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Analysis of the Magnetic Properties of $RFe_{11}Ti$ and $RFe_{11}TiH$ (R=Tb,Ho)

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The values of crystalline-electric-field parameters A_{nm} for $RFe_{11}TiH_x$ (R=Tb,Ho) (x=0,1) are obtained by fitting calculations to the magnetization curves along the crystal axes at 4.2 K and higher temperatures. The insertion of H element in $RFe_{11}Ti$ significantly affects CEF parameters A_{nm}. By using exchange field $2\mu_B H_{ex}$ derived by inelastic neutron scattering and fitted A_{nm}, the calculations reproduce the experimental curves well.

Key words : crystalline-electric-field, rare-earth 3d metal compound, magnetization curve

1. Introduction

During the last few years, the magnetic properties of the intermetallic compounds $RFe_{11}Ti$ and their hydrides $RFe_{11}TiH$ have been studied extensively [1-8]. Many experimental investigations of the intrinsic magnetic properties of $RFe_{11}TiH_x$ (x=0,1) single crystals have been reported [1-7]. The insertion of hydrogen element in $RFe_{11}Ti$ significantly changes their Curie temperature, saturation magnetization and magnetic anisotropy. Nikitin et al evaluated the values of Ho-Fe exchange field $2\mu_B H_{ex}$ and crystalline-electric-field (CEF) parameters A_{nm} in $HoFe_{11}TiH_x$(x=0,1) by fitting calculations to the magnetization curves along the crystal axes at 4.2 K and concluded that hydrogenation leads to the increase in $2\mu_B H_{ex}$ of about 10% [1]. Inelastic neutron scattering (INS) experiments on $RFe_{11}Ti$ and $RFe_{11}TiN$ (R=Gd,Er) show that insertion of N element leads to the decrease in $2\mu_B H_{ex}$ of about 14% [9]. INS experiments on Gd_2Fe_{17} and $Gd_2Fe_{17}M_x$ (M: C, N, D (deuterium)) also show that insertion of light interstitial element (C, N, D) reduces the Gd-Fe exchange field at Gd site in Gd_2Fe_{17} [10]. In this connection, the insertion of light interstitial elements leads to the enhancement of Curie temperature and the lattice expansion of the compounds.

This work is to evaluate the values of CEF parameters A_{nm} of $RFe_{11}TiH_x$ (R=Tb,Ho) (x=0,1) from the experimental magnetization curves at 4.2 K and higher temperatures on the basis of the single-ion model and analyse the experimental data.

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2. Method of Analysis

$RFe_{11}TiH_x$ (x=0,1) have a $ThMn_{12}$-type tetragonal structure with space group *I4/mmm*. There is one rare-earth ion site with the point symmetry D_{4h}. The Hamiltonian of the rare-earth ion consists of the CEF interaction, the R-Fe exchange interaction and the Zeeman energy, i.e.

$$H=H_{CEF}+2\mu_B\hat{S}\bullet\vec{H}_{ex}+\mu_B(\hat{L}+2\hat{S})\bullet\vec{H}, \quad (1)$$

In the coordinate system with x and z axes along the [100] and [001] axes, respectively, the CEF interaction can be expressed as

$$H_{CEF}=\sum_{n=2,4,6}A_{n0}\sum_j C_{n0}(\theta_j,\varphi_j) + \sum_{n=4,6}A_{n4}\sum_j (C_{n4}(\theta_j,\varphi_j)+C_{n-4}(\theta_j,\varphi_j)), \quad (2)$$

Here

$$C_{nm}(\theta_j,\varphi_j)=[4\pi/(2n+1)]^{1/2}Y_{nm}(\theta_j,\varphi_j), \quad (3)$$

$Y_{nm}(\theta_j,\varphi_j)$ are the spherical harmonics, θ_j and φ_j are the polar and azimuthal angles of the position vector of jth 4f electron. It is assumed that the rare-earth ion is trivalent and $\vec{H}_{ex}$ is proportional to the magnetic moment of the Fe sublattice $\vec{M}_{Fe}$. The matrix elements of Eq. (1)

are calculated by using the irreducible-tensor-operator technique. For a given applied field $\vec{H}$ and a direction of $\vec{H}_{ex}$, the eigenvalues E_i and eigenfunctions $|n_i\rangle$ (i = 1, 2, ···, 2J+1) are obtained by diagonalizing the (2J+1) × (2J+1) matrix. The free energy for $RFe_{11}TiH_x$ (x=0,1) is given by

$$F(\vec{H}, \vec{M}_{Fe}, T)=-kT\ln Z+K_1(T)Sin^2\theta_{Fe}-\vec{M}_{Fe}(T)\cdot\vec{H}, \quad (4)$$

Where

$$Z=\sum_i \exp(-E_i/kT), \quad (5)$$

and $K_1(T)$ is the magnetocrystalline anisotropy constant of the Fe sublattice. $M_{Fe}(T/T_C)/M_{Fe}(0)$ and $K_1(T/T_C)/K_1(0)$ for $RFe_{11}Ti$ and $RFe_{11}TiH$ are taken to be same as those for $LuFe_{11}Ti$ and $LuFe_{11}TiH$, respectively [4]. The value of $M_{Fe}(0)$ is taken to be 20.1 μ_B/*f.u.* for $RFe_{11}Ti$ and 20.6 μ_B/*f.u.* for $RFe_{11}TiH$, respectively. $K_1(0)$=23.6 K/f.u. for $RFe_{11}Ti$ and $K_1(0)$=26.6 K/f.u. for $RFe_{11}TiH$ is same as that for $LuFe_{11}Ti$ and $LuFe_{11}TiH$, respectively [4]. The equilibrium direction of $\vec{M}_{Fe}$ is determined from minimization of $F(\vec{H}, \vec{M}_{Fe}, T)$, and the magnetic moments of the rare-earth ion and of $RFe_{11}TiH_x$ (x=0,1) are given by

$$\vec{M}_R=-\mu_B\sum_i \langle n_i|\vec{L}+2\vec{S}|n_i\rangle\exp(-E_i/kT)/Z, \quad (6)$$

$$\vec{M}=\vec{M}_R+\vec{M}_{Fe}. \quad (7)$$

The value of the exchange field $2\mu_B H_{ex}$ in $RFe_{11}TiH_x$ (R=Tb,Ho) (x=0,1) were estimated as follows. The value of $2\mu_B H_{ex}$(T ≈ 20 K) derived by INS experiment is about 470 K, 343 K and 250 K for $SmFe_{11}Ti$ [11], $GdFe_{11}Ti$ [9] and $ErFe_{11}Ti$ [9]. It is known that the value of $2\mu_B H_{ex}$ decrease monotonically across the rare-earth series from Pr to Er in many Rare-earth-Fe intermetallic compounds [12, 13]. By extrapolating the value for $SmFe_{11}Ti$, $GdFe_{11}Ti$ and $ErFe_{11}Ti$ according such a variation in $2\mu_B H_{ex}$ across the $RFe_{11}Ti$ series, the value of $2\mu_B H_{ex}$(T ≈ 20 K) for $TbFe_{11}Ti$ and $HoFe_{11}Ti$ is estimated to be about 285 K and 260 K, respectively. It is assumed that the decrease of $2\mu_B H_{ex}$ induced by insertion of N element is about five times of the decrease by insertion of H in $RFe_{11}Ti$ as in R_2Fe_{17} compounds [10]. The value of $2\mu_B H_{ex}$(T ≈ 20 K) for $TbFe_{11}TiH$ and $HoFe_{11}TiH$ is estimated to be about 277 K and 253 K.

The values of CEF parameters A_{nm} were evaluated by fitting calculations to the magnetization curves at 4.2 K and higher temperatures for the single crystals of $RFe_{11}TiH_x$ (R=Tb,Ho) (x=0,1).

3. Results and Discussion

Table 1 lists the fitted values of CEF parameters A_{nm}. It can be seen that the value of A_{20} of $RFe_{11}TiH$ increase about 77%~113% towards negative value compared with that of corresponding $RFe_{11}Ti$. In this connection, a Mössbauer study of ^{155}Gd in $GdFe_{11}TiH_x$ (x=0,1) has shown that hydrogenation leads to about 53% increase of the electric field gradients at Gd ion site, which are proportional to A_{20} [8]. Hydrogenation has also significant effect on the high-order CEF parameters A_{nm}. The significant changes of A_{nm} cause the change of magnetic aniso-

Table 1. The fitted values of CEF parameters A_{nm}

	A_{20}	A_{40}	A_{44}	A_{60}	A_{64}
$TbFe_{11}Ti$	-90 K	-4 K	-100 K	2 K	-87 K
$TbFe_{11}TiH$	-160 K	-110 K	34 K	20 K	-25 K
$HoFe_{11}Ti$	-61 K	-122 K	-176 K	432 K	-13 K
$HoFe_{11}TiH$	-130 K	-85 K	-80 K	62 K	-10 K

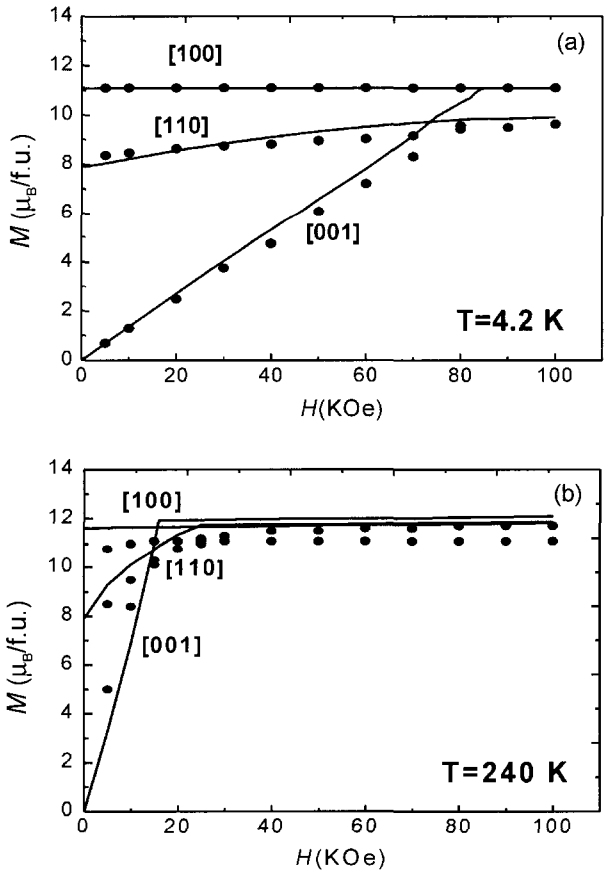

Fig. 1. (a) magnetization curves of $TbFe_{11}Ti$ at 4.2 K: ● experimental data from [2]; —— calculation. (b) magnetization curves of $TbFe_{11}Ti$ at 240 K: ● experimental data from [6]; —— calculation.

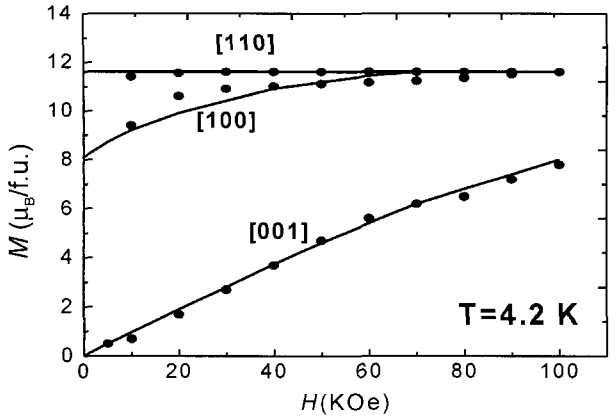

Fig. 2. magnetization curves of $TbFe_{11}TiH$ at 4.2 K: ● experimental data from [2]; —— calculation.

tropy. For example, the change of the easy magnetization direction induced by insertion of H for $TbFe_{11}Ti$, which is from the [100] to the [110] axis, mainly originates from a sign change of A_{44}.

Fig. 1 to 4 show the comparison of the calculations with the experimental data. The symbols represent the experimental data, and the full curves the calculations. Fig. 1(a) and 1(b) show the magnetization curves along the [100], [110] and [001] axes at 4.2 K and 240 K for $TbFe_{11}Ti$. Fig. 2 shows the magnetization curves along the [100], [110] and [001] axes at 4.2 K for $TbFe_{11}TiH$. Fig. 3(a) and 3(b) show the magnetization curves along the [100], [110] and [001] axes at 4.2 K and 290 K for $HoFe_{11}Ti$. Fig. 4(a) and 4(b) show the magnetization curves along the [100], [110] and [001] axes at 4.2 K and 300 K for $HoFe_{11}TiH$. At 4.2 K, a field-induced first-order magnetization process (FOMP) in the [001] direction was found by our calculation for $HoFe_{11}TiH$. It may be as in the FOMP in the $DyFe_{11}Ti$ [14] that a continuous increase of the magnetization instead of a discontinuous jump in the experimental curves is observed for magnetic field around the critical field. The predicted FOMP by calculation need to be confirmed by the singular-point-detection technique in $HoFe_{11}TiH$.

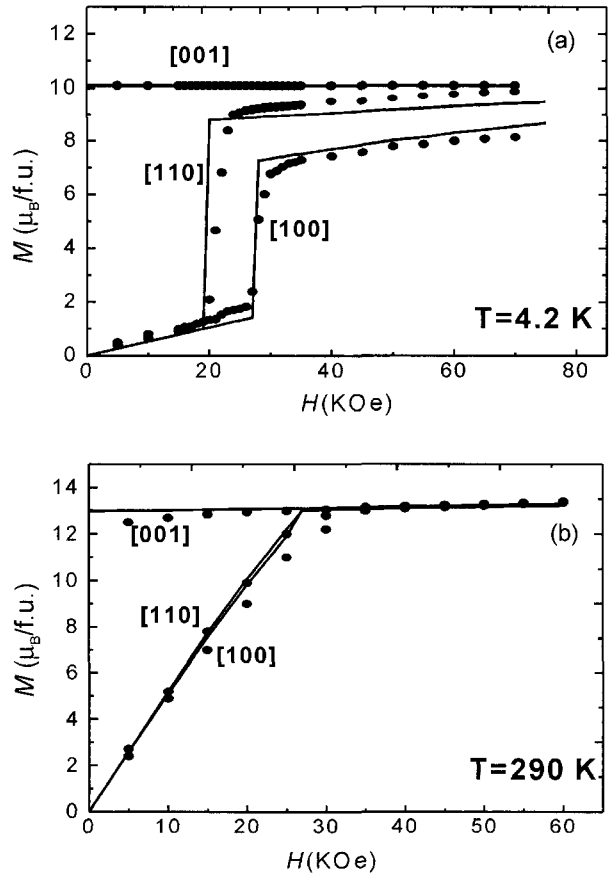

Fig. 3. (a) magnetization curves of $HoFe_{11}Ti$ at 4.2 K: ● experimental data from [1]; —— calculation. (b) magnetization curves of $HoFe_{11}Ti$ at 290 K: ● experimental data from [6]; —— calculation.

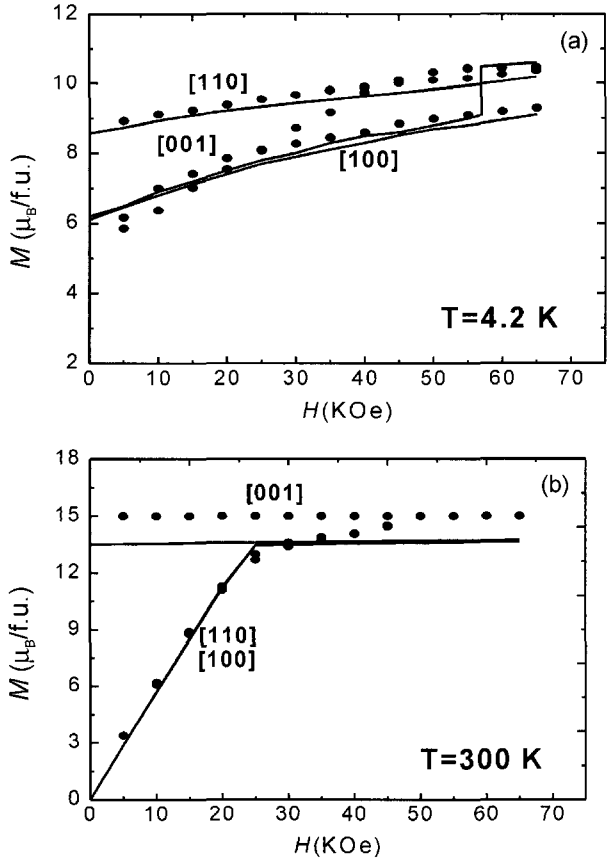

Fig. 4. (a) magnetization curves of $HoFe_{11}TiH$ at 4.2 K: ● experimental data from [1]; —— calculation. (b) magnetization curves of $HoFe_{11}TiH$ at 300 K: ● experimental data from [7]; —— calculation.

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