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Lack of static lattice distortion in $Tb_2Ti_2O_7$

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Abstract

We investigated the possibility of temperature-dependent lattice distortions in the pyrochlore compound $Tb_2Ti_2O_7$ by measuring the internal magnetic field distribution, using muon spin resonance, and comparing it to the susceptibility. The measurements are done at temperatures as low as 70 mK and external fields up to 6 kG. We find that the evolution of the width of the field distribution can be explained by spin susceptibility only, thus ruling out a temperature-dependent hyperfine coupling. We conclude that lattice deformations are absent in $Tb_2Ti_2O_7$.

(Some figures in this article are in colour only in the electronic version)

Despite its short-range AFM correlations at temperatures lower than 100 K, $Tb_2Ti_2O_7$ remains in a fluctuating paramagnetic spin-liquid state down to 70 mK [1]. This very unusual state of matter has attracted considerable attention of experimentalist and theorist alike. Recently it was demonstrated that, under pressure [2], high magnetic field [3], or both [4], $Tb_2Ti_2O_7$ does order magnetically. The possibility that this order stems from magneto-elastic coupling was considered in all cases [2–4]. This type of coupling allows the lattice to distort in order to relieve the magnetic frustration, concomitantly lowering the total system energy. Thus the magneto-elastic coupling, which is a small perturbation to the spin Hamiltonian, can select one ground state out of the macroscopically degenerate ground states [5, 6]. In the most spectacular case the magneto-elastic coupling leads to long-range spin order accompanied by a new lattice structure such as in $ZnCr_2O_4$ [7] and $CdCr_2O_4$ [8]. An alternative and less dramatic possibility is a selection of a ground state with short-range spin order and short-range lattice deformation. In the latter case the original lattice structure is preserved on the average. This might be the situation in $Y_2Mo_2O_7$ where evidence for lattice deformations were found by several methods [6, 9, 10].

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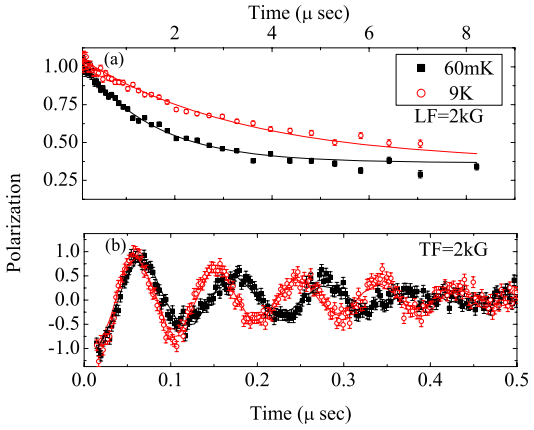

Figure 1. The time dependence of the muon polarization in the (a) longitudinal and (b) transverse directions at an applied field of 2 kG and two different temperatures. The timescale is different between the two directions. The solid lines are fits to equations (1) and (2).

In this work we study the possible existence of magneto-elastic coupling in $Tb_2Ti_2O_7$ using the muon spin resonance (μSR) technique. The basic idea is to investigate the nature of the changes in the local environment of the muon as the temperature decreases. We determine whether only the spin polarization is changing or whether the lattice is involved as well. Electronic spin polarization contributes to the shift of the muon spin rotation frequency. Lattice distortions are responsible for muon spin polarization relaxation. Comparing these two quantities provides information on the presence or absence of magneto-elastic coupling.

Transverse (TF) and longitudinal field (LF) μSR measurements were performed with powder samples on the GPS and LTF spectrometers at Paul Scherrer Institute, Switzerland. The measurements were carried out with the muon spin tilted by 50° relative to the direction of the applied magnetic fields, and positron data were accumulated in both the forward–backward (longitudinal) and the up–down (transverse) directions simultaneously. This allowed us to determine both transverse and longitudinal muon spin relaxations rates. In figure 1 we show the LF (panel (a)) and the TF (panel (b)) data at two temperatures and applied field of 2 kG. The TF data are shown in a reference frame rotating at a field of 1.5 kG. Several aspects can be seen in the raw data. From the timescale it is clear that the transverse relaxation is greater by far than the longitudinal one. The longitudinal relaxation increases as the temperature decreases, as was observed previously [11]. Finally, the transverse relaxation increases and the muon rotation frequency decreases upon cooling.

The μSR LF polarization is best described by the root exponential

$$P_{\mathrm{LF}}(t) = A_{\mathrm{LF}} \exp(-(t/T_1)^{\frac{1}{2}}) + B_{\mathrm{LF}} \tag{1}$$

where the parameter A_{LF} is set by taking into account the tilt of the muon spin relative to the longitudinal magnetic field, T_1 is the longitudinal relaxation time, B_{LF} is the background, and t is time. Similarly, the TF polarization is best fitted by a root exponential superimposed on a cosine oscillation

$$P_{\mathrm{TF}}(t) = A_{\mathrm{TF}} \exp(-(t/T_2)^{\frac{1}{2}}) \cos(\omega t + \phi) + B_{\mathrm{TF}}. \tag{2}$$

Here T_2 is the transverse relaxation time. The other parameters have the same meaning as in equation (1). The quality of the fits is presented by the solid lines in figure 1.

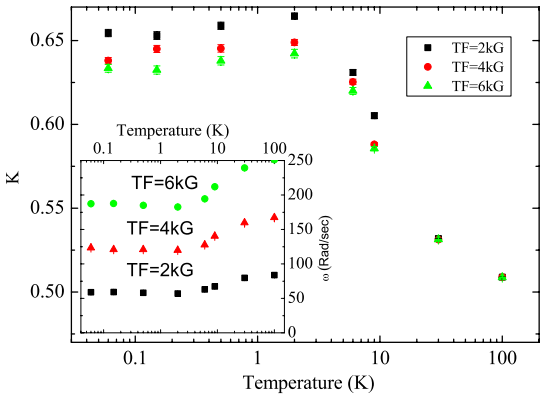

Figure 2. Inset: the muon rotation frequency as a function of temperature for different applied fields. Main figure: the shift in the rotation frequency defined in equation (3).

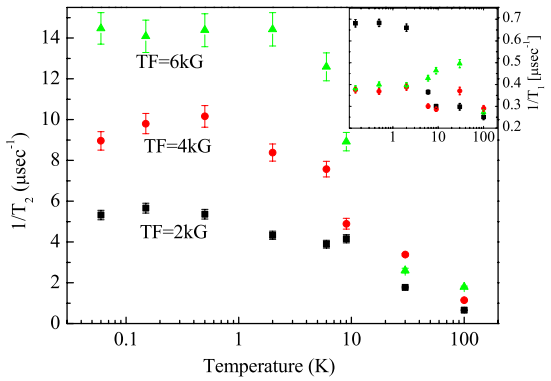

Figure 3. The muon spin transverse relaxation rate T_2^{-1} as a function of temperature for three different fields. Inset: the muon spin longitudinal relaxation rates T_1^{-1} at the same temperatures and fields.

Data were collected in the temperature range 60 mK to 100 K and in three fields, of 2, 4, and 6 kG. The frequency ω as a function of temperature for the three different fields is depicted in the inset of figure 2. The frequency shift,

$$K = (\omega_0 - \omega)/\omega_0, \tag{3}$$

is shown in the main panel for the same temperatures and fields. Similarly, we present T_2^{-1} in figure 3, and in the inset T_1^{-1}. The most important aspect of the data is that all quantities saturate as the temperature decreases below $\sim$2 K.

The shift and LF relaxation have a simple interpretation. K is a result of the sample magnetization, and T_1^{-1} stems from dynamic field fluctuations. The relaxation T_2^{-1} is a bit more involved. It is a result of both static field inhomogeneities on the timescale of one muon spin turn, and dynamically fluctuating fields. However, since T_1^{-1} is an order of magnitude lower than T_2^{-1}, the contribution to the TF relaxation from dynamic fluctuations is negligible. Therefore, T_2^{-1} could be analysed in terms of field inhomogeneities only.

T_2^{-1} is related to lattice deformation via the muon coupling to its neighbouring spins and the system's susceptibility. To demonstrate this relation let us assume for simplicity that the muon is coupled only to one electronic spin **S** (extension to multiple couplings is trivial). In this

case the magnetic field experienced by the muon is a sum of the external field **H** and the field from the neighbouring electron $\mathbf{H}_{\rm int} = g\mu_{\rm B}\mathbf{AS}$, where g is the spectroscopic splitting factor, and $\mu_{\rm B}$ is the Bohr magneton. Here $\mathbf{A} = \mathbf{A}(\mathbf{r})$ is the coupling between the muon and electron spin, which we assume depends on the distance between them, although at present the origin of this coupling is unknown. In a mean field approximation, $\mathbf{S} \rightarrow \langle\mathbf{S}\rangle = \chi\mathbf{H}/g\mu_{\rm B}$. Therefore, the muon experiences a magnetic field $\mathbf{B} = (1 + \mathbf{A}\chi)\mathbf{H}$. Assuming that the susceptibility and the couplings are isotropic, the time evolution of a muon spin due to static field inhomogeneity is given by

$$P_{\rm TF}(t) = \int \cos[\gamma_\mu(1 + \{\langle A\rangle + \delta A\}\chi)Ht]\rho(\delta A)\,{\rm d}\delta A \tag{4}$$

where $\langle A\rangle$ is the mean coupling, $\gamma_\mu = 85.162$ Mrad T^{-1} s^{-1} is the muon gyromagnetic ratio and ρ is the distribution of coupling variations. From equations (2) and (4) we find, using inverse Fourier transform, that:

(I) The frequency shift is given by

$$K = \langle A\rangle\chi. \tag{5}$$

(II) The distribution ρ can be expressed as

$$\rho(\delta A) = f(\sigma/\{2\pi|\delta A|\})/(2|\delta A|) \tag{6}$$

where the effective width of the distribution, σ, is given by

$$\sigma = 1/\left|T_2\chi\gamma_\mu H\right|, \tag{7}$$

$$f(x) = \sqrt{x}\left[\sin(\pi x/2)\left\{1 - 2FS(\sqrt{x})\right\} + \cos(\pi x/2)\left\{1 - 2FC(\sqrt{x})\right\}\right] \tag{8}$$

and FS and FC are the Fresnel functions [12]. Thus, according to equation (7), if the ratio between T_2^{-1} and K remains constant as the temperature decreases, the most likely conclusion is that σ is temperature independent, and no lattice deformations are present. This conclusion would be wrong only if by accident $\langle A\rangle/(T_2K)$ turns out to be temperature independent while $(T_2K)^{-1}$ does vary with T. To eliminate this possibility χ must be measured directly, as discussed below. It should be pointed out that the same calculation could be done by assuming that χ is distributed instead of A. In this case a proportionality between T_2^{-1} and K would mean a temperature independent distribution of χ throughout the lattice. This would equally role out lattice distortion upon cooling.

In figure 4 we show $(T_2\gamma_\mu H)^{-1}$ versus K with the temperature as an implicit parameter. The two quantities are linearly dependent at all fields. Allowing for a base line shift, which does not originate in localized spins, will lead to a proportionality relation between these quantities. This stands in strong contrast to $Y_2Mo_2O_7$, where the muon transverse relaxation grows as a function of χ faster than exponentially. This is demonstrated in the inset of figure 4 using data from [10] on a semi-log scale. We conclude that in $Tb_2Ti_2O_7$ the muon transverse relaxation has the same temperature dependence as the shift. In fact, by calculating σ for each data point using equation (7) we find that $\Delta\sigma/\overline{\sigma}$ is 15% for $Tb_2Ti_2O_7$ and 115% for $Y_2Mo_2O_7$, where $\overline{\sigma}$ and $\Delta\sigma$ are the average and standard deviation of σ respectively. $\Delta\sigma/\overline{\sigma}$ is a measure of the relative change in the distance variations due to temperature changes.

We also performed DC susceptibility measurements using a cryogenic SQUID magnetometer, and home-built Faraday balance [13] in a dilution refrigerator (DR). In the Faraday balance the powder sample is attached to a metallic sample holder, which serves as one of the plates of a capacitor, and is suspended on a spring. The sample experience both a field H and a field gradient of 1 T m^{-1}. The capacitance C is measured as a function of H and $\chi \propto {\rm d}C^{-1}/{\rm d}H$.

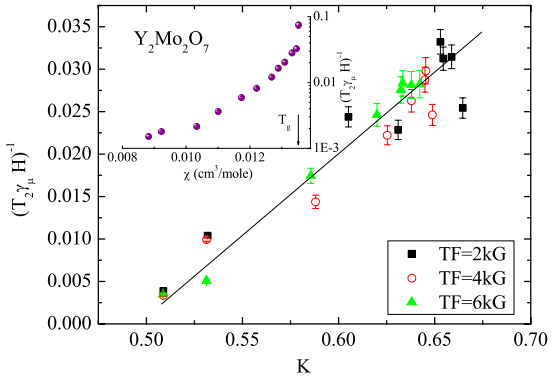

Figure 4. Demonstrating the linear relation between the muon spin relaxation rate normalized by the field $(T_2\gamma_\mu H)^{-1}$ versus the shift in muon spin rotation frequency K, which is proportional to the susceptibility χ. The inset shows the same type of measurement on a semi-log scale in the pyrochlore compound $Y_2Mo_2O_7$ taken from [10].

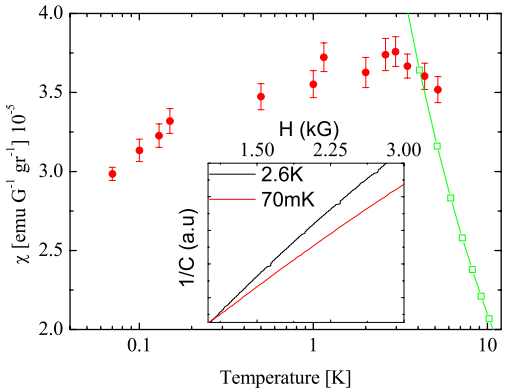

Figure 5. The DC susceptibility as measured by SQUID, and Faraday balance mounted in a dilution refrigerator. The two measurements do not overlap, probably due to the difficulty of cooling the large amount of powder in the balance. Nevertheless a decrease in χ is observed as the temperature decreases. The inset shows raw inverse capacitance versus applied field. The slope is proportional to χ.

In the inset of figure 5 we show raw data of C^{-1} versus H at 70 mK and 2.6 K. At all fields the slope at 70 mK is smaller than at 2.6 K. This means that at some temperature, in between these two, the susceptibility starts to decrease. In figure 5 we show the temperature-dependent susceptibility obtained by both experimental methods. The two measurements do not overlap in the 2–5 K region. We attribute this problem to difficulties in cooling of the entire volume of the sample in the DR. In other words, the mean temperature of the sample T_s is higher than the DR temperature T_{DR}. Nevertheless, as T_{DR} decreases, and so does T_s, the susceptibility does not increase. In fact, below $T_{DR} \sim 0.2$ K, the susceptibility decreases. A saturated susceptibility and the decrease below ∼0.2 K is in rough agreement with previous measurements of Luo *et al* [14]. This result shows qualitatively that T_2^{-1} and χ do not vary substantially with T below 2 K, which supports our previous conclusions.

To summarize, we compare the transverse relaxation rate resulting from internal field distribution to susceptibility measured by the shift in the muon rotation frequency and DC

susceptibility. We find that the relaxation rate has the same temperature dependence as the susceptibility. This indicates that the only reason for increasing relaxation upon cooling is an increase in the electronic moment size. Therefore, there is no evidence for lattice deformation in $Tb_2Ti_2O_7$ that is static on the timescale of 0.1 μs.

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