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μSR and Mössbauer studies of transverse spin freezing

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Abstract

Partially frustrated ferromagnets exhibit two magnetic transitions. T_{xy} marks the ordering of transverse spin degrees of freedom in the plane perpendicular to the axis defined by ferromagnetic order established at T_c. Zero-field muon spin relaxation (ZF-μSR) is sensitive both to the increase in static order that occurs at T_{xy} as the transverse spin components order, and also to the fluctuations associated with that ordering. The dynamic and static signatures coincide, confirming all of the features expected to be associated with the transition. We have used ZF-μSR to establish detailed phase diagrams for two bond-frustrated systems: a-Fe_xZr_{100-x} and a-$Fe_{90-x}Ru_xZr_{10}$, and a-$(Fe_{100-x}Mn_x)_{78}Si_8B_{14}$, a site-frustrated system. In addition, we have used selective excitation double Mössbauer (SEDM) spectroscopy to probe the dynamics at T_{xy} and confirm that ZF-μSR yields the same values, both for T_{xy}, and, more significantly, for the fluctuation rates.

Keywords: Spin glasses; Frustration; Magnetic ordering; Metallic glasses; Mössbauer spectroscopy

1. Introduction

The ground state of a magnetic system containing competing exchange interactions is non-collinear: the interactions are frustrated and this frustration interferes with the development of a collinearly ordered magnetic ground state. Severe frustration leads to isotropic spin freezing and the system is a spin glass; however at low levels of frustration, where the ferromagnetic interactions dominate, a net ferromagnetic component persists, and the non-collinear order develops separate from, and at a temperature below that at which the ferromagnetism appears. It is the nature of this non-collinear ordering and the manner in which it arises that form the subject of this work.

Mean field theory predicts two magnetic transitions at intermediate frustration [1]. The upper transition (T_c) marks the onset of ferromagnetic order at a conventional phase transition, while the lower one (T_{xy}) marks the ordering of spin components in the plane perpendicular to the ferromagnetic order [2][1]. Experimental systems are generally dominated by short-range interactions (rather than the infinite-range interactions associated with mean-field models) and it is possible to

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[1] A third line is present in the mean-field phase diagram, however this is a mathematical artefact and reflects a failure of the replica-symmetry methods employed to solve the model.

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introduce frustration in two distinct ways [3]: using sites or bonds. In a site-frustrated system, an antiferromagnetic dopant is introduced randomly into an otherwise ferromagnetic system, while bond frustration arises through the convolution of a distance-dependent exchange interaction with a random distribution of bond lengths or interaction distances. Numerical simulations have confirmed that the basic phenomenology arising from the two mechanisms is the same, however one significant difference has been identified: the transverse ordering induced by bond-frustration is isotropic (an *xy*-spin-glass) [4], while that arising from site frustration is antiferromagnetic [5].

A typical phase diagram for bond-frustrated a-$Fe_{90-x}Ru_xZr_{10}$ is shown in Fig. 1. With increasing frustration (Ru content), T_c falls and T_{xy} rises to meet it. Beyond $x_c \sim 2.3$ the two transitions merge and the system is a spin-glass.

The predicted signatures of transverse spin freezing are: (1) Long-range ferromagnetic order established through a phase transition at T_c and persists through T_{xy}. These systems are *not* 're-entrant' in the sense that they do not acquire and subsequently lose long-range order on cooling. (2) Ordering of spin components perpendicular to the ferromagnetic order at T_{xy}. (3) Both transitions (at T_c and T_{xy}) are accompanied by a peak in the appropriate susceptibility. This peak is divergent at T_c, while the situation at T_{xy} is less clear [4,6].

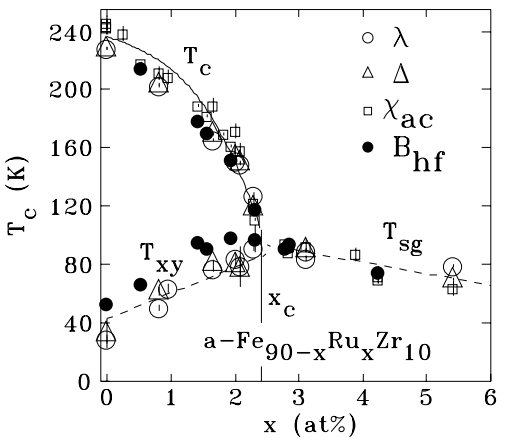

Fig. 1. Magnetic phase diagram for a-$Fe_{90-x}Ru_xZr_{10}$. Transition temperatures are taken from μSR (λ, Δ), AC-susceptibility (χ_{AC}) and Mössbauer spectroscopy (B_{hf}).

The first signature, while not obviously related to transverse spin freezing, is essential. If ferromagnetic order does not appear, it is impossible to define a transverse plane. Furthermore, without ordering at T_c it is meaningless to consider *changes* in that order. Ferromagnetic ordering has been confirmed by the observation of micron-sized domains, both within the sample plane by Lorentz microscopy [7], and through the sample thickness by neutron depolarisation [8]. Ferromagnetic order is observed at all temperatures below T_c, and is only absent when the level of frustration becomes too high for ferromagnetic order to develop. Such fully frustrated systems are then never ferromagnetic and enter a spin glass state on cooling [9]. There is no evidence for 're-entrance'.

Direct experimental evidence for the second signature in bond-frustrated systems comes from Mössbauer spectra collected on field cooling through T_{xy}. The magnetic field is used to orient the ferromagnetic order above T_{xy}, and then the ordering of components perpendicular to this order is detected through changes in line intensities in the Mössbauer spectrum. Data from AuFe [10] and *a*-Fe–Zr alloys [11] have clearly confirmed the basic nature of the transition at T_{xy}. Furthermore, by exploiting the electric field gradient in crystalline AuFe as an internally defined axis, it was possible to show that transverse spin freezing occurs spontaneously and does not require an external field to define the ferromagnetic ordering direction [12]. Polarised neutron diffraction on a single crystal of Fe_2MnSi has demonstrated that site frustration also leads to transverse spin freezing and that this order is antiferromagnetic [13]. A similar conclusion was reached in a Mössbauer study of transferred hyperfine fields in ^{119}Sn-doped a-$(Fe_{1-x}Mn_x)_{78}Sn_2Si_6B_{14}$ [14].

Secondary support for ordering of transverse spin components comes from comparison of bulk magnetisation and hyperfine fields from Mössbauer spectroscopy. Both a-$Fe_{90-x}Ru_xZr_{10}$ [15] and a-$(Fe_{1-x}Mn_x)_{78}Sn_2Si_6B_{14}$ [14] exhibit a clear break in plots of the hyperfine field (B_{hf}) vs. temperature at T_{xy}, while no such feature is seen in the magnetisation. This means that the increase in total spin length seen by B_{hf} does not appear in the ferromagnetic direction and therefore occurs in the

xy-plane. Both spin-glass and antiferromagnetic ordering of the transverse components are consistent with this observation.

The work presented here deals with the third signature. Our starting point is the prediction by numerical simulations that transverse spin freezing should be associated with a significant fluctuation peak [4]. We have exploited the excellent sensitivity of μSR to both static and dynamic magnetic behaviour to demonstrate that this dynamic peak is indeed present and coincides with the expected growth in static order at T_{xy} [16–18]. The μSR measurements are made without having to apply an external field (c.f. the Mössbauer data above) and therefore provide an unperturbed window onto the ordering behaviour of partially frustrated magnetic systems. In addition we have developed a modified Mössbauer technique: selective excitation double Mössbauer (SEDM) spectroscopy [19] which is sensitive to dynamics and allows us to make a direct comparison between fluctuations observed at an interstitial impurity (the muon) and at the nucleus of the atom with the fluctuating moment.

2. Experimental methods

Samples were prepared by arc-melting followed by melt-spinning. ZF-μSR measurements were made on the M13 and M20 beamlines at TRIUMF. Field-zero was set to 1 μT using a flux-gate magnetometer. Samples were 170–200 mg cm^{-2} thick over a 16 mm diameter active area. Histograms containing $\sim 4 \times 10^7$ events were fitted using a conventional non-linear least-squares minimisation routine.

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. The asymmetry will follow the Kubo–Toyabe (K–T) form [20]:

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

with $\alpha = 2$, so that Δ/γ_μ is the rms field. This function (see insets to Fig. 2 at 110 and 5 K)

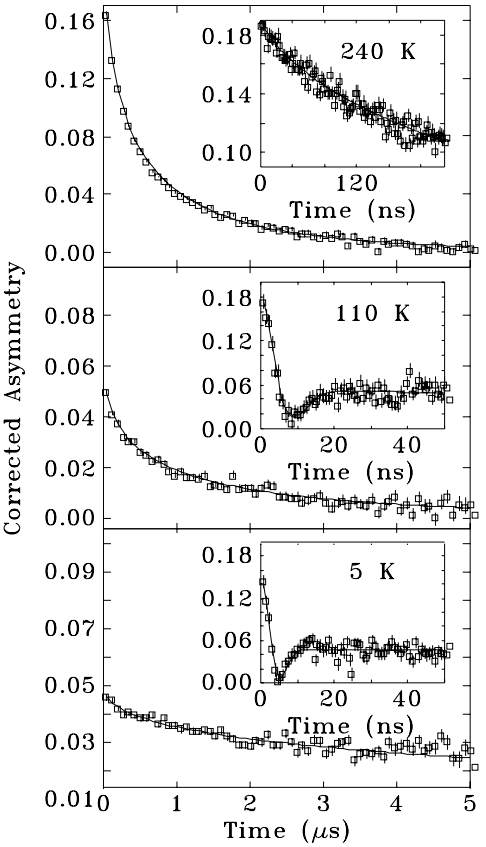

Fig. 2. Typical μSR asymmetry patterns for a-$Fe_{91.5}Zr_{8.5}$ at 240 K (above T_c), 110 K (below T_c but above T_{xy}) and 5 K (below T_{xy}). Insets show the early time region of the data where, for $T < T_c$, the static K–T minimum is observed. Solid lines are fits to functions described in the text.

exhibits a minimum at $\Delta \times t = \sqrt{3}$ then recovers to 1/3 for long times. Fluctuations lead to an exponential dephasing of the muon polarisation:

$$A_d = A_o \exp(-\lambda t), \qquad (2)$$

where λ is an effective relaxation rate. In cases where both static order and fluctuations are present, the asymmetry decays according to the product:

$$A = A_d \times G_z. \qquad (3)$$

We caution that the preceding expression is an approximation and only holds if $\Delta \gg \lambda$ [21]. This condition is not met near T_c, especially in the highly frustrated samples, and a proper treatment

of the dynamics is essential to avoid falling into the trap of using a stretched exponential to analyse the data [22].

Selective excitation double Mössbauer (SEDM) spectra were collected using a 2 GBq ^{57}CoRh source. Sample temperatures of 20–100 K were obtained with a closed cycle refrigeration system. Counting times were 8–10 days. The source was driven at a constant Doppler velocity chosen to correspond to a specific resonance line in the absorption spectrum, and a resonant conversion electron detector mounted on a drive operated in constant acceleration mode was used to detect the scattered radiation [19].

3. Results

Fig. 2 illustrates a primary strength of μSR: 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. The static K–T contribution is confined to the first 20 ns, while the dynamic decay is spread over the remaining 5 μs.

T_c is well defined and can be assigned to the temperature at which either λ diverges or Δ ceases to be zero on cooling. These values typically agree within the experimental error of 1–2 K. Furthermore, T_cs determined by μSR are in full agreement with values determined from bulk measurements (magnetisation and susceptibility) and by Mössbauer spectroscopy. This works at all compositions and levels of frustration and serves to confirm that μSR is a consistent probe of the magnetic ordering (see Fig. 1).

On cooling below T_c, the dynamic relaxation rate falls as the fluctuations freeze out. In non-frustrated samples, this slow-down continues down to the lowest temperatures measured (typically 5 K in this work) and λ falls below 0.1 MHz. In frustrated materials the decline is interrupted by a second peak: T_{xy}. This peak is clearly visible in Fig. 3 and provides an unambiguous signature of T_{xy}.

The ordering of the transverse spin components also leads to an increase in the total ordered spin.

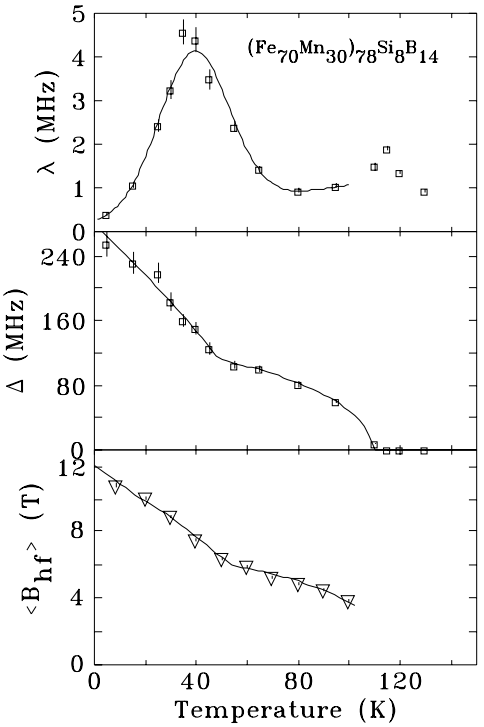

Fig. 3. Dynamic relaxation rates (λ) and static rate (Δ) derived from μSR compared with the average hyperfine field ($\langle B_{hf} \rangle$) derived from Mössbauer measurements on a-$(Fe_{70}Mn_{30})_{78}Si_8B_{14}$.

Both the static rate (Δ) and the average hyperfine field ($\langle B_{hf} \rangle$) derived from Mössbauer measurements may show a break in their temperature dependences, as this extra order develops (lower panels of Fig. 3). However, they are far less well localised and in both low frustration samples (where the transverse contribution is small) and high frustration samples (close to x_c where the two transitions merge) the break is often indistinguishable from the normal increase on cooling. In general, we have found agreement within $\sim$5 K for the three alloy systems we have studied, with any inconsistency between the break point and the peak in $\lambda(T)$ being dominated by difficulties in fitting the position of the break. Various determinations of both T_c and T_{xy} are summarised as a phase diagram in Fig. 1. In none of the systems studied here have we found any evidence for a third transition below T_{xy}. Our static and dynamic

signatures coincide in both site-and bond-frustrated materials, and we have not been able to confirm the earlier claim for a third transition in the a-Fe–Mn-glass system [23].

In order to establish that the muons are seeing a real fluctuation peak at T_{xy} we turn to a probe that is sensitive to the fluctuations of the actual moments, rather than the magnetic field at an interstitial site. In a procedure analogous to optical pumping, SEDM spectroscopy uses one Mössbauer event to populate a specific hyperfine level in the target nucleus and then a second such event to Mössbauer-analyse the radiation re-emitted during the decay [19]. Changes in the magnetic environment of the probe nucleus while it is in the excited state ($\sim$100 ns) lead to additional lines in the observed spectrum. Both the intensity and width of these lines are used to determine the average relaxation rate. The data in Fig. 4 show precise agreement between the relaxation rates determined by the two techniques [24], and confirm that the muons are indeed seeing the fluctuations of the iron moments.

Finally, our initial μSR work on a-Fe_xZr_{100-x} revealed a significant discrepancy between T_{xy} values obtained from μSR and applied-field Mössbauer spectroscopy [17]. This led to speculation that the applied field was suppressing T_{xy}. Subsequent longitudinal field μSR (LF-μSR) confirmed the dramatic impact of an applied field, and data in Fig. 5 show that we were able to follow T_{xy} as a field suppressed it by a factor of four [25]. Furthermore, it is clear that existing theoretical predictions of this behaviour derived from mean-field calculations (the G–T and A–T lines shown on Fig. 5) [26] are inaccurate, and this rather severe failure should be addressed.

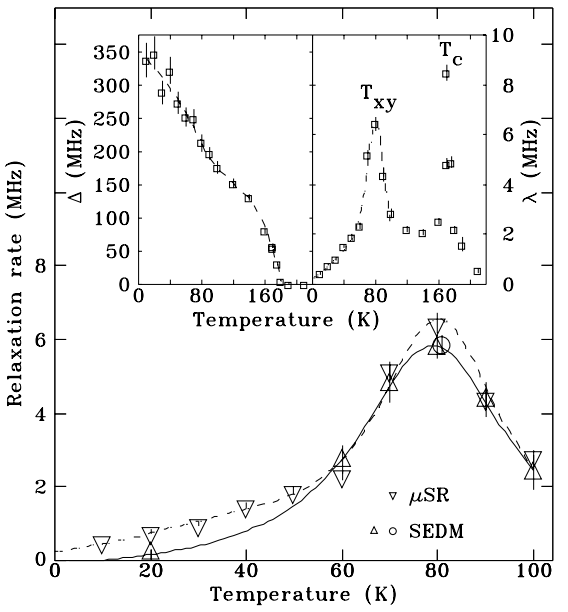

Fig. 4. Inset: Temperature dependence of the static (Δ) and dynamic (λ) relaxation rates of a-$Fe_{92}Zr_8$. Body: Relaxation rates from ZF-μSR fits (∇) and SEDM fits ($\triangle$, $\bigcirc$) around T_{xy}.

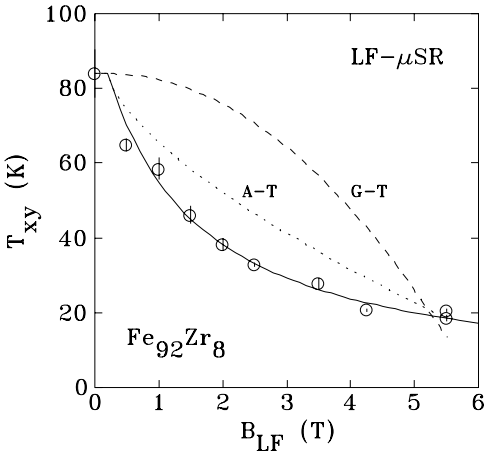

Fig. 5. Field dependence of T_{xy} measured by LF-μSR in a-$Fe_{92}Zr_8$. The solid line is a phenomenological fit. Dashed (G–T) and dotted (A–T) lines show mean-field predictions, scaled to agree at 0 and 5 T.

4. Future directions

μSR has clearly provided significant insight into the ordering at T_{xy}, however, one major question remains unanswered: is the event at T_{xy} a phase transition? Indications from early numerical simulations were that it represented a change in short-range order only [4], making it essentially dynamic in nature. However, more recent simulations may be more consistent with there being a phase transition at T_{xy} [6]. Furthermore, we have found that there is a peak in the out-of-phase susceptibility (χ'') at T_{xy}, and this peak coincides (within experimental error of $\sim$2 K) with T_{xy} from μSR. This agreement between measurements on

timescales separated by six decades is hard to reconcile with a dynamic, short-ranged ordering of transverse spin components. A complete scaling analysis of the fluctuation peak shapes in μSR and more extensive numerical simulations are clearly needed to make progress on this issue.

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