Partial magnetization plateau emerging from a quantum spin ice state in Tb$_2$Ti$_2$O$_7$

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The quantum spin ice model applied to Tb$_2$Ti$_2$O$_7$ predicts that magnetic fields applied along the [111] axis will induce a partial magnetization plateau [H. R. Molavian and M. J. P. Gingras, J. Phys.: Condens. Matter 21, 172201 (2009)]. We test this hypothesis using ac magnetic susceptibility and muon-spin relaxation measurements, finding features at 15 and 65 mT agreeing with the predicted boundaries of the magnetization plateau. This suggests that Tb$_2$Ti$_2$O$_7$ is well described by a quantum spin ice model with an effective exchange constant of $J_{eff}$ = 0.17(1) K.

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Geometrically frustrated magnets are materials in which geometrically-induced competition between interactions prevents local magnetic moments from ordering down to temperatures well below the energy scale of those interactions. If dynamic magnetic fluctuations persist to low temperatures, the quantum fluctuations of the Tb$_3^{3+}$ ions [$\Delta l/l \sim 10^{-4}$] is observed at 4.2 K and can be understood in terms of the crystal field levels of the Tb$_3^{3+}$ ions [24]. X-ray diffraction studies point to structural fluctuations in zero magnetic field [22] and find a cubic-tetragonal structural phase transition that can be resolved in magnetic fields $B \gtrsim 25$ T [22]. Inelastic neutron scattering measurements made in magnetic fields applied along the [110] direction show a feature at 0.04 THz indicating the presence of a tetragonal lattice distortion [27].

To test for the [111] magnetization plateau in Tb$_2$Ti$_2$O$_7$ we have carried out ac susceptibility and muon spin relaxation ($\mu$SR) experiments using magnetic fields applied along the Tb$_2$Ti$_2$O$_7$ [111] crystal axis. These provide complementary information on the magnetic susceptibility at two distinct timescales.

Our ac susceptibility measurements were performed using a custom made coil set, thermally anchored to the mixing chamber of an Oxford Instruments dilution refrigerator through immersion in liquid $^4$He. For our $\mu$SR experiments [25] the field was parallel to the initial muon spin polarization [longitudinal field (LF)] in the ISIS measurements (0.04 $< T < 10$ K) and with the initial muon spin polarization partially rotated [LF and transverse
field (TF) for measurements (0.025 < T < 0.9 K) using the Low Temperature Fridge (LTF) spectrometer (Paul Scherrer Institute, Switzerland). The measured parameter is the time-dependent muon decay asymmetry, A(t), recorded in positron detectors on opposite sides of the sample. Having subtracted the background and normalized the signal, this provides a measure of the spin polarization \( P(t) = [A(t) - A_{bg}]/[A(0) - A_{bg}] \) of the muon ensemble as a function of time. For all our \( \mu \)SR experiments the \( \text{Tb}_2\text{Ti}_2\text{O}_{7} \) crystals, grown using a floating zone furnace, were arranged in a mosaic and attached to a silver backing plate using a thin layer of GE varnish. The silver plate gives a temperature and field-independent background signal that can easily be identified and subtracted from the asymmetry data.

The ac susceptibility data are shown in Figure 1 (a) for the real part \( \chi' \) and (b) for the imaginary part \( \chi'' \). The data recorded at 100 and 125 mK are equivalent within error (125 mK data not shown) but there is a clear separation between these data and those recorded at 68 mK in both components of the susceptibility. In \( \chi' \) the separation is clear below \( \sim 200 \) mT and grows towards zero field. This can be compared to the theoretically predicted magnetization curves \[12\] in the inset to Fig. 1 where a separation between the predictions for 50 and 100 mK emerges below \( \sim 300 \) mT. The temperature dependence is more pronounced in \( \chi'' \), where at 68 mK the susceptibility rises to a peak at 15 mT but at \( \geq 100 \) mK it falls monotonically. As with \( \chi' \), at 68 mK \( \chi'' \) remains distinct from that at 100 mK up to \( \sim 300 \) mT, in accordance with the behavior predicted for the magnetization. Any feature at the upper boundary of the magnetization plateau is indistinct, also as predicted. Previous measurements of the ac susceptibility indicated a partial spin freezing at low temperature and a frequency dependence consistent with our results \[7, 29\].

The \( \mu \)SR data recorded in longitudinal field are shown in Fig. 2 (a) and can all be described by the equation:

\[
P_z(t) = e^{-\lambda t},
\]

where \( \lambda \) is the muon spin relaxation rate. The background is weakly relaxing in zero applied field but effectively constant in longitudinal applied field, consistent with the expected behaviour of the silver sample holder. That this equation describes the data over the whole measured field range demonstrates that the magnetic fields remain dynamic on the timescale probed by muons. The transverse field data shown in Fig. 2 (b) take a similar form except for the muon spin precession. They can be described by the equation:

\[
A_x(t) = A_x e^{-\lambda x t} \sin(\omega_x t) + A_{bg} e^{-\lambda bg t} \sin(\omega_{bg} t),
\]

where the first term describes the signal from the sample and the second term describes the background signal. The oscillations are sinusoidal rather than the conventional cosinusoidal behavior because of the detector geometry used and the angular precession frequencies are related to the magnetic fields experienced by the muons as \( \omega = \gamma \mu_B B \) (\( \gamma/2 \pi = 135.5 \text{ MHz/T}^{-1} \)).

The magnetic fluctuations in \( \text{Tb}_2\text{Ti}_2\text{O}_{7} \) have previously been shown to be fast compared with the range of timescales probed by muons \[3, 34, 32\], that is to say that the system is paramagnetic on the muon timescale, and the relaxation rate \( \lambda \) can therefore be related to the distribution width of magnetic fields at the muon stopping
site $\Delta$, the fluctuation time $\tau$, and the applied longitudinal field $B_{\text{LF}}$ by the sum of Redfield’s equation \cite{33} and a field-independent relaxation rate, $\lambda_0$:

$$\lambda = \frac{2\gamma_B^2 \Delta^2 \tau}{1 + \frac{\gamma_B^2 B_{\text{LF}}^2 \tau^2}{B_{\text{LF}}}} + \lambda_0.$$  \hspace{1cm} (3)

We include the field-independent relaxation rate, $\lambda_0$, because our data appear to tend to a field-independent value above 0.25 T, which is consistent with the behaviour seen in that field range in Ref. \cite{30}.

For the $T \leq 2$ K longitudinal field data, three regions can be identified in the field dependent relaxation rates shown in Fig. 3. At small fields up to $B_1 \sim 15$ mT, $\lambda$ increases to a peak, then falls steeply to a kink at $B_2 \sim 60$ mT, followed by a more gradual fall between 60 and 250 mT. The fields at which the peak and kink are observed are consistent with the boundaries of the magnetization plateau predicted in Ref. \cite{12} (see the inset to Fig. 1). For comparison, we plot the trend $\lambda(B_{\text{LF}}) \propto B^{-1}$ previously found for polycrystalline data at 100 mK \cite{31} as a solid line in Fig. 3. This is similar to the behaviour seen in our data above $\sim 70$ mT.

Using equation (3) we can test predictions for the field dependence of $\Delta$ and $\tau$. The simplest assumption is that neither depends on field, which effectively describes both the 5 and 10 K data. Below 5 K this model does not work over the whole field range. However, it is effective between the peak ($B_1$) and kink ($B_2$) shown in Fig. 3 thereby suggesting that a plateau exists in the local magnetic field distribution in the anticipated field range. Fitting (with $\lambda_0 = 0$) leads to $\Delta \sim 12$ mT and $\tau \sim 13$ ns at low-temperature, the fit for 25 mK being shown as the dashed line in Figure 3. Including the $\lambda_0$ values estimated using the data above 150 mT ($\sim 0.5$ MHz) reduces the value of $\Delta$ by around 15 % and increases $\tau$ by around 10 %. Independent of this, the quality of the fits is poorer for the 1.5 and 2 K data, which is consistent with thermal fluctuations breaking up the low temperature state. Above $\sim 65$ mT it is not possible to describe the data using equation (3) and the same parameters as between 15 and 60 mT. This strongly suggests that either the field distribution or the fluctuation timescale has changed.

To visualize the field-dependence of the local magnetic field distribution $\Delta$ under the assumption that the fluctuation timescale $\tau$ is independent of the weak magnetic fields being applied along [111], we can rearrange equation (3) into the form:

$$\Delta = \sqrt{(\lambda_0 - \lambda_0)(1 + \gamma_B^2 B^2 \tau^2)/2\gamma_B^2 \tau}.$$  \hspace{1cm} (4)

Fig. 4 shows the resulting $\Delta$ values assuming $\tau = 20$ ns at all temperatures. This $\tau$ value is suggested by the fits to the higher temperature data although the form of these results is only weakly dependent on $\tau$ for $12 < \tau < 25$ ns, with the plateau region consistently evident in the $T \leq 2$ K data. We expect the fits to equation (3) in the plateau region will underestimate $\tau$ since $\Delta$ is assumed constant, which is unlikely at non-zero temperature. The $\lambda_0$ values for $T \leq 2$ K were estimated by fitting equation (3)
with constant $\Delta$ and $\tau$ to the highest field data available, $\mu_0 H \gtrsim 150$ mT, and seeking a value that did not change significantly with the low-field boundary of the fitting window. In the inset to Fig. 1 we show the theoretical magnetization curves from Ref. [12]. There is good agreement in the position of the plateau region. Since we measure the local magnetic field distribution width $\Delta$ rather than $M$, it is not possible to make quantitative comparisons of the magnitude.

We also carried out muon-spin rotation measurements from 2.5 to 250 mT at 25 mK with the aim of finding evidence for the level crossing predicted to occur at the upper boundary of the magnetization plateau by the theory described in Ref. [12]. No evidence for such an effect was found, which could be due to correlations between neighbouring tetrahedra or, alternatively, that the primary relaxation mechanism for the muon spin is always the large distribution of fluctuating local fields. Instead we observed a significant negative frequency shift between the precession frequencies in the sample and the background, $K = (\omega_b - \omega_{bg})/\omega_{bg}$. Above $\sim 7.5$ mT, $-0.65 < K < -0.625$ with negligible field dependence. This value is consistent with previous measurements at higher fields [30, 31], which attributed the effect to the large sample magnetization.

In conclusion, both ac susceptibility and $\mu$SR measurements of the dynamic magnetic fields in Tb$_2$Ti$_2$O$_7$ show features suggesting that a partial magnetization plateau emerges for fields between 12 and 65 mT applied along the [111] axis. Comparing these fields to the predictions of the quantum spin ice model suggests an effective exchange [34]. Our results provide a further quantitative test for this model.

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