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Specific heat of ytterbium iron garnet $Yb_3Fe_5O_{12}$ at low temperatures

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Abstract

The state of the Yb^{3+} ion of the ytterbium iron garnet ($Yb_3Fe_5O_{12}$) is suitable for the development of theoretical models with rather good approximations. Using the theory of two-level paramagnetic ions, the total and magnetic specific heat at low temperatures has been calculated. The influence of tensors **g** and **G** on the specific heat of YbIG has been examined. A comparison has been made of our results and those obtained with a model based on of the approximation of Weiss Molecular-Field (WMF) and experimental data.

Key Words: Ytterbium iron garnet YbIG, specific heat

1. Introduction

Rare-earth iron garnets (RIGs) are among some of the important magnetic materials and have extensive applications in materials science and technology. They are ferrimagnets, and are used in magnetic recording device and show giant magneto restriction at low temperatures. Similarly, the garnets have important applications in laser industry. By substituting various rare earth ions into the garnet lattice one can study the effect of these ions on the macroscopic properties. Within rare earth iron garnets a variety of magnetic interactions, which provide a detailed test for any proposed theoretical model [1].

The ytterbium iron garnet ($Yb_3Fe_5O_{12}$) is of particular interest because (i) the lowest doublet of Yb^{3+} in YbIG is separated from the excited states by an energy on the order of 600 cm^{-1} and thus does not contribute to the specific heat below 4 K; (ii) the interesting parts of the diagrams are confined in the low temperature range 2–30 K, mainly due to its very low inversion (compensation) temperature T_I ($\approx$7–8 K); and (iii), the state of the Yb^{3+} ion is suitable for the development of theoretical models with rather good approximations and generally this compound can be considered as a good example for the study of different kinds of magnetic phase [2–4].

In this paper, we are interested in the calculation of the specific heat of YbIG in the low temperatures

range 2–40 K. The influence of the values of the **g** and **G** tensors on the specific heat of the compound in question has been examined.

2. Theory of specific heat for YbIG

2.1. Two-level paramagnetic ions model for YbIG

The repartition of atoms of YbIG is as follow: the Fe^{3+}, located on a and d sites, are strongly internally coupled $(500K)$ and, at low temperature, effectively form a single rigid saturated sub lattice. The state of any give Fe^{3+} is that of the sublattice as a whole, the back reaction of neighboring Yb^{3+} ions being negligible. This justifies a Mean-field approach to the Yb-Fe interactions. The Yb ions, on six inequivalent c sites, interact negligibly among themselves and form a non-cooperative "paramagnetic" system. The lowest Kramer's doublet of Yb^{3+} is well separated from other states and with subtle corrections its spin Hamiltonian describes the behavior. Interactions within the ground doublet are given by exchange **G** and paramagnetic tensors **g**. The anisotropy of YbIG has its origin in these interactions.

Per the argument above, our calculation will be a straightforward application of the theory of two-level paramagnetic ions in a exchange field without an external magnetic field.

With consideration of the crystal field, exchange interaction and the external magnetic field, the Hamiltonian of the Yb^{3+} is written in the form [5, 6]

$$H = H_c + H_{exch} + H_{ext}. \tag{1}$$

The strongest values of crystal field product the split of energies, and the system may be reduced to the fundamental level which can be simulated as a spin fictive $S = \frac{1}{2}$ without a crystal field term. So, a Kramer's doublet in presence of a magnetic field and exchange interaction is

$$H_{ext} = -\mu_B \vec{S}\mathbf{g}\vec{H} \tag{2}$$

$$H_{exch} = -\vec{S}\mathbf{G}\vec{u} \tag{3}$$

Here, $\vec{u} = \frac{\vec{M}_{Fe}}{M_{Fe}}$ is a unit vector and **g** and **G** are diagonal matrices in local coordinates which satisfy the relation

$$\mathbf{G} = \mu_B M_{Fe}\mathbf{gn} \tag{4}$$

where **n** are the coefficients of molecular field and reduces to a constant in the isotropic case. The behavior of the ground doublet of Yb^{3+} on {q} site is given by the relation

$$(H)_q = (H_{exch} + H_{ext})_q = -\vec{S}_q\vec{\Delta}_q \tag{5}$$

where

$$\vec{\Delta}_q = -\mu_B \mathbf{g}_q\vec{H} + \mathbf{G}_q\vec{u} \tag{6}$$

is the level splitting of a site on the i^{th} Yb sublattice ($i = 1, 6$). Following this expression, all thermodynamic quantities are easily deduced. Using an approach based on a two-level system, the partition function for a {q} site can be written

$$Z_q = Tr\left[e^{-\beta(H)_q}\right], \beta = \frac{1}{k_B T}. \tag{7}$$

For a Kramer's doublet of Yb^{3+} on a {q} site, we have

$$Z_q = e^{\frac{\Delta_q}{2k_BT}} + e^{-\frac{\Delta_q}{2k_BT}}. \tag{8}$$

Then, the free magnetic energy for the ion of Ytterbium is

$$F(T)_q = -k_BTln(Z_q) = -k_BTln\left(2\cosh\left[\frac{\Delta_q}{2k_BT}\right]\right). \tag{9}$$

The total partition function for a six in equivalents sites is

$$Z = \prod_{q=1}^{6} Z_q \tag{10}$$

Thus, the total free magnetic energy of Yb^{3+} and Fe^{3+} ions in the presence of an external magnetic field is

$$F(T) = -k_BT\sum_{q=1}^{6}\left(ln\left(2\cosh\left[\frac{\Delta_q}{2k_BT}\right]\right)\right) - \vec{M}_{Fe}\vec{H} \tag{11}$$

From equations (6) and (11) (i) the effective fields at each of the six in equivalent Yb sites were determined via the **G** and **g** tensors and then the Yb-ion free energies were calculated; and (ii) the principal local axes of the **g** and **G** tensors are appropriate to the i^{th} sublattice.

2.2. Lattice specific heat: theoretical survey

For magnetic insulators such as the garnets the Hamiltonian of the system can be written in first approximation by [7]

$$H = H_{latt} + H_{magn} + H_{nucl}, \tag{12}$$

where H_{latt} depends only on the nuclear position and momentum coordinates, H_{magn} describes the behavior of the unpaired (magnetic) electrons, and H_{nucl} describes the interaction of the nuclear spins with the electron spins. From the equation of the Hamiltonian, the total specific heat resulting is given by

$$C = C_{latt} + C_{magn} + C_{nucl}, \tag{13}$$

where C_{latt}, C_{latt} and C_{nucl} are the lattice, the magnetic, and the nuclear specific heat, respectively. The problem of this equation is that the C_{latt} term cannot be determined directly. This limitation can be overcome by assuming the C_{latt} of rare earth garnets could be taken from that of LuIG. Hence, we replace the lattice term C_{latt} with the experimental values of LuIG, that is

$$C_{YbIG} = C(LuIG) + C_{magn} + C_{nucl} \tag{14}$$

The nuclear term can be chosen as

$$C_{nucl} = \frac{0.0158}{T^2}. \tag{15}$$

The magnetic term can be deduced from the theory of two-level paramagnetic ions (equations (1)), by using the well-known thermodynamic relation

$$C_{magn} = -T^2 \frac{d^2}{dT^2} F(T). \tag{16}$$

The application of the approximation of Weiss-Molecular- Field (WMF) gives

$$C_{magn_{WMF}} = 6R \left(\frac{\bar{E}_1}{k_B T}\right)^2 \frac{e^{-\frac{\bar{E}_1}{k_B T}}}{\left(1+\frac{\bar{E}_1}{k_B T}\right)^2}, \text{with } \overline{E}_1 = 25cm^{-1}. \tag{17}$$

In what follows, the total and the magnetic computed specific heat, without an external magnetic field, and for different choice of the values of tensors **g** and **G**, are shown. A comparison is made of our results with those obtained by the WMF approximation.

3. Numerical simulation

The total and magnetic heat specific computed of YbIG for different values of **g** and **G** was obtained by theory of two-level paramagnetic ions in an exchange field. The physical quantities used in the computation are listed in Table 1.

Table 1. Principal values of tensors **G** and **g** used in YbIG calculations on the local axis.

Author	G (cm^{-1})	g
Wickersheim[8]	11.625.729.9	2.853.603.78
Kolmakova[9]	25.52912	3.73.33.0
Fillion[10]	31.1527.0612.25	3.753.653.1

The results of our computed specific heat are presented and discuss below. They are plotted and compared with those obtained by Harris et al. [7].

In Figure 1 is plotted the total specific heat of YbIG by using the model of two-level paramagnetic ions for different choice of values of **g** and **G**, together with data from the theory of WMF approximation, in the temperature range 1.5–22 K. Results obtained by us is also depicted in the same figure for comparison. In the region above 10 K, we see that (i) the experimental values are lower than theoretical values; (ii) the specific heat calculated from the values of [9–10] are very close to the data of [7]; and (iii) the two-level model using values from Kolmakova [9] is in better agreement with the experimental results than the WMF approximation. In the region below 10 K, the WMF approximation is closer to the curve of specific heat of [8] than the model of two-level paramagnetic ions.

In Figure 2 is show the zoom of the specific heat of YbIG at very low temperatures. Observe that, below 4 K, the specific heat becomes exceeds what one would expect on the basis of the WMF and the two-level models.

Figure 3 shows the magnetic specific heat of YbIG computed with the two-level model and the WMF approximation. The figure compares the experimental magnetic specific heat data extracted from the data of [8]. The two-sites energy levels have been determined and compared with those obtained by the model of

WMF and the experimental data. Our values, E_1 and E_2, can be compared with those deduced from other experiments (see Table 2).

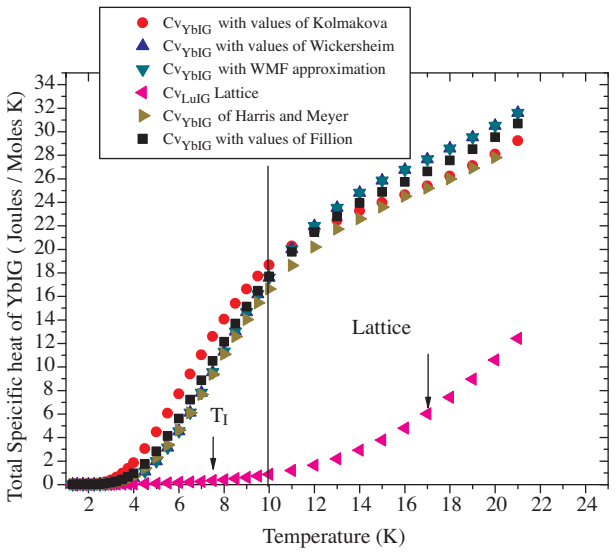

Figure 1. Total specific heat of YbIG in the range 1.5–2.2 K.

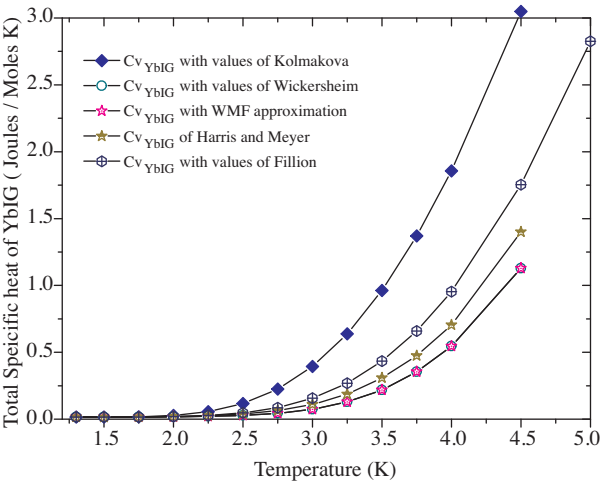

Figure 2. Total specific heat of YbIG in the temperature range 15–4.5 K.

In Table 2, we can see that the values of $\bar{E}$ predicted with our calculation based on two-level paramagnetic ions are much approached with the value extracted with the Weiss-Mean-Field (WMF) model (see Figure 4).

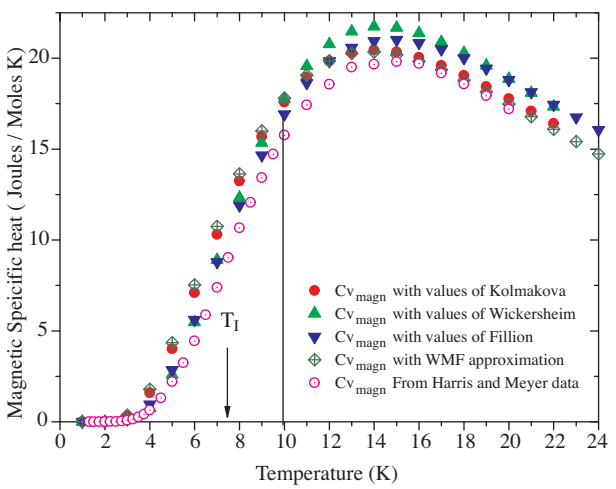

Figure 3. The magnetic specific heat of YbIG in the temperature range 1.5–22 K.

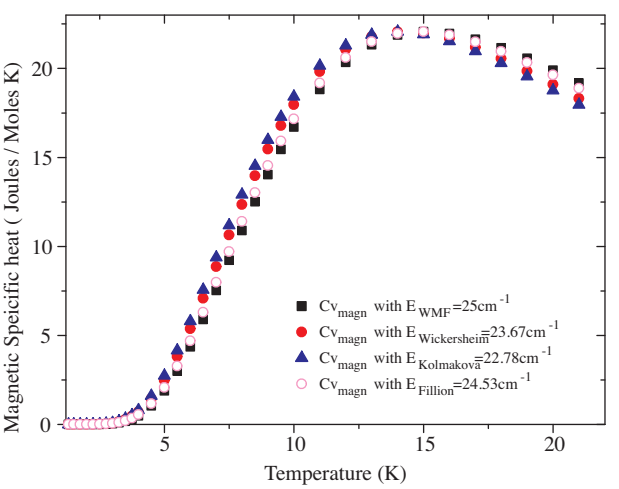

Figure 4. The computed magnetic specific heat of YbIG versus temperature, using the Weiss Mean Field approximation (equation 17).

Table 2. Values of splitting of the lowest doublet of Yb^{+3} in YbIG.

Source	$E_1\,(cm^{-1})$	$E_2\,(cm^{-1})$	$\bar{E}\,(cm^{-1})$
Specific heat (4–20 K)	...	...	25.0
Magnetization (2.2–100 K)	...	...	25.5
Infrared absorption (1.5 K)	23.4	26.4	24.9
Optical absorption (77 K)	22.1	25.3	23.7
Susceptibility (550–1450 K)	...	...	108
Our work			
•Wickersheim[8]	20.58	28.49	24.53
•Kolmakova[9]	17.67	27.88	22.78
•Fillion[10]	22.03	25.32	23.67

4. Conclusions

This paper examined the influence of tensors **g** and **G** on the calculated specific heat of YbIG. These parameters must be accurate because any deviation in the values of **g** and **G** would not give and reproduce the compensation temperature T_I of YbIG. In the region above 10 K the experimental curve lies below the theoretical one, possibly as a result of the uncertainty in the lattice specific heat; but in the region below 4 K the specific heat becomes larger than what one would expect on the basis of the WMF and tom-level paramagnetic models. The different choice of the values of **g** and **G** can affect the results of the total specific heat of YbIG. Finally, from Figure 1, we can see that the T_I temperature describes the reorientation of spin, but not a transition phase.

Appendix

The values of **g** and **G** are estimated in the local axis, but the measure of the specific heat in the laboratory is made in the global axis. For this, we used the transformation matrix (MT) from the global axis to local axis. The matrixes are

$$MT\,(C_1) = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} \\ 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{pmatrix}, MT\,(C_1') = \begin{pmatrix} 1 & 0 & 0 \\ 0 & \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \\ 0 & -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} \end{pmatrix}, MT\,(C_2) = \begin{pmatrix} 0 & 1 & 0 \\ -\frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \end{pmatrix}$$

$$MT\,(C_2') = \begin{pmatrix} 0 & 1 & 0 \\ \frac{1}{\sqrt{2}} & 0 & \frac{1}{\sqrt{2}} \\ \frac{1}{\sqrt{2}} & 0 & -\frac{1}{\sqrt{2}} \end{pmatrix}, MT\,(C_3) = \begin{pmatrix} 0 & 0 & 1 \\ \frac{1}{\sqrt{2}} & -\frac{1}{\sqrt{2}} & 0 \\ \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \end{pmatrix}, MT\,(C_3') = \begin{pmatrix} 0 & 0 & 1 \\ \frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \\ -\frac{1}{\sqrt{2}} & \frac{1}{\sqrt{2}} & 0 \end{pmatrix},$$

where C_i and C_i', with $i = (1, 2, 3)$, are the local six sublattice of Yb^{3+}.

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