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Magnetic Properties of Gd-Sc Single Crystal Alloys

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Magnetization, magnetic susceptibility, electrical resistivity, thermal expansion, specific heat and magnetocrystalline anisotropy measurements have been made on $Gd_{75}Sc_{25}$ single crystals. Low isofield a-c susceptibility data for the *c*-axis sample exhibit the existence of antiferromagnetism, which is suppressed ferromagnetically by the applied field of 30 Oe. Electrical resistivity, thermal expansion and specific heat data show an anomaly at T_N. Magnetocrystalline anisotropy measurements show that the easy direction changes from the basal plane to the *c*-axis at 87 K with increasing temperature.

I. Introduction

A study of alloy systems of heavy rare-earth metals with a nonmagnetic constituent should provide fruitful information to aid in understanding the complex magnetic interactions in the rare-earth metals. The free-electron RKKY indirect-exchange model has been applied to the rare-earth metals with reasonable success. It reveals that nearest-neighbor interactions are ferromagnetic and next-nearest-neighbor interactions are antiferromagnetic. If we apply these criteria to Gd alloyed with nonmagnetic Sc, we would expect the nearest-neighbor interactions to decrease more than the next-nearest-neighbor interactions. Hence, the Curie temperature of this alloy system should decrease in accord with simple dilution theory and the magnetic ordering should became antiferromagnetic at sufficiently high concentrations of the non-magnetic component. Nigh *et al.*[1] showed that the alloys of higher Gd content were ferromagnets and these of lower Gd content were antiferromagnets. For the most part, experiments are in agreements with theoretical predictions. However, there were some differences in the detailed behavior of Gd–Y alloy systems.[2),3),4)] In the alloys of higher Gd content, the low isofield magnetization data for the *a*-axis showed two different Curie-Weiss regimes, which suggests double ferromagnetism. This paper deals with the similar detailed observations for Gd–Sc alloy system.

We report here the results of detailed measurements of magnetization, low field a–c susceptibility, electrical resistivity, thermal expansion, specific heats and magnetocrystalline anisotropy on $Gd_{75}Sc_{25}$ single crystals.

II. Experimental Procedure

The alloys were prepared by arc-melting together carefully weighed amounts of the constituents. The single crystals were grown by a thermal annealing described by Nigh.[5] The arc-melted button ingots were supported by a tantalum sheet and annealed for five days just below the melting point of the alloys in the quartz tube. For measurements of magnetization, the single-crystals were cut out rectangular parallelepipeds, $3 \times 3 \times 5$ mm, with the long dimensions oriented along the crystallographic axis to be placed in the direction of the magnetic field. For electrical resistivity and thermal expansion measurements the single-crystals were shaped into the rectangular parallelepipeds, about $1 \times 1 \times 15$ mm, with the long dimension oriented along the *a*- and *c*-axes. For measurement of magnetocrystalline anisotropy, the single crystal was shaped into the sphere of about 3 mm in diameter.

III. Results

The magnetization was measured in the temperature range from 77K to room temperature by means of a vibrating-sample magnetometer. In Figures 1and 2, we give the magnetization per gram,σ_g ,versus temperature at different applied fields for the *b*- and *c*-axes of $Gd_{75}Sc_{25}$, respectively. Isofields magnetization curves along the *b*-axis show knee at the ordering temperature, which are clearly shifted to lower temperatures by increasing fields up to 2 kOe.

Figures 3 and 4 show the low isofield a-c susceptibility normalized by the each value of T_N versus temperature at different a–c applied fields for the *b*- and *c*-axes of $Gd_{75}Sc_{25}$, respectively. We can see one ordering temperature at T_N for *b*-axis curves

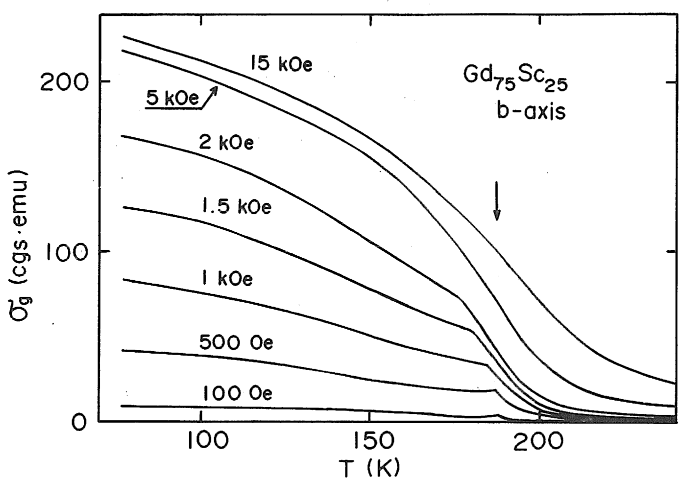

Fig. 1. Isofield magnetization vs temperature for $Gd_{75}Sc_{25}$ along the *b*-axis.

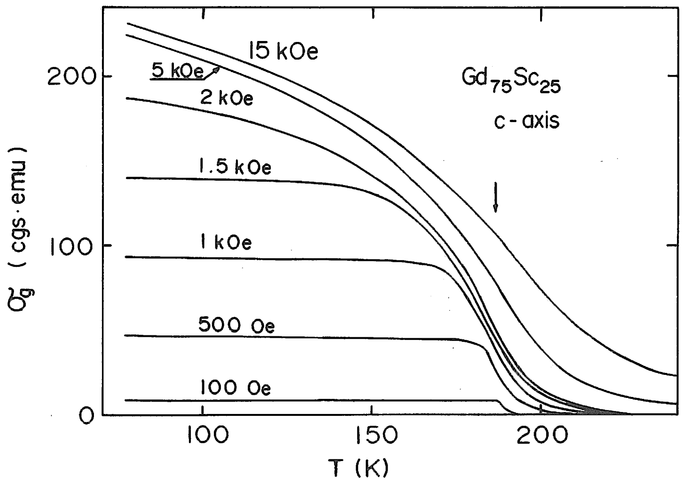

Fig. 2. Isofield magnetization vs temperature for $Gd_{75}Sc_{25}$ along the *c*-axis.

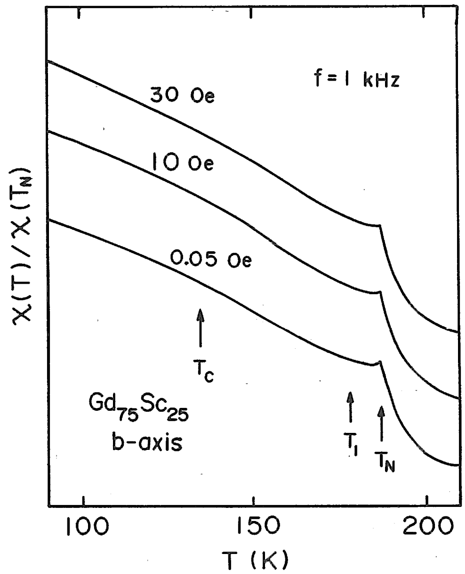

Fig. 3. Low isofied a-c susceptibility normalized by the each value of T_N vs temperature for $Gd_{75}Sc_{25}$ along the *b*-axis.

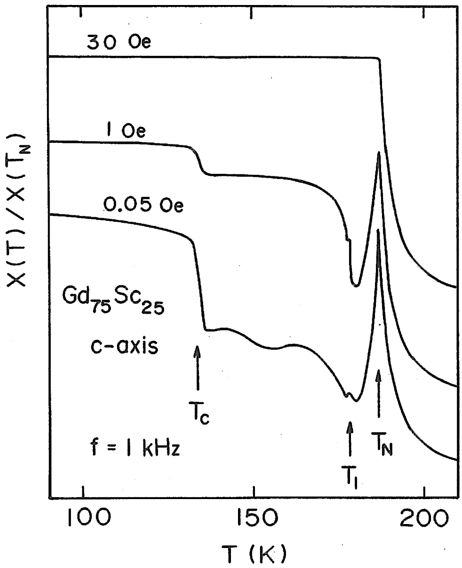

Fig. 4. Low isofield *a-c* susceptibility normalized by the each value of T_N vs temperature for $Gd_{75}Sc_{25}$ along the *c*-axis.

in Fig. 3. On the other hand, three ordering temperatures appear for the *c*-axis susceptibility curves up to 1 Oe in Fig. 4. Sharp peaks were found at the highest ordering temperature, T_N of 187 K. Small peaks appeared at T_I of 178 K up to 1 Oe. The *c*-axis susceptibility increase suddenly at T_C of 135 K as the temperature

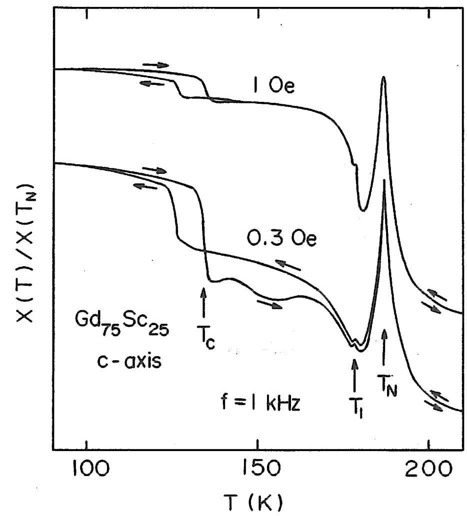

Fig. 5. Low isofield a-c susceptibility normalized by the each value of T_N for decreasing and increasing temperature.

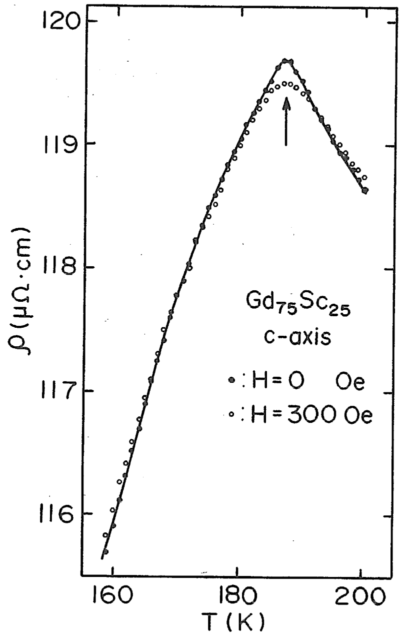

Fig. 6. Erectrical resistivity vs temperature for $Gd_{75}Sc_{25}$ along the *c*-axis. Open circles represent the magnetoresistance in the field at 300 Oe along the current direction.

decreasing.

Thermal hysteresis of isofield susceptibility curves for *c*-axis sample are shown in Fig. 5. The lowest transition temperature T_C obtained by the cooling and warming process are 126 K and 135 K, respectively. The susceptibility curve for 0.3 Oe in the warming process shows two small peaks between the transition temperatures of T_I and T_C.

The electrical resistivity for $Gd_{75}Sc_{25}$ single crytal was measured by the standard four-prove method. The enlarged electrical resistivity curve along the *c*-axis exhibit a peak at the highest ordering temperature T_N in Fig. 6. The effect of an external magnetic field in the *c*-axis direction on the electrical resistivity along the *c*-axis near T_N is shown. The external field supresses the resistivity.

Figure 7 shows the thermal expansion versus temperature with reference to that of 273 K along the *a*– and *c*-axes. The curves have the inflection point at the highest transition temperature T_N. Figure 8 shows the thermal expansion coefficients along the *a*- and *c*-axes as a function of temperature. The values of *a*-axis are positive and take a cusp at the magnetic transition temperature T_N, on the contraly the values of *c*-axis are negative and take a dip at T_N.

The low-temperature calolimeter used in this work is the usual isolation

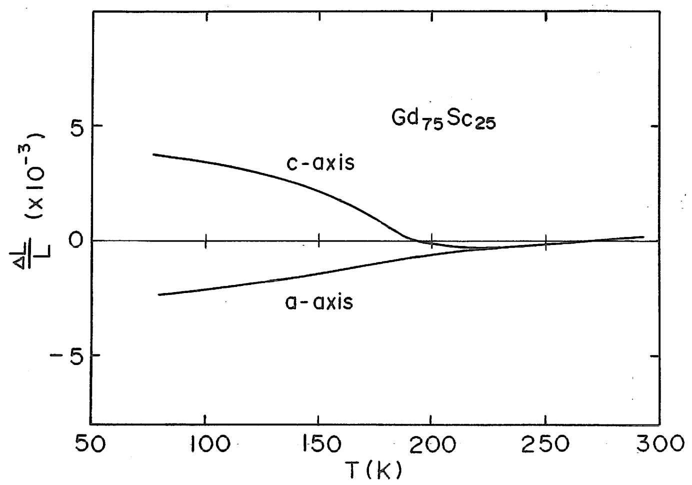

Fig. 7. The thermal expansion measured along the *a*- and *c*-axes as a function of temperature.

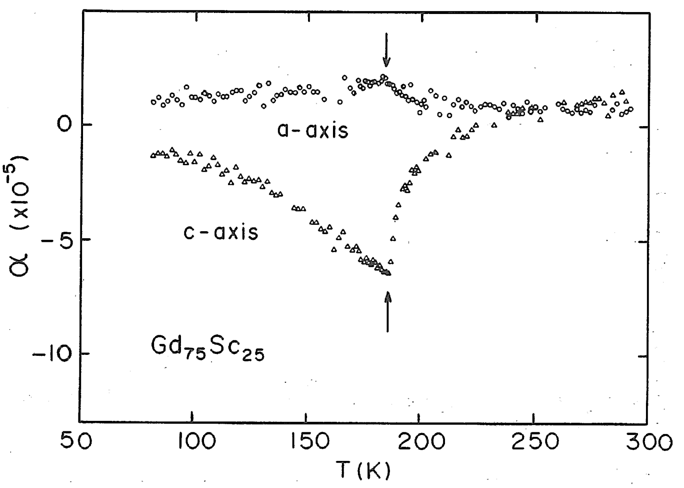

Fig. 8. The thermal expansion coefficients measured along the *a*- and *c*-axes as function of temperature.

heat-pulse type. The specific-heat results for $Gd_{75}Sc_{25}$ are shown in Fig. 9. Specific-heat peak was found close to the highest ordering temperature T_N. The specific-heat anomaly shows a λ-type second-order phase transition.

The magnetocrystalline anisotropy was measured in the temperature range from 77 K to room temperature by means of an automatic torque magnetometer. The measurement was done by the application of fields up to 14 kOe in the *a–c* plane. The values of the anisotropy constant K_1 and K_2 are plotted as a function of temperature

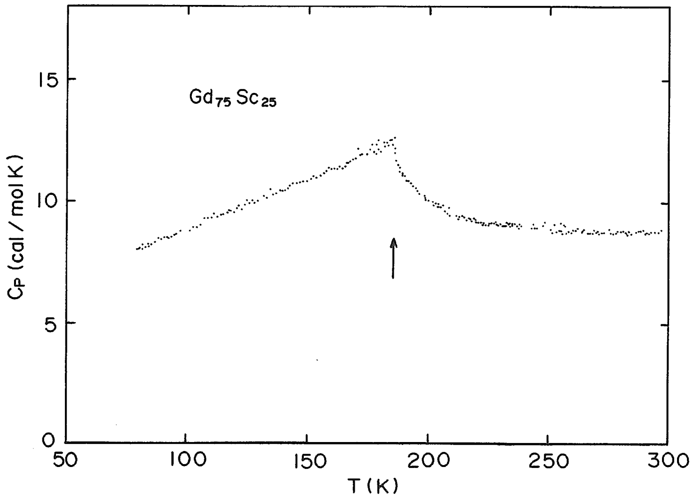

Fig. 9. Specific-heat vs temperature for $Gd_{75}Sc_{25}$ alloy.

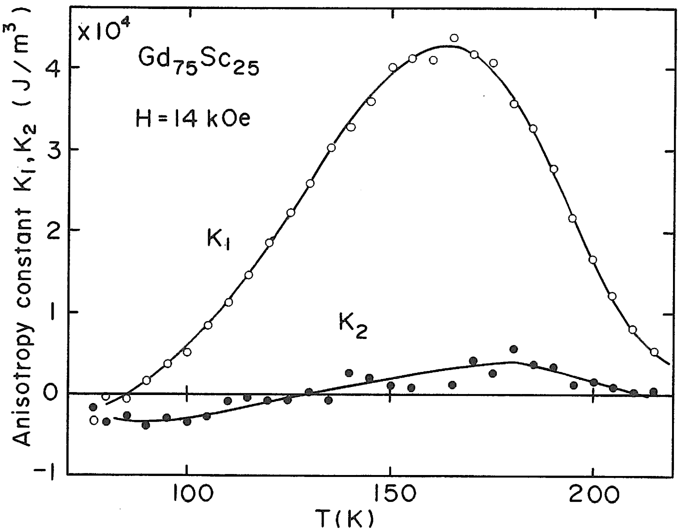

Fig. 10. A plot of anisotropy constants K_1 and K_2 vs temperature.

in Fig. 10. The values of K_1 take a maximum at about 160 K and change the sign at 87 K. The easy direction changes from the *c*-axis to the basal plane at 87 K with decreasing temperature.

IV. Discussion

Bates *et al.*[6] have made measurements of the neutron diffraction for the $Gd_{62.4}Y_{37.6}$ single crystal alloy and found a complex intermediate phase. It appears that the transition from antiferromagnetic to ferromagnetic as the temperature decreases proceeds via at least one intermediate phase. The phase implies a modulated *c*-axis moment and the coexistence of a cone structure, but whose structure was not yet fully understood.

Blaga *et al.*[7),8] have made detailed measurements of ultrasonic attenuation and electrical resistivity in Gd–Y alloy system. The anomalies at the Néel temperature are most unusual and indicate and interesting competition between ferromagnetic and antiferromagnetic phases. There is a possibility that Gd–Sc alloys 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. The results of low isofield a–c susceptibility for *c*-axis show the similar complex intermediate phase. The susceptibility curves of Gd–Sc alloys, however, do not show the same behavior as Gd–Y alloys which exhibited the double ferromagnetism.[2] The susceptibility curves exhibit the existence of antiferromagnetism, which is supressed ferromagnetically by the field of 30 Oe. The facts that applied fields of over 30 Oe are sufficient to suppress the phenomenon and that magnetization data on single crystals are needed to observe this phenomenon explains why it was overlooked in earlier investigation. The phase transitions at T_I and T_C could not observe in our measurements of electrical resistivity, thermal expansion and specific heat,because the energies of the transitions are too small and our experimental techniques were not so precise.

It will be helpful to have the results of neutron scattering experiments now underway.

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