Quantum theory of the strong magneto-optical effect of Ce-substituted yttrium iron garnet

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Calculations of the Faraday rotation (FR) and ellipticity spectra in the wavelength region between 360 and 1260 nm caused by the Ce^{3+} ions in the Ce-substituted yttrium iron garnets based on the quantum theory are presented. The calculation of the temperature dependence of FR at 633 and 1150 nm wavelengths is presented as well. The Faraday effect contributed by the Ce^{3+} ions is caused mainly by the intraionic electrical dipole transitions between the $4f$ and $5d$ configurations. The effect of the crystal field (CF), superexchange interaction, and spin-orbit coupling on the magneto-optical (MO) effect has been discussed. The energy differences between the lowest and second CF-split $5d$ levels and the lowest CF-split $4f$ level are 1.36 and 3.10 eV, respectively. Therefore there are anomalous rotatory dispersions at about 1.36 and 3.10 eV. The difference of the transition intensity for the right- and left-handed circularly polarized light between the lowest CF-split $5d$ level and the ground state is negative, while the corresponding value for the transition between the second CF-split $5d$ level and the ground state is positive. So the FR is positive over the whole photon energy region between 1.36 and 3.10 eV and is negative outside this region. Since the energy differences between the CF splitting levels of the excited configuration are comparable with the energy difference between the mean energies of the excited and ground configurations, the MO coefficient varies with temperature. The admixing of different multiplets of the ground configuration is another cause which makes the MO coefficient vary with temperature.

I. INTRODUCTION

Many experimental investigations on the magneto-optical (MO) effect and the magnetic properties of rare-earth- (RE-) substituted iron garnets have been carried out. Recently, it has been found that Ce-substituted yttrium iron garnet (Ce:YIG) has a very large Faraday rotation (FR) in the visible and near-infrared regions. The factor per Ce ion is 2×10^4 deg/cm at a wavelength 633 nm and -1.3×10^4 deg/cm at 1150 nm.[1,2] The specific efficiency of the Ce^{3+} ions is much larger than that of Bi^{3+} ions, the champion of the MO enhancement for the past 20 years. Thus Ce:YIG film is a very promising MO material for application in the region from the visible to the near infrared. It is found that the Ce substitution prominently enhances the Faraday effect not only in the infrared region at $h\nu = 1.4$ eV, but also in the ultraviolet region at $h\nu = 3.1$ eV. Owing to the resonance dispersion effect, the FR varies rapidly as the frequency is swept over the 1.4 and 3.1 eV regions, and this is accompanied by a large Faraday ellipticity.[2] But the origin of the giant MO effect of Ce:YIG is not clear yet. Therefore it is of great importance to study the mechanism of the contribution of the RE ions in such compounds to the MO effect.

It is known that the FR in Ce:YIG is mainly due to the Ce sublattice. According to Refs. 1–3, the single-ion model is valid for the FR caused by the RE sublattice in R:YIG when the temperature is not very low and the concentration of the RE ion is not very large and the FR

due to the RE sublattice in Ce:YIG and Pr:YIG is strongly dependent on the wavelength. Weller, Reim, and Schrijner[4] have found from x-ray photoemission spectroscopy and MO spectroscopy that the $4f \rightarrow 5d$ transitions enhance the MO effect considerably in Nd-Fe-Co and Yb-Fe-Co. Jacobs, Krupke, and Weber[5] have revealed a strong lowering of the $4f \rightarrow 5d$ transitions in Ce^{3+} ions from 50 000 cm^{-1} in the free-ion state to 22 000 cm^{-1} in the dodecahedral site in yttrium aluminum garnet (YAG). Based on all these facts, we think that the MO effect in Ce:YIG is caused mainly by the ultraionic electric dipole (ED) transition between the $4f$ and $5d$ configurations of the Ce^{3+} ions which are split by the crystal field (CF) and the superexchange interaction.

II. CALCULATION OF THE FARADAY ROTATION AND ELLIPTICITY AT ROOM TEMPERATURE

As mentioned above, the Faraday effect contributed by the Ce sublattice is assumed to be caused mainly by the intraionic ED transitions between the $4f$ and $5d$ configurations. Therefore it is necessary to calculate the energy levels split by the CF and superexchange interaction and the corresponding wave functions. The perturbation Hamiltonian of a Ce^{3+} ion in the crystal is

$$H' = V_c + \xi l \cdot s + 2\mu_B H_m \cdot s , \qquad (1)$$

where $\xi l \cdot s$ is the spin-orbit coupling, the third term in the right-hand side is the Zeeman energy, H_m is the molecular field, and V_c is the CF energy and is given by

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

The Ce^{3+} ions are at the c sites of the lattice; their environment has D_2 symmetry. Thus, every CF-split level is doubly degenerate.

The choice of the proper set of parameters of the CF upon the Ce^{3+} ions is a crucial step. The values obtained with a point-charge model are too small to obtain correct results. A similar situation was also found when studying the magnetic properties of the RE-substituted garnets. Nekvasil et al.[6] have determined a set of parameters ($A_{20}\langle r^2 \rangle$, $A_{2\pm2}\langle r^2 \rangle$, etc.) for the CF upon the Sm^{3+} ions in SmIG by fitting the experimental results about the magnetic phase transition in SmIG. It is expected that the parameters of the CF upon the light RE ions in different R:YIG compounds will change gradually with the atomic number of the RE ions. So we determined the parameters of the CF upon the Ce^{3+} ions according to the values of the CF parameters obtained by Nekvasil et al. for the Sm^{3+} ions in SmIG and the ratios of the values of $\langle r^2 \rangle$, $\langle r^4 \rangle$, and $\langle r^6 \rangle$ of different RE ions. The nonzero parameters are shown in Table I. The values of ξ for the $4f$ and $5d$ configurations and the difference of the mean energies of these two configurations have been taken from the paper by Starostin.[7] According to Refs. 8 and 9, the value of the molecular field $\mathbf{H}_m$ at room temperature has been taken to be 510 kOe.

Because the superexchange interaction is less than the spin-orbit coupling and the CF effect, we carried out the perturbation calculation in the following order of priorities: $V_c + \xi l \cdot s$ and $2\mu_B \mathbf{H}_m \cdot s$. The CF-split energy levels and the corresponding wave functions have been obtained by solving the secular equation

$$\| \langle \varphi_i | V_c + \xi l \cdot s | \varphi_j \rangle - E\delta_{ij} \| = 0 \ . \tag{3}$$

In calculating the splitting of the $4f$ states, both $^2F_{5/2}$ and $^2F_{7/2}$ multiplets have been included. As for the $5d$ states, the $^2D_{3/2}$ and $^2D_{5/2}$ multiplets have been included. The calculated levels are given in Table II.

The Zeeman splitting of the states of the $4f$ configuration caused by the superexchange interaction was obtained by solving the following secular equation:

$$\| \langle \psi_i | V_c + \xi l \cdot s + 2\mu_B \mathbf{H}_m \cdot s | \psi_j \rangle - E\delta_{ij} \| = 0 \ . \tag{4}$$

Because the admixing of the lowest three CF levels owing to the superexchange interaction is not negligible, the bra and ket in Eq. (4) include the six wave functions of the lowest three levels. Compared with the superexchange interaction, the energy differences between the higher lev-

TABLE I. The values of the parameters of the CF upon the Ce ions (in cm^{-1}).

	$A_{20}\langle r^2 \rangle$	$A_{2\pm2}\langle r^2 \rangle$	$A_{40}\langle r^4 \rangle$	$A_{4\pm2}\langle r^4 \rangle$	$A_{4\pm4}\langle r^4 \rangle$
$4f$	-1363	340	-9889	588	1520
$5d$	-4200	1500	-166900	9250	62730
	$A_{60}\langle r^6 \rangle$	$A_{6\pm2}\langle r^6 \rangle$	$A_{6\pm4}\langle r^6 \rangle$	$A_{6\pm6}\langle r^6 \rangle$	
$4f$	4288	-310	1120	153	

TABLE II. The values of the CF-split energy levels of the $4f$ and $5d$ configurations (in cm^{-1}), taking the average energy of the $4f$ shell as zero.

$4f$	-2321	-1655	-1236	47.35	448.5	1574	3142
$5d$	8670	22702	65065	73689	79148		

els and these levels are large enough so the error caused by the neglect of the higher levels is very small. The calculated results at room temperature (294 K) are listed in Table III. The effect of the exchange interaction on the splitting of the $5d$ states has been neglected.

When the damping can be neglected, the complex FR per unit length caused by the ED transitions is[10,11]

$$\varphi = \theta_F + i\psi = \frac{\pi\omega}{c\bar{n}} \frac{(\bar{n}^2+2)^2}{9} (\chi^- - \chi^+) \ . \tag{5}$$

Here the Lorentz-Lorenz correction is incorporated. In Eq. (5), θ_F is the FR, ψ is the Faraday ellipticity, χ^- and χ^+ are the electric susceptibility for right- and left-handed circularly polarized light, $\bar{n}$ is the mean refractive index of the material, ω is the angular frequency of the light wave, and c the velocity of the light in vacuum.

As pointed out by Dionne and Allen,[12] for the magnetic garnets at energies below 2.5 eV, the imaginary part of the diagonal element of the permittivity tensor $\varepsilon_0'' \approx 0$, and the real part of the diagonal element $\varepsilon_0' \approx \bar{n}^2$. For YIG, $\bar{n} \approx 2.2$.[13] That is, the index of refraction is very much greater than the extinction coefficient. Furthermore, both ε_0' and ε_0'' are only modestly changed by rare-earth substitution. These features make Eq. (5) accurate for calculating the MO effect of Ce:YIG in the energy region considered.

By inserting the quantum-mechanical expressions for ac susceptibility into Eq. (5), we obtain

$$\theta_F = \frac{N\pi(\bar{n}^2+2)^2}{9\bar{n}c\hbar} \sum_{n,g} A_{ng} \frac{\omega^2(\omega_{ng}^2 - \omega^2 - \Gamma_{ng}^2)}{(\omega_{ng}^2 - \omega^2 + \Gamma_{ng}^2)^2 + 4\omega^2\Gamma_{ng}^2} \rho_g \ , \tag{6}$$

$$\psi = \frac{N\pi(\bar{n}^2+2)^2}{9\bar{n}c\hbar} \sum_{n,g} A_{ng} \frac{\omega\Gamma_{ng}(\omega_{ng}^2 + \omega^2 + \Gamma_{ng}^2)}{(\omega_{ng}^2 - \omega^2 + \Gamma_{ng}^2)^2 + 4\omega^2\Gamma_{ng}^2} \rho_g \tag{7}$$

for each type of ion present in the material, where

$$A_{ng} = |\langle n|V_-|g \rangle|^2 - |\langle n|V_+|g \rangle|^2 \ . \tag{8}$$

In Eqs. (6)–(8), N is the number of ions per unit volume, Γ_{ng} are the half-widths of resonance lines, $|g \rangle$ are the CF- and exchange-interaction-split $4f$ states with energy E_g, $|n \rangle$ are the CF-split $5d$ states with energy E_n, and $\hbar\omega_{ng} = E_n - E_g$. $V_\pm$ are the ED moment operators for right- and left-handed circularly polarized light:

$$V_\pm = e(x \pm iy) \ . \tag{9}$$

ρ_g is the occupation probability of the state $|g \rangle$ and is given by

$$\rho_g = e^{-E_g/kT} \Big/ \left[\sum_g e^{-E_g/kT} \right] = \rho_0 e^{-E_g/kT} . \qquad (10)$$

The values of the occupation probabilities of the lowest six CF- and exchange-interaction-split states of the $4f$ configuration are listed in Table III.

Figure 1 shows the energy levels of the $4f$ and $5d$ configurations and the ED transitions between these two configurations. It should be noted that (1) the admixing of the lowest three CF-split $4f$ levels by superexchange interaction has been considered; (2) the Zeeman splitting of the $5d$ states is neglected, so every level is doubly degenerate. The Zeeman splitting of the $5d$ states only leads to a very small change of ω_{ng}. Therefore the error caused by the neglect of the Zeeman splitting of the $5d$ states is very small.

It can be seen from Fig. 1 that the degenerate $4f$ levels are split into sublevels due to the superexchange interaction. When a plane-polarized electromagnetic wave propagates through the crystal, the transition probabilities for the right- and left-handed circularly polarized light between these sublevels and the $5d$ levels are the same. Hence there will be no Faraday effect if there is no superexchange interaction, but the Zeeman splitting makes the occupation probabilities of these sublevels different. This results in a Faraday effect. It is seen from Table III that the occupation probabilities of the third and other higher CF- and superexchange-interaction-split $4f$ levels are small. Therefore the Faraday effect comes mainly from the ED transitions from the lowest two $4f$ states to the $5d$ states.

Recently, Dionne and Allen[12] have analyzed the origins of the giant FR and ellipticity in Bi-substituted garnets. They show that the transitions in the Fe^{3+} ions
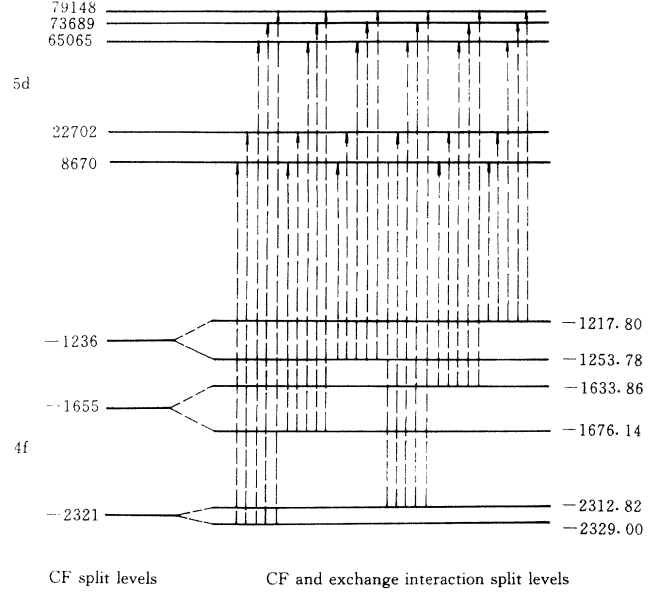

FIG. 1. Energy level and ED transition diagram for Ce^{3+} ions in Ce:YIG at room temperature. The data on both sides are the values of the energy in cm^{-1}. It should be noted that the Zeeman splitting of the $5d$ states is neglected; every $5d$ level is doubly degenerate.

which are influenced by covalent interactions with Bi ions are of diagmagnetic type. As will be discussed in Sec. III, the molecular field upon the rare-earth ions in YIG is very much less than the molecular field upon the Fe^{3+} ions in YIG. Therefore, the Zeeman splitting of the Ce^{3+} ions is very much smaller than the splitting of the

TABLE III. The energies, occupation probabilities (ρ_g), and wave functions of the lowest six CF- and exchange-interaction-split levels of the $4f$ configuration at 294 K. Here the representation $|J, J_z\rangle$ is used; so, for example, $|\frac{5}{2}, \frac{3}{2}\rangle$ represents the wave function $|J = \frac{5}{2}, M_J = \frac{3}{2}\rangle$.

Energy (cm^{-1})	ρ_g	Wave function
-2329.00	$0.498\,12$	$0.087\,30\|\frac{5}{2},\frac{3}{2}\rangle + 0.985\,07\|\frac{5}{2},-\frac{1}{2}\rangle - 0.092\,57\|\frac{5}{2},-\frac{5}{2}\rangle$ $+0.022\,34\|\frac{7}{2},\frac{7}{2}\rangle - 0.049\,98\|\frac{7}{2},\frac{3}{2}\rangle - 0.096\,79\|\frac{7}{2},-\frac{1}{2}\rangle$ $+0.032\,60\|\frac{7}{2},-\frac{5}{2}\rangle$
-2312.82	$0.460\,15$	$-0.085\,78\|\frac{5}{2},\frac{5}{2}\rangle + 0.985\,98\|\frac{5}{2},\frac{1}{2}\rangle + 0.085\,40\|\frac{5}{2},-\frac{3}{2}\rangle$ $-0.031\,32\|\frac{7}{2},\frac{5}{2}\rangle + 0.096\,59\|\frac{7}{2},\frac{1}{2}\rangle + 0.048\,76\|\frac{7}{2},-\frac{3}{2}\rangle$ $-0.022\,09\|\frac{7}{2},-\frac{7}{2}\rangle$
-1676.14	$0.020\,396$	$-0.181\,27\|\frac{5}{2},\frac{3}{2}\rangle + 0.120\,60\|\frac{5}{2},-\frac{1}{2}\rangle + 0.951\,20\|\frac{5}{2},-\frac{5}{2}\rangle$ $-0.030\,57\|\frac{7}{2},\frac{7}{2}\rangle + 0.108\,50\|\frac{7}{2},\frac{3}{2}\rangle + 0.028\,72\|\frac{7}{2},-\frac{1}{2}\rangle$ $-0.185\,15\|\frac{7}{2},-\frac{5}{2}\rangle$
-1633.86	$0.016\,59$	$0.949\,97\|\frac{5}{2},\frac{5}{2}\rangle + 0.114\,08\|\frac{5}{2},\frac{1}{2}\rangle - 0.188\,84\|\frac{5}{4},-\frac{3}{2}\rangle$ $+0.184\,52\|\frac{7}{2},\frac{5}{2}\rangle - 0.029\,34\|\frac{7}{2},\frac{1}{2}\rangle - 0.114\,05\|\frac{7}{2},-\frac{3}{2}\rangle$ $+0.031\,05\|\frac{7}{2},-\frac{7}{2}\rangle$
-1253.78	$0.002\,582$	$0.776\,61\|\frac{5}{2},\frac{3}{2}\rangle - 0.075\,71\|\frac{5}{2},-\frac{1}{2}\rangle + 0.206\,17\|\frac{5}{2},-\frac{5}{2}\rangle$ $+0.036\,21\|\frac{7}{2},\frac{7}{2}\rangle - 0.581\,44\|\frac{7}{2},\frac{3}{2}\rangle + 0.009\,02\|\frac{7}{2},-\frac{1}{2}\rangle$ $-0.095\,75\|\frac{7}{2},-\frac{5}{2}\rangle$
-1217.80	$0.002\,165$	$0.214\,61\|\frac{5}{2},\frac{5}{2}\rangle - 0.073\,97\|\frac{5}{2},\frac{1}{2}\rangle + 0.775\,02\|\frac{5}{2},-\frac{3}{2}\rangle$ $+0.097\,38\|\frac{7}{2},\frac{5}{2}\rangle - 0.009\,21\|\frac{7}{2},\frac{1}{2}\rangle + 0.580\,48\|\frac{7}{2},-\frac{3}{2}\rangle$ $-0.035\,95\|\frac{7}{2},-\frac{7}{2}\rangle$

Fe^{3+} ions caused by superexchange fields. As a result, the difference between the occupation probabilities of the lowest two $4f$ levels of the Ce^{3+} ions is not large. So the MO effect of Ce^{3+} ions in YIG has paramagnetic features. As a comparison, the sketch of the spectral transitions for the Fe^{3+} ions in Bi-substituted YIG is given in Fig. 2.

It is seen from Fig. 1 that below 4 eV there are two resonance frequencies (at 1.36 and 3.10 eV, respectively). They correspond to the transitions from the lowest and second CF levels of the $5d$ configuration to the ground state. The value of A_{ng} is negative for the transition from the lowest $5d$ CF level to the ground state, while it is positive for the transition from the second $5d$ CF level to the ground state. Therefore the FR is positive over the whole region between these two resonance frequencies, and is negative outside this region. For simplicity, it is assumed that the transitions from the lowest $5d$ CF level to various $4f$ levels have the same half-width Γ_1. The corresponding value for the second $5d$ CF level is assumed to be Γ_2. The values of Γ_1 and Γ_2 are determined by fitting the measured Faraday rotation and ellipticity spectra obtained by Gomi, Furuyama, and Abe.[1,2] The values of $\hbar\Gamma_1$ and $\hbar\Gamma_2$ obtained are 0.15 and 0.187 eV, respectively. These values are between the values obtained by Gomi, Furuyama, and Abe[1,2] and those obtained by Kucera, Bok, and Nitsch.[14] The energy differences between the higher $5d$ CF levels and the ground state are larger than 4.0 eV, so the values of the half-width for the transitions from these levels to the $4f$ states are assumed to be zero. The value of $\langle r \rangle_{4f5d}$ used is 1.01 Å to fit the measured values of the FR. It is a little larger than the corresponding value of the free Ce^{3+} ions.[7,15] We think the extension of the radial wave function of the Ce^{3+} ion in the garnets may be one cause for the actual CF being very much larger than that calculated with the point-charge model. Furthermore, Kucera[16] and Jacobs, Krupka, and Weber[5] argued that the covalency of the $5d$ states with the neighboring ions might be large. Therefore we think this value is reasonable. The calculated Faraday rotation and ellipticity spectra contributed by the Ce^{3+} ions for $Y_2CeFe_5O_{12}$ at room temperature are shown in
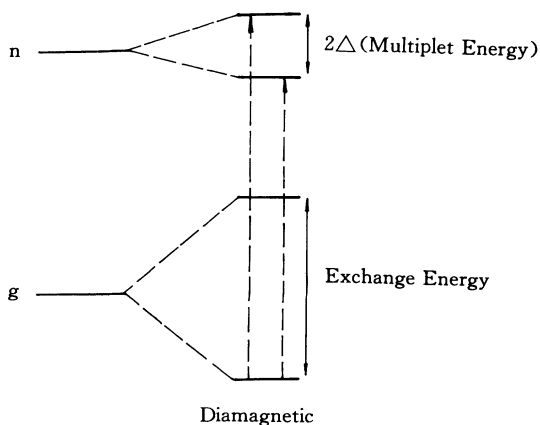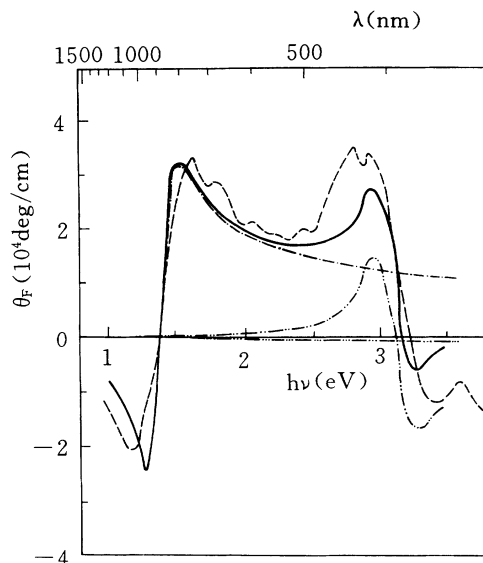
FIG. 3. Faraday rotation spectrum contributed by the Ce^{3+} sublattice of $Y_2CeFe_5O_{12}$ at room temperature. ——— FR caused by the lowest $5d$ level$\rightarrow 4f$ states transitions (the left part of this curve is coincident with the curve of the total FR); ————— FR caused by the second $5d$ level$\rightarrow 4f$ states transitions; —————FR caused by the other higher $5d$ levels$\rightarrow 4f$ states transitions; ——— total FR; — — — measured values obtained by Gomi, Furuyama, and Abe (Ref. 2).

Figs. 3 and 4, respectively. Besides the total FR, the FR caused by the transitions from the lowest $5d$ level, the second $5d$ level, and other higher levels to various $4f$ states are also shown in Fig. 3. It should be noticed that the left part of the curve of the FR caused by the transi-

FIG. 2. Sketch of the spectral transitions for iron ions in Bi-substituted garnets [after Dionne and Allen (Ref. 12)].
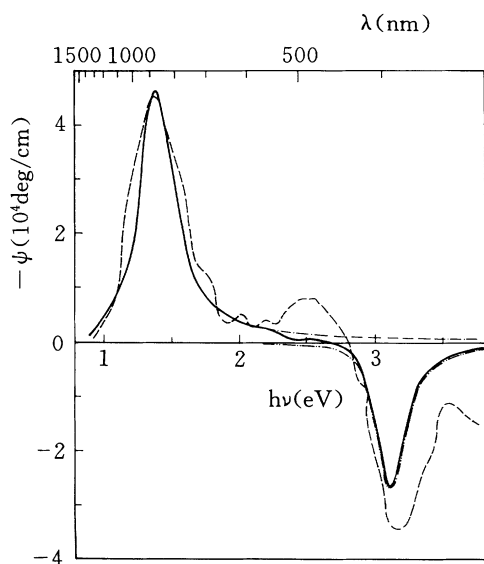
FIG. 4. Same as Fig. 3, but for the Faraday ellipticity spectrum. The contribution of the third and other higher $5d$ levels is neglected. The left part of the curve for the ellipticity caused by the transitions from the lowest $5d$ level$\rightarrow 4f$ states is coincident with the curve of the total ellipticity.

TABLE IV. The calculated values of the magnetic moment $\bar{m}$, FR θ_F at 633 and 1150 nm wavelengths, and the MO coefficient C_e in the temperature range from 50 to 294 K. In the calculations, the admixing of the $^2F_{5/2}$ and $^2F_{7/2}$ multiplets is taken into account. $\bar{m}$ is in μ_B/ion, θ_F in deg. cm^{-1} per Ce ion, and C_e in (deg. cm^{-1} per Ce ion)/(μ_B/ion).

T	$\bar{m}$	θ_F(633 nm)	θ_F(1150 nm)	$|C_e|$(633 nm)	$|C_e|$(1150 nm)
294	0.038	2.03×10^4	-1.26×10^4	53.4×10^4	33.2×10^4
255	0.0455	2.81×10^4	-1.71×10^4	61.8×10^4	37.6×10^4
200	0.060	4.42×10^4	-2.75×10^4	73.7×10^4	45.8×10^4
150	0.0857	6.85×10^4	-4.36×10^4	79.9×10^4	50.9×10^4
100	0.14	11.42×10^4	-7.46×10^4	81.6×10^4	53.5×10^4
50	0.29	23.35×10^4	-15.60×10^4	80.5×10^4	53.8×10^4

tions from the lowest $5d$ level is coincident with the curve of the total FR. In Fig. 4, besides the total ellipticity, the ellipticity caused by the transitions from the lowest $5d$ level and the second $5d$ level is also shown. The contribution of the higher $5d$ levels to the ellipticity is very small and is neglected. The left part of the curve of the ellipticity caused by the transitions from the lowest $5d$ level is coincident with the curve of the total ellipticity. From these figures it can be seen that the MO effect comes mainly from the ED transitions between the lowest two $4f$ levels and the lowest two $5d$ levels.

The measured spectra obtained by Gomi, Furuyama, and Abe[2] are given in these figures as well. It can be seen that the calculated curves are in good agreement with the measured curves. It should be noticed that both the FR and the Faraday ellipticity of about 1.3 eV obtained by Kucera, Bok, and Nitsch[14] are larger than these values. This difference may be caused by measurement errors of the cerium concentration, because the values are very low in the samples used by Kucera, Bok, and Nitsch.

III. TEMPERATURE DEPENDENCE OF THE FARADAY ROTATION

For many RE-substituted garnets, the FR by the RE sublattice increases as the temperature decreases. At the same time, the magnetization of the RE sublattice (M_C) increases as well. The MO coefficient, i.e., the ratio of FR to M_C, usually increases as temperature decreases. However, Crossley, Cooper, and Page[17] have pointed out that the MO effect caused by each magnetic ion is proportional to its magnetic moment evolution in a wide range of circumstances (especially the temperature and magnetic-field dependences). But there are some reasons to make the MO coefficient vary with temperature. If either the absolute values of the perturbation energies of the excited configuration ΔE_n or the corresponding

values of the ground configuration ΔE_g are not much less than the difference between the mean energies of the excited and ground configurations, the MO coefficient will vary with temperature. The admixing of different multiplets of the ground configuration by the CF also makes the MO coefficient vary with temperature. In this section, this problem is studied comprehensively.

In the RE-substituted garnets, there are three exchange interactions, i.e., the R-Fe exchange interaction between the RE and Fe sublattices, the R-R exchange interaction, and the Fe-Fe exchange interaction. The Fe-Fe exchange interaction is much larger than the R-Fe exchange interaction, while the latter is in turn much larger than the R-R exchange interaction. Therefore the Zeeman splitting of the RE ions depends mainly on the R-Fe exchange. The resultant magnetization of two Fe sublattices is well represented by the magnetization of YIG $(M_{\rm YIG})$. Therefore the molecular field $\mathbf{H}_m$ acting upon the Ce^{3+} ions can be expressed as

$$\mathbf{H}_m = n M_{\rm YIG} \ . \tag{11}$$

According to Néel,[18] to the first-order approximation, n can be expressed as

$$n = n_0(1 + \gamma T) \ . \tag{12}$$

According to Leycuras et al.,[19] $\gamma = -1.3 \times 10^{-3}$. The values of $M_{\rm YIG}$ at various temperatures have been obtained by Gonano.[20] The values of the magnetic moment of a Ce^{3+} ion at various temperatures,

$$\bar{m} = \sum_g \langle g|(l_z + 2s_z)\mu_B|g \rangle \rho_0 e^{-E_g/kT} \tag{13}$$

have been calculated.

The calculated values of $\bar{m}$, θ_F, and the MO coefficient in the temperature range from 50 to 300 K with the admixing of the $^2F_{5/2}$ and $^2F_{7/2}$ multiplets taken into ac-

TABLE V. Same as Table IV, but the admixing of the $^2F_{5/2}$ and $^2F_{7/2}$ multiplets is neglected.

T	$\bar{m}$	θ_F(633 nm)	θ_F(1150 nm)	$|C_e|$(633 nm)	$|C_e|$(1150 nm)
294	0.0191	0.724×10^4	-0.505×10^4	37.9×10^4	26.4×10^4
255	0.0227	1.00×10^4	-0.682×10^4	44.1×10^4	30.0×10^4
200	0.0310	1.59×10^4	-1.07×10^4	51.2×10^4	34.5×10^4
150	0.0461	2.48×10^4	-1.68×10^4	53.8×10^4	36.4×10^4
100	0.0775	4.16×10^4	-2.86×10^4	53.7×10^4	36.9×10^4
50	0.1676	8.97×10^4	-6.26×10^4	53.5×10^4	47.4×10^4

count are given in Table IV. The corresponding values when such admixing is neglected are given in Table V.

From these tables, it can be seen that (1) the calculated MO coefficient is dependent on the temperature; (2) since the splitting of the excited configuration owing to the CF is very large (see Table II), the variation of the MO coefficient with temperature comes mainly from the splitting of the excited configuration; (3) the admixing of the $^2F_{5/2}$ and $^2F_{7/2}$ multiplets also makes the FR increase and the MO coefficient vary with temperature.

IV. THE EFFECT OF THE SPIN-ORBIT COUPLING

There are some ambiguities about the effect of the spin-orbit coupling on the MO behavior. It is usually considered that the spin-orbit coupling is a very important factor in determining the MO behavior of materials. So far, the explanation of the huge enhancement of the MO effect of the bismuth or lead ions contained in the iron garnets is restricted to qualitative arguments about the large value of spin-orbit coupling for the $6p$ electrons of the Bi^{3+} and Pb^{2+} ions and the importance of the covalent $2p$-$6p$ bond between the valence shells of the Bi^{3+} and O^{2-} ions.[21] Liu, Zuo, and Le[22] have pointed out that the MO effect in ferromagnetic materials originates mainly from the splitting of the excited states caused by the spin-orbit coupling. To clarify this problem, the effect of the spin-orbit coupling on the FR by the Ce^{3+} ions has been studied. First, we set the value of the spin-orbit coupling parameter ξ of the $4f$ states to zero and keep other parameters unchanged in calculating the FR. Then we set the parameter ξ of the $5d$ states to zero in the calculation. Finally, both the spin-orbit coupling parameters of the $4f$ and $5d$ states are set to zero. The calculated results are listed in Table VI. We can see from the table that the effect of the spin-orbit coupling of the ground configuration on the FR is very large, while the effect of the spin-orbit coupling of the excited configuration is very small. Similar calculations for the FR in CeF_3 have been made. In CeF_3, the environment of the Ce^{3+} ion has C_{3v} symmetry. It is found that the effect of the spin-orbit coupling of both the $4f$ configuration and the $5d$ configuration on the MO behavior of CeF_3 is small.

V. CONCLUSIONS AND DISCUSSIONS

From the calculated results, it can be seen that the MO behavior of the Ce^{3+} ion in Ce:YIG is well explained by the model used in this paper. The Faraday effect contributed by the Ce^{3+} ions is caused mainly by the intraionic ED transitions between the lowest two CF- and exchange-interaction-split levels of the $4f$ configuration and the lowest two CF-split levels of the $5d$ configuration.

TABLE VI. The influence of the spin-orbit coupling on the Faraday rotation in Ce:YIG at room temperature (ξ in cm^{-1}, θ_F in deg. cm^{-1} per Ce ion).

$\xi(4f)$	$\xi(5d)$	$\theta_F(633\ nm)$	$\theta_F(1150\ nm)$
644	1000	2.0×10^4	-1.25×10^4
644	0	1.87×10^4	-1.19×10^4
0	1000	0.23×10^3	-0.146×10^3
0	0	0.089×10^2	-0.137×10^2

The CF and the superexchange interaction are two very important factors in determining the FR in the garnets. The effect of the splitting of both the ground and excited states due to the CF on the MO behavior of Ce:YIG is great, but if the CF is weak enough, the effect of the splitting of the excited configuration caused by the CF will be small. The splitting of the excited states caused by the superexchange interaction and the applied magnetic field is usually negligible.

The spin-orbit coupling is another factor in determining the FR in materials. It is found that the spin-orbit coupling of the ground configuration is very important for the FR in Ce:YIG, while the effect of the spin-orbit coupling of the excited configuration on the FR is very small.

In Ce:YIG, the very large splitting of the states caused by the CF makes the MO coefficient vary with temperature. The admixing of different multiplets of the ground configuration is another cause which makes the MO coefficient vary with temperature. The environment of the RE ions in R:YIG has D_2 symmetry. Therefore, in the case of non-Kramers ions, all CF-split levels are nondegenerate. The nondegenerate CF states will barely contribute to the Faraday effect if the admixing of different CF states is negligible. Hence, for the non-Kramers RE ions in R:YIG, the Faraday effect is caused by the admixing of different CF states owing to the superexchange interaction. This is another reason for the difference of the MO behavior of the Ce^{3+} ion and Pr^{3+} ion in YIG being so large, in spite of Pr being next to Ce in the periodic table.

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