Interpretation of the temperature dependence of the electron-paramagnetic-resonance linewidth in Ag-Mn spin-glasses

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We present a theory for the temperature dependence of the electron-paramagnetic-resonance linewidth in Ag-Mn spin-glasses in the region above T_g. Our approach differs from the microscopic theory of Levy *et al.* in that we use two-spin correlation functions which are compatible with the results of neutron spin-echo studies on Cu-Mn spin-glasses. The analysis, which is also applicable to paramagnetic resonance in other spin-glasses which have similar spin dynamics, pertains to the region $1.1T_g \lesssim T \lesssim 3T_g$. We obtain scaling behavior for the critical part of the linewidth and the dynamic line shift with effective exponents for the characteristic time and zero-frequency linewidth which are in agreement with the results obtained for Ag—2.6 at. % Mn. We also make a connection between the linewidth and the effective correlation time measured in muon-spin-rotation studies. We find that the effective exponent for the correlation time in Ag—1.6 at. % Mn is consistent with the exponents for the resonance linewidth.

I. INTRODUCTION

In two recent papers[1,2] data for the electron-paramagnetic-resonance (EPR) linewidth have been reported for various Ag-Mn spin-glasses. According to Ref. 2, above T_g the critical part of the linewidth for a Ag—2.6 at. % Mn sample obeys a dynamic scaling relation of the form

$$\Gamma_c \propto [(T-T_g)/T_g]^{-\gamma} G(\omega\tau) , \tag{1}$$

where ω is the resonance frequency and $\tau=\tau_0[(T-T_g)/T_g]^{-\delta}$, T_g being the temperature associated with the maximum in the zero-field susceptibility. The authors find $\gamma=1.5\pm0.1$, $\delta=2.5\pm0.3$, and $\tau_0=3\times10^{-12}$ s. The purpose of this paper is to outline a theory for the linewidth which accounts for the values of γ and δ provides a qualitative description of the scaling function G. In addition, we establish a connection between the critical part of the EPR linewidth and an effective correlation time measured in muon-spin-rotation (μSR) studies.[3]

Recently, Levy *et al.* have presented a microscopic theory for the EPR linewidth in spin-glasses with long-range interactions.[4] The theoretical linewidth, while increasing as T approaches T_g, does not display the scaling behavior found experimentally in Ag-Mn.[5] Our approach is *identical* to that of Ref. 4 up to the point of expressing the linewidth as a time integral over a four-spin correlation function. We *differ* from Ref. 4 only in our treatment of the four-spin function. Rather than decoupling the four-spin function in mode space and using the Kirkpatrick-Sherrington model[6] for the two-spin functions, we follow an earlier approach[7] by decoupling in reciprocal space. In addition, instead of the Kirkpatrick-Sherrington dynamics we use functional forms for the two-spin functions which fit the data obtained from neutron spin-echo (NSE) studies carried out on Cu-Mn.[8] Implicit in our analysis is the assumption that the same functional forms are appropriate for Ag-Mn. Although the analysis has been developed for Ag-Mn and Cu-Mn, we expect it to be applicable to other spin-glasses which have two-spin functions that behave similarly to those in Cu-Mn.

II. SELF-ENERGY

In the approach developed in Ref. 7, where the decoupling of the four-spin function is done in reciprocal space, the self-energy term associated with the EPR linewidth and dynamic line shift is proportional to an expression of the form

$$(T\chi_T)^{-1}\sum_q A_q \int_0^\infty e^{-i\omega t}\,|S(q,t)|^2 dt , \tag{2}$$

where χ_T is the isothermal transverse susceptibility and A_q is a coefficient, which depends on the lattice structure, symmetry-breaking interactions, etc. The symbol $S(q,t)$ denotes the spatial Fourier transform of the two-spin function

$$\langle S_{\alpha i}(t)S_{\alpha j}(0)\rangle,\quad \alpha=x,y,z . \tag{3}$$

In this expression the angular brackets denote a configurational average over all distributions of Mn ions as well as a thermal average. The former is necessary to ensure that the correlation functions depend on the relative separation of the spins, and thus has a well-defined Fourier transform. On the other hand, if the decoupling is done in mode space, as in Ref. 4, (2) is replaced by

$$(T\chi_T)^{-1}\int_0^\infty e^{-i\omega t}\left|\sum_\lambda \langle S_x^\lambda(t)S_x^\lambda(0)\rangle\right|^2 dt , \tag{4}$$

where $\langle S_x^\lambda(t)S_x^\lambda(0)\rangle$ is the two-spin function in the basis λ which diagonalizes the interaction matrix.[6]

The neutron spin-echo measurements for Cu—5 at. % Mn showed that in the range 0.045 $\text{Å}^{-1} \le q \le 0.36$ Å^{-1} $S(q,t)$ was q independent and varied with time in a manner which could be parametrized as

$$\langle S(q,t)S(-q,0)\rangle = \langle S(q)S(-q)\rangle E_M^{-1}\int_0^{E_M} dE \exp[-(t/\tau^*)e^{-E/kT}] , \quad (5)$$

with $E_M \approx 300$ K and $\tau^* = 6\times10^{-14}$ s. Equation (5) has been found to fit the data at 50, 36, and 30 K corresponding to $T=1.8$, 1.3, and $1.1T_g$, respectively ($T_g=27.5$ K), but failed to fit the data closer to T_g. In very recent work on Cu—5 at. % Mn, Uemura *et al.*[9] showed that the q dependence extended to $q\approx 4$ Å^{-1}. In our analysis we will assume that in Ag-Mn $S(q,t)$ is also q independent for all q values which make significant contributions to the critical part of the self-energy. We will further assume that the time dependence of $S(q,t)$ can be parametrized as in Eq. (5) with E_M and τ^* as adjustable parameters. As will be shown below, the critical exponents obtained in the EPR studies on Ag-Mn are consistent with $E_M \approx 10kT_g$, a value similar to that found in Cu-Mn.[8] Strictly speaking, both the two-spin correlation function and the susceptibility are field dependent. However, the scaling behavior observed experimentally shows that the self-energy relevant to the EPR analysis is only weakly field dependent (on a logarithmic scale) for fields up to $0.5T$. Because of this we appear to be justified in approximating both the two-spin function and the transverse susceptibility by their zero-field limits.

With Eq. (5) for the two-spin function, the critical part of the self-energy is written as

$$I_c(\omega,T)=C(T\chi_T)^{-1}\int_0^\infty e^{-i\omega t}\left[E_M^{-1}\int_0^{E_M} dE \exp[-(t/\tau^*)e^{-E/kT}]\right]^2 dt , \quad (6)$$

where C is a constant independent of ω and T which involves the symmetry-breaking interaction responsible for the relaxation of the total moment. Prior to considering the integral over t in Eq. (6) we write

$$\begin{aligned} E_M^{-1}\int_0^{E_M} dE \exp[-(t/\tau^*)e^{-E/kT}] &= (kT/E_M)\int_{(t/\tau^*)e^{-E_M/kT}}^{t/\tau^*} u^{-1}e^{-u}du \\ &= (kT/E_M)[E_1((t/\tau^*)e^{-E_M/kT})-E_1(t/\tau^*)] , \end{aligned} \quad (7)$$

where $E_1(x)$ is the exponential integral.[10] With τ^* in the range of 10^{-14} s we can neglect the second term in square brackets on the right-hand side of (7) in comparison with the first for values of the time (or the reciprocals of the frequencies) which are of interest. Thus we have

$$E_M^{-1}\int_0^{E_M} dE \exp[-(t/\tau^*)e^{-E/kT}] \approx (kT/E_M)E_1((t/\tau^*)e^{-E_M/kT}) . \quad (8)$$

Following Ref. 4 we identify the critical parts of the linewidth, $\Gamma_c(\omega,T)$, and dynamic line shift, $\Delta_c(\omega,T)$, with the real and imaginary parts of $I_c(\omega,T)$, respectively. We find

$$\Gamma_c(\omega,T)=C(T\chi_T)^{-1}(kT/E_M)^2\tau^* e^{E_M/kT} \times\int_0^\infty \cos(\omega\tau y)E_1^2(y)dy \quad (9)$$

and

$$\Delta_c(\omega\tau)=C(T\chi_T)^{-1}(kT/E_M)^2\tau^* e^{E_M/kT} \times\int_0^\infty \sin(\omega\tau y)E_1^2(y)dy , \quad (10)$$

where

$$\tau=\tau^* e^{E_M/kT} . \quad (11)$$

We postpone discussion of Eqs. (9)—(11) until the next section.

III. SCALING AND EXPONENTS

Equations (9) and (10) show that both the linewidth and line shift can be expressed in scaling form as

$$\Gamma_c(\omega,T)=f(T)G^{\Gamma}(\omega\tau) \quad (12)$$

and

$$\Delta_c(\omega,T)=f(T)\omega\tau G^{\Delta}(\omega\tau) , \quad (13)$$

with the characteristic time τ, given by Eq. (11), being identified with the maximum decay time characterizing the evolution of $\langle S(q,t)S(-q,0)\rangle$.

Plots of the two functions $G^{\Gamma}(\omega\tau)$ and $G^{\Delta}(\omega\tau)$, normalized to unity at $\omega\tau=0$, are given in Fig. 1. Also shown is the asymptotic behavior of the experimental scaling function for Ag—2.6 at. % Mn reported in Ref. 2 along with the functional form

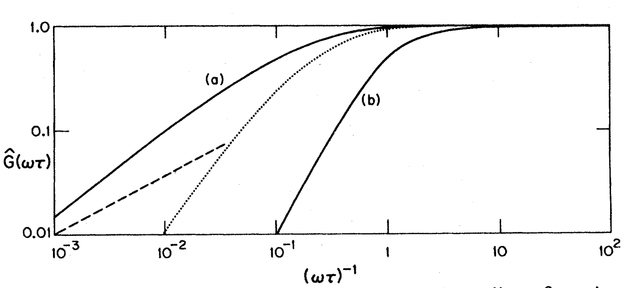

FIG. 1. Log-log plot of the normalized scaling functions $\hat{G}(\omega\tau)=G(\omega\tau)/G(0)$ vs $(\omega\tau)^{-1}$. (a) $\hat{G}^{\Gamma}(\omega\tau)$, Eq. (12) with $G^{\Gamma}(0)=2\ln 2$. (b) $\hat{G}^{\Delta}(\omega\tau)$, Eq. (13) with $G^{\Delta}(0)=\ln 2-0.5$. The dotted line is $[1+(\omega\tau)^2]^{-1}$, the result obtained for both $\hat{G}^{\Gamma}$ and $\hat{G}^{\Delta}$ for exponential decay. The dashed line indicates the asymptotic behavior of $\hat{G}^{\Gamma}(\omega\tau)$ for Ag—2.6 at. % Mn that was reported in Ref. 2.

$$G(\omega\tau)=[1+(\omega\tau)^2]^{-1}\ , \tag{14}$$

which is obtained for both G^{Γ} and G^{Δ} assuming that $|S(q,t)|^2$ decays as $\exp(-t/\tau)$. A comparison of the scaling function obtained from Eq. (9) with the data of Ref. 2 indicates that the calculated curve is in reasonable qualitative agreement with experiment in that it reproduces the high-frequency tail which is present for $\omega\tau \gg 1$. It should be noted, however, that the single exponential, although inadequate at high frequencies, does give a better fit to the data in the region $\omega\tau \lesssim 1$.

In analyzing the temperature dependence it should be emphasized that the experimental data reported in Ref. 2 are confined to the region $0.1 \lesssim (T-T_g)/T_g \lesssim 4.5$ with power-law behavior limited to $(T-T_g)/T_g < 3$. Although not immediately obvious the functional forms obtained for the characteristic time τ and the low-frequency limit of the linewidth do show effective power-law behavior in the region $0.2 \lesssim (T-T_g)/T_g \lesssim 2$.

In order to extract effective exponents we consider the behavior of a log-log plot of $T^n \exp(E_M/kT)$ versus $(T-T_g)/T_g$. Taking $x=\ln[(T-T_g)/T_g]$ we have

$$\ln(T^n e^{E_M/kT}) = n\ln[T_g(1+e^x)] + (E_M/kT_g)(1+e^x)^{-1}\ . \tag{15}$$

For $n=0$, the appropriate value for the characteristic time, the right-hand side of (15) has an inflection point at $x=0$ $(T=2T_g)$, with the slope at the inflection point equal to $-E_M/4kT_g$. We use the slope at the inflection point to define an effective exponent for the characteristic time, δ_{eff}. Taking $E_M=10kT_g$, as found in Cu-Mn over a range of concentrations, we obtain $\delta_{\text{eff}}=2.5$ in agreement with experiment.

Using the negative of the slope at $x=0$ $(T=2T_g)$ to define effective exponents for general n we find

$$-\frac{d}{dx}\{n\ln[T_g(1+e^x)]+(E_M/kT_g)(1+e^x)^{-1}\}\bigg|_{x=0} = (E_M/4kT_g) - n/2\ . \tag{16}$$

In applying this analysis to the low-frequency limit of the EPR linewidth, we neglect the temperature dependence of the product $T\chi_T$ (Ref. 1) so that

$$\Gamma(0,T) \sim T^2 e^{E_M/kT}\ , \tag{17}$$

corresponding to $n=2$. From Eq. (16), evaluated with $E_M=10kT_g$, we obtain an effective exponent for the linewidth $\gamma_{\text{eff}}=1.5$, which is also in agreement with experiment.

Although the effective exponent is only characteristic of the slope at $T=2T_g$ the behavior of $e^{E_M/kT}$ and $T^2 e^{E_M/kT}$ is reasonably well approximated by a power law over the interval $0.3 \lesssim (T-T_g)/T_g \lesssim 2.5$. This is evident in Fig. 2 which is a log-log plot of $(T/T_g)^n e^{-5} \times \exp(10T_g/T)$ versus $(T-T_g)/T_g$ for $n=0$, 1, and 2.

Finally, if we equate Eq. (11) at $T=2T_g$, $E_M=10kT_g$, to the measured value of the characteristic time at $2T_g$, 3×10^{-12} s, we find $\tau^*=2\times10^{-14}$ s, which is comparable to the value of 6×10^{-14} s inferred for Cu–5 at. % Mn.

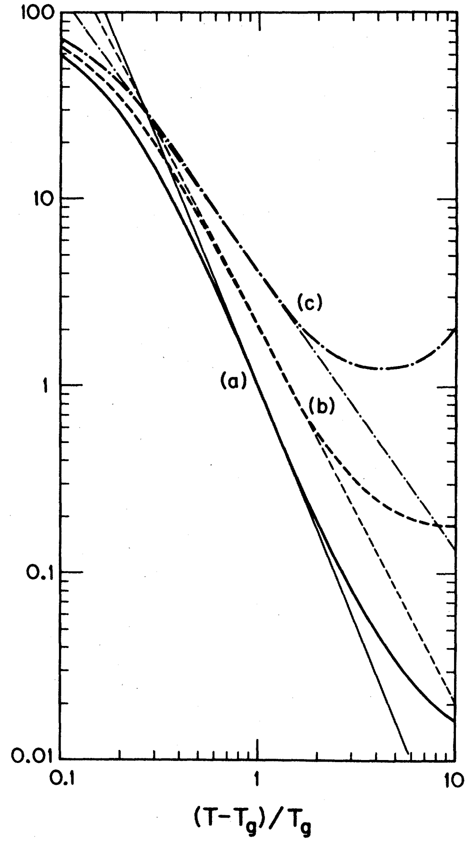

FIG. 2. Log-log plots of $(T/T_g)^n \exp[10T_g/T-5]$ vs $(T-T_g)/T_g$. (a) $n=0$, (b) $n=1$, (c) $n=2$. In each case the corresponding straight line is tangent to the curve at $T=2T_g$ and indicates the region of approximate power-law behavior with effective exponent 2.5 (a), 2.0 (b), and 1.5 (c). Curves (a) and (c) describe the characteristic time and low-frequency limit of the EPR linewidth, respectively, while curve (b) characterizes the temperature dependence of the effective correlation time inferred from μSR studies.

IV. μSR STUDIES

The theory outlined in Sec. III accounts for the effective exponents of the characteristic time and the low-frequency limit of the EPR linewidth. As indicated in the Introduction, there is a close connection between the EPR, μSR, and NSE results. Provided the q dependence of the two-spin correlation functions can be overlooked, as appears to be the case in Cu-Mn and assumed to be the case in Ag-Mn, the effective correlation time inferred in μSR studies is given by[3]

$$\hat{\tau} = \langle S(q)S(-q)\rangle^{-1}\int_0^\infty \langle S(q,t)S(-q,0)\rangle dt\ , \tag{18}$$

which reduces to

$$\hat{\tau} = \tau^*(kT/E_M)e^{E_M/kT}\ , \tag{19}$$

with the postulated correlation function and $E_M \gg kT$.

Using Eq. (16) and taking $E_M=10kT_g$ as in the EPR

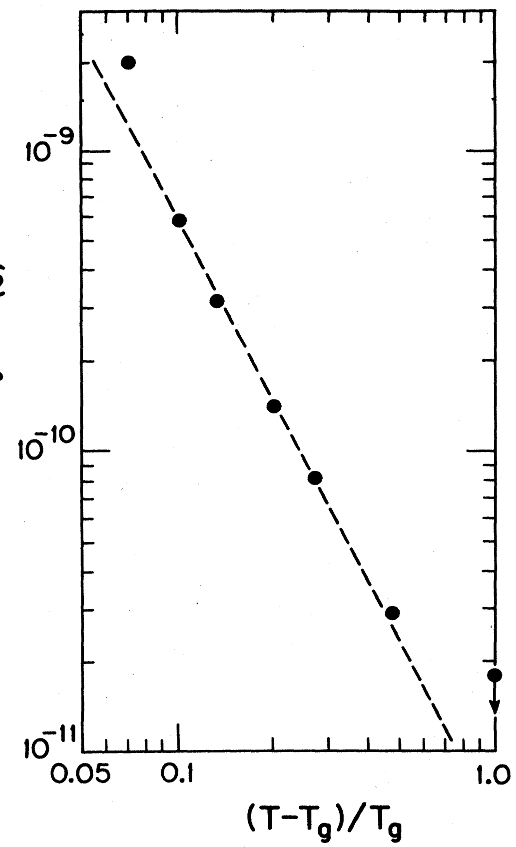

FIG. 3. Log-log plot of $\hat{\tau}$ vs $(T-T_g)/T_g$. $\hat{\tau}$ is the effective correlation time inferred from μSR studies of Ag—1.6 at. % Mn and is equal to the time integral of the spin autocorrelation function for q-independent dynamics [Ref. (3)]. The data points are from Ref. 11 with the arrow indicating that the value at $T=2T_g$ is probably an upper limit. The dashed line has slope -2, which corresponds to an effective exponent equal to 2.0.

analysis we predict an effective exponent equal to 2 for μSR data in the neighborhood of $T=2T_g$. In Fig. 3 we display a log-log plot of $\hat{\tau}$ for An—1.6 at. % Mn.[11] The arrow attached to the point at $T=2T_g$ indicates that the value plotted is probably an upper limit since a nearly identical value is obtained for $T=3.3T_g$.[12] Omitting that point and the one for $(T-T_g)/T_g=0.07$, the remaining five points fall approximately on a straight line of slope 2 corresponding to an effective exponent equal to 2, in agreement with our prediction.

Extrapolating the straight line in Fig. 3 to $T=2T_g$ we obtain the value $\hat{\tau}=6\times10^{-12}$ s. Using Eq. (19) and taking $E_M=10kT_g$ we infer $\tau^*(1.6\text{ at. }\%)=2\times10^{-13}$ s. Assuming τ^* scales inversely with T_g, this value corresponds to $\tau^*=2\times10^{-13}$ s $[(7.5\text{ K})/(10.24\text{ K})]=1.5\times10^{-13}$ s for the Ag—2.6 at. % Mn sample studied in Ref. 2, which is in order-of-magnitude agreement with the value of τ^* inferred from the EPR analysis.

V. DISCUSSION

The analysis developed in the preceding sections provides an interpretation of the temperature dependence of the data for Ag-Mn in the range $1.1T_g \lesssim T \lesssim 3T_g$. Using two-spin functions which are compatible with the results of NSE studies on Cu-Mn spin-glasses, we are able to account for the scaling behavior and the temperature dependence of the low-frequency linewidth and the characteristic time. In addition, we can explain the temperature dependence of the effective correlation time measured in μSR experiments.

The dynamic scaling may be the most surprising result to emerge to date from EPR studies of spin-glasses. It can only come about if the four-spin correlation function in the self-energy integral is insensitive to the presence of applied fields of the strengths employed in the experiment. When this is the case, changing the resonance frequency has the effect of probing the frequency dependence of the Fourier transform of the four-spin correlation function. The weak field dependence was taken into account implicitly in our analysis when we made use of two-spin functions obtained from zero-field NSE measurements.

The q independence of the long but finite wavelength spin dynamics in Cu-Mn is also difficult to understand. One can only surmise that the anisotropic terms in the Hamiltonian play a more important role than one would naively expect with the consequence that the total spin is not even an approximate constant of the motion.[13] It should be noted that the temperature range over which the theory is applicable coincides with the range over which it is possible to obtain meaningful values for the critical part of the linewidth. Below $1.1T_g$ the resonance line shape becomes non-Lorentzian.[1] Above $(3-4)T_g$ the critical part of the width is only a few percent of the total width.

As discussed in the Introduction our theory differs from that of Levy *et al.*[4] only in the treatment of the four-spin function appearing in the self-energy. Although we utilize different decoupling schemes this distinction is unimportant as long as the two-spin functions are only weakly q dependent. The important difference is that we use an experimentally determined two-spin function, whereas in Ref. 4 the spin dynamics is assumed to be that of the Kirkpatrick-Sherrington model.

In assessing the accuracy of the theory one must keep in mind the approximate nature of the fit to the NSE data provided by Eq. (5). As emphasized by Mezei,[8] the parametrization with a temperature-independent E_M breaks down in the limit as T approaches T_g. This fact may explain the persistence of the straight-line behavior in Fig. 3 relative to what one would expect from curve (b) in Fig. 2. Thus the question of whether a calculation of the EPR absorption spectrum which utilized the correct two-spin function would remain valid down to T_g remains open.

Although the theory works well in the case of Ag-Mn the agreement with the EPR results for Cu-Mn obtained to date appears to be less satisfactory. In particular, data for Cu—3 at. % Mn show that the ratio of the linewidth to the relative g shift $\Delta g/g$, which is assumed to be dynamic in origin, is approximately described by the equation

$$\Gamma_c g/\Delta g=[0.75+14.7(T/T_g-1)^{1.3}]\times10^{11}\ \text{s}^{-1}\ . \tag{20}$$

The near straight-line behavior of Eq. (20), when plotted against T, differs from the results obtained from Eqs. (12) and (13) in the limit $\omega\tau \ll 1$. From these equations we obtain

$$\Gamma_c g/\Delta g = [G^{\Gamma}(0)/G^{\Delta}(0)](\tau^*)^{-1}e^{-10T_g/T}, \tag{21}$$

which deviates significantly from straight-line behavior. We note, however, that with

$$G^{\Gamma}(0)/G^{\Delta}(0) = 2\ln 2/(\ln 2 - 0.5), \tag{22}$$

the value of τ^*, inferred by matching (20) and (21) at $T = 2T_g$, is 3×10^{-14} s, which is comparable to the NSE value of τ^*. Although it is possible that the form hypothesized for the two-spin functions may be a better fit to the actual functions in Ag-Mn than in Cu-Mn, a more plausible explanation, assuming the g shift is entirely dynamic in origin, is the breakdown of the condition $\omega\tau \ll 1$ as $T \to T_g$. If we scale τ for Cu—5 at. % Mn, $6\times 10^{-14}e^{10T_g/T}$ s, by the ratio of the glass temperatures for the 5 at. % and 4 at. % samples (27.5 K)/(18.9 K), we find that with the experimental frequency, 9.3 GHz, $\omega\tau > 1$ for $T < 1.9T_g$. On the basis of this result we suggest that a reanalysis of the EPR experiments in Cu-Mn along the lines of Ref. 2 would be worthwhile.[15]

Finally, we emphasize that the theory in the form presented is expected to be qualitatively applicable only to those metallic spin-glasses with long-range interactions which have spin dynamics similar to Cu-Mn and Ag-Mn. Whether or not EPR in insulating spin-glasses with short-range interactions can be described by a modified theory, with appropriate two-spin functions and symmetry-breaking interactions, is also an open question.

Note added in proof. Recent μSR studies of the amorphous spin-glass $Pd_{75}Fe_5Si_{20}$ show that the effective correlation time varies as $(1-T_g/T)^{-2}$ over the internal $1.1T_g \lesssim T \lesssim 2.2T_g$ [C. Y. Huang (private communication)]. When plotted against the reduced temperature $T/T_g - 1$ the data are characterized by an effective exponent approximately equal to 1.4, indicating that the spin dynamics in this system are different from that of the Ruderman-Kittel-Kasuya-Yosida spin-glasses.

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