MAGNETIC ANISOTROPY OF A TERBIUM SINGLE CRYSTAL

K. P. BELOV and Yu. V. ERGIN

J. Exptl. Theoret. Phys. (U.S.S.R.) 50, 560-564 (March, 1966)

The temperature variation of the magnetic anisotropy energy near the temperature of destruction of magnetic ordering (the Curie point, Θ_2) is estimated from magnetization isotherms of a single crystal of terbium, taken along the axes of easy and of difficult magnetization. The effective anisotropy field leads to the phenomenon of an "apparent" lowering of the Curie temperature along the axis of difficult magnetization by about 30°.

. MAGNETIC and neutron-diffraction studies^[1, 2] have shown that terbium is ferromagnetic below the temperature $\Theta_1 = 221^{\circ} K$ and paramagnetic above the temperature $\Theta_2 = 230^{\circ}$ K; in the temperature interval Θ_1 to Θ_2 , or 221 to 230°K, a helicoidal magnetic structure is observed in it; the resultant magnetic moment of each basal plane is turned through a certain angle with respect to the moment of a neighboring plane. A sufficient, small magnetic field (~ 300 Oe) destroys this magnetic structure, and the magnetic moments, which lie in the basal plane, become parallel to one another. Because of the small energy of the helicoidal structure, the behavior of terbium near the point of destruction of the magnetic spin ordering, Θ_2 , is very similar to the behavior of an ordinary ferromagnet near the Curie point. A peculiarity of the magnetic properties of terbium is the presence of a huge energy of uniaxial anisotropy. This anisotropy is present not only below the temperature Θ_2 , but also at temperatures above Θ_2 ; that is, in the paramagnetic range. The paramagnetic susceptibility and the paramagnetic Curie points along the axes of difficult and of easy magnetization are different.

Direct measurements of the magnetic anisotropy of terbium in the ferromagnetic range are very difficult, since they require very strong fields. As is shown below, however, the size of the magnetic anisotropy energy can be estimated from magnetization curves of a single crystal of terbium, taken in the Curie-point region. The method used for this purpose is Landau's thermodynamic method, which was applied earlier to the estimation of the anisotropy of a single crystal of gadolinium.^[3]

2. The single crystal of terbium contained less than 0.1% impurities. It was oriented by the Laue method. The magnetization measurements were carried out by a null method with a magnetometer of the Domenicali type, in fields up to 15,000 Oe. The paramagnetic susceptibility was measured with the same magnetometer in a field of 10,000 Oe. To eliminate the influence of the magnetocaloric effect, the magnetization measurements were made under isothermal conditions.

3. The equation that describes the magnetization curves of a ferromagnet near the Curie point, with allowance for uniaxial anisotropy energy, can be obtained from the theory of second-order phase transitions.^[41] For a ferromagnet with an axis of easy magnetization lying in the basal plane, the thermodynamic potential of the system can be written in the form

$$\Phi = \Phi_0(T) + \frac{1}{2}\alpha\sigma^2 + \frac{1}{4}\beta\sigma^4 + k'\sigma_z^2 - \sigma \mathbf{H}, \qquad (1)$$

where k' is a coefficient proportional to the anisotropy energy, σ is the specific magnetization, and α and β are thermodynamic coefficients that depend on the temperature and pressure. Upon minimizing this potential and neglecting the anisotropy energy in a plane perpendicular to the axis of easy magnetization, we get two equations, which describe the paraprocess magnetization along the axis of difficult magnetization (H = H_z = H_c) and perpendicular to it (H = H_x = H_b):

$$\alpha_{\bar{c}}\sigma_{\bar{c}} + \beta_{\bar{c}}\sigma_{\bar{c}}^3 = H, \qquad \alpha_{\bar{b}}\sigma_{\bar{b}} + \beta_{\bar{b}}\sigma_{\bar{b}}^3 = H, \tag{2}$$

where $\sigma_{\overline{c}}$ and $\sigma_{\overline{b}}$ are the specific magnetizations along the \overline{c} and \overline{b} axes of the crystal, measured in the external magnetic field; $\alpha_{\overline{c}}$ and $\alpha_{\overline{b}}$ are thermodynamic coefficients along the same axes; and the coefficients $\beta_{\overline{c}}$ and $\beta_{\overline{b}}$ are, to a first approximation, equal. Here

$$\alpha_{\overline{c}} = \alpha_{\overline{b}} + 2k'. \tag{3}$$

The Curie point, according to Landau's theory, is taken to be the temperature at which the coefficient α vanishes. The coefficient $\alpha_{\overline{b}}$ is determined by extrapolation of the curves of H/σ as function of σ^2 to the axis of ordinates at each temperature.^[5] The coefficient $\alpha_{\overline{c}}$ cannot be de-



FIG. 1. Magnetization isotherms of a single crystal of terbium along the axis **b**.

termined by this method, because in the fields at present available it is not possible to magnetize a terbium crystal to saturation along the axis of difficult magnetization. The value of the coefficient 2k' can be estimated, however, from data on the anisotropy of the paraprocess susceptibility χ_n = $d\sigma_i/dH$. Here σ_i is the specific magnetization of the paraprocess. It is easily seen that

$$\chi_n = 1 / (\alpha + 3\beta\sigma^2) \tag{4}$$

and consequently

$$2k' = (1/\chi_{\bar{c}} - 1/\chi_{\bar{b}}) - 3\beta \ (\sigma_{\bar{c}}^2 - \sigma_{\bar{b}}^2), \tag{5}$$

where $\sigma_{\overline{c}}$ and $\sigma_{\overline{b}}$ are the specific magnetizations along the axes \overline{c} and \overline{b} of the crystal, measured in the field H, and $\chi_{\overline{c}}$ and $\chi_{\overline{b}}$ are the specific paraprocess susceptibilities along the same axes in the same field H.

The anisotropy energy (per cm^3) is calculated in the following form:



FIG. 2. Dependence of H/ σ on σ^2 for a single crystal of terbium along the axis $\bar{\mathbf{b}}$.



FIG. 3. Temperature dependence of the coefficient α of a single crystal of terbium along axes \overline{b} and \overline{c} .

$$K = \rho k' \sigma^2 = \rho k' (\sigma_s + \sigma_i)^2, \tag{6}$$

where ρ is the density of the metal, σ_s is the specific spontaneous magnetization, and σ_i is the specific paraprocess magnetization. Thus the anisotropy energy can be divided into two parts,

$$K = K_s + K_H, \tag{7}$$

where K_s is the anisotropy energy connected only with the existence of σ_s , whereas K_H is an energy that depends on the field in consequence of the paraprocess.

The "apparent" Curie-point shift in ferromagnetic crystals was predicted theoretically in ^[6] and was first observed experimentally in a single crystal of gadolinium. ^[3] It must be remembered, however, that according to Landau's theory, ^[4] the Curie point is an isolated point in the (T, H) plane, and its value does not depend on the external magnetic field or on the internal magnetic anisotropy field. Introduction of the concept of "apparent" anisotropy of the Curie point enables us to estimate the temperature variation of the magnetic anisotropy energy of rare-earth ferromagnets near the Curie point Θ_2 ; it is at present difficult to do this by a direct method, except for gadolinium.

4. Figures 1 and 2 show magnetization curves and curves of H/σ as function of σ^2 for a single crystal of terbium, along the axis of easy magnetization (the axis b). It is clear that Eq. (2) is well fulfilled for sufficiently large magnetizations; that is, we have to do with the paraprocess.

From the H/ σ curves were calculated the temperature variation of the coefficient $\alpha_{\bar{b}}$ (Fig. 3) and the spontaneous magnetization of the terbium single crystal along the axis \bar{b} (insert in Fig. 2). The Curie point along this axis, determined from the condition that $\alpha_{\bar{b}}$ must vanish, was $\Theta_{2\bar{b}}$ = 227.5 ° K.

The temperature dependence of the coefficient 2k', determined near the Curie point according to formula (5), is shown in Fig. 4. By adding the value of the coefficient 2k', calculated in this way



FIG. 4. Temperature dependence of the coefficient k' and of the constants K, K_s , and K_H for a single crystal of terbium near the Curie point Θ_2 .



FIG. 5. Temperature dependence of the reciprocal of the paramagnetic susceptibility of a single crystal of terbium along axes \overline{b} and \overline{c} .

at each temperature, to the corresponding value of $\alpha_{\rm b}^-$, we obtained (Fig. 3) the temperature variation of the coefficient $\alpha_{\rm c}^-$.

We did not succeed in making measurements of the value of $\chi_{\overline{c}}$ at temperatures below 215°K because of the large ponderomotive forces that acted on the specimen when it was magnetized along the axis of difficult magnetization. As is evident from Fig. 3, approximate extrapolation of the $\alpha_{\overline{c}}(T)$ curve to the temperature axis gives a value $\Theta_{2\overline{c}}$ = 198°K for the Curie point in the direction of the axis \overline{c} . Thus, because of the influence of the effective field due to magnetic anisotropy, the destruction of magnetic order along the axis \overline{c} in a single crystal of terbium occurs at a temperature about 30° lower than in the direction of the axis \overline{b} . This value is in agreement with the anisotropy of the paramagnetic Curie point along the axes of difficult and of easy magnetization (Fig. 5).

The temperature variation of the anisotropy constant, determined according to formula (6), is shown in Fig. 4. It is clear that the constant K_s , connected with the spontaneous magnetization, decreases monotonically and vanishes at the point $\Theta_{2\bar{b}}$. The constant K_H (H = 10 kOe), which depends on the field, has a maximum right at the Curie point and vanishes only far away from it.

Thus we have for the first time succeeded in estimating experimentally the value and the temperature variation of the anisotropy energy of a single crystal of terbium. Its value even near the Curie point Θ_2 is very large—of order 10^7 erg/cm^3 . It is clear also that the value of K depends on the external magnetic field. In a single crystal of terbium the "apparent" shift of the Curie point is appreciably larger (~30°) than in a single crystal of gadolinium (~1.5°).^[3]

We thank Professor A. S. Borovik-Romanov for discussion of the problem.

¹D. E. Hegland, S. Legvold, and F. H. Spedding, Phys. Rev. **131**, 158 (1963).

²W. C. Koehler, J. Appl. Phys. **36**, 1078 (1965); W. C. Koehler, J. W. Cable, E. O. Wollan, and M. K. Wilkinson, J. Phys. Soc. Japan **17**, Suppl.

B-III, 32 (1962).

³K. P. Belov, Yu. V. Ergin, R. Z. Levitin, and A. V. Ped'ko, JETP **47**, 2080 (1964), Soviet Phys. JETP **20**, 1397 (1965).

⁴ L. D. Landau and E. M. Lifshitz, Statisticheskaya fizika (Statistical Physics), Nauka, 1964; Elektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Fizmatgiz, 1959.

⁵K. P. Belov, Magnitye prevrashcheniya (Magnetic Transitions), Fizmatgiz, 1959.

⁶ E. R. Callen and H. B. Callen, J. Phys. Chem. Solids **16**, 310 (1960); E. R. Callen, Phys. Rev. **124**, 1373 (1961).

Translated by W. F. Brown, Jr. 73