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Theoretical analysis of the anisotropy of the magnetization of the Ho^{3+} ion in holmium iron garnet single crystals*

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The spontaneous magnetization of the Ho^{3+} ion in holmium iron garnet (HoIG) single crystals in the temperature range of 4.2–294 K along the directions [111], [110], and [100] are calculated, taking into account the effects of six magnetically inequivalent sites occupied by the Ho^{3+} ions based on the quantum theory. The calculated results show that the magnetization of the Ho^{3+} ion in HoIG is obviously anisotropic. The theoretical results are in agreement with those of experiments. A primary interpretation of the anisotropy of magnetization of the Ho^{3+} ion in HoIG is put forward.

Keywords: Anisotropy, magnetization, crystal field, superexchange interaction, rare-earth sublattice-site

PACC: 7820L, 7530G, 7550G

1. Introduction

In recent years, much attention has been paid to rare-earth-substituted yttrium iron garnets $\{R_xY_{3-x}\}[Fe_2](Fe_3)O_{12}$ (R: YIG, R^{3+} represents a trivalent rare earth ion) mainly because these garnets have a very strong enhancement of the Faraday rotation(FR).[1−6] Different from light rare earth ions, the heavy rare earth ions Ho^{3+} can completely substitute the Y^{3+} ions in yttrium iron garnets to form the compound holmium iron garnet $Ho_3Fe_5O_{12}$ (HoIG) single crystals because of the lanthanide contraction. It should be noted that in HoIG the obvious anisotropy of the magnetization and of the magneto-optical (MO) effects was observed besides a strong increase of the FR, especially at low temperature.[7−8] 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 variations of these quantities, they discovered that both the magnetization and the MO effects in HoIG and in HoIG:Al obviously showed anisotropy. However, theoretical interpretation of this anisotropic phenomenon has not been reported and so further experimental study is confined.

Like other rare earth iron garnets, HoIG belongs to space group Ia3d[9] and has three different magnetic lattices whose respective magnetic moments are denoted by M_d (tetrahedral), M_a (octahedral), M_c (dodecahedral). The Fe^{3+} ions at d sites are strongly coupled antiferromagnetically with the Fe^{3+} ions at a sites; this interaction determines the Curie temperatures which are nearly the same in all rare earth iron garnets (RIGs, R^{3+} represents a trivalent rare earth ion) (560 K). The moment of the rare earth ions is generally antiparallel to the resultant moment of the Fe^{3+} ions; in many RIGs, there is a compensation temperature, T_{comp}, at which the ferrite magnetization is zero; for HoIG, T_{comp} is 136 K.[7]

The rare earth ions, which occupy the 24c crystal sites, are equivalent in crystallography. However, an external magnetic-field or a molecular field will decrease the symmetry and, in general, the 24c lattice is divided into six magnetically inequivalent sublattices under influence of the field, denoted by $C_1C_2C_3$ and $C_1'C_2'C_3'$.[10] The purpose of this work is to calculate the spontaneous magnetization of the Ho^{3+} lattice based on the quantum theory and give an interpreta-

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tion on the anisotropy of the magnetization induced by the Ho^{3+} ions in HoIG. In this study, the different contributions of the six sublattices of the Ho^{3+} ions are calculated.

2. Theory and calculation

The magnetization of the Ho^{3+} ion is simply determined by the successive splittings of the $4f^{10}$ configuration induced by the crystal-field (CF) and $Ho^{3+}-Fe^{3+}$ superexchange interactions. So the perturbation Hamiltonian upon Ho^{3+} ion is

$$\hat{H} = \hat{H}_{\mathrm{C}} + \hat{H}_{\mathrm{ex}}, \tag{1}$$

where $\hat{H}_{\mathrm{C}}$ is the CF Hamiltonian which can be expressed as

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

and A_{kq} is the CF parameter and $Y_{kq}(\theta, \varphi)$ is the spheric harmonics. The CF Hamiltonian can further be expressed as

$$\begin{aligned}\hat{H}_{\mathrm{C}} = & \sum_{k=2,4,6} A_{k0} r^k Y_{k0} + \sum_{k=2,4,6} A_{k\pm2} r^k Y_{k\pm2} \\ & + \sum_{k=4,6} A_{k\pm4} r^k Y_{k\pm4} + A_{6\pm6} r^6 Y_{6\pm6},\end{aligned} \tag{3}$$

because the Ho^{3+} ion has an environment with D_2 symmetry. The CF interactions upon the six magnetically inequivalent sites are the same because the local environments around the six sites are completely identical.

The splitting of the ground term induced by the CF interaction is calculated by solving the following secular equation:

$$\left\| \left\langle JJ_z | \hat{H}_{\mathrm{C}} | J'J_{z'} \right\rangle - E_{\mathrm{C}} \delta_{JJ'} \delta_{J_Z J_{Z'}} \right\| = 0, \tag{4}$$

where $|JJ_z\rangle$ is the wavefunction of free Ho^{3+} ions in the coupled representation, E_{C} is the energy of the CF-split level. Thus, the energies and wavefunctions of the CF-split levels in the ground term of the Ho^{3+} ions in HoIG can be obtained by solving Eq.(4).

Néel theory[11] reduces to the assumptions that the superexchange interaction acting on the rare earth ions in rare earth iron garnets can be expressed as

$$\hat{H}_{\mathrm{ex}} = 2\mu_{\mathrm{B}} \boldsymbol{H}_{\mathrm{ex}} \bullet \hat{S}, \tag{5}$$

where μ_{B} represents Bohr magneton, $\hat{S}$ is the total spin angular momentum operator of the Ho^{3+} ion, $\boldsymbol{H}_{\mathrm{ex}}$ is the exchange field (or molecular field) and can be expressed as

$$\boldsymbol{H}_{\mathrm{ex}} = n_0 \left(1 + \gamma \mathrm{T}\right) \boldsymbol{M}_{\mathrm{Fe}}. \tag{6}$$

Here $\boldsymbol{M}_{\mathrm{Fe}}$ represents the resultant spontaneous magnetization of the two Fe^{3+} ions sublattices, $\boldsymbol{M}_{\mathrm{Fe}} = \boldsymbol{M}_{\mathrm{d}} - \boldsymbol{M}_{\mathrm{a}}$, $\boldsymbol{H}_{\mathrm{ex}}$ is along the direction of $\boldsymbol{M}_{\mathrm{Fe}}$. In the following, for the Ho^{3+} ions at the six sublattices, we use such local coordinates, the directions of their axes being given in Table 1. Then the CF parameters of Eq.(3) are the same for all the six sublattices. Therefore, the wavefunctions, expressed in local coordinates, of the CF-split levels are the same for all the Ho^{3+} ions.

Table 1. The directions for the axes of the local coordinates for the six sublattices of the 24*c* lattice in garnets*.

sites	$\boldsymbol{u}$	$\boldsymbol{v}$	$\boldsymbol{w}$
C_1	[0 1 1]	[0 $\bar{1}$ 1]	[1 0 0]
C_2	[1 0 1]	[1 0 $\bar{1}$]	[0 1 0]
C_3	[1 1 0]	[$\bar{1}$ 1 0]	[0 0 1]
C_1'	[0 1 $\bar{1}$]	[0 1 1]	[1 0 0]
C_2'	[$\bar{1}$ 0 1]	[1 0 1]	[0 1 0]
C_3'	[1 $\bar{1}$ 0]	[1 1 0]	[0 0 1]

*expressed with the directional numbers with respect to the three edges of the cubic crystal cell; symbols C_1,C_2, C_3, and C_1', C_2', C_3' express the six sublattices; u, v, w express unit vectors along the axes of the local coordinates.

Equation (6) can be expressed as

$$\begin{aligned}\boldsymbol{H}_{\mathrm{ex}} = H_0 \Big[& \cos\left(\boldsymbol{M}_{\mathrm{Fe}}, \boldsymbol{u}\right) \boldsymbol{u} + \cos\left(\boldsymbol{M}_{\mathrm{Fe}}, \boldsymbol{v}\right) \boldsymbol{v} \\ & + \cos\left(\boldsymbol{M}_{\mathrm{Fe}}, \boldsymbol{w}\right) \boldsymbol{w} \Big].\end{aligned} \tag{7}$$

In this formula H_0 is the absolute value of the exchange field, $H_0 = n_0 \left(1 + \gamma T\right) M_{\mathrm{Fe}}$, $\cos\left(\boldsymbol{M}_{\mathrm{Fe}}, u\right)$ denotes the cosine of the included angle between the vectors $\boldsymbol{M}_{\mathrm{Fe}}$ and $\boldsymbol{u}$.

The energies and wavefunctions of the CF split and superexchange-interaction-mixed levels of the ground term of the Ho^{3+} ion are obtained by solving the following secular equation:

$$\left\| \left\langle \varphi_{\mathrm{ci}} \right| \hat{H}_{\mathrm{C}} + \hat{H}_{\mathrm{ex}} \left| \varphi_{\mathrm{cj}} \right\rangle - E_g \delta_{\mathrm{ij}} \right\| = 0, \tag{8}$$

where $|\varphi_{\mathrm{ci}}\rangle$ and $\langle \varphi_{\mathrm{ci}}| \hat{H}_{\mathrm{C}} |\varphi_{\mathrm{ci}}\rangle$ are the eigen wavefunction and eigen energy obtained by solving Eq.(4), respectively.

The ionic magnetic moment, at temperature T, is given by

$$m = -\mu_{\mathrm{B}} \sum_{g} \langle g|L_H + 2S_H\,|g\rangle\, \rho_g, \tag{9}$$

where L_H and S_H are the component of the orbital and spin angular momenta along the direction of the exchange field; $|g\rangle$ is the CF-split and superexchange interaction-mixed (or split) state whose energy and occupation probability are denoted by E_g and ρ_g, respectively; ρ_g is expressed as

$$\rho_g = \exp\left(-E_g/kT\right) \Big/ \sum_{g} \exp\left(-E_g/kT\right). \tag{10}$$

The magnetic moments of Ho^{3+} ions at the six sublattices at temperature T are calculated when the exchange field is along a certain direction (e.g. [111] direction). Then the average value for the magnetic moments of the Ho^{3+} ions at the six sublattices when the exchange field is along this direction, is reported and compared with the measured value. Finally, the spontaneous magnetization is obtained of the Ho^{3+} ions in HoIG over the whole temperature range (294–4.2 K) when they are magnetized along [111], [110], and [100] directions.

3. Results of calculations

In the calculation, to gain the proper set of the CF parameters for the Ho^{3+} ion is a crucial step. Following the previous works on other R:IG (Pr:IG, Nd:IG and so on),[1,2] these parameters are determined according to the values of CF parameters for the Sm^{3+} in SmIG deduced from the magnetic phase transition experiments by Nekvasil *et al*[12] and the ratios of the values of $\langle r^2\rangle$, $\langle r^4\rangle$, $\langle r^6\rangle$ of the Ho^{3+} ion and the Sm^{3+} ion. The determined non-zero parameters are listed in Table 2.

Table 2. The non-zero parameters of the CF for the Ho^{3+} ions in HoIG (in cm^{-1}).

	$\langle r^2\rangle A_{20}$	$\langle r^2\rangle A_{2\pm2}$	$\langle r^4\rangle A_{40}$	$\langle r^4\rangle A_{4\pm2}$	$\langle r^4\rangle A_{4\pm4}$	$\langle r^6\rangle A_{60}$
4f(Ho^{3+})	−331.5	239.2	−2708.2	−845.6	633.0	135.2
	$\langle r^6\rangle A_{6\pm2}$	$\langle r^6\rangle A_{6\pm4}$	$\langle r^6\rangle A_{6\pm6}$			
4f(Ho^{3+})	−674.4	886.7	33.7			

The ground term $4f^{10}$ (^{5}I) 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. The energy gaps between the ground multiplet and the excited multiplets ($^5I_7$, $^5I_6$, $^5I_5$, $^5I_4$) are so large that the mixing of the ground multiplet with other multiplets caused by the CF can be completely neglected. For instance, the energy of the lowest 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 $^5I_8$ multiplet (487 cm^{-1}).[13] So in the calculation of the energies and wavefunctions of the CF-split levels of the ground multiplet $4f^{10}$ ($^5I_8$) the influence of the excited multiplets ($^5I_7$ etc.) will completely be neglected. The calculated energies of the CF-split levels of the ground multiplet $4f^{10}$ ($^5I_8$) are given in Table 3, in which the energy of the lowest CF-split $^5I_8$ level is taken to be zero. The energies and the wavefunctions of the lowest four CF-split levels are listed in Table 4. In this table, the average energy of the $^5I_8$ multiplet is taken to be zero.

Table 3. The experimental[13] and calculated energies of the CF-split levels of the ground multiplet $(4f^{10})^5I_8$ of the Ho^{3+} ion in HoIG (in cm^{-1}).

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

Table 4. The energies and wavefunctions of the lowest four CF-split levels of the ground multiplet of the Ho^{3+} ion.

Energy/cm^{-1}	Wave function
−322.1149	$0.1840\|8,8\rangle$-$0.5842\|8,6\rangle$-$0.0780\|8,4\rangle - 0.3447\,\|8,2\rangle + 0.0000\,\|8,0\rangle + 0.3447\,\|8,-2\rangle$
	$+0.0780\|8,-4\rangle + 0.5842\,\|8,-6\rangle - 0.1840\,\|8,-8\rangle$
−293.2931	$0.2178\,\|8,8\rangle - 0.6271\,\|8,6\rangle - 0.0485\,\|8,4\rangle - 0.2387\,\|8,2\rangle + 0.0008\,\|8,0\rangle$
	$-0.2387\,\|8,-2\rangle - 0.0485\,\|8,-4\rangle - 0.6271\,\|8,-6\rangle + 0.2178\,\|8,-8\rangle$
−247.2352	$-0.3107\,\|8,7\rangle - 0.3508\,\|8,5\rangle - 0.4689\,\|8,3\rangle - 0.2460\,\|8,1\rangle + 0.2460\,\|8,-1\rangle$
	$+0.4689\,\|8,-3\rangle + 0.3508\,\|8,-5\rangle + 0.3107\,\|8,-7\rangle$
−231.5948	$0.5388\,\|8,7\rangle - 0.2473\,\|8,5\rangle + 0.3749\,\|8,3\rangle - 0.0895\,\|8,1\rangle - 0.0895\,\|8,-1\rangle$
	$+0.3749\,\|8,-3\rangle - 0.2473\,\|8,-5\rangle + 0.5388\,\|8,-7\rangle$

Since the Ho^{3+} ion is of non-Kramers' type, the ground multiplet $4f^{10}$ ($^5I_8$) is split into seventeen singlets by the CF with D_2 symmetry. However, there are only fourteen energy levels that have been measured. The highest three levels may not be observed in the experiment. From Table 3 it can be seen that the agreement between the calculated and experimental values is satisfactory. A nondegenerate level has no contribution to the magnetic moment of the Ho^{3+} ion. But from Table 3, it can be seen that the energy gaps between the lowest two levels, between the third and the fourth levels, between the fourth and the fifth levels, between the fifth and the sixth levels, and between the seventh and the eighth levels (for simplicity, the energies and the corresponding wavefunctions of the 5–8 th levels are not listed in Table 4) are small. The two levels in every couple will be mixed with each other by the superexchange interaction and the so-mixed levels will now have a non-negligible contribution to the magnetic moment. Of course, these levels can also be mixed with levels of other couples by the superexchange interaction and have extra contributions to the magnetic moment. However, these contributions are negligible.

Guillot *et al*[14] showed that both the values of n_0 and γ in Eq.(6) are temperature independent. However, they also showed that the values in the temperature range $T < T_{\rm comp}$ are different from those in temperature range $T > T_{\rm comp}$. We determine the values of n_0 and γ by fitting the 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. The determined values are as follows. In the temperature range $T > T_{\rm comp}$, $n_0 = 23000\,{\rm Oe}\mu_{\rm b}^{-1}\,{\rm mol}^{-1}$ and $\gamma = -0.4 \times 10^{-3}\,{\rm K}^{-1}$; in the range of $T < T_{\rm comp}$ $n_0 = 25000\,{\rm Oe}\mu_{\rm b}^{-1}\,{\rm mol}^{-1}$ and $\gamma = -1.6 \times 10^{-3}\,{\rm K}^{-1}$; for $T = T_{\rm comp}$, $n_0 = 23500\,{\rm Oe}\mu_{\rm b}^{-1}\,{\rm mol}^{-1}$ and $\gamma = -0.4 \times 10^{-3}\,{\rm K}^{-1}$. These values are close to the corresponding values determined by Guillot *et al* ($T > T_{\rm comp}$, $n_0 = 11700\,{\rm Oe}\mu_{\rm b}^{-1}\,{\rm mol}^{-1}$ and $\gamma = -0.4\times10^{-3}\,{\rm K}^{-1}$; $T < T_{\rm comp}$, $n_0 = 12600\,{\rm Oe}\mu_{\rm b}^{-1}\,{\rm mol}^{-1}$ and $\gamma = -1.3 \times 10^{-3}$). The values of $\boldsymbol{M}_{\rm Fe}$ is given by Gonano *et al*[15] from the NMR experiment. Finally, the calculated values of the magnetization of a 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. For comparison the experimental values deduced by Guillot *et al*[14]

Table 5. The values of the calculated and measured magnetic moment of a Ho^{3+} ions in HoIG at various temperatures ($T = 294 - 4.2\,{\rm K}$) along [111], [110], [100] directions (in $\mu_{\rm B}$/ion).

T/K	294	255	200	150	136	100
m(cal.[111])	0.493	0.621	0.872	1.237	1.410	1.795
m(cal.[110])	0.493	0.621	0.872	1.235	1.407	1.788
m(cal.[100])	0.493	0.620	0.870	1.230	1.398	1.768
m(meas.[111][14])	0.666	0.800	1.034	1.414	1.577	2.112
T/K	77	50	21	4.2	4.2	
m(cal.[111])	2.338	3.317	4.790	5.136	6.467(meas.[7])	
m(cal.[110])	2.321	3.259	4.525	4.758	5.433(meas.[7])	
m(cal.[100])	2.273	3.103	3.985	4.111	4.333(meas.[7])	
m(meas.[111][14])		3.669	5.741			

from the M_{Fe}/M_C curve and the values of M_{Fe} at various temperatures (294–4.2 K) when the exchange field is along the [111] direction and the measured values by Fadly *et al*[7] at $T = 4.2$ K (magnetized along [111], [110], and [100] directions) are listed in the table as well.

In order to make a comparison between the calculated and experimental values clearer, Figs.1 and 2 are given as follows.

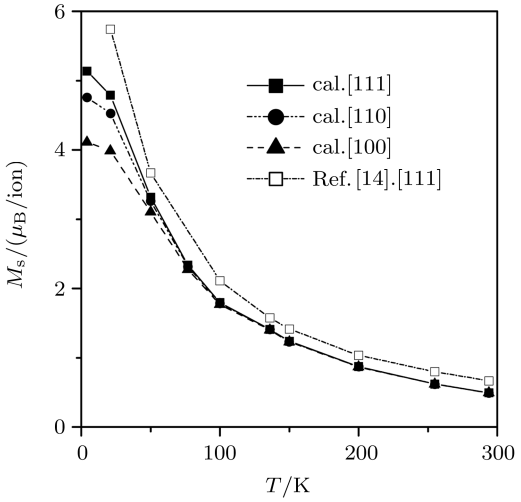


Fig.1. Variation of the spontaneous magnetization of the Ho^{3+} ion with temperature in HoIG.

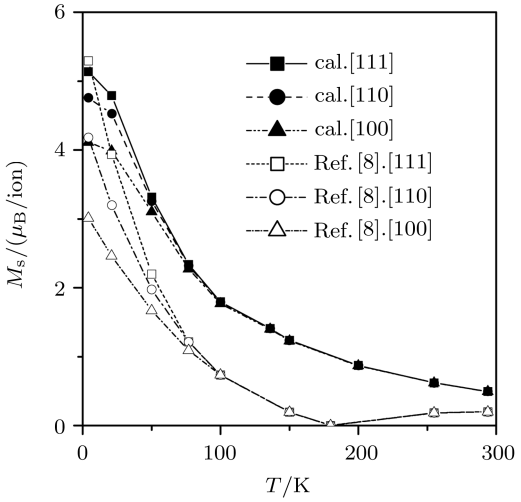


Fig.2. Temperature dependence of the spontaneous magnetization of the HoIG:Al (measured) and of the Ho^{3+} ion in HoIG (calculated).

Figure 1 shows the variation of the calculated spontaneous magnetization (M_s) of the Ho^{3+} sublattice in HoIG with temperature when magnetized along the [111], [110], and [100] directions, and the experimental values[14] along the [111] direction. Three salient features can be observed in this figure. First, the anisotropy of the magnetization of the Ho^{3+} sublattice appears to be very obvious, especially at low temperatures ($T < 100$ K). As shown in this figure and Table 5, the magnetization of the Ho^{3+} sublattice is maximal when magnetized along the [111] direction, while it is minimal when magnetized along [100] direction and is intermediate when magnetized along the [110] direction. Second, the gap between the two curves obtained when magnetized along the [111] direction and [110] direction is smaller than the corresponding gap between the two curves obtained when magnetized along the [110] direction and [100] direction. Finally, magnetized along whichever directions, the curve of the magnetization versus temperature is nonlinear and presents a sharply assurgent tendency. Although the calculated magnetization curve (magnetized along the [111] direction) is below the corresponding measured curve, the tendency of the calculated curves are in agreement with that of the experimental.

A comparison between the temperature dependence of the measured M_s of the HoIG:Al[8] and that of the calculated M_s of the Ho^{3+} sublattice in HoIG are given in Fig.2. Taking into account the anisotropy discussed above, the general tendency of the theoretical curves is in very good agreement with that of the measured curves, especially at low temperatures ($T < 100$ K). But it can be seen that the strength of the anisotropy of M_s of the HoIG:Al is greater than that of the Ho^{3+} sublattice in HoIG. This may be due to the fact that when non-magnetic Al^{3+} ions partly substitute the Fe^{3+} ions (especially those at the d sites) the symmetry of the lattice will be changed and then the anisotropy increases.

From Fig.2, it can also be seen that the experimental curve has a compensation temperature but the calculated curve has not. The reason for this may be the fact that the magnetization of the HoIG:Al is the resultant magnetization of the magnetic moment of the Ho^{3+} sublattice and the resultant moment of the Fe^{3+} ions. Because the Ho^{3+} moment is generally antiparallel to the Fe^{3+} resultant moment, there exists a compensation temperature ($\sim$180 K)[8] in the HoIG:Al single crystals, at which the bulk magnetization vanishes. In this paper, however, only the magnetization of the Ho^{3+} sublattice in HoIG is calculated, so the theoretical curve has no compensation temperature.

As shown in Table 5, the agreement between our calculated ones and the experimental values deduced

by Fadly *et al*[7] (at $T = 4.2\,\mathrm{K}$) is also satisfactory.

4. Discussions and conclusion

Our theoretical calculation about the magnetization of the Ho^{3+} ions leads to a preliminary interpretation of the anisotropy of the magnetization of the Ho^{3+} ions in HoIG. It shows that the obvious anisotropy of the magnetization of the Ho^{3+} ions originates mainly from the effects of the crystal field. From Eq.(7) it can be seen that when the crystal is magnetized along different directions, the Zeeman splitting will be different (though the magnitude of the exchange interaction remains unchanged). So the magnetization is dependent on the magnetized direction. Furthermore, also from Eq.(7) it can be seen that when the crystal is magnetized along a certain direction, the Zeeman splitting of the Ho^{3+} ions at different sublattices is different. Hence, to interpret the observed results we have to take into account the different contributions of the different Ho^{3+} sublattices. But it must be noted that the agreement between the calculated and the experimental values is not very good. This may be attributed to the anisotropy of the exchange field, which will be studied in the future.

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