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Abstract
The pronounced anisotropy of the magnetization caused by the Ho^{3+} ions in the ferrimagnetic holmium iron garnet has been investigated based on quantum theory. The strong anisotropy of the magnetization of the Ho^{3+} ions originates mainly from the effect of the crystal field upon the Ho^{3+} ions and the anisotropic Ho^{3+}–Fe^{3+} superexchange interaction. Following the expression of the Yb^{3+}–Fe^{3+} exchange interaction used by Alben, the anisotropy of the Ho^{3+}–Fe^{3+} exchange interaction is defined by three principal values of the exchange tensor G. Because the six Ho^{3+} sublattices are magnetically non-equivalent, we calculate the magnetic quantities of the Ho^{3+} at the six sublattices and compare the average value of the so-obtained six quantities with the measured values. The calculated results are in good agreement with experiments. An interpretation on the anisotropy of the magnetic properties of HoIG is given.

1. Introduction

In recent years, much attention has been paid to rare-earth-substituted yttrium iron garnets $\{RE_x Y_{3-x}\}[Fe2](Fe_3)O_{12}$ (RE:YIG, RE^{3+} represents trivalent rare earth ions) mainly because RE^{3+} ions cause a very strong enhancement of the Faraday rotation in these iron garnets [1–5]. The heavy rare earth ions substituted in iron garnet single crystals induce not only a strong increase of the Faraday rotation but also an obvious anisotropy [6–8]. Fadly *et al* [7] measured the spontaneous magnetization M_s and the spontaneous FR Φ_s in HoIG single crystals in 1979 and Ostoréro *et al* [8] measured these quantities in HoIG:Al single crystals in 1994. From the temperature dependence of these quantities, they discovered that both the magnetization and the MO effects in HoIG and HoIG:Al obviously present anisotropy.

Rare earth iron garnets of formula $RE_3Fe_5O_{12}$ (REIG) belong to the space group $Ia3d$ [9]. The magnetic ions are distributed over three crystallographic sites with sublattice magnetizations M_a (octahedral site $16a$: [Fe^{3+}]), M_d [tetrahedral site $24d$: (Fe^{3+})] and M_c (dodecahedral site $24c$: {Re^{3+}}). The Fe[a]–Fe[d] superexchange interaction determines the Néel temperature ($T_N = 560$ K) for all REIGs. Furthermore, in heavy rare earth iron garnets, there

is a compensation temperature T_{comp}. At T_{comp} the bulk magnetization vanishes. In pure HoIG, $T_{comp} = 137$ K [8]. The magnetic behavior of the rare earth ions is mainly determined by the c–d superexchange interaction [10]. In general, the rare earth ions, which occupy the $24c$ crystal sites, are equivalent in the crystallography. However, an external magnetic field or an exchange field will decrease the symmetry and the $24c$ lattice is divided into six magnetically non-equivalent sublattices denoted by $C_1C_2C_3$ and $C_1'C_2'C_3'$ [11]. Hence, the magnetic quantity of the Ho^{3+} ions in various sublattices has to be calculated. Because the Ho^{3+} ions distribute uniformly among the six sublattices of the $Ho_3Fe_5O_{12}$ crystal, we calculate the simple arithmetic average values of the magnetic quantity of the Ho^{3+} ion in the six sublattices and compare the average values to the measured results. In the following, we report only the average values.

2. Approach

2.1. The CF-split energy levels and the corresponding wavefunctions

The magnetic behavior of the rare earth ions in the rare earth iron garnets arises from the split levels of the ground

Table 1. The values of the parameters of the CF upon the 4f electrons of the Ho^{3+} ions in HoIG (in cm^{-1}).

$A_{20}\langle r^2\rangle$	$A_{2\pm2}\langle r^2\rangle$	$A_{40}\langle r^4\rangle$	$A_{4\pm2}\langle r^4\rangle$	$A_{4\pm4}\langle r^4\rangle$	$A_{60}\langle r^6\rangle$	$A_{6\pm2}\langle r^6\rangle$	$A_{6\pm4}\langle r^6\rangle$	$A_{6\pm6}\langle r^6\rangle$
−331.5	239.2	−2708.2	−845.6	633.0	135.2	−674.4	886.7	33.7

Table 2. The calculated and measured [12] energies of the CF-split levels of the ground multiplet $(4f^{10})^5I_8$ of the Ho^{3+} ions in HoIG (in cm^{-1}).

E_c(cal)	0	28.8	74.9	90.5	107.2	114.4	154.3	160.9	402.1
E_c(exp)	0	29	66	78	95	110	145	160	402
E_c(cal)	419.6	428.2	436.0	542.2	546.2	553.5	707.7	709.4
E_c(exp)	413	424.5	436.5	461	487

configuration [2]. The ground term $4f^{10}(^5I)$ of free Ho^{3+} ions contains five multiplets $^5I_8$, $^5I_7$, $^5I_6$, $^5I_5$ and $^5I_4$, the $^5I_8$ multiplet being the ground multiplet. According to Johnson *et al* [12], in HoIG, the energy of the lowest crystal field (CF)-split level of the first excited multiplet $^5I_7$ is 5198 cm^{-1}, 4711 cm^{-1} higher than the highest CF-split level of the $^5I_8$ multiplet (487 cm^{-1}). The energy gap between them is so large that the CF and superexchange interaction induced mixing of the ground multiplet with $^5I_7$ and other higher-lying multiplets can be completely neglected. Therefore the spontaneous magnetization of the Ho^{3+} ions is simply determined by the successive splitting of the multiplet $^5I_8$ caused by the CF and Ho^{3+}–Fe^{3+} superexchange interaction. Because the superexchange interaction is less than the CF effect, the perturbation calculation has to be carried out with the following order of priorities: CF interaction and superexchange interaction.

The perturbation Hamiltonian upon the Ho^{3+} ions is

$$\hat{H} = \hat{H}_{CF} + \hat{H}_{ex}. \tag{1}$$

Here $\hat{H}_{ex}$ is the superexchange interaction Hamiltonian and will be discussed in section 2.2. $\hat{H}_{CF}$ is the crystal field Hamiltonian which can be expressed as

$$\hat{H}_{CF} = \sum_{k,q} A_{kq} r^k Y_{kq}(\theta, \varphi). \tag{2}$$

A_{kq} is the CF parameter and $Y_{kq}(\theta, \varphi)$ is the spherical harmonic. Because the local environment of the six magnetically non-equivalent Ho sites is identical, the CF upon the Ho sites in the six sublattices is the same.

The choice of the proper set of CF parameters is a crucial step. Following the previous works on other R:YIG (Ce:YIG, Pr:IG, Nd:IG and so on) [1–3], these parameters are determined according to the values of the CF parameters for the Sm^{3+} in SmIG deduced from the magnetic phase transition experiments by Nekvasil *et al* [13] and the ratios of the values of $\langle r^2\rangle$, $\langle r^4\rangle$, $\langle r^6\rangle$ of different rare earth ions. Table 1 shows the non-zero parameters we used.

The CF-split energy levels and the corresponding wavefunctions are obtained by solving the following secular equation:

$$\|\langle J J_z|\hat{H}_{CF}|J'J'_z\rangle - E_{CF}\delta_{JJ'}\delta_{J_zJ'_z}\| = 0 \tag{3}$$

where $|J J_z\rangle$ is the wavefunction of free Ho^{3+} ions in the coupled representation and E_{CF} is the energy of the CF-split level. The calculated levels are given in table 2, taking the energy of the lowest CF-split $^5I_8$ level to be zero. The experimental values [12] are given in table 2 as well.

2.2. The energies and wavefunctions of the CF-split and superexchange interaction mixed levels

$\hat{H}_{ex}$ can be expressed as [14]

$$\hat{H}_{ex} = 2\mu_B \boldsymbol{H}_{exch} \cdot \hat{\boldsymbol{S}}. \tag{4}$$

Here, μ_B is the Bohr magneton, $\hat{\boldsymbol{S}}$ is the total spin angular momentum operator of a Ho^{3+} ion and $\boldsymbol{H}_{exch}$ is the exchange field. The Fe^{3+} ions, located on a and d sites, are strongly coupled due to the Fe^{3+}–Fe^{3+} superexchange interaction (T_N = 560 K) and, at low temperature, effectively form a single rigid saturated sublattice. The state of any given Fe^{3+} ion is that of the sublattice as a whole. The Fe^{3+}–Ho^{3+} interaction is relatively weak, so the back reaction of Ho^{3+} ions on the Fe^{3+} ions is negligible. Furthermore, the superexchange interaction between the different Ho^{3+} ions is very much weaker than the Fe^{3+}–Ho^{3+} interaction and generally is neglected in the analysis. So, the exchange field can be expressed as

$$\boldsymbol{H}_{exch} = n_0 (1 + \gamma T) \boldsymbol{M}_{Fe}. \tag{5}$$

Here n_0 is the exchange-field coefficient and γ is a coefficient that describes its thermal evolution; M_{Fe} is the resultant spontaneous magnetization of the Fe^{3+} sublattices. It should be noted that the exchange-field coefficient is different from the classical molecular-field coefficient [2]. In the molecular-field approximation, the Zeeman Hamiltonian is written as $\mu_B \boldsymbol{H}_m \cdot (2\boldsymbol{S} + \boldsymbol{L})$ and the molecular field is $\boldsymbol{H}_m = n'_0(1 + \gamma' T)\boldsymbol{M}_{Fe}$. From the comparison with equation (5), it can be known that $\boldsymbol{H}_m$, n'_0 and γ' are different from $\boldsymbol{H}_{exch}$, n_0 and γ.

$\boldsymbol{H}_{exch}$ is along the direction of $\boldsymbol{M}_{Fe}$, and in turn along the direction of the applied magnetic field. In the following, for the Ho^{3+} ions at the six sublattices denoted by C_1, C2, C_3 and C'_1, C2′, C'_3, we use the local coordinates expressed by $\boldsymbol{u}, \boldsymbol{v}, \boldsymbol{w}$ (unit vectors along the axes of the local coordinates). The directions of these axes are expressed with the directional numbers with respect to the three edges of the cubic crystal cell. Then the CF parameters in equation (2) are the same for all six Ho sublattices. Therefore, the wavefunctions of the CF-split levels of the Ho^{3+} ions at the six sublattices are the same when they are expressed in local coordinates.

In the local coordinates, for each of the six non-equivalent Ho sites, equation (5) can be expressed as

$$H_{\text{exch}} = n_0(1 + \gamma T)|M_{\text{Fe}}|[\cos(M_{\text{Fe}}, u)u$$
$$+ \cos(M_{\text{Fe}}, v)v + \cos(M_{\text{Fe}}, w)w]. \quad (6)$$

In this formula $\cos(M_{\text{Fe}}, u)$ denotes the cosine of the separation angle between the vectors M_{Fe} and u. It can be seen from equation (6) that, no matter what direction the exchange field is along, the intensity of the Ho^{3+}–Fe^{3+} exchange interaction is identical for the same Ho site (e.g. C_1, etc). In other words, equation (5) presents an isotropic exchange interaction.

Following the expression of the anisotropic Yb^{3+}–Fe^{3+} exchange interaction used in the study of the phase transition in YbIG by Alben [15], we introduce three principal values of the exchange tensor G (G_u, G_v and G_w) to define the anisotropic Ho^{3+}–Fe^{3+} exchange interaction:

$$H_{\text{exch}} = n_0(1 + \gamma T)|M_{\text{Fe}}|\frac{M_{\text{Fe}}}{|M_{\text{Fe}}|} \cdot [G_u uu + G_v vv$$
$$+ G_w ww] \quad (5i)$$

where uu, vv, ww are dyad. Equation (5i) can be expressed further as

$$H_{\text{exch}} = n_0(1 + \gamma T)|M_{\text{Fe}}|[G_u \cos(M_{\text{Fe}}, u)u$$
$$+ G_v \cos(M_{\text{Fe}}, v)v + G_w \cos(M_{\text{Fe}}, w)w]. \quad (6i)$$

It can be seen that, if $G_u = G_v = G_w = 1$, equation (6i) turns back to equation (6), whereas, if G_u, G_v and G_w are not identical, equation (5i) presents an anisotropic exchange interaction. It means that the intensity of the Ho^{3+}–Fe^{3+} exchange interaction upon a Ho^{3+} ion depends on the relative orientation of M_{Fe} to the local coordinates (u, v, w) of the considered Ho^{3+} ion.

Then the energies and wavefunctions of the CF-split and superexchange interaction mixed levels of the ground multiplet of the Ho^{3+} ion can be obtained by solving the following secular equation:

$$\|\langle \psi_{\text{ci}}|\hat{H}_{\text{CF}} + \hat{H}_{\text{ex}}|\psi_{\text{cj}}\rangle - E_g \delta_{ij}\| = 0 \quad (7)$$

where $|\psi_{\text{ci}}\rangle$ and $\langle \psi_{\text{ci}}|\hat{H}_{\text{CF}}|\psi_{\text{ci}}\rangle$ are the eigenwavefunctions and eigenenergies obtained by solving equation (3), respectively.

The principal values of G_x, G_y and G_z of the exchange tensor G used by Alben [15] in the study of YbIG came from near-infrared measurements. The corresponding values for the anisotropic Ho^{3+}–Fe^{3+} exchange interaction in HoIG have not been determined so far. We determine these values by fitting the experimental results of the magnetization of the Ho^{3+} ion in HoIG. The obtained values are 0.7, 1 and 1.3, respectively. The ratio of G_x, G_y and G_z used by Alben [15] is $G_x:G_y:G_z = 1:2.22:2.57$, while our ratio is $G_x:G_y:G_z = 1:1.43:1.86$. It can be seen that the differences between our three values are smaller than those used by Alben [15].

Guillot *et al* [16] showed that both the values of n_0 and γ are temperature-independent. Nevertheless, they also pointed out that the values in the temperature range $T < T_{\text{comp}}$ may be different from those in the temperature range $T > T_{\text{comp}}$. We determined the values of n_0 and γ by fitting the

Table 3. The energies (in cm^{-1}), occupation probabilities (ρ_g), magnetic moment (in μ_{B}/ion) and wavefunctions of the lowest two CF-split and superexchange interaction mixed levels of the ground multiplet at 294 K, the Ho^{3+} ion being at the C_1 site and being magnetized along the [111] direction.

Energy	ρ_g	m	Wavefunction
−328.9371	0.1698	4.2658	$0.0880\lvert 8, 8\rangle + 0.0173\lvert 8, 7\rangle - 0.3048\lvert 8, 6\rangle + 0.0293\lvert 8, 5\rangle - 0.0542\lvert 8, 4\rangle + 0.0327\lvert 8, 3\rangle - 0.2282\lvert 8, 2\rangle + 0.0242\lvert 8, 1\rangle + 0.0001\lvert 8, 0\rangle - 0.0327\lvert 8, -1\rangle + 0.4006\lvert 8, -2\rangle - 0.0614\lvert 8, -3\rangle + 0.0896\lvert 8, -4\rangle - 0.0628\lvert 8, -5\rangle + 0.7737\lvert 8, -6\rangle - 0.0372\lvert 8, -7\rangle - 0.2533\lvert 8, -8\rangle$
−288.6066	0.1394	−2.7192	$0.2562\lvert 8, 8\rangle + 0.0387\lvert 8, 7\rangle - 0.7863\lvert 8, 6\rangle + 0.0700\lvert 8, 5\rangle - 0.0933\lvert 8, 4\rangle + 0.0656\lvert 8, 3\rangle - 0.3636\lvert 8, 2\rangle + 0.0300\lvert 8, 1\rangle + 0.0004\lvert 8, 0\rangle - 0.0069\lvert 8, -1\rangle - 0.0810\lvert 8, -2\rangle + 0.0043\lvert 8, -3\rangle - 0.0036\lvert 8, -4\rangle + 0.0155\lvert 8, -5\rangle - 0.3696\lvert 8, -6\rangle + 0.0041\lvert 8, -7\rangle + 0.1419\lvert 8, -8\rangle$

experimental values of the magnetic moment of the Ho^{3+} ion and considering the corresponding values of n_0 and γ of other rare earth ions [1–3]. In the temperature range $T > T_{\text{comp}}$, $n_0 = 23\,000$ Oe μ_{B}^{-1} mol^{-1} and $\gamma = -0.4 \times 10^{-3}$ K^{-1}; in $T < T_{\text{comp}}$, $n_0 = 25\,000$ Oe μ_{B}^{-1} mol^{-1} and $\gamma = -1.6 \times 10^{-3}$ K^{-1}; for $T = T_{\text{comp}}$, $n_0 = 23\,500$ Oe μ_{B}^{-1} mol^{-1} and $\gamma = -0.4 \times 10^{-3}$ K^{-1}. These values are close to the corresponding values determined by Guillot *et al* [16] ($T > T_{\text{comp}}$, $n_0 = 11\,700$ Oe μ_{B}^{-1} mol^{-1} and $\gamma = -0.4 \times 10^{-3}$ K^{-1}; $T < T_{\text{comp}}$, $n_0 = 12\,600$ Oe μ_{B}^{-1} mol^{-1} and $\gamma = -1.3 \times 10^{-3}$). But it should be noted that, although the values of n_0 and γ present a discontinuity at T_{comp}, the effective field acting on the Ho^{3+} ions remains a continuously increasing function in the whole temperature range (294–4.2 K). The values of M_{Fe} are given by Gonano *et al* [17] from the NMR experiment. By solving the secular equation (equation (7)) with an anisotropic exchange interaction (equation (6i)), we obtain the energies and wavefunctions of the CF-split and superexchange interaction mixed levels of the Ho^{3+} ion at the C_1 sublattice (between $T = 294$ and 4.2 K), M_{Fe} being along the [111] direction. Some results are listed in tables 3 and 4. For an Ho^{3+} at any sublattice and under an applied magnetic field along any direction, similar results are obtained as well.

2.3. The magnetic moment of Ho^{3+} ions

We calculate the value of the magnetic moment of an Ho^{3+} ion at various temperatures T by using the following formula:

$$m = -\mu_{\text{B}} \sum_g \langle g|L_H + 2S_H|g\rangle \rho_g \quad (8)$$

where L_H and S_H are the components of the orbital and spin angular momenta along the direction of the exchange field H_{exch}. $|g\rangle$ is the CF-split and superexchange interaction mixed

Table 4. The same as table 3 but $T = 4.2$ K (only those of the lowest level is given).

Energy	ρ_g	m	Wavefunction
−338.0075	1.0000	5.3962	$0.0555\lvert 8, 8\rangle + 0.0197\lvert 8, 7\rangle - 0.2065\lvert 8, 6\rangle + 0.0335\lvert 8, 5\rangle - 0.0439\lvert 8, 4\rangle + 0.0394\lvert 8, 3\rangle - 0.1812\lvert 8, 2\rangle + 0.0326\lvert 8, 1\rangle + 0.0004\lvert 8, 0\rangle - 0.0504\lvert 8, -1\rangle + 0.4013\lvert 8, -2\rangle - 0.0988\lvert 8, -3\rangle + 0.0940\lvert 8, -4\rangle - 0.1032\lvert 8, -5\rangle + 0.8052\lvert 8, -6\rangle - 0.0614\lvert 8, -7\rangle - 0.2652\lvert 8, -8\rangle$

state whose energy and occupation probability are denoted by E_g and ρ_g, respectively. ρ_g is given by

$$\rho_g = e^{-E_g/kT} \bigg/ \sum_g e^{-E_g/kT} = \rho_0\, e^{-E_g/kT}. \qquad (9)$$

The magnetic moments of an Ho^{3+} ion at any one of the six sublattices at temperature T are calculated when the exchange field is along a certain direction (e.g. the [111] direction). Then the average value of the magnetic moments of the Ho^{3+} ions at the six sublattices, when the field is along this direction, is obtained. Finally, the M_s of the Ho^{3+} ions in HoIG in the temperature range from 4.2 to 294 K, the field being along the [111], [110] and [100] three directions, are obtained and compared with the results measured by some authors.

3. Results and conclusions

For a non-Kramers' ion (Ho^{3+}), its ground multiplet should be split into 17 singlets by the CF with D_2 symmetry. However, only 14 measured energy levels were observed [12] (the highest three levels might be missed in the experiments). The calculated results are in very good agreement with experiments (table 2). A non-degenerate level has no contribution to the magnetic moment. However, from table 2, it can be seen that the energy gaps between the lowest two levels and between many other neighboring levels are small. The two neighboring levels with a small energy gap will be mixed with each other by the superexchange interaction and the so-mixed levels will have a non-negligible contribution to the magnetic moment.

The theoretical values of the magnetization of an Ho^{3+} ion in HoIG in the whole temperature range (294–4.2 K) when magnetized along the [111], [110] and [100] directions are listed in table 5. The following measured values are also listed in the table. They are: (1) the values deduced by Guillot *et al* [16] from the observed M_{Fe}/M_c curve and the values of M_{Fe} [17] at various temperatures (294–21 K) when the field is along the [111] direction; (2) the values obtained by Fadly *et al* [7] at 4.2 K (magnetized along [111], [110] and [100] directions); (3) the values deduced by Guillot *et al* [9] from the M_{HoIG} curve and the values of M_{Fe} in the temperature range from 4.2 to 150 K (magnetized along the [111] and [100] directions). The theoretical results are in good agreement with experiments.
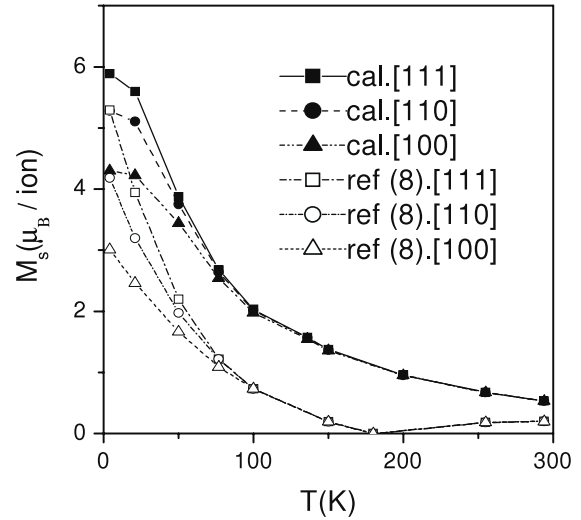
Figure 1. Temperature dependence of the spontaneous magnetization of HoIG:Al (measured [8]) and of the Ho^{3+} ion in HoIG (calculated).

Ostoréro *et al* [8] discovered that the M_s of the HoIG:Al presents a very pronounced anisotropic character and attributed this anisotropy to the Ho^{3+} ions. The theoretical $M_s(T)$ curves of the Ho^{3+} sublattices in HoIG are in good agreement with the measured $M_s(T)$ curves of the HoIG:Al [8] (figure 1). In figure 1, three salient features should be noticed. First, the M_s of the Ho^{3+} ions in HoIG appears quite anisotropic, especially at low temperature ($T < 100$ K). As shown in this figure and table 5, when the crystal is magnetized along the [111] direction the magnetization is maximal, while the one when it is magnetized along the [100] direction is minimal. That is to say, [111] is the easy direction of magnetization, [100] the hard direction and [110] the intermediate direction. Second, the magnetization gap between the [111] direction and the [110] direction is smaller than the one between the [110] and [100] directions. Third, the anisotropy of M_s decreases rapidly with the increase of the temperature and disappears at about 100 K. It is also noted that the strength of the anisotropy of M_s of the HoIG:Al is greater than that of the Ho^{3+} sublattices in HoIG. For example, at $T = 4.2$ K, the difference between the values of the M_s of the Ho^{3+} ions in HoIG when magnetized along the [111] direction and along the [100] direction is 1.585 μ_B/ion. However, for the measured M_s of HoIG:Al, the corresponding difference is around 2.280 μ_B/ion. Although there are some errors in both the calculations and measurements, we think that this disagreement might be mainly due to the following fact. The partial substitution of the Fe^{3+} ions (especially those at the d sites) by non-magnetic Al^{3+} ions will lead to distortion of the crystal lattice and then enhances the anisotropy. From figure 1 it can also be seen that the experimental curves have a compensation temperature but the calculated curves have not. The reason for this difference is as follows. The observed magnetization of HoIG:Al is the resultant magnetization of both the Ho^{3+} sublattices and the Fe^{3+} sublattices. Because the Ho^{3+} moment is generally antiparallel to the resultant Fe^{3+} moment, there exists a compensation temperature ($\sim$180 K [8]) in the observed

Table 5. The calculated and measured values of the magnetic moment of an Ho^{3+} ion in HoIG in the temperature range from 294 to 4.2 K when magnetized along the [111], [110] and [100] directions (in μ_B/ion).

T (K)	294	255	200	150	137	100
m (cal.[111])	0.532	0.675	0.957	1.374	1.572	2.031
m (cal.[110])	0.532	0.674	0.956	1.370	1.566	2.017
m (cal.[100])	0.532	0.673	0.952	1.359	1.549	1.975
m (meas.[111] [16])	0.666	0.800	1.0340	1.4140	1.577	2.112
m (meas.[111] [9])				1.299	1.510	2.155
m (meas.[100] [9])				1.299	1.510	1.993

T (K)	77	50	21	4.2	4.2
m (cal.[111])	2.678	3.875	5.594	5.884	6.467 (meas. [7])
m (cal.[110])	2.643	3.751	5.104	5.274	5.433 (meas. [7])
m (cal.[100])	2.544	3.444	4.226	4.299	4.333 (meas. [7])
m (meas.[111] [16])		3.669	5.741		
m (meas.[111] [9])	3.052	3.708	5.965	6.400	
m (meas.[100] [9])	2.362	3.272	4.315	4.450	
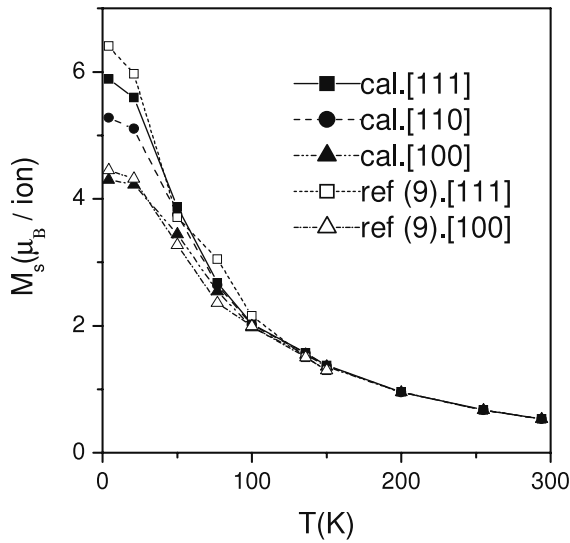
Figure 2. Variation of the spontaneous magnetization of an Ho^{3+} ion in HoIG versus temperature.

Figure 3. Temperature dependence of the spontaneous magnetization of an Ho^{3+} ion in HoIG when magnetized along the [111] direction. The symbols with a star (cal.[111]*, etc) denote the results obtained supposing the Ho^{3+}–Fe^{3+} exchange interaction to be isotropic.

$M_s(T)$ curve. However, the reported theoretical M_s is only that of the Ho^{3+} sublattice in HoIG, so the theoretical curves have no compensation temperatures.

Figure 2 shows the calculated M_s of the Ho^{3+} ions in HoIG when magnetized along the [111], [110] and [100] directions in the temperature range from 4.2 to 294 K and the measured M_s of the Ho^{3+} ions in HoIG along the [111] and [100] directions in the 4.2–150 K temperature range [9]. Obviously, the [111] direction is the easy axis and the [100] direction is the hard axis. The difference between the two directions is evident, that is to say, the anisotropy is strong. From this figure and table 5, it can be seen that the theoretical values when magnetized along the [111] or [100] directions are in good agreement with the experimental results reported in [9] as well.

A comparison between the theoretical M_s of the Ho^{3+} ions in HoIG when magnetized along the [111] direction and the corresponding experimental data deduced by Guillot *et al* [16] is given in figure 3. As shown in this figure and table 5, the agreement between them is also very good.
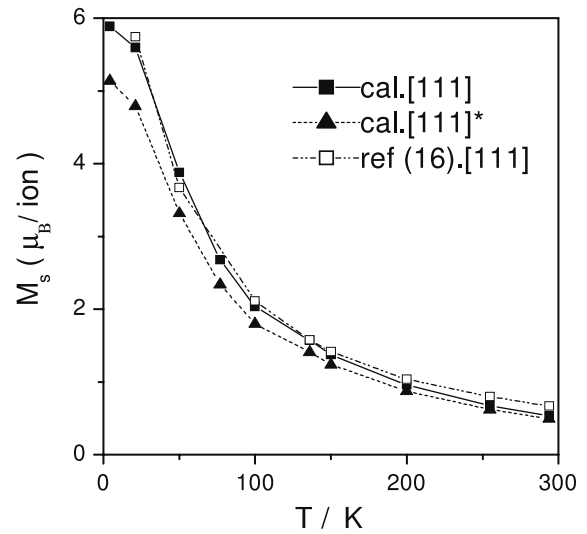
Now we will have some comments about the RE^{3+}–Fe^{3+} exchange interaction. In previous studies on the magnetic or magneto-optical properties of other rare earth iron garnets, the RE^{3+}–Fe^{3+} exchange interaction was generally considered isotropic. However, Alben [15] pointed out that the Yb^{3+}–Fe^{3+} exchange interaction in YbIG is anisotropic and it is an important origin of the macroscopic anisotropic properties of YbIG. Because YbIG and HoIG crystals have the same structure and both Ho and Yb are heavy rare earth elements, in this work we suppose the Ho^{3+}–Fe^{3+} exchange interaction in HoIG to be also anisotropic. To reveal the relative importance of the anisotropy of the Ho^{3+}–Fe^{3+} exchange interaction, the M_s of the Ho^{3+} ions in HoIG when magnetized along the [111], [110] and [100] directions are also calculated supposing the Ho^{3+}–Fe^{3+} exchange interaction to be isotropic (i.e. supposing $G_u = G_v = G_w = 1$, but keeping other parameters unchanged). The obtained results are shown in figures 3 and 4 by the curves with symbols like 'Ho cal. [111]*'. From these
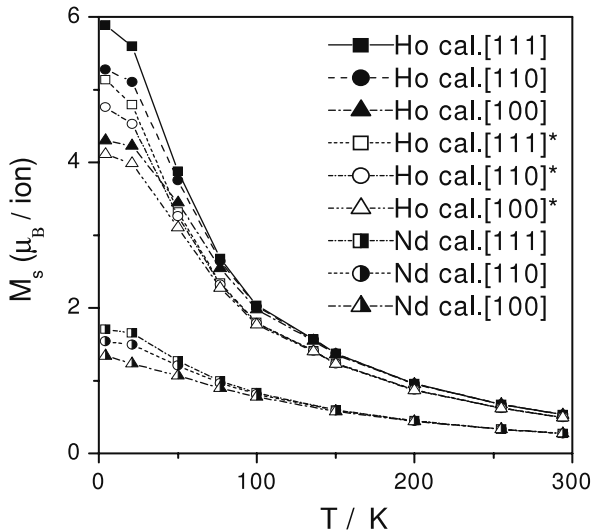
Figure 4. Temperature dependence of the spontaneous magnetization of an Ho^{3+} ion in HoIG and of a Nd^{3+} ion in Nd:YIG. The meaning of the symbols is the same as figure 3 (also see the text).

two figures, we can see that the calculated M_s is smaller than the corresponding values obtained when supposing $G_u = 0.7$, $G_v = 1$, $G_w = 1.3$. However, the tendency of the curves is basically unchanged. The calculated M_s is still anisotropic even when the Ho^{3+}–Fe^{3+} exchange interaction is isotropic. It means that the crystal field upon the Ho^{3+} sublattice alone can induce the magnetic anisotropy.

By analyzing the calculated results, we find out that the contribution of the CF effect to the magnetic anisotropy of the HoIG crystal is greater than the contribution of the anisotropic Ho^{3+}–Fe^{3+} exchange interaction. However, the latter is still important and the two contributions are not counteractive but additive. In addition, at low temperature, the anisotropic exchange interaction increases the magnitude of M_s. Finally, we conclude that the obvious anisotropy of the magnetization of the Ho^{3+} sublattices in HoIG originates mainly from both the CF effect and the anisotropy of the Ho^{3+}–Fe^{3+} exchange interaction.

To justify the correctness of the theory used, we also calculate the M_s of the Nd^{3+} ions in Nd:YIG when magnetized along the [111], [110] and [100] directions in the temperature range from 4.2 to 294 K by using the same theory. Here, the values of G_u, G_v and G_w are 0.7, 1 and 1.3, respectively. The values of CF parameters and n_0, γ are given by Zhang *et al* [3]. The values of M_{Fe} are given by Gonano *et al* [17] from the NMR experiment.

The results are given in figure 4. It can be seen that the calculated M_s of the Nd^{3+} ions in Nd:YIG presents a similar anisotropic character at low temperature. However, two important points should be noted. First, the $M_s(T)$ curves of the Nd^{3+} ions are much gentler than those of the Ho^{3+} ions. Second, the anisotropy is very weak. For instance, the difference between the magnetizations when magnetized along the [111] direction and the [100] direction is 0.366 μ_B/ion at $T = 4.2$ K, being about a fifth of that of the Ho^{3+} ion in HoIG. This may be the reason why little attention has been paid to the anisotropy of the magnetization of the RE^{3+} ions in the light rare earth iron garnets (e.g. Nd:YIG). At the same time, this result implies that the character of the RE^{3+} ions itself may have a bearing on the anisotropy of its M_s in REIGS.

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