

with lower dispersion would increase the speed of response and possibly enable the direct observation of hot carrier transient effects. In current experiments, a second optical pulse in the infrared is used to turn off the switch, in a similar manner to the previous low-voltage measurements,¹ to make an optical gate with a variable-duration transmission window.

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High-efficiency KrF excimer laser*

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Efficient high-power laser emission has been observed at 249 nm from a KrF excimer laser obtained by an electron-beam-pumped mixture of Ar, Kr, and NF₃ (1300:130:1) at a total pressure of 2.25 atm. An energy of 1.5 J was extracted in a 125-nsec (FWHM) pulse from a 100-cm³ volume, using a coaxial electron-beam laser. Laser efficiency was estimated to be nearly 15% based on energy deposition in the gas. Over-all electrical efficiency was ~1%.

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Gaseous excimers have been recognized to be a rich source of new laser transitions. These molecules are particularly important because they are characterized by repulsive or very weakly bound ground electronic states and offer the possibility of tunable high-power high-efficiency lasers in the visible and ultraviolet spectral regions. Due to repulsive ground states, saturation intensities are expected to be high. High efficiency is partly due to transitions terminating in the ground state and consequently yielding high quantum efficiencies. High saturation intensity may also be useful in decreasing the effect of quenching and thereby increasing the efficiency.

Nonradiative losses from the laser upper state appear to be a serious problem in achieving high-efficiency emission from several uv lasers. Effects of this loss may be minimized by having a larger rate of stimulated emission compared to that of quenching. However, in lasers using bound-bound transitions, such as Ar-N₂¹ and XeF,² the saturation intensity limits the rate of stimulated emission. With a bound-free transition, a larger rate of stimulated emission is possible for successful competition against nonradiative losses. In support of the above discussion, we wish to report the achievement of efficient high-power laser emission from the krypton fluoride (KrF) excimer at 249 nm obtained by an electron-beam-pumped mixture of argon, krypton, and NF₃ (1300:130:1) at total pressures from 2 to 3.5 atm. KrF laser action has been previously reported by Ewing and Brau³ using a mixture of Ar, Kr, and F₂. We believe the use of NF₃ is more desirable since it is not as corrosive as fluorine and does not absorb at the laser emission wavelength.

Fluorescence and initial laser experiments were carried out using an arrangement described earlier¹ for the Ar-N₂ transfer laser. The emission spectrum of KrF obtained from a mixture of 2 Torr NF₃ and 3.4 atm Ar containing 10% Kr is shown in Fig. 1(a). Spectra of the rare-gas halides such as KrF arise⁴ from transitions originating at a Coulombic upper electronic state and terminating on a repulsive or weakly bound ground state. The absence of any sharp structure in the KrF emission spectrum indicates that the transition is of the bound-free type. Furthermore, the narrow width (4 nm) of the emission band is understandable in terms of the transition terminating in a relatively flat portion of the repulsive potential curve. The slight band intensity alteration appearing as actually two bands is not inconsistent⁵ with a bound-free transition. Due to a large degree of vibrational relaxation at high pressures,

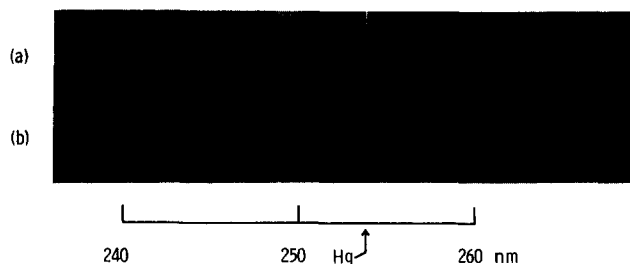


FIG. 1. (a) Emission spectrum of KrF from a mixture of 2 Torr NF₃ and 3.4 atm Ar with 10% Kr obtained by electron-beam pumping. (b) Laser spectrum of the same mixture. The intensity has been attenuated by a factor of 10⁴ to prevent film saturation.

emission is expected primarily from $v'=0$ of the upper state. A very faint band is also visible [Fig. 1(a)] towards the short-wavelength side separated by nearly 350 cm^{-1} . This separation agrees with vibrational level spacings of the upper state estimated by Tellinghuisen.⁵

The laser spectrum consists of a single narrow band at 249 nm. Laser emission was so intense that it had to be attenuated by a factor of 10^4 to prevent film saturation. Spectral line narrowing with simultaneous increase in intensity, observed with properly aligned cavity mirrors, gives conclusive evidence of laser oscillations. Laser output energy was measured to be 100 mJ in a 10-nsec (FWHM) pulse at a total pressure of 3.4 atm. An electron energy deposition calculation¹ indicates that nearly 2 J was absorbed by the gas, giving a laser efficiency of 5%.

Additional experiments were performed with a coaxial electron gun.⁶ The optical cavity, consisting of two 75% partial reflectors, produced an output energy of 1.5 J (125-nsec FWHM) at a total pressure of 2.25 atm (Ar:Kr:NF₃ at 1300:130:1). A one-dimensional electron-beam deposition calculation based on Berger and Seltzer⁷ shows that nearly 10 J of beam energy was absorbed by the 100-cm³ gas volume, giving an estimated efficiency of 15%. Since the stored energy in the Marx generator was about 200 J, an over-all electrical efficiency near 1% was achieved.

In the electron-beam-excited mixture of Ar, Kr, and NF₃, energy is primarily deposited in Ar. For every 91 eV lost by the high-energy electron, 3.5 Ar⁺ ions and one Ar* (³P metastable) are created.⁸ Through a unique set of kinetic processes at high pressures, most Ar⁺ species are⁹ converted into Ar*. Energy transfer from Ar* to Kr occurs¹⁰ with 100% efficiency. The 10% mixture of Kr also absorbs some energy directly from the electron beam which is converted into Kr* by processes similar to those in Ar. The estimated Kr* density is $\sim 10^{15}/\text{cm}^3$.

The Xe* + F₂ reaction, which seems to give a high ($\geq 60\%$) yield¹¹ of XeF*, is similar⁴ to the formation of an alkali halide in an alkali-halogen reaction. Since the fluorine donor properties of NF₃ appear to be similar to those of F₂, the Kr* + NF₃ reaction is also expected¹¹ to efficiently form KrF*. After emission to the ground state, KrF is dissociated. Free fluorine atoms may combine with NF₂ to reform NF₃. This is consistent with the observation that the same gas mixture containing only 2 Torr of NF₃ could be used over 20 times without any reduction in intensity.

The radiative lifetime of KrF* is expected to be similar to that measured by Searles and Hart¹² for XeBr (~ 20 nsec). These data combined with the linewidth of 4 nm give an estimated stimulated emission cross section of $5 \times 10^{-17}\text{ cm}^2$. With an intracavity power of 8

MW/cm² in the coaxial device, the stimulated emission rate is found to be $\sim 5 \times 10^8/\text{sec}$ which is estimated to be higher than the quenching rate of KrF* by Kr (or possibly NF₃). Since the terminal laser state is expected to dissociate in a time typical of molecular vibrational time period ($\sim 10^{-13}$ sec), KrF laser saturation intensity should be $\sim 10^{12}\text{ W/cm}^2$. Thus it is possible to increase the intracavity power until the stimulated emission rate is significantly higher than the quenching rate.

Achievement of high efficiency in KrF* is aided by the lack¹³ of NF₃ absorption at this wavelength. Photoionization, which plagues the Xe₂⁺ excimer laser, may be absent in the KrF laser. Since the energy of the laser photon is 5 eV and the binding energy of the KrF* is similar¹¹ to that for RbF (~ 6 eV) photoionization should not occur in this laser. In the electron-beam-pumped laser nearly 20 eV is required to create Ar*. Since virtually every Ar* may result in a 5-eV KrF laser photon, the theoretical efficiency is expected to be $\sim 25\%$ which is consistent with the observation using the coaxial electron-gun laser.

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