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RARE GAS HALIDE LASERS FOR FUSION

FINAL TECHNICAL REPORT

For the Period

1 March 1979 - 29 February 1980

C. Duzy, J. Hsia, H. Hyman, J. Jacob, D. Klimek, J. Parks and D. Trainor
April 1980

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RARE GAS HALIDE LASERS FOR FUSION

Final Technical Report
for the period 1 March 1979 - 29 February 1980

C. Duzy, J. Hsia, H. Hyman, J. Jacob, D. Klimek,
J. Parks and D. Trainor

AVCO EVERETT RESEARCH LABORATORY, INC.
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April 1980

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FOREWORD

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ABSTRACT

In an effort to reliably extend our understanding of KrF* lasers, we collected specific, detailed experimental data and carried out theoretical calculations to explore and document those issues we identified as likely to be important in the short pulse operating regime. These included the effects of: (1) fuel burnup, (2) electron quenching (3) gain/absorption at high current densities, (4) photoionization, (5) accessibility of the lying levels of KrF* by laser flux, (6) temperature on fluorescence efficiency.

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I. INTRODUCTION

Several of the national laboratories and a growing number of industrial research laboratories, supported by the Department of Energy, have been investigating the suitability of lasers for use in inertial confinement fusion, ICF, applications. Largely as a result of pellet modeling calculations and comparisons to new experimental results, various characteristics of the sought after laser have emerged and are summarized in Table 1.

A leading contender for this mission is the KrF* laser which operates at 249 nm. This rare gas halide exciplex has been researched extensively, since it was first lased⁽¹⁾ at AERL in 1975. These studies have shown it to be efficient and scalable. Unfortunately, it is not intrinsically capable of producing the short pulses needed for this application. In the long pulse length regime ($\sim 1 \mu s$), it is very efficient ($\eta_{int} \sim 10\%$) and many of the relevant kinetic issues and scaling constraints have been explored and quantified.⁽²⁾ At issue, for this application, is at how short a pulse length can one operate a high energy, long gain length, KrF* laser without significant loss in intrinsic efficiency. Once the optimal pulse length is established, the laser can be coupled to pulse shortening techniques,⁽³⁾ such as Raman pulse compression and/or angular multiplexing, to achieve the very short pulses needed for efficiently coupling the photon energy into the pellet. It was the purpose of this contract to address the kinetic constraints of this scaling exercise, i.e., to answer the question, what are the relevant reaction mechanisms and chemical processes that govern short pulse, efficient KrF* laser operation?

Once these features were identified, we subsequently measured the appropriate rate constants, cross sections, etc. which allow reliable, quantitative model projections to be made for the short pulse, high energy inertial confinement fusion application.

The results in our research efforts to date are summarized in this report.

TABLE I
DOE ICF LASER REQUIREMENTS

● Energy	1 - 5 MJ
● Pulse Length	~ 10 ns
● Efficiency	> 1%
● Wavelength	$0.2 \mu < \lambda < 3 \mu$
● Repetition Rate	1 - 10 Hz

II. BACKGROUND

Avco Everett Research Laboratory, Inc. (AERL) has been very active in the research and development of numerous rare gas halide lasers including KrF*. As a result of the earlier DARPA-supported research at AERL, the dominant formation and quenching kinetics were well documented.^(4,5) The stimulated cross section was measured and the dominant absorbers in the active medium were identified.⁽⁶⁾ This information, when combined with the results from other laboratories, provided sufficient data to permit accurate modeling of long pulse length operation for KrF* lasers.⁽⁷⁾ AERL constructed a comprehensive code to predict laser performance over a wide range of operating conditions. The predictions of this DARPA-supported code are presented in a plot of intrinsic efficiency as a function of e-beam current density in Figure 1. A description of the work⁽⁷⁾ that lead to these results is included in Appendix A. In plotting these curves, $J_{eb} \tau_p$ has been kept constant at 7.5×10^{-6} Coulombs/cm²; i.e., the pump pulse energy into the laser mixture has been held constant. For this pump energy, half of the F₂ was predicted to be burned up by the end of the pulse. (Later in Section IV, it will be pointed out that this code underestimated the F₂ dissociation. Several other code features have also altered as a result of this project.)

Similar modeling efforts were carried out by AERL under IRAD funding for the ArF* laser candidate. Our calculation showed that ArF* could be significantly more efficient than KrF*. Since any detailed understanding of KrF* laser kinetics contains as a subset the kinetics of argon/fluorine mixtures, we elected to provide as much information about ArF* as was feasible as part of this limited program.

In an effort to reliably extend our understanding of KrF* lasers to the short pulse operating regime (indicated by the shaded region in Figure 1), we collected specific detailed, experimental kinetic data to explore and document those issues we identified as likely to be important in this regime. These included the effects of:

- 1) Fuel burnup
- 2) Electron quenching
- 3) Gain/absorption at high current densities

Figure 1 Scaling Map for the KrF* Laser

- 4) Photoionization
- 5) Accessibility of the higher lying levels of KrF^* by laser flux
- 6) Temperature on fluorescence efficiency

All of these topics have been individually studied as part of this program and the details of each task are presented in Section IV. In the following section, the results of this program are utilized to present new kinetic code projections for the short pulse operating regime. These kinetic codes were thoroughly tested by requiring that they effectively model the experimentally measured fuel burnup, gain/absorption and fluorescence efficiency. By fitting a wide variety of data, collected on a number of different apparatus, by different researchers, a consistent and reliable rate package evolved. This collection of rate constants and cross sections includes those measured or calculated as part of this program and represents our best understanding of this laser system to date.

The exceptions to a complete understanding include the issue of accessibility of the higher lying vibrational levels of KrF^* by the laser flux and the influence of temperature on laser performance (see Section V).

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III. SCALING PROJECTIONS

In this section, we present the predictions of the kinetic rate package with regard to the intrinsic efficiency of the KrF* laser as a function of pulse length (see Figure 2). As before, in plotting these curves, $J_{\text{eb}, \text{tp}}$ has remained fixed at 7.5×10^{-6} Coulombs/cm² and the optimum output coupling has been adjusted throughout.

A comparison of these predictions to those previously published⁽⁷⁾ is provided in Figure 3 for a nominal length of one meter. Notice the predicted decreased efficiency is essentially constrained to the short pulselengths for the particular mixture considered. The predicted efficiency at the ~ 12 A/cm² region where the AERL/DARPA-supported one-meter experiments were performed is essentially unchanged.

In addition to the long pulselength results there are a number of short pulselength, high efficiency, experimental results that have been reported recently in the literature. A comparison of our model predictions for those experimental conditions indicated by the various authors is presented in Table II. The agreement with the 0.6 μ s results of AERL⁽⁷⁾ and the 50 ns results of Livermore⁽⁹⁾ is outstanding. The relatively high efficiency reported by the Sandia⁽¹⁰⁾ group is hard to reconcile with the Livermore result and the predictions of our code. High efficiency is certainly possible in short gain length lasers, especially when operated at higher fluorine densities $> 0.2\%$ (see Figure 4). Under these conditions, the increased F₂ causes a corresponding decrease in the steady-state electron number density and detrimental effects such as electron quenching are reduced. However, the accompanying increase in absorption precludes this solution for the scale sizes appropriate to produce the energy levels needed for this application.⁽³⁾

In the following section, the individual program Tasks are listed and a discussion of the individual results are presented. Most of these individual efforts were integrated into the kinetic code with the exception of the effect of determining the accessibility of the KrF*(B) state to the laser photons. These experiments are not complete and further research is necessary. It is partly for this reason that the code predictions above ~ 60 A/cm² has been shaded in Figure 2.

Figure 2 Scaling Map for the KrF* Laser (New Code)

Figure 3 Comparison of Old Code and New Code for a 1-Meter Laser Length

TABLE II
AERL
LASER OSCILLATOR CODE PREDICTIONS

	EXPERIMENTAL DATA					AERL CODE	
	GAIN LENGTH (CM)	PRESSURE (ATM)	DEPOSITION (MW/CM ³)	η_{INT} (ENERGY)	η_{INT} (POWER)	η_{INT} (ENERGY)	η_{INT} (POWER)
LLL	50	1.5	.7	9%	11%	9%	11%
LLL	50	2	1.2	9%	11%	9%	11%
SANDIA	40	1.3	1.8	12.8%		9%	
SANDIA	40	3.3	7	9%		6.5%	
AERL	100	1.7	0.13	9%		9%	

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Figure 4 KrF* Efficiency vs F₂ Density for a 50 cm Laser Length

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IV. TECHNICAL TASKS

A. F_2 BURNUP IN ArF^* AND KrF^* LASERS

The objective of this task is to experimentally measure the rates of F_2 destruction in typical e-beam excited ArF and KrF laser mixtures. These rates are important because they directly impact the intrinsic efficiency and specific energy achievable with these lasers. In addition, the results of these experiments are used to test our understanding of the various kinetic processes leading to F_2 destruction and to assess the predictive capability of existing ArF and KrF laser model codes. Once the predictive capability of these codes is established, then they can be used to accurately predict the ultimate performance achievable with large scale versions of these lasers.

Previously, (11) we have reported on the results of F_2 burnup measurements taken in binary mixtures of F_2 and Ar . Two independent methods were used for those measurements. The density of F_2 remaining in the laser mixture immediately after e-beam excitation was measured using UV absorption and plasma return current techniques. These techniques have been described in detail in an earlier report. (12) In summary, the two sets of results obtained using these F_2 diagnostic techniques agreed with each other to within the experimental uncertainty. The measured F_2 burnup rates were compared with DARPA-developed model predictions (see Section II). The comparison suggested that additional triatomic reaction processes must be included in the model to correctly account for the measured F_2 burnup rates, e.g., $e + Ar_2F^* \rightarrow$ products.

Similar measurements were subsequently taken in $F_2/Kr/Ar$ mixtures. For these experiments, the primary method used was the plasma return current technique. Selected points were checked using UV absorption to verify the validity of the measurements. UV absorption measurements were tedious because scatter in the data required averaging over many shots for each fixed e-beam energy deposition. Plasma return current measurements were straightforward and less time-consuming in comparison.

Figure 5 shows examples of typical plasma return current data. The top oscillogram shows the e-beam current density in the cell measured with a Faraday cup. The current continuously increased throughout the pulse due to diode closure in the cold cathode e-gun. The bottom oscillograms show the floating potentials that were generated by the plasma return current in two

E-BEAM CURRENT

2.5 A/cm²/DIV

TIME 200 nS/DIV

BACK PLATE
FLOATING POTENTIAL
0.2% F₂/99.8% Ar
2 ATM

250 V/DIV

BACK PLATE
FLOATING POTENTIAL
0.4% F₂/99.6% Ar
2 ATM

250 V/DIV

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Figure 5 Plasma Return Current Measurements of F₂ Burnup

mixtures with different F_2 concentrations. These show that the initial floating potential is proportional to the F_2 density in this range of F_2 concentrations and is independent of e-beam current density, as expected from the theory presented previously.⁽¹²⁾ The decrease in the floating potential later in the pulses is due to the decrease in electron attachment rate brought about by destruction of F_2 . This decrease is a measure of F_2 destruction once allowance is made for the variation in F_2 attachment rate constant due to the rise in temperature (see Appendix B) and electron mobility.⁽¹¹⁾

Data obtained in 2 atm mixtures of 0.4% F_2 /4% Kr/95.6% Ar are summarized in Figure 6. Also shown is the F_2 burnup calculated by the KrF model code after the addition of Kr_2F^* quenching kinetics. The dominant kinetic reactions included in the code are shown schematically in Figure 7. The added reactions are shown by dashed arrows. For these calculations we have used, for the Kr_2F^* radiative lifetime, a value of 165×10^{-9} s and a F_2 quenching rate constant of 3×10^{-10} cm³/s as measured by Chen and Payne.⁽¹³⁾ An electron quenching rate constant of 1×10^{-7} cm³/s similar to that found⁽¹¹⁾ for Ar_2F^* was assumed in the model. Within the scatter of the data, the measured F_2 burnup rate in 0.4% F_2 /4% Kr/95.6% Ar is the same as that found in 0.4% F_2 /99.6% Ar mixtures. Also, for these mixtures, the code predicted a slightly larger F_2 burnup rate ($\sim 3\%$) for the tertiary mixture. This small difference cannot be resolved by the present measurements.

In conclusion, we have measured the F_2 burnup rates in typical e-beam excited ArF and KrF laser mixtures. The results were compared with predictions of the existing ArF and KrF laser codes. The comparisons suggested that reactions involving the excited triatomics Kr_2F^* and Ar_2F^* must also be included in the laser model codes and that when these reactions are included, the code correctly predicts the F_2 burnup rate to within the accuracy of the experiments.

This gives further confidence in the code's general applicability and suitability for scaling calculations.

Figure 6 F₂ Burnup Measurement Results in Typical KrF Laser Mixture

Figure 7 Schematic of Dominant KrF Kinetics

B. ELECTRON QUENCHING

The rare gas halide lasers in general, and KrF in particular, have been extensively investigated by a number of research groups. As a result, much information has been accumulated on their formation and heavy particle quenching kinetics⁽⁴⁾ absorption⁽⁶⁾ and the impact of temperature on laser efficiency.⁽¹⁴⁾ For KrF* lasers ($\lambda = 249$ nm), this information has provided suitable input for models to make reliable predictions which agree with existing experimental data and project operating characteristics for large scale lasers.⁽²⁾ Having a reliable model allows one to optimize laser operating conditions (e-beam current, temperature, pressure, output coupling, path length, mixture, ect.) for use in a particular application, e.g., the design of a high-power, high-energy KrF* laser for inertial confinement fusion applications. In this approach, the high-energy laser pulse needs to be delivered to the target in ~ 10 ns. A number of publications have outlined potential methods to achieve such short laser pulses.⁽³⁾ Generally, pulse compression techniques are easier to accomplish for small compression ratios necessitating operation of the KrF* laser in a 50-100 ns pulse mode. This mode requires very high pump power densities which results in very large steady state electron densities. In order to limit the detrimental effect of electrons collisionally quenching the upper laser level, one might choose to operate at higher atom densities (e.g., F₂). This, however, will limit efficiency and place a length scaling limit on the gain length, since absorption and quenching also increase with increasing fluorine density. High efficiency operation of KrF* lasers has been reported^(10,15,16) in short pulse regimes for laser devices of < 60 cm length operating with F₂ densities near $\sim 0.3\%$. However, in order to optimize the KrF* and ArF* laser's performance and project to large gain length operation, a knowledge of the electron quenching rate constant is necessary.

We have measured under this task the electron quenching rate constants for the collisional relaxation of the upper laser levels of KrF* and ArF*, i.e.,

and found them to be near 2×10^{-7} cm³/s. These are slightly smaller than the value we reported⁽¹⁸⁾ for

of 4.0×10^{-7} cm³/s and larger than the theoretical predictions of Hazi, Resigno and Orel.⁽¹⁷⁾

The apparatus has been described in detail in earlier publications. (18-20) It consists of a spatially uniform, high energy e-beam originating from a broad area cold cathode which enters the reaction cell through a 2 mil Kapton foil. The reaction gas consisted of binary mixtures of argon and fluorine, in the case of ArF*, or krypton and fluorine for the KrF* experiments. The mixture pressure was chosen so that the closely coupled electronic states and their vibrational/rotational manifolds are well mixed. In binary mixtures, the formation kinetics are well documented and proceeds largely through ion-ion recombination mechanisms. Once formed, the RgF* molecules can radiate and be quenched by heavy particles or electrons. These electrons are present as a result of ionization in the gas by the primary e-beam. Through a variety of energy transfer collisions, these secondary electrons eventually attain average energies near 1 eV for the typical conditions used in these experiments. (20)

The resulting rare gas halide fluorescence is monitored at right angles to the e-beam direction by an optically filtered, appropriately attenuated, photodiode: a typical KrF* fluorescence spectrum and the filter transmission curve are shown in Figure 8. In the case of ArF*, the spectrally filtered signal impinges upon a glass plate coated with sodium salicylate. In this manner, the sensitivity of the photodiode was increased for 193 nm signal processing. The resulting temporal signals are recorded on a Tektronic 7844 oscilloscope. Viewing is restricted to a 1.5 mm slab immediately next to the foil and appropriate precautions were taken to insure the signals observed were free of amplified spontaneous emission. By choosing conditions (pressure, mix, e-beam current density), the effect of electrons on the quenching can be made relatively more important and, therefore, the corresponding rate constant more easily measureable.

In principle, to obtain a rate constant for electron quenching of the RgF* molecules, one uses a Stern-Vollmer type analysis, viz.,

$$\frac{1}{\text{signal}} = \text{constant} + k_{n_e} \tau [n_e] \quad (1)$$

and plots $1/\text{signal}$ vs n_e and relates the slope to the radiative lifetime, τ , and the rate constant for electron quenching, k_{n_e} . The electron number density in these experiments can be varied in two ways. First, one can perform experiments at various e-beam currents and obtain suitable ranges of electron densities as a direct result of changes in the pump power. Also, for dissociative attachment dominated experimental conditions such as

Figure 8 Fluorescence Spectra of KrF^* for a Mixture of 0.4 Torr of F_2 in 190 Torr of Kr at an E-Beam Current Density of 5 A/cm³. Also shown (smooth curve) is the filter used to monitor the sidelight emission.

these, the electron number density is inversely proportional to the F_2 number density. Variations in n_e can thus be accomplished through variations in F_2 . This has the desired effect of varying n_e but unfortunately also has a minor effect on the constant in Eq. (1), which contains all the heavy particle quenching including the quenching of RgX^* by F_2 . Separating the quenching due to F_2 , Eq. (1) reduces to a modified expression, viz.,

$$\left\{ \frac{1}{\text{signal}} = AB [F_2] \right\} \quad \text{vs } n_e \quad (\text{or vs } \frac{1}{F_2}) \quad (2)$$

A typical plot is presented in Figure 9 for 190 Torr of krypton at 7.6 amps/cm² and variations in F_2 density from 0.5 to 4 Torr. The slope of 2.9×10^{-7} s provides a value for $k_{el} \tau_{rad}/k_{ATT}$. To reduce these data, values of $\tau_{rad} = 6.5$ ns and $k_{ATT} = 4.5 \times 10^{-9}$ cm³/s were used. (20)

A summary of our experimental results is presented in Table III. Each of the measured rate constants were obtained from a computer least square fit to data similar to that shown in Figure 9. Each of the data points on these plots represents the average of 3 to 5 individual separate mixtures run at the same conditions. A consistent set of rate constants were obtained over a wide variation in J_{eb} , pressure and F_2 density. In the case of KrF^* quenching by electrons, a rate constant of $2.0 \pm 0.2 \times 10^{-7}$ cm³/s was observed. The ArF^* results were considerably more difficult to obtain due to the short radiative lifetime, $\tau = 2.5$ ns, rapid two-body quenching (e.g., F_2), shorter wavelength, and smaller stopping power of argon.

In general, however, these rate constants are large compared to theoretical studies on electron quenching by Hazi, Resigno and Orel. (17) These authors predict a value six to seven times lower for KrF^* than what was measured in these experiments. Similarly, they predict a value for XeF^* lower than has been measured (18) yet the measured ratio for $(XeF + e)/(KrF + e)$ is similar to the theoretical calculations. Part of the difficulty may lie in the fact that these measurements involve the relaxation of the collisionally mixed C- and B-states and their appropriate vibrational manifolds. Under such conditions, the appropriate radiative lifetime to choose to deduce a rate constant is not evident. (21)

Figure 9 Modified Stern-Vollmer Plot of 1/Signal (Corrected for F₂ Quenching) vs Electron Density (or 1/F₂ Density)

TABLE III
SUMMARY OF EXPERIMENTAL RESULTS

Reaction	J_{eb} (A/cm ²)	P_{Total} (atm)	$k_{el} \times 10^7$ (cm ³ /s)
KrF* + e →	15.3	0.25	2.0
		0.125	1.7
	7.6	0.25	2.2
		0.125	<u>2.2</u>
		Avg = < 2.0 ± 0.2 >	
ArF* + e →	44.4	0.25	1.6
	10.1	0.25	3.0

The likely mechanisms operable in these quenching reactions involve superelastic collisions,

and/or electron dissociative attachment processes

Deactivation of KrF^* by electrons as a result of superelastic collisions formed the basis for the theoretical effort of Hazi, Resigno and Orel⁽¹⁷⁾ and these results are consistent with this loss mechanism. These experimental rate constants are quite large to be entirely due to dissociative attachment processes for ~ 1 eV electrons in a room temperature gas.

Regardless of the nature of the quenching process, the magnitude of these measured rate constants suggest that electron quenching is a significant loss mechanism when the electron density, n_e , is in excess of 3×10^{14} #/cm³. Such is the case when these lasers are operated with high input powers as in UV preionized discharge and high current e-beam pumped systems.

C. GAIN/ABSORPTION MEASUREMENTS

The purpose of this task is to provide absorption/gain data over a wide range of operating conditions (in particular e-beam intensity) such that an extensive data base is generated with which to test and refine the kinetic code predictions.

The apparatus used for these measurements is shown in Figure 10. The $F_2/Kr/Ar$ mix is contained in a nickel coated aluminum cell. The e-beam enters the cell through a 2 mil kapton foil. The irradiated mix is then probed with a frequency doubled nitrogen-pumped dye laser, (Molelectron UV24-DLII). The doubled dye laser beam is first spacially adjusted and defined by using the beam expander and iris. A portion of the beam is then split off to a reference detector by means of a quartz flat. The remainder of the beam is used to probe the laser. The probe can be used in the four-pass configuration shown in Figure 10 or in a two-pass arrangement. The total effective path length through the irradiated region with the four-pass system is 60 cm. This length is necessary for absorption measurements under low pump flux conditions. A more convenient two-pass arrangement is sufficient for gain measurements and absorption measurements under high pump flux conditions.

After traversing the cell, the probe beam is then directed to the probe photodiode. KrF^* fluorescence emission viewed by this detector is reduced by placing the photodiode 2 m from the cell and using an iris diaphragm to restrict the detector area to that exposed to the probe beam. Filters which pass the doubled frequency, but block the primary frequency of the dye are located at the photodiodes. It is also necessary to place a diffusing screen a few centimeters from the detector face in order to maintain detector linearity. The signals from the reference and probe photodiodes are then sent to an oscilloscope in a shielded screen room for recording.

Gain measurements were made by tuning the probe laser to 248.5 nm. The wavelength was calibrated using a 0.5-meter monochromator and a mercury lamp. Using this method, the wavelength was determined to within ± 0.1 nm. Sharp absorption lines within the fluorescence profile have been observed.⁽²²⁾ To assure that the gain measurements were not influenced by accidental overlap with these features, measurements were periodically taken 1-nm on either side of 248.5 nm.

In order to evaluate the absorption at line center, measurements were taken at 240 and 260 nm. The reported absorption is then taken to be the average of the measured values at these two wavelengths.

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Figure 10 Schematic of Absorption Experiment

The e-beam current densities used were 20, 55, and 110 A/cm². The necessary decrease in current density from the 110 A/cm² maximum was accomplished by using attenuation screens. All the currents were directly measured using a carbon beam collector connected to ground through a Pearson current transformer.

Both absorption and gain measurements were taken using laser mixes of 0.6% F₂/4% Kr/95.4% Ar and 0.2% F₂/4% Kr/95.8% Ar, at 1, 2, and 3 atm total pressure, and current densities of 20, 55, and 110 A/cm². The absorption measurement results are tabulated, along with the corresponding model predictions, in Table IV. The gain results are given in Table V. Graphical representations of the experimental and code results for the 2 amagat case are shown in Figures 11 and 12.

Because of the cell geometry, it was not possible to make direct absolute energy deposition measurements for the region probed by the dye laser. For this reason the e-beam stopping power used in the computer code was adjusted to match the gain data at 20 and 55 A/cm². The code's reliability at these depositions was confirmed by its ability to predict laser efficiencies measured at AERL⁽²⁾ under conditions where accurate absolute depositions were known.

The principle absorbers for the deposition and pressure range studied in these experiments are Kr₂F*, Ar**, Kr** and F⁻. The absorption by F⁻ decreases as e-beam current increases. The Kr₂F* absorption predominates at all conditions but its importance increases with increasing pressure. Kr** and Ar** absorption increases with increasing e-beam current and decreasing pressure.

Theoretical analysis of the electronic spectrum of Kr₂F*⁽²³⁾ indicates that the absorption of Kr₂F* at the KrF* wavelength should be similar to that of Kr₂⁺. Using the cross section calculated⁽²⁴⁾ for Kr₂⁺ as a starting point, the Kr₂F* absorption cross section was adjusted, along with the F₂ quenching rate, until a good match was obtained between predicted and observed absorption in the regions where Kr₂F* absorption was most important. A cross section of 4.2×10^{-18} cm² yielded the best results. This is very similar to the calculated value for Kr₂⁺ (3.2×10^{-18} cm²); however the F₂ quenching rate constant (2×10^{-10} cm³/s) used for this "best fit" is about half the value reported in the literature.⁽²⁵⁾ This is consistent however with the limited data on the F₂ dependence (at various temperatures) of Kr₂F* emission which will be presented later in this report.

TABLE IV

SUMMARY OF ABSORPTION MEASUREMENTS AND CODE PREDUCTIONS (%/cm)

 J_{eb} (A/cm²)

	1		2		3		← Amagats
	0.2	0.6	0.2	0.6	2.0	0.6	← % F ₂
110	0.86	0.73	1.8	1.3	2.2	1.6	Exp.
	0.91	0.61	1.6	1.1	2.4	1.6	Code
55	0.41	0.32	0.91	0.60	1.1	0.89	Exp.
	0.53	0.36	1.0	0.64	1.6	0.86	Code
20	0.21	0.18	0.42	0.26	0.53	0.38	Exp.
	0.24	0.17	0.50	0.27	0.72	0.36	Code

TABLE V

SUMMARY OF GAIN MEASUREMENTS AND CODE PREDICTIONS (%/cm)

 J_{eb} (A/cm²)

	1		2		3		← Amagats
	0.2	0.6	0.2	0.6	0.2	0.6	← %F ₂
110	9.1	8.3	14	11	15	11	Exp.
	7.9	8.8	11	10	11	9.8	Code
55	5.1	5.3	6.6	5.1	6.6	5.3	Exp.
	5.2	4.7	6.6	5.4	6.2	5.0	Code
20	1.9	1.4	2.3	1.6	2.2	1.4	Exp.
	2.2	1.7	2.6	1.9	2.3	1.8	Code

Figure 11 Small-Signal Gain, Experimental Results and Model Predictions

Figure 12 Total Absorption Measurement, Experimental Results and Model Predictions

Although the absorption cross sections for the various excited states of Ar and Kr were calculated as part of this program (see Section IV.D), the total contribution from $\text{Ar}^* + \text{Kr}^*$ is still uncertain since the population distribution among the various excited, absorbing states is not known. For this reason, the Ar^* and Kr^* cross sections were varied within the calculated variance range until a good fit to the data in the regions where these species are important was obtained.

The results listed in Table IV and V show that the code is capable of predicting gain and absorption over a broad range of pressure, F_2 density, and pump flux.

D. PHOTOIONIZATION CROSS SECTIONS FOR EXCITED STATES OF THE RARE GAS ATOMS

1. Introduction

Under intense pumping conditions, which are required for short-pulse operation of the rare gas halide lasers, significant population densities of the high-lying atomic levels are expected. Photoionization of these states by the laser photons can subsequently lead to reduced laser extraction efficiency. The relevant cross sections for photoionization of these excited states of the rare gases (Ne, Ar, Kr and Xe) have thus been calculated as part of the present program and the results are presented in this section of the final report.

Recent theoretical interest in the photoionization of excited atomic states has resulted from the importance of such processes in the performance of rare gas-halide exciplex lasers,⁽²⁶⁾ as well as from the discovery of unexpected features in excited-state photoionization cross section.⁽²⁷⁾ Both of these aspects of the problem have led us to investigate the photoionization of the first excited s-, p-, and d-states of the heavier rare gases (excluding helium). Specifically, calculations have been carried out for processes of the type

$$X^*(...p^5 n\ell) + h\nu \rightarrow X^+(...p^5) + e$$

with $X^* = \text{Ne}^*(n\ell = 3s, 3p, 3d)$; $\text{Ar}^*(n\ell = 4s, 4p, 3d)$; $\text{Kr}^*(n\ell = 5s, 5p, 4d)$; and $\text{Xe}^*(n\ell = 6s, 6p, 5d)$.

Results of calculations on the $\text{Ar}^*(4s)$ and $\text{Kr}^*(5s)$ states (which include the $^3P_{0,2}$ metastable levels) as well as on the $\text{Ar}^*(4p)$ and $\text{Kr}^*(5p)$ states were first reported by Hyman.⁽²⁶⁾ These calculations revealed the existence of a Cooper minimum in the s-state cross sections. In addition, it was pointed out that the excited p-state photoionization cross sections were very much larger than those of the s-states over the range of photon energies relevant to the rare gas-halide lasers ($h\nu \lesssim 10$ eV). Calculations on the photoionization of the metastable $X^*(ns^3P_{0,2})$ states of the rare gases by McCann and Flannery⁽²⁸⁾ confirmed the existence of the Cooper minimum for argon and krypton and indicated similar behavior for the neon and xenon cross sections. A more elaborate theory has been applied to the neon metastable photoionization problem by Hazi and Rescigno.⁽²⁹⁾ By including many-electron correlation effects within an LS-coupling scheme, they found a finite, non-zero value at the minimum of the cross section, as contrasted with the zero value obtained from the single-channel, central-field methods.^(26,28) Finally, Hartquist⁽³⁰⁾ has carried out a quantum defect calculation in

the $j\ell$ -coupling scheme for the photoionization of the four $X^*[ns(J)]$ levels of the heavier rare gases, and his results are in qualitative agreement with those discussed above. The only relevant experimental results consist of upper bounds on the $Ar^*(4s^3P_{0,2})$ and $Kr^*(5s^3P_{0,2})$ cross sections at threshold, reported by Dunning and Stebbings. (31)

The present calculations are based upon a modification of the method used earlier (see Ref. 26); we therefore briefly review the theory in the next section. The results of our cross-section calculations are presented in Section D.3, where they are compared with the existing work in the literature. Aside from the Cooper minimum appearing in each of the s-state cross sections, additional minima have been found in two other cases: for photoionization of the $Ne^*(3p)$ and $Xe^*(5d)$ states. These features will be discussed in detail. Finally, we will indicate the relevance of our results to the physics of rare gas-halide lasers. Section D.4 contains a summary and conclusions.

2. Method

As described previously, (26) we have averaged over all J-levels of each excited-state configuration, so that the excited states can be simply represented in the form $X^*(...p^5n\ell)$, where $n\ell$ denotes the orbital of the active electron. The cross section for photoionization of the $N\ell$ th state is then given by the expression

$$\sigma_{n\ell}(\epsilon) = 2.69 \times 10^{-18} (I_{n\ell} + \epsilon) \frac{\ell+1}{2\ell+1} R_{\ell+1}^2 + \frac{\ell}{2\ell+1} R_{\ell+1}^2 \quad (\text{cm}^2) \quad (2)$$

where $I_{n\ell}$ is the ionization potential and ϵ is the energy of the ejected photoelectron (all energies are in Rydbergs). R is the radial matrix element, defined by

$$R_{\ell+1} = \int_0^\infty P_{n\ell}(r) r P_{\epsilon, \ell+1}(r) dr \quad (3)$$

where $P(r)$ is the solution of the radial Schrodinger equation:

$$\left[\frac{d^2}{dr^2} - \frac{\ell(\ell+1)}{r^2} + \epsilon + V(r) \right] P(r) = 0 \quad (4)$$

We take the central-field potential to be of the form

$$V(r) = V_{\text{ion}}(r) + \frac{\alpha}{(r^2 + r_0^2)^2} \quad (5)$$

where the first term is the potential of the parent ion, $X^+(\dots p^5)$, which was constructed from the Hartree-Fock wavefunctions of Clementi and Roetti, (32) while the second term is a polarization potential, with α the core polarizability and r_0 an effective cutoff radius. The value of r_0 was taken to be the Hartree-Fock mean radius of the outer shell of the core. In our earlier work, (26) α was estimated by scaling from the known polarizabilities of analogous atoms and ions, whereas in the present work a different procedure has been used. We have regarded α as a parameter to be determined from a least-squares fit of calculated binding energies to the experimentally-known energies (33) of the excited atomic states. A sufficient number (~ 15) of excited-state configurations were included so as to span about an order-of-magnitude in binding energy for each atom. The results of this method are given in Table VI, which lists r_0 (in units of a_0 , the Bohr radius, α (in a_0^3), and the average percent deviation between the calculated and experimental energies. The fit is very satisfactory, particularly since we varied only one parameter in the central-field potential. The values of the polarizability are very different from those estimated previously, (26) and in general are much larger than one would expect for the "true" dipole polarizability of the core. This results from the fact that with our present semi-empirical method, the polarization term in expression (5) not only accounts for the actual core polarization effect, but must also simulate exchange, the quadrupole potential of the open-shell core (for non s-states), the over estimate of nuclear screening due to the use of fully-relaxed ionic wave functions to construct $V_{\text{ion}}(r)$, and other small effects.

TABLE VI
PARAMETERS FOR EXCITED STATES OF THE RARE GASES

<u>Atom</u>	<u>$r_0(a_0)$</u>	<u>$\alpha(a_0^3)$</u>	<u>Average Fit (%)</u>
Ne	0.8759	2.610	0.5
Ar	1.559	15.64	0.5
Kr	1.843	28.82	0.7
Xe	2.222	53.80	1.4

Using the values listed in the table, the Schrodinger Eq. (4) was solved numerically subject to the boundary conditions: $P(0) = 0$, and for large r , $P(r) \rightarrow 0$ for the bound excited states and $P(r) \rightarrow \pi^{-1/2} \epsilon^{-1/4} \sin [\epsilon^{1/2} r - \ell\pi/2 - \epsilon^{-1/2} \ln(2\epsilon^{1/2} r) + \arg r (\ell + 1 - i\epsilon^{-1/2}) + \delta\ell(\epsilon)]$ for the continuum states, where $\delta\ell(\epsilon)$ is the phase shift. Since the same potential, $V(r)$, was used for all bound and continuum states, the wave functions are automatically orthogonal.

3. Results and Discussion

The $\text{Ne}^*(3s)$ photoionization process has received the most extensive treatment, and Figure 13 shows our cross section plotted together with those of McCann and Flannery⁽²⁸⁾ and Hazi and Rescigno⁽²⁹⁾ (both length and velocity forms). Away from the Cooper minimum all of the calculations agree reasonably well. The fact that Hazi and Rescigno included all open channels arising from the coupling between the open-shell core and the ejected p-electron results in a non-zero minimum. All purely central-field methods (including ours and that of Ref. 28) yield a zero value at the minimum.

Figure 14 shows the s-state photoionization cross sections for the heavier rare gases as a function of the wavelength of the incident photon. They all exhibit a Cooper minimum near threshold, which is indicative of strong cancellation effects in the radial matrix element. Such cancellation effects make the cross sections highly sensitive to the details of the model. Thus, the fact that our present values of α for argon and krypton are different from those used previously, in turn causes our present cross sections to differ in detailed form from those obtained earlier.⁽²⁶⁾ They are, however, qualitatively similar, particularly in that they are very small in absolute magnitude. Our present results are also in quite good agreement with those of McCann and Flannery.⁽²⁸⁾ A direct comparison with Hartquist's calculations⁽³⁰⁾ is difficult, due to his use of $j\ell$ -coupled states; however, he also finds Cooper minima near threshold and his cross sections are small. In addition, our results for argon and krypton are consistent with Dunning and Stebbings⁽³¹⁾ upper-bound experimental values ($1.1 \times 10^{-18} \text{ cm}^2$ for Ar and $4.9 \times 10^{-19} \text{ cm}^2$ for Kr) at threshold, although this can only be considered a rough order-of-magnitude comparison.

The photoionization cross sections for the first excited p-states are shown in Figure 15. They are clearly considerably larger than those for the s-states. The neon $3p \rightarrow \epsilon d$ transition has a minimum at $\epsilon = 0.37 \text{ Ry}$, which causes the total cross section first to drop off rapidly, and then to level off due to the small $3p \rightarrow \epsilon s$ contribution. This minimum was not observed for the corresponding $np \rightarrow \epsilon d$ transitions in argon, krypton, or xenon. In

Figure 13 Photoionization Cross Sections for $\text{Ne}^*(2p^5 3s)$ vs Photoelectron Energy (ϵ): ——— present work, ——— McCann and Flannery (Ref. 28), --- Hazi and Rescigno (Ref. 29) length form, Hazi and Rescigno (Ref. 29) velocity form.

Figure 14 Photoionization Cross Sections vs Wavelength of Incident Photon for Excited s-States: --- Ne* (2p⁵3s), — Ar* (3p⁵4s), — Kr* (4p⁵5s), — Xe* (5p⁵6s).

Figure 15 Photoionization Cross Sections vs Wavelength of Incident Photon for Excited p-States: --- $\text{Ne}^*(2p^5 3p)$, — $\text{Ar}^*(3p^5 4p)$, — — $\text{Kr}^*(4p^5 5p)$, — . — $\text{Xe}^*(5p^5 6p)$.

order to more fully understand this behavior, we have plotted the relevant oscillator strength distributions in Figure 16. The strengths of some of the discrete $np \rightarrow n'd$ transitions are included in the form of histograms, constructed according to the technique described by Fano and Cooper.⁽³⁴⁾ These plots clearly show how the minimum present in the continuum for neon shifts into the discrete portion of the spectrum for the heavier species.

Finally, the d-state cross sections are plotted in Figure 17. They are also large near threshold and fall rapidly with decreasing wavelength. Xenon has been found to exhibit minima in both the $5d \rightarrow \epsilon p$ and $5d \rightarrow \epsilon f$ transitions, analogous to the behavior of the cesium 5d-state photoionization cross section studied by Msezane and Manson.⁽²⁷⁾ In both xenon and cesium, the dipole matrix element for the $5d \rightarrow \epsilon p$ transition is negative at threshold and goes through zero at low ejected electron energy ($\epsilon = 0.31$ Ry for Xe and $\epsilon = 0.074$ Ry for Cs). Due to the fact that the $5d \rightarrow \epsilon p$ contribution to the total cross section is very much smaller than that due to the $5d \rightarrow \epsilon f$ transition, the structure resulting from this $d \rightarrow p$ minimum does not show up on the plot of Figure 17. The $5d \rightarrow \epsilon f$ matrix element starts out positive and changes sign at $\epsilon = 0.75$ Ry, yielding the sharp minimum in the xenon cross section shown in the figure (this is analogous to the cesium minimum occurring at $\epsilon = 0.164$ Ry). In the cesium case there is a second sign change (made necessary by the fact that the matrix element must eventually become positive for large energies) at 7.4 Ry. Since our calculations only covered the range up to $\epsilon < 1$ Ry, we did not observe this second $d \rightarrow f$ minimum, but it must presumably exist at some higher energy where the matrix element again goes positive. In general, the similarity between the $Xe^*(5d)$ and $Cs^*(5d)$ photoionization cross sections is not surprising since the 5d-electron lies well outside the respective cores, which are themselves not too dissimilar in structure.

As indicated in the introduction, excited-state photoionization processes in the rare gases can significantly impact the performance of rare gas-halide lasers. This will be especially true for the laser operating regime suitable for inertial confinement fusion (ICF) applications. A laser driver for ICF must operate in the short-pulse regime, necessitating intense e-beam pumping, which in turn can lead to large population densities of excited rare gas atoms. As an example, for a typical atmospheric pressure krypton-fluoride laser mixture, with an e-beam current ~ 100 A/cm², absorption from excited atomic states can account for $\geq 50\%$ of the total photoabsorption of laser photons in the medium.⁽³⁵⁾ A knowledge of the relevant photoionization cross sections is therefore important for modeling and thereby optimizing such lasers. We have listed in Table VII the cross sections pertinent to the

Figure 16 Oscillator Strength Distributions in the Discrete and Continuous Spectra for the Transitions $np \rightarrow n'd(ed)$.

Figure 17 Photoionization Cross Sections vs Wavelength of Incident Photon for Excited d-States: --- $\text{Ne}^*(2p^5 3d)$, — — — $\text{Ar}^*(3p^5 3d)$, — $\text{Kr}^*(4p^5 4d)$, — · — $\text{Xe}^*(5p^5 5d)$.

TABLE VII

PHOTOIONIZATION CROSS SECTIONS FOR EXCITED RARE GAS ATOMS
RELEVANT TO THE KrF AND XeCl LASERS

		Cross Section (10^{-18} cm^2)	
<u>Atom</u>	<u>State</u>	<u>$\lambda = 240 \text{ nm (KrF)}$</u>	<u>$\lambda = 308 \text{ nm (XeCl)}$</u>
Ar*	$3p^5 4p$	5.4	
	$3d$	2.4	
Kr*	$4p^5 5p$	5.2	
	$4d$	2.8	
Ne*	$2p^5 3p$		2.3
	$3d$		0.6
Xe*	$5p^5 6p$		7.6
	$5d$		9.3

krypton-fluoride (KrF) and xenon-chloride (XeCl) lasers, operating at $\lambda = 248$ nm and $\lambda = 308$ nm, respectively (these currently appear to be the leading candidates for short-wavelength ICF laser drivers). The KrF and XeCl lasers typically use argon and neon, respectively, as buffer gases. The table includes only the p- and d-state cross sections, since photoionization from the s-states is negligible at the wavelengths of interest.

4. Summary and Conclusions

Calculations have been carried out to obtain photoionization cross sections for the first excited s-, p-, and d-states of the heavier rare gases. Wave functions were determined from a semi-empirical method, in which an effective core polarizability was adjusted to fit the computed eigenvalues to the known excited-state binding energies for each atom. This method should give a good representation of the wave functions at large radii, the region of configuration space relevant to the calculation of dipole matrix elements. A number of interesting features have been found in the various cross sections, and these have been described and analyzed. In addition, the role of excited-state photoionization in short-pulse rare gas-halide lasers for fusion was briefly considered. The relevant cross sections were shown to be large ($\sim 10^{-18}$ - 10^{-17} cm²), and thus these processes are important for modeling and understanding the performance of such lasers.

E. ACCESSIBILITY OF THE $\text{KrF}^*(\text{B})$ STATE TO LASER PHOTONS

1. Introduction

In this section we will discuss the accessibility of the upper level manifold of KrF^* to laser photons. The upper level of KrF^* is formed by ion-ion recombination and rare gas metastable reactions with F_2 . As a result, a large fraction of the energy flows through the KrF^* high-lying vibrational manifold before it resides preferentially in the lower levels as dictated by the mixture temperature. Under high pump conditions, and, hence, large electron densities, the lifetime of KrF^* could be reduced sufficiently such that equilibrium may not be established. Under these conditions, a significant fraction of the energy may be lost due to quenching processes before the lower lasing levels are populated. The accessibility of these states will depend on two issues (a) the rate of vibrational relaxation, and (b) the probability of stimulating the higher levels by laser flux. In this section we will present experimental results that strongly indicate that finite vibrational relaxation could impact the laser performance at the higher pump intensities required for inertial confinement fusion applications.

The effect of finite vibrational relaxation can be considered in a simple way by analysing the following rate equations.

$$\frac{dN_o}{dt} = - \frac{(N_o - N_{oe})}{\tau_v} - \frac{N_o}{\tau_o} - \frac{N_o}{\tau_s} \quad (6)$$

and

$$\frac{dN}{dt} = R - \frac{N}{\tau} - \frac{N_o}{\tau_s} \quad (7)$$

where N_o is the population of the zeroth vibrational level of the $\text{KrF}(\text{B})$ state; N_{oe} is the equilibrium population of the zeroth level, $N_{oe} = \Theta_o N$, where Θ_o is the partition function and $N = \sum_v N_v$, τ is the lifetime of the upper level, τ_s is the stimulated lifetime of the upper level, R is the pumping rate for producing the excited states and $1/\tau_v$ is the vibrational relaxation rate.

Equations (6) and (7) can be solved under steady-state conditions to give:

$$\frac{N}{N_0} = \frac{1 + (\phi/\phi_s) \frac{1}{\beta}}{(1 + \phi/\phi_s)} \quad (8)$$

where ϕ is the cavity flux and ϕ_s is the saturation flux defined as the flux necessary to reduce the upper state population by a factor of two, N_0 is the upper state population density when $\phi = 0$ and

$$\beta = 1 + \theta_0 \phi/\phi_v \quad (9)$$

Note that when vibrational relaxation is infinitely fast, $\tau_v \rightarrow 0$ and $\beta \rightarrow \infty$. Under these conditions the depression of the KrF* sidelight fluorescence will scale with $(1 + \phi/\phi_s)^{-1}$. Also, from Eq. (8), if vibrational relaxation is finite, as $\phi \rightarrow \infty$, N_0 will asymptote to β .

To investigate these phenomena directly, we performed experiments on a one meter laser device. The e-beam excited gas mixture was 0.2% F₂, 4% Kr and 95.8% Ar at a total pressure of 1 atm. The 400 kV, 24 A/cm² beam entered the laser cavity through a 0.001" Kapton foil. To insure uniform deposition of the electrode beam into the gas through the (4 x 100) cm² foil, a guide magnetic field was imposed of 1 to 2 kG.

The characteristic 1 μ s pulse length of this device was decreased to 400 ns to reduce the energy on the cavity optics. The cavity consisted of a high feedback output coupler (91.78% or 99.92% reflector) in conjunction with a MAX reflector. With this arrangement cavity fluxes of 3 MW/cm² were achieved without encountering severe damage on individual optical components.

The resulting sidelight fluorescence was monitored at a sidelight viewing port located near the edge of the excitation region closest to the output coupler (see Figure 18). The sidelight and laser flux were monitored with various calibrated photodiodes. Appropriate filters were used to isolate the spectral region of interest and protect the detectors from signals whose intensities were too great (i.e., nonlinear effects). This arrangement allows us to determine the cavity flux in the region of interest in a simple way. The laser energy was monitored by a Scientech model 362 power/energy meter.

Care was taken to insure the sidelight viewing was restricted to excitation regions in the cavity that experienced the full cavity flux, i.e., there was no radiation coming from molecules that could not be stimulated due to being in an area outside the flux. This was checked by performing a series of experiments whereby metal attenuators were placed over the e-beam foil to completely stop the electrons from entering the cell for various cavity heights, e.g., the normal 4 cm (full cavity) was reduced to 1.4 and 0.75 cm. In all cases, the results were the same (see Figure 19).

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Figure 18 Schematic Diagram of Experiment. The e-beam emerges through the foil out of the plane of the figure.

These data show the sidelight depression (signal in the absence of flux, N_0 , to signal in the presence of cavity flux, N) as a function of cavity flux.

The saturation flux for these data in Figure 19 was 0.95 MW/cm^2 . From the data, it is clear N_0/N does not vary linearly with ϕ/ϕ_s which it would do if KrF was totally homogeneously broadened. From the asymptote, we estimate β lies between 4 and 5 (curve marked β). Since from Eq. (9), β is related to the vibrational relaxation time, we can make an estimate of τ_v . At room temperature $\theta_0 \sim 0.75$ and $\tau \approx 4.3 \text{ ns}$, so $\tau_v \approx 0.8 \text{ ns}$. Assuming $(kQ)^{-1} = \tau_v$ and $Q = \text{Ar} = 2.49 \times 10^{19} \text{ particles/cm}^3$, $k \sim 5 \times 10^{-11} \text{ cm}^3/\text{s}$ for a two-body rate constant and $2 \times 10^{-30} \text{ cm}^6/\text{s}$ for a three-body rate constant. This means that even if $\text{KrF}^*(v)$ were being relaxed in almost every collision, for conditions where the quenching lifetime is short, this effect could be considerable.

Should the laser flux at a fixed wavelength be capable of assessing all vibrational levels of the $\text{KrF}^*(B)$ state, then a vibrational relaxation model falls short of providing an explanation for these data. Consider the case presented in Figure 20, where the KrF fluorescence band is depicted along with the calculated contributions from the first few vibrational levels. Assuming that each of these levels exhibit gain at the laser wavelength, then the sidelight depression equation reduces to

$$N_0/N = \frac{1}{\sum_i \frac{\theta_i}{1 + \phi/\phi_{si}}} \quad (10)$$

where ϕ_{si} is the saturation flux appropriate for the i^{th} level. Consider the fit to these data with this model when ϕ_{si} for $i > 0$ is made larger than ϕ_{s0} by about a factor of two or three because of the anticipated reduced gain (see Figure 19).

It is apparent that neither model entirely fits the data perfectly, but these results do indicate that incomplete coupling is evident. It is possible that both effects occur simultaneously, but vibrational relaxation effects are more reasonable (e.g., $\phi_{si} = 20 \phi_{s0}$ to fit the data). To determine the contributions of each of these mechanisms, additional experimentation is necessary. Such experiments should include observations of sidelight spectral behavior as a function of cavity flux for different gas mixtures, pressures and various spectral widths. To predict the impact of these issues on the efficiency of the KrF^* laser at pump intensities near 10^6 W/cm^2 , more information is needed.

Figure 19 Sidelight Ratio vs Cavity Flux. The dashed line shows the expected behavior for a homogeneously broadened laser.

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Figure 20 Spectral Fluorescence Intensity for KrF*(B) Radiation as a Function of Wavelength. Depicted is Contributions from $v = 0$ and $v = 1$.

F. TEMPERATURE DEPENDENCE OF KrF^* FLUORESCENCE

It is the purpose of this Task to investigate experimentally the increase in fluorescence efficiency of KrF^* as the laser gas mixture is heated in a special reaction cavity passivated for minimal chemical effects on the fluoride.

1. Experimental

The temperature dependences of the KrF^* and Kr_2F^* emission were measured in the apparatus shown schematically in Figure 21. The $\text{F}_2/\text{Kr}/\text{Ar}$ mixture is excited by an e-beam from the Marx bank entering the cell through a 2-mil kapton foil. The e-beam characteristics are 20 A/cm², 250 kV with a pulse length of 120 ns. There is a 1400 G guide magnetic field coil around the cell which improves the system reliability and simplifies the energy deposition analysis.

The experiments were performed over the temperature range 300°K to 420°K. The temperature of the cell is controlled by means of cartridge heaters in the cell body and window holders. The fill line was also maintained at the cell temperature to assure temperature equilibration of the gas mix with a minimum of wait time between filling the cell and firing the Marx bank.

Because of the enhanced reactivity of F_2 at elevated temperatures, the aluminum cell was nickel coated. The thermal stability of F_2 , over the temperature range of interest, was confirmed by a series of experiments which measured the F_2 density (in the Kr/Ar mixtures used) as a function of time. The density was monitored by measuring the absorption of the 253.7 nm Hg line by the F_2 .

The KrF^* and Kr_2F^* fluorescence emission was measured using an ITT calibrated S5 photodiode. A narrow band (~ 20 nm), dielectric filter was used to observe the KrF^* emission and a Corning O-52 blocking filter was used in front of the Kr_2F^* detector.

2. Results and Discussion

Measurements of the KrF^* and Kr_2F^* emission were obtained at 315, 365, and 420°K, and total densities of 1, 2, and 3 amagats, for the two mixes, 0.26% $\text{F}_2/4\%$ $\text{Kr}/95.7\%$ Ar and, 0.79% $\text{F}_2/4\%$ $\text{Kr}/95.2\%$ Ar . The results for the 0.26% F_2 mix are shown in Figure 22. Qualitatively, one can observe the expected decrease in the three-body rate constants with temperature as a decrease in Kr_2F^* emission at elevated temperatures. At high densities, 2 and 3 amagats, the three-body relaxation of KrF^* is dominant and one can

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Figure 21 Schematic of Experimental Setup (Temperature Dependent Fluorescence Measurements)

Figure 22. Temperature Dependence of KrF* and Kr₂F* Emission

see the increase of Kr_2F^* emission as the rate of these relaxation processes are reduced. At 1 amagat, other processes which reduce the KrF^* emission are dominant and the expected increase is not observed.

The three-body reactions of interest are

The rate constants for these reactions has been measured (36) at room temperature and are 6.5×10^{-31} and $8 \times 10^{-32} \text{ cm}^6/\text{s}$, respectively. Additional important quenching processes are

with the corresponding rate constants of $7.8 \times 10^{-10} \text{ cm}^3 \text{ sec}^{-1}$ (4) and $2 \times 10^{-7} \text{ cm}^3 \text{ sec}^{-1}$ (37). Considering the above reactions, the steady-state expression for the fluorescence intensity from KrF^* is

$$S_{\text{KrF}^*} = \frac{\alpha(\text{Ar})}{1 + \{k_1(\text{Kr})(\text{M}) + k_2(\text{Ar})(\text{M}) + k_3(\text{F}_2) + k_4(e^-)\}\tau_1}$$

where α is a constant, (M) is the total gas density (cm^{-3}), and τ_1 is the radiative lifetime of KrF^* ($6.5 \times 10^{-9} \text{ s}$). The electron density can be estimated by knowing the e-beam stopping power and current ($5.75 \times 10^3 \text{ eV cm}^{-1} \text{ atm}^{-1}$, 20 A/cm^2), electron attachment rate by F_2 ($4.5 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$), (see Appendix B) and the fraction of F_2 in the mix. The electron density is thus calculated to be $9.3 \times 10^{13} \text{ cm}^{-3}$ and $3.1 \times 10^{13} \text{ cm}^{-3}$ for the 0.26% F_2 and 0.79% F_2 mixes respectively.

The above expression can be used to estimate the fluorescence emission, but because of corrections necessary to account for slight non-steady state behavior ($\sim 10\%$ for 1 amagat), and amplified spontaneous emission (variations of ~ 5 to 10% over these experimental conditions), the computer code which was updated for this contract was used to evaluate the data. All the rate constants, except those for the three-body reactions, were assumed to

be temperature independent, and the room temperature values were used. The three-body rate constants were evaluated at each temperature by using the experimentally determined room temperature rate constants given above, normalized to the temperature dependence functions calculated by Shui.⁽³⁸⁾ The rate constants used are given in Table VIII. In Figure 23, the experimentally measured KrF* emission, along with the corresponding model predictions, is plotted versus pressure for the three temperatures used. As the density is increased, there is a rollover in KrF* fluorescence intensity due to the three-body relaxation processes. When the temperature is increased, this rollover is significantly reduced. As can be seen, the model results which use the theoretically determined temperature dependence agree well with experiment.

The model predictions shown in Figure 23 indicate that the increase in KrF* emission with temperature is due to a corresponding reduction in the three-body quenching rates. Further evidence to support this assumption can be obtained from the quantitative analysis of the Kr₂F* fluorescence intensity. Kr₂F* is produced primarily* by reaction (11), and reaction (12) followed by the exchange reaction:

It is postulated that the rate constant for this reaction is on the order of $5 \times 10^{-10} \text{ cm}^3 \text{ sec}^{-1}$. This is based on the fact that the reaction is exothermic and that the rate constant for the reaction:

has been measured⁽¹⁴⁾ to be $1.6 \times 10^{-9} \text{ cm}^3 \text{ sec}^{-1}$. In this case, reaction (15) should dominate over other ArKrF* loss processes. The effect of temperature on the formation of KrF*, (or relaxation of KrF* not involving Kr₂F* production) can be factored out of the analysis of the Kr₂F* emission by looking at the ratio Kr₂F*/KrF*. A plot of this ratio versus temperature for the 0.26% F₂ mix at 3 amagats is shown in Figure 24. The temperature variation of this ratio, normalized to the corresponding low temperature point, was the same for all pressures and F₂ densities used in this study. As was the case for the absorption and gain data reported earlier, it was necessary to use the value of $2 \times 10^{-10} \text{ cm}^3 \text{ sec}^{-1}$ for the F₂ quenching of Kr₂F* in order to get the proper F₂ dependence of the Kr₂F*/KrF* ratio.

* < 10% could be formed via Ar₂F* followed by exchange reactions.

TABLE VIII

TEMPERATURE DEPENDENCE OF THREE-BODY RATE CONSTANTS

°K	k (cm ⁶ sec ⁻¹)	k (cm ⁶ sec ⁻¹)
	KrF* + Kr + M	KrF* + Ar + M
300	6.5×10^{-31}	8.0×10^{-32}
315	4.9×10^{-31}	6.9×10^{-32}
365	2.8×10^{-31}	4.3×10^{-32}
420	1.8×10^{-31}	2.7×10^{-32}

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Figure 23 Temperature Dependence of the Three-Body Quenching of KrF* Fluorescence

Figure 24 Temperature Effect on the Ratio of Kr₂F^{*}/KrF^{*} Fluorescence

Again, there is good agreement between theory and experiment. If one assumes that the relaxation processes for Kr_2F^* are temperature independent, the variation in $\text{Kr}_2\text{F}^*/\text{KrF}^*$ should follow the variation with temperature of the weighted average of the rate constants for reactions (11) and (12). The results indicate that the theoretically determined temperature dependences yield rate constants that are within 20% of the values needed to obtain complete agreement between theory and experiment. This correction in the temperature reduction of the three-body rate constants would also improve the agreement between model and experiment shown in Figure 23.

In summary, the results reported here indicated that, with only minor adjustments, the theoretical predictions of Shui(38) give a reliable description of the temperature dependence of the three-body reactions. However, there does seem to be other processes varying with temperature as evidenced by the low pressure KrF^* results. Additional research into this area will be necessary for good predictions of the temperature dependence of laser efficiency.

V. SUMMARY

In carrying out this program, we have made extensive contributions to further the understanding of the impact of a number of key kinetic issues important for efficient operation of KrF* lasers (see Figure 25). These include the effects of fuel burnup and the magnitude of electron quenching and photoionization processes. Also, an extensive data base was collected regarding gain/absorption features at a variety of wavelengths, current densities, pressures, mixture components and mole fractions. Based on these results, additional reaction-rate constants and cross sections are now available with which to model the krypton fluoride laser. The effect of these new data can be appreciated by the information contained in Figure 26. Shown is the sidelight fluorescence data with and without laser flux normalized at the peak intensity. The new code predicts the shape of the experimental data quite well, but predicts a greater sidelight depression than was experimentally observed. The issue of the accessibility of the higher lying levels of KrF* by the laser flux is not complete and therefore has not been included in the model. Additional experimentation is needed to establish the impact of this feature on efficient short-pulse operation. Also, the experiments on the temperature influence on KrF* fluorescence should be extended to include temperature effects on laser performance. Overall, these kinetic issues suggest that efficient high-power, large-aperture lasers can be built at efficiencies consistent with ICF requirements. Clearly, other system constraints must be considered in designing such a device, but reliable kinetic modeling is reasonably assured.

- PULSE LENGTH DETERMINED BY F₂ BURNUP

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Figure 25 Key Kinetic Issues Concerning KrF* Laser Performance

Figure 26 Sidelight Fluorescence with and without Laser Flux. Also shown are the code predictions (old and new).

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APPENDIX A

PULSE SHAPE AND LASER ENERGY EXTRACTION FROM E-BEAM PUMPED KrF*

1. INTRODUCTION

The physical processes which determine the performance of KrF* lasers have been intensively researched during the last two years. As a result of this research, for e-beam excitation intensities in the $1-5 \times 10^5 \text{ W/cm}^3$ regime, the dominant formation and quenching kinetics are well understood, (1,2) the stimulated cross section has been measured, and the dominant absorbers in the active medium have been identified. (3) The above information allows one to construct a comprehensive model which can be used to predict laser performance over a wide range of laser operating conditions. Experimental verification of the predictive capability of such a model then provides an independent check on the various kinetics leading to medium gain and absorption, as well as a check on the validity of power extraction model.

In this report, a description of such a code is presented. The predictions of the code are compared with experimental results obtained with a 1 m e-beam excited KrF* laser device. The comparison includes the temporal shape of the KrF* laser output and sidelight pulses, the output pulse energy, and intrinsic laser efficiency. Also included is a detailed description of the 1 m laser device. The model predicts the laser energy and power extracted and efficiencies to within 10% of the experimental values. A detailed comparison of the predicted and measured sidelight emission, however, show evidence of processes not previously addressed. These include (a) the effect of finite vibrational relaxation and (b) electron quenching of KrF*. Although for the experimental conditions discussed in this article the effect of these processes on the overall laser performance is about 10%. They may become important at pump powers $> 5 \times 10^5 \text{ W/cm}^3$.

In Section 2 we summarize the formation and quenching kinetics. In Section 3 we discuss how the kinetics are coupled to power extraction by the laser cavity flux. In Section 4 the experimental apparatus and a detailed comparison between the results and model are presented. In Section 5 a discussion of the processes not previously investigated will be addressed. In Section 5 a scaling map for the KrF* laser is also presented.

2. KrF* LASER KINETICS

Figure A-1 shows a block diagram which summarizes the important processes included in the computer code. The e-beam energy results in the formation of electron and rare gas ion pairs. The secondary electrons are rapidly lost via dissociative attachment with the halogen donor to form F^- . These ionic states eventually result in the formation of KrF^* .⁽¹⁾ The dominant formation kinetics and corresponding rates are listed in Table A-1 (Refs. 4-7). Once the KrF^* is formed, it can radiatively decay, be quenched by the rare gases or halogen donor or be stimulated by the cavity flux. The rate constants for the dominant formation and quenching processes have been measured and checked by comparing the measured KrF^* fluorescence efficiency calculated using these rates for various Ar/Kr/F₂ mixtures. The dominant quenching reactions and rate constants used in the code are given in Table A-2. (Refs. 1 and 8). Inclusion of these processes also enables the code to predict the number densities of species which absorb at the laser wavelength. The calculated medium absorption has been compared with measured values for various mixtures and e-beam excitation levels.⁽³⁾ The three important absorbing species are F₂, F^- and Kr_2F^* . The absorption cross sections of these species⁽⁹⁻¹¹⁾ are listed in Table A-3. What remains to be verified is the ability of the code predict power extraction in the presence of intrinsic medium absorption and overall laser performance for various levels of e-beam excitation and laser gas mixtures. In the following section this ability is checked by comparing the predictions with experimental results. This comparison provides a verification of the validity of the power extraction model as well as an independent check of the various kinetics leading to laser medium gain and absorption.

3. POWER EXTRACTION

The rate of change of the cavity flux is given by

$$\frac{\partial \phi_{\pm}}{\partial t} \pm c \frac{\partial \phi_{\pm}}{\partial x} = \left(\frac{g_0 - \alpha_0}{1 + (\phi_{+} + \phi_{-})/\phi_s} \right) \alpha \phi_s \quad (A-1)$$

here $\phi_{+}(\phi_{-})$ is the cavity flux propagating in the positive (negative) x direction, ϕ_s is the saturation flux, g_0 is the small signal gain, α_0 and α are the saturable and nonsaturable absorption coefficients of the medium. Absorption by F₂, F^- , Kr_2^+ and Ar_2^+ is nonsaturable, while the absorption by Kr_2F^* is saturable with the same saturation flux as the lasing transition, if KrF^* is the precursor of the Kr_2F^* .⁽¹⁾ Equation (A-1) may be simplified by integrating over x. The temporal variation of the laser cavity flux is then determined by the following differential equation

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Figure A-1 A Block Diagram of the KrF* Laser Code

TABLE A-1

DOMINANT FORMATION RATES

			<u>Ref.</u>
Ar	$\left\{ \begin{array}{l} \rightarrow \text{Ar}^+ \\ + e_s + e \end{array} \right\}$	26.4 eV/electron-ion pair	(4)
$\vec{e} +$			
Kr	$\left\{ \begin{array}{l} \rightarrow \text{Kr}^+ \end{array} \right\}$	24.1 eV/electron ion pair	(5)
$e + \text{F}_2$	$\rightarrow \text{F}^- + \text{F}$	$3 \times 10^{-9} \text{ cm}^3/\text{s}$	
$\text{Ar}^+ + \text{F}^- + (\text{M})$	$\rightarrow \text{ArF}^* + (\text{M})$	$(10^{-7} + 10^{-6} P) \text{ cm}^3/\text{s};$	(6,7)
$\text{Kr}^+ + \text{F}^- + (\text{M})$	$\rightarrow \text{KrF}^* + (\text{M})$	$P \leq 1 \text{ atm } 1.1 \times 10^{-6} \text{ cm}^3/\text{s}$	
$\text{Ar}_2^+ + \text{F}^- +$	$\rightarrow \text{ArF}^* +$		
$\text{Kr}_2^+ + \text{F}^- +$	$\rightarrow \text{KrF}^* +$	$P = \text{pressure in atm}$	

TABLE A-2
DOMINANT QUENCHING RATES

KrF* + F ₂	→ Products	7.8 x 10 ⁻¹⁰ cm ³ /s ⁻¹	<u>Ref.</u>
KrF* + 2Kr	→ Kr ₂ F* + Kr	6.7 x 10 ⁻³¹ cm ⁶ /s ⁻¹	
KrF* + Kr + Ar	→ Kr ₂ F* + Ar	6.5 x 10 ⁻³¹ cm ⁶ /s ⁻¹	(8)
KrF* + 2Ar	→ Products	7 x 10 ⁻³² cm ⁶ /s ⁻¹	

TABLE A-3
DOMINANT ABSORBING SPECIES

<u>Species</u>	<u>Cross Section</u> (cm ²)	<u>Ref.</u>
F ₂	1.3 x 10 ⁻²⁰	(9)
F ⁻	5.6 x 10 ⁻¹⁸	(10)
Kr ₂ F	1.6 x 10 ⁻¹⁸	(11)

$$\frac{d\langle\phi_c\rangle}{dt} = c \left(\frac{g_o - \alpha_o}{1 + \langle\phi_c\rangle/\phi_s} - g_c - \alpha \right) \langle\phi_c\rangle \quad (A-2)$$

where $\langle\phi_c\rangle$ is the spatially averaged cavity flux, g_c the cavity loss. In writing Eq. (A-2) we have assumed that (1) the variations of gain and absorption along the cavity are small (12) and (2) the temporal variation of ϕ_c is small during one round trip in the cavity. The photon round trip time in the cavity was 10 ns, whereas, as will be shown subsequently, the cavity flux varied on a time scale of ~ 50 ns. The numerical solution of Eq. (A-1) was obtained by following the temporal buildup of laser cavity flux from KrF* spontaneous fluorescence. The initial value of the isotropic KrF* fluorescence was determined from the code describing the formation and quenching kinetics. The predicted shape of the laser pulse was only weakly dependent on the calculated value of initial flux, since the transit time of the flux in the laser cavity was short compared to the time scale of the e-beam current density variations.

4. EXPERIMENTAL RESULTS AND COMPARISON WITH MODEL PREDICTIONS

The data were obtained using a 1-m e-beam excited laser. A cross section of the cold cathode e-gun ($V_{eb} \approx 250$ keV), $J_{eb} \approx 12$ A/cm²) and the laser chamber are shown in Figure 4-2. The e-gun was powered by a Marx generator which produced beam pulselengths from 0.5 to 1 μ s. The cathode consisted of 1-mil thick tantalum blades placed on edge and separated by 1 cm. The anode was made from stainless steel mesh. A 2-mil thick Kapton foil supported by a 90% transmitting support structure separated the laser cavity from the e-gun vacuum chamber. The foil window had an aperture of 10 x 100 cm. A ground beam collector situated 8.5 cm from the foil collected the beam electrons after they traversed the active laser volume.

The laser chamber was constructed from Lucite which was passivated with F₂ before each series of laser experiments. An optical cavity was formed by two dielectrically-coated quartz (Dynasil) flats located outside the laser. These mirrors were separated by 1.5 m. A planar optical cavity was chosen so that the resultant transverse intensity profile of the laser beam was indicative of the e-beam energy deposition uniformity across the laser aperture. Laser cavity windows were uncoated quartz (Dynasil) flats which were accurately aligned parallel to the optical cavity mirrors.

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Figure A-2 A Cross-Sectional View of the E-Beam and Laser Cavity

A dc magnetic field, provided by two external field coils, was used to confine the e-beam against transverse beam diffusion caused by small angle scattering of the electrons by the foil and laser gas mixture. The field was directed parallel to the desired e-beam direction and measured 800 G at the center of the cavity. The e-beam power deposited was calculated from measured voltage and transmitted current traces shown in Figure A-3. The beam energy deposited was also calculated from measurements of the cavity overpressure taken after the acoustic disturbance had dissipated. These measurements were corrected for power radiated from the laser cavity and they agreed with theoretical calculations to within $\pm 5\%$.

Figure A-4 shows the comparison of the code predictions and experimental observations for a typical case. The laser mixture for this case contained 0.2% F₂, 4% Kr and 95.8% Ar at a total mixture pressure of 1.5 atm. The optical cavity output coupling was 0.66. The peak e-beam current density after attenuation by the anode screen and foil support structure was 11.5 A/cm². The top trace in Figure A-4 is the KrF* sidelight pulse shape that was monitored in the absence of laser flux by an S5 photodiode. A narrow band filter ($\Delta\lambda = 5$ nm) centered at 249 nm was placed in front of the photodiode. For comparison, the sidelight pulse shape predicted by our numerical code is also shown. The predicted curve was normalized to the experimental curve at the peak value. The center traces are a comparison of the sidelight under laser conditions.

The bottom traces in Figure A-4 show the predicted and experimental laser pulse shapes. Experimentally, the laser energy was measured using a Scientec calorimeter and the pulse shape was monitored by a photodiode. The laser power coupled out was computed by insuring that the integrated power was just the measured output energy. The area of the laser beam was determined from burn pattern measurements so that the output flux could be computed. Notice that the predicted pulse turns on about 40 ns earlier than the experimental pulse. In part this is because of the assumptions made in writing Eq. (A-2). As the laser flux increases, the sidelight is depressed. This depression, caused by the laser cavity flux, is smaller than predicted. The experimental value is 0.45 compared to the theoretical value of 0.33.

5. DISCUSSION

The experimental sidelight amplitude is expected to be larger than the predicted value because fluorescence from the upper and lower edges of the e-beam is not saturated as well as that in the center. However, because of the guide magnetic field, we expect the edge effects to be of the order 5-10% which is much less than the 30-35% difference seen experimentally. A possible explanation

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Figure A-3 Temporal Shape of the E-Beam Voltage and Current Density

Figure A-4 The Top Two Traces are a Comparison between Numerically Predicted and Measured KrF* Sidelight Pulse Shapes Under Nonlasing Conditions. The middle two traces are the sidelight pulse shapes under lasing conditions. The bottom traces are a comparison of the predicted and measured laser flux extracted.

for this difference is the finite vibrational and perhaps rotational relaxation of the upper laser level. This can be seen by analyzing the following rate equations:

$$\frac{dN_o}{dt} = - \frac{(N_o - N_{oe})}{\tau_v} - \frac{N_o}{\tau_o} - \frac{N_o}{\tau_s} \quad (A-3)$$

and

$$\frac{dN}{dt} = R - \frac{N}{\tau} - \frac{N_o}{\tau_s} \quad (A-4)$$

where N_o is the population of the zeroth level; N_{oe} is the equilibrium population of the zeroth level $N_{oe} = \theta_o N$. Where θ_o is the Boltzmann factor and $N = \sum_v N_v$, τ is the lifetime of the upper level, τ_s is the stimulated lifetime of the upper level, R is the pumping rate and τ_v is the vibrational relaxation rate.

Equations (A-3) and (A-4) can be solved under steady-state conditions to give:

$$N/N_o = \frac{\{1 + (\phi/\phi_s) \frac{1}{\beta}\}}{(1 + \phi/\phi_s)} \quad (A-5)$$

where ϕ_s is the saturation flux defined previously, N_o is the upper state population density when $\phi = 0$ and

$$\beta = 1 + \theta_o \tau/\tau_v \quad (A-6)$$

Note that for $\tau_v \rightarrow 0$ $\beta \rightarrow \infty$ and the depression of the sidelight will be $(1 + \phi/\phi_s)^{-1}$. In our model we have assumed $\tau_v = 0$. To explain the 25-35% difference in the depression we require that

$$\frac{\phi}{\phi_s} \frac{1}{\beta} \approx 0.25-0.35 \quad \text{or} \quad \beta \approx 8.5 \quad (A-7)$$

The vibrational spacing of KrF^* is estimated⁽¹³⁾ to be 310 cm^{-1} . Hence, at room temperature $\theta_o \sim 0.76$. Using the value of β given by Eq. (A-7) the ratio $\tau/\tau_v \sim 10$. The KrF^* lifetime in the laser mixture is $\sim 2.9 \text{ ns}$. Hence, we can estimate τ_v the vibrational relaxation time to be $\sim 0.29 \text{ ns}$. Assuming that Ar is responsible for the vibrational relaxation, this relaxation time corresponds

to a two-body rate constant of $\sim 10^{-10}$ cm³/s or a three-body rate constant of $\sim 2.7 \times 10^{-30}$ cm⁶/s. This finite vibrational relaxation rate will decrease the extraction efficiency by $1/\beta$ (Ref. 14) which is small for this experiment and so we will ignore its effect in the subsequent discussion.

However, it should be noted that this decrease in extraction efficiency can be substantial for very high e-beam pumping intensities ($> 10^6$ W/cm³) where the electron density is high and electron quenching of KrF* can be significantly lower τ . Also the transfer time between the B and C states of KrF have a similar effect on the extraction efficiency. Again at low pump powers the effect is small, but it could be substantial at the higher pump power.

Figure A-5 shows the predicted time dependence of the formation efficiency η_F , fluorescence efficiency η_{FL} , the laser intrinsic efficiency η_{INT} , and the laser extraction efficiency η_{EXT} . η_F is the efficiency with which KrF* is created and η_{FL} is the spontaneous power radiated divided by the e-beam power deposited. η_{EXT} is the ratio of the laser flux extracted to the number of photons available, and η_{INT} is just the product $\eta_{EXT}\eta_F$, which also is equal to the laser output power divided by the e-beam power deposited. Interestingly enough, by the end of the pulse the fluorescence efficiency is 7.5%, while the intrinsic efficiency is almost 9%. The reason that $\eta_{INT} > \eta_{FL}$ is that under lasing conditions the cavity flux is large enough to stimulate the KrF* before it can radiatively decay or be quenched by F₂ and the rare gases. Finally, η_{EXT} and η_{INT} are zero until the laser turns on, which is to be expected.

We have used our code to predict the laser output for a variety of pumping conditions. These predictions are shown in Figure A-6 which is a plot of the intrinsic efficiency as a function of e-beam current density. In plotting these curves we have kept $J_{eb}\tau_p$ constant at 7.5×10^{-6} Coulombs/cm², i.e., the pump pulse energy into the laser mixture was held constant. For this pump energy half the F₂ is burnt by the end of the pulse.

Notice that for a given laser length of < 2 m the laser efficiency first rises as the e-beam current density increases. This increase is because the absorbing species F⁻ increases only as the square root of the pump power whereas the gain increases linearly. However, the curves reach a peak and then the efficiency decreases as the pump power is increased further. The reason for this decrease is that at the higher current densities the secondary electron density increases. These secondary electrons can quench the KrF*. In the model we have assumed a quenching rate constant of 2×10^{-7} cm³/s.⁽¹⁵⁾ The electron quenching of KrF* has yet to be determined. It is partly for this reason

Figure A-5 Plots Showing the Predicted Values of η_{EXT} , η_F , η_{INT} and η_{FL} . Note that the curve for η_{EXT} is divided by two.

Figure A-6 Scaling Map for the KrF* Laser

that we have shaded the region in Figure A-6 corresponding to current densities $> 30 \text{ A/cm}^2$. The efficiency for longer laser lengths also decrease. This decrease is because the product of the absorption and the laser length is greater than unity. (14)

For the present laser device the highest output energy and the highest intrinsic efficiency were observed with a laser filling pressure of 1.7 atm (0.2% F_2 /4% Kr/95.8% Ar), an average transmitted current of 11.5 A/cm^2 (13.5 A/cm^2 peak) and an optical cavity output coupling of 0.71. Under these conditions a total output energy of 102 J was obtained with an active laser volume of 8.5 l (12 J/L) and the intrinsic efficiency was 9%. For these conditions the code predicted an intrinsic efficiency of 9.3% and an energy extraction density of 12.5 J/L.

In conclusion, we have shown that a comprehensive computer code has been developed which can predict all the important characteristics of e-beam pumped KrF lasers for e-beam current densities $< 30 \text{ A/cm}^2$. The predictive capabilities of the code have been verified by detailed comparisons with experiment. The code should prove valuable in achieving optimal laser design as well as projecting the ultimate energy scalability of these lasers. For current densities $> 30 \text{ A/cm}^2$ the electron quenching of KrF^* and other species could become important.

APPENDIX A

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$$\left\{ \left\langle \frac{\phi_c/\phi_s}{1 + \phi_c/\phi_s} \right\rangle - \frac{\langle \phi_c/\phi_s \rangle}{1 + \langle \phi_c/\phi_s \rangle} \right\} / \left\langle \frac{\phi_c/\phi_s}{1 + \phi_c/\phi_s} \right\rangle \ll 1$$

where $\langle F \rangle = 1/2 \int F(x) dx$. For the example discussed subsequently this ratio < 0.1 .

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APPENDIX B

ELECTRON DISSOCIATIVE ATTACHMENT RATE CONSTANTS FOR F_2 AND NF_3 AT 300 AND 500°K

There are now available a large number of attachment rate constants as a function of average electron energy for many halogen containing molecules. Many of these measurements were prompted by the use of these halogen compounds as donors for the production of excited rare gas halide molecules in e-beam⁽¹⁾ and discharge⁽²⁾ excited laser devices. Recently, experiments on these laser devices at AERL have indicated significant improvement in laser performance and intrinsic efficiency when these laser gas mixtures were heated to temperature near 500°K.^(3,4) Specifically, the xenon fluoride laser (350 nm) showed efficiency increases from 2.2 to over 5% when heated.⁽³⁾ Production of excited XeF^* in these experiments occurs primarily through ion channels.⁽⁵⁾ As neon is the buffer, most of the e-beam power results in the formation of neon electron-ion pairs, viz

where e_p is the primary and e_s the secondary electrons respectively. These energetic secondary electrons⁽⁶⁾ subsequently undergo a variety of energy transfer collisions and eventually attain average energies near 1 eV under typical laser operating conditions when pumped by a high energy e-beam.⁽⁷⁾ Their likely fate is to dissociatively attach to halogen containing species with reasonably fast rate constants producing the halogen negative ions, e.g.,

and

A knowledge of the electron energy dependence of these reactions is important for laser modeling and can be obtained by following the reaction progress at fixed background pressures of some non-reactive buffer (e.g., N_2) as a function of applied electric field. This buffer permits reliable modeling of the mean electron energy. Also useful, in view of the laser efficiency vs temperature

results, ⁽³⁾ is the rate constant's functional dependence on (ambient) laser mixture temperature.

We report here measurements of electron attachment to NF₃ and F₂ at various applied electric fields at 300 and 500°K in an atmosphere of N₂. These measurements were obtained by measuring the steady state current in the discharge circuit with known amounts of attaching gas present. First, measurements were made with chlorine (10 Torr) present; then, with one Torr of NF₃ or F₂ present. In this manner, the ratios of the discharge currents were obtained. Knowledge of the attachment rate constant for chlorine⁽⁸⁾ allowed us to then determine the value of the rate constants for F₂ and NF₃. Since the chlorine attachment rate constant was previously measured in the same device by observing the decay of the current after the e-beam was shut off, it was an absolute measurement requiring only a knowledge of the chlorine density (measured in absorption) and the temporal behavior of the current with a calibrated oscilloscope time base (Tektronix 7844). In this regard, the rate constants reported here are directly tied to the values for those previously reported rate constants. This technique for measuring these rates was prompted by the substantially faster rate for attachment to F₂ or NF₃, such that current decays were limited by the finite turn-off time of the e-gun (t ~ 10 ns).

The experimental apparatus is the same as used in the earlier measurements.⁽⁸⁾ The gas mixture is ionized by a beam of fast electrons having an energy of about 150 KeV and a current density near 0.3 A/cm². About 40 ns after the e-beam is fired, a capacitor that was precharged to the desired voltage is switched across the anode and cathode of the discharge cell. This discharge cell contains the reaction mixtures (e.g., 1 Torr NF₃ or 10 Torr Cl₂, etc., in 1 atm of N₂) which were premixed in teflon lined, stainless steel, 5 liter gas cylinders and allowed to mix thoroughly before use in these experiments. The N₂ (New England Oxygen, 99.997%), F₂ (A.R. Products, 98%) Cl₂ (Matheson 99.965%) and NF₃ (Rocketdyne, 98%) were used without further purification.

The data consisted of measuring the steady-state current I_s, in the discharge circuit, which, after correction for the known e-beam current, can be related directly to the electron number density. At steady state,

$$\frac{dn_e}{dt} = 0 = S - \alpha n_e n_+ - k_{ATT} N_E [H]$$

where S is the source term describing the rate of ionization by the primary e-beam current density, which was kept constant in these experiments, α the electron-ion recombination rate constant

and n_e the electron density, n_+ the positive ion density (N_4^+), ⁽⁹⁾ k_{ATT} the attachment rate constant and $[H]$ the number density of attaching specie present. Under our experimental conditions, $k_{ATT} n_e [H] \gg \alpha n_e n_+$ and therefore

$$S = k_{ATT} n_e [H]$$

or

$$S \propto k_{ATT} I_S [H]$$

since $n_e \propto I_S$. By operating with S held constant (fixed e-beam current and constant mixture pressure, i.e., N_2 at 1 atm) and measuring I_S for the various halide compounds added, the ratios for the respective attachment rate constants were obtained, e.g.,

$$\frac{k_{NF_3}}{k_{Cl_2}} = \frac{I_S^{Cl_2}}{I_S^{NF_3}} \frac{Cl_2}{NF_3}$$

These measurements were recorded as a function of applied discharge voltage (to vary the electron energy) and ambient mixture temperature)to obtain values for k_{ATT} at elevated temperature).

In addition, these steady-state data were analyzed to obtain n_e directly through estimates of the drift velocity and discharge current density. The rate constants deduced in this way agreed with the method described above to within 30%. The differences are most likely due to the accuracy with which we can measure the discharge current density (i.e., the active area).

If one utilizes a Boltzmann code ⁽¹⁰⁾ containing cross sections from swarm data for electron impact processes in N_2 as a function of applied electric field to deduce an average electron energy appropriate for these uniformly excited e-beam sustained discharges, one obtains the values for the NF_3 and F_2 rate constants as a function of average (mean) electron energy as shown in Figures B-1 - B-4. For comparison we include the recent rate constant results ^(11,12) available in the literature at room temperature and comparable electron energies. Also shown is the functional dependence of the rate constant calculated utilizing the absolute cross section data of Chantry. ⁽¹³⁾ The agreement for fluorine is within a factor of two and for NF_3 within 20%

There are no elevated temperature data with which to compare (see Figures B3 and B4). From the theoretical effort of Hall, ⁽¹⁴⁾ which suggested increased dissociative attachment rate constants from higher vibrational states of F_2 , one would predict a 10%

Figure B-1 Summary of Dissociation Attachment Rate Constants for F_2 as a Function of Average Electron Energy at 300°K ; +, Ref. 11; Δ , Ref. 12; —, Ref. 13; ●, this work.

Figure B-2 Summary of Dissociative Attachment Rate Constants for NF_3 as a Function of Average Electron Energy at 300°K :
 —, Ref. 13; ■, this work..

Figure B-3 Temperature Effect on Attachment Rate Constant for $e + F_2$: ●, 300°K, ○, 500°K

Figure B-4 Temperature Effect on Attachment Rate Constant for $e + NF_3$: $\blacksquare$, 300°K; $\square$, 500°K

increase due to Boltzmann population changes in the apparent rate constant, rather than the nearly 25% increase observed. Clearly, more information on the temperature dependence of various attachment processes would be useful.

APPENDIX B

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