Magnetoresistance and Field-Induced Phase Transitions in the Helical Antiferromagnetic State of Dysprosium*

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Field-induced magnetic phase transitions in the helical phase of dysprosium have been studied through basal-plane magnetoresistance measurements. Certain details of the measurement technique were found to be of profound importance: it is essential that the sample's internal magnetic field have high homogeneity, and that the single-crystal specimen be mounted in such a way that it may distort freely under the influence of thermal and magnetostrictive stresses. Throughout this investigation, the magnetic field was applied in the basal plane. Anomalies in the magnetoresistance, which are correlated with the field which collapses the helix at a critical value H_c, have been observed. For temperatures greater than 130 °K, and fields greater than H_c, anomalies have been observed which are believed to result from transitions from fan to ferromagnetic ordering. Resistance deviations as large as 4% were observed at 20 kOe, and may be important to the interpretation of microwave-absorption measurements.

In the temperature range of approximately 179–85 °K, dysprosium has been found to exhibit helical antiferromagnetic order.[1] In this state, the magnetic moments in each *c* plane (dysprosium 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. The electrical, magnetic, and magnetoelastic properties of this state have been extensively studied, and the reader is referred to works by Cooper,[2] Elliott,[3] and Tebble and Craik[4] for general information. Herpin and Meriel[5] were among the first to study helical antiferromagnets, using the methods of neutron diffraction and magnetization to characterize the properties of $MnAu_2$. They showed that the application of a magnetic field along the axis of order had little effect, but a field applied perpendicular to the helical axis was capable of collapsing the helix at a critical-field value referred to as H_c. Further, they showed that an intermediate "fan" state could exist in a range of applied fields between H_c and a limiting value, denoted as H_f, of sufficient intensity to collapse the fan into complete ferromagnetic alignment. Magnetization studies of single crystals of dysprosium by Behrendt *et al.*[6] showed that the critical field to collapse the helix varied with temperature to an approximate maximum of 11 kOe, a fortuitously convenient value for experimentation. A steplike increase of the magnetization occurs at H_c, and this is accompanied by a similar change in the size and shape of the crystal, due to magnetostriction.[7] Although both the magnetization and magnetostriction data show rounding for applied fields in excess of H_c, no value for H_f has been deduced from these measurements.

A number of investigators[8–16] have considered theoretical analysis of magnetization processes of a helical antiferromagnet. Such studies generally suggest that a fan state should exist. Neutron-diffraction methods have not yet been applied to a study of the magnetization process of dysprosium, but such experiments are now planned[17] and should provide data of great value in determining under what conditions the fan, or possibly other intermediate states, may exist.

Our studies of the magnetoresistance of single-crystal dysprosium were initially stimulated by microwave-absorption and resonance studies of the helical phase, which provided data difficult to interpret using available models.[18] The essential point in this regard is that a microwave spectrometer cannot separately distinguish between a field-dependent resistance change and a magnetic process, since both will give rise to changes in the microwave-cavity quality radiation factor, *Q*.

Previous magnetoresistance measurements[19] have indicated that anomalies exist at H_c, and we considered it essential to obtain such data on samples of the same geometry used in resonance experiments, in order to sort out the various effects. What was found was that the resistance was an extremely sensitive probe of changes in magnetic order of the system. These studies have provided the first direct evidence for the existence of an intermediate state, and a measurement of H_c over the entire temperature range for which the material is antiferromagnetic.

Three *c* plane discs were spark cut from two single crystals grown by different commerical firms. Subsequently, the surfaces were spark planed to remove surface damage brought about by the cutoff procedure. The final step in the sample preparation was to chemically polish the surfaces to further reduce surface damage. Sample A had a diameter of 6.0 mm and a thickness of 0.12 mm, and samples B and C had diameters of 8.0 mm and thicknesses of 0.24 mm.

A modified four-probe ac method was used to measure the sample resistance as a function of applied magnetic field. Two small sharp-pointed brass probes placed 3 mm apart, located symmetrically about the center along a crystallographic axis, were used to provide the necessary electrical contacts. Four wires were attached to the two probes. One pair was used to provide connections to a constant current (50-mA) ac source at 10 kHz. The remaining pair was connected to the input of a 10-MΩ-input-impedance lock-in amplifer used to measure the voltage drop across the sample. Tests indicated this to be a reliable way to measure the resistance, although only two contacts were actually attached to the surface of the specimen. This sample-contact system did not give rise to any observable end effects,[20] as can be seen by comparing our data to that given by Mackintosh and Spanel.[19]

The sample was mounted in a copper-can assembly, in such a way that it could distort freely under the influence of thermal and magnetostrictive stresses. This sample mounting was found to be a critical and essential part of the experimental procedure. When the sample was constrained, sharp anomalies at field-induced transitions were smeared out and became unresolvable. The general features of the curves were highly reproducible over the course of this study, approximately 20 runs. Occasionally it was possible to resolve the first resistance anomaly into two distinct peaks. This point is presently a subject of further study.

A variable-temperature Dewar system employing cold helium gas was used to provide an ambient temperature for the copper sample enclosure slightly lower than the desired operating temperature. A heater in intimate contact with the sample enclosure provided for fine control of the temperature, which was stabilized to ±10 m °K using an automatic regulator system employing a field-insensitive capacitance thermometer.[21] The temperature was measured using a calibrated gold-iron versus chromel-P thermocouple with an accuracy of ±0.5 °K. Magnetoresistance curves were plotted directly on an x-y recorder. Although we report here only the data taken between 179 and 85 °K, measurements were taken from 300 to 4.2 °K during the various runs, starting from room temperature.

Our typical longitudinal-magnetoresistance curves (not shown) have the same general shape as

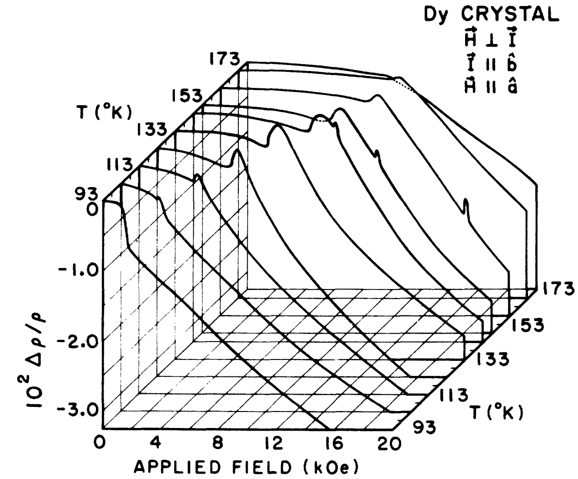

FIG. 1. Summary of typical transverse-magnetoresistance measurements for several temperatures. Broad resistance anomalies are correlated with the critical field H_c. Peaked resistance anomalies observed at the several higher temperatures are believed to be due to spin fluctuations at the transition field H_f. In general, the data indicate that a magnetic field has a small effect on the basal-plane resistance of the distorted helix ($H < H_c$), but does have comparatively large effects on the fan ($H_c < H < H_f$) and ferromagnetic ($H > H_f$) states.

data given by Mackintosh and Spanel.[19] Transverse-magnetoresistance data in the basal plane apparently have not been published, and our data for this case differ from the longitudinal results in several important details. In general, the transitions at the critical field H_c are ound to be sharper. Narrow peaked resistance anomalies at higher fields, indicating phase transitions, have also been observed. Typical magnetoresistance results for the helical phase are shown in Fig. 1. In general, the data show that the sample's resistance decreases slowly, until at a critical value of the magnetic field, a large resistance anomaly occurs, usually resulting in a local maximum. With further increase of the applied field, the resistance decreases more rapidly, and at a higher temperature, a second peaked anomaly is observed.

If one compares this data to the data on magnetization[6] and magnetostriction,[7] it is easy to see that the first resistance anomaly occurs at the critical field H_c which collapses the helicoid. The critical-field resistance anomaly appears to be completely insensitive to the relative directions of the current and field in the basal plane. To verify the result, we have studied the following four configurations: $\vec{I} \parallel \vec{H} \parallel \hat{a}$, $\vec{I} \parallel \vec{H} \parallel \hat{b}$, $\vec{I} \perp \vec{H} \parallel \hat{a}$, and $\vec{I} \perp \vec{H} \parallel \hat{b}$, and see no consistent differences. Use of this experimental technique has permitted the determination of a field which "destroys" the helix to within 1 °K of the Néel temperature. The curve bounding

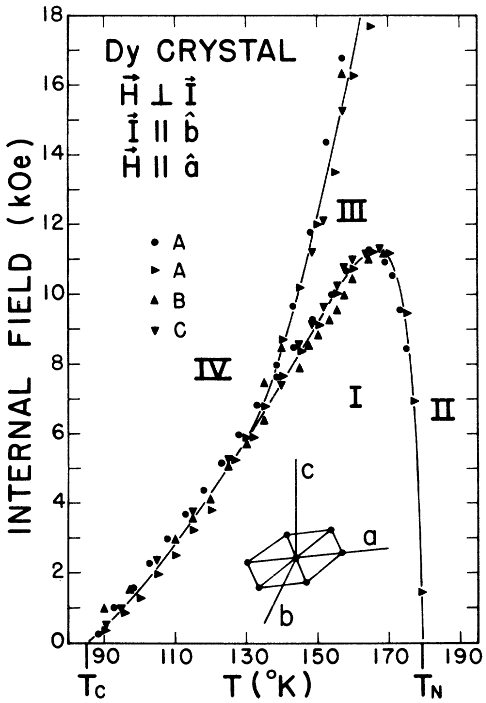

FIG. 2. Schematic H-T phase diagram for dysprosium deduced from typical magnetoresistance data from three different samples. The labels I, II, III, and IV refer to the antiferromagnetic, paramagnetic, fan, and ferromagnetic states, respectively. It has not yet been possible to draw a boundary separating regions II and III. The symbols used on the graph indicate data from samples A, B, and C.

region I in Fig. 2 summarizes these results.

The curve separating regions III and IV in Fig. 2 was drawn by connecting the H-T data points for which sharp resistance anomalies were observed for fields greater than H_c. We believe that these local resistance maxima are the result of spin fluctuations at the transition field H_f, which results in large increases in the sample's resistance. Interpreted in this way, these data provide the first direct evidence for an intermediate (fan) state in dysprosium. It is interesting to note that the field for which a discontinuity in the slope of the magnetostriction data at 144 °K[7] is observed, correlates with our value for H_f, providing appropriate demagnetization corrections are made. We expect this correlation to hold for all temperatures between 130 and 160 °K. Clearly, there must be another line separating regions II (paramagnetic) and III (fan), but our data have provided no information which would enable the drawing of such a boundary. Neutron-diffraction experiments may be required to resolve this question.

The resistance anomalies observed could also result from changes in superzones[22–24] which result from the different periodicity of the chemical and magnetic unit cells. In previous work on terbium,[25] a magnetic field has been shown to suppress the superzones. It appears that final diagnosis of the origin of these resistance effects must await further neutron-diffraction work and detailed studies of the Fermi surface.

It is perhaps important to note that anomalies corresponding to H_f were observed only with one current-field configuration, $\vec{I} \perp \vec{H} \parallel \hat{a}$. Further, it is interesting to note a slight change in the character of the field-dependent resistance deviation between 120 and 133 °K. The basal-plane anisotropy,[26] which varies as a high power of the reduced magnetization, becomes negligible in this temperature interval.

The magnetoresistance data presented here are for increasing field; hysteresis is generally observed which vanishes as T approaches T_N.

In summary, it has been shown that resistivity measurements provide a sensitive means to monitor field-induced phase transitions in rare-earth metals. These studies have indicated the existence of an intermediate (fan) state for temperatures greater than 130 °K. Further, it is noted that the interpretation of microwave-spectroscopy measurements cannot be done without careful consideration of the basal-plane magnetoresistance, which may vary by more than 4% in a 20-kOe field range. The sample geometry is seen to be a factor of considerable importance, for, if the sample is cylindrical, as in the case of the measurements of Ref. 19, inhomogeneous internal magnetic fields may not allow the observation of important effects. Details of the sample mounting were also found to be of considerable importance, suggesting that magnetostriction plays a vital role in these transitions.

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