

# Broadband Tunable Excimer Lasers

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In recent years, there has been considerable interest in the development and use of excimer lasers for a variety of applications. The word *excimer* is a contraction of "excited dimer," which refers to an excited diatomic molecule, composed of two atoms of the same type. In fact, almost all of the important excimers are actually "exciplexes" in the sense that they are composed of two different atoms. The most important of these excimers consists of a rare-gas halogen combination, such as xenon fluoride or krypton fluoride. In the ground state, these molecules are very unstable, and the two atoms dissociate very quickly. However, if there is available a positively charged ion, such as  $\text{Xe}^+$ , and a matching negatively charged ion, such as  $\text{F}^-$ , then the electrostatic attraction between the two can cause them to come together forming an excited excimer molecule, such as  $\text{XeF}^*$ . This molecule looks and acts very much like an alkali-halide molecule, such as  $\text{NaCl}$  or  $\text{KF}$ . The excited excimer state is metastable in the sense that it can exist for several hundreds of nanoseconds before the extra electron on the  $\text{F}^-$  ion returns to the  $\text{Xe}^+$  ion, and the molecule dissociates. During such an electronic transition, the energy of the molecule is reduced, and the excess energy can be emitted in the form of a photon. The photon can be emitted by spontaneous emission, or, if the excimer molecule is inside an optical cavity, by stimulated emission, thus giving rise to laser action.

Most excimer lasers in existence today operate on relatively narrow-band transitions in the violet and ultraviolet region of the spectrum. The most important of these is the  $\text{ArF}$  laser at 193 nm,  $\text{KrF}$  at 249 nm,  $\text{XeCl}$  at 308 nm, and  $\text{XeF}$  at 351 nm. All of these laser species may be conveniently excited by electric discharge and have seen numerous scientific, industrial, and medical applications in the past few years. Pulse energies on the order of 1 J, with a 10 to 100 ns pulse width, are typical for current commercial excimer lasers. Electric to optical conversion efficiencies are on the order of 1%. Recently, an electron beam pumped  $\text{KrF}$  laser, "Aurora," at the Los Alamos National Scientific Laboratory, delivered 3000 J with a pulse width of 400 ns.

There are also several excimer lasers that possess a broadband output emission that extends over several tens of nanometers. One of the most promising of this class of excimer laser is the  $\text{XeF}$  ( $\text{C} \rightarrow \text{A}$ ) laser. In order to understand the difference between the narrow-band and the broadband excimers, one must examine the energy-level diagram for such molecules, as depicted in Fig. 1. The rare-gas halogen excimers

actually have two first excited states labeled B ( $\Omega = 1/2$ ) and C ( $\Omega = 3/2$ ). There are also two ground states, X ( $\Omega = 1/2$ ) and A ( $\Omega = 3/2$ ). In most excimers, the C state lies sufficiently higher than the B state, so that at room temperature, it is essentially empty. Selection rules dictate that a radiative transition can occur between the B and X states, or between the C and A states. Since the B state is the only populated upper level, the  $\text{B} \rightarrow \text{X}$  transition is the only one observed for most excimers.  $\text{XeF}$  is unique, however, in the fact that the C state lies slightly below the B state. There is a small binding energy in  $\text{XeF}$ , even for the neutral X ground state, and as a result, the  $\text{B} \rightarrow \text{X}$  transition is relatively narrow banded. The A state is highly repulsive, however, and even a small deviation from the bottom of the potential well in the C-state energy curve will result in a significant wavelength shift on the  $\text{C} \rightarrow \text{A}$  transition. Also, because of the steep rise of the A-state energy curve above that for the X state, the  $\text{C} \rightarrow \text{A}$  emission band is shifted toward the long wavelength region of the spectrum, as compared to the  $\text{B} \rightarrow \text{X}$  transition.

There is yet another class of excimers that also exhibits broadband emission. These are the triatomic excimers such as  $\text{Kr}_2\text{F}$  at  $435 \pm 30$  nm and  $\text{Xe}_2\text{Cl}$  at

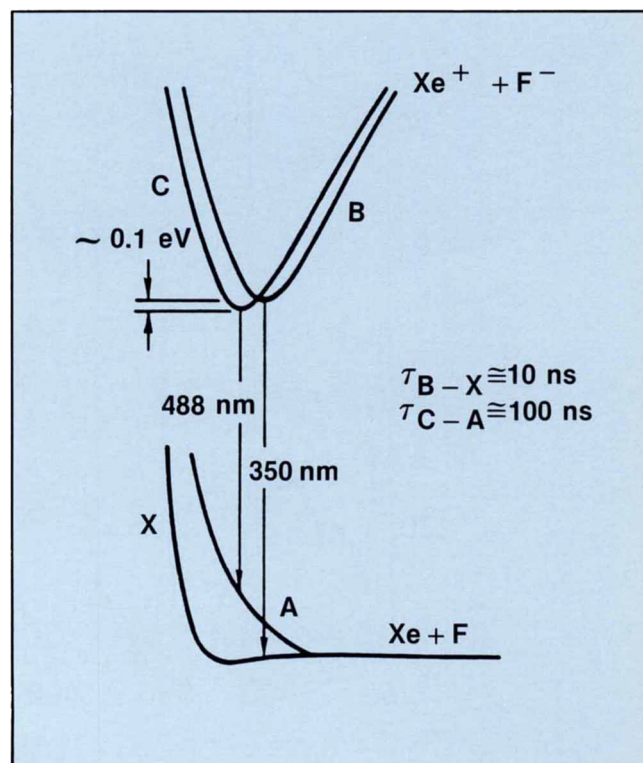


Fig. 1 Energy as a function of interatomic separation for the excimer  $\text{XeF}$ , showing both the  $\text{B} \rightarrow \text{X}$  and the  $\text{C} \rightarrow \text{A}$  transitions.

520  $\pm$  35 nm. Their emission bands, as well as those for some other trimers, which have not been made to lase, are depicted in Fig. 2. Shown also in this figure are the narrow-band emissions from several of the diatomic excimers. A typical energy-level diagram for a triatomic excimer is shown in Fig. 3. In the trimers, the ground state is always highly repulsive, and as a result, the emission is characteristically broadbanded. The trimers are formed via a reaction between the excited diatomic molecule, such as KrF\* and a second rare-gas atom. The reaction also requires a third body, such as Ar, in order to conserve energy and momentum. Because this formation process requires a three-body reaction, it only takes place at relatively high pressures.

Because these lasers are wavelength tunable, and because they operate in the blue-green to blue region of the electromagnetic spectrum, they have several potential applications. One of these is for underwater optical communications and ranging. The transmission of seawater is at a maximum near 480 nm, and, thus, blue-green laser light is ideal for underwater communications with submarines. Intense, wavelength tunable excimer lasers in the visible may also have numerous other potential applications in laser isotope separation, surface chemistry for semiconductor processing, and in laser-assisted chemistry. Many chemical activation energies, as well as impor-

tant absorption bands, lie within the visible region. Because the absorption bands are very narrow, a tunable laser is often the only practical method for getting just the right wavelength for a particular application.

In order for these lasers to be of practical use, they must meet certain performance criteria. For communications between a satellite and an underwater submarine, somewhere between 1 and 10 J of energy will be required per pulse. In order to keep the laser volume to some reasonable size, the output energy density should be about 1 J/liter. If one assumes a 1,000 Hz pulse rate at 5 J/pulse and a 1% conversion efficiency, it would take 500 KW to power the laser, not to mention that one would need to provide 495 KW of cooling! Laser efficiency is a very important consideration for such a system and is one of the most important criteria for evaluating a particular laser candidate.

Excitation of the tunable rare-gas halide excimers has been accomplished in several ways. Among these are direct electrical discharge, electron beam pumping, and photolytic or optical excitation. Because these excimer lasers are characteristically broadband, their optical gain is quite low. In the absence of any losses, the small-signal gain per unit length,  $g$ , of an excimer may be calculated from the expression

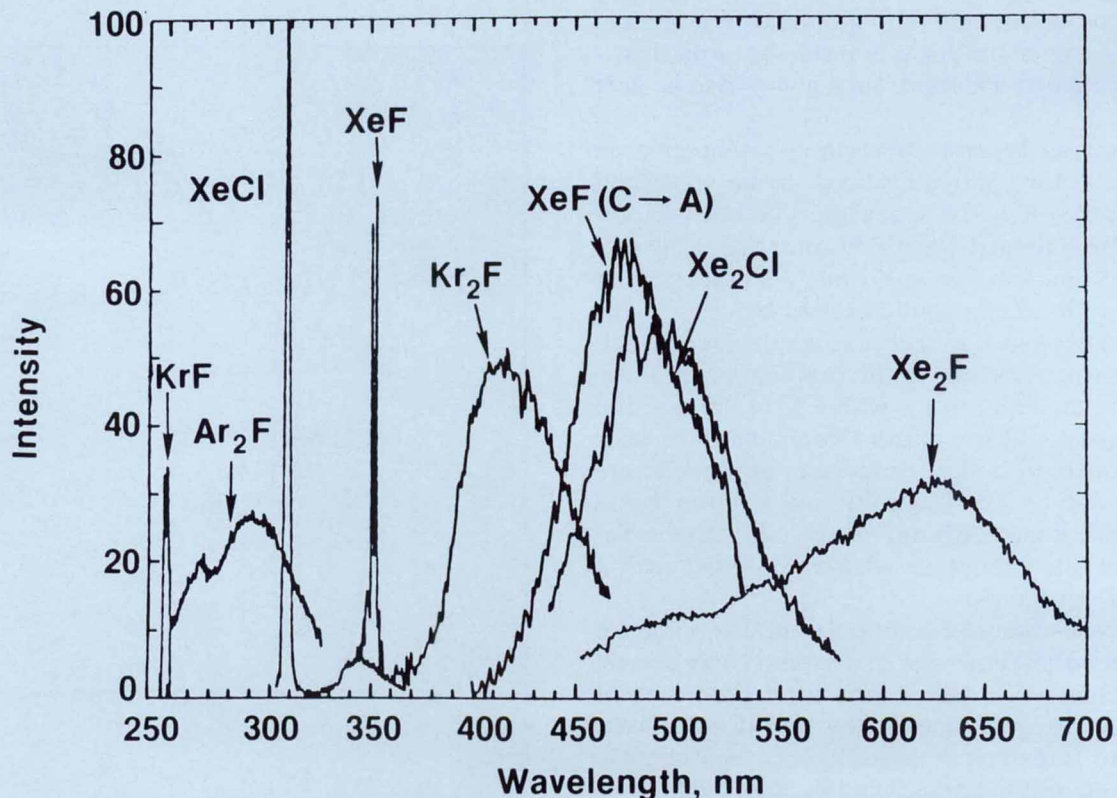


Fig. 2 Fluorescence spectra of several diatomic and triatomic excimers.



$$g = \sigma N^* = \frac{\lambda^4}{8\pi c \tau \Delta\lambda} N^*$$

Here,  $\sigma$  is the stimulated emission cross section, and  $N^*$  is the excited state number density.  $\lambda$  is the wavelength of the peak of the fluorescence emission curve, and  $\Delta\lambda$  is its full width at half maximum.  $\tau$  is the excited state lifetime, and  $c$  is the speed of light. The fluorescence emission curve for  $\text{XeF}(\text{C} \rightarrow \text{A})$  is shown in Fig. 4. It has a  $\Delta\lambda$  of about 70 nm, with a center wavelength near 485 nm. Since the excited state lifetime is about 100 ns, the stimulated emission cross section,  $\sigma$ , is about  $1 \times 10^{-17} \text{ cm}^2$ . Thus, in order to obtain a gain of 1% per cm (without even considering the effect of possible absorbers in the laser medium), one must achieve an excited state population of about  $1 \times 10^{15} \text{ cm}^{-3}$ . This requires about 4 MW/liter of pumping power just to account for the losses due to spontaneous emission. Conversion inefficiencies through various loss mechanisms can increase this number by a factor of 10 or more. Thus, in order to achieve laser action with one of these broadband excimers, very large pumping powers are required.

One of the most effective ways of achieving large excitation energy densities is through the use of a high-current relativistic electron beam (e-beam). An e-beam generator consists of two main components. The first of these is a Marx-bank, which is a set of

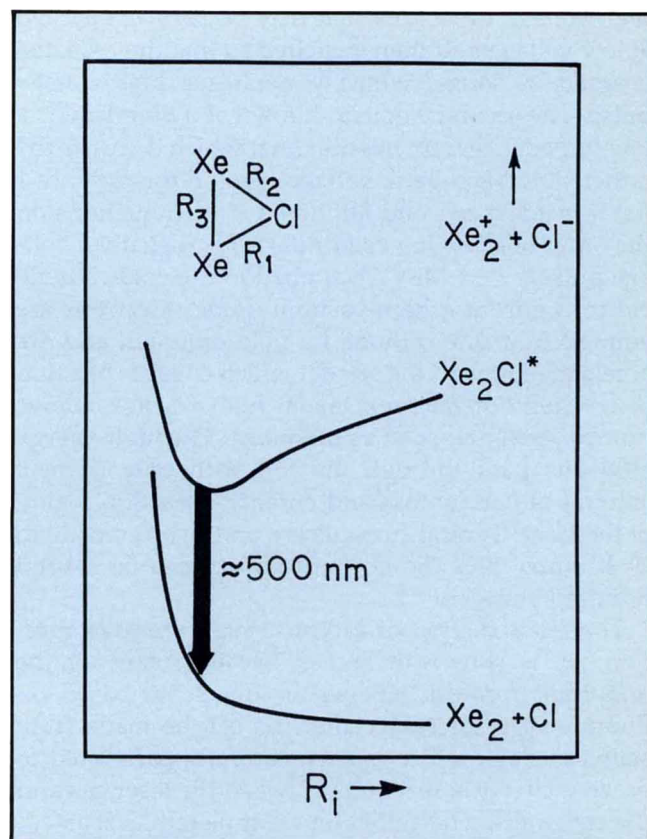


Fig. 3 Energy-level diagram of the triatomic rare-gas halogen excimer  $\text{Xe}_2\text{Cl}$ .

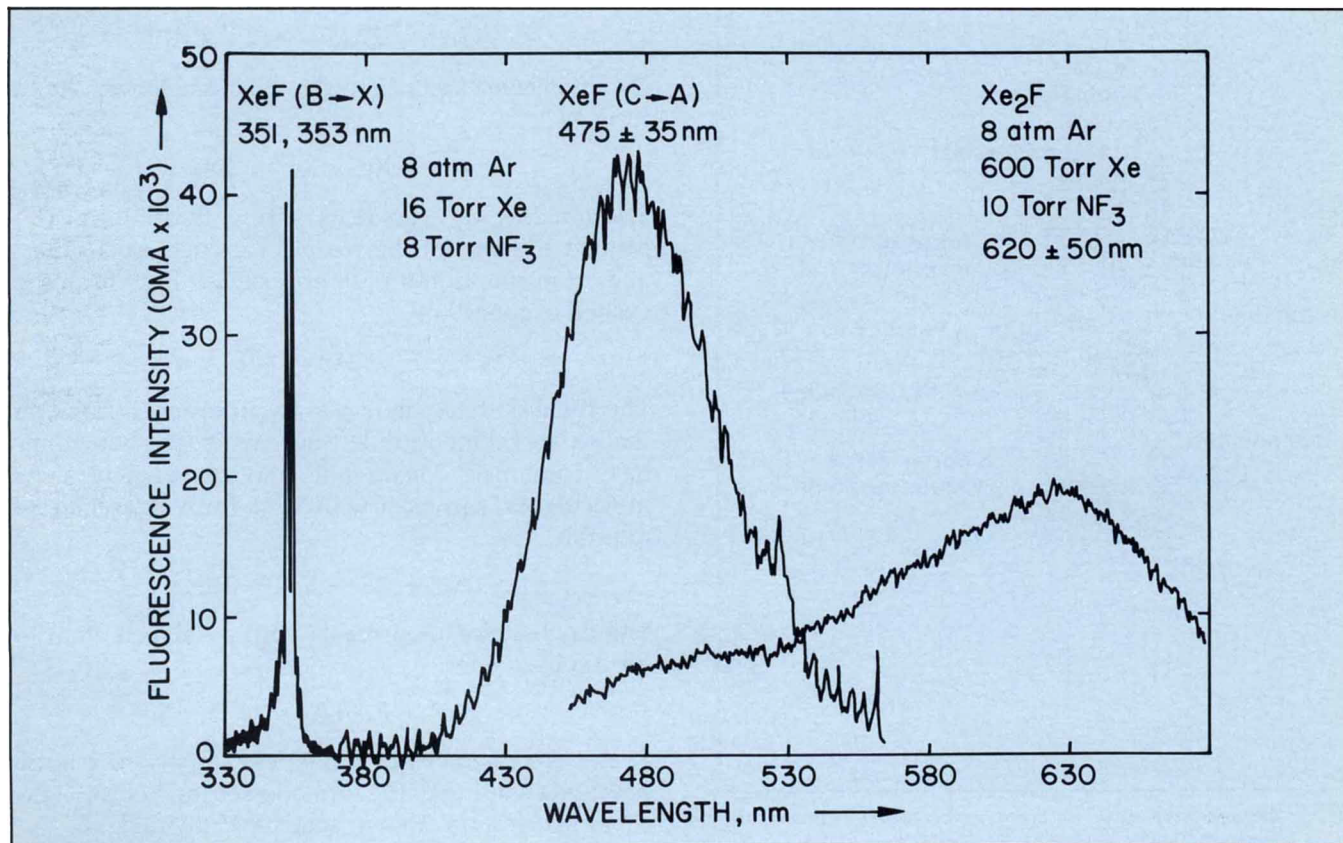


Fig. 4 The fluorescence emission curve for  $\text{XeF}(\text{C} \rightarrow \text{A})$ . The center wavelength is at about 475 nm, and the width of the emission curve at half-maximum is 70 nm. Also shown in the figure are the emission spectra for the narrow-band  $\text{B} \rightarrow \text{X}$  transition and the triatomic  $\text{Xe}_2\text{F}$  emission.

high-voltage capacitors that may be parallel charged at low voltage and then switched so that they are discharged in series, which generates a high-voltage pulse. The second element, known as a Blumlein, is a low-impedance transmission line, which converts the rather slow Marx-bank voltage-rise (on the order of 1  $\mu$ s) into a fast rise and fall time pulse (transitions on the order of a few nanoseconds). This (negative) voltage pulse ( $\sim -1$  MeV) is applied to the cold-cathode electron gun of a high-vacuum diode. Electrons are emitted from the cathode by field emission and are accelerated toward the anode, which consists of a thin (a few tens of microns) metal foil of a low atomic number material (such as titanium). The high-energy electrons pass through the foil with only a small amount of energy loss and enter the reaction region of the laser. Typical currents are on the order of 10 to 20 K amps, and the current density can be several hundred amps/cm<sup>2</sup>.

The cross section of a typical high-pressure reaction cell is shown in Fig. 5. Because many of the important rare-gas halogen excimers are based on fluorine, it is necessary that the cell be made from stainless steel, which is then carefully passivated to prevent chemical reactions between the laser gas and the cell walls. The cell is first completely evacuated, and then a careful mix of various gases (such as Xe,

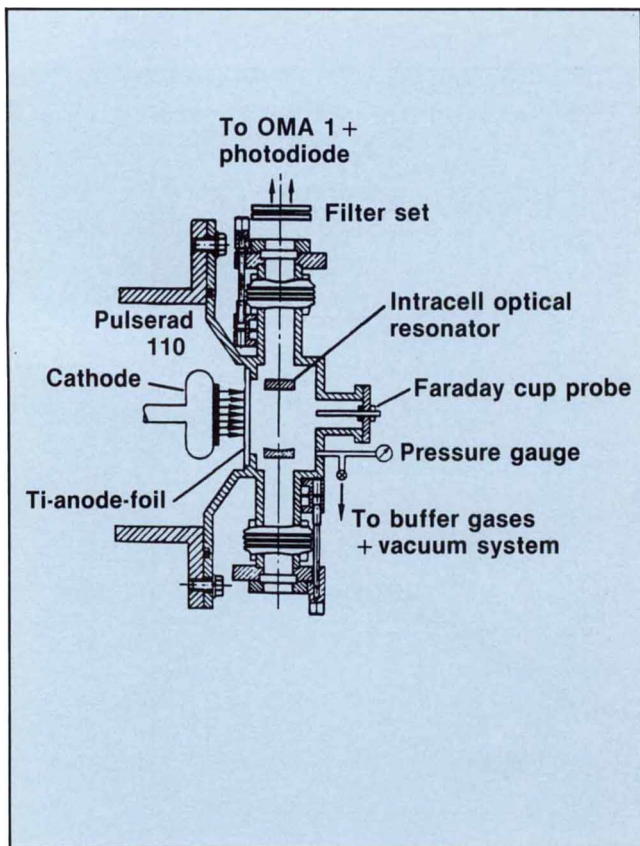
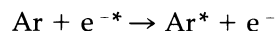
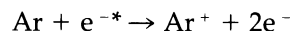


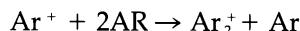
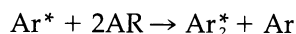
Fig. 5 Cross-sectional view of a high-pressure e-beam excited excimer laser cell. The Pulserad 110 is the electron beam accelerator. The optical cavity is located within the cell and can be aligned by means of external adjustments.

Kr, NF<sub>3</sub>, F<sub>2</sub>, and Ar) is admitted, with the final pressure in the six to ten atmosphere range. The high-energy e-beam interacts with the laser gases, creating various excited species, which then react to form the upper level excimers. A pair of mirrors located within the cell form the optical cavity and provide the feedback and coupling necessary to make the laser operate. Wavelength tuning can be achieved with either a prism or a diffraction grating as one of the reflecting elements. In order to achieve very narrow linewidths (0.1 cm<sup>-1</sup>), an injection locked oscillator-amplifier scheme has been used.

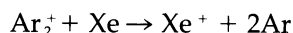
The laser gas mixture for most rare-gas halogen excimer lasers consists of a large fraction of a relatively light background or buffer gas (often helium, neon, or argon) into which smaller amounts of the active rare gas (Xe or Kr) and the halogen donor (NF<sub>3</sub>, F<sub>2</sub>, or CCl<sub>4</sub>) are added. For instance, for an e-beam excited XeF(C→A) laser, a typical gas mixture might consist of 6 atm Ar, 16 Torr Xe, and 8 Torr NF<sub>3</sub>. When the high-energy electrons (e<sup>\*</sup>) enter the reaction cell, they collide primarily with the buffer gas, forming excited buffer gas species



The excited neutral or ionized Ar can then react further yielding



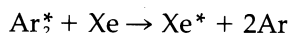
The molecular argon ion, Ar<sub>2</sub><sup>+</sup> then reacts with Xe to form Xe<sup>+</sup>



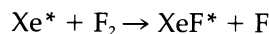
The ionized Xe<sup>+</sup> can react with a fluorine ion (F<sup>-</sup> formed by reaction between an excited electron e<sup>-</sup> and a fluorine donor F<sub>2</sub> + e<sup>-</sup> → F<sup>-</sup> + F) to form the excited excimer XeF.



The third body Ar is necessary in order to carry off the excess momentum left over when the two atoms have combined. Meanwhile, the excited neutral Ar molecule Ar<sub>2</sub><sup>\*</sup> can react with Xe to form an excited Xe neutral



The excited Xe<sup>\*</sup> then reacts with a halogen atom to form the excimer



These, and somewhat other less important energy pathways leading to the formation of the XeF(B,C) excimer species, are shown schematically in Fig. 6.

Evidence suggests that the XeF excimer is formed primarily in the B state, perhaps in highly excited



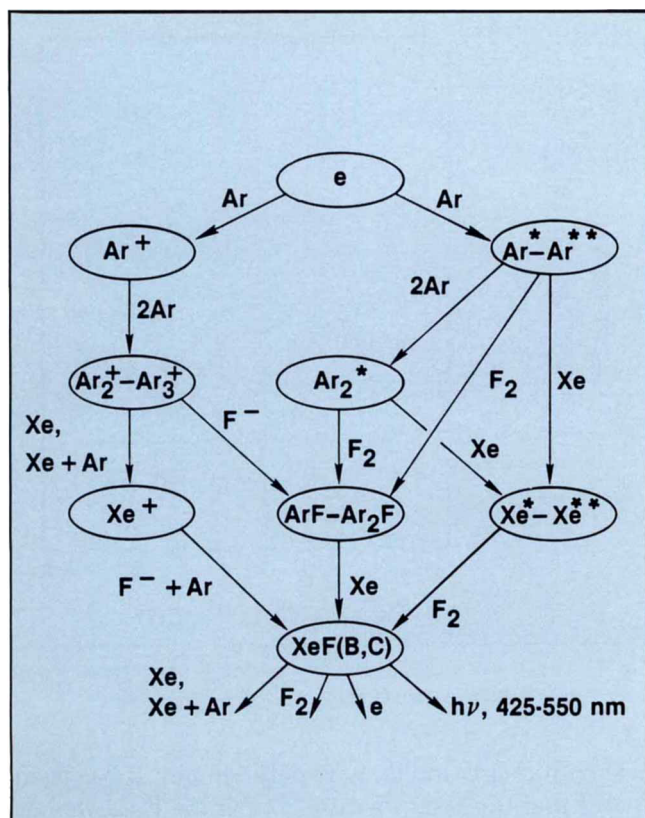


Fig. 6 Dominant formation kinetics of e-beam pumped XeF (B,C). The input energy enters at the top of the reaction chain in the form of relativistic electrons. Reactions occur as indicated by the arrows, utilizing the species alongside each arrow to form the products enclosed in the ovals.

vibrational levels. Rapid relaxational quenching by both the buffer gas, as well as secondary electrons from the e-beam, quickly relax the molecule to its lowest vibrational state, however. The same species also create rapid mixing and thermalization between the B and C excited states. In just a few nanoseconds, most (over 95% at 300° degrees F) of the XeF exists as XeF(C) and is available for laser action. Overall, the channeling of electron beam input energy into excited XeF(C)\* can occur with an efficiency from 5% to 10%.

Unfortunately, subsequent processes are active in quenching the XeF and removing it from the system. A three-body reaction among XeF, Xe, and Ar can occur, leading to the formation of the trimer Xe<sub>2</sub>F



In fact, reactions of this type are the most important formation channels for the triatomic excimers and are used extensively whenever a trimer species is desired. They represent a loss channel for XeF, however, and thus limit the concentration of Xe, which one can tolerate in the laser gas mixture.

Various other species in the gas mixture can also quench, or remove, XeF. The most significant of these is quenching by the halogen donor gas (NF<sub>3</sub> or F<sub>2</sub>). Both of these gases have a large quenching rate constant for XeF. This leads to a trade-off with regard to

both Xe and halogen donor concentration. Too little of either, and not enough XeF is formed for optimum laser operation. Too much, and the XeF, which is produced, is too quickly removed by quenching. Thus, each gas has its individual optimum concentration, which is the best average between these two competing functions: formation and quenching. The buffer gas, argon, is a mild quencher of XeF, and removal by this species is usually not important when compared to the other quenchers. Removal by secondary electrons is not entirely negligible, and an improvement in laser performance can be obtained whenever the electron concentration is reduced.

Once the excited XeF has been relaxed into the C state, it may then undergo a transition to the A state, giving off a photon as it does. This then is the basic kinetic chain leading from the input electron beam to the emitted blue-green photons. If one looks at the temporal relationship between the excitation pulse, the fluorescence emission in the visible, and the XeF(C→A) laser emission, one notices a striking feature. Whereas the fluorescence pulse reaches its peak at about the time the excitation pulse has ended (indicating the buildup of excited XeF comes about as a result of direct integration of the excitation energy), the laser emission is delayed by a significant amount of time from the fluorescence. This temporal relationship is shown in Fig. 7 for a typical XeF(C→A) laser. The reason for the long delay in the laser emission buildup becomes clear when one takes a look at the optical gain evolution with time for an e-beam excited XeF(C→A) laser.

Figure 8 shows the gain as a function of time for two different gas mixtures in the cell. In these experiments, an argon ion laser beam at 488 nm is sent through the active region of the laser cell. By measuring the increase or decrease of the intensity of the

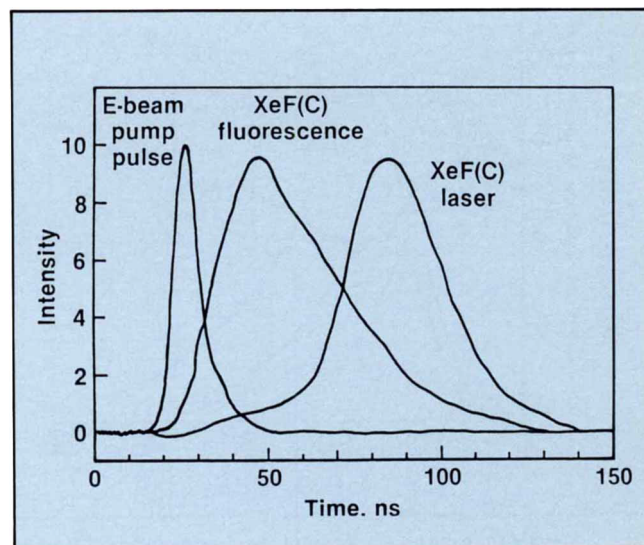


Fig. 7 Normalized temporal characteristics of the e-beam excitation pulse (as measured by a Faraday cup within the reaction cell), the XeF(C) fluorescence emission, and the XeF(C→A) laser pulse.

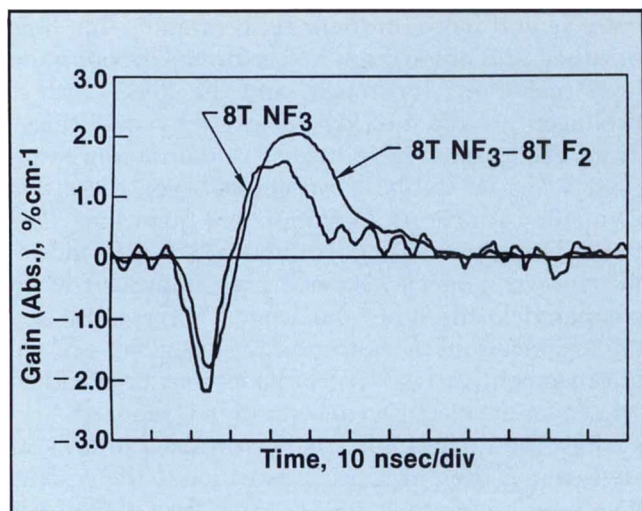


Fig. 8 Temporal evolution of the net gain of the XeF(C→A) laser as measured with a 488 nm Argon ion laser probe beam. The cell contained 16 Torr of Xe, 6 Atm of argon, and, in one case, 8 Torr of F<sub>2</sub> and, in the other, 8 Torr each of F<sub>2</sub> and NF<sub>3</sub>.

transmitted beam, the gain or absorption at any given instant can be measured. Both mixtures show an initial strong *negative* gain, or absorption, followed by a period of positive gain, during which the laser pulse can build up out of spontaneous emission. For the gas mixture containing only NF<sub>3</sub>, the period of positive gain exists for only about 20–25 nsec, and the peak gain is only about 1.5% cm<sup>-1</sup>. When the combined mixture of 8 Torr NF<sub>3</sub> and 8 Torr F<sub>2</sub> is used, the gain lasts for about 40 nsec, and increases to about 2% cm<sup>-1</sup>. With a low gain laser, such as this broadband excimer, a change of even 1/2% cm<sup>-1</sup> in gain can have a significant effect on the laser output. This behavior

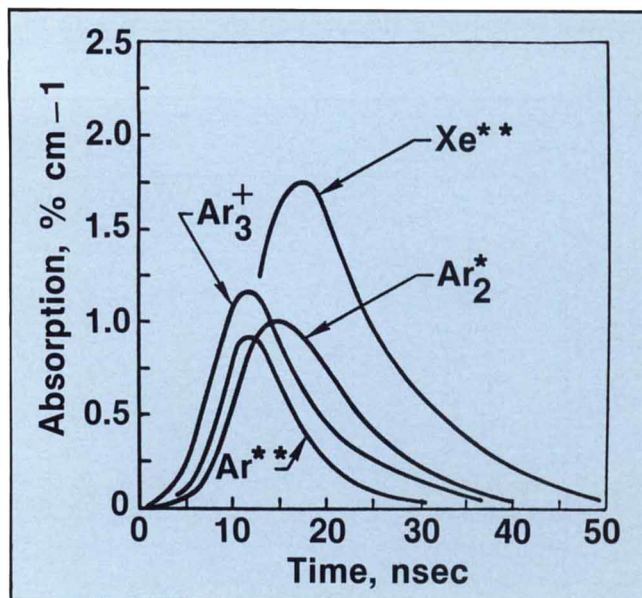


Fig. 10 Computed temporal evolution of the absorption caused by several of the important absorbing species in an e-beam excited XeF(C→A) laser mixture.

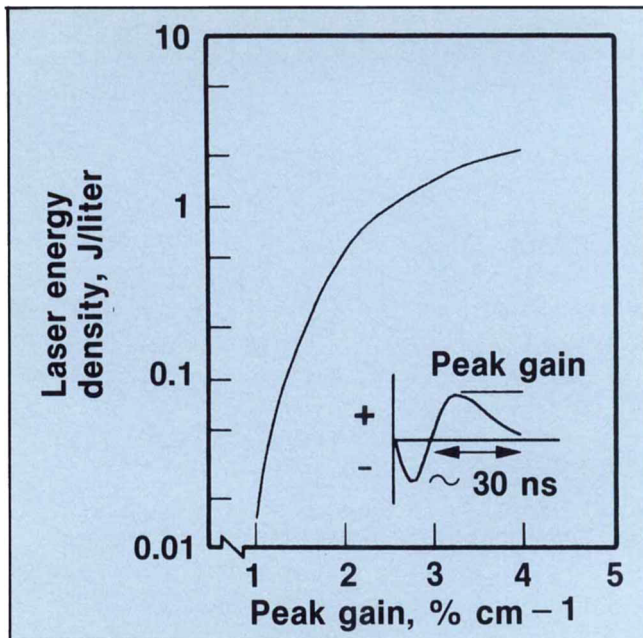


Fig. 9 Variation of peak laser pulse energy density as a function of peak gain for a small gain laser.

is shown clearly in Fig. 9. Experimentally, it has been found that the laser mixture containing F<sub>2</sub> as well as NF<sub>3</sub> yields about two orders of magnitude more output power than does the one with only NF<sub>3</sub>.

Absorption measurements made at several wavelengths, coupled with detailed kinetic modeling of the laser gas mixture, provide the insight required to understand the limits of XeF(C→A) laser performance, and why a combination of halogen donor species improves the situation. Studies show a number of ionized and excited species evolving after the e-beam

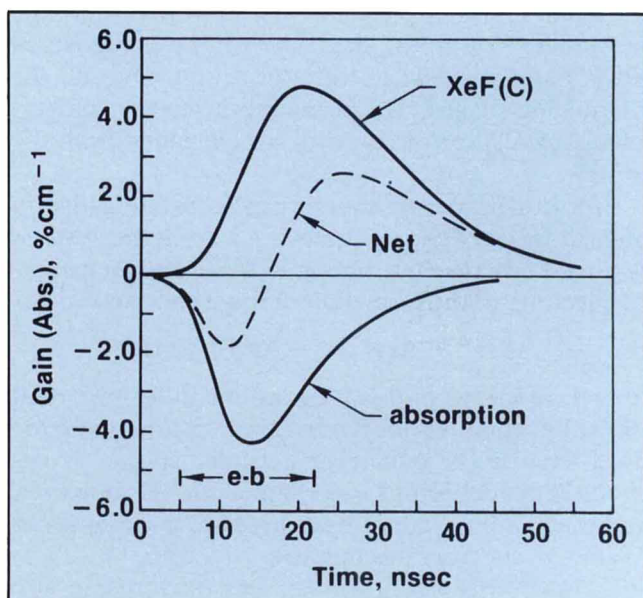


Fig. 11 Calculated gain and absorption for a XeF(C→A) laser using experimentally measured rate constants. The dotted curve shows the difference between the gain and the absorption, and represents the net gain of the system.



excitation pulse, several of which exhibit *broadband* optical absorption in the blue-green region of the spectrum. Figure 10 shows the contribution of several of the most important species as a function of time during and after excitation of an XeF(C $\rightarrow$ A) laser gas mixture. During the period of the e-beam excitation, Ar<sub>3</sub><sup>+</sup>, Ar<sup>\*\*</sup> (the 4p, 3d, and higher excited states of Ar) and the excited argon molecule Ar<sub>2</sub><sup>+</sup>(<sup>3</sup>Σ<sup>+</sup><sub>u</sub>) all contribute substantial optical loss. In the afterglow of the e-beam pulse, it is essentially excited xenon that is responsible for the optical absorption at the XeF(C $\rightarrow$ A) laser wavelength. Note that the absorbing species shown in Fig. 10 are intimately involved in the sequence of events leading to the production of the XeF excimer molecule (Fig. 6).

Knowledge of the temporal evolution of the dominant broadband absorbing species, combined with that of the XeF(C) population, permits calculation of the net optical gain as shown in Fig. 11. Initially, the broadband absorption processes more than offset the positive contribution of the excited XeF(C), and a net optical loss results. As the populations of various absorbers are reduced, however, the gain becomes positive until such time as the XeF(C)\* population is depleted, after which the gain decays to zero. Even during the period of net positive gain, however, the absorption due to Xe<sup>\*\*</sup> (Fig. 10) substantially reduces the value of the peak gain from what it would be in the absence of any absorbers.

As mentioned above, a combination of two fluorine donors, NF<sub>3</sub> and F<sub>2</sub>, considerably improves the laser performance because of two separate actions. In the first place, the dissociative attachment, which leads to

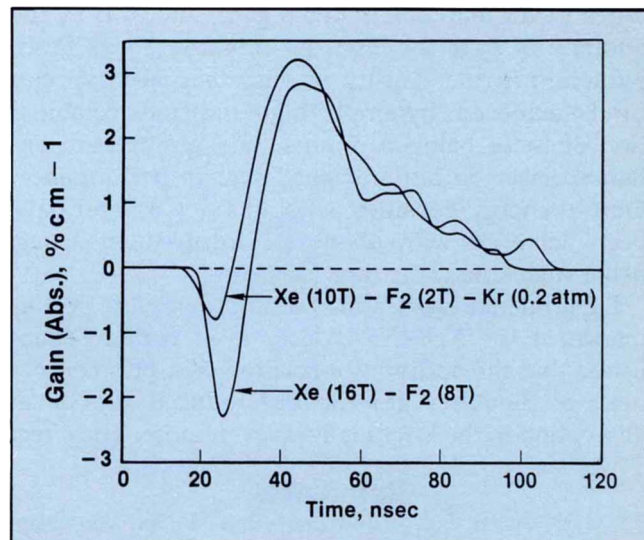


Fig. 13 Temporal evolution of the net gain measured at 488 nm for two different laser gas mixtures. The first is for an optimized Ar, Xe NF<sub>3</sub>, F<sub>2</sub> mixture, while the second is for a mixture composed of 6.5 atm Ar, 10 Torr Xe, 8 Torr NF<sub>3</sub>, 2 Torr F<sub>2</sub>, and 0.2 Atm Kr.

the formation of an F<sup>-</sup> ion by reaction with secondary electrons, is a strong function of electron energy for both donor species. In the case of NF<sub>3</sub> and F<sub>2</sub>, these rates are complementary in the sense that F<sub>2</sub> has a higher dissociative attachment rate at low electron energies, while NF<sub>3</sub> reacts better with higher energy electrons (see Fig. 12). Thus, by using a combination of the two halogen donors, strong formation of the F<sup>-</sup> ion will occur for a longer time as the temperature (energy) of the secondary electron gas cools down. F<sub>2</sub> is also a very good quencher of Xe<sup>\*\*</sup>. In fact, it is

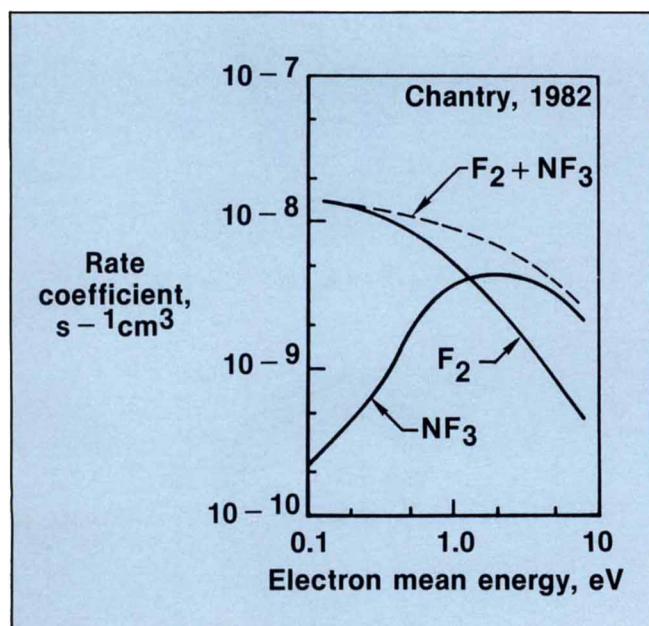


Fig. 12 Dissociative attachment rate for electrons for NF<sub>3</sub> and F<sub>2</sub> as a function of electron energy (adapted from P. Chantry in *Applied Atomic Collision Physics*: vol. 3. *Gas Lasers*, edited by H. S. W. Massey, E. W. McDaniel and B. Bederson, Academic, New York, 1982).

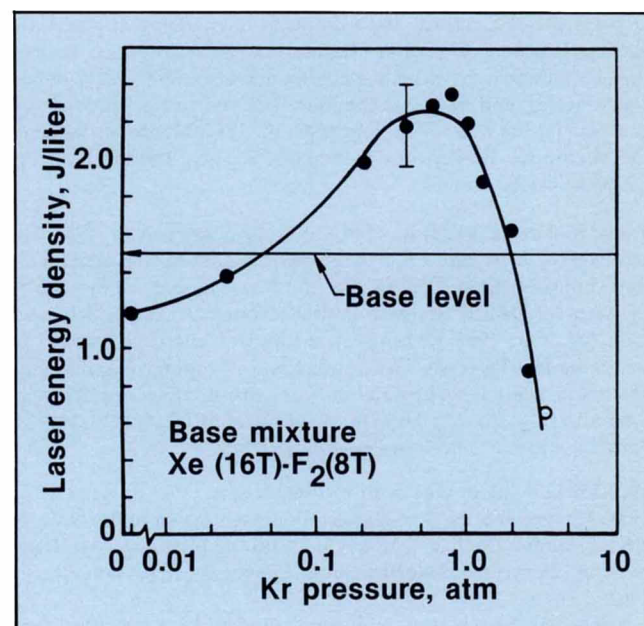


Fig. 14 XeF(C $\rightarrow$ A) laser output energy density as a function of krypton partial pressure for an optimized gas mixture. The base level is the maximum output energy density that could be achieved without the addition of krypton to the mixture.

much better than  $\text{NF}_3$  in this regard, and thus by the addition of  $\text{F}_2$  to the laser gas mixture, a significant reduction in the density of absorbing  $\text{Xe}^{**}$  species can be achieved. By employing a judicious combination of these halogen donors, one can achieve enhanced laser output and good system performance. Output energy densities of about  $1.5 \pm .3$  J/liter have been achieved with about 1% intrinsic efficiency, using such kinetic mixture "tailoring."

Experimental work aimed at improving the performance of the  $\text{XeF}(\text{C} \rightarrow \text{A})$  laser even further established that the addition of krypton as a fifth component of the laser gas mixture reduced the initial absorption in the laser medium even more. The effect

of krypton is shown in Fig. 13. Not only is the initial absorption reduced, but the peak gain is slightly higher and lasts for a somewhat longer time, resulting in a substantial increase in laser power (Fig. 14). The main reason for this significant change is that krypton appears to provide strong channels for the removal of the excited argon species, which are causing the absorption during the early part of the e-beam excitation. These findings provide vivid evidence of the importance of broadband absorbing processes in excimer lasers. Indeed, control of broadband transient absorption, which is a fundamental problem, is the single, most important factor in the operation of electrically excited excimer lasers.

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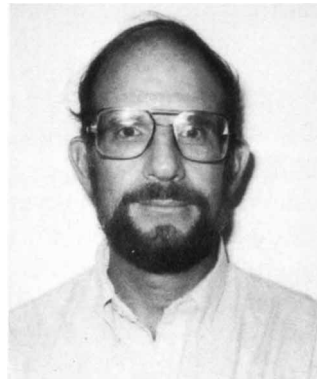
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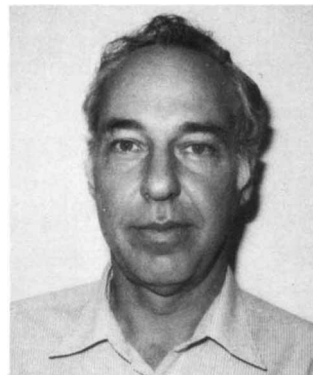
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William L. Wilson, Jr.



Frank K. Tittel



William L. Nighan