Structure, exchange interactions and magnetic anisotropy of $Er_2Co_{17-x}Ga_x$ (x = 0-8) compounds

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Bing Liang†, Bao-gen Shen, Fang-wei Wang, Shao-ying Zhang, Zhao-hua Cheng, Tong-yun Zhao and Wen-shan Zhan

State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, People's Republic of China

Abstract. The structural and magnetic characteristics of Er_2Co_{17}-based ternaries, in which Co is partly substituted by Ga atoms, have been investigated by means of x-ray powder diffraction (XRPD) and magnetic measurements. XRPD patterns show that all samples are single phase with the rhombohedral Th_2Zn_{17}-type structure except the sample with $x = 8$ which contains a small amount of hcp Co and rare-earth-rich phase as impurity phases. The Ga substitution for Co in these compounds leads to a monotonic increase in unit cell volume and a monotonic decrease in Curie temperature. The saturation magnetization is found to approximately linearly decrease with increasing Ga concentration. For $Er_2Co_{17-x}Ga_x$ compounds with $x = 4$, 5 and 6, compensation points are observed in the corresponding temperature dependence of the magnetization. The values of compensation temperature are found to shift to higher temperature with increasing Ga content. The intersublattice-molecular-field coefficients, n_{Er-Co}, have been determined on the basis of magnetization curves measured at the compensation temperature. The exchange-coupling constants J_{Er-Co} have been calculated by analysing the field dependence of the magnetization and by using the method based on magnetic-ordering temperatures. It has been found that the Ga substitution for Co in these compounds has little influence on Er–Co exchange interaction. XRPD patterns on magnetically aligned samples with $x \leqslant 5$ show that the easy magnetization direction at room temperature corresponds to the c-axis.

1. Introduction

Rare-earth (R) transition-metal (T) compounds of the 2:17 type, with T = Co, or Co + other transition metals, which can serve as the principal phase in permanent magnets, have been extensively studied during the past few decades [1–11]. Especially, the magnetocrystalline anisotropies of these compounds are of substantial technological and theoretical interest. A strongly uniaxial magnetocrystalline anisotropy is required to achieve high coercivities. The R_2Co_{17} binary compounds exhibit uniaxial anisotropy only for R = Sm, Er and Tm. It has been noted previously [2, 12] that the easy magnetization direction (EMD) follows the sign of the second Stevens factor, α_J, of the R^{3+} ion in the R_2Co_{17} intermetallics. The Co-sublattice anisotropy is too weak to affect this result. In previous investigations, various elemental substitutions (such as Ga, Al and Si) for Fe in the R_2Fe_{17} compounds were systematically investigated [13–20]. The results showed that the substitution of these nonmagnetic atoms for Fe has a profound influence on the magnetic properties, especially on determining the EMD of these compounds. As an extension of our previous work,

† Corresponding author. Present address: Max-Planck-Institut für Metallforschung, Heisenbergstrasse 1, D-70569 Stuttgart, Germany. E-mail address: liang@axphys.mpi-stuttgart.mpg.de

we study the effect of substitution of nonmagnetic atoms on the structure and magnetic properties of the Co-based 2:17-type compounds in order to gain more valuable knowledge about the fundamental aspects of the magnetism in R–T compounds. Here we select R = Er because the study of the R 3d coupling parameter is easier to perform on materials with antiparallel coupling in the heavy R 3d compounds. We select Ga as the substituting element since Ga atoms can substitute for Co up to a quite high concentration without changing the crystal structure. In this paper, we study the influence of Ga substitution in $Er_2Co_{17-x}Ga_x$ ($0 \leqslant x \leqslant 8$) compounds on the structure and the magnetic properties, such as Curie temperature, compensation temperature, saturation magnetization, exchange-coupling strength between the 4f and 3d moments and magnetocrystalline anisotropy.

2. Experiment

Samples with composition of $Er_2Co_{17-x}Ga_x$ ($x = 0, 1, 2, 3, 4, 5, 6, 7$ and 8) were prepared by arc-melting from raw materials of at least 99.9% purity under highly purified argon atmosphere. The ingots were turned over and remelted at least three times to ensure their homogeneity. As-cast alloys were sealed in quartz tubes and annealed at 1300 K for 72 hours, then rapidly cooled to room temperature.

X-ray powder diffraction (XRPD) with Cu Kα radiation was used to determine phase purity, crystal structure and lattice parameters. Magnetic anisotropy studies were undertaken by using XRPD methods on samples which had been magnetically oriented. The oriented samples were prepared by mixing the powder with epoxy resin and then aligning at room temperature in an external field of 1 T.

The magnetization curves were measured at 5 K using a superconducting quantum interference device (SQUID) in magnetic fields up to 7 T. The saturation magnetization has been derived by extrapolation of the fitting of the experimental data of M–H curve towards saturation. The Curie temperature and the compensation temperature were derived from the temperature dependence of magnetization measured with the SQUID ($5 \text{ K} \leqslant T \leqslant 300 \text{ K}$) and a Faraday magnetic balance ($T \geqslant 300 \text{ K}$).

3. Results and discussion

XRPD patterns indicate that all samples are almost single phase of the rhombohedral Th_2Zn_{17}-type structure except the sample with $x = 8$ which contains a small amount of hcp Co and rare-earth-rich phase as impurity phases. For the compounds of $x > 8$, small amounts of an unidentified secondary phase show up. As an example, figure 1(a) shows the typical XRPD patterns of $Er_2Co_{17-x}Ga_x$ samples with $x = 2$ and 5. The lattice constants, a and c, derived from the XRPD patterns, are illustrated in figure 2 as a function of Ga concentration and also listed in table 1 together with the c/a-ratio. The lattice constant c increases monotonically from $x = 0$ to 4, remains almost constant between $x = 4$ and 6, and then decreases between $x = 6$ and 8, while the lattice constant a increases monotonically with increasing Ga content. The unit cell volume shows a virtually linear expansion at a rate of 8.15 Å^3 per Ga atom, as can be seen in figure 1. This variation in lattice constants is very similar to those observed in other $R_2Co_{17-x}Ga_x$ compounds with R = Y, Pr, Gd and Tb [21–23] and also in $R_2Fe_{17-x}Ga_x$ [16] compounds.

The saturation magnetization μ_s of $Er_2Co_{17-x}Ga_x$ compounds at 5 K are presented in table 2 and are shown in figure 3 as a function of Ga concentration. It can be seen that the μ_s decreases almost linearly with increasing Ga concentration and the slope of decline is almost
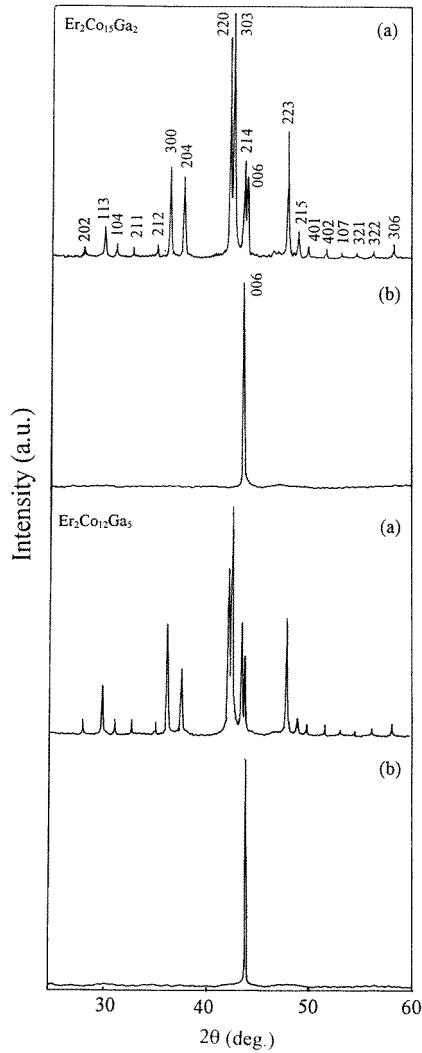
Figure 1. The typical XRPD patterns of unoriented (a) and oriented (b) $Er_2Co_{17-x}Ga_x$ powders with $x = 2$ and 5.

the same as those in the case of other $R_2Co_{17-x}Ga_x$ compounds, such as R = Y, Pr, Gd and Tb [21–23]. From figure 3 one can also observe that the decline in saturation magnetization is greater than expected due to the simple magnetic dilution of the Co sublattices, which is derived from extrapolating to the Er moment in Er_2Ga_{17}. This behaviour implies that the decrease in μ_s is not only due to the reduced Co content but also due to the transfer of electrons of Ga atoms to the 3d vacancies in cobalt atoms [24]. The magnetic moment μ_s of $Er_2Co_{17-x}Ga_x$ compounds can be described as

$$\mu_S = (17 - x)\mu_{Co} - 2\mu_R. \tag{1}$$

If the μ_{Co} values in $Er_2Co_{17-x}Ga_x$ compounds are assumed to be equal to those in $Y_2Co_{17-x}Ga_x$ compounds [22], the values of μ_{Er} can be determined from (1). As can
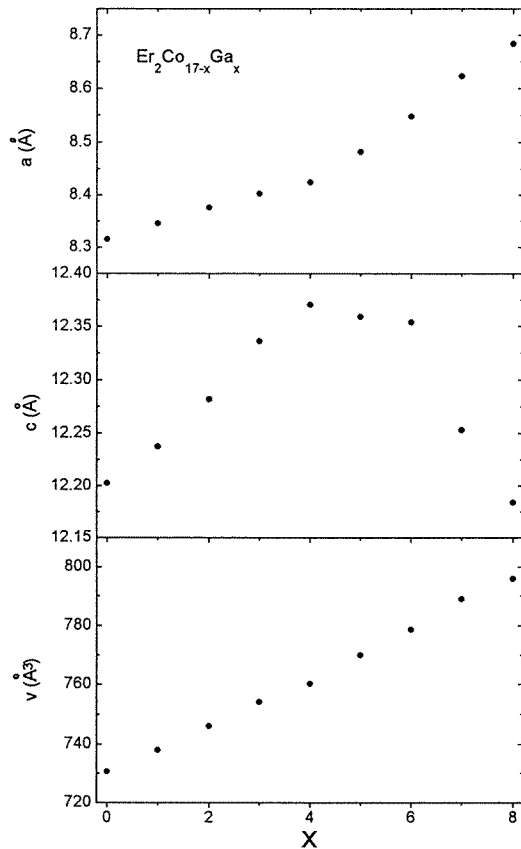
Figure 2. Ga-concentration dependence of the lattice parameters of $Er_2Co_{17-x}Ga_x$ compounds.

Table 1. The lattice constants, a, c, the unit cell volume, v, and c/a-ratio of $Er_2Co_{17-x}Ga_x$ compounds.

Compounds	a (Å)	c (Å)	v (Å^3)	c/a
Er_2Co_{17}	8.315	12.203	730.67	1.47
$Er_2Co_{16}Ga$	8.345	12.237	737.99	1.47
$Er_2Co_{15}Ga_2$	8.375	12.282	746.04	1.47
$Er_2Co_{14}Ga_3$	8.402	12.336	754.19	1.47
$Er_2Co_{13}Ga_4$	8.424	12.371	760.20	1.47
$Er_2Co_{12}Ga_5$	8.482	12.359	769.99	1.46
$Er_2Co_{11}Ga_6$	8.524	12.354	777.45	1.45
$Er_2Co_{10}Ga_7$	8.623	12.253	789.00	1.42
$Er_2Co_9Ga_8$	8.684	12.184	795.88	1.40

be seen in table 2, μ_{Er} derived in this way are found to be slightly lower than free-ion magnetic moment and decrease with increasing Ga content. The decrease of Er moments implies that 4f electrons are also slightly influenced by Ga atoms.

It is found from figure 3 that for the $Er_2Co_{17-x}Ga_x$ compounds with $x \cong 3.8$ the saturation magnetization becomes zero owing to mutual compensation of the Er-sublattice and Co-sublattice moments. When Ga content x is higher than 3.8, the Er-sublattice

Table 2. The magnetic parameters for Er$_2$Co$_{17-x}$Ga$_x$ compounds.

Compounds	T_C (K)	μ_s (μ_B fu^{-1}) (at 5 K)	μ_R (μ_B)	gJ (μ_B)	μ_{Co} (μ_B)	T_{comp} (K)	EMD (at room temperature)
Er$_2$Co$_{17}$	1199	11.13	8.8	9.0	1.69		c-axis
Er$_2$Co$_{16}$Ga	1069	8.11	8.42	9.0	1.56		c-axis
Er$_2$Co$_{15}$Ga$_2$	932	5.23	8.14	9.0	1.43		c-axis
Er$_2$Co$_{14}$Ga$_3$	793	1.91	8.27	9.0	1.32		c-axis
Er$_2$Co$_{13}$Ga$_4$	602	−1.11	8.06	9.0	1.15	35	c-axis
Er$_2$Co$_{12}$Ga$_5$	396	−3.56	7.47	9.0	0.95	54	c-axis
Er$_2$Co$_{11}$Ga$_6$	251	−5.97	7.47	9.0	0.81	67	—
Er$_2$Co$_{10}$Ga$_7$	128	−8.17	7.46	9.0	0.67		—
Er$_2$Co$_9$Ga$_8$	25	−10.50	7.29	9.0	0.45		—
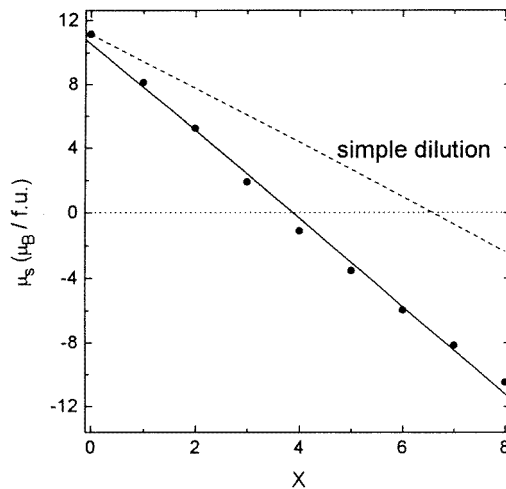
Figure 3. Ga-concentration dependence of the saturation magnetization of Er$_2$Co$_{17-x}$Ga$_x$ compounds at 5 K.

moment at 5 K is larger than that of the Co sublattice. However, since the indirect Er–Er, Er–Co exchange interactions are weaker than the direct Co–Co exchange interaction, the Er-sublattice moment drops off much more rapidly as a function of temperature than that of the Co sublattice, and the moments of the two sublattices will cancel at certain temperature. This can be clearly observed from the temperature dependence of the magnetization in figure 4 for Er$_2$Co$_{17-x}$Ga$_x$ compounds with $x = 4$, 5 and 6. The compensation temperature at which the two sublattice moments are equal is 35 K, 54 K and 67 K for $x = 4$, 5 and 6, respectively. The Ga substitution results in a decrease in the moment of the Co sublattice; as a consequence, the compensation point of Er$_2$Co$_{17-x}$Ga$_x$ compounds shifts to higher temperature with increasing Ga content.

The magnetic coupling between the magnetic moments in a lattice consisting of N_R rare-earth atoms and N_T transition-metal atoms is commonly described by means of a two-sublattice mean field model. The intersublattice-coupling strength n_{RT} can be determined experimentally by several different methods. One of them is the high-field magnetization measurements performed on finely powdered samples (HFFP) that are free to orient their magnetization according to the applied field. This so-called HFFP method has been used to
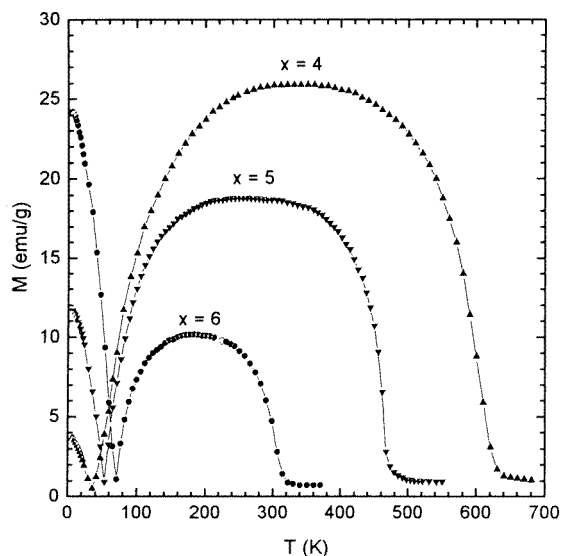
Figure 4. Temperature dependence of magnetization of $Er_2Co_{17-x}Ga_x$ compounds with $x = 4$, 5 and 6.

determine the accurate value of n_{RT} in a fairly large number of different compounds [25–27] recently. The behaviour expected as a function of the applied field can be characterized as follows: for the fields lower than $\mu_0 H_{crit,1} = |M_T - M_R| n_{RT}$, the moment configuration is strictly antiparallel and the magnetization is equal to $M_S = |M_T - M_R|$. For the fields higher than $\mu_0 H_{crit,2} = |M_T + M_R| n_{RT}$, the moment configuration is parallel and the magnetization is equal to $M_S = |M_T + M_R|$. For the fields between the two critical fields, the two sublattice moments commence bending towards each other and the magnetization is given by $M_S = \mu_0 H / n_{RT}$, and thus the coupling constant can be derived directly from the slope, dH/dM. This behaviour can be clearly illustrated in figure 5. But for many compounds, the second critical field is far above the accessible field range for magnetization measurements. Even the first critical field is not accessible to us. However, we have the possibility of reducing $|M_T - M_R|$ by appropriate substitutions with, for instance, non-magnetic elements. In the ideal case that $M_T = M_R$, the magnetization curve is a straight line with a slope of $1/n_{RT}$, therefore realistic values for n_{RT} can be obtained without the requirement of high fields. In fact, the condition of $M_T = M_R$ is found at the compensation temperature, so that n_{RT} can be straightforwardly derived from the $M–H$ curve within a field up to 7 T. The field dependence of the magnetization of $Er_2Co_{17-x}Ga_x$ compounds close to the compensation temperatures is given in figure 6. It is found that the $M–H$ curves measured close to the various compensation temperatures are approximately linear and parallel to each other in all samples except at the lowest fields. From a fit to the linear parts, we obtain the slope which according to the model is proportional to n_{RT}^{-1}. If only the nearest-neighbour interactions are considered and assuming that the R–T exchange coupling is spatially isotropic and distance independent in the nearest-neighbour sphere, the microscopic R–T exchange-coupling constant, J_{RT}, is related to the macroscopic intersublattice molecular-field coefficient n_{RT} via the following expression [25]:

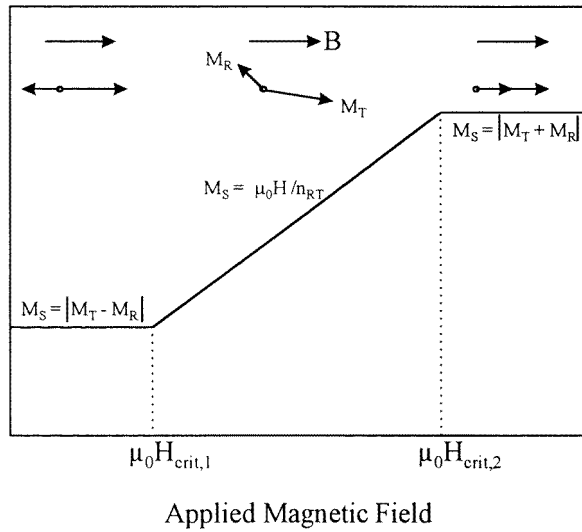$$J_{RT} = -\frac{N_T g_R \mu_B^2}{Z_{RT}(1 - g_R)} n_{RT}. \tag{2}$$

Figure 5. Field dependence of the resultant magnetic moment of a two-sublattice system. For the different ranges the related sublattice-moment configurations are indicated.
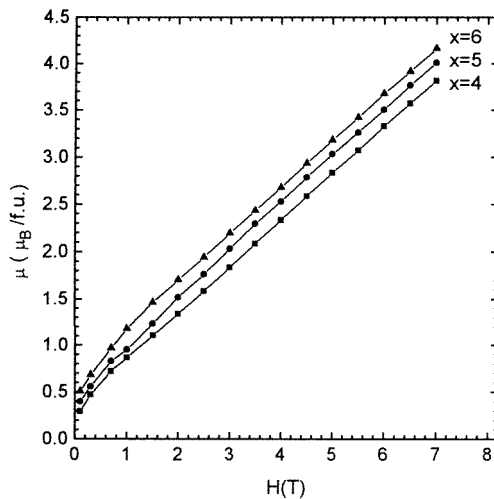

Figure 6. Field dependence of the magnetization of Er$_2$Co$_{17-x}$Ga$_x$ compounds with $x = 4$, 5 and 6 close to their compensation temperatures.

Here g_R is the Landé factor, $N_T (=17)$ is the number of transition metal T (T = Co and Ga) atoms per formula unit and $Z_{RT} (=19)$ is the number of nearest T atom neighbours of an R atom. The value of Z_{RT} is related to $Z_{TR} (=2)$ via the relation $N_R Z_{RT} = N_T Z_{TR}$ [28]. The values of n_{Er-Co} and J_{Er-Co} determined in this way have been listed in table 3.

It is also possible to determine the intersublattice-coupling strength from the difference in magnetic-ordering temperature of R$_2$T$_{17}$ compounds in which R has a magnetic moment and in which R is non-magnetic [29–31]. It is well known that the Curie temperature of R–T

Table 3. Magnetic coupling constants for $Er_2Co_{17-x}Ga_x$ compounds, n_{Er-Co} and J_{Er-Co}, in which $J_{Er-Co}^{(1)}$ are derived from M–H curves and $J_{Er-Co}^{(2)}$ is derived from the fitting of M–T curves with a mean field calculation.

Compounds	n_{Er-Co} (Tfu μ_B^{-1})	$-J_{Er-Co}^{(1)}$ ($\times 10^{-23}$ J)	$-J_{Er-Co}^{(2)}$ ($\times 10^{-23}$ J)
Er_2Co_{17}			9.55
$Er_2Co_{16}Ga$			11.04
$Er_2Co_{15}Ga_2$			10.94
$Er_2Co_{14}Ga_3$			10.71
$Er_2Co_{13}Ga_4$	2.08	10.34	10.23
$Er_2Co_{12}Ga_5$	2.21	10.99	11.59
$Er_2Co_{11}Ga_6$	1.99	9.92	9.31
$Er_2Co_{10}Ga_7$			9.93

compounds is determined by the T–T, R–T and R–R interactions. In the high-temperature limit of the mean field model, one may neglect the R–R interaction since it is of the RKKY type which is comparatively weaker than T–T and R–T interaction. From the corresponding values of the 3d spin S_T and the Curie temperature of $R_2Co_{17-x}Ga_x$ compounds in which R is non-magnetic, we can obtain the exchange-coupling constant J_{RT} by using the standard mean field formula [32]:

$$J_{RT}^2 = 9k^2 T_{C,R}(T_{C,R} - T_{C,0})/4Z_{RT}Z_{TR}S_T(S_T + 1)(g_R - 1)^2 J_R(J_R + 1) \tag{3}$$

where $T_{C,R}$ and $T_{C,0}$ denote the Curie temperatures of the compound with a magnetic rare-earth element and non-magnetic element (Y, La, or Lu), respectively. k is the Boltzmann constant. Here the T_C of $Y_2Co_{17-x}Ga_x$ compounds [22] are used as $T_{C,0}$. The values of J_{Er-Co} calculated in this way are presented in table 3 for comparison with those derived from relation (2). Within experimental accuracy, the values of J_{Er-Co} derived from the two methods are in good agreement with each other.

It can be seen from table 3 that the values of J_{Er-Co} are hardly affected by the Ga substitution. Similar effects were also observed in other $R_2Co_{17-x}Ga_x$ compounds [22, 23] and in iron-based 2:17-type compounds substituted by Ga or Al atoms [15, 18, 33–36]. Recent band-structure calculations performed by Brooks *et al* [37, 38] have shown that the key role in the R–T exchange coupling is played by the 5d electrons of the R component. On the basis of band-structure calculations performed by Brooks *et al*, Jacobs *et al* [15] offered a better explanation which can be applied to explain the constant behaviour of J_{RT} in various series of pseudo-binaries or pseudo-ternaries, such as in compounds of $R_2Fe_{17-x}M_x$ (M = Ga, Al, or Si) [33–35] and $R_2Fe_{14-x}Al_xB$ [36]. It is conceivable that the hybridization of these d bands with the s, p electron states of Ga leads to band broadening of comparable magnitude or, alternatively, to comparable reductions of the average 3d spin and average 5d spin. Since Brooks *et al* showed that the overall 4f–3d coupling strength depends on the ratio of the average 3d and 5d spins, there may be cancellation of the effect of both spin reductions, leaving the strength of J_{RT} almost unchanged.

The Curie temperature of $Er_2Co_{17-x}Ga_x$ compounds is also presented in table 2. T_C is found to decrease monotonically with increasing Ga concentration. This situation is dramatically different from that observed in various $R_2Fe_{17-x}M_x$ (M = Ga, Al and Si) compounds where the Curie temperature first strongly increases with x before decreasing at higher x value [16–20]. The relatively low T_C in R_2Fe_{17} compounds may be due to the existence of antiferromagnetic coupling between Fe moments on certain sites [39], which leads to weak overall Fe–Fe exchange interactions. The preferential substitution of

Ga, Al or Si on these sites can reduce the antiferromagnetic coupling and consequently enhance the T_C, while the magnetic coupling between Co moments in the metal system is always ferromagnetic, independent of Co moment and structure, leaving no room for antiferromagnetic types of 3d moment interaction that could reduce the Curie temperature. It is well known that for the iron- and cobalt-based rare-earth (R) transition-metal (T) compounds, the T–T interaction is the strongest one and primarily dominates the Curie temperature. The Ga substitution for Co increases the Co–Co interatomic distance and decreases monotonically the Curie temperature. This implies that the Co–Co interatomic distances are located at the right side of the Pauling–Slater curves; the increase of Co–Co distance in $Er_2Co_{17-x}Ga_x$ compounds weakens the Co–Co exchange interaction. Meanwhile, the magnetic dilution is another reason for the rapid decrease in T_C.

In order to investigate the room-temperature EMD of these compounds, the magnetic-field-oriented samples of $Er_2Co_{17-x}Ga_x$, whose T_C are above room temperature, were checked by x-ray diffraction. All samples investigated here are easy-c-axis anisotropy. As an example, figure 1(b) illustrates the room-temperature XRPD patterns of $Er_2Co_{17-x}Ga_x$ compounds with $x = 2$ and 5. The drastic increase of the (00l) peak and the complete disappearance of the (hk0) and of all (hkl) reflections reveal that the samples possess uniaxial anisotropy at room temperature. It is well known that in the R–T intermetallic compounds, both the R sublattice and the T sublattice have contributions to the bulk anisotropy energy. The former is mainly determined by the crystalline electric field (CEF) and the R–T exchange interactions and is very strongly temperature dependent, whereas the latter originates from incompletely quenched angular momentum of the 3d electrons, which, compared to the former, is less temperature dependent. To a first approximation, the total magnetocrystalline anisotropy is the sum of the first-order anisotropy constants of the R and T sublattices. In the case of the Er_2Co_{17} compound

$$K_1[\text{total}] = K_1[\text{Er}] + K_1[\text{Co}]. \tag{4}$$

The rare-earth sublattice anisotropy can be expressed [40] in crystal field terms as

$$K_1[\text{Er}] = -(3/2)\alpha_J \langle r_{4f}^2 \rangle \langle 3J_Z^2 - J(J+1) \rangle A_2^0 \tag{5}$$

where α_J is the second-order Stevens factor and A_2^0 is the second-order crystal field parameter which is determined predominantly by the rare-earth valence electron charge asphericity [41]. Because α_J is positive for the Er^{3+} ion and A_2^0 is negative for the underlying structure, a positive $K_1[\text{Er}]$ corresponds to easy-axis anisotropy. It is found before that $K_1[\text{Co}]$ is negative for the R_2Co_{17} compound [4, 5, 7], giving a negative contribution to the total magnetocrystalline anisotropy. However, $K_1[\text{Co}]$ is comparatively small for temperatures well below the Curie temperature; the positive $K_1[\text{total}]$ is responsible for the easy-axis anisotropy of Er_2Co_{17} compound at room temperature. Earlier investigations showed that the preferential substitution of non-magnetic atoms on certain Co sites can weaken the planar anisotropy of the Co sublattice and change $K_1[\text{Co}]$ from negative to positive in a certain substitution concentration range [7]. This means that the $K_1[\text{total}]$ will become more positive with the substitution of non-magnetic atoms in the case of $Er_2Co_{17-x}Ga_x$ compounds. Thus, no spin reorientation is expected to occur when the temperature varies between low temperature and Curie temperature since the sign of $K_1[\text{total}]$ remains unchanged with increasing Ga concentration. Indeed, no spin-reorientation transition is observed in the temperature dependence of the magnetization of $Er_2Co_{17-x}Ga_x$ compounds from 1.5 K up to T_C.

4. Conclusions

The substitution of non-magnetic atoms Ga for Co has a profound influence on the magnetic properties of the Co-based 2:17-type compounds. The $Er_2Co_{17-x}Ga_x$ compounds remain single phase of the rhombohedral Th_2Zn_{17} type up to $x = 8$. The Ga substitution increases the unit cell volume and the Co–Co interatomic distance; as a consequence, the Co–Co exchange coupling decreases with increasing Ga concentration. The rapid decrease in T_C results from both the decrease of exchange interaction and the magnetic dilution. The saturation magnetization is found to approximately linearly decrease with increasing Ga concentration, and the decline in saturation magnetization is faster than expected due to the simple magnetic dilution of the Co sublattices. The compensation temperatures are observed in the temperature dependence of magnetization for $Er_2Co_{17-x}Ga_x$ compounds with $x = 4$, 5 and 6 and the values of T_{comp} are found to shift to higher temperature with increasing Ga concentration. The intersublattice-molecular-field coefficients, n_{Er-Co}, are derived on the basis of magnetization curves measured at the compensation temperature. The exchange-coupling constants J_{Er-Co} are determined by analysing the field dependence of the magnetization and by using the method based on magnetic-ordering temperatures. Within experimental accuracy, the values of J_{Er-Co} derived from the two methods are in good agreement with each other. It is found that the Ga substitution for Co in $Er_2Co_{17-x}Ga_x$ compounds has little influence on Er–Co exchange interaction. XRPD patterns on magnetically aligned samples with $x \leqslant 5$ show that the easy magnetization direction at room temperature corresponds to the c-axis

Acknowledgments

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