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## Absolute Determination of Spins of Neutron Resonances and the Hyperfine Coupling Constant in $\text{Er}^{167\frac{1}{2}}$

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The spins of the four lowest neutron resonances in  $\text{Er}^{167}$  have been determined by measuring the transmission of polarized neutrons through a sample of polarized erbium nuclei. The determination of the spins depends solely on the transmission effect and is independent of any assumption about the sign of the nuclear magnetic moment or the direction of the effective field at the target nuclei. The spins were found to be  $J=I+\frac{1}{2}=4$  for the 0.460-eV resonance and  $J=I-\frac{1}{2}=3$  for the resonances at 0.584, 6.10, and 9.6 eV. The nuclear polarization of the sample, obtained as function of the sample temperature at a fixed energy, was fitted to a theoretical curve, using the magnetic and electric hyperfine splitting constants as fitting parameters. The values found for the magnetic and electric hfs constants are, respectively,  $A/k = -0.085 \pm 0.0005^\circ\text{K}$  and  $P/k = -0.005 \pm 0.001^\circ\text{K}$ . Taking the nuclear magnetic moment of  $\text{Er}^{167}$  to be  $-0.56\mu_N$ , the corresponding effective magnetic field at the nucleus is  $7.26 \times 10^6 \text{Oe}$ .

### I. INTRODUCTION

Transmission measurements with polarized neutrons and targets of polarized nuclei offer a simple and straightforward way to determine the spins of low-energy neutron resonances. At low neutron energies, only  $s$ -wave-type interactions between neutrons and target nuclei need to be considered. Hence, a resonance in the slow-neutron cross section corresponds to the formation of a compound nucleus with a definite total angular momentum  $J$ , where  $J$  is limited to  $I \pm \frac{1}{2}$ ,  $I$  being the spin of the target nucleus.

While, in general, the sign of the nuclear polarization, i.e., the sign of the nuclear magnetic moment and the direction of the effective field at the nucleus, must be known in order to make an unambiguous determination of the resonant spin, it is possible in some cases to make an absolute determination, provided the neutron cross section contains at least two observable resonances of opposite spin states. Preliminary results<sup>1</sup> on erbium depended on the assumption of a negative nuclear magnetic moment and a positive effective magnetic hyperfine field. The present work makes use of the fact that the two lowest resonances in the neutron cross section of  $\text{Er}^{167}$  have opposite spin and

that, therefore, the spins can be determined directly from the measurements of the transmission effect without any additional assumptions.

Since erbium has a very large magnetic hyperfine field, large transmission effects could be obtained which allowed the determination of the magnitude and the sign not only of the magnetic hfs constant but also of the electric hfs constant.

### II. THEORY OF TRANSMISSION EXPERIMENTS

#### A. Absolute Spin Determination

The theory describing the interaction of polarized neutrons and polarized nuclei has been developed elsewhere in detail.<sup>2-4</sup> Therefore, it is only necessary to present some of the pertinent equations here.

In order to represent the measurements with polarized neutrons parallel and antiparallel to the spin of the target nuclei, it is convenient to define a quantity  $\langle \mathcal{E} \rangle$ , the transmission effect,

$$\langle \mathcal{E} \rangle = \frac{\mathcal{T}_P - \mathcal{T}_A}{\mathcal{T}_P + \mathcal{T}_A} = \frac{C_P - C_A}{C_P + C_A}, \quad (1)$$

where  $\mathcal{T}_P$ ,  $\mathcal{T}_A$  and  $C_P$ ,  $C_A$  are, respectively, the transmissions and counting rates for parallel and antiparallel combination of neutron and target spin.

It can be shown<sup>2</sup> that the transmission for the parrallel and antiparrallel spin interaction, respectively, is given by

$$\tau_P = e^{-\alpha t} [\cosh kt - (\tau f_n^0 - \nu) \sinh kt], \quad (2a)$$

$$\tau_A = e^{-\alpha t} [\cosh kt + (\tau \varphi f_n^0 - \nu) \sinh kt]. \quad (2b)$$

In these equations  $f_n^0$  is the polarization of the neutron beam;  $\varphi$  is the efficiency of neutron spin reversal;  $\alpha = N\sigma + D$ , where  $N$  is the number of target nuclei per  $\text{cm}^3$ ,  $\sigma$  the total neutron cross section, and  $D$  the depolarization factor ( $D^{-1}$  is the mean free path for spin reversal);  $\kappa = (N^2\sigma^2\rho^2 + D^2)^{1/2}$ , where  $p = \rho f_N$ , where  $f_N$  is the nuclear polarization and  $\rho$  is a statistical weighting factor which has the values  $-1$  or  $I/(I+1)$  for  $J = I - \frac{1}{2}$  or  $J = I + \frac{1}{2}$ , respectively;  $\tau = N\sigma p/\kappa$ ; and finally  $\nu = D/\kappa$ .

Combining Eqs. (1), (2a), and (2b), one gets

$$\langle \mathcal{E} \rangle = -\frac{1}{2}(1 + \varphi) f_n^0 \frac{\tau \sinh kt}{\cosh kt + [\nu - \frac{1}{2}(1 - \varphi) \tau f_n^0] \sinh kt}. \quad (3)$$

If  $D = 0$  and  $\varphi \approx 1$ , and if  $f_N$  is small, Eq. (3) reduces to

$$\langle \mathcal{E} \rangle = -\frac{1}{2}(1 + \varphi) f_n^0 \tanh(N\sigma p f_N t). \quad (4a)$$

Although only an approximation, Eq. (4a) may serve to illustrate the main points of the experimental method. For thin targets, Eq. (4a) can be further reduced to

$$\langle \mathcal{E} \rangle \approx -\frac{1}{2}(1 + \varphi) f_n^0 (N\sigma p f_N t). \quad (4b)$$

Equation (4b) shows that the sign of  $\langle \mathcal{E} \rangle$  is determined by the weighting factor  $\rho$  if the sign of  $f_N$ , the nuclear polarization, is known. If the magnitude of  $f_N$  is also known, a measurement of the

magnitude of  $\langle \mathcal{E} \rangle$  will determine the value of  $\rho$  and, therefore, the spin state  $J$ . However, more often than not there is doubt about the sign of  $f_N$  as well as its magnitude.

If there are two closely spaced resonances,  $a$  and  $b$ , the spin states can easily be found by measuring the quantity  $\langle \mathcal{E} \rangle_a / \langle \mathcal{E} \rangle_b$ , where  $\langle \mathcal{E} \rangle_a$  and  $\langle \mathcal{E} \rangle_b$  are the measured transmission effects at the resonances  $a$  and  $b$  obtained under identical conditions, i.e., the same sample temperature and the same external magnetic field.

In this case

$$\frac{\langle \mathcal{E} \rangle_a}{\langle \mathcal{E} \rangle_b} = \frac{\rho_a (N\sigma t)_a}{\rho_b (N\sigma t)_b}. \quad (5)$$

The values for  $(N\sigma t)_{a,b}$  can be found easily from transmission measurements with unpolarized neutrons and unpolarized targets. Since the resonances  $a$  and  $b$  have opposite spin values, and the possible  $\rho$  values are  $-1$  and  $I/(I+1)$ , the quantity  $\langle \mathcal{E} \rangle_a / \langle \mathcal{E} \rangle_b$  can have only one of the two values  $-I/(I+1)$  or  $-(I+1)/I$  if we neglect, for the moment, the factor arising from the  $(N\sigma t)_{a,b}$  values.

Similar simplified equations were used successfully in the case of terbium,<sup>5</sup> where the measured transmission effects were relatively small; however, in the general case, and in particular for large transmission effects, i.e., whenever the hyperbolic functions cannot be approximated by their arguments, the exact equations must be used. Furthermore, Eq. (3) must be extended to take into account the finite energy resolution of the neutron spectrometer and the Doppler broadening of the resonant cross section. Equation (3) then becomes

$$\langle \mathcal{E} \rangle = -\left(\frac{1 - \varphi}{2}\right) f_n^0 \frac{\int R(E - E') \tau e^{-\alpha t} \sinh(\kappa t) dE'}{\int R(E - E') e^{-\alpha t} \{ \cosh(\kappa t) + [\nu - \frac{1}{2}(1 - \varphi) \tau f_n^0] \sinh(\kappa t) \} dE'}, \quad (6)$$

where  $R(E - E')$  is the spectrometer resolution function,<sup>6</sup> and where the cross section  $\sigma$  is calculated using the Doppler-broadened Breit-Wigner formula. Equation (5) has to be extended in the same way,  $\langle \mathcal{E} \rangle_a / \langle \mathcal{E} \rangle_b$  becoming the ratio of two expressions calculated using Eq. (6). For large values of  $\kappa t$  in Eq. (6), this ratio becomes a function of  $f_N$  and, therefore, of the sample temperature  $T$ . The  $\rho$  ratio also depends on the instrumental resolution, the more so the further apart the energies  $a$  and  $b$  are.

### B. Hyperfine Interactions

Once the  $\rho$  value, i.e.,  $J$ , is known one can ob-

tain a plot of  $f_N$  versus  $T$  by using the measured dependence of  $\langle \mathcal{E} \rangle$  as function of  $T$  and the use of Eq. (6). This information can be used to determine the hyperfine-splitting constants. If we describe the nuclear part of the hyperfine interaction by the spin Hamiltonian

$$\mathcal{H} = A S_z I_z + P [I_z^2 - \frac{1}{3} I(I+1)], \quad (7)$$

where  $A$  and  $P$  are the magnetic and electric splitting constant, respectively, the nuclear polarization is given by

$$f_N = \frac{1}{I} \frac{\sum m \exp[-(\frac{1}{2} A m + P m^2)/kT]}{\sum \exp[-(\frac{1}{2} A m + P m^2)/kT]}, \quad (8)$$

For  $P = 0$ , Eq. (8) reduces to the Brillouin function

$$f_N = B_I(\beta) = \frac{2I+1}{2I} \coth\left(\frac{2I+1}{2}\beta\right) - \frac{1}{2I} \coth\left(\frac{\beta}{2}\right) \quad (9)$$

with  $\beta = A/(2kT)$ . Unless single crystals and relatively high external magnetic fields are used, most samples will be only partially magnetized and Eqs. (8) and (9) must include a constant  $k_m$  to take account of this incomplete magnetization. We have then

$$f'_N = k_m f_N, \quad (10)$$

where  $f_N$  is given by Eq. (8) or (9).

### III. APPARATUS AND SAMPLE

The equipment used in the experiment consisted of a crystal spectrometer to obtain monochromatic polarized neutrons by Bragg reflection off the (111) and (220) planes of a magnetized Co-Fe crystal, and a demagnetization cryostat for cooling of the nuclear sample. For any details, the reader is referred to previous descriptions.<sup>2,4</sup>

Two different samples were used, both cut from the same metal ingot obtained from the Lindsay Chemical Company. The purity of the metal is stated to be 99.9%. The samples were cut as rectangular slabs 0.005 and 0.014 in. thick. Special attention was given to the mounting of the sample to assure a good thermal connection to the cooling salt.<sup>7</sup>

Since the neutron beam contains a certain fraction of second-order neutrons (higher orders can be neglected) which can greatly alter the transmission measurements, second-order resonance filters were used to reduce the second-order fraction of the beam to a negligibly small percentage. These filters consist of resonance absorbers that have strong isolated resonances at the second-order energy, while the transmission at the first-order energy is relatively high.

The cooling of the sample was accomplished by adiabatic demagnetization of an iron ammonium sulfate salt. The temperature was measured by measuring the susceptibility of the cooling salt. The susceptibility is directly proportional to the inverse of the magnetic temperature. The data of Cooke, Meyer, and Wolf<sup>8</sup> were used to convert the magnetic temperature to the thermodynamic temperature.

### IV. EXPERIMENTAL RESULTS

Figure 1 shows the neutron transmission of both samples used, 0.005 and 0.014 in. thick, over the energy range of the first two resonances. The measured points were obtained using In, Dy, Re, Te, Hf, and Gd as resonance second-order beam

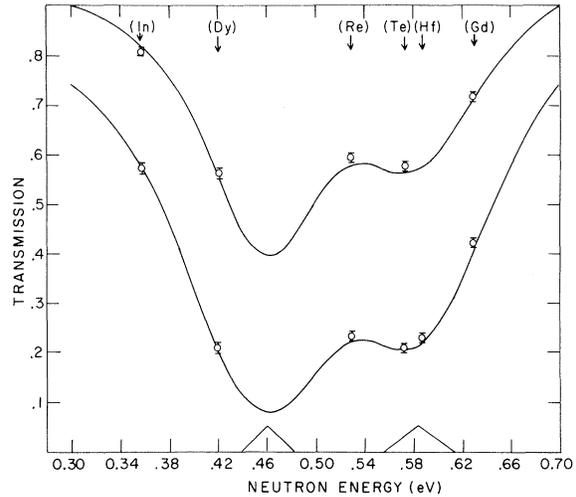


FIG. 1. Neutron transmission for 0.014- and 0.005-in. samples of natural erbium metal. Curves are calculated using the parameters from Ref. 9. Second-order filters used are indicated at top.

filters. The solid lines are the calculated transmission for unpolarized neutrons. The agreement is very good and the measurements serve as a sensitive check of the resonance parameters used,<sup>9</sup> the sample thickness, and the effectiveness of the second-order filters.

Erbium metal exhibits a strong magnetic anisotropy<sup>10,11</sup> which has a depolarizing effect on the neutron beam. This depolarization varies with energy and, therefore, does not cancel in Eq. (5). Figure 2 shows the depolarization factor  $D$  as function of energy.  $D$  was measured in a double-scattering experiment, using a second magnetized Co-Fe crystal as a second Bragg reflector. If  $f_n^0$  is the polarization of the neutron beam and  $f_n$  the polarization of the beam after transmission through the sample, then<sup>2</sup>

$$f_n = f_n^0 e^{(-2Dt)}, \quad (11)$$

where  $D$  is the depolarization factor and  $t$  the sample thickness.

The factor  $D$  is also needed in Eq. (6) in order to calculate the transmission effect as a function of energy.

Figure 3 shows the transmission effect measured at the six different energies for which second-order filters were available. A first inspection of the data shows that the spin states of the two first resonances must be opposite, which immediately eliminates two  $\rho$ -value combinations. The two solid lines in Fig. 3 are calculated using Eq. (6) representing the two possible cases of  $J$  values, i.e.,  $I + \frac{1}{2}$ ,  $I - \frac{1}{2}$ , and  $I - \frac{1}{2}$ ,  $I + \frac{1}{2}$ , for the first two resonances. As we have made no assumption

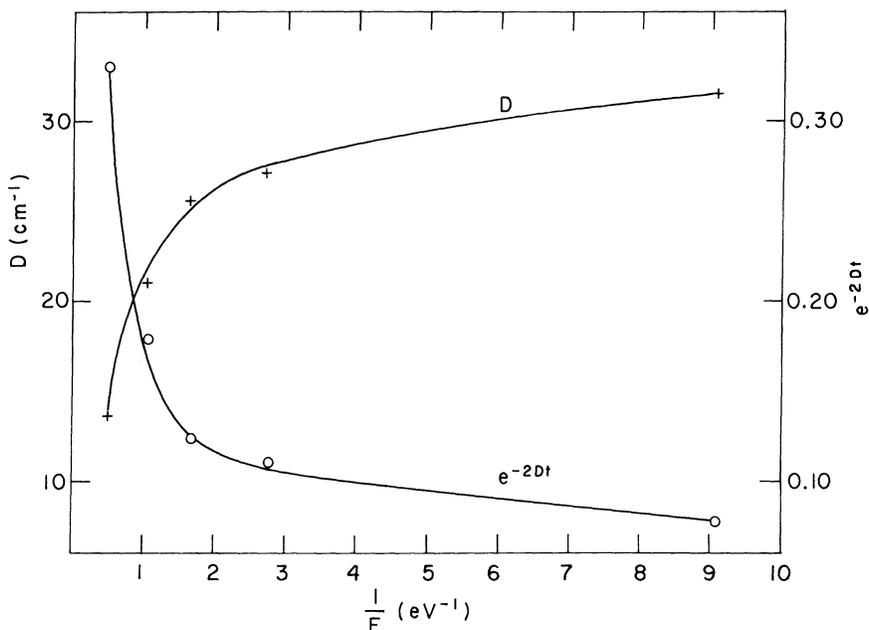


FIG. 2. Depolarization factor  $D$  as function of energy, as determined from the ratio of neutron beam polarization  $f_n/f_n^0 = e^{-2Dt}$ .

about the sign of either the nuclear magnetic moment or the direction of the effective magnetic field at the nucleus, the reflections of the calculated curves about the  $\langle \mathcal{E} \rangle = 0$  axis are equally possible.

If we now form the ratio  $R$  of the calculated  $\langle \mathcal{E} \rangle$  at  $E = 0.4278$  eV and  $E = 0.5838$  eV, two points at which the transmission effect has been measured, we find  $R_I = -1.099$  for curve I and  $R_{II} = -2.094$  for curve II. The ratio of the experimental points is

$R_{\text{exp}} = -1.044 \pm 0.03$ . The experimental results, therefore, are consistent only with the spin assignment of curve I, i.e.,  $J = I + \frac{1}{2} = 4$  for the 0.460-eV resonance and  $J = I - \frac{1}{2} = 3$  for the 0.584-eV resonance. Transmission effects of  $\langle \mathcal{E} \rangle = -3.5\%$  and  $\langle \mathcal{E} \rangle = -1.9\%$  were measured at the 6.10- and 9.6-eV resonances, respectively. The negative sign of the measured effect determines immediately the compound spin of both resonances to be also  $J = I - \frac{1}{2} = 3$ . In order to determine the hyperfine coupling con-

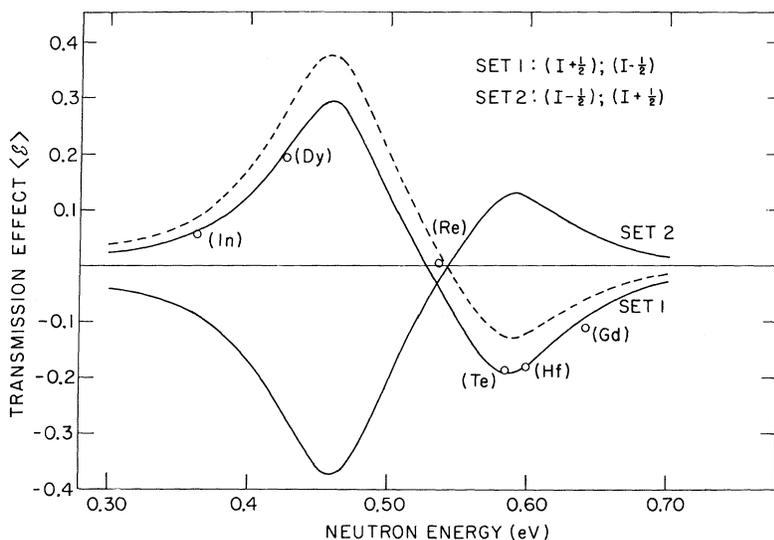


FIG. 3. Transmission effect as function of energy. The solid curves are calculated using Eq. (6). Sets 1 and 2 refer to the two possible combinations of  $\rho$  values. The dashed curve is the reflected curve for Set 2. The second-order filters used are indicated in parenthesis.

stants, the transmission effect was measured as function of the sample temperature. The results are shown in Fig. 4. The upper curve (II) was obtained with the 0.014-in. sample at 0.428 eV, while the lower curve (I) gives the data obtained with the 0.005-in. sample at 0.584 eV. The ordinate is the nuclear polarization  $f_N$  which is obtained from  $\langle \mathcal{E} \rangle$  in the manner described in Sec. II B. The difference in magnitude of  $f_N$  for the two samples comes from the factor  $k_m$  in Eq. (10). At the external magnetic fields used here (17 kOe) erbium metal is only partially magnetized. The magnetization along the crystalline  $a$  and  $b$  axis shows a very abrupt change in this region<sup>11</sup> and we must expect appreciable differences in magnetization for our two samples even at constant external field, because of the geometry-dependent demagnetization fields as well as differences in the previous magnetic history of the sample.

We have fitted the data to Eq. (8) using a least-squares-fit method<sup>7</sup> and obtained the values  $A/k = -0.085 \pm 0.0005^\circ\text{K}$  and  $P/k = -0.005 \pm 0.001^\circ\text{K}$  for the thin sample. The values for the thick sample lie 5% lower. The thicker sample represents a greater heat load on the cooling sample, and since the temperature is measured indirectly through the susceptibility of the cooling salt, the possible temperature gradient is likely to be more significant. We have, therefore, not included the data obtained with the thick sample in our final analysis. If we arbitrarily let  $P/k = 0$  and fit our data to Eq. (9), the  $A/k$  value reduces to  $A/k = -0.075^\circ\text{K}$  as the best fitting value. However, the fit to the data be-

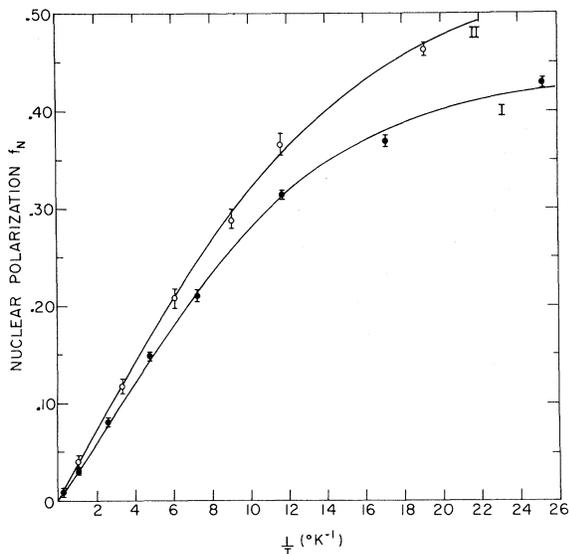


FIG. 4. Measured nuclear polarization as function of the inverse absolute temperature. The curves are calculated using Eq. (8).

TABLE I. Summary of spins determined in this work.

Neutron energy (eV)	$J$
0.460	4
0.584	3
6.10	3
9.6	3

comes decidedly poorer.

## V. DISCUSSION

The spins of four neutron resonances in  $\text{Er}^{167}$  have been determined absolutely. The determinations are independent of any assumption about the sign of the nuclear magnetic moment or the internal effective field. The results are summarized in Table I.

The hyperfine coupling constants are directly related to effective magnetic and electric fields at the site of the nucleus, and in the case of the rare earths these fields are determined mostly by the behavior of the  $4f$  electrons. The information that can be obtained from the magnetic coupling constant is essentially the product  $\mu H_{\text{eff}}$ . We will assume here that we can get an accurate value for the nuclear magnetic moment (n.m.m.)  $\mu$  from other measurements and will, therefore, be able to measure the effective field  $H_{\text{eff}}$ .

Unfortunately, it is very difficult to obtain values for the rare-earth n.m.m.'s by direct measurements. The procedure has usually been to measure the hyperfine coupling by magnetic-resonance methods and use more or less accurate theoretical estimates of the hyperfine fields. These estimates, of course, depend largely on the assumptions

TABLE II. Summary of nuclear magnetic moments of  $\text{Er}^{167}$ . Values are in units of nuclear magnetons. p.r. means paramagnetic resonance and a.b.r. means atomic-beam resonance.

$\mu$	Method	Reference
0.11	p.r.	a
0.48	p.r.	b
0.50	p.r.	c
0.50	Hartree-Fock $\langle r^{-3} \rangle$	d
0.58	p.r.	e
-0.564	a.b.r.	f
0.563	p.r.	f
-0.5647	a.b.r.	g

<sup>a</sup>See Ref. 12.

<sup>b</sup>See Ref. 13.

<sup>c</sup>See Ref. 14.

<sup>d</sup>See Ref. 15.

<sup>e</sup>See Ref. 16.

<sup>f</sup>See Ref. 17.

<sup>g</sup>See Ref. 18.

TABLE III. Comparison of the hfs results of the present work with previously published values. All values of  $H_{eff}$  for which  $a'$  is also given have been calculated using  $\mu = -0.56\mu_N$ .

$a' = A/2k$ (°K)	$H_{eff}$ ( $10^6$ Oe)	Method	Reference
	7.60	Mössbauer effect	a
	7.71	Mössbauer effect	b
0.0473	8.10	specific heat	c
0.0547	9.34	specific heat	d
-0.0451	7.72	calc. from	e
0.0452	7.70	electron-spin res.	
0.0456	7.79	calc. from electron-spin res.	f
	8.4	calc. $\langle r^{-3} \rangle_{eff}$	g
-0.0425	7.26	polarized neutron transmission	present work

<sup>a</sup>See Ref. 19.

<sup>b</sup>See Ref. 20.

<sup>c</sup>See Ref. 21.

<sup>d</sup>See Ref. 22.

<sup>e</sup>See Ref. 23.

<sup>f</sup>See Ref. 24.

<sup>g</sup>See Ref. 25.

about the  $4f$  electron wave functions, and calculations by different authors differ by as much as 25%.

Table II summarizes some available data for the n.m.m. of  $\text{Er}^{167}$ . All of the older data<sup>12-15</sup> are obtained from paramagnetic-resonance measurements, using various assumptions to calculate  $\langle r^{-3} \rangle$ . The results of Bleaney,<sup>13</sup> Elliot and Stevens,<sup>14</sup> and those of Freeman and Watson<sup>15</sup> agree very well, although the courses of their calculations are quite different. There is no agreement with the value of Hutchison and Wong,<sup>12</sup> although their calculations reproduce their experimental results. In an effort to arrive at more realistic  $\langle r^{-3} \rangle$  values, Lindgren<sup>16</sup> has used  $4f$  electron wave functions that were adjusted to fit results on  $\text{Pr}^{3+}$  and  $\text{Tm}^{3+}$ . His  $\langle r^{-3} \rangle$ 's are about 20% smaller, and consequently the value for the n.m.m. is larger. Bleaney<sup>17</sup> has recently recalculated his earlier results<sup>13</sup> based on  $\langle r^{-3} \rangle$  values obtained by interpolating between some direct n.m.m. measurements. The new values are, not too surprisingly, in very good agreement with the most recent direct determination of the n.m.m. of  $\text{Er}^{167}$  obtained by atomic-beam triple magnetic resonance.<sup>18</sup> For our purposes, we will use the value of  $\mu = -0.56\mu_N$ .

There are several previous determinations of the hyperfine constant in erbium by the specific-heat method, as well as field determinations by the Mössbauer method, and calculations based on magnetic-resonance measurements. In order to compare the different results we have calculated

$H_{eff}$ , and  $a'$  where applicable, in all cases using the value  $\mu = -0.56\mu_N$  except in the Mössbauer determinations. Table III gives a summary of previous and present measurements. The Mössbauer measurements<sup>19,20</sup> were done by observing the splitting of the 80.6-keV transition in  $\text{Er}^{166}$ ; both measurements do observe a quadrupole interaction. The specific-heat measurements<sup>21,22</sup> were made down to 0.4 and 0.5°K, respectively. These temperatures were not low enough to allow an extraction of information on the quadrupole interaction from the data. Bleaney<sup>23</sup> estimated hyperfine interactions based on data from electron-spin resonance and electron-nuclear double-resonance measurements on salts and atomic beam measurements on free ions. He also included a quadrupole interaction of  $P/k = -0.003^\circ\text{K}$ . Kondo<sup>24</sup> calculated effective magnetic fields, using available electron-spin-resonance data. Cohen and Wernick<sup>25</sup> calculated an effective field using an effective  $\langle r^{-3} \rangle$  value obtained by extrapolation from  $\text{Tm}^{169}$  in order to interpret their Mössbauer data on  $\text{Er}^{166}$ .

While the specific-heat measurements lie somewhat higher, the agreement is, in general, good for all measurements listed.

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