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DOUBLE UMBRELLA STRUCTURE IN TERBIUM IRON GARNET

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(TITRE) ET RESUME EN FRANCAIS :

Double structure parapluie dans les grenats de fer et de terbium

La structure magnétique en parapluie du grenat de fer et de terbium (T6IG) a été étudiée par diffraction de neutron de 4,2 K à 650 K, domaine qui couvre à la fois la température de Néel (T_N = 550 ± 10 K) et la température de compensation (T_{comp} = 244 ± 2 K).

Pour $T > T_N$, les pics observés ont seulement une contribution nucléaire. Pour $T_{comp} < T < T_N$, les résultats peuvent être interprétés selon la théorie de Néel du ferri : magnétisme. En dessous de T_{comp}, trois points importants peuvent être notés : des raies de surstructure apparaissent qui sont interdites par le groupe d'espace cubique Ia3d (ex : (110),(310) (411,330),(530,433)) alors que d'autres raies de surstructure sont absentes contrairement aux autres grenats de terre rare lourdes (ex : (200,(600,442),(622)) ; la réflection la plus forte (110) s'éteint près de T_{comp}. Les modes magnétiques associés au groupe d'espace de symétrie rhomboédrale $R\bar{3}c$ ont été calculés ; les 2 sites magnétiques non équivalents C_1 et C_1' de l'ion Tb^{3+} ont été décrits en choisissant comme axes x, y et z : $[\bar{2}11]$, $[0\bar{1}1]$ et [111]. A 4,2 K, les paramètres de la double structure en parapluie sont trouvés égaux à m_1 = 8,1 μ_B ; θ_1 = 30°79, ϕ_1 = 180° et m_1' = 8,9 μ_B, θ_1' = 28°07 et ϕ_1' = 0°. Ces paramètres conduisent à un parfait accord avec les mesures d'aimantation spontanée pour les 3 directions cristallographiques : [100], [110] et [111].

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ABSTRACT

The umbrella magnetic structure of the terbium iron garnet (TbIG) has been studied using neutron diffraction experiments performed in the 4.2 to 650 K temperature range which covers both the Néel temperature (T_N = 550 ± 10 K) and the compensation temperature (T_{comp} = 244± 2 K).

When $T > T_N$, the observed peak intensities originate from nuclear contribution only. When $T_{comp} < T < T_N$, the results have been interpreted in the Néel theory of ferrimagnetism. Below T_{comp}, three salient features have been observed : there appear superstructure lines forbiden by the cubic space group Ia3d, such as (110), (310), (411,330), (530,433) whereas the other superstructure peaks (200), (600,442) and (622) are found absent contrary to other heavy rare earth garnets ; the strongest reflexion (110) vanishes near T_{comp}. The magnetic modes associated to the rhombohedral symmetry space group $R\bar{3}c$ have been calculated ; the two inequivalent magnetic sites C_1 and C_1' of the Tb^{3+} ion have been described choosing the $[\bar{2}11]$, $[0\bar{1}1]$ and $[111]$ as x, y and z axes. At 4.2 K, the parameters of the "double" umbrella structure are found equal to $m_1 = 8.18\mu_B$; $\vartheta_1 = 30.79°$; $\varphi_1 = 180°$ and $m_1' = 8.90\mu_B$; $\theta_1' = 28.07°$ and $\phi_1' = 0°$. They lead to a perfect agreement with the spontaneous TbIG magnetization measurements for the three main crystallographic directions : [100], [110] and [111].

INTRODUCTION

The rare earth iron garnets $\{R_3^{3+}\}[Fe_2^{3+}](Fe_3^{3+})\ O_{12}$ (RIG) are known to present ferrimagnetic properties at room temperature. These properties are directly related to the distribution of the magnetic ions over three different crystallographic sites of the cubic space group Ia3d |1| : The R^{3+} ions are in the 24c (dodecahedral) while the Fe^{3+} ions occupy the 16 a (octahedral) and the 24d (tetrahedral) sites. The strongest a-d superexchange interactions constrain the sublattices magnetization M_a and M_d to be antiparallel ; they are independent of the nature of the rare earth so the Néel temperature is nearly the same for all iron garnets (T_N = 550 ±10K). The super-exchange interactions between the iron and the rare earth ions are an order of magnitude smaller than the first ones and lead M_c to be antiparallel to (M_d-M_a). The bulk garnet magnetization is thus given by $|(M_d-M_a) - M_c|$; it may vanish at some compensation temperature (T_{comp}) lower than T_N as found in the heavy rare earth iron garnets. In $Tb_3Fe_5O_{12}$, T_{comp} is equal to (244± 2K).

At low temperature, the classical ferrimagnetic model must be replaced by a new magnetic configuration in which the R^{3+} magnetic moments are no more antiparallel to the resultant Fe^{3+} magnetization. It results from the presence of two types of anisotropies. First, the R^{3+} free ion energy levels are strongly influenced by the crystalline field due to the ligands |2|; secondly owing to the orbital degeneracy of the R^{3+} ions, the superexchange interactions Fe^{3+} - R^{3+} is strongly anisotropic (detailed references can be found in |3|). Such a situation has been described phenomenologically by both $\underline{g}$ and $\underline{G}$ (anisotropic exchange) tensors using R^{3+} fictive spins.

In TbIG, previous studies using X rays and neutrons diffraction experiments powder samples at low temperatures showed a conical arrangement of the Tb^{3+} magnetic moment associated to a small lattice distortion : TbIG becomes rhombohedral, so at 6.75 K, the rhombohedral angle of the cubic distorted cell is α_r = 89°52'30" together with a slight cell contraction. The local symmetry of the R^{3+} ion passes from $D_2(222)$ to $C_2(2)$ |4||5| |6||7|.

As the a-d exchange coupling are too strong to be disturbed by the non collinear Tb^{3+} ordering ,the $[Fe_2^{3+}]$ and the (Fe_3^{3+}) magnetic moments lie along the [111] direction which remains to be the macroscopic easy axis of the garnet |8| . Several years ago, one of us |4||5| has proposed that the Tb^{3+} moments are aligned along the generatrixes of one cône : at 4.2 K, the cône angle with [111] is equal to 30 degrees , the moment value

being $8.5\mu_B$. This structure was based on the non-observation of the (200) super-structure line.

As the position sensitive detectors (P.S.D.) technique allow now to collect precisely neutron diffraction lines of low intensity, it was decided to confirm (or to deny) the absence of the (200) line and to study the thermal evolution of the other superstructure lines as recently performed in ErIG and HoIG [9] [10] .

EXPERIMENTAL

Neutron diffraction experiments on the powder sample TbIG have been performed at the "Siloé" reactor of the Nuclear Center of Grenoble using a P.S.D. technique. In the cryostat, ten values of temperature (4.2, 20, 54, 68, 80, 109, 131, 160, 208 and 244 K) were studied, while in the furnace T was equal to 283, 400, 453 and 614 K. A wavelength of 2.49 Å was used, the time of counting for each temperature was in the order of ten hours, and all the experiments were performed without magnetic field using filters to avoid $\lambda/2$ contamination.

The magnetization of a flux grown single crystal (the sample is a sphere of 5.5 mm of diameter) was measured in d-c magnetic field H up to 200 kOe from 4.2 to 300 K at the Service National des Champs Intenses (SNCI, Grenoble). H was applied parallel to [111], [100] and [110] directions successively.

RESULTS AND DISCUSSION

A detailed analysis of the magnetic properties will be given in a forthcoming paper. Only the results obtained at 4.2 K are presented (fig. 1) ; they show evidence of two types of properties : high anisotropy in this garnet and when H is parallel to the [100] direction, two abrupt transitions are induced by an internal field (external field minus demagnetizing field) equal to 66.6 and 82.8 kOe respectively . It is worth to note that these transitions which, to our knowledge were never mentioned before, reduce to one transition only when H is tilted of about ten degrees with respect to the [100] direction.

The spontaneous magnetizations which have been determined by extrapolating the M(H) curves to zero internal field are found equal to $34.53\mu_B.mole^{-1}$, $28.35\mu_B.mole^{-1}$, $20.17\mu_B.mole^{-1}$ for the [111], [011] and [100] directions respectively ; here one mole is defined as $2(Tb_3Fe_5O_{12})$. For the [111] direction, our determination agrees perfectly with previous results [11], [12] ; no informations concerning the two other directions were available in the litterature down to 4.2 K.

The neutron diffraction will be now discussed. Whatever the temperature is, all the observed reflexions (h k l) are indexed by $h + k + l = 2n$ which is the general extinction rule of the cubic space group Ia3d-(O_h^{10}) with eight $R_3Fe_5O_{12}$ formulas per cell. The lattice parameter a was deduced for each temperature from the values of the diffraction angles ; the results are in excellent agreement with those obtained by X-rays diffraction |6| , |7| , so at 283 K, a is found equal to (12.433 ±0.004) Å . When T (614 K) is higher than T_N, only the nuclear contribution exists. Attention was paid to the Lorentz correction, to the thermal agitation parameters ; thus the observed peak intensities fitted very well the absolute values calculated with the following oxygen atomic positions (x = - 0.0279 ; y = 0.0555 ; z = 0.1505) which have been previously determined at 673 K by one of us |5| . Such a fit has allowed the determination of the scale factor.

When $T_{comp} < T < T_N$, the observed spectra have been well interpreted within the ferrimagnetic Néel model which is well described by the magnetic modes F belonging to the three dimensional irreducible representation T_{1g} of the space group Ia3d with the wave vector $\vec{k} = [000]$, |5|. The calculation of this magnetic contribution was made with the following choices : the three magnetic form factors were issued from the reference |13|, the iron magnetic moments were assumed equal to the values obtained in YIG |14|, the Tb^{3+} sublattice magnetization is equal to the values measured either on powder samples |15| or on single crystals |12| . Working in these hypothesis leads to a good fit of the neutron intensities with a reliability factor defined as $R = \Sigma |(I_{obs} - I_{calc})| / \Sigma I_{obs}$ of about 10 % for the three values of T.

Below T_{comp}, there appear several superstructure lines which are forbidden by the garnet nuclear space group Ia3d ; they correspond to (h k l) equal to (110), (310), (411, 330), (530, 433) respectively. Three salient points are to be noted. First the (200), (600, 442) and (622) reflexions were never observed in spite of a long counting time (ten hours) and of a very good temperature stability (10^{-1} degree). So it can be concluded that such an absence is an intrinsec property of the TbIG garnet since the (200) peak has been evidenced in other garnets |5|, |16|. Secondly, the (222) reflexion was not observed which, contrary to the first point, is a common feature for all iron garnets. The third point is concerned with the thermal evolution of the (110) reflexion which is the strongest among all superstructures: as shown in the figure 2, it is clear to see that this line vanishes near T_{comp}, similar behaviours were observed in ErIG and HoIG |9| |10| . It should be noted that at 208 K, the (110) reflexion amounts to 0.6 % with respect the (211) line.

To discuss the magnetic structure at low temperatures, it is of great importance to make two remarks : the cubic magnetic modes of the T_{1g} representation do not lead to intensities which fit the experimental data; X rays diffraction having shown evidence of a rhombohedral distorsion the highest symmetry group of Ia3d to be considered in representation analysis is the subgroup $R\bar{3}c$ - (D^6_{3d}) |4| . The R^{3+} atomic positions in both groups are given in table 1. As the superstructure lines are also indexed with $k + k + l = 2n$, the wave vector $\vec{k}$ of the magnetic lattice is $\vec{k} = [000]$, (chemical cell = magnetic cell) ; thus the primitive translation of the crystallographic lattice (I), $(1|\vec{R}_n) = (1\ |1/2,1/2,1/2)$ is also a primitive translation of the magnetic lattice (I). Furthermore, the absence of the (222) superstructure reflexions implies that the inversion $(\bar{1}|000)$ becomes a magnetic translation : the spins of two atoms which are related by this inversion symmetry are parallel. Finally, the 24 R^{3+} ions are distributed over six magnetically inequivalent sublattices noted C_j and C'_j (j = 1,2,3) ; C_2 and C_3 are related respectively to C_1 by a rotation of 120 and 240 degrees about the 3-fold symmetry [111] axis (i.e. for C'_j). When the symmetry is reduced from Ia3d to $R\bar{3}c$, the preceeding magnetic inequivalency becomes a crystallographic inequivalency : the sublattices (C) corresponding to the sites (6e) (i.e. for the sublattices (C') to the sites (6e')).Using the macroscopic method of Bertaut |17|,|18| , the calculation of the rhombohedral magnetic modes have been performed for the different magnetic sites : R^{3+} (24c→ 6e and 6e'), Fe^{3+} (24d→ 12f) , Fe^{3+} (16a→ 2b and 6d). The representation analysis of $R\bar{3}c$ - (D^6_{3d}) , $\vec{k} = [000]$ space group will be published in a future paper. The irreductible representation T_{1g} is reduced to $A_{2g} + E_g$; the two sites R^{3+} basis vectors which belong to the one dimensional representation A_{2g} are given in the table 2 and thus without exception all magnetic components of the three types of magnetic ions may be coupled in an hamiltonien of order two.

For visualizing the rhombohedral symmetry $[\bar{2}11]$, $[0\bar{1}1]$ and [111] directions are choosen to be the x,y,z axes where $xz(\pi)$ is a g tensor plane. From the analysis of the R^{3+} basis vectors, it is concluded that four magnetic configurations are possible as indicated schematically for C_1 and C'_1 in the figure 3 : these two R^{3+} moments lie in the (π) plane while the Fe^{3+} spins are obviously aligned along the [111] direction. Due to the fact that in TbIG the (200), (600, 442) and (622) lines were not observed, the x components of the C_1 and C'_1 sites must be opposite and thus only two configurations (fig. 3c and 3d) have to be considered. Introducing the spherical coordinates (m_1, θ_1, ϕ_1) for C_1, C_2, C_3 and $(m'_1, \theta'_1, \phi'_1)$ for C'_1, C'_2,C'_3, the refinements of all

the twenty line intensities observed at 4.2 K was carried out for the two allowed configurations. It appears unambiguously that only the structure in which the C_1 moment lies between [111] and [100] whereas C_1' moment between [111] and [011] fits perfectly with a reliability factor R= 6 %. The parameters of the "double umbrella" structure were found to be : $m_1 = (8.18 \pm 0.02)\mu_B$; $\theta_1 = (30.79 \pm 0.02)°$; $\phi_1 = 180°$ and $m_1' = (8.90 \pm 0.02)\mu_B$; $\theta_1' = (28.07 \pm 0.05)°$; $\phi_1' = 0°$.

The ferrite spontaneous magnetizations along the main crystallographic directions [100] , [110] and [111] associated to this "double umbrella" structure are in very good agreement with those obtained from magnetic measurements ; not as an example, that for [111] the neutron analysis gives $34.64\mu_B.mole^{-1}$ which can be compared to $34.53\mu_B.mole^{-1}$ of the magnetic measurements.

In conclusion, a comparison of our "double umbrella" with the "single cône" structure proposed several years ago by one of us [5] is useful. At first, it should be noted that a complete agreement has been found between old and new observed spectra at 4.2 K. In a first approximation, the single cône structure was an experimental magnetic structure determination based on the magnetic modes of the cubic space group as a starting point. The authors did not take into account the detailed informations which can be deduced from the crystallographic distortion evidenced by F. Sayetat [6] [7] . In this work, it is clear that the magnetic modes associated to the rhombohedral space group $R\bar{3}c$ - (D_{3d}^6) allow to find two acceptable values of m_1 and m_1' which interpret perfectly the garnet magnetization. Furthermore, it is important to remark that in the whole temperature range (4.2 - 244 K), all the new observed spectra have been fitted within the rhombohedral model; the temperature dependence of the "double umbrella" structure will be discussed in a next paper. Here it is noted only that even the difference between the m_1 and m_1' moment values is rather small at 4.2 K, it becomes more important above 20 K.

REFERENCES

| 1| "Physics of Magnetic Garnets", edited by Paoletti, North-Holland Publishing Co (1978).

| 2| W.P. Wolf, M. Ball, M.T. Hutchings, M.J.M. Leask, A.F.G. Wyatt, J.Phys.Soc.Japan, 17 (1962) 443.

| 3| I. Veltrusky, V. Nekvasil, J.Phys.C., 13 (1980)1685.

| 4| F. Tcheou, E.F. Bertaut, A. Delapalme, F. Sayetat and H. Fuess, Int.Conf. on Rare Earth Elements, Colloq. CNRS N° 180, Vol. II, (1970) 313.

| 5| F. Tcheou, Thesis University of Grenoble (France), (1972), N° A.O. 7632.

| 6| F. Sayetat, J.Appl.Phys.Vol. 46, N°8 (1975) 3619.

| 7| F. Sayetat, Thesis University of Grenoble (France), (1974) N° A.O. 9677.

| 8| R.F. Pearson, J. Appl.Phys., Vol. 33, N°3 (1962) 1236.

| 9| M. Guillot, F. Tcheou, A. Marchand, P. Feldmann, C.R.Acad.Sci.Paris, 291, (1980) 227.

|10| M. Guillot, F. Tcheou, A. Marchand and P. Feldman, J.M.M.M., 31-34 (1983) 631.

|11| S. Geller, H.J. Williams, R.C. Sherwood, J.P. Remeika and G.P. Espinosa, Phys.Rev.131 (1963)1080.

|12| M. Guillot and H. Le Gall, J. de Phys. 38 (1977)871.

|13| R.E. Watson and A.J. Freeman, Acta Cryst. 14 (1961) 27.

|14| R.L. Gonano, Phys.Rev. 156 (1967) 521.

|15| R. Pauthenet, Ann.Phys. 3 (1958) 424.

|16| S.J. Pickart, H.A. Alperin and A.G. Clark, J.Appl.Phys. 41 (1970) 1192.

|17| E.F. Bertaut, Acta Cryst., A24 (1968) 217.

|18| E.F. Bertaut, J. de Phys. 32 (1971) C1-462.

$Ia3d - (O_h^{10})$			
	Sites	Symmetry	Positions
Tb^{3+}	24c	222	Site C : $\pm(\frac{1}{8},0,\frac{1}{4})$; + CP Site C' : $\pm(\frac{3}{8},0,\frac{3}{4})$; + CP
$R\bar{3}c - (D_{3d}^6)$			
Tb^{3+}	6e	2	$\pm(X,\frac{1}{2}-X,\frac{1}{4})$; $X=\frac{3}{8}$; + CP
	6e'	2	$\pm(X,\frac{1}{2}-X,\frac{1}{4})$; $X=\frac{7}{8}$; + CP

Table 1 - Atomic positions of the Tb^{3+} ions in the cubic space group $Ia3d - (O_h^{10})$ and in the rhombohedral space group $R\bar{3}c - (D_{3d}^6)$: C.P. represents the clock-wise permutations.

Sites (6e)	Sites (6e')
$\Psi_{11}(I) = (S_1+S_7)_X + (S_2+S_8)_Y + (S_3+S_9)_Z$	$\Psi_{11}(IV) = (S_4+S_{10})_X + (S_5+S_{11})_Y + (S_6+S_{12})_Z$
$\Psi_{11}(II) = (S_1+S_7)_Y + (S_1+S_7)_Z + (S_2+S_8)_Z + (S_2+S_8)_X + (S_3+S_9)_X + (S_3+S_9)_Y$	$\Psi_{11}(V) = (S_4+S_{10})_Y + (S_4+S_{10})_Z + (S_5+S_{11})_Z + (S_5+S_{11})_X + (S_6+S_{12})_X + (S_6+S_{12})_Y$

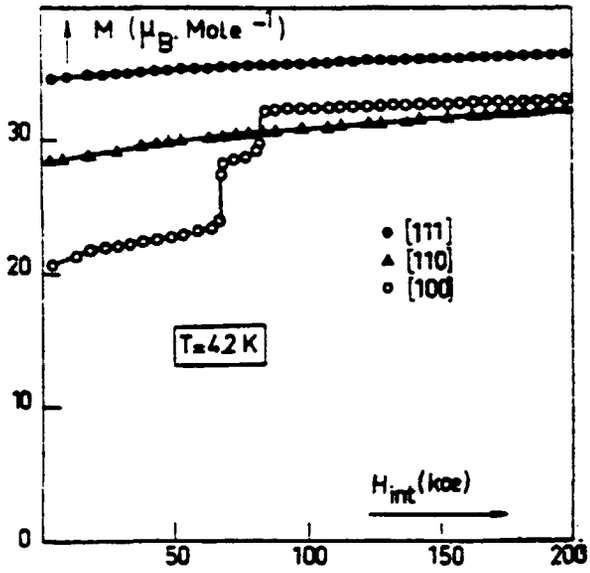
Table 2 - Magnetic modes (A_{2g} : $\vec{k} = [000]$)

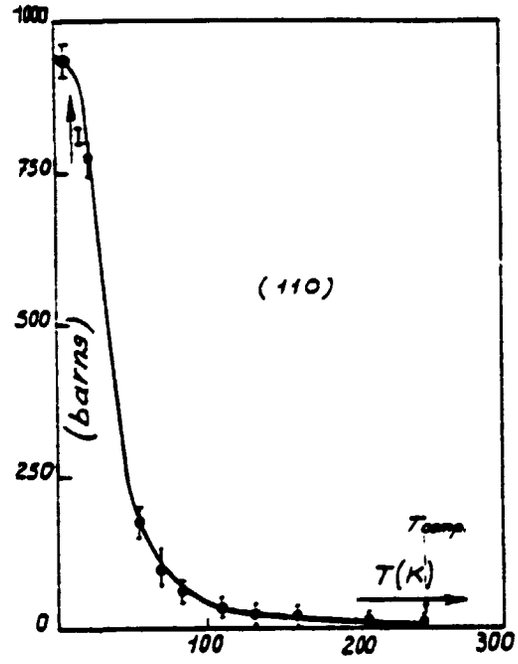
FIGURE CAPTIONS

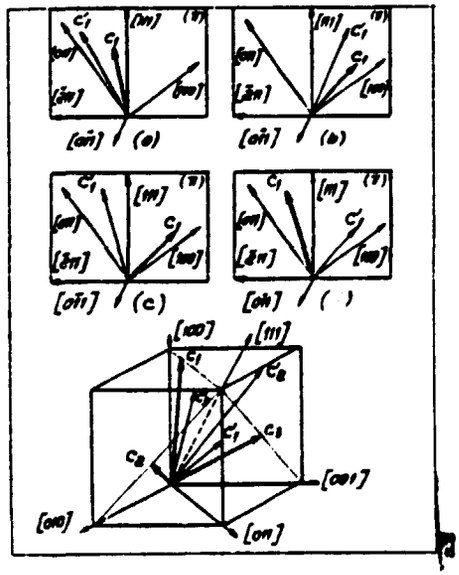
1 - Internal field dependence of the TbIG magnetization at 4.2 K for three field directions.

2 - The evolution of the (110) super-structure line due to the onset of the non collinear structure.

3 - Four magnetic configurations in the A_{2g} - $\vec{k} = [000]$ "Double umbrella" magnetic structure in TbIG


M (μ_B. Mole^{-1})
30
20
10
0
50
100
150
200
• [111]
▲ [110]
○ [100]
T=4.2 K
H_{int}(koe)


1000
750
500
250
0
(barns)
(110)
100
200
300
T(K)


(a)
(b)
(c)
[100]
[111]
[001]
[010]
[011]