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DYNAMICS OF SPIN GLASSES DETECTED BY MUON SPIN RELAXATION

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Abstract

Muon spin depolarization measurements in dilute-alloy spin glasses AuFe, CuMn and AgMn are surveyed. After a brief review of earlier transverse-field experiments, we introduce the principles and results of zero-field μ SR which determined absolute values of the correlation time τ_c of M_n (or Fe) moments from $\tau_c \sim 10^{-10}$ sec (at $T \sim 1.2T_g$) to $\tau_c \sim 10^{-5}$ sec (at $T \sim 0.5T_g$) with a rapid change of τ_c around the susceptibility-cusp temperature T_g . Finally, we describe details of our recent μ SR measurements in finite longitudinal magnetic fields, which generally confirmed the zero-field results and revealed a coexistence of fast and slow spin fluctuations just below T_g .

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Muon spin rotation and relaxation (μ SR) measurement has extensively been applied to the research of solids [1,2]. Depolarization of muon spins reflects the amplitude, randomness, and dynamical fluctuations of local magnetic fields in the specimen, and thus, muons give us important information on magnetic materials as a microscopic probe. In this article, we will show how the μ SR method attacks the spin glass problem [3-6], focusing on positive muon (μ^+) experiments in typical dilute-alloy spin glass systems as AuFe or CuMn (around 1 at.%).

I. TRANSVERSE-FIELD μ SR (TF- μ SR) IN SPIN GLASSES

The first μ SR experiment in spin glasses was carried out by Murrick, Flory and Kossler [7] in CuMn and AuFe. They measured the muon spin precession by applying transverse external magnetic fields of $85 \sim 1890$ Oe, and observed the depolarization of muon spins from the damping of the precession envelope, shown in Fig. 1(a). They found a rapid change of the depolarization rate around T_g , attributed it to the static component of random local fields on μ^+ , and deduced the temperature dependence of its averaged width W , shown in Fig. 1(b), which exhibits a sharp increase at T_g when approached from higher temperatures. Recently such TF- μ SR measurements have been performed also by Emmerich et al. [8] in CuMn and by Brown et al. [9] in AgMn, yielding results qualitatively similar to the earlier experiment.

The principle of this TF- μ SR is illustrated in Fig. 2(a). Applying a transverse external magnetic field H_T perpendicularly to the initial direction of muon spins, we stop the polarized muon beam in the specimen (dotted piece of Fig. 2). With the mean lifetime $\tau_\mu = 2.2$ μ sec, positive muon (μ) decays into a positron (e^+) emitting it preferentially to the muon spin direction. We observe the time spectrum of these positrons (by the counter E), which exhibits the muon spin precession around H_T

$$E(t) = N \cdot \exp(-t/\tau_\mu) \{1 + A \cdot G_X(t) \cos(\gamma_\mu H_T t + \phi)\}, \quad (1)$$

where A is the initial asymmetry of muon decay (usually ~ 0.2), γ_μ is the gyromagnetic ratio $2\pi \times 1.35 \times 10^4$ /sec/Oe of μ^+ , and $G_X(t)$, the transverse spin relaxation function, represents the depolarization of muon spins in the specimen.

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In TF- μ SR, which exactly corresponds to the " T_2 " measurement in magnetic resonance, spin depolarization is caused both by static inhomogeneous local fields and by fluctuating random fields, thus it is difficult to distinguish the static and dynamical effects. Furthermore, $G_X(t)$ is observed only upon the sinusoidal modulation, so it is usually difficult to deduce the detailed shape of the relaxation function in this experiment. Therefore, all the TF- μ SR results were fitted by assuming the simplest case, $G_X(t) = \exp(-t/T_2)$. In all three TF- μ SR experiments, $1/T_2 [= \gamma_\mu W$ in Fig. 1(b)] keeps rather constant values at lower temperatures ($T \sim 0.7T_g$), which are well explained by the static random fields on μ^+ from surrounding Mn or Fe moments. Though they enlightened only little about dynamical aspects, these experiments provided some important microscopic information around T_g , which we will revisit later.

II. ZERO-FIELD μ SR(ZF- μ SR) IN SPIN GLASSES

In 1978 at TRIUMF (Vancouver), we established [10,11] a completely different method to observe muon spin depolarization. As shown in Fig. 2(b), we measure the decay positrons by forward (EF) and backward (EB) counters, applying a longitudinal magnetic field H_L or even without applying any field. As long as muon spins keep the polarization the asymmetry EF:EB is about 1.2:0.8 (after some corrections for the solid angle factors), but it becomes 1:1 when muon spins are depolarized.

$$EF(t) = N \cdot \exp(-t/\tau_\mu) \{1 + A \cdot G_Z(t)\}$$

$$EB(t) = N \cdot \exp(-t/\tau_\mu) \{1 - A \cdot G_Z(t)\} \quad (2)$$

Thus the longitudinal muon spin relaxation function $G_Z(t)$ can directly be observed from the ratio $(EF-EB)/(EF+EB)$. We first applied this method to " T_1 " measurements in MnO [10] and in MnSi [12], and also to some basic studies of relaxation phenomena in nuclear dipolar systems [13,14].

To realize μ^+ in dilute-alloy spin glasses, the following information is helpful. (a) No effect of muon diffusion: μ^+ is known to rest stationarily at an interstitial site of pure Cu between $T = 2 \sim 70$ K [15], and similar situations can be expected in other fcc noble metals (Au or Ag [16]). (b) Random and uniform occupation of interstitial sites by muons: This assumption well explained the TF- μ SR data [7-9], and will be confirmed also in ZF- μ SR. (c) Dominant atomic dipolar fields on μ^+ :

Because of the weak coupling between conduction electrons and muon spins (Knight shift ~ 50 ppm for μ^+ in Cu [17]) is more than 40 times smaller than ~ 2300 ppm for the host Cu nuclei [18]), averaged RKKY field on μ^+ in 1 at.% CuMn is only around 10 G. Atomic dipolar field is around 100 G, and nuclear dipolar field from Cu nuclei is around 4 G.

Static random local field on a probe from dilute moments in such a system has a Lorentzian distribution ($HWHM$ a/γ_μ) for dilute limit [19,20] (effective for Mn or Fe concentrations $c \lesssim 3$ at.%).

$$P^L(H_i) = (\gamma_\mu/\pi) \cdot \{a/(a^2 + \gamma_\mu^2 H_i^2)\}, \quad (i=x,y,z) \quad (3)$$

and this will be a good approximation even for muons surrounded by the "ordered" moments [21]. When local field is static, the zero-field muon spin relaxation function has a simple form

$$g_Z(t) = \frac{1}{3} + \frac{2}{3}(1-at)\exp(-at) \quad (4)$$

as calculated by Schenck [22] and Kubo [23]. Here the 1/3 component represents muons stopped at a site where local field is rather parallel to the initial polarization direction (z-axis), while the damped oscillation of the 2/3 component is caused by the x- and y-components of random fields.

The effect of dynamical fluctuations of Mn (or Fe) moments was considered by Uemura [24,25], who proposed to decompose $P^L(H_i)$ into some sub-distributions, each of which associated to different muon sites as shown in Fig. 3. When surrounding moments fluctuate randomly, the local field at each site will be modulated within such a sub-distribution.

For simplicity, a Gaussian shape was assumed to represent each sub-distribution, and the muon spin relaxation function at each site was calculated by using the strong collision approximation for the dynamical modulation of random fields with the rate ν [14,25]. Then by taking the spatial average over the probability to find each muon site, the zero-field spin relaxation function $G_Z^G(t, a, \nu)$ for muons in a spin glass was analytically deduced as shown in Fig. 4. A recent computer simulation work [26] shows a good agreement with this treatment. Relaxation functions for TF- μ SR and longitudinal-field μ SR (LF- μ SR) can be calculated in a similar way [24,27], as can the line shape of host NMR in spin glasses.

Fig. 4 demonstrates some nice features of ZF- μ SR in spin glasses:

- i) In ZF- μ SR, static case (broken line of Fig. 4) can clearly be distinguished from dynamical modulations, thanks to the characteristic 1/3 component. ii) This 1/3 component reflects slow modulations ($v/a < 1$) of local fields as [25]

$$G_z(t) \sim \frac{1}{3} \exp(-\frac{2}{3} vt) \quad (5)$$

since local fields may no longer keep muon polarization after a modulation. Within the time window $t \leq 10 \mu\text{sec}$ (~ 5 mean muon lifetimes) of μ SR, we can observe $v \sim 10^5 - 10^7 / \text{sec}$ from this effect. Note that any dynamical effect slower than $\sim 20 \mu\text{sec}$ looks "static" in μ SR.

- iii) The "narrowing" effect, starting at $v/a \sim 2$, changes the shape of $G_z^S(t)$ very slowly with increasing v/a , due to a wide variety of local fields at different sites. This enables us to measure up to $v \sim 10^{11} / \text{sec}$. Thus in ZF- μ SR, we can deduce the correlation time τ_c of Mn (or Fe) moments, as $\tau_c = 1/v$, in a very wide range between 10^{-5} sec and 10^{-11} sec.

Zero-field μ SR, measured without perturbing the system by the applied magnetic fields, is an ideal method to probe spin glasses, in which the "cusp" of susceptibility is known to be appreciably rounded by an external field as small as 200 Oe [3]. At TRIUMF, we performed ZF- μ SR in AuFe (1.0 and 1.4 at.%) and CuMn (1.1 at.%) [28], and succeeded to observe a detailed shape of relaxation functions as shown in Fig. 5. In AuFe , where nuclear dipolar moments of Au are negligibly small, the observed shape of $G_z(t)$ fitted well to the model function $G_z^S(t)$, as illustrated by the solid lines of Fig. 5. The characteristic line shape for the almost static case was observed at $T \sim 0.5 T_g$, a slow modulation (τ_c in μsec region) was found at $T \sim 0.7 T_g$, depolarization rate was changed rapidly by the narrowing effect around T_g , and almost no relaxation was observed above $T = 1.3 T_g$, suggesting a fast spin fluctuation of Mn moments at higher temperatures. Similar results were obtained in CuMn (ZF- μ SR data is included in Fig. 7 of the next section), except for the effect of nuclear dipolar fields of Cu which slightly modified the observed line shape [28]. The characteristic shape of $G_z(t)$ for static nuclear dipolar fields [14,29], observed at $T > 1.3 T_g$, assumed that μ^+ is not diffusing. The averaged local field a/γ_μ (~ 61 G in 1% AuFe , ~ 110 G in CuMn), deduced from the data, agreed well to the calculated values of static atomic dipolar fields [25] from Mn or Fe moments.

This supports the assumption of uniform occupation of interstitial sites by muons, and so does the observed static line shape which indicates the Lorentzian local field.

The correlation time τ_c of Fe (or Mn) moments, thus deduced from ZF- μ SR spectra, is shown in Fig. 6, where we also included the recent results of ZF- μ SR in AgMn (1.6 at.%) by Brown et al. [30] obtained at LAMPF (Los Alamos). All the data of τ_c in three different systems can be scaled with the reduced temperature T/T_g (c.f., $T_g = 12.8\text{K}$ for 1.4% AuFe , 7.6K for AgMn) as shown in Fig. 6(b); another "scaling law" [31] in spin glasses. τ_c exhibits a rapid change around T_g and varies from 10^{-6} to 10^{-10} sec within a small interval $0.8 T_g < T < 1.2 T_g$. The sharpness of this change corresponds to the index $N \sim 2$ when we assume the power law behavior above T_g , while the slope of τ_c around T_g gives $E_a \gtrsim 20 kT_g$ and to $\lesssim 10^{-15}$ sec when fitted with the Arrhenius law $\tau_c = \tau_0 \exp(E_a/kT)$. τ_c at $T = T_g$ is around $10^{-8} \sim 10^{-9}$ sec for all the specimens. A slow spin fluctuation ($\tau_c \sim 10^{-5} - 10^{-6}$ sec) is observed at $T \leq 0.7 T_g$. To grasp a general feature, we started with the simplest model function $G_z^S(t)$, which assumes a single value of τ_c and an exponential auto-correlation function for Mn (or Fe) moments. A similar model function [32] was assumed for the analysis of AgMn data. Yet within such limitations, ZF- μ SR has determined the absolute value of τ_c , for the first time, in a wide temperature region around T_g . See ref. [25] for details of ZF- μ SR in spin glasses.

III. LONGITUDINAL-FIELD μ SR (LF- μ SR) IN SPIN GLASSES

To investigate further details of dynamical aspects, we have performed the μ SR measurement in CuMn by applying finite longitudinal magnetic fields $H_L = 0 - 640$ Oe [33]. When random fields H_i are static, the applied longitudinal fields $H_L > H_i$ will "decouple" the random fields, then help keep the muon polarization and change the shape of $G_z(t)$ appreciably [23]. In contrast, for fluctuating random fields ($v/a > 1$), the depolarization rate Γ at each muon site is proportional to [34]

$$\Gamma \propto v / (v^2 + \omega_L^2), \quad \omega_L = \gamma_\mu H_L \quad (6)$$

so $G_z(t)$ will not be much changed by H_L in the case of fast fluctuations $v > \omega_L$. In this way, we can clearly distinguish the dynamical and static effects in LF- μ SR, as in the " T_1 " measurements of magnetic resonance.

Fig. 7 shows the observed spectra. In this time region $t \leq 1$ μ sec, the effect of nuclear dipolar fields is negligibly small, and completely decoupled by $H_L \approx 40$ Oe. Attached solid lines represent the longitudinal-field relaxation functions [27], calculated in a similar way as $G_z^S(t)$, with the values of ν deduced from ZF- μ SR at each temperature. At $T > T_g$, the observed data did not depend much on the external fields, indicating that muon spins are mainly depolarized by the fluctuating random fields. This confirms the value of τ_c in Fig. 6 as a good measure. In a clear contrast, the observed data depended remarkably on H_L below T_g . At $T = 10$ K, the field dependence of $G_z(t)$ was much greater than the simplest model function (upper line for $H_L = 640$ Oe, lower for $H_L = 0$ Oe), suggesting an important static effect. $G_z(t)$ at lower temperatures exhibited a clear decoupling effect and fitted well to the model functions for slow spin fluctuations in μ sec region.

In a certainly high longitudinal field, where the static effects are appreciably eliminated, the depolarization rate becomes maximum for $\nu = \omega_L$, and so we can select the dynamical contribution by "gating" with H_L as illustrated in Fig. 8. The fast depolarization observed at 10 K in $H_L = 640$ Oe suggests an important dynamical effect at $T \sim 0.9T_g$ coexisting with the static effect found in Fig. 7.

Fig. 9 shows an attempt to reproduce the observed field dependence at $T = 10$ K by model functions representing some different situations [27]. As we mentioned earlier, "single τ_c " model is obviously not adequate. Then we tried to average such functions over a wide variety of τ_c , which is denoted as "clusters" in Fig. 9, because this model covers muons in different large clusters fluctuating with different τ_c . This model improved the situation, but still is not quite adequate. Finally we considered the model where random fields at each site can be expressed by the vector sum of static component and fluctuating component. The lines in Fig. 9 represent the case with 40% of static component plus 60% of random fields fluctuating with $\nu = 3 \times 10^8$ /sec. This treatment is very close to "mean field approximation", and we expect to observe this line shape if each surrounding Mn moment can also be described by such sums of static and dynamical components. It is interesting to note that the estimated static component $m = 0.3 \sim 0.4$ corresponds to the "mean field order parameter" $q = 0.09 \sim 0.16$,

which looks reasonable at $T \sim 0.9T_g$ [6]. It should be noted, however, this model also covers another physical situation where Mn moments with different values of τ_c are distributed microscopically in the system. The agreement of this model to the data is still not complete, suggesting the real system to be somewhat more complicated.

In spin glasses known of its sensitivity to the external fields, H_L may have changed the dynamics of the system itself, which we neglected in the above arguments. This aspect can be checked, to some extent, when we revisit TF- μ SR. If the system itself is changed, we will see the same reduction of depolarization rate in TF- μ SR measured with $H_T = H_L$. If the observed change of $G_z(t)$ can mainly be ascribed to the decoupling effect, the depolarization rate of TF- μ SR will not depend much on H_T . The available data of TF- μ SR [7-9] seems to be supporting the latter case. For $H_T \leq 200$ Oe, a more careful fitting of the data will be required since H_T is no longer dominant compared to the random fields ~ 100 G. We will also note that the effect of induced polarization of Mn moments in H_L or H_T , estimated from dc susceptibility, is negligibly small compared to the observed relaxation rate, as described in ref. [9]. Such field effects on the spin dynamics of spin glasses, however, need further theoretical and experimental investigations to be fully understood.

IV. DISCUSSIONS AND CONCLUSIONS

In this way, the combination of ZF, LF, and TF- μ SR experiments has enlightened the dynamical fluctuations of spin glasses. Fig. 6 represents a general tendency of the averaged value of τ_c . A fast slowing down of τ_c is observed above T_g . $\tau_c \sim 10^{-8}$ sec, deduced at T_g , agrees well with the Mössbauer experiments [35] which found the "freezing" of spins around T_g with their time window $t \sim 10^{-8}$ sec. The slow modulation in μ sec region, found at $T \sim 0.7T_g$, explains the disappearance of the host NMR spin echo signal [36] as a dynamical effect. Even when we assume a constant static component below T_g , the fluctuating component estimated in LF- μ SR becomes slower than $\tau_c \sim 3 \times 10^{-9}$ sec below T_g . These results support a picture that spin fluctuation keeps slowing down through T_g . It is a challenging problem to investigate how such a "slowing down" is realized with a large change of magnetic entropy above T_g observed in calorimetric measurements [37].

The observed LF- μ SR data exhibits a clear contrast of the two regions, $T > T_g$ and $T < T_g$, suggesting a possible change of spin dynamics at T_g . Just below T_g , a coexistence of static and dynamical contributions is found to depolarize muon spins. Such a study of distribution of τ_c , however, becomes difficult for μ SR in the region $\tau_c \approx 10^{-6}$ sec and $\tau_c \leq 10^{-10}$ sec, where the shape of relaxation functions does not vary much with the change of τ_c . Murañi [38] has suggested the distribution of τ_c , even above T_g , based on the neutron results [39]. The superparamagnetic-cloud model [40] was very successful in explaining the remanent magnetization, observed mainly at $T \leq 0.5T_g$. The μ SR data around T_g , with much sharper change of τ_c , suggests that the direct continuation of this model to higher temperatures is not adequate. The size of the clouds, for example, may have to be changed for different temperature regions.

μ SR has also been applied to insulator spin glasses [41,42], revealing much more continuous change of τ_c . Earlier results in Pd alloys [43] suggest a possibility to extend μ SR also in such systems. Further μ SR investigations will be fruitful to clarify more detailed dynamical aspects of spin glasses.

It is our pleasure to acknowledge helpful conversations with most of the contributors listed in the references.

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FIGURE CAPTIONS

- Fig. 1. (a) Muon spin precession signal in CuMn (0.7%) observed in transverse external magnetic fields. (b) Width W of random fields estimated from depolarization rate $1/T_2$ of muon spins as $\gamma_\mu W = 1/T_2$. Both (a) and (b) from *Murrock et al.* [7].
- Fig. 2. Schematic view of experimental methods of (a) transverse-field μ SR and (b) zero- and longitudinal-field μ SR. An incoming polarized muon is identified by M counter, and stopped in a specimen (dotted piece). Decay positrons are recorded by counters E, EF and EB, and the measured time spectra reflect muon spin polarization in the way as shown in the figure.
- Fig. 3. Different muon sites in a spin glass with each different variable range of random fields on μ^+ .
- Fig. 4. Zero-field muon spin relaxation function $G_Z^{SG}(t)$ in a spin glass. Static relaxation function (eq. 4) is indicated by the broken line, and the dynamical cases are illustrated with the normalized modulation rate v/a . From *Uemura* [24].
- Fig. 5. Zero-field muon spin relaxation functions observed in AuFe (1 at.%). Solid lines represent model functions $G_Z^{SG}(t, a, v)$ with $a/\gamma_\mu = 61$ G and with the values of v given in Fig. 6(a). From *Uemura et al.* [28].
- Fig. 6. Correlation time τ_c of Fe (or Mn) moments in dilute-alloy spin glasses AuFe, CuMn and AgMn. Ac susceptibility measured in a piece of the same specimen is attached in (a), and (b) shows the comparison of τ_c in three systems plotted versus reduced inverse temperature T_g/T . *AuFe* and *CuMn* from *Uemura et al.* [28], and *AgMn* from *Brown et al.* [30].
- Fig. 7. Muon spin relaxation functions in CuMn observed in longitudinal magnetic fields $H_L = 0 - 640$ Oe [33]. Solid lines represent zero- and longitudinal-field relaxation functions of "single τ_c " model for $a/\gamma_\mu = 115$ G and for the values of τ_c in Fig. 6 estimated from ZF- μ SR at each temperature.
- Fig. 8. Muon spin relaxation function in CuMn observed in $H_L = 640$ Oe, plotted with a longer time range ($t \lesssim 5$ μ sec) [33]. $G_Z(t)$ selectively reflects the dynamical component of fluctuating random fields with $v \sim \omega_L$.
- Fig. 9. Comparison of some attempts to reproduce the observed field dependence of $G_Z(t)$ at $T = 10$ K ($T/T_g \sim 0.9$) in CuMn by different model functions corresponding to different physical situations.

Fig. 1(a)

Fig. 1(b)

Fig. 2

Fig. 3

Fig. 7