Transverse spin freezing in Ruthenium-doped a$-Fe_{90}Zr_{10}$

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Muon spin relaxation (μSR) has been used to study the ferromagnet to spin glass crossover in a$-Fe_{90-x}Ru_xZr_{10}$. We find clear evidence of two magnetic transitions in both the dynamic and static behaviour of the muon polarisation decay: a ferromagnetic phase transition at T_c, followed by transverse spin freezing at T_{xy}. Comparison with earlier Mössbauer data shows that the break in $B_{hf}(T)$ is indeed due to transverse spin freezing, and that both Mössbauer spectroscopy and μSR yield the same values for T_{xy}.

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

I. INTRODUCTION

Of all transition metals (TM) surveyed, ruthenium leads to the most rapid destruction of ferromagnetic order in a$-Fe_{90-x}TM_xZr_{10}$ alloys[1]. Ruthenium is non-magnetic, and forms a stable ternary a-Fe-Ru-Zr glass over a very wide composition range[2]; it therefore provides an ideal system in which to study the ferromagnet (FM) to spin glass (SG) crossover. Mössbauer[3] and neutron depolarisation[4] measurements have shown that there is a complete loss of long-range FM order by a critical doping level of $x_c \sim 2.5$. Since the loss of order in the binary a-Fe-Zr system is due to the development of exchange frustration[5], we would expect the same mechanism to be active in the Ru-doped system and therefore transverse spin freezing should occur below T_c in those alloys that are close to the FM–SG crossover[6].

The defining characteristic of transverse spin freezing is the appearance of SG order at a well defined temperature (T_{xy}), and in the plane perpendicular to the axis defined by the existing FM order. The SG order is in addition to, and co-exists with, the FM order throughout the material. The local moment increases, but the system becomes non-collinear and so the magnetisation is unaffected. This behaviour is most easily observed using Mössbauer spectroscopy in an applied field of several Tesla where spin components are seen to develop perpendicular to the applied field on cooling through T_{xy}[5]. An alternative signature has been provided by numerical simulations[7], which showed 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. This prediction was recently confirmed by μSR measurements on binary a$-Fe_xZr_{100-x}$ alloys where the dynamic signature of transverse spin freezing was clearly observed and shown to coincide with an increase in the static order[8]. The systematic deviation between the applied field Mössbauer values for T_{xy} and those derived from zero-field μSR indicates that the system is severely perturbed by an applied field and emphasises the need to use zero-field methods to follow the ordering at T_{xy}.

Doping a$-Fe_{90}Zr_{10}$ with ruthenium leads to a rapid reduction in T_c and magnetization while the local Fe moment is largely unaffected[1,4]. Behaviour that suggests increasing frustration. However, only indirect evidence has been presented for transverse spin freezing in a$-Fe_{90-x}Ru_xZr_{10}$. The temperature dependence of the average hyperfine field (B_{hf}) shows a break in slope well below T_c, at a temperature labelled T_{br}. The composition dependence of T_{br} is consistent with the expected behaviour of T_{xy}[4]. Such a break in slope could be associated with the increase in local moment that occurs when the transverse spin components order; however we observed no such break in the binary a-Fe-Zr system[5]. By contrast, the a$-(Fe_{1-x}Mn_x)_{78}Sn_2Si_6B_{14}$ system exhibits a striking break in slope at T_{xy}[9].

In order to confirm that transverse spin freezing does indeed occur, and to compare the static and dynamic behaviour in this system with that observed in the binary a-Fe-Zr system, we have made μSR measurements through the FM–SG crossover region in a$-Fe_{90-x}Ru_xZr_{10}$. We show that the dynamic and static signatures of transverse spin freezing are present, and coincide in temperature. The results are also compared with zero field Mössbauer data and confirm that the break in $B_{hf}(T)$ is associated with transverse spin freezing.

II. EXPERIMENTAL METHODS

Ribbons of a$-$Fe$_{90-x}$Ru$_x$Zr$_{10}$ were prepared by arc-melting followed by melt-spinning[4]. Cu K$_\alpha$ x-ray diffraction 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 susceptometer/magnetometer. Sample compositions were checked by electron microprobe analysis and some loss of ruthenium (typically 0.1 – 0.4 at.%) was noted. All compositions quoted here are actual, rather than nominal values.

Mössbauer measurements were obtained on a constant-acceleration spectrometer using a 1 GBq ^{57}Co**Rh** source. Temperatures between 12 K and room temperature were achieved using a vibration-isolated closed-cycle refrigerator. Spectra were fitted using Window's method[10].

Zero-field μSR measurements were made on the M20 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 were 170–200 mg cm^{-2} thick over a 16 mm diameter active area. Histograms containing 1–4 $\times 10^7$ events were acquired with a timing resolution of 0.8 ns.

As before[8], data above T$_c$ were fitted using a stretched exponential:

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

while below T$_c$ the presence of static order required the inclusion of a Kubo-Toyabe (K–T) function[12]:

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

and the data ws fitted as the product:

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

a procedure that is valid as long as $f_L \gg \lambda$ Ref.[13].

III. RESULTS AND DISCUSSION

Typical μSR patterns for a$-$Fe$_{89.2}$Ru$_{0.8}$Zr$_{10}$ are shown in Fig. 1 at 220 K (above T$_c$), 180 K (below T$_c$ but above T$_{xy}$) and 45 K (below T$_{xy}$). Above T$_c$ there is no static order, but magnetic fluctuations couple to the muon spin and cause an exponential decay in the observed polarisation. The inset shows that the exponential continues to the earliest time measured ($\sim$1.5 ns), and no static contribution is apparent. Below T$_c$, fluctuations (e.g. magnons) continue to depolarise the muons, but the characteristic K–T minimum is observed at early times (inset), indicating the presence of static magnetic order. Finally, below T$_{xy}$, fluctuations are still present and the K–T minimum due to static order has moved to still earlier time, indicating an increase in the local field at the muon sites. The static relaxation rate at 45 K is 260 MHz, however it is clear from the inset to Fig. 1, that our resolution is more than adequate to provide reliable values at far greater relaxation rates. Fig. 1 also serves to demonstrate a major advantage of the zero-field μSR technique: static and dynamic effects can both be studied in a single measurement, but their signatures in the data are completely independent, and therefore do not interfere. The static contributions in Fig. 1 are complete by 40 ns, whereas the dynamic exponential decay extends across the remaining 4 μs.

Values for static relaxation rates in the samples studied here are shown in Fig. 2. The most immediate observation is that as the ruthenium content, x, is increased, the temperature at which static magnetic order develops moves down rapidly. A second feature is apparent below T$_c$ in most of the curves: there is a distinct break in the temperature dependence for $x \leq 2.0$. Δ(T) was fitted using the sum of two Brillouin functions (modified to allow for a distribution of exchange[14]) each with different onset temperatures and saturation values. These fits are shown as solid lines in Fig. 2. This behaviour is consistent with transverse spin freezing, and matches that seen previously in a $-$ Fe $-$ Zr[8].

Localising a break in slope is not always easy, nor does it provide a convincing demonstration that a significant change in magnetic order has occurred. The behaviour of the dynamic relaxation rate, also shown in Fig. 2, is far clearer. Above T$_c$, the muons are depolarised by dynamic fluctuations in the local magnetisation. These fluctuations diverge at T$_c$. We found that a simple Curie-Weiss function ($\lambda \propto (\mathrm{T}-\mathrm{T_c})^{-1}$) gave a good description of λ(T) as we approached T$_c$ from above. This served to locate T$_c$, however no attempt was made to determine the critical exponent, nor were the data adequate for such an analysis. Below T$_c$ magnon–muon interactions serve to depolarise the muons. As the temperature is lowered, these freeze out and λ should fall to zero. This was observed in a$-$Fe$_{89}$Zr$_{11}$[8], which does not exhibit transverse spin freezing, and was only seen here for $x > 3$ where we are fully in the spin glass regime and transverse spin freezing is also absent. For the other samples, the steady decline in λ(T) is interrupted by a broad peak (right-hand side of Fig. 2). This peak was most easily fitted as a Gaussian with a width of $\sim$20–25 K. This peak clearly reflects a substantial increase in the fluctuation rate (at least two orders of magnitude above the background in the equivalent region in a$-$Fe$_{89}$Zr$_{11}$[8]), however it is equally clearly not divergent. This observation is in striking agreement with the predictions of numerical simulation work[7], where T$_{xy}$ was shown to be associated with a *non-critical* peak in the fluctuation rate.

Transition temperatures are summarised in the phase diagram shown as Fig. 3. The basic pattern is exactly that expected for a partially frustrated magnetic system exhibiting transverse spin freezing[5–7,15]. T$_c$ falls, and

T_{xy} rises to meet it as the frustration level (in this case controlled by the ruthenium content) is increased. One key result is the close agreement between the transition temperatures derived from the static and dynamic effects in the μSR signal. Both T_c and T_{xy} are clearly associated with fluctuations and with a marked change in the local, static order. There is no evidence for further transitions below T_{xy}. There is however, one interesting difference between the theoretical predictions and the phase diagram presented in Fig. 3. T_c(x) and T_{xy}(x) are usually shown as having a constant slope until they meet, then the spin glass transition T_{sg}(x) continues horizontally[5–7,15]. Here we see that T_{xy}(x) levels off and T_c(x) drops abruptly by nearly a factor of two in the last 0.5 at.% to join it. This observation indicates that the region close to the FM–SG crossover would bear closer theoretical investigation.

Since the static rate (Δ) and B_{hf} both exhibit a break in slope, and both reflect local static order, it is of interest to compare their behaviour. Fig. 4 shows the temperature dependences of Δ and B_{hf} for x $\sim$1.0 and $\sim$1.6, with each data set normalised to its value extrapolated to T=0 K. In each case the two measurements track reasonably closely, but the behaviour of B_{hf}(T) is generally smoother, with a less well-defined break at T_{xy}. Despite the operational simplicity of the Mössbauer measurements, it is clear that μSR provides clearer insight into the static behaviour, and in addition dynamics can be probed independently.

The solid symbols in Fig. 3 summarise the results of zero-field Mössbauer measurements of T_c and T_{br}[4]. The consistency between T_{br} from B_{hf}(T), and T_{xy} from Δ(T) and λ(T), confirms that the break in slope in the Mössbauer data is indeed due to transverse spin freezing. Furthermore, the agreement here is far better and more consistent than that found when T_{xy} values from μSR and applied field Mössbauer data were compared for a$-Fe_xZr_{100-x}$[8]. There, a substantial deviation was observed, and the gap grew as the crossover region was approached. These two data sets taken together strongly suggest that the transverse spin freezing transition is suppressed by an external applied field, and serve to emphasise the need to use zero-field techniques in its determination.

IV. CONCLUSIONS

μSR provides clear evidence of two, and only two, magnetic transitions in a$-Fe_{90-x}Ru_xZr_{10}$. 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 non-critical peak in the fluctuations at T_{xy} predicted by numerical simulations. The decline in T_c on approaching x_c is more rapid than simple models predict, and suggests that the collapse of long-ranged order may be more abrupt than previously expected. Comparison with earlier Mössbauer data shows that the break in B_{hf}(T) is due to transverse spin freezing, and the consistent agreement between these two zero-field measurements, in contrast with those made in an applied field, suggests that transverse spin freezing is significantly suppressed by an external field.

V. ACKNOWLEDGMENTS

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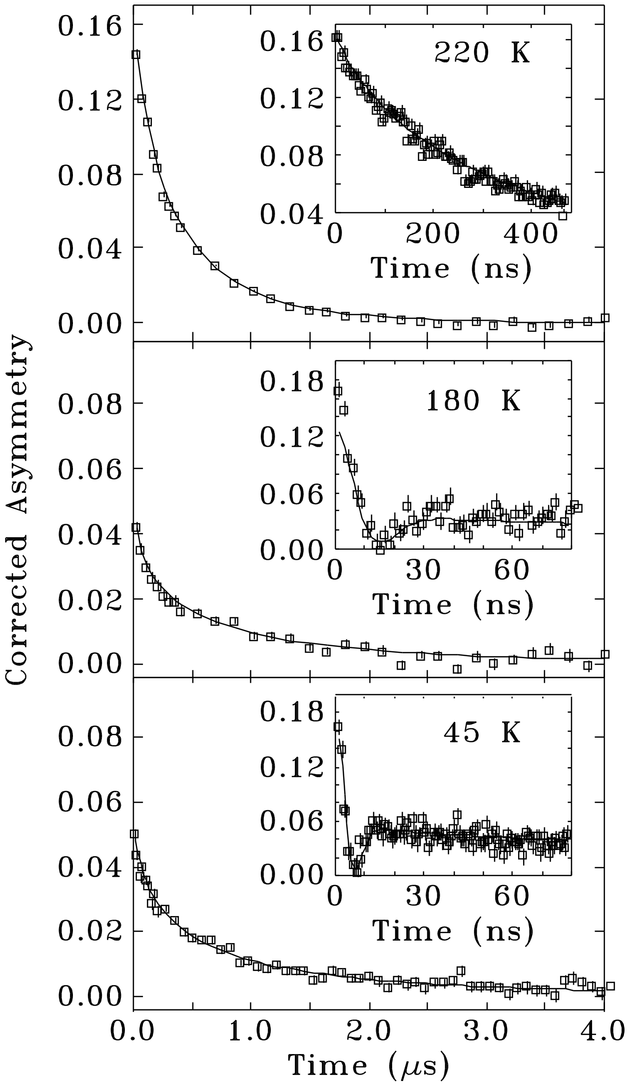

FIG. 1. Typical μSR patterns for a$-$Fe$_{89.2}$Ru$_{0}$.8Zr$_{10}$ at 220 K (above T$_c$), 180 K (below T$_c$ but above T$_{xy}$) and 45 K (below T$_{xy}$). Insets show the early time region of the data where, for T $<$ T$_c$, the static K–T minimum is observed. Solid lines are fits to functions described in the text.

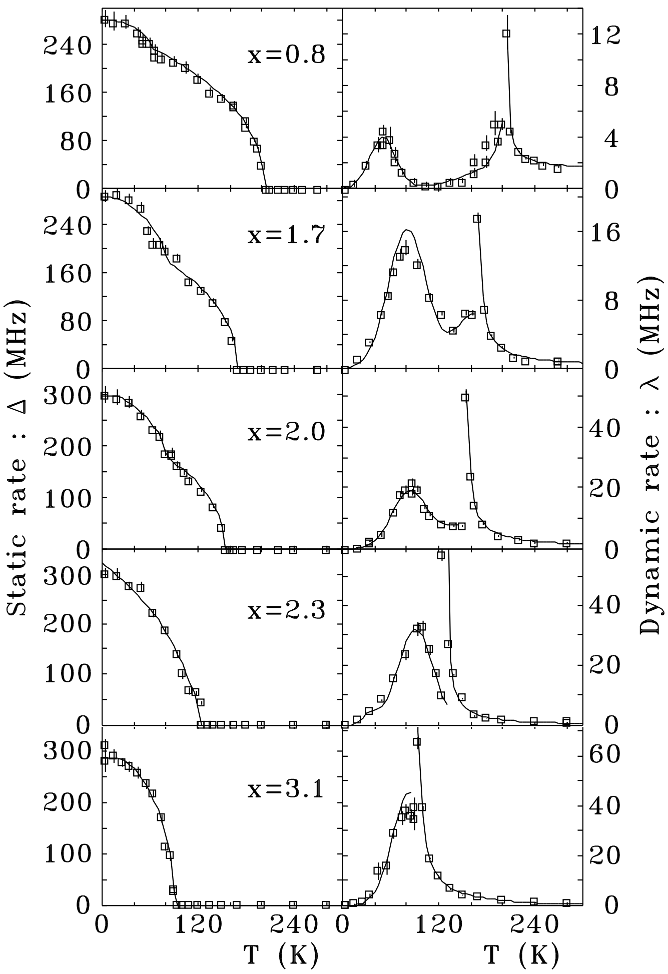

FIG. 2. Temperature dependence of the static (Δ) and dynamic (λ) relaxation rates in a$-$Fe$_{90-x}$Ru$_x$Zr$_{10}$ showing the steady reduction in ordering temperature with increasing ruthenium content, and the effects of transverse spin freezing for x $\leq$ 2.75. Solid lines are fits described in the text. Note, by x=4, Δ(T), and λ(T) show only a single ordering event.

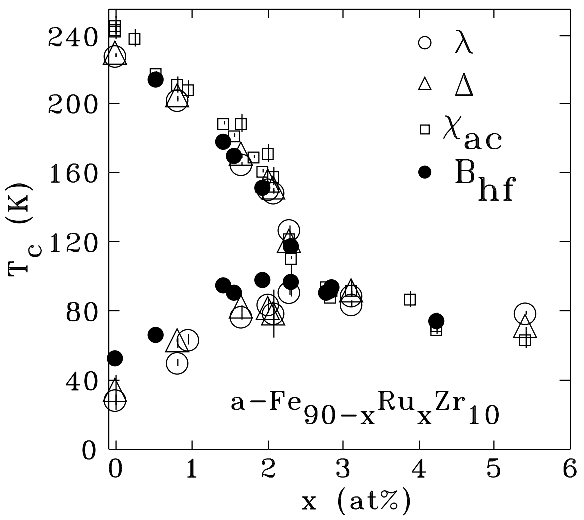

FIG. 3. Magnetic phase diagram for a$-Fe_{90-x}Ru_xZr_{10}$ showing transition temperatures derived from static and dynamic μSR data and χ_{ac} measurements. T_c and T_{xy} obtained on an independently prepared series of alloys using zero-field Mössbauer spectroscopy are also shown. All compositions derived from electron microprobe analysis.

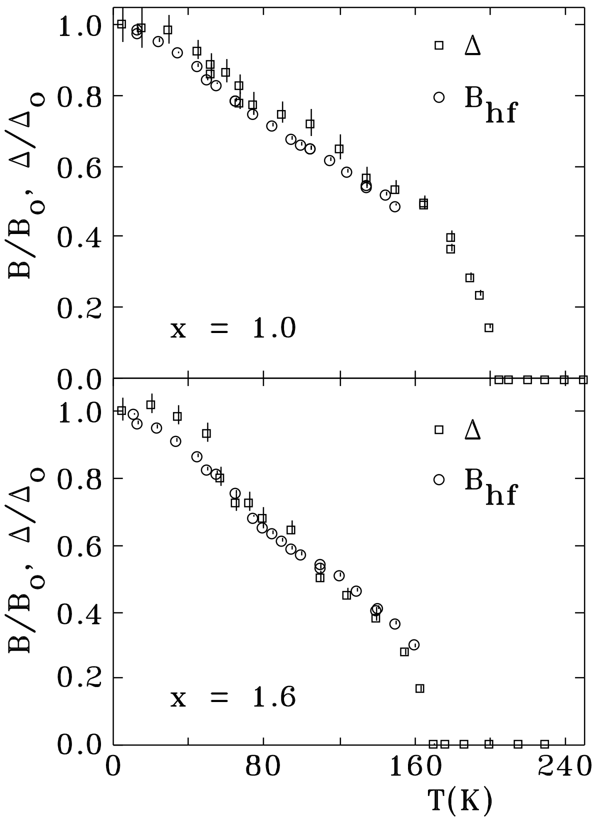

FIG. 4. Comparison of Mössbauer hyperfine fields (B_{hf}) and static relaxation rates (Δ) for Top: $x = 1.0$ (B_{hf}) and $x = 0.8$ (Δ); Bottom: $x = 1.6$ (B_{hf}) and $x = 1.7$ (Δ). Each data set has been normalised to its value extrapolated to T=0 K.