served. In the present experiments two peaks are found separated by only about 40 gauss so that it seems unlikely that the broadening is extreme. The tail of the resonance extends to about 400 gauss with no additional structure. This may be related to the characteristic of the magnetic method that even unbroadened lines possess apparent magnetic widths which are proportional to the applied magnetic field.

Although the interpretation is admittedly incomplete, the extreme sharpness of the resonance is apparent. In further study, involving the development of a Doppler shift drive, we hope to measure a number of the energy shifts and level splittings mentioned in previous paragraphs.

We wish to thank S. D. Stoddard and R. E. Cowan for preparation of the ZnO source buttons and for compacting the enriched ZnO absorber. The generous cooperation of the cyclotron group is gratefully acknowledged. W. E. Keller and J. G. Dash each contributed a number of ideas to the experiment.

---

†Work done under the auspices of the U. S. Atomic Energy Commission.

[1]R. L. Mössbauer, Z. Physik 151, 124 (1958); Naturwissenschaften 45, 538 (1958); Z. Naturforsch. 14a, 211 (1959).

[2]D. E. Nagle, P. P. Craig, and W. E. Keller, Nature (to be published).

[3]R. V. Pound and G. A. Rebka, Phys. Rev. Letters 4, 397 (1960).

[4]R. V. Pound and G. A. Rebka, Phys. Rev. Letters 4, 337 (1960); B. D. Josephson, Phys. Rev. Letters 4, 341 (1960).

[5]O. C. Kistner and A. W. Sunyar, Phys. Rev. Letters 4, 412 (1960).

[6]G. Heiland, E. Mollwo, and F. Stöckmann, Solid-State Physics, edited by F. Seitz and D. Turnbull (Academic Press, New York, 1959), Vol. 8, p. 191.

[7]H. Kopfermann, Kernmomente (Akademische Verlagsgesellschaft, Frankfurt am Main, 1956).

OPTICAL AND MICROWAVE-OPTICAL EXPERIMENTS IN RUBY

T. H. Maiman
Hughes Research Laboratories, Malibu, California
(Received April 22, 1960)

Several recent papers[1-4] have reported optical and microwave-optical measurements in ruby (Cr^{+++} in Al_2O_3). We wish to report here some new experiments concerning the fluorescent relaxation processes in this crystal. Reported here also are the first observations of ground-state population changes in ruby due to optical excitation and the detection of optical absorption between two excited states in this crystal.

The predominant processes which ensue in a fluorescent material when it is irradiated at an appropriate wavelength are shown in Fig. 1. W_{13} is the induced transition probability per unit time due to an exciting radiation and the S_{mn} are decay rates which include both radiative and nonradiative processes. In this crystal S_{21} is easily obtained from the decay rate of the fluorescent level (^2E) after an exciting source is turned off. The lifetime for this process is about 5 msec. Varsanyi, Wood, and Schawlow[2] have further demonstrated that this lifetime is almost entirely due to spontaneous emission, i.e., S_{21} is approximately the Einstein A coefficient A_{21}.

An approximate value for the rate S_{32} was obtained in the following way. A crystal of ruby was irradiated with 5600A radiation causing absorption into the lower band ($^4A_2 \rightarrow {}^4F_2$). The sample used was a one-centimeter cube cut from a boule of standard pink ruby supplied by the Linde

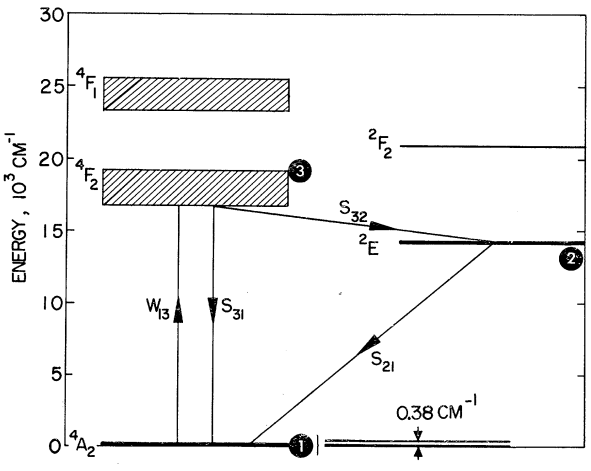

FIG. 1. Pertinent features of the energy level diagram in ruby.

Company, with a concentration of approximately 0.05 weight percent of Cr_2O_3 to Al_2O_3. Two components of radiation re-emitted from the crystal were observed in a direction perpendicular to the exciting beam: that due to re-emission of the incoming radiation (spontaneous decay from $^4F_2$) and fluorescence (spontaneous decay from ^2E). The intensity of the first component is proportional to $h\nu_{31}N_3A_{31}$, where A_{31} is the A coefficient for $^4F_2 \rightarrow {}^4A_2$ and is calculated from measurements of absorption coefficient and linewidth for this transition ($A_{31} \sim 3\times10^5$/sec). Similarly the fluorescent intensity is proportional to $h\nu_{21}N_2A_{21}$. By a measurement of the ratio of these two components and the use of an auxiliary condition applicable to steady-state conditions $N_2S_{21} = N_3S_{32}$ and also the use of the approximation $S_{21} = A_{21}$, we find $S_{32} \sim 2\times10^7$/sec.

A measurement of fluorescent quantum efficiency, i.e., the number of fluorescent quanta emitted compared to the number absorbed by the crystal from the exciting beam, yielded a value near unity. This result reconfirms the evidence that the life of level 2 is near radiative and also implies that $S_{32} >> S_{31}$. The experiment was not accurate enough to yield a precise value but does indicate that the nonradiative process $(S_{31} - A_{31}) < 4\times10^6$/sec.

Calculations utilizing the previous results indicated that population changes in the ground state of ruby due to optical excitation would be easily observed. This conclusion was verified in the following experiments. A ruby crystal was mounted between parallel silvered plates to form a microwave cavity resonant at the ground-state zero-field splitting (11.3 kMc/sec). About half the cavity losses were due to magnetic absorption as evidenced by an increase in cavity Q when a small magnet was brought near the ruby. The reflection coefficient of the cavity was monitored on an oscilloscope while a short pulse (200 μsec) of light from a flash tube irradiated the crystal. The magnitude of the microwave magnetic absorption was observed to decrease abruptly and then return to equilibrium with a time constant of about 5 msec (see Fig. 2). We attribute this effect to temporary depletion of this ground state population with subsequent decay back from the fluorescent level. The experiment was performed at room temperature where the thermal relaxation times in the ground state of ruby are the order of a microsecond; in the time scale of the experiment, therefore, Boltzmann equilibrium in these levels is maintained.

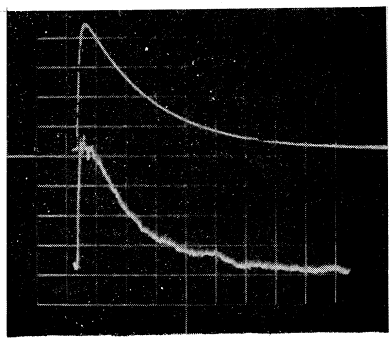

FIG. 2. The upper trace was obtained from a photomultiplier monitoring the R-line fluorescence ($^2E \rightarrow {}^4A_2$) in ruby. The lower trace is a recording of the reflection coefficient of a microwave ruby cavity during the same time interval. The time scale is 2 milliseconds per division. Both recordings followed an intense light pulse which excited the crystal via $^4A_2 \rightarrow {}^4F_2$.

A repetition of the above experiment at liquid helium temperatures is being planned. At this temperature we would expect to be able to observe directly any preferential depopulation of the ground sublevels due to polarized light and also any preferential repopulation of these levels since the thermal relaxations times would then be 30-100 msec.

To verify further the depletion of ground-state population observed in the previous experiment an independent measurement was made. A beam of monochromatic light of wavelength 4100 A was transmitted through a ruby crystal and partially absorbed due to the transition $^4A_2 \rightarrow {}^4F_1$. When the intense pulse of radiation at 5600 A was turned on, the 4100A radiation passing through the crystal abruptly increased and subsequently decayed in about 5 msec just as the microwave signal in the previous experiment. This result was expected since the temporary reduction in ground-state population caused the crystal to become more transparent to the 4100A radiation until the fluorescent level decayed to normal. In both experiments a population change of about 3% was estimated.

An unexpected result was observed when the probe wavelength was changed from 4100 A to 3600 A. In this case a decrease in light intensity emerging from the crystal was observed. This implies that the crystal became more absorbing even though the ground-state population was decreased. We can explain this last effect when it is realized that 3600A radiation can cause transitions from the fluorescent level (^2E) to a high-

lying charge transfer band (not shown in the figure). Consequently, we conclude that we were observing transitions between two excited optical states. The fact that the abruptly increased 3600A absorption also decayed with a 5-msec time constant is consistent with and strengthens the above conclusion.

The author gratefully acknowledges helpful discussions with G. Birnbaum, R. W. Hellwarth, L. C. Levitt, and R. A. Satten and is particularly indebted to I. J. D'Haenens for technical assistance in performing the experiments.

[1]I. Wieder, Phys. Rev. Letters 3, 468 (1959).
[2]F. Varsanyi, D. L. Wood, and A. L. Schawlow, Phys. Rev. Letters 3, 544 (1959).
[3]S. Geschwind, R. J. Collins, and A. L. Schawlow, Phys. Rev. Letters 3, 545 (1959).
[4]J. Brossel, S. Geschwind, and A. L. Schawlow, Phys. Rev. Letters 3, 548 (1959).

NONADIABATIC THEORY OF THE SCATTERING OF ELECTRONS FROM HYDROGEN

A. Temkin*
Theoretical Division, Goddard Space Flight Center, National Aeronautics and Space Administration
Washington, D. C.
(Received April 6, 1960)

Rosenberg, Spruch, and O'Malley[1] (RSO) have recently calculated rigorous upper bounds, $a_S < 6.23$ and $a_t < 1.92$, for the singlet and triplet scattering lengths in the scattering of electrons from atomic hydrogen,[2] under the assumption that the H ion has only one bound state, with singlet spin. The fact that these upper bounds are 15% below the results of most of the elaborate calculations thus far performed[3] indicates that we are far from an adequate quantitative theory of this three-body problem.

Those calculations which yield scattering lengths close to these upper bounds make use of the adiabatic hypothesis, according to which the distortion of the atom follows the instantaneous position of the scattered electrons.[4,5] Objections to the validity of the adiabatic hypothesis have recently been raised.[6] We shall outline in this note a rigorous (and hence nonadiabatic) series for the phase shift. The proposed series serves two purposes: First, because it is rapidly convergent it provides a method for calculating phase shifts reliably. This is so because methods can be given for accurately approximating the higher order terms. Second, the successive terms of the projected series can be put in a one-to-one correspondence with an adiabatic series which has been derived for dealing with this problem.[4,7] It can be demonstrated that the main contribution to the nonadiabatic terms comes from the functions which enter the adiabatic development. This provides a justification for the qualitative validity of the adiabatic theory and an explanation of the fact that the adiabatic calculations give scattering lengths in better accord with the RSO upper bounds.

The method is an extension of a method used originally by Breit and more recently by Luke, Meyerott, and Clendenin[8] for calculating the energy of some excited states of 2-electron atoms and ions. One makes a decomposition of the zero angular momentum wave function $\Psi(r_1 r_2 \theta_{12})$,

$$\Psi(r_1 r_2 \theta_{12}) = \frac{1}{r_1 r_2} \sum_{l=0}^{\infty} (2l+1)^{1/2} \Phi_l(r_1 r_2) P_l(\cos\theta_{12}), \qquad (1)$$

and reduces the Schrödinger equation to the infinite set of coupled equations (for $r_1 > r_2$)

$$\left\{\left[\sum_{i=1}^{2} \frac{\partial^2}{\partial r_i^2} - \frac{l(l+1)}{r_i^2} + \frac{2}{r_i}\right] - 1 + k^2 - (2l+1)\sum_n \frac{r_2^n}{r_1^{n+1}} \int_0^{\pi} P_l^2 P_n \sin\theta_{12} d\theta_{12}\right\} \Phi_l$$

$$= \sum_{m(\neq l)} \left\{[(2l+1)(2m+1)]^{1/2} \sum_n \left(\int_0^{\pi} P_l P_m P_n \sin\theta_{12} d\theta_{12} \right) \frac{r_2^n}{r_1^{n+1}} \right\} \Phi_m. \qquad (2)$$

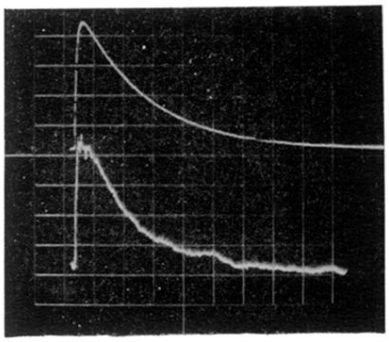

FIG. 2. The upper trace was obtained from a photomultiplier monitoring the R-line fluorescence ($^2E \rightarrow {}^4A_2$) in ruby. The lower trace is a recording of the reflection coefficient of a microwave ruby cavity during the same time interval. The time scale is 2 milliseconds per division. Both recordings followed an intense light pulse which excited the crystal via $^4A_2 \rightarrow {}^4F_2$.