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Magnetic properties and magnetoresistance of $HfFe_6Ge_6$-type $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds*

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The magnetic properties and magnetoresistance effects of $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds have been studied by magnetic properties and resistivity measurements in applied magnetic fields up to 5T. The compounds with x=0.1, 0.2, 0.4 and 0.5 order antiferromagnetically at 425, 428, 430 and 432K, respectively, and there are second magnetic phase transitions below 100K. The compound with x=0.6 exhibits a transition from ferrimagnetic to antiferromagnetic, then to ferrimagnetic state again with decreasing temperature. Furthermore, it displays a field-induced metamagnetic transition, and its threshold field decreases with increasing temperature. The magnetoresistance curve of the compound with x=0.6 in applied magnetic fields up to 5T is presented and the magnetoresistance effects are related to the metamagnetic transitions.

Keywords: magnetic property, magnetoresistance effect
PACC: 7525, 7530E, 7530V

1. Introduction

The structure and magnetic properties of the RMn_6X_6 (R=rare earth metal, X=Ge, Sn) compounds have been studied extensively.[1–3] Most of them crystallize in the hexagonal $HfFe_6Ge_6$-type structure (space group $P6/mmm$), which consists of alternate (001) layers containing R and Mn atoms, in which the R atoms form hexagonal planes (H), Mn atoms form Kagome nets (K) stacked along the c-axis with the sequence $...KHKKHK...$[1] The evolution of the magnetic behaviour as a function of the R element is more complicated in relation to the different strengths of the R-Mn interactions and to the magnetocrystalline anisotropy of the R atom.[2] In order to clarify the effect of substitution of Gd for Dy atom on the magnetic properties of $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds, we have studied their magnetic properties and magnetoresistance (MR) in detail.

2. Experimental

The $Dy_{1-x}Gd_xMn_6Ge_6$ compounds (x=0.1–0.6) were prepared by arc-melting in a high-purity argon atmosphere. The elements used were of at least 99.9% purity. An excess of Dy, Gd and Mn was added to compensate for the loss of weight during the arc-melting. The ingots were remelted at least three times to ensure the homogeneity. They were subsequently sealed in an evacuated quartz tube and annealed at 1073K for ten days, then quenched into water. The crystal structure of them were measured by using x-ray diffraction (XRD) with Cu$K\alpha$ radiation. The magnetic properties were measured by a vibrating sample magnetometer (VSM) and a superconducting quantum interface device (SQUID). The MR measurement was carried out by means of the standard four-probe DC method with an electric current of 100 mA

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parallel to the magnetic field. The sample used in the MR measurement was a square cross-sectional rod with dimensions of $10\times2\times2\text{mm}^3$.

3. Results and discussion

Analysis of the XRD patterns indicates that the $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds are isotypic of $HfFe_6Ge_6$ in the whole Gd concentration range investigated. The lattice parameters do not vary significantly, but increase with the Gd content. These values are listed in Table 1.

Table 1. Lattice parameters and magnetic data of $Dy_{1-x}Gd_xMn_6Ge_6$ compounds.

x	a/nm	c/nm	c/a	V/nm^3	$T_{N,C}$/K	T_t/K	M_s/(Am^2/kg)
0.1	0.5224	0.8161	1.5622	0.1929	425	83	1.08
0.2	0.5226	0.8164	1.5622	0.1931	428	84	1.90
0.4	0.5228	0.8168	1.5624	0.1933	430	86	2.98
0.5	0.5230	0.8172	1.5625	0.1936	432	88	6.05
0.6	0.5232	0.8176	1.5627	0.1938	450	90	7.89

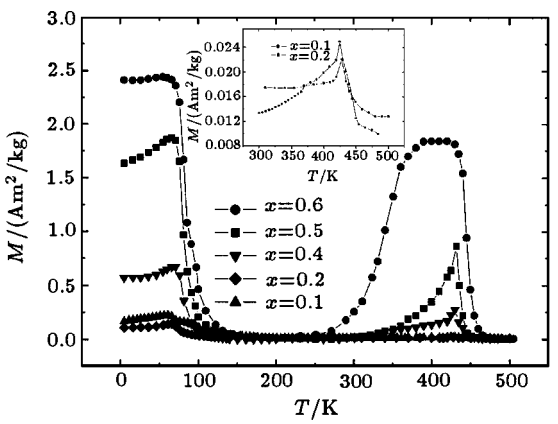


Fig.1. Thermal variation of the magnetization for the $Dy_{1-x}Gd_xMn_6Ge_6$ compounds under an applied field of 0.01T (the inset shows the M-T curves near T_N for x=0.1 and 0.2).

The thermal variation of magnetization is presented in Fig.1. The compounds with x=0.1, 0.2, 0.4 and 0.5 order antiferromagnetically at 425, 428, 430 and 432K, respectively. Above T_C=450K, the compound with x=0.6 is paramagnetic. At temperatures just below T_C, where the magnetization reaches a comparatively high value, this compound is ferrimagnetically ordered with a basically ferromagnetic Mn- sublattice moments coupled antiparallelly to basically ferromagnetically aligned (Dy,Gd)- sublattice moments, and at mediate temperatures it is antiferromagnetically ordered. Below 100K, all the above compounds display more or less enhancement of magnetization, which should be regarded as a transition from antiferromagnetic to ferrimagnetic or incommensurate cone state in which both the (Dy,Gd)- and Mn- sublattices might have the ferromagnetic component along the easy axis. These components are antiparallel and lead to a net moment in the easy direction.[3] The temperature of this transition increases with Gd content from 83K for x=0.1 to 90K for x=0.6. These transition temperatures (T_t) are listed also in Table 1.

It is worth noting that the ferrimagnetic or ferromagnetic components of the compounds (x=0.1–0.6) increase with the Gd content in the region of 300–450K and below 100K. The magnetic coupling energy between the R- and Mn- sublattice moments is proportional to $n_{R\text{Mn}}M_RM_{\text{Mn}}$, which in turn is proportional to $(g-1)<J>_R M_{\text{Mn}}$. The quantity $(g-1)J$ is the largest for $R\equiv$Gd and falls off rapidly towards both ends of the lanthanide series.[1] Thus, the coupling energy of (Dy,Gd)- and Mn- sublattice coupling is stronger with increasing the substitution of Gd for Dy. The increase of the coupling energy between the (Dy, Gd)- and Mn- sublattices leads to a constructive interaction with a ferrimagnetic alignment between both sublattices.

The magnetization curves of $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds at 5K measured by SQUID are shown in Fig.2. The spontaneous magnetizations are measured. This feature is in good accordance with the low-temperature ferrimagnetic or incommensurate cone structure. The spontaneous magnetizations (M_s) of $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds, obtained by extrapolating the high-field part of the isotherm to zero field, increase with the content of Gd. These values are given in Table 1. The magnetization curves of the compounds with x=0.5 and 0.6 display a much more ferromagnetic or ferrimagnetic behaviour. The magnetic isotherms of the compound with x=0.6 at 100 and 150K were measured by increasing the field from 0 to 5T, then decreasing the field from 5 to −1T, as shown in Fig.3.

It is seen that the magnetic isotherms at 100 and 150K display distinct hysteresis. This suggests that a transition from an antiferromagnetic to a ferrimagnetic or conical state can be induced by an external magnetic field. Furthermore, the coercive fields decrease from 0.11 to 0T with increasing temperature from 100 to 150K. The compound (x=0.6) isotherm curves have been recorded at various temperatures in order to check the evolution of the metamagnetic process (Fig.4). The thermal variation of the threshold field is depicted in the inset of Fig.4. It is worth noting that the threshold field decreases with increasing temperature.

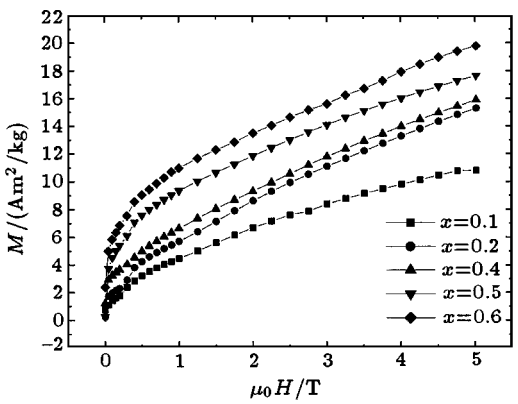


Fig.2. Magnetization versus applied field for the $Dy_{1-x}Gd_xMn_6Ge_6$ compounds at 5K.

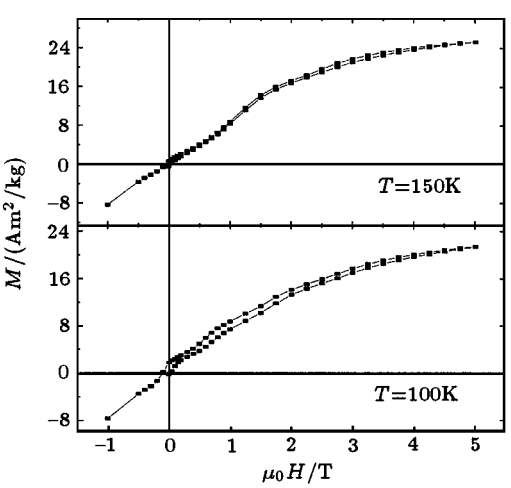


Fig.3. Magnetic isotherms for the $Dy_{0.4}Gd_{0.6}Mn_6Ge_6$ compound measured at 100 and 150K.

The thermal variation of MR for the compound with x=0.6 is presented in Fig.5. The MR values are positive in low temperatures, and are negative in higher temperatures. The occurrence of positive MR value is puzzling because MR is generally characterized by a negative value. Similar anomalies have been studied in a few ferromagnetic materials by Mallik *et al*,[4,5] in $LaMn_2Ge_2$ and $SmMn_2Ge_2$. According to their point of view, the application of H presumably induces more spin fluctuations (in contrast to the usual influence of H to suppress spin fluctuations) in a stable conical arrangement, as a result of which the resistivity (ρ) in the presence of a field is larger. This view may be used to interpret our experimental results. As mentioned above, the magnetic structures of $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds may be in a cone structure at low temperatures. In addition, the curve of MR versus temperature exhibits three peak values, 1.24%, –1.25% and –2.66% at 15, 65 and 280K, respectively. As already observed in related Mn compounds,[6] large values of MR are observed in phases with metamagnetic transitions.

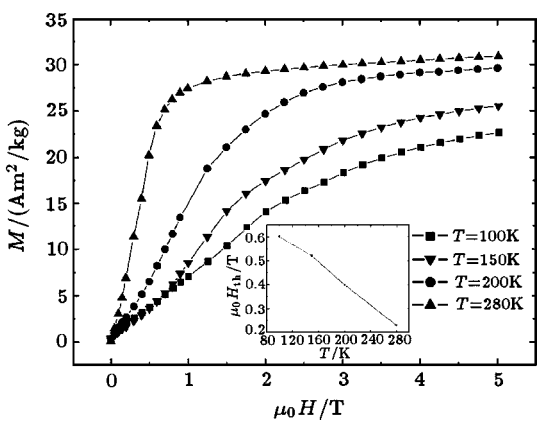


Fig.4. Magnetization versus applied field for the $Dy_{0.4}Gd_{0.6}Mn_6Ge_6$ compound at various temperatures (the inset shows the thermal variation of the threshold field).

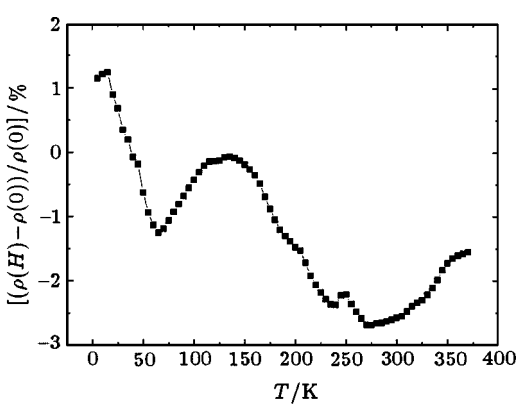


Fig.5. MR versus temperature for the $Dy_{0.4}Gd_{0.6}Mn_6Ge_6$ compound.

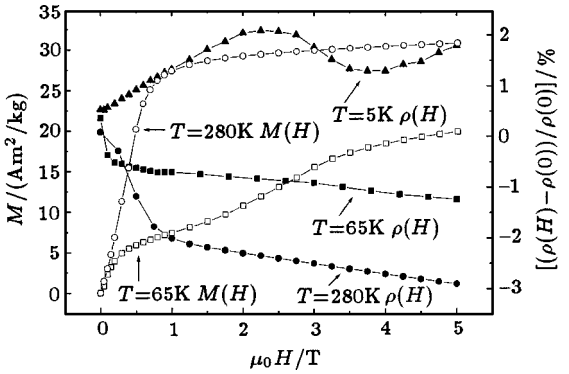


Fig.6. Magnetization and MR versus magnetic field at 5, 65 and 280K for the $Dy_{0.4}Gd_{0.6}Mn_6Ge_6$ compound.

The field dependence of MR and the magnetization at 65 and 280K, and that of MR at 5K of the compound with x=0.6 are plotted in Fig.6. The value of MR at 5K is positive in the applied field range, which is compatible with the curve of MR versus temperature. The absolute value of MR at 280K first increases sharply with increasing field in the magnetic field range below 1T, but at 0.23T there is an inflection with respect to the threshold field, then increases linearly with further increase of field from 0.2 to 5T. The sharp increase of the negative MR shows that the compound with x=0.6 undergoes a transition from the antiferromagnetic to the ferrimagnetic state. Comparing the magnetization curves with the MR curves, we can see that the threshold field (0.23T) occurring in magnetization is almost the same as that occurring in the MR effect. The MR of the compound at 65 K decreases continuously, mirroring the magnetization trend.

In summary, the substitution of Gd for Dy atom yields a significant effect on the intersublattice couplings and the magnetic properties of the $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds, and hence the $Dy_{1-x}Gd_xMn_6Ge_6$ (x=0.1–0.6) compounds display abundant magnetic behaviours. The compounds with x=0.1, 0.2, 0.4 and 0.5 order antiferromagnetically at 425, 428, 430 and 432K, respectively; and there are the second magnetic phase transitions below 100 K. The transition temperatures increase with the Gd content from 83K for x=0.1 to 90K for x=0.6. The compounds with x=0.6 exhibit a transition from ferrimagnetic to antiferromagnetic, then to ferrimagnetic or cone state with decreasing temperature. Furthermore, it displays a field-induced metamagnetic transition, and the threshold field decreases with increasing temperature. An obvious MR effect is observed for the compound with x=0.6, and the absolute values with respect to the temperatures of metamagnetic transition (65 and 280K) are 1.25% and 2.66%.

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