

extrapolation that we have made, particularly in view of the close agreement (within about 2%) between the experimental and theoretical values of the linear term. Rigorously there always remains the experimental possibility of a transition, possibly accompanied by the appearance of superfluidity, at temperatures lower than those of our measurements; but we feel that the data presented here make this possibility now unlikely.

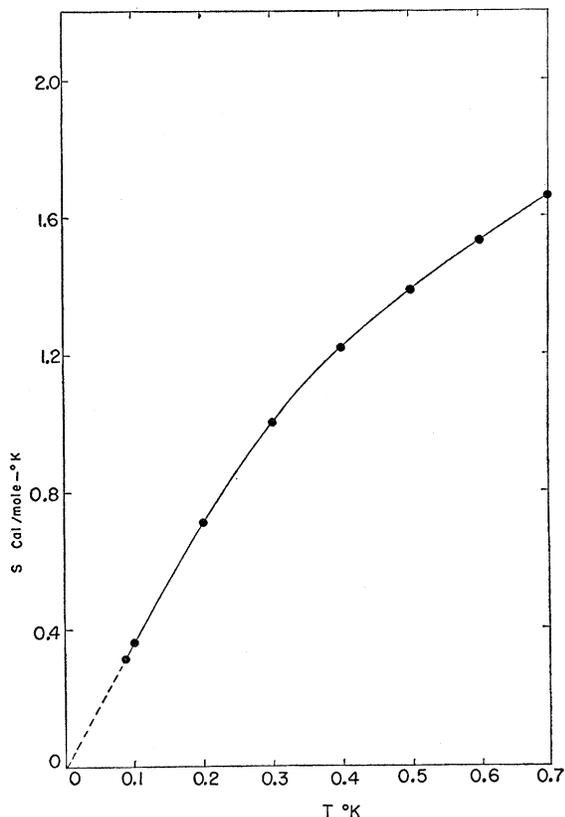


FIG. 2. Molar entropy of liquid He<sup>3</sup> at a pressure of 12–15 cm Hg, calculated from the present specific heat measurements by using a linear extrapolation from 0.085°K to 0°K.

It is generally agreed<sup>10</sup> that the condition leading to superfluidity in He<sup>4</sup> is the absence of low-lying energy states apart from the collective phonon excitations. It follows from the linear Fermi-type specific heat that in He<sup>3</sup> an abundance of low single-particle excitations exists down to 0°K, thus preventing the occurrence of superfluidity, a conclusion inherent in the original statistical approach of London.<sup>11</sup>

The marked deviation from an ideal Fermi function above about 0.2°K indicates a more complex character for the excitations at these higher temperatures. It is anticipated that further insight into the nature of the excitations will result from measurements on the liquid under pressure, which are now in progress. Further discussion is therefore postponed.

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## Electron Nuclear Double Resonance of Neodymium\*

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THE method of electron nuclear double resonance has recently been employed by Feher<sup>1</sup> for investigation of some solid state problems. In this letter we discuss the application of the method to an investigation of Nd<sup>3+</sup> in LaCl<sub>3</sub> crystals.

Both Nd<sup>143</sup> and Nd<sup>145</sup> have nuclear spin 7/2. The lowest crystal field state of Nd<sup>3+</sup> (*f*<sup>3</sup>, *I*<sub>9/2</sub>) in LaCl<sub>3</sub> is a Kramers doublet described approximately by<sup>2</sup>

$$0.941|\pm 7/2(-0.339)|\mp 5/2\rangle.$$

The energies of the hyperfine levels are therefore given by<sup>3</sup>

$$W(M, m) = -\frac{1}{2}hcA + hcP\{M^2 + m^2 + 2Mm - 5\} \\ \pm \frac{1}{2}\{[(M+m)hc(A-2P) + g_1\beta H]^2 \\ + (hcB)^2[16 - (M+m)^2]\}^{1/2} - g_N'\beta Hm, \quad (1)$$

for the case in which **H** is parallel to the *c* axis of the

LaCl<sub>3</sub> crystal.  $A$ ,  $B$ ,  $P$ , and  $g_{||}$  are parameters in the spin-Hamiltonian ( $S=\frac{1}{2}$ )

$$\mathcal{H} = \beta \{ g_{||} H_x S_x + g_{\perp} [H_x S_x + H_y S_y] \} + hc \{ A S_x I_x + B [S_x I_x + S_y I_y] + P [I_x^2 - \frac{1}{3} I(I+1)] \} - g_N' \beta_N \mathbf{H} \cdot \mathbf{I}. \quad (2)$$

It has been pointed out by Bleaney<sup>4</sup> that in the second-order perturbation theory treatment of the hyperfine patterns of paramagnetic resonance spectra, the cross-product term between the matrices of the magnetic perturbation,  $\beta \mathbf{H} \cdot (\mathbf{L} + 2\mathbf{S})$ , and of the nuclear spin perturbation,  $a \mathbf{I} \cdot \mathbf{J}$ , must be taken into account. This will be seen to introduce the term (notation of Elliott and Stevens<sup>5</sup>)

$$4\beta^2 \beta_N g_N H m \langle r^{-3} \rangle \langle J || A || J \rangle \langle J || N || J \rangle \sum_i \frac{\langle \varphi_j | J_k | \varphi_i \rangle}{W_j - W_i}$$

in the energy in the case in which only the crystal field states,  $\varphi_j$ , of the lowest multiplet  $^{2S+1}L_J$  are taken into account. This term has the same dependence on nuclear magnetic moment and on external field strength as does the usual term  $-g_N \beta_N H m$  representing the direct effect of the external field on the nucleus. For fixed orientation of the crystal the two are therefore experimentally indistinguishable. This effect is similar to the paramagnetic chemical shift of an ordinary nuclear resonance with the exception that the intervals  $W_j - W_i$  between the crystal field levels are relatively small and the apparent external field seen by the nucleus may be much larger than the actual external field.

We may write

$$g_N' = g_N \left( 1 + 4\beta^2 \langle r^{-3} \rangle \langle J || A || J \rangle \langle J || N || J \rangle \times \sum_i \frac{\langle \varphi_j | J_k | \varphi_i \rangle}{W_j - W_i} \right). \quad (3)$$

TABLE I. Frequencies of second resonance for Nd<sup>145</sup>. It is assumed that  $A$  is positive.

$m$	$ \mathbf{H} $ for normal paramagnetic resonance at 9218 Mc sec <sup>-1</sup> (gauss)	Frequency for second resonance (Mc sec <sup>-1</sup> )
-7/2	2146.7	412.17
		406.22
-5/2	1999.3	413.58
		406.97
		405.58
		...
+1/2	1570.0	393.56
		389.60
		388.80
		385.75
+3/2	1428.2	387.88
		384.64
		384.00
		384.00
		381.50

In our experiments the transitions

$$(\frac{1}{2}, m) \rightarrow (\frac{1}{2}, m \pm 1) \quad \text{and} \quad (-\frac{1}{2}, m) \rightarrow (-\frac{1}{2}, m \pm 1) \quad (4)$$

were induced by introducing a second field, with frequency matching these transitions, into the cavity in which normal paramagnetic resonance of the transition

$$(-\frac{1}{2}, m) \rightarrow (+\frac{1}{2}, m) \quad (5)$$

was occurring at approximately 9000 Mc sec<sup>-1</sup>. The second field was frequency-modulated at frequencies from  $1.25 \times 10^5$  to  $4 \times 10^2$  cycle sec<sup>-1</sup>. The ac component of the microwave carrier at this modulation frequency was detected by crystal diode or bolometer. When any of the transitions (4) occurred, populations of the nuclear states fluctuated at the modulation frequency, producing a signal. The second resonances have a width of  $\sim 1 \times 10^5$  cycle sec<sup>-1</sup>.

In Table I are given the frequencies so far observed at which the second resonance for Nd<sup>145</sup> occurs when the  $c$  axis of the crystal is parallel to  $H$ . The remainder of the measurements are in progress

$g_{||}$  and  $A$  have been previously measured<sup>2</sup> for Nd<sup>145</sup> and found to be 3.996 and 0.0264 cm<sup>-1</sup>, respectively. Using these values, the data in Table I are fit by (1), using the following values of the remaining parameters and assuming  $A$  to be positive:  $B = 0.01034 \pm 0.00002$  cm<sup>-1</sup>,  $g_{N||}' = -0.345 \pm 0.006$ , and  $P = (-1.27 \pm 0.30) \times 10^{-6}$  cm<sup>-1</sup>. Using the formula of Elliott and Stevens<sup>5</sup> and  $\langle r^{-3} \rangle = 42 \times 10^{24}$  cm<sup>-3</sup>, the quadrupole moment is given by

$$Q = 0.0104 \times 10^{-24} \text{ cm}^2.$$

The crystal field states of Nd<sup>145</sup> in LaCl<sub>3</sub> have previously been calculated<sup>3</sup> using the optical data of Sayre, Sancier, and Freed<sup>6</sup> for Pr<sup>143</sup> in LaCl<sub>3</sub> to obtain values of the crystal field parameters which were extrapolated to the case of Nd<sup>145</sup>. Using these states, we have calculated the values of  $g_{N||}'$  and  $g_{N\perp}'$ . We find

$$g_{N||}' = g_N (1 + \langle r^{-3} \rangle 3.946 \times 10^{-26}), \quad (6a)$$

$$g_{N\perp}' = g_N (1 + \langle r^{-3} \rangle 6.967 \times 10^{-26}). \quad (6b)$$

(The contribution of the  $J=11/2$  state is also taken into account in this calculation.) The experimental value of  $A$  for Nd<sup>145</sup> gives

$$g_N \langle r^{-3} \rangle = -7.600 \times 10^{24} \text{ cm}^{-3}, \quad (7)$$

not allowing for any  $s$  contribution. This value cannot be combined with (6a) to obtain reliable numerical values of  $g_N$  and  $\langle r^{-3} \rangle$  because of the uncertainty with respect to the amount of  $s$  contribution and the very great sensitivity of the numerical values to the assumed amount of this contribution. Taking  $42 \times 10^{24}$  cm<sup>-3</sup> as the value<sup>7</sup> of  $\langle r^{-3} \rangle$ , (6a) gives  $g_N = -0.130$  and (7) gives  $g_N = -0.181$ .

Measurements of  $g_{N\perp}'$  would enable us to find both  $g_N$  and  $\langle r^{-3} \rangle$  with good accuracy from (6a) and (6b). Extensive observations of second resonance in the per-

pendicular orientation have been made on both  $\text{Nd}^{146}$  and  $\text{Nd}^{148}$  and will be discussed in a complete paper on this subject. The main difficulty in the analysis of the data for the perpendicular orientation is in the fitting of the results to the spin-Hamiltonian (2). Formulas of fourth or higher order perturbation theory must be used to obtain sufficiently accurate values of the parameters inasmuch as, particularly in the case of  $\text{Nd}^{143}$ , the normal second- and third-order contributions to the hyperfine effects are very much larger than that coming from  $g_{N1}\beta_N\mathbf{H}\cdot\mathbf{I}$ .

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### Anomalous Behavior of $\text{Al}^{27}(p,\alpha)\text{Mg}^{24}$ Differential Cross Sections\*

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THE partial success of Butler's direct interaction theory<sup>1</sup> in fitting the experimental  $\text{C}^{12}(\alpha,p)\text{N}^{15}$  angular distributions suggested an investigation of  $(p,\alpha)$  reactions. The differential cross sections of  $\text{Al}^{27}(p,\alpha_0)\text{Mg}^{24}$  ( $Q=1.60$  Mev) and  $\text{Al}^{27}(p,\alpha_1)\text{Mg}^{24}$  ( $Q=-0.23$  Mev) have been measured for protons of approximately 11 Mev from the Brookhaven National Laboratory 60-inch cyclotron. Alpha-particle groups are separated from proton and deuteron groups by a  $(dE/dx)$  vs  $E$  proportional counter scintillation counter telescope. This counter, the scattering chamber, and associated equipment have been described elsewhere.<sup>2</sup> Beam energy and energy spread are determined by range measurements using the range curves of Aron, Hoffman, and Williams. The initial proton energy varies between 10.3 Mev and 11.0 Mev depending on cyclotron operation conditions, and the energy spread is approximately 200 kev. Lower proton energies are obtained by degrading the beam with aluminum absorbers.

Figure 1 shows the angular distributions measured for the  $\text{Al}(p,\alpha)$  reactions leading to the ground and first excited states of  $\text{Mg}^{24}$  respectively, both at 10.97 and 10.87 Mev incident proton energy. Figure 2 shows

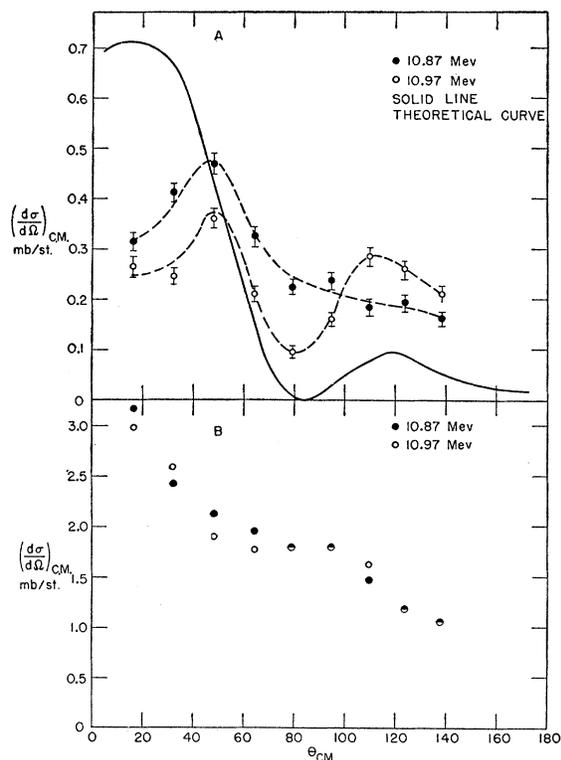


FIG. 1. (A) Angular distributions of alpha particles from  $\text{Al}^{27}(p,\alpha_0)\text{Mg}^{24}$  ( $Q=+1.60$  Mev) for 10.97-Mev and 10.87-Mev protons. Theoretical curve calculated from Butler's<sup>1</sup> Eq. (58) for  $r=4.50\times 10^{-13}$  cm and for 10.9-Mev protons.  $\mathbf{Q}=(23/27)\mathbf{k}_p - (23/24)\mathbf{k}_\alpha$ . (B) Angular distributions of alpha particles from  $\text{Al}^{27}(p,\alpha_1)\text{Mg}^{24}$  ( $Q=+0.23$  Mev) for 10.97-Mev and 10.87-Mev protons.

the differential cross sections at laboratory angle  $45^\circ$  for these reactions as well as for protons scattered elastically from aluminum, as a function of proton energy. The curves in Fig. 2 were obtained after a major cyclotron shutdown and it was not possible to bring the beam energy up to its previous value of 10.97 Mev. The estimated maximum experimental error is 15% for the differential cross sections. The absolute bombarding energy is believed to be known to 100 kev. Energy changes are known to 2%.

The theoretical curve in Fig. 1 was calculated from Eq. (58) of Butler's paper<sup>1</sup> for 10.9-Mev protons. The nuclear radius used,  $r=4.50\times 10^{-13}$  cm, was that which gave the best fit to the minimum and second maximum. It was not possible to reproduce the first maximum with a reasonable value for the radius. Butler's theoretical expression predicts less forward peaking than is observed for  $(\alpha,p)$  reactions.<sup>1,3</sup> Hunting and Wall<sup>3</sup> obtain a much improved fit to their  $(\alpha,p)$  data with the expression  $\exp(-Q^2/Q_0^2)|j_l(Qr)|^2$  for the differential cross section, taking the Fermi momentum into account. For  $\text{Al}^{27}(\alpha,p)\text{Si}^{30}$  they required a radius of  $4.98\times 10^{-13}$  cm for a fit to their data. However, for  $\text{Al}^{27}(p,\alpha_0)\text{Mg}^{24}$  The Butler theory predicts more forward peaking than