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Magnetization and specific heat studies of the low temperature anomalies in terbium iron garnet $Tb_3Fe_5O_{12}$

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Abstract. The macroscopic magnetic properties of the terbium iron garnet ($Tb_3Fe_5O_{12}$) single crystals were studied at low temperature by magnetizations $M_T(H)$ in magnetic fields up to 80 kOe and applied along the easy axis <111>. Maxima are observed near ~ 65 and ~ 47 K, respectively in *the curves of the* initial magnetic susceptibility $\chi_0(T)$, and the parameter $|b|(T)$ of the bH^2 term are associated with the non linear variation of the $M_T(H)$ magnetizations. From the specific heat $C_p(T)$ measurements, a great peak is observed near 57 K in the excess contribution of the Tb^{3+} ions $C_p^{ex}(T)$ to $C_p(T)$ which vanishes in the 170–208 K temperature range. The results are discussed with respect to the simultaneous signs of the "low-temperature point" T_B predicted by Belov at 58 K and the 'Schottky anomaly' detected experimentally by the presence in the $C_p^{ex}(T)$ curve of a $1/T^2$ term for $T > T_B$.

1. Introduction

The rare earth iron garnets $RE_3Fe_5O_{12}$ or REIGs (where RE^{3+} is a trivalent rare earth ion or the yttrium) or their mixed compounds are distinguished today by new fundamental interests [1], [2]. This is the case of $Tb_3Fe_5O_{12}$ or TbIG who presents large magnetodielectric effects in low external magnetic fields (H_{ex} < 0.2 T) at 2 K [3]. Furthermore, a significant coupling between the magnetic exchange and ligand-field excitations near 60–80 K with two distinct behaviours above and below 150 K were also reported [4]. It was shown recently [5], [6] that despite the <111> direction is the easy axis (EA) of magnetization at all temperatures, the magnetic structure changes from collinear to a 'double-umbrella' structure below 160 K. This change, well interpreted on basis of group theory analysis [7], is followed by an anomalous magnetic behaviour around 54 K. In this paper, one expects that this low-temperature anomalous behavior to be reflected with anomalies on some pertinent parameters, such as magnetic susceptibility, which may lead to the proximity of the so-called 'low-temperature point' of Belov T_B predicted at 58 K for TbIG [8 and refs. therein]. But we wish to focus on another important effect arising from the 'Schottky effect' which is able to give rise to anomaly in the magnetic specific heat contribution due to Tb^{3+} ions only in the in the region of T_B.

2. Experimental details and results

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The as-prepared TbIG samples are two spherical single crystals grown by the standard flux method. The first one (sample 1) (diameter: 5 mm; weight: 427.00 mg) is the same that used recently for the magnetization measurements $M_T(H)$ in high DC magnetic fields H_{ex} up to 200 kOe [9]. Whereas the second one (sample 2), it consists of a small sphere (diameter: 3 mm; weight 197.88 mg). Using a conventional magnetometer at the Institute Néel CNRS Grenoble, France, the $M_T(H)$ curves were measured from 4.2 K to 300 K with external magnetic field H_{ex} up to 80 kOe and applied along the EA <111>. The curves $M_T(H)$ versus the internal magnetic field H are overall reported in $\mu_B/2Tb_3Fe_5O_{12}$. The compensation temperature T_{comp} at which M^{total}(TbIG) vanishes is equal to 243.5 K [9] and 249 K for samples 1 and 2, respectively. The specific heat $C_p(T)$ measurements were performed on both samples using an AC calorimeter based on the compensation of power method from 5 K to 300 K. The measuring principle is based on the differential $C_p(T)$ between the sample and a reference. The increase of temperature (T) can be made according to variable speeds. Several runs with a rising of T with a speed of 1 K/mn were applied and a run was made with a more precision between 4.2 K and 100 K at a slower speed of 0.5 K/mn. Herein, the two best reproducible runs labeled as "data 1" and "data 2" in J/K mol (one mole = 2TbIG) for samples 1 and 2, respectively, are reported.

T*ypical* $M_T(H)$ curves obtained for the sample 1 at different values of T are given in Fig. 1. At 4.2 K, the sample is apparently saturated at 3.5 kOe, whereas the saturation is not attained for $T > 4.2$ K.

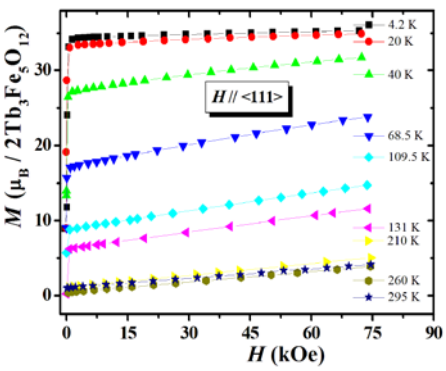

Figure 1. $M_T(H)$ vs H in the 4.2–300 K range.

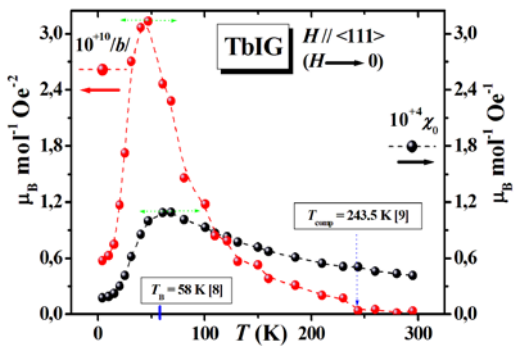

Figure 2. $10^{+4}\chi_0(T)$ and $10^{+10}|b|(T)$ vs T.

A non linear variation is shown with the exception of the T-region (220–260 K) below and above T_{comp} where the expected canted phases occur [9]. Both collinear and coaxial phases can be described using the previous expansion $M_T(H) = M_S(T) + \chi_0(T)H + b(T)H^2$ [9]. $M_S(T)$ identified with the spontaneous magnetization is equal to 34.35 μ_B/mol at 4.2 K. The initial magnetic susceptibility $\chi_0(T)$ is obtained by extrapolation at $H = 0$, and reaches the lowest value of 1.74×10^{-2} μ_B mol^{-1} kOe^{-1} at 4.2 K. Take into account the slight curvature of the curves, the parameter $b(T)$ is obtained. It is well-known that super-exchange interaction of Fe^{3+} in octahedral [a] and Fe^{3+} in tetrahedral (d) sites are almost identical for all REIGs [8]. Thus, we can determine the *spontaneous magnetization* M_{Tb} of the Tb^{3+} ions in dodecahedral {24c} site as $M_{Tb} \approx M^{total}$(TbIG) – M(YIG) in the basis of the two-sublattice model with the resultant of iron sublattices magnetization $M_{Fe} \approx M$(YIG). *At 4.2 K, the existence of a great difference in* M_{Tb} *between that (44.35* μ_B mol^{-1}*) of the coaxial structure and the calculated value in the hypothesis of both the collinear structure and the free ion value (54* μ_B mol^{-1}*) clearly reflects the effect of the* "double-umbrella" structure *of the* Tb^{3+} *moments, which is in agreement with the NPD results [5-7]. The plots of* $10^{+4}\chi_0(T)$ and the absolute values of b parameter $10^{+10}|b|(T)$ *are shown in Fig. 2.* Both curves increase when T decreases and show a maximum near ~ 65 K and ~ 47 K, respectively. *The measured* C_p(T) *data 1 and 2 plotted in the inset of Fig. 3 show a good agreement with those reported previously on powder sample for* T *< 25 K [10]. One can see a qualitative agreement from 20 K to 200 K with a few discrepancy of about 6% in the* T*-range 200–298.15 K, as well as for the data 2, with the results obtained on powder sample prepared by the standard high-*T *solid-state*

reaction method [11]. Above 25 K, the C_p(T) *data 1 are in good agreement with those measured on a sample prepared by a sol-gel powder method [12].*

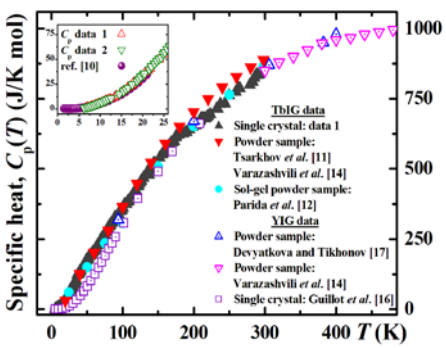

Figure 3. Plots of C_p data 1 with those found in the literature for TbIG and YIG. In inset, C_p data 1 and 2 are compared those in [10].

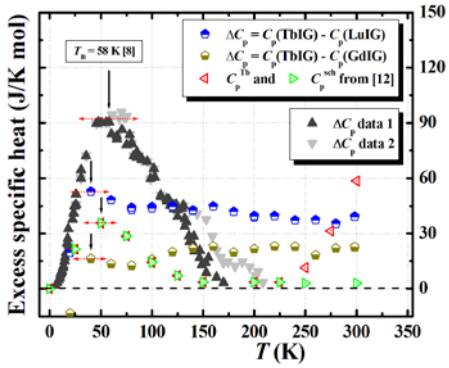

Figure 4. Plots of ΔC_p data 1 and 2 compared with other determinations of ΔC_p and with the calculated C_p^{Tb} and C_p^{sch} contributions [12].

3. Phenomenological analysis of the specific heat and discussion

As for all REIGs, C_p^{total}(TbIG) can be written simply as follows

$$C_p^{total}(\text{TbIG}) = C_p^{nuc} + C_p^{reg} + C_p^{mag} + C_p^{sch} \quad (1)$$

where C_p^{nuc} arises from the interactions between nuclear and electron spins, C_p^{reg} named as "regular part" in [13], [14] represents both lattice and dilatation contributions, C_p^{magn} is the cooperative part associated with ferrimagnetic disordering process, C_p^{sch} is the magnetic non-cooperative electronic Schottky contribution caused by redistribution of 4f-electrons in the Tb^{3+} ions over the excited levels during the increasing of T. Define $C_p^{M_{Fe}}$ and C_p^{Tb} are related to the contributions of M_{Fe} and M_{Tb} sublattice magnetization, and then, the magnetic contribution C_p^{magn} resulting from the competition between three types of anisotropies (spin-orbit coupling, crystalline field and super-exchange interactions) is simply described as $C_p^{magn} = C_p^{M_{Fe}} + C_p^{Tb}$. Then, C_p^{nuc} can be neglected above 6 K for TbIG [10], C_p^{total}(TbIG) can be expressed as follows

$$C_p^{total}(\text{TbIG}) = C_p^{reg} + C_p^{M_{Fe}} + C_p^{Tb} + C_p^{sch} \quad (2)$$

So, on the basis of the Debye model, the evaluation of C_p^{reg} cannot be simply obtained [15]. We assume that TbIG has the same value of C_p^{reg} with that of YIG. Besides this, the resultant M_{Fe} is saturated below T_{comp} for TbIG [9]. Meanwhile, M_{Fe} can not affect C_p^{total}(TbIG). Therefore, we can put $C_p^{reg} + C_p^{M_{Fe}} \approx C_p$(YIG) in equation (2). The expected excess magnetic contribution C_p^{ex} can be written as follows

$$C_p^{ex} = \Delta C_p \approx C_p^{total}(\text{TbIG}) - C_p(\text{YIG}) = C_p^{Tb} + C_p^{sch} \quad (3)$$

It must be noted that, in C_p(YIG), neither the 'low-T' of Belov T_B nor 'Schottky effect' are expected. The reason is that the ground state of the Y^{3+} ion is in a S-state, but it is not the case for the magnetic Tb^{3+} ion. In Fig 3, we have plotted the C_p(YIG) data measured on single crystal from 5.3 K to 208.6 K [16] with those reported for YIG powder samples [14], [17] in order to smooth more data in the large T-range. It is noted that the Debye temperature θ_D and lattice parameters vary negligibly through the REIGs [18]. Since θ_D is proportional to $(\bar{m})^{-1/2}$, where $\bar{m}$ is an average mass of the atoms in the unit cell, and θ_D(TbIG) is estimated to be larger than θ_D(YIG) by about 15% if $\bar{m}$ is assumed proportional to the molecular weight. A large excess in the C_p^{ex} curve is observed in Fig. 4 with a broad peak reaching ~ 93 J/K mol nearby 57 K for both data 1 and 2. After a rapid decreasing with an increasing of T, C_p^{ex} disappears near 170 and 208 K, respectively. After subtracting those of LuIG which was considered as an isostructural baseline [13], [14], the C_p^{ex} data deduced by the C_p^{total}(TbIG) data are also

shown in Fig. 4, where a maximum (about ~ 53 J/K mol) is observed at near 45 K. The differences between maxima and low-*T*-anomalies possibly arise from the large difference between atomic mass of Y and Lu elements. Then, one can estimate that θ_D(YIG) should be 15% larger than θ_D(LuIG). Moreover, due to the 'low-*T* point' at T_B = 58 K [8] and the 'Schottky effect' [15], anomalies are overlapped and appear as a continuum-type over the entire C_p^{ex} curve, and the transition at T_B is then obscured. For separating both excess from each other in the C_p^{ex} curve, C_p^{sch} can be tentatively appraised experimentally using C_p(GdIG) as a baseline [14]. Herein, C_p^{RE} and $C_p^{M_{Fe}}$ in these two REIGs having similar magnetization functions are roughly comparable. In addition, as Gd^{3+} is in ${}_8S^{7/2}$ ground state, Schottky contribution is not expected too, and the C_p^{sch} contribution can be assumed as $C_p^{sch} \approx C_p^{total}$(TbIG) – C_p(GdIG). A small 'knee' of 17 J/K mol appears at 45 K in the C_p^{sch} curve plotted in Fig. 4 with both calculated curves of C_p^{Tb} and C_p^{sch} contributions [12]. The calculation of C_p^{sch} using the generalized equation of Moretti and Ottonello [15] was carried out. Here, the magnetic splitting energies $\overline{E}_1$ = 37.04 cm^{-1} and $\overline{E}_{1'}$ = 109.34 cm^{-1} associated with the two inequivalent sublattices C_1 and C'_1, respectively, were used [5]-[7]. Only one maximum of ~36 J/K mol is observed at 50 K in both curves with about 10 J/K mol subsisting at 150 K. C_p^{Tb} curve appears again at T > 210 K and reaches ~ 59 J/K mol at 300 K. The discrepancies between amplitudes of the maxima could be due to the inconsistency of the values of $\overline{E}_1$ and $\overline{E}_{1'}$ compared with those proposed before in low-*T* 36 and 50 cm^{-1}, respectively [10]. The differences above 150 K may be due to the effect of C_p^{reg} which is further fairly inaccurate in this high-*T* region, and the dilatation of the lattice contribution becomes not negligible [18].

4. Conclusion

In this study, magnetization, magnetic susceptibility along <111> and specific heat measurements, anomalies are reported in TbIG single crystals at temperatures in the neighbourhood (± 10 K) of the "low-*T* point", which are due to the simultaneous effect of the 'Schottky anomaly' near 57 K on the excess specific heat. These results imply that precise calculations of the magnetic contribution based on the NPD refined parameters of the two Tb^{3+} sublattices with a best determination of the Schottky effect based on more reliable magnetic energy levels are needed.

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