

dependence of the transfer integral  $V_{ij}$ . The spatial dependence is characterized by  $\gamma_{ij}$  in our study. As seen in Fig. 3, the value of  $T_e$  depends not only upon  $\gamma$  between the substrate and the sputtered atom ( $\gamma_a$ ) but rather strongly also upon  $\gamma$  between the substrate atoms themselves ( $\gamma_s$ ).

(4) The time interval within which the studied process of ionization occurs is found to be  $4 \times 10^{-14}$  sec for  $\gamma_a = 1.5 \text{ \AA}^{-1}$  and about  $2 \times 10^{-14}$  sec for  $\gamma_a = 3 \text{ \AA}^{-1}$ . The ionization in a real system involves energy exchange and, hence, energy drain to the colder lattice in this time interval. In view of our limited knowledge of the electronic structure in the collision cascade,<sup>8</sup> it is difficult, at present, to determine reliably the speed with which electronic excitations spread in the cascade region. If one assumes that this region resembles, in the short time interval, a highly disordered solid the excitations may spread by a hopping process and travel in the time  $\tau$  the distance  $d$  given by

$$d = (lv_e\tau)^{1/2}, \quad (5)$$

where  $l$  is the length of the hop and  $v_e$  is the electron velocity. The distance  $d$  clearly depends sensitively upon the electronic structure of the substrate, being smaller for heterogenous materials like oxides and larger for clean metals.

For the electronic bandwidth of 3 eV,  $l$  equal to the interatomic distance  $3 \text{ \AA}$ , and  $\tau = 2 \times 10^{-14}$  sec, one gets from (5) the value of  $d$  equal to  $10 \text{ \AA}$ . This length is not far from the dimension of our model and hence, the outlined conclusions are likely to be valid also in real semi-infinite systems. The LTE should thus be understood in many experimental situations as being local on the atomic scale, i.e., the parameters  $T_e$  and  $\phi$  depend sensitively through the values of  $\gamma$  on the electronic structure of the sputtered atom and its immediate atomic surrounding.

<sup>1</sup>C. A. Andersen and J. R. Hinthorne, *Anal. Chem.* **45**, 1421 (1973).

<sup>2</sup>Z. Jurela, *Int. J. Mass Spectrom. Ion Phys.* **12**, 33 (1973).

<sup>3</sup>J. M. Schroer, T. N. Rhodin, and R. C. Bradley, *Surf. Sci.* **34**, 571 (1973).

<sup>4</sup>Z. Šroubek, *Surf. Sci.* **44**, 47 (1974).

<sup>5</sup>A. Blandin, A. Nourtier, and D. W. Hone, *J. Phys. (Paris)* **37**, 369 (1976).

<sup>6</sup>J. K. Nørskov and D. I. Lundqvist, *Phys. Rev. B* **19**, 5661 (1979).

<sup>7</sup>Z. Šroubek, J. Zavadil, F. Kubec, and K. Žďánský, *Surf. Sci.* **77**, 603 (1978).

<sup>8</sup>K. Wittmack, *Surf. Sci.* **85**, 69 (1979).

## Zero-Field Spin Relaxation of $\mu^+$ as a Probe of the Spin Dynamics of AuFe and CuMn Spin-Glasses

Y. J. Uemura, T. Yamazaki, R. S. Hayano, and R. Nakai

*Department of Physics, University of Tokyo, Bunkyo-ku, Tokyo, Japan*

and

C. Y. Huang

*Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87545*

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The zero-field spin relaxation of positive muon is measured in spin-glasses AuFe and CuMn. A stochastic theory of muon spin relaxation in a random dilute spin system is formulated to deduce the correlation time of Fe (or Mn) moments. The observed correlation time increases rapidly from  $10^{-10}$  sec (at  $T \sim 1.2T_g$ ) to  $10^{-5}$  sec (at  $T \sim 0.5T_g$ ), showing a sharp slowing down of spin fluctuation around  $T_g$ .

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In this paper, we report on the first attempt to measure the dynamical spin fluctuation of spin-glasses AuFe and CuMn by the zero-field spin relaxation of positive muon ( $\mu^+$ ).<sup>1</sup> Longitudinal spin-relaxation function  $G_z(t)$  of  $\mu^+$  can be direct-

ly obtained by the time-differential measurement of the forward/backward decay positrons emitted from  $\mu^+$  stopped in a specimen. This technique<sup>2</sup> allows us to observe the muon-spin relaxation with and without external magnetic field, and

thus must be a very suitable tool to investigate spin dynamics of spin-glass systems. On the other hand, the conventional (transverse-field) muon-spin-rotation method, applied to spin-glasses,<sup>3,4</sup> cannot discriminate between static inhomogeneous field broadening and dynamic field modulation.

Muon spin is mainly relaxed by the atomic dipolar field of impurity spins rather than through the Ruderman-Kittel-Kasuya-Yasuda (RKKY) interaction.<sup>3,4</sup> For the Gaussian distribution of random local field (e.g., nuclear dipolar broadening), the static zero-field relaxation function was given theoretically by Kubo and Toyabe<sup>5</sup> as

$$G_z^{KT}(t) = \frac{1}{3} + \frac{2}{3}(1 - \Delta^2 t^2) \exp(-\frac{1}{2}\Delta^2 t^2), \quad (1)$$

where  $\Delta^2 \equiv \gamma_\mu^2 \langle H_i^2 \rangle$ ,  $H_i$  being the amplitude of one component of local field ( $\gamma_\mu = 8.51 \times 10^4$  sec/G). The dynamical modulation of local field was taken into account by using the strong collision approximation,<sup>6</sup> yielding  $G_z^{KT}(t, \Delta, \nu)$  for the Markovian modulation with the rate  $\nu$ . To obtain muon relaxation function  $G_z^{SG}(t)$  in dilute moments, we have only to integrate  $G_z^{KT}(t, \Delta, \nu)$  over the probability distribution function  $P(\Delta)$  of  $\Delta$  as

$$G_z^{SG}(t, a, \nu) = \int_0^\infty G_z^{KT}(t, \Delta, \nu) P(\Delta) d\Delta, \quad (2)$$

$$P(\Delta) = (2/\pi)^{1/2} (a/\Delta^2) \exp(-a^2/2\Delta^2).$$

We assumed this  $P(\Delta)$  so that the local field may have a Lorentzian distribution<sup>7</sup> with the half width at half maximum  $a/\gamma_\mu$ , after integration over  $P(\Delta)$ . Each spatial configuration of dilute spins

surrounding  $\mu^+$  corresponds to different values of  $\Delta$ . The numerical calculation of  $G_z^{SG}(t)$  is shown in Fig. 1.

For the static case,  $G_z^{SG}(t)$  becomes

$$G_z^{SG}(t) = \frac{1}{3} + \frac{2}{3}(1 - at) \exp(-at) \quad (3)$$

as given by Kubo.<sup>8</sup> Here the rapid reduction to  $\frac{1}{3}$  corresponds to the static inhomogeneous broadening, while one component of local field parallel to the initial muon-spin direction helps hold the asymmetry to  $\frac{1}{3}$  afterwards. This remaining  $\frac{1}{3}$  asymmetry is relaxed as  $\sim (\frac{1}{3}) \exp(\frac{2}{3}\nu t)$  by slow modulation ( $\nu/a = 0 \sim 0.1$ ) of local field. In contrast, the beginning part of  $G_z^{SG}(t)$  is not much changed as far as  $\nu/a \leq 1$ , until the narrowing effect starts (at  $\nu/a \sim 2$ ) to reduce its decay rate. The narrowing, however, takes place much more slowly with increasing  $\nu/a$  compared to the nuclear dipolar system, because of the wide distribution of local field. For 1 at.% CuMn or AuFe,  $a$  is roughly  $10^7$ /sec; thus within the time window of muon measurement (30 nsec to  $10 \mu$ sec) we can observe very wide range of  $\nu$  ( $10^5 - 10^{11}$ /sec). Assuming a random (spatially uncorrelated) fluctuation of impurity spins, the self-correlation time  $\tau_c$  of Fe (or Mn) moment is given by  $1/\nu$ . Detailed stochastic theory of  $\mu^+$  in spin-glass will be reported elsewhere.<sup>9</sup>

At TRIUMF, we observed  $G_z(t)$  of  $\mu^+$  under zero field ( $< 1$  Oe) in AuFe (1.0 at.%,  $T_g = 9.1$  K deduced from the observed cusp of ac susceptibility; 1.4 at.%,  $T_g = 12.8$  K: made by rapid cool

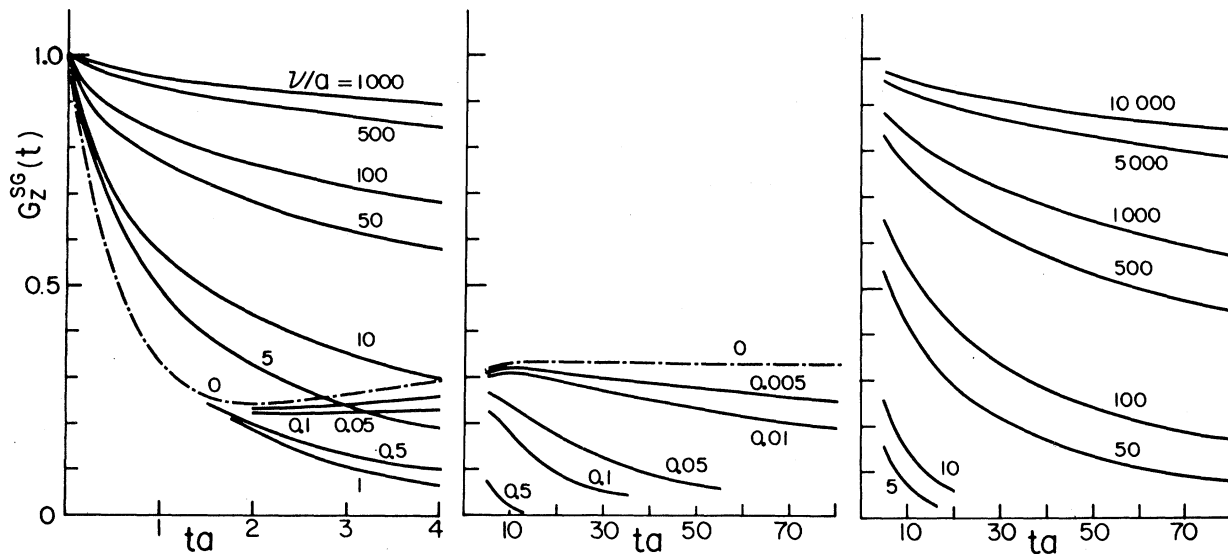


FIG. 1. The zero-field spin-relaxation function  $G_z^{SG}(t)$  for  $\mu^+$  in dilute random moments displayed with the normalized modulation rate  $\nu/a$ . Broken line shows the static case.

down after arc melting) and  $\text{CuMn}$  (1.1 at.%,  $T_g = 10.8$  K; quenched from 930 C). The samples were checked by chemical analysis, ac susceptibility (very sharp cusp), and x-ray microanalysis (homogeneous impurity concentration among many different spots of  $10 \mu\text{m}$  diameter). The temperature was stabilized within  $\pm 0.1$  K throughout the measurement.

As shown in Fig. 2(a), the observed  $G_z(t)$  in  $\text{AuFe}$  (1.0 at.%) is well fitted by  $G_z^{SG}(t)$  of Eq. (2). The averaged static local field  $a = 5.2 \pm 0.3 \mu\text{sec}^{-1}$  ( $a/\gamma_\mu = 61$  Oe) deduced from the spectra of 4.5 and 6 K corresponds well to the calculated dipolar field from Fe moments  $a = 5.1 \mu\text{sec}^{-1}$ , suggesting a random selection of interstitial sites by muons. The characteristic  $\frac{1}{3}$  component observed at 4.5 K is relaxed by a dynamical modulation ( $1/\nu \sim 1 \mu\text{sec}$ ) at 6 K. The narrowing effect, starting just below  $T_g$ , rapidly reduces the relaxation rate with increasing temperature above  $T_g$ . This rapid change of  $G_z(t)$  is observed at  $T \sim 9.5$  K in 1.0%  $\text{AuFe}$ , while at  $T \sim 13$  K in 1.4%  $\text{AuFe}$ . We should then ascribe it to the spin fluctuation of Fe moment rather than to the muon diffusion.

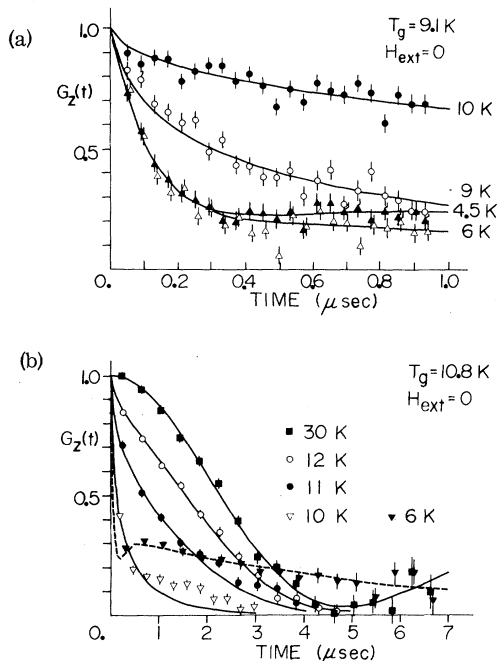


FIG. 2. The zero-field relaxation function of  $\mu^+$  observed (a) in  $\text{AuFe}$  (1.0 at.%) and (b) in  $\text{CuMn}$  (1.1 at.%). The solid line corresponds to  $G_z^{SG}(t)$  for  $a = 5.2/\mu\text{sec}$  in  $\text{AuFe}$  and  $G_z^{SG}(t)G_z^{KT}(t)$  for  $a = 10.2/\mu\text{sec}$ ,  $\Delta = 0.36/\mu\text{sec}$  in  $\text{CuMn}$ , and the broken line of  $\text{CuMn}$  shows  $G_z^{SG}(t)$ .

$\mu^+$  is known to be frozen at an interstitial site of Cu (Ref. 10) in the temperature region of our interest, and similar situation can be expected in another fcc noble metal Au indirectly<sup>11</sup> though the negligibly small nuclear dipolar moment of Au is not helpful for the direct measurement of muon diffusion. Thus we assumed no effect of muon diffusion and obtained  $\tau_c$  from  $1/\nu$ . The effect of diffusion, if ever present, should be limited to the region of very slow modulation ( $\nu \leq 0.3 \mu\text{sec}^{-1}$ ). This aspect can actually be confirmed in the case of  $\text{CuMn}$  (1.1 at.%) shown in Fig. 2(b) at  $T = 30$  K, where  $G_z(t)$  exhibits a typical Kubo-Toyabe function  $G_z^{KT}(t)$  of Eq. (1) for static nuclear dipolar field with  $\Delta \sim 0.36 \mu\text{sec}^{-1}$  expected in pure Cu. The effect of local field  $H_{\text{Mn}}$  from Mn spins is eliminated by the fast spin fluctuation ( $\tau_c \leq 10^{-11}$  sec) at this temperature ( $T \sim 3T_g$ ).

In  $\text{CuMn}$ , as well as in  $\text{AuFe}$ , the rapid change of  $G_z(t)$  was observed when we cooled down through  $T_g$ . This is due to  $H_{\text{Mn}}$ , reflecting the change of  $G_z^{SG}(t)$  with increasing  $\tau_c$ , and  $G_z(t)$  is fitted well by  $G_z^{SG}(t)[G_z^{KT}(t)]$  for Cu [solid lines of Fig. 2(b)] for  $\nu \geq 10^7/\text{sec}$ . At lower temperatures, the observed spectra exhibited  $G_z^{SG}(t)$ , with the slow relaxation of  $\frac{1}{3}$  asymmetry, since nearly static field of  $H_{\text{Mn}}$  decoupled the nuclear dipolar field  $H_{\text{Cu}}$  (cf.  $H_{\text{Mn}} \gg H_{\text{Cu}}$ ) in this case. At any temperature, the possible contribution of  $H_{\text{Cu}}$  is almost negligible [ $G_z^{KT}(t) \sim 1$ ] in the beginning part ( $t \leq 0.5 \mu\text{sec}$ ) of  $G_z(t)$ , thus we can obtain  $\tau_c$  and  $a = 10.2 \pm 0.8 \mu\text{sec}^{-1}$  (calculated atomic dipolar field  $a = 11.4 \mu\text{sec}^{-1}$ ). The role of  $H_{\text{Cu}}$  was checked by applying a small longitudinal field of  $\sim 30$  Oe, which is enough to decouple  $H_{\text{Cu}}$  selectively.

In this way,  $\tau_c$  is directly deduced from  $G_z(t)$  over wide temperature region as plotted in Figs. 3(a) and 3(b). We can draw the following conclusions:

(i) *Rapid change of  $\tau_c$  near  $T_g$ .*— $\tau_c$  changed drastically, about two orders of magnitude, within the region  $0.9T_g < T < 1.1T_g$ . The sharpness of this change roughly corresponds to  $n \sim 2$  for  $\tau_c \propto [T/(T - T_g)]^n$  when we compare the paramagnetic region ( $T > 1.03T_g$ ) to the power-law behavior of critical slowing down [see Fig. 3(b)]. When we assume the Arrhenius law  $\tau_c = \tau_0 \exp(E_a/kT)$ , the slope of  $\tau_c$  around  $T_g$  gives  $E_a \geq 20kT_g$ .

(ii) *Scaling with  $T_g$ .*— $\tau_c$  for different system and impurity concentration can roughly be scaled by  $T_g$  of each sample.  $\tau_c$  at  $T = T_g$  is  $\sim 10^{-8}$  sec, which must be a characteristic number for spin-glasses.

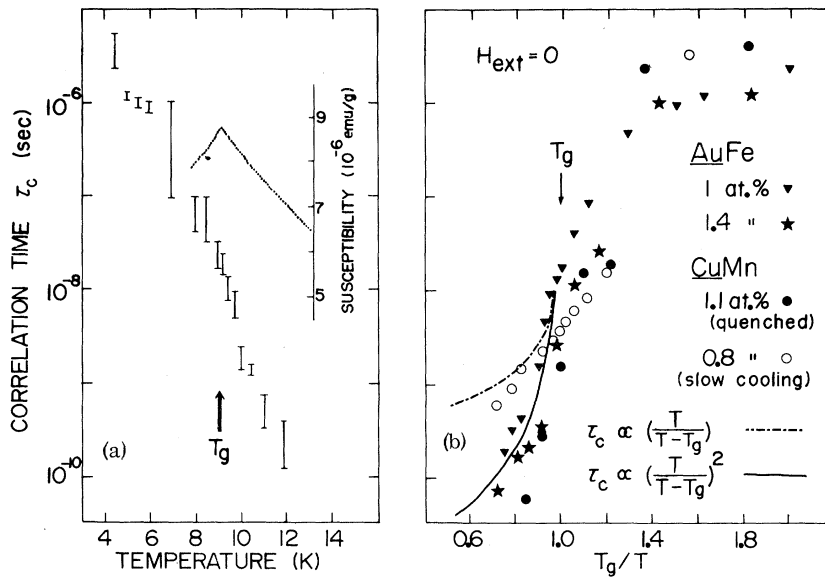


FIG. 3. (a) Correlation time  $\tau_c$  of Fe moment in AuFe (1.0 at.%,  $T_g = 9.1$  K). The error bar includes the systematic error and the scattering of data among a few measurements. (b)  $\tau_c$  plotted vs reduced temperature  $T_g/T$ . Statistical error is roughly the size of each point.

(iii) *Slow correlation in the freezing phase.*

—The impurity spins are fluctuating slowly ( $\tau_c \sim \mu\text{sec}$ ) in the freezing phase ( $0.5T_g < T < 0.8T_g$ ).

(iv) *Dependence on heat treatment of samples.*

—We have also performed the same experiment in CuMn (0.5, 0.8, 1.8, and 2.4 at.%) made by slow cooling down after melting. Compared to these samples, the rapidly-cooled samples (AuFe and quenched CuMn) showed much sharper change of  $\tau_c$  around  $T_g$  [see Fig. 3(b)], better fit of  $G_z(t)$  to  $G_z^{SG}(t)$ , and sharper cusp of ac susceptibility. These features are explained by the inhomogeneous impurity concentration (varying from 70% to 130% of averaged concentration in the region of  $\sim 100 \mu\text{m}$ ) found in slowly-cooled CuMn by means of an x-ray microanalyzer.

In AuFe and quenched CuMn,  $G_z(t)$  slightly deviates from  $G_z^{SG}(t)$  in the region  $0.8T_g < T < 0.9T_g$  [e.g., Fig. 2(b), at 10 K], which might be attributed to the possible effect of the Edwards-Anderson order parameter<sup>9,12</sup> and/or to the spatial distribution of  $\tau_c$ . These aspects will be clarified in future with advanced techniques (much higher statistics, lower backgrounds, longer time range). In this report, we confined ourselves to the simplest case of the exponentially decaying autocorrelation function of impurity spins.

The present work has provided new information in the dynamical aspects and revealed the drastic change of spin dynamics around  $T_g$ . The general features are consistent with the neutron

experiment,<sup>13</sup> and can be compared with the EPR experiment by Salamon.<sup>14</sup> The previous muon-spin-rotation results<sup>3</sup> can be understood in our viewpoint. Thus we have demonstrated a unique capability of zero-field muon spin rotation probing spin-glasses.

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<sup>1</sup>A preliminary stage of this work was reported in T. Yamazaki, Y. J. Uemura, M. Takigawa, and C. Y. Huang, *J. Magn. Magn. Mater.* **15-18**, 407 (1980).

<sup>2</sup>R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, K. Nagamine, T. Yamazaki, and H. Yasuoka, *Phys. Rev. Lett.* **41**, 421 (1978); R. S. Hayano *et al.*, *Phys. Rev. Lett.* **41**, 1743 (1978).

<sup>3</sup>D. E. Murnick, A. T. Fiory, and W. J. Kossler, *Phys. Rev. Lett.* **36**, 100 (1976).

<sup>4</sup>K. Nagamine, N. Nishida, S. Nagamiya, O. Hashimoto, and T. Yamazaki, *Phys. Rev. Lett.* **38**, 99 (1977).

<sup>5</sup>R. Kubo and T. Toyabe, in *Magnetic Resonance and Relaxation*, edited by R. Blinc (North-Holland, Amsterdam, 1967), p. 810.

<sup>6</sup>Y. J. Uemura, R. S. Hayano, J. Imazato, N. Nishida, and T. Yamazaki, *Solid State Commun.* **31**, 731 (1979); R. S. Hayano, Y. J. Uemura, J. Imazato, N. Nishida, T. Yamazaki, and R. Kubo, *Phys. Rev. B* **20**, 850 (1979).

<sup>7</sup>R. E. Walstedt and L. R. Walker, *Phys. Rev. B* **9**, 4857 (1974).

<sup>8</sup>R. Kubo, private communication.

<sup>9</sup>Y. J. Uemura, to be published.

<sup>10</sup>O. Hartmann, *Hyperfine Interact.* **6**, 203 (1979).

<sup>11</sup>R. H. Heffner, J. A. Brown, C. Y. Huang, S. E. Kohn, M. Leon, C. E. Orsen, and M. E. Schillaci, *Bull. Am. Phys. Soc.* **25**, 345 (1980).

<sup>12</sup>S. F. Edwards and P. W. Anderson, *J. Phys. F* **5**, 965 (1975), and **6**, 1927 (1976).

<sup>13</sup>F. Mezei and A. P. Murani, to be published.

<sup>14</sup>M. B. Salamon, *J. Magn. Magn. Mater.* **15**, 147 (1980).

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## ERRATUM

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REGGE SLOPE AND THE  $\Lambda$  PARAMETER IN QUANTUM CHROMODYNAMICS: AN EMPIRICAL APPROACH VIA QUARKONIA. W. Buchmüller, G. Grunberg, and S.-H. H. Tye [*Phys. Rev. Lett.* **45**, 103 (1980)].

The factor  $-(1/\rho_2)$  on the right-hand side of Eq. (4) should be replaced by  $-(1/\rho^2)$ .

Figure 2 has the wrong scale in  $r$ . The corrected figure is given below.

