Pressure effect on magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$

E. G. Gerasimov* and N. V. Mushnikov
Institute of Metal Physics, Ural Division of the Russian Academy of Sciences, st. S. Kovalevskya, 18, 620219 Ekaterinburg, Russia

T. Goto
Institute for Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa 277-8581, Japan
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The magnetization of a quasisingle-crystalline $La_{0.75}Sm_{0.25}Mn_2Si_2$ sample was measured under high pressures to $P=1.2$ GPa. The compound exhibits four different spontaneous magnetic phase transitions: the ordering of Sm sublattice at a critical temperature $T_{Sm}=14$ K, the ferro-antiferromagnetic phase transition at $T_{AF}=150$ K, the Curie temperature of the ferromagnetic phase at $T_C=305$ K, and finally, the transition to the paramagnetic state at $T_P=403$ K. The changes of the critical temperatures of the spontaneous magnetic phase transitions under pressure are −0.6, +203, −12, and ~−60 K/GPa, respectively. The temperature range of ferromagnetic phase shrinks with increasing pressure, and at $P \geqslant 0.6$ GPa no ferromagnetic ordering of Mn magnetic moments is observed. For the antiferromagnetic phase, application of a relatively low magnetic field produces a sharp first-order field-induced metamagnetic transition. The critical field of the metamagnetic transition depends on the direction of applied field and increases with increasing pressure. The nature of various magnetic phase transitions in RMn_2X_2 compounds is discussed using a thermodynamic analysis of the obtained results and a phenomenological model of the step change of Mn-Mn interlayer exchange interaction at the transition.

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I. INTRODUCTION

The intermetallic compounds of the RM_2X_2 family (R is rare-earth metal; M is one of the late $3d$ or $4d$ transition metals; X=Si or Ge) exhibit an extremely wide spectrum of the physical phenomena including giant magnetoresistance, superconductivity, spontaneous, and field-induced magnetic phase transitions.[1] The RM_2X_2 compounds crystallize in the tetragonal $ThCr_2Si_2$-type structure (space group $I4/mmm$). Atoms of each sort in the lattice lie within equivalent separate atomic planes (layers) staked along the tetragonal **c** axis in the strict sequence: -*M*-*X*-*R*-*X*-*M*-. Because of the naturally layered crystal structure these compounds can be considered as unique model objects for studying the physical phenomena that are intrinsic for multilayer structures and quasi-two-dimensional magnetic materials.[2]

Among the RM_2X_2 family, only in the compounds with M=Mn does the $3d$ transition metal have a nonzero magnetic moment. A wide variety of magnetic structures and magnetic phase transitions is observed for RMn_2X_2 compounds. One of the most unusual features of magnetism of RMn_2X_2 compounds is a strong dependence of the interlayer Mn-Mn exchange interaction on the distance between Mn atoms. From the analysis of the magnetic properties of ternary RMn_2X_2 compounds with different R and X elements, it was concluded that the type of interlayer Mn-Mn magnetic ordering correlates with the change of the crystallographic **a** lattice parameter that determines the intralayer Mn-Mn distance $d_{Mn\text{-}Mn}=\mathbf{a}/\sqrt{2}$.[1,3] There exists some critical in-plane Mn-Mn distance $d_c \approx 0.285-0.287$ nm, above which (i.e., at $d_{Mn\text{-}Mn} > d_c$) the Mn magnetic moments in the adjacent layers are ordered ferromagnetically (F), while for $d_{Mn\text{-}Mn} < d_c$ the antiferromagnetic (AF) ordering is realized. Inside each layer a canted ferromagnetic structure with the canting angle $\Theta \approx 45° - 60°$ is usually formed and the resulting Mn magnetic moment of the layer is directed along the **c** axis. At the same time, no correlations were found between the type of Mn ordering and the value of the crystallographic **c** parameter which determines the interlayer Mn-Mn distance.

For the RMn_2X_2 compounds for which $d_{Mn\text{-}Mn} \approx d_c$ the first-order phase transition between antiferromagnetic and ferromagnetic phases may occur by changing temperature or pressure. The nature of this transition remains unclear. On the one hand, it was assumed that the phase transition may be driven by a change of the electronic structure of the compounds, since the value of d_c is close to the critical value at which the delocalization of $3d$ electrons of Mn occurs in binary Mn-based alloys.[1,4,5] However, up to now no experimental evidences of the change of the electronic structure were found for RMn_2X_2 compounds at the AF-F transition. In particular, the value of the Mn magnetic moment practically remains unchanged at the AF-F transition. On the other hand, the AF-F phase transition in RMn_2X_2 can be qualitatively described in a model of the localized magnetic moments in the assumption of a strong dependence of the interlayer Mn-Mn exchange interaction from the intralayer Mn-Mn distances.[6–9]

Earlier we investigated the magnetic properties and magnetic structures of a series of $La_{1-x}Sm_xMn_2Si_2$ compounds in which the Mn-Mn distance changes continuously from $d_{Mn\text{-}Mn} > d_c$ to $d_{Mn\text{-}Mn} < d_c$ with increasing the Sm concentration.[9,10] In the present paper we studied the effect of hydrostatic pressure on magnetic properties of a quasisingle crystal $La_{0.75}Sm_{0.25}Mn_2Si_2$ for which $d_{Mn\text{-}Mn} \approx d_c$ to reveal the origin of the spontaneous and field-induced magnetic phase transitions in the layered RMn_2X_2 compounds.

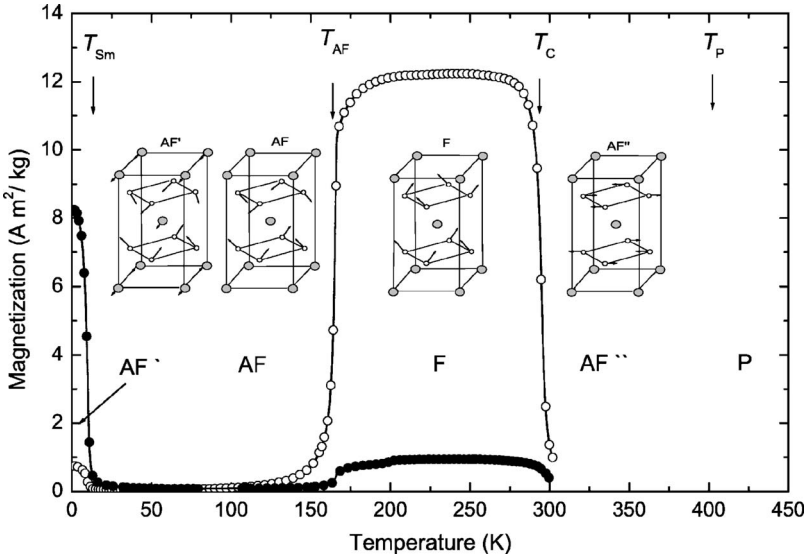

FIG. 1. Temperature dependence of the magnetization of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis (open symbols) and in the basal plane (closed symbols) in magnetic field 5 mT. The magnetic structures for different magnetic states taken from Ref. 10.

II. EXPERIMENTAL DETAILS

The $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound was prepared from high-purity elements using induction melting under an argon atmosphere followed by annealing at $T=1273$ K for one week. The sample composition was checked by powder x-ray diffraction, which confirms the $ThCr_2Si_2$-type structure characteristic of $La_{1-x}Sm_xMn_2Si_2$ compounds.

The quasisingle crystal samples in the form of plates were selected from large grains of the ingot. According to the x-ray Laue analysis, the tetragonal **c** axis of the samples was directed perpendicular to the plate plane, while the **a** axes were disoriented by a small angle within the plate plane. The size of the sample selected for the magnetization measurements under pressure is approximately $1.5\times1.5\times0.8$ mm^3 with a weight 18 mg.

The $La_{0.75}Sm_{0.25}Mn_2Si_2$ sample was compressed in a Teflon capsule filled with a liquid pressure medium fluorinert in a nonmagnetic high-pressure clamp cell made of a Ti-Cu alloy. The pressure produced in the sample at low temperatures was calibrated by measuring the Meissner effect of Pb. The uncertain in-pressure value caused by thermal expansion of the cell and pressure medium with increasing temperature is estimated to be below 10% for maximum available pressure 1.2 GPa. The total magnetization of the sample and the surrounding pressure cell was measured with an extraction-type magnetometer within temperature range 1.5–310 K in magnetic fields up to 9 T produced by a superconducting magnet. Magnetic measurements were carried out both along the tetragonal **c** axis and in an arbitrary direction within the basal plane.

III. EXPERIMENTAL RESULTS

A. Effect of external pressure on spontaneous magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$

Figure 1 shows temperature dependences of the magnetization $M(T)$ for $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured at ambient pressure in a small magnetic field $\mu_0H=5$ mT applied along the **c** axis and in the basal plane. The magnetic structures which are realized in the compound at different temperatures were determined earlier from magnetic neutron diffraction and thermal expansion measurements.[10,11] As seen from Fig. 1, four different magnetic states are observed for $La_{0.75}Sm_{0.25}Mn_2Si_2$. The critical temperatures of the magnetic phase transitions are: $T_P=405$ K, $T_C=305$ K, $T_{AF}=160$ K, and $T_{Sm}\approx14$ K. At temperatures $T>T_P$, the $La_{0.75}Sm_{0.25}Mn_2Si_2$ is in the paramagnetic state (P), at $T_C<T<T_P$ the collinear antiferromagnetic ordering of the Mn magnetic moments within the layer (in-plane antiferromagnetic state, AF″) is realized, at $T_{AF}<T<T_C$ a canted ferromagnetic structure of Mn magnetic moments is formed inside each Mn layer with ferromagnetic ordering of the magnetic moments in the adjacent layers (F), at $T_{Sm}<T<T_{AF}$ the interlayer antiferromagnetic ordering (AF) is observed, and at $T<T_{Sm}$ the Sm magnetic moments order ferromagnetically and oriented in the basal plane, which leads to a distortion of the AF structure in the Mn sublattice and formation of a new antiferromagnetic AF′ state. The AF AF′, AF, and F magnetic states and AF′-AF, AF-F, and F-AF″ phase transitions can be clearly distinguished from temperature dependence of the magnetization (Fig. 1). At the same time, the AF″ magnetic state and the AF″-P phase transition practically cannot be identified by bulk magnetic measurements.[12] The antiferromagnetic ordering of Mn moments above T_C was proved by magnetic neutron diffraction and Mössbauer studies.[12,13] Recently, an anomaly on temperature dependence of the thermal expansion coefficient[11] and heat capacity[14] was found at T_P which was attributed to the disappearance of the long-range antiferromagnetic order. These experiments give a macroscopic confirmation of existence of the AF″-P transition in RMn_2X_2 compounds.

Application of external hydrostatic pressure leads to a strong increase of T_{AF} and a small decrease of T_C temperatures [Fig. 2(a)]. The temperature range of the existence of ferromagnetic ordering shrinks with increasing pressure. At $P=0.6$ GPa the ferromagnetic phase disappears and the $M(T)$ dependence is characteristic of an antiferromagnet. It shows a maximum at 286 K [Fig. 2(b)], which may be attributed to the Néel temperature T_N. At a further increase of

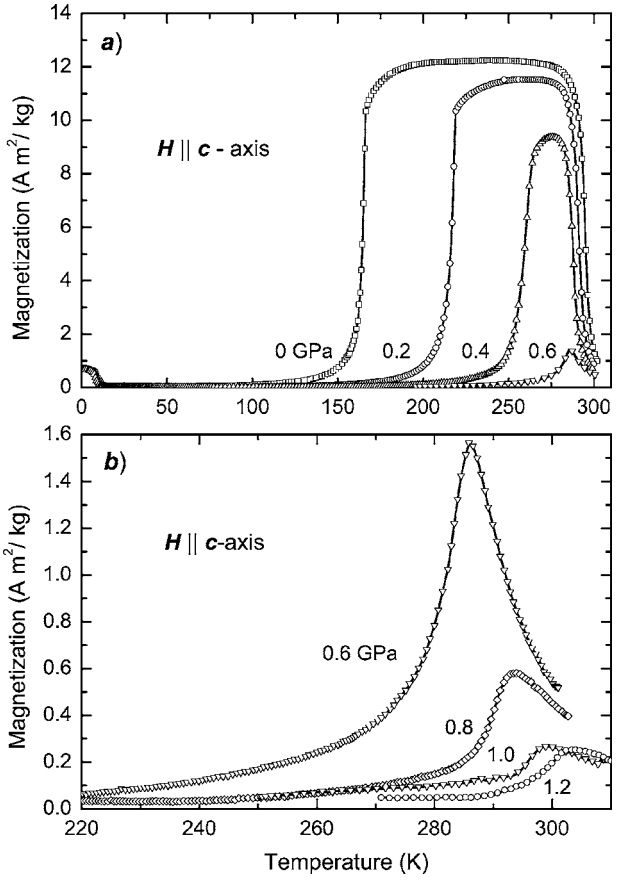

FIG. 2. Temperature dependence of the magnetization of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis in magnetic field 5 mT for different pressures $P \leqslant 0.6$ GPa (a) and $P \geqslant 0.6$ GPa (b).

pressure the Néel temperature of the compound increases, while the amplitude of the magnetization peak at the transition gradually suppresses [Fig. 2(b)]. The ordering temperature of Sm sublattice is weakly affected by pressure. This ordering is probably caused by RKKY long-range indirect magnetic exchange interaction, which is insensitive to the small interatomic distance change produced by external pressure. The value of T_{Sm} slowly decreases with increasing pressure (Fig. 3).

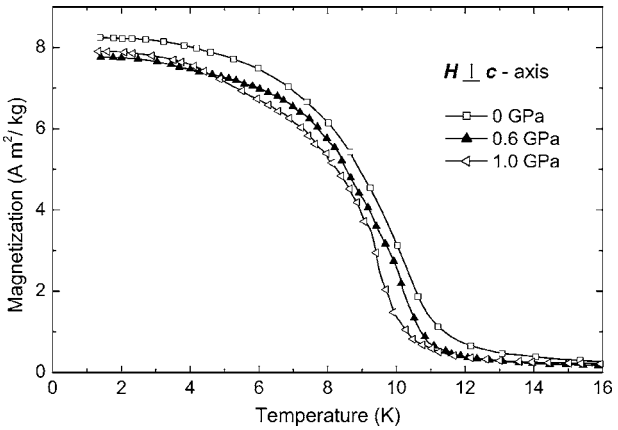

FIG. 3. Temperature dependence of the magnetization of $La_{0.75}Sm_{0.25}Mn_2Si_2$ in magnetic field 5 mT applied in the basal plane for different pressures.

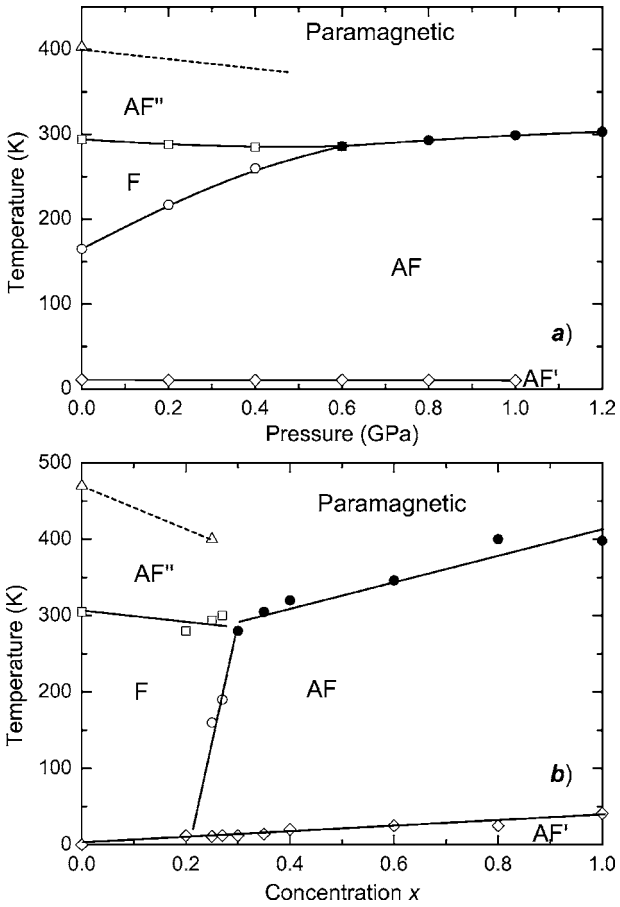

FIG. 4. *P-T* magnetic phase diagram of $La_{0.75}Sm_{0.25}Mn_2Si_2$ (a) and *x-T* magnetic phase diagram of $La_{1x}Sm_xMn_2Si_2$ (b) taken from Ref. 9.

Figure 4(a) represents the *P-T* magnetic phase diagram of $La_{0.75}Sm_{0.25}Mn_2Si_2$ where all the above results of pressure effect on spontaneous magnetic phase transitions are summarized. For the comparison, in Fig. 4(b) we show the *x-T* magnetic phase diagram of $La_{1-x}Sm_xMn_2Si_2$ system in which the lattice parameters and the elementary cell volume gradually decrease with increasing Sm concentration.[9] One can see that the *P-T* phase diagram of $La_{0.75}Sm_{0.25}Mn_2Si_2$ is qualitatively similar to the *x-T* phase diagram of the $La_{1-x}Sm_xMn_2Si_2$ series. The critical temperatures of magnetic transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ change with increasing pressure similarly with those for the $La_{1-x}Sm_xMn_2Si_2$ system with increasing Sm content. An exception is T_{Sm} temperature which increases in the $La_{1-x}Sm_xMn_2Si_2$ system as a result of an increasing of the Sm magnetic ions concentration. Generally, influence of the external pressure on the magnetic state of $La_{0.75}Sm_{0.25}Mn_2Si_2$ is equivalent to the "chemical" pressure. Decreasing of Mn-Mn interatomic distances in both cases leads to the stabilization of antiferromagnetic interlayer coupling between Mn magnetic moment projections on the **c** axis, decreasing T_C and increasing T_N temperatures. The applied pressure strongly affected the interlayer Mn-Mn exchange interaction and changes its sign for some temperatures.

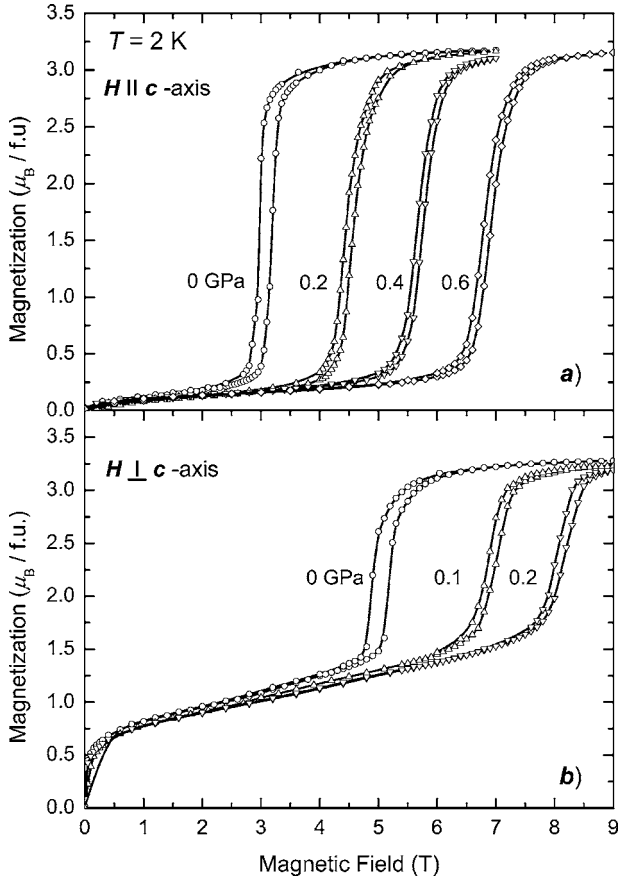

FIG. 5. Magnetization curves of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis (a) and in the basal plane (b) at $T=2$ K for different pressures.

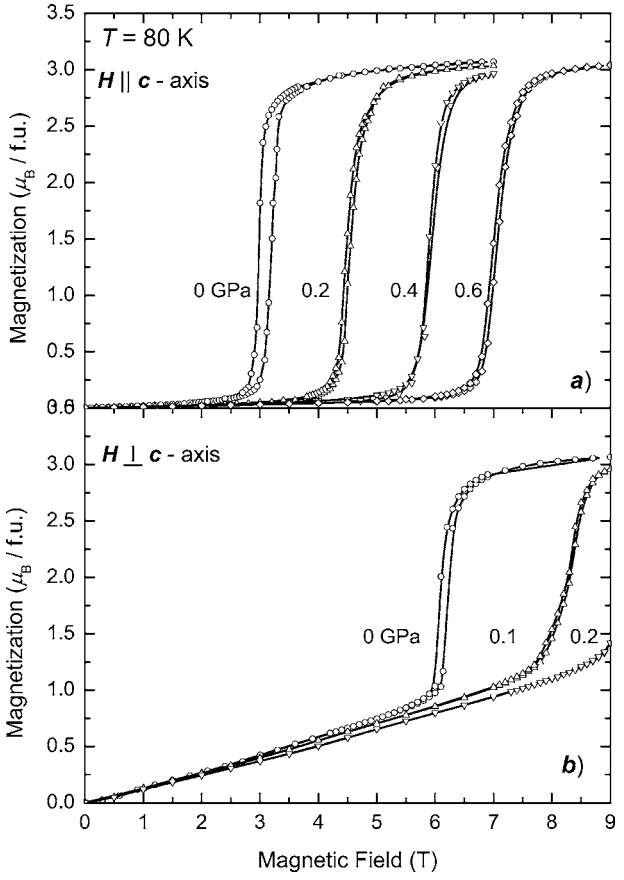

FIG. 6. Magnetization curves of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis (a) and in the basal plane (b) at $T=80$ K for different pressures.

B. Influence of pressure on field-induced magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound

Figures 5(a) and 5(b) show the magnetization curves of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured parallel and perpendicular to the **c** axis in the antiferromagnetic AF′ state at $T=2$ K for different pressures. At critical magnetic fields $H_{\parallel}$, $H_{\perp}$, the magnetization curves of the compound in both directions exhibit a jump corresponding to the field-induced AF′-F magnetic phase transition. A small hysteresis around the transition testifies that the AF′-F field-induced transition is of a first-order type. When the magnetic field is applied in the basal plane, a weak ferromagnetic component appears in the small field. It cannot be attributed to a Sm magnetic sublattice moment only, which is estimated to be $\sim 0.1\ \mu_B$. Perhaps, because of a Sm-Mn exchange interaction a ferromagnetic component of a Mn magnetic moment occurs in the plane, which corroborates with neutron diffraction data.[10] The value of the Mn saturation magnetic moment in the field-induced F state $\mu_{Mn} \approx 1.6\ \mu_B/Mn$ is close to the value of the z component of Mn magnetic moments $\mu_z \approx 1.88\ \mu_B/Mn$ determined for AF′ state at $T=4.2$ K by neutron diffraction.[10] Thus, at AF′-F field-induced phase transition there appears a change of the interlayer magnetic Mn-Mn ordering without a substantial change of the canting angle of the intralayer angular ferromagnetic structure. The value of the spontaneous ferromagnetic component in basal plane $\mu_F \approx 0.3\ \mu_B/Mn$ is also in very good agreement with the y component of Mn magnetic moments $\mu_y \approx 0.4\ \mu_B/Mn$ determined from neutron diffraction experiments.[10] The critical field of the AF′-F transition increases with increasing pressure, which indicates a promotion of the antiferromagnetic exchange interaction between layers.

Figures 6(a) and 6(b) show the magnetization curves of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis and in the basal plane at $T=80$ K in the antiferromagnetic AF state. Similarly to the AF′ state, the distinct magnetization jumps appear on magnetization curves, corresponding to the field-induced AF-F phase transition. The transition shifts to higher fields with increasing pressure. The value of the Mn saturation magnetic moment in the field-induced F state $\mu_{Mn} \approx 1.5\ \mu_B/Mn$ is close enough to the value of the z component of the Mn magnetic moment on the **c** axis $\mu_z \approx 1.76\ \mu_B/Mn$ determined for $T=78$ K in the AF state using neutron diffraction experiments.[10]

In the ferromagnetic F state at ambient pressure the magnetization curves indicate a considerable uniaxial magnetic anisotropy with the easy axis directed along the **c** axis of the crystal. At $T=250$ K the compound in zero field is a ferromagnet, but the pressure-induced antiferromagnetic state appears at $P=0.4$ GPa (Fig. 7). The applied magnetic field in-

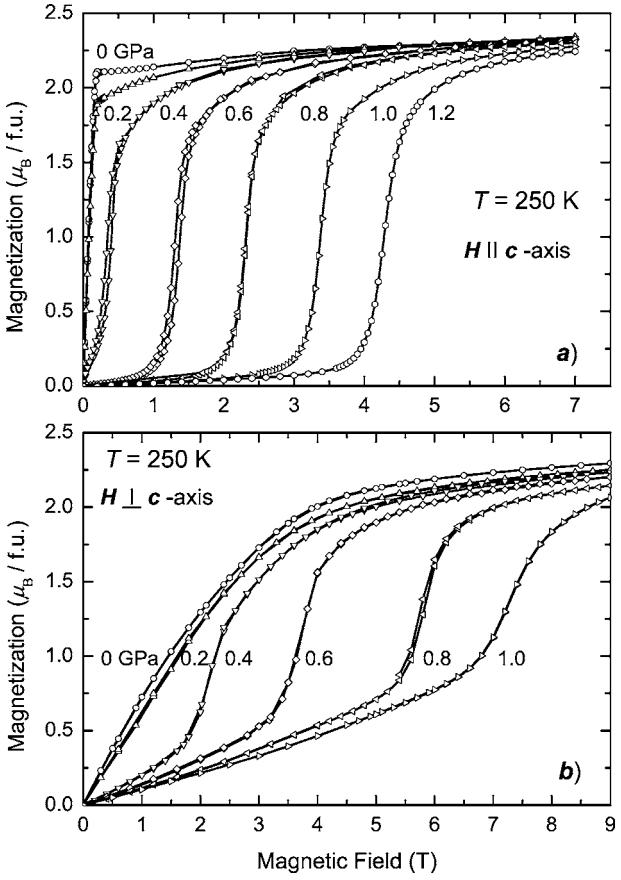

FIG. 7. Magnetization curves of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured along the **c** axis (a) and in the basal plane (b) at $T=250$ K for different pressures.

duces the reentrant AF-F magnetic phase transition. The critical field of the transition increases with increasing external pressure.

Temperature dependences of both the critical fields of metamagnetic transition and the anisotropy field H_A of $La_{0.75}Sm_{0.25}Mn_2Si_2$ are shown in Fig. 8. For comparison, the dashed line in Fig. 8 shows temperature dependence of the anisotropy field of $LaMn_2Si_2$.[15] It is seen that in the temperature range of ferromagnetic ordering of $La_{0.75}Sm_{0.25}Mn_2Si_2$, the anisotropy fields of the two compounds practically coincide. By our opinion, there are no reasons to believe that the anisotropy field of $La_{0.75}Sm_{0.25}Mn_2Si_2$ differs considerably from that of $LaMn_2Si_2$ at low temperatures where the F-AF transition does not allow us to determine the value of H_A.

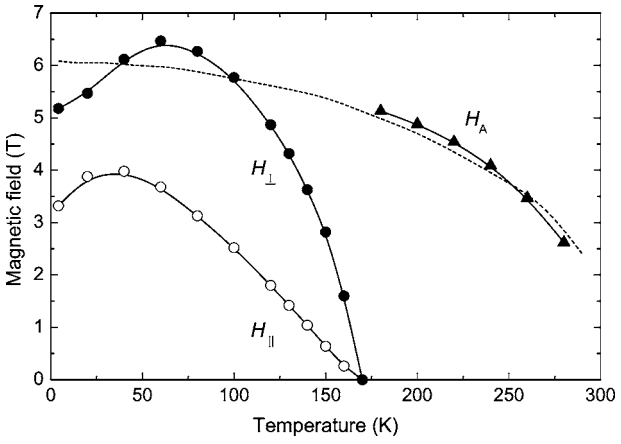

FIG. 8. Temperature dependences of the critical fields of metamagnetic transition $H_{\|}$, $H_{\perp}$ and magnetic anisotropy field H_A for $La_{0.75}Sm_{0.25}Mn_2Si_2$. The dashed line shows the temperature dependence of magnetic anisotropy field for $LaMn_2Si_2$ taken from Ref. 11.

Thus, a characteristic feature of the field-induced AF-F and AF′-F phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ is their appearance when the critical magnetic field is applied both along ($H_{\|}$) and perpendicular ($H_{\perp}$) to the easy **c** axis of crystal, although the values of $H_{\|}$ and $H_{\perp}$ are different. These values are comparable or lower than the anisotropy field of the compound. External pressure promotes the antiferromagnetic interlayer Mn-Mn exchange interaction and leads to the increase of critical fields of the metamagnetic transitions.

IV. DISCUSSION

In order to explain the observed unusual magnetic properties of $La_{0.75}Sm_{0.25}Mn_2Si_2$ we have to consider at least three different exchange interactions in the Mn sublattice of RMn_2X_2 compounds. The competition of intralayer antiferro- and ferromagnetic Mn-Mn exchange interactions causes the formation of canted ferromagnetic structure inside each layer and determines the critical temperature of transition in the paramagnetic state T_P, the Néel T_N, and Curie T_C temperatures. The interlayer Mn-Mn exchange interaction determines the type of interlayer magnetic Mn-Mn ordering and the temperature (critical field, pressure) of AF-F transition.

The present data of pressure effect on spontaneous magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ are in good agreement with the data obtained previously for $SmMnGe_2$-like compounds in which $d_{Mn\text{-}Mn} \approx d_c$ and where it was observed the change of sign of the interlayer Mn-Mn exchange interaction with increasing temperature.[16–21] In Table I we summarize the values of critical temperatures of magnetic phase transitions and their pressure derivatives for $La_{0.75}Sm_{0.25}Mn_2Si_2$, $La_{0.3}Y_{0.7}Mn_2Ge_2$, $SmMn_2Ge_2$, and $Nd_{0.35}La_{0.65}Mn_2Si_2$ compounds. For all these compounds the $d_{Mn\text{-}Mn}$ is very close to d_c and spontaneous AF-F magnetic phase transition is observed. As seen from Table I, the values of pressure derivative of critical temperatures of spontaneous magnetic phase transitions in these compounds are very close to each other. The strongest pressure derivative (or the dependence on the lattice parameters change) is observed for the temperature of AF-F phase transition.

Let us analyze the effect of pressure on spontaneous magnetic phase transitions in Mn sublattice of $La_{0.75}Sm_{0.25}Mn_2Si_2$ using the thermodynamic relations. For AF-P, F-AF″, and AF″-P second-order spontaneous phase transitions at T_N, T_C, and T_P, the Erenfest relation should be fulfilled

$$\frac{dT_{C,N,P}}{dP} = TV\left(\frac{\Delta\alpha_V}{\Delta C_P}\right), \tag{1}$$

where V is the volume, $\Delta\alpha_V$ is the difference between the values of a thermal expansion coefficient above and below

TABLE I. The critical temperatures of magnetic phase transitions and their pressure derivatives for different $R\mathrm{Mn}_2X_2$ compounds.

	$La_{0.75}Sm_{0.25}Mn_2Si_2$ (this work)		$La_{0.3}Y_{0.7}Mn_2Si_2$ (Ref. 20)		$SmMn_2Ge_2$ (Refs. 16–19)		$Nd_{0.35}La_{0.65}Mn_2Si_2$ (Ref. 21)	
	T (K)	dT/dP (K/GPa)	T (K)	dT/dP (K/GPa)	T (K)	dT/dP (K/GPa)	T (K)	dT/dP (K/GPa)
T_R	11	−0.6			100	−1.6	50	
$T_{F\text{-}AF}$	160	+203	180	+190	196	+171	220	+209
T_C	294	−12	345	−12	348	−11	295	−24
		($P<0.6$ GPa)		($P<0.82$ GPa)		($P<1$ GPa)		($P<0.4$ GPa)
T_N	286	+28	330	+57	337	+31	295	+31
		($P\geqslant 0.6$ GPa)		($P\geqslant 0.82$ GPa)		($P\geqslant 1$ GPa)		($P\geqslant 0.4$ GPa)
T_P	403	−60[a]			385	?	395	+20

[a]Estimated value (see text).

the critical temperature (T_C, T_N, T_P), ΔC_P is the value of a kink of specific heat at the phase transition. According to the available data on the change of the thermal expansion coefficient in $R\mathrm{Mn}_2X_2$ compounds at magnetic phase transitions, $\Delta\alpha_V<0$ at $T=T_C$ and $\Delta\alpha_V>0$ at $T=T_N$.[11] Therefore, from Eq. (1) it follows that $(dT_C/dP)<0$ and $(dT_N/dP)>0$, in agreement with experiment (see Table I).

Up to now little experimental information is available for the AF″ state and AF″-P phase transition in $R\mathrm{Mn}_2X_2$. Using the thermodynamic relation (1) and recent experimental data on change of thermal expansion coefficient of the $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound at T_P $\Delta\alpha_V=-7.5\times10^{-6}<0$,[11] it is possible to assume that $(dT_P/dP)<0$. Additionally, the expected decrease of critical temperature T_P with increasing pressure for $La_{0.75}Sm_{0.25}Mn_2Si_2$ is corroborated with decreasing of T_P observed for $La_{1-x}Sm_xMn_2Si_2$ when Sm concentration increases [Fig. 4(b)]. Alternatively, a positive change of T_P under pressure was observed in $Nd_{0.35}La_{0.65}Mn_2Si_2$.[21] However, this result may be caused by a wrong definition of T_P value from bulk magnetic measurements which are insensitive to AF″-P transition. This transition temperature in $R\mathrm{Mn}_2X_2$ compounds may be determined accurately by neutron diffraction and Mössbauer studies, the measurements of thermal expansion and heat capacity.[11–14] On the basis of the thermodynamic relation (1) we estimate the (dT_P/dP) value using the relations between the step changes of thermal expansion and the specific heat coefficients at $T_P(\Delta\alpha_V^P, \Delta C_P^P)$ and $T_C(\Delta\alpha_V^C, \Delta C_P^C)$

$$\frac{dT_P}{dP}\approx\frac{dT_C}{dP}\frac{T_P}{T_N}\frac{\Delta\alpha_V^P}{\Delta\alpha_V^C}\frac{\Delta C_P^C}{\Delta C_P^P}. \tag{2}$$

Using the values of $\Delta\alpha_V^P/\Delta\alpha_V^C\approx0.6$ and $\Delta C_P^C/\Delta C_P^P\approx6.3$ obtained for $SmMn_2Ge_2$[11,14] we estimate roughly the value $(dT_P)\approx5\times(dT_C/dP)=-60$ K/GPa. The dashed line with a corresponding slope on the P-T magnetic phase diagram [Fig. 4(a)] shows the expected variation of T_P under pressure for the $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound.

For the first-order AF-F magnetic phase transition the Clausius-Clapeyron thermodynamic relation should be valid

$$\frac{dT_{AF}}{dP}=-\frac{\Delta V}{\Delta M}\left(\frac{dT_{AF}}{dH}\right). \tag{3}$$

Using experimental values of $dT_{AF}/dP=203$ K/GPa, $\Delta M=15.6$ Am^2/mole, and $(dT_{AF}/dH)=25.5$ K/T, we estimate the value of $\Delta V=1.24\times10^{-7}$ m^3/mol. Therefore, the F-AF magnetic phase transition in $La_{0.75}Sm_{0.25}Mn_2Si_2$ should be accompanied by a significant volume change $\Delta V/V\approx2.3\times10^{-3}$.

The thermodynamic analysis of influence of pressure on spontaneous magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ shows that intralayer Mn-Mn exchange interactions are also sensitive to the critical distance between Mn atoms in $R\mathrm{Mn}_2X_2$ compounds. When the distance changes from $d_{\text{Mn-Mn}}>d_c$ to $d_{\text{Mn-Mn}}<d_c$ the sign of interlayer exchange interactions changes from positive to negative. Simultaneously, the pressure derivative for intralayer Mn-Mn exchange interaction changes the sign: The in-plane interaction increases with increasing pressure (or with decreasing of interatomic Mn-Mn distances) for $d_{\text{Mn-Mn}}<d_c$ and decreases for $d_{\text{Mn-Mn}}>d_c$ (Fig. 4, Table I). This may indicate the change of electronic band structure of the compounds.

It should be noted that the existence of the intermediate magnetically ordered AF″ state which separates the ferromagnetic and paramagnetic states is a characteristic feature of the compounds with $d_{\text{Mn-Mn}}>d_c$. For the compositions with $d_{\text{Mn-Mn}}<d_c$ no features of magnetically ordered phase were found above the region of stability of the AF phase, and the transition at T_N shall be considered as the order-disorder phase transition. However, it is unclear how the pressure (or concentration) dependence of T_P looks like in the vicinity of the critical distance d_c. We plan to study this region of the phase diagram in more details in the nearest future.

Usually, the metamagnetic transition in an antiferromagnet appears when the magnetic anisotropy is high in comparison with exchange interaction responsible for antiferromagnetic ordering. In this case the transition appears only for the field applied along the antiferromagnetic axis. For $La_{0.75}Sm_{0.25}Mn_2Si_2$ we observe first-order AF-F and AF′-F field-induced phase transitions when magnetic field is oriented both along and perpendicular to the **c** axis of the crystal. At the same time, the critical field values are lower than the magnetic anisotropy field. These features are characteristic of the itinerant electron antiferromagnets in which the first-order magnetic phase transition is caused by peculiari-

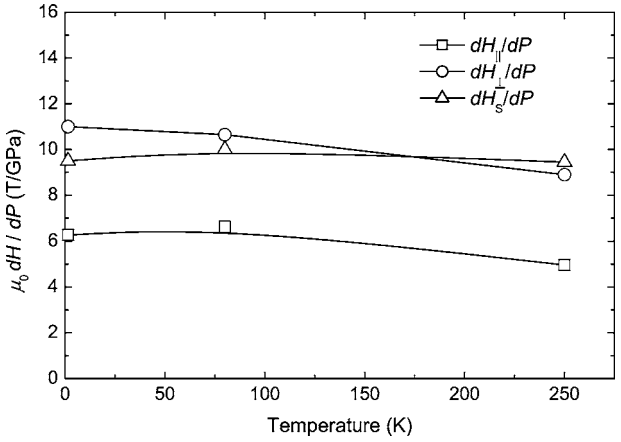

FIG. 9. Dependence of critical fields from pressure at various temperatures.

ties of electronic band structure. If we assume that at the point of phase transition there appears a step change of the interlayer Mn-Mn exchange interactions then for any mechanism of this change we can derive the following expression for critical fields of metamagnetic transitions:[9]

$$H_{\parallel}^{0} = \tfrac{1}{2}(H_{\text{Mn-Mn}}^{\text{AF}} + H_{\text{Mn-Mn}}^{\text{F}}) = H_{\text{Mn-Mn}}^{\text{AF}} + \tfrac{1}{2}\Delta H, \tag{4}$$

$$H_{\perp}^{0} = H_{\text{S}} - \sqrt{H_{\text{S}}^{2} - H_{\text{S}}(H_{\text{A}} + 2H_{\parallel}^{0})} = H_{\text{S}} - \sqrt{-H_{\text{S}}\Delta H}, \tag{5}$$

where $H_{\text{S}} = H_{\text{A}} + 2H_{\text{Mn-Mn}}^{\text{AF}}$ is the saturation field and ΔH is the change of the field of Mn-Mn interlayer exchange interaction at the transition from antiferromagnetic to ferromagnetic interlayer ordering. It is evident from Eq. (5) that ΔH is always negative. The value of $H_{\text{Mn-Mn}}^{\text{AF}}$ is accepted to be positive in the model, while $H_{\text{Mn-Mn}}^{\text{F}}$ may be positive or negative.[9] For a limiting case $\Delta H = 0$ Eqs. (4) and (5) describe the traditional metamagnetic transition, when the magnetization jump appears only for the field applied along the antiferromagnetic axis. For the nonzero ΔH value the metamagnetic transition appears in both directions. Finally, the ferromagnetic ground state stabilizes when $H_{\text{Mn-Mn}}^{\text{AF}} + H_{\text{Mn-Mn}}^{\text{F}} \leqslant 0$.

By taking into account the influence of a molecular exchange field of Sm sublattice H_{Sm}, Eqs. (4) and (5) may be rewritten as follows:

$$H_{\parallel} \approx H_{\parallel}^{0} - \frac{H_{\text{Sm}}^{2}}{H_{\text{S}}^{2}}\left(H_{\parallel}^{0} + \tfrac{1}{2}H_{\text{A}}\right), \tag{6}$$

$$H_{\perp} = H_{\perp}^{0} - H_{\text{Sm}}. \tag{7}$$

Differentiating Eqs. (6) and (7) by pressure and assuming H_{Sm} and H_{A} values to be pressure-independent we obtain the following approximate relations:

$$\frac{dH_{\parallel}}{dP} \approx \frac{dH_{\parallel}^{0}}{dP} \approx \frac{dH_{\text{Mn-Mn}}^{\text{AF}}}{dP} + \frac{1}{2}\frac{d\Delta H}{dP} \approx \frac{dH_{\text{Mn-Mn}}^{\text{AF}}}{dP}, \tag{8}$$

$$\frac{dH_{\perp}}{dP} \approx \frac{dH_{\perp}^{0}}{dP} \approx \frac{dH_{\text{S}}}{dP} - \frac{d\sqrt{-H_{\text{S}}\Delta H}}{dP} \approx 2\frac{dH_{\text{Mn-Mn}}^{\text{AF}}}{dP}. \tag{9}$$

Therefore, the growth of the critical field of metamagnetic transition with pressure is determined mainly by pressure dependence of the molecular field of Mn-Mn interlayer exchange interaction. Additionally, the pressure derivative of the critical field in the basal plane is expected to be twice of that along the **c** axis. In Fig. 9 we show the values of pressure derivatives of the critical fields of $La_{0.75}Sm_{0.25}Mn_2Si_2$ measured at different temperatures. It is seen that the experimental results are in good agreement with the above model.

It should be noted that in spite of a good qualitative description of the AF-F phase transition in the framework of localized magnetic moment models[6–9] in the assumption of a strong dependence of interlayer Mn-Mn exchange interaction on interatomic distances, the origin of this transition in RMn_2X_2 remains unclear. It is difficult to understand within these models why the interlayer Mn-Mn exchange interactions are sensitive mainly to the in-plane Mn-Mn distances[20] and what is the reason of appearance of the critical distance d_c between Mn atoms. Moreover, the AF-F magnetic phase transition possesses the features characteristic of itinerant electron antiferromagnets: (i) The transition is accompanied by a considerable volume change; (ii) the field-induced transition is observed both parallel and perpendicular to the antiferromagnetic axis; and (iii) alongside with the change of sign of interlayer Mn-Mn exchange interaction we observe the change of sign of pressure derivative of intralayer Mn-Mn exchange interactions. Band structure calculations for RMn_2X_2 compounds reveal a correlation between density of states at the Fermi level and the type of interlayer Mn-Mn magnetic ordering.[5] According to the analysis in the Hubbard model for the compounds with layered crystal structure,[4] the sign of this interaction may depend on density of states at Fermi level, while the value of the Mn magnetic moment may remain unchanged at the transition. For a deeper understanding of the nature of the AF-F transition, the studies of AF″ magnetic state and dependence of electron structure of RMn_2X_2 compounds on interatomic distances are strongly desirable.

V. CONCLUSION

We have studied the magnetization under pressure for a quasisingle crystal $La_{0.75}Sm_{0.25}Mn_2Si_2$ for which the in-plane Mn-Mn distance is very close to the critical distance of ferro-antiferromagnetic transition. The effect of external hydrostatic pressure on different spontaneous magnetic phase transitions in the $La_{0.75}Sm_{0.25}Mn_2Si_2$ compound is found to be very similar to that of "chemical" pressure in the $La_{1-x}Sm_{0.25}Mn_2Si_2$ system in which lattice parameters gradually decrease with increasing x. Similar to other RMn_2X_2 compounds in which Mn-Mn distances are close to the d_c critical value, pressure exhibits the strongest influence on the temperature of the antiferromagnetic-ferromagnetic first-order phase transition, which confirms a strong dependence of the interlayer Mn-Mn exchange interaction on Mn-Mn distance.

First-order field-induced metamagnetic transition in the compound has the features that are characteristic for itinerant antiferromagnets. It appears when the field is applied both

parallel and perpendicular to the antiferromagnetic axis and is accompanied by a considerable volume change. Additionally, the thermodynamic analysis of the obtained results allows us to conclude that with a decreasing of interatomic Mn-Mn distances the intralayer Mn-Mn exchange interaction increases for $d_{\text{Mn-Mn}} < d_c$ and decreases for $d_{\text{Mn-Mn}} > d_c$.

The influence of pressure on the field-induced magnetic phase transitions in $La_{0.75}Sm_{0.25}Mn_2Si_2$ and other RMn_2Si_2 compounds can be described within a phenomenological model based on the assumption of jump changes of the interlayer Mn-Mn exchange interaction at the transition. The model predicts the metamagnetic transition in both directions of a single crystal and anisotropic behavior of the pressure derivative of critical field.

*Electronic address: gerasimov@imp.uran.ru

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