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Reply to “Comment on ‘Field dependence of the transverse spin freezing transition’ ”

D. H. Ryan
Physics Department and Centre for the Physics of Materials, McGill University, 3600 University Street, Montreal, Quebec, Canada H3A 2T8

J. M. Cadogan
School of Physics, The University of New South Wales, Sydney, NSW 2052, Australia

J. van Lierop
Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973-5000

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Muon spin relaxation (μSR) is a more reliable technique for tracking the field dependence of T_{xy} than Mössbauer spectroscopy. It yields consistent behavior and does not suffer from fitting artifacts. In addition it can be performed both with and without an applied field. Apparent differences between applied-field Mössbauer results and those from μSR derive entirely from difficulties associated with the analysis of the Mössbauer data.

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In attempting to explain the differences between T_{xy} derived from their applied field Mössbauer data and our longitudinal-field muon spin relaxation (LF-μSR) results,[1] Kaptás *et al.*[2] appeal to possible magnetic inhomogeneities or the effects of differing sample production and handling procedures. Neither of these claims is valid. They further argue that Mössbauer spectroscopy is inherently superior as, in principle, it allows direct estimation of the demagnetizing field. However, as we show here, the uncertainties and instabilities introduced by the data analysis procedures far outweigh any possible advantages.

I. SAMPLE RELATED ISSUES

There is no evidence to support a claim that *a*-Fe-Zr alloys are either magnetically or chemically inhomogeneous. We have used neutron depolarization,[3] μSR,[4,5] and most recently, selective excitation double Mössbauer spectroscopy (SEDM),[6] to demonstrate that these materials are, without doubt, magnetically homogeneous. Furthermore, small angle neutron scattering data above T_c indicate that these glasses show minimal effects of chemical clustering.[7] Indeed, the most commonly cited evidence for magnetic inhomogeneity, namely bimodal hyperfine field distributions ($P(B_{hf})$), is readily shown to be a mathematical artifact of the fitting process.[8–10]

The data used to construct the magnetic phase diagrams of *a*-Fe-Zr (Ref. 1) and *a*-Fe-Ru-Zr (Ref. 11) are derived from samples prepared over a seven-year period by several different people on two different melt-spinning systems. The materials used ranged in age from months to five years at the time of measurement. In many cases, several years elapsed between the ac-susceptibility, Mössbauer, and μSR measurements. Despite this, the phase diagrams exhibit excellent agreement between the results derived from different techniques, and values obtained from samples of different ages prepared by different people are fully consistent. We observe a smooth evolution in behavior that depends only on composition, with no evidence for aging or handling effects.

Finally, both mean-field calculations[12] and numerical simulations[13] indicate that the magnetic behavior is controlled by the level of frustration present, and that this frustration affects T_c and T_{xy} equally strongly. Therefore T_c provides as good an indicator of basic sample condition as T_{xy}. It is trivial to determine T_c, and as noted above, ac-susceptibility, bulk magnetization, Mössbauer spectroscopy, neutron depolarization, and μSR all yield consistent values. Since our T_c’s agree with those of Kaptás *et al.*,[2] and we have eliminated both sample aging and preparation conditions as sources of variation, it is clear that only the reliability of the procedure used for determining T_{xy} remains as the origin of the apparent discrepancies.

II. METHOD RELATED ISSUES

μSR is perfectly suited to the study of transverse spin freezing. It is a bulk technique that probes both dynamic and static behavior simultaneously. We have confirmed that it yields correct values for T_c in both *a*-Fe-Zr (Ref. 1) and *a*-Fe-Ru-Zr (Ref. 11). Investigation of a polysaccharide iron complex allowed us to demonstrate that in a magnetically inhomogeneous system, μSR could be used to distinguish the nonmagnetic shells from the magnetic cores, both blocked and dynamic.[14] We observed no evidence for magnetic inhomogeneity in *a*-Fe-Zr. Finally, the *a*-Fe-Ru-Zr system exhibits a break in the temperature dependence of B_{hf} that can be used to locate T_{xy} in zero field using Mössbauer spectroscopy. These zero field values for T_{xy}, and those obtained by μSR are in perfect agreement.[11] Indeed, it was the obvious discrepancy between our zero-field μSR (Refs. 4,15) and applied field Mössbauer data[8] that led us to investigate the effects of an applied field on T_{xy}.

Determination of T_{xy} using μSR (both in zero field, and with an applied longitudinal field) involves locating a peak in the muon relaxation rate that is sampled at 10–15 temperature points and typically stands well over ten standard deviations above any background.[11,15] The changes in muon relaxation rate can be seen by eye, in the raw data, *without* recourse to computer analysis. SEDM in zero applied field has been used to check both the relaxation rates from μSR and the value of T_{xy}. Perfect agreement was obtained.[6] We also confirmed the location of T_{xy} by using a break in the temperature dependence of the static field experienced by the muons. The two values typically agree within 5 K. The observed behavior in both the static and dynamic signatures is fully consistent with numerical simulations.[13]

By contrast, determining T_{xy} from applied field Mössbauer spectroscopy in these magnetic glasses is far less straightforward. One must identify the field (or temperature) at which the second and fifth lines of the disorder-broadened six-line pattern disappear. $P(B_{hf})$ is strongly asymmetric (skewed towards low fields), its shape is not known *a priori*, and it is affected by the applied field. Lines two and five overlap the low-field tails of lines one and six, respectively, and their low-field tails in turn overlap lines three and four. The fitted intensity of lines two and five is strongly correlated with the form of the low-field tail used, or assumed, in fitting $P(B_{hf})$. While the average hyperfine field is generally a robust parameter, the deconvolution routines employed to determine $P(B_{hf})$ for magnetic glasses are prone to oscillations which render the form of $P(B_{hf})$ unreliable. Mathematical instabilities in the deconvolution procedure,[8,10] the effects of line overlap, and the limitations of first-order perturbation expansions[9] all combine to greatly reduce the reliability of the analysis used to track the intensities of lines two and five in the applied field spectra. Far fewer points are available, and the uncertainties on any given measurement are much larger than for μSR, so that actual, rather then quoted, ambiguities in T_{xy} are often 10 K or more (see Fig. 2 of Vincze *et al.*[16]). In addition, the external field, essential to the measurement, strongly affects the magnetic structure (it can suppress the transverse ordering entirely) so that $P(B_{hf})$ is necessarily changed. While it is true that, in principle, the demagnetizing field could be determined from the decrease in the average B_{hf}, the method relies on B_{hf} and the external field simply adding, with no field-induced changes in $P(B_{hf})$. Unfortunately, the severe effects of an applied field on the magnetic order, and thus on $P(B_{hf})$, make this assumption invalid and render the procedure unreliable. Furthermore, in both **Au**Fe (Refs. 17,18) and *a*-Fe-Zr,[19] the external field often leads to an *increase* in B_{hf}. Even for ferromagnetic *a*-$\mathrm{Fe}_{88}\mathrm{Zr}_{12}$ the fitted field shift has not saturated by 7 T.[20] Thus, the ‘‘inherent control’’ claimed as an advantage, does not actually exist.

There are significant inconsistencies in the data shown in Fig. 1 of the preceding comment[2] that serve to underline the problems encountered in using Mössbauer spectroscopy to follow the effects of an applied field on T_{xy}. First, their T_{xy} for $x=8$ extrapolates to essentially the same zero-field value as that of the more frustrated $x=7$ alloy. By contrast, our value lies nicely between those of the $x=7$ and $x=9$ samples. Second, their field dependence for $x=8$ is severely curved, yet for both $x=7$ and $x=9$, the dependence is linear. While there appear to be no strong predictions for the particular functional form of $T_{xy}(B)$, it would seem reasonable to expect that the behavior would be consistent from sample to sample, or at least that it would evolve in a consistent manner with composition. We recently checked the field dependence of T_{xy} for $10\geqslant x\geqslant 7$ using LF-μSR,[21] and confirmed that the same form is in fact observed across this composition range.

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