Faraday rotation in Co^{3+}-substituted magnetic garnets

Faraday rotation in Co^{3+}-substituted magnetic garnets

Luke McAven[1] and Kiminari Shinagawa[2]

[1] School of Information Technology and Computer Science, University of Wollongong,
Wollongong 2522, Australia
[2] Department of Physics, Toho University, Funabashi city, Chiba 274 8510, Japan

E-mail: lukemc@uow.edu.au and sinagawa@ph.sci.toho-u.ac.jp

Abstract
Magnetic garnets containing Co^{3+} at a tetrahedral site exhibit Faraday rotation.
The temperature dependence of the rotation has recently been measured, and
an attempt was made to describe the mechanism of the rotation. An error in
the theoretical calculation led to significant disparity between the experimental
results and the theoretical predictions of the model. Several of the distinct
features of the spectra were missing. We apply the same mechanism to show
that the features are described better than previously suggested.

1. Introduction and background

Magnetic garnets with substituted tetrahedral cobalt-ion sites are known to exhibit Faraday
rotation (FR). However, until recently (Shinagawa *et al* 1997, 1999, Takeuchi *et al* 1999)
experimental measurements had been performed at room temperature (Egashira and Manabe
1972, Saito *et al* 1987).

Investigations of the Co^{3+} system at room temperature (Saito *et al* 1987) showed that
the dispersive-shaped FR spectra at around 1.3 μm arise due to the tetrahedral crystal-field
transitions ^{5}E $\rightarrow$ ^{5}T$_2$ of the Co^{3+} ions. Taking the strong-crystal-field approximation (Sugano
et al 1970), and including the spin–orbit interaction and the lower-symmetry (S_4) crystal field,
explained the room temperature FRS well.

Takeuchi *et al* (1999) measured the temperature dependence, in the range 60–300 K, of the
FR of a Co^{3+}-substituted magnetic garnet system, (YGd)$_3$Fe$_{5-0.07}$Co$_{0.07}$O$_{12}$. They analysed
the results using the molecular-field approximation which had been successfully applied to
Co^{2+}-substituted magnetic garnets (Shinagawa *et al* 1997, 1999). The measurements showed
several changes in the dispersive-shaped FRS as the temperature is decreased.

(1) There appears to be a very slight shifting of the FRS spectra centre from about 1.3 μm to
shorter wavelengths.
(2) There is a distinct sharpening of the positive rotation peak centred at about 1.4 μm.

(3) The negative rotation peak centred at about 1.2 μm becomes saturated. This is most noticeable in the small difference in this region between temperatures of 120 and 60 K (figure 2 of Takeuchi *et al* 1999).

Takeuchi *et al* (1999) provide fitted parameters and conclude that the low-symmetry crystal field plays an important role in the FRS of magnetic garnets containing tetrahedral Co^{3+}. This conclusion is consistent with the discussion in Saito *et al* (1987) on the significance of the low-symmetry crystal-field parameter in determining overall shape of the FRS.

However, the calculated temperature dependences of Takeuchi *et al* (1999) do not describe well the three FRS temperature dependences described above. Sharpening at the high-wavelength peak occurs but results in an symmetrically unphysical sharpening at the low-wavelength peak. We have since found errors in their calculations. The order of the spin–orbit terms was incorrectly assigned. Furthermore, calculations of the transition strengths, as given here in figure 3, were incorrect. This was traced to flaws in the programs used in the analysis of Takeuchi *et al* (1999). We compare characteristics of correctly calculated spectra with the experimental features identified above and give a new set of parameters from fitting. The fit to the experimental data suggests the model needs modification to fully represent the characteristics of temperature dependence in FR.

The outline of this paper is as follows. In section 2 we outline the model we take of the energy levels and states in the Co^{3+}-substituted garnet. Section 3 describes the transitions allowed for the left and right circularly polarized light, the difference between which is critical in the FRS.

Section 4 describes the modelling of the temperature dependence of the FR in this system. We present our set of parameters in section 5 and discuss the goodness of fit.

We summarize our conclusions in section 6.

2. Energy levels and eigenstates

The energy level diagram for Co^{3+} at a tetrahedral site in magnetic garnet is given in figure 1. We shall discuss the various interactions and labellings associated with the splittings in that diagram. Note that the effective spin–orbit interaction should be defined as $-\lambda(L \cdot S)$, reversing the order of the excited states from those given in figure 3 of Takeuchi *et al* (1999).

The ground and excited states of tetrahedral Co^{3+} $(3d)^6$ are labelled by ^5E ($^5E(t_2)^3(e)^3$ in full) and $^5T_2$ ($^5T_2(t_2)^4(e)^2$ in full) respectively. The excitations from the ground to the excited states are thus charge transfer transitions from an e orbital to a t_2 orbital. As indicated in the diagram the states are separated by 10 Dq. The most significant interaction here is the spin–orbit one. The excited state is equivalent to the $^5P(L = 1)$ atomic state, so the spin–orbit interaction splits it into three states which we associate with $J = 1$, 2 and 3. The three states are shifted by 3λ, λ and -2λ respectively, where λ is negative. The ground state is not split by the spin–orbit interaction.

The tetrahedral site in magnetic garnets is distorted to an S_4 symmetry (Wood and Remeika 1967). Takeuchi *et al* (1999) mention that tetrahedral Co^{3+} $(3d)^6$ is a Jahn–Teller ion, which may imply that the low-symmetry distortion around Co^{3+} is large compared with that around Fe^{3+} $(3d)^5$. Like Takeuchi *et al* (1999), we assume the symmetry around Co^{3+} to still be S_4. The S_4 low-symmetry crystal field further splits the excited state J-multiplets, as demonstrated in figure 1. In figure 2 we give calculated splitting of the excited state $^5T_2$ due to the low-symmetry crystal field. The curves differ significantly from those in figure 4 of Takeuchi *et al* (1999). Although we use different parameters (see table 2), this difference is significantly due to the now correct level order (under spin–orbit) being used.
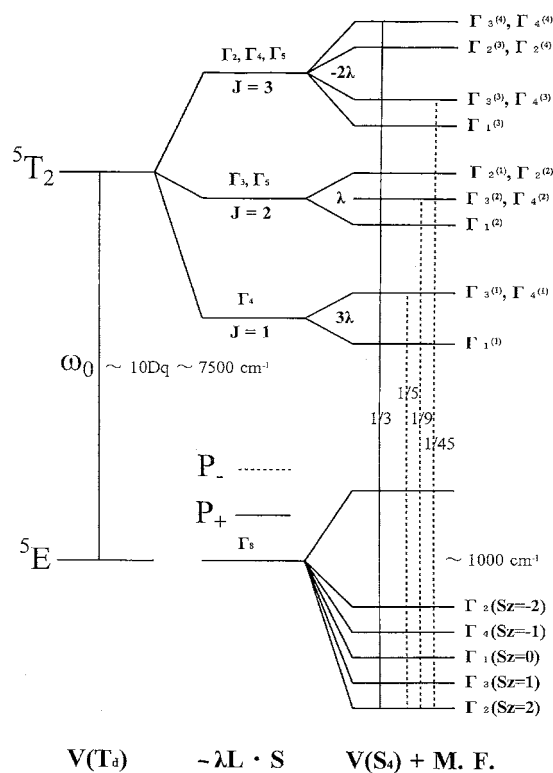
Figure 1. A schematic energy diagram of a tetrahedral Co^{3+} ion under the spin–orbit interaction, the low-symmetry crystalline field (V_{S_4}) and the molecular field (MF). The diagram includes allowed transitions for the P$_+$ (——) and P$_-$ (- - - -) polarizations of light, at 0 K.
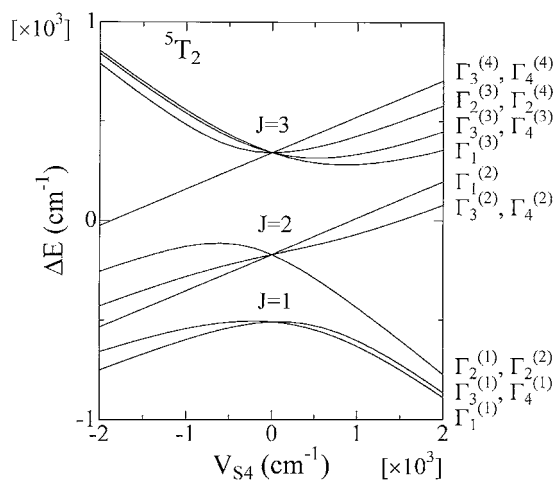

Figure 2. Calculated energy levels of the $^5T_2$ excited state of the Co^{3+} ion for varying strengths of the low-symmetry crystalline field (V_{S_4}). In this calculation we neglect the molecular field.

Table 1. The selection rules for left (P$_-$) and right (P$_+$) polarized light.

P$^+$: Γ_4	P$^-$: Γ_3
$\Gamma_1 \longrightarrow \Gamma_4$	$\Gamma_1 \longrightarrow \Gamma_3$
$\Gamma_2 \longrightarrow \Gamma_3$	$\Gamma_2 \longrightarrow \Gamma_4$
$\Gamma_3 \longrightarrow \Gamma_1$	$\Gamma_3 \longrightarrow \Gamma_2$
$\Gamma_4 \longrightarrow \Gamma_2$	$\Gamma_4 \longrightarrow \Gamma_1$

The lower-symmetry crystal-field interaction also affects the ground state, splitting it into two levels $^5\Gamma_1$ and $^5\Gamma_2$. Experimentally the former is known to be of lower energy (Wood and Remeika 1967), approximately 1000 cm^{-1} lower.

The final splitting that we consider affects the ground state, and is associated with the molecular field originating from the sublattice magnetization of Fe^{3+}. This perturbation is small but nevertheless plays an important role in the modelling of the temperature dependence of the FR, as we shall describe in section 4. Although from our point of view this is the most significant effect of the molecular field, the field also causes the excited $\Gamma_3 + \Gamma_4$ levels to split (as seen in figure 3 of Takeuchi *et al* 1999).

Now let us turn to the exact form of the Hamiltonian we use, so that we can describe the parameters we are using in the model. To obtain the eigenvalues and eigenfunctions of the excited state $^5T_2$, the matrix of spin–orbit and S_4 crystal-field interactions needs to be diagonalized, as in Saito *et al* (1987) and Takeuchi *et al* (1999). The major crystal-field term is assumed to be associated with the spherical harmonic function $Y_0^{(2)}(\theta, \phi)$, so the first-order perturbation to the Hamiltonian is

$$\mathcal{H}' = \sum \zeta (l \cdot s) + B \sum Y_0^{(2)}(\theta, \phi). \tag{2.1}$$

B is a simple coefficient which is a measure of the departure from the tetrahedral field. Matrix elements of this Hamiltonian for the excited states contain a λ term associated with the spin–orbit interaction, and a second term in the form of a coefficient multiplied by a reduced matrix element and by B. The reduced matrix element, $\langle \alpha (t_2^4 e^2)^5 T_2 \| Y^{(2)} \| \alpha (t_2^4 e^2)^5 T_2 \rangle$, is a constant for all of the $^5T_2$ states, so we combine the reduced matrix element and B into a new parameter V_{S_4}.

The molecular-field perturbation is added afterwards in the form

$$\mathcal{H}'' = -AS_z \mathcal{H}_z^{ex}. \tag{2.2}$$

A is an exchange parameter proportional to the exchange interaction between Co^{3+} and Fe^{3+}. The exchange Hamiltonian, $\mathcal{H}^{ex}$, will be discussed more in section 4. Most significantly, it splits the ground state multiplet into five levels, associated with spin projections of the $S = J = 2$ into $S_z = J_z = -2, -1, 0, 1, 2$ from top to bottom.

3. Allowed transitions

FR depends on the different transitions allowed for left and right circularly polarized light. The transitions for left and right circularly polarized light transform according to Γ_3 and Γ_4 respectively, and the transitions allowed are therefore as given in table 1. The left and right circularly polarized lights are associated with P$_-$ and P$_+$ respectively. In figure 3 we give strengths for the transitions from the ground levels. These transition strengths are given as a function of the low-symmetry field (V_{S_4}), and are given for both P$_+$ and P$_-$ polarizations. This figure differs from the corresponding figure 5 of Takeuchi *et al* (1999), and again it is not entirely due to the use of different parameter values. In particular, the $\Gamma_1(S_z = 0)$ curves for the two polarizations should coincide, but do not in Takeuchi *et al* (1999).
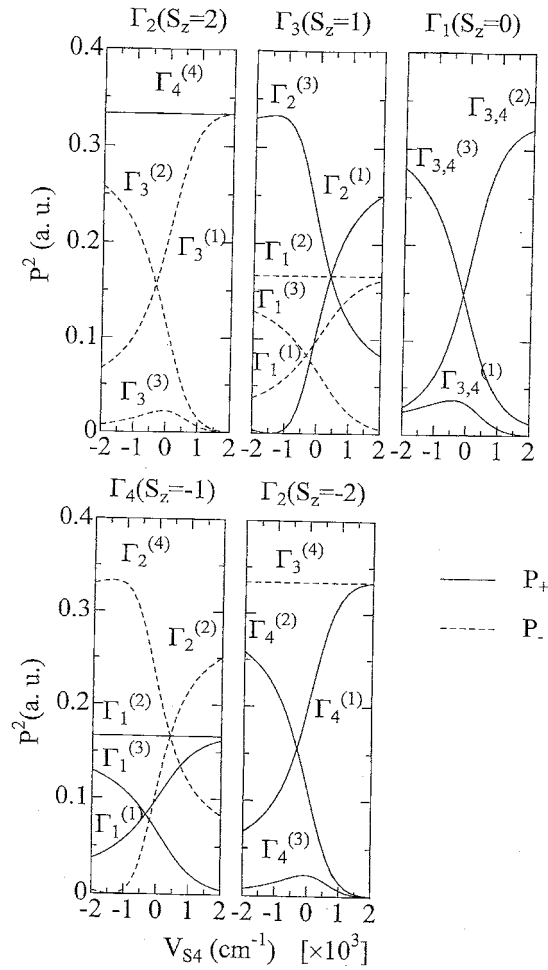
Figure 3. Strengths for P$_+$ (——) and P$_-$ (- - - -) transitions from the ground levels for varying strengths of the low-symmetry crystalline field (V_{S_4}).

4. The temperature dependence of the Faraday rotation

The predominant influence on the FR, as regards temperature variation, is through the occupancy of the ground states. Recalling from section 2 that the levels of the ground state multiplet are associated with spin projections, the electron population probability for the S_z-state is

$$\rho_{S_z} = \frac{\exp(-E_{S_z}/kT)}{Z} \tag{4.1}$$

where the partition function is

$$Z = \sum \exp\left(-\frac{E_{S_z}}{\kappa T}\right). \tag{4.2}$$

E_{S_z} is the energy of the S_z-level in the ground multiplet. κ is the Boltzmann constant.
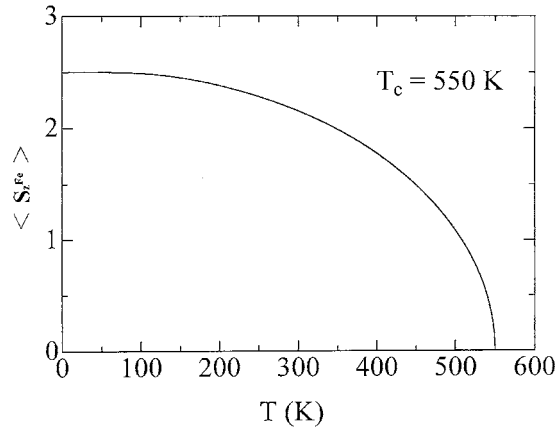
Figure 4. The temperature dependence of the thermal average value of an iron spin. We have taken the Curie temperature, T_C, to be 550 K.

At a given temperature T and frequency ω, the FR is given by

$$\theta(\omega) = \frac{C}{2} \sum_{i,S_z} \rho_{S_z} \frac{\omega^2(\omega(i)^2 - \omega^2 - \Gamma(i)^2)(P_-(i)^2 - P_+(i)^2)}{(\omega(i)^2 - \omega^2 + \Gamma(i)^2)^2 + (2\omega\Gamma(i))^2} \tag{4.3}$$

where C is a constant and $P(i)^2$, $\omega(i)$ and $\Gamma(i)$ represent the transition strengths, the transition frequency and the damping constant for the ith transition, respectively.

Setting the average energy of the ground multiplet to zero, the energies in the multiplet are given by

$$E_{S_z} = -A\langle S_z^{\text{Fe}}\rangle S_z \tag{4.4}$$

where $\langle S_z^{\text{Fe}}\rangle$ is the thermal average value of an iron spin S_z. We take $\langle S_z^{\text{Fe}}\rangle$ to be (Shinagawa *et al* 1997) given by the solution to the pair of equations

$$\langle S_z^{\text{Fe}}\rangle = S \cdot B_S(x), \qquad S = \tfrac{5}{2} \tag{4.5}$$

$$x = \frac{3\langle S_z^{\text{Fe}}\rangle}{S+1} \frac{T_c}{T}. \tag{4.6}$$

The use of the Brillouin function for $S = 5/2$ arises due to the adopting of the molecular-field approximation. We have used a value of $T_c = 550$ K for the Curie temperature. This corresponds to a temperature dependence for $\langle S_z^{\text{Fe}}\rangle$ as shown in figure 4.

5. Our parameters

In figures 5 we present the model fitted to the experimental data at $T = 300$, 180 and 60 K. The parameters used for those fits are given in table 2.

Having corrected the calculations of Takeuchi *et al* (1999), we see that improvements to the fitting can be made. Noticeably the new parameters allow for dampening of the lower-wavelength peak, while maintaining the sharpening of the higher-wavelength peak. A modification of the model to shift the centre of the spectra to lower wavelengths as the temperature decreases is mathematically simple, but this issue seems relatively insignificant and the effect is not well quantified. It is more important that the calculated left low-temperature curve is noticeably to the right of the experimental data. This is the main aberration, although
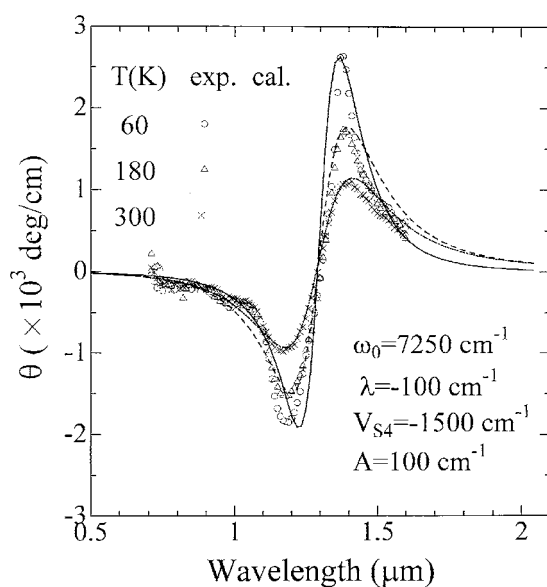
Figure 5. The fitted calculated curves and the experimental FR at the temperatures 60, 180 and 300 K.

Table 2. The parameters used to calculate the FR spectra.

T	ω_0	Γ_1	Γ_2	Γ_3	λ	$V(S_4)$	A
60	7250	1050	1050	525	-100	-1500	100
180	7250	1600	1600	700	-100	-1500	100
300	7250	1800	1800	850	-100	-1500	100

the slopes off to the right of all the curves on the right are somewhat inconsistent with the experimental data. It would seem that a refinement of the model is required. It is difficult to determine whether the parameters would retain significance in a modified model, since almost as good fits can be obtained using different parameters.

6. Summary

We have corrected the calculations of Takeuchi *et al* (1999) and reanalysed the temperature dependence, in the range 60–300 K, of the FR of a Co^{3+}-substituted magnetic garnet system.

We are able to model some of the characteristics of the FR curve's temperature dependence but have difficulties with some others. A modification to the model is needed to rectify the discrepancies.

Acknowledgments

References

Egashira K and Manabe T 1972 *IEEE Trans. Magn.* **50** 646–8

McAven L F and Shinagawa K 2001 presented at *1st Joint European Magnetic Symp. (Grenoble, Aug. 2001)*

Saito T, Tamanoi K, Shinagawa K and Tsushima T 1987 *J. Magn. Soc. Japan* **11** 245–8

Shinagawa K, Matsubayashi S, Ando K, Saito T and Tsushima T 1999 *J. Magn. Magn. Mater.* **196–197** 595–6

Shinagawa K, Simsa Z, Saito T and Tsushima T 1997 *Japan. J. Appl. Phys.* **36** L213–15

Sugano S, Tanabe Y and Kamimura H 1970 *Multiplets of Transition-Metal Ions in Crystals* (New York: Academic)

Takeuchi A, Ando K, Shinagawa K, Saito T and Tsushima T 1999 *IEEE Trans. Magn.* **35** 3139–41

Wood D L and Remeika J P 1967 *J. Chem. Phys.* **46** 3595–602