

A discharge-pumped ArCl superfluorescent laser at 175.0 nm

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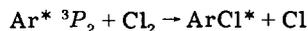
Initial observations have been made of laser action in ArCl excimers pumped by a high-voltage fast-rise-time Blumlein discharge circuit. The new laser emits at 175.0 nm and operates at atmospheric pressure. The gas from which the molecules are formed consists of 1% Cl₂, 15% Ar, and 84% He. A gain coefficient of 0.012 cm⁻¹ has been determined for ArCl.

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Observations of numerous rare-gas halide lasers (XeF, XeCl, XeBr, KrF, ArF, and KrCl) have been made using electron beam excitation.¹⁻¹⁰ Some of these molecules have also exhibited stimulated emission in discharge-pumped systems.¹¹⁻¹⁵ This paper reports the first observations of superfluorescent emission from ArCl at 175.0 nm. This wavelength is the shortest yet obtained from rare-gas halide excimers. It was produced with a fast (2.5 ns current rise time) Blumlein discharge circuit, which also was capable of exciting XeF, KrF, ArF, and KrCl with no difficulty. (This is also the first report of discharge pumping of KrCl.)

While the basic operating principles of the traveling-wave Blumlein discharge system have been described previously¹⁶ for use in generating vacuum ultraviolet emission from H₂ and CO, the system used for this work was extensively modified to produce a uniform discharge at higher pressures. These modifications included the creation of a gas chamber capable of handling several atmospheres of pressure and new electrodes raised from the polyethylene Blumlein dielectric. The electrodes, constructed from stainless steel, were 160 cm long, 2.5 cm high, and could be varied in separation from 1 to 7 cm. Electrically the Blumlein circuit was triggered by the sequential firing of nine Mylar switches timed to produce a traveling wave of excitation in the direction of proposed pulse propagation (this has also been called the "swept gain" mode of operation). The resulting voltages across the electrodes could be varied from about 80 to 110 kV. Several electrode shapes were tried before settling on a nearly flat pair which gave a uniform discharge free from arcing with pressures in the $\frac{1}{2}$ -1-atm range.

The emission spectra of the rare-gas halides were first studied by Golde and Thrush¹⁷ and Velazco and Setser.¹⁸ The emission spectrum of ArCl formed by the reaction



is given by Golde and Thrush and shows a rather broad-band (~8.0 nm) spectrum with a maximum at 175.0 nm and a secondary peak at about 169.0 nm. Ewing and Brau also observed the emission spectra of the rare-gas monohalides and realized the possibility of uv and vacuum uv lasers.¹⁹ They were able to estimate the features of a number of these lasers, and some of these lasers have now been realized. Because of an interest in short wavelengths the work reported here is an initial attempt at investigating those rare-gas halides which

are predicted to lase in the vacuum ultraviolet.

Initial experiments with this device were carried out in XeF, and the spectrum at 355 nm showed the discrete lines characteristic of transitions between stable molecular levels. The gas mixture used was 1.5% NF₃, 5% Xe, and then filled with He to make total pressures of up to 500 Torr. Attempts to generate emission from KrF produced poor results with NF₃, and attempts to generate ArF with NF₃ produced only a weak spontaneous spectrum accompanied by numerous lines attributable to the excitation of NF₃ or its decomposed fragments. Substitution of pure F₂ eliminated all difficulties in producing stimulated emission in KrF and ArF as shown in Fig. 1. Gas mixtures consisted of about 1% F₂, 10% Ar or Kr, and the rest He to make pressures of about 1 atm. It is possible that pressures above 1 atm may produce higher powers, but this was not investigated because of our interest in obtaining shorter wavelengths. Therefore, Cl₂ was substituted for F₂, and the laser emission spectra shown in Fig. 1 were obtained from KrCl and ArCl. Using similar gas mixtures and pressures with Cl₂ as used with F₂, lasing was easily obtained.

All of the excimers lased in the superfluorescent (or amplified spontaneous emission) mode of operation. No resonant cavity was needed to obtain lasing, although as Burnham¹¹ has indicated, considerable improvement could be expected in the output when resonant cavities are employed. The emission was extracted through a LiF window and traversed a vacuum path of about 1 m

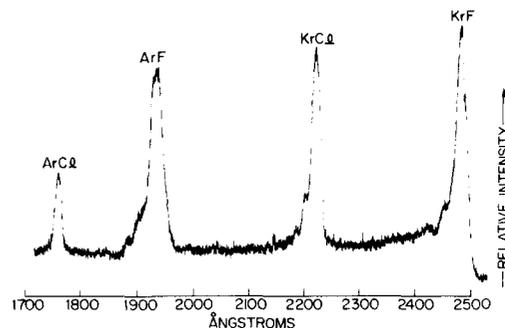


FIG. 1. Spectra of discharge-pumped superfluorescent laser emission from four rare-gas halides. Each laser line was recorded in vacuum on the same piece of Kodak 101-01 film by placing the appropriate gases in the discharge chamber. Hg lines were used for calibration.

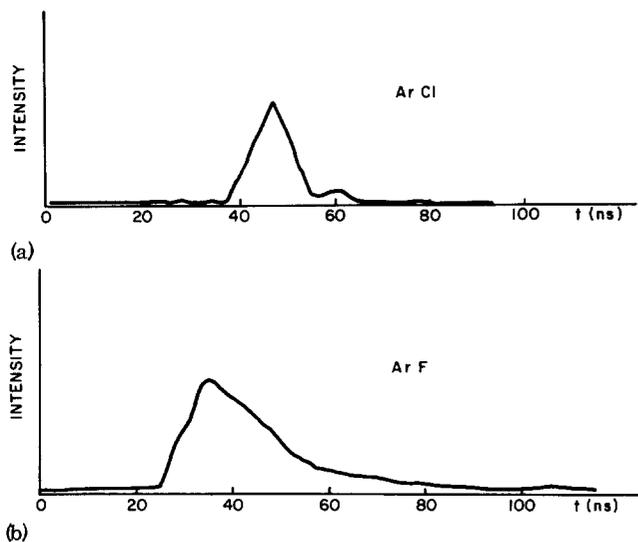


FIG. 2. Traces of typical time resolved emission waveforms from (a) ArCl and (b) ArF.

in length before entering a McPherson Model 225 1-m vacuum spectrograph. The dispersed radiation was recorded on Kodak 101-01 vacuum uv film. A Hg lamp was used to calibrate the wavelengths.

Evidence of amplification was obtained by numerous methods. The most straightforward method consists of measuring the output as a function of length of excited gas. For lengths below saturation intensity an exponential dependency would occur when amplification takes place. By placing an absorbing material (a glass plate) across the discharge channel to keep emission from one end of the discharge from reaching the remaining excited gas, the effective length can be varied without changing the discharge parameters anywhere along the length of the electrodes. Observations were made of exponential behavior when the amplifying length was cut from 160 to 80 cm. These indicate unsaturated amplification for the excimers investigated and allow measurement of the small-signal gain coefficients of 0.012 cm^{-1} for ArCl, 0.017 cm^{-1} for ArF, 0.018 cm^{-1} for KrCl, and 0.019 cm^{-1} for KrF.

The temporal history of the amplified output was monitored both by a photomultiplier-spectrograph system and by the use of bandpass filters manufactured by Acton Research which were placed over an ITT 4018 photodiode with a sapphire window. Typical pulses for ArCl and ArF are shown in Fig. 2. The output pulse widths of KrF and ArF were longer (FWHM $\sim 20 \text{ ns}$) than the pulse widths of KrCl and ArCl (FWHM $\sim 10 \text{ ns}$). Similar observations can be found in the comparison of KrF and KrCl by Murray and Powell.⁹

There is likely to be a considerable improvement in

the output power of ArCl and the other rare-gas halides by optimizing electrode structure, gas mixture, and resonator configuration. Present energy estimates made with a Molelectron calorimeter (J3-02) indicate about 0.2 mJ for ArCl, 1 mJ for ArF, 1.3 mJ for KrCl, and 1.7 mJ for KrF from this system. Therefore, the peak power generated in these gases ranges from 20 to 90 kW.

Numerous applications of ArCl might be expected as performance improves. One application is that of pumping doped crystals to produce lasing over wavelengths tunable from 175 to 260 nm.²⁰ Such tunable wavelengths would likely have numerous applications in photochemistry and isotope separation. As Ewing and Brau predict,¹⁹ it is likely that other rare-gas halides can be made to lase at even shorter vacuum ultraviolet wavelengths. Several candidates which remain to be tried are KrBr which should lase at 203 nm, KrI at 185 nm, ArBr at 161 nm, and NeF at 107 nm.

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