Local Observation of the Impurity-Impurity Interaction in Paramagnetic *Au*Fe Alloys

A. Metz, J. Kapoor, and D. Riegel
Hahn-Meitner-Institut G.m.b.H., 14109 Berlin, Germany

W. D. Brewer
Fachbereich Physik, Freie Universität Berlin, 14195 Berlin, Germany
(Received 3 June 1994)

From the nuclear spin rotation of implanted ^{54}Fe nuclei, we have determined the local susceptibility, the spin dynamics, and the width of the distribution of local magnetic fields of $Au_{1-x}Fe_x$ ($x = 10^{-4}$–5×10^{-2} at.%) spin glasses far above the freezing temperature. The width of this distribution was found to be proportional to the freezing temperature. The mean field freezing temperature, as obtained from the field distribution, was found to be a factor of 5 higher than the experimentally observed freezing temperature.

PACS numbers: 75.30.Et, 75.50.Lk, 76.80.+y

The behavior of dilute metallic spin glasses is governed by the magnetic impurity-impurity interaction, which is generally assumed to be of the Ruderman-Kittel-Kasuya-Yosida (RKKY) type. Because of the oscillating character of this interaction, the average local field is close to zero, and the width of the local field distribution has emerged as a natural measure of the spin glass freezing temperature T_f. This idea has been widely accepted since the early spin glass model of Edwards and Anderson (EA) [1], but recent numerical simulations [2] have indicated that the actual situation might be more complicated. In view of these theoretical uncertainties there is a clear need for reliable experimental data on J_0, the strength of the impurity-impurity interaction, especially since the available experimental information was obtained from bulk measurements, and depends upon additional assumptions [3].

In this Letter, we report on investigations of the local magnetic response of Fe moments in *Au*Fe spin glasses far above T_f, by means of the time-differential perturbed γ-ray distribution (TDPAD) method. We have chosen the *Au*Fe system because it is a prototype of a metallic spin glass, and because ^{54}Fe is an excellent probe for investigations with the TDPAD method. We have studied the local susceptibility, the spin dynamics, and—as the central point of this paper—the distribution of local magnetic fields. From the latter quantity, we can determine the interaction strength J_0.

The TDPAD method probes the static and the dynamic response at the nuclear site of the recoil implanted Fe atoms via the spin rotation and its damping of an excited and aligned nuclear isomer (here ^{54}Fe) in an external magnetic field. The method permits microscopic measurements at the site of the magnetic impurity, and allows investigations of the magnetic response over a wide range of temperatures, both in the concentrated as well as in the extremely dilute regime. Measurement at the impurity site is an important feature, since the interaction between the impurities cannot be predicted in a reliable way from the interaction between a single impurity and the conduction electrons [4]. Because of these features, the application of TDPAD to spin glasses yields essentially new results that cannot be obtained with other nuclear methods such as Mössbauer effect (ME), impurity nuclear magnetic resonance (NMR), or muon spin rotation (μSR).

Most ME experiments, using the ^{54}Fe probe, have been directed towards estimating the freezing temperatures from the onset of a peak splitting in the spectrum, e.g., [5], and towards an understanding of static and dynamic interactions in *Au*Fe alloys around and below T_f [6].

Impurity NMR studies have been carried out by a number of authors. Such studies, however, require a sufficiently long relaxation time and are therefore restricted to very low temperatures in the frozen state [7], to very high temperatures [8], or to systems with high Kondo temperatures like *Au*V [9]. This is the main reason that impurity NMR studies in the paramagnetic state of a spin glass, which would allow a direct comparison of the interaction strength J_0 to T_f, have not been reported thus far.

Our TDPAD experiments were performed at the VICKSI accelerator of the Hahn-Meitner-Institut in Berlin, using a pulsed ^{12}C beam of 42 MeV energy to produce ^{54}Fe ions by the heavy ion reaction ^{45}Sc(^{12}C, $p2n$)^{54}Fe, followed by recoil implantation into the *Au*Fe alloys. This technique produces extremely dilute ^{54}Fe nuclear probes (concentration $\ll$ 1 ppm) with nuclear spin 10^+ and lifetime 360 ns, which allow the detection of the magnetic response via the observation of spin rotation spectra $R(t)$ in an external magnetic field B_{ext}. Details of methodical aspects can be found in Ref. [10].

In Figs. 1 and 2 we show some typical $R(t)$ spectra for different temperatures and concentrations. The spectra were fitted to the function $R(t) = A_{22}e^{-\Gamma t}\cos 2(\omega_L t - \phi)$. From the temperature dependence of the Larmor frequencies,

$$\omega_L(T) = \hbar^{-1} g_N \mu_N B_{\text{ext}}[1 + B_{\text{hf}}\chi_{\text{loc}}(T)/g\mu_B S], \quad (1)$$

0031-9007/94/73(23)/3160(4)$06.00

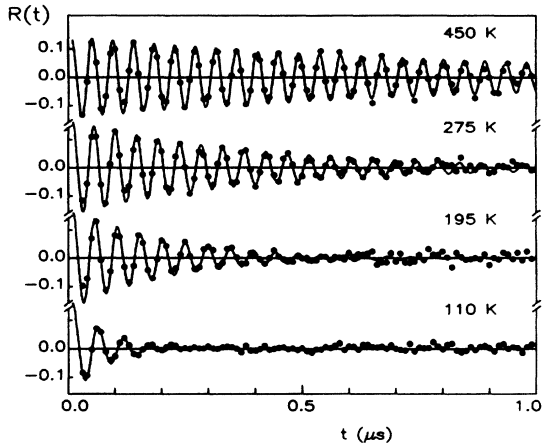

FIG. 1. Temperature dependence of the spin rotation spectra of ^{54}Fe in an *Au*Fe(0.8%) alloy recorded at $B_{ext} = 2$ T.

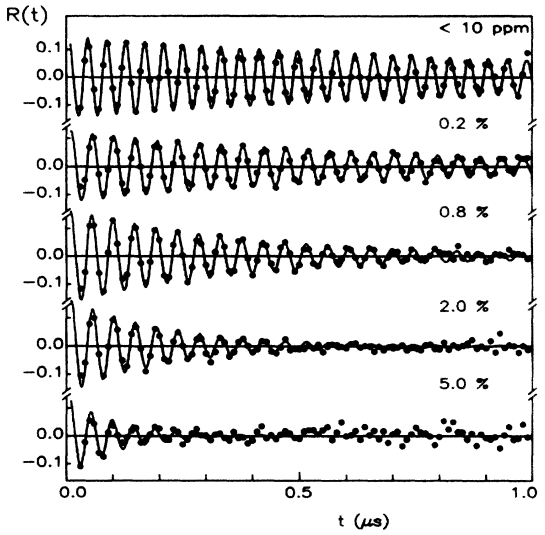

FIG. 2. Spin rotation spectra of Fe in various *Au*Fe alloys, with concentrations ranging from below 10 ppm to 5 at. %. All spectra were recorded at room temperature and $B_{ext} = 2$ T.

the local susceptibilities $\chi_{loc}(T)$ can be extracted. For concentrations up to 5 at. % Fe, the local susceptibilities can be fitted by a Curie law: $\chi_{loc}(T) = (g\mu_B)^2 S(S+1)/3k_BT$. Using $g_N = 0.728$ for the nuclear g factor of the ^{54}Fe isomer and an effective spin $S = 1.4$ from bulk susceptibility measurements [11], we obtain $B_{hf} = -22$ T for the hyperfine field at $T = 0$ K, in good agreement with the results given in Ref. [12].

The observation of only one component with well defined frequency and constant amplitude A_{22} (independent of temperature and concentration) implies that all Fe atoms contributing to $R(t)$ were located on substitutional sites. By comparison with the A_{22} observed for ^{54}Fe in liquid metals, one can deduce that about 80% of the implanted ^{54}Fe contribute to $R(t)$ in *Au*Fe alloys. We note that the implanted ^{54}Fe probes are statistically distributed and—because of the rapid decay—are not subject to possible chemical short range order effects.

Most sensitive to the interaction between the magnetic Fe ions is the damping of the $R(t)$ spectra. It turns out to be unexpectedly large, exhibiting a strong dependence on the temperature (see Fig. 1, for example), and increases with concentration (Fig. 2). Figure 3 shows a log-log plot of the damping constant Γ as a function of the temperature. The Γ vs T curves change from a T^{-1} dependence for Fe in pure Au to a T^{-2} dependence for the *Au*Fe(5 at. %) alloy. Furthermore, we have measured Γ as a function of the external magnetic field and found (except for Fe in pure Au) a roughly linear dependence of Γ on B_{ext}.

The observed damping can be interpreted as being caused by two independent physical phenomena: $\Gamma = \Delta 2\omega_L + \tau_N^{-1}$. The nuclear magnetic spin relaxation rate τ_N^{-1} is due to dynamic fluctuations of the atomic Fe spin, whereas $\Delta 2\omega_L$ scales with the field-induced static inhomogeneous broadening arising from the interactions between the Fe impurities.

Dynamic spin fluctuations are most clearly seen for Fe in pure Au. Here Γ can be identified with the field independent rate τ_N^{-1}. The observed τ_N^{-1} nicely follows Korringa-type behavior, $\tau_N^{-1} \propto \tau_J \propto T^{-1}$ (Fig. 3), where the atomic Fe spin relaxation time τ_J arises from the exchange interaction between the Fe moment and conduction electrons. Employing the relation $\tau_N^{-1} = 2\tau_J (g_N \mu_N B_{hf}/\hbar)^2 (S+1)/S$, one can deduce τ_J of Fe in pure Au at 300 K to be 1.9×10^{-13} s. This value is of the same order of magnitude as an estimate $\tau_J = 4.2 \times$

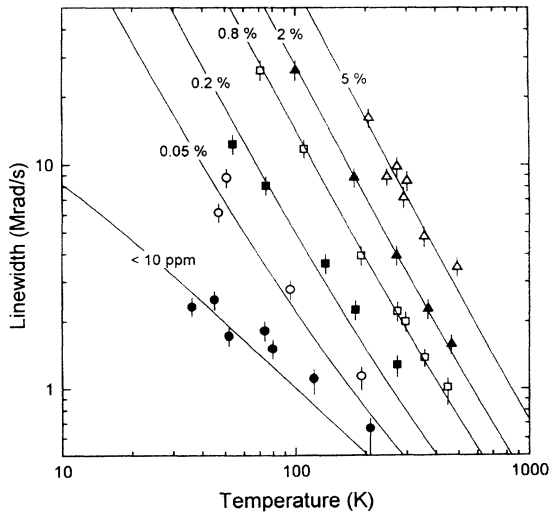

FIG. 3. Log-log plot of the damping constant Γ of the spin rotation spectra, recorded at $B_{ext} = 2$ T, vs temperature for various concentrations.

10^{-13} s from resistivity measurements [13], and compatible with Mössbauer linewidth results [6]. Now, applying the relation for the Korringa relaxation of a local moment, $\tau_J = \hbar/4\pi\,(2l+1)[J_2 N(E_F)]^2 k_B T$, and taking for the density of states at the Fermi surface the value for pure Au, $N(E_F) = 0.136$ states/spin eV, we can determine the exchange parameter to be $J_2 = 0.36(2)$ eV. Here, we have assumed that the $l = 2$ orbital component dominates the s-d interaction, and have included a factor $(2l+1)$ to account for the fact that the orbital substates contribute independently to the relaxation process [14]. The value for J_2 thus obtained can be compared with the coupling parameter derived from the Kondo effect using the relation $k_B T_K = E_F \exp[1/2J_2 N(E_F)]$. With $E_F = 5.51$ eV and $T_K = 0.18$ K [15], we calculate $J_2 = 0.29$ eV, in reasonable agreement with the value found from the spin dynamics.

Estimates for the spin dynamics were also obtained for the more concentrated alloys by measuring the field dependence of the damping, and subsequent extrapolation to zero field, in order to separate the field-independent dynamic rate from the inhomogeneous line broadening. Using this procedure, we found no significant indications for a change of τ_J in the more concentrated alloys, which implies that there is no evidence for dynamic correlations between the Fe local moments at temperatures above $8T_f$, where our data were acquired. Such dynamic correlations would lead to a decrease of the atomic spin rate, and have been observed in *Cu*Mn by means of neutron scattering [16] and in *Au*Fe by means of μSR [17] at temperatures up to 4 times T_f.

Even at the moderate impurity concentration of 0.2 at. % Fe, the damping is dominated by the inhomogeneous broadening, which is clearly measurable over a broad range of concentrations in the paramagnetic regime of the *Au*Fe spin glasses well above T_f. The static broadening $\Delta\omega_L$ reflects the distribution of local magnetic fields and is the result of the interaction between the magnetic impurities, which is defined by the Hamiltonian

$$H = -J_0 \sum_{i<j} F(R_{ij})\,\mathbf{S}_i \cdot \mathbf{S}_j\,, \tag{2}$$

where J_0 is the coupling constant and $F(R)$ is the oscillatory range function. When averaging over many impurity configurations, as we do in our TDPAD experiments, we measure the effect of a distribution of local fields. The resulting distribution of Larmor frequencies has a half-width $\Delta\omega_L$, which is given by (see Ref. [9])

$$\Delta\omega_L = \hbar^{-1} g_N \mu_N B_{\rm hf} J_0 \langle F\rangle (B_{\rm ext}/g\mu_B S)[\chi_{\rm loc}(T)/g\mu_B]^2, \tag{3}$$

where $\langle F\rangle$ is half the width at half the maximum of the distribution of local amplitudes generated by the range function $F(R)$ [9]. This expression is based on a mean field approximation, which is appropriate in the temperature range investigated because of the absence of correlations between the spins, as shown by our results on the spin dynamics. The square of $\chi_{\rm loc}$ arises from the effect of impurity-impurity interactions, and gives rise to the observed T^{-2} dependence. Walstedt and Walker [18] have studied this distribution in detail for the case of RKKY interactions, where $F(R)$ can be approximated by $F(R) = \cos(2k_F R)/(2k_F R)^3$. Below 0.1 at.%, the distribution has a Lorentzian line shape and is centered around zero. The data in Fig. 3 can be well fitted by the function $\Gamma(T) = \tau_N^{-1}(T) + \Delta 2\omega_L(T)$, in which $\tau_N^{-1}(T)$ is assumed to have the value obtained for extremely dilute Fe in Au, as described above. From these fits one can directly obtain the parameter $J_0\langle F\rangle$, inserting the known values for $B_{\rm hf}$ and S into Eq. (3).

The parameter $J_0\langle F\rangle$ is an important quantity, since it determines the width of the distribution of local magnetic fields, and defines $T_{\rm mf} \equiv J_0\langle F\rangle/k_B$, the mean field estimate for the spin glass freezing temperature. In Fig. 4, we show a log-log plot of $T_{\rm mf}$ and T_f vs concentration, where $T_{\rm mf}$ is obtained from our results, and the T_f curve represents an extensive set of experimental data [19]. By inspection of Fig. 4 it can be seen that T_f and $T_{\rm mf}$ have a very similar concentration dependence. The ratio $T_{\rm mf}/T_f$ lies between 4.6 and 7.0, the average value being 5.0 over the investigated concentration range. This shows that $T_{\rm mf}$ and T_f are proportional to each other, but the spin glass freezing sets in only at a temperature well below the mean field estimate $T_{\rm mf}$.

In order to be able to make a comparison with other results, we have to extract the value of J_0 and make an assumption concerning $\langle F\rangle$. Following Ref. [18], we obtain for a monovalent fcc metal $\langle F\rangle = x/18\pi$, where x is the concentration of magnetic impurities. Using this relation, which is valid in the dilute limit, and inserting the experimental value for $J_0\langle F\rangle$ at 0.05 at.%, we obtain for J_0 a value of 30 eV. Such a strong coupling is not consistent with simple RKKY coupling, where J_0 is given by $J_0 = 9\pi J_2^2 (2l+1)^2/E_F$. This expression was used by Larsen [19], who found accidental agreement with the prediction $T_f \cong T_{\rm mf}$ from the EA model. A stronger interaction is predicted by the double resonance mechanism [4], where J_0 is given by $J_0 =$

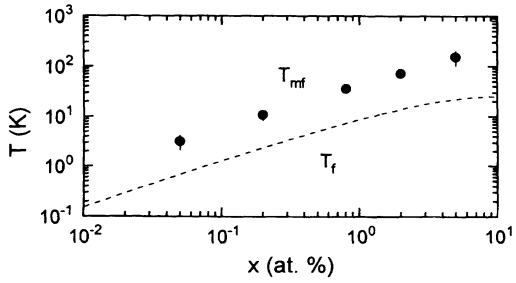

FIG. 4. Log-log plot of the mean field estimate $T_{\rm mf}$, as obtained from the distribution of local fields, and of the experimental freezing temperature T_f vs Fe concentration x (see text).

$100E_F(\sin^2\eta_+ + \sin^2\eta_-)/\pi S^2$, and $\eta_{+(-)}$ is the d-wave phase shift for the spin up (down) resonance. For AuFe, with $S \approx 1.5$, $\eta_+ \approx \pi$, and $\eta_- \approx 2\pi/5$, this yields $J_0 \approx 80$ eV. A direct comparison with first principles calculations [20] is not possible since these are limited to near neighbors, the present numerical accuracy being insufficient to predict the asymptotic behavior.

We can also compare our results with a recent numerical simulation of freezing behavior in RKKY-type spin glasses, carried out by Matsubara and co-workers [2]. These authors calculated T_f of a 5 at.% spin glass alloy with $S = 1$ and obtained $T_f \cong 7 \times 10^{-5} J_0/k_B$. Using our value for J_0 in this expression and applying a spin correction factor S^2, we find $T_f = 50$ K, which is a factor of 2 higher than the experimental value for a 5 at.% AuFe spin glass [19], but well below $T_{\mathrm{mf}} = 150$ K.

We suggest that the difference between T_{mf} and T_f is caused by the fact that, upon approaching the freezing temperature, the impurity spins form increasingly extended clusters, where frustration is partly relieved [2]. Thus the width of the distribution of local magnetic fields is reduced to far below the mean field value by a decrease of both the magnitude of the local fields themselves and of the number of possible spin configurations. In order to obtain a better understanding of the relation between T_f and T_{mf}, further experimental and numerical studies of the local field distribution and its concentration dependence, both near and far above T_f, would be desirable.

To summarize, we have explored the applicability of the TDPAD method to the study of spin glass problems, using the typical alloy AuFe, and have learned that this technique is very sensitive to interactions between local moments, far above the spin glass temperature T_f. Thus it enabled us to determine, for the first time, the strength of this interaction *at the impurity site itself.* Our results show that current theoretical models do not provide a quantitative estimate of this interaction strength. In addition, we have been able to show that the widely accepted mean field estimate of the spin glass freezing temperature is much higher than the actual freezing temperature in AuFe spin glasses. A study of the field dependence of the spin rotation damping revealed no indications of dynamic spin correlations above $8T_f$.

Finally, we like to point out that the method can be applied generally to study magnetic impurity-impurity interactions and impurity spin dynamics in other magnetic alloys, e.g., to compare accurately Fe-Fe interactions with Fe-Mn or Fe-Cr interactions in noble metal based spin glass systems. Using nuclear probes in rare earth isotopes (see Ref. [10]), it also seems possible to extend such studies to rare earth spin glasses in the paramagnetic phase.

The authors wish to thank K. D. Gross, P. H. Dederichs, Yi Li, F. Mezei, C. Pappas, and R. Zeller for support of this work and for valuable discussions. We also thank Peter Szymkowiak for his expert help with sample preparation.

[1] S. F. Edwards and P. W. Anderson, J. Phys. F **5**, 965 (1975).
[2] F. Matsubara and M. Iguchi, Phys. Rev. Lett. **68**, 3781 (1992); M. Iguchi *et al.*, Phys. Rev. B **47**, 2648 (1993).
[3] Valuable experimental reviews can be found in K. H. Fisher, in *Numerical Data and Functional Relationships in Science and Technology,* edited by K.-H. Hellwege, Landolt-Börnstein, New Series, Group III, Vol. 15a (Springer-Verlag, Berlin, 1982), p. 289; and J. A. Mydosh, *Spin Glasses, an Experimental Introduction* (Taylor & Francis, London, 1993).
[4] B. Caroli, J. Phys. Chem. Solids **28**, 1427 (1967); Q. Zhang and P. M. Levy, Phys. Rev. B **34**, 1884 (1986).
[5] U. Gonser *et al.*, J. Appl. Phys. **36**, 2124 (1965); C. E. Violet and R. J. Borg, Phys. Rev. **149**, 540 (1966).
[6] C. Meyer, F. Hartmann-Boutron, Y. Gros, and I. A. Campbell, J. Magn. Magn. Mater. **46**, 25 (1985).
[7] H. Alloul and P. Mendels, Phys. Rev. Lett. **54**, 1313 (1985).
[8] R. E. Walstedt and W. W. Warren, Phys. Rev. Lett. **31**, 365 (1973).
[9] A. Narath, A. C. Gossard, and J. H. Wernick, Phys. Rev. Lett. **20**, 795 (1968); A. Narath and A. C. Gossard, Phys. Rev. **183**, 391 (1969).
[10] D. Riegel and K. D. Gross, in *Nuclear Physics Applications on Materials Science,* edited by E. Recknagel and J. C. Soares, NATO Advanced Study Institutes Ser. E, Vol. 144 (Kluwer Academic, Norwell, MA, 1988), p. 327.
[11] C. M. Hurd, J. Phys. Chem. Solids **28**, 1345 (1967); A. F. J. Morgownik and J. A. Mydosh, Solid State Commun. **47**, 321 (1983); C. N. Guy, J. Phys. F **7**, 1505 (1977).
[12] P. Steiner *et al.*, Solid State Commun. **13**, 1507 (1973).
[13] I. A. Campbell, P. J. Ford, and A. Hamzic, Phys. Rev. B **26**, 5195 (1982).
[14] B. Caroli, J. Phys. F **5**, 1399 (1975).
[15] J. G. Pérez-Ramírez and P. Steiner, J. Phys. F **7**, 1573 (1977).
[16] A. P. Murani, Phys. Rev. Lett. **41**, 1406 (1978).
[17] I. A. Campbell *et al.*, Phys. Rev. Lett. **72**, 1291 (1994).
[18] R. E. Walstedt and L. R. Walker, Phys. Rev. B **9**, 4857 (1974).
[19] U. Larsen, Phys. Rev. B **18**, 5014 (1978).
[20] A. Oswald, R. Zeller, P. J. Braspenning, and P. H. Dederichs, J. Phys. F **15**, 193 (1985).