PHYSICAL REVIEW B VOLUME 13, NUMBER 3 1 FEBRUARY 1976

Magnetoresistance and field-induced phase transitions in the helical and conical states of holmium*

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(Received 22 July 1974)

Magnetoresistance measurements on single crystals of holmium are reported for the helical antiferromagnetic and conical ferromagnetic phases. Longitudinal and transverse measurements were made with the magnetic field applied in the a, b, and c planes, including nine different field-current configurations. Data with the current parallel to the c direction and the applied field parallel to the easy b direction showed the magnetoresistance to be as large as 40%. Basal-plane resistance anomalies have been observed, which when compared to magnetostriction, magnetization, and neutron-diffraction data can be correlated with field-induced transitions, permitting the construction of schematic H-T phase diagrams. The results of these measurements are in good general agreement with previous work, with the exception that evidence for an additional stable intermedite state has been obtained. The basal-plane data were isotropic with respect to the current direction, and highly anisotropic with respect to the direction of the applied magnetic field. In order to observe sharp resistance anomalies, it was essential to mount the samples in such a way as to provide for relief of thermal and magnetostrictive stresses. All samples studied were in the form of thin disks, with the magnetic field applied in the plane of the disk to minimize the demagnetizing effects.

INTRODUCTION

In the temperature range of approximately 133–20 °K, holmium has been found to exhibit simple helical antiferromagnetic order.[1] In this state, the magnetic moments in each c plane (holmium is hcp) are ordered ferromagnetically, but the direction of the magnetization rotates by a temperature-dependent pitch angle from layer to layer, generating a helical structure. Below 20 °K, the projections of the magnetic moments on the basal planes retain the helical character, but the magnetization in each layer also has a component in the c direction, giving rise to a net moment, and a conical ferromagnetic state with a pitch angle of 30°. Application of a magnetic field of sufficient intensity in the basal plane of the specimen leads to a succession of structure modifications.[2,3] With increasing magnetic field, a sequence of distinct stable "fan" states may be obtained. Ultimately, at least for fields applied in the easy (b) direction, simple ferromagnetic order is achieved. A description of the various states has been provided by combining neutron-diffraction data with magnetization data,[4] yielding schematic H-T phase diagrams for two different basal-plane field alignments. It is to be noted that neutron-diffraction data in Ref. 2 from the two specimens studied differed somewhat, particularly for $\vec{H} \parallel \vec{a}$.

Sherrington[5] has proposed that intermediate spin orderings may be found in holmium, including modifications of both the antiferromagnetic helix, and ferromagnetic cone states. In both cases the state modification involved tilting the magnetization in each layer, about an axis in the c plane. The analysis was based upon the stability of the helical and conical states to magnon mode softening at wave vectors $q = q_0$, where q_0 is the wave vector associated with the periodic order. Previously, Woods *et al.*[6] had discussed the relative stability of the two phases (helix and cone) as a function of axial anisotropy, indicating a range of anisotropy for which the helical and conical states would be unstable. There apparently exists no detailed analysis of this problem considering applied-field effects, which would be pertinent to our experiments.

Magnetization studies of single crystals of holmium showed that the critical field H_c to collapse the helix varied with temperature to an approximate maximum of 17 kOe at 75 °K. A steplike increase of the magnetization occurs at H_c, and this is accompanied by a change in the size and shape of the crystal, due to magnetostriction.[7]

Depending on the temperature, both the magnetization and magnetostriction data display knees, suggesting the existence of intermediate stable spin configurations. The neutron experiments were instrumental in unraveling the details of these complex structures. We find that the magnetoresistance measurements are reliable, sensitive probes to determine the boundaries of the various phases.

EXPERIMENTAL PROCEDURE

Six holmium disks, two specimens each of a, b, and c plane orientations were prepared from a crystal grown by a commercial firm with a stated purity of 99.9%. The temperature dependence of the resistivity was found to be in excellent agreement with published data,[4] indicating that the crystal's quality was comparable to those previously studied. In order to minimize the demagnetizing fields, thin samples were employed. However, for thickness less than 0.3 mm, the specimen became extremely fragile and easily damaged. The dimension of the samples studied varied from 0.4 to 0.7 mm in thickness and from 6.4 to 7.0 mm in diameter.

A variable temperature Dewar employing cold helium gas was used to provide the desired operating temperature for the specimen. Temperature stabilization was achieved to ±10 mK using an automatic regulator system employing a field-insensitive capacitance thermometer[8] with General Radio 716-C capacitance bridge. The output of the bridge was detected synchronously with a lock-in amplifier and the output signal used to control a remotely programmable power supply driving a heater in intimate physical contact with the sample. This provided for fine control of the temperature. The temperature was measured and set at zero applied field to an accuracy of ±0.5 °K using a 0.3% gold-iron vs Chromel-P thermocouple[9] and a Leeds and Northrup $K5$ potentiometer.

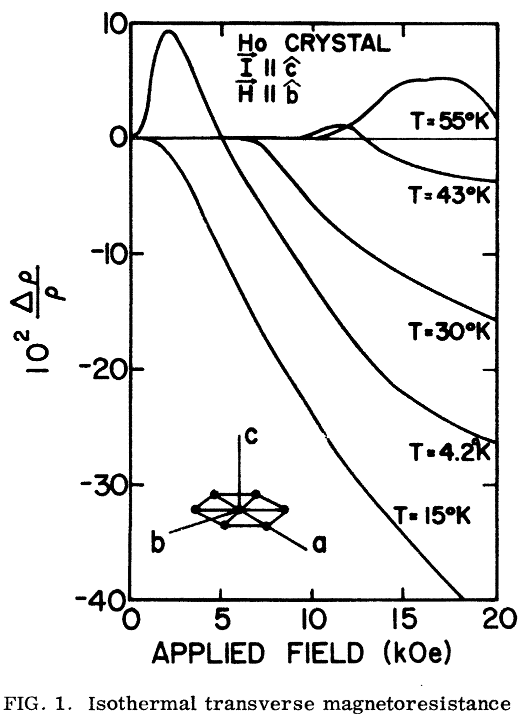

FIG. 1. Isothermal transverse magnetoresistance curves measured parallel to the c axis of holmium as a function of field applied parallel to the b direction. Identical results are obtained when the magnetic field is applied along the a axis. The 40% negative magnetoresistance at 15 °K for an applied field at 18 kOe may be associated with the suppression of the electron energy gaps by the magnetic field applied in the basal plane.

A modified four-probe ac method was used to measure the sample's resistance, employing two sharp pointed brass probes as electrical contacts. Two small nylon C-clamps supported the crystal, and the probes were placed approximately 3 mm apart, symmetrically about the center of the crystal along one of the crystallographic directions. Two wires were attached to each probe, one pair provided a "constant" ac current, and the other pair was used to measure the voltage drop across the sample. An ac current (50 mA rms) at 10 kHz was used and the voltage drop across the sample was synchronously detected by a lock-in amplifier whose output was connected to the y axis of an x-y recorder. The x axis of the recorder was driven by the magnet power supply, which provided a voltage proportional to the magnetic field strength.

As previously reported for dysprosium,[10] it was found to be essential to mound the sample in such a way that it could distort freely under the influence of thermal and magnetostrictive stresses. When the sample was constrained by rigidly fixing both nylon C-clamps to the base of the probe, peaked resistance anomalies were smeared out and became unresolvable. This condition was immediately obvious from data with $T < T_N$, and such runs were terminated.

RESULTS

Several typical isothermal curves of the transverse magnetoresistance for the c axis as a function of magnetic field applied along the b direction are shown in Fig. 1. The application of a magnetic field in the b direction in holmium destroys the magnetic periodicity leading to a ferromagnetic state. This should suppress any exchange energy gaps associated with the helical order, which may make a large contribution to an enormous 40% negative magnetoresistance for the c direction observed at 15 °K with an applied field of 18 kOe.

Measurements of this configuration at helium temperatures were intriguing. For small applied fields, the magnetoresistance was positive, reaching a maximum of approximately 10% before decreasing to a negative value of approximately 27% at 20 kOe. A maximum was observed in both holmium and dysprosium below approximately 7 °K. The origin of this effect is not presently understood.

The abrupt changes in slope, and peaks in the magnetoresistance curves at 55, 43, and 30 °K (Fig. 1) apparently correspond to transitions from one stable state to another. The c-axis transverse magnetoresistance was found to be relatively insensitive to the onset of structure modifications. Sharp anomalies were observed in other measurement configurations which will be discussed later.

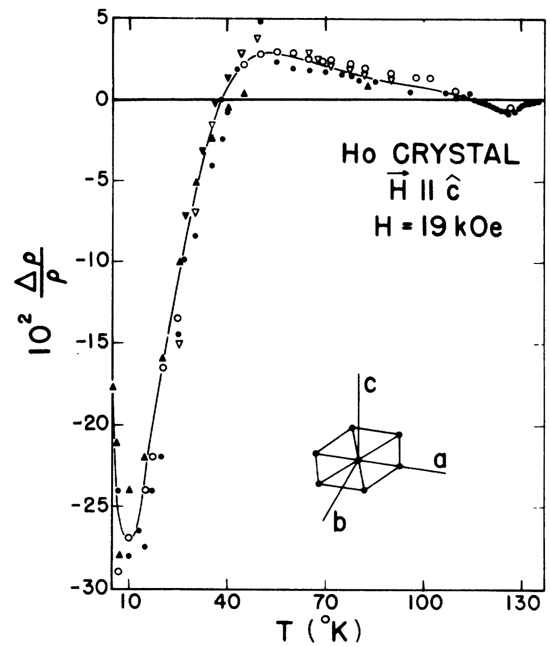

FIG. 2. Longitudinal magnetoresistance measured parallel to the c axis of holmium at 19 kOe as a function of temperature. The magnetoresistance changes sign at approximately 40 and 120 °K. X-ray and neutron-diffraction experiments do not indicate any crystal- or magnetic-structure modification at these temperatures. The various symbols correspond to different runs.

When the magnetic field was applied along the c direction, the isothermal curves of the longitudinal magnetoresistance (not shown) varied monotonically with the applied magnetic field. Figure 2 illustrates the temperature dependence of the longitudinal magnetoresistance for the c direction at 19 kOe. The magnetoresistance changes sign at 120 and 40°K, however, neutron-diffraction measurements[2] have given no indication of any crystal or magnetic structure modification at these temperatures.

The effect of spin-lattice coupling on resistivity has been dramatically illustrated by the analysis of apparently inconsistent data for the a and c directions in gadolinium.[11] The authors showed that a large contribution to the c axis data was due to anomalous lattice contraction. Correcting for this effect, they showed that the spin-disorder scattering contributions to the a- and c-axis data were similar. They argued [Ref. 11, Eq. (1)] that the temperature derivative of the resistivity contains a term proportional to the thermal expansion coefficient.

It is perhaps worthwhile to consider a modest extension of their argument to illustrate the role of magnetostriction in magnetoresistance measurements at constant pressure:

$$\left(\frac{\partial \rho_i}{\partial H}\right)_P = \sum_j \left(\frac{\partial \rho_i}{\partial L_j}\right)_H \left(\frac{\partial L_j}{\partial H}\right)_P + \left(\frac{\partial \rho_i}{\partial H}\right)_{L_j}.$$

Here ρ_i is the resistivity in the ith direction and L_j are the lattice constants. It is well known that large lattice distortions accompany the field-induced collapse of ordered spin structures and data for the field derivatives of the lattice constants can be easily determined from field-dependent magnetostriction measurements.[7] One may easily see that the derivatives $(\partial L_j/\partial H)_P$ are sharply peaked at a field which gives rise to a "steplike" change in the lattice constants. As a result, if the coefficients $(\partial \rho_i/\partial L_j)_H$ are monotonic, there will be a sharply peaked contribution to $(\partial \rho_i/\partial H)_P$. This is consistent with a "steplike" change in the magnetoresistance. It is now evident that if the sample is rigidly constrained "steplike" anomalies in the magnetoresistance due to the magnetostriction will be suppressed. It is difficult to make an estimate of $(\partial \rho_i/\partial H)_{L_j}$ since for spin systems ordered with a nonzero wave vector, we expect that one of the substantial contributions to $(\partial \rho_i/\partial H)_{L_j}$ arises from the field-induced distortion of the spin configurations. Application of a field will, in general, tend to change the harmonic content of a spin structure, and contribute to superzone effects and spin-disorder scattering.

The field-induced spin structure modifications involve order-order magnetic transitions which may be of first or second order. In such cases that the magnetization is continuous, we expect that spin fluctations at the transition field will give rise to peaked resistance anomalies. On the other hand, if the magnetization is discontinous, the resistivity may have a steplike change due to changes in spin-disorder scattering. Unfortunately, complex changes in the conduction bands may accompany the field-induced magnetic phase transitions. As a result, it may be difficult to predict the behavior of the resistivity in the vicinity of such a transition.

Magnetoresistance measurements in the basal plane for temperatures greater than 20°K were found to be isotropic with respect to the direction of the current and highly anisotropic with respect to the direction of the applied magnetic field. In addition, application of a magnetic field in the basal plane introduces dramatic features in the isothermal magnetoresistance data. For temperatures less than 20°K, the magnetoresistance is nearly isotropic. Figure 3 illustrates the effects of the field in the b direction with the magnetic field applied in the a direction. A peak in the magnetoresistance occurs below approximately

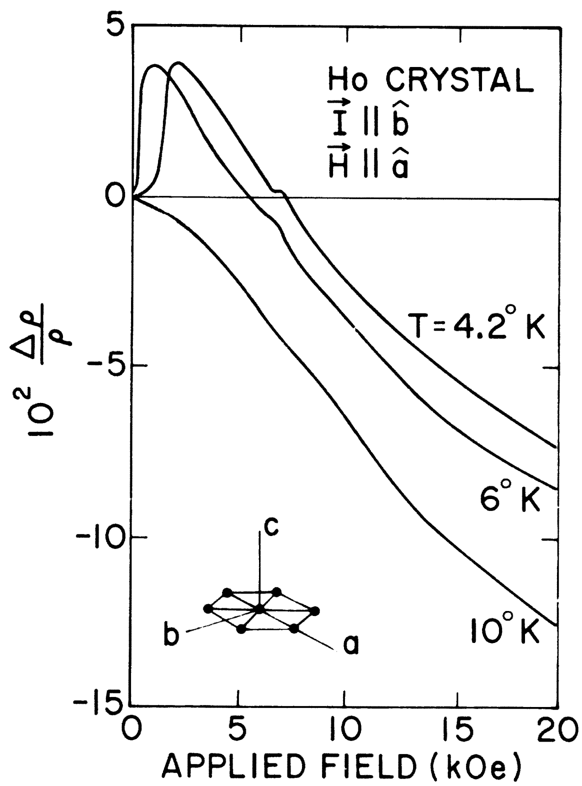

FIG. 3. Typical isothermal curves of the transverse magnetoresistance measured parallel to the b direction of holmium with the magnetic field applied parallel to the hard a axis as a function of temperature. In the ferromagnetic region, for temperatures less than 20 °K, the magnetoresistance in the basal plane is essentially isotropic with respect to the a and b directions. The anomalies in the curves are believed to be due to field-induced magnetic phase transitions. For the complete H-T phase diagram, see Figs. 5 and 8.

7 °K. It is interesting to note that this peak shifts to higher magnetic fields for lower temperatures. The origin and the behavior of this peak cannot be explained by the limited available theoretical analysis appropriate to the field-induced structure modifications in holmium.

The correlation of the magnetization, magnetostriction neutron-diffraction results, when compared to the magnetoresistance data involving anomalies suggests that the magnetoresistance may be the most sensitive indicator of structure modifications. It is to be noted that the neutron data were correlated with magnetization data to construct the phase diagrams in Ref. 2. In addition, the magnetization and magnetostriction are obviously insensitive to second-order transitions. However, both the neutron and magnetoresistance[13] techniques are sensitive to second-order magnetic phase transitions. Therefore, we assumed that resistance anomalies in isothermal magnetoresistance curves indicated transition fields, and used these data to construct H-T phase diagrams, correcting for the small demagnetizing fields associated with the sample geometry. An example of the data reduction is shown in Fig. 4 which illustrates typical longitudinal isotherm curves of the resistivity for the a direction.

The magnetoresistance was not a strong function of the applied magnetic field for fields less than a certain value. At a critical field H_1 a discontinuous change in the resistivity appeared. This corresponds to the so-called "helix–helix+ I" transition discussed in Fig. 8 of Ref. 2. As the magnetic field was increased, another discontinuity in the resistivity was observed. The value of the ap-

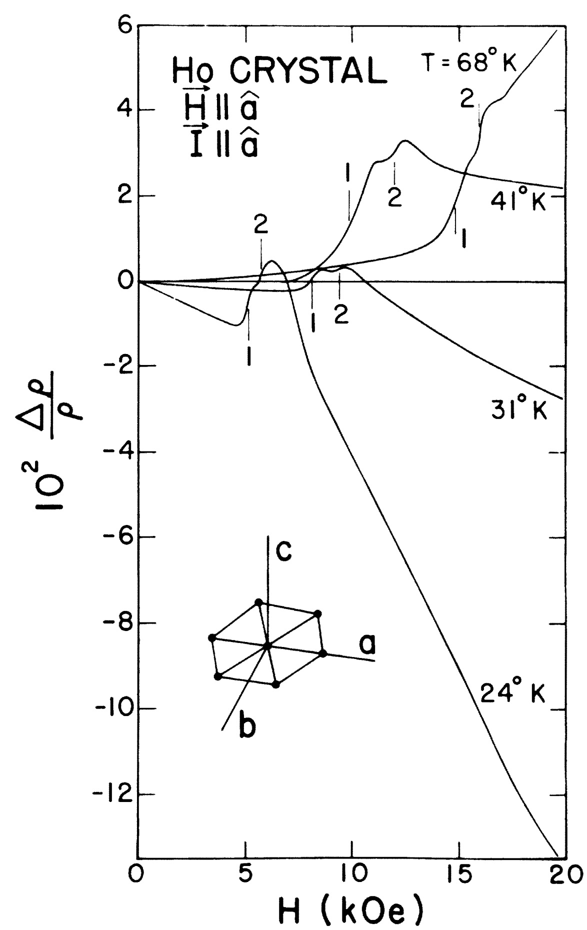

FIG. 4. Longitudinal magnetoresistance data for the hard a axis of holmium as a function of the applied magmetic field in the antiferromagnetic region. The transverse data with the same field orientation yielded identical results. The fields at which an abrupt increase in magnetoresistance occurs, H_1, correspond to "helix-helix + I" transitions. The fields at which a second abrupt increase occurs are referred to as H_2. In the figure, the fields H_1 and H_2 are labeled as 1 and 2, respectively, for each temperature.

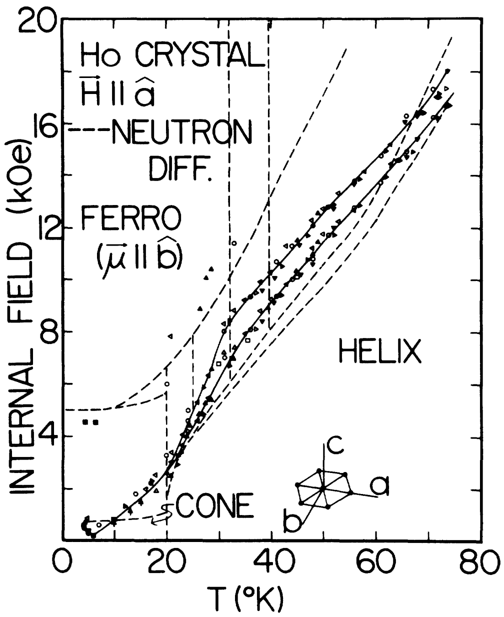

FIG. 5. Schematic H-T phase diagram for single-crystal holmium deduced from the magnetoresistance data with the magnetic field applied parallel to the hard ***a*** axis. The dashed lines are the results of the neutron-diffraction experiments, which are included for comparison. The open and closed symbols are from the longitudinal and transverse magnetoresistance data, respectively, and the various symbol shapes on the figure are from data on different crystals.

plied magnetic field H_2 for which this discontinuity occurs, corresponds to the transition from "helix+ I" to a state with a ferromagnetic component. Occasionally it was possible to resolve a third transition at higher fields. The schematic phase diagram for the applied magnetic field along the a axis obtained from data similar to that illustrated in Figs. 3 and 4 is given in Fig. 5. The neutron-diffraction results indicate a direct transition from "helix" to "helix plus ferro" between 20 and 25 °K. The phase diagrams, obtained from the magnetoresistance measurements, suggest the existence of an intermediate state, possible "helix + I," in this temperature region. Although the general agreement between the phase diagram determined here and the neutron-diffraction data is somewhat unsatisfactory, the phase diagram is in excellent agreement with other experimental observations, such as magnetization[4] and microwave absorption.[12] This suggests that the neutron-diffraction data maybe in error, perhaps owing to internal-field inhomogeneity and the sample-mounting procedures employed.

The most complex magnetization process was indicated when the magnetic field was applied along the b direction. For the temperatures great-

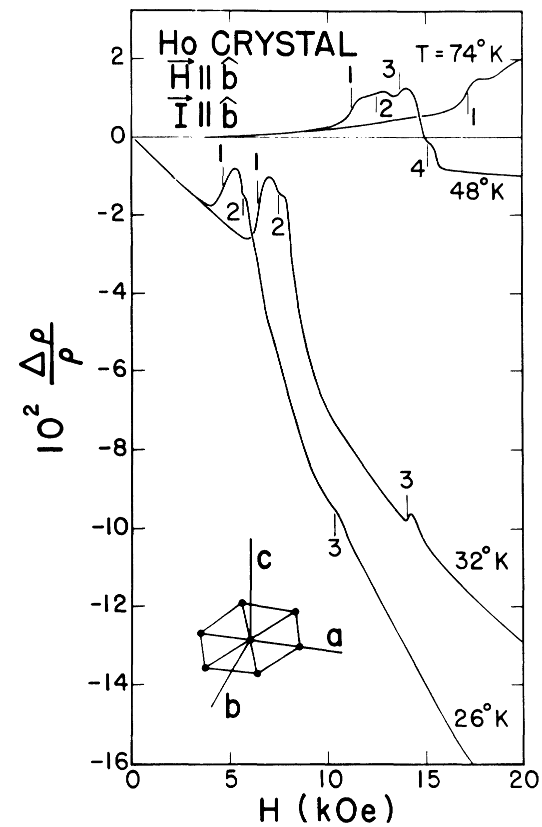

FIG. 6. Several isothermal plots of the longitudinal magnetoresistance for the easy b direction of holmium as a function of the applied magnetic field in the antiferromagnetic region. The transverse data with the same direction of the applied field yielded identical results. The abrupt changes in slope and peak, corrected for the demagnetizing field, correspond to the transitions between different stable magnetic states. In the figure, 1, 2, 3, etc. correspond to transition fields H_1, H_2, H_3, etc.

er than 45 °K, the spin system could apparently exist in as many as five different stable states for different strengths of the applied magnetic field. Figure 6 illustrates the transitions between these states as were identified from the longitudinal isotherm curves of the magnetoresistance for the b direction. The data presented here are for increasing field; when the measurements were made with decreasing applied magnetic field, hysteresis was generally observed. The most striking features of the hysteresis was observed at temperatures of approximately 40 °K. As shown in Fig. 7, the field separations between subsequent transitions were different for opposite senses of the applied magnetic field sweep, and in general, the response of the spin structure to the increasing applied magnetic field was quite different than for

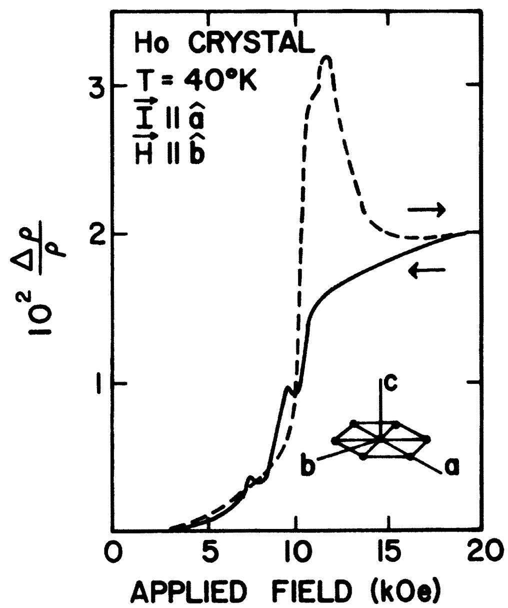

FIG. 7. Hysteresis in the transverse magnetoresistance for single-crystal holmium measured parallel to the *b* axis as a function of magnetic field applied parallel to the *a* direction at 40 °K. Not only do the transition fields shift to lower values for decreasing field sweeps, but the field separations between the various transitions are different for opposite sweep senses of the applied magnetic field. The dashed and solid lines are for increasing and decreasing field, respectively.

a decreasing field.

The schematic *H*-*T* phase diagram deduced from these measurements with the applied magnetic field parallel to *b* direction is illustrated in Fig. 8. The phase diagram obtained from the neutron-diffraction data[2] is also presented for comparison.

The intermediate magnetic phase transitions, for temperatures greater than 20°K, observed by the neutron-diffraction experiments are in excellent agreement with the results obtained from the analysis of the magnetoresistance data. The field required for the transition from "cone" to "cone plus ferro" (Fig. 8 of Ref. 2) decreases approximately linearly with decreasing temperature. The analysis of the magnetoresistance data indicates an increase in this field below 7°K. This was also observed when the magnetic field was applied along the *b* direction as was illustrated in Fig. 3 in agreement with the magnetization data.[4]

Several features of the isothermal magnetoresistance curves of Fig. 6 which correspond to the "helix+ I-I+II" (Fig. 8 of Ref. 2) transitions, above approximately 45°K, persist to lower temperatures than previously reported. It appears that the neutron-diffraction experiments were unable to resolve this transition.

CONCLUSION

Although the results of a general calculation of the magnetoresistance of a magnetic specimen with a periodic spin structure is not available, experimental measurements of the magnetoresistance of holmium have been shown to provide substantial information about the *H*-*T*-plane phase diagram. When a spin structure undergoes a field-driven phase transition, or a structure modification which is a monotonic function of the applied field, changes in the spin-disorder scattering are easily observed. In particular we have shown how abrupt changes in the lattice constants at critical values of the applied field lead to steplike contributions to the magnetoresistance. As a result, the magnetoresistance measurements are highly sensitive to field-induced phase transitions, which lead to substantial changes in the magnetization, and therefore, the magnetostriction.

In the event that a field-induced transition occurs with a continuous magnetization, spin fluctuations accompanying the transition can give use to sharp

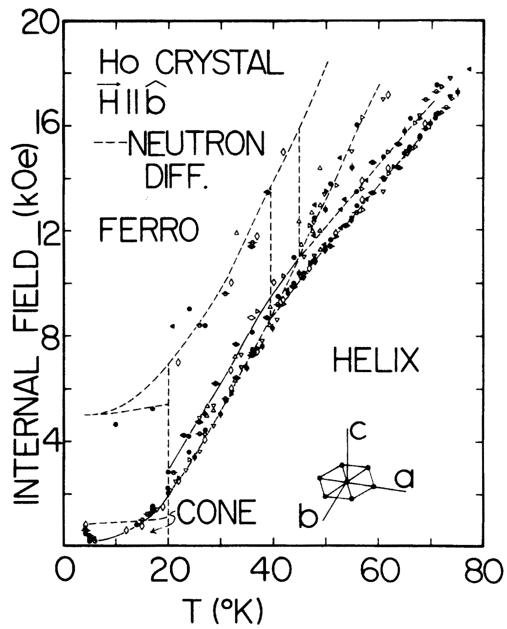

FIG. 8. Schematic *H*-*T* diagram for single-crystal holmium deduced from the magnetoresistance data with the magnetic field applied parallel to the easy *b* direction. The dashed lines are the results of the neutron-diffraction experiments, which are included for comparison. The open and closed symbols were obtained from the longitudinal and transverse magnetoresistance data, respectively, and the various symbol shapes on the figure are from data on different crystals. The data suggest the existence of an additional field-induced stable state not observed in the neutron-diffraction experiments.

peaks in the magnetoresistance.[10] Therefore, magnetoresistance measurements can be used as an effective tool to determine the boundaries of the various phases of a complex spin structure in the H-T plane. We find excellent general agreement with the previously published H-T phase diagram, which was deduced from neutron-diffraction and magnetization data. However, our data suggest the persistance of an intermediate state to a lower temperature than previously reported. We also observed an increase of the field required for the "cone" to "cone plus ferro" transition for temperatures less than 7 °K. This result is consistent with the magnetization of Strandberg *et al.*[4] We also observed large hysteresis in the transition fields in the neighborhood of 45 °K. At other temperatures, the hysteresis was observed to be substantially smaller. Unfortunately, the magnetoresistance does not yield definitive data about the type of spin structure, except possibly for a simple ferromagnetic configuration, for which a calculation predicts a negative mangetoresistance.[13] Our data are consistent with this result.

In order to conclude anything more precise about the response of the conduction electrons to the various magnetic structures, more detailed data on magnetostriction and magnetization will be required. Further, additional neutron-diffraction experimentation would illuminate the character of the various magnetic phases of holmium.

*Supported in part by the National Science Foundation.
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