

Transmission of Polarized Neutrons through Samples of Polarized Ho^{165} Nuclei*

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Transmission experiments were carried out with polarized-monochromatic neutrons and polarized Ho^{165} nuclei using polycrystalline samples of holmium-ethyl sulfate and holmium metal. The spins of the two excited states of the compound nucleus corresponding to the neutron resonances at 3.92 and 12.8 eV have been assigned as $J=I+\frac{1}{2}=4$. The nuclear polarization obtained for the metal sample has the same sign and magnitude as in the holmium-ethyl sulfate. The hyperfine constant A/k was calculated to be $0.61 \pm 0.06^\circ\text{K}$ for the ethyl sulfate and $0.65 \pm 0.08^\circ\text{K}$ for the metal. The depolarization of the neutron beam in passing through the magnetized samples has been studied at thermal energies.

I. INTRODUCTION

FEW reliable methods exist for measuring the total angular momentum J of the compound-nuclear states which correspond to resonances in slow neutron ($l=0$) cross sections. Although a small cross section due to resonance scattering is always present, the cross section at such resonances is predominantly due to radiative capture. In principle, J can be determined from precise measurements of both the total and the scattering cross sections through the resonance region; however, in only a few favorable cases^{1,2} is the scattering cross section sufficiently large to yield data of the required precision. Assignments of J values based on the spectrum of capture gamma rays emitted at the resonance must, for the present, be regarded with skepticism, except in rare instances³ where special constraints permit a unique choice.

The most straightforward method for measuring the J values of resonances makes use of polarized neutrons and polarized-target nuclei. The transmission of a monochromatic polarized neutron-beam through a sample containing polarized nuclei will depend on the J value of the resonance and on the relative polarization of the neutrons and the nuclei; e.g., if $J=I+\frac{1}{2}$ (where I is the spin of the target nucleus and J is the spin of the compound state) the transmission will be lower for neutron and nuclear spins polarized parallel than for antiparallel polarization. Hence, the J value can be determined by observing the change in transmission which occurs when the relative polarization is changed from parallel to antiparallel. Experiments of this type

have already been done with Mn^{55} , In^{115} , Sm^{149} , Eu^{151} , and $\text{Gd}^{155,157}$ resonances.^{4,5}

In this paper we shall report the initial results obtained with equipment designed to make transmission measurements with polarized-monochromatic neutron beams and polarized-nuclear samples. Two resonances in the Ho^{165} cross section, at 3.92 and 12.8 eV, have been investigated. In addition, we have studied the depolarization of the neutron beam which results from the magnetic structure of the samples, and several interesting features relating to the hyperfine interactions in holmium-ethyl sulfate and holmium metal.

It is known from paramagnetic resonance experiments⁶ that the hyperfine splitting is extremely large in the case of diluted holmium ethyl sulfate. Thus, reasonably large nuclear polarization of Ho^{165} in holmium-ethyl sulfate can be expected at temperatures which can be achieved with a liquid-helium bath; i.e., $\sim 1^\circ\text{K}$.

When the present work was started, very little was known about the hyperfine splitting in holmium metal. Spectroscopic measurements⁷ indicated that the hyperfine splitting of the ground state was large. As will be seen, our transmission measurements on holmium metal at 0.95°K confirm this, and, in fact, yield a quantitative value for the hyperfine-interaction constant. This value is consistent with the recent specific heat measurements on holmium metal by Gordon, Dempsey, and Soller.⁸

When the neutron beam passes through the magnetized samples it is partially depolarized as a result of the gross-magnetic structure of the crystallites. It was necessary to make a careful study of the depolarization effect for proper interpretation of the data. The varia-

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¹ B. N. Brockhouse, Can. J. Phys. **31**, 432 (1953).

² H. L. Foote, Jr., dissertation University of Utah, 1954 (unpublished); R. E. Wood, Phys. Rev. **104**, 1425 (1956); J. A. Moore, *ibid.* **109**, 417 (1958).

³ H. H. Landon and E. R. Rae, Phys. Rev. **107**, 1333 (1957).

⁴ S. Bernstein, L. D. Roberts, C. P. Stanford, J. W. T. Dabbs, and T. E. Stephenson, Phys. Rev. **94**, 1243 (1954); J. W. T. Dabbs, L. D. Roberts, and S. Bernstein, *ibid.* **98**, 1512 (1955); L. D. Roberts, S. Bernstein, J. W. T. Dabbs, and C. P. Stanford, *ibid.* **95**, 105 (1954).

⁵ A. Stolovy, Phys. Rev. **118**, 211 (1960); Bull. Am. Phys. Soc. **5**, 294 (1960); **6**, 275 (1960).

⁶ J. M. Baker and B. Bleaney, Proc. Phys. Soc. (London) **A68**, 1090 (1955).

⁷ V. B. Belionin, Optics and Spectroscopy **5**, 236 (1958).

⁸ J. E. Gordon, C. W. Dempsey, and T. Soller, Phys. Rev. **124**, 724 (1961).

tion of the depolarization with neutron velocity, with sample thickness, and with magnetizing field yields some information on the domain size in the crystallites.

II. EQUIPMENT

The equipment used in the experiments⁹ consisted of a crystal spectrometer for obtaining monochromatic, polarized neutrons by Bragg reflection, and a cryostat for cooling the nuclear sample. The cryostat was positioned on the spectrometer "arm," and had evacuated re-entrant windows to permit the Bragg reflected-neutron beam to pass through the polarized nuclear sample. The monochromating crystal, which served also to polarize the reflected neutrons,¹⁰ was a face-centered cubic single crystal¹¹ of the alloy 92% Co-8% Fe. Most data were taken with the (111) reflection which ideally yields approximately 100% neutron polarization; however, some data were taken with the (220) reflection which yields better energy resolution at the cost of decreased intensity and polarization (~85%). The reflected neutrons are polarized parallel to the direction of magnetization of the crystal.

The polarization direction of the reflected beam is adiabatically rotated from vertical to horizontal by means of a magnetic guide field as it passes along the arm of the spectrometer. This rotation is necessary because the nuclear target in the cryostat is polarized in the horizontal direction. The rotation can be made either to the right or the left by means of a mechanical device which links together the successive-permanent-magnet elements of the guide field.

The polarization of the reflected beam can be measured by installing an analyzer unit in place of the regular BF₃ detector. The analyzer consists of a small Bragg spectrometer containing a Co-Fe crystal similar to the monochromating crystal. The polarization is measured by comparing the intensity of the second Bragg reflection when the first and second crystals are magnetized successively parallel and antiparallel. The intensity of the second reflection drops off rapidly with neutron energy, and, hence the beam polarization can be measured by this means only for energies ≤ 0.5 ev. The polarization for the (111) reflection was normally about 0.95; and the efficiency ϕ for changing the neutron polarization from parallel to antiparallel was about 0.98.

Although the cryostat was designed for cooling samples to temperatures of $\approx 0.01^\circ$ to 0.1°K , this

feature was not used in the present measurements since adequate nuclear polarizations were obtained at the liquid-helium bath temperatures. The liquid-helium bath could be maintained at a temperature of 0.95°K by pumping with an oil diffusion pump, and a mechanical pump. The samples were mounted on a lifting device, and could be moved in and out of the beam. Thus absolute transmission measurements could be made at the low temperature.

The spectrometer and cryostat will be described more completely in a separate paper.⁹

III. SAMPLES

Deuterated-holmium-ethyl sulfate, $\text{Ho}(\text{C}_2\text{H}_5\text{SO}_4)_3 \cdot 9\text{D}_2\text{O}$, was made from holmium oxide supplied by Lindsay Chemical Company. The oxide contained at most 0.1% impurities; mainly Dy- and Er-oxide. The crystals of holmium-ethyl sulfate contained at most 2% normal water. Hence, the scattering of neutrons due to hydrogen atoms has been substantially reduced. A powdered sample of holmium-ethyl sulfate 5.0-cm high, 2.4-cm wide, and 0.95-cm thick was prepared.

The holmium metal, which was also obtained from Lindsay, had the same impurities as the above-mentioned oxide. The metal was remelted in a tantalum boat using a vacuum furnace. The transmission experiments were carried out with a slab 1.6-cm high, 2.4-cm wide, and 0.137-cm thick. The depolarization was studied with samples 0.032- and 0.062-cm thick. All these samples were carefully cut from the same melt.

IV. TRANSMISSION OF POLARIZED NEUTRONS THROUGH SAMPLES OF POLARIZED NUCLEI

A. The Case of Monoenergetic Neutrons

The polarization of the neutron beam is defined by

$$f_n \equiv (w_+ - w_-)/(w_+ + w_-), \quad (1)$$

where w_+ and w_- are, respectively, the numbers of neutrons parallel and antiparallel to the applied magnetic field H_0 . As the beam passes through the nuclear sample neutrons in both spin states are lost by capture or scattering. In addition, some neutrons can change from one spin state to the other due to magnetic interactions within the sample, especially in polycrystalline samples with strong anisotropic-magnetic properties. The losses of neutrons from the beam are given by the simultaneous-differential equations:

$$\begin{aligned} dw_+ &= [-w_+N\sigma(1+\rho f_N) - D(w_+ - w_-)]dx, \\ dw_- &= [-w_-N\sigma(1-\rho f_N) + D(w_+ - w_-)]dx, \end{aligned} \quad (2)$$

where N is the number of target nuclei per cm^3 , σ is the appropriate total cross section, ρ is a statistical weighting factor which has the value -1 or $1/(I+1)$ depending on whether the spin associated with the cross section is $I - \frac{1}{2}$ or $I + \frac{1}{2}$, $f_N = \langle I_z \rangle / I$ is the nuclear polarization along the direction of the polarizing field H_0 , and D^{-1} is the mean free path for spin reversal.

⁹ A complete description of the spectrometer and cryostat will be submitted to the Review of Scientific Instruments in the near future. Brief descriptions were given by H. Marshak, C. A. Reynolds, F. J. Shore, and V. L. Sailor, *Bull. Am. Phys. Soc.* **3**, 17 (1958), and V. L. Sailor, H. Marshak, F. J. Shore, C. A. Reynolds, and H. Postma, *ibid.* **6**, 275 (1961).

¹⁰ R. Nathans, C. G. Shull, G. Shirane, and A. Andresen, *J. Phys. Chem. Solids* **10**, 138 (1959).

¹¹ The authors are grateful to Dr. R. M. Bozorth and Dr. A. J. Williams of the Bell Telephone Laboratories for growing and cutting the first Co-Fe crystal used in this experiment. A second specimen was purchased from the Virginia Institute for Scientific Research, 2820 Grove Ave., Richmond 21, Virginia.

Equation (1) can be solved for w_+ and w_- by straightforward methods:

$$w_{\pm} = e^{-\alpha t} \{ w_{\pm}^0 [\cosh \kappa t \mp \tau \sinh \kappa t] + w_{\mp}^0 \nu \sinh \kappa t \}, \quad (3a,b)$$

where t is the sample thickness, $\alpha = N\sigma + D$, $\kappa = (N^2\sigma^2 p^2 + D^2)^{1/2}$, $\tau = N\sigma p / \kappa$, $\nu = D / \kappa$, and $p = \rho f_N$. The intensities of the two spin components of the neutron-beam incident on the target are w_+^0 and w_-^0 .

The initial polarization of the neutron beam is defined as:

$$f_n^0 = (w_+^0 - w_-^0) / (w_+^0 + w_-^0). \quad (4)$$

After passing through the target the polarization of the neutron beam, obtained from Eqs. (3a,b), is

$$f_n = \frac{f_n^0 - (\tau + \nu f_n^0) \tanh \kappa t}{1 - (\tau f_n^0 - \nu) \tanh \kappa t}. \quad (5)$$

If $N\sigma p \ll D$, Eq. (5) reduces to

$$f_n = f_n^0 \exp(-2Dt), \quad (5a)$$

which affords the means for measuring D directly at an energy where the nuclear cross section σ of the sample is negligible.

Let \mathcal{T}_p and \mathcal{T}_a denote the transmissions of the sample for the two cases where the neutrons are polarized, respectively, parallel and antiparallel to the external-magnetic field H_0 at the sample. Now

$$\mathcal{T}_{p,a} = \frac{(w_+ + w_-)_{p,a}}{w_+^0 + w_-^0} = \frac{C_{p,a}}{C_0}, \quad (6)$$

where C_0 is the counting rate obtained with no sample in the beam, and C_p and C_a are the counting rates obtained with the neutrons polarized, respectively, parallel and antiparallel to H_0 . Substituting Eqs. (3a,b) into (6), we obtain for the transmissions:

$$\mathcal{T}_p = e^{-\alpha t} [\cosh \kappa t - (\tau f_n^0 - \nu) \sinh \kappa t], \quad (7a)$$

and

$$\mathcal{T}_a = e^{-\alpha t} [\cosh \kappa t + (\tau f_n^0 + \nu) \sinh \kappa t]. \quad (7b)$$

In these formulas, f_n^0 denotes the neutron polarization if the beam is polarized parallel to H_0 . If the neutron polarization is reversed with respect to this external field, the beam polarization is given by $-\phi f_n^0$, where ϕ is the efficiency for reversing the neutron polarization.

In the following sections we will make use of the *transmission effect*, defined by

$$\mathcal{E} = (\mathcal{T}_p - \mathcal{T}_a) / (\mathcal{T}_p + \mathcal{T}_a) = (C_p - C_a) / (C_p + C_a). \quad (8)$$

Combining Eqs. (7a,b) and (8), we obtain

$$\mathcal{E} = -\frac{\frac{1}{2}(1+\phi)f_n^0\tau \tanh \kappa t}{1 - [\frac{1}{2}(1-\phi)f_n^0 - \nu] \tanh \kappa t}. \quad (9)$$

If $\kappa t \ll 1$, Eq. (9) reduces to

$$\mathcal{E} = -\frac{1}{2}(1+\phi)f_n^0(1-Dt)N\sigma p t; \quad (9a)$$

or if $D=0$, and $\phi \approx 1$, Eq. (9) reduces to

$$\mathcal{E} = -\frac{1+\phi}{2}f_n^0 \tanh(N\sigma p t). \quad (9b)$$

B. Effect of Spectrometer Resolution

The preceding equations were derived on the assumption that the neutron beam is monoenergetic. However, the Bragg reflected beam of the spectrometer contains a distribution of energies given by the normalized resolution function

$$R(E' - E) = E'^{-\eta} \exp\left[-\frac{4 \ln 2}{(\Delta E)^2}(E' - E)^2\right]. \quad (10)$$

The quantity ΔE is the width of the distribution determined primarily by the collimation of the beam. The term $E'^{-\eta}$ takes account of various intensity factors in the system which are energy dependent; e.g., detector sensitivity, reactor spectrum, and crystal reflectivity. For this spectrometer in the energy range 0.5 to 15 eV, η varies from 2 to 2.5 and can be determined for each energy from a curve of the open beam counting rate as a function of energy.

The corrected expressions for \mathcal{T}_p , \mathcal{T}_a , and \mathcal{E} are obtained from a convolution of (10) with Eqs. (6) and (8). The resulting formulas are somewhat complicated, and need not be presented here because simpler approximations are adequate for most resonances.

The transmission $\langle \mathcal{T} \rangle$ for unpolarized neutrons passing through a target of unpolarized nuclei is given by

$$\langle \mathcal{T} \rangle = \int_0^\infty R(E' - E) e^{-N\sigma t} dE' / \int_0^\infty R(E' - E) dE'. \quad (11)$$

For the case where $Dt \ll 1$ and $\phi \approx 1$ the transmission effect may be written in the form

$$\langle \mathcal{E} \rangle = -\frac{1}{2}(1+\phi)(1-Dt)f_n^0 p \times \frac{\int_0^\infty R(E' - E) e^{-N\sigma t} \sinh(N\sigma p t) dE'}{\int_0^\infty R(E' - E) e^{-N\sigma t} \cosh(N\sigma p t) dE'}. \quad (12)$$

For small values of p , this expression, to good approximation, can be written as

$$\langle \mathcal{E} \rangle = -\frac{1}{2}(1+\phi)(1-Dt)f_n^0 p \times \frac{\int_0^\infty R(E' - E) e^{-N\sigma t} N\sigma t dE'}{\int_0^\infty R(E' - E) e^{-N\sigma t} dE'}. \quad (12a)$$

It should be noted that σ is a function of E' , and, in the case of a neutron resonance, is the Doppler-broadened Breit-Wigner cross section. The integrals in Eqs. (11), (12), (12a) must be evaluated numerically.

V. POLARIZATION OF HO¹⁶⁵

A. Polarization of Ho Nuclei in Holmium-Ethyl Sulfate

The electronic ground state of Ho³⁺ in holmium-ethyl sulfate is a non-Kramers doublet. Its hyperfine splitting is described by the following spin Hamiltonian with effective spin $S' = \frac{1}{2}$:

$$\mathcal{H} = g_{11}\mu_B H_z S'_z + A S'_z I_z + \mathcal{H}', \quad (13)$$

where \mathcal{H}' contains terms which are small compared to the first two. These small terms describe the splitting of the non-Kramers doublet due to the electric-crystal field, the nuclear-quadrupole interaction, the interaction between the Ho³⁺ ions, and the direct interaction between the external magnetic field and the nucleus. From paramagnetic resonance experiments⁶ on Ho³⁺ incorporated in yttrium-ethyl sulfate, Baker and Bleaney found that $g_{11} = 15.36$, $I = 7/2$, and $A/k = 0.48^\circ\text{K}$. They concluded from a detailed study¹² that the sign of A is positive. This conclusion also follows from the theory of rare-earth ions,¹³ if one assumes the large nuclear magnetic moment of Ho¹⁶⁵ is positive. Hence, the nuclear spins are polarized parallel to the applied magnetic field. If a large external field is applied, the nuclear polarization can be calculated to a reasonable degree of accuracy from the first two terms of the spin Hamiltonian. We used a field of 10 koe in our measurements. This gives an electronic magnetization close to saturation for most of the crystallites of the holmium-ethyl-sulfate sample. The nuclear polarization¹⁴ f_N is then given by

$$f_N = B_I(A/2kT) \left\langle \cos\theta \tanh\left(\frac{g_{11}\mu_B H_0 \cos\theta}{2kT}\right) \right\rangle_{\text{av}}, \quad (14)$$

where θ is the angle between the c axis of a crystallite and the external magnetic field H_0 . The Brillouin function $B_I(\beta)$ is given by

$$B_I(\beta) = \frac{2I+1}{2I} \coth\left(\frac{2I+1}{2}\beta\right) - \frac{1}{2I} \coth\left(\frac{\beta}{2}\right), \quad (15)$$

where the argument $\beta = A/2kT$.

Equation (14) contains an averaging over the random orientation of the crystallites which must be done numerically. Using Eq. (14), the nuclear polarization at temperature of 0.95°K can be calculated using the values of A/k , and g_{11} given above. The calculated

value at 0.95°K is

$$f_N = 0.493 B_I(0.252) = 0.176. \quad (16)$$

It will be seen in Sec. VII that this result is in qualitative agreement but slightly lower than our experimental value.

B. Nuclear Polarization in Holmium Metal

If we assume that the hyperfine splitting of holmium metal can be described by the same effective-spin Hamiltonian Eq. (13), the nuclear polarization in the metal will be expressed as

$$f_N = B_I(\beta) M/M_\infty. \quad (17)$$

Here M is the magnetization of a polycrystalline-metal sample and M_∞ is the saturation magnetization of a single crystal along an axis of easy magnetization.

VI. DEPOLARIZATION OF THE NEUTRON BEAM

A. Theory

The equations derived in Sec. IV show that it is important to determine the loss in polarization of the neutron beam as it passes through the sample, i.e., to measure the parameter D . Within a sample, which is not completely magnetized, the most important cause of depolarization arises from the crystal-magnetic anisotropy and the random orientation of the crystallites or domains. As a neutron progresses through the sample, it encounters internal fields which are oriented in random directions relative to the external-field direction. Thus each crystallite or domain will have a finite component of internal field at right angles to the external field about which a new precession will occur. A classical treatment of the resulting depolarization is given by Halpern and Holstein.¹⁴ In this theory two general cases are considered: (1) The neutrons make a small part of one Larmor precession in each domain or (2) they make more than one. The first case leads to the following approximate expression of the depolarization parameter D :

$$D \approx (\mu_n/\hbar)^2 \langle B_{i\perp}^2 \delta^2 \rangle_{\text{av}} n/v^2, \quad (18)$$

where μ_n is the neutron magnetic moment, $B_{i\perp}$ is the component of the induction B_i within domain i perpendicular to the original beam polarization, δ is the domain size, n is the number of domains per unit length along the neutron flight path, and v is the neutron velocity. If the neutrons perform more than one Larmor precession in each domain, the depolarization is approximately given by

$$D \approx \langle B_{i\perp}^2/B_i^2 \rangle_{\text{av}} n. \quad (19)$$

The obvious difference, which one should expect, between these two equations is that the former is energy dependent, i.e., D is inversely proportional to the neu-

¹² J. M. Baker and B. Bleaney, Proc. Roy. Soc. (London) **A245**, 156 (1958).

¹³ R. J. Elliott and K. W. H. Stevens, Proc. Roy. Soc. (London) **A218**, 553 (1953).

¹⁴ Q. Halpern and T. Holstein, Phys. Rev. **59**, 960 (1941).

tron energy, while the latter is energy independent. The theory of Halpern and Holstein is only approximately correct, since it neglects atomic periodicities in the induction within the domain and the demagnetization fields.

B. Experimental Results

The beam polarizations, f_n^0 with the sample out of the beam and f_n with the sample in, were measured by means of a Bragg reflection from a second Co-Fe crystal. The reflected intensity decreases rapidly with neutron energy and, hence, the depolarization can only be studied in the range from thermal energies up to ~ 0.5 ev. Consequently, it is necessary to obtain the depolarization at the Ho resonances (3.92 and 12.8 ev) from an extrapolation of data taken at much lower energies.

Measurements of the depolarization were done at 0.062 ev for two foils of holmium metal, 0.032- and 0.062-cm thick. These foils at a temperature of 0.95°K were magnetized in a field of 17.9 koe. At 0.062 ev the neutron cross section is small for holmium. The values of D obtained for both foils were the same within the experimental errors, the average value of D being $17.3 \pm 0.9 \text{ cm}^{-1}$. The depolarization of the thicker sample was measured at various neutron energies. It turned out that the observed depolarization depends strongly on the neutron energy E as is indicated in Fig. 1. For the highest energies $\ln(f_n/f_n^0)$ tends to vary inversely with E . This variation is in agreement with formula (18). The linear extrapolation of the highest energy data gave $D = 1.67/E \text{ cm}^{-1}$, where E is in electron volts. An estimate of the domain size δ can be obtained from these depolarization measurements with the aid of formula (18). In the domain i the induction $\mathbf{B}_i = \mathbf{H}_0 + 4\pi\mathbf{M}_i \approx 38 \text{ koe}$ is obtained from the magnetization data of a holmium single crystal,¹⁵ in which we have made the approximation that the magnetization of a domain is confined to an easy direction in the basal plane, which is at random in the polycrystalline sample. The averaged value of B_{it}^2 is then approximately $0.61B_i^2$. Under these conditions, the domain size is found to be $\sim 0.4 \times 10^{-4} \text{ cm}$.^{15a}

The depolarization in the holmium-ethyl-sulfate sample, at a temperature of 0.95°K , was measured at 0.062, 0.117, and 0.267 ev as a function of the applied magnetic field H_0 . The ratio f_n/f_n^0 is plotted in Fig. 2 as a function of H_0 . As expected, it was found that the depolarization decreased when H_0 was increased. The ratio f_n/f_n^0 is, within the errors, the same at 0.062 and 0.117 ev, while it was found that at 0.267 ev the depolarization is somewhat less. These experimental data indicate, that the depolarization is negligible at the two Ho resonances of 3.92 and 12.8 ev if an external field of 10 koe or more is applied.

¹⁵ B. L. Rhodes, S. Legvold, and F. H. Spedding, Phys. Rev. **109**, 1547 (1958).

^{15a} Note added in proof. The average grain size in this sample was $4.5 \times 10^{-2} \text{ cm}$; thus, there are approximately 10^9 domains per grain.

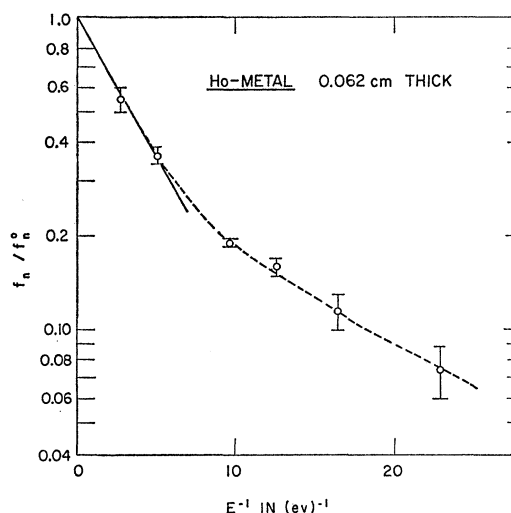


Fig. 1. The ratio f_n/f_n^0 , measured for holmium metal at various neutron energies. All the data were taken for an external field of 17.9 koe and at a temperature of 0.95°K .

The large depolarization in the polycrystalline sample of holmium-ethyl sulfate must be explained primarily in terms of the internal fields of the crystallites. However, it is not possible to explain the data with the aid of formulas (18) and (19) with reasonable values of the internal field. Probably this sample constitutes a case which falls between the two extremes expressed by formulas (18) and (19).

VII. TRANSMISSION EFFECTS AT THE RESONANCES

The transmission of unpolarized neutrons, measured as a function of neutron energy for the polycrystalline sample of holmium ethyl sulfate, is shown in Fig. 3. The two resonances at 3.92 and 12.8 ev were clearly resolved.

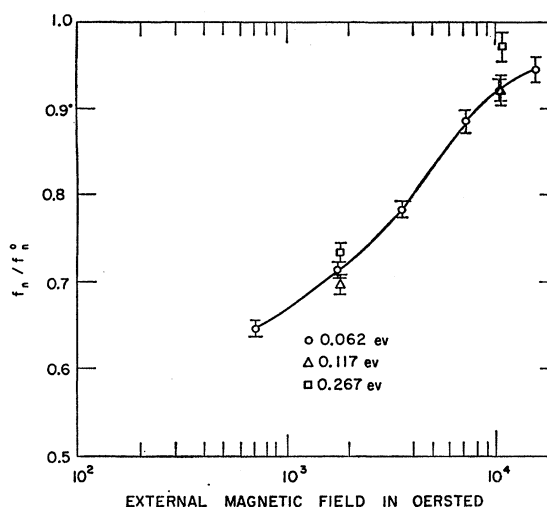


Fig. 2. The ratio f_n/f_n^0 , measured for holmium-ethyl sulfate as a function of the external field. The data were taken for three neutron energies at a temperature of 0.95°K .

TABLE I. Transmissions and transmission effects for the holmium resonances. Columns 3 and 6 list the experimental values. Column 5 lists the transmission calculated from Eq. (11), which takes account of the spectrometer resolution. The good agreement between columns 3 and 5 gives confidence to the values of p which we derived with the aid of Eq. (12) from the $\langle \epsilon \rangle$ measurements listed in column 6.

Neutron energy (ev)	Sample	$\langle T \rangle_{\text{meas}}$	$N\sigma_0 t$	$\langle T \rangle_{\text{calc}}$	$\langle \epsilon \rangle_{\text{meas}}$	J
3.92	Ho-ethyl sulfate	0.62 ± 0.02	9.30	0.648	-0.040 ± 0.003	4
	Ho metal	0.378 ± 0.015	42.9	0.386	-0.079 ± 0.005	
12.8	Ho-ethyl sulfate	0.61 ± 0.06			-0.065 ± 0.028	4
	Ho metal	0.50 ± 0.05			-0.052 ± 0.017	

The resonance at 3.92 ev was found with neutrons reflected from either (111) or (220) planes of the cobalt-iron crystal. The 12.8 ev resonance could only be observed with the (220) reflection, because the resolution was too poor with the (111). In addition, with Co(111) the Bragg angle corresponding to 12.8 ev is only $1^\circ 7'$ which results in a very large background from the splatter of the unreflected beam.

At these resonances, the reflected beam is not monoenergetic due to the finite resolution of the spectrometer [see Eq. (10)]. At 3.92 ev the width of the resolution function was $\Delta E = 0.970$, and 0.594 ev for the (111) and (220) reflections, compared to the natural width¹⁶ of the resonance, $\Gamma = 0.087$ ev. Thus, Eqs. (11) and (12) must be used in the analysis of the measured values of $\langle T \rangle$ and $\langle \epsilon \rangle$. In computing the resolution-corrected values of $\langle T \rangle$ and $\langle \epsilon \rangle$ with Eqs. (11) and (12), the Doppler-broadened cross section must be used.¹⁷ In a

solid, the Doppler broadening depends on an effective temperature T_{eff} rather than the actual temperature. In the limit of very low temperatures,¹⁸ $T_{\text{eff}} \approx \frac{1}{3}\Theta$ where Θ is the Debye temperature, which for Ho metal¹⁹ was taken to be 161°K. Calculated transmissions listed in column 5 of Table I are seen to be in reasonable agreement with the observed transmissions which are listed in column 3.

The transmissions measured for holmium-ethyl sulfate must be corrected for the attenuation of the beam due to the presence of the ethyl-sulfate groups and the heavy-water molecules in the sample. The attenuation due to these components was measured at 2.42 and 2.38 ev, where the cross section of holmium is relatively small. The measurements yield the values 0.508 ± 0.010 and 0.492 ± 0.010 for the (111) and (220) reflections, respectively. As expected, these transmissions are equal to within the statistical uncertainty. The experimental value is in reasonable agreement with an estimate obtained by adding cross sections of the nuclei in the ethyl-sulfate groups and the heavy water, which yields the value of 0.530.

In order to obtain the transmission-effects $\langle \epsilon \rangle$, as defined in Sec. IV, we measured the intensity of the transmitted beam polarized alternately parallel and antiparallel to the external field H_0 . The external field was 10 koe in case of holmium-ethyl sulfate and 17.9 koe for the holmium metal. Background intensities were determined by rotating the monochromating crystal 2° off the Bragg angle. The values obtained for $\langle \epsilon \rangle$ are listed in column 6 of Table I. From the values of $\langle \epsilon \rangle$ listed in the table, some conclusions can be made directly. Both resonances showed transmission effects of the same sign. Thus, the spins of the two resonances are the same. Since the Ho nuclei are polarized parallel to the applied field, the spins²⁰ must be $J = I + \frac{1}{2} = 4$.

To obtain the value of $p[(7/9)f_N]$ corresponding to the measured value of $\langle \epsilon \rangle$, we have computed a

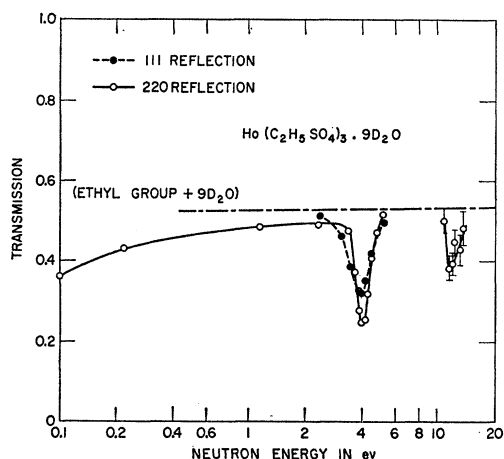


FIG. 3. Transmission of holmium-ethyl sulfate as a function of neutron energy. The sample thickness was $Nt = 0.994 \times 10^{21}$ holmium nuclei per cm^2 . The Co(220) reflection gives a lower transmission than the Co(111) at the 3.92-ev resonance since it gives better resolution. The dot-dashed line gives the calculated transmission of the ethyl sulfate and D_2O components of the sample.

¹⁶ Parameters for the 3.92-ev resonance in Ho^{165} were measured by R. I. Schermer (unpublished). The parameters are: $E_0 = 3.924 \pm 0.010$ ev; $\sigma_0 = 9360 \pm 200$ b; $\Gamma = (87 \pm 2) \times 10^{-3}$ ev; $\Gamma_n = (2.2 \pm 0.1) \times 10^{-3}$ ev; $\Gamma_\gamma = (85 \pm 2) \times 10^{-3}$ ev.

¹⁷ A recent discussion of Doppler broadening of neutron resonances may be found in the paper Br. A. Bernabei, L. B. Borst, and V. L. Sailor, Nuclear Sci. and Eng. 12, 63 (1962).

¹⁸ W. E. Lamb, Jr., Phys. Rev. 55, 110 (1939).

¹⁹ B. C. Gerstein, M. Griffel, L. D. Jennings, R. E. Miller, R. E. Skochdopole, and F. H. Spedding, J. Chem. Phys. 27, 394 (1957).

²⁰ Due to an error made during the tune-up period of the equipment, the spins of these resonances were erroneously reported to be $J = I - \frac{1}{2} = 3$. The values for both resonances listed in the paper, H. Postma, H. Marshak, V. L. Sailor, F. J. Shore, and C. A. Reynolds, Bull. Am. Phys. Soc. 6, 275 (1961), should be corrected to read $J = I + \frac{1}{2} = 4$.

family of curves of $\langle \mathcal{E} \rangle$ as a function of p and $N\sigma_0 t$ for the 3.92-ev resonance. The resonance parameters given in reference 16 were used in the calculations, as well as the constants $\Delta E = 0.970$ ev, $f_n^0 = 0.95$, and $\phi = 1$. For holmium-ethyl sulfate $Dt = 0$, while for the polycrystalline metal the value $Dt = 0.058$ was obtained from a linear extrapolation of the data in Fig. 1. The theoretical relationships between $\langle \mathcal{E} \rangle$ and p as calculated for the two samples are shown in Fig. 4. Also the experimental values of $\langle \mathcal{E} \rangle$ are given in this figure.

For the holmium-ethyl sulfate, the experimental value of $\langle \mathcal{E} \rangle$ with the aid of the computed curve yields the result $p = 0.168 \pm 0.013$. From this, the hyperfine splitting $A/k = 0.61 \pm 0.06^\circ\text{K}$ is obtained.

In order to calculate the hyperfine splitting constant for the metal, it is necessary to know the value of M/M_∞ . From a magnetization curve of polycrystalline holmium, as given by Henry,²¹ the magnetization at 17.9 koe is estimated to be 0.80 of saturation; however, unpublished data on our sample indicate that 0.66 is a more realistic value.^{21a} This value is corrected for a small demagnetization field. The apparent-saturation magnetization of a polycrystalline sample is approximately $\frac{3}{4}$ that of a single crystal, if one assumes that the magnetization of a domain is confined to one of the axes of easy magnetization in the basal plane, which is randomly oriented. Hence we can conclude, that $M/M_\infty \approx 0.49$. From the experimental value of $\langle \mathcal{E} \rangle$ we derived $p = 0.177 \pm 0.013$. Using the estimated value of M/M_∞ , we finally obtain the hyperfine splitting constant $A/k = 0.65 \pm 0.08^\circ\text{K}$.

VIII. CONCLUSIONS

One of the objectives of spin assignments of resonance levels is to determine whether there are relationships between the spins and other resonance parameters, especially the radiation width Γ_γ . At the moment, such a comparison is not possible in the case of Ho^{166} , since the resonance parameters have not been measured accurately for the 12.8-ev resonance. On the basis of the multiplicity of capture gamma rays, Draper and Springer²² suggested that the spin of the 3.92-ev resonance might be $J = I - \frac{1}{2} = 3$. This is in disagreement

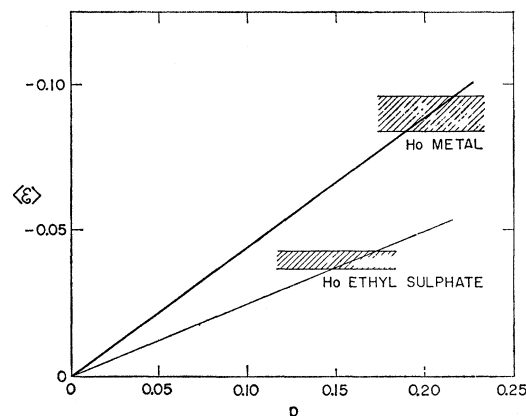


FIG. 4. The transmission effects $\langle \mathcal{E} \rangle$ versus $p [= (7/9)f_N]$ for the two samples of holmium-ethyl sulfate and holmium metal. The shaded regions give the experimental values of $\langle \mathcal{E} \rangle$.

with our experimental result. However, it is unlikely that one can distinguish between the two spin states on the basis of gamma multiplicities.

The hyperfine splitting constants, $A/k = 0.61 \pm 0.06$ and $0.65 \pm 0.08^\circ\text{K}$, obtained for concentrated holmium-ethyl sulfate and holmium metal are equal within the errors. The specific heat measurements⁸ on holmium metal gave the value of $A/k = 0.618^\circ\text{K}$. Apparently, the hyperfine splitting constant is larger for the concentrated holmium-ethyl-sulfate and the metal as compared to those of diluted-holmium-ethyl sulfate and diluted-holmium chloride.^{6,23} From the g_{11} values of the diluted salts it has been concluded that the $|\pm 7\rangle$ ground state doublet of Ho^{3+} is admixed with $|\pm 1\rangle$ and $|\pm 5\rangle$.¹² The larger values of A/k can be understood if the ground state of holmium is a $|\pm 7\rangle$ or a $|\pm 8\rangle$ doublet, instead of the given mixture. Since A depends linearly on g_{11} in the case of the rare earth ions, the values of A/k expected for these states are 0.55 and 0.62°K . The latter agrees best with the experimental values.

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²¹ W. E. Henry, Phys. Rev. **117**, 89 (1960).

^{21a} Note added in proof. Justification for this value is contained in V. L. Sailor, R. I. Schermer, F. J. Shore, C. A. Reynolds, H. Marshak, and Hans Postma, Phys. Rev. (to be published).

²² J. E. Draper and T. E. Springer, Nuclear Phys. **16**, 27 (1960).

²³ W. Low, *Paramagnetic Resonance in Solids* (Supplement 2 of Solid-State Physics) (Academic Press Inc., New York and London, 1960), p. 130.