# Comparison of spin-glass dynamics determined by the muon-spin-relaxation and neutron spin-echo techniques

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Impurity-spin-correlation functions obtained from muon-spin-relaxation and neutron spin-echo experiments are found to agree quantitatively above and below the "glass" temperature $T_g$. The agreement confirms the power-law decay of correlations in time near and below $T_g$. Features of the experimental results which depend on (quasi)static impurity-spin correlations suggest that the spatial distribution of thermally averaged impurity-spin magnitudes is quite uniform below $T_g$. The stochastic behavior of dipolar fields at muon sites is essentially the same as that of the impurity spins themselves at all temperatures.

## I. INTRODUCTION

The dynamic behavior of disordered spin systems possessing random exchange is a subject of considerable current interest. In simple theoretical models for crystalline (nonrandom) spin systems, such as ordered ferromagnets, it is found that the spin-autocorrelation function $S_\sigma(t)=\langle \vec{\sigma}(t)\cdot\vec{\sigma}(0)\rangle$, for the stochastic time dependence $\vec{\sigma}(t)$ of the impurity spins, decays exponentially in time (outside of the critical region).[1] In disordered systems, however, dynamic theories[2] and simulations[3] predict an algebraic (power-law) decay in time. Therefore, it is of interest to pursue experimentally the distinction between dynamics in ordered and disordered systems.

Recently, two relatively new experimental techniques, neutron—spin-echo (NSE) scattering and muon-spin relaxation ($\mu$SR), have been used to probe spin dynamics in metallic spin-glasses. These are dilute magnetic alloys (e.g., *Ag*Mn, *Cu*Mn, and *Au*Fe), which exhibit a characteristic cusp in the ac susceptibility[4] at the glass freezing temperature $T_g$. The NSE technique, introduced by Mezei and Murani,[5] measures the spin-correlation function $S_\sigma(\vec{q},t)$ directly, where $\vec{q}$ is the neutron momentum transfer. The $\mu$SR method[6] yields spin-lattice—relaxation rates of local probes (implanted positive muons), which are sensitive to fluctuations of the (dipolar) local field $\vec{h}(t)$ at muon sites. This yields a broad average over all $\vec{q}$, as in nuclear magnetic resonance (NMR). The time dependence of the local-field correlation function $S_h(\vec{q},t)$, averaged over all $\vec{q}$, can be measured indirectly using $\mu$SR in a longitudinal applied field, and the result can be compared to $S_\sigma(\vec{q},t)$ obtained using NSE in zero field. The two techniques are complementary in the time domain, since NSE is most sensitive to correlation times in $S_\sigma(\vec{q},t)$ between $10^{-8}$ and $10^{-12}$ s, whereas $\mu$SR is most sensitive to times between $10^{-4}$ and $10^{-11}$ s.

In this paper, the $\mu$SR and NSE measurements of impurity-spin-correlation functions in metallic spin-glasses are compared. The results are also compared to dynamical theories. Preliminary results of this analysis have been previously presented.[7]

## II. MUON-SPIN-RELAXATION DATA

Recently, the longitudinal field dependence of the muon-spin-lattice—relaxation rate $\lambda_\parallel$ has been reported[8] for $T<T_g$ in the spin-glass $Ag_{100-x}Mn_x$, $x=1.6$, 3, and 6 at. %. The technique, which is analogous to a measurement of $T_1$ in NMR, has been described elsewhere.[9,10] The measurements were carried out in applied fields between 0.15 and 5 kOe ($g\mu_B H \ll k_B T$) and at temperatures between $0.3T_g$ and $0.92T_g$. It was found that $\lambda_\parallel=K(T)\omega_\mu^{\nu-1}$, where $\omega_\mu$ is the Larmor frequency of the muon and $K(T)$ is a temperature-dependent constant. The measured values of $\nu$ are given in Table I. Combining these results in the temperature range of $0.3\le T/T_g\le 0.66$ yields $\nu=0.54\pm0.05$. For $T/T_g=0.92$ one obtains $\nu=0.24\pm0.02$.

For muons at rest in the sample, one may argue on very general grounds[11] that $\lambda_\parallel$ is proportional to the noise power $J_h(\omega_\mu)$ in the fluctuating field at the muon Larmor frequency $\omega_\mu$, assuming that the applied field does not appreciably affect the impurity-spin dynamics. The assumption of stationary muons is known to be true at the temperatures of interest.[12] The assumption of field-

TABLE I. Values of the power-law exponent $\nu$ of the noise spectrum below $T_g$ in $Ag_{100-x}Mn_x$ spin-glasses, $1.6\le x\le 6$ at.%, obtained from $\mu$SR measurements as described in Sec. II of the text. Data is from Ref. 8.

| $T/T_g$ | $\nu$ |
|---|---|
| 0.30 | 0.59±0.15 |
| 0.46 | 0.51±0.07 |
| 0.66 | 0.55±0.07 |
| 0.92 | 0.24±0.02 |

 

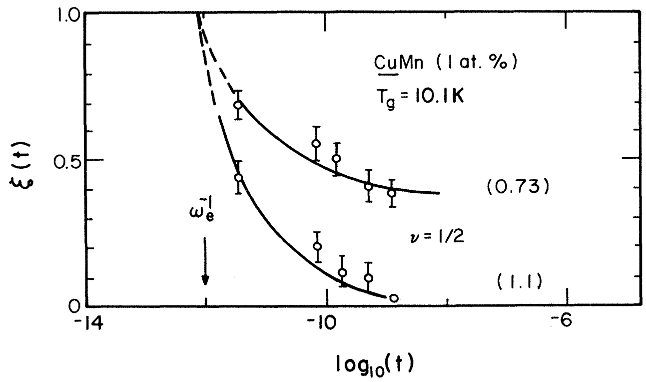


FIG. 1. Impurity-spin-correlation functions $\xi(t)$ obtained from neutron–spin-echo measurements (Ref. 15) for $T/T_g \leq 1.1$ in spin-glass $Cu_{99}Mn_1$. The curves are fits to the functional form of Eqs. (2) and (3) with parameters as indicated. The dotted lines indicate that Eq. (2) is rigorously correct for $\omega_e t \gg 1$. The numbers in parentheses are the reduced temperatures $T/T_g$.

independent dynamics is reasonable for small fields and temperatures well below $T_g$, and is borne out by NMR measurements in *Cu*Mn (Ref. 13) and *Ag*Mn (Ref. 14). Near $T_g$, a direct effect of the field on $J_h(\omega_\mu)$ cannot be ruled out, as discussed below.

## III. NEUTRON SPIN-ECHO DATA

Figures 1 and 2 give NSE results[5,15] for $S_\sigma(\vec{q},t)$ in $Cu_{99}Mn_1$, $T_g = 10.1$ K (Fig. 1), and in $Cu_{95}Mn_5$, $T_g = 27.5$ K (Fig. 2). The $S_\sigma(\vec{q},t)$ data were found to be essentially independent of $\vec{q}$ over the measured range ($0.045 \leq q \leq 0.36$ Å$^{-1}$), and we shall henceforth write $\xi(t) \equiv S_\sigma(q,t)$. The form of $\xi(t)$ was found to be exponential for $T \gg T_g$, but a long-time tail developed near and below $T_g$. The curves in Figs. 1 and 2 are model fits as discussed below.

## IV. COMPARISON OF NSE AND $\mu$SR: $T < T_g$

In order to compare the NSE measurements of $\xi(t)$ to $\mu$SR spin-lattice–relaxation measurements, we take a simplified form for the total spectral density,

$$\tilde{J}_h(\omega) = A\delta(\omega) + BJ_h(\omega) , \quad (1)$$

of muon local-field fluctuations. Here, the first term is related to the quasistatic response of the system, and the second term gives the dynamic fluctuation density. In real systems, the first term may possess a nonzero (but small) width which cannot be detected using finite probe frequencies. Taking the Fourier transform of $\tilde{J}_h(\omega)$ yields the autocorrelation function $S_h(t)$ of the muon local field

$$S_h(t) = \int_{-\infty}^{\infty} d\omega\, \tilde{J}_h(\omega) e^{i\omega t} = A + (1-A)F(t) , \quad (2)$$

where $F(0) = 1$ and $F(\infty) = 0$ such that $A = S_h(\infty)$.

In a real spin-glass, an upper limit to the spectrum of fluctuation frequencies exists. We take this upper limit to be of the order of an exchange frequency $\omega_e \simeq k_B T_g/\hbar$ ($\sim 10^{12}$ s$^{-1}$ for $x \simeq 0.01$ in *Cu*Mn and *Ag*Mn). The $\mu$SR data[8] imply a power-law form $J_h(\omega) \propto \omega^{\nu-1}$, such that

$$F(t) \simeq (\omega_e t)^{-\nu}, \quad \omega_e t \gg 1 . \quad (3)$$

With the use of the form of the correlation function given by Eqs. (1) and (2), and the time dependence implied by the $\mu$SR results [Eq. (3)], values of $S_h(t)$ are obtained and compared with the measured $\xi(t)$ from NSE. This procedure assumes that the form of the correlation function is the same for fluctuations of the impurity spins and the muon local fields; evidence for the validity of this assumption will be given below in Sec. V where NSE measurements in in *Cu*Mn will be compared to $\mu$SR measurements in *Ag*Mn, *Au*Fe, and *Cu*Mn. It must also be assumed that measured frequencies, rates, inverse times, etc., can be scaled by the respective glass temperatures of each sample.

Least-squares fits of the NSE data have been performed using Eqs. (2) and (3), with free parameters $A$, $\omega_e$, and $\nu$. In general, the NSE data are not sufficiently precise to determine all of these parameters accurately, and instead it must be decided whether a physically reasonable set of parameters is consistent with both $\mu$SR and NSE experimental results. The curves in Figs. 1 and 2 give typical fits. It can be seen that the form of Eqs. (2) and (3) represent the NSE data well, using the estimates $\omega_e = 10^{12}$ s$^{-1}$ for $Cu_{99}Mn_1$ and $5\times 10^{13}$ s$^{-1}$ for $Cu_{95}Mn_5$. For $T \leq 0.7T_g$ reasonable fits are obtained with $\nu \simeq \frac{1}{2}$, which is the value obtained from the $\mu$SR data,[8] although the neutron data do not determine $\nu$ well. One can see from Figs. 1 and 2 that at $T/T_g \simeq 0.7$, for example, $A(T)$ is $\sim 0.65$ for $Cu_{95}Mn_5$ and $\sim 0.4$ for $Cu_{99}Mn_1$. Therefore, the NSE data do not obey the expected scaling law, as noted previously.[15]

Below $T_g$, the short-time behavior of the longitudinal muon-spin-relaxation function $G_{\parallel}(t)$ provides direct experimental information on the distribution of *static* muon local fields, and is therefore related to the long-time behavior of $S_h(t)$ [i.e., the value of $A$ in Eq. (2)]. Zero-field measurements of $G_{\parallel}(t)$ in *Ag*Mn,[10] *Au*Fe,[16] and *Cu*Mn[17] for impurity concentrations of $\sim 1$ at.% have determined the width $a(T)$ of the Lorentzian distribution of static local fields in each of these systems. Below $T_g$, $a(T)$ increases with decreasing temperature from $a(T_g) \sim 0$ to the frozen-spin value $a(0)$ for $T \ll T_g$. Figure 3 gives the dependence of $[a(T)/a(0)]^2$ on reduced temperature $T/T_g$ for the above systems. Within about 20% uncertainty, the data scale well. We note that for

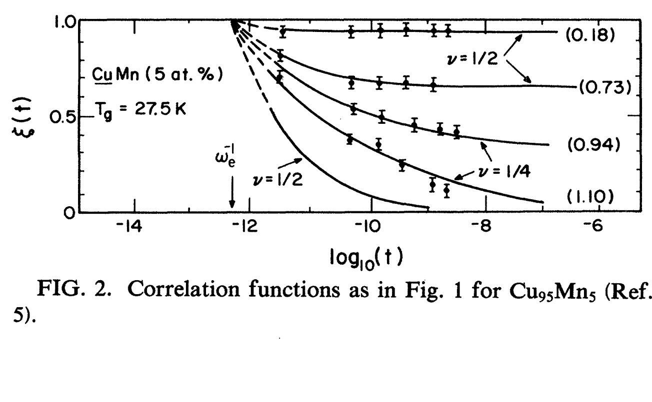


FIG. 2. Correlation functions as in Fig. 1 for $Cu_{95}Mn_5$ (Ref. 5).

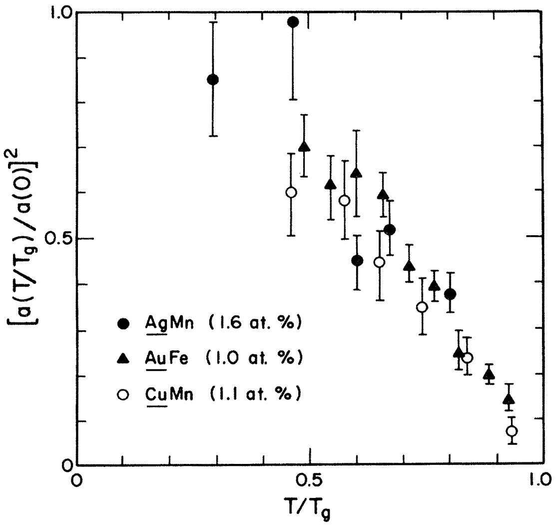


FIG. 3. Dependence of the square of the reduced muon local-field distribution width $[a(T)/a(0)]^2$ on reduced temperature $T/T_g$ in three metallic spin-glass systems at zero field (Refs. 10, 16, and 17).

$T/T_g=0.7$, the value of $[a(T)/a(0)]^2 \simeq 0.4$ agrees well with the NSE value of $A(T/T_g=0.7)$ for $Cu_{99}Mn_1$ (Fig. 1), but is significantly smaller than the value of $A(T/T_g=0.7)=0.65$ obtained for $Cu_{95}Mn_5$ (Fig. 2).

Walstedt and Walker[18] have shown that under some general assumptions ($r^{-3}$ interaction, low impurity concentration) the linewidth $a(T)$ of a probe spin (muon, nucleus) in a magnetic alloy is related to the impurity-spin magnitude by

$$a(T) \propto \langle \, | \langle \vec{\sigma}_i \rangle_T | \, \rangle_i \ . \tag{4}$$

Here $\langle \ \rangle_T$ and $\langle \ \rangle_i$ signify thermal and spatial averages, respectively. Equation (4) is valid in the rapid-fluctuation limit, i.e., for impurity-spin thermal-fluctuation rates much larger than the resultant probe-spin linewidth $a(T)$,[18] which seems to be well established for metallic spin-glasses.[10,16,17] Moreover, $A(T)$, defined in Eq. (2), is given by

$$A(T)=\langle \, | \langle \vec{\sigma}_i \rangle_T |^2 \rangle_i / S^2 \ . \tag{5}$$

Comparison of $a(T)$ ($\mu$SR) and $A(T)$ (NSE) then yields information on the *spatial* distribution of spin magnitudes $\langle \sigma_i \rangle_T$, as can be established by considering two extreme cases.[16]

(a) The $\langle \vec{\sigma}_i \rangle_T$ are all the same. Then

$$\langle \, | \langle \vec{\sigma}_i \rangle_T |^n \rangle_i = | \langle \vec{\sigma}_i \rangle_T |^n \tag{6}$$

for any $n$, and

$$A(T)=[a(T)/a(0)]^2 \ . \tag{7}$$

As noted above, this is consistent with $\mu$SR data for $x \simeq 1$ at. % and the NSE results for $Cu_{99}Mn_1$ at $T/T_g=0.7$.

(b) A fraction $f$ of the spins are saturated ($| \langle \vec{\sigma}_i \rangle_T | = S$), and the remaining fraction is "free" ($\langle \vec{\sigma}_i \rangle_T = 0$). Then

$$\langle \, | \langle \vec{\sigma}_i \rangle_T |^n \rangle_i = f S^n \tag{8}$$

for any $n$, and

$$A(T)=a(T)/a(0)=f \ . \tag{9}$$

The comparison between $\mu$SR and NSE data suggests, therefore, that the impurity spins are distributed in direction, but not in magnitude, below $T_g$. This, in turn, seems consistent with a "percolation" picture of the transition at $T_g$ only if the "infinite cluster," the formation of which is taken to define $T_g$, quickly encompasses essentially all impurity spins as the temperature is lowered below $T_g$. More NSE results at different temperatures in samples with $x \simeq 0.01$, as well as $\mu$SR data at higher concentrations, would be useful in extending this conclusion.

## IV. COMPARISON OF NSE AND $\mu$SR: $T > T_g$

The form of $\xi(t)$ as measured by NSE changes for $T > T_g$, and eventually tends toward an exponential for $T \gg T_g$. Spin probes in rapidly fluctuating local fields are not sensitive to the functional form of $S_h(t)$,[11] but yield an effective correlation time $\tau$ defined by

$$\tau \equiv \int_0^\infty [S_h(t)/S_h(0)] dt \ . \tag{10}$$

Values of $\tau$ have been obtained for *Ag*Mn (Ref. 10), *Au*Fe (Ref. 19), and *Cu*Mn (Ref. 19) from zero-field $\mu$SR measurements. Figure 4 is a scaled plot of $\tau T_g$ vs $T/T_g$ reproduced from Ref. 10.

Mezei[15] has obtained good fits of $\xi(t)$ data obtained from NSE in $Cu_{95}Mn_5$ to the form

$$\xi(t)=\frac{1}{E_M}\int_0^{E_M} \exp[-t/(\tau_0 e^{E/T})] dE \ , \tag{11}$$

which was motivated by considerations of distributions of barrier heights for activated hopping (However, its validi-

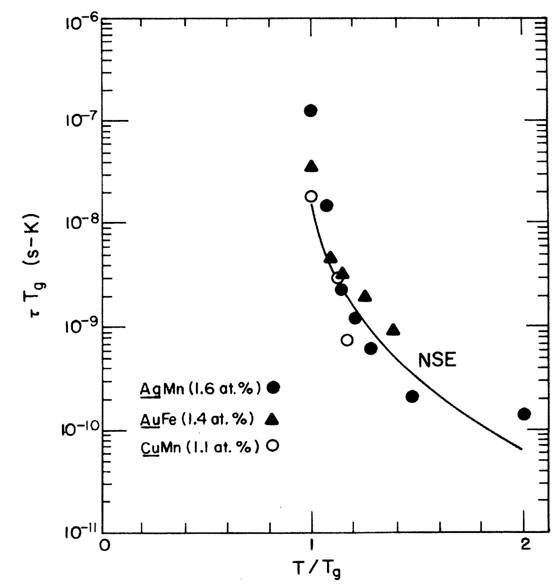


FIG. 4. Dependence of the scaled effective correlation time $\tau T_g$ on reduced temperature $T/T_g \geq 1$ in three metallic spin-glass systems (Refs. 10 and 19) at zero applied field. Data points are results of $\mu$SR measurements. The curve is the fit of Eq. (11) to NSE data for $Cu_{95}Mn_5$.

ty is not necessarily a demonstration of the importance of such barriers.) A good fit was obtained for $E_M=300$ K and $\tau_0=6\times10^{-14}$ s. Scaled values of these parameters, together with Eqs. (10) and (11), yield the curve in Fig. 4. There is good agreement between the $\mu$SR and NSE data, with *no* adjustable parameters. This confirms the similarity between muon local-field and impurity-spin stochastic behavior assumed above over a wide range of values of $\tau$.

## V. CONCLUSIONS

Muon-spin-relaxation and neutron—spin-echo measurements of local-field and impurity-spin-correlation functions $\xi(t)$ in metallic spin-glasses give consistent results for temperatures above and below $T_g$. The time-dependent component of $\xi(t)$ decays as $t^{-\nu}$, $\nu\simeq\frac{1}{2}$, below $T_g$. This behavior is consistent with recent mean-field theories of spin-glass dynamics,[2] as well as with earlier Monte Carlo simulations.[3]

For *Cu*Mn, *Ag*Mn, and *Au*Fe spin-glasses with similar values of $T_g$ ($\pm30\%$), scaled static $\mu$SR linewidths agree well below $T_g$. Futhermore, the relation $A(T)=[a(T)/a(0)]^2$, expected for a uniform distribution of impurity-spin—moment magnitudes, is obeyed for impurity concentrations $x\simeq1$ at. %. This agreement breaks down, however, for comparison with NSE data on $Cu_{95}Mn_5$, where, as noted by Mezei and Murani,[5,15] the larger value of $A(T)$ implies an increase in static spectral density with increasing impurity concentration.

Near the glass temperature, the relation $\xi(t)\propto t^{-\nu}$ still seems to hold, but $\nu$ deviates from $\frac{1}{2}$ for the *Ag*Mn $\mu$SR and $Cu_{95}Mn_5$ NSE data. Curiously, the $Cu_{99}Mn_1$ NSE data still yield $\nu\simeq\frac{1}{2}$ at $T/T_g\simeq1.1$. Longitudinal-field $\mu$SR experiments are not reliable indicators of the value of $\nu$ near $T_g$, however, since $J_h(\omega_\mu)$ may be directly affected by the field. Above $T_g$, where $\xi(t)$ tends toward an exponential form, $\mu$SR and NSE data yield consistent correlation times in zero field, as seen in Fig. 4.

NSE and $\mu$SR measurements are therefore in reasonable quantitative agreement in metallic spin-glasses. This implies (1) that the $q$ independence of $S_\sigma(q,t)$, obtained from NSE measurements for a restricted range of $q$, apparently holds over the wider range of $\vec{q}$ values important for $\mu$SR, (2) a consistent time dependence of $S_\sigma(\vec{q},t)$ is obtained for measuring times $\leq10^{-9}$ s (NSE) and $\geq10^{-9}$ s ($\mu$SR), and (3) perhaps most importantly, more confidence in the reliability of the experimental determinations is warranted than could be given to either technique taken by itself.

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