Nonlinear Response in Quantum Spin Glasses

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We examine the behavior of the dynamic nonlinear response of a quantum spin glass in an exactly solvable fully connected quantum spherical model with random Gaussian bond distribution. In the quantum critical regime where the microscopic energy scale is set entirely by temperature the nonlinear response was found frequency independent and nonsingular. On the contrary, the genuine static nonlinear susceptibility diverges everywhere on the critical boundary with unusual violation of the universal scaling by the double logarithms at the zero-temperature critical point. Implications for experiments on quantum dipolar spin glasses are also noted. [S0031-9007(97)04608-5]

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The study of quantum phase transitions is the central theme of current research in condensed matter and statistical physics (see, e.g., Ref. [1] for an account of recent developments). These transitions, governed by quantum fluctuations, appear in the vicinity of zero temperature when an external parameter is varied. In the presence of disorder, as in quantum spin glass (SG), one encounters new features that are usually absent in pure system. Quantum fluctuations may permit the system to pass from one local minimum of the free energy to another via tunneling through barrier (as opposed to the conventional thermally driven activation process) at rates that do not vanish as temperature T approaches zero. The canonical example of a quantum SG is the quantum Sherrington-Kirkpatrick (SK) model [2] in a transverse field [3]. Here, quantum criticality gives rise to a number of novel physical effects such as introduction of quantum channels for relaxation, continuously depressing the phase transition temperature down to $T = 0$ which recently became accessible experimentally. Examples include the dipolar Ising magnet $LiHo_xY_{1-x}F_4$ in a transverse field [4] and the so-called proton glasses [5] being the mixtures of ferroelectric and antiferroelectric hydrogen bonded materials such as $Rb_{1-x}(NH_4)_x(H_2P)_4$ [6].

An essential ingredient in understanding quantum disordered systems in general (and quantum SG in particular) is the determination of their critical properties especially near $T = 0$. Typically, the conventional linear response, i.e., ac susceptibility χ does not diverge at the freezing temperature T_c but merely exhibits a cusp. The *nonlinear* response $\chi_{\rm n1}$, however, being the higher order correlation function should have a critical singularity at T_c [7,8] making it an indispensable physical quantity accessible in experiments which probes the SG phase transition. Usually, at the classical SG transition a scaling behavior $\chi_{\rm n1} \sim (T - T_c)^{-\gamma}$ was found, with γ ranging from 0.9 to 3.8 [9]. Strikingly, this behavior is qualitatively different in experimental measurements of the nonlinear susceptibility of the disordered dipolar *quantum* Ising magnet $LiHo_xY_{1-x}F_4$ as the critical boundary is approached [10]: the sharp divergence measured in the classical limit (i.e., for small transverse field) becomes suppressed and effectively disappears as $T \rightarrow 0$, raising the question of whether a well-defined SG transition still occurs.

On the theoretical side, there are only a few studies on the nonlinear response in quantum spin glasses: perturbation expansions for the SK model in a transverse field Γ yield near the critical point $\Gamma_c(T = 0)$ the nonlinear susceptibility in the scaling form $\chi_{\rm n1} \sim (\Gamma - \Gamma_c)^{-\gamma}$ with an estimate for γ between 0.29 and 0.75 [11]; Landau-type theory results in $\gamma = 1$ [12], whereas a real-space renormalization-group analysis gives the value $\gamma = 1.16$ [13]. Monte Carlo simulations for transverse Ising SG models in two [14] and three [15] dimensions signal even stronger divergence of $\chi_{\rm n1}$ at the $T = 0$ transition point in clear contradiction with experimental findings. Notably, none of these works explored the *dynamic* properties of the nonlinear response close to the $T = 0$ critical point. This, however, appears to be of paramount importance while discussing the quantum SG transition scenario since in actual experiments the peculiar behavior of $\chi_{\rm n1}$ was probed at a *finite* frequency ω [10].

In this Letter we argue that the anomalous behavior of the nonlinear susceptibility appears to be a tantamount of a finite temperature paramagnetic-SG transition near the onset of a $T = 0$ paramagnetic phase. In this so-called quantum critical (QC) regime [16] where the microscopic energy scale μ is set entirely by temperature [and $\mu(T) \rightarrow 0$ as $T \rightarrow 0$] the nonlinear response was found ω independent and nonsingular over the frequency range $\mu(T) \leq \omega \leq \Lambda_\omega$ where $\Lambda_\omega \sim \Gamma$ is the upper energy cutoff. On the contrary, the genuine *static* nonlinear susceptibility $\chi_{\rm n1}(\omega = 0)$ diverges everywhere on the critical boundary with unusual violation of the universal scaling by the double logarithms at the $T = 0$ critical point. We present exact calculations of the frequency dependent nonlinear response; this will be established for a model system—a solvable quantum spherical SG model with Gaussian distributed, infinite-ranged two-spin interactions [17]. Our discussion will be in the context of the following quantum SG Hamiltonian:

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$$H = \frac{\Delta}{2}\sum_i \Pi_i^2 - \sum_{i<j} J_{ij}\sigma_i\sigma_j - h\sum_i \sigma_i\,, \qquad (1)$$

where the variables σ_i ($i = 1,\ldots,N$) are associated with spin degrees of freedom and canonically conjugated to the "momentum" operators Π_i such that $[\sigma_i, \Pi_j] = i\delta_{ij}$. The coupling Δ regulates the strength of quantum fluctuations ($\Delta \to 0$ corresponds to the classical limit), and h is a longitudinal field used to define various susceptibilities but usually set to zero. Furthermore, the J_{ij}, connecting all sites, are assumed to be statistically independent between different links with first $\langle J_{ij}\rangle_{\rm av} = 0$ and second moment $\langle J_{ij}^2\rangle_{\rm av} = J^2/N$, where $\langle\cdots\rangle_{\rm av}$ denotes the random average. Finally, Eq. (1) is supplemented by the mean spherical constraint

$$\left\langle\left\langle \frac{1}{N}\sum_{i=1}^N \sigma_i^2\right\rangle_T\right\rangle_{\rm av} = 1\,, \qquad (2)$$

with $\langle\cdots\rangle_T$ being the ensemble average. Note that the choice of Eq. (1) is not incidental here—the model it describes is just a spherical version of the transverse Ising SG where the stronger condition $(\sigma_i^z)^2 = 1$ is replaced by the sum rule (2) and the parameter Δ straightforwardly translates into the transverse field Γ [18].

The evaluation of the statistical properties is achieved by expressing the partition function $Z = \mathrm{Tr}\, e^{-H/k_BT}$ in terms of the functional integral in the Matsubara "imaginary time" τ ($0 \le \tau \le 1/k_BT \equiv \beta$). We obtain $Z = \int D\Xi$ and $\langle\cdots\rangle = Z^{-1}\int D\Xi\ldots$ where

$$\int D\Xi\ldots \equiv \int \prod_i [D\sigma_i(\tau)] \times \delta\left(\sum_{i=1}^N \sigma_i^2(\tau) - N\right) e^{-\int_0^\beta d\tau\, S_\sigma(\tau)}\ldots \qquad (3)$$

with the Euclidean action of the form

$$S_\sigma(\tau) = \frac{1}{2\Delta}\sum_i \left(\frac{\partial\sigma_i}{\partial\tau}\right)^2 - \sum_{i<j} J_{ij}\sigma_i(\tau)\sigma_j(\tau)\,. \qquad (4)$$

To assure the spherical constraint the functional analog of the Dirac δ-function representation $\delta(x) = \int_{-\infty}^{+\infty}(dv/2\pi)e^{ivx}$ is implemented introducing the Lagrange multiplier $v(\tau)$, thus adding an additional quadratic term (in σ fields) to the action (4) and allowing one to perform N independent traces over σ_i. To accomplish this we take the diagonal representation for the random symmetric matrix J_{ij}, namely, $\sum_i J_{ij}\phi_j^\lambda = J_\lambda\phi_j^\lambda$ with the real orthonormal eigenvectors ϕ_i^λ (here, $\lambda = 1,\ldots,N$ and J_λ is the λth eigenvalue). In the thermodynamic limit $N \to \infty$ the saddle point $v(\tau) \equiv v_0$ becomes exact yielding for the spherical constraint (2)

$$1 = \frac{1}{\beta}\sum_{\omega_\ell}\sum_\lambda \chi_\lambda(\omega_\ell) + \frac{1}{N}\sum_\lambda h_\lambda^2\chi_\lambda^2(0)\,, \qquad (5)$$

where the sum runs over the (Bose) Matsubara frequencies $\omega_\ell = 2\pi\ell/\beta$ ($\ell = 0, \pm1, \pm2, \ldots$), $\chi_\lambda(\omega_\ell) = (2v_0 - J_\lambda + \omega_\ell^2/\Delta)^{-1}$ is the "shattered" dynamic susceptibility, $h_\lambda = h\sum_j \phi_j^\lambda$, and on the average, using the orthonormality of the eigenvectors ϕ_j^λ, one can replace h_λ^2 by h^2.

The lowest order linear response, i.e., the dynamic ac susceptibility

$$\chi(\omega, h) = \frac{1}{N}\sum_\lambda \int_0^\beta d\tau\, e^{i\omega_\ell\tau}\langle\sigma_\lambda(\tau)\sigma_\lambda(0)\rangle_T \bigg|_{i\omega_\ell\to\omega+i0^+}, \qquad (6)$$

subject to the spherical constraint (5) never becomes critical as $h \to 0$. To access the criticality one has to consider a quantity which couples to the SG order and plays the role in a SG that the uniform susceptibility does in a ferromagnet. It is given by the quantum mechanical *four*-spin correlation function—the spin glass susceptibility (sometimes also called "order parameter" or Edwards-Anderson susceptibility),

$$\begin{aligned}\chi_{\rm SG}(\tau_1 - \tau_2, \tau_3 - \tau_4) &= \sum_{ij}\langle\langle\sigma_i(\tau_1)\sigma_j(\tau_2)\rangle_T \times \langle\sigma_i(\tau_3)\sigma_j(\tau_4)\rangle_T\rangle_{\rm av} \\ &= \frac{1}{\beta^2 N}\sum_{\omega_\ell,\omega_\ell'} e^{i\omega_\ell(\tau_1-\tau_2)+i\omega_\ell'(\tau_3-\tau_4)} \times \sum_\lambda \chi_\lambda(\omega_\ell)\chi_\lambda(\omega_\ell')\,. \qquad (7)\end{aligned}$$

In the thermodynamic limit $N \to \infty$, the summation over eigenstates can easily be done by observing

$$\frac{1}{N}\sum_\lambda\ldots \to \int_{-2J}^{2J} d\epsilon\, \rho(\epsilon)\ldots\,, \qquad (8)$$

where $\rho(\epsilon) = \lim_{N\to\infty} N^{-1}\sum_\lambda\langle\delta(\epsilon - J_\lambda)\rangle_{\rm av}$ is the averaged density of states describing the eigenvalue spectrum of the random matrix J_{ij}. For Wigner ensemble of Gaussian distributed J_{ij} with zero mean the famous semicircle law emerges with $\rho(\epsilon) = (2\pi J^2)^{-1}\sqrt{4J^2 - \epsilon^2}$ for $|\epsilon| < 2J$ leading to

$$\chi(\omega_\ell) = \frac{1}{2J^2}\left[2v_0 + \frac{\omega_\ell^2}{\Delta} - \sqrt{\left(2v + \frac{\omega_\ell^2}{\Delta}\right)^2 - 4J^2}\,\right]. \qquad (9)$$

Inferring Eqs. (7) and (9) we can manipulate the Fourier transformed SG susceptibility into

$$\chi_{\rm SG}(\omega_\ell, \omega_\ell') = \frac{\chi(\omega_\ell)\chi(\omega_\ell')}{1 - J^2\chi(\omega_\ell)\chi(\omega_\ell')}\,, \qquad (10)$$

showing that $\chi_{\rm SG}(0,0)$ indeed exhibit singularity where $1 = J\chi(0)$ which happens at $h = 0$ on the critical line $T_c(\Delta)$ where the Lagrange multiplier $2v_0(T,\Delta)$ "sticks" at the upper limit of the eigenvalue spectrum ($J_\lambda^{\rm max} = 2J$) of the random matrix J_{ij} [19].

Though $\chi_{\rm SG}$ is not directly measurable in experiments it becomes accessible via the nonlinear response $\chi_{\rm nl}$. By expanding $\chi(\omega, h)$, Eq. (9) in powers of the field h one obtains

$$\chi(\omega, h) = \chi(\omega, 0) + \sum_{k=1}^{\infty} \frac{1}{2k!} \chi_{2k+1}(\omega) h^{2k}, \qquad (11)$$

with $\chi_3(\omega) \equiv \chi_{\rm nl}(\omega)$ being the lowest order frequency dependent nonlinear response,

$$\chi_{\rm nl}(\omega) = \chi_{\rm SG}(\omega, \omega) v_h^{(2)}, \qquad (12)$$

where

$$v_h^{(2)} \equiv \left. \frac{d^2 v_0(h)}{dh^2} \right|_{h=0} = \beta \frac{\sum_\lambda \chi_\lambda^2(0)}{\sum_{\omega_\ell} \sum_\lambda \chi_\lambda^2(\omega_\ell)} \qquad (13)$$

originates from the differentiation of the spherical constraint (5) with respect to the magnetic field h. Explicitly, after performing summation over eigenstates and Matsubara frequencies one obtains

$$\frac{1}{N} \sum_\lambda \chi_\lambda^2(0) = \frac{1}{J^2} \left(\frac{v_0}{\sqrt{v_0^2 - J^2}} - 1 \right) \qquad (14)$$

and

$$\frac{1}{N\beta} \sum_{\omega_\ell} \sum_\lambda \chi_\lambda^2(\omega_\ell) = \frac{1}{4} \int_{-2J}^{2J} d\epsilon\, \rho(\epsilon) \times \left\{ \frac{\sqrt{\Delta}}{(2v_0 - \epsilon)^{3/2}} \coth\left(\frac{\beta\sqrt{\Delta(2v_0 - \epsilon)}}{2} \right) - \frac{1}{2} \frac{\beta\Delta}{2v_0 - \epsilon} \left[1 - \coth^2\left(\frac{\beta\sqrt{\Delta(2v_0 - \epsilon)}}{2} \right) \right] \right\}. \qquad (15)$$

Consider now a $T = 0$ phase transition between the paramagnetic and SG phases, induced by varying the parameter Δ. Introducing the energy parameter $\mu = \sqrt{2\Delta(v_0 - J)}$ measuring the distance, in parameter space, from criticality ($\mu = 0$ defines the transition point). We obtain $\chi_{\rm SG}(0, 0) \sim 1/\mu$ close to the critical point. The singularity of $\chi_{\rm nl}(0)$ turns out to be stronger, however—the *additional* divergency comes from the second derivative $v_h^{(2)}$ of the Lagrange multiplier $v_0(h)$ [cf. Eq. (13)]. It is readily seen that the expression (15) is singular *at* $T = 0$ critical point. Indeed, at the zero temperature

$$\frac{1}{N\beta} \sum_{\omega_\ell} \sum_\lambda \chi_\lambda^2(\omega_\ell) \to \int_{-\Lambda_\omega}^{\Lambda_\omega} \frac{d\omega}{\sqrt{\omega^2 + \mu^2}} \sim \ln\left(\frac{\mu}{\Lambda_\omega^{1/2}} \right), \qquad (16)$$

where Λ_ω denotes the frequency cutoff. Therefore, [cf. Eqs. (13), (14), and (15)] one obtains $v_h^{(2)} \sim 1/(\mu \ln \mu)$, and consequently we infer from Eq. (12) that $\chi_{\rm nl} \sim 1/(\mu^2 \ln \mu)$. With the constraint on the spin length (5) implying that the gap μ vanishes logarithmically faster as $\Delta \to \Delta_{c0} \equiv \Delta_c(T = 0)$ [20], $\mu \sim [(\Delta - \Delta_{c0})^{-1} \ln(\Delta - \Delta_{c0})]^{-1/2}$, we find for the nonlinear response

$$\chi_{\rm nl}(\omega = 0) \sim \frac{(\Delta - \Delta_{c0})^{-1} \ln(\Delta - \Delta_{c0})}{\ln[(\Delta - \Delta_{c0})^{-1} \ln(\Delta - \Delta_{c0})]}, \qquad (17)$$

with novel and unusual violation of the universal scaling by the *double* logarithms [21].

However, the experimental results for the nonlinear susceptibility are derived from ac values of the nonlinear susceptibility at a *finite* probing frequency. Therefore, to examine this issue we have calculated the dynamic nonlinear response by analytically continuing $\chi_{\rm nl}(\omega_\ell)$ to the domain of *real* frequencies $\chi_{\rm nl}(\omega) = \chi'_{\rm nl}(\omega) + i\chi''_{\rm nl}(\omega)$ with real (imaginary) part $\chi'_{\rm nl}(\omega)$ [$\chi''_{\rm nl}(\omega)$]. From Eqs. (9), (10), and (12) we obtain

$$\chi'_{\rm nl}(\omega) = \begin{cases} \frac{1}{J^2}\left(\frac{2v_0 - \omega^2/\Delta}{\sqrt{(2v_0 - \omega^2/\Delta)^2 - 4J^2}} - 1 \right) v_h^{(2)} & \text{for } \omega^2 \mu^2, \\ -v_h^{(2)} J^2 & \text{for } \mu^2 \le \omega^2 \le \mu^2 + 4\Delta v_0, \\ \frac{1}{J^2}\left(\frac{2v_0 - \omega^2/\Delta}{\sqrt{(2v_0 - \omega^2/\Delta)^2 - 4J^2}} + 1 \right) v_h^{(2)} & \text{for } \omega^2 > \mu^2 + 4\Delta v_0, \end{cases} \qquad (18)$$

and

$$\chi''_{\rm nl}(\omega) = \frac{\mathrm{sign}(\omega)}{J^2} \Theta\left(1 - \left| \frac{v_0}{J} - \frac{\omega^2}{2\Delta J} \right| \right) \times \frac{(2v_0 - \frac{\omega^2}{\Delta}) v_h^{(2)}}{\sqrt{[2(J - v_0) + \frac{\omega^2}{\Delta}][2(J + v_0) - \frac{\omega^2}{\Delta}]}}, \qquad (19)$$

where $\Theta(x)$ is the unit step function.

Consider now the situation in the vicinity of the zero-temperature paramagnetic-SG transition. Raising the temperature at $\Delta = \Delta_{c0}$ one enters the QC regime in which the physics is dominated by the $T = 0$ quantum critical point. Here the temperature is the most significant energy scale and the system "feels" the *finite* value of T before becoming sensitive to the deviation of Δ from Δ_{c0} [12]. In particular, for the energy parameter μ which defines the frequency scale one obtains from the constraint equation (2) $\mu(T) \sim k_B T / \ln^{1/2}(\Lambda_\omega / k_B T)$ thus implying that

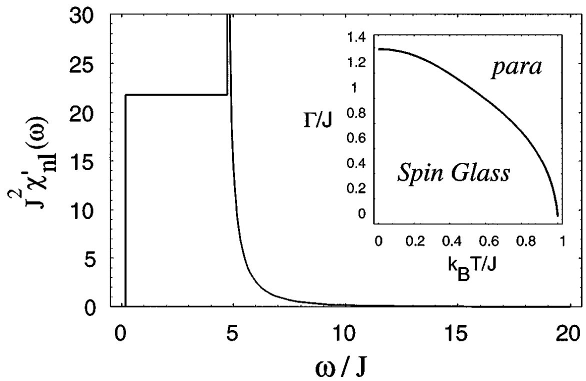

FIG. 1. Frequency dependence of the real part of the dynamic nonlinear response $\chi'_{nl}(\omega)$ in the quantum critical regime: $T/J^{\lambda}_{max} = 0.1$ at the critical point $\Delta \equiv \Delta_c(T = 0) = 9\pi^2/16$ (which translates into the transverse field $\Gamma/J \approx 1.5$; see Ref. [18]). The inset shows the Γ-T phase diagram of the quantum spherical SG model, Eq. (1), for $h = 0$.

$\mu(T) \to 0$ as $T \to 0$. The frequency behavior of $\chi'_{nl}(\omega)$ in this region is very remarkable (see Fig. 1). For frequencies $\mu(T) \leq \omega \leq 2\sqrt{\Delta J}$ [where the imaginary part $\chi''_{nl}(\omega)$, Eq. (19), is nonzero] we infer from Eq. (18) that $\chi'_{nl}(\omega) = -v_h^{(2)}/J^2$, i.e., the nonlinear susceptibility is frequency *independent* and nonsingular. In the opposite high frequency region $\omega > 2\sqrt{\Delta J}$ we find nonlinear response vanishing as $|\chi'_{nl}(\omega)| \sim 1/\omega^4$. Interestingly, this behavior is consistent with available experimental findings for disordered dipolar magnet $LiHo_xY_{1-x}F_4$. Measurements of the ω dependence of $\chi'_{nl}(\omega)$ for transverse field which is 1% above the critical field strength marking the transition to the SG phase close to $T = 0$ critical point indicate the frequency-independent nonlinear response [22]. Experimental data clearly illustrate the crossover between high-ω and low-ω (frequency-independent) behaviors—in remarkable agreement with theoretical prediction for $\chi'_{nl}(\omega)$ made here. Finally, we emphasize that the behavior near the QC point is quantitatively different from the case where $\chi_{nl}(\omega)$ is measured by a finite probing frequency with response falling out of equilibrium before the transition temperature—a situation typically appearing at the classical critical point; in this case $\chi_{nl}(\omega)$ will be frequency dependent showing a maximum at the freezing transition (see Ref. [23]).

In summary, we explored the dynamic nonlinear response in a quantum spin glass modeled by the exactly solvable spherical bond-disordered quantum spin system with infinite connectivity. We have argued that some puzzling aspects of the behavior of the nonlinear susceptibility appear to receive a natural explanation as the novel $T = 0$ quantum glass regime is approached.

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