An investigation on the magneto-optic properties of terbium gallium garnet under high magnetic field*

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The superexchange interaction on a magnetic ion may be represented by an effective field $\boldsymbol{H}_{\rm m} = \lambda \boldsymbol{M}$ in some paramagnetic materials, here λ is the coefficient of effective field and $\boldsymbol{M} = \chi \boldsymbol{H}_{\rm e}$ with χ being the magnetic susceptibility and $\boldsymbol{H}_{\rm e}$ being the applied field. The variation of the equivalent $\lambda\chi$ with the dynamic applied field is given and the crystal field-splitting levels of the excited configuration $4f^7 5d^1$ of the Tb^{3+} ion are calculated in the $Tb_3Ga_5O_{12}$. By means of the effective field $\boldsymbol{H}_{\rm m}$ and the applied field $\boldsymbol{H}_{\rm e}$, the Faraday rotation of $Tb_3Ga_5O_{12}$ at 6 K and 41 K, under the high magnetic field and at 0.63 μm wavelength, are presented. Our calculated results are in agreement with the experimental data.

Keywords: terbium gallium garnet, high applied field, superexchange interaction, magneto-optical properties

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1. Introduction

The terbium gallium garnet $Tb_3Ga_5O_{12}$(TGG) has attracted much attention for several decades due to some of its merits as being chemically stable, mechanically strong and large magnitudes of magneto-optical effect which may be used in the optical isolators, modulators, highly efficient laser matrices, acousto-optics devices, and so on.[1,2]. In the previous semiempirical works,[3,4] the superexchange interaction between rare-earth ions in gallium garnet and the crystal field (CF)-splitting levels of the excited configuration $4f^7 5d^1$ of Tb^{3+} ions were neglected and the oscillator strength for allowed $f - d$ transitions was only taken as a constant. In this paper, when the wavelength is 0.63 μm, we show the Faraday rotation (FR) of TGG at 6 K and 41 K under the high magnetic field based on the quantum theory. The CF-splitting levels of the excited configuration $4f^7 5d^1$ of Tb^{3+} ions and the superexchange interaction between rare-earth ions in gallium garnets are taken into account. Our calculated results are supported by the experimental data.

2. Calculations of CF and Zeeman-splitting levels for Tb^{3+} ions in the TGG

The Hamiltonian of Tb^{3+} ions in TGG crystal is

$$\hat{H} = \hat{H}_0 + \hat{H}_{\rm c} + \hat{H}_{\rm m}. \tag{1}$$

Here $\hat{H}_0$ is the Hamiltonian of the free Tb^{3+} ions, $\hat{H}_{\rm c}$ is the CF Hamiltonian expressed as

$$\hat{H}_{\rm C} = \sum_{k,q} A_{k,q} \langle r^k \rangle Y_{k,q}(\theta, \varphi). \tag{2}$$

Here $A_{k,q}\langle r^k \rangle$ are the CF parameters, $\langle r^k \rangle$ the average values of r^k and $Y_{k,q}$ the spherical harmonic functions. $\hat{H}_{\rm m}$ is the Zeeman effect Hamiltonian in Eq. (1).

The structure of TGG crystal is described by space group O_h^{10},[5] the Tb^{3+} ions are located at six nonequivalent dodecahedral positions whose environment has orthorhombic symmetry with point group D_2. As a result, Eq. (2) is simplified as: for $4f^8$ configuration,

$$H_{\rm C} = \sum_{k=2,4,6} A_{k,0} r^k Y_{k,0} + \sum_{k=2,4,6} A_{k,\pm 2} r^k Y_{k,\pm 2} + \sum_{k=4,6} A_{k,\pm 4} r^k Y_{k,\pm 4} + A_{6,\pm 6} r^6 Y_{6,\pm 6}; \tag{3}$$

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For $4f^7 5d^1$ configuration,

$$H_{\rm C} = \sum_{k=2,4} A_{k,0} r^k Y_{k,0} + \sum_{k=2,4} A_{k,\pm 2} r^k Y_{k,\pm 2} + A_{4,\pm 4} r^4 Y_{4,\pm 4}. \quad (4)$$

The parameters of CF acting on the bound configuration of Tb^{3+}ions in gallium garnet were proposed by Guillot *et al.*[6] For the excited configuration, the CF-parameters are listed in Table 1, which were obtained by fitting the excitation spectra.[7]

Table 1. The values of the CF parameters upon Tb^{3+} ions (in cm^{-1}).

	$A_{20}\langle r^2\rangle$	$A_{2\pm 2}\langle r^2\rangle$	$A_{40}\langle r^4\rangle$	$A_{4\pm 2}\langle r^4\rangle$	$A_{4\pm 4}\langle r^4\rangle$	$A_{60}\langle r^6\rangle$	$A_{6\pm 2}\langle r^6\rangle$	$A_{6\pm 4}\langle r^6\rangle$	$A_{6\pm 6}\langle r^6\rangle$
$4f$	–128.4	267.9	–2555.9	294.2	1116.6	677	–155	1045	–4
$5d$	–3063	1180	–139845	72999	5412				

The CF-split levels and the corresponding wave functions of Tb^{3+} ions in gallium garnet have been obtained by solving the following secular equation

$$\|\langle\varphi_a|\hat{H}_{\rm c}|\varphi_b\rangle - E_{\rm g}^{(0)}\delta_{ab}\| = 0. \quad (5)$$

Here $\langle\varphi_a|$ and $\langle\varphi_b|$ are the wave functions of the free Tb^{3+} ions. In the calculation the admixing of the multiplets $4f^8 {}^7F_6$ and ${}^7F_5$ has been taken into account. The energies of the lowest eight CF-split levels for the $4f^8$ configuration of Tb^{3+} ions in TGG are –217.88, –215.96, –179.71, –179.31, –178.26, –173.96, 25.29 and 46.19 cm^{-1},respectively.

The $4f^7 5d^1$ configuration of Tb^{3+} ions forms the multiplets ${}^9D_J$ (J = 6, 5, 4, 3, 2) and ${}^7D_J$ (J = 5, 4, 3, 2, 1) and the levels of the ${}^9D$ term are lower than that of the ${}^7D$. The minimum interval of the spin-allowed $4f^7 5d^1$ ${}^7D - 4f^8$ ${}^7F$ transition of Tb^{3+} ion is 38462 cm^{-1}[7] in TGG. When the admixing of the multiplets $4f^7 5d^1$ ${}^7D_5$ and ${}^7D_4$ is taken into account, the corresponding calculated value is 38460 cm^{-1}.

The definition of six nonequivalent crystal sites at which Tb^{3+} ions are located are shown in Table 2.[3]

Table 2. Orientation of local symmetry axes for six inequivalent dodecahedral positions of rare-earth ions in rare-earth garnets.

unit vector	C1	C2	C3	C4	C5	C6
u	[100]	[010]	[001]	[100]	[010]	[001]
v	[01$\bar{1}$]	[$\bar{1}$01]	[1$\bar{1}$0]	[011]	[101]	[110]
				[0$\bar{1}$1]	[10$\bar{1}$]	[$\bar{1}$10]
w	[011]	[101]	[110]			

For the TGG crystal, the Néel temperature is 0.25 K,[8] therefore the TGG is paramagnetic at 6 K and the magnetic susceptibility χ of the crystal conforms to the Curie–Weiss law, suggesting that the superexchange interaction between the electrons in different magnetic ions plays an important role in the crystal, which is similar to the case of rare earth trifluorides.[9,10] The superexchange interaction can be described by an effective field $\boldsymbol{H}_{\rm m}$, i.e.

$$\boldsymbol{H}_{\rm m} = \rho \boldsymbol{M}. \quad (6)$$

Here ρ is the coefficient of the effective field, which is related to the temperature. $\boldsymbol{M}$ is the magnetization induced by the applied field $\boldsymbol{H}_{\rm e}$. The total field $\boldsymbol{H}_a$ acting on the magnetic ions is the sum of the applied field $\boldsymbol{H}_{\rm e}$ and the effective field $\boldsymbol{H}_{\rm m}$,

$$\boldsymbol{H}_a = \boldsymbol{H}_{\rm e} + \boldsymbol{H}_{\rm m} = (1 + \lambda\chi)\boldsymbol{H}_{\rm e} = (1 + \chi_{\rm m})\boldsymbol{H}_{\rm e}. \quad (7)$$

The applied field $\boldsymbol{H}_{\rm e}$ is along the [110] axis. The Zeeman effect Hamiltonian is given by

$$\hat{H}_{\rm m} = \mu_{\rm B}[H_{\rm e}(\hat{L}_{\rm z} + 2\hat{S}_{\rm z}) + \chi_{\rm m} H_{\rm e} 2\hat{S}_{\rm z}]. \quad (8)$$

Here $\mu_{\rm B}$ is the Bohr magneton, $\chi_{\rm m} = \lambda\chi$ is equivalent magnetic susceptibility, $\hat{L}$ and $\hat{S}$ are the orbital and spin angular momentum operators, respectively.

It is a crucial step to determine the equivalent magnetic susceptibility $\chi_{\rm m}$ in this work. Wang *et al.*[11] took the $\chi_{\rm m}$ as a constant in the investigations of the magneto-optical properties for the ErGG, which can not describe the magneto-optical behaviour

of TbGG completely. Since the rate of field variation is 10^7–10^8 T/s in the experiments,[3] the magnetic susceptibility χ and $\chi_{\rm m}$ will change obviously with the dynamic applied field $\boldsymbol{H}_{\rm e}$.[12] Therefore the $\chi_{\rm m}$ is approximately expressed as

$$\chi_{\rm m} = a + bH_{\rm e}^{-1} + cH_{\rm e}. \tag{9}$$

Here a, b and c are the constants determined by fitting the measured values of the FR at 6 K[3] with the optimum values being –0.502, 1.114×10^2 kOe and -8.220×10^{-4} (kOe)$^{-1}$ (1 Oe=79.5775 A/m), respectively.

As a perturbation, the effective field $\boldsymbol{H}_{\rm m}$ induces the splitting of the CF energy levels with the applied field $\boldsymbol{H}_{\rm e}$. For the six nonequivalent crystal sites, the Zeeman–splitting levels can be obtained by solving the secular equation

$$\|\langle\varphi_{\rm i}|\hat{H}_{\rm m}|\varphi_{\rm j}\rangle - E_{\rm g}^{(1)}\delta_{ij}\| = 0. \tag{10}$$

Here $\langle\phi_{\rm i}|$ and $\langle\phi_{\rm j}|$ are the wave functions corresponding to CF-splitting energy levels $E_{\rm g}^{(0)}$. Since the average energy of $4f^7 5d^1$ configuration is more than 60000 cm^{-1}, which is much large than that of the $4f^8$ configuration, the effect of the superexchange interaction on the $4f^7 5d^1$ configuration can be neglected.

3. Calculation of Faraday rotation

The megnetic-optic (MO) effect of the TGG crystals in the visible region is mainly due to the intraionic parity allowed electric dipole transition between the $4f^8$ and $4f^7 5d^1$ configurations, whose contribution to the Faraday rotation (FR) per unit length is[13]

$$\theta_{\rm F} = \frac{N\pi e^2}{9\hbar c}\frac{(\bar{n}^2+2)^2}{\bar{n}}\sum_{g,n}\frac{\omega^2}{\omega_{ng}^2-\omega^2} \times \{|\langle n|V_-|g\rangle|^2 - |\langle n|V_+|g\rangle|^2\}\rho_{\rm g}. \tag{11}$$

Here N is the number of ions per unit volume, $\bar{n}$ the average refractive index of TGG, ω the angular frequency of the light wave, $\langle g|$ a set of ground states with energy $E_{\rm g}$, $|n\rangle$ the excited states with energies E_n, $\hbar\omega_{ng} = E_n - E_{\rm g}$, $eV_\pm$ are the electric dipole moment operators for the right- and left-hand circularly polarized light, respectively,

$$eV_\pm = e(x \pm {\rm i}\, y). \tag{12}$$

$\rho_{\rm g}$ is the occupation probability of the state $|g\rangle$ and can be expressed as

$$\begin{aligned}\rho_{\rm g} &= \rho_0 {\rm e}^{-E_{\rm g}/kT} = \rho_0 {\rm e}^{-(E_{\rm g}^{(0)}+E_{\rm g}^{(1)})/kT} \\ &= \rho_0 {\rm e}^{-E_{\rm g}^{(0)}/kT}\left(1 - \frac{\mu_{\rm B}}{kT}\langle g|(H_{\rm e}(\hat{L}_{\rm z}+2\hat{S}_{\rm z}) + H_{\rm e}\chi_{\rm m}2\hat{S}_{\rm z})|g\rangle\right).\end{aligned} \tag{13}$$

By solving Eqs. (11)–(13), the average values of

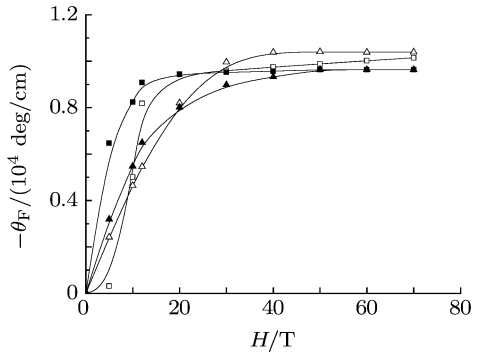


Fig. 1. Dependence of the Faraday rotation on the magnetic field in $Tb_3Ga_5O_{12}$. The experimental values at 6 K[3] (—■—), the calculated values at 6 K without taking the exchange interaction into account (—□—), the experimental values at 41 K[3] (—▲—), the calculated values at 41 K without taking the exchange interaction into account (—△—).

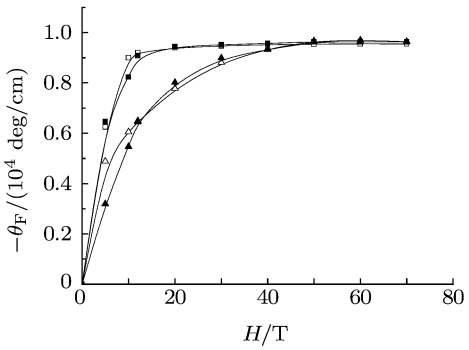


Fig. 2. Dependence of the Faraday rotation in $Tb_3Ga_5O_{12}$ on the magnetic field. The experimental values at 6 K[3] (—■—), the calculated values at 6 K obtained by taking the exchange interaction into account (—□—), the experimental values at 41 K[3] (—▲—), the calculated values at 41 K obtained by taking the exchange interaction into account (—△—).

FR for the six nonequivalent crystal sites in TGG are obtained at 6 K and 41 K when the wavelength is

0.63 μm. Figures 1 and 2 show the results without and with the superexchange interaction, respectively, which are compared with the experimental data.[3] In the calculation, the value of $\langle r \rangle_{4f5d}$ of the Tb^{3+} ion in TGG is taken as 0.0172 nm.

4. Discussion and conclusion

In this paper, the specific FR of TGG crystals at 6K and 41K when the wavelength is 0.63 μm have been studied based on the quantum theory. It can be seen from Fig. 2 that our calculated results are in agreement with the experimental data.

The superexchange interaction between the electrons in different magnetic ions plays an important role in the terbium gallium garnet, which is described as an effective field $\boldsymbol{H}_{\rm m}$. The high order terms of the dynamic applied field has an important effect on FR.

It is concluded from Eqs. (7) and (9) that the total field $\boldsymbol{H}_a$ acting on the magnetic ions changes linearly in the low applied field, which is attributed to the constant $\chi_{\rm m}$,[11] and that when the applied field becomes larger the $\boldsymbol{H}_a$ increases with the square of applied field.

The effective field and the applied field lead to the splitting of the ground state, which affects the occupation probability of an electron in the ground state, resulting in the fact that the slopes of $\mathrm{d}\theta_{\rm F}/\mathrm{d}H$ are different at 6 K and 41 K.

The temperature dependence of the effective field coefficient λ originates mainly from the superexchange interaction between the electrons in different magnetic ions and the spin-fluctuations of the magnetic ions.[14−16] The temperature dependence effect of the effective field coefficient λ on FR is smaller than that of high order terms of the applied field under high applied field. Therefore the temperature dependence of the effective field coefficient λ is neglected in this work, leading to that the calculated values of FR in lower applied field at 41 K are not in good agreement with the experimental data. The variation of the equivalent magnetic susceptibility $\chi_{\rm m}$ with the dynamic applied field $\boldsymbol{H}_{\rm e}$ and the temperature simultaneously will be investigated further.

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