# Relaxation dynamics of quantum spin glasses: Role of heat-bath coupling

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We have studied the role of heat-bath coupling in a quantum spin glass described by a previously developed random-bond random-field Ising model in a transverse field, which accounts for quantum tunneling of protons in a disordered hydrogen-bonded ferroelectric. In contrast to the stochastic model considered earlier, it is assumed that proton tunneling is not directly affected by the thermal fluctuations and thus remains coherent. The basic mechanism that renders the system dissipative is heat-bath-assisted jumps of protons across the potential barrier. An effective single-spin Hamiltonian is obtained on the basis of a thermofield dynamic approach in the short-time limit. A resolvent expansion of the underlying time-evolution operator is then set up and the resulting resonance line shape calculated to leading order in the perturbation theory. It is shown that at low frequencies the line shape exhibits a singular contribution that diverges as $\sim |\omega|^{-1/2}$, but disappears in the classical limit of zero tunneling frequency. This singularity is due to coherent tunneling motion and is thus a typical quantum effect which could be observed in the NMR, NQR, or EPR line shapes in the appropriate temperature range.

## I. INTRODUCTION

A quantum spin glass is a system with quenched disorder described by cooperative random interactions between quantum spins. The subject is of considerable contemporary interest and is discussed extensively in the literature.[1–16] A complete theoretical understanding of a quantum spin glass requires very elaborate and complicated techniques, and therefore, it is considered important to analyze the statistical mechanics of simple models. A typical example of these is a disordered Ising model in the presence of a transverse field, which describes quantum tunneling processes.[1–16]

A particular physical realization of this model is found in so-called proton glasses, i.e., randomly mixed hydrogen-bonded ferroelectric and antiferroelectric crystals, such as $Rb_{1-x}(NH_4)_xH_2PO_4$, also known as RADP. Here the proton can occupy two sites in an O—H···O bond which are customarily mapped onto the two states of an Ising pseudospin variable $S^z = \pm 1/2$. The static cooperative glasslike behavior of this system is described by a Hamiltonian for the interactions between all the pseudospins at different sites, which is characterized by both random bonds and local random fields.[2] The additional feature, which distinguishes RADP from its deuterated analog D-RADP, is the presence of a transverse field which accounts for quantum tunneling of the proton from one site of the bond to the other. In spite of the accumulation of a large body of experimental data on the deuterated systems D-RADP,[17–19] no extensive studies have been made to date on the tunneling motion of the proton in RADP at low temperatures and, in particular, on the way this motion is affected by the stochastic dynamics due to coupling to the heat bath.

A step toward understanding the basic quantum processes in disordered systems has recently been made by the present authors[20] in a stochastic theory of the resonance line shape in proton glasses. In Ref. 20, henceforth referred to as I, we speculated also on the feasibility of comparing various resonance experiments such as NMR, nuclear quadrupole resonance (NQR), and EPR with the theory. One important issue which still remains open is concerned with the precise nature of the dissipative interaction of the system with the surrounding heat bath. The purpose of this paper is mainly to address this particular question.

It is important at this stage itself to amplify what exactly is the issue concerning the role of the heat bath. The point is that the proton can be transferred from one site to another in an O—H···O bond not merely by the quantum tunneling, but also by a classical, thermally activated mechanism. Indeed, it is the latter which dominates the dynamics of the deuteron in the D-RADP. In proton glasses, however, one expects to see a competition between the coherent tunneling processes and the incoherent thermal activation. The latter, of course, originates from thermal fluctuations that render the system dissipative. In I we have assumed that this heat-bath coupling not only causes random hoppings of the proton from one site to another, but also leads to incoherence in the tunneling process. This is by no means a unique way in which the heat bath can affect the dynamics of the system. In fact, one can imagine it is only the hopping of the proton across a potential barrier that is aided by the stochastic forces from the heat bath, while tunneling remains unaffected and, therefore, coherent. This mechanism of relaxation, different from the one assumed in I, may at first sight appear not too relevant for line shapes, as the heat bath plays only an indirect role. However, we argue in this paper that in proton glasses, which are characterized by quantum interactions, different forms of heat-bath couplings lead to qualitatively different line shapes.

The outline of the paper is as follows: In Sec. II we introduce the basic Hamiltonian following the notation of I and also specify the terms describing the interaction with the heat bath. We then set up a resolvent expansion of the underlying time-evolution operator that enters the relevant line-shape expression. The calculational details are presented in Sec. III and in the Appendix. In Sec. IV we exhibit various numerical plots of the line shape for different values of the parameters. Finally, our main conclusions are summarized in Sec. V.

## II. MATHEMATICAL FORMULATION

The Hamiltonian describing the proton glass system may be written as

$$\mathcal{H} = \Delta\omega \sum_i I_i^z S_i^z - \frac{1}{2}\sum_{ij} J_{ij} S_i^z S_j^z - \Omega \sum_i S_i^x - \sum_i f_i S_i^z, \tag{1}$$

where $I^z$ is a nuclear spin operator whose eigenvalues determine the resonance lines and $\Delta\omega = \omega_L - \omega_R$ is the difference between the resonance frequency in the left and right potential minimum of the proton. $J_{ij}$ is the infinite-ranged quenched random interaction between the pseudospins $S_i^z$, $\Omega$ the tunneling frequency, and $f_i$ represents the random longitudinal field at site $i$. The random interaction $J_{ij}$ and random fields $f_i$ are assumed to be independently distributed according to their respective Gausssian distributions,

$$\mathcal{P}(J_{ij}) = \frac{1}{\sqrt{2\pi J^2}} \exp\left(-\frac{J_{ij}^2}{2J^2}\right), \tag{2}$$

$$\mathcal{P}(f_i) = \frac{1}{\sqrt{2\pi\Delta}} \exp\left(-\frac{f_i^2}{2\Delta}\right). \tag{3}$$

In what follows we shall denote by $[\cdots]_d$ an average with respect to combined distributions (2) and (3). As in I, we want to treat the Hamiltonian (1) within the mean-field theory of quantum spin glasses, which we expect to be appropriate in the context of an infinite-range model.[5,9] This then allows us to carry out further discussion in terms of an effective single-spin Hamiltonian. We adopt here the results of Ref. 7, which have been obtained using the thermofield dynamic (TFD) approach and a short-time approximation for the dynamic self-interaction. The effective single-spin Hamiltonian in the present case reads

$$\mathcal{H}_S = \Delta\omega I^z S^z - hS^z - \Omega S^x. \tag{4}$$

Here $h = h(z)$ is an effective field acting along the $z$ axis and is due to the nonzero spin-glass order parameter $q$,

$$h(z) = \tfrac{1}{2} J z \sqrt{q + \tilde{\Delta}}, \tag{5}$$

where $z$ is the excess static Gaussian random field,[7] and $\tilde{\Delta} \equiv 4\Delta/J^2$. The mean-field equations for the local polarization $p(z)$ and the spin-glass order parameter $q$ are[7]

$$p(z) = r(z) \tanh[\tfrac{1}{2}\beta h_0(z)] \tag{6}$$

and

$$q = \int_{-\infty}^{+\infty} \frac{dz}{\sqrt{2\pi}} e^{-z^2/2} p(z)^2, \tag{7}$$

with

$$h_0(z) = \sqrt{\Omega^2 + h(z)^2} \tag{8}$$

and

$$r(z) = h(z)/h_0(z). \tag{9}$$

In order to describe the dissipative dynamics of the heat bath, we generalize the Hamiltonian in (4) as

$$\mathcal{H}_0 = \mathcal{H}_S + \mathcal{H}_I + \mathcal{H}_B, \tag{10}$$

where $\mathcal{H}_I$ describes the interaction between the spin subsystem and the heat bath. We assume the following type of interaction:

$$\mathcal{H}_I = gbS^x. \tag{11}$$

In (11), $b$ is an operator which acts in the Hilbert space of the heat-bath Hamiltonian $\mathcal{H}_B$ and $g$ is a multiplicative coupling constant. The exact nature of the operators $b$ will not be specified here, except that it is pertinent to remark that the coupling is designed to drive transitions between the eigenstates of $S^z$. In the context of the physical model this mimicks the hopping of the proton from one site to another in the O—H$\cdots$O bond, and is expected to lead to Glauber kinetics for the underlying Ising model if the tunneling were absent. Furthermore, as the coupling term is chosen such that it commutes with the term proportional to $\Omega$, the heat bath does not interfere directly with the tunneling process. This scenario of dissipative dynamics is quite distinct from the one envisaged in I.

As described in I, the effects of dynamics are contained in the bath-averaged time-development operator of the system whose Laplace transform is denoted by $[\hat{U}(s)]_{\text{av}}$, where the variable $s$ is related to the applied frequency $\omega_{\text{app}}$ measured relative to the resonance frequency $\bar{\omega} = (\omega_L + \omega_R)/2$. The average $[\hat{U}(s)]_{\text{av}}$ was evaluated in I using the stochastic theory approach.[21] In the present paper, on the contrary, we adopt a system-plus-reservoir approach in order to give a proper treatment to the coherent tunneling term, and systematically "project out" the bath degrees of freedom. This can be most conveniently achieved by writing a resolvent expansion of $[\hat{U}(s)]_{\rm av}$ in which the interaction term $\mathcal{H}_I$ is treated perturbatively. As discussed in detail in Ref. 22, such an expansion yields the following general expression for $[\hat{U}(s)]_{\rm av}$:

$$[\hat{U}(s)]_{\rm av} = [s - i\mathcal{L}_S + \hat{\Sigma}(s)]^{-1}, \tag{12}$$

where $\mathcal{L}_S$ is the Liouville operator associated with the spin Hamiltonian $\mathcal{H}_S$ in (4) and $\hat{\Sigma}(s)$ is the so-called relaxation matrix, to be specified below. While it is possible to calculate $\hat{\Sigma}(s)$ to arbitrary order in the perturbation theory, it suffices for our purpose to use the expansion to second order in $\mathcal{H}_I$, which gives

$$\hat{\Sigma}(s) = \left(\mathcal{L}_I \frac{1}{s - i\mathcal{L}_S - i\mathcal{L}_B}\mathcal{L}_I\right)_{\rm av}. \tag{13}$$

In the next section we will present the delails of the calculation.

## III. RESONANCE LINE SHAPE

As shown in I, the line shape is obtained from the correlation function

$$\hat{C}(s) = \mathrm{Tr}(\rho_S I^-(0)\{[\hat{U}(s)]_{\rm av} I^+(0)\}), \tag{14}$$

where $\rho_S$ is the density operator associated with the system Hamiltonian $\mathcal{H}_S$, and is given by

$$\rho_S = \exp\left(-\beta\mathcal{H}_S\right)/Z_S, \tag{15}$$

where $Z_S$ is the corresponding partition function. It should be noted that $\rho_S$ is not diagonal in the representation of $S^z$. Also, in writing (14), we have neglected the influence of the term proportional to $\Delta\omega$ on $\rho_S$ because usually $\mid \beta\Delta\omega \mid << 1$. The final step is to write out the trace over the eigenstates of $I^z$ and $S^z$, and obtain

$$\hat{C}(s) = \sum_{\nu\nu'\mu} \langle \mu \mid \rho_S \mid \nu\rangle \times (\nu + \tfrac{1}{2}; \mu - \tfrac{1}{2} \mid [\hat{U}(s)]_{\rm av} \mid \nu' + \tfrac{1}{2}; \nu' - \tfrac{1}{2}), \tag{16}$$

where the states $\mid \pm\frac{1}{2}\rangle$ are the eigenstates of $I^z$, while the states with the Greek indices $\mu$, $\nu$, etc., are the eigenstates of $S^z$. Note that the Hamiltonian (10) is completely diagonal in the $I^z$ representation, and hence maintaining track of the states $\mid \pm\frac{1}{2} >$ is a matter of mere bookkeeping. It is then clear from Eq. (12) that the evaluation of the matrix elements of $[\hat{U}(s)]_{\rm av}$ involves the inversion of the matrix which in the present problem has a dimension 4×4, as the pseudospin $S^z$ is only a half-valued operator. The first piece of this is the matrix of $(+- \mid \mathcal{L}_S \mid +-)$, which has the 4×4 representation as follows:

$$\begin{pmatrix} +\frac{1}{2}\Delta\omega & 0 & +\frac{\Omega}{2} & -\frac{\Omega}{2} \\ 0 & -\frac{1}{2}\Delta\omega & -\frac{\Omega}{2} & +\frac{\Omega}{2} \\ +\frac{\Omega}{2} & -\frac{\Omega}{2} & -h & 0 \\ -\frac{\Omega}{2} & +\frac{\Omega}{2} & 0 & +h \end{pmatrix}, \tag{17}$$

where the rows and columns are labeled by $\mid \mu\nu)$ which take the values $\mid ++)$, $\mid --)$, $\mid +-)$, and $\mid -+)$, respectively.

The next nontrivial object is the relaxation matrix $\hat{\Sigma}(s)$. In accordance with our earlier statement about the density operator, we neglect the influence of the term proportional to $\Delta\omega$ on the relaxation matrix $\hat{\Sigma}(s)$. Then the latter is an operator in the Hilbert space of $S^z$ alone. Furthermore, we treat the heat bath in the Markovian approximation, which implies that

$$\hat{\Sigma}(s) \approx \hat{\Sigma}(0) = \int_0^{\infty} dt\, \{\mathcal{L}_I \exp\left[i\left(\mathcal{L}_S + \mathcal{L}_B\right)t\right]\mathcal{L}_I\}_{\rm av}, \tag{18}$$

where in (18) $\mathcal{L}_S$ is associated with the Hamiltonian

$$\mathcal{H}_S \approx -hS^z - \Omega S^x. \tag{19}$$

Finally, the influence of the heat bath is assumed purely dissipative; i.e., the relaxation matrix $\hat{\Sigma}(s=0)$ is totally real. Neglecting then all imaginary components, and after some lengthy algebra involving the matrix representation of Liouville operators, and regrouping of terms as correlation function of heat-bath operators $b$, we can write down all the elements of $\hat{\Sigma}(s=0)$. A representative sample is

$$\left(++ \mid \hat{\Sigma}(0) \mid ++\right) = \frac{1}{8}g^2 \int_{-\infty}^{+\infty} dt \left[\left(1 + \frac{h}{h_0}\right)e^{-ih_0 t} + \left(1 - \frac{h}{h_0}\right)e^{ih_0 t}\right]\langle\langle b(t)b(0)\rangle\rangle, \tag{20}$$

where $h_0$ is defined in (8), and

$$\langle\langle b(t)b(0)\rangle\rangle \equiv \mathrm{Tr}\left[\rho_B e^{i\mathcal{H}_B t} b(0) e^{-i\mathcal{H}_B t} b(0)\right], \tag{21}$$

$\rho_B$ being the density operator of the heat bath.

As mentioned earlier, we make no attempt to calculate the bath correlation function (21). Instead, we simply parametrize it in terms of a phenomenological relaxation rate $\lambda$ by first making use of the following Kubo relation:

$$\int_{-\infty}^{+\infty} dt\, e^{+ih_0 t}\langle\langle b(t)b(0)\rangle\rangle = e^{\beta h_0}\int_{-\infty}^{+\infty} dt e^{-ih_0 t}\langle\langle b(t)b(0)\rangle\rangle. \quad (22)$$

Using (22), we can write

$$\int_{-\infty}^{+\infty} dt\, e^{\pm ih_0 t}\langle\langle b(t)b(0)\rangle\rangle = \lambda\frac{e^{\pm\beta h_0/2}}{e^{+\beta h_0/2}+e^{-\beta h_0/2}}, \quad (23)$$

where

$$\lambda \equiv \int_{-\infty}^{+\infty} dt\left(e^{+ih_0 t}+e^{-ih_0 t}\right)\langle\langle b(t)b(0)\rangle\rangle = \int_{-\infty}^{+\infty} dt\, e^{ih_0 t}\left[\langle\langle b(t)b(0)\rangle\rangle + \langle\langle b(0)b(t)\rangle\rangle\right]. \quad (24)$$

The quantity written in the square brackets is the time-symmetric correlation function. Now, according to our stated objective, the fluctuations in the heat bath are characterized by frequencies which are assumed to be much larger than the frequency associated with $h_0$, in which case $\lambda$ becomes real, approximately given by the following expression:

$$\lambda \approx \int_{-\infty}^{+\infty}\left[\langle\langle b(t)b(0)\rangle\rangle + \langle\langle b(0)b(t)\rangle\rangle\right]. \quad (25)$$

It may be remarked that $\lambda$ is related to the so-called Kubo correlation time $\tau_c$ via

$$\lambda = 2\tau_c\langle\langle b^2\rangle\rangle, \quad (26)$$

where

$$\tau_c \equiv \int_{-\infty}^{+\infty} dt\frac{\langle\langle b(t)b(0)\rangle\rangle + \langle\langle b(0)b(t)\rangle\rangle}{2\langle\langle b^2\rangle\rangle}. \quad (27)$$

Collecting all the terms and introducing the Liouville operator $(S^z)^\times$ (Ref. 22) associated with $S^z$, the matrix of $\hat{\Sigma}(s=0)$ in the 4×4 space of $(S^z)^\times$ can be written as

$$\frac{\lambda}{2}\begin{pmatrix} 1+\frac{h}{h_0}(p_- - p_+) & -1-\frac{h}{h_0}(p_- - p_+) & 0 & 0 \\ -1+\frac{h}{h_0}(p_- - p_+) & 1-\frac{h}{h_0}(p_- - p_+) & 0 & 0 \\ 0 & 0 & +2 & -2 \\ 0 & 0 & -2 & +2 \end{pmatrix}, \quad (28)$$

where $p_\pm$ are respective probabilities for the states $|\pm\rangle$.

Combining (17) and (28) it becomes evident that in order to obtain the matrix $[\hat{U}(s)]_{\rm av}$, we have to invert the following matrix denoted by $M$:

$$\begin{pmatrix} i\omega - \frac{i}{2}\Delta\omega + \frac{\lambda}{2}(1-p) & -\frac{\lambda}{2}(1-p) & -i\frac{\Omega}{2} & +i\frac{\Omega}{2} \\ -\frac{\lambda}{2}(1+p) & i\omega + \frac{i}{2}\Delta\omega + \frac{\lambda}{2}(1+p) & +i\frac{\Omega}{2} & -i\frac{\Omega}{2} \\ -i\frac{\Omega}{2} & +i\frac{\Omega}{2} & i(\omega+h)+\lambda & -\lambda \\ +i\frac{\Omega}{2} & -i\frac{\Omega}{2} & -\lambda & i(\omega-h)+\lambda \end{pmatrix}. \quad (29)$$

Here $p = [h(z)/h_0(z)]\,(p_+ - p_-)$ represents the local polarization for a given value of excess random field $z$. We have performed this task analytically and the detailed expressions for the elements of the inverse matrix $M^{-1}$ are given in the Appendix. Using these matrix elements, the relevant correlation function can be obtained from the expression (16). Finally, we recall that the observed line shape is obtained by averaging the correlation function over the distribution of local polarization $p$. In view of Eq. (6), this step is tantamount to performing averaging over the excess static noise field $z$ which is present in the effective single-spin Hamiltonian (4). We have, for the spectral line shape,

$$J(\omega) = \frac{1}{4\pi}\int_{-1}^{+1} dp\, W(p)J(\omega,p), \quad (30)$$

where

$$J(\omega,p) = \mathrm{Re}[\hat{C}(s=i\omega)], \quad (31)$$

and $\hat{C}(s)$ is given by (16). According to Eq. (16) and using the corresponding matrix elements of the density operator $\rho_S$ defined in (15) and of $[\hat{U}(s=i\omega)]_{\rm av}$ (see the Appendix), we finally obtain the expression for the line shape at given configuration of local polarizations $p$:

$$J(\omega,p) = \frac{8\bar{\lambda}}{\Delta\omega}\frac{(1-p^2)Q_0 + \bar{\Omega}^2X^2Q_1 + \bar{\Omega}X\sqrt{q+\tilde{\Delta}z(p)}Q_2}{\left[(\omega^2-1)^2+\bar{\lambda}^2(\omega-p)^2\right]Q_0 + \omega^2\bar{\Omega}^2X^2Q_3}, \quad (32)$$

where:

$$Q_0 = [X^2(q+\tilde{\Delta})z^2 - \omega^2]^2 + 4\bar{\lambda}^2\omega^2,$$

$$Q_1 = [\omega^2 - X^2(q+\tilde{\Delta})z^2]p^2 + 2\omega(\omega+p),$$

$$Q_2 = 4(1-\omega^2)(\omega+p) + 2\omega[\omega^2 - X^2(q+\tilde{\Delta})z^2](1-p^2) - 4\bar{\lambda}^2p^2(\omega-p) - 2\omega p^2\bar{\Omega}^2X^2,$$

$$Q_3 = 2(\omega^2-1)[X^2(q+\tilde{\Delta})z^2 - \omega^2] + 4\bar{\lambda}^2\omega(\omega-p) + \omega^2\bar{\Omega}^2X^2.$$

Here $z(p)$ represents the values of $z$ which satisfy the Eq. (6), $\omega$ represents a dimensionless frequency $\omega_{\rm app}/\Delta\omega$, and the dimensionless parameters are defined as $\bar{\Omega} \equiv 2\Omega/J$, $\bar{\lambda} \equiv \lambda/\Delta\omega$, and $X \equiv J/\Delta\omega$. The distribution of local polarization $W(p)$ for the present model has been calculated earlier[10] using the TFD approach and the same approximation for the dynamic self-interaction as adopted here. For the case $S^z = \pm 1/2$ it reads

$$W(p) = \frac{4}{\beta J\sqrt{2\pi(q+\tilde{\Delta})}}e^{-z^2/2} \times \left[\left(\frac{h(z)}{h_0(z)}\right)^2 + \frac{2\bar{\Omega}^2}{\beta h(z)h_0(z)^2}p - p^2\right]^{-1}, \quad (33)$$

where $z$ is a function of $p$ in view of Eq. (6). In the limit of zero tunneling $\Omega \to 0$, the expression for $J(\omega,p)$ [Eq. (31)] reduces to the known result for the classical Ising spin glass with Glauber dynamics.[23]

## IV. RESULTS AND DISCUSSION

Equations (32) and (33) imply that at low frequencies the line shape $J(\omega)$ will be dominated by the behavior of $J(\omega,p)$ at small values of $\omega$ and $p$. From Eq. (32) we find for $\omega \ll 1$, $p \ll 1$ the leading contribution

$$J(\omega,p) = \frac{8\bar{\lambda}}{\Delta\omega}\frac{Ap^2}{Bp^4 + C\omega^2}, \quad (34)$$

with

$$A = 4X\bar{\Omega}^2/\tanh(\bar{\beta}\bar{\Omega}), \quad (35)$$

$$B = \left[\bar{\Omega}X/\tanh(\bar{\beta}\bar{\Omega})\right]^4, \quad (36)$$

$$C = 4\bar{\lambda}^2, \quad (37)$$

and $\bar{\beta} = 1/\bar{T}$, where $\bar{T} = 4T/J$.

Inserting (34) into Eq. (30) and replacing $W(p)$ by its value at $p = 0$, we obtain after carrying out the remaining integration the leading singularity of the line shape function:

$$J(\omega)_{\rm sing} = \frac{2}{\pi}\frac{\bar{\lambda}}{\Delta\omega}W(0)\frac{A}{B}\frac{1}{\sqrt{2\hat{\omega}}}\left[\frac{1}{2}\ln\left(\frac{1+\hat{\omega}-\sqrt{2\hat{\omega}}}{1+\hat{\omega}+\sqrt{2\hat{\omega}}}\right) + \arctan\left(\frac{\sqrt{2\hat{\omega}}}{\hat{\omega}-1}\right) + \pi\Theta(1-\hat{\omega})\right]. \quad (38)$$

Here

$$\hat{\omega} = |\omega|(C/B)^{1/2} \quad (39)$$

and $\Theta(x)$ is the usual unit-step function. Thus we see that the line-shape function diverges at low frequencies as

$$J(\omega) \sim |\omega|^{-1/2}. \quad (40)$$

This implies that the correlation function $C(t)$ introduced in I, which is the inverse Laplace transform of $\hat{C}(s)$, behaves at long times as

$$C(t) \sim t^{-1/2}. \quad (41)$$

The singularity of $J(\omega)$ at small $\omega$ is a quantum effect not present in the classical limit $\bar{\Omega} \to 0$. This can easily be seen from the expression (32) in which the singular behavior disappears when $\bar{\Omega} \to 0$. Specifically, the parameter $A$ in Eq. (34) vanishes for $\bar{\Omega} \to 0$, whereas $B$ and $C$ both have a finite limit. The physical reason for the singularity of $J(\omega)$ in the quantum case is the fact that at $p = 0$ the field $h(z)$ in the Hamiltonian (4) disappears. Hence in this limit the heat-bath coupling term (11) commutes with the tunneling term, the latter being the only remaining static field acting on these spins. The protons represented by these pseudospins can tunnel coherently between the two potential minima without experiencing any damping due to the heat bath. This situation is quite different from the one envisaged in I, where by assumption the heat-bath coupling term was such that always had an incoherent effect on the pseudospin Hamiltonian $\mathcal{H}_S$, even in the limit $p \to 0$.

The difference between the complete expression for the line shape (30) and its singular part (38) is a well-behaved function of $\omega$ and can easily be evaluated numerically.

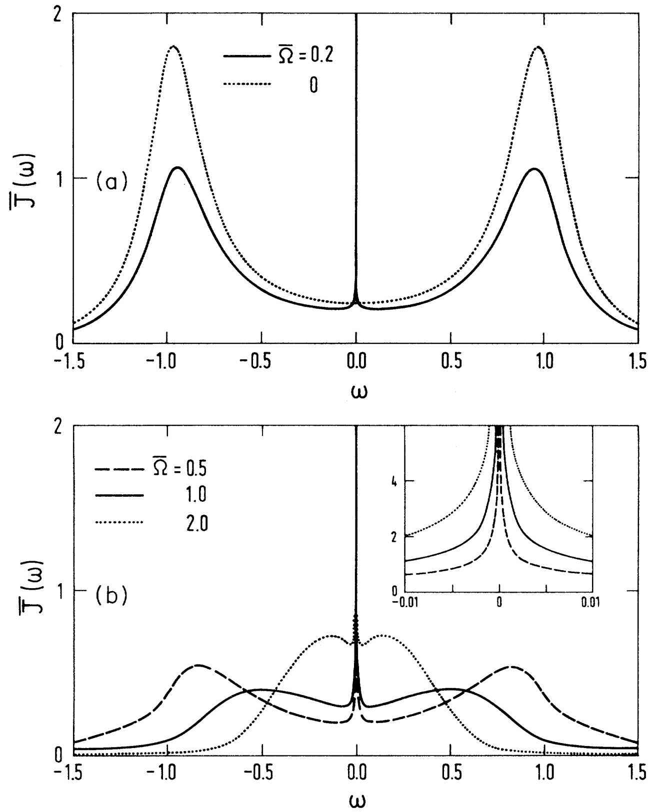


FIG. 1. Calculated line shape $\bar{J}(\omega) \equiv \Delta\omega J(\omega)$ vs reduced frequency $\omega/\Delta\omega$ for fixed temperature $T/J = 1.0$, random-field variance $\tilde{\Delta} \equiv 4\Delta/J = 0.35$, relaxation rate $\bar{\lambda} \equiv \lambda/\Delta\omega = 0.4$, and $X \equiv J/\Delta\omega = 0.01$, and various values of tunneling frequency $\bar{\Omega} \equiv 2\Omega/J$, as indicated. (a) Comparison between classical ($\bar{\Omega} = 0$) and a quantum system ($\bar{\Omega} = 0.2$). (b) Line shape in the quantum case for three values of $\bar{\Omega}$. Inset: enlarged view of the central singularity.

The results obtained for various representative values of the model parameters are shown in Figs. 1–3. In Figs. 1(a) and 1(b) the tunneling frequency $\bar{\Omega}$ has been varied from zero up to the value 2.0 with the reduced

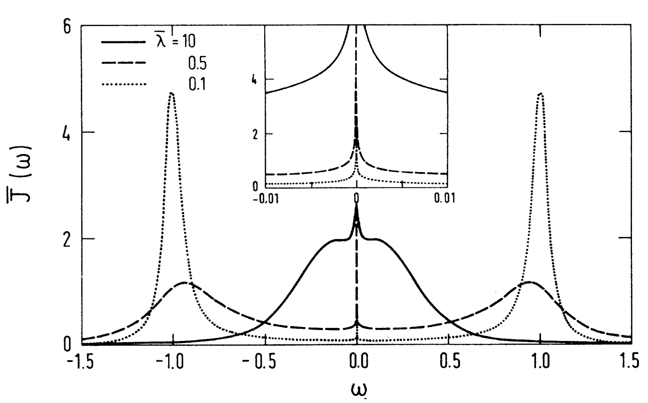


FIG. 2. Same as Fig. 1, but for fixed $\bar{\Omega} = 0.1$, $T/J = 1.0$, $\tilde{\Delta} = 0.35$, $X = 1.2$, and three values of $\bar{\lambda}$, as indicated. Inset: enlarged view of the central singularity.

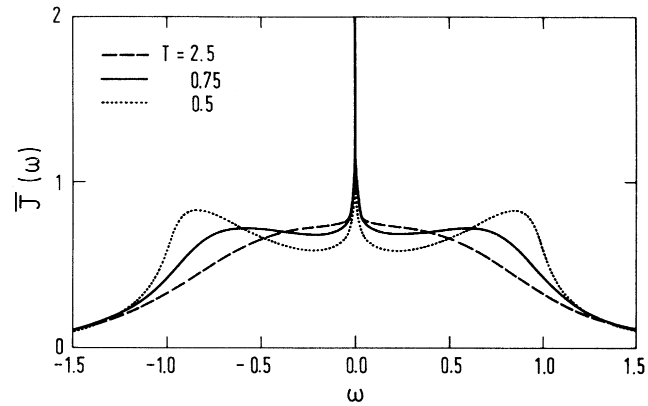


FIG. 3. Illustration of the temperature dependence of the line shape, evaluated at three different values of $T/J$, as indicated, and with $\bar{\Omega} = 0.1$, $\tilde{\Delta} = 0.35$, $\bar{\lambda} = 1.5$, and $X = 1.2$.

temperature $T/J$, random-field variance $\tilde{\Delta}$, and the relaxation rate $\bar{\lambda}$ all being fixed. As shown in Fig. 1(a) the central divergent component is not present in the classical case $\bar{\Omega} = 0$, but reappears at the smallest nonzero value of $\bar{\Omega}$. The two side peaks near $\omega/\Delta\omega = \pm 1$ broaden with increasing $\bar{\Omega}$ and the peak positions move closer to the center. The inset in Fig. 1(b) shows an enlarged view of the central singularity. In Fig. 2 the parameter $\bar{\lambda}$, which effectively measures the frequency of random fluctuations due to the heat bath on a frequency scale set by $\Delta\omega$, has been varied at a fixed value of $\bar{\Omega} = 0.1$. The case $\bar{\lambda} = 0.1$ corresponds to the slow-motion regime in which the quantum tunneling effects are dominant. On the other hand, for large values of $\bar{\lambda}$ one approaches the fast-motion limit $\bar{\lambda} \to \infty$ and the line shape tends to the static value $J(\omega) \to W(\omega)/\Delta\omega$. The same feature has been found for the stochastic model discussed in I, as well as in the case of Glauber dynamics of a classical spin glass.[23] Here the function $W(p)$ represents the static local polarization distribution as given by Eq. (33), which itself contains quantum effects.

Increasing the temperature leads to a gradual loss of the three-peak structure, as illustrated in Fig. 3. All the temperatures have been chosen such that the system is always well above the characteristic temperature $T_0 = J/4$, where in the classical limit the system would undergo a freezing transition into the nonergodic phase and the present approach would break down. From Eq. (38) one can see that the amplitude of the singular part of the line shape decreases with rising temperature and at the same time the frequency scale determined by the expression (39) expands. Thus for $T \to \infty$ the central peak progressively loses significance and, as expected, quantum effects cannot be observed.

## V. CONCLUSIONS

We have considered the dynamics of a quantum spin glass described by the transverse Ising model with random-bond random-field type of disorder, in which quantum effects are introduced through the transverse field representing the tunneling frequency $\Omega$ of, say, protons in a hydrogen-bonded proton glass. Specifically, we have studied the effects of the coupling between the quantum subsystem and the surrounding heat bath on the resonance line shape. We have introduced a type of coupling to the heat bath which merely assists thermally activated hopping of the proton in an O—H···O bond, leaving the quantum tunneling process untouched and hence fully coherent. This is to be contrasted with the stochastic model studied in I, where both hopping and tunneling were assumed to be directly influenced by the heat bath. The present type of coupling leads to a dynamic model, which thus describes an alternative approach to equilibrium in a quantum spin glass. As in I, the effective single-spin Hamiltonian has been obtained within the thermofield dynamic approach and a short-time approximation for the dynamic self-interaction, which are applicable to the ergodic spin-glass phase above the instability surface.[7] The spin-glass phase below the instability surface is characterized by a set of order parameters[24] and an effective single-spin Hamiltonian of the form (4) becomes inapplicable.

One of our main conclusions is that at low frequencies the line shape exhibits a singular part, which diverges as $\sim |\omega|^{-1/2}$. The corresponding functional form of this singularity has been obtained analytically. In physical terms it represents the contribution of coherent tunneling processes, which are not broadened by the heat-bath coupling, and is thus a genuine quantum effect. The singularity disappears in the classical case $\Omega \to 0$. The general result for the line shape has been obtained by a numerical integration of the remaining nonsingular part and is shown in Figs. 1–3. As in I, one can vary the relevant model parameters, in particular, the coupling strength $\bar{\lambda}$, and thus drive the system from the slow-motion regime into the fast-motion regime. In the former case, the spectrum is characterized by a three-peak structure, which in the fast-motion limit reduces to a single-peak one. In both cases, the central singularity is superimposed on the spectrum. In the high-temperature limit, the amplitude of the singularity as well as its width decrease with temperature, and thus quantum effects gradually disappear.

In real systems, both mechanisms of heat-bath coupling, i.e., the stochastic type studied in I and the coherent tunneling plus thermal hopping studied here, could be effective, depending on the temperature range. We expect that in general the present mechanism is dominant at low temperatures, whereas at high temperatures some incoherence in the tunneling process due to direct effects of the heat bath should be included. In a previous work[23] we offered some suggestions regarding the experimental verification of the predicted line shape in classical deuteron glasses such as D-RADP through EPR, NMR, and NQR techniques, which in general terms remain valid in the present case as well. Specifically, one should focus on the undeuterated proton glass RADP and search for the appearance of a sharp central line as the temperature is lowered. If the line shape could be fitted to an expression like our Eq. (39), this would amount to a verification of the coherent tunneling process. As a suitable experimental technique one could use high-resolution NMR of protons via chemical shift tensor effects or ENDOR (i.e., electron nuclear double resonance) spectroscopy.

In addition to the ferroelectric proton glasses, the recently studied ferromagnetic system $\mathrm{LiHo}_x\mathrm{Y}_{1-x}\mathrm{F}_4$ (Ref. 25) offers another possible realization of the transverse Ising spin glass, where the external magnetic field in the transverse direction causes mixing between the ground and excited states, thus inducing quantum effects similar to tunneling. It should be noted, however, that the model studied here is explicitly applicable to ferroelectric glasses only, since the random local field is allowed in ferroelectrics but not in ferromagnets. The appropriate modifications of the model in order to deal with the latter case could easily be made by including a static parallel magnetic field.

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## APPENDIX

Matrix elements of $[\hat{U}(i\omega)]_{\mathrm{av}}$:

$$\begin{aligned} U_{11} = &-\frac{\lambda}{2}(1+p)(\omega^2-h^2) - 2\lambda\omega(\omega+\Delta\omega/2) \\ &+i[(\omega+\Delta\omega/2)(h^2-\omega^2) \\ &\quad +\lambda^2\omega(1+p)+\omega\Omega^2/2], \end{aligned} \tag{A1}$$

$$U_{12} = \frac{\lambda}{2}(1-p)(h^2-\omega^2) + i\omega[\lambda^2(1-p)+\Omega^2/2], \tag{A2}$$

$$U_{13} = -\frac{\Omega}{2}\lambda p(\omega-h) - i\frac{\Omega}{2}(\omega+\Delta\omega/2)(\omega-h), \tag{A3}$$

$$U_{14} = \frac{\Omega}{2}\lambda p(\omega+h) + i\frac{\Omega}{2}(\omega+\Delta\omega), \tag{A4}$$

$$U_{21} = -\frac{\lambda}{2}(1+p)(\omega^2-h^2) + i\omega[\lambda^2(1+p)+\Omega^2/2], \tag{A5}$$

$$\begin{aligned} U_{22} = &-\frac{\lambda}{2}(1-p)(h^2-\omega^2) - 2\omega(\omega-\Delta\omega/2) \\ &+i[(\omega-\Delta\omega/2)(h^2-\omega^2) \\ &\quad +\lambda^2\omega(1-p)+\omega\Omega^2/2], \end{aligned} \tag{A6}$$

$$U_{23} = -\frac{\Omega}{2}\lambda p(\omega-h) + i\frac{\Omega}{2}(\omega-\Delta\omega/2)(\omega-h), \tag{A7}$$

$$U_{24} = \frac{\Omega}{2}\lambda p(\omega + h) - i\frac{\Omega}{2}(\omega - \Delta\omega/2)(\omega + h), \quad \text{(A8)}$$

$$U_{31} = i\frac{\Omega}{2}(\omega + \Delta\omega/2)(h - \omega), \quad \text{(A9)}$$

$$U_{32} = i\frac{\Omega}{2}(\omega - \Delta\omega/2)(\omega - h), \quad \text{(A10)}$$

$$\begin{aligned} U_{33} = & -\lambda\left[\omega^2 - (\Delta\omega)^2/4\right] + \lambda(p\Delta\omega/2 - \omega)(\omega - h) \\ & + i\{\lambda^2\omega - \left[\omega^2 - (\Delta\omega)^2/4\right](\omega - h) \\ & -\lambda^2 p\Delta\omega/2 + \omega\Omega^2/2\}, \end{aligned} \quad \text{(A11)}$$

$$\begin{aligned} U_{34} = & -\lambda\left[\omega^2 - (\Delta\omega)^2/4\right] \\ & -i\left[\lambda^2 p\Delta\omega/2 - \omega(\lambda^2 + \Omega^2/2)\right], \end{aligned} \quad \text{(A12)}$$

$$U_{41} = +i\frac{\Omega}{2}(\omega + \Delta\omega/2)(\omega + h), \quad \text{(A13)}$$

$$U_{42} = -\frac{\Omega}{2}(\omega - \Delta\omega/2)(\omega + h), \quad \text{(A14)}$$

$$\begin{aligned} U_{43} = & -\lambda\left[\omega^2 - (\Delta\omega)^2/4\right] \\ & -i\left[\lambda^2 p\Delta\omega/2 - \omega(\lambda^2 + \Omega^2/2)\right], \end{aligned} \quad \text{(A15)}$$

$$\begin{aligned} U_{44} = & -\lambda\left[\omega^2 - (\Delta\omega)^2/4\right] - \lambda(\omega - p\Delta\omega/2)(\omega + h) \\ & + i\{\lambda^2(\omega - p\Delta\omega/2) \\ & - \left[\omega^2 - (\Delta\omega)^2/4\right](\omega + h) + \omega\Omega^2/2\}. \end{aligned} \quad \text{(A16)}$$

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