Transverse spin freezing in *a*-$Fe_xZr_{100_x}$studied using muon spin relaxation

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Analysis of the muon spin relaxation (μSR) signal from a range of a-Fe_xZr_{100-x} alloys shows that the entire sample orders at T_c and that the volume fraction that could be associated with isolated nonmagnetic clusters is less than 3%. Furthermore, the coincidence between the static and dynamic μSR signatures of transverse spin freezing provides a clear confirmation of predictions from numerical models. [S0021-8979(00)48808-0]

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 (SG) 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 SG order in the plane perpendicular to the FM order.[1] 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_c. This picture has emerged from mean field calculations,[2] numerical simulations,[3] and experimental measurement.[4]

Despite the simplicity of this description, and the remarkable quantitative agreement between the numerical and experimental results,[4] some issues remain unresolved. The most serious, 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.[4,5] A second issue relates to magnetic inhomogeneity. It has been argued that the behavior of partially frustrated magnets is due not to frustration, but 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.[6] Finally, numerical simulations predict that the freezing of transverse spin components should be accompanied by significant, but noncritical, magnetic fluctuations. A direct search for such fluctuations in χ_{ac} is complicated by the dominant response of the FM order.[7]

Muon spin relaxation (μSR) can be used to probe magnetic systems in much the same way as Mössbauer spectroscopy, except that the muon probes the transferred field in the time domain, rather than energy. This difference makes μSR particularly sensitive to both static order and magnetic fluctuations. Zero-field (ZF) μSR used here, allows us to probe changes in magnetic order with no field applied, closer to the conditions used in theoretical models of partially frustrated magnets. Furthermore, since the muons are implanted at random locations throughout the sample, they are sensitive to spatial variations in magnetic order.

II. EXPERIMENTAL METHODS

Meter-length ribbons 1–2 mm wide of a-Fe_xZr_{100-x} were prepared by arc melting followed by melt spinning. Sample compositions were checked by electron microprobe. Cu K_α x-ray diffraction and room temperature ^{57}Fe Mössbauer spectra were used to confirm the absence of crystalline contamination. Basic magnetic characterization was carried out on a Quantum Design Physical Properties Measurement System. T_c and σ_s were found to be consistent with standard values.[4,8]

ZF–μSR measurements were made on the M13 beamline at TRIUMF. Sample temperature was controlled between 5 and 300 K in a conventional He-flow cryostat. Field-zero was set to better than 0.01 mT using a Hall probe and confirmed by the μ^+ precession signal in pure silver. Samples were $\sim$200 mg cm^{-2} thick and 16 mm in diameter. The time dependence of the μ^+ spin polarization was followed by plotting the asymmetry (A) between detectors placed in the forward (F) and backward (B) directions rela-

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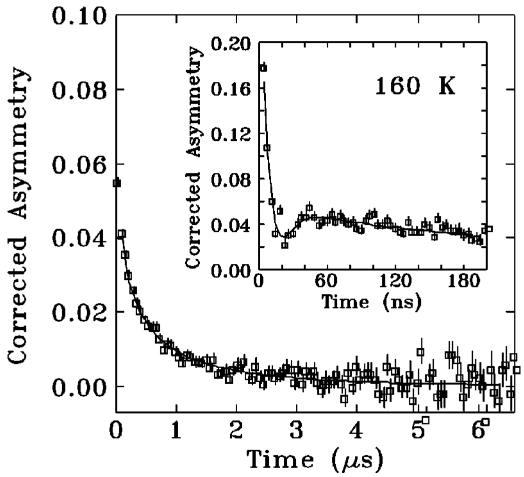

FIG. 1. Typical μSR signal, with fit, observed below T_c for a-$Fe_{92}Zr_8$. Inset shows the early-time region where the K–T minimum characteristic of static order is visible.

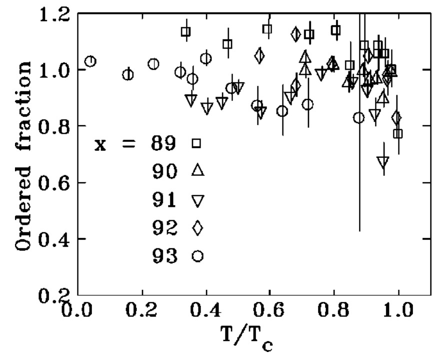

FIG. 2. Temperature dependence of the ordered magnetic fraction for the five a-Fe_xZr_{100-x} alloys studied here.

tive to the initial μ^+ flight direction ($A=F-B/F+B$) as a function of time. $A(T)$ was fitted using a conventional nonlinear least-squares minimization routine.

III. RESULTS AND DISCUSSION

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 distribution of static local fields, the asymmetry, $A(T)$, will follow the Kubo–Toyabe (K–T) form[9]

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

where Δ/γ_μ is the rms field. This function (see inset to Fig. 1) exhibits a minimum at $\Delta t=\sqrt{3}$ then recovers to $\frac{1}{3}$ for long times. We modeled the shallowing of the K–T minimum observed near T_c by introducing a scaling power α that serves to smoothly interpolate between the form expected for a Gaussian distribution of fields ($\alpha=2$) and that from a Lorentzian distribution ($\alpha=1$).[10] α was found to start close to 1 right at T_c but recovered to 1.8 ± 0.2 within 30 K below T_c. Similar behavior has been seen in FeNiCr, where it was fitted by summing Lorentzian and Gaussian contributions.[11] Above T_c, there will be no magnetic order and, hence, no static field. However the presence of neighboring moments that fluctuate in time leads to a dephasing of the muon polarization by a process analogous to spin-lattice relaxation in nuclear magnetic resonance (T_1) and an exponential decay of the asymmetry is observed:

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

where λ is an effective relaxation rate. This stretched exponential gave an excellent fit to the data. β was generally close to 1 above T_c but fell as low as 0.3 immediately below T_c. 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.,

$$A=A_dG_z^G.$$

The data in Fig. 1 illustrate a primary strength of ZF–μSR: static and dynamic magnetic effects can be observed simultaneously and are well separated in the data. In Fig. 1, the static K–T contribution is confined to the first 60 ns, while the dynamic decay is spread over the remaining 7 μs. A maximum of four parameters were generally varied in fitting the μSR data: λ and β describe dynamic effects and were always present, while Δ and α were included below T_c to reflect the presence of static order.

Inhomogeneous ordering, (i.e., the presence of clusters), is completely inconsistent with the μSR 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,[12] but μSR now allows us to rule out the presence of nonordered clusters for $T_{xy}<T<T_c$. If some fraction of the muons stopped in sites with no static field, then the K–T asymptote would lie above the $\frac{1}{3}$ expected, and 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. When the model was extended to allow for a nonmagnetic component, there was no improvement in the fit quality, and the fitted magnetic fraction was 0.95–1.05 in all cases below T_c (see Fig. 2). 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.

The dynamic relaxation rate for $x=92$ derived from fits in which the ordered component was fixed at 1.0 is shown in Fig. 3. There are clearly two peaks in the fluctuation rate. The upper one coincides with T_c determined by ac susceptibility, and is also associated with the onset of static order in the μSR data as can be seen from the plot of Δ on the same figure. The lower temperature peak in λ is also associated with an increase in Δ and we attribute this feature to transverse spin freezing. We fitted $\Delta(T)$ using a combination of a modified Brillouin function combined with a linear term to allow for the increase associated with ordering of transverse spin components. This signature is in perfect accord with

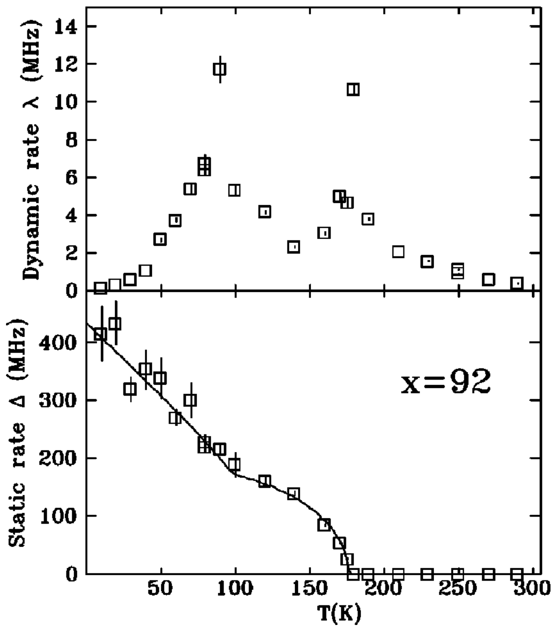

FIG. 3. Results of fitting μSR data for a-$Fe_{92}Zr_8$. Top: dynamic relaxation rate (λ), bottom: static relaxation rate (Δ). Solid line is a fit to a modified Brillouin function. Note that the onset of magnetic order and the abrupt increase in Δ both coincide with peaks in λ.

qualitative descriptions of transverse spin freezing,[4,8] and is precisely that expected from numerical modeling:[3] an increase in static order accompanied by a peak in the fluctuation rate. The second transition was observed for $x=90$, 91, and 92, and in each case, there is a distinct break in slope in $\Delta(T)$ at the same temperature at which a maximum is observed in $\lambda(T)$. These results demonstrate that there is single magnetic event below T_c in partially frustrated magnets, and that it is associated with an increase in static ordered moment, and accompanied by a peak in the fluctuation rate.

We saw only a single peak (at T_c) for $x=89$ as the frustration effects are sufficiently weak that T_{xy} lies below 5 K, or more likely is absent entirely. Remarkably, only a single event was observed at $x=93$ suggesting that this composition is actually a SG, in contradiction of previous results that indicated that the critical composition for the FM–SG crossover lay at $x\sim 94$.[4,5] These results are summarized in Fig. 4. A similar study of site-frustrated a-$(Fe_{0.74}Mn_{0.26})_{75}P_{16}B_6Al_3$ did not find agreement between the static and dynamic estimates of T_{xy} and concluded that there was an additional transition in this system.[13] However, our numerical simulations of site-frustrated systems predict essentially the same behavior as seen here: two transitions, FM followed by transverse spin freezing.[14] The origin of this discrepancy remains unclear.

The observations below T_{xy} are also inconsistent with ordering of clusters. If the moments in the clusters are similar in magnitude to those in the matrix, then there should be no change in Δ at T_{xy}, merely an improvement in the fits as the K–T asymptote moved to $\frac{1}{3}$. If we assume that the increase in Δ at T_{xy} is in fact due to ordering of clusters, then the local moments in those clusters must be *much* larger than

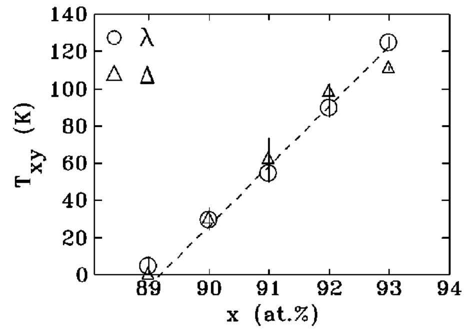

FIG. 4. T_{xy} deduced from μSR data on a-Fe_xZr_{100-x}.

those in the matrix (about a factor of two 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.[1,4,5] Furthermore, the early-time fit following the K–T decay would be extremely poor if the bulk of the sample was still not ordered below T_c.

In conclusion, μSR provides clear evidence of 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. The presence of nonordered clusters below T_c can be ruled out.

ACKNOWLEDGMENTS

This work was supported by grants from the Natural Sciences and Engineering Research Council of Canada, Fonds pour la Formation de Chercheurs et l'Aide à la Recherche, Québec, and the Australian Research Council.

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