KINETICS MODELING AND INTERPRETATION OF EXPERIMENTAL RESULTS FOR XeF

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SECTION I

INTRODUCTION

In this report we present a detailed discussion of a computer model of the electron beam pumped XeF laser which was developed for Los Alamos National Laboratory during FY86. In its present form, it should provide useful modeling and scaling of XeF laser experiments currently of interest. We anticipate that additional modifications will be required for long pulse ($t_p > 5 \mu\text{sec}$) simulations.

The model includes all of the important energy transport processes which contribute to the formation and quenching of XeF in the B-state (upper laser level), X-state (lower laser level), C-state (which mixes with the B-state as a loss mechanism), and the unbound (repulsive) A-state to which the C-state decays. The gain of the B to X transition is resolved for the three most important laser (vibrational) lines: (0-3) at 353 nm, (0-2) and (1-4) at 351 nm.

The sections which follow describe the kinetics model and computer code for electron beam pumped XeF lasers which use gas mixtures composed of argon (the buffer), xenon, and fluorine. The model includes the effects of three temperatures: the neutral gas, electron, and vibrational (particularly of the XeF states and the absorbers). The temperatures, energy flow, and chemistry (plasma and neutral) are calculated in a self-consistent manner; balances of energy, matter, and charge are tracked by the code and output with each print cycle.

The model includes the time history of all of the important absorbers: Ar_2^+ , Xe_2^+ , Ar^* and Xe^* (s and p states), F_2 , F^- , and Xe_2F^* .

Energy extraction in the form of lasing is computed using a modified Rigrod analysis which treats multiline lasing. This extraction model assumes a spatially uniform laser flux and is therefore a one dimensional (in time) treatment. The more general extraction model which treats non-uniform spatial effects (laser flux and pumping by a non-

uniform electron beam) is being developed, and will be integrated with excimer model in the FY87 reporting period.

The computer code was written in FORTRAN77 on a DEC MICROVAX II which uses the VAX/VMS operating system. A version of the code is also available which runs on the Los Alamos CRAY computers (1A, 1S, or XMP using CTSS).

Access to the CRAY version of the computer code and its use are described in the user's manual provided with this report. We have not provided the source listing of the code with this report, since it is readily available at Los Alamos to any validated user (in the open partition) by following the instructions in the user's manual.

SECTION II

PHYSICAL MODEL OF THE XeF(B-X) SYSTEM

A. Basic Characteristics of the XeF Laser

Laser action in XeF was first reported by Ault, Bradford and Bhaumik [1], and Brau and Ewing [2]. Both groups used high energy electrons to pump the medium, which contained argon, xenon, and a fluorine donor. Later, Champagne and Harris [3] showed that replacing the argon with neon improved the laser efficiency. Hsia *et al.* [4] have shown a dramatic increase in efficiency by heating the laser medium to 450°K. Unfortunately, these improvements can be costly to implement. The purpose of this work is to develop an XeF kinetics model to explore the advantages of low pressure, room temperature operation.

The most efficient laser transitions for the XeF laser occur on the $B \rightarrow X$ system (see Fig. 1). Because the X-state is bound, $E_B \sim 1100 \text{ cm}^{-1}$, a number of distinct lines appear due to transitions to the vibrational states in the lower level. This is quite different from the KrF laser which appears to lase on one effective line at 249 nm; this is because its ground state is repulsive in nature and does not have any bound vibrational levels. Throughout this report, we shall refer to the XeF laser at 352 nm, with the understanding that we are talking about the B-X transitions which occur at 353 nm due to the $v = 0$ to $v' = 3$ state (v is the vibrational state in the lower laser level) and the 351 nm transition due to the two transitions $v = 1$ to $v' = 4$ and $v = 0$ to $v' = 2$ which happen to overlap.

The XeF ($C \rightarrow A$) has also been made to lase at 483 nm. However, the overall efficiency is not very good, while also requiring suppression of the B-X lines and operation at high pressures $> \sim 5 \text{ atm}$ to help populate the C-state by quenching of the B-state. It is interesting to note that a comparable laser transition occurs in KrF, even though the B-state and C-state in KrF essentially overlap. Lasing on the C-A transition is inefficient because the stimulated emission cross-sections are very small ($\sigma \sim 10^{-17} \text{ cm}^2$)[5].

Lasing transitions of interest of XeF(B) to XeF(X) are at 351 nm (0-2) and (1-4) and at 353 nm (0-3). Numbers in parentheses are the vibrational quantum numbers.

Fig. 1 Simple Energy Level Diagram of the Important Electronic States of XeF

The processes which lead to the formation of the upper XeF levels are still being debated. The principal uncertainty is in deciding how much of the XeF^* formed through ion-ion recombination or the various neutral channels goes into the C-state directly. It is well known that the C-state is formed from quenching of the B-state by the neutral species (it has recently been shown that electrons probably don't contribute much to this process), so that operating at low pressure can minimize this loss. In this work we have chosen to allow all of the XeF^* to be initially in the XeF(B) state and then through quenching to form the XeF(C) state as suggested by Finn *et al.* [6].

In later sections, we will wish to calculate gain coefficients for each transition, and from these the resulting laser intensities [see Section III]. The energy of the various states in a vibrational manifold v are given by:

$$\epsilon_v = h \omega_e (v + 1/2) - h \omega_e x_e (v + 1/2)^2 \quad (1)$$

where h is Planck's constant and the ω_e and $\omega_e x_e$ coefficients for the XeF states are given in Table I (see Ref. [7]). The relative population for each vibration manifold is given by:

$$n(v) = N_0 \left[\frac{e^{-\epsilon_v / kT_v}}{1 - e^{-\epsilon_v / kT_v}} \right] \quad (2)$$

where N_0 is the total population of the appropriate electronic state of XeF, T_v is the vibrational temperature, and k is the Boltzmann constant. Generally we shall assume that T_v is in equilibrium with the neutral gas temperature.

We can now generate an expression for the small signal gain coefficient for an XeF ($\text{B} \rightarrow \text{X}$) transition for an initial vibrational level v' and final of v'' :

Table I
Spectroscopic Constants for the Important
XeF Electronic States

		state			
		X	C	B	D
T_e	(cm^{-1})	0	28023	28811.0	38051.4
ω_e	(cm^{-1})	225.40	346	309.02	350.14
$\omega_e x_e$	(cm^{-1})	10.87	2.2	1.505	1.899
B_e	(cm^{-1})	0.19326	0.170	0.14670	0.1608
R_e	($\text{\AA}$)	2.293	2.45	2.631	2.513

$$g_o(v' \rightarrow v'') = \sigma_s [n_B(v') - n_X(v'')] \quad (3)$$

where σ_s is the stimulated emission cross-section for $B \rightarrow X$ transition ($\sigma_s \sim 5A^{-2}$) [8, 9] and n_B is the number density of B states in the v' th level, n_X is the number density of X states in the v'' th level. Therefore, g_o is in cm^{-1} . This will be used in the section on energy extraction. We now turn our attention to the processes which form XeF^* in an e-beam pumped laser.

B. Basic Formation Processes for XeF^*

The electron beam transfers most of its energy into the laser medium through ionization ($\sim 60\%$) and excitation of the metastables ($\sim 15\%$); the rest is usually wasted as heat or lost radiation. A more detailed discussion of the energy channeling is given by Lorents [10]. For the sake of discussion, consider a binary mixture of Xe and F_2 . Since F_2 usually comprises less than 1% of the mix, only the energy flow through Xe will be considered. The e-beam electrons are denoted by e_p , the Xe ions by Xe^+ ; then the basic reactions for e-beam excitation are (the references for the reactions that follow are given in Table II in Section III):

where the secondary electrons produced by ionization are denoted by e_s . The species Xe^+ and Xe^* are precursors for the formation of XeF^* along with the secondary electrons e_s since they are used in the conversion of F_2 to ionic F^- in the dissociative recombination reaction:

This reaction, followed by the three-body ionic recombination reaction, leads directly to the formation of XeF^* (most probably the B but possibly the C state [6]):

where m is any third body, usually Xe or Ar. The harpoon reaction is another formation channel from the neutrals:

Additional reactions can occur which compete for the Xe^+ and Xe^* :

However these new dimer species can undergo reactions which result in formation of XeF^* :

It is tempting at this point to consider a mixture of Xe/F₂ as a serious laser mixture (similar to Kr/F₂ for the KrF* laser) because the formation efficiency η_{form} of XeF as described above is very high. The energy to form an electron - ion pair in Xe is about 22 ev, with an additional yield of 50% for the metastable Xe*. Thus,

$$\eta_{\text{form}} = \frac{(3.52 \text{ ev}) (1.5)}{22 \text{ ev}} = 24\% \quad (14)$$

Unfortunately, this high formation efficiency does not convert into efficient lasing action because the Xe₂⁺ ion is a strong broad band absorber of the 352 nm radiation, laser efficiencies of only 1% or less have been reported [11]. An additional more practical problem is the very high expense of the xenon gas. For these reasons, the xenon is always diluted to about 1% in a buffer gas of argon or neon. Some attempts to use krypton as a buffer have been reported but Kr₂⁺ is also a strong absorber at 352 nm (see Fig. 2) so that no benefit of the higher formation efficiency is realized. The formation of XeF* is less direct in the presence of an Ar buffer, because there are a similar set of reactions to those above with Ar replacing Xe at the appropriate places. These reactions result in the creation of the new species Ar*, Ar⁺, Ar₂^{*}, Ar₂⁺ and ArF*. However there are a series of replacement reactions that occur, such as:

Fig. 2 Theoretical Dimer Ion Absorption Coefficients Due to
H. H. Michels *et al.* (J. Chem. Phys. 71 5053 (1979))

which produce the excited Xe containing species. In addition, two new species ArXe^* and ArXe^+ are formed:

which react with F_2 and F^- , respectively, to form XeF :

The principal disadvantage of the Ar buffer gas is that it takes more energy to form the Ar states. Thus the formation efficiency will be lower with Ar present. We also need to include the fact that the branching ratio for Ar^* harpoon reaction is less than 100% [12]. (The measured value is about 60%.) This gives a formation efficiency of about 16%. A further complication occurs due to the formation of Ar_2^+ , which is an absorber at 352 nm (see Fig. 2). This will be discussed in the next section.

C. Quenching and Absorption Losses

We have shown that the XeF(B) states can be formed efficiently. The extraction of B-state energy is complicated by a number of processes, which compete with stimulated emission of this state. The first process is termed quenching, because the reaction results in the destruction of the XeF^* without the emission of a photon. The principal quenchers are Xe and Ar in the three body recombination reactions [8]:

The second reaction is quickly followed by:

An additional complication of these reactions is the formation of the trimer Xe_3F^* , which is an absorber of 352 nm. The other important quenchers are diatomic fluorine F_2 , atomic fluorine F , and the secondary electrons e_s ; their quenching reactions are:

The most significant feature to note in these reactions is that F_2 and e_s are essential species in the formation of XeF^* . The F species is also formed in the reaction chain leading to XeF^* . These facts make it impossible to avoid quenching as a loss mechanism. Quenching by F_2 is part of the reason that the XeF laser mixture is composed of very little

F₂. However, the species F₂ is also the key species controlling the electron density through the dissociative attachment process:

since ionic recombination of the dimer ions is relatively slow:

The amount of F₂ required for a given operating point will depend on two factors: the pump rate which will determine the rate of production of the secondary electrons, and the duration of the pumping, since F₂ is used up or burned. The combination of these two factors is the specific energy loading (joules per cm³), that is the pump rate (watts per cm³) and the pulse duration (seconds). For efficient laser operation in DOD devices, usually an F₂ density of $1.0 \times 10^{17} \text{ cm}^{-3}$ is required per 100 joules per litre of input energy. This density of F₂ represents the tradeoff between F₂ and e_s quenching. The process is dynamic because F₂ quenching will dominate at the beginning of the pumping, and then shift to e_s and F dominated quenching, since the F₂ density is decreasing, while the electron and F densities are increasing. In long pulse XeF lasers it may be possible (or necessary) to recycle the F atoms.

Another important characteristic of F₂ is that it absorbs the 352 nm laser light; this makes minimizing its density even more important, because absorption will limit the length of the laser cavity, which impacts the flux values which can be considered in a design. F₂

is not the only species which absorbs the laser photons. All of the important ionic precursor species (Ar_2^+ , Xe_2^+ , F^-), some of the excited neutral species (Ar^{**} , Xe^{**} , Xe_2^* , Ar_2^* , ArXe^*), and the triatomic Xe_2F^* have appreciable cross-sections and impact the total absorption present in a given operating point. The absorber Xe_2F^* is partially saturable since it is formed from some of the XeF^* state which can be destroyed in the lasing process. The other absorbers are formed as essential precursors to XeF^* and are thus non-saturable; they can only be minimized through a judicious choice of the laser mix and pump rate.

The important issues to remember are: (1) the principal absorbers in the laser medium are the species which are also the essential precursors for forming XeF^* , (2) the absorbers limit the flux which can be present in the laser medium, and (3) that quenching of XeF^* can be a serious loss mechanism for a low flux laser. A quantitative discussion of these issues will be presented in the next section on energy extraction and fluorescence.

D. Energy Extraction

In this section we shall present a microscopic view of the stimulated emission process in rate equation form; this will facilitate a discussion on the relative time scales for lasing, quenching, and fluorescence. In addition, we shall recast the microscopic equations into the more familiar macroscopic equations to lead into the discussions of the following sections.

The basic reaction for the stimulated emission of a photon $h\nu$, whose number density we shall denote by N_ν , from XeF^* is:

This process is an amplification of the original photon, because the stimulated photon has the same phase and direction of propagation as the original photon, and is the reason the factor of 2 is appropriate on the right side of the equation. The above reaction is

characterized by the stimulated emission cross-section σ_s which has an effective value of $5.33 \times 10^{-16} \text{ cm}^2$ based on a fluorescence lifetime of 12.0 nanosecs.[13, 14]

In addition to quenching as a loss mechanism, the XeF^* states can spontaneously decay (fluorescence) with a lifetime of 12.0 nanosec, the flux in an oscillator actually builds-up from the fluorescence. The spontaneously emitted photons are undesirable because they are emitted in all directions with random phase. This is sometimes called optical noise; the rate of this reaction:

is just the inverse of the lifetime τ_R of the state:

$$\tau_R^{-1} = 8.00 \times 10^7 \text{ sec}^{-1} \quad (31)$$

The rate for a typical three body quenching reaction at 2 atmospheres is:

$$\tau_Q^{-1} \sim 1.6 \times 10^8 \text{ sec}^{-1} \quad (32)$$

which is comparable. Detailed calculations with the XeF^* kinetics code show that the upper state lifetime τ_u is about 4.2 nanosec when all processes, excluding lasing, are summed together. From this quantity τ_u and the stimulated emission cross-section σ_s , the

photon flux ϕ_{sat} necessary to cause the same combined loss of XeF^* can be determined:

$$\phi_{\text{sat}} = \frac{1\nu}{\sigma\tau} \quad (33)$$

or

$$\phi_{\text{sat}} = 282 \text{ kw/cm}^2 \quad (34)$$

Since the rates which determine the upper state lifetime can depend on temperature and on the species density, the value for the saturation flux can vary with time, pump rate, or gas mixture.

The absorption process is the exact opposite of lasing, since the photon in the reactants is lost and no other photons are formed. Some typical photoabsorption reactions for the XeF^* laser are:

In each reaction there is a rate coefficient which is the product of the cross-section and the speed of light. If we combine all of these reactions together and define the absorption coefficient α as the sum of the absorption processes, we can write the equations for the photon density N_ν and XeF^* in terms of the quantities discussed in this section:

$$\frac{dN}{dt} = c\sigma_s \left(\text{XeF}^*(\text{B}) - \text{XeF}(\text{X}) \right) N_v - c\alpha N_v + \frac{\text{XeF}(\text{B})}{\tau_r} \quad (38)$$

$$\frac{d[\text{XeF}(\text{B})]}{dt} = [\text{formation}] - \frac{[\text{XeF}(\text{B})]}{\tau_u} - c\sigma_s \left(\text{XeF}(\text{B}) - \text{XeF}(\text{X}) \right) \quad (39)$$

The first equation can be rewritten by defining the small-signal gain coefficient g_0 , the laser flux ϕ_L , and E_v is the photon energy.

$$\frac{d\phi_L}{dt} = (g_0 - \alpha) \phi_L + \frac{\Delta\Omega}{4\pi} \cdot \frac{\tau_u}{\tau_r} g_0 \cdot c \cdot \phi_s \quad (40)$$

where $\phi_s = \frac{E_v}{\sigma_s \tau_u}$ is the saturation flux

and $\Delta\Omega$ is the solid angle that the laser cavity intercepts. If the photons are being amplified in an oscillator there is an additional loss term which must be subtracted from the equation to account for photons which are outcoupled or absorbed in the mirrors; this quantity is called the cavity loss g_c . It is calculated from the equation:

$$g_c = \frac{1}{2l_c} \ln(M^2) \quad (41)$$

where l_c is the distance between the mirrors and M is the magnification of the resonator. This discussion represents a simplification of the usual macroscopic case, because the transition from our microscopic equations implicitly assumed that the XeF^* (the gain) and the photon density (the flux) were uniform throughout the pumped medium. For some

devices, such as the Tahoma laser at Spectra Technology, Inc., where the experiments discussed in Section IV were performed, this is a reasonable approximation which we call the uniform flux approximation. We shall also need to generalize this discussion since the XeF is a multiline laser. This will be presented in the next section.

SECTION III

BASIC EQUATIONS

The physics equations are given in this section. These are the same equations upon which the computer code is based for a simulation of an electron beam pumped XeF laser.

We begin by choosing a laser gas mixture. The input parameters are the partial fractions f_i (of the mixture composed of argon, xenon and fluorine). Because excimer lasers are sometimes operated at initial temperatures above room temperature, it is more useful to define the amount of gas present in amagats N_A rather than pressure, since pressure scales with temperature. One amagat is the density of a gas at STP and corresponds to Loschmitt's number $N_L = 2.69 \times 10^{19}$ particles per cm^3 . This is equivalent to Avagadro's number 6.023×10^{23} in a standard volume of 22.4 litres. The determination of the initial gas densities n_i is then a straightforward calculation;

$$n_i = f_i \cdot N_A \cdot N_L \quad (42)$$

The pressure P is then calculated from the ideal gas law;

$$P = n_O k T_g \quad (43)$$

where n_O is the total density, k is the Boltzmann constant $1.38 \times 10^{-23} \text{ J/}^\circ\text{K}$ and T_g is the gas temperature in $^\circ\text{K}$.

In a laser experiment the optical mirrors determine the characteristic cavity loss of the oscillator g_c , also called the cavity gain since this is the threshold gain required for lasing. The laser flux ϕ_L , for the uniform flux model, is computed from the differential equation:

$$\frac{d\phi_L}{dt} = c (g - g_c) \phi_L \quad (44)$$

where c is the speed of light and g is the net gain in the cavity, i.e., small signal gain g_0 less the loss in the laser medium. The term g_c is determined from the reflectivities of the laser mirrors R_1 R_2 and the distance between them [15]:

$$g_c = -\frac{1}{2l_c} \ln (R_1 R_2) \quad (45)$$

If R_1 is the output mirror reflectivity, the above equation can be written in the alternate forms:

$$\begin{aligned} g_c &= -\frac{1}{2l_c} \ln [R_2(1 - F_1)] \\ &= \frac{1}{2l_c} \ln [R_2(M^2)] \end{aligned} \quad (46)$$

where F_1 is the output coupling fraction and M is the magnification of the resonator cavity.

We need to modify the above analysis to include the multiline lasing character of the XeF laser. Because of the number of vibrational states involved, this can become a major task. The reason is that the lasing process by nature drives the upper and lower levels out of vibrational equilibrium. A complete treatment would require a relaxation analysis of how the energy is redistributed. We can avoid this complication by realizing that the relaxation to Boltzmann equilibrium will be dominated by two body collisions with the argon buffer gas which will occur on a time scale that is a fraction of a nanosecond for laser

gas pressures of two atmospheres or more. The lasing process will occur on a time scale of 1 to 2 nanoseconds for typical laboratory lasing conditions (i.e., a flux of 3-4 times the saturation flux). Hence, it is a reasonable approximation to assume that the distribution relax infinitely fast so that they are always in a Boltzmann equilibrium, and just make allowance for the depletion of the total XeF(B) manifold and the increase in the total XeF(X) manifold. The gain coefficient for the 353 nm transition is designated $g(353)$ and can be computed from

$$g(353) = \sigma_s [N(B, v=0) - N(X, v=3)] \quad (47)$$

where $N(B, v=0)$ is the fraction of the total XeF(B) population in the $v=0$ vibrational state. Similarly, $N(X, v=3)$ is the fraction of the total XeF(X) population in the $v=3$ vibrational state. If we call $\phi(353)$ the laser cavity flux at 353 nm and designate $s(353)$ as the source of the initial flux (either captured fluorescence or an injected flux), the equations for the buildup of flux, X-state population, and depletion of the XeF(B) state are:

$$\frac{d\phi(353)}{dt} = c [g(353) - g_c] \phi(353) + s(353) \quad (48)$$

$$\frac{d[XeF(X)]}{dt} = \frac{[XeF(B)]}{\tau_u} + \frac{g(353) \phi(353)}{h\nu} - \frac{[XeF(X)]}{\tau_Q} \quad (49)$$

$$\frac{d[XeF(B)]}{dt} = R - \frac{[XeF(B)]}{\tau_u} - \frac{g(353) \phi(353)}{h\nu} \quad (50)$$

where R is the rate of formation of the XeF(B) state, τ_u is the total quenching rate of the B-state (including fluorescence), τ_Q is the total quenching rate of the X-state, and $h\nu$ is the photon energy for 353 nm. When the lasing occurs on the 351 nm transition, we get a similar set of equations if we assume that the $v' = 1$ to $v'' = 4$ transition overlaps the $v' = 0$ to $v'' = 2$ transition, which is a good approximation. The gain coefficient $g(351)$ becomes:

$$g(351) = \sigma_s [N(\text{B}, v = 0) - N(\text{X}, v = 2)] + \sigma_s [N(\text{B}, v = 1) - N(\text{X}, v = 4)] \quad (51)$$

where $N(\text{B}, v = 1)$ is the fraction of XeF(B) population in the $v = 1$ vibrational state, $N(\text{X}, v = 4)$ is the fraction of the XeF(X) states in the $v = 4$ vibrational level and so forth. The density fractions are determined by the method described in Section II. When there is lasing on both lines simultaneously, the equations become:

$$\frac{d\phi(353)}{dt} = c [g(353) - g_c] \phi(353) + s(353) \quad (52)$$

$$\frac{d\phi(351)}{dt} = c [g(351) - g_c] \phi(351) + s(351) \quad (53)$$

$$\frac{d[\text{XeF(X)}]}{dt} = \frac{[\text{XeF(B)}]}{\tau_u} + \frac{g(353)\phi(353)}{h\nu} + \frac{g(351)\phi(351)}{h\nu'} - \frac{[\text{XeF(X)}]}{\tau_Q} \quad (54)$$

$$\frac{d[\text{XeF(B)}]}{dt} = R - \frac{[\text{XeF(B)}]}{\tau_u} - \frac{g(353)\phi(353)}{h\nu} - \frac{g(351)\phi(351)}{h\nu'} \quad (55)$$

where all quantities are as above and $h\nu'$ is the photon energy at 351 nm. The equations above constitute the multiline extraction model in the present XeF code.

The pumping rate of the electron beam is a complicated function of the mixture composition and density, the energy of the electrons, and the current density of the beam. An accurate determination of this information for a given experimental setup requires the use of a Monte Carlo code (see Appendix B), which is relatively expensive to use (> 10 min per calculation on the CRAY and > 400 minutes on a VAX 11/780). Fortunately, Lorents [10] has developed a theory for the energy partitioning within mixtures of noble gases, and Berger and Seltzer [17] have tabulated the relative stopping powers of the noble gas as well as some other useful gases and materials. For mixtures of Ar/Xe, we neglect F₂ because it is less than 1% of the mixture, the relative partitioning η_i of e-beam deposited energy is;

$$\eta_{\text{Ar}} = [\text{Ar}] / ([\text{Ar}] + [\text{Xe}]) \quad (56)$$

$$\eta_{\text{Xe}} = 2.62[\text{Xe}] / ([\text{Ar}] + [\text{Xe}]) \quad (57)$$

Where [Ar] is the argon density, [Xe] is the xenon density and the factor of 2.62 is due to Berger and Seltzer, it shows that xenon has nearly four times the stopping power of argon. Within the partitioning of power through each species, there is an additional partitioning into ionization and metastable yields; for argon there is a 28% yield of metastables, for xenon 50% [10]. The measured values for the energy required to produce an electron-ion pair in argon is 26.2 eV and for xenon 21.9 eV so that energy deposition through xenon is much more efficient than through argon, because more excited states are produced (1.50 vs 1.28) for less energy. There is also wasted energy in this process; for example, for xenon the ionic state is at 12.4 eV, the metastable state is at 8.5 eV, so that 16.65 eV is actually

usable of the 21.9 eV required to make the electron-ion pair. The rest of the energy goes into the secondary electrons. Most of this energy is then lost as heat into the neutral gas through momentum transfer, which cools the electrons to about 1.0 eV (Ramsauer effect). We have verified this through time dependent Boltzmann code calculations [18].

The reaction package we have developed for the XeF* kinetics code is given in Table II and a schematic of the energy flow is given in Fig. 3. It is comprised of 37 species within a set of 144 reactions; the last 5 reactions which refer to electron beam processes are handled as a special case within the code, the first 139 reactions use a very general format for the reaction rate coefficients RC(i) of the ith reaction:

$$RC(i) = A \cdot T^{\alpha} \exp\left(-\frac{38.68\beta}{T}\right) T_e^{\alpha_1} \exp\left(-\frac{\beta_1}{T_e}\right) \quad (58)$$

Where A, α , β , α_1 , and β_1 are the parameters which are read in from the XEFMODEL file, the variable T is the neutral gas temperature divided by 300 (room temperature) and T_e is the electron temperature in electron volts. The amount of flexibility is probably more than the user would be able to specify, so that in practice usually one or two parameters are used. An important point to remember when considering the photoabsorption or stimulated emission processes is that the cross-sections must be multiplied by the speed of light and entered in the A parameter, since the code treats all the reactions as kinetic processes.

Once the reaction rates are calculated the species derivatives are computed by determining the rates of change of all the species in each reaction and adding for a formation process F, or subtracting for a depletion process D, the appropriate rate of change from the equation for the N_i species' derivatives:

$$\frac{dN_i}{dt} = \sum_j F_{ji} - \sum_k D_{ki} \quad (59)$$

Table II Rate Constants for the XeF Ar/Xe/F₂ Model

Reaction	Rate constant	Reference
<u>Neutral reaction</u>		
$\text{Ar}^* + 2\text{Ar} \rightarrow \text{Ar}_2^* + \text{Ar}$	3.3 E-32 cm ⁶ /s	[19]
$\text{Xe}^* + 2\text{Ar} \rightarrow \text{ArXe}^* + \text{Ar}$	1.0 E-33	[19]
$\text{Xe}^* + \text{Xe} + \text{Ar} \rightarrow \text{Xe}_2^* + \text{Ar}$	2.3 E-32	[20]
$\text{Xe}^* + 2\text{Xe} \rightarrow \text{Xe}_2^* + \text{Xe}$	1.7 E-32 for ¹ Σ state	[21]
	3.0 E-32 for ³ Σ state	[21]
$\text{ArXe}^* + \text{Xe} \rightarrow \text{Xe}_2^* + \text{Ar}$	1.0 E-10 cm ³ /s	[22]
$\text{Ar}^{**} + \text{Ar} \rightarrow \text{Ar}^* + \text{Ar}$	1.0 E-10	[23]
$\text{Xe}^{**} + \text{Ar} \rightarrow \text{Xe}^* + \text{Ar}$	1.0 E-10	[24]
$\text{Xe}^{**} + \text{Xe} \rightarrow \text{Xe}^* + \text{Xe}$	1.0 E-10	[24]
$\text{Ar}^* + \text{Xe} \rightarrow \text{Xe}^* + \text{Ar}$	2.1 E-10	[25]
$\text{Ar}^* + \text{Xe} + (\text{Ar}) \rightarrow \text{ArXe}^* + (\text{Ar})$	5.0 E-11	Estimation
$\text{Ar}_2^* + \text{Xe} \rightarrow \text{Xe}^* + 2\text{Ar}$	2.4 E-10	[19]
$\text{Ar}_2^* + \text{Xe} \rightarrow \text{ArXe}^* + \text{Ar}$	5.0 E-11	Estimation
$\text{Xe}^* + \text{Xe} \rightarrow 2\text{Xe}$	3.5 E-15	[26]
$\text{Xe}^{**} + 2\text{Xe} \rightarrow \text{Xe}_2^{**} + \text{Xe}$	1.0 E-31 cm ⁶ /s	[27]
$\text{Ar}^{**} + 2\text{Ar} \rightarrow \text{Ar}_2^{**} + \text{Ar}$	1.0 E-31	[27]

Table II (cont'd)

Reaction	Rate constant	Reference
$\text{Xe}_2^{**} + \text{M} \rightarrow \text{Xe}^* + \text{Xe} + \text{M}$	$1.0 \text{ E-11 cm}^3/\text{s}$	[27]
$\text{Ar}_2^{**} + \text{M} \rightarrow \text{Ar}^* + \text{Ar} + \text{M}$	1.0 E-11	[27]
$\text{Xe}_2^* (^1\Sigma) + \text{M} \rightarrow \text{Xe}_2^* (^3\Sigma) + \text{M}$	1.3 E-13	[28]
M: Rare-gas atom		
<u>Penning ionization</u>		
$\text{Xe}^* + \text{Xe}^* \rightarrow \text{Xe}^+ + \text{Xe} + \text{e}$	$5.0 \text{ E-10 cm}^3/\text{s}$	[29]
$\text{Xe}_2^* + \text{Xe}_2^* \rightarrow \text{Xe}_2^+ + 2\text{Xe} + \text{e}$	5.0 E-10	[29]
$\text{Ar}^* + \text{Ar}^* \rightarrow \text{Ar}^+ + \text{Ar} + \text{e}$	5.0 E-10	[30]
$\text{Ar}_2^* + \text{Ar}_2^* \rightarrow \text{Ar}_2^+ + 2\text{Ar} + \text{e}$	5.0 E-10	[19]
<u>Charge transfer</u>		
$\text{Ar}^+ + 2\text{Ar} \rightarrow \text{Ar}_2^+ + \text{Ar}$	$2.5 \text{ E-31 cm}^6/\text{s}$	[27]
$\text{Ar}^+ + \text{Xe} + \text{Ar} \rightarrow \text{ArXe}^+ + \text{Ar}$	1.0 E-31	[31]
$\text{Xe}^+ + 2\text{Ar} \rightarrow \text{ArXe}^+ + \text{Ar}$	1.0 E-31	[32]
$\text{Xe}^+ + \text{Xe} + \text{Ar} \rightarrow \text{Xe}_2^+ + \text{Ar}$	2.0 E-31	[33]
$\text{Xe}^+ + 2\text{Xe} \rightarrow \text{Xe}_2^+ + \text{Xe}$	2.5 E-31	[27]
$\text{Ar}_2^+ + \text{Xe} \rightarrow \text{Xe}^+ + 2\text{Ar}$	$1.0 \text{ E-9 cm}^3/\text{s}$	[33]
$\text{ArXe}^+ + \text{Xe} \rightarrow \text{Xe}_2^+ + \text{Ar}$	1.0 E-11	[35]

Table II (cont'd)

Reaction	Rate constant	Reference
$\text{ArXe}^+ + \text{Xe} \rightarrow \text{Xe}^+ + \text{Ar} + \text{Xe}$	5.0 E-10	[35]
<u>Dimer-ion - electron recombination</u>		
$\text{Ar}_2^+ + e \rightarrow \text{Ar}^{**} + \text{Ar}$	$8.8 \text{ E-8 } T_e^{-0.67} \text{ cm}^3/\text{s}$	[36]
$\text{Xe}_2^+ + e \rightarrow \text{Xe}^{**} + \text{Xe}$	$2.0 \text{ E-7 } T_e^{-0.5}$	[36]
$\text{ArXe}^+ + e \rightarrow \text{Xe}^{**} + \text{Ar}$	$2.0 \text{ E-7 } T_e^{-0.5}$	[32]
Te: mean electron temperature (in electron volts)		
<u>Radiation</u>		
$\text{Ar}_2^* \rightarrow 2\text{Ar} + h\nu$	6.00 E+7 s^{-1}	[37]
$\text{Xe}_2^* \rightarrow 2\text{Xe} + h\nu$	1.82 E+8 for $^1\Sigma$ state	[37]
	1.00 E+7 for $^3\Sigma$ state	[37]
$\text{ArXe}^* \rightarrow \text{Ar} + \text{Xe} + h\nu$	5.00 E+7	Estimate
$\text{Xe}^{**} \rightarrow \text{Xe}^* + h\nu$	1.49 E+7	[34]
$\text{ArF}^* \rightarrow \text{Ar} + \text{F} + h\nu$	2.50 E+8	[38]
$\text{XeF}^* (\text{B}) \rightarrow \text{XeF} (\text{X}) + h\nu$	8.00 E+7	[39]
$\text{XeF}^* (\text{C}) \rightarrow \text{Xe} + \text{F} + h\nu$	1.1 E+7	[39]
$\text{XeF}^* (\text{B}) + h\nu \rightarrow \text{XeF} (\text{X}) + 2h\nu$	5.33 E-16 cm^2	See text
$\text{Xe}_2\text{F}^* \rightarrow 2\text{Xe} + \text{F} + h\nu$	4.00 E+6 s^{-1}	[40]

Table II (cont'd)

Reaction	Rate constant	Reference
<u>F₂ - electron reaction</u>		
$F_2 + e \rightarrow F + F^-$	1.3 - 2.0 E-9 cm ³ /sec (function of F ₂ density)	Time dependent
$F_2 + e \rightarrow F + F + e$	2.18 E-9 cm ³ /sec	Boltzmann calculation
<u>Three-body ion-ion recombination</u>		
$Ar^+ + F^- + Ar \rightarrow ArF^* + Ar$		
$Ar_2^+ + F^- + Ar \rightarrow ArF^* + 2Ar$		
$Xe^+ + F^- + Ar \rightarrow XeF^* + Ar$	2 E-6 cm ³ /sec	[effective two body rate] [41]
$Xe_2^+ + F^- + Ar \rightarrow XeF^* + Xe + Ar$		
$ArXe^+ + F^- + Ar \rightarrow XeF^* + 2Ar$		
<u>Xe - F₂ reaction</u>		
$Xe^* + F_2 \rightarrow XeF^* + F$	2.0 E-10	[19]
$Xe^{**} + F_2 \rightarrow XeF^* + F$	5.6 E-10	[19]

Table II (cont'd)

Reaction	Rate constant	Reference
<u>XeF* relaxation</u>		
$\text{XeF}^* + \text{Xe} \rightarrow \text{Xe} + \text{F} + \text{Xe}$	$2.1 \text{ E-12 cm}^3/\text{s}$	[42, 13, 43]
$\text{XeF}^* + \text{Ar} \rightarrow \text{Xe} + \text{F} + \text{Ar}$	2.0 E-13	[42, 13, 43]
$\text{XeF}^* + \text{F}_2 \rightarrow \text{Xe} + \text{F} + \text{F}_2$	8.0 E-10	[42, 13, 43]
$\text{XeF}^* + \text{e} \rightarrow \text{Xe} + \text{F} + \text{e}$	2.0 E-7	[42, 13, 43]
$\text{XeF}^* + 2\text{Ar} \rightarrow \text{products}$	$3.0 \text{ E-33 cm}^6/\text{s}$	[42, 13, 43]
$\text{XeF}^* + \text{Xe} + \text{Ar} \rightarrow \text{Xe}_2\text{F}^* + \text{Ar}$	1.5 E-31	[42, 13, 43]
$\text{XeF}^* + 2\text{Xe} \rightarrow \text{Xe}_2\text{F}^* + \text{Xe}$	4.0 E-31	[42, 13, 43]
$\text{XeF (X)} + \text{Ar} \rightarrow \text{Xe} + \text{F} + \text{Ar}$		
$\text{XeF (X)} + \text{Xe} \rightarrow \text{Xe} + \text{F} + \text{Xe}$	$\tau = 200 \text{ ps}$	Estimate See Ref. [58]
$\text{XeF (X)} + \text{F}_2 \rightarrow \text{Xe} + \text{F} + \text{F}_2$		
<u>Xe₂F* relaxation</u>		
$\text{Xe}_2\text{F}^* + \text{Ar} \rightarrow \text{products}$	$3.0 \text{ E-14 cm}^3/\text{s}$	[43]
$\text{Xe}_2\text{F}^* + \text{Xe} \rightarrow \text{products}$	5.0 E-13	[43]
$\text{Xe}_2\text{F}^* + \text{e} \rightarrow 2\text{Xe} + \text{F} + \text{e}$	2.0 E-7	[43]

Table II (cont'd)

Reaction	Rate constant	Reference
<u>Absorption</u> (rate constant is the cross-section times the speed of light)		
$\text{Ar}_2^+ + h\nu \rightarrow \text{Ar}^+ + \text{Ar}$	4.3 E-7	[44]
$\text{Xe}_2^+ + h\nu \rightarrow \text{Xe}^+ + \text{Xe}$	1.032 E-6	[44]
$\text{ArXe}^+ + h\nu \rightarrow \text{Ar}^+ + \text{Xe}$	4.5 E-7	[45]
$\text{Ar}_2^* + h\nu \rightarrow \text{Ar}_2^+ + e$	4.2 E-7	[46]
$\text{Xe}_2^* + h\nu \rightarrow \text{Xe}_2^+ + e$	4.2 E-7	[47]
$\text{ArXe}^* + h\nu \rightarrow \text{ArXe}^+ + e$	4.5 E-7	[48]
$\text{Xe}^{**} + h\nu \rightarrow \text{Xe}^+ + e$	3.0 E-7	[49]
$\text{Xe}^* + h\nu \rightarrow \text{Xe}^+ + e$	3.0 E-9	[50]
$\text{Ar}^{**} + h\nu \rightarrow \text{Ar}^+ + e$	1.8 E-7	[49]
$\text{F}^- + h\nu \rightarrow \text{F} + e$	6.0 E-8	[51]
$\text{F}_2 + h\nu \rightarrow \text{F} + \text{F}$	1.98 E-10	[52]
$\text{Xe}_2\text{F}^* + h\nu \rightarrow \text{products}$	1.8 E-7	[53]

Table II (cont'd)

Reaction	Rate constant	Reference
<u>Electron reaction</u>	($T_e = 1.25$ ev)	
$\text{Ar} + e \leftrightarrow \text{Ar}^* + e$	3.00 E-13 1.46 E-9	
$\text{Ar} + e \leftrightarrow \text{Ar}^{**} + e$	1.27 E-13 5.11 E-10	
$\text{Ar}^* + e \leftrightarrow \text{Ar}^{**} + e$	2.43 E-7 9.19 E-7	
$\text{Ar} + e \rightarrow \text{Ar}^+ + 2e$	1.01 E-15	
$\text{Ar}^* + e \rightarrow \text{Ar}^+ + 2e$	4.49 E-10	
$\text{Ar}^{**} + e \rightarrow \text{Ar}^+ + 2e$	4.95 E-9	
$\text{Ar}_2^* + e \rightarrow \text{Ar}_2^+ + 2e$	4.95 E-9	Time dependent Boltzmann code calculation [see text]
$\text{Xe} + e \leftrightarrow \text{Xe}^* + e$	2.16 E-12 7.23 E-8	
$\text{Xe} + e \leftrightarrow \text{Xe}^{**} + e$	2.15 E-12 1.68 E-10	
$\text{Xe}^* + e \leftrightarrow \text{Xe}^{**} + e$	3.96 E-7 1.88 E-7	
$\text{Xe} + e \rightarrow \text{Xe}^+ + 2e$	1.37 E-12	
$\text{Xe}^* + e \rightarrow \text{Xe}^+ + 2e$	6.16 E-10	
$\text{Xe}^{**} + e \rightarrow \text{Xe}^+ + 2e$	2.26 E-7	
$\text{Xe}_2^* + e \rightarrow \text{Xe}_2^+ + 2e$	6.26 E-10	

Table II (cont'd)

Reaction	Rate constant	Reference
$\text{ArXe}^* + e \rightarrow \text{ArXe}^+ + 2e$	6.26 E-10	
$\text{Xe}_2^* + e \rightarrow \text{Xe}_2^{**} + e$	1.0 E-7 cm ³ /s	[27]
$\text{Xe}_2^* (^3\Sigma) + e \rightarrow \text{Xe}_2^* (^1\Sigma) + e$	8.8 E-8	[27]
$\text{Xe}_2^* (^1\Sigma) + e \rightarrow \text{Xe}_2^* (^3\Sigma) + e$	3.0 E-7	[27]
$\text{Xe}_2^{**} + e \rightarrow \text{Xe}_2^* (^3\Sigma) + e$	1.0 E-6	[27]
$\text{Xe}_2^{**} + e \rightarrow \text{Xe}_2^* (^1\Sigma) + e$	3.0 E-7	[27]
$\text{Ar}_2^* + e \rightarrow \text{Ar}_2^{**} + e$	1.0 E-7	[27]
$\text{Ar}_2^{**} + e \rightarrow \text{Ar}_2^* + e$	1.0 E-6	[27]
$\text{Xe}_2^* + e \rightarrow 2\text{Xe} + e$	1.0 E-9	[27]
$\text{Ar}_2^* + e \rightarrow 2\text{Ar} + e$	1.0 E-9	[27]
$\text{ArXe}^* + e \rightarrow \text{Ar} + \text{Xe} + e$	1.0 E-9	[34]

where the sums are over all the contributing reactions. Since the equations are coupled first order equations, they can be solved using a variety of numerical integration schemes. The integrator presently being used is a modified Euler scheme described in the paper by Johnson et al. [15]; in this paper there is also a discussion of tests of other integration schemes.

In addition to integrating the species densities, we also integrate the input pump power, the energy lost to fluorescence E_{rad} , temperatures, T_g and T_e , and the laser output flux I_{out} . These equations are shown below.

$$E_{\text{dep}} = \int P_{\text{eb}} dt \quad (60)$$

where P_{eb} is the e-beam pump power.

$$E_{\text{rad}} = \sum_i \int \epsilon_i \frac{dN_i}{dt} dt \quad (61)$$

(the sum is over all of the radiating species) where ϵ_i is the energy of the photon of the i th radiating species

$$T_g = \frac{2}{3} \sum_i \frac{\Delta E_i \cdot CT(i)}{c_v} \quad (62)$$

where ΔE_i is the net energy of the i th reaction whose rate in the forward direction is $CT(i)$, c_v is the heat capacity at constant volume of the gas. The output laser flux ϕ_{out} is given by [15] (for an alternate expression see Ref. [16]):

$$\phi_{\text{out}} = \left(\frac{1 - R_1 R_2}{1 + R_1 R_2} \right) \phi_L \quad (63)$$

where ϕ_L is the laser cavity flux, R_1 and R_2 are the mirror reflectivities, the output energy fluence is then,

$$E_{\text{out}} = \int \phi_{\text{out}} dt \quad (64)$$

For the multiline case there is an equivalent set of equations for each flux.

Some additional quantities of interest are: the performance parameters for XeF* formation efficiency η_{form}

$$\eta_{\text{form}} = \frac{\epsilon_{\text{XeF}^*} \cdot \sum_i \text{CT}(i)}{P_{\text{eb}}} \quad (65)$$

where ϵ_{XeF^*} is the energy of the XeF photon (3.52 eV) and the summation is over all the formation reactions of rate CT(i), P_{eb} is the e-beam input power, and the instantaneous laser efficiency η_P :

$$\eta_P = \frac{\phi_{\text{out}} \cdot A}{P_{\text{eb}} \cdot V} \quad (66)$$

where ϕ_{out} is laser output flux, P_{eb} is the beam power, A is the beam area, V is the laser volume, and the total laser efficiency η_{eff} which is the output energy E_{out} divided by the input energy E_{inp}

$$\eta_{\text{eff}} = E_{\text{inp}}/E_{\text{out}} \quad (67)$$

We have now covered the basic equations and features of our kinetics model which are included in the computer code. In the next section, we shall discuss some recent experimental results for the XeF laser. We shall present results of our computer simulations of these experiments. We shall also propose some experiments which would be useful in benchmarking the code.

SECTION IV

NEW THEORETICAL AND EXPERIMENTAL RESULTS

A. Discussion of Model Development During FY86

During the past year, we worked on a number of projects related to improving the kinetics modelling of the XeF laser. The principal new contributions are: (1) calculation of the rates for the electron dependent processes using a time dependent Boltzmann solver, (2) analysis of electron beam energy deposition experiments with subsequent benchmarking of a Monte Carlo electron transport code, (3) analysis of recent energy extraction measurements in argon buffered XeF laser mixtures (some of this work is classified; it will be reported elsewhere).

B. Modelling of Recent Electron Density Measurements

The need for a time dependent treatment of the secondary electron energy transport processes was discovered when we tried modelling the electron density measurements of Kimura *et al.* [54] (this paper is not yet published; it comprises Appendix A). There are two important results of this work to consider: (1) the electron density in the XeF electron beam pumped XeF (with molecular fluorine) laser is a factor of 2-4 times that predicted with models using published values for the dissociative attachment rate of fluorine (typically $3-8 \times 10^{-9} \text{ cm}^3/\text{sec}$), (2) the burnup rate of the fluorine is much faster than expected. Both of these effects are critical issues, since the higher electron density means that all of the rates for the electron quenching processes are wrong, and the faster burnup rate for the fuel means that more fuel will be required for long pulse operation. In our initial attempt to explain the electron density results, we discovered that the dissociative attachment rate had to be in the range of $1.8-2.0 \times 10^{-9} \text{ cm}^3/\text{sec}$. We could not predict this value using the steady state Boltzmann treatment described by Johnson *et al.* [15]. In addition, this value is nearly a factor of two less than that measured by Hsia *et al.* [4]. We ultimately solved the problem by realizing that the steady state Boltzmann model could not properly account for the energy transport out of the high energy electron beam. Using a

time dependent model developed by Morgan [18], we were able to predict the lower value for the dissociative attachment rate. When incorporated in the kinetics model, we were able to predict the results of Fig. 4. The agreement with the electron density measurements is good during the first half of the time history. The late time behavior is progressively poor because the fluorine molecules are dissociating by another means other than dissociative attachment. We believe that the electron excitation of the F_2 to a repulsive electronic state is occurring. A literature search is presently underway to find the cross-section for this process so that we can include it in the Boltzmann code calculations.

During the course of this work, we noticed that the time dependent Boltzmann calculation came to an approximate steady state rather quickly, so that it was not necessary to update the excited state densities more than a few times during the course of the kinetics history. The electron temperature effectively clamped at 1.25 electron volts (ev), and the dissociative attachment rate varied less than 10 percent. This is an important result because we were able to greatly reduce computation time (nearly an order of magnitude). In fact, the set of rates presented in Table II can be used to model most cases (this eliminates the costly Boltzmann calculation). We shall continue to test this idea as more data becomes available. As a consequence of this work, we have developed a new set of electron quenching rates. Basically, the rates are lower than the old experimental literature recommends. Interestingly, the new values are more in line with theoretical predictions [55].

C. Modelling of Electron Beam Energy Deposition

An extremely important part of any laser experiment is the determination of the amount of energy deposited in the laser volume because it is essential for the calculation of the laser efficiency. For an electron beam pumped excimer laser, this is determined from a change in pressure in the laser volume. The reason this works is that, except for the energy which escapes as radiation (fluorescence), all of the input energy eventually ends up as heat. The resulting temperature rise is measurable as a pressure change (the acoustical

Fig. 4 A Comparison of Predicted and Measured Electron
Densities for Electron Beam Pumped Mixtures of Ne/Xe/F₂

energy is negligible). However, this can only give a value which is averaged over the entire volume of gas. What is really required is the deposition over the laser extraction volume. The obvious solution is to baffle the pumped region to be only the extraction volume. Usually this is not practical because of cost, or because of complications due to parasitics.

During this year, we have finished analyzing the results of an experiment by Mandl and Salesky (this work was originally performed in September 1984 and funded by this program) on spatially resolved e-beam energy deposition in rare gases. The details of this work are presented in Appendix B. The most important consequences of this work are that 3-dimensional Monte Carlo electron transport calculations coupled with either pressure rise measurements or Faraday cup measurements of the e-beam current density (with known beam energy) are sufficient to determine the spatial energy loading (or pumping) for a given laser experiment. This is particularly important for benchmarking the predictive capabilities of a kinetics code. Of course, the time history of the e-beam voltage and current must be measured, and the fluorescence loss taken into account. We believe that this technique will be invaluable to the experimental laser development at Los Alamos for electron beam pumped devices. This will be particularly true for the chemical laser program where knowledge of the excitation rates is very important.

D. Modelling of Energy Extraction in Ar/Xe/F₂ Mixtures

In the spring of 1985, a series of XeF (351-353 nm) energy extraction measurements were performed at Spectra Technology, Inc. (STI) (see Appendix C). The principal reason for these measurements was to explore the use of argon buffered mixtures as an option to conventional Ne buffered mixtures. The nearly factor of two higher electron stopping power of argon over neon results in operating pressures of only two atmospheres with argon versus four atmospheres of neon. Since ultimately we shall be interested in building very large devices with an output aperture in excess of 1 meter, the lower pressure Ar buffered mixtures are desirable from the point of view of output window

design and cost, whether the window is a quartz window, a shuttered window or an aerodynamic window. The laser gas mixtures considered were composed of Ne/Xe/F₂, Ne/Xe/NF₃, Ar/Xe/F₂ and Ar/Xe/NF₃. The neon buffered mixtures considered were chosen to be typical of current research at AERL, MLT and SDI, and to serve as a benchmark for the measurements with the argon buffered mixtures.

Qualitative results from these experiments indicate that for the higher pump rates $> 150 \text{ kw/cm}^3$ and pump times $> 0.5 \text{ sec}$ resulting in energy loading of $> 0.075 \text{ J/cm}^3$, the Ar buffer mix performs nearly as well as the Ne buffered. Although this is a surprising result in view of the published literature on XeF which always shows about a 2 (neon) to 1 (argon) ratio of performance (laser efficiency), none of these experiments were performed at the higher pump rates which we achieved. An exception to this is the work of Champagne *et al.* [56]. However, because he used brewster angle windows, which are very lossy, the argon buffer mixture never performed very well. This is discussed further in Appendix C. The neon buffered mixture follows the trend of the input pump pulse and is nearly constant through the lasing history. The enhanced behaviour (the constantly increasing output) of the Ar mixture is probably due to heating of the gas which has an effect on the upper XeF(B) vibrational state populations and more importantly the lower XeF(X) state population since it is weakly bound and can dissociate [58]. Different heating rates will occur because the Ar buffered mixture has a lower heat capacity due to the lower density (pressure) of the mixtures. Another important effect could be due to the decreasing absorption cross-section of Ar_2^+ with temperature [44]. We believe that there is a combination of effects which make the Ar buffer mixtures look even more promising as a driver option to KrF. A detailed report of the experimental results can be found in Appendix C.

Figure 5 is a kinetics code simulation of the experimental results for the Ar/Xe/F₂ mix (as present in Appendix C) using the model described in this report. The results are in reasonably good agreement with the data; however the enhanced performance with time is

Fig. 5 Kinetics Code Simulation of the Electron Beam
Pumped Tahoma Laser Experiment

not modelled very well. At present, we have not resolved this issue but we shall pursue the problem. In Fig. 6 we present predictions of gain and absorption for the laser experiment. At present these measurements are not available for the Tahoma device. However, these predictions appear to be consistent with the measurements of Champagne [59]. Additional measurements of gain and absorption for the 351 - 353 nm region are critical to future code development and benchmarking. We have performed simulations of the test using this code; the qualitative agreement appears to be quite good. However, a critical evaluation cannot be made until the final calibration of the data is made. This work will be reported elsewhere in a memorandum.

Fig. 6 Kinetics Code Prediction of Gain and Absorption at the
351 nm XeF Transition for Electron Beam Pumped
Mixtures of Ar, Xe, and F₂

SECTION V

DERIVATION OF THE SPATIALLY DEPENDENT MULTILINE EXTRACTION MODEL

In Section III above, we developed the simple macroscopic model for laser extraction from the microscopic kinetic equations for stimulated emission by photons. The model was extended to include extraction from a simple flat-flat (or nearly flat-flat) oscillator which was uniformly pumped, with a spatially uniform flux. This model has been adequate for our modelling of energy extraction from a small (< 1 meter gain length) low gain ($g_0 L < 4$) device. However, for much larger devices, the uniform flux model is probably not adequate, because small aspect ratios (L/D) of < 5 , and high gains $g_0 L > 5$, will introduce ASE effects. In addition, long gain lengths > 2 meters will introduce the Ananov effect (or mode build up) which may require injection locking to aid in the flux buildup.

A. Single Line Extraction Model

In this section, we shall generalize the treatment of the spatially dependent stimulated emission process reported by Salesky *et al.* [57]. We begin by generalizing the single line case and then shall generate the equations for a two line laser. The spatial dependence (in say the z direction) of the flux distribution will allow for the fact that the photons move at the finite speed of light. This can be accomplished by generalizing the time derivative in Eq. (44) to the case of a transport equation:

$$\frac{d}{dt} \rightarrow c \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \quad (68)$$

The simple flux becomes:

$$\left(c \frac{\partial}{\partial z} + \frac{\partial}{\partial t} \right) \phi(z, t) = c (g - \alpha) \phi(z, t) + S \quad (69)$$

where c is the speed of light, g is the gain, α is the absorption, and S is a source term either internal or external; these quantities can all be functions of space and time. Next we expand the laser flux into a left-going ϕ_- and a right-going component ϕ_+ :

$$\phi = \phi_- + \phi_+ \quad (70)$$

so that the above equation becomes two equations:

$$\left(c \frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right) \phi_+(z, t) = c (g - \alpha) \phi_+(z, t) + S_+ \quad (71)$$

$$\left(-c \frac{\partial}{\partial z} + \frac{\partial}{\partial t}\right) \phi_-(z, t) = c (g - \alpha) \phi_-(z, t) + S_- \quad (72)$$

Note that we have also expanded the source terms into two (left-going and right-going) components. Also the negative sign on the second equation is the result of choosing the right-going direction as the $+z$ direction. The generalization into quasi-modes for the transverse angular spread in a cylindrically symmetric resonator is accomplished by expanding the fluxes ϕ_- and ϕ_+ into the quasi mode components:

$$\phi_- = \sum_k \phi_{-k} \quad , \quad \phi_+ = \sum_k \phi_{+k} \quad (73)$$

We shall identify the source terms S_- and S_+ with that part of the fluorescence from XeF^* which is moving to the left or right, and which is in the zeroth quasi-model which

represents the solid angle intercepted by the resonator $\Omega_0^2/(4\pi)$. Thus we obtain the equations:

$$c \frac{\partial \phi_{+k}}{\partial z} + \frac{\partial \phi_{+k}}{\partial t} = c (g - \alpha) \phi_{+k} + \frac{1}{2} \frac{\Omega_0^2}{4\pi} \frac{\tau_u}{\tau_r} c g \phi_{\text{sat}} \delta_{k_0} \quad (74)$$

$$- c \frac{\partial \phi_{-k}}{\partial z} + \frac{\partial \phi_{-k}}{\partial t} = c (g - \alpha) \phi_{-k} + \frac{1}{2} \frac{\Omega_0^2}{4\pi} \frac{\tau_u}{\tau_r} c g \phi_{\text{sat}} \delta_{k_0} \quad (75)$$

The additional factors in the source terms represent the spontaneously emitted photons, which are produced at the rate:

$$\frac{dN_v^{\text{se}}}{dt} = \frac{[\text{XeF}^*]}{\tau_R} \cdot f_B \quad (76)$$

where N_v^{se} is the density of emitted photons, $[\text{XeF}^*]$ is the XeF^* density, τ_R is the radiative lifetime, f_B is a factor less than 1 which depends on the vibrational state, ϕ_{sat} is the saturation flux defined in Section (III), g is the gain coefficient defined below, and τ_u is the upper state lifetime. We need an additional differential equation for the gain g to complete the model. We can obtain this by rewriting the kinetic equation for the XeF^* density $[\text{XeF}^*]$:

$$\frac{d[\text{XeF}^*]}{dt} = [\text{formation}] - \frac{[\text{XeF}^*]}{\tau_u} - c\sigma_s [\text{XeF}(\text{B}) - \text{XeF}(\text{X})] \quad (77)$$

Where the formation term is the sum of all of the reactions producing XeF^* , the quenching term is the sum of all of the reactions which destroy $[\text{XeF}^*]$ with

$$\frac{1}{\tau_u} = \frac{1}{\tau_r} + \sum_i \frac{1}{\tau_{Q_i}} \quad (78)$$

If we multiply through by σ_s and convert the photon density N_v to a flux ϕ and introduce the value for $\phi_{\text{sat}} = E_v/(\sigma_s \tau_u)$, this equation becomes

$$\frac{\partial g}{\partial t} = -\frac{g}{\tau_u} \left(1 + \frac{\phi}{\phi_{\text{sat}}}\right) + R(t) \quad (79)$$

where $R(t)$ is the formation term multiplied by the appropriate factors. These equations comprise the single line spatially dependent extraction model, along with the boundary conditions:

$$\phi_{+,k}(z, t=0) = \phi_{-,k}(z, t=0) = 0, \quad \text{all } k \quad (80)$$

which says that initially all the fluxes are zero,

$$\phi_{+,k}(z_m, t) = \phi_{-,k}(z_m, t), \quad \text{all } k \quad (81)$$

which says that the flux in the + z direction gets reflected by the mirror at z_m , and

$$\phi_{+,k}(0, t) = \frac{1}{M} \left(\phi_{-,k-1}(0, t) + \delta_{k,N}(0, t) \right) \quad (82)$$

which accounts for the fluxes at the feedback mirror at $z = 0$. Here the $k - 1^{\text{th}}$ quasi-model gets reflected (- to + direction) and becomes the k^{th} mode in the + direction. M is the magnification, z_m is the position of the collimating mirror, $z = 0$ is the position of the feedback mirror, and N is the Ananev number and the number of the self-replicating mode.

$$N = \frac{\ln\left(\frac{D^2}{\lambda R}\right)}{\ln(M)} \quad (83)$$

where λ is laser wavelength, D is the diameter, R the radius of the collimating mirror. To obtain the case of injection locking the first boundary condition is rewritten as;

$$\phi_{+,k}(z, t = 0) = \phi_{-,k}(z, t = 0) = \phi_{in} \delta_{kN} \quad (84)$$

This model has been coded as the subroutine NJORDY; it requires the subroutine EXTRACT1 to interface with the kinetics part of the code. The solution in NJORDY is accomplished by dividing the z -direction into a number of sheets, through which the flux propagates, the time to propagate from one sheet to the next represents a characteristic time or minimum time step which NJORDY can consider.

B. Multiline Extraction Model

The equations for the multiline laser line model for XeF can be obtained from the equations above by adding an index m , where m is the number of laser lines, to the fluxes $\phi_{\pm}^k$, the gain g , the absorption coefficient, α , formation rate R , and the saturation flux ϕ_{sat} . This will result in the set of coupled equations below:

$$c \frac{\partial \phi_m^{k+}}{\partial z} + \frac{\partial \phi_m^{k+}}{\partial t} = c (g_m - \alpha_m) \phi_m^{k+} + \frac{\Omega_o^2}{8\pi} \frac{\tau_u}{\tau_r} c g_m \phi_{sat} \delta_{k,0} \quad (85)$$

$$-c \frac{\partial \phi_m^{k-}}{\partial z} + \frac{\partial \phi_m^{k-}}{\partial t} = c (g_m - \alpha_m) \phi_m^{k-} + \frac{\Omega_o^2}{8\pi} \frac{\tau_u}{\tau_r} c g_m \phi_{sat}^m \delta_{k,0} \quad (86)$$

$$\frac{\partial g_m}{\partial t} = -\frac{g_m}{\tau_u} \left(1 + \frac{\phi_m}{\phi_{sat}^m}\right) + R_m(t) \quad (87)$$

There is a set of m boundary conditions like those given above. The solution of this set of equations can be quite difficult if the gain equation contains contributions from laser lines which interact with each other, as in the case of a deuterium fluoride (DF) chemical laser. For the case of the XeF laser, one such complication can be avoided by treating the two transitions at 351 nm as sufficiently overlapping to be one transition (see the discussion in Section III). The other complication is that lasing drives the vibrational manifolds of the XeF(B) state and XeF(X) state out of Boltzmann equilibrium, thus making a relaxation calculation necessary to provide the appropriate pump term ($R^m(t)$) for each gain equation. We shall incorporate the infinitely fast relaxation approximation discussed in Section III. By doing this, we can use a Boltzmann weighting factor to calculate the gain terms from the total populations of the XeF(B) and XeF(X) states. We can also get the appropriate pumping terms ($R^m(t)$) by weighting the total production of XeF(B) states. The Boltzmann factor f_B is given by:

$$f_B = \left[e^{\epsilon/kT_v} - 1 \right]^{-1} \quad (88)$$

where T_v is the vibrational temperature of the vibrational manifold, and ϵ is the energy of the vibrational state of interest.

The work on writing a computer subroutine implementing these equations has not been finished at the time of this report. We anticipate completing the computer model for the XeF laser early in FY87.

SECTION VI

THE XeF KINETICS CODE ARCHITECTURE

As much as possible we have tried to create a modular code in which each physics task is separated into a separate subroutine. The use of modular architecture has a number of advantages, the most important being easier understanding, especially for the new user, of what the code is supposed to do and how it is doing it. Modularity also makes it easier to add new subroutines or modifications to the model. A brief description of each subroutine is given below.

There are user input specifications or decisions (yes or no answers in the input menu) which determine what calculations are to be performed during a computer run. These decisions affect which subroutines will be used in the calculation. The first option allows the user to specify a calculation of the kinetic parameters without energy extract. With this option enabled, the code will generate the time history of the small-signal gain. If an extraction calculation is required, a second option must be specified to determine which extraction model to use. The question in the menu asks if the uniform flux model is requested. If the answer is yes, the subroutine EXTRACT is used (at present this is the only laser option). If the answer is no, the subroutine EXTRACT1 is used; this then calls the subroutine NJORDY. The third option determines whether the vibrational dependence of the XeF(B) state is to be used. This option determines whether the subroutine ALPHA or the subroutine ALPHA1 which treats the vibrational effect is to be used. This option is always enabled for XeF.

A simplified chart which shows the branch and return points in the calling sequence is depicted in Fig. 7. The main module XEFKIN handles all input and output file manipulation, including the plot file which is generated with the CGSHIGH graphics package. All plot related calls are performed in this module. The only other module called from XEFKIN is the subroutine DRIVER. The DRIVER routine contains the integration algorithm. At the beginning of each run, DRIVER calls subroutine ENERGY to initialize

Fig. 7 Schematic of the Kinetics Code Architecture
with the Scheme of the Subroutines

the energy changes for each reaction, RATES to generate the starting rate values, and DSPDT2 to initialize the derivative values. Once the main integration loop has been entered, DRIVER calls DSPDT which sets up the electron beam energy input channeling. DSPDT then calls either ALPHA or ALPHA1 depending whether the vibrational option is required in the XeF(B) state gain calculation. In addition, these subroutines calculate the total absorption coefficients. DSPDT then calls DSPDT2 to determine the updated values for the derivatives of the various species densities including the various photon densities. DSPDT2 calls RATES to obtain the current rates based on the current temperatures, neutral gas and electron. The next call in DSPDT is optional. It is a call to EXTRACT. The subroutine EXTRACT is called only if a lasing calculation is specified and if the uniform extraction model has been specified. The next subroutine call originates in the DRIVER subroutine. It is an optional call (in the present XeF code this is disabled) to the subroutine EXTRACT1. This call is executed only if the lasing option is enabled and the resonator extraction model is required. If called, EXTRACT1 serves as an interface to the NJORDY subroutine, which performs Korff's calculation of the injection-locked (an option) three-dimensional (cylindrically symmetric) unstable resonator model.

In addition to integrating the species densities, DRIVER also integrates the temperatures, the output flux, and the input power. Driver returns to the main XEFKIN after the user specified major time interval, to load the plotting arrays, and to dump the current values to the output file. In addition, the current status of the integrator is also given as an aid to future optimization. The state of the balances of the original input gases Ar, Xe and F₂, in their current form, are given to demonstrate conservation of matter. The energy balance (and the power balance) are shown as well as the charge balances, plus and minus. At the end of the user specified time limit, date headers are generated, followed by the writing of the plot files. A more complete discussion of the files and the use of the code is presented in the user's manual for the code.

SECTION VII

CONCLUSIONS

We have developed a sophisticated and useful model for the XeF laser. The work has included improvements in the physics of the model which include the consideration of the vibrational levels of the XeF B and X states. This allows for the treatment of the multiple laser line character of XeF. We have developed the equations to calculate the time dependence of gain on the many vibrational laser transitions observed in the XeF laser. In addition, a model for multiline lasing has been developed and implemented in the computer code. Temperature dependence of the absorption of the dimer ions Xe_2^+ and Ar_2^+ improves the model predictions for gas temperatures in excess of 400°K. These temperatures will occur during the pumping of the new generation of SDI laser devices. We have developed but not yet implemented a 3-dimensional time dependent extraction model which treats the effects of non-uniform flux, ASE and mode buildup in large low aspect ratio (< 10) lasers such as the Los Alamos large aperture module LAM. The extraction model also treats the effect of injection locking by an external laser. A complete discussion of the physics in the model is presented as well as a list of the reactions and their rate coefficients. Comparisons with the recent experiments of Salesky and Kimura (see Appendix C) and some older work of Champagne *et al.* have been given and discussed. A number of new phenomena have been observed and reported.

A computer code which implements the physics model was developed in a modular architecture with a user friendly interface. The code was written in FORTRAN 77 with minimal Cray FORTRAN extensions. The modular form separates each physics task into its own subroutine. Modularity in a code makes it easier to implement multiple task development (such as the 3D extraction model), even when done by more than one person. It also makes the code more understandable to the new user. The user friendly interface makes the various input options more accessible to the interactive user. The new input-output file structure allows the user to identify and label all of the input and output

(including the plot file) files with a unique three character prefix code. This is particularly useful for archival management or multiple case tests.

This report also serves as a documentation of our efforts over the past few years to improve the level of sophistication of modeling in XeF, in particular the predictions of multiline gain and energy extraction. As much as possible we have presented a discussion of the basic physical processes which are important in our view, and our method for dealing with them. In the future, we hope to continue this effort by proposing new experiments to benchmark the present model, and to help identify new areas for experimental and theoretical work. We shall also complete the work on the multiline spatially dependent extraction model. The fluorine chemistry in the model will be improved upon to allow for improved predictions of long temporal pulse pumping ($> 5 \mu\text{sec}$) of XeF.

SECTION VIII

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APPENDICES

APPENDIX A

Electron Density Measurements of Electron-Beam Pumped XeF and KrF Laser Mixtures

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ABSTRACT

A CO₂ quadrature interferometer is used to measure the temporal evolution of the electron density in XeF and KrF electron-beam pumped laser mixtures (nonlasing). Measurements are obtained using a 300 kV electron beam which delivers ≥ 10 A/cm² at the foil for pulse durations ≤ 650 nsec (FWHM). For typical XeF mixtures (99.35% Ne/0.5% Xe/0.15% F₂ at 2888 torr) the electron density varies between $3\text{--}4.5 \times 10^{14}$ cm⁻³ during the pulse. The electron density is found to increase as the initial F₂ concentration is decreased. Both 10% Kr in argon diluent and argon-free KrF mixtures are tested. The electron density for these mixtures has the same magnitude and behavior as the typical XeF mixture.

Several computer models of electron-beam (e-beam) pumped XeF and KrF lasers exist.¹⁻⁴ All the models rely on verification using experimental data on the laser performance, for example the laser output flux and efficiency. The models have been very useful in improving the understanding of the complex plasma chemistry which occurs in these lasers. One key area is the role of the primary and secondary electrons which are very important in the formation of the excimers, both through ionic and metastable channels,⁵ and in quenching of excited states.⁶ Despite the importance of the electrons, none of the codes have been validated with regard to their predictions of the electron densities, primarily because of the lack of experimental data.

This letter reports the first measurements of electron densities in e-beam pumped XeF and KrF (10% Kr and argon-free) laser mixtures under nonlasing conditions. Electron density measurements have been previously reported in discharge pumped XeCl⁷⁻⁸ and XeF;⁹ and, recently, the electron density in e-beam pumped Ar/F₂ and Ar/HCl has been measured.^{10,11} An earlier letter¹² gives the electron density results for e-beam pumped XeCl laser mixtures.

The experimental system is described in more detail elsewhere.¹² Briefly, the gas chamber of the Tahoma laser system¹³ is placed in one arm of a CO₂ (10.6 μ m) quadrature interferometer.¹⁴ The electron density is determined from the refractive index change of the plasma due to the presence of free electrons. In this particular experiment, one fringe

shift corresponds to an electron density of $3.0 \times 10^{14} \text{ cm}^{-3}$.

The electron beam source is a carbon felt, cold cathode at 300 kV with a peak emission current of $\approx 23 \text{ kA}$. The current density at the foil (0.13 mm aluminized Kapton) is $\geq 10 \text{ A/cm}^2$. A pressure rise technique¹⁵ is used to calibrate the power deposition in the gas mixtures. In the XeF gas mixtures, the deposition is $\approx 180 \text{ kW/cm}^3$. In the KrF gas mixtures, the deposition is kept constant at $\approx 120 \text{ kW/cm}^3$ by adjusting the total pressure of the gas mixtures to compensate for the different electron stopping powers of Ar and Kr. All measurements are performed at 294°K.

Figure 1 shows typical time dependent electron density results for XeF as a function of the F_2 halogen donor. The XeF mixture is 0.5% Xe in Ne diluent to a total gas pressure of 2888 torr. For reference, Fig. 2 shows a normalized typical trace of the excimer sidelight fluorescence for the 0.15% F_2 XeF mixture (the same trace is also representative of the KrF* sidelight fluorescence for the data given in Fig. 3). The top of Fig. 3 gives a normalized typical pulse shape of the e-beam current. The e-beam pulse length does vary slightly shot-to-shot; some of the data curves (e.g. the 0.15% and 0.20% F_2 mixtures in Fig. 1) represent data for pulse lengths shorter than shown in Fig. 2 and the top of Fig. 3.

As seen in Fig. 1, as the initial halogen concentration is reduced, the electron density tends to increase more rapidly towards the end of the pulse. This is not surprising, since as the F_2 is depleted (burned-up),

due to the dissociative attachment with electrons, less F_2 is available to control the electron density. Without the attacher, the electron density appears to approach the higher densities found in e-beam pumped pure rare gas mixtures.¹⁶ Dissociative recombination of the electrons with dimer ions does not help control the electron density because the dimer ion number density tends to be too small^{2,3} for the conditions of this experiment. The rapid increase in the electron density as the halogen is depleted has been predicted in e-beam sustained discharge models,⁴ but the increase tends to be at least an order of magnitude larger than the results in Fig. 1. Since the mechanism for electron population growth in discharge lasers (metastable ionization leading to electron avalanche and arcing) is very different from that in e-beam pumped systems, care must be taken when comparing the two pumping schemes.

From past experience¹⁷ on the Tahoma laser device, for these pulse lengths, the optimum laser performance on XeF occurs at F_2 concentrations of 0.10-0.15%. At higher F_2 concentrations, although the electron density may decrease, there is a significant increase in F_2 quenching and F_2 absorption which reduces the laser efficiency. The electron density profiles in Fig. 1 do not display a peak in the density at the beginning of the pulse as is seen in the XeCl data.¹²

The electron density results for the KrF mixtures are given in Fig. 3. The mixtures are 89.7% Ar/10% Kr/0.27% F_2 at 1007 torr, and 99.8% Kr/0.4% F_2 at 865 torr. For both mixtures, the F_2 partial pressure

is 2.7 torr, corresponding to a density of $8.9 \times 10^{16} \text{ cm}^{-3}$. The electron density behavior is very similar between the two KrF mixtures. The slight increase in density towards the end of the pulse is apparently due to F_2 burn-up.

To investigate the effects of high energy loading, measurements are performed on the 10% Kr and Kr/ F_2 mixtures for e-beam pulse lengths of 650 nsec (FWHM). The results are depicted in Fig. 4. The F_2 concentration has been increased in the 10% Kr mixture to 5 torr ($1.6 \times 10^{17} \text{ cm}^{-3}$), and in the Kr/ F_2 mixture to 4 torr ($1.3 \times 10^{17} \text{ cm}^{-3}$). At this higher F_2 concentration, the electron density for the Ar/Kr/ F_2 mixture does not increase after 300 nsec as seen in Fig. 3, but remains essentially constant. On the other hand, the Kr/ F_2 mixture still displays a gradual increase in the electron density towards the end of the pulse. Although the initial F_2 concentration in the Kr/ F_2 mixture is slightly less than in the Ar/Kr/ F_2 mixture, this tendency for the electron density to be higher in the Kr/ F_2 mixtures is consistent with model predictions² and long pulse extraction measurements.¹⁸ Kr^+ is formed more efficiently in Kr/ F_2 than Ar^+ is in Ar/Kr/ F_2 ,¹⁵ which means more electrons are freed per unit of power deposition. Higher gas heating effects in the Kr/ F_2 mixtures also slightly increase the dissociative attachment rate constant of F_2 ,¹⁹ which leads to a faster depletion of the halogen fuel and an increase in the electron density.

The KrF mixtures given in Figs. 3 and 4 represent optimum mixtures on the Tahoma laser. For the optimum XeF and KrF laser mixtures, the electron

density varies between $3-4.5 \times 10^{14} \text{ cm}^{-3}$. The density can be estimated analytically from the expression,

$$n_e = \frac{P_i / (eW_i)}{k_a [F_2]}, \quad [1]$$

where n_e is the electron density, P_i is the input power deposition, (eW_i) is the energy loss per ion pair produced,²⁰ k_a is the F_2 attachment rate constant, and $[F_2]$ is the F_2 number density. Using a typical F_2 attachment rate constant of $2 \times 10^{-9} \text{ cm}^3/\text{sec}$,¹⁹ and values for (eW_i) of 38 eV and 26 eV for Ne and Ar, respectively,²⁰ we find that electron densities of $1-2 \times 10^{14} \text{ cm}^{-3}$ are predicted which are 2-3 times smaller than measured.

Although Eq. 1 may predict lower electron densities, preliminary comparisons²¹ with a computer model² yield good agreement in overall magnitude and temporal behavior with the data. The model uses a time-dependent Boltzmann equation solution²² and indicates that the secondary electron energy distribution is significantly non-Maxwellian for the pump conditions of this experiment. It also predicts that the F_2 dissociative attachment rate constant is lower than that assumed in Eq. 1, which leads to higher electron densities.

The reasons the electron distribution is non-Maxwellian are complex because the distribution is affected by a variety of interrelated kinetic processes. For example, the electron dissociative attachment rate constant of F_2 is directly dependent on gas temperature¹⁹ and inversely dependent on

electron temperature.^{19,23} However, Rozenberg, *et al.*¹⁰ show that the dissociative attachment rate constant is also a function of the F_2 concentration and e-beam current density, and that adding F_2 has the net effect of increasing the average electron temperature, caused by removal of low energy electrons through attachment. This affects the electron energy distribution and, at the concentrations and densities used in this work, may lead to a lower attachment rate constant. Other implications of the electron density results are reported elsewhere.²⁴

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FIGURE CAPTIONS

FIGURE 1. Typical electron densities of XeF mixtures (0.5% Xe, balance Ne to 2888 torr) for various initial F_2 concentrations. Data points are not shown; however, a representative error bar is indicated.

FIGURE 2. Normalized waveform of typical XeF^* and KrF^* sidelight fluorescence emission for the data given in Figs. 1 and 3.

FIGURE 3. Typical electron densities of KrF mixtures (89.7% Ar/10% Kr/0.27% F_2 at 1007 torr, and 99.8% Kr/0.4% F_2 at 865 torr). The top trace is the normalized e-beam current pulse. Data points are not shown; however, a representative error bar is indicated.

FIGURE 4. Typical electron densities of KrF mixtures (89.5% Ar/10% Kr/0.5% F_2 at 1007 torr, and 99.4% Kr/0.6% F_2 at 865 torr). The top trace is the normalized e-beam current pulse. Data points are not shown; however, a representative error bar is indicated.

Fig. 1

Fig. 2

Fig. 3

Fig. 4

APPENDIX B

E-BEAM DEPOSITION STUDIES OF THE RARE GASES^{*}

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ABSTRACT

The spatial distribution and absolute values of the stopping power of the rare gases Kr, Ar and Ne were determined in 275 kV e-beam excited gas mixtures. The measured values were compared with 1D and 3D Monte Carlo code calculations. Excellent agreement was found in all cases.

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I. INTRODUCTION

The determination of the absolute laser efficiency and the sidelight fluorescence efficiency are critical in testing laser models and using these models to scale to high power. The accuracy of these measurements depends on accurately measuring the laser output energy and the fluorescence emitted. It also depends on accurately determining the excitation energy deposited into the gas. It is the determination of the input energy which is typically the more difficult measurement.

In this paper we will describe a series of measurements of energy deposition for e-beam excited rare gases. Deposition measurements were carried out for various rare gas fluoride laser mixes. The mixtures consisted mainly of a rare gas and some small fluorine impurity needed to form the upper laser level. The rare gases studied were Kr, Ar and Ne. Monte Carlo calculations were performed and compared to the measurements.

In this study the effects of atomic number, gas density and stopping power variations of about a factor of four were observed affording a wide range over which to check our measurements and model. The rare gases studied are of particular interest in that they are the major constituents of the key rare gas excimer laser systems now being investigated for scaling to high power, i.e., XeF, KrF, XeCl and ArF.

II. APPROACH

A one meter device, described previously⁽¹⁾ was used to measure both the total energy deposited into the gas by the e-beam and the spatial distribution of the deposited energy. The experimental parameters are listed in Table I. Our original method of measuring the spatial distribution of deposited energy used a photomultiplier to monitor the excimer fluorescence through two pinholes. The setup gave a 0.5 cm resolution and necessitated

taking a large number of laser shots to map out the entire deposition spatial profile. The present method replaces the point by point measurement with a single photograph of the entire fluorescence profile. The photograph is densitometrized and linearized to give the entire spatial profile in one exposure. This leads to an improvement in the determination of spatial profile.

Corrections are made, where appropriate, for the varying fluorescence efficiency with deposition. These spatial profiles are put onto an absolute scale by determining the total energy deposited into the gas using the measured jump in pressure caused by e-beam heating of the laser mixture. The results are compared with Monte Carlo code calculations.^(2,3)

III. EXPERIMENT

a. Total Energy Deposition in E-Beam

The maximum energy available from the e-beam is the product of the e-beam voltage and current integrated over the pulse duration. Figure 1a is an oscillogram showing the I/V characteristics of the Marx Bank used to drive the e-beam. Not all the energy is necessarily deposited into the gas since some electrons may not be stopped in the cavity but instead hit the rear wall laser cell. In any volume element of excited gas the energy deposited is the product of the stopping power and the e-beam current integrated over the pulse duration. It is the energy deposited in the various volume elements which raises the gas temperature and results in a pressure pulse. A typical pressure pulse as measured on a Validyne pressure transducer is shown in Figure 1b. The initial overshoot in pressure is due to shock waves produced by the expanding e-beam heated gas. We ignore this overshoot and determine

the initial pressure rise by making a least squares fit to the long time exponential decay of the pressure pulse and extrapolating back to $t = 0$. It has been shown⁽⁴⁾ that for large volume cavities (our cavity is 13 cm deep by 7 cm high) the pressure jump method should be accurate. The energy deposited in the cavity is determined from the heat capacity of the gas and the ratio of e-beam excited to total system volume.

Some fraction of the energy deposited into the gas does not go into heat but instead is radiated by the excimer. Figure 1c shows temporal traces of excimer radiation -- in this case 351 nm and 353 nm XeF radiation. For XeF this is the main source of radiation. In calculating the total energy deposited into the gas the radiation loss must be accounted for. In general, the rare gas halides have appreciably lower fluorescence efficiencies than the rare gas excimers. Rare gas excimers can, under certain conditions, have fluorescence efficiencies in excess of 50%.⁽⁵⁾ It is for this reason that, although we are studying stopping power in the rare gases, small amounts of halogen fuel were added. This essentially channels all the radiation into the rare gas halides and substantially lowers the fluorescence efficiency without significantly affecting the stopping power. Thus small errors in determining the fluorescence efficiency will not add significantly to the uncertainty in the determination of the stopping power.

b. Spatial Profiles of E-Beam Deposition

Once the total energy deposited in the cavity is determined one must measure the spatial profile of the deposition. Knowing these two quantities allows one to determine the spatial distribution of the stopping power.

In order to determine the spatial distribution of the e-beam deposition in the gas a setup as shown in Figure 2 was used. The technique involves

photographing the sidelight of the rare gas halide emission. A filter at the rare gas halide wavelength (e.g., 353 nm for XeF) is placed in front of the camera and the emission from the center of the cavity is focussed onto the film plane. In order to reduce amplified spontaneous emission and prevent problems of parallax the 1 meter e-beam of our laser was masked down to ~ 4 cm width. A spatial marker plate was placed inside the cavity, close to the masked e-beam to allow the unambiguous determination of the spatial distribution of the fluorescence.

A photograph showing a typical e-beam profile is displayed in Figure 3. The radiation, KrF, in this case, is filtered at 249 nm. The "v" groove of the spatial marker plate locates the cavity center line. The distance between marker slits is 2 cm. These photographs are made on Polaroid Type 55 positive-negative film. The negative is densitometered and, using a linearization program⁽⁶⁾ written for this type film, we generate linearized plots of the fluorescence. A typical plot is given in Figure 4. This is for a gas mixture of almost pure Kr (99.7% Kr and 0.3% F₂) at 1.0 ama. The laser cavity is 13 cm deep and although the fluorescence signal can be seen across the full extent of the cavity, reflections cause ambiguities at either end. The signals are therefore only plotted over their useful range, 2-12 cm.

The spatial distribution of the emission together with the measured value of the total energy deposition allows an absolute determination of the spatial distribution of the stopping power. Figure 4 is labeled with the values of stopping power determined from our measurements. Kr has the highest Z of any gas that was studied in this program and resulted in the largest variation of stopping power across the cavity (~ factor of 4). Over such a

large range of stopping power there is a small change in fluorescence efficiency. We used a previously developed KrF code⁽¹⁾ to calculate the change in fluorescence efficiency with stopping power. This correction is plotted (dashed curve) in Figure 4.

The e-beam used for these measurements⁽¹⁾ was guided by in a 4 kg magnetic field allowing one to treat this problem as a 1D instead of 3D calculation. In order to check the validity of this assumption both 1D and 3D Monte Carlo calculations of the deposition were made using these codes.^(2,3) Little difference was found in the deposition predicted by the two codes at this high magnetic field. The 3D calculations tend to predict slightly lower deposition due to the spreading of the E-beam. These results may give a slightly better fit to the data although both 1D and 3D codes give good agreement. The results of the 1D calculations are plotted as the square data points and the 3D calculations as diamond data points in Figure 4. Similar measurements at 0.8 ama gave comparable agreement.

Deposition was also measured for almost pure argon. Mixtures of 95.7% Ar with 4% Kr and 0.3% F₂ at densities between 0.5 and 1.5 ama were used to again form the KrF rare gas excimer. Using the procedures outlined above a determination was made of the stopping power of this mixture. The measured and 1D and 3D Monte Carlo code calculated values of the stopping power for 1.5 ama are plotted in Figure 5.

Measurements in almost pure Ne were made at densities ranging from 2 to 3.9 ama with mixtures of 99.35% Ne with 0.5% Kr and 0.15% F₂. In this case the radiation is from the XeF rare gas excimer at 353 nm. Comparison between the measured and calculated stopping power for a 3 ama density case is shown in Figure 6.

IV. CONCLUSIONS

Excellent agreement between measured and calculated stopping power in Kr, Ar and Ne gases has been demonstrated. For our experimental conditions where a large 4 kG magnetic field is used there is little difference between the 1D and 3D Monte Carlo Code calculations. The major advantage of the 1D code is that it runs about an order of magnitude faster. Measurements were also made in 99.7 Kr/0.3 F₂ mixtures with He added at densities between 1 ama and 3 ama. Excellent agreement between measured and calculated stopping power was also found for these cases, however, the 1 ama Kr stopping power is much higher than the He stopping power, even at the 3 ama density. The differences between the stopping powers measured with and without the He were small. Higher densities of He were not used since the He reduces the KrF* fluorescence and makes the measurements impractical. Similarly, almost pure Xe mixtures were not used because Xe is a very effective quencher of XeF* radiation at high density and would also significantly reduce the fluorescence.

These measurements allow a determination of the stopping power of the rare gases Kr, Ar and Ne, to better than 10% and therefore allow accurate determination of the laser efficiencies and fluorescence efficiencies of some of the most important excimer lasers. This should help improve the code development and understanding of these laser systems.

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FIGURE CAPTIONS

1. Measurements used in determination of total energy deposition by e-beam. These particular measurements are for 3 ama of 99.35 Ne/0.5 Xe/0.3 F₂; (a) oscillogram showing Marx Bank current and voltage characteristics, (b) oscillogram of Validyne pressure transducer output displaying pressure jump caused by e-beam heating of cavity gas, (c) oscillogram of photodiode signals giving temporal display of sidelight from the two major uv bands of XeF, 353 nm and 351 nm.
2. Experimental setup used to determine the spatial distribution of the radiation from e-beam excited rare gas-halide excimer mixtures. E-beam is masked off to 4 cm width.
3. Typical photograph of spatial distribution of excimer radiation. This case is for KrF. Note "v" groove marks centerline of laser cavity.
4. Linearized densitometer trace of KrF fluorescence (1.0 ama, 0.3 F₂/99.7 Kr). Absolute scale for stopping power is determined from pressure jump analysis. Dashed line shows corrections for fluorescence change with pump rate. Square data points show 1D, 275 kV, Monte Carlo code calculations. Diamond data points are 3D Monte Carlo code results.
5. Measured and calculated stopping power for KrF mixture (0.3 F₂/4 Kr/95.7 ar) at 1.5 ama.
6. Measured and calculated stopping power for XeF mixture (0.15 F₂/0.5 Xe/99.35) at 3 ama.

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TABLE I
EXPERIMENTAL PARAMETERS

E-BEAM

VOLTAGE	275 kV
CURRENT	11.4 A/cm ²
CROSS SECTION	35 cm x 100 cm
PULSE DURATION	0.6 - 1.25 μ s

MAGNETIC FIELD	4 kG
PUMPED VOLUME	4.6
CHAMBER VOLUME	14.7
GAS PRESSURE	0.5 - 4 AMA
FOIL THICKNESS	3 mil (Aluminized Kapton)

MARX OUTPUT
VOLTAGE
91.8 kV/div

MARX CURRENT
5.3 kA/div

PRESSURE ~ 52,000 BARVES/DIV
(0.75 psi/div)

(b)

L4863

FIGURE
1

L484E

FIGURE

2

FIGURE

M2137

FIGURE

M2138

FIGURE

M2122

FIGURE
6

APPENDIX C

Improved Performance in Argon Diluent XeF Lasers

by

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ABSTRACT

In electron-beam pumped XeF lasers, neon has been preferred over argon as the diluent. However, for the same energy deposition, the argon mixture can be operated at a lower pressure than neon, thereby easing design requirements in large aperture lasers. Under high input energy conditions ($>100 \text{ J/l}$), significant intrapulse heating appears to occur in the argon mixture which substantially improves its overall laser efficiency. Presented are room temperature extraction measurements comparing neon and argon diluent mixtures under high energy loading conditions (160 kW/cm^3 , $1 \text{ } \mu\text{sec}$ laser pulse lengths). It is found that the laser output stays relatively constant during the pump pulse for the neon mixture, but the output for the argon case increases almost linearly during the pulse. The laser efficiency in the argon mixture is also higher when using NF_3 rather than F_2 for the halogen fuel.

When first demonstrated, electron-beam pumped XeF lasers used argon as the diluent.¹⁻² Later, Champagne and Harris³ discovered that using neon as the diluent improved the laser performance. Hsia, et. al.⁴ found that operating the laser at elevated temperature further dramatically improved the performance. Thus, current wisdom in regards to obtaining the best performance from XeF is to operate the laser with neon diluent at >3 amagats and at 400-450°K.

There are a number of reasons why neon is better than argon as the buffer gas. The quenching rate of neon is lower than argon,⁵⁻⁶ and the absorption at 350 nm of Ne_2^+ is lower than Ar_2^+ .⁷ There is also a higher electron density produced in the argon mixtures, due to the lower ionization potential of argon,⁸ which leads to higher electron quenching. However, under certain circumstances argon may be an attractive alternative to neon. For the same energy deposition, an argon diluent mixture can be operated at a lower total pressure than a neon mixture. This is because the electron stopping power of argon is almost a factor of two greater than neon.⁹ For large aperture lasers, operating at lower pressures considerably eases the design requirements for the windows and foils. Another advantage of using argon diluent, which will be demonstrated in this paper, is that the laser efficiency with argon diluent can be comparable, and perhaps even higher, than the neon mixture during high input energy conditions (>100 J/l).

Measurements are made using the Tahoma laser which has been described elsewhere.¹⁰⁻¹¹ The 300 kV electron beam delivers >10 A/cm² at the foil

(0.076 mm Kapton) in an active volume 3.8 cm dia. x 70 cm. Power deposition is measured using a pressure rise technique and is kept constant at $\approx 160 \text{ kW/cm}^3$ for all gas mixtures by adjusting the total pressure to compensate for the different electron stopping powers of argon and neon. The electron beam pulse length is variable from 0.35 - 1.2 μsec ; this leads to maximum laser pulse lengths of approximately 1 μsec . One sided, anti-reflection coated windows are used; the rear mirror is a 5 m, $>99\%$ reflector; and the output mirror is a flat 50%R reflector (the optimum output coupling determined from earlier work¹²). All measurements are performed at an initial gas temperature of 294°K.

For the neon mixture, the laser efficiency is essentially the same when using either F_2 or NF_3 .¹²⁻¹³ However, for the argon mixture higher efficiency is obtained when using NF_3 rather than F_2 . This may be because the argon mixture is more sensitive to losses due to UV absorption by F_2 . Figure 1 shows the time averaged laser efficiency (laser output/energy deposited in gas) as a function of the NF_3 concentration for the argon diluent case.

Figure 2 are photodiode traces of the laser output and sidelight fluorescence for the neon and argon diluent mixtures. The gas mixtures are 6 torr F_2 , 15 torr Xe, and 2624 torr Ne; and 4 torr NF_3 , 15 torr Xe, and 1493 torr Ar. The typical electron beam voltage and current waveforms for the data shown in Fig. 2 are given in Fig. 3. Note, the current is relatively constant and lasts $>1 \mu\text{sec}$.

For the neon mixture [Fig. 2(a)], the laser output is relatively constant, but terminates after approximately 0.8 μ sec. The decrease in the sidelight fluorescence towards the end of the pulse suggests that F_2 is being consumed ("burned up") which leads to increased electron quenching. Similar behavior has been observed on this device in Kr/ F_2 mixtures.¹⁴ Addition of more F_2 to the neon mixture tends to lengthen the laser pulse, but it also decreases the laser efficiency due to increased F_2 quenching and absorption.

The laser output for the argon mixture [Fig. 2(b)] displays a nearly linear increase during the pulse duration. This linear increase is observed when using either NF_3 or F_2 . The argon mixture sidelight fluorescence is lower than the neon mixture fluorescence, which is consistent with a lower formation efficiency⁶ and/or higher quenching by argon and electrons. The magnitude of the argon mixture fluorescence is the same for both NF_3 and F_2 . Unlike the neon mixture fluorescence, the argon mixture fluorescence is relatively constant during the entire electron beam pump pulse.

We believe the linear increase in the laser output for the argon diluent mixture is the result of intrapulse gas heating occurring due to the lower heat capacity of the argon mixture. A temperature rise in the argon mixture of 150°-185°K occurs during the 1.0-1.2 μ sec pump pulse. This temperature rise may improve the laser performance for the same reason elevated temperature operation improves the neon mixture. Heating removes

the XeF lower state bottlenecking which tends to suppress lasing on the 351 nm lines.^{12,15} The absorption at 350 nm of Ar_2^+ also decreases with increasing temperature.¹⁶

It should be noted that the temperature rise in the neon mixture during the 1.0-1.2 μsec pump pulse is 90°-110°K. Although lower than in the argon mixture, this temperature rise should still lead to a detectable improvement in the laser output during the pump pulse. However, this improvement is not observed in the data; the reason is not understood at this time.

It is interesting to note that the laser output pulse measured by Champagne, et. al.¹⁷ also tends to increase over time. Their experiment used $\text{NF}_3/\text{Xe}/\text{Ar}$ with similar electron beam characteristics, but used Brewster angle windows. The high optical losses from the Brewster angle windows may account for their apparent lower efficiency as compared to this work. A similar problem with Brewster angle windows has been observed in KrF laser experiments.¹⁸

For the argon mixture, the linear increase of the laser output means the laser efficiency tends to improve with pump pulse length or, equivalently, with higher energy loading. Plotted in Fig. 4 is the time averaged laser efficiency as a function of laser pulse width for both diluents. The efficiency of the neon mixture is nearly constant with pulse width, although it may decrease slightly as the pulse length approaches

1 μ sec due to the effects of F_2 burn-up. The efficiency for the argon mixture increases with pulse width and actually surpasses the neon efficiency after roughly 0.7 μ sec. This can be seen more clearly in Fig. 5 which plots the laser power efficiency during the pump pulse for the data shown in Fig. 2. Power efficiency is defined as the ratio of the output power density (W/cm^2) to the input power density (W/cm^2).

Although not measured in this work, it would be illuminating to obtain the time-dependent lasing spectrum of the argon mixture to see if lasing on the 351 nm lines increases with time. It would be also useful to perform extraction measurements at elevated temperature to determine if even better performance can be obtained using argon.

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FIGURE CAPTIONS

FIGURE 1. Laser output efficiency as a function of NF_3 concentration for the argon diluent mixture. The xenon concentration is 1 percent; total pressure is 1520 torr. Also plotted as an open circle is the efficiency when using F_2 as the halogen fuel.

FIGURE 2. Laser photodiode oscilloscope traces of the laser output (top trace) and sidelight fluorescence (bottom trace). (a) is 0.23% F_2 /0.57% Xe/99.2% Ne at 2645 torr. (b) is 0.26% NF_3 /1.0% Xe/98.7% Ar at 1510 torr. The time scale is 200 nsec/div.

FIGURE 3. Typical electron beam voltage and current waveforms for the data shown in Fig. 2. Top trace is current at 10 kA/div; bottom trace is voltage at 100 kV/div. The time scale is 200 nsec/div.

FIGURE 4. Laser efficiency of the neon and argon mixtures as a function of the laser pulse length. Open circles are for neon diluent (F_2 fuel); closed circles are for argon diluent (NF_3 fuel).

FIGURE 5. Peak power efficiency during the pump pulse for the data given in Fig. 2. Open circles are for the neon diluent mixture; closed circles are for the argon mixture.

Fig. 1

(a)

(b)

Fig. 2

Fig. 3

Fig. 4

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