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Magnetoelastic properties of substituted $Er_{1-x}Gd_xMn_6Sn_6$ intermetallic system

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ABSTRACT

The forced magnetostriction of polycrystalline samples of $Er_{1-x}Gd_xMn_6Sn_6$ ($0 \le x \le 1$) intermetallics with hexagonal $HfFe_6Ge_6$-type structure is investigated in the temperature range of 77–480 K. Gd substitution has a significant effect on interatomic distances and especially on inter-sublattice R–Mn couplings. The replacement of Er by Gd results in increasing the ordering temperature followed by reinforcement of the R–Mn coupling, as well as decreasing the magnetostriction values owing to the S-state character of Gd^{3+} ions. The results show that the contribution of Er sublattice to anisotropic magnetoelastic effects is positive, while that of Gd and Mn is negative. All the examined samples exhibit considerable magnetovolume anomalies at the ordering temperature (T_C=338, 381, 412 and 434 K for the samples with x=0, 0.2, 0.6 and 1.0, respectively). While the unsubstituted sample exhibits metamagnetic transitions, Gd-contained compounds do not show this behavior, owing to the strong Gd–Mn coupling. The experimental results obtained are discussed in the framework of the two-magnetic sublattice by bearing in mind the lattice parameter dependence of the interlayer Mn–Mn exchange interaction in these layered compounds. From the temperature dependence of magnetostriction values and considering the magnetostriction equation for a hexagonal structure, we attempt to determine the signs of some of the magnetostriction constants for these compounds and the influence of Gd substitution on them.

1. Introduction

RMn_6Sn_6 ternary intermetallic compounds with R=Sc, Y and rare-earth elements (except for R=Pm and Eu whose compounds have never been prepared) have attracted considerable attention in recent years, owing to their interesting magnetic properties. They have been extensively studied using a wide variety of methods including magnetization measurements [1–3], neutron diffraction [4], Mössbauer spectroscopy [4], NMR spectroscopy [5], transport, magnetotransport [6] and magneto-optical measurements [7], and also by theoretical studies on their electronic structure [8]. All these compounds crystallize in the hexagonal $HfFe_6Ge_6$-type structure with space group P6/mmm (Fig. 1). This crystal structure can be described as layers of R and Mn atoms alternately stacked along the *c*-axis in the sequence Mn–(R,Sn)–Mn–Sn–Sn–Sn–Mn. The magnetic structure of RMn_6Sn_6 compounds consists of two interacting subsystems: one of them is composed of R atoms whereas Mn atoms form the other. The observed complex magnetic behavior of these compounds with various magnetic phase transitions originate from the temperature-dependent competition between the Mn–Mn, R–Mn and R–R interactions, as well as from the magnetocrystalline anisotropies of the R and Mn sublattices. Both the intralayer Mn–Mn interaction (J_0) which is the strongest, and the interlayer Mn–Mn exchange interaction through the Mn–Sn–Sn–Sn–Mn slab (J_1) are always positive (ferromagnetic), while the nature of that within the Mn–(R,Sn)–Mn slab (J_2) depends on the Mn–Mn interatomic distances and so is very sensitive to the R element [9,10]. The R–Mn coupling is negative for heavy R elements and strongest for R≡Gd [11,12], with the same order of magnitude as the interlayer Mn–Mn. Among the RMn_6Sn_6 family, the compound with R≡Er has a complex behavior displaying several transitions: spontaneous (temperature-induced) transitions characterized by antiferromagnetism below T_N=352 K and a transition to ferrimagnetic state at about 75 K, as well as metamagnetic (field-induced) ones in its ordered state [1]. In order to better understand the contributions of the two sublattices to these magnetic behaviors, we decided to study the effect of Gd substitution for Er on structure and on certain magnetic and magnetoelastic properties of $Er_{1-x}Gd_xMn_6Sn_6$ ($0 \le x \le 1$) compounds. Since the Mn sublattice favors an easy plane anisotropy and Er and Gd both reveal an easy plane behavior in the whole ordered state as well [4], there will be no competition between the two sublattice anisotropies and consequently no spin reorientation process is expected in the compounds studied. As a result of the strong dependence of Mn–Mn interlayer

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interactions on interatomic distance, one may expect that these magnetic transitions involving variation of arrangement of Mn moments, are likely to be accompanied by anomalies in the magnetoelastic behaviors. Therefore, in the present work, we report the effect of Gd substitution for Er on the field-induced magnetovolume effects of $Er_{1-x}Gd_xMn_6Sn_6$ ($0 \le x \le 1$) compounds. The influence of this substitution on their structure, thermal expansion and spontaneous magnetostriction has been reported elsewhere [13].

2. Experiments

$Er_{1-x}Gd_xMn_6Sn_6$ ($0 \le x \le 1$) polycrystalline samples were prepared by arc melting of the constituent elements under high-purity argon atmosphere in a water-cooled copper hearth. The details of samples preparation have been described previously in Ref. [13]. The purity and microstructure of the prepared samples were examined using X-ray powder diffraction (XRD) with monochromatic Cu Kα radiation ($\lambda \sim 1.5406$ °) in the 2θ range of 20–90° in a continuous scan mode with a step width of 0.05° and using scanning electron microscopy (SEM) (Leo 1450VP, Carl Zeiss SMT, Germany). For structural characterization, analysis of the obtained XRD data was performed using the Rietveld refinement method, through the Fullprof software. The Linear thermal expansion (TE) normalized to 77 K ($\Delta l/l = (l_T - l_{77K})/l_{77K}$) and magnetostriction (MS) were measured using the strain-gage Wheatstone bridge technique on disk-shaped samples with a diameter of about 6 mm and thickness of about 2 mm in the temperature range of 77–520 K and magnetic fields up to 1.5 T. The accuracy of these measurements was better than 2×10^{-6}. The longitudinal ($\lambda_{\parallel}$) and transverse ($\lambda_{\perp}$) magnetostrictions of the samples were measured parallel and perpendicular to the applied magnetic field, respectively. The anisotropic magnetostriction $\Delta\lambda$ (λ_t) and volume magnetostriction $\Delta V/V$ (ω) were calculated directly from the relations $\lambda_t = \lambda_{\parallel} - \lambda_{\perp}$ and $\lambda_{\perp} = \lambda_{\parallel} + 2\lambda_{\perp}$. It should be noted that no significant difference was observed between the strains measured in the plane and perpendicular to the plane of the disc of the samples (at room temperature), suggesting the absence of any preferred orientation effects.

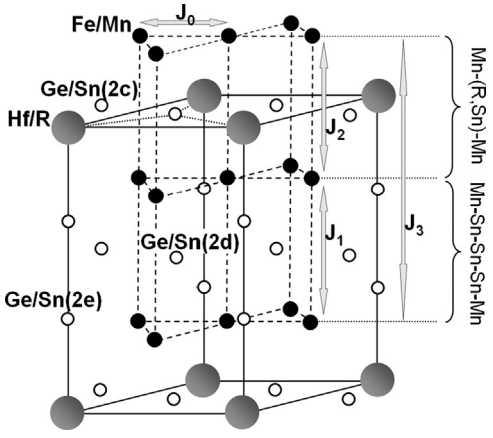

Fig. 1. Schematic representation of the $HfFe_6Ge_6$-type crystal structure and different Mn–Mn magnetic interactions in RMn_6Sn_6 compounds.

3. Results and discussion

As previously reported [13], the samples are pure single-phase with $HfFe_6Ge_6$-type structure (S.G. P6/mmm). The refined lattice parameters obtained from the Reitveld analysis of XRD patterns that are summarized in Table 1 show that the replacement of Er by Gd in $Er_{1-x}Gd_xMn_6Sn_6$ compounds causes the lattice constants to increase. This occurs due to the larger atomic radius of Gd compared with Er [14].

The zero-field linear thermal expansion dl/l of $Er_{1-x}Gd_xMn_6Sn_6$ samples in the temperature range of 77–520 K [13] has revealed considerable anomalies (notable volume expansions) at Néel-like transition points of the related compounds (T_N) and also at $T_M = 309$–311 K where the Mn moments experience collapse-like reduction [15], whereas trivial anomalies occurred at Curie-like transition points (T_C). Consequently, with regard to the magnetic arrangement of sublattices at the transition temperatures of the samples, it can be concluded that the spontaneous magnetovolume effects in these compounds originate mainly from the antiferromagnetic interlayer Mn–Mn exchange interactions, whereas the intralayer ferromagnetism does not influence these magnetovolume effects. The values of the transition temperatures of the studied samples estimated from the thermal expansion measurements are summarized in Table 1. As seen, the ordering temperature increases with increasing Gd content. This behavior has been discussed in detail in our previous paper [13].

The longitudinal $\lambda_{\parallel}$ and transverse $\lambda_{\perp}$ magnetostrictions of $Er_{1-x}Gd_xMn_6Sn_6$ samples were measured as a function of the applied magnetic field at certain typical temperatures. As representatives, the $\lambda_{\parallel}$ and $\lambda_{\perp}$ isotherms of the samples at two selected temperatures are presented in Fig. 2. For the $ErMn_6Sn_6$ sample, except for the region near to the antiferromagnetic–ferrimagnetic transition point (about 77 K), there is no significant difference between the MS measured parallel ($\lambda_{\parallel}$) and perpendicular ($\lambda_{\perp}$) to the applied magnetic field. This means that, apart from the low temperature region, MS is almost isotropic. As seen, Gd substitution causes the longitudinal and transverse MS to be opposite in sign (except for the region around the ordering temperature), and whereas for the sample with $x = 0.2$ there is no significant difference between their magnitudes, the difference grows with Gd content so that for the sample with $x = 1$, $|\lambda_{\parallel}| \approx 3|\lambda_{\perp}|$. Furthermore, the comparison of the results shows that the linear MS decreases with Gd substitution. This is reasonable since Gd^{3+} is an S state ion ($L = 0$) with a spherical symmetric 4f charge density. Consequently, loss of the interaction of the anisotropic electronic cloud of 4f electrons with the crystalline field which is the origin of MS effect in heavy rare earths makes the MS of Gd approximately two orders of magnitude smaller than the other rare earth

Table 1
Rietveld refined lattice parameters and magnetic transition temperatures of $Er_{1-x}Gd_xMn_6Sn_6$ samples obtained from zero-field thermal expansion, TE [13] and volume magnetostriction, MS, measurements under magnetic fields up to 1.5 T (Fi = ferrimagnetic, AF = antiferromagnetic).

Gd context (x)	a ( )	c ( )	V (^{3})	c/a	TE			MS	Magnetic order
					T_{C1} (K)	T_N (K)	T_C (K)	T_t (K)	
0	5.52743	9.02036	238.671	1.6319	≈ 77	340	–	338	Fi–AF
0.2	5.52835	9.01982	238.736	1.6316	164	335	383	381	Fi–AF–Fi
0.6	5.53266	9.02602	239.274	1.6314	–	–	419	412	Fi
1	5.54671	9.04348	240.956	1.6304	–	–	434	434	Fi