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Equilibrium dynamics of a paramagnetic cluster

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The transverse autocorrelation function at an arbitrary temperature is calculated rigorously for a system with an arbitrary number of quantum spins, each of which is coupled to all the other spins with equal exchange. It is shown that the cluster approximation, formed by a model for a real magnet, explains short- and intermediate-time (up to the time to reach the spin-diffusion regime) experimental measurements in the paramagnetic phase.

For more than forty years of theoretical research on the equilibrium dynamics of quantum Heisenberg magnets at high temperatures, the behavior at intermediate times (that is of the order of the inverse value of exchange between a spin and its surrounding) has received no reasonable explanation. The Bloembergen suggestion[1] to describe an equilibration of a higher excited spin system as some random-walk process was followed by several attempts concentrating mainly on investigation of the longest-time behavior, in the so-called spin-diffusion regime. The synthetic adoption approach,[2–8] in which a suitable function (from the point of view of the expected long-time behavior) is used to fit certain moments known from the direct short-time expansion, could not describe experimental measurements on an intermediate time scale.[9–12] A comparison of the mode-coupling theories[13–16] with experiment also shows the same remarkable discrepancy (see experimental work[11] for review). All these theoretical approaches show a monotonic time decay or a negative value of the transverse pair-spin autocorrelator, but the experimental curve is positive and has a minimum at some intermediate time.[11] The most representative paper of the mode-coupling approach[15] is used the clear physical idea that each spin moves in a randomly varying effective magnetic field produced by its neighbors. This idea is very natural on the largest time scale, but must be reexamined on an intermediate time scale, when it seems more suitable to use a cluster mean field with a nonrandom but self-consistent effective magnetic field created by the surrounding spins.

To test this hypothesis we can restrict ourselves to the following cluster model. Let a system of N $\frac{1}{2}$ spins be described by the uniform-range Heisenberg Hamiltonian, where N is the number of spins in the exchange sphere of an original magnet:

$$\hat{\mathscr{H}}=-(J/2N)(\hat{\sigma})^2, \tag{1}$$

where $JN/(N-1)$ is an exchange strength between the spin and its surrounding and $\hat{\sigma}=\Sigma_j^N\hat{\mathbf{s}}$ is the total spin operator of the system. Suppose the system is in thermal equilibrium with an inverse temperature β. An investigation of the static expectation values of the system is (in the limit $N\gg 1$) the subject of mean-field theory for magnets with a long-range exchange.[17,18] A question arises: is it possible to create a dynamical mean-field approach for those magnets? Or, returning to the system of N uniformly coupled spins, what is the equilibrium dynamics? The uniform feature of the exchange makes the spatial correlations homogeneous, which allows one to express any spin correlator in terms of autocorrelation functions. The study of the dynamic properties of such a model as its anisotropic generalization (the so-called van der Waals model) was pioneered by Dekeyser and Lee.[19–21] In Refs. 20 and 21 they answered the "thermodynamic" ($N\rightarrow\infty$) part of the question posed above: it was shown, first, that in the limit $N\rightarrow\infty$, the dynamics of a single spin and the total remaining spin are coupled to each other linearly;[20] that allowed them subsequently to determine the spin autocorrelation function analytically.[21] However, this direct scheme, which builds on a straightforward resolution of the single-spin dynamics at $N=\infty$, fails to describe the cluster (finite-N) situation.

In this paper, we use a functional integral formalism with the model described by the Hamiltonian (1) to obtain *an exact expression for the temporal dependence of the transverse-pair autocorrelator at arbitrary temperature and number of spins.* We show that the cluster approximation, formed by the model for a real magnet (N plays the role of the number of spins in the exchange sphere) explains the short- and intermediate-time experimental measurements in the paramagnetic phase.

Our starting point is the transverse pair autocorrelator in the following well-defined form:

$$\mathscr{K}(t,\beta)\equiv\frac{1}{2}\frac{\mathrm{Tr}\{[\hat{s}^-(0)\hat{s}^+(t)+\hat{s}^+(0)\hat{s}^-(t)]e^{-\beta\hat{\mathscr{H}}}\}}{\mathrm{Tr}[e^{-\beta\hat{\mathscr{H}}}]}, \tag{2}$$

where $\hat{s}^{\pm}(t)$ is the usual notation for up- and down-spin operators in the Heisenberg representation $\hat{s}^{\pm}(t)=e^{it\hat{\mathscr{H}}}\hat{s}^{\pm}e^{-it\hat{\mathscr{H}}}$, $\hat{s}^{\pm}=\hat{s}^x\pm i\hat{s}^y$. The Hamiltonian (1) commutates with the full spin of the system $\hat{\sigma}$. It means that we can classify the eigenstates of the Hamiltonian by the value of the spin angular momentum L, that can take the positive values $L=N/2$, $N/2-1,\ldots,$ and its projection on the z axes M, that can take the values $M=-L,-L+1,\ldots,L$. The energy level $E_L=-JL(L+1)/2N$ corresponding to the value of full spin L thus has $(2L+1)$-fold degeneracy. This $|L,M\rangle$ representation of the eigenstates would be very convenient for a calculation of a diagonal matrix element (or a trace) defined in terms of the full spin operator only. Except for a calculation of a diagonal matrix element of a one-spin operator that does not commutate with the Hamiltonian, as in (2), this representation is not suitable.

To avoid the effect of the mixing of the $|L,M\rangle$ eigenstates by the one-spin operators $\hat{s}^{\pm}$, we will formulate a technique that maps the initial $\hat{\sigma}$-phase space into some unbounded functional one. The resulting one-particle quantum mechanics turns out to be exactly solvable. Here we present a sketch of the method, postponing details to a more detailed publication. Let us also note, to avoid misunderstanding, that the method we use has nothing to do with the Bethe-ansatz or inverse-scattering approaches.

We begin from the infinite-temperature case in which the correlator (2) is reduced to

$$\mathcal{K}^0(t;N)=\mathrm{Tr}[e^{-it\hat{\mathcal{H}}}\hat{s}^{+}e^{it\hat{\mathcal{H}}}\hat{s}^{-}]. \tag{3}$$

In order to calculate the N-spins trace we perform Hubbard-Stratonovich transformations of the evolution operators from the definition (3)

$$e^{-it\hat{\mathcal{H}}}\propto\int\mathcal{D}\varphi_1\exp\left(-\frac{iN}{2}\int_0^t dt'\varphi_1^2\right)\prod_j\mathcal{A}_j^{(1)}, \tag{4}$$

$$e^{it\hat{\mathcal{H}}}\propto\int\mathcal{D}\varphi_2\exp\left(-\frac{iN}{2}\int_0^t dt'\varphi_2^2\right)\prod_j\mathcal{A}_j^{(2)}, \tag{5}$$

$$\mathcal{A}_j^{(1,2)}(t)=T\exp\left(i\int_0^t dt'\boldsymbol{\varphi}_{1,2}(t')\hat{\mathbf{s}}_j\right), \tag{6}$$

factoring the initial trace to a product of local ones (here and further we measure t in the units of J^{-1}). Due to an absence of any time dependence in the Hamiltonian (1) we had no time-ordered exponentials in (3). Still, after the Hubbard-Stratonovich transformation, T exponents appeared [in $\mathcal{A}_j^{(1,2)}(t)$]. This stems from the noncommutativity of the full spin of the system $\hat{\sigma}$ with an arbitrary one-site operator $\hat{\mathbf{s}}_i$, and reflects the multiplicativity (in discretized time) of the functional measures in (4) and (5). At first glance, those time-ordered exponentials produce real difficulties. Indeed, we cannot calculate them explicitly as functionals of $\boldsymbol{\varphi}_{1,2}$. However, a substitution does exist which recasts the time-ordered exponentials (6) into the products of the usual exponentials (see Refs. 22 and 23) by means of the following change of variables $\boldsymbol{\varphi}\to(\rho,\psi^{\pm})$ in the functional integrals (4) and (5):

$$\varphi_{1,2}^z=\rho_{1,2}\mp 2\psi_{1,2}^+\psi_{1,2}^-,$$
$$\varphi_1^-=\psi_1^-,\quad \varphi_1^+=-i\dot{\psi}_1^+ +\rho_1\psi_1^+-(\psi_1^+)^2\psi_1^-,$$
$$\varphi_2^+=\psi_2^+,\quad \varphi_2^-=-i\dot{\psi}_2^- -\rho_2\psi_2^- -(\psi_2^-)^2\psi_2^+, \tag{7}$$

where $\varphi^{\pm}=(\varphi^x\pm i\varphi^y)/2$. Equation (7) deals with the difficulty, mentioned above, that it is impossible to express T exponentials in terms of the usual functions of initial variables $\boldsymbol{\varphi}$; in general, ρ and $\psi^{\pm}$ cannot be expressed solely in terms of $\boldsymbol{\varphi}$ from (7). But to perform changes of variables $\boldsymbol{\varphi}\to(\rho,\psi^{\pm})$ in the functional integrals (4) and (5) it is not necessary to invert (7). Explicit expressions for $\mathcal{A}^{(1,2)}(t)$ and the Jacobian of (7) in terms of the new variables ρ and $\psi^{\pm}$ may be found by analogy with what has been explained in detail in Refs. 22 and 23. After a calculation of the N local tracers, we arrive at the following functional representation for $\mathcal{K}^0(t;N)$:

$$\mathcal{K}^0=\mathrm{const}\int\mathcal{D}\rho_{1,2}\mathcal{D}\psi_{1,2}^{\pm}e^{\mathcal{S}_0}\mathcal{B}^{N-1}\mathcal{C},$$

$$\mathcal{S}_0=-\frac{iN}{2}\int_0^t[\rho_1^2-\rho_2^2-4i\dot{\psi}_1^+\psi_1^-+4i\psi_2^+\dot{\psi}_2^-]dt+\frac{i}{2}\int_0^t(\rho_1-\rho_2)dt,$$

$$\mathcal{B}=\mathrm{Tr}[\mathcal{A}^{(1)}\mathcal{A}^{(2)}]=2\cos\left[\frac{i}{2}\int_0^t(\rho_1+\rho_2)dt'\right]+e^{(i/2)\int_0^t(\rho_1-\rho_2)dt'}\left(\psi_1^+ + i\int_0^t\psi_2^+e^{i\int_0^t\rho_2 dt'}\right)\times\left(\psi_2^-+i\int_0^t\psi_1^-e^{-i\int_0^t\rho_1 dt'}\right),$$

$$\mathcal{C}=\mathrm{Tr}[\mathcal{A}^{(1)}\hat{s}^+\mathcal{A}^{(2)}\hat{s}^-]=e^{(i/2)\int_0^t(\rho_1-\rho_2)dt'}. \tag{8}$$

Normalization of the functional integral (8) depends on N only, and it can be fixed by an evident condition $\mathcal{K}^0(t=0)=1$. The transversal fields $\psi_{1,2}^{\pm}$ have no dynamics at all. Indeed, the functional integral in (8) remains the same if the fields $\psi_2^+(t')$, $\psi_1^-(t')$ at an arbitrary moment $0<t'<t$ are replaced by $\psi_2^+(t)$, $\psi_1^-(t)$ correspondingly. But the dynamics of the ρ fields keeps it very nontrivial. However, it turns out that the functional integral (8) over the ρ fields is of the Feynman-Kac type[24] and we can calculate it explicitly. The problem is transformed to a calculation of some matrix element of the accompanying one-dimensional quantum mechanics with the Hamiltonian

$$\hat{\mathcal{H}}_{\mathrm{ac}}=-(1/2N)\,\partial_\xi^2+(N/2)\,e^{-\xi}. \tag{9}$$

Let us note that a similar matrix element with respect to exactly the same quantum mechanics appears in the calculation of the multipoint densities correlator in $1d$ localization.[25] The wave function (and all the matrix elements correspondingly) is calculated explicitly. In total we therefore obtain the following answer for the equilibrium transverse pair correlator at infinite temperature:

$$\mathcal{K}^0(t;N)=\frac{2}{3}\left\{\frac{1}{2}+\left(\cos\frac{t}{2N}\right)^N-\frac{1}{N}\left[\left(\cos\frac{t}{2N}\right)^{N-2}-1\right]-(N-1)\sin^2\frac{t}{2N}\left(\cos\frac{t}{2N}\right)^{N-2}\right\}. \tag{10}$$

$\mathcal{K}^0(t;N)$ is periodic in time; starting from unit a zero time it relaxes to a minimum, then restores up to a plateau ($\frac{1}{3}$ at $N\to\infty$), and becomes again unit at $t_{\mathrm{per}}=4\pi N$. The direct calculations of $\mathrm{Tr}[e^{-it\hat{\mathcal{H}}}\hat{s}^+e^{it\hat{\mathcal{H}}}\hat{s}^-]$ at small N confirm the result (10).

In the limit of large N, when t_{per} is not reached, an intermediate asymptotic takes place. Thus, at $t=\tau\sqrt{N}$, $\tau\sim 1$, $N\gg 1$ we have a smooth relaxation depending on N via τ only

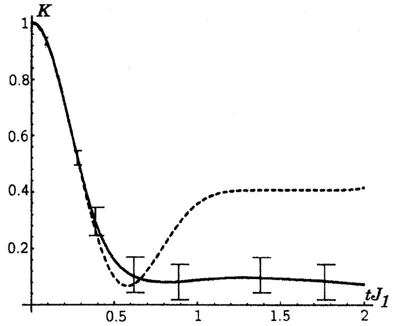

FIG. 1. The full curve is the Fourier transform of the measured data for the spectral densities of the transversal autocorrelation function at relatively high temperature in $Rb_2CuBr_4*H_2O$. J_1 is the value of the nearest-neighbor exchange in the magnet. The dashed line is the cluster result (10) with an appropriate choice of parameters J and N.

$$\mathcal{K}^0(t)\approx\tfrac{1}{3}\{1+2e^{-\tau^2/8}-(\tau^2/2)e^{-\tau^2/8}\}. \tag{11}$$

The result (11) is characterized by a Gaussian bump and a nonvanishing tail at $\tau\to\infty$ (in strong agreement with Ref. 21). Let us note that the Gaussian bump, which as we will see below takes place also at lower temperatures, has also been obtained by Belinicher and L'vov for the Green function of dispersion-free magnons in long-range quantum magnets.[26] The asymptotic value of $K^0(\tau)$ at $\sqrt{N}\ll t\ll N$ ($\frac{1}{3}$ at $N\to\infty$) stems from eigenstates of the initial quantum mechanics (1) with the zero full spin $\sigma=0$. The nonvanishing tail is just an artifact of the nonergodicity of our model. Thus, in the case of a more realistic long-range model the plateau can be realized in the thermodynamic limit only as an intermediate-time asymptotic.

The present results give approximations for a real quantum magnet. The thermodynamic ($N=\infty$) result (11) forms the zeroth order with respect to the small parameter $1/N$ cluster approximation (N plays the role of the number of spins in the exchange sphere; let us note that in $3d$ even in the case of the nearest-neighbor interaction $1/N$ is a reasonable enough small parameter). However, (10) can give us more if we consider N as the number of spins in the exchange sphere of a real magnet. The experimental observations[11] support this statement. The experimental curve for the transversal autocorrelator in $Rb_2CuBr_4*H_2O$ (which is three-dimensional bcc, $s=\frac{1}{2}$) repeats with very good agreement the dependence (11) in the interval of times from zero up to the moment of time when $\mathcal{K}^0(t;N)$ reaches its minimal value [see Fig. 1 for comparison of some experimental measurements and (11) with an appropriate choice of constants J and N]. Later in time the dependence (11) deviates from the experimental curve, which restores the value approximately on 0.05 units to cross over further into a spin-diffusion tail at the largest times.

The behavior (11) looks similar to the calculations[27] for spin in a classical random field. However, in our case there was no external randomness at all. And the constant of Gaussian relaxation, which had been external in Ref. 27, is defined in our consideration by means of the dynamics itself.

In a recent paper[28] we investigated the long-time dynamics of an arbitrary-exchange quantum Heisenberg model at infinite temperature. It was shown that the quantum spin pair correlator (in the para-phase due to unbroken symmetry of the Hamiltonian it is just the transverse correlator multiplied by $\frac{3}{4}$) is equal to the correlator of a classically evaluated vector field $\boldsymbol{\phi}_k(t)$

$$\dot{\boldsymbol{\phi}}_k=\sum_j J_{kj}[\boldsymbol{\phi}_k\times\boldsymbol{\phi}_j], \tag{12}$$

averaged over the initial conditions $\boldsymbol{\phi}_k(0)=\mathbf{p}_k$ with respect to the Gaussian measure

$$\prod_k d\mathbf{p}_k\exp\left\{-\frac{1}{2s(s+1)}\sum_i \mathbf{p}_i^2\right\}. \tag{13}$$

This "classical" problem, remaining strongly nonlinear at an arbitrary exchange J_{ij}, becomes linear and exactly solvable for the uniform exchange $J_{ij}=J/N$. Indeed, in the uniform case the right-hand side of (12) is $J[\boldsymbol{\phi}_k\times\mathbf{P}]/N$, where $\mathbf{P}=\Sigma_k\mathbf{p}=\Sigma_k\boldsymbol{\phi}$ is the integral of motion. The classical motion of a spin turns out to be just *the uniform precession around the total spin of the system.* It is remarkable that the $\frac{1}{3}$ universal (at $N\to\infty$ only) tail has in these terms a very simple explanation: $\overline{\cos^2\theta}=\frac{1}{3}$ stems from the longitudinal (parallel to the full spin of the system) single-spin projection. It yields at $N\gg 1$, $t=\tau\sqrt{N}$ and spin $s=\frac{1}{2}$ exactly the same answer (11) for the transverse autocorrelator. Let us note that the above-mentioned physical picture of the linear single-spin dynamics was described by Dekeyser and Lee.[20] Thus, we obtained first, a good physical picture resulting in (11), and second, that the transition to the classical model is valid not only at a large enough time,[28] but also for a long enough exchange rate.

Armed with this understanding of the infinite temperature case we can go forward to a finite temperature. It is possible to show that the approach resulting in (10) is generalized on finite temperatures. Indeed, the substitution in the first exponential in the right-hand side of (3) $t_1=t-i\beta$ instead of t produces $\mathcal{K}^+(t,\beta)$, the real part of which gives $\mathcal{K}(t,\beta)$ (2). Thus, omitting the details of the calculation we write here the result

$$\begin{aligned}\mathcal{K}^+(t,\beta)=&\frac{1}{\sqrt{\beta}Z(\beta)}\int_{-\infty}^{+\infty}dt'(\cosh t')^N e^{-2t'^2N/\beta}\\&\times\left[\frac{2Ne^{-t'}}{\beta}\left(\frac{4t'^2N}{\beta}-t'-1\right)-4\exp\left(\frac{\beta}{8N}\right.\right.\\&\left.+\frac{2it'}{\beta}(t-i\beta)+\frac{(t-i\beta)^2}{2\beta N}\right)\left(1+\frac{N}{\beta}-\frac{4t'^2N^2}{\beta^2}\right.\\&\left.\left.+\frac{(4it'N+t-i\beta)(t-i\beta)}{\beta^2}\right)\right],\end{aligned} \tag{14}$$

where $Z(\beta)$ is the partition function being defined from this expression with the condition $\mathrm{Re}[\mathcal{K}^+(0,\beta)]=1$.

In the intermediate asymptotic $t=\tau\sqrt{N}$, $\tau\sim 1$, $N\gg 1$ the saddle-point approximation of (14) gives a generalization of (10) to the finite-temperature paramagnetic case $(4-\beta)N\gg 1$

$$\mathcal{K}(t,\beta)\approx\tfrac{1}{3}\{1+2[1-\tau^2(4-\beta)]e^{-\tau^2/2(4-\beta)}\}, \tag{15}$$

when the only solution of the saddle-point equation

$$\tanh t^* = 4t^*/\beta, \tag{16}$$

is $t^*=0$ [in the low-temperature case two nonzero solutions of (16) appear, and one of them has to be chosen as the saddle point in the corresponding $N \gg 1$ calculations]. In this case $\beta<4$, $Z(\beta) \propto (4-\beta)^{-3/2}$. Thus, we conclude that at $\beta=4$ there is a peculiarity of usual phase transition type (of course a phase transition exists only in the limit of a large number of spins $N \gg 1$). It is clear that the static critical exponent $\frac{3}{2}$ is just the mean field (by construction) one but its evaluation is useful for a control of the really complicated dynamical calculations. We see that the squared inverse time of Gaussian relaxation (15) goes linearly to ∞ with $4-\beta \to 0$. The results (14) and (15) are shown graphically in Fig. 2.

To conclude, for the N-spin uniform exchange quantum model we have rigorously calculated the temporal dependence of the transverse pair autocorrelator at an arbitrary number of spins and temperatures. In the para-phase the correlator shows Gaussian relaxation from 1 at $t=0$ via a minimum to a (universal $=\frac{1}{3}$ at $t \sim \sqrt{N} \to \infty$) plateau (see Fig. 2).

The results are obtained by a method that is nothing more than *a quantum cluster dynamical mean-field* one and that by the construction turns out to be *exact*.

From the point of view of possible applications to long-range quantum magnets those exact results are also unique in generating a starting dynamical approximation for the para-phase. The possibility of having an exact result for a general N is very important. Indeed, the full answer (14) gives an approximation to the problem with a large but finite radius of exchange in an infinite magnet, when N plays the role of a number of spins in the exchange sphere. The physical picture accompanied by this approximation is clear from the classical model (12) and (13): the dynamics of a spin is defined by its uniform precession around the full spin of the system. A comparison of the approximation, which gives for the equilibrium autocorrelation function in the paramagnetic phase the Gaussian relaxation from 1 at $t=0$ via the minimum at $t=t_m$ (for $\beta=0$, ≈ 0.07 at $t_m \approx 3.5\sqrt{N}/J$) to a plateau, with the corresponding experimental curve[11] gives a coincidence at short and intermediate (up to t_m) times; the experimental results show that the cluster approximation fails immediately after t_m, when a spin-diffusion regime begins to shape. We expect the universality of the minimum in temporal behavior of the autocorrelation in a more realistic (Heisenberg) model with a finite exchange range. To prove it precise analytical calculations as experimental measurements describing cross-over between the cluster and spin-diffusion regimes are required.

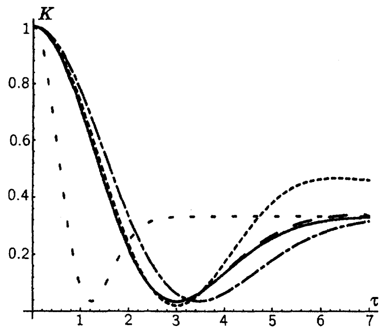

FIG. 2. Transverse pair autocorrelator as a function of τ. The six collected plots correspond to different value of inverse temperature β and number of spins N in the cluster: the full line: $\beta=1$, asymptotic $N \to \infty$; the long-dashed line: $\beta=1$, $N=100$; the dashed line: $\beta=1$, $N=10$; the dashed-dotted line: $\beta=0$, asymptotic $N \to \infty$; the dotted line: $\beta=3.5$, asymptotic $N \to \infty$.

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