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2008 J. Phys.: Condens. Matter 20 445219

(http://iopscience.iop.org/0953-8984/20/44/445219)

J. Phys.: Condens. Matter **20** (2008) 445219 (6pp) doi:10.1088/0953-8984/20/44/445219

Positive magnetoresistance and large magnetostriction at first-order antiferro–ferromagnetic phase transitions in RMn_2Si_2 compounds

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Received 8 July 2008, in final form 17 September 2008
Published 10 October 2008
Online at stacks.iop.org/JPhysCM/20/445219

Abstract
The magnetostriction and magnetoresistance associated with the field-induced and spontaneous first-order antiferro–ferromagnetic (AF–F) phase transitions have been studied for quasi-single-crystalline samples of $La_{0.25}Sm_{0.75}Mn_2Si_2$, $La_{0.25}Y_{0.75}Mn_2Si_2$ and $La_{0.27}Y_{0.73}Mn_2Si_2$ compounds with natural layered $ThCr_2Si_2$-type structure. It was found that both the spontaneous and field-induced AF–F transitions are accompanied by a large volume magnetostriction $\Delta V/V \approx 2 \times 10^{-3}$ and anisotropic linear changes of the lattice parameters $\Delta a/a \approx 1.6 \times 10^{-3}$, $\Delta c/c \approx -0.75 \times 10^{-3}$. The field-induced AF–F magnetic phase transition has been observed in magnetic fields applied both along the c-axis and in the basal plane, and the magnetostriction value is virtually independent of the direction of applied field. It has been found also that the magnetoresistance is positive in these compounds (the value of the electrical resistance in the ferromagnetic state is higher than that in the antiferromagnetic state) for the fields applied both along the c-axis and in the basal plane. The value of the magnetoresistance observed along the c-axis is 30 times as high as that in the basal plane. The obtained results indicate that the electronic band structure changes are likely responsible for the AF–F magnetic phase transitions observed in the RMn_2X_2 compounds.

1. Introduction

The intermetallic RMn_2X_2 (R is a rare earth element, X is Si or Ge) compounds crystallize in the $ThCr_2Si_2$-type structure (space group $I4/mmm$). For this structure, different atoms lie in separate atomic planes (layers) stacked along the c-axis in the next strong sequence: –Mn–X–R–X–Mn– [1]. The natural layered crystal structure gives an opportunity to consider these compounds as unique model objects for studying the phenomena which are characteristic of monoatomic multilayered films. Particularly, these crystalline layered compounds are suitable for studying the origin of the magnetoresistance associated with magnetic ordering changes in the multilayered structures.

Among the various magnetic phase transitions observed in RMn_2X_2 compounds, the greatest attention is attracted by the antiferro–ferromagnetic phase transition (AF–F) at which the change of interlayer ordering of Mn magnetic moments occurs. As was proposed in [1, 2], the AF–F phase transition in the RMn_2X_2 compounds is related to the existence of a critical intralayer Mn–Mn distance $d_c \approx 0.28$–0.287 nm (at room temperature). For $d_{Mn-Mn} > d_c$, the adjacent Mn layers are ferromagnetically ordered, while for $d_{Mn-Mn} < d_c$ an antiferromagnetic order is observed [1, 2]. Inside each layer, the Mn moments are usually ordered in a canted ferromagnetic structure with the canting angle $\theta \approx 45°–60°$ which remains unchanged through the AF–F phase transition; the net magnetic moment of the layer is always directed along the c-axis. Among ternary RMn_2X_2 compounds, the condition $d_{Mn-Mn} \approx d_c$ is only satisfied for the $SmMn_2Ge_2$ compound for which the spontaneous and field-induced AF–F phase transitions are observed [2]. The condition $d_{Mn-Mn} \approx d_c$ can also be reached in the quasi-ternary $R_{1-x}R'_xMn_2X_2$ and $RMn_2Ge_{1-x}Si_x$ compounds where interatomic distances can be varied gradually by alloying.

The nature of the AF–F phase transition remains unclear. On the one hand, the transition can result from variation of the electronic band structure in the compounds [1, 3]. However, any direct evidence of the band structure changes are still absent. For example, it was shown that a correlation exists between the lattice parameters and the density of states (DOS) at the Fermi level for these compounds, but no appreciable changes of the DOS was detected at the AF–F magnetic phase transition [4]. On the other hand, the AF–F phase transition can be described with an assumption of the existence of a strong dependence of the intralayer Mn–Mn exchange interactions on the interatomic distances [5].

The study of the magnetostriction and magnetoresistance of the RMn_2X_2 compounds may clarify the origin of the AF–F phase transition. To our knowledge, there have not yet been any direct measurements of the magnetostriction at the AF–F phase transition for RMn_2X_2 compounds, while the data on the magnetoresistance are scanty and inconsistent. Thus far, a negative magnetoresistance was observed at the field-induced AF–F phase transition for polycrystalline $Sm_{0.9}Y_{0.1}Mn_2Ge_2$ [5], single-crystalline $Gd_{0.925}La_{0.075}Mn_2Ge_2$ [6] and polycrystalline $La_{0.75}Sm_{0.25}Mn_2Si_2$ [7] samples. The negative magnetoresistance means that the electrical resistance value is higher in the AF state than in the F state. One can conclude from these data that the spin-dependent conductivity electron scattering mechanism is the main contributor to the magnetoresistance of the compounds. In contrast to these results, a positive magnetoresistance was reported at the AF–F phase transition for single-crystalline film of the $SmMn_2Ge_2$ in the basal plane [8] and for polycrystalline $SmMn_2Ge_2$ [9]. Moreover, magnetoresistance of different signs was observed for polycrystalline $SmMn_2Ge_2$ when the measurements were performed on the same sample for different time periods [10].

The aim of the present work was to study the magnetoelastic properties and to re-investigate the magnetoresistance using bulk quasi-single-crystalline $La_{1-x}R_xMn_2X_2$-type samples (R = Sm, Y), for which $d_{Mn-Mn} \approx d_c$ and, therefore, the AF–F magnetic phase transition can be induced by both temperature change and the application of a relatively weak magnetic field.

2. Experimental details

The $La_{0.75}Sm_{0.25}Mn_2Si_2$, $La_{0.25}Y_{0.25}Mn_2Si_2$ and $La_{0.73}Y_{0.27}Mn_2Si_2$ compounds were obtained from initial high-purity components by induction melting in an argon atmosphere followed by annealing at $T = 1293$ K for 1 week. X-ray diffraction showed that all the compounds are single-phase and have the $ThCr_2Si_2$-type structure.

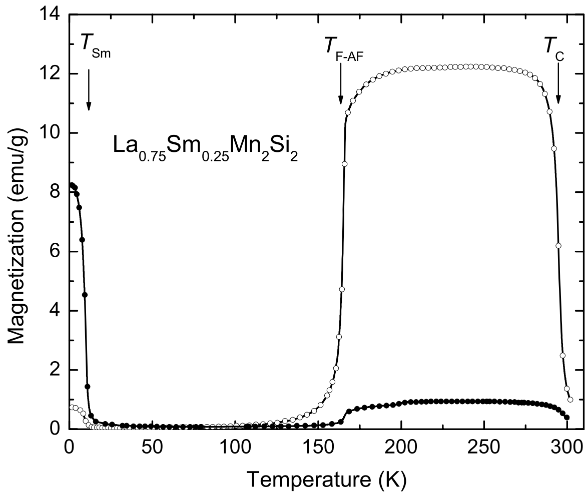

Figure 1. Temperature dependences of magnetization for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the c-axis (close symbols) and in the basal plane (open symbols) on cooling in magnetic field $H = 50$ Oe.

Quasi-single-crystalline samples with the shape of plates were selected from massive polycrystalline ingots. X-ray Laue analysis showed that the tetragonal c-axis for all samples was directed perpendicular to the plane of the plates, while in the plane of the plate a small misorientation of the a-axes was observed.

Magnetic measurements were performed using both a commercial SQUID and vibrating sample magnetometers. The electrical resistivity was measured in a superconducting coil magnet in magnetic fields directed perpendicular to the electrical current for measurements of the electrical resistance in the basal plane (R_a) and parallel to the electrical current for measurements of the electrical resistance along the c-axis (R_c). The measurements were performed by applying the electrical current $I = 1$–10 mA using the standard four-probe method. The contacts where prepared using gold wires and silver paste. The magnetoresistance $\Delta R/R$ at the AF–F phase transition was defined as $\Delta R/R = ((R(\mathrm{F}) - R(\mathrm{AF}))/R(\mathrm{AF})) \times 100\%$, where R (F) and R (AF) are the electrical resistances in the temperature- or magnetic field-induced F state and in the AF state, respectively.

3. Experimental results

Figure 1 shows the temperature dependence of the magnetization along the c-axis and in the basal plane measured by cooling in magnetic field $H = 50$ Oe for $La_{0.75}Sm_{0.25}Mn_2Si_2$. Four various magnetic ordered states are observed with critical temperatures of the magnetic phase transitions $T_C = 294$ K, $T_{AF} = 160$ K and $T_{Sm} \approx 14$ K. According to the previous results of the neutron scattering and magnetic experiments [11, 12], at $T_{AF} \leqslant T \leqslant T_C$ ferromagnetic (F) interlayer Mn–Mn ordering exists in the compound. For

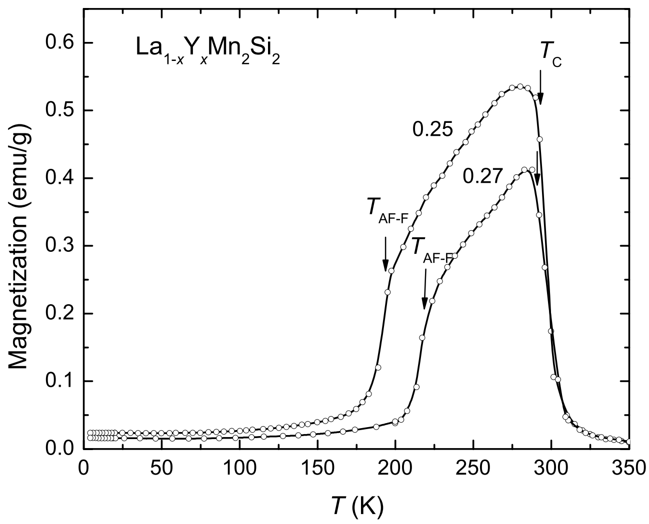

Figure 2. Temperature dependences of magnetization for $La_{0.75}Y_{0.25}Mn_2Si_2$ and $La_{0.73}Y_{0.27}Mn_2Si_2$ measured along the c-axis in magnetic field $H = 50$ Oe.

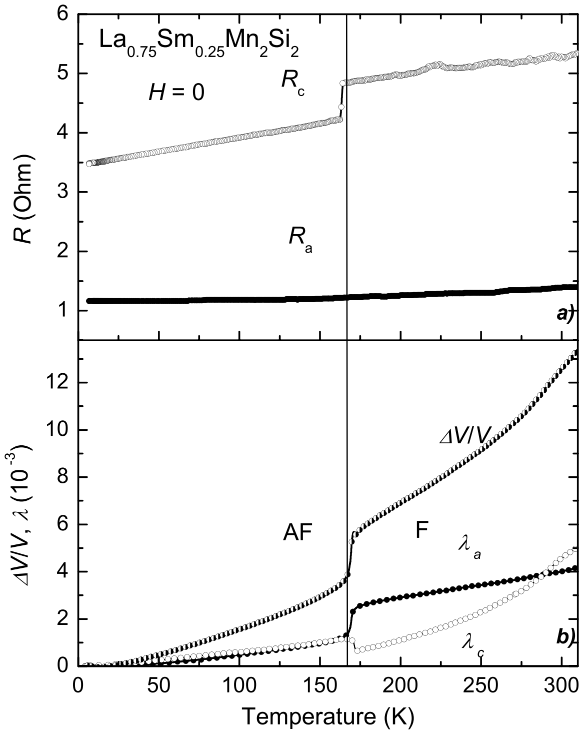

Figure 3. Temperature dependences of electrical resistance (a) and spontaneous linear magnetostriction (b) for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the c-axis (R_c, λ_c) and in the basal plane (R_a, λ_a). The volume spontaneous magnetostriction was calculated as $\Delta V/V = 2\lambda_a + \lambda_c$.

$T < T_{AF}$ the ferromagnetic type of interlayer Mn–Mn ordering changes to the antiferromagnetic (AF), and for $T < T_{Sm} = 14$ K ferromagnetic ordering of Sm magnetic moments occurs in the basal plane, which results in a small distortion of the AF structure in consequence of the ferromagnetic Sm–Mn exchange interactions and the appearance of a small in-plane ferromagnetic component of the magnetic moment (AF′ state).

Figure 2 shows temperature dependences of the magnetization measured along the c-axis on cooling in magnetic field $H = 50$ Oe for $La_{0.25}Y_{0.25}Mn_2Si_2$ and $La_{0.73}Y_{0.27}Mn_2Si_2$ compounds. As can be seen from figure 2, two different magnetic states with critical temperatures T_{AF} and T_C can be clearly separated. Spontaneous magnetization directed along the c-axis appears below the Curie temperature T_C. At $T < T_{AF}$, the spontaneous magnetization disappears. According to neutron scattering experiments [13], at $T = T_{AF}$ a spontaneous AF–F phase transition occurs.

The temperature dependence of the electrical resistance $R(T)$ measured along the c-axis exhibits a jump at the spontaneous AF–F phase transition for $La_{0.75}Sm_{0.25}Mn_2Si_2$ (figure 3(a)). The $R(T)$ dependence measured in the basal plane shows no noticeable anomalies at $T = T_{AF}$. The resistance value in the F state appears to be smaller than that in the AF state. For the other magnetic phase transitions at T_{Sm} and T_C we did not observe any anomalies of the $R(T)$ dependence either along the c-axis or in the basal plane.

Figure 3(b) shows the temperature dependence of the spontaneous linear magnetostriction for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured in zero magnetic field along the c-axis (λ_c) and in the basal plane (λ_a) on heating from 4.2 to 310 K. In this figure we also show the temperature dependence of the spontaneous volume magnetostriction $\Delta V/V = \lambda_c + 2\lambda_a$. As can be seen, the spontaneous AF–F first-order phase transition is accompanied by large discontinuous changes of the spontaneous linear magnetostriction, which is negative along the c-axis $\Delta\lambda_c \approx -0.75 \times 10^{-3}$ and positive in the basal plane $\Delta\lambda_a \approx 1.5 \times 10^{-3}$. In spite of $\Delta\lambda_a$ and $\Delta\lambda_c$ having different signs, the AF–F phase transition is accompanied by a large increase of the volume $\Delta V/V \approx 2.0 \times 10^{-3}$. Similar to the resistance measurements, no noticeable changes of either linear or volume magnetostriction was observed at the AF–AF′ phase transition.

At $T < T_{Sm}$, the application of a magnetic field induces the first-order AF′–F magnetic phase transition for $La_{0.75}Sm_{0.25}Mn_2Si_2$. The transition appears as a jump at the magnetization curves and is observed both along the c-axis and in the basal plane when the magnetic field reaches its critical values $H_\parallel$ and $H_\perp$, respectively (figure 4 (a)). The field-induced AF′–F phase transition is also accompanied by an abrupt increase of the sample length in the basal plane $\Delta\lambda_a \approx 1.5 \times 10^{-3}$ and by contraction of the sample along the c-axis $\Delta\lambda_c \approx -0.75 \times 10^{-3}$ (figure 4(b)). The values of the magnetostriction jumps at the field-induced AF′–F phase transition coincide with those at the spontaneous AF–F phase transition. The magnetic field dependence of the electrical resistance measured along the c-axis and in the basal plane also shows jumps in the critical magnetic fields which correspond to the critical fields $H_\parallel$ and $H_\perp$, respectively (figure 4(c)). The magnetoresistance value measured with increasing magnetic field applied along the c-axis $\Delta R_c/R_c \approx +3\%$ is about 30 times as high as that in the basal plane $\Delta R_a/R_a \approx +0.1\%$. When the magnetic field decreases, the $R_c(H)$ dependence along the c-axis also shows a jump, but the jump has the opposite sign to that of the initial magnetizing. Therefore, the residual resistance in zero

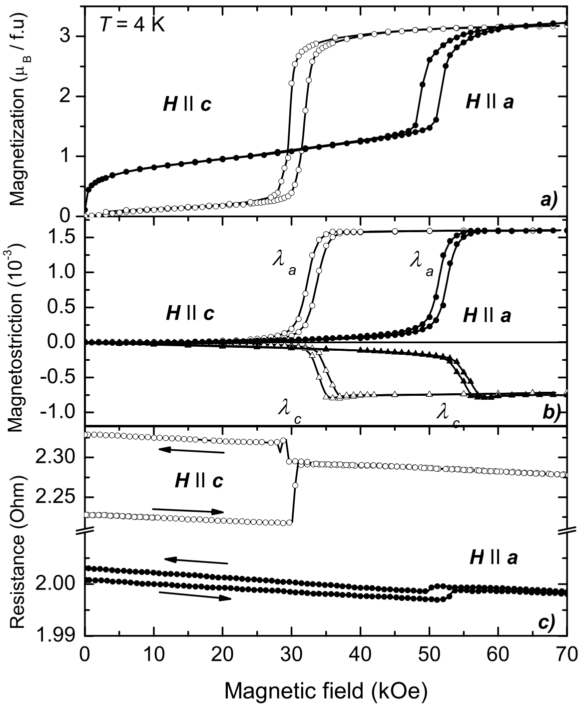

Figure 4. Magnetization curves (a) and variation of the linear magnetostriction (b) and electrical resistance (c) with magnetic field for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured in magnetic field directed along the *c*-axis (open symbols) and in the basal plane (closed symbols) at $T = 4$ K.

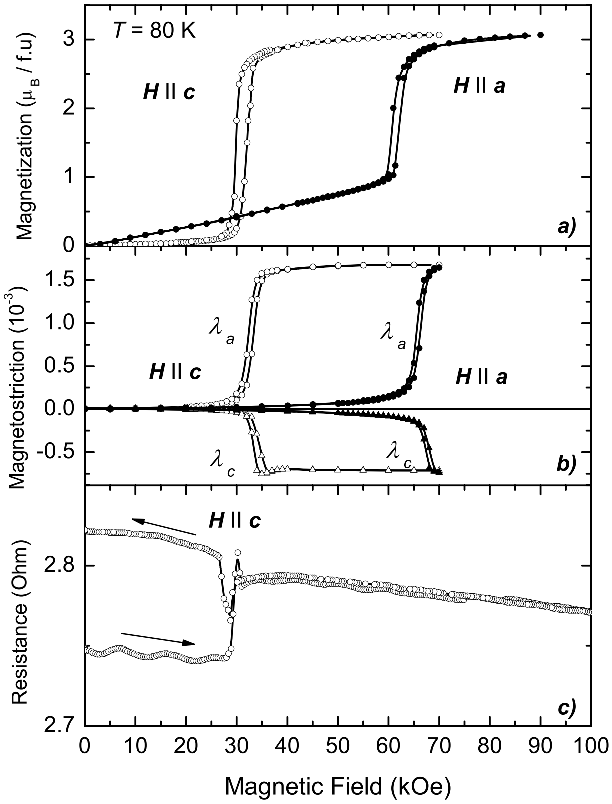

Figure 5. Magnetization curves (a) and variation of the linear magnetostriction (b) and electrical resistance (c) with magnetic field for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured in magnetic field directed along the *c*-axis (open symbols) and in the basal plane (closed symbols) at $T = 80$ K.

magnetic field increases considerably. In the basal plane, for field-up and field-down branches of the $R_a(H)$ dependence, the sign of the electrical resistance jumps remains almost the same, but the residual electrical resistance also slightly increases. In the magnetic fields above the critical fields $H_{\parallel}$, $H_{\perp}$, the magnetoresistance is negative (the electrical resistance decreases with increasing magnetic field). Such behavior is characteristic of most antiferro and ferromagnets.

At $T_{\mathrm{Sm}} < T < T_{\mathrm{AF}}$, the magnetization curves for $La_{0.75}Sm_{0.25}Mn_2Si_2$ also show jumps which correspond to the first-order field-induced AF–F magnetic phase transition (figure 5(a)). The AF–F phase transition is also accompanied by an increase of the magnetostriction in the basal plane $\Delta\lambda_a \approx 1.5 \times 10^{-3}$ and by a decrease of the magnetostriction along the *c*-axis $\Delta\lambda_c \approx -0.75 \times 10^{-3}$ (figure 5(b)). The magnetostriction values also coincide with the anomalies of the lattice expansion arising at the spontaneous AF–F phase transition. The dependence of electrical resistance on the magnetic field also shows a positive jump $\Delta R_c/R_c \approx +8.5\%$ at the AF–F phase transition. As in the case of the AF′–F phase transition, $\Delta R_c/R_c$ changes sign on decreasing the magnetic field and the residual resistance in zero magnetic field increases (figure 5(c)).

Since the magnetoresistance in the basal plane is very small, we measured the magnetoresistance for $La_{0.25}Y_{0.25}Mn_2Si_2$ and $La_{0.73}Y_{0.27}Mn_2Si_2$ along the *c*-axis only. Figures 6(a) and (b) show magnetization curve and variation of the electrical resistance with magnetic field measured along the *c*-axis for the $La_{0.73}Y_{0.27}Mn_2Si_2$ compound.

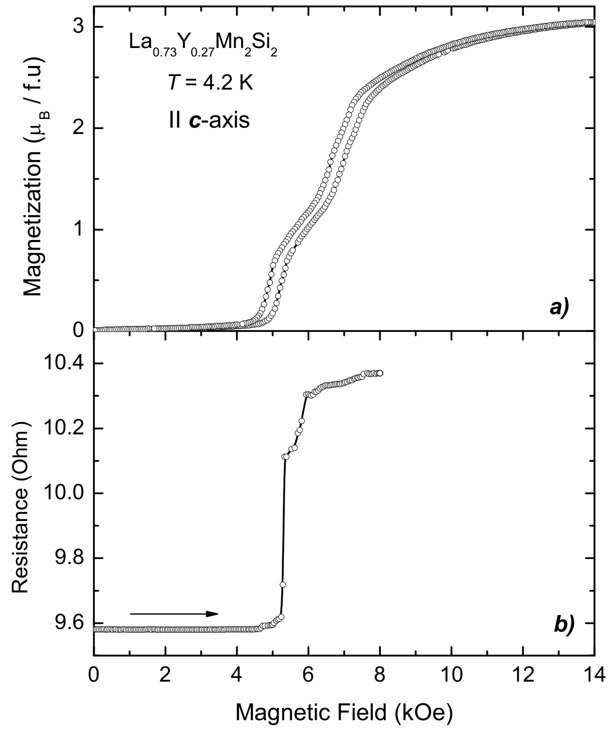

Figure 6. Magnetization curve (a) and dependence of electrical resistance on magnetic field (b) for $La_{0.73}Y_{0.27}Mn_2Si_2$ measured in a magnetic field directed along the *c*-axis at $T = 4.2$ K.

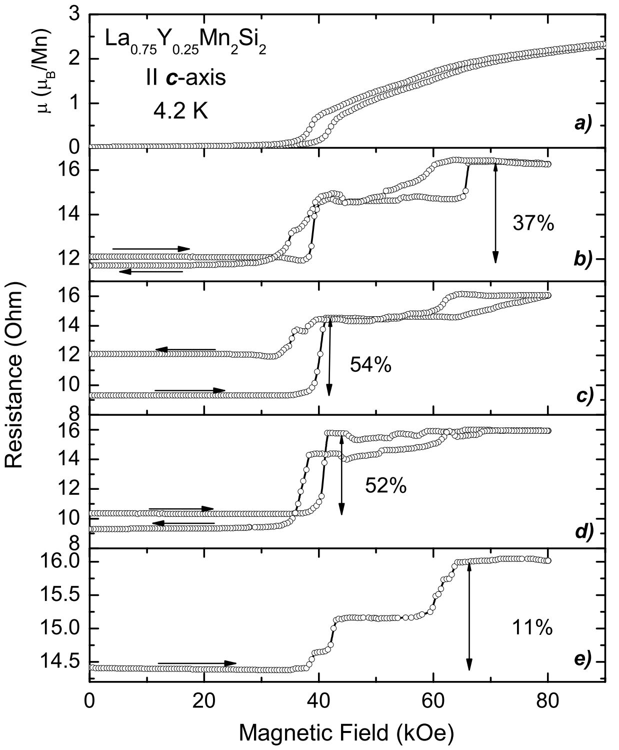

Figure 7. Magnetization curve (a) and dependences of electrical resistance on magnetic field (b)–(e) for $La_{0.75}Y_{0.25}Mn_2Si_2$ measured in a magnetic field directed along the c-axis at $T = 4.2$ K.

The magnetization curve exhibits a two-step jump which corresponds to the AF–F phase transition. The nature of the two-step character of the field-induced AF–F transition in the $La_{0.73}Y_{0.27}Mn_2Si_2$ compound is unclear at this time. According to the results of neutron scattering experiments [13], in the $La_{1-x}Y_xMn_2Si_2$ compounds an increase of Y concentration does not just lead to the change of the interlayer Mn–Mn magnetic ordering type (AF–F phase transition). The noncollinear intralayer Mn–Mn ordering disappears with increasing x. It may be expected that for $La_{1-x}Y_xMn_2Si_2$ compounds with $x = 0.27$ for which $d_{\mathrm{Mn-Mn}} \approx d_c$, the magnetic field induces both the AF–F phase transition and a change of noncollinear Mn–Mn intralayer ordering. This could result in the two-step character of the field-induced AF–F phase transition in $La_{0.73}Y_{0.27}Mn_2Si_2$. The main jump of the electrical resistance is observed in the magnetic field corresponding to the first jump on the magnetization curve (figure 6(b)). As in the case of $La_{0.75}Sm_{0.25}Mn_2Si_2$, the magnetoresistance $\Delta R_c/R_c \approx +8\%$ is positive.

Figure 7(a) shows magnetization curve measured along the c-axis for the $La_{0.75}Y_{0.25}Mn_2Si_2$ compound. As in the case of $La_{0.73}Y_{0.27}Mn_2Si_2$, the field-induced AF–F transition has a two-step shape. In order to reveal the effect of microcracks appearing at the transition, the variation of the electrical resistance with magnetic field for $La_{0.75}Y_{0.25}Mn_2Si_2$ was measured four times. As can be seen from figures 7(b)–(e), the magnetoresistance is positive in all cases but the shape of the dependences and absolute values of the magnetoresistance are significantly different.

Table 1. Magnetoresistance values at the AF–F transition for RMn_2X_2 compounds.

Compound	$\Delta R/R$ (%)	T_{AF} (K)	Reference
$Sm_{0.9}Y_{0.1}Mn_2Ge_2$ (polycrystal)	−10	185	[5]
$SmMn_2Ge_2$ (quasi-single-crystal film in the basal plane)	+8	100	[8]
$SmMn_2Ge_2$ (polycrystal)	+5; −7	110–140	[9, 10]
$La_{0.75}Sm_{0.25}Mn_2Si_2$ (polycrystal)	−27	150	[7]

4. Discussion

The performed studies of the magnetostriction for quasi-single-crystalline $La_{0.75}Sm_{0.25}Mn_2Si_2$ show that both spontaneous and field-induced AF–F phase transitions are accompanied not only by large linear anisotropic changes of the lattice parameters $\Delta a/a \approx 1.6 \times 10^{-3}$, $\Delta c/c \approx -0.75 \times 10^{-3}$, but also by large change of the volume $\Delta V/V \approx 2 \times 10^{-3}$. The measured volume change at the AF–F phase transition quantitatively agrees with the value estimated earlier using thermodynamical relations between the dependence of the critical temperature T_{AF} on external pressure and the lattice volume [12]. The field-induced AF–F phase transition is observed in magnetic fields directed both along the c-axis and in the basal plane and is accompanied by similar distortions of the crystal lattice.

The obtained results on the magnetoresistance are not so unambiguous. At first, it should be noted that the magnetoresistance observed at the AF–F phase transition is always positive for all studied compounds (measuring both along the c-axis and in the basal plane). Second, the magnetoresistance value measured along the c-axis is much larger than that in the basal plane. Moreover, irreversible changes of the residual electrical resistance in magnetic field were observed and the magnetoresistance value strongly varies from one measurement to another.

As was mentioned in the introduction, different signs of the magnetoresistance were reported for RMn_2Si_2 compounds. The results concerning the magnetoresistance at the AF–F phase transition for RMn_2Si_2 compounds are summarized in table 1 (here we do not include the $Gd_{0.925}La_{0.075}Mn_2Ge_2$ compounds [6] for which the Gd sublattice can give a considerable contribution to the magnetoresistance). As can be seen from table 1, the positive sign of the magnetoresistance determined in our study for quasi-single-crystalline compounds coincides with the only result obtained for a single-crystalline $SmMn_2Ge_2$ film [8]. The disagreement is mainly observed for polycrystalline samples. It is obvious that the strong magnetostriction distortions accompanying the AF–F phase transition complicate the magnetoresistance measurements for RMn_2Si_2 compounds because of the cracks appearing in the brittle samples. This may affect both the residual electrical resistance value and the sign of the magnetoresistance. For polycrystalline samples where the boundaries of crystallites serve as the sources of microcracks, the crackling is expected to be more pronounced than that for single crystals. Therefore, the wide scattering of the magnetoresistance data observed for polycrystalline samples and irreversible changes of the magnetoresistance observed by us for quasi-single-crystalline samples may be attributed to changes of the morphology of the sample because of the large magnetostriction deformations.

It is known that the spin-dependent magnetoresistance arises from scattering of conduction electrons on magnetic moments. For noncollinear magnetic ordering, the electrical resistance, as a rule, is larger than that for collinear ordering of the moments. In the antiferromagnetic state, the spin-dependent part of the electrical resistance is also larger than in the ferromagnetic state. For the studied RMn_2Si compounds we observe an opposite behavior.

The magnetoresistance observed for RMn_2Si_2 may be due to a change of the electronic band structure at the AF–F phase transition. The transition is accompanied by a positive in-plane magnetostriction (figure 4) which leads to an increase in the Mn–Mn interatomic distance and, therefore, to the narrowing of the 3d-band. Since the Mn-ion magnetic moment remains virtually unchanged [11, 13], one can expect an increasing density of states at Fermi level. Within the Mott s–d scattering model [14] the conduction s-electrons may fill empty states in the d-band. The probability of filling increases with increasing density of states at the Fermi level. This approach could explain the positive sign of the magnetoresistance. The observed large volume magnetostriction and field-induced AF–F phase transitions both along the *c*-axis and in the basal plane can also provide the evidence for the electronic band structure changes at the AF–F phase transition. However, a spin-dependent mechanism of scattering should also be considered including a strong anisotropy of the magnetoresistance, since the magnetoresistance is high along the *c*-axis, but it is very small in the basal plane where the intralayer ferromagnetic structure remains unchanged at the AF–F phase transition.

5. Conclusion

The results of studies of the magnetostriction and magnetoresistance for quasi-single-crystals $La_{0.75}Sm_{0.25}Mn_2Si_2$, $La_{0.25}Y_{0.25}Mn_2Si_2$ and $La_{0.73}Y_{0.27}Mn_2Si_2$ compounds at the AF–F phase transition presented in this work can be summarized as follows. The spontaneous and field-induced AF–F first-order phase transition in the Mn sublattice is accompanied by large linear and volume magnetostrictions. The magnetoresistance is positive at the AF–F phase transition both along the *c*-axis and in the basal plane. The magnetoresistance is strongly anisotropic, its value along the *c*-axis is much higher than that in the basal plane. The results obtained allow us to conclude that the AF–F phase transition in the RMn_2Si_2 compounds arises as result of electronic structure changes.

Acknowledgments

This study was supported by the RAS Program (project no. 01.2.006 13391), RFBR (grant Nos. 06-02-16951, 05-02-16087), by the Priority Program 'Quantum Macrophysics' and by Ural and Siberian Divisions of RAS (grant no. 32-2006).

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