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Competing exchange interactions and magnetic anisotropy of $La_{1-x}Tb_xMn_2Si_2$

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ABSTRACT

Crystal structure, magnetization and magnetic susceptibility have been studied for the $La_{1-x}Tb_xMn_2Si_2$ ($0 \le x \le 1$) polycrystalline and quasi-single crystalline samples. It has been shown that, at low temperature $T = 4.2$ K, substitution of the terbium for lanthanum leads to recurred change of the type of interlayer Mn-Mn magnetic ordering. For the compounds with $x < 0.2 \approx x_{c1}$ the manganese magnetic moments of adjacent layers are ordered ferromagnetically, in the concentration range $0.2 < x < x_{c2}$ (the critical concentration x_{c2} is between 0.4 and 0.6) the antiferromagnetic order is realized, while for $x > x_{c2}$ the Mn sublattice is again ferromagnetically ordered and, due to the negative Tb–Mn interaction, ferrimagnetic structure is formed. Using the magnetization data, the concentration magnetic phase diagram has been suggested. The observed variation of the type of magnetic ordering has been explained in terms of the change of interatomic Mn-Mn distances and a competition of the Tb–Mn, Mn–Mn and Tb–Tb interlayer exchange interactions.

1. Introduction

Intermetallic compounds with general formula RM_2X_2, where R is the rare earth metal; M is the transition 3d-, 4d-, or 5d-metal; X is Si or Ge, crystallize in the body-centered tetragonal $ThCr_2Si_2$-type structure. The structure consists of the single-atomic layers of different elements, stacked along the crystallographic c-axis in the sequence -M-X-R-X-M-. The natural layered structure is considered to be responsible for very exciting variety of physical properties ranging from superconductivity to heavy-fermion behavior [1].

The compounds RM_2X_2 for M=Mn attract the greatest attention since they demonstrate a wide variety of magnetic structures and magnetic phase transitions. One of the reasons of the transitions is due to a strong dependence of the type of interlayer Mn-Mn ordering on the lattice parameters and on the in-plane Mn–Mn distance d_{Mn-Mn}. For the RMn_2X_2 compounds with $d_{Mn-Mn} < d_c \approx$ 0.285–0.287 nm, the resultant Mn magnetic moments of adjacent Mn layers are ordered antiferromagnetically along the c-axis, while for $d_{Mn-Mn} > d_c$ the ferromagnetic ordering is typically realized [1,2]. Within the Mn layers, collinear ferromagnetic, collinear antiferromagnetic or canted ferromagnetic ordering can form. Variation of the crystal structure parameters by means of the external hydrostatic pressure or "chemical pressure" in the quasi-ternary $R_{1-x}R'_xMn_2X_2$ and $RMn_2(Si_{1-x}Ge_x)_2$ systems allows to control magnetic structures and magnetic phase transitions in these compounds [1,3–8].

In addition to the effects of interatomic distances, an important role in formation of magnetic structures in RMn_2X_2 compounds belongs to a competition between the interlayer Mn–Mn, R-Mn, and R–R exchange interactions and magnetic anisotropy. As an example, neutron diffraction studies of the magnetic structure of ternary $TbMn_2Si_2$ compound showed that at low temperatures $T < 51$ K when the Tb moments are magnetically ordered, the canted ferromagnetic ordering of the Mn moments is formed, in spite of the in-plane Mn–Mn distance is below its critical value [9,10]. For the temperature range 51 K $< T <$ 64 K the Tb sublattice is still in the magnetically ordered state, while the resultant Mn magnetic moments of adjacent layers form collinear antiferromagnetic order. Above 64 K, the magnetic order of the Tb sublattice is destroyed by thermal fluctuations, whereas the collinear antiferromagnetic interlayer Mn–Mn coupling persists up to the Neel temperature T_N, the value of which is estimated to be 501–550 K [9,10]. Such competition of the Mn-Mn, Tb–Mn, and Tb–Tb inter- and intralayer exchange interactions is responsible for unexpectedly high values of the magnetocaloric and the exchange bias effects [11,12]. Due to the strong Tb-Mn exchange interaction, the change of the type of the interlayer Mn–Mn coupling in

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$TbMn_2Si_2$ is accompanied by much lower anomalies of the lattice parameters [12] than those observed, *e.g.*, in the $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound [13,14]. Moreover, when the Mn-Mn interlayer coupling changes from antiferro- to ferromagnetic with increasing temperature, both the *c* and *a* lattice constants decrease for the $TbMn_2Si_2$ [12], whereas the *c* parameter decreases and both the *a* parameter and the lattice volume increase for the $La_{0.75}Sm_{0.25}Mn_2Si_2$ [13]. All these finding indicate that, depending on the values of the *R*-Mn and Mn–Mn exchange interactions, the spontaneous change of the interlayer Mn-Mn coupling in the cases when $d_{Mn-Mn} \approx d_c$ and $d_{Mn-Mn} < d_c$ can be caused by different physical mechanisms.

In the present paper, in order to reveal the origin of magnetic phase transitions in layered RMn_2X_2 compounds, we studied magnetic properties of quasi-ternary $La_{1-x}Tb_xMn_2Si_2$ intermetallics. With this system, by increasing the terbium content *x* ($0 \le x \le 1$), we can gradually decrease the Mn–Mn interatomic distance from $d_{Mn-Mn} > d_c$ in $LaMn_2Si_2$ down to $d_{Mn-Mn} < d_c$ in $TbMn_2Si_2$, and simultaneously enhance the Tb–Mn and Tb–Tb exchange interactions.

2. Experimental details

The alloys $La_{1-x}Tb_xMn_2Si_2$ with the Tb content $x=0$, 0.1, 0.2, 0.25, 0.27, 0.4, 0.6, 0.8, and 1.0 were prepared by induction melting of the constituents in an argon atmosphere followed by annealing at 900 °C for one week. Powder X-ray diffraction analysis was performed at room temperature with the DRON-6 diffractometer in Kα Cr radiation.

For the compositions $x = 0$, 0.1, 0.2, 0.27, 0.4, 0.8, and 1.0, small quasi-single crystal plates with the mass of 7–12 mg were split from massive ingots. X-ray back-scattered Laue patterns confirmed that the plates consist of several crystallites, the tetragonal *c*-axis of which is oriented perpendicular to the plate plane, while the *a*-axes of crystallites are partially disoriented within the plane of the plate.

The magnetization measurements of quasi-single crystal samples were performed with Quantum Design MPMS5-XL SQUID magnetometer in magnetic fields up to 50 kOe in the temperature range from 2 up to 320 K. Magnetic studies for temperatures 300–725 K in magnetic field 10 kOe were performed for random polycrystalline samples using high-temperature high-sensitive vibrating sample magnetometers.

3. Results and discussion

According to the powder X-ray diffraction analysis, all the studied alloys are single phase with the tetragonal $ThCr_2Si_2$-type structure (space group *I*4/*mmm*). The Tb ion has smaller atomic radius than the La ion. Therefore, the lattice parameters of $La_{1-x}Tb_xMn_2Si_2$ compounds gradually decrease with increasing the Tb content (see Fig. 1). However, the *c*/*a* ratio increases with increasing *x*. It means that the crystallographic *c* parameter and interlayer distances decrease slower than the *a* parameter and the intralayer distances. For the compounds with $x \le 0.2$, the in-plane Mn–Mn distance $d_{Mn-Mn} = a/\sqrt{2}$ is appreciably larger than the critical value d_c=0.287 nm, while for the Tb content $x \ge 0.27$ it became lower than d_c. Hence, the change of the interlayer Mn-Mn coupling in $La_{1-x}Tb_xMn_2Si_2$ can be expected at a critical concentration $x_{c1} \approx 0.2$.

Temperature dependences of the magnetization *M*(*T*) of $La_{1-x}Tb_xMn_2Si_2$ samples measured in small magnetic field H=50 Oe applied along the *c*-axis are shown in Fig. 2. For the

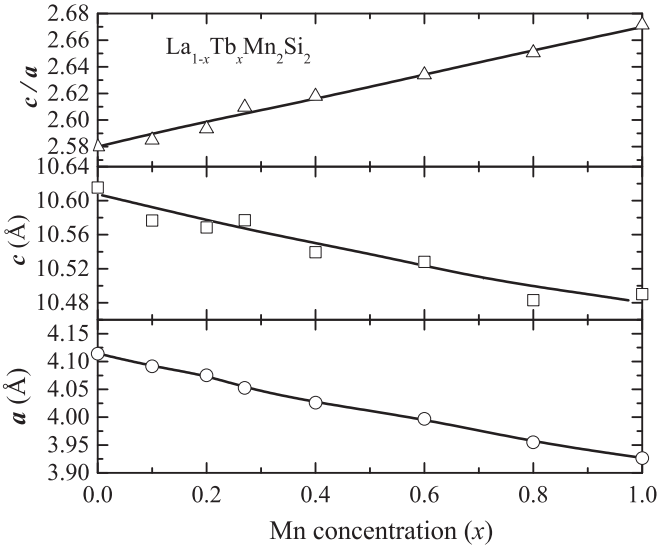

Fig. 1. Concentration dependence of the lattice parameters of $La_{1-x}Tb_xMn_2Si_2$ compounds at room temperature.

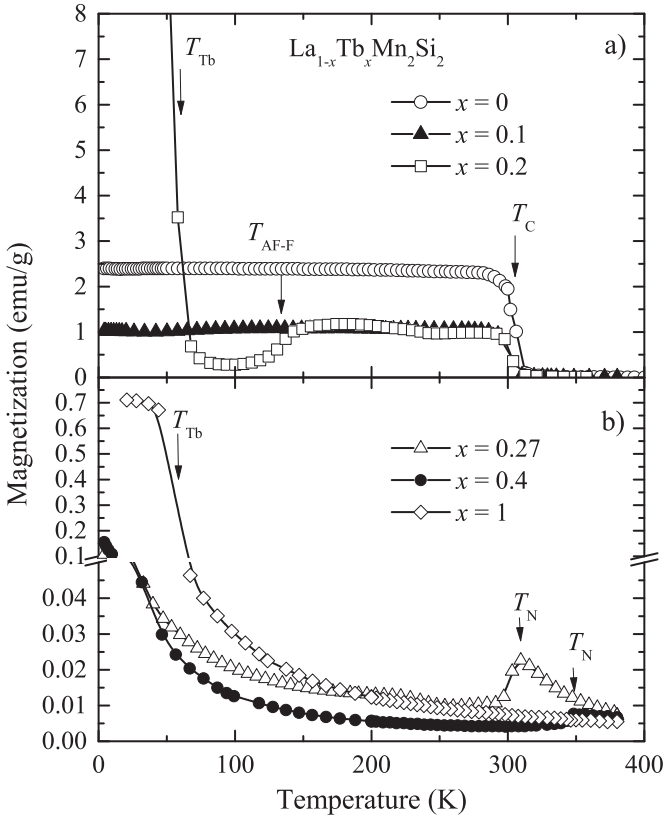

Fig. 2. Temperature dependences of the magnetization of single crystals $La_{1-x}Tb_xMn_2Si_2$ for x = 0, 0.1 and 0.2 (a) and x =0.27, 0.4 and 1 (b), in magnetic field H = 50 Oe, applied along the *c*-axis.

compounds with $x < 0.2$ the shape of the *M*(*T*) dependence is typical of ferri- or ferromagnets. Sharp decrease of the magnetization near 300 K corresponds to the Curie temperature T_C (Fig. 2a). For the compound with x=0.2, decreasing temperature leads to the magnetization decrease at the temperature $T_{AF-F} \approx 125$ K. For further temperature decrease, the magnetization sharply increases at $T_{Tb} \approx 50$ K. Here we defined the critical temperatures as the temperatures at which the derivative *dM*/*dT* has an extremum. The in-plane Mn-Mn distance d_{Mn-Mn} in the compound with x=0.2 is