

The present results can also be compared with those of Thompson and Myers.³⁰ They assumed a band-independent exchange splitting, which was adjusted to yield the experimental value of D and then computed the entire spin-wave spectrum. They found that a band splitting of 0.91 eV was necessary to yield a correct value of D and (at the same time) 0.6 d holes per atom. The same difficulty concerning the spin splitting exists in both calculations.

V. FINAL REMARKS

We have applied a combined tight-binding and pseudopotential scheme to the calculation of the energy band structure of nickel. This procedure is different in some respects from similar procedures employed by others in that the overlap matrix is not assumed to be a unit matrix, and in that an attempt is made to calculate the tight-binding parameters in a more realistic manner. The resulting band structure is used in a t -matrix calculation to obtain the spin splitting of the highest d band and the spin-wave effective mass.

Numerous problems remain. We may not have dealt adequately with the complex system of overlapping bands that exist in nickel. For example, we have con-

sidered the bands to be defined strictly in order of increasing energy, and have not considered the complex problems which result from the crossing or the close approach of bands.⁵² We have ignored the existence of electronlike portions of the Fermi surface, have neglected bands above the d bands altogether; have treated the holes in nickel as a low-density system, although the validity of this is not established; and have not included spin-orbit coupling or any interaction between electrons on different lattice sites. The critical reader will undoubtedly raise additional objections of his own. However, we recall that the spin-wave reciprocal effective mass is the difference of two quantities; there is no guarantee that one will even obtain $D > 0$ (stability against spin-wave excitations). That the results are not unreasonable is perhaps an indication that the most essential features of the problem are understood. We believe that further work in this field will be rewarding.

ACKNOWLEDGMENTS

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⁵² J. Callaway and A. J. Hughes, Phys. Rev. **156**, 860 (1967).

Green's Function Theory of a Heisenberg Ferromagnet with Strong Dipole-Dipole Interactions: Magnetization of GdCl_3 [†]

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We have investigated the magnetization as a function of T and H of a spin system with both isotropic Heisenberg exchange and dipole-dipole interactions for $S \geq \frac{1}{2}$ and a hexagonal crystal structure. The Green's functions for such a system, recently derived by Becker, are decoupled in a first-order random-phase approximation. For both Heisenberg-dipole and simple-dipole crystals, we find a lowering of the magnetization below saturation at $T=0$. The Curie-Weiss temperature θ and the ordering temperature T_c are calculated and compared with experimental values on GdCl_3 . In the limit of zero exchange, we obtain a condition for the type of lattice and shape of domains which make ferromagnetic ordering possible.

I. INTRODUCTION

THE Heisenberg model has been analyzed with the technique of double-time thermodynamic Green's functions (GF) by several authors in various approximations.¹ In this model, the isotropic exchange interaction

is assumed to be between nearest magnetic neighbors only. In most real crystals there are, however, long-range interactions such as magnetic dipole-dipole interactions which can influence the thermodynamic properties quite remarkably. An example of such a system is the ferromagnetic insulator GdCl_3 where

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¹ For a review of the results see S. V. Tyablikov, *Methods in the Quantum Theory of Magnetism* (Plenum Press, Inc., New York, 1967), p. 247 ff.

isotropic exchange interactions and dipole-dipole interactions are of comparable magnitude.

GdCl₃ has been intensively studied experimentally and theoretically. Wolf *et al.*² measured the magnetization and the static susceptibility as a function of the applied field H in the temperature region of liquid helium and nitrogen and found a ferromagnetic phase transition at $T_c = 2.20^\circ\text{K}$. The magnetic specific heat was determined by Boyd and Wolf³ and Clover and Wolf⁴ using the high-frequency relaxation method. Because the Gd³⁺ ions are in orbital s states, crystal field effects are unimportant in GdCl₃. Indeed, Birgeneau and co-workers⁵⁻⁷ have shown in a series of papers that the resonance measurements on Gd³⁺ pairs in the isomorphous salts LaCl₃ and EuCl₃ can be completely explained in terms of an isotropic exchange between nearest and next-nearest neighbors and by dipole-dipole interactions. The exchange constants extrapolated from these measurements for GdCl₃ agree quite well with those derived from bulk measurements of the high-temperature specific heat of GdCl₃ by Clover and Wolf.⁴

The theoretical interest in GdCl₃ stems from the fact that various theoretical approximations may be tested by direct comparison with experiment because all terms in the Hamiltonian are well known and the structure is quite simple. Marquard and Stinchcombe⁸ calculated the unrenormalized spin-wave spectrum using an extension of the Holstein-Primakoff⁹ transformation theory. Marquard¹⁰ has given a high-temperature series expansion for the susceptibility and specific heat of GdCl₃. With the technique of two-time thermodynamic Green's functions, Becker,¹¹ henceforth referred to as I, calculated the frequency and wave-vector-dependent susceptibilities $X_{xx}(\omega, \mathbf{k})$ and $X_{yy}(\omega, \mathbf{k})$, and the spin-wave energies renormalized by the magnetization and the transverse-spin correlation.

In this paper, we use the random-phase form of the Green's functions, derived in I, to obtain the static magnetization $M(H, T)$, the critical temperature T_c and the high-temperature series expansion for the zero-field susceptibility of GdCl₃. The random-phase approximation (RPA) is used as a decoupling scheme for the two-time Green's functions,¹ which gives an over-all good description of the thermodynamic properties in the entire temperature region and in the entire external

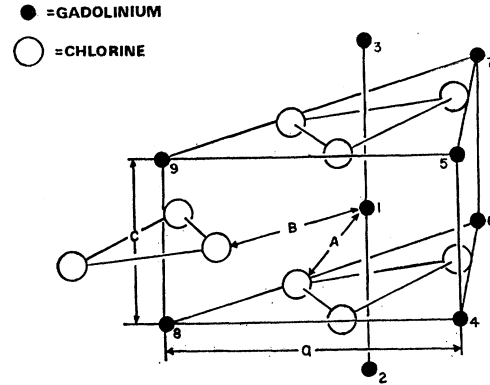


FIG. 1. Crystal structure of gadolinium trichloride. The ions 2 and 3 are the nearest neighbors of ion 1, the ions 4-9 are the next-nearest neighbors of ion 1.

magnetic field region. The high-temperature series expansion derived from the RPA Green's functions agrees with the leading terms of the exact expansion.¹ However, it is well known that RPA gives rise to an incorrect description of the spin-wave interactions at $T \ll T_c$ leading to a spurious $T^{3(S+1/2)}$ term in the magnetization. Also the spin dynamics in the critical region are obviously incorrect. Fortunately, for the high spin value $S = \frac{7}{2}$ of Gd³⁺ the spurious term in the low-temperature magnetization is of the order T^{12} and the thermodynamic properties at low temperatures are well described by RPA. Since the high- and low-temperature properties are correct to a high order it will be quite interesting to compare the critical temperature T_c calculated in RPA with the experimental value. The calculated thermodynamic properties are compared with those measured by Wolf *et al.*² For a crystal with pure dipole-dipole interaction, we calculate the same properties and also give a simple condition, valid in this approximation, for the type of lattice and shape of domains which make ferromagnetic ordering possible.

II. LATTICE STRUCTURE AND HAMILTONIAN

The lattice structure of GdCl₃ was determined by Zachariasen¹² and is shown in Fig. 1. The lattice is hexagonal with two Gd³⁺ ions in the unit cell. The two nearest Gd³⁺ neighbors are placed along the c axis at a distance $c = 4.105 \text{ \AA}$ and interact antiferromagnetically with the central ion.⁵⁻⁷ The six next-nearest neighbors are at a distance $b = 4.721 \text{ \AA}$ and at an angle $\theta = 64.2^\circ$ to the c axis. They interact ferromagnetically with the central ion. The Hamiltonian of this system is given by

$$\mathcal{H} = -\hbar\gamma H \sum_{i=1}^N S_i^z + \sum_{ij} I_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \frac{1}{2} \frac{(\hbar\gamma)^2}{4\pi\mu_0} \sum_{ij} \frac{1}{r_{ij}^3} \times \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{3(\mathbf{r}_{ij} \cdot \mathbf{S}_i)(\mathbf{r}_{ij} \cdot \mathbf{S}_j)}{r_{ij}^2} \right), \quad (1)$$

¹² W. H. Zachariasen, J. Chem. Phys. **16**, 254 (1948).

² W. P. Wolf, M. J. M. Leask, B. Mangum, and A. F. G. Wyatt, J. Phys. Soc. Japan **17**, 481 (1961).

³ E. L. Boyd and W. P. Wolf, J. Appl. Phys. **36**, 1027 (1965).

⁴ R. B. Clover and W. P. Wolf, Solid State Commun. **6**, 331 (1968).

⁵ R. J. Birgeneau, M. T. Hutchings, and W. P. Wolf, J. Appl. Phys. **38**, 957 (1967).

⁶ M. T. Hutchings, R. J. Birgeneau, and W. P. Wolf, Phys. Rev. **168**, 1026 (1968).

⁷ R. J. Birgeneau, M. T. Hutchings, and W. P. Wolf (unpublished).

⁸ C. D. Marquard and R. B. Stinchcombe Proc. Phys. Soc. (London) **92**, 665 (1967).

⁹ T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

¹⁰ C. D. Marquard, Proc. Phys. Soc. (London) **92**, 650 (1967).

¹¹ E. Becker Physik Kondensierten Materie **8**, 216 (1968).

where H is an external magnetic field parallel to the c axis, γ is the gyromagnetic ratio, \mathbf{r}_{ij} is the displacement vector between ion i and j , μ_0 is the vacuum permeability. Using the crystal symmetry of GdCl_3 , Eq. (1) may be reduced to

$$\mathcal{H} = -\hbar\gamma H \sum_{i=1}^N S_i^z - \frac{1}{2} \sum_{ij} (A_{ij} S_i^z S_j^z + B_{ij} S_i^+ S_j^-) + \frac{1}{2} \sum_{ij} (D_{ij}^+ S_i^- S_j^- + D_{ij}^- S_i^+ S_j^+), \quad (2)$$

where the coefficients are given in spherical coordinates r_{ij} , ϕ_{ij} , θ_{ij} by

$$\begin{aligned} A_{ij} &= 2(\alpha_{ij} - I_{ij}), \\ B_{ij} &= -(\alpha_{ij} + 2I_{ij}), \\ \alpha_{ij} &= -\frac{1}{2} \frac{(\hbar\gamma)^2}{4\pi\mu_0} \frac{1}{r_{ij}^3} (3 \cos^2 \theta_{ij} - 1), \\ D_{ij}^\mp &= -\frac{3}{4} \frac{(\hbar\gamma)^2}{4\pi\mu_0} \frac{1}{r_{ij}^3} \sin^2 \theta_{ij} e^{\mp 2i\phi_{ij}}. \end{aligned}$$

The last term in (2) does not commute with the Zeeman energy. Therefore the eigenstates of the Hamiltonian are not angular momentum states and the magnetization at $T=0$ will be lowered below the full saturation magnetization.

III. GREEN'S FUNCTIONS IN A RPA

The spectral density $\langle\langle A; B \rangle\rangle_{\hbar\omega}$ of the two-time thermodynamic Green's function (GF) of the operators A and B is related to the time-dependent correlation function $\langle BA(t) \rangle$ by¹³

$$\langle BA(t) \rangle = \lim_{\epsilon \rightarrow 0} i \int_{-\infty}^{\infty} d\omega \frac{\langle\langle A; B \rangle\rangle_{\hbar\omega+i\epsilon} - \langle\langle A; B \rangle\rangle_{\hbar\omega-i\epsilon}}{e^{\beta\hbar\omega} - 1} e^{-i\omega t}. \quad (3)$$

The spectral densities follow from their equation of motion¹³

$$\hbar\omega \langle\langle A; B \rangle\rangle_{\hbar\omega} = \frac{1}{2\pi} [\langle[A, B]\rangle + \langle\langle[A, \mathcal{H}]; B\rangle\rangle_{\hbar\omega}]. \quad (4)$$

In paper I the Green's functions $\langle\langle S_i^\pm; S_j^\pm \rangle\rangle_{\hbar\omega}$ and $\langle\langle S_i^\pm; S_j^\mp \rangle\rangle_{\hbar\omega}$ were calculated from the equation of motion (4) with the Hamiltonian (2). From the interaction terms in the Hamiltonian higher-order functions occurred which will be approximated¹⁴ by

$$\begin{aligned} \langle\langle S_i^z S_j^+; S_r^- \rangle\rangle_{\hbar\omega} &\rightarrow \langle S_i^z \rangle \langle\langle S_j^+ S_r^- \rangle\rangle_{\hbar\omega}, \\ \langle\langle S_i^z S_j^-; S_r^+ \rangle\rangle_{\hbar\omega} &\rightarrow \langle S_i^z \rangle \langle\langle S_j^- S_r^+ \rangle\rangle_{\hbar\omega}, \end{aligned} \quad (5)$$

¹³ D. N. Zubarev, Soviet Phys.—Usp. 3, 320 (1960).

¹⁴ In I, the GF were decoupled with a technique developed by H. B. Callen [Phys. Rev. 130, 890 (1963)]. The RPA decoupling in (5) is a special case. In a later paper, the thermodynamic properties will be calculated using the Callen-decoupling scheme.

where $\langle S_i^z \rangle$ is the spin-expectation value for a single ion.

After a transformation to reciprocal space we obtained in I the correlation functions, using (3) in the form

$$\psi^{+-}(\mathbf{k}) \equiv \sum_j \langle S_i^+ S_j^- \rangle e^{i\mathbf{k} \cdot (\mathbf{r}_i - \mathbf{r}_j)} = 2\langle S^z \rangle \phi(\mathbf{k}), \quad (6)$$

where the function $\phi(\mathbf{k})$ is related to the energies

$$F(\mathbf{k}) = \hbar\gamma H + 3\langle S^z \rangle \alpha(0) - \langle S^z \rangle [\alpha(0) - \alpha(\mathbf{k})] - 2\langle S^z \rangle \{I(0) - I(\mathbf{k})\}, \quad (7)$$

$$E(\mathbf{k}) = [F(\mathbf{k}) + \langle S^z \rangle D(\mathbf{k})]^{1/2} [F(\mathbf{k}) - \langle S^z \rangle D(\mathbf{k})]^{1/2}, \quad (8)$$

by the equation

$$\phi(\mathbf{k}) = \frac{1}{2} \left(\frac{F(\mathbf{k})}{E(\mathbf{k})} - 1 \right) + \frac{F(\mathbf{k})}{E(\mathbf{k})} \frac{1}{e^{\beta E(\mathbf{k})} - 1}. \quad (9)$$

The coefficients $\alpha(\mathbf{k})$, $D(\mathbf{k})$, $\phi(\mathbf{k})$, and $I(\mathbf{k})$ are the Fourier transforms of the interaction terms α_{ij} , $D_{ij} = D_{ij}^+ + D_{ij}^-$, and I_{ij} . $\phi(\mathbf{k})$ has the form of a quasi-boson occupation number. If we now introduce

$$\phi_0 = (1/N) \sum_{\mathbf{k}} \phi(\mathbf{k}), \quad (10)$$

we get the general relation¹⁵

$$\langle S^z \rangle = \frac{(S - \phi_0)(1 + \phi_0)^{2S+1} + \phi_0^{2S+1}(1 + S + \phi_0)}{(1 + \phi_0)^{2S+1} - \phi_0^{2S+1}}. \quad (11)$$

The set of Eqs. (7)–(11) may be solved self-consistently to give $\langle S^z \rangle(T, H)$.

The summation in (10) extends over the first Brillouin zone in the reciprocal lattice. Usually this sum is carried out over a sphere with the same volume as the true Brillouin zone.⁸ However, for the hexagonal lattice this is a very poor approximation. We approximate the Brillouin zone by a cylinder of radius $3.82/a$ and height $4\pi/C$ to make the numerical calculations tractable. Here $a = 7.36 \text{ \AA}$.

IV. HIGH-TEMPERATURE EXPANSION

For high temperatures, $\langle S^z \rangle \ll 1$ and therefore $\phi_0 \gg 1$, which makes an expansion of (11) in powers of ϕ_0^{-1} possible.

$$\langle S^z \rangle = \frac{1}{3} S(S+1) (\phi_0^{-1} - \frac{1}{2} \phi_0^{-2} + \dots). \quad (12)$$

If we retain only terms linear in the applied field H , we can expand $\langle S^z \rangle$ in powers of β

$$\langle S^z \rangle = \hbar\gamma H \sum_{j=1}^{\infty} C_j \beta^j. \quad (13)$$

In powers of β , we also have

$$\phi_0 = \frac{1}{N} \sum_{\mathbf{k}} \frac{F(\mathbf{k})}{\beta E_{-}^2(\mathbf{k})} + O(\beta), \quad (14)$$

¹⁵ H. B. Callen, Ref. 14.

and comparing terms of equal powers of β we have

$$\langle S^z \rangle = \frac{1}{3} S(S+1) \frac{\hbar \gamma H}{kT} \left[1 + \frac{\theta_M}{T} + \left(1 - \frac{\Delta}{\theta_M^2} \right) \frac{\theta_M^2}{T^2} + \dots \right]. \quad (15)$$

For the zero-field susceptibility

$$\chi = \lim_{H \rightarrow 0} \frac{1}{\mu_0} \frac{M}{H} = \lim_{H \rightarrow 0} \frac{1}{\mu_0} \frac{\hbar \gamma N}{H} \langle S^z \rangle, \quad (16)$$

we have therefore

$$\chi = \frac{C}{T} \left[1 + \frac{\theta_M}{T} + \left(1 - \frac{\Delta}{\theta_M^2} \right) \frac{\theta_M^2}{T^2} + \dots \right], \quad (17)$$

where

$$kC = \frac{1}{3} S(S+1) [(\hbar \gamma)^2 / \mu_0] N \quad (18)$$

is the Curie constant. The linear expansion coefficient θ_M is identical with the Curie temperature of the molecular field theory:

$$\theta_M = C(L^{zz} - N_z) - (4/3k)S(S+1)(I_1 + 3I_2). \quad (19)$$

L^{zz} is a dimensionless dipole-field factor (see Appendix) and N_z is the demagnetization factor. θ_M is shape-dependent due to N_z .

The series (17) deviates from the Curie-Weiss law in the quadratic term by the factor $(1 - \Delta/\theta_M^2)$, where

$$\Delta = \left[\frac{1}{3} S(S+1) \right]^2 \frac{1}{N} \sum_{\mathbf{k}} \{ [\alpha(\mathbf{k}) + 2I(\mathbf{k})]^2 + D^2(\mathbf{k}) \}. \quad (20)$$

Using the values for the dipole field factors given in the Appendix and the exchange constants $I_1 = 0.039$ k°K, $I_2 = -0.048$ k°K determined by Clover and Wolf,⁴ we calculate from (19) a value $\theta_M = 3.17^\circ\text{K}$. The experimental value for the corresponding terms in the high-temperature susceptibility is⁴: $\theta = (2.96 \pm 0.28)^\circ\text{K}$. The summation in (20) must be performed numerically, and we get

$$\Delta = (2.9 \pm 0.3)^\circ\text{K}^2.$$

Marquard¹⁰ has calculated the high-temperature susceptibility by an expansion of the partition function and found

$$\chi^{-1} = (T/C) [1 + B/kT + B_2/(kT)^2 + \dots], \quad (21)$$

where

$$B_1 = 21[I_1 + 3I_2] - 0.9647 + CN_z, \quad (22)$$

$$B_2 = 231[I_1^2 + 3I_2^2] + 6.377I_1 - 2.723I_2 + 0.4321. \quad (23)$$

B_1 is exactly $-\theta_M$, whereas $B_2 = 2.758$ (°K)².

From their high-temperature susceptibility measurements, Clover and Wolf⁴ derived a value

$$\Delta_{\text{exp}} = 1.9 \pm 0.2 (\text{°K})^2. \quad (24)$$

V. CRITICAL TEMPERATURE

In the molecular field theory, the critical temperature T_c is equal to the paramagnetic Curie-Weiss temperature θ_M , but actually T_c is much lower. Because experimentally the critical temperature of GdCl_3 is known to an accuracy of 0.01°K ,^{16,17} it offers a good test of the theory.

At the critical point we have for $H=0$ again $\phi_0 \gg 1$, and we may write for $T < T_c$

$$\langle S^z \rangle = |T - T_c|^\gamma \sum_{n=0}^{\infty} C_n (T_c - T)^n. \quad (25)$$

At $H=0$, the energies $F(\mathbf{k})$ and $E(\mathbf{k})$ are proportional to $\langle S^z \rangle$, and we get from (9) and (10) by expanding in powers of $\Delta = T_c - T$ the relation

$$kT_c = \frac{1}{3} S(S+1) \left(\frac{1}{N} \sum_{\mathbf{k}} \frac{f(\mathbf{k})}{\epsilon^2(\mathbf{k})} \right)^{-1}, \quad (26)$$

where

$$f(\mathbf{k}) = F(\mathbf{k}) / \langle S^z \rangle = 2[\alpha(0) - I(0)] + \alpha(\mathbf{k}) + 2I(\mathbf{k}),$$

$$\epsilon^2(\mathbf{k}) = E^2(\mathbf{k}) / \langle S^z \rangle^2 = f^2(\mathbf{k}) - D^2(\mathbf{k}).$$

The summation in (26) must be performed numerically. With the dipole sums given in the Appendix and the exchange constants of Clover and Wolf⁴ we get a critical temperature¹⁸

$$T_c = (2.48 \pm 0.12)^\circ\text{K},$$

if we consider the uncertainties in the values of I_1 and I_2 . The calculated value of T_c is still 12% higher than the measured value, indicating that the RPA is not a good decoupling for the Green's functions in the critical region. Another source of error is the approximation of the Brillouin zone by a cylinder. From (26), we can also calculate the Curie temperature of a pure dipole system by taking $I_1 = I_2 = 0$. For the same dipole sums as in the case of GdCl_3 we then get $T_c = 0.88^\circ\text{K}$. (The corresponding value of the molecular field theory is 0.96°K .)

¹⁶ A. F. G. Wyatt, Ph.D. thesis, Oxford University, 1963 (unpublished).

¹⁷ E. Becker *et al.*, Phys. Letters **19**, 86 (1965).

¹⁸ In the calculation of T_c it was assumed that at $T = T_c$ the crystal forms domains with a demagnetization factor $N_z = 0$. Actually the magnetic transition temperature of GdCl_3 is independent of the crystal shape [F. Kuch, Ph.D. thesis, Technische Hochschule Darmstadt, Germany, 1967 (unpublished)], supporting the above assumption. However, there is still the possibility that the dipole sums $\alpha(0)$ within a ferromagnetic domain are different from the sums of infinitely long thin ellipsoidal sample used in the evaluation of (26).

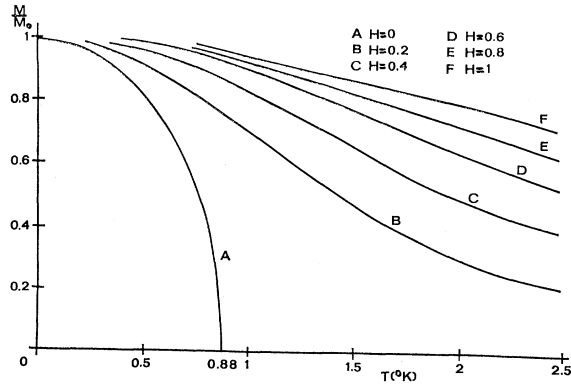


FIG. 2. Calculated magnetization M/M_0 for a hexagonal crystal with \mathbf{H} parallel to the c axis and dipole-dipole interaction only; lattice spacing as in the case of GdCl_3 .

For a dipole crystal, the condition that ϕ_0 be real also yields the condition

$$L^{zz} - L^{yy} - N^z \geq 0$$

for ordering at $T=0$ to be possible. All cubic crystals have $L^{xx} = L^{yy} = L^{zz} = \frac{1}{3}$ and in this approximation we get at best a metastable ferromagnetic ground state.

VI. MAGNETIZATION

The magnetization $M(H, T)$ is obtained by self-consistent numerical solution of the Eqs. (7)–(11) on a computer. Since the dipole sums are shape-dependent, we take an infinitely long thin ellipsoidal sample with $N_z = 0$. First, we calculate the magnetization of the crystal with the exchange interaction turned off. The

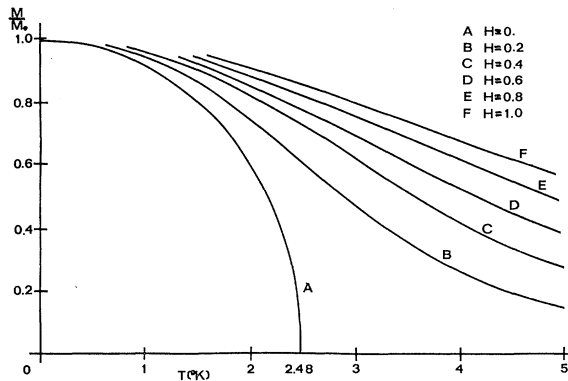


FIG. 3. Calculated magnetization M/M_0 for GdCl_3 with \mathbf{H} parallel to the hexagonal c axis.

result is shown in Fig. 2. At $T=0$ the magnetization is $0.99M_0$ for $H=0$ and it drops to zero at $T_c = 0.88^\circ\text{K}$.

Figure 3 gives the magnetization of the GdCl_3 sample. For zero magnetic field the $T=0$ magnetization is $0.995M_0$. This is in disagreement with the measurements of Wolf *et al.*,² who found a lowering of $\sim 18\%$ below M_0 for $T \rightarrow 0$. But the determination of the zero-field magnetization by the authors is not very accurate. It is expected that the magnetization measurements for GdCl_3 will be performed again with better samples. Figures 2 and 3 show also the calculated field dependence of the magnetization. In both cases the magnetic field is in units of $M_0/\mu_0 = 8440$ Oe and is varied from 0 to 1 in steps of 0.2.

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We thank Professor W. P. Wolf and R. B. Clover for helpful discussions.

APPENDIX: DIPOLE SUMS

The dipole sums $\alpha(\mathbf{k})$ and $D(\mathbf{k})$ are analytically simple only at $\mathbf{k}=0$ and for $10/R \lesssim k \ll 1/a$, where R is the size of the sample and a is a distance of the order of magnitude of the lattice constant. We have in these two regions (see paper I) for $S = \frac{7}{2}$

$$\alpha(0) = (2C/21)(L^{zz} - N_z),$$

$$\alpha(\mathbf{k}) = (2C/21)(L^{zz} - \cos^2\theta_k), \quad 10/R \lesssim k \leq a$$

$$D(0) = (2C/21)(L^{yy} - L^{xx} + N_y - N_x),$$

$$D(\mathbf{k}) = (2C/21)(L^{yy} - L^{xx} + \sin^2\theta_k), \quad 10/R \lesssim k \leq a.$$

Here C is the Curie constant (18), and θ_k is the angle between the \mathbf{k} vector and the z axis. N_x, N_y, N_z are the demagnetization factors. The dipole field factors are calculated in I:

$$L^{xx} = 0.2669, \quad L^{yy} = 0.1601, \quad L^{zz} = 0.5692.$$

For the sample used in our calculation, we have

$$N_x = N_y = \frac{1}{2}, \quad N_z = 0.$$

The small region $k \lesssim 10/R$ in the reciprocal lattice is unimportant for the thermodynamic properties. In the region $k \lesssim 1/a$ the \mathbf{k} dependence of $\alpha(\mathbf{k})$ and $D(\mathbf{k})$ is not given by the above expressions. Numerical calculations show that the deviations are small. We therefore, for $k < 0$, assume the dipole sums in the whole Brillouin zone to be of the same analytical form as in the region $10/R \lesssim k \ll 1/a$.