Muon Spin Relaxation Study of Reentrant Spin Glasses: Amorphous-Fe$_{1-x}$Mn$_x$

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We have performed an extensive muon spin relaxation study of the well known (Fe$_{1-x}$Mn$_x$)$_{75}$P$_{16}$B$_6$Al$_3$ reentrant spin glass. We find a strong increase of the dynamic as well as static muon depolarization rate at the paramagnetic to ferromagnetic transition temperature, $T_c$, and at $T_f$ ($T_f < T_c$) that corresponds to the onset of strong bulk magnetic irreversibilities. We find no critical dynamic signature of a freezing of the transverse $XY$ spin components at an intermediate temperature $T_{xy}$ ($T_f < T_{xy} < T_c$) where extra static order not contributing to the magnetization starts developing.

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Frozen random disorder added to competing interactions leads to random frustration in condensed matter systems [1], a feature that is detrimental to the stability of the long-range ordered state otherwise present in the disorder-free pure material. Random frustration is ubiquitous in condensed matter physics [1]: it arises in random magnets [2], disordered Josephson junction arrays in a magnetic field [3], mixed molecular systems [4], and in partially UV polymerized lipid membranes [5]. One expects these systems to share similar thermodynamic behavior and, in particular, to display a long-range ordered phase with low-temperature glasslike properties [6,7]. Hence, an improved understanding of the simpler randomly frustrated ferromagnets would contribute to our overall understanding of several other types of weakly frustrated systems.

The mean-field theory of weakly randomly frustrated Heisenberg ferromagnets predicts a sequence of three transitions [8]. Upon cooling, a transition from a paramagnetic phase to a collinear ferromagnetic state first occurs at $T_c$, below which the system develops a nonzero bulk magnetization, $M$ (e.g. along the $z$ direction). At a lower temperature, $T_{xy}$, the transverse $XY$ spin components perpendicular to $M$ freeze in random directions, with no decrease of $M$ at $T < T_{xy}$. The temperature vs disorder, $x$, boundary, $T_{xy}(x)$, is usually referred to as the Gabay-Toulouse line [8]. Finally, strong longitudinal irreversibilities develop at a third temperature, $T_f$ ($T_f < T_{xy}$), with again no decrease of $M$ at $T < T_f$ [9,10,11]. The boundary $T_f(x)$ in the mean-field theory of Heisenberg spin glasses is a remnant of the longitudinal freezing seen below the Almeida-Thouless line in weakly frustrated Ising spin glasses [8]. In mean-field theory, $T_{xy}$ is a true thermodynamic (transverse spin-glass) phase transition, while $T_f$ is a sharp crossover temperature rather than a transition per se. Systems exhibiting such mixed ferromagnetic and spin glass behavior are commonly referred to as “reentrant spin glasses” [10,11]. An extensive experimental effort using macroscopic (AC and DC magnetization) and microscopic (neutrons, Mössbauer spectroscopy and EPR) probes have been used in the past twenty years to search for the above three transitions in real materials, and to determine their nature [1,2,3,4]. The ferromagnetic and freezing transitions at $T_c$ and $T_f$ are most easily observed through anomalies of the magnetic susceptibility [2,3,5]. Signatures of the transverse freezing transition at $T_{xy}$ have apparently been seen in Au$_{81}$Fe$_{19}$ via magnetic Bragg peak intensity [9,14] and EPR [11] measurements, in amorhous Fe$_{100-x}$Bi$_x$ and in (Fe$_{0.85}$Ni$_{0.15}$)$_{1-x}$Mn$_x$ [12] using Mössbauer spectroscopy, and in amorphous (Fe$_{1-x}$Mn$_x$)$_{75}$P$_{16}$B$_6$Al$_3$ (a$-$Fe$_{1-x}$Mn$_x$) using neutron depolarization, small angle neutron scattering and inelastic neutron scattering [9,15]. However, there is at present very little evidence for a clear signature of these three transitions as demonstrated via a single experimental technique on a given material. An exception may be the experiments mentioned above on Au$_{81}$Fe$_{19}$ [10,11], where three “features” that may be associated with $T_c$, $T_{xy}$ and $T_f$ have been seen. This raises the long standing and still main question at stake in the problem of reentrant spin glasses:

In real materials, as opposed to mean-field theory, are $T_{xy}$ and $T_f$ really two distinct transitions or thermodynamic features, or, are they the same one observed at different time and length scales?

To help resolve this issue, we have performed an extensive $\mu$SR study of the well known a$-$Fe$_{1-x}$Mn$_x$ reentrant ferromagnetic spin glass material [16]. $\mu$SR is an ideally suited technique to study the above question as it allows to extract simultaneously the amount of static spin order and to monitor the level of the spin dynamics. Specifically, we have searched for three well defined signatures of transitions at $T_c$, $T_{xy}$ and $T_f$ in the muon relaxation spectrum. Barsov et al. [13] performed earlier $\mu$SR study in Fe$_2$Ni$_{80-x}$Cr$_x$ and (Fe$_x$Mn$_{1-x}$)Pt$_3$ alloys. They found no signature of anomaly in dynamic muon spin relaxation rate $\lambda$ corresponding to $T_{xy}$ in the former system, while some broad feature which might be related to $T_{xy}$ has been seen in the latter. Their results were, however, not quite conclusive, due to very small variable range of $\lambda \leq 1.5 \mu s^{-1}$ in their data and to the lack of re-
sults for the contribution from the static depolarization rate.

In order to build a clear comparison between real systems and theory, we have focused our experimental efforts on an archetype reentrant ferromagnetic spin glass material, such as $a$-$Fe_{1-x}Mn_x$, which has been investigated by several other techniques [10]. The disorder-temperature phase diagram of $a$-$Fe_{1-x}Mn_x$ is shown in the inset panel of Fig. 1. We have investigated reentrant spin glass samples with concentration $x = 0.26$ and 0.30, and a strongly frustrated sample, $x = 0.41$, which shows full blown spin-glass behavior [6].

$$\lambda = \frac{\gamma A}{\mu}$$

The second term in square brackets for $A_s(t)$ describes the $\mu^+$ spin depolarization arising from the static local magnetic fields statistically not parallel to the incoming muon spin at the muon sites. Consequently, the long-time limit of the static relaxation function, $\lim_{t \to \infty} A_s(t) = 1/3$ for an amorphous ferromagnet, as occurs in a spin glass [4]. The probability distribution of local fields is characterized by a temperature, $T$, dependent RMS field value $\Delta(T) / \gamma \mu \gamma$, where $\gamma$ is the muon gyromagnetic ratio. $\Delta(T)$ is a measure of the average static electronic moment, $\bar{S}_i$, with $\Delta(T) \propto [ < \bar{S}_i >^2 ]_{T_1}$, where $< ... >_T$ is a Boltzman thermal average, and $[ ... ]$ is an average over the spin sites $i$. Hence, $\Delta(T)$ is zero in the paramagnetic phase, and increases as the thermal fluctuations diminish and the spins $\bar{S}_i$ become increasingly more static. $A_d(t)$ describes the fluctuating dynamical local field which leads to a muon spin depolarization at a rate $\lambda(T)$, where $\lambda(T)$ is an effective $1/T_1$ spin-lattice relaxation rate. $\alpha(T)$ and $\beta(T)$ are temperature dependent fitting parameters. To avoid a non-unique and an over-parametrization of our data, and to keep the physical interpretation of the data as transparent as possible, we analysed our ZF data with Eq.(1), which is qualitatively compatible with our $\mu$SR spectra for an amorphous ferromagnetic material devoid of muon spin precession signal. Below $T_c$, a strong depolarization is observed at early times due to the onset of static magnetic order. The ZF spectra were fitted with Eq. 1 using $\Delta(T)$, $\lambda(T)$, $\alpha(T)$ and $\beta(T)$ as fitting parameters.

Figures 1a) and 1b) show the temperature dependence of the dynamical relaxation rate, $\lambda(T)$, of the two reentrant samples, $x = 0.26$ and 0.30. We observe two clear peaks at temperatures that correspond reasonably well to the Curie temperature, $T_c$, and the onset of strong magnetic irreversibility temperature, $T_f$, respectively, as determined by magnetization measurements. For the spin glass sample, $x = 0.41$, there is a single peak in $\lambda$ whose temperature coincides with the spin-glass transition temperature $T_f$ found in magnetization measurements (Fig. 1c). The location of the peaks in $\lambda(T)$ for the three samples studied are reported in the phase diagram shown in Fig. 1.

Above $T_c$, the full asymmetry is relaxing dynamically. Below $T_c$, a static intradomain magnetization sets in, and at “long times”, only 1/3 of the asymmetry is re-
laxing with a dynamical rate $\lambda(T)$. Hence, a restricted fit to the long time tail of the asymmetry for $T < T_c$ reveals the behavior of $\lambda(T)$ in the most accurate way. By analogy with NMR relaxation in conventional magnets we attribute most of the observed spin relaxation below $T_c$ to muon spin-flip quasi-elastic scattering such as due to two-magnon Raman processes, for example. For the $x = 0.26$ and $x = 0.30$ reentrant samples, we have found that $\lambda(T)$ does not show any feature in a temperature range around the temperature where previous neutron experiments on $a$-$Fe_{1-x}Mn_x$ found an anomaly ascribed to a transverse spin freezing transition $[3]$. The peak in $\lambda$ seen at $T \approx T_f$, and the subsequent rapid decrease in the muon depolarization rate below $T_f$ indicate that there is a considerable slowing down of the spin dynamics as the system goes through $T_f$, which is then followed by a sizeable reduction in the density of low-lying excitations below $T_f$. Recall that the most naive interpretation for the transition at $T_f$ is that of a remnant of the Almeida-Thouless line in an Ising spin glass $[2]$. In this context, it is interesting that we see such a dramatic change of spin dynamics at a well characterized temperature for which mean-field theory predicts no transition $[2]$. This change in the spin dynamics is reminiscent of the anomalous behavior of the spin wave stiffness constant showing a minimum in inelastic neutron scattering experiments, and which is unexplained by theory $[16]$. Hence, both inelastic neutron scattering and the $\mu$SR results show a modification in the nature of the spin excitations below $T_f$ in $a$-$Fe_{1-x}Mn_x$. For the $x = 0.41$ spin glass sample (Fig. 1c), we have found that the dynamical exponent $\beta(T)$ reaches a value very close to $1/3$ right at $T = T_f$, as observed in recent $\mu$SR experiments on $AgMn$ and $AuFe$ dense spin glasses $[7]$.

Figures 2b) and 2c) show the temperature dependence of the static relaxation rate, $\Delta(T)$, for the reentrant samples, $x = 0.26$ and 0.30. We expect $\Delta$ to scale like the static RMS internal field for $T_{xy} < T < T_c$, $[2]$, with the latter itself proportional to the magnetization, $M$. The solid line in Fig. 2b) connecting the magnetization data (circles) shows the expected behavior of $\Delta(T)$ assuming $\Delta \propto M$, taking the magnetization, $M$, data from Ref. $[2]$, and fixing an overall proportionality factor using the values of $\Delta$ and $M$ at 130K. The results for $\Delta(T)$ and $M$ depart from each other at $T \approx T_c \approx 200K$ since $M$ was obtained in a field of 2000Oe, and this has the largest effect close to $T_c$. We also find that above 70K, $\Delta(T)$ scale proportionally to the internal longitudinal field, $B_{ND}$, deduced from neutron depolarization measurements $[3]$. However, $\Delta$ departs from the rescaled $M$ for a temperature close to and below $T = 70K$. This indicates an increase of the static spin order at $T < T_{xy} \approx 70K$. A comparison between magnetization data and hyperfine field in Mössbauer data for an $x = 0.235$ a-FeMn sample is shown in Fig. (2a). Overall, the behavior we observe in the static order parameter $\Delta$ is qualitatively very similar to what is observed in Mössbauer experiments $[8]$ of $a$-FeMn and other reentrant spin glasses, where the measured hyperfine field, $B_{Moss}$, starts deviating slowly from the bulk magnetization at $T_{xy}$. However, here in the $\mu$SR experiment, in contrast to most previous experimental probes used to study reentrant spin glasses, we monitor simultaneously both the static and dynamical behavior of the magnetic moments.

The above data for $\Delta(T)$ were determined by fitting the time dependence of the recovery of $G(t)$ from its minimum at a time $t_{\min} = (1 + \alpha)^{1/\alpha}/\Delta(T)$ towards the $1/3$ value. In $\mu$SR measurements, there is a “dead time” intrinsic to the experimental technique, which is of the order of 10ns, and for which there are no positron counts available. Setting this dead time to $t_{\min}$, we find that for $\alpha = 2$, the largest $\Delta$ we can extract reliably is of the order of 200$\mu$s$^{-1}$. To estimate the value of $\Delta(T = 4K)$ independently, we applied longitudinal fields, $H_{LF}$, and studied the field dependence of a decoupling ratio $r = A(t; T = 4K)/A(t; T = 270K)$ at a time $t \sim 0.05\mu$s, and fitted $r$ using the field-depenciency of the Kubo-Toyabe formula for Gaussian fields $[8]$. This is a reasonable procedure since we have $\Delta = 0$ at 270K, $1/\lambda(T = 270K) \gg (0.05\mu)s^{-1}$ and also $\lambda(T = 4K) \gg (0.05\mu)s^{-1}$. $r$ is given by

$$r = 1 - \frac{2\Delta^2}{\omega^2_0} + \frac{2\Delta^3}{\omega^3_0} \exp(-\frac{\omega^2_0}{2\Delta^2}) \times \int_0^{\omega_0/\Delta} \exp(u^2/2)du$$

In the analysis, we assumed that the internal field, and hence $\omega_0 = 0$ for an applied magnetic field, $H_{LF}, H_{LF} \leq$
Our results suggest that technique to study the static and dynamic spin behavior rate ∆. The sample was bolted in the sample holder to prevent it from reorienting while in the ferromagnetic phase and in strong \( H_{ LF} \). The results are shown in Fig. 3. Using \( D \) and ∆(\( T = 4K \)) as fitting parameters, we found \( D = 9.23kG \), a reasonable value for \( \mathrm{a-FeMn} \) for \( x = 0.26 \) for the geometry considered [1], and ∆(\( T = 4K \))/\( \gamma_\mu \) = 0.27T, giving ∆(\( T = 4K \)) = 230(\( \mu s \))–1, in reasonable agreement with the result obtained by fitting the time dependence of \( A(t) \). In principle, this procedure could be used to extract ∆(\( T \)) reliably for all \( T < T_c \), but would take a prohibitively long total beam-time. Hence, we confirm that ∆(\( T \rightarrow 0 \)) is above the apparent flattening of ∆ ≈ 180(\( \mu s \))–1 seen in the range 70K\(< T < 100K \), and that extra static that is not parallel to the longitudinal magnetization develops in the range 50< \( T < 100K \).

![FIG. 3. Longitudinal field dependence of the asymmetry ratio, \( A(T = 4K)/A(T = 270K) \). The fits are done using the decoupling Kubo-Toyabe Eq. (2) for the known demagnetization field \( D \approx 9.23kG \) for the geometry used in the \( \mu \)SR experiment, and for various choices of the static relaxation rate ∆.](image)

In conclusion, we have used the muon spin relaxation technique to study the static and dynamic spin behavior in the amorphous metallic \( \mathrm{FeMn} \) to search for three distinct transitions in reentrant spin glass materials. We have found a strong increase of the dynamic as well as static muon depolarization rate at the paramagnetic to ferromagnetic transition temperature, \( T_c \), and at \( T_f \) (\( T_f < T_c \)) that corresponds to the onset of strong bulk magnetic irreversibilities. However we observed no dynamical (critical slowing down) signature of a freezing of the transverse \( XY \) spin components at an intermediate temperature \( T_{xy} \) (\( T_f < T_{xy} < T_c \)). Despite the absence of critical dynamics at \( T_{xy} \), our results show unambiguously that in a reentrant spin glass, extra static moment which does not contribute to the magnetization, develops smoothly with no critical behavior within the time window of the probe at a temperature \( T_{xy} \) (\( T_f < T_{xy} < T_c \)). Our results suggest that \( T_f \) and \( T_{xy} \) are due to distinct thermodynamic “features” intrinsic to the system, and are not simply arising from a single transition observed at different time and length scales when comparing results obtained from different experimental techniques.

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