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Drastic change of the electrical resistivity related to the novel magnetic phase transition in α-Sm_2S_3

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Abstract. Magnetization and electrical resistivity of α-Sm_2S_3 have been investigated by using single crystals. The temperature dependence of the magnetization; $M(T)$, demonstrates a magnetic transition at around T = 4.5 K. The $M(T)$ shows abrupt rise below 5K with decreasing temperature in the low magnetic filed of H = 100 Oe. Zero-field-cooled $M(T)$ shows a sudden decrease with marking a maximum at 4.5 K, while field-cooled $M(T)$ keeps steady rising around this temperature. The magnetization $M(H)$ in the magnetic field along the c-axis shows a ferromagnetic hysteresis at the lower temperature than T_C = 4.5 K, although its magnitude is much smaller than the value for the full Sm^{3+} moment. The resistivity $\rho(T)$ indicates a double peak having the maxima at T = 4.65 and 3.3 K in no magnetic field. The ratio of ρ(4.65 K) to ρ(6.0 K) is extremely large as it exceeds 100. The double peak diminishes rapidly with increasing magnetic field. The negative giant magneto-resistance effect having the magneto-resistance ratio of −99 % at H = 10 kOe has been observed at T = 4.5 K.

1. Introduction

Novel physical properties related to magnetic transitions in a series compounds α-R_2S_3 (R = Gd, Tb and Dy) have been investigated within recent years [1-9]. These compounds and α-Sm_2S_3 have the same crystal structure, shown in figure 1, having an orthorhombic symmetry that belongs to the space group *Pnma* [10]. There are two crystallographically inequivalent Sm sites labeled Sm1 and Sm2 in this structure. The Sm1 atoms form planes with buckling in the *ab*-plane. The Sm2 atoms are connected to these planes. The decorated planes are stacked along the *c*-axis. Magnetic and electrical measurements on α-Sm_2S_3 single crystal have been carried out. In this paper, we present novel magnetic transition and drastic change of the electrical resistivity of α-Sm_2S_3.

2. Experimental

The powder sample of α-Sm_2S_3 was prepared from Sm_2O_3 by a sulfurizing method [1, 11]. The single crystals of α-Sm_2S_3 were grown by a chemical transport reaction method with iodine as a carrier [1, 12]. The single crystal orientation was confirmed by X-ray (Cu K_α-radiation) diffraction measurements. Typical length of needle shaped single crystals having longitudinal directions along the *b*-axis is about a few millimeters and a typical diameter of hexagonal cross sections in the *ac* plane is a little less than one millimeter. The magnetization measurement was performed using an rf-SQUID magnetometer (Quantum Design). The magnetic field is applied along each direction of the crystal axes. The

electrical resistivity in a magnetic field was measured by a dc two-probe method. These measurements were made in the temperature range of 1.8-300 K and the magnetic field range of 0-10 kOe.

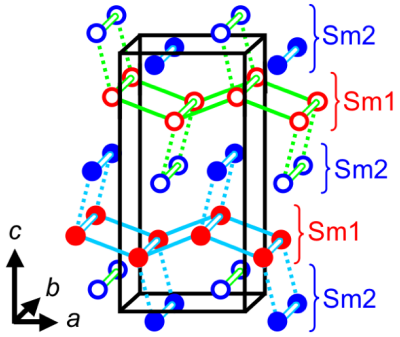

Figure 1. Crystal structure of α-Sm_2S_3, in which S atoms are omitted for clarity. Sm1 site atoms and Sm2 site atoms are stacked alternately along the *c*-axis. The open circles represent Sm atoms on (0, 1/4, 0) plane and the filled circles are Sm atoms on (0, 3/4, 0) plane.

3. Results and discussion

3.1. Magnetization

The magnetization of α-Sm_2S_3 at higher temperatures than 10 K is almost constant with having small values. Although weak temperature dependence of magnetic susceptibility at about T = 100-300 K is often seen in paramagnetic compounds including Sm^{3+} [13-15], the feature in the present case is more marked and seen in wider temperature region. There is a possibility of +2 valence for one Sm site.

The temperature dependence of the magnetization per one Sm atom below 10 K is shown in figure 2, where the magnetic field of 100 Oe is applied along the *c*-axis. The magnetization was measured in the processes of zero field cooling (Z.F.C.) and field cooling (F.C.). The Z.F.C. magnetization shows a sharp peak at 4.5 K (see the inset of figure 2), while the F.C. magnetization keeps rising around 4.5 K with decreasing temperature. After showing a shoulder at 3.7 K, the F.C. magnetization increases again and tends to be saturated at lower temperatures. The value of M at 1.8 K is 0.055 μ_B, which is as small as 7.7 % of full saturation moment 0.71 μ_B for Sm^{3+}. The $M(T)$ curve measured in the magnetic field of 100 Oe applied along the *a*- or *b*- axis, resembles $M(T)$ for $\boldsymbol{H}//\boldsymbol{c}$ qualitatively; however, the magnitude for $\boldsymbol{H}//\boldsymbol{a}$ (or $\boldsymbol{b}$) is more than an order smaller than that for $\boldsymbol{H}//\boldsymbol{c}$.

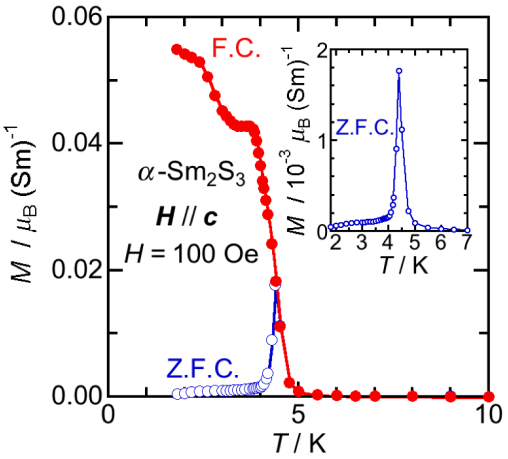

Figure 2. The temperature dependence of magnetization for α-Sm_2S_3 in the field of 100 Oe along the *c*-axis. The Z.F.C. and F.C. curves are shown. The Z.F.C. curve magnified is also shown in the inset.

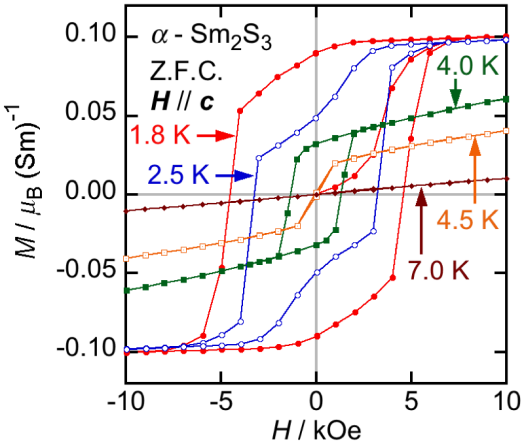

Figure 3. Magnetization curves at various temperatures for α-Sm_2S_3 within the field of 10 kOe along the *c*-axis. Initial magnetization process from H = 0 kOe is also shown only for the data obtained at 1.8 K.

Next, we show the magnetization as a function of magnetic field. Figure 3 shows the magnetization curves $M(H)$ at various temperatures. The $M(H)$ curve obtained at 7.0 K is linear to the magnetic field, while the $M(H)$ at 4.5 K shows a very small hysteresis around $H = 0$ kOe. The $M(H)$ curves at the lower temperatures than 4.5 K show ferromagnetic hystereses clearly. In the $M(H)$ curve at 1.8 K, the gentle slope of the initial process from $H = 0$ kOe becomes steeper around $H = 3.5$ kOe and then the curve tends to be saturated. The saturated value at $H = 10$ kOe is 0.10 μ_B. If we assume this value originates from only one Sm site, it corresponds to 0.20 μ_B per Sm atom on the Sm site considered. The value of 0.20 μ_B is about 28 % of full saturation moment. The magnitude of the residual magnetization is as large as 90 % of saturated value, and the coercivity is about 4.5 kOe. These hystereses demonstrate the existence of ferromagnetic phase below 4.5 K with having magnetic domains. We consider a weak ferromagnetic (WF) transition is brought about at 4.5 K by ordering the *c*-axis components of canted magnetic moments on one Sm site. When the sample is cooled down with no magnetic field, magnetic domains must be formed; thus, the magnitude of the Z.F.C. magnetization below 4.5 K is rather small as shown in figure 2. When the sample is cooled in a magnetic field; however, even low magnetic field of 100 Oe prevents the magnetic system from forming magnetic domains. Each of the series compounds α-R_2S_3 (R = Gd, Tb and Dy) shows antiferromagnetic (AF) ordering at $T = 9.9$ K [1, 2], 12.6 K [5] and 11.5 K [7]. It is considered that the AF ordering occurs in $R1$ site. That is confirmed by neutron diffraction experiments for α-Gd_2S_3 [3] and α-Tb_2S_3 [6]. Each $R2$ site moments show characteristic ordering at the different or same temperatures [1, 5, 7]. Assuming the preference of Sm1 site moments for AF ordering also in α-Sm_2S_3, we speculate the WF ordering occurs in Sm2 site at 4.5 K and AF ordering occurs in Sm1 site at 3.7 K. We premise that the Sm1 site has magnetic moments; namely, the valence of Sm is +3 at the low temperature region around 5 K.

3.2. Electrical resistivity

The temperature dependence of the electrical resistivity; ρ, of α-Sm_2S_3 in high temperature region is rather weak and the signs of $d\rho/dT$ are opposite at both sides of 240 K ($d\rho/dT > 0$ at $T > 240$ K). Such behavior is commonly observed in the series compounds α-R_2S_3 (R = Gd, Tb and Dy) [1, 5, 8].

Figure 4 shows temperature dependence of the electrical resistivity at lower temperatures than 10 K in various magnetic fields perpendicular to the *b*-axis. The dc electric current along the *b*-axis is 1.0 μA. The resistivity $\rho(T)$ indicates a double peak having the maxima at $T = 4.65$ and 3.3 K in no magnetic field. First peak (at the higher temperature) is extremely large; thus, the ratio of the maximum ρ(4.65 K) to ρ(6.0 K) exceeds 100. As for the second peak, the ratio of ρ(3.3 K)/ρ(6.0 K) is about 20. The double peak is reduced and shifted toward higher temperature by increasing the applied field. The applied field of 10 kOe diminishes the peaks significantly. However, the first peak remains with having the ratio of ρ(5.2 K)/ρ(6.0 K) = 9 (see the inset of figure 4).

Next, we show the change of the electrical resistivity against the magnetic field at 4.5 K in figure 5. The magneto-resistance (MR) ratio defined by $[\rho(H)-\rho(0)]/\rho(0)$ is plotted as a function of H. Negative giant magneto-resistance (GMR) has been found. We can see, for examples, the fairly large MR ratio about −20 % at $H = 100$ Oe and the extremely large one of −99 % at $H = 10$ kOe in figure 5.

Similar anomalous behavior in the electrical resistivity is seen also in α-Dy_2S_3 [8]. However, the present case is more drastic in the magnitude of the change. As temperature decreases, the $\rho(T)$ starts rising just above WF transition temperature in both the cases. Short-range ordering of R moments and change of magnetic structure must affect carriers in its scattering. Important difference is that the ferromagnetic hysteresis is not seen in the $M(H)$ of α-Dy_2S_3. It is considered that the Sm2 layers (WF) and Sm1 layers (paramagnetic or non-magnetic or AF) are stacked alternately along the *c*-direction in α-Sm_2S_3. We speculate several layers form a magnetic domain having the nano-sized thickness. As for α-Sm_2S_3, the formation and annihilation of the magnetic domains might have a great influence on the drastic change of the electrical resistivity.

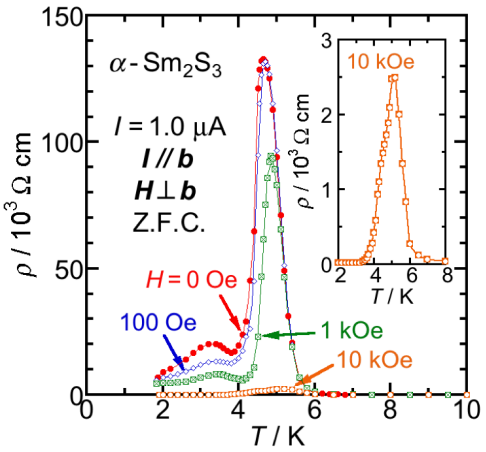

Figure 4. The temperature dependence of the electrical resistivity of α-Sm_2S_3 in the various magnetic field perpendicular to the *b*-axis. The inset shows the magnified curve for H = 10 kOe.

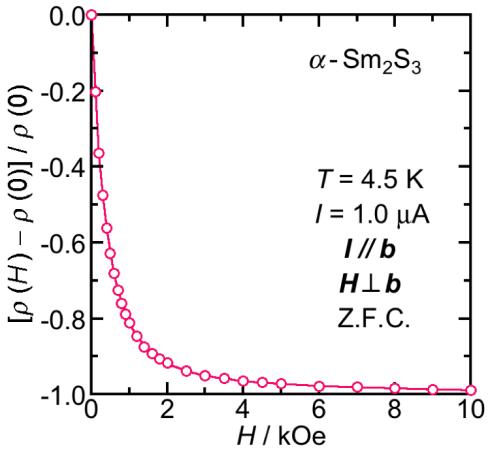

Figure 5. The magnetic field dependence of the magneto-resistance ratio of α-Sm_2S_3 at 4.5 K.

4. Conclusions

We have found a characteristic weak ferromagnetic transition at T_C = 4.5 K and related drastic changes of electrical resistivity induced by temperature and magnetic field in α-Sm_2S_3. Negative GMR effect having extremely large MR ratio of −99 % at H = 10 kOe has been observed at T = 4.5 K. It is considered that the change of magnetic structure including the formation and annihilation of magnetic domains influences the drastic change of the electrical resistivity significantly.

Acknowledgements

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