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The anisotropic Dy–Fe exchange interaction in $\mathbb{D}y(Fe_{11}Ti)$

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Abstract. The values of the isotropic and anisotropic Dy–Fe exchange interactions and the CEF parameters for $Dy(Fe_{11}Ti)$ are evaluated from the magnetization curves along the crystal axes measured on the single crystal in the range of temperatures between 4.2 and 300 K. The anisotropic exchange interaction is two orders smaller in magnitude than the isotropic exchange interaction but is comparable with the CEF interaction and apparently affects the magnetization processes.

1. Introduction

Among the $R(Fe_{11}Ti)$ (R≡rare earth) compounds, a more detailed experimental investigation of the magnetization curves has been reported for a single crystal of $Dy(Fe_{11}Ti)$ [1]. In the same work, the curves along the crystal axes at different temperatures have been fitted by calculations based on the single-ion model, in which the Dy–Fe exchange interaction, as usual, has been taken as isotropic. For a satisfactory fit of the calculation, Hu *et al* [1] were obliged to assume that the absolute value of the main crystalline-electric-field (CEF) parameter A_{20} should decrease by 30% on increase in temperature from 0 to 300 K, which seems to be unreasonably large.

In this connection, it is noted that the value of the magnetocrystalline anisotropy (MCA) constant for $Y(Fe_{11}Ti)$ and $Lu(Fe_{11}Ti)$ is as large as 2.1 K per Fe atom at 4.2 K [2, 3]. Such a large MCA reveals that an appreciable part of the orbital angular moments of Fe atoms remains at some crystallographic sites, and so it is expected that the anisotropy of the R–Fe exchange interaction and the magnetic moments of the Fe atoms in a $R(Fe_{11}Ti)$ compound would not be so small that they can be neglected. Anisotropy of the magnetic moments of the Fe atoms, indeed, has been observed for a number of $R(Fe_{11}Ti)$ compounds [2]. The observation of anisotropy of the exchange interaction is not so straightforward.

This paper shows that the difficulty of the unreasonably large variation in the fitted A_{20} with temperature met in the above-mentioned work can be overcome by taking into account the anisotropy of the Dy–Fe exchange interaction.

2. Method of analysis

$R(Fe_{11}Ti)$ has a $ThMn_{12}$-type tetragonal crystal structure belonging to the space group $I4/mmm$, in which there is one rare-earth ion site with the point symmetry D_{4h}. The Hamiltonian of the rare-earth ion consists of the R–Fe exchange interaction, the CEF interaction and the Zeeman energy, i.e.

$$\mathcal{H} = \mathcal{H}_{ex} + \mathcal{H}_{CEF} + \mu_B(L + 2S) \cdot H. \tag{1}$$

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In the coordinate system with the x and z axes along the [100] and [001] axes, respectively, the R–Fe exchange interaction in the general form [4] and the CEF interaction can be expressed as

$$\mathcal{H}_{ex} = 2\mu_B \boldsymbol{S} \cdot n_{RFe} \boldsymbol{M}_{Fe}[1 + p\sin^2(\theta_{Fe})] \tag{2}$$

$$\mathcal{H}_{CEF} = \sum_{n=2,4,6} A_{n0} \sum_j C_{n0}(\theta_j, \varphi_j) + \sum_{n=4,6} A_{n4} \sum_j [C_{n4}(\theta_j, \varphi_j) + C_{n-4}(\theta_j, \varphi_j)]. \tag{3}$$

Here

$$C_{nm}(\theta_j, \varphi_j) = [4\pi/(2n+1)]^{1/2} Y_{nm}(\theta_j, \varphi_j) \tag{4}$$

$Y_{nm}(\theta_j, \varphi_j)$ are the spherical harmonics, θ_j and φ_j are the polar and azimuthal angles of the position vector of the jth 4f electron, n_{RFe} is the molecular-field coefficient, $\boldsymbol{M}_{Fe}$ and θ_{Fe} are the magnetic moment of the Fe sublattice and the angle between $\boldsymbol{M}_{Fe}$ and the c axis, respectively, and p is the anisotropy of the R–Fe exchange interaction. The rare-earth ion is assumed to be triply ionized. The matrix elements of equation (1) are calculated using the irreducible-tensor-operator technique [5]. For a given applied field $\boldsymbol{H}$ and a given direction of $\boldsymbol{M}_{Fe}$, the eigenvalues E_i and eigenfunctions $|n_i\rangle (i = 1, 2 \ldots, 2J+1)$ are obtained by diagonalizing the $(2J+1) \times (2J+1)$ matrix. The free energy for $R(Fe_{11}Te)$ is given by

$$F(\boldsymbol{H}, \boldsymbol{M}_{Fe}, T) = -kT \ln Z + K_1(T)\sin^2\theta_{Fe} - \boldsymbol{M}_{Fe}(T) \cdot \boldsymbol{H} \tag{5}$$

where

$$Z = \sum_i \exp(-E_i/kT) \tag{6}$$

and K_1 is the MCA constant of the Fe sublattice. $M_{Fe}(T)/M_{Fe}(0\text{ K})$ and $K_1(T)/K_1(0\text{ K})$ with $M_{Fe}(0\text{ K}) = 20.4\mu_B$ FU^{-1} and $K_1(0\text{ K}) = 23.3$ K FU^{-1} ($1\text{ K} = 1.38 \times 10^{-23}$ J) were taken to be the same as the corresponding values for $Lu(Fe_{11}Ti)$ [3]. The equilibrium direction of $\boldsymbol{M}_{Fe}$ is determined from minimization of $F(\boldsymbol{H}, \boldsymbol{M}_{Fe}, T)$, and the magnetic moments of the rare-earth ion and of $R(Fe_{11}Ti)$ are given by

$$\boldsymbol{M}_R(T) = -\mu_B \sum_i \langle n_i|\boldsymbol{L} + 2\boldsymbol{S}|n_i\rangle \exp(-E_i/kT)/Z \tag{7}$$

$$\boldsymbol{M}(T) = \boldsymbol{M}_R(T) + \boldsymbol{M}_{Fe}(T). \tag{8}$$

The values of the exchange-field parameters n_{RFe} and p and the CEF parameters A_{20}, A_{40}, A_{44}, A_{60} and A_{64} were evaluated from a fit of the calculations to the experimental data. The values of the parameters were also evaluated by neglecting the anisotropy of the Dy–Fe exchange interaction (i.e. for $p = 0$) for reference. A_{20} was allowed to vary with temperature in both cases, while the higher-order CEF parameters are treated as invariant since the CEF interaction terms are less important and become increasingly insignificant with increase in temperature.

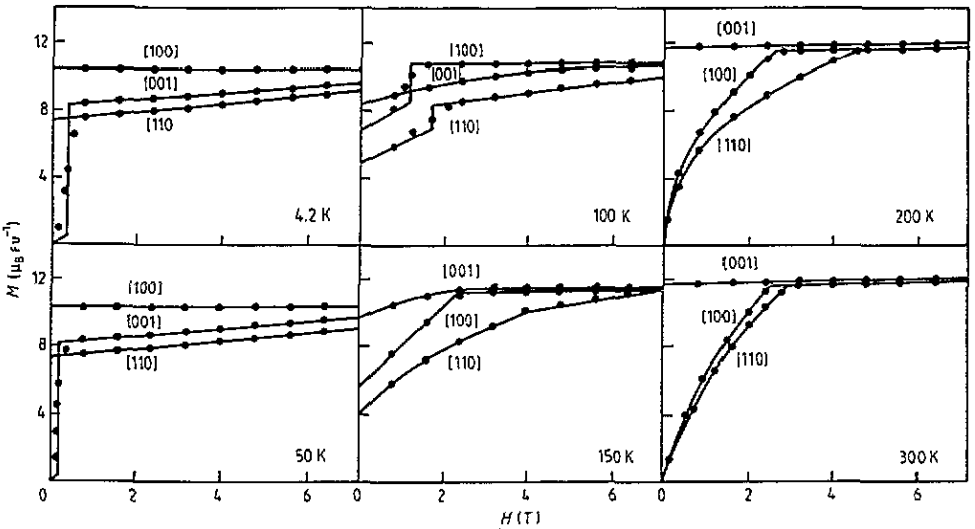

Figure 1. Magnetization curves for $Dy(Fe_{11}Ti)$ in the range of temperatures between 4.2 and 300 K: ●, experimental data from Hu *et al* [1]; ——, calculations

3. Results and discussion

Figures 1 and 2 show the comparison of the calculations with the experimental data for $Dy(Fe_{11}Ti)$. The symbols represent the experimental data, and the full curves the calculations. The calculations with $p = 0$ simulate the experimental data as well. Figure 1 shows the magnetization curves along the [100], [110] and [001] axes at 4.2, 50, 100, 150, 200 and 300 K. Figure 2 shows the temperature dependences of the components of spontaneous magnetization M_s along the [100] and [001] axes, and of the angle between the direction of M_S and the [001] axis. The experimental data in the figures are from the work of Hu *et al* [1]. Table 1 lists the fitted values of the molecular-field coefficient n_{RFe} and the CEF parameters A_{nm} evaluated by taking into account ($p \neq 0$) or by neglecting ($p = 0$) the anisotropy of the Dy–Fe exchange interaction. Those fitted by Hu *et al* [1] verified in this work are also included in the table. It reveals that, for $p = 0$, a series of the sets of n_{RFe} and A_{nm} in which $A_{20}(0\ K)$ also satisfies the condition $-50\ K \lesssim A_{20}(0\ K) \lesssim -25\ K$ can also simulate the experimental curves if A_{20} is allowed to vary with temperature. The decrease in $|A_{20}(T)|$ on increase in temperature is larger for sets with larger values of $|A_{20}(0\ K)|$ such as $|A_{20}(300\ K) - A_{20}(0\ K)|/|A_{20}(0\ K)| = 24\%$, 28% and 30% for the sets with $A_{20}(0\ K) = -27$ K, -36 K and -47 K (see the last three rows in the table), respectively. In any case, as the decreases are unreasonably large, it is concluded that the experimental curves cannot be reproduced reasonably by neglecting the anisotropy of the Dy–Fe exchange interaction. It can be seen from the table that the anisotropic exchange interaction is two orders smaller in magnitude than the isotropic exchange interaction, as for $Gd_xY_{1-x}Co_5$ and $R_2Co_{14}B$ (R=Pr, Nd or Gd) [4]. The fitted values, especially of $A_{20}(0\ K)$, are affected by taking into account the anisotropy of the exchange interaction. Figure 3 shows the variation in the fitted A_{20} with temperature. The broken curve is obtained by neglecting the anisotropy of the exchange interaction with the set of parameters $p = 0$, $n_{RFe} = 7.5$ K FU μ_B^{-2}, $A_{20}(0\ K) = -27$ K, $A_{40} = -110$ K, $A_{44} = 160$ K, $A_{60} = 140$ K and $A_{64} = 15$ K, which represents the most slowly decreasing curve, on

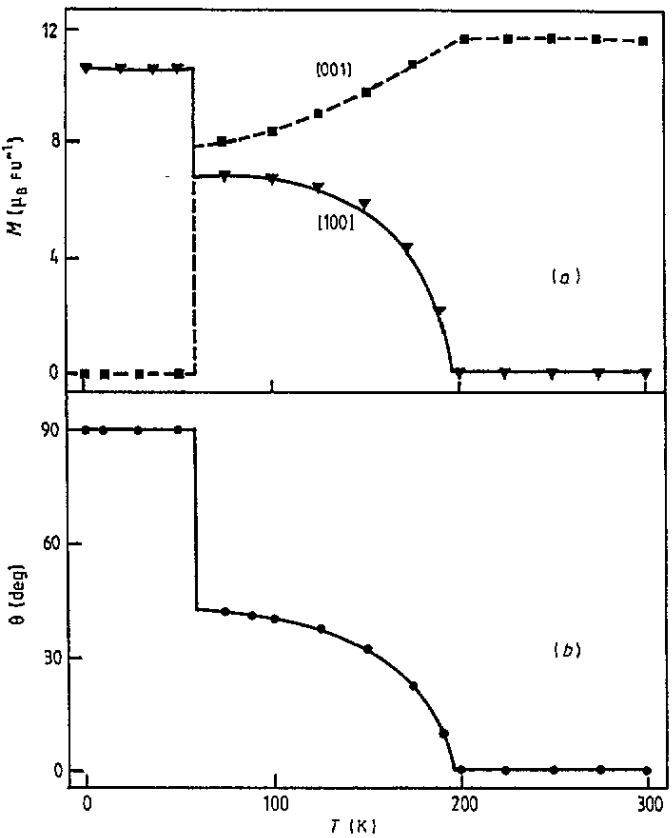

Figure 2. Temperature dependence (*a*), of the components of spontaneous magnetization along the [001] and [100] axes and (*b*) of the angle between the direction of the spontaneous magnetization and the [001] axis for $Dy(Fe_{11}Ti)$: ●, ■, ▼, experimental date from Hu *et al* [1]; ——, calculations

increase in temperature of the sets with $p = 0$ (a decrease of 24% at 300 K). The full curve is obtained by taking into account the anisotropy ($p = -0.0066$). When the anisotropy of the Dy–Fe exchange interaction is taken into account, the value of $|A_{20}(T)|$ decreases by 7% on increase in temperature from 0 to 300 K, which would be reasonable. In fact, Czjzek *et al* [6] observed a variation in the electric field gradients at the Gd ion sites in $Gd_2Fe_{14}B$, which are proportional to A_{20}, by ±5% on increase in temperature from 0 to 120 K. The large decreases in $A_{20}(T)$ at higher temperatures for $p = 0$ can be explained as follows. Although the anisotropic exchange interaction is much smaller than the isotropic exchange interaction, it is still comparable with the CEF interaction in strength. The θ_{Fe} dependence of the exchange interaction, $2\mu_B S \cdot n_{RFe} M_{Fe} p \sin^2(\theta_{Fe}) \simeq 2\mu_B S n_{RFe} M_{Fe} p \sin^2(\theta_{Fe})$, is similar to that of the CEF interaction term of the coefficient A_{20} [7],

$$\left\{\left[-\left(\frac{3}{4}A_{20}\alpha_j\right)\sum_{J_z}[3J_z^2 - J(J+1)]\exp\left(\frac{-2\mu_B(g_J-1)J_z n_{RFe}M_{Fe}}{kT}\right)\right]\right.$$
$$\left.\times\left[\sum_{J_z}\exp\left(\frac{-2\mu_B(g_J-1)J_z n_{RFe}M_{Fe}}{kT}\right)\right]^{-1}\right\}\sin^2\theta_R.$$

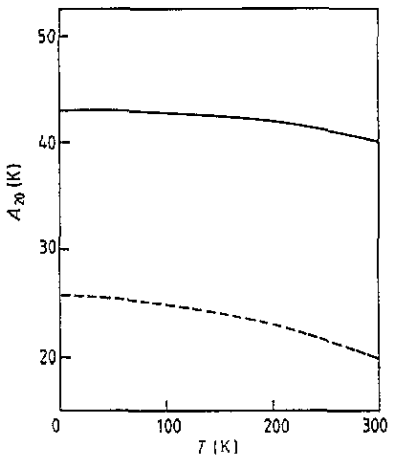

Figure 3. Temperature dependences of the fitted A_{20} for $Dy(Fe_{11}Ti)$: ——, line deduced by taking into account the anisotropy of the Dy–Fe exchange interaction ($p = -0.0066$): – – –, line deduced by neglecting the anisotropy of the Dy–Fe exchange interaction with the set of parameters $p = 0$, $n_{RFe} = -7.5\mu_B^{-2}$ K FU, $A_{20}(0\text{ K}) = -27$ K, $A_{40} = -110$ K, $A_{44} = 160$ K, $A_{60} = 140$ K and $A_{64} = 15$ K.

Table 1. The fitted values of the Dy–Fe exchange interaction and CEF parameters for $Dy(Fe_{11})Ti$.

n_{RFe} (K FU μ_B^{-2})	p	A_{20}(0 K) (K)	A_{40} (K)	A_{44} (K)	A_{60} (K)	A_{64} (K)	Reference
−7.5	−0.0066	−43	−100	135	140	15	This work
−7.5	0	−27	−110	160	140	15	This work
−6.9	0	−36	−120	152	170	10	This work
−5.9	0	−47	−130	150	209	5	[1], verified by this work

The anisotropy of the exchange interaction affects the characteristics of the magnetization curves mainly similarly to the way in which the A_{20} CEF interaction does and so affects the fitted value of A_{20} mostly when the anisotropy is taken into account. Since the anisotropic exchange interaction decreases apparently with increase in temperature in proportion to $\langle S_R(T)\rangle M_{Fe}(T)$, neglect of the interaction causes the unreasonably large decrease in the fitted parameter $-A_{20}$.

Acknowledgments

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