"HELICAL" ANTIFERROMAGNETISM OF GADOLINIUM

K. P. BELOV and A. V. PED'KO

Moscow State University

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Magnetic properties of gadolinium were measured in weak fields. The magnetization of gadolinium in weak fields was found to fall rapidly between $\Theta_1 = 210°K$ and the Curie point $\Theta_2 = 290°K$. The magnetization isotherms, $\sigma(H)$, recorded over this range of temperatures, had marked kinks, similar to those occurring in dysprosium at critical fields representing disappearance of "helical" antiferromagnetism. It is concluded that a helical spin configuration exists in gadolinium between Θ_1 and Θ_2 and that this configuration can be destroyed by very weak fields ($H_C \approx 0$—15 Oe).

1. Recent work[1] has shown that the rare-earth metals Dy, Tb, Ho, Er, and Tm have two magnetic transitions. Below a certain temperature Θ_1 ($\Theta_1 = 85°K$ for Dy) these metals are ferromagnetic, above Θ_1 they are antiferromagnetic, and above Θ_2 ($\Theta_2 = 179°K$ for Dy) they become paramagnetic.

Neutron diffraction studies[2] have shown that between Θ_1 and Θ_2 Dy, Er, and Ho possess what is known as a helical spin configuration, which is a special form of antiferromagnetism. In the case of gadolinium the spin configuration has not been established.[3] It has been usually assumed that gadolinium is a "normal" ferromagnet although its electron and crystal structures are identical with those of dysprosium, erbium, and holmium. We shall show below that gadolinium also has the two transition points Θ_1 and Θ_2 and antiferromagnetism between these temperatures. There is, however, one important difference: the antiferromagnetism of gadolinium is destroyed even by weak fields. This instability explains why the antiferromagnetism of gadolinium was not discovered earlier.

2. To obtain reliable magnetization curves in weak fields, measurements were carried out on toroidal samples of polycrystalline Gd. Figure 1 gives the temperature dependence of the specific magnetization, obtained in fields of 0.28 Oe and above. At the temperature $\Theta_1 \approx 210°K$ the magnetization in weak fields (0.28—1.12 Oe) falls sharply and has a very small value up to $\Theta_2 = 290°K$. The transition at Θ_1 does not appear in stronger fields (2.8—112 Oe) although the curves obtained in such fields show clearly some anomaly. In the strongest fields used by the authors (500—2000 Oe) the magnetization curves have a normal Weiss form (Fig. 1).

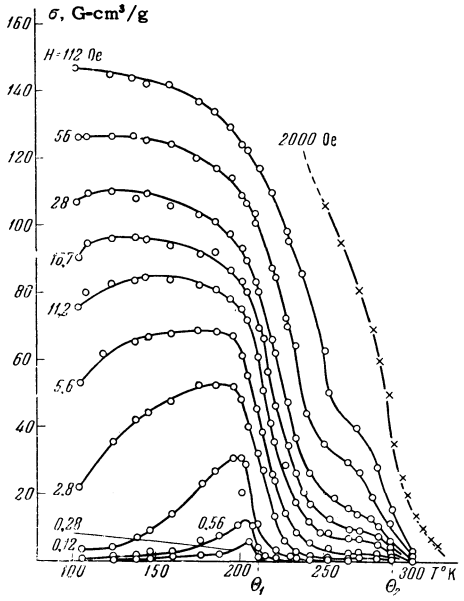

FIG. 1. The temperature dependence of the magnetization of gadolinium in various fields; Θ_1 is the temperature of appearance of antiferromagnetism, Θ_2 is the Curie point.

It has been suggested[3] that gadolinium is antiferromagnetic between Θ_1 and Θ_2 and that the antiferromagnetism exists only in the absence of applied magnetic fields; very weak fields are sufficient to destroy this antiferromagnetism. By analogy with dysprosium, the antiferromagnetism should disappear at a definite critical field H_C.

Figure 2 gives the magnetization isotherms of gadolinium recorded between Θ_1 and Θ_2; they are very similar to the isotherms of dysprosium. The curves of Fig. 2 show definite kinks at critical fields H_C and the magnetization rises more rapidly above these kinks. This can be seen clearly in Fig. 3 where the abscissa scale is enlarged. Below

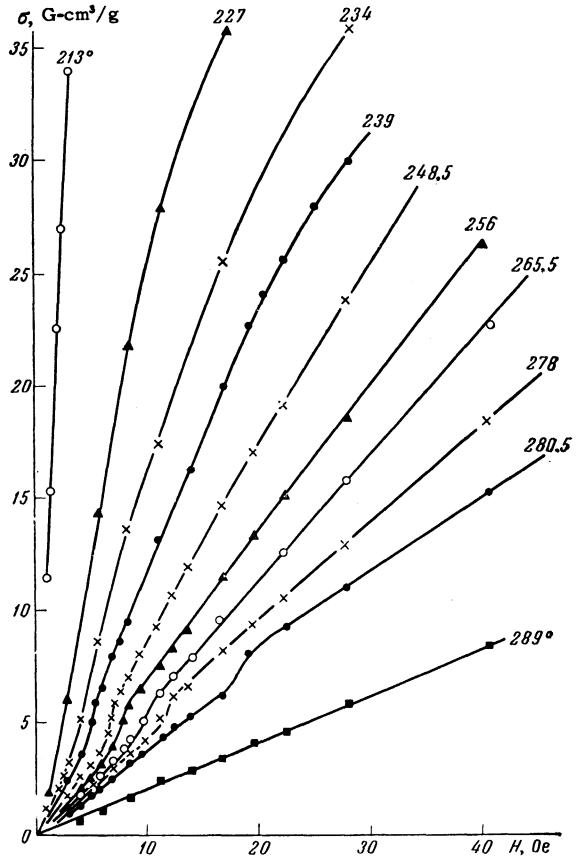

FIG. 2. The magnetization isotherms of gadolinium in the temperature interval Θ_1–Θ_2 in fields up to 40 Oe.

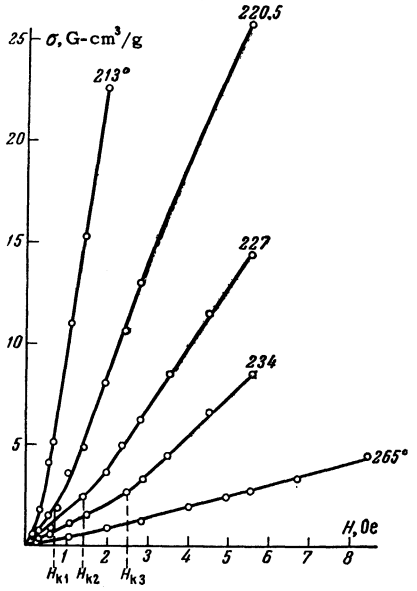

FIG. 3. The magnetization isotherms of gadolinium in the temperature interval Θ_1–Θ_2 in fields up to 8 Oe.

Θ_1 the $\sigma(H)$ curves are continuous as in typical ferromagnets. On increase of temperature, though still below Θ_1, reduction of the magnetic anisotropy constant causes an increase of the susceptibility (Fig. 4), which is again typical of ferromagnets. This should be compared with the decrease of the susceptibility with increase of temperature between Θ_1 and Θ_2.

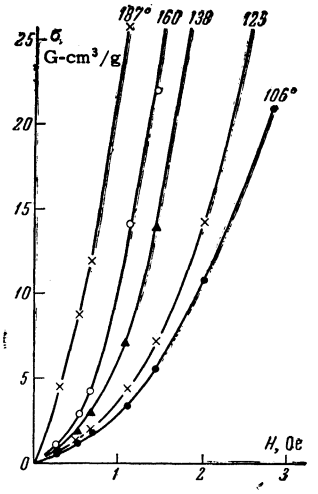

FIG. 4. The magnetization isotherm of gadolinium below Θ_1.

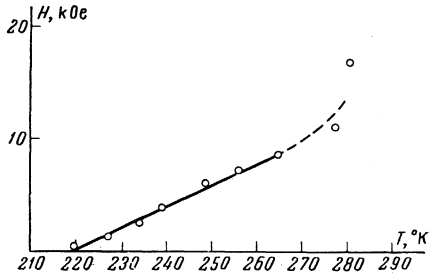

FIG. 5. The temperature dependence of the critical field H_c.

The curves of Fig. 2 indicate that the critical field H_c rises with temperature (Fig. 5), as in the case of dysprosium. In gadolinium the values of H_c (0—15 Oe) are three orders of magnitude smaller than in the case of dysprosium (0—11000 Oe).

3. The magnetic properties of gadolinium between Θ_1 and Θ_2 can be accounted for by assuming the helical spin configuration which is known to occur in the lattices of rare-earth metals. In each layer of atoms of the basal plane of a hexagonal gadolinium crystal, spins are oriented in parallel (ferromagnetism). The directions of the resultant magnetizations of neighboring planes made an angle α_0 and this indicates a helical spin configuration. Enz [4] showed that a nonzero angle between the magnetization directions of neighboring layers of atoms is the result of competition between two exchange interactions: positive between atoms of neighboring layers, and negative between atoms in the same layer. The angle α_0 is given by [4]

$$\cos\alpha_0 = -J_1/4J_2, \qquad (1)$$

where the J's are the exchange parameters: $J_1 > 0$ and $J_2 < 0$.

Internal magnetic fields acting in basal planes (effective fields of magnetic and magnetoelastic anisotropies) tend to reduce the angle α_0, i.e., they tend to destroy the helical configuration; this destruction does indeed take place at $T < \Theta_1$. An external magnetic field $H = H_c$ also tends to reduce the angle α_0 and destroy the helical spin configuration. With increase of temperature the magnetic anisotropy constants and the magnetostriction in the basal plane are reduced and therefore the corresponding effective internal fields are also reduced. Thus stronger external fields are needed to destroy the helical configuration, i.e., H_c should rise with increase of temperature as, indeed, is observed (Fig. 5). The very small values of H_c for gadolinium indicate that the angle α_0 of this metal is very small.

Equation (1) shows that a helical spin configuration with a very small angle α_0 appears when the negative exchange interaction (J_2) is small compared with the positive interaction ($J_2 \approx -J_1/4$).

[1] Behrendt, Legvold, and Spedding, Phys. Rev. **109**, 1544 (1958); Elliott, Legvold, and Spedding, Phys. Rev. **94**, 1143 (1954); Rhodes, Legvold, and Spedding, Phys. Rev. **109**, 1547 (1958); Thoburn, Legvold, and Spedding, Phys. Rev. **112**, 56 (1958); Elliott, Legvold, and Spedding, Phys. Rev. **100**, 1595 (1955).

[2] Wilkinson, Koehler, Wollan, and Cable, J. Appl. Phys. Suppl. to **32**, No. 3, 48S (1961).

[3] Belov, Levitin, Nikitin, and Ped'ko, JETP **40**, 1562 (1961), Soviet Phys. JETP **13**, 1096 (1961).

[4] U. Enz, J. Appl. Phys. Suppl. to **32**, No. 3, 22 (1961).

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14