Synthesis of rare gas-halide mixtures resulting in efficient XeF($C \rightarrow A$) laser oscillation

W. L. Nighan
United Technologies Research Center, East Hartford, Connecticut 06108
F. K. Tittel, W. L. Wilson, Jr., N. Nishida, Y. Zhu, and R. Sauerbrey
Electrical Engineering Department and Rice Quantum Institute, Rice University, Houston, Texas 77251

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Significantly improved XeF($C \rightarrow A$) laser performance has been achieved using electron beam excitation of complex, multicomponent gas mixtures specifically tailored so as to reduce medium transient absorption in the blue-green region. Use of Ar and Kr together as the effective rare gas buffer-energy transfer species, along with a combination of NF_3 and F_2 to produce the desired F-donor molecule characteristics, has permitted synthesis of near optimum medium properties for which XeF(C) is produced efficiently while transient absorption is minimized. With this technique we have achieved laser pulse energy density and intrinsic efficiency of 2.2 ± 0.3 J/l and $\sim 1.5\%$, respectively, values that are comparable to those of the $B \rightarrow X$ rare gas-halide lasers.

Short pulse electron beam (e-beam) excitation of the blue-green XeF($C \rightarrow A$) laser transition is characterized by a period of strong transient absorption during the excitation pulse, followed by the development of net gain and subsequent laser oscillation in the afterglow regime.[1] Until recently, the combination of a relatively short duration of the net gain region ($\lesssim 50$ ns) and a relatively low value of the peak gain ($\lesssim 2\%$ cm^{-1}) has limited the efficiency of this tunable laser to unacceptably low levels. However, by selectively tailoring kinetic processes through the use of a unique two-halagon mixture containing *both* NF_3 and F_2 so as to reduce transient absorption, a very significant improvement in XeF($C \rightarrow A$) laser performance has been achieved.[1,2] Indeed, e-beam excitation of such two-halogen Ar-Xe mixtures has yielded laser pulse energy density in excess of 1.0 J/l, corresponding to intrinsic electrical-optical energy conversion efficiency estimated to be in the 0.5%–1.0% range. In this letter we report further improvement in XeF($C \rightarrow A$) laser performance resulting from the addition of Kr to an Ar-Xe-NF_3-F_2 mixture. This multicomponent mixture has permitted synthesis of near optimum medium properties resulting in XeF($C \rightarrow A$) laser pulse energy density and intrinsic efficiency of 2.2 ± 0.3 J/l and $\sim 1.5\%$, respectively. This improvement was obtained without increasing the energy deposited in the gas by the e-beam. Thus, a level of XeF($C \rightarrow A$) laser performance has been achieved which, for the first time, is comparable to that typical of UV $B \rightarrow X$ rare gas-halide laser transitions.

In this investigation laser excitation was provided by an electron beam having an energy of 1 MeV and a pulse duration of 10 ns (full width at half-maximum). The e-beam current density at the center of the optical axis was ~ 250–300 A cm^{-2}, as measured with a Faraday probe. A stable, intracell optical resonator was used consisting of a totally reflecting ($R > 99.6\%$) mirror having a radius of curvature of either 0.5 or 1.0 m, separated by 12.5 cm from a flat output mirror having a reflectivity of 95%, a value found to be optimum for the present conditions. The active region was the ~ 28 cm^3 volume defined by the clear aperture (1.9 cm diameter) and the pumped length (10 cm). Specific details of this experimental arrangement and related diagnostic apparatus are described in Ref. 1.

A cw Ar-ion laser was used[1] to measure the temporal evolution of the gain/absorption at several wavelengths throughout the blue-green region. Figure 1 shows a representative gain-absorption profile at 488 nm for an optimized Ar-Xe-NF_3-F_2 mixture under conditions for which the laser pulse energy density and intrinsic efficiency were typically 1.5 ± 0.3 J/l and $\sim 1.0\%$, respectively. Comprehensive analysis[1] of medium kinetic processes indicates that for these conditions the initial period of strong absorption is primarily the result of photoionization of the $4p$, $3d$, and higher lying states of Ar, and of the Xe $6p$, and $5d$ states, along with photodissociation of $Ar_2(^3\Sigma_u^+)$ and Ar_3^+. These broadband absorption processes more than offset the positive contribution of XeF(C) excimer molecules during and immediately following the period of e-beam excitation. Although the Ar-related absorption processes decay rapidly, photoionization of Xe excited states is very significant even after the gain

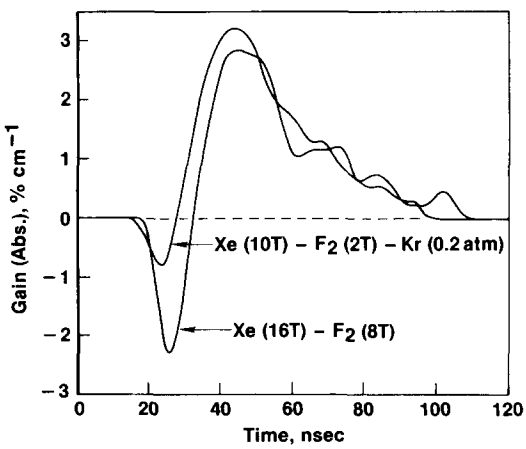

FIG. 1. Temporal evolution of the net gain measured at 488 nm for mixtures comprised of 6.5 atm Ar, 8 Torr NF_3, and partial pressures of Xe and F_2 optimized with and without Kr as indicated. The measured e-beam current density on the optical axis for these conditions was typically 275 ± 25 A cm^{-2}, corresponding to a volumetric energy deposition estimated to be ~ 150 J/l (Ref. 1).

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becomes positive, thereby substantially reducing the peak gain value from that possible due to XeF(C) alone.[1]

During the course of this work it was found that addition of Kr dramatically reduced the initial absorption of the previously optimized Ar-Xe-NF_3-F_2 mixture.[1] Based on our analysis of the change is measured absorption in pure Ar and in Ar-Xe mixtures, it appeared that the most likely explanation for this observation was that Kr reduced the concentrations of both $Ar_2(^3\Sigma_u^+)$ and Ar_3^+ during the e-beam excitation pulse, while providing additional decay channels for the Xe $6p$ and $5d$ states. Both gain and fluorescence data indicated that the additional quenching of XeF(C) by Kr was also significant, however, so that the magnitude and duration of the gain typical of our initial experimentation with Kr were not improved. Thus, in these early studies, no increase in laser pulse energy/efficiency was achieved with mixtures containing Kr.[1]

Because the influence of Kr on the measured blue-green absorption in various mixtures was found to be so pronounced, recent efforts have focused on systematic measurement and evaluation of gain temporal profiles and laser pulse energy, while varying the fractional concentrations of Xe, F_2, and NF_3 in mixtures of Ar and Kr at a *total* pressure[3] of 6.5 atm. Although the Kr pressure was varied from a few Torr to several atm, particular emphasis was placed on Kr pressures in the 0.1–1.0-atm range for which the absorption peak during the e-beam excitation pulse was found to be minimized. Presented in Fig. 1 is the measured gain profile for a *reoptimized* mixture containing 0.2 atm Kr, excited under conditions essentially identical to those of the mixture without Kr, also shown in the figure. The significant reduction in absorption during the excitation pulse with Kr in the mixture is readily apparent, as is the higher value of peak gain and the increase in gain duration to about 80 ns. These features were found to be typical of Kr pressures throughout the entire 0.1–1.0-atm range.

Measurements show that the magnitude of the absorption minimum (Fig. 1) with Kr in the mixture is relatively insensitive to the specific values of Xe or F_2 pressure, but that the peak gain (and its rate of decay) is dependent on the concentrations of these species. Indeed, the *optimum* concentrations of both Xe and F_2 are found to be significantly lower than their values in the absence of Kr.

Although our measurements indicate that the primary role of Kr is reduction of the concentrations of Ar and Xe related species that absorb in the blue-green region,[1] the measured rate of rise of the XeF($C \rightarrow A$) fluorescence and its peak value both are significantly higher with Kr in the mixture, particularly for large F_2 concentrations. Analysis shows that these effects are much more pronounced than would be expected on the basis of faster XeF B/C state mixing[4] due to Kr, suggesting that XeF(B, C) formation is enhanced when Kr is present. One possible explanation for this observation is that Xe displacement reactions involving either KrF or Kr_2F are more effective than their Ar counterparts.[5] However, the enhancement in XeF(C) formation with Kr in the mixture appears to be of less importance than the reduction in transient absorption.

Presented in Fig 2 is the dependence of laser pulse energy density on Kr pressure for the specific Ar-Xe-F_2-NF_3 mixture found to be optimum for Kr pressures in the 0.1–1.0-atm range.[3] The four-component mixture optimized in the absence of Kr consistently results in laser pulse energy density values of 1.5 ± 0.3 J/l for the present conditions, a value not unduly sensitive to either the radius of curvature of the total reflector or of the transmission of the output coupler. Figure 2 shows that as the Kr pressure is increased above ~0.05 atm for the reoptimized mixture, the laser pulse energy increases above the ~1.5 J/l base level of the reference no-Kr mixture. For Kr pressures in the 0.4–0.9-atm range, a broad maximum in laser energy density is achieved, corresponding to ~50% increase over that of the reference mixture, a result that is consistent with the trend exhibited by the measured gain/absorption profiles. The maximum 2.2 ± 0.3 J/l laser energy density typical of these conditions corresponds to an intrinsic energy conversion efficiency estimated[1] to be approximately 1.5%.

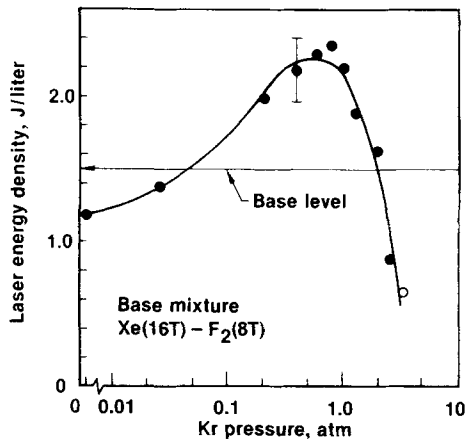

FIG. 2. Laser pulse energy density dependence on Kr pressure as measured using a calibrated vacuum photodiode detector. The mixture was comprised of Ar and Kr at a total pressure of 6.5 atm, 8 Torr NF_3, 10 Torr Xe, and 2 Torr F_2. The indicated base level refers to the mixture optimized in the absence of Kr; the symbol (O) indicates the measured laser energy with no Ar in the mixture.

Measurements were also carried out for Kr pressures in the 3–4-atm range with *no* Ar in the mixture, a condition for which the energy deposited by the e-beam would be very nearly equivalent to Ar-buffered mixtures at a *total* pressure of 6.5 atm.[3] In these tests, with the Xe and F_2 pressures again reoptimized, the maximum laser energy density was in the 0.5–0.7-J/l range, a value much lower than that of either the no-Kr reference mixture or the optimized mixture with Kr present.[6] With the Kr pressure in the 3–4-atm range, addition of varying amounts of either Ne or Ar up to pressures of several atm resulted in laser pulse energy values about the same as those using Kr alone, although the energy deposited in such cases was substantially increased. This is in contrast to the situation for either the Ar-buffered reference mixture or the optimized Ar-Kr mixture, for which it was found that increasing the energy deposition by increasing either the e-beam current density or the Ar pressure resulted in higher laser output. Indeed, for the optimum conditions of Fig. 2, but with the Ar pressure increased from ~6.5 to 8.5 atm, the laser pulse energy was found to increase from 2.2 J/l to ~3.0 J/l. Thus, all of our evidence indicates that the use of Ar and

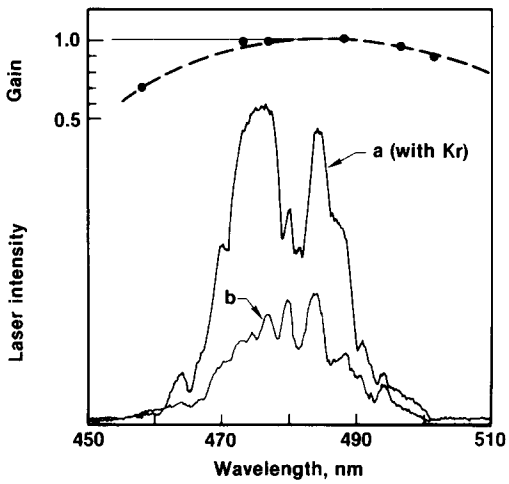

FIG. 3. XeF($C \rightarrow A$) laser spectra for (a) the mixture optimized with Kr present and (b) for the optimum mixture in the absence of Kr. Also shown is the wavelength dependence of peak zero-field gain normalized to its value at 488 nm.

Kr *together* as an effective buffer-energy transfer species results in a medium that, when electrically excited, is characterized by significantly lower concentrations of excited-ionized species that absorbed in the blue-green region than is the case using either Ar or Kr alone.

Presented in Fig. 3 are the time integrated laser spectra for optimum mixtures with and without Kr for conditions similar to those in Fig. 1. Although there are some differences, the two spectra are generally similar except for a somewhat deeper absorption valley centered near 480 nm in the Ar-Kr mixture. Also shown in this figure is the measured wavelength dependence of the peak value of the zero-field gain,[1] normalized to its value at 488 nm. The relatively weak wavelength dependence of the gain suggests that *efficient* tuning[7] of the electrically excited XeF($C \rightarrow A$) laser may be possible throughout a large portion of the 450–510-nm range.

This investigation has shown that a combination of rare gases (Ar + Kr) and fluorine molecules ($NF_3 + F_2$) permits synthesis of near optimum XeF($C \rightarrow A$) laser properties for which XeF(C) can be produced efficiently (5%–10%) while transient absorption is minimized. The resulting optical extraction efficiency of 20%–25% is unique for an electrically excited XeF($C \rightarrow A$) laser. Indeed, the values of laser energy density (2–3 J/l) and intrinsic efficiency (1%–2%) typical of the present electron beam excited XeF($C \rightarrow A$) laser medium are comparable to those of other blue-green lasers such as HgBr($B \rightarrow X$) and wavelength shifted XeCl($B \rightarrow X$) or XeF($B \rightarrow X$). If comparable performance levels can be achieved using discharge excitation, the XeF($C \rightarrow A$) laser may become a competitive, tunable optical source for the blue-green region of the spectrum. Additionally, it is likely that mixture synthesis of the type employed in this investigation will find application as a means to improve the performance of other laser systems.[8]

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[3] With an Ar pressure of 6.5 atm the addition of Kr up to pressures of ~0.3 atm results in an insignificant increase in *e*-beam energy deposition. In order to account for the effect of Kr at higher pressures, the Ar partial pressure was reduced so as to maintain a total pressure of 6.5 atm. This approximation to constant energy deposition becomes relatively poor for Kr pressures $\gtrsim$ 1.0 atm since the *e*-beam stopping power of Kr is about twice that of Ar. However, our experiments show that, for Kr pressures above ~2.0 atm, the maximum attainable laser energy (< 1.0 J/l) for an optimized mixture is relatively insensitive to either the partial pressures of various rare gas buffer species, the total pressure, or the energy deposited. See text.

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