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Observation of transitions to spin-slip structures in single-crystal holmium

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We present the results of magnetization measurements on single-crystal holmium using a SQUID magnetometer in the temperature range from 4.2 to 140 K in magnetic fields from 0.01 to 3 T applied along the *b* axis. Our magnetization data shows the Néel temperature to be $T_N=132$ K. In addition, we observe anomalies in the temperature dependence of the magnetization at 21, 42, and 98 K. These anomalies can be accounted for within the spin-slip model; Bohr *et al.* [Physica **140A**, 349 (1986)] have pointed out the existences of ferrimagnetic structure's spin slip and Cowley and Bates [J. Phys. C **21**, 4113 (1988)] additional structures that breaks the hexagonal symmetry.

The magnetic structure of holmium was first determined by Koehler *et al.*[1] Since then, the magnetic properties of holmium have been studied by several authors using a variety of experimental techniques.[2] In zero field, holmium has a basal-plane helical antiferromagnetic state between the Néel temperature, $T_N=132$ K, and the Curie temperature, $T_c=20$ K.[1] However, a significant amount of thermal hysteresis is known to be present at this transition. Below T_c, a component of the magnetic moment tilts out of the plane along the *c* axis, forming a conical ferromagnetic spiral structure. Several authors have observed anomalies at 96 and 24 K in ultrasonic attenuation,[3,4] thermal expansion,[5] and specific-heat measurements[6] using single crystals of holmium. Steinitz *et al.*[7] have observed anomalies at 16.5, 19, and 22 K, in zero-field specific-heat and thermal-expansion measurements on the same sample used in the present study. Using synchrotron radiation, Gibbs *et al.*[8] have observed higher-order commensurate phases with $Q=\frac{2}{11}c^*$ and $Q=\frac{5}{27}c^*$ and interpreted these results using the spin-slip model. Recently, Cowley and Bates[9] have performed a high-resolution neutron-diffraction study of holmium. They found that a parametrized spin-slip model could account for the intensity of the large number of diffraction peaks they observed. Bohr *et al.*[10] have pointed out that some commensurate spin-slip structures are ferrimagnetic with a net moment in the basal plane of holmium, in particular the structure with a spin-slip spacing of 11 atomic layers. This was further developed by Bates *et al.*,[11] who showed that structures characterized by an average spin-slip spacing $b=(3N-1)$ layers break the hexagonal symmetry and suggested this to be the reason for observed anomalies at 19.8, 24.5, 40.5, and 97.4 K in ultrasonic velocity measurements; *b* being 11, 8, 5, and 2, respectively.

We undertook an attempt to observe these anomalies through magnetization measurements on a well-characterized single crystal of holmium, using a SQUID (superconducting quantum interference device) magnetometer. By applying the magnetic field parallel to the *b* axis (easy axis) in the basal plane, we had very high sensitivity to basal-plane ferromagnetism as well as some sensitivity to structures that breaks the hexagonal symmetry. The sample, prepared by D. Fort of the University of Birmingham, was provided by H. Astrom of the Royal Institute of Technology in Stockholm, Sweden and had a residual resistivity ratio (RRR) between 80 and 90. This compares with the RRR of about 24 of the Dalhousie University sample examined in earlier papers.[5,12] It is in the shape of a half-disk of diameter 6 mm, thickness of 1.05 mm, and mass 97.8 mg. We have measured the temperature dependence of the magnetization in the temperature range from 4.2 to 140 K in a SQUID magnetometer supplied by Quantum Design, Inc. of San Diego, California. Data with **H** along the *c* axis in the temperature range from 85 to 130 K have been previously reported.[12]

The temperature dependences of the magnetization at various constant magnetic fields applied parallel to the *b* axis are shown in Fig. 1 in the temperature range from 5 to 140 K. For $H=0.01$ T and $H=0.1$ T the magnetization should be referred to the scale to the right of the figure and for $H=0.5$ T and $H=3$ T to the scale to the left of the figure, see the arrows in the figure. An enlargement of the data, normalized for display purposes, in the 5–40-K range is shown in Fig. 2. In a field of 0.01 T we observe a pronounced peak at 20 K. This peak is by far the strongest signal observed and it seems plausible that it corresponds to the ferrimagnetic structure with one spin-slip for every 11 atomic layers ($Q=\frac{2}{11}c^*$). In a 0.1-T field this peak is replaced by a broad feature between 19 and 24 K. Though the broad peak may be the signature of the two overlapping peaks, it cannot be ruled out that it is the external magnetic field that stabilizes the $Q=\frac{2}{11}c^*$ ferrimagnetic structure over the corresponding temperature interval. Locking to this structure has previously

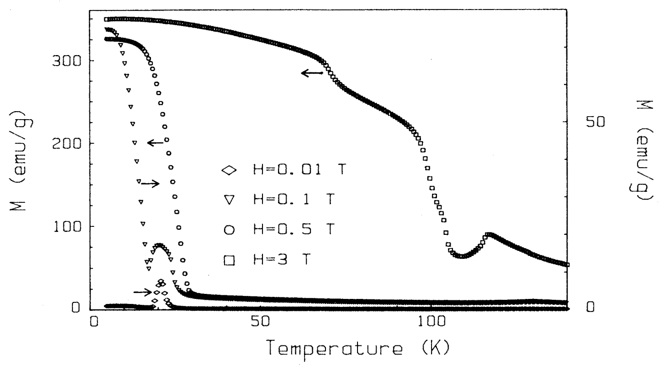

FIG. 1. Magnetization (M) as a function of temperature from 5 to 140 K at four different magnetic fields along the b axis of the Ho single crystal. For $H=0.01$ T and $H=0.1$ T the magnetization should be referred to the scale to the right of the figure and for $H=0.5$ T and $H=3$ T to the scale to the left of the figure, see the arrows in the figure.

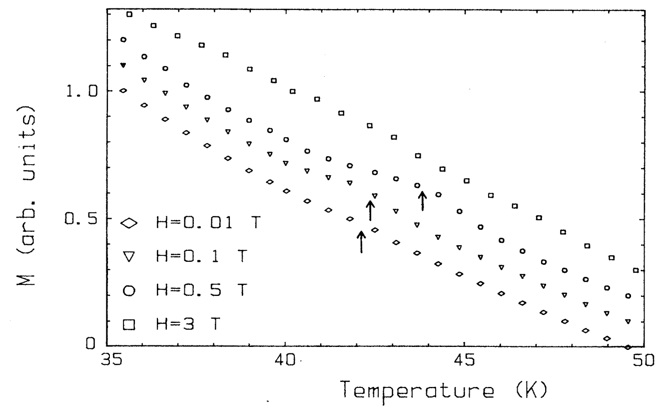

FIG. 3. Normalized magnetization (M) as a function of temperature from 35 to 50 K at four different magnetic fields along the b axis of the Ho single crystal.

been observed by Koehler *et al.*[13] For $H=0.1$ T a rise in the magnetization is seen below $T=17$ K. This is the transition to the fan structure observed by Koehler *et al.*[13] At higher fields this transition happens at correspondingly higher temperatures. For $H=3$ T the structure is ferromagnetic at 4.2 K.[13] The enlargement of the normalized results in the range 35 to 50 K (Fig. 3) shows a small broad peak at 42 K for 0.01 T. This anomaly shifts to higher temperatures at higher fields, as shown by arrows in Fig. 3. At 42 K the Q vector of the magnetic spiral is known to be approximately $Q=0.20c^*$.[1,9,11] This corresponds to a spin-slip separation of 5. Figure 4 shows the normalized magnetization in the temperature range 90–105 K at various fields. We observe an anomaly at 98 K in 0.01 T which shifts to higher temperatures as the field increases. This anomaly at 98 K is very weak, but reproducible, and is clearly observed in the dM/dT vs T curve (at 0.01 T) shown in the inset of Fig. 4. At 98 K the Q vector is known to be approximately $Q=0.25c^*$ (Refs. 1, 9, and 11) corresponding to a spin-slip separation of 2.

In conclusion, we have observed anomalies in the temperature dependence of magnetization at 21, 42, and 98 K in low magnetic fields. To our knowledge this is the first time these transitions have been observed using magnetization measurements. It appears reasonable to us that the anomalies that we observe at these temperatures in the magnetization should be due to transitions to commensurate spin-slip structures that are characterized by spin-slip spacings of 11, 5, and 2 layers, respectively. The structure with a spin-slip spacing of 11 atomic layers is ferrimagnetic as pointed out by Bohr *et al.*[10] Further, while the structures with an average spacing of 5 or 2 atomic layers do not have a net moment, they still break the hexagonal symmetry as pointed out by Bates *et al.*[11] A detailed study of the field and temperature dependence of magnetization in the entire temperature range from 4.2 to 140 K for all three a, b, and c crystal axes will be presented elsewhere.

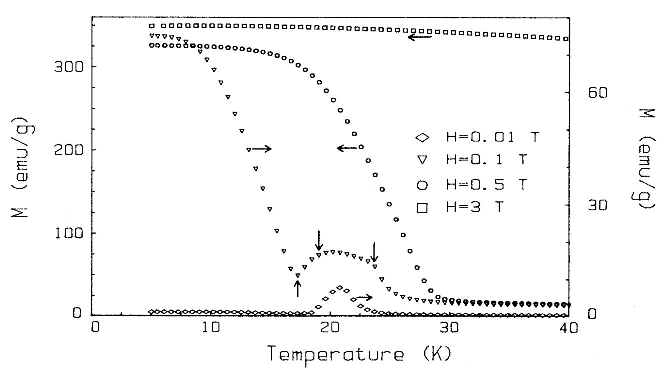

FIG. 2. Normalized magnetization (M) as a function of temperature from 5 to 40 K at four different magnetic fields along the b axis of the Ho single crystal. For $H=0.01$ T and $H=0.1$ T the magnetization should be referred to the scale to the right of the figure and for $H=0.5$ T and $H=3$ T to the scale to the left of the figure, see the arrows in the figure.

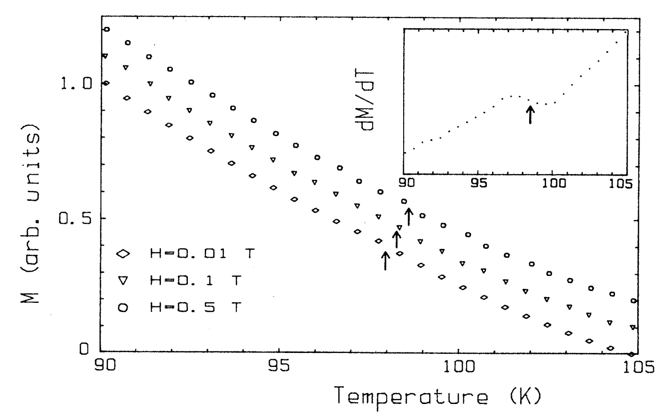

FIG. 4. Normalized magnetization (M) as a function of temperature from 90 to 105 K at three different magnetic fields along the b axis of the Ho single crystal. The inset shows the dM/dT between 90 and 105 K in a field of 0.01 T.

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