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Theory of high-temperature nuclear-spin relaxation in spin-glasses

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Using a soft-spin model with random exchange couplings and a Langevin equation to describe impurity spin dynamics, a high-temperature result for the host nuclear-spin relaxation rate T_1^{-1} in a spin-glass is obtained. The result shows that T_1^{-1} diverges as $(T-T_g)^{-1}$, where T_g is the spin-glass transition temperature, as T_g is approached from above. This is in general agreement with the very few data available.

Most of the current interest in the subject of spin-glasses started with the Edwards-Anderson mean-field theory,[1,2] which predicted a very peculiar kind of phase transition. In this type of phase transition, the order that sets in below the transition temperature T_g is in time rather than the usual order in space. Because of this, in the study of spin-glasses, impurity dynamics takes a special significance. In fact, a host of different experimental methods, including μ^+ depolarization,[3,4] Mössbauer effect,[5] neutron scattering,[6] ESR,[7] and NMR,[8–11] have been employed to investigate impurity spin dynamics in these systems. Among these have been the NMR studies of the nuclear-spin relaxation rate T_1^{-1} of the host metal.[9–11] Measurements of T_1^{-1} can yield information about the transverse response of the impurity spin at the nuclear Larmour frequency ω_n. In this paper, we present a high temperature ($T > T_g$) calculation of T_1^{-1} in a spin-glass. Our result shows that the host nuclear-spin relaxation rate diverges as $(T-T_g)^{-1}$ as T_g is approached from above. This appears to be in agreement with the very few data available.[9–11]

We begin by specifying the model we employ. To describe the impurity spin system we use a soft-spin model with random exchange interactions. A mean-field theory will be employed to solve this model and the impurity spin dynamics will be described by a Langevin equation. The interaction of the conduction electron with the host nuclear spin $\vec{\tau}_n$ and the impurity spin will be treated in second-order perturbation theory. There is some apparent redundancy involved in treating the impurity-impurity interaction and the conduction electron-impurity coupling as independent, since impurity-impurity interaction is itself a result of treating the conduction electron-impurity coupling in second order, giving us RKKY (Ruderman-Kittel-Kasuya-Yosida[12]) interaction among impurities. However, a recent calculation by Jayaprakash *et al.*,[13] using a thermodynamic scaling approach, shows that if one starts out with noninteracting impurities (but interacting with electrons via Kondo coupling), as the cutoff is reduced to construct an effective Hamiltonian, one gets, in addition to the rescaled Kondo (impurity-electron) coupling, the RKKY couplings between the impurities. The Hamiltonian that we implicitly use in these calculations should therefore be understood as the renormalized Hamiltonian rather than the one with bare couplings. Further, we will assume that the system is dilute enough that, while considering bulk host nuclear-spin relaxation, any direct couplings between the host nuclear spin and the impurity spin can be neglected.

As stated above, to deal with the impurity-impurity interactions, we employ a soft-spin model[14] with random exchange interactions. This model has been used before to describe a spin-glass.[15] The Hamiltonian is written as

$$H=-\frac{1}{2}\sum_{ij}J_{ij}\vec{S}_i\cdot\vec{S}_j-g\mu_B\sum_i\vec{S}_i\cdot\vec{H}_i \ . \tag{1}$$

Here $\vec{H}_i$ is the external magnetic field at the site i. The spin variables $\vec{S}_i$ are classical, with variable length, and an associated probability

$$P(\vec{S}_i)=\exp[-\tfrac{1}{2}r_0\vec{S}_i^2-\tfrac{1}{4}u(\vec{S}_i^2)^2] \ . \tag{2}$$

The couplings J_{ij} are random and each of these have an independent Gaussian distribution with zero mean. As remarked in Ref. 15, this model has a formal similarity with Landau-Ginzburg ϕ^4-field theory. This can be seen by observing that the thermal averages involve calculating the partition function

$$Z=\int\prod_{i=1}^{N}d^3S_iP(\vec{S}_i)\exp(-\beta H) \ . \tag{3}$$

Z, upon using Eqs. (1) and (2) for H and $P(\vec{S}_i)$, can be written as[15]

$$Z=\int\prod_{i=1}^{N}d^3S_i\exp(-H_{\rm eff}) \ , \tag{4}$$

where the effective (dimensionless) Hamiltonian $H_{\rm eff}$

is given by

$$H_{\text{eff}}=\frac{1}{2}\sum_{ij}[\,(r_0\delta_{ij}-K_{ij})\vec{S}_i\cdot\vec{S}_j + \tfrac{1}{2}u\,\delta_{ij}(\vec{S}_i^{\,2})^2+\vec{h}_i\cdot\vec{S}_i\delta_{ij}]\ , \tag{5}$$

where we have defined the dimensionless random coupling $K_{ij}\equiv\beta J_{ij}$ and the reduced field $\vec{h}_i = g\mu_B\vec{H}_i/k_BT$. From the above, it is clear that the form of this effective Hamiltonian is the same as that occurring in Landau-Ginzburg ϕ^4 theory. Here our spin variables, however, are defined on a discrete set of lattice points. This formal similarity helps in the perturbation expansion, where one treats both u and K_{ij} as small.

Turning to the problem of nuclear-spin relaxation, we remark that in order to make contact with the experiment, in addition to the thermal averaging, one has to calculate the average over the different configurations. For example, the average over the distributions of K_{ij}'s or J_{ij}'s, we will denote such an average by a set of angular brackets with a subscript c. Thus the experimental nuclear-spin magnetization recovery is proportional to

$$\langle\exp[-t/T_1(J_{ij})]\rangle_c \equiv\int\prod_j dJ_{ij}\,P(J_{ij})\exp[-t/T_1(J_{ij})]\ , \tag{6}$$

where $T_1^{-1}(J_{ij})$ is the nuclear-spin relaxation rate for a fixed configuration.

To decide what should be used for $T_1(J_{ij})$, we remark that if one treats the impurity-conduction electron problem in a molecular-field approximation, one gets two impurity contributions to the nuclear-spin relaxation rate, namely, the BGS (Benoit, de Gennes, and Silhouette) rate and the GH (Giovannini and Heeger) rate.[16] As discussed in Ref. 16, it is difficult to evaluate the relative sizes of the two contributions. However, at high temperatures and for impurity concentration x comparable to those of some of the data[10] that we refer to at the end, experiments[17] indicate that the BGS is the dominant one. We assume that it remains dominant as T_g is approached from above, as there is no apparent reason to expect otherwise. This rate can be written as[16]

$$\frac{1}{T_1(J_{ij})}=Cx\frac{k_BT}{\hbar\omega_n}\,\text{Im}\chi_{J_{ij}}(\omega_n)\ , \tag{7}$$

where $\hbar\omega_n$ is the nuclear Zeeman splitting and $\chi_{J_{ij}}(\omega_n)$ is the impurity transverse dynamic susceptibility for a fixed configuration. In this paper we shall restrict ourselves to the case of negligible applied field, considering only linear response. In such a case, the transverse and longitudinal susceptibilities become identical, and therefore we shall omit the adjective transverse in the remaining part of this paper. C, for our purposes, is a constant, whose actual value has no significance for our calculations. For further details about C, see Ref. 16.

Having decided upon $T_1(J_{ij})$, we substitute the above expression in Eq. (6) and expand the exponential for small t to get

$$\langle\exp[-t/T_1(J_{ij})]\rangle_c=1-t\left(Cx\frac{k_BT}{\hbar\omega_n}\right)\langle\text{Im}\chi_{J_{ij}}(\omega_n)\rangle_c+\frac{t^2}{2!}\left(Cx\frac{k_BT}{\hbar\omega_n}\right)^2\langle(\text{Im}\chi_{J_{ij}}(\omega_n)^2\rangle_c+\cdots\ . \tag{8}$$

We shall show later that in a certain approximation [basically neglecting terms of $O(1/N)$], all the terms of the above equation which are higher than first order in time t are simply related to the first-order term in t. Thus we first focus on this first-order term. It involves the calculation of the imaginary part of the configuration-averaged impurity dynamic susceptibility $\langle\chi_{J_{ij}}(\omega_n)\rangle_c$.

In order to obtain impurity dynamic susceptibility, we use a Langevin equation[18] to describe the impurity spin dynmaics, introducing a kinetic coefficient Γ_0 and a random noise source $\vec{\xi}_i(t)$. The equation is

$$\frac{\partial\vec{S}_i}{\partial t}=-\Gamma_0\frac{\partial H_{\text{eff}}}{\partial\vec{S}_i}+\vec{\xi}_i(t)\ , \tag{9}$$

where H_{eff} is given by Eq. (5) and $\vec{\xi}_i(t)$ satisfies the following conditions:

$$\langle\vec{\xi}_i(t)\rangle=0\ , \tag{10}$$

$$\langle\vec{\xi}_i(t)\cdot\vec{\xi}_j(t')\rangle=2\Gamma_0\delta(t-t')\delta_{ij}\ . \tag{11}$$

It is usually assumed that the source of this noise is provided by the lattice. However, in metallic spin-glasses, the conduction electrons are also a source of noise.

To calculate the susceptibility, one substitutes the effective Hamiltonian Eq. (5) in the Langevin Eq. (9) to obtain an equation of motion for $\vec{S}_i$. This equation can be solved,[15,18–20] and the result is a Dyson's equation for the configuration-averaged full Green's function $\langle G_{ij}(\omega)\rangle_c$ (Ref. 15):

$$\langle G_{ij}(\omega)\rangle_c=g_i^0(\omega)\delta_{ij}-g_i^0(\omega)\,\Sigma_i(\omega)\langle G_{ij}(\omega)\rangle_c]\ , \tag{12}$$

where the self-energy $\Sigma_i(\omega)$ for $T>T_g$ is given by[15]

$$\Sigma_i(\omega)=3uG_{ii}(\omega=0)-\sum_l\Delta_{il}\langle G_{ll}(\omega)\rangle_c\ ,$$

where $g_i^0(\omega)=(i\omega/\Gamma_0+r_0)^{-1}$ is the zeroth-order Green's function and Δ_{ij} is essentially the square of the width of the Gaussian distribution for K_{ij}'s.

To obtain the imaginary part of the response function, we Fourier transfer the above equation and absorb the zero-frequency part $\Sigma(k,0)$ of the self-energy in the zero-order correlation function by defining $G_0(k)=[r_0+\Sigma(k,0)]^{-1}$. The result is

$$G^{-1}(k,\omega)=G_0^{-1}(k)-i\omega\Gamma_0^{-1}+\Sigma(k,\omega)-\Sigma(k,0) \ , \tag{14}$$

where $G(k,\omega)$ is the Fourier transfer of $\langle G_{ij}(\omega)\rangle_c$. Next, we define the physical kinetic coefficient by[15]

$$\Gamma^{-1}(\omega)=\Gamma_0^{-1}+\frac{\partial\Sigma(k,\omega)}{\partial(-i\omega)} \ . \tag{15}$$

We are interested in the response at the nuclear Larmour frequencies ω_n. Since $\hbar\omega_n$ is small compared to other energy scales in the problem (e.g., k_BT_g), we can consider only the low-frequency response function. If we now integrate the above equation from $\omega=0$ to ω, and use the result in Eq. (14), we obtain for low frequencies

$$G^{-1}(k,\omega)=G_0^{-1}(k)-i\omega\Gamma(0) \ . \tag{16}$$

This immediately gives the imaginary part of $G(k,\omega)$, in the limit $\omega\to 0$, as

$$\mathrm{Im}G(k,\omega)=\frac{\omega}{\Gamma(0)}G_0^2(k) \ . \tag{17}$$

To study the temperature dependence of this, we remark that for $T>T_g$, we essentially have free spins and G_0 is regular ($\sim 1/k_BT$). Thus for any possible singularity that may appear as T_g is approached from above, we examine the T dependence of $\Gamma^{-1}(0)$. It has been shown in Ref. 15 that $\Gamma^{-1}(0)$ diverges at T_g and for $T\gtrsim T_g$ is given by[15]

$$\Gamma^{-1}(0)\cong 2\Gamma_0^{-1}(T/T_g-1)^{-1} \ . \tag{18}$$

Substituting this value of $\Gamma^{-1}(0)$ in Eq. (17), and Fourier transforming back to real space, we finally obtain the configuration-averaged response function for $T\gtrsim T_g$ and $\omega\to 0$ as

$$\langle\mathrm{Im}G_{ij}(\omega)\rangle_c=\frac{2\omega}{\Gamma_0}G_0^2\frac{T_g}{T-T_g}\delta_{ij} \ . \tag{19}$$

Since

$$\langle\mathrm{Im}\chi_{J_{ij}}(\omega)\rangle_c=g\mu_B\langle\mathrm{Im}G_{ij}(\omega)\rangle_c \ ,$$

this completes our calculations of the first-order term in time t in Eq. (8).

As a next step in our calculation, we turn to the second-order term in Eq. (8). It is essentially given by $\langle[\mathrm{Im}G_{ij}(\omega)]^2\rangle_c$. We now show it is simply related to $\langle\mathrm{Im}G_{ij}(\omega)\rangle_c$. For this purpose, we write it as

$$\langle[\mathrm{Im}G_{ij}(\omega)]^2\rangle_c=-\tfrac{1}{4}\{\langle[G_{ij}(\omega)]^2\rangle_c - 2\langle G_{ij}(\omega)G_{ij}^*(\omega)\rangle_c + \langle[G_{ij}^*(\omega)]^2\rangle_c\} \ . \tag{20}$$

We first evaluate the first term $\langle[G_{ij}(\omega)]^2\rangle_c$ on the right-hand side of the above equation. Noting that since $\langle K_{ij}\rangle_c=0$, one must pair up K_{ij}'s to get nonzero results, one gets a self-consistent equation for $G_{2,ij}(\omega)\equiv\langle[G_{ij}(\omega)]^2\rangle_c$, which can be represented by Fig. 1. There a single line stands for $\langle G_{ij}(\omega)\rangle_c$, while two lines with a circle around them represent $G_{2,ij}(\omega)$. The equation itself is

$$G_{2,ij}(\omega)=\langle G_{ij}(\omega)\rangle_c^2+\langle G_{ii}(\omega)\rangle_c^2\sum_l\Delta_{il}G_{2,lj}(\omega) \ . \tag{21}$$

To solve this equation, we Fourier transform to k space and use the fact that there is no spatial correlation, i.e., $\langle G_{ij}(\omega)\rangle_c=G(\omega)\delta_{ij}$, to get

$$G_2(k,\omega)=\frac{G^2(\omega)}{1-\Delta(k)G^2(\omega)} \ . \tag{22}$$

Now we assume that $\Delta(k)=\Delta(0)\delta_{k,0}$, which simply means that all distribution $\mathcal{P}(K_{ij})$'s have the same width. With this assumption, if we Fourier transform back to real space, we obtain

$$G_{2,ij}(\omega)=\langle G_{ij}(\omega)\rangle_c^2\delta_{ij}+\frac{1}{N}\frac{G^4(\omega)\Delta(0)}{1-\Delta(0)G^2(\omega)} \ . \tag{23}$$

Clearly, the second term on the right-hand side is of the order $1/N$ smaller than the first and hence can be neglected. Thus the above equation reduces to

$$G_{2,ij}(\omega)\equiv\langle[G_{ij}(\omega)]^2\rangle_c=\langle G_{ij}(\omega)\rangle_c^2 \ . \tag{24}$$

Similarly, it can be shown that

$$\langle G_{ij}^*(\omega)G_{ij}(\omega)\rangle_c=\langle G_{ij}^*(\omega)\rangle_c\langle G_{ij}(\omega)\rangle_c$$

and $\langle(G_{ij}^*)^2\rangle_c=\langle G_{ij}^*\rangle_c^2$. From the use of the above three results, it follows that to leading order in $1/N$

$$\langle[\mathrm{Im}G_{ij}(\omega)]^2\rangle_c=\langle\mathrm{Im}G_{ij}(\omega)\rangle_c^2 \ . \tag{25}$$

As we already have a result for $\langle\mathrm{Im}G_{ij}(\omega)\rangle_c$ [Eq. (19)], we have completed the calculation of the second-order term in Eq. (8).

Following the same argument, it can be shown that

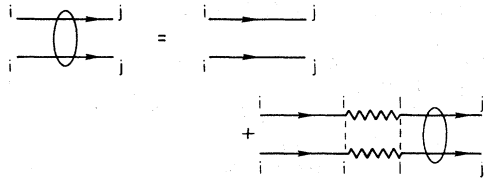

FIG. 1. Graphical representation of the self-consistent Eq. (20) for $G_{2,ij}=\langle G_{ij}^2\rangle_c$. A single line represents $\langle G_{ij}\rangle_c$, while two lines tied up by a circle stand for $G_{2,ij}$.

to leading order in $1/N$,

$$\langle [\mathrm{Im}G_{ij}(\omega)]^n \rangle_c = \langle \mathrm{Im}G_{ij}(\omega) \rangle_c^n \ . \tag{26}$$

Thus all higher-order terms are related in a simple way with the first-order term in Eq. (8) and results such as Eqs. (25) and (26) allows us to resum the series in Eq. (8). The result is that we get back an exponential form for the nuclear magnetization recovery $M(t)$ for $T \gtrsim T_g$ given by

$$M(t) = M(0)e^{-t/T_1} \ , \tag{27}$$

where

$$\frac{1}{T_1} = \frac{\gamma}{T - T_g} \ , \tag{28}$$

with $\gamma = (2g\mu_B k_B/\hbar\Gamma_0)Cx$. In arriving at this result we have also used Eq. (19) for $\langle \mathrm{Im}G_{ij}(\omega_n) \rangle_c$. This is our high-temperature result for nuclear-spin relaxation rate T_1^{-1} in a dilute spin-glass, which shows that T_1^{-1} diverges as $(T - T_g)^{-1}$ as T_g is approached from above and nuclear-spin relaxation has an exponential form.

This result seems to be in agreement with the very incomplete data available,[9] which does appear to show a divergence near T_g. However, this data suffers from some intrinsic time resoltuion difficulties.[21] There is also some data available in very dilute spin-glasses[10] which again show a divergence in the neighborhood of T_g. Here, also, there is a problem in directly comparing this zero-field theory, because of the presence of finite magnetic fields ($\mu_B H \sim k_B T_g$) in these experiments. However, as we will discuss in some detail elsewhere,[22] these experiments are not inconsistent with the above result. The only clear-cut zero-field data is by Chen and Slichter.[11] They have recently observed a maximum in T_1^{-1} at T_g in a similar spin-glass system. Further, their data is also consistent with the mean-field exponent of -1 that we have obtained here.

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