Critical slowing down in the geometrically frustrated pyrochlore antiferromagnet Gd$_2$Ti$_2$O$_7$

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Abstract

Longitudinal-field muon spin relaxation experiments have been carried out in the paramagnetic state of single-crystal Gd$_2$Ti$_2$O$_7$ just above the phase transition at $T_m = 1.0$ K. At high applied fields the exponential relaxation time $T_1$ is proportional to field, whereas $T_1$ saturates below a crossover field $B_c$ that is $\sim 2.5$ T at 1.5 K and decreases as $T_m$ is approached. At low fields the relaxation rate increases markedly as the freezing temperature is approached, as expected for critical slowing down of the spin fluctuations, but the increase is suppressed by applied field. This behavior is consistent with the very long autocorrelation function cutoff time implied by the low value of $B_c$.

Key words: geometrical frustration, $\mu$SR, pyrochlores, Gd$_2$Ti$_2$O$_7$

1. Introduction

In the pyrochlore titanate Gd$_2$Ti$_2$O$_7$ the Gd$^{3+}$ local moments occupy corner-shared tetrahedral sites, leading to geometrical frustration, enormous ground-state degeneracy, and extreme sensitivity to mechanisms that break this degeneracy [1]. Phase transitions are not expected for a pyrochlore lattice of spins coupled by a Heisenberg exchange interaction [2,3]. But Gd$_2$Ti$_2$O$_7$ exhibits a zero-field magnetic phase transition at $T_m = 1.0$ K [4]; below this temperature a complex field-temperature phase diagram is found [5,6]. The transition temperature is only slightly dependent on applied field, and the data suggest a multicritical point at $\sim 1.5$ T and $\sim 0.86$ K. Similar behavior emerges from theories based on dipolar interactions between Gd$^{3+}$ moments [5,7].

Although Mössbauer data suggest a first-order transition at $T_m$ [8], the specific heat and susceptibility measurements that establish the phase diagram show no evidence for a discontinuous jump or hysteresis at the transition for any field, suggesting that all transitions are of second order. Zero-field muon spin relaxation ($\mu$SR) measurements in the
ordered state of Gd$_2$Ti$_2$O$_7$ [9] indicate that strong dynamic fluctuations persist to low temperatures.

This paper reports a μSR study of critical dynamics in the paramagnetic state ($T \gtrsim T_m$) of Gd$_2$Ti$_2$O$_7$, which was motivated by the above results and the possibility of strong low-frequency fluctuations resulting from frustration in this compound.

2. Experimental Results

Longitudinal-field μSR experiments were carried out in single-crystal Gd$_2$Ti$_2$O$_7$ at the M15 channel, TRIUMF, Vancouver, Canada. Data were taken at temperatures just above $T_m$ in fields $B$ of up to 4 T applied parallel to the (111) direction. Exponential relaxation was observed at all temperatures and fields.

Figure 1 gives the field dependence of the exponential relaxation time $T_1$ at three temperatures just above $T_m$. Above a crossover field $B_c$ that decreases from $\sim 2.5$ T at 1.5 K to $\sim 1$ T at 1.1 K $T_1$ is proportional to field (fit lines in Fig. 1). Below $B_c$ $T_1$ tends towards a constant except perhaps at 1.1 K. A linear field dependence of $T_1$ (with no crossover at low fields) was also found at low temperatures in the diluted pyrochlore Ising antiferromagnets (Tb$_p$Y$_{1-p}$)$_2$Ti$_2$O$_7$, 0.21 $\leq p \leq$ 1 [10], which do not order magnetically down to $\sim 50$ mK. Such behavior has been called “cooperative paramagnetism”, since the interactions that couple the paramagnetic spins strongly affect the spin dynamics but do not cause the spins to order. Our measurements suggest that similar cooperative paramagnetism characterizes the high-temperature phase of Gd$_2$Ti$_2$O$_7$.

In general the muon relaxation rate $1/T_1$ in a paramagnet is given by [11]

$$1/T_1 \propto (T/\omega_\mu) \sum_q |H_{\text{hf}}(q)|^2 \chi''(q, \omega_\mu),$$  \hspace{1cm} (1)

where $H_{\text{hf}}(q)$ is the spatial Fourier transform of the transferred hyperfine interaction between a muon and the paramagnetic spin system, $\chi''(q, \omega_\mu)$ is the dissipative component of the complex spin susceptibility, and $\omega_\mu = \gamma_\mu B$ is the muon Zeeman frequency, where $\gamma_\mu = 852 \, \mu s^{-1} \, T^{-1}$ is the muon gyromagnetic ratio. In the sum over $q$ of Eq. (1) $\chi''(q, \omega_\mu)$ is weighted by the hyperfine coupling factor (form factor) $|H_{\text{hf}}(q)|^2$. If $H_{\text{hf}}(q)$ depends significantly on $q$, the effective weighting changes as $\chi''(q, \omega_\mu)$ evolves from a $q$-independent form at high temperatures (appropriate to independently-fluctuating local moments) to a form with a peak at the ordering wave vector as the transition is approached. The physical picture for this behavior is that the fluctuating local field at a given muon site is the resultant of transferred hyperfine fields from a number of neighboring spins, and will therefore be modified by any change in spatial correlation between the fluctuations of these neighboring spins.

If either $|H_{\text{hf}}(q)|^2$ or $\chi''(q, \omega_\mu)$ is substantially independent of $q$, however, the temperature and field dependencies of $1/T_1$ reflect those of the local dissipative susceptibility $\chi''(\omega) = \sum_q \chi''(q, \omega)$. The $q$ dependence of $H_{\text{hf}}$ can be estimated qualitatively by comparing the paramagnetic-state hyperfine field $A_{\text{hf}}$, which gives an estimate of the effective coupling for uncorrelated spins, with the zero-field μSR frequency or frequencies observed in the magnetically ordered state, which yield the effective coupling for the ordered-state spin configuration.

In Gd$_2$Ti$_2$O$_7$ $A_{\text{hf}} = 18.9(2) \, mT/\mu_B$ from transverse-field μSR experiments at high temperatures [12]. In the magnetically ordered state, where Gd moments are oriented parallel to local (111) directions [8], two muon frequencies, 21.2(1) mT/\mu_B and 27.7(1) mT/\mu_B (assuming an ordered Gd$^{3+}$...
increases markedly as $T$ of the spin fluctuations (i.e., divergence of field dependence of magnetic susceptibility) near a magnetic transition, and is interpreted as evidence for critical slowing down near a phase transition [4,5,6], and critical slowing down would be expected at any applied field as the transition line is approached. The apparent suppression of critical slowing down is, however, consistent with the cutoff power-law divergence scenario outlined above: for $B \gtrsim 1/\gamma_B \tau_c$, the power-law behavior of $\chi''(\omega)/\omega$ near a phase transition [14], with a cutoff time $\tau_c$ that controls $1/T_1$. Critical slowing down would not be suppressed in systems where $\tau_c$ is sufficiently small, i.e., $\tau_c \ll 1/\gamma_B B$ for measurements at applied field $B$ (cf. Fig. 2).

3. Conclusions

Our results suggest that spin fluctuations above the magnetic phase transition in Gd$_2$Ti$_2$O$_7$ are associated with cooperative paramagnetic behavior similar to that found in (Tb$_p$Y$_{1-p}$)$_2$Ti$_2$O$_7$ [10], with the difference that a long-time cutoff causes saturation of the fluctuation spectrum at low frequencies (low applied fields). It is the anomalously low frequency of this crossover that suppresses the critical slowing down and permits observation of the high-field power-law field dependence. To our knowledge this behavior has not been anticipated theoretically.

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