Appl. Phys. B 35, 227–231 (1984)

Applied Physics B Photophysics and Laser Chemistry

A New Blue-Green XeF($C \rightarrow A$) Excimer Laser Amplifier Concept

Y. Nachshon and F. K. Tittel

Department of Electrical Engineering and Rice Quantum Institute
Rice University, Houston, TX 77251, USA

Received 29 May 1984/Accepted 23 July 1984

Abstract. The feasibility of using the electrically excited XeF($C \rightarrow A$) excimer medium as an efficient wideband amplifier in the blue-green region of the spectrum has been investigated. Calculations show that for an intense blue-green optical flux input the amplification characteristics of XeF($C \rightarrow A$) improve, as a result of both bleaching of the pump induced absorbers and by additional production of XeF(C) level population. The removal of one of the major absorbing species, Xe** in the XeF ($C \rightarrow A$) laser mixture by means of a pulsed ruby laser probe beam has been demonstrated.

PACS: 42.55, 42.60

The XeF($C \rightarrow A$) rare gas halide excimer transition has been shown to be an efficient broadband laser in the blue-green region of the spectrum [1–3]. A laser output energy density of about 3 J/l with an intrinsic electrical-optical efficiency of $\sim 1.5\%$ has been achieved using selective tailoring of the kinetic processes in an electron beam excited high pressure argon-buffered mixture containing both NF_3 and F_2 as halogen donors. In these mixtures the concentration of electrons which quench the XeF(C) level population and the density of absorbing species in the blue-green are significantly reduced. However, the efficiency is still limited mainly by pump-induced absorbing species of Ar and Xe [2]. In this paper we suggest a further improvement in the efficiency of the XeF($C \rightarrow A$) laser that can be achieved by bleaching the absorbers with intense blue-green radiation [4].

Of the rare gas halide molecules, XeF is unique in that the C level lies 0.1 eV below the B level. Therefore, in thermal equilibrium, at 300 K, more than 97% of combined population of B and C states resides in the C-state; and the branching to the XeF(C) level is very efficient [2]. However, the gain for $C \rightarrow A$ transition is low ($< 3\%$/cm) since the cross-section for stimulated emission is low ($\sim 10^{-17}$ cm^2) and the linewidth is ~ 60 nm (FWHM). Hence, the build-up time of the optical flux of such a laser is slow, thereby limiting its efficiency. On the other hand, the superradiance effects are limited because the gain for the XeF($C \rightarrow A$) transition is small. Moreover, since the number density of the B level is small, the superradiance for the $B \rightarrow X$ transition is small, too. Therefore, it is possible to consider the XeF($C \rightarrow A$) excimer as a potentially efficient wideband blue-green amplifier.

Basic Concept

Two types of pump induced transient absorbers may be considered, as shown in Fig. 1. One type is the narrowband absorption as indicated by the transition between $N_x^* + h\nu \rightarrow N_x^{**}$, where N_x^* is the number density of absorbers. This absorption leads to a structure superimposed on the fluorescence and laser spectra. For a homogeneous linewidth, the discrete absorption will not affect the output power significantly since the laser can operate between absorbing frequencies. Broadband absorption, on the other hand, depicted by $N_x^{**} + h\nu \rightarrow N_x^+$ in Fig. 1, reduces the optical gain over the entire spectral range of the laser. This process can significantly reduce the output power of the laser. However, an intense photon flux can bleach the narrowband absorption and, in fact, remove the structure from the laser output spectrum. Since reduction or removal of the broadband absorption should make the most noticeable improvement in the laser output, we shall consider only this type of absorption [4].

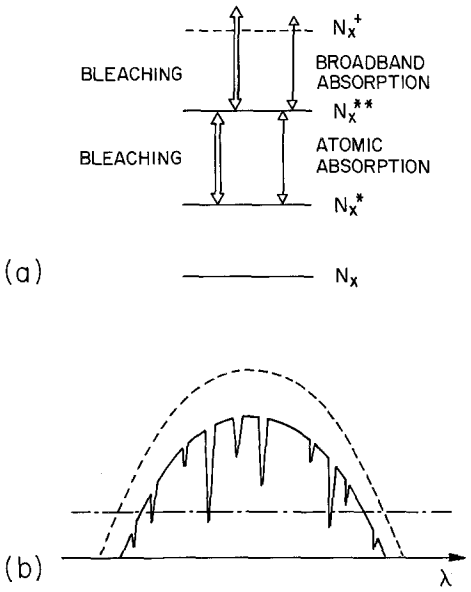

Fig. 1. (a) Schematic diagram of atomic and broadband absorption. (b) Stimulated emission cross-sections due to the XeF($C\rightarrow A$) transition (-----), broadband absorption (-·-), and the combine contribution including atomic absorption (——)

Let N_c be the number density of XeF(C)-level population, while σ_c and σ_x are the stimulated emission cross-section in the blue-green spectrum region for the XeF(C) level and an equivalent absorber, respectively. The net gain – g, for a homogeneously broadened line is

$$g(\nu)=\sigma_c(\nu)N_c-\sigma_x(\nu)N_x. \tag{1}$$

We assume that the upper absorption level is unbound, which is true for the major absorber $Xe^{**}+h\nu \rightarrow Xe^+ + e$.

Let us now examine the amplification process for this excimer system under intense blue-green illumination. A short (<2 ns) optical input pulse $I(\nu)$ is assumed. During this period there is very little change in the production or quenching of the XeF(C) level or density of the absorbers, as a result of the kinetic processes induced by the e-beam pulse. The only effect will result from the interaction of the blue-green radiation with these species. Therefore, the rate equations for N_c and N_x can be written as

$$\frac{dN_c}{dt}=-\left[\frac{\sigma_c(\nu)}{h\nu}I(\nu)\right]N_c, \tag{2}$$

$$\frac{dN_x}{dt}=-\left[\frac{\sigma_x(\nu)}{h\nu}I(\nu)\right]N_x \tag{3}$$

ν and $I(\nu)$ are the same for both equations. Therefore, if $\sigma_x>\sigma_c$, the rate of removal of the absorbers is faster than the decay rate of XeF(C) due to radiation. Hence the rate at which gain decreases due to saturation by the optical flux is smaller than the rate without absorbers. This increases the effective saturated intensity. Furthermore,

$$\frac{dg}{dt}=\frac{I}{h\nu}(\sigma_x^2N_x-\sigma_c^2N_c). \tag{4}$$

For $\sigma_x^2N_x>\sigma_c^2N_c$, the gain increases as the intensity increases. Assuming a short length amplifier, such that the photon flux intensity is nearly constant along the optical axis, one can define the total input energy flux $\Delta E=I\cdot\Delta t$. From equations 2 to 4 one abtains for $dg/dE>0$:

$$\frac{\sigma_x^2N_x(t_0)}{\sigma_c^2N_c(t_0)}>\exp\left[\frac{\Delta E}{h\nu}(\sigma_x-\sigma_c)\right], \tag{5}$$

where t_0 is the time at which the input or bleaching pulse first enters the amplifier.

In the case of an e-beam pumped XeF($C\rightarrow A$) laser, Xe** is the principal absorber [1, 2] with $\sigma_c=10^{-17}\,\mathrm{cm}^{-2}$ and $\sigma_x=2\times10^{-17}\,\mathrm{cm}^{-2}$. Therefore, for a sufficiently small E, dg/dE is positive when $N_x(t_0)/N_c(t_0)>1/4$.

Calculations of the net gain g/g_0 (where $g\equiv g(t_0+\Delta t)$ and $g_0\equiv\sigma_0N_c(t_0)$) as a function of the photon energy flux E, for several ratios of $N_x(t_0)/N_c(t_0)$ for the above conditions are shown in Fig. 2a. For example, for $N_x/N_c=1/2$, $g(E=0)=0$. However, as E increases, g increases, since the absorber number density decreases faster than the XeF(C) level population. For a large E, the gain decreases due to saturation of the XeF(C) level population. As the ratio σ_x/σ_c increases, dg/dE increases for the same N_x/N_c.

Furthermore, let us define the relative energy gain R_E as the ratio of the additional energy from an amplifier and the maximum available energy for the same amplifier without absorbers (i.e., $N_x=0$; $E\rightarrow\infty$). The maximum available energy for a given amplifier is defined by $A\cdot1\cdot N_c\cdot h\nu$, where A is the beam cross-section and 1 is the amplifier length. It is clear that R_E is always smaller than unity. For the case when $\sigma_x>\sigma_c$ and $N_x(t_0)<N_c(t_0)$

$$R_E<1-\frac{N_x}{N_c}. \tag{6}$$

However, a net energy gain can be achieved for an amplifier even if $g(E=0)$ is less than zero providing that $N_x(t_0)<N_c(t_0)$. The total relative energy gain R_E for a one meter amplifier assuming an effective gain $g_0=4\%/\mathrm{cm}$, is given in Fig. 3a for different ratios of $N_x(t_0)/N_0(t_0)$ for the above conditions.

Photoionization of Xe** not only removes one of the principle absorbers in the laser medium, but also

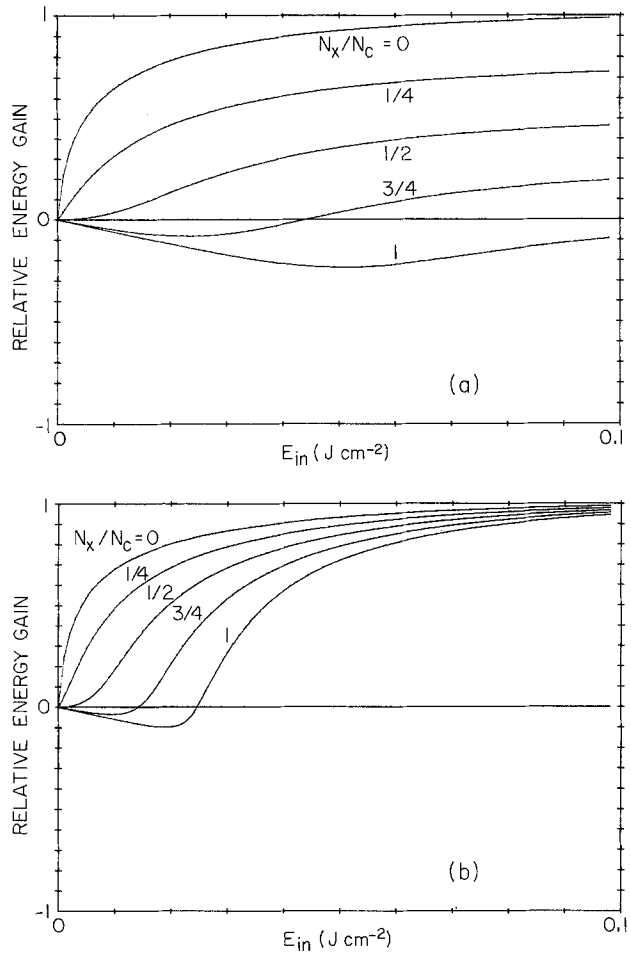

Fig. 2. (a) Net gain as a function of the input photon energy for several values of $N_x(t_0)/N_c(t_0)$, (b) Net gain for the above conditions taking into consideration the fast reaction between Xe^+ and F^-

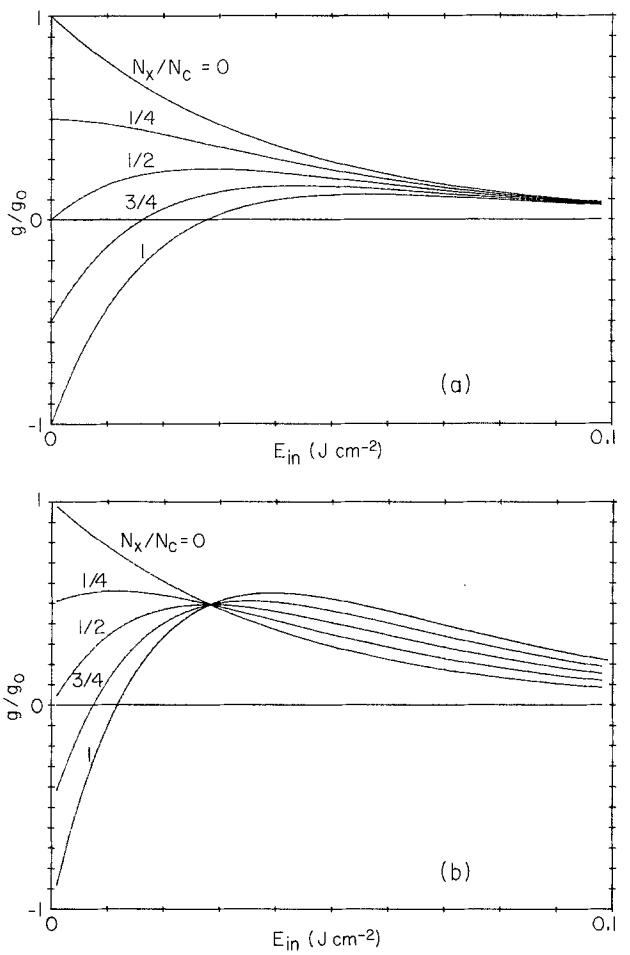

Fig. 3. (a) Net relative energy gain for a 1-m amplifier as a function of the input photon energy for four values of $N_x(t_0)/N_c(t_0)$. (b) Net relative energy gain for the above conditions taking into consideration the fast reaction between Xe^+ and F^-

produces a faster build-up of the XeF(C) level population since the reaction

$$Xe^+ + F^- + Ar \rightarrow XeF^* + Ar \tag{7}$$

is much faster than the reaction

$$Xe^{**} + F_2 \rightarrow XeF^* + F. \tag{8}$$

The characteristic time for reaction (7) is ~ 1 ns for the conditions stated in [1]. One can assume an optical pulse which is longer than 1 ns but still shorter than the other production and quenching rates of the XeF(C) level population. For these conditions any photon absorbed by Xe** will end up as a XeF(C) excimer. The gain rises faster due to reaction (7). Calculation of the net gain (g/g_0) as a function of the energy flux E, taking this effect into account is given in Fig. 2b and that of the modified total relative energy gain, R_E in Fig. 3b. In this case the net relative energy gain is limited to $R_E < 1$, since any photon that is absorbed produces a XeF(C) level excimer that can produce another photon. For a longer pulse (~ 10 ns) one can no longer neglect the effect of the production and quenching of the XeF(C) level population from the e-beam kinetics.

In addition to the Xe** electron beam-excited Ar and Xe mixtures contain yet other absorbers in the blue-green region [2], such as Ar_2^*, Ar**, and Ar_3^+

$$Ar_2^* + h\nu \rightarrow Ar^{**} + Ar, \tag{9}$$

$$Ar^{**} + h\nu \rightarrow Ar^+ + e, \tag{10}$$

$$Ar_3^+ + h\nu \rightarrow Ar^+ + 2Ar. \tag{11}$$

Intense blue-green light will not remove the Ar species absorbers as efficiently as it removes Xe**, since both Ar_2^* and Ar** absorb the radiation and Ar^+ forms Ar_3^+ via Ar_2^+ by means of a three-body collisions with Ar.

Thus the net gain lies somewhere between the case represented in Fig. 2a and b. The effective gain characteristics will depend upon the number densities of the different absorbers and their absorption cross sections. Similarly, the net relative energy gain for a 1-m amplifier falls between the cases represented in Fig. 3a and b.

The Ar-related absorbers species decay faster than Xe** [2]. Therefore, the effect of bleaching the absorbers is more noticeable in the tail of the XeF($C \rightarrow A$) fluorescence. Moreover, as reported in [2], the addition of Kr quenches Ar-related absorbing species. A five-component rare gas halide mixture, consisting of Ar, Kr, Xe, NF_3, and F_2 may lend itself to an effective e-beam excited wideband amplifier in the blue-green region [3].

The stimulated emission cross-section for the XeF($C \rightarrow A$) transition and the ionized Xe($6p$) and Xe($5d$) levels, as a function of wavelength, are depicted in Fig. 4 [5, 6]. At a frequency ν, away from the peak of the XeF($C \rightarrow A$) transition, the ratios between $\sigma_x(\nu)$ and $\sigma_c(\nu)$ increases. Therefore, any photon at this frequency will bleach the absorbers at nearly the same rate as at the peak frequency but will increase the saturation of the XeF($C \rightarrow A$) transition. Good synchronization of two intense illumination pulses, one off the XeF($C \rightarrow A$) transition peak and a second delayed pulse centered at the peak, should produce improved amplifier characteristics. This scheme can be used in a laser where the end reflector designed for the XeF($C \rightarrow A$) transition wavelength would be transmitting outside the laser wavelength region.

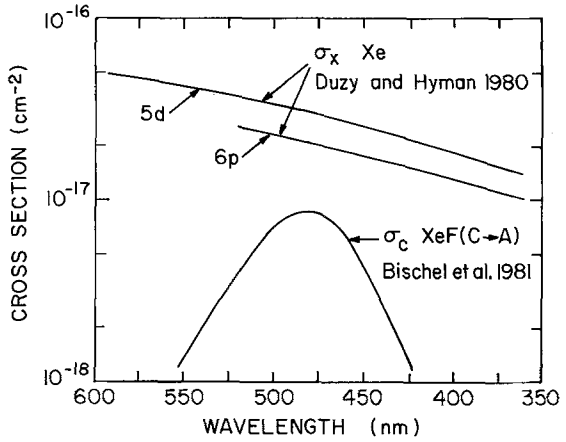

Fig. 4. Stimulated emission cross-section for the XeF($C \rightarrow A$) transition [4] and to the ionized Xe($6p$) and Xe($5d$) levels [3]

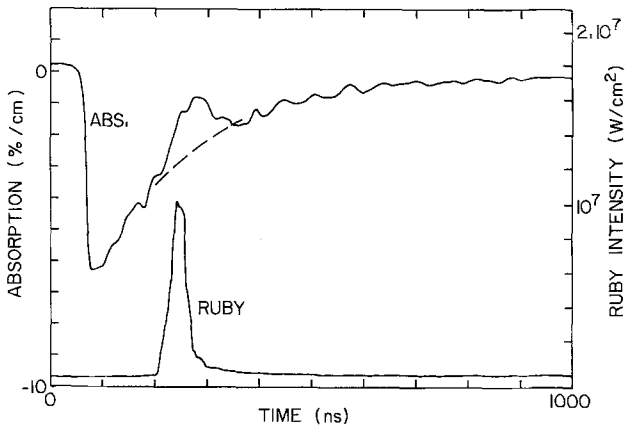

Fig. 5. Absorption measurements at 488 nm for a mixture of 16 Torr Xe and 6 atm Ar. The perturbation in the absorption during the decay time is due to the photoionization of Xe** by the ruby laser

Feasibility Demonstration

To demonstrate the effect of bleaching on the Xe** levels, the optical absorption at 488 nm was measured using an Ar ion laser probe for a mixture of 16 Torr Xe and 6 atm Ar [1, 2]. In this mixture the absorption after the e-beam pumping pulse is due to Xe** alone. The decay rate is ~300 ns. During that time the upper levels of Xe** are populated due to the pumping by electrons. This has also been demonstrated in the infrared by the atomic Xe ($5d$–$6p$) laser operating at 1.733 μm transition [7, 8]. During the decay time, a Q-switched ruby laser beam at 694.3 nm was directed into the reaction cell colinear with the Ar ion probe laser. The ruby laser pulse width was 40 ns with an intensity of ~10 MW/cm^2. The effect upon absorption by the ruby laser pulse is shown in Fig. 5. The absorption decreases during the ruby pulse and rapidly returns to its original value just after input laser pulse.

The ruby laser can ionize only the upper level of Xe($5d$), as shown in Fig. 6. This level, which is the upper laser

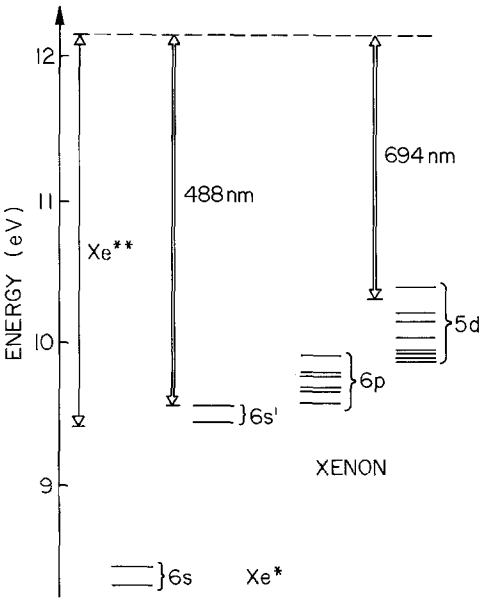

Fig. 6. Energy diagram of Xe*, low levels of Xe** and Xe^+

level of neutral atomic Xe at 1.733 μm, is highly populated [7, 8]. The visible absorption under these conditions is due to photoionization of Xe** [2]. Since the electrons couple together all of the $5d$ levels efficiently, ionization of the Xe($5d$) upper level by the ruby laser will result in a decrease of the population of the other Xe** levels. An absorption measurement using an 488 nm Ar ion laser beam is influenced by the population of several levels, as shown in Fig. 6. Under the experimental conditions, Xe^+ becomes Xe** by a three-body collision with Ar through $ArXe^+$. This process is very fast, as is demonstrated in Fig. 5. The reduction in the absorption during the ruby pulse is a net effect of the 694.3 nm flux intensity, the fast reaction leading to the production of Xe** and the electron coupling among the Xe** levels. The reaction time that produces Xe** from Xe^+ is of the order of 30 ns.

For XeF laser mixtures, the F_2/NF_3 halogen donors will attach electrons at the tail of the XeF($C \rightarrow A$) fluorescence. Therefore, only the lower lying levels of Xe** will be populated, and the 694.3 nm photons would not have enough energy to ionize Xe**. However, the blue-green photons of the laser can ionize the low-lying levels of Xe**. In the presence of F_2/NF_3, the ionized Xe will react with ionized F to produce another XeF(C) level. An excimer laser-pumped dye laser at (say) 425 nm, an excimer laser or the frequency doubled output of a Nd:YAG laser output with an energy output of >100 mj would permit an even more direct experimental evaluation of the wideband XeF($C \rightarrow A$) amplifier concept.

Two other potential broadband amplifiers in the blue-green region are the trimers, Xe_2Cl and Kr_2F. The stimulated emission cross-sections are similar to the XeF($C \rightarrow A$) transition since the radiative lifetime and the linewidth are similar. Moreover, the gain in the blue-green region is $\sim 2\%$/cm [4], which implies efficient pumping to the upper lasing state ($N = g/\sigma$). Therefore, one can use a combination of the XeF($C \rightarrow A$), Xe_2Cl, and Kr_2F to produce a convenient broadband amplifier system for intense visible light.

Acknowledgements. This work was supported in part by the Office of Naval Research, the National Science Foundation and the Welch Foundation. The authors are grateful to W. L. Nighan and W. L. Wilson for helpful discussions.

References

1. W.L. Nighan, Y. Nachshon, F.K. Tittel, W.L. Wilson, Jr.: Appl. Phys. Lett. **42**, 1006 (1983)
2. Y. Nachshon, F.K. Tittel, W.L. Wilson, Jr., W.L. Nighan: J. Appl. Phys. **56**, 36 (1984)
3. Y. Nachshon, N. Nishida, F.K. Tittel, W.L. Wilson, W.L. Nighan, G. Marowsky: Conf. on Laser and Electro-Optics, Technical Digest **154** (1984)
4. F.K. Tittel, G.Marowsky, W.L. Wilson, Jr., M.C. Smayling: IEEE J. QE-**17**, 2268 (1981), also in *Excimer Lasers*, 2nd. ed., ed. by Ch. K. Rhodes, Topics Appl. Phys. **30** (Springer, Berlin, Heidelberg, New York 1984) Chap. 6
5. C. Duzy, H.A. Hyman: Phys. Rev. A **22**, 1878 (1980)
6. W.K. Bischel, D.J. Eckstrom, H.C. Walker, Jr., R.A. Tilton: J. Appl. Phys. **52**, 4429 (1981)
7. S.A. Lawton, J.B. Richards, L.A. Newman, L.Specht, T.A. De Temple: J. Appl. Phys. **50**, 3888 (1979)
8. N.G. Basov, A.Y. Chugunov, V.A. Danilychev, I.V. Kholin, M.N. Ustinovsky: IEEE J. QE-**19**, 126 (1983)