Probing the Spin-Spin Dynamical Autocorrelation Function in a Spin Glass above T_g via Muon Spin Relaxation

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We report zero and longitudinal field muon spin relaxation measurements in the metallic spin glass AgMn(0.5 at.%) at $T > T_g$. We find that muon polarization obeys a time-field scaling relation which allows us to distinguish between three possible forms of the spin-spin dynamical autocorrelation function: a power law, a stretched exponential, and a cutoff power law. We also discuss the evolution of the muon relaxation line shape as the temperature approaches T_g. [S0031-9007(96)00913-1]

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It is now fully recognized that there are profound similarities linking the ordering processes in all the different forms of glassy systems, and that the key to a comprehensive understanding of the glass transition lies in the dynamics [1]. In order to make meaningful and detailed comparisons, it is essential to establish precisely the empirical rules which govern the dynamics in each glass, and, in particular, in spin glasses. The most important facet of spin glass dynamics is the autocorrelation function $q(t) = \langle \mathbf{S}_i(t) \cdot \mathbf{S}_i(0) \rangle$, since cross correlations are null. Below the spin glass transition temperature T_g, there is a general consensus that $q(t)$ decays as a power law (PL), namely,

$$q(t) - q(\infty) \sim ct^{-\alpha} \tag{1}$$

after some microscopic time, on the order of 10^{-14} sec [2]. However, at temperatures above T_g, where $q(\infty) = 0$, the shape of $q(t)$ is the subject of ongoing debate. The purpose of this study is, therefore, to establish the form of $q(t)$ above T_g.

Theoretical predictions and empirical parametrization of experimental data have included both the PL [3,4] and the stretched exponential (SE) [4,5]

$$q(t) \sim c \exp[-(\lambda t)^{\beta}]. \tag{2}$$

General scaling rules near a phase transition imply a cutoff power law (CPL) [6]

$$q(t) \sim ct^{-\alpha} f(\lambda t), \tag{3}$$

with λ tending to zero as $T \to T_g$. The CPL is often approximated by the Ogielski form (OF) $q(t) \sim ct^{-\alpha} \exp[-(\lambda t)^{\beta}]$ which is observed in numerical work on Ising spin glasses [7].

The difference between these functions is fundamental, and should be emphasized. The PL is time-scale invariant, and dynamical modulations should be observed in any time window. The SE, on the other hand, has a well defined time scale given by $1/\lambda$. Finally, the CPL is time-scale invariant only at times much shorter than $1/\lambda$.

These three candidates for $q(t)$ are frequently used to analyze transport and relaxation phenomena in other disordered systems [8]. In spin glasses, however, accurate experimental data are lacking for $T > T_g$ and at times shorter than 1 μsec. For example, the neutron spin echo measurements of Mezei and Murani [9], which probe $q(t)$ directly, were successfully fitted by both a power law decay [3,10] and a stretched exponential [11]. In our experiment we use both the zero field (ZF) and longitudinal field (LF) muon spin relaxation (μSR) technique to analyze the dynamics of the canonical Heisenberg spin glass system AgMn at $T > T_g$. We find clear evidence that $q(t)$ is compatible with the OF, implying the possibility of a universal dynamic behavior for the whole class of spin glass systems.

In the LF and ZR μSR technique, one measures the asymmetry A in the spatial distribution of positrons emitted from the muon decay as a function of time t from the moment the muon enters the sample, while changing the applied longitudinal field $H \| \hat{\mathbf{z}}$. The muon spin polarization $P_z(H, t)$ is given by $A(H, t)/A_0$ [12]. This technique was used, for the first time, to map $q(t)$ in AgMn by MacLaughlin *et al.* below T_g where they found a PL [2]. However, at $T < T_g$ both static and dynamical fields contribute to the relaxation of $A(H, t)$, and the interpretation of the data is not trivial. This complication does not exist above T_g, and, as we shall demonstrate, the field variations of the relaxation rate (see Fig. 1) probe only the Fourier transform (FT) of $q(t)$. Since the field dependence of $A(H, t)$ is expected to be most pronounced at large values of t, we choose to perform our measurements at the ISIS facility in RAL, where the pulsed beam allows for the largest value of t (~20 μsec). The pulse, however, has a width of 70 nsec which distorts the first ~0.2 μsec of the spectrum. The longitudinal fields available at ISIS allow the FT of $q(t)$ to be measured from $f \equiv \gamma_\mu H/2\pi =$ 1.7 to 54.2 MHz ($\gamma_\mu/2\pi = 13.55$ MHz/kG is the muon gyromagnetic ratio). The ZF measurements provide the same information at nominally $f = 0$. However, a stray field ~1 G ($f \sim 10$ kHz) could not be ruled out. As a result of the simplified experimental conditions we are able to demonstrate that the μSR data obeys simple

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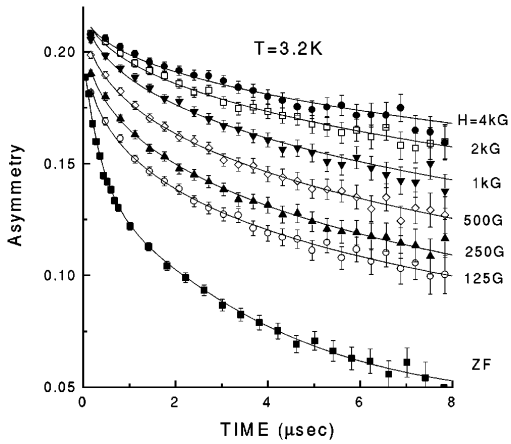

FIG. 1. Asymmetry $\propto P(H,t)$ obtained in AgMn(0.5 at. %) at $T = 3.2$ K ($T_g = 2.95$ K) in zero field and a geometric series of fields. The solid lines are guides for the eye.

scaling relations (see Fig. 2) which, as mentioned before, could be explained only by the CPL correlation function.

We choose the metallic spin glass AgMn since muons do not relax in pure silver, and the internal field from the small Ag nuclear moment can be neglected. The concentration of Mn is selected so that the dynamical phenomena we are seeking could be observed in the μSR time window. This selection has to be done carefully; if the concentration is too high, the Mn spin-spin coupling and the Mn-spin muon-spin couplings are so strong that an external field on the order of 1 kG has no effect on the polarization [e.g., AgMn(7 at. %) [13]], whereas for low concentrations the averaged internal field at the muon site could be so small that the muon does not relax within its lifetime [in the limit AgMn(0 at. %) = Ag]. In addition, we must make sure that the external field has no considerable impact on $q(t)$. We anticipate that reasonable conditions would be in AgMn(0.5 at. %), where $T_g = 2.95$ K [14]. In this concentration the electronic Zeeman energy in our highest field is still an order of magnitude smaller than a typical spin-spin coupling estimated from T_g.

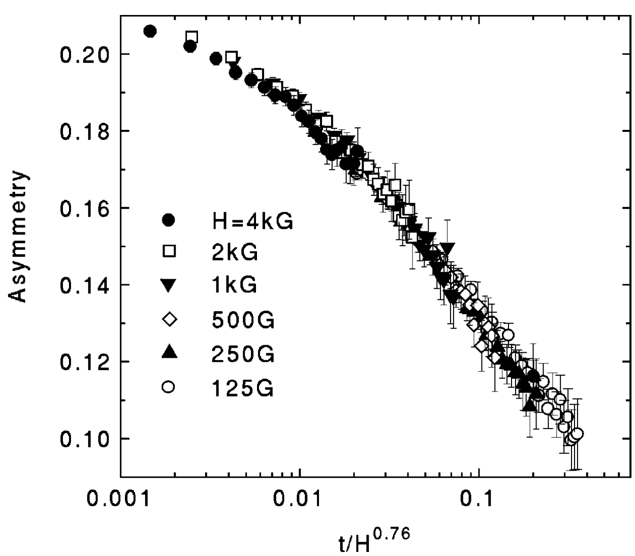

FIG. 2. The muon asymmetry in AgMn(0.5 at. %) at $T = 3.2$ K and various applied magnetic fields H.

Our analysis of the muon polarization relaxation proceeds in three steps, as outlined in the pioneering work of Uemura *et al.* [15]. First, we write an expression for the muon polarization in a given environment. A muon which experiences an internal field $\mathbf{B}(t)$, with a local time averaged second moment $\Delta^2 = \gamma_\mu^2 \langle B_i^2 \rangle$ (i is the special direction), relaxes according to

$$P_z(H,t) = \exp[-2\Delta^2 \tau(H) t], \quad (4)$$

where the correlation time τ is given by

$$\tau(H) = \frac{1}{\langle \mathbf{B}^2 \rangle} \int_0^\infty \langle \mathbf{B}(t) \cdot \mathbf{B}(0) \rangle \cos(\gamma_\mu H t) dt \equiv c\,\iota(H), \quad (5)$$

and the fast fluctuation limit ($\Delta\tau \ll 1$) is assumed. Next, we replace the field correlation function with the spin correlation function, namely,

$$\frac{\langle \mathbf{B}(t) \cdot \mathbf{B}(0) \rangle}{\langle \mathbf{B}^2 \rangle} = q(t). \quad (6)$$

This was demonstrated to be a proper procedure by Heffner and MacLaughlin [10]. Finally, the parameters Δ and τ might vary from one muon site to another, hence we should average their possible values. However, most authors assume that τ is not site dependent [3,10,15]. For reasons that will soon become clear, we use a weaker assumption where the site dependence of τ enters only through the prefactor c in the correlation functions. Thus, the sample-averaged polarization $\overline{P}_z$ is given by

$$\overline{P}_z(H,t) = \int\int \rho(\Delta, c) \exp[-\Delta^2 c\,\iota(H) t]\, dc\, d\Delta, \quad (7)$$

where $\rho(\Delta, c)$ is the probability that the muon is experiencing a given Δ, and a correlation function with a prefactor c. Equation (7) is intentionally written in a way which does not imply any specific order of averaging.

Using Eqs. (5) and (6), we can predict the field dependence of ι for the different functional forms of $q(t)$. The case where $q(t)$ decays as a PL, with $\alpha < 1$, is trivial, and the correlation time satisfies $\iota(H) \propto 1/H^{1-\alpha}$. The case where $q(t)$ decays as a SE is more complicated to analyze. However, in the asymptotic limit $\omega \equiv \gamma_\mu H \gg \lambda$, it obeys $\iota(H) \propto 1/H^{1+\beta}$ [16]. Finally, for the CPL, $\iota(H) \propto 1/H^{1-\alpha}$ asymptotically since it is indistinguishable from the PL at $t \ll 1/\lambda$. However, unlike the PL, $\iota(H)$ does not diverge at $H \to 0$ due to the cutoff. As an example, the FT of the PL, SE, and OF for $\alpha = 1/2$ and/or $\beta = 1/2$, where it can be obtained analytically, is depicted in the inset of Fig. 3. The important features

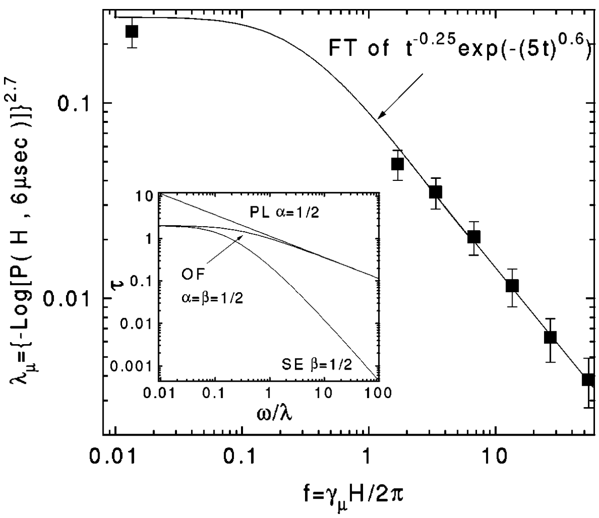

FIG. 3. λ_μ (see text) as a function of $f = \gamma_\mu H/2\pi$ on a log-log plot. The solid line is the Fourier transform of the function described in the figure. The inset shows the Fourier transform of a power law with $\alpha = 1/2$, a stretched exponential with $\beta = 1/2$, and Ogielski form with $\alpha = \beta = 1/2$.

in this figure are (I) the PL and the OF have the same asymptotic behavior which is different from the SE, and (II) the SE and OF are finite at $\omega \to 0$.

In all of the above cases, the muon polarization given by Eq. (7) asymptotically obeys the scaling relation

$$\overline{P}_z(H,t) = \overline{P}_z(t/H^\gamma), \tag{8}$$

where for the PL and the CPL $\gamma = 1 - \alpha$, and for the SE $\gamma = 1 + \beta$. As we demonstrate next, Eq. (8) describes our data at $T > T_g$ quite accurately. The value of γ would therefore distinguish between the different shapes of the correlation functions.

In Fig. 1 we present the asymmetry $A(H,t)$ at $T = 3.2$ K both in a geometrical series of fields and in ZF. The geometrical series was taken between 4 kG (the highest field available at ISIS) and 125 G by repeatedly dividing the field by 2. The values of the fields are accurate to within 1 G. A clear difference can be seen between the curves, and the higher the field the weaker the relaxation. The solid lines are guides for the eye. We demonstrate the validity of Eq. (8) in Fig. 2, where the asymmetry is shown on a semilogarithmic plot as a function of $t/H^{0.76}$. Because of the pulse width, only data points with $t > 0.2$ μsec are presented. The value of $\gamma = 0.76$ is chosen so that the muon polarizations obtained at different fields overlap. This figure demonstrates that the scaling relation of Eq. (8) is valid for over 3 orders of magnitude in t/H^γ. Since $\gamma < 1$ (the accuracy in γ is ± 0.05), we conclude that within our frequency range the spin-spin correlation function is well approximated by either a PL or the CPL [provided that $\lambda < O(1)$ MHz], with $\alpha = 0.24 \pm 0.05$. This conclusion is achieved without assuming a specific functional form for the muon polarization.

In a similar experiment at $T = 3.0$ K (not shown), we found $\alpha = 0.1 \pm 0.05$. Thus, the critical slowing down is manifested in a decrease of α as T is lowered towards T_g. The value $\alpha \simeq 0.1$ as $T \to T_g$ can be compared with critical exponents data on AgMn. Because $\alpha = (d - 2 + \eta)/2z$ [7], Lévy's ac susceptibility data [14] (taken at frequencies 10^4 times lower than the μSR frequencies) imply $\alpha = 0.13 \pm 0.02$, in excellent agreement with the present result.

However, according to Eq. (8), an instantaneous relaxation of the muon spin should occur as $H \to 0$. This is obviously not the case, as can be seen from the zero field data in Fig. 1, which relaxes within a finite time. Therefore, as suggested by Refregier, Ocio, and Bouchiat [17], there must be a crossover from a high frequency range where $\iota(H) \propto 1/H^{1-\alpha}$, to a low frequency range, where $\iota(H)$ is bounded. Indeed, the CPL does provide a crossover between the $\omega \gg \lambda$ and $\omega \ll \lambda$ regions. This could also explain why no field dependence of the muon relaxation is seen, above T_g, in samples with higher concentration of Mn; for a given value of $(T - T_g)T_g$, λ increases with increasing Mn spin-spin coupling, and therefore increases with Mn concentration. At ~10% of Mn, the frequency range accessible for the μSR shifts into the low frequency limit, where $\iota(H) =$ const.

A complementary approach for analyzing our data is to fit the asymmetries with a specific functional form. One such form, which has been very successful in fitting μSR data at high concentration of magnetic impurity, is

$$A(H,t) = A_0 \exp[-(\lambda_\mu t)^{\beta_\mu}]. \tag{9}$$

The subscript μ reflects the fact that λ_μ and β_μ are parameters of the muon relaxation function, and not of $q(t)$. In order to demonstrate Eq. (8), we must fit the data taken at different fields with a common β_μ and A_0 (and base line) and show that $\lambda_\mu(H) \propto \iota(H) \propto H^{-\gamma}$. In practice this is a rather difficult program to execute, especially for the early time data. We believe that this difficulty arises from a combination of effects: (I) the pulse structure of the beam, and (II) small changes in A_0 (and base line), as the field is altered by more than an order of magnitude, due to changes in the positrons' trajectories. While these effects spoil the fit by a specific theoretical function, they cause only a minor change in the appearance of Fig. 2. The field dependence of λ_μ could still be obtained from the late time data by plotting $\lambda_\mu = \{-\ln[P(H,t_1)]\}^{1/\beta_\mu}$ as a function of H, as shown in Fig. 3. We have used $t_1 = 6$ μsec since the error bars are reasonably small, and $\beta_\mu = 0.37$ (see Fig. 4). The nominal ZF point is placed at $H = 1$ G. Although at present our data are not sufficient to distinguish between different cutoff functions, we show for demonstration purpose the numerical FT of the OF with $\alpha = 0.25$ (obtained from Fig. 2), $\beta = 0.6$, and $\lambda = 5$ MHz.

In the inset of Fig. 4 we present the asymmetry obtained at various temperatures by cooling the sample

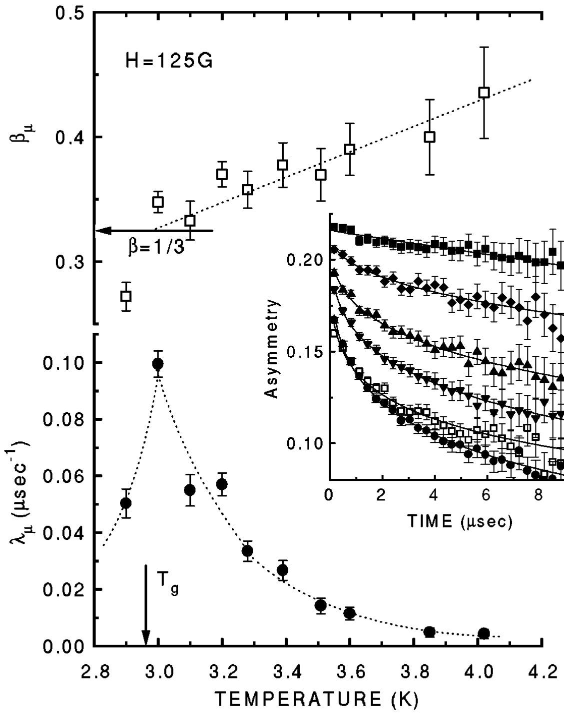

FIG. 4. The temperature dependence of the parameters λ_μ and β_μ in AgMn(0.5 at. %). The dotted lines are guides for the eye. The inset shows the asymmetry obtained at various temperatures approaching T_g. The solid lines are fits by Eq. (9).

in $H = 125$ G. When the field is constant and only the temperature is changed, we find no difficulty in fitting the data by Eq. (9). The solid lines are such fits where A_0 is determined from the 30 K data and held fixed. These fits provide a very good description of the data. A clear change in the parameters takes place between 3.0 and 2.9 K. The asymmetry at 2.9 K relaxes faster at the beginning but recovers and even exceeds the 3.0 K asymmetry towards the end of the measuring time. This recovery of the relaxation is a signature of a phase transition, and $T_g = 2.95 \pm 0.05$ K determined by μSR is in agreement with the known value of T_g [14].

The parameters λ_μ and β_μ are presented in Fig. 4 as a function of temperature. The dotted lines are guides to the eye. The relaxation rate λ_μ reaches its maximum value at $T = 3.0$ K, again indicating a phase transition. In addition, it is clear that, as the temperature is lowered towards $T_g, \beta_\mu \rightarrow 1/3$. This trend was first noticed by Campbell *et al.* in samples with relatively high concentration (>7%) of magnetic impurities [13]. Our result therefore suggests that $\beta_\mu = 1/3$ at T_g is common to a wide range of concentrations in AgMn.

It is now also clear why it is necessary to postulate the distribution of the prefactor c; if we had a unique value for c $(= c_0)$ then according to Eqs. (7) and (8) we would have $\overline{P}(H,t) = \overline{P}(c_0 t/H^\gamma)$, where $\overline{P}(t)$ is a temperature independent function. This is in contrast to the experimental observation. Therefore, the evolution of the line shape [namely, $\beta_\mu(T)$] must be controlled by the evolution of the distribution of c. A theory which relates Eq. (9) to Eq. (7) would be useful for further analysis of our data. This theory, however, must apply to both diluted and concentrated samples, as demonstrated here.

We have shown that the muon polarization obeys the scaling relation of Eq. (8) above T_g. By assuming that, up to a prefactor c, all the muons are experiencing the same correlation function, we conclude that, out of the three possibilities for $q(t)$ given in Eqs. (1)–(3), the cutoff power law [Eq. (3)] describes our data best. We also found that the muon polarization function decays as a stretched exponential with $\beta_\mu \rightarrow 1/3$ as $T \rightarrow T_g$, and that $\beta_\mu(T)$ is determined by the temperature dependence of the distribution of c.

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