

has been done for the direct interband transitions above the energy gap. For more accurate analysis of the Faraday rotation in *p*-type Ge, it is also necessary to take statistics into account since the dispersion must be weighted by the number of empty states. In spite of these assumptions, however, the above approximate results seem to be reasonable because they can explain the major features of the experimental observations, whereas the classical theory for bound electrons cannot. Lastly, in order to account for the proper sign of the interband Faraday rotation, it is necessary to consider in detail the individual magnetic levels of the two sets of the bands involved in transition, including spin, particularly in the presence or absence of spin-orbit splitting. The analysis would presumably account for the apparent difference between the observation in GaAs as contrasted to InSb and InAs.¹

We would like to thank Dr. G. S. Heller, Dr. H. J. Zeiger, and Dr. L. M. Roth for helpful discussions in connection with this problem.

*This work was done in part at Lincoln Laboratory, operated with support from the U. S. Army, Navy, and Air Force.

†This work was done in part at the National Magnet Laboratory, operated with support by the Air Force through the Air Force Office of Scientific Research.

¹S. D. Smith, T. S. Moss, and K. W. Taylor, *J. Phys. Chem. Solids* **11**, 131 (1959); R. N. Brown and B. Lax, *Bull. Am. Phys. Soc.* **4**, 133 (1959); A. K. Walton and T. S. Moss, *J. Appl. Phys.* **30**, 951 (1959); B. Hartmann and B. Kleman, *Arkiv Fysik* **18**, 75 (1960); M. Cardona, *Phys. Rev.* **121**, 752 (1961).

²M. J. Stephen and A. B. Lidiard, *J. Phys. Chem. Solids* **9**, 43 (1958).

³B. Lax and S. Zwerdling, *Progress in Semiconductors*, edited by A. F. Gibson (Heywood and Company, Ltd., London, 1960), Vol. 5, p. 224; B. Lax, *Proceedings of the Prague Semiconductor Conference*, August, 1960 (to be published).

⁴L. M. Roth, B. Lax, and S. Zwerdling, *Phys. Rev.* **114**, 90 (1959).

⁵R. J. Elliott, T. P. McLean, and G. G. Macfarlane, *Proc. Phys. Soc. (London)* **72**, 553 (1958); B. Lax, L. M. Roth, and S. Zwerdling, *J. Phys. Chem. Solids* **8**, 311 (1959); E. Burstein, G. S. Picus, R. F. Wallis, and F. J. Blatt, *J. Phys. Chem. Solids* **8**, 305 (1959), and *Phys. Rev.* **113**, 15 (1959); S. Zwerdling, B. Lax, L. M. Roth, and K. Button, *Phys. Rev.* **114**, 80 (1959).

⁶H. B. Briggs and R. C. Fletcher, *Phys. Rev.* **87**, 1130 (1952); W. Kaiser, R. J. Collins, and H. Y. Fan, *Phys. Rev.* **91**, 230(A) and 1380 (1953).

HYPERFINE INTERACTIONS IN THE GROUND STATE AND FIRST EXCITED STATE OF Dy¹⁶¹ IN DYSPROSIUM IRON GARNET*

R. Bauminger, S. G. Cohen, A. Marinov, and S. Ofer
Department of Physics, The Hebrew University, Jerusalem, Israel
(Received April 13, 1961)

The hyperfine Zeeman splittings in the recoil-free absorption spectrum¹ of the 26-keV γ rays from Dy^{161m} ($\tau_{1/2} \sim 3 \times 10^{-8}$ sec)² in Dy¹⁶¹ situated in dysprosium iron garnet (DIG) have been observed. This is possible since in the rare earth iron garnets (5Fe₂O₃·3M₂O₃) the sublattice of rare earth ions below the Curie temperature shows a temperature-dependent spontaneous magnetization.³ Nearly all the rare earth elements can be incorporated in iron garnets and since an appreciable recoil-free efficiency is expected at liquid air temperature for the γ transition from the first excited state of many rare earth nuclei, the method is of general application. Values for the effective magnetic field at the nucleus (H_{eff}) and the magnetic and quadrupole moments of the ground state and

first excited state of Dy¹⁶¹ are derived. At 85°K a large nuclear quadrupole interaction has been observed, but at 300°K the quadrupole interaction is much smaller. This strongly temperature-dependent interaction is interpreted as being largely due to the average electric field gradient produced by the partially aligned orbitals of the 4f electron shell and correlated with the spontaneous magnetization (and H_{eff}) within each domain. This gradient will have an axis of symmetry parallel to H_{eff} .

The recoil-free absorption spectrum of polycrystalline DIG, isotopically enriched in Dy¹⁶¹, was measured using a source of Dy^{161m} which showed an unsplit emission line of relatively narrow width. This was prepared as follows: 6.7-day Tb¹⁶¹ was separated by ion exchange

from neutron-irradiated Gd_2O_3 enriched in Gd^{160} , electroplated on copper from a solution of the chloride in pyridine, covered with evaporated copper, and the "sandwich" heated in hydrogen at about $900^\circ C$. With this source the linewidth of the unsplit recoil-free absorption spectrum in a dysprosium aluminum alloy at room temperature was about 4 mm/sec, which is, therefore, an upper limit to the source linewidth. The source (in all measurements at $300^\circ K$) was vibrated on a loud-speaker membrane at 65 cycles/sec and the absorption of the 26-kev radiation recorded automatically as a function of relative velocity as described elsewhere.⁴

Figure 1 shows the absorption spectrum obtained in DIG at $85^\circ K$. Well-resolved lines extending to velocities of about 10 cm/sec in each

direction are seen. The positions of the peaks were confirmed in separate runs extending to smaller velocities. The Dy^{161} ground state is known to be $5/2^-$ and the first excited state has been thought to be $5/2^+$.⁵ The analysis of the results shown in Fig. 1(A) in terms of transitions between the Zeeman levels of the two nuclear states demands that both levels have a spin $5/2$ and that the magnetic moments have opposite sign. The measurements of Park⁶ indicate a negative magnetic moment for the ground state. It was assumed that each level was split according to the expression,⁷

$$W = mg_I \mu_n H_{\text{eff}} + \frac{1}{4} eqQ [3m^2 - I(I+1)] / [I(2I-1)].$$

Table I shows the identification of the transitions between the Zeeman levels. There are a

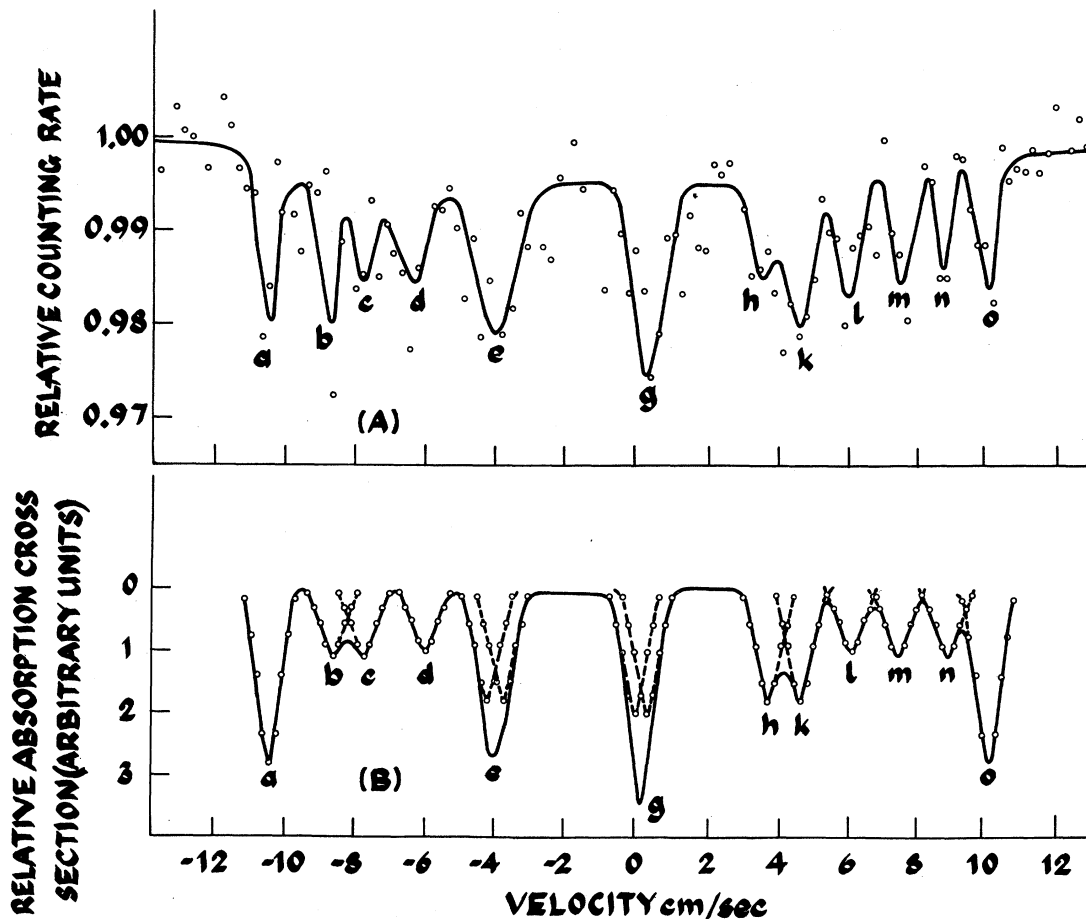


FIG. 1. (A) The absorption by dysprosium iron garnet at liquid air temperature of the 26-kev γ ray emitted at room temperature in the decay of Tb^{161} , diffused into copper, as a function of relative velocity between source and absorber. The statistical error on each point is about 0.3%. (B) Reconstructed spectrum assuming the results of the least-square analysis of the experimental peak energies, the theoretical relative transition probabilities, and a Gaussian line shape.

Table I. Identification of observed transitions between nuclear Zeeman levels at liquid air temperature.

Peaks observed	Transitions between Zeeman substates	Peaks observed	Transitions between Zeeman substates
a	-5/2 → -5/2	h	+1/2 → +3/2
b	-3/2 → -5/2	k	+3/2 → +1/2
c	-5/2 → -3/2	l	+3/2 → +3/2
d	-3/2 → -3/2	m	+3/2 → +5/2
e	{ -3/2 → -1/2 -1/2 → -3/2	n	+5/2 → +3/2
g	{ -1/2 → +1/2 +1/2 → -1/2	o	+5/2 → +5/2

total of 16 possible electric dipole transitions. The calculated Wigner coefficients determining the transition probabilities for two of the transitions (+1/2 → +1/2, and -1/2 → -1/2) are very small and they cannot be observed. The remaining transitions are all observed but at two positions in the spectrum two transitions overlap [*e* and *g* in Fig. 1(A)].

Table II shows the results of a least-squares analysis of the experimental peak energies giving $g\mu_n H_{\text{eff}}$ and $eqQ/4$ for both nuclear levels, and ΔE , the chemical shift between source and absorber.⁸ Figure 1(B) shows the reconstructed spectrum obtained from these values, using the appropriate theoretical relative transition probabilities, and assuming a Gaussian line shape. The agreement between Figs. 1(A) and 1(B) is seen to be very satisfactory. Measurements in DIG at 300°K shows a similar but much narrower spectrum corresponding to a smaller H_{eff} ; the analysis of the results at 300°K is also given in Table II.⁹

Using the value of (-0.37 ± 0.05) nm for the Dy^{161} ground-state magnetic moment obtained by Park,⁶ we get $(+0.42 \pm 0.08)$ nm for the

magnetic moment of the first excited state, $H_{\text{eff}}(85^\circ\text{K}) = (3.5 \pm 0.55) \times 10^6$ oe, and $H_{\text{eff}}(300^\circ\text{K}) = (7.5 \pm 1.5) \times 10^5$ oe. $H_{\text{eff}}(85^\circ\text{K})/H_{\text{eff}}(300^\circ\text{K})$ is then (4.6 ± 1) , which is close to the value 4 for the ratio of the partial spontaneous magnetizations of the dysprosium sublattice at the two temperatures deduced by Pauthenet from magnetization measurements using the Néel model.³ This illustrates again the proportionality between H_{eff} and spontaneous magnetization.^{10,4} The experimental values for the magnetic moment and their signs for the two nuclear states are in agreement with the predictions of Nilsson's unified model.⁵

The results show a remarkably large quadrupole interaction ($eqQ_1/4 = +120$ Mc/sec) at 85°K and a relatively much smaller interaction at 300°K ($eqQ_1/4 < 20$ Mc/sec). This is interpreted as due largely to the average field gradient produced by the 4*f* electrons which should be partially aligned below the Curie temperatures. Assuming that J of the rare earth ion (the ground state of the Dy^{3+} is ${}^6H_{15/2}$) in the garnet is a good quantum number and neglecting crystalline field effects on the 4*f* shell,¹¹ then the electric field gradient

Table II. Summary of results for the hyperfine interactions and chemical shifts observed in dysprosium iron garnet at 300°K. Q_1 and g_1 correspond to the ground state of Dy^{161} ; Q_2 and g_2 correspond to the 26-keV level of Dy^{161} .

Temperature (°K)	ΔE (Mc/sec)	$eqQ_1/4$ (Mc/sec)	$eqQ_2/4$ (Mc/sec)	$g_1\mu_n H_{\text{eff}}$ (Mc/sec)	$g_2\mu_n H_{\text{eff}}$ (Mc/sec)
85	8 ± 10	120 ± 30	90 ± 30	-400 ± 40	+455 ± 40
300	12 ± 10	< 20	< 20	-84 ± 20	+97 ± 20

produced by this mechanism will be equal to the Boltzmann average of the electric field gradient produced by the ion in the various J_z states. In the Weiss field approximation, eqQ will then be given by

$$eqQ = e^2Q \langle 1/r^3 \rangle a \left[\sum_{J_z = -J}^{J_z = +J} [3J_z^2 - J(J+1)] \exp(-J_z g\beta H/kT) \right] / \left[\sum_{J_z = -J}^{J_z = +J} \exp(-J_z g\beta H/kT) \right], \quad (1)$$

where a is the matrix element $\langle \sum_i (3\cos^2\theta_i - 1) \rangle$ for the ion in the state $J_z = J$,¹² and H is the temperature-dependent Weiss field.¹³ The expression in brackets can be regarded as a modified Brillouin function giving the average of $[3J_z^2 - J(J+1)]$ instead of the average of J_z appearing in the usual Brillouin function

$$M/M_S = \left[\sum_{J_z = -J}^{J_z = +J} J_z \exp(-J_z g\beta H/kT) \right] / \left[\sum_{J_z = -J}^{J_z = +J} \exp(-J_z g\beta H/kT) \right], \quad (2)$$

which should describe the dependence of the spontaneous magnetization of the dysprosium sublattice on temperature. The functions of the argument $x = g\beta H/kT$ have been computed for $J = 15/2$, and using Pauthenet's values for the spontaneous magnetization to fix x from Eq. (2) at 85°K and 300°K, Eq. (1) shows that $eqQ(85^\circ\text{K})/eqQ(300^\circ\text{K})$ should equal about 8, consistent with experiment. Moreover, taking $\langle 1/r^3 \rangle = 74 \times 10^{+24} \text{ cm}^3$,¹⁴ $a = 2/3$ (assuming LS coupling),¹² we obtain from the experimental value of $eqQ_1/4$ at 85°K, a value of about $2 \times 10^{-24} \text{ cm}^2$ for Q_1 . This lies between the value $3 \times 10^{-24} \text{ cm}^2$ obtained for Q_1 from measurements of $B(E2)$ in Coulomb excitation¹⁵ and the value 1.1×10^{-24} obtained by Park⁶ from spin resonance experiments. We consider that further investigations leading to a knowledge of the necessary corrections to the simple treatment outlined above will permit accurate estimate of the electric field gradients and nuclear quadrupole moments.

We would like to thank M. Schieber from the Department of Electronics of the Weizmann Institute for preparing the DIG and A. Moustachi for help in chemical problems.

*Supported in part by the U. S. Air Force, Air Research and Development Command, through its European Office.

¹R. L. Mössbauer, *Z. Physik* **151**, 124 (1958).

²S. Ofer, P. Avivi, R. Bauminger, A. Marinov, and S. G. Cohen, *Phys. Rev.* **120**, 406 (1960).

³R. Pauthenet, *Ann. Phys.* **3**, 424 (1958).

⁴R. Bauminger, S. G. Cohen, A. Marinov, and S. Ofer, *Phys. Rev.* **122**, 743 (1961).

⁵B. R. Mottelson and S. G. Nilsson, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **1**, No. 8 (1959).

⁶J. G. Park, *Proc. Roy. Soc. (London)* **A245**, 118 (1958).

⁷This is an approximation to first order in the ratio of the quadrupole interaction (assumed to be axially symmetric) to magnetic interaction. The arguments presented in this Letter lead us to believe that q is predominantly axially symmetric and parallel to H_{eff} . To the extent that this is true, the use of the expression is justified even if the ratio of interactions is not small.

⁸O. C. Kistner and A. W. Sunyar, *Phys. Rev. Letters* **4**, 412 (1960).

⁹The definition of the lines in Fig. 1(A) shows that there must be a fast thermal exchange between the various Stark levels of the Dy ion excited at 85°K. Nevertheless the linewidths are broader than those observed at 300°K, and this might be ascribed to a slower relaxation between the Stark levels at 85°K.

¹⁰D. E. Nagle, H. Frauenfelder, R. D. Taylor, D. R. F. Cochran, and B. T. Matthias, *Phys. Rev. Letters* **5**, 364 (1960).

¹¹A detailed theoretical treatment should take into account the possibility of partial decoupling of the orbital angular momentum from the aligned spin angular momentum under the combined influence of the exchange interactions acting on the spins and the crystal field interactions. There is some evidence for a partial quenching of the orbital magnetic moment in the garnets. (See reference 3.)

¹²K. W. Stevens, *Proc. Phys. Soc. (London)* **A65**, 209 (1952); a is equal to the factor α of Stevens, multiplied by $[3J^2 - J(J+1)]$, which in this case is equal to 105.

¹³We neglect the atomic core distortion factor which has not yet been calculated for rare earth states. [R. M. Sternheimer, *Phys. Rev.* **95**, 736 (1954).]

¹⁴B. Bleaney, *Proc. Phys. Soc. (London)* **A68**, 937 (1955).

¹⁵N. P. Heydenburg and G. F. Pieper, *Phys. Rev.* **107**, 1297 (1957).