Symmetry analysis of the magnetic structures in TbIG and Tb:YIG at low temperature

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Abstract. Magnetic structures in TbIG and in terbium-yttrium ferrite garnet Tb$_{3.7}$Y$_{2.63}$IG are revisited using Bertaut’ representation analysis for the highest crystallographic subgroups of the space group Ia$\overline{3}$d with the wave vector $\mathbf{k} = 0$. Powder neutron diffraction experiments performed only on TbIG are combined with single crystal magnetization measurements. Three field-induced phase transitions are observed at 4.2 K in high magnetic fields up to 150 kOe along the $<001>$ direction while $<111>$ remain the easy axis of magnetization. New parameters of the previous Tb$^{3+}$ double umbrella magnetic structure are found at 5 K in a modified model of the rhombohedral subgroup R$\overline{3}$c. From magnetizations on Tb$_{0.37}$Y$_{2.63}$IG single crystal, non-collinear magnetic structure is evidenced at 4.2 K along the easy axis $<001>$. The parameters of a ‘square pyramid’ umbrella are predicted using both the basis vectors of the tetragonal subgroup $I_4/\text{acd}$ and a model based on the effective spin Hamiltonian with anisotropic exchange $G$ and magnetic $\tilde{g}$ tensors with the requirement $g_x G_x < g_y G_y << g_z G_z$. Anisotropy of magnetization is observed by comparison with the hard axis $<111>$ below $T_a = 80$ K where the earlier spin realignment phase transitions $<001>\leftrightarrow<uuw>\leftrightarrow<111>$ occur between $T_1 = 40$ K and $T_2 = 140$ K. The basis vectors of the monoclinic subgroup C2/c are tentatively proposed for the low symmetry angular phases $<uuw>$. All results are compared to previous works.

1. Introduction
The rare earth iron garnets $\{\text{RE}^{3+}\}[\text{Fe}^{3+}\text{Fe}^{3+}]\text{O}_{12}$ or REIG are ferrimagnetic materials with a crystallographic structure of the cubic space group Ia$\overline{3}$d-(O$\overline{h}$$_{10}$ No. 230, where the rare earth ions RE$^{3+}$ or the Yttrium Y$^{3+}$ are in the dodecahedral $\{24c\}(222)$ site and the two iron ions Fe$^{3+}$ in the octahedral $\{16a\}(\overline{3})$ and the tetrahedral $\{24d\}(\overline{4})$ sites [1 and Refs. herein]. They are now the subject of a great interest after the famous paper of Hur et al. [2] which have revealed that TbIG exhibits a magnetodielectric (MD) response at low temperature as well when a very small external magnetic field ($H_{ex} < 0.2$ T) is applied. This large MD effect appears to be correlated with the previous magnetostriction and high anisotropic behavior of this compound [3, 4]. Recently, Kang et al. [5] have discovered in a high external field up to 100 kOe a possible coupling between the magnetic exchange
and ligand-field excitations which occurs at a specific temperature range situated between 60 and 80 K with a distinct behavior above and below another characteristic temperature (140 K). In the same time, Louca et al. [6], using neutron scattering on TbIG single crystal have found a surprisingly result that the previously suggested reduced symmetry from the cubic to a rhombohedral structure in cooling below $T_N$ (568 K) cannot adequately describe the data in any range while a tetragonal phase is more appropriate. Zhang et al. [7], by introducing the spin fluctuations in the improved exchange field, the magnetic and magneto-optical properties of the Tb$^{3+}$ ions in TbIG and Tb:YIG have been calculated without taking into account the differences between the two non collinear magnetic structures which appear at low temperature. Due to these recent controversies, it is imperative to revisit the magnetic ordering of the Tb$^{3+}$ ions in TbIG and in a mixed terbium-yttrium ferrite garnet compound of the Tb$_{x}$Fe$_{3}$O$_{12}$ system (hereafter Tb$_{x}$Y$_{1-x}$IG) with precise experiments combined by symmetry analysis.

2. Experimental

The neutron diffraction experiments on TbIG powder sample were performed at ILL Grenoble, using the diffractometer D1B with the value of the wavelength $\lambda$ equal to 2.52 Å. All patterns are recorded in the 5–300 K temperature range. The resolution of the multidetector is equal to 0.2°. We present only the results of the treatment of the pattern at $T = 5$ K. The other patterns used to reexamine the temperature evolution of the double umbrella magnetic structure will be published elsewhere [8]. For the magnetic study the samples are two spherical single crystals. These two samples have been grown from a PbO/PbF$_{2}$/B$_{2}$O$_{3}$ solvent using a spontaneous nucleation method with the mixture of pure oxides Fe$_{2}$O$_{3}$, Y$_{2}$O$_{3}$ and Tb$_{2}$O$_{3}$. The electron microprobe analysis performed directly on the sphere gives the terbium concentrations ($x = 3$) for the pure TbIG and ($x = 0.37$) for the mixed compound Tb$_{0.61}$Y$_{2.63}$IG with respectively the associated weights and diameters: (0.4313 g; 5.5 mm) and (0.0413 g; 3.1 mm). The two samples was oriented along the [111] and [001] crystallographic directions by the X-ray Laue technique with an error less than 1°. The isothermal magnetizations $M_s(H)$ in function of the internal magnetic field $H$ (the applied field $H_a$ minus the demagnetizing field of the sphere) are performed at the LNCMI Grenoble. For $x = 3$, the high dc applied field produced by a Bitter magnet is varied by steps of 2 kOe up to 150 kOe at 4.2 K. For $x = 0.37$, a superconducting magnet with $H_a$ up to 80 kOe is used at 4.2 and 40 K and also around 80 K where the magnetization anisotropy is assumed to appear. Some of our unpublished $M_s(H)$ data [9] are also chosen in order to complete the study. All results are reported in Bohr magneton ($\mu_B$) for two formula unit’s 2Tb$_{1-x}$Y$_{x}$IG.

3. Results for TbIG

3.1. High field magnetization measurements at 4.2 K

The $M_s(H)$ curves at 4.2 K are plotted in the figure 1. The <111> direction is confirmed as the easy axis of magnetization. Along the <100> direction, when the field is increasing three critical fields $H_{c1}$, $H_{c2}$ and $H_{c3}$ appear at respectively ~ 20, 68 and 104 kOe. These results are slightly different to our previous results [3, 4] where only two field-induced phase transitions have been reported at $H_{c1}$ (66.66 kOe) and $H_{c2}$ (82.49 kOe). These transitions are characterized by a first order nature with the onset for the first time in dc fields of a hysteresis around $H_{c1}$ and also $H_{c2}$. The technical saturation magnetization $M_s[001] = 31.60 \mu_B$/mol is found for $H >> H_{c2}$ by extrapolating the $M_s(H)$ curve to zero internal field $H$. The resulting terbium magnetization $M_\text{ Tb} = 41.60 \mu_B$/mol leads to $6.93 \mu_B$/Tb$^{3+}$ related to a reduction of 23% by comparison with the free ion value ($9 \mu_B$).

3.2. Powder neutron diffraction pattern at 5 K

The pattern recorded at 5 K is presented in the figure 2. The observed reflections (hkl) are indexed with the general extinction rule ($h + k + l = 2n$) of Ia$\overline{3}$d with the wave vector $k = 0$. Four
superstructure lines \((110)^{2}, (310)^{2}, (411,330)^{2}\) and \((433,530)^{2}\) appear without ambiguity. Contrary to previous findings at \(1.5\) K \([10, 11]\) and \(4.2\) K \([3]\) we have also evidenced additionally the previous observed superstructure lines \((200)^{2}\) and \((600,442)^{2}\) with a sensibility \((0.5\) and \(1\%\) respectively to \((110)^{2}\)) in the same order of magnitude \([12]\). These two lines are not resolved at \(13\) K (not show) \([8]\).

3.3. Representation analysis

We have reported on table 1, the notations \(C_{j}\), \(C'_{j}\) \((j = 1-3)\) and \(\{i = 1-6\}\) \([13, 14]\), the numbers and the crystallographic positions of the RE\(^{3+}\) ions in the Wyckoff site \(\{24c\}\). The local axis \((U,V,W)\) in the \(D_{2h}(222)\) symmetry are identified to the parameters \(g_{\alpha}(\alpha = x,y,z)\) of the magnetic tensor \(\mathbf{g}\). The Bertaut’ representation analysis of the cubic space group \(Ia\overline{3}d-(O_{h}^{10})\) \([15]\) was applied in the past by Tchéou et al. \([10]\) and Bertaut et al. \([11]\) in order to determine the magnetic modes of the Tb\(^{3+}\) ions in TbIG. However, in their original works, two three-dimensional irreductible representations were used: \(\Gamma_{4g}\) = \(T_{1g}\) and \(\Gamma_{5g}\) = \(T_{2g}\). The representation analysis of \(Ia\overline{3}d\) has been developed later completely by us taking into account the coupling between the Tb\(^{3+}\) ions with the two iron ions Fe\(^{3+}\) in sites \([16a]\) and \(24d)\) \([8, 9]\). In the mixed Tb\(_{x}\)Y\(_{3-}\)IG system \((0 < x \leq 3)\), \(\Gamma_{4g}(T_{i})\) can describe in a first approximation the magnetic structures below \(T_{N}\). Using the earlier linear combinations of the spins \([15]\): \(F = +S + S_{j+1} + S_{j+3} + S_{j+6} + S_{j+9}\); \(G = +S - S_{j+1} + S_{j+6} - S_{j+9}\); \(C = +S + S_{j+1} - S_{j+6} - S_{j+9}\); and \(A = +S - S_{j+1} - S_{j+6} + S_{j+9}\) with \((j = 1-3)\), the basis vectors and the multiplicity \((m)\) for the RE\(^{3+}\) inequivalent sites are listed on table 2. The chemical cell is equal to the magnetic cell, thus the primitive translation noted \((1/2,1/2,1/2)\) of the crystallographic lattice \((I)\) is a primitive translation of the magnetic lattice \((I)\). The spin vectors \(S_{j}\) and \(S_{j+12}\) \((j = 1-3)\) are coupled ferromagnetically. The forbidden superstructure lines \((222)^{2}\) and \((622)^{2}\) are not observed in the figure 2. Then, the symmetry operation \((\overline{1}\) 0 0) is an inversion center and the magnetic modes \(C_{j}\) and \(A_{j}\) are equals to zero. The spins vectors \(S_{j}\) and \(S_{j+12}\) \((j = 1-3)\) are coupled ferromagnetically in the magnetic modes \(F\) and \(G\). Consequently, the site \(\{24c\}\) splits into six magnetically inequivalent sublattices noted \(C_{j}\) and \(C'_{j}\) \((j = 1-3)\) (see table 1) so that in any of a given point the crystalline field effect being exactly the same: \(C_{j}\) and \(C'_{j}\) are related to \(C_{i}\) by a rotation of \(120\) and \(240^\circ\) around the 3-fold symmetry \([111]\) axis (and respectively for \(C'_{1}\), \(C'_{2}\) and \(C'_{3}\)).

3.4. Investigation of the novel double umbrella for TbIG

![Figure 1. TbIG: \(M_s(H)\) curves at \(4.2\) K.](image)

![Figure 2. TbIG: D1B pattern at \(5\) K.](image)
The appearance of the three critical fields $H_c$, $H_c$, and $H_c'$ at 4.2 K along the <100> direction (see figure 1), confirms that the true subgroup of Ia $\overline{3}$ d is R $\overline{3}$ c-(D $\overline{6}$ d 3) No. 167, in agreement with the previous observed rhombohedral distortions [16]: the irreducible representation ($\Gamma_3 = T_1$) is reduced to ($\Gamma_2 = A_2$) + E, where the E modes are not considered. The preceding magnetic inequivalency becomes a crystallographic inequivalency: the sublattices (C) corresponding to the site 6e(2) and C'j to the site 6e'(2) of R $\overline{3}$ c in positions ±(X,1/2–X,1/4) (and cyclic permutation) with $X = 3/8$ and $X = 7/8$ respectively. The associated basis vectors described by the ferromagnetic mode $f_i = S_j + S_{j+6}$ (j = 1-3) in the rhombohedral axis X, Y, Z are gathered on table 2.

| Notations of the sublattices C_j, C'_j (j = 1-3) and {i=1-6} from refs. [13], [14] | Numbers and positions of the RE ions in the Wyckoff site {24c} | Local axis in D2(222) symmetry |
|---|---|---|
| C_1 | 1/8,0,1/4 | 100 |
| 3 | 7/8,0,3/4 | 100 |
| 1 | 5/8,1/2,3/4 | 011 |
| 4 | 3/8,1/2,1/4 | 011 |
| C'_1 | 3/8,0,3/4 | 100 |
| 2 | 5/8,0,1/4 | 011 |
| 4 | 7/8,1/2,1/4 | 011 |
| 3 | 1/8,1/2,3/4 | 011 |
| C_2 | 1/4,1/8,0 | 010 |
| 3 | 3/4,7/8,0 | 101 |
| 5 | 3/4,5/8,1/2 | 101 |
| C'_2 | 3/4,3/8,0 | 010 |
| 4 | 1/4,5/8,0 | 101 |
| 6 | 1/4,7/8,1/2 | 101 |
| C_3 | 0,1/4,1/8 | 001 |
| 5 | 0,3/4,7/8 | 110 |
| 1 | 1/2,3/4,5/8 | 110 |
| C'_3 | 0,3/4,3/8 | 001 |
| 6 | 0,1/4,5/8 | 110 |
| 2 | 1/2,1/4,7/8 | 110 |
| 24 | 1/2,3/4,1/8 | 110 |

We suggest a new combination of these magnetic basis vectors and we propose four modified double umbrella models for the six sublattices C_j and C'_j (table 3a). Around the 3-fold symmetry [111] axis, the sublattices C_j and C'_j are situated in the three glide planes e, e.3 and e.3' of the space group R $\overline{3}$ c.
Table 2. Splittings of Wyckoff positions, symmetry, multiplicity m and basis vectors.

| Space groups | Sites | Symmetry | m | Basis vectors of RE$^{3+}$ ions with k = 0 |
|--------------|-------|----------|---|-----------------------------------------|
| Ia 3 d\n($\Gamma_q = T_{1q}$) | 24c | (222) | 6 | $\Psi_{11} = F_{1x}$, $\Psi_{12} = F_{2x} + F_{3x}$, $\Psi_{15} = G_{2x} + G_{3y}$, $\Psi_{21} = F_{x}$, $\Psi_{22} = F_{y} + F_{y'}$, $\Psi_{25} = G_{x} + G_{y'}$, $\Psi_{31} = F_{z}$, $\Psi_{32} = F_{z}$, $\Psi_{33} = G_{z} + G_{y'}$ |
| R $\bar{3}$ c\n($\Gamma_s = A_s$) | 6e | (2) | 3 | $\Psi_{11} = f_{x} + f_{z}$, $\Psi_{12} = f_{z} + f_{x}$, $\Psi_{15} = f_{x} + f_{y}$, $\Psi_{21} = f_{x} + f_{x}$, $\Psi_{22} = f_{x} + f_{y}$, $\Psi_{25} = f_{x} + f_{x}$, $\Psi_{31} = f_{x} + f_{y}$, $\Psi_{32} = f_{x} + f_{y}$, $\Psi_{33} = f_{x} + f_{x}$ |
| C2/c\n($\Gamma_s = A_s$) | 8f | (1) | 2 | $\Psi_{11} = G_{x} + G_{y}$, $\Psi_{12} = G_{x} + G_{y}$, $\Psi_{15} = G_{x} + G_{y}$, $\Psi_{21} = f_{x} + f_{y}$, $\Psi_{22} = f_{x} + f_{y}$, $\Psi_{25} = f_{x} + f_{y}$, $\Psi_{31} = f_{x} + f_{y}$, $\Psi_{32} = f_{x} + f_{y}$, $\Psi_{33} = f_{x} + f_{y}$ |
| I4 / acd\n($\Gamma_s = A_s$) | 16e | (2) | 4 | $\Psi_{11} = G_{y} + G_{z}$, $\Psi_{12} = G_{y} + G_{z}$, $\Psi_{15} = G_{y} + G_{z}$, $\Psi_{21} = f_{x} + f_{y}$, $\Psi_{22} = f_{x} + f_{y}$, $\Psi_{25} = f_{x} + f_{y}$, $\Psi_{31} = f_{x} + f_{y}$, $\Psi_{32} = f_{x} + f_{y}$, $\Psi_{33} = f_{x} + f_{y}$ |
| 8b | (222) | 2 | $\Psi_{11} = G_{y} + G_{z}$, $\Psi_{12} = G_{y} + G_{z}$, $\Psi_{15} = G_{y} + G_{z}$, $\Psi_{21} = f_{x} + f_{y}$, $\Psi_{22} = f_{x} + f_{y}$, $\Psi_{25} = f_{x} + f_{y}$, $\Psi_{31} = f_{x} + f_{y}$, $\Psi_{32} = f_{x} + f_{y}$, $\Psi_{33} = f_{x} + f_{y}$ |

Table 3. TbIG: validation of the novel double umbrella model at 5 K.

| (a) Four modified models for the RE$^{3+}$ ions in $A_{2g}$ |
|-------|
| f′≠f and a≠a′ |
| (6e): $C_i : S_i = S_{i,a} = \hat{n} \pm a \mathbf{g}_i$ |
| C′$_i : m'_i = S_i = (f′ - a/3)\mathbf{n}$ |
| \(- (2\sqrt{2})/3 \hat{a} \mathbf{p}_i\) |
| $I_{600,442}^{(120)} \cap_1 = A(a - a')^2$ |
| Ag$^2$ with the coefficient A |
| = 256 \{0.275(Tb$^3$)$^2\}$ |
| $I_{600,442}^{(120)} \cap_1 = B(a - a')^2$ |
| Be$^2$ with the coefficient B |
| = 789.10 \{0.275(Tb$^3$)$^2\}$ |

Each glide plane containing [111] (unit vector $\mathbf{n}$) and one of the rhombohedral axis {1 1 1}, [1 1 1], [1 1 1] represented by the unit vectors $\mathbf{g}_j$, (j = 1-3). In this condition, we have: $c = (\mathbf{n}, \mathbf{g}_j)$, $c.3 = (\mathbf{n}, \mathbf{g}_j)$ and $c.3' = (\mathbf{n}, \mathbf{g}_j)$. The moments $C_1$ and $C_1'$ (figure 3) are drawn in the (Tl) plane associated to the glide plane c.3 which remains a principal plane of the magnetic tensor $\mathbf{g}$. In contrast, the cubic axis [100] and [011] cease to be principal axes. Furthermore, the perpendicular projection of [1 1 1] (unit vector...
The vector $g_1$ on the plane (111) is along the low symmetry axis $\hat{2}11$ (unit vector $p_1$) and equivalent directions. We choose the mixed system of crystalline axis between Ia $\bar{3}$ d and R $\bar{3}$ c: [111] (unit vector $m$); the low symmetry axis $\hat{2}11$ (unit vector $p_1$) and the perpendicular to the $(\pi)$ plane of the local axis [011] (not drawn in the figure 3). If one takes into account the smallness superstructure lines (200) and (600,442)* at 5 K (table 3b), new parameters are found for $C_1 (m, \theta_1, \phi_1)$ and $C'_1 (m', \theta', \phi')$ sites with a good reliability factor $R$ (table 3c) for a refinement based on all the reflections. The results obtained in the subgroup R $\bar{3}$ of Ia $\bar{3}$ d [12] lead to values of the components $m_{1x}$ and $m_{1z}$ above the free Tb$^{3+}$ ion value (9 $\mu_B$). Previous calculated temperature evolution [17] is not consistent with the onset of all the superstructure lines [8].

**Figure 3.** TbIG: the novel model at 5 K.

**Figure 4.** Tb$_{0.37}$Y$_{2.63}$IG: $M_s(H)$ at 4.2 and 40 K.

### 4. Results for Tb$_{0.37}$Y$_{2.63}$IG

#### 4.1. High field magnetization measurements

For Tb$_{0.37}$Y$_{2.63}$IG, two typical isothermal magnetization curves $M_s(H)$ at 4.2 and 40 K along [111] and [001] directions are presented in the figure 4. For $H$ parallel to [111], we observe an increasing of the magnetization corresponding to the rotation of the magnetization from the spontaneous direction. So, field-induced phase transitions at a critical field $H_c$ are observed at 4.2 (~ 17 kOe) and 40 K (~ 11 kOe) in increasing fields contrary to our previous magnetization measurements at 1.5 K on the same sample [18]. We can define by extrapolating the $M_s(H)$ curves to zero internal field $H$, firstly the spontaneous magnetization $M_s[111] = 2.86 \mu_B$/mol for $H < H_c$, and secondly the technical saturation magnetization $M_s[111] = 3.86 \mu_B$/mol for $H >> H_c$. The rapid approach to saturation observed along [001] shows that the easy axis is in this direction and remains until 40 K as evidenced by comparison of the curves and consequently, [111] is the hard axis. At 4.2 K, the value of $M_s[001] = 4.88 \mu_B$/mol gives the resulting $M_{Tb^{3+}} = 5.12 \mu_B$/mol with $<m>_{Tb^{3+}} = 6.92 \mu_B$/Tb$^{3+}$ which is in agreement with the above pure TbIG value contrary to that found previously [19] (6.52 $\mu_B$/Tb$^{3+}$) for a similar composition ($x \sim 0.37$). An average reduction of 23.1 % is obtained by comparison with the free ion. The terbium moments have a non
collinear ordering at 4.2 K. The thermal variation of the magnetization difference $\Delta M_1(T) = \{M_{[111]} - M_{[001]}\}(T)$ is plotted in the figure 5. We can observe a rapid decreasing in the curve after a maximum at $T_1 = 40$ K. A particular inflexion point is observed around 80 K and $\Delta M_1 = 0$ in the region above $T_2 = 140$ K when the [111] direction becomes the easy axis until room temperature.

Figure 5. $\text{Tb}_{0.37}\text{Y}_{2.63}$ IG: the anisotropic properties

This inflexion point is always present in the thermal variation of the second magnetization difference curve $\Delta M_2(T) = \{M_{[001]} - M_{[111]}(1/3)^{1/2}\}(T)$ (figure 5). A magnetization anisotropy is evidenced in the plot of the third magnetization difference curve $\Delta M_3(T) = \{M_{[001]} - M_{[111]}\}(T)$ which reaches a maximum value of 1.02 $\mu_B$/mol at 4.2 K. This new result gives the first confirmation that non collinear magnetic structures take place in the so-called angular phases $<uuw>$ below the typical temperature of anisotropy $T_a = 80$ K. The spin reorientation phase transitions $<001>\leftrightarrow<111>$ introduce another complexity due to the low symmetry of the $<uuw>$ phases [18, 20] where the iron sublattice magnetization $M_{Fe}$ lies in an arbitrary direction in one of the cubic $\{011\}$ type planes.

4.2. Representation analysis

Rhombohedral distortions have been observed until 50 K for $x = 1.17$ in the previous X-ray analysis of magnetoelastic distortions of the system $\text{Tb}_x\text{Y}_{3-x}$ IG for single crystals in the 5–300 K temperature range [21]. For $x \leq 0.7$ no tetragonal distortions which are about one order smaller than rhombohedral distortions have been revealed (also for monoclinic distortions). The high $<001>$, $<111>$ and low $<uuw>$ symmetry phases observed by magnetic measurements must be connected with the allowing maximal subgroups of $\text{Ia\bar{3}d}$. They are obtained by the ‘symmetry lowering device’ [22, 23]: the tetragonal $\text{I4}/\text{acd}$ ($\Gamma_2 = \text{A}_{2g}$) below $T_1$ and the rhombohedral $\text{R\bar{3}c}$ ($\Gamma_2 = \text{A}_{2g}$) above $T_1$ (table 2) are concerned. When a spin reorientation phase transition temperature region exists, the space group must be a subgroup of the two groups at the ends [24] with the requirement that the two ends of the spin reorientation phase transitions are characterized by second order nature in the $T$-range (40–140 K) [18]. Despite the previous theoretical predictions [20] that the nature of the upper transition at the end
of the spin reorientation phase transition \(<\text{uuw}\leftrightarrow<\text{111}\> might be first order like the transition observed at \(T_{\text{tr}} = 14.5 \, \text{K}\) in DyIG [25, 26], good agreement is found with another second order transition at \(T_{c} = 135 \, \text{K}\) on Tb_{0.26} Y_{2/3} IG single crystal [27]. The monoclinic space group (C2/c) (\(\Gamma_{g} = A_{g}\)) [23] (table 2) is a subgroup of the two space groups I\(4_{/}\text{acd}\) and R \(\overline{3}\) c: it may be used in the intermediate T-range.

4.2.1. Tetragonal basis vectors of I\(4_{/}\text{acd}\) (\(\Gamma_{g} = A_{g}\)).
Using the above linear combinations \(F_{i}\) and \(G_{i}\), the tetragonal basis vectors for the two RE\(^{3+}\) inequivalent sites (8b) and (16e) are given on table 2. A ‘square pyramid’ umbrella structure is found in the subgroup I\(4_{/}\text{acd}\) (\(\Gamma_{g} = A_{g}\)): site (8b) \(C\) with \(S_{y} = S_{z} = + S_{x} = + S_{z} = (0,0,F)\) along [001]; site (16e): \(C\)\(_{1}\)' with \(S_{y} = + S_{z} = (0,a,b)\); \(S_{y} = + S_{z} = (0,-a,b)\) in the (100) plane; \(C\)\(_{1}\)'\(_{2}\) with \(S_{y} = + S_{z} = (a,0,b)\); \(S_{y} = + S_{z} = (-a,0,b)\) in the (010) plane with the general condition: \(F \neq (a^{2} + b^{2})^{1/2}\).

4.2.2. Monoclinic basis vectors of C2/c (\(\Gamma_{g} = A_{g}\)).
The magnetic structures which appear in the intermediate T-range are complicated. A \(T_{c} = 40 \, \text{K}\), the further splitting will depend on the choice of the monoclinic axis of the subgroup C2/c of I\(4_{/}\text{acd}\). We choose at first the case of ‘unique axis c, cell choice 3’ of the subgroup I\(112_{/}\text{b}\) [23] with the origin at \(\Gamma\), a cubic binary axis 2 = \((2\, x, 0,1/2,0)\) in 0,0,z and a glide plane \(b = (\Gamma.2\, x, 0,1/2,0)\) in \(x,y,0\). We obtain the corresponding sites: (8f) \(C_{1}\), \(C_{1}\)',; (8f) \(C_{2}\), \(C_{2}\)',; (4e) \(C_{1}\) and (4e)' \(C_{1}\)'. At \(T_{c} = 140 \, \text{K}\), we choose in this second case the following generators for the non-isomorphic subgroup R\(12_{/}\text{c}(\text{C2/c})\) of R \(\overline{3}\) c [23]: the identity (\(\Gamma\)), the inversion (\(\Gamma.2\, x, 0,1/2,0)\) in \(x,y,0\), a diagonal binary axis 2 = \((2\, x, y, z)\) in \(x,y,0\) and a glide plane (\(\Gamma.2\, x, y, z)\) in \(x,y,0\). We obtain the following additional splitting of the sites \(C_{1}\), \(C_{2}\), \(C_{1}\)', \(C_{2}\)'\(_{1}\), \(C_{2}\)'\(_{2}\): sites (8f) \(C_{1}\), \(C_{1}\)',; (8f) \(C_{2}\), \(C_{2}\)',; (4e) \(C_{1}\) and (4e)' \(C_{1}\)'\(_{2}\). The corresponding basis vectors are given on table 2. This preliminary result of the Bertaut’ representation analysis of C2/c provides a first but an incomplete answer in our research of the temperature evolution of magnetic structures in Tb\(_{0.37}\) Y\(_{2/3}\) IG.

4.3. Investigation of the ‘square pyramid’ umbrella for Tb\(_{0.26}\) Y\(_{2/3}\) IG.

4.3.1. The effective spin Hamiltonian model.
We suppose that the quasi doublet model with a pseudo spin \(S = 1/2\) seems to apply to the Tb\(^{3+}\) ion, suggesting that the next energy level is lying at least 80 cm\(^{-1}\) above [18]. The contribution of the RE\(^{3+}\) ions of the site \(\{i\}\) in the \(\{i = 1-6\}\) notation [13, 14] to the Zeeman and exchange energies is represented by the spin Hamiltonian: \(\hat{\mathbf{H}} = + \mu_{B} \mathbf{H} \mathbf{g} \mathbf{S} + (–\mathbf{M}_{w} / \mu_{B}) \mathbf{G} \mathbf{S} \). In the system of the local axis \((\mathbf{U}, \mathbf{V}, \mathbf{W})\) the anisotropic exchange \(\mathbf{G}\) tensor has the same axis as the magnetic tensor \(\mathbf{g}\). If the magnetic field vector \(\mathbf{H}\) is equal to zero, the components of the magnetic moment \(\mathbf{m}\) are reduced to: \(m_{z} = (\mu_{B}/2) \times n_{i} \mathbf{g}_{i} \mathbf{G}_{i} / (\sum n_{i} \mathbf{g}_{i} \mathbf{G}_{i})^{1/2}\) (by omitting the indice \(i\)). The anisotropy of these two tensors is assumed to be similar and diagonal and we shall consider in our investigation only six parameters \(g_{i}\) and \(G_{i}\) (\(i = x,y,z\)). The similar reduction (\(\sim 23 \%\)) obtained for our two terbium concentrations \((x = 3)\) and \((x = 0.37)\) imply that the new parameters of TbIG at 5 K along the [111] axis confirmed by the recent method of validation of magnetic structures [28] can be used. For the sublattice \(C_{1}\), in the notation \(C_{1}\)\(_{j}\) (\(j = 1-3\)) the moment vector \(\mathbf{m}_{j}\) with the value \((8.07 \, \mu_{B})\) is in the plane \((\mathbf{U}, \mathbf{W}) = ([100],[011])\) and originate an angle of 30.79 and 23.95\(^{\circ}\) with [111] and [100] respectively; in this case, we have: \(g_{i} \mathbf{G}_{i} / g_{j} \mathbf{G}_{j} = \tan(23.95^{\circ})/\sqrt{2} = 0.3134\). In the same manner for the sublattice \(C_{1}\)', the moment vector \(\mathbf{m}_{j}'\), \((8.99 \, \mu_{B})\) is in the plane \((\mathbf{U}, \mathbf{V}) = ([100],[011])\) and originate with [111] an angle of 28.07\(^{\circ}\) and with
[011] and [100] the angles of 7.19 and 82.81° respectively. We have then: \( g_x G / g_y G = \tan(82.81°) / \sqrt{2} = 5.605 \). We obtain a particular requirement from the following inequalities: \( g_x G / g_y G << g_y G \) with \( 1 < 3 \ll 18 \). For the sublattices \( C_1 \) (or \( C_5 \)), the moment vector \( m'_i \) (or \( m_i \)) which is in the same equivalent plane \((U, W) \equiv (U, V) = ([100],[011])\), the ratio of the components \((W/V)\) is proportional to \((g_x G / g_y G) = 0.056\) with \( \delta = \arctan(0.056) = 3.2° \), the angle with the local axis \( V = [011] \). In the tetragonal model, if we take into account the angle of \( 45° – 3.2° \) = 41.8° with [001] we can find the value 8.49 \( \mu_B \) for the moments of the sublattices \( C_1, C' \) and \( C_2, C'_2 \). The moment vectors \( m_i \) and \( m'_i \) of the sublattices \( C_i \) and \( C'_i \) are parallel to the [001] axis with the new specific condition for the low moment value: \( F < (a^2 + b^2)^{1/2} < 9 \mu_B \) (free ion value). We can assume that if \( g_x = 0 \), we obtain \( \Theta = +45° \) and the moment vector \( m'_i \) is aligned directly along the direction of the local axis \( V = [011] \). Finally at 4.2 K, the predicted parameters of our single ‘square pyramid’ umbrella (figure 6) with \( M_{x_3} \) along [00T] are defined by: site (8b), \( C'_y C' \); \( m_i = m'_i = 8.1\pm0.02 \mu_B \) and \( \Theta = 0° \); site (16e), \( C'_y C' \) and \( C_y C'_y \); \( m_i = m'_i = 8.97\pm0.02 \mu_B \) and \( \Theta = 45° \) respectively in the (100) and (010) planes.

4.3.2. Discussion.

Previous parameters obtained with crystal field calculations on \( \text{Tb}_{0.25} \text{Y}_{0.75} \text{IG} \) single crystal [29] give a calculated average moment \( <m>_{\text{calc}} = 7.66 \mu_B / \text{Tb}^{3+} \) close in agreement with their measured magnetization value \( <m>_{\text{exp}} = 7.69 \mu_B / \text{Tb}^{3+} \). But, two different conical arrangements are found: the sublattices \( C_y C'_y \) and \( C_y C'_y \) are decoupled and situated in two planes with respectively (3, 5) and (4,6) in the \{i = 1-6\} notation of ref. [14]. Their model is also based only the two anisotropy constants \( K_1 \) and \( K_2 \) where only first order transitions exist. At the opposite, our results are in good agreement with the magnetization measurements and the required conditions on the three anisotropy constants \( K_1, K_2 \) and \( K_3 \) > 0 in their relative form \( K_1 = K_1/K_2 \) and \( K_3 = K_1/K_2 \) found previously [18, 20]. NMR investigations are made also on the same compound \( \text{Tb}_{0.25} \text{Y}_{0.75} \text{IG} \) [30]. The value of \( <m>_{\text{exp}} = 6.62 \mu_B / \text{Tb}^{3+} \) leads to a poor agreement with \( <m>_{\text{calc}} \), and with the constraint of our model in which the four large moments build the ‘square pyramid’ umbrella while the two low moments are along [001].

5. Conclusion

The magnetic structures in \( \text{TbIG} \) and \( \text{Tb}_{0.25} \text{Y}_{0.75} \text{IG} \) have been revisited using the Bertaut’s representation analysis. Taking into account the observed smallness superstructure lines (200), (600,442), the refinement of the TbIG powder neutron diffraction data at 5 K give rises new parameters of the double umbrella in the subgroup \( R \overline{3} c \) along the easy axis [111]. A good agreement is found with the high field magnetization at 4.2 K where a third critical field \( H_{32} \) is observed and added to previous \( H_{14} \) and \( H_{12} \) along the hard axis [001]. For \( \text{Tb}_{0.25} \text{Y}_{0.75} \text{IG} \), a critical field \( H_{c} \) is observed at 4.2 and 40 K along the hard axis [111]. Parameters of a ‘square pyramid’ umbrella structure are predicted at 4.2 K in the tetragonal subgroup \( I4/acd \) where the easy axis belongs to the high symmetry phase \( (<001>) \) until \( T = 40 \) K. For \( T > T_c \) (140 K), the easy axis is in the high symmetry phase \( (<111>) \) and the magnetic structures can be described in the subgroup \( R \overline{3} c \). Between the two preceding second order spin reorientation phase transition temperatures where the angular and low symmetry phases \( <uuv> \) exist, an anisotropy of magnetization is observed below a specific temperature of anisotropy \( T\alpha \) (80 K). This result seems to be associated to a change of the spontaneous and the field-induced non collinear magnetic structures. Basis vectors of the subgroup \( C2/c \) are proposed for the intermediate \( T\alpha \) range. Such results look reasonable and are indeed a first approximation without neutron diffraction study on \( \text{Tb}_{0.25} \text{Y}_{0.75} \text{IG} \).
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