μSR study of transverse spin freezing in a$-Fe_xZr_{100-x}$

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μSR has been used to study magnetic ordering in the partially frustrated a$-$Fe$_x$Zr$_{100-x}$ alloy system. We find clear evidence of two magnetic transitions in both the dynamic and static behaviour of the muon polarisation 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.

76.75.+i 75.50.Bb 75.50.Lk 75.50.Kj

I. INTRODUCTION

The random addition of antiferromagnetic (AF) 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 magnetisation nor long range order. At lower levels of frustration the system exhibits characteristics of both extremes as long-ranged FM order co-exists 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 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 Tesla) external field to define the FM ordering direction[4,5]. An elegant demonstration of zero-field transverse spin freezing in **Au**Fe[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$-$Fe$_x$Zr$_{100-x}$ using an external field of <6 T[7].

A second issue relates to magnetic inhomogeneity. It has been argued that the behaviour 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 non-collinearity[8]. This idea can be traced back to the metallurgically unstable spin-glass alloys such as **Au**Fe where a cluster description is natural[9]. Even without appealing to clusters, the irreversibilities and non-collinear order that develop at T$_{xy}$ are suggestive of spin glasses and have led to these systems being mis-named "re-entrant" spin glasses implying the ordering sequence: paramagnet $\rightarrow$ ferromagnet $\rightarrow$ spin-glass. However, it has been shown that realistic models fail to yield re-entrant behaviour[3,10], and neutron depolarisation has been used to confirm that there is no loss of FM order below T$_{xy}$[11], even at the FM–SG boundary[12].

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 non-critical, magnetic fluctuations. Unfortunately, a direct search for such fluctuations using *ac*-susceptibility (χ_{ac}) is complicated by the dominant response of the FM order[13]. The FM response can be suppressed by using an applied field, and a field-dependent susceptibility peak at T$_{xy}$ has been reported[14], but was not seen in a later study[15].

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 $a-Fe-Zr$ system can be tuned all of the way from FM to the FM–SG crossover boundary at a critical composition of $x_c \sim 93$ at.% by simply varying the iron content. Third, μSR 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, μSR 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 μSR 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$_x$Zr$_{100-x}$ 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 characterisation was carried out on a commercial extraction magnetometer (Quantum Design PPMS). T$_c$ and σ_S were found to be consistent with standard values[4,16].

Zero-field μSR 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 μ^+ precession signal in a pure silver blank. Samples consisted of $\sim$15 layers of 20μm 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 polarised μ^+ 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 μ^+ polarisation is conventionally followed by plotting the asymmetry (A) between scintillation detectors placed in the forward (F) and backward (B) directions relative to the initial μ^+ flight direction ($A = (F - B)/(F + B)$) as a funtion 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 non-linear least-squares minimisation routine to functional forms described below.

III. DATA ANALYSIS

Many excellent descriptions of ZF-μSR 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_x, then the muon spin will precess about the field at the Larmor frequency $f_L = B_x \gamma_\mu$ where γ_μ = 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 (K-T) form[19]:

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

where Δ/γ_μ is the rms field. This function (see inset to Fig. 1) exhibits a minimum at $\Delta \times t = \sqrt{3}$ 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 neighbouring moments that fluctuate in time will lead to a dephasing of the muon polarisation by a process analogous to spin-lattice relaxation in NMR (T$_1$), and an exponential decay of the asymmetry:

$$A_d = A_o \exp(-\lambda t)$$

is observed where λ 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 $f_L \gg \lambda$[20]) *i.e.*

$$A = A_d * G_z^G.$$

The data in Fig. 1 illustrate a primary strength of μSR: static and dynamic magnetic effects can be observed simulaneously and they are sufficiently well-separated in the data that they can be distinguished with great reliability. 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.

In order to fit our data, the two basic functions described above had to be modified. First, below T$_c$, we found that the dynamic decay departed significantly from a simple exponential form, and that a stretched exponential:

$$A_d = A_o \exp\left(-(\lambda t)^{\beta}\right)$$

gave a far better description of the data. β was generally close to 1 above T$_c$ but fell as low as 0.3 below T$_c$[21]. Such values for β 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 K-T minimum that was too shallow to be reproduced by the standard form. This behaviour has been attributed to an excess of low-field sites (beyond that given by the assumed Gaussian distribution)[23]. We modelled the shallowing of the K-T minimum by introducing a scaling power α 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:

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

and α was found to start close to 1 right at T$_c$ but recovered to 1.8 ± 0.2 within 30 K below T$_c$. While it affects the detailed shape of the K-T decay, α has little effect on the value of Δ derived from a given data set. Similar behaviour 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, eff, which fixes the $t = \infty$ background for fully depolarised muons. Both parameters were determined from data obtained close to but above T$_c$ where we observed an exponential decay (with no static K-T contribution) that is fast enough to be complete during the available 7 μs 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 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.

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 λ(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 $x \geq 90$. 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, non-critical, 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 Δ(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 λ(T). As expected, the size of this break increases as it moves to higher temperatures with increasing frustration. Furthermore, since Δ *increases* on cooling through T$_{xy}$, the local order must grow as the transverse components order. A similar increase in $< B_{hf} >$ is seen in Mössbauer spectra[4], while neutron depolarisation[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" behaviour. We have fitted Δ(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 behaviour, 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 λ(T) and Δ(T) are summarised as a phase diagram (Fig. 4). The agreement between the static and dynamic μSR signatures and also χ_{ac} data confirms that we are indeed detecting the onset of order at T$_c$. Furthermore, λ(T) and Δ(T) also yield the same value for T$_{xy}$ in each case (the average deviation is less than 5 K), clearly demonstrating that the lower fluctuation peak is also associated with changes in the static order, as predicted by numerical simulations[3]. In all cases however, T$_{xy}$ is more easily identified from the peak in λ than from the break in Δ(T), underlining the benefits of using μSR to study transverse spin freezing. A similar study of site-frustrated a$-(Fe_{74}Mn_{26})_{75}P_{16}B_6Al_3$ did not find agreement between the static and dynamic estimates of T$_{xy}$ and concluded that there was an additional transition in this system[27]. However, our numerical simulations of site-frustrated systems predict essentially the same behaviour as seen here: two transitions, FM followed by transverse spin freezing[28]. The origin of this discrepancy remains unclear.

Comparison of the applied-field Mössbauer results[4] for T_{xy} with the μSR 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 μSR 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 depolarisation signal in this alloy, indicating the absence of, or at best very small, domains[11,12]. We emphasise 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 non-magnetic 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 depolarisation measurements[11,12], but μSR now allows us to rule out the presence of non-ordered clusters for $T_{xy} < T < T_c$. As the maximum asymmetry and detector efficiencies are determined above T_c, the $\frac{1}{3}$ point on the K–T decay is completely defined. If some fraction of the μ^+ were to stop in sites with no static field, then the observed asymptote would lie above the expected $\frac{1}{3}$, 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 non-magnetic 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[29]. 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 non-ordered clusters below T_c. In addition, the behaviour 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 change in the value of Δ at T_{xy}, merely an improvement in the fits as the entire volume of the sample exhibited static order. The K–T asymptote would simply move to $\frac{1}{3}$. For the increase in Δ 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 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 were still not ordered below T_c.

In conclusion, μSR 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 behaviour of the muon polarisation 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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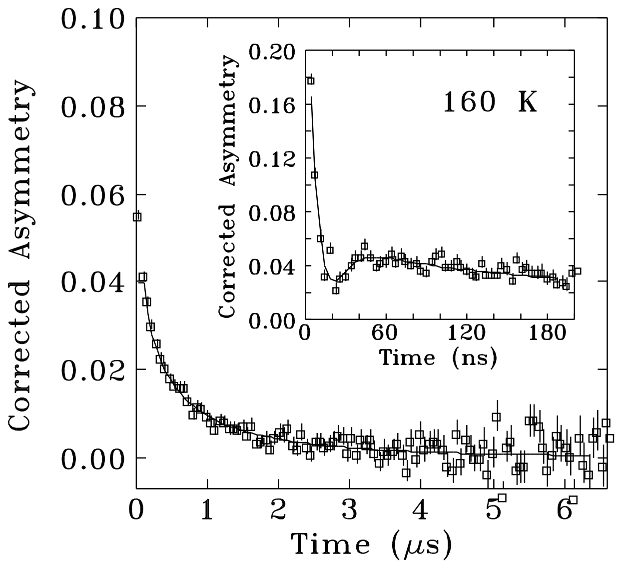

FIG. 1. Typical μSR signal, with fit, observed below T_c for a$-Fe_{92}Zr_8$. Inset shows the early-time region where the K-T minimum characteristic of static order is visible. Note the clear separation in time-scales for the static and dynamic contributions.

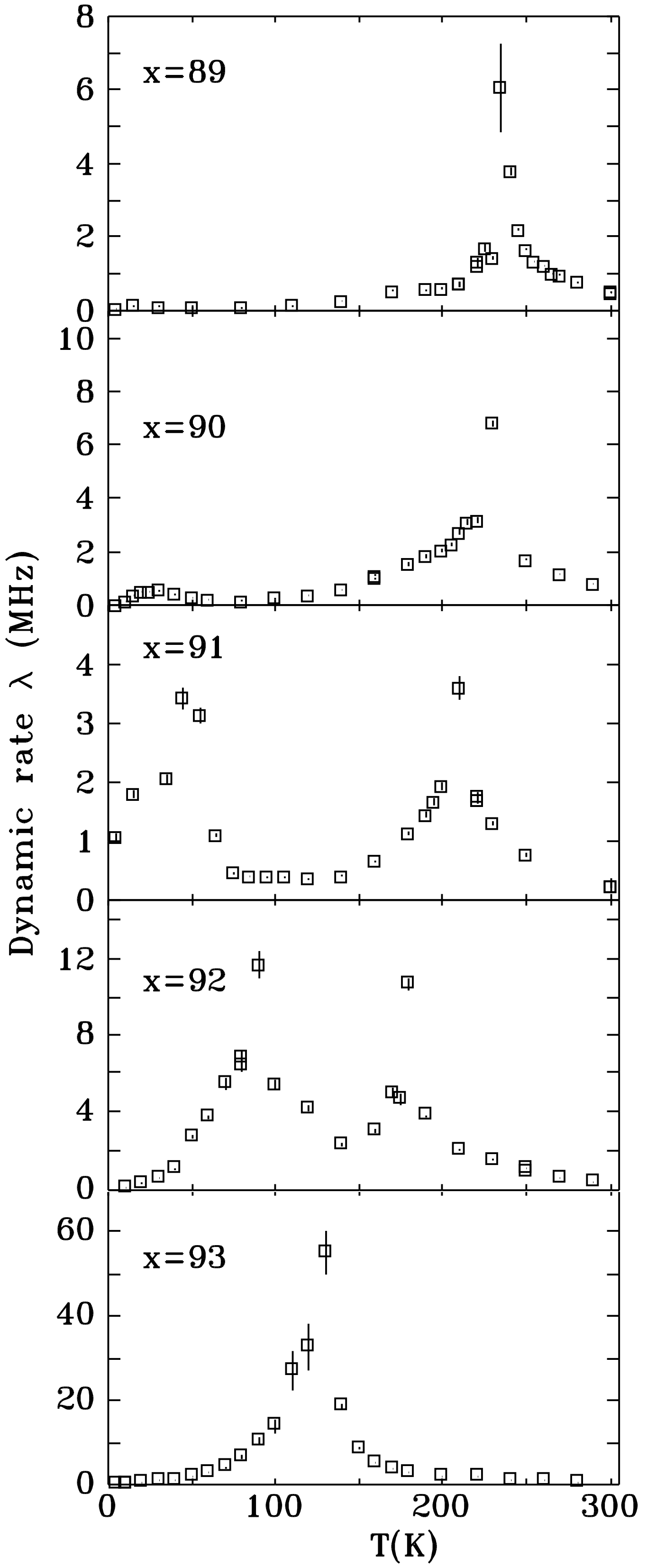

FIG. 2. Temperature dependence of the dynamic relaxation rate (λ) showing the high temperature cusp (T_c) merging with the lower temperature feature (T_{xy}) with increasing frustration.

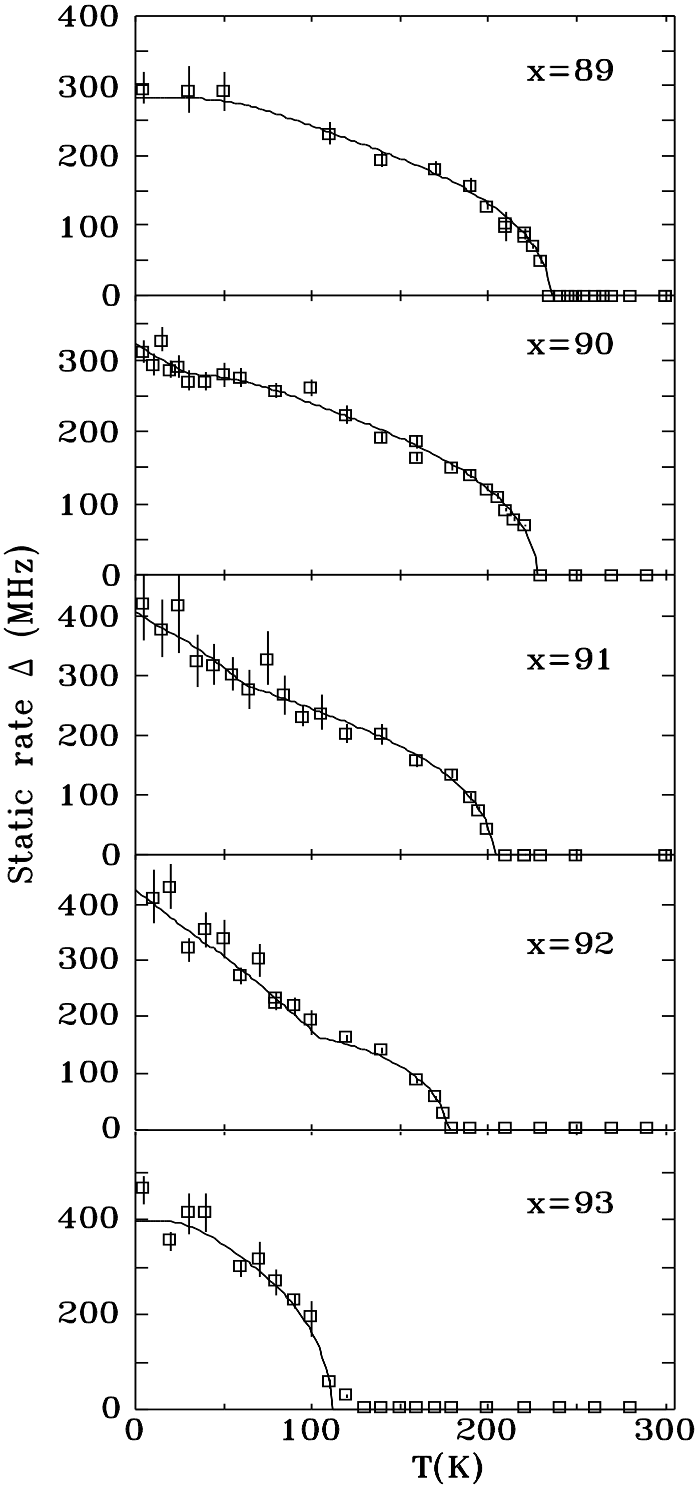

FIG. 3. Temperature dependence of the static relaxation rate (Δ) showing the steady reduction in ordering temperature with increasing frustration and the effects of transverse spin freezing for $90 \geq x \geq 92$. Solid lines are fits to a modified Brillouin function with a linear term to include ordering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

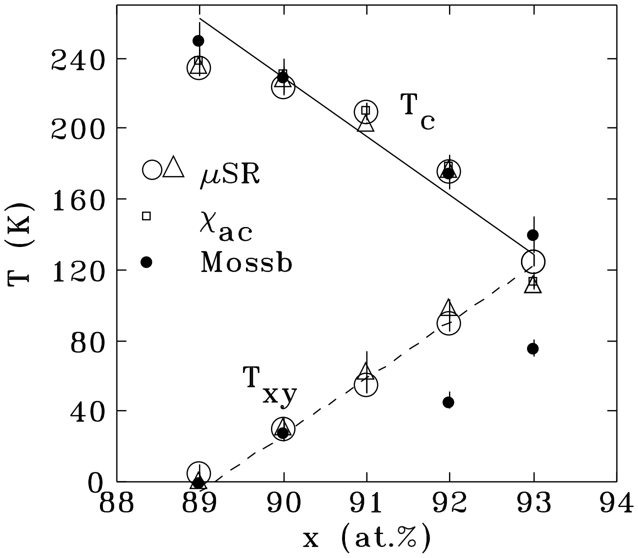

FIG. 4. Magnetic phase diagram for a$-$Fe$_x$Zr$_{100-x}$ showing T$_c$ and T$_{xy}$ deduced from μSR data. Open circles mark values derived from the peak in λ, while the open triangles reflect values derived from fitting Δ(T). T$_c$s derived from χ_{ac} measurements on the same samples are also shown 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[4].