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Magnetic phase transition and the corresponding magnetostriction of intermetallic compounds $R\mathrm{Mn_2Ge_2}$ (R=Sm, Gd)*

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The temperature dependence of lattice constants a and c of intermetallic compounds $R\mathrm{Mn_2Ge_2}$ (R=Sm, Gd) is measured in the temperature range 10–800K by using the x-ray diffraction method. The magnetoelastic anomalies of lattice constants are found at the different kinds of spontaneous magnetic transitions. The transversal and longitudinal magnetostrictions of polycrystalline samples are measured in the pulse magnetic field up to 25T. In the external magnetic field there occurs a first-order field-induced antiferromagnetism–ferromagnetism transition in the Mn sublattice, which gives rise to a large magnetostriction. The magnitude of magnetostrictions is as large as 10^{-3}. The transversal and longitudinal magnetostrictions have the same sign and are almost equal. This indicates that the magnetostriction is isotropic and mainly caused by the interlayer Mn–Mn exchange interaction. The experimental results are explained in the framework of a two-sublattice ferrimagnet with the negative exchange interaction in one of the sublattices by taking into account the lattice constant dependence of interlayer Mn–Mn exchange interaction.

Keywords: rare-earth intermetallics, magnetic phase transition, magnetostriction
PACC: 7530K, 7580

1. Introduction

In recent years, the ternary intermetallic compounds $R\mathrm{Mn_2Ge_2}$, where R is the rare-earth element or Y, have been extensively studied due to their unusual magnetic properties. $R\mathrm{Mn_2Ge_2}$ crystallizes in a body-centred-tetragonal $\mathrm{ThCr_2Si_2}$-type structure (space group $I4/mmm$), which consists of R, Ge and Mn layers, stacked in the sequence of R–Ge–Mn–Ge–R- along the c-axis.[1] From the magnetic point of view, $R\mathrm{Mn_2Ge_2}$ compounds have two different magnetic subsystems: R and Mn subsystems, and their magnetic properties are mainly determined by these two subsystems and their interactions. In $R\mathrm{Mn_2Ge_2}$ there are different kinds of exchange interactions. Many studies[2,3] have shown that the interlayer Mn–Mn exchange interaction is very sensitive to the intralayer Mn–Mn distance and, consequently the lattice constant a. When a is larger than a critical value a_k, the interlayer Mn–Mn exchange interaction is ferromagnetic, otherwise it is antiferromagnetic. For this reason, the Mn sublattice in most light rare-earth compounds has a ferromagnetic structure, whereas the intrinsic magnetic structure of the Mn sublattice in compounds with heavy rare earths is antiferromagnetic.

In $\mathrm{SmMn_2Ge_2}$, the lattice constant a is very close to the critical value a_k at room temperature. As a result, with the decrease of temperature to a certain value, the interlayer Mn–Mn couple changes its sign due to thermal contraction, giving rise to the first-order magnetic transition of the Mn sublattice from a ferromagnetic to an antiferromagnetic state and accompanying the abrupt decrease of lattice constant a. When the temperature continues to decrease to T_2, the Mn sublattice reenters the ferromagnetic state because of the Sm–Mn exchange interaction.[3–5] In the heavy rare-earth compounds (R=Gd, Tb, Dy), the R–Mn exchange interaction is antiferromagnetic and its value has the same order as the interlayer Mn–Mn exchange interaction. With the reduction of temperature, the Mn sublattice undergoes a first-order magnetic transition from an antiferromagnetic to a ferromagnetic state as a competition result of R–Mn and interlayer

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Mn–Mn exchange interaction. For $GdMn_2Ge_2$ this transition occurs at 96K.[6]

When the interlayer Mn–Mn exchange interaction is antiferromagnetic, the magnetic structure of the Mn sublattice may change under the action of an external magnetic field. This kind of field-induced magnetic transition has been found in $SmMn_2Ge_2$,[3,4] $GdMn_2Ge_2$ [7,8] and $DyMn_2Ge_2$.[9,10]

In this paper we study the magnetoelastic properties of $SmMn_2Ge_2$ and $GdMn_2Ge_2$ compounds caused by the spontaneous and field-induced magnetic transitions in the Mn sublattice by measuring the temperature dependence of lattice constants and transversal and longitudinal magnetostrictions.

2. Experimental details

The polycrystalline samples of $SmMn_2Ge_2$ and $GdMn_2Ge_2$ were prepared by induction melting of the constituent elements in a water-cooled crucible under argon atmosphere. Melting was repeated several times in order to ensure homogeneity. Then the ingots were sealed in a quartz tube for further annealing at 750°C for one week. The quality of the samples was verified by x-ray diffraction (XRD), and the samples were found to be approximately single phase.

The temperature dependence of the lattice constants of $SmMn_2Ge_2$ and $GdMn_2Ge_2$ was measured in the temperature range of 10–800K using the XRD method which was carried out on a Geigerflex diffractometer with Fe$K\alpha$ radiation. The transversal and longitudinal magnetostrictions were measured by the induction method in a pulse magnetic field up to 25T in the temperature range of 10–300K. In order to determine the accurate temperature of spontaneous magnetic transition, we also measured the ac magnetic susceptibility in a low field (up to 10^{-3}T).

3. Results and discussion

$SmMn_2Ge_2$: According to many magnetic measurements,[3,4] the Mn sublattice of $SmMn_2Ge_2$ orders ferromagnetically at T_C=341K. When the temperature decreases to T_{t1}=150K, the Mn sublattice transforms to an antiferromagnetic state with a first-order transition due to thermal contraction, which causes the interlayer Mn–Mn exchange interaction to change its sign. As the temperature continues to reduce, the Mn sublattice reenters the ferromagnetic state at T_{t2}=100K. At the same time, the Sm sublattice becomes ferromagnetically ordered and ferromagnetically coupled with the Mn sublattice.

Figure 1 shows the temperature dependence of lattice constants a and c of $SmMn_2Ge_2$, measured using the XRD method. It can be seen from Fig.1 that, at T_{t1}, when the first-order ferromagnetism–antiferromagnetism transition occurs in the Mn sublattice, the lattice constant a decreases abruptly, while at T_{t2} (when the Mn sublattice recovers from the antiferromagnetic state to the ferromagnetic state) a increases abruptly. At these transition points the lattice constant c almost does not change. These results are in agreement with those in Refs.[3] and [4], confirming that the interlayer Mn–Mn exchange interaction is sensitive to the lattice constant a. The change of the lattice constant $\frac{\Delta a}{a}$ is about 0.15%. From Fig.1 one can see that, above the Curie temperature T_C=341K, there is a negative magnetoelastic anomaly on curve $c(T)$ at T_N ≈380K. Recently, neutron diffraction and Mössbauer studies[2,11,12] have shown that, for light rare-earth compounds, the Curie temperature T_C is not the transition temperature of paramagnetism–ferromagnetism; above T_C, the compounds have some kind of antiferromagnetic structure, and at a higher temperature T_N (T_N ≈380K for $SmMn_2Ge_2$) the compounds become magnetically disordered.[12] Our studies confirm the existence of a high-temperature antiferromagnetic phase. The $c(T)$ curve indicates that the transition from a high-temperature antiferromagnetic phase to a paramagnetic phase is connected with the lattice constant c.

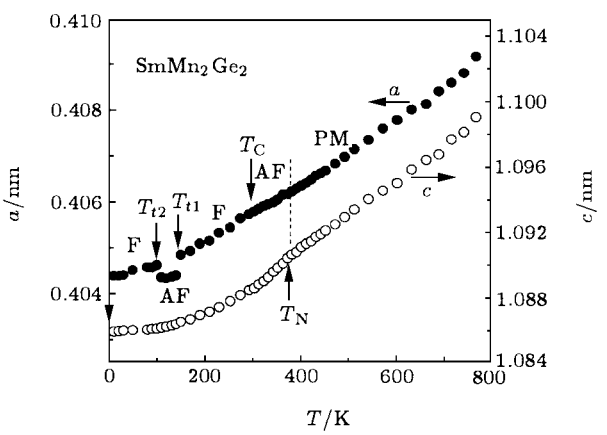


Fig.1. The temperature dependence of lattice constants a and c of the intermetallic compound $SmMn_2Ge_2$.

Figure 2 illustrates the transversal and longitudinal magnetostrictions of $SmMn_2Ge_2$. It can be seen that, in the temperature range $100 < T < 150$K and at a critical magnetic field, the transversal and longitudinal magnetostrictions suddenly increase, but out

of this temperature range the magnetostrictions are small and almost do not change. As mentioned above, for $100 < T < 150$K, the Mn sublattice has an antiferromagnetic structure and Sm is magnetically disordered. According to Refs.[3] and [4], under the external magnetic field, the Mn subsystem transforms from an antiferromagnetic to a ferromagnetic state with a first-order transition. Apparently, the abrupt increase of magnetostriction is related to this field-induced transition. It can also be seen from Fig.2 that the magnetostriction caused by this antiferromagnetism–ferromagnetism transition is isotropic ($\alpha_{\parallel} \approx \lambda_{\perp}$). The temperature dependence of saturation magnetostriction is presented in Fig.3. From the $a(T)$ and $c(T)$ curves (Fig.1) one can see that the first-order spontaneous antiferromagnetism–ferromagnetism transition is accompanied by the abrupt increase of lattice constant a, while c is almost not changed. This suggests that the magnetostriction caused by the field-induced antiferromagnetism–ferromagnetism is mainly caused by the lattice constant a. As the magnetic anisotropy in $100 < T < 150$K is small (the Sm subsystem is magnetically disordered in this temperature range), the transition from an antiferromagnetic to a ferromagnetic state occurs in all field directions with respect to the crystalline axis.[2] Under this condition the transversal and longitudinal magnetostrictions are equal and can be expressed as

$$\lambda_{\parallel} = \lambda_{\perp} \approx 2\Delta a/3a. \quad (1)$$

Here $\Delta a/a$ is the lattice constant change caused by the temperature-induced antiferromagnetism–ferromagnetism transition ($\Delta a/a \approx 0.15\%$). From Eq.(1) we can obtain $\lambda_{\parallel}$, $\lambda_{\perp} = 1.0 \times 10^{-3}$, which is in agreement with the experimental data. A small difference of transversal and longitudinal magnetostrictions may be caused by the textural structure of polycrystalline samples. The influence of the textural structure on the crystalline structure and magnetic properties of intermetallic compounds $R\mathrm{Mn_2Ge_2}$ has been reported in Ref.[13].

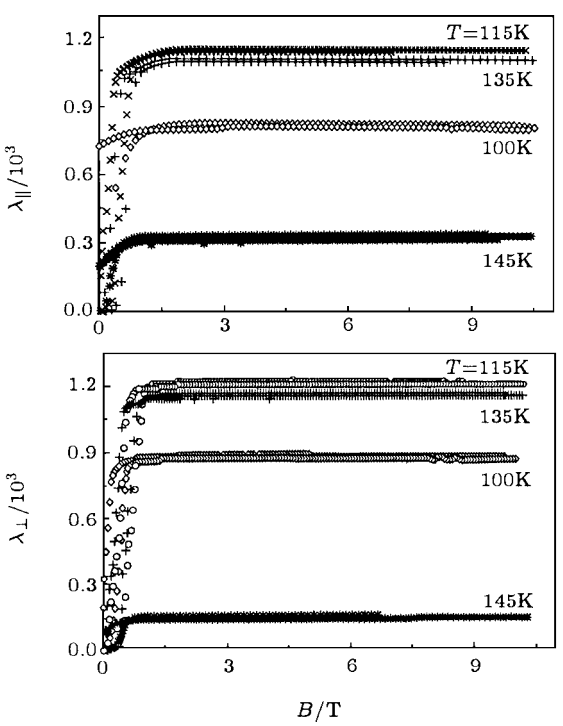


Fig.2. The transversal and longitudinal magnetostrictions of polycrystalline sample $SmMn_2Ge_2$.

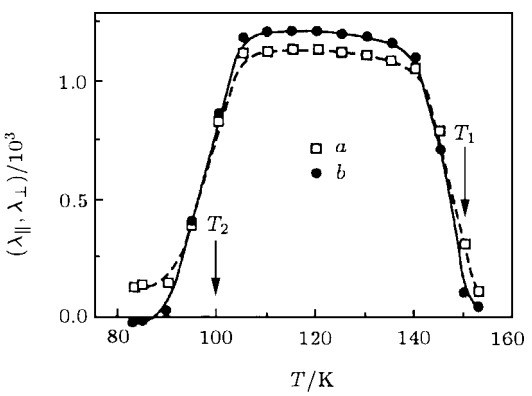


Fig.3. The temperature dependence of longitudinal (a) and transversal (b) saturation magnetostrictions of $SmMn_2Ge_2$. Arrows indicate the temperatures of the magnetic phase transition.

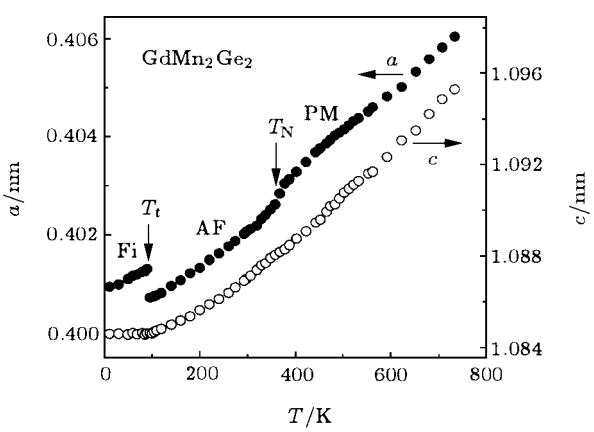


Fig.4. The temperature dependence of lattice constants a and c of intermetallic compound $GdMn_2Ge_2$.

$GdMn_2Ge_2$: According to Ref.[6], at $T_N = 365K$, $GdMn_2Ge_2$ is antiferromagnetically ordered. As temperature decreases to T_t=96K, the Mn sublattice transforms from an antiferromagnetic to a ferromagnetic state with a first-order transition. At the same time the Gd subsystem becomes ferromagnetically

ordered and antiferromagnetically coupled with the Mn subsystem. Figure 4 presents the temperature dependence of lattice constants a and c. It can be seen from Fig.4 that at T_N the paramagnetism–antiferromagnetism transition in the Mn sublattice is accompanied by a negative magnetoelastic anomaly $a(T)$. At T_t the first-order antiferromagnetism–ferromagnetism transition in the Mn sublattice is accompanied by the abrupt increase of a, $\Delta a/a \approx 10^{-3}$, which is almost the same as that observed in $SmMn_2Ge_2$.

The transversal and longitudinal magnetostrictions of $GdMn_2Ge_2$ are displayed in Fig.5. It can be seen from Fig.5 that, when the magnetic field arrives at a critical value, the transversal and longitudinal magnetostrictions suddenly increase. Above T_t=96K, the transversal and longitudinal magnetostrictions are all positive, while below this temperature they are negative. As mentioned above, when $T > T_t$, the Mn sublattice has an antiferromagnetic structure. According to the single crystalline $GdMn_2Ge_2$ studies,[7] when an external magnetic field is applied along the c-axis, a first-order magnetic phase transition occurs in the Mn sublattice from an antiferromagnetic to a ferromagnetic state. Below T_t, $GdMn_2Ge_2$ has a ferrimagnetic structure. When the external magnetic field is applied perpendicular to the c-axis, the compound transforms from linear ferrimagnetic to the nonlinear triangular magnetic structure, in which the Gd magnetic moment is along the field direction, and the two Mn magnetic moments are split apart from each other and form an angle with Gd moment.[7,8] It is obvious that the observed sudden changes of transversal and longitudinal magnetostrictions above and below T_t are caused by these field-induced first-order magnetic transitions. The negative transversal and longitudinal magnetostrictions below T_t are due to the fact that the magnetic field destroys the ferromagnetic structure of the Mn sublattice. In contrast, above T_t the magnetic field makes the Mn subsystem transform from the antiferromagnetic state to the ferromagnetic state; this gives rise to a positive magnetostriction. (In the temperature range of 96 < T <240K, we did not obtain the saturation magnetostriction, since in this temperature range the critical field of the antiferromagnetism–ferromagnetism transition is larger than the maximum field we can have.) That the transversal and longitudinal magnetostrictions have the same sign and are almost equal indicates that the magnetostriction is isotropic and mainly caused by the exchange interaction. The small difference of the transversal and longitudinal magnetostrictions may come from the textural structure as in the case of $SmMn_2Ge_2$. Comparing Figs.4 and 5 one can see that the magnitude of the saturation magnetostriction of $GdMn_2Ge_2$ has the same order of magnitude as the lattice constant change $\Delta a/a$ caused by the temperature-induced first-order ferromagnetism–antiferromagnetism transition. This also suggests that the magnetostriction is mainly caused by the magnetic structure change of the Mn sublattice.

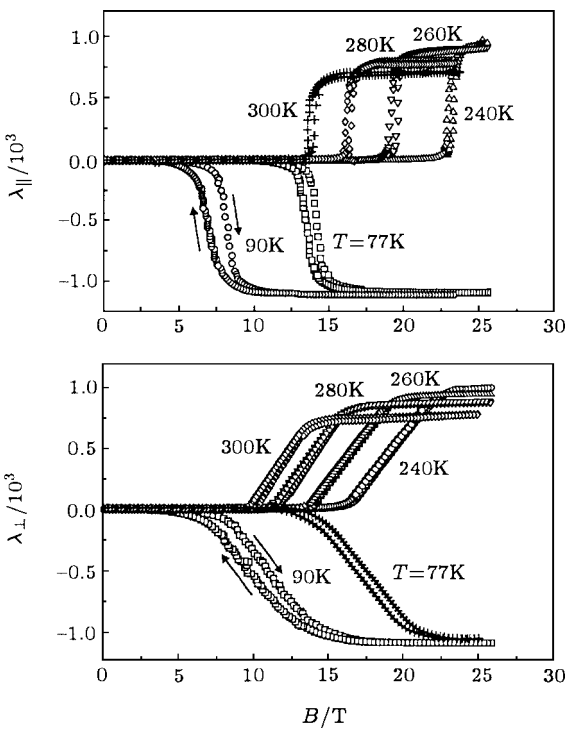


Fig.5. The transversal and longitudinal magnetostrictions of the polycrystalline sample $GdMn_2Ge_2$.

4. Conclusion

The temperature dependence of lattice constants a and c measured by the XRD method shows that the different kinds of spontaneous magnetic transitions in $SmMn_2Ge_2$ and $GdMn_2Ge_2$ are accompanied by the magnetoelastic anomalies of a and c. The maximum magnetoelastic anomaly observed on curve $a(T)$ is caused by the first-order ferromagnetism–antiferromagnetism transition in the Mn sublattice, and $\Delta a/a \approx 10^{-3}$. Under an external magnetic field, there occurs the first-order field-induced antiferromagnetism–ferromagnetism transition in the Mn sublattice, which gives rise to a large magnetostriction. That the transversal and longitudinal magnetostrictions have the same sign and are almost

equal to each other indicates that the magnetostriction is isotropic and is mainly caused by the interlayer Mn–Mn exchange interaction. The saturation magnetostriction has the same order of magnitude as the $\Delta a/a$ aroused by the temperature-induced first-order antiferromagnetism–ferromagnetism transition in the Mn sublattice. This suggests that the large magnetostriction is mainly due to the change of lattice constant a caused by the change in the magnetic structure of the Mn sublattice.

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