

## SOME MAGNETIC PROPERTIES OF A GADOLINIUM SINGLE CRYSTAL

V. I. CHECHERNIKOV, IULIU POP, I. V. BUROV, and E. M. SAVITSKIĬ

Moscow State University

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The magnetic properties of a gadolinium single crystal have been investigated from 20 to 1000°C. It was established that the magnetic susceptibility measured in a direction perpendicular to the *c* axis is larger than in the parallel direction. Magnetization curves were measured in both directions near the magnetic transformation temperature (17°C). Existence of a critical field  $H_c = 3000$  Oe, at which the magnetization abruptly increases, was observed. The size of the magnetization jump decreases with increase of temperature and vanishes at  $T = 100^\circ\text{C}$ . The data obtained show the presence in gadolinium of a complicated spin configuration.

1. The magnetic properties of polycrystalline gadolinium in the high-temperature region have been studied in many researches.<sup>[1-3]</sup> These investigations made it possible to obtain some information about the electronic structure and magnetic properties of this metal. Meanwhile many questions, in particular the question of the existence of a helical spin configuration in gadolinium, remain still not definitely settled. It has recently been shown that in many rare-earth metals (Dy, Tb, Ho, Er, Tu), there are two critical temperatures ( $\Theta_1$  and  $\Theta_2$ ), in the interval between which these metals are in an antiferromagnetic state.<sup>[4-7]</sup> Investigations by neutron diffraction<sup>[8]</sup> have shown that between temperatures  $\Theta_1$  and  $\Theta_2$ , there is a helical spin structure, which constitutes a peculiar form of antiferromagnetism. Belov and coworkers also detected two phase-transition temperatures in polycrystalline gadolinium (cf. <sup>[9]</sup>).

It therefore seemed particularly interesting to make a study of a single crystal of gadolinium. An investigation of the magnetic properties was carried out in the temperature interval from 17 to 1000°C.

A single crystal of gadolinium was obtained by the method of recrystallization-annealing at temperature 1200°C over a period of 20 hours. The annealing was done in a vacuum at residual pressure  $2 \times 10^{-6}$  mm Hg.<sup>[10]</sup> The raw material for preparation of the single crystal was metallic gadolinium, prepared by reaction of gadolinium fluoride with calcium. After this the gadolinium was subjected to distillation in a vacuum. The distilled metal contained the following impurities: fluorine  $\leq 0.0065\%$ , oxygen  $\leq 0.07\%$ , nitrogen

$\leq 0.01\%$ , neodymium  $\leq 0.1\%$ , and samarium  $\leq 0.4\%$ .

Figure 1 shows the microstructure of the distilled gadolinium. As can be seen, the grains have thin boundaries, and both the body and boundaries of the grains are practically free of nonmetallic inclusions; these, as is known, impede grain growth. The annealing produced a coarse-grained specimen with grain sizes from 3 to 12 mm diameter. A single crystal was cut from the specimen and was oriented by the Laue method. After each cutting operation, the surface of the metal was etched with a solution of nitric acid and alcohol, to a depth of 0.1 to 0.2 mm, to eliminate surface hardening.

FIG. 1. Microstructure of gadolinium ( $\times 200$ ).

2. Metallic gadolinium, as is known, has a hexagonal close-packed lattice of the magnesium type, with the parameters  $a = 3.6360 \pm 9 \text{ \AA}$ ,  $c = 5.7826 \pm 6 \text{ \AA}$ ,  $c/a = 1.59$ .<sup>[11]</sup> The single-crystal gadolinium was placed in a holder in such fashion that

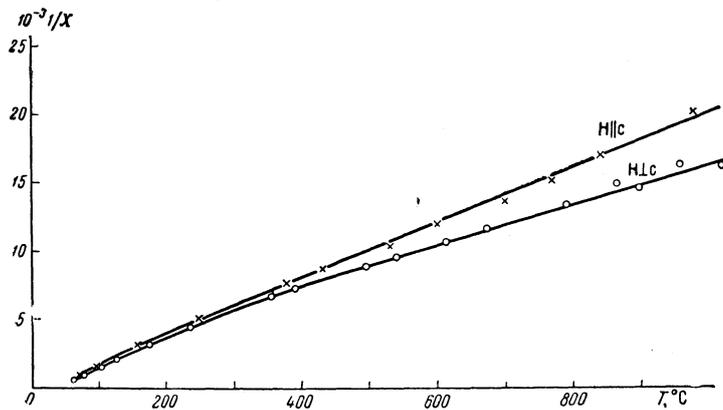


FIG. 2. Dependence of  $1/\chi$  on  $T$  for a single crystal of gadolinium.

the magnetic field was directed either parallel or perpendicular to the  $c$  axis, i.e.,  $H \parallel c$  or  $H \perp c$ . Measurements of the temperature dependence of the magnetic susceptibility were made in vacuum, by the Sucksmith method, in the temperature interval from 17 to 1000°C. In the immediate vicinity of the 17°C phase-transition point (with  $T > 17^\circ\text{C}$ ), magnetization curves (to about 7000 Oe) were determined.

Figure 2 shows the dependence of the reciprocal of the specific susceptibility,  $1/\chi$ , on temperature for two different orientations of the crystal. The magnetic susceptibility measured in the perpendicular direction was found to be larger than in the parallel. This difference was most pronounced in the high temperature region. Below 400°C the difference is less clearly evident. In the temperature interval from 17 to 100°C, we also measured isotherms of magnetization, which are shown in Fig. 3. The magnetic field was directed perpendicular and parallel to the  $c$  axis. On the magnetization isotherms, at  $H_C = 3000$  Oe, there occur breaks, above which the magnetization increases faster. Similar breaks had been observed earlier in other rare-earth metals, [4-7] in the range in which there is a helical spin structure. With in-

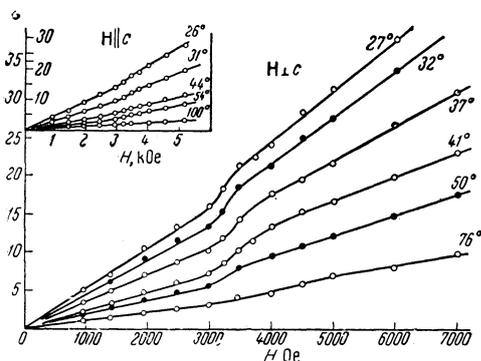


FIG. 3. Magnetization isotherms for a single crystal of gadolinium.

crease of temperature, the size of the magnetization jump in gadolinium decreases, and at about 100°C it completely disappears. At and above this temperature, the susceptibility is independent of the direction of the magnetic field. Thus the temperature  $\Theta = 100^\circ\text{C}$  is probably the temperature below which there exists a complicated electronic configuration, which breaks down in the presence of a critical field  $H_C$ . On the magnetization curves taken in a parallel field ( $H \parallel c$ ), similar breaks are observed, but they are less clearly marked (Fig. 3). It should be emphasized that the size of the critical field  $H_C$  in both cases remains constant, and equal to 3000 Oe, as the temperature increases.

3. The results of the investigation of the temperature dependence of magnetic susceptibility of a single crystal of gadolinium, in the high-temperature range, point to the existence in this rare-earth metal of a complicated electronic configuration. In all probability, the magnetic moments in gadolinium are oriented with respect to the  $c$  axis at some angle that is less than  $90^\circ$ . These moments are arranged antiparallel at definite periods; this leads to the occurrence of an antiferromagnetic phase. As experiment has shown, remnants of the antiferromagnetic configuration persist at temperatures above  $17^\circ\text{C}$  and vanish only at about  $100^\circ\text{C}$ .

The possibility is not ruled out that in gadolinium there is a significantly more complicated spin structure than in other rare-earth metals.

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<sup>1</sup>W. Klemm and H. Bommer, *Z. Anorg. Chem.* **231**, 138 (1937).

<sup>2</sup>S. Arajs and R. V. Colvin, *J. Appl. Phys.* **32**, 336S (1961).

<sup>3</sup>V. I. Chechernikov, *FMM* **13**, 3 (1962).

<sup>4</sup>Behrend, Legvold, and Spedding, Phys. Rev. **109**, 1544 (1958).

<sup>5</sup>Elliott, Legvold, and Spedding, Phys. Rev. **94** 1143 (1954).

<sup>6</sup>Elliott, Legvold, and Spedding, Phys. Rev. **100**, 1595 (1955).

<sup>7</sup>Rhodes, Legvold, and Spedding, Phys. Rev. **109**, 1547 (1958).

<sup>8</sup>Wilkinson, Koehler, Wollan, and Cable, J. Appl. Phys. **32**, 48S (1961); Cable, Wollan, Koehler, and

Wilkinson, J. Appl. Phys. **32**, 49S (1961).

<sup>9</sup>K. P. Belov and A. V. Ped'ko, JETP **42**, 87 (1962), Soviet Phys. JETP **15**, 62 (1962).

<sup>10</sup>Savitskii, Terekova, Naumkin, and Burov, Tsvetnye metally (Nonferrous Metals), 1963, p. 5.

<sup>11</sup>Spedding, Daane, and Herrmann, Acta Cryst. **9**, 559 (1956).

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