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## Temperature and high magnetic field dependence of the Faraday rotation in the terbium iron garnet

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**Résumé.** — La rotation Faraday a été mesurée à  $1,15 \mu$  dans le ferrite grenat de terbium, suivant la direction [111]. Deux types d'expériences ont été réalisées : en champ continu faible (20 kOe), l'évolution de la rotation a été étudiée entre 4,2 et 300 K ; en champ magnétique intense transitoire (1 200 kOe) la variation de la rotation sous l'action du champ a été mesurée à la température ambiante. L'ensemble des résultats est analysé en fonction des contributions induites par les transitions dipolaires magnétiques et électriques qui sont proportionnelles aux aimantations des sous-réseaux magnétiques. Les contributions des sous-réseaux  $\text{Fe}^{3+}$  sont supposées identiques à celles du YIG et les contributions des ions  $\text{Tb}^{3+}$  sont calculées. Pour l'ion terre rare, la contribution des transitions dipolaires magnétiques est calculée en tenant compte de l'influence du champ cristallin sur le facteur de Landé. Celle des transitions dipolaires électriques apparaît comme une fonction de la température et du champ appliqué.

**Abstract.** — The Faraday rotation of terbium iron garnet at  $1.15 \mu$  wavelength is presented. The experimental results have been obtained along the [111] easy direction both in low d.c. field 20 kOe as a function of temperature between 4.2 and 300 K and at room temperature as a function of the applied field (up to 1 200 kOe). The results are analysed in terms of magnetic and electric dipole contributions from the three magnetic sublattices. The  $\text{Fe}^{3+}$  contributions are considered to have the same values as in YIG and the total  $\text{Tb}^{3+}$  contributions are obtained. The contribution of the magnetic dipole transitions of the R.E. has been calculated when the influence of the crystalline field effect on the Landé factor is taken into account ; the electric dipole contribution is found to be strongly temperature and magnetic field dependent.

**Introduction.** — Since the first crystallographic description by Bertaut and Forrat [1] and the first magnetic property analysis by Pauthenet [2], the rare-earth iron garnets (RIG) and the yttrium iron garnet (YIG) have been extensively studied. From the optical property investigation by Dillon [3], it was shown that the ferrimagnetic garnets exhibited a strong circular magnetic birefringence (CMB) in the near infrared and visible ranges. Later Matthews *et al.* [4] established that the Faraday rotation was directly connected with the magnetic properties.

The unit cell contains eight formula  $\text{R}_3\text{Fe}_5\text{O}_{12}$  with 24  $\text{Fe}^{3+}$  ions in the d sites (tetrahedral), 16  $\text{Fe}^{3+}$  in the a sites (octahedral) and 24  $\text{R}^{3+}$  in the c sites (dodecahedral). The sublattice magnetizations are noted  $\mathbf{M}_d$ ,  $\mathbf{M}_a$ ,  $\mathbf{M}_c$  respectively. The  $\text{Fe}^{3+}$  ions in

the two different sites are strongly coupled anti-ferromagnetically by their own mutual interactions ; the moment of the rare-earth sublattice is antiparallel to the resultant  $\text{Fe}^{3+}$  magnetization, there is a compensation temperature for which the ferrite magnetization is zero ; below the compensation temperature, the ferrite magnetization direction is given by  $\mathbf{M}_c$ .

For all temperatures we can write :

$$|\mathbf{M}_{\text{ferrite}}| = |(\mathbf{M}_d - \mathbf{M}_a) - \mathbf{M}_c|. \quad (1)$$

The RIG have [111] easy, [100] hard and [110] intermediate directions.

As discussed in [5] and [6], the Faraday rotation  $\Phi_F$  is induced both by the electric and magnetic dipole transitions and is proportional to the three sublattice magnetizations.

$\Phi_F$  can be written for temperature  $T$  greater than the compensation temperature :

$$\Phi_F = \Phi^e + \Phi^m = -(A_e + A_m) |\mathbf{M}_a| + (D_e + D_m) |\mathbf{M}_d| - (C_e + C_m) |\mathbf{M}_c|. \quad (2)$$

Below  $T_{\text{comp}}$ , all the moments change sign,

$$\Phi_F = \Phi^e + \Phi^m = (A_e + A_m) |\mathbf{M}_a| - (D_e + D_m) |\mathbf{M}_d| + (C_e + C_m) |\mathbf{M}_c|. \quad (3)$$

In the eqs. (2) and (3),  $A_e$ ,  $D_e$ ,  $C_e$ ,  $A_m$ ,  $D_m$ ,  $C_m$  are the M.O. coefficients induced by the electric and magnetic dipole transitions in the octahedral, tetrahedral and dodecahedral sites respectively :  $|\mathbf{M}_a|$ ,  $|\mathbf{M}_d|$  and  $|\mathbf{M}_c|$  are given in Bohr magnetons.

The magneto-optical effects are associated with the influence of the magnetic perturbation due to the electron spins on the permittivity tensor  $\varepsilon_{ij}$  of the medium. In the dia- or paramagnetic systems, the tensor  $\varepsilon_{ij}$  is directly related to the applied field. In ferromagnetic crystals  $\varepsilon_{ij}$  is determined by the spins of the unfilled shells coupled by exchange interactions ; thus the electric dipole induced by the electric dipole transitions is spin dependent. Details about the origin of the Faraday rotation are given in the ref. [7]. Note that by using the hypothesis that the magnetization  $M$  is proportional to the spin  $S$ , the relation  $\varepsilon_{ij}(S)$  becomes equivalent to  $\varepsilon_{ij}(M)$ . For a ferrimagnetic crystal, such a relation is valid for each magnetic sublattice and the Faraday rotation associated to the electric dipole transitions is found to be proportional to the corresponding magnetic moments as expressed in the relations (2) and (3).

In this paper, we report experimental results obtained along the [111] easy direction under different conditions (Section 1). The magnetic and magneto-optic properties of the  $\text{Tb}^{3+}$  ions are described using some assumptions which are discussed in the section 3. The influence of a low d.c. magnetic field on the Faraday rotation is presented in section 2 ; the rare-earth contribution to the total rotation can therefore be deduced. Under low external fields, the temperature dependence of either the *magnetic rotation* (Section 4) or the *electric rotation* (Section 5) will be determined and discussed. In the sections 6 and 7, the high magnetic field dependences of these two contributions are analysed in detail at room temperature. Finally conclusions regarding previous works on the same subject are presented ; note that the model proposed in this paper has been used to analyse the Faraday rotation evolutions in other heavy rare-earth garnets.

**1. Experimental.** — We have studied the Faraday rotation of TbIG when the applied field  $H_a$  is parallel to the [111] direction ; the experiments were performed at  $1.15 \mu$  wavelength. This wavelength lies at the

edge of the garnet window [3]. In low continuous magnetic fields up to 20 kOe, the measurements of the Faraday rotation were made on single crystals in the 4.2-295 K range. Some experiments have been performed when the applied field is parallel to the [001] direction but these results are only given for comparison and will not be analysed in detail in this paper. At room temperature, the Faraday rotation was measured in very high transient pulsed fields up to 1 200 kOe on slices up to  $100 \mu$  thick, the field was produced by capacitor bank discharge in a single turn coil [8].

Magnetic measurements along the direction [111] have been presented in a previous paper [9].

**2. Spontaneous Faraday rotation in TbIG.** — The figures 1 and 2 show the variation of  $\Phi_F$  versus the applied field  $H_a$  ; for the [111] direction,  $\Phi_F$  is linear ; for the [001] direction, the evolution of  $\Phi_F$  is found to be complex. To determine the spontaneous Faraday rotation,  $\Phi_F$  (TbIG), defined as the rotation associated with the spontaneous magnetization, we extrapolate the linear part of the curves to  $H_a = 0$ .  $\Phi_F$  (TbIG) versus temperature is plotted in figure 3. If we define the rotation in YIG as positive [5], we see on figures 1 and 2 that the Faraday rotation in TbIG is negative

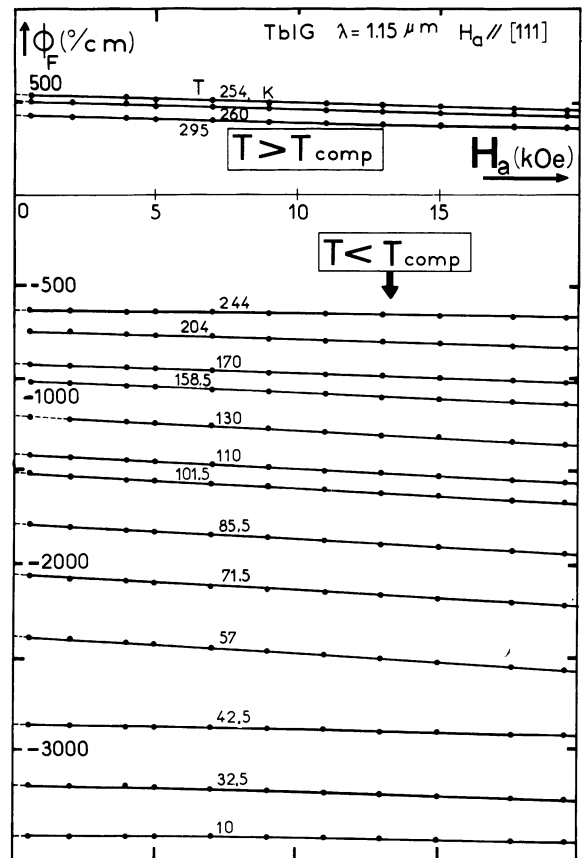


Fig. 1. — Magnetic field dependence of the Faraday rotation in TbIG up to 20 kOe at  $1.15 \mu$  wavelength in the 10-295 K temperature range for the field applied along the [111] direction.

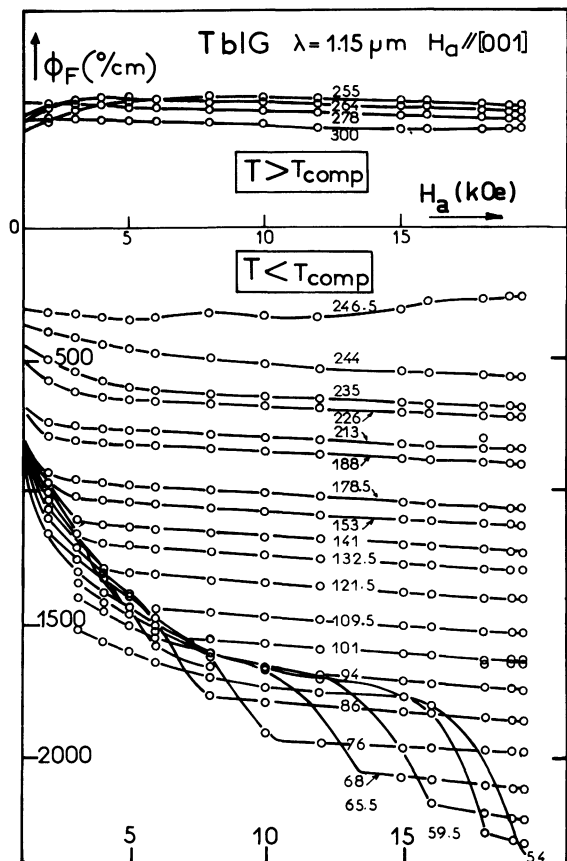


Fig. 2. — Magnetic field dependence of the Faraday rotation in TbIG at 1.15 μ wavelength in 57-300 K temperature range for the field applied along the [001] direction.

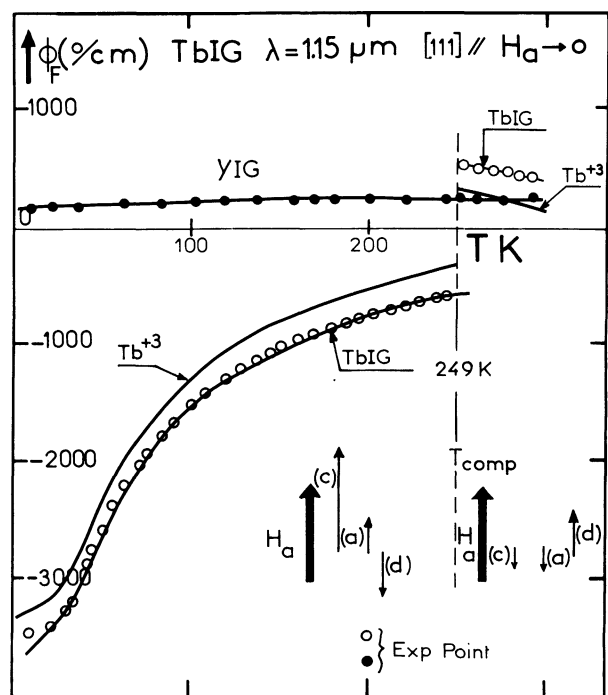


Fig. 3. — Temperature dependence of the spontaneous Faraday rotation in YIG and in TbIG at 1.15 μ wavelength.

below the compensation temperature and positive above this temperature. This sharp change of sign is contained in eqs. (2) and (3).

As previously observed by Crossley and co-workers [5, 6] in the 100-450 K temperature range, the rotation in TbIG is much larger than that of YIG (for example, at 295 K,  $\Phi_F(\text{TbIG}) = 435 \text{ deg. cm}^{-1}$ ,  $\Phi_F(\text{YIG}) = 285 \text{ cm}^{-1}$  at 1.15 μ wavelength). The rapid increase below 200 K shows the strong effect of the  $\text{Tb}^{3+}$  ion : at very low temperature, the Faraday rotation becomes very large, 3 460  $\text{deg. cm}^{-1}$ .

For the field applied along [001] (Fig. 2), the Faraday rotation curves  $\Phi_F(H_a)$  become complex for  $T < 100 \text{ K}$ .

This evolution may be related to the Cotton Mouton effect ( $\Phi_{CM}$ ) results which have been published some years ago [10, 11] (Fig. 4). Such results are explained by the presence of the umbrella magnetic structure : below 100 K,  $\mathbf{M}_d - \mathbf{M}_a$  lies in the [111] direction while the  $\text{Tb}^{3+}$  moments have a conical arrangement (the cone angle is equal to 30°, the  $\text{Tb}^{3+}$  magnetic moment length is 8.5  $\mu_B$ , this is deduced from the component along [111] which is equal to 7.35  $\mu_B$ ) [12]. Therefore, when the magnetic field is applied along the [001] axis, the umbrella arrangement is slowly destroyed and all the magnetic moments tend to align parallel to  $H_a$ . Consequently, the tempe-

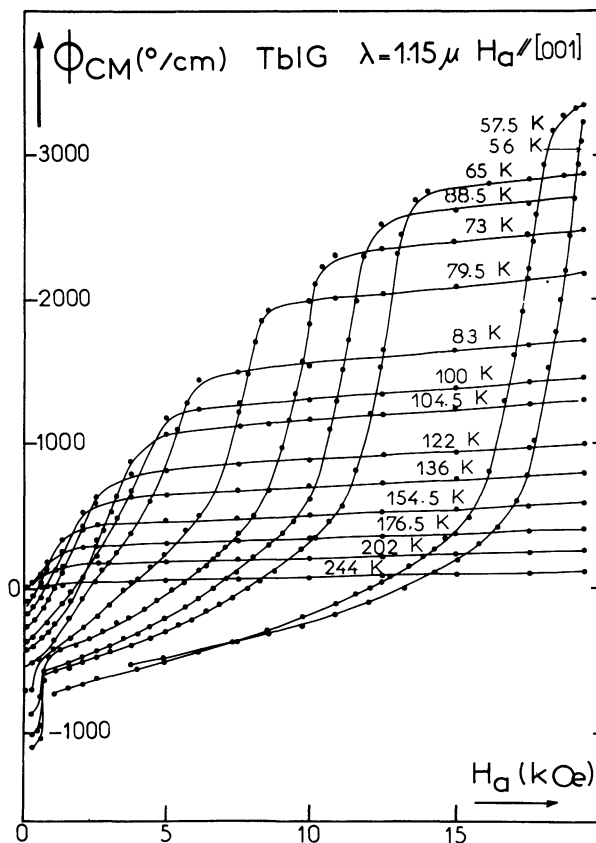


Fig. 4. — Magnetic field dependence of the Cotton Mouton rotation in TbIG at 1.15 μ wavelength in 57-295 K temperature range for the applied field along the [001] direction.

temperature evolutions of the saturation field either of  $\Phi_F$  or of  $\Phi_{CM}$  are related only to the magnetic structure evolution and must be identical. This point of view is confirmed by the experimental results as is shown on figure 5.

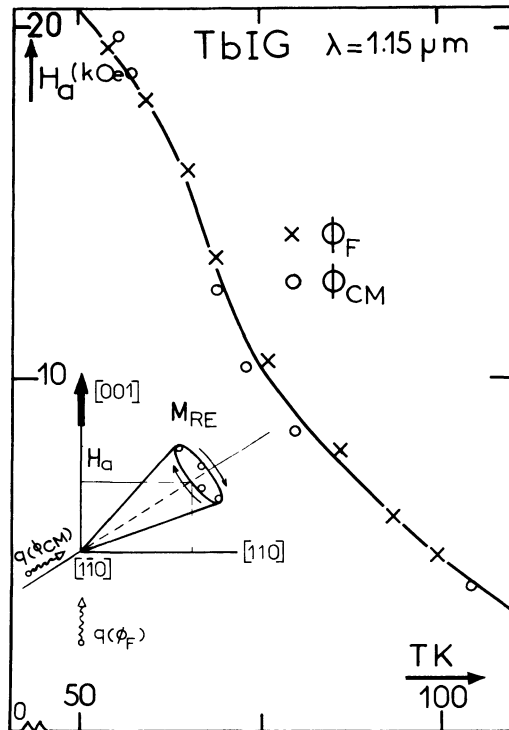


Fig. 5. — Temperature dependence of the saturation magnetic field either of the Faraday rotation or of the Cotton Mouton effect in TbIG; the field is applied along the [001] direction. In insert, the orientation of the *umbrella* magnetic structure of the  $Tb^{3+}$  sites is compared to both directions of the laser beam (Faraday and Cotton Mouton geometry).

**3. Hypothesis.** — To understand and analyse the rare-earth contribution to the Faraday rotation, we must separate the  $Fe^{3+}$  contributions from the total rotation. We suppose this contribution is the same in all rare-earth garnets. In other words, we take the first two terms of eq. (2) as for the Faraday rotation in YIG as written in [12].

This hypothesis can be justified from both optical and magnetic considerations. We have seen that the Faraday rotation in YIG is determined by the conjugate effect of the crystalline field and super-exchange coupling [14]. The similarity of the experimental absorption curves for the different garnets shows that the transition (with which the energy levels of the magnetic electrons of the  $Fe^{3+}$  are concerned) are weakly perturbed by the nature of the rare-earth ions [3, 15].

Previous absorptions measurements on YIG have been limited by the available sample thickness to below  $20\,000\text{ cm}^{-1}$  [3, 15, 16]. Wood and Remeika have related their results to an addition of crystal

field ( $3d \rightarrow 3d$  within  $Fe^{3+}$ ) and charge transfer { oxygen ( $2p$ )-iron ( $3d$ ) } transitions [15]. The charge transfer analysis have been discussed by Clogston [17] and by Wickerheim and Lefever [16]. The crystal field effects are interpreted in the  $10\,000\text{--}32\,000\text{ cm}^{-1}$  range with a splitting of the energy levels of the  $Fe^{3+}$  in the two magnetic sites (using first the cubic approximation, then the effects of a trigonal distortion and spin orbit interactions [15, 18]). More recently many iron garnets have been grown by epitaxy and spectra reported up to  $40\,000\text{ cm}^{-1}$  by Scott and Page [19]; a least square fitting was used to determine the major components of the YIG spectrum; all transitions below  $35\,000\text{ cm}^{-1}$  involve  $Fe^{3+}$  pairs while the first charge transfer transition between the oxygen  $2p$  band and the iron d-like bands occurs at  $35\,600\text{ cm}^{-1}$  (in this transition octahedral  $Fe^{3+}$  is involved).

One of the most interesting point of these spectra analyses is the important role of the superexchange term. For gallium doped YIG, Wood and Remeika [15] have observed that the intensities at several important points in the spectra are roughly proportional to the square of the fraction of  $Fe^{3+}$  ions present. This means that these intensities are characteristic of an  $Fe^{3+}\text{-O}^{2-}\text{-Fe}^{3+}$  association rather than an individual ion. On the other hand, Scott and Page [19] justify the most intense  $Fe^{3+}$  pair transitions by the fact that the dominant magnetic interactions between  $Fe^{3+}$  are superexchange through  $O^{2-}$ . Such a conclusion was already adopted by Wemple *et al.* [20]. All the previous results may be valid for the garnets where a rare-earth ion substitutes the  $Y^{3+}$  ion; the absorption spectra show in addition (in the  $10\,000\text{--}30\,000\text{ cm}^{-1}$  range) some rather sharp absorption lines which may be identified with electronic transitions ( $4f \rightarrow 4f$ ) [15]. With regard to the exchange interactions between  $Fe^{3+}$  ions, Pauthenet [2] has shown that the values of the Néel point lie in the  $560\text{ K}$  range whatever the rare-earth ion; its value is determined by a strong exchange term which describes the superexchange interactions  $Fe^{3+}\text{-O}^{2-}\text{-Fe}^{3+}$  and may be considered as a constant in the various garnets.

Finally, we may admit that the magnetic and magneto-optical properties of the  $Fe^{3+}$  ions are determined mainly by the crystal field and the super-exchange term and are independent of the nature of the rare-earth present in the dodecahedral site. A reasonable assumption is to consider that the contribution of  $Fe^{3+}$  ions to  $\Phi_F$  is the same in all rare-earth garnets; this contribution is considered to have the same value as measured in YIG [13, 14]. We obtain from the eqs. (2) and (3) :

$$|\Phi_F(Tb^{3+})| = |\Phi_F(TbIG) - \Phi_F(YIG)| = |(C_e + C_m)M_c| \quad (4)$$

where  $\Phi_F(Tb^{3+})$  represents the total Faraday rotation produced by the  $Tb^{3+}$  ions of the dodecahedral site.

In ref. [15], it was shown that changing the nature of the rare-earth produces a very small decrease in the frequency of the first absorption maximum of the  $\text{Fe}^{3+}$  ion; this frequency is equal to  $10\,900\text{ cm}^{-1}$  in YIG and  $11\,100\text{ cm}^{-1}$  in TbIG respectively. In the relation (4), we have assumed that to a first approximation, the total Faraday rotation produced by the  $\text{Fe}^{3+}$  ions is not modified by this small variation (less than 2%).

The eq. (4) shows that from the experimental variations of  $\Phi_F$  (TbIG) and  $\Phi_F$  (YIG) (as a function either of the applied field or of the temperature) we can deduce the different evolutions of the rare-earth contribution. In the following sections, the gyromagnetic and the gyroelectric contributions of the rare-earth sublattice are analysed.

**4. Temperature dependence of the « spontaneous magnetic » rotation in the dodecahedral site.** — In this section, the gyromagnetic contribution of the  $\text{Tb}^{3+}$  sublattice is calculated; it is given by  $|C_m M_c|$ .

Krinchik and Chetkin [21] have shown that in the infrared, the rotation in YIG does not depend on frequency (non *dispersive term*); this rotation is induced by the magnetic dipole transitions with an amplitude given, for a two sublattices system, by the following relation :

$$\Phi^m = -2\pi \frac{\bar{n}}{c} (\gamma_1 M_1 + \gamma_2 M_2) \quad (5)$$

which is valid for frequencies higher than the exchange resonance and where

$$\gamma_1 = g_1 e/2mc \quad \text{and} \quad \gamma_2 = g_2 e/2mc$$

are the gyromagnetic ratios of the two magnetic ions.

In TbIG, measurements of the Faraday rotation carried out in the wavelength interval from 6 to  $7.5\ \mu$  by Chetkin *et al.* [22] established that  $\Phi_F$  does not depend on frequency for  $\lambda > 6\ \mu\text{m}$ . (Experiments were run in the 25-350 K temperature range and in a field up to 8 kOe.)

The  $\text{Fe}^{3+}$  ions in the two different sites (a) and (d) are so strongly coupled by their mutual interactions that we may consider that the two  $\text{Fe}^{3+}$  sublattices form a rigid sublattice. In this case, eq. (5) established for a two sublattices system can be used in TbIG when we note  $\mathbf{M}_1 = \mathbf{M}_d + \mathbf{M}_a$  and  $\mathbf{M}_2 = \mathbf{M}_c$ . This choice remains valid in the presence of a very intense applied field (1 000 kOe). In a previous work [14], we have shown that the transition from the ferri-magnetic structure to a non colinear structure (with the angle between  $H_a$  and the  $\text{Fe}^{3+}$  sublattice moments is less than ninety degrees) occurs in the 1 700 kOe range. In other words, the first term of eq. (5) is proportional to the classical coefficients  $A_m$  and  $D_m$  of eqs. (2) and (3) with

$$|A_m| = |D_m| = 9.15\text{ deg. cm}^{-1} \mu_B^{-1} \quad [10].$$

The magnetic dipole transition coefficient in the dodecahedral site is now given from eq. (5) by :

$$C_m(\text{Tb}^{3+}) = -\frac{2\pi\bar{n}}{c} \cdot \gamma_2 = -\frac{\pi\bar{n}e}{mc^2} g_2 \quad (6)$$

which corresponds to the numerical values :

$$C_m = 4.575 \times g_2\text{ deg. cm}^{-1} \mu_B^{-1} \quad (7)$$

$$\Phi^m(\text{Tb}^{3+}) = 4.575 \times g_2 \cdot |\mathbf{M}_c| \quad (8)$$

where  $|\mathbf{M}_c|$  is always given in Bohr magneton. We have taken for the mean refractive index  $\bar{n}$  the YIG value : 2.2.

To obtain a quantitative agreement of experimental and theoretical values (given by eq. (8)), Chetkin *et al.* [22] had to assume that the  $g_2$  factor of the  $\text{Tb}^{3+}$  ion is less than its free ion value 1.5, and is equal to 1.1. This conclusion is confirmed by our magnetization and susceptibility measurements along the [111] direction [9]. The effects of the crystal field produced by the dodecahedrally surrounding  $\text{O}^{2-}$  ions are appreciable not only at low temperature but also in the 100-300 K range. Consequently, using the mean value of Chetkin *et al.* ( $g_2 = 1.1$ ), we can write from eq. (7),  $C_m = 5.03\text{ deg. cm}^{-1} \mu_B^{-1}$ . Note we have neglected the small temperature dependence of the  $g_2$  factor which was obtained by Chetkin *et al.* [22] (in the 100-300 K range, the variations are less than 5%).

The values of  $|\mathbf{M}_c|$  obtained along the [111] direction in zero external field are given in the table I of the ref. [9]; using these results and taking  $C_m = 5.03\text{ deg. cm}^{-1} \mu_B^{-1}$ , we calculate from the eqs. (7) and (8) the spontaneous magnetic contribution in the dodecahedral site as a function of the temperature. The figure 6 represents this variation; the magnetic contribution of the  $\text{Fe}^{3+}$  ions (issued from ref. [14]) have been also reproduced.

For comparison the experimental points (as simply photographed from the published graph) given by Chetkin *et al.* [22] have been plotted. The agreement between our curve and this experimental variation is acceptable. Note that these authors give no indication about the choice of the thermal variation of  $|\mathbf{M}_c|$ .

Table I. —  $A_e, D_e, C_e$  are given in  $\text{deg. cm}^{-1} \mu_B^{-1}$  for one mole  $2\text{ R}_3\text{Fe}_5\text{O}_{12}$ .

	$A_e$	$D_e$	$C_e$
TbIG (ref. [6])	$-9 \pm 15$	$-10 \pm 11$	$-84.4 \pm 2.5$
	5.3	- 6.36	- 89
	5	3.70	- 72
	- 438.5	- 319	- 108.6
	- 95.5	- 75.1	- 91
	- 85.88	- 63.9	- 85
	$-395 \pm 5$	$245 \pm 4$	- 40
YIG (ref. [6])	$-40.3 \pm 1.7$	$-21.4 \pm 1.2$	
(ref. [26])	$-50 \pm 3$	$-29 \pm 2$	

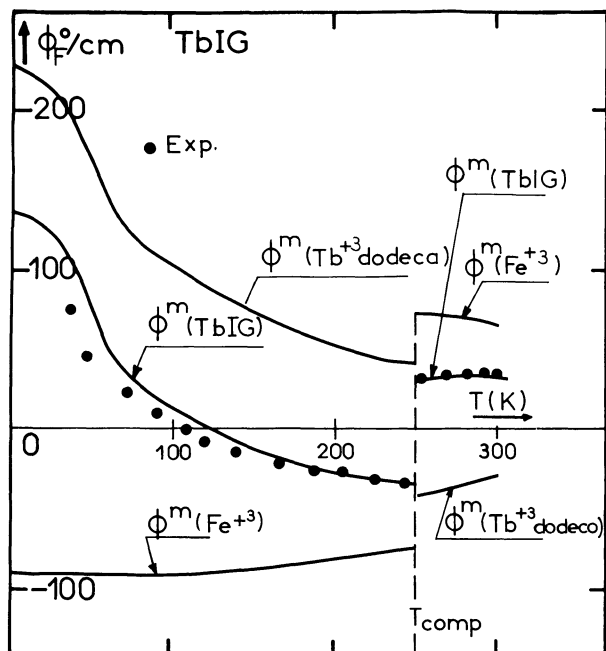


Fig. 6. — Temperature dependence of the *spontaneous magnetic* Faraday rotation for the  $Tb^{3+}$  sublattice and for the  $Fe^{3+}$  resultant moment.

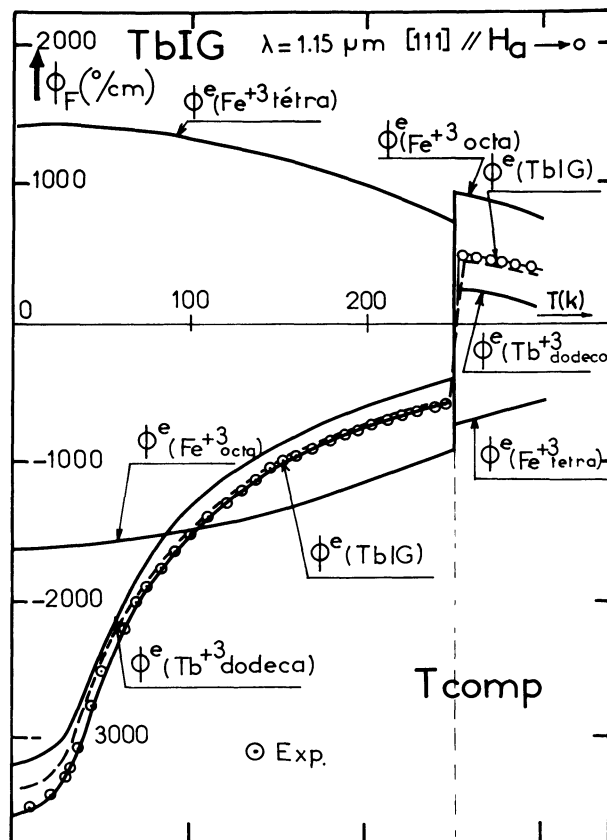


Fig. 7. — Temperature dependence of the *spontaneous electric* Faraday rotation for the  $Tb^{3+}$  sublattice and for the two  $Fe^{3+}$  sublattices.

**5. Temperature dependence of the « spontaneous electric » rotation in the dodecahedral site.** — The experimental and the theoretical values of the magnetic contribution for the  $Tb^{3+}$  ion have been established in the preceding paragraph, so we can obtain the spontaneous electric Faraday rotation,  $\Phi^e$ , in the dodecahedral site from the experimental total rotation given by the eq. (4). As shown on figure 7, one fact may be noted immediately : at  $1.15 \mu$ , the greater part of the rotation in TbIG is produced by the electric dipole transitions.

Therefore, the first order M.O. coefficient  $C_e$  induced by the electric dipole transitions may be deduced by comparing the values of  $\Phi^e$  with  $|M_c|$  given by the table I of the ref. [8] at each temperature. As shown on figure 8, the temperature dependence of  $C_e$  appears clearly ;  $C_e$  decreases from a maximum value ( $-86 \text{ deg. cm}^{-1} \mu_B^{-1}$ ) at very low temperature to  $-30 \text{ deg. cm}^{-1} \mu_B^{-1}$  at room temperature ; in the  $4.2\text{-}300 \text{ K}$  temperature range,  $C_e$  is a linear function of the temperature.

We may compare the magnitude of these electric dipole transition parameters with those which Cooper *et al.* [5, 6] have measured for  $Tb^{3+}$  in diamagnetic hosts (at  $1.15 \mu$  wavelength) :  $38.5$  and  $56.3 \text{ deg. cm}^{-1} \mu_B^{-1}$  in TbGaG and TbAlG respectively at  $300 \text{ K}$ . For TbIG, they have calculated  $C_e = -84.4 \pm 2.5 \text{ deg. cm}^{-1} \mu_B^{-1}$  with the following assumptions : the  $g_2$  factor of  $Tb^{3+}$  keeps its free ion value and  $C_e$  is temperature independent. Some comments about these results are given in the conclusion.

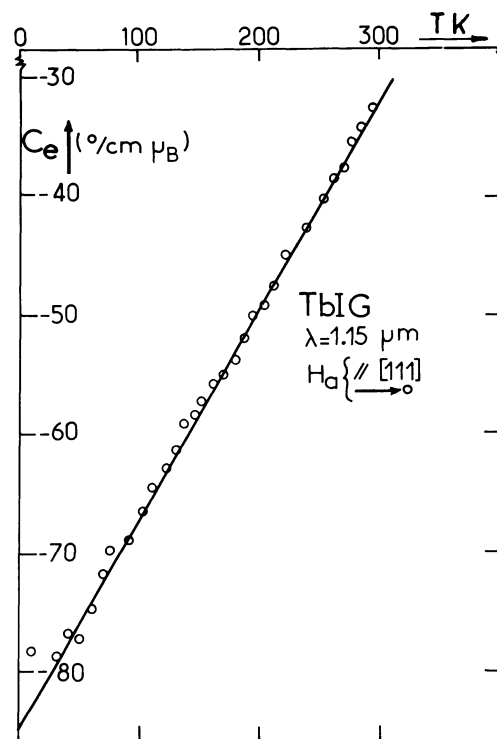


Fig. 8. — Temperature dependence of the first order M.O. coefficient induced by electric dipole transitions in the dodecahedral site ( $Tb^{3+}$  ion).

**6. High magnetic field dependence of the « magnetic rotation » of the Tb<sup>3+</sup> ions in the dodecahedral site.** — Consider, now, the evolution of the total Faraday rotation of the TbIG under the action of intense transient magnetic field at room temperature. We suppose that the eq. (4) remains valid and we can deduce the high field dependence of the Tb<sup>3+</sup> rotation from the experimental curves obtained for TbIG and YIG. In other words, we assume that the exchange field produced by the rare-earth sublattice on the two Fe<sup>3+</sup> sites may be neglected compared to the applied field. Consequently, the evolutions of the Fe<sup>3+</sup> sublattices are taken to be identical in both TbIG and in YIG.

The Tb<sup>3+</sup> magnetic contribution is given by eqs. (7) and (8) and to evaluate  $\Phi^m$ , we must analyse the magnetic field dependences of  $C_m$  and  $M_c$ . The magnetic field dependence of the first order M.O. coefficient,  $C_m(H_a)$  associated with the magnetic dipole transition can be deduced from the introduction of the Wangness [23] calculations in eq. (5) :

$$C_m = - 2 \pi \frac{\bar{n}}{c} \cdot \gamma_2 \left( 1 - \frac{\gamma_2 P^2}{\omega^2} \right) \quad (9)$$

with :

$$P = H_a + n(M_1 + M_2) \quad (10)$$

where  $M_1$  and  $M_2$  represent always  $M_d$ ,  $M_a$  and  $M_c$  respectively (as in relation (5)).

In eq. (10),  $n$  represents a mean molecular field coefficient such that the resultant of the molecular fields due to Fe<sup>3+</sup> (a) and Fe<sup>3+</sup> (d) respectively is given by  $n |M_d - M_a|$  [2].  $n$  is of the order  $17 \times 10^3 \text{ Oe } \mu_B^{-1}$  [2, 9], the correcting term :  $\gamma_2 P^2/\omega^2$  may be neglected for radiations in the optical range. From this, we can conclude that the change of the Tb<sup>3+</sup> magnetic Faraday rotation under high magnetic field is produced by the evolution of the sublattice magnetization  $M_c(H_a)$  only.

In zero external field, we have established that the temperature dependence of the spontaneous Tb<sup>3+</sup> sublattice magnetization can be written (eq. (2) of ref. [8]) :

$$|M_c| = \chi \cdot n |M_d - M_a| = \frac{C \cdot n}{T - \theta_p} \cdot M(\text{YIG}) \quad (11)$$

where the crystalline field effects are absorbed into the susceptibility  $\chi$ ;  $C$  represents the Curie constant determined along the direction [111];  $\theta_p$  is the paramagnetic temperature which is proportional to the superexchange interactions between rare-earth ions [2].

Numerically, we have found :  $C = 1.79 \times 10^{-2} \text{ deg. } (\mu_B \text{ mole}^{-1} \text{ Oe}^{-1})^{-1}$   $n = 16.3 \times 10^3 \text{ Oe } \mu_B^{-1}$ ,  $\theta_p = - 40 \text{ K}$  (these values correspond to the external field parallel to the easy [111] direction) [9].

The analysis of the temperature and field dependence of the magnetization of Néel ferrimagnets has been developed by A. E. Clark and E. Callen [24]. In TbIG at room temperature,  $M_c$ ,  $M_a$ ,  $M_d$  are colinear,

regardless of the applied field (the *angled* phase in which the sublattice magnetizations form angles with the field less than ninety degrees occurs in the vicinity of the compensation temperature 249 K). For  $H_a < n |M_d - M_a|$ ,  $M_c$  lies antiparallel to the field in a net field  $|n(M_d - M_a) - H_a|$ . In this region, the rare-earth moment decreases paramagnetically with increasing applied field. At  $H_a = n |M_d - M_a|$ , the rare-earth sublattice is demagnetized. For  $H_a > n |M_d - M_a|$ , further increase in field causes  $M_c$  to be parallel to the applied field. Therefore, we obtain from the eq. (11) (after numerical substitution) that the field evolution of the rare-earth sublattice is given at 300 K by :

$$|M_c(H_a)| = 5.265 \times 10^{-5} \{ \mp n(M_d - M_a) \pm H_a \} \quad (12)$$

where  $M_a$  and  $M_d$  are now functions of the applied field. We will use the variation of the sublattice magnetizations  $M_a$  and  $M_d$  which have been calculated in ref. [9] when the simplified hypothesis which neglects the intra-sublattice interaction is accepted. In figure 9, we have plotted the calculated field dependences of both sublattices and total magnetization of YIG up to 1 300 kOe at room temperature; the values of the exchange integrals which describe the

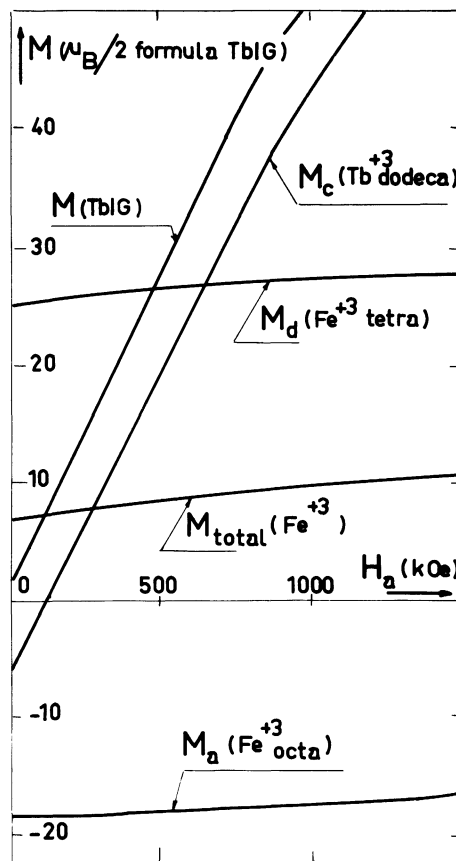


Fig. 9. — Calculated high magnetic field dependence of the three sublattices magnetization at room temperature.



superexchange have been taken to be  $J_{aa} = J_{dd} = 0$  and  $J_{ad} = 14 \text{ cm}^{-1}$  [14].  $M$  (YIG) increases from  $7 \mu_B$  ( $H_a = 0$ ) to  $10.5 \mu_B$  in a field up to 1 300 kOe. As we have  $(M_d - M_a) = 7.4 \mu_B$  when  $H_a = 110$  kOe, we deduce from the relation (12) that  $M_c(H_a)$  vanishes at this field value. In the high field range, we have for example,  $H_a = 1 000$  kOe,  $M_d - M_a = 9.75 \mu_B$ ,  $M_c = 44.2 \mu_B$ ;  $M$  (TbIG) =  $53.95 \mu_B$ . After multiplication of the values of  $|M_c|$  by

$$C_m = 5.03 \text{ deg. cm}^{-1} \mu_B^{-1},$$

we obtain the  $\text{Tb}^{3+}$  magnetic contribution as a function of the applied field (Fig. 10).

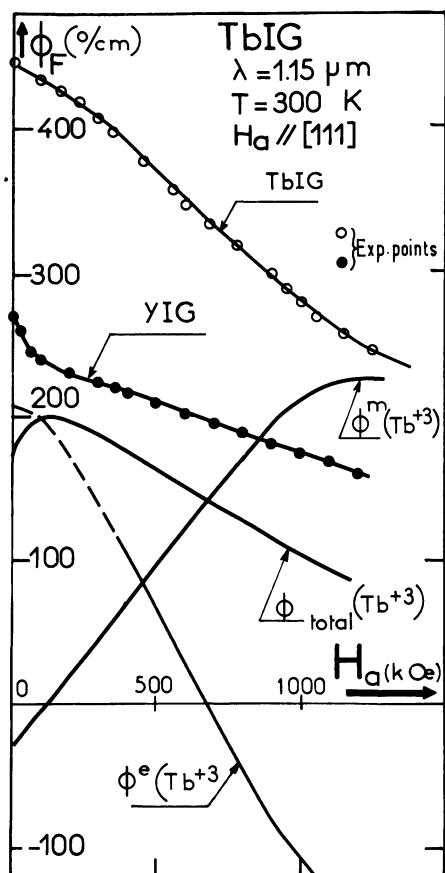


Fig. 10. — High magnetic field dependence of the Faraday rotation in TbIG and in YIG (at  $1.15 \mu\text{m}$  wavelength and 300 K : experimental results), calculated high field variation of both contributions (electric and magnetic) in the dodecahedral site.

**7. High magnetic field dependence of the « electric » rotation in the dodecahedral site.** —  $\Phi^m(H_a)$  has been calculated in the preceding section, so we can evaluate the rotation induced by electric dipole transitions,  $\Phi^e(H_a)$  from eq. (4) (where  $\Phi_F(\text{Tb}^{3+})$  is deduced from the experimental data in TbIG and YIG). The result is represented in figure 10.

We remark that  $\Phi^e(H_a)$  is zero in the 670 kOe range even though the field in which the rare-earth sublattice is demagnetized ( $n |M_d - M_a|$ ) is equal

to 110 kOe). Such a result could be explained in terms of a difference between the molecular field  $H_m$  and the exchange field  $H_{\text{exch.}}$ ; the latter is an effective field which acts on the total magnetic moment when the exchange field acts on spin moment only. From ref. [25],  $H_{\text{exch.}}$  is related to  $H_m$  by :

$$H_{\text{exch.}} = \frac{g_2}{2(g_2 - 1)} \cdot H_m.$$

Taking, as in section 4,  $g_2 = 1.1$  and  $H_m = 110$  kOe, we find  $H_{\text{exch.}} = 590$  kOe; this agrees reasonably with the value deduced from the experimental data. This result may be related to the fact that in systems of electrons coupled by exchange interactions, the electric dipole induced by the electric dipole transitions are proportional to the electric permittivity.

The figures 9 and 10 suggest an electric dipole coefficient strongly field dependent ( $\Phi^e(H_a)$  decreases when  $M_c(H_a)$  increases). This point seems to be confirmed when we return to the results obtained in d.c. fields (Section 1). Using the experimental variations of the figure 1,  $\Delta\Phi_F/\Delta H_a$  is found to be always negative; the variation  $|\Delta\Phi_F/\Delta H_a|^{-1}$  is a linear function of the temperature which is given by  $|\Delta\Phi_F/\Delta H_a|^{-1} = (T - \theta)/1.25$  (Fig. 11). (Note that for  $T > T_{\text{comp}}$  it is difficult to determine this evolution with accuracy.)

$$C_e(T) = C_e(0) + \alpha T \quad (13)$$

with  $C_e(0) = -86 \text{ deg. cm}^{-1} \mu_B^{-1}$  and

$$\alpha = 0.16 \text{ deg. cm}^{-1} \mu_B^{-1} \text{ K}^{-1}.$$

Therefore, from eqs. (2) and (3), we obtain :

$$\frac{\Delta\Phi_F}{\Delta H_a} = |(C_e(0) + C_m)\chi| + |\alpha T\chi| + |M_c| \frac{\Delta C_e}{\Delta H_a}. \quad (14)$$

We see that  $|\Delta\Phi_F/\Delta H_a|$  will be proportional to  $\chi$  if the compensation of the last two terms of eq. (14) occurs. Using :

$$\chi = \frac{1.79 \times 10^{-2}}{T + 40}$$

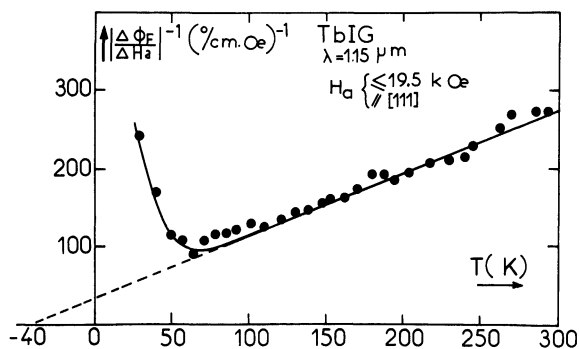


Fig. 11. — Temperature dependence of  $|\Delta\Phi_F/\Delta H_a|^{-1}$  deduced from the  $\Phi_F(H_a)$  curves (Fig. 1).

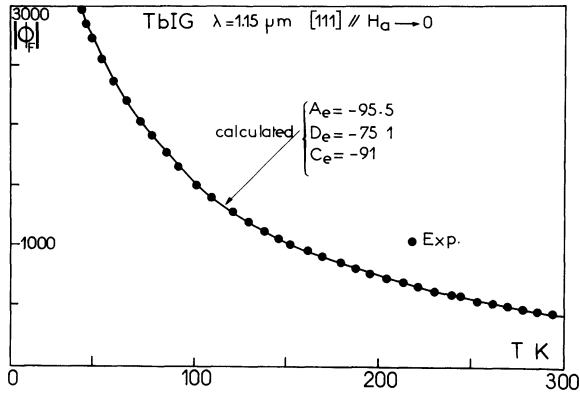


Fig. 12. — Temperature dependence of the absolute value of the spontaneous Faraday rotation in TbIG at 1.15 μm wavelength for the field applied along the [111] direction. The calculated curve is given by eq. (2) and (3) where the six coefficients  $A_e$ ,  $D_e$ ,  $C_e$ ,  $A_m$ ,  $D_m$ ,  $C_m$  are temperature independent.

(given by the magnetic study of ref. [9]) and  $\alpha = 0.16$ , this condition will be satisfied for :

$$\frac{C_e}{H_a} = \frac{-0.29 \times 10^{-2}}{(T + 40)} \times \frac{1}{M_c}$$

Finally, with the values of  $|M_c|$  given in ref. [9], we calculate  $\Delta C_e = -1$  (at 100 K),  $\Delta C_e = -2.3$  (at 200 K), and  $\Delta C_e = 4 \text{ deg. cm}^{-1} \mu_B^{-1}$  (300 K) when the field increases up to 10 kOe. Note that the first term of eq. (13) gives us :

$$\left| \frac{\Delta \Phi}{\Delta H_a} \right|^{-1} = \frac{T + 40}{1.40}$$

which is in good agreement with the experimental curves of figure 11 which can be fitted to :

$$\frac{T + 40}{1.25}$$

In view of the results, we can say that the first order M.O. coefficient  $C_e$  induced by the electric dipole transitions is at the same time temperature and applied field dependent.

**8. Conclusion.** — To separate the gyroelectric and the gyromagnetic contributions of the three magnetic sublattices to the Faraday effect, we made the assump-

tion that the temperature and the magnetic field affect the coefficients  $A_e$ ,  $D_e$  and  $C_e$ . Such an assumption is just the opposite of previous works where  $A_e$ ,  $D_e$  and  $C_e$  were obtained from a least square fit to sublattices magnetization data [5, 6, 26]. Some comments about this difference are thus necessary. Our precise magnetization data [9] have been taken into account to perform the least square analysis again. First, it is quite clear that the three parameter values which are given in refs. [5, 6] form a suitable solution of the problem. But it must be admitted that this solution is not unique; a list of solutions is proposed in the table I. We have no reasons to believe that all the possible solutions have been obtained. Nevertheless, it is clear that the sign of  $A_e$  and  $D_e$  cannot be found (either  $A_e$  and  $D_e$  both negative or  $A_e$  and  $D_e$  both positive are determined) and it is impossible to conclude that the  $\text{Fe}^{3+}$  contribution is positive when the magnetic moment is antiparallel to the light beam direction as is observed for paramagnetic ions [6].

The experimental curve perfectly fits the curve which is calculated for  $A_e = -95.5 \text{ deg. cm}^{-1} \mu_B^{-1}$ ,

$$D_e = -75.1 \text{ deg. cm}^{-1} \mu_B^{-1}$$

and

$$C_e = -91 \text{ deg. cm}^{-1} \mu_B^{-1}$$

Nevertheless, the quality of this agreement is accidental as it is proved in table II. We start from the experimental results ( $\Phi_F$ ,  $M_a$ ,  $M_d$ ,  $M_c$ ) which have been obtained at three temperatures 100, 204 and 294 K; the values relative to 100 and 294 K are supposed to be constant when a very small variation ( $10^{-3}$ ) either of  $M_a$  or of  $M_d$  or of  $M_c$  is simulated at 204 K ( $\Phi_F$  is taken to be unchanged). As is shown in the table II, the corresponding variations of  $A_e$ ,  $D_e$  and  $C_e$  are important. So we can conclude that the least square analysis where  $A_e$ ,  $D_e$  and  $C_e$  are temperature independent leads us to a great number of solutions for the fitting parameters and they have no correlation to the physical problem.

The fundamental hypothesis of our work is that we consider that in the rare-earth garnets, the total contribution to the Faraday rotation of the two  $\text{Fe}^{3+}$  sublattices is well represented by the YIG

Table II. — All the values are expressed relative to one mole  $2 \text{ R}_3\text{Fe}_5\text{O}_{12}$ ; the sublattice moments are in Bohr magneton;  $A_e$ ,  $D_e$ ,  $C_e$  in  $\text{deg. cm}^{-1} \mu_B^{-1}$ .

$T$	$M_a$	$M_d$	$M_c$	$A_e$	$D_e$	$C_e$
100	19.7	29.5	20.4			
294	17.78	25.1	6.45	- 95.5	- 75.1	- 91
204	19	27.64	10.6			
204	19 + 0.05	27.64	10.6	- 119	- 92.3	- 93.2
204	19	27.64 - 0.04	10.6	- 109	- 84.9	- 92.2
204	19	27.64	10.6 ± 0.02	- 102	- 80.4	- 91.7

Faraday rotation. In this case,  $C_e$  is found to be strongly temperature dependent. Since this publication was submitted for publication, the same model has been applied to other heavy rare-earth garnets : DyIG, HoIG, ErIG. In each of those, it has been shown that  $C_e$  presents a linear temperature evolution in the spontaneous state and is strongly applied field dependent. The first results relative to DyIG and HoIG have been published in the refs. [27, 28, 29]; they will be developed in detail in forthcoming papers together with the study of ErIG. So, it is clear that the TbIG properties are confirmed by the results which have been obtained on other rare-earth garnets and on YIG [13, 30].

Until now, we have been unable to explain the microscopic origin of temperature and applied field evolutions of the electric dipole transitions coefficients. As in the ferrite, the rare-earth presents a paramagnetic behaviour,  $C_e$  is equal to  $VT/\chi(T - \theta_p)$ , where  $V$  is the Verdet constant. Therefore,  $C_e$  have no temperature dependence for  $T > \theta_p$  if only  $\chi$  and  $V$  are exactly identical temperature functions [31]. Let us remark in this last reference that this conclusion was not verified perfectly, *the anomaly in  $\chi/V$  at room temperature might be blamed on a slight of absorption centres with temperature or something of the kind* (Van Vleck and Hebb in ref. [31]). The paramagnetic temperature dependent Verdet constant is given by the proportionality  $V \sim F(\mu H/kT) |1/(v^2 - v_t^2)|$  when the term  $F(\mu H/kT)$  describes the occupancy of various fundamental multiple levels (it is related to the magnetic susceptibility);  $v$  and  $v_t$  are the frequency of measurement and frequency of the main transition giving rise to the Faraday effect [32]. In the terbium aluminate garnet (TbAlG which is paramagnetic above 1.3 K),  $V$  has been measured in the visible spectral region (between 0.405 and 0.607  $\mu$ ) at different temperature values : 1.4 and 4.2 K [33], 77 and 300 K [34]. In this last temperature range,  $1/V$  is not proportional to  $1/\lambda^2$  as is the case at 1.4 and at 4.2 K [33]; therefore, the transition frequencies which are involved in the Faraday rotation are not temperature independent; in other words, we deduce from the results of refs. [33] and [34], that the product  $VT$  may be not a constant in the paramagnetic garnet.

In this work, we have analysed the temperature and magnetic field dependences of the contributions of the rare-earth  $Tb^{3+}$  ion in TbIG. Using the values of the rare-earth sublattice magnetic moment given by the magnetic study and the Lande factor value which takes the crystalline field effect into account, it is possible to calculate the magnetic dipole contributions as a function either of the temperature or of the applied field. Note that the temperature dependence of the Lande factor has been neglected in the calculation of the gyromagnetic contribution. Also the electric dipole transition contribution has been found to be strongly temperature and applied field

dependent. The whole study has been carried out with the justified hypothesis in which the  $Fe^{3+}$  contributions in TbIG are somewhat identical from those in YIG. Such a result confirms previous work on YIG [13, 30] and on other garnets [27, 28, 29].

Until now, the microscopic origin of the temperature and field dependences of the Faraday rotation has not been explained. If we summarize the theoretical situation, it has been shown theoretically that the rotation caused by an ion is proportional to its magnetic moment even in the presence of an exchange field [35], thus it is reasonable to analyse the rotation of the RIG in terms of sublattice magnetizations. Nevertheless, the understanding of the exact nature of the transitions giving the strong electric dipole transitions and high Faraday rotation remains unknown. Due to both electric and magnetic transitions, the interactions of the light with the crystal can produce elastic (phonons), magnetic (magnons), electric (plasmons and excitons) or electromagnetic (helicons) waves in the medium.

In the studies of spin-photon scatterings (in the visible and infrared range), the response of the medium can be described by the spin dependent electric polarizability [7, 35] and by the magnetic dipole contribution [21, 22]. As for the spin electric polarizability, T. Moriya [35] has proposed a theory of the Raman scattering of light for insulating compounds of transition elements; it is interesting to note that the scattering efficiency is found to decrease with increasing temperature in the antiferromagnetic iron-group fluorides (such a dependence is related to the temperature variation of the phenomenological uniaxial anisotropy constant). Further theoretical studies need to be done to determine the origin of the temperature and field evolutions of the Faraday rotation.

The temperature and field dependences of the Faraday rotation have been analysed in terms of sublattice magnetizations. It should be noticed that N. F. Kharchenko *et al.* [36] have introduced an additional term  $F(\omega)H$  to the eqs. (2) and (3) to investigate the magnetic field dependence of the Faraday rotation of GdIG in the vicinity of the magnetic compensation temperature (285.5 K) at 0.6328  $\mu$ . The electric dipole contribution of the gadolinium sublattice to the Faraday rotation is small in the visible region of the spectrum; so the additional term is due to the action of the magnetic field on the excited high temperature states of the crystal and  $F$  is given to be  $-2.3 \text{ deg.cm}^{-1} \mu_B^{-1}$  when  $C_e$  is equal to  $-2 \text{ deg.cm}^{-1} \text{ kOe}^{-1}$ . In TbIG the greater part of the terbium sublattice rotation is produced by the electric dipole transitions and it is reasonable to analyse the results with eqs. (2) and (3). Recently, a new form of additional terms has been proposed to analyse the Faraday rotation of YIG single crystal ( $\lambda = 0.6328 \mu$ ) [37]. The additional terms of eqs. (2) and (3) are assumed to be a linear combination of

$aM_a^2 H_{\text{eff}}$  and  $dM_d^2 H_{\text{eff}}$  where  $a$  and  $d$  are constant parameters and  $H_{\text{eff}}$  an effective field given by  $H - 4\pi M$  ( $M$  is the total magnetization per unit volume). Such a model seems to improve the quality of the *fit* and is preferred to a field dependence of  $A$  and  $D$  which is adopted in the ref. [38].

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