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Magnetism and magnetoresistance of Er_{1-x} $Sm_xMn_6Ge_6$ (x=0.2–1.0) compounds*

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Magnetic and transport properties of $Er_{1-x}Sm_xMn_6Ge_6$ (x=0.2-1.0) have been investigated by x-ray diffraction (XRD) and magnetization measurement. Analysis of the XRD patterns indicates that the samples with $x \leq 0.4$ mainly consist of $HfFe_6Ge_6$-type phase and the samples with $0.6 \leq x \leq 1.0$ mainly consist of YCo_6Ge_6-type phase (P6/mmm). The lattice constants and the unit cell volume increase with increasing Sm content. The antiferro–ferri–ferromagnetic transitions can be observed with increasing Sm content. The samples with x=0.2 and 0.4 order antiferromagnetically at 420 and 425K, respectively. The samples with x=0.6, 0.8 and 1.0 order from ferri- to ferromagnetically over the whole magnetic ordering temperature range. The corresponding Curie temperatures are 435, 441 and 446K, respectively. The magnetoresistance (MR) isotherms of the sample with x=0.8, measured at various temperatures, are analysed. The magnitude of MR is found to be positive below 55K and gradually increases to a relatively large value of about 5.02% at 5K in a field of 5T as the temperature is lowered. A possible explanation for the positive MR is given.

Keywords: magnetism, positive magnetoresistance
PACC: 7525,7530E, 7530V

1. Introduction

The crystallographic and magnetic properties of the RMn_6Ge_6 (R=Y, Nd, Sm, Gd-Lu) compounds have been studied extensively. They are isotypic either to $HfFe_6Ge_6$ (R=Y, Gd–Lu)[1] or to YCo_6Ge_6 (R=Nd, Sm).[2] The magnetic behaviour of this family of compounds is of particular interest due to the different strengths of the R–Mn interaction and to the magnetocrystalline anisotropy of the R atom.[3] Magnetic measurement and neutron diffraction studies have shown that the magnetic properties and magnetic structures of RMn_6Ge_6 are very sensitive to the Mn–Mn interatomic distance and the nature of the R elements. Among them, $ErMn_6Ge_6$ orders antiferromagnetically below T_N=475K and undergoes a first-order spin reorientation transition at T_t=80K. The sequences of phases found by neutron diffraction are: (a) a triple skewed spiral phase in the range 1.4–80K; the structural unit consists of Mn(+)-Er(-)-Mn(+) triple layers which change their moment direction collectively along the direction of the wavevector; (b) a complex distorted skewed spiral phase in the region 80–160K; and (c) a collinear antiferromagnetic phase in the region 160–200K.[3] Such a complicated magnetic structure suggests that the magnetic interactions within the slab are very sensitive to the nature of the R element and slight modifications of the chemical bond.

Intensive study of spin-dependent transport in magnetic multilayers has been stimulated by the observation that antiferromagnetically coupled layers can show a dramatic decrease in resistivity when the layers are forced into ferromagnetic alignment by the application of an external magnetic field. The recognized interpretation is that giant magnetoresistance (MR) is usually attributed to spin-dependent scattering inside the bulk or at the interface,[4] the overall

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resistivity being lower for parallel than for antiparallel arrangement of the magnetic moments. However, it has been reported that some of the layered compounds exhibit large positive MR behaviour, in spite of the fact that they are ferromagnetic at low temperatures.[5−10] In this paper, we report the effect of substitution of Sm for Er on the magnetic and transport properties of $Er_{1-x}Sm_xMn_6Ge_6$ ($0.2 \leq x \leq 1.0$) series and offer the possible explanations of the positive MR at low temperatures.

2. Experiment

$Er_{1-x}Sm_xMn_6Ge_6$ (x=0.2–1.0) polycrystalline samples were synthesized by arc-melting in a high-purity argon atmosphere. The raw materials used were of at least 99.9% purity. An excess of Er, Sm and Mn over the stoichiometric amount was added to compensate for the loss of weight during the arc-melting. The ingots were re-melted at least three times to ensure the homogeneity. They were subsequently sealed in an evacuated quartz tube and annealed at 1073K for ten days, and then quenched in water. The crystal structures of the samples were measured by XRD with CuK_α radiation. The magnetic properties were measured by a vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID). Specific heat measurements were performed by the semi-adiabatic heat pulse method in the temperature interval 5–150K in the presence of 0 or 5T fields. The MR measurement was carried out by means of the standard four-probe dc method with an electric current of 100 mA parallel to the magnetic field. The sample used in the MR measurement was a rod with square cross-section having dimensions of $10\times2\times2$ mm^3.

3. Results and discussion

Analysis of the XRD patterns shows that the samples with $x \leq 0.4$ mainly consist of $HfFe_6Ge_6$-type phase and the samples with $0.6 \leq x \leq 1.0$ mainly consist of YCo_6Ge_6-type phase (P6/mmm). The lattice constants of all compounds are listed in Table 1. As observed, the replacements of Er by Sm are characterized by the increase of lattice constants due to the larger atomic radius of Sm compared with Er. The increase of the lattice constants means lengthening of the Mn–Mn and R–Mn interatomic distances.

Table 1. Structural and magnetic data for $Er_{1-x}Sm_xMn_6Ge_6$.

x	a/nm	c/nm	c/a	V/nm^3	$T_{N,C}$/K	Structure type	M_S/(A·m^2·kg^{-1})
0.2	0.5224	0.8152	1.5605	0.1929	420	$HfFe_6Ge_6$	–
0.4	0.5228	0.8164	1.5616	0.1932	425	$HfFe_6Ge_6$	–
0.6	0.5237	0.8180	1.5620	0.1943	435	YCo_6Ge_6	33.1
0.8	0.5243	0.8188	1.5617	0.1949	441	YCo_6Ge_6	57.5
1.0	0.5245	0.8194	1.5622	0.1952	446	YCo_6Ge_6	67.5

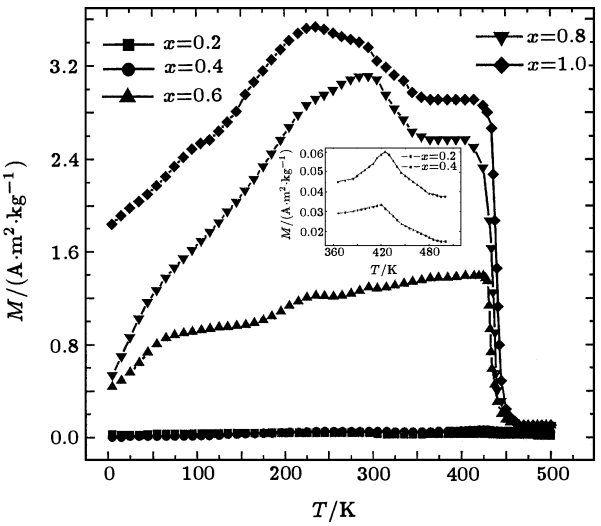


Fig.1. Temperature dependence of magnetization for $Er_{1-x}Sm_xMn_6Ge_6$ in a magnetic field of 0.01T (the inset shows the magnetization for the samples with x=0.2 and 0.4).

The temperature dependence of the magnetization measured under a field of 0.01T is shown in Fig.1. Two content regions with different magnetic properties appear: (1) for x=0.2 and 0.4, the samples have antiferromagnetic properties; (2) for x=0.6, 0.8 and 1.0, the samples order from ferri- to ferromagnetically over the whole magnetic ordering temperature range. The magnetic data are also listed in Table 1.

The interpretation is evidenced by the magnetic isotherms of the samples with x=0.6 and 0.8 measured at various temperatures, as shown in Figs.2 and 3, respectively. It is seen that the magnetic isotherms of the sample with x=0.6 display the typical ferrimagnetic behaviour, and the spontaneous magnetization M_S, obtained by extrapolating the high-field parts of the isotherm to zero field, increases first, reaches the maximum of 44.6A·m^2·kg^{-1} at 280K, and then decreases at higher temperatures. For the sample with

x=0.8, the magnetization increases faster at low fields and approaches saturation at high fields, indicating the ferri-/ferromagnetic character. The spontaneous magnetization M_S decreases with increasing temperature from 57.5A·m^2·kg^{-1} at 5K to 46.7A·m^2·kg^{-1} at 305K.

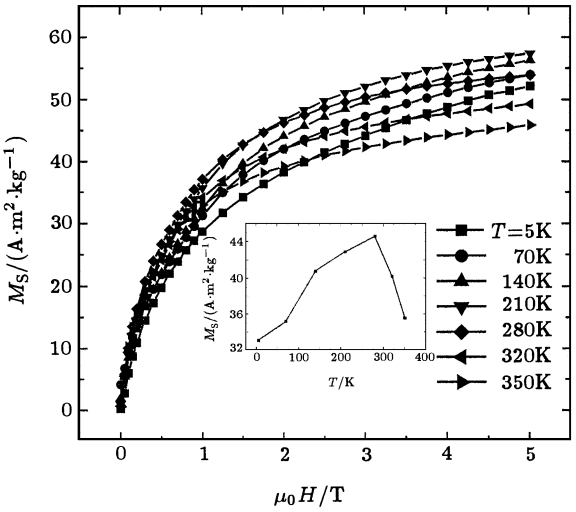


Fig.2. Magnetic field dependence of magnetization for $Er_{0.4}Sm_{0.6}Mn_6Ge_6$ at various temperatures (the inset shows the temperature dependence of the spontaneous magnetization).

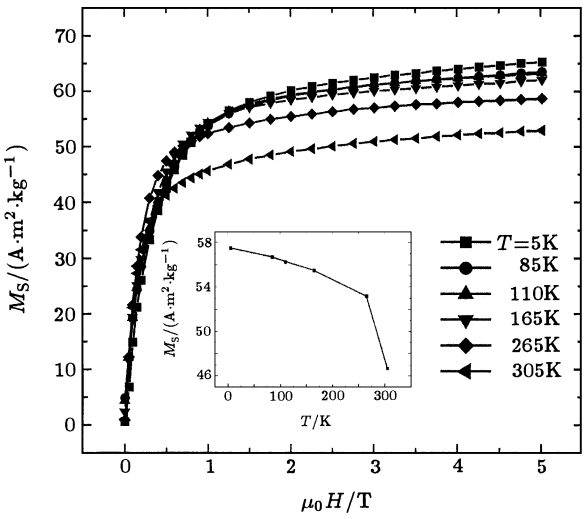


Fig.3. Magnetic field dependence of magnetization for $Er_{0.2}Sm_{0.8}Mn_6Ge_6$ at various temperatures (the inset shows the temperature dependence of the spontaneous magnetization).

A transition from an antiferromagnetic to a ferromagnetic state can be observed with the increase of Sm content. Two reasons can be used to interpret this experimental result: (1) for the coupling between Mn–Mn layers, as mentioned above, the substitution of Sm for Er leads to an increase of the lattice constants, the expansion of the Mn–Mn interlayer distance results in the ferromagnetic coupling of them; (2) for the coupling between R–Mn layers, as generally observed in R-3d intermetallic compounds, the Sm–Mn couplings are positive. This exchange interaction is very strong, causing the Sm moments to order simultaneously with the Mn moments. The strong interaction has been attributed to the larger exchange interaction between the 4f and 5d electrons in the light rare earth, resulting in an increase in the spatial extent of the 4f shell with the rare earth atomic number.[11] Thus the coupling energy between R and Mn sublattices is stronger with the increase of the substitution Sm for Er atom. The increase of the coupling energy leads to a constructive interaction with a ferromagnetic alignment between both sublattices.

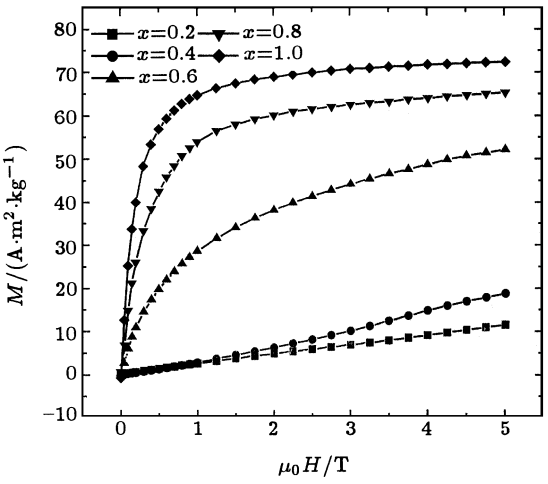


Fig.4. Magnetic field dependence of magnetization for $Er_{1-x}Sm_xMn_6Ge_6$ at 5K.

Figure 4 shows magnetization as a function of applied field at 5K. For x=0.2 and 0.4, the magnetization increases linearly in the investigated field range. For x=0.6, 0.8 and 1.0, the magnetization increases relatively faster at low fields and approaches saturation at high fields, which indicates their ferri-/ferromagnetic characters. The spontaneous magnetization M_S is presented in Table 1. It is seen that the M_S of the sample (x=0.6, 0.8 and 1.0) increases with the increase of Sm content. Neutron diffraction studies on the magnetic ordering of $ErMn_6Ge_6$[3] and $Tb_{0.8}Nd_{0.2}Mn_6Ge_6$[12] have shown that the low temperature phases are a triple skewed spiral and a triple conical spiral, respectively, in which the triple unit of Mn–R–Mn moments changes collectively their orientation by a constant angle along the c axis. The Er moment and the resultant moment component of the

Tb and Nd moments oriented antiparallelly, ordering opposite to the line bisecting the Mn–Mn interplanar angle. Thus we see that the low temperature magnetic structure of $Er_{1-x}Sm_xMn_6Ge_6$ ($x \leq 0.4$) is similar to that of $ErMn_6Ge_6$ and the Er and Sm moments may order antiparallelly. With further increase of the Sm content, the stronger positive coupling of Sm–Mn leads to the decrease of the Mn–Mn interlayer angle and then leads to the appearance of the ferrimagnetic in-plane component which increases with the Sm content, and reaches the maximum at x=1.0, i.e. the in-plane orientation of the Sm and Mn moments is parallel. Thus the M_S of them increases with increasing Sm content.

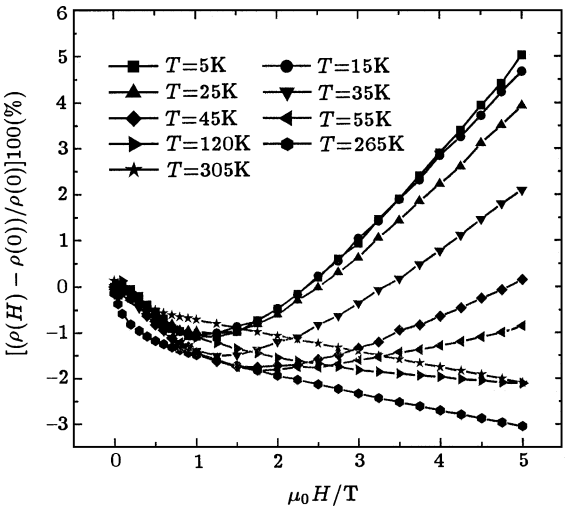


Fig.5. Magnetic field dependence of MR for $Er_{0.2}Sm_{0.8}Mn_6Ge_6$ measured at various temperatures.

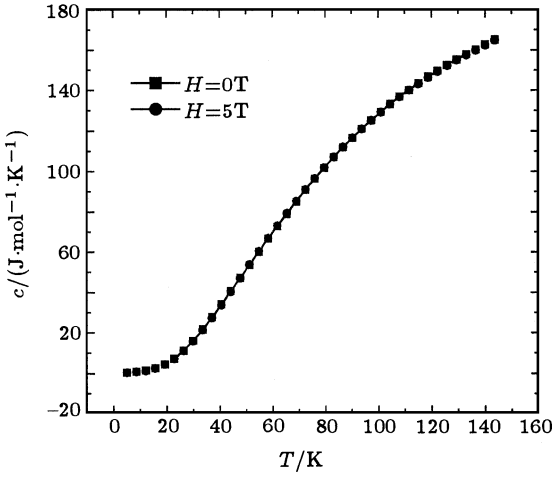


Fig.6. Specific heat (c) as a function of temperature (T) for $Er_{0.2}Sm_{0.8}Mn_6Ge_6$.

The normalized MR=[ρ(H)-ρ(0)]/ρ(0) for the sample with x=0.8 as a function of applied field at various temperatures is given in Fig.5. We note that the MR changes gradually from a small negative value of about-0.85% at 55K to a relatively large positive value of about 5.02% at 5K in a magnetic field of 5T. It is seen that the sign of MR is positive at 5T below the temperature of 55K. We have measured its zero-field and magnetic-field (5–150K) specific heat (c) (Fig.6) and surprisingly we do not see any noticeable anomaly around the corresponding temperature and its value does not vary with magnetic field. It is known that the application of magnetic field induces more spin fluctuations and accordingly strengthens the spin fluctuation contribution to the electron specific heat. Since the application of magnetic field does not effect the specific heat, we may exclude the possibility that the positive MR arises from the spin fluctuation. We have considered that the origin of positive sign may be due to the impurity phase of $SmMn_2Ge_2$ present in the sample. A similar experimental phenomenon has been found in layered $SmMn_2Ge_2$ magnet studied by Sampathkumaran *et al.*[13] According to their point of view, it results from a phase transition below 30K attributable to magnetic ordering from the Sm sublattice. But this possibility is less likely owing to the following reasons: (1) it can be seen from Figs.3 and 6 that the sample with x=0.8 orders ferri-/ferromagnetically over the whole magnetic ordering temperature range, i.e. no phase transition appears at the corresponding temperature; (2) the impurity phase of $SmMn_2Ge_2$ has not been observed in the XRD of the sample with x=0.8. The positive MR may arise if the presence of a field induces some pseudogaps in some portions of the Fermi surface.[14] It may also be due to the influence of Lorentz force on the conduction electrons, as has been suggested for (Dy,Sm)Mn_6Ge_6 compound.[15] It is also clear that in the temperature range above 55K, the MR is negative. It increases with increasing field and does not saturate up to 5T. This indicates that the sample does not acquire a collinear ferromagnetic state in the range of fields studied.

In summary, the substitution of Sm for Er leads to an increase of the lattice constants and the unit cell volume and strengthens the interlayer ferromagnetic coupling between Mn–Mn and (Er,Sm)–Mn. Thus the transition from an antiferromagnetic to a ferromagnetic state can be observed with increasing Sm content. The samples with x=0.2 and 0.4 order antiferromagnetically at 420 and 425K, respectively. The

samples with x=0.6, 0.8 and 1.0 order from ferri- to ferromagnetically over the whole magnetic ordering temperature range. The corresponding Curie temperatures are 435, 441 and 446K, respectively. The origin of positive MR at low temperatures may be due to the influence of magnetic field on the Fermi surface or due to the Lorentz force on the conduction electrons.

References

[1] Bucholz W, Schuster H U and Anorg Z 1981 *Allg. Chem.* **482** 40

[2] Olenitch R R, Akselrud L G and Yarmoliuk Ya P 1980 *Dopov. Akad. Nauk Ukr. RSR* A **2** 84

[3] Schobinger-Papamantellos P, Andre G, Rodriguez-Carvajal J and Buschow K H J 1995 *J. Alloys Compounds* **219** 176

[4] Levy P M, Zhang S and Fert A 1990 *Phys. Rev. Lett.* **65** 1643

[5] Mallik R, Sampathkumaran E V and Paulose P L 1997 *Appl. Phys. Lett.* **71** 2385

[6] Verbanck G, Temst K, Mae K Schad R, Van Bael M J, Moshchalkov V V and Bruynseraede Y 1997 *Appl. Phys. Lett.* **70** 1477

[7] Mallik R, Sampathkumaran E V, Paulose P L and Nagarajan V 1997 *Phys. Rev.* B **55** R8650

[8] Das I and Sampathkumaran E V 1994 *Phys. Rev.* B **49** 3972

[9] Das I and Sampathkumaran E V 1995 *Phys. Rev.* B **51** 1308

[10] Sampathkumaran E V and Das I 1995 *Phys. Rev.* B **51** 8631

[11] Belorisky E, Fremy M A, Gavignan J P, Givord D and Li H S 1987 *J. Appl. Phys.* **61** 3971

[12] Schobinger-Papamantellos P, Andre G, Rodriguez-Carvajal J, Duong N P and Vuschow K H J 2001 *J. Magn. Magn. Mater.* **231** 121

[13] Sampathkumaran E V, Paulose P L and Mallik R 1996 *Phys. Rev.* B **54** R3710

[14] Das I and Rawat R 2000 *Solid State Commun.* **115** 207

[15] Wang R W, Zhang L G, Zhang S Y, Yao J L, Liu B D and Shen B G 2004 *J. Phys.* D; *Appl. Phys.* **37** 385