Mean-field theory of nuclear-spin relaxation in the spin-glass phase

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Based on Edwards-Anderson mean-field theory a calculation of nuclear-spin relaxation in the spin-glass phase is presented. It is shown that most of the features of the available data can be understood within this mean-field theory. In particular, a nonexponential form of nuclear magnetization recovery is obtained, in agreement with experiment. Further, it is shown that the impurity spin-correlation time τ_e changes rapidly from $\hbar\tau_e^{-1} \gg k_B T_g$ to $\hbar\tau_e^{-1} \gg k_B T_g$ around T_g (T_g is the spin-glass transition temperature), which agrees, qualitatively, with the result obtained from recent μ^+-meson depolarization experiments.

Nuclear-spin relaxation of the host metal in dilute magnetic alloys provides useful information about the impurity-spin dynamics. In particular, it can be useful in the study of spin-glasses,[1,2] where impurity-spin dynamics assumes a more important role. In a recent work[3] we presented a theory of high-temperature nuclear-spin relaxation in these alloys. The purpose of this paper is to present calculations for low temperatures, i.e., for temperatures T less than the spin-freezing temperature T_g. The theory is restricted to the case of zero applied magnetic field.

The basic assumptions of the present calculations are the same as those of Ref. 3. However, instead of using a random exchange model and a Langevin equation, we use a simple time-dependent form of Edwards-Anderson (EA) mean-field theory[4,5] to describe the impurity-impurity interaction part of the problem. We assume an impurity spin $\vec{S}$ of one-half and negligible applied magnetic field. The effect of the finite field and $S \neq \frac{1}{2}$ will be discussed qualitatively. At the end of the paper we will also discuss some of the recent developments in the theory of spin-glasses and their relevance to the calculation described here.

The data that will be discussed most extensively here is the data taken on dilute Cu-Mn spin-glasses by Bloyet *et al.*,[6] as it appears to be the most complete. For quick reference their data for the two lowest-field strengths have been reproduced in Fig. 1. Unfortunately, in these experiments substantial fields are present. Very recently, Chen and Slichter[7] have reported some zero-field measurements on a different spin-glass system. As we will discuss later, the zero-field limit of the results of Bloyet *et al.*[6] does not appear inconsistent with the findings of Chen and Slichter.[7] We will show that most features of the available data[6] for $T < T_g$ can be understood within the mean-field theory. In particular, a nonexponential form of nuclear magnetization recovery, which is practically the same as that seen by Bloyet *et al.*,[6] is obtained. Further, the temperature dependence of the impurity-spin self-correlation time τ_e obtained by comparing the theory with the experiment[6] is very close to the temperature dependence of τ_e obtained by Uemura *et al.*[8] from μ^+-meson depolarization experiments.

Turning to our calculations of nuclear-spin relaxation, we assume[9] that the dominant impurity con-

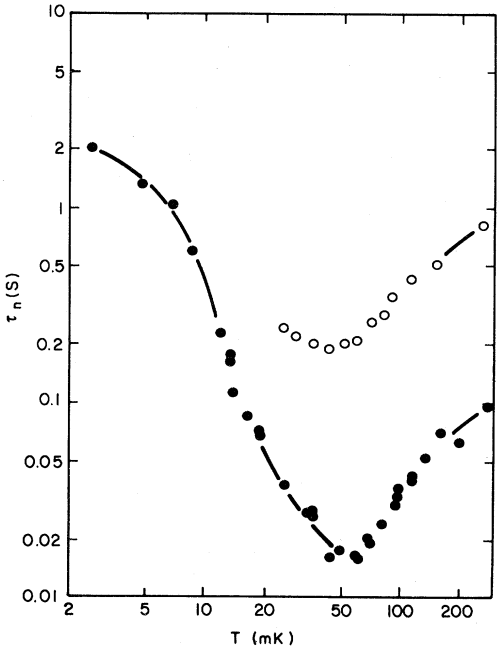

FIG. 1. Experimental results of Ref. 6. τ_n is a mean-nuclear-spin relaxation time. Solid circles represent the data at $H \sim 275$ G (310 kHz), while the open circles stand for the data at $H \sim 1330$ G(1.5 MHz). In both cases the impurity concentration $X = 43 \times 10^{-6}$ and $T_g = 86$ mK.

tribution to the host-nuclear-spin relaxation is given by the Bionett, de Gennes, and Silhouette[10] (BGS) rate. This can be written as

$$T_1^{-1}=C\frac{k_BT}{\hbar\omega_n}\mathrm{Im}\chi(\omega_n)\ . \tag{1}$$

Here $\hbar\omega_n$ is the nuclear Zeeman splitting and $\chi(\omega)$ is the impurity transverse dynamic susceptibility. C, for the purpose of our discussion, can be treated as a constant. Now the problem of calculating T_1^{-1} reduces to the calculation of impurity transverse susceptibility in the spin-glass phase. To do this we employ a time-dependent form of EA mean field. This generalization of EA mean-field theory has been used before in different contexts.[11] The effective mean-field Hamiltonian for an impurity spin S can be written as

$$H=-g\mu_B\vec{h}_0(\vec{r})\cdot\vec{S}\ , \tag{2}$$

where $\vec{h}_0(\vec{r})=\sqrt{q/3}(J/\mu_B)\vec{r}$ is the effective random magnetic field seen by a given impurity due to all others. $\vec{r}$ is a random vector with an associated probability $(2\pi)^{-3/2}\exp(-r^2/2)$. J is the width of the Gaussian distribution assumed for the couplings J_{ij} between the impurities. q is the EA order parameter which for $S=\frac{1}{2}$ satisfies the following equation[5]

$$\tfrac{3}{4}-q=(2\pi)^{-3/2}T'\sqrt{5/q}\times\int d^3r\,re^{-r^2/2}\tanh\left[\frac{2}{T'}\sqrt{q/5}r\right]\ , \tag{3}$$

where $T'=T/T_g$ is the reduced temperature and T_g is the transition temperature defined by $k_BT_g=\sqrt{15/12}J$.

In the usual study of the transverse response of a single spin one applies a large static field and then perturbs this spin by impressing on it a small rotating rf field $\vec{h}_{\mathrm{ext}}(t)$ perpendicular to the direction of the external static field. In the model we are considering there is no external static field. The only static field seen by a given spin $\vec{S}$ is the field $\vec{h}_0(\vec{r})$ due to all other impurities. Thus in our calculations $\vec{h}_0$ replaces the external static field. In the presence of a small rotating rf field $\vec{h}_{\mathrm{ext}}(t)$, the equation of motion for the ith component S_i of a spin $\vec{S}$ becomes[12]

$$i\hbar\frac{dS_i}{dt}=[S_i,H_0]-i\hbar\tau_e^{-1}[S_i(t)-(g\mu_B)^{-1}\chi_0h_{i,\mathrm{tot}}(t)]\ , \tag{4}$$

where χ_0 is the static susceptibility calculated with H_0 only. $h_{i,\mathrm{tot}}(t)$ is the ith component of the net instantaneous field, i.e., $\vec{h}_{\mathrm{tot}}(t)=\vec{h}_0+\vec{h}_{\mathrm{ext}}(t)$. As shown by Spencer and Orbach[12] this relaxation to the total instantaneous field, rather than to the static one alone, is required in order to obtain the correct static limit of the dynamic transverse susceptibility $\chi(\omega)$. Here τ_e^{-1} is the impurity relaxation rate. There are two possible sources of this relaxation process. The most obvious one is the Korringa relaxation of the impurity spin due to its interaction (Kondo) with the conduction electrons. The other could result from the part of the impurity-impurity interaction which has been left out of the mean-field theory. We know of no calculation of τ_e^{-1} in a spin-glass; therefore, we will treat it as a parameter of the theory to be determined by the experiments.

For the actual calculation of $\chi(\omega)$, we take the z axis along the static field $\vec{h}_0$ and

$$\vec{h}_{\mathrm{ext}}(t)=\vec{h}_{\mathrm{ext}}^{+}[h^+=|h^+|(\hat{x}+i\hat{y})/\sqrt{2}]$$

with $|\vec{h}^+|\ll|\vec{h}_0|$. With the use of Eq. (4) and following the same steps as in Ref. 11 to calculate $\langle s^+(\omega)\rangle$ and the retarded spin-flip propagator up to first order in h^+, it is straightforward to obtain the transverse dynamic susceptibility $\chi(\omega,\vec{r})$ as[12]

$$\chi(\omega,\vec{r})=(g\mu_B)^2\frac{\omega_0+i\tau_e^{-1}}{\omega_0-\omega+i\tau_e^{-1}}\frac{\langle s_z\rangle}{\hbar\omega_0}\ , \tag{5}$$

where $\langle s_z\rangle$, as calculated with H_0 only, is

$$\langle s_z\rangle=\tfrac{1}{2}\tanh(\beta\hbar\omega_0/2)\ . \tag{6}$$

Here $\hbar\omega_0=\sqrt{q/3}(J/\hbar)r$ and $\beta=1/k_BT$. Now the use of the imaginary part of Eqs. (5) and (6) in Eq. (1) gives T_1^{-1} as

$$T_1^{-1}(r)=C(g\mu_B)^2k_BT\frac{\hbar\tau_e^{-1}}{(\hbar\omega_0-\hbar\omega_n)^2+(\hbar\tau_e^{-1})^2}\times\frac{\tanh(\beta\hbar\omega_0/2)}{\hbar\omega_0/2}\ . \tag{7}$$

To proceed further, we remark that a fixed value of $\vec{r}$ corresponds to a fixed configuration of the bonds J_{ij}. To make contact with experiments, one has to average over different configurations. This means one has to calculate an average over the random vector $\vec{r}$. For fixed $\vec{r}$ the nuclear magnetization recovery $M(t)$ is proportional to $\exp[-t/T_1(r)]$. The experimental nuclear magnetization recovery shape is thus obtained by averaging this quantity to obtain

$$M(t)=M_0\sqrt{2/\pi}\int_0^\infty dr\,r^2e^{-r^2/2}e^{-t/T_1(r)}\ . \tag{8}$$

Equation (8), together with Eq. (7), then describes the nuclear-spin relaxation in a spin-glass. However, Eq. (7) can further be simplified by noting that

$\hbar\omega_n/\hbar\omega_0 \sim \mu_n H/k_B T_g$, where μ_n is the nuclear magneton and H is the externally applied magnetic field. Even for the largest field strengths for the experiments[6] discussed in this paper $\hbar\omega_n/\hbar\omega_0 \sim 10^{-3}$. Thus $\hbar\omega_n$ in the denominator of Eq. (7) can be neglected and Eq. (7) becomes

$$T_1^{-1}(r) = B \frac{\hbar\tau_e^{-1}/k_B T_g}{qr^2 + \frac{5}{16}(\hbar\tau_e^{-1}/k_B T_g)^2} \times \frac{\tanh[(2/T')\sqrt{q/5}r]}{(2/T')\sqrt{q/5}r}, \quad (9)$$

where $B = 5C(g\mu_B)^2/16k_B T_g$.

Equations (9) and (8) are our central results describing nuclear-spin relaxation in spin-glasses for $T < T_g$ for the case of negligible applied field and an impurity spin of one-half. These can be used to extract information about the impurity-spin correlation time τ_e.

Next we show how one can obtain the nonexponential form for $M(t)$ for $T \ll T_g$, which is seen experimentally.[6] To arrive at this form we assume that for $T \ll T_g$, $\hbar\tau_e^{-1} \ll k_B T_g$. This assumption is consistent with the random freezing of impurity spin below T_g and is supported by μ^+-meson depolarization experiments,[8] which indicates that τ_e^{-1} is scaled by T_g. Further, we note that for $T \ll T_g$, $q \sim 1$, and because of the weight factor $r^2\exp(-r^2/2)$ in Eq. (8) the values of r that are important ~ 1. Thus, in the denominator of Eq. (9), $qr^2 \sim 1$, while due to the assumption made above $(\hbar\tau_e^{-1}/k_B/T_g)^2 \ll 1$, and therefore, $(\hbar\tau_e^{-1}/k_B T_g)^2$ in the denominator of Eq. (9) can be neglected. In addition, for $T \ll T_g$ (i.e., $T' \ll 1$), $\tanh[(2/T')\sqrt{q/5}r] \sim 1$. With these simplifications Eq. (9) for $T \ll T_g$ reduces to

$$\left[\frac{1}{T_1(r)}\right]_{T \ll T_g} = \frac{\sqrt{5}}{2} B \frac{\hbar\tau_e^{-1}}{k_B T_g} \frac{T'}{q^{3/2} r^3}, \quad (10)$$

and hence Eq. (8) for $M(t)$ takes the form

$$[M(t)]_{T \ll T_g} = M_0 \sqrt{2/\pi} \int_0^\infty dr\, r^2 e^{-r^2/2} e^{-t/\alpha r^3}, \quad (11)$$

where

$$\alpha = \frac{2}{\sqrt{5}B} \frac{q^{3/2}}{T'} \left[\frac{\hbar\tau_e^{-1}}{k_B T_g}\right]^{-1}. \quad (12)$$

Equation (11) is our theoretical result for $T \ll T_g$, while the form of nuclear magnetization observed by Bloyet *et al.*[6] is

$$[M(t)]_{\text{expt}} = M_0 e^{-a[1-(1+t/a\tau_n)^{1/2}]} \quad (13)$$

with $a = 0.3$ and τ_n a mean-nuclear-spin relaxation time. In Fig. 2 we plot the result of numerical integration involved on the right-hand side of Eq. (11). We have plotted $\ln[M(t)/M_0]$ as a function of (t/α) for small t. For comparison, in Fig. 3 we plot the same quantity as obtained from the experiments, Eq. (13), as a function of $(t/\alpha\tau_n)$. These two graphs clearly show that the form of our result in Eq. (11) and the experimental expression in Eq. (13) are the same for small time t, i.e., for the values of t which

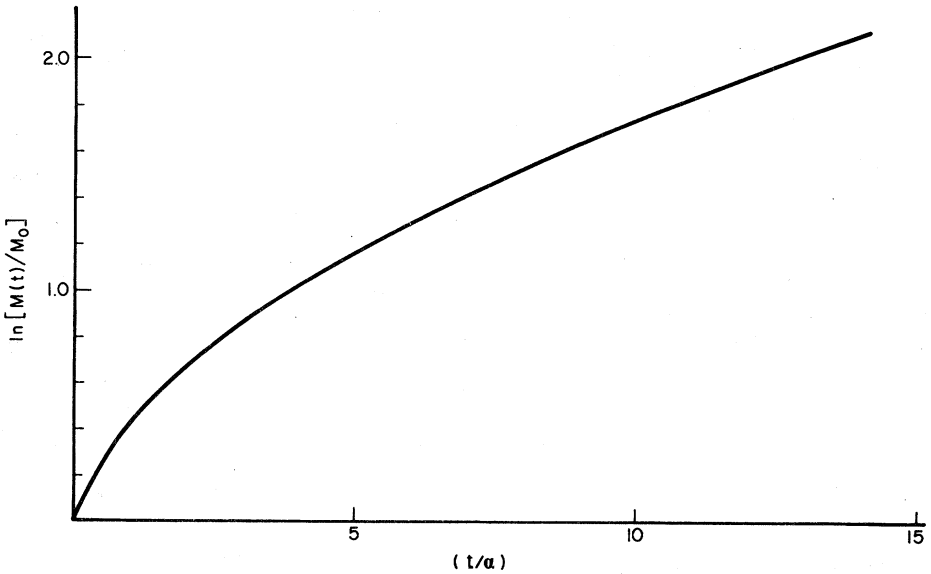

FIG. 2. Form of nuclear-spin magnetization $M(t)$ as predicted by Eq. (11) for $T \ll T_g$. Plotted is $\ln[M(t)/M_0]$ as a function of t/α.

are important from the experimental point of view. This rather excellent agreement suggests that the assumption $\hbar\tau_e^{-1} \ll k_B T_g$ for $T \ll T_g$ made earlier is correct.

We now turn to a discussion of the temperature dependence of τ_e. A quantitative comparison of Figs. 2 and 3 gives $\tau_n = 1.4\tau_n$. Then with the use of Eq. (12) one finds that

$$\tau_n \propto (q^{3/2}/T')(k_B T_g/\hbar\tau_e^{-1}) .$$

Since for $T \ll T_g$, q is a very slow varying function of temperature, the temperature dependence of τ_n mostly represents the variation of the impurity relaxation rate τ_e^{-1} with temperature. Thus given τ_n as a function of T, one can calculate $\tau_e^{-1}(T)$ for $T \ll T_g$.

Now we compare the lowest-field result ($H_0 \sim 275$ Oe) of Ref. 6 with our zero-field theory as regards the temperature dependence and try to extract the temperature dependence of τ_e. Since these fields are still substantial ($\mu_B H \sim k_B T_g$) and spin-glasses are known to be sensitive to the applied fields,[2] only a qualitative comparison is appropriate. For very low T ($T \ll T_g$), τ_n varies approximately as $1/T$, which gives τ_e^{-1} as a constant. As the temperature increases τ_n starts falling as $1/T^2$, and than at a much faster rate τ_e^{-1} correspondingly increases until $\hbar\tau_e^{-1}/k_B T_g$ becomes comparable to 1. At this point the $(\hbar\tau_e^{-1}/k_B T_g)^2$ term in the denominator of Eq. (9) can no longer be neglected. As τ_e^{-1} continues to increase the $(\hbar\tau_e^{-1}/k_B T_g)^2$ term in the denominator of Eq. (9) starts to dominate. Since now $\tau_n \propto (\hbar\tau_e^{-1}/k_B T_g)^{-1}$, τ_n starts increasing. This behavior gives the minimum in the experimental data[6] shown in Fig. 1.

It should be mentioned that as T_g is approached from below and $\hbar\tau_e^{-1} \to k_B T_g$, the shape of the nuclear magnetization recovery changes. However, this can be accounted for by giving a small r dependence to τ_e^{-1} near T_g. This r dependence of τ_e^{-1} means that near T_g, instead of a single relaxation time τ_e, we need to consider a distribution of relaxation times $\tau_e^{-1}(r)$ which is not at all unreasonable (see the discussion of the end of the paper). Near T_g there is also some temperature dependence due to $\tanh[(2/T')\sqrt{q/5r}]$, which is not crucial until $T \to T_g$ ($T' \to 1$), $q \to 0$, and one recovers the exponential decay for the nuclear magnetization. Thus the general shape of the experimental curve τ_n vs T shown in Fig. 1 can be explained by assuming that for $T \ll T_g$, $\hbar\tau_e^{-1} \ll k_B T_g$, while for $T \gg T_g$, $\hbar\tau_e^{-1} \gg k_B T_g$.

Although our theory is restricted to the case of negligible applied field, the field dependence of the data[6] shown in Fig. 1 can be understood, qualitatively, by appealing to what is known about the effects of a magnetic field on the properties of a spin-glass. Since our calculations involve the susceptibility its field dependence is particularly valuable. Generally, the effect of finite fields is to broaden the transition

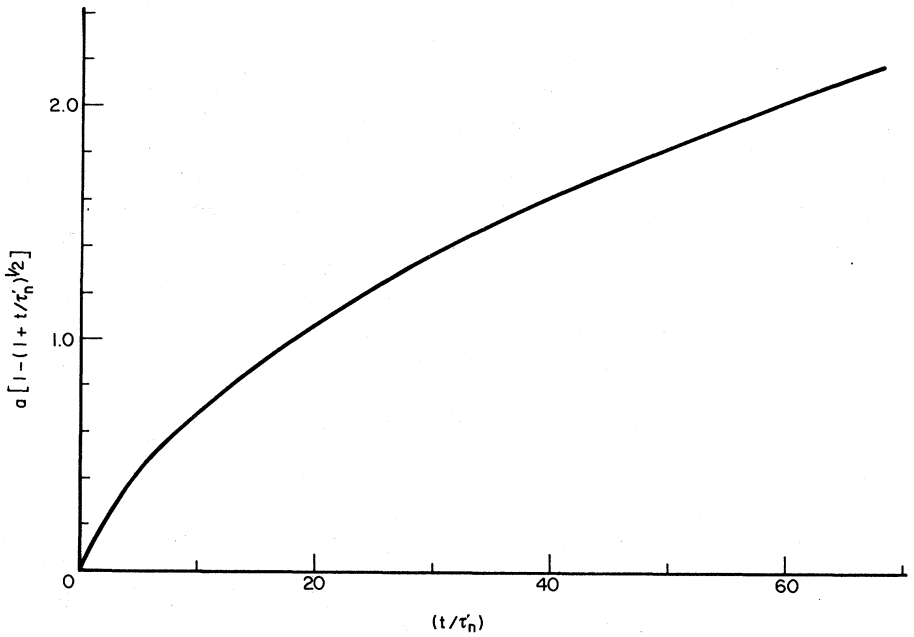

FIG. 3. Experimental curve Eq. (13) (Ref. 6) for nuclear-spin magnetization $M(t)$. Plotted is $\ln[M(t)/M_0]$ as a function of $t/\tau_n' = t/a\tau_n$, with $a = 0.3$.

by making the order parameter nonzero for $T > T_g$.[13] In addition, it shifts the maximum of the susceptibility from $T = T_g$ to lower temperatures.[13,14] Now if we compare the experimental results of Ref. 6 (Fig. 1) for the two lowest-field strengths ($H_0 \sim 275$ and ~ 330 Oe), we find indications of both of these characteristics. The maximum in τ_n seems to shift to lower temperatures as H is increased and, also, the minimum appears to be less pronounced. This trend would indicate that in the limit of zero applied field, the minimum would occur at the spin-freezing temperature and the transition would be a sharp one. This means that the condition $\hbar\tau_e^{-1}/k_B T_g \sim 1$ would occur at the transition temperature and the change from $\hbar\tau_e^{-1} \ll k_B T_g$ would take place over a more restricted range of temperatures around T_g. It is very comforting to note that the recent zero-field experiments by Chen and Slichter[7] on a very different spin-glass system do show a minimum at the transition temperature. Thus these latter experimental results are not inconsistent with the results of Bloyet *et al.*

Another difference between our theory and the experimental situation is the impurity (Mn) spin. The impurity spin for the samples considered is $\frac{5}{2}$ and not $\frac{1}{2}$ as our theory assumes. The major effect of having $S > \frac{1}{2}$ is to increase the field dependence. This would not change any of our qualitative conclusions.

As indicated before the picture that the impurity-spin self-correlation time τ_e changes from $\hbar\tau_e^{-1} < k_B T_g$ to $\hbar\tau_e^{-1} \gg k_B T_g$ around T_g as the temperature is increased is in agreement with the recent μ^+-meson depolarization experiments by Uemura *et al.*[8] In fact, if we use their results for the impurity correlation time $\tau_e(T)$, after properly rescaling with T_g, in our result Eq. (12) we obtain the curve for τ_n vs T shown in Fig. 4. This has a shape which is very similar to the one seen experimentally[6] for $T < 0.5 T_g$ and for the lowest field (Fig. 1).

To summarize, based on EA mean-field theory[4,5] we have derived an expression, Eq. (8), which along with Eq. (9) describes nuclear-spin relaxation in a dilute spin-glasss for the case of negligible applied field. The theory is valid for $T < T_g$. This can be used to extract the temperature dependence of the impurity-spin self-correlation time τ_e. In spite of the fact that our theory is restricted to the zero-field case, the main features of the available data,[6] which involves finite fields, can be understood if one assumes that τ_e^{-1} changes around T_g, such that for $T \ll T_g$, $\hbar\tau_e^{-1} \ll k_B T_g$, and for $T \gg T_g$, $\hbar\tau_e^{-1} \gg k_B T_g$.

We believe it is important to point out that in Ref. 6 it was incorrectly assumed that

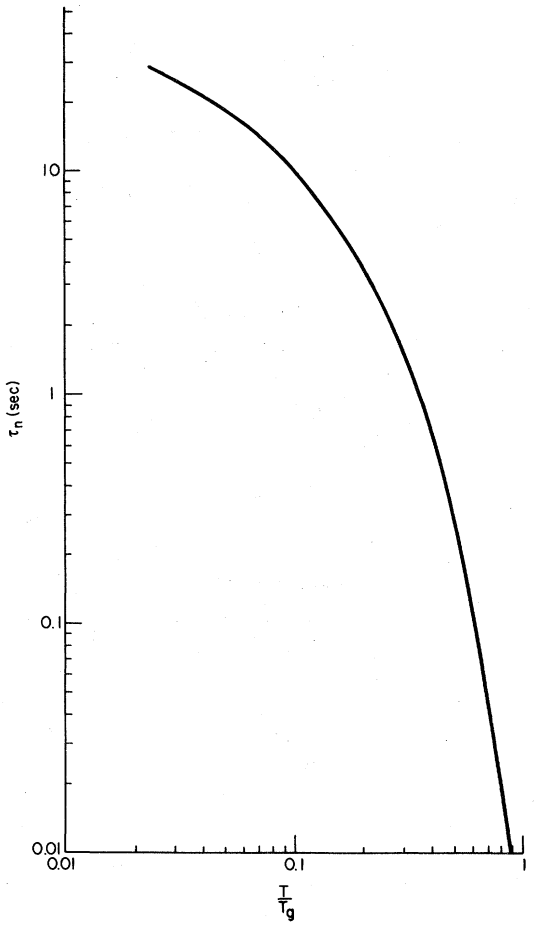

FIG. 4. τ_n as a function of temperature as given by Eq. (12), with the use of the temperature dependence of τ_e as measured by μ^+-depolarization experiments (Ref. 9). τ_e has been scaled with proper T_g.

$$\mathrm{Im}\chi(\omega_n) \sim \tau_e^{-1}/(\omega_n^2 + \tau_e^2) ,$$

and therefore, it was argued that the minimum (see Fig. 1) corresponds to $\omega_n \sim \tau_e^{-1}$. However, as we have mentioned before, ω_n is small compared to other energy scales, namely ω_0 (as well as $g\mu_B H$) and hence it drops out. The minimum actually corresponds to

$$\hbar\tau_e^{-1}/\tau\omega_0 = \hbar\tau_e^{-1}/k_B T_g \sim 1 .$$

Since the estimation of Bloyet *et al.*[6] of τ_e^{-1} rested on an incorrect assumption, their estimates of τ_e^{-1} are in error by approximately 3 orders of magnitude.

We conclude by discussing some of the recent developments in the theory of spin-glasses and their relevance to our calculations. The work during the past few years has shown that the EA order parameter, which is purely static, is not sufficient to describe the spin-glass phase completely.[15,16] In par-

ticular, computer simulations[15] by Kirkpatrick and Sherrington of an infinite range model of the same authors[17] have indicated that the EA order parameter decays to zero. Instead, several authors have proposed dynamic theories.[18–23] In these theories the spin-glass phase is characterized by time-persistent spin correlations, which decay over macroscopically large time scales. It is commonly assumed that this slow relaxation occurs in a distribution of many large time scales.[19,20] Further, instead of using only one order parameter, one employs a large number of order parameters, q being one of them.[19,24]

At present these theories are not capable of yielding any estimates of the time scales for the slow relaxation. However, the authors of Refs. 18 and 19 cite susceptibility measurements[25] in support of this slow relaxation. The experiments[25] show that the magnetization, as predicted by EA mean-field theory for $T < T_g$, decays to its true equlibirium (i.e., truly time-independent) value in a time, which is of the order of hours. Since the time of measurements in nuclear-spin relaxation is very small ($\sim 10^{-7}$ s), this slow relaxation is unlikely to affect the validity of our results. We are implicitly assuming that q remains the most important order parameter for the short times involved in T_1^{-1} measurements. As regards the distribution of many time scales our calculations indicate that taking into account this distribution is perhaps more important near T_g than far below T_g (recall that near T_g we had to give a small r dependence to τ_e^{-1}).

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