STIMULATED OPTICAL EMISSION FROM EXCHANGE-COUPLED IONS OF Cr^{+++} IN Al_2O_3

Irwin Wieder and Lynn R. Sarles
Varian Associates, Palo Alto, California
(Received December 19, 1960)

The advantages of chromium ions in aluminum oxide crystals (ruby) for double resonance experiments have been pointed out[1,2] and utilized in several microwave-optical experiments.[3-5] Recently, stimulated emission[6] and coherence[7] of the characteristic "R_1" fluorescence at 6943 A has been observed in pink ruby (~0.07% Cr^{+++} by weight). We wish to report the observation of stimulated emission at wavelengths 7010 A and 7040 A from transitions in red ruby (~0.7% Cr^{+++} by weight) which arise[8] from exchange coupling between neighboring chromium ions.

The observations were made by monitoring the fluorescence of a $\frac{1}{8}$-in.$\times 1\frac{1}{2}$-in. red crystal rod following an intense excitation flash. By virtue of the broad optical absorption bands and the efficient radiationless transfer to sharp excited states, these levels accumulate an excess population which may satisfy the requirements[9] for stimulated optical emission. At high excitation energy the fluorescence at 7010 A and 7040 A exhibits the initial decrease from the natural radiative lifetime which is characteristic of stimulated emission. However, the decay curves show significant differences from the previous results with the R_1 line. Figure 1 is a comparison of the R_1 line in pink ruby, the 7010A line in red ruby, and the R_1 line in red ruby, all under similar conditions of excitation at 77°K. The decay curves at low excitation are all similar in appearance to Fig. 1(c), and in two cases the shapes change progressively with increasing excitation until they reach the curves shown in Fig. 1(a) and Fig. 1(b). A plot of the areas under the fluorescence curves of R_1 and 7010 A as a function of flash energy is shown in Fig. 2.

Our interpretation of the data is as follows: It is apparent that the R_1 line in red ruby does not exhibit stimulated emission. This follows from the fact that the absorbed flash energy is not sufficient to overcome the ground-state population, and is corroborated by the approximately linear relationship between R_1 and flash energy in Fig. 2. (The increased linewidth in red ruby would not change conditions enough to prevent stimulated emission from occurring.) The behavior of the 7010A line (and 7040A line) is a little more complicated. The fact that it exhibits enhanced emission while the R_1 line in this sample does not is a result of the relatively unpopulated terminal state for the 7010A transition (negligible optical absorption for this wavelength at 77°K). Since the number of atoms radiating 7010A light saturates at intermediate energy (Fig. 2), it appears that the total number of equivalent ion pairs which have this line in their spectrum is excited at this energy. This number

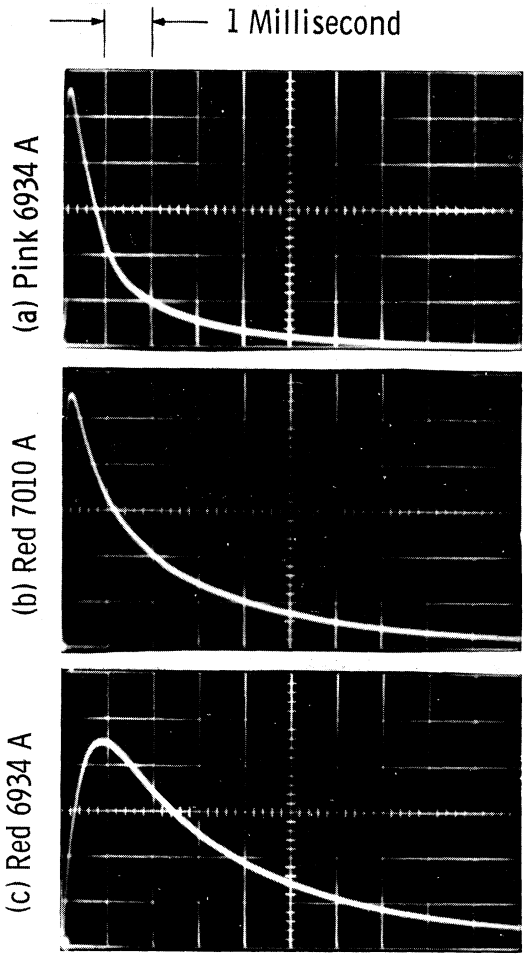

FIG. 1. Fluorescence decay curves of pink and red ruby taken at 77°K under approximately the same high excitation conditions.

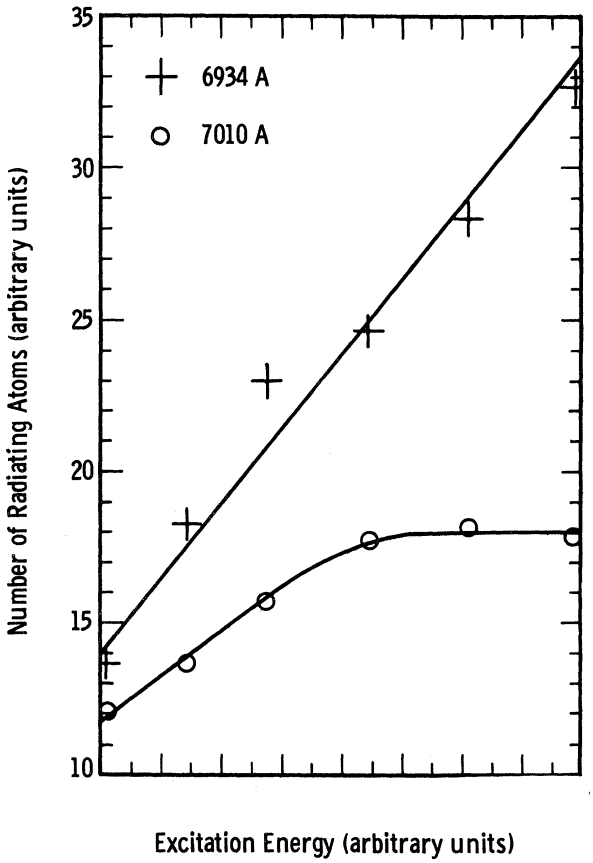

FIG. 2. A plot of area under fluorescence decay curves for R_1 and 7010 A in the same red ruby at 77°K as a function of excitation flash energy.

is not sufficiently greater than the threshold number needed for stimulated emission to exhibit the degree of lifetime shortening observed in pink ruby [compare Fig. 1(a) and Fig. 1(b)].

These experimental results also yield information on the excitation processes of the coupled ions. Since more than half of the single ions are in the ground state even at high excitation, direct excitation via the broad absorption bands is not likely for those ion pairs near the center of the rod. On the other hand, thermal transfer of excitation between single ions and paired ions is ruled out by the saturation behavior of the 7010A line. One possibility which seems promising is radiative excitation by trapped 6934A light. As a preliminary check we have observed absorption lines in the red sample which overlap the 6934A line and have also observed the 7010A fluorescence when the red rod is excited by a pulse of 6934A light from a pink sample. Further experiments to clarify the nature of the excitation processes of exchange-coupled ions are in progress.

[1]I. Wieder, Proceedings Ann Arbor Conference on Optical Pumping, June, 1959 (unpublished).
[2]A. L. Schawlow, Proceedings of the Quantum Electronics Conference, Shawanga, New York, September, 1959 [Quantum Electronics, edited by C. H. Townes (Columbia University Press, New York, 1960)].
[3]I. Wieder, Phys. Rev. Letters 3, 468 (1959).
[4]S. Geschwind, R. J. Collins, and A. L. Schawlow, Phys. Rev. Letters 3, 545 (1959).
[5]T. H. Maiman, Phys. Rev. Letters 4, 564 (1960).
[6]T. H. Maiman, Nature 187, 493 (1960); British Communications and Electronics 7, 674 (1960).
[7]R. J. Collins et al., Phys. Rev. Letters 5, 303 (1960).
[8]A. L. Schawlow, D. L. Wood, and A. M. Clogston, Phys. Rev. Letters 3, 271 (1959).
[9]A. L. Schawlow and C. H. Townes, Phys. Rev. 112, 1940 (1958).

SIMULTANEOUS OPTICAL MASER ACTION IN TWO RUBY SATELLITE LINES

A. L. Schawlow and G. E. Devlin
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received December 19, 1960)

We have obtained pulsed maser oscillations simultaneously at wavelengths of 7009 A and 7041 A in concentrated ruby. The lines involved are two of the strongest of the satellite or neighbor lines,[1] which have been known and studied for many years but are not yet fully analyzed.[2-5] It is known[5] that these lines disappear at low concentrations, but appear strongly in emission at chromium concentrations of the order of 0.5%. In absorption, they are weak and disappear entirely at low temperatures.

Since these lines terminate on states which are far enough above the ground states to be empty at low temperatures and since their upper states can be populated easily by white light illumination, it was proposed that the strongest of them be used for an optical maser.[6] A very low estimate of the fluorescence efficiency (0.1%) was used, but with the advantage of an empty lower state, pulsed operation seemed possible at low

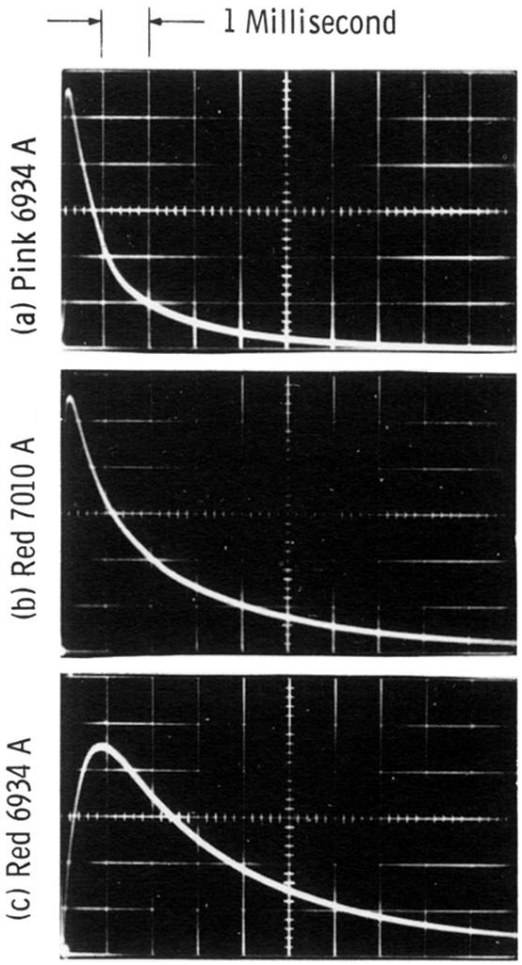

FIG. 1. Fluorescence decay curves of pink and red ruby taken at 77°K under approximately the same high excitation conditions.