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TITLE LIQUID EXCIMERS: LASING Xe_2 and Kr_2 IN LIQUID ARGON

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Liquid Excimers: Lasing Xe_2 and Kr_2 in Liquid Argon

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ABSTRACT

By pumping a small cryogenic cell with a 40-ns, 1 MeV e-beam pulse, we have excited lasing in dilute mixtures of xenon or krypton in liquid argon. The lasing occurred at 175 nm for the excimer Xe_2 and 147 nm for Kr_2 .

ARTICLE

The first excimer laser was demonstrated in early 1970 by Shank et al. [1-3], who lased optically-pumped polyatomic dye molecules. (The distinction between exciplex and excimer has apparently fallen into disuse.) This first lasing was followed in the same year by Basov et al. [4] with the first true excimer laser; a beam-excited Xe_2 (at 175 nm) in liquid xenon. The mainstream of excimer laser research, however, followed the gaseous rare gas-halogen excimer forms established in 1975 [5,6], which resulted in the

development and commercialization of a new family of effective UV lasers.

Having the excimers in liquid form offers the advantage of reducing the volume of the lasing medium. The fluorescence work of Rhodes' group [7,8] and ours [9] also showed that in the liquid form, polar excimers are red-shifted by large amounts, so that liquid excimer lasers would also offer a whole new set of tunable short wavelengths. (These lasers are tunable in two senses; tunable within the broad lasing line, and solvent-tunable, as the red shift is dependent on the molecular weight of the solvent.) Liquid excimers should retain the classic excimer efficiencies, since liquid densities usually produce radiative decay rates that are lower than collisional pumping rates. In this letter we report the lasing of Xe_2 (175 nm) and Kr_2 (147 nm) in a liquid argon host. Other recently published reports of liquid excimer lasers include Shahidi et al. (XeF in liquid argon, [10]) and Loree et al. (XeO in liquid argon, [11]).

A schematic of the cryogenic laser cell is shown in Figure 1. The vacuum-isolated cell was cooled by pulses of liquid nitrogen vapor regulated by a thermocouple-driven temperature controller which maintained cryogenic temperatures to better than $\pm 1\text{K}$. The 1 MeV, 40-ns e-beam pump pulse entered the side of the cell through a 50 μm nickel foil. The e-beam energy

deposited in the lasing medium was 0.3 J, measured calorimetrically. The penetration depth of the electrons in liquid argon limited the active volume to about a 2 mm depth; the height was 2 mm and the length 19 mm; thus the pump power density was on the order of 100 MW/cm^3 . Both mirrors were mounted directly on the ends of the cell and sealed with spring-loaded calcium-lead alloy gaskets to the diamond-turned faces of the OHFC copper body. This sealing system survived many pressure and thermal cycles from room temperature down to nominal operating conditions (104 K and 2.8 atm) and back. The high-reflector mirror was overcoated aluminum film with a concave 10-cm radius and the output coupler was flat, varying from a bare magnesium fluoride window to another aluminum HR as required.

Early in these investigations we found that if the argon host were contaminated by oxygen at tens of parts per billion, the only emission that would appear would be the fluorescence of ArO in the green, no matter what the ostensible mix was. This acute sensitivity to impurity oxygen was not found by Rhodes' group in optically-pumped mixes [12], but has been noted in studies of solid rare gases [13]. In our case it proved sufficient to purify research-grade argon gas by flowing it through a Hydrex Ti gettered purifier. As most of our mixes contained less than 1% of the lasing species, our mixing was usually carried out in two

steps. A primary mix was made with several percent of the lasant (all mix values are room temperature pressure ratios) which was further diluted with argon to the desired concentration. When the final mix was felt to be well homogenized, it was directly condensed in the cold optical cell. We tried to maintain the accuracy of the mix by always starting runs at the lowest concentrations and moving monotonically to richer mixes. This procedure minimized the influence of residual gases in the system on the concentrations.

The reproducibility of the lasing results was greatly improved when we added the pump shown in Figure 1. The stirring afforded by the centrifugal pump both prevented thermal striations and kept the xenon or krypton from forming large ice crystals (both are solids at the operating temperature of the laser). The efficacy of the circulation could be appreciated by aligning a He-Ne laser beam through the cell when it was equipped with a pair of windows. When the pump was turned off, the He-Ne beam broke up in a matter of seconds as thermal layering took place. The 10-mm diameter pump was run at 1500 rpm by a magnetically-coupled external motor.

The temporal measurements of the lasing and the sidelight fluorescence were made with ITT F4115 VUV photodiodes in vacuum. Energies were also inferred from the photodiode signals after calibration (in the exact

geometries used) with measured ArF laser beams at 193 nm. No corrections were made for the reduced photodiode response at the liquid excimer wavelengths. The spectral measurements were made with a vacuum spectrometer; the output was viewed by a Tracor Northern intensified diode array with a magnesium fluoride input window. This setup successfully detected Ar₂ fluorescence at 130 nm.

The lasing line of Xe₂ centered on the fluorescence peak at 175 nm and narrowed to 1.0 nm wide at half-maximum (the fluorescence was 9.0 nm wide). Note that in the gas phase the lasing occurs at 172 nm; the red shift in the liquid is small because Xe₂ (and Kr₂) molecules are homopolar [8]. In contrast, the red shift of the lasing line for the polar molecule XeF in liquid argon is 53 nm.

Figure 2 illustrates the temporal characteristics of the sidelight and lasing. The fluorescence reflects the 40-ns pump pulse shape with a depletion notch at the moment of lasing; the lasing pulse was about 4 ns wide. Both of these Xe₂ figures are data taken with a 0.1% mix, which was near optimum for energy and reproducibility, although mixes from 0.05 to 15% Xe lased. In Figure 3 we show the change in the lasing wavelength as the xenon concentration was increased. This saturating red shift was caused by the increasing average molecular weight of the local universe

surrounding each lasing molecule.

The best Xe/Ar mixes would lase with a bare magnesium fluoride window as the output coupler. The output from this setup was attenuated by several 90-degree reflections (from bare magnesium fluoride windows) and detected by a previously-calibrated (at 193 nm) photodiode. This measurement indicated a reproducible energy output of 10 mJ. To cross-check this number, we put a piece of burn paper on the output window; when compared to burns taken at 193 nm, this rough energy measurement suggested a value greater than 5 mJ. A less quantitative but more dramatic measure of the lasing energy was the clean hole blown in the Al coating of the high reflector after some tens of shots. At 10 mJ, the tiny lasing volume produced better than 100 J/l, a beam with 10 MW/cm², and a gain of at least 35% cm.

The mixes of Kr/Ar clearly had less gain than the Xe₂, and required a pair of high reflectors to induce lasing; at 147 nm the "HR's" were down to 80% reflectivity and leaked a useful amount of the beam for diagnostics. Figure 4 shows the temporal narrowing of the lasing compared to the fluorescence. The time behavior again suggests the uncomplicated nature of these simple excimers. The lasing wavelength occurred at the peak of the fluorescence, and was narrowed from 8.0 nm to 1.0 nm at half-maximum. The range of

concentrations that lased was about 0.3-20% Kr. The red shift with increasing concentration was not measured. The lasing was rather erratic, but comparisons of the leakage signal through a common aluminum-coated HR showed the energy to be down an order of magnitude from the Xe₂ output. This would imply an energy density in the medium of some tens of joules per liter.

Both media exhibited apparently straightforward kinetics, in contrast to XeC, which lased after the pump pulse was finished and did so in a series of gain-switched spikes [11]. While these are not new lasing molecules [4,14], forming the excimers in an argon host is a technique that may have favorable physical and economic consequences. We have, in these preliminary experiments, demonstrated high energy densities and wavelengths that can be tuned (both within the line and by solvent effects) to new vacuum UV regions. The issue of reabsorption of the emission by the excimer molecule, an important limitation in the gaseous rare gas excimers, was not examined in these dilute liquid mixes. If a specific need arises, these are potentially efficient and useful lasing systems.

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Xe_2 TIME HISTORIES

Xe Lasing Solvent Shift (Xe in Ar)

Kr₂ TIME HISTORIES