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Magnetic structure of $La_{1-x}Tb_xMn_2Si_2$ compounds

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ABSTRACT

The magnetic structure of $La_{1-x}Tb_xMn_2Si_2$ compounds with $0 \leq x \leq 1$ has been studied by powder neutron diffraction and magnetization measurements on single-crystal samples. We demonstrate that at 4.2 K, with increasing the Tb concentration, a canted ferromagnetic structure of $LaMn_2Si_2$ changes at $x > 0.2$ to a canted antiferromagnetic structure. The antiferromagnetic in-plane component of Mn moment decreases with increasing concentration x and vanishes at $x > 0.5$. $TbMn_2Si_2$ is characterized by a collinear in-plane Mn ordering and ferrimagnetic structure, in which the Mn sublattice possesses the ferromagnetic interlayer alignment along the easy c-axis. Neutron diffraction study does not reveal a long-range magnetic order of Tb moments for the compounds with $x \leq 0.4$. Our results show that for concentrations $0.2 < x \leq 0.4$, a competition of the interlayer Tb-Mn, Mn-Mn exchange interactions and strong uniaxial magnetic anisotropy leads to formation of a frustrated magnetic state of Tb ions, which prevents magnetic ordering in the Tb sublattice.

1. Introduction

Intermetallic compounds RM_2X_2 (R is the rare earth element or Y; M is the 3d-, 4d-, or 5d-transition metal; X is Si or Ge) crystallize in the naturally layered body-centered tetragonal $ThCr_2Si_2$-type structure (space group $I4/mmm$). In this structure, mono-atomic layers of different elements are stacked along the crystallographic c-axis in the strict sequence -M-X-R-X-M-. The layered structure is considered to be responsible for quite an exciting variety of physical properties observed in these compounds [1,2].

Diversity of types of magnetic ordering and phase transitions were found in the RM_2X_2 compounds for M = Mn [1–3], where Mn atoms carry magnetic moment. Systematic study of different ternary and pseudoternary RMn_2X_2 compounds shows that the exchange interactions strongly depend on the in-plane Mn-Mn distance $d_{Mn\text{-}Mn}$. For the compounds with $d_{Mn\text{-}Mn} < d_c \approx 0.285$–$0.287$ nm, the axial component of Mn magnetic moments of adjacent Mn layers is ordered antiferromagnetically, while for $d_{Mn\text{-}Mn} > d_c$, the ferromagnetic ordering of the Mn magnetic moments of the Mn layers along the c-axis is realized [1,2,4]. Additionally, for several RMn_2X_2 systems with smaller lattice parameter a, the second critical distance $d_{c2} \approx 0.282$–0.284 nm was experimentally verified [5–7]. In the compounds with $d_{c2} < d_{Mn\text{-}Mn} < d_c$, both intralayer and interlayer magnetic couplings are antiferromagnetic, which leads to the formation of a canted antiferromagnetic structure. For the compounds with $d_{Mn\text{-}Mn} < d_{c2}$, no intralayer in-plane spin component is observed, Mn moments form a collinear ferromagnetic order within Mn monolayers, while the interlayer Mn-Mn coupling remains antiferromagnetic. Both the spontaneous and field-induced changes of the interlayer Mn-Mn magnetic ordering in the compositions with $d_{Mn\text{-}Mn} \approx d_c$ are accompanied by considerable volume and anisotropic lattice deformations [8]; therefore, the RMn_2X_2 compounds can be considered as materials for magnetostriction applications [9]. A variety of magnetic phase transitions makes these systems also attractive for magnetocaloric studies [10–14].

Acting upon the interatomic distances by means of external hydrostatic pressure [15,16] or “chemical pressure” in the quasi-ternary $R_{1-x}R'_xMn_2X_2$ and $RMn_2(Si_{1-x}Ge_x)_2$ systems [1–3,5–7] is considered to be a way to control magnetic structures and magnetic phase transitions in these compounds. Recently, we have performed magnetic measurements for the $La_{1-x}Tb_xMn_2Si_2$ system for which the lattice parameters decrease, while the Tb-Mn and Tb-Tb exchange interactions increase, with increasing x [17]. In this paper, in order to clarify the origin of different types of magnetic ordering in RMn_2X_2, we have studied magnetic structures of the $La_{1-x}Tb_xMn_2Si_2$ compounds for different x using powder neutron

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diffraction and magnetization measurements on single-crystal samples.

2. Samples and experimental details

The alloys $La_{1-x}Tb_xMn_2Si_2$ with the concentration $0 \le x \le 1$ were prepared by induction melting of the constituents in an argon atmosphere followed by annealing at 900°C for one week. According to the powder X-ray diffraction analysis, all the studied alloys are single-phase with the tetragonal $ThCr_2Si_2$-type structure. For the magnetization studies, the quasi-single-crystal samples in the form of plates with the mass of 7–12 mg were selected from large grains of the ingot. X-ray back-scattered Laue analysis confirmed that the plates consist of several crystallites, the tetragonal *c*-axes of which are oriented strictly perpendicular to the plate plane, while the *a*-axes of crystallites are partially disoriented within the plane of the plate. In order to avoid rotation in magnetic field, the quasi-single crystals were glued in a cube of the epoxy resin, the *c*-axis of the samples being parallel to the cube edge.

The magnetization measurements of quasi-single-crystal samples were performed in the Center of Collective Use of IMP UB RAS with Quantum Design MPMS5-XL SQUID magnetometer in magnetic fields up to 50 kOe at different temperatures.

Neutron powder diffraction studies have been carried out at 4.2 and 293 K on the D-3 diffractometer with the neutron wavelength $\lambda = 0.2429$ nm at a horizontal channel of the IVV-2M reactor (Zarechny, Russia). The data were analyzed with the Rietveld-type refinement FullProf program.

3. Results and discussion

Based on the magnetic phase diagram of the $La_{1-x}Tb_xMn_2Si_2$ system obtained from magnetization measurements [17], we selected for neutron diffraction studies compositions with the Tb content $x = 0$, 0.1, 0.2, 0.27, 0.4, and 1.0. For these compounds, the ordering temperature for the Mn magnetic moments exceeds room temperature, while the Tb moments can order well below 100 K. Neutron diffraction patterns of all the selected samples collected at 4.2 and 293 K are given in Fig. 1. The diffraction patterns at 4.2 K contain the magnetic reflections originated from two magnetic sublattices, whereas at 293 K, only the Mn moments contribute to the magnetic neutron scattering.

Using the data of symmetry analysis [18] and the Rietveld refinement of neutron diffraction patterns, we determined magnetic structures which form in the $La_{1-x}Tb_xMn_2Si_2$ compounds at different Tb content. The structures are schematically shown in Fig. 2.

In $ThCr_2Si_2$-type structure (space group *I*4/*mmm*), nuclear and magnetic contributions to the observed intensities obey the following reflection conditions [3,5,6]:

(1) (*hkl*) with $h + k + l = 2n$ for nuclear reflections.
(2) (*hkl*) with $h + k + l = 2n$ and $h + k = 2n$ and $l = 2n$ for ferromagnetic ordering between the adjacent Mn planes (e.g. (112), (200) reflections).
(3) (*hkl*) with $h + k = 2n + 1$ for antiferromagnetic ordering of Mn atoms within the (001) planes (e.g. (101), (103) reflections).
(4) (*hkl*) with $h + k + l = 2n + 1$ for antiferromagnetic ordering between the adjacent Mn planes (e.g. (111), (113) reflections).

At 4.2 K (Figs. 1a and 2a) for $LaMn_2Si_2$, the reflections conditions (2) and (3) are satisfied, which corresponds to a canted ferromagnetic structure within the Mn layers and ferromagnetic interlayer

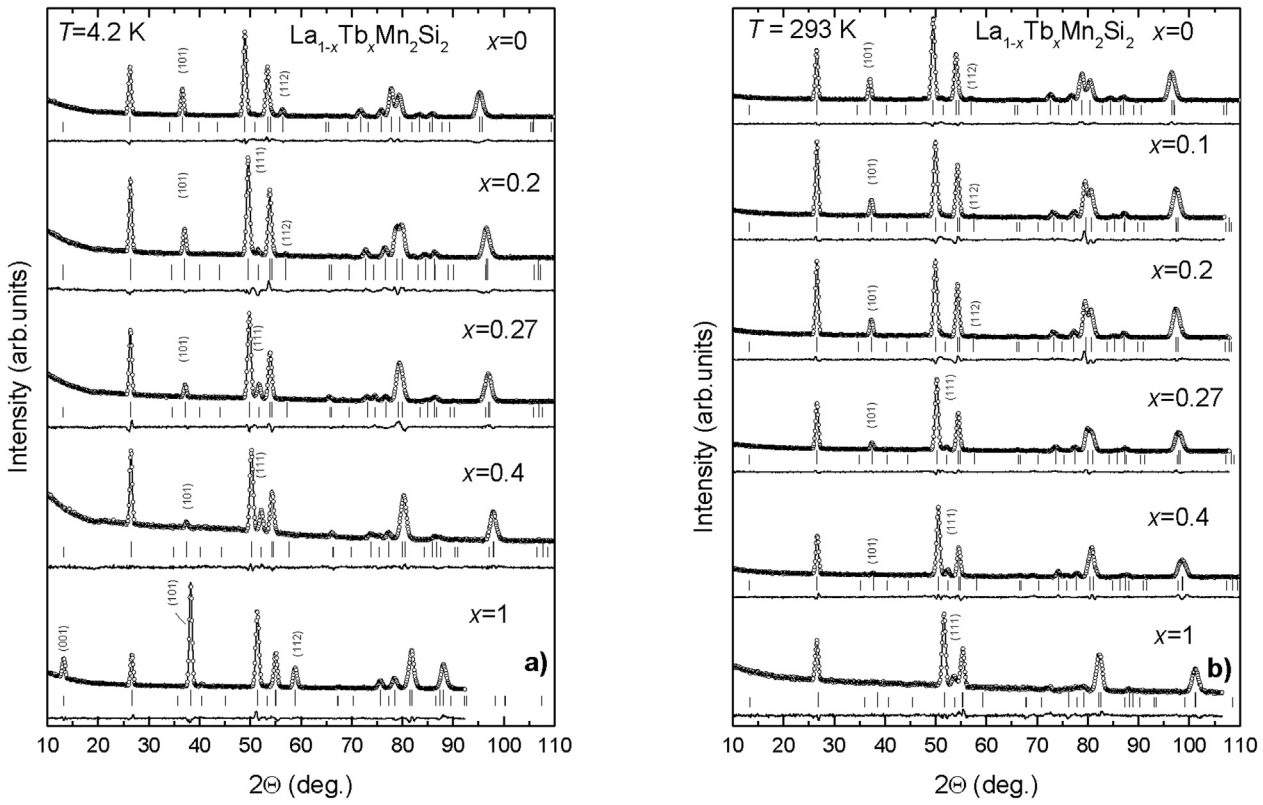

Fig. 1. Neutron diffraction patterns of $La_{1-x}Tb_xMn_2Si_2$ at 4.2 K (a) and 293 K (b). Points are experimental data, bold lines are Rietveld refinement with FullProf. Below the patterns, vertical lines point to the positions of magnetic and nuclear reflections, thin solid line shows the difference between the experimental and calculated patterns.