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25 October 1965

3p.

Fourth Quarterly Letter Report . no. 4

High Energy Crystalline Laser Materials .

R. C. Ohlmann .

ARPA Order 30662

Contractor: Westinghouse Elec. Corp.
Contract Date: 10/16/64
Amount of Contract: \$58,845

Contract No. Nonr. 34658(00)

Contract Expiration Date: 10/16/65
Project Scientist: Dr. R. C. Ohlmann
Phone: (412) 242-1500, Ext. 414

The object of the contract is to perform research using spectroscopic techniques on samples of potential laser materials as indicated below:

- (1) Double doped samples (rare earths and transition metal ions) of $YAlO_3$, $CeAlO_3$, $LaAlO_3$, and $Y_3Al_5O_{12}$ (YAG).
- (2) U^{4+} in CeO_2 and ThO_2 .
- (3) Eu^{2+} , Gd^{2+} , Sm^{2+} , Ce^{2+} or Mn^{2+} in grossularite, $Ca_3Al_2(SiO_4)_3$.

The ultimate objective is to obtain improved laser materials for high energy lasers.

During the fourth quarter of this contract, the instrumentation for studying near-infrared fluorescence was completed and the fluorescence of Ho^{3+} in $Y_3Al_5O_{12}$ at wavelengths of from 1.9 to 2.15 microns was studied. The excitation spectrum of the fluorescence at 2.0975μ from crystals double doped with chromium and holmium showed that energy transfers from Cr^{3+} to Ho^{3+} in this host material. Particularly evident is the existence of a broad excitation band at $6000\text{\AA}$ where Cr^{3+} absorbs and Ho^{3+} does not absorb. This agrees with

* This work was performed as a part of Project DEFENDER under the joint sponsorship of the Advanced Research Projects Agency, the Office of Naval Research, and the Department of Defense.

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the results recently published⁽¹⁾ on the coherent oscillations from Ho^{3+} in

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1. L. F. Johnson, J. E. Geusic and L. G. Van Uitert, Appl. Phys. Letters 7, 127 (1 Sept. 1965).
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YAlG , where it was reported that a sample sensitized with 0.5% Cr^{3+} had about 60% of the pulse threshold of an unsensitized sample.

The fluorescence of crystals of YAlG containing manganese has been studied. Both Mn^{2+} and Mn^{4+} emission is evident. The overlap of Mn emission wavelengths with rare-earth excitation wavelengths suggest that it may be possible to sensitize Nd^{3+} with Mn^{2+} and Ho^{3+} with Mn^{4+} . However, the Mn concentration in the crystals obtained up to this date is not high enough for efficient energy transfer to occur.

Since GdAlO_3 doped with Cr may be a high-power Q-spoiled laser material, techniques for obtaining single crystals have been developed. Boules over an inch long were pulled from the melt after solving the problem of considerable supercooling which occurs in this material. Absorption, fluorescence and excitation spectra have been obtained on relatively clear 3mm thick slabs of $\text{GdAlO}_3:\text{Cr}$ (0.2%) down to 30°K. The five R-lines in emission are about 10Å wide and about 10-12Å spacing and do not narrow appreciably at low temperatures. It appears their relative intensities may be following a Boltzmann temperature distribution which indicates interaction rate between the states causing the emission is at least faster than the spontaneous emission rate (decay time ~18 milliseconds at 77°K). The long decay time, moderate line width and absorption strength which allows 0.5% Cr concentration in a laser crystal means that its capability of energy-storage per unit volume is an order-of-magnitude greater than ruby.

Crystals of grossularite were obtained from Linde which were hydrothermally grown and were doped with europium and samarium under reducing conditions. The fluorescence and excitation spectra of this material indicated, however, that the ions were primarily in the trivalent state. These spectra and their analysis will appear in the final report.

A search for fluorescence from uranium ions (at 1 atomic percent) in CeO_2 and ThO_2 from 4500Å to 3.2 microns was unsuccessful at both 300°K and 77°K. The sensitivity was such that a fluorescence emission less than 1% of that of Ho^{3+} in YAlG at 2.09μ would have been observed.

The preparation of materials for this contract was under the direction of Dr. R. Mazelsky and the spectroscopic studies and overall direction were supervised by Dr. R. C. Ohlmann.