Studies of $\text{R}_2\text{Ti}_2\text{O}_7$ ($\text{R}=\text{Gd}$ and $\text{Yb}$); new results

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Abstract

Specific heat and muon spin rotation and relaxation data are presented for two geometrically frustrated systems: Gd$_2$Ti$_2$O$_7$ which antiferromagnetically orders and Yb$_2$Ti$_2$O$_7$ which presents dynamical short range correlations at low temperature. The muon data help to characterize the spin dynamics of these two compounds.

Key words: Magnetism, frustration, pyrochlore.

Geometrically derived magnetic frustration arises when the spatial arrangement of the spins is such that it prevents the simultaneous minimization of all interaction energies [1]. Compounds of interest for the investigation of frustration are, for example, the crystallographically ordered pyrochlore structure compounds $\text{R}_2\text{Ti}_2\text{O}_7$ where the rare earth ions (R) form a sub-lattice of corner sharing tetrahedra.

We shall focus on two pyrochlore compounds Gd$_2$Ti$_2$O$_7$ and Yb$_2$Ti$_2$O$_7$. Reports of experimental data concerning these two systems were already published by some of us, see Refs. [2,3,4,5]. A summary of the muon spin rotation
and relaxation (μSR) data has been recently provided [6]. Here we report unpublished data obtained from specific heat and μSR measurements. The latter measurements were either performed at the ISIS μSR facility in UK or at the Swiss Muon Source of the Paul Scherrer Institute in Switzerland. We stress that the specific heat measurements are essential to test the sample quality in geometrically frustrated systems. For example, in the case of Gd$_2$Ti$_2$O$_7$, the heat treatment affects the specific heat data in two ways. First it notably increases the intensity of the signal and second it slightly decreases the temperature of the two peaks corresponding to the magnetic transitions. As far as μSR spectra are concerned, the values for the measured local fields are noticeably changed by the heat treatment.

In a first step we consider Gd$_2$Ti$_2$O$_7$. Figure 1 shows the specific heat measurements and the variation of entropy which is deduced from these data. The entropy variation up to \( \simeq 5 \) K is only 90% of its expected value for a spin 7/2 system. This missing entropy is probably related to the persistent spin dynamics observed in this system deep in the ordered state [2]. The specific heat structure observed at \( T_{c1} \simeq 1.02 \) K for Gd$_2$Ti$_2$O$_7$ corresponds to a long-range magnetic ordering of the Gd magnetic moments as shown by the spontaneous muon spin precession detected below \( T_{c1} \) in zero-field. In fact, we observe two Bessel-like oscillations, each characterized by the maximum field at the muon site, \( B_{\text{max}} \); see Fig. 2. Note that the transition at \( T_{c2} \simeq 0.74 \) K is hardly visible in the temperature dependence of \( B_{\text{max}} \). This is not surprising since the difference between the magnetic structures adopted by Gd$_2$Ti$_2$O$_7$ above and below \( T_{c2} \) is small: the change concerns only a quarter of the Gd$^{3+}$ moments which are disordered above \( T_{c2} \) and very weakly ordered below [7].
Fig. 2. Temperature dependence of $B_{\text{max}}$ measured for Gd$_2$Ti$_2$O$_7$ (two fields per temperature point can be resolved up to 0.6 K and only one field at higher temperatures) and the associated exponential damping rate $\lambda_X$ for the higher $B_{\text{max}}$ value. The damping rate is negligible for the smaller $B_{\text{max}}$. The line is the result of a fit for the larger field with $B_{\text{max}}(T) \propto [1 - (T/T_{c1})^\alpha]^\beta$, setting $\alpha = 2$ and $\beta = 0.365$ as expected for a 3D Heisenberg antiferromagnet characterized by a second order phase transition at $T = T_{c1}$. The fit is numerically reasonable and physically justified since $B_{\text{max}}$ is expected to track the order parameter. The contact field term, which might have a temperature dependence different from that of the order parameter, should indeed be negligible for an insulating compound such as Gd$_2$Ti$_2$O$_7$.

Although the thermal behavior of the spin-lattice relaxation, $\lambda_Z$, at high temperature is not directly related to the frustrated nature of the compounds of interest, it is still of interest. We present $\lambda_Z(T)$ above $T_{c1}$ for Gd$_2$Ti$_2$O$_7$ in Fig. 3. When the temperature is decreased from room temperature towards $T_{c1}$, $\lambda_Z(T)$ first slightly decreases and reaches a local minimum around 10 K, before the slowing down of the spin dynamics causes a marked increase of $\lambda_Z(T)$ when approaching $T_{c1}$. The initial slight decrease is the signature of the antiferromagnetic spin correlations which build up in Gd$_2$Ti$_2$O$_7$ as the system is cooled down. It is fitted to a model in which the spin correlation function is expanded versus $(k_B T)^{-1}$ [6,3] (see line in Fig. 3).

A remarkable output of most $\mu$SR measurements for geometrically frustrated magnetic materials has been the observation of persistent spin dynamics at
Fig. 3. The spin-lattice relaxation rate $\lambda_Z$ versus temperature measured in the paramagnetic phase for crystals of Gd$_2$Ti$_2$O$_7$. The data have been recorded for two orientations of the initial muon polarization, parallel to either a [111] or [110] crystallographic axis: they show no difference. The line is the result of a fit with $\lambda_Z(T) = \lambda_{Z}^{(ss)}(1 + T_0/T)$ where $\lambda_{Z}^{(ss)} = 1.45 (37) \mu s^{-1}$ and $T_0 = -2.3 (6) \text{ K}$. The further increase of $\lambda_Z$ as the sample is cooled down below $\simeq 10 \text{ K}$ reflects the slowing down of the magnetic fluctuations when approaching $T_{c1}$ from above. This effect is not accounted for by the model.

low temperature. That dynamics has recently been shown to be the signature of a large density of states at low energy characterized by a gap linear in temperature [2]. The persistent dynamics is usually recognized by the observation of a finite value for $\lambda_Z$. This is the case for Gd$_2$Ti$_2$O$_7$ [2]. If the spin dynamics is particularly slow, $P_Z(t)$ is no longer stretched-like but Kubo-Toyabe-like. This type of relaxation function was found in Yb$_2$Ti$_2$O$_7$ [4] below the temperature of the specific heat peak shown in Fig. 4. The spin dynamics is then revealed by the finite slope of $P_Z(t)$ at large time. In order to ascertain the interpretation of the Yb$_2$Ti$_2$O$_7$ spectra, we have investigated their field dependence. As shown in Fig. 5, the relaxation is quenched by a longitudinal field. The fits presented as full lines in this figure are done using the function 

$$a_0 P_Z(t) = a_1 P_{GBG}(t) + a_2$$

where $P_{GBG}(t)$ is the Gaussian-broadened Gaussian function [8] and the second term ($a_2$) represents the background signal which mainly arises from muons stopped in the silver sample holder. A conventional Kubo-Toyabe function instead of $P_{GBG}(t)$ does not properly account for the data. This is illustrated by the dashed line in Fig. 5 which is the best fit for the spectrum recorded in a 2 mT longitudinal using the Kubo-Toyabe function: the dashed line curve differs notably from the data for $t \leq 1 \mu s$. It means that the muon spins detect a field distribution influenced by disorder. This probably results from the geometrical frustration of the magnetic interactions. We also note that a dynamical spin-glass model [9] does not either provide a fit of the early times data as good as the $P_{GBG}(t)$ function.

The model behind this Gaussian-broadened Gaussian function assumes that
the muons see different environments, each of them being characterized by a dynamical Kubo-Toyabe function associated with a field width $\Delta/\gamma_\mu$. To account for the slight differences between the environments, $\Delta$ is then assumed to be Gaussian distributed with a mean-value $\Delta_0$ and a width $w$. The common parameters for the five spectra presented in Fig. 5 are the initial asymmetries of the $\mu$SR signals related to the sample and the background, respectively $a_1 = 0.170$ and $a_2 = 0.065$, $\Delta_0 = 4.9 \mu s^{-1}$ ($\Delta_0/\gamma_\mu = 5.7 \text{ mT}$), the correlation frequency of the field at the muon $\nu = 0.85 \text{ MHz}$ and $w/\Delta_0 = 0.38$. Only the field values are changed. As expected from the important effect of modest fields in the shape of the $\mu$SR spectra, $\nu$ is found in the megahertz range. The remarkable result is that the fits work only if we take the values for the field smaller than the actual applied field. We have $B_{\text{meas}} = 2.5, 5.0, 9.2, 17.2$ and $36 \text{ mT}$ instead of $B_{\text{ext}} = 10, 20, 50, 100$ and $200 \text{ mT}$ respectively. This remarkably feature has already been encountered for a Kagome-like material [10]. It has been attributed to the intermittent nature of the spin dynamics. In our case the ratio $B_{\text{meas}}/B_{\text{ext}}$ depends on $B_{\text{ext}}$ which would imply that the applied field affects the system. We note that $\Delta_0/\gamma_\mu$ is reduced compared to the high temperature field width of $80 \text{ mT}$ [11] measured above the specific heat anomaly at $\simeq 0.25 \text{ K}$. This reduction is consistent with the sporadic nature of the field at the muon. More work is yet needed for an interpretation of the magnitude of this reduction.

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Fig. 5. Effect of a longitudinal magnetic field on the longitudinal polarization function $P_Z(t)$ recorded in Yb$_2$Ti$_2$O$_7$ at low temperature. The spectra are presented in absolute scale, i.e. they are not shifted vertically. The different lines are results of fits described in the main text.

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