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# Spin Configurations

## Neutron Diffraction Investigation of Magnetic Ordering in Dysprosium

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Neutron diffraction measurements on a single crystal of dysprosium show that the magnetic structure in the antiferromagnetic region between 179° and 87°K closely resembles a helical-type arrangement of the atomic moments. In this arrangement the moments within a hexagonal layer are aligned parallel and point in a direction perpendicular to the  $c$  axis of the crystal. The moment direction in adjacent layers is rotated by a specific angle which is dependent on the temperature of the sample. A slight modification of this structure exists below about 140°K, and a transition to ferromagnetism occurs at 87°K.

MEASUREMENTS of the magnetic properties<sup>1-3</sup> and specific heat<sup>4</sup> of polycrystalline dysprosium showed that this metal is ferromagnetic below 85°K, antiferromagnetic between 85°K and 179°K, and paramagnetic above 179°K. A very thorough single crystal magnetic investigation by Behrendt, Legvold, and Spedding<sup>5</sup> confirmed these results and showed that

dysprosium is highly anisotropic with the spontaneous magnetic moments always oriented parallel to the planes of the hexagonal layers. In addition, these single crystal measurements showed that the ferromagnetic-antiferromagnetic transition can be raised to higher temperatures by external magnetic fields perpendicular to the  $c$  axis and that anisotropy exists within the hexagonal layers at all temperatures below approximately 110°K.

Neutron diffraction investigations have been initiated on the same single crystal of dysprosium, and the preliminary results are in agreement with the magnetic measurements. The major magnetic behavior is indicated in Fig. 1(a), which shows the scattered neutron intensity near the position of the (002) reflection as a function of the sample temperature. Above 179°K there is no magnetic coherent scattering and only the (002) nuclear reflection is observed. Below 179°K antiferromagnetic reflections appear on each side of the nuclear reflection, and, as shown for the (002<sup>-</sup>) reflection, the intensities increase with a Brillouin-type dependence until a temperature of about 90°K is reached. At this temperature, the antiferromagnetic reflections begin to disappear and the magnetic scattering is found in the (002) reflection, thereby indicating a transition to the ferromagnetic state. In these experiments most of the antiferromagnetic-ferromagnetic transition occurred in a temperature interval of about three degrees, and the temperature at the halfway point was 87°K. A thermal hysteresis was observed in the transition, so that for increasing sample temperatures, the corresponding value was 92°K.

In the antiferromagnetic region, the magnetic structure, which causes the satellite reflections on each side of the nuclear reflections, corresponds to a helical-type arrangement of the magnetic moments. The moments are oriented in a direction perpendicular to the hexagonal  $c$  axis and form ferromagnetic sheets of moments in the hexagonal layers with a specific angle of rotation between moments in adjacent layers. The separation of a given pair of satellites provides a direct measurement of the interlayer turn angle, and the variation of this angle with temperature is shown in Fig. 1(b). At the Néel temperature, the angle is about 43.2°, and it

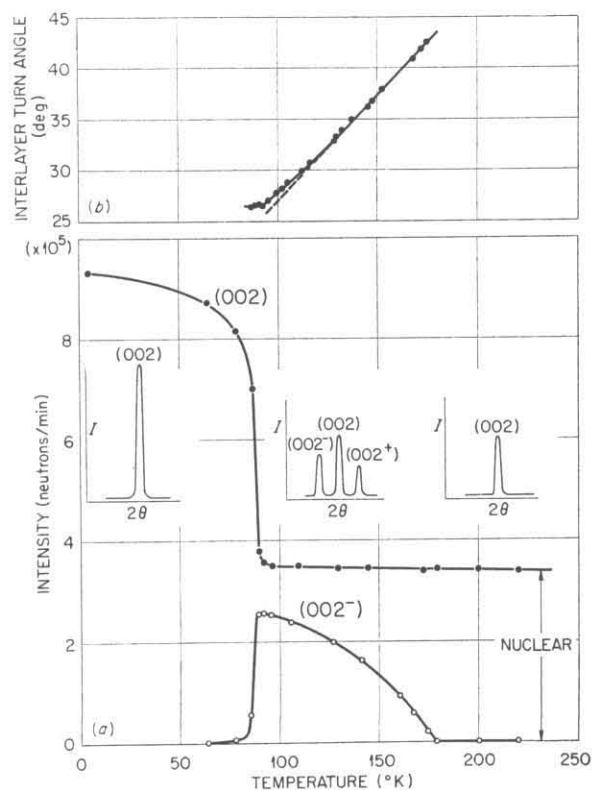


FIG. 1. (a) Temperature variation of the intensities of the (002) and (002<sup>-</sup>) reflections. (b) Temperature variation of the interlayer turn angle in the helical arrangement of magnetic moments.

<sup>1</sup> F. Trombe, *Compt. rend.* **221**, 19 (1945).

<sup>2</sup> F. Trombe, *Compt. rend.* **236**, 591 (1953).

<sup>3</sup> J. F. Elliott, S. Legvold, and F. H. Spedding, *Phys. Rev.* **94**, 1143 (1954).

<sup>4</sup> M. Griffel, R. E. Skochdopole, and F. H. Spedding, *J. Chem. Phys.* **25**, 75 (1956).

<sup>5</sup> D. R. Behrendt, S. Legvold, and F. H. Spedding, *Phys. Rev.* **109**, 1544 (1958).

decreases to about  $26.5^\circ$  at the temperature at which the transition to the ferromagnetic state starts to occur. Although the variation with temperature is nearly linear, there is a definite departure from linearity at about  $130^\circ\text{K}$ . In the ferromagnetic region, dysprosium appears to be a normal ferromagnet with the magnetic moments parallel or closely parallel to the hexagonal layers. The total amount of coherent magnetic scattering is essentially continuous across the transition and indicates merely a reorientation of the moments in this process. However, the value of the saturation atomic magnetic moment perpendicular to the  $c$  axis that was obtained for both the antiferromagnetic and ferromagnetic structures is slightly less than the maximum value of  $10.0\mu_B$  that was observed in the magnetic measurements. It is not yet known if this difference, which amounts to about  $0.5\mu_B$ , is significant.

In addition to the major characteristics of the magnetic structures indicated in Fig. 1, a minor modification of the helical structure must exist between  $140^\circ\text{K}$  and the ferromagnetic transition, since additional very weak antiferromagnetic reflections were observed in this temperature region. These reflections appear to be second harmonics of the  $(00l)$  satellites with an intensity variation that is characteristic of a second-order transition. The very weak intensities cause an uncertainty in the value of the transition temperature of perhaps 10 degrees. The occurrence of these reflections could be

associated with the nonlinear variation of the data in Fig. 1(b), and it is reasonable to assume that they may also be connected with the anisotropy in the basal plane that was observed in the magnetic measurements below  $110^\circ\text{K}$ . However, the exact changes in the magnetic structure that cause these reflections have not yet been determined.

Neutron diffraction experiments were also performed with the single crystal in an external magnetic field. When the field was applied parallel to the  $c$  axis of the crystal, the magnetic properties were the same as those observed in zero field. However, as shown in the magnetic measurements, when the field was applied perpendicular to the  $c$  axis the ferromagnetic transition was raised to higher temperatures. In this connection, it is interesting to note that the variation of the turn angle with temperature shown in Fig. 1(b) was not affected by the field. As the ferromagnetic transition was raised to higher temperatures, it occurred at the value of the turn angle corresponding to that temperature.

#### ACKNOWLEDGMENTS

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### Neutron Diffraction Study of Metallic Erbium

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Neutron diffraction measurements were made on erbium single crystals in the temperature range  $298^\circ$ – $4.2^\circ\text{K}$ . The material is antiferromagnetic below  $80^\circ\text{K}$  and ferromagnetic below  $20^\circ\text{K}$ . In the antiferromagnetic region, the magnetic scattering consists of satellite reflections corresponding to a modulation of the magnetic scattering amplitude along the  $c$  axis. The spacing and intensity distribution of these satellites show two distinct subregions of antiferromagnetism. In the upper region, between  $80^\circ$  and  $52^\circ\text{K}$ , the data suggest a sinusoidal modulation of the magnitude of the  $c$ -axis component of magnetic moment with a period of  $3.5c_0$ . Between  $52^\circ$  and  $20^\circ\text{K}$  the wavelength of the modulation varies continuously from  $3.5^\circ c_0$  to  $4.0c_0$ . In addition, there is a squaring up of the modulation and a simultaneous ordering of the component of the moment normal to the  $c$  axis. Below  $20^\circ\text{K}$  the material is basically ferromagnetic with a moment of  $7.2\mu_B$  directed parallel to the  $c$  axis.

MAGNETIC studies performed on polycrystalline<sup>1</sup> and single-crystal<sup>2</sup> samples of metallic erbium indicate an antiferromagnetic transition temperature of  $80^\circ\text{K}$  and a paramagnetic Curie temperature of  $42^\circ\text{K}$ . Critical field type magnetization isotherms were observed, and extrapolation of the results to zero field yielded a Curie temperature of  $20^\circ\text{K}$ . A neutron diffrac-

tion investigation<sup>3</sup> of polycrystalline erbium showed the presence of both the ferromagnetic and antiferromagnetic states, but because of the complexity of the patterns, the antiferromagnetic structure could not be determined. Single-crystal neutron diffraction measurements with sample temperatures ranging from  $298^\circ$  to  $4.2^\circ\text{K}$  have now been made, and the results form the basis of this report.

The magnetic neutron scattering from erbium shows

<sup>1</sup> J. F. Elliott, S. Legvold, and F. H. Spedding, *Phys. Rev.* **100**, 1595 (1955).

<sup>2</sup> S. Legvold (private communication).

<sup>3</sup> W. C. Koehler and E. O. Wollan, *Phys. Rev.* **97**, 1177 (1955).