Low temperature spin dynamics of geometrically frustrated antiferromagnets $Y_2Mo_2O_7$ and $Y_2Mo_{1.6}Ti_{0.4}O_7$ studied by muon spin relaxation

S. R. Dunsiger
Department of Physics, University of British Columbia, British Columbia V6T-1Z1, Canada

R. F. Kiefl
Department of Physics, University of British Columbia, British Columbia V6T-1Z1, Canada and Canadian Institute for Advanced Research, University of British Columbia, British Columbia V6T-1Z1, Canada

K. H. Chow
Clarendon Laboratory, Oxford University, Parks Road, Oxford OX1 3PU, United Kingdom

B. D. Gaulin
Department of Physics and Astronomy, McMaster University, Hamilton, Ontario L8S 4M1, Canada

M. J. P. Gingras
TRIUMF, 4004 Westbrook Mall, Vancouver V6T 2A3, Canada

J. E. Greedan
Department of Chemistry, McMaster University, Hamilton, Ontario L8S 4M1, Canada

A. Keren, K. Kojima, and G. M. Luke
Department of Physics, Columbia University, New York, New York 10027

W. A. MacFarlane
Department of Physics, University of British Columbia, British Columbia V6T-1Z1, Canada

N. P. Raju
Department of Chemistry, McMaster University, Hamilton, Ontario L8S 4M1, Canada

J. E. Sonier
Department of Physics, University of British Columbia, British Columbia V6T-1Z1, Canada

Y. J. Uemura and W. D. Wu
Department of Physics, Columbia University, New York, New York 10027

The spin dynamics of geometrically frustrated pyrochlore antiferromagnets $Y_2Mo_2O_7$ and $Y_2Mo_{1.6}Ti_{0.4}O_7$ have been investigated using muon spin relaxation. In $Y_2Mo_2O_7$ a dramatic slowing down of the Mo^{4+} spin fluctuations occurs as one approaches the spin freezing temperature T_F of 22 K from above. At lower temperatures the spins freeze into a disordered magnetic state similar to that found in a spin glass but with a small residual muon spin relaxation rate at low temperatures. This residual relaxation rate is larger in $Y_2Mo_{1.6}Ti_{0.4}O_7$ where $T_F=15$ K. These results suggest that there is a nonzero density of states for magnetic excitations in these systems near zero energy. © *1996 American Institute of Physics.* [S0021-8979(96)05808-4]

Recently, there has been considerable interest in the behavior of geometrically frustrated antiferromagnets in which the natural antiferromagnetic coupling between ions is uniformly frustrated by the geometry of the lattice. This is unlike the random frustration occurring in conventional disordered spin glasses. In three dimensions, the most well studied systems are pyrochlores, where magnetic ions occupy a lattice of corner sharing tetrahedra. Monte Carlo simulations[1] have shown that Heisenberg spins on a lattice of corner sharing tetrahedra have a classical ground state with a macroscopic degeneracy, since the lowest energy spin configuration requires only that $\Sigma_{i=1}^{4}\mathbf{S}_i=0$ for each tetrahedron. Villain argued the classical ground state is a cooperative paramagnet,[2] with only short range spin-spin correlations and no ordering for $T>0$ K. However, additional factors such as magnetic anisotropy, further nearest neighbor exchange and thermal fluctuations[3] can lift the degeneracy. A variety of magnetic ground states have been observed in different pyrochlores. These include a non-collinear long range order (LRO) in which the spins on each tetrahedron point away from the center and a dense spin glass where the moments are frozen with no LRO.

Bulk magnetic susceptibility measurements on the pyrochlores $Y_2Mo_2O_7$[4] and $Y_2Mo_{1.6}Ti_{0.4}O_7$ show a cusp-like behavior at T_F with strong irreversibilities below $T_F=22$ K and 15 K, respectively, typical of conventional spin glasses. Strong diffuse scattering, indicating the presence of short range correlations, has been observed in wide angle neutron scattering measurements on the isostructural compound $Tb_2Mo_2O_7$.[5,6] No LRO is observed below T_F.

We report on the results of an investigation of the low temperature magnetic properties of $Y_2Mo_2O_7$ and $Y_2Mo_{1.6}Ti_{0.4}O_7$ using muon spin relaxation. Below T_F a large static internal magnetic field develops which has a very broad distribution, such that no coherent muon spin precession is observed. At the same time, the muon spin relaxation rate $1/T_1$ decreases according to a power law with decreasing temperature. The most remarkable feature in the data is the

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presence of a residual spin relaxation rate in both samples at low temperatures, which is evidence for a nonzero density of magnetic states near zero energy. These results may be compared with μSR experiments on conventional spin glasses AuFe and CuMn.[7]

Pyrochlores crystallize with a fcc structure containing eight formula units per conventional unit cell and space group $Fd\bar{3}m$. The ions on the 16d site form a network of corner sharing tetrahedra; the 16c sites constitute an identical sublattice, displaced by (1/2,1/2,1/2). Mo^{4+} ions occupy the 16c site, while the Y^{3+} ions occupy the 16d site. The Mo^{4+} ion has a magnetic moment of $\sim 1\mu_B$, whereas Y^{3+} and Ti^{4+} are diamagnetic. The oxygen vacancy concentration, which is likely the main source of crystalline disorder in these materials, is below the limit of 1% detectable by neutron and x-ray diffraction.

The μSR measurements on pressed polycrystalline pellets were made at TRIUMF in a ^{4}He gas flow cryostat for temperatures above 2K and in an Oxford Instruments model 400 top loading dilution refrigerator (DR) for lower temperatures. In a μSR experiment the observed quantity is the time evolution of the muon spin polarization, which depends on the ensemble averaged distribution of internal magnetic fields and their temporal fluctuations. The present measurements were made in a small external field directed along the initial polarization direction (longitudinal field) to reduce sensitivity to small random static nuclear magnetic dipolar fields in the sample, sample holder and cryostat. Details on the μSR technique may be found elsewhere.[8]

Figure 1(a) shows several typical μSR spectra of $Y_2Mo_2O_7$ in a longitudinal applied field of 0.02 T. Above $T_F = 22$ K the observed spin relaxation is attributed to rapid fluctuations of the internal magnetic field due to Mo^{4+} moments in the paramagnetic phase. When $\nu \gg \Delta$ (defined below) the relaxation function [see $P_z(t)$ in Fig. 1] for each magnetically equivalent site i can be described by a single exponential $e^{-\lambda_i t}$ with a relaxation rate[8]:

$$\lambda_i = \frac{4\pi\Delta_i^2 \nu_i}{\nu_i^2 + 2\nu_L^2}, \tag{1}$$

where $\Delta_i = \gamma_\mu B_i$ is the gyromagnetic ratio of the muon ($2\pi \times 135.54$ MHz/T) times the rms internal magnetic field B_i at site i. ν_i is the fluctuation rate of the internal field, and $\nu_L = \gamma_\mu B_{\text{ext}}$ is the Larmor frequency of the muon in the external magnetic field. Note that λ_i is only weakly dependent on the applied field provided $\nu_i \gg \nu_L$; this is consistent with the absence of any observed field dependence above T_F. Just above T_F, $P_z(t)$ deviates somewhat from a single exponential [see for example T=27.5 K spectrum in Fig. 1(a)] and is better described by a stretched exponential form $e^{-(\lambda t)^\beta}$ with β near 0.4. In conventional spin glasses in the limit of rapid spin fluctuations, a square root exponential relaxation is a natural consequence of the large number of magnetically inequivalent sites arising from the broad distribution of distances to the magnetic moments. In a system of dense moments such as $Y_2Mo_2O_7$ it is still possible to have multiple sites and multi-exponential behavior, but the ratio of amplitudes for the various components and the corresponding relaxation rates are temperature independent. The data for $Y_2Mo_2O_7$ near T_F could not be fit with this latter restriction, indicating that the observed non-exponential behavior possibly originates from a distribution of correlation times τ_i associated with different finite spin clusters. Similar behavior has recently been observed in other dense spin glasses AgMn and AuFe.[9] μSR spectra for $Y_2Mo_{1.6}Ti_{0.4}O_7$ are shown in Fig. 1(b), where the diamagnetic Ti^{4+} ions are substitutional impurities on the B site, introducing random disorder into the system.

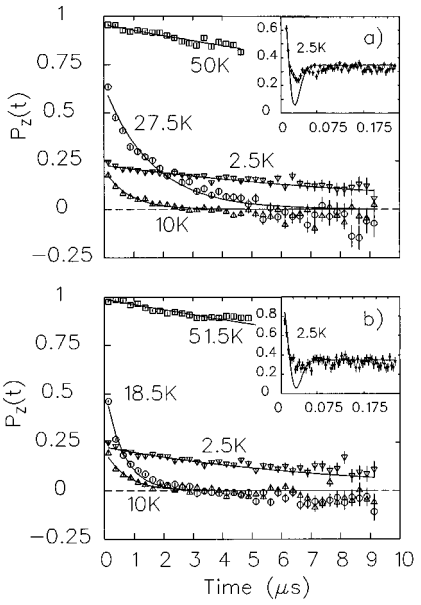

FIG. 1. Long-time dependence of the muon polarization, $P_z(t)$. Solid lines are best fits to the data. The inset shows the short-time behavior at $T = 2.5$ K for (a) $Y_2Mo_2O_7$ and (b) $Y_2Mo_{1.6}Ti_{0.4}O_7$.

The muon spin polarization function below T_F (see insets in Fig. 1) is characterized by rapid depolarization of 2/3 of the initial polarization in the first 0.05 μs, followed by slow relaxation of the remaining 1/3 component. This is a characteristic signature of a highly disordered magnetic state in which the moments are quasi-static on the timescale of the muon lifetime. For example, the muon polarization function for a single magnetic site with a Gaussian distribution of static internal fields is given by a Kubo-Toyabe function.[8] The curves in the insets of Fig. 1 show a fit of the early time data at 2.5 K to this function, modified slightly to include the small external field of 0.02 T. This does not change the form of the function appreciably provided B_{ext} is much less than B_i. Note that the 2.5 K spectrum for $Y_2Mo_2O_7$ was taken with better statistics. The best fit to the data corresponds to an average field strength $\sqrt{8/\pi}\Delta/\gamma_\mu = 0.105(5)$ T for both compounds. The dip in $P_z(t)$ at 0.032 μs is not however as deep as predicted by the Kubo-Toyabe function, indicating the distribution of internal fields is more complicated than a single Gaussian.

Muon spin relaxation results from the exchange of energy with magnetic excitations. A first order process, in which the muon absorbs or creates an excitation with an energy equal to the muon Zeeman energy, is normally suppressed in conventional long range ordered systems, since it

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requires excitations at energies less than the smallest gap. In a second order process (Raman magnon scattering) involving inelastic scattering of an excitation, application of Fermi's Golden rule gives:

$$1/T_1 \propto \int_0^\infty dE n\left(\frac{E}{k_B T}\right)\left[n\left(\frac{E}{k_B T}\right)+1\right]M^2(E)\rho^2(E), \qquad (2)$$

where the muon Zeeman energy has been neglected, $\rho(E)$ is the density of states and $M(E)$ is the matrix element for inelastic scattering of an excitation of energy E causing a muon spin flip. In a spin glass, $n(E/k_BT)$ is the probability distribution for the intravalley excitations, assumed to be Bose, within one of a macroscopic number of metastable states or "valleys." The power law behavior of $1/T_1$ is primarily determined by the energy dependence of $\rho(E)M(E)$, as intervalley transitions are thought to be important only in the mK range.[10] For example, an energy gap in $\rho(E)$ generally leads to an exponential variation of $1/T_1 \sim T\exp(-E_g/k_bT)$ at low temperatures, where T is much less than E_g/k_b. If $\rho(E)$ and $M(E)$ have power law dependences with powers l and m, respectively, then Eq. (2) implies $1/T_1$ varies as $T^{2(l+m)+1}$. The low temperature linear specific heat observed in $Y_2Mo_2O_7$ (Ref. 4) suggests $\rho(E)$ is flat or at least weakly dependent on energy.

Figure 2(a) shows the average muon spin relaxation rate in $Y_2Mo_2O_7$ obtained from fits to a single exponential relaxation function over a restricted time interval of 0.05 to 6 μs. There is a sharp rise in the average $1/T_1$ as one approaches $T_F = 22$ K from above. This is attributed to critical slowing down of the Mo^{4+} fluctuations. Below T_F, $1/T_1$ decreases gradually as the magnetic excitations freeze out. The curve in Fig. 2(a) shows the best fit of the data below 12 K to a simple power law form, $\lambda = \lambda_0 + AT^n$, with exponent $n = 2.1(3)$. A similar analysis of the $Y_2Mo_{1.6}Ti_{0.4}O_7$ data below 12 K yields a value of $n = 2.03(6)$, as illustrated in Fig. 2(b). The spin freezing temperature, as seen by a peak in $1/T_1$, occurs at $\sim$15 K in this sample. The muon spin depolarization rate is roughly temperature independent below 1 K, but at a higher value of $\sim 0.05\ \mu s^{-1}$, as compared to $0.02\ \mu s^{-1}$ in $Y_2Mo_2O_7$. The small but finite residual relaxation at the lowest temperatures implies there is an appreciable density of excitations close to zero energy. We suggest that the mechanism giving rise to temperature independent muon spin depolarization is enhanced by the addition of random disorder. However, the power law behavior is unaffected within the accuracy of the measurements.

Computer simulations by Ching *et al.*[11] on insulating Heisenberg spin glasses $Eu_xSr_{1-x}S$ ($x = 0.54$ and 0.40) have indicated the density of states $\rho(\epsilon)$ may be peaked at low energies. The geometrically frustrated kagomé lattice system $SrCr_8Ga_4O_{19}$ has also recently been studied using μSR. Dynamic spin fluctuations are observed without static freezing, even at 100 mK, well below $T_F = 3.5$ K.[12]

In summary the data for $Y_2Mo_2O_7$ and $Y_2Mo_{1.6}Ti_{0.4}O_7$ are consistent with spin glass behavior as shown by:

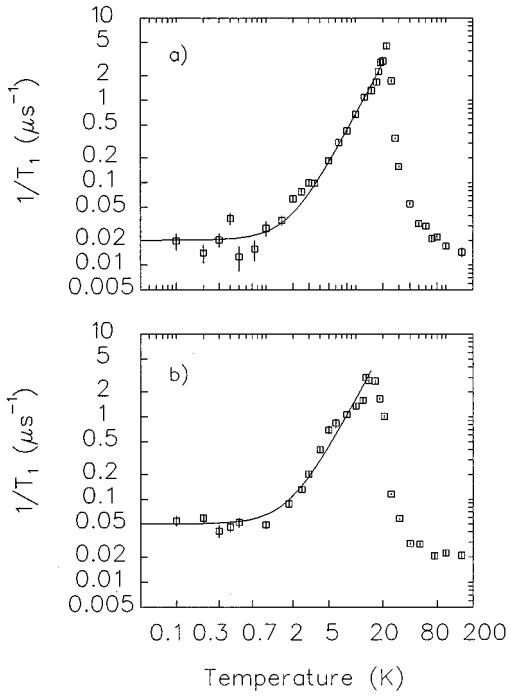

FIG. 2. Dynamical relaxation rate $1/T_1$ vs temperature for (a) $Y_2Mo_2O_7$ and (b) $Y_2Mo_{1.6}Ti_{0.4}O_7$in a longitudinally applied field of 0.02 T. The solid line is the best fit to the data assuming a power law functional form.

(1) the critical slowing down of the spin fluctuations and nonexponential muon spin relaxation near T_F,
(2) a broad distribution of static internal fields below T_F and
(3) the weak power law behavior in $1/T_1$ below T_F.

In both systems there appears to be a residual relaxation which persists down to very low temperatures. This indicates there is a nonzero density of states for magnetic excitations close to zero energy.

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