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ENERGY TRANSFER IN SOLID LASER MATERIALS

Technical Summary Report No. 2

1 Aug - 31 Oct 64

ARPA Order No. 225 (78)

W. W. Hansen Laboratories of Physics
Stanford University
Stanford, California

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INTRODUCTION

This report is the Second Technical Summary Report under Contract Nonr 225(78), which began on 1 May 1964, and it reports the period of 1 August through 31 October 1964. At the present time there are three projects active under this contract:

1. Ultraviolet excitation of ruby
   A. Nature of the excited states in ruby
   B. Energy transfer between levels and between ions in ruby
2. Energy transfer between unlike ions in lanthanum fluoride

The Responsible Investigator for this contract is A. L. Schawlow.

OBJECTIVE

The purpose of this investigation is to study methods of exciting solid materials for visible, and possibly ultraviolet, maser action, and of energy transfer processes between energy levels and between ions.

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Low-speed wind-tunnel tests of a large-scale inflatable structure paraglider (Essais en soufflerie à faible vitesse d'un aéro-voilier à structure gonflable à grande échelle).

GAMSE (B.), MORT (K.W.) et YAGGY (P.P.).
NASA Technical Note, no D-2859 (6/65), 30 p., 9 fig. et photos, 1 tabl., 3 réf. bibl. [Me. 62].

Résultats d'essais concernant les performances et les caractéristiques aérodynamiques longitudinales de cet aéro-voilier faisant partie du programme de recherche sur la récupération d'un véhicule spatial du type capsule.
I. ULTRAVIOLET EXCITATION OF RUBY

The purpose of this investigation is to study the suitability of light for pumping ruby lasers. Wherever the peak intensity of flash lamps is increased by operating at high currents, the greatest fractional increase occurs in the ultraviolet region. We are, therefore, investigating the nature of the excited states in ruby, and the processes of energy transfer between them.

A. NATURE OF THE EXCITED STATES IN RUBY

(G. K. Klauminzer, H. W. Moos, P. L. Scott)

The previous report described the investigation, in progress at that time, of the $^2E \rightarrow ^2T_2$ absorption in ruby. To review briefly, the experiment was undertaken to determine if absorption from the excited state $^2E$ does indeed occur at wavenumbers 14,400 cm$^{-1}$ less than absorption from the ground state to the same upper levels. Components of the two levels $^2E$ and $^2T_2$ were accurately located by absorption from the ground state. Energy differences were computed as those where $^2E \rightarrow ^2T_2$ absorption ought to occur.

When radiation of the desired wavelength was passed through ruby pumped with a flashlamp, sharp absorptions appeared at positions corresponding within experimental error to those calculated. Line widths also agreed with those expected. Thus it appears that the simple energy difference explanation suffices, at least for this low energy transition.
In addition to locating the $^2E \rightarrow ^2T_2$ absorption lines, we measured their intensities in $\delta$ and $\pi$ polarizations. The infrared radiation traversing the ruby was polarized by transmission through a stack of microscope slides set at the Brewster's angle. Results were in qualitative agreement with those predicted from selection rules calculated by a method similar to that of Tanabe and Sugano.1

The oscillator strength for the transition was calculated after the population of the $^2E$ level was determined. The latter was found by observing the absorption from the ground state to the $^4T_2$ band when the pumping lamp was flashed. The decrease in absorption upon pumping corresponded to a 5% depopulation of the ground state, or, alternately, a 5% population of the $^2E$ level assuming an efficiency of 1. With this figure the oscillator strength was determined to be approximately $2 \times 10^{-5}$.

B. ENERGY TRANSFER BETWEEN LEVELS AND BETWEEN IONS IN RUBY

(J. L. Emmett, R. L. Greene, S. A. Johnson and W. M. Yen)

Work on a direct test of pure ultraviolet light for laser excitation is awaiting the completion of a high intensity pulse excitation apparatus. It is expected that this apparatus will be available within the next quarter. It will use a double-pulsed flash lamp with xenon or oxygen filling. A liquid filter solution will remove the visible light.

Work on spatial energy transfer will be resumed shortly.
II. ENERGY TRANSFER BETWEEN UNLIKE IONS IN LANTHANUM FLUORIDE

(W. C. Scott, W. M. Yen)

We have continued studies of the low temperature (4.2°K) linewidths and lineshapes of optical transitions of 4f ions in doubly doped solids and find that additional insight may be gained on the energy transfer mechanisms that occur between the impurity systems. In particular we have concentrated our efforts on the 5985 Å (3P0 → 3H6) transitions of Pr3+ in an LaF3 matrix doubly doped with Pr3+ and Nd3+. It was found by some of us that in the singly doped LaF3:Pr3+, because of the relatively long lifetimes of the initial and terminal states of the 5984 Å transition, the width and shape of this transition is inhomogeneously broadened by lattice imperfections, i.e., the lineshape is Gaussian.2 It was further reported3 that with the addition of Nd3+ into the system LaF3:Pr3+, the 5985 Å Pr3+ transition begins to exhibit a broader homogeneous (or Lorentzian) component which at higher Nd3+ concentration completely dominates the lineshape. We construed that this component is evidence of a Pr3+(3H6) → Nd3+(4I13/2) transfer process, where the width of the component reflects the transfer probability. The magnitude of the Lorentzian portion of the line indicates the number of Pr3+ ions that are engaged in the transfer process.4

We obtained a new set of LaF3 samples where the Pr3+ concentration was kept at 0.1% and the Nd3+ concentration was varied from 0% to 5% in discrete steps. The samples were obtained from Varian Associates and
were grown in the same run in order to assume minimal sample variation. Detailed widths and shape studies were made of the $5985 \AA$ Pr$^{3+}$ transition at $4.2^0K$ in fluorescence and using a 1.8 m. Jarrel-Ash scanning spectrometer.

In order to separate the narrow inhomogeneous component from the broader homogeneous one, the scan traces $G(\omega - \omega_0)$ were Fourier transformed about the $\omega_0$ center frequency into a time domain. It is well known$^5$ that a Gaussian $G(\omega)$ will transform into a Gaussian $F(t)$, whereas a Lorentzian will transform into an exponential $e^{-\alpha t}$ where $\alpha$ is connected to the linewidth. It is then a simple matter to obtain the separation using semi-log $t$ and $t^2$ plots and extrapolating. In so doing, we find that the homogeneous component is, in most cases, describable by a single exponential and that the inhomogeneous component is to within 15% a constant in all the samples.

The decay constant $\alpha$ increases linearly with Nd$^{3+}$ concentration up to $\sim 2\%$ Nd$^{3+}$ and then shows a saturation effect. The ratio of the homogeneous to inhomogeneous component should yield the portion of Pr$^{3+}$ ions engaged in the transfer process. Unfortunately, because of initial wing deviations$^2,6$ from the Gaussian lineshape in the $5985 \AA$ line of LaF$_3$:Pr$^{3+}$ no such analysis is possible until the wing deviations are better understood. At a Nd$^{3+}$ concentration of $\sim 0.5\%$ the homogeneous component completely dominates the line.

Work will continue on the detailed theoretical aspects of this problem.
REFERENCES

5. See, for example, A. Abragam, Principles of Nuclear Magnetism (Clarendon Press, Oxford, 1961) Chapter III.