MUON SPIN RELAXATION MEASUREMENTS IN KAGOMÉ LATTICE SYSTEM $SrCr_8Ga_4O_{19}$

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Dynamical spin fluctuations in $SrCr_{8-x}Ga_{4+x}O_{19}$, a frustrated spin system on a kagomé lattice, is examined by the longitudinal field muon spin relaxation technique. This system shows a spin-glass (SG)-like cusp in the susceptibility at $T_g = 3.5(2)$ K. The slowing down of Cr spin fluctuations is found to occur over a very wide temperature range $T_g < T < 30T_g$. As $T/T_g \to 0$ these fluctuations remain without static polarization (order parameter). Such strong fluctuations below T_g have not been observed before in a conventional SG system.

Recently, the layered oxide $SrCr_{8-x}Ga_{4+x}O_{19}$ (SCGO(x)) has been examined experimentally by several groups, as a physical realization of antiferromagnetic (AFM) interaction between localized moments (Cr^{3+}, $S = \frac{3}{2}$) on the geometrically frustrated kagomé lattice. Various measurements show that this system behaves like a spin glass (SG). However, some emerging features of the data, suggest that SCGO(x) is fundamentally different from a typical SG. For example, the ratio of Curie-Weiss to Néel temperatures is as high as 150, the heat capacity below T_g has an unusual temperature dependence ($C(T) \propto T^2$) [1], and the inelastic spectral weight of neutron scattering is larger than the elastic portion, below T_g [2]. The promise for new physics comes also from theoretical arguments [3]. The classical ground state is highly degenerate [4], and this degeneracy might be responsible for the strong dynamical fluctuations below T_g.

In view of this unusual phenomenon it is essential to probe the dynamical spin fluctuations of this system. Since the energy resolution used in the neutron measurement (50 GHz) limits the sensitivity to fast fluctuations, a different method is needed to study the slower part of the spectrum. μSR is free from the resolution problem; the limitation on the time window comes only from the μ^+ lifetime (2.2 μs^{-1}), and the available external field. In this technique a wide range of spin fluctuations (0.1 to 200 MHz), and the possible existence of static effects can be detected. This paper reports a μSR experiment in geometrically frustrated SG, and demonstrates clearly the difference between SCGO(x) and typical SG systems. Our data shows that the slowing down of spin fluctuations occurs in a very wide temperature range $T_g < T < 30T_g$, and that dynamic fluctuations remain with significant spectral weight even at $T = 0.01T_g$.

In a muon spin relaxation/rotation (μSR) experiment, a beam of 100 % polarized muons is stopped, one by one, in a specimen. The muon comes to rest within 10^{-10} sec, in a crystallographic site, and its spin evolves in the local field until the μ^+ decays. The decay positron is emitted preferentially along the spin direction, and by detecting more than 10^6 positrons, the time dependence of the muon polarization can be reconstructed. The μSR experiment can be done in several geometries: In the longitudinal field (LF) configuration, both an externally applied magnetic field H_L, and the initial muon polarization $P_z(0)$ are pointing along the beam direction $\hat{z}$. Due to field inhomogeneities, and non equilibrium population of the muon spin, $P_z(t)$ is expected to relax with a temperature dependent timescale. In the transverse field (TF) μSR configuration, the external magnetic field H_T is applied perpendicular to the muon polarization. As a result, the muon spin precesses in the local field H_{loc} with frequency $\nu_\mu = (\gamma_\mu/2\pi)H_{loc}$.

The muon polarization $P_z(t) = G_z^G(t, \Delta, \nu, H_L)$ in a SG environment is described in detail in Ref. [5]. In brief, three parameters determine the time dependent of the polarization function: the width of the instantaneous field distribution $\Delta^2 = 1/3\gamma_\mu^2\langle B^2\rangle$, which is approximated by a gaussian for concentrated moments, the fluctuation frequency ν of this field, and the external longitudinal field H_L. Two extreme cases, static and fast dynamic, are of special importance. In the static limit $\nu \ll \Delta_s$, the muon spin depolarizes on a time scale of $1/\Delta_s$ due to spatial inhomogeneities of the magnetic field. As $H_L = \omega_L/\gamma_\mu$ increases from 0, the relaxation rate of $P_z(t)$ decreases, and no longer depends on the applied field when $\omega_L \geq 5\Delta_s$. At such ω_L, the internal static field is completely decoupled, and $G^G(t, \Delta, 0, H_L) = 1$ within experimental resolution. In the fast dynamic limit ($\nu/\Delta_d > 10$), the polarization is given by $\exp(-t/T_1)$ where

$$1/T_1 = 2\Delta_d^2\nu/(\nu^2 + \omega_L^2). \tag{1}$$

In this case, the relaxation rate $1/T_1$ will be insensitive to the external field, as long as $\omega_L^2 \ll \nu^2$. When static and fast dynamic fields coexist, $P_z(t)$ is given by $P_z(t) = G^G(t, \Delta_s, 0, H_L, t) \exp(-t/T_1)$. The polarization function in this situation depends on the external field only if $1/\Delta < 2T_1$, since otherwise the dynamic part relaxes completely before the static part changes significantly. Longitudinal fields make the static part only flatter; therefore when $1/\Delta > 2T_1$, $P_z(t)$ will show no field dependence. Thus, the continuous application of external field can limit the possible values of the static local field and the fluctuation rate.

Our powder sample was prepared by heating a stoichiometric mixture of $SrCO_3$, Cr_2O_3 and Ga_2O_3 with 99.9% purity at 1350°C in air for 24hr. The glass temperature $T_g = 3.5(2)$ K in our sample was determined by magnetization measurement, and the concentration of Cr ions, $x = 0.15(25)$ was inferred from the x dependence of T_g as described in Ref. [6]. The μSR experiment was performed at TRIUMF using a gas-flow cryostat at high temperatures ($T \geq 3$ K), and in an Oxford dilution refrigerator at low temperatures ($0.04 < T < 7.0$ K).

The polarization function at 3 K, and in several longitudinal magnetic fields is shown in Fig 1b. Although some field dependence for $0 < H_L <$ 100 G was seen at long t, the spectra is clearly insensitive to external field higher than 100 G. Since there is no 1/3 component recovery in zero field and no decoupling in longitudinal fields [5], the relaxation at $T = 3$ K is attributed to fast fluctuating electronic moments. The field dependence between 0 and 100 G is due to either small static fields from the Ga nuclei and/or muons that stop outside of the sample.

The polarization function at $H_L = 100$ G and several temperatures is shown in Fig 1a. The relaxation clearly increases as the system is cooled down from 100 K to temperatures below the SG transition. We fit the data with $P_z(t) = \exp(-t/T_1)$ and plot $1/T_1$ as a function of temperature in Fig. 2. Between 5 K and 100 K the relaxation rate changes by three orders of magnitudes in time scale; the slowing down of spin fluctuations occurs in a very wide temperature range, and $1/T_1$ saturates below 5 K on the logarithmic scale (but increases slowly on a linear scale). The saturation temperature is somewhat higher than T_g observed in the susceptibility measurement. These two phenomenon, the range of slowing down and the saturation of $1/T_1$, are in contrast to a typical SG behavior. For example, in *Cu*Mn no relaxation is observed when $T > 3T_g$, and $1/T_1$ drops sharply below T_g [5]. In *Cu*Mn(5 at.%) the relaxation rate reaches $\sim 1\%$ of its maximum value when the temperature is decreased to $\sim 25\%$ of T_g.

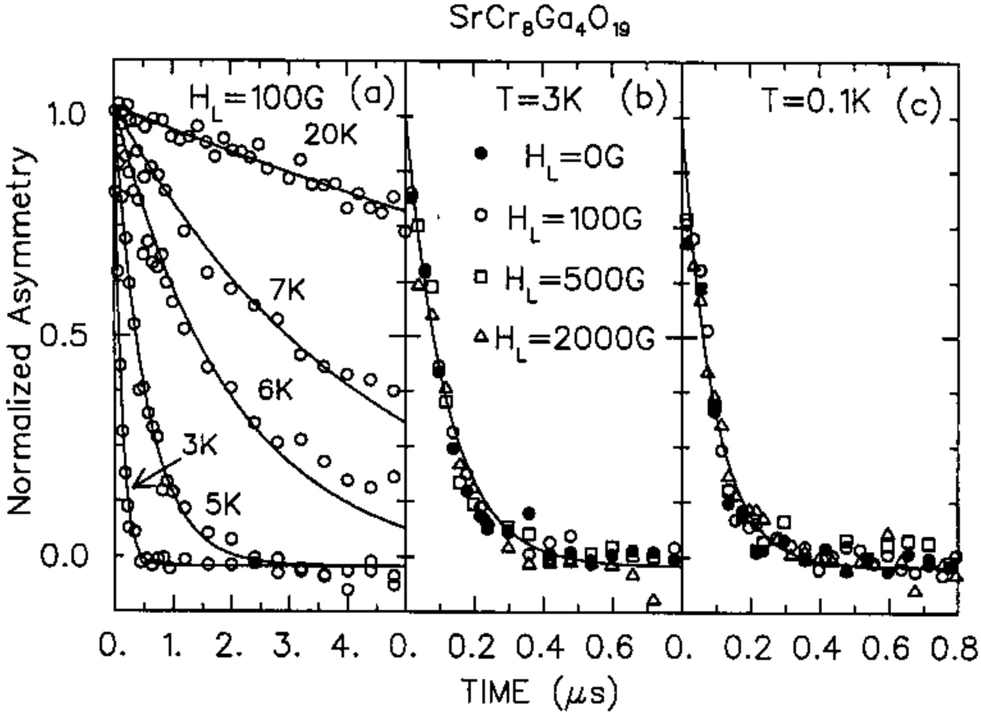

Fig. 1. Longitudinal field μSR data in $SrCr_8Ga_4O_{19}$.

The temperature dependence of the relaxation rate above the transition temperature is fitted with two models: critical exponent and activated dynamics. The fits are shown in Fig. 2. The critical exponent fit (broken line) is done using the equation $T_1^{(0)}/T_1 = [T/(T - T_c)]^\nu$, and the values $T_c = 4.00(5)$ K and $\nu = 2.77(1)$ were found. The activated dynamics fit (solid line) is done using $\ln(T_1^{(0)}/T_1) = [E_a/(T - T_c)]^{1+\psi\nu}$. This model was proposed on theoretical grounds [7] for a two dimensional SG, and was observed in $Rb_2Cu_{1-x}Co_xF_4$ [8]. In a dynamically activated process the barrier height B scales with correlation length as $B \propto \xi^\psi$, and ξ scales with temperature as $\xi \propto (T - T_c)^{-\nu}$. In this type of process, the logarithm of the relaxation rate follows a power law. The best fit when T_c was free to change gave: $T_c = 0.007(5)$ K, $\psi\nu = 0.68(1)$ and $E_a = 11.34(1)$ K. Keeping $T_c = 0$ in the fit gave $\psi\nu = 0.68(1)$ and $E_a = 11.4(1)$ K. It should be noted, however, that the activation formula gives a good fit only above $T \sim 5$ K.

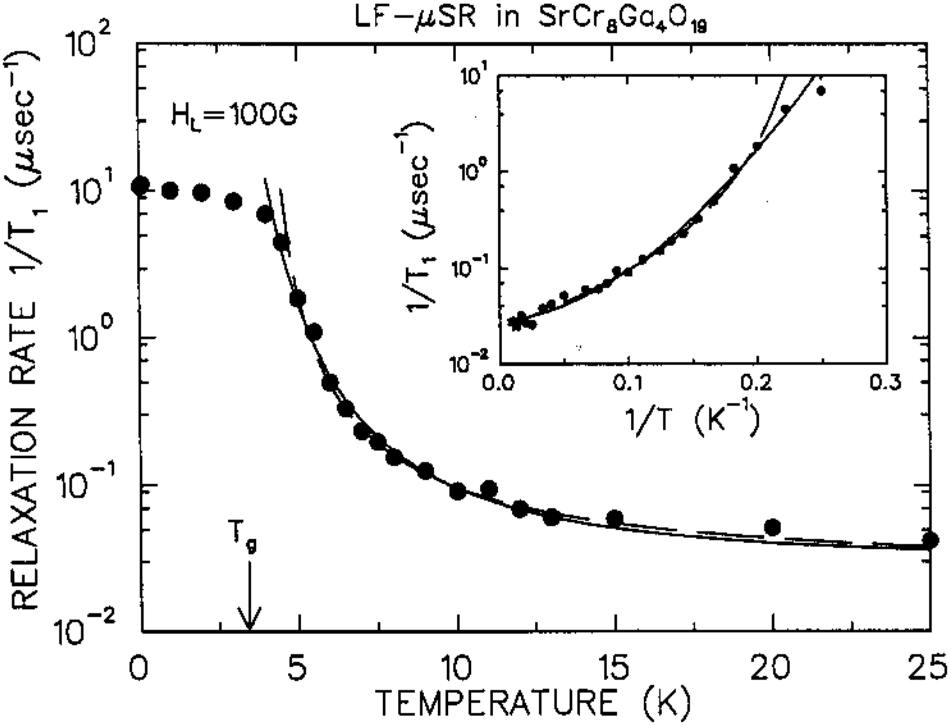

Fig. 2. Relaxation rate $1/T_1$ as a function of temperature in $SrCr_8Ga_4O_{19}$.

The application of several longitudinal magnetic fields, including zero field (ZF), at temperatures well below the SG transition (0.1 K) is demonstrated in Fig. 1c. The spectra show no dependence on the externally applied field and the relaxation rate is on the order of 10 μsec^{-1}. If the relaxation in ZF was due to static inhomogeneities, generated by the frozen part of the Cr electronic moments as well as the nuclear spin, Δ would have been $\sim$ 200 G. In this case the polarization would have shown strong dependence on longitudinal fields in the range of 0 to 2 kG. The relaxation must therefore be due to some combination of static and dynamic fluctuating fields. According to the discussion following Eq.1, a lower limit on the fluctuation rate $\nu > 170$ MHz (ω_L of 2 kG), and an upper limit on the static field width $\Delta < 5$ μsec^{-1} ($\langle B^2 \rangle^{\frac{1}{2}} < 100$ G) can be obtained.

The small magnitude of the static field could *a priori* be an accidental consequence of the muon site. We studied this possibility by transverse field TF-μSR experiment, and by numerical simulations. In the TF-μSR configuration, we measured the muon rotation frequency as a function of temperature $\nu_\mu(T)$ and use the linear relation $\Delta\nu_\mu(T) = g\Delta M(T)$ to evaluate g; $M(T)$ is the sample magnetization measured at the same external field as the muon precision frequency. By the same linear relation we then estimate the muon frequency shift, and the local field, for fully polarized spins. This method predicts that fully polarized spins will produce an internal local field of 12 kG. In the numerical simulation, the orientation of the Cr moments was allowed to vary randomly. The local field at a site was calculated by summing the dipolar field from all moments in a sphere of radius 10 Å. We found $\langle B^2\rangle^{1/2} > 3$ kG in all sites. These two methods show that the order of magnitude of the internal field is 10 kG. We therefore conclude that the small static field below T_g is not due to the muon site, but rather is an indication of fluctuating moments.

The time scale of the fluctuation rate ν can be evaluated by Eq. 1 and the estimated value of the instantaneous local field. Taking $\omega_L = 0$, and $1/T_1 = 10.8(4)\ \mu\text{sec}^{-1}$ we find that $\nu \sim 50$ GHz at $T \to 0$. This is compatible with the lower limit on the fluctuation rate $\nu \gg 170$ MHz. It should be noted that such slow fluctuations would be considered static by the neutron scattering experiment due to their energy resolution. It is very surprising that the full-amplitude spin fluctuations still exist and account for most of the muon polarization relaxation even at 40 mK (0.1% of T_g).

References

[1] A. P. Ramirez, G. P. Espinosa and A. S. Cooper, Phys. Rev. Lett **64**, 2070 (1990).

[2] C. Broholm, G. Aeppli, G. P. Espinosa and A. S. Cooper, Phys. Rev. Lett. **65**, 3173 (1990).

[3] J. T. Chalker, P. C. W. Holdsworth and E. F. Shender PRL **68(6)** 1992; S. Sachdev, Phys. Rev B **45**, 12377 (1992); D. Huse, A. D. Rutenberg, Phys. Rev. **B** 45, 7536 (1992).

[4] A. B. Harris, C. Kallin and J. Berlinsky, Phys. Rev. B **45**, 2899 (1992), and references theirin.

[5] Y. J. Uemura, T. Yamazaki, D. R. Harshman, M. Senba and E. J. Ansaldo, Phys. Rev B **31**, 546 (1985).

[6] B. Martínez *et al.*, Phys. Rev. B **46**, 10786 (1992).

[7] D. S. Fisher, J. Appl. Phys. **61** (8), 3672 (1987).

[8] C. Dekker, A. F. M. Arts, H. W. de Wijn, A. J. van Duyneveldt and J. A. Mydosh, Phys. Rev. Lett. **61**, 1780 (1988).