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DISCHARGE PHYSICS, FORMATION AND
QUENCHING PROCESSES OF XeF^+

FINAL TECHNICAL REPORT

MASTER

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ABSTRACT

This report discusses the discharge physics and the formation and quenching processes in typical XeF laser mixtures. We have determined that the discharge physics is dominated by electron impact excitation and ionization of the rare gas metastables. The ionization of the metastables impacts the discharge stability directly while their excitation strongly affects the efficiency of producing XeF^* . From our discharge experiments we have determined that $\text{Xe}^+(6p)$ states react with NF_3 to produce XeF^* with a branching ratio of 0.7. The rate-constant for this reaction is $4 \times 10^{-10} \text{ cm}^3/\text{sec}$. The formation and quenching processes have been determined by analyzing the dependence of $(\text{B}^2\Sigma_{1/2}^+ \rightarrow \text{X}^2\Sigma_{1/2}^+)$ radiation on the partial pressures of Ar, Xe, and F_2 . In the experiments the XeF^* was produced by a high energy E-beam. We have determined the two and three body quenching rates by Ar to be $8 \pm 4 \times 10^{-13} \text{ cm}^3/\text{sec}$ and $1.5 \pm 0.5 \times 10^{-32} \text{ cm}^6/\text{sec}$. Xe quenches XeF^* with a three body rate of $3 \pm 1.5 \times 10^{-31} \text{ cm}^6/\text{sec}$; the third body was mainly argon.

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1. DISCHARGE MODELING OF THE XeF^* LASER

A. INTRODUCTION

Laser action in XeF^* was first achieved at AERL by pure E-beam pumping.⁽¹⁾ Subsequently Burnham et al.⁽²⁾ at NRL and Sutton et al.⁽³⁾ at Aerospace obtained laser action by UV preionization, avalanche discharge pumping. More recently we have obtained 40 mJ of laser energy in an E-beam ionized, avalanche discharge.⁽⁴⁾ We have also investigated in detail the discharge physics of the XeF laser containing approximately 99.5% Ar, 0.4% Xe and 0.1 - 0.2% NF_3 .

The physics of rare gas/halogen discharges is dominated by the excited species when the fractional population of the rare gas metastables exceeds 10^{-9} .⁽⁵⁾ For example, an important process in XeF laser discharges is the excitation of low lying metastables to higher lying states. This process can strongly influence the secondary electron energy distribution and therefore the efficiency of producing the metastables (which react with NF_3 to form XeF^* -- the upper laser level). The dominant ionization mechanism in XeF laser discharges is ionization of the xenon and argon metastables, i.e., two-step ionization. For this discharge, metastable ionization rates of $1-5 \times 10^7 \text{ sec}^{-1}$ are typical so that this process strongly influences discharge stability. These important excited state processes will be described in more detail subsequently.

B. SIMPLIFIED XeF KINETICS

In this section the dominant kinetics of the XeF laser discharge will be discussed. The high energy electrons $\vec{e}$ ionize the mixture forming mainly argon ions

The secondary electrons e_s are rapidly lost by attachment to NF_3 to form the negative halogen ion

The secondary electrons gain energy in the applied electric field and most of the discharge energy initially goes into producing argon metastable Ar^*

There are three principal reactions by which the argon metastables are lost

The rate constant for reaction (I-4) has been measured to be

1.8×10^{-10} and 3×10^{-10} cm^3/sec for $\text{Ar} (^3\text{P}_2)$ and $\text{Ar} (^3\text{P}_0)$ respectively. (6)

Reaction (I-5) has a three-body rate constant of 1.7×10^{-32} cm^6/sec for $\text{Ar} (^3\text{P}_1)$, 0.9×10^{-32} cm^6/sec for $\text{Ar} (^1\text{P}_1)$ and 1.6×10^{-32} cm^6/sec for $\text{Ar} (^3\text{P}_2)$. (7)

Assuming a gas kinetic mixing rate of these states by electrons, the loss rates of Ar^* by reactions (I-4) and (I-5) will be only marginally affected. So we will assume a mean rate of 2×10^{-10} cm^3/sec for reaction (I-4) and 10^{-32} cm^3/sec for reaction (I-5). The argon metastables react with NF_3 to form ArF^* with a rate constant of 1.4×10^{-10} cm^3/sec . (8) The

branching ratio for this reaction is yet to be determined. Extrapolating from reactions of Xe^* and Kr^* with NF_3 ⁽⁷⁾ one might conclude that reaction (I-6) has a branching ratio of about 0.5. At a mixture pressure of four atmospheres with 0.4% Xe and 0.1% NF_3 , the rates for reactions (I-4), (I-5) and (I-6) are 8×10^7 , 2.5×10^7 and $1.4 \times 10^7 \text{ sec}^{-1}$. So about 15% of the Ar^* is channeled into ArF^* . Most of the energy in ArF^* will produce XeF^* by the following reaction

The remainder of the energy is presumably radiated on the ArF^* bands. As reaction (I-7) proceeds with a rate constant $\sim 10^{-9} \text{ cm}^3/\text{sec}$, 75% of the ArF^* is to be channeled into XeF^* .

The argon excimers Ar_2^* will be deactivated at least as rapidly by Xe to form highly excited xenon states Xe^{**} . This means that 70-80% of the Ar_2^* will end up as Xe^{**} . The remainder of the Ar_2^* will be radiated. So we can conclude that the argon metastables transfer their energy to the xenon metastables with a 75% efficiency. The Xe^{**} thus formed will react with the NF_3 to form XeF^* . The branching ratio and rate constant for this reaction are not known. However, from our discharge data we have inferred a reaction rate constant of $4 \times 10^{-10} \text{ cm}^3/\text{sec}$ and a branching ratio of 0.7.

There are, of course, inefficiencies that arise from discharge pumping. The most severe are the electron impact excitation of the argon and xenon metastables

The rare gas metastables are similar to the alkalis and probably have large impact cross section to higher lying levels. Figure I-1 shows the expected cross section for Xe^* (Cs). Also shown in Fig. I-1 are the cross sections for the excitation of ground state Xe and Ar and the ionization of Xe^* (Cs).

C. DISCHARGE PHYSICS

As we have stated earlier, the physics of the discharge is strongly affected by the electron impact excitation and ionization of the rare gas metastables. To model these effects, we have treated the xenon metastables as cesium and the argon metastables as potassium, an analogy used successfully in predicting the emission spectra of rare-gas monohalides.

Some of the electron-impact cross sections used in our model are shown in Fig. I-1. The cross section for excitation from the 6s configuration to the 6p configuration in Cs (Xe^*) has a peak value of $90 \text{ \AA}^2$ at 8 eV. The peak value of the metastable-excitation is 40 times the peak value of the argon excitation cross section. More important, however, is the ability of most of the electrons to excite the 6s - 6p transitions -- which have a threshold of 1.5 eV -- but only the high energy tail of the electron energy distribution can produce metastables from the ground state.

In Figs. I-2, I-3 and I-4 we show the predictions of the Boltzmann code. Figure I-2 shows the percentage of energy that goes into producing Xe^* as a function of the fractional metastable population $\text{Xe}^*/(\text{Kr} + \text{Ar})$ for electric fields of 2-6 kV/cm atm. It is apparent from Fig. I-2 that the efficiency of producing the metastables is a strong function of the Xe^* population. For example, the efficiency of forming Xe^* is almost 45% when the fractional population is 10^{-5} and the electric field is 2 kV/cm atm. This

Fig. 1-1 Electron-Impact Cross Sections of Cesium (Xe^*) and Xenon as Functions of Electron Energy

Fig. 1-2 Percentage of Discharge Energy into Xe^* as a Function of Fractional Xe^* Population for Various Electric Fields

Fig. I-3 Ionization Rate as a Function of Fractional Xe^* Population

Fig. I-4 Average Electron Energy as a Function of Fractional Xe^* Population

efficiency decreases to less than 10% when the fractional population is increased to 10^{-4} . The decrease in efficiency can be made up by increasing the electric field. Figure I-3 shows the ionization rate as a function of the fractional metastable population. Figure I-4 is a plot of the average electron energy as a function of the fractional metastable population. Notice that the electrons cool as the metastable population increases. The cooling effect is much stronger at smaller electric fields.

Using the rate constants predicted by the Boltzmann code, we have developed a *self-consistent kinetics code* that follows the temporal evolution of the secondary electrons, positive and negative ions, Ar^* , Ar_2^+ , Xe^{**} and XeF^* . We couple our kinetics code to a simultaneous set of differential equations that describe the electrical circuit. The outputs of this code include the temporal evolution of the discharge current and voltage and the XeF^* fluorescence for a given preionization level, capacitor charge voltage and gas mixture.

The predictions of this discharge model have been compared with our XeF^* laser discharge experiments. The top trace in Fig. I-5 is the E-beam current in the discharge cavity. The lower trace is the fluorescence as observed by a photomultiplier after the signal passes through a 1/4 meter Jarrel Ash monochromator tuned to 3520 Å. The cavity was filled with a 1 atm mix of 99.4% Ar, 0.4% Xe and 0.2% NF_3 . The dashed trace is the prediction of the code. The amplitude of this predicted trace is adjusted to closely match the measured fluorescence. This amplitude normalization was necessary as we did not have an absolute calibration on the fluorescence emanating from the discharge. For subsequent comparisons between experiment and theory no further adjustments were made.

FLUORESCENCE CALIBRATION

Fig. I-5 Measured and Predicted Fluorescence for Pure E-Beam Pumping.
The discharge cavity contained 99.4% Ar, 0.4% Xe and 0.2% NF_3 .

Once the XeF^* fluorescence amplitude was normalized, we measured the magnitude and efficiency of discharge produced XeF^* fluorescence enhancement. Figure I-6 shows the experimental results and theoretical predictions when the $0.3 \mu\text{F}$ capacitor is charged to 8 kV. The top trace is the discharge voltage. The second trace is the discharge current. The third trace is the XeF^* fluorescence. By the end of the pulse the enhancement in the fluorescence is 2.5. The metastables are being produced with the same efficiency as by pure E-beam pumping. Note that the predicted fluorescence is 30% higher than the measured fluorescence. The reduced measured fluorescence can be explained if the branching ratio from Xe^{**} (see reaction I-4) is 0.7. Further, to predict the current in the discharge we have used a rate constant for reaction (I-4) of $4 \times 10^{-10} \text{ cm}^3/\text{sec}$. If a slower rate constant is used the predicted discharge current does not reach a steady state value.

D. CONCLUSIONS

In conclusion, our discharge model predicts that rare gas metastables can be produced with high efficiency (60%) as long as the fractional metastable population is kept sufficiently small ($< 2-3 \times 10^{-5}$). We have also inferred that Xe^{**} reacts with NF_3 to produce XeF^* with a branching ratio of 0.7. The rate constant for this reaction is $4 \times 10^{-10} \text{ cm}^3/\text{sec}$.

Fig. I-6 Measured and Predicted Fluorescence When Capacitor is Charged to 8 kV

II. FORMATION AND QUENCHING OF XeF^*

A. INTRODUCTION

Efficient scaling of the XeF^* laser⁽¹⁻⁵⁾ to high average power requires knowledge of the processes responsible for the formation and quenching of the upper laser level. From the formation kinetics one can determine the upper state production efficiency. The quenching kinetics enables one to choose the appropriate mix and determine the laser saturation flux.⁽⁹⁾ For efficient laser power extraction the cavity flux should be much greater than the saturation flux. In this section we report on measurements of the quenching rates of XeF^* by Ar and Xe. These rates were obtained by analyzing the dependence of XeF^* fluorescence intensity on the partial pressures of Ar, Xe and F_2 . We have determined the rate constants for the following processes:

The third body M was mainly argon.

B. EXPERIMENTAL APPARATUS

Argon, xenon and fluorine were premixed and excited by a high energy E-beam. The resulting XeF^* fluorescence amplitude was measured by a photodiode via a 5 nm bandpass filter centered at 352 nm. To ensure that the emission was representative of the overall kinetic processes, we repeated the experiments with a 50 nm bandpass filter. This changed the

amplitude of the photodiode signal by a constant factor over the whole pressure range of interest. Hence we concluded that the attenuation of XeF^+ ($\text{B}^2\Sigma_{1/2}^+ \rightarrow \text{X}^2\Sigma_{1/2}^+$) radiation observed through the 5 nm bandpass filter was constant when the mixture pressure was varied. The mixtures used contained mainly Ar, 7.6 torr F_2 , and varying amounts of Xe (from 7.6 torr to 150 torr). For a given run however we kept the xenon concentration fixed and increased the argon partial pressure from 0.5 atmospheres up to 4 atmospheres because at lower pressures the spectra changed radically.⁽⁸⁾ This change is possibly due to the lack of collisional relaxation of the manifold of upper states at low gas pressures.

Research grade (Matheson) Ar and Xe were used without any further purification. The gases were analyzed by Gollob Analytical Service, Inc., and found to have less than 100 ppm of O_2 , N_2 , H_2O and CO_2 impurities. The F_2 was 98% pure. The electron gun apparatus used in these experiments has been described previously.⁽⁴⁾ This device produces an electron beam with an energy of 150 keV and a current density of $0.2 - 5 \text{ A/cm}^2$ for 300 nsec. The cross sectional dimensions of the electron beam were 2 cm x 22 cm. The beam interacted with the mixture in a teflon cell 0.5 cm deep in the beam direction; this short dimension insured a linear increase in the energy deposition with pressure for pressures up to 4 atmospheres.

C. ANALYSIS OF DATA

Table II-1 lists the dominant formation kinetics. The high energy electrons deposit their energy into F_2 , Xe and Ar in a ratio roughly proportional to their mass densities. One important question in analyzing the experimental data is to decide the major kinetic pathways of the positive ions Ar^+ and Xe^+ . Reaction (II-3) gives a formation rate for ArF^+ that is proportional to

TABLE II-1

DOMINANT FORMATION REACTIONS

$[F^-] [Ar^+]$. Since $[F^-] \sim [Ar^+] \sqrt{I_{eb}}$ (I_{eb} = E-beam current) one expects that ArF^* formation and hence XeF^* (via reaction II-6) by this path to be proportional to I_{eb} . Similarly reaction (II-7) gives a production rate of XeF^* linearly proportional to I_{eb} . The data showed XeF^* fluorescence to vary linearly with E-beam current from 0.2 A/cm^2 to 5 A/cm^2 for the full range of mixtures and pressures sued, which is thus consistnet with the foregoing discussion. It is also known, however, that reaction (II-4) and (II-8) are significant sinks for Ar^+ and Xe^+ , especially at the higher pressures. Only by hypotheisizing reactions (II-5) and (II-9) can we maintain a total mechanism of ion chemistry which leaves XeF^* , and hence its fluorescence, linearly proportional to I_{eb} . Thus alternate species such as Ar_2F^* and Xe_2F^* cannot be major products of reactions (II-5) and (II-9).⁽¹⁰⁾

Once XeF^* is formed, it can radiate (with spontaneous lifetime of 16 nsec)⁽¹¹⁾ or be quenched by F_2 , Xe or Ar. Recently Brashears, Setser and DesMarteau⁽¹²⁾ have measured the quenching rate constants of XeF^* by F_2 and Xe and found them to be $3.3 \times 10^{-10} \text{ cm}^3/\text{sec}$ and $2.9 \times 10^{-11} \text{ cm}^3/\text{sec}$ respectively. All processes involved reach a steady state on a time scale much less than the 300 nsec E-beam pulse. So the photodiode signal can be written as

$$S = \frac{\alpha + \alpha_{Ar} P_{Ar}}{\gamma + \xi_{F_2} P_{F_2} + \xi_{Xe} P_{Xe} + \xi_{Ar} P_{Ar} + \delta_{Xe} P_{Xe} P + \delta_{Ar} P_{Ar} P} \quad (II-1)$$

where P_{F_2} , P_{Ar} and P_{Xe} are the pressures of F_2 , Ar and Xe respectively. P is the total pressure, α_{Ar} is proportional to the efficiency of generation of XeF^* from the E-beam energy deposited in the Ar. α is the contribution to the XeF^* formation from the E-beam energy deposited in Xe. Both α and

α_{Ar} are constants for constant P_{Xe} and P_{Ar} . γ is the inverse spontaneous lifetime. ξ_{F_2} , etc., are the two body quenching rate constants of F_2 etc. δ_{Xe} and δ_{Ar} are the three body quenching rate constants by argon and xenon. We have ignored the three body quenching by F_2 because of the low fluorine concentrations used. For example, a three body rate constant of 10^{-30} cm⁶/sec would lower the measured two body rate constant by Ar by 20%. It is very difficult to obtain this rate constant because of the rapid quenching of XeF^* by F_2 .⁽¹²⁾

For each experimental run the signal intensity was measured as a function of P_{Ar} keeping P_{F_2} and P_{Xe} constant. (A run consists of a series of experimental data for various values of P_{Ar} .) Seven different runs were made for $P_{Xe} = 7.6$ up to 152 torr. For each run the data so obtained was fitted to a polynomial

$$S = \sum_n A_n P_{Ar}^n \quad (II-2)$$

In Eq. (II-2) we have assumed $P_{Ar} = P$. Typically it was found that a 3rd order polynomial adequately fitted the data. We are only interested in the first two coefficients A_0 and A_1 given by

$$A_0 = \alpha / \zeta \quad (II-3)$$

and

$$A_1 = \frac{\alpha_{Ar}}{\zeta} - \frac{\eta \alpha}{\zeta^2} \quad (II-4)$$

We have abbreviated $\zeta = (\gamma + \xi_{F_2} P_{F_2} + \xi_{Xe} P_{Xe})$ and $\eta = \xi_{Ar} + \delta_{Xe} P_{Xe}$. By using Brashears, Setser and DesMarteau's results⁽¹²⁾ for ξ_{F_2} and ξ_{Xe} and assuming $\eta \alpha / \zeta \alpha_{Ar} \ll 1$ (this assumption, which allows neglecting the second term in Eq. (II-4), was verified a posteriori), α and α_{Ar} were then calculated for each run from (I-3) and (II-4).

We now define a quantity $L(P_{Ar})$ given by

$$L(P_{Ar}) = \frac{a + a_{Ar} P_{Ar}}{S} - \xi \quad (II-5)$$

From Eq. (II-1)

$$L(P_{Ar}) = \eta P_{Ar} + \delta_{Ar} P_{Ar}^2 \quad (II-6)$$

By at least squares fit, of a quadratic in P_{Ar} , to the experimental values of $L(P_{Ar})$, η and δ_{Ar} were evaluated for each run. By plotting η vs P_{Xe} , we obtained values for ξ_{Ar} and δ_{Xe} .

D. RESULTS AND CONCLUSIONS

Figure II-1 shows the temporal variation of the E-beam current and XeF^* fluorescence as detected by the photodiode. Figure II-2 shows a typical curve of the measured photodiode signal as a function of Ar pressure for the special case of 22.8 torr Xe and 7.6 torr F_2 . Figure II-3 shows a reduction of the data shown in Fig. II-2 to the form given by (II-6). Also shown is at least squares fit to the reduced data to obtain the value of η and δ_{Ar} . Finally, in Fig. II-2, Eq. (II-1) is plotted with the derived values of a_{Ar} , ξ_{Ar} , etc. From these derived values we have also determined that for this case $(\eta a)/(\xi a_{Ar}) \approx 10^{-2}$. The value of δ_{Ar} was obtained by averaging the results of the many different runs performed. The large possible errors in the measured rates is because the loss of XeF^* by two and three body quenching are the same order of magnitude over our range of experiments.

Using the quenching rates given above, one can compute the saturation flux ϕ_s for an arbitrary mixture of Ar/Xe/ F_2 from the following expression⁽¹³⁾

Fig. II-1 Typical Experimental Data. The top trace is the E-beam current and the lower trace is the photodiode response.

Fig. II-2 The Open Circles are the Experimental Measurements of the XeF^+ Radiation Detected by a Photodiode. Each experimental value shown above is the average of many data points. The signal was through a 5 nm bandpass filter centered at 352 nm. The curve is a plot of Eq. (II-1).

Fig. II-3 The Open Circles are the Data Shown in Fig. II-2 Reduced to Eq. II-5. The curve is a least squares fit to a quadratic in P_{Ar} .

$$\phi_s = \frac{h\nu\gamma}{\sigma_s} \left[1 + \left(\xi_{F_2} P_{F_2} + \xi_{Xe} P_{Xe} + \xi_{Ar} P_{Ar} \right) \gamma^{-1} + \left(\delta_{Xe} P_{Xe} + \delta_{Ar} P_{Ar} \right) P \gamma^{-1} \right] \quad (II-7)$$

where σ_s is the stimulated cross section and $h\nu$ is the photon energy. From the XeF^* spontaneous spectra we have estimated that $\sigma_s \approx 4 \times 10^{-16} \text{ cm}^2$. Hence we can evaluate $h\nu\gamma/\sigma_s$ to 0.084 MW/cm^2 . For a typical laser mix containing 0.1% F_2 , 0.5% Xe and 99.4% Ar at a total pressure of two atmospheres, one obtains $\phi_s \approx 0.13 \text{ MW/cm}^2$. It should be noted that this saturation flux is correct in the limit that the ground state population is negligible.

In most E-beam pumped XeF^* laser experiments performed to date NF_3 has been used as the halogen bearing compound. (14, 15, 16) Brashears et al. (12) have determined that the NF_3 quenching rate of XeF^* is 16-17 times slower than F_2 . So ϕ_s for a mix containing NF_3 will be somewhat smaller than the value calculated above. Further, NF_3 does not absorb at 350 nm. (17) In spite of these two considerations, the highest laser efficiencies obtained in XeF^* have been about 5%. (16, 17) The maximum expected efficiency is 17%. (18) A possible explanation for the lower efficiencies obtained experimentally is photoabsorption due to charged particles (for example F^-) (19) and excited states (for example Xe^* , Ar^* , etc.). (20)

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