XeF laser characteristics studied at elevated temperatures

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The electron-beam-pumped XeF laser operating on the B-X and C-A transitions shows a strong temperature dependence. Fluorescence, gain, and laser output measurements in $Ar/Xe/NF_3$ and $Ne/Xe/NF_3$ mixtures are reported for temperatures from 300 to 600 K. Elevated temperatures cause stronger mixing between the B and C states as well as among the vibrational levels within the B state. Together with the higher ground-state dissociation rate this leads to an improvement of the B-X lasing for 300k $< T <$ 500 K, whereas the C-A laser emission ceases under these conditions.

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INTRODUCTION

In the last few years electron-beam-pumped excimer lasers have been developed as highly efficient and scalable laser systems.[1] Much of the attention has focused on the xenon fluoride excimer because it exhibits laser action in the ultraviolet and the visible spectrum. The spectroscopic studies of several groups have led to modifications to the potential curves as calculated by Dunning and Hay.[2] According to Tellinghuisen et al.[3,4] the ground state is bound with a well depth of about 120 cm⁻¹ and Kligler et al.[5] found that the C state lies below the B state by about 700 cm⁻¹ as depicted in Fig. 1. This has resulted in obtaining laser emission in the blue-green on the bound-free C→A transition at 486 nm in addition to the stimulated emission on the prominent bound-bound B→X laser transition at 351 nm first observed by Ault et al.[6] and Brau and Ewing.[7] Laser action on the C→A transition was demonstrated by Bischel et al.[8] with photodisso-

ciative excitation of XeF_2, by Ernst and Tittel[9] in e-beam-pumped $Ar/Xe/NF_3$ mixtures and by Fisher et al.[10] and Burnham[11] in discharge-pumped $He/Xe/NF_3$ mixtures.

The B→X laser performance under long e-beam pulse excitation (1 μs) could be improved by using neon as buffer gas instead of argon, as shown by Champagne and Harris.[12] Furthermore, Hsia et al.[13] found that the B→X laser efficiency increases at elevated temperatures by as much as a factor of 2. Explanations of these two results are based on investigations of the B→X transition only. However, laser emission occurs on both the B→X amd C→A transitions and the energy separation between the B and C states is small. The coupling and the excitation mechanism of these two states must be taken into account. In this work we investigated the influence of different buffer gases and temperatures on the excited-state formation, B and C state mixing processes, and lower level removal. Detailed fluorescence, gain, and laser output measurements for both transitions at temperatures between 300 and 600 K are reported. Both argon and neon were used as buffer gases.

EXPERIMENT

A 1-MeV 20-kA electron-beam pulse of 8 ns duration generated by a Physics International Pulserad 110 electron-beam accelerator entered the stainless-steel reaction cell through a 50-μm-thick Ti foil to transversely excite a 6-cm-long gas column. The cell could be heated to about 700 K. The e-beam apparatus and the high-temperature setup are described in more detail in Ref. 14. Light emitted from the excited volume was monitored time resolved with a photodiode, over a 30-nm-wide spectral range for each band selected by a $\frac{1}{4}$-m monochromator placed in front of the diode. Simultaneously, spectra were observed with an optical multichannel analyzer (OMA).

For laser experiments a prealigned resonator was inserted into the cell, which had a high-reflectance 1-m curved end mirror and a 95% reflectance output coupler for the B→X or a 98% one for the C→A transition.

Gain measurements were performed using an argon ion laser as a cw probe laser. After passing the cell the beam was directed through several apertures to a monochromator with a fast photomultiplier placed 6.5 m away in a Faraday cage.
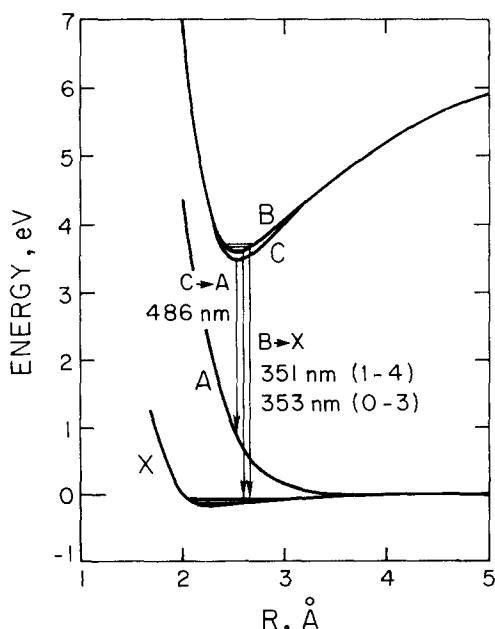

FIG. 1. Partial energy level diagram of XeF showing B→X and C→A laser transitions.

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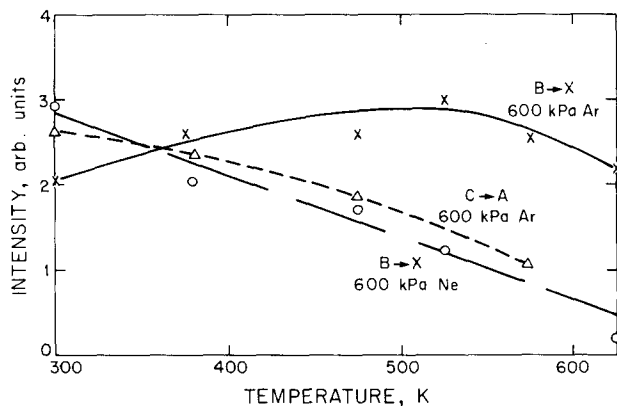
FIG. 2. Fluorescence peak intensity versus temperature for mixtures with 16 Torr Xe and 8 Torr NF_3.

FIG. 4. Small-signal gain of the $B{\rightarrow}X$ and $C{\rightarrow}A$ transitions versus temperature for mixtures with 16 Torr Xe and 8 Torr NF_3.

A brick wall and lead shielding helped to avoid x-ray noise generated by the e-beam accelerator. Influence of fluorescence light from the cell could not be detected and attenuation of the laser ligh with filters prevented the photomultiplier from saturation.

Gain on the $B{\rightarrow}X$ and $C{\rightarrow}A$ transitions was measured using the argon ion laser lines at 351 and 488 nm, respectively. The experiments were performed with mixtures of 16 Torr Xe, 8 Torr NF_3, and 400 and 600 kPa Ar or Ne.

RESULTS

All our measurements showed a strong temperature dependence which means that there is an adjustment of the particle velocity distribution to the environmental temperature on a nanosecond time scale even though the equilibrium in the gas mixture has been strongly disturbed by the short electron-beam excitation pulse. This must be due to the high particle density in the plasma. In order to distinguish the temperature effects on the upper level population of the transitions from those on the population inversion, i.e., including the lower level removal, the results for fluorescence and gain/laser measurements are discussed separately.

Fluorescence

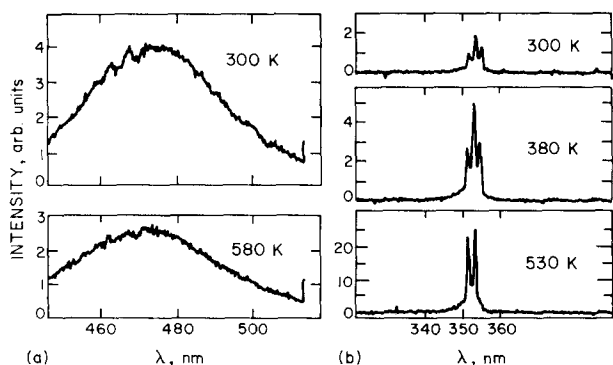The temperature dependence of the measured fluorescence peak intensity is depicted in Fig. 2. This plot reflects

the B and C state population densities. For 600 kPa argon buffer gas the $B{\rightarrow}X$ fluorescence increases with temperature up to 530 K and then starts to decrease. At the same time the $C{\rightarrow}A$ fluorescence decreases over the whole temperature range. Between 300 and 530 K the $B{\rightarrow}X/C{\rightarrow}A$ fluorescence ratio changes by a factor of 3.5, which compares to a change of the B/C population ratio by a factor of 4 in the case of a Boltzmann distribution.

With 600 kPa neon buffer gas the $B{\rightarrow}X$ fluorescence intensity at room temperature was as high as that for argon buffer gas at the optimum temperature of 520 K. For Ne the fluorescence intensity decreased for elevated temperatures.

FIG. 3. Fluorescence spectra of (a) the $C{\rightarrow}A$ transition at 300 and 580 K and (b) the $B{\rightarrow}X$ transition at 300, 380, and 530 K for a mixture of 16 Torr Xe, 8 Torr NF_3, and 600 kPa Ar.
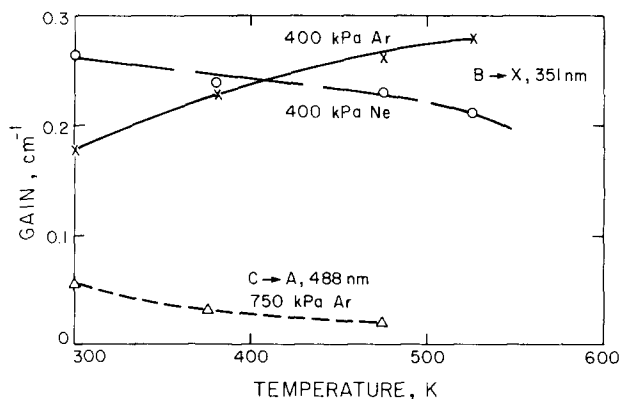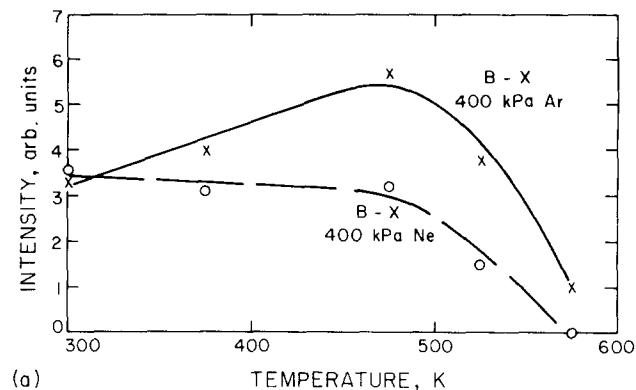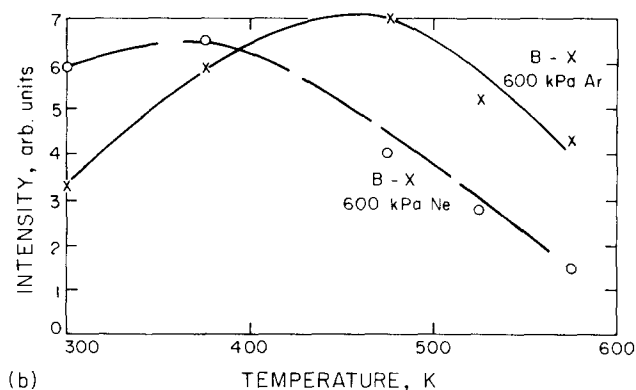
FIG. 5. $B{\rightarrow}X$ laser peak intensity versus temperature for mixtures of 16 Torr Xe, and 8 Torr Nf_3, (a) for 400 kPa and (b) 600 kPa buffer gas pressure of Ar and Ne.
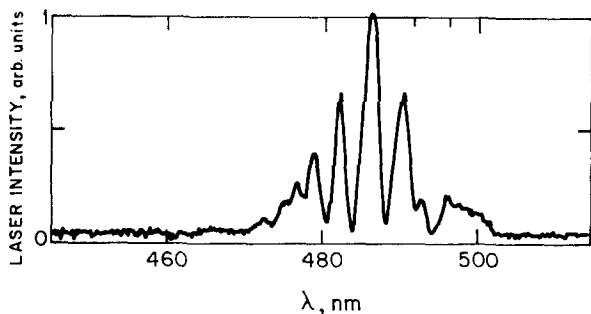
FIG. 6. Laser output spectrum of the $C \rightarrow A$ transition for a mixture of 16 Torr Xe, 8 Torr NF$_3$, and 600 kPa Ar at room temperature.

The observed $C \rightarrow A$ flluorescence was extremely small for Ne. Therefore, all $C \rightarrow A$ measurements were only performed with Ar buffer gas.

Spectra obtained with the OMA proved to be useful for observing temperature-dependent changes of the spectral structure within the bands. As seen in Fig. 3 (a) emission from the $C \rightarrow A$ transition appears as a continuum-like band centered around 475 nm with a FWHM of about 40 nm at room temperature. At 580 K the intensity is obviously lower and the emission band is slightly broader with less pronounced absorption dips. However, the spectral structure of the $B \rightarrow X$ fluorescence shown in Fig. 3 (b) for 600 kPa Ar buffer gas changes considerably with temperature. For a wavelength calibration in Fig. 3 (b) the argon ion laser lines at 351.1 and 363.8 nm were used. At room temperature three bands appear at 351, 353, and 355 nm, due to the 1-4 (weakly 0-2), 0-3 (weakly 1-6), and 0-4 vibrational bands.[3] A band at 349 nm, due to the 2-5 transition with a Franck-Condon factor four times as high as the 0-4 transition,[4] does not show up. This indicates that in our experiment the vibrational state $v' = 2$ was only weakly populated under all conditions. With increasing temperature the 351-nm band grows more intensely than the 353-nm band to reach a ratio of unity at 580 K. The 355-nm band disappears for $T > 480$ K, which shows that 600 kPa Ar buffer provides strong vibrational relaxation at room temperature, highly populating the $v' = 0$ level. With higher temperatures an increasing vibrational mixing leads to a stronger $v' = 1$ state population. A slightly increasing separation between the 351- and 353-nm bands for $T > 530$ K indicates that at elevated temperatures the 353-nm emission is determined primarily by the 1-6 transition (data and assignment used for this conclusion are taken from Refs. 3 and 4).

With Ne buffer gas instead of Ar at the same pressure, a similar temperature-dependent change of the $B \rightarrow X$ spectrum was observed. The main difference was that with Ne the 351-nm band was stronger than with Ar for all temperatures, which shows that the vibrational relaxation is less strong with Ne. As with Ar, the 349-nm band was not observed and the 355-nm emission vanished for $T > 480$ K.

Gain and laser emission

Temperature-dependent small-signal gain measured with an argon ion probe laser at 351 and 488 nm is depicted in Fig. 4. At a moderate buffer gas pressure of 400 kPa gain on the $B \rightarrow X$ transition increased by 50%, when the gas mixture with Ar buffer was heated from 300 to 530 K. With Ne buffer a slight decrease of the gain was observed. To get measurable gain on the $C \rightarrow A$ transition at elevated temperatures a fairly high buffer gas pressure of 750 kPa Ar had to be used. At this pressure the gain at 480 K had only about 30% of the room-temperature value and above 480 K became too small to be observed. The $B \rightarrow X$ gain above 530 K could not be measured because of the constantly changing direction of the probe laser beam due to air turbulence at the cell windows.

The $B \rightarrow X$ laser peak intensity at 400 kPa buffer gas pressure behaved very much like the measured gain with temperature, as is depicted in Fig. 5(a). For Ar buffer there is a 60% increase of the laser output at 480 K as compared to 30 K. At higher buffer gas pressures of 600 kPa the temperature has an even stronger influence shown in Fig. 5(b). With Ar buffer gas the $B \rightarrow X$ laser emission at 480 K is about 120% higher than at room temperature. With 600 kPa Ne buffer gas the laser emission slightly improves with temperature between 300 and 380 K in contrast to the fluorescence (cf. Fig. 2) and starts to fall off only for $T > 380$ K. With both buffer gases the laser output decreases rapidly beyond 500 K, independent of the pressure. Above 600 K the mirror coatings started to deteriorate. Laser emission on the $C \rightarrow A$ transition was not observed at elevated temperatures since the low gain could not overcome the cavity losses.

A laser spectrum of the $C \rightarrow A$ transition at room temperature is shown in Fig. 6. Absorption bands, probably due to transient species, cause the line structure of the laser output, which was centered at 486 nm. The envelope has a FWHM of 12 nm. Similar absorption features in the weak near-threshold emission with Ne or Kr buffer gases indicate that it is the xenon or fluorine that are involved in the absorption rather than some buffer gas species.

$B \rightarrow X$ laser spectra show a strong change with temperature. As shown in Fig. 7(a) laser action with 600 kPa Ar buffer gas at 300 K occurs mainly on the 0-3 transition at 353 nm. With increasing temperature the 351-nm output is enhanced and finally becomes dominant. A similar change appears with 600 kPa Ne buffer, depicted in Fig. 7(b), except that the 351-nm laser output is always stronger than with Ar buffer gas. A similar temperature-dependent change of the laser line intensity ratio was observed in Ref. 13. The stronger increase of the $B \rightarrow X$ stimulated emission with temperature than for the fluorescence indicates that the $B \rightarrow X$ laser performance is improved both by upper level mixing and the higher dissociation rate of the ground state at elevated temperatures.

DISCUSSION

The experimental results reported in this work lead to several conclusions which can be compared with other results reported in the literature. The fact that the $(B \rightarrow X / C \rightarrow A)$ fluorescence ratio increases for a mixture using a light buffer gas as collision partner (i.e., Ne instead of Ar) and also for elevated temperatures strongly supports the result of Kligler et al.[5] that the C state lies below the B state.

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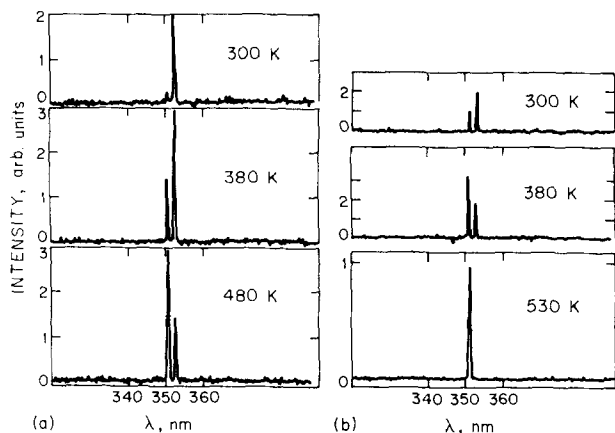
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FIG. 7. Laser output spectra of the $B{\rightarrow}X$ transition at 300, 380, 480, and 530 K for (a) a mixture of 16 Torr Xe, 8 Torr NF$_3$, and 600 kPa Ar and (b) a mixture of 16 Torr Xe, 8 Torr NF$_3$, and 600 kPa Ne.

The temperature behavior of the two $B{\rightarrow}X$ laser lines agrees well with the transition assignment by Tellinghnisen et al.[3]

Most reaction kinetic studies in the past did not include the C state. A recent publication of Finn et al.[15] considered the role of the C state and reported that about 80% of the excitation energy goes directly into the B state. Our experimental results agree with this conclusion and the enhanced C population at high pressure seems to be due to $B{\rightarrow}C$ quenching,[16] especially if a heavier collision partner like Ar is involved. However, our measurements indicate that energy transferred into the C state need not be lost. The back reaction $C{\rightarrow}B$ can be strongly enhanced by heating. Therefore, the $B{\rightarrow}X$ laser efficiency is improved at elevated temperatures, not only by the higher dissociation rate of the ground state as predicted by Finn et al. in Ref. 17 and found by Hsia et al.,[13] but also by the stronger B-C mixing which increases the laser emission by populating the B state more effectively. At room temperature Ne is the better buffer gas for the $B{\rightarrow}X$ laser. An important reason for this is that neon is a less efficient $B{\rightarrow}C$ quencher and exhibits lower absorption.[18]

Of course, when comparing these results with other investigations the intensity and duration of the excitation pulse should be considered. As, for example, Chang and Champagne[19] pointed out, the absorption in the excited mixtures differs significantly whether the e-beam pulse is maintained during the whole reaction time or decays very fast as in our case. Fluorescence and laser emission occur about 10 ns after the 8-ns-long excitation pulse by which time the broadband absorption should have decreased according to Refs. 19 and 20.

The fluorescence measurements with Ne buffer gas (depicted in Fig. 2) show that for elevated temperatures for the

B state population is not increased by an enhanced $C{\rightarrow}B$ back reaction since the C state is only weakly populated. In fact, the $B{\rightarrow}X$ fluorescence decreases for increased temperatures which may be due to strong vibrational mixing within the B state and a population distribution over many levels with $v' > 1$. In this case laser action is only enhanced by stronger lower level removal.

For the e-beam-pumped XeF $B{\rightarrow}X$ laser it has been shown that quenching losses due to high buffer gas pressures which are needed for the efficient energy transfer can be removed by heating the laser gas mixture. This will also increase the ground-state dissociation rate. On the other hand, $C{\rightarrow}A$ laser action should be enhanced by cooling of the high-pressure mixture.

ACKNOWLEDGMENTS

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