C.14 Phonon-Assisted Laser Transitions and Energy Transfer in Rare-Earth Laser Crystals,[1] H. P. Jenssen and A. Linz, *Center for Materials Science and Engineering and Department of Electrical Engineering, Massachusetts Institute of Technology, Cambridge, Mass.*

Energy transfer between rare-earth ions in crystals is a process that can be successfully used in rare-earth laser crystals as evidenced by successful CW operation[2] of Ho^{3+}:YAG at 77 K and by the recently reported 2.06-μm Ho^{3+}:YLF laser operated at room temperature.[3] In both cases Er^{3+} and Tm^{3+} were used as sensitizers.

In this work an extensive study of energy transfer and phonon interaction in rare-earth-doped $LiYF_4$ (YLF) has been conducted. First the crystal-field split energy levels of the rare-earth ions were determined by liquid He temperature absorption and fluorescence measurements. It was found that only in very few cases could the spectra be explained using the static crystal field model alone. The vibronic transitions were very strong, especially for ions near either end of the rare-earth series. The effective density of phonon states[4] for YLF was obtained from the vibronic transitions accompanying the Pr $^3H_4 \rightarrow {}^3P_0$ transition at 5°K. The effective density of states reaches maximum at 300 cm^{-1} and then rapidly falls off at higher energies. The resonance between a vibronic transition and an electronic transition enormously enhances the absorption or fluorescence at that energy. The Pr $^3H_4 \rightarrow {}^1D_2$ transition is a good example of this in YLF. Here two of the electronic levels of Pr $^1D_2$ are separated by 300 cm^{-1} and the resonance effect is clearly evident in the absorption spectra. In the Ho $^5I_7 \rightarrow {}^5I_8$ emission spectrum at 2.065 μm there is also resonance between electronic transitions and vibronic transitions. McCumber[5] treated the phonon-terminated laser as observed in Ni^{2+}:MgF_2. A similar treatment for the Ho^{3+}:YLF case shows that the stimulated emission cross section here is increased by the presence of resonant vibronics, by a factor of up to 4.5 over what would be expected from the measured absorption cross section.

A new technique for investigating energy transfer was used. Crystals with high enough concentration of sensitizer and activator ions to assure near neighbor pairs were used to measure transfer rates as a function of energy gap. In a crystal containing equal concentrations of Ho^{3+} and Er^{3+}, for example, five possible transfer steps can be measured with energy gap ranging from near zero to about 1800 cm^{-1}. The transfer rates were obtained by measuring the sensitizer lifetime with and without the activator present, and also by quantum efficiency measurements. An exponential dependence of transfer rates with respect to energy gap was found. The multiphonon relaxation rates in YLF were also measured and had the same dependence on energy gap as the transfer rates, but were 5×10^4 higher for a given energy gap.

[1] This work was supported in part by the Office of Naval Research under Contract N00014-67-A-0204-0044 and in part by the Advanced Research Projects Agency under Contract SD-90 and DAHC 15-67C0222.

[2] L. F. Johnson, J. E. Geusic, and L. G. Van Uitert, *Appl. Phys. Lett.*, vol. 8, p. 200, 1966.

[3] E. P. Chicklis, C. S. Naiman, R. C. Folweiler, D. R. Gabbe, H. P. Jenssen, and A. Linz, *Appl. Phys. Lett.*, vol. 19, p. 119, 1971.

[4] E. Cohen, L. A. Riseberg, and H. W. Moos, *Phys. Rev.*, vol. 175, p. 521, 1968.

[5] D. E. McCumber, *Phys. Rev.*, vol. 134, p. A299, 1964.

SESSION D

Monday, May 8, 1972 2:00–5:00 P.M.

Ultraviolet and X-Ray Lasers

D.1 Generation of Ultraviolet Radiation in Phase-Matched Metal Vapors *(Invited)*, G. C. Bjorklund, D. S. Carson, S. E. Harris, A. H. Kung, R. B. Miles, and J. F. Young, *Microwave Laboratory, Stanford University, Stanford, Calif.*

This paper will report theoretical and experimental work on tripling and mixing of laser radiation in alkali metal vapors. Theory predicts and experiments confirm a third-order nonlinear susceptibility about 10^5 times greater than that of previously studied inert gases. It is of key importance that the high localized oscillator strengths of vapors such as rubidium, cesium, sodium, etc., cause the refractive indices at frequencies below their fundamental resonance line to be greater than their refractive indicies at the third-harmonic of these frequencies. It is then possible to add an inert buffer gas to attain phase matching.

Experiments and calculations are now underway with the objectives of generating tunable radiation in the spectral region of 1800 to 2500 Å and of generating a number of discrete ultraviolet and vacuum ultraviolet wavelengths between 1100 and 3300 Å. These experiments will differ from that of our previous work by in each case allowing two of the generating frequencies to span the atomic resonance line. This results in roughly a factor of 100 increase in coherence length and allows higher alkali metal vapor pressures to be employed. In particular cases it will not be necessary to use any buffer gas whatsoever. For instance, the process of mixing two photons of 5320-Å light with one photon of 6220-Å light in sodium vapor to generate one photon at 1863 Å has a coherence length of 12 cm at a sodium vapor pressure of about 15 torr. The low losses of these metal vapors above their ionization potential should allow this technique to be extended to about 300 Å. Theoretical studies of the effect of atomic saturation both in destroying phase matching and causing thermal defocusing are also underway. Under certain conditions, these effects will limit the allowable incident energy density to a much lower value than that given in our earlier publication.

D.2 Electron-Beam Gas-Laser Excitation, R. W. Dreyfus and R. T. Hodgson, *IBM Thomas J. Watson Research Center, Yorktown Heights, N. Y. 10598.*

This paper reports an approach that successfully introduces extremely high power in a short pulse into laser systems. We used a commercial field-emission electron-beam device to excite N_2, Ne, and H_2 superradiant gas lasers at 3371, ~3300, and 1161–1613 Å, respectively. In this paper, the various laser characteristics, the overall behavior of electron beams, and the energy transfer to the gas are considered in detail.

The experimental arrangement is relatively simple. A 175-cm-long 1- or 2-cm ID tube is bolted to the face of a Febetron[1] 706 electron-beam generator. This device emits ~10^4 A of ~400-keV electrons in a 3-ns pulse. The 1-cm^2 electron beam would normally diverge, but the electrons are constrained by an ~6-kG axial magnetic field to propagate 160 cm down the tube. The tube is filled with the different gasses at pressures ranging from 5 to 400 torr. If the pressure is too high (e.g., ~70 torr N_2), a well-defined beam does not propagate the whole length of the tube, even though the range of a single 400-keV electron is very much larger than the tube length. Evidence of beam instabilities have been noted on the measured beam current, and these instabilities presumably lead to loss of the beam.

A number of criteria are used to test for lasing action. They include spectral analysis of the light, fluorescence time reduction, high power output, and unidirectional emission of the light.

Since the N_2 laser light at 3371 Å is in an experimentally accessible spectral region, and the appropriate collision cross sections are known, in this paper we will concentrate on the N_2 laser characteristics. The next paper will give further details on vacuum ultraviolet laser transitions in H_2 as demonstrated by spectral analysis of the emitted light.

The light output from electron-beam bombarded N_2 at 20 torr was recorded spectrographically and with a 0.4-ns rise-time calibrated photodiode. Typical N_2 laser spectra consisting of some rotational lines of the 0-0 band ($C^3\Pi_u - B^3\Pi_g$) in the second positive system were recorded near 3371 Å. The photodiode showed 60-kW pulses with 2-ns pulse lengths. This is considerably shorter than the 35-ns fluorescence time for the $C^3\Pi_u$ state. When a magnetic mirror was used to reflect the electrons at the end of the tube, the pulse lengthened to ~7 ns.

The angular distribution of the emitted light was measured to be 10 mrad by directly exposing Polaroid film at various distances from the end of the tube. This divergence corresponds to light emitted at a point source near the electron gun which is restricted by the aperture at the far end of the tube. The energy was estimated to be ~450 ergs from the film density.

The laser power is estimated theoreti-

[1] Field Emission Corporation.