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Field dependence of the transverse spin freezing transition

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Transverse spin freezing in a-$Fe_{92}Zr_8$ has been studied using longitudinal field muon spin relaxation in fields of up to 5.5 T. The fluctuations associated with freezing of the transverse spin components are confirmed as a robust signature of T_{xy}. A $1/B$ dependence for $T_{xy}(B)$ is observed, in qualitative disagreement with all current theoretical descriptions of the transition.

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T_{xy} marks the temperature at which a partially frustrated three-dimensional Heisenberg magnet develops static spin components perpendicular to the ferromagnetic order established at T_c. Between T_c and T_{xy} the system is ferromagnetic, while below T_{xy} the magnetic structure is characterized by coexisting, and mutually perpendicular, ferromagnetic and xy-spin-glass ordering.[1–3]

Comparisons between theoretical predictions and observed behavior yield good, qualitative agreement on the form of the phase diagrams and the nature and sequence of orderings. Furthermore, semiquantitative agreement between scaled transition temperatures and noncollinearity has been demonstrated for both bond[4] and site[5] frustrated systems. However, detailed, quantitative tests are lacking, primarily because it is impossible to map the simplified exchange and moment distributions employed in the models onto the unknown distributions present in the real materials. Furthermore, changing the sample composition in order to map out a phase diagram necessarily modifies these distributions, again in an unknown way.

The work presented here addresses the issue of quantitative comparison by focussing on a single sample and using an applied field to modify the ordering behavior directly. We confirm that T_{xy} persists in substantial external fields but is strongly suppressed. The functional form of this suppression provides a severe test of existing models of partially frustrated magnetic systems. By tracking T_{xy} in an external field at fixed frustration and composition, we are able to restrict our attention to two thermodynamically relevant variables: B and T.

We have previously shown that zero-field muon spin relaxation (ZF-μSR) can be used to locate T_{xy} through both the increase in static order and the peak in the fluctuation rate associated with the ordering of the transverse spin components.[6–8] The data presented below confirm that the fluctuation peak is readily observed in a substantial applied field, and is therefore a robust signature of the transition.

a-Fe_xZr_{100-x} is a well-characterized, metallurgically stable, partially frustrated Heisenberg magnet.[4] Its phase diagram (Fig. 1) shows that it is ferromagnetic at $x=88$, and enters the fully frustrated spin-glass state by $x=92.8$. Neutron depolarization has confirmed that at all compositions where ferromagnetic order is established at T_c, this order persists through T_{xy} and down to the lowest temperatures examined (~ 5 K).[9,10] ZF$-\mu$SR has further shown that the magnetic order is uniform, with no evidence for magnetic segregation.[7] The system therefore provides an ideal test bed for the study of transverse spin freezing. The composition dependence of T_{xy} has been determined using ZF-μSR,[6–8] exploiting both the increase in static order and the fluctuation signature predicted by numerical simulations.[3] The observed form of the phase diagram differs from that predicted by mean-field theory in two major respects:[11] (i) there is no evidence for a third transition below T_{xy}; and (ii) T_c appears to be a stronger function of x than T_{xy}, the reverse of the mean-field prediction. Both features have also been observed in Ru-doped a-$Fe_{90-x}Ru_xZr_{10}$.[8] Better agreement is found with the more realistic numerical simulations, in that only two transitions (T_c and T_{xy}) are predicted, however, that work was not detailed enough to address the precise form of the frustration dependences of the transitions.

Longitudinal-field muon spin relaxation (LF-μSR) measurements were made on the M20 beamline at TRIUMF. Sample temperature was controlled between 5 and 300 K in a He-flow cryostat. The sample was 16 mm in diameter and 200 mg cm^{-2} thick. Histograms containing $1-4\times10^7$ events were acquired with a timing resolution of 0.8 ns. Longitudinal fields (i.e., parallel to the initial muon polarization) of up to 5.5 T were applied using a superconducting solenoid. In all cases, the field was applied well above T_{xy} and the measurements made on field cooling, to eliminate any possible sample history effects.

For a complete description of μSR methodology, the

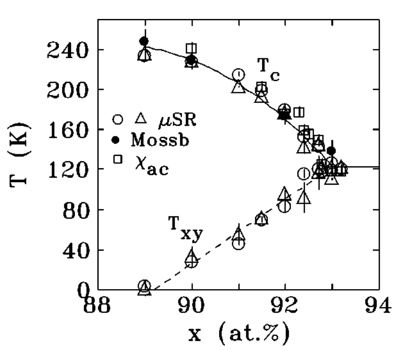

FIG. 1. Magnetic phase diagram for a-Fe_xZr_{100-x}.

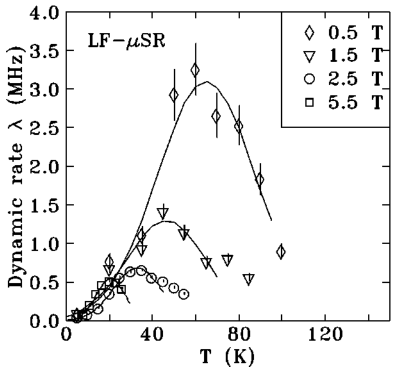

FIG. 2. Temperature dependence of the dynamic relaxation rate in a-$Fe_{92}Zr_8$ for a number of representative fields. Note that a clear maximum, defining T_{xy}, is observed in all cases.

reader is referred to a number of excellent reviews.[12] In our earlier zero-field work on this system, the time-dependent asymmetry between the forward and backward counters was fitted using a product of a static Kubo-Toyabe function (K-T)[13] and an exponential decay reflecting dynamics.[6] The application of a significant longitudinal field affects the K-T function in two ways. Firstly, it modifies the shape of the function, sharpening the observed minimum and moving the K-T contribution to earlier times.[13–15] Secondly, as the field magnetises the sample parallel to the muon polarization, it greatly reduces the amplitude of the K-T term, eliminating it entirely in the limit of perfect alignment. As a result, the static K-T contribution was not resolved in most of the measurements made here, and the analysis concentrated on the dynamic term.

Since a longitudinal field will favor ferromagnetic order over xy-spin-glass ordering, we expect that T_{xy} will be suppressed by an applied field. The temperature dependence of the relaxation rate (λ), shown in Fig. 2 for a number of fields, indicates that a peak corresponding to T_{xy} was readily observed in all fields used, confirming that fluctuations provide a robust signature of T_{xy}. The fluctuation peak is reduced in amplitude and, as expected, moves to lower temperatures as the field is increased.

Theoretical predictions for the field dependence of T_{xy} are limited to mean-field calculations. Unfortunately, the infinite-ranged interactions inherent to the mean-field approximation obliterate many subtle effects of exchange frustration. For example, such models are unable to distinguish bond[4] and site[5] frustrated systems. While short-ranged numerical simulations are more accurate, and correctly reproduce the different ordering behavior due to bond[3] and site[16] frustration, results for applied fields are not currently available. We are therefore forced to restrict our comparisons to the predictions of mean-field models.

Frustration in the mean-field models is characterized by J_o, the ratio between the mean and width of the (assumed) Gaussian exchange distribution. The solution of Gabay and Toulouse[1] yields three transitions in a partially frustrated system (i.e., $J_o \geqslant 1$): a ferromagnetic (FM) phase transition at T_c, followed by two more (FM→M1 and M1→M2), at lower temperatures. The M1 state is characterized by coexisting ferromagnetic and transverse spin-glass ordering and so the FM→M1 boundary (often called the $G-T$ line) corresponds to T_{xy}. The third transition, M1→M2, is associated with spontaneous replica symmetry breaking, and marked by the onset of strong longitudinal irreversibility. It is generally referred to as the $A-T$ line.[17] Subsequent work[18] has identified an instability in the model at the $G-T$ line, and casts doubt on the existence of the $A-T$ line. Given the close relationship between the in-field and zero-field phase diagrams,[19] the presence or absence of the $A-T$ line in field can be related to the existence of the M1→M2 transition in zero field. Experimentally, the third transition (M1→M2) is not seen, and given the otherwise perfect agreement between the mean-field and numerical phase diagrams, is it likely that this line is an artifact of the model.

For the fully frustrated, $J_o=0$ case, two lines with distinct field dependences are predicted. The upper ($G-T$) line marks the onset of transverse spin freezing[1,20] and should scale as:

$$T_{GT} \propto T_o \left(1-\frac{B^2}{A_{GT}}\right),$$

where A_{GT} is a constant, and T_o is the transition temperature in zero field. Similarly, the lower ($A-T$) line marks the onset of replica symmetry breaking,[1,17] and should scale as:

$$T_{AT} \propto T_o \left(1-\frac{B^{2/3}}{A_{AT}}\right).$$

These two transition lines are in fact surfaces, and are continuations of the $G-T$ and $A-T$ lines predicted in zero field for $J_o \geqslant 1$. Given that we did not observe any evidence for the $A-T$ line in zero field,[6–8] and the prediction that replica symmetry fails on the $G-T$ line,[18] we do not expect our shift in T_{xy} to track with the $A-T$ prediction. Furthermore, the experimental ordering behavior at T_{xy} corresponds closely with that predicted at the $G-T$ line. For $J_o \geqslant 1$, the mean-field theory has to be modified to include a nonzero magnetization.[21] This leads to a field dependence for the generalized $G-T$ line of the form:

$$T_{FH} \propto T_o \left(1-\frac{B}{A_{FH}}\right).$$

Unfortunately, none of the three predicted forms describes the observed field dependence of T_{xy} shown in Fig. 3. Even the most likely candidate, T_{FH}, which lies between the $G-T$ and $A-T$ forms, does not come close to the data.

The observed field dependence is smooth and appears to be saturating at high fields, suggesting a function of the form $T_{xy} \propto 1/B$. If we introduce a scaling parameter, J_s, both to adjust the rate of decline and also to cut off the divergence at zero field, then the field shift can be described by the simple function:

$$T_{xy}^B = T_{xy}^0 \left[1-\frac{B}{(J_s+B)}\right]$$

where the superscripted '0' and 'B' mark the values in zero field and an applied field. It is clear from the solid line in

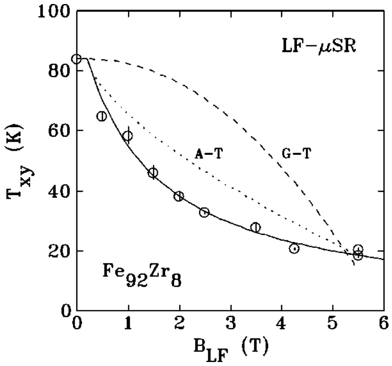

FIG. 3. Field dependence of T_{xy} measured by LF-μSR in a-$Fe_{92}Zr_8$. The solid line is a phenomenological fit described in the text. Dashed ($G-T$) and dotted ($A-T$) lines show the mean-field predictions discussed in the text, corrected for B_D and scaled to agree at 0 and 5 T.

Fig. 3 that this function fits the data remarkably well. In making the fit, T_{xy}^0 was left as an adjustable parameter, on principle, and the value returned was within 1 K of that measured in zero field, providing further confirmation that the function is a reasonable description of the data. The scaling parameter, J_s, took a value of 1.5 ± 0.3 T, which is consistent with the 1.36 T saturation polarization of this alloy.[4] The short horizontal section near $B=0$ reflects the effects of demagnetizing fields (B_D) which force the internal field to be zero until the sample is saturated. The fit yields $B_D = 0.2\pm0.4$ T, somewhat below the $\sim$1.36 T expected for this material with the field applied perpendicular to the sample plane. The field-cooling procedure adopted during the measurements allows us to rule out coercivity effects. However, the ribbons were not clamped perfectly flat, nor was it possible to orient the sample precisely perpendicular to the applied field. Even a slight misalignment would lead to a substantial reduction in the effective demagnetizing factor, therefore a reduced value for B_D is expected.

The conclusions of this work are straightforward. T_{xy} can be followed in a significant applied field and it is strongly suppressed. Existing theoretical predictions for the functional form of this suppression are incorrect. Further work in this area will take two parallel tracks. We will extend the LF-μSR work to samples with higher and lower levels of frustration in order to map out the complete $T_{xy}(J_o, B)$ surface. In addition, we will extend the numerical simulations to include externally applied fields.

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