SPIN REORIENTATION AND EXCHANGE COUPLING IN THE $Dy_{1-x}Er_xFe_{10}Si_2$ COMPOUNDS*

P. STEFAŃSKI, A. KOWALCZYK, D. SKORYNA, A. SZLAFEREK
AND A. WRZECIONO

Institute of Molecular Physics, Polish Academy of Sciences
Smoluchowskiego 17/19, 60-179 Poznań, Poland

The structural and magnetic properties of $Dy_{1-x}Er_xFe_{10}Si_2$ are investigated. X-ray analysis reveals that these compounds are of the tetragonal $ThMn_{12}$ structure. In this structure the rare earth atoms occupy one crystallographic position 2(a). The unit cell contains 26 atoms. The spin reorientation temperature, T_{SR}, was measured from the temperature dependence of the initial susceptibility using an ac bridge of mutual inductance of the Harsthorn type. Dy^{+3} and Er^{+3} have opposite contributions to the entire magnetic anisotropy. The spin reorientation temperature is found to be about 271 K in $DyFe_{10}Si_2$ and 48 K in $ErFe_{10}Si_2$. The values are discussed applying the crystal field model. The value of the rare earth–transition metal exchange coupling constant J_{RFe}/k_B derived from the mean-field model analysis of the Curie temperature is about -13 K. The Fe–Fe exchange integral is much higher and is equal to about 75 K.

PACS numbers: 75.30.Gw, 75.50.Gg

1. Introduction

Recently, many groups of researchers have investigated the iron-rich ternary compounds of the type $R(Fe,T)_{12}$ in which R is a rare earth element and T is a stabilizing element such as titanium, vanadium, chromium, silicon, etc. All these compounds crystallize in the $ThMn_{12}$ type of structure [1]. In this structure the R atom occupies one kind of crystallographic site, ie. 2(a), the other atoms occupy 8(i), 8(j) and 8(f) positions.

In the rare earth–transition metal compounds (R–T), the contribution to the net anisotropy from the R sublattice generally dominates at lower temperatures, whereas the T-sublattice anisotropy dominates at high temperatures. Also, the R-sublattice anisotropy decreases much faster with increasing temperature than the T-sublattice anisotropy. In the $RFe_{10}Si_2$ compounds, the Fe anisotropy favours the c-axis in the whole range of temperature [2]. The easy magnetization

---

direction (EMD) of the R sublattice depends on the crystal electric field (CEF) interaction and the exchange field experienced by the R ion.

In this paper the competition between the opposite contributions of different rare earth sublattices to the entire anisotropy is investigated. For this purpose we used two ions with different signs of second-order Stevens factors: $Dy(\alpha_J < 0)$ and $Er(\alpha_J > 0)$.

2. Experimental

The samples were prepared by induction melting of stoichiometric amounts of the constituents in a water-cooled boat under atmosphere of argon. The ingots were inverted several times to insure homogeneity. Then the samples were wrapped in a Ta foil, sealed in a quartz tube filled with argon and annealed for two weeks at $900^\circ C$ and then rapidly cooled to room temperature. X-ray diffraction was performed on powdered samples using Cu K_α radiation. Diffraction patterns showed only characteristic lines of the $ThMn_{12}$ structure.

The spin reorientation (SR) temperature, T_{SR}, was determined from measurements of the temperature dependence of the initial susceptibility using an ac bridge of mutual inductance of the Harsthorn type. The intensity of alternating field used was 20 A/m and frequency — 15 Hz.

The Curie temperature was determined by measuring the temperature dependence of magnetization (M) under a weak external field. The value of T_C was obtained by plotting M^2 versus T and extrapolating to $M^2 = 0$.

3. Results and discussion

X-ray diffraction patterns revealed that the compounds crystallize in the tetragonal $ThMn_{12}$-type structure.

The T_C values decrease when Dy atoms are replaced by Er ones (Table). This decrease in T_C may be related to the reduction of the lattice parameters with the increasing Er content. Because most of the Fe–Fe distances in $RFe_{10}Si_2$ compounds are less than 2.5 Å, a decrease in lattice parameters would mean a decrease in Fe–Fe distance and consequently, results in reduction of the exchange interactions and a decrease in T_C.

In order to derive information regarding the magnetic coupling constants J_{FeFe} and J_{RFe} between the rare earth (R) and Fe moments in the $RFe_{10}Si_2$ compounds, we have performed a standard mean-field analysis of the observed Curie temperatures. In this method T_C can be expressed as

$$3k_B T_C = a_{FeFe}^2 + (a_{FeFe}^2 + 4a_{RFe}a_{FeR})^{1/2}, \tag{1}$$

where

$$a_{FeFe} = Z_{FeFe}J_{FeFe}S_{Fe}(S_{Fe} + 1), \tag{2}$$

$$a_{RFe}a_{FeR} = Z_{RFe}Z_{FeR}S_{Fe}(S_{Fe} + 1)(g_J - 1)^2 J(J + 1)J_{RFe}^2. \tag{3}$$

Equation (1) is obtained after neglecting weak R–R interactions. From the crystal structure of $ThMn_{12}$, it follows that each R atom has an average number

TABLE
Curie temperatures and spin-reorientation temperatures of the $Dy_{1-x}Er_xFe_{10}Si_2$ compounds.

x	T_C [K]	T_{SR} [K]
0.0	568	271
0.25	562	218, 134
0.50	557	207
1.0	549	48

$Z_{RFe} = 17$ Fe nearest neighbours while each Fe atom has an average number of $Z_{FeR} = 2$ R nearest neighbours and an average number of $Z_{FeFe} = 8$ Fe neighbours. $S_{Fe} = 0.65$ is the quasi-spin of the Fe ions. Using the experimental data of T_C (535 K for $YFe_{10}Si_2$ [2]) one finds $J_{FeFe}/k_B = 75$ K. This value of the coupling constant was subsequently used in Eq. (1) when applied to $DyFe_{10}Si_2(T_C = 568$ K) and $ErFe_{10}Si_2(T_C = 549$ K). This leads to the values of J_{RFe}/k_B equal to 13 K and 12 K for $DyFe_{10}Si_2$ and $ErFe_{10}Si_2$, respectively.

The value of the uniaxial anisotropy constant K_1 due to the rare earth sublattice is given by the equation [3]

$$K_1 = (-3/2)\alpha_J \langle r^2 \rangle A_2^0 \langle O_2^0 \rangle + 5\beta_J \langle r^4 \rangle A_4^0 \langle O_4^0 \rangle + (-21/2)\gamma_J \langle r^2 \rangle A_6^0 \langle O_6^0 \rangle. \quad (4)$$

It is valid in this class of compounds in the large exchange limit [4]. α_J, β_J, γ_J are the Stevens factors [5], $\langle r^n \rangle$ are Hartree–Fock radial integrals [6], A_n^m are crystal electric field potentials. O_n^m are Stevens equivalent operators [4].

The magnitude and sign of K_1 is mainly determined by the first term in Eq. (4). Knowing that $A_2^0 < 0$ [7] in this class of compounds for the rare earth with $\alpha_J < 0$ (Nd, Tb, Dy, Ho), the rare earth anisotropy constant K_1 has a negative sign (when neglecting higher order terms in Eq. (4)) and for these ions SR transition should appear. For $DyFe_{10}Si_2$ $T_{SR} = 271$ K (see Table). For Er^{+3} ion ($\alpha_J > 0$) SR transition appears as a result of domination of the second and third term in Eq. (4). In this class of compounds for Er^{+3} ion the higher order crystal field terms has a remarkable influence on the anisotropy [8].

In $Dy_{1-x}Tm_xFe_{10}Si_2$ samples there exist two rare earth sublattices each of them manifesting SR transition. A "mixing" of SR takes place. It appears that for $x = 0.25$ the SR at $T_{SR} = 134$ K comes from Er-sublattice. The Er anisotropy is enhanced by the Dy planar anisotropy. SR at 218 K comes from Dy-sublattice. While lowering Dy content T_{SR} coming from Dy-sublattice shifts to lower temperatures and as a result of competition of Dy- and Er-sublattice anisotropies with axial Fe-sublattice anisotropy SR transition is produced at 207 K.

References

[1] D.B. de Mooij, K.H.J. Buschow, *Philips J. Res.* **42**, 246 (1987).

[2] P. Stefański, A. Wrzeciono, *J. Magn. Magn. Mater.* **82**, 125 (1989).

[3] P.-A. Lindgård, O. Danielsen, *Phys. Rev. B* **11**, 351 (1975).

[4] P. Stefański, A. Wrzeciono, *Phys. Status Solidi B* **151**, K195 (1989).

[5] M.T. Huthings, *Solid State Phys.* **16**, 277 (1964).

[6] A.J. Freeman, R.E. Watson, *Phys. Rev.* **127**, 2058 (1962).

[7] K.H.J. Buschow, D.B.de Mooij, M. Brouha, H.H.A. Smit, R.C. Thiel, *IEEE Trans. Magn.* **MAG-24**, 227 (1964).

[8] P. Stefański, A. Kowalczyk, *Solid State Commun.* **77**, 397 (1991).