LA-UR-86-2760

Received by OSTI

SEP 0 3 1986

CONF-8606131--8

Los Alamos National Laboratory is operated by the University of California for the United States Department of Energy under contract W-7405-ENG-36

LA-UR--86-2760

DE86 015321

TITLE: μ^+SR IN AMORPHOUS SPIN GLASSES $Pd_{75}Fe_5Si_{20}$ AND $Pd_{75}Fe_5P_5$

AUTHOR(S): J. H. Brewer, D. P. Spencer, C. Y. Huang, Y. J. Uemura, and H. S. Chen

SUBMITTED TO: Muon Spin Relaxation, Uppsala Sweden, June 23-27, 1986, Proceedings of

μ^+SR IN AMORPHOUS SPIN GLASSES $Pd_{75}Fe_5Si_{20}$ AND $Pd_{75}Fe_5P_{20}$

J.H. Brewer and D.P. Spencer,* TRIUMF and Departments of Physics and Chemistry, University of British Columbia, Vancouver, B.C., Canada V6T 2A3

C.Y. Huang, Los Alamos National Laboratory, Los Alamos, New Mexico, USA 87545

Y.J. Uemura, Brookhaven National Laboratory, Upton, New York, USA

H.S. Chen, AT&T Bell Laboratories, Murray Hill, New Jersey, USA

The zero-field muon spin relaxation function $G_{zz}(t)$ has been measured as a function of reduced temperature $t = T/T_g$ in the amorphous metallic spin glasses $Pd_{75}Fe_5Si_{20}$ and $Pd_{75}Fe_5P_{20}$. The results are in qualitative agreement with earlier measurements on dilute alloy spin glasses, including an onset of static order below T_g and a $[t/(t-1)]^2$ dependence of the correlation time τ_c above T_g. Both samples have the same $\tau_c(t)$ above T_g and almost identical static width $\Delta_s \rightarrow \Delta_0 \approx 43\ \mu s^{-1}$) as $T \rightarrow 0$, but the t-dependence of Δ_s near T_g differs markedly.

Considerable previous μ^+SR work on dilute-alloy spin glasses/1/ has revealed several "universal" characteristics of muon relaxation in these systems. First, above T_g every μ^+ sees a fluctuating local field due to nearby paramagnetic moments, but (as long as the muons do not diffuse) the sequence of local fields seen is selected randomly from a *different* distribution for each muon, depending upon how close it is to the moment(s) furnishing the field. The individual local distributions may be treated as gaussian, while the instantaneous overall distribution must be lorentzian. The distribution of gaussian widths that generates an overall lorentzian was described by Uemura;/1/ if the moments generating the local fields all fluctuate at the same rate, the resulting muon relaxation function has a "root exponential" behaviour $G_{zz}^{SG}(t) = \exp[(-\lambda t)^{\frac{1}{2}}]$ in the limit of fast fluctuations. The data so far are in agreement with this picture, and all yield a temperature dependence of the fluctuation rate $1/\tau_c$ consistent with a $\tau_c \sim [t/(t-1)]^2$ dependence, where t is the "reduced temperature" $t = T/T_g$.

Second, below T_g some paramagnetic moments stop fluctuating before others (depending upon their local couplings), with the result that the local field seen by a muon appears on average to have a static component and a fluctuating component. This is conventionally described phenomenologically by an

Edwards-Anderson/2/ "order parameter" Q and a single correlation time τ_c so that the autocorrelation function of the local field B_l has the putative form

$$<B_l(0)\ B_l(t)> = <B_l(0)^2>[Q + (1 - Q)\ \exp(-t/\tau_c)]. \qquad [1]$$

Of course, this is a naive empirical description of the "freezing" of different moments, in which the fact that different moments freeze at different temperatures is absorbed into the temperature dependence of Q, and the probable concomitant distribution of correlation times is ignored. However, this picture has served to fit the existing data admirably, with the possible exception of the region very close to T_g.

We have now extended these studies to the *amorphous* spin glass systems $Pd_{75}Fe_5Si_{20}$ and $Pd_{75}Fe_5P_{20}$, with remarkably similar results.

The surface muon beams of the M9 and M20 channels at TRIUMF/4/ were essential to this experiment in several ways. First, the short range straggling ($\approx$ 30 mg/cm^2) of 4.1 MeV μ^+ allowed all of the incident muons to be stopped in the spin-glass material, which was in the form of continuous ribbons 0.5-2 mm wide and < 0.1 mm thick, crumpled into "nest"-shaped targets about 50-500 mg/cm^2 thick and 2-5 cm in diameter, enclosed in thin Mu-metal foil to exclude stray magnetic fields. Second, because no anti-counter is needed with a surface muon beam, the resultant time spectra $F(t)$ and $B(t)$ (for "Forward" and "Backward" muon-decay positron detectors, respectively) contained undistorted data at very early times (within < 10 ns after the muon's arrival in the target). Finally, a 3 m Wien filter was used to eliminate an otherwise problematic positron contamination; as a result, $F(t)$ and $B(t)$ had very low backgrounds. These backgrounds were determined from t < 0 bins and subtracted from $F(t)$ and $B(t)$ to form the "pure" muon-decay time spectra $F'(t)$ and $B'(t)$, which were then combined to form the "μSR signal"

$$A(t) = [B'(t) - F'(t)]/[B'(t) + F'(t)], \qquad [2]$$

which was the quantity fitted to the theoretical formula below.

Temperatures were regulated to within about 0.2 K (near T_g) in a Janis Super-varitemp He-flow cryostat. All the data for a given sample were taken in one series of runs on the same day, without touching anything but the temperature control, and the temperature points were taken in random order. This procedure was essential to assure the constancy of "systematic" empirical variables, which could then be held fixed (see below).

The *nonmagnetic* amorphous metal $Pd_{80}P_{20}$ was also investigated in zero and transverse field. The substitution of P for Si in this alloy allowed us to check the monmagnetic version for μ^+ diffusion as manifest in "motional narrowing" of μ^+ relaxation by P nuclear dipoles. We found no evidence for fast diffusion; in fact, the relaxation of the muons (which appears gaussian and is never very fast) seems to *increase* slightly with increasing T (at temperatures well above T_g), suggesting that the muons migrate from shallow traps at low T to (nearby) deeper traps at higher T. In any case, μ^+ hopping is much too slow to confound the determination of local field correlation times in the spin glass samples.

The ZF-μSR spectra $A(t)$ for $Pd_{75}Fe_5Si_{20}$ and $Pd_{75}Fe_5P_{20}$ spin glass samples were fitted to the theoretical function

$$A(t) = [(a+1)A_0G_{zz}^{sg}(T) - (a-1)]/[(a+1) - (a-1)A_0G_{zz}^{sg}(t)], \quad [3]$$

where A_0 is the empirical maximum asymmetry, a is the empirical normalization ratio N_F^0/N_B^0 (commonly called the "baseline") and $G_{zz}^{sg}(t)$ is the spin glass relaxation function given by Refs. /1,3/:

$$G_{zz}^{sg}(t,\Delta_0,Q,\tau_c) = \frac{1}{3}\exp(-\sqrt{D}) + \frac{2}{3}\left[1 - \frac{S}{\sqrt{D+S}}\right]\exp(-\sqrt{D+S}) \quad [4]$$

where $D = 4\,\Delta_d^2 t\tau_c$, $S = \Delta_s^2 t^2$, $\Delta_s^2 = Q\,\Delta_0^2$, $\Delta_d^2 = \Delta_0^2 - \Delta_s^2$, Δ_0 is the static width at $T\rightarrow 0$, Q is the mean-field order parameter /1,2/ and τ_c is the correlation time for the fluctuating local field (presumed to be the same as the Fe spin correlation time). This function holds for all T provided (i) $1/\tau_c >> \Delta_0$ everywhere; and (ii) the general assumptions of Uemura /1,3/ in constructing $G_{zz}^{sg}(t)$ are valid for amorphous systems.

The rather clumsy form of Eq. (3) is necessary to correctly account for differences in solid angle, counter efficiency, etc., between the two telescopes. The empirical asymmetry A_0 was first allowed to vary freely; then the weighted mean of A_0 taken from $T > T_g$ runs was used as a fixed parameter in the final "global" analysis, with the logic that A_0 is a function only of the beam polarization, target and counter geometry, which were not changed. The baseline a was always allowed to vary freely, since such systematic effects as intermittent discriminators can cause a to fluctuate dramatically; in fact only a slight monatonic increase of a with T below $\approx$12 K was observed, probably due to the increasing density of the helium exchange gas at lower T, which slightly changes the muon stopping distribution.

Thanks to the very early time information available with surface muons, the initial rapid decay of $A(t)$ below T_g was directly observed (see Fig. 1), fixing the effective static width Δ_s and allowing the use of Eq. (4) to determine Δ_0, Q and τ_c.

The resultant values of Δ_s are plotted in Fig. 2; the tendancy toward a value $\Delta_0 \approx 43\ \mu s^{-1}$ at $T = 0$ is apparent. Therefore the final "global" analyses were performed with the

$T = 0$ static width fixed to this value. With this ambiguity removed it was also possible to extract reliable values of the correlation time τ_c below T_g and check self-consistently whether the criterion $1 \gg \Delta_0 \tau_c$ is valid.

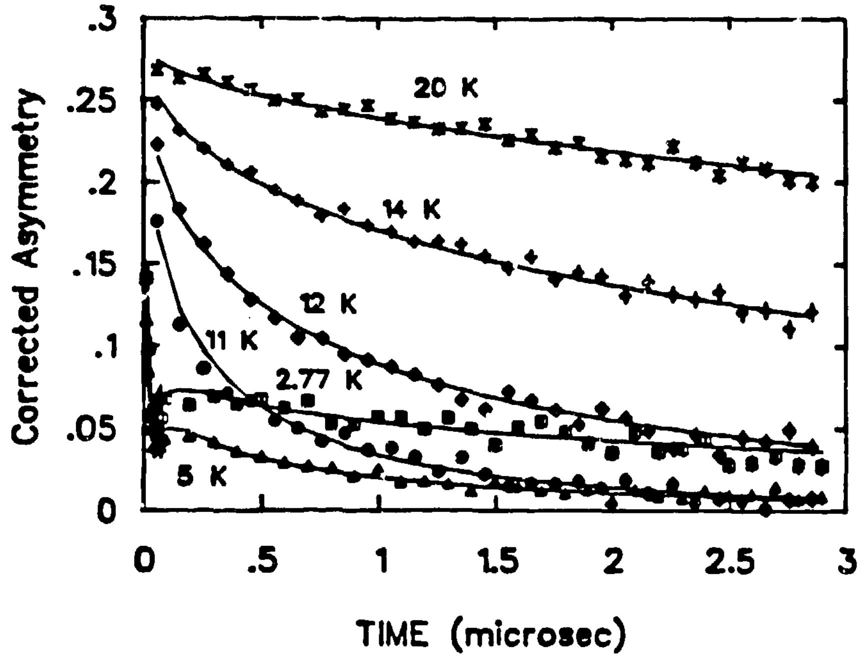

Figure 1. Examples of ZF-μ^+SR time spectra $A(t)$ for the amorphous spin glass $Pd_{75}Fe_5P_{20}$ at several temperatures. The fitted curves are $A(t) = A_0\, G_{zz}^{sg}(t)$.

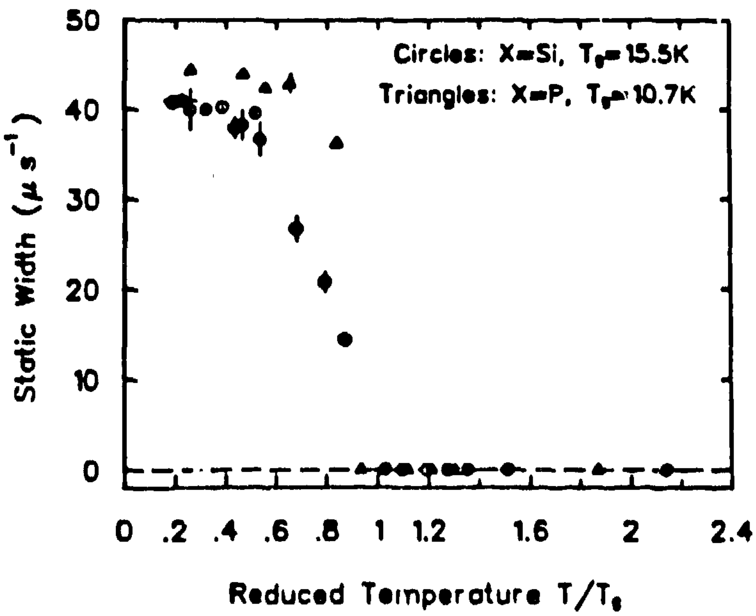

Figure 2. Dependence of the static width $\Delta_s = Q\Delta_0$ upon the reduced temperature $t = T/T_g$ in the two spin glass samples $Pd_{75}Fe_5Si_{20}$ [open circles, T_g = 15.5(5)K] and $Pd_{75}Fe_5P_{20}$ [triangles, T_g = 10.7(9)K].[9]

The largest value of $\Delta_0\tau_c$ obtained in this way was ≈ 1/6, which is uncomfortably close to the region in which the model [4] breaks down, but perhaps still meaningful. Ideally the data for $T < T_g$ should be fitted with the "time-dependent order" model of Uemura,/2/ but unfortunately that model has not yet been rendered into a "fittable" form.

Above T_g the best fit was always obtained with $Q = 0$, as expected; in fact, the disappearance of Q was used to confirm the location of T_g. This removed the ambiguity of the extra parameter and allowed very precise determination of τ_c (given, of course, the fixed value of $\Delta_0 = 43\ \mu s^{-1}$) above T_g. The "root exponential decay" characteristic of spin glasses in the dynamical limit /1,2/ is beautifully displayed in all spectra above T_g, several of which are plotted in Fig. 1 for early times.

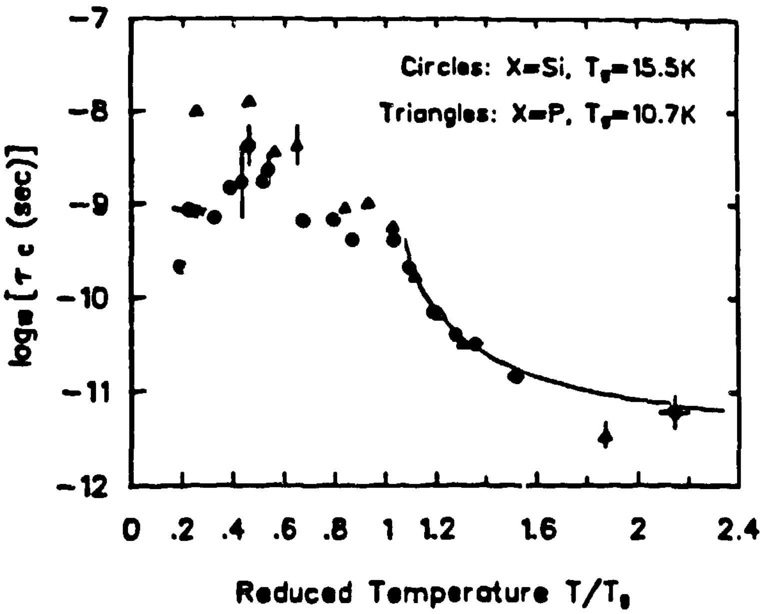

Figure 3. Correlation time τ_c as a function of reduced temperature t for the two spin glass samples $Pd_{75}Fe_5Si_{20}$ [T_g = 15.5(5)K; open circles] and $Pd_{75}Fe_5P_{20}$ [T_g = 10.7(9)K; triangles].

The correlation time τ_c is displayed in Fig. 3 as a function of reduced temperature $t = T/T_g$, including the $T < T_g$ results for completeness; the solid line shows the excellent agreement above T_g with a power law

$$\tau_c \sim \{t/(t-1)\}^2 \quad \text{or} \qquad [5a]$$

$$\tau_c \sim [T/(T-T_g)]^2 \qquad [5b]$$

as observed earlier for dilute-alloy spin glasses./1/ A recent theoretical estimate of the critical exponent by Huber/5/ is in good agreement with our results.

Perhaps the most striking aspect of Fig. 3 is the fact that $\tau_c(t)$ has not only the same shape, but the same *values* at the same

t for both samples. This is a strong indication not only that the "scaling universality" observed for dilute-alloy spin glasses/1/ applies to amorphous systems as well, but also that the corresponding "reduced temperature" t is really a "fundamental" parameter characterizing these spin glasses. The marked difference in the t-dependence of Δ_s for the two samples is equally strong evidence that the *range* of the magnetic order is profoundly influenced by the identity of "X" (P or Si).

Every experiment should have one result which disagrees dramatically with expectations, and while this one very nearly violates that rule, it did not disappoint us. Most runs are fitted superbly by Eq. (4), but the $Pd_{75}Fe_5Si_{20}$ run at 14.2 K, which was both the closest to T_g and the highest statistics (1.8 M events/spectrum), does not. This means that the model breaks down very near to T_g, which may be the first hint of something unusual in spin glass ordering at the transition temperature. Longitudinal field experiments /1,2/ could help to clarify the mechanisms at work, but a new model for $G_{zz}^{sg}(t)$ is probably necessary.

A good candidate for such a model springs from the "stretched exponential" autocorrelation function used to describe translational order in glassy materials./6,7/ We have so far characterized the local field B_l in terms of the simple autocorrelation function [Eq. (1)] in which the order parameter Q -- a sort of microscopic local magnetization -- is used as an empirical measure of the fraction of the randomly-oriented B_l which is static, as opposed to fluctuating with a correlation time τ_c. This is very similar to using the stretched exponential autocorrelation function

$$\langle B_l(0)\, B_l(t)\rangle = \langle B_l(0)^2\rangle \exp[-(t/\tau_c)^{1-n}] \qquad [6]$$

with $n < 1$. Such a function has the same qualitative shape as that of Eq. (1) -- i.e., an initially exponential decay with a nearly static "tail" at long times. It is not clear whether the data available would allow us to distinguish between these two descriptions, and in any case there is as yet no analytical formulation of the relaxation functions $G_{zz}(t)$ that would result from such a model.

The different t-dependence of Q in the two samples is surprising, particularly in view of their virtually identical Δ_0 and $\tau_c(t)$. This could be interpreted as a difference between indirect Fe-Fe coupling through Si or P intermediaries, affecting the dynamic range of spin-glass ordering.

This work was supported by the Natural Sciences and Engineering Research Council of Canada, by the Canadian National Research Council (through TRIUMF) and by the U.S. Department of Energy.

REFERENCES

* *Current address*: Dept. of Radiation Oncology, University of Chicago Medical Center, Box 440, 5841 S. Maryland Ave., Chicago, Ill., USA 60637.

/1/ Y.J. Uemura, T. Yamazaki, D.R. Harshman, M. Senba and E.J. Ansaldo, Phys. Rev. B31, 546 (1985).
/2/ S.F. Edwards and P.W. Anderson, J. Phys. F5, 965 (1975); *ibid.*, 6, 1927 (1976).
/3/ Y.J. Uemura, Ph.D. thesis, Univ. of Tokyo (1982).
/4/ J.L. Beveridge, J. Doornbos, D.M. Garner, D.J. Arseneau, I.D. Reid and M. Senba, Nucl. Instr. & Methods in Phys. Research A240, 316 (1985).
/5/ D.L. Huber, Phys. Rev B 31, 4420 (1985).
/6/ K.L. Nagi and C.T. White, Phys. Rev. B 20, 2475 (1979); A.S. Arrott and B. Heinrich, J. Magn. Magn. Mater. 31-34, 1084 (1983).
/7/ R.V. Chamberlin, G. Mozurkeiwich and R. Orbach, Phys. Rev. Lett. 52, 867 (1984).