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[21]Meterological conditions were approximately: 70–80 °F, 60% relative humidity, moderate winds. Dates: 14–16 August 1979.

A triatomic Xe_2Cl excimer laser in the visible

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A new triatomic rare-gas halide laser using the molecular exciplex Xe_2Cl is described. Laser emission centered at 518 nm with a special bandwidth of 30 nm and peak power of about 2 kW was obtained from an electron-beam-excited high-density $Ar/Xe/CCl_4$ mixture.

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Rare-gas halide excimers have been shown to be efficient high-power laser media at several fixed wavelengths in the UV.[1] However, the usefulness of these lasers in a number of applications would be greatly enhanced if they were tuned continuously over broad wavelength bands in both the UV and the visible. Recently there has been considerable interest in a tunable blue-green diatomic XeF ($C \rightarrow A$) excimer laser.[2–5] In addition, triatomic rare-gas halides have been proposed as laser media which in principle may provide UV and visible wavelength tunability.[6,7] The possibility of electron-beam-excited laser action in Xe_2Cl has been suggested by Tang, Lorents, and Huestis,[8,9] who reported transient gains of about 6% per pass at 514.5 nm in the afterglow following an initial absorption peak.

In this letter we describe the first observation of laser action in Xe_2Cl from electron-beam-excited high-pressure $Ar/Xe/CCl_4$ mixtures. The experiments were carried out in a stainless-steel cell attached to the field emission diode of a Pulserad Model 110 electron beam generator as described in detail in Ref. 10. A beam of 1-MeV electrons with a pulse width of 10 ns (FWHM) and a current density of 0.8 kA/cm^2 was injected transversely into the laser medium through a 50-μm-thick titanium anode foil over an area of 10 cm^2. This resulted in an input beam energy of 80 J. The laser cavity was formed by two reflectors positioned in the cell as shown in Fig. 1. The sapphire or fused-silica vacuum windows were placed external to the cavity, thereby avoiding reflection losses which are significant for a low-gain laser medium. One reflector was a 1-m-radius mirror ($R > 99.8\%$ from 480 to 510 nm), while the other was a flat output mirror ($R = 98\%$). The two mirrors were separated by 10 cm and the optical axis was 1.5 cm from the anode foil. The bellows

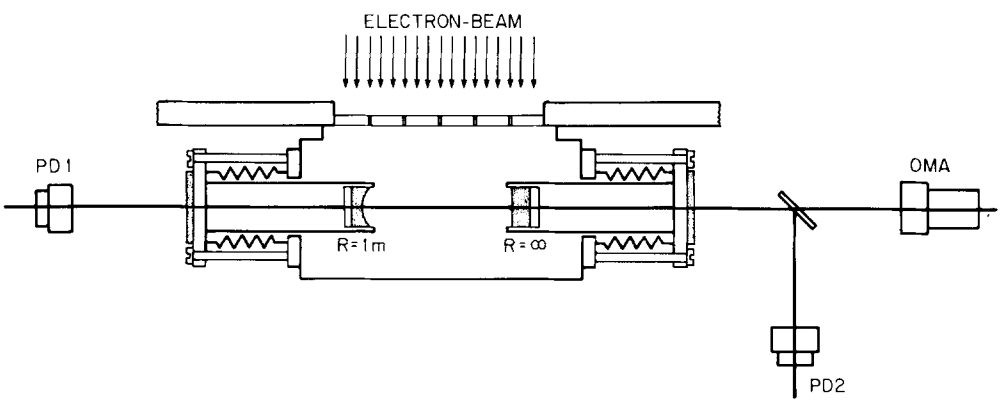

FIG. 1. Schematic of electron-beam-pumped laser cell and associated diagnostic instrumentation.

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design of the mirror mounts permitted convenient external alignment of the intracell resonator. The fluorescence and laser signals were monitored in the axial direction by ITT F4000 S-5 photodiodes (PD_1 and PD_2) and simultaneously by spectrograph or a calibrated PAR OMA 1 optical multichannel analyzer.

Transitions of interest in emission from XeCl and Xe_2Cl excimers are shown in Fig. 2, in which the potential-energy curves are presented for these species. In the case of XeCl [Fig. 2(a)] both an intense, narrow-linewidth $B \rightarrow X$ transition and a weaker, broadband $C \rightarrow A$ transition are observed at 308 and 345 nm, respectively.[11] For triatomic Xe_2Cl broadband emission centered at 500 nm occurs between strongly bound ionic upper states and repulsive ground states,[7-9] as shown in Fig. 2(b).

Fluorescence measurements were used to determine the optimum composition of a gas mixture for enhanced Xe_2Cl emission as compared to XeCl emission, which is always present, since it can be considered as a precursor in the reaction chain finally leading to the formation of Xe_2Cl.[9] The range of pressures of argon, xenon, and CCl_4 evaluated was 4–9 atm, 100–750 Torr, and 0.5–10 Torr, respectively. Low xenon pressures (< 100 Torr) favored increased XeCl excited-state production. A time-integrated fluorescence spectrum obtained with the OMA for a typical mixture of 6.5-atm Ar, 200-Torr Xe, and 1-Torr CCl_4 is shown in Fig. 3. The Xe_2Cl spontaneous emission has a spectral bandwidth of approximately 100 nm (FWHM) and exhibits weak absorption features.

On the other hand, the Xe_2Cl laser spectrum which has a center wavelength of 518 nm shows significant spectral narrowing from 100 nm (spontaneous emission) to 30 nm (laser). In addition, greatly enhanced intracavity absorption features similar to the diatomic XeF ($C \rightarrow A$) laser was observed. The laser output spectrum is red shifted relative to the fluorescence spectrum by approximately 20 nm. Since the reflectivity of the laser mirrors remains nearly constant over this spectral range, cavity pulling effects can be excluded. Instead, the observed shift is due to the spectral dependence of the stimulated emission cross section. Furthermore, the cavity enhanced absorption features shown in Fig. 3 can be compared to the visible atomic and molecular absorption characteristics for the electron-beam-pumped argon diluent reported in Ref. 12. Two observed absorption peaks at 519 and 537 nm are in good agreement with the published data on molecular absorptions in argon.[12] In fact the intracavity laser enhancement of absorption features reveals additional features at 493, 503, and 532 nm which may be attributed to enhanced Ar absorption or to transient absorptions of the other components (Xe, CCl_4) not observed by the single-pass absorption measurements.

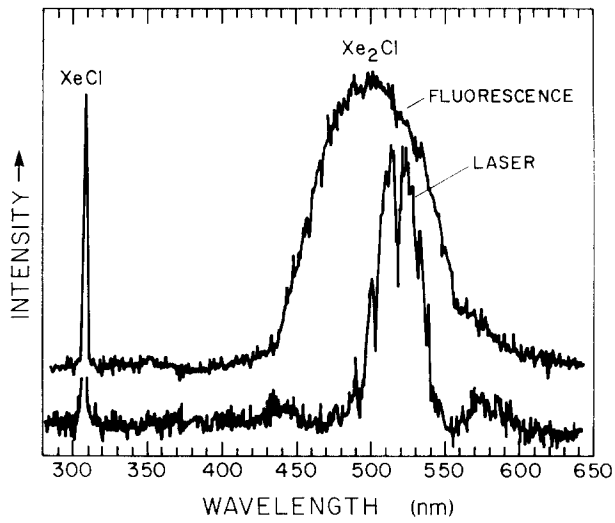

FIG. 3. Fluorescence and laser spectrum of Xe_2Cl and XeCl ($B \rightarrow X$) of an $Ar/Xe/CCl_4$ mixture of 6.5 atm, 200 Torr, and 1 Torr, respectively. All intensities are shown in arbitrary units, not corrected for the spectral response of the OMA Model 1205D/01 vidicon detector.

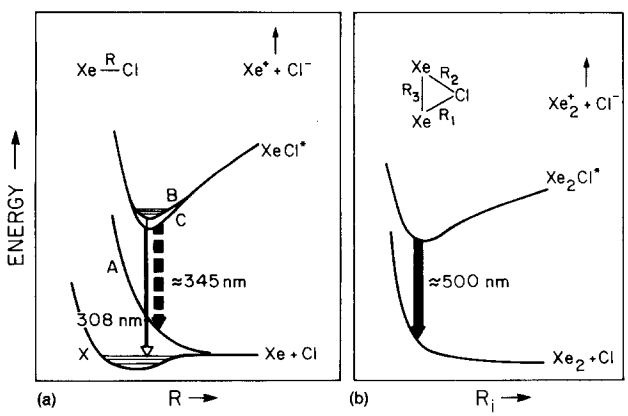

FIG. 2. Simplified potential-energy diagram of the diatomic excimer XeCl and triatomic excimer Xe_2Cl.

At present laser operation is close to threshold and limited by the low signal gain, the short time available for the buildup of stimulated emission and stable cavity oscillations, and by visible absorption effects. The peak laser power as measured by a fast photodiode was approximately 2 kW.

In summary, we have demonstrated the operation of the first triatomic excimer laser using an electron-beam-pumped $Ar/Xe/CCl_4$ mixture. However, to obtain broadband tunability in the blue-green it will be necessary to develop a resonator with a low-loss dispersive element for the Xe_2Cl laser. This will require further suppression of the stimulation of XeCl transitions and visible absorbing species by optimization of the rare-gas halide mixture and the electron beam characteristics. Also, experiments are in progress aimed at developing a XeCl ($C \rightarrow A$) laser tunable in the UV from about 330 to 370 nm based on preliminary fluorescence measurements. This will require a cavity centered at 345 nm but lossy at 308 nm in order to suppress the main XeCl ($B \rightarrow X$) transition.

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High-resolution saturated absorption spectroscopy with coherent trains of short light pulses

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The feasibility of saturated absorption with two counterpropagating trains of short light pulses from a frequency-locked, synchronously pumped, mode-locked, cw dye laser is demonstated. In contrast with the experiments using a single-frequency laser, several velocity classes contribute to the signal, thus increasing the signal-to-noise ratio. In addition, the advantages reported for two-photon spectroscopy (precise frequency scale, increase of the second harmonic, etc.) are available in this new technique.

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In Doppler-free saturation spectroscopy cw or pulsed quasimonochromatic laser beams were used exclusively until now. In that case only one velocity class of gas molecules participated in the generation of the saturation signal. We report in this letter the results of saturated-absorption experiments with two counterpropagating trains of short light pulses from a mode-locked cw dye laser, where several velocity classes of molecules contribute simultaneously to the signal.

We performed the experiments on iodine molecules, using a synchronously pumped, mode-locked cw laser whose jitter was reduced by a servo-locking technique.[1,2] The 1.6-MHz recorded linewidths of the individual modes are comparable to that of a frequency-locked single-mode cw dye laser.[3] The possibility of measuring line separation with great accuracy leads to a precise determination of the hyperfine constants, which are an order of magnitude better than in the previous measurements using an external optical frequency marker.

While two-photon excitation of a gas of atoms or molecules with a coherent train of standing-wave light pulses can be viewed as a linear interaction between an effective field and a two-level system[4] and analyzed in the time domain, the fundamental nonlinear character of saturated absorption prevents a similar simple treatment. However, if the dipole relaxation time of the investigated transition is much larger than the delay time between two pulses, in such a way that one molecule experiences many pulses before the decay of the coherences induced by the first pulse, the interaction can then be easily understood in the frequency domain, and a very clear analysis can be made. The two counterpropagating beams can be described as a superposition of phase-locked oscillating laser modes which are equally spaced in frequency. The saturating beam, propagating along the z axis, perturbs the thermal equilibrium of the velocity classes v_z of molecules resonant with each of the laser modes. The equally spaced holes burned in the velocity distribution are then analyzed by the frequency comb of the probe beam propagating in the opposite direction. When the laser mode comb is translated in frequency, the transmission of the probe beam will be enhanced each time at least one of its modes interacts with the same molecules as does one mode of the saturating beam. If n denotes the number of laser modes, one obtains then for each transition ω_0 $2n-1$ equally spaced fringes free of first-order Doppler broadening, with a fringe spacing equal to half the laser mode spacing. From this simple picture it can easily be seen that several velocity classes contribute to each fringe. The number of these contributions increases when considering the fringes closer and closer to the fringe comb center where it is equal to the number of

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