Dynamical Crossover in an Ising Spin Glass above T_g: A Muon-Spin-Relaxation Investigation of $Fe_{0.05}TiS_2$

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We investigate the temperature dependence of the spin-spin dynamical autocorrelation function of the Ising spin glass $Fe_{0.05}TiS_2$ through field dependent muon-spin lattice relaxation measurements. We successfully analyze the results using the Ogielski function, namely, $t^{-x}\exp(-[t/\tau]^y)$ as employed in numerical simulations. The experimental estimates of x, y, and τ are compared with those from simulations. Our major finding is that in this system the correlation function changes its nature from Ogielski to a form indistinguishable from pure stretched exponential upon cooling close to T_g, indicating a dynamical crossover.

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The Ising model with random interactions is used to describe spin glasses (SG) as well as other objects on a lattice with random interactions, because it incorporates phase transition, frustration, and metastability with a minimal number of degrees of freedom. As a result there has been a tremendous effort over the years to calculate and simulate the dynamical and thermodynamical properties associated with this model. Since the simulations of Ogielski [1] it became clear that the dynamical autocorrelation function of Ising SG $q(t) \equiv \langle \mathbf{S}(t)\mathbf{S}(0)\rangle$ cannot be characterized simply by an exponential decay, or even by exponential decays with a distribution of correlation times. Similar results were obtained later by other simulations [2,3], and theoretical explanations were proposed [4]. The first experimental evidence for this phenomenon was provided by the neutron spin echo (NSE) experiments of Mezei and Murani [5]; more recent studies are the muon-spin-relaxation (μSR) work of Keren *et al.* [6,7] and the NSE measurements of Pappas [8]. However, these experiments were done on metallic spin glasses where the spins are Heisenberg and therefore cannot be compared directly with Ising simulations or theories. Relaxation was measured using ac susceptibility on an Ising system but this technique is restricted to a maximum T very close to T_g [9]. The purpose of the present work is to overcome these difficulties and to determine the functional form and the parameters of the autocorrelation decay in an Ising spin glass as a function of temperature over a wide range of T including the vicinity of T_g. As far as we know this is the first attempt of any kind to determine the correlation function in a real Ising spin glass material over a wide T range. We find a qualitative agreement between simulations and experiment, but there are significant quantitative differences.

The determination of $q(t)$ is done using μSR measurements [T_1]. In a muon T_1 measurement one determines the time t it takes the polarization $P_z(H,t)$ of a muon to reach thermal equilibrium with the system under investigation, from an initial nonequilibrium polarization $P_z(H,0) = 1$, while applying an external field $\mathbf{H}$ along the initial muon polarization. The muon polarization, in turn, is proportional to the asymmetry $A(H,t)$ of the decay positrons in the direction parallel and antiparallel to the initial muon polarization.

Our study is conducted on $Fe_{0.05}TiS_2$ which is a well-established Ising spin glass [10–12]. It was chosen since a muon is sensitive to the magnetic fields generated by nuclear moments, so it is best to work with spin glasses which have low or zero nuclear moments in order to eliminate parasitic nuclear terms which can obscure the local magnetic moment effects. Nuclear moments in $Fe_{0.05}TiS_2$ are all zero or small. In Fig. 1 we show magnetization measurements in a single crystal of $Fe_{0.05}TiS_2$ using a SQUID magnetometer. The units are arbitrary for presentation purposes. We have used field cooled (FC) and

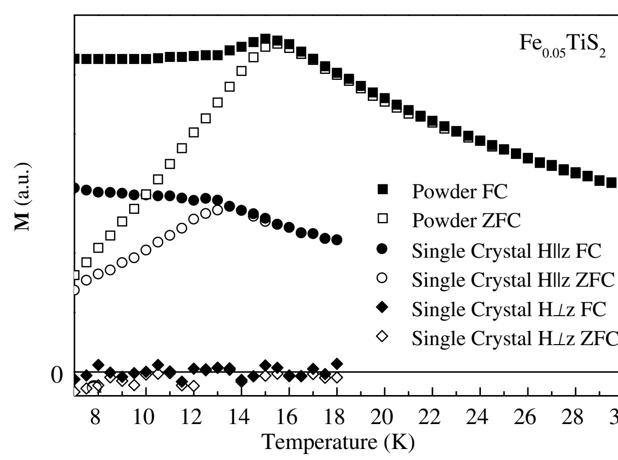

FIG. 1. Field cooled and zero field cooled magnetization measurements in a single crystal and powder of $Fe_{0.05}TiS_2$ for two field orientations.

zero field cooled (ZFC) procedures, and the field is applied both parallel and perpendicular to the easy axis **z**. When the field is perpendicular to **z** we cannot detect magnetization in either the FC or the ZFC measurements. In contrast, when the field is parallel to **z** the compound behaves as a standard SG with a cusp at $T_g = 13.5$ K and deviation between the FC and ZFC measurements below T_g. For our μSR measurements we used a powder of the same nominal concentration, which had a $T_g = 16$ K. The magnetization data in the powder are also depicted in Fig. 1. The value of T_g and the highest field applied in our experiment obey the condition $\mu_{\mathrm{eff}} H < kT_g$, where $\mu_{\mathrm{eff}} \simeq 4\mu_B$ [11] is the magnetic moment of the Fe ions. This condition ensures that the experiment is in the zero field limit, as far as the electronic system is concerned [12].

Representative $A(H, t)$ measurements in $T = 25.09(1)$ and 17.05(1) K and at various fields are shown in Figs. 2(a) and 2(b) on a semilog scale. Clearly, H affects the time dependence of the muon polarization very strongly, especially at 17 K. As H increases, the relaxation rate of the asymmetry decreases. In addition, at 17 K, wiggles are observed in the data at early times and low fields.

Our preliminary data analysis relies on the expression

$$P_z(H, t) = P_z[t/T_1(H)], \tag{1}$$

where $1/T_1$ is given by the Fourier transform (FT) of the field-field dynamical autocorrelation function

$$\Phi(t) \equiv \gamma_\mu^2 \langle \mathbf{B}_\perp^d(t) \mathbf{B}_\perp^d(0) \rangle \tag{2}$$

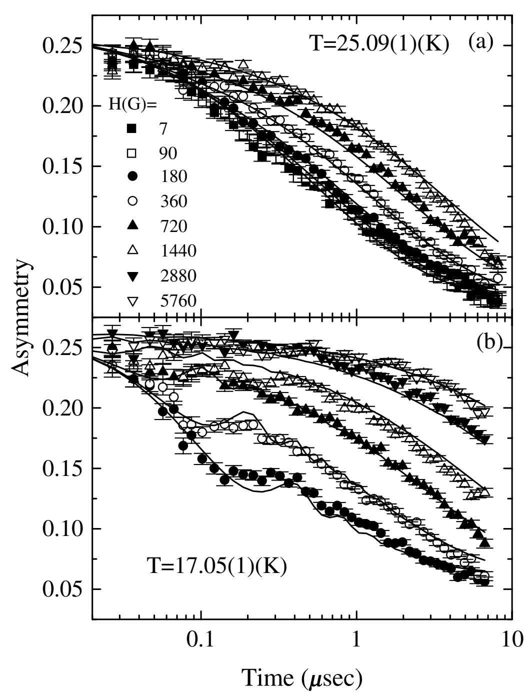

FIG. 2. Asymmetry versus time on a semilogarithmic scale in two temperatures for various longitudinal magnetic fields. The solid lines are fits to the model described in the text.

evaluated at $\gamma_\mu H$ (for detailed discussion, see Ref. [7]). Here $\mathbf{B}_\perp^d$ is the dynamical part of the magnetic field at the muon site perpendicular to $\mathbf{H}$; $\gamma_\mu = 85.162$ MHz/kG is the muon gyromagnetic ratio. This expression is valid in the *long time* limit, namely, when $\Phi(t)$ is much smaller than its initial value. Since there are no cross correlations in spin glasses we expect $\Phi(t) \propto q(t)$. If, for example, $q(t)$ is exponential, then $\Phi(t) = 2\Delta^2 \exp(-t/\tau)$, where $2\Delta^2 = \gamma_\mu^2 \langle \mathbf{B}_\perp^2 \rangle$, and

$$\frac{1}{T_1(H)} = \frac{2\Delta^2 \tau}{1 + (\gamma_\mu \tau H)^2}. \tag{3}$$

However, in a spin glass, the muon could stop in a variety of environments and *a priori* can experience different instantaneous fields or correlation times. By allowing for a distribution ρ of Δ and/or τ we can obtain the average polarization

$$\overline{P}(H, t) = \iint \rho(\Delta, \tau) P\left(\frac{2\Delta^2 \tau t}{1 + (\gamma_\mu \tau H)^2}\right) d\Delta d\tau. \tag{4}$$

The important aspect of this expression is that for asymptotic fields $\gamma_\mu H \gg 1/\tau^{\min}$, where $\tau^{\min}$ is the shortest correlation time in a distribution of exponential relaxations, we expect the scaling relation

$$\overline{P}(H, t) = \overline{P}(t/H^\gamma) \tag{5}$$

with $\gamma = 2$. This value of γ is obtained regardless of the distribution $\rho(\Delta, \tau)$. Therefore, any strong deviation from this scaling relation is an indication that exponential relaxations with a distribution of correlation times is not an adequate description of the spin autocorrelation function and other options should be explored.

Two interesting options are (i) a cutoff power law correlation, namely, $\Phi(t) \sim t^{-x} f(t/\tau)$, where f is a cutoff function; the FT of this function falls off like $1/H^{1-x}$ at high fields, so $\gamma = 1 - x < 1$. (ii) The stretched exponential $\Phi(t) \sim \exp[-(t/\tau)^y]$; the FT of this function behaves like $1/H^{1+y}$ at high fields, therefore $\gamma = 1 + y > 1$.

Indeed, the field time scaling relations of Eq. (5) are shown to hold at *high fields* and *late times* in Figs. 3(a) and 3(b) for the corresponding data sets presented in Fig. 2. We find that γ is very different from 2 in both cases; at $T = 17$ K, $\gamma = 1.2(1)$, and at $T = 25$, $\gamma = 0.6(1)$. Therefore, as mentioned above, the correlation function cannot be described by exponentials with single or multiple correlation times. While γ's which are smaller than 2 were found before [6,8], this is the first observation of γ that changes from below 1 to above 1 as a function of temperature. It indicates a dynamical crossover upon cooling in the asymptotic behavior of the Fourier transform of the correlation function, or, equivalently, in the early time behavior of $\Phi(t)$.

However, Eq. (5) cannot account for the wiggles seen in the data in Fig. 2(b) and the data seem to hold more information. Physically, the relaxation proceeds as a succession of abrupt single spin flip events. The source of the

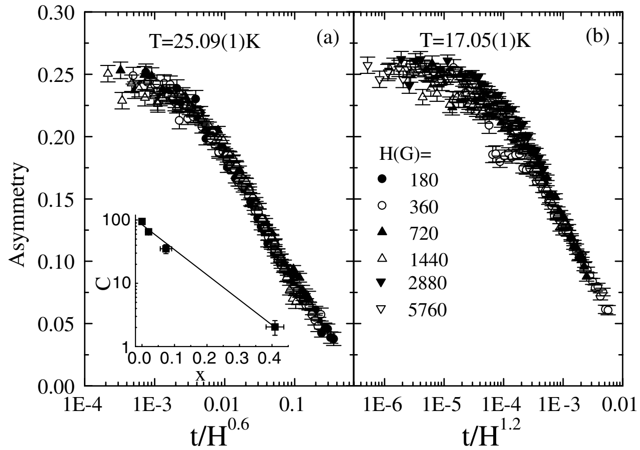

FIG. 3. The asymmetry from Fig. 2 plotted as a function of t/H^{γ} for fields higher than 180 G. (a) At $T = 25.09$ K all data sets collapse into one function for $\gamma = 0.6$. (b) The same happens in $T = 17.05$ K provided that $\gamma = 1.2$. The inset shows the parameter C on a semilog scale vs the power x as obtained from the fits of Eq. (8) to the data.

wiggles are those muons for which the transverse magnetic field has not yet flipped abruptly during a muon rotation period. These muons will oscillate around the vector sum of both internal and external fields at a frequency $\omega = \gamma_{\mu}|\mathbf{H}+\mathbf{B}^d|$. But since $\mathbf{B}^d$ is a random field with zero average, its contribution will show up as a damping of the wiggles, while the frequency of the wiggles will be at $\omega = \gamma_{\mu}H$. Roughly, if there is a cutoff time τ, then by the time $t > \tau$ an abrupt flip has occurred for each muon. Therefore, a wiggle pattern will not be seen for longer times. As a result, the observation of a "wiggle" pattern up to time t is the proof that a significant fraction of local moments have not flipped abruptly within this time scale, and the additional information they provide is a model independent estimate of the cutoff time.

In order to account for both the scaling and the wiggles simultaneously we must apply the equation developed in Ref. [7]. There it was shown that the relation between $A(H, t)$ and $\Phi(t)$ is via the expression

$$A(H, t) = A_0 \exp\{-[\Gamma(H, t)t]^{\beta}\} + B_g, \tag{6}$$

where

$$\Gamma(H, t)t = \int_0^t (t - t')\Phi(t')\cos(\gamma H t')dt'. \tag{7}$$

At *late times,* one finds that $\Gamma(H, t) \to 1/T_1(H)$ restoring Eq. (1). In addition, Eqs. (6) and (7) lead to the dynamical Kubo-Toyabe behavior under the appropriate site averaging [13]. As for the other parameters: A_0 is the initial asymmetry, B_g is background due to muons that missed the sample, β is a phenomenological fit parameter introduced since at high temperatures, where $\Gamma(H, t) = 1/T_1(H)$, the muon relaxation is a stretched exponential. In order to account for the data at all temperatures we assume the Ogielski correlation function

$$\Phi(t) = 2ct^{-x} \exp[-(t/\tau)^y]. \tag{8}$$

Although this function is not well defined at $t = 0$, the integral in Eq. (7) exists. According to our scaling based conclusions above, the fits should yield $x \simeq 0$ and $y = 0.2$ at $T = 17$ K, and $x = 0.4$ and $y \geq 0$ at $T = 25$ K.

We fit Eq. (6) with Eqs. (7) and (8) to the data. $\beta = 0.62(3)$; A_0 and B_g are global fit parameters for all fields and temperatures. The parameters x, y, τ, and c are common to all fields at a given temperature. Fit results for two cases are shown by the solid lines in Fig. 2 and the fit parameters are presented in Fig. 4. While the fit parameters seem to be numerous they are determined nearly uniquely by different regions of the data in a model independent way. As was explained above, x and/or y are set by the late times (past wiggles) and high fields; τ is set by the early time low field wiggles. The parameter c is set by the initial relaxation rate. Indeed, the error bars on these parameters are very small. For comparison we also present the simulation parameters from Ogielski [1].

At $T = 17$ K the fit yields $x \simeq 0$ and $y = 0.35$. These values are in agreement with the scaling prediction. We thus find $y \simeq 1/3$ at $T \to T_g$ as was obtained by Ogielski's simulations and by nearly every other simulation known to us. In addition, at $T > T_g$ the values of x are clearly bigger than zero and are increasing with increasing T. In this

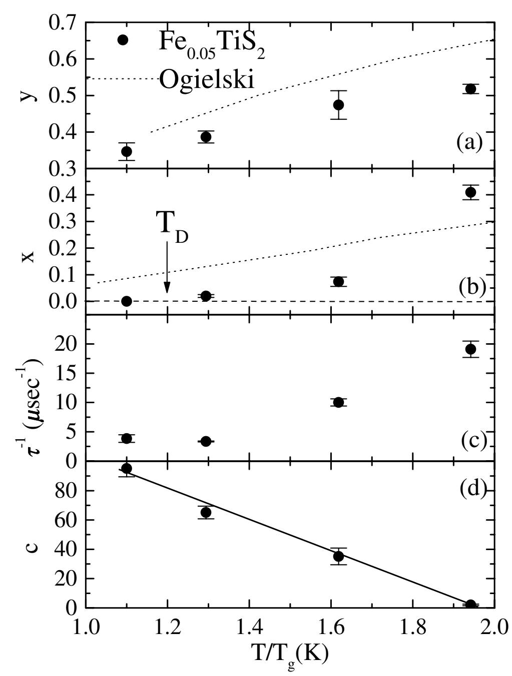

FIG. 4. The temperature dependence of the parameters (a) y, (b) x, (c) τ^{-1}, and (d) c when accounting for the μSR data using the correlation function given in Eq. (8). The simulation results of Ogielski are also shown. The temperature where the crossover from an Ogielski to stretched exponential correlation takes place is marked in panel (b) by T_D.

temperature range our results are in a qualitative agreement with Ogielski's simulations and with more recent results [3], although there are significant quantitative departures from the simulations results. In particular, the simulations failed to anticipate the $x \simeq 0$ limit at $T \rightarrow T_g$ and the dynamical crossover from Ogielski to a stretched exponential dominated behavior. An estimate of the crossover temperature T_D is presented Fig. 4(b). The very low power law exponent $x(T_g)$ in this system is consistent with a value estimated directly from NSE measurements on another $FeTiS_2$ sample [14]. As for the parameter τ, it has an interesting behavior. It increases with decreasing temperature but appears to saturate close to $T/T_g = 1$. A similar T independent correlation time close to T_g was found in AgMn [7] and the simulations of [3]. Since close to T_g the wiggles span nearly the entire μSR time window, the saturation of τ might be an experimental effect. We conclude that within our time window, the most active parameter above the SG transition is the exponent y.

We now discuss the parameter c. From dimensional analysis (and other considerations [7]) this parameter can be written as $c = \Delta^2 \tau_e^x$, where τ_e has dimensions of time. When $x = 0$, $c = \Delta^2$. From the value of c in $T = 17$ K we find $\langle \mathbf{B}_\perp^2 \rangle = 0.026$ kG^2, namely, the local field is of the order of 100 G. This was confirmed independently by measurements at temperatures well below T_g (not shown) where the internal field is static and $\langle \mathbf{B}_\perp^2 \rangle$ is the only unknown variable. Finally, $\langle \mathbf{B}_\perp^2 \rangle$ should be temperature independent. If τ_e is also T independent we expect a $\log(c)$ against the x plot to be a straight line. Indeed, such a linear dependency is consistent with the data as shown in the inset of Fig. 3(a). The slope of the line, which is $\log\tau_e$, gives $\tau_e = 0.14(6)$ nsec.

In summary, we demonstrate that a description in terms of exponentially decaying correlation functions with a distribution of correlation times cannot explain our muon depolarization data. The Ogielski function is much more suitable for this purpose. We have managed to extract all the parameters for this function and to study their temperature dependence. We find that close to T_g the power law part becomes invisibly small and the correlation is dominated by a stretched exponential with exponent near $1/3$. The slowing down of the spin fluctuation is the combined result of a stretching out of the exponent and a lengthening time scale.

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