

The internuclear distance between dipole partners expands as the temperature is increased. By approximating $C(T_n)$ as a linear function between adjacent ambients, $\Delta D_a(T_n)$, the change that occurs in this distance as the ambient is increased from its lowest value of 78°K to T_n can be expressed as follows:

$$\Delta D_a(T_n) = (\alpha_a/2) \sum_{m=n}^4 [C(T_m) + C(T_{m+1})][T_m - T_{m+1}],$$

for $n=0, 1, 2, 3, 4$ and $T_{m+1} < T_m$. (B9)

The results of Eq. (B9) show that the change in the internuclear distance D_a is so small over the entire temperature range of the study that it can be regarded as a constant. Using the value of D_a computed in Appendix A, the moment of an elementary cadmium sulfide dipole at temperature T_n can be expressed as follows:

$$M(T_n) = D_a k_a(T_n). \quad (\text{B10})$$

The values that were obtained for $k_a(T_n)$, $k_a(T_n)$, $\Delta D_a(T_n)$, and $M(T_n)$ are indicated in Table II.

Optically Induced Magnetization in Ruby*

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The magnetization of ruby in a magnetic field at 300°K is changed when the ruby is optically pumped with linearly polarized radiation from a Q-switched ruby laser. With a magnetic field parallel to the trigonal axis, laser light polarized parallel to this axis induces transitions from the spin $\pm \frac{1}{2}$ levels of the 4A_2 ground state to the levels of the $\bar{E}({}^2E)$ excited state. The change in M_z is linearly proportional to H except near the anticrossing points of the 4A_2 spin levels at 2.07 and 4.14 kG. At these field strengths, there is an enhancement of the magnetization caused by state mixing of the 4A_2 wave functions. When H has a small component perpendicular to the z axis, a magnetization is detected parallel to this component in the vicinity of 4.14 kG. The effect requires a long ground-state relaxation time. In a separate experiment, the relaxation of M_z was found to vary from 0.13 μsec in zero magnetic field to a constant value of 0.57 μsec for fields above 60 G.

1. INTRODUCTION

LINEARLY polarized radiation from a Q-switched ruby laser has been used to optically pump a ruby crystal. When the crystal is in a magnetic field, the changes in population of the energy levels 4A_2 , $\bar{E}$, and $2\bar{A}$ of the chromium ions cause a change in the magnetization. Expressions for the field dependence of the magnetization are obtained in Sec. 2 for the case that both the laser polarization and the magnetic field are parallel to the optic axis. The experimental results discussed in Sec. 3 show the solid-state analog of anticrossing-state mixing experiments in gases and indicate that the relaxation time of the ground state is long compared to the laser pulse length. Quantitative measurements of the relaxation rate in small magnetic fields, using circularly polarized laser radiation, are reported in Sec. 4.

2. THEORY

The splitting of the 4A_2 , $\bar{E}$, and $2\bar{A}$ energy levels of ruby in a magnetic field parallel to the optic axis is shown Fig. 1. The ground state, with an effective spin of $\frac{3}{2}$, is described by the spin Hamiltonian¹

$$\mathcal{H} = g_{11}({}^4A_2)\beta H \cos\theta S_z - D[S_z^2 - \frac{1}{3}S(S+1)] + \frac{1}{2}g_{\perp}({}^4A_2)\beta H \sin\theta[e^{-i\varphi}S_+ + e^{i\varphi}S_-]. \quad (1)$$

The g values are given in Table I, and the zero field splitting $2D$ is 0.3824 cm^{-1} . The magnetic field makes

TABLE I. The g values of Cr^{3+} in Al_2O_3 .

Energy level	g_{11}	$g_{\perp}$
4A_2	1.9840 ± 0.0006^a	1.9867 ± 0.0006^a
$\bar{E}({}^2E)$	-2.445 ± 0.001^b	$< 0.2^c$
$2\bar{A}({}^2E)$	1.48 ± 0.08^d	...

^a Reference 1. ^b Reference 4. ^c References 4 and 5. ^d Reference 6.

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¹ A. A. Manenkov and A. M. Prokhorov, Zh. Eksperim. i Teor. Fiz. 28, 762 (1955) [English transl.: Soviet Phys.—JETP 1, 611 (1955)]; J. E. Geusic, Phys. Rev. 102, 1252 (1956).

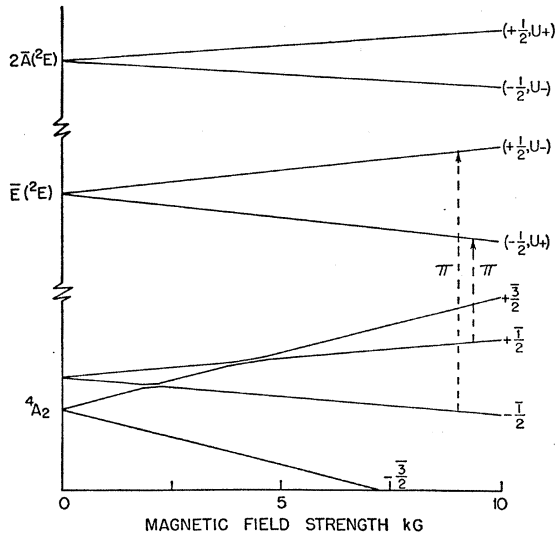


FIG. 1. Zeeman splitting of the 4A_2 , $\bar{E}$, and $2\bar{A}$ states of Cr^{3+} in ruby with H parallel to the optic axis. The transitions between 4A_2 and $\bar{E}$ for light polarized parallel the optic axis are shown. Values of $\bar{n}$ are shown with bars.

an angle θ with the optic axis, and an azimuthal angle φ with the x axis. If θ is zero, the Hamiltonian is diagonal, and the energy levels are linear functions of the field. The spin $+\frac{3}{2}$ level then crosses the spin $-\frac{1}{2}$ and $+\frac{1}{2}$ levels at 2.07 and 4.14 kG, respectively. If there is a perturbation with off-diagonal matrix elements, the levels will not intersect, but will remain continuous functions of the field. The wave function associated with each level then is a linear combination of the wave functions of the two levels in the absence of the perturbation. At the field corresponding to the closest approach of the energy levels, the wave functions consist of equal amplitudes of the unperturbed wave functions. It is convenient to adopt the notation of Schulz-DuBois² in identifying the energy levels by a quantum number $\bar{n}$ ranging from $-\frac{3}{2}$ to $+\frac{3}{2}$ in order of increasing energy as shown in Fig. 1. The number $\bar{n}$ coincides with the quantum number n of the Zeeman Hamiltonian in the high-field limit.

We shall consider the third term of Eq. (1) as a perturbation and obtain the field dependence of the energy levels and the wave functions near 4.14 kG. Since $\Delta S = 1$, the degeneracy is removed in first order. The solution of the secular determinant yields the energy levels

$$W = g_{11}({}^4A_2)\beta H \cos\theta \pm \frac{1}{2}[\Delta^2 + |2V|^2]^{1/2}, \quad (2)$$

where $\Delta = g_{11}({}^4A_2)\beta H \cos\theta - 2D$ is the separation of the unperturbed levels, and the absolute value of the off-diagonal matrix element of the perturbation is $|V| = \frac{1}{2}\sqrt{3}g_1({}^4A_2)\beta H \sin\theta$. The positive sign before the radical sign corresponds to the $\bar{n} = +\frac{3}{2}$ level. The eigen-

functions are

$$|+\frac{3}{2}\rangle_{\bar{n}} = a|+\frac{3}{2}\rangle + be^{+i\varphi}|+\frac{1}{2}\rangle, \quad (3a)$$

$$|+\frac{1}{2}\rangle_{\bar{n}} = -be^{-i\varphi}|+\frac{3}{2}\rangle + a|+\frac{1}{2}\rangle. \quad (3b)$$

The relative phase, φ , is determined by the direction of the perturbing magnetic field. The real coefficients a and b are

$$a = |2V| \{ |2V|^2 + [(|2V|^2 + \Delta^2)^{1/2} - \Delta]^2 \}^{-1/2}, \quad (4a)$$

$$b = [(|2V|^2 + \Delta^2)^{1/2} - \Delta] \times \{ |2V|^2 + [(|2V|^2 + \Delta^2)^{1/2} - \Delta]^2 \}^{-1/2}. \quad (4b)$$

The $\bar{E}$ and $2\bar{A}$ states have an effective spin of $\frac{1}{2}$.³ The g values are given in Table I.⁴⁻⁶ The negative value of $g_{11}(\bar{E})$ has been discussed by Clogston.⁵ In contrast with spin systems having a positive g value, the effective $+\frac{1}{2}$ spin state has a lower energy than the $-\frac{1}{2}$ state in a magnetic field.⁷

McCumber and Sturge⁸ have measured the positions of the optical absorption lines, and their widths, as functions of temperature. Between 77 and 400°K, $\bar{E}$ and $2\bar{A}$ are thermally broadened by a two-phonon Raman process. The population of the two levels, after being disturbed, will reach thermal equilibrium in a time short compared to the duration of the laser pulse length. Both absorption lines have a Lorentzian shape.⁹ The linewidth of the 4A_2 to $2\bar{E}$ transition is 11 cm^{-1} at 300°K.¹⁰

The experimental evidence of Sec. 4 indicates that the relaxation time between the Zeeman levels of the ground state is much longer than the pulse length. Since the radiative lifetime of the excited state is approximately 3 msec^{11,12} the chromium ions excited by laser radiation remain in the excited state for a period long compared to the pulse length.

Consider now the high field region ($H > 5 \text{ kG}$), where state mixing is absent. As shown in Fig. 1, radiation polarized parallel to the optic axis induces transitions from the $\bar{n} = \pm\frac{1}{2}$ levels of 4A_2 to $\bar{E}$ with equal dipole strength.^{3,6} Owing to the higher population of the lower level, the absorption coefficient of this level will be slightly larger. It will be useful to separate this popula-

³ S. Sugano and Y. Tanabe, J. Phys. Soc. Japan 13, 880 (1958).

⁴ S. Geschwind, R. J. Collins, and A. L. Schawlow, Phys. Rev. Letters 3, 545 (1959).

⁵ A. M. Clogston, Phys. Rev. 118, 1229 (1960).

⁶ S. Sugano and I. Tsujikawa, J. Phys. Soc. Japan 13, 899 (1958).

⁷ M. H. L. Pryce, Phys. Rev. Letters 3, 375 (1959).

⁸ D. E. McCumber and M. D. Sturge, J. Appl. Phys. 34, 1682 (1963).

⁹ A. L. Schawlow, *Quantum Electronics, Proceedings of the Third International Congress*, edited by P. Grivet and N. Bloembergen (Columbia University Press, New York, 1964), p. 645.

¹⁰ A. L. Schawlow, *Advances in Quantum Electronics*, edited by J. R. Singer (Columbia University Press, New York, 1961), p. 50.

¹¹ T. H. Maiman, R. H. Hoskins, I. J. D'Haenens, C. K. Asawa, and V. Evtuhov, Phys. Rev. 123, 1151 (1961).

¹² G. H. Dieke and L. A. Hall, J. Chem. Phys. 27, 465 (1964).

² E. O. Schulz-DuBois, Bell System Tech. J. 38, 271 (1959).

tion dependence of the absorption coefficient,

$$\alpha(\pm\frac{1}{2}, \nu) = \alpha_0(\pm\frac{1}{2}, \nu) \exp[\mp(g_{11}(^4A_2)\beta H/2kT)], \quad (5)$$

where $\alpha_0(\bar{n}, \nu)$ is the absorption coefficient of each level in zero field. The $\alpha_0(\bar{n}, \nu)$ are shown in Fig. 2 for a field of 6 kG. Except at $\nu = \nu_0$ the absorption coefficient of one component exceeds the other.

The laser wavelength coincides with the center of the absorption line only if the two rubies have the same temperature. Measurements indicate, however, that the flash lamp heats the laser ruby by 10°K. Using a shift of 0.1 cm⁻¹/°K,^{8,13} the peak of the fluorescent line of the laser ruby will shift to longer wavelength by approximately 1 cm⁻¹ from the room temperature value before Q-switched laser action occurs. This is consistent with the wavelength shift observed during normal laser action with a similar laser head.¹⁴

When the absorption does not significantly deplete the laser intensity, the rate of depopulation per unit volume of the $\bar{n} = \pm\frac{1}{2}$ levels is

$$dN(\pm\frac{1}{2}, t)/dt = \alpha(\pm\frac{1}{2}, \nu_L)(\eta/h\nu_L)I(\nu_L, t). \quad (6)$$

Here η is the refractive index and $I(\nu_L, t)$ is the laser intensity at time t in vacuum. Integration of Eq. (6) yields the number of ions excited up to time t .

$$\Delta N(\pm\frac{1}{2}, t) = \alpha(\pm\frac{1}{2}, \nu_L)\mathfrak{N}(t), \quad (7)$$

where $\mathfrak{N}(t)$ is the number of laser photons which have passed through the crystal

$$\mathfrak{N}(t) = (\eta/h\nu_L) \int_0^t I(\nu_L, t') dt'. \quad (8)$$

The resulting magnetization parallel to the optic axis is

$$\begin{aligned} \Delta M_z^{(1)}(H, t) = & [g_{11}(\bar{E})^2 + g_{11}(2\bar{A})^2 - 2g_{11}(^4A_2)^2] \\ & \times \beta^2 H (8kT)^{-1} [\alpha_0(+\frac{1}{2}, \nu_L) + \alpha_0(-\frac{1}{2}, \nu_L)] \mathfrak{N}(t) \\ & + \frac{1}{2} g_{11}(^4A_2) \beta [\alpha_0(+\frac{1}{2}, \nu_L) - \alpha_0(-\frac{1}{2}, \nu_L)] \mathfrak{N}(t). \end{aligned} \quad (9)$$

It has been assumed that instantaneous thermal equilibrium is reached between the $2\bar{A}$ and $\bar{E}$ levels. Since they are separated by only 29 cm⁻¹, essentially one half of the excited ions are distributed over the spin doublet $2\bar{A}$ and one half over the spin doublet $\bar{E}$. The series expansion of the Boltzmann factor in Eq. (5) has also been used in the derivation of Eq. (9).

The magnetization of the excited state is nearly cancelled by the decrease in the ground-state magnetization resulting from the different initial populations of this state. The main contribution to the magnetization results from the second term of Eq. (9) which describes the preferential excitation as shown in Fig. 2. Using the values given above, it is approximately 100 times larger than the first term.

¹³ I. D. Abella and H. Z. Cummins, J. Appl. Phys. **32**, 1177 (1961).

¹⁴ T. P. Hughes, Nature **195**, 325 (1962); T. P. Hughes and K. M. Young, *ibid.* **196**, 332 (1962).

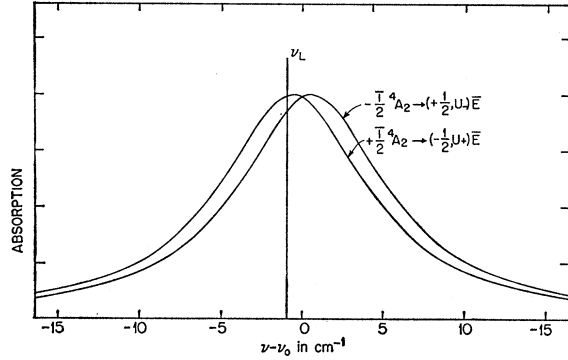


FIG. 2. The absorption coefficients $\alpha_0(+\frac{1}{2}, \nu)$ and $\alpha_0(-\frac{1}{2}, \nu)$ for a field of 6 kG. Values of $\bar{n}$ are shown with bars.

With the conditions shown in Fig. 2, the second term is linear in the field. Expansion of the Lorentzian absorption coefficients yields

$$\alpha_0(+\frac{1}{2}, \nu_L) - \alpha_0(-\frac{1}{2}, \nu_L) \approx 2\alpha_0(+\frac{1}{2}, \nu_0) [g(^4A_2) + |g(\bar{E})|] \times \beta H (\nu_0 - \nu_L) / hc(\Delta\nu)^2, \quad (10)$$

where $\alpha_0(+\frac{1}{2}, \nu_0)$ is the maximum absorption in zero field at the center frequency ν_0 . Similar expressions for the magnetization may also be found for the field regions between 0 to 2 kG and 2 to 4 kG where state mixing is negligible. Although the labels of the absorption coefficients are different, the absorption still occurs from the Zeeman spin $\pm\frac{1}{2}$ levels. The magnetization in these regions is also a linear function of the field.

Consider next the magnetization near 4.14 kG. From Eq. (3), the absorption coefficients of the two interacting levels are proportional to the square of the amplitude of the spin $+\frac{1}{2}$ Zeeman wave functions

$$\alpha(+\frac{3}{2}, \nu_L) = b^2 \alpha_0(+\frac{1}{2}, \nu_L) \exp\{-[W(+\frac{3}{2})/kT]\}, \quad (11a)$$

$$\alpha(+\frac{1}{2}, \nu_L) = a^2 \alpha_0(+\frac{1}{2}, \nu_L) \exp\{-[W(+\frac{1}{2})/kT]\}. \quad (11b)$$

Here $\alpha_0(+\frac{1}{2}, \nu_L)$ is the absorption coefficient of the $+\frac{1}{2}$ level which would be obtained in the absence of state mixing. Taking $W(+\frac{3}{2}) = W(+\frac{1}{2})$, the magnetization parallel to the optic axis is

$$\begin{aligned} \Delta M_z(H, t) = & \Delta M_z^{(1)}(H, t) \\ & + 2\beta g_{11}(^4A_2) a^2 b^2 \alpha_0(+\frac{1}{2}, \nu_L) \mathfrak{N}(t). \end{aligned} \quad (12)$$

The first term is the magnetization which would occur in the absence of state mixing [Eq. (9)]. The additional magnetization has its maximum value $\frac{1}{2}\beta g_{11}(^4A_2) \times \alpha_0(+\frac{1}{2}, \nu_L) \mathfrak{N}(t)$ when Δ is zero and $a = b$. It is reduced to one half its peak value when $\Delta = \pm|2V|$, corresponding to a width between the half-amplitude points of $4|V|/g_{11}(^4A_2)\beta$ G. This term is the result of the change in the magnetic moment of the $\bar{n} = +\frac{1}{2}$ level and $\bar{n} = +\frac{3}{2}$ level. The total excitation rate from these two levels remains equal to the excitation rate from the $+\frac{1}{2}$ level away from the crossing region.

When state mixing is absent, a magnetization perpen-

dicular to the axis does not exist. The perturbation which resulted in the wave functions of Eq. (3) creates perpendicular components of magnetization near 4.14 kG,

$$\Delta M_x(H, t) = \sqrt{3}g_1(^4A_2)\beta ab(b^2 - a^2) \times \alpha_0(+\frac{1}{2}, \nu_L)\mathcal{N}(t) \cos \varphi, \quad (13a)$$

$$\Delta M_y(H, t) = \sqrt{3}g_1(^4A_2)\beta ab(b^2 - a^2) \times \alpha_0(+\frac{1}{2}, \nu_L)\mathcal{N}(t) \sin \varphi, \quad (13b)$$

ΔM_x and ΔM_y have their extrema when $\Delta = \pm |2V|$. The magnetization is parallel to the direction of the perturbing field for $\Delta < 0$ ($H < 4.14$ kG), and antiparallel when $\Delta > 0$. The perturbing field does not, however, determine the peak amplitude.

The situation near 2.07 kG is somewhat different. Since ΔS is two, a magnetic perturbation couples the spin $+\frac{3}{2}$ and $-\frac{1}{2}$ wave functions in second order

$$|+\frac{1}{2}\rangle_{\bar{n}} = c|+\frac{3}{2}\rangle + d|-\frac{1}{2}\rangle, \quad (14a)$$

$$|-\frac{1}{2}\rangle_{\bar{n}} = -d^*|+\frac{3}{2}\rangle + c|-\frac{1}{2}\rangle, \quad (14b)$$

where c and d are functions of the field. The magnetization parallel to the optic axis is

$$\Delta M_z(H, t) = \Delta M_z^{(1)}(H, t) + 4\beta g_{11}(^4A_2)c^2d^2\alpha_0(-\frac{1}{2}, \nu_L)\mathcal{N}(t), \quad (15)$$

where $\alpha_0(-\frac{1}{2}, \nu_L)$ is the absorption coefficient of the $\bar{n} = -\frac{1}{2}$ level in the absence of state mixing. Magnetization components perpendicular to the optic axis do not occur in lowest order.

The perturbation due to a field component perpendicular to the optic axis has been used as an example. Other perturbations, such as the dipolar interaction between chromium ions, and with the Al^{27} nuclei, will also prevent the levels from intersecting. An additional magnetization parallel to the optic axis would thus occur even in the absence of a field component perpendicular to this axis. However, ΔM_x and ΔM_y do not exist unless the perturbation has a preferred direction perpendicular to the axis.

The effects due to state mixing are quite analogous to level anticrossing effects observed in resonance fluorescence.¹⁵ In this case the Zeeman levels of the excited state are tuned by an external magnetic field. The wave functions of the states are coupled by off-diagonal matrix elements of the magnetic hyperfine interaction, which prevents the levels from intersecting. The angular distribution of resonance radiation in the presence of state mixing differs from the radiation pattern without state mixing, but the total resonance fluorescence remains unchanged. In contrast to the present experiment, coherence effects are also observed when the levels cross.¹⁶ The resonance fluorescence experiments

have been used to accurately determine the zero field splittings of the excited states from the field required for crossing.¹⁶ It follows from the above that the quantity $D/g_{11}(^4A_2)$ may be similarly obtained.

3. EXPERIMENTAL RESULTS

The experimental configuration is shown in Fig. 3. Radiation from a Q -switched ruby laser, 0.2 J in 25×10^{-9} sec, was polarized parallel to the direction of the magnetic field by an air-spaced Glan prism. A 50-cm focal-length lens slightly reduced the diameter of the beam at the position of the sample. A small fraction of the beam which was deflected by the glass slide to the photomultiplier served as a monitor of the laser intensity.

The ruby sample was a Y -cut, 0.635-mm-diam laser rod with 0.04% Cr_2O_3 concentration. The birefringence of the ruby was used to align the optic z axis parallel to the direction of the field.

A transverse pickup coil, consisting of seven turns wound on a phenolic cylinder, was used to detect the time rate of change of the magnetization. The orientation of the coil could be adjusted to detect the magnetic signal parallel to either the z or x axis. In order to eliminate the spurious signals which were detected when the laser radiation was incident on parts of the coil wire, the coil shielded with opaque electrical tape.

The signals obtained with the plane of the pickup coil oriented perpendicular to the optic axis is shown in Fig. 4. In regions where state mixing of the wave functions is negligible, the signal is a linear function of the field, and is directed parallel to the field. The additional magnetic signals occurring at the level anticrossings of the ground state are clearly evident. Equations (9) and (12) indicate that the excess signal at 2.07 kG should have twice the maximum amplitude of the signal at 4.14 kG. The narrow width of the 2.07-kG signal prevented an accurate determination of its maximum amplitude so that a numerical comparison of the peak amplitudes could not be made.

The difference in width at half-amplitude of the two peaks is due to the different order in which the levels are coupled. The smallest width at half-amplitude, obtained by careful alignment, was 300 G at 4.14 kG, and less than 40 G at 2.07 kG. The residual width is attributed to the mosaic structure occurring in ruby grown by the flame fusion technique.⁹ The various mosaic pieces will have their optic axes displaced

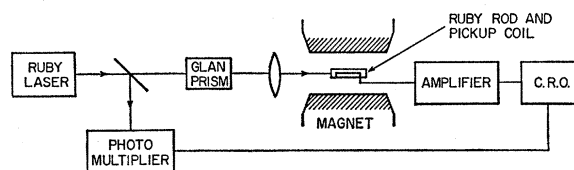


FIG. 3. Schematic diagram of the apparatus.

¹⁵ T. G. Eck, L. L. Foldy, and H. Wieder, Phys. Rev. Letters 10, 239 (1963); K. E. Lassila, Phys. Rev. 135, A1218 (1964).

¹⁶ F. D. Colegrove, P. A. Franken, R. R. Lewis, and R. H. Sands, Phys. Rev. Letters 3, 420 (1959); P. A. Franken, Phys. Rev. 121, 508 (1961).

slightly from the direction of the magnetic field, thus creating a field component perpendicular to this axis in each mosaic piece. The field dependence of the additional signal is approximately Gaussian with slightly broadened wings, and thus does not correspond to the predicted dependence. A Gaussian dependence would be expected from a random distribution of mosaic pieces. Introducing a small perpendicular field component by rotating the crystal several degrees appreciably broadened the wings of the curve, giving somewhat better agreement with Eq. (12).

In another experiment, the temperature of the sample ruby was varied from 300 to 77°K. The measurements were made in a field of 6 kG. As the crystal was cooled from 300°K, the signal amplitude at first increased to approximately twice the 300°K signal, but then decreased as the temperature was lowered further. No signal was obtained at 77°K. These results follow

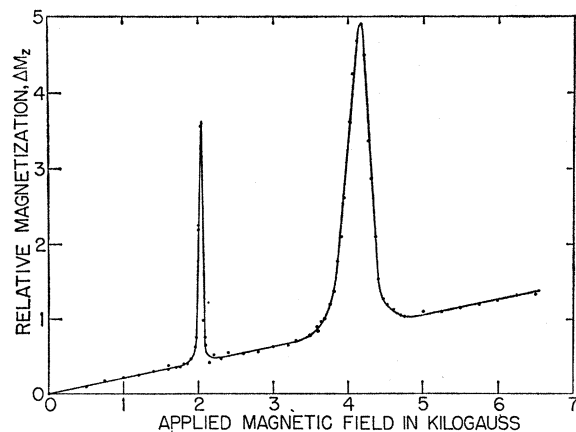


FIG. 4. The laser-induced magnetic signal parallel to the z axis as a function of magnetic field.

directly from the known temperature dependence of the line position and linewidth. Cooling narrows the absorption line, thus increasing the preferential depopulation of the $\bar{n} = +\frac{1}{2}$ level. The shift towards shorter wavelength moves the absorption line further away from the laser wavelength.

Signals due to ΔM_x , obtained by rotating the coil by 90°, were observed near 4 kG. Similar tests made with an axially wound coil also indicated the presence of a component in the y direction. The magnetization parallel to the x axis was eliminated by tilting the rod to give a 100-G component in the y direction and aligning the x axis perpendicular to the field direction. The signal from ΔM_y for this orientation is shown in Fig. 5. ΔM_y is parallel to the field component H_y for fields less than 4.14 kG and reversed polarity when the direction of H_y was reversed. The dashed curve was calculated from Eq. (13b) using $|2V| = 150$ G, and a constant $\alpha_0(+\frac{1}{2}, \nu_L)$. Agreement between the curves is satisfactory near the center, but deviations appear

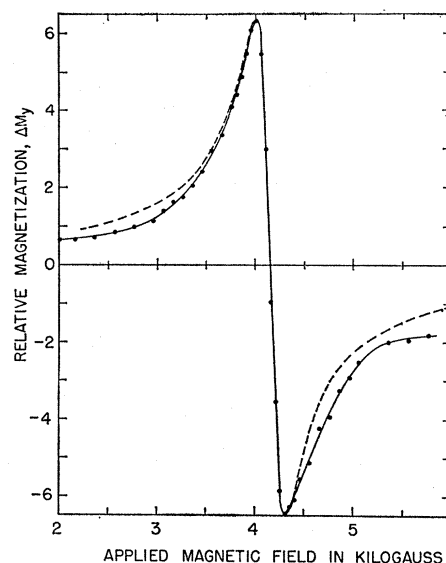


FIG. 5. The laser-induced magnetic signal parallel to the y axis as a function of magnetic field. The dashed curve is obtained from Eq. (13b).

in the wings. The deviation is due to the increase of $\alpha_0(+\frac{1}{2}, \nu_L)$ with increasing field strength.

In all measurements the signal amplitude was linearly proportional to the laser intensity.

4. RELAXATION EFFECTS

The additional magnetization at 2.07 and 4.14 kG is conclusive evidence that the relaxation time of the ground-state is long compared to the duration of the laser pulse. If this relaxation time had been short, the populations of the two excited states, and the ground state would individually be given by a Boltzmann distribution. In this case the direction of the change in the magnetization would be opposed to the direction of the magnetic field (the static magnetization would decrease). This is contrary to the observed direction and to the predictions of Eq. (12). A long ground-state relaxation time is also in agreement with the results obtained by paramagnetic-resonance-saturation methods. Extrapolation of the low-temperature measurements¹⁷ to 300°K yields a value lying between 0.1 and 1.0 μ sec.

Additional information is obtained from the time dependence of the magnetic signal. When the ground-state relaxation time is very long compared to the laser pulse length, the magnetization is proportional to the time integral of the laser intensity. This follows directly from the derivation of Eq. (9). The detected emf is then proportional to the instantaneous laser intensity, as observed. For a relaxation time comparable to the pulse

¹⁷ J. C. Gill and R. J. Elliot, *Advances in Quantum Electronics*, edited by J. R. Singer (Columbia University Press, New York, 1961), p. 399; J. H. Pace, D. F. Sampson, and J. S. Thorp, *Phys. Rev. Letters* 4, 18 (1960); I. J. D'Haenens and C. K. Asawa, *J. Appl. Phys.* 33, 3201 (1962).



FIG. 6. Top: The effect of a magnetic field on the signal level. Bottom: Monitor of the laser intensity. The horizontal sweep is 10^{-7} sec/cm.

(a) $H = 0$ G (b) $H = 190$ G

length, the magnetization would be a more complicated function of time.

These conclusions are only valid if the radiative lifetime of the excited state is long. If the relaxation time for transitions between all the levels had been short compared to the laser pulse length, the magnetization would be directly proportional to the instantaneous laser intensity. In the limit of an extremely short relaxation time, a change in the magnetization would not occur.

The observed emf is a single pulse, having approximately the shape of a laser pulse. In addition, a small decay signal was observed after the termination of the laser pulse. It is caused by the relaxation of the ground-state magnetization.

Cross relaxation may occur when the energy separation between the Zeeman levels are equal.¹⁸ The magnetization was measured as a function of field in the vicinity of several harmonic points. No unusual change in either the peak amplitude or the decay signal was

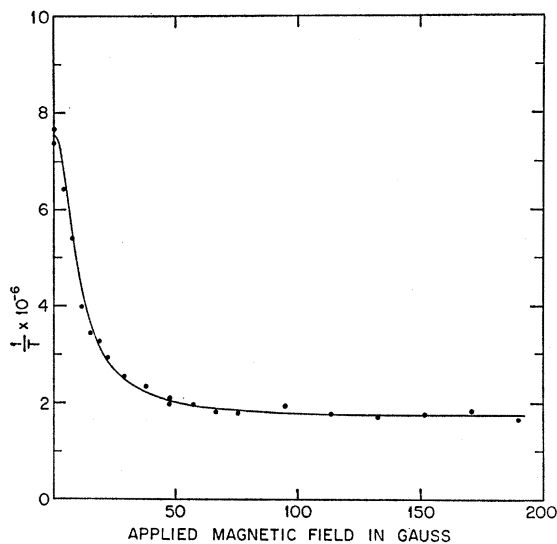


FIG. 7. The relaxation rate of the decay of the magnetic signal as a function of magnetic field. The curve is Eq. (16).

¹⁸ N. Bloembergen, S. Shapiro, P. S. Pershan, and J. O. Artman, *Phys. Rev.* **114**, 445 (1959).

observed. The cross relaxation time is thus longer than the laser pulse length.

Since the signal-to-noise ratio was relatively small, it was not possible to obtain a quantitative measure of the relaxation time in the above experiment.

Hull and Smith¹⁹ have recently measured the alignment of Cr^{3+} ions in ruby using circularly polarized ruby-laser radiation propagating parallel to the optic axis. The magnetization is directed parallel to this axis, and is an order of magnitude larger than the signals obtained above. They measured a relaxation time of a few tenths of a microsecond.

This configuration was used to measure the relaxation time at 300°K as a function of magnetic field parallel to the optic axis of a 0.04% concentration *z*-cut ruby rod. Figure 6 is a reproduction of a double-exposure photograph showing on top the signal for fields of 0 and 190 G. The monitor of the laser pulse is shown below. The signals are qualitatively similar to the signals obtained previously with the laser radiation polarized parallel to the optic axis. The peak amplitude is not affected by the field, but there is a difference in the decay of the magnetization after the termination of the laser pulse. The signal amplitude decreases exponentially in time with a single time constant.

The relaxation rate is shown as a function of field in Fig. 7. The curve is a plot of

$$T^{-1} = 1.75 \times 10^{-6} + 5.8 \times 10^{-6} [1 + (H/10.5)^2]^{-1}. \quad (16)$$

The relaxation time increases from $T_2 = 130 \times 10^{-9}$ sec in zero field, to $T_1 = 570 \times 10^{-9}$ sec at 60 G. Cross relaxation effects are quite apparent in the low-field region. The relaxation rate appears to be constant for fields larger than 60 G. At 1400 G, T_1 was 550×10^{-9} sec, which is within the experimental error of the value obtained at 60 G.

Light polarized perpendicular to the optic axis depopulates the spin $\pm \frac{3}{2}$ levels in addition to the spin $\pm \frac{1}{2}$ levels.^{3,6} It was not possible to separate the individual components and measure the relaxation rate of a single level. A detailed analysis of the relaxation mechanism in ruby for small magnetic fields will not be given here. It should be noted that if the cross-relaxation rate could be expressed as the overlap of individual lines, it would have a Gaussian field dependence^{18,20} since the microwave resonance line has this shape.

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¹⁹ G. F. Hull, Jr. and J. Smith, *Bull. Am. Phys. Soc.* **9**, 447 (1964).

²⁰ P. S. Pershan, *Phys. Rev.* **117**, 109 (1960).