

INFLUENCE OF PRESSURE ON THE MAGNETORESISTANCE AND THE CRITICAL FIELD OF A DYSPROSIUM SINGLE CRYSTAL

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Submitted December 9, 1969

Zh. Eksp. Teor. Fiz. 59, 351-355 (August, 1970)

An investigation was made of the influence of pressure on the magnetoresistance of a dysprosium single crystal in the temperature range 85-177°K. It was found that the pressure-induced shift of the critical field depended strongly on the temperature. A thermodynamic analysis of the experimental data for dysprosium showed that the increase in the critical field with increasing pressure, observed in the range 110-140°K, could be explained by an enhancement of the exchange interaction. A correlation was found between the values of dH_{CR}/dp and dH_{CR}/dT , as well as between the magnetostriction and dH_{CR}/dT . This correlation showed that the decrease in H_{CR} below 160°K, which led to the transition of dysprosium to the ferromagnetic state, was due to an anomalous increase in the lattice parameter c below the temperature of the transition from the paramagnetic to the helicoidal antiferromagnetic state.

INVESTIGATIONS of the influence of pressure on the magnetic properties of rare-earth metals are of considerable interest because the magnetoelastic energy of these metals should play a far greater role in those metals than it does in classical ferromagnets. This is because of the very high value of the magnetostriction of rare earths.^[1-4] Of particular interest is the influence of the magnetoelastic energy on the transition from the helicoidal antiferromagnetic to the ferromagnetic state, observed at a temperature Θ_1 in the absence of a magnetic field, and on the critical field H_{CR} , which destroys the helicoidal magnetic structure and induces ferromagnetic ordering. We shall concentrate our attention on the temperature dependence of dH_{CR}/dp of a dysprosium single crystal throughout the range of temperatures in which the helicoidal structure exists in fields $H < H_{CR}$. This approach eliminates the magnetic anisotropy effects and makes it possible to obtain more information than that which could be deduced from the available data on $d\Theta_1/dp$ in weak fields,^[2,5,6] and on dH_{CR}/dp in polycrystalline samples.^[5]

The critical field was determined by measuring the magnetoresistance in fields up to 15 kOe at pressures $p = 1$ or 5000 atm. Hydrostatic pressure was established using the method described by Balla and Brandt.^[7] A dysprosium single crystal, measuring $0.9 \times 0.5 \times 2.5$ mm, was placed in a nonmagnetic beryllium-bronze bomb. The long axis of the sample, along which the magnetic field was applied, coincided with one of the crystallographic directions lying in the basal plane. The pressure was transmitted by a mixture of kerosene and transformer oil. The electrical resistance and the magnetoresistance were determined by the potentiometric method. The pressure was measured with a Manganin manometer.

It is evident from Figs. 1 and 2 that the negative magnetoresistance increases rapidly from a certain critical value of the magnetic field H_{CR} . The corresponding decrease in the resistance caused by the application of the field is due to the transition, at

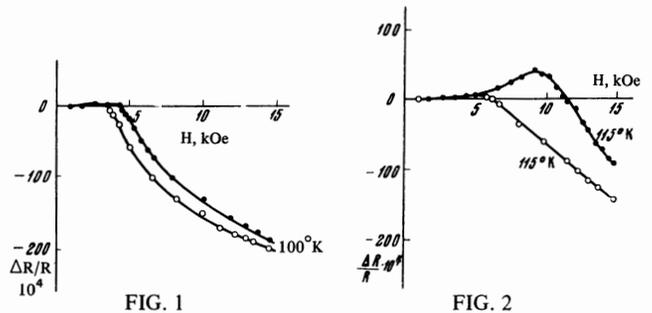


FIG. 1. Longitudinal magnetoresistance of a dysprosium single crystal along a direction lying in the basal plane, recorded at 100°K at pressures of 1 atm (open circles) and 5000 atm (black dots).

FIG. 2. Longitudinal magnetoresistance of a dysprosium single crystal along a direction lying in the basal plane, recorded at 115°K at pressures of 1 atm (open circles) and 5000 atm (black dots).

$H = H_{CR}$, from the helicoidal to the ferromagnetic state, in which conduction electrons are scattered less strongly by magnetic inhomogeneities. The application of a hydrostatic pressure of 5000 atm shifts the steep part of the curve parallel to itself in the direction of stronger magnetic fields; the shift is 1500 Oe at 100°K and 5000 Oe at 115°K. This means that H_{CR} increases by the same amount. Figure 3 shows the temperature dependence of dH_{CR}/dp . Near the temperature $\Theta_2 = 177^\circ\text{K}$, corresponding to the transition from the paramagnetic to the helicoidal antiferromagnetic state, the effect of pressure is small but it increases at lower temperatures, reaching a maximum at 120°K; then it decreases again on approach to $\Theta_1 = 85^\circ\text{K}$.

According to our experimental results and those published previously,^[2] the critical field H_{CR} , whose value is governed by the exchange interaction between layers, decreases very strongly from 10.5 kOe to zero when the temperature is reduced from 160 to 85°K. Below Θ_2 , dysprosium exhibits also a strong increase in the lattice parameter c , which represents the dis-

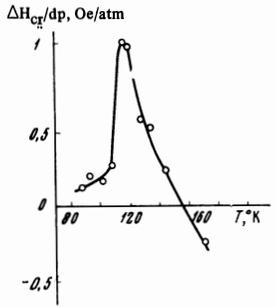


FIG. 3. Temperature dependence of the pressure-induced shift of the critical field of a dysprosium single crystal.

tance between the magnetic layers in the helicoidal structure.^[4] This suggests that the strong temperature dependence of the critical field (and of the helicoid angle) is due to the strong dependence of the energy of the exchange interaction between these layers on the interlayer distance, which increases anomalously below Θ_2 . There should be a correlation between the values of dH_{CR}/dp and dH_{CR}/dT , since in the first case, H_{CR} increases because of the decrease in c under pressure; and in the second case, H_{CR} increases because of the decrease in c due to the heating in the interval $\Theta_1 - \Theta_2$.

We shall investigate the pressure and the temperature dependences of the critical field and establish a relationship between dH_{CR}/dp and dH_{CR}/dT by considering the thermodynamic potential terms which depend on the deformation along the c axis. Following the theory given in^[8,9], we shall assume that the helicoidal structure is due to the exchange interaction. We thus find that the thermodynamic potential contains the following terms: the free energy of the exchange interaction; the magnetoelastic component of the exchange interaction energy, which takes account of the strong dependence of this energy on the parameter c ; the elastic energy; the component of the free energy due to the change in the entropy as a result of the thermal expansion; and the work done by the external magnetic field and by the stress along the c axis (p_c):

$$\Phi = \Phi_0 + F_{exc}(I_s, \omega, T) - \gamma(\omega)I_s^2\lambda_c + \frac{1}{2}E\lambda_c^2 - TE\alpha_c\lambda_c + p_c\lambda_c - IH. \quad (1)$$

Here, I_s is the saturation magnetization; ω is the angle between the magnetic moments of the neighboring layers for $\lambda_c = 0$; $\gamma(\omega)$ is the magnetoelastic constant of the exchange interaction between the layers; $\lambda_c = (c - c_0)/c_0$ is the relative deformation along the c axis (c_0 is the value of the lattice parameter c at 0°K in the absence of any influence of the exchange forces and external stresses); E is Young's modulus; α_c is the thermal expansion coefficient along the c axis at $T > \Theta_2$.

The use of a thermodynamic potential which allows only for the dominant deformation along the helicoid axis (the c axis) has been justified for dysprosium in connection with the magnetostriction,^[2] the temperature dependence of the helicoid angle,^[2,10] and the thermal expansion.^[4,10] In contrast to these investigations, we shall include the external magnetic field energy in the calculations of dH_{CR}/dp and dH_{CR}/dT but we shall not specify the dependences of the exchange parameters and of the magnetoelastic constant γ on

the angle ω in order to make our results more generally applicable.

The condition for a minimum of the thermodynamic potential (1), given by $\partial\Phi/\partial\lambda_c = 0$, yields the relative elongation along the c axis:

$$\lambda_c = \gamma(\omega) \frac{I_s^2}{E} + T\alpha_c - \frac{p_c}{E}. \quad (2)$$

Hence, we can find the magnetostriction $\Delta\lambda_c$ due to the transition at $H = H_{CR}$ from the helicoidal to the ferromagnetic state:

$$\Delta\lambda_c = -\frac{I_s^2}{E} \Delta\gamma(\omega), \quad (3)$$

where $\Delta\gamma(\omega)$ is the change in the magnetoelastic constant at this transition. The helicoidal structure parameter ω changes discontinuously at $H = H_{CR}$ and, therefore, we may assume that the phase transition considered is of the first kind. The thermodynamics of phase transitions of the first kind predicts that $dH_{CR}/dp = \Delta V/\Delta I$. Using the experimental observation that dysprosium is characterized by $\Delta V \approx \Delta\lambda_c$,^[3] we find the quantity of interest to us:

$$\frac{dH_{CR}}{dp} = \frac{I_s^2}{E\Delta I} \Delta\gamma(\omega). \quad (4)$$

Substituting Eq. (2) in Eq. (1), we obtain the thermodynamic potential in the form

$$\Phi = \Phi_0 + F_{exc}(I_s, \omega, T) - \frac{1}{2}E\lambda_c^2 - IH, \quad (5)$$

where λ_c is given by Eq. (2). Equation (5) can be used to calculate the change in H_{CR} due to cooling. This change is the consequence of the temperature dependence of the magnetoelastic energy and is given by:

$$\frac{dH_{CR}}{dT} = \Delta \left(\frac{\partial\Phi}{\partial T} \right)_p \frac{1}{\Delta I} = -\frac{\partial}{\partial T} \Delta \left(\frac{E\lambda_c^2}{2} \right) \frac{1}{\Delta I} = -\Delta \left(\lambda_c \frac{\partial\lambda_c}{\partial T} \right) \frac{1}{\Delta I}. \quad (6)$$

We shall now find whether the temperature dependence of the critical field of dysprosium can be explained by the magnetoelastic energy of the exchange interaction between layers. We shall estimate dH_{CR}/dT using the known values of the magnetostriction^[2,3] and of the thermal expansion coefficients:^[3,4] $\lambda_{2C} = 8.9 \times 10^{-3}$, $\alpha_{2C} = 6.3 \times 10^{-5}$, $\lambda_{1C} = 7.1 \times 10^{-3}$, $\alpha_{1C} = 3.5 \times 10^{-5}$, $\Delta I = 1.97 \times 10^3 \text{ G}$.^[3] Here, and later, we shall use the subscript 1 for the phase with the helicoidal structure and subscript 2 for the ferromagnetic phase. Substituting these experimental values in Eq. (6), we find that $dH_{CR}/dT = 1.2 \times 10^2 \text{ Oe/deg}$, which is close to the value of $dH_{CR}/dT = 1.5 \times 10^2 \text{ Oe/deg}$ found by us for dysprosium in the range $110-140^\circ\text{K}$, and also found in other investigations.^[2]

This result shows that the decrease in H_{CR} of dysprosium due to its cooling in the $\Theta_1 - \Theta_2$ range can be explained as follows. According to the experimental results,^[2-4] cooling below Θ_2 increases the spontaneous deformation of the lattice λ_{1C} and λ_{2C} , and increases the magnetostriction $\Delta\lambda_c$ which accompanies the destruction of the helicoidal order in a field $H = H_{CR}$. The discontinuity in the magnetoelastic energy of the exchange interaction, observed at $H = H_{CR}$,

$$\Delta E_{me} = \frac{1}{2}(E\lambda_{2c}^2 - E\lambda_{1c}^2)$$

increases when the temperature is lowered and this reduces the energy barrier separating the helicoidal

and ferromagnetic structures so that the value of H_{CR} decreases at the rate given by Eq. (6). Other mechanisms have less effect on the temperature dependence of H_{CR} in the $\Theta_1 - \Theta_2$ range, with the exception of the temperatures close to Θ_1 and Θ_2 .

Using $\lambda_{2c} = \lambda_{1c} + \Delta\lambda_c$ and Eq. (4), we obtain a formula relating dH_{CR}/dp and dH_{CR}/dT :

$$\frac{dH_{CR}}{dp} \approx -\frac{1}{\alpha_{2c}E} \frac{dH_{CR}}{dT} - \frac{\alpha_{1c}}{\Delta I} \left(1 - \frac{\alpha_{1c}}{\alpha_{2c}}\right). \quad (7)$$

We shall now substitute in Eq. (7) the published experimental values: $E = 7.5 \times 10^{11}$ dyn/cm²,^[2] $\alpha_{2c} = 6.3 \times 10^{-5}$, $\alpha_{1c} = 3.5 \times 10^{-5}$,^[2-4] $\Delta I = 1.97 \times 10^3$ G at 130°K.^[3] We shall also substitute $dH_{CR}/dT = 1.5 \times 10^2$ Oe/deg, the value obtained by us for dysprosium in the range 110–140°K. These substitutions give $dH_{CR}/dp = 1.6$ Oe/atm. This estimate is in order-of-magnitude agreement with the experimental data on the pressure-induced shift of the critical field in the temperature range 110–140°K. The influence of the magnetic anisotropy in the basal plane on the value of H_{CR} becomes appreciable below 110°K. At these temperatures, the application of a pressure increases the anisotropy constant and, therefore, dH_{CR}/dp decreases. Above 140°K, the effects of the change in the saturation magnetization due to heating and due to the pressure-induced shift of Θ_2 become appreciable. These effects are ignored in Eq. (7).

Thus, our thermodynamic analysis of the experimental data for dysprosium gives the following information. First, the increase in the critical field with increasing pressure can be explained, except at temperatures close to Θ_1 and Θ_2 , by a strong enhancement of the exchange interaction between layers, which occurs when the distance between them is reduced. Secondly, the observed correlation between the values of dH_{CR}/dp and dH_{CR}/dT , as well as that between the magnetostriction and dH_{CR}/dT , shows that the strong temperature dependence of the critical field and the consequent strong temperature dependence of the helicoidal structure periods associated with H_{CR} , can all be explained by the dependences of H_{CR} and ω on the lattice parameter c , which increases anomalously below Θ_2 .

According to the theory due to Dzyaloshinskiĭ,^[9] the

transition from helicoidal antiferromagnetism to ferromagnetism at Θ_1 is due to the temperature dependence of the exchange parameters. Our results are in agreement with this theory^[9] and they show that the temperature dependence of the parameters of the exchange interaction between layers, which governs the value of H_{CR} , can be explained by the dependence of these parameters on c , which increases rapidly below Θ_2 . The strong reduction in H_{CR} caused by cooling below 160°K results finally in the destruction of the helicoidal structure and the establishment of the ferromagnetic order.

The authors are grateful to Professor K. P. Belov for suggesting this problem and for his interest in this investigation.

¹K. P. Belov, R. Z. Levitin, S. A. Nikitin, and A. V. Ped'ko, Zh. Eksp. Teor. Fiz. 40, 1562 (1961) [Sov. Phys.-JETP 13, 1096 (1961)].

²K. P. Belov, R. Z. Levitin, and S. A. Nikitin, Usp. Fiz. Nauk 82, 449 (1964) [Sov. Phys.-Usp. 7, 179 (1964)].

³S. Legvold, J. Alstad, and J. Rhyne, Phys. Rev. Lett. 10, 509 (1963).

⁴F. J. Darnell, Phys. Rev. 130, 1825 (1963); 132, 1098 (1963).

⁵L. I. Vinokurova and E. I. Kondorskiĭ, Izv. Akad. Nauk SSSR, Ser. Fiz. 28, 537 (1964); L. I. Vinokurova, Dissertatsiya (Dissertation), Moscow State University, 1964.

⁶H. Bartholin and D. Bloch, J. Phys. Chem. Solids 29, 1063 (1968).

⁷D. Balla and N. B. Brandt, Zh. Eksp. Teor. Fiz. 47, 1653 (1964) [Sov. Phys.-JETP 20, 1111 (1965)].

⁸S. V. Vonsovskiĭ and Yu. A. Izyumov, Usp. Fiz. Nauk 77, 377 (1962); 78, 3 (1962) [Sov. Phys.-Usp. 5, 547, 723 (1963)].

⁹I. E. Dzyaloshinskiĭ, Zh. Eksp. Teor. Fiz. 46, 1420 (1964); 47, 336 (1964) [Sov. Phys.-JETP 19, 960 (1964); 20, 223 (1965)].

¹⁰P. C. Landry, Phys. Rev. 156, 578 (1967).

Translated by A. Tybulewicz