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Magnetic and magnetotransport properties of intermetallic SmMn_2Si_2

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The magnetic properties of naturally layered intermetallic compound SmMn_2Si_2 with textured structure have been studied. There exist a ferromagnetic transition at 35 K and two antiferromagnetic transitions at 120 and 230 K. The antiferromagnetic state below 230 K exhibits different magnetoresistance, with a negative magnetoresistance of 3%–4% for current I applied perpendicular to the c axis and with a positive magnetoresistance effect of about 4%–6% for current I parallel to the c axis. The observed magnetoresistance is likely to be related to magnetovolume effects. In the ferromagnetic state, a positive magnetoresistance with a maximum increase of 22% under an applied field of 5 T is observed at 4 K, and both $H \perp I$ and $H \parallel I$ configurations show positive magnetoresistance. © 1997 American Institute of Physics. [S0021-8979(97)73808-8]

I. INTRODUCTION

Naturally layered intermetallic ternary compounds, RMn_2X_2 (R =rare earth and X =Si, Ge), have become of renewed interest recently because of the large magnetoresistance (MR) effect^{1–3} which quite resembles that of the artificially layered magnetic thin film.^{2,4} The crystal structure of the compounds is of body-centered tetragonal ThCr_2Si_2 type. The structure is formed by the stacking of atomic layers perpendicular to the c axis with the sequence $-\text{Mn}_2\text{-X-R-X-}$. In this system, much attention has been paid to SmMn_2Ge_2 because it has been shown to undergo several magnetic phase transitions at high and low temperatures^{1–3,5,6} and exhibits interesting magnetotransport behavior.^{1–3} For the Si containing system, little work has been done on SmMn_2Si_2 (Ref. 6) or Si-substituted $\text{SmMn}_2(\text{Ge}_{1-x}\text{Si}_x)_2$.⁷ The study by Szytula and Szott⁶ indicated that SmMn_2Si_2 is an antiferromagnet with $T_N=398$ K. However, the low-temperature magnetic properties were not reported. The Si-substituted $\text{SmMn}_2(\text{Ge}_{1-x}\text{Si}_x)_2$ was studied over the composition range $x=0$ –0.15 only.⁷ Thus the low-temperature magnetic and magnetotransport behaviors of SmMn_2Si_2 have not been well studied. In this study, SmMn_2Si_2 has been prepared and its low-temperature magnetic phase transitions and magnetoresistive effects have been investigated.

II. EXPERIMENTAL DETAILS

SmMn_2Si_2 samples were prepared by arc melting of stoichiometric mixture of starting materials (purity >99.9%) in an Ar atmosphere. The melting procedure was first to form Mn–Si alloy and then to form the final products by adding Sm metal. The samples were subsequently sealed into evacuated quartz tubes and annealed at 800 °C for a week. The resulting samples were of textured structure in which the c axis is easy to identify based on the layered structure. X-ray diffraction analysis indicated that the samples were single phase with body-centered tetragonal ThCr_2Si_2 structure of lattice parameters $a=0.3990$ and $c=1.0509$ nm. Composi-

tion analysis by x-ray photoemission spectroscopy (XPS) also verified that the stoichiometry of the samples was in 1:2:2.

Magnetization measurements as a function of temperature between 4 and 300 K, and as a function of applied field up to 5 T, were carried out by means of a SQUID magnetometer (Quantum Design). Measurements of the resistance without or with an applied field up to 5 T were taken by the standard four-lead method.

III. RESULTS AND DISCUSSION

The magnetic properties of RMn_2X_2 are sensitive to Mn–Mn intralayer distance with a critical value 0.2865 nm. The compounds with the distance less than 0.2865 nm are antiferromagnetic while those are ferromagnetic for the distance larger than the critical value.⁷ The Mn–Mn intralayer distance in SmMn_2Ge_2 is 0.2873 nm, larger than the critical value. It is ferromagnetic with a Curie temperature of 340 K.⁶ As it is cooled below 340 K, the ferromagnetic phase persists down to 150 K, then it orders antiferromagnetically, which is retained down to 100 K. As the sample is further cooled to below 100 K, a ferromagnetic phase reenters.¹ For SmMn_2Si_2 , its Mn–Mn intralayer distance is 0.2821 nm, less than the critical value of 0.2865 nm. This compound is antiferromagnetic with a Néel temperature of 398 K.⁶ However, the low-temperature magnetic properties of this compound have not been well studied. Figure 1 gives the first detailed view of the magnetic properties of SmMn_2Si_2 at low temperatures.

The temperature dependence of the magnetic susceptibility of the sample SmMn_2Si_2 in an applied field of 100 G is shown in Fig. 1. The sample was cooled down from room temperature in an applied field before magnetization measurement. As seen from Fig. 1, the compound undergoes several magnetic phase transitions at low temperatures. These transitions occur at about 35, 120, and 230 K, which correspond to a ferromagnetic phase transition at ~ 35 K, and two antiferromagnetic phase transitions at ~ 120 and ~ 230 K, respectively.

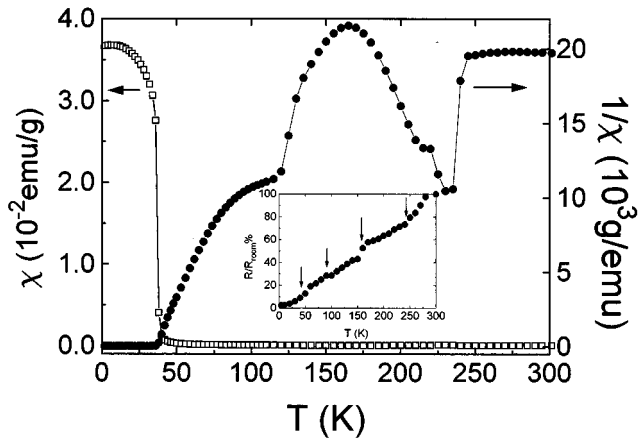


FIG. 1. Temperature dependence of the magnetic susceptibility and reciprocal susceptibility for SmMn_2Si_2 . The inset shows the temperature-dependent resistance at zero field. The arrows mark the temperature where anomalies in the resistance were observed.

It is known that at the transition point of antiferromagnetic state and the ferromagnetic state (see Fig. 1), a corresponding anomaly in the resistance is observed.¹⁻³ The temperature dependence of the resistance of SmMn_2Si_2 , measured when cooling from room temperature to 4 K, is shown in the inset of Fig. 1, which shows corresponding transitions at those transition temperatures indicated by arrows, ~ 35 , ~ 100 , and ~ 230 K. In addition to the transitions at about 35, 100 (120 K), and 230 K, an anomaly of the resistance at about 165 K was observed with a drop of the resistance. It is unclear what caused this anomaly at 165 K. From the magnetic susceptibility data shown, it does not appear that the anomaly is of magnetic origin.

Figure 2 shows the magnetic field dependence of magnetization at temperatures 4.2, 220, and 300 K. At 4.2 K, the magnetization increases sharply up to about 0.5 T, and then increases linearly up to the maximum applied field of 5.0 T without saturation. For the magnetization at 220 K, a linear relationship between the magnetization and the applied field is found. The magnetization at 300 K also changes linearly with applied field, both consistent with antiferromagnetic states.

The magnetoresistance measurements were performed at 220 and 4 K as a function of applied field up to 5 T. Because the samples are of textured structure, two configurations $H \parallel c$ and $H \perp c$ were employed when the applied current was perpendicular to the c axis. Figure 3 shows the field dependence of the resistance at 220 K, plotted as $(R - R_0)/R_0\%$ versus applied field H . At this temperature, the sample is in the antiferromagnetic state. A negative magnetoresistance was observed for $H \parallel c$ with a value of -3% and for $H \perp c$ with a value of -4.2% in the field up to 5 T applied for the first time. Thereafter, the MR value changes within a small range between -4.2% and -5.4% with field cycling between -5 and 5 T and does not come back to 0 at $H=0$ (not shown here). The inset of Fig. 3 shows the field dependence of the resistance at 220 K, measured with a configuration of $H \perp c$ when applied current was parallel to the c axis. A positive magnetoresistance of about 6% at 5 T was observed. Resem-

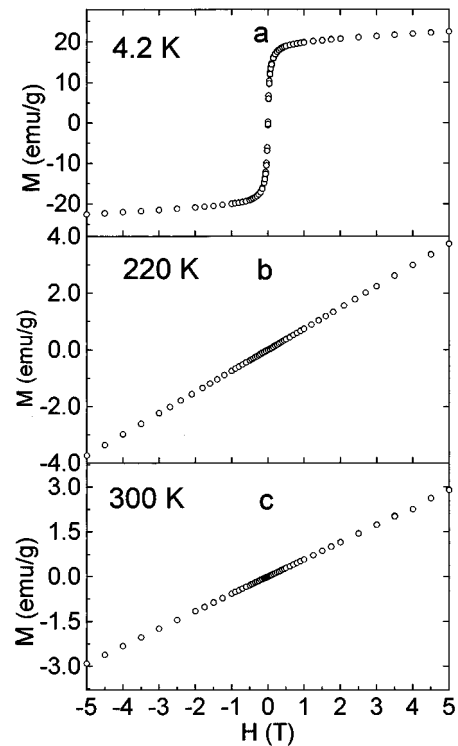


FIG. 2. Magnetization of SmMn_2Si_2 as a function of applied field at 4.2 K (a), 220 K (b), and 300 K (c).

bling the negative MR behavior, the positive MR value shifts between 4% and 6% with further field cycling and does not go back to 0 at $H=0$. This behavior that the MR changes monotonically initially with the applied field and does not go back to zero at $H_0=0$ upon further cycling was also observed in other layered systems, for example, AgSn/Co multilayer film.⁸ While such behavior in the multilayer systems, for example in AgSn/Co, is understood in that Co has a remanent moment, the cause for such behavior in SmMn_2Si_2 cannot be explained. As shown in Fig. 2(b), no observable remanence can be seen in its magnetization. Further investigation is needed in order to understand the mechanism

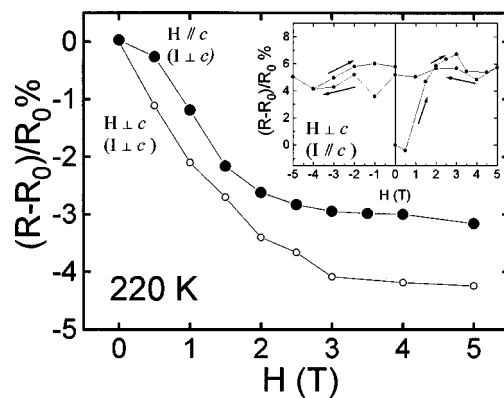


FIG. 3. Field dependence of the resistance of textured SmMn_2Si_2 at 220 K. Applied current was perpendicular to the c axis. The inset shows the field-dependent resistance where applied current was parallel to the c axis.

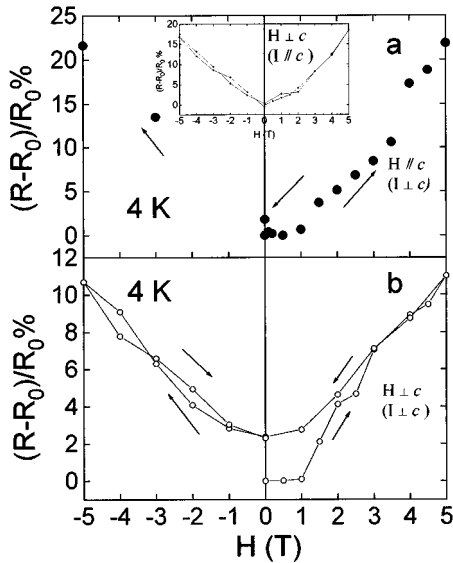


FIG. 4. Resistance as a function of applied field for textured SmMn_2Si_2 at 4 K. Applied current was perpendicular to the c axis. The inset shows the field-dependent resistance where applied current was parallel to the c axis.

for such hysteretic behavior in the magnetoresistance of SmMn_2Si_2 .

In the antiferromagnetic state, the resistance is higher than in the ferromagnetic state and a negative magnetoresistance effect has been observed in the polycrystalline SmMn_2Ge_2 related compounds,^{1,3} which can be understood on the basis of a spin-valve mechanism.⁴ However, in Fig. 3, both positive and negative magnetoresistance effects are observed when the current is applied along different directions (perpendicular or parallel to the c axis of the textured sample). Therefore the above mechanism cannot explain the observed results here. Similar results have been found by van Dover *et al.* who revealed that the antiferromagnetic state has a lower resistance than the ferromagnetic state when the current is along the a direction of a single crystal SmMn_2Ge_2 (Ref. 2) and it has a higher resistance when the current is along the c direction.⁹ A mechanism related to band-structure effects has been used to explain the observed results.^{2,3}

The sample enters the ferromagnetic state when cooled below 35 K. Figure 4 shows the resistance-field dependence of the sample at 4 K. With $H \parallel c$, an increase in MR of about 22% is observed at 5 T [Fig. 4(a)]. For $H \perp c$ [Fig. 4(b)], an increase in MR of 11% at applied field up to 5 T is found. Thereafter, the MR cycles reproducibly with field, with a subsequent minimum 2.3% at $H=0$. The inset of Fig. 4(a) is the field dependence of the resistance at 4 K, measured with

the same configuration as that of the inset of Fig. 3. A positive MR with an increase of 18% at 5 T is found. The MR cycles reproducibly with field and does not saturate with applied field up to 5 T. The MR observed in the ferromagnetic state is believed to be the anisotropic magnetoresistance.¹⁰

IV. CONCLUSIONS

Magnetization measurements between 4 and 300 K for the compound SmMn_2Si_2 indicated a ferromagnetic transition at 35 K, two antiferromagnetic transitions at 120 and 230 K, consistent with the resistance-temperature dependence. MR measurements in the antiferromagnetic state of 220 K revealed different magnetoresistive behaviors for different directions of the applied current with respect to the c axis of the textured samples. For $I \perp c$, a negative MR effect was observed with a decrease of about $-3\% \sim -4\%$ in the resistance at an applied field of 5 T. For $I \parallel c$, a positive MR effect was found with an increase of about $4\% \sim 6\%$ in the resistance. These MR effects cannot be explained in terms of a spin-valve mechanism.⁴ The observed MR is likely to be related to band-structure effects,² probably related to magnetovolume effects as proposed recently.³ Further research using a single crystal of SmMn_2Si_2 is needed in order to obtain data for comparison with those of a SmMn_2Ge_2 single crystal. For MR in the ferromagnetic state of 4 K, a positive MR effect was always observed, with a maximum increase of 22% at 5 T. This MR behavior is similar to that of the anisotropic magnetoresistance found in ordinary ferromagnets.

ACKNOWLEDGMENTS

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