of the $V:\text{MgF}_2$ medium. Pure MgF_2 has a relatively large thermal conductivity at room temperature ($\kappa = 0.2 \text{ W/cm/K}$) which becomes much larger at reduced temperature⁶⁸ ($\kappa = 0.3$ at 200 K, and $\kappa = 3$ at 100 K).

$V:\text{MgF}_2$ Power Amplifiers. To obtain some feel for the overall efficiency performance which might be achieved with a $V:\text{MgF}_2$ power amplifier, a conceptual design of a $V:\text{MgF}_2$ amplifier analogous to the 34-cm-aperture Nova Nd:glass-box amplifier⁶⁹ was examined. Basically, this amplifier consists of two slabs of active material held at the Brewster angle with respect to the direction of propagation of the extraction pulse. The slabs are held in a rectangular box with a nominal 34-cm aperture and walls which are silver-coated metal. The two slabs are face-pumped from above and below using ten xenon flashlamps. The flashpumps are 110 cm long, with a 1.5-cm bore, and are excited for 500 μs . An examination was made of three $V:\text{MgF}_2$ amplifiers each of which differed in the optical depth of the active $V:\text{MgF}_2$ slabs. The important design parameters for each case are summarized in Table 7-8. The design with the highest value of ρl (1.6×10^{21} ions/cm²) is the most efficient. For this design, each

Table 7-8. Estimated parameters for a two-disk $V:\text{MgF}_2$ amplifier.

Parameter	Units	Doping (10^{20} ion/cm ³)		
		0.5	1.0	6.7
Disk thickness	cm	5.8	4.4	2.4
Disk minor dimension	cm	34.3	35.5	37
Disk major dimension	cm	61.5	63.7	66.4
Disk area	cm ²	2110	2260	2460
Disk volume	cm ³	12240	9945	5905
Optical depth	$10^{20}/\text{cm}^2$	2.9	4.4	16
Gain coeff	m ⁻¹	1.7	2.2	3
Stored energy density	J/cc	0.38	0.49	0.67
Energy stored per disk	J	4650	4875	3955
No. of lamps per amp		10	10	10
Input pump energy	kJ	97	97	45
Excitation efficiency	%	10	13	17

slab is 2.4 cm thick and stores about 4000 J of energy. The estimated small-signal gain coefficient for this case is 3 per meter and the excitation efficiency of the 4T_2 upper-laser level is 17% in a fully loaded cavity. The efficiency performance estimated for the 34-cm V:MgF₂ box amplifier is summarized in Table 7-9. The overall efficiency of producing the energy stored in the upper-laser level at the end of the 500- μ s pump pulse is broken down into six multiplicative sub-efficiencies:

- E_E – The bank-to-flashlamp transfer efficiency.
- E_R – The efficiency of converting flashlamp discharge energy into light.
- E_{TR} – The efficiency of transferring the flashlamp light to the V:MgF₂ medium.
- E_A – The fraction of energy incident on the V:MgF₂ medium that is absorbed.
- E_Q – The fraction of absorbed energy reaching the upper laser level.
- E_D – The fraction of energy having reached the upper laser level which does not decay away during the duration of pump excitation.

As the V²⁺ ion-doping density increases from 0.5×10^{20} ions/cc to 6.7×10^{20} ions/cc, the net production efficiency increases from 11 to 17%. Multipass extraction calculations indicate that extraction efficiencies of >60% can be achieved with B-integrals of <2 radians. A 10-ns-extraction pulse with a super-Gaussian top-hat spatial profile is used and thermally induced stress-birefringence losses must be kept adequately small. The maximum permissible pulse-repetition rate of a V:MgF₂ axial-gradient power amplifier, constrained by fracture stress of the amplifier medium, has been examined in a preliminary fashion using a simple model described elsewhere.³⁹ Parallel V:MgF₂ plates of thickness, t , pumped uniformly with a thermal heat

load of Q (J/cc), at pulse-repetition frequency, R (Hz), have been considered in these model calculations. The plates are separated by a 1-cm channel through which helium at 200 K flows at 0.2 Mach. The induced isothermal stress is plotted in Fig. 7-60 as a function of disk thickness, with the thermal power loading, QR (W/cc), as a parameter. For pumping conditions of interest, $Q > 0.7$ J/cc. Thus, plates of 2.5-cm thickness pulsed at a 3-Hz rate will experience a surface stress of 3000 psi (compare with a 7000 psi fracture stress for MgF₂). Thicker plates would have to work at correspondingly lower rates. One could maintain the same repetition rate and slab thicknesses by lowering the temperature of the cooling gas. For example, by reducing the helium temperature from 200 to 100 K, the thermal conductivity of MgF₂ increases by a factor of three. For the 100-K cooling temperature, the vertical scale of Fig. 7-60 is reduced by a factor of three. The power required to cool the plates is estimated to be less than 10% of the power removed from the plates.⁴⁶

Based on preliminary analyses and the conceptual laser systems outline given above, a number of key issues have emerged. Figure 7-59 indicates that to achieve an attractive excitation efficiency, the optical depth of the V:MgF₂ laser medium must be in

Fig. 7-60. Steady-state thermal stress in MgF₂ slabs under uniform thermal loading, Q (J/cc), at a pulse repetition rate, R (Hz), and cooled by flowing gaseous helium at 1 atm and 0.2 Mach in a 10-mm-wide channel.

Table 7-9. Projected excitation efficiencies of V²⁺MgF₂ 34 cm-box amplifiers.

Symbol	Sub-efficiency	Doping (10^{20} ions/cc)		
		0.5	1.0	6.7
E_E	Bank flashlamp	0.85	0.85	0.85
E_R	Rad. eff. lamps	0.80	0.80	0.80
E_{TR}	Cavity transfer eff.	0.65	0.71	0.75
E_A	Absorption eff.	0.46	0.52	0.64
E_Q	Quantum defect	0.67	0.67	0.67
E_D	Fluor. decay	0.78	0.79	0.80
E_{TOT}	Total eff.	0.11	0.13	0.17

the range of 3 to 5×10^{20} ions/cm². Heat removal considerations indicate V:MgF₂ slab thicknesses not greater than a few centimeters can be utilized so that the V²⁺ ion density in the slabs must then exceed 2×10^{20} ions/cc (i.e., 1 wt% VF₂). The highest V²⁺ ion density in MgF₂ reported to date in the literature⁵⁸ is 5×10^{19} ions/cc. Recently,⁷¹ researchers at the Lincoln Laboratory have developed⁵³ techniques for growth of high-optical-quality MgF₂ single crystals doped with the divalent transition metal ions Ni²⁺ and Co²⁺. Using V:MgF₂ melts in a vertical-gradient freeze configuration, Lincoln Laboratory has grown small crystals (2-cm-diam $\times$ 4-cm-long cylinders) with V²⁺ ion densities up to 2×10^{20} ion/cc (1 wt% VF₂). Preliminary growth runs revealed some gradients in V²⁺ distribution in the crystals, but at this time it is not thought to be a fundamental constraint. Preliminary discussions with Lincoln Laboratory and other crystal growers indicate good technical prospects for the growth of large (30 to 60 cm) V:MgF₂ crystals. One technique is the heat exchanger method (HEM) which has been used to grow⁷⁰ 300-mm-diam, high-optical-quality sapphire and silicon. Given continued successful progress in the V:MgF₂ growth area, additional key issues in laser design will be addressed in much more detail. V:MgF₂ materials and amplifier system design are two of the key issues which require further attention. With respect to V:MgF₂ materials, the following items are important:

- Measurement of stress-optical parameters.
- More refined measurements of mechanical properties (fracture stress, Poisson's ratio, etc.).
- Determination of excited-state spectral absorption cross sections.

Amplifier and system design characteristics of importance are

- Optimization of pump flashlamps.
- Efficient pump cavities.
- Parasitic-ASE control in V:MgF₂ slabs.
- Slab holding cryogenic cooling structures.
- Cyclical surface stress.

Recent experimental measurements performed at the Lincoln Laboratory⁷¹ show that the lifetime of the ⁴T₂ upper-laser level remains near the radiative lifetime of 2.5 ms for temperatures up to 200 K. A search of the literature has identified many cryogenic liquids matching the index of

refraction of MgF₂ (1.38) that can serve both to cool and to suppress parasitic oscillations in V:MgF₂ amplifier slabs. Preliminary V:MgF₂ systems analyses indicate that at the MJ level, overall system efficiency in excess of 5% should be realizable. Using moderately well-developed power-conditioning technologies (millisecond pulse-forming-network capacitor banks), high current ignitions, and low-stressed xenon flashlamps—alderated for $>10^9$ shot life—and assuming V:MgF₂ amplifier material costs at \$3/cc, systems costs of $<\$100$ M/MJ are projected. On the basis of a positive resolution to issues raised in our initial studies of the V:MgF₂ systems approach, we expect to address the key issues cited above during 1980. We will also continue to search for alternatives to V:MgF₂ with higher potential for development as an advanced laser driver.

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Group VI Lasers

During 1979 we completed our studies of Group VI atom lasers excited by photolysis of simple molecules. In this period we concentrated experimentally on Se(^1S) lasers produced by 172-nm photodissociation of COSe. The energy levels and laser transitions of selenium are shown in Fig. 7-61. We studied lasers operating on both the 489-nm, $^1\text{S}_0 - ^1\text{P}_1$ "transauroral line" and the 777-nm, $^1\text{S}_0 - ^1\text{D}_2$ "auroral line." The laser output energies that we obtained, as reported in the 1978 Annual Report,⁷² confirmed that the photodissociation yield of Se(^1S) atoms is at least 0.6 under practical laser conditions.

The kinetic performance of ^1S metastable atoms at the high concentrations ($\approx 10^{16} \text{ cm}^{-3}$) necessary for laser operation was discussed in the 1977 and 1978 Annual Reports.^{72,73} We found previously that the large fluences of VUV radiation, used to dissociate the fuel molecules, produced

Fig. 7-61. Energy levels of selenium. The laser transitions of interest are at 488.7 nm and 766.8 nm. Photoionization of Se(^1S) can occur by absorption of the 172-nm pump radiation.

Fig. 7-62. Cutaway view of the electron-beam source and high-pressure rare-gas cell used to produce rare-gas-excimer radiation.

significant concentrations of free electrons by photoionization of the product ^1S atoms.⁷⁴ This photoionization process is indicated schematically in Fig. 7-61. The free electrons can rapidly deactivate the ^1S atoms and thus greatly limit laser performance. We observed that small concentrations of SF_6 can counteract this problem by attaching the photoelectrons so that their quenching is not significant. For optimum Se-laser performance, we photolyzed 1 Torr of COSe mixed with 5 Torr of SF_6 and 50 Torr of CO. The CO provided deactivation of the lower state as well as electron cooling. The latter enhanced electron attachment by SF_6 .

In 1979 we investigated the effect of rare gas additives on Se-laser performance. We observed that collision broadening and collision-induced emission both effect the $\text{Se}(^1\text{S})$ gain. We also demonstrated operation of XeSe and KrSe collision-induced lasers for the first time. These devices are analogous to the rare-gas-oxide lasers that we have previously studied.⁷⁵⁻⁷⁷

Theoretical studies of issues important for Group VI lasers continued at a low level in 1979. We calculated the cross sections for inelastic collisions of electrons with sulfur atoms. These calculations, together with the previous work on oxygen

atoms,⁷⁸ add to our general knowledge of superelastic collisions. Such processes have great importance in gaseous-laser media.

Selenium Laser Experiments. A cutaway of the apparatus that we used for photolytic excitation is shown in Fig. 7-62. The VUV pump radiation, xenon excimer emission around 172 nm, was produced by an electron beam which excited pure rare gases. The major components of the electron-beam source (Maxwell Pocobeam) were a Marx bank, a 10- Ω coaxial water pulse-forming line, a self-breakdown output switch, and a field emission diode. The nominal voltage of the electron beam was 1 MeV for a duration of 50 ns. The anode of the diode was simply the high-pressure rare-gas cell as shown in Fig. 7-62. A two-bladed carbon cathode formed an electron-beam over an area of 500 mm $\times$ 75 mm. The cathode was directly opposite a ribbed structure which supported a 0.127-mm-thick titanium foil through which an electron-beam current of approximately 50 kA entered the gas.

The excited rare gas was contained in a 500-mm-long $\times$ 150-mm-high $\times$ 30-mm-deep volume. The energy deposition profiles and energy transfer efficiencies for this device were given previously.⁷⁷ Because the energy deposition was largest near the foil, we designed the photolytic cell so that the entrance was aligned as close as possible to that surface. The parallel offset between the leading edge of the entrance window and the foil was about 4 mm. The longitudinal distance from the end of the electron-beam-pumped region to the photolytic cell window is about 170 mm.

A cutaway of the photolytic cell is shown in Fig. 7-63. Laser mirrors were held internal to the cell and separated by 120 mm. A metal trough limited the photolysis volume of the laser to about 6.5 cm³. We measured the laser output power using vacuum photodiodes and compared this power to the laser output energy measured by a volume-absorbing calorimeter.

Identical photolytic cells were placed at each end of the rare-gas excimer source. We used one cell for the laser and kinetics studies and the other to house a calorimeter which gave the VUV pump energy on every shot. The time dependence of the VUV light was measured with a photodiode which detected light scattered from the calorimeter. The calorimeter consisted of a 1-mm-thick $\times$ 85 mm $\times$ 15 mm carbon collector which had a single thermocouple attached to its back side. The calorimeter

Fig. 7-63. Cutaway view of the photolytic cell used for experiments with Se lasers. The laser mirrors are mounted within the cell.

element was made of pyrolytic graphite to be damage-resistant and relatively black to the VUV radiation. The carbon collector, held in a vacuum, was suspended on threads directly behind the MgF_2 entrance window. We calibrated the calorimeter response by heating the carbon with a 0.2 s pulse of 488-nm, Ar^+ laser radiation. The calorimeter was illuminated by an expanding Ar^+ beam whose spatial profile matched the dimensions of the VUV window aperture. We used the volume-absorbing laser calorimeter to measure the energy of the Ar^+ laser pulse before expansion.

The calorimeter response was checked against a pressure-rise technique designed to measure the VUV energy. This technique utilizes the change in the number of molecules when N_2O is photodissociated. Photodissociation of N_2O around 172 nm produces a nitrogen molecule and an oxygen atom. The oxygen atom can react with N_2O to

form either N_2 plus O_2 or two NO molecules. In either case the net result is a gain of exactly one molecule per absorbed photon.⁷⁹ Knowing the cell volume, we used the chemical pressure-rise to determine the VUV energy entering the photolytic cell. Using N_2O at one atmosphere pressure, where its absorption coefficient around 172 nm is about 2.5 cm^{-1} , we observed pressure rises on the order of 0.1 Torr with a differential capacitance manometer. The VUV energies, indicated by the pressure-rise and the carbon calorimeter, agreed within 10% over the range of energies used in the experiments.

We varied the output fluence of Xe_2 radiation by changing the Xe pressure in the main cell, thereby varying the electron-beam power deposition. For pressures below 7 atm, the Xe_2 output from the ends of the cell was simple fluorescence. For higher pressures the output could be called superfluorescence: that is, it exhibited the effects of medium gain. Superfluorescent operation was indicated by a gain-narrowing of the Xe_2 spectrum and a rapid change in the VUV output energy with Xe pressure. The maximum VUV energy transmitted into the photolytic cell was about 2 J at 13 atm of Xe.

We optimized the laser performance on both the $\text{Se } ({}^1\text{S}_0 - {}^3\text{P}_1)$ and $\text{Se } ({}^1\text{S}_0 - {}^1\text{D}_2)$ lines by varying the photolytic gas mixture, the VUV pump fluence, and the output coupling of the laser mirrors. As previously noted, the optimum gas mixture was a 1-5-50 Torr mix of COSe , SF_6 , and CO . The laser output was approximately constant for VUV fluences between 125 and 200 mJ/cm^2 . The COSe , which has a saturation fluence for dissociation of about 14 mJ/cm^2 , was completely bleached over the laser aperture for these fluences. (We measured the COSe absorption cross section for Xe_2 superfluorescence to be $8.4 \pm 1.0 \times 10^{-17} \text{ cm}^2$ implying a saturation fluence, $h\nu/\sigma$, of $14 \pm 2 \text{ mJ}/\text{cm}^2$.) Above 200 mJ/cm^2 the laser output slowly declined, presumably because of kinetic losses associated with photoionization. Laser mirrors having about 1% transmission gave optimum output energy. Mirror reflectivity variations indicated that the single-pass gain was about 5% over 80 mm for the transauroral line and slightly less for the auroral line. For optimum conditions the output energy was 25 mJ for

Fig. 7-64. Time dependence of the Se transauroral laser for (a) high-intensity and (b) low-intensity outputs. The end of the photolytic pulse is indicated by the arrows.

the blue line and 16 mJ for the infrared line. The time dependence of the transauroral laser is shown in Fig. 7-64(a).

Figure 7-64(b) illustrates the effect on the laser performance of adding rare-gas diluent to the photolytic mixture. With the addition of 300 Torr of Xe, the peak transauroral power was lowered from about 30 kW to 1 W. Also, the delay and pulse width increased with the addition of Xe. These effects were caused simply by lowering the gain of the laser transition by line broadening. When the gain was sufficiently small, such as in Fig. 7-64(b), the laser was strictly in a build-up mode and extracted an insignificant amount of energy from the medium. A simple rate equation formulation describes the laser intensity in that case.

Omitting the spontaneous source term, the laser intensity, I_L , is described by

$$\frac{dI_L}{dt} = \sigma_s S I_L - \tau_c^{-1} I_L \quad (42)$$

where the cavity loss rate, τ_c^{-1} , is the round-trip cavity loss divided by the round-trip transit time. The stimulated emission gain cross section is σ_s . For non-saturating conditions, the Se(¹S) population, S , decays only by collisional processes. The peak laser intensity occurs at a time, t_p defined by the condition $\sigma_s S(t_p) = \tau_c^{-1}$. Expanding the Se(¹S) time dependence in a Taylor series about t_p and using this relationship, we find to lowest order

$$\frac{dI_L}{dt} = -\tau_c^{-1} (t - t_p) I_L \quad (43)$$

where $\tau_c^{-1} = \dot{S}(t_p)/S(t_p)$ is the instantaneous decay rate of the Se(¹S) density. The time dependence of the laser intensity around t_p is given by

$$I_L(t) = I_L(t_p) \exp[-1/2 \tau_c^{-1} (t - t_p)^2]. \quad (44)$$

Fig. 7-65. Delay time from the end of the photolytic pulse to the peak of the laser pulse as a function of Xe and He additive pressure for both the transauroral and auroral lines. The variations are caused by both line broadening and collision-induced emission.

Therefore, for non-saturating conditions the laser pulse should be Gaussian-shaped with a FWHM of $2.355 \tau_c^{-1} \tau_c$. A Gaussian pulse shape is a good fit to the observed pulses, as shown in Fig. 7-64(b). Knowing the Se(¹S) decay time, τ_s , we have also used the FWHM of such pulses to determine the cavity decay time and, hence, the total mirror loss for practical conditions.

Figure 7-65 shows the time delay to the maximum laser intensity for both the auroral and transauroral Se lasers as a function of either He or Xe addition. Aside from the rare-gas addition, we used the optimum mixture, VUV fluence, and mirror reflectivities as previously discussed. Note for the $1S_0 - 3P_1$ laser that Xe causes a greater delay than He for a given pressure. This indicates a more rapid gain reduction, presumably from the line broadening caused by the addition of Xe rather than He. As the laser delays approached their peak values with increasing rare-gas addition, the laser power began a rapid decline which covered many orders of magnitude. We interpret this to mean that at the maximum delay the laser undergoes a transition from the saturating to the build-up regime. With further rare-gas addition, and hence lower overall gain, the delay then decreases because the crossover with the cavity loss occurs earlier in time. In contrast to the transauroral behavior, the laser delays on the $1S_0 - 1D_2$ line were essentially identical for Xe and He additions. For this case we believe that the reduction in gain caused from line broadening by Xe is opposed by an increase in gain caused by

Fig. 7-66. Peak intensity of the Se auroral-line laser versus Xe pressure added to the normal photolytic mixtures. The increased intensity for Xe pressures above 1000 Torr indicates operation of a collision-induced XeSe laser.

collision-induced emission. Collision-induced effects were not significant for the transauroral line.

We investigated the effect of collision-induced emission on the Se (¹S₀ - ¹D₂) laser by adding large concentrations of Xe to the gas mixture and using the conditions that were otherwise optimum. Figure 7-66 shows the peak laser intensity measured by the photodiode as a function of Xe addition. For Xe pressures above 1000 Torr, the laser output increased rather than decreased with Xe addition. A laser mixture composed with 5000 Torr of Xe, and allowed to mix for four days, produced an output energy approximately 75% of that obtained for low-pressure mixtures. Assuming that above 1000 Torr of Xe, the gain increased rather than decreased with Xe pressure, the variation of the pulse shape and delay with added Xe were analogous to the low-pressure behavior. As judged by the time delay to the peak intensity, the gain in the high-pressure regime equaled the Doppler-broadened gain at approximately 3500 Torr of Xe.

In the region where the gain increases with Xe pressure because of collision-induced emission, the lasing species should be labeled XeSe. The collision-induced laser spectrum was distinctly different from the atomic spectrum, supporting this designation. The peak emission was at 777.26 ± 0.1 nm, to the red of the atomic line, and had a resolvable width of about 0.2 nm, as shown in Fig. 7-67. We also ob-

Fig. 7-67. Spectra of Se (¹S₀ - ¹D₂) atomic laser (unresolved) and the XeSe collision-induced laser.

tained infrared oscillation at 4000 Torr of Kr, implying operation of a KrSe laser. The slight shift in the XeSe laser wavelength from the atomic line is similar to that previously observed for KrO laser operation.⁸⁰ We speculate that a red-shift in the peak gain may be caused by the contribution of the ¹Σ⁺ - ¹π molecular transition, whereas the broader ¹Σ⁺ - ¹Σ⁺ transition is generally blue-shifted from the atomic line.^{81,82}

Electron-Sulfur Inelastic Collisions. Previous kinetic studies have established that the deexcitation of S(¹S) and Se(¹S) by electrons is a serious loss mechanism unless molecular buffers are employed to reduce the density of electrons in the photolytic mixture. In our original analysis,⁷⁴ we estimated the cross sections for superelastic electron-sulfur and selenium collisions by extrapolating the theoretical cross sections available for the analogous electron-oxygen system.⁷⁸ More recently, we have undertaken a new theoretical study of electron-sulfur collisions with the goal of obtaining more accurate deexcitation cross sections for S(¹S). We used the close-coupling technique to describe the dynamics of the scattered electron and multiconfiguration wave functions to represent the (3s² 3p⁴) ³P, ¹D and ¹S states. The resulting superelastic cross sections for ¹S → ¹D and ¹S → ³P transitions are shown in

Fig. 7-68. Theoretical cross sections for the deexcitation of $S(S)$ by low energy electrons.

Fig. 7-68 for impact energies of 0 to 15 eV. The broad dips in the cross sections near 3 to 4 eV are due to the $3s3p^6\ ^2S$ resonance state of S^- . Since the description of this resonance in the present work is not very accurate, we expect the position and width of this feature to change somewhat at higher levels of approximation. At low energies, the present cross section for the dominant $1S \rightarrow 1D$ transition is about a factor of 4 higher than our previous estimate and yields a thermal rate coefficient of $4 \times 10^{-8} \text{ cm}^3 \text{ s}^{-1}$ for the electron deactivation of $S(S)$.

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Increased Bandwidth for Fusion Lasers

Broadband irradiation of fusion targets could suppress stimulated Brillouin effects and permit more efficient coupling to targets at higher intensities than with single frequency lasers. There is increasing interest in converting narrowband lasers such as Nd:glass and KrF to broadband sources and we have recently begun to address this issue seriously.

We feel that one of the best candidates for broadband conversion of fusion lasers is forward-stimulated-rotational Raman scattering ($\Delta J = 2$) in hydrogen or deuterium. We have begun an investigation of its characteristics. Stimulated-rotational Raman scattering converts a single-laser frequency input to a multiline spectrum centered on the original laser frequency with an interline spacing of 587 cm^{-1} for hydrogen and 297 cm^{-1} or 414 cm^{-1} for deuterium. If the target Brillouin gain bandwidth is less than the interline spacing, these multiline sources are equivalent to broadband irradiation. In 1979 we concentrated on concepts and theory and will proceed to experiments on small Nd:glass and KrF lasers in 1980. This includes possible installation of a broadbanding system on the Argus laser for target experiments if the results of initial experiments are promising.

A broadband rotational Raman converter would be applied to the output of a fusion laser as shown in Fig. 7-69. A Raman laser injects a forward-traveling pulse coincident with a single-frequency laser-pump pulse. The two pulses propagate together through a converter cell filled with hydrogen or deuterium. Parametric coupling between the two pulses is strong because of the small frequency shift and the very low dispersion of hydrogen. Parametric interactions quickly generate many orders of stimulated Stokes and anti-Stokes Raman scattering to the red and blue sides of the pump pulse, propagating collinearly with it. The process is very efficient since the rotational shift is small. The rotational gain is highest for pump and

Fig. 7-69. Multiple wavelength generation by rotational Raman scattering. A Raman laser injects a forward-traveling pulse coincident with a single-frequency laser pump-pulse. The two pulses propagate together through a converter cell filled with hydrogen or deuterium.

Table 7-10. Predicted gain in cm/GW for hydrogen vibrational and rotational Raman scattering.

Parameter	λ (nm)		
	249	694	1064
Rotation: linear polarization	3.5	1.0	0.6
Rotation: circular polarization	5.3	1.4	0.9
Vibration: either polarization	7	1.4(1.5) ^a	0.7

^aMeasured value (Ref. 88).

first Stokes injected signals with opposite circular polarizations. Parallel linear polarization can be used but with only 2/3 the gain for opposite circularly polarized beams.

We estimate the gain for stimulated-rotational Raman scattering in hydrogen using the following:

- The Placzek relation for wavelength scaling of Raman cross sections.⁸³
- Literature values for broadening coefficients and pressure narrowing from Doppler collapse.^{84,85}

- Experimental comparison of the relative gains of rotational and vibrational scattering in hydrogen.⁸⁶⁻⁸⁸ The predicted vibrational and rotational gains in hydrogen are independent of pressure when it is high enough that the Doppler effect has been completely suppressed and the line width is determined solely by pressure broadening. These predicted gains are shown in Table 7-10. A converter for the 249-nm KrF laser may require careful design to avoid competition from super-

Fig. 7-70. H₂ Raman gain vs density. At low pressures, both rotational, S, and vibrational, Q, gains are a function of pressure. Vibrational gain falls off dramatically at low pressures where Doppler broadening is important.

fluorescent vibrational Raman scattering. This is not likely to be an issue for 1064-nm Nd:glass applications. At low pressures, the gains are a function of pressure, as shown in Fig. 7-70. The vibrational gain falls off dramatically at low pressures, where Doppler broadening is important, since the vibrational shift (4155 cm^{-1}) is much larger than the

rotational shift. There is less experimental data available for deuterium but it is expected to show similar results.

For pulses less than a few nanoseconds the line width of the Raman amplifier is sufficiently narrow that the Raman amplifier can become transient, thus reducing the gain. This occurs when the response time of the medium is not short compared to the pulse lengths of the pump and Stokes pulses. Standard analysis of transient parametric amplifiers indicate that the transiency condition is essentially

$$\Delta t \tau \approx \sqrt{G/2\pi}$$

where G is the small-signal, long-pulse-width, gain exponent and τ is the pulse width. However, in the multiline case appropriate to SRRS in H_2 the appropriate condition for transiency is $\tau/T_2 \gtrsim 1$. Thus, for H_2 at 1 atm, the device will be transient for 1-ns pulses since $T_2 \sim 7.2$ ns for this case. Transient effects could influence the choice of operating pressure, as shown in Fig. 7-71, forcing a ns-pulse converter to operate above 3 Amagats. Converters for use with large lasers and longer pulses could operate at atmospheric pressure, or below.

The theory of the generation of many high-order sidebands, through parametric coupling in forward Raman scattering, is complex. We shall summarize briefly some conclusions here.

A set of parametrically coupled Raman orders evolves according to the equation

$$\frac{\delta E_j}{\delta z} = \frac{\nu_j}{\nu_0} (\beta^* E_{j+1} - \beta E_{j-1}) \quad (45)$$

where $\beta = 1/2Qe^{i\phi}$ is a coupling parameter ("material excitation") proportional to the product of all adjacent field amplitudes $\sum_j E_j^* E_{j-1}$. On further approximation, Q is roughly the Stokes gain per unit length, γl_0 . The subscripts, j , here refer to the pump for $j = 0$, to anti-Stokes for $j > 0$, and the Stokes for $j < 0$. If the Raman shift is very small so that $\nu_j \approx \nu_0$ and the dispersion of the index of refraction is zero, one may show that

$$E_n(z, \zeta) = \sum_p e^{i(n-p)\phi} F_p^{\text{in}}(\zeta) J_{p-n}(zQ) \quad (46)$$

where z is the distance propagated through the converter and ζ is the running time, $t - z/c$. J_n is a Bessel function of order n , and

Fig. 7-71. H_2 Raman line width vs density. Transient effects are a factor in choice of operating pressure. Nanosecond-pulse converters must operate above 3 Amagats, whereas, converters with large lasers and longer pulses can operate at, or below, 1 atm.

$$Q = \frac{1}{2} \epsilon_0 C \gamma \int_{-\infty}^{\infty} dt' / T_2 I_p^{\text{in}}(t') F_s^{\text{in}}(t') \times \exp - [(t - t')/T_2] \quad (47)$$

where γ is the monochromatic forward steady-state gain coefficient, approximately 10^{-9} cm/W for H_2 at 1.064 nm. The number of waves in the output is approximately $2zQ$ and the conversion efficiency to a higher order wave becomes

$$I_n(t, z) = I_0(t) J_n^2(zQ) \quad (48)$$

where I_0 is the pump intensity and I_n is the intensity in order, n . For a representative 1064-nm experiment using a pump at 10^8 W/cm² and a 1-m converter cell, we predict an output consisting of about nine waves. These higher-order waves, of course, violate the $\nu_j \approx \nu_0$ assumption for 1064-nm experiments, so the applicability of this analysis is uncertain until experiments confirm it. However, the prediction is not in conflict with typical Raman oscillator experiments in the literature.

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Chemically Pumped Iodine Lasers

The ($^2P_{1/2} - ^2P_{3/2}$) transition in atomic iodine at $1.315 \mu\text{m}$ has been considered in the past as a possible laser fusion driver.⁸⁹ This laser is excited by flashlamps, has a reasonable storage lifetime, and can be cooled by convective transport. The primary disadvantage for fusion-reactor applications has been the relatively low efficiency. Recently, a chemical process which leads to the formation of excited iodine has been discovered.⁹⁰ This type of laser operates satisfactorily in a cw mode but may not operate in the pulsed mode required for fusion applications. As part of the evaluation of this laser system, we are considering alternatives to the present techniques of chemical production that are more suitable for pulsed operation.

The chemically pumped iodine laser derives its energy from the chemical reaction of O_2 (gas) which is bubbled through a basic (NaOH) solution of hydrogen peroxide (H_2O_2) to form $\text{O}_2(^1\Delta)$. The excited oxygen molecules are mixed with I_2 and the following reaction sequence results in a population inversion in the I-atoms:

If the fraction of system oxygen molecules in the excited $^1\Delta$ -state exceeds 15% (20% at 500 K), the equilibrium population of I-atoms, due to reaction (3), will be inverted. In practice, the $\text{O}_2(^1\Delta)$ concentration is 40%, although theoretically the $\text{Cl}_2 - \text{H}_2\text{O}_2$ reaction could yield 80%.

Higher $\text{O}_2(^1\Delta)$ concentrations have been achieved by using aromatic molecules, which absorb ultraviolet radiation over a broad band of energies and then transfer the excitation to O_2 .

Wasserman et al.⁹¹ have observed that in a vapor-phase mixture of octafluoronaphthalene (C_{10}F_8) and O_2 , irradiated with a mercury or xenon flashlamp, the steady state fraction of O_2 in the $^1\Delta$ state was as high as 70%. The C_{10}F_8 , whose ground state is a singlet, absorbs photons with energies $>30\,000 \text{ cm}^{-1}$ to populate several excited singlet states. Such states rapidly decay (through inter-system crossing) to very long-lived triplet metastable states. These can transfer their energy to the ground $^3\Sigma$ -state of O_2 to produce $\text{O}_2(^1\Delta)$. The steady-state population of excited O_2 molecules has been measured for many different aromatic sensitizers. Several produced $^1\Delta$ fractions in excess of 40%; results with octafluoronaphthalene were by far the best with 70%.

It is expected that mixtures at about one Torr each of O_2 , I_2 , and C_{10}F_8 in several atmospheres of an Ar-gas buffer, when irradiated with a flashlamp, could produce significant populations of $\text{I}(^2P_{1/2})$. This system has two advantages over the $\text{Cl}_2 - \text{H}_2\text{O}_2$ system. First, since only the I_2 molecules are dissociated in this process, a regeneration step is unnecessary and the problems associated with obtaining a homogeneous mixture of I_2 and $\text{O}_2(^1\Delta)$ in a flowing system can be avoided. Second, the C_{10}F_8 will serve as a reservoir for energy used during the extraction step to dissociate any I_2 that is re-formed. The most effective quencher of $\text{I}(^2P_{1/2})$ in this system is I_2 . Maintaining its concentration at a low level can increase the storage time of the energy in the laser.

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Laser Theory and Design

In this section, we present the results of analytical and experimental programs that have addressed specific technical issues associated with rare-gas-halide pulse-compressor laser systems. These programs include kinetics experiments and

Fig. 7-72. Schematic of the LAMP kinetics facility.

modeling, theoretical studies of specific reaction processes, and computational simulations of parasitics and transient laser dynamics.

LAMP KrF Kinetics Experiments and Analyses

Because the KrF laser is the most efficient high-power short-wavelength laser available, there has been much effort devoted to understanding the formation and quenching kinetics of this excimer. The kinetic scheme in an electron-beam-pumped KrF laser is sketched in "KrF Amplifiers," above. In an effort to assemble a complete model for Ar/Kr/F₂ kinetics, we have performed experiments on Ar/F₂, Kr/F₂, and Ar/Kr/F₂ mixtures using a short-pulse low-current electron beam. The experimental fluorescence data were analyzed using a computational kinetic model, and a self-consistent set of kinetic data was constructed. Calculations

were then performed to model experiments appearing in the published literature, and the model was found to give good agreement with the published results. With such a kinetic model, we may explore the parametric space of operation for a KrF laser for gain, loss, and efficiency in terms of Ar/Kr/F₂ fractions, total pressure, gas temperature, and electron-beam power deposition and pulse length.

Experiments. Mixtures of Ar, Kr, and F₂ at total pressures of 250 to 2000 Torr were excited by a short-duration electron beam, and their fluorescent spectra were studied with temporal and spectral resolution. A block diagram of the apparatus is shown in Fig. 7-72. The electron gun has 80 keV of energy and provides an electron pulse of 24-ns duration, with rise and fall times of 1 ns. The current density at the foil is approximately 0.3 A/cm^2 . The pulse repetition rate is variable up to 50 pulses per second (pps) but was run typically at 10 pps. The electron beam enters the cell through a narrow slit, $5 \times 30 \text{ mm}$, covered with a thick foil. The fluorescent signal is viewed through LiF lenses, using a

McPherson 300-mm monochromator, and the signal is detected with an RCA photomultiplier (GaAs photosurface), recorded on a Tektronix 7912 transient recorder, and averaged on a PDP 11 digital computer.

Experimentally, it was found that the total time resolution of the system was better than 5 ns. The signal was averaged between 100 and 400 times, depending on the signal strength, and the computer calculated the logarithm of the signal, displayed it, and fitted a straight line (weighted by the signal strength) in an interval selected by the operator. In this way, the decay times were calculated. The integrated signal was obtained electronically with a resistance-capacitance (RC) integrator and averaged by photographing the face of an oscilloscope. It should be pointed out that the computer models were fitted to the whole fluorescence curve, not merely to its decay. In particular, the quantities used in the fitting were integrated intensity, peak intensity, delay of the peak intensity, decay rates, and deviations from exponential decay.

In our experiments, we used binary mixtures of Ar/F₂ and Kr/F₂ and ternary mixtures of Ar/Kr/F₂. In the binary mixtures, total pressures of 250, 500, 1000, and 1950 Torr and F₂ concentrations of 0.035, 0.1, and 0.3% were systematically explored. In the ternary mixtures, several "cuts" were made with concentrations of Kr and F₂ in the ranges of 0.3 to 3% and 0.135 to 0.3%, respectively, but not all combinations were measured.

Emissions were measured on the peaks of ArF⁺ (B-X) at 195 nm (the apparent red shift from 193 nm was caused by the falling sensitivity of the photomultiplier) and on the Ar₂F band at 295 nm. In KrF, all three bands were monitored: the D-X at 221 nm, the B-X at 249 nm, and the C-A at 280 nm. Also monitored were the Kr₂F emission at 380 nm and the Kr(4p'[1/2]₁ → 4s'[1/2]₂) J = 0 line at 772.4 nm. In ternary mixtures, there is an overlap between the Ar₂F and KrF (C-A) emissions, and they cannot always be distinguished.

Sample spectra are shown in Fig. 7-73 for a mixture of Ar at 1000 Torr and 0.1% F₂, together with the results of our computer modeling. The spectra in these cases were averaged 100 times. The time dependence of these spectra is typical: the ArF emission "integrates" the electron-beam pulse, immediately starts to decay in an exponential way, and later decays in a way that is slower than exponential

Fig. 7-73. Typical experimental and computed fluorescence profiles for ArF and Ar₂F.

(more like $1/t$). The Ar₂F emission peaks with a delay and becomes very nearly exponential at later times.

Model. The kinetic model used in this analysis includes electron-beam excitation and ionization processes, electron-impact processes due to thermal electrons, heavy-particle kinetics, and photon processes. A Maxwellian electron-energy distribution is assumed, and an energy balance is used to compute the gas and electron temperatures. The computer program reads in symbolic chemical reactions, rate-coefficient data, and electron-impact data; constructs rate equations for the chemical species; and solves for the time-dependent populations. By extensive use of interactive computer graphics, the program can quickly determine decay rates and integrated intensities for comparison with the experimental data.

Our treatment of electron impact processes has been described in a previous annual report.⁹² The basic formation and quenching kinetics of ArF and KrF in electron-beam-pumped Ar/Kr/F₂ mixtures are well understood and have been described in

detail by Rokni et al.⁹³ The basic kinetic scheme is described in "KrF Amplifiers," above. In our electron-beam experiment, operated at gas pressures of greater than 500 Torr, the primary rare-gas halide (RGH) formation process is positive-negative ionic recombination. A general description of the process is presented in "Ion-Ion Recombination," below. As there are no experimental data on these recombination rates, we use the theory of Flannery^{94,95} with data on ionic mobilities to calculate the ion-ion recombination rates. Since the effective two-body rate is a function of pressure and temperature, this calculation is performed during operation of the kinetics code.

Results of Experiments and Modeling. The experiments were performed systematically on Ar F₂, Kr F₂, and Ar Kr F₂ mixtures, and decay rates, time-integrated intensities, and delay times of the diatomic and triatomic RGH exciplexes were measured. Values were established for key rate coefficients in the model by varying tentative coefficient values systematically until a good match was obtained between calculational and experimental values.

In the Ar F₂ mixture, even at 500 Torr, the kinetic chain for formation of ArF consists of rapid formation of the dimer ion ArF₂⁺ from Ar⁺ and subsequent recombination with F⁻ to form ArF. Due to the very short radiative lifetime of ArF (4 ns) (Ref. 96), its apparent decay rate in the afterglow is a measure of its formation rate. Flannery and Yang find that the effective two-body rate for ArF₂⁺ + F⁻ + Ar → ArF + Ar increases linearly with pressure at pressures of less than 0.5 atm, becoming diffusion limited at higher pressures with a peak of $k = 3.1 \times 10^{-7} \text{ cm}^3 \text{ s}^{-1}$ at $p = 1.6 \text{ atm}$. In the model calculations, shown in Fig. 7-74, we found that a recombination rate coefficient equal to one-third the Flannery and Yang⁹⁵ value gives the best results. We have used a constant scale factor of 1/3 for all pressures, although, in reality, the recombination rate coefficient probably is a function of pressure.

Due to the low beam-current density, the electron density in the plasma is in the range of 10^{12} to 10^{13} electrons/cm³ and, of course, diminishes very rapidly in the afterglow as a result of F₂ dissociative attachment. Because of this rapid diminution, electron quenching of the excimer is negligible, and dis-

Fig. 7-74. ArF decay rates in the electron-beam afterglow vs F₂ content for three total gas pressures.

sociative recombination of the dimer ion is not competitive with ion-ion recombination, especially in the afterglow. Additionally, due to weak excitation, the myriad other excited state-excited state processes may be neglected.

The integrated ArF intensity, as a function of pressure, is sensitive to the rate of

which is the primary Ar₂F formation process. The rate coefficient for this reaction was found in this work to be $2 \times 10^{-31} \text{ cm}^6/\text{s}$. Because the stable configuration of the Ar₂F molecule is triangular, the probability that this molecule is formed directly by ion recombination is small.

The primary quenching of Ar₂F in these experiments is due to F₂ because Ar₂F has a long radiative lifetime (185 ns). Although the experimental data give an apparent rate of $2 \times 10^{-10} \text{ cm}^3/\text{s}$ for F₂ quenching, the model calculations use a value of $4 \times 10^{-10} \text{ cm}^3/\text{s}$ for a good fit. Due to the rapid formation rate of Ar₂F, the apparent and actual rates differ, as does the extrapolated zero pressure rate. Comparisons of experimental data and model calculations are shown in Figs. 7-73 and 7-75. In Fig. 7-73, the experimental and computed curves are normalized in peak amplitude only, and it may be seen that the predicted delay time to peak fluorescence is in excellent agreement with the experiment.

Fig. 7-75. Ar_2F decay rates in the electron-beam afterglow vs total gas pressure for three F_2 contents.

As we might expect, the kinetics of Kr/F_2 mixtures are nearly identical to those of Ar/F_2 mixtures. Again, we found the ionic recombination rates to be slower than the theory predicts, about one-fourth the Flannery and Yang⁹⁵ value. The kinetics of the $\text{Ar}/\text{Kr}/\text{F}_2$ mixture are more complicated. For high Kr fractions (3%), KrF is formed by Kr_2^+ ionic recombination, with a rate equal to 0.7 times the theoretical rate.⁹⁵ At very low Kr fractions (0.3%), KrF is formed primarily by ArKr^+ ionic recombination. The rate here is one-half that predicted by using the Flannery⁹⁴ theory.

Both Ar and Ar^+ have roles in the kinetic chain that leads, ultimately, to the formation of KrF . In considering these roles, we direct our attention to the Stern-Volmer plot in Fig. 7-76. From the dashed line, we can infer a rate coefficient of $3.3 \times 10^{-10} \text{ cm}^3/\text{s}$ for the displacement reaction

The deviation at low partial pressure of Kr is due to the interception of the Ar_2^+/F^- recombination channel by charge transfer from Ar_2^+ to Kr . Because of this, we observe a simple exponential decay of ArF fluorescence in the experiment. Our analysis obtained a rate coefficient for the displacement reaction of $7.5 \times 10^{-10} \text{ cm}^3/\text{s}$, which agrees with other experimental values.⁹⁷ Consequently, most ArF at

Fig. 7-76. Stern-Volmer plot of reciprocal of ArF integrated intensity in $\text{Ar}/\text{Kr}/\text{F}_2$ mixture.

high Kr partial pressures is formed by the "harpoon" reaction

A rate coefficient of $4 \times 10^{-10} \text{ cm}^3/\text{s}$ was obtained from our analysis of the harpoon reaction, and this is in agreement with the most recent measurement of Kolts and Setser.⁹⁸

The complete set of rate coefficients deduced from these experiments and analyses has been presented in "KrF Amplifiers," above. These results were used to prepare model predictions that, when compared with experimental data obtained both from the literature and from our own experiments, have shown good agreement between predicted and actual results. This agreement gives us reason to have confidence in the predictive capabilities of this $\text{Ar}-\text{Kr}-\text{F}_2$ kinetic model.

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Rare-Gas Halide Kinetics

We have performed small-scale experiments on photolytic and electron-beam-pumped rare-gas halide lasers to

- Improve our general understanding of the rare-gas halide excimers.
- Study possible means for increasing the efficiency of our baseline system, the KrF laser.
- Investigate electron-beam-pumped XeCl lasers as possible replacements for KrF devices.

The XeCl system has the advantage of operating at longer wavelengths than KrF, 308 nm compared with 249 nm, where thin-film dielectric coatings are considerably more resistant to optical damage. However, past experiments⁹⁹ in the electron-beam-pumped mode have shown that the XeCl medium has an efficiency that is approximately one-half the efficiency of KrF. The best XeCl efficiency was found at an operating pressure of 4 atm, which is considerably higher than we normally use for KrF.

In 1980, we are continuing to explore the parameters of the XeCl system, and we are investigating three fundamental issues of rare-gas halide lasers:

- The partition of energy between the B and C electronic states.
- Electron deactivation of the excited states.
- Background absorption in the excited medium that limits the output power and efficiency of the laser.

Because these three issues are common to, and closely related for, all rare-gas halide excimers, we have not limited our attention to the primary candidate molecules, KrF and XeCl. For example, the photolytically pumped XeF laser has provided information on all three issues.

XeF Experiments. We used the MEG II facility, described in "Group VI Lasers," above, to produce XeF by the photodissociation of XeF₂. Our experiments differ from the Group VI laser work primarily by the use of XeF₂ rather than COSe (carbonyl selenide) as a photolytic fuel. XeF₂ is a

Fig. 7-77. Schematic potential-energy curves for XeF.

transparent crystal having a vapor pressure of 4 Torr at room temperature. It is readily available commercially and is stable with respect to Xe plus F₂ by 1.23 eV (ΔH_f°).¹⁰⁰ In a system with nonreactive walls, the lifetime of XeF₂ should be infinite. However, by using a mass spectrometer, we found that the one-half decay time of XeF₂ in our photolytic cell was about 30 min. To study questions regarding XeF₂ loss, we performed photolytic experiments by slowly flowing our buffer gases over a temperature-controlled bed of XeF₂ crystals.

XeF₂ absorbs in a strong band centered at 160 nm. We found the XeF₂ absorption cross section for Xe₂ superfluorescence at 172 nm to be 8.5×10^{-18} cm², corresponding to a saturation fluence of 135 mJ/cm². Black and co-workers¹⁰¹ have found that the quantum yield of XeF is unity over this absorption band. For unknown reasons, the photodissociation of XeF₂ produces almost entirely XeF B-state molecules with little yield of XeF C-state molecules. This, then, provides a convenient technique for studying B- and C-state mixing and for separating properties (such as deactivation) of these two states.

The relevant energy levels of XeF are shown in Fig. 7-77. The conventional XeF laser operates on the B-X transition near 350 nm. Recently, laser oscillation has also been demonstrated¹⁰² for the very

Fig. 7-78. Fluorescence spectrum of XeF, obtained by electron-beam pumping an Ar-Xe-NF₃ mixture, showing the sharp B-X transition and the broadband C-A transition.

broad C-A transition centered near 480 nm. Typical fluorescence spectra for these two transitions are shown in Fig. 7-78.

Equilibrium at 300 K between the XeF B- and C-state populations can be established by collisions following the photolysis of XeF₂. The ratio of the fluorescence powers on the two transitions then implies the ratio of the B- and C-state populations, provided the ratio of the spontaneous emission coefficients is known. From the population ratio, the energy splitting between, and relative positions of, the two states are immediately determined. This technique has been used^{103,104} to establish the B-C splitting for XeF and to demonstrate that the C state is actually lower.

We have verified these results by using pulsed photolysis of XeF₂ at 172 nm. The ratio of the C-A to the B-X fluorescence intensity is plotted vs buffer-gas pressure in Fig. 7-79. Note that at low

Fig. 7-79. C-A/B-X fluorescence intensity ratio vs buffer-gas pressure for XeF lasers. The curves were calculated by using a simple model.

Fig. 7-80. Time dependence of XeF B-X and XeF C-A lasers pumped by 172-nm photolysis of XeF₂. The pump fluence was 150 mJ/cm², and the photolytic mixture was composed of 2 Torr of XeF₂, 80 Torr of SF₆, and 80 Torr of N₂.

pressure the ratio is very close to zero, implying that the photolysis produces mostly XeF B-state molecules. Collisional equilibrium between the B and C states is established for N₂ and SF₆ pressures above 1000 Torr and for Ar pressures considerably above 1000 Torr. At equilibrium, the intensity ratio approaches 3 (Refs. 103 and 104). The radiative lifetime of the C-A transition is about 100 ns, and that of the B-X transition is about 13 ns (Ref. 101); this implies that the XeF C-state population is 20 times the B-state population at thermal equilibrium at room temperature. If a Boltzmann distribution is assumed, the C state is lower by 650 ± 50 cm⁻¹. We have modeled the approach to equilibrium by varying the rates for the mixing process $\text{XeF(B)} + \text{M} \xrightarrow{\text{kin}} \text{XeF(C)} + \text{M}$. The backward rate was determined from the forward rate by the energy splitting and detailed balance. We computed the solid curves in Fig. 7-79 by using k_m equal to 1.2×10^{-11} cm³/s for SF₆ and N₂ and 2.1×10^{-12} cm³/s for Ar. These values are about a factor of two smaller than those found by Brashears and Setser.¹⁰³

The fact that the XeF C state is about 3 kT lower than the B state has profound importance for XeF lasers. To extract energy efficiently on the B-X transition, we must depend strongly on mixing processes. It is generally assumed, but not well proven, that electrons provide "complete mixing" between the B and C states in electron-beam-excited rare-gas halide lasers. Nonetheless, it is not surprising that heating improves the performance¹⁰⁵ of the 350-nm transition of XeF, since heating increases the equilibrium ratio between the B and C states.

The B-C splitting is probably smaller and less important for XeCl and KrF lasers than for XeF lasers.

We have also performed laser experiments on the B-X and C-A transitions using photolysis of XeF₂. Again, the diagnostics and the experimental setup were the same as those described in "Group VI Lasers," above. We were primarily interested in the XeF C-A laser because it has only recently been made to oscillate,¹⁰² but we were also interested in this laser in the fusion context. It has a broad bandwidth, which may be important for target coupling, and a small, but usable, energy storage time because of its 100-ns radiative lifetime. Using mirrors with high reflectivity, and operating at 480 nm with 2% output coupling, we obtained the results shown in Fig. 7-80. With a photolytic mixture of 2 Torr of XeF₂, 80 Torr of SF₆, and 80 Torr of N₂, and with a 172-nm pump fluence of 150 mJ/cm², we found an output of 8 mJ on the C-A transition with a peak power of 270 kW. This corresponds to a specific output energy greater than 1 J/litre, since the laser volume was about 6.5 cm³. The output energy on the B-X transition was approximately 100 times lower for these conditions. Taking into account the number of pump photons that were absorbed in the laser volume, we calculated that the C-A laser efficiency was 2.5% with respect to the absorbed energy at 172 nm. This is more than an order of magnitude smaller than the quantum-limited efficiency of 36%. We have no explanation for this discrepancy; however, we did not carefully optimize C-A output with respect to all parameters.

Fig. 7-81. Spectrum of XeF C-A laser pumped by 172-nm photolysis of XeF_2 . The absorption lines are caused by excited Xe atoms.

The XeF C-A laser spectrum is shown in Fig. 7-81. Although gain narrowing is evident, the laser spectrum is still quite broad. Perhaps the most interesting features of the spectrum are the various absorption lines. These absorptions (aside from the three unidentified features indicated by question marks) are caused by Xe atoms excited to the 6s level, the lowest excited configuration of Xe. It is impossible to create a Xe^* atom from XeF_2 with only a single photon at 172 nm; hence, we assume that Xe^* is created by the absorption of a "second photon" by the product XeF. Absorption by XeF could create either $\text{XeF}^+ + e$ or $\text{Xe}^{**} + \text{F}$. Either process ultimately leads to Xe^* . Discharge-pumped¹⁰⁶ and electron-beam-pumped¹⁰⁷ XeF C-A lasers also show these absorption lines. In the latter cases, the absorptions are very strong and obliterate most of the laser spectrum.

Photoabsorption of the vacuum ultraviolet (VUV) pump radiation by the product states causes the production of ions and free electrons. Because this phenomenon is reminiscent of our work on Group VI lasers, we investigated the effect of adding SF_6 , which rapidly attaches and removes electrons, to the laser mixture to determine if this addition would improve the XeF output. The influence of SF_6 is much less dramatic in this case than in the Group VI laser studies. The C-A laser usually performed better using mixtures containing SF_6 than it did with mixtures containing pure N_2 ;

however, this improvement was not dramatic. It is likely that electron recombination with XeF^+ is relatively fast; this is not the case in Group VI systems, where electron attachment to SF_5 is necessary for rapid electron removal.

We also measured the XeF C-A gain using a 488-nm Ar^+ laser. We found a peak gain of $6\% \text{ cm}^{-1}$ for a 2-Torr XeF_2 , 1000-Torr SF_6 mixture. This gain is consistent with unity quantum yield to XeF and a C-A gain cross section of about $1 \times 10^{-17} \text{ cm}^2$, as computed from the line width and radiative lifetime.

XeCl Experiments. We performed experiments on XeCl using both photolytic and electron-beam pumping. In the photolytic work, we used Kr_2 radiation at 146 nm to excite Xe via its 147-nm resonance line. The excited Xe reacted with Cl_2 to form XeCl, which then combined with Xe to form the trimer Xe_3Cl . The Xe_3Cl trimer emits a broad band, resembling the C-A transition of XeF, and is interesting not only as a potential laser candidate¹⁰⁸ but also as an important absorber at 308 nm, the primary laser transition of XeCl. Because of its ionic character, Xe_3Cl should absorb much like Xe_2^+ (Ref. 109).

We observed both XeCl B-X emission and Xe_3Cl emission using the photolytic technique. The B-X emission was maximum for a mixture of 2 Torr of Cl_2 and 100 Torr of Xe, while the Xe_3Cl intensity was strongest for a mixture of 2 Torr of Cl_2 , 400 Torr of Xe, and about 3 atm of Ar. However, the fluorescence intensities were an order of magnitude below the XeF intensity observed from XeF_2 photolysis. The fluorescence was probably weaker because of imperfect coupling of the Xe absorption to the Kr_2 emission; only a relatively small fraction of the Kr_2 radiation was strongly absorbed by the Xe, and the remainder of the spectrum was absorbed weakly. At the high Xe pressures required to absorb most of the Kr_2 input, the strongest absorption was concentrated at the input window and was inaccessible.

We obtained a relatively weak laser oscillation on the XeCl B-X transition by photolytic pumping, but we were unsuccessful in Xe_3Cl laser attempts, presumably because of the weak pumping rate. Also, since Xe was used as a precursor of Xe_3Cl , it seems likely that Xe^* absorption, such as found in

the XeF C-A laser, would be very significant for Xe₂Cl oscillation.

We have recently begun studies of electron-beam-pumped XeCl lasers. Previous experiments have shown that the XeCl B-X transition can operate with a medium efficiency (5%) approximately one-half that of KrF.⁹⁹ Measurements of gain and absorption in the spectral region around 308 nm suggest that the gain/loss ratio is about 10 for a Ne-Xe-HCl mixture.¹⁰⁰ The absorption appears to be broadband in its spectrum and may be caused by rare-gas molecular ions.

During 1979, we studied XeCl fluorescence as a function of gas mixture and electron-beam current density, and laser experiments are planned for 1980. The XeCl fluorescence was found to depend strongly on the HCl concentration; the XeCl intensity was maximum at 3.5 Torr of HCl for all Xe-Ne mixtures. Higher HCl pressures apparently caused strong deactivation of XeCl relative to radiative decay. (The HCl quenching coefficient of XeCl was recently determined¹⁰¹ to be $8 \times 10^{-10} \text{ cm}^3/\text{s}$.) The XeCl fluorescence exhibited a broad maximum at Xe pressures between 25 and 200 Torr. In contrast to previous laser studies,⁹⁹ we found that 4 atm of Ne was not required for optimum fluorescence. Using optimum HCl and Xe concentrations, we found that the XeCl fluorescence efficiency was roughly constant between 1 and 4 atm of Ne and then declined at higher pressures. The peak in the laser efficiency previously found at 4 atm was likely caused by using a constant-ratio mixture; the peak occurred because of the variation in HCl partial pressure and does not represent a fundamental limitation.

We also studied XeCl fluorescence as a function of electron-beam current density for Ne-Xe-HCl and Ne-Xe-Cl₂ mixtures. We varied the current density by placing a perforated-carbon beam stop in the electron-beam drift region; the beam profile became uniform after passing through this plate. Figure 7-82 shows the current-density variation for mixtures containing 3.3 Torr of either HCl or Cl₂, 66 Torr of Xe, and 3300 Torr of Ne. The Y-axis in Fig. 7-82 is proportional to fluorescence efficiency since we have plotted the peak signal divided by current. The current-density variations for HCl and Cl₂ mixtures are clearly quite different. The

Fig. 7-82. Fluorescence efficiency vs current density for electron-beam pumping of mixtures of 3.3 Torr of either HCl or Cl₂, 66 Torr of Xe, and 3300 Torr of Ne.

fluorescence efficiency with HCl increases with increasing current up to about 100 A/cm²; for Cl₂, the efficiency is constant at low currents and then declines when the current rises above 20 A/cm². Electron deactivation presumably causes the rollover in efficiency at high currents for both cases. Judged by the XeCl fluorescence, HCl should be a better laser fuel at current densities of around 100 A/cm². We believe that the differences between HCl and Cl₂ are primarily caused by their attachment properties.

We are presently modeling the kinetics of the XeCl laser medium.

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ASE in Bidirectional Amplifiers

In 1979, we extended our study of optical field dynamics and amplified spontaneous emission (ASE) to include bidirectional and folded amplifiers. The bidirectional amplifier is a mirrorless amplifier through which two oppositely directed optical pulses pass simultaneously. The pulses overlap spatially and fill the entire amplifier volume. This device has better control of parasitic oscillations and ASE than unidirectional amplifiers because the medium is filled throughout with signal fluence of greater than a saturation intensity, and the gain is therefore loaded at every point. A very similar device is the folded amplifier, which has a mirror only at one end. The folded amplifier is also similar to the regenerative amplifier; indeed, it would be such an amplifier if the mirror were moved sufficiently far back that the signal fluences did not overlap itself inside the medium. In cw operation, the folded amplifier is exactly equivalent to one-half of the bidirectional amplifier (Fig. 7-83). Placing a 100% reflective mirror at the exact midpoint of a bidirectional amplifier creates a folded amplifier that has exactly the same signal fluences as the bidirectional amplifier. On reflection, it is clear that the full optical behavior of the folded amplifier—including ASE and various parasitic waves—must be identical to the equivalent bidirectional amplifier because of the imaging properties of plane mirrors. Because the bidirectional amplifier is symmetric, the right-traveling waves of all kinds at the midplane match the left-traveling waves exactly. This situation is exactly recreated by a 100% reflectivity plane

Fig. 7-83. The bidirectional and folded configurations.

Table 7-11. Equivalence parameters of folded and bidirectional amplifier.

	Folded amplifier	Bidirectional amplifier
Length	L	$2L$
Aperture	A	A
Gain	α	α
Loss (nonsaturating)	γ	γ
Input per side	I_{in}	I_{in}
Total input	I_{in}	$2I_{in}$
Power gain	P	P
Output per side	I_{out}	I_{out}
Total output	I_{out}	$2I_{out}$
Extraction efficiency	η	η
Gain loading	$\phi(z)$	$\phi(z)$
Stored power	P_{av}	$2P_{av}$

mirror placed at the midplane. As a consequence, there exists an exact equivalence, which is illustrated in Table 7-11. This table gives the parameters of the equivalent bidirectional amplifier required to calculate the properties of a given folded amplifier. It also permits us to translate folding into bidirectional calculations.

The effects of ASE on bidirectional amplifier performance have been calculated in a manner similar to the calculations carried out last year¹¹² on unidirectional amplifiers. The transverse variations in the gain and optical fields are neglected, and the walls are assumed either to be perfectly absorbing or to have a reflectivity of at most a few percent. The only modification to the mirrorless amplifier model is to include a second signal wave traveling in

Fig. 7-84. Extraction efficiency vs input intensity in units of I_{sat} for a unidirectional amplifier with $\alpha_0/\gamma = 15$, $\gamma L = 1$, and $\tau/\tau_R = 0.33$. The curves are parametric in the aspect ratio, a , defined in terms of the solid angle of the exist aperture at the input $\delta\Omega = \pi a^2$.

Fig. 7-85. Extraction efficiency vs input intensity in units of I_{sat} for a bidirectional amplifier with $\alpha_0/\gamma = 15$, $\gamma L = 1$, and $\tau/\tau_R = 0.33$. The curves are parametric in the aspect ratio, a , which is defined in the text. Also plotted is the ideal case of a unidirectional amplifier without ASE effects (curve labeled A), for comparison. (The difference between A and the $a = 0.05$ curve is due partially to ASE and partially to the different intensity distributions in the uni- and bidirectional amplifiers.)

the opposite direction to the first wave. The resultant code is almost identical in accuracy and time to the unidirectional code described previously.¹¹² To illustrate the improved control of ASE in bidirectional amplifiers, we plot in Figs. 7-84 and 7-85 the extraction efficiency of uni- and bidirectional amplifiers as a function of input intensity. It is clear from the comparison case (labeled A) in Fig. 7-85

that the debilitating effects of ASE are less for the bidirectional case, and, moreover, that the peak extraction efficiency is greater for narrow bidirectional amplifiers than for unidirectional amplifiers, even excluding ASE. These curves are plotted parametrically in the aspect ratio, a , defined in terms of the solid angle of the output aperture at the input

$$\delta\Omega = \pi a^2.$$

From the general comparative study of bidirectional and unidirectional amplifiers we have carried out, it is clear that both amplifiers have similar extraction characteristics but that the bidirectional amplifier offers the advantage of large signal fluence everywhere in the gain medium. Thus, unlike the unidirectional case, no region of the amplifier is unloaded, and the high small-signal gain is suppressed everywhere. This allows greater parasitic control, and the parasitic limits on the amplifiers allow larger aspect ratios for the bidirectional case. In turn, the greater transverse dimension permits a larger total energy per amplifier module.

Author: D. Eimerl

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112. *Laser Program Annual Report—1978*, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-50021-78 (1979), p. 8-14.

Transient Laser Dynamics

Among the various analytical problems posed by the RAPIER program, perhaps the most challenging is that regarding injection locking. Injection locking was used to control the laser line width in initial experiments on the Raman compressor, a device that requires a narrow pump-laser line width ($\sim 0.1 \text{ cm}^{-1}$). By contrast, the pump-laser medium (KrF), left to itself, lases with a wide line width ($\sim 50 \text{ cm}^{-1}$).

During the initial experiments, it soon became apparent that injection locking provided two additional benefits: it could be used to control the spatial beam quality, which is important because of the propagation distances involved in a compressor-stacker system; and it produces a large power gain, which can lead to a reduction in the number of components in the KrF amplifier chain. To realize these potential benefits of injection locking, it became necessary to develop analytical techniques that

Fig. 7-86. Signal and ASE dynamics in the transient oscillator. The relationship of signal output to input and the use of retarded times for the ASE components are shown.

could relate parameters of the laser source and amplifier to resultant line width and spatial beam quality.

Transient Injection Locking Analysis. One promising technique of injection locking involves the use of unstable resonator optics to set up what is essentially a three-pass folded amplifier. In general, the pulse length of the source for injection locking does not have the same pumping time as the KrF amplifier. Therefore, the analysis must consider the transient control of a high-magnification unstable resonator in a high-gain laser medium; in particular, the analysis must address the relationship of the laser aperture to the degree of control that can be obtained with various optical configurations.

To achieve the above requirements, a code was developed that treats amplified spontaneous emission (ASE) in a fully transient manner. The analysis of ASE in the transient case is similar to the analyses previously presented¹¹³ in the steady-state case; namely, an analysis must be made of those rays that are emitted in the body of the medium, travel through the medium, experience gain, reach a saturation intensity, and then impinge on the wall. These rays affect the history and steady-state value of the gain through the usual mechanism of gain saturation; however, they cannot be represented as

a source term in the differential equations of energy transport in a way that does not require a prior knowledge of the solution of those equations. Consequently, we adopted an integrodifferential equation approach, which is the time-dependent generalization of the integral-equation formulation of the steady-state problems analyzed in 1979, and the comments made in the 1978 annual report¹¹³ with respect to the limitations and idealizations of this approach apply here also.

The basic dynamics of the problem are illustrated in Fig. 7-86. The signal field enters from the left, experiences a number of reflections, and at each reflection makes a contribution to the output fluence. The output is, therefore, a sum over the signal field evaluated at various retarded times and is given by

$$I_{out}(t) = \sum_j I_{in}(t - j\tau) \quad (54)$$

where τ is the round-trip time of the cavity. Similarly, the ASE at the output is an integral, over the complete history of the cavity, of the sources, the gain experienced at earlier times, and that portion of the source that couples with the exit aperture:

$$I_{ase} = \int (\text{source})(\text{coupling})(\text{retarded gain}) \quad (55)$$

To unfold the contribution to the ASE from the region $x_{ret} > L$, where L is the cavity length, we unfold the resonator as illustrated in Fig. 7-87. Consider a resonator with plane mirrors. The contribution to the ASE for time $t' > t - \tau/2$ is given by the extreme right box. For times $t - \tau > t' > t - \tau/2$, this contribution is given by the second adjacent box. In this latter box, the waves are traveling to the left in the cavity but to the right in the unfolded scheme described here. The ASE originating at earlier times is evaluated in boxes successively further to the left. The contributions from the different boxes differ not only in the time at which the source is evaluated but also, and more importantly, in the fact that the further to the left a box lies, the smaller the coupling into the output aperture. The ASE from the remote past will most likely impinge on a wall because only those rays close to the optical axis of the cavity will remain in the cavity for a sufficient time to exit through the output aperture. Thus, since the ASE is

Fig. 7-87. The unfolded resonator. The signal propagates to the right, the ASE is isotropic, and only a portion of the ASE at x reaches the plane at x' , rather than impinging on a wall. The gain is also unfolded. Each box corresponds to a single pass across the cavity.

isotropic, the relevant portion to include is simply given by the solid angle of the exit aperture as seen at the point of origin through many reflections. It might be said that remote images of the ASE source at the exit aperture have very small solid angles. Thus, in the unfolded scheme using plane mirrors, the ASE is evaluated as if the unfolded resonator were a superfluorescer and there were lossy elements at the boundaries between the boxes.

A similar argument holds for the signal. The signal propagates to the right in the unfolded resonator as if it were traveling in a medium of length Nl where N is the number of boxes. However, from Eq. (54), the output at any time is given by the sum of the signal fields at the points corresponding to the exit aperture, labeled f in Fig. 7-87. Similarly, the backward fluence is given by the sum of the signal fields at the points labeled b (Fig. 7-87). Of course, the gain and loss due to the medium must be unfolded so that every other box has the same gain and loss as a function of position. Adjacent boxes have inverted gain and loss. Namely, for the j th box,

$$\alpha_{j+2}(x) = \alpha_j(x) = \alpha_{j \pm 1}(L - x) \quad (56)$$

Thus, the gains at the points labeled x in Fig. 7-87 are the same.

Returning briefly now to the evaluation of the ASE, it should be noted that the ASE evaluated in

the third box from the right is identical to that in the box furthest to the right because the medium is unfolded as well as the resonator itself. Thus, the ASE needs to be evaluated only in the two furthest-right boxes, from which the total forward and backward traveling components are obtained. Thus, the total optical field at any point x (Fig. 7-87) is the sum of the values for signal fluence obtained from the appropriate point in each box and from the two ASE terms

$$I_{tot} = \sum_j I + I_{ase}^f + I_{ase}^b \quad (57)$$

The gain and loss of the medium respond dynamically through stimulated emission, in the usual manner, to this total local irradiance. Thus, if a kinetic model of the medium is included in the code, it must use I_{tot} as the appropriate optical irradiance in the kinetic equations.

It is possible to design a treatment of transverse parasitic oscillations in the code by including a transverse irradiance, $I_t(x)$, that experiences gain in the medium, $\alpha(x)$, loss at the walls, and some coupling to the spontaneous emission. However, the relevance of such a term to real parasitic waves is unclear.

Figure 7-88 shows a space-time schematic of the unfolded resonator that illustrates the different manners in which signal fluence and ASE are

Fig. 7-88. Space-time schematic of the unfolded resonator, showing the calculational procedure for signal, ASE, and gain matrix.

treated in the code. The signal propagates at each time step, using the gain evaluated at the relevant time. However, the code reevaluates the ASE completely at each time step by using the complete space-time history of the medium, which is stored by the code and built up as the code runs. In this way, the rays of ASE that are lost by impinging on the cavity walls are treated correctly at each time step. The complete space-time history of the gain so obtained not only gives a great depth of insight into the transient laser dynamics but also is necessary to treat the ASE effectively.

Integro-differential Equations of the Transient Model. To set up the calculation, the total pumping time of the gain medium is used to determine the equivalent n of passes, where $n = 2L/\tau$. The unfolded resonator is divided into n segments, or boxes, and is consequently nL in length. The signal input is specified on the extreme left edge of the unfolded resonator and propagates according to the familiar equation of energy transport,

$$\left[\partial/\partial z + \partial/\partial(ct) \right] I(z, t) = g I(z, t) \quad (58)$$

For KrF, the simplest kinetic model is a one-state model,

$$(\partial/\partial t + 1/z) \alpha = \frac{P}{\tau} - (n/h\nu) I_{\text{tot}} \quad (59)$$

for the gain, α , and $\gamma = \text{constant}$ for the losses. Thus, the net gain, g , is

$$g = \alpha - \gamma \quad (60)$$

The ASE at position x is a straightforward statement of Eq. (55):

$$I_{\text{ase}}(x, t) = (z/z_r) \int_0^x dx' a(x', t') \times \frac{\delta\Omega(x, x')}{4\pi} \exp[G(x, x', t)] \quad (55a)$$

where the retarded time is

$$t' = t - (x - x')/c \quad (61)$$

and $\delta\Omega(x, x')$ is the solid angle of the cavity cross section at x' as seen at the point x . The gain term in Eq. (61) is

$$G(x, x', t) = \int_x^{x'} dx'' [a(x'', t'') - \gamma(x'', t'')] \quad (62)$$

where the retarded time is

$$t'' = t - (x' - x'')/c \quad (63)$$

These expressions for total intensity, ASE, at medium-gain saturation, combined with the definitions of output irradiances, constitute the unfolded resonator model. These equations define the transient laser dynamics model used to analyze the A-amplifier experiments.

A representative set of calculations made by using this transient laser dynamics model is shown in Figs. 7-89 through 7-94. Figure 7-89 shows the experimental arrangement that we want to model. In the calculation, the A-amplifier cavity was not filled completely with the gain medium but contained both a region of gain medium and a roughly equal region of empty space. The small signal gain in the cavity was approximately e^5 ; it was pumped [see Eq. (59)] by a term, P , which approached its steady-state value exponentially in about one round trip.

The pump function is plotted in Fig. 7-90. Note that the gain is only one-half that shown because the cavity is only half filled; namely, $L_{\text{medium}} = 1/2 L_{\text{cavity}} = 1/2 L$.

The injected pulse shape is shown in Fig. 7-91. This pulse shape is a simulation of the experimentally obtained injection pulse. The gain history of the medium is shown in Fig. 7-92. The horizontal axis (number of passes) is time measured, and the z -axis (going back into the page) represents space.

Fig. 7-89. Experimental arrangement of A-amplifier, which is modeled by the transient laser dynamics code.

Fig. 7-90. The pump function, P in Eq. (59), which is used in the one-state kinetic model for KrF.

The imprint of the gain of the waves as they propagate back and forth in the cavity is evident.

Figure 7-93 plots the output of the cavity. In this case, the transient character of the process is very clear: in 14 passes, the steady-state configuration has not been reached.

Figure 7-94 plots the ASE output, which is very small in this case. The peaks correlate with the gain. The ASE ultimately falls off as the cavity fluence builds up.

A complete set of runs was made with the unfolded resonator model to predict the injection-locking power requirement for the A-amplifier. The

Fig. 7-91. A simulated input pulse shape for the injection-locking experiments.

injected power required to achieve equal levels of ASE energy and signal energy in the output was predicted to be 0.3 mJ, compared with the ex-

Fig. 7-92. Excited-state population (KrF) in the transient oscillator. The imprint made on the gain by waves in the cavity is evident.

Fig. 7-93. Output of the oscillator. The transient behavior has persisted throughout the operation of the laser, and no steady-state configuration is reached.

Fig. 7-94. ASE output of the oscillator. Correspondence with the graph of the excited-state population (Fig. 7-92) is evident.

perimental value of 0.1 mJ. The difference is attributable to the assumptions of the model; namely, the modeling of the unstable resonator by an

"equivalent" plane oscillator, and the assumption of transverse uniformity of the medium.

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Analysis of Parasitic Oscillations

An important part of the task of designing multimegajoule lasers is to establish criteria for the threshold, the rate of growth, and the suppression of parasitic oscillations. It is not yet possible to devise simple, meaningful models that will give significant information about parasitic oscillations in a real laser; however, we have devised two simple, idealized model problems that should give us some insight into the limitations of more realistic model systems. These two idealized models are the "glint" model and the "diffuse-wall" model.

The Glint Model. Any real laser design is likely to have certain closed paths involving multiple reflections off of various structural elements. The more obvious of these paths can easily be discovered, by examination or by trial and error, and then eliminated or baffled. However, a real system can be expected to still retain a certain number of glints, i.e., discrete, brightly reflecting spots such as bolt heads, sharp corners, or random bright patches.

To model these glints in a generalized way, we can define each glint as a diffuse point reflector (i.e., as a reflector that would uniformly reflect any incident light back out into 2π steradians) and assign to each glint a capture area or cross section. This cross section would define how much light the glint would capture from an incident wave and how much light it would reflect back out. We could then specify a gain medium, with some simple geometry and a given gain coefficient, and randomly distribute some number of glints with given cross sections around the surface of the gain medium. A simple computer simulation could then be made by assuming some initial light fluence impinging on each glint and calculating the reflected and amplified fluence reaching each glint as a result of the initial fluence reflecting from all the other glints. The simulation would be iterated as necessary to determine if the fluence levels hitting the glints would grow or decay with repeated iterations.

Since the glint locations and strengths would presumably be random (chosen by the computer

before each run), the calculations would have statistical or Monte Carlo characteristics, perhaps matching the nature of the real physical problem. The object of this exercise would be to discover empirically what ranges of gain coefficients and total glint cross sections would lead to apparently stable or unstable behavior. We could then specify a tentative maximum total glint cross section allowable in a real laser.

Detailed calculations using this glint model have not yet been carried out.

The Diffuse-Wall Model. The diffuse-wall model assumes a gain medium bounded by a certain number of diffusely reflecting walls. The computer simulation for the diffuse-wall model is similar to that for the glint model, described above, but with the glints replaced by a suitable number of small subareas on the diffuse walls. The significant question, of course, is what combinations of laser gain, diffuse-wall reflectivity, and geometry lead to growing or decaying intensity distributions in simple iterations of the model.

This model has a certain reasonableness, in that the diffuse walls may represent a limiting best-case model for the foils unavoidably present in an electron-beam laser; if a wall is truly diffuse, its exact shape or slope should not matter. If the computer simulations indicate that diffuse walls are acceptable, a straightforward definition of wall reflectivity characteristics could be made, towards which laser designers and materials suppliers could strive.

In the diffuse-wall model, we assume a rectangular gain volume that has dimensions of length, L , width, w , and height, h ; that has two diffusely reflecting walls with total power reflectivity of R ; and that is entirely filled with a uniform gain medium having a power-gain coefficient of g_0 (Fig. 7-95). Whether this is a saturated or an unsaturated gain coefficient is immaterial to these calculations; only the geometrical ratios w/L and h/L and the axial gain value g_0L are significant in the analysis.

In the simplest case, the gain medium is assumed to have two diffusely reflecting, opposed walls in the (x,y) or (w,L) planes, as shown in Fig. 7-95. These walls are assumed to have a uniform lambertian reflectivity.

The analysis is carried out by assuming an initial intensity distribution of laser radiation, $I_0(x_0,y_0)$, incident on one of the two walls. This distribution is presumably incoherent in its spectral

Fig. 7-95. Basic model for diffuse parasitic oscillations. The volume is assumed to be filled with a uniform gain medium having power gain coefficient, g_0 , and the two walls are assumed to be diffusely reflecting with total power reflectivity, R .

and angular characteristics, but, because of the diffuse reflectivity of the surface, the detailed nature of the incident radiation is irrelevant.

Given the distribution $I_0(x_0, y_0)$ on the lower wall, we can evaluate the reflected, amplified intensity $I(x, y)$ reaching each point (x, y) on the upper plane from all possible points (x_0, y_0) on the lower plane. For the computer evaluation, both distributions are, of course, sampled at a suitable number of discrete points on the two planes. We can then iterate this procedure back and forth between the two walls in a procedure reminiscent of the Fox and Li¹¹⁴ procedure for finding optical resonator mode patterns, except that incoherent intensities rather than coherent fields are iterated and the iteration kernel contains only gain and not phase shift.

We have used the diffuse-wall model to estimate, for a specified uniform power gain $= \exp(g_0 L)$ in the axial (x) direction, and for a specified geometry, what overall or total diffuse reflectivity (R_{th}) for the side walls will just create a threshold condition for a diffuse parasitic oscillation in this structure.

The general result obtained is that the intensity distribution, $I(x, y)$, on the walls rapidly converges on successive bounces to a simple, smooth pattern with a weak maximum or minimum (depending on circumstances) in the center. The total power in this distribution then grows (or decays) by a constant factor on each successive bounce, with the growth

Fig. 7-96. The total diffuse power reflection factor, R_{th} , at which a diffuse parasitic oscillation will just reach threshold, is plotted vs the axial gain of the laser medium for a rectangular gain volume with height and width equal to one-half the length. The two lines indicate computer simulations with different numbers of total array points (N).

or decay rate per bounce depending on the axial gain coefficient, $g_0 L$, and the geometrical factors, w/L and h/L .

We interpret this result by the following rationale. If the intensity pattern grows by a growth factor, F , in one transit, then a net total power reflection coefficient, $K = R_{th} = 1/F$, for the diffusely reflecting wall would just cancel this growth. (By the reflectivity coefficient, R , we mean the net fraction of the incident power that is reflected diffusely into a lambertian pattern by the wall.) For this set of parameters— $g_0 L$, w/L , and h/L —the value of $R_{th} = 1/F$ presumably represents a threshold reflectivity for the growth of a diffuse mode of parasitic oscillations having the distribution $I(x, y)$. Lower wall reflectivities ($R < R_{th}$) should then mean stability—and higher reflectivities should mean instability—for a particular gain and geometry. The main outputs of this analysis are plots of the threshold wall reflectivity, R_{th} (%), vs the axial gain, $G = \exp(g_0 L)$, for different geometrical ratios, as presented in Figs. 7-96 through 7-98.

It might appear that the net growth ratio per bounce, i.e., FR for $R > R_{th}$, could be interpreted as the physically real growth rate per bounce for the buildup of the same transient intensity distribution,

Fig. 7-97. Curves similar to Fig. 7-96 for three lasers having the same total volume of laser medium but different height/width ratios.

$I(x,y,t)$, for reflectivities away from the threshold value. However, this is not a valid interpretation. To calculate $I(x,y,t)$ on any one bounce in the computer simulation, we use $I_0(x_0,y_0,t_0)$ one bounce earlier. However, there are significantly different transit times for light from different points (x_0,y_0) on the input plane to points (x,y) on the output plane. Hence, in a transient case, we would need to know $I_0(x_0,y_0,t_0)$ for different earlier times (t_0) and for different pairs of points (x,y) and (x_0,y_0) . This problem is avoided exactly at threshold, where $FR = FR_{th} = 1$ and, therefore, the intensity distribution should be stationary in time if $R = R_{th}$.

Figure 7-96 plots, as a typical result, the threshold diffuse total reflectivity for parasitic oscillation vs the axial power gain for the laser medium. This plot is for a typical geometry in which the height and width of the laser volume are both equal to one-half the length, h . The results shown are independent of absolute size of the laser and depend only on overall total gain. At low gains, of course, the parasitic growth coefficient is so small that oscillations would not occur even with 100% (diffusely) reflecting walls. In Fig. 7-96, the two curves represent 32 and 128 array points (4 by 8 cells and 8 by 16 cells, respectively, in one quadrant of each plane), and the close agreement of these curves in-

Fig. 7-98. Threshold reflectivity vs width/height ratio at two different axial gain values, maintaining a constant total laser volume such that $wh = 0.25 L^2$.

icates that the computer simulation appears to have adequate precision.

Figure 7-97 shows plots of similar results for three cases with the same total volume of laser medium but different ratios of height to width. Figure 7-98 shows threshold reflectivity vs width/height ratio for constant volume and two fixed gains. Somewhat surprisingly, a wide, low-profile geometry is more stable against this kind of diffuse parasitics than is a tall, narrow geometry. The reason presumably is that the longer transverse gain path in the latter case (at constant volume and constant g_0) more than makes up for the larger leakage out of the two open sides.

The main conclusion to be drawn from these results would seem to be that, for a high-gain medium, even totally diffuse reflecting walls must have a quite low total reflectivity if the laser is to be stable against a diffuse kind of parasitic oscillation. Varying the laser geometry can help some to improve this situation, but not a great deal.

This model assumes, of course, a highly ideal, purely diffuse wall material. Whether such a material can be obtained is another question. The assumption of totally diffuse reflection greatly simplifies the calculation procedure, since we need to know only the total intensity striking any cell of the

surface without needing to know the direction of incidence. Attempting to apply this procedure to a wall model in which the reflected distribution depended upon the incident direction would make the formulation considerably more complex, though probably still tractable, and would require substantially larger computer codes. Including a genuinely specular, coherent reflected component would become still more difficult because of the problem of handling the phases of any coherent waves.

Another problem still basically lacking any solution is how to formulate general, meaningful models for parasitic oscillations (other than this model and the glint model) that would be reasonably close to real lasers and yet not be simply a brute-force analysis of a specific, detailed laser design. Until these general, meaningful models are developed, the best approaches to analyzing parasitic oscillations seem to be the glint and diffuse-wall models.

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Electron-KrF and Electron-XeF Collision Processes

Much of the recent theoretical modeling¹¹⁵⁻¹¹⁸ of the rare-gas fluorides—e.g., KrF and XeF—has pointed to the importance of various electron-excimer scattering processes in electron-beam-pumped or discharge-pumped lasers. These processes include direct quenching of the ionic excimer states to the lower, dissociative, covalent states; mixing of the B, C, and D excited states; and dissociative attachment of electrons to the excimers. The possible role of negative-ion resonance states in electron collisions with rare-gas halides has also been recognized.^{118,119} However, these earlier studies produced only indirect experimental information and no quantitative estimates of rates or cross sections for any of these processes. To obtain such quantitative data, we calculated cross sections and rate coefficients for the electron-impact deexcitation of KrF and XeF excimers via the optical y allowed transitions

where $Rg = Kr$ or Xe .

In our study,¹²⁰ we used a modified impact-parameter technique, along with molecular wave functions calculated from first principles (ab initio), to calculate the superelastic cross sections for Eq. (64). Recent applications of this technique to optically allowed excitations in simple diatomic molecules (e.g., H_2) yielded cross sections that were within 25% of those calculated with the best available techniques (the close-coupling technique and the distorted-wave technique),¹²¹ even at energies within several electron volts of the excitation threshold. The section, "Electron Scattering Processes," below, presents a more detailed discussion of the modified impact-parameter technique and its application to H_2 and N_2 .

As a first step, we used large, ab initio configuration-interaction (CI) wave functions to calculate the Born cross sections and electric-dipole transition moments for the $1^2\Sigma^+ \rightarrow 2^2\Sigma^+$ and $1^2\Pi \rightarrow 2^2\Pi$ transitions without accounting for spin-orbit coupling.

In the case of KrF, all electrons were treated explicitly, and the wave functions, energies, and transition moments that we obtained are the same as those obtained by Hay and Dunning.¹²² In the case of XeF, we employed a relativistic effective core potential¹²³ to describe the inner, inactive electrons of Xe, but we used the same type of CI wave functions for the valence electrons that we used for KrF. We also evaluated the excitation cross sections using the calculated transition energies and dipole transition moments in the modified impact-parameter formulas. The cutoff in the impact parameter was chosen to reproduce the Born cross sections at an impact energy of 100 eV. The superelastic cross sections were then calculated by using the transition energies and dipole moments obtained previously^{122,124} with spin-orbit coupling. Since the impact-parameter technique fails very close to threshold, the calculated cross sections between 2 and 3 eV were fitted to the threshold law, $\sigma \sim E^{-1/2}$, appropriate for the transitions in Eq. (64). Figures 7-99 and 7-100 display, for KrF and XeF, respectively, the resulting deexcitation cross sections for the five strongest transitions.

Finally, we used these quenching cross sections, and an assumed Maxwell-Boltzmann electron distribution, to obtain temperature-dependent rate

$$e^- + RgF(B, C, D) \rightarrow e^- + RgF(X, A), \quad (64)$$

Fig. 7-99. Cross sections for direct quenching of KrF excimers by electrons.

Fig. 7-100. Cross sections for direct quenching of XeF excimers by electrons.

coefficients. Direct electron deactivation of the excimer states is dominated by the B-X transition, with maximum rate coefficients of $3 \times 10^{-8} \text{ cm}^3/\text{s}$ and $7 \times 10^{-8} \text{ cm}^3/\text{s}$ for KrF and XeF, respectively. The rate coefficients obtained with the Maxwellian velocity distribution depend only weakly on temperature between 0 and 5 eV. The rate coefficients are sufficiently large so that, at typical electron densities ($3 \times 10^{15} \text{ electrons/cm}^3$), the quenching rates due to electron collisions are comparable to other deexcitation processes previously considered.¹¹⁷

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Vibrational Quenching of Excited Rare-Gas Halide Molecules by Charge-Exchange Collisions

The early RGH spectra taken by Golde¹²⁵ and by Velazco and Setser¹²⁶ show a very broad continuum extending far into the ultraviolet. These spectra, taken at very low pressures (several Torr), also exhibit a large degree of vibrational and rotational excitation. Spectra taken at higher pressures (1 to 5 atm) show a much narrower band, with well-defined structure attributed to bound-to-free transitions from individual vibrational levels of the upper state. An interesting feature of the RGH spectra is that they require only relatively small increases in pressure to collapse the Golde spectra to the high-pressure spectra of Murray and Powell.¹²⁷ This phenomenon raises a question as to how the vibrational-state distribution, which initially is at the top of the potential well, can relax so rapidly at these relatively low pressures ($\sim 100 \text{ Torr}$). Also, it is important to understand the effects of different buffer gases on the rate of vibrational relaxation. We know experimentally that the emission spectrum of electron-beam-excited KrF appears quite different when neon is substituted for argon as a

buffer gas.¹²⁸ The Ne-Kr-F₂ spectrum shows considerably more vibrational excitation than the Ar-Kr-F₂ spectrum. Discharge RGH lasers typically use helium as a buffer gas, which may be even less effective than either neon or argon as a quencher. Conditions in which the higher vibrational levels of the upper laser levels are significantly populated because of a relatively inefficient quencher could lead to laser inhomogeneity and lower efficiency.

It is known that discharge RGH lasers have a medium efficiency lower than that of electron-beam-pumped RGH lasers, and we want to determine the contribution of buffer-gas effects to this difference. Also, long-pulse operation is more efficient than short-pulse operation, again suggesting a relationship to the vibrational relaxation time for the upper level. The RGH molecule, after being formed in extremely high vibrational levels, must relax through hundreds of levels to the lowest several levels that account for most of the population observed in emission. The radiative lifetime of the III₁ 2 upper laser level of KrF is ~7 ns (Ref. 129) and for ArF is ~4 ns (Ref. 130). If the vibrational relaxation were essentially a diffusion down the dense manifold of states, it could not compete with quenching losses from the higher levels due to superelastic-electron or heavy-particle collisions. Obviously, what is needed is a mechanism that allows for large jumps down the vibrational manifold.

Of several possible mechanisms for vibrational quenching of excited RGH molecules, one that appears to allow for transfer of several quanta per collision involves symmetric charge exchange between the thermal Rg (Rare-gas) atoms of the buffer gas and the Rg⁺ of the RgF molecule. We developed a model that allows us to determine whether this mechanism can account for the observed rapid quenching. The model was applied to KrF and demonstrated that relaxation to the lower vibrational levels occurs in tens of nanoseconds at 1 atm. The model is general and applies to any RGH laser (e.g., ArF, XeF, XeCl), in addition to KrF.

A schematic of the charge-exchange model is shown in Fig. 7-101. We assume that the charge exchange occurs in a C_{2v} geometry (r₁ = r₂), where the two diabatic surfaces, Rg' + RgF and Rg'F + Rg,

Fig. 7-101. Schematic model for charge-exchange quenching of KrF by Kr. The Kr⁺ and F⁻ represent the initial vibrating molecule with internuclear separation, r₁. The thermal Kr atom, a distance r₂ from the F⁻ ion, will exchange its charge with Kr⁺ when r₁ = r₂. The initial vibrational velocity of the F⁻ ion is given by v_i, and the final velocity of the new diatomic molecule is given by v_f.

cross. Charge exchange for r₁ < r₂ does not conserve energy and corresponds to jumping to the upper surface.

The adiabatic surface for the collinear exchange reaction for Ar' + ArF shows the slight hump at C_{2v} geometries corresponding to the avoided crossing. After charge exchange occurs between the Rg atoms, the final vibrational energy can be estimated by projecting the F⁻ velocity vector, v_i, onto the new internuclear axis (r₂ in Fig. 7-101). The thermal velocity of the Rg buffer gas is negligible (~√kT/m). If all C_{2v} geometries are equally probable, then, on the average, one-half of the F⁻ kinetic energy remains as vibration and one-half appears as rotation. The Rg' atom originally bound to the F⁻ carries away all of its own kinetic energy. Therefore, for a given r (= r₁ = r₂),

$$\begin{aligned}
 & \text{(vibrational energy after collision)} \\
 &= 1/2 \left[1/2 m_F \left(\frac{m_R}{m_F + m_{Rg}} \right)^2 v_i^2 \right] \\
 &= \frac{m^2}{2m_F} \frac{1}{2} \text{kin. (r)} \quad (65)
 \end{aligned}$$

where v_i is the initial relative velocity of Rg^+ and F^- , and E_v^{kin} is the initial vibrational kinetic energy. The rate constant for this exchange reaction is the rate at which thermal Rg atoms penetrate a sphere of radius r around F^- . Since there is no activation energy, this rate is given by

$$K(r) = \left(\frac{8kT}{\pi\mu} \right)^{1/2} \sigma, \quad (66)$$

where the cross section for a particular internuclear separation, r , is $\sigma = \pi r^2$. The average rate constant, K , is given by the probability that the Rg^+ and F^- ions have a separation, r , multiplied by the rate for that value of r ,

$$\begin{aligned} \langle K \rangle &= \int_0^\infty |\psi_E(r)|^2 K(r) dr \\ &= \left(\frac{8\pi kT}{\mu} \right)^{1/2} \int_0^\infty |\psi_E(r)|^2 r^2 dr, \end{aligned} \quad (67)$$

where $\psi_E(r)$ is the vibrational wave function for energy, E .

Now that the average rate for a quenching collision is expressed in terms of a simple integral, a similar expression can be found for the average energy transferred in a collision, $\langle E_T \rangle$. This is just the rate at which energy is carried off divided by the rate of reaction,

$$\langle E_T \rangle = \frac{\langle K_{E_T} \rangle}{\langle K \rangle}. \quad (68)$$

The average, $\langle K_{E_T} \rangle$, is found from the rate constant for the reaction occurring at the particular internuclear separation, r , multiplied by the average energy carried off at that r , all of which is multiplied by the probability that Rg^+F^- is at the separation, r :

$$\langle K_{E_T} \rangle = \int_0^\infty |\psi_E|^2 \left(1 - \frac{\mu^2}{2m_F} \right) E_v^{\text{kin}}(r) K(r) dr. \quad (69)$$

The term E_v^{kin} can be found from the eigenvalue for the vibrational level, E , and the unperturbed potential energy curve, $V(r)$, for the excited state, $E_v^{\text{kin}} = E - V(r)$. Thus, the following expression is obtained for $\langle K_{E_T} \rangle$:

$$\langle K_{E_T} \rangle = \left(\frac{8\pi kT}{\mu} \right)^{1/2} \int_0^\infty |\psi_E(r)|^2 \left(1 - \frac{\mu^2}{2m_F} \right) [E - V(r)] r^2 dr. \quad (70)$$

Finally, from Eq. (68), the average energy transferred per collision is

$$\langle E_T \rangle = \left(1 - \frac{\mu^2}{2m_F} \right) \frac{\int_0^\infty |\psi_E(r)|^2 (E - V(r)) r^2 dr}{\int_0^\infty |\psi_E(r)|^2 r^2 dr}. \quad (71)$$

The model has been used to calculate the rates of vibrational quenching for various levels of KrF . Figure 7-102 shows three typical vibrational levels and wave functions for the Dunning and Hay (Ref. 129) $2\Sigma^+$ KrF potential curve. The formulas derived above were used to calculate average cross sections, energy transferred per collision, and average rate constants for these three levels, as well as for other representative levels in the well.

In Fig. 7-103, we show the average vibrational energy lost per collision of a thermal Kr atom with a KrF molecule having a total vibrational energy, E . The average rate constant for the charge-exchange collision is plotted in Fig. 7-104. For vibrational levels near the dissociation limit, the diatomic molecule spends most of its time near the outer turning point of its vibrational motion, and the collision rate is found to be large, since the cross section is given by πr^2 . However, relatively little energy is carried off because the velocity of the Kr^+ , to which the electron is transferred, is small. Levels near the bottom of the well also are not rapidly quenched by this mechanism because, again, the Kr^+ is moving slowly. A maximum of 2500 cm^{-1} /collision is found to be removed for collisions with diatomic molecules in levels roughly halfway down the well.

The amount of time required to deexcite a KrF molecule from a level near the dissociation limit to some final total energy, E_f , is given by

$$t_R = \int_{E_i}^{E_f} \left(\frac{dE}{dt} \right)^{-1} dE = - \int_{E_i}^{E_f} \frac{dE}{\langle K_{E_T} \rangle [\text{Kr}]} \quad (72)$$

In Fig. 7-105, the relaxation time, t_R , normalized to the Kr pressure, is plotted as a function of the final-state energy. At a total Kr pressure of 1 atm, the

Fig. 7-102. Three representative vibrational wave functions for the $^2\Sigma^+$ state of KrF. The potential energy curve is displayed as a function of the internuclear separation, r . The wave functions shown are for quantum numbers $v = 10$, $v = 69$, and $v = 142$.

Fig. 7-103. Average energy transferred, $\langle E_T \rangle$, during the Kr + KrF exchange reaction as a function of initial vibrational energy, E , of KrF.

Fig. 7-104. Average rate, $\langle K \rangle$, for the Kr + KrF exchange reaction as a function of initial vibrational energy, E , of KrF.

Fig. 7-105. The final vibrational energy, E_f , of KrF as a function of relaxation time, t_R (normalized by the pressure of Kr)

model predicts quenching to the lowest vibrational levels in a few tens of nanoseconds.

From these results, we conclude that the charge-exchange mechanism is very important in vibrational deexcitation of RGH molecules, particularly through the intermediate levels, where 10 collisions can result in the loss of more than 3 eV. A refined model would include the dependence of the symmetric charge-exchange probability on the separation between the Rg atoms. This inclusion might result in the collision rate being slightly smaller and in the energy carried away per collision being larger.

An important additional consequence of the large exchange rate is that the B, C, and D states will be mixed by this process. The singly occupied p-orbital of the newly created ion will have a distribution in orientations, so that the electronic state can be changed during the collision.

Finally, although the cross sections for asymmetric charge exchange are smaller, they may also contribute significantly to vibrational deexcitation. Particularly for a KrF laser where the buffer gas is Ar, we would expect much of the initial vibrational cooling to proceed down the ArF ladder before asymmetric charge exchange produces KrF. For He

Fig. 7-106. Schematic of vibrational relaxation in KrF for different buffer gases. The potential energy curves for NeF and ArF are shown along with that for KrF. Arrows indicate possible relaxation processes. The displacement reaction $\text{NeF} + \text{Kr} \rightarrow \text{Ne} + \text{KrF}$ produces continuum vibrational states of KrF (denoted by the hatching).

or Ne buffer gases, the HeF and NeF vibrational manifolds lie above the KrF dissociation limit; thus, vibrational quenching by charge exchange will be much less efficient than in the Ar case. This charge-exchange mechanism is illustrated in Fig. 7-106.

Further calculations, using the diatomics-in-molecules method to generate the potential energy surfaces and using classical trajectories, are being made to test the charge-exchange model.

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Ion-Ion Recombination

In electron-beam-pumped RGH lasers, the upper laser level is formed primarily by recombination of positive rare-gas ions with negative halogen ions, according to

where X represents a rare gas (e.g., Ar, Kr, Xe), Y represents a halide (e.g., F, Cl), and M represents any of the rare gases used as buffers in these lasers. At subatmospheric pressures, recombination is a true three-body process. At pressures greater than several atmospheres, however, the reaction becomes a diffusion-controlled process; i.e., the reaction rate is controlled by the rate at which the positive and negative ions drift toward each other, due to their mutual electrostatic interaction, in the presence of the background gas. Hence, the effective two-body reaction rate increases linearly with pressure at low pressures and is some decreasing function of pressure in the diffusion-limited regime. RGH lasers, as envisioned for use in laser fusion, operate in the intermediate-pressure range of 1 to 4 atm.

The theory that we currently use in our kinetic model for calculating ion recombination rates is that of Flannery,¹³¹ which is based on the earlier work of Bates and Flannery,¹³² of Natanson,¹³³ and of Thomson.¹³⁴ Using the latest theory, Flannery and Yang¹³⁵ have calculated some of the recombination rates for the rare-gas fluorides. The effective two-body recombination rate calculated for $Kr^+ + F^- + Ar \rightarrow KrF^* + Ar$ by Flannery and Yang¹³⁴ is shown as line (5) in Fig. 7-107. Wadehra and Bardsley¹³⁶ have made calculations of the formation rates of ArF and KrF using a similar theory. To treat the intermediate-pressure range, all of these theories assume that the effective two-body recombination rate, α (cm³/s), is related to the effective bimolecular (low-pressure) rate, α_t , and to the diffusion-limited (high-pressure) rate, α_d , by

$$\frac{1}{\alpha} = \frac{1}{\alpha_t} + \frac{1}{\alpha_d}. \quad (74)$$

All of these theories also use the concept of critical radius in calculations of the recombination rate. According to this concept, two ions will recombine if one of the ions loses energy by colliding with a neutral particle when this ion is within some critical radius, r_c , of the other ion.

Fig. 7-107. Effective two-body recombination rate for $Kr^+ + F^- + Ar$. Line (1), Bardsley and Wadehra¹³⁷; line (2) $N = 10^{13}$ cm⁻³; line (3) $N = 10^{14}$ cm⁻³; line (4) $N = 10^{15}$ cm⁻³; line (5), Flannery and Yang¹³⁵.

Various points of view have been taken regarding the value of r_c ; consequently, in the various theories, r_c has assumed values in the range of

$$\frac{1}{4} \frac{e^2}{kT} \leq r_c \leq \frac{5}{12} \frac{e^2}{kT}. \quad (75)$$

This range of values has led to a disparity among the recombination rates calculated by using the various theories.¹³⁷ As an added complication, we have inferred from our own experiments¹³⁸ on excited states of ArF and KrF that the rates provided by the Flannery theory are too fast by factors of from two to four.

Because of the importance of ionic recombination in RGH lasers, and the uncertainty in the theoretical results that we currently use, we have undertaken to improve our ability to compute the reaction rates. In high-power lasers, where the charged-particle densities may exceed 10^{15} particles/cm³, we expect that screening of the ion-ion attraction potential by electrons and by other ions may be significant. Screening would have the effect of slowing down the formation rate of the RGH lasing species. Our recombination calculations involve Monte Carlo simulation of the drift of the positive and negative ions toward each other in the presence of the background gas. Instead of a critical radius

for recombination, we are using a critical energy, E_c . If the relative total energy of two ions— $1/2\mu v_r^2 + V(r)$, where μ = reduced mass, v_r = relative speed, and $V(r)$ = interaction potential—is less than E_c when one of them suffers a collision with a neutral atom, the ions are assumed to recombine. We take $E_c = -10kT$ (where k is Boltzmann's constant and T is temperature), although trial calculations have shown that the recombination rate is independent of the critical energy when $E_c \gtrsim -5kT$. This critical energy concept, then, is superior to the critical radius concept. Monte Carlo calculations by Bardsley and Wadehra¹³⁹ have given the recombination rate coefficient shown by curve (1) in Fig. 7-107.

In our extension of the recombination calculations to the regime of high densities of charged particles, we use shielded potentials and the radial-distribution-function concept that is fundamental to the theory of liquids¹⁴⁰ and of one-component plasmas.¹⁴¹ The radial-distribution function, $g(r)$, for two particles is defined such that $\rho g(r)$ is the average density around one particle when the other particle is a distance r away. Then, $\rho g(r) \rightarrow \rho$ as $r \rightarrow \infty$, where ρ is the average particle density in the system. The most simple formulation using a shielded potential is the nonlinear Debye-Huckel theory, which provides analytic forms for both the ion-ion shielded potential and the radial-distribution function. A more accurate approach than the Debye-Huckel theory involves solving the hypernetted chain (HNC) equation for a plasma composed of several charged species and a neutral species. Using shielded potentials and radial-distribution functions, we can formulate the problem to simulate recombination in a laser plasma, rather than just in a neutral background gas, which is assumed by all the other theories.

Preliminary calculations have been performed using the Debye-Huckel shielded potential. For a plasma containing several species of charged particles, the Debye length is given by

$$\lambda_D = \left(4\pi e^2 \sum_j \frac{z_j^2 N_j}{kT_j} \right)^{-1/2} \quad (76)$$

where z_j , N_j , and T_j are, respectively, the charge, number density, and temperature of species j .

Typically, the ion temperature is 300 to 600 K, and the electron temperature is greater than 12 000 K. Hence, the plasma shielding is due primarily to the ions.

Monte Carlo simulations are used to compute the recombination probability, w , for two ions that are initially a distance r_0 apart. The recombination rate is then¹³⁹

$$\alpha = \pi r_0^2 \bar{v} w g(r_0) \quad (77)$$

where $\bar{v}$ is the mean particle speed and $g(r_0)$ is the radial distribution function at r_0 . For a Coulomb potential,¹³⁹

$$g(r_0) = e^{-\frac{e^2/r_0}{kT}} \left(1 + \frac{r_0^2 \bar{v} w}{4eK} e^{-\frac{e^2/r_0}{kT}} \right) \quad (78)$$

where K is the sum of the mobilities of the ions in the background gas. The first term is the well-known Boltzmann factor, and the second term represents a density increase at $r = r_0$ due to the recombination "sink" at $r = 0$ (Ref. 139).

For the Debye potential, $V(r) = (-e^2/r)e^{-r/\lambda_D}$, a similar result can be derived:

$$g(r_0) = \exp\left(\frac{-e^2}{r_0 kT} e^{-r_0/\lambda_D}\right) + \frac{r_0^2 \bar{v} w}{4K kT} \int_{r_0}^{\infty} \exp\left(\frac{-e^2}{rkT} e^{-r/\lambda_D}\right) \frac{dr}{r^2} \quad (79)$$

Trial calculations have shown that, for $\lambda_D \gtrsim r_0$, the recombination probability, w , is approximately the same as that for the Coulomb ($\lambda_D \rightarrow \infty$) potential. For smaller λ_D , we must compute the particle trajectories using the shielded potential. Using the probabilities computed by Bardsley and Wadehra,¹³⁹ we have used Eqs. (77) and (79) to estimate the effects of screening in cases where ion densities are 10^{13} , 10^{14} , and 10^{15} ions/cm³. These densities are typical of laser plasmas and have corresponding Debye lengths of $\lambda_D = 5200 a_0$, $1640 a_0$, and $520 a_0$, respectively, where $a_0 = 0.52 \text{ \AA}$ (Bohr radius). In these calculations, $r_0 = 1000 a_0$. The results are shown as curves (2), (3), and (4) in Fig. 7-107. The plasma shielding effects are evident at an ion density of only 10^{13} ions/cm³. The recombination rate for $N = 10^{15}$ ions/cm³ is expected to be too large because $\lambda_D \ll r_0$ in this case. Additionally, an ion density this large is well out of the range of applicability of the Debye-Huckel theory.

For this and larger ion densities, we must solve the HNC equation to obtain the correct interatomic potential and $g(r)$ and then perform trajectory calculations using this potential.

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Heavy-Particle Quenching Rates for RGH Lasers

Several heavy-particle reaction rates that may be significant for modeling RGH laser systems cannot be measured directly. However, useful upper bounds for these rates can be generated by using the collision-complex model, and we have used this model to determine whether several reactions involving Ar₂F and Kr₂F are important in understanding the kinetics of the laser.

The collision-complex model assumes that the cross section for the process is given by

$$\sigma = \sigma_{cc} P \quad (80)$$

where σ_{cc} is a capture cross section for forming the complex and P is the probability that, once the complex is formed, the particular reaction will take place, i.e., a branching ratio. (A branching ratio is the ratio of the number of products ending up in a particular state to the total number of products

ending up in all states.) Therefore, since $P \leq 1$, an upper bound for the reaction cross section is given by σ_{cc} .

The capture cross section for low-energy (thermal) collisions depends only on the long-range interaction between the colliding species. If the long-range attraction falls off more rapidly than $1/r^4$, there will be a barrier in the effective potential,

$$V_{\text{eff}}(r) = \frac{C}{r^n} + \frac{v(s+1)}{2\mu r^2} \quad (81)$$

for the non-zero impact parameters. The first term on the right-hand side of Eq. (81) is the leading term in the multipole expansion of the intermolecular potential.

For each collision energy, E , a maximum impact parameter exists such that only collisions with $b < b_{\text{max}}$ will experience a close encounter or capture. Thus, we define

$$\sigma_{cc} = \pi b_{\text{max}}^2 = \pi \left[\frac{n^2 C^2}{4E^2} \left(\frac{n}{n-2} \right)^{n-2} \right]^{1/n} \quad (82)$$

The rate constant for close collisions, assuming a Boltzmann distribution of velocities, is

$$k_{cc}(T) = \left(\frac{k}{\pi\mu} \right)^{1/2} \beta^{3/2} \int_0^\infty dl l \sigma_{cc}(l) e^{-\beta l} \quad (83)$$

The quenching reactions of interest will have rate constants that are bounded by k_{cc} .

We have calculated rate constants for several quenching processes of two types:

- "Self-quenching" processes, reactions (4) and (5) of Table 7-12, for which the long-range attraction is dominated by the dipole-dipole interaction.

- Quenching by rare gas atoms or by F₂, for which the dipole-induced-dipole term dominates.

For the first case (self-quenching), the parameters C and n in the effective potential [Eq. (81)] are $D^4/4$ and 6, respectively, corresponding to the angle-averaged Boltzmann-weighted interaction between two permanent dipoles. The similarly averaged attraction for the second case (rare-gas or F₂ quenching) is given by $C = D^2 \alpha$ and $n = 6$. Here, D is the dipole moment of the excited molecule, chosen to be 10 debye for all reactions considered, and α is the polarizability of the quencher. The molecular parameters are given in

Table 7-12. Upper bounds of rate constants for several important quenching reactions.

	Theory (cm ³ /s)	Experimental (cm ³ /s)
1) Ar ₂ F* + Ar → products	4.0 × 10 ⁻¹⁰	1.2 × 10 ⁻¹³
2) Ar ₂ F* + Kr → products	3.6 × 10 ⁻¹⁰	—
3) Ar ₂ F* + F ₂ → products	4.3 × 10 ⁻¹⁰	4 × 10 ⁻¹⁰
4) Ar ₂ F* + Ar ₂ F* → products	3.4 × 10 ⁻⁹	—
5) Kr ₂ F* + Kr ₂ F* → products	2.5 × 10 ⁻⁹	—
Polarizabilities		
$\alpha(\text{Ar}) = 1.63 \times 10^{-24} \text{ cm}^3$		
$\alpha(\text{Kr}) = 2.48 \times 10^{-24} \text{ cm}^3$		
$\alpha(\text{F}_2) = 1.33 \times 10^{-24} \text{ cm}^3$		
Dipole moments		
$D(\text{Ar}_2\text{F}) = D(\text{Kr}_2\text{F}) \approx 10 \text{ debye}$		

Table 7-12. Putting these parameters in Eqs. (82) and (83), we obtain the following expressions for the capture rate constants:

$$k_a(F) = \left(\frac{2\pi}{\mu}\right)^{1/2} (54)^{1/3} \beta^{1/6} \Gamma\left(\frac{4}{3}\right), \quad (84)$$

for the first case; and

$$k_b(F) = \left(\frac{2\pi}{\mu}\right)^{1/2} (54)^{2/3} \alpha^{1/3} \beta^{1/6} \Gamma\left(\frac{5}{3}\right), \quad (85)$$

We note that, for both types of reaction, the temperature dependence of the rate is very weak, but we emphasize that the quenching rate may have quite a different dependence, since the branching probability, P , defined in Eq. (80), can be a function of temperature.

Our analytical results are shown in Table 7-12, along with the molecular parameters used in the calculations, and two available experimental results are also shown in the table. The low quenching rate observed for reaction (1) implies that most close collisions do not produce quenching ($P \ll 1$). The more likely result of such encounters is the displacement of an argon atom,

a reaction that we discuss in connection with vibrational quenching in "Vibrational Quenching of Excited Rare-Gas Halide Molecules by Charge-Exchange Collisions," above.

Although the collision-complex model does not address the problem of predicting the products of reactions, it can produce useful upper bounds for these processes.

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Electron-F₂ Collision Processes

Kinetic studies of electron-beam-excited fluorine-rare-gas mixtures have established that the dominant mechanism for the formation of rare-gas fluoride excimers is ion-ion recombination, in which the negative ions are produced by dissociative attachment of low-energy electrons to F₂. Electronic excitation of F₂ by low-energy-electron impact is also an important process in these systems. In particular, since F₂ can be dissociated by excitation of the lowest ³Π_g state—the calculated vertical excitation energy to this level is only 3.35 eV (Ref. 142)—cross sections for this process are crucial in determining the low-energy-electron distributions and atomic-fluorine concentrations in gas discharges.

The data presently available on electron-F₂ scattering processes are quite limited. Although the results of the most recent experimental studies on dissociative attachment to the lowest vibrational state of F₂ are all in qualitative accord,¹⁴³⁻¹⁴⁵ some discrepancies remain and, as yet, no experimental data have been obtained on attachment to vibrationally excited F₂. Furthermore, no experimental cross sections and only indirect theoretical information¹⁴⁶ are available for the dis-

sociation of F_2 by electron impact. Therefore, we undertook an extensive theoretical study of the electronic structure of F_2 and the behavior of F_2 in collisions with low-energy electrons.

We calculated cross sections for the excitation of the lowest ${}^3\Pi_u$ state of F_2 by electrons, of from 5 to 40 eV, using a form of the distorted-wave approximation.¹⁴⁷ In this approximation, molecular continuum functions are first computed in the field of an appropriate potential, which we took to be the static-exchange potential for elastic $e^- + F_2$ scattering. The distorted waves, evaluated at the appropriate incident and final electron energies, were then combined with the molecular ground and excited target states, respectively, in computing the scattering amplitude for the inelastic transition. This formulation of the inelastic problem has been applied successfully to several transitions in $e^- + He$ scattering¹⁴⁸ and, more recently, to several low-lying states in H_2 (Ref. 149) and N_2 (Ref. 150). In all of these studies, where experimental data are available for comparison, the distorted-wave approximation was found to faithfully reproduce the shapes of the angular distributions and to provide integrated cross sections that fell within a factor of two of experimental data, even close to threshold.

From previous studies of the elastic scattering of electrons by F_2 in the static-exchange approximation,¹⁵¹ we know that there is a broad shape resonance of ${}^2\Sigma_u^+$ symmetry at 2.2 eV. We also know that such a resonance is most probably an artifact of the static-exchange model and that a proper treatment of polarization effects would lead to a more attractive interaction potential and a disappearance of the resonance, since the F_2^- ion is actually bound at $R = 2.68 a_0$, the equilibrium internuclear separation of F_2 (Ref. 152). Since we computed our distorted waves with the elastic static-exchange potential, we expected a significant enhancement in the inelastic cross sections at low energy. This, indeed, turned out to be the case, as shown in Fig. 7-108, where the upper curve is the total cross section obtained in the previously described model.

We next attempted to take some account of target polarization by constructing a static-exchange potential with orbitals taken from a self-consistent-field (SCF) calculation on the F_2^- negative ion. In effect, the target orbitals were allowed to adjust to the presence of an additional electron. The lower curve in Fig. 7-108 corresponds to the total

Fig. 7-108. Cross section for electron-impact dissociation of F_2 by excitation of the lowest ${}^3\Pi_u$ state. Upper curve: distorted-wave approximation with F_2 orbitals. Lower curve: distorted-wave approximation with F_2^- orbitals.

cross section obtained in this model. The artificial low-energy enhancement has clearly disappeared. It is our feeling that the lower curve is the more reliable result at low energy, although a proper treatment of the R -dependence of the problem could build in some resonance structure near threshold, since F_2^- becomes unbound at a value of R slightly less than $2.68 a_0$ (Ref. 152).

The processes of vibrational excitation and dissociative attachment to both ground and excited F_2 molecules are amenable to theoretical analysis, since they can be calculated by using resonant scattering theory. The first step in the study of these processes is to calculate accurate potential energy curves for the ground states of $F_2({}^1\Sigma_g^+)$ and $F_2({}^3\Sigma_g^+)$. As previously mentioned, the molecular negative ion is stable for internuclear separations greater than $1.4 \text{ \AA}$ ($2.68 a_0$); however, the ion autodetaches an electron to form F_2 at small internuclear distances. Indeed, the rate of autodetachment and the energy separation between F_2 and F_2^- are the two critical parameters that determine the magnitudes of the various cross sections and the branching ratio for vibrational excitation and electron attachment in collisions of a low-energy electron with F_2 .

We undertook a careful study of these two parameters as functions of internuclear distance. We found that calculating an accurate value for the

Fig. 7-109. F_2/F_2^- potential-energy curves.

electron affinity of molecular fluorine is a difficult task because we cannot rely on a cancellation of errors when comparing two systems with different numbers of electrons. Our best results to date, shown in Fig. 7-109, indicate that the electron affinity of F_2^- is approximately zero near the equilibrium geometry ($R = 2.68 a_0$) of F_2 . This result is consistent with experimental observations of a very large electron attachment rate near threshold.

Currently under study are the effect of F_2 initial vibrational excitation on the dissociative attachment cross section and the determination of the branching ratio for the decay of the unstable F_2^- ion.

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Advances in Atomic and Molecular Theory

There are often cases where the direct measurement of a basic process related to gas lasers or laser photochemistry is either technically infeasible or prohibitively expensive. In such cases, a mathematical model can provide a useful means for understanding and quantitatively assessing the importance of a particular phenomenon. The ability to construct such a model in a timely fashion depends critically on the existence of accurate supporting theory and computational technology.

Members of the Theoretical Atomic and Molecular Physics (TAMP) group have provided ongoing support in both the theoretical and the computational areas by pursuing fundamental research activities that address a broad range of problems in low-energy atomic and molecular physics. In this section, we summarize key developments that have taken place during 1979 in the areas of atomic and molecular structure, photodissociation, electron-atom and electron-molecule scattering, and resonance theory. We also report developments on the TAMP group's remote computational facility.

Progress in the theoretical field has, in many instances, been the result of fruitful collaborations between members of the TAMP group and outside consultants. The latter are cited in the following pages as major contributors.

Molecular Structure

Several new and important structure codes are being used by the TAMP group to study the potential-energy surfaces of molecular excited states. In particular, a new effective-core-potential (ECP) integral routine was written at the University of Washington, under contract, and has been installed on the CDC 7600 at LLL. The new program

Fig. 7-110. Comparison of potential-energy curves for the ground state of Kr_2 calculated with an effective core potential (ECP) and with the all-electron (AE) calculation of Wadt (Ref. 156).

will allow us to properly treat systems with occupied *f*-orbitals, such as the lanthanides and actinides.

For accurate calculation of diatomic potential curves, we have installed a Slater orbital integral program. This program, DERIC, is being used to recalculate and extend our study of the excited states of rare-gas diatomic molecules.^{153,154} A significant advancement in determination of the ECP for the rare-gas atoms has given us added confidence in our results. We previously were troubled by potential energy curves that were too "soft" when compared to all-electron (AE) calculations. The ECPs were recalculated, using the prescription of Christiansen et al.,¹⁵⁵ and the resultant good agreement with the AE calculations of Wadt¹⁵⁶ for Kr_2 is shown in Fig. 7-110. Exact agreement was not expected, since the ECP calculation employ a superior basis set. Calculations are in progress on Ar_2 , Kr_2 , and Xe_2 .

AE calculations were carried out with our existing codes to determine the interaction potential for two nitrogen molecules.¹⁵⁷ The intermolecular

potential, Φ , plays an important role in interpreting N_2 data at high pressures. Seven different orientations were studied, and a large degree of anisotropy was discovered. Definitions of the seven geometries for the N_2 - N_2 interactions are shown in Fig. 7-111, and the calculated energies are plotted in Fig. 7-112. The linear geometry I. suffers a curve crossing at very close distances and exhibits a break in the potential curve at 4 to 5 Bohr radii (a_0). The nature of this curve crossing and of the curves for excited states of N_4 are now under study.

Calculations on triatomic molecules, such as COS (carbon oxysulfide), CO_2 , CS_2 , and N_2O , are also continuing. Our progress on COS was discussed in the 1977 annual report.¹⁵³ A problem common to each of these molecules is the influence of polarization functions on the excitation energies and, of course, on the potential curves for the excited states. Our work on the photodissociation of these molecules requires accurate excited-state potential surfaces. A comparison of the excitation energies for a basis set with and without 3d-polarization functions shows a shift of 0.8 eV for the valence states. The lowest Rydberg state is shifted much less. This has led to some confusion in the literature concerning assignment of the optical and electron-impact spectra.^{158,159}

To understand the reason for this shift in energy, we are calculating the potential surfaces of CO_2 in a good basis set that includes polarization functions. While the bending curves are crucial for interpreting the spectra, we have only completed the symmetric-stretch curves at present. These curves are shown in Fig. 7-113. The $^1\Pi_g$, $^1\Pi_u$, and $^1\Sigma_u^+$ states have Rydberg character, while the rest are valence states; the position of the minima are consistent with this situation. Correlation errors between states are not accounted for in these curves, so the excitation energies are too large. It is the $^1\Pi_g$ and $^1\Delta_u$ states that have been assigned incorrectly in the literature.

CI curves are being calculated for both the symmetric-stretch and the bending coordinates. Once consistent excitation energies are obtained for CO_2 , we should be able to understand the remaining triatomic molecules.

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Fig. 7-111. Definitions of seven geometries employed in the calculations of interaction potentials for two N_2 molecules at distance r . The angle ϕ is measured between the two N_2 molecules after being projected onto a single plane. The first and second molecules make the angles θ_1 and θ_2 , respectively, with this projection plane.

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Theory of Photodissociation of Small Molecules

The photolysis of small molecules can yield internally excited fragments that can initiate chemical reactions, radiate light and other electromagnetic radiation, or emit electrons. Detailed understanding of these processes is particularly important in developing gas lasers.

We have written a computer code with which we can calculate, as functions of incident photon

Fig. 7-112. Interaction potentials calculated *ab initio* for the seven orientations of two N_2 molecules that are depicted in Fig. 7-111. The [4s3pld] s.c.f. was used for the calculation of Φ .

energy, photodissociation cross sections and final-state distribution probabilities for linear triatomic molecules. The program requires as input the equilibrium geometry and vibrational-force constants of the initial state and the potential-energy surface for the final dissociating state.

The cross section for photodissociation of a molecule is proportional to the square of a matrix element that involves overlap of the initial bound and the final scattering wave functions for nuclear motion—provided the transition strength is constant in the Franck–Condon region, which is generally the case. The initial-state wave function can be calculated in the harmonic approximation by using spectroscopically determined molecular geometry and force constants. We calculate the final scattering state with outgoing boundary conditions by using the R-matrix propagation technique developed by Light and Walker¹⁶⁰ for molecular collision processes. This technique solves the scattering problem by generating piecewise analytic wave functions whose logarithmic derivatives (R-matrix) are propagated from the collision region to the various asymptotic final arrangements. We modified this one-dimensional code¹⁶¹ so that we can calculate overlaps between the scattering and initial-state wave functions. These overlaps are propagated along with the R-matrix. In the

Fig. 7-113. Symmetric-stretch potential-energy curves for the ground and excited states of CO_2 .

asymptotic region, we apply proper boundary conditions, which normalize the overlap integrals. Thus, we obtain partial and total photodissociation probabilities that are proportional to the cross sections.

We have used this program to model a CO_2 system. The initial vibrational wave functions were generated by using spectroscopic data from Herzberg.¹⁶² The upper state dissociates to CO in its ground electronic state and is generated by using a LEPS-type functional that has a 1.27-eV barrier to reaction. The resultant surface is shown in Fig. 7-114. The asymptotic limit of $CO + O$ corresponds to -11.24 eV. The equilibrium internuclear separation, R_{CO} , in the ground state is 2.20 a_0 . We calculated dissociation probabilities from the ground state (Fig. 7-115) and from a state with one quantum of excitation in the asymmetric stretch (Fig. 7-116). We have plotted, in addition to the total probabilities (upper curves), the partial probabilities for populating the lowest three vibrational levels ($v_f = 0, 1, 2$) of the CO fragment as a function of photon energy above the energetic dissociation threshold.

The structure in Fig. 7-115 is due to excitation of different vibrational levels of the final-state symmetric stretch in the Franck–Condon region. The peaks can be shown to coincide with the eigenvalues of the upper-state symmetric stretch. We note that the partial probabilities do not peak precisely at the

Fig. 7-114. Excited-state potential-energy surface for collinear CO_2 .

same photon energies; in fact, the energy dependence is remarkably different for different final levels.

The symmetric triatomic system is interesting in that the degrees of freedom are approximately separable within the Franck-Condon region. Using this fact, Pack¹⁶³ and, later, Heller¹⁶⁴ postulated that the observed structure in the absorption spectra could be explained in terms of these quasi-separable degrees of freedom. Our calculations show that regions of the upper surface away from the Franck-Condon region can also be very important,

since the final-state interactions result in very dissimilar partial cross sections. The total cross section, then, could be more sensitive than had been expected to characteristics of the whole upper surface. We find (Fig. 7-116) that putting one quantum of energy in the asymmetric mode of the initial state completely washes out the vibrational structure. The extra energy in the dissociating mode eliminates the constructive interference seen in the ground-state spectrum.

Our computer code will be used to study the CO_2 system in more detail so that we can better understand what is a notoriously confusing spectrum.

Fig. 7-115. Total or partial probabilities for photodissociation of ground-state CO_2 , each shown as a function of excess photon energy above the energetic dissociation threshold. Each of the lower three curves shows the probability of populating a single fragment state (partial cross section); the top curve (the total cross section) is the sum over the partial cross sections—i.e., the sum over all possible final states.

Modeling of excited-state surfaces must progress further before the analytical techniques described above can be applied to those molecules that are important to our laser systems.

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Fig. 7-116. Total or partial probabilities for photodissociation of CO_2 with one quantum of energy in the asymmetric stretch mode. The curves are defined as in Fig. 7-115.

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Electron Scattering Processes

Cross sections and rate constants for electron-atom and electron-molecule collision processes are fundamental to the modeling of energy flows in electron-beam-pumped and discharge-pumped

laser systems. The TAMP group has, therefore, made a substantial effort to both develop and acquire the improved computational technology necessary for making quantitative predictions about a wide variety of atomic and molecular systems in different energy regions.

The low-energy region, roughly characterized by impact energies less than the ionization potential, requires a rigorous quantum mechanical treatment that takes proper account of electron-exchange and polarization effects. The numerical close-coupling technique is the most reliable tool for studying electron-atom collisions in this region; consequently, we have acquired the atomic-close-coupling code, IMPACT,¹⁶⁵ developed at University College London, and have installed this code on our CRAY computer. IMPACT has been used to generate cross sections for the electron-impact deexcitation of the ³⁵S level of atomic sulfur, and these results are presented in "Group VI Lasers," above.

In the case of low-energy electron-molecule collisions, the formulation of practical solution techniques is still an active area of research, and no standard codes are generally available. We have, therefore, continued with our work in this area and have helped to develop, in collaboration with others, a variational approach that relies both on numerical and analytic (bound-state) techniques for implementation.

At high energies (> 100 eV), the Born approximation is generally valid. The Born code that we have developed is both efficient and flexible; since it is interfaced with our molecular-structure codes, this Born code can provide generalized oscillator strengths¹⁶⁶ that incorporate complex CI wave functions.

The intermediate energy range (from about the ionization potential to about 100 eV) is perhaps the most difficult area for numerical work because of the large number of electronically accessible states and the characteristically rapid oscillation of the continuum wave functions. A significant advance in this area was our extension of the impact-parameter technique¹⁶⁷ to treat electron-impact excitation of optically allowed molecular states. We have also developed distorted-wave techniques¹⁶⁸ that are particularly useful for studying transitions at intermediate impact energies between optically forbidden states. Our theoretical studies of electron-impact dissociation of molecular fluorine (discussed in "Electron-F₂ Collision Processes," above) were

carried out by using a distorted-wave approximation.

In the course of our work on low-energy electron-molecule collisions, we found that the Schwinger variational principle offers a number of distinct advantages over other stationary expressions.¹⁶⁹ Most notably, the Schwinger expression for the scattering amplitude is independent of the normalization employed in the trial wave function. This feature is particularly attractive for studying electron-molecule scattering, where discrete basis functions can be used advantageously in solving the collision problem. Several square integrable (1,2) techniques have been introduced, but they all seem to require large numbers of basis functions to achieve adequately converged results.^{170,171} Furthermore, the Schwinger principle does not suffer from the spurious singularities that arise in algebraic variational techniques of the Kohn type,¹⁷² since the principle depends on the fact that the exact wave function satisfies the Lippmann-Schwinger integral equation. Indeed, it can be shown that using a trial function (ψ_t) with only linear parameters in the Schwinger variational principle,

$$\psi^S(r) = \sum_{i=1}^N c_i \phi_i(r), \quad (87)$$

is equivalent to solving the Lippmann-Schwinger equation (for the scattering amplitude) exactly for the separable potential of the form¹⁷³

$$V^S(r, r') = \sum_{i,j=1}^N \langle r | V | \phi_i \rangle \langle \phi_j | V | r' \rangle \underline{Y}_{ij}^{-1} \quad (88)$$

In Eq. (88), $\underline{Y}$ is the $N \times N$ matrix representation of the potential in the basis ϕ_i . Thus, for the scattering amplitude, the Schwinger principle gives a stationary expression that is the exact solution for a potential of finite rank and, hence, does not show any singularities at physical energies.

The Schwinger variational expression for the T-matrix can be written, in atomic units, as

$$\langle k' | T^S | k \rangle = \frac{\langle k' | U | \underline{\psi}_k^+ \rangle \langle \underline{\psi}_k^- | U | k \rangle}{\langle \underline{\psi}_k^- | U - U G_0^+ U | \underline{\psi}_k^+ \rangle} \quad (89)$$

where G_0^+ is the free-particle Green's function for the outgoing (incoming) wave boundary condition,

$k^{(\pm)}$ is a corresponding trial scattering wave function, and $U = 2V$, with V being the effective interaction between the target and the incident electron. We have applied the Schwinger principle to $e^- + H_2$ scattering, using trial wave functions expanded in a finite set of Cartesian-Gaussian functions.¹⁷⁴ With such a choice of functions, we were able to develop efficient analytical approximation schemes for evaluating all of the required matrix elements. Using basis sets of modest size (5 to 10 functions), we were able to obtain results for $e^- + H_2$ scattering, in the static-exchange approximation,¹⁷⁴ that agree well with the results of an exact numerical integration of the corresponding coupled integrodifferential equations.¹⁷⁵

We have also used efficient numerical schemes for evaluating the Schwinger variational expression with an arbitrary trial wave function. In this connection, the Schwinger principle can be iterated to converge to an exact result.¹⁷⁶ We are currently using this approach to study low-energy electron scattering by strongly polar diatomic molecules.¹⁷⁷

Until quite recently, the Born approximation was the only practical technique for calculating cross sections for electronically inelastic electron-molecule collisions. Most previous applications of this technique used rather simple descriptions of the molecular excited states, even though it is well known that configuration interaction among excited states may profoundly influence calculated optical oscillator strengths and, presumably, electron collision strengths. To remedy this oversimplification, we have developed an efficient and flexible Born code that is completely interfaced with our molecular-structure codes. Consequently, we have the capability to use very large *ab initio* CI wave functions to describe molecular excited states in electron-scattering calculations. For example, in our study of electron quenching processes in rare-gas halide lasers,¹⁷⁸ we used the Born code to obtain generalized oscillator strengths and integrated cross sections for the ionic, 2Σ , excited states of KrF and XeF—two complex molecules, each of which requires over 1000 electron configurations to accurately describe its electronic states.

From previous work on electron-atom collisions, it is well known that the Born approximation overestimates the integrated cross sections for optically allowed transitions at low impact energies (less than 50 eV). Unfortunately, this is precisely the energy region that is most important for modeling

electronically excited gas lasers. Consequently, we have extended the semiclassical impact-parameter technique¹⁶⁷ to include electron-molecule collisions and have derived a modified expression for the cross section that automatically satisfies reciprocity.

For molecules, the Born-Oppenheimer separation of electronic and rotational-vibrational motions is employed. The cross section for exciting (or deexciting) a given electronic state is obtained by averaging the purely electronic cross section over all possible orientations of the molecule. The molecular target electrons are treated quantum mechanically, while the scattered electron is treated classically, using straight-line trajectories. The molecular-transition probability is calculated by using first-order, time-dependent perturbation theory. Exchange between the target electrons and the projectile is ignored. This technique is only applicable to spin-allowed transitions for which long-range electric multipole potentials dominate the electron-molecule interaction. A requirement that energy and angular momentum be conserved during collision leads to expressions for excitation and deexcitation cross sections that satisfy reciprocity.

At high impact energies, the modified cross section formulas reduce to previous results.^{167,179} To avoid overestimating the partial cross sections for small impact parameters, a lower cutoff in the impact parameter is employed. The value of this cutoff should be roughly equal to the radius of the target; a precise cutoff value can be determined by normalizing to the Born approximation at high energy, where the impact-parameter formula and the Born approximation have the same limiting behavior. The use of such a cutoff is justified¹⁶⁷ for optically allowed transitions where long-range forces dominate and where large impact parameters contribute a significant fraction of the total cross section.

We successfully applied the modified impact-parameter technique to several optically allowed transitions in H_2 , F_2 , N_2 (Ref. 180), KrF, and XeF (Ref. 178). Figure 7-117 compares our cross sections, computed with the impact-parameter technique, with those obtained with other techniques for the

transition in H_2 . It is clear that our technique yielded values for cross sections that were within 25% of those calculated with the best available techniques,^{181,182} even at energies within several

Fig. 7-117. Cross sections for electron-impact excitation of $X^1\Sigma_g^+ \rightarrow B^1\Sigma_u^+$ transition in H_2 ; (---) impact-parameter technique; (—) modified impact-parameter technique; (O) Born approximation (Ref. 181); (□) two-state close-coupling technique (Ref. 181); (△) distorted-wave approximation (Ref. 182).

Fig. 7-118. Cross sections for electron-impact excitation of $X^1\Sigma_g^+ \rightarrow c^1\Sigma_u^+$ transition in N_2 . Present results: (—) impact-parameter technique with CI wave function; (●) Born approximation with CI wave function. Previous results: (○) Born approximation with single-configuration wave function (Ref. 183); (□) experimental results of Chutjian et al. (Ref. 184); (△) experimental result of Aarts and de Heer (Ref. 185).

electron volts of the excitation threshold. Figure 7-118 compares our cross sections for the

transition in N_2 with previous theoretical and experimental results. In this case, we found that inclusion of configuration mixing in the description of the N_2 excited states was essential for obtaining accurate cross sections.

The examples cited above demonstrate the utility and flexibility of both our Born scattering code and the modified impact-parameter technique.

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Advances in the Theory of Molecular-Resonance Phenomena

Resonant autoionizing states of molecules and autodetaching states of molecular negative ions play crucial roles in many processes involving low-energy electrons, e.g., dissociative attachment, dissociative recombination, and Penning ionization.¹⁸⁶ In electrically excited lasers, such processes often

constitute important pathways to forming the upper laser level. For example, in rare-gas fluoride lasers, the dissociative attachment of electrons to F_2 is the major mechanism for forming F^- ions.¹⁸⁷ Thus, accurate cross sections for various resonant processes are required for the kinetic modeling of discharge-pumped or electron-beam-pumped gas lasers.

During the past few years, we have taken an active role in developing two efficient and practical techniques for the theoretical study of molecular resonances: the Stieltjes imaging technique for calculating the decay lifetimes of autoionizing states^{188,189} and the complex-basis-function technique^{190,191} for treating resonance states of many electron systems. During 1979, we made significant progress with both of these techniques.

We used electronic-resonance parameters, determined with the Stieltjes technique, to compute cross sections for the resonant vibrational excitation of N_2 by low-energy electrons. Also, we developed an iterative SCF procedure that uses complex basis functions to compute energy and decay width of shape resonances, and we applied this procedure to the $2p^\circ$ state of Be^+ . The complex-basis-function technique was also extended to treat the direct photoionization of simple molecules. Numerical results obtained for the photoionization of H_2^+ suggest that further investigations of this technique will be worthwhile.

It is well known that the vibrational excitation of small molecules by low-energy electrons is dominated by negative-ion shape resonances. One of the widely used techniques for calculating vibrational-excitation cross sections is the 300-MERANG model,¹⁹² in which the motions of the nuclei are described by a complex potential. The real and the imaginary parts of this potential are the electronic energy and the decay width, respectively, of the negative-ion resonance. Most previous applications¹⁸⁶ of this technique involved semiempirical procedures, in which the two resonance parameters were determined by fitting the calculated cross sections to experimental results.

We calculated the vibrational-excitation cross sections for N_2 by using this complex-potential model with resonance parameters obtained from *ab initio* studies of the $2\Pi_g$ resonance of N_2^- . For this particular study, our two main objectives were to:

- Test the validity of approximations, implicit in the complex-potential model, for describing nuclear motion.

- Test the dependence of the computed cross sections on the *ab initio* resonance parameters used in the model.

We employed three different sets of resonance parameters in our study. For the first set of parameters, we extracted the resonance energy and resonance width of $N_2(2\Pi_g)$ from the R-matrix calculations of Schneider and co-workers.¹⁹³ We determined the second set of parameters by using the Stieltjes imaging technique, along with a CI description of N_2 . We took the third set of resonance parameters from previous work,¹⁸⁹ in which polarization of the N_2 target during collision was neglected.

Our results show that inclusion of target polarization in the calculation is critical if realistic vibrational-excitation cross sections are to be obtained. The $v = 0 \rightarrow v = 1$ and the $v = 0 \rightarrow v = 3$ excitation cross sections obtained from our present work are compared in Fig. 7-119 with cross sections obtained from previous R-matrix calculations.¹⁹³ As may be seen, the structures of the cross sections compare very favorably for the two computational techniques. However, because the cross section structure is a function of the vibrational levels of the N_2 compound state, we found the fine structure to be sensitive to the N_2 potential-energy curve used in the calculations. Figure 7-120 compares our total cross section for the ground vibrational state of N_2 with the experimental results of Kennerly and Bonham.¹⁹⁴ We obtained our theoretical cross section as a sum over all final vibrational levels, using an elastic ($v = 0 \rightarrow v = 0$) cross section appropriately modified with the correct energy dependence near the excitation threshold. The agreement with experiment is reasonable.

Our work shows that the complex-potential model, used with *ab initio* electronic-resonance parameters, can accurately provide resonant vibrational-excitation cross sections. For the case of N_2 , the approximations underlying this model are certainly valid, and our description of the $2\Pi_g$ negative-ion resonance appears to be correct.

The complex-basis-function technique is a mathematical device that enables us to treat autoionizing states of atoms and molecules simply as bound states.¹⁹⁵ In its original form, the technique proceeds by constructing an analytical continuation of the Hamiltonian operator through a transformation of the coordinates of each electron

Fig. 7-119. Comparison of cross sections for vibrational excitation from the $v = 0$ level of N_2 by electron impact: (---) present calculations using the complex-potential, or BOOMERANG, technique; (—) R-matrix results of Schneider, Le Dourneuf, and Vo Ky Lan (Ref. 193).

Fig. 7-120. Comparison of experimental and theoretical cross sections for electron scattering from the ground vibrational state of N_2 : (---) experimental data of Kennerly and Bonham (Ref. 194); (—) current theoretical result from the complex-potential model.

according to $r \rightarrow re^{i\theta}$. This nonhermitian operator can have complex eigenvalues and normalizable eigenvectors corresponding to autoionizing states. The fact that the resonance-state vectors can be made normalizable by this technique has enabled investigators to assume standard bound-state CI conditions in implementing the technique. The original technique has been modified in practice to make tractable the calculations on multielectron systems.¹⁹⁰ Even with these modifications, however, CI expansions are often found to converge slowly in resonance calculations, and the required basis sets typically are larger than those customarily used in bound-state applications. The problem hinges on the lack of suitable procedures for generating basis sets that can adequately describe complex resonance states.

The Hartree-Fock SCF technique is frequently employed in bound-state applications to generate a set of occupied and virtual orbitals that can then be used in further CI studies. We are currently experimenting with a similar strategy in the complex-coordinate procedure. By formulating SCF equa-

Fig. 7-121. Complex energy of the $(1s^2 2s^2 2p) \ ^2P^\circ$ state of Be⁺ as a function of rotation angle, θ , in radians. Lower curve refers to a calculation in which all orbitals are scaled; upper curve refers to a calculation in which only the outer 2p orbital is expanded in a complex basis. (Note: 1 hartree = 4.360×10^{-18} J.)

tions in the context of the complex-coordinate approach, we have found interesting cases (shape resonances) where the generalized equations can be made to converge for a metastable state, whereas the usual SCF equations would collapse to a lower state of the same symmetry. Figure 7-121 shows the behavior of the complex SCF energy, as a function of the rotation angle, θ , for the autodetaching $^2P^\circ$ state of Be⁺. The main curve in Fig. 7-121 was obtained from a complex-coordinate calculation, in which all the orbitals are rotated (standard procedure). The more slowly varying curve, above the main curve, was obtained by using a procedure that we developed,¹⁹⁰ in which the tightly bound core orbitals are left unrotated and only the resonance 2p orbital is expanded in terms of complex-scaled basis functions. Since exact calculations carried out on a complete basis would give a

Fig. 7-122. Photoionization cross section of H_2^+ for polarization parallel to the molecular axis. The superposed curves are for calculations in which the rotation angle is varied from 15° to 25° in 2.5° increments. Open squares are the exact results. (From Ref. 197.)

result independent of the rotation angle, θ , our modified procedure is clearly seen to represent a dramatic improvement over the original complex-coordinate procedure.

We have also extended the complex-basis-function technique to compute direct photoionization of simple molecules.¹⁹⁶ We make use of the fact that the photoionization cross section can be expressed as an expectation value of the resolvent over the square-integrable function

$$\lim_{\epsilon \rightarrow 0} e^{-i\epsilon} \sigma(\omega) = \frac{4\pi\omega}{c} \left(\psi_0 \left| \mu \frac{1}{H - I_0 - \omega - i\epsilon} \mu \right| \psi_0 \right), \quad (90)$$

where ψ_0 is the initial state of the system, with energy E_0 ; μ is the dipole operator; and ω is the photon frequency. The use of complex basis functions allows us to obtain meaningful approximations to the right-hand section of Eq. (90), at real photon energies, by inverting the finite-basis-set representation of the Hamiltonian operator. Figure 7-122 shows the photoionization cross section of H_2^+ that we calculated by using the

complex-basis-function technique. Further applications of this new technique to molecular systems more complex than H_2 will be the subject of future work.

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TAMP Remote Computational Facility

The TAMP group is responsible for providing timely theoretical support, expertise on and awareness of current advances in theory, and state-of-the-art computational methods and facilities for analysis of laser-related problems. One of the facilities provided by the TAMP group is the remote computational facility (RCF). The RCF was designed to be used for problems involving dynamic

Fig. 7-123. Schematic of the TAMP group's remote computing facility (RCF).

calculations made with classical trajectory techniques. These problems are typified by repetitive floating-point calculations that require little input/output activity. The RCF provides a machine that not only is dedicated for these problems but also is connected to the Octopus network for data transport.

During 1979, the RCF was made available for production calculations; consequently, it has been used essentially full time, and several areas of ex-

pansion are planned for 1980. The two major projects for which the RCF has been used are the classical trajectory study of chemical reactions relating to isotope separation (see "The Reaction of Vibrationally Excited HBr and I," below) and Monte Carlo glass simulations (see "Monte Carlo Glass Simulations," above).

A diagram of the RCF is shown in Fig. 7-123. The main computer is a PDP 11-55, which has an attached AP-120B array processor. The classical trajectory calculations were run only on the PDP 11-55 at a speed 10 to 15 times slower than on a CDC 7600. On the other hand, the Monte Carlo glass-simulation calculations involved heavy use of the AP-120B and were made at speeds within a factor of two of the CDC 7600. The computing speeds of several popular mainframes are compared with that of the TAMP group's PDP 11-55 in Fig. 7-124.

During 1980, we plan to greatly enhance the RCF's disk-storage capability so that a greater variety of projects can make use of the facility. The time-sharing capability will be increased by adding another partition in memory to the PDP 11-55. We have ordered new additions to our software library for the AP-120B, and we will install VERSATEC graphics routines on the PDP 11-55.

Two new projects that are expected to begin using the RCF in 1980 involve molecular-structure calculations (the first to be made on the RCF) and the development of a classical photodissociation program for studying polyatomic molecules.

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Fig. 7-124. Computing speeds of several popular mainframes compared with the TAMP group's RCF.

Energy and Military Applications

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Energy and Military Applications

8

Introduction

Before we can use laser-fusion energy as a primary power source, we must bridge the gap between the ICF experimental program and such eventual applications as the generation of electric power or the testing of the vulnerability of military hardware to x rays and neutrons. In a program dominated by complex physics experiments, it is easy to lose sight of the final applications of research. Likewise, users of energy such as the utilities often fail to understand or appreciate the technology requirements imposed by the constraints of physics. We must understand both the current physics basis of ICF and the engineering required to realize a potential application. The former is assured by our intimate connection with experimental work at LLNL. The latter is accomplished by our cooperative work with experts from industry and universities to conceptually design future power plants or test

facilities. These design studies are our most useful tool for early detection of potential problems, and for seizing opportunities brought forward by innovation that can dramatically reduce development time.

Finally, we work with industry to plan the required technology development programs and strategies which will use our nation's resources most effectively.

In 1978 we described the conceptual design of a 1000-MW_e electricity generating station based on the HYLIFE chamber.

In this section, we review our progress in the conceptual design of subsystems of the HYLIFE electric power plant, and present an economic-tradeoff analysis for integrating these subsystems into a complete plant.

HYLIFE is a particularly attractive concept for a Liquid-Metal-Wall (LMW) reaction chamber, which will convert fusion radiation into heat without suffering excessive wall damage, promising a 30-yr life without wall replacement (see Fig. 8-1).

Fig. 8-1

Many areas of analysis identified as having the greatest uncertainty are treated in more detail in this report.

We first describe the advantages and disadvantages of the HYLIFE system, emphasizing the direction we should take in future research and development to assure that the positive features are realized and the negative ones minimized. To gain perspective, we review the conceptual designs of liquid-metal-protected fusion-reactors of various groups published in the last decade. We then present a set of guidelines for 1-MW-reactor design that attempt to codify what we have learned into a coherent strategy for conceptual design.

Next we present new or revised analyses of the fluid mechanical and structural performance of the HYLIFE reactor system, including the effect of vibrations on the stability of jets; the response of the fluid wall to the energy deposition of x-rays, debris, and neutrons; and the subsequent thermal and mechanical response of the wall. We then describe the evolution of the HYLIFE configuration, showing the simplifications we have introduced in the pressure vessel, nozzle plate, and lithium-circulation system.

We emphasize the flexibility inherent in the HYLIFE approach to reactor design. Because the vapor pressure of lithium and lead-lithium mixtures at 500°C spans the range of 10^{-5} to 10^{-2} Torr, both laser and heavy-ion-beam drivers can be accommodated. Also, either mechanical or electromagnetic liquid-metal pumps may be used with HYLIFE. The recirculated power and total system efficiency are substantially affected by the use of different drivers, pumps, and operating temperatures; we also explore these relationships.

As a new technology matures from the physics to the engineering stage, materials problems often start to dominate development times and influence engineering decisions. Even if we are successful in minimizing the need for new materials, the uncertainties of using common materials in new regimes in a nuclear environment demand that we pay early attention to the properties and compatibilities of low-activity structural materials and heat-transfer fluids. Accordingly, we present results of our experimental and analytical investigation into the equation of state of lithium and the properties of graphite, Pb-Li alloys, and 2-1/4 Cr-1 Mo steel. We also give a development scenario for ASME code qualification for a ferritic steel carrying lithium or lead in a nuclear environment.

In 1979 we conducted parametric economic studies to help define the operating parameters of a power plant. It is inadequate to arbitrarily set a requirement for recirculated-power fraction or fusion-energy gain (product of pellet gain and driver efficiency), when the costs and input energy requirements for different types of drivers and reaction-chamber concepts vary so dramatically. Furthermore, we need a consistent methodology for combining the driver, reactor, and pellet manufacturing systems, when each system has better performance or lower cost in different (and often mutually exclusive) operating regimes of yield, pulse energy, or repetition rate. In our economic analysis, we use the minimum cost of electricity to choose a consistent operating regime, which leads to some valuable conclusions about configuring an ICF system in spite of uncertainties in cost estimates.

Finally, we also expanded our conceptual design to include a short-wavelength laser system for fusion. Together with the Advanced Quantum Electronics Group and an industrial team from Bechtel National, Hughes Aircraft Company, and Physics International, Inc., we arrived at a design for a KrF-driven, stacker/compressor laser that will be extremely valuable in guiding our search for high-leverage areas to reduce cost, complexity, and technology development time. We used near term technology for a 1.5-MJ, 2-Hz, 10^7 -shot life design suitable for an engineering test facility. With additional development and capital cost to achieve greater life, this modular laser facility could be expanded to the 3.0-MJ-size characteristic of a full-scale fusion-power-plant driver.

As we demonstrate ignition of ICF targets in the mid-80's, and progress toward the high gains required for commercial power, the engineering challenge will become clearer and stronger. The analyses we perform now will allow us to create a strategy and to plan the critical proof-of-concept experiments required to proceed with confidence on the engineering development path that will ensure the realization of fusion power.

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A Conceptual Design Strategy for Liquid-Metal-Wall Inertial Fusion Reactors

Introduction

As we approach demonstration of the feasibility of igniting high-gain inertial-fusion targets, we must look ahead to the technology that will be required to bridge the gap between an understanding of the fusion physics and the operation of a fusion power plant. Design concepts of future reaction chambers and power plants are key elements in helping us anticipate and resolve problems in fusion power-plant technology. These concepts are our most valuable tools for assessing the technical feasibility, and economic and social costs of fusion power.

In this set of articles, we describe a particularly promising design concept for a pulsed-fusion energy-conversion chamber—the HYFIFE system. The dominant feature of this concept is the thick, free-flowing blanket of liquid metal used to protect the structure of the inertial-fusion reactor. An example of the liquid-metal-wall (LMW) concept is shown in Fig. 8-2. After several years of cooperative study with our colleagues in industry and in universities we have begun to distill our understanding of this concept into a design strategy that will allow the fusion engineering community to find the path of minimum development effort.

The most striking feature of inertial fusion, in terms of its impact on reactor design, is that the confinement of the fusion plasma is decoupled from the functions of the reaction-chamber wall and structure. Because the physics of energy absorption, implosion, and ignition of the plasma are physically separable from the chamber conditions and structural requirements, inertial fusion gives us an extraordinary degree of flexibility in comparison with the stringent demands placed on wall materials by

magnetically confined plasmas. Although propagation of driver energy (either laser or ion beams) through the chamber to the target still depends on chamber pressure, and the design of the first wall still depends on the nature of the radiation from the fusion pellet, these constraints can be readily accommodated.

In inertial fusion, fluids (gases or liquids) can be placed inside the reaction chamber to protect the wall from ablative-material loss caused by energy deposition of short-range-fusion radiation (25 to 35% in soft x rays and debris) and, in some cases, radiation damage caused by deeply penetrating x rays and neutrons. However, the use of protective fluids combined with the pulsed-energy release leads to engineering constraints and techniques that are radically different from those in magnetic fusion. In inertial fusion the understanding of the dynamic response of fluids and structures becomes more important with far less emphasis placed on development of tougher new materials and on high-thermal performance of cooled-blanket modules.

Wall life can be extended by using either gases or liquids in the fusion chamber. Gases are attractive because they can be pumped in and out of the chamber quickly enough to achieve a high repetition rate (up to 5 Hz) and they require relatively little pumping power. However, gases are effective only in protecting the walls against the minor fraction of fusion radiation in x rays and debris; the neutrons penetrate the wall unaffected. The main principle of the gas-filled chamber¹ is to have the gas absorb the short-duration pulse of x rays and debris, and then to reradiate the energy to the walls over a sufficiently long time that the wall ablation is prevented or minimized. Unfortunately, indications are that rare gases, at densities that will allow propagation of the laser beam, turn transparent at high temperatures so rapidly that there is little advantage.² Apparently, higher pressures are required than laser propagation will allow. At higher

Fig. 8-2. Elevation cross section of HYLIFE chamber.

densities favorable to propagation of channeled light-ion beams,³ the gas-filled chamber appears to be promising. The upper-density limit then becomes the impulsive loading on the wall caused by a fireball-driven shock wave.

On the other hand, liquids will not only absorb x rays and debris, they will also attenuate and absorb the major energy-carrying component—the neutrons. Because most of the fusion energy is absorbed by the liquid metal, separate heat-transfer systems for the chamber gas and blanket coolant are not required. Of the liquid metals, lithium, lead, and lead-lithium solutions are the most attractive.

LMW chambers can span the chamber pressure range of 10^{-5} to 10^{-1} Torr because of the low vapor pressure at temperatures between 375

and 500°C, which leads to an environment suitable for propagation of either laser or heavy-ion beams. The LMW flow pattern can be arranged to cover almost the entire area of the chamber wall, leaving apertures just large enough to allow entry of the pellet and driver beams.

The LMW design can be made insensitive to the energy spectra of, and the partitioning of energy between, x rays, ions, and neutrons. This is an attractive feature of the design, because we know little about the characteristics of a reactor-size pellet at this current, early stage of inertial fusion.

The main advantage of liquid-filled chambers over gas-filled chambers, however, stems from the high attenuation (up to 95%) of the neutron energy before the neutrons reach the solid structure. The high attenuation leads to an order-of-magnitude increase in wall life and a corresponding reduction in

mass of activated steel, compared with either gas-filled inertial-fusion reactors or any magnetic-fusion reactor. In this set of articles, we explore these attractive features of LMW chambers in more detail, discuss the difficulties that arise in their design, and review some conceptual designs that have been proposed to deal with the technical issues we identify. Finally, we summarize this discussion by stating what has been learned in a set of guidelines for LMW reactor design.

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Positive and Negative Features of LMW Chambers

We can build a solid foundation for a design strategy by listing the attractive and unattractive features of LMW chambers, and then searching for a conceptual design approach and physical configuration that will enhance the positive aspects and minimize the negative aspects.

The positive features are as follows:

- **Wall protection.** The structure is protected from short-range fusion products by a layer of liquid metal thick enough to absorb all the debris and all but the hardest x rays over more than 98% of the solid angle. This obviates the need for fastening hot, brittle, sacrificial wall materials to cold supporting structures.

- **Self-renewing.** The liquid wall is continuously self-renewing. If required, it can be replaced entirely in the time between pulses. Remote replacement of activated solid walls is avoided.

- **Neutron absorption.** The neutrons are attenuated and moderated in the liquid metal before they reach solid surfaces. Less than 5% of the neutron energy, with a highly degraded spectrum, need reach the structure.

- **Direct energy deposition.** The energy from all types of fusion radiation (photons, ions, and neutrons) is deposited directly into the heat-transfer medium, without having to first pass through a solid surface. The heat-transfer liquid is a metal with high-heat capacity and high-heat-conduction coefficient.

- **Tritium breeding.** Using lithium and lead-lithium mixtures, we can breed sufficient tritium in the LMW to eliminate the need for supplemental blanket modules.

- **Low activation of LMW.** The main activation product of lithium is the tritium needed in the reactor-fuel cycle.

- **Beam propagation.** The low vapor pressure of liquid metals at energy-conversion temperatures of interest (400 to 500°C), coupled with the ability to direct liquid flow to avoid the path of propagating beams, allows the use of either laser or heavy-ion beams in a mode approximating vacuum propagation.

- **Self-pumping.** Most of the gas in the chamber arises from ablation caused by absorption of x rays and debris. This gas is self-pumped by condensation on the very large surface area of cold liquid in the chamber. Although vacuum pumps are required for the noncondensable, nonabsorbed gases (primarily helium), the volume flow is small, the recovery of heat is not required, and the pumping power is negligible.

The positive features of LMW chambers lead to advantages for a power-producing technology:

- The lack of wall ablation and the large attenuation and moderation of neutrons allow the design of a structure that will not require replacement for the entire 30-yr life of the power plant.

- *The low temperature and the low-neutron dose* allow the use of common low-alloy steels, which leads to two extremely important implications: no new exotic material need be developed and the cost of the required materials qualification program is minimized.

- The combination of long structural life and low-chromium, nickel-free steel leads to a very low total mass of activated structure—perhaps over a factor of 20 less than a 316-SS Tokamak.¹

- Because more than 95% of all the fusion energy released is absorbed in the free-flowing liquid metal, separate heat-transfer systems for gas

cooling, wall cooling, and blanket cooling are unnecessary.

- The heat-transfer loops have low maximum temperature and pressure.

- Simple, high-power-density chamber configurations are possible that do not require complex blanket modules having large temperature and pressure gradients, close tolerances, and miles of weldments.

- Because LMW chambers will allow either laser or heavy-ion drivers, there is an advantage in reduced development risk.

LMW chambers also have negative aspects that must be minimized either by clever design or early attention to experiment and development. Most of these are specific to liquid-metal systems. We list them because the strategy must be designed to minimize their impact on the feasibility of the entire system. The negative aspects are as follows:

- Flow rates. Increasing the liquid metal flow rate results in a large increase in capital cost because of increases in pumps and piping, additional containment volume, and recirculated electricity required for pumping power. New programs are required for developing pumps for metals other than sodium.

- Liquid-metal hazards. A large inventory of liquid metal (10^3 tons) is required. Although the amounts are comparable to the requirements of an LMFB the size of SUPERPHENIX, the additional constraints of using lithium or lead compared with sodium are a concern. Fire and toxicity of lithium are potential hazards in the extreme event of breach of containment. Dispersal by a lithium fire of tritium and dissolution of radioactive target elements in liquid metal are of concern in postulating the maximum credible accident.

- Pulsed stresses. The presence of fluid in an inertial-fusion chamber provides a mechanism for creating fluid momentum, which causes impact stresses in the structural wall.

- Repetition rate. The repetition rate of LMW chambers is limited by the time required to inject a new LMW into the chamber. Smaller chambers do not improve the repetition rate much because the thickness of liquid metal required to stop neutrons and the practical upper limit on

velocity are relatively inflexible. The rate of condensation of vaporized liquid may also set a limit, but it now appears that the flow conditions are more restrictive than the condensation problem.

- Chemistry. Corrosion products and the elements from fusion targets will be mixed in the liquid metal and be susceptible to neutron activation. Also, although pure liquid metals are relatively benign in contact with candidate steels, the corrosion rate is considerably enhanced by the presence of impurities (e.g., nitrogen in lithium or oxygen in sodium). Therefore, the chemistry must be controlled. Methods of extracting these materials must be devised. Tritium appears to be one of the easiest to extract. The problem of extracting or separating the materials from the fluid has not been evaluated for either LMW or gas-filled chambers; separation from a liquid metal may prove to be easier than from high-flow-rate vacuum pumps. In any event, we would prefer to minimize the activated material circulating through the primary heat-transfer loop because of the hazard to maintenance personnel.

- Secondary loop. A secondary heat-transfer loop appears to be required to isolate the primary liquid-metal loop from the steam feed-water loop. Although accident safety is our first concern, the inclusion of an intermediate sodium loop also allows cold trapping of diffused hydrogen isotopes to very low levels during normal operation.

- Irradiation geometry. LMW chambers are attractive if replaceable beam tubes are not needed (e.g., if the liquid wall can be separated in a few places to admit the driver beams and pellet). Also, it would be awkward if not impossible to use LMWs if tens of symmetrically arranged beams were required. These chambers work best with few-sided irradiation of reasonably small solid angle, although each aperture may admit a cluster of high-frequency beams.

These limitations of the LMW concept add to the basis for formulating the design strategy in that they form the boundaries of design space. They also are the penalty function in that they represent the costs or factors to be minimized.

In the past, the fusion community has focused on such problems as short wall life, high tritium inventory, and large chemical energy storage as major factors. However, in each case, we see in the HYLIFE concept solutions that allow shifting of emphasis to less severe design limitations.

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Review of LMW Chambers

The history of LMW chambers extends back to 1970, when Christofilos⁵ suggested that a continuously renewing wall of liquid metal might solve the wall damage problem anticipated in an Astron magnetic fusion reactor. The first suggestion of LMW chambers for inertial fusion was the BLASCON reactor shown in Fig. 8-3. It was introduced by Fraas⁶ in 1971 and refined in 1972.⁷ It featured a vortex of swirling lithium contained in a pressure vessel, arranged so that the fusion pellet and laser beam entered from the top, down a long narrow hole in the center of the vortex. They imagined that a vessel 3 m in diameter and 25 cm thick would be appropriate for a 200-MW_e, 1- to 10-Hz system. However, calculations showed that the wall would suffer permanent deformation from the impact of shock waves driven by the energy absorption. A key contribution of this early paper was the suggestion of introducing bubbles into the lithium to attenuate the shock wave. Furthermore, they performed experiments in 1971 with chemical explosives in water containing bubbles, which indicated wall-pressure reductions of a factor of 2.5 or more.

Two concepts published in the mid-1970s that we do not discuss in detail are of the "wetted wall" variety. The first, by Baird and Anderson,⁸ was introduced in order to remedy the pellet constraint of BLASCON. Their reactor consisted of a rotating container of molten lithium, the free surface of which was to form a paraboloid of revolution. The surface was to act as a first wall and as a mirror to reflect the laser light and uniformly irradiate a pellet located at the focus. This reactor and the more practical spherical wetted-wall concept of Booth et al.⁹ are in a separate class of LMW reactors because they have porous structural walls between the pellet and the neutron-absorbing blanket. The wetted-wall concepts have not received much attention recently because they seem to share the worst problems of both dry-wall and LMW chambers (e.g., high-neutron damage, difficulty in getting uniform wall coverage, and difficulty in vacuum pumping be-

Fig. 8-3. The BLASCON-ICF liquid metal chamber.

tween pulses). Because they have a distinct set of engineering issues, we do not include them in our design strategy.

Other LMW chamber designs based on a "falling rain" concept have been introduced. Seifritz called his concept the "falling evaporating molten salt blanket"¹⁰ in which the molten salt contains both lithium and uranium or thorium fluorides to breed fissile materials. Burke¹¹ introduced a similar concept in late 1974. His concept was designed to be driven by a 100-MJ electron beam igniting a fusion pellet placed in an expendable diode structure fuel assembly. The reactor, shown in Fig. 8-4, was intended to contain yields of from 20 to 200 GJ using a LMW consisting of a two-phase flow of helium and lithium 5 m thick. The cylindrical chamber has a 10-m-radius blast shield inside a 15-m-radius primary pressure vessel. Because each pellet is enclosed in a new evacuated diode assembly, the high (near atmospheric) helium pressure is of no consequence. Burke argues that for such large energy gains the cost of sacrificial diodes is tolerable or even inconsequential. He also discusses the possibility of performing direct MHD energy conversion by including a conical channel and superconducting magnets at the bottom of the reactor, arranging the liquid wall to obtain the greatest amount of vaporization and then converting plasma kinetic energy into electricity.

From today's viewpoint, the two design features that were incorporated to deal with large

Fig. 8-4. Burke's electron-beam-drive liquid-wall concept.

blast effects—a two-phase blanket and a double-shell chamber—are desirable and required. Calculations of induced momentum and resultant impact stress in the wall were given in a later publication by Kot and Burke¹² indicating that peak pressures in the lithium blanket can be reduced from the 17 000 bars experienced at zero void fraction to 33 bars at 5% void fraction down to 9 bars at 50% void fraction of helium bubbles. They suggest that the impulse delivered to the wall because of direct neutron heating can be kept below 0.05 bars by this method.

In 1977 two groups published significant design results. Maniscalco et al.¹³⁻¹⁵ reported results of scoping studies that surveyed several inertial-fusion reactor designs and concluded that the "waterfall" reactor shown in Fig. 8-5, which uses a liquid-lithium blanket and a ferritic-steel structure, shows the greatest promise. The major contribution of this work is in the quantification of the lithium thickness required to obtain a 30-yr life in 2-1 4 Cr-1 Mo steel. A method was developed to properly account for the effect of neutron spectrum moderation in both the pellet and lithium in determining the displacements per atom (dpa) and helium production rates.

Fig. 8-5. The early I.I.I. lithium-waterfall concept.

Maniscalco and his associates at I.I.I. envisioned a free-falling annulus of liquid lithium, with replaceable beam tubes extending through the fall. In this design, the top is protected by a vortex of lithium. A double-wall chamber is used to isolate the pulsed-tensile loading of the inner wall from the outer wall, which experiences a predominantly steady compressive load. A deficiency of this design involves the particular configuration of the LMW.

Subsequent calculations show that a uniform annulus is inappropriate. The plasma column formed from the vaporized liquid metal pushes the liquid out radially so that, in conjunction with the velocity produced by neutron deposition, the momentum delivered to the wall is unacceptably high.

The other important concept of 1977 is the Boiler and Moderator (BAM) of Powell, Lazareth,

Fig. 8-6. Liquid-wall boiler-and-moderator (BAM) concept.

and Fillo,¹⁶ shown in Fig. 8-6. They envisioned a vertical cylinder with a radius of from 1.5 to 4 m, with the height three times the width, and with the ability to contain 10 GJ yields every few seconds. In this concept, an LMW of lithium, ${}^6\text{LiBe}$, or lead with a small percentage of ${}^6\text{Li}$ falls vertically, leaving a gap between the liquid and solid structure. The LMW consists of two zones: an inner spray zone several centimeters thick of droplets designed to absorb the x rays and debris, and a thicker, continuous zone of liquid metal that attenuates and moderates the neutrons. The ends of the cylinder are closed off by either an intermittent flow or a magnetically driven device that pinches off the annulus of liquid metal on a pulsed basis.

Powell and his associates at Brookhaven National Laboratory (BNL) devoted their attention to the two most important problems: the momentum imparted to the LMW when the fusion energy is absorbed and the rate of condensation of hot plasma. Regarding the first problem, they conclude that the momentum delivered to the liquid by the outward thrust of the high-pressure gas exceeds that from the reaction force caused by the initial ablation, but that it can be tolerated for nominal-size chambers. Regarding the second problem, they conclude that rapid condensation of the hot plasma in

the inner spray zone is likely to substantially decrease the outward thrust on the liquid. Unfortunately, they do not calculate the momentum induced by absorption of the neutron energy, which can dominate. They do, however, give a method for decreasing this momentum should it prove to be important—that is, to make the entire LMW a dense two-phase flow.

To calculate condensation time, the BNL group assume the conditions of a heat-conduction-limited process. They calculate the time required for all energy originally in x rays and debris to be conducted into the droplets, assuming that the spray zones containing droplets are 5 cm thick and 1 cm in diameter. They calculate condensation times ≤ 1 ms assuming a rate of heat transfer from the vapor to the drop that is considerably faster than conduction time within the drop and conclude that condensation rates are unlikely to be a problem.

In 1978 Burke¹⁷ proposed the heavy-ion beam driven reactor shown in Fig. 8-7. The chamber consists of a horizontal cylindrical drum that is rotated mechanically. The lithium blanket is maintained at the wall by centripetal force, the pellet and beams are inserted from the ends of the cylinder. A rotating seal is suggested that will allow cold lithium to be pumped in, while hot lithium and the exhaust are removed at outflow weirs at the two ends. No calculations or documentation yet exist for this intriguing design.

The most recent conceptual power plant design, and the one for which the most detailed calculations have been performed, uses the HYLIFE chamber designed by L.L.^{18,19} A drawing of the chamber is shown in Fig. 8-2. This 20-man-year study (in conjunction with university and industrial contractors) resulted in a simple chamber design that can conceptually be operated with yields of several thousand megajoules up to a few times a second. A lithium energy-conversion blanket consisting of a dense array of 20-cm-diam jets is injected into the chamber between each pulse, providing an effective blanket thickness of 1 m between the fusion pellet and the inner steel wall. The 14-MeV neutron flux is reduced by a factor of 200 by this lithium blanket, allowing a 5-m-radius inner wall to operate 21 full-power years (30 yr at 70% availability) at an integrated neutron energy flux of 0.3 MW m^{-2} . Because the wall flux in the absence of lithium protection would be 5.7 MW m^{-2} , including effects of the pellet, the power density within the

Fig. 8-7. HIB reactor with centrifugal blanket.

reactor vessel is very high, approaching that of a fission reactor. A common low-alloy-ferritic steel (2-1/4 Cr-1 Mo) is used throughout. The tritium-breeding ratio is controllable between 1.0 and 1.7.

The fluid configuration in the chamber is an annular hexagonal-close-packed array (50% packing fraction) of 20-cm-diam lithium jets, resulting in effective shock isolation from the effects of the fusion pulse. This configuration minimizes the wall stress resulting from impact of lithium accelerated by the high-pressure, x-ray and debris blowoff gas, because the hot gas merely blows through the array of jets. The force of the volumetric expansion of fluid resulting from the neutron absorption is primarily absorbed in liquid-liquid interactions of colliding jets. Finally, the enormous area of the fluid at relatively cold temperatures (500°C) acts as an effective condensation pump, avoiding the need for mechanical vacuum pumping of hot plasma.

The characteristics of the power plant are as follows:

- Yield per pulse 2700 MJ
- Pulse repetition rate 1 Hz

- Net electrical power 1068 MW_e
- Recirculated power 14.8%
- Net electrical efficiency 33.6%

The HYBEE power plant requires 16 liquid-metal pumps to inject the lithium into the 5-m-radius chamber. The lithium recirculation pumping power is ~20 MW_e—only 1.6% of the gross electric power.

The most valuable results from the above studies are the engineering issues that were uncovered. We discuss the most important issues individually in the next article.

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Guidelines for the Design of an LMW Chamber

In the paper from which this design strategy has been condensed,²⁰ we discussed the engineering constraints and dominant tradeoffs that must be made in designing an LMW chamber. Because much of the methodology is given in detail in other articles in this annual report, the methodology will not be repeated here. (See articles beginning with "New or Revised Calculations of the High-Yield

Lithium-Injection Fusion-Energy Converter (HY-IEEE) System" and ending with "Material Considerations for Liquid-Wall ICF Energy Converters" in this section.)

Also, because different portions of the HY-IEEE design evolved at different rates, we do not yet have a perfectly self-consistent design. The small inconsistencies which do remain provide a strong motivation to consider a proper design methodology to prevent future inconsistencies. We therefore present a summary of what we have learned in the form of a suggested design strategy.

Oversimplification is a risk in giving a set of rigid design guidelines because the tradeoffs are inevitably more complex than stated. There are many exceptions to the guidelines that follow, yet the alternative of stating all the important factors to be globally optimized is unconstructive. With this proviso, we offer the following guidelines as a basic strategic framework for pulsed-fusion chamber design using a damage-proof, reestablishable LMW.

- Select a fusion yield and repetition rate based on desired electric-power output and reasonable assumptions for pellet gain and driver characteristics, and estimates of recirculated power requirements. The resulting design will almost certainly suggest a more attractive combination of yield and repetition rates, leading to an iterative design procedure.

- Select a liquid metal based on the tradeoff between pumping-power requirements and potential safety considerations. Lead requires approximately ten times more recirculated pumping power than lithium, but a lithium system must be designed to minimize the potential hazard of the stored chemical energy of hundreds of tons of lithium.

- Select a thickness of liquid metal and radius of steel liner to achieve a structural life equal to power plant life based on radiation damage constraints. This will minimize the total mass of activated structural material.

- Provide high enough flow velocity to ensure reestablishment of the LMW, and provide sufficient fluid area to recondense the vaporized liquid within the desired interpulse time.

- Choose a low-alloy, low-activation, low-corrosion, low-radiation-damage ferritic steel to minimize the required materials-characterization-and-development effort.

- Use a double-shell chamber configuration to separate the functions of withstanding pulsed-radiation and stress loadings from those of the vacuum and containment vessel, which must withstand a steady compressive load

- Choose the highest operating temperature and corresponding vapor pressure allowed by driver propagation constraints, and by the strength and corrosion rate of the steel chosen

- Because the imposition of any liquid in the chamber provides a mechanism for creating momentum that can be transferred to the structure, seek an LMW configuration that will minimize the induced momentum while absorbing the fusion energy. A two-phase flow will reduce both the momentum imparted to the fluid and the wall stress resulting from the impact

- The thickness of the steel liner and the inner radius of the LMW are chosen simultaneously in order to meet two criteria on allowed stresses. Increasing the liner thickness will reduce the impact stress, but will also increase the peak wall temperature and thermal stress (and decrease the wall strength). Moving the LMW farther from the pellet and closer to the inner liner will reduce the impact stress without affecting the wall strength or thermal stress, but at a cost of increased flow-rate requirements. Therefore, select the steel-liner thickness that results in positioning the LMW closest to the pellet, while meeting both constraints on allowable stress. This strategy minimizes the required mass-flow rate of liquid metal.

- Use high-f number-focusing optics stacked into closely packed arrays in an irradiation geometry that irradiates the pellet from only a few directions to minimize the area of penetration through the LMW.

- In designing the balance of a plant, use an inert atmosphere and many separate steel-lined concrete cells to minimize the hazards of accidental liquid-metal leakage. Use an intermediate heat exchanger loop to ensure the physical separation of radioisotopes in the LMW loop from the steam loop.

The emphasis in the design strategy given above is placed on features that minimize social and economic costs while meeting the engineering constraints. Minimizing the mass of activated material and minimizing the power recirculated for pumping are the primary examples. Most problems in inertial-fusion-reactor design (e.g., radiation

damage, impact stress, and thermal stress) can be solved by increasing the liquid-metal flow rate and inventory, but liquid-metal pumping and plumbing are expensive in both component cost and required containment volume. Therefore, innovations in the design of the chamber and loop configurations to minimize the liquid-metal flow rate; and thus pumping cost, liquid-metal building space, and recirculated power have extremely high leverage.

We hope that this simple strategy will form the basis of a broad industrial effort to design and engineer more sophisticated LMW reactors, which will lead to a technically feasible inertial power plant, constructed of readily available materials, at acceptable economic and social cost.

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New or Revised Calculations of the High-Yield Lithium-Injection Fusion-Energy (HYLIFE) System

During 1979 we increased the sophistication of analytical models used in the HYLIFE design. We also extended our simple models to screen the design parameters and select those that were optimum for our analyses. Finally, we addressed several design issues for the first time.

In this set of articles, we consider one complete fusion-pulse-response sequence in the HYLIFE chamber. In the first two articles, we describe the vibrational modes of the chamber and its components, and the effect of nozzle vibration on lithium-jet stability.

In the third and fourth articles, we describe the x-ray and debris-induced blowoff from the LMW array, the formation of a lithium plasma near the chamber axis, and the subsequent expansion of the plasma through the jet array that makes up the LMW in the HYLIFE concept.

Fig. 8-8. HYLIFE-target chamber (1978 version) used in preliminary vibrational analysis.

In the fifth article, we discuss the absorption of reaction energy by the LMW as a function of LMW density and position. In the sixth article, we discuss the neutron-induced disassembly of a thick, annular LMW (all), and the effect of fall position on wall stresses. To accommodate larger fusion yields, we segmented the LMW into a 50%-packing-fraction array of jets. In the seventh article, we analyze the response of the segmented LMW to the fusion reaction.

In the eighth and ninth articles, we summarize the first structural-wall (FSW) design procedures, and describe the tradeoffs between LMW array position and wall thickness. Finally, in the last article, we discuss generation of both condensable and noncondensable vapors in the chamber.

Vibrational Analysis of the HYLIFE Chamber

We performed a preliminary structural analysis of the HYLIFE chamber described in the 1978 Laser Program Annual Report²¹ to determine the natural frequencies of components that make up the target-chamber assembly.

We evaluated the natural frequencies of the target-chamber assembly (Fig. 8-8) using the pipe-element option of the SAP V finite-element computer code.²² The assembly includes the primary tank, first wall, graphite blankets, and closure plug. We used a coarse-mesh, simplified model; thus the results are only preliminary.

The lithium-inlet and -outlet piping were not included in the model because they were assumed to be flexible relative to the target-chamber-assembly support ring; therefore it was assumed they would not influence assembly frequencies. Individual items, such as the orifice plate, guide baffle, nozzle plate, and splash baffles were not specifically modeled; but their mass effects were included. We determined the frequencies of these components with simplified hand calculations.

The shape, mass, and fundamental frequency of each component are shown in Fig. 8-9. The first vertical natural frequency of the assembly is 25 Hz. None of the frequencies is close to the repetition rate of 1 Hz.

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Fig. 8-9. Results of preliminary vibrational analysis.

Component (weight in 10^3 kg)	Description	Weight (kg)
		First horizontal mode frequency (Hz)
Primary tank (1000)		10.0
First wall (160)		8.7
Plug (29)		11.9
	8.7
		First vertical mode frequency (Hz)
Slosh baffle (87)		142
Orifice plate (32)		41
Guide baffle (71)		176
Nozzle plate (54)		70
Splash baffle (200)		58

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Experimental Investigation of the Stability of Liquid-Sheet Jets

Introduction. A potential problem arises in the application of liquid jets in the HYLIFE converter. The reactor vessel and jet nozzles could sustain large amplitude vibrations from "ringing" of the entire structure caused by the pulsed nature of laser-fusion microexplosions. The University of California-Davis theoretically and experimentally analyzed the stability of jets under forced vibrations as a part of the HYLIFE design study. Jet breakup would allow neutrons, ions, and x rays to stream directly onto the structure and to thereby drastically shorten the useful life of the chamber.

Extensive studies of the breakup of fan-shaped-sheet jets and sprays have been made by Dombrowski and Munday as part of spraying and atomization studies.²³ However, for our geometry of a nearly constant-width sheet jet, we found only two relevant stability analyses in the literature. In 1953, Squire²⁴ published a theoretical study of the linear temporal stability of a horizontal sheet jet. Two years later, Hagerty and Shea²⁵ independently published a similar theoretical analysis along with experimental data directly relevant to our study. Their experiments employed a very thin, horizontal sheet of water with a thickness of only approximately 1.6 mm and with jet velocities of approximately 3 to 5 m/s. The jet nozzle was vibrated transverse to the flow direction at frequencies of up to 200 Hz. Measurements of the exponential growth rates agreed very well with the theoretical predictions.

In the current experimental study, we have extended the work of Hagerty and Shea to better simulate the HYLIFE converter environment. We have included the following:

- Thicker sheet-water jets.
- Large-amplitude forced excitation up to several hundred Hz.
- Vertical (downward) jet orientation.
- Experiments at 20-kPa pressure in a vacuum vessel as well as at 1 atm.

We briefly review the key results of the linear-temporal-stability theories of Refs. 24 and 25. Then we describe an alternative simplified spatial-growth-rate theory referred to as "trajectory theory," which

was developed to explain larger-amplitude, forced-excitation data. Finally, the experimental apparatus is described and results are compared with various theories.

Theory. Linear-temporal-stability theories developed in Refs. 24 and 25 are essentially equivalent, and both are basically extensions of the theories of Rayleigh²⁶ and Lamb²⁷ for temporal growth of waves on a single free surface to the present case of two free surfaces. Linear theory only permits two types of waves to form on the two free surfaces:

- Waves that are in phase (sinuous mode).
- Waves that are 180° out of phase (dilatational mode).

Sinuous waves always have a higher growth rate in the unstable regime and therefore tend to be the dominant waves. Also, linear theory predicts that both the growth rates at a given frequency and the critical frequency decrease significantly as ambient air pressure is reduced. In a vacuum, the sheet jet should be stable to all disturbance frequencies.

In actual experiments, waves grow in space rather than in time. Calculated temporal growth rates are often transformed to spatial growth rates by dividing by wave velocity for comparison with experiments. Thus for liquid-sheet jets when wave velocity is essentially equal to jet velocity, linear theory predicts a spatially growing, exponential wave envelope for unstable cases.

Our experiments purposely imposed large-amplitude forced excitation on sheet-jet nozzles to simulate the worst situation for laser-fusion application. The resulting wavy motion of sheet jets had a very large amplitude; peak amplitudes were often an order of magnitude larger than the jet thickness as illustrated in the photograph of the edge view of the sheet jet shown in Fig. 8-10. Clearly, linear-stability-theory results are suspect under these conditions and will be shown to only partially agree with experimental results. Consequently, we decided to develop a simple first approximation to a spatial-stability theory.

Our approach was to first consider the fluid particle at the crest of the wave shortly after the flow leaves the nozzle. We then assumed that the wave speed was approximately equal to the vertical component of the local jet velocity. This assumption implies that a fluid particle that starts in the wave crest stays in the wave crest, and that the trajectory of the particle traces the envelope of the waves.

We performed an experiment to verify this approximation, because it is one of the key assumptions of the trajectory theory. Dimples were formed in the wave crests on one side of the flow as the waves passed a point a few centimeters below the nozzle exit. The dimple remained in the wave crest on the side where the dimple was originally produced, even for very-large-amplitude waves.

We then assumed that the particle that remains in the wave crest is the one that leaves the nozzle with the maximum initial transverse velocity at the time the vibrating nozzle passes through the vertical axis.

Equations of motion in the vertical and horizontal directions can be formulated for aerodynamic forces caused by the air-pressure difference across the wavy jet and the surface-tension forces on the fluid particle in the wave crest. If aerodynamic and surface tension forces are linear and continuity is applied, the equation of motion in the horizontal direction is a linear second-order differential equation with time-dependent (or space-dependent) coefficients. This equation has been solved numerically for our specific nozzle geometries and flow conditions.^{28,29}

For the special case of high initial jet velocities (≥ 5 m/s), the velocity and jet thickness changes in the fall times of the experiments are very small. After a short distance, the trajectory theory solution to the wavy-jet envelope approaches a simple exponential that has the growth rate predicted by the spatial interpretation of the linear-temporal-stability theory. However, the envelope amplitude predicted with spatial-trajectory theory is larger than that given by the linear theory.

Linear-stability theory can be modified to give essentially the same results as the spatial-trajectory theory by changing the initial conditions to flat sheet surfaces with surface velocity distributions that are initially spatially sinusoidal. Since this is an unusual initial condition for a linear-stability analysis, it was perceived as the one that would give the correct transformation to our spatially growing wave situation only after the simple trajectory theory gave such good agreement with experimental results.

Experimental Apparatus. A schematic diagram of the experimental apparatus is shown in Fig. 8-11. The flow pipe enters the vacuum vessel vertically and is attached to various nozzles used in the experiments by means of a coupling that provides a

Fig. 8-10. Edge view of a typical near-sinusoidal exponentially growing waveform of the 2.5-mm-thick sheet jet with forced excitation of the nozzle at 70 Hz, initial velocity of 4.75 m/s, forcing amplitude of 1.06 mm, and pressure of 100 kPa.

smooth transition on the inner surface. Two nozzles were fabricated from 50.8-mm (2-in.) cast acrylic tubes heated and pressed at one end to form a constant-slit exit (constant jet thickness). Jet thicknesses were 2.5 mm and 7.9 mm for the two nozzles. A water-jet ejector pump was used to

Fig. 8-11. Schematic diagram of the water-flow system, vacuum-tank apparatus, and forced-excitation instrumentation used in the sheet-jet experiments.

vacuum pump the experimental chamber to approximately 20 kPa.

The nozzles were vibrated transversely by means of an electromechanical shaker mounted outside the vacuum tank and attached to the nozzle by a rod that passes through a bellows-type feedthrough into the vacuum chamber.

Water exits the nozzle and falls approximately 0.7 m to a small pool at the bottom of the vacuum tank. For the tests at atmospheric pressure, the water then simply flows out from a drain pipe in the bottom end plate to a drainage sump. For subatmospheric tests, water is pumped to the drainage sump and a constant head of approximately 0.3 m of water is maintained in the bottom of the vacuum tank. The experimental apparatus is isolated from pump vibrations.

Basic wave-form amplitude data were obtained from photographs of the edge of the wavy-sheet jet (Fig. 8-10). This photograph is the superposition of a long time-exposure photo used to define the wave

envelope and a short flash exposure to show up the wave form and permit measurement of wave lengths.

For vibrational amplitudes large enough to be read from photographs, measured values agreed with those calculated from accelerometer readings to within approximately $\pm 5\%$. Only accelerometer readings were used for smaller vibrational amplitudes.

In reading the amplitude of the mean-wave envelope at various vertical positions, it was necessary to subtract the thickness of fluid accumulations in sheet edges, as well as fluid lumps or "blobs" near the edge regions of the wave crests. The fluid lumps were often present at very large wave amplitudes and low jet velocities. Reasonably accurate corrections were possible after careful visual study of the jet-wave shapes using a stroboscope. The estimated accuracy of the corrected mean-wave amplitude data is ± 0.75 mm for the thick jet (sheet thickness = 7.9 mm) and ± 1.0 mm for the thin jet (sheet thickness = 2.5 mm). However, this is similar to a

Fig. 8-12. Typical 2.5-mm sheet jet experimental results for the mean envelope of the waveform (excluding fluid blobs) compared with the linear theory (LT) and the trajectory theory (TT).

bias error, since the eye can integrate and smooth the data read from the photographs quite well.

Experimental Results. We conducted experiments over a range of forcing frequencies from 40 to 300 Hz, at flow velocities up to 5 m/s. The forcing amplitudes ranged from very small values of only 1% up to 40% of the jet thickness to simulate severe vibration of the reactor-nozzle plate. Resultant mean amplitudes of the water jets were as high as 200 times the forcing amplitude. Even under these extreme conditions, the sheet jets showed a remarkable resistance to breakup or droplet formation for fall distances of over 0.5 m.

Results of a typical experiment, where the waveforms were sinusoidal like, are shown in Fig. 8-12. The results for the growing waveform at 100-kPa pressure agree very well with those of a simple spatial-trajectory theory (TT-B), while the linear theory (LT-A) yields the correct growth rate of the wave envelope but substantially underpredicts the wave amplitudes because of the incorrect choice of

the initial condition mentioned above.

At low pressure, wave amplitude is limited by surface-tension forces, as can be seen from the 20-kPa test.

The excellent agreement between the data collected in the 25-cm fall distance and the simple-trajectory theory for the thin (2.5 mm) nozzle was found to hold for a wide variety of initial amplitudes provided that the amplitudes were less than approximately one-half the initial-jet thickness and that the waveform was not too far from sinusoidal in shape. This appears to confirm the two key assumptions made in developing the trajectory theory:

- Wave speed equals the jet speed so that a fluid particle that starts in the crest stays in the crest.
- The fluid particle that is in the wave crest at the end leaves the nozzle with the maximum initial transverse velocity.

For the thick nozzle, the initial jet thickness was 7.9 mm (compared with 2.5 mm for the thin nozzle). This large thickness was chosen because one of the basic objectives of this research was to study thicker jets than those employed by Hagerty and Shea (1.6 mm). Our thicker jets were limited by the apparatus to lower velocities (typically 1.65 m/s) so that spatial variations of the velocity and thickness were expected to be much more important than for the thin nozzle. A typical waveform for large-amplitude excitation of the thick nozzle is shown in Fig. 8-13. The waveform is almost a sawtooth with fluid blobs projecting outward from the wave crests. These fluid blobs were primarily at the edges rather than the faces of the sheet, with much smaller fluid accumulations along the entire wave crests on the faces. They stayed attached to the main waveform for tens of wavelengths and fell with the main waves. During the growth of these blobs, the sheet thickness varied continuously along the jet, being notably larger at the wave crests. The dramatic appearance of this highly nonsinusoidal waveform when viewed with a strobe can be appreciated from Fig. 8-13; the unexpected stability against breakup into droplets cannot yet be fully explained.

For highly nonlinear and nonsinusoidal, sawtooth-like waveforms produced on the thick (7.9-mm) sheet jets at high-excitation amplitudes,

Fig. 8-13. Edge view of a typical sawtooth waveform of the 7.9-mm-thick sheet jets with forced excitation at 70 Hz, initial velocity of 1.65 m/s, forcing amplitude ≈ 1.27 mm, and pressure of 100 kPa.

none of the theoretical models fully explains the observed wave amplitudes, and further research in this area is needed. Work on a nonlinear theory for the wave growth is now in progress at the University of California-Davis.³⁰ Additional small-scale water-jet experiments are underway at UC Davis to extend our knowledge of thicker jets and to investigate other jet breakup modes.

Summary. The scaling laws implied by the trajectory theory (and the modified linear theory) indicate that the envelope of the waves on thin-sheet jets in a vacuum environment will be limited in amplitude by surface tension forces. Extrapolations of the trajectory theory indicate the HYLIFE jets will not break up or coalesce before the fusion pulse. Nonlinear-theory development and thicker jet experiments are now being pursued to increase our understanding.

Ultimately, however, some large-scale water jet experiments at higher Reynolds-and-Weber numbers, and with fall lengths of several meters, will also have to be performed. These experiments will verify the scaling laws, providing us with increased confidence that the lithium jets will not break up or coalesce before the fusion pulse is initiated.

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Central Region Pressurization Caused by X-Ray and Debris-Generated Lithium Blowoff

Introduction. In this article,³¹ we describe the behavior of the lithium blowoff plasma generated by the absorption of the 25 to 35% of the fusion energy that is unable to penetrate beyond a very

thin layer in the innermost exposed jets. The specific energy of the lithium in this thin layer increases by several orders of magnitude; and the instantaneous pressure may exceed 100 GPa. A strong shock wave then radiates from the short-range deposition region, but is quickly captured and attenuated by a rarefaction centered at the free inner surface. The blowoff velocity at the tail of the rarefaction is estimated to be ~ 40 km/s from earlier calculations³² using a 750-MJ target and a 2-m-fall inner radius. However, this figure is too low for the higher-energy-density systems now being considered. Moreover, the velocity continues to increase as the lithium blowoff implodes on the chamber axis, which is a consequence of the energy-focusing effect of the convergent geometry. At lithium-implosion time, the kinetic energy thus acquired is reconverted to internal energy. Much, if not most, of the energy is in the form of radiation.

The subsequent energy flow is controlled both by radiation transport and hydrodynamic motion. In our initial calculations (a 750-MJ chamber with 2-m-fall inner radius), the mean free path for radiation was large compared with the dimension of the reaction chamber so that the central-cavity lithium plasma was considered to be optically thin and the emission approximation valid. If the energy and mass density in the cavity are sufficiently high, this approach no longer works; and the diffusion approximation must be considered.

The Design Basis: HYLIFE. Our previous calculations³² of the lithium implosion and blowback were based on a 750-MJ pulse deposited in a lithium annulus with inner and outer radii of 2.0 and 2.6 m, respectively. The thickness of lithium for which $e \geq e_c$ (the cohesive energy, ~ 23 MJ/kg) was $\sim 10^{-2}$ mm (this is a measure of the blowoff mass) so that the lithium-plasma density in the central cavity at blowoff-implosion time was on the order of 10^{-6} Mg/m³. Figure 8-14 shows the Rosseland mean opacity for lithium as a function of temperature and density; the data derive from calculations performed at Los Alamos Scientific Laboratory by W. Huebner.³³ For $\rho = 10^{-6}$ Mg/m³, the maximum value for K_R is $\sim 10^3$ m²/kg at a temperature slightly below 1 eV. For higher and lower temperatures, the opacity decreases sharply; at 10 eV, $K_R \sim 1$ m²/kg and at 0.125 eV (the lowest temperature for which we have tabulated data), $K_R \sim 10^{-6}$ m²/kg. Thus the radiation-mean free path is bounded by $1 \text{ m} < \lambda_R = (K_R \rho)^{-1} < 10^9 \text{ m}$, so that

the emission approximation appears fully justified. (Calculations based on the Planck mean opacity—cf. Fig. 8-15—would reduce the minimum bound by approximately one order of magnitude.)

Current reactor designs call for greater energy per pulse and a more compact fall configuration. The HYLIFE system employs a 2700-MJ thermonuclear pulse in a close-packed, annular-jet array (300 jets, each 200 mm in diameter) that surrounds the pellet on an inner radius of ~ 0.5 m. In this case, the thickness of the penetration layer ($e \geq e_c$) is approximately 300 times higher and the central cavity cross section four times less than before, so that the lithium-plasma density at implosion is 1200 times higher. The minimum value of λ_R is then $\sim 10^{-3}$ m, which is considerably less than the cavity radius so that, at least at temperatures near 1 eV, the plasma cannot reasonably be considered optically thin. This has a very strong influence on the energy transport and the plasma motion in the cavity.

Calculations with the Emission Approximation.

Each plasma zone ($e \geq e_c$) in the cavity is considered to be a ring source that radiates to the relatively cool ($e < e_c$) residual fall. Radiant energy released by the lithium implosion is absorbed initially by the first interior zone in the fall that is opaque. This zone then heats and expands until it becomes transparent, at which point no further radiant absorption is allowed and the next adjacent two-phase (liquid-vapor) or liquid zone acts as the radiation sink. This process is coupled with the energy flow by hydrodynamic motion and continues until the radiation flow no longer influences the motion.

Figure 8-16 shows the Lagrangian motion within the Eulerian reactor cavity as a function of time. The Eulerian-cavity radius (ECR) is defined as that surface to which the inner boundary of the innermost lithium annulus (or, approximately, the innermost row of jets) is congruent at $t = 0$. Proceeding in the counterclockwise direction from the initial state in Fig. 8-16, the inner boundary of the annular fall has imploded on the cylindrical axis at $t = 1.5 \mu\text{s}$; the collapse velocity is almost 400 km/s, corresponding to an energy of ~ 80 GJ/kg. The fluid initially contained within a 1-mm-thick annulus, at a radius of 0.5 m (the first zone), now occupies the cavity core to a radius of 166 mm. The

Fig. 8-14. The Rosseland mean opacity for lithium as a function of temperature and density.

peak temperature in the core is ~ 100 eV and the peak pressure ~ 1 GPa. Approximately 200 Lagrangian zones have entered the Eulerian cavity.

By $t = 5 \mu\text{s}$, the peak temperature has dropped to ~ 40 eV. Figure 8-17, which is a graph of the energy radiated from source to sink as a function of time, shows that 30% of the initial plasma energy has been emitted. (E_0 is the total energy initially contained in fluid for which the specific internal energy exceeded the cohesive energy.)

Most of the radiant emission occurs in the first $50 \mu\text{s}$ as plasma continues to move toward the axis and to compress the core. The work done thereby

sustains the radiation transport, which, as seen in Fig. 8-17, is essentially completed in this interval. The energy absorbed by the two-phase liquid-vapor region (60% of E_0 at $50 \mu\text{s}$) causes this material to heat, expand, and accelerate towards the axis. In Fig. 8-16, the region is represented by relatively wide zones that appear near the ECR beginning at $t = 10 \mu\text{s}$. The significance of this is that the inward-moving, two-phase fluid has a mass that exceeds that of the outward-moving, low-density plasma by several orders of magnitude. By $60 \mu\text{s}$, the boundary between the two regions is approximately half way between the axis and the ECR. However, this boundary is unstable and the resulting mixing and

Fig. 8-15. The Planck mean opacity for lithium as a function of temperature and density.

molecular transport would be expected to cool and condense the hot plasma. This, in turn, will reduce the requirement for mechanical exhausts—a vexing feature of drywall ICF systems. (The ambient pressure between pulses must be as low as 10^{-4} Torr for some of the driver systems under consideration.)

The spatial distribution of pressure, temperature, and particle velocity at $t = 60 \mu\text{s}$ is shown in Figs. 8-18 to 8-20. These results are discussed in the following paragraphs where the equivalent results obtained using the diffusion approximation are presented. The time basis of $60 \mu\text{s}$ was chosen for the comparison because the diffusion results at

this time served as initial conditions for the cross flow of plasma through the annular array of jets that constitutes the liquid-metal wall in the HYLIFE design (see the following article).

Calculations with the Diffusion Approximation.

In our earlier calculations with the diffusion approximation, we used an explicit method devised by Trulio.³⁴ This method allows somewhat larger time steps than would otherwise be computed when the mean free path for radiation, λ_R , is of the order of or greater than the zone size. In the limit of $\lambda_R \rightarrow \infty$ —for example, photon propagation in a near

Fig. 8-16. Lagrangian motion within the Eulerian-reactor cavity as a function of time, as calculated with the emission approximation.

vacuum—diffusion theory leads to the physically absurd result that for a field of finite energy density, the radiant-energy flux is unbounded. Actually, no matter what the value of λ_R , the magnitude of the photon-energy flux cannot exceed $c\rho e$ even if all the photons were to move in one direction. In Trulio's method, the time step is proportional to the zone size, Δx , rather than to $(\Delta x)^2$, and is independent of λ_R when $\lambda_R \rightarrow \infty$. Still, calculations were far too costly; and it was necessary to implement implicit differencing. Our method is based on a scheme

derived by Christy,³⁵ in which a simple Newton-Raphson-like iteration procedure is used for direct solution of an implicit form of the nonlinear diffusion equation, coupled with explicit Lagrangian hydrodynamics. Our method differs mainly in that we solve the diffusion equation separately, in sequence with the hydrodynamics. This permits an arbitrary, time-dependent coordinate system to be employed, without affecting any conservation requirement; mass, momentum, and energy are exactly conserved. Other minor differences are the boundary conditions, the use of a flux limiter, and

Fig. 8-17. The ratio of the total energy radiated to the total energy initially contained in the core gas as a function of time, as calculated with the emission approximation.

Fig. 8-18. Radial distribution of pressure in the Eulerian-reactor cavity at $t = 60 \mu s$ as calculated with the emission approximation

the employment of tabular data, rather than a simple analytical form, for the opacity. The specific boundary conditions and finite difference equations used are listed and explained in Ref. 31.

Figure 8-21 shows the Lagrangian motion within the Eulerian reactor cavity, calculated with the diffusion approximation, and should be contrasted with Fig. 8-16. Major differences are apparent as early as at $t = 5 \mu s$, the first frame shown

Fig. 8-19. Radial distribution of temperature in the Eulerian-reactor cavity at $t = 60 \mu s$, as calculated with the emission approximation.

Fig. 8-20. Radial distribution of particle velocity in the Eulerian-reactor cavity at $t = 60 \mu s$, as calculated with the emission approximation.

after the implosion. The radiant energy generated near the axis diffuses rapidly into the cooler incoming plasma, but only until a temperature of ~ 1 eV is reached. At that point, as is seen in Fig. 8-14, the plasma becomes effectively opaque. This blocks the hot interior core from radiating to the cool liquid or

Fig. 8-21. Lagrangian motion within the Eulerian-reactor cavity as a function of time, as calculated with the diffusion approximation.

liquid-vapor region, as was the case with the emission approximation. No inward-moving mass of cool fluid is produced and the hot-cavity plasma moves outward unopposed against the main fall.

Figures 8-22 to 8-24 depict the spatial distribution of pressure, temperature, and particle velocity at $t = 60 \mu s$ and should be compared with the equivalent emission-approximation plots (Figs. 8-18 to 8-20). The cavity pressure in both cases is approximately the same, 0.4 GPa; but the volume in which this pressure is contained in the emission case is

only approximately 40% as large as for diffusion. This is consistent with Fig. 8-17, which shows that, in the emission case, 60% of the energy initially contained in the cavity has been radiated to the cool zones by $t = 60 \mu s$; effectively, none of the cavity energy has escaped in the diffusion case. Comparison of the temperature profiles reveals the same conclusion. With diffusion-limited-radiation flow, Fig. 8-23 shows the peak temperature in the core at $60 \mu s$ to be more than 15 eV, dropping off gradually to the 1 eV level at a radius of 0.32 m; a 100-mm-wide opaque layer (the dark, densely packed region

Fig. 8-22. Radial distribution of pressure in the Eulerian-reactor cavity at $t = 60 \mu\text{s}$, as calculated with the diffusion approximation.

Fig. 8-23. Radial distribution of temperature in the Eulerian-reactor cavity at $t = 60 \mu\text{s}$, as calculated with the diffusion approximation.

in Fig. 8-21) insulates the cavity from the rest of the fall. By contrast, Fig. 8-19 shows the peak temperature at $60 \mu\text{s}$ with emission-limited-radiation flow to be 2 eV.

Finally, comparison of the particle velocity traces in Figs. 8-20 and 8-24 shows in more detail the principle sketched in Figs. 8-16 and 8-21. With diffusion at $60 \mu\text{s}$, the leading edge of the core plasma has blown back almost to the ECR and the peak outward radial velocity exceeds 5 km/s. With

Fig. 8-24. Radial distribution of particle velocity in the Eulerian-reactor cavity at $t = 60 \mu\text{s}$, as calculated with the diffusion approximation.

emission, the cooler fluid is rushing inward at speeds up to 4 km/s and the outward core plasma motion is on the verge of being arrested.

Conclusion. Radiation transport strongly affects cavity motion in the period immediately following lithium implosion. If the emission approximation were valid, the reactor designer's task would be easier. The core plasma would be immersed in cooler elements from the main fall, its momentum countered, and its subsequent influence very much reduced. Mixing and molecular transport would likely cool and condense the plasma within the Eulerian-cavity radius. Unfortunately, use of the emission approximation is probably overly optimistic for the HYLIFE system. For this design, radiation diffusion seems to be a better—although still not totally accurate—approximation for the transport. With diffusion, the hot-core plasma will blow back against, and interact with, the main fall. This will increase the outward-directed momentum of the fall. Thus the impulse imparted to the walls of the reactor will be greater. The heat-transfer problem will be more complicated, although not necessarily more worrisome from a practical standpoint.

The main problem with both approximations to the transport equation is that neither works over the entire region of interest. Generally, emission is

better near the axis where the temperature is high and the density low, and diffusion is better elsewhere. There are several methods for describing radiation transport in optically thin and thick regions that are usefully accurate in intermediate regions as well. The simplest of these is the Variable-Eddington method developed by Freeman¹⁶ and his associates. Even better, albeit more expensive, are the Monte Carlo methods pioneered by Fleck.¹⁷ We have not pursued these for several reasons, however. First, no reliable opacity data are available for lithium in the near-liquid or two-phase liquid-vapor regions; and the employment of more sophisticated transport algorithms is not likely to produce more accurate results without these data. Second is the assumption that the flow is strictly one-dimensional. This assumption is especially questionable in the implosion phase where, even if the inner fall surface was initially quite smooth, Rayleigh-Taylor instabilities would almost certainly develop. Given that close-packed jets constitute the actual geometry, so that a top view would show a scalloped inner surface, the energy density at implosion time is likely to be much less concentrated near the axis. This would tend to lower and to equalize the core temperature and density, and favor a diffusion-like transport of radiation. In any case, it does not seem reasonable to ignore deviations from one-dimensional geometry if a very accurate solution is required.

The diffusion approximation in one-dimensional cylindrical symmetry seems to represent a worst-case scenario and is therefore useful for preliminary designs of the reaction chamber. The initial conditions for the LMW motion are based on the energy deposition from the D-T pellet in the plane passing through the pellet's equator and normal to the axis of the fall. The energy, of course, falls off with increasing distance from this plane so that axial gradients exist from the beginning. The radial energy and momentum flow in practice are thus certain to be less than the one-dimensional calculations predict.

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38. The x-ray and debris-induced blowoff from the inner jets and the resultant lithium plasma generation near the chamber axis are completed within tens of microseconds (see the preceding article). Under these conditions, the main impulse is imparted to the jet array by the expanding plasma within, at most, several hundred microseconds. During this period, axial, and circumferential motion can safely be neglected so that, to first approximation, the problem reduces to one-dimensional motion with cylindrical symmetry.

Outward Drag on the Jet Array by Expanding Lithium Plasma

The Duct Analogy.³⁸ The x-ray and debris-induced blowoff from the inner jets and the resultant lithium plasma generation near the chamber axis are completed within tens of microseconds (see the preceding article). Under these conditions, the main impulse is imparted to the jet array by the expanding plasma within, at most, several hundred microseconds. During this period, axial, and circumferential motion can safely be neglected so that, to first approximation, the problem reduces to one-dimensional motion with cylindrical symmetry.

Initially, the x-ray-induced blowoff from the jet array is primarily directed inward; the flow collapses on the cylinder axis with leading-edge velocities that may exceed 100 km/s. The collapsing flow absorbs the debris. The hot, high-pressure plasma created by the implosion then expands against the jets. Until the first jets are contacted, the flow cross section varies linearly with the radius. The same is true after the plasma breaks clear of the last jets. In these regions, a homeomorphic mapping exists to one-dimensional flow between two intersecting planes that enclose an acute angle. There are one-to-one correspondences between the position vector describing the axial distance from the intersection and the reactor-cavity radius vector, and between the distance separating the two planes and the free-flow cross section. A similar analogy can be made within the packed columnar array, where the flow area is constricted by the presence of the jets. In this case, the inclined surfaces are not planes,

Fig. 8-25. Duct representation of the HYLIFE free-flow cross section depicted as a function of the chamber radius a : the equatorial plane passing through the target pellet.

Fig. 8-26. Cumulative jet perimeter as a function of chamber radius.

however, but wavy walls. To illustrate, Fig. 8-25 shows the "duct" representation of the HYLIFE midplane, neglecting the asymmetry along the axis of the beam path.

We computed the free-flow cross section in the region containing the jets by tracing a series of concentric circles, each 10 mm apart, beginning at a radius of 0.4 m (just inside the innermost row of

jets), and ending at a radius of 2.71 m (just beyond the outermost jets). At each radius, the aggregate are length not intercepted by a portion of any jet is the free-flow cross section. The cumulative jet surface was obtained by sweeping outward along a radius vector from the innermost jets. If there are n jets in the array, the surface function varies from 0 at $r = R_0$ to πD (where D is the jet diameter) at $r = R_1$. Figure 8-26 shows how the surface function behaves in the HYLIFE jet array.

The equations of motion describing the flow in such a variable area duct are given in Ref. 38 for a moving coordinate system with velocity, s . The equations reduce to an Eulerian representation when $s = 0$ and a Lagrangian representation when $s = u$, the particle velocity. For numerical solution, it is often convenient to "optimize" the motion of the coordinate mesh as would be the case, for example, when high resolution about a moving discontinuity is required. In general, s equals neither u nor 0. Although the computer code, AFTONCF, was written to allow a completely arbitrary grid-motion algorithm, a Lagrangian formulation was found to give satisfactory results in solving the duct flow of interest here.

The impulse delivered to the jets is caused partly by skin friction and partly by form or profile drag. The former is the time integral of all shearing stresses taken over the surface of the jets, whereas the latter derives from normal stress or pressure. Even a simplified Navier-Stokes representation of the turbulent shear-interaction region between the jets is proscribed by the one-dimensional-flow restriction. Instead, it is assumed that the main effect of viscosity is to slow the mean flow and can therefore be simulated by reducing the pressure gradient in the direction of motion. Quasi-steady parallel-flow theory^{38,39} was used for this purpose.

The specific-internal energy consists of the sum of the material specific energy and the radiation specific energy. The latter is important only near the cavity axis where the plasma implosion gives rise to very high temperature and low density; in this region, the matter energy may constitute less than half of the total. Under these conditions, radiation transport cannot be neglected. We employ the diffusion approximation in our formulation. In the jet region, the radiation energy is much less than the

matter energy; and although it is not necessarily true that the radiation-flux term is negligible, it is far less important than in the central cavity.

Numerical Solution. Numerical solution of the one-dimensional equations of motion in a generalized coordinate system has been discussed at length in Refs. 40 and 41. Extension of the method to include flow in a variable-area duct has also been accomplished in examining the fluid mechanics of the impulsive-water cannon.⁴²

In simple one-dimensional flows, the rate at which volume is swept out by a fluid surface is exactly determined by the inner product of that surface with the particle velocity. The surface is a single-valued function of position and conservation of space is assured when an appropriate time-averaged position is used, depending on whether the symmetry is linear, cylindrical, or spherical. In a duct with arbitrary shape, the cross section cannot be based on any average position coordinate; overlap errors will lead to severe numerical instabilities if this is done, especially near inflection points or discontinuities. Reference 38 shows how to avoid this difficulty for both Lagrangian and non-Lagrangian coordinate frames.

In this calculation, radiation transport was only allowed in the central cavity; heat transfer from the plasma to the jets was excluded. Cursory analysis indicates that convective cooling, at least, is of little importance in the time period in which the main impulse is delivered to the jets. Radiative cooling, on the other hand, likely cannot be ignored; subsequent calculations are planned to explore its influence. The problem is complicated by disassembly and particulation via isochoric heating (resulting from the neutron deposition) and by lithium-transport-property uncertainties in the two-phase region.

The implicit, semiempirical universal-resistance law, derived by Prandtl⁴³ for smooth pipes at very large Reynolds numbers, was used to estimate the friction coefficient, C_f , in the jet region $R_0 \leq r \leq R_1$; for all other r , $C_f \equiv 0$. Within the jet array, an equivalent diameter was calculated based on the local-flow cross section and jet-contact surface. Ignoring Mach number effects in all likelihood introduces a very substantial error; indeed, it may be the case that Mach number effects predominate

Fig. 8-27. Initial pressure in the cavity (at 60 μ s) before contact with the jet array.

Fig. 8-28. Initial density in the cavity (at 60 μ s) before contact with the jet array.

over those associated with the Reynolds number.⁴⁴ It will be seen below, however, that form drag so dominates skin friction that a large uncertainty in C_f can be tolerated.

Our initial conditions derive from the preceding article. The leading edge of the outward moving plasma arrived at the inner jets in just over 60 μ s. The pressure, density, temperature, and particle velocity in the inner cavity are shown in Figs.

Fig. 8-29. Initial temperature in the cavity (at 60 μ s) before contact with the jet array.

Fig. 8-30. Initial particle velocity in the cavity (at 60 μ s) before contact with the jet array.

8-27 to 8-30. The sharp jump in density and concomitant drop in temperature are connected with the earlier radiation flow.

For this problem, 214 Lagrangian zones defined the plasma, the leading edge of which was initially (at 60 μ s) assumed to be a free surface ($P = 0$) located at $r = 0.43 \text{ m} \approx R_0$. Zonal thickness ranged from a minimum of 0.3 mm to a maximum

of 21 mm at the axis. When the leading edge of the plasma made contact with the cavity wall at $r = R_2 \approx 5.0 \text{ m}$, the outer boundary condition was switched from free surface to rigid wall ($u = 0$)—the condition used throughout at the inner boundary on the cylinder axis.

Results. Figure 8-31 shows an overview of the calculation. Each pie segment displays the computational mesh at a discrete time. On entering the field of jets, the plasma is accelerated by the convergence effect. The average velocity of the leading edge in the period $60 < t < 100 \mu\text{s}$ is 17 km/s; the velocity increases to 25 km/s in the next 50 μs interval. Thereafter, once free of the flow constriction, the acceleration is very much reduced. At 250 μs , the leading edge is traveling at 26.4 km/s just before contact with the cavity wall. The main mass of fluid is indicated by the dark band in each pie segment, which is observed to have almost cleared the jet region at 350 μs . The mesh distribution behind this band illustrates the alternating regions of flow compression and expansion as the plasma moves through each "row" of jets.

Figure 8-32 shows that the period of momentum exchange between the plasma and jets is virtually terminated at approximately 350 μs . The total impulse, I_T , includes contributions from form drag and skin friction. The impulse imparted by form drag is defined as:

$$I_f = \iint P dA_j dt \quad (1)$$

where $A_j = 2\pi r - A(r)$.

Figure 8-33 depicts the impulse due to form drag through 360 μs when the calculation was terminated. Although the jets were not arranged in concentric rows, their distribution was highly correlated with discrete radial positions. Thus it is not surprising that the spatial distribution of impulse retains this discrete character.

The impulse caused by viscous drag is defined by

$$I_v = \iint C_f \rho u |u| d\phi dt \quad (2)$$

Fig. 8-31. Fluid motion as a function of time. Each pie segment displays the Lagrangian computational mesh at a discrete time.

where ρ and u are the plasma density and velocity, respectively, and ϕ is the jet surface function. The viscous-drag impulse is plotted in Fig. 8-34, through $360 \mu\text{s}$. There are no negative contributions to this integral in the time frame of interest since the particle velocity throughout was directed radially outward. The most important point to note when comparing Figs. 8-33 and 8-34 is the scale of the ordinate, which is three orders of magnitude larger in Fig. 8-33. This means that the overwhelming contribution to the momentum exchange between plasma and jets derives from form drag and that, as

previously noted, even gross errors in the estimate of the friction coefficient have little or no effect.

The impulse imparted to the jet array is by no means uniformly distributed. Figure 8-33 shows that roughly two-thirds of the total is associated with the first "row" of jets, which constitutes only approximately 4% of the total mass. The second row, with approximately 6% of the jet mass, receives approximately 8% of the impulse; and the other seven rows share the balance. The average-velocity increment resulting from the drag (obtained by dividing the impulse imparted to a row by the jet mass contained therein) on the first row is 210 m/s ,

Fig. 8-32. Total impulse (form drag and skin friction) imparted by the vapor to the jets, I_T , as a function of time normalized to the peak value at $360 \mu\text{s}$, I_T^* .

Fig. 8-33. Radial distribution of impulse caused by form drag at $t = 360 \mu\text{s}$.

but only 17 m/s for row number two and correspondingly less for the remaining rows. The average velocity increment imparted to all the jets (obtained by dividing the total impulse by the total jet mass) is just under 14 m/s for this configuration. This gives a crude measure of the specific effect of drag since the lithium will eventually impact the wall not as a discrete series of jets; but as a continuous annulus of fluid in which momentum

Fig. 8-34. Radial distribution of impulse caused by skin friction at $t = 360 \mu\text{s}$.

gradients have been smeared by multiple particle impacts. A still better measure is obtained by superposing the drag-impulse distribution on the overall LMW disassembly problem, in which short-range spall and long-range isochoric heating are also modeled. When this was done,⁴⁵ the drag contribution for this jet configuration was approximately 20% of the total impact velocity.

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Neutron-Induced Specific Energy Deposition at the Chamber Midplane

Introduction. A major factor determining the hydrodynamic response of the LMW is the neutron-energy deposition. In this article, we describe the Monte Carlo⁴⁶ neutronics calculations used to provide input to the hydrodynamic calculations. We also describe an empirical relationship that predicts the midplane neutron-induced specific-energy-deposition profile as a function of the array location (minimum distance from the pellet) and packing fraction for an array with an effective thickness of 1 m. (Effective thickness equals the packing fraction times the actual thickness.)

The Neutronics Model. One sixth of the hexagonal array of jets modeled is shown in Fig. 8-35. The dots indicate the center of the jet positions at the chamber midplane (i.e., the plane of the fusion pellet). We are particularly interested in conditions along the chamber midplane since the maximum specific-energy deposition occurs here. This, then, will give a worst-case calculation for the hydrodynamic response. The jet diameter is 0.20 m at the midplane and the pitch is 0.27 m, giving a 50% packing fraction. The total number of jets is 300. No deviation from a standard close-packed-hexagonal array is made to allow for beam paths.

The array of discrete jets is modeled as a series of concentric right-circular-annuli 8-m high. There are nine annuli of liquid lithium separated by annuli of lithium vapor as shown in Fig. 8-36. Each liquid-density annulus represents one of the hexagonal "rows" of jets in Fig. 8-35. Each of these zones is divided into three segments by horizontal boundaries at ± 0.5 m referenced to the midplane. This gives top (+4.0 to +0.5), center (+0.5 to -0.5), and bottom (-0.5 to -4.0) sections of each annular zone. Thus there are 51 zones (27 liquid, 24 vapor) representing the jet array. This configuration allows us to examine the row-by-row specific-energy deposition, and its variation with vertical position.

Figure 8-37 shows this vertical division of the annular zones and the other zones that are included to give a reasonable representation of the HYLIFE chamber, particularly in the radial direction. A 14.1-MeV-neutron source was uniformly distributed throughout a D-T zone compressed to $\rho R = 3.0$ g/cm². This results in ~ 2.1 MeV deposited within the pellet, and thus corresponds to a neutron-energy fraction of 68% (12.0/17.6).

The radius to the center of each liquid-rich annulus is set equal to the average radius of the jet in the corresponding hexagonal row. The inner and outer radii are then set such that the volume (and mass) of each annulus is equal to the volume (and

Fig. 8-35. Position of jet centers (one-sixth of jet array).

Fig. 8-36. "Pie slice" of lithium zones at midplane.

mass) of the jets in the corresponding hexagonal row. The volume of the i th liquid annulus is thus,

$$V_i = V_1 (N_i/300)$$

where V_1 = total lithium volume in the jet array and N_i = number of jets in the i th hexagonal row.

Fig. 8-37. Neutronics model.

Since this neutronics model represents an array of vertically injected jets, the volume was calculated on that basis. Vertical injection requires a nozzle with a large ratio of flow area to total area; 85% was assumed in this case. To obtain a 50% packing fraction at the midplane of the 8-m chamber, the jets must be injected at 6.44 m/s. This results in a chamber transit time of 0.78 s and midplane velocity of 10.95 m/s. Since we specified 300 jets with 0.2-m diameters at the midplane, the total flow rate is $103.2 \text{ m}^3/\text{s}$. The volume of lithium in the jet array is then $(103.2 \text{ m}^3/\text{s})(0.78 \text{ s}) = 80.5 \text{ m}^3$.

Table 8-1 lists the number of jets per hexagonal row, the lithium volume per row, the average radius to the jets in each row, the inner and outer radii of the corresponding annular zone in the neutronics model, and the thickness of each annulus. Since the thicknesses of the annuli are constant over the length of the chamber in the model, we underestimate the actual amount of lithium in the top section by 7.5 m^3 , and overestimate the amount in the center and bottom sections by 0.7 and 6.8 m^3 , respectively. This is not expected to significantly affect the results, particularly those for the center section that is used in the hydrodynamics calculations.

Neutronic Results. Table 8-2 lists the resulting specific-energy deposition for the three zones of each annulus and for each annulus as a whole. The total energy deposited in each annulus is also given. The specific-energy deposition and total-energy deposition are based on a 2700-MJ-yield pellet, an array inner radius of 0.4 m, and an array packing fraction of 50%. The actual array packing fraction is slightly lower than the packing fraction of the model since changing the jets to annuli moves the

Table 8-1. Parameters of jet array and neutronics model.

Row	N (jets/row)	Vol (m^3)	Avg radius (m)	Inner radius (m)	Outer radius (m)	Thickness (m)
1	12	3.22	0.5027	0.4390	0.5664	0.1274
2	18	4.83	0.7445	0.6800	0.8090	0.1290
3	24	6.44	0.9883	0.9235	1.0531	0.1296
4	30	8.05	1.2328	1.1678	1.2978	0.1300
5	36	9.66	1.4777	1.4127	1.5427	0.1300
6	42	11.27	1.7228	1.6577	1.7879	0.1302
7	48	12.88	1.9681	1.9030	2.0332	0.1302
8	54	14.49	2.2134	2.1483	2.2785	0.1302
9	36	9.66	2.4038	2.3638	2.4438	0.0800
	300	80.50				

Table 8-2. Neutron energy-deposition; yield = 2700 MJ; inner array radius = 0.4 m; array packing fraction = 50%.

Row	Zone	By zone	By row	E-Total (MJ)
		E/m (kJ/kg)	E/m (kJ/kg)	
1	Top	202		580
	Center	1548	370	
	Bottom	200		
2	T	106		423
	C	695	180	
	B	107		
3	T	65		318
	C	349	101	
	B	67		
4	T	42		235
	C	189	60	
	B	41		
5	T	27		176
	C	109	37	
	B	27		
6	T	18		130
	C	64	24	
	B	18		
7	T	12		97
	C	39	16	
	B	12		
8	T	8		70
	C	22	10	
	B	8		
9	T	6		32
	C	15	7	
	B	6		
				2060

Table 8-3. Energy density in central section from neutron heating; yield = 2700 MJ.

Row	Energy density (kJ/kg)			
	±0.5 m	±0.2 m	±0.1 m	±0.05 m
1	1548	1856	1930	1974
2	695	771	794	810
3	349	373	377	371
4	189	196	198	197
5	109	111	111	113
6	64	66	65	66
7	39	38	37	37
8	22	23	23	22
9	15	16	16	16

array inner and outer radii together slightly; the model packing fraction is 55.5% and the model inner radius is 0.439 m for this geometry.

The highest energy density is ~1550 kJ/kg; it occurs in the center section of the innermost annulus. This drops by two orders of magnitude to 15 kJ/kg in the center section of the outermost an-

Fig. 8-38. Specific energy deposition as a function of central zone height for the inner two jet rows. Inner row average radius = 0.5 m and array packing fraction = 50% were used for a 2700-MJ target.

nulus. For the innermost annulus, the energy density is nearly eight times higher in the center section than the top or bottom sections. For the outermost annulus there is less than a factor of three difference. The average-energy density for the entire array is 53 kJ/kg; the total neutron energy deposited in the nine lithium annuli is 2060 MJ.

Since the variation in the energy deposition over the length of the inner rows was so great, we predicted that the 1.0-m height of the central section was too large, resulting in a significant underestimate of the midplane condition. Three additional two-dimensional neutronic calculations with 0.4-, 0.2-, and 0.1-m-wide central bands were done to determine the trend in specific-energy density as the bandwidth approaches zero. The results are compared in Table 8-3. As expected, the energy density in the innermost row increases with decreasing bandwidth, while the results for the outer zones are relatively constant. Remember that these are the average-energy densities for zones that are 0.13 m thick in the radial dimension.

Figure 8-38 is a plot of the specific-energy deposition as a function of the height of the central band for the first two rows. Extrapolating to a zero-thickness central band gives 2030 kJ/kg for row 1 and 820 kJ/kg for row 2. These extrapolated values for rows 1 and 2 and the results of the calculation with a central section width of 0.1 m for rows 3 through 9 were used as input for the hydrodynamic calculations.

Fig. 8-39. Neutron-energy-deposition profiles for the 0.1-m midplane section of four arrays.

Scaling Law for Other Array Configurations.

We performed several additional two-dimensional neutronics calculations to investigate the variation of the energy-deposition profile as a function of the location and packing fraction of the jet array. The results of these calculations were used to develop an empirical relationship to describe the neutron-induced, specific-energy deposition profile as a function of these parameters.

To investigate the variation with the position of the array, three calculations were made in which the radial position of the annuli listed in Table 8-1 were

increased by 0.5, 1.0, and 1.5 m. The total thickness of the blanket region was held constant at 2.0 m and the height of the midplane section was 0.1 m in all cases. The resulting specific-energy deposition profiles are shown in Fig. 8-39. Note that except for the smallest inner radius ($R_i = 0.439$ m), the energy deposition essentially shows a simple exponential decline through the array. We also ran calculations with a varying array packing fraction. The expres-

Fig. 8-40. Empirical equation fit to TARTNP data for a solid annulus with inner radius = 1.939 m. Data points are TARTNP data; solid line is the prediction.

Fig. 8-41. Empirical equation fit to TARTNP data for a 55.5%-packing-fraction array with inner radius = 0.439 m. Data points are TARTNP data; solid line is the prediction.

sion we formulated based on the results of these calculations is

$$\frac{E}{m} = 1660 E_{TN} \left(\frac{1}{R_i + 2S} \right)^2 \exp \left\{ \left[0.035 \left(\frac{\rho}{\rho_0} \right)^{0.6} - 3.7 \times 10^{-5} R_i \right] (25 - x) \right\}, \quad (3)$$

where $x = (R - R_i)$ = the depth into the array at the

Fig. 8-42. Empirical equation fit to TARTNP data for a 41.2%-packing-fraction array with inner radius = 1.939 m. Data points are TARTNP data; solid line is the prediction.

midplane, cm; R_i = the array inner radius, cm; (ρ/ρ_0) = the array packing fraction; and E_{TN} = the total fusion-energy yield, MJ. The form of the equation is based on the following observed results.

- The peak specific-energy deposition for a given fusion-energy yield scales as R_i^{-2} where R_i is greater than the inner radius of the array.

- The specific-energy deposition decreases exponentially with the distance from the inner radius of the array, x . (This pure exponential dependence is somewhat perturbed for small values of R_i .)

- The coefficient of x in the exponential does not scale directly with the packing fraction; that is, for a given array location, the slopes of the deposition profiles are not directly proportional to the packing fraction as might be expected.

- The coefficient of x in the exponential is a function of both the packing fraction and the location of the array, R_i ; that is, for a given packing fraction, a semilog plot of the deposition profile shows straight lines with slopes that are not equal. The slopes decrease with increasing R_i , indicating more energy is deposited in the outer zones. This geometric effect is discussed in Ref. 47.

Three examples of how the expression fits the results of the neutronics calculations are shown in Figs. 8-40 to 8-42. The fit is close enough to be used as input for hydrodynamic calculations of alternative array configurations within certain constraints. Note that this expression breaks down for

small values of (ρ/ρ_0) . Thus for

$$(\rho/\rho_0) \leq 1.1 \times 10^{-5} R_i^{5/3} \quad (4)$$

the expression predicts an exponentially increasing function, a physical impossibility. The expression should only be used for $(\rho/\rho_0) > 0.4$, and $R_i > 0.4$ m. Its accuracy improves with both the increasing packing fraction and array radius.

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Annular-Fall Response to a 750-MJ Fusion Pulse

In this article, we describe the initial calculations of the response to a 750-MJ fusion pulse of a 60-cm-thick annular fall with an inner radius of 2 m. The results from these calculations led us to divide the LMW into 300 jets (HYLIFE).

Most important aspects of the fall response are completed in the time required for an acoustic wave to traverse the fall thickness. For a 60-cm-thick fall, this amounts to slightly over 130 μ s, during which the vertical motion may safely be neglected. The most severe interaction is at the horizontal plane intersecting the equator of the pellet. In our (conservative) calculations, we use a cylindrically symmetric system with no axial relief. Figure 8-43 schematically depicts the initial-value problem. Both the inner and outer surfaces of the annular fall are assumed to be free. After energy deposition, the inner surface will implode on the cylinder axis and the outer surface will contact the first wall, which, insofar as the fluid is concerned, remains rigid.

Numerical Model. The lithium was treated as an inviscid compressible fluid. Account was taken of the liquid-vapor phase change and of ionization of the gas. A detailed account of the lithium equation of state is given in "Analytic Lithium Equation of State" later in this section. We used the AFTON code,^{48,49} which allows the conservation equations to be numerically integrated on any convenient

Fig. 8-43. Schematic representation of fall disassembly as a one-dimensional initial-value problem with cylindrical symmetry. Inset illustrates subsequent motion.

mesh rather than one that is embedded in the material or fixed in space. For this problem, accurate resolution of the short-range deposition required extremely fine zoning initially at the inner radius; the first-zone thickness was $\sim 0.3 \mu$ m. However, it was necessary to extend the mesh over the entire annulus to simultaneously account for the long-range deposition. The strategy was to satisfy the initial conditions on a mesh, each successive radially increasing element of which was 2% wider than its predecessor. With this geometric progression and with the use of 500 zones, the initial thickness of the last zone at the outer radius was ~ 13 mm. Both inner and outer fall radii were taken to be Lagrangian surfaces so that no lithium was convected across these. The mesh lines in between were gradually allowed to move towards an equal spacing as the motion progressed. In this way, the shock zone that developed from the short-range deposition was always contained in an "optimally" fine-mesh structure as it traversed the fall annulus; and the cost of the calculation was low. A typical disassembly calculation used ~ 5 min of machine time on our CDC 7600 computer.

Fig. 8-44. Typical neutron-energy-deposition profile at the pellet plane for a 750-MJ-fusion pulse.

A variant on this scheme allowed slightly better resolution of the traveling pulse to be achieved and helped minimize the aliasing error.⁴⁹ An artificial Lagrangian interface was inserted so that, together with the Lagrangian inner radius, an initially thin layer was enclosed in which all the short-range deposition was included. All fluid contained within this layer was free to move in Lagrangian fashion; the "accordion" rezone described above was only applied between the artificial interface and the outer radius. Whenever a fluid zone on the outer side of the artificial interface expanded (nominally 10%) below normal density, the interface counter was incremented to include the expanded material within the Lagrangian core region. With this dynamic rezoning technique we were able to maintain the minimum resolution of the shock pulse, as it arrived

Fig. 8-45. Spatial-velocity profile at spall time (133 μ s). The dashed curve is the result from long-range penetration only. The solid curve represents combined long- and short-range penetration. The curves are truncated at $R = R_0$; peak gas velocity in the core region ($R < R_0$) at this time is much higher.

at the outer radius of the annulus, to approximately 1 mm.

Fall Disassembly. If the neutron-energy-deposition profile shown in Fig. 8-44 is assumed to instantaneously apply to the annular-lithium fall, the fall is then isochorically heated; the pressure generated ranges from a maximum of 21 MPa at the inner radius to approximately 3 MPa at the outer radius. (In the one-dimensional neutronics calculation that included an outer graphite reflector, the energy deposition in the fall was 88% complete at 1 μ s and 96% complete at 5 μ s, which is effectively instantaneous compared with the acoustic wave traversal time.) Since both inner and outer radii are free surfaces, rarefactions centered at these locations then move into the fall interior. The relief wave from the inner radius is of sufficient strength to cavitate the fluid behind it; the jump-off velocity at the inner surface is 9 m/s. The outer part of the fall does not immediately cavitate, since it is influenced by the initially positive pressure gradient. The two rarefactions reflect at the geometric midpoint in the fall and the reflected relief wave does finally cavitate the outer portion. The elapsed time is just under 135 μ s, which is the time required for an acoustic wave to traverse the annulus. The fall is now split into two halves, with the inner half moving inward toward the symmetry axis and the outer

Fig. 8-46. Pressure profile in the fall at 1 μ s. The peak pressure (in the pulse) is more than 0.8 GPa.

half moving toward the reactor wall. The peak velocity of this latter segment (at the outer surface) is slightly more than 7 m/s. The velocity profile is shown by the dashed curve in Fig. 8-45.

With the short-range deposition superposed, the motion at the inner-fall surface is much more violent. For realistic penetration layers, the debris mass is insignificant in comparison with that of the lithium: conservation of momentum and energy then suggests that virtually all the debris slap is converted to internal energy of the lithium in the penetration layer. (In reality, the debris arrives considerably later than the x rays and is absorbed in ablated Li plasma that has already decoupled from the fall: neglecting the decoupling is conservative.) The magnitude of the specific-internal energy is then far more than the specific energy of cohesion for lithium (23 MJ/kg). The peak pressure generated is of the order of 100 GPa. A shock moves into the main fall, but is quickly attenuated by a rarefaction centered at the free inner surface. This rarefaction typically traverses the initial penetration layer in less than 1 ns. In so doing, only a small fraction of the short-range energy deposition couples to the main fall (beyond the initial penetration layer); more than 90% is blown back into the inner cavity in the form of lithium-gas kinetic energy.

The energy that couples to the fall goes mainly to heating fluid adjacent to the initial penetration layer and into the traveling pulse. Figure 8-46 shows

Fig. 8-47. Pressure applied to a first (structural) wall located just beyond the fall outer boundary by the outward-moving lithium fall with velocity profile as in Fig. 8-45.

the pressure profile in the fall at 1 μ s; the peak pressure in the pulse has already dropped below 1 GPa. When the pulse arrives at the outer fall radius, the reflection from this surface causes a spall to occur. The velocity distribution at this point is shown as the solid curve in Fig. 8-45. The peak velocity in the spall layer is more than 60 m/s.

Wall Stress in a Wall just Beyond the Fall. The pressure applied to a first structural wall located just beyond the fall ($R_w \sim 2.6$ m) by the outward-moving portion of the lithium fall is shown in Fig. 8-47. This result was obtained by applying a rigid-wall boundary condition to the outer-fall radius at the instant of spall (when the pressure in the outward-moving fall had everywhere vanished).

That a rigid-wall boundary condition is appropriate here may be deduced from the ratio of the mechanical impedance of the actual wall to that of lithium. For 2-1/4 Cr-1 Mo steel, this ratio is more than 17, so that the error in the impact Hugoniot is a maximum of $\sim 5\%$.

As can be seen in Fig. 8-47, the wall loading decreases sharply from the impact pressure of ~ 150 MPa to zero in 7 μ s. The much smaller pressure pulse that begins 15 μ s later, however, persists for a much longer period. The initial spike results mainly from the short-range deposition. The 15- μ s zero-pressure period is caused by the "cavitation tail" behind the spall layer. The fluid contained in this region must be recompressed before significant

Fig. 8-48. First-wall-hoop stress as a function of time from impact given the loading history of Fig. 8-47.

momentum transport to the wall can be effected. The overall loading time of $150 \mu\text{s}$ reflects the $15\text{-}\mu\text{s}$ delay. When subtracted from the total, the difference accounts for the time it takes an acoustic wave originating at the wall to traverse half the original-fluid annulus, reflect at the fall split, and return to the wall.

For an internally pressurized cylinder, neglecting end effects, the equation of motion is

$$PR_w - \sigma_\theta \delta = \rho_w R_w \delta \ddot{u} \quad (5)$$

where $P = P(t)$ is the applied pressure, R_w is the cylinder radius, σ_θ is the hoop stress, δ is the wall thickness, ρ_w is the wall density, and $\ddot{u}$ is the displacement. Equation (5) is strictly applicable to the case where $\delta/R_w \ll 1$, which, for any realistic ICF reactor, will always be satisfied. If we now require linear elastic constitutive behavior,

$$\sigma_\theta = E u / R_w \quad (6)$$

where E is Young's modulus, Eq. (5) reduces to

$$\ddot{u} + \omega^2 u = F / (\rho_w \delta) \quad (7)$$

where $\omega = \sqrt{E / \rho_w R_w^2}$. The natural period of the wall structure is $T \equiv 2\pi / \omega = 2\pi R_w \sqrt{E / \rho_w}$. Equation (7) is subject to the initial conditions:

$$u(0) = \dot{u}(0) = 0 \quad (8)$$

Given $P(t)$, this equation is easily integrated by conventional methods. Figure 8-48 shows the variation of hoop stress with time for the loading history of Fig. 8-47 and an assumed wall thickness of $50 \mu\text{m}$. The peak stress of 114 MPa occurs at just under 1 ms , long after the applied pressure has ceased. The reason for the delay is that the duration of load application, t_* , is very much less than the wall period T . Consider the result when the forcing function takes the Heaviside form:

$$P(t \rho_w \delta) = \begin{cases} F_0 & t < t_* \\ 0 & t \geq t_* \end{cases} \quad (9)$$

The solution is then

$$u = \left(F_0 / \omega^2 \right) \begin{cases} (1 - \cos \omega t) & t < t_* \\ \{ \cos \omega(t - t_*) - \cos \omega t \} & t \geq t_* \end{cases} \quad (10)$$

If t_{**} is defined as the time at which u is a maximum in the period $t > t_*$,

$$(t_{**} - t_*) / t_* = (1 / \omega t_*) \tan^{-1} \{ \sin \omega t_* / (1 - \cos \omega t_*) \} \quad (11)$$

$$u(t_{**}) = (PR / \delta) \sqrt{2(1 - \cos \omega t_*)} \quad (12)$$

and

$$\sigma(t_{**}) / \sigma(t_*) = \sqrt{2(1 - \cos \omega t_*)} \quad (13)$$

Equation (13) shows that the inertial "overshoot" grows without bound as $t_* / T \sim \omega t_* \rightarrow 0$. In particular, for the system defined by the parameters of Fig. 8-48, $t_* \approx 150 \mu\text{s}$, $E = 200 \text{ GPa}$, $\rho_w = 7.8 \text{ Mg m}^{-3}$ and $R_w = 2.6 \text{ m}$ so that $\omega = 1.95 \times 10^3 \text{ s}^{-1}$, $\omega t_* = 0.293$ and $T = 3.23 \text{ ms}$. Substituting into Eq. (11) yields $(t_{**} - t_*) / t_* = 4.88$, which is within 8% of the value of 4.5 measured from the Fig. 8-48 profile (derived not with step-loading, but with the

internal-pressure history in Fig. 8-47). For $\omega t_* = 0.293$, Eq. (13) predicts a stress-amplification factor just under 7, whereas the Fig. 8-47 loading history yields 4.5.

Equation (12) may be simplified when $\omega t_* \ll 1$; in this case $\sqrt{2(1 - \cos \omega t_*)} \rightarrow \omega t_*$; we then have the important result that

$$q_{0, \text{max}} (\omega t_* \ll 1) = (P_t/6) \sqrt{1/\rho_w} \quad (14)$$

Equation (14) indicates that when the loading time is small compared with the natural period of the wall, the maximum hoop stress is proportional not to the applied pressure, but to the impulse. If the "average" pressure required by Eq. (14) is taken as the overall energy density (at the plane of the pellet, including both long- and short-range deposition) in the cavity bounded by the outer-fall radius, and the loading time is assumed to be the acoustic-travel time across the annulus, the predicted peak-hoop stress is just over 110 MPa—within 3% of the Fig. 8-48 value of 114 MPa.

From Eq. (14), we may also estimate the relative influence of the long- and short-range deposition on the peak-hoop stress. Since the impulse is the determining factor, the area under the curve in Fig. 8-47 implies that the relative contributions are more or less equal. This was verified by numerically integrating Eqs. (7) and (8) and suppressing that portion of the pressure history that follows the spike so that the load vanished after the first $7 \mu\text{s}$. The result, as shown in Fig. 8-49, was a reduction of the peak stress from 114 MPa (total) to 48 MPa (from short-range deposition).

In addition to the stress from the liquid impact, there is a stress caused by the high gas pressure in the chamber center that is transmitted through the liquid to the wall. Most of the short-range energy deposition is blown back into the annular core. Virtually all blowback is in the form of gas kinetic energy less than a microsecond after the initial deposition. If the residual liquid-lithium annulus is assumed to bound an adiabatic cavity, all blowback energy must eventually be in the form of gas pressure, which then exerts an outward radial force on the fall. The equilibrium energy density in an 8-m tall, 4-m-diam cavity would be $\sim 2.4 \text{ MPa}$ if all short-range deposition becomes blowback. If we conservatively take the blowback fraction as 0.9 and

Fig. 8-49. First-wall-hoop stress as a function of time from impact given only the spike loading portion of Fig. 8-47 (suppressing the long-range deposition).

assume ideal gas behavior ($\gamma = 5/3$), the equilibrium pressure, P_0 , would be $(5/3 - 1) \times 0.9 \times 2.4 = 1.4 \text{ MPa}$. If there is no heat transfer or axial relief, the pressure will persist, and we find, by Eq. (12), that the dynamic-hoop stress in the wall can attain the limiting value $2P_1 R_w/\delta$, where P_1 is the equivalent pressure at the wall, ($P_1 = P_0 R_0/R_w$). For a 50-mm-thick wall, the peak-hoop stress from gas pressure under the above conditions would be 58 MPa. The total peak-hoop stress is then 172 MPa—approximately three times the allowable stress.

Wall Stress When there is a Gap Between the Wall and Fall. Suppose we relax the assumption that the chamber-wall radius and the outer radius of the lithium annulus are coincident. If the gap is large enough, the inward-moving portion of the fall will be halted and accelerated radially outward by the internal-gas pressure. It will catch and recompact the outward half of the fall, and the resulting slug of liquid will continue to be accelerated until it impacts the wall.

For an isentropic expansion of the internal gas, the impact-Hugoniot pressure is

$$P = \left\{ \frac{3\rho c^2 P_0}{\left(\frac{R_1}{R_0} \right)^2 - 1} \left[1 - \left(\frac{R}{R_0} \right)^{-4/3} \right] \right\}^{1/2} \quad (15)$$

where P_0 = the initial core-gas pressure, r = the inner radius at the time of interest, $\rho = 485 \text{ kg/m}^3$ is the liquid density, and $c = 4.5 \text{ km/s}$ = the liquid sound speed. Equation (15) is not valid for values of r less than the point where the slug is formed. In this intermediate region, the stress is a combination of the impact stress from the outer half of the fall plus the impact stress from the reversed inner half of the fall. At large values of r , the impact pressure approaches an asymptote, $[3P_0c^2\rho_r(r_1/r_0) - 1]^{1/2}$. However, mass conservation requires the fall to foreshorten so that the load application period continues to diminish with increasing wall radius. The wall stress is found by combining Eqs. (14) and (15).

Before evaluating the wall stress, we must determine the appropriate wall thickness as a function of wall radius. The thermal stress and wall temperature is found for the surface-cooled wall by the methods described in "First Wall Design Using Several Procedures and LMW Geometries" later in this section. That article also summarizes the two design criteria as:

$$\sigma_i \leq \frac{1}{2} S_f$$

and

$$\sigma_{th} + \sigma_i \leq S_Y \quad (16)$$

where S_Y and S_f are the yield and fatigue strengths of the steel at its peak temperature, and σ_{th} and σ_i are the thermal and impact stresses. If the impact stress is assumed to the maximum allowable ($1/2 S_f$), the maximum allowable thermal stress is then $S_Y - 1/2 S_f$. The maximum allowable wall thickness is found from the thermal stress; the allowable wall thickness increases directly with wall radius. Thus, in Eq. (14), as we increase the wall radius (or fall-to-wall gap), pressure, time, and inverse-wall-thickness all decrease, and the hoop stress decreases. Unfortunately, a wall radius larger than 25 m is required even if the initial gas pressure is arbitrarily reduced by a factor of ten (Fig. 8-50).

A complicating factor is that the acceleration of the fall by the low-density-core gas is Taylor unstable. Thus the gas will likely penetrate the annulus before the latter can attain its limiting velocity. This would somewhat reduce the impact Hugoniot.

Erosion. In writing Eq. (5), we have tacitly ignored the stress distribution within the wall. Since the wave speed in steel exceeds 5 km/s, one acoustic transit across a 50-mm-thick wall takes less than 10 μs , so that 100 or so transits occur in t_{fall} , the time to reach the peak stress. This is a more than adequate time for the stress within the wall to equalize. Nevertheless, Fig. 8-47 shows that in the first few microseconds, the inner surface of the wall is subjected to an impact pressure of at least 150 MPa (numerical dispersion inherent in this calculation assures us that this is indeed a lower bound). Moreover, if the outer radius of the lithium fall is separated from the wall by a significant distance, the spall layer will not reach the wall as a continuous slug, but rather as liquid fragments or drops. In this case, the local-impact pressure may exceed the one-dimensional Hugoniot value by as much as a factor of three because of the impact geometry.⁵⁰ These localized impacts will be of microsecond or submicrosecond duration so that any individual drop will be of little interest. Over the 30-yr anticipated life, however, there will be on the order of 10^9 fusion pulses, implying at least an equivalent number of droplet impacts. Hancox and Brunton⁵¹ have demonstrated considerable erosion damage to stainless steel by water-drop impacts at 90 m/s after only 10^5 to 10^6 impacts. At high temperature, in the lithium environment, and with 10^3 to 10^4 times as many impacts, we can speculate that the situation will not be improved.

One way of mitigating the erosion damage would be to provide a gap between the main fall and the wall, and to place a quiescent layer of lithium in this gap. The Hugoniot-impact pressure of the spall fragments on the buffer layer would be only half that on the steel wall; and if the buffer layer were several times as thick as the fragments, the rate of momentum transfer to the steel wall could be very much reduced.

Summary. It can be seen from Fig. 8-50 that a 750-MJ fusion pulse produces almost 10 times the ASME-allowable alternating stress ($\sim 60 \text{ MPa}$) in the 8-m-high, 5.2-m-diam, 1.6-cm-thick chamber (see "High-Cycle Fatigue Strength of 2-1/4 Cr-1 Mo Ferritic Steel at Elevated Temperatures" later in this section). Increasing the chamber radius (for the same fall geometry) does not help for reasonable chamber radii and surface-cooled walls. Further,

Fig. 8-50. First-wall-hoop stress as a function of wall radius for a 60-cm-thick fall with a 2-m inner radius. The two curves show the stress when 90% and 9% of the short-range fusion-project energy contributes to the chamber pressure (90% is the more reasonable). The curves are not shown for radii less than the radius required for slug formation; the limiting case of a negligible fall-to-wall gap is shown. Wall thickness is set by thermal stress considerations for each chamber radius; $\delta(\text{cm}) = 0.594 R_w(\text{cm})$. See the article "First Wall Design Using Several Procedures and LMW Geometries" for an explanation of the structural design procedures.

recondensation of the vaporized lithium is not optimized because of the lack of fragmentation of the solid fall. To reduce the wall stresses, enhance vapor condensation, and produce more thermal power in a single chamber, we have proposed segmenting the

annular fall into a large number of discrete jets (the HYLIFE concept).

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Dissipation of Kinetic Energy by the HYLIFE Jet Array

Introduction. Our early fluid-wall designs had the lithium injected as one-continuous-annular fall surrounding the pellet. Since the specific-internal energy from the neutron deposition is typically 10 to 100 times higher at the inner radius than at the outer, this configuration tends to produce steep, positive-velocity gradients in the liquid. This is quite undesirable as it leads to high-speed spall layers; and the first wall is then bombarded with potentially erosive drops or fragments. The situation can be very much improved if the continuous annulus is replaced with a series of concentric annuli, separated by gaps. The reason is that the gaps allow pressure relief from surfaces that are interior to the main fall boundaries. This promotes momentum exchange between fluid elements moving in opposite directions and effectively smears or averages the velocity distribution in the fall. A further design improvement is the HYLIFE geometry in which an annular close-packed array of jets is employed. This allows the hot, high-pressure lithium plasma, deriving from the x-ray deposition and debris interaction, to vent from the cavity formed by the inner radius of the innermost annulus.

Our current state of analysis includes central core pressurization and venting calculations for the HYLIFE-jet array. However, the neutron-induced disassembly calculation has only been performed on the concentric annuli geometry that most closely resembles the HYLIFE jet array. We recognize that the neutron-induced disassembly calculation is conservative since it neglects azimuthal relief in the jet collisions. The theory developed for the neutron-induced disassembly is also applied to calculate the

pressure exerted on the first wall by the impact of the cavitating fluid.

Two Annuli Separated by a Void Gap: The Model. The problem is sketched in Fig. 8-51. At time $t = 0$ two liquid annuli—each of thickness Δ and separated by a void gap of width $\hat{a}$ —are instantaneously heated. We assume for the present that the heating is uniform, so that the initial specific-internal energy within the Lagrangian region $R_0 \leq r \leq R_1$ is e_0 . Furthermore,

$$e_0 \ll e_c \quad (8)$$

where e_c = the cohesive energy of the fluid (2.55 eV = 0.41 MJ/kg for Li). We assume further that the equation of state (EOS) describing the slightly expanded liquid phase is of the Grüneisen form with a Clausius-Clapeyron-like term employed for the two-phase liquid-vapor region (see "Analytic Lithium Equation of State"):

$$P = \text{Max} \left\{ \rho_0 C_0^2 (\eta - 1) + \gamma_0 \rho_0 e, \right. \\ \left. P_c \exp(1.4 - 1.481 e_c / e) \right\} \quad (18)$$

where ρ_0 is the reference liquid density; γ_0 is the Grüneisen parameter; C_0 is the sound speed at the melting point; and the constant $P_c = 100 \text{ MPa}$. P , ρ , and e are the pressure, density, and specific-internal energy, respectively; and $\eta = \rho / \rho_0$. For $\eta \rightarrow 1$, $\gamma_0 e / C_0^2$, Eq. (17) implies that $P \rightarrow 0$ (i.e., the Clapeyron term in Eq. 18(b) behaves like a tensile cutoff). In this case, the EOS is similar to models used to describe porous materials²²; as will be seen, the analogy is quite relevant.

Initially, $\eta = 1$ so that the pressure developed is $P_0 = \gamma_0 \rho_0 e_0$. The inner and outer radii of each annulus are free surfaces from which centered rarefactions then move into the annuli interiors. The free surface particle speed is

$$U_1 \approx P_0 / \rho_0 C_0 = \gamma_0 e_0 / C_0 \quad (19)$$

If we neglect the divergent geometry for the present, U_1 is also the speed along the trailing characteristic (tail of the expansion fan), which is itself moving at speed $C_0 - U_1$ into the fluid. Behind the tail, conditions are everywhere uniform and

Fig. 8-51. Two concentric liquid rings separated by a void gap; suddenly heated at $t = 0$; at $t = t_0$, when the pressure has just released; and at $t \geq t_1$, when collision occurs.

energy conservation requires the specific-internal energy to have decreased to

$$\epsilon_1 = \epsilon_0 - \frac{1}{2} U_1^2 = \epsilon_0 (1 - \gamma_0 M_0 / 2) \quad (20)$$

where we have defined the Mach number parameter

$$M_0 = \gamma_0 \epsilon_0 / c_0^2. \quad (21)$$

The leading characteristic (head of the expansion fan) arrives at the center of each annulus at $t_0 \approx \Delta / 2C_0$, and the pressure is reduced to zero everywhere shortly thereafter (although this may take up to twice as long when the spatial distribution of energy is initially nonuniform). The particle velocity distribution at this point will appear approximately as sketched in Fig. 8-52. In actuality, the interaction of the reflected characteristics at the center of the fall will create a nonsimple wave region that will result in cavitation occurring there. It can be shown,⁵³ however, that the width of the cavitation region [i.e., that region at the center of each annulus in which negative pressures would be generated but for the cutoff in Eq. (18)] is approximately $\Delta(M_0/2)(1 - M_0/2)$. Since Eq. (17) implies $M_0 \ll 1$, we are justified in neglecting this effect.

Each fluid annulus will then begin dividing in half with the outer half moving outward and the inner half inward. The Lagrangian element defined by $R_a \leq r \leq R_b$ will collide with the element defined by $R_c \leq r \leq R_d$ in a time increment $t_1 - t_0 = \delta/2U_1 = \delta C_0 / 2\gamma_0 \epsilon_0$. These two fluid elements are shown

shaded in Fig. 8-51 for $t \geq t_0$; for clarity, the boundaries of the remaining fluid elements are identified with dashed lines. Maximum momentum interaction will be achieved if the collision occurs after pressure relief has been fully accomplished in each segment (i.e., if $t_1 - t_0 > t_0$). This inequality can be rewritten as

$$\delta \geq M_0. \quad (22)$$

Upon collision, shocks will reflect with speed S from the contact interface, which is a line of symmetry in the nondivergent case. The particle velocity and specific-internal energy behind the shock then regain their initial values, i.e.,

$$v_2 = u \quad (23)$$

$$\epsilon_2 = \epsilon_0 \quad (24)$$

Restricting attention to the righthand or outward side of the contact interface, mass and momentum conservation require

$$\rho_1 v_1 S = \rho_2 v_2 S \quad (25)$$

and

$$P_2 + \rho_2 S^2 = \rho_1 (S - v_1)^2, \quad (26)$$

where due account has been taken of the fact that both P_1 and U_2 are zero.

Formal solution of Eqs. (18), (25), and (26) yields

$$v_2 = \lambda v_1 \sqrt{\lambda^2 - 1} \quad (27)$$

and

$$\gamma_2 S (t_0 - P_2/P_0) = \gamma_1 \gamma_2 M_0 v_2 = \gamma_1 v_1. \quad (28)$$

where

$$\lambda = \frac{1}{2} \left[(1 - M_0) + \gamma_1 (1 + M_0^2) \right] \quad (29)$$

and

Fig. 8-52. Spatial distribution of particle velocity at $t = t_0$, when pressure has just released (slab geometry).

$$B = \eta_1(1 - M_0) \quad (30)$$

The quantity η_1 represents the average density of the fluid elements at the moment of the collision (ahead of the shocks). In a linear field, this is readily determined by solving Eq. [18(a)] with $P_1(\eta_1, c_1) = 0$. The result is

$$(\eta_1)_{\max} = 1 - M_0(1 - \gamma_0 M_0/2) \quad (31)$$

The subscript (max) is appended to signify that this is the maximum value η_1 can take if the pressure is to be everywhere zero before collision. We note that $\eta_1 < (\eta_1)_{\max}$ even for linear slab geometry if account is taken of the cavitation developed at the midpoint of each annulus by the reflected rarefaction fan (neglected here because of the assumption that $M_0 \ll 1$). That $\eta_1 < (\eta_1)_{\max}$ for cylindrical or spherical flows is easy to demonstrate. In a divergent flow field, the flow cross section for the radially outward-moving segment continues to increase; and since the pressure is uniformly zero, there can be no compensating acceleration. Then, by continuity, the density must decrease in this segment. The EOS [Eq. (18)] is not symmetric about $(\eta_1)_{\max}$, however, so the converging flow in the radially inward-moving segment will be accelerated with $\eta_1 \approx (\eta_1)_{\max}$ in this segment. At contact, the average density of the ensemble must then be less than the maximum and will decrease with increasing initial separation distance, δ .

Since it is clear on physical grounds that

$$(\eta_1)_{\max} = \eta_2 + 1 \quad (32)$$

Eqs. (26) and (28) can be simplified somewhat by taking the cavitated Hugoniot to be approximately equal to the reference isochore; that is, by setting

$$\eta_2 = 1 \quad (33)$$

so that

$$S/c_0 = P_2/P_0 = \eta_1 M_0/(1 - \eta_1) \quad (34)$$

Fig. 8-53. Relative error in computing the pressure by assuming the Hugoniot to be approximated by the reference isochore.

The relative error in the pressure is then

$$\epsilon = [(P_2)_{\text{exact}} - P_2] / (P_2)_{\text{exact}} \\ = (\eta_1/\eta_2) \{ (1 - \eta_2) / (1 - \eta_1) \} \quad (35)$$

where $(P_2)_{\text{exact}}$ derives from Eq. (28).

The error, ϵ , is plotted as a function of η_1 in Fig. 8-53 with η_2 evaluated from Eqs. (27), (29), and (30), using $M_0 = 10^{-3}$. This is the approximate value of M_0 derived when the total-neutron-energy deposition per unit height (at the horizontal plane passing through the equator of the fusion pellet) is averaged over the lithium fuel mass per unit height. Figure 8-53 shows that the approximation of Eq. (33) results in at most a 20% relative error in the calculation of the collision pressure. Even this result is somewhat misleading because the maximum relative error occurs when the pressure gradient $\partial P / \partial \eta_1$ is very steep. Figure 8-54, which is a plot of the collision pressure as a function of the precollision density, shows that Eqs. (33) and (34) are indeed a very good approximation to the exact solution.

Figure 8-54 also shows that a very small decrease in the precollision density produces a very large decrease in the collision pressure. Cavitating

Fig. 8-54. Collision pressure as a function of precollision density.

the fluid by only 1% below $(\eta_1)_{\max}$ (≈ 0.99 for $M_0 = 10^{-2}$) decreases the shock pressure by almost 40%.

The physical picture may be clarified somewhat by the P-V diagram sketched in Fig. 8-55 ($V = 1/\eta$). Point A corresponds to the initial state attained upon energy deposition [(i.e., $P_A = P_0(V = 1, e = e_0)$]. The fluid is then isentropically expanded along the path $A \rightarrow B + C$. Point B corresponds to the "linear" geometry case [i.e., $P_B = P[\eta_1 = (\eta_1)_{\max}, e = e_1]$]. Collision occurs at point C. The path $C \rightarrow D$ is a Rayleigh line and no intermediate states are possible. Conservation of energy requires that the area $\overline{AEBO}$ equals $\overline{CDO}$; it is therefore clear that P_D , the collision pressure, rapidly decreases as V_C increases. The shaded area $\overline{CEF}$ represents the energy dissipated by the collision.

When the shocks have reached the Lagrangian surfaces (R_A and R_D in Fig. 8-51), they are reflected as rarefactions behind which the particle speed will be

$$U_3 \approx P_2/\rho_2 C_0 \quad (36)$$

The net impulse eventually imparted to the wall is proportional to the total outward-directed momentum $\rho_0 \Delta(U_1 + U_3)/2$. Had the energy been deposited in a singular annular fall without imbedded void gaps, the outward-directed momentum would have been simply $\rho_0 \Delta U_1$. Thus the impulse

Fig. 8-55. Disassembly and collision sketched in the pressure-specific volume plane.

ratio is $(U_1 + U_3)/2U_1$ or, employing Eqs. (33), (34), and (36),

$$1/I_0 = [1 + M_0 \eta_1 (1 - \epsilon_1)/2] \quad (37)$$

If the colliding fluid masses are cavitated by only 1% below $(\eta_1)_{\max}$, the impulse ratio for $M_0 = 10^{-2}$ is 0.75.

Numerical Experiments. Equations (34) and (37) predict the pressure and outward-directed momentum given η_1 , the average density just before collision. For slab geometry and $M_0 \ll 1$, Eq. (31) provides a satisfactory estimate of this quantity. No such simple estimate is evident when the geometry is divergent because of the absence of symmetry about the radius of collision and the inhomogeneity of the colliding fluid packets. In general, η_1 will depend on the geometry (inner annular radius, annular thickness and gap width) as well as on M_0 . On the other hand, numerical solution of the initial value problem posed in Fig. 8-51 by finite difference methods is quite straightforward. Figure 8-56 depicts the results of a number of such calculations made with the AFTON code.⁵³ The impulse ratio, $1/I_0$, is plotted as a function of the ratio of the gap width to thickness ratio, δ/Δ . Two different initial energy deposition functions were used—a "typical" distribution and a uniform distribution. In the former, e_0 varied from ~ 2 MJ/kg at the inner radius (0.5 m in all cases) to ~ 0.02 MJ/kg at the outer

Fig. 8-56. Impulse imparted to the wall as a function of gap-to-annulus thickness ratio. Numerical results are computed for 1 and 9 gaps, and for uniform and nonuniform spatial-energy deposition. The ordinate is the ratio of the gapped-to-ungapped geometry outward momenta; the open circles are the uniform energy deposition; the solid circles are the "typical" neutron-energy-deposition profile.

radius. In the latter, e_0 was constant throughout the fall and equal to the mass-averaged energy derived from the nonuniform case (0.225 MJ/kg , corresponding to $M_0 = 10^{-2}$). For both deposition functions, calculations were first made for a single annular fall ($\delta/\Delta = 0$). The total outward-directed momentum for the nonuniform deposition was 42% higher than for the uniform case (the peak outward velocity at the outer radius was almost five times higher). The ordinate values for the points shown in Fig. 8-56 (open for uniform deposition and shaded for nonuniform) were derived by dividing the total outward-directed momentum obtained with the fall initially split (into $n + 1$ annuli separated by n void gaps; $n = 1, 9$) by the total outward-directed momentum for the single annular geometry and the same initial-energy distribution and total mass. The

multiannulus problems were generated from the single annulus problem after the energy was deposited. This was done for n gaps by dividing the single annulus into $n + 1$ Lagrangian segments and inserting the gaps of desired thickness, beginning with the innermost gap and proceeding outward. After each gap was inserted, the thickness of all succeeding segments and the mesh spacing by which they were delineated were appropriately foreshortened to conserve mass.

Considering first the single-gap calculations, it is remarkable that although the impulse depends very much on the initial spatial energy distribution, the impulse ratio is affected very little; its value is determined strictly by the mass-averaged energy (represented by M_0) and the geometry. Using the

numerical data derived from the calculations at $M_0 = 10^{-2}$, we found that a very good fit was obtained with the empirical expression

$$I/I_0 = \{1 + 1/[1 + (\delta/\Delta)/4M_0]\}^{1/2} \quad (38)$$

as evidenced in Fig. 8-56. To test the M_0 dependence, we increased this parameter by one order of magnitude, to 10^{-1} , and used the code to calculate I_0 (single annular fall) and I (1 void gap, $\delta/\Delta = 0.25$); the ratio I/I_0 so determined was 0.801. This agrees with the value of 0.808 predicted by Eq. (38) to within better than 1%.

If Eqs. (37) and (38) are equated, it is possible to "experimentally" deduce the parameter η_1 for the single gap case

$$\eta_1(R_0 = 0.5 \text{ m}) = [1 + M_0 + (\delta/\Delta)/4]^{-1} \quad (39)$$

For $\delta/\Delta \rightarrow 0$, $\eta_1 \rightarrow (1 + M_0)^{-1} \approx 1 - M_0$ for $M_0 \ll 1$, which is the correct density ratio in this limit.

Figure 8-56 also shows the effect of gap thickness on the impulse ratio when the number of gaps is increased. A much smaller relative gap spacing is required to effect a given impulse reduction when the number of gaps is increased from 1 to 9. Also, very little is to be gained by increasing the void to annulus thickness ratio above 0.25 when $M_0 = 10^{-2}$. Figure 8-57, which is a cross plot of Fig. 8-56, shows that little is to be gained by splitting the fall into more than 10 or so concentric annular segments.

Multiannular Fall Disassembly in a 1000-MW_e Reactor. Figure 8-58 shows the calculated mass-averaged outward-directed velocity, $\bar{U}$, as a function of time for two different liquid-metal-wall designs in a HYLIFE reactor; the ordinate was obtained by dividing the total outward-directed momentum by the outward-moving fluid mass.

In the design labeled "actual," 300 jets (each of 200-mm diam) arrayed around the D-T pellet in a hexagonal close-packed annulus, were simulated by nine concentric annuli separated by void gaps. The innermost annulus was located at a radius of 0.5 m and was associated with the first 12 jets so that it contained 4% of the total jet mass. The second annulus had 6% of the mass, the third 8%, and so on

Fig. 8-57. Impulse imparted to the wall as a function of the gap number for different gap-to-annulus thickness ratios and an impact Mach number of 10^{-2} (cross plot of the data in Fig. 8-56).

Fig. 8-58. Calculated mass-averaged outward-directed fall velocity as a function of time for two different fall designs in a 1000-MW_e ICF reactor. The "actual design" locates annular rings in the region of the jet array. The "minimum gap design" reduces the spacing between the rings to reduce numerical errors and run time.

up until the eighth, which had 18%. Each of these was separated by a void gap of 118 mm. The outermost annulus contained the remaining 12% of the mass and was located 88 mm from its inner adjacent annulus.

For $t \leq 60 \mu\text{s}$, the problem is dominated by x-ray deposition and ionic debris from the D-T pellet, which penetrate only a very thin layer in the innermost exposed jets (inner annulus). The details of the resulting implosion and blowback are discussed in

Fig. 8-59. Spatial distribution of particle velocity and density (shaded) for the "actual" design of Fig. 8-58 at $t = 60 \mu s$.

"Central Region Pressurization Due to X-Ray- and Debris-Generated Lithium Blowoff" and "Outward Drag on the Jet Array Expanding Lithium Plasma" earlier in this section. In brief, a hot, high-pressure lithium plasma is created in the core that then expands until contact is made with the first wall (located two or more meters beyond the last row of jets). Coupled with the jet disassembly induced by x-ray spall and neutron heating, this results in a complicated three-dimensional flow. A quasi-one-dimensional method was, however, devised to calculate a first approximation of the drag impulse exerted on the jet array by the venting plasma. This impulse, which was effectively terminated by $t \approx 350 \mu s$, was then superimposed on the radial motion

of the concentric annuli caused by the isochoric heating. The object was to obtain a reasonable estimate of the impulse imparted to the first wall by the lithium.

For the "actual" design, the spatial distributions of particle velocity and density are shown in Fig. 8-59 at $t = 60 \mu s$, when the plasma first arrived at the innermost annulus. An Eulerian inner boundary was imposed at $r = 0.5 m$, since the cavity region was calculated separately to determine the blowback and cross flow. All other annular boundaries were Lagrangian until gap closure was attained, after which a gradual, accordion-like⁵⁴ rezone was employed. It can be seen that at $60 \mu s$

Fig. 8-60. Spatial distribution of particle velocity and density (shaded) for the design "actual" of Fig. 8-58 at $t = 1$ ms.

the fast-moving x-ray-spall layer from the first "row" has already made contact with the second row. The peak outward-directed velocity at this time exceeds 2 km/s. Also noteworthy in Fig. 8-59 is the splitting evidenced in the remaining annuli by the nonuniform neutron heating (the fluid density is shown by the shaded overlay; the reference liquid density was 0.518 Mg/m^3).

By $t = 1$ ms, Fig. 8-58 shows that the average outward-directed velocity has decreased to approximately 52 m/s. Figure 8-60 illustrates the velocity and density distribution. Contact has been made by

all but the last three annuli. By $t = 5$ ms, $\bar{U}$ in Fig. 8-58 has dropped to its minimum value of 36 m/s, and Fig. 8-61 shows that all of the fluid annuli have joined. The outward-moving fluid has begun to compact, a process that is essentially completed 10 ms later as seen in Fig. 8-62. At this point, virtually the entire fluid mass (at the plane of the D-1 pellet) is at near-liquid density and is moving outward at a uniform velocity of 38 m/s.

Numerical Error. Most practical schemes for the solution of arbitrary initial value problems in continuum mechanics are dispersive (i.e., if the solution is thought of as being expanded in Fourier series with time-dependent coefficients, different

Fig. 8-61. Spatial distribution of particle velocity and density (shaded) for the "actual" design of Fig. 8-58 at $t = 5$ ms.

components travel with different speeds). In particular, components whose wavelengths are of the same order as the spatial mesh discretization interval are always falsified somewhat. In most cases, this is of little interest as long as the amplitude of the falsified components remains small everywhere; a condition that is usually easy to meet with a judicious choice of artificial viscosity and timestep. Unfortunately, however, numerical dispersion may lead to serious error in the isochoric-heating problem. The reason is that the small decreases in the average density that result when square corners are rounded or smeared can cause large decreases in the collision pressure (cf. Fig. 8-54). In numerical experiments performed with just two colliding slabs

(linear geometry) and with $M_0 \ll 1$, so that the actual physical dissipation must be negligible, upwards of 500 zones were required to assure this result. The calculation described in Figs. 8-59 to 8-62 did initially employ 500 zones; but only approximately 300 delineated the liquid and two-phase liquid-vapor region, the other 200 having blown off early (within the first few microseconds) with the plasma into the core cavity. Further, that calculation involved at least $n(n+1)/2 = 36$ collisions in a divergent geometry.

The magnitude of the dissipation (physical and numerical) resulting from the divergent geometry might be determined by minimizing the gap width

Fig. 8-62. Spatial distribution of particle velocity and density (shaded) for the "actual" design of Fig. 8-58 at $t = 15$ ms.

between annuli. Figure 8-56 shows that if the gap width is of the order of M_0 , in accord with Eq. (22), the dissipation is very much reduced without affecting the desirable spatial averaging of momentum. Accordingly, a multiannular fall geometry was set up with the same mass and energy distribution as for the actual design described above, but with much reduced gap spacing. The performance of this "minimum gap" design is also shown in Fig. 8-58. With the gap spacing reduced so that the first collisions occur everywhere nearly simultaneously (and in a time period not much exceeding the time for initial-pressure relief), the average outward-directed velocity is determined much more rapidly than with the actual design. The minimum value of $\bar{U}$ of 41.5 m/s occurs at approximately 300 μ s. By 1 ms, the available potential energy of collision has been reconverted to kinetic energy; and $\bar{U}$ has stabilized at 41.5 m/s. This may be directly compared with the 38 m/s obtained with the actual design, since the identical cross-flow impulse was superimposed on both problems. The 10% difference is probably an acceptable margin of error, considering the crudeness of the model.

There has been some recent progress with the development of numerical methods that are (practically) nondispersive^{55,56}; Keith Miller's moving finite element (MFE) method⁵⁶ appears to be particularly promising in this regard. They have not yet, however, been adapted to problems as complex as those under consideration here. Alternatively, the

magnitude of the numerical dispersion can be estimated by a very finely zoned calculation of the "actual" geometry. This remains to be done.

Impact on the First Wall. Our main concern is that the hoop stress that results from fluid impact does not exceed the maximum allowable working stress. If the first wall were located at 2.3 m (i.e., directly behind the nearly compacted fluid packet in Fig. 8-62) the impact pressure would be $P_3 \approx \rho_0 \bar{U} C_0$. For lithium, $\rho_0 = 0.518 \text{ Mg/m}^3$, $C_0 = 4.5 \text{ km/s}$; and if we conservatively take $\bar{U} = 41.5 \text{ m/s}$, $P_3 = 96.7 \text{ MPa}$. This pressure will persist for a duration $\tau = 2\ell/C_0$, where ℓ is the thickness of the fluid packet (from Fig. 8-62, $\ell = 2.3 - 1.8 = 0.5 \text{ m}$), so that $\tau \approx 0.22 \text{ ms}$. The natural period of the wall is $T = 2\pi R_w / \sqrt{E_w / \rho_w}$, where R_w is the wall radius, E_w is the Young's modulus, and ρ_w is the density. If 2-1/4 Cr-1 Mo steel is used with $R_w = 2.3 \text{ m}$, $T = 3.16 \text{ ms}$ so that $\tau/T \ll 1$. In this case, the peak-hoop stress, $\sigma_{\max} = (P_3 \tau / \delta_w) \sqrt{E_w / \rho_w}$, where δ_w is the wall thickness (see the preceding article for a derivation of this formula). Assuming $\delta_w = 50 \text{ mm}$, the peak-hoop stress under these conditions would be 1.9 GPa—over 30 times the recommended alternating stress for the ferritic steel.

Now suppose the first wall is relocated at 5-m radius. Since the pressure in the outward-moving fluid packet in Fig. 8-62 is everywhere zero, no further acceleration is possible and the packet will coast at the speed U until impact. The speed is uniform throughout so the thickness of the fluid packet will be maintained. But continuity requires that as the field diverges, the density must drop. If the density is $\sim \rho_0$ at $t = 15 \text{ ms}$, impact will occur at $15 + (5 - 2.3) / 0.415 = 80 \text{ ms}$ at which time the density ratio will be $\eta = \rho / \rho_0 = (2.3^2 - 1.8^2) / (5^2 - 4.5^2) = 0.43$. Physically, we expect that cavities will be initially formed within the bulk liquid. If η is sufficiently small, the fluid will appear as a rain of drops or fragments; but this is probably not the case here.

The impact of the cavitated liquid on the (assumed rigid) wall is exactly analogous to the symmetrical liquid impacts described by Eq. (34). Then, $P_3 = \rho_0 \bar{U}^2 \eta / (1 - \eta)$. With $\eta = 0.43$ the impact pressure $P_3 = 0.67 \text{ MPa}$, more than 100 times less than obtained when R_w was 2.3 m, but still $2/(1 - \eta) = 3.5$ times the Bernoulli pressure. Also from Eq. (34), the compaction speed, S , will be $\bar{U} \eta / (1 - \eta) = 31.3 \text{ m/s}$, so that the pressure will persist for a period of $\tau \approx \ell / S + \ell / C_0 \approx \ell / S = 16 \text{ ms}$. And if $R_w = 5 \text{ m}$, $T = 6.9 \text{ ms}$ and $\tau/T > 1$. In this case, the

peak-hoop stress is simply $\sigma_{\max} = 2P_3R_w/\delta_w = 134 \text{ MPa}$.

The 134-MPa-hoop stress is approximately twice the ASME-allowable alternating stress. The peak stress can be decreased to the ASME allowable by doubling the wall thickness to 10 cm and internally cooling the wall to limit the peak temperature. Alternatively, the jet-array inner radius can be somewhat increased to reduce the energy deposition and consequently the wall loading (see "First-Wall Design Using Several Procedures and LMW Geometries" later in this section).

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Design Method for the HYLIFE Reaction Chamber

The design of the HYLIFE reaction chamber is based on the use of standard materials and established design practices. We chose 2-1/4 Cr-1 Mo ferritic steel as the material because of its widespread use in the nuclear industry, good high-temperature properties, resistance to hydrogen (tritium) embrittlement, compatibility with flowing lithium, and low-radiation damage and activation characteristics.

We chose the design method in the American Society of Mechanical Engineers (ASME) Boiler and Pressure Vessel Code because of its worldwide acceptance and conservative approach. Strict compliance with the basic ASME code is not possible because high-temperature, high-fatigue-cycle material property data covering our operating conditions have not been accepted for inclusion in the code. However, ASME Code Case N-47 (1592-10)⁵⁷ documents an ASME-accepted design method,

which, when coupled with Booker's fatigue data,⁵⁸ offers a logical, conservative approach to designing critical HYLIFE components.

The most highly stressed structural component in the HYLIFE reaction chamber is the first structural wall (FSW). Its operating conditions include not only impulsive loads that occur following each fusion reaction, but thermal loads caused by neutron energy deposited throughout the surface-cooled FSW. The FSW is not a containment structure; and it can be argued that compliance with the ASME code is not necessary. For this reason, we have outlined in the following article alternative design methods that may be utilized, should a less-stringent method be desired.

The purpose of the FSW is to isolate the vacuum vessel (and reflector) from impulsive loads that could result in a fatigue failure of the containment structure. The FSW is designed to last the entire 30-yr-expected life of the reaction chamber without replacement (10^9 cycles at a 1-Hz repetition rate). Maximum temperatures up to $\sim 810 \text{ K}$ are reached in FSW.

In August 1979 we documented⁵⁹ a method of including thermal stresses using the ASME Pressure Vessel and Piping Code,⁶⁰ Section III. This method classifies thermal stresses as secondary because they are self-balancing. The code requires that the total stress be less than three times an allowable-design-stress intensity, S_m , when these thermal stresses are added to primary membrane, bending, and local stresses. The allowable-design-stress intensity is tabulated for various materials in the ASME code. An additional requirement is imposed for cyclic operation in that the peak-to-peak sum of all cyclic stresses present must be less than twice an allowable-stress value, S_a , obtained from design fatigue curves. The major difficulty with Section III is that a maximum-temperature limit of 540 K is imposed on both the S_m and S_a values for 2-1/4 Cr-1 Mo ferritic steel. Because our operating temperatures exceed 540 K, we suggested a method for extrapolating S_m to higher temperatures, which was consistent with other sections of the ASME code. Left unresolved at that time was the question of how to determine the value of S_a at higher temperatures.

Hovingh⁶¹ pointed out that the ASME-design method for fatigue should only be used in conjunction with their published design fatigue curves. The

ASME-design-fatigue curves include adjustments from the original data to account for combined mean and cyclic stress, using a modified Goodman diagram. Reference 62 is the basis for the fatigue design method used in the ASME code.

In this article, we describe the relevant HYLIFE operating conditions and a method for determining limiting stresses of HYLIFE components. The method is used with recent high-temperature fatigue data (see also "High-Cycle Fatigue Strength of 2-1/4 Cr-1 Mo Ferritic Steel at Elevated Temperatures") to select the first-structural-wall thickness and the location of the liquid-metal-jet array. Our proposed method for determining limiting stresses follows ASME Code Case N-47 as much as possible. The main deviation is in that we extend Code Case N-47 to include higher temperatures and numbers of fatigue cycles.

Even though ASME Code Case N-47 includes fatigue data to 10^6 cycles at temperatures exceeding 860 K for 304 and 316 stainless steels and Ni-Fe-Cr alloy 800H, there are no high-temperature fatigue data for 2-1/4 Cr-1 Mo ferritic steel included, and in no case, is there data that extend to our needed value of 10^9 cycles. Perhaps this is because of large variations of properties in ferritic steel with various heat treatments, or as Brinkman⁶³ suggests, because of the lack of established fatigue curves used with the ASME elastic cumulative-damage criterion. However, Booker's⁵⁸ continuously cycled-fatigue data extend to ~860 K and are expected to be included in Code Case N-47. His data and the data of Brinkman⁶⁴ and Jaske⁶⁵ are in agreement.

Although one cannot now comply completely with the basic ASME code or Code Case N-47, if 2-1/4 Cr-1 Mo ferritic steel is used at temperatures exceeding 540 K, it should be possible to more closely comply when Booker's data are added to Code Case N-47. Unfortunately, when these data are added, they will probably include only continuous cycling data up to 10^6 cycles. If we are to design the actual HYLIFE FSW with the ASME procedures, fatigue data up to slightly over 10^9 cycles must be gathered for 2-1/4 Cr-1 Mo ferritic steel; and any derating caused by the 775 K lithium environment must also be determined.

HYLIFE Operating Conditions. The operating conditions include a slow warmup lasting several

Fig. 8-63. Variation of the first-structural-wall surface and center temperatures with time.

days, during which the reaction chamber is brought to its nominal temperature of 775 K. A lithium preheater will be required until the temperature exceeds the melting point of lithium (450 K). After this, lithium circulation can be started, and either the preheater or circulation-loop heat exchangers used to reach operating temperature. The rate of temperature rise will be kept low so that thermal stresses will stay within allowable bounds at all locations within the chamber. The necessary vacuum for laser-beam propagation of 13 Pa (0.1 Torr) inside the chamber will be established either during or after the warmup.

Next, the fusion reactions will be initiated at one-second intervals. A startup will be followed by an operating period. The temperature of the FSW surface and center are shown as a function of time in Fig. 8-63. These temperatures are based on a 50-mm-thick FSW, which is cooled on both inner and outer surfaces.^{66,67} Thermal conductivity and a uniform heat generation of 35 W/m² K and 4.0 MW/m³ have been incorporated. The maximum temperature difference between the center and surfaces, and the variation of the center temperature between two fusion reactions, are related to peak thermal stresses in the FSW. The rate of center temperature rise above the surface temperature and the

phase lag between these two temperatures are not particularly important.

After the operating period, the fusion reactions will be stopped and the chamber cooled down slowly, using essentially the reverse of the procedure used for warmup. Shutdowns will be scheduled for maintenance, inspection, repair, and replacement of components in the reaction chamber or elsewhere in the power plant. The frequency of these shutdowns is not known; but for this study, we have considered 100 shutdowns during the 30-yr-chamber life.

The peak-thermal stress^{66,67} from the temperature difference shown in Fig. 8-63 is 86 MPa. This thermal stress is self-equalizing and decreases as the FSW material creeps. A smaller 2-MPa-cyclic thermal stress resulting from the 1-Hz thermal cycles is superposed. There is insufficient time available for creep to have an effect on the smaller, but higher-frequency thermal stress.

Stress in the FSW from lithium impact depends on several factors including the inner radius of the liquid-metal jet array and the FSW thickness. We first used estimates from the previous article for the impact stress that would result if the inner radius of the jet array were 0.4 m. Next we used the scaling relationships discussed in the following article to adjust the FSW stress for varying FSW thickness and jet-array inner radius. This procedure assumes that the momentum dissipation and the relative weighting of neutron and blowthrough-induced momenta do not vary with small changes in jet-array inner radius. Specific values of 50 mm and 550 mm were selected for the FSW thickness and jet-array inner radius to satisfy creep fatigue and other requirements. Calculations of the impact stress are continuing in an attempt to reduce uncertainties.

Variation of the lithium-impact hoop stress with time is shown in Fig. 8-64. The sum of all damping effects is considered to be 2% of critical damping.⁶⁸ The actual magnitude of damping would necessarily need to be determined by experiment; however, Newmark and Hall⁶⁹ consider 2% of critical damping to be a conservative value for welded-steel structures. Expressed as a magnitude decrement, 2% of critical damping is equivalent to a peak-magnitude decay to 88% of its previous value during a single cycle; that is, the peak stress, σ , decays as

$$\frac{\sigma}{\sigma_0} = e^{-\alpha t} \quad (40)$$

where σ_0 is the initial peak stress. The value of α is related to the fraction of critical damping, ξ , and the natural circular frequency of the FSW, ω_n , as

$$\alpha \approx \xi \omega_n \quad (41)$$

with

$$\omega_n = \sqrt{\frac{E}{\rho R^2}} \quad (42)$$

Here E is the modulus of elasticity equal⁶⁷ to 160 GPa; ρ is the density of the FSW material equal to 7860 kg/m³; and R is the radius of the FSW equal to 5 m. For our case $\xi = 0.02$, $\omega_n = 900 \text{ s}^{-1}$, and the stress-amplitude ratio between two successive cycles is 0.88. The amplitude of the stress decays to 10% of its initial value in approximately 1/8 second—well before the next fusion reaction occurs.

The peak-radial-impact stress is less than 10% of the peak-hoop-impact stress because the FSW is a thin-walled vessel. There is also a steady-state stress in the axial direction from the static weight of the wall. This steady-state stress is approximately 800 kPa, which is less than 2% of the peak-hoop-impact stress and well below creep limitations. Because of the relatively large magnitude in the peak-hoop-impact stress, both radial and axial stresses are neglected in this analysis.

For our case, we have from Fig. 8-64 10⁹ cycles each of the stresses resulting from lithium impacts varying from plus to minus 60, 53, 47, 41, 36 . . . MPa. To each of these stresses, we should add the 2-MPa thermal stress caused by the 1°C temperature change in the FSW wall following each fusion reaction (see Fig. 8-63). We also have 100 cycles of startup where we must add the 86-MPa thermal stress to the lithium-impact stresses.

The sum of all FSW surface stresses included in the analysis is shown schematically for the surface of the FSW as a function of time in Fig. 8-65. The slowly varying nature of the curves is caused by the thermal stresses resulting from startup and shutdown. The rapidly varying stresses are caused by the lithium-impact loads. The 2-MPa thermal stress at 1 Hz is of such small magnitude that its variation is

Fig. 8-64. Variation of FSW lithium-impact hoop stress following a single-fusion reaction.

lost when compared with the lithium-impact stresses.

After the 1-Hz fusion reactions begin, the temperature difference between the center and the surface of the FSW increases (see Fig. 8-63). Correspondingly, the thermal stress at the surface increases in a positive direction (tension) and the thermal stress at the center (not shown in Fig. 8-65) increases in a negative direction (compression). However, creep occurs continuously and relaxes these thermal stresses so that they slowly reach a maximum and then decay. These thermal stresses continue to decay until shutdown occurs, at which time the temperature gradient in the FSW vanishes. Residual stresses appear as compression at the surface and tension at the center, and the absolute value of the temperature in the FSW decreases as the chamber cools. When the fusion reactions begin again, the thermal stresses reappear.

Summarizing, we must allow for the stresses shown in Fig. 8-65, which include the effect of creep relaxation. The impact stresses are independent of small plastic deformations in the FSW. For this reason, they are classified as primary stresses in the ASME code. In contrast, thermal stresses relax

themselves with minor plastic deformations and thus are classified as secondary stresses. For example, the plastic strain necessary for relief of the 86-MPa thermal stress that occurs during the startup and shutdown periods is only 0.05%. A tabular summary of these primary and secondary stresses is included in Table 8-4.

Method of Determining Limiting Stresses in HYLIFE Components. We have conservatively chosen to satisfy the intent and methodology of ASME Code Case N-47 (1592-10),⁵⁷ which is required for containment structures. In essence, this code case lists three criteria, which should be satisfied, and a fourth criterion, which does not apply to HYLIFE conditions. These criteria are applied to our most highly stressed component, the first structural wall, even though it is not itself a containment structure.

● The first criterion involves creep-fatigue evaluation where we determine the number of cycles necessary to cause a failure under fluctuating primary loading conditions. Using the nomenclature of Ref. 57, we must satisfy

$$\sum_{j=1}^P \left(\frac{n}{N_D} \right)_j + \sum_{k=1}^q \left(\frac{t}{T_D} \right)_k \leq D \quad (43)$$

where n is the number of applied cycles of loading conditions, j ;

N_D is the number of design allowable cycles of loading condition, j , obtained from design fatigue curves, described later;

t is the time duration of the steady-creep load condition, k ;

T_D is the allowable time at a given effective stress from the steady-creep load, k , determined for an inelastic analysis;

D is the total creep-fatigue allowable-damage limit (dimensionless).

Addressing the second term, we note that Code Case N-47 shows a minimum stress-to-rupture strength of 165 MPa for 100 h and 58 MPa for 3×10^5 h (34 yr) at 590°C. Our only steady stress is that caused by the weight of the FSW (~800 KPa), which is below the minimum stress-to-rupture strength by a factor of over 70. The 80- to 90-MPa secondary thermal stress occurring during startup and shutdown operation decays because of creep relaxation. Hence, the second term is negligible.

Fig. 8-65. Schematic of stress-time signature at the surface of the HYLIFE FSW.

Table 8-4. Summary of significant primary and secondary FSW stresses.

	Primary hoop impact stress (MPa)	Secondary thermal stress at FSW surfaces (MPa)	Total hoop stress at FSW surfaces (MPa)
Normal operation (10 ⁹ cycles)	-60 to +60	+2	-58 to +62
	-53 to +53	+2	-51 to +55
	-47 to +47	+2	-45 to +49
	-41 to +41	+2	-39 to +43
	-36 to +36	+2	-34 to +38
Peak conditions during startup-shutdown operation (100 cycles)	-60 to +60	-84 to +88	-144 to +148
	-53 to +53	-84 to +88	-137 to +141
	-47 to +47	-84 to +88	-131 to +135
	-41 to +41	-84 to +88	-125 to +129
	-36 to +36	-84 to +88	-120 to +124

When this term is negligible, the value of D is unity so that Eq. (43) becomes

$$\sum_{j=1}^p \left(\frac{n}{N_{Dj}} \right) \leq 1. \quad (44)$$

In determining N_{Dj} , the ASME code specifies that the equivalent strain range

$$\Delta \epsilon_{\text{equiv}} = \frac{\sqrt{2}}{2(1+\nu)} \left[\left(\Delta \epsilon_r \right)^2 + \left(\Delta \epsilon_\theta \right)^2 + \left(\Delta \epsilon_z \right)^2 + \frac{3}{2} \left(\Delta \gamma_{r\theta}^2 + \Delta \gamma_{\theta z}^2 + \Delta \gamma_{rz}^2 \right) \right]^{1/2} \quad (45)$$

be used with an approved fatigue curve. Here ν is Poisson's ratio and $\Delta \epsilon_r$, $\Delta \epsilon_\theta$, and $\Delta \epsilon_z$ are the dif-

ferences between the peak-positive strain and peak-negative strain in each of the three coordinates. $\Delta\gamma_{\theta\theta}$, $\Delta\gamma_{\theta z}$, and $\Delta\gamma_{zz}$ are the corresponding differences in shear stresses. The ASME code uses a multiplier of $\sqrt{2}/3$ rather than $2\sqrt{2}/2(1 + \nu)$. Booker⁵⁸ points out that the $\sqrt{2}/3$ multiplier applies only to plastic conditions and not in general. When a plastic material is present, $\nu = 0.5$ and the two multipliers are identical.

When we consider a thin-walled vessel, such as the FSW, subjected to uniform internal radial forces, a state of one-dimensional stress exists with

$$\begin{aligned}\Delta\epsilon_{\theta} &= \frac{\Delta\sigma_{\theta}}{1} \\ \Delta\epsilon_r &= \Delta\epsilon_z = -\frac{\nu\Delta\sigma_{\theta}}{1} \\ \Delta\gamma_{\theta\theta} = \Delta\gamma_{\theta z} = \Delta\gamma_{zz} &= 0\end{aligned}\quad (46)$$

so that Eq. (45) becomes

$$\Delta\epsilon_{\text{equiv}} = \frac{\Delta\sigma_{\theta}}{1} \quad (47)$$

Campbell,⁷⁰ who wrote the section on creep fatigue in Code Case N-47, suggests that the number of fusion pulses permitted is best determined by using the total number of load cycles during normal operation shown in Table 8-4 with Eqs. (44) and (47), and the fatigue property curves relating the number of allowed cycles, N_D , to the equivalent strain range, $\Delta\epsilon_{\text{equiv}}$.

Booker,⁵⁸ Brinkman,⁶⁴ and Jaske⁶⁵ are involved in a continuing program that includes specifying fatigue properties of 2-1/4 Cr-1 Mo ferritic steel at high temperatures and large numbers of cycles. Their results agree with each other, and Booker's work is being approved for inclusion in the ASME code. Booker's relationships for continuously cycled plastic and elastic strain range ($\Delta\epsilon_p$ and $\Delta\epsilon_e$) vs cycles to failure (N_D) at 860 K are

$$\begin{aligned}\Delta\epsilon_p &= \begin{cases} 9.71 N_D^{0.370} & N_D > 10^4 \\ 176.5 N_D^{0.720} & N_D < 10^4 \end{cases} \\ \Delta\epsilon_e &= \begin{cases} 0.298 N_D^{0.0287} & N_D > 10^4 \\ 0.606 N_D^{0.0959} & N_D < 10^4 \end{cases}\end{aligned}\quad (48)$$

Fig. 8-66. Variation of allowed $\Delta\epsilon_{\text{equiv}}$ with low values of N_D using Booker's correlations of $\Delta\epsilon_p$ and $\Delta\epsilon_e$.

with the total-strain range ($\Delta\epsilon_t$) equal to $\Delta\epsilon_p$

$$\Delta\epsilon_t = \Delta\epsilon_p = \Delta\epsilon \quad (49)$$

Booker also documents $\Delta\epsilon_p$ and $\Delta\epsilon_e$ values at 800 K that are between 17 and 43% higher than those of Eq. (48), but we conservatively use the 860 K values.

Following normal ASME Code procedures, the allowed equivalent-strain range, $\Delta\epsilon_{\text{equiv}}$, is equal to $\Delta\epsilon_t$ lowered by the most conservative of the safety factors, 20 on N_D or 2 on $\Delta\epsilon_p$. Below approximately 10^3 cycles the former criterion is most restrictive and above 10^3 cycles the latter criterion is most restrictive. Figures 8-66 and 8-67 show the variation of allowed $\Delta\epsilon_{\text{equiv}}$ with the number of design allowable cycles, N_D , for a temperature of 860 K. The same information is plotted on these two curves to improve readability over the range of 10 decades. The curves are smoothed in the transition region between the two regimes where the most restrictive criterion for calculating $\Delta\epsilon_{\text{equiv}}$ changes. Inclusion of the curves beyond $N_D = 10^6$ cycles in the ASME Code is not expected in the near future. However, Booker has documented data at cycles up to 10^6 and extrapolates his curves to 10^9 . An extrapolation slightly beyond 10^9 cycles in Fig. 8-67 is needed for

Fig. 8-67. Variation of allowed $\Delta\epsilon_{\text{equiv}}$ with high values of N_D using Booker's correlations of $\Delta\epsilon_p$ and $\Delta\epsilon_c$.

correct analysis using Eqs. (44) and (47) under HYIHF design conditions.

Application of the creep-fatigue criterion to the FSW starts with the total-loading conditions for normal operations in Table 8-4. We calculate the equivalent strain range, $\Delta\epsilon_{\text{equiv}}$, using Eq. (47). We then use Figs. 8-66 and 8-67 to determine the number of design allowable cycles, N_D , for each loading cycle. These values are tabulated in Table 8-5. The last column on the right tabulates values of n/N_D . According to Eq. (44), the value of $\sum n/N_D$ must be less than unity, which it is. Note that because of the logarithmic nature of the data a small change in the magnitude of the primary loads makes a substantial change in $\sum n/N_D$.

The inner radius of the liquid-metal-jet array was chosen to be 550 mm for our design because a significantly smaller radius would not permit Eq. (44) to be satisfied. A larger inner radius could be used; but this increases the flow rate of lithium and the pumping power.

The impact stress could also have been reduced by increasing the FSW wall thickness above the selected value of 50 mm. However, then the second criterion could not be satisfied without internal cooling because the secondary-thermal stress in-

Table 8-5. Calculations of creep-fatigue and operating conditions for the FSW.

Creep-fatigue conditions			
Total stress cycle (MPa)	$\Delta\epsilon_{\text{equiv}}$ (%)	N_D	n/N_D
-58 to +62	0.075	4×10^{10}	0.025
-51 to +55	0.066	$\sim 10^{12}$	0.001
-45 to +49	0.059	—	—
-39 to +43	0.051	—	—
-34 to +38	0.045	—	—
$\sum n/N_D \sim 0.030$			
Operating conditions			
Primary impact stress (MPa)	Actual time (h) ^a	Allowable time (h) ^b	Time ratio ^c
60	430	10 000	0.043
53	760	25 000	0.031
47	970	58 000	0.017
41	1100	140 000	0.008
36	1200	300 000	0.004
0.100			

^aBetween this stress level and next lower stress level.

^bAt this stress value.

^cOf actual to allowable.

creases more rapidly than the primary-impact stress decreases.

• The second criterion that needs to be satisfied in Code Case N-47 is associated with maximum strain limits. The code permits satisfaction of this requirement using either an elastic or an inelastic analysis. If the elastic analysis is used, the sum of the primary and secondary stresses must be less than the yield strength (Fig. 8-68 shows an ASME-allowed yield strength, S_y , of 160 MPa at our operating temperature of 810 K). The inner radius of the liquid-metal jet array in conjunction with the thickness of the FSW were selected so that this criterion is satisfied (148-MPa total primary and secondary stresses). Future calculations using a more complex inelastic analysis where 1% total strain is permitted may result in a less stringent strain limit criterion.

• The third criterion applies to what the ASME calls "operating conditions." This criterion specifies that the cumulative time effect of the primary membrane stress intensity, P_m , be less than

Fig. 8-68. Strength properties of 2-1/4 Cr-1 Mo ferritic steel from ASME Code Case N-47.

an allowable limit of general primary-membrane stress intensity, S_{mt} . The values of S_{mt} and other strengths are shown in Fig. 8-68. We combined the primary-impact-stress levels of Table 8-4 with the values of S_{mt} at our design temperature of 810 K to form a ratio of actual time to allowable time for each loading condition as shown in Table 8-5. The total sum of these ratios must be less than unity. In our case, this sum is a very acceptable 0.10, indicating that this criterion offers no restriction. Other operating condition restraints include local and bending stresses; but these restraints are less stringent for the HYLIFE FSW.

● A fourth criterion applies to "design conditions" in which the steady-primary load is limited to a maximum allowable stress intensity, S_0 . Essentially all of the primary load on the HYLIFE inner liner is cyclic with a period of approximately 7 ms so that although not explicitly stated, this criterion is not applicable. Rather the cyclic nature of the load is considered in the "operating conditions" criterion. The steady-primary load caused by the weight of the FSW is only 800 KPa, which is negligible in respect to the allowed 53 MPa for S_0 at 810 K.

Summary. The intent and methodology included in ASME Code Case N-47 are followed in designing HYLIFE components and, in particular, the first structural wall. Two criteria are limiting: a

creep-fatigue criterion, which is based on the cumulative damage method and Booker's fatigue data for 2-1/4 Cr-1 Mo ferritic steel, complies with anticipated revisions to the ASME code except that the number of cycles for HYLIFE conditions exceeds 10^6 . Fatigue data at over 10^8 cycles exists so that extrapolation to the HYLIFE design point of 10^9 cycles appears reasonable.

A second criterion associated with strain limits restricts the total peak primary plus secondary stress to the yield strength of the material. A further revision using an inelastic analysis limit of 1% strain may result in a relaxation of this criterion.

Application of the method to the HYLIFE FSW shows an acceptable life of over 30 yr for a 50-mm-thick FSW and an inner radius of the liquid-metal-jet array of 550 mm.

The HYLIFE LMW design offers flexibility in case impact stresses should be revised after further analysis. Any reasonable revision can be accommodated by changing the FSW thickness and the inner radius of the liquid-metal-jet array.

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First-Wall Design Using Several Procedures and LMW Geometries

Introduction. Preceding articles in this section describe in detail the cyclic loading of the HYLIFE first structural wall (FSW) for a single-point design (liquid-metal-jet-array inner radius of 0.5 m, effective array thickness of 0.8 m, and a packing fraction of ~50%). In the preceding article, we outlined a design procedure that would be ASME approved if the 2-1/4 Cr-1 Mo fatigue properties were in the code. That method did not predict a 30-yr life from mechanical strength considerations for the geometry used in the detailed hydrodynamic calculations. Yet we can show from simple hydrodynamic modeling that increasing the array inner radius increases the wall life when all other factors are held constant.

The radiation damage life is approximately 68 yr at 70%-capacity factor based on the methods described in Ref. 71, and a 1-m effective liquid-metal-jet-array thickness. For a 0.8-m LMW, the radiation damage lifetime of the FSW is 37 yr. Thus mechanical design is more restrictive than radiation damage for these thick LMWs.

We describe here a simple model that can determine a more optimum design point for future sophisticated calculations. The simple model is normalized to the single-point calculation using more sophisticated analyses. We also show a range of mechanical design models including the ASME method we chose in the previous article.

Wall Stresses. For energy deposited in a material during a period of time so short that the pressure cannot relieve itself during the deposition time, the pressure rise at any position x is

$$\Delta p(x) \approx \rho \Gamma \left[\frac{q'''(x)}{\rho} \right], \quad (50)$$

where ρ and Γ are the density and the Grüneisen constant of the material, respectively; and $q'''(x)/\rho$

is the energy deposition per unit mass at position x . A relief wave moves into the material from the free surface and spalls the surface if the tensile strength is exceeded. The velocity of the spalled material at any position x is given by

$$v(x) = \frac{1}{2\rho c\delta} \int_{2x}^{2(x+\delta)} \Delta p(x') dx', \quad (51)$$

where c is the acoustic velocity of material, δ is the thickness of the spalled layer, and $p(x')$ is given by Eq. (50).

If the pressure distribution is smeared to a uniform level through the liquid, a simple equation for the slug velocity results:

$$v = \frac{\Gamma}{c} \left(\frac{E}{m} \right). \quad (52)$$

Here, E/m is the fusion energy deposited in the liquid per unit mass. This equation assumes that all the LMW momentum is consolidated into one outward moving slug (as shown in the article "Dissipation of Kinetic Energy by the HYLIFE Jet Array").

The fusion energy absorbed in the liquid blanket is

$$E = E_n [1 - \exp(-\mu t)], \quad (53)$$

where μ is the neutron-energy-attenuation coefficient, t is the effective blanket thickness to neutrons, and E_n is the fusion-neutron energy including exothermic energy from neutron interactions in the liquid blanket. We do not include the short-range, fusion-product-produced energy deposited in the liquid since that energy primarily accumulates in the core gas that blows through the array. (In "Outward Drag on the Jet Array by Expanding Lithium Plasma," we showed that this blow-through accounts for approximately 20% of the wall loading. We will account for that liquid momentum in the model's normalization factor.)

For a cylindrical liquid-metal jet array the energy per unit mass at the horizontal plane of the fusion event is

$$\frac{E}{m} = \frac{F}{\rho \pi h (R_1^2 - R_0^2) f} \left(\frac{2\pi R_0 h}{4\pi R_0^2} \right) = \frac{1}{2\pi R_0 (R_1^2 - R_0^2) f \rho l}. \quad (54)$$

Fig. 8-69. Slug velocity and density at impact with a 5-m-radius first structural wall. The curves are for a 1.0-m effective-lithium-array radius (packing fraction times actual thickness). The single data point is for an effective-array thickness of 78 cm at a packing fraction of 50%. The considerable reduction in slug velocity for the increase in array thickness is a result of the increased array mass (or decreased energy density). The slug density was calculated assuming the slug formed with initial outer radius of 15 cm greater than the initial array outer radius for the 50%-packing-fraction array.

where R_0 and R_1 are the inner and outer radii of the liquid blanket; and f is the packing fraction of the liquid between R_0 and R_1 . For a continuous blanket of liquid, $f = 1$.

The velocity of the slug midplane is given by combining Eqs. (52) to (54) as

$$v = \frac{\alpha \Gamma E_n}{2\pi \rho c f R_0 \left(R_1^2 - R_0^2 \right)} [1 - \exp(-\mu c)] \quad (55)$$

where α is an empirical factor that normalizes this model to the more sophisticated models. We use a value of $\alpha = 0.28$ based on the minimum-gap calculation given in "Dissipation of Kinetic Energy by the HYLIFE Jet Array" earlier in this section ($R_0 = 0.5$ m, $R_1 = 1.626$ m, $f = 0.835$, $E_n = 2277$ MJ, $v = 41.5$ m/s, $c = 4.5$ km/s, $\rho = 485$ kg/m³, and $\Gamma = 0.9$).

The jet-array geometry vents the short-range-fusion-product-produced core gas before slug formation, and the slug will cavitate as it coasts at a constant velocity to the FSW because of the diverging geometry (i.e., slug thickness will be un-

Fig. 8-70. The peak FSW hoop stress is a function of both array location and wall thickness. The effective-array thickness for this plot is 1.0 m; the FSW radius is 5.0 m, and the fusion energy is 2700 MJ.

changed). The impact pressure will persist longer than a first-structural-wall period because of the cavitating slug's recompaction time, and the peak stress will be $2PR/\delta$, where R and δ are the FSW radius and thickness, and P is the impact pressure. From the article "Dissipation of Kinetic Energy by the HYLIFE Jet Array,"

$$P = \rho v^2 [\eta/(1 - \eta)] \quad (56)$$

where ρ is the normal-liquid density, and η is the ratio of the cavitating-liquid density to the normal-liquid density.

If the slug forms with outer radius approximately 15 cm greater than the original-array outer radius (as it did in the "actual" calculation), the density ratio, η , at impact may be found from simple geometry.

Figure 8-69 shows the velocity and cavitating to normal density ratio as a function of array inner radius for the 2-m thick, 50%-packing-fraction array, and 5-m FSW radius. The geometry used in the hydrodynamic calculations (0.8-m effective LMW thickness) is also shown.

The impact stress, $2PR/\delta$, is shown in Fig. 8-70 as a function of array inner radius and FSW

Fig. 8-71. The peak thermal stress and peak temperature vs. wall thickness for a 5.0-m-radius wall, 1.0-m effective-array thickness, and 2700-MW fusion power. The deviation in thermal stress from parabolic at larger thicknesses is due to the reduction of the modulus of elasticity with increasing temperature.

Fig. 8-72. Properties of 2-1/4 Cr-1 Mo steel vs temperature.

thickness. We calculated the thermal stress using the methods of Ref. 72 with internal-FSW heat source (4 MW m^3), coefficient of expansion ($1.19 \times 10^{-5} \text{ K}^{-1}$), Poisson's ratio (0.318), thermal conductivity (32.5 W/m-K), and convection coefficient ($14,700 \text{ W/m}^2\text{-K}$). The elastic modulus was allowed to vary with temperature. We plot the peak-FSW temperature and thermal stress as a function of FSW thickness in Fig. 8-71.

Design Limits—ASME Method. The ASME-design procedure described in the article "Design Method for the HYLIFE-Reaction Chamber" can be simplified to two criteria for this scaling model.

Fig. 8-73. Design space for a 1-m effective array thickness, 5.0-m-FSW radius; 1-Hz, 2700-MJ fusion pulses; and 30-year life. In the yellow region, the sum of alternating and thermal stresses is less than yield strength. In the blue region, the alternating stress is less than one-half the fatigue strength. The allowable design point must lie in the green region which satisfies both criteria. The point selected is the point meeting both criteria with the least inner radius.

First, the impact stress must be less than one-half the fatigue strength. Second, the sum of the impact and thermal stresses must be less than the yield strength.

Both the yield strength and the fatigue strength are functions of temperature. In Fig. 8-72 we show the temperature dependence of these strengths and also of the elastic modulus used to calculate the thermal stress. The fatigue strength was calculated for 10^{10} cycles (10^9 fusion pulses, 10 rings per pulse) using Booker's data (see the article "High-Cycle Fatigue Strength of 2-1/4 Cr-1 Mo Ferritic Steel at Elevated Temperatures").

We have combined the FSW stresses and design limits in a single plot (Fig. 8-73). This graph shows the allowable region for FSW thickness and jet-array inner radius for a 2-m thick, 50%-packing-fraction array absorbing the 2700 MJ, 1-Hz-fusion pulse and a 5-m-wall radius. To minimize pumping power and lithium/tritium inventories, we choose the allowable point with the smallest array inner radius, 0.56 m. The corresponding FSW thickness is 48 mm. We must also consider the effect of effective

Fig. 8-74. Four design procedures for the first structural wall. All but the ASME method collapse to the fatigue strength when the thermal stress is relieved by creep. (The ASME method collapses to one-half the fatigue strength.)

array thickness (packing fraction times actual thickness) and the FSW radius before selecting an optimum design point. We will select the point that results in minimum lithium flow area (resulting in minimum pumping power and lithium/tritium inventories) and in minimum building size.

Other Design Methods. The first structural wall is not a containment vessel and thus need not meet the requirements of the ASME code. Failure of the FSW would produce an economic penalty, but it would not endanger either plant integrity or the environment. Therefore, other methods of establishing design limits can be considered. We have examined several alternatives:

- The Goodman Law.
- The Modified Goodman Law.
- The Soderberg Law.

These alternatives are discussed in Ref. 73. The allowable alternating stress is plotted in Fig. 8-74 for each method.

We have conservatively assumed that creep will not relieve the thermal stress in all cases. If creep-relief is instantaneous, the ASME method would produce an allowable alternating stress curve equal to one-half the fatigue strength; and the other methods would produce an allowable curve equal to

the fatigue strength. Recent creep calculations indicate the thermal stress will be largely relieved within a few days after each system start-up (see preceding article).

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Removal of Noncondensable Gases from the HYLIFE Chamber—Initial Conditions

A laser-driven fusion reactor must be restored to an equilibrium pressure of less than $\sim 10^{-1}$ Torr between fusion pulses. Similarly, a ballistic-propagation, heavy-ion, beam-driven fusion reactor must be restored to a pressure of less than $\sim 10^{-4}$ Torr between pulses. The vapor in the chamber just before the fusion pulse has the following sources:

- Helium from fusion reactions in the target.
- Unburned deuterium and tritium from the target.
- Other gases from the target.
- Helium and tritium from neutron-lithium reactions in the LMW.
- Hydrogen that diffuses from the steam loop through the sodium loop to the lithium-flow loop.
- Lithium vapor at the vapor pressure corresponding to the liquid temperature.

Helium from Fusion Reactions. The target helium production rate is

$$R_1 = \left(\frac{P_{1N}}{1.6 \times 10^{19} \text{ MJ/MeV}} \right) \left(\frac{1 \text{ reaction}}{17.6 \text{ MeV}} \right) \times \left(\frac{4 \text{ g He}}{6.02 \times 10^{23} \text{ reactions}} \right) \quad (57)$$

For a 2700-MW_{TN} chamber, the target helium production rate is 6.4 mg/s. This helium will

probably be absorbed in lithium vaporized by the x-ray pulse; hence, it will be in the vapor to be pumped rather than being entrained in the liquid wall.

Unburned Deuterium and Tritium in the Target.

For a 30% burn fraction, unburned fuel enters the chamber at a rate of 7.4 mg/s of deuterium and 11.1 mg/s of tritium. This gas will probably not be entrained in the LMW.

Other Gases from the Target. Our Monte Carlo neutronics calculations show approximately 0.08 mg/s of hydrogen is produced from nuclear reactions in the target, such as $d(n,2n)p$ reactions. In addition, compounds used in the pusher and ablator layers of the target will be vaporized during the fusion pulse with some products remaining as vapor after cooling. Since no specific target designs are being considered in this preliminary estimate, we simply double the mass and partial pressure of the other target gases (helium and unburned fuel).

Helium and Tritium Produced in the LMW. For each D-T reaction in the target, between 1.0 and 1.75 tritium atoms are produced in the LMW (depending on the tritium breeding ratio—TBR). Since each tritium production reaction is accompanied by the production of one atom of helium, the LMW-helium production rate equals the product of the target helium (or neutron) production rate and the TBR. For the maximum TBR of 1.75, the LMW-production rates are 11.1 mg/s of helium and 8.3 mg/s of tritium. These gases are entrained in the LMW and must bubble out. In time, however, the amount of He bubbling out of the LMW will equal the production rate in the LMW. The tritium will be removed from the liquid lithium outside the chamber by other means. Because of its low remaining concentration in the liquid, the tritium will have a very low partial pressure above the liquid; we assume it to be zero for this preliminary analysis.

Hydrogen Diffuses from the Steam Loop through the Sodium Loop to the Lithium Flow Loop. Design studies of the Clinch River Breeder Reactor Project indicate that hydrogen can be cold trapped in the sodium loop to a steady-state concentration of 0.126 weight part per million that corresponds to a partial pressure of hydrogen of approximately 3×10^{-5} Torr. At this pressure above the sodium, the hydrogen diffusion rate into the lithium loop will be (see Ref. 74)

Fig. 8-75. Partial pressure of hydrogen over 775-K liquid lithium as a function of hydrogen concentration in the liquid.

$$\phi \left(\frac{\text{cm}^3 (\text{STP})}{\text{sec}} \right) = \frac{CA}{X} \left(p_1^{1/2} p_2^{1/2} \right) \quad (58)$$

We use $A = 24,500 \text{ m}^2$, $X = 3 \text{ mm}$, and $C = 3 \times 10^{-2} [\text{cm}^3 (\text{STP}) \text{ mm}] (\text{sec m}^3 \text{ Li} \cdot \text{T}^{-1/2})$. p_1 is the hydrogen pressure above the sodium (3×10^{-5} Torr), and p_2 is the hydrogen pressure above the lithium.⁷⁵

$$p_2 (\text{Torr}) = 1.33 \times 10^{-9} [C(\text{ppm})]^{4/3} \quad (59)$$

This equation is plotted in Fig. 8-75. The hydrogen diffusion rate is 0.12-mg/s when the hydrogen concentration in the 775 K lithium is maintained at 1 ppm by extraction with the bred tritium. The partial pressure of hydrogen associated with this concentration is 1.3×10^{-9} Torr.

Lithium Vapor. For a portion of the interpulse time, high lithium pressure will exist in the chamber. This pressure must be accommodated by the pump with subsequent condensation of the pumped hot lithium vapor beyond or within the pump, or the pump must be blocked during the high-pressure periods. This problem has not yet been addressed in our study.

The vapor pressure above lithium at 775 K is $\sim 3.3 \times 10^{-3}$ Torr. The vapor pressure above 83 atomic percent Pb-17 atomic percent Li is $\sim 1.3 \times$

10^{-5} Torr (see "Properties of Lead-Lithium Solutions").

Total Noncondensable Gas Load. Neglecting hot-vaporized lithium, Table 8-6 lists partial pressures and vapor masses that will exist in the ~ 1000 m³ chamber assuming the vapor temperature is 775 K.

We cannot pump the vapors associated with the liquid-metal-wall vapor pressures below the pressures listed since new vapor would simply form from the liquid. Note that for the pure lithium reactor (i.e., laser-driven reactor), the species that we want to pump are only $\sim 8\%$ of the vapor mass in

the chamber after one fusion pulse, and as we pump, that fraction decreases. Thus we should allow some pressure buildup before pumping. If laser propagation limits the pressure to 10^{-4} Torr, over 200 s of steady operation are possible without hindering the beam.

Details of the pump design are needed to determine mass flow rate and the initial and final pressure after pumping cycle for the laser-driven chamber. However, it is clear that if the pressure is allowed to build for ~ 10 s, the fusion-pulse-source vapor will be over 50% by mass of the vapor pumped. The mass flow rate will be of order 100 mg/s, or the new vapor added each second. (We neglect any hot vaporized lithium above the pressure.) The initial and final pump cycle pressures will thus be of order 10^{-2} .

If ion-beam propagation limits the pressure to 10^{-4} Torr, either the temperature must be lowered or a liquid metal with lower vapor pressure must be chosen (such as 83% lead-17% lithium; see article "Properties of Lead-Lithium Solutions"). For the lead-lithium liquid, one pulse raises the chamber pressure to approximately five times the drive propagation limit. Therefore, after each pulse, ~ 100 mg must be pumped at a pressure near 10^{-4} Torr.

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Evolution of the HYLIFE Fusion-to-Electricity Converter

Overview

Generation of economical electric power from inertial-confinement fusion shows increasing promise because of potential modifications incorporated into the HYLIFE reaction chamber.⁷⁶ LLNL and the Energy Systems Group of the Rockwell International Corporation studied modifications in which flexibility of the design and simplification in

Table 8-6. Gas loads in the HYLIFE chamber (vapor temperature = 775K). Liquid metal vaporized by the fusion pulse is not included as it is assumed to be condensed by the LMW itself. In practice, hydrogen isotopes may also condense to equilibrium values in the LMW; therefore, this table represents upper bound loads. The nonfusion sources are based on the partial pressure associated with concentrations in the liquid. In the case of hydrogen entering from the H₂O and Na loops, the diffusion rate is too low to sustain the pressure shown if the chamber is vacuum pumped.

	Vapor mass (mg)	Partial pressure (Torr)
From fusion pulses (per pulse)		
Target He	6.4	5.4×10^{-5}
LMW He	11.1 (max)	9.3×10^{-5} (after bubbling out)
Target D ^a	7.4	6.2×10^{-5}
Target T ^a	11.1	6.2×10^{-5}
Other target gases (including protium)	25 (?)	1.8×10^{-4} (?)
Total pulsed	60	4.5×10^{-4}
Nonfusion sources: pure Li HYLIFE		
Equilibrium Lithium vapor	690	3.3×10^{-3}
Hydrogen from H ₂ O and Na loops ^a	0.00008 (max)	1.3×10^{-9} (max)
Nonfusion sources: 83% Pb - 17% Li HYLIFE		
Equilibrium Lead-lithium vapor	80	1.3×10^{-5}
Hydrogen from H ₂ O and Na loops ^{a,b}	0.00003 (max)	1.3×10^{-9} (max)

^a Hydrogen isotopes are considered diatomic. Assuming all hydrogen (protium) in the Pb-Li loop originates in the Na/H₂O loops; none is entrained from the fusion pulse or produced by nuclear reactions. Also assumes hydrogen isotopes removed from LMW down to 1 ppm.

Fig. 8-76. Comparison of the (a) original and (b) modified HYLIFE reaction chamber.

construction were emphasized. The modified chamber includes a liquid-metal (lithium) jet array as shown in Fig. 8-1. This jet array acts as: (1) a renewable first wall and blanket to shield metal components from x-ray and neutron exposure, (2) a tritium breeder to replace tritium burned during the fusion process, and (3) an absorber and transfer medium for fusion energy. Over 90% of the energy produced in the reaction chamber is absorbed in the liquid-metal-jet array.

The effectiveness of the jet array for shielding allows the use of structural components without replacement over the entire 30-yr expected lifetime of the power plant. These structural components are made from low-alloy, 2-1/4 Cr-1 Mo ferritic steel that is compatible with flowing lithium, is resistant to hydrogen (tritium) embrittlement, and produces few radioisotopes. The liquid lithium is circulated through 16 external loops, where both the tritium

and the energy produced in the reaction chamber can be removed.

As originally conceived,^{77,78} the HYLIFE chamber is pulsed every second by a 2700-MJ fusion reaction. A deuterium-tritium pellet is injected into the chamber and laser or heavy-ion beams initiate the fusion reactions. The fusion energy from the reaction is absorbed in the circulating liquid lithium. A comparison of the original and modified designs is shown in Fig. 8-76.

The liquid lithium enters the modified chamber radially through equally spaced 1.2-m-inside-diam pipes near the top. The lithium free falls into a pool, flows vertically downward through a nozzle plate that forms the jet array, and falls through the main body of the chamber. It then collects at the bottom where it flows into pipes and is pumped back into

Table 8-7. Comparison of parameters—original and modified HYLIFE chambers.

Parameter	Original design	Modified design
Fusion energy yield per shot, MJ	2700	2700
Repetition rate, Hz	1	1
Lithium flow rate in jet array, m ³ /s	93	142
Lithium flow rate for first structural wall, reflector, and vacuum vessel cooling, m ³ /s	36	18
Total lithium flow rate, m ³ /s	129	160
Lithium temperature (average), K	770	770
Temperature rise in lithium per shot, K	12	10
Total lithium inventory, m ³	2700	2500
Pump efficiency	0.72 (mechanical)	0.40 (EM)
Pumping power, MW _e	20	40
Shielding at midplane, m of lithium	1.06(+0.54, -0.20)	1.00(+0.40, -0.02)
Packing fraction at midplane ^a	0.50	0.48
Jet cross section	Circular	Rectangular
Jet velocity at nozzle plate, m/s	4.4 (curved)	5.5 (vertical)
Annular weir velocity at nozzle plate, m/s	—	7.1
Inner jet array radius at midplane, m	0.439	0.55
Vacuum vessel:		
Inner radius, m	5.435	5.435
Wall thickness, mm	80	70
Nozzle plate thickness, m	0.4	.8
First structural wall:		
Inner thickness, mm	5.0	5.0
Wall thickness, mm	35	50
Graphite reflector:		
Inner radius, m	5.085	5.085
Wall thickness, mm	300	300

^aPacking fraction is defined as the ratio of liquid to total cross-sectional area in the region of the liquid metal jet array.

the chamber at the top. At the midplane, where the fusion reactions occur, the jet array has an effective mean thickness of 1 m with a packing fraction of one-half. The orifice plate and splash suppressor included in our original design are not required in the modified design because vertical jets can tolerate liquid-lithium head fluctuations without coalescing, and because of the use of electromagnetic pumps which are resistant to pressure pulses from the fusion reactions.

The advantage of the jet array over a solid annulus or several separate concentric annular rings is that high-pressure gases, generated by short-range energy deposition from the fusion reactions, can vent through the interstices between the jets. Even though the energy deposition in the lithium closest to the fusion reaction is so large that these jets partially disassemble before the high-pressure gases can vent through the interstices, a circumferential density variation exists with the lowest value occurring in the regions of the interstices. These regions have lower resistance to gas flow so that the venting process is still enhanced. Less outward-directed liq-

uid momentum is produced, reducing the momentum transferred to the first structural wall (FSW). The impact affects only the FSW, so that essentially a steady loading exists on the vacuum vessel.

HYLIFE Component Design

The simplicity and flexibility of the HYLIFE-chamber design are key advantages over other proposed chambers. In the modified-HYLIFE design, the vacuum vessel contains only a nozzle plate, FSW, and graphite reflectors. Either laser or heavy-ion fusion drivers may be used. The driver beams enter horizontally into the chamber through openings in the vacuum vessel, FSW, and graphite reflectors. Fuel pellets are injected horizontally next to one of the beams. (Vertical injection through the top access port is also possible.)

In the following paragraphs, we describe major components as well as the characteristics of the modified HYLIFE chamber. Design parameters for the original and modified chambers are compared in Table 8-7.

Nozzle Plate. The flow of liquid lithium is vertically downward with the jet cross section being

Fig. 8-77. Cross-sectional and elevation views of the modified nozzle plate.

generally rectangular as shown in Fig. 8-77. This geometry offers the advantages of simplicity, a nearly uniform shielding of the FSW and other components, and an increased tolerance to the variation of liquid-lithium head above the nozzle plate. Except for the driver-beam injection areas, the FSW is protected by a minimum of one meter of lithium at midplane.

In the original design, the jets had a radial velocity component, so that a variation in liquid-lithium head could have caused the jets to coalesce. An additional orifice plate above and separate from the nozzle plate was required to assure a uniform lithium head. With the vertical jets of the modified design, the only effects of a change in liquid-lithium head are variations of jet velocity and jet cross-sectional area below the nozzle plate. The centerline location of each jet is independent of spatial or temporal changes in the liquid lithium head. Hence, the jets remain separate.

The innermost zone of the modified nozzle plate is protected from neutrons, debris, and x rays by an annular weir flow over the inner radius. Thus this portion of the nozzle plate is also shielded by one meter of lithium or more. The weir flow is designed to break up in the first 2 m of flow to allow horizontal injection of the driver beams and venting of high-pressure gas.

Changes in the shape, location, and initial velocity of the jets and weir can be made simply by minor redesign of the jet-flow channels and weir deflector, and by variations of the lithium head above the nozzle plate. Construction as well as strength is considered in the design. The modified-nozzle plate consists of 33-mm-thick extruded-angle plates welded together. The part of the nozzle plate through which the jet array passes contains essentially straight-through penetrations. The outer portion is of similar structure, except the bottom and top are closed by flat plates to prevent the through-flow of lithium and to give additional strength. Some circulation occurs in the outer region to keep the maximum metal temperatures below 800 K. This circulation is directed to the FSW, reflector, and vacuum vessel to cool them as well.

The nozzle plate is simply supported from the FSW by a series of gussets. Circulative cooling around and through the gussets keeps peak temperatures below 800 K. The nozzle plate central hole equalizes pressure from the fusion reaction on

the top and bottom. Also, it makes the center of the chamber accessible. Graphite plug shielding problems are reduced because of the increased distance from the fusion reactions, and the nozzle-plate inner-region shielding problem is mitigated by the addition of the weir flow.

A more detailed stress analysis is in progress to determine the nozzle-plate thickness. This analysis considers the weight of lithium and the nozzle plate local-pressure fluctuations, liquid-lithium expansion and absorption of neutrons in the nozzle plate and the surrounding lithium. A conservative preliminary design set the thickness at approximately 1 m.

Alternative designs for the nozzle plate were developed by the Energy Systems Group of the Rockwell International Corporation⁷⁹ in which a grouping of spray nozzles (instead of an annular flow) is used to protect the inner portion from damage by x rays (see Fig. 8-78). The spray is directed down and outward but the quantity is not sufficient to prevent neutron damage. Hence, the inner portion of the plate is designed for periodic replacement.

Two concepts were considered in the outer portion of the alternate-nozzle-plate designs—a tubular structure shown in Fig. 8-78 or a box structure (not shown). The box structure is made rigid to withstand the cyclic loads without failure. The lower strength tubular structure is designed to be resilient and, thus, tends to reduce cyclic loads following each fusion reaction.

There are other nozzle plate designs that may be used in the HYLIFE converter. Because several nozzle plate designs could perform their functions for the lifetime of the energy converter, the final selection will be based, at least in part, on the manufactured cost.

Vacuum Vessel, First Structural Wall, and Reflector. Essentially all cyclic impulse loads from the fusion reactions are absorbed by the liquid-metal-jet array, FSW, and nozzle plate. Any cyclic pressure load imposed on the upper head is small (less than 0.1 MPa), because the gas flow through the central hole in the nozzle plate dissipates throughout the entire upper cavity inside the vacuum vessel.

The main body contains tubes for injection of the driver beams, and a full-diameter bolted flange with gaskets, to permit remote disassembly and reassembly of components, should that be necessary. The nozzle plate and FSW may be assem-

Fig. 8-78. Alternate tubular-nozzle-plate concept.

bled as a single unit before installation in the vacuum vessel. The beam and target injection tubes are attached to the vacuum vessel with seals and bolted flanges to ease remote assembly and disassembly. All seals are located above the vessel's lithium-liquid level, exposing them to lithium vapor only and, thus, maximizing their lifetime. The seals are maintained at a temperature greater than the saturation temperature to prevent condensation. No seal is necessary in the flange connecting the upper and lower parts of the FSW (see Figs. 8-76 and 8-77). The top access port may be used for inspection and minor repairs.

A space separates the lithium liquid from the vacuum vessel below the bottom of the FSW to minimize neutron-induced-lithium-pressure oscillations on the pumps. Similarly, the free-fall of lithium from the inlets near the top of the vacuum vessel to the pool above the nozzle plate also helps to reduce neutron-induced-lithium-pressure oscillations on the pumps.

The lower part of the vacuum vessel is now shaped so that the lithium hits at an oblique angle to the vertical axis of the vessel. The liquid surface forms a cusp-like shape at the outlet thus minimizing splash back into the region of the fusion reactions. Previously, a splash baffle was included in the design.

A 300-mm-thick graphite reflector (Figs. 8-76 and 8-77) is attached to the inside surface of the vacuum-vessel wall. It is encased in a thin-metal container and is cooled internally with lithium. The reflector is mounted to the vacuum vessel with brackets at the top. Supports at the bottom are slotted to allow for expansion.

A 50-mm-thick FSW is positioned inside the reflector and may be attached to the top head for easy removal. This wall is the most highly stressed component and is designed to last the entire 30-yr life of the reaction chamber. However, it can be replaced if necessary, with little expense and

downtime, as compared with replacing the vacuum vessel.

The thickness of the FSW is dictated primarily by temperature and stress considerations. As the thickness increases, the temperature in the wall increases and therefore the allowable design stress decreases. However, the cyclic stress caused by lithium impact also decreases. Thermal stresses, which increase as the peak temperature difference through the liner increases, were found to be of secondary importance for our design conditions because they are largely self-relieved after startup.

The FSW design procedure, documented in "Design Method for the HYLIFE-Reaction Chamber" is based on the creep-fatigue analysis⁸⁰ of the ASME Code Case N-47. Determination of the lithium-impact stress, which occurs about 10^9 times over the 30-yr lifetime of the chamber, is shown in the article "Dissipation of Kinetic Energy by the HYLIFE Jet Array." Wall temperature and thermal stress are determined from Ref. 78. Alternate design procedures may be utilized because the FSW is not a containment structure. However, we chose the ASME method because of its conservatism and its acceptance in industry. Several alternate design procedures are outlined in "First-Wall Design Using Several Procedures and LMW Geometries" earlier in this section. Other, more complex, types of FSW designs, which may permit a smaller inner radius or an increased safety factor, are being considered.

Lithium Circulation. The circulation system consists of 16 separate loops, each containing an electromagnetic (EM) pump. Inlets and outlets are 1.2-m-diam pipes, equally spaced around the circumference of the vacuum vessel. All joints are welded; and no valves are present in the loops. The inlets are located above the lithium pool, so that if a pump failure occurs in one loop, the other loops can continue to operate normally (see Fig. 8-76). Efficiency of the EM pump is conservatively assumed to be 40% (see "A Comparison of Mechanical and Electromagnetic Pumps").

Although mechanical pumps are desirable because of their advanced state of development and demonstrated high efficiency, their ability to withstand neutron-induced pressure pulsations transmitted through the liquid lithium from the fusion reactions is questionable. The internal rotating

parts of mechanical pumps are particularly susceptible to failure from repeated fluid impact and local cavitation caused by these pressure pulsations. We circumvented the problem by using EM pumps, in which the fluid essentially flows through a straight channel. Large EM pumps that have flow rates of 8 to $10 \text{ m}^3/\text{s}$ must be developed for the circulation loops; however, smaller-sized pumps (0.1 to $0.2 \text{ m}^3/\text{s}$) are utilized in numerous applications today.⁸¹

Characteristics of the liquid-metal jet array and cooling streams inside the chamber are listed in Table 8-8. The channel between the FSW and reflector contains a lithium stream and a space. Near the bottom, the lithium stream is kept adjacent to the outer surface of the FSW by flow guides attached directly to the liner. This arrangement reduces the required flow rate of lithium without appreciably increasing the temperature rise in the lithium as it passes through the chamber. Further, the adjacent space prevents direct transmission of impact forces from the fusion reaction to the reflector. Similarly, the channel between the reflector and the vacuum vessel also contains a lithium stream and a space. However, no flow guides are required because gravity acts to keep the coolant adjacent to the vacuum vessel near the bottom. The reflector is also cooled internally to keep its maximum temperature below allowable limits.

Materials Considerations. The selection of 2-1/4 Cr-1 Mo, low-alloy, ferritic steel as the structural material in the HYLIFE chamber is based on considerations of strength, fabricability, cost, resistance to corrosion and radiation damage, and susceptibility to neutron activation. This steel has adequate strength⁸² at operating temperatures up to 870 K (see "High-Cycle Fatigue Strength of 2-1/4 Cr-1 Mo Ferritic Steel at Elevated Temperatures"). It is used extensively for nuclear-power-plant heat exchangers and steam generators because of its good fabricability, low cost, and resistance to hydrogen embrittlement. Whitlow, et al.,⁸³ report that the weight loss of this alloy is approximately one-third that of 300-series stainless steel when in contact with flowing lithium at 810 K. Hot lithium attacks steel at the grain boundaries, producing exposed grains at the surface. Selle and Olson⁸⁴ suggest that material dissolution increases with nickel and chrome content; 2-1/4 Cr-1 Mo ferritic steel contains no nickel and only 2-1/4% chrome.

Because iron has a lower cross section for helium producing (n, α) reactions than does nickel,

Table 8-8. Characteristics of the lithium circulation in the modified HYLIFE chamber.

	Fluid temperature rise through chamber (K)	Lithium flow rate (m ³ /s)	Fluid velocity at nozzle plate lower surface (m/s)	Fluid velocity at midplane (m/s)	Radial thickness at nozzle plate lower surface (mm)	Radial thickness at midplane (mm)
Annular weir flow } Liquid-metal jet array }	10.3	10.6 132.0	7.1 5.5	11.4 10.4	316 300	200 190 to 215
FSW inner surface cooling	6.9	3.4	6.2	10.8	17	10
FSW outer surface cooling	5.0	3.4	6.2	10.8	17	10
Reflector internal surface cooling	6.5	7.1	—	—	—	—
Vacuum vessel inner surface cooling	1.2	3.7	6.2	10.8	17	10
		160.2				

the rate of helium production is half that of Type-316 austenitic steel. Rockwell International studied the effects of helium on the tensile properties of alloys by irradiating miniature tensile specimens with alpha particles. Austenitic samples with low helium concentrations (40 appm) lost ductility more rapidly with increasing temperature than did ferritic steels with the same helium content.^{85,86} Thus both the production rate and degrading effects of He are less in ferritic steels than austenitic steels.

The displacement-damage rate in the HYLIFE-chamber wall is essentially independent of whether a ferritic or austenitic steel is used. Radiation-induced swelling is generally assumed to be a linear function of the number of displacements per atom (dpa) after some incubation dpa. Ion-irradiation experiments of ferritic alloys indicate that they resist swelling better than Type-316 SS. The 2-1.4 Cr-1 Mo alloy showed only 0.8% swelling after 115 dpa at 825 K.⁸⁷

The absence of nickel is also an advantage with respect to neutron activation, because no ⁵⁸Co or ⁶⁰Co isotopes are generated. These cobalt isotopes have long half-lives; and their presence complicates the shielding of components in the lithium circulation loops. If nickel were present in the structural components, it would be leached and activated to form cobalt isotopes that could then be deposited throughout the lithium circulation loops.

We performed two-dimensional activation calculations of the steel, using the TARTNP (50-group) neutronics code⁸⁸ with the Fusion Reactor Isotope Production (FRIP) code.⁸⁹ Results, showing total curies from all activation products present at shutdown and total gamma-energy-emission rate

Table 8-9. Activation of components in the modified HYLIFE reaction chamber after 30 yr of operation.

	Total Ci at shutdown (Ci/kW _{th})	Total gamma energy emission rate (γ-MeV/kW _{th} -h)
Vacuum vessel	1.3	5.9 × 10 ¹³
Nozzle plate	1.6	1.0 × 10 ¹⁴
First structural wall	9.0	5.4 × 10 ¹⁴
Totals	11.9	7.0 × 10¹⁴

for the modified HYLIFE chamber, are listed in Table 8-9. Induced activity in terms of bulk curies is an important consideration for long-term storage and other environmental effects. The gamma energy output rate is coupled to the component cooling requirement immediately after shutdown and to the biological shielding requirements for hands-on maintenance. In comparison with the HYLIFE value of 11.9 Ci/kW_{th} at shutdown (Table 8-9), a Tokamak with a 316-stainless-steel wall and a liquid-metal fast-breeder reactor (LMFBR) would have 1000 and 5000 Ci/kW_{th}, respectively.

The modified design reduces the lithium inventory which in turn reduces the amount of tritium present in the reaction chamber. Both are desirable from the standpoints of safety, cost, and conservation of resources.

Summary. The HYLIFE design offers flexibility and simplified construction, which results in improved promise for economic generation of electricity from inertial confinement fusion power. A

key feature is the inclusion of a liquid metal (lithium) jet array that absorbs over 90% of the energy produced and breeds tritium to replace that used in the fusion reactions. Structural components are shielded by the jet array from debris, x-ray, and neutron exposure.

The use of jets dissipates much of the liquid momentum imparted to the lithium; thus impact loads on the FSW are minimized. All structural components are designed to last the entire 30-yr lifetime of the chamber, without replacement, using common 2-1/4 Cr-1 Mo ferritic steel. A full-diameter flange is included, which permits remote disassembly and reassembly of components, should this ever be required. Electromagnetic pumps reduce the susceptibility to damage caused by pressure pulses from the fusion reactions, relative to mechanical pumps. Lithium and tritium inventory is minimized to reduce hazards and expense.

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Alternative HYLIFE Design Options

As we have developed our calculational models and applied them to the HYLIFE-point design, we have had to make design choices that were sometimes arbitrary because of lack of information or resources. In this section, we briefly summarize some of the alternatives in four areas of HYLIFE: steam cycle, driver, liquid-metal pump, and first-wall material.

We have selected a superheated-steam cycle because of its high efficiency. In the first article of this subsection, we present the potential penalties for using a less efficient, but possibly more reliable, saturated-steam cycle.

A strong, positive feature of the HYLIFE converter is its potential to accommodate either a laser or heavy-ion-beam driver system. The higher-efficiency ion-beam driver requires a considerably lower chamber pressure for propagation; thus either the liquid-metal-wall composition or temperature must be changed. [A high-pressure (~1 Torr) propagation window has been theoretically predicted, but not verified.] In the second article, we present energy balances to show the effect of such changes on plant efficiency and circulating power.

In our 1978 design, we used low-development-risk mechanical pumps. In our current design, we use electromagnetic (EM) pumps, which have several potential operating advantages. However, because of the low level of development, the efficiency of large EM pumps is quite uncertain; we have used an estimated value of 40% in our design. In the third article we compare the two pump types, and describe a large EM pump that has a theoretical efficiency of 66%.

Finally, we briefly examine the advantages and limitations of using Nb-1% Zr for the first-structural-wall material while retaining 2-1/4 Cr-1 Mo ferritic steel for most other components.

Steam Cycle Options

We have considered reducing the lithium temperature in the reaction chamber to allow use of a heavy-ion-beam driver, to increase material strength, and to reduce corrosion. This temperature reduction lowers electrical-conversion efficiency and, hence, lowers net plant efficiency. The actual reduction in electrical-conversion efficiency depends on the steam-cycle design. Cycles with higher efficiencies are accompanied by deleterious system impacts that may balance their efficiency advantage.

Four steam-cycle and heat-transfer systems were selected for investigation. These systems are based on design parameters from General Electric and Atomics International prototype-large-breeder-reactor (PLBR) studies. The four types of steam-cycle and heat-transfer systems considered are the following:

I. A saturated-steam cycle with a constant temperature difference of 182°C between the reaction-chamber outlet and steam-turbine inlet.

II. A saturated-steam cycle in which the temperature difference between the reaction-chamber outlet and the steam-turbine inlet is reduced linearly from 182°C to 44°C.

III. A superheated-steam cycle with a constant temperature difference of 44°C between the reaction-chamber outlet and the steam-turbine inlet. Turbine inlet pressure is kept constant as reaction-chamber temperature decreases until saturated conditions occur. The turbine inlet pressure is then decreased along the saturation line.

IV. A superheated-steam cycle with a constant temperature difference of 44°C between the

Fig. 8-79. Thermal efficiency and net-plant efficiency vs. reaction-vessel lithium-outlet temperature.

reaction-chamber outlet and the steam-turbine inlet. The steam-turbine-inlet pressure is reduced as reaction-chamber temperature decreases to maintain a constant superheat of 110°C.

The calculated thermal efficiencies for the four cycles and the corresponding laser-driven-HYLIFE converter net electrical efficiencies are plotted on Fig. 8-79. (Thermal efficiency equals gross electric power divided by gross thermal power; net electrical efficiency equals net electric power divided by fusion-chamber thermal power; and the fusion-chamber thermal power is the fusion power plus its enhancement in the blanket.) From this graph, the selection of steam cycle III would logically follow. However, there are disadvantages that make the choice less sure. Both superheat cycles require higher-temperature steam-turbine operation, which we expect to lead to more turbine failures and resultant power-plant downtime. The cost of that downtime has not been quantified; however, it could be large enough to make a lower-efficiency steam cycle more economical in the long run.

Figure 8-80 shows the increase in primary and secondary heat-transfer-system flow rates as reaction-chamber outlet-temperature decreases. The increase in system size and pumping power is another cost of lower wall temperature. Here, the superheat cycles (III and IV) have a slight advantage over the saturated cycles.

Fig. 8-80. Primary and secondary coolant-loop flow rates as a function of reaction-vessel lithium-outlet temperature.

Fig. 8-81. Steam-generator heat-transfer area as a function of reaction-vessel lithium-outlet temperature.

Figure 8-81 shows the increase in steam-generator heat-transfer area required as reaction-chamber outlet temperature decreases. Steam cycle II requires much more area than the other three cycles as reactor-outlet temperature decreases.

Overall, when the reaction-chamber fluid temperature is reduced, all four cycles have lower efficiencies with higher flow rates, pumping powers,

and heat-exchanger areas. The superheated-steam cycle with constant turbine-inlet pressure until saturation (cycle III) is superior to the other three cycles in all these areas. However, its higher turbine temperatures could result in higher maintenance costs and lower availability. Detailed optimization and tradeoff studies will be required before the steam cycle or reaction chamber exit temperature can be finalized.

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Energy Balance for Heavy-Ion-Beam Driven HYLIFE Converter Systems

We have examined the effect on the power-plant energy balance of using an ion beam rather than laser-driven HYLIFE converter. The ion beam probably cannot propagate through "pure lithium" at the 500°C vapor pressure. For a pure-lithium LMW, the temperature must be reduced to 400°C to reduce the vapor pressure and permit beam propagation. Alternatively, the makeup of the LMW fluid can be changed to a composition of about 99 wt% lead-1 wt% lithium, which has a low enough vapor pressure at 500°C to permit ion-beam propagation.

Our baseline HYLIFE-converter energy balance is shown in Fig. 8-82. A 4.5-MJ, 5%-efficient driver is used with a pellet gain of 600. (At 4.5 MJ, the target design gain curves range from 100 to 900; thus we have chosen an optimistic but potentially achievable gain.) Other assumptions and calculations used to produce the baseline energy balance were described in the 1978 Laser Fusion Annual Report.⁹⁰ Operating the lithium fall at 500°C provides turbine-inlet steam conditions at 454°C and 15.2 MPa, which produces a 39%-electrical-conversion efficiency. After power that is recycled within the plant to run the laser system and auxiliary equipment is subtracted from the output power, the HYLIFE converter has a net efficiency of 33.6%.

When the temperature of the lithium LMW is reduced to 400°C so that the heavy-ion beam can propagate through the lithium vapor, the electrical conversion efficiency of the plant declines. The actual-conversion efficiency is dependent on the

Fig. 8-82. Laser-driven HYLIFE-fusion-power-plant energy balance, "pure lithium" LMW at 500°C.

steam cycle conditions chosen as discussed in the preceding article. For the purpose of comparing energy balances for the heavy-ion-beam driver, we have selected an electrical-conversion efficiency of 34%, which is in the midrange of possible efficiencies. For ease of comparison, we have used the same 4.5-MJ driver energy and gain-pellet design point as in the laser case; however, using an approximate heavy-ion-beam-driver efficiency of 25% results in a gain-driver efficiency product of 150, which is much higher than the value of 30 for the baseline laser system. Significant savings in recycled electric power are produced. Nevertheless, the net-plant efficiency is 30.8% compared with the 33.3% efficiency for the laser-driven design. A diagram of the energy balance for a HYLIFE converter using a 400°C-

lithium LMW and heavy-ion-beam driver is shown in Fig. 8-83.

The use of a liquid-metal wall made up largely of molten lead with a small portion of lithium added for tritium breeding has the advantage of allowing operation at 500°C with the heavy-ion-beam driver. However, increased pumping power is required for the liquid-metal jet array and primary heat-transfer system because of the increased fluid density (up by a factor of 18) and the reduced heat capacity (down by a factor of approximately 27). We are just beginning to explore the use of lead-lithium within HYLIFE; the energy balance shown in Fig. 8-84 is a preliminary illustration of the type of changes that

Fig. 8-83. Ion-beam-driven HYLIFE-fusion-power-plant energy balance. "pure lithium" LMW at 400°C.

will occur with the lead-lithium fall. Blanket gain increases somewhat because of the neutron multiplication effect of lead, but lithium primary pumping power increases because of the reduced heat capacity of lead. The LMW pumping power increases because of the higher fluid density, although the LMW flow rate is reduced by half because of the increased stopping power of the lead-lithium fluid. The thermal energy reclaimed as pump heating increases significantly with the added pumping power. Overall, the net efficiency of 30.4% is slightly less than the 30.8% efficiency of the 400°C lithium LMW. Redesign of the primary heat-transfer system could probably substantially reduce that

system's pumping power. (A 6% decrease in lead-lithium pumping power would increase the net efficiency to 30.8%.) The use of a lead-lithium mixture remains an attractive option if a heavy-ion beam driver is chosen within the ICF program.

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90. *Laser Program Annual Report—1978*, Lawrence Livermore Laboratory, Livermore, Calif., UCRL-50021-78 (1979) pp. 9.2 to 9.75.

Fig. 8-84. Heavy-ion-beam-driven HYLIFE-fusion-power-plant energy balance, 99.3 Wt% Pb and 0.7 Wt% Li LMW at 500°C.

A Comparison of Mechanical and Electromagnetic Pumps

Our initial design for the HYLIFE converter included large mechanical pumps to recirculate the liquid-metal flow that forms the jet array and primary heat-transfer system. We chose mechanical pumps because these large pumps are currently under extensive development for the LMFBR program. As a result, less extrapolation from the state of the art will be required than for electromagnetic (EM) pumps. In addition, large mechanical pumps probably will have higher efficiencies than large EM pumps, although this is not certain because of the lack of large EM-pump

development. An examination of EM pumps for use as full recirculation and primary heat-transfer system pumps showed them to have the following general advantages over mechanical pumps:

- No penetrations through liquid-metal containment.
- No bearings in liquid metal.
- No seals.
- No need for a cover gas within the pump.

Two further advantages of EM pumps are unique to the HYLIFE converter design. First, EM pumps will allow reduction in the volume of lithium, and hence, tritium. Second, EM pumps have no moving components in the working fluid that could be prone to cavitation damage. These

pumps will, therefore, be less affected by pressure pulses transmitted through the piping from the reaction chamber. The updated HYLIFE design discussed earlier in this set of articles uses EM pumps.

Three types of EM pumps have been used to pump liquid metal. The conduction type, called a Faraday EM pump, uses electrodes attached directly to the pipe walls. This pump is very inefficient and probably applicable only to use in the laboratory. The linear-induction type, called the Einstein-Szilard pump, is now the most commonly used EM pump for liquid metals. In this pump, coils along the outside of a pipe section produce a moving electromagnetic field within the liquid-metal flow. The third type of EM pump is the moving windings type, of which the helical-rotor is the most prominent (Fig. 8-85). In this pump, a steady-electric field is set up by a coil, and the coil is rotated by a motor to create a moving field in the liquid-metal flow, which is in an annulus around the coil. This moving-field type of pump appears to be the most efficient of the three; theoretically, it could have a higher efficiency than mechanical pumps.

In an EM pump, electromagnetic fields pass through the liquid-metal containment boundary (pipe walls) to act on the liquid metal, thereby producing the pumping action. In mechanical pumps, seals are used to separate oil from the liquid metal; a cover of gas over the liquid metal is required to protect the seals from direct contact with liquid metal. In a HYLIFE converter, some of this cover gas migrates through liquid metal to the reaction chamber. Equipment would be required to remove the cover gas from the reaction chamber. A hydrostatic-type liquid bearing is needed in mechanical pumps to stabilize the rotating elements in the liquid metal. The development of materials and designs for the liquid-metal bearing is avoided by use of the EM pump.

We expect liquid-metal volume within the EM pump to be less than that within the mechanical pump. Furthermore, since the mechanical pump is much more susceptible to cavitation damage, it requires an inlet pressure that, in HYLIFE, must be supplied by a static head. The reduced inlet pressure to the EM pump will reduce the liquid-metal volume of the system. When added to the reduction

Fig. 8-85. Helical-rotor electromagnetic-pump schematic.

in volume achievable within the pumps themselves, a significant liquid-metal volume reduction—and thus radioactive-inventory reduction—might be achieved.

Large pressure pulses are expected to result from the fusion pulses in the reaction chamber. These pulses can propagate through the liquid-metal system, causing damage to mechanical pumps. Pulse-damping mechanisms must be included in the design of the HYLIFE plant when mechanical pumps are used. The use of EM pumps may eliminate the need for pulse damping.

EM pumps are probably less efficient than mechanical pumps. An energy-flow diagram for a large liquid-metal mechanical pump is shown in Fig. 8-86. This energy flow diagram is based on actual test data for the fast-flux test-facility pumps. A similarly sized Einstein-Szilard EM pump was designed by the General Electric Company, but was not tested. However, it was designed to have a net pump efficiency of 40%, and test efficiencies are normally less than expected.

We have considered using a helical-rotor EM-pump design for HYLIFE. With this pump, there might be no efficiency disadvantage compared with mechanical pumps. An energy-flow diagram for a helical-rotor pump is shown in Fig. 8-87. However, the high pump efficiency in this case may only reflect the preliminary nature of the sign.

Alternatively, advanced mechanical pumps currently under design permit reduced liquid-metal

Fig. 8-86. Energy flow diagram for large mechanical pump for liquid-metal service based on FFTF-pump results

volume and have lower cavitation susceptibility. These advanced mechanical pumps include inducers (axial-pumping stages) for operation at higher-suction-specific speeds. In the HYLIFE plant, this allows the pumps to rotate faster, thus making a smaller, less-expensive pump. The lithium volume within the pump and within the lines to and from the chamber can be reduced because of the lower-inlet-head requirement and higher-rotation speed. The efficiency of advanced mechanical pumps remains much the same as that of current mechanical pumps.

Both pump-energy flow diagrams in Figs. 8-86 and 8-87 have a very high efficiency when recirculating power is added to the efficiency of the pump. However, capital funds must be expended to increase the size of *almost all* power plant components to circulate and thus recover the pump heat.

Both advanced-mechanical and EM pumps could have higher efficiencies than the values used in our studies (our estimates were conservative). If

Fig. 8-87. Energy flow diagram for large electromagnetic pump for liquid metal service based on helical-rotor design.

so, a principal disadvantage in using a liquid-metal wall of high-Pb concentration, Pb-Li solution as the LMW would be eliminated (see the preceding article).

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Nb-1% Zr as a First-Structural-Wall Material in the HYLIFE Converter Concept

A self-consistent comparison of the mechanical design response of the HYLIFE first structural wall was made using the current ferritic-steel wall and a Nb-1% Zr wall. Both materials were evaluated at 800 K. The energy generation per unit volume, q''' , in the Nb-1% Zr is approximately three times that in the 2-1/4 Cr-1 Mo ferritic steel.⁹¹ However, the

thermal-stress parameter, $E/\alpha q'''/[k(1-\nu)]$, is approximately the same for both alloys because of the lower elastic modulus, E , the lower linear-thermal expansion coefficient, α , and the higher thermal conductivity, k , of the Nb-1% Zr.

While refractory metals such as niobium tend to exhibit an endurance limit, such that the strain range to failure is independent of the number of cycles for a lifetime greater than,⁹² for example, 10^7 cycles; there is no evidence⁹³ of an endurance limit for 2-1/4 Cr-1 Mo. The ratio of the fatigue strength of Nb-1% Zr is 1.3 times higher than that of 2-1/4 Cr-1 Mo for a lifetime of 10^{10} cycles at 800 K. The fatigue strength of the Nb-1% Zr alloy is based on 1962 to 1963 high-temperature high-cycle fatigue experiments conducted in sodium and lithium by AiResearch Manufacturing Division, Garrett Corporation in Phoenix, Arizona.⁹⁴

If the loading of the first structural wall is caused by the impact of expanded lithium, fatigue strength is the figure of merit. Thus a Nb-1% Zr first structural wall of the same design as in the HYLIFE converter can tolerate a fusion event approximately 30% greater than the 2-1/4 Cr-1 Mo ferritic steel first structural wall.

If the impact on the first structural wall is from a noncavitating slug of liquid lithium, the figure of merit of a structural material is its fatigue strength divided by its acoustic velocity. The ratio of this figure of merit for Nb-1% Zr to that of 2-1/4 Cr-1 Mo is approximately 1.5, such that the Nb-1% Zr can tolerate one-half again as large a fusion event as the 2-1/4 Cr-1 Mo.

The use of a Nb-1% Zr alloy as the first structural wall will allow fusion events at least 30% higher than the 2-1/4 Cr-1 Mo ferritic steel using the HYLIFE converter concept. This will result in a smaller chamber or a lower pulse rate for a given overall power, and hence, a lower circulating power fraction because of the lower pumping power required.

The disadvantages of Nb-1% Zr are the increased cost and activity of the structural wall relative to a steel wall of the same dimensions. However, the Nb-1% Zr is more corrosion resistant and can operate with a higher power density than a steel-wall liner, so that the total system cost may be less. The niobium liner would be the only exotic

material used in the converter, with 2-1/4 Cr-1 Mo ferritic steel used in all other components.

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Material Considerations for Liquid-Wall ICF Energy Converters

Accurate knowledge of material properties at power-plant operating conditions throughout the plant life is crucial to the commercial application of fusion energy. Material selection and qualification is an expensive, long-lead-time procedure, which we have already begun. We have identified 2-1/4 Cr-1 Mo ferritic steel as a promising structural material for the HYLIFE converter.

Neither 2-1/4 Cr-1 Mo nor any other material is qualified in the ASME Boiler and Pressure Vessel Code for use as a fusion-power-plant first structural wall because no previous applications have combined pulsed radiation, cyclic stresses, and liquid-metal corrosion. The first article in this set of articles describes the process used to specify, code-qualify, and derate the first structural wall material. Specification of material includes determining the composition and heat treatment to be used. Corrosion resistance, radiation damage resistance, and thermal and mechanical properties are considered in this phase. Once the material is specified, rigorous mechanical testing is required for code-

qualification. Finally, the derating of properties caused by corrosion and radiation must be determined. For stainless steel used in a sodium environment, this procedure has taken 15 years, and work remains to be done. For ferritic steel to be used in lithium, we are just beginning the work; in the first in this set of articles, we describe our planning and progress.

The next two articles describe the results of 2-1/4 Cr-1 Mo ferritic steel corrosion and fatigue strength experiments at the Colorado School of Mines and the Oak Ridge National Laboratory. Then, two articles present the LLNL-developed equation of state for lithium and our preliminary analysis of the properties of lithium-lead mixtures. Finally, we describe some of the properties of graphite, which is used as a neutron reflector in our HYLIFE design.

Structural Material Selection and Qualification in a Liquid-Metal Environment

Introduction. Selection of a suitable material for containment of high-temperature lithium has included investigations in most major alloy systems.^{95,96} Pure iron has excellent corrosion resistance in the unstressed condition; however, its low strength and poor-creep resistance make it unsuitable as a structural material. Refractory metals also have good corrosion resistance; but high cost and poor fabricability discourage their use. High-nickel and cobalt alloys were investigated because of their good creep resistance; however, these alloys experience major selective dissolution attack in the presence of liquid lithium.⁹⁶

Much of the research in this area has been focused on ferrous alloys, including carbon, low-alloy, and stainless steels. Steels containing low levels of carbon (e.g., austenitic-stainless steels) have, in general, shown good corrosion resistance.

Ferrous alloys offer good mechanical properties and are available, economical, and easily joined. Regulatory codes governing the manufacture of reactor vessels from many ferrous alloys are available. The development of new ferrous alloys or the use of nonferrous alloys could cause substantial delays in the commercial development of fusion power.

We have initially selected a ferritic steel containing no nickel and only 2.25% Cr (2-1/4 Cr-1 Mo). Nickel increases the radiation damage, general corrosion in lithium, and storage time after shut-down of fusion reactor steel. Chromium is an element not found in the U.S. (about 98% of known commercial deposits are in South Africa, Rhodesia, and the U.S.S.R.).

Before 2-1/4 Cr-1 Mo ferritic steel can be used in the construction of a commercial HYLIFE converter, three stages of qualification must be completed:

- Confirmation of the corrosion resistance of the alloy and selection of a heat treatment.
- Performance of the mechanical property tests necessary to fully code-qualify the alloy and heat treatment. (The 2-1/4 Cr-1 Mo material is ASME-code qualified at lower temperatures and fatigue cycles.) These tests are not conducted in a liquid-metal or radiation environment.
- Determination of the derating of code properties caused by pulsed-radiation damage, liquid-metal corrosion, and nonstandard cyclic-loading patterns.

The structural material qualification process is long term; however, it should be carefully planned to minimize costs. For example, early in the program, we should screen a number of alloy and heat-treatment combinations using inexpensive experiments. As the number of alloys and heat treatments being considered decreases, more expensive and lengthy experiments can be used.

We have begun the material qualification program with modest experiments at the Colorado School of Mines (CSM).^{97,98} The HYLIFE material qualification program at CSM is only one part of a larger program at CSM, Oak Ridge National Laboratory (ORNL), and elsewhere to develop 2-1/4 Cr-1 Mo ferritic steels for coal-gasification systems and breeder reactors. Thus the early costs are shared among several programs.

Information Required for Alloy and Heat-Treatment Selection. Before measuring mechanical properties required for full ASME code-qualification, we must perform a variety of studies to determine the corrosion resistance and radiation damage susceptibility of the material:

- Carbide stability studies.
- Localized, severe attack by the liquid metal.
- Ductile-to-brittle transition phenomena.

- Transition joint weldments.
- General corrosion.
- Interstitial element transfer.
- Creep/fatigue and rapid-loading/fatigue interactions.

- Erosion.
- Melting practice effects.
- Radiation effects synergistic to liquid metal compatibility. Note that the interstitial element transfer and melting practice effects are still important in the area of sodium technology.

We have listed the studies in a sequence that minimizes cost by scheduling the more difficult and expensive tests after reducing the number of materials being considered. Our ongoing experiments at CSM have addressed the first seven areas. We note that in addition to preparing for the ASME-code-required testing, these studies also are the basis for the third phase of qualification (derating of properties because of corrosion, radiation effects, and nonstandard cyclic-loading).

Because the nature of the liquid-metal and steel interaction depends on the liquid-metal composition and temperature as well as the alloy and heat treatment, CSM is using lithium and a variety of lead-lithium liquids at various temperatures for the corrosion studies.

In the following paragraphs, we discuss each factor listed above.

- *Carbide stability* heads the list of these factors because the usual carbide in a ferritic steel, cementite (Fe_3C), is unstable in lithium. The precursor carbide to cementite (epsilon-carbide) is even more rapidly attacked than cementite. The precursor carbide becomes an important factor at temperatures below 500°C. Carbides that are stable in lithium solutions can be created by alloying the ferritic steel with an element such as niobium or by suitable heat treatment of the conventional ferritic steel. Both approaches are being studied.

- *Severe attacks by the liquid metal at localized sites*, such as crack tips or grain boundaries, can increase corrosion damage, and the damage will be difficult to monitor before an unexpected catastrophic failure. Localized, severe attack by the liquid metal can be a consequence of steel microstructure, interstitial impurities within the liquid, impurities at the grain boundaries within the steel and state of stress. The problem is complicated because more than one of these variables are present when localized attack is noted, and experiments that

isolate the causal variable are often extremely difficult to perform. Both heat treatments and niobium stabilization have greatly reduced severe attack along grain boundaries (see "Liquid-Lithium Corrosion of 2-1/4 Cr-1 Mo Steel Weldments").

- *The ductile-to-brittle transition (DBT) temperature* is the temperature at which the energy required to impact fracture a Charpy V-notch specimen greatly decreases with decreasing temperature. Below the DBT temperature, the material fractures in a brittle manner without absorbing much energy. Above the DBT temperature, much more energy is required, and the fracture is ductile. Therefore, we want to be sure that the HYLIFE structure is always above the DBT temperature when it absorbs impacts. The DBT phenomena can occur even at constant temperature, depending on strain rate, notch severity, absorption of interstitial elements, radiation damage, and migration of impurities to the crack site. An example of DBT dependence on temperature and strain rate is shown in Fig. 8-88. Finally, all DBT phenomena are sensitive to the microstructure of the steel.

When ferritic steels are exposed to lithium in fusion applications, each of the above factors can come into play. For example, fatigue caused by repetitive lithium impact can extend a crack, making the notch severe enough to create brittle crack propagation conditions. Then, the crack extension per cycle suddenly increases; and evidence of brittle cleavage becomes apparent when the crack surface is examined metallographically.

Atomic (rather than molecular) hydrogen diffuses into steel easily. The HYLIFE steel will be subject to hydrogen embrittlement (e.g., DBT) both by hydrogen injected into the steel during fluid impacts (since atomic hydrogen will be dissolved in the lithium), and by hydrogen produced in the steel by neutron reactions. Lithium-injected nitrogen can also embrittle the steel.

When impurities such as sulfur, oxygen, arsenic, and antimony segregate at the grain boundaries, the alloy can become susceptible to the DBT phenomena. In these cases, the brittle fracture is intergranular rather than transgranular.

The microstructure of heat-affected zones (HAZ) adjacent to welds is often quite susceptible to deleterious environmental effects. Post-weld heat

Fig. 8-88. Ductile-to-brittle transition as a function of both temperature and strain rate. High strain rate extends the brittle regime to higher temperatures.

treatments used to convert the carbides in the HAZ to stable forms alter the microstructure of the base alloy and influence DBT behavior away from the HAZ.

Any consideration of DBT phenomena must include dynamic effects such as strain rate. In cyclic-loading tests, care must be taken to produce realistic strain rates. Early indications from CSM lithium-environment sinusoidal-fatigue tests⁹⁷ suggest that, at sufficiently high pulse rates, grains cleave in a brittle mode even at temperatures as high as 500°C. A continuous-sinusoidal fatigue test produces much smaller strain rates than the impact-and-rest loading of the HYLIFE first structural wall for identical pulse rates (Fig. 8-89). These preliminary results give some indication as to propagation of fatigue cracks; but they do not address brittle fracture phenomena under static loading conditions or

crack initiation phenomena under fatigue conditions.

- *Transition joint weldments* will be required between ferritic and austenitic stainless steel components. F-M pump components, flow meters, and expansion bellows will probably be constructed of high-nickel (austenitic) stainless steel because of magnetic field and ductility constraints. The usual transition joint weldment material between ferritic steel and high-nickel stainless steel is a nickel-base alloy. However, since nickel-base alloys dissolve rapidly in lithium, an iron-base alloy will probably be needed as the transition joint material. Dilution of the ferritic steel by the transition joint alloy will occur during welding. Carbide stability, which is the most critical metallurgical consideration, will be affected by dilution of ferritic steel.

- *General corrosion* is expected to be a significant problem with high-Pb concentration liquids. Precipitation of the corrosion product, which may be radioactive, around the loop is perhaps the most serious aspect of general corrosion.

Fig. 8-89. Effect of cyclic loading pattern on strain rate. For equal frequency, a pulse-and-hold-loading pattern (solid lines) produces a higher strain rate than a continuous-sinusoidal-loading pattern (dashed line). In HYLIFE, the amplification is of order 200 rather than the factor of 3 shown here.

● *Interstitial element transfer* has, for years, dominated sodium compatibility concerns. Almost all research effort in this area has concentrated on carbon transfer. In particular, the weakening associated with carbon transfer out of the 2-1/4 Cr-1 Mo alloy has claimed the attention of metallurgists. In the case of lithium, interstitial

Fig. 8-90. The effect of frequency on crack growth rates for 2-1/4 Cr-1 Mo ferritic steel (from Ref. 97).

transfer will also occur; but hydrogen and nitrogen transfer are now thought to be more important than carbon transport. Nitrogen effects are in the initial stages of investigation.

● *Fatigue* studies are now being performed at CSM in high-nitrogen lithium in a temperature range that includes 400°C. These studies not only provide information on specific mechanical properties, but also give insight into the problem of optimizing metallurgical structure. This additional benefit derives from the extreme sensitivity of rapid-loading and fatigue effects to slight change in metallurgical parameters. One specific test in this area measures crack area advancement per cycle as a function of cyclic frequency for a sinusoidally applied stress. In lithium, the least crack advancement per cycle is often found at intermediate values of cyclic frequency. In other environments in the 500°C range, the high crack advancement per cycle is expected to occur only at low cyclic frequencies, where creep makes fatigue more severe. (Fig. 8-90).

● *Erosion* needs to be studied in terms of four phenomena:

- Droplet impact.
- Cavitation of lithium-vapor bubbles on solid surfaces.
- Impact of solid particles carried in flowing lithium.
- Corrosion and erosion by rapidly flowing lithium.

Our present understanding of lithium compatibility leads us to predict that erosion in lithium systems will differ markedly from erosion phenomena in sodium.

● *Steel melting practice effects* must be considered in studies of interstitial element transfer between liquid metals and structural alloys. We know from sodium compatibility studies that the rate of decarburization associated with vacuum melted 2-1/4 Cr-1 Mo ferritic steel is higher than the decarburization rate of electro-slag remelted alloy.

● *Pulsed radiation effects* may be more important in lithium than in sodium. Indications are that lithium compatibility is also affected by the state of stress in the structural alloy, whereas the stress state is not important for sodium compatibility.

Lead-Lithium Alloys. As discussed in the article entitled "Energy Balance for Heavy-Ion-Beam Driven HYLIFE Converter Systems," 83 at.% Pb-17 at.% Li may have advantages over pure lithium in an ion-beam-driven system. This eutectic composition is expected to act much differently than pure lithium. The large negative deviation from the chemical ideality associated with lead-lithium solutions indicates that the lithium chemical activity in the liquid is miniscule (see "Properties of Lead-Lithium Solutions" later in this section). One could estimate that this eutectic is similar to that of pure lead with respect to compatibility; but since metallic bonding has been replaced by ionic bonding to some extent, the analogy with pure lead may be misleading. Experiments are now underway on 2-1/4 Cr-1 Mo in binary Pb-Li melts ranging from pure lithium to the 83% lead composition.

Information Required for Code Qualification. Selection of a new alloy or of a new heat treatment of an alloy means that initial vessel designs cannot be performed "according to code." In practice, new alloys or unusually heat-treated alloys used in pressurized or nuclear systems must be added to the accepted codes. This requires years of mechanical property measurement and experimentation to obtain physical properties. All measurements must be repeated enough times to have statistical validity. The material properties required to perform a stress analysis using the accepted design codes are shown in Table 8-10. For other than an isothermal

analysis, the specific heat, thermal diffusivity, thermal emissivity, and thermal conductivity of the material are also required.

When a material must be added to the ASME code rapidly, a "code case" may be initiated, but the code case is effective for three years only. The procedure for obtaining approval of a material by "code case" requires five conditions.

● A pending ASTM specification for the material.

● Material commercially available to specification.

● Both reasonable demand for material by industry and concurrent urgency for approval by means of a code case.

● Request for approval clearly describing material in ASTM specification form.

● Inquirer furnishing the code committee with certain data as specified.

Degrading Caused by Liquid Metal, Radiation, or Nonstandard Fatigue Loading. Degradation of properties in an environment (such as lithium) is handled in the codes by making the "owner" of a plant responsible for considering property degradation. The code only concerns itself with a material before use in an application. This point is illustrated below by the following quote from the code.

From Article NCA-1000, Scope of Section III
"...The rules of this Section provide requirements for new construction and include consideration of mechanical and thermal stresses due to cyclic operation. They do not cover deterioration which may occur in service as a result of radiation effects, corrosion, erosion or instability of the material. These effects shall be taken into account with a view to realizing the design or the specified life of the components..."

Considerations such as radiation damage and corrosion by liquid lithium are vital to the designer of a fusion plant, but they do not impact the lengthy process of getting a material into the ASME code.

Summary. To qualify a ferritic steel in our pulsed-radiation, cyclic-loading, liquid-metal environment, we must initially explore the corrosion and radiation susceptibility of various alloys and heat treatments. When enough understanding is achieved to select the alloy and heat treatment, specific properties must be measured in a standard environment (without radiation, liquid metal, or nonstandard cyclic loading) for extension of the

Table 8-10. Data required for inclusion of the structural material in design codes.

ASME boiler and pressure vessel code (ASME B & PVC)				
	Section III		Section VIII Division I	ANSI B 31.1 Power piping code
	Class 1	Classes 2 & 3		
Minimum tensile strength at room temperature	↑	↑	↑	↑
Minimum yield strength at room temperature	↑	↑	↑	↑
Tensile strength at operating temperature	Yes	Yes	Yes	Yes
Yield strength at operating temperature	Yes	Yes	Yes	Yes
Modulus of elasticity at operating temperature	Yes	Yes	Yes	Yes
Shear modulus	Yes	Yes	Yes	Yes
Poisson's ratio	Yes	Yes	Yes	Yes
Fatigue curves at temperatures of interest (continuously cycled and with hold time)	Yes	Yes	Yes	Yes
Density	Yes	Yes	Yes	Yes
Coefficients of expansion	Yes	Yes	Yes	Yes
For temperatures above the creep range:				
Av stress for a creep rate of 0.01%/h	—	Yes	Yes	Yes
Av and minimum stresses for rupture at 10 ⁵ h	—	Yes	Yes	Yes
Stress vs time-to-rupture curves for temperature and hold times of interest	Yes	—	—	—
Isochronous stress-strain curves for temperatures and hold times of interest	Yes	—	—	—
Tertiary-creep-initiation stress level for temperatures and hold times of interest	Yes	—	—	—

Notes: Section III, ASME B & PVC is for nuclear pressure vessels, Class 1 is the most stringent of the three classes.
Section VIII, Division I ASME B & PVC is for non-nuclear pressure vessels.

code to the required temperatures and number of cycles. Simultaneously, the effects of the nonstandard environment must be measured to allow safe, licensable design.

We are now in the early stages of the qualitative understanding phase with experiments and analyses at the Colorado School of Mines and the Energy Technology Engineering Center. Since these studies are also being done for other energy and industrial programs, our present annual cost is low. However, as the alloy and heat-treatment matrix is reduced and as the testing program becomes specific to our application, our funding must expand. In addition, questions such as the effects of prolonged pulsed 14-MeV neutron radiation may be unanswerable without major new test facilities.

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Liquid-Lithium Corrosion of 2-1/4 Cr-1 Mo Steel Weldments

Investigations of the susceptibility of 2-1/4 Cr-1 Mo ferritic steel to corrosion by liquid lithium at the Metallurgical Engineering Department of the Colorado School of Mines continue to be supported

Fig. 8-91. Corrosion of 2-1/4 Cr-1 Mo ferritic steel by 755-K lithium.

by I.I.I. During the last year, we have emphasized evaluation of the effect of post-weld heat treatment and niobium additions on lithium corrosion susceptibility.

Briefly, the procedure has been to expose small gas-tungsten-arc (GTA) weldments, post-heat treated under various metallurgical conditions, to static liquid lithium containing 17.6% of lead by weight. Lead is a candidate material for use in heavy-ion-beam-driven fusion pellets. The exposure temperature for these tests is 775 K; exposure times of up to 1600 h are used. Low nitrogen levels of ~50 ppm were maintained throughout these experiments.

The metallurgical conditions for weldments of both 2-1/4 Cr-1 Mo ferritic steel and 2-1/4 Cr-1 Mo ferritic steel with a small amount of Nb added (niobium-stabilized), were varied by post-weld heat treatments representing both the minimum and maximum temperatures specified by ANSI code.

Isothermally transformed and tempered coupons were preheated, GTA-welded, and then given one of three post-weld heat treatments: (1) none, (2) 1 h at 85 K, or (3) 10 h at 1035 K. The susceptibility of these various metallurgical conditions to lithium corrosion are summarized in Fig. 8-91.

Weldments of the niobium-stabilized 2-1/4 Cr-1 Mo were found to be quite resistant to corrosion by lithium-17.6% lead at 775 K. Only slight penetration (near the limit of measurement) was observed after 1600 h; the heat-affected zones of the welds were not preferentially attacked. Post-weld heat treatments did not significantly improve the performance of the niobium-stabilized 2-1/4 Cr-1 Mo ferritic steel weldments.

In contrast, the effects of thermal stabilization are significant for 2-1/4 Cr-1 Mo that contains no niobium. Without post-weld heat treatment, heat-affected zones of 2-1/4 Cr-1 Mo GTA weldments were observed to be very susceptible to lithium attack. Rapid intergranular penetration along previous austenitic grain boundaries occurred. This

penetration is thought to be associated with grain boundary films of Fe_3C , a compound that is thermodynamically unstable in the presence of lithium. Figure 8-91 graphically illustrates how post-weld heat treatment (which has been shown to convert Fe_3C to the more stable carbides M_{23}C_6 , Mo_2C , and M_6C) improves the lithium corrosion resistance of 2-1/4 Cr-1 Mo ferritic-steel weldments. A long-time, stress-relief heat treatment at 1035 K is shown to produce a weldment almost as resistant to lithium attack as a niobium-stabilized alloy. Although both the thermally stabilized, conventional 2-1/4 Cr-1 Mo and the Nb-stabilized alloy are resistant to lithium corrosion, the conventional steel is favored since it results in less neutron activation.

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High-Cycle Fatigue Strength of 2-1/4 Cr-1 Mo Ferritic Steel at Elevated Temperatures

The inner structures of inertial-fusion energy converters will experience pulsed loads for a very large number of cycles. Thus the lifetime of these structures will be determined in part by their fatigue strength. We have chosen a low-alloy ferritic steel (2-1/4 Cr-1 Mo) for the reaction chamber because of its corrosion resistance to lithium, low activity, and low susceptibility to radiation damage. The engineering-design criterion for this material may be dominated by the low stress levels required to attain a long fatigue life. Accordingly, we have obtained from ORNL⁹⁹ new high-temperature, high-cycle-fatigue strain-range data for 2-1/4 Cr-1 Mo that they have characterized for the design of nuclear steam generators. These fatigue data were accepted by the ASME for consideration of inclusion in ASME Code Case N-47.¹⁰⁰

The data considered by ORNL were limited to the following criteria:

- Material conformed to the applicable ASME specifications on chemical and mechanical properties.

- Data were from fully reversed, push-pull, constant-amplitude, strain-controlled fatigue tests.

- Data were obtained at a strain rate approximately equal to $4 \times 10^{-3} \text{ s}^{-1}$.

The strain range data were fit to a Manson-Coffin power law of the form

$$\Delta \epsilon_e + \Delta \epsilon_p = A_e(T) N_f^{c_e} + A_p(T) N_f^{c_p} \quad (60)$$

that relates the elastic and plastic strain ranges, $\Delta \epsilon_e$ and $\Delta \epsilon_p$, to the cycles to failure, N_f , using log-centered regression analyses. The exponents on the power law were independent of the temperature, while the coefficients were a function of temperature.

No systematic temperature dependence on the cycles to failure for a given strain range occurs below 700 K and in the range between 811 and 866 K. For temperatures between 700 and 811 K, ORNL recommends that the strain range be interpolated linearly in $\log \Delta \epsilon$ vs $1/T$ space for a given cyclic life.⁹⁹ Since it is likely that the high-cycle fatigue strength at 866 K is actually lower than at 811 K, we have used the temperature scaling for the entire temperature range.

The fatigue strength of 2-1/4 Cr-1 Mo as a function of temperature for various cycle lifetimes is shown in Fig. 8-92. The temperature dependence of Young's Modulus is included in the determination of stress. For an ASME-pressure-vessel-code design,^{100,101} the fatigue limit derived from the data is reduced by a factor of two as a safety margin.

The HYLIFE-converter first structural wall must operate for a lifetime of between 10^9 and 10^{10} cycles at temperatures between 770 and 840 K. Note that the reduction of the strength for an increase from 10^9 to 10^{10} cycle is less than 10%, while an increase in temperature from 770 to 840 K results in a fatigue strength reduction of 21%. Finally, based on the ASME procedure for 10^{10} cycles and peak-wall temperatures of 740 and 840 K, we must design for maximum impact stresses of 80 to 50 MPa, using the ASME design procedure. While using only the fatigue-strength curves (no safety factor), we must design for maximum-impact stresses of 160 to 100 MPa.

Author: J. Hovingh

Major Contributors: J. H. Pitts and J. A. Blink

Fig. 8-92. Fatigue strength of 2-1/4 Cr-1 Mo steel as a function of te

various cycle lifetimes.

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Analytic Lithium Equation of State

Our equation-of-state (EOS) model includes the liquid, liquid-vapor, and ionized-gas regions. Pressure (P), temperature (T), and sound speed (C) are computed as functions of density (ρ) and specific internal energy (e).

We first constructed accurate tables of P and e for lithium over the range $10^{-6} \leq \rho \leq 2 \text{ Mg m}^{-3}$ and $454 \leq T \leq 3.7 \times 10^8 \text{ K}$. Such a wide range of density and temperature cannot be handled with any single equation-of-state model; therefore, our tables derive from three separate models. These include a soft-sphere model for the low-pressure liquid and

vapor region, an ionization-equilibrium model for low-density-ionized gas, and a pseudo-potential model for the liquid metal up to high density and temperature. A detailed discussion of these models and their assembly into a tabular-data base is given in Ref. 102.

To numerically integrate the equations of motion, it is convenient to have pressure as an analytic function of density and energy. Our method is to divide the (ρ, e) plane into regions where analytic forms $P(\rho, e)$ known to be adequate can be used. These are then fitted smoothly together to form a complete equation of state. We take the melting point ($T = 454 \text{ K}$, $\rho_0 = 0.518 \text{ Mg m}^{-3}$) to be the zero of energy.

There are three principal regions:

1. (liquid) $\rho > \rho_0$ for all e .
2. (gas) $\rho \leq \rho_0$ and $e > e_c = \text{cohesive energy}$.
3. (two-phase) $\rho \leq \rho_0$ and $e \leq e_c$.

These regions are shown in Fig. 8-93. Note that Region 3 is the most complex and is subdivided into further regions.

Region 1 (Liquid). An adequate model for Region 1, with $\rho > \rho_0$ and energies up to the maximum expected in these calculations, is the

Fig. 8-93. The energy-density plane is divided into three regions for constructing an analytic equation of state.

Grüneisen model.¹⁰³ For our purposes, it is convenient to use the shock Hugoniot as a reference instead of the zero-Kelvin isotherm:

$$P = P_H \left(1 + \frac{\gamma_0 x}{2} \right) + \gamma_0 \rho_0 e \quad (61)$$

where

$$P_H = \rho_0 C_0^2 \gamma_0 / (1 - S \gamma_0^2)$$

and

$$x = 1 - \frac{\rho_0}{\rho}$$

Here C_0 is the sound speed at the melting point, S is the slope of the shock velocity-particle velocity line and γ_0 is the Grüneisen parameter. The values of these parameters are listed in Table 8-11. The melting point of lithium is chosen as the standard state. Equation (61) is in satisfactory agreement with our data base up to pressures of 100 GPa.

Region 2 (Gas). In Region 2, the material lies in the fluid state above the liquid-vapor region. We want the pressure to join smoothly to the Grüneisen model at $\rho = \rho_0$ and to the ideal gas at $P = 0$. Since the equation of state for an ideal gas with no inter-

Table 8-11. Material parameters for the analytic lithium EOS.

ρ_0	0.518 Mg/m ³
ν_0	0.9
C_0	4.5 km/s
S	1.113
R	1.198 kJ/kg-K
e_c	23.03 MJ/kg
e_l	14 MJ/kg

Fig. 8-94. A comparison of pressure vs. specific internal energy at $\rho = 10^{-6}$ Mg/m³ for the ideal gas model, the tabular-data base values, and our analytic fit to the data base.

nal degrees of freedom is $P = (2/3)(e - e_c)\rho$, a suitable form of the EOS in Region 2 is

$$P = \frac{2}{3}(e - e_c)\rho + \left[\gamma_0 \rho_0 e - \frac{2}{3}(e - e_c)\rho_0 \right] (\rho/\rho_0)^k \quad (62)$$

The exponent $k = 1.5$ was determined from the best overall fit to the tabular data base. Since the specific energy of the low-density gas in the reactor cavity can reach 10 GJ/kg, the lithium atoms can be nearly completely ionized. Although $P = (2/3)(e - e_c)\rho$ is correct for both the low-temperature (atomic Li) and high-temperature (fully ionized Li) limits, there are strong deviations from ideality in the partial ionization region as shown in Fig. 8-94. The two

regions of strong deviation represent ionization of the $2S^1$ and $1S^2$ electron shells

The deviations are sufficiently large that they should be taken into account, which we have done by fitting the bimodal pressure curve in Fig. 8-94 with a pair of log-normal functions. Thus,

$$P = \frac{2}{3}(e - e_c) \rho + g(e) + \left[\gamma_0 \rho_0^* \left(\frac{2}{3}(e - e_c) \rho_0 g(e) \right) (\rho / \rho_0)^h \right], \quad (63)$$

where

$$g(e) = 1 - 0.7 \exp[-2(\gamma - 1.233)^2] \\ - 0.7 \exp[-0.5(\gamma - 4.468)^2]$$

and

$$\gamma = \ln(e/e_c).$$

Equation (63) is compared with Eq. (62) and with the tabular data base in Fig. 8-94.

Region 3 (Two-phase). Region 3 contains the expanded liquid and the liquid-vapor, two-phase region. Here we need functions that show the sharply varying behavior characteristic of the two-phase boundary.

In the slightly expanded liquid phase ($\rho \gtrsim \rho_0$), an appropriate simple form is

$$P = \rho_0 \epsilon_0^2 (\rho / \rho_0 - 1) + \gamma_0 \rho_0 e. \quad (64)$$

Equation (64) joins smoothly with the Region 1 EOS [Eq. (61)]; and the linear-density dependence is adequate for the narrow-density range over which Eq. (64) holds true. In the high-density part of the two-phase region itself, we have fitted a Clausius-Clapeyron-like form to the tabular data base:

$$P = P_c \exp(1.4 - 1.481 e_c / e), \quad (65)$$

where $P_c = 100 \text{ MPa}$. At lower densities, this density-independent form must be replaced by a density-dependent pressure that drops toward zero

faster than the ideal gas in Region 2. Again, a simple form in rough agreement with the data base is

$$P = \gamma_0 \rho_0 e^{\epsilon_0 / \rho_0} (\rho / \rho_0)^h, \quad (66)$$

Equation (66) is continuous with the Region 2 EOS [Eq. (63)] at $e = e_c$. The region of validity of Eqs. (64), (65), and (66) is determined by their points of intersection. One further algebraic form is needed to make Eq. (65) join smoothly with Region 2. This form is constructed by interpolating between Eq. (63) and Eqs. (64) to (66) across the gap $e_c < e < e_1$. The equation of state is now complete. The function $P(\rho, e)$ over the range $0.1 e_c < e < 10 e_c$ is shown in Fig. 8-95.

Temperature. For radiation-transport calculations, we need the temperature $T(\rho, e)$ for each point on the (ρ, e) plane. In Region 1, we use the following simple algorithm with the temperatures being given in Kelvins:

$$T = (e - e_0) / 3R, \quad (67)$$

where

$$e_0 = e_{00} + \epsilon_0 e_{00} \gamma_0 + \frac{1}{2} \left(\epsilon_0^2 + \gamma_0^2 e_{00}^2 \right) \gamma_0^2 + \frac{1}{6} \left(4 \epsilon_0^2 + \gamma_0^2 e_{00}^2 \right) \gamma_0^3$$

and

$$\epsilon_{00} = 3R(454) = 1362R.$$

In Region 2 (gas), we use

$$T = T_g + (\rho / \rho_0)^{0.2} (T_s - T_g), \quad (68)$$

where

$$T_g = (0.5 + 3.5 e_c / e) (e - e_c) / 6R$$

and

$$T_s = e / 3R + 454.$$

In this region, however, the temperature may not be strictly a function of the specific-internal energy and the density. For high values of temperature and low values of density, the matter energy will constitute only a small fraction of the total, the balance being

Fig. 8-95. Pressure as a function of density for various values of specific internal energy.

in the form of radiation. Our hydrocode supplies the density and the combined energy, $e_T = e + (a/\rho)T^4$, where $a = 4\sigma/c$; σ is the Stefan-Boltzmann constant and c is the speed of light. We then substitute Eq. (68) and effect an iterative solution for e and T . This procedure is unnecessary in Regions 1 and 3 because the density is too high in the former and the temperature is too low for the radiation term to be important in the latter. When the radiation average is computed, the radiation contribution to the pressure, $aT^4/3$, is included in the total pressure supplied to the hydrocode. The term T_g in Eq. (68) deals approximately with the transition

from an atomic Li gas (1 particle/atom) at low temperature to a fully ionized gas (4 particles/atom) at high temperature.

In Region 3 (two-phase), we use

$$T = (\rho/\rho_0)^{0.2} (e/3R + 454). \quad (69)$$

The three temperature formulas [Eqs. (67) to (69)] are adequate for the computation of radiation

transport; but they are not thermodynamically consistent with the more complex pressure equation of state.

Sound Speed. The sound speed in any region derives simply from

$$C(\rho, e) = \left[\left(\frac{\partial P}{\partial \rho} \right)_e + \frac{P(\rho, e)}{\rho^2} \left(\frac{\partial P}{\partial e} \right)_\rho \right]^{1/2}. \quad (70)$$

The appropriate pressure function is used to evaluate Eq. (70).

Summary. Overall, the low-pressure liquid, two-phase, and vapor regions are adequately represented since they are approximately fitted to an accurate table. Also, we have taken pains to fit the high-temperature ionized gas with reasonable accuracy since this region can be entered by the hot gas in the reactor cavity. The most uncertain region is the dense liquid at high temperatures. At pressures above 100 GPa, the Grüneisen model breaks down, and there is no simple model to replace it.

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Properties of Lead-Lithium Solutions

Researchers at the Energy Technology Engineering Center have reviewed the properties of Pb-Li solutions as a function of temperature and Pb concentration in the lithium. These solutions are of interest because Pb will be present to some extent in both ion-beam and laser-driven ICF targets; therefore, Pb will be present as an impurity in a flowing-lithium wall. In addition, Pb-Li solutions containing ~80 at.% Pb are strong candidates for an ion-beam-driven HYLIFE converter and a viable alternative to a "pure" Li wall for a laser-driven converter. We summarize the properties of Pb-Li solutions below: design tradeoffs between Li and

Table 8-12. Phases of the Pb-Li system at 500°C.

Composition (w/o Pb)	Phase
0 to 60	liquid
60 to 88	liquid plus solid
88 to 91.8	solid
91.8 to 95	liquid plus solid
95 to 100	liquid

Pb-Li solutions are discussed in the article "Energy Balance for Heavy-Ion Beam-Driven HYLIFE Converter Systems."

Liquid-to-Solid Transitions. Although Li melts around 180°C and Pb melts at 327°C, the Pb-Li system at 500°C is not liquid at all compositions (Table 8-12). We note the presence of three eutectics in the Pb-Li system:

- At 23 wt% Pb, between nearly pure Li and $\text{Li}_{22}\text{Pb}_3$, with a 177°C-melting point.
- At 95 wt% Pb, between Li_8Pb_3 and a solid solution of LiPb deficient in Pb, with a 464°C-melting point.
- At 99.3 wt% Pb, between stoichiometric Li-Pb and a solid solution of Pb that contains lithium, with a 235°C-melting point.

As a liquid of a given composition is slowly cooled below its liquidus temperature, a solid of a different composition will begin to precipitate and the liquid composition will shift. (At a eutectic, the solid and liquid compositions are identical.) Phase diagrams indicate the temperatures and compositions at which solids can precipitate from a cooling-liquid solution of any composition. In Fig. 8-96, we show the regions around the 23 wt% Pb and 99.3 wt% Pb eutectics.

On the low-Pb side of the figure, we are uncertain as to the exact position of the liquidus line which delineates the compositions at which $\text{Li}_{22}\text{Pb}_3$ solid can exist in the liquid; however, that line is approximately correct. This curve shows that if we slowly cool liquid lithium with a Pb content greater than zero but less than 23 wt% (equals 1 at.%) Pb, solid, pure lithium will precipitate out before massive solidification at the eutectic composition. Thus if we attempt to "cold trap" liquids with less than 23 wt% Pb between 177°C and 180.7°C, we would increase the Pb content of the solution toward the eutectic and trap solid lithium. Cold-trapping liquids with compositions somewhat over 23 wt% Pb will decrease the Pb content of the liquid

Fig. 8-96. Phase diagram of the Pb-Li system for Pb concentrations less than 40 and greater than 99 Wt%. Eutectics are shown at 23 and 99.3 Wt% Pb.

toward the eutectic composition and trap the solid $\text{Li}_{22}\text{Pb}_5$. Therefore, we cannot cold trap Pb-Li liquids to under 23 wt% Pb.

Note that cold trapping may be required to keep other impurities, such as halide atoms, at a low level. In addition, the presence of impurities, even in the hundreds of ppm range, may allow cold trapping of Pb compounds with those impurities. Finally, the liquidus lines are such that no unwanted precipitation of any binary Pb-Li compound can occur at temperatures even as low as 250°C until a Pb-concentration exceeding 35 wt% is reached. This is a definite operating advantage.

Turning to the high-Pb side of the figure, we first note that the exact composition of the highest Pb eutectic cannot be established from the literature to closer than ± 0.05 wt%. The gap representing all liquid phase at 250°C is only ~ 0.2 wt% wide for this eutectic as compared with an ~ 35 wt% wide gap for the 23 wt% Pb. This means that for the high-Pb eutectic, Pb buildup of only ~ 0.1 wt% in the liquid from target debris can be cold trapped using a small side stream cooled to $\sim 250^\circ\text{C}$. At 300°C, a 0.5 wt% Pb increase above the eutectic would precipitate out solid lead. At temperatures above 327°C, increasing the Pb content above the high-Pb eutectic will not

cause solid to form. Thus loop operation with the 99.3 wt% Pb eutectic should probably be confined to minimum temperatures greater than 327°C unless cold-trapping for Pb removal is included.

Liquid-to-Vapor Transitions. If the Pb-Li system obeyed the ideal solution law (Raoult's law), the vapor pressure above a Pb-Li solution would be

$$P = N_{\text{Li}} P_{\text{Li}} + N_{\text{Pb}} P_{\text{Pb}} \quad (71)$$

where N_{Li} and N_{Pb} are the atom fractions of lithium and lead in the liquid; and P_{Li} and P_{Pb} are the vapor pressures of the pure elements at the given temperature. Since HYLIFE-converter concepts based on heavy-ion-beam drivers are sensitive to the total metal-vapor pressure, we investigated¹⁰⁴ the ideality of Pb-Li liquids. In an actual solution, both terms in Eq. (71) are multiplied by functions of the liquid composition and temperature. If the functions are 1.0, the solution is "ideal"; if the functions are greater than 1.0, the solution is said to have "positive deviation from ideality" and a higher than expected vapor pressure. In the case of the Pb-Li system, we found electrochemical data¹⁰⁵⁻¹⁰⁸ that

Fig. 8-97. Vapor pressure above Pb-Li liquids at 500°C as a function of liquid composition. The curve labeled actual is based on electromotive force data. For the 83 at.% Pb eutectic, the vapor pressure is approximately 2% of the pressure predicted by Raoult's law.

indicate a profound negative deviation from ideality.

We can see the engineering impact of this negative deviation by comparing the ideal and actual vapor pressures of Pb-Li solutions at 500°C (Fig. 8-97). The profound negative deviation from ideality means that the total vapor pressure of the high-Pb eutectic (83 at.% = 99.3 wt% Pb) at 500°C is down from that predicted from ideality by a factor of ~40. A total vapor pressure of approximately 1.3×10^{-5} Torr (1.7 mPa) at 500°C is indicated, which is less than for pure molten Pb vapor (1.58×10^{-5} Torr = 2 mPa) or pure Li ($\sim 3.3 \times 10^{-3}$ Torr = 0.44 Pa).

The curves of Fig. 8-98 show the vapor composition associated with a given liquid composition at 500°C. For the 83 at.% Pb eutectic liquid composition, the negative deviation from ideality results in a vapor that is rich in Pb (~96 at.% Pb) rather than lithium; ideal solution vapor would be only ~3 at.% Pb.

The standard method of presenting vapor pressure and vapor concentration is shown on Fig. 8-99. To use this figure, select the liquid composition on the x-axis, move up to the liquid curve and read the vapor pressure. Then move across to the

Fig. 8-98. Vapor composition above Pb-Li liquids at 500°C as a function of liquid composition. At the 83 at.% Pb eutectic, the vapor is almost pure Pb compared with the almost pure Li vapor predicted by Raoult's law.

Fig. 8-99. Vapor pressure and composition as a function of liquid composition plotted in the standard manner.

vapor curve and read the vapor composition from the x-axis.

The profound negative deviation from the ideal solution in terms of vapor pressure also applies to chemical activity. Thus, if a pure-Li system is converted to an 83 at.% Pb eutectic, the chemical activity of the 17 at.% Li of the solution will be reduced by a factor of 1400 (the negative deviation

Fig. 8-100. Density of the Pb-Li system for Pb concentrations below 60 wt%.

of the lithium component). The remaining 83 at% will be reduced from the chemical activity of pure Li to approximately that of pure Pb.

The negative deviation from an ideal solution is thought to be caused by conversion from metallic to ionic bonding. If so, the electrical conductivity will be reduced and electromagnetic pumping will be more expensive. Conversely, MHD head losses caused by fluid direction changes in a static-magnetic field will be reduced.

Liquid Properties. Engineering properties of most interest to HYLIFE converter designers include density, specific heat, and sound velocity. The energy requirements for the fall pump and, to a lesser extent, the primary heat-transfer circuit are quite sensitive to liquid density. In Fig. 8-100, the density of liquid Pb-Li at 500°C is given as a function of weight percent Pb for low concentrations. The density of liquid at the temperature at which solid just begins to precipitate (i.e., the liquidus temperature) is also given. Note that the specific gravity of melt at liquidus temperature exceeds 1.0 above ~52 wt% Pb. Reactions of hot reactive metals with water are usually milder when the freezing liquid is able to sink under the water.

Fig. 8-101. Sound speed in the liquid Pb-Li system at 500°C vs wt% Pb.

Based on experimental data at 99.175 wt% Pb and pure Pb liquid, we estimate the density of the high-Pb eutectic to be 9.3, 9.23, and 9.16 Mg m⁻³ at 300, 400, and 500°C, respectively.

We have found no data as yet on heat capacity of the high-Pb eutectic.

Speed-of-sound measurements are important for both liquid-hammer calculations and in calculating the time sequence of events within the HYLIFE converter chamber. This quantity is plotted for lithium-rich solutions at 500°C in Fig. 8-101 as a function of weight percent Pb.

Authors: N. J. Hoffman (Rockwell International, Energy Technology Engineering Center) and J. A. Blink

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Fig. 8-102. Irradiation-induced volume change in graphite (initial porosity = 5%).

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Properties c. Graphite

To maximize the neutron economy of the HYLIFE converter, a graphite-neutron reflector is used between the first structural wall and the vacuum vessel. To aid in the design of this reflector, we are collecting design data for various graphites.

The crystalline structure of graphite is generally highly anisotropic. For this reason, radiation-induced property changes are strongly related to the structure. An interstitial occupying a position between basal planes, has a very low ac-

Fig. 8-103. Irradiation-induced volume change in graphite (initial porosity = 20%).

tivation energy of 0.45 eV for motion parallel to the basal planes, but a large energy (2.8 eV) is required for motion across the planes.¹⁰⁹

Figures 8-102 and 8-103 (from Ref. 109) show the behavior of graphite with 5 and 20% initial porosity irradiated at temperatures between 400 and 1600°C. We can see that in the case of 5%-initial porosity at 400°C, the volume increases by 7% at a dose of 50 dpa. In the case of 20%-initial porosity, there is a decrease in volume that levels at ~7% in the 20 to 50 dpa range.

Tests have been conducted on three kinds of graphite. For H-451 graphite, thermal and mechanical properties were measured at several

irradiation temperatures of up to 1800 K with neutron fluences up to $1.1 \times 10^{26} \text{ n/m}^2$ (11 dpa). H-451 nuclear-grade graphite made by Great Lakes Carbon Corporation is a near-isotropic, coarse-grained, extruded product that was developed for the HTGR. It has a density of 1.77 Mg/m^3 (77% of theoretical density). After continuous fast-neutron irradiation, the H-451 first shrinks, then expands.¹¹⁰ Another near-isotropic nuclear-grade graphite was tested at Dounreay Fast Reactor between 600 to 1000 K at neutron fluences to $5 \times 10^{26} \text{ n/m}^2$ (50 dpa).¹¹¹ Similar results were obtained.

Table 8-13(a). H-451 graphite: thermal and mechanical properties.

Property	Direction in log	Unirradiated value at:					Value of irradiation to 10^{26} n/m ² at:				
		300 K	800 K	1300 K	1800 K	2300 K	300 K	800 K	1300 K	1800 K	2300 K
Thermal conductivity (W/m-K)	Axial	141	82	58	48	42	—	25	34	41	42
	Radial	134	79	55	46	40	—	23	31	39	40
Thermal expansivity (10^{-6} K ⁻¹)	Axial	4.0	4.0	4.6	5.0	5.4	—	4.6	2.6	2.5	—
	Radial	4.5	4.5	5.1	5.5	5.9	—	5.2	2.9	2.8	—
Specific heat, C_p (J/kg-K)	—	710	1620	1890	1990	2050	710	1620	1890	1990	2050
Tensile strength (MPa) ^a	Axial	13.7	15.4	17.1	18.9	20.6	—	23.1	28.4	26.7	—
	Radial	10.8	12.2	13.5	14.9	16.2	—	18.3	22.4	21.1	—
Flexural strength (MPa) ^a	Axial	20.3	22.9	25.4	28.0	30.5	—	34.4	42.1	39.6	—
	Radial	17.4	19.6	21.8	24.0	26.1	—	29.4	36.2	33.9	—
Compressive strength (MPa) ^a	Axial	51.9	58.4	64.9	71.4	77.9	—	87.6	107.6	101.0	—
	Radial	49.6	55.8	62.0	68.4	74.9	—	83.7	102.8	96.7	—
Young's modulus (GPa) ^a	Axial	7.9	8.5	9.1	9.7	10.3	—	19.1	25.0	19.4	—
	Radial	6.9	7.4	8.0	8.5	9.0	—	16.7	22.0	17.0	—
Poisson's ratio ^a	All	0.11	0.11	0.11	0.11	0.11	—	0.11	0.11	0.11	—
Tensile fatigue limit (99% survival to 10^5 cycles, MPa) ^{a,b}	Axial	6.0	6.8	7.5	8.3	9.0	—	11.8	14.5	13.6	—
	Radial	5.4	6.1	6.8	7.4	8.1	—	9.9	12.1	11.4	—

^aData at high temperatures extrapolated.^bTension-compression stress cycle.

Table 8-13 (b). POCO AXF graphite: thermal and mechanical properties.

Property	Unirradiated value at:					Value of irradiation to 10^{26} n/m ² at:				
	300 K	800 K	1300 K	1800 K	2300 K	300 K	800 K	1300 K	1800 K	2300 K
Thermal conductivity (W/m-K)	100	63	45	37	32	—	18	25	31	32
Thermal expansivity (10^{-6} K ⁻¹)	7.1	7.1	7.6	8.1	8.5	—	7.7	2.4	2.8	—
Tensile strength (MPa)	33	37	41	46	50	—	56	67	61	—
Flexural strength (MPa)	55	62	69	76	83	—	95	113	101	—
Compressive strength (MPa)	110	124	137	152	165	—	189	225	201	—
Young's modulus (GPa)	11.9	12.8	13.7	14.6	15.5	—	29.8	36.8	25.6	—
Poisson's ratio	0.25	0.25	0.25	0.25	0.25	—	0.25	0.25	0.25	—
Tensile fatigue limit (99% survival to 10^8 cycles, MPa)	16	17	19	22	24	—	—	—	—	—

Table 8-13 (c). LTI pyrocarbon: typical thermal and mechanical properties.^a

Property	Unirradiated value at:					value of irradiation to 10^{26} n/m ² at:				
	300 K	800 K	1300 K	1800 K	2300 K	300 K	800 K	1300 K	1800 K	2300 K
Thermal conductivity (W/m-K)	10.0	7.1	7.5	7.5	7.5	10.0	7.1	7.7	12.1	12.8
Thermal expansivity (10^{-6} K ⁻¹)	3.8	5.7	—	—	—	—	6.0	—	4.0 ^b	—
Tensile strength (MPa)	250	280	320	290	265	—	330	380	350	—
Flexural strength (MPa)	380	420	480	440	400	—	500	575	530	—
Compressive strength (MPa)	760	840	960	880	800	—	1000	1150	1060	—
Young's modulus (GPa)	18	19.4	20.7	20.0	18.5	—	50.0	50.0	36.0	—
Poisson's ratio	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22	0.22
Tensile fatigue limit (MPa, 99% survival to 10^8 cycles)	250	280	320	290	265	—	330	380	350	—

^aLTI pyrocarbon properties are adjustable within fairly wide ranges through proper selection of deposition conditions, and the properties above are to be regarded only as typical mid-range values.^bEstimate.

AXI grade graphite produced by the POCO Graphite Company has a finer grain, a strength approximately three to four times higher, and a fatigue life approximately 50% higher than H-451 in the unirradiated condition. It also shrinks and then expands during irradiation.

Chemically vapor deposited (CVD) pyrocarbon (such as LTI pyrocarbon) has high strength and good stability under neutron irradiation. It does not shrink or expand during irradiation as much as H-451 and AXI and is not as highly anisotropic as the more familiar pyrolytic graphite.

Table 8-13 (a,b,c) shows values for the thermal and mechanical properties of unirradiated and irradiated graphites and isotropic pyrocarbon. H-451 at 800 K has a higher thermal expansion after irradiation while the thermal conductivity is lower. The Young's modulus, after irradiation, is more than twice the unirradiated value, and the tensile fatigue life after irradiation is also higher. The overall effect of these changes is that the thermal stress for a given power in irradiated H-451 is 12 times higher than in the unirradiated condition, but the strength increases only by a factor of 1.5.

The same trends in the results are observed for POCO AXI graphite after irradiation. The LTI pyrocarbon shows less change in the properties after irradiation to 10^{26} n/m².

When graphite is irradiated under an external stress, it undergoes plastic deformation in the direction of the stress in addition to the dimensional changes that would occur without stress. Observed irradiation creep strains are at least an order of magnitude larger than any strain found in graphite in the absence of irradiation.¹¹²

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Cost-Performance Studies of an Inertial-Fusion Power Plant

Introduction

In this analysis, we developed a methodology for determining an attractive set of operating parameters for an inertial-fusion plant when the performance of the individual subsystems pull the system in opposing directions. For example, both laser and heavy-ion beam drivers are more efficiently utilized at higher repetition rates, even though liquid-metal-wall chambers are more attractive for low-repetition-rate, high-yield operation. We also explore questions of degree (e.g., how much additional capital cost is it worth to obtain a marginal one percent of driver efficiency?). We also sought a common basis from which to compare different drivers (and eventually, different reactor concepts) having quite disparate efficiency and cost characteristics. Finally, we explored the conditions that underlie the truth of such commonly expressed (and often contradictory) statements as:

- A fusion-energy gain (pellet gain times driver efficiency) of ten is all that is required for economical fusion power.
- Operating at a higher repetition rate can always make up for lower yield caused by lower pellet gain.
- Higher driver efficiency is always preferred to lower efficiency.

In each case, different variables are held constant.

We chose an economic-tradeoff analysis as a consistent basis for comparison because it is the only sufficiently general way to evaluate the combined performance of the subsystems, while at the same time assessing the penalties associated with off-optimum operation. There are risks in using an economic analysis; however, because our estimates of the inertial-fusion subsystems are this

Fig. 8-104. Logic flow diagram for inertial-confinement-fusion economic studies.

early stage of development are uncertain, as are estimates of some of the technical performance parameters. Even with uncertainties in both estimates, we can explore the complicated interrelationships between the cost and performance of subsystems that make up a power plant, identify attractive operating regimes, test the sensitivity to our assumptions, and determine the factors that have the greatest leverage on reducing power-generation costs.

Our figure of merit for this analysis is the minimum cost of electrical power. The cost of electrical power is dominated by the carrying charges

on the capital investment in energy technologies such as fusion that are not fuel intensive. Therefore, the analysis consists primarily of determining the dominant dependencies of the total costs of a reactor, driver, and pellet factory on the operating parameters, and combining them in a simple, consistent, computer model of coupled equations. This model is illustrated in Fig. 8-104, which shows the logic-flow diagram used in this analysis. From looking at the reactor cost, we see that it depends on the pulse repetition frequency (prf), chamber size, and plant-power level. The power level is a function of the energy multiplication in the chamber, the prf, and the pellet yield. The yield is the product of driver energy and pellet gain, which in turn, is a

Table 8-14. Financial parameters.

Cost of equity financing	14%
Equity to total capital ratio	50%
Cost of debt financing	8%
Debt to total capital ratio	50%
Income tax rate	50%
Calculated discount rate	9%
Method of tax depreciation	SYD
Project life	30 yr
Calculated fixed charge rate	15%
General inflation rate	7%
Construction period	
Reactor	10 yr
Driver, pellet factory	5 yr
Escalation rate	
Reactor	9%
Driver, pellet factory	7%

function of the driver energy. The price that influences the reactor cost also determines the lithium flow rate and, thus, the lithium-circulation pumping power. This affects the net electric-power output, which, together with the cost of the major components, determines the cost of electricity, and so on through the calculation.

The level of tradeoff-analysis given here is a compromise: a more complicated analysis is not warranted because of the magnitude of uncertainties, and a more simplistic analysis is inadequate to capture the essential interrelationships. In the future, we will emphasize obtaining better data for the system performance and cost models to improve the quality of the conclusions.

Cost Basis

We concentrate primarily on the capital costs that dominate generation costs for an ICF power plant. Briefly, the total capital costs of the three major subsystems are calculated as follows. (The details of the cost methodology are described in Reference 113.) Estimated construction costs (ECC) are first determined in 1979 dollars. Adding 25% to the estimated construction cost to account for contingency and design allowances gives us the direct capital cost (DCC). Indirect construction costs totaling 35% of the direct capital cost are added. Interest and escalation during construction are included as they occur to give the total investment at the time of commercial operation. This total capital cost is then deflated and expressed in 1979 dollars.

The financial parameters used in this study are listed in Table 8-14. Note that the discount rate is 9% and the calculated fixed-charge rate is 15%. We

Fig. 8-105. Calculated pellet gain as a function of driver energy.

have used a longer construction period and a higher escalation rate for the reactor than for the driver and pellet factory.

Pellet Gain Relationships

We begin the analysis with the calculated pellet gain (ratio of fusion-energy output to beam-energy input) as a function of the driver energy. We investigate the sensitivity of the economics to a variation in pellet gain by comparing the results obtained using an optimistic-gain curve with those obtained using more conservative-gain curves. These curves are shown in Fig. 8-105.

Pellet gain is wavelength-dependent for lasers and ion-energy-dependent for accelerators. Therefore, gain relationships for other driver-pellet combinations may differ from the particular example used here. Conservative calculations indicate that pellets for short-wavelength lasers are expected to fall within the conservative-gain band indicated; pellets for ion energies currently envisioned are expected to fall within a slightly lower conservative-gain band. Depending on the physics uncertainties, pellets for either driver might approach the optimistic-gain curve. Also, as we gather experience, we would expect to advance our knowledge from the conservative bands to higher gain

curves. To bound the potential performance of the ICF system, we compare the economic results for the optimistic-gain curves with the lower boundaries of the respective conservative-gain bands.

Pulse Repetition Rate

Throughout our analysis, the reactor-fusion power is fixed at 2700 MW_f. Even though economies of scale might favor an even larger system, we have selected this power since it results in standard-size electric-power plant (~1 GW_e). Also, by holding the power constant, the cost of the balance of plant, which is a major portion of the reactor cost, remains constant. This eliminates uncertainties in scaling one of the major cost components of the ICF power plant.

The prf required to achieve 2700 MW_f of fusion power for the different pellet-gain curves is shown in Fig. 8-106. This is simply the fusion power divided by the pellet yield corresponding to the different gain curves. The required prf is shown for the

Fig. 8-106. Required pulse-repetition frequency for 2700-MW fusion power.

optimistic-gain curve and also for the lower boundaries of the conservative-gain bands. Very simply, lower-pellet yields result in higher required prf's.

Total Capital Cost of Major Subsystems

For a given pellet-gain relationship, the operating point of the power plant is completely specified by the driver energy. By specifying how the cost of the drivers, reactor, and pellet-factory scale with these coupled parameters, we can express the results of the economic analysis solely as a function of driver energy.

Figure 8-107 shows estimates of the total capital cost of the short-wavelength laser (SWL) and heavy-ion-beam (HIB) drivers as a function of the driver energy. Remember that the total capital cost includes both indirect construction costs and time-related costs. The total capital cost of the SWL is estimated to be \$352 million at 1 MJ (ECC = \$200 million), and the cost scales with driver energy to the 0.8 power. The total capital cost of a 1-MJ HIB is \$616 million (ECC = \$350 million) and its cost scales with energy to the 0.4 power.¹¹⁴ Note that beyond ~4 MJ, the HIB has a lower capital cost than the SWL. The cost of the drivers is taken to be independent of the prf for both lasers and heavy-ion beams.

While we use the solid lines of Fig. 8-108 in the analysis, we have investigated the sensitivity of the system economics to a $\pm 50\%$ uncertainty in the cost estimates. These uncertainties are indicated by the dashed lines.

Figure 8-108 shows the total capital cost of both the reactor and pellet factory as a function of the prf. The reactor-cost scaling is based on the HYLIFE design.¹¹⁵ The cost includes the cost of the HYLIFE chamber, lithium-circulation pumps, and balance of plant. The $\pm 20\%$ uncertainty band investigated in the sensitivity analysis is shown.

The size of the HYLIFE chamber is set by a radiation-damage constraint on the first structure wall and is therefore constant, since fusion power is fixed in this analysis.

The cost of the lithium-circulation pumps is taken to be proportional to the required flow rate through the HYLIFE chamber. As the prf increases, the interpulse-time decreases; lithium must be injected at a higher velocity to reestablish the jet array between fusion energy pulses. As a result, the

Fig. 8-107. Total capital cost of drivers.

lithium flow rate, and therefore the cost of lithium pumps, increases with increasing prf. In addition, the electric power consumed by the lithium circulation system increases with increasing flow rate. At 1 Hz, the total capital cost of the reactor is \$1830 million (ECC = \$900 million) with \$120 million for lithium pumps.¹¹⁶

The cost of the pellet factory and the scaling with the pellet-production rate and pellet yield are uncertain. Fortunately, a $\pm 50\%$ variation in pellet-factory cost only has a $\pm 3\%$ effect on cost of electricity. The cost relationship (Fig. 8-108) used in this study is based on a total capital cost of \$176 million (ECC = \$100 million) at 1 Hz. We assume that the

cost is independent of the pellet yield and that 70% of the cost scales with the square root of the production rate. The $\pm 50\%$ uncertainty band is shown.

Based on this cost relationship for the pellet factory, the cost per pellet is shown in Fig. 8-109 as a function of the production rate. It shows a cost of \$1.2 per pellet at 1 Hz and 30¢ per pellet at 10 Hz. This is only the cost per pellet required to pay for the capital investment in the pellet factory. Material feed costs are expected to be negligible; and operation and maintenance costs will have to be minimized.

Fig. 8-108. Total capital cost of pellet factory and reactor.

Fig. 8-109. Cost per pellet as a function of production rate.

Total Power-Plant Cost and Net Electric Power

Figures 8-110 and 8-111 show the total power-plant capital cost in billions of dollars and the net electric power in MW_e as a function of the driver energy in MJ. Figure 8-110 is for a 5%-efficient SWL driver; Fig. 8-111 is for a 25%-efficient HIB driver. Both are based on the same optimistic-gain curve.

These figures illustrate how the total capital cost curve passes through a minimum near 2 MJ. Increasing the driver energy from 1 MJ lowers the required prf and, therefore, decreases the cost of the reactor and pellet factory. The driver cost, however, is increasing with increasing energy, which causes

Fig. 8-110. Total capital cost and net electric power of laser-driven plant.

Fig. 8-111. Total capital cost and net electric power of HIB-driven plant.

the total cost curve to begin to increase. The minimum occurs where, as driver energy is increased, the marginal increased driver cost equals the marginal decreased cost of reactor plus pellet factory for the corresponding decrease in prf. The minimum total capital cost is lower for the SWL-driven power plant than for the HIB-driven plant because of the lower cost of a SWL at lower energy per pulse.

However, the net electric power—the power available for distribution to the electric grid—is equally important. It is equal to the total electric

power generated (gross power) minus the power required to operate the plant (recirculating power). The power required by the drivers, lithium-circulation pumps, and other plant auxiliaries (e.g. secondary sodium-coolant pumps) make up the recirculating power requirements. Auxiliary power is 76 MW_e—too large to be omitted.

The electric power required by the driver, P_D , is related to the fixed fusion power, P_F by $P_D = P_F/(\eta_D G)$, where η_D is the driver efficiency and G is the pellet gain. Increasing the driver energy increases the pellet gain and, therefore, reduces the recirculating power requirement of the driver. It also lowers the prf, which reduces the power consumption of the lithium-circulation pumps as previously discussed. The auxiliary power requirements are fixed, since they are dependent only on the fixed-gross thermal power of the power plant.

Figures 8-110 and 8-111 show in both cases that the net electric power rises rapidly between 1 and 2 MJ and then asymptotically levels off. The net electric power of the HIB-driven plant is higher than the net power of the SWL-driven plant because of the higher efficiency of the HIB driver.

Cost of Electricity

The cost of electricity in cents/kWh as a function of the driver energy in MJ is plotted in Fig. 8-112. A 70%-capacity factor has been used. There

Fig. 8-112. Cost of electricity for the different pellet gain relationships and drivers.

are two sets of curves; one for the optimistic-gain curve and the other for the lower boundaries of the conservative-gain bands.

For the optimistic-gain curve, the cost of electricity curves are essentially the same for the SWL- and HIB-driven plants. Both pass through shallow minima near 2.5 MJ. The benefit of higher net electric power for the more efficient HIB system is canceled by the added capital cost of the driver in this case.

For the conservative-gain curves, the cost of electricity is higher and the minima are shifted toward higher driver energies. In this range of driver energies, the HIB has a dual advantage of lower capital cost and higher net-electric-power generation. As a result, the increase in the cost of electricity is much less for the HIB-driven plant.

Multiple Reactor Power Plants

Up to this point, we have considered a power plant that consists of a single reactor with a dedicated driver and pellet factory. If, on the other hand, more than one reactor is serviced by a single driver and pellet factory, the cost of these subsystems is shared and the overall cost of electricity is reduced.

The cost of electricity as a function of the number of reactors serviced by a single driver and pellet factory is shown in Fig. 8-113. It is important to note that the abscissa is the number of full-size (~ 1 GW_e) reactors, each of which must have its own

balance of plant because of such factors as maximum turbine size. It should not be confused with the concept of a power plant made up of many smaller, individual fusion chambers associated with a single balance of plant.

Figure 8-113 is based on the optimistic-gain curve. Recall that the driver costs are independent of prf and the pellet factory cost increases according to the scaling shown in Fig. 8-108. As indicated, doubling the number of reactors reduces the generation cost by $\sim 17\%$ and tripling the number results in an $\sim 24\%$ reduction. In this case, there is little incentive to have more than three reactors serviced by a single driver and pellet factory.

One disadvantage to this approach is that as the number of reactors increases, the total amount of electric power dependent on the reliability of a single driver increases. In fact, the number of reactors will have to be limited to the maximum loss of load that the utility system can manage; a more reliable, and perhaps more expensive, driver may be required if several reactors are dependent on it. The following curves consider a dual-unit power plant (two reactors) as the base case.

Selection of an Operating Regime

We define the acceptable operating regime of the power plant as the range of driver energies and corresponding system parameters that result in a cost of electricity that is within 5% of the minimum on the cost curve.

Figures 8-114 to 8-117 give the cost of electricity curves for the different drivers and gain curves. Curve A in each figure is the base case analysis; that is, the analysis with the appropriate scaling relationship for the HYLIFE chamber, pellet factory, and drivers. Curve B shows the results when the reactor cost is fixed and the lithium-circulation pumping power is not considered. Curve B, therefore, shows the sensitivity of the results to the assumed HYLIFE-scaling relationships. Curve C fixes the cost of the pellet factory, as well as the reactor cost. This, then, gives some indication of the effect of the pellet factory scaling relationship on the resulting cost of electricity curves.

Curves B and C in these figures do not correspond to workable conceptual designs; they are included to indicate the sensitivity to the assumptions.

Fig. 8-113. Cost of electricity from multireactor power plant.

Fig. 8-114. Selection of operating point based on optimistic gain curve.

Fig. 8-115. Selection of operating point based on optimistic gain curve.

In comparing curve C to curve A, we note that failing to include the prf dependence of the reactor and pellet factory will only affect the cost of electricity by 10%, but will lead to errors of a factor of two in the determination of driver energy and required prf.

The limit lines labeled a, b, and c intersect curves A, B, and C, respectively, at the point where the cost of electricity is 5% higher than the minimum. These limit lines, therefore, bound the

Fig. 8-116. Selection of operating point based on conservative gain curves.

Fig. 8-117. Selection of operating point based on conservative gain curves.

operating regime for each curve. In general, one would choose an operating point near the left boundary, as this will generally result in a smaller amount of total capital to be raised. There may also be technology limits, such as the chamber reestablishment rate, which would prevent choosing the lower capital cost. The bottom scale on each graph gives the operating parameters of interest. Note that since these curves are based on a power plant consisting of two reactors with a single driver, the driver prf is double the chamber prf. Fusion-energy gain is the product of pellet gain and driver efficiency. In the past, it has been used as a figure of merit for determining economic feasibility ($\eta Q \geq 10$) and for comparing the attractiveness of different driver systems. As we will see, in a coupled economic analysis such as this, the fusion energy gain is not a relevant parameter in itself.

Comparing curve A in Figs. 8-114 and 8-115 shows that in both cases the operating regime corresponds to driver energies from 2 to 4 MJ, driver-pulse rates less than 4.8 Hz, and chamber prf's less than 2.4 Hz. The cost of electricity is approximately the same in both cases even though the fusion-energy gain is a factor of five higher (142 compared with 28) with the HIB driver. These figures also illustrate that if, for example, the condensation rate prevents the HYLIFE chamber from operating at 2.4 Hz, there is little penalty in the cost of electricity in going to higher driver energies and lower chamber prf's.

Figures 8-116 and 8-117 show the electricity-cost curves based on the conservative-pellet-gain relationships. The operating regime is now shifted to high driver energies: > 7 MJ for the SWL and > 8 MJ for the HIB driver. As in the optimistic-gain figures, these conservative-gain figures illustrate the strong dependence of the operating regime on the reactor scaling relationship. Again, the minimum cost of electricity is relatively insensitive to the scaling. For the SWL (Fig. 8-116), the operating regime corresponds to $\eta Q > 6.6$, whereas for the HIB (Fig. 8-117), a fusion-energy gain as high as 20 is not within the operating regime. Whether either of these systems is economically feasible will depend on the cost of electricity from competing energy sources.

Sensitivity Analysis

To help identify those factors with a high leverage for decreasing (or increasing) the cost of

electricity, we examined the sensitivity of the power generation cost to the primary system parameters and major cost components.

Figure 8-118 shows the sensitivity to the driver efficiency for the optimistic-gain curve. The HIB-driven plant operates in a regime where the electricity cost is essentially independent of the driver efficiency over a wide range of values. This reflects the very low recirculating-power requirements of this driver-pellet combination. The SWL-driven system, on the other hand, shows a strong dependence on the laser efficiency. The 5% efficient laser used in our analysis is at the knee of the curve. Doubling the efficiency to 10% reduces the cost of electricity by less than 8%; but decreasing the efficiency to 2.5% increases the cost of electricity by $\sim 17\%$.

The bar chart in Fig. 8-119 shows the sensitivity of the cost of electricity to the gain curves. The cost is given for the two types of drivers, each with three different gain curves: the optimistic curve and the upper and lower boundaries of the respective conservative gain bands. The results are given for both single and dual-unit plants.

It is clear that the SWL-driven system is more sensitive to variations in pellet gain than the HIB-driven plant. For single-unit plants there is over a 100% variation with the SWL and a nearly 40% variation with the HIB. For dual-unit plants, the

Fig. 8-118. Cost of electricity as a function of driver efficiency. The curves do not join in the 10-15% range because the cost of the SWL is less than the cost of a HIB driver at the driver energies represented here.

Table 8-15. Electricity cost sensitivity (optimistic gain curve).

Parameter	Change in parameter (%)	Change in cost of electricity			
		SWL		HIB	
		¢/kWh	%	¢/kWh	%
Reactor cost	±20	±0.8	±13	±0.8	±12
Driver cost	±50	±0.84	±13	±0.96	±15
Pellet factory cost	±50	±0.23	±3.7	±0.22	±3.5

Table 8-16. Electricity cost sensitivity (conservative gain curves).

Parameter	Change in parameter (%)	Change in cost of electricity			
		SWL		HIB	
		¢/kWh	%	¢/kWh	%
Reactor cost	±20	±1.2	±9.2	±0.9	±16
Driver cost	±50	±3.2	±24	±1.8	±21
Pellet factory cost	±50	±0.39	±3.0	±0.31	±3.6

Fig. 8-119. Sensitivity to pellet gain.

cost of electricity varies by ~80% with a SWL; but with a HIB driver the variation is less than 30%.

Note that the cost advantages of sharing the driver between two reactors is more significant with the lower pellet gain curves. This follows since the operating regimes with the conservative-gain curves require very large, expensive drivers.

Tables 8-15 and 8-16 show the variation of the cost of electricity, in cents/kWh and as a percentage of the base value, for the indicated variation in the cost of the major subsystems of the power plant. Table 8-15 is for the optimistic-gain curve and Table 8-16 is for the lower boundaries of the respective-gain bands. For the optimistic gain, the reactor and driver costs have comparable leverage in determining the cost of electricity. The pellet factory is relatively unimportant. With the conservative-gain curves, the drivers become the high leverage items, because these systems require large driver energy.

Conclusions

Based on our analyses, we have formed the following conclusions:

- The higher efficiency of a heavy-ion-beam driver is especially important in the case of the lower, conservative gain, since in this regime, high driver energies are required to obtain reasonable yields. As one goes to higher driver energies, heavy-ion beams have the dual benefit of lower recirculated power and lower capital cost. However, for the optimistic gain, the lower capital cost of the laser at low-pulse energy (1 to 3 MJ) compensates for the higher required input energy, and leads to comparable electricity costs.

- The range of acceptable operating parameters is quite dependent on both the pellet-gain curves and cost scaling relationships; and we

have been unable to find a simple criterion for choosing operating parameters that omit cost considerations.

- A fusion-energy gain of greater than 10 is not a sufficient condition for determining economic feasibility. In this analysis, fusion-energy gains corresponding to the minimum cost of electricity varied from a low of 6.6 with a laser in the conservative-gain case to a high of 142 for a heavy-ion beam with an optimistic-gain curve. That is, because of cost considerations, a higher-efficiency driver does not necessarily allow operating at a lower gain or vice versa.

- The commonly perceived economic penalties of operation at high driver energies (5 to 8 MJ) and lower repetition rates (≤ 6 -Hz driver, ≤ 3 -Hz chamber) may not exist. The decreasing cost of the reactor and pellet factory with decreasing prf tends to balance the increasing cost of the driver. This is particularly true for dual-unit plants where the driver cost is shared.

- Electricity costs will be less sensitive to uncertainties in pellet gain with a higher efficiency driver.

- For high gains, the reactor is the major cost component and, therefore, holds the most leverage for reducing the cost of electricity. For the conservative-gain case, the driver cost becomes a comparable leverage item, especially for a single-unit plant.

- Two or three full-size reactors serviced by a single driver and pellet factory are cost effective, and are increasingly so for lower pellet gains because the driver cost is then proportionally larger.

In the future, we shall update this analysis as new and better information becomes available on the cost and performance of the major subsystems of an ICF power plant. More detailed studies on KrF lasers and pellet factories will be available soon. The dependence of driver cost on prf will be added. Finally, we intend to include an analysis of a reactor system with multiple smaller chambers making up a single, 1-GW_e power plant.

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Conceptual Design of a KrF-Fusion-Laser System

Our goal is to design a conceptual short-wavelength laser system for repetitively pulsed experiments in laser fusion to compress a KrF-laser pulse, which can be efficiently generated in the pulse length regime of several hundred nanoseconds, to the 20-ns time scale required to drive a fusion target. The two techniques that are combined to compress the pulse are 1) an angle-coded optical pulse-stacking scheme to compress the pulse a factor of four in time, and 2) a backward-wave Raman amplifier, which compresses the pulse another factor of five in time. The physics of these two techniques are discussed in Section 7 of this report.

Our objective was to identify and examine the key technical issues for short-wavelength fusion-laser technology and to indicate areas where significant innovation or development is needed to reduce cost and technical risk. The approach was to perform a conceptual design of a KrF-fusion-laser system suitable for an experimental test facility (ETF), the first repetitively pulsed-fusion driver. We chose design goals that were representative of such a near-term facility: an output energy of 1.5 MJ, a repetition rate of 2 Hz, a pulse length of 20 ns, an efficiency of 5%, and a lifetime of 10^7 shots. Choosing an ETF system allowed us to use near-term technology to form more concrete technology and cost assessments. Because we think that further innovation is desirable and probably required to make this system compelling, we chose a configuration that seemed to fulfill the design goals and eschewed further optimization.

This conceptual design is the collaborative effort of Bechtel National, Inc., which provided the

Fig. 8-120. Architecture of the hybrid KrF pulse-stacker/Raman compressor design for a nominal 1.5-MJ/2-Hz-fusion driver. Only one-half of the system is shown (not to scale).

system integration, mechanical design, building, layout, and costing; Physics International, which provided the power conditioning and costing; Hughes Aircraft Co., which provided the optical and alignment system and costing; and Lawrence Livermore Laboratory, which provided the KrF amplifier and Raman compressor performance parameters, the ETF design goals, and overall direction. Because of the brevity of this summary, we urge the reader to obtain the final report.¹¹⁷

The architecture of the hybrid-laser system is shown in Fig. 8-120. As can be seen, we have divided the laser into two subsystems: the pulse stacker and the Raman compressor. A KrF amplifier and preamplifier operating at $\lambda = 249 \text{ nm}$ prepares 100-ns pulses, which are fed into 14 beam lines, each 120 m long, extending radially outward from a central hub building. The KrF amplifiers and pulse-forming lines are located at the ends of the beam lines in a perimeter building. The laser pulses make three round trips in the beam tubes. In an input delay line, the pulses first reflect off of staggered mirrors, to produce a 100-ns train of four 100-ns pulses. These are then double-passed

through the KrF amplifier, which is in turn pumped for 400 ns by the e-beam. On the final pass, the amplified pulses are incoherently recombined into a side-by-side array of 100-ns pulses. The 14 arrays of 2-hy-2 pulses totaling 1.9 MJ are then simultaneously directed from the hub building through the Raman-compressor cell containing methane. In the meantime, a Stokes pulse has been prepared that is 20 ns in duration, at a wavelength of $\lambda = 268 \text{ nm}$. This Stokes pulse is counter-propagated through the Raman cell and amplified to the 1.5-MJ level. This final-pulse compression of a factor of five is accomplished in the Raman cell with 80%-conversion efficiency.

A more detailed view of the building layout is provided in Fig. 8-121. The perimeter building contains the KrF amplifiers, Marx banks, associated power supplies, gas-cooling and circulation equipment, and energy stores for the guide magnets required for the uniform deposition of e-beam energy in the KrF medium. The hub building contains the primary and tertiary mirrors for all 14 amplifier channels; it is the point where the laser pulses are reassembled and pointed at the Raman-compressor cells. The flow loops for circulating and cooling the methane gas are shown. Brewster-angle windows of fused silica are used in the Raman cell. The entire

Fig. 8-121. Schematic layout of the perimeter building, which houses the KrF amplifiers and Marx banks; the hub building with its large mirror arrays; and the flow loops, which circulate methane through the Raman cells.

Raman tunnel is 180 m long, to allow for synchronous double-passing of both the 100-ns KrF-pump beams and the 20-ns Stokes beam within the Raman cell.

The nominal performance specifications for the final KrF amplifiers is given in Table 8-17. These were used during the study, although subsequent calculations have been somewhat less optimistic. The output energy of 160 kJ per 2-by-2-by-3 m³ amplifier does not include mirror losses (5% each), window losses (2% surface), or fill factors (90%), which were separately accounted for. The final amplifier has a nominal-energy gain of 20. Each amplifier is equipped with rectangular magnetic-field coils, each 5 m by 3.3 m in size and separated by 3 m, which provides a 1-kG field in the active medium. This gives 20% field uniformity, and leads to 90%-deposition efficiency. It requires 0.65 MJ of stored energy per pulse, transferred resonantly from a 10-kV capacitor bank. The 15% energy loss estimated per shot leads to a power requirement of 2.8 MW for all 14 amplifiers. In addition, 5.6 MW of power are required for the gas circulation and cooling systems, which circulate the gas at 8 m/s.

In Table 8-18, we give the nominal performance parameters for the CH₄ Raman compressor, which must accommodate all 14-beam arrays. A length of 15 m of Raman-scattering medium is required to efficiently deplete the 100-ns-KrF pulse. Because of the high conversion efficiency (80%), there is little heat deposited and only a slow flow is required. The one-atmosphere cell pressure and the absence of large overpressures allow the use of a large expanse of fused-silica windowpanes, arranged in 2-by-7 arrays.

For the e-beams and power conditioning, we examined both pulse-forming networks (PFN) and pulse-forming lines (PFL) in the range 100 ns to 1 μ s pulse durations. For the 400-ns duration chosen, the PFL is preferable even though the energy must be stored twice (in the Marx generator and again in the liquid pulse-forming line) and there is a requirement for a high-voltage output switch. This is because the number of parallel elements required to keep the inductance sufficiently low increases with the inverse square of the pulse duration in a PFN. As the number of Marxes increases, the inductance of connections rises suf-

Table 8-17. Nominal performance specifications for each KrF final amplifier.

Aperture	2 m $\times$ 2 m
Length	3 m
Output fluence	4 J/cm ²
Intrinsic efficiency	10%
Pump time	470 ns
E-beam energy	1.3 MeV
Amplifier output energy	160 J
Stacker compression factor	4 $\times$
Stacker output pulse time	100 ns
Ar/Kr/F ₂ mixture	0.937/0.060/0.003 atm

Table 8-18. Nominal performance specifications for Raman compressor using methane.

Stokes pulse length	20 ns
Extraction efficiency	80%
Compression factor	5 $\times$
Pump pulse length (cell length)	100 ns (15 m)
Cell pressure	1 atm
Stokes output fluence	4 J/cm ²
KrF pump fluence	5 J/cm ²
Output energy	1.5 MJ
KrF energy	1.9 MJ
Output aperture (2 $\times$ 7 beam array)	3.3 m $\times$ 11.6 m

ficiently rapidly that no practical solution can be found at the 400-ns-pulse length.

In the KrF-laser system, there are 28 e-beams and PFL's, each called a "side", since each KrF amplifier is pumped from two sides. In each PFL, the energy is first stored in eight oil-immersed Marx generators, which then charge eight water dielectric pulse-forming lines on a 2- μ s time scale. Each output line bifurcates, driving two diodes. There are 14 e-beam apertures, measuring 3 m by 2 m, each consists of 16 modular diodes. Each side stores 1.3 MJ (for a total of 36-MJ stored energy per pulse) in 120- μ F, 50-kV capacitors. The energy losses are given in Fig. 8-122, which also shows the relative sizes of components. The overall-electrical-system efficiency is 57%.

The system is designed to operate for 10⁷ shots with only routine maintenance. The capacitors are derated a factor of two in voltage to attain this lifetime. The Marx switches will erode approximately 100 g per switch in 10⁷ shots at an erosion rate of 50 μ g/coulomb; changing the gas pressure

Fig. 8-122. The size, configuration, and component efficiencies are shown for a 400-ns pulse-forming line. Two of these are required to supply the electron beams of each of the 14 KrF amplifiers.

during the lifetime, can compensate for widening of the gap.

The output switch for each PFL is gas insulated and command-triggered, and operates through six breakdown channels each carrying 53 kA. This switch is under development but does not now exist in the required form.

A 900-ns PFL was also designed and costed for a 9-pulse stacker option; should the longer pulse length electrical system prove to be substantially less expensive. Unfortunately, we were unable to separately optimize the KrF amplifier design and architecture for each pulse length. However, the cost per joule stored was practically identical, leading to a selection of the 400-ns, 4-pulse stacker on the basis of a less costly optical system.

The configuration of the optical system is the key to the pulse-compression scheme. To take advantage of the long pulse-length KrF laser, we feed the final amplifier by an optical delay line that stacks a sequence of N short pulses of length τ through the amplifier at slightly different angles. By overfilling the amplifier with each pulse, the energy is extracted over the entire volume for a pulse length of $N\tau$, which allows efficient pumping of the amplifier by long-pulse electron beams. Angular multiplexing of the short pulses allows them to be separated spatially at the output of the amplifier where their relative delays are removed in a second

optical delay line. The pulses are reassembled into a spatial matrix rather than being superposed, since the latter would require very careful control on the interbeam coherence. The general architecture of this pulse stacker is shown in the perspective view of Fig. 8-123.

Further pulse compression, beam cleanup, and prepulse isolation are accomplished by using the KrF laser as a pump for a double-pass backward-wave Raman-scattering cell filled with methane. This cell converts the 248-nm, 100-ns, 1.9-MJ-KrF pump pulse of moderate wavefront quality to a 268-nm, 20-ns, 1.5-MJ Raman-shifted pulse with a smooth wavefront with negligible prepulse energy.

Effective prepulse isolation is accomplished because there can be very little energy directed toward the pellet in the absence of an input (at the Stokes frequency) propagating counter to the KrF-pump beam in the Raman cell. The Raman input to the cell requires only 1.5 kJ to control the 1.5-MJ output; this input in turn is generated in a front end that requires an input of only 1.6 J. This million-fold gain accounts for the effective prepulse isolation control of the Raman cell. The high quality of the output wave-front is essentially independent of the pump wave-front quality.

Many options were considered; among these were a 4-pulse versus a 9-pulse stacker, different types of mirror substrate, several alignment schemes, solid versus aerodynamic windows, and different beam-path gases. The baseline design chosen is a compact 4-pulse stacker that minimizes the complexity of the mirror mounts and alignment system, while fitting in a beam tube only 2.1 m by 6.2 m in cross section.

We selected an image alignment system that uses, as point sources of light, laser diodes mounted on each mirror array. Consider three sequential mirrors in the optical train (M_1 , M_2 , M_3). The light sources are mounted on the edges of mirror M_1 ; and a video camera is mounted on M_3 . The mirror M_2 is aligned by looking at the reflected image in M_2 of the point sources located on the mirror M_1 . The precision video camera or CCD on M_3 is used to compare the image with a reference perfect-alignment image; and an error signal is generated. A servo drives the mirror mount until the actual and

reference images agree. The entire optical train is aligned by sequentially continuing this process down the train. All four mirrors of each array are prealigned and fixed relative to each other, so the entire mount is aligned in one operation. The sensitivity and signal to noise ratios of this system are adequate with commercially available components.

Dielectric-coated, aluminum-substrate, water-cooled mirrors were chosen. The nominal fluence limits imposed were 5 J cm^{-2} at 248 nm and 100 ns, and 4 J cm^{-2} for the Stokes pulse at 268 ns and 20 ns. The final design has 5.9 J cm^{-2} on one of the KrF stacker mirrors. The 10^7 -shot lifetime required for multilayer dielectric reflectors at this fluence is orders of magnitude beyond the current state of the art. At 248 nm and 20 ns, the best commercial coatings have survived 10^2 shots at 2 J cm^{-2} and single shots at 10 J cm^{-2} . Clearly, this is an area requiring technology development.

The KrF amplifier windows are constructed of panes of fused silica with convex curvature to withstand the transient pressure differences. They are 10-cm thick—limited by the temperature profile across the thickness. We also investigated aerodynamic windows of argon gas, to eliminate the need for high-fluence solid windows. However, the gas used for a flowing curtain cannot contain fluorine, because of its large absorption coefficient. Our calculations showed that flowing curtains of pure argon in contact with the laser gas resulted in such high mixing rates that the costs of recovering fluorine would be unacceptable.

Perhaps the most important result of the study is the identification of areas requiring significant or early technological development. We briefly list some of the most important, deferring detailed discussion to the design report.¹¹

- Development of an output switch for a pulse-forming line capable of $\geq 10^7$ shot life at 2.7-MV, 0.3-TW, 2-Hz operation.
- Control of insulator breakdown and development of effective-modularized diodes.
- Experimental definition of minimum magnetic-guide-field requirements.
- Development of dielectric coatings in ultraviolet capable of withstanding 10^7 shots in the 4 to 6 J cm^{-2} range, for both mirrors and windows.
- Development of credible designs for fused-silica windows capable of surviving e-beam and pressure loadings, or development of aerodynamic

Fig. 8-123. Perspective view of the optical pulse-stacker system to be enclosed in beam tubes. The KrF amplifier is located just in front of the secondary mirrors.

windows with appropriate control over mass flow and composition.

- Determine need for helium in beam tubes versus adequacy of dry nitrogen or air.
- Definition of required optical homogeneity of KrF amplifier medium and beam conditioning gas, and minimum-alignment tolerances to adequately fill the Raman cell.
- Definition of shielding requirements for both x ray and noise.

- Properly determine the effects of employing long-pump-pulse technology by varying the amplifier sizing and stacker configuration to optimize around less-expensive PFN technology.

The cost of the development time and effort represented in the above abbreviated list is significant; but the technology is still new, and it is highly probable that important innovations lie ahead that can greatly reduce the technological risk, required development time, and eventual capital cost.

In conclusion, we observe that the overall efficiency of the laser system (3%) was somewhat less than anticipated. An accounting of the primary energy losses is given in Table 8-19. It is possible that the e-beam deposition efficiency could be raised (from 57% to perhaps 65%); but it is difficult to see where any great improvements could be made. The losses from windows, mirrors, and fill factors are already optimistic. The KrF-extraction pumping and efficiency at 400 ns probably cannot be bettered, and may be somewhat lower. The steady-state power losses caused by gas and water circulation and cooling, and magnetic-field establishment are 10% effects. Finally, although a pure stacker might save the 80%-efficiency factor allocated to Raman conversion, we do not know if the consequent reduced beam quality and prepulse isolation that a pure stacker implies would provide acceptable target performance.

The costs are summarized in Table 8-20. They correspond to \$2.67 per joule, which is consistent with previous estimates. It is interesting to note that the costs of the three major subsystems (optical, electrical, and building/mechanical) have total costs that are within 10% of one another. This implies that there is no single area that could have significant leverage in reducing costs. We must seek

Table 8-19. Laser system output and efficiency.

Laser electrical efficiency		
Source	Efficiency (%)	Power (MW)
Wall plug input	100	78.6
E-beam deposition	57	44.8
Intrinsic KrF medium	10	4.48
Windows and fill factors	90	4.03
Mirrors	82	3.28
Raman conversion	80	2.63
Total laser output	3.3	2.63 (1.3 MJ/shot)
Ancillary equipment losses (MW)		
E-beam wall plug		78.6
Gas flow blowers		5.6
Magnetic guide fields		2.8
Cooling water pumps		0.7
Total system input power		87.7
Overall system efficiency $\frac{2.63 \text{ MW}}{87.7 \text{ MW}} = 3.0\%$		

Table 8-20. Stacker-compressor laser system costs (400-ns pulsed power).

	Cost (millions)
E-beam generator systems	\$ 98.7
Optical systems	99.4
Buildings and ancillary equipment	87.2
Front end (14% of above)	40.7
Total field	326
Home office	34
Total	360
Contingency (25%)	90
Construction cost, fourth quarter	\$450

to reduce costs by finding improvements in all areas—or seek a major innovation in the conceptual architecture.

Finally, we mention an inconsistency in the design philosophy that should be remedied in future work. We intended the laser system for use in an ETL, for which a lifetime of 10^7 shots is adequate. However, there is a tradeoff between economy and efficiency that has received little attention. The cost per unit output energy is probably the best figure of merit for an ETL system; but we did not have the time or funds to explore some alternatives to reduce this cost (250 \$/joule output in a 20-ns pulse in the ultraviolet). For example, welded steel beamlines are probably not needed if nitrogen could be used

instead of helium, separate delay mirrors for the input and output stackers are probably redundant, and facilities savings may be possible by including separate Raman cells in each beamline. In a brief, good-brush study of this kind, we were unable to arrive at a point design of a recommended laser system. Instead, the greatest benefit of the study lies in the technology issues identified and alternative solutions proposed to attain a truly attractive short-wavelength laser driver.

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Advanced Isotope Separation

9

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Advanced Isotope Separation

9

Introduction

The goal of the advanced isotope separation (AIS) program at LLL is to demonstrate an economic and energy-efficient laser isotope separation (LIS) process for enriching uranium. The feed for LIS enrichment plants can be either natural uranium (0.7% ^{235}U assay) or the depleted tails from conventional enrichment facilities (0.25% ^{235}U). The product is uranium fuel for light water reactors (LWRs).

The physical principle behind LIS is the selective absorption of laser radiation by atoms or molecules containing the isotope of interest. This is possible because isotopic substitution produces shifts in the absorption spectrum (typically 5 to 10 GHz for atomic uranium) which are larger than the line widths (0.1 to 1.0 GHz) attainable with powerful, tunable laser sources. Consequently, it is simple to preferentially excite or ionize only the desired isotope.

The mainline LIS process developed at LLL operates on uranium in the form of its atomic vapor. The rich electronic spectrum in atomic uranium (10^4 to 10^5 transitions) makes its atomic vapor especially suitable as a working medium for LIS and allows us to consider operating lasers anywhere from the infrared to the ultraviolet.

The essential features of the atomic vapor laser isotope separation (AVLIS) process are depicted schematically in Fig. 9-1. Steps in the separation process are as follows:

1. Uranium vapor (produced by evaporating uranium metal) streams into the separation zone.

2. Visible lasers, tuned precisely to the ^{235}U transition, selectively photoionize the ^{235}U in the vapor.

3. Positively charged ^{235}U ions are attracted and collected by a product collector plate.

4. ^{238}U vapor, which is not ionized by the laser radiation, is unaffected by the electrical forces and streams onto a tails collector.

Spectroscopy experiments at LLL in early 1974 showed the scientific feasibility of selectively isolating desired isotopes of uranium through photoionization. Since 1974, we have concentrated on developing laser, vaporizer, and extractor systems that can be

scaled to large size. We have identified a set of baseline laser transitions and have accurately measured all parameters in the photoionization process. We have also successfully completed macroscopic enrichment experiments, demonstrating that the baseline photoionization process agrees with our theoretical expectations. Finally, an extensive development program, carried out both at LLL and at Union Carbide Corporation-Nuclear Division (Oak Ridge), has demonstrated that atomic uranium of the required densities can be vaporized and collected in a reliable and routine fashion.

Fig. 9-1

The integrated process development facilities that have been or shortly will be brought on line at LLL are shown in Fig. 9-2. The SPP-II system, which provides primary laser power, was first operated in 1976. The Regulus vaporizer/process chamber came on line in 1978. These two systems have been combined in our recent enrichment runs. The Venus laser power amplification system will be integrated into our process simulation experiments in 1980. The Mars system, a higher-throughput vaporizer and process chamber, is scheduled for completion in 1981.

On the basis of major advances during the last five years, we are convinced that AVLIS can meet its very low capital cost, as well as its very low projected cost per separative work unit (\$10-40/SWU); the AVLIS process derives this cost-effectiveness from its ability to obtain large separation factors (and hence high enrichment rates) within a single stage.

Fig. 9-2

The major reduction in enrichment cost that AVLIS represents will influence the entire nuclear fuel cycle. Introduction of AVLIS into the "once-through" LWR fuel cycle, represented in Fig. 9-3, will significantly increase uranium resource utilization, providing the following benefits:

- Substantially reduced capital investment in enrichment plants, electrical generating capacity, mining, and milling.
- A more competitive US position in world markets, improving both balance of payments and proliferation control.
- Expanded energy supplies from an extended LWR economy.

The AVLIS process, in fact, may offer one of the few options in the entire energy field for actually reducing an existing fuel cost.

The current level of AVLIS technology points to commercial deployment in the early 1990s. The interim development program for meeting this date must be an aggressive one, as delays that defer commercialization until the mid-1990s or later will erode the benefits of introducing AVLIS. With this in view, the AVLIS program at LLL is structured into four progressive stages:

- Technology Evaluation (1978-80). In this phase, we have used research equipment to obtain the basic physics data needed for adequately defining the LIS process. We are investigating process parameter scaling to decide on the best approaches for developing system components.
- Technology Demonstration (1980-85). During this stage, we will scale component hardware to near-plant size, extending component operating lifetimes into the hundreds and thousands of hours. We will address issues of engineering scale, collect system performance data, optimize process performance, and seriously begin to resolve the problems associated with system lifetime.
- Engineering Demonstration (1984-88). This phase calls for us to increase the component and subsystem lifetimes, reduce hardware costs, improve component reliability and

Fig. 9-3

maintainability, and optimize the system performance under plant operating conditions.

- **Economic Demonstration (1987-92).** This final phase marks the actual use of AVLIS technology in producing cost-effective separative work.

We are now nearing the end of the technology evaluation phase. We have measured the relevant scientific parameters for a completely integrated process and demonstrated uranium enrichment in a number of process experiments. We have begun increasing component sizes and addressing the full-scale system integration issues on which a full technology demonstration depends. Nevertheless, we still have considerable scaling to do for the planned technology demonstration.

Within the AVLIS program, we are developing alternate subsystems to back up our mainline technology. A prominent example of these process options is the development of both copper vapor and rare gas halogen lasers as primary drivers. To the extent that program schedules and funding permit, we will continue to explore parallel approaches in the areas of lasers, vaporizers, extractors, and collectors.

While these options give the program flexibility and underwrite the performance and cost goals of future enrichment systems, it should be emphasized that the system components for the mainline AVLIS process already exist.

In this section we present only a few selected highlights of the AVLIS program; this review emphasizes supporting research at the expense of component development studies. More detailed descriptions of all aspects of uranium AVLIS at LLL are given in classified publications.

“Laser Development” describes part of our work on copper vapor and rare gas halogen lasers. For these well-established laser systems, our experiments are directed toward increasing laser power output and identifying the phenomena that limit laser lifetime.

In “Atomic Vapor Spectroscopy” we describe two experiments that apply new techniques to uranium spectroscopy; these experiments are part of LLL’s extensive and continuing uranium atom spectroscopy effort.

“Advanced Laser Research” presents examples of LLL research ranging from entirely theoretical studies of the kinetics of laser discharges to experimental work that seeks to extend the spectral range covered by tunable laser sources.

The fundamental physical event in an AVLIS process is the interaction of a free atom with laser radiation. This basic process has been much studied but is far from completely understood. The subsection entitled “Photon-Atom Dynamics” describes theoretical studies of the deflection of atoms and the pumping of multilevel atoms by lasers.

In the next section, “Collision Physics and Chemistry,” we present two chemical kinetics studies: a continuation of our survey of the single-collision chemistry of ground-

state uranium atoms, and a calculation of the kinetics of a representative atom-molecule reaction.

Over the years, we have surveyed the field of laser photochemistry with the aim of identifying alternate applications for the laser technology developed in support of the AVLIS program. An example of this work is presented in the concluding article, "Alternate Laser Applications: H₂S Cleanup in Fuel Feed Streams by Laser Photolysis."

Authors: J. I. Davis and R. C. Stern

Laser Development

Copper Vapor Laser Overview

The AVLIS process uses pulsed lasers to photoionize free uranium atoms. Because the absorption line widths in uranium vapor are narrow and the isotope shifts are small, continuously tunable laser sources are required. Early in the AVLIS program, pulsed dye lasers were identified as having both the requisite tunability and the potential for high output power at high pulse repetition frequencies (prf's). Copper vapor lasers (CVLs) were identified about the same time as good candidates to pump the dye lasers, offering several immediate advantages:

- Good overlap with the absorption spectra of suitable laser dyes.
- High prf capability.
- Potential for scaling to high power.

The past year has been extremely important for copper laser development. Not only did we activate a new, large CVL facility but also realized major advances in the performance of copper vapor lasers.

Venus Facility. Additional laser power is needed if we are to continue our progress in understanding the AVLIS process and to increase our capability for uranium enrichment. The Venus power-amplification facility (Fig. 9-4), now being built for full operation in late 1980, is expected to deliver more than 400 W of copper laser power for isotope separation experiments. The Venus system consists of three dye power amplifiers energized by an array of 32 copper vapor lasers. Low-power dye laser beams transported from an adjacent laboratory are injected into the Venus dye amplifiers, increased in average power, and then combined onto a common axis before being propagated to the isotope separation chamber. The copper laser and dye systems were described in last year's annual report.¹

Following installation of the copper laser modules in the Venus facility early in 1979, work was directed toward our September goal of producing more than 250 W of copper pump laser power from the combined Venus/SPP-II facilities. Successful demonstration was considered vital in that its attainment would require simultaneous high performance of a large number of the copper lasers on the Venus spaceframe. We surpassed the goal, producing more than 300 W of copper laser power from the combined facilities. Of this, Venus provided 260 W with 21 laser heads operating. The Venus lasers were configured into four master-oscillator-power-amplifier (MOPA) chains comprising as many as six lasers. Their successful operation at this power level confirmed many of our system philosophies: e.g., that high levels of pump laser power can be achieved with a large number of laser units and that the multiple-laser approach is feasible. The success reinforced our confidence in the design approach that allows scaling of the copper laser output power per aperture by means of a MOPA chain. We were well satisfied with the human engineering and operability of the system. Control was based on the use of a master console. We demonstrated initial use of our diagnostics and computer control subsystem. The results suggest that our design goal of more than 400 W of average power will be met during 1980 with the Venus copper laser system.

Performance. Attempts to improve the average power output of copper vapor lasers have been limited in the past by the requirement—for all self-terminating lasers—that the metastable lower level of the laser transition must be deactivated. Until recently it was believed this could be done only by collisional deactivation after diffusion of the copper metastables to the walls; consequently, at high prf's, the bore diameters of cylindrical discharge lasers were limited to 25 to 30 mm. To circumvent this restriction we turned to more complex annular configurations that allow an increased active volume.

Fig. 9-4. Venus laboratory.

An annular copper vapor laser designed and built at LLL produced record power outputs in 1979.

Recent experiments at LLL and elsewhere (Israel and the Soviet Union) have suggested that using neon buffer gas at high operating voltages creates conditions in which volumetric deactivation takes place. This result is extremely significant because it removes the requirement for wall proximity, thus allowing the construction of simple, large-bore cylindrical CVLs. The performance of one of these devices, described below, has confirmed that large-bore CVLs can be operated at high average powers with pulse repetition frequencies greater than 4 kHz.

Our ability to obtain high stored powers using large-bore cylindrical lasers allows us ultimately to build MOPA chains using simple spherical optics and to incorporate advances in copper laser reliability technology—achieved with smaller 15-W lasers—directly into large-bore cylindrical lasers. Significant advances in copper laser lifetime were demonstrated during 1979. We have also been able to demonstrate excellent long-term performance of copper lasers using inexpensive and easily obtainable materials. Finally, our experimental and theoretical work on the electrical engineering of high-power copper lasers has produced several circuit designs that not only allow us to operate at the

higher voltages required for large-bore cylindrical lasers but also point toward circuits of like capability that can be built at significantly less cost.

These successes have increased our confidence that copper vapor laser technology can be advanced from its present state to a level appropriate for incorporation in an AVLIS plant.

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Advanced Copper Vapor Laser Experiments

In the past year we have conducted several experiments which influenced our understanding of the copper vapor laser and its scaling potential. Past efforts at LLL in developing high-power CVLs have been concentrated on an annular discharge design, in a buffer of low-pressure helium. The use of this geometry was motivated by the belief that the metastable lower level in CVLs is deactivated between pulses by diffusion to the walls; an annular configuration that minimizes the diffusion distance

Fig. 9-5. Schematic drawing of large-bore copper vapor laser (CVL).

without restricting the bore volume would thus allow higher power output. To verify this result, an annular CVL constructed at LLL has been successfully operated with output powers in the range from 25 to 35 W for a 73-mm-diameter discharge.²

In the past two years, however, publications by Bokhan³ and Isaev⁴ in the Soviet Union and by Smilanski⁵ in Israel have reported enhanced power output from small-diameter CVLs using high-pressure neon discharges. Experiments on a 22-mm-i.d. CVL conducted this past June at LLL corroborate the foreign results. Generally, laser output peaks in the range from 20 to 50 Torr and does not decrease significantly up to 100 Torr, even at prf's as high as 12 kHz. (We found that operation of the CVL with a neon buffer gas is successful only when a large electric field is applied across the discharge tube. Typical applied fields are of the order of 200 V/cm—approximately twice that used in the helium-buffered lasers investigated in the past.)

The high-pressure neon results contradict our earlier understanding that the metastable lower level in CVLs is deactivated by diffusion to the walls, since fundamental mode diffusion times in neon above 50 Torr are more than five times longer than the interpulse period. In fact, the neon data strongly suggest that deactivation of the metastable level in high-pressure neon takes place by means of volumetric deactivation, i.e., through depopulation by one of the plasma species.

This promise of a volumetric deactivation process in high-pressure neon prompted us to conduct experiments on a CVL of larger diameter, whose active volume was more than seven times that of other neon-copper lasers previously investigated at LLL.

Figure 9-5 is a schematic drawing of the laser tube. The alumina discharge tube is 73 mm in i.d. and 1.22 m in length, although it was estimated during our investigation that only the center 560 to 640 mm contributes to gain length because of a limited hot zone. The discharge tube is wrapped with a metallic heat shield, which ensures that the waste heat produced by the plasma raises the alumina tube temperature to its operating temperature of 1725 K. This large-bore CVL was operated with neon at 10 to 60 Torr. The laser electronics include thyatron-switched command charging of the 16-nf main-discharge capacitance. The energy stored on the main capacitance is discharged through the laser by triggering a single HY-53 thyatron.⁶ Typical charging voltages for neon operation were 12 to 18 kV.

Oscillator Performance. We first evaluated the performance of this large-bore CVL head as an oscillator; preliminary results were encouraging. Flat-flat optics were employed using a 99% reflector at 510.6 and 578.2 nm and a 4% reflecting quartz output coupler. We found that the oscillator optimized at a prf of 4 kHz, with neon buffer pressure in the range of 25 to 35 Torr (though there was a tendency for the optimum neon pressure to shift to higher values at higher applied voltages across the discharge tube).

A representative beam profile, taken at a 4-kHz prf with neon at 25 Torr, is displayed in Fig. 9-6. Under these optimum conditions the output beam was uniform across its entire 73-mm diameter. A uniform laser beam at 4 kHz and 25 Torr neon in a 73-mm-i.d. discharge strongly suggests that there is a volumetric deactivation of the metastable level; this in turn increases the likelihood that the laser can be scaled to even larger diameters.

Fig. 9-6. Radial intensity profile of large-bore CVL oscillator beam, with neon at 25 Torr and prf of 4 kHz. Beam is uniform across its 73-mm inner diameter.

The oscillator produced up to 55 W average power from the green and yellow lines. Laser efficiency remained constant at 0.58% up to the maximum input power of 9.6 kW. These results, achieved at a 4-kHz prf with 32-Torr neon, are summarized in Fig. 9-7, with oscillator output plotted against input power. The 55-W output power is nearly twice that achieved previously in a low-pressure annular CVL of approximately the same outside diameter.

Amplifier Performance. Since there is high interest in operating CVLs in sequence as both master oscillators and power amplifiers, we have also operated a large-bore CVL head as an amplifier, and measured its stored power.⁷ The optical arrangement for this stored-power measurement employed a 25-mm 15-W CVL with unstable resonator optics as the oscillator, whose 1-inch beam was then expanded to fill the inner diameter of the large-bore laser head. Output power from this oscillator-amplifier configuration was measured as a function of the 25-mm-oscillator output; the results, at a 4-kHz prf, are plotted in Fig. 9-8.

The data we obtained fit the steady-state saturation formula

$$\ln G = g_0 \ell - \frac{\Delta P}{P_{\text{sat}}} \quad (1)$$

Fig. 9-7. CVL oscillator output power vs electrical input power. Efficiency remains constant.

for a homogeneously broadened line, where g_0 is the small signal gain; P_{sat} is the saturation power, ℓ is the active gain length of approximately 600 mm, G is the large signal gain equal to $P_{\text{out}}/P_{\text{in}}$, and the extracted power ΔP is equal to $P_{\text{out}} - P_{\text{in}}$. For the data in Fig. 9-8, the formula yields a saturation fluence of approximately $70 \mu\text{J}/\text{cm}^2$ and a small signal gain of about 1%/mm; these values are in general agreement with previous measurements in low-pressure helium CVLs at LLL.⁷

Using the figure data in Eq. (1) allows us to estimate the maximum stored power that one can extract from an amplifier under arbitrarily large input powers. Figure 9-8 shows that the stored power in the large-bore laser head used as an amplifier, equal to the extracted power at $G = \text{large signal gain} = \text{unity}$, was 67 W; this same head produced 37 W average power as an oscillator. These data imply that there is 80% more power available from this CVL when used as an amplifier than when used as an oscillator.

We have used the results of this stored-power measurement to estimate the stored power, efficiency, and energy density of the large-bore CVL that achieved 55 W as an oscillator:

$$P_{\text{stored}} = 100 \text{ W} ,$$

$$n = \frac{P_{\text{stored}}}{P_{\text{supply}}} = 1.1\% ,$$

$$e_d = 10.5 \mu\text{J}/\text{cm}^3 ;$$

the energy density is calculated assuming a hot-zone length of 600 mm.

Metastable Deactivation. The initial success of the large-bore high-pressure-neon CVL has stimulated interest in understanding the kinetics of the deactivation of the metastable lower laser level. Two conjectured reactions are discussed below.

One possible reaction involves deactivation of copper metastables by neon directly:

In this reaction Cu^* represents the copper ^2D metastables, Ne represents neon atoms, and ΔE is the released kinetic energy of approximately 1.5 eV. The reaction rate per copper metastable is

$$R = k[\text{Ne}] . \quad (2)$$

Rate constants for this process with other rare gases have been measured.⁸ All reaction rates are less than $5 \times 10^{-16} \text{ cm}^3/\text{s}$. With neon at 50 Torr, however, this rate constant results in a characteristic quenching time of 7 ms, which is much too slow to deactivate the copper metastable level between discharge pulses, since a typical interpulse period is less than 250 μs .

The metastable lower level in CVLs might also be deactivated by superelastic collisions with electrons during the interpulse period. The reaction for this process is written as

and has a characteristic rate equal to

$$R_{\text{se}} = k_{\text{se}}[e] . \quad (3)$$

The rate constant for this collision process with copper metastables has not been measured, but typical rate constants for other metastables (such as helium) are in the neighborhood of $10^{-9} \text{ cm}^3/\text{s}$. Applying this value with an electron density on the order of $10^{13}/\text{cm}^3$ results in a collision rate of ap-

Fig. 9-8. Results of stored-power measurement using large-bore CVL as amplifier. Note that whereas the average output of this CVL as an oscillator was 37 W, its output as an amplifier is measured here as 67 W.

proximately 10^4 s^{-1} ; this value is comparable to the pulse repetition rates we see in high-pressure neon.

For superelastic collisions to be the dominant deactivation process during the interpulse period, the electron temperature must drop to near-thermal levels ($\sim 2000 \text{ K}$). Then, for the appropriate copper ground state density of $\sim 7 \times 10^{14} \text{ cm}^{-3}$, a Saha equilibrium (between the 2000-K electrons, the copper ground state, and the metastable states) would yield metastable densities of 10^{11} to 10^{12} cm^{-3} . These densities are low enough to permit population inversion during the next discharge pulse.

While superelastic collisions between electrons and copper metastables produce the necessary deactivation rate, the process does not have helium or neon as one of the reactants and hence cannot help to explain the differences between helium and neon laser performance. These differences must therefore be attributed to significantly different afterglow characteristics, such as electron density and temperature, which are themselves strongly affected by differences in electronic structure, recombination rates, and elastic collision cross sections. Further work, to improve our understanding of afterglow characteristics and thus to optimize laser performance, is in progress.

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Rare Gas Halogen Laser Overview

Discharge rare gas halogen lasers are efficient and scalable drivers that can be used both as intrinsically narrow-line tunable sources and as pump sources for dye lasers. The range of wavelengths available from RGH lasers makes them applicable for one-, two-, and three-step AVLIS processes.

The major technology issues related to the development of RGH lasers for AVLIS can be roughly divided into three areas: materials compatibility and closed-cycle chemistry, power conditioning, and flow-related phenomena. Our vehicle for studying these issues has been the closed-cycle test bed (CCTB),⁹ a UV-preionized RGH laser comprising a halogen-compatible closed-loop flow system and thyatron-switched power conditioning modulator. This system was used during the past year to investigate to first order the closed-cycle characteristics of four RGH lasers, with emphasis on the detailed behavior of XeCl. To date, XeCl has exhibited the best average power and lifetime behavior: the XeCl laser seems especially suited for the eventual AVLIS applications. Work on the CCTB on flow-related laser effects and power conditioning development will continue in the near future.

In addition to studies of closed-cycle laser behavior, we have investigated advanced techniques for exciting and preionizing RGH lasers which would result in better laser performance and relax

the requirements on the thyatron switches in high-power, conventional-discharge laser designs. The advanced technique under investigation uses a low-current-density, short-pulse electron beam as both the preionizer and the main switch in the laser. The scientific feasibility of this electron-beam-switched discharge (EBS) technique had been demonstrated¹⁰ in single-pulse laser experiments. Considerable effort was devoted this year to developing a wire-ion-plasma (WIP) electron gun for EBS excitation, whose primary characteristics and advantage over other gun designs are relative simplicity, ruggedness, and ultimate reliability. The WIP gun is capable of operating at the peak current density and repetition rate required for high-average-power RGH laser devices. The emphasis in the coming year will be to explore characteristics of the EBS laser concept under conditions of flow and continuous pulse rates.

Author: T. Kan

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XeCl Laser Development/Closed-Cycle Test Bed Experiments

Closed-cycle-flow rare gas halide lasers have produced unprecedented average laser power in the ultraviolet (155 W at 249 nm).¹¹ Volume and prf scaling will lead to devices of considerably higher power. Issues facing the development of high-average-power RGH lasers can be divided into those inherent in high-average-power operation and those related to operating lifetime and reliability. Among the former are pulse power switching, optics damage, and the difficulties associated with high-prf operation, particularly flow clearing and acoustic damping. The latter include gas contamination, materials compatibility, optics degradation, and the lifetime of the high-power electronic components. Our major tool for average-power RGH laser development at present is the closed-cycle test bed (CCTB) system, with which we are seeking to understand the mechanisms that limit laser power and

Fig. 9-9. Closed-cycle test bed laser flow loop. Overall length of loop is 3 m; attached at cross is turbomolecular vacuum pump.

operating life. This work is directed toward developing practical designs and operating techniques for high-average-power devices.

CCTB Laser Performance. Figure 9-9 shows the CCTB flow loop, which is described in detail elsewhere.^{12,13} The laser gas at about 2-atm pressure is circulated around the loop by a 300 liter/s tube-axial fan. The gas flow velocity in the active volume is ~ 20 m/s. In the laser head are 60-cm-long transverse discharge electrodes separated by 2 cm and connected along their entire length to rows of mica capacitors which, together with two HY-53 ceramic hydrogen thyratrons, comprise the main discharge pulser. The storage capacitors are command-charged to about 38 kV, storing approximately 15 J. The resulting discharge current pulse is ~ 25 ns long with a peak of ~ 20 kA. Ultraviolet preionization is provided by an arc array downstream of the electrodes.

Early in 1979 the maximum laser energy obtained with the CCTB was 80 mJ with KrF; the

Table 9-1. CCTB peak laser performance.

System	Mixture (partial pressures) (Torr)	Energy (mJ)	Average power	
			W	Hz
XeCl	HCl, Xe, He (3.5/70/1350)	100	54	600
	HCl, Xe, Ne, He (3.5/70/700/650)	125	55	600
KrF	F ₂ , Kr, He (3/150/1350)	145	28	300
XeF	F ₂ , Xe, He (3.8/15/1435)	45	16	500

peak average laser power was 23 W at 600 Hz with XeCl. Improvements to the discharge pulser, discharge electrodes, and laser optics then produced significant improvements in the peak laser performance. Listed in Table 9-1 are the present maximum low-prf laser energies and peak average laser powers for three RGH systems. The highest average power—55 W at 600 Hz—was obtained with XeCl using a helium/neon gas mixture at about 1:1.

Fig. 9-10. Laser energy prf-dependences for KrF and XeCl with helium buffer gas mixtures. Dashed lines are constant average-power contours. As explained in text, these curves have structures above about 200 Hz that are not resolved here.

Figure 9-10 shows the prf dependence of the laser pulse energy for KrF and XeCl; these data were derived from steady-state power measurements. Above about 600 Hz, persistent arcing occurs downstream of the electrodes because of insufficient flow clearing; the XeCl energy drops rapidly. At prf's above about 200 Hz, these curves have detailed structures not shown in Fig. 9-10: the maximum departure from the curves is a laser energy reduction of 20%. Diagnostic experiments indicated that the structures could not be attributed to mechanical vibration, suggesting acoustic resonances as the likely cause. To investigate, we mounted a fast pressure transducer near the discharge anode, discovering strongly resonant acoustic oscillations. For example, at prf's near 500 Hz, these oscillations persist from shot to shot with amplitudes such that the resulting density inhomogeneities are estimated to be $\Delta\rho/\rho \approx 5 \times 10^{-3}$, with scale lengths of a few centimeters. The mechanism for the corresponding decrease in laser energy, rather than degraded optical homogeneity, is believed to be degraded discharge stability from the density gradients associated with acoustic os-

cillations. Supporting evidence includes our observation that the decrease in laser energy results mainly from a shortening of the laser pulse length rather than from a reduction in the peak laser power. Further support comes from optical homogeneity measurements and visual observation of the discharge. Our findings make it increasingly clear that acoustic damping is essential to successful operation of these lasers at higher prf's.

Operating Lifetime Studies. Much of the the concern about the practicality of high-average-power rare gas halide lasers has historically centered on lifetime issues, particularly on limitations of gas lifetimes from contamination and halogen consumption. Our first approach to this problem was to build the system as cleanly as possible, using common vacuum materials (aluminum alloys and stainless steels) and O-ring seals. During initial system operation we surveyed the lifetimes of single gas fills for operation at 250 Hz, using the four most promising RGH systems. The shortest half-power lifetimes were observed with ArF and XeF (15 and

Fig. 9-11. Mass spectra showing contaminant buildup during XeF laser run. Vertical axis is linear in density; scale depends somewhat on species. CF_3 and SiF_3 are presumably produced from CF_4 and SiF_4 in ionizer of mass spectrometer.

30 min, respectively); the KrF lifetime, 2 h, was considerably better. The longest lifetime was with XeCl, in which the power decreased 20% during a 2.5-h run. Much of this dropoff may have resulted from coatings deposited on the laser windows during the run.

We then began examining specific contamination problems in detail, installing an on-line mass spectrometer sampling system in the loop as our gas composition diagnostic. The use of this diagnostic has been mainly qualitative, for monitoring changes in the gas composition during long runs. The mass spectra data in Fig. 9-11 show one of the most dramatic effects we've seen: a buildup of the contaminants CF_4 and SiF_4 during an XeF run, evidenced by their cracking products CF_3^+ and SiF_3^+ . The carbon probably comes from low-level hydrocarbon and atmospheric contaminants; a possible source of the silicon is a high-silicon aluminum casting alloy in the fan. Changes in the gas composition during XeCl runs are much less pronounced but show a slow buildup of CCl_4 .

From the lifetime results and mass spectrometer data, it appears that although there are particular problems with some of the fluorine

systems, our design has been relatively successful in maintaining gas purity. It is also apparent that gas contamination is not an operating lifetime limitation in this device with XeCl.

To further investigate the practical lifetime limits in high-average-power RGH lasers, we devoted considerable effort to achieving relatively long XeCl laser run times on the CCTB. The most troublesome problems were found to be electrode erosion and window coating. The surfaces of the discharge electrodes won't be able to maintain halogen passivation layers, thus will be continuously eroded by discharge sputtering and halogen corrosion. The preferred electrode material would combine high melting temperature for sputtering resistance and relatively nonvolatile halogen corrosion products for corrosion resistance. Unfortunately the refractory metals—otherwise an obvious choice—generally yield very volatile halides and thus resist chemical attacks quite poorly. Of the materials tested to date with the CCTB, the best results by far have been with nickel. Nickel-plated aluminum electrodes and solid nickel (201) electrodes with water cooling channels have performed well; each has withstood some 10^7 shots with erosion limited to a slight flattening of the ends.

The next limit of concern was deposition of a coating on the fused silica windows. This coating is laid down in a beam pattern, presumably involving a photochemical reaction, and has a very reflective, metallic appearance. We found the deposition rate and composition to depend on the discharge electrode material, the slowest rate being for nickel electrodes. X-ray fluorescence analysis of the coating deposited during runs with nickel electrodes showed it to be mostly chlorine, probably in the form of chlorinated hydrocarbons. Isolating the window surfaces from the flow-loop gas (from HCl) was an obvious solution. An arrangement of baffles in the window mounts, together with a flow of clean gas across the window, has dramatically reduced the deposition. Flowing 3.5 liters/min of He-5%Xe past each window, we operated the laser for 4 h at a prf of 525 Hz, producing about 40 W for the entire run. After the run the windows were found to have about 3% absorption at 308 nm because of a coating; no apparent absorption was found after this coating was removed. The total fluence through the windows during the run was about 0.5 MJ/cm².

To reduce gas consumption, we have been evaluating techniques to purify the gas vented from the loop and return it past the windows, maintaining the loop's purity. Preliminary success has been achieved using the scheme shown in Fig. 9-12. Flow-loop gas is bled from the center of the fan shaft seal and flowed through a calcium trap¹⁴ at 500 to 600°C, followed by a cooldown and a cold

trap. The calcium quantitatively removes the HCl, atmospheric contaminants, and hydrocarbons. The resulting clean gas is used to purge the window mounts and fan motor housing. Makeup HCl is added to the loop in proportion to the bleed flow. This configuration allows the system to operate in chemical steady state with a gas change in the loop every 45 min, limiting the consumption to 1.7 g/h of HCl. With this scrubber system, laser power in excess of 35 W at ~500 Hz has been maintained for 2 h.

As we learn more about operating characteristics and lifetime limits of closed-cycle-flow rare gas halide lasers, we become increasingly more confident that they will realize their potential as practical high-average-power UV sources.

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Fig. 9-12. Gas scrubber arrangement for closed-flow window purge.

Electron-Beam Switching for Rare Gas Halogen Lasers

An advanced technique for exciting and preionizing discharge rare gas halogen (RGH) lasers is being studied for improving laser performance and simplifying the power conditioning requirements for high-average-power devices. This electron-beam-switched discharge (EBS) technique, shown in Fig. 9-13 and previously described in some detail,¹⁵ uses a low-current-density, short-pulse electron beam as both the preionization source and the fast switch in laser excitation.

The main differences between preionized discharges and the EBSD lie in the timing and

Fig. 9-13. Circuit for RGH laser using electron-beam-switched discharge (EBSD) excitation.

magnitude of the various electrical inputs to the discharge circuit (see Fig. 9-13). For preionized discharges, there is at first no voltage across the electrodes. Before application of the voltage, the laser volume is preionized with UV radiation or a weak electron beam, producing an electron density of 10^8 to 10^{10} cm^{-3} between the electrodes. The main electrode voltage is applied a few tenths of a microsecond after preionization. The rise time of this voltage must be rapid because the discharge breaks down as the voltage rises. Breakdown times are generally 10 to 100 ns.

For EBSD operation, the discharge volume is not preionized. The primary capacitors are charged relatively slowly over a period of several microseconds; the voltage is kept low enough to preclude breakdown. At the appropriate time, an electron beam pulse of short duration and high intensity—substantially more than that needed for preionization—is applied to the volume to initiate the discharge. Electron-beam current densities greater than about 0.1 A/cm^2 are required, corresponding to an electron density of approximately 10^{12} cm^{-3} . After breakdown, the subsequent discharge phases are similar for both EBSD and preionization. The discharge current and voltage are determined by the discharge kinetics and characteristics of the external power supply.

The chief advantage of the electron-beam-switched discharge is reduction of the peak power handled by primary switch S_1 . The EBSD mode of operation allows pulse-charging of capacitor C_2 in about $1 \mu\text{s}$ compared with the 100-ns regime required for conventional preionized discharges. This advantage would not be so important if the fast pulse switching problems were merely transferred to

Fig. 9-14. Wire-ion-plasma (WIP) gun concept.

the electron beam system. Fortunately, the electron beam power required for switching the discharge is substantially less than the discharge power itself. Experiments have shown that beam current densities of $< 1 \text{ A/cm}^2$ in a short pulse (100 ns) are adequate for switching.

Wire-Ion-Plasma Electron Gun. The peak electron beam powers required for EBSD are relatively modest and can be generated by several different techniques. While both thermionic and cold-cathode guns can supply the requisite peak power, the added requirements of high reliability and long life for AIS applications have led us to develop the wire-ion-plasma (WIP) electron gun technology.¹⁶ The WIP gun concept, originally devised by Hughes as a continuous electron gun preionizer for CO_2 lasers, is shown in Fig. 9-14. The electron source for the high-energy electron beam is generated at the molybdenum cathode surface as secondary electrons emitted by bombardment of fast He^+ ions. The He^+ ions, generated in the wire-ion plasma source located at ground potential, are extracted from the plasma through a transparent grid and then accelerated to full cathode potential. When they impact the cathode surface, approximately 15 e^- secondaries are generated per ion. These secondary electrons in turn are accelerated backwards by the cathode field through the transparent plasma chamber and the foil. The helium gas density in the gun is maintained at 10 to $100 \mu\text{m}$, consistent with the simultaneous demand for high-voltage holdoff and adequate He^+ density for the desired electron beam current.

Fig. 9-15. Effect of grid depth on temporal behavior of electron-beam current density. Scales on the traces have been normalized for equal heights on the pulse leading edge, to highlight differences in tail behavior.

The WIP gun has two primary advantages over other electron gun sources. The absence of thermionic or field emitters permits extreme ruggedness and long life. Also, the high-voltage accelerating field is maintained at dc while the electron beam is controlled at ground potential by the wire-ion plasma source. These features suggest excellent prospects for the high reliability and long life required for AIS applications.

Repetitive Operation and Grid Control. To be useful for RGH laser excitation, the WIP gun—originally devised for cw operation—must be capable of pulsed operation at kilohertz repetition rates for the requisite current density and pulse length. The major uncertainty about repetitive operation stems from the unknown characteristics of the wire plasma under pulsed excitation. A program initiated with Hughes this year successfully demonstrated that the gun is capable of generating repetitive pulses at 10 kHz in burst-mode operation with peak current densities of 1 A/cm^2 in 200-ns pulses.¹⁶

The dynamics and design of the wire-ion plasma and associated grid structure are crucial to detailed behavior of the electron beam pulse. For a fixed gun geometry and gun voltage, the WIP current density controls, to first order, the electron-beam current density. The WIP current pulse length, together with the plasma recombination time and grid depth, in turn controls the electron-beam pulse length. Figure 9-15 presents oscillograms of the time integral of the electron

beam's current pulse measured with a Faraday cup for grid configurations at two different depths: 2 and 4 cm. While both grids produce a pulse with a short leading edge of about 200 ns, the pulse from the shallow grid has a much longer tail—lasting up to about 1 ms—which results from insufficiently rapid recombination of the plasma in the wire plasma chamber. The long tail would lead to increased foil dissipation and heating. However, as the oscillograms indicate, the tail can be reduced or eliminated by appropriate grid design.

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Atomic Vapor Spectroscopy

Survey Spectroscopy of Atomic Uranium Emission Spectrum with Fourier Transform Spectrometer

This year we have investigated the emission spectrum of atomic uranium, using the Fourier transform spectrometer (FTS) at Kitt Peak

National Observatory (KPNO). The primary purpose of this study was to obtain accurate relative intensities of the uranium emission spectrum over the widest possible spectral range. Such intensity data can then be used to determine the branching ratio for transitions: when the lifetime of the upper level is known, the transition probability or absorption cross section can be determined.

Cross sections of optical transitions are needed for evaluating potential processes of uranium I.I.S. Cross sections of other transitions in neutral and singly ionized uranium are desirable for diagnostic purposes, such as

- Determining atom concentrations in various levels of neutral uranium in a vaporization source.

- Determining the concentration of singly ionized species before and after laser radiation.

Our secondary purpose was to record the ^{235}U spectrum with the FTS, which allows rapid survey of the effect of hyperfine structure (hfs) on a large number of potential transitions. Although the data obtained are not of sufficiently high resolution to determine the precise profile of any one line, they do make possible a preliminary assessment of many potentially useful transitions.

The investigation discussed in this section is the result of a collaborative effort between I.L.L. and KPNO. A contract was established under which I.L.L. would procure for KPNO an ultraviolet beam splitter with associated optics, in order to extend the FTS wavelength coverage down to at least 2500 Å, from its 1978 limit of 4000 Å. In return, KPNO would supply line lists of ^{238}U with accurate relative intensity in the range from 3800 Å to 1.2 μm. The desirable range is 2500 Å to 3.5 μm, the full range of the uranium line list developed by Engelman and Radziemski at Los Alamos Scientific Laboratory (LASL).¹⁷

Experimental. The Kitt Peak FTS was designed by J. Brault to conduct high-resolution solar spectroscopy, but it has also been employed successfully to obtain spectra of laboratory sources. The FTS has a linear relative intensity response over a 10^5 dynamic range: because of its high resolution capability, it is well-suited for obtaining accurate relative intensities in the exceedingly complex uranium emission spectrum. With the addition of three interchangeable beam splitters plus optics, and with various detectors, the FTS can cover the

Table 9-2. Wavelength ranges covered by Kitt Peak FTS, equipped with various optics and detectors.

Beam splitter	Optics	Detectors	Range
Quartz	UV	Silicon	2 200-11 000 Å
Glass	visible	Silicon	4 000-11 000 Å ^a
CaF ₂	infrared	Indium antimonide	0.7-5.5 μm ^b

^aUpper limit extended to 2.5 μm with indium antimonide detector.

^bUpper limit extended to 9.5 μm with cooled lead antimony telluride detector.

wavelength range from 2200 Å to 9.5 μm (45 400 to 1000 cm⁻¹). The interchange of beam splitters and optics involves considerable effort, but as Table 9-2 shows, considerable range is available with a single set. The instrument has a 1-m maximum path difference, giving a maximum resolution ($\nu/\Delta\nu$) of 3×10^6 .

The FTS instrument requires a spectrum source that is stable in intensity for the duration of the scan (10 to 60 min). We used a hollow cathode lamp that fulfills this requirement very well, and which is also less subject to self-absorption than other available sources such as the arc or electrodeless lamp. Self-absorption acts to reduce the relative intensity of lines combining with low-lying or metastable levels of the atom; this reduction in intensity leads to inaccurate (low) branching ratios for such transitions. The cathodes used were depleted uranium (99.8% ^{238}U) cylinders 38 mm long by 12.7 mm in diameter, with a 6.5-mm hole through the center length of the cylinder; the cathodes were made at LLL.

Three scans were taken of this source (see Table 9-3). The first scan covered the full spectral range from 2400 to 12 000 Å. The other two covered the effective ranges from 3500 to 7000 Å and from 5600 to 11 800 Å. After each of these scans, the spectrum of a calibrated tungsten filament lamp and a quartz halogen tungsten lamp also were recorded, to determine the spectral response or efficiency of the spectrometer. The calibrated tungsten lamp output is very low in the UV range and is useful only to about 4500 Å (22 000 cm⁻¹); the quartz halogen tungsten lamp enabled us to determine the spectrometer efficiency to about 3850 Å (26 000 cm⁻¹). An argon mini-arc continuum source that would have enabled determination of the spectrometer efficiency down to 2500 Å (40 000 cm⁻¹) was delivered in time for our planned August run, but the

Table 9-3. Record of FTS scans taken July 29 to August 2, 1979, at Kitt Peak National Observatory.

Record number	Source	Range (Å)	Useful range (Å)
7/31/79-1	^{238}U hollow cathode	2 400-12 000	4 000-11 700
2	Mini-arc	2 400-12 000	unstable
3	Quartz iodine	2 400-12 000	3 850-12 000
4	NBS tungsten	2 400-12 000	4 500-12 000
8/1/79-1	—	—	—
2	^{238}U hollow cathode	3 500- 7 000	3 850- 6 500
3	^{238}U hollow cathode	5 600-12 000	5 800-11 700
4	NBS tungsten	5 600-12 000	5 600-12 000
5	NBS tungsten	3 500- 7 000	4 500- 6 800
6	Quartz halogen	3 500- 7 000	3 850- 6 800
7	^{238}U EDL	3 500- 7 000	
8	^{235}U EDL	3 500- 7 000	
9	^{235}U EDL	5 600-12 000	
10	70% ^{235}U , 30% ^{238}U EDL	3 500- 5 000	3 600- 4 950 ^a
8/2/79-1	70% ^{235}U , 30% ^{238}U EDL	3 500- 7 000	
2	^{235}U EDL	3 500- 5 000	3 600- 4 950 ^a
3	70% ^{235}U , 30% ^{238}U EDL	3 500- 7 000	

^aHigh-resolution scans.

necessary power supply did not arrive until November. Thus, our intensity data are most reliable for wavelengths above 3900 Å (below 25 630 cm⁻¹).

For observation of the ^{235}U spectrum, we used electrodeless lamps (EDLs). We would have preferred to use a ^{235}U hollow cathode, because it has low Doppler broadening and self-absorption characteristics. It was ruled out because of lack of materials to assemble a 99% ^{235}U cathode and because of the problems associated with handling such a radioactive source (the Kitt Peak FTS is located at a site with essentially no provision for handling relatively large amounts of radioactive material). We used EDLs containing 99.7% ^{235}U , and containing 70% ^{235}U plus 30% ^{238}U as iodides; the total amount of ^{235}U was less than 2 mg. A lamp containing ^{238}U (depleted uranium) was also used. A total of seven scans were made using these EDLs as sources (see Table 9-3).

Results. The raw data from the FTS scans were reduced by Brault and Testerman of KPNO. The final format for the intensity data are line lists and Gould plots (of uncorrected intensity vs wavelength); for the ^{235}U emission survey data, only Gould plots were made. The line lists contain the wave number, wavelength, line width (FWHM), corrected intensity, and equivalent width (intensity times line width) for each observed spectral feature. The equivalent width values represent the integrated intensity for the lines and are the values used as the relative intensities.

The lists were received as hard copy and as magnetic tape. Three semifinal listings were generated from scans 7/31/79-1, 8/1/79-2, and 8/1/79-3, covering the wavelength ranges shown in Table 9-3. Because of the large range covered by the 7/31/79-1 scan, the resolution was lower and ghosts are more probable than for the other two scans, so there is a greater chance that blends and ghosts may have distorted the calculated intensities from this scan. We have therefore combined the 8/1/79-2 intensities from 3525 to 6200 Å with the 8/1/79-3 intensities from 6200 to 11 800 Å to form our working line list of relative intensities. The list contains over 50 500 line positions, with approximately 47 000 entries in the range of 3900 to 11 800 Å where the relative intensities are believed reliable.

This working list has been combined with the LASL line list mentioned above, which contains energy level assignments, isotope shift, and other spectroscopic information, to produce a master listing that now contains reliable relative intensities for the range from 3900 to 11 800 Å (25 600 to 8450 cm⁻¹). The intensity scale runs from about 1 for the weakest features in the FTS spectra to 16 890 for the strongest feature, the 5915-Å line of uranium.

Branching Ratios. The master list is sorted by all known upper levels of uranium; for each such upper level there is a list of all known transitions from that level. Table 9-4 is an example of the relative intensity data for the 19 783 cm⁻¹, J = 6 even level of neutral uranium. The wave number

Table 9-4. Relative intensities of transitions from $19\,783\text{ cm}^{-1}$, $J = 6$ even level of neutral uranium.

Wave number	Wavelength	Upper level	J	Lower level	J	Relative intensity	Intensity rank	Branching ratio	Isotopic shift
3 876.996	25 786.128	19 783	6	15 906	6				
3 979.023	25 124.943		6	15 804	6				
4 241.316	23 571.158		6	15 542	5				
4 429.670	22 568.886		6	15 353	7				
4 812.816	20 772.187		6	14 970	5				
5 220.982	19 148.258		6	14 562	5				
6 380.799	15 667.737		6	13 402	6				0.400
6 421.939	15 567.368		6	13 361	6				
6 957.019	14 370.045		6	12 826	7				0.500
8 106.297	12 332.714		6	11 677	7				
8 326.024	12 007.248		6	11 457	6				0.600
8 493.069	11 771.085		6	11 290	5	0			
9 494.718	10 529.286		6	10 288	6	54	9	0.0059	
9 528.339	10 492.134		6	10 254	5	82	8	0.0089	
9 702.307	10 304.003		5	10 081	5	24	13	0.0026	
9 714.160	10 291.430		6	10 069	7	201	7	0.022	0.150
11 664.702	8 570.5177		6	8 118	7	4 003	1	0.437	0.485
11 919.130	8 387.5691		6	7 864	5	405	5	0.0442	-0.046
12 457.216	8 025.2688		6	7 326	7	47	10	0.0052	
12 777.802	7 823.9192		6	7 005	6	366	6	0.040	-0.015
13 534.306	7 386.5968		6	6 249	6	43	12	0.0047	
14 021.255	7 130.0638		6	5 762	5	1 363	2	0.149	-0.020
15 507.639	6 446.6571		6	4 275	6	7	14	0.0007	
15 982.505	6 255.1114		6	3 800	7	46	11	0.005	
19 163.010	5 216.9343		6	620	5	1 176	4	0.128	-0.040
19 783.336	5 053.3502		6	0	6	1 345	3	0.147	-0.045

and wavelength of each transition are given in the first two columns, the upper level and its J value are given in columns 3 and 4, the lower level and its J value are given in columns 5 and 6, the relative intensity is given in column 7, and the intensity rank in column 8. Column 9 shows the branching ratio BR_{ik} , given by

$$BR_{ik} = \frac{I_{ik}}{\sum_n I_{in}}, \quad (4)$$

where $\sum_n I_{in}$ is the sum of the intensities of all known transitions from the upper level i , and I_{ik} is the intensity of the particular transition for which the branching ratio is being determined. The final column in Table 9-4 lists the isotope shift, in cm^{-1} .

As mentioned earlier, we obtained no relative intensities at wavelengths above $11\,800\text{ \AA}$. For the $19\,783\text{ cm}^{-1}$ level (Table 9-4), there are 11 infrared transitions with no measured relative intensity. Although infrared transitions are generally weak, branching ratios determined assuming that they are zero may still be subject to considerable error, as any intensity in infrared transitions will reduce the

ratios. To obtain reliable relative intensity data in the infrared region, we plan to use the Kitt Peak FTS to observe the uranium emission spectrum out to approximately $50\,000\text{ \AA}$. The hollow cathode source conditions will be set as close as possible to the conditions used to obtain the visible and ultraviolet data. The infrared observations will overlap our previous measurements in the region from 7000 to $12\,000\text{ \AA}$, so that the new measurements can be corrected to the same relative intensity scale. These measurements are scheduled to take place in August or September 1980, when an infrared beam splitter is scheduled to be installed in the FTS.

Transition Probabilities and Cross Sections. If we know the radiative lifetime τ of the upper level i , the transition probability A_{ik} can be determined from the branching ratio by the relation

$$A_{ik} = \frac{BR_{ik}}{\tau_i}. \quad (5)$$

The absorption cross section σ_{ik} at the peak of a purely Doppler-broadened line can be calculated from the relation

Fig. 9-16. Two Kitt Peak FTS scans of a 5-cm⁻¹ region of uranium emission spectrum, to assess broadening of transitions due to ²³⁵U hyperfine structure (hfs). Upper scan is of pure ²³⁵U; lower scan is of 70% ²³⁵U, 30% ²³⁸U. Effect of hfs on line width is most apparent for line at right (wave number 21 768).

$$\sigma_{ik} = \frac{2}{\Delta\nu_D} \sqrt{\frac{\ln 2}{\pi}} \frac{\lambda_0^2 g_i^2}{8\pi g_k} A_{ik} \quad (6)$$

where $\Delta\nu_D$ is the Doppler width of the line; λ_0 is the wavelength of the transition (in cm⁻¹); and g_i and g_k are the degeneracies ($2J + 1$) of the upper and lower levels, respectively.

Since the hollow cathode emission includes lines from the singly ionized spectrum of uranium along with lines from the neutral spectrum, branching ratios can be determined for singly ionized uranium also. To determine cross sections for singly ionized uranium, it will be necessary to

measure the lifetimes of its transition levels; this may be possible using laser spectroscopy techniques.

Effects of Hyperfine Structure. The Gould plots of the ²³⁵U and 70% ²³⁵U, 30% ²³⁸U FTS scans provide a method to quickly assess the broadening of the ²³⁵U transition due to the hfs effects. Figure 9-16 shows two scans of a 5-cm⁻¹ (~1 Å) region of the uranium spectrum at 4593 Å (21 766 cm⁻¹), in which there are three prominent uranium transitions. The upper scan is of pure ²³⁵U, while the lower scan is from a 70% ²³⁵U, 30% ²³⁸U source; the ²³⁸U transitions are identified on the lower scan. Some broadening of the ²³⁵U lines is present for

each transition but is most evident for the line on the right. The isotope shift (^{238}U -transition wave number minus ^{235}U -transition wave number) is positive for the transition on the left and negative for the other two transitions. As this is a relatively simple portion of the very complicated uranium emission spectrum, one can see the value of a high-resolution spectrometer for studying the uranium spectrum.

Conclusions. Our observation of the uranium emission spectrum on the high-resolution Kitt Peak FTS has produced the most reliable relative intensity data available for the spectral region from 3900 to 11 800 Å. These data can be used to obtain branching ratio data for a large number of transitions in neutral and singly ionized uranium, and thence to screen for transitions for use in laser isotope separation processes and diagnostics. Observations in the region from 11 800 to 50 000 Å, which are needed to improve the branching ratio data, should be completed by the end of 1980.

Observations of ^{235}U emission spectra with the FTS have provided data useful for rapidly assessing the broadening of the ^{235}U lines due to the effects of hyperfine structure.

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Characterization of Uranium Hollow Cathode Source and Demonstration of ASE in Uranium Vapor

Cathode sputtering is known to be a simple way of producing high densities of refractory materials. Hollow cathode tubes of moderate length can be used as laser sources, or for the study of related high-density physics phenomena such as amplified spontaneous emission (ASE), radiation trapping, beam propagation, and a variety of kinetic processes. The production of uranium atoms by discharge sputtering is a much simpler and therefore

Fig. 9-17. Cross section of uranium hollow cathode device (length 100 mm).

much more desirable source for high-density physics experiments than electron-beam production of uranium vapor; in our experience, cathode sputtering provide reliable continuous operation with minimum maintenance.

We have evaluated the performance of a uranium hollow cathode source to measure the density of obtained uranium as a function of certain discharge parameters. We have also characterized the lifetime of laser-excited uranium atoms within the discharge, in order to verify that the source performance is satisfactory for studying ASE and radiation propagation physics in domains of interest to the AIS program.

The device consists of a channeled bar of uranium metal 100 mm in length; the channel is approximately 2 by 6 mm in cross section. This cathode is placed in a vacuum system evacuated to 10^{-7} Torr, after which approximately 1 Torr of rare gas is readmitted. A positively biased electrode is positioned approximately 10 mm above the channel and held at several hundred volts; either a dc discharge (from 0 to 700 mA) or a pulsed discharge (of several amperes peak current) is then applied (see Fig. 9-17).

We have measured the density of ground-state uranium atoms produced under both pulsed and continuous-wave discharge conditions. This was

done by measuring the absorption of cw laser light (from a Coherent Radiation model 599 dye laser) beamed down the cathode channel. Figure 9-18 shows the attenuation results for three such experiments, one at 5915 Å and two at 6056 Å, with both krypton and xenon at 1.5 Torr used as buffer gases. Using a cross section of $3 \times 10^{-14} \text{ cm}^2$ for uranium at 6056 Å, we found that a discharge of 700 mA produced a density of $2 \times 10^{12} \text{ atoms/cm}^3$. A Doppler width of 800 MHz was measured under dc conditions. We find that atom density appears to scale linearly with current.

Figure 9-19 shows two oscilloscope traces that record the evolution of the uranium ground-state population following a pulsed discharge. Note that a high density of sputtered uranium persists for as long as a few milliseconds. With our present hollow cathode we were unable to maintain the linear scaling needed to obtain higher densities from higher peak currents (on the order of tens of amperes). We achieved densities near 4×10^{12} with currents of 2 A (pulse width = 300 μs); higher currents or longer pulse durations resulted in parasitic discharges to various vacuum fittings and mechanical mounts attached to the hollow cathode. It is expected that improved electrical isolation, coupled with a longer pulse discharge, will substantially increase obtainable density.

We found also that the density of sputtered uranium atoms produced within a xenon discharge at a given dc current is larger than that obtained

within a krypton discharge at the same current. This is consistent with previous work¹⁸ and is probably due to a combination of xenon's more favorable momentum-transfer cross section and the large charge-exchange cross section between uranium and

Fig. 9-18. Absorption of representative cw dye laser beams in uranium hollow cathode.

Fig. 9-19. Evolution of ground-state population following 10-A, 50-μs pulsed discharge at (a) 5915 Å and (b) 6056 Å. In (a), xenon pressure is 1.8 Torr; center line of scope trace equals 100% absorption. High density of sputtered uranium persists for several ms. In (b), xenon pressure is 1.4 Torr; center line of scope trace again equals 100% absorption. High density persists for nearly 5 ms.

Fig. 9-20. (a) Lamb dip resonance width of 25 MHz at 5915 Å in 20-mA dc discharge: krypton pressure 0.6 Torr. (b) Lamb dip resonance widths at 5915 Å in a 2-A peak current dc discharge with 1.23-ms delay between probe and discharge: krypton pressure 1 Torr. Resonances as narrow as 30 MHz were observed in several other scans.

xenon ions. This latter property would ensure a more rapid crossover into a uranium ion rather than a rare-gas-ion sputtering regime; this in turn would increase the momentum transfer per sputtering event even further.¹⁹

Demonstration of Amplified Spontaneous Emission. Efficient photoionization of atomic vapor requires that process transitions be chosen which minimize the possibility of ASE. Unfortunately, the high densities, strong oscillator strengths, and long vapor paths used in the AVLIS process are all favorable to the production of ASE. Due to the complexity of the uranium spectrum, the best argument which can be made against the occurrence of ASE under process conditions for any given set of transitions is an experimental demonstration of its absence.

For the hollow cathode source to be useful for ASE experiments, it must first produce electrically excited uranium atoms with a lifetime in excess of several nanoseconds. To demonstrate that the lifetime of the excited atoms is sufficiently long, we measured the width of the Lamb dip resonance at 5915 Å both in the cw discharge and also at various delay times after the initiation of the pulsed discharge. Figure 9-20 shows that the resonance width is about 25 MHz (FWHM); since the lifetime of electronically excited atoms is the HWHM of the Lamb dip spectrum, the lower lifetime limit in the discharge is on the order of 10 ns.

Our measurements indicate that this width does not seem to vary measurably as a function of the delay after initiation of the discharge. This

suggests that the width may well be instrument-limited, but since the excited-atom lifetime is at least 10 ns, the exact width is probably not important. Also note that that we are most probably observing not electronic relaxation, but translational relaxation, to which the Lamb dip line width is very sensitive; our experimental results are consistent with this observation.

To demonstrate that hollow cathodes can be used to study the effects of ASE in multistep photoionization schemes we have selected for study a strong transition, $7006\text{ cm}^{-1} - 33\,571\text{ cm}^{-1}$ (Ref. 20), in which the excited state can be accessed from the ground state by the frequency-doubled output of a dye laser. To ensure that inversion is uniform along the entire length of the cathode, the number of photons delivered must be larger than the number of atoms irradiated ($n_p > n_a$); we have therefore used the doubled output of a Quanta-Ray YAG pumped dye laser with a per-pulse energy of 3 mJ and a line width of about 5 GHz; the pump wavelength is 2978 Å. The radiation is focused into the cathode through a 100-nm lens. Approximately 100 μJ of laser light is delivered into the cell on resonance during each pulse. Under the conditions of our experiment, irradiation over a cross section of approximately 2 mm by 2 mm then gives $n_p \sim 10^{14} > n_a \sim 10^{13}$.

The hollow cathode is operated with dc currents ranging from 0 to 1000 mA. Xenon at 1.5 Torr is used as the sputtering gas. Amplified spontaneous emission is detected with a Moletron LP-141 detector placed approximately 250 mm

Table 9-5. Strong emission channels from representative highly excited electronic states in uranium vapor.

Upper state (cm^{-1})	Lower state (cm^{-1})	Emission intensity from LASL line list	gA value
33 571	0	245	—
	3 800	328	—
	7 006	343	12.0
	12 826	321	—
33 341	0	369	1.9
	4 275	329	—

from the end of the hollow cathode. A Corning 4-97 filter isolates the detector from the laser light while transmitting about 75% of the atomic emission between $3700 \text{ \AA} < \lambda < 5700 \text{ \AA}$. The tabulated emission spectrum from the experimentally accessed levels is summarized in Table 9-5.

The $33\,341\text{-cm}^{-1}$ transition is included to determine whether the observed signal from $33\,571\text{-cm}^{-1}$ results from a discharge artifact (the $33\,341\text{-cm}^{-1}$ level is not expected to exhibit ASE, as its strongest transition is back to the ground state and therefore inversion cannot be achieved by optical pumping unless one resorts to the use of π pulses). In early experiments performed with a poor vacuum system, the application of intense laser radiation to the discharge channel caused arcing: the sudden passing of high currents through the device subsequently showed up on the detector as a delayed signal. The use of a clean vacuum (base pressure less than 10^{-7} Torr) eliminated this artifact, but the transition at $33\,341\text{-cm}^{-1}$ is still used before each experiment to verify that the condition remains eliminated.

Figure 9-21 gives the detector signal intensity as a function of cathode current (a current of 1 A corresponds to an atom density of about 3×10^{12} ground-state uranium atoms cm^{-3} ; density decreases linearly with decreasing current over the range from 100 to 1000 mA). The ASE was too weak to be observed visually, but the signal was observed to arrive promptly at the detector (rather than in delayed fashion, as was the case with the arc-related signal). Furthermore, using a series of Corning filters, the ASE signal was isolated to $3700 \text{ \AA} < \lambda < 5700 \text{ \AA}$, while arc-related signals are apparent at all wavelengths. Importantly, consistent with its assignment as being due to ASE, the signal was spatially coincident with the laser pump beam. Signal intensity was more or less independent of detector distance from the cathode, and decreased as laser pump intensity or cathode current was

Fig. 9-21. ASE intensity as a function of hollow cathode current.

decreased. The slope in Fig. 9-21 shows the ASE threshold, followed by a signal intensity rising more rapidly than a unity slope, which would indicate saturated gain. A rough estimate of the point at which ASE threshold is reached yields $20 < n^* \sigma I < 100$.

Finally we measured the ASE wavelength using a 0.3-m monochromator. A survey of the dispersed ASE showed not only the expected peak at 376 nm but also an emission at 482 nm (± 5 nm) of approximately equivalent intensity. We observed no other wavelengths between $350 < \lambda < 700$ nm. According to recent Kitt Peak data, the ratio of the Einstein coefficient times degeneracy (gA) value from the $7006\text{-cm}^{-1} \leftarrow 35\,571\text{-cm}^{-1}$ (376 nm) transition to the gA value from the $12\,826\text{-cm}^{-1} \leftarrow 35\,571\text{-cm}^{-1}$ (482 nm) transition is 1.45 to 1. All the other emission channels from the $33\,571\text{-cm}^{-1}$ level are substantially weaker. The 482-nm signal also had a threshold behavior and increased with density; unfortunately, the variation of the two presumed ASE spectral bands as a function of uranium density could not be mapped independently as, at this stage

of the experiments, the channel of the hollow cathode collapsed due to repeated cycling of the cathode through the uranium phase change temperature at 930 K.

Now that we have shown that the presence of ASE can be monitored using a hollow cathode as a vapor source, we are in a position to study its influence on process operating conditions. The experimental arrangement used in these studies should prove invaluable in the study of high-density physics phenomena relevant to the AVLIS process. We have already proven its value in studying ASE and expect it to be similarly useful in a variety of beam-propagation experiments.

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Advanced Laser Research

Pulse Amplification of a Continuous Wave Laser

Many experiments in high-resolution spectroscopy require both the very narrow line widths characteristic of continuous wave (cw) dye lasers and the high peak powers characteristic of pulsed lasers. The most common laser source used in such experiments is a pulsed oscillator with sufficient intracavity dispersion to produce output on a single longitudinal mode despite the high gain produced by high-peak-power pulsed-laser pumping.^{21,22} A few authors have tried the more straightforward method of amplifying a narrow-band cw dye laser with one or more high-gain amplifiers pumped by pulsed lasers.²³⁻²⁵ The difficulty with the latter scheme has always been the very low power of the cw beam; most of the amplifier output comes from amplified spontaneous emission (ASE), requiring

dispersive elements to filter the amplification. Thus considerations of both efficiency and signal-to-noise ratio have favored the pulsed oscillator.

Single-mode cw ring dye lasers now produce over five times more power than the older standing-wave cavity lasers. The frequency-stabilization and scanning features of newer cw dye lasers also make pulsed amplification attractive. We have therefore conducted a series of experiments on pulsed amplification of a cw ring laser, using both nitrogen and copper vapor lasers to pump the dye amplifier.

A typical experimental setup is shown schematically in Fig. 9-22. A commercial argon ion laser is used to pump a commercial Rhodamine 6G ring dye laser; the single-mode output of this laser is focused into a dye cell of rectangular cross section. The gain length in the ring dye laser is 8 mm; the width and flow rates are adjusted so that the pumped volume is replaced between pulses. For nitrogen pumping, amplifying 5900-Å light, the dye is Rhodamine 6G in ethanol solution; for CVL pumping, amplifying 6000-Å light, the dye is Kiton Red S (also in ethanol). Temporal properties of the amplified signal are analyzed with a fast photodiode whose output is displayed on an oscilloscope; in the CVL experiments, amplifier output power is also measured on a power meter. Spectral properties of the signal are characterized with a Fabry-Perot interferometer (preceded, in the CVL case, by a monochromator).

Table 9-6 gives the experimental results of this single-stage amplification process for both CVL and nitrogen laser pumping. The results for the two systems are similar. We obtain optimum results when the cw laser beam remains focused along the entire length of the amplifier (i.e., when the Rayleigh range equals half the dye cell length). This corresponds to a focal-spot diameter of 60 μm . Approximately 0.6% of the pump energy is extracted at the cw laser frequencies. Background ASE accounts for about 14% of the energy emitted into the solid angle of the laser beam. The bandwidth of the ASE is about 80 Å, giving an ASE relative brightness (intensity per unit bandwidth) of less than 10^{-6} . The cw laser's 40-MHz line width is reproduced in the CVL case, while a nearly Fourier-limited line width of 130 MHz is obtained in the nitrogen case. The laser beam transverse profile is in each case about twice the Gaussian diffraction limit, due both to the exponential transverse gain profile in the amplifier, and the cw beam's deviation from a Gaussian

Fig. 9-22. Experimental setup for pulsed amplification of continuous wave laser.

profile. Both amplifiers exhibit output pulses shorter than their pump pulses, because of lower gains in the tail of the pump pulse.

The different properties of the pump lasers determine the properties of the amplified signal. The nitrogen laser has higher pulse energy and shorter pulse duration than the CVL. Consequently the amplified pulse energy and peak power are higher with the nitrogen laser pump. On the other hand, the high pulse repetition rate of the CVL provides much higher average power in the amplified beam. Further amplification is of course possible; a second (identical) stage of nitrogen pumping amplification has produced a 200- μ J pulse, while second-stage CVL pumping gives a 50- μ J pulse. Given these variations, the choice of pump laser should be dictated primarily by the requirements of the spectroscopy experiment being performed.

The amplifier properties observed for CVL pumping compare less favorably with the properties of a CVL-pumped single-mode oscillator. The efficiency of the oscillator is better: about 2%, compared to 0.6% for the amplifier. ASE for the oscillator is only 2% of the laser output, compared to 14% for the amplifier. The line width of the oscillator is larger than the pulsed CVL amplifier: 60 MHz vs 40 MHz (or even 60 MHz vs approximately 20 MHz, corresponding to Fourier-limited

Table 9-6. Summary of experimental parameters and results, using nitrogen and copper vapor lasers to pump the dye amplifier.

Parameter	CVL	Nitrogen
Energy in pump pulse	0.7 mJ	3 mJ
Duration	~30 ns	~8 ns
Repetition rate	6000 Hz	30 Hz
cw dye laser line width	<40 MHz	<40 MHz
cw dye laser focal-spot diameter	60 μ m	60 μ m
cw dye laser intensity at focus	2×10^4 W/cm ²	2×10^4 W/cm ²
Energy in amplified pulse	4 μ J	18 μ J
Duration	18 ns	5 ns
Line width	<40 MHz	<130 MHz
Peak power	220 W	3600 W
Average power	24 mW	<0.54 mW
Extraction efficiency	0.6%	0.6%
ASE energy fraction per pulse	14%	14%
ASE bandwidth	80 Å	80 Å
ASE relative brightness	<10 ⁻⁶	<10 ⁻⁶
Beam quality	3X	3X

amplification of a more stable cw laser). Still, the properties of CVL oscillators and amplifiers are similar enough that considerations of convenience and intended application should determine the individual experimenter's choice of laser system.

In summary, we find that pulsed amplification of a single-mode cw laser provides a convenient source of tunable, narrow-band, pulsed radiation.

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ArF Excimer Laser Injection-Locking and Amplifier Energy Extraction

Much attention has been paid recently to line-narrowed, tunable radiation from excimer lasers for photochemical, spectroscopic, and laser fusion applications.²⁶⁻²⁹ Interest has been expressed, in particular, in the ArF excimer laser, which provides UV photon energies (6.4 eV). In studying tech-

niques for generating useful outputs from ArF laser oscillators, we have achieved very narrow line widths (0.25 cm^{-1}), broad, continuous tuning ranges (360 cm^{-1}), and close to diffraction-limited output from an injection-locked amplifier. In ArF oscillator-amplifier experiments we have also determined such important laser scaling parameters as small signal gain, saturation fluence, optical energy density, and nonsaturable loss. In one application we used the injection-locked amplifier to generate tunable radiation efficiently from the VUV (1238 Å) to the visible (4412 Å) using stimulated Raman scattering in H_2 (see following article on tunable VUV generation by high-order Raman frequency shifting).

The ArF oscillator and amplifier transverse discharge laser heads, along with the narrow-line oscillator characteristics, have been previously described.³⁰ The oscillator and amplifier preionization and discharge circuits for these experiments are switched by individual thyatrons to achieve precise temporal synchronization among all components. The discharge volumes are both $100 \times 2.0 \times 0.5 \text{ cm}$. The laser gas-flow system combines fluorine, argon, and helium buffer in an optimum ratio (1:40:360) and is capable of 30 liters/min longitudinal flow, allowing continuous 10-Hz operation.

Frequency-selective optics for the oscillator consist of two prisms and two etalons in a half-symmetric (flat and convex mirror) unstable

Fig. 9-23. Unstable resonator configuration for injection-locked ArF amplifier. Energy is injected through 1.0-mm hole in R_1 and extracted off scraper mirror; resonator magnification of 4.0 corresponds to 94% geometric output coupling.

resonator configuration. The oscillator's narrow-band (0.25 cm^{-1}), continuously tunable (from 1928 to 1940 Å FWHM) radiation is used to injection-lock the confocal unstable resonator-amplifier through a hole in the concave cavity mirror (see Fig. 9-23). The hole was chosen to be the diameter of the first Fresnel zone (1.0 mm) and acts as a spatial filter for the injected signal.

Our experiments show that this configuration improves the output beam quality as well as the energy extraction. Injecting 2.0-mJ broad-band (120 cm^{-1}) energy into the amplifier cavity doubles the output energy from 45 to 90 mJ; the amplifier beam divergence decreases from 40 to 7 times the diffraction limit. The pulse length also increases from 8 to 16 ns, so the improvement in output energy can be

attributed to minimizing the optical buildup time within the resonator.

We have also determined the injection-locked amplifier spectral characteristics. Typical results are shown in Fig. 9-24. The amplifier line width was reduced from 120 cm^{-1} to 15 cm^{-1} by injecting a 200- μJ 20-cm^{-1} -line-width signal from the oscillator. The effective amplifier tuning range, determined by the points at which fluorescent output becomes significant, is 360 cm^{-1} (13.5 Å). These spectra [Fig. 9-24(a)] were recorded in the film plane of a 1.0-m spectrometer using a linear optical multichannel analyzer (OMA) with a resolution of 50 cm^{-1} . Higher-resolution spectra ($\sim 5\text{ cm}^{-1}$) were determined by photographing the ring pattern produced

Fig. 9-24. (a) Laser line width with and without injection-locking. Lower trace shows amplifier line width and relative intensity as 20 cm^{-1} -line-width injected signal is tuned. (b) Amplifier line width (0.45 cm^{-1}) with 0.25-cm^{-1} injected signal is given by Fabry-Pérot ring pattern shown. Resolution is 0.15 cm^{-1} .

(a)

Analyzer etalon free
spectral range: $1,150\text{ cm}^{-1}$

(b)

by an analyzer etalon (finesse of 10). Injecting a 30- μ J, 0.25-cm⁻¹-FWHM line width signal into the amplifier resulted in a 32-mJ, 0.45-cm⁻¹ output [Fig. 9-24(b)]. Table 9-7 summarizes these measured values with the amplifier operating at less than optimum output energy. The output line widths in the table are broader than the input because of a difference in measurement technique. Oscillator line widths were determined in earlier experiments using an OMA to record the ring pattern and represent the FWHM value. Amplifier line widths represent the full width (zero to zero) of the photographed ring. This results in a measurement which, depending somewhat on film exposure, is roughly a factor of 2 greater than the FWHM determined with an OMA. Taking this factor of 2 into account, the injection-locked amplifier spectral characteristics closely match those of the injected signal, as expected.

The ultimate utility of RGH lasers in high-energy and average-power applications depends on oscillator-amplifier staging. One of the chief issues concerning RGH amplifiers is the level of nonsaturable loss relative to the small signal gain. Nonsaturable losses result from such mechanisms as photoionization of constituent atoms or molecules, photodissociation of rare gas dimer ions or halogen molecules, and photodetachment of negative rare gas ions. These mechanisms are potentially significant in all RGH media and can severely limit the obtainable amplifier output fluence and extraction efficiency. Therefore, we have investigated single-pass ArF amplifier characteristics using the unstable resonator laser as a probe. The low divergence of the unstable resonator input effectively discriminates between the amplifier output signal and its highly divergent single-pass fluorescence, so the small signal gain can be accurately determined. The oscillator output energy is also sufficient to saturate the gain medium strongly, so effects due to significant nonsaturable loss can be measured.

The effect of nonsaturable losses can be modeled by assuming a steady-state, homogeneous extraction. Since ArF is homogeneous and its optical pulse length (15 ns) is considerably longer than its upper-level lifetime (2 ns), this assumption is reasonable. The growth of the field $I(z)$ in the amplifier is then given by

$$\frac{1}{I(z)} \frac{\partial I(z)}{\partial z} = \frac{g_0}{1 + I(z)/I_s} - \alpha_0 \quad (7)$$

Table 9-7. Injection-locked unstable resonator energy and line width at 1933 Å.

Measured injected signal		Measured injection-locked output	
Line width (cm ⁻¹) (FWHM)	Energy (μJ)	Line width (cm ⁻¹) (FWHM)	Energy (mJ)
—	0	120.0	24
20.0	~200	15.0	43
1.0	~500	2.2 ^a	40
0.25	~30	0.45 ^a	32

^aValues measured on film; FWHM is approximately one-half this value.

where g_0 , α_0 , and I_s are the small signal gain, nonsaturable loss, and saturation flux, respectively. Integration of Eq. (7) yields the amplifier gain saturation characteristics:

$$\ln G = g_0 \psi(1 - \Gamma) - \frac{1}{\Gamma} \ln \left[\left(1 - \frac{\Gamma I(z=0)}{(1 - \Gamma) I_s} \right) / \left(1 - \frac{\Gamma I(z=L) G}{(1 - \Gamma) I_s} \right) \right] \quad (8)$$

In this equation, G is the gain $I(z=L)/I(z=0)$ and $\Gamma = \alpha_0/g_0$. In the limit where the nonsaturable losses are small or the amplifier is not saturated, i.e., $(\Gamma/1 - \Gamma) I(z=0)/I_s \ll 1$, expansion of the last term in Eq. (8) yields a form similar to the lossless ($\alpha_0 = 0$) case:

$$\ln G \approx g_0 \psi(1 - \Gamma) - \frac{I(z=0)(G - 1)}{I_s(1 - \Gamma)} \quad (9)$$

In this approximation, one expects that a graph of experimental data with $\ln G$ and $I(z=0)(G - 1)$ as the ordinate and abscissa will yield a straight line. The intercept and slope are related to small signal gain and saturation flux. Significant nonsaturable losses result in a distinct deviation from a linear saturation curve at high levels of saturation and limit the potential extraction efficiencies.

Figure 9-25 shows experimental saturation data for ArF. The data derive from varying the optical input energy E_1 to the amplifier by inserting calibrated neutral density filters into the probe beam and measuring the resulting output energy E_2 . Also, the probe beam area (2.0 × 0.5 cm) closely matches the gain volume; hence, the data represent both temporal and spatial averages. Window transmission is accounted for, so energies are values on inside surfaces. The dashed curve is a least-

Fig. 9-25. ArF amplifier gain saturation data. Dashed curve is least-squares fit to data using Eq. (9). Solid curves show calculated effect [Eq. (8)] of varying gain to nonsaturable loss ratio (g_0/α_0). For negligible losses, small signal gain and energy density are given by ordinate and abscissa intercepts, respectively. Inverse slope yields saturation fluence.

squares fit to the data using $g_0\ell(1 - I')$ and $I_s(1 - I')$ as fit parameters and assuming low nonsaturable losses [Eq. (9)]. Obviously the fit is quite good even to high levels of saturation. The solid curves in Fig. 9-25 are calculated from Eq. (8) assuming different values of I' but using the least-squares fit values of $g_0\ell(1 - I')$ and $I_s(1 - I')$. Comparison of the data with these curves places an approximate lower bound on the gain-to-loss ratio: ($I/I' \lesssim 50$). The least-squares fit parameters (assuming I' is negligible) yield the small signal gain (7.0%/cm), saturation fluence (24.5 mJ/cm²), and energy density (1.72 J/l) for the ArF medium (laser gain volume is 0.10 cm³). The important consequence of low non-

saturable loss in ArF is that extremely high optical extraction efficiencies are possible. In these experiments 81% of the available energy was extracted (139 mJ/172 mJ).

The amplifier electrical efficiency can be estimated from circuit voltage and capacitance data. There are two types of capacitors: a 50-nF capacitor with 21-J stored energy (dc-charged to 29 kV) is switched by the thyatron across 17-nF barium titanate peaking capacitors which are closely coupled to the discharge electrodes. The actual

energy transferred to the peaking capacitors (also at 29 kV) is only 7.2 J because of a nonoptimal circuit design and unavoidable switch losses. The energy in the barium titanate capacitors may be less than 7.2 J due to a characteristic decrease in capacitance with increasing voltage. The derating factor is 1.4 at 29 kV, but the pulse-charged (~ 100 ns) factor is not known. Hence, the ArF excited-state production efficiency relative to the energy in the peaking capacitors is 2.4%, or perhaps as high as 3.4% if the 1.4 derating factor applies. The efficiency drops to 0.82% when circuit losses are considered.

The saturation fluence for ArF can be calculated since the cross section σ , spontaneous lifetime τ_s , and ArF excited-state quenching rates k are experimentally known. The dominant quenching reaction at the gas concentration and pressure relevant to these experiments [Fig. (9-25)] is

where $k\tau_s \approx 7.8 \times 10^{-18} \text{ cm}^3$ (Refs. 31, 32). The saturation flux is given by

$$I_s = \frac{hc}{\lambda \sigma \tau_s} (1 + k\tau_s n_{\text{F}_2})^{-1}, \quad (11)$$

where n_{F_2} is the fluorine number density ($1.27 \times 10^{17} \text{ cm}^{-3}$). Using 12 Å-ns (Ref. 29) for $\sigma\tau_s$, one obtains a saturation flux of 1.71 mW/cm² which, for the 15-ns FWHM optical pulse width in these experiments, corresponds to a fluence of 25.6 mJ/cm². This is in excellent agreement with our experimental value (24.5 mJ/cm²).

These experiments have demonstrated ArF injection-locked and linear amplifiers with sufficient beam quality (7 times diffraction limit), tuning range (360 cm⁻¹), line width (0.3 cm⁻¹ FWHM), output energy (160 mJ), and potential efficiency (2 to 3%) to be extremely useful in a variety of new photochemical and spectroscopic applications. The low nonsaturable loss ($\geq 1.4 \times 10^{-3} \text{ cm}^{-1}$) also allows efficient energy extraction at high saturation levels and a simple linear amplifier scaling strategy to achieve high pulse energy.

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Tunable VUV Generation by High-Order Raman Frequency Shifting

During 1979 we extended our experimental studies and initiated new theoretical work on high-order stimulated Raman scattering (SRS). Our original motivation, as described in last year's annual report,³³ was a need for a versatile technique to generate broadly tunable coherent UV and infrared light for our atomic and molecular spectroscopy and chemistry experiments. One such experiment, involving H₂S photodissociation, is described at the end of this section. Another application being studied at LLL is the use of tunable UV and infrared sources to increase the effective bandwidth of fusion lasers, for enhanced coupling to fusion targets (see Section 7, "Increased Bandwidth for Fusion Lasers"). We have also used the stimulated Raman generator this year to study alternate LIS processes, covering spectral regions in atomic uranium that were previously inaccessible with tunable dye lasers. In this section we highlight experimental work on VUV generation, using Raman shifting of the output of the ArF laser system described in the preceding section. We also present a theoretical description of the nonlinear parametric processes involved in the observed high-order Raman effect.

Experimental. The shortest wavelengths observed in our earlier Raman work, using the fixed outputs available from a Nd:YAG laser and a dye laser, were restricted to wavelengths greater than 1700 Å. The high-order nonlinear processes were limited by optical dispersion through phase matching conditions. One solution to this difficulty in efficiently generating wavelengths farther into the VUV is to use a pump with a shorter wavelength, to

Fig. 9-26. ArF-pumped stimulated vibrational Raman spectra; shaded portion indicates vacuum ultraviolet (VUV) spectrum.

minimize the effects of dispersion and maximize the Raman gain. This approach has been investigated using the specially designed tunable ArF oscillator with injection-locked amplifier described in the previous article.

We have used broad-band radiation from the ArF laser, with SRS in H_2 , to efficiently generate tunable radiation from the VUV (1237 Å) to the visible (4412 Å); a portion of this Raman spectrum is shown in Fig. 9-26. In our experiment, a 500-mm ($\lambda/20$) lens of good optical quality is used to focus a collimated pump beam into the center of a 1-m Raman cell containing 1 to 40 amagats of high (99.999%) purity H_2 . The output Stokes and anti-Stokes beams are refocused by a 250-mm-focal-length lens, and then dispersed by a 60° fused silica prism or injected into a vacuum spectrometer. The scattered radiation is linearly polarized parallel to the pump. Circular polarization of the pump induces high-order rotational Raman components [$\nu_r(S_1) = 587 \text{ cm}^{-1}$] and reduces the conversion efficiency of the purely vibrational Raman lines.

Threshold for first Stokes (S_1) generation is observed to be independent of focusing geometry (as expected). Figure 9-27, which shows pump thresh-

Fig. 9-27. Threshold energy for first Stokes (S_1) generation as a function of I_2 density.

Fig. 9-28. Stokes (S) and anti-Stokes (AS) energy dependence on ArF pump energy.

old energy vs H_2 density, indicates that we are in the transient gain regime for S_1 generation. The data are fit (broken line) to a transient gain model³⁴ assuming a square pump pulse. A transient gain analysis is applicable since the ratio of laser pulse length to medium response is less than the steady state gain factor.

Under these conditions, the small signal intensity gain g (total gain = e^g) is then given by

$$g = 2 \left(2 \sqrt{\frac{2 T_P}{G_{se} T}} - \frac{T_P}{G_{ss} T_2} \right) G_{ss}, \quad (12)$$

where G_{ss} is the steady state small signal amplitude gain at intensity I (Ref. 34). The steady state gain is just $G_{ss} = \gamma I l = \gamma E \eta / \lambda_p$, where γ is the Raman amplitude gain coefficient equal to $\gamma = \gamma_0 \rho / \Delta \nu$. Here γ_0 is a molecular parameter proportional to ν_{ss} and $\Delta \nu$ is the pressure- or density-dependent Raman line width. The parameter η incorporates the focusability or uniphase character of the pump beam having pulse duration T_p . For a diffraction-limited Gaussian beam, $\eta = \pi$. The density-dependent relaxation rate $1/T_2 = C_0 \rho$, where ρ is density.

Our transient gain model assumes that threshold is achieved for a small signal gain g_0 independent of pump energy, gas pressure, or focusing geometry. The energy at which threshold is achieved is just

$$E_{TH} = \left(\frac{\lambda_p}{4\gamma_0} \right) \left(\frac{\Delta \nu}{\rho^2} \right) \left(\frac{E_0^2}{\eta} \right). \quad (13)$$

For our data the fit parameter $g_0^2/\eta = 244$. This indicates a small signal gain of $g_0 = 10.5$ is needed for the threshold to be reached, since we independently calibrated the pump beam to be $7 (= \pi/\eta)$ times its diffraction limit. We see for a diffraction-limited beam that less than 1 mJ/cm^2 is needed to reach the S_1 threshold at the H_2 densities of interest ($\rho \cong 1$ to 5 amagats). The threshold small signal gain is large because the Raman cell operates in an ASE mode. Because of its wavelength scaling ($E_{th} \propto \lambda_p \lambda_s$), however, threshold occurs at very low ArF pump energy.

Once threshold for S_1 is exceeded [where S_n (AS_n) is the n th Stokes (anti-Stokes) wave], the higher-order component waves emerge at various cone angles; these increase with rising Raman order and gas pressure due to phase matching requirements and dispersion. This phenomenon (which motivates the theoretical discussion that concludes this article) suggests that the high-order processes can be described by a parametric interaction.

We have observed that although the efficiencies of conversion to the vibrational Raman components depend on the spatial properties and energy in the pump as well as on the H_2 gas pressure, they are insensitive to the f -number of the input optical system. Figure 9-28 shows the energy conversions to various Raman orders as pump energy is increased to fifteen times S_1 threshold. This figure illustrates an important feature observed in every experiment

Fig. 9-29. Fourth anti-Stokes (1463 Å) energy as a function of H_2 pressure.

we have performed with various wavelength pumps and temporal formats; all the processes seem to be completely saturated, since the output energies for all Raman orders vary linearly with pump energy (except near threshold). The conversion efficiencies to S_1 and S_2 are 30% each at 32-mJ pump energy. Even S_3 (10%) and AS_1 (5%) have reasonable conversion efficiencies.

In the linear regime the conversion efficiency increases with decreasing pressure. On the other hand, energy threshold increases dramatically with decreased pressure. As a general rule, the higher the pump energy the lower the pressure at which we operate the Raman cell to maximize the conversion efficiency to any order. This effect is illustrated in Fig. 9-29 for AS_4 at 1463 Å.

With 90 mJ of broad-band ArF radiation from the injection-locked system, we have observed AS_6 (1304 Å) and AS_7 (1237 Å). The experimental observations of wavelengths shorter than 1789 Å (AS_1) were made by focusing the output of the Raman cell with MgF_2 optics into a 1.3-m vacuum spectrometer. The VUV Raman orders were monitored at the exit slit of the vacuum spectrometer by a channeltron detector. We estimate that after AS_1 the energy decreases by a factor of about 10 per anti-Stokes order (Fig. 9-30).

Theoretical. Besides being a versatile spectroscopic and diagnostic tool, the high-order stimulated Raman effect will open up new areas in quantum electronics. It is important therefore to

Fig. 9-30. Anti-Stokes energy as a function of order. AS energy falls exponentially as AS order increases.

develop a theoretical model to describe the multiwave Raman effect.

A starting point for the analysis is Maxwell's set of equations relating n waves (at $\omega_n = \omega_0 + n\omega_R$, where ω_0 and ω_R are the pump and Raman frequencies, and $n = 0, \pm 1, \pm 2, \dots$). We will restrict the discussion in the following ways:

- We consider only plane waves that are time-bandwidth-limited.
- We couple the waves through a four-wave process only, ignoring the mechanism (two-wave, etc.) by which the input waves are generated.

- We ignore optical dispersion contribution from the H_2 gas.

- We ignore backward wave generation.

- We ignore issues related to dispersion and the effect of focusing.

Although these restrictions considerably simplify the experimental study above, they will enable us to understand the cogent features of a highly nonlinear process. In any case, since conversion into higher orders by means of the parametric process is degraded by optical dispersion, the analysis will provide an upper bound on the conversion efficiencies that can be achieved.

Under these conditions, Maxwell's equations

$$\frac{\partial \epsilon_n}{\partial z} = -2\pi i \frac{\omega_n}{c} (\chi \epsilon)_n = -2\pi i \frac{\omega_n}{c} \sum_p \chi_{np} \epsilon_p, \quad (14)$$

where the traveling wave at ω_n is

$$\epsilon_n = \epsilon_n e^{i\phi_n} e^{i\omega_n \left(t - \frac{z}{c}\right)}$$

and (χ) is the complex molecular susceptibility ($\chi' - i\chi''$), become

$$2 \frac{\partial F_n(Z, \tau)}{\partial Z} = b(n) \left\{ F_{n+1}(Z, \tau) - F_{n-1}(Z, \tau) \right\}, \quad (15)$$

Here,

$$F_n = \epsilon_n e^{-in\phi},$$

$$\phi = \text{phase} \left\{ \sum_K \epsilon_{K+1}(0, t) \epsilon_K(0, t) e^{i(\phi_{K+1} - \phi_K)} \right\},$$

$$b(n) = 1 + \frac{n\omega_R}{\omega_0}, \quad (\text{Manley-Rowe Factors})$$

$$\tau = t - \frac{Z}{c},$$

and Z , the "parametric gain" coefficient that determines the conversion efficiency to any Raman order, is given by

$$Z = - \int_0^z \frac{2\pi}{c} \omega_0 \chi'' dz.$$

Z can be calculated from a knowledge of the field strengths $\epsilon_K(Z, t)$ and the relevant molecular dynamics.

We can express in a formal way the solution to Eq. (15) in a power series expansion such as

$$\Gamma_n(Z, \tau) = \sum_K \sum_p \frac{(B^K)_{np}}{K!} \left(\frac{Z}{2}\right)^K \Gamma_p(0, \tau) \quad (16)$$

or

$$\epsilon_n(Z, \tau) = \sum_K \sum_p e^{in\phi} \frac{(B^K)_{np}}{K!} \left(\frac{Z}{2}\right)^K \epsilon_p(0, \tau), \quad (17)$$

where the coefficients of the matrix B are

$$B_{nm} = b(n) \left\{ \delta_{n-1,m} - \delta_{n+1,m} \right\}.$$

As an example, suppose only

$$\epsilon_0(0, t) \gg \epsilon_{\pm 1}(0, t)$$

are present at the input of the cell, and the parametric gain Z is small. Then, for the anti-Stokes waves,

$$\epsilon_n(Z, \tau) = e^{in\phi} \frac{\left\{ \prod_{K=1}^n \left(1 + \frac{K\omega_R}{\omega_0} \right) \right\} \left(\frac{Z}{2}\right)^n}{n!} \epsilon_0(0, \tau), \quad (18)$$

or

$$\frac{\Gamma_n(Z, \tau)}{\Gamma_0(0, \tau)} = \frac{\left\{ \prod_{K=1}^n \left(1 + \frac{K\omega_R}{\omega_0} \right) \right\}^2 \left(\frac{Z}{2}\right)^{2n}}{(n!)^2}. \quad (19)$$

From the molecular dynamics, Z depends only on the fields through the ratio $\epsilon_{-1}(0, t)/\epsilon_0(0, t)$, which is constant if $\epsilon_{-1}(0, t)$ is initially generated via a two-wave exponential process. It can also be shown that Z approaches, in a distance short compared to the Rayleigh length (depth of focus) in the experiment, a value independent of the path length. This critical distance is just ω_0/ω_R times the distance required for

a steady state small signal Raman gain of 1 at pump intensity I_0 .

As a result of coupling the molecular response and the generated Raman waves, we find that

- AS_n output energy scales linearly with input energy.
- The conversion efficiency to any order is "independent" of the cell length.

This last theoretical result reinforces our experimental observation that the process is insensitive to the f -number of the input optical system.

We have outlined only some of the features of the theoretical model. We are presently completing an analytic solution to the case of zero optical dispersion; the results will aid us in understanding high-order nonlinear Raman processes while providing important scaling laws requisite for future applications.

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Photon-Atom Dynamics

Coherent Deflection of Atoms in a Standing Light Wave

Deflection of a beam of neutral atoms by a resonant electromagnetic wave, or by a combination of resonant and static fields, has been a subject of interest since the advent of higher-power tunable lasers.³⁵

When a strong, resonant electromagnetic wave irradiates an atomic beam, absorption-emission processes proceed at two distinct rates:

- Photons are absorbed from, and emitted into, the applied fields at the induced transition rate Ω .
- Photons are (occasionally) spontaneously emitted in random directions at the spontaneous rate γ .

Deflection or scattering of the atomic beam results when momentum is transferred from the

field to the atoms, the rate of momentum transfer depending on the nature of the applied field.

If the applied field consists of a single plane wave, momentum is transferred to the atoms at the spontaneous rate γ . This momentum transfer (i.e., radiation pressure) proceeds at the spontaneous rate because absorption followed by induced emission into the same field mode involves no net transfer of momentum. Absorption followed by spontaneous emission, however, transfers an average of one quantum of momentum for each spontaneous event (isotropic spontaneous emission does not carry away the momentum acquired by the atom through absorption).

If the applied field is composed of two or more plane waves, an atom can absorb a photon from one of the plane waves, and, through induced emission, emit the photon into a different plane wave, with a resultant transfer of momentum at the induced rate Ω . Because in a strong applied field Ω may exceed γ by many orders of magnitude, it is expected that deflection processes operating at the induced rate will be more efficient and more rapid than processes operating at the spontaneous rate.

Earlier models³⁶ of the deflection process treated a two-level atom in a classical standing-wave monochromatic field, restricted in three ways:

- The frequency of the field ω matches the Bohr transition frequency of the atom (i.e., exact resonance).

- The incident atomic beam is aligned parallel to the plane-wave phase surfaces.

- The time duration of the atom/field interaction is sufficiently brief so that the transverse kinetic energy of deflection (i.e., energy from momentum transverse to the wave fronts) remains much smaller than the interaction energy $\hbar\omega$.

Within these limits, we find that the field deflects the atoms coherently: during each Rabi-cycle period $1/\Omega$, the atomic transverse momentum increases by $\hbar\omega/c$.

In more recent work³⁷ we have succeeded in relaxing all these restrictions. After outlining the theoretical model enabling us to do this, we illustrate our results for the case in which the deflection process is followed over a long period of time—much longer than the time during which the third restriction would apply. We conclude with a very brief description of our results when the restrictions on initial transverse momentum and field frequency are also lifted.

Theoretical Model. We decompose the standing wave into two oppositely directed travelling waves, whose amplitudes are now quantized. The basis states of the system are defined as products of the two field states $\phi_1(N_1)$ and $\phi_2(N_2)$, containing N_1 and N_2 photons respectively, and of the atom state $\phi_{\pm}(p)$, describing excitation (+ means excited, - means unexcited) and transverse momentum p . For simplicity, we assume that the fields contain equal numbers of photons and that they are oppositely directed, with photon propagation vectors $\vec{k}$ and $-\vec{k}$.

Consider, for example, the time development of a single initial state, ψ_0 , defined by

$$\psi_0 = \phi_1(N_1) \phi_2(N_2) \phi_-(\vec{p}_0). \quad (20)$$

The atom is initially in the ground electronic state and can absorb a photon from either of the two fields. Absorption of a photon from the field $\vec{k}$ reduces the occupation number of that field by one, promotes the atom from the ground state to the excited state, and increases its momentum by $\hbar\vec{k}$. The resulting state of the system is defined by

$$\psi_1 = \phi_1(N_1 - 1) \phi_2(N_2) \phi_+(\vec{p}_0 + \hbar\vec{k}). \quad (21)$$

Similarly, absorption of a photon from the other field changes the state of the system to

$$\psi_{-1} = \phi_1(N_1) \phi_2(N_2 - 1) \phi_+(\vec{p}_0 - \hbar\vec{k}). \quad (22)$$

From the state ψ_1 , the atom can be stimulated to emit a photon into either field. If it emits a photon $\vec{k}$, then the system returns to ψ_0 ; if, however, it emits a photon $-\vec{k}$, the system wave function becomes

$$\psi_2 = \phi_1(N_1 - 1) \phi_2(N_2 + 1) \phi_-(\vec{p}_0 + 2\hbar\vec{k}). \quad (23)$$

Similarly, a new state is generated from ψ_{-1} by the emission of a photon $\vec{k}$, namely

$$\psi_{-2} = \phi_1(N_1 + 1) \phi_2(N_2 - 1) \phi_-(\vec{p}_0 - 2\hbar\vec{k}), \quad (24)$$

and so on.

A general wave function of the system at arbitrary time can be written as a superposition of basis states

$$\psi(t) = \sum C_n(t) \psi_n e^{-i\alpha_n t}, \quad (25)$$

where $C_n(t)$ is the probability amplitude for finding the system in the state n , and α_n is the phase associated with that state.

The general wave function must satisfy the Schrödinger equation, and this places certain restrictions on the amplitudes C_n and phases α_n . Ultimately, the following equation, representing the "equations of motion" of the system, must be satisfied:

$$i \frac{dC_n}{dt} = \left[b(qn + n^2) \pm \frac{\Delta}{2} \right] C_n + \frac{\Omega}{2} (C_{n-1} + C_{n+1}), \quad (26)$$

where state n is even (+) or odd (-). The coefficient of C_n on the right side of the equation is the energy of n in the absence of the field. It has two parts: one related to the change in kinetic energy of the atom with respect to the initial state, and the other related to the difference Δ between the laser frequency and the atomic absorption frequency (in the absence of these two effects, all states have the same energy, since photons are just being passed around between the fields and the atom). Parameters b and q are defined by

$$b \equiv \frac{\hbar k^2}{2M},$$

and

$$q \equiv \frac{p_0}{\hbar k}.$$

Any given state ψ_n is radiatively connected only to its nearest neighbors, $\psi_{n\pm 1}$. The coupling is given by the atom field's interaction energy $\hbar\Omega/2$, which gives rise to the off-diagonal terms in Eq. (26).

When treated as described above, the atom/field system is analogous to a highly excited anharmonic oscillator, or to a sequentially linked N-level atom.³⁸

Application. Equation (26) takes on an analytically soluble form if we can neglect the diagonal terms with respect to the off-diagonal terms, such that:

$$\left| b(qn + n^2) \pm \frac{\Delta}{2} \right| \ll \frac{\Omega}{2}. \quad (27)$$

Under this restriction,

$$C_n(t) = i^n J_n(\Omega t), \quad (28)$$

where $J_n(\Omega t)$ is the Bessel function of order n , whose argument is Ωt .

As previous articles have noted,^{35,36} the population flow of the Bessel function follows a dominantly double-peaked pattern, symmetrically distributed about the initial state $n = 0$. As the process evolves, the population reaches and then passes levels having an increasingly large value of $|n|$. There is never an appreciable population in levels much beyond the leading edge of the probability distribution—levels for which the order of the Bessel function exceeds its argument $n = \Omega t$.

The Bessel-function approximation remains valid as long as we can neglect detuning relative to the Rabi frequency. Even if $\Delta = 0$ and $q = 0$, the population will arrive, in due time, at levels for which bn^2 no longer remains negligible compared with Ω , and approximation (27) then fails.

Although the equations of motion are no longer analytically soluble when bn^2 becomes comparable in magnitude to Ω , they can be solved numerically. Figure 9-31 displays a relief view of the probability distribution $P_n(t)$ as a function of time, obtained by the numerical solution of Eq. (26) for the parameter choices $\Omega = 1$, $b = 0.05$. In Fig. 9-31 we observe two peaks of probability moving away from $n = 0$, in accord with the Bessel-function approximation. Note that we use Ω as our unit of frequency, and Ω^{-1} as our unit of time in the figure.

We have previously shown^{35,36} that first-order atomic deflections are achieved in the Bessel-function regime with a field intensity of 10^6 W/cm², in a time shorter than the natural lifetime of the excited atom. This pattern of probability flow ceases on reaching the turning-point levels, where detuning equals the Rabi frequency:

$$|n_{\max}| = \sqrt{\Omega/b}. \quad (29)$$

For the present example, this formula gives $|n_{\max}| = 4.5$. The time of this failure of the Bessel-function approximation is roughly $t = |n_{\max}|/\Omega \cong 5$. At longer times, the probability distribution undergoes more complicated variations, but it remains almost entirely within the bounds of $\pm |n_{\max}|$, which is the

Fig. 9-31. Distribution of population as a function of time, obtained by the numerical solution of Eq. (26) for parameters $\Omega = 1$ and $b = 0.05$.

maximum number of photons that can be transferred from one field to the other.

In interpreting the time-dependent dynamics displayed in Fig. 9-31, it is useful to think of the probability $P_n(t)$ as a fluid which, following initial release from confinement at $n = 0$, flows as a wave packet toward larger $|n|$, until it rebounds from a soft barrier at $|n_{\max}|$. The probability then sloshes back toward $n = 0$ where it subsequently piles up. The probability wave continues to propagate back

and forth, but the peaks tend to spread out and diminish in amplitude.

Lifting the constraints on initial transverse momentum p_0 and field frequency detuning Δ also yields interesting results. For $0 < p_0 < p_{\max} = \sqrt{2Mk\Omega}$ and short interaction times, probability distribution is again symmetric about the incident atomic direction. The turning points, however, are no longer symmetric about that direction, and the maximum deflection is larger than when $p_0 = 0$. For sufficiently long times, the time-averaged population distribution peaks at both p_0 and $-p_0$; peaking becomes more pronounced as p_0 approaches $p_{\max}$.

When $p_0 = p_{\max}$, the distribution remains sharply peaked about p_0 . After sufficient time, however, the population can "tunnel through" the forbidden region and appear in a sharp peak about $p = -p_0$. This population oscillation is an illustration of what, for the N-level atom, would be termed a multiphoton resonance. The condition for this resonance is the Bragg condition for waves of wavelength equal to the atomic de Broglie wavelength, scattering from a "lattice" of periodicity equal to the optical wavelength.

Frequency-detuning only slightly alters the dynamics, as long as the detuning is much smaller than the Rabi frequency. For larger detunings, dramatic changes occur: the maximum deflection increases and the Bragg scattering occurs on a much shorter time scale.

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N-Level Photon-Atom Dynamics

Previous annual reports^{39,40} have described our work aimed at understanding the mechanisms whereby laser radiation excites and subsequently ionizes an atom. As this work has shown, the dynamics of coherent radiative atomic excitation differ qualitatively from those of systems excited incoherently: the retention of phase memory during coherent excitation leads to oscillatory or fluctuating excited-state populations, whereas incoherently excited populations typically tend monotonically toward equilibrium values. This is behavior appreciated since Einstein's early work. Although much of our effort is directed toward modeling as accurately as possible the precise conditions expected for practical operation of laser isotope separation facilities, the complexity of such models often hampers understanding of the connection between measurable results (e.g., ionized fraction) and experimentally controllable conditions.

To enlarge our understanding of coherent excitation, we have continued to supplement our numerical modeling with studies of a simpler mathematical idealization of coherent excitation, the N-level atom.⁴¹⁻⁴⁴ This simple model neglects all homogeneous relaxation effects (such as spontaneous emission or collisions) and idealizes the field-atom interaction as a linked succession of transitions driven simultaneously by monochromatic light tuned to (or near) the Bohr resonance frequencies. Although practical application to real experimental conditions must include such complications as nonmonochromatic pulsed lasers—and encountering spatially and temporally inhomogeneous flowing densities of absorbing atoms, each of which may possess degenerate hyperfine-split energy levels—the possibility of analytic solutions for the simple N-level atom nevertheless continues to motivate study. During the past year we examined two interesting analytically soluble cases,^{43,44} each of which provides further insight into coherent excitation.

As described in previous articles,^{39,44} our models express the excitation probability $P_n(t)$ of level n at time t (for $1 \leq n \leq N$) as the absolute square of a complex-valued probability amplitude $C_n(t)$ obtained as the solution to the rotating-wave-approximation (RWA) time-dependent Schrödinger equation

$$i \frac{d}{dt} C_n(t) = \sum_m W_{nm} C_m(t), \quad (30)$$

where, for the present cases of sequential excitation of nondegenerate levels, the RWA Hamiltonian matrix W is tridiagonal (and symmetric). Diagonal elements W_{nn} have as their real parts the cumulative detunings Δ_n of $n - 1$ successive lasers away from the corresponding sum of $n - 1$ Bohr transition frequencies, and have as their negative imaginary parts half the probability loss rate γ_n from level n :

$$W_{nn} = \Delta_n - \frac{i}{2} \gamma_n. \quad (31)$$

The nonzero off-diagonal elements of W , those that link adjacent levels, are proportional to the dipole transition moment d_n linking levels n and $n + 1$ and to the electric field amplitude $\mathcal{E}_n$ whose carrier frequency closely matches the Bohr frequency for the $n \rightarrow n + 1$ transition:

$$W_{n,n+1} = W_{n+1,n}^* = -d_n \mathcal{E}_n / \hbar \equiv \frac{1}{2} \Omega_n \quad (32)$$

Thus, the N -level atom comprises a sequence of $N-1$ values for the Rabi frequencies Ω_n and N values for the cumulative detunings Δ_n and loss rates γ_n , each of which, in principle, is adjustable. Although this model is highly idealized as a description of laser excitation, it provides useful insights into details of coherent excitation and into the relationships between physically adjustable parameters and population variations.

The first of our models⁴³ exploited the mathematical equivalence of a spin- J system to an $N = (2J + 1)$ -level sequence in which the Rabi frequency varies according to the formula

$$\Omega_n = \Omega_0 [n(N - n)]^{1/2}, \quad (33)$$

and the cumulative detuning and loss rate increase linearly with n :

$$\Delta_n = \Delta_0 n \quad \gamma_n = \gamma_0 n. \quad (34)$$

One can express the solutions $C_n(t)$ for this model in terms of rotation matrices. It is not difficult to show⁴³ that the eigenvalues λ_n of the RWA Hamiltonian are evenly spaced,

$$\lambda_n = n \left[|\Omega_0|^2 + \left(\Delta_0 - \frac{i}{2} \gamma_0 \right)^2 \right]^{1/2}, \quad (35)$$

so that when $\gamma_0 = 0$ the system is exactly periodic. (Although general two- and three-level systems are periodic for any choice of Ω_n and Δ_n , lack of commensurable eigenvalues generally prevents periodicity for larger N .⁴⁰⁻⁴²)

Fig. 9-32. Effect on populations $P_n(t)$ for an $N = 9$ level spin- J model of varying initial conditions. Successive frames show time evolution of system prepared, respectively, in levels 1, 2, 3, 4, and 5. Labels identify curves for specific levels n .

Unlike other analytically soluble resonant cases ($\Delta_n = 0$) of the N -level atom (for $N > 3$), the choice of Rabi frequencies [Eq. (33)] leads to complete population inversion (in the time interval $\tau = \pi/\Omega_0$, independent of the number of levels). This combination of periodicity and complete inversion leads to an N -level "area theorem": if all the Rabi frequencies share a common arbitrary time dependence $\Omega_n(t) = f(t)\Omega_n$, then complete inversion occurs for times such that the time integral of $f(t)$ is an odd-

Fig. 9-33. (a) N -level vector model shows initial population distribution as a cone in abstract space; for population in level n , cone height along Z axis is $M = n - 1/2(N + 1)$ and cone side length is $[J(J + 1)]^{1/2} = 1/2(N^2 - 1)^{1/2}$. Oriented as shown here, cone represents population inversion. As time progresses, cone precesses about an axis Z'' whose Euler angles are $\alpha = \arctan \text{Im}(\Omega_0)/\text{Re}(\Omega_0)$ and $\beta = \arctan [|\Omega_0/(\Delta_0 - (i/2)\gamma_0)|]$. (b) Precession when there is no detuning or loss, so that Z'' coincides with X axis. During one cycle, population moves from initial level n_0 into level $N - n_0 + 1$.

integer multiple of π . Interestingly, if population begins in level n_0 , then a π -area pulse places all the population into level $(N - n_0 + 1)$ (see Fig. 9-32). No other levels ever contain all the population at one instant of time. We have devised a simple N -level "vector model" portrait of the probability flow, a generalization of the oft-used two-level vector model of Feynman, Vernon, and Hellwarth (see Fig. 9-33).

The linear-in- n behavior of the cumulative detuning [1:q. (34)] befits Doppler-shift detuning and thus allows an analytic solution in this important case. The analogy with a spin- J system puts no restriction on spacing of atomic energy levels: only the detuning enters the equation.

Our second soluble model occurs in considerations of limiting cases. Although solutions to the loss-free Schrödinger equation continue varying with time indefinitely, the time-averaged probabilities

$$\bar{P}_n(\tau) = \frac{1}{\tau} \int_0^\tau dt |c_n(t)|^2 \quad (36)$$

approach asymptotic limits within a few population oscillations⁴⁰; thus, the limiting value $\bar{P}_n \equiv P_n(\infty)$ provides a useful indicator of average population. The simplicity of computing $\bar{P}_n$ from eigenvalues and eigenvectors of the RWA Hamiltonian^{41,42} makes this infinite-time average a particularly useful monitor of population dynamics: in the study of laser-induced chemical reactions at low densities, the values $\bar{P}_n$ measure the availability of excited population for reactions. Thus it is useful to understand the conditions on the Rabi frequencies and detunings that maximize the value of $\bar{P}_n$ for some level (or levels) n .

For simplicity we assume that population initially resides in the first of the N levels. Previously we noted⁴¹ that if all Rabi frequencies are equal, the population averages of an N -level system tend toward the rate-equation equilibrium value of $\bar{P}_n = 1/N$, and that if Rabi frequencies increase with n , then $\bar{P}_n$ decreases with n (although significant level-to-level variations may occur). We observed that sequences placing the largest Rabi frequencies in the middle levels tend to concentrate populations in the end levels—as though population tended to avoid large Rabi frequencies. This observation suggests a strategy for placing maximum population into the

Fig. 9-34. Examples of N -level behavior when first and last Rabi frequency are much smaller than remaining Rabi frequencies: $\Omega_1 = \Omega_{N-1} = 0.02 \Omega_n$, with $\Sigma \Omega_i^2 = 1/t_0$. Frames show $N = 4, 5, 10$, and 11. Population flows periodically between levels $n = 1$ and $n = N$ (see Ref. 44 for analysis of these curves).

most excited state; namely, to employ a sequence of Rabi frequencies in which the first and last steps have the smallest Rabi frequencies. We found that, as these Rabi frequencies became much smaller than the remaining Rabi frequencies, the system tends toward very simple periodic behavior in which $\bar{P}_n$ is concentrated almost entirely in the two equally populous levels $n = 1$ and $n = N$. Interestingly, we discovered a striking difference between even- N and odd- N systems: for even N , the limiting system is equivalent to an effective two-level atom whereas an odd- N system approaches the behavior of a three-level atom (see Fig. 9-34). Using matrix partitioning techniques we developed an analytic description of these limiting examples.⁴⁴

The analytical results of these two soluble models have extended our understanding of coherent excitation and of the way in which adjustments in laser power and frequency alter this excitation. We are now attempting to generalize our ob-

servations to more general sequences of detunings and Rabi frequencies.

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Collision Physics and Chemistry

Crossed Molecular Beams Studies of Gas-Phase Uranium Atom Reactions

In last year's report on the gas-phase chemistry of uranium,⁴⁵ we observed that uranium exhibits a wide range of reactivity (<1 to 35 Å^2) with a variety of small inorganic and organic oxidizers and halocarbons.⁴⁶ Uranium in the gas phase typically behaves like other heavy metals, such as barium and tin, which have been extensively studied.

During the past year, we reexamined a number of uranium-atom reactions in which more than one reaction product is thermodynamically allowed; in several cases (reactions with SO_2 , H_2O , and low-chain-length alcohols) we observed more than one product. We also investigated the reactions of uranium with other halocarbons and halogen acids. Finally, we refined our analysis of the laboratory angular distributions of UO from reactions of U with O_2 , NO, SO_2 , NO_2 , $(\text{CH}_3)_2\text{CO}$, H_2O , CH_3OH , and $\text{C}_2\text{H}_5\text{OH}$ to understand the dynamics of these reactions better and to improve our estimates of the corresponding total reaction cross sections.

Halogenated Methanes and Halogen Acids. Last year we described the reactions of uranium atoms with a set of halogenated methanes (CCl_4 , CH_3I , CF_3Br , CF_3Cl , and CF_4); we observed that reactivity decreased in the order I, Br, Cl, F. During the past year we investigated the reactions of uranium with CF_3I , CH_3Cl , SF_6 , HF, and HCl, again using crossed molecular beam scattering in the molecular beam facility (see last year's report⁴⁵ for a detailed description of the apparatus and technique). A complete list of all 10 halogen-transfer reactions studied to date, with corresponding relative cross sections (estimated from ratios of integrated laboratory fluxes) measured at thermal energy, is provided in Table 9-8. For these reactions we crossed a 2550-K uranium-atom beam with a 350-K reagent beam; the most probable incident collision energies are in the range 2 to 3 kcal/mole.

Large uranium halide signals are observed from the $\text{U} + \text{HCl}$ and $\text{U} + \text{HF}$ reactions (numbers 7 and 8 in Table 9-8). Reaction 8 is, in fact, the only reaction observed to date to produce significant UF product. (Some UF is observed in reaction 9; however, the signal is just above the background noise level, and we assign only an upper limit to the relative cross section.) The UX-product angular dis-

Table 9-8. Relative cross sections for reactions of uranium with halogenated methanes and halogen halides.

Reaction	Relative cross section ^a
(1) $\text{U} + \text{CH}_3\text{I} \rightarrow \text{UI} + \text{CH}_3$	1.0
(2) $\text{U} + \text{CF}_3\text{I} \rightarrow \text{UI} + \text{CF}_3$	0.5
(3) $\text{U} + \text{CF}_3\text{Cl} \rightarrow \text{UCI} + \text{CF}_3$	<0.03
(4) $\text{U} + \text{CH}_3\text{Cl} \rightarrow \text{UCI} + \text{CH}_3$	0.2
(5) $\text{U} + \text{CF}_3\text{Br} \rightarrow \text{UF} + \text{CF}_3$	0.2
(6) $\text{U} + \text{CCl}_4 \rightarrow \text{UCI} + \text{CCl}_3$	0.5
(7) $\text{U} + \text{HCl} \rightarrow \text{UCI} + \text{H}$	0.8
(8) $\text{U} + \text{HF} \rightarrow \text{UF} + \text{H}$	1.2
(9) $\text{U} + \text{SF}_6 \rightarrow \text{UF} + \text{SF}_5$	<0.06
(10) $\text{U} + \text{CF}_4 \rightarrow \text{UF} + \text{CF}_3$	NR ^b

^aEstimated from ratios of integrated laboratory fluxes.

^bNR = no reaction product observed.

tributions from the HCl and HF reactions are peaked at, and are only 10 to 20% wider than, the distribution of centroids (incident center of mass-velocity vectors). This is not unexpected, since the very light H atoms (the other product) cannot carry off a significant amount of linear momentum. Hence, in this case, the laboratory angular distributions of UX product are determined mainly by kinematic considerations and reveal little about the reaction dynamics of these reactions.

Comparison of the reactivity of the halogenated methanes reveals two significant trends:

- The reactivity of uranium with both the CF_3X and the CH_3X series of reagents decreases in the order I, Br, Cl. In all cases only the weakest C-X bond is broken, as we never detect any UI or UI.

- We observe that for $\text{X} = \text{Cl}$ and $\text{X} = \text{I}$, CF_3X is considerably less reactive than the corresponding CH_3X .

Reaction Products from $\text{U} + \text{SO}_2$, $\text{U} + \text{NO}_2$. Laboratory angular distributions of UO formed in the reactions of uranium with O_2 , NO, SO_2 , and NO_2 have been previously presented.^{45,46} In the first two reactions, UO is the only possible neutral product (UN from $\text{U} + \text{NO}$ is thermodynamically forbidden by 23 kcal/mole). However, for $\text{U} + \text{SO}_2$ and $\text{U} + \text{NO}_2$, neutral products other than UO are thermodynamically allowed (see Table 9-9). In both cases the most exothermic channel is the one yielding UO_2 , a reflection of the large U-O bond energy. Exothermicities cannot be assigned to the reactions producing UXO ($\text{X} = \text{S}$ or N), since these triatomics have not been characterized; it is likely, however, that both products are thermodynamically

Table 9-9. Relative product fluxes for various reaction channels in $U + SO_2$ and $U + NO_2$ systems.

Possible channels, $U + SO_2$	$\Delta H(\text{kcal/mole})$	Possible channels, $U + NO_2$	$\Delta H(\text{kcal/mole})$
$U + SO_2 \rightarrow UO + SO$	-50	$U + NO_2 \rightarrow UO + NO$	-109
$U + SO_2 \rightarrow UO_2 + S$	-97	$U + NO_2 \rightarrow UO_2 + N$	-130
$U + SO_2 \rightarrow US + O_2$	+13	$U + NO_2 \rightarrow UN + O_2$	-22
$U + SO_2 \rightarrow USO + O$	?	$U + NO_2 \rightarrow UNO + O$	?

Relative product fluxes at 10° LAB		
UO	UO_2 and US	USO
100	82.6	5.7

Relative product fluxes at 10° LAB			
UO	UO_2	UN	UNG
100	1.0	1.0	0.7

allowed. Measurements of relative cross sections for each reaction channel, i.e., the chemical branching ratios, provide sensitive tests of theories of chemical reaction dynamics.

The reaction products from $U + SO_2$ and $U + NO_2$ were identified using crossed molecular beam scattering. An effusive source located in the main chamber 6 mm from the scattering center served as the gas source, providing high-intensity gas beams at the cost of angular resolution. The detector (an electron-bombardment quadrupole mass spectrometer) was positioned at $\theta_{LAB} = 10^\circ$, close to the most probable center of mass of the colliding pair. The resolution of the quadrupole mass spectrometer was adjusted to be constant in the mass range of interest.

Because we intersect two thermal beams, the distribution of relative collision energies is broad. The most probable collision energy depends on the mass of the collision partner; for both SO_2 and NO_2 , the distribution peaks between 2 and 3 kcal/mole. These energies are quite small compared to the reaction exothermicity.

For SO_2 , we observed relative product signals at UO, UO_2 /US, and USO masses, in the ratio of 100:83:6; this indicates three reaction channels—two major and one minor—in the $U + SO_2$ reaction. Since both UO_2 and US have the same molecular mass (270 amu), further experiments were required to distinguish between these products. In this case, we exploited the different distribution of atomic S and O isotopes. Based on the abundance of ^{34}S , if US were the only product at mass 270, then the ratio of product signals at masses 272 and 270 should be approximately 0.044. If, however, the reaction produces UO_2 instead, then we should expect, based on the isotopic distribution for ^{16}O , ^{17}O , and ^{18}O , a much smaller ratio of

0.0041. We actually measured $\text{Signal}(272 \text{ amu}) / \text{Signal}(270 \text{ amu}) = 0.0064 (\pm 0.0016)$, indicating that the product at mass 270 amu is predominantly 94% ($\pm 4\%$) UO_2 . Essentially no US is formed. Since the US channel is predicted to be modestly endothermic, these observations confirm our prior expectations.

Angular distributions of both UO and UO_2 from $U + SO_2$ are shown in Fig. 9-35. At the low electron energies (10 to 15 eV) typically used for ionization in our experiments, U^+ , UO^+ , and UO_2^+ ions arise predominantly from direct ionization of the corresponding neutral.⁴⁷ As a check, we increased the ionizer energy to 30 eV and observed that both the UO and UO_2 signals increased. If there were significant impact-dissociation of UO_2 in the ionizer producing UO^+ , we should have seen a decrease in the UO_2 signal. Consequently we conclude that the UO^+ and UO_2^+ ion signals are good monitors of the $U + SO_2 \rightarrow UO$ and $U + SO_2 \rightarrow UO_2$ reactions, respectively.

At angles near $\theta_{LAB} = 0^\circ$ (the straight-through uranium beam), some of the UO product may arise from elastic scattering of the small ($\geq 0.5\%$) UO contaminant in the U oven, rather than from the $U + SO_2$ reaction. To estimate the elastic component, we scale the observed uranium-atom scattering by the measured ratio of UO/U in the beam. The UO curve in Fig. 9-35 shows the corrected UO scattering. Adjustments to the UO signal are required only at $\theta_{LAB} = \pm 5^\circ$ and do not change the overall shape of the curve.

Both angular distributions shown in the figure are considerably broader than the distribution of centroids (centers of mass of the colliding pairs); the FWHM of the UO_2 product distribution is approximately 30% wider than that of the UO distribution. Since the fragment released in forming

Fig. 9-35. Laboratory angular distributions for UO and UO₂ produced in the reaction of U and SO₂. Points indicate experimental data; dashed line indicates calculated distribution of centroids for collision pairs.

UO₂ is lighter than that released in forming UO, this result implies that the absolute amount of energy released into product translation is larger for UO₂ than for UO. The UO₂ distribution peaks near the most probable centroid (distribution of centroids is shown by the lightly dashed curve), which suggests a long-lived collision complex. Such a mechanism might be expected, as it would allow sufficient time for both S-O bonds to be broken and for two U-O bonds to be formed. The UO is slightly forward-peaked—confirming our previous observations^{45,46}—and indicates a direct interaction.

We next looked for similar chemical branching in the U + NO₂ system (see Table 9-9 for possible

reaction products). Product signals on the order of only 1% of the UO signal were observed at UN and UO₂ masses. Thus, since UO is the only significant neutral product of the U + NO₂ reaction, we conclude that this system differs dramatically from the U + SO₂ system.

The striking difference in UO:UO₂ product ratios from SO₂ and NO₂ is not explained by thermodynamics; nor can the differences be explained by simple geometric considerations. Consider the space-filling models of U, NO₂, and SO₂ shown in

Fig. 9-36. Space-filling models of U, NO₂, and SO₂.

Fig. 9-36; here each atom is bounded by its van der Waals radius. Using a hard-sphere model for the interatomic potentials, one can calculate the fraction of uranium-atom collisions that strike the various atoms in the triatomic reagents; results are shown in Table 9-10. Note that the hard-sphere total cross sections for SO₂ and NO₂ are close to equal. These model results do not show any feature that would explain the absence of UO₂ product from NO₂. For example, a hypothesis that collisions with O lead to stripping (UO product), while collisions with S or N are required for UO₂ formation, would still predict significant UO₂ product from both reagents.

Attempts to correlate this striking pattern of reactivity with the electronic structure of the triatomic reagent are already underway.

Table 9-10. Analysis of U + NO₂ and U + SO₂ hard-sphere collisions.

Parameter	NO ₂	SO ₂
Total cross section, Å ²	62.9	66.9
Collisions with both O atoms, %	81.7	68.9
Collisions with central atom, %	18.3	32.0

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Reaction of Vibrationally Excited HBr and I

The effects of the vibrational energy of reactants on the reaction rate have been treated theoretically by Polanyi.⁴⁸ He and coworkers later carried out classical trajectory calculations for reactions $HX + Y \rightarrow HY + X$ where X and Y are halogen atoms.⁴⁹⁻⁵¹ Johnson and Winter studied the $O + H_2 \rightarrow OH + H$ reaction in a similar fashion.⁵² For these endoergic reactions the vibrational energy of the reactants significantly accelerated the rate of reaction. Birely and Lyman tried to systematize the effect of vibrational energy but found no correlations.⁵³ Arnoldi et al. demonstrated experimentally that vibrational excitation can dramatically accelerate the reaction between HCl and Br, and were able to observe isotopic enrichment of ³⁵Cl with this technique.⁵⁴ Badcock et al. studied the reaction of HBr with I using sequential laser excitation of the HBr molecules, and observed bromine isotopic enrichment after selective excitation.⁵⁵

The primary reaction is

where $n = 0, 1, 2, 3, 4$; the bromine atoms are scavenged by undissociated I₂ in the reaction

Fig. 9-37. Contour plot of LEPS empirical surface fitted to ab initio calculations of potential energy surface; X indicates saddle point.

Badcock et al. estimated that for the primary reaction, $v = 2$ and $v = 3$ accounted for the majority of the products; they also predicted a rate enhancement of about 10^9 (Ref. 55).

To study this reaction theoretically we have used accurate configuration interaction calculations to determine a potential energy surface,^{56,57} which we then fit to a London-Eyring-Polanyi-Sato (LEPS) empirical surface (shown in Fig. 9-37). The ab initio calculations of the potential surface predict

a reaction barrier of 20.4 kcal/mole. The position of the barrier is $R_{\text{HI}} = 1.79 \text{ \AA}$ and $R_{\text{HBr}} = 1.72 \text{ \AA}$ for a collinear saddle point. The fitted LEPS surface gave Sato parameters of -0.08, 0.20, and 0.0.

Using this surface we carried out classical trajectory calculations with HBr initially in vibrational levels $v = 0, 1, 2, 3$ or 4, using a 300-K Boltzmann distribution over the rotational states. The cross section for producing HI + Br was obtained at collision energies of 1, 2, 4, 6, 8, and 10 kcal/mole; 2000

Fig. 9-38. Calculated cross sections for reaction $\text{HBr}(v) + \text{I} = \text{HI} + \text{Br}$, with initial vibration state $v = 2, 3$, and 4.

trajectories were calculated at each energy. Confirming the experimental findings of Badcock et al.,⁵⁵ we did not observe any reaction until $v = 2$. For $\text{HBr}(v = 2)$ the reaction is still 1.84 kcal/mole endoergic, while $\text{HBr}(v = 3)$ and $\text{HBr}(v = 4)$ are exoergic by 4.96 and 11.51 kcal/mole, respectively.

The calculated reactive cross sections for each of these three initial vibrational states are shown in Fig. 9-38. The reactions for $v = 3$ and $v = 4$ are not sensitive to collision energy, indicating that the internal reagent energy is readily available to overcome the reaction barrier. The cross section for $v = 2$ is the most sensitive of the three, since only a small amount of translational energy is required for reaction. The considerable scatter of the $v = 2$ data results from the small number of reactive trajectories. The higher vibrational levels appear to be well behaved.

At this time the low-energy (< 1 kcal/mole) regions of the cross sections have not been adequately determined, which has prevented us from calculating the rate constants for all initial vibrational levels. To integrate the full cross section (and hence produce the rates), we must estimate the

low-energy cross sections from the long-range attractive forces between HBr and I ; this work is now in progress.

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Alternate Laser Applications: H_2S Cleanup in Fuel Feed Streams by Laser Photolysis

During the past year we performed a series of experiments to examine the scientific feasibility and cost-effectiveness of purifying synthesis gas with a photochemical technique employing efficient ultraviolet lasers. Synthesis gas (mainly H_2 and CO), an important chemical feedstock for the synthesis of methanol, methane, glycols, and other hydrocarbons, often contains trace amounts of sulfur compounds (mainly H_2S with some COS and CS_2). Such impurities are extremely (and undesirably) reactive with the metallic catalysts used in the above chemical syntheses; in fact, sulfur "poisoning" of these catalysts often leads to rapid degradation of catalytic reactivity, and a shorter catalytic lifetime.⁵⁸

For certain catalysts, such as the copper-based catalysts used to synthesize low-pressure methanol from synthesis gas, the H_2S impurity level must be kept at a level of 0.5 ppm or lower to ensure a

Fig. 9-39. Experimental arrangement for measuring UV absorption cross sections of synthesis gas.

catalytic lifetime of one year.⁵⁹ Synthesis gas purity of less than 1 ppm is commonly achieved by an effective but rather expensive chemical process using ZnO. The laser purification technique described here could be used as a direct substitute for this ZnO process, while retaining the conventional precursor scrubbing method for the removal of the bulk of H₂S and other acid gases in the synthesis gas stream.⁶⁰

The laser scheme is based on the photodissociation of H₂S (Refs. 61-63) by either an RGH or a Raman-shifted RGH laser, in the reaction

In the presence of a metal or a metal-oxide surface placed in the flow stream, the HS fragment could undergo surface reactions, leaving the sulfur atoms either bound as a metal sulfide:

or deposited as solid polymeric sulfur:

where M is a metallic surface or surface-adsorbed species.

Feasibility. There are two scientific issues pertaining to the economics of this purification process.

- The central spectroscopic issue is that of selectivity: the degree of unwanted laser absorption by synthesis gas (particularly CO) and by spectator impurities (such as C₂H₄, CH₃OH, O₂) whose removal is not beneficial to extending catalyst lifetimes. These impurities are either intrinsic to the actual gasification process or are introduced during the prescrubbing of H₂S (to less than 10 ppm) by conventional sulfur-removal methods.

- The other issue, of equal importance, is the quantum yield of H₂S removal: demonstrating efficient conversion of the HS fragments to metal sulfide or polymeric sulfur.

Figure 9-39 is a schematic of the experimental arrangement used to determine whether or not H₂S can be selectively dissociated with a UV laser while in the presence of synthesis gas. We measured absolute absorption cross sections for the synthesis gas from 190 to 250 nm, using ArF and KrF lasers and a Raman-shifted Rhodamine 6G dye laser with H₂ as the Raman medium. Figure 9-40 shows the absorption cross sections of H₂S (Ref. 61) and synthesis gas (free of H₂S) measured at various laser wavelengths. The laser fluences are roughly 0.1

J cm^2 or lower. The synthesis gas used here is a special makeup mixture of research-grade H_2 , CO , CH_4 , C_2H_4 , CO_2 , and O_2 gases, from Matheson; its composition was similar to that of an actual synthesis gas from a coal gasifier. For comparison, absorption cross sections are also plotted for molecular CO near the region of 180 to 260 nm. The cross section ratio ($\sigma_{\text{H}_2\text{S}}/\sigma_{\text{syn}}$) was found to be roughly 10^7 in the region of 210 to 220 nm; absorption by synthesis gas at 193 nm is primarily due to the Cameron band of CO .⁶⁴

Our UV absorption data suggest, therefore, that a spectroscopic selectivity of 10^7 is feasible if one uses a laser from 210 to 220 nm. The best candidate appears to be an H_2 -Stokes-shifted ArF laser⁶⁵ at 210 nm. Other laser systems, such as an anti-Stokes Raman-shifted KrF laser at 225 nm or a KrCl laser at 220 nm,⁶⁶ would also serve the purpose. One might also selectively dissociate H_2S using a tunable, line-narrowed ArF laser⁶⁵ injection-locked to a region between CO absorption lines. It is not clear, however, that such a system would yield similarly feasible spectroscopic selectivity.

Fig. 9-40: UV absorption cross sections of H_2S and synthesis gas; absorption cross sections for molecular CO near 180 to 260 nm are also plotted. For simplicity, rotational structures and width of each band are not shown here.

To determine the quantum yield of H_2S removal, we have performed photolysis experiments on H_2S at various synthesis-gas densities. The objective of these experiments is to determine the ratio

$$\frac{[\text{H}_2\text{S}] \text{ removed from synthesis gas}}{[h\nu] \text{ absorbed by H}_2\text{S molecules}}$$

Figure 9-41 is a schematic of the photolysis experiment using an ArF laser. The concentration of the H_2S in the gas is determined by monitoring the attenuation of the 193-nm radiation before and after photolysis. We used clean stainless steel or copper tubing for collection or polymerization of sulfur; these results are presented in Fig. 9-42. Note that the quantum yield of H_2S removal is near unity at low pressure (~ 10 Torr) and decreases to roughly 0.4 at 1 atm. The partial pressure of H_2S for these experiments was kept at 2 mTorr. We were able to improve the quantum yield slightly by using tubes of smaller diameter.

Little is known about the reactivity of HS radicals with metal surfaces. Our low-density photolysis data seem to indicate that surface reactions (38) and (39) above are quite fast. At low synthesis-gas density, most of the HS radicals initially produced by the photolysis pulse were transformed into metal sulfide or polymeric sulfur, and hence removed from the gas stream. Additional HS radicals might also have been generated by reactions⁶⁷ between H_2S and H atoms on the collector surface:

This is consistent with our experimental findings that the measured quantum yield of H_2S removal is near unity under low-density conditions. The smaller quantum yield observed at higher synthesis-gas density may result from the recombination reaction of HS radicals prior to their polymerization on the metal surface:

Rate constants reported for reaction (40) have given the result $k = 3 \times 10^{-11} \text{ cm}^3/\text{molecule} \times \text{s}$ (Refs. 67, 68). Based on this value, we can make a

Fig. 9-41. Experimental apparatus for measuring quantum yield of H_2S removal.

rough estimate that under our experimental conditions, with pressure near 1 atm, a significant fraction of the HS radicals can recombine in the gas stream; this reduces the overall quantum yield of H_2S removal to ~ 0.5 , in good agreement with our experimental findings.

Cost Analysis. Based on the spectroscopic selectivity and quantum yield for sulfur removal, we were able to make a more realistic cost analysis for an H_2S cleanup process, using a Stokes-shifted ArF laser at 210 nm, with H_2 as the scattering medium. The estimated laser purification costs for various H_2S concentrations are listed in Table 9-11.

Clearly, the projected purification cost is a function of many parameters such as spectroscopic selectivity, quantum efficiency for H_2S removal, laser fluence, saturation factors, laser efficiency, Raman conversion efficiency, laser-equipment capital, replacement and repair costs, and photon utilization efficiency. Values for these parameters are given along with comparison costs for a conventional chemical method⁵⁹ using ZnO at 623 K. Note that laser purification appears to be uneconomic for an initial H_2S level much greater than 1 ppm. At an impurity level of less than 1 ppm, however, the laser process becomes more economical than the current ZnO process.

Fig. 9-42. Quantum yield of H_2S removal at various synthesis gas pressures. Quantum yield = ratio of number of H_2S molecules removed from gas per number of laser photons absorbed by gas.

For the synthesis of methanol ($\text{CO} + 2\text{H}_2 \rightarrow \text{CH}_3\text{OH}$), the cost increase due to the addition of the laser purification process will amount to roughly 1% of the current methanol chemical value (\$175/tonne). The cost for the laser process results mainly from capital and maintenance charges of the

Table 9-11. Comparison of synthetic gas purification costs: laser photolysis vs ZnO.

H ₂ S impurity levels	Laser purification (\$/mole of S removed) ^a	ZnO purification (\$/mole of S removed)		
		Bulk gas heating cost ^b	ZnO material cost	ZnO unit capital charges ^c
10 ppm → 1 ppm	<10	2	0.5	d
1 ppm → 0.1 ppm	10-20	20	3	20
0.1 ppm → < 0.05 ppm	<100	200	~15	NA ^d

^aCalculated for photon cost = \$1/mole (laser capital and maintenance); laser photon flux = 0.5 J/cm²; quantum yield of H₂S removal = 0.5; Raman shifting efficiency = 0.5; photon utilization efficiency = 0.5 (for 10 ppm, $\ell = 10$ m), 0.35 (1 ppm, $\ell = 10^2$ m), 0.04 (0.1 ppm, $\ell = 10^2$ m).

^bHeat provided by synthesis gas at 7 mil/kWhr, negligible if using waste heat.

^c25% of process unit investment.

^dData not available.

laser system. On the other hand, the cost for the chemical method using a ZnO guard bed is due mostly to capital charges for the ZnO reactor and operating charges for bulk gas heating (required for promoting the ZnO/H₂S reaction at the surface). The cost of bulk gas heating becomes negligible if one uses waste heat.

Conclusion. The laser purification technique presented here has the promise of leading to a process economically competitive with conventional ultrapurification methods. An optimum photolysis selectivity of near unity can be attained using a UV source at 210 to 220 nm; the proposed laser systems (Raman-shifted ArF or KrF lasers) have previously demonstrated over 1% electrical efficiency, which is an order of magnitude higher than that of a conventional low-power UV lamp (such as a Zn lamp). Laser purification is a potentially practical approach for achieving sulfur impurity levels <0.1 ppm. This nearly complete cleanup of the catalytic poison H₂S from synthesis-gas stream could greatly extend the lifetimes of copper-based low-pressure catalysts currently used in the synthesis-gas manufacture of methanol and other chemical feedstocks.

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