Study of magnetic properties of single crystal holmium in weak magnetic fields

O.V. Snigirev, A.M. Tishin and A.V. Volkozub

Department of Physics, Moscow State University, 119899 Moscow, USSR

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The study of the magnetisation of high purity single crystal holmium in weak magnetic fields of $10^{-10^3}$ A/m has been made over a temperature range of 4.2-200 K. The data obtained indicate the presence of a spontaneous magnetic moment in basal plane at $T < 20$ K. Thermal hysteresis of magnetic moment has been observed in the vicinity of magnetic phase transitions paramagnetism-helicoidal antiferromagnetism-conical ferromagnetism with the wave vector along the $c$-axis. The existence of this hysteresis indicates that both transitions are of first order.

1. Introduction

It is established that in the temperature range below $T = \Theta_1 = 20$ K conical ferromagnetic ordering takes place in holmium. For example, at 4.2 K the magnetic moment components of holmium in the basal plane and along the hexagonal axis $c$ are equal to 9.5 and $1.7\mu_B$, respectively [1,2]. Neutron-scattering study has shown that the application of a magnetic field $H > 4 \times 10^5$ A/m in the basal plane at $T = 4.2$ K results in the transformation of the conical ordering into the ferromagnetic structure [2]. It was stated in refs. [1,2] that at zero field the temperature $\Theta_1$ is the conical ferromagnetism–helicoidal antiferromagnetism first-order transition point.

The experiments made in recent years using neutron diffraction and synchrotron radiation on holmium substantially transform the data about this low temperature magnetic phase transition [3–6]. For example, the values of the wave vector of the cone structure at 4.2 K obtained by different authors are essentially different (see, for instance, refs. [6,1,5]). The additional first-order phase transition has been found between two commensurable values of the wave vector $2/11$ and $1/6$ in terms of $c^*$ [3] in the temperature range below $\Theta_1$.

The helicoidal antiferromagnetic structure is destroyed in holmium at $T = \Theta_2 = 132$ K. In the region of $T < 50$ K under neutron-scattering examination [1], magnetic satellites of the 5th and 7th order are observed which indicate strong deflection of magnetic moments to the direction of the axes of easy magnetisation in the basal plane. A thorough study of the behavior of the 3rd order satellites allowed [6] to assume the asphericity in the magnetisation distribution of holmium.

The data about the character of the magnetic phase transition at point $\Theta_2$ are also essentially different. It was stated in ref. [9] that the character of the phase transition from the paramagnetic state to the helicoidal structure depends on the kind of magnetic interaction. If the exchange energy exceeds significantly the spin–orbit or dipole–dipole forces, then only the first-order transition is possible. The character of the phase transition can also be influenced by short-range order fluctuations that increase as they approach this second-order transition point [9,10]. Similar conclusions are obtained by calculations carried out by the renormalisation group method [11]. Experi-
mental data on neutron diffraction \[12\], heat capacity \[13\], thermal expansion \[17\] demonstrated this transition to be a second-order one. At the same time, the results of the investigation of heat expansion \[14\], latent heat transition \[16\], muon spin relaxation \[15\] and neutron diffraction \[5\] indicate the presence of a first-order transition.

The analysis of these data as well as data about the effect of nonmagnetic impurities on the magnetic properties of holmium \[18,19\] indicates that the perfection of the structure and the purity of the sample have a noticeable effect on the temperature of magnetic phase transitions, the character of the helicoidal antiferromagnetism-paramagnetism phase transition and other properties.

The purpose of this paper is to perform a careful study of the magnetic behaviour of holmium in a temperature region of 4.2 K < \( T \) < 200 K in weak magnetic fields in \( 10^{-1} \text{ A/m} \), which do not transform magnetic ordering.

2. Experiments

The measurements of the magnetic moment have been carried out by the SQUID-susceptometer \[7,8\]. A sample with the form of a thread \( \approx 2 \text{ mm} \) long and with a mass of 9.5 mg was glued on to the quartz support and placed in a magnetometer ampoule. The magnetic moment was measured in constant magnetic field applied in basal plane \( (H \parallel b) \). The absolute error of the measured moment did not exceed 10%. The sensitivity of the installation to the magnetic moment was \( 5 \times 10^{-12} \text{ A m}^2/\text{Hz}^{1/2} \).

Since, as mentioned above, structural perfection and sample purity can essentially influence the results of the investigation, single crystal holmium thread was used in the work reported here. The thread was obtained by the vacuum sublimation technique in the graphite-heated resistance furnace at remnant pressure \( \approx 10^{-6} \text{ Torr} \).*

Holmium was purified when it was precipitated on water-cooled condenser in the form of crystal druses grown not closely together. The impurity composition of sublimated holmium was checked by mass-spectrometric and extraction techniques. The analysis of the content of 20 impurity elements showed that the main number of impurities (17 from 20) was within \( 10^{-3} \text{ to } 10^{-5} \) at\%, the contents of the impurities of copper, europium and neodymium was \( 10^{-2} \) at\%. Our investigation showed that the ratio of the sample specific resistances at \( T = 300 \) and 4.2 K was \( R_{300}/R_{42} \approx 50 \). It follows from the X-ray data that the sample was oriented in the basal direction along the \( b \)-axis.

3. Results and discussion

The temperature dependences of the sample magnetic moment on cooling and heating in the magnetic field \( H = 104 \text{ A/m} \) are shown in fig. 1. On cooling from the paramagnetic phase a maximum is observed on the curve \( M(T) \) at \( T = 18.5 \text{ K} \). As the temperature increases the momentum first decreases to the point \( T = 19.3 \text{ K} \) and then increases taking a maximum value at \( T = 21.1 \text{ K} \). As can be seen from fig. 1, on thermocycling a hysteresis is observed that extends to temperatures into the paramagnetic state. A similar character of the curves \( M(T) \) was observed as the field increases up to \( 10^3 \text{ A/m} \), no significant displacement in the temperature of maxima and minimum of the magnetic moment being observed (see fig. 2).

In the temperature range of 14–19 K a number of additional peculiarities is observed on sample heating from helium temperatures (see fig. 3). It was assumed in refs. \[3,4\] that repetitive defects (spin slipping) may appear in the magnetic structure of holmium and some other heavy rare-earth metals. It was shown that within the framework of this model one can account for the experimentally observed features of the behaviour of the wave vector. Such deviations of the magnetic ordering of holmium may result in ferrimagnetic ordering in the region of commensurable magnetic structure \[3\].

* The sample prepared by O.D. Chistyakov at Moscow Institute of Metallurgy.
Fig. 1. Temperature dependences of the magnetic moment of a holmium single crystal on cooling and heating in the magnetic field $H = 104 \, \text{A/m} \ (H \parallel b)$.

Fig. 2. Temperature dependences of the magnetic moment of a holmium single crystal on sample heating in magnetic field. 1: $H = 29 \, \text{A/m}$; 2: $H = 104 \, \text{A/m}$; 3: $H = 223 \, \text{A/m}$; 4: $H = 940 \, \text{A/m} \ (H \parallel b)$. 
The presented data seem to indicate the presence of a spontaneous magnetic moment in the basal plane at temperatures lower than 20 K and may be accounted for by the existence of ferromagnetic ordering within the limits of the basal plane. It should be noted that the presence of hysteresis at $T > 10$ K is in sufficiently good agreement with the neutron-graphical data [3] and
supports the possibility of the coexistence of magnetic phases in the temperature range of $\approx 10$–$20$ K.

The anomalies presented in fig. 3 seem to indicate a still more complex character of the reconstruction of the magnetic ordering. It must be noted that the points of their positions, to a certain degree, coincide with the temperatures at which there are anomalies of calorimetric data.

As is evident from fig. 4, the temperatures of paramagnetism–antiferromagnetism transitions on heating and cooling are quite different ($\Theta_2 = 133$ K on heating and $\approx 120$ K on cooling the sample). The transition is accompanied by vanishing of the temperature hysteresis at $T = 162$ K. The data obtained in this work and presented in figs. 1 and 4 indicate that paramagnetism–antiferromagnetism and antiferromagnetism–ferromagnetism transitions are first-order phase transitions.

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References


