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Abstract.   We study here the dynamic susceptibility of a model quantum glass. A statistical mechanical treatment, based on a mean-field analysis of the transverse Ising model, is able to account for the competition between disorder and frustration, on the one hand, and quantal fluctuations on the other. Our results derived for the dynamic susceptibility are in qualitative agreement with experimental measurements on a recently studied dipolar-coupled quantum glass, namely $LiHo_x Y_{1-x} F_4$, whose quantum term can be manipulated in the laboratory.

1. Introduction

Quantum glasses are a class of complex systems of great current interest in condensed matter physics. These materials exhibit various glass-like properties arising from quenched disorder and frustration. In addition, they also display quantum behaviour, particularly at low temperatures, for which tunnelling becomes important. There are two important questions relating to this class of problem:

(i) do quantum interactions preserve or destroy the glass transition and

(ii) what—if there are any—are the influences of the quantum transitions on the relaxational phenomena in these glassy systems?

Not much is known about this subject, as only a few systems have been subjected to careful experimental investigation. However, we are fortunate in that these few systems include the recently much-studied dipolar-coupled magnetic glasses [1] and proton glasses [2]. In this paper we will mainly focus our attention on the former, and present results on the theoretically calculated dynamic susceptibility, which can be compared directly with experiments.

Before we introduce these materials, we briefly discuss the 'classical' Ising glass or spin glass, by way of background to the subject. The classical spin-glass systems, such as AuFe or CuMn, are known to possess complicated free-energy landscapes involving deep valleys and randomly distributed barriers [3, 4]. As a result they have novel phase transition characteristics requiring the introduction of unusual order parameters. Equally intriguing are their time-dependent properties, captured by certain correlation functions which decay over time as 'stretched exponentials' of the so-called Kohlrausch–William–Watts form, with characteristic relaxation times that obey the Vogel–Fulcher relation [5]. Study of the thermodynamics of such systems has necessitated the invention of new statistical mechanical techniques such as the 'replica' method for evaluating the relevant partition function. In recent years much attention has been focused on truly non-equilibrium phenomena (not studied in this paper) in classical spin glasses, characterized by what is called 'aging',

which manifests itself through violation of stationarity [6, 7]. Quantum effects are further expected to enrich the observed phenomena, as different minima of the free energy, which are otherwise disjoint classically, can be linked by quantum tunnelling. Thus, quantum mechanics provides additional routes to relaxation which, taken together with the ubiquitous influence of disorder and frustration, can cause fascinating new effects worthy of detailed investigation, especially at low temperatures.

One quantum spin glass, which has been the subject of recent experimental studies, is the randomly diluted rare-earth material $LiHo_x Y_{1-x} F_4$, in which Ho has a magnetic moment whereas Y is non-magnetic [1]. When $x = 1$, one has a pure system in which the Ho moments interact via dipolar coupling, leading to ferromagnetic order at a Curie temperature $T_C = 1.53$ K. If Y is randomly substituted in place of Ho, the magnetic bonds are broken at random sites and, because the dipolar interaction depends on the inter-moment vector distances, the sign of the interaction randomly alternates between positive and negative. Thus we have what we may call a 'magnetic glass' in which there is random distribution of non-magnetic bonds as well as magnetic bonds of both ferromagnetic and antiferromagnetic types. In this respect, the system at hand is quite different from a metal-based spin glass (such as AuFe) in that the disorder in the interaction is not caused by oscillatory RKKY interaction between conduction electrons [8], but is due to random non-magnetic substitution of dipolar-coupled moments.

Although the rare-earth moments of Ho^{3+} in $LiHo_x Y_{1-x} F_4$ are dipolar coupled, because of the presence of a large crystal field only the lowest doublet of the 17 crystal-field-split states is appreciably populated at low temperatures ($T < 2$ K). Furthermore, the sign of the crystal field is negative and hence the lowest doublet corresponds to the highest spin states. Consequently, the off-diagonal terms of the dipolar interaction are effectively 'quenched' [9], yielding the so-called 'truncated dipolar Hamiltonian' [10]. The latter is of the Ising form, leading to an Ising interaction amongst spins along the crystallographic *c*-axis. In the experiment of Wu *et al*, a small magnetic field transverse to the *c*-axis is applied in the laboratory which causes admixture of the two eigenstates of the crystal-field-split doublet. Since this field couples to the component of the spin perpendicular to the *c*-axis, the transverse Ising model provides a valid description of the statistical mechanics of the system. We shall present here a theoretical treatment of the transverse Ising model that is able to account for this system in terms of phase changes as well as relaxational behaviour. Another similar system is a random admixture of hydrogen-bonded ferroelectric and antiferroelectric crystals—such as $Rb_{1-x}(NH_4)_x H_2 PO_4$ [2]. This system is called a proton glass as the basic random interaction is between protons, each of which can occupy two sites in an O–H...O bond. Quantum effects are important because the proton can tunnel from one site of the bond to the other. Our theoretical description can also be applied to proton glasses, with, however, an appropriately defined bond distribution. These results have been discussed in a separate paper [11].

Therefore, we see that the transverse Ising model with random bond strengths provides a valid theoretical framework for describing the physical properties of quantum glasses, such as magnetic and proton glasses. This model has been the subject of extensive theoretical activity in recent years. There have been a few studies, both analytical [12, 13] and numerical [14, 15], carried out to investigate the quantum phase transition at $T = 0$ between the paramagnetic and the spin-glass phase as the strength of the uniform transverse field is varied. These studies are focused on the scaling properties of the non-linear susceptibility near the 'quantum critical point'. However, the recent experiments [1] have not probed this regime. In this paper, we concentrate just on the frequency-dependent susceptibility of $LiHo_x Y_{1-x} F_4$ at temperatures above the glass transition temperature, which has been

experimentally measured, using a simple theoretical framework.

The outline of the paper is as follows. In section 2 we introduce the transverse Ising model and the distribution function of its interaction parameters which are applicable to magnetic glasses. The mean-field theory of this model is discussed in section 3. Section 4 is devoted to a discussion of relaxation phenomena in thermal equilibrium in terms of the dynamic susceptibility. The results and discussion are presented in section 5. Finally, in section 6, we present a few concluding remarks.

2. The Hamiltonian

The Hamiltonian for the transverse Ising model can be written as

$$H_s = - \sum_{i,j=1}^{N} J_{ij} \sigma_i^z \sigma_j^z - \Omega \sum_{i=1}^{N} \sigma_i^x \tag{1}$$

where J_{ij} is the interaction between randomly distributed holmium moments and Ω is proportional to the square of the field perpendicular to the c-axis applied in the laboratory [1]. In the absence of the transverse field, the Hamiltonian of equation (1) reduces to that of the Sherrington–Kirkpatrick (SK) model for the classical spin glass [16]. In equation (1), the interaction energy J_{ij} for magnetic glasses, recalling its origin in dipolar coupling, is given by [17, 10]

$$J_{ij} = g^2 \mu^2 (1 - 3\cos^2(\theta_{ij}))/r_{ij}^3 \tag{2}$$

where g is the gyromagnetic ratio, μ is the Bohr magneton, and r_{ij} and θ_{ij} are respectively the magnitude and the polar angle of the vector $(\bar{r}_i - \bar{r}_j)$ connecting sites i and j. Because the magnetic sites are randomly located, the angle θ_{ij} is also randomly distributed between 0 and π causing the sign of J_{ij} to fluctuate between positive and negative.

In addition to this effect of disorder and frustration, the last term in equation (1) induces tunnelling effects by mixing the eigenstates corresponding to the σ_z-operator. This introduces quantum dynamics in the system, making H_s the prototype Hamiltonian of a quantum glass. In the experiments of Wu *et al*, this is realized by the application of a magnetic field H_t perpendicular to the c-axis. This field can cause an admixture of the crystal-field-split states yielding in perturbation theory a spin Hamiltonian that depends on σ_x with a prefactor Ω which is quadratic in H_t^2. The transverse field Ω plays the role of a disordering field, like the temperature T. This is because as Ω increases, it causes an increasingly large amount of energy to be available to order the spins along the z-axis. Thus, even at zero temperature there is a critical value Ω_c such that for $\Omega > \Omega_c$ the system goes into a paramagnetic phase. This phenomenon is called a quantum phase transition because it happens due to purely quantal fluctuations. These phase transitions are fundamentally different from classical phase transitions. While unambiguous experimental study of these systems is quite difficult, LiHoF$_4$ provides a clean model magnet for which high-precision, controlled investigation of the quantum critical phenomenon can be carried out.

It is customary to assume that the distribution of J_{ij} is Gaussian:

$$P(J_{ij}) = \frac{1}{\sqrt{2\pi\Delta^2}} \exp\left(-\frac{(J_{ij} - J_0)^2}{2\Delta^2}\right). \tag{3}$$

In the experimentally studied compounds, we associate ferromagnetic and antiferromagnetic bonds with the coupling constants $+J$ and $-J$, and let $x/2$ be the concentration of these

bonds (assumed to be present in equal strength) and $1 - x$ be the concentration of non-magnetic bonds [18]. Equating the mean and the variance of the Gaussian distribution, equation (3), with those of the trimodal distribution of $(\pm J, 0)$ then requires

$$J_0 = 0 \qquad \Delta^2 = xJ^2. \tag{4}$$

Together with equation (3), equation (1) can describe the quantum system of the magnetic glass whose behaviour we are interested in. In an earlier paper, we presented results on the imaginary part of the susceptibility $\chi''(\omega)$, which however did not include the concentration dependence of J_0 and Δ^2 [19]. The present paper is a complete treatment, directly relatable to the measurements of Wu *et al* [1]. With the above background which gives us motivation for carrying out the mean-field analysis of the transverse Ising model, we discuss the mean-field theory for the model in the next section before turning our attention to dissipative phenomena. Indeed, as we will show, mean-field theory works remarkably well in qualitatively reproducing the data on dynamic susceptibility, especially above the glass transition temperature T_g (=0.1 K) of $LiHo_xY_{1-x}F_4$.

3. Mean-field theory

Classical spin glasses have been studied extensively using the replica method, which involves new ideas such as replica-symmetry breaking, the Edwards–Anderson order parameter and ergodicity. The use of this method in quantum problems is difficult due to the non-commutativity of operators appearing in the Hamiltonian, although some progress has been made in recent years [20–26]. A different treatment using a thermofield dynamics approach has been given by Kopec [27]. We refrain from repeating the rather formidable arguments of the thermofield dynamics method, and immediately rewrite the single-site Hamiltonian for the transverse Ising spin glass:

$$H_s = -h\sigma^z - \Omega\sigma^x. \tag{5}$$

Here h is an effective field acting along the z-axis due to the non-zero spin-glass order parameter q:

$$\begin{aligned} h(\xi) &= \tilde{\Delta}\xi\sqrt{q} \\ \tilde{\Delta} &= \Delta\sqrt{N} \end{aligned} \tag{6}$$

where ξ is the excess static noise arising from the random interactions J_{ij}. The mean-field equations for the local polarization and the Edwards–Anderson order parameter q are

$$p(\xi) = \frac{h(\xi)}{h_0(\xi)}\tanh(\beta h_0(\xi)) \tag{7}$$

and

$$q = \int_{-\infty}^{\infty} \frac{d\xi}{\sqrt{2\pi}} \exp(-\xi^2/2)[p(\xi)]^2 \tag{8}$$

with

$$h_0(\xi) = \sqrt{\Omega^2 + h^2(\xi)}. \tag{9}$$

The way to calculate any physically observable quantity is to compute the corresponding quantity in accordance with the statistical mechanical prescription for a given realization of the disorder, and then to perform an averaging over the underlying probability distribution of the disorder. In the mean-field approximation the disorder is represented entirely in terms of the local polarization p (cf. equation (7)). Thus all physical quantities are to be first

calculated for a given value of p and the results averaged over the distribution of p, defined by $W(p)$. The latter is defined as

$$W(p) = \frac{1}{N} \sum_i \delta(p - \langle \sigma_i^z \rangle) = \int D\xi \; \delta(p - p(\xi)) \tag{10}$$

which for the case where $\sigma_z = \pm 1$ yields

$$W(p) = \frac{1}{\beta J \sqrt{2\pi q}} \exp\left(-\frac{1}{2}\xi_0^2(p)\right) \left[\frac{h^2(\xi_0)}{h_0^2(\xi_0)} + \frac{\Omega^2}{\beta h(\xi_0)h_0^2(\xi_0)} p - p^2\right]^{-1} \tag{11}$$

where the function $\xi_0 = \xi_0(p)$ is the inverse of $p(\xi)$, which satisfies equation (7) for p in the interval $[-1, +1]$. It is evident that for $\Omega = 0$, equations (5)–(9) reduce to the corresponding equations for the SK model for the region of phase space above the Almeida–Thouless surface, where replica symmetry is assumed [4]. Again, in the limit where $\Omega = 0$, $W(p)$ reduces to the corresponding expression for the pure Ising model [4].

It is interesting to note that equations (7)–(11) can also be obtained from a generalization of the heuristic argument of Southern [28] for the pure Ising/SK model to the disordered transverse Ising model. As has been discussed by Fischer and Hertz (chapter 3 of reference [4]), this argument is incorrect. Yet, and quite remarkably, the results survive the close theoretical scrutiny of the replica-symmetric theory [16] or the corresponding dynamic formulation of the SK model [29–31]. The fact is that the results obtained in the crude mean-field theory are correct (even though their derivation is incorrect!) above the Almeida–Thouless line. Below that line, the physics is rather complicated, as the assumption of replica symmetry breaks down, and analytical treatments are difficult. We expect to encounter similar and more intractable obstacles in the quantum glass problem due to the presence of the additional transverse term in the SK model. Again, within the TFD approach [27], one finds that below an Almeida–Thouless-like surface there is no stable quantum glass solution, whereas above that surface one arrives at the mean-field solutions given by equation (7) and equation (8). We shall assume here the approximate validity of these solutions near the quantum glass temperature T_g. The heuristic argument of Southern for the pure Ising/SK model estimates T_g to be equal to Δ, the square root of the variance of the interactions J_{ij}. In the quantum glass problem, the transverse field Ω reduces T_g until finally, at a critical value Ω_c, $T_g = 0$. One can try to improve the results by taking into account the Onsager terms as in the Thouless–Anderson–Palmer equations [32], but these equations are known to reduce to the SK equations above the instability surface. Since we are interested in comparing our analytical results with those obtained by Wu *et al* at temperatures just above the glass transition temperature, equation (7) and equation (8) are relevant and sufficient for our simple mean-field calculation.

The Hamiltonian of equation (5) describes the reversible quantum dynamics of the system. This is in itself interesting, and indeed has been the subject of attention in investigations using the Ising model for a transverse field [35]. However, our aim is to study the dynamic susceptibility response which is related to the power absorbed by the system. Hence, it is necessary to introduce irreversible effects leading to a dissipative irreversible dynamics of the system. In order to include a dissipative dynamics, we have to expand the scope of the Hamiltonian in equation (5) by including the coupling to the surrounding heat bath:

$$H_0 = H_S + H_I + H_B \tag{12}$$

where H_I describes the interaction between the spin system and the heat bath.

The question is, what form of the interaction Hamiltonian should be chosen H_I? In order to answer this question, it is useful to first diagonalize H_S (in equation (5)) by rotating the coordinate system about the y-axis in a clockwise direction by an angle $\theta = \arctan(\Omega/h)$. The corresponding rotation operator is

$$U_Y^R = \exp(-i\theta\sigma^y). \tag{13}$$

Note that in the rotated frame, the total Hamiltonian is

$$\tilde{H}_0 = \tilde{H}_S + \tilde{H}_I + H_B \tag{14}$$

where the subsystem Hamiltonian in the rotated frame is given by

$$\tilde{H}_S = h_0\sigma^z \tag{15}$$

where h_0 is given by equation (9). It is now easy to deduce the form of the interaction $\tilde{H}_I$ in the rotated frame, as long as we restrict consideration to Glauber-like relaxational dynamics. As has been shown by Heims [36] and Kawasaki [37], the corresponding form is

$$\tilde{H}_I = gb\sigma^x \tag{16}$$

where b is a bath operator (describing phonons) which acts on the Hilbert space of the heat-bath Hamiltonian H_B and g is a multiplicative coupling constant. The operator σ^x, being entirely off-diagonal in the representation in which σ^z (in equation (15)) is diagonal, causes heat-bath-induced flips, *à la* Glauber. We are now ready with the answer to the question posed at the beginning of this paragraph. 'Unrotating' equation (16), we find, in the laboratory frame,

$$H_I = \frac{gb}{h_0}[h\sigma^x - \Omega\sigma^z]. \tag{17}$$

It is evident that when tunnelling is absent (i.e. $\Omega = 0$), H_S reduces to the Ising Hamiltonian in the mean-field approximation (cf. equation (1)), and the coupling with the heat bath assumes the form of equation (16). In that limit, we recover the Glauber kinetics. The full interaction term in equation (17), however, describes two entities:

(i) spin flips of the Glauber type via σ^x and
(ii) incoherent tunnelling via σ^z.

This completes the discussion on the motivation behind choosing the form given in equation (17).

4. Frequency-dependent susceptibility and the correlation function

We now turn our attention to the central issue of the present work, namely the computation of the dynamic susceptibility $\chi(\omega)$. This, in linear response theory, adapted to single-site mean-field approximation, is given by [34]

$$\chi(\omega) = \beta \lim_{\delta \to 0, \, s \to -i\omega+\delta}[1 - s\tilde{C}(s)] \tag{18}$$

where $\tilde{C}(s)$ is the Laplace transform of the correlation function defined as

$$C(t) = \langle\sigma^z(0)\sigma^z(t)\rangle_{eq}. \tag{19}$$

Here the angular brackets denote the appropriate quantum and statistical averaging. The quantity s is related to the applied frequency ω, δ is a small real-valued parameter included

to ensure convergence of the Laplace transforms and β is the inverse temperature. $C(t)$ can be explicitly written for the equilibrium ensemble as

$$C(t) = \text{Tr}\left(\rho_{eq}\sigma^z(0)e^{iH_0t}\sigma^z(0)e^{-iH_0t}\right) \tag{20}$$

where H_0 is the total Hamiltonian as in equation (12) and Z_0 is the corresponding partition function. In the rotated frame the correlation function reads

$$C(t) = \text{Tr}\left(\tilde{\rho}_{eq}\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right]e^{i\tilde{H}_0t}\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right]e^{-i\tilde{H}_0t}\right) \tag{21}$$

where $\tilde{H}_0$ is the total Hamiltonian in the rotated frame, as given by equation (14). Assuming that the subsystem is weakly coupled to the heat bath, we can factorize the density matrix and write the correlation function as

$$C(t) = \frac{1}{Z_s}\text{Tr}\left(e^{-\beta\tilde{H}_s}\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right]\text{Tr}\left[\frac{e^{-\beta H_B}}{Z_B}\left[U(t)\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right]\right]\right]\right) \tag{22}$$

where $U(t)$ is the time-development operator. The Laplace transform of $C(t)$ reads

$$\tilde{C}(s) = \frac{1}{Z_s}\text{Tr}\left(e^{-\beta\tilde{H}_s}\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right][\tilde{U}(s)]_{av}\left[\frac{h}{h_0}\sigma^z - \frac{\Omega}{h_0}\sigma^x\right]\right). \tag{23}$$

In equation (23), $[\tilde{U}(s)]_{av}$ denotes the Laplace transform of the time-development operator of the system. As discussed extensively in reference [34], it is the physics of a given problem that decides the nature of the time-development operator. In the present context, we have adopted a system-plus-reservoir approach in order to give a proper treatment of the dissipative interaction term and systematically 'project out' the bath degrees of freedom. This can be most conveniently achieved by writing a resolvent expansion of $[\tilde{U}(s)]_{av}$ in which the interaction term H_I is treated perturbatively. Such an expansion yields the following general expression for $[\tilde{U}(s)]_{av}$ [34]:

$$[\tilde{U}(s)]_{av} = [s - iL_s + \tilde{\Sigma}(s)]^{-1} \tag{24}$$

where L_s is the Liouville operator associated with the spin Hamiltonian H_s, defined in equation (5), and $\tilde{\Sigma}(s)$ is the so-called relaxation matrix, to be specified below. While it is possible to evaluate $\tilde{\Sigma}(s)$ to arbitrary orders in perturbation theory, it suffices for the purpose of obtaining Markovian dynamics to use an expansion up to second order in H_I, which yields

$$\tilde{\Sigma}(s) = \left[L_I\frac{1}{s - iL_s - iL_B}L_I\right]_{av}. \tag{25}$$

Writing out the trace over the subsystem, we obtain

$$\tilde{C}(s) = \frac{1}{Z_s}\frac{1}{h_0^2}\sum_{\mu,\nu,\mu',\nu'}e^{\beta h_0\mu}\langle\mu|h\sigma^z - \Omega\sigma^x|\nu\rangle(\nu\mu|[\tilde{U}(s)]_{av}|\nu'\mu')\langle\nu'|h\sigma^z - \Omega\sigma^x|\mu'\rangle. \tag{26}$$

In writing this equation, we have used the properties of the Liouville operators (refer to chapter 1 of reference [34]) as well as the notation

$$(\mu\nu|L|\mu'\nu') = \delta_{\nu\nu'}\langle\mu|H|\mu'\rangle - \delta_{\mu\mu'}\langle\nu'|H|\nu\rangle \tag{27}$$

where L is a Liouville operator corresponding to the Hamiltonian H. In order to make further progress, we need to compute the matrix elements of $[U(s)]_{av}$. The latter, being a super-operator in the space of the subsystem alone, is characterized by 16 elements, because the subsystem is itself restricted to a two-dimensional Hilbert space in the present case. Using equation (27), the matrix elements of L_S can be written as

$$(\nu\mu|L_S|\nu'\mu') = h_0(\mu - \nu)\delta_{\nu\nu'}\delta_{\mu\mu'}. \tag{28}$$

Explicitly

$$L_S = \begin{pmatrix} 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 \\ 0 & 0 & -2h_0 & 0 \\ 0 & 0 & 0 & 2h_0 \end{pmatrix} \tag{29}$$

where the rows and columns labelled by $|\nu\mu\rangle$ take the values $|++\rangle$, $|--\rangle$, $|+-\rangle$ and $|-+\rangle$ respectively.

The next step is the evaluation of the relaxation matrix. Treating the heat bath in the Markovian approximation, it is possible to approximate the relaxation matrix as follows:

$$\tilde{\Sigma}(s) \approx \tilde{\Sigma}(0) = \int_0^\infty dt \; L_I(\exp[i(L_S + L_B)t])L_I. \tag{30}$$

A typical sample, as evaluated from equation (30), is of the form

$$W_{++} = (++|\tilde{\Sigma}(0)|++) = g^2 \int_{-\infty}^{+\infty} dt \; [e^{+ih_0t}\langle b(0)b(t)\rangle + e^{-ih_0t}\langle b(t)b(0)\rangle] \tag{31}$$

where the bath correlations are defined as

$$\langle b(t)b(0)\rangle \equiv \text{Tr}[\rho_B e^{iH_Bt} b(0) e^{-iH_Bt} b(0)] \tag{32}$$

where ρ_B is the density operator of the heat bath. Furthermore, the quantities W_{+-} and W_{-+} are related as follows:

$$W_{+-} = e^{2\beta h_0} W_{-+}. \tag{33}$$

Equation (33) expresses the detailed balance of the transitions, as it can be rewritten as

$$p_-^{eq} W_{+-} = p_+^{eq} W_{-+} \tag{34}$$

where $p_\pm^{eq}$ denote the equilibrium probabilities:

$$p_\pm^{eq} = \frac{e^{\pm\beta h_0}}{e^{+\beta h_0} + e^{-\beta h_0}}. \tag{35}$$

Having fixed the detailed-balance condition, we now turn our attention to evaluating the bath correlation functions. All elements of $\tilde{\Sigma}(0)$ can be expressed in terms of certain bath correlation functions [33]. These correlations are not evaluated here explicitly but are simply parametrized in terms of a phenomenological relaxation rate λ by first making use of the following Kubo relation:

$$\int_{-\infty}^{+\infty} dt \; e^{+ih_0t}\langle b(t)b(0)\rangle = e^{2\beta h_0} \int_{-\infty}^{+\infty} dt \; e^{-ih_0t}\langle b(t)b(0)\rangle. \tag{36}$$

Using equation (36), one can now define λ as

$$\int_{-\infty}^{+\infty} dt \; e^{\pm ih_0t}\langle b(t)b(0)\rangle = \lambda \frac{e^{\pm\beta h_0}}{e^{+\beta h_0} + e^{-\beta h_0}} \tag{37}$$

where

$$\lambda \equiv \int_{-\infty}^{+\infty} dt \; (e^{+ih_0t} + e^{-ih_0t})\langle b(t)b(0)\rangle = \int_{-\infty}^{+\infty} dt \; e^{+ih_0t}(\langle b(t)b(0)\rangle + \langle b(0)b(t)\rangle). \tag{38}$$

We assume that the fluctuations in the heat bath are characterized by frequencies which are much larger than the frequency associated with h_0, i.e. the fluctuations in the bath

have a very short lifetime. Under this Markovian assumption, λ becomes real and can be approximated by the following expression:

$$\lambda \approx \int_{-\infty}^{+\infty} \mathrm{d}t \ (\langle b(t)b(0)\rangle + \langle b(0)b(t)\rangle). \tag{39}$$

In order to obtain the matrix $[U(s)]_{av}$, we have to invert the matrix M, which is given by equation (24) as

$$M = \begin{pmatrix} s + \lambda p_-^{eq} & -\lambda p_-^{eq} & 0 & 0 \\ -\lambda p_+^{eq} & s + \lambda p_+^{eq} & 0 & 0 \\ 0 & 0 & s + \dfrac{\lambda}{2} + 2ih_0 & -\dfrac{\lambda}{2} \\ 0 & 0 & -\dfrac{\lambda}{2} & s + \dfrac{\lambda}{2} - 2ih_0 \end{pmatrix} \tag{40}$$

where $p^{eq} = p_+^{eq} - p_-^{eq}$ is the net polarization in the rotated frame, $p_\pm^{eq}$ are given by equation (35) and λ is as given by equation (39). The matrix M is easily invertible and yields the following bath-averaged time-development operator $[U(s)]_{av}$ for the system that we have considered:

$$[U(s)]_{av} = \begin{pmatrix} \dfrac{1}{s(s+\lambda)}\begin{pmatrix} s + \lambda p_+^{eq} & \lambda p_-^{eq} \\ \lambda p_+^{eq} & s + \lambda p_-^{eq} \end{pmatrix} & \begin{matrix} 0 & 0 \\ 0 & 0 \end{matrix} \\ \begin{matrix} 0 & 0 \\ 0 & 0 \end{matrix} & \dfrac{1}{s(s+\lambda) + 4h_0^2}\begin{pmatrix} s + \dfrac{\lambda}{2} - 2ih_0 & \dfrac{\lambda}{2} \\ \dfrac{\lambda}{2} & s + \dfrac{\lambda}{2} + 2ih_0 \end{pmatrix} \end{pmatrix}. \tag{41}$$

We are now ready to insert equation (41) in equation (26). It is clear from the structure of equation (41) that the cross terms involving the product of h and Ω vanish, yielding

$$\tilde{C}(s) = \frac{1}{s(s+\lambda)}\left(s\frac{h^2}{h_0^2} + \lambda p^2\right) + \frac{\Omega^2/h_0^2}{s(s+\lambda) + 4h_0^2}\left[(s+\lambda) + 2ip\frac{h_0^2}{h}\right] \tag{42}$$

where p, the local polarization, is given by equation (7) and

$$p_+^{eq} - p_-^{eq} = \frac{h_0}{h}p. \tag{43}$$

With the correlation function at hand, the dynamic susceptibility for a given realization of the polarization is found, using equation (18), to be

$$\chi(\omega, p) = \beta\left[1 - \frac{(-i\omega h^2/h_0^2 + \lambda p^2)}{(i\omega + \lambda)} - i\omega\frac{(\Omega^2/h_0^2)[\lambda - i(\omega - 2ph_0^2/h)]}{(\omega^2 - 4h_0^2) + i\omega\lambda}\right]. \tag{44}$$

The measured susceptibility is obtained by averaging equation (44) over the polarization distribution function $W(p)$ given by equation (11):

$$\chi(\omega) = \frac{1}{4\pi}\int_{-1}^{1} \mathrm{d}p \ W(p)\chi(\omega, p). \tag{45}$$

In particular, the absorptive susceptibility obtained from the imaginary part of equation (44) for a given configuration of local polarization p is given by

$$\chi''(\omega, p) = \beta\left[\frac{\omega\lambda}{\lambda^2 + \omega^2}\left[\frac{h^2}{h_0^2} - p^2\right] + \frac{4\omega\lambda\Omega^2}{(4h_0^2 - \omega^2)^2 + \omega^2\lambda^2}\left[1 - \frac{\omega p}{2h}\right]\right]. \tag{46}$$
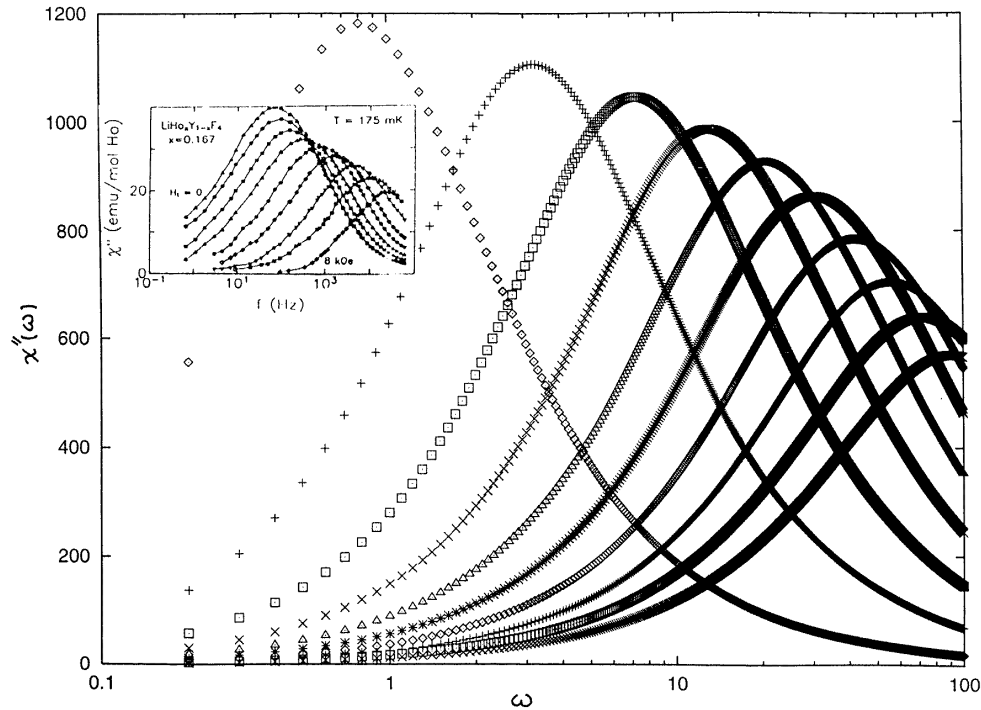
This equation is the main focus of our paper.

Figure 1. The imaginary part of the dynamic susceptibility, $\chi''(\omega)$, as a function of ω. The curves, in descending order of amplitude, correspond to different values of Ω/J, namely 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.8, 0.9 and 1.0. The temperature T and the relaxation rate λ are chosen to be 1.1 and 50, both in units of J. The inset shows the experimental data for comparison (see reference [1]).

5. Results and discussion

The result of numerical evaluation of the imaginary component of the dynamic susceptibility, $\chi''(\omega)$, averaged over the polarization is presented in figure 1 for the experimentally relevant value of x, namely $x = 0.167$ in $LiHo_x Y_{1-x} F_4$ as measured by Wu *et al*. The results were obtained at a temperature such that $T/J = 1.1$. As discussed in section 3, with this choice of T/J we have ensured that the temperature selected is just above the glass transition temperature. The experimental measurements of Wu *et al* are in this regime. The relaxation rate λ/J has been chosen to be 50. The curves, in descending order of their amplitude, correspond to different values of Ω/J ranging from 0.1 (the largest-amplitude curve) to 1.0 (the smallest-amplitude curve). The dynamic susceptibility response around the peak value indicates a shift in the frequency as Ω/J is increased. In addition to the reduction in amplitude, there is a clear indication of broadening of the peaks as quantum effects are enhanced (figure 1). These effects can be associated with a broadening of the energy eigen-levels of σ^z due to the presence of the transverse field. It can also be observed that the low-frequency tails of $\chi''(\omega)$ are greatly suppressed, implying that quantum routes to relaxation affect the long-time dynamics of the system. The data exhibit qualitative similarities with the experimental data (shown in the inset of figure 1) obtained by Wu *et al* as regards the features mentioned above.
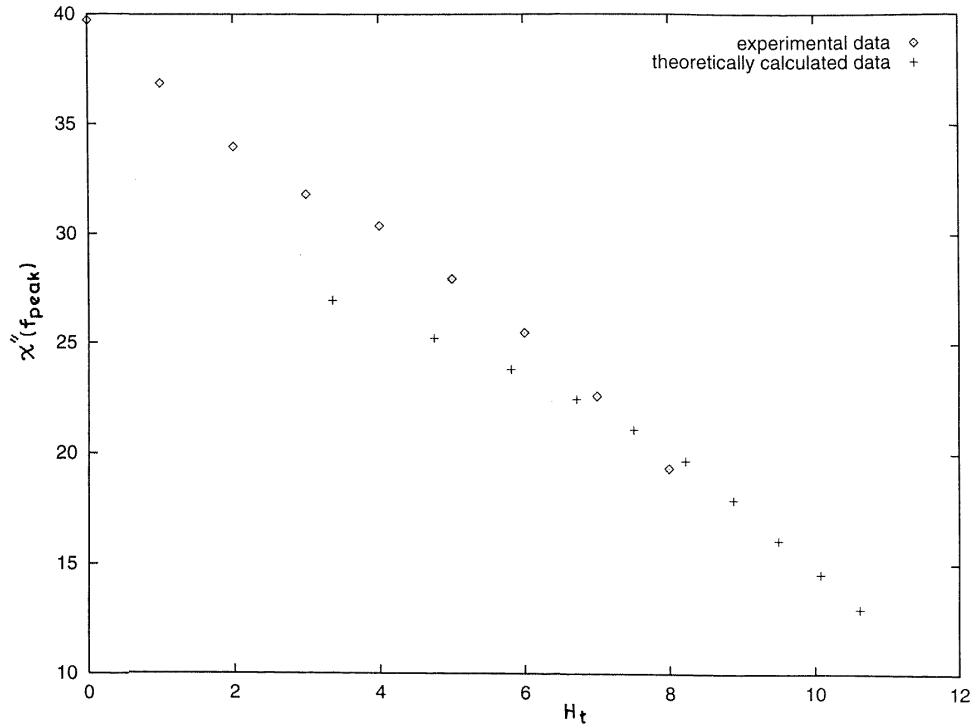
Figure 2. Comparison of the peak value of the imaginary part of the susceptibility as a function of the transverse field for the experimental data (read out from reference [1]) and our theoretically computed data.

It can be seen from figure 1 that the non-Debye response which is so characteristic of glassy systems is driven towards a symmetric Debye response of $\chi''(\omega)$ as quantum effects are enhanced. In this sense, the effect of the quantum tunnelling term is very similar to that of the temperature. We find that a large value of T/J results in the above situation for much smaller ratios of Ω/J. We also find that the quantum characteristics of the shift in the peak frequency and broadening of the response for a fixed value of Ω/J are less conspicuous for larger values of T/J. In order to further facilitate comparison of our theoretically obtained data with the experimental data of Wu *et al*, we plot the peak values of the susceptibility versus the applied transverse field for the two sets of data (figure 2). Both of these sets of data can be fitted to straight-line equations. The equation of the best-fit line governing the experimental data is

$$\chi''(f_{peak}) = -2.5151H_t + 40.1174$$

while that governing the theoretically obtained data is

$$\chi''(\omega_{peak}) = -1159.294\sqrt{\Omega/J} + 1741.134\,64.$$

(Recall that $H_t = \Omega^2$ [1].) The task now is to match the experimental parameters with those in our model. In order to determine the relation between $\chi''(f_{peak})$ and $\chi''(\omega_{peak})$ and H_t and $\sqrt{\Omega/J}$, we match the intercepts on the axes of the best-fit lines corresponding to both sets of data. In this way we try to obtain scale factors for the x- and y-coordinates for the theoretical data. By means of this exercise, we have ensured that the two sets of data

lie in the same parameter range. For low values of the applied field we could not observe perceptible quantum effects in our model calculation, but it was possible to measure the effects of transverse fields stronger than those considered in the experiment. From figure 2, it is clear that the agreement between the experimental data (squares) and the theoretical data (plus signs) improves as the values of the applied field H_t become larger.
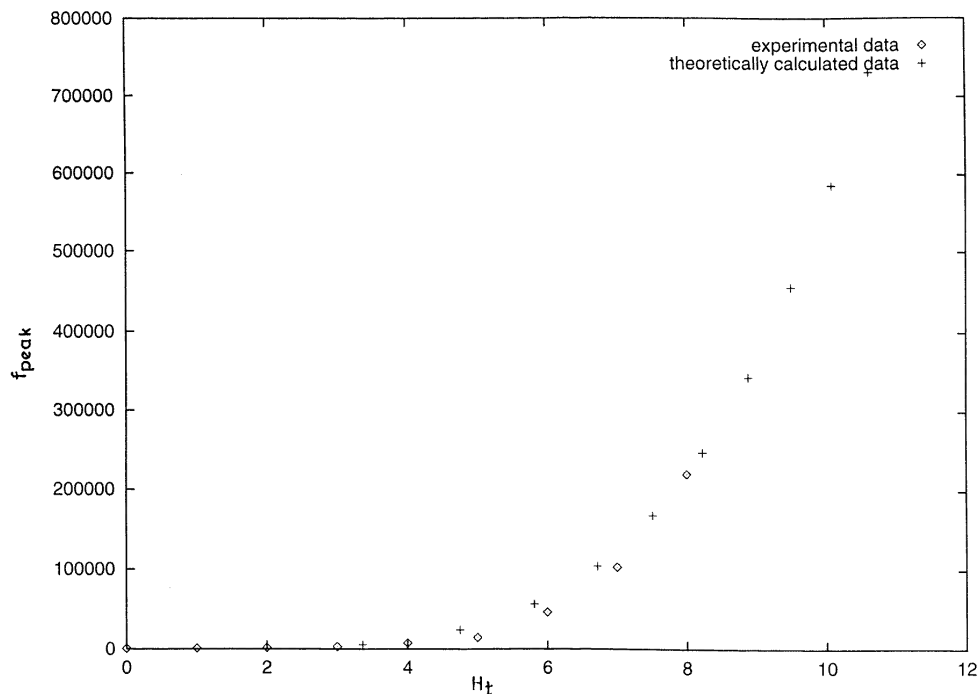

Figure 3. Comparison of the frequency corresponding to the peak susceptibility response as a function of the transverse field for the experimental data and our theoretically computed data.

We have also compared f_{peak} and ω_{peak} as functions of the applied transverse field in figure 3. For non-zero values of the applied field, the experimental data fit to a power law

$$f_{peak} = 2.2595 H_t^{5.523\,65}.$$

Our theoretical data also fit to a power law governed by the equation

$$\omega_{peak} = 90.0221 \sqrt{\Omega/J}^{\,4.237\,82}.$$

From figure 2, we find that the experimental and theoretical values of $\chi''(f_{peak})$ agree best for $H_t = 8$. Hence, setting $f_{peak}(H_t = 8)$ and $\omega_{peak}(H_t = 8)$, we obtain a scale factor for our set of theoretical data to reduce it to fit within the range of the experimental data. Figure 3 shows the experimental and the scaled theoretical data. A good agreement between these data is clearly brought out in this graph.

The relaxation parameter λ effectively measures the frequency of random fluctuations due to the heat bath on a scale that is set by the strength of the disorder J. The case for which $\lambda = 1.0$ corresponds to a slow-motion regime in which the quantum tunnelling effects seem important. We have observed a double-peaked structure in our numerical evaluation of $\chi''(\omega)$ for values of λ in this range. Such a situation has not been observed experimentally and it may be of interest to investigate the experimental regime in which

a double-peaked structure of $\chi''(\omega)$ is observed. We have checked that the results are not reproducible when λ is small; hence the heat-bath coupling of the kind assumed by us is critical in bringing out the essential features of the quantum and relaxational dynamics for $LiHo_x Y_{1-x} F_4$, which is known to exhibit spin-glass properties at $x = 0.167$ for low temperatures. We have also checked for these effects in a ferromagnet in a transverse field, in the absence of disorder. The computation of $\chi''(\omega)$ for different values of the transverse field yields results which are qualitatively different from those observed in the experiments of Wu *et al*. These observations indicate that disorder and quantum effects are crucial in bringing out the experimentally observed features.

6. Conclusions

Analytical techniques for studying dynamical properties at non-zero temperatures are few. Of late, numerical techniques such as the quantum Monte Carlo method are being used to explain the static properties of the transverse Ising model. The study of dynamical aspects using the quantum Monte Carlo method is difficult since it is necessary to analytically continue imaginary-time correlations to real-time correlations. Therefore, we have used a simple mean-field approach to examine the dynamic relaxation of a quantum system in the presence of disorder. It is able to account for the competition between disorder and frustration, on the one hand, and quantal fluctuations on the other, in a dipolar-coupled rare-earth magnet whose quantum term can be manipulated in the laboratory. There is good agreement between our results and the experimental results of Wu *et al* above the glass transition temperature.

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