Nuclear Resonance Absorption in Dy^{161} Situated in Dy$_2$O$_3$ and Dysprosium Iron Garnet*

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The recoil-free resonant absorption of the 26-kev γ ray ($T_{\frac{1}{2}}\sim 3\times 10^{-8}$ sec) emitted in the decay of Tb^{161} by absorbers containing Dy^{161} has been investigated. High Mössbauer efficiencies at room temperatures have been observed for sources and absorbers in the form of oxide and rare earth iron garnet. The line shapes obtained were very broad, of the order of 100 times the natural widths, and showed no resolved lines. The broad lines are interpreted as due to a wide complicated hyperfine spectrum whose details have been smoothed out by transitions between magnetic sublevels induced by paramagnetic relaxation. In the rare earth iron garnet, the exchange field acting on the rare earth ion should decouple the nuclear and electron spins. The effective magnetic field at the nucleus in the rare earth garnet is about 2×10^6 oe.

W E have investigated the recoil-free resonant absorption of the 26-kev γ ray (5/2− → 5/2+, $T_{\frac{1}{2}}=2.8\times 10^{-8}$ sec),[1,2] emitted in the decay of Tb^{161} by absorbers containing Dy^{161}. The very large efficiencies observed for recoil-free emission and absorption with sources and absorbers in the form of oxide, or rare earth iron garnet, even at room temperatures, permit a study of the structure of the absorption lines. The dysprosium ions are strongly paramagnetic (for the free ion the ground state is $h_{15/2}$ and the magnetic moment is 10.6 μ_B) and one would expect the hyperfine interaction to lead to a complex splitting of the emission and absorption lines. On the other hand, the strongly temperature-dependent paramagnetic relaxation in solids should broaden the sharp hyperfine levels by inducing transitions between the magnetic sublevels, and if the spin correlation times are very short compared to the nuclear precession period, even reduce the over-all width of the hyperfine splitting.[3] The present case is favorable for studying these phenomena in different crystalline environments. In this work absorption lines of the order of 100 times the natural line width have been observed at room temperatures, but there is no evidence of detailed line structure. Measurements in rare earth garnets are of particular interest, in view of the ferrimagnetic nature of these materials and the existence of a moderately large exchange field acting on the rare earth ions.[4,5]

Tb^{161} (half-life 6.9 days) was prepared by irradiating Gd$_2$O$_3$ enriched in Gd^{160} with thermal neutrons in a reactor. Gd^{161} is formed by neutron capture and decays with a half-life of 3.6 min to Tb^{161}. As sources in the resonance experiments, the irradiated Gd$_2$O$_3$ containing the Tb^{161} was used, and also a mixed rare earth poly-crystalline iron garnet (5Fe$_2$O$_3\cdot$3M_2O$_3$) containing 5% irradiated gadolinium and 95% natural dysprosium. Dysprosium oxide (Dy$_2$O$_3$) or polycrystalline dysprosium iron garnet were used as absorbers.

The 26-kev radiation was detected with a large argon-filled proportional counter. Relative uniform motion between source and absorber with velocities between 0.01 and 10 cm/sec were provided by a carriage driven by an accurately machined eccentric wheel rotating uniformly. Only the central part of the motion was used, over a distance of about 1 cm, and the transmission of the absorber was separately measured for both positive and negative velocities using fast mercury relays to operate the scalers. A 1000-cycle oscillator was used to time the counts.

In Fig. 1 a plot of the counting rates obtained at 300°K, with a 40-mg/cm^2 Dy$_2$O$_3$ absorber and a Gd$_2$O$_3$ source, for different source velocities is shown. From this figure it is seen that the maximum relative change in counting rate is about 6%. This number is to be corrected because of the dilution of the effect caused by the background in the gamma-ray spectrum. The corrected maximum effect in this case is then (8.5±1)%.

The shape of the line approximates that of an isosceles triangle with a width at half maximum of 1.3 cm/sec. The simplest although crude interpretation for this shape is that both the emission and absorption lines have approximately rectangular shapes with widths of 1.3 cm/sec. This width is 50 times larger than the half-width of the natural line calculated from the mean life of the level. No evidence of subsidiary resonances was found up to the highest velocity used—10 cm/sec. The absence of a detailed line structure is interpreted as being due largely to paramagnetic relaxation.

Figure 2 shows the maximum change in counting rate, corrected for dilution, as a function of the thickness of Dy$_2$O$_3$ absorber. The continuous curve drawn in Fig. 2 is the graph of the function $0.23(1-e^{-12.5x})$, where x is the absorber thickness in g/cm^2. The approximately exponential character of the absorption of the resonant radiation is consistent with the assumption made above that the absorption line has a rectangular shape. The

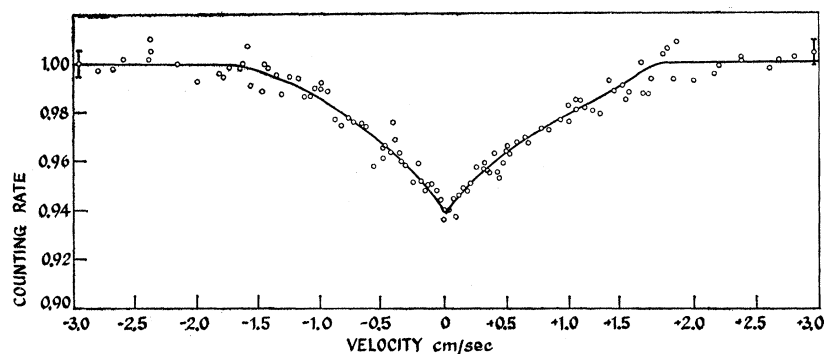[1] G. Hansen, O. Nathan, O. B. Nielsen, and B. K. Sheline, Nuclear Phys. **6**, 630 (1958).
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FIG. 1. The absorption by Dy_2O_3 of the 26-kev γ ray emitted in the decay of Tb^{161} as a function of relative velocity between source and absorber.

magnitude of the effect thus extrapolated for an infinitely thick absorber is then $f_1 = 0.23$ and this is therefore the probability for emission of recoil-free photons from the source at room temperature. The experiments showed that the mass absorption coefficient for the resonant radiation in the Dy_2O_3 absorber is 12.5 cm^2/g. The calculated mass absorption coefficient for radiation at the resonance energy, assuming the absorption line to have an unperturbed natural Breit-Wigner shape, is 560 cm^2/g and the conversion coefficient of the 26-kev transition is three, as is to be expected for a pure $E1$ transition.[1] Assuming the interpretation of the line shape given in the previous paragraph, the probability f_2 for recoil-free absorption in the absorber can be calculated from the ratio of the area of the experimental absorption line to the theoretical area of the natural absorption line. The value obtained in this way for f_2 is about 0.9. This is a much more uncertain figure than the value for f_1 given above. From the experimental value $f_1 = 0.23$ for the source of Dy in Gd_2O_3 we obtain a Debye temperature of about 230°K using the Debye-Waller relation.[6]

In the measurements with source and absorber (40 mg/cm^2) in the form of iron garnets at 300°K, the maximum relative change in counting rate was $(4\pm1)\%$ and the line had a much greater over-all width (width at half maximum 3.6 cm/sec), but showed, within the statistical errors, no sign of a line structure.

Some experiments were also carried out at liquid air temperatures, with the oxide source and absorber. The maximum relative change in counting rate corrected for dilution, using a 40-mg/cm^2 absorber, was in this case $(5\pm1)\%$, significantly less than the result at room temperatures. This result was somewhat surprising since the efficiencies for recoil-free emission and absorption are expected to be larger at lower temperatures, in accordance with the Debye-Waller factor. The results can be explained, however, if we assume that the over-all broadening is greater at the lower temperatures, leading to a small average cross section for absorption. The present measurements of the line shape at liquid air

temperatures, although still inexact, support this conclusion.

Park[7] has investigated the hyperfine splittings of the electron spin resonance absorption in dysprosium, for Dy^{161} situated in dysprosium acetate at liquid helium temperatures, and obtained $A_x = 0.038$ cm^{-1}. Assuming splittings of this value for the nuclear ground state in dysprosium oxide and ignoring for simplicity the splitting of the excited state (the nuclear magnetic moment is not known), one expects a maximum over-all width of about 4 cm/sec, for the absorption line, which is larger than that observed for the oxides. On the other hand, one expects several Stark levels to be excited at room temperatures, and the hyperfine splitting will be different for each level. Moreover, at room temperatures, the over-all hyperfine splittings may be reduced as a consequence of paramagnetic relaxation, and this might explain the evidence for line narrowing at room temperatures mentioned above. The fact that the experimental line width is of the same order as the width expected from the results of Park, shows that the electron spin correlation time τ is not very small compared to the nuclear precession frequency in the hyperfine field. We conclude that τ is not very small compared with 10^{-10} sec.
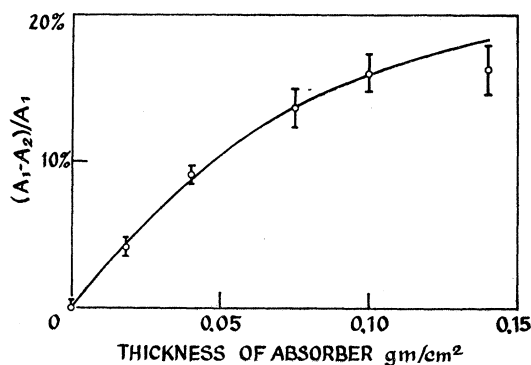

FIG. 2. The points represent the resonance absorption as a function of the thickness of Dy_2O_3 absorber. The solid curve is the graph of the function $0.23(1 - e^{-12.5x})$, where x is the absorber thickness in g/cm^2.

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In the case of the garnets, an exchange field of the order of 10^5 oe acts on the rare earth ion, due to the interaction between the iron and rare earth sub-lattices.[4,5] This exchange field is sufficient to produce saturation magnetization of rare earth ions at room temperatures, so that one would still expect that para-magnetic relaxation might broaden the individual absorption lines. At lower temperatures, however, and possibly even at liquid air temperatures, one might expect to see some line structure, in view of the magnitude of the exchange field acting on the rare earth ion.

The exchange field should decouple the nuclear and electron spins. The over-all maximum width of the nuclear ground state can then be written as $2 \mu H_{av}$, where H_{av} is the effective field at the Dy nuclei averaged over the different Stark levels operative at room temperatures. If we ignore, again, the hyperfine splittings of the excited state, and take $\mu = 0.35$ nm,[7] then, from the experimental width of the absorption line with garnets at room-temperatures, we obtain $H_{av} \sim 2 \times 10^6$ oe.

ACKNOWLEDGMENTS

Paramagnetic Resonance Spectrum of Mn²⁺ in ZnSiF₆:6H₂O, Δm = ±1 Transition*

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In the spectrum of Mn²⁺ in ZnSiF₆:6H₂O a number of weaker lines are observed at intermediate angles of the magnetic field H with respect to the crystal axis, in addition to the 30 allowed transitions $\Delta M = \pm 1$, $\Delta m = 0$. These lines have been measured and assigned to $\Delta M = \pm 1$, $\Delta m \pm 1$ transitions. The relatively strong intensity is explained and the intensity of the lines is shown to be proportional to $(D/A)^2 \cos^2\theta \sin^2\theta$. These forbidden transitions can be utilized for dynamic polarization of manganese nuclei.

INTRODUCTION

THE paramagnetic resonance spectrum of Mn²⁺ in single crystals of zinc fluosilicate (ZnSiF₆:6H₂O) has been analyzed by Bleaney and Ingram.[1] The spectrum consists of 5 groups of 6 lines each corresponding to $\Delta M = \pm 1$ and $\Delta m = 0$. In addition Bleaney and Ingram observed a number of weaker lines when the external magnetic field was not directed along or perpendicular to the trigonal axis of the crystal. We have studied these absorption lines and are able to assign these to $\Delta M = \pm 1$, $\Delta m = \pm 1$ transitions. The relatively strong intensity of these so-called "forbidden" transitions is explained. These transitions can be utilized for dynamic polarization of manganese nuclei.

THEORY

The conventional spin Hamiltonian pertaining to an ion in an axial symmetry is given by[1,2]

$$\mathcal{H} = g\beta \mathbf{H} \cdot \mathbf{S} + D[S_z^2 - \tfrac{1}{3}S(S+1)] + F(a)$$
$$+ A S_z I_z + B(S_x I_x + S_y I_y)$$
$$- \gamma \beta_N \mathbf{H} \cdot \mathbf{I} + Q'[I_z^2 - \tfrac{1}{3}I(I+1)]. \quad (1)$$

In the case of manganese $S = I = \tfrac{5}{2}$. The symbols in (1) have their usual meaning. Taking the direction of the magnetic field as the axis of quantization, the allowed transitions pertaining to the electronic part of the spectrum are given to second order by

$$M = \pm\tfrac{5}{2} \leftrightarrow \pm\tfrac{3}{2} \quad H = H_0 \mp 2D(3\cos^2\theta - 1) - (32D^2/H_0)\cos^2\theta \sin^2\theta + (D^2/H_0)\sin^4\theta \mp 2pa,$$

$$M = \pm\tfrac{3}{2} \leftrightarrow \pm\tfrac{1}{2} \quad H = H_0 \mp D(3\cos^2\theta - 1) + (4D^2/H_0)\cos^2\theta \sin^2\theta - (5/4)(D^2/H_0)\sin^4\theta \pm \tfrac{5}{2}pa, \quad (2)$$

$$M = +\tfrac{1}{2} \leftrightarrow -\tfrac{1}{2} \quad H = H_0 + (16D^2/H_0)\cos^2\theta \sin^2\theta - (2D^2/H_0)\sin^4\theta,$$

where H is the magnetic field at which the transition is observed, $H_0 = h\nu/g\beta$, $p = 1 - 5\Phi$, $\Phi = l^2m^2 + m^2n^2 + n^2l^2$, (l,m,n) being the direction cosines of the magnetic field relative to the cubic axes.

In addition we need the matric elements resulting from the interaction between the nucleus and the electron

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