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The Gd–Fe exchange interaction in $GdFe_6Ga_6$

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Abstract

We have investigated the Gd–Fe exchange-coupling strength in $GdFe_6Ga_6$ by means of magnetization measurements on single-crystalline powder particles that are free to rotate into their minimum-energy direction in the applied magnetic field. The experiments have been carried out at 4.2 K and at high magnetic fields up to 55 T in order to achieve bending of the Gd- and Fe-sublattice moments out of their initially collinear antiparallel configuration. The bending starts around 40 T and, from the magnetization behavior above this field value, the strength of the Gd–Fe exchange coupling J_{GdFe}/k_B has been determined to be −8.5 K. This value is discussed and is compared with earlier results on the RFe_6Ga_6 compounds with R = Dy, Ho, Er and Tm.

Keywords: $GdFe_6Ga_6$; Exchange-coupling constant; High-field magnetization

1. Introduction

The ternary RFe_6Ga_6 compounds crystallize in the tetragonal $ThMn_{12}$ structure in which the rare-earth (R) atoms occupy the 2a sites, Fe atoms the 8f sites and Ga atoms the 8i sites. The 8j site is statistically occupied by an equiatomic mixture of Fe and Ga [1]. The RFe_6Ga_6 compounds in which R is a heavy-rare-earth element are ferrimagnetic and have Curie temperatures around $T_C = 500$ K. At low temperature, they all exhibit easy-plane anisotropy. In a previous investigation [2], the exchange-coupling strength J_{RFe}/k_B between the Fe and the R-sublattice in ferrimagnetic RFe_6Ga_6 compounds has been studied by means of magnetic measurements in applied magnetic fields up to 35 T on single-crystalline fine powders of several of these compounds that were free to orient themselves in the applied field. This so-called free-powder technique has been intensively applied to determine the intersublattice-coupling strength in many series of ferrimagnetic R (heavy rare earth)–T (transition metal) compounds [3]. From the results reported for the Gd compound in Ref. [2], it is clear that the highest field of 35 T, available in that study, is below the critical field strength necessary to break the collinear ferrimagnetic configuration of the Gd and Fe-sublattice moments and to make them bend towards each other. In the present study, we have extended the range of magnetic-field strengths up to 55 T, enabling to reach the critical field for $GdFe_6Ga_6$.

2. The free-powder method

In a mean-field description, the free energy of a compound consisting of two magnetic sublattices with moments $\vec{M}_R$ and $\vec{M}_T$ is represented by

$$E = E_R^{an} + E_T^{an} + n_{RT}\vec{M}_R\vec{M}_T - (\vec{M}_R + \vec{M}_T)\vec{B} \qquad (1)$$

The first two terms represent the anisotropy energies of the R- and T-sublattices, with sublattice moments, respectively. The third term describes the 3d–4f exchange interaction and contains the intersublattice molecular-field coefficient n_{RT}. The last term in Eq. (1) represents the Zeeman energy in an applied field B.

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In general, at low temperatures, the R-sublattice anisotropy strongly dominates the T-sublattice anisotropy in R–T compounds. Therefore, usually the approximation is justified that the T-sublattice anisotropy can be neglected. Under these circumstances, in the magnetization process of a single crystal or a powder consisting of single-crystalline particles that can freely rotate in the applied field, the R-sublattice will always be in its easy direction, for all possible moment orientations of the two sublattices. In this simple case, no change in anisotropy energy is involved in the magnetization process and equation (1) can be simplified to

$$E = n_{\mathrm{RT}} M_{\mathrm{R}} M_{\mathrm{T}} \cos\alpha - MB \tag{2}$$

with M given by

$$M = \sqrt{M_{\mathrm{R}}^2 + M_{\mathrm{T}}^2 + 2M_{\mathrm{R}}M_{\mathrm{T}}\cos\alpha} \tag{3}$$

The equilibrium directions of $\vec{M}_{\mathrm{R}}$ and $\vec{M}_{\mathrm{T}}$ can be determined by minimizing the free energy in Eq. (2) with respect to α. For ferrimagnetic compounds, at low fields, the configuration of the moments of the two sublattices is perfectly antiparallel and the magnetization is $M = |M_{\mathrm{T}} - M_{\mathrm{R}}|$. Above a first critical field, $B_{\mathrm{cr},1} = n_{\mathrm{RT}}|M_{\mathrm{T}} - M_{\mathrm{R}}|$, the sublattice moments start to bend towards each other and the magnetization is represented by

$$M = n_{\mathrm{RT}}^{-1} B \tag{4}$$

This relationship allows for a straightforward determination of the parameter n_{RT}. Beyond a second critical field, which usually is too high to be experimentally accomplished, $B_{\mathrm{cr},2} = n_{\mathrm{RT}}|M_{\mathrm{T}} + M_{\mathrm{R}}|$, forced ferromagnetic alignment of the two sublattice moments is reached, corresponding to a magnetization equal to $M = M_{\mathrm{T}} + M_{\mathrm{R}}$.

3. Experimental

The compound $\mathrm{GdFe_6Ga_6}$ has been prepared by arc melting and subsequent annealing for 2 weeks at 1300 K, i.e. in the same way as the $\mathrm{RFe_6Ga_6}$ compounds with R = Dy, Ho, Er and Tm that have been reported in Ref. [2]. X-ray diffraction showed the sample to consist mainly of the $\mathrm{RFe_6Ga_6}$ phase with the tetragonal $\mathrm{ThMn_{12}}$-type of crystal structure (space group *I*4/*mmm*, no. 139). The magnetization was measured at 4.2 K in fields up to 5 T in a SQUID magnetometer and at 4.2 K in pulsed fields up to 55 T with pulse duration of 20 ms in KYOKUGEN (the Research Center for Materials Science at Extreme Conditions) at Osaka University. The magnetization measurements were performed on free-powder particles put loosely in the sample holder so that they could orient themselves into the equilibrium direction in the applied field. The powders consist of sieved particles with size smaller than 20 μm in order to ensure single crystallinity.

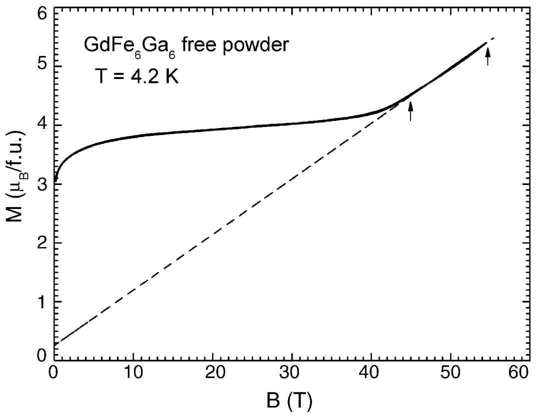

Fig. 1. Magnetization of a free powder of $\mathrm{GdFe_6Ga_6}$ at 4.2 K in applied fields up to 55 T. The dashed line corresponds to the slope of the high-field part, as explained in the text.

4. Results and discussion

Fig. 1 shows the magnetization at 4.2 K of a free-powder sample of $\mathrm{GdFe_6Ga_6}$ measured in fields up to 55 T. The spontaneous magnetization, determined by extrapolating the linear part of the magnetization in the ferrimagnetic state between 10 and 30 T to zero field, equals about 3.7 μ_{B}/f.u. If we assume the Gd magnetic moment in $\mathrm{GdFe_6Ga_6}$ to be equal to the free-ion value of 7 μ_{B} and to be aligned fully antiparallel with the Fe-sublattice moment, this leads to about 10.6 μ_{B} for the Fe-sublattice magnetic moment. Around 40 T, the magnetization starts to exhibit upward curvature, followed by a linear part between 45 and 55 T that satisfies $M = 0.094B + 0.28$, represented by the dashed line in Fig. 1. By means of the relation $M = n_{\mathrm{RT}}^{-1}B$, derived in the previous section, the molecular-field coefficient is found to be $n_{\mathrm{GdFe}} = 10.6$ T f.u./μ_{B}. The small intercept with the magnetization axis of 0.28 μ_{B}/f.u. is not intrinsic. It should be attributed to magnetically saturated impurity phase or to experimental uncertainty in the determination of the magnetic isotherm.

The molecular-field coefficient n_{RT} is related to the exchange-coupling constant J_{RT}, appearing in the interaction Hamiltonian, $H = -\sum 2J_{\mathrm{RT}}\vec{S}_{\mathrm{R}}\vec{S}_{\mathrm{T}}$ by [3]:

$$n_{\mathrm{RT}} = \frac{J_{\mathrm{RT}} Z_{\mathrm{RT}} (g_{\mathrm{R}} - 1)}{N_{\mathrm{T}} \mu_{\mathrm{B}}^2 g_{\mathrm{R}}} \tag{5}$$

here, g_{R} is the Landé factor, Z_{RT} the number of nearest T neighbors to an R atom, for which $Z_{\mathrm{RT}} = 10$, i.e. half of the number of nearest T neighbors in $\mathrm{RT_{12}}$ (equal to 20), has been taken, N_{T} the number of T atoms per formula unit, being 6 for $\mathrm{RFe_6Ga_6}$ compounds.

The values of the molecular-field coefficients n_{RT} and the exchange-coupling parameters J_{RT} for the presently investigated compound $\mathrm{GdFe_6Ga_6}$ are given in Table 1, together with the results on the compounds with R = Dy, Ho, Er and Tm, as reported in Ref. [2]. It can be seen that the strength of the R–Fe coupling does not change monotonously with

Table 1
Intersublattice-coupling constants and exchange-interaction parameters of RFe_6Ga_6 compounds as derived from free-powder high-field magnetization measurements

R	$g_R J$ (μ_B)	$g_R/(g_R - 1)$	n_{RFe} (T f.u./μ_B)	$-J_{RFe}/k_B$ (K)
Gd	7	2	10.6	8.5
Dy	10	4	4.0	6.5
Ho	10	5	3.8	7.7
Er	9	6	3.1	7.4
Tm	7	7	2.2	6.2

The values for the compounds with R = Dy, Ho, Er and Tm are from Ref. [2].

the atomic number Z of the R element. In this respect, it should be remarked that it is difficult to estimate the accuracy of the presented J_{RFe} values. One reason is that it is impossible to precisely control the mass during the melting procedure, so that the eventual composition may differ from the intended stoichiometric composition. Deviations from stoichiometry will have direct, but unpredictable, influence on the magnetization and, consequently, on the value of J_{RFe}. Furthermore and less important, for the free-powder magnetization measurements, high magnetic fields are requested in which magnetization measurements are understandably not equally accurate as low-field measurements. In particular, in the Amsterdam high-field installation, the magnetization measured in equivalent field pulses may differ by a small contribution, which is proportional to the field. Although this uncertainty is small, it hampers a determination of the magnetic susceptibility with an accuracy better than about 1%.

It has been reported by Beloritzky et al. [4] that in a given series of R–T compounds, the value of the R–T exchange-coupling parameter J_{RT} decreases with increasing atomic number Z of the R element. This dependence was explained in terms of an increasing 4f–5d distance with increasing Z and, therefore, a decreasing 4f–5d interaction. The values for the exchange-coupling strength J_{RFe} in the RFe_6Ga_6 series, as presented in Table 1, indeed exhibit the expected decrease of J_{RFe} with increasing Z. The fact that the decrease is not monotonous is attributed to the experimental uncertainties and inaccuracies described above.

On the basis of experimental results on $Ho(Fe, V)_{12}$ and $Er(Fe, V)_{12}$ compounds, the values for the exchange coupling J_{RT} in the hypothetical compounds $HoFe_{12}$ and $ErFe_{12}$ have been estimated in Ref. [2] to be $-J_{HoFe}/k_B = 7$ K and $-J_{ErFe}/k_B = 6$ K. The results in Table 1 indicate that the strength of exchange coupling in the corresponding RFe_6Ga_6 compounds is of very similar magnitude.

Acknowledgments

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