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(4) ENERGY TRANSFER IN SOLID LASER MATERIALS

(9) Technical Summary Report No. 4,

1 Feb - 30 Apr 65.

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INTRODUCTION

This report is the fourth Technical Summary Report under Contract Nonr 225(78), which began on 1 May 1964, and it reports the period of 1 February through 30 April 1965. 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, and
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.

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, P. L. Scott)

Measurements of absorption from the metastable ^2E level of Cr^{3+} ions in Al_2O_3 (pink ruby) have been continued. In particular, the polarizations of the ^2E to $^2T_2$ transitions have been measured.

As described previously, the chromium ions are excited to the metastable state by a flash of light from a xenon lamp. A smaller probe light is pulsed about 2 milliseconds after the first flash, in order to measure the absorption produced by populating the ^2E levels.

With the aid of the pulsed probe lamp, our preliminary measurements of polarization have been confirmed with greatly improved accuracy. Qualitatively, they are in accord with crystal field theory. Those lines predicted to be σ-polarized (electric vector $\perp$ crystalline c-axis are σ-polarized), while those lines predicted to be Π-polarized

Π-polarized (electric vector // crystalline c-axis). The simple theory assumes the equality of certain matrix elements, and this leads to a prediction that all σ-polarized lines should be eight times stronger than the corresponding Π-polarized lines. Experimentally, they are nearly equal. However, the equality of these matrix elements is really just an assumption; other reasonable assumptions can account for the observed results.

Crystal field theory has well-known limitations. However, the present results show that it is adequate to describe the absorption of ruby, not only from the ground state but also from the metastable ^2E state, at least to the $^2T_2$ levels.

B. ENERGY TRANSFER BETWEEN LEVELS AND BETWEEN IONS IN RUBY

(R. L. Greene, A. L. Schawlow, 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 pumping 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.

For most of this period, the comparison of the two sheath materials was handicapped by poor reproducibility. The difficulty was caused by the temperature rise in the ruby rod after flashing. This heating occured not only because of pumping light absorption in the ruby rod,

but also because of hot air from the flashlamp. The lamp is directly below the ruby and so continues to heat the crystal for some time after each flash. When the ruby is heated, the spectral line broadens, so that less light amplification is produced by a given number of excited atoms.

A thermocouple was then attached to the ruby rod. Then each flash after the first one was delayed for some minutes until the thermocouple retured to a chosen temperature. This was better, but still not dependable as the rod temperature could be, and apparently was, nonuniform. Finally, some consistency was obtained by waiting for fifteen minutes or so between flashes. The thermocouple then indicated that the ruby rod had time to reach and hold a fixed temperature for several minutes. However, this procedure is quite tedious, and so better cooling will be provided.

The results so far are quite preliminary, but appear to indicate that the peak laser power is about 25 percent greater with the glass sheath than with the quartz. As shown earlier by Gires and Mayer [Ann. de Radioelectric 18, 112 (April 1963)], ultraviolet light can be absorbed by chromium ions in the metastable ^2E state. Presumably, the ions are then excited to a still higher state, from which they may later revert to the ^2E state. If all of the ions return within the 10 microseconds duration of the flash, and do not excite any others in the process, there should be little change in the laser intensity. Gires and Mayer state that the excited atoms return to the metastable state within 50 microseconds. Our results indicate that at least some of them do not return within 10 microseconds. Moreover, the laser pulse,

when the quartz sheath is used, has a weak tail for about 20 microseconds after the main pulse. Possibly this indicates a time constant of the order of 20 microseconds for the return to the ^2E state. However, these results are very tentative and will require much investigation.

II. ENERGY TRANSFER BETWEEN UNLIKE IONS IN LaF_3

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

We have investigated several mechanisms which allow rapid energy transfer between impurity ion system in the search for likely homogeneous interaction responsible for the broadening we have observed in the 5985 Å Pr^{3+} in doubly doped $LaF_3:Pr^{3+}$, Nd^{3+}. We have also measured the lifetime of the $^3P_0$ state (originating state of the 5985 Å line) of Pr^{3+} as a function of Nd^{3+} concentration, and find that the broadening does not occur in this state. This made it necessary to determine the positions of Nd^{3+} states which are energetically near the terminating $(^3H_6)_1$ state of the Pr^{3+} transition. This was accomplished during this quarter.

The magnitude of the broadening implies very strong coupling between the ions. Weaker interactions (e.g., quadrupole-dipole, phonon-phonon) may thus be ruled out. This leaves the dipole-dipole interaction and the spin-exchange interaction as likely candidates for the energy transfer process. Our calculations indicate that the dipolar interactions lead to widths of the order 10^{-3} cm^{-1}, whereas the spin-exchange interaction leads to a width of ~ 1 cm^{-1}. The magnitude of the latter is roughly equivalent to what one observes experimentally. We may conclude that transfer of excitation to the Nd^{3+} ion occurs at the

$(^3H_6)_1$ Pr^{3+} state via spin-exchange interactions.

Results of this work have been written up and submitted for publication in Physical Review under the title "Optical Linewidth and Lineshape Studies of Energy Transfer Mechanisms in Rare-Earth Ions" (W. M. Yen, R. L. Greene, W. C. Scott, and D. L. Huber). A brief version of the work will be presented at the Physics of Quantum Electronics Conference, San Juan, Puerto Rico, June 28-30, 1965, and will appear in the proceedings of the conference.