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Magnetic and thermodynamic properties of selected SmT_2X_2 ternaries

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Abstract

Four ternary SmT_2X_2 (T = Fe, Co; X = Si, Ge) compounds have been synthesized in polycrystalline form, characterized by X-ray powder diffraction and studied in detail by means of magnetization and specific heat measurements in the temperature range 200–300 K and in magnetic fields up to 9 T. All the four compounds have been found to crystallize in the tetragonal $ThCr_2Si_2$-type structure with the space group *I*4/*mmm*. Both germanides, $SmCo_2Ge_2$ and $SmFe_2Ge_2$, exhibit a susceptibility maximum at ∼14 and ∼6 K, respectively, and a corresponding specific heat anomaly, which can be attributed to the transition from paramagnetic to antiferromagnetic state. Whereas the magnetization of $SmCo_2Ge_2$ is nearly linear with magnetic field increasing up to 9 T, the other germanide at 2 K undergoes a clear metamagnetic transition in magnetic fields above 2 T. Also in case of $SmCo_2Si_2$ magnetization and specific heat data can be conceived with antiferromagnetism at temperatures below $T_N \sim 17$ K. The $SmFe_2Si_2$, however, is apparently ferromagnetic below ∼3.5 K as evidenced by anomalies in the temperature dependence of the specific heat and ac-susceptibility and their evolution in magnetic fields, as well as by the evolution of magnetization curves with temperature.

Keywords: SmT_2X_2 compounds; Antiferromagnetic ordering; $ThCr_2Si_2$-type structure

1. Introduction

The ternary intermetallics RET_2X_2 (RE—rare earth, T—transition d-metal, X—p-metal) often crystallize in the tetragonal $ThCr_2Si_2$-type with RE ions occupying the 2a (0,0,0) positions, transition-metal ions sitting on the 4d (0,1/2,1/4) sites and and the X ions on 4e (0,0,*z*) ones with *z* close to 0.38.

In these materials, a wide variety of ground states is observed ranging from unconventional superconductivity to strong ferromagnetism [1]. The large number of available RET_2X_2 compounds offers a large playground for systematic studies of behavior of the rare earth ions exposed to various chemical surroundings of T and X species while the symmetry of the RE-ion neighborhood remains intact by chemical composition.

Despite the intensive studies focused on the RET_2X_2 compounds performed by many researchers in the past little is known about Sm based compounds. The only well known examples are the $SmMn_2Ge_2$, which undergoes several magnetic phase transitions in a wide temperature range and exhibits a giant magnetoresistance [2,3] and its silicide analogue, $SmMn_2Si_2$, where the magnetic transitions have been reported at 398 K (AF ordering), 230 and 120 K (order-to-order transitions), and 35 K (transition to ground-state ferromagnetic state) [4,5].

While stable manganese magnetic moments are formed and antiferromagnetically ordered in these compounds [4], Fe has been reported to be nonmagnetic in the whole $REFe_2Si_2$ series [6]. The $SmFe_2Si_2$ compound was reported to be nonmagnetic down to 4.2 K [7].

When alloying Mn with Fe in the transition-metal sublattice (up to the ratio of 1:1) [8], the high-temperature ferromagnetic state in the germanide is suppressed and the ordering temperature of the low-temperature FM phase becomes reduced. The lack of solid information on the

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Table 1
Lattice parameters for studied SmT_2X_2 compounds

	$SmFe_2Si_2$	$SmCo_2Si_2$	$SmFe_2Ge_2$	$SmCo_2Ge_2$
a	3.95983	3.93241	4.0523	4.0123
c	10.0093	9.9877	10.3213	10.1124
V	39.635	39.276	41.825	40.574
a/*c*	2.528	2.54	2.547	2.52

SmT_2X_2 compounds motivated us to synthesize the 122 silicides and germanides with T = Fe and Co.

2. Sample preparation and characterization; experimental

The polycrystalline samples of $SmCo_2Si_2$, $SmFe_2Si_2$, $SmCo_2Ge_2$ and $SmFe_2Ge_2$, were prepared by arc melting of stoichiometric amounts of constituent elements under protective Ar atmosphere (purity 6N). The purity of components was at least 3 N. Typical sample mass of 2–3 g was used and each sample was remelted for about three times in order to obtain good homogeneity of the sample.

The obtained materials were characterized by means of microprobe and X-ray diffraction (using Cu Kα with monochromator to eliminate Fe extinction) techniques, the lattice parameters are given in Table 1. All compounds were found to crystallize in the tetragonal $ThCr_2Si_2$-type structure with the space group *I*4/*mmm*. In the Fe based materials a tiny amount of grain boundary precipitates of the Fe_xX_y-type ferromagnetic at room temperature has been detected by microprobe.

The specific heat was measured in the temperature range 2–300 K on plate shaped samples of a typical mass of ∼5 mg using double relaxation technique. The measurements of magnetic properties were performed on randomly oriented fine powder fixed by nonmagnetic glue. Magnetization and susceptibility data have been corrected by subtracting the spurious signal from ferromagnetic impurity. These experiments were performed using PPMS-9T apparatus from Quantum Design in the Joint laboratory for Magnetic Measurements in Prague (http://195.113.32.128/jlms/En/Default_en.htm).

3. Results and discussion

The temperature dependence of the specific heat measured for $SmCo_2Ge_2$ compound shows a broad maximum around ∼14 K, which corresponds to the temperature of the cusp observed on the temperature dependence of magnetic susceptibility as can be seen in Fig. 1. These temperature dependences remain intact in magnetic fields up to 9 T, which is in agreement with the linear dependence of magnetization on magnetic fields up to 9 T observed at temperatures down to 2 K. These results can be tentatively attributed to paramagnetism $SmCo_2Ge_2$ at temperature above 14 K and the transition to antiferromagnetic state around this temperature. The observed linearity of magnetization curve up to 9 T suggests that the metamagnetic transition connected with suppression of antiferromagnetism is lifted to considerably higher magnetic fields, which is consistent with the relatively high value of T_N and expected low value of Sm magnetic moment.The low-temperature upturn seen in Fig. 1 both in *C*/*T* (specific heat over temperature) and susceptibility data may indicate an onset of a magnetic phase transition below 2 K. This speculation, however, should be tested by measurements to temperatures in mK range. Also the silicide counterpart of this compound shows clear attributes of antiferromagnetism (see Fig. 2). This material is characterized by a sharp peak at ∼17 K in the temperature dependence of the specific heat which roughly coincides with the cusp on the temperature dependence of the susceptibility. Also this material is characteristic by the invariability of specific heat and susceptibility anomalies around T_N and linearity of magnetization with respect to applied magnetic fields up to 9 T, which points to a high value of critical field for metamagnetism in these two antiferromagnets.

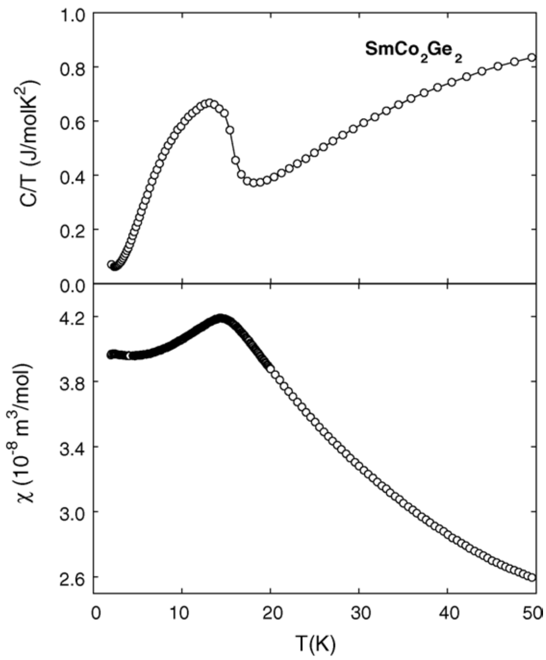

Fig. 1. The temperature dependence of *C*/*T* in zero field (up) and the magnetic susceptibility (down) in 9 T of the $SmCo_2Ge_2$ compound.

The striking difference in specific-heat anomaly between the $SmCo_2T_2$ silicide and germanide may indicate the difference in microstructure of the two materials. Whereas the sharp specific-heat peak observed for $SmCo_2Si_2$ maybe considered as a hallmark of a well stoichiometric material without extended crystal structure defects the broad feature on the temperature dependence of the specific heat of $SmFe_2Ge_2$ may indicate certain spread of off-stoichiometry over the sample.

Magnetization data collected for the $SmFe_2Ge_2$ compound also provide strong arguments for antiferromagnetism

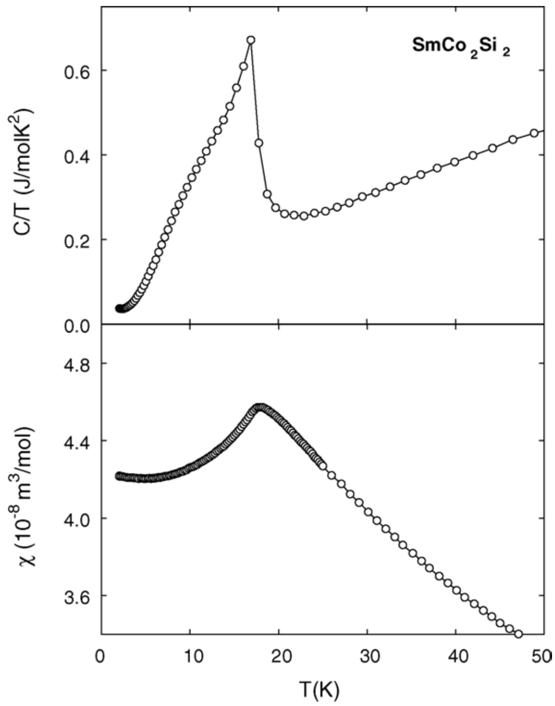

Fig. 2. The temperature dependence of *C*/*T* in zero field (up) and the magnetic susceptibility (down) in 9 T of the $SmCo_2Si_2$ compound.

at low temperatures. The temperature dependence of magnetization (Fig. 3) peaks at ∼6 K (T_N) and shows some additional structure between 4 and 5 K. These features can be tentatively attributed to magnetic phase transitions in this material. The upper one may be identified with the transition from high-temperature paramagnetism to the low-temperature antiferromagnetism and the lower one is probably marking an order-to-order (AF1–AF2) transition. The magnetization curve at 2 K (see Fig. 4) clearly confirms the low-temperature metamagnetism in $SmFe_2Ge_2$ by showing a significant metamagnetic transition above 2 T followed by a slow saturation with further increasing magnetic field.

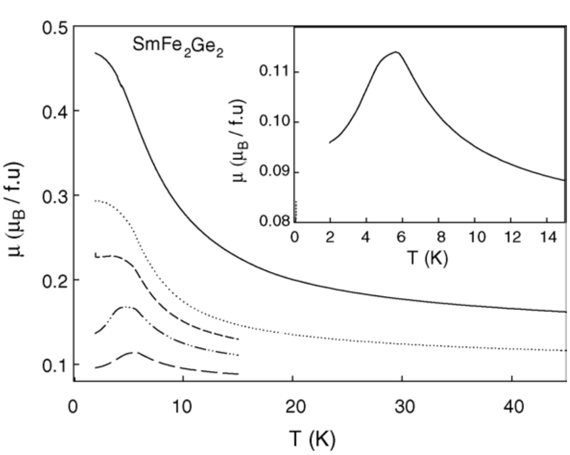

Fig. 3. The temperature dependence of the magnetization of the $SmFe_2Ge_2$ compound in various fields—1, 2, 3, 4 and 9 T (from lower to upper curve). Inset shows the low-temperature detail of the magnetization in 0.5 T.

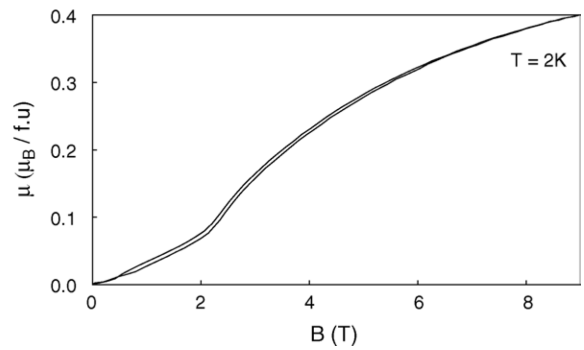

Fig. 4. The magnetization curve of the $SmFe_2Ge_2$ compound at 2 K.

The metamagnetic transition becomes smeared out with the increasing temperature and vanishes above T_N.

The last studied compound, $SmFe_2Si_2$ shows strikingly different behavior in comparison with the previous three SmT_2X_2 counterparts. The specific heat in magnetic fields and temperature evolution of magnetization curves indicate ferromagnetic ordering in this compound below $T_C = 3.5$ K. The Curie temperature is marked by a narrow peak in the temperature dependence of the specific heat and a sharp maximum in the real part of the ac-susceptibility (Fig. 5). The specific-heat anomaly becomes gradually broadened and it is lifted to higher temperatures when the magnetic

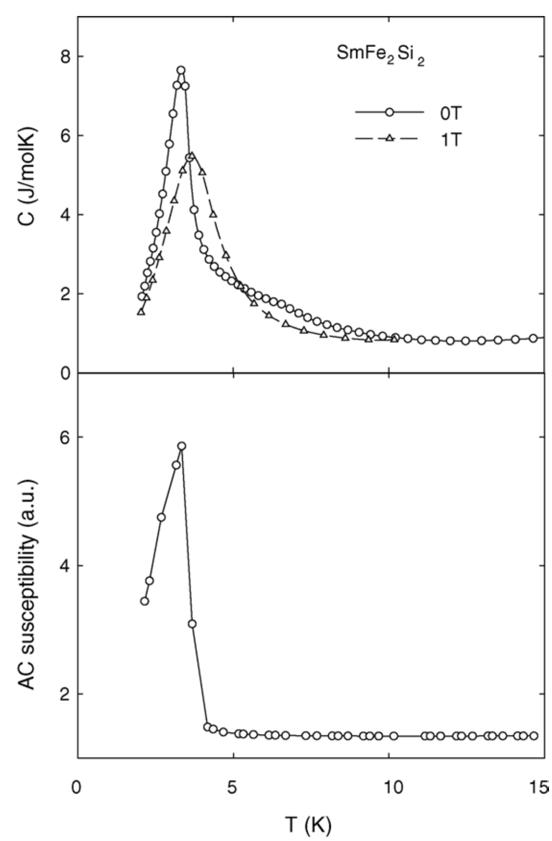

Fig. 5. The temperature dependence of the specific heat (up) and the real part of ac-magnetic susceptibility (down) of the $SmFe_2Si_2$ compound.

field is applied. The ferromagnetic ordering is expected to originate from weakly interacting Sm magnetic moments, whereas Fe sites remain nonmagnetic similar to other $REFe_2X_2$ compounds.

4. Conclusions

We have prepared four novel tetragonal compounds of SmT_2X_2-type (T = Fe and Co, X = Si and Ge) in polycrystalline form and characterized the materials by microprobe and powder X-ray diffraction.

All the four compounds crystallize in the tetragonal $ThCr_2Si_2$-type structure. Both germanides, $SmCo_2Ge_2$, $SmFe_2Ge_2$ and $SmCo_2Si_2$ shows a susceptibility maximum and a corresponding specific-heat anomaly at ∼14, 6 and 17 K, respectively. These features are attributed to magnetic phase transition from paramagnetic to the low-temperature antiferromagnetic state. Whereas the magnetization of $SmCo_2Ge_2$ and $SmCo_2Si_2$ is nearly linear with magnetic field increasing up to 9 T, the other germanide at 2 K undergoes a clear metamagnetic transition in magnetic fields above 2 T. In contrast to the the previous three SmT_2X_2 counterparts, the $SmFe_2Si_2$ is apparently ferromagnetic below ∼3.5 K as evidenced by anomalies in the temperature dependence of the specific heat and ac-susceptibility and their evolution in magnetic fields, as well as by the evolution of magnetization curves with temperature.

For further investigation of these compounds monocrystalline samples are desirable to investigate the role of magnetocrystalline anisotropy in these compounds.

Acknowledgements

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