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# High dc fields magnetisation of YbIG at low temperatures

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## Abstract

Magnetisation measurements have been performed on a single crystal of ytterbium iron garnet ( $\text{Yb}_3\text{Fe}_5\text{O}_{12}$  or YbIG) with dc magnetic fields up to 16 T applied along the  $\langle 110 \rangle$  direction at temperatures up to 50 K. The magnetic phase transitions, between the different so called canted and collinear phases, have been observed. The position of the lines of the phase diagram together with the nature of the corresponding transitions have been precisely determined. In particular the line separating the two canted phases is found to be of first order with a characteristic jump in the magnetisation which tend to vanish only at the highest fields. These results are compared with previous studies in higher pulsed fields and recent ac susceptibility measurements in fields lower than 4 T, together with the theoretical predictions. © 1998 Elsevier Science S.A.

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## 1. Introduction

The rare earth iron garnets exhibit several magnetic phase transitions induced by an external magnetic field leading to rather complicated phase diagrams depending on the field direction. The ytterbium iron garnet ( $\text{Yb}_3\text{Fe}_5\text{O}_{12}$ ) is of particular interest because, firstly, the interesting parts of the diagrams are confined in the low temperatures range (2–30 K), mainly due to its very low inversion (or compensation) temperature  $T_1$  ( $\cong 7$ –8 K), and secondly, the state of the  $\text{Yb}^{3+}$  ion is suitable for the development of theoretical models with rather good approximations [1–3] and generally this compound can be considered as a good example for the study of different kinds of magnetic phase transitions. Previous experimental work has been reported [4–8] which show a general qualitative agreement with the theoretical predictions especially for the  $\langle 111 \rangle$  and  $\langle 100 \rangle$  directions of the applied magnetic field. But most of them were made in high pulsed fields or limited to 4 or 8 T in static fields. Moreover, for the  $\langle 110 \rangle$  direction, there is still a controversy about the existence and the nature of some of the transitions lines. It was then of a great interest to obtain new reliable experimental data in high fields. In the present work, the phase diagram of YbIG is investigated

by means of precise magnetisation measurements in static magnetic fields up to 16 T applied along the  $\langle 110 \rangle$  direction.

## 2. Experimental

Using the extraction technique in superconducting magnets, the magnetisation has been measured in high dc magnetic fields up to 16 T from 1.5 K to 50 K. The sample is a spherical single crystal obtained by the standard PbO/PbF<sub>2</sub> flux method (diameter: 3.1 mm, weight: 108.45 mg) and oriented along a  $\langle 110 \rangle$  crystallographic direction by the X-ray Laue technique within an error of  $< 1^\circ$ . All the magnetisation results are reported in Bohr magneton by the  $\text{Yb}_3\text{Fe}_5\text{O}_{12}$  formula unit and the magnetic field  $H$  is the external applied one.

## 3. Results and discussion

The field dependence of the magnetisation  $M_T(H)$  for some sample temperatures are shown in Fig. 1. As expected, the curves are almost linear and identical for 2 K and 7.8 K up to 16 T. The fact that they extrapolate to the origin is characteristic of a type {2} canted phase which originates from domains of spontaneous magnetisation

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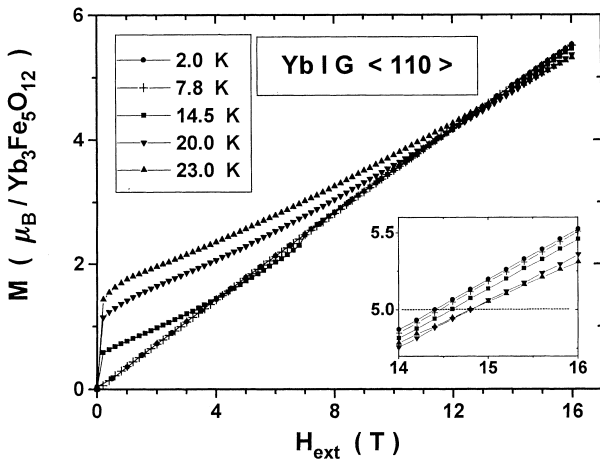


Fig. 1. Magnetisation of YbIG at several temperatures versus magnetic fields up to 16 T.

along easy  $\langle 111 \rangle$  directions perpendicular to the  $\langle 110 \rangle$  field [6]. At 14.5 K the curve in high fields is also of the same type but, below 6.92 T, the lower part of the curve is non linear and extrapolates to a spontaneous value, indicating that the corresponding type  $\{1\}$  canted phase originates from other  $\langle 111 \rangle$  directions closer to  $\langle 110 \rangle$  (at  $35^\circ.3$ ) [6]. In the vicinity of the transition field, the magnetisation undergoes a rapid variation which can be taken as a magnetisation jump  $\Delta M \cong 0.18 \mu_B/\text{f.u.}$  between the two canted phases ( $\{1\} \leftrightarrow \{2\}$ ). Above 20 K there is no such transition in the whole field range. At these temperatures the high field phase is of the collinear type and the slopes of the curves are temperature dependent as shown in the insert of Fig. 1. On the other hand, it is interesting to detail the low field behaviour near the inversion (or compensation) temperature  $T_i$ . In Fig. 2, two typical curves, at 4 K (Fig. 2a) and 11 K (Fig. 2b), are plotted together with their derivatives  $(dM/dH)_T$  with respect to the magnetic field  $H$  obtained by simple numerical differentiation. In each case, a transition field  $H_t$  is well defined by a sharp peak in the differential susceptibility, more characteristic of a first order transition of the same type than previously observed at 14.5 K (Fig. 1) with a magnetisation jump between the two canted phases (Fig. 2c).

In order to further examine the lines in the diagram, especially those which are almost vertical (i.e. parallel to the  $H$  axis), the magnetisation has been recorded at constant external magnetic field  $H$ , while the temperature is varied by steps from 2 K to 50 K. The results for 0.85 T and 7.97 T, respectively, typical of the low field and the high field regions, are reported in Fig. 3. At  $H=0.85$  T (Fig. 3a), three transitions are clearly observed. The first two, accompanied by a sharp peak in the derivative  $(dM/dT)_H$ , correspond to the first order transition lines between the canted phases on each side of the inversion temperature  $T_i$ . The third one, on the high temperature side, scarcely visible on the  $M_H(T)$  curve, is determined by a kink at 22.5 K in the derivative  $(dM/dT)_H$  and correspond to the

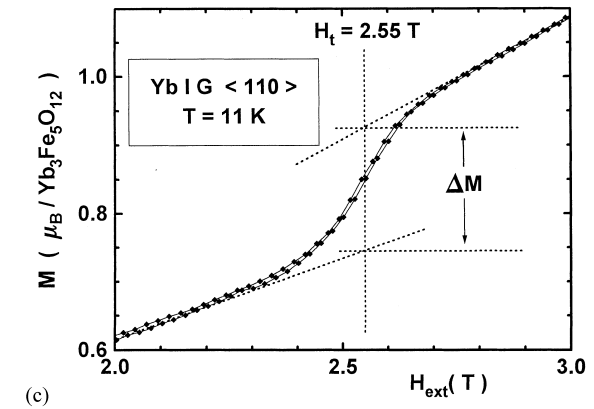
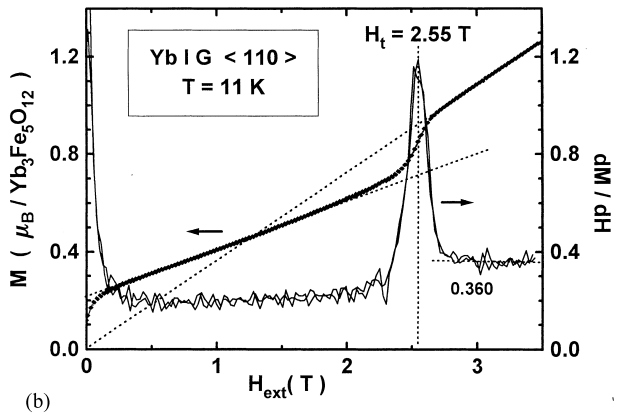
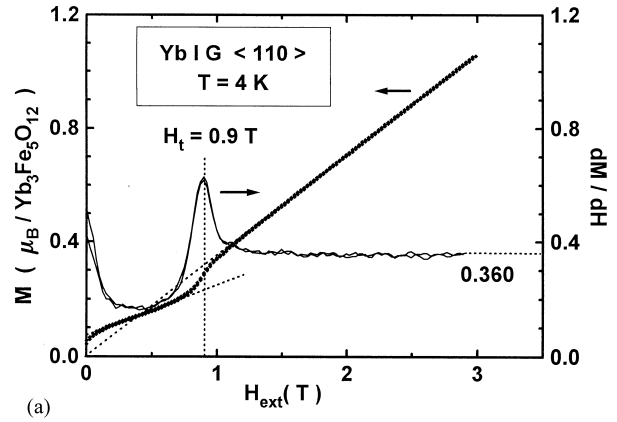


Fig. 2. Magnetisation and differential susceptibility curves of YbIG in fields up to 3.5 T; (a):  $T=4$  K; (b):  $T=11$  K; (c): Determination of the magnetisation jump on the 11 K  $M_T(H)$  curve.

second order transition line between the high temperature  $\langle 110 \rangle$  collinear phase and a type  $\{1\}$  canted phase. At  $H=7.97$  T, besides the second order transition occurring at 18 K, only one first order transition exists at 15 K with a magnetisation jump  $\Delta M \cong 0.15 \mu_B/\text{f.u.}$  as defined in the Fig. 3c.

All the transition points obtained from either  $M_T(H)$  or  $M_H(T)$  are reported in Fig. 4 and they are in excellent agreement with each others except in the immediate vicinity of the inversion point which is detailed in the insert. It comes mainly from the difficulty to assign the

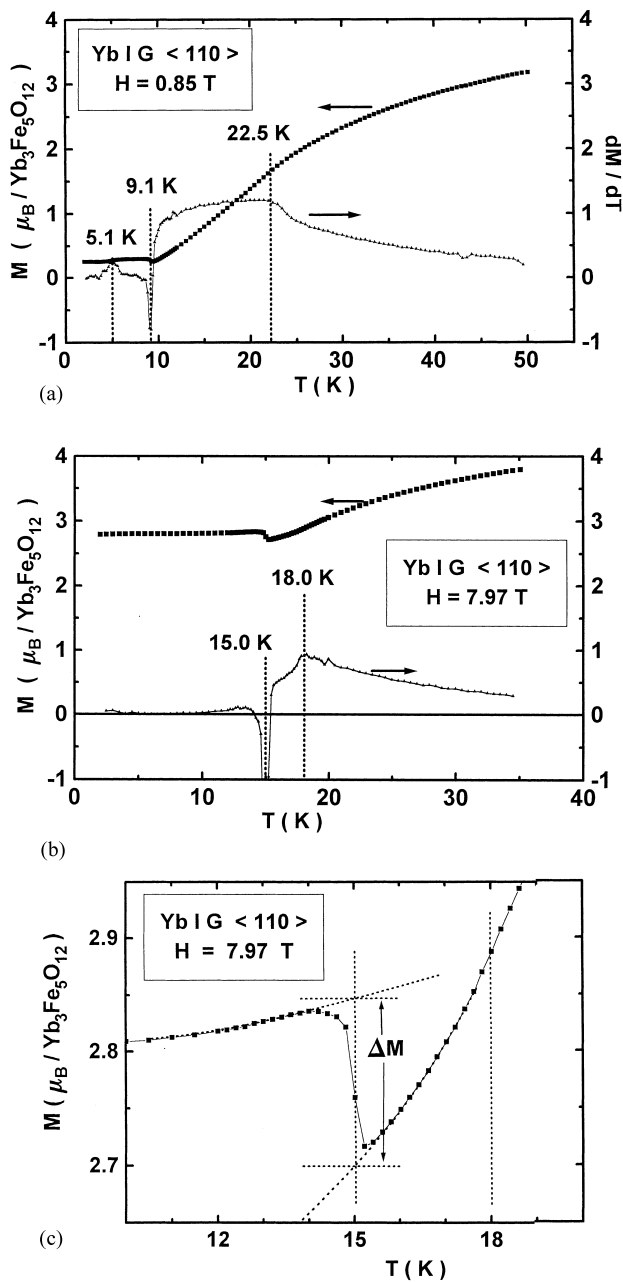


Fig. 3. Thermal variations of the YbIG magnetisation  $M_H$  and its derivative  $(dM/dT)_H$  at constant external fields; (a):  $H=0.85$  T; (b):  $H=7.97$  T; (c): Determination of the magnetisation jump on the  $7.97$  T  $M_H(T)$  curve.

transition points on the  $M_H(T)$  curves in the very low field region ( $H < 0.4$  T). However, two features can be pointed out; (i): the  $M_T(H)$  curves show a rather marked anomaly, the field of which,  $H_t$ , has an extended minimum for  $0.34$  T around  $T=7.8$  K; (ii): the two lines, given by the  $M_H(T)$  curves points, collapse progressively as  $H$  is decreased, for  $0.052$  T to a curve with only a well marked second order-like kink at  $T=7.82$  K. This is in favour of the existence, at least very close to  $T_1=7.82$  K, of a second

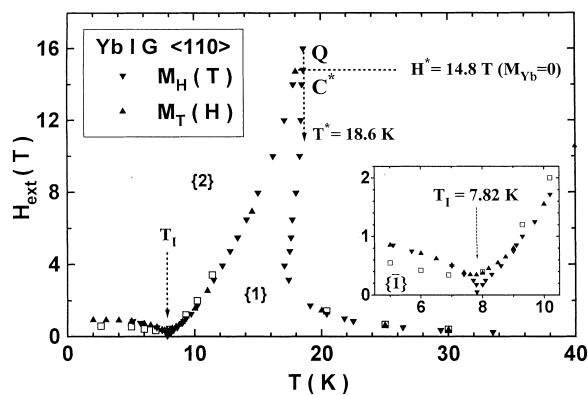


Fig. 4. Experimental magnetic phase diagram of YbIG for the  $\langle 110 \rangle$  direction (open squares are from [8]).

order transition line separating the two inverse canted phases  $\{1\}$  and  $\{\bar{1}\}$ , starting at  $T_1$  and going upward.

All these features are to be connected with the fact that, especially close to the inversion point  $T_1$ , the free energy of the three different phases are very close to each other and, due to the small size of the total magnetisation, its dependence with the magnetic fields is very small. Therefore, the effects of impurities or local inhomogeneities, for example a little amount of Pb substituted on rare earth or iron sites, may overcome the variations due to the field and change the relative stability of the phases, enlarging the range where they can coexist together.

It is important to note that no measurable hysteresis is ever observed, even in this low field region, in accordance with the recent 1 kHz ac susceptibility measurements of Ref. [8] which have been partly reported on the same figure for comparison. Despite the excellent agreement for the second order outer line, there are some discrepancies in the position of the two first order transition lines. But they can be explained by a smaller value of the 0 K spontaneous magnetisation of their sample, inducing smaller transition fields at very low temperatures and a correlated shift of the inversion point and related lines to the left. Therefore, the hysteresis observed in pulsed fields [5,6] may only be the result of some time after-effects which can appear at such high field variation rates ( $\sim 10^6$  T s $^{-1}$ ) if the associated relaxation times reach the  $10^{-7}$  s range at these low temperatures. In that case the transition is expected closer to the instability limit line of the phase than on the centre line of equal free energy. It is also important to point out that all the first order transitions are observed with an extension over some range of field and temperature, making them less sharp than they might be in theory, smoothing the associated differential susceptibility peaks and yielding to some uncertainties for extracting the right value of the associated magnetisation jump coming from the necessity to extrapolate the behaviour of each phase to the centre line as shown on Fig. 2c and Fig. 3c. Nevertheless, such a magnetisation jump can be unambigu-

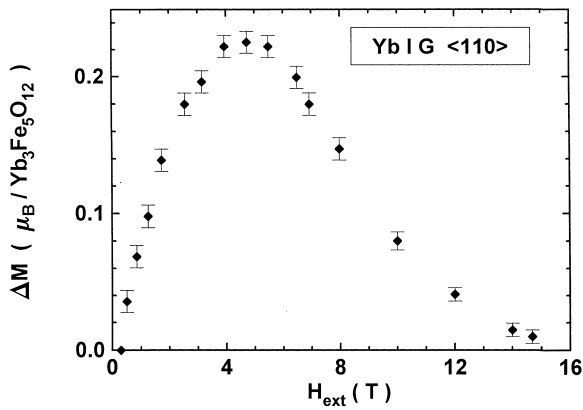


Fig. 5. The magnetisation jump along the upper transition line between the canted phases {2} and {1} as a function of the magnetic field.

ously defined and followed up to 14.7 T as seen on Fig. 5. For higher fields, the situation becomes less clear because the two transitions are too close and cannot be distinguished so easily. Only the point  $C^*$  ( $H^*=14.8$  T,  $T^*=18.6$  K, Fig. 4) where the horizontal line in the collinear phase ( $M=5\mu_B/\text{f.u.}=M_{\text{YIG}}$ ,  $M_{\text{Yb}}=0$ ) meets the second order line with the occurrence of the canted phase {2} can be well defined by the point where the slope of the  $M_T(H)$  magnetisation curves which vary strongly in the collinear phase become almost temperature independent in the canted phase (insert of Fig. 1).

It seems that, close to the point  $C^*$ , in the canted region, the magnetisation of the phase {2} maintains an excess of  $\sim 0.05\mu_B/\text{f.u.}$  over the value in the collinear phase which is by continuity the starting value of phase {1}. But it is not clear if the rapid variation of  $M$  between 17 K and 18.5 K observed on  $M_H(T)$  is to be taken as the transition itself (normal first order enlarged), as a rapid change preceding a

symmetry-affected first order transition in the Alben classification [1], or a second order transition according to the Loos theory [2].

As a conclusion, our upper line canted-canted is of first order up to 14.7 T, and seems to continue, with may be a change in nature, up to 16 T, yielding to a critical point  $Q$  ( $H_Q \cong 16$  T,  $T_Q \cong 18.6$  K) above the  $C^*$  point. For a better understanding of this critical region, higher resolution experiments in higher magnetic fields would be needed.

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