Journal of Alloys and Compounds 676 (2016) 74–79

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

Magnetostriction of $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound

N.V. Mushnikov*, E.G. Gerasimov

Institute of Metal Physics UB RAS, S. Kovalevskaya Str. 18, 620990, Ekaterinburg, Russia

ARTICLE INFO

Article history:
Received 1 February 2016
Received in revised form
15 March 2016
Accepted 17 March 2016
Available online 18 March 2016

Keywords:
Rare-earth intermetallics
Magnetic phase transition
Magnetostriction
Magnetovolume coupling
Magnetic ordering

ABSTRACT

The magnetostriction and magnetization are studied for a quasi-single crystalline sample of the $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound which has the spontaneous first-order antiferromagnetic to ferromagnetic (AF–F) phase transition at a temperature of 160 K. For the AF state, the field-induced AF–F transition is observed in magnetic fields directed both along the easy *c*-axis and in the basal plane. The transition is accompanied by both the volume change $\Delta V/V = 2.8 \times 10^{-3}$ and an anisotropic lattice distortion. The anisotropic magnetostriction is negative in magnetic fields applied along the *c*-axis and positive, in the basal plane. In small magnetic fields, both the volume and anisotropic magnetostrictions are proportional to the square of magnetization. In magnetic fields applied along the *c*-axis, the field-induced canting of the magnetic moments leads to much larger magnetoelastic deformations than in the basal plane. The values of the magnetovolume coupling constants both along the *c*-axis and in the basal plane are in agreement with those estimated from the thermodynamic relationships. Very small volume and linear strains are observed for the ferromagnetic state at 250 K. The obtained results suggest that the magnetoelastic effects in these compounds are mainly due to the change of mutual orientations of the Mn magnetic moments of adjacent layers.

1. Introduction

Ternary intermetallic compounds RMn_2X_2 (R is a rare earth element, X is Si or Ge) crystallize in a body-centered tetragonal $ThCr_2Si_2$-type structure (space group $I4/mmm$). In this structure, the R, Mn and X atoms form separate layers stacked along the *c*-axis in the sequence –Mn–X–R–X–Mn– [1]. The compounds are of considerable interest due to a variety of magnetic structures and magnetic phase transitions. In most cases, at low temperature the magnetic moments of the Mn atoms form a canted ferromagnetic ordering within each Mn layer, while the resulting magnetic moments of adjacent Mn layers can be oriented both parallel and antiparallel to each other, thus forming canted ferromagnetic (F) or canted antiferromagnetic (AF) structures [2,3]. At elevated temperatures, a collinear in-plane antiferromagnetic structure (AF″) is observed with the antiferromagnetic alignment of the Mn atoms within the layer [4]. Additionally, magnetic R atoms form a low-temperature ferromagnetic ordering that leads to a distortion of the AF structure due to the R–Mn exchange interaction (we denote such ordering as AF′). As a limiting case, the strong R–Mn exchange interaction can provide the ferromagnetic alignment of the Mn moments [2–4].

A number of studies have shown that the type of magnetic ordering of RMn_2X_2 strongly depends on the Mn–Mn intralayer distance, d_{Mn-Mn}, and consequently, on the lattice constant $a = \sqrt{2}d_{Mn-Mn}$ [1,3,5]. For the compounds in which d_{Mn-Mn} is larger than $d_c = 2.87$ Å, the intralayer in-plane alignment is antiferromagnetic and the interlayer coupling is ferromagnetic. When d_{Mn-Mn} is between 2.84 and 2.87 Å, both the intralayer and interlayer couplings are antiferromagnetic. If the distance d_{Mn-Mn} is smaller than 2.84 Å, there is no intralayer in-plane spin component and the interlayer coupling remains antiferromagnetic. For a particular case when the Mn–Mn intralayer distance is close to d_c, the AF–F magnetic phase transition can be induced by changing temperature (as a result of the lattice thermal expansion) or by application of magnetic field. For ternary RMn_2X_2 compounds, the condition $d_{Mn-Mn} \approx d_c$ is satisfied for the $SmMn_2Ge_2$ and $GdMn_2Ge_2$ compounds in which both the spontaneous and field-induced AF–F phase transitions are observed [6,7]. The condition $d_{Mn-Mn} \approx d_c$ can be also fulfilled in the quasi-ternary $R_{1-x}R'_xMn_2X_2$ [8,9] and $RMn_2Ge_{1-x}Si_x$ [10,11] compounds where the interatomic distances can be finely tuned by changing the composition.

As a result of a strong dependence of the Mn–Mn exchange interactions on the interatomic distance, the AF–F magnetic phase

* Corresponding author.
E-mail address: mushnikov@imp.uran.ru (N.V. Mushnikov).

http://dx.doi.org/10.1016/j.jallcom.2016.03.128
0925-8388/

transition is accompanied by a considerable change in the lattice parameters [12], with the ferromagnetic state corresponding to a larger volume V of the lattice. The spontaneous volume magnetostriction upon the AF–F magnetic phase transition amounts to $\omega = \Delta V/V = 2 \times 10^{-3}$. In the AF state, near the transition temperature, the forced volume magnetostriction of the same order can be gained in a relatively small magnetic field [13]. Such considerable magnetoelastic deformation is of interest due to its potential applications in actuators, sensors and magnetomechanical devices [14]. Although the value of the magnetic-field-induced strain in $R\mathrm{Mn_2}X_2$ is lower than that observed for the magnetostructural transitions in the Ni–Mn–(Ga,In,Sn,Ge) Heusler-type alloys [15,16] or $\mathrm{Gd_5(Si_xGe_{1-x})_4}$ alloys [17], the AF–F magnetic phase transition in $R\mathrm{Mn_2}X_2$ is much less hysteretic and highly reversible, which favors practical applications. On the other hand, theoretical models of the spontaneous and field-induced first-order AF–F transition for this compounds consider magnetoelastic phenomena to be very important [18,19]. For the quasi-ternary $R_{1-x}R'_x\mathrm{Mn_2}X_2$ compositions, the spontaneous magnetostriction can promote phase separation [20,21]. Therefore, accurate measurements of the magnetostriction can shed light on the origin of magnetic phase transitions in $R\mathrm{Mn_2}X_2$.

Direct measurements of the magnetostriction accompanying the AF–F phase transition have been reported for the $\mathrm{SmMn_2Ge_2}$ [22,23], $\mathrm{GdMn_2Ge_2}$ [22,24] and $\mathrm{Gd_{1-x}Sm_xMn_2Ge_2}$ [25] compounds. The linear magnetostriction of the order of 1×10^{-3} has been found for the temperature ranges in which the applied magnetic field changes the magnetic state of the Mn system. The measurements were performed on polycrystalline samples. The analysis of the obtained data has been performed on the assumption that magnetic anisotropy of the compounds is negligibly weak and magnetostriction is completely isotropic and is determined by the magnetoelastic change of the lattice parameter a:

$$\Delta L \Big/ L_{\parallel} = \Delta L/L_{\perp} = 2\Delta a/3a, \tag{1}$$

where $\Delta L/L_{\parallel}$ and $\Delta L/L_{\perp}$ are the relative sample-length changes in the direction parallel and perpendicular to the applied field, respectively. However, the magnetization measurements on the aligned powder samples [9,26] and single crystals [27,28] revealed a considerable magnetocrystalline anisotropy due to the manganese sublattice. Besides, upon the spontaneous AF–F phase transition not only the a parameter undergoes an abrupt change but the c parameter as well. The lattice expands in the basal plane and shrinks along the c-axis [29,30]. Therefore, the transition in magnetic field should have been accompanied also by a considerable anisotropic magnetostriction. However, the anisotropic contribution to the magnetostriction can be separated only from the measurements performed on single-crystalline samples.

In the present paper, we studied the magnetostriction of a quasi-single crystal $\mathrm{La_{0.75}Sm_{0.25}Mn_2Si_2}$ sample for which the in-plane $d_{\mathrm{Mn-Mn}}$ distance is close to its critical value. For different magnetic structures realized in this compound at different temperatures, we separate the volume and anisotropic contributions to the magnetostriction. Comparing the magnetization and magnetostriction curves in the low-field regions, we determined the magnetoelastic coupling constants and estimated applicability of theoretical models that take into account the dependence of the value and sign of exchange interactions on the interatomic distances in $R\mathrm{Mn_2}X_2$.

2. Experimental details

The $\mathrm{La_{0.75}Sm_{0.25}Mn_2Si_2}$ compound was prepared by induction melting of high-purity constituent elements in argon atmosphere followed by annealing at $T = 1273$ K for one week. Powder X-ray diffraction measurements confirmed the single-phase state with the $\mathrm{ThCr_2Si_2}$-type structure and the lattice parameters $a = 4.084$ Å and $c = 10.597$ Å at room temperature. The in-plane Mn–Mn distance amounts to 2.89 Å, which is only slightly above the critical distance d_c characteristic of the $R\mathrm{Mn_2}X_2$ compounds.

A quasi-single crystal sample in the form of plate was selected from the massive polycrystalline ingot. The X-ray Laue analysis showed that the tetragonal c-axis was directed perpendicular to the plate plane, while the a-axes were disoriented within the plane of the plate.

The magnetization measurements were performed with an extraction-type magnetometer in steady magnetic fields up to 9 T produced by a superconducting magnet. The magnetostriction measurements were carried out using WK-09-031CF-350 strain gauges (Micro-Measurements, USA). A gauge glued on the sample and a reference gauge glued on a quartz plate were connected to a dc Wheatstone bridge. The uncertainty in the absolute value of magnetostriction is estimated to be less than 5%. For the magnetic field applied along the c-axis we measured the field-induced change of the sample length $\Delta L/L$ both along the c-axis and in the basal plane. When the magnetic field was applied in an arbitrary direction in the basal plane (hereafter this direction will be denoted as a-axis, though it may decline from the [001] axes of separate crystallites of our quasi single-crystalline sample), the magnetoelastic deformation was measured in three mutually perpendicular directions: along the c-axis, parallel, and perpendicular to the field direction in the basal plane.

3. Results and discussion

As it was shown in previous studies, several magnetic structures and magnetic phase transitions were observed in $\mathrm{La_{0.75}Sm_{0.25}Mn_2Si_2}$ at different temperatures [27,31]. The magnetic phase diagram obtained from the magnetization measurements

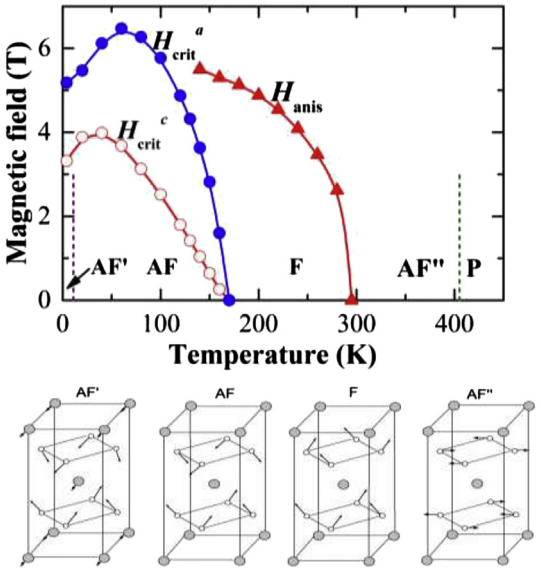

Fig. 1. H–T magnetic phase diagram of the $\mathrm{La_{0.75}Sm_{0.25}Mn_2Si_2}$ compound. The symbols AF (AF′, AF″), F and P stand for the antiferromagnetic, ferromagnetic and paramagnetic phases, respectively. Orientations of the Mn magnetic moments for different magnetic phases are schematically shown in lower part of the figure. H^a_{crit} and H^c_{crit} denote the critical field of the AF (AF′) – F transition along the a and c axes, respectively. H_{anis} is the anisotropy field measured along the hard a direction of the F phase.