Muon spin resonance study of transverse spin freezing in \(a\text{-Fe}_x\text{Zr}_{100-x}\)

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\(\mu\)SR has been used to study magnetic ordering in the partially frustrated \(a\text{-Fe}_x\text{Zr}_{100-x}\) alloy system. We find clear evidence of two magnetic transitions in both the dynamic and static behavior of the muon polarization decay. The results are in perfect agreement with a description of the ordering in terms of a ferromagnetic phase transition at \(T_c\), followed by transverse spin freezing at \(T_{xy}\). We have confirmed the presence of a peak in the fluctuations at \(T_{xy}\) predicted by numerical simulations. This peak grows in strength and moves towards \(T_c\) with increasing frustration.

I. INTRODUCTION

The random addition of antiferromagnetic exchange interactions to an otherwise ferromagnetic (FM) Heisenberg spin system leads to a loss of FM order through the effects of exchange frustration. In extreme cases, a spin glass is formed with random isotropic spin freezing and neither net magnetization nor long-range order. At lower levels of frustration the system exhibits characteristics of both extremes as long-ranged FM order coexists with spin-glass (SG) order in the plane perpendicular to the FM order. On warming such a system from \(T=0\) K, the SG order first melts at \(T_{xy}\) followed by the loss of FM order at \(T_S\). This picture has emerged from mean-field calculations, numerical simulations, and experimental measurement. Despite the simplicity of this description, and the remarkable quantitative agreement between the numerical and experimental results, some issues remain unresolved. The most serious of these relates to differences between the theoretical and experimental procedures. The theoretical work is generally carried out in zero field, while experiments aiming to detect the freezing of transverse spin components at \(T_{xy}\) rely on a significant (typically 2–5 T) external field to define the FM ordering direction. An elegant demonstration of zero-field transverse spin freezing in AuFe (Ref. 6) that used the local electric field gradient to define the initial \((T>T_{xy})\) ordering direction, was overshadowed by the metallurgical instability of the Au-Fe system, but remains as perhaps the only zero-field evidence to date. The procedural differences leave open the possibility that the observation of spin components freezing perpendicular to the FM order at \(T_{xy}\) is due, at least in part, to the presence of the applied field used to orient the FM order. Indeed it has been claimed that the transverse components, and hence \(T_{xy}\), can be eliminated entirely in \(a\text{-Fe}_x\text{Zr}_{100-x}\) using an external field of \(<6\) T.

A second issue relates to magnetic inhomogeneity. It has been argued that the behavior of partially frustrated magnets is due not to frustration, but rather to the presence of magnetically isolated clusters embedded in the FM matrix. These clusters order at some lower temperature (i.e., \(T_{xy}\)) destroying the FM order and leading to noncollinearity. This idea can be traced back to the metallurgically unstable spin-glass alloys such as \(\text{AuFe}\) where a cluster description is natural. Even without appealing to clusters, the irreversibilities and noncollinear order that develop at \(T_{xy}\) are suggestive of spin glasses and have led to these systems being misnamed “re-entrant” spin glasses implying the ordering sequence: paramagnet→ferromagnet→spin glass. However, it has been shown that realistic models fail to yield reentrant behavior, and neutron depolarization has been used to confirm that there is no loss of FM order below \(T_{xy}\), even at the FM-SG boundary.

Finally, numerical simulations predict that although the freezing of transverse spin components does not represent a phase transition, it should be accompanied by significant, but noncritical, magnetic fluctuations. Unfortunately, a direct search for such fluctuations using ac susceptibility (\(\chi_{ac}\)) is complicated by the dominant response of the FM order. The FM response can be suppressed by using an applied field, and a field-dependent susceptibility peak at \(T_{xy}\) has been reported, but was not seen in a later study.

The work reported here addresses these three issues in several ways. First, by working with melt-spun metallic glasses, we are using extremely stable and uniform materials in which we have seen no change in \(T_c\) over a 10-year period in some samples. Second, frustration in the \(a\text{-Fe}_x\text{Zr}\) system can be tuned all of the way from FM to the FM-SG crossover in some samples. Third, \(\mu\)SR provides a local probe of static magnetic properties that can be used in zero applied field. Fourth, since the muons stop at random locations throughout the sample, they are sensitive to spatial variations.
in magnetic order and so can be used to detect magnetic inhomogeneities. Fifth, $\mu$SR is also sensitive to fluctuations in magnetic order, even in the presence of significant static order, and so it can be used to search for the fluctuation signature of transverse spin freezing, again in zero applied field.

We have been able to detect both $T_c$ and $T_{xy}$ in both the static and dynamic channels of the $\mu$SR data. We are able to rule out magnetic inhomogeneities as the ordering mechanism at $T_{xy}$. Our results confirm the presence of a fluctuation peak at $T_{xy}$ as predicted by numerical simulations. We find that the dynamic and static signatures of transverse spin freezing coincide, and conclude that there is a single magnetic transition below $T_c$ in these partially frustrated materials. These results are fully consistent with the predictions of numerical simulations.$^3$

II. EXPERIMENTAL METHODS

Meter-length ribbons 1–2 mm wide of $a$-$Fe_2Zr_{100-x}$ were prepared by arc melting appropriate amounts of the pure elements (Fe 99.97% and Zr 99.5%) under Ti-gettered argon, followed by melt spinning in 40 kPa helium with a wheel speed of 55 m/s. Sample compositions were checked by electron microprobe and found to be $\sim 0.1$ at. % Fe-rich of nominal in all cases. Cu $K\alpha$ x-ray diffraction on an automated powder diffractometer, and room temperature $^{57}$Fe Mössbauer spectra were used to confirm the absence of crystalline contamination. Basic magnetic characterization was carried out on a commercial extraction magnetometer (Quantum Design PPMs). $T_s$ and $\sigma_S$ were found to be consistent with standard values.$^4,16$

Zero-field $\mu$SR (ZF-$\mu$SR) measurements were made on the M13 beamline at TRIUMF. Sample temperature was controlled between 5 K and 300 K in a He-flow cryostat. Field-zero was set to better than 0.1 mT using a Hall probe and confirmed using the $\mu^+$ precession signal in a pure silver blank. Samples consisted of $\sim 15$ layers of 20-$\mu$m thick ribbons clamped between copper rings to give thicknesses of 170–200 mg cm$^{-2}$ over a 16 mm diameter active area. A pure silver (99.99%) mask$^{17}$ prevented stray muons from striking any of the mounting hardware. Essentially 100% spin polarized $\mu^+$ were implanted with their moments directed in the forward direction (i.e., along $z$). The subsequent decay $e^+$ is emitted preferentially along the moment direction. The time dependence of the $\mu^+$ polarization is conventionally followed by plotting the asymmetry ($A$) between scintillation detectors placed in the forward ($F$) and backward ($B$) directions relative to the initial $\mu^+$ flight direction $[A = (F-B)/(F+B)]$ as a function of time. Histograms containing 1–4 $\times 10^7$ events were acquired with timing resolutions of either 0.625 ns (well below $T_c$) or 1.25 ns (below $T_c$ to 300 K). The time-dependent asymmetry was then fitted using a nonlinear least-squares minimization routine to functional forms described below.

III. DATA ANALYSIS

Many excellent descriptions of ZF-$\mu$SR exist$^18$ so for the purposes of the analysis described here we note only the following. If a muon comes to rest at a site with a local field $B_z$, then the muon spin will precess about the field at the Larmor frequency $f_L = \gamma_BB_z$ where $\gamma_B = 135.5$ MHz/T is the gyromagnetic ratio of the muon. This precession leads to a periodic oscillation of the observed asymmetry and was used here to set the field at the sample to zero by observing the precession in a pure silver sample. The materials studied here are both structurally disordered (i.e., glassy) and magnetically disordered as a result of exchange frustration; therefore we expect a distribution of local fields to be present. For a system with an isotropic Gaussian distribution of static local fields, the asymmetry will follow the Kubo-Toyabe (KT) form:$^19$

$$G^G_\alpha(\Delta,t) = \frac{1}{3} + \frac{2}{3} [1 - (\Delta t)^2] \exp\left(-\frac{(\Delta t)^2}{2}\right),$$

where $\Delta/\gamma_B$ is the rms field. This function (see inset to Fig. 1) exhibits a minimum at $\Delta ti = \sqrt{3}$ then recovers to $\frac{1}{3}$ for long times. The asymptotic value reflects the fact that, on average, $\frac{1}{3}$ of the muons will have their moments parallel to the local field and therefore do not precess. Above $T_c$, there will be no magnetic order and hence no static field. However, the presence of neighboring moments that fluctuate in time will lead to a dephasing of the muon polarization by a process analogous to spin-lattice relaxation in NMR ($T_1$), and an exponential decay of the asymmetry:

$$A_d = A_0 \exp(-\lambda t),$$

is observed where $\lambda$ is an effective relaxation rate. In cases where both static order and fluctuations are present, the asymmetry decays according to the product of the two functions (as long as $f_L \gg \lambda$), i.e.,

$$A = A_d * G^G_\alpha.$$

The data in Fig. 1 illustrate a primary strength of $\mu$SR: static and dynamic magnetic effects can be observed simultaneously and they are sufficiently well separated in the data that they can be distinguished with great reliability. In Fig. 1, the static KT contribution is confined to the first 60 ns, while the dynamic decay is spread over the remaining 7 $\mu$s.

In order to fit our data, the two basic functions described above had to be modified. First, below $T_c$, we found that the

![FIG. 1. Typical $\mu$SR signal, with fit, observed below $T_c$ for $a$-$Fe_2Zr_8$. Inset shows the early-time region where the KT minimum characteristic of static order is visible. Note the clear separation in time-scales for the static and dynamic contributions.](Image 354x580 to 522x733)
dynamic decay departed significantly from a simple exponential form, and that a stretched exponential:

\[ A_d = A_o \exp[-(\lambda t)^\beta] \]

gave a far better description of the data. \( \beta \) was generally close to 1 above \( T_c \) but fell as low as 0.3 below \( T_c \).\(^{21}\) Such values for \( \beta \) probably reflect a logarithmic, rather than exponential decay implying hierarchically constrained dynamics,\(^{22}\) and suggesting that the presence of frustration affects the spin dynamics even above \( T_{xy} \), i.e., in the ferromagnetic phase. Second, the static contribution close to \( T_c \) had a KT minimum that was too shallow to be reproduced by the standard form. This behavior has been attributed to an excess of low-field sites (beyond that given by the assumed Gaussian distribution).\(^{23}\) We modeled the shallowing of the KT minimum by introducing a scaling power \( \alpha \) that serves to interpolate smoothly between the decay form expected for a Gaussian distribution of fields (\( \alpha = 2 \)) and that characteristic of a Lorentzian distribution (\( \alpha = 1 \)).\(^{24}\) The form used was

\[ G^O(\Delta, t) = \frac{1}{3} + \frac{2}{3} \left[ 1 - (\Delta t)^\alpha \right] \exp\left( -\frac{\Delta t}{\alpha} \right) \]

and \( \alpha \) was found to start close to 1 right at \( T_c \) but recovered to \( 1.8 \pm 0.2 \) within 30 K below \( T_c \). While it affects the detailed shape of the KT decay, \( \alpha \) has little effect on the value of \( \Delta \) derived from a given data set. Similar behavior was seen in FeNiCr, where it was fitted by summing Lorentzian and Gaussian contributions.\(^{25}\)

Finally, there are two instrumental effects set by the experimental geometry that do not appear in the theoretical models. These are the maximum asymmetry, \( A_o \), which sets the initial signal at \( t = 0 \), and the relative efficiency of the \( F \) and \( B \) detectors, \( \text{eff} \), which fixes the \( t = \infty \) background for fully depolarized muons. Both parameters were determined from data obtained close to but above \( T_c \), where we observed an exponential decay (with no static KT contribution) that is fast enough to be complete during the available 7 \( \mu \)s time window used. Once determined for a given sample, they were fixed for the entire data set. A maximum of four parameters were therefore varied in fitting the \( \mu \text{SR} \) data: \( \lambda \) and \( \beta \) describe dynamic effects and were always present, while \( \Delta \) and \( \alpha \) were included below \( T_c \) to reflect the presence of static order.

IV. RESULTS AND DISCUSSION

The dynamic relaxation rates shown for each alloy in Fig. 2 clearly illustrate the evolution from ferromagnet at \( x = 89 \) to spin glass at \( x = 93 \). \( T_c \) is marked by a clear cusp in \( \lambda(T) \) that moves down in temperature as the frustration level increases. At the same time, a broad feature develops at a much lower temperature for \( x \gtrsim 90 \). This peak both grows in amplitude and moves to higher temperatures with increasing \( x \) and hence frustration. Finally the two features merge at \( x = 93 \) as the system becomes a spin glass. These results are in perfect accord with both qualitative descriptions of transverse spin freezing\(^{4,16}\) and numerical simulations,\(^{3}\) which predicted a broad, noncritical, fluctuation peak at \( T_{xy} \) as the transverse spin components order. A second clear prediction of transverse spin freezing models is an increase in the net ordered moment below \( T_{xy} \) as the transverse components add to the ferromagnetic order established at \( T_c \). This effect is clearly visible in Fig. 3, where \( \Delta(T) \) is plotted for the five alloys. For \( 90 \leq x \leq 92 \) (i.e., those samples in which transverse spin freezing is observed) there is a distinct break in slope at the same temperature at which the lower temperature maximum is observed in \( \lambda(T) \). As expected, the size of this break increases as it moves to higher temperatures with increasing frustration. Furthermore, since \( \Delta \) increases on cooling through \( T_{xy} \), the local order must grow as the transverse components order. A similar increase in \( \langle B_{xy} \rangle \) is seen in Mössbauer spectra,\(^{4}\) while neutron depolarization\(^{11,12}\) shows that long-range FM order is not lost below \( T_{xy} \). We can therefore rule out any loss of order, and hence “re-entrant” behavior. We have fitted \( \Delta(T) \) using a combination of a modified Brillouin function\(^{26}\) with a linear term to allow for the additional increase associated with the ordering of the transverse spin components. This is the simplest function that reproduces the observed behavior, and in the absence of a detailed theoretical prediction for the expected form, it is sufficient to allow us to estimate the onset of transverse spin freezing.

The results of our analysis of \( \lambda(T) \) and \( \Delta(T) \) are summa-
FIG. 3. Temperature dependence of the static relaxation rate ($\Delta$) showing the steady reduction in ordering temperature with increasing frustration and the effects of transverse spin freezing for $90 \geq x \geq 92$. Solid lines are fits to a modified Brillouin function with a linear term to include ordering of transverse spin components.

FIG. 4. Magnetic phase diagram for $a$-Fe$_{93}$Zr$_{100-x}$ showing $T_c$ and $T_{xy}$ deduced from $\mu$SR data. Open circles mark values derived from the peak in $\lambda$, while the open triangles reflect values derived from fitting $\Delta(T)$. $T_{xy}$ is derived from $\chi_{ac}$ measurements on the same samples and are also shown as small open squares. Values obtained on an independently prepared series of alloys and measured using applied-field Mössbauer spectroscopy are shown for comparison as solid symbols (Ref. 4).

Comparison of the applied-field Mössbauer results$^4$ for $T_{xy}$ with the $\mu$SR values shown in Fig. 4 reveals a significant difference. The Mössbauer results are systematically lower, and the gap grows with increasing frustration. Since the onset of transverse spin freezing is clearly observed in the Mössbauer data, it is difficult to attribute the differences in derived $T_{xy}$’s to problems with the analysis. Indeed, the gap appears to grow as the signature gets stronger. We are forced to conclude that the field used in the Mössbauer measurements tends to lead to a stronger FM ordering and suppresses the onset of transverse spin freezing. The effect becomes more severe with increasing frustration and, in the case of $a$-Fe$_{93}$Zr$_7$, which is extremely close to $x_c$ where $T_c$ and $T_{xy}$ merge, the applied field leads to a 40% reduction in $T_{xy}$. The $\mu$SR data indicate that $a$-Fe$_{93}$Zr$_7$ is at the FM-SG crossover boundary, rather than well below it, as previously concluded from in-field measurements.$^4$ This revised conclusion is also more consistent with the observation of a very weak neutron depolarization signal in this alloy, indicating the absence of, or at best very small, domains.$^{11,12}$ We emphasize that moving $x_c$ from 94 at. % to 93 at. % reflects only a refinement of our understanding of this system; all of the observed phenomena remain in complete agreement with the predictions of transverse spin freezing models.

Inhomogeneous ordering (i.e., the presence of nonmagnetic clusters), is completely inconsistent with the $\mu$SR data. In cluster models, a significant volume of the sample does not order at $T_c$, but forms isolated, rapidly fluctuating clusters that freeze out around $T_{xy}$, causing a loss of long-ranged order. This loss of order below $T_{xy}$ has already been ruled out by neutron depolarization measurements.$^{11,12}$ but $\mu$SR now allows us to rule out the presence of nonordered clusters for $T_{xy} < T < T_c$. As the maximum asymmetry and detector efficiencies are determined above $T_c$, the $\frac{1}{2}$ point on the KT decay is completely defined. If some fraction of the $\mu$’s were to stop in sites with no static field, then the observed asymptote would lie above the expected $\frac{1}{2}$, and the fitted function would necessarily lie below the data. Since the static decay was typically complete within 60 ns even a modest offset between the fit and subsequent dynamic decay would be obvious. No such offset was observed in any of the data sets (see for example Fig. 1). When the model was extended to allow for a nonmagnetic component below $T_c$, there was no
improvement in the fit quality, and the fitted magnetic fraction was 0.95–1.05 in all cases below $T_c$. In all samples, the asymptotic value was attained immediately below $T_c$ and no changes were observed on subsequent cooling. Our analysis indicates that less than 3% of the sample volume could be present as nonordered clusters below $T_c$. In addition, the behavior of $\Delta(T)$ below $T_{xy}$ is also inconsistent with ordering of clusters. If the moments in the clusters were similar in magnitude to those in the matrix, then there would be no improvement in the fits as the entire volume of the sample exhibited static order. The KT asymptote would simply move to $1$, the fits as the entire volume of the sample exhibited static order. The KT asymptote would simply move to $\frac{1}{2}$. For the increase in $\Delta$ at $T_{xy}$ to be due to ordering of clusters, the local moments in those clusters must be much larger than those in the matrix (about a factor of 2 larger for $x=92$ in Fig. 3), and the clusters must occupy a significant volume fraction of the sample for their ordering to dominate below $T_{xy}$. The ordering of a significant volume fraction of much larger moments would have a profound and unmistakable effect on the Mössbauer spectra of these alloys that is simply not observed. Furthermore, the early-time fit following the KT decay would be extremely poor if the bulk of the sample were still not ordered below $T_c$.

In conclusion, $\mu$SR provides clear evidence of two, and only two, magnetic transitions in iron-rich $a$-Fe-Zr alloys. The two transitions are observed in both the dynamic and static behavior of the muon polarization decay. The results are in perfect agreement with the description of the ordering in terms of a FM-phase transition followed by transverse spin freezing. We have confirmed the presence of a peak in the fluctuations at $T_{xy}$ predicted by numerical simulations.

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17. We confirmed that the silver used gave no time-dependent $\mu$SR signal down to 5 K.
21. $\beta$ was largely constant well below $T_c$ and did not affect the determination of $T_{xy}$.