EFFECT OF MAGNETIC FIELD ON THE CRYSTAL STRUCTURE OF DYSPROSIUM

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The effect of magnetic fields up to 16 kOe on the crystal structure of polycrystalline dysprosium in the paramagnetic (300°K), antiferromagnetic (145°), and ferromagnetic (77°K) states is studied. It is shown that a first-order antiferromagnetism-ferromagnetism transition accompanied by lowering of symmetry of the crystal lattice from hexagonal to rhombic occurs at 145°K in a field of ~10 kOe. A certain type of dynamic texture is formed in the ferromagnetic state, which is related to the tendency of the magnetic moments to become oriented at a minimal angle with respect to the field.

A study of the crystal structure of metals in magnetic fields, together with the measurements of such properties as magnetization, coercive force, magnetostriction, etc., can yield valuable information concerning the thermodynamics and kinetics of magnetic phase transitions, the electronic and magnetic structure of metals, the magnetic deformation of the crystal lattice, etc. However, no experiments on the influence of the magnetic field on the crystal structure of metals have yet been performed, although the x-ray technique in moderately strong stationary magnetic fields (up to 20 kOe) has been well worked out for such objects as liquid crystals[1]. The study of the magnetic structure of metals in external magnetic fields has recently become part of the practice of x-ray structure analysis[2].

Great interest attaches to an investigation of the effect of a magnetic field on the crystal structure of rare-earth metals, in which transitions occur between varying types of magnetic structures under the influence of temperature and pressure, as well as that of the magnetic field. The optimal object for the study of the influence of the magnetic field on the crystal structure is dysprosium. Neutron-diffraction investigations[3] have shown that in the interval 85–178°K there is observed in dysprosium a helicoidal magnetic structure of the type of a simple helix, namely, the magnetic moments lie in the basal planes (001) of a compact hexagonal (hcp) lattice, and the angle of the helicoid increases from 43.2° at T_N to 26.5° at T_C; below 85°K, the magnetic moments are collinearly ordered in the basal planes. It has been shown by x-ray diffraction[4] that a rhombic distortion of the hcp lattice takes place at the Curie point, and a jump of the atomic volume is observed; this means that the antiferromagnetism–ferromagnetism transformation (with respect to temperature) is a first-order phase transition. The helicoidal magnetic structure of dysprosium is destroyed by a magnetic field[5], and the maximum field required to destroy the antiferromagnetic structure (H_{cr}) is ~11 kOe (at T_N). The question of the nature of the antiferromagnetism–ferromagnetism phase transition in a magnetic field and of the ensuing structure changes remains open, and the present paper is devoted mainly to its solution.

EXPERIMENTAL PROCEDURE

The object of the investigation was 99.2% pure polycrystalline dysprosium. The investigated sample, in the form of a disc of 4 mm diameter and 1 mm thick, cooled by a stream of nitrogen vapor[6], was placed in the gap of an electromagnet fed with direct current. The sample was rotated in a plane perpendicular to the direction of the incident x-ray beam, and the incident and reflected beams (S_0 and S) and the magnetic field intensity vector (H) were all in one plane (Fig. 1). Such a geometry has made it possible to obtain x-ray diffraction patterns by the back-photography method, in which the reflecting planes make small angles with the field[1)], and the recorded diffraction pattern pertains to crystallographic planes whose normals lie in the same plane as the vector H (so-called longitudinal photography).

The temperature was measured with a differential chromel-alumel thermocouple and stabilized accurate to 1–2°K. The magnetic field intensity ranged from 0 to 16 kOe, and was maintained during the experiment with accuracy ±0.1 keV. Diffraction reflections from the planes (203), (210), (211), and (105) of the hcp dysprosium lattice were registered in chromium radiation. The accuracy with which the distances between planes (Δd) were measured was $\pm 1 \times 10^{-4}$ Å. The intensity of the diffraction lines was estimated visually.

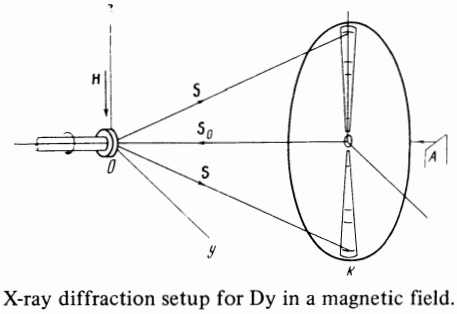

FIG. 1. X-ray diffraction setup for Dy in a magnetic field. O – sample, K – cassette with film, A – x-ray tube.

1) In this case this angle lies in the range ~ 6 – 18°.

As a rule, all the photographs were obtained in an increasing magnetic field intensity.

RESULTS

The variation of the diffraction pattern as a function of the magnetic field intensity in the paramagnetic (300°K), antiferromagnetic (145°K), and ferromagnetic (77°K) regions are shown in Fig. 2.

In the paramagnetic region, no noticeable change is observed in the x-ray interference intensity as a function of the magnetic field intensity.

In the antiferromagnetic region, in fields 0—9 kOe, there are likewise no changes in the form of the x-ray patterns, but in a field of ~10 kOe the diffraction pattern changes appreciably: all the x-ray lines shift noticeably, the diffraction peak from the plane $(210)_h$[2] splits into two components. The splitting of the $(210)_h$ line is evidence of a lowering of the symmetry of the crystal lattice to rhombic[4]. It is seen from Fig. 2, however, that the remaining diffraction lines do not split. Further increase of the magnetic field intensity leads to a redistribution of the intensity between the diffraction lines $(150)_r$ and $(310)_r$ (the former weakens, the latter becomes stronger) and a noticeable attenuation of the $(151)_r$ line[3].

In the ferromagnetic region, even without an applied magnetic field, the most intense diffraction peaks (the "former" hexagonal lines $(203)_h$ and $(210)_h$) are split into two components, since the crystal lattice has rhombic symmetry in this temperature range[4]. With increasing magnetic field intensity, an attenuation of the diffraction lines $(043)_r$ and $(151)_r$ is observed, as a result of which the diffraction pattern becomes similar to the x-ray pattern obtained at 145°K in a maximum field, although a noticeable redistribution of the intensities of the $(310)_r$ and $(150)_r$ lines, which take place at 145°K when $H \geq 12$ kOe, could not be observed even in a field of ~16 kOe.

In addition, at 77°K, a certain smearing of the diffraction maxima is observed with increasing magnetic field intensity, as a result of which the accuracy with which the positions of the peaks are determined is somewhat lower. The smearing of the lines was observed also at 145°K and $H > 10$ kOe.

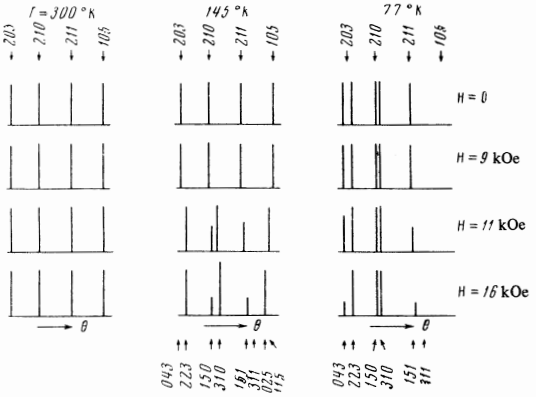

FIG. 2. X-ray patterns of dysprosium. The heights of the lines are proportional to the intensities of the x-ray reflection. The hexagonal indices are indicated on the top and the rhombic ones on the bottom.

In addition to the visual estimate of the intensities of the x-ray lines, we measured the interplanar distances as functions of the applied field. The curves

$$d_{(203)_h}(H) \text{ and } d_{(210)_h}(H) \text{ or } d_{(223)_r}(H),\ d_{(150)_r}(H) \text{ and } d_{(310)_r}(H)$$

were used to calculate the periods a, b, and c for the hcp or rhombic lattices of dysposium. The obtained field dependences of the lattice parameters are shown in Figs. 3 and 4.

In the paramagnetic region, the dependence of the periods a and c of the hcp lattice on the magnetic field intensity is quite weak, but a certain compression of the crystal lattice along the hexagonal axis is noticeable.

In the antiferromagnetic region, the dependence of the periods of the crystal lattice on the magnetic field intensity has a rather complicated character. Whereas the parameters a and c of the hcp lattice change quite little with the field in fields 0—9 kOe, a jumpwise lowering of the symmetry to rhombic takes place at $H = 10$ kOe (H_{cr}), as already noted. At $H > H_{cr}$, the period b increases noticeably with the field, while the period c decreases, and the period a changes insignificantly. The degree of rhombic distortion of the hcp lattice (b/a) naturally increases in this case (Fig. 5).

In the ferromagnetic region, no strong dependence of the period of the rhombic lattice on the magnetic field intensity is observed. The ratio of the periods (b/a) is practically independent of the field, and its

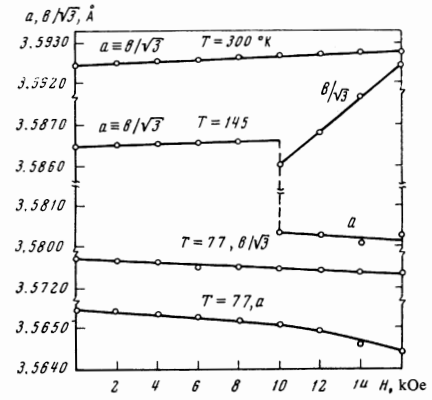

FIG. 3. Field dependence of the periods a and b.

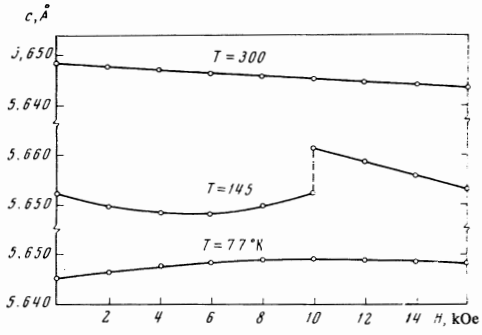

FIG. 4. Field dependence of the period c.

[2] The subscripts h and r stand for hcp and rhombic lattices.

[3] The geometry of the setup does not make it possible to register the line $(311)_r$, which is the second component of the split $(211)_h$ peak.

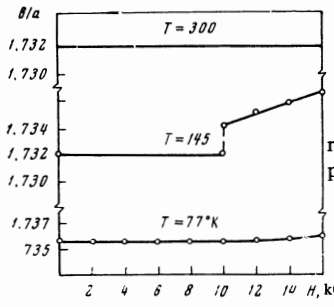

FIG. 5. Field dependence of the ratio of the periods b/a in the basal plane.

magnitude at 77°K corresponds approximately to the value of 145°K in the maximum field.

We note in conclusion that in the ferromagnetic region there is a certain disparity between the experimental and calculated values of the interplanar distances, although no such disparity is observed in the paramagnetic and antiferromagnetic states. Apparently, the strong magnetic anisotropy of dysprosium in the ferromagnetic state leads to an appreciable difference between the effective magnetic field acting on the different grains.

DISCUSSION

The results allow us to draw a qualitative picture of the behavior of polycrystalline dysprosium in moderately strong constant magnetic fields.

When $T \gg T_N$ (300°K in our experiments), the crystal structure of dysprosium is stable in fields up to 16 kOe. This agrees with the results of the magnetic measurement[5]: strong magnetostriction in the paramagnetic state is observed only in very strong fields (up to 150 kOe[7]).

Then $T_C < T < T_N$, the hcp lattice is stable in fields up to 10 kOe (145°K). It can be assumed that in such fields there occurs a distortion of the helicoidal magnetic structure, namely, the angle between the directions of the magnetic moment in the neighboring basal planes decreases[8], and at a magnetic field intensity 10 kOe (H_{cr}) a helicoidal structure of the type of a simple helix becomes unstable. When $H > H_{cr}$, the magnetic moments are collinearly ordered in the basal planes, while at H_{cr} the symmetry of the crystal lattice drops to rhombic and the periods of the lattice change jumpwise (Figs. 3--5). Thus, the antiferromagnetism—ferromagnetism transformation in dysprosium in a magnetic field is a first-order phase transition.

Attention is called to the fact that at 145°K, certain lines are missing from the diffraction picture of rhombic dysprosium in fields stronger than H_{cr}; in addition, in the ferromagnetic region there is a redistribution of the intensity between the diffraction maxima (Fig. 2). This phenomenon can be treated as formation of a specific "magnetic" texture in the polycrystalline sample: the magnetic moments, tending to become aligned at a minimum angle to the field, determine the orientation of the rhombic domains in polycrystalline dysprosium. This texture is dynamic, for owing to the rotation of the sample in the magnetic field (Fig. 1) the angles between the easy-magnetization axes and the field change all the time. In spite of the large magnetic viscosity of dysprosium[9], the low rate of rotation of the sample does not hinder the formation of a magnetic texture. An increase in the rate of rotation of the sample can apparently lead to a destruction of the texture. The fact that at a lower temperature (77°K) the texture effects are less strongly pronounced (Fig. 2) may be connected with the increased domain dimensions (and consequently with the decrease of their mobility) with decreasing temperature[10].

The increased smearing of the lines on the x-ray patterns of ferromagnetic dysprosium (at 77°K and at 144°K in fields stronger than 10 kOe) with increasing field may be connected with the appearance of large magnetoelastic stresses[5].

DEDUCTIONS

We have investigated the influence of a magnetic field of intensity up to 16 kOe on the crystal structure of polycrystalline dysprosium in the paramagnetic, antiferromagnetic, and ferromagnetic states. The main results of this investigation are as follows:

1. In the paramagnetic and ferromagnetic states, the crystal lattices (hcp and rhombic, respectively) of the dysprosium are stable.

2. In the antiferromagnetic state, when the critical value of the magnetic field intensity is reached, the change of the magnetic structure is accompanied also by a distortion of the crystal lattice (with a lowering of its symmetry to rhombic).

3. The antiferromagnetism—antiferromagnetism transformation in the magnetic field is a phase transition of first order.

4. In a magnetic field and in the ferromagnetic state, a dynamic magnetic structure is produced because of the tendency of the magnetic moments to become oriented at a minimum angle to the magnetic field intensity vector.

CONCLUSION

The purpose of this investigation was to consider the fundamental aspects of the behavior of paramagnetic, antiferromagnetic, and ferromagnetic dysprosium in a magnetic field. We propose to study in the future the temperature and angular dependences of the observed structure effects. Quantitative information concerning the magnetic deformation of the crystal lattice should yield analogous investigations for single crystals of dysprosium.

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