Observation of an intermediate phase in dysprosium near the Néel point by neutron diffraction

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The magnetic structure of dysprosium near the point of magnetic disordering has been studied as a function of the thermal history of the sample by neutron diffraction. An intermediate vortex phase appears during cooling from the paramagnetic phase and then converts into a helicoidal phase.

The heavy rare-earth metals terbium, dysprosium, and holmium have a simple spiral magnetic structure below the magnetic disordering temperature.[1] Recent studies of the heat capacity,[2] the magnetic susceptibility,[3] and the linear-expansion coefficient[4] of dysprosium near the Néel point T_N, however, have revealed several anomalies: a difference in the critical behavior of the heat capacity and of the electrical resistance below and above[2] T_N; the appearance of an additional, anomalous magnetic moment, perpendicular to the base plane[3]; and a jump in the elongation along the hexagonal axis about 6 K below the Néel point.[4] In Refs. 3 and 4, the presence of anomalies depended on the thermal history of the sample and the direction in which the temperature was being changed (a heating or a cooling). The observed anomalies in the magnetic susceptibility and in the thermal expansion are seen only when the sample is being cooled from the paramagnetic phase. It has been shown that all the experimental data can be explained under the assumption that a magnetic vortex structure similar to that described in Ref. 5 for an easy-plane magnetic material arises in the dysprosium near the disordering temperature.

A phase transition accompanied by the formation of a vortex state is characteristic of two-dimensional systems and is a consequence of the absence of a long-range magnetic order from such systems. The suggestion of the existence of a vortex state thus contradicts the experiments observations of a spiral structure. This contradiction mandated a more-detailed neutron-diffraction study of the magnetic structure of dysprosium near T_N.

The experiments were carried out on the neutron diffractometer of the Institute of Physics of the Academy of Sciences of the Georgian SSR. The sample is a plate whose plane is perpendicular to the hexagonal C axis, cut from a block of single crystals from which a sample used previously[4] in measurements of the linear-expansion coefficient had been cut. The ratio of the electrical resistances of this sample at 300 and 4.2 K is 17. The experiments are carried out in series with an identical thermal history. In one series of experiments, the sample is cooled to 130 K, i.e., into the region of the helical state, and then heated to the temperature of interest before the measurement is taken. In another set of experiments, the sample is heated to 200 K, i.e., into the region of the

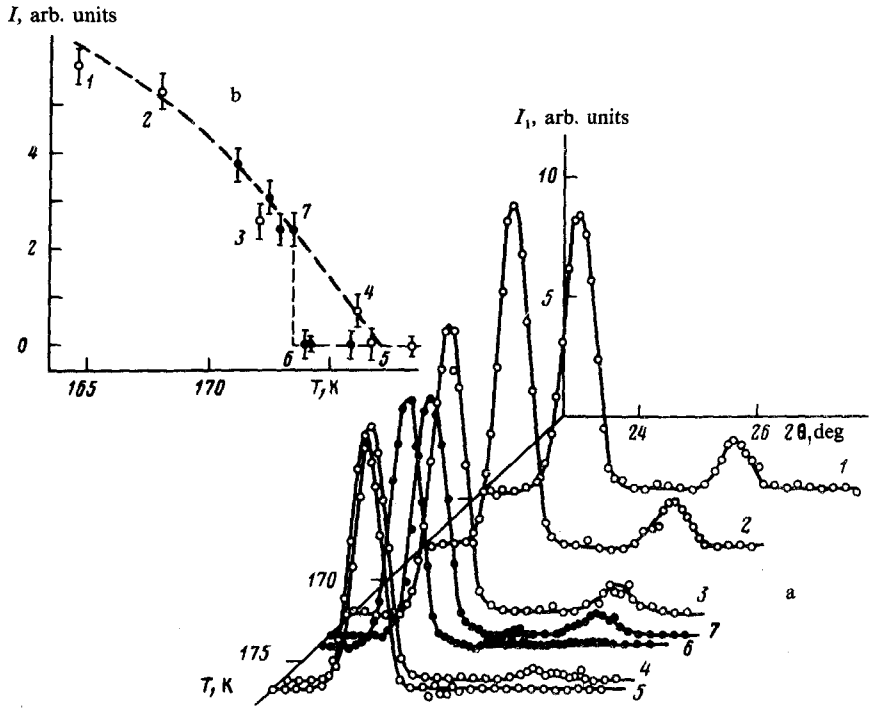

FIG. 1. Neutron diffraction patterns of dysprosium near the Néel point. a: Intensity versus the angle at fixed temperatures. b: Temperature dependence of the integral intensity of the magnetic satellite. ○—Experimental points recorded during heating; ●—during cooling of the sample.

disordered state, and then cooled to the temperature of interest. The accuracy at which the temperatures are determined and held constant is $\cong 0.1$ K in all the experiments. On each neutron diffraction pattern we measure the intensity of the (002) nuclear reflection, that of the magnetic satellite, and also the angular position of the magnetic satellite with respect to the nuclear reflection.

Figure 1 shows some characteristic neutron diffraction patterns and a plot of the integral intensity of the magnetic satellite [I] versus the temperature near the disordering point. We see from this figure that the temperature dependence of the intensity of the magnetic satellite takes different forms, depending on whether the sample is heated from the helicoidal phase or cooled from the paramagnetic phase. In the case of the heating, the satellite is seen up to 176.2 K (curve 4), and as the temperature is raised, the intensity of this satellite falls to the background level, disappearing at the Néel point. During cooling, the satellite does not appear before 174 K (curve 6), and at 173.7 K its intensity changes abruptly and significantly (curve 7). The values of T_N, found from the minimum of the anomaly in the linear-expansion coefficient, are identical during the heating and cooling of the sample,[4] 178.5 K. The abrupt increase in the intensity of the magnetic satellite at 173.7 K, in agreement with the jump in

length observed near this temperature,[4] indicates that there is a second phase transition in the dysprosium during cooling, in addition to the phase transition at the Néel point.

According to these results, as a sample is heated, the helicoidal long-range order of the spiral structure persists up to T_N, while during cooling, despite the phase transition at 178.5 K, the long-range order of the spiral-structure type does not appear before 713.7 K. This behavior means that in the temperature interval 173.7–178.5 K a magnetic structure different from a helicoidal structure exists when the sample is cooled. Consequently, the absence of a long-range order from the intermediate phase, which exists in the interval 178.5–173.7 K, during the cooling, the appearance of an additional magnetic moment along the C axis (Ref. 3), and the behavior of the heat capacity which is described approximately by a Kosterlitz-Thouless function,[2] indicate that a vortex state similar to that described in Ref. 5 arises in this interval.

On the theoretical side, the question of a spatial ordering of vortices present in a sufficiently high concentration is an open question. According to estimates in Ref. 3, in the case of the appearance of a lattice of vortices, the period of this lattice would be greater than the period of the spiral structure of dysprosium. The slight anomaly between the nuclear reflection and the satellite on neutron diffraction patterns 6 and 7 may be evidence of such an ordering.

We might also note that the satellite observed during the heating of the sample apparently has a shape more complex than Gaussian (curves 3 and 4). A splitting of the satellite may be a consequence of the commensurability[6] of the magnetic spiral structure and of the crystal structure at 173 K (Refs. 7 and 8), but this question requires a more detailed experimental and theoretical study.

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