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SELF-ABSORPTION AND TRAPPING OF SHARP-LINE RESONANCE RADIATION IN RUBY

F. Varsanyi, D. L. Wood, and A. L. Schawlow
Bell Telephone Laboratories, Murray Hill, New Jersey
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As an aid in planning a microwave-optical double resonance experiment,[1,2] we have examined by high-resolution optical spectroscopy the details of the sharp-line fluorescence of ruby (Cr^{3+} in Al_2O_3).

The only transition which appears in the fluorescence of dilute ruby at 4°K is from $^2E(\bar{E})$ to $^4A_2$ and appears at 6934 A. Selection rules for this transition have been calculated,[3] and partially verified for absorption.[4] The Zeeman levels to be discussed here are those occurring for a magnetic field parallel to the symmetry axis and are shown in Fig. 1.

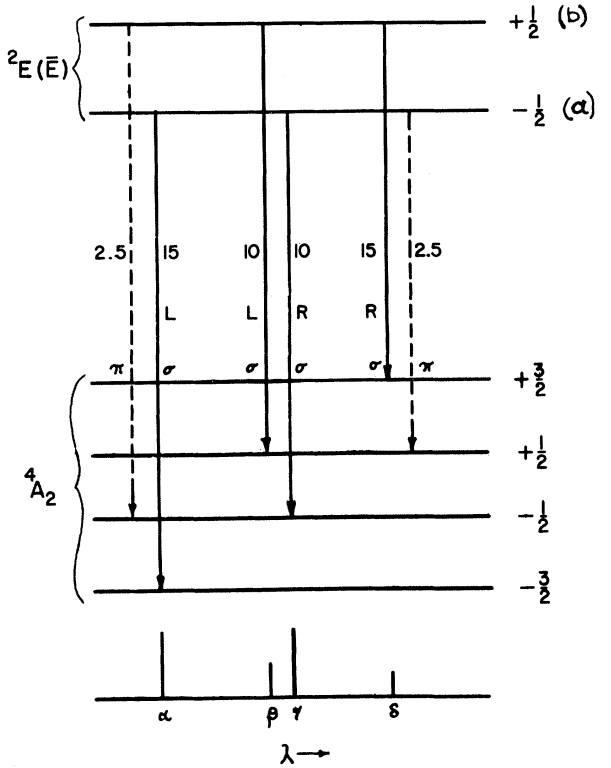

FIG. 1. Energy levels and transition probabilities for Zeeman levels involved in the resonance lines of ruby. $H \| c$ axis.

Figure 2(a) gives the predicted and observed intensities for a dilute ruby containing about 10^{-6} Cr per Al, and showing no color. For the theoretical patterns complete thermalization among the Zeeman levels is assumed in both ground and excited states. The figure shows that the selection rules predicting a 3:2 ratio of components α to γ and of δ to β are confirmed. Moreover, use of the α to δ and γ to β ratios shows that the population of atoms in the upper Zeeman level of the excited state is only slightly above that of a Boltzmann distribution. The effective spin temperature is 1.77°K. If it is assumed (although this is doubtful) that the excita-

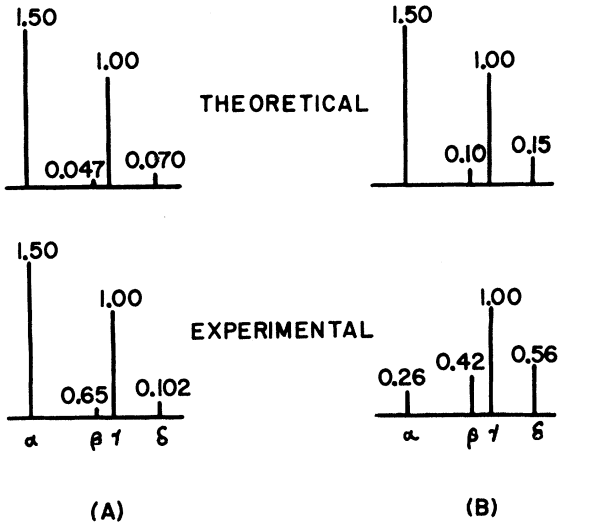

FIG. 2. (a) Theoretical and observed intensity ratio of Zeeman components in very dilute ruby sample (white sapphire). $H = 30\,000$ gauss; $T = 1.58$°K. (b) Theoretical and observed intensity ratio of Zeeman components in ruby containing 0.05% Cr_2O_3. $H = 30\,000$ gauss; $T = 2.1$°K.

tion process populates the two levels equally, and that the actual population is obtained by a balance between spin-lattice relaxation and radiative decay, the spin-lattice relaxation time is about 2×10^{-4} second. Also the crystal may have been heated above the helium bath temperature by the exciting radiation, so the actual relaxation time may be somewhat less. In any case, it seems that there is nearly complete thermalization between the Zeeman levels of the excited state during its relatively long lifetime of several milliseconds.

The great disparity between calculated and observed intensities for a ruby containing 0.05% Cr_2O_3 by weight is shown in Fig. 2(b). This disparity is explained by self-absorption which differs from one component to the next because of the Boltzmann population distribution of the ground-state levels. Thus component α terminates on the ground level ($m = -3/2$), which is populated by 85% of the atoms and is strongly absorbed. At the other extreme the component δ terminates on the $+3/2$ level, which is 8.3 cm^{-1} above it, and contains only 0.3% of the atoms, so that δ is much less absorbed. While self-absorption is often encountered in gaseous luminescence, it is seldom possible for a gas to be cold enough to have appreciably different populations in individual Zeeman levels.

Quantitatively, we find that an absorption coefficient of about 25 cm^{-1} for the α component produces satisfactory agreement with the observed intensity ratio of γ to α in the 0.05% ruby spectrum. A direct absorption measurement at 77°K, where only half of the atoms are in the $\pm3/2$ state and line widths have increased from 0.25 to 0.32 cm^{-1}, gave $k = 13$ cm^{-1} in good agreement. The radiative lifetime of the excited state was also measured in the very dilute sample, and in finely divided and dispersed 0.05% ruby, and was found to be 4.3 ± 0.3 milliseconds. This gives an oscillator strength $f = 7.5\times10^{-7}$ for the same transition, which then gives $k = 11$ cm^{-1} for 0.05% ruby. This satisfactory agreement indicates that the observed lifetime is indeed predominantly radiative.

It has been possible in the 0.05% ruby to observe for the first time in a solid the trapping of resonance radiation. At 77°K the radiative lifetime is found to vary smoothly from 4.3×10^{-3} second for a fine, dispersed powder to 15×10^{-3} second for a whole boule, a very long lifetime. The observation of trapping confirms the predominantly radiative character of the decay of the excited state, and the strong self-absorption of the resonance radiation. The ratio of lifetimes with and without trapping is only about 4, showing that other modes of decay such as radiation in vibrational sidebands and nonradiative processes also occur.

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OPTICAL DETECTION OF PARAMAGNETIC RESONANCE IN AN EXCITED STATE OF Cr^{3+} IN Al_2O_3

S. Geschwind, R. J. Collins, and A. L. Schawlow
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received November 12, 1959)

We have observed paramagnetic resonance in an excited metastable state of Cr^{3+} in Al_2O_3 by a method of optical detection which should have wide applicability to the general problem of studying paramagnetic resonance in excited states of ions in solids. This method makes use of the selective reabsorption in the ground-state Zeeman levels of the fluorescent light from the excited states in solids at very low temperatures.[1] Referring to Fig. 1 of the preceding Letter,[1] at 1.6°K, the lower of the two Zeeman levels of the excited state $\bar{E}(^2E)$ will be much more heavily populated. Let us first assume there is no reabsorption so that the theoretical