First-order magnetic transition in $Yb_2Ti_2O_7$

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The very nature of the ground state of the pyrochlore compound $Yb_2Ti_2O_7$ is much debated, because experimental results demonstrate evidence for either a disordered ground state or a long-range ordered ground state. Indeed, the delicate balance of exchange interactions and anisotropy is believed to lead to competing states, such as a quantum spin liquid state or a ferromagnetic state which may originate from an Anderson-Higgs transition. We present a detailed magnetization study demonstrating a first-order ferromagnetic transition at 245 and 150 mK in a powder and a single-crystal sample, respectively. Its first-order character is preserved up to applied fields of $\sim$200 Oe. The transition stabilizes a ferromagnetic component and involves slow dynamics in the magnetization. Residual fluctuations are also evidenced, the presence of which might explain some of the discrepancies between previously published data for $Yb_2Ti_2O_7$.

I. INTRODUCTION

Magnetism affected by geometrical frustration is an active field due to the ability to generate new and unusual magnetic phases [1]. In this context, the pyrochlore oxide materials $R_2M_2O_7$ (R = rare earth, M = metal) form a very rich family in which a large diversity of new physics can be explored [2]. Specifically, the rare-earth ions lie on the vertices of corner sharing tetrahedra, forming the highly frustrated pyrochlore lattice. Depending on the rare-earth element, the anisotropy of the spins as well as the exchange and dipolar interactions can be varied so that different model Hamiltonians can be studied within this structure. One of the most spectacular realizations is the spin-ice phase (mainly studied with R = Dy and Ho; M = Ti) [3,4] in which the local spin arrangement obeys the ice rule (two spins point into and two spins point out of every tetrahedron in the structure) and which possesses a macroscopically degenerate ground state. This state is induced by the strong uniaxial anisotropy along the local $\langle 111 \rangle$ axes of the tetrahedra, combined with a resultant ferromagnetic interaction. With these ingredients and in the presence of strong transverse fluctuations, a new magnetic state is expected to be stabilized, the quantum spin ice (QSI) in which exotic excitations are predicted [5–7].

$Yb_2Ti_2O_7$ has been proposed as a good candidate for stabilizing the QSI state [8,9]. Indeed, the exchange in $Yb_2Ti_2O_7$ is highly anisotropic, with a strong ferromagnetic component akin to the Ising exchange of spin ice [8,10,11], despite an XY-like anisotropy perpendicular to the local $\langle 111 \rangle$ directions [12,13]. At low temperature, using a model Hamiltonian with anisotropic exchange parameters deduced from experiments, a first-order phase transition towards a long-range ferromagnetic order is predicted [9,14–16].

Experimentally, the existence of a long-range magnetic ordering in this compound is debated, suggesting a fragile ground state with respect to perturbations. In an early study, a peak was observed around 210 mK in the specific heat of a polycrystalline sample [17]. It was later shown to be associated with a first-order transition and an abrupt slowing down of the fluctuations in the low-temperature phase [18].

Below the transition, depending on the nature of the samples (single crystal or polycrystal) and the crystal growth conditions, different results have been obtained. Some neutron scattering measurements demonstrate ferromagnetic long-range order (LRO) [14,19] while others do not [20–22]. A discrepancy is also observed in muon spin relaxation measurements (μSR) where an anomaly at the transition is present [18,23] or not [24]. In the meantime, it was shown that the peak in specific heat strongly depends on the samples [25,26] so that the presence of a transition towards a long-range order might depend on the sample quality.

It has been suggested that the specific heat anomaly, however, does not necessarily correspond to a magnetic ordering [24,25]. It is therefore essential to probe another thermodynamic quantity which should be more sensitive to the magnetic nature of the transition: the magnetization. In this article, we show that the magnetization of $Yb_2Ti_2O_7$ presents a first-order transition in both a powder sample and a single crystal which was shown to develop additional magnetic intensity on structural peaks [14]. The first-order nature of the transition invoked in previous studies [14,18,24] is proved by the existence of a small thermal hysteresis (of a few millikelvins in width). The transition is accompanied by strong time-dependent effects. The magnetization value below the transition temperature is consistent with the stabilization of a ferromagnetic ordering with a reduced spontaneous moment, suggesting a strongly fluctuating spin component. Significantly the first-order behavior occurs below the peak in the specific heat where only a deviation in the susceptibility is observed.

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II. EXPERIMENTAL DETAILS

A. Sample preparation

Three samples were measured: (i) a compacted powder sample; (ii) a crushed powder sample mixed with Cu grease to ensure a good thermal coupling, both obtained from the same synthesis; and (iii) a single crystal, with the magnetic field applied along the [100] and [110] axes. These samples are the same as those used in Ref. [23] (and Refs. [14,19] in the case of the single crystal).

Polycrystalline samples of $Yb_2Ti_2O_7$ were synthesized using a solid state reaction method. Stoichiometric quantities of Yb_2O_3 and TiO_2 powders were repeatedly ground, pressed into pellets, and sintered at 1300 °C for several days. The method used for the growth of the single crystal is described in Ref. [14].

The shape of these samples is irregular, thus preventing an accurate correction for demagnetizing effects. Using the formula provided for parallelepipeds [27], the demagnetizing factors N could be nevertheless estimated:

(i) For the single crystal, N is within the range 2.7 to 3.5 and 4.7 to 5.3 (cgs units) for measurements along the [100] and [110] directions, respectively.

(ii) The compacted powder has an elongated shape, giving approximately $N \approx 2$ (cgs units).

(iii) For the crushed powder, the estimation is much harder, but N should be larger than the value for the compacted powder.

B. Magnetic measurements

Magnetization and ac susceptibility measurements were performed down to 70 mK on two superconducting quantum interference device SQUID magnetometers equipped with dilution refrigerators developed at the Institut Néel [28]. The setups can measure absolute values of the magnetization by the extraction method. One magnetometer is devoted to low-field measurements (0–4 kOe) and the second to high-field measurements (0–80 kOe).

The samples were attached to a copper tress suspended from the mixing chamber of the dilution fridge and thermalized with Apiezon grease. The crushed powder was prepared in a copper pouch and mixed with Apiezon grease (for thermalization and to avoid reorientation of the powder under the magnetic field).

III. RESULTS

A. Magnetic characterization below 4.2 K

Figure 1 shows the magnetization measured in a small field as a function of temperature up to 4 K for all the samples. Above 1.5 K, the susceptibility follows a Curie-Weiss law (see inset of Fig. 1), consistent with previous results above 2 K [12,19,29,30]. Some differences are obtained between the field orientations. These differences are mainly due to demagnetization effects that depend on the shape of the sample, although a small contribution from the exchange anisotropy and the magnetocrystalline anisotropy of the system is also expected [31].

Below 1.5 K, the susceptibility continues to increase with decreasing temperature but deviates from the Curie-Weiss
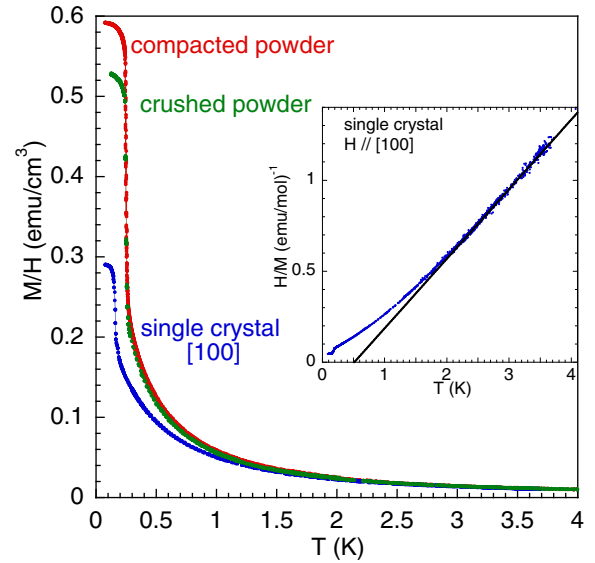

FIG. 1. (Color online) M/H vs T for the single crystal (H // [100]) and for the two powder samples. The applied field was 5 Oe below 1 K and 100 Oe above (where the magnetization M is linear in H for such fields). Inset: H/M vs T for the single crystal. The line is a fit to a Curie-Weiss law H/M (emu^{-1} mol) $= -0.195 + 0.382T$ for $T > 1.5$ K.

law. Then at 250 mK for the powder (165 mK for the single crystal), the magnetization M increases abruptly and reaches a plateau at low temperature. This very sharp increase immediately suggests a magnetic transition, especially as it matches approximately with the anomaly in specific heat [23].

The magnetization curves up to 80 kOe and down to 90 mK were performed for the compacted powder and with the field aligned along the [100] and [110] directions for the single crystal. Below 2 K, the magnetization is almost saturated above 60 kOe and reaches about 1.75 μ_B/Yb, as shown in Fig. 2.
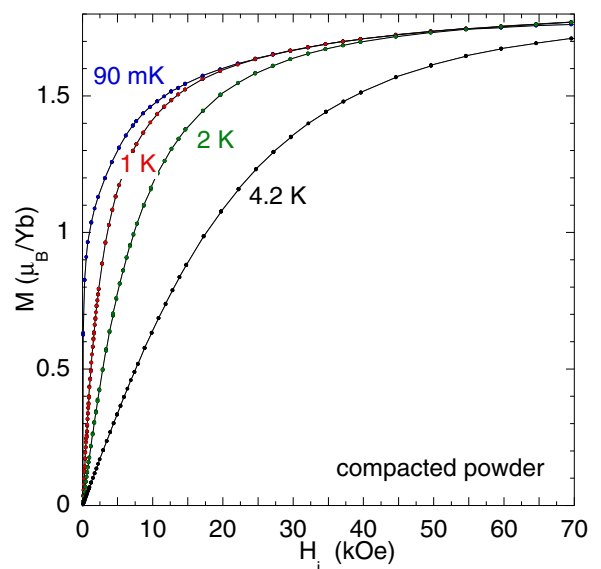

FIG. 2. (Color online) M vs H_i for the compacted powder for temperature between 90 mK and 4.2 K. The internal field $H_i = H - NM$ was calculated with $N = 1.68$ (cgs units) [32].
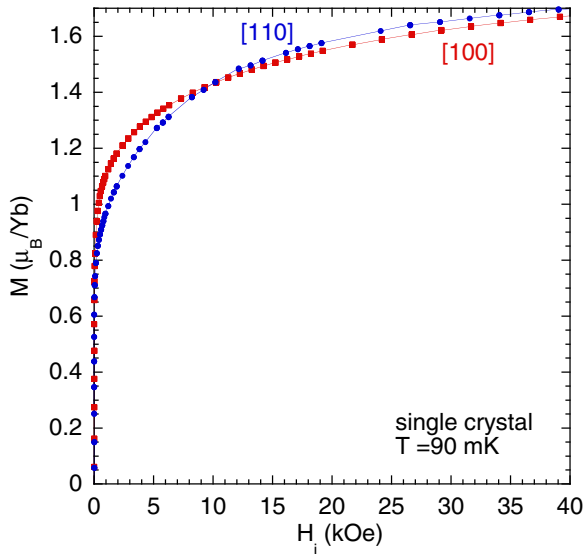
FIG. 3. (Color online) M vs H_i for the single crystal at 90 mK. The internal field H_i was calculated with $N_{[100]} = 3.38$ and $N_{[110]} = 3.8$ (cgs units) [32].

FIG. 4. (Color online) M vs H_i for the single crystal at $T = 80$ mK and with H applied along the [100] direction. H_i was calculated with $N_{[100]} = 3.38$ (cgs units) [32].

Above 2 K, no clear anisotropy is observed. Below 1 K, the magnetization curves start to separate significantly. In particular, in the low-field region, the magnetization increases much faster when the field is applied along the [100] direction, as can be seen in Fig. 3.

B. The ferromagnetic ordering

In an ordered ferromagnetic state, a system has a spontaneous magnetization and the initial intrinsic susceptibility is expected to diverge. The measured value of the bulk susceptibility is then equal to the inverse of the demagnetizing factor N. For the four measurements performed (two powder samples and two orientations of a single crystal), the measured M/H value (which can be considered as the susceptibility in such small fields) on the plateau is larger for samples with the smaller demagnetizing factors, as expected, and is in the range of $1/N$ (see Fig. 1). In particular, preliminary measurements were performed on a roughly parallelepipeded crystal, along its long direction which corresponds to an arbitrary crystallographic direction. In that case, the demagnetization factor was better characterized [$N \approx 3.2$ (cgs units)], and the value at the M/H plateau matches the $1/N$ value, revealing the ferromagnetic nature of the transition.

In a ferromagnetic LRO phase, the ferromagnetic ordered component can be associated with the spontaneous magnetization which is in turn deduced from magnetization curves M vs H, provided that the field is applied in the direction of the ordered moment. In Yb$_2$Ti$_2$O$_7$, the determination of this ordered component appears especially difficult, since the magnetic structure of the LRO phase is not known precisely.

To address this question, we have analyzed our M vs H magnetization curves for the powders and for the single crystal measured along the [100] direction, and using a less comprehensive data along the [110] direction, as we did not perform precise measurements at very low fields. It is worth no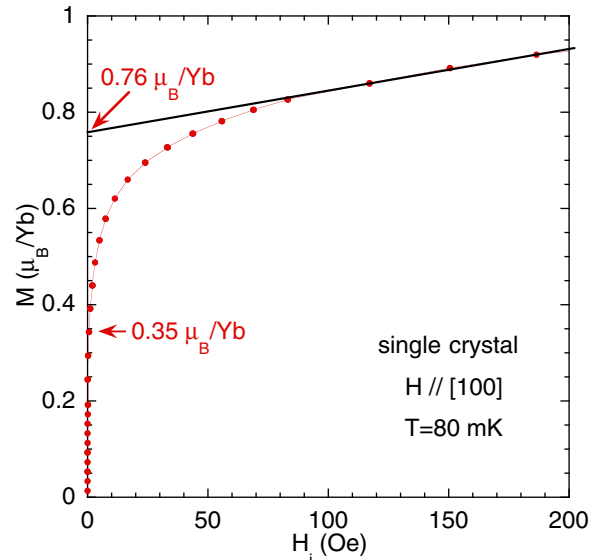ting that the magnetization as a function of field deviates very quickly from the demagnetization line ($=H/N$), in agreement with the fast decrease of the M/H plateau value observed in the M vs T curves when the field is increased, as will be detailed below. This indicates that the value of the spontaneous moment is quite small.

To make quantitative comparisons, we have plotted the magnetization as a function of the internal field H_i, supposing, as stated previously, that the demagnetizing factors N equal the values of H/M in very low field. The first conclusion is that the [100] direction seems to be the easy axis of magnetization, suggesting that the moments are mainly oriented along the [100] direction. This result seems reasonable since it is approximately the obtained direction for the magnetic moments in Yb$_2$Sn$_2$O$_7$ [33].

So, supposing that the ferromagnetic component is parallel to the [100] direction, we can estimate the spontaneous moment from the low-temperature curve as shown in Fig. 4. Strictly speaking, the spontaneous magnetization M_s corresponds to the value at which the curve leaves the zero internal field slope. This would lead in the present case to a spontaneous moment of about 0.35 μ_B/Yb. However, due to the curvature around the zero internal field, another criterion is usually considered: the spontaneous magnetization may be interpreted as the intercept of the slope of the M vs H_i curve at larger field with the zero field axis. This has been done in Fig. 4 and gives a value of about 0.8 μ_B/Yb. Note, there is no clear break in the magnetization curve which can be used to fix the field range over which to carry out the extrapolation. The smooth shape of the M vs H curves may be due to the presence of a fluctuating component in the magnetization.

The above analysis gives a ferromagnetic ordered moment in the range of 0.35 and 0.8 μ_B/Yb at 80 mK in the single crystal, to be compared to the 1.75 μ_B/Yb value of the magnetization in high field. The same kind of analysis in the powder gives a moment three times smaller, which is consistent with the hypothesis of an ordered moment along [100]. A

possible temperature dependence of the spontaneous moment must be considered, since an increase in the ordered moment may be expected as the temperature is reduced further below T_C. For the single crystal, our analysis was carried out at 80 mK which corresponds to $T_C/2$. The same procedure was followed at 110 mK and the results were found to be comparable. For the powder sample, we performed the analysis between 80 (about $T_C/3$) and 200 mK, and no significant dependence of the spontaneous moment with temperature was observed. These results suggest that the spontaneous moment will not increase significantly at lower temperature and point out the first-order nature of the transition.

C. First-order transition and time-dependent effects

A detailed study of the magnetization around the transition has been performed. To ensure accurate results, measurements had to be performed with well-controlled temperature regulation and extremely slow cooling and warming rates. The protocol was the following: (i) regulate at a given temperature, (ii) take a large number of measurements (between 40 and 100) so that the magnetization reaches equilibrium at this temperature, and (iii) change the temperature with a step of 5 or 2 mK depending on the measurements. The temperature was ramped between 80 and 400 mK, cooling and warming the sample. The equivalent ramping rate is between 9 and 18 mK/h. The obtained magnetization as a function of temperature for the single crystal is shown in Fig. 5. It can be seen that at the transition, at a fixed temperature, a strong relaxation occurs. As shown in the inset of Fig. 5 where the magnetization is
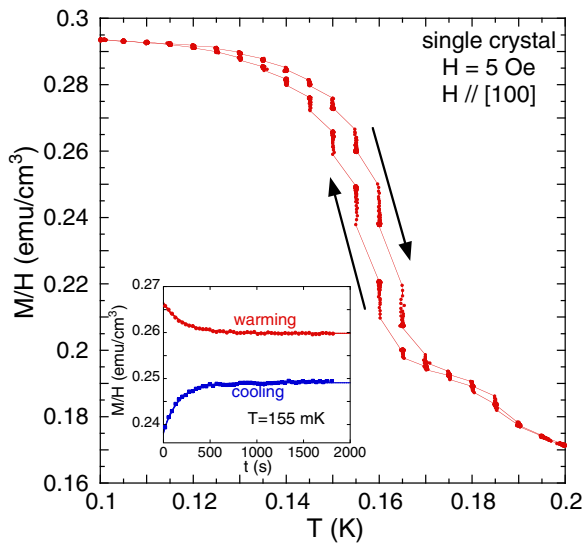

FIG. 5. (Color online) M/H vs T for the single crystal in an applied field $H = 5$ Oe parallel to the [100] axis at the proximity of the transition. The temperature was swept in steps of 5 mK and 100 extractions were made at each temperature ($\sim$30 min at each temperature). Inset: Isotherm as a function of time t at $T = 155$ mK when warming (red circles) and when cooling (blue squares). The lines are fitted to the exponential: $\frac{M}{H}(t) = \frac{M_{eq}}{H} - \frac{\Delta M}{H} \exp(-t/\tau)$. When warming $\tau = 207$ s, $\frac{M_{eq}}{H} = 0.260$ emu cm^{-3}, and $\frac{\Delta M}{H} = -6.58 \times 10^{-3}$ emu cm^{-3}. When cooling $\tau = 165$ s, $\frac{M_{eq}}{H} = 0.249$ emu cm^{-3}, and $\frac{\Delta M}{H} = 1.08 \times 10^{-2}$ emu cm^{-3}.
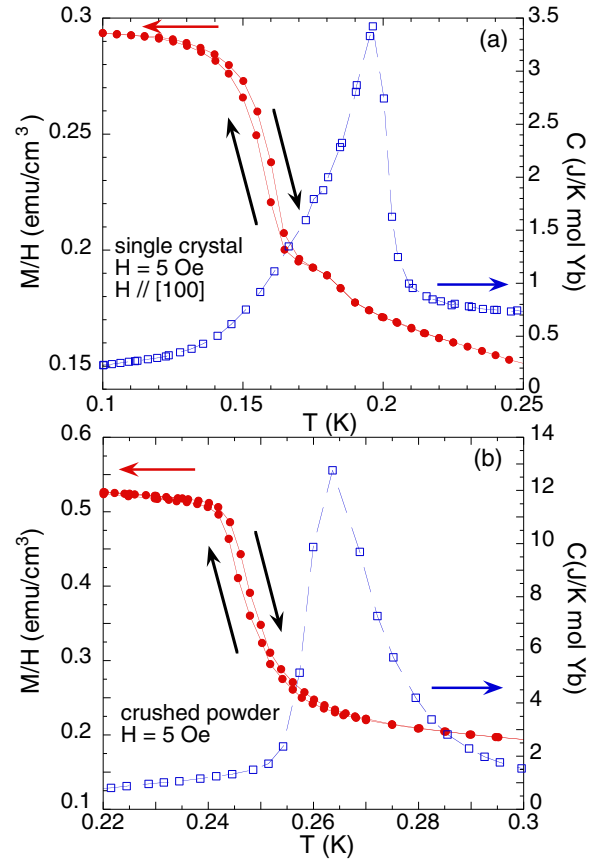

FIG. 6. (Color online) (a) M/H vs T for the single crystal in an applied field $H = 5$ Oe parallel to the [100] axis, extracted from Fig. 5 with only the equilibrium value of the magnetization plotted compared to the specific heat data. (b) The equivalent data for the crushed powder.

plotted as a function of time, at 155 mK, the equilibrium magnetization is reached after times as long as 600 s.

Figure 6(a) shows the equilibrium values of the magnetization at the transition (obtained from Fig. 5) as a function of temperature for the single crystal. It can be seen that a small hysteresis is present (which is much narrower than that for a fast temperature sweep), indicating a first-order like behavior. Also shown is the specific heat data on the same crystal. A subtle change of slope occurs in the magnetization at the peak in specific heat, while the first-order transition develops below this peak. The bump observed at $\approx$180 mK before the sharp increase is not present in the magnetization of the powder sample as shown in Fig. 6(b) and might be due to a sample inhomogeneity, a consequence of difficulties in sample preparation [25,26].

From the magnetization, it appears, that the first-order transition occurs around 150 mK in this single crystal. The transition extends over about 20 mK and the hysteresis width is about 3 mK. For the powder sample, the transition occurs around 245 mK, but the width of the transition and the width of the hysteresis are similar.

Zero-field-cooled–field-cooled (ZFC-FC) magnetization shows an irreversibility below the temperature of the transition (see Fig. 7) [34]. In ordered materials, such irreversibility is
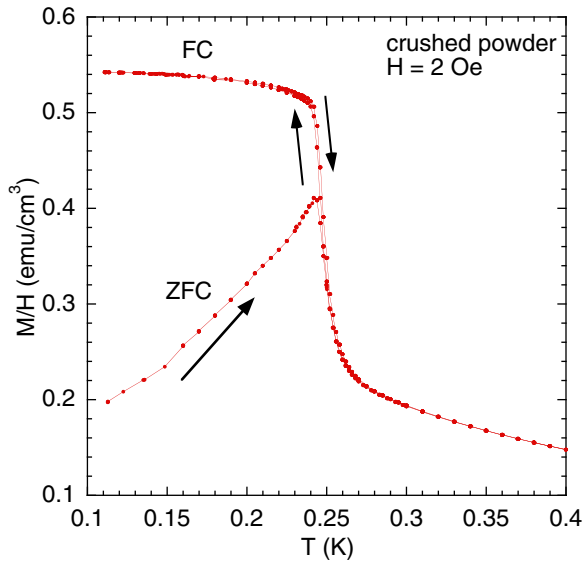
FIG. 7. (Color online) M/H vs T for the crushed powder for $H = 2$ Oe measured with the ZFC-FC procedure.

FIG. 8. (Color online) M/H vs T for the compacted powder for several fields between 2 and 500 Oe. Inset: M/H vs T for the single crystal for several fields between 30 and 1000 Oe, applied along the [110] direction.

often ascribed to domain freezing. The ZFC-FC irreversibility is strongly reduced when the applied field is increased and is suppressed at about 500 Oe. This suggests that small fields are enough to overcome the barriers involved in the freezing. In addition, hysteresis loops were performed. A small hysteresis opens below the transition. In the powder samples, the hysteresis is about 20 Oe in width at 80 mK (giving a coercive field of 10 Oe) and closes at about 500 Oe. The hysteresis is 10 Oe in width at 200 mK and disappears at the transition. In the single crystal, with the field applied along [100], the hysteresis is smaller (less than 5 Oe in width at 80 mK). These results are consistent with the ZFC-FC measurements. They imply that the domain pinning is quite weak and that the reversal of the magnetization in an applied field mainly occurs by a continuous rotation of the moments.

The effect of the field on the transition has also been investigated. For the powder samples, a field as small as 10 Oe is enough to reach the regime of nonlinear susceptibility, resulting in a smaller value of M/H (see Fig. 8). However, up to at least 20 Oe, the transition temperature and the transition and hysteresis widths remain constant. At 50 Oe, the transition starts to become smoother and shifts to higher temperatures, but the hysteresis persists. Above 250 Oe, the hysteresis is lost. The "step" in the magnetization continues reducing in magnitude and broadening.

The results are qualitatively similar in the single crystal (see inset of Fig. 8) and independent of the direction of the applied field. However, the transition in the single crystal appears to be less sensitive to the magnetic field, except the bump at 180 mK which is suppressed in 10 Oe. The susceptibility remains linear and the transition is unchanged up to 30 Oe. Above 50 Oe, the transition broadens and starts to shift to higher temperatures (which seems analogous to the reported behavior of the ac susceptibility in Yb$_2$Sn$_2$O$_7$ [35]). Above approximately 200 Oe, the hysteresis disappears (similar to the powder samples) and, finally, the amplitude of the "step" decreases significantly above 500 Oe. The main conclusions
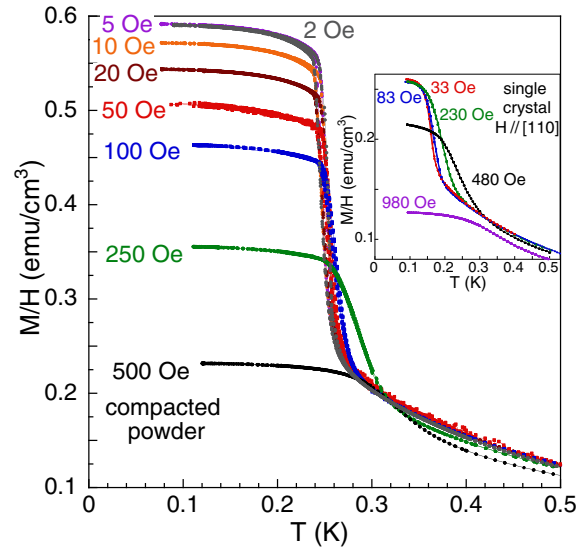
from this field-induced behavior are (i) the hysteresis and so the first-order character are preserved up to an applied field of ~200 Oe and (ii) the increase of the applied field broadens and shifts the magnetization step to higher temperatures, as expected for ferromagnetic transitions. Above 200 Oe, from the magnetization measurements alone, we cannot conclude
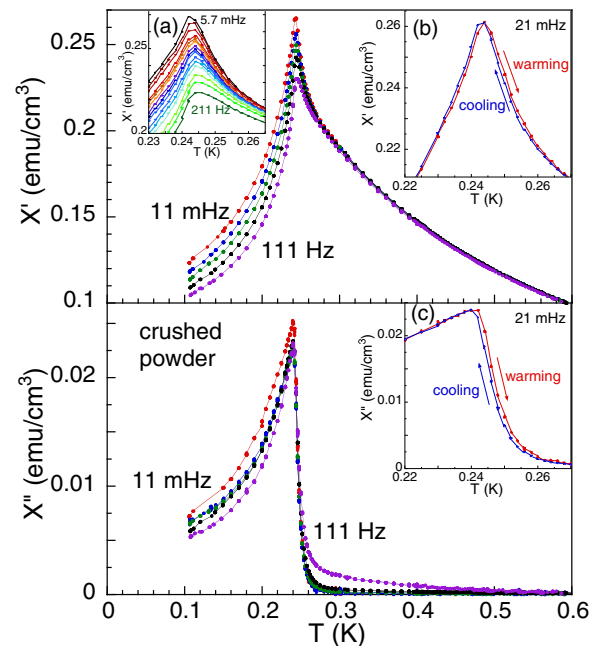

FIG. 9. (Color online) The ac susceptibility, in phase χ' (top) and out-of-phase χ'' (bottom), vs T for the crushed powder sample. Data were collected while cooling. The amplitude of the ac field is 1 Oe. (a) Magnification of the χ' peak for frequencies between 5.7 mHz and 211 Hz. (b) χ' and (c) χ'' vs T at the transition at 21 mHz for the cooling and warming ramps.

whether the "step" is a signature of a phase transition or rather of a crossover to a field-polarized state.

The dynamics at the transition have been studied via ac susceptibility and are shown for the powder sample in Fig. 9. χ' shows a sharp peak, which is associated with the onset of the out-of-phase part χ'' of the susceptibility. In the measured frequency range (5.7 mHz to 2.11 kHz), no frequency dependence of the peak position is observed (in both the powder and crystal), but the amplitude of the peak increases a little when the frequency decreases [see Fig. 9(a)]. A small hysteresis is observed in χ' and χ'' [see Figs. 9(b) and 9(c)], in the same temperature range as the magnetization. The characteristics of the ac susceptibility are consistent with the picture of a first-order magnetic transition. The χ'' onset would then be the signature of the dissipation at the transition. The temperature of the χ' peak gives a transition temperature $T_c = 243 \pm 1$ mK. These results are in strong contrast with ac measurements in $Yb_2Sn_2O_7$ [36], where a glassy behavior was reported.

IV. DISCUSSION AND CONCLUSIONS

Our results show an agreement between thermodynamic measurements, i.e., magnetization and specific heat, supporting a first-order transition in $Yb_2Ti_2O_7$, at 245 mK and 150 mK for the powder and the single crystal, respectively, and involving a LRO ferromagnetic component. This is qualitatively consistent with neutron measurements performed in the single crystal [14,19], although our results (transition temperature, hysteresis width, and value of the ordered moment) do not match quantitatively.

These features, a first-order transition resulting in a small ferromagnetic ordered magnetic moment suggesting a fluctuating component, are reminiscent of other pyrochlore compounds which exhibit long-range order [2]: $Gd_2Sn_2O_7$, the archetype of a dipolar Heisenberg pyrochlore antiferromagnet [37,38], and $Tb_2Sn_2O_7$, an ordered spin ice [39]. In these compounds, persistent spin dynamics are observed but the ordered moment is much larger than that in $Yb_2Ti_2O_7$, most probably resulting in a more robust long-range ordering.

The fragility of the ordered state in $Yb_2Ti_2O_7$ results in a sample dependence of the transition. The reasons for this dependence are under debate and are of great importance to understand the mechanisms of the ordering in $Yb_2Ti_2O_7$. Questions immediately arise from the results reported here: magnetization measurements definitely show that a first-order magnetic transition occurs in these samples around the vicinity of the peak in specific heat. Does suppression of the ordering temperature in the single crystal suggest that disorder induced by the growth process increases spin fluctuations? Do other samples which exhibit a peak in specific heat, but no evidence of long-range ordering in neutron scattering [25], also show evidence of a transition in magnetization data? The analysis of our single-crystal results as a function of temperature may give a preliminary answer to this question: the presence of a reversible bump before the step in the magnetization seems to indicate a partial ordering first, at the specific heat maximum, before the achievement of the transition at lower temperature. Thus it might be possible that, in other samples [24,25] in which the microscopic probes (neutron scattering, μSR) do not detect LRO, such a partial ordering could occur at the specific heat peak but without ending in a transition at lower temperature. So that the existence of a peak in specific heat may not involve a long-range magnetic ordering. If a LRO transition was also present in the magnetization in these samples, the presence of strong fluctuations might be the clue to explain why the thermodynamic probes (magnetization, specific heat) do show the evidence of a transition while the microscopic ones (neutron scattering, μSR) do not. Further magnetization measurements would be needed in these samples to answer this question.

Finally, it is interesting to discuss the origin of the transition itself. Theoretical work predicts a first-order character of the transition [9,14,15], in agreement with the above results. However, the mechanism for the transition is still debated: a Higgs mechanism from a Coulomb phase [14] or a confinement of the excitations from a thermal spin liquid state [15], for example, have been proposed. It might be of great interest to consider theoretically the effects of the magnetic field on the transition in the different scenarios and to compare them with the dependence (temperature and order) on the magnetic field reported above.

In conclusion, we have shown that in both powder and single-crystal samples, a first-order magnetic transition occurs in $Yb_2Ti_2O_7$. The transition width is about 20 mK while the hysteresis at the transition is about 3 mK. The value of the dc susceptibility below the transition indicates the existence of a spontaneous magnetization and so of a ferromagnetic component. The transition remains first order up to about 200 Oe. In larger fields the magnetization anomaly softens and shifts to higher temperatures.

ACKNOWLEDGMENTS

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