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Magnetocrystalline Anisotropy of Gd-Y-Sc Alloys

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Magnetocrystalline anisotropy measurements have been made on a single-crystal sample of $Gd_{75}Y_{17.5}Sc_{7.5}$ alloy over the temperature range from 77 to 240 K using a torque magnetometer. It is shown that at temperatures greater than T_c^1(=191K) the apparent easy direction of magnetization in the low field is *c*-axis. Below this temperature the easy direction lies on a basal plane. In the temperature range between T_c^1 and T_c (=219K), the anisotropy constant K_1 increase initially up to about 3kOe and then decrease with increasing field. This corresponds to the change in the apparent easy direction from *c*- to *a*-axis with increasing magnetic field, which in itself is unusual behaviour for ferromagnet.

§ 1. Introduction

Gadolinium is the only rare earth which is a simple ferromagnet.[1] The magnetic ordering is believed to take place by a long range oscillatory indirect exchange interaction of the RKKY-type between the 4*f* electrons via polarization of the conduction electrons.[2] The 4*f* shell electrons in Gd metal have essentially no orbital moments in the ground state, so that the Gd metal exhibits a relatively weak magnetocrystalline anisotropies[3] about 10^{-3} times smaller than other heavy rare earth metals. Recently, Cable and Koehler[4] have made measurements of the neutron diffraction for ^{160}Gd single-crystal and found that above 235 K long-wavelength spin-orientation fluctuations exist and only the average easy direction of magnetization is the *c*-axis. It is interesting to study the magnetic properties of the Gd alloyed with nonmagnetic elements such as Y and Sc in order to have the information of dilution effect of the long-wavelength spin-orientation fluctuation region.

The single crystals of Gd-Y[5),6)] and Gd-Y-Lu[7)] were investigated and a new magnetic ordering phenomenon was found. The low isofield magnetization data for the *a*-axis samples showed two different Curie-Weiss regimes whereas similar data for an *a*-axis Gd single crystal showed a single Curie-Weiss regime. The two Curie-Weiss regimes encountered are suggestive of double ferromagnetism which places the moments on the surface of a cone around the *c*-axis at T_c while the basal-plane component is random down to a lower temperature T_c^1 below which simple ferromagnetism obtains.

It seemed strange to us that Gd-Sc alloy did not show the same behavior as Gd-Y alloys because Sc has the same hcp crystal structure as Y with only a slightly smaller atomic volume than Y. This prompted us to see if adding some Y to Gd-Sc might yield magnetic behavior similar to Gd-Y. We report here the results of magnetocrystalline anisotropy measurements on Gd-Y-Sc single crystals.

§2. Sample Preparation

The single crystals used in this study were prepared in bulk form starting with the highest purity Ames Laboratory polycrystalline Gd, Y and Sc metals. The major impurities in the starting Gd, given in ppm atomic, were O=220, H=310, C=80, N=10, F=25 and Fe=26. All other metallic impurities were less than 10 ppm atomic. The yttrium and scandium were of compararable purity. The alloys were prepared by arc melting the mixture of the corresponding elements in a purified argon atomosphere. The single crystals were grown by a thermal annealing procedure described by Nigh.[8] For measurement of magnetocrystalline anisotropy, the single-crystal was shaped into the sphere of about 6.9 mm in diameter. The deviation from the true sphere was less than 0.5% in diameter. The orientation of the crystal was determined by X-ray Laue reflection method.

§3. Magnetocrystalline Anisotropy Results

The magnetocrystalline anisotropy was measured in the temperature range from 77 to 240 K by means of an automatic torque magnetometer which utilizes an unbonded strain gauge for the conversion of torque to electricity. The measurement was done by the application of fields up to 12 kOe in the *a*-*c* plane.

In Fig. 1 is schematically shown the automatic torque magnetometer. Here S is a sample. The torque of the sample is transferred to the U-gauge by a specimen holder made of quartz pipe C which is pulled up by phosphorus bronze wire P and a large weight W. The force acting on the specimen was detected in a range from 10 to 3×10^3 dyne.

The torque curves in the *a*-*c* plane for $Gd_{75}Y_{17.5}Sc_{7.5}$ alloy at 200 Oe are shown in Fig. 2. Below T_c^1 (=191 K), the apparent easy direction of magnetization, where the torque curve intersects the abscissa changing the sign from the positive to the negative value with increasing the rotation angle, is the *a*-axis, and the hard axis is the *c*-axis. Above T_c^1, the easy direction is the *c*-axis.

In Fig. 3 the easy direction referred to the *c*-axis, θ, which was found by torque measurements at 200 Oe is plotted as a function of temperature. The easy direction in the low field changes suddenly from the *a*-axis to *c*-axis at T_c^1 with increasing temperature.

Figure 4 shows the torque curves at 192 K, at which the magnetic structure has a

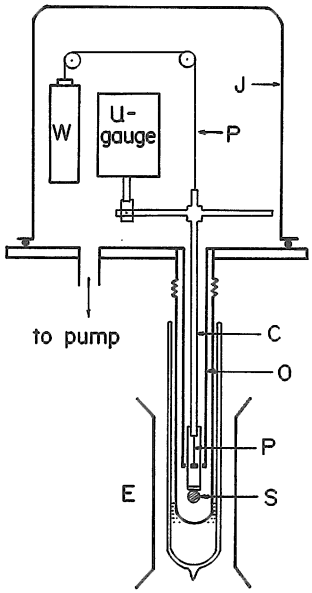

Fig. 1. Block diagram of a automatic torque magnetometer.

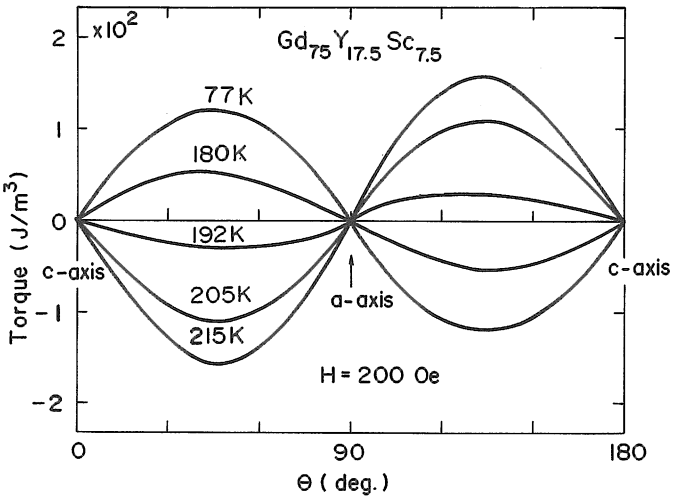

Fig. 2. Torque curves for $Gd_{75}Y_{17.5}Sc_{7.5}$ alloy at 200 Oe for various temperatures.

double ferromagnetic arrangement. In low fields, the apparent easy axis is the *c*-axis. In the fields from 6 to 8 kOe, the anisotropy energy surface, which is obtained from the integral of torque curve, have the two minimums of the stable and metastable states. At 6 kOe, the stable state is *c*-axis and the matastable state is *a*-axis. At 8 kOe, on the other hand, the stable state is *a*-axis and the metastable state is *c*-axis. The apparent easy direction of magnetization changes from the *c*- to *a*-axis with increasing magnetic

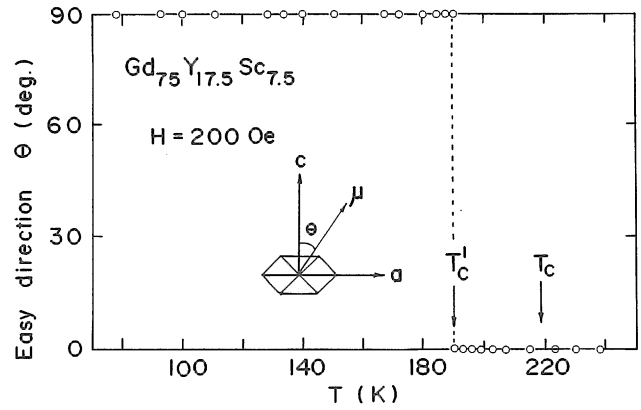

Fig. 3. Easy direction at 200 Oe vs temperature.

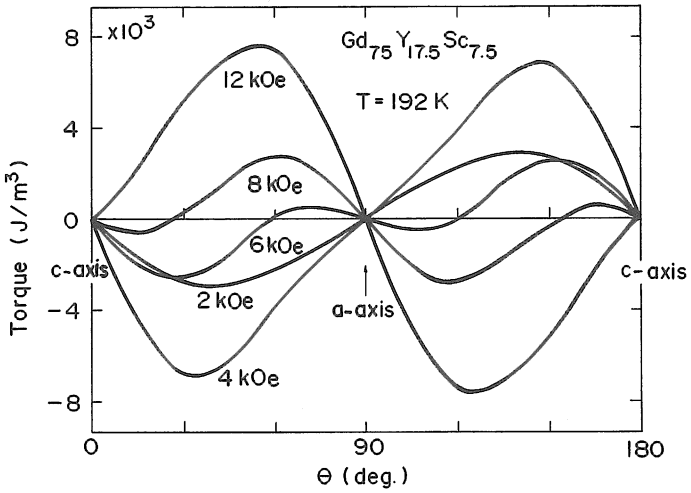

Fig. 4. Torque curves for $Gd_{75}Y_{17.5}Sc_{7.5}$ alloy at 129K for various applied field.

field. At the field of 12 kOe, the easy axis is the a-axis, and the torque curve is similar to that below T_c^1.

For a hexagonal crystal in the ferromagnetic state, the magnetocrystalline anisotropy energy is approximately written, according to the crystal symmetry, in the formula

$$E_a = K_0 + K_1 \sin^2\theta + K_2 \sin^4\theta + K_3 \sin^6\theta + K_4 \sin^6\theta \cos 6\phi + \cdots,$$

where K_1, K_2, K_3 and K_4 are the anisotropy constants, and θ and ϕ are the angles of the magnetization vector and its projection to the basal plane referred to the c- and a-axes, respectively. Assuming that ϕ is constant and the last term may be included in the previous term as $K_3' = K_3 + K_4 \cos 6\phi$, then the torque curve is given by the equation

$$L=-\frac{\partial E_a}{\partial\theta}$$

$$=-\left(K_1+K_2+\frac{15}{16}K'_3\right)\sin 2\theta+\left(\frac{K_2}{2}+\frac{3}{4}K'_3\right)\sin 4\theta-\frac{3}{16}K'_3\sin 6\theta+\cdots.$$

Using this equation, we determined the values of the anisotropy constants by Fourier analysis of the torque curves obtained in this measurement.

The values of the anisotropy constant K_1 vs applied field for various temperatures are shown in Fig. 5. Below T_c^1, K_1 decrease with increasing field which is expected for ferromagnetic anisotropy. In the temperature range from T_c^1 to T_c, K_1 increase initially and then decrease with increasing field, which in itself is unusual behaviour for ferromagnet. This corresponds to the change in the easy direction from the c- to a-axis with field at constant temperature. Above T_c, K_1 increase with increasing field which is expected for paramagnetic anisotropy.

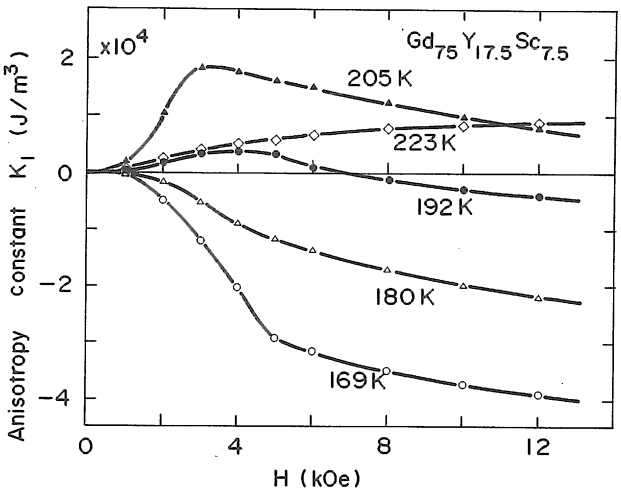

Fig. 5. Apparent anisotropy constant K_1 vs applied field for various temperatures.

§4. Discussion

The magnetic anisotropy of Gd is small compared to that of the other rare-earth metals. This is in accordance with the fact that the orbital moment of the half-filled $4f$ shell is zero. In first order there is no spin-orbit coupling for the localized $4f$ electrons. Part of the magnetic moment arises from polarization of the nonlocalized $5d$ electrons. These electrons are mainly responsible for the anisotropy of Gd and Gd-Y (Sc, Lu) alloy systems.

Corner *et al.*[3] have made measurements of the magnetocrystalline anisotropy of Gd and found the unusual behaviour of the field dependent anisotropy K_1 at temperatures close to the Curie temperature, at which the K_1, initially positive, decrease with

increasing field. They conclude that these unusual behaviour comes from paramagnetic alignment. They pointed out, however, that the effect of the temperature dependence of paramagnetic contribution to K_1 changes the sign at T_c as the temperature is increased. The paramagnetic contribution might not change the sign. It is difficult to conclude that the unusual field dependence of K_1 comes only from paramagnetic contribution below T_c.

Franse *et al.*[9] have made measurements of the pressure dependence of magnetic anisotropy for high purity Gd and suggested that the anisotropy energy consists of two parts; a regular part that contains only a few Fourier components of low degree and that can therefore be described with a few anisotropy constants, and an anomalous part that is connected with the movement through the Fermi level of a peak in the density-of-states curve.

The anisotropy constant K_1 for $Gd_{75}Y_{17.5}Sc_{7.5}$ decrease with increasing field below T_c^1, which is expected for a ferromagnetic anisotropy. Above the Curie temperature T_c, K_1 increase with increasing field which is expected for the paraprocess resulting from the alignment of atomic moments against thermal fluctuations by the applied field. In the temperature range from T_c^1 to T_c, K_1 increase initially and then decrease with increasing field. The magnetostriction constant along the c-axis for $Gd_{75}Y_{25}$ alloy is positive in value at all the temperature range and has a large maximum value near the Curie temperature.[10] The unusual field dependence K_1 in the double ferromagnetic region is probably caused by the magnetostriction which produce the movement through the Fermi level of a small peak in the density of electron states curve.

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