Low temperature spin reorientation in dysprosium iron garnet

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Abstract. The spin reorientation (SR) phase transition in dysprosium iron garnet (Dy$_3$Fe$_5$O$_{12}$ or DyIG) have been studied by specific heat $C_p(T)$ and high field magnetisation measurements $M_f(H)$ and $M_h(T)$ on single crystals at low temperature. A first order SR is observed with a sharp jump at $T_{SR}=14.5\pm0.5$ K in the $C_p(T)$ curve which corresponds to a spontaneous change from the high temperature (HT) easy direction $<111>$ to an $<uuw>$ angular low temperature (LT) phases. Above $T_{SR}$, the magnetic structure is described by the irreducible representation (IR) $A_{2g}$ of the rhombohedral space group $R\overline{3}c$. Below $T_{SR}$, the magnetic structure changes in the monoclinic the space group C2/c with the IR $A_{g}$. When the field $H$ is kept aligned along the hard symmetry directions $<100>$ and $<110>$, we obtain respectively the variation of the angular positions $\theta(T)$ and $\theta'(T)$ from the total spontaneous magnetisation down to 1.5 K ($\theta=39.23^o$ and $\theta'=30.14^o$) and the results are in good agreement with the previous observations in low fields. When the sample is allowed to rotate freely on itself, the critical field $H_c(T)$ between the HT$<111>$ and the LT$<uuw>$ angular phases permits us to precise the transition line up to 15 T and 40 K between the so called canted field induced (FI) and the associated collinear magnetic phases. The experimental magnetic phase diagram (MPD) is precisely determined in the ($H_c-T$) plane and the domains of the existence and the stability of the two magnetic phases are specified.

Introduction
This work take place in the general magnetic study of the crystal field (CF) and the Fe$^{3+}$–RE$^{3+}$ anisotropic exchange (AE) interaction effects in rare earth iron garnets RE$_3$Fe$_5$O$_{12}$ (or REIG where RE$^{3+}$ is a rare earth ion or Y$^{3+}$) which are until now very attractive materials for their fundamental investigations [1]. The many interesting magnetic properties at low temperature which is often taken in account including strong magnetocrystalline anisotropy, spontaneous non-collinear magnetic structures (double umbrella-type), SR and FI magnetic phase transitions and their complicated experimental MPD in the vicinity of the compensation temperature, have been thoroughly reviewed by Guillot [2] and Zvezdin [3]. The occurrence of the so-called $<uuw>$ and $<uv0>$ angular LT phases where the iron sublattices magnetisation $M_{Fe}$ lies on a low symmetry direction are studied experimentally [4,5,6] and theoretically [7,8] by using different phenomenological models based on the generalized spin effective Hamiltonian broadly applicable to such magnetic transitions. So, the revisiting of these SR phase transitions would give a good chance to improve our knowledge in several topics especially detailed more information about the real non collinear magnetic structure of the RE$^{3+}$ at low temperature.
Experimental
The specific heat $C_p(T)$ has been performed on single crystal grown by standard PbO/PbF$_2$ method (disk of thickness: 1 mm; weight: 0.09074 g) without applied magnetic field and in the 1.25 K-40 K temperature range. Using the extraction technique in superconducting magnets, the magnetisations of two spherical single crystals of DyIG grown by the same method (weights: 0.37682 g and 0.20308 g and diameters respectively: 4.5 mm and 3 mm) have been measured in the 1.5 K-50 K temperature range and in high dc magnetic fields up to 16 T. The magnetic field was applied on the sample allowed to rotate freely on itself and along the three main crystallographic directions <111>, <110> and <100> which were checked by X-ray Laue technique within an error less than one degree. All the magnetisation results are reported in Bohr magneton by the 2(Dy$_3$Fe$_5$O$_{12}$) formula units and $H$ is the internal magnetic field. The magnetisations have been also performed on powder sample. By X-ray and neutron diffraction experiments at room temperature, the lattice constant of the powder sample (a=12.403 Å) was found in good agreement with previous values [9].

Results and discussion
The variation of the specific heat $C_p(T)$ versus $T$ is plotted in the figure 1. The results are in a good agreement with the data obtained on powder sample from 1.3 K to 16 K where the Schottky levels have been calculated [10]. At first, the SR transition is observed with a sharp jump at $T_{SR}=14.5\pm 0.5$ K in our $C_p(T)$ curve which correspond to a spontaneous change from the HT easy direction <111> to the <uuw> angular LT phases as reported and predicted previously [4-8]. At this first order transition temperature $T_{SR}$ a drastic change of the real structure is expected: the rhombohedral distortion of the R 3 c space group with only two inequivalent magnetic sites [9] is no longer valid below $T_{SR}$ and the symmetry is lowered with the appearance of at least four inequivalent sites of the monoclinic space group C2/c [6]. The more recent study of the Mössbauer spectra of DyIG [11] clearly confirm also that the SR phase transition occur at 15 K down 4.2 K but reveal in the 45 K-295 K temperature range, that the Mössbauer spectral components are consistent with a crystal with a reduced symmetry from cubic to rhombohedral but with the space group R 3. From the detailed of the magnetisations measurements in low and high magnetic fields and the neutron diffraction experiments, only two types of essential properties are presented in this discussion:

![Figure 1. Plot of the $C_p(T)$ vs $T$.](image1)

![Figure 2. Plots of $M_T(H)$ vs $H$ at 1.5 K.](image2)

1) Above 120 K and below $T_N=563$ K, we can describe the collinear ferrimagnetic order by the F modes of the irreducible representation (IR) $\Gamma_{4g}(T_{1g})$ belonging to $\text{Im} 3 d$ [12].
2) Below 120 K, the onset of the antiferromagnetic component of Dy$^{3+}$ moments, leads to a magnetic spin configuration of a double umbrella-type. So, the highest subgroup R 3 c of the cubic space group
is used in the representation analysis of Bertaut [13]. The opening of the double umbrella magnetic structure is followed by an increase of the rhombohedral distortion which modify the cubic description $\Gamma_4(T_1g) \rightarrow A_2g$ and the crystallographic and magnetic glide planes $c$ and $c'$ respectively are conserved. The change of the symmetry below $T_{SR}$ is a characteristic of a first order transition, and the basis vectors of the IR $A_g$ belonging to the monoclinic space group $C2/c$, are able to describe the non collinear magnetic structures in the LT<uuw> angular phases. The isothermal magnetisation curves $M_T(H)$ are reported as a function of the internal magnetic field at 1.5 K (Figure 2). It can be seen that the three main crystallographic directions $<111>, <110>$ and $<100>$ are non easy directions compared to the case of the free sphere where the magnetisation is always greater than when the field is applied along the main directions. The difference $\Delta M_S = M_S(\text{free}) - M_S(<111>)$ (where $M_S(\text{free})$ is the true spontaneous magnetisation and $M_S(<111>)$ is the forced one obtained with $H>H_c$) which equal zero for $T>T_{SR}$ is a constant reliable parameter for $T<T_{SR}$ $(0.52 \mu_B/2f.u)$ in the 10 K-1.5 K temperature range.

**Figure 3.** Plots of $\theta(T)$ ● and $\theta'(T)$ ▲ vs $T$.

**Figure 4.** Plots of $M_T(H)$ vs $H$ at 20.0 and 25.0 K.

When the field $H$ is kept aligned along the hard symmetry directions $<100>$ and $<110>$, we obtain respectively the thermal variations of the angular positions $\theta(T)$ and $\theta'(T)$ of the total magnetisation down to 1.5 K (see figure 3) where the results ($\theta=39.23^\circ$ and $\theta=30.14^\circ$) are in good agreement with previous observations in low fields [4,5]. It is shown, that the stable phases are of the <uuw> type below $T_{SR}$ and of the <111> type above. The rapid variation of the iron sublattices magnetisation direction $M_{Fe}$ in the vicinity of $T_{SR}$ confirms the character of a first order of the SR transition. The early studies focused on the FI phase transitions around the compensation or inversion temperature ($T_{comp}$ or $T_{I}=218.5$ K) of this compound [14]. The new observation of these FI phase transitions are obtained in the $T_{SR}$-50 K temperature range. The sample is aligned along <111> and is allowed to rotate freely on itself at 20.0 and 25.0 K (Figure 4). The transitions between HT<111> and LT<uuw> phases are observed on the curves $M_T(H)$ by a change in the susceptibilities at a critical field $H_c$. The magnetisation $M_{\text{ext}}(T)$ has been recorded at constant external magnetic field $H_{\text{ext}}=1, 2, 3, 4, 5, 8, 10.5$ and 13 T (Figure 5). The domains of the existence and the stability of the two magnetic phases are now specified and the transition line precised up to 15 T between the canted FI and the associated collinear magnetic phases experimental MPD in the ($H_c-T$) plane (Figure 6). The temperature of this transition between the HT<111> and the LT<uuw> phases increase linearly with the magnetic field and seems to disappears for $H>15$ T and $T>45$ K.
4. Conclusion

The SR phase transition in dysprosium iron garnet has been studied by specific heat and high field magnetisation measurements at low temperature. A sharp jump is observed at $T_{SR}=14.5\pm0.5$ K in the $C_p(T)$ curve which correspond to a spontaneous change from the HT<111> easy direction to an LT<uw> angular phases as predicted and previously reported. The first order transition <111>↔<uw> is explained by the transition IR $A_2$↔IR $A_g$ from R$3c$ to C2/c space groups. When the field $H$ is kept aligned along <100> and <110>, we obtain respectively the variation of the angular positions $\theta(T)$ and $\theta'(T)$ from the total spontaneous magnetisation down to 1.5 K ($\theta=39.23^\circ$ and $\theta'=30.14^\circ$) and the results are in good agreement with previous observations in low fields. When the sample is allowed to rotate freely on itself, the transition line $H_{c}(T)$ is precised up to 15 T and 40 K between the canted FI and the associated collinear magnetic phases. The domains of the existence and the stability of the two magnetic phases are specified in the experimental MPD $(H_c,T)$.

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