

EXPERIMENTAL TEST OF THE SPIN-WAVE THEORY OF A FERROMAGNET

A. C. Gossard, V. Jaccarino, and J. P. Remeika

Bell Telephone Laboratories, Murray Hill, New Jersey

(Received July 24, 1961)

A detailed theory of the thermodynamic properties of an idealized model of a ferromagnet, in which there exist only temperature-independent ferromagnetic nearest neighbor interactions having the form $J \sum_i \vec{m}_i \cdot \vec{S}_i$, has been given.¹ There has hitherto been no satisfactory test of the theory, primarily because no known ferromagnetic materials satisfied the assumptions of the model. Recently, Tsubokawa² has discovered an insulating ferromagnet, CrBr_3 , which closely approx-

imates the model. Using nuclear magnetic resonance (NMR) techniques, we have made a precise measurement of the temperature dependence of the magnetization of this compound.

CrBr_3 has the DO_5 (BiI_3) hexagonal layer structure³ shown in Fig. 1 and is prepared by heating powdered chromium in an atmosphere of bromine at 800°C .² Tsubokawa has measured several of its magnetic properties. In particular he has found a ferromagnetic Curie point $T_C = 37^\circ\text{K}$ and a saturation magnetization of $3\mu_B$ per Cr^{3+} ion as would be expected for the $3d^3$ 4S configuration in an octahedral crystal field. For the purely ionic 4S configuration a 375 000-oersted negative internal field at the Cr nucleus would be expected as a result of the exchange polarization of inner closed-shell s electrons by the aligned d electrons.⁴ Since overlap of the Cr and Br wave functions and covalent bonding reduce the magnitude of the internal field at the Cr nucleus, we searched for the Cr^{53} NMR in the frequency region below 85 Mc/sec using a frequency-swept modified Robinson marginal oscillator.⁵

An oscilloscope tracing of the spectrum observed at 58.038 Mc/sec ($H_{\text{hfs}} = 241\,220$ oe) and 1.34°K in zero external field is shown in the upper corner of Fig. 2. Three distinct absorptions are seen, whose separations and intensities correspond to the allowed transitions for $I = \frac{3}{2}$ and a nuclear electric quadrupole interaction $h^{-1}e^2q_{zz}Q = 1.184$ Mc/sec.

The occurrence of sharp satellites indicates a uniform orientation of the internal magnetic field with respect to the axes of the electric field gradient tensor. From this, and the external field dependence of the frequency for resonance, it may be concluded that the nuclei participating in the resonance are situated in the body of the domains and not in the domain walls. Enhancement of the resonant absorption occurs from domain rotation rather than wall motion.⁶ The sign of the magnetic field at the nuclei was determined to be negative with respect to the direction of magnetization from the observed decrease of the resonance frequency with increasing external field. The temperature dependence of the Cr^{53} NMR is shown in Fig. 2. The resonance was observed up to 15°K .⁷

Because of the hfs interaction $A\vec{I} \cdot \vec{S}$ between

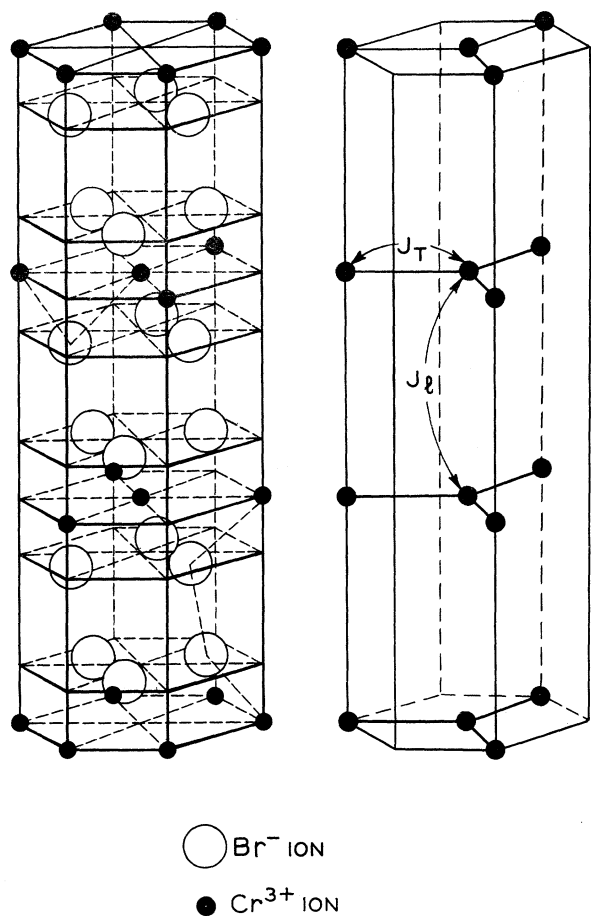
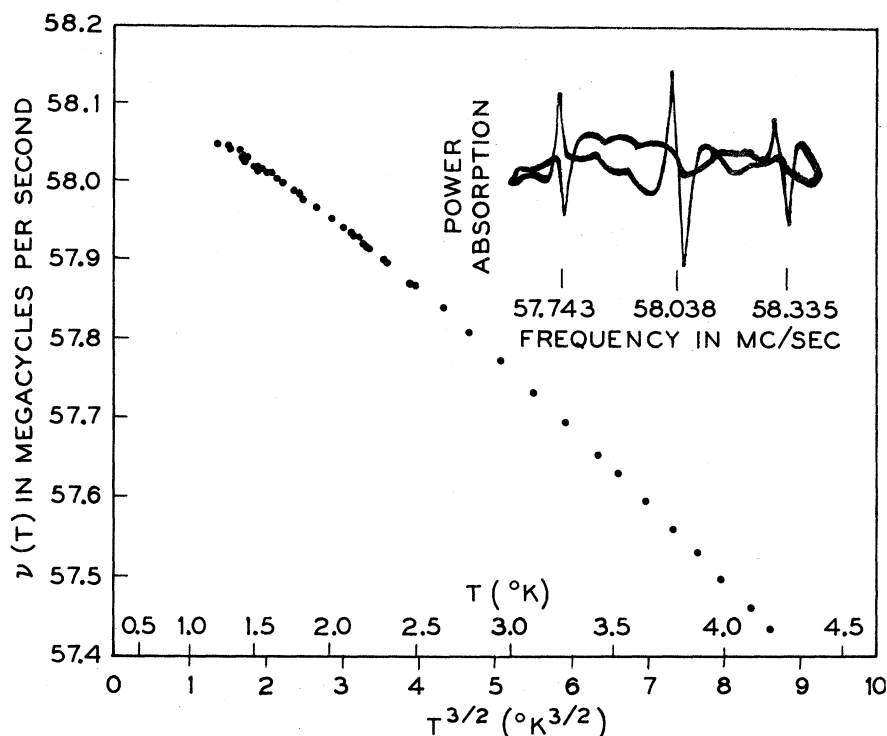


FIG. 1. Crystal structure of CrBr_3 (from Tsubokawa). The Br^- ions form a nearly perfect close-packed hexagonal structure. The Cr^{3+} ions occupy two-thirds filled hexagonal layers with two Br^- layers separating each Cr^{3+} layer. Each Cr^{3+} is surrounded by a nearly perfect octahedron of Br^- ions. At the right a simplified model used to calculate the spin-wave dispersion law is shown.

FIG. 2. Cr^{53} nuclear resonance frequency vs temperature in CrBr_3 in zero applied magnetic field. An oscillographic picture of the resonances at 1.34°K is shown above. Differences in the shape of the resonances on forward and return sweep are caused by passing through the lines in times comparable to nuclear relaxation times.



the ionic spin $\tilde{S}$ and the nuclear spin $\tilde{I}$, NMR is observable at a temperature $T < T_c$ at a frequency $\nu(T) = \hbar^{-1} A \langle S \rangle_T$, where $\langle S \rangle_T$ is the thermal average of $\tilde{S}$. Since $M(T) = N g \beta \langle S \rangle_T$, we expect $\nu(T)$ to be proportional to $M(T)$ provided that A depends neither implicitly nor explicitly on T . The temperature dependence of hfs interactions has been investigated in other magnetic solids.⁸ The results of such studies lead us to conclude that the variation of A between 0° and 4°K , in CrBr_3 , is considerably less than the intrinsic precision of our experiment (1 part in 30 000).

The spin-wave prediction⁹ for the temperature dependence of the spontaneous magnetization $M(T)$, for $T \ll T_c$ is,

$$M(T) = M(0) [1 - aT^{3/2} - bT^{5/2} - cT^{7/2} - dT^4 - eT^{9/2} - \dots]. \quad (1)$$

The half-odd-integral terms beyond $T^{3/2}$ result from the discreteness of the lattice, while the T^4 term is the lowest spin-wave interaction term. In the case of cubic symmetry with no external field or anisotropy, the coefficients a , b , c , d , and e are functions only of J , S and the lattice geometry. In the DO_5 lattice, however, several exchange interactions are involved. Nearest neighbor spins within a layer are strongly coupled by superexchange through a single inter-

vening ion. Exchange between spins in adjacent layers is weaker because two intervening ions in series separate the interacting spins. Calculation of the expected magnetization coefficients is thus complicated by the fact that some weak exchange is expected even for spins not directly above one another. To simplify calculation of the coefficients, we have adopted a model of the lattice shown at the right in Fig. 1. Only two exchange interactions, J_l and J_t , are considered in the spin-wave calculation. For $J_l \ll J_t$ we obtain $a = 0.0130 J_l^{-1/2} J_t^{-1}$ and $b = 0.000140 J_l^{-3/2} J_t^{-1}$. In the presence of a magnetic or anisotropy field, H , the $T^{3/2}$ coefficient in the same formalism becomes

$$(a/2.612) \sum_n [n^{-3/2} \exp(-ng\beta H/kT)].$$

The other coefficients are modified similarly.

In Fig. 2 the results of the NMR measurements are plotted against $T^{3/2}$ for $T < 0.11 T_c$. At the lowest temperatures the effect of the anisotropy-modified $T^{3/2}$ term is seen to predominate. Using Dillon's observed value¹⁰ of 6850 oersteds for the anisotropy field, a least-squares fit of the form

$$\nu(T) = \nu(0) \left[1 - \frac{a}{2.612} \sum_n \frac{e^{-ng\beta H/kT}}{n^{3/2}} T^{3/2} - \frac{b}{1.341} \sum_n \frac{e^{-ng\beta H/kT}}{n^{5/2}} T^{5/2} \right],$$

was made. $\nu(0)$ was determined to be 58.096 Mc/sec, corresponding to a field at the nucleus of 241 460 oersteds. a was $(2.544 \pm 0.067) \times 10^{-3} \text{ }^\circ\text{K}^{-3/2}$, b was $(3.03 \pm 1.04) \times 10^{-5} \text{ }^\circ\text{K}^{-5/2}$, and the mean square deviation of the data points from the fit was 0.0016 Mc/sec. Inclusion of $T^{7/2}$ and T^4 terms does not significantly improve the fit. From a and b we calculate that $J_t = 5.44^\circ\text{K}$ and $J_l = 0.88^\circ\text{K}$. The smallness of J_l relative to J_t , inferred from inspection of the superexchange "paths," is thus confirmed, and manifests itself in the magnetization curve as an unusually large $T^{5/2}$ term.

We wish to thank R. J. Blume for the circuit of the Robinson-type spectrometer, J. L. Davis for experimental assistance, E. M. Kelley for aid in growing the crystals, and Dr. L. R. Walker for theoretical counsel.

- ¹F. Bloch, *Z. Physik* **74**, 295 (1932).
- ²Ichiro Tsubokawa, *J. Phys. Soc. Japan* **15**, 1664 (1960).
- ³A. Braekken, *Kgl. Norske Videnskab. Selskab, Forh.* **5**, No. 11 (1932).
- ⁴V. Heine, *Phys. Rev.* **107**, 1002 (1957).
- ⁵F. N. H. Robinson, *J. Sci. Instr.* **36**, 481 (1959); R. J. Blume (to be published).
- ⁶A. M. Portis, A. C. Gossard, R. H. Lindquist, and M. Rubinstein (to be published).
- ⁷R. G. Barnes and S. L. Segel have observed Br^{81} nuclear quadrupole resonance in the paramagnetic state of CrBr_3 . R. G. Barnes and S. L. Segel, *Phys. Rev. Letters* **3**, 462 (1959).
- ⁸G. B. Benedek and J. Armstrong, *J. Appl. Phys.* **32**, 106S (1961); V. Jaccarino and L. R. Walker (to be published).
- ⁹Freeman J. Dyson, *Phys. Rev.* **102**, 1230 (1956).
- ¹⁰We are indebted to J. F. Dillon for communication of this result before publication.

PHASE TRANSITION IN MERCURY TELLURIDE*

J. Blair and A. C. Smith

Department of Electrical Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts

(Received July 17, 1961)

An abrupt change in the resistivity of mercury telluride has been observed at a pressure of about 15 000 kg/cm² at room temperature. This change is ascribed to a transition between two polymorphic phases of HgTe. The resistivity changes essentially reversibly by a factor of 10^4 to 10^5 at this transition. No information about the structure of the high-pressure phase has been obtained.

Figure 1 shows the variation of the resistivity of a sample of HgTe as a function of pressure. The resistivity was measured with a four-probe method using a precision potentiometer. At 16 000 kg/cm², the resistivity increases by a factor greater than 10^4 . The transition takes place very slowly and the points above 16 000 kg/cm² do not represent an equilibrium situation. The time that elapsed while the sample was in the high-pressure phase was several hours and the resistivity drifted upward consistently. As the pressure was decreased, the reverse transition occurred at 12 000 kg/cm². The resistivity decreased by a factor of 10^4 , but did not quite return to its original value.

When the sample was examined, several cracks were found. These cracks presumably account for the difference in the atmospheric pressure values of the resistivity before and after the application of pressure. The sample was initially a single crystal and if the transition nucleated at a num-

ber of points, the sample would be polycrystalline after the transition. The strains arising from misorientation of the crystallites would produce the observed cracks.

The resistivity of this sample before the pressure experiment was 1.5×10^{-3} ohm cm. Meas-

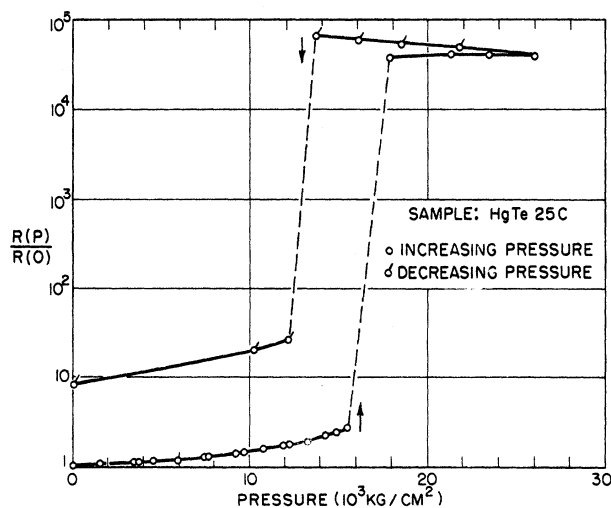


FIG. 1. Normalized resistance of HgTe vs pressure. Arrows show pressures at which transition occurred.