# Phase transition in holmium "in terms of the magnetic field"

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An x-ray diffraction study of the crystal structure of holmium at 77–292 K is carried out in magnetic fields up to 16 kOe. At $T = 77$ K and $H = 9$ kOe a discontinuous change occurs in the crystal lattice parameters ($\Delta a < 0$, $\Delta C > 0$). This first-order phase transition "in terms of the magnetic field" is ascribed to the distortion of the helicoidal antiferromagnetic structure.

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It is well known that the helicoidal antiferromagnetic structure (a simple helix, SS) which is typical of many rare earth and transition metals and alloys at $T_c < T < T_N$ is destroyed in a moderately high magnetic field. In several theoretical papers[1–5] it was shown that the direct transition from a helicoidal antiferromagnetic state to a ferromagnetic state, which takes place in the transition "with respect to temperature" at $T_c$ (strictly speaking, at $\Theta_1$) can not occur in a magnetic field. The "transition with respect to the magnetic field" should occur in two stages: from the helicoidal state to the intermediate "fan" state (first-order transition) and from the fan phase into the ferromagnetic state (second-order transition).

The distortion of the helicoidal magnetic structure in a magnetic field has been observed in a neutron-diffraction study of $MnAu_2$[6] and holmium.[7] Unfortunately the $H-T$ diagrams of holmium given in[7] are not exact because they contain two-phase regions, points of quaternary equilibrium, etc.

Holmium is obviously the best object (at least among rare earth metals) where one can search for the phase transition between the helicoidal and intermediate phases, because the magnetic properties of holmium have been investigated thoroughly enough.[8–11] The behavior of the magnetic properties, as well as the results of neutron-diffraction measurements in a magnetic field,[7] lead to the assumption that there should be an intermediate magnetic state. In the absence of a magnetic field holmium has a helicoidal SS structure in the temperature region 20-132 K and an FS conical structure if $T < 20$ K.[12] The present paper presents the results of an x-ray study of the structural effects in holmium caused by the application of an external magnetic field up to 16 kOe in the temperature region 77–300 K.

The procedure used for the study of the structure in a magnetic field was described by us earlier.[13] The object of the investigation was a polycrystalline sample of holmium 99.5% pure. The x-ray photographs were taken in the radiation of a chromium anticathode. The geometry of the experiment made it possible to record reflections from the planes (210), (105), (203), (212), and (114) of the hexagonally close-packed lattice of holmium. The lattice parameters were calculated from the (210) and (105) lines; the relative error in the measurements of the parameters $a$ and $c$ did not exceed $\sim 5 \cdot 10^{-5}$ (at room temperature).

X-ray studies of holmium in a magnetic field were performed at $T < T_N$ (77, 115, 130 K) as well as at $T > T_N$ (145 and 292 K). The results of the measurements of the parameter $c$ are given in Fig. 1. At all temperatures (except $T = 77$ K) a very small variation of the lattice parameters was observed (the dependences of $a$, $c$, and $c/a$ on $H$ were approximated by straight lines because the magnetostriction was very weak). Actually the difference between the measured values of the magnetostriction coefficient ($a_i^{-1} da_i/dH$) and zero was not larger than the experimental error at $T > 77$ K. On the other hand the magnetostriction coefficients of other helicoidal antiferromagnets—dysprosium and terbium—are relatively large, especially near $T_N$.

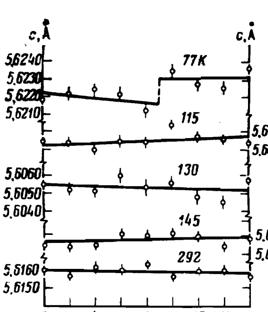


FIG. 1. Isotherms of the field dependence of the parameter $c$ of holmium.

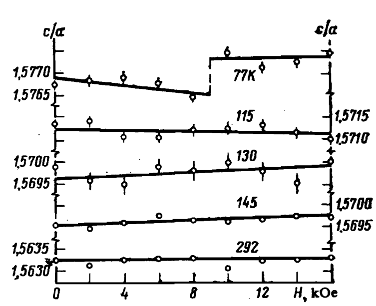


FIG. 2. Isotherms of the field dependence of the axis ratio $c/a$ of holmium.

At 77 K a decrease of the parameters $a$ and $c$ was observed in magnetic fields up to 9 kOe. At $H \sim 9$ kOe, small (but well pronounced) jumps of parameters of the crystal lattice of holmium take place: $\Delta a/a \simeq -(10 \pm 10) 10^{-5}$, $\Delta c/c \simeq (25 \pm 10) \cdot 10^{-5}$. The jump is most pronounced at the axis ratio $(c/a)^{-1}\Delta(c/a) \simeq (35 \pm 10) \cdot 10^{-5}$ (see Fig. 2) It should be emphasized that the abrupt change of the lattice parameters of holmium at 77 K in the 9-kOe magnetic field was repeatedly reproduced in a number of experiments (Figs. 1 and 2 show the results of one of the experiments at 77 K).

The jump of the parameters of the crystal lattice shows unequivocally that the first-order phase transition "in terms of the magnetic field" takes place in holmium at 77 K. Using the Clapeyron-Clausius equation

$$q = \frac{T\Delta V}{dT/dp}, \qquad (1)$$

where $\Delta V = 2\Delta a + \Delta c$ is the change of the volume at the phase transition, one can calculate the transition heat $q$. Using the measured value of $\Delta V = 1.49 \cdot 10^{-3}$ Å$^3$ and the value $dT_c/dP = -0.39$ deg/cal taken from[16] we get $q = 4.25$ cal/mole. We should stress that the substitution of $dT/dP$ for the SS→FS transition in Eq. (1) is not completely justified because it appears that in this case the helicoidal structure is not transformed into a conic structure (see below). This is therefore only a crude estimate of $q$. Using the well known corollary of the same Clapeyron-Clausius equation

$$\Delta\sigma = \Delta V \Big/ \frac{dH_{cr}}{dT}\frac{dT}{dp}, \qquad (2)$$

one can estimate the jump $\Delta\sigma$ of the magnetization in the first-order phase transition. The value of $dH_{cr}/dT$ for the SS→FS transition is $2.5 \cdot 10^2$ Oe/deg and the estimate of $\Delta\sigma$ gives $1.65\ \mu_B$.

What changes of the magnetic structure can be related to the first-order phase transition "in terms of the magnetic field" observed in holmium? It is difficult to assume that this transition is due to the transformation of the helicoidal antiferromagnetic structure into a conic ferromagnetic one, because the change of the volume at the phase transition at $T = 77$ K, and $H = 9$ kOe is appreciably larger than the value of $\Delta V$ in the SS→FS transition.[17] 1) The critical field causing the transition is considerably smaller than the value of $H_{cr}$ causing the ferromagnetic transformation.[8–10] Apparently, the phase transition in holmium is due to a transformation of the helicoidal phase into some intermediate phase (probably into the "fan" structure).[6,7] This agrees with the presence of singularities on the $\sigma(H)$ curves of holmium in the temperature region $T_c < T < T_N$, preceding the transition into the ferromagnetic state[9]; to be sure, these singularities were observed at lower temperatures and in stronger fields than in our experiments.

1) The value of $\Delta V$ in the antiferromagnetic–ferromagnetic transition increases almost linearly with decreasing temperature.[14]


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