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Effect of Cr substitution on the magnetic properties of $SmMn_2Ge_2$

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Abstract

The magnetic properties of polycrystalline $SmMn_2Ge_2$ and $Sm(Mn_{1-x}Cr_x)_2Ge_2$ have been investigated using magnetization, electrical resistance and thermal expansion measurements. A magnetic phase diagram for $Sm(MN_{1-x}Cr_x)_2Ge_2$ has been established. It is found that the antiferromagnetic phase in $SmMn_2Ge_2$ disappears for Cr concentrations of less than 5 at.% when Mn is partially substituted by cr. It is found that there are two ferromagnetic (FM) phases in $Sm(Mn_{a-x}Cr_x)_2Ge_2$. The high temperature FMI phase is due to Mn moment ordering while the lower temperature FMII phase is due to Mn as well as Sm moment ordering.

Keywords: Re-entrant ferromagnetism; Magnetic phase diagrams; Thermal expansion

1. Introduction

Intermetallic ternary compounds with the formula RMn_2Ge_2 (where R is a rare earth element) crystallize in body-centered tetragonal structures. Over the last few years several research groups have done studies to determine the magnetic properties of these materials. A detailed review of the properties of these compounds can be found in Refs. [1,2]. The magnetic properties of these compounds are sensitive to the intralayer Mn–Mn distances. The compounds with intralayer Mn–Mn distance $R^a_{Mn-Mn} < 0.2865$ nm are antiferromagnetic while compounds with $R^a_{Mn-Mn} > 0.2865$ nm are ferromagnetic.

In particular $SmMn_2Ge_2$ exhibits re-entrant ferromagnetism. It is found that $SmMn_2Ge_2$ is ferromagnetic below $T_c = 350$ K and goes into an antiferromagnetic phase on cooling below $T_2 = 150$ K. Further decrease in temperature leads to a ferromagnetic phase transition below $T_1 = 100$ K. In an effort to understand the nature of the magnetic phases in $SmMn_2Ge_2$ Duraj et al. [3,4] have studied the magnetic properties of $SmMn_2Ge_2$ by partial substitution of Sm with non-magnetic Y and magnetic Gd. They found that by increasing the Y or Gd concentration in $Sm_{1-x}L_xMn_2Ge_2$ (where L = Y or Gd) the temperature range of the ferromagnetic phase decreases while the antiferromagnetic range increases. This effect has been related to a decrease in intralayer distance R^a_{Mn-Mn} with increasing concentration of Y or Gd.

In this article we report the effect of partial substitution of Mn by cr in $Sm(Mn_{1-x}Cr_x)_2Ge_2$. Magnetization measurements as function of temperature and magnetic field are utilized to determine the magnetic phase diagram of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ system with chromium concentrations from $x = 0$ to $x = 0.50$.

2. Experimental detail

Polycrystalline samples were synthesized by arc melting of proper amounts of the constituent elements under an Ar atmosphere. An excess amount of Mn was added to each sample to compensate for loss (due to low vapor pressure) during arc melting. The purities of the elements were 99.9% for Sm, 99.99% for Mn and Cr and 99.999% for Ge. The ingots were remelted 4 or 5 times by turning over each time to ensure homogeneity. Samples were then wrapped in Ta foil and annealed under vacuum (not more than 10^{-7} Torr) at 800 °C for 7 days. Magnetization measurements were carried out by means of a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, CA) over the temperature range 5–395 K and in the field range 0.5–55 kG.

Resistivity measurements were made using the four-probe method over the temperature range 5–395 K. To carry out the experiments, the SQUID was used to control the temperature together with two external devices, a Keithley 220 current source and a Keithley 181 nanovoltmeter. Thermal expansion measurements were made by using a capacitance dilatometer over the temperature range 77–290 K.

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3. Results and discussion

3.1. $SmMn_2Ge_2$

The magnetization M of a zero field cooled sample of a polycrystalline $SmMn_2Ge_2$ as a function of temperature in applied field of 500 G is shown in Fig. 1(a). We observed (i) a ferromagnetic transition at $T_c = 341$ K, (ii) a ferro- to antiferromagnetic transition at $T_2 = 158$ K and (iii) an antiferro- to a ferromagnetic transition below $T_1 = 104$ K. These transition temperatures are consistent with earlier measurements of several researchers [5–8]. The phase transitions at T_1 and T_2 for $SmMn_2Ge$ are also observed in electrical resistivity (Fig. 1(c)) and thermal expansivity $\Delta l/l$ measurements (Fig. 1(d)). A step change in the thermal expansivity at $T_1 = 104$ K and again at $T_2 = 153$ K for $kSmMn_2Ge_2$ clearly

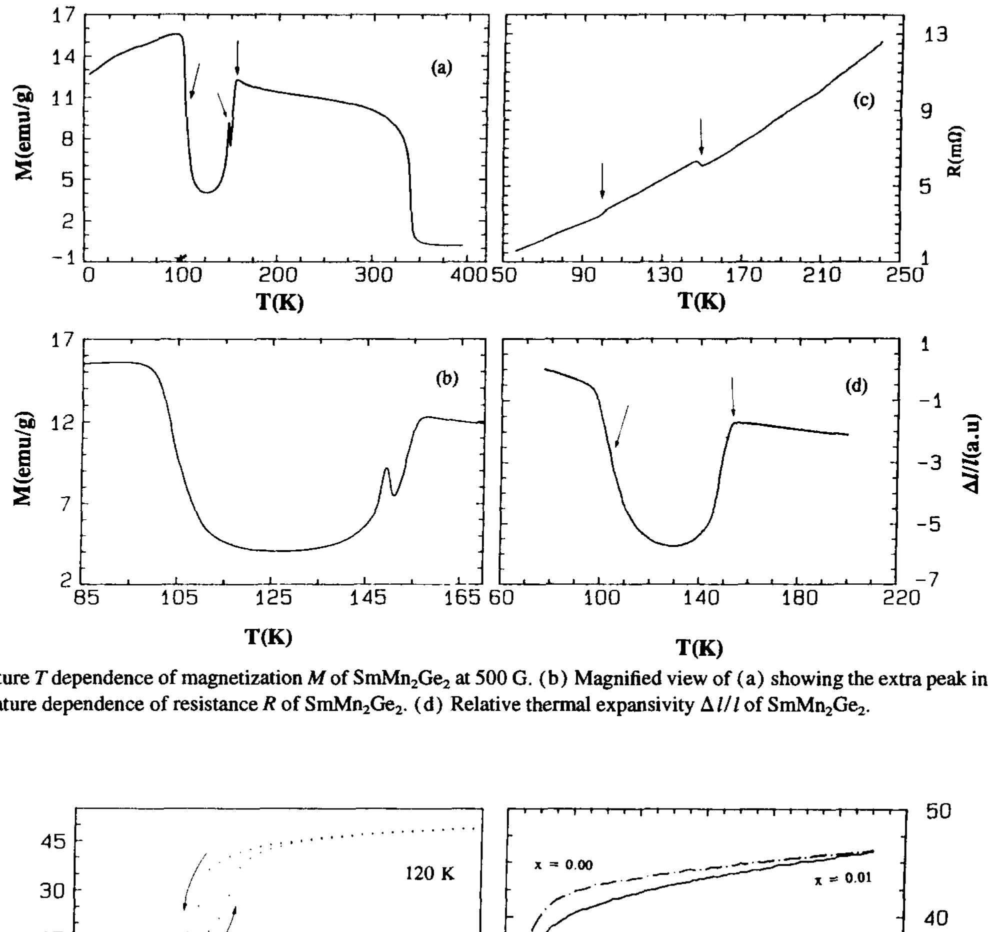

Fig. 1. (a) Temperature T dependence of magnetization M of $SmMn_2Ge_2$ at 500 G. (b) Magnified view of (a) showing the extra peak in the antiferromagnetic region. (c) Temperature dependence of resistance R of $SmMn_2Ge_2$. (d) Relative thermal expansivity $\Delta l/l$ of $SmMn_2Ge_2$.

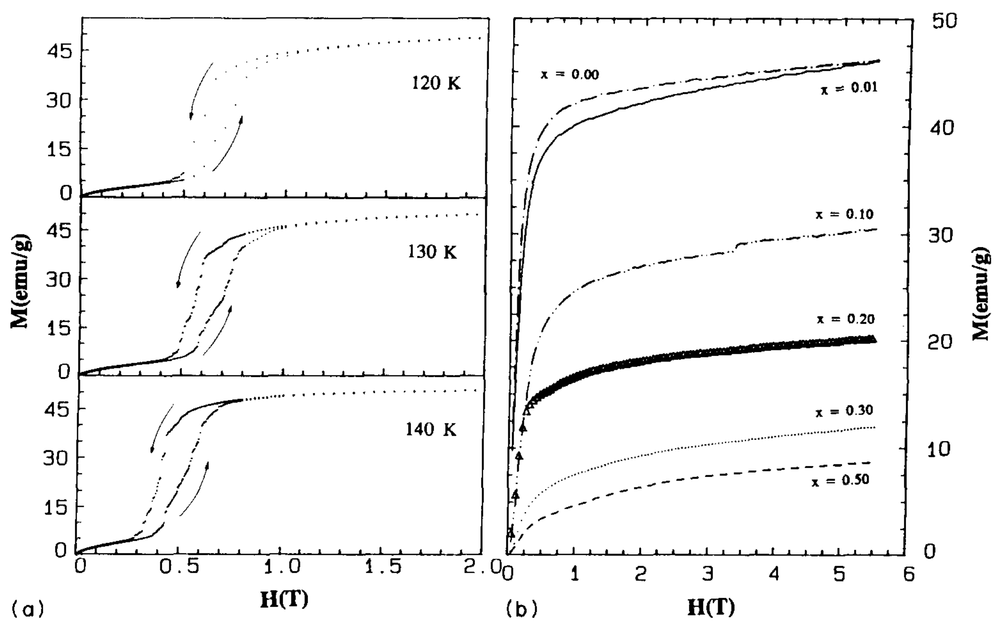

Fig. 2. (a) Magnetization M as a function of applied magnetic field H at different temperatures in the antiferromagnetic region of $SmMn_2Ge_2$. (b) Magnetization as a function of applied magnetic field of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ for $x = 0.0$–0.5 at 5 K.

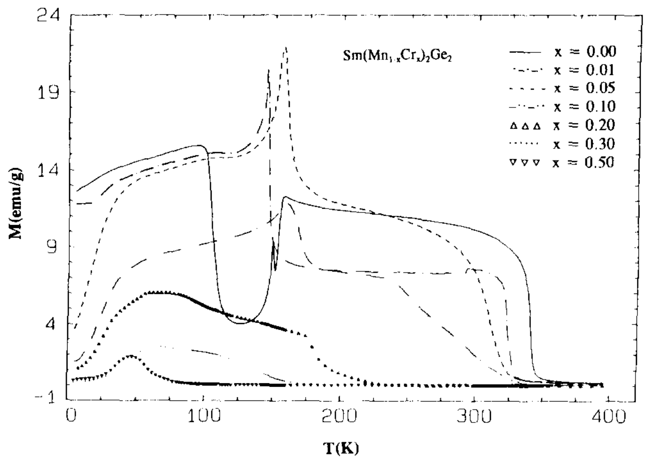

Fig. 3. Magnetization M of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ samples as a function of temperature T for $x = 0.0$–0.5 at 500 G.

suggests first-order phase transitions from ferro- to antiferromagnetic order with increasing temperature at T_1 and from anti- to ferromagnetic with increasing temperature at T_2.

From the magnetization measurements on single-crystal $SmMn_2Ge_2$ Fujii et al. [9] conclude that below $T_c = 340$ K the magnetic moments are ordered ferromagnetically along the c axis. In the antiferromagnetic region $T_1 < T < T_2$ the Mn interlayer coupling is antiferromagnetic along the c axis with Sm moments being disordered. Finally, below T_1 Mn as well as Sm moments are ordered ferromagnetically along the ⟨110⟩ direction.

In magnetization data (Fig. 1(a)) we observe a clear sharp peak at $T_{SR} = 149$ K just below the ferro- to antiferromagnetic transition. This suggests that at $T_{SR} = 149$ K there is a spin reorientation of Mn moments from the c axis ⟨001⟩ direction to the basal plane {110⟩ direction which appears as a stable antiferromagnetic phase down to T_1. However, this point needs further investigation.

A magnetic field induced transformation from antiferromagnetic phase to ferromagnetic phase is achievable with modest applied magnetic fields. The magnetization M as a function of applied magnetic field at constant temperatures $T_1 < T < T_2$ in the antiferromagnetic phase is presented in Fig. 2(a). It can be seen that as the field increases the sample goes from an antiferromagnetic to a ferromagnetic order with a sharp increase in magnetization at a critical field and that the field-induced transition is complete above 1 T. We also observe a field hysteresis suggesting that the field-induced transition from the antiferro- to ferromagnetic state is first order.

3.2. $Sm(Mn_{1-x}Cr_x)_2Ge_2$

The magnetic properties of Cr-substituted $Sm(Mn_{1-x}Cr_x)_2Ge_2$ were investigated for Cr concentrations $x = 0$, 0.01, 0.05, 0.1, 0.2, 0.3 and 0.5. In Fig. 3 we present the magnetization of zero field cooled samples as a function of temperature at a constant applied magnetic field of 500 G. From magnetization data (Fig. 3) we make the following observations: (i) the antiferromagnetic phase disappears for Cr concentrations $x \geqslant 0.05$; (ii) the temperature T_c of the paramagnetic to ferromagnetic transition decreases monotonically with increasing Cr concentration; (iii) for $x = 0.5$ there is only one magnetic transition at 47 K. The overall magnetic properties of these compounds can be seen in Fig. 3 for a Cr concentration $x = 0.10$ for example. As the temperature decreases below $T_{c1} = 260$ K the system goes from a paramagnetic to a ferromagnetic phase and further a decrease in temperature leads to a second ferromagnetic phase below

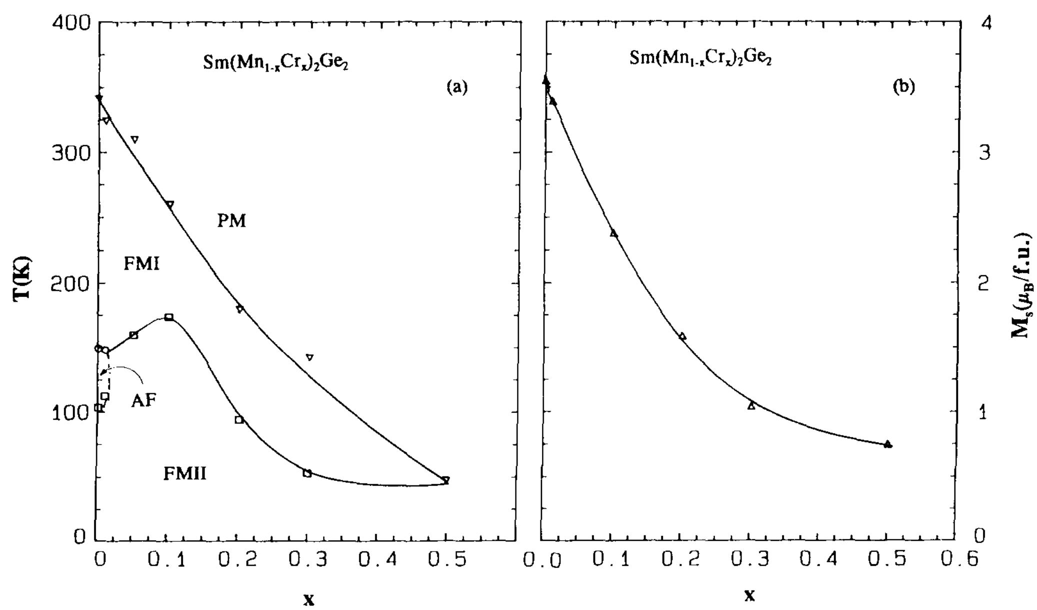

Fig. 4. (a) Magnetic transition temperatures of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ as a function of x. Different phases, PM, FMI, AF and FMII, have been designated. (b) Saturation magnetization M vs. x of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ samples at 5 K.

$T_{c2} = 174$ K. The ferromagnetic phase in the range $T_{c2} < T < T_{c1}$ is due to the ferromagnetic alignment of the manganese moments; below T_{c2} the manganese as well as samarium magnetic moments are ordered ferromagnetically.

The magnetic phase diagram determined from magnetization data of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ as a function of cr concentration x is presented in Fig. 4(a). We notice that there are primarily three magnetic phases: (1) the paramagnetic phase (PM), (2) the ferromagnetic phase I (FMI) which is probably due to Mn moment ordering and (3) the ferromagnetic phase II (FMII) due to Mn as well as Sm moment ordering. For a small range of cr concentration ($x < 0.05$) there is an antiferromagnetic phase (AF). This AF phase is presently being investigated in detail.

The magnetization M as a function of applied magnetic field for $Sm(Mn_{1-x}Cr_x)_2Ge_2$ at a constant temperature of 5 K is presented in Fig. 2(b). The magnetization decreases with increasing Cr concentration. The saturation magnetization M_s determined from Fig. 2(b) is seen in Fig. 4(b). M_s decreases monotonically as the concentration of Cr increases increases and approaches a constant value for $x \geqslant 0.03$.

4. Summary

We have studied the magnetic properties of polycrystalline $SmMn_2Ge_2$. We have found a sharp peak at $T_{SR} = 149$ K just below the ferro- to antiferromagnetic transition ($T_2 = 158$ K). This sharp peak at $T_{SR} = 149$ K may be due to spin reorientation of Mn moments. We have investigated the magnetic phase diagram of $Sm(Mn_{1-x}Cr_x)_2Ge_2$. It is found that the antiferromagnetic phase in $SmMn_2Ge_2$ completely disappears for Cr concentrations of less than 5 at.%. This result confirms the suggestion by Fujii et al. [9] and Szytula and Leceijewicz [1,2] that the magnetic properties of $SmMn_2Ge_2$ are very sensitive to in-plane Mn–Mn distances. More recently, Brabers et al. [10] have clearly demonstrated the dependence of magnetic phase transitions on the Mn–Mn distances in $SmMn_2Ge$ and related compounds. We have determined the magnetic phase diagram of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ as a function of Cr concentration. A more detailed investigation of the magnetic properties of $Sm(Mn_{1-x}Cr_x)_2Ge_2$ for $x \leqslant 0.05$ is currently being pursued.

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