ENERGY TRANSFER IN SOLID LASER MATERIALS

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INTRODUCTION

This report is the sixth Technical Summary Report under Contract Nonr 225(78), which began on 1 May 1964, and it reports the period of August through 31 October 1965. At the present time there are two projects active under this contract:

1. Ultraviolet excitation of ruby
   - A. Nature of the excited states in ruby
   - B. Energy transfer between unlike ions in LaF_3
2. Excited state spectroscopy

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.

I. ULTRAVIOLET EXCITATION OF RUBY

The purpose of this investigation is to study the suitability of ultraviolet 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

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

To evaluate the effects of the ultraviolet radiation in laser pumping or in removing atoms from the metastable state, a ruby rod was surrounded by a transparent cylindrical sheath. When the sheath is of quartz, it transmits radiation down nearly to 2000 Å. When the sheath is pyrex, all radiation below about 3000 Å is absorbed, while longer wavelengths are transmitted as well as through the quartz sheath. In other experiments, a cylindrical liquid filter sheath of either water or copper sulfate solution contained in quartz was used.

With the various filters, the laser output from the ruby rod was measured at several flashlamp energies.

During this period the difficulties reported previously were overcome and consistent results were obtained. It was found that the integrated laser output with the quartz sheath is 30% to 40% less than

with the pyrex sheath. The output with no sheath is comparable to that with the quartz sheath. Thus, removal of some of the short wave ultraviolet is found to increase the ruby output.

In the second experiment the ruby rod was pumped while surrounded first with a filter of distilled water and then with a filter of $CuSO_4$ solution. The $CuSO_4$ concentration was adjusted so that the transmission between 3500 Å and 5000 Å matched that of the water. The transmission of the $CuSO_4$ solution was negligible below 2800 Å. The output was lower for the $CuSO_4$ filter at low flashlamp pumping levels (100 J), but gave a 15% greater output at higher pumping levels (300 J). The initially lower output with the $CuSO_4$ filter can be understood, since the distilled water and the $CuSO_4$ solution absorb differently in the region of the usual green absorption (3000 Å to 5600 Å). At higher pumping energies with correspondingly more ultraviolet light, this difference appears to be overcome by the ultraviolet absorption from the ^2E state.

Both of these experiments lead to the conclusion that intense ultraviolet light is detrimental to ruby laser output. More detailed experiments will be necessary to determine the exact mechanism that causes this effect.

Meanwhile, a report of this work and related work sponsored by the U. S. Air Force has been accepted for publication in Applied Optics as a Letter to the Editor.

B. ENERGY TRANSFER BETWEEN UNLIKE IONS IN LaF_3

(W. M. Yen, R. L. Greene, W. C. Scott)

Optical linewidth and line-shape studies of 4f impurity ions have been made in doubly-doped systems. It was found that interactions which involve excitation transfer between the impurity systems produce homogeneous broadening and line-shape changes in the optical transition.

In particular, we have studied the behavior of the 5985 Å line of Pr^{3+} in doubly-doped $LaF_3:Pr^{3+}$, Nd^{3+} as a function of Nd^{3+} concentration. The 5985 Å Pr^{3+} transition at 4.2°K involves two long lived electronic states of the Pr^{3+} ion; it follows that in the singly-doped $LaF_3:Pr^{3+}$ the transition shows a Gaussian shape which is characteristic of inhomogeneous strain and imperfection broadening. As Nd^{3+} is added to the system, a broader Lorentzian component begins to appear in the 5985 Å Pr^{3+} transition and eventually (Nd^{3+} concentrations of ~ 1% or above) completely dominates the width and shape of the transition. The Lorentzian component of the transition line-shape was successfully separated using a Fourier transform technique, and the width of this component was studied as a function of the Nd^{3+} concentration. The width is found to increase <u>linearly</u> with increasing Nd^{3+} concentrations, the linewidth saturates and approaches an asymptotic value of ~ 2 cm^{-1}. We construe this homogeneous component to be evidence of a rapid short range energy transfer mechanism between the Pr^{3+} and Nd^{3+} systems at the terminating $(^3H_6)_1$ level of the 5985 Å Pr^{3+} transition. Various mechanisms have been investigated as causes of the energy transfer process; it is concluded that the spin exchange interaction is the most probable cause for the transfer. Results of other spectroscopic measurements on the doubly-doped $LaF_3:Pr^{3+}$, Nd^{3+} system have also been obtained.

An account of this work has now been published.[1]

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[1] W. M. Yen, R. L. Greene, W. C. Scott, and D. L. Huber, Phys. Rev. <u>140</u>, A1188 (1965).

II. EXCITED STATE SPECTROSCOPY

(J. L. Emmett, S. Johnson)

In the course of our various investigations on energy transfer mechanisms, a need inevitably develops for information on lifetimes of excited states. This is the sort of information that we must obtain to explain the harmful effects of ultraviolet radiation on ruby laser action. In other words, we lack a basic diagnostic tool.

Some excited-state spectroscopy has been performed by G. K. Klauminzer (Lt., U.S.A.F., now recalled to active duty). However, the point-by-point method which he used is too slow and tedious for general use. The slowness is particularly objectionable when exploring for totally unknown excited-state absorptions.

We have, therefore, begun serious consideration of the design of a fast gated spectrograph, which could record an entire spectrum at a given time after an initiating pulse. Such an instrument might use a gated image intensifier followed by a photographic plate. At least one commercial image intensifier camera could be used in this way.

Another method of excited-state spectroscopy would use a gated image orthicon tube. In this case, the light intensity at all parts of the spectrum would be stored simultaneously on the tube target. It could then be scanned into a digital memory bank (Enhancetron), and the process repeated many times for better signal-to-noise ratio. The orthicon would have the advantage of high quantum efficiency characteristic of photocathodes. Unlike a scanning photoelectric instrument, however, it would record the entire spectrum at one time. It would thus make most efficient use of the light available for short pulse spectroscopy.