Spin dynamics in a frustrated magnet

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180° spin flips have been identified as the dominant fluctuation mechanism at the transverse spin freezing transition in partially frustrated a$-Fe_{92}Zr_8$. The form of the selective excitation double Mössbauer spectra, coupled with the perfect agreement with zero-field muon spin relaxation data, eliminates other relaxation forms.

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Competition between majority ferromagnetic (FM) exchange and low levels of antiferromagnetic (AF) exchange leads to frustration and non-collinear ordering. At high temperatures, there is a paramagnetic to ferromagnetic phase transition (at T_c) where the z component of the moments order collinearly. Subsequently, at a lower temperature (T_{xy}), transverse spin freezing occurs: the x and y components of the moments freeze out and the system enters a non-collinear state where ferromagnetic and spin-glass order coexist [1].

The unusual magnetic phenomenon of transverse spin freezing was initially observed in iron rich Au-Fe alloys with zero and applied-field Mössbauer measurements [2,3], however, results were overshadowed by the metallurgical instability of the Au-Fe system. Conclusive evidence of transverse spin freezing was demonstrated in the iron-rich a-Fe_xTM_{100-x} (TM=Zr,Hr) metallic glasses with applied-field transmission Mössbauer spectroscopy [4] where the spin components were observed to develop perpendicular to the field on cooling through T_{xy}. Although there is remarkable quantitative agreement between numerical [5] and experimental [4] results, simulations have only been carried out in zero field while the transmission Mössbauer experiments were carried out in a significant applied field (2–5 T).

An alternative, zero-field signature of T_{xy} has been identified by numerical simulations, which predict magnetic fluctuations associated with the freezing of transverse spin components [5]. Searches by χ_{ac} [6–8] are complicated by the dominant ferromagnetic order, and results were inconclusive. By contrast, zero-field muon spin relaxation (ZF-μSR) provides a complete separation of static and dynamic magnetic behaviour and allows the magnetic ordering to be probed under conditions identical to those used in the numerical simulations. ZF-μSR measurements on a-Fe_xZr_{100-x} [9] and a-$Fe_{90-x}Ru_xZr_{10}$ [10] have confirmed that the static and dynamic signatures of T_{xy} are both present as predicted, and ruled out any contribution from magnetic inhomogeneities [11].

While remarkable agreement exists between theory and experiment in terms of the static and basic dynamic magnetic behaviour of frustrated magnets, the detailed nature of the fluctuations at T_{xy} remains unknown. Furthermore, neither mean-field calculations [12], nor Monte-Carlo simulations [5] are capable of addressing this fundamental issue. The simplest possibility might be a precession that gradually slows on cooling, but other forms are easily imagined, such as rapid moment reversals that block at T_{xy} much as in a superparamagnetic fine particle system. Although ZF-μSR is uniquely sensitive to magnetic relaxation in the presence of static order, it cannot be used to differentiate between specific moment fluctuation modes (e.g. spin flips, continuous easy-axis relaxation, etc.).

In this letter we present a direct comparison between ZF-μSR and selective excitation double Mössbauer (SEDM) spectroscopy measurements on a-$Fe_{92}Zr_8$ around T_{xy} ($\sim$80 K). The results show that the relaxation rates measured by these two distinct techniques are in perfect agreement, and that the dominant fluctuation mode is clearly moment reversals and not precessional rotation.

a-Fe_xZr_{100-x} is a well-characterized, metallurgically stable, partially frustrated Heisenberg magnet [4]. Its phase diagram [9] determined from χ_{ac}, applied-field transmission Mössbauer, and ZF-μSR spectroscopy, shows that it is ferromagnetic at $x = 88$, and enters a fully frustrated spin-glass state by $x = 92.8$. μSR data show that the $x = 92$ composition is well suited for a study of spin dynamics at T_{xy}, as ferromagnetic order is established at $T_c \sim 180$ K, and $T_{xy} \sim 80$ K. Spectra of the 16 mm diameter, 200 mg cm^{-2} thick sample of melt spun ribbons were collected at TRIUMF on the M13 beamline in a He-flow cryostat. Time histograms of 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). A typical ZF-μSR spectrum of a-$Fe_{92}Zr_8$ is shown in Fig. 1.

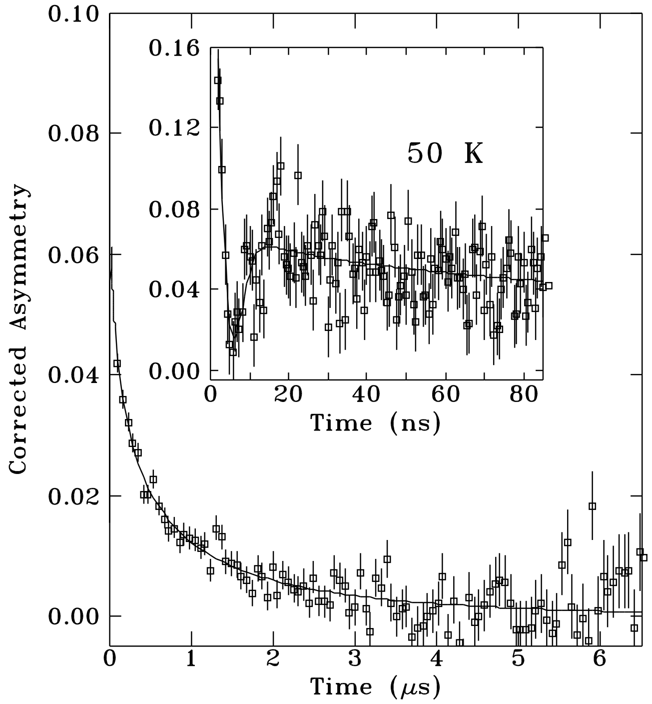

FIG. 1. Typical ZF-μSR spectrum of a-$Fe_{92}Zr_8$ at 50 K (below T_{xy}). Inset shows the early-time region where the KT minimum characteristic of static order is visible. Note the clear separation in time-scales for the static and dynamic contributions.

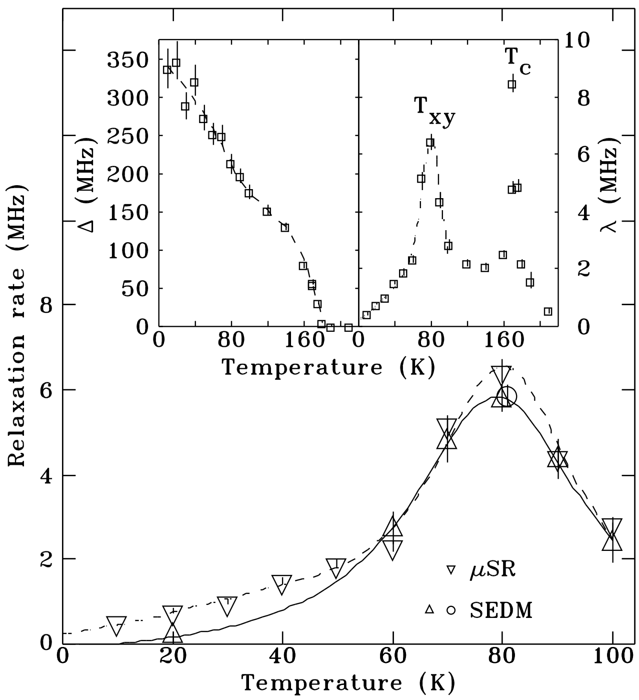

FIG. 2. Inset: Temperature dependence of the static (Δ) and dynamic (λ) relaxation rates of a-$Fe_{92}Zr_8$. Dashed lines are guides to the eye. Body: Relaxation rates from ZF-μSR fits ($\bigtriangledown$) and SEDM fits ($\bigtriangleup$,$\bigcirc$) around T_{xy}. Lines are guides to the eye.

Since the static and dynamic μSR signals are quite distinct [9], a product function was used to fit the data, i.e. $G_z(t) = \{\frac{1}{3} + \frac{2}{3}(1-(\Delta t)^\alpha)\exp(-\frac{(\Delta t)^\alpha}{\alpha})\} \times \exp(-\lambda t)$, where for $\alpha = 1$ the term in $\{...\}$ denotes a Kubo-Toyabe (KT) asymmetry form for a Lorentzian distribution of static local fields [13] and for $\alpha = 2$ the term in $\{...\}$ is the KT asymmetry form for a Gaussian distribution of static local fields with Δ/γ_μ the rms field [13]. λ represents a $1/T_1$ type spin relaxation rate.

At T_c, $\alpha = 1$ was found to describe the Lorentzian KT functional form of the spectra. Within 30 K of T_c, a Gaussian distribution of local fields, with $\alpha = 2$, described the spectra. This variation of α around T_c indicates an evolution in the local moment distribution as the collinear static order is established below T_c. Results from fits to the spectra for a-$Fe_{92}Zr_8$ are shown in Fig. 2. Examining $\Delta(T)$ on cooling, we see that the static magnetic field is zero until T_c is reached and collinear ferromagnetic order is established for the z component of the moments. A steady increase in Δ occurs on further cooling, until T_{xy} is reached, where a more dramatic rise in the static magnetic field occurs as the x and y components of the moments order with their time-average no longer being zero.

A more marked signature of moment behavior is provided by the relaxation rate, $\lambda(T)$, in Fig. 2. As the temperature approaches the paramagnetic to ferromagnetic phase transition from above, magnetic fluctuations occur over a continuum of time- and length-scales, and ultimately diverge at T_c. The relaxation rate measured by ZF-μSR reflects this divergence (it is well fitted by a Curie-Weiss C/(T-T_c) form) and yields $T_c = 180\pm5$ K (in excellent agreement with χ_{ac} data that measures fluctuations on a much slower time scale). Below T_c, fluctuations are much weaker and the muon depolarisation is dominated by the effects of magnons. By 0.9 T_c in a-$Fe_{89}Zr_{11}$ (a composition that exhibits no frustration effects) λ is below 1 MHz [9] and slows to less than 0.1 MHz. However, in the a-$Fe_{92}Zr_8$ frustrated magnet studied here, this decline in $\lambda(T)$ is cut off by the approach of T_{xy}. Fluctuations build up and $\lambda(T)$ increases to a second maximum at T_{xy}. The behavior around T_{xy} is clearly non-critical as $\lambda(T)$ does not diverge. Indeed, the behavior is quite rounded and is well fitted by a Gaussian peak (dashed line in Fig. 2). Below T_{xy} all fluctuations gradually freeze out and λ approaches zero.

In order to investigate the detailed nature of the fluctuations around T_{xy}, we turn now to selective excitation double Mössbauer (SEDM) spectroscopy. SEDM is a modified Mössbauer technique that relies on Mössbauer analyzing the radiation re-emitted from the sample following resonant pumping into a selected hyperfine state.

As with ZF-μSR, static and dynamic disorder can be completely distinguished as they each yield distinct and unambiguous spectral signatures [14,15]. Our recent innovations (e.g. a three order of magnitude improvement in effective counting times, and a spectrometer that can run continuously without periodic energy calibrations [15,16]), have removed SEDM from it's standing as a fringe spectroscopic technique, and allowed us to apply it to problems of current interest [14,15].

SEDM spectra of the same a-$Fe_{92}Zr_8$ sample used for ZF-μSR measurements were collected using a 2 GBq ^{57}Co**Rh** source. Sample temperatures of 20 to 100 K were obtained with a closed cycle refrigeration system. Counting times were 8–10 days. For all temperatures the leftmost line (corresponding to the $m_g = -\frac{1}{2} \rightarrow m_e = -\frac{3}{2}$ transition, line #1) was pumped as it has the largest absorption cross-section, thus maximizing the signal and reducing collection times. To unequivocally fix the type of moment oscillations, the second line (corresponding to the $m_g = -\frac{1}{2} \rightarrow m_e = -\frac{1}{2}$ transition, line #2) was also pumped at T_{xy}. Typical spectra obtained while pumping lines #1 and #2 at 80 K are shown in Fig. 3.

At 20 K, the absence of magnetic fluctuations is confirmed by the observation of a single emission line in the a-$Fe_{92}Zr_8$ SEDM spectrum. Since the $m_e = -\frac{3}{2}$ level is pumped, in a static magnetic field, the radiation re-emitted when the excited nucleus decays to the ground state is at the same energy as the pump energy. This is because the static magnetic field present at the ^{57}Fe nucleus induces hyperfine splitting in the $I_g = \frac{1}{2}$ ground state and the $I_e = \frac{3}{2}$ excited state, and selection rules ($\Delta m_I = 0, \pm 1$) result in the $m_g = -\frac{1}{2}$ ground state as the only possible decay from the $m_e = -\frac{3}{2}$ excited state.

a-$Fe_{92}Zr_8$ is chemically and magnetically disordered, and this static disorder leads to broadened absorption lines in the transmission spectra that reflect a convolution between the source linewidth and hyperfine field distribution. The SEDM spectra of a material with static disorder also show a broadened re-emission line (compared to that of α-Fe) however it is substantially narrower than that observed by transmission as only part of the field distribution is probed [14]. The 20 K SEDM spectrum of the a-$Fe_{92}Zr_8$ is correctly described by the asymmetric Gaussian distribution of hyperfine fields determined from the 20 K transmission spectrum. We thus have a complete description on the static magnetic disorder to be convolved with the x and y moment fluctuations which deveolp around T_{xy}.

The onset of spin dynamics has a significant impact on the SEDM spectra in Fig. 3 and these spectra provide incontrovertible evidence of 180° moment reversals as the source of moment fluctuations around T_{xy}. While only line #1 at ~ -3 mm/s is pumped in the top SEDM spectrum, a new line (#6) is clearly present at $\sim +3$ mm/s. This is a direct indication that the magnetic field at the excited nucleus has reversed direction during the lifetime ($\sim$ 100 ns) of the excited state, so that the projection of I_e onto the field has changed sign. That is, while the $m_e = -\frac{3}{2}$ state was initially populated, after the magnetic field reversal, the populated state has changed to $m_e = +\frac{3}{2}$ which then decayed to the $m_g = +\frac{1}{2}$ state, giving line #6 at $\sim +3$ mm/s. The observed SEDM spectra undeniably indicate that moments are undergoing 180° spin flips near T_{xy}. *No* amount of static disorder can lead to this additional line at $\sim +3$ mm/s.

Confirmation that 180° moment flips are associated with the fluctuations at T_{xy} is possible by examining another transition. Line #2 ($m_g = -\frac{1}{2} \rightarrow m_e = -\frac{1}{2}$) has the next largest nuclear cross-section and so offers the next best possible signal for the collection time. In a static magnetic field, pumping a Mössbauer nucleus into the $m_e = -\frac{1}{2}$ excited state results in a decay into the $m_g = -\frac{1}{2}$ and $m_g = +\frac{1}{2}$ ground states: lines #2 and #4 appear in a SEDM spectrum when line #2 has been driven. Examining the bottom SEDM spectrum of a-$Fe_{92}Zr_8$ in Fig. 3 when line #2 is driven, we see that lines #2 and #4 appear as expected. However, lines #3 and #5 are also clearly present. Again, reversal of the magnetization direction during the lifetime of the excited state leads to population of the $m_e = +\frac{1}{2}$ excited state, which then decays to the $m_g = +\frac{1}{2}$ and $m_g = -\frac{1}{2}$ ground states yielding lines #3 and #5. The behavior inferred from driving line #1 is thus fully confirmed.

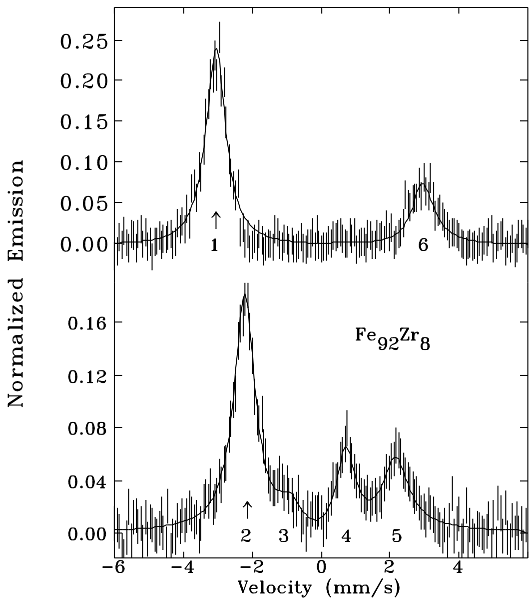

FIG. 3. SEDM spectra at $T_{xy} = 80$ K when the $m_e = -\frac{3}{2}$ state is populated (top) and when the $m_e = -\frac{1}{2}$ state is populated (bottom). Pump energies are indicated by the ↑.

The form of the SEDM spectra clearly indicates that 180° moment reversals are the dominant contribution to the Mössbauer-observed relaxation behaviour Although moment flip rates are essentially a function of the SEDM drive and return line intensities, the static disorder established below T_c needs to be included for a complete, physical description of the spectra. With a model based on Sack's [17] stochastic quantum mechanical formalism, a more quantitative description of the relaxation lineshape, including static disorder measured from transmission Mössbauer spectra, allows accurate information to be extracted from the full SEDM lineshape. Baseline, pump line intensity, excited-state linewidth, and relaxation rate are the only free parameters in the least-squares fits. Results of fits to the SEDM data are shown in Fig. 2. Relaxation rates determined when line #1 is driven are shown by the $\triangle$ and the rate when line #2 was driven at 80 K is shown by the $\bigcirc$ in Fig. 2. While ZF-μSR is, in principle, sensitive to all forms of magnetic relaxation, and has a wider frequency window than the Mössbauer effect, the fitted SEDM relaxation rates shown in Fig. 2 are in perfect agreement with the ZF-μSR rates. This perfect correspondence between results that consider all modes of moment fluctuations (ZF-μSR), and only 180° moment reversals (SEDM) leads inveitably to the conclusion that, around T_{xy}, moment reversals are the prevailing relaxation mechanism. It is likely that the slightly higher relaxation rate (compared to the SEDM value) seen by ZF-μSR at 20 K reflects the presence of some fluctuations other than moment reversals.

Additional evidence for 180° moment reversals being the dominant relaxation mechanism is provided by the fitted linewidth of the SEDM spectra. With the source and detector linewidths characterised using standard samples [16], and the static disorder for the current sample determined from transmission Mössbauer spectra, we observe a temperature-independent sample linewidth of 0.10 ± 0.01 mm/s that is fully consistent with the lifetime (100 ns) of the excited state of the Mössbauer nucleus. This constant linewidth allows us to rule out the presence of any small-amplitude moment fluctuation modes, as they would lead to a significant increase in linewidth even if they did not induce mixing of the $m_e = \pm\frac{3}{2}$ states [18].

To summarize, in a-$Fe_{92}Zr_8$, a paramagnetic to ferromagnetic phase transition at T_c results in collinear ordering of the z component of the moments. The static magnetic disorder from this collinear ordering is present in both ZF-μSR and Mössbauer effect measurements, while moment fluctuations, measured with ZF-μSR, exhibit a Curie-Weiss divergence. At the lower temperature, T_{xy}, transverse spin freezing happens. The non-critical magnetic fluctuations from the x and y components of moments slowing into the measuring time window of both the muon and Mössbauer effect have been detected at T_{xy}, with transverse spin freezing indicated by slowing of fluctuations with cooling. 180° moment flips have been explicitly identified as the dominant fluctuation mechanism of transverse spin freezing. Although an average over all possible spin fluctuation modes is measured with ZF-μSR, SEDM shows different spectral signatures for different relaxation phenomena, and yields a model independent measure of the spin relaxation rate. The excellent agreement between fluctuation rates from ZF-μSR and SEDM provides strong evidence that the same magnetic relaxation phenomenon is detected with the two different probes.

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