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Effect of Nitrogen on XeF($C \rightarrow A$) and Xe_2Cl Laser Performance

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Abstract—**Experiments demonstrating the effect of nitrogen on electron-beam pumped XeF($C \rightarrow A$) and Xe_2Cl laser performance are reported. The laser power of the XeF($C \rightarrow A$) laser decreased with increasing nitrogen pressure, whereas the Xe_2Cl laser power increased by a factor of three at an optimum nitrogen pressure of 200 torr. Atomic absorptions in both laser spectra are decreased by the addition of nitrogen. The kinetic mechanisms leading to the observed behavior are discussed.**

I. Introduction

RECENTLY, broadband rare gas halide lasers utilizing either the $C \rightarrow A$ transition of diatomic XeF* [1]-[4] centered around 480 nm or trimers, such as Xe_2Cl at about 520 nm [5], have been developed. For the XeF($C \rightarrow A$) laser, high power output and wavelength tunability have been demonstrated [6]-[8]. In the case of Xe_2Cl, gain measurements [9] and laser action [5] were reported.

Both excimer lasers suffer from absorptions by metastable xenon species [7], [10] which reduce their output power and impair their tuning capability. Recently, Eckstrom *et al.* [6], [7] demonstrated that these absorptions can be eliminated in the XeF($C \rightarrow A$) laser spectrum when XeF_2 is excited photolytically to form XeF* and nitrogen is used instead of argon as the buffer gas. In this work the effects of adding nitrogen to $Ar/Xe/NF_3$ and $Ar/Xe/CCl_4$ mixtures on the electron beam-pumped XeF($C \rightarrow A$) and Xe_2Cl laser performance are reported. The fluorescence and laser output characteristics for both laser mixtures are described.

Manuscript received April 19, 1982. This work was supported by the Office of Naval Research, the National Science Foundation, the Robert A. Welch Foundation, and the Deutsche Forschungsgemeinschaft.

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II. Experimental Setup

Details of the experimental arrangement were the same as those described in [10]. Mixtures of high purity gases were transversely pumped by an electron beam. In the case of XeF($C \rightarrow A$), 8 torr NF_3, 16 torr Xe, and varying amounts of nitrogen were mixed in 6 atm of argon buffer gas. Typical gas mixtures for Xe_2Cl were 1.5 torr CCl_4, 300 torr Xe, 6 atm Ar, and varying amounts of nitrogen. The maximum electron beam energy was 1 MeV and the pump pulse duration was 10 ns. The typical maximum electron beam current density, measured by a Faraday cup on the optical axis of the reaction cell was 200 A/cm^2. An area of 10 cm X 2 cm was pumped by the *e*-beam.

The optical emission from the reaction cell was monitored by two fast vacuum photodiodes (ITT F 4000S). Color glass filters were used to reject the strong fluorescence from the XeF($B \rightarrow X$) or XeCl($B \rightarrow X$) transitions. The photodiode signals were recorded by a Tektronix R7912 Transient Digitizer. The rise time of the detection system was about 2 ns. A 0.25 m Jarrel Ash spectrometer in conjunction with an OMA 1 optical multichannel analyzer was used to observe the temporally integrated emission spectra.

For the XeF($C \rightarrow A$) laser experiments, mirrors with a reflectivity of 98 percent between 420 nm and 500 nm and radii of curvature of 1 and 10 m, respectively, were used. In the case of the Xe_2Cl laser, a mirror with a reflectivity of greater than 99.5 percent at 520 nm and a radius of 5 m and an output coupler with 98 percent reflectivity at 520 nm and a radius of curvature of 10 m were mounted inside the laser cell. In both cases the distance between the mirrors was 14 cm.

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III. Results

A. $XeF(C \rightarrow A)$

The variation of the time-integrated $XeF(C \rightarrow A)$ laser output power with the addition of small amounts of nitrogen was studied. When the entire spectral range of the laser pulse was considered, the laser power decreased monotonically with nitrogen up to 200 torr (Fig. 1). However, when only the short wavelength portion of the laser pulse was considered (453 to 470 nm), the laser power increased when up to 20 torr N_2 was added. Thus, although the reflectivity of the laser mirrors decreased to about 97 percent at 460 nm, the output power from the $XeF(C \rightarrow A)$ laser increased by about 30 percent in this spectral region. The laser power decreased, however, when additional N_2 was added to the $Ar/Xe/NF_3$ laser mixture. Because the laser temporal pulse shape did not change with the addition of nitrogen, the peak laser power behaved the same as the laser energy. When pure nitrogen was used as a buffer gas, no laser action was observed for the pump power density available. Furthermore, the fluorescence from the $XeF(C)$ state around 480 nm decreased when the nitrogen pressure increased. This fluorescence decrease was less pronounced when higher amounts of xenon were used.

The laser spectrum changes considerably when N_2 is added to the gas mixture. Fig. 2(a) shows a laser spectrum without nitrogen, while in Fig. 2(b), 50 torr N_2 was added to a standard gas mixture. Besides the increase of the laser power in the short wavelength part of the spectrum, the absorption features in Fig. 2(b) are considerably richer than in Fig. 2(a).

Many of the absorptions in Fig. 2(a) have been attributed to absorption of metastable xenon species mainly into the Xe-Rydberg series. With no nitrogen present in the gas mixture and in agreement with earlier observations [7], [10], only absorptions from the $Xe(6s^3P_0)$-level and the $Xe(6s^3P_1)$-level (weaker) are observed. However, when nitrogen is added [Fig. 2(b)] absorption lines beyond the ionization limit of the $^3P_0 \rightarrow nf$ $(\frac{3}{2})$ Rydberg series at 462.4 cm [11] can be observed because the laser spectrum now extends to shorter wavelengths. Most of these lines can be attributed to transitions from the $Xe(6s^3P_2)$-level into various $Xe(6p)$ and $Xe(7p)$-states. Several absorption lines are probably due to xenon and argon ions [12].

The absorption valleys observed in the laser spectrum with nitrogen are not quite as deep as in the absence of nitrogen. However, no complete elimination of the absorption lines could be achieved as observed by Bischel *et al.* [7] with photolytic pumping.

B. Xe_2Cl

The Xe_2Cl fluorescence and laser characteristics behaved quite differently compared with $XeF(C \rightarrow A)$. First the fluorescence intensity was independent of the nitrogen admixture up to N_2 pressures of 1 atm in a normal Xe_2Cl gas mixture. With an admixture of 50 torr nitrogen the Xe_2Cl fluorescent intensity as a function of CCl_4 donor pressure was the same as in the absence of nitrogen. Furthermore, only very weak additional quenching of Xe_2Cl fluorescence by N_2 was observed. The quenching rate constant for Xe_2Cl by N_2 was determined to be $(7 \pm 2) \times 10^{-14}$ cm$^3 \cdot$ s^{-1}.

Considerable improvement of the Xe_2Cl laser output was observed, however, when nitrogen was added to the gas mixture as shown in Fig. 3. A threefold increase of the laser power could be achieved by the addition of 200 torr of nitrogen. The decrease in laser power with increasing N_2 pressure above 200 torr is consistent with earlier results where a failure of the Xe_2Cl laser using nitrogen as a buffer gas was reported [10]. In order to get reliable results, several shots with different gas mixtures were used for every experimental data point depicted in Fig. 3. The error bar gives the statistical error in the maximum of the curve.

As in the case of fluorescence, the Xe_2Cl laser intensity shows the same dependence on the CCl_4 pressure with and without the admixture of 200 torr nitrogen. Intense UV-superradiance

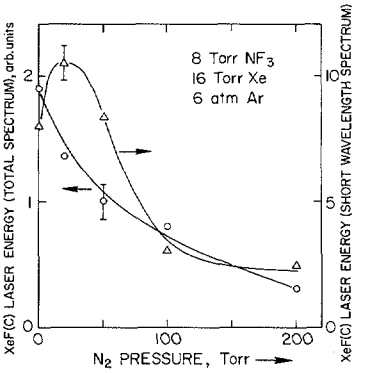

Fig. 1. $XeF(C \rightarrow A)$ laser energy as a function of the nitrogen admixture. The curve corresponding to the left vertical scale depicts the total laser energy. For the curve corresponding to the right vertical scale, the laser energy spectrum was integrated between 453 and 470 nm.

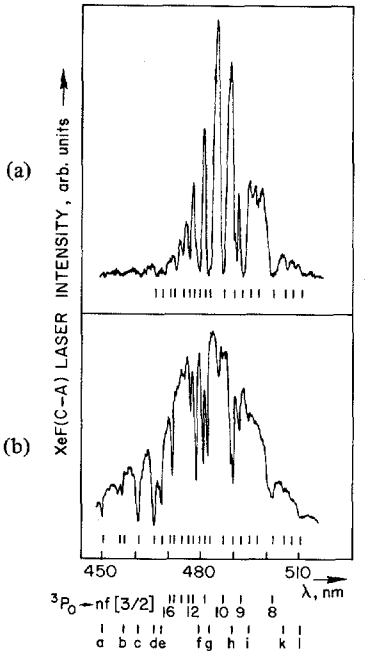

Fig. 2. $XeF(C \rightarrow A)$ laser spectra for a mixture of 8 torr NF_3, 16 torr Xe, and 6 atm Ar (a) and for the same mixture with 50 torr N_2 added (b). Besides the $^3P_0 \rightarrow nf$ $[\frac{3}{2}]$, Rydberg series (n is the principal quantum number) the following absorption lines were identified:

a) $^3P_0 \rightarrow 6p'$ $[\frac{3}{2}]$	*e*) $^3P_2 \rightarrow 6p'$ $[\frac{3}{2}]$	*i*) Ar^+
b) Xe^+	*f*) $^3P_2 \rightarrow 7p$ $[\frac{3}{2}]$	*k*) $^3P_0 \rightarrow 11p$ $[\frac{3}{2}]$
c) Xe^+	*g*) $^3P_1 \rightarrow 7p$ $[\frac{3}{2}]$	*l*) Xe^+
d) $^3P_2 \rightarrow 7p$ $[\frac{5}{2}]$	*h*) $^3P_1 \rightarrow 6p'$ $[\frac{3}{2}]$	

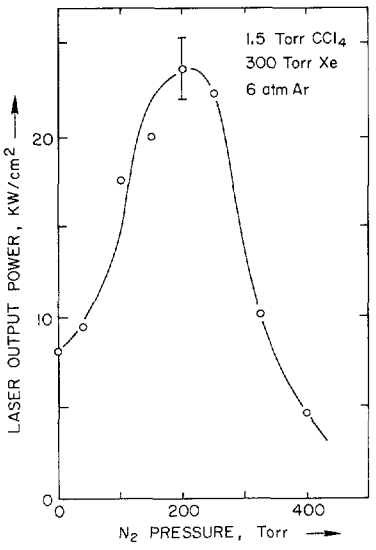

Fig. 3. Xe_2Cl laser power as a function of nitrogen pressure.

on the $XeCl(B \rightarrow X)$-transition at 308 nm which was observed for CCl_4 pressures of more than 1.5 torr without nitrogen was strongly quenched by 50 torr of nitrogen. When neon was used as a buffer gas, no laser action could be observed. The addition of nitrogen did not alter this behavior.

The time-integrated laser spectrum of Xe_2Cl was strongly influenced by nitrogen (Fig. 4). Without nitrogen, the laser spectrum shows deep absorption valleys [Fig. 4(a)]. The most distinct absorption line is due to the transition between the $Xe(6s^3P_0)$ and the $Xe(7f\ [\frac{3}{2}])$ level at 516.4 nm [10]. When 200 torr nitrogen is added, this absorption line is eliminated completely [Fig. 4(b)]. The Ar^+ absorption line at 514.5 nm is, however, not affected by nitrogen admixture. The absorption decrease of the xenon line is fully saturated at less than 100 torr of N_2. The laser power, however, increases up to 200 torr of nitrogen in the gas mixture.

IV. Discussion

The primary role of N_2 as an additive in the present $XeF(C)$ and Xe_2Cl laser experiments can be best understood by consideration of two factors: 1) the influence of N_2 on the production of XeF and Xe_2Cl and 2) the effect of N_2 on the production and loss of the primary absorbing species Xe*, Xe_2^*, and Xe_2^+. Evaluation of the loss of the neutral absorbers is relatively straightforward. Assuming that the measured [13] quenching coefficient of $1.9 \times 10^{-11}\ s^{-1}\ cm^3$ for $Xe(^3P_2)$ by N_2 is also representative of the other $Xe(6s)$ levels, N_2 pressure of 100 torr results in a Xe* quenching time constant of about 20 ns, a time characteristic of the observed lasing onset for the present conditions. Further, since the dimer Xe_2^* is produced from Xe*, it follows that the presence of N_2 will also result in a reduction of both the production and loss of this broadband absorber. For these reasons, the presence of N_2 at the levels typical of this experiment is expected to reduce both discrete and broadband absorption resulting from Xe* and Xe_2^* in both laser mixtures. However, a comparison of Figs. 2 and 4 reveals significant qualitative differences in the changes in absorption observed when N_2 is added to $XeF(C)$ and Xe_2Cl mixtures. Additionally, $XeF(C)$ fluorescence and laser output was found to decrease in the presence

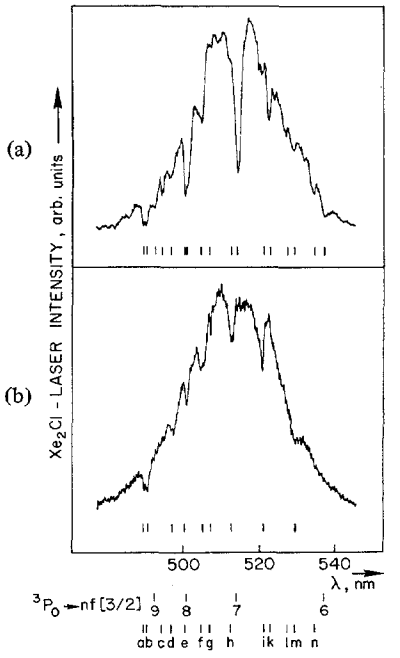

Fig. 4. Xe_2Cl laser spectra for a mixture of 1.5 torr CCl_4, 300 torr Xe, and 6 atm Ar (a) and for the same mixture with 200 torr N_2 added (b). Besides the $^3P_0 \rightarrow nf\ [\frac{3}{2}]$, Rydberg series of Xe* the following absorption lines have been identified [12]:

a) $^3P_0 \rightarrow 13p\ [\frac{1}{2}]$
b) $^3P_2 \rightarrow 7p\ [\frac{5}{2}]$
c) Ar^+
d) $^3P_0 \rightarrow 12p\ [\frac{1}{2}]$
e) Ar^+
f) Ar*
g) $^3P_0 \rightarrow 11p\ [\frac{3}{2}]$
h) Ar^+
i) Ar*
k) Xe^+
l) Xe^+
m) Xe^+
n) $^3P_0 \rightarrow {}^\circ 10d\ [\frac{3}{2}]$.

of N_2 while Xe_2Cl laser output increased although Xe_2Cl fluorescence remained relatively unaffected. These observations are best explained by considering the effect of N_2 addition on charged particle loss processes in the two laser mixtures.

Fig. 5 shows the electron energy dependence of the rate coefficient for dissociative electron attachment [14] to NF_3 and CCl_4. In the absence of N_2 the electron mean energy for both the XeF and Xe_2Cl laser mixtures is expected to be in the 2-3 eV range, typical of *e*-beam excited rare gases [15]. This is a mean energy for which the attachment coefficients for NF_3 and CCl_4 are both very large and of comparable magnitude. Under these circumstances ion production by the *e*-beam is balanced almost entirely by ion-ion recombination in both laser mixtures. Modeling of the present experimental conditions shows that for a quasi-steady electron mean energy of about 2-3 eV during and slightly after the *e*-beam excitation pulse, the negative ions F^- and Cl^- resulting from NF_3 and CCl_4 dissociative attachment recombine with Xe^+ and Xe_2^+ to form XeF and XeCl, respectively, the latter being the precursor of Xe_2Cl [10]. Thus, in the absence of N_2, attachment is balanced by positive ion-negative ion recombination in both mixtures.

Upon addition of N_2 to these laser mixtures at the 1-5 percent fractional concentration levels of this experiment, calculations show that the electron mean energy will be substantially lower (a few tenths of an eV) during and after the excitation pulse as a result of electron cooling due to N_2 vibrational excitation. Examination of Fig. 5 shows that the effect of such a reduction in electron temperature on negative ion pro-

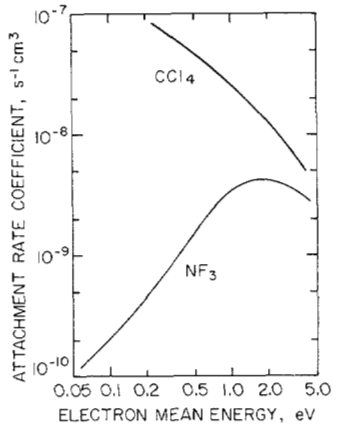

Fig. 5. Electron attachment coefficients for NF_3 and CCl_4 as a function of the mean electron energy (adapted from Chantry [14]).

duction (electron loss) is dramatically different in the XeF(C) and Xe_2Cl mixtures containing NF_3 and CCl_4, respectively. In the former, electron cooling from the few eV range to a few tenths of an eV results in an order-of-magnitude decrease in the rate coefficient for NF_3 dissociative attachment with the result that F^- production and subsequently XeF formation decreases, a result consistent with our observation that XeF($C \rightarrow A$) fluorescence decreases as the N_2 concentration increases. The quenching of XeF(C) by N_2 should be negligible even at nitrogen pressures of the order of 100 torr [16]. Additionally, in this case the decreased attachment results in a much larger electron density so that electron dissociative recombination with Xe_2^+ dominates the charged particle loss, i.e., in the limit of a very low attachment coefficient ion, production by the e-beam is balanced by electron-ion recombination rather than by attachment. Since the product of $e + Xe_2^+$ recombination [17] is Xe*, although N_2 serves to quench Xe* and Xe_2^*, its presence in the XeF mixtures also contributes to an increase in the production of these absorbers. While the net effect of N_2 on the density of absorbing species may be beneficial, when considered in light of the large decrease in Xe^+-F^- recombination, addition of N_2 results in a decrease in XeF(C) laser output.

In marked contrast to the situation in XeF mixtures in which NF_3 is the halogen donor, Fig. 5 shows that electron cooling in the Xe_2Cl mixture due to N_2 addition results in an order-of-magnitude increase in the CCl_4 attachment coefficient. Although the rate coefficient for Cl^- production increases sharply as the electron energy decreases, the net effect of this increase on the absolute magnitude of ion-ion recombination (and therefore XeCl production) is of second-order importance since for this mixture attachment is the dominant electron loss process throughout the entire 0.2-2.0 eV electron energy range of importance. Thus, for the conditions of the Xe_2Cl mixture with or without N_2, XeCl formation by way of ion-ion recombination is sensibly constant, being determined by the magnitude of the e-beam excitation pulse. That is, so long as attachment is always the dominant electron loss process, the magnitude of the attachment coefficient itself is not particularly important. This interpretation is consistent with our observation that Xe_2Cl fluorescence is relatively insensitive to N_2 addition over a broad range of N_2 fractional concentration. Although an enhanced electron attachment coefficient does not affect the XeCl* production, modeling of the present experimental conditions shows that Xe_2^+, Xe*, and Xe_2^+ populations decrease with increasing electron attachment coefficient. Therefore, discrete and broadband absorption by these species are reduced by the addition of nitrogen, causing a significant increase in the Xe_2Cl laser output. These absorption processes are particularly important because recent calculations by Stevens and Krauss [19] show that the absorption of Xe_2Cl laser light by the Xe_2Cl molecule is negligibly small.

Clearly, high energy pulsed e-beam excitation of the high pressure mixtures typical of the XeF and Xe_2Cl lasers considered here results in a highly ionized, highly excited plasma medium of a type for which there is relatively little experience. Nonetheless, the differences in the magnitude and the electron energy dependence of the attachment coefficients in NF_3 and CCl_4 in response to the electron cooling certain to occur with N_2 addition to rare gas mixtures are so large as to dominate over other known reactions. In conclusion, the present observations accompanying the addition of N_2 to XeF(C) and Xe_2Cl laser mixtures using NF_3 and CCl_4, respectively, can be interpreted in a satisfactory manner on the basis of changes associated with the different attachment characteristics of the two laser mixtures and the quenching of absorbers by nitrogen.

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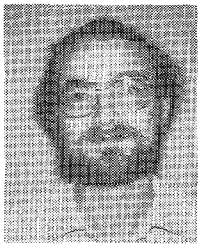

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Frank K. Tittel (SM'72), for a photograph and biography, see p. 902 of the May 1982 issue of this JOURNAL.

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Generation of Short Optical Pulses in Semiconductor Lasers by Combined dc and Microwave Current Injection

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Abstract—**We report picosecond pulse generation in low threshold buried optical guide lasers using combined direct and microwave current excitation. The pulse widths were obtained as a function of direct current for several levels of RF excitation using lasers 125 and 380 μm long. The pulses have a Gaussian shape with full widths at half maximum intensity ranging from 19 to 57 ps. The pulse widths were obtained from the second harmonic autocorrelation. The experimental results are in reasonable agreement with the theory of short pulse formation in sinusoidally modulated lasers. The pulse width decreases with increasing dc and microwave current, and decreasing laser length. The shortest pulses were obtained with a 125 μm long laser using 0.25 W RF at 1 GHz and 35 mA dc bias. Multiple pulses are emitted at high dc excitation levels. Using the above laser at the indicated current levels the emission consists of a burst of ~10 pulses which are separated by 30 ps, and has an approximately exponentially decaying amplitude. Saturable absorption was introduced in the lasers by degradation and results in shortening the pulses. A comparison is made of the pulse widths obtained for sinusoidal microwave current modulation and for pulsed excitation where the excitation is obtained from a step recovery diode. Reduced pulse widths are obtained for short current pulse excitation.**

Manuscript received December 14, 1981; revised May 14, 1982.
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I. INTRODUCTION

THERE are several methods for obtaining short optical pulses from semiconductor lasers. We describe the particularly simple technique, yielding picosecond pulses at gigahertz repetition rates, which utilizes combined dc and microwave current injection [1]-[7]. We present a theory of pulse formation in modulated lasers with the emphasis on the dependence of the pulse width on the dc and microwave current and the other laser parameters. Experimentally, a new low threshold buried optical guide (BOG) laser geometry is utilized. Pulses widths ranging from 19 to 57 ps are obtained. Similar excitation schemes have been used to obtain pulses of 30 ps duration [3], 14 ps duration [5], and 28 ps duration [6] using various laser geometries. Short pulses have also been obtained using short (50-150 ps) excitation pulses obtained from microwave current driven step recovery diodes (comb generators) [8]-[10] as well as other short electrical pulses [11], [12]. We find that the pulse widths obtained by the pulsed current injection are somewhat shorter than for direct microwave current modulation.

Considerably shorter pulses have previously been obtained

0018-9197/82/0900-1340$00.75