A study of spin dynamics in the a-$Fe_{90}Sc_{10}$ spin glass

J. van Lierop[a)]
Centre for the Physics of Materials, Physics Department, McGill University, 3600 University Street, Montreal, Quebec H3A2T8, Canada and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973-5000

D. H. Ryan
Centre for the Physics of Materials, Physics Department, McGill University, 3600 University Street, Montreal, Quebec H3A2T8, Canada

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

Spin dynamics in a-$Fe_{90}Sc_{10}$ have been examined with zero-field muon spin relaxation (ZF-μSR) and selective excitation double Mössbauer (SEDM) spectroscopy. Analysis of the μSR data yields $T_{sg} \sim 105$ K in agreement with both χ_{ac} and conventional Mössbauer data. SEDM shows fluctuation rates in good agreement with μSR, but reveals an abrupt change in relaxation behavior from spin-flip to diffusive above 90 K ($0.9T_{sg}$).
[DOI: 10.1063/1.1456437]

I. INTRODUCTION

Many iron-rich amorphous alloys exhibit magnetic frustration and noncollinear ordering due to the competition between majority ferromagnetic exchange and low levels of antiferromagnetic exchange. At high temperatures there is a paramagnetic to ferromagnetic phase transition (T_c) where the z component of moments orders collinearly. At lower temperatures, transverse spin freezing happens where x and y components of the moments freeze out (T_{xy}) and the system enters a noncollinear state with coexisting ferromagnetic and spin-glass order. If the frustration level is high enough, the two transitions merge to form a single transition (T_{sg}) from paramagnet to spin glass.

Amorphous iron-rich Fe–Sc alloys[1] are unique among the iron-rich early transition metal–iron glasses as they do not exhibit a strong composition dependence of the magnetic ordering temperature, which for the composition range of a-Fe_xSc_{100-x} with $89 \leqslant x \leqslant 91$ is essentially constant at ~ 100 K.[2] Applied-field Mössbauer and susceptibility measurements indicate that the material is a borderline spin glass, just sufficiently frustrated to destroy the ferromagnetic order.[3–5] The observation of a $B^{2/3}$ dependence to the average hyperfine field (from transmission Mössbauer results) has led to the suggestion that a-Fe–Sc undergoes a transition from a cluster spin glass at low temperatures to a superparamagnetic and then paramagnetic state with rising temperature.[6,7] However this argument has been questioned,[8] and applied-field transmission Mössbauer and magnetization results indicate that the a-Fe–Sc system undergoes a transition directly from spin glass to paramagnet.[5]

The main shortcoming with the the above experiments is the use of large external magnetic fields that can cause significant changes in the magnetic structure.[5] Indeed, in the a-Fe–Zr alloys, the effect of large applied fields has been shown to severely affect T_{xy}.[9] To avoid possible difficulties resulting from external applied fields, we have used two zero-field techniques to examine magnetic ordering in a-$Fe_{90}Sc_{10}$. Zero-field muon spin relaxation (ZF-μSR) spectroscopy provides a complete separation of the static and dynamic magnetic properties so that the magnetic fluctuations associated with the freezing of spin components[10] can be detected. Additionally, selective excitation double Mössbauer (SEDM) spectroscopy, another zero-field technique that provides distinct spectral signatures from static and time-dependent magnetism[11,12] has been used to determined the nature of the fluctuations below T_{sg}.

II. EXPERIMENTAL METHODS

The sample preparation is described in Ref. 5. Basic magnetic characterization was carried out on a commercial susceptometer/magnetometer. ^{57}Fe transmission Mössbauer spectra were collected on a constant acceleration spectrometer using a 1 GBq ^{57}CoRh source and fitted using an asymmetric Gaussian hyperfine field distribution. SEDM spectra were collected using a 2 GBq ^{57}CoRh source over an 8–10 day period (exciting line No. 1) on our improved SEDM spectrometer[12] and fitted using a model that incorporates both static disorder determined from transmission Mössbauer fit results and a two-level relaxation model.[13] Temperatures between 20 and 100 K were achieved using a vibration-isolated closed-cycle refrigerator. ZF-μSR spectra of the 16-mm-diameter, 200-mg-cm^{-2} thick sample of the same ribbons were collected at TRIUMF on the M13 beamline in a He-flow cryostat. ZF-μSR spectra were fitted using the full dynamic Kubo–Toyabe or spin-glass function.[14]

III. RESULTS AND DISCUSSION

Typical ZF-μSR spectra for a-$Fe_{90}Sc_{10}$ are shown in Fig. 1. Magnetic fluctuations couple to the muon spin and cause an exponential decay of the observed polarization. The

a)Electronic mail: johan@bnl.gov

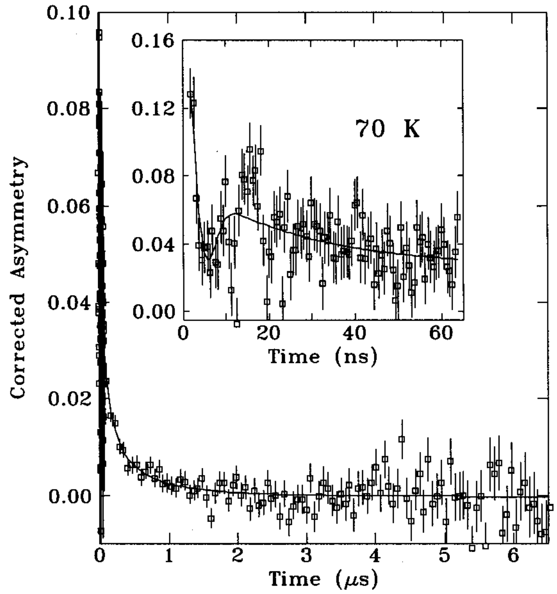

FIG. 1. Typical μSR spectrum for a-$Fe_{90}Sc_{10}$ at 70 K. Inset shows the early time region of the data where the static Kubo–Toyabe minimum with fast relaxation is observed. Solid lines are fits to functions described in Ref. 14.

inset shows that this exponential decay changes character at early times. A characteristic Kubo–Toyabe minimum is observed, indicating the presence of static magnetic order.[15] Since the static and dynamic μSR signals overlapped, the full dynamic Kubo–Toyabe and spin-glass functions[16] were used to fit the data. Results are shown in Fig. 2 where Δ/γ_μ represents the static rms field and the fitted relaxation rate describes a $1/T_1$ -type spin relaxation rate.

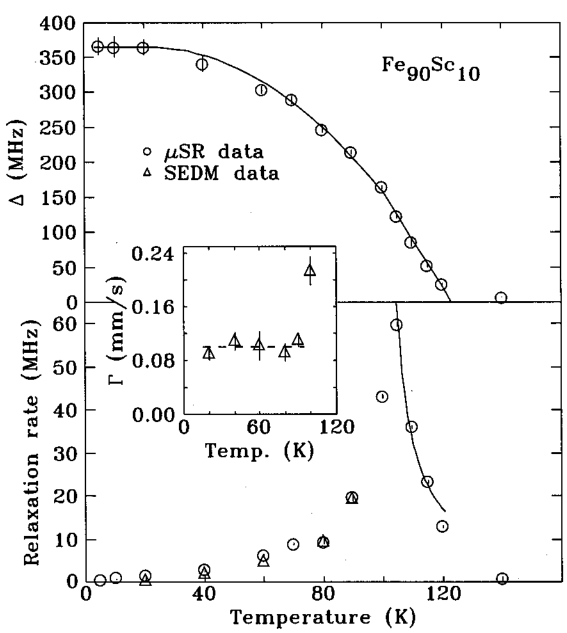

FIG. 2. Top: Temperature dependence of the static (△) relaxation rates for a-$Fe_{90}Sc_{10}$. Lines are guides to the eye. Bottom: Temperature dependence of the dynamic ZF-μSR relaxation rate (○) and SEDM relaxation rate (△). Inset shows the temperature dependence of the fitted line widths (Γ) of the SEDM spectra.

As the temperature approaches the paramagnetic to spin-glass transition from above, magnetic fluctuations occur over a continuum of time and length scales and ultimately diverge at 105 ± 5 K. Examining $\Delta(T)$ in Fig. 2, as we cool from 200 K there is a gradual, linear increase in Δ as some of the magnetic fluctuations slow into the measuring time window of the muon. $\Delta(T)$ changes once the transition is reached. On cooling past the transition a steady increase in Δ occurs until around 20 K where the static field is constant and all fluctuations have ceased. A more marked signature of moment behavior is provide by the ZF-μSR relaxation rate (○) in Fig. 2 that shows the divergence of magnetic fluctuations at all time and length scales at ~105 K. At temperatures below this, muon depolarization is dominated by the magnetic fluctuations slowing and becoming static. The temperature dependences of $\Delta(T)$ and $\lambda(T)$ are consistent with the behavior of a highly frustrated magnet that undergoes a single transition from paramagnet to spin glass at T_{sg}. The sharp cusp in $\lambda(T)$ (Fig. 2) is especially indicative of a magnetic transition, consistent with the observed $\lambda(T)$ transition from paramagnet to transverse spin freezing in the a-Fe–Zr[17] and a-Fe–Ru–Zr[18] systems. Additional evidence of a transition occurring around 100 K is provided by χ_{ac} data ($T_{sg}=100\pm1$ K) that measure fluctuation on a much slower time scale. The interpretation that a-Fe–Sc is a cluster spin glass is inconsistent with the fitting procedure used for the μSR data where a continuous distribution of hyperfine fields is assumed.[14] A magnetic cluster system will exhibit a distribution of relaxation rates as different sized clusters relax at different rates. Some of these clusters fluctuate either too fast or too slow to fall into the measuring time of the muon, and a more complex model is required to fit the depolarization of the muons.[19] Such behavior is not seen here.

SEDM spectra (Fig. 3) of the same a-$Fe_{90}Sc_{10}$ sample were collected to investigate the nature of the fluctuations below T_{sg}. At 20 K, the absence of magnetic fluctuations is confirmed by the observation of a single emission line in the SEDM spectrum, consistent with the ZF-μSR data [i.e., $\Delta(T)$ is at its maximum and λ is essentially zero]. The chemical and magnetic disorder leads to broadened absorption lines in the transmission spectrum 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[12]) however it is substantially narrower than that observed by transmission as only part of the field distribution is probed. The 20 K SEDM spectrum of the a-$Fe_{90}Sc_{10}$ is correctly described by the asymmetric Gaussian distribution, $P(B_{hf})$, of hyperfine fields determined from the 20 K transmission spectrum. This demonstrates that we have a complete description of the static magnetic disorder to be convolved with the moment fluctuations which develop as T_{sg} is approached.[11,12]

An unambiguous SEDM spectral signature results from spin dynamics below T_{sg}, seen in the spectra up to 90 K ($0.9T_{sg}$) in Fig. 3. Only line No. 1 is excited, but emission from line No. 6 is clearly present as well. This is a direct indication that the magnetic field at the excited nucleus has

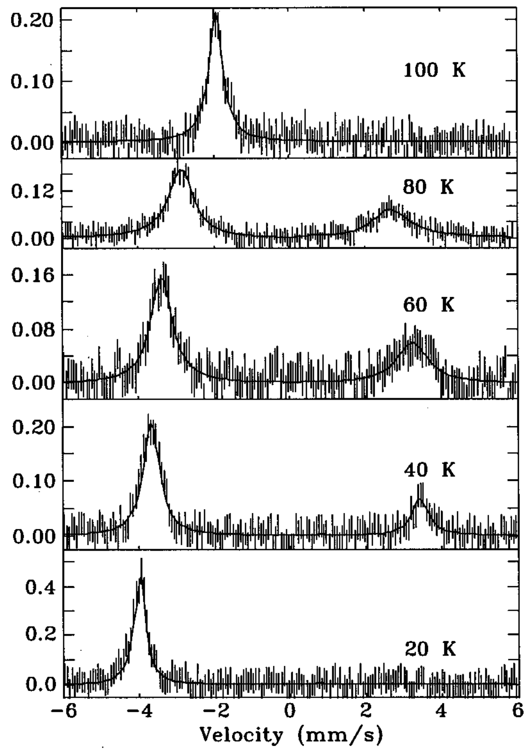

FIG. 3. SEDM spectra of a-$Fe_{90}Sc_{10}$. At each temperature the pump energy was centered on the line at the left.

reversed direction during the lifetime (97 ns) of the excited state.[11] No amount of static disorder can lead to this additional line. Most striking is what happens to the SEDM spectrum at 100 K. The echo line (No. 6) has disappeared (Fig. 3), however, the fitted line width is much broader then that of the lower temperature SEDM fits (inset to Fig. 2). This behavior is consistent with the fluctuations changing behavior from a spin-flip to a continuous easy-axis relaxation.[20]

Results of fits to the SEDM data are shown in Fig. 2. ZF-μSR is, in principle, sensitive to all forms of magnetic relaxation and has a wider frequency window than the Mössbauer effect, however the fitted SEDM and ZF-μSR relaxation rates up to 90 K are in perfect agreement. This consistency between results leads to the conclusion that both SEDM and μSR are measuring the same time-dependent phenomena. Additional evidence is provided by the fitted line width of the SEDM spectra. With the source and detector line widths characterized using standard samples,[12] and the static disorder for the a-$Fe_{90}Sc_{10}$ determined from transmission Mössbauer spectra, we observe a temperature-independent sample line width of 0.10 ± 0.01 mm/s until 90 K that is fully consistent with the lifetime of the excited state of the Mössbauer nucleus. At 100 K ($0.95T_{sg}$) the fluctuation mechanism seems to change from spin-flips to a continuous easy-axis relaxation. This is suggested by the sudden increase in the fitted line width of the 100 K SEDM spectrum. The $P(B_{hf})$ does not completely describe the magnetism of the system, and the fitted line width is too large to reflect only the lifetime of the Mössbauer nucleus excited state. The static field determined from the μSR results (Δ) at 100 K is large enough to guarantee hyperfine field splittings that can be observed with SEDM,[12] and a simulated transmission spectrum with a ~40 MHz (spin-flip) relaxation rate is not consistent with the measured transmission spectrum at 100 K. This is strong evidence that the features observed in the SEDM spectrum at 100 K are due to a real change in the magnetic dynamics and that the clear increase in linewidth must be due to dynamic effects.

In conclusion, a-$Fe_{90}Sc_{10}$ undergoes a paramagnetic to spin-glass transition at $T_{sg} \sim 105$ K. We find no evidence for cluster glass behavior in either the μSR or the SEDM data. Additionally, the fluctuations from the moments slowing into the measuring time window of both the muon and Mössbauer effect have been detected at T_{sg}. Moment flips of 180° have been explicitly identified as the dominant fluctuation mechanism up until 0.95 T_{sg}, where the fluctuations change behavior from spin-flip to continuous easy-axis relaxation. 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.

ACKNOWLEDGMENTS

This work was supported by grants from the Natural Sciences and Engineering Research Council of Canada, Fonds pour la Formation de Chercheurs et l'Aide à la Recherche, Québec, The Australian Nuclear Science and Technology Organization, and in part under the auspices of the U.S. Dept. of Energy, Division of Materials Sciences, Office of Basic Energy Sciences under Contract No. DE-AC02-98CH10886.

[1] R. K. Day, J. B. Dunlop, C. P. Foley, M. Ghafari, and H. Pask, Solid State Commun. **56**, 843 (1985).
[2] D. Wiarda and D. H. Ryan, J. Appl. Phys. **76**, 6189 (1994).
[3] H. Ren and D. H. Ryan, J. Appl. Phys. **73**, 5494 (1993).
[4] H. Ma, Z. Wang, H. P. Kunkel, G. Williams, D. H. Ryan, and J. O. Ström-Olsen, J. Magn. Magn. Mater. **104-7**, 89 (1992).
[5] H. Ren and D. H. Ryan, Phys. Rev. B **51**, 15885 (1995).
[6] M. Ghafari, R. K. Day, J. B. Dunlop, W. Keune, and A. C. McGrath, Hyperfine Interact. **54**, 533 (1990).
[7] M. Ghafari, B. Stahl, S. H. Banihashemi, M. Müller, and H. Hahn, Hyperfine Interact. **126**, 15 (2000).
[8] L. E. Wenger and J. A. Mydosh, Phys. Rev. B **29**, 4156 (1984).
[9] D. H. Ryan, J. van Lierop, M. E. Pumarol, M. Roseman, and J. M. Cadogan, Phys. Rev. B **63**, 140405 (2001).
[10] J. R. Thomson, H. Guo, D. H. Ryan, M. J. Zuckermann, and M. Grant, Phys. Rev. B **45**, 3129 (1992).
[11] J. van Lierop and D. H. Ryan, Phys. Rev. Lett. **86**, 4390 (2001).
[12] J. van Lierop and D. H. Ryan, Rev. Sci. Instrum. **72**, 3349 (2001).
[13] J. van Lierop and D. H. Ryan Phys. Rev. B **65**, 104402 (2002).
[14] J. van Lierop, D. H. Ryan, and J. Gallego (unpublished).
[15] R. Kubo and T. Toyabe, in *Magnetic Resonance and Relaxation*, edited by R. Blinc (North-Holland, Amsterdam, 1967), p. 810.
[16] Y. J. Uemura, T. Yamazaki, D. R. Harshman, M. Senba, and E. J. Ansaldo, Phys. Rev. B **31**, 546 (1985).
[17] D. H. Ryan, J. M. Cadogan, and J. van Lierop, Phys. Rev. B **61**, 6816 (2000).
[18] D. H. Ryan, J. M. Cadogan, and J. van Lierop, Phys. Rev. B **62**, 8638 (2000).
[19] J. van Lierop, D. H. Ryan, M. E. Pumarol, and M. Roseman, J. Appl. Phys. **89**, 7645 (2001).
[20] A. M. Afanas'ev and V. E. Sedov, Hyperfine Interact. **56**, 1425 (1990).