Local Spin Relaxation within the Random Heisenberg Chain

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Finite-temperature local dynamical spin correlations $S_{nn}(\omega)$ are studied numerically within the random spin-1/2 antiferromagnetic Heisenberg chain. The aim is to explain measured NMR spin-lattice relaxation times in $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$, which is the realization of a random spin chain. In agreement with experiments we find that the distribution of relaxation times within the model shows a very large span similar to the stretched-exponential form. The distribution is strongly reduced with increasing T, but stays finite also in the high-T limit. Anomalous dynamical correlations can be associated with the random singlet concept but not directly with static quantities. Our results also reveal the crucial role of the spin anisotropy (interaction), since the behavior is in contrast with the ones for the XX model, where we do not find any significant T dependence of the distribution.

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One-dimensional (1D) quantum spin systems with random exchange couplings reveal interesting phenomena fundamentally different from the behavior of ordered chains. Since the seminal studies of antiferromagnetic (AFM) random Heisenberg chains (RHCs) by Dasgupta and Ma [1,2] using the renormalization-group approach and further development by Fisher [3], it has been recognized that the quenched disorder of exchange couplings J leads at lowest energies to the formation of random singlets with vanishing effective $\tilde{J}$ at large distances. The consequence for the uniform static susceptibility χ^0 is the singular Curie-type temperature (T) dependence, dominated by nearly uncoupled spins at low T and confirmed by numerical studies of model systems [4], as well as by measurements of $\chi^0(T)$ on the class of materials being the realizations of RHC physics, in particular the mixed system $BaCu_2(Si_{1-x}Ge_x)_2O_7$ [5–7].

Recent measurements of NMR spin-lattice relaxation times T_1 in $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$ [6] reveal a broad distribution of different T_1 resulting in a nonexponential magnetization decay being rather of a stretched-exponential form. In connection to this, the most remarkable is the strong T dependence of the T_1 span becoming progressively large and the corresponding distribution non-Gaussian at low T. It is evident that in a random system T_1, which is predominantly testing the local spin correlation function $S_{nn}(\omega \to 0)$, becomes site n dependent and we are therefore dealing with the distribution of T_{1n} leading to a nonexponential magnetization decay.

Theoretically the behavior of dynamical spin correlations in RHCs has not been adequately addressed so far. There is (to our knowledge) no established model result and moreover no clear prediction for the behavior of dynamical ($\omega \neq 0$) spin correlations at $T > 0$ in RHCs. It seems plausible that the low-T behavior should follow from the random-singlet concept and its scaling properties, discussed within the framework of the renormalization-group approaches [2,3,8,9]. Still, the relation to singular static correlations as evidenced, e.g., by $\chi^0(T)$ diverging at $T \to 0$, and low-ω dynamical correlations is far from clear.

One open question is also the qualitative similarity to the behavior of the random anisotropic XX chain invoked in several studies [4,8–10]. The latter system is equivalent to the more elaborated problem of noninteracting (NI) spinless fermions with the off-diagonal (hopping) disorder [11,12].

In the following we present results for the dynamical local spin correlation function $S_{nn}(\omega)$, in particular for its limit $s = S_{nn}(\omega \to 0)$ relevant for the NMR T_1, within the AFM RHC model for $T > 0$, obtained using the numerical method based on the density-matrix renormalization group (DMRG) approach [13]. At high $T \geq J$, distribution of s reveals a modest but finite width qualitatively similar both for the isotropic and the XX chain. On the other hand, the low-T variation established numerically is essentially different. While for the XX chain there is no significant T dependence, results for the isotropic case reveal at low $T \ll J$ a very large span of s values and corresponding T_{1n}, qualitatively and even quantitatively consistent with NMR experiments [6].

We study in the following the 1D spin-1/2 model representing the AFM RHC,

$$H = \sum_i J_i (S_i^x S_{i+1}^x + S_i^y S_{i+1}^y + \Delta S_i^z S_{i+1}^z), \qquad (1)$$

where J_i are random and we will assume their distribution as uncorrelated and uniform in the interval $J - \delta J \leq J_i \leq J + \delta J$, with the width $\delta J < J$ as the parameter. In the following we will consider predominantly the isotropic case $\Delta = 1$, but as well the anisotropic XX case with $\Delta = 0$. The chain is of the length L with open boundary

conditions as useful for the DMRG method. We further on use $J = 1$ as the unit of energy as well as $\hbar = k_B = 1$.

Our aim is to analyze the local spin dynamics in connection with the NMR spin-lattice relaxation [6]. In a homogeneous system the corresponding relaxation rate $1/T_1$ is expressed in terms of the q-dependent spin correlation function,

$$\frac{1}{T_1} = \sum_{q\alpha} A_\alpha^2(q) S^{\alpha\alpha}(q, \omega \to 0), \tag{2}$$

where $A_\alpha^2(q)$ involve hyperfine interactions and NMR form factors [6]. In the Supplemental Material [14] we show that the dominant dynamical $\omega \to 0$ contribution at low T is coming from the regime $q \sim \pi$. Therefore the variation $A_\alpha^2(q)$ is not essential and the rate depends only on the local spin correlation function $1/T_1 \propto S^{zz}_{\mathrm{loc}}(\omega \to 0)$. In a system with quenched disorder the relaxation time becomes site dependent, i.e., T_{1n}; hence, we study in the following the local correlations $S_{nn}(\omega)$ and the distribution of local limits $s = S_{nn}(\omega \to 0)$ and related relaxation times $\tau = 1/s$ where

$$S_{nn}(\omega) = \frac{1}{\pi} \mathrm{Re} \int_0^\infty dt e^{\imath\omega t} \langle S_n^z(t) S_n^z(0) \rangle. \tag{3}$$

In order to reduce finite-size effects we study large systems employing the finite-temperature dynamical DMRG (FTD-DMRG) [13,15,16] method to evaluate the dynamical $S_{nn}(\omega)$, Eq. (3). To reduce edge effects we choose the local site n to be in the middle of the chain, $n = L/2$. The distribution of s is then calculated with $N_r \sim 10^3$ different realizations of the system with random J_i. More technical detail on the calculation can be found in the Supplemental Material [14].

We start the presentation of the results with typical examples of $S_{nn}(\omega)$. In Fig. 1 we show calculated spectra for a system with $L = 80$ sites, $T = 0.5$, $\Delta = 1$, and three different realizations of J_i, i.e., the homogeneous system with $J_i = 1$ and two configurations with $\delta J = 0.7$. Spectra for the uniform system are broad and regular at $\omega \sim 0$, agreeing with those obtained with other methods [17], while $S_{nn}(\omega)$ for random case strongly depend apart from δJ also on the local J_i, $i \sim n$. In particular, spectra with both J_{n-1} and J_n small have a large amplitude at the relevant $\omega \sim 0$, while spectra with one large J_{n-1} or J_n have most of the weight at high-ω and a small amplitude at $\omega \sim 0$ (elaborated further in the conclusions). For the following analysis it is important that $s = S_{nn}(\omega \to 0)$ can be extracted reliably.

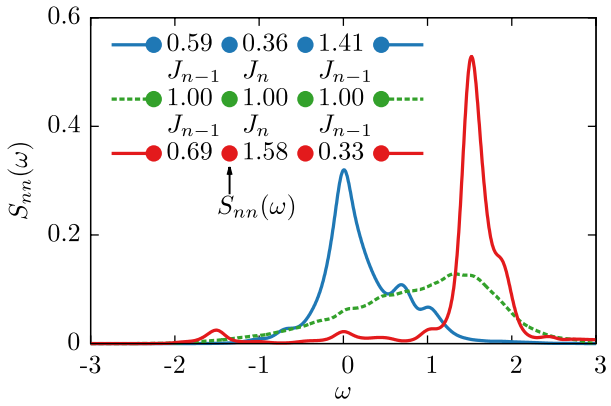

FIG. 1 (color online). Dynamical local spin correlations $S_{nn}(\omega)$ for different configurations of J_i. Shown are spectra for the homogeneous case $\delta J = 0$ and two configurations with $\delta J = 0.7$, calculated for $T = 0.5$ and $L = 80$ sites.

Results for $T \geq J$.—Before displaying results for the most interesting $T < J$ regime, we note that even at $T \gg J$ one cannot expect a well-defined $\tau = \tau_0$ but rather a distribution of values. One can understand this by studying analytically local frequency moments within the high-T expansion and using Mori's continued fraction representation [18] with the Gaussian-type truncation level of $l > 3$ [19,20] (see [14] for more details). In the inset of Fig. 2 we present the high-T result for the probability distribution function of s PDF(s) and compare it with the numerical results evaluated for $T = 1$. Several conclusions can be drawn from the results presented in Fig. 2. (a) The agreement of PDF(s) obtained via the analytical approach and numerical FTD-DMRG method is satisfactory having the origin in quite broad and featureless spectra $S_{nn}(\omega)$ at $T \geq J$. Still we note that median values of s (s_{med}) differ between both approaches and that for $T \gg J$ (unlike $T \leq J$) contribution of $q \to 0$ can become essential [14,21]. (b) PDF(s) becomes quite asymmetric and broad for $\delta J \geq 0.5$. (c) Consequently, also the distribution of local relaxation times PDF(τ) has a finite but modest width for $T \to \infty$. This seems to be in qualitative agreement with NMR data for $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$, where the width was hardly detected at high T [6].

Results for $T < J$.—More challenging is the low-T regime which we study using the FTD-DMRG method

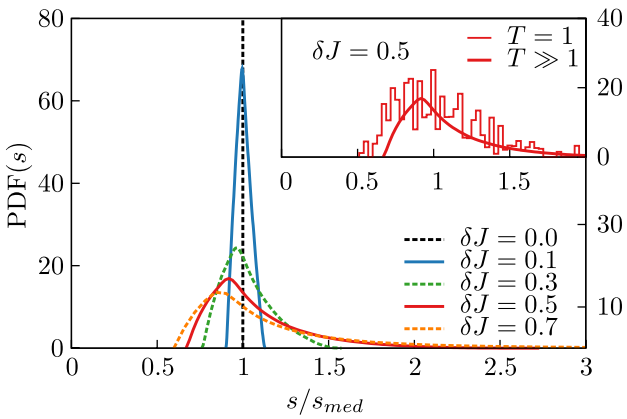

FIG. 2 (color online). Probability distribution function of local relaxation rates PDF(s) at $T \gg 1$ evaluated using the moment expansion for different δJ. Inset: Comparison of the analytical and FTD-DMRG result for $\delta J = 0.5$, $L = 20$ with a full basis and averaged over $N_r = 10^3$ realizations.

for typically $L = 80$ and $N_r \sim 10^3$. Besides the isotropic case ($\Delta = 1$), we also investigate for comparison the XX model ($\Delta = 0$). As the model of NI fermions with the off-diagonal disorder [10,11], it can be easily studied via full diagonalization on much longer chains with $L \sim 16\,000$. The PDF for $T < J$ can become very broad and asymmetric. Hence, we rather present results as the cumulative distribution function $\mathrm{CDF}(x) = \int_0^x dy \mathrm{PDF}(y)$. Further we rescale x values to the median defined as $\mathrm{CDF}(x_{\mathrm{med}}) = 0.5$. Results for CDF($s$) are presented in Fig. 3. Note that $\mathrm{PDF}(\tau) = \mathrm{PDF}(s)/\tau^2$. Panels in Fig. 3 represent results for the isotropic case $\Delta = 1$ with (a) fixed $T = 0.2$ and varying $\delta J = 0.1$–0.9, while in (b) $\delta J = 0.7$ is fixed and $T = 0.1$–0.5. The inset of Fig. 3(b) displays the T dependence (for fixed $\delta J = 0.7$) of the CDF for the XX chain.

We first note that within the XX chain CDF(s) are essentially T independent. This appears as quite a contrast to, e.g., static $\chi^0(T)$ which exhibits a divergence at $T \to 0$ [4,14]. Results for the isotropic case $\Delta = 1$ in Figs. 3(a) and 3(b) are evidently different. The span in the CDF becomes very large (note the logarithmic scale) either by increasing δJ at fixed T or even more by decreasing T at fixed δJ. From the corresponding PDF one can calculate the relaxation function $R(t) = \int ds \mathrm{PDF}(s) e^{-ts}$, which is in fact the quantity measured in the NMR as a time-dependent magnetization recovery [6]. As in experiment, the large span in our results for low T can be captured by a stretched exponential form, $R(t) \approx \exp[-(t/\tau_0)^\Gamma]$, where Γ and τ_0 are parameters to be fitted for a particular PDF(s) and corresponding $R(t)$. It is evident that $\Gamma \ll 1$ means large deviations from the Gaussian-like form, and in particular very pronounced tails in the PDF(s), both for $s \gg s_{\mathrm{med}}$ as well as a singular variation for $s \to 0$. In the latter regime $1/\tau_0$ can deviate substantially from the average of local $1/\tau$. It should be also noted that the stretched exponential form, is the simplest one capturing the large span of s values. It is also used in the experimental analysis [6], but the corresponding PDF(s)s reveal somewhat enhanced tails for $s > s_{\mathrm{med}}$ relative to calculated ones in Figs. 3(a) and 3(b), and the opposite trend for $s < s_{\mathrm{med}}$. This suggests possible improvements and a description beyond the stretched exponential form, which we leave as a future challenge. More details can be found in the Supplemental Material [14].

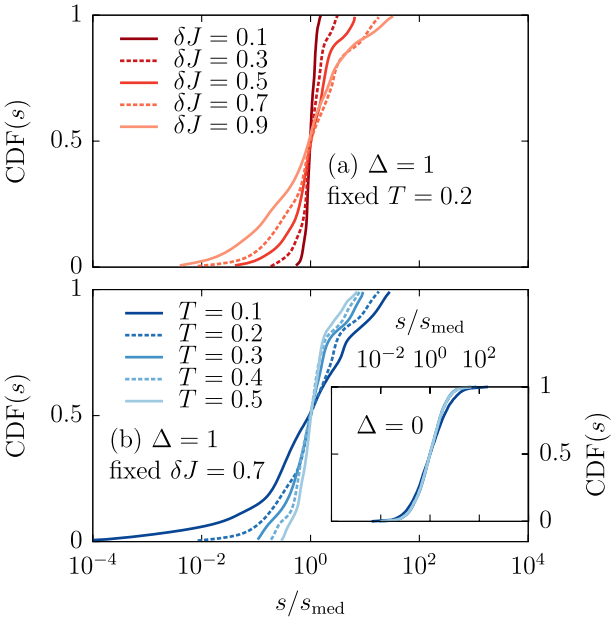

FIG. 3 (color online). Cumulative distribution function of s. Shown are FTD-DMRG results for $\Delta = 1$: (a) for fixed $T = 0.2$ and various δJ, (b) for fixed $\delta J = 0.7$ and various $T \leq 0.5$. Inset of (b): full diagonalization results for $\Delta = 0$, $\delta J = 0.7$ and various T.

Results for the fitted exponent $\Gamma(T)$ for $\Delta = 1$ as extracted from numerical PDF(s) for various δJ are shown in Fig. 4(a). They confirm experimental observation [6] of increasing deviations from simple exponential variation ($\Gamma = 1$) for $T \ll J$. While for $T > J$, $\Gamma \lesssim 1$ for modest $\delta J < 0.7$, low-T values can reach even $\Gamma < 0.5$ at lowest reachable $T < 0.1$. Note that in such a case values of s are distributed over several orders of magnitude.

Of interest for the comparison with experiment is also the T variation of fitted $1/\tau_0$. Results are again essentially different for $\Delta = 0$ and $\Delta = 1$. τ_0 (as well s_{med}) for $\Delta = 0$ follows well the Korringa law $1/\tau_0 \propto T$ for $T < 0.5$, as usual for the system of NI fermions with a constant density of states (DOS) (divergent DOS at $E \to 0$ could induce a logarithmic correction). On the other hand, for the isotropic ($\Delta = 1$) chain with no randomness $\tau_0 = \tau$ it should follow $1/\tau \sim$ const for $T < J$ [17,22]. Similar behavior is

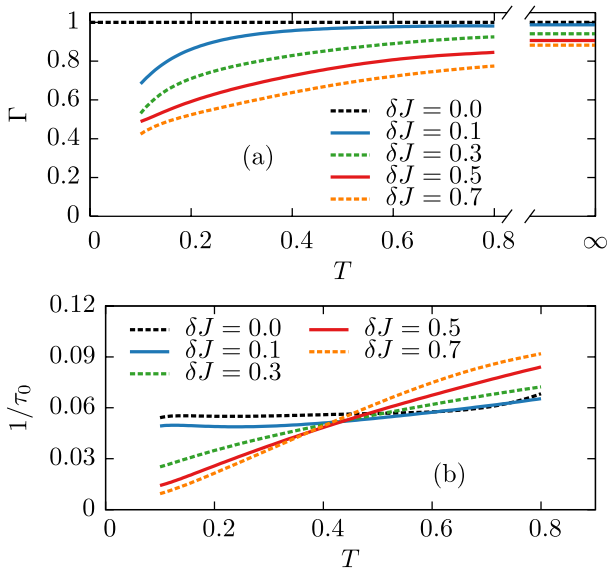

FIG. 4 (color online). (a) Exponent Γ vs T obtained from PDF(s) data for different δJ and isotropic case $\Delta = 1$. (b) T dependence of fitted $1/\tau_0$ for $\Delta = 1$ and different δJ.

observed for weak disorder $\delta J = 0.1$ as shown in Fig. 4(b). However, with increasing randomness δJ, $1/\tau_0$ becomes more T dependent and increases with T. Such T dependence in the RHC of $1/\tau_0$ is, although in agreement with experiment, in apparent contrast with diverging $\chi^0(T \to 0)$. This remarkable dichotomy between static and dynamical $\omega \to 0$ behavior can be reconciled by the observation that in a random system $S(q, \omega \sim 0)$ reveals, besides the regular par, also a delta peak at $\omega = 0$ (not entering $1/T_1$), which can be traced back to diagonal matrix elements [14] being an indication of a nonergodic behavior (at least at low T). Note that more frequently studied static $S(q)$ (equal-time correlation) [23] represents a sum rule containing both parts. Also, the relation $\chi^0(T) = S(q = 0)/T$ in spite of divergent $\chi(T \to 0)$ leads to vanishing $S(q = 0)$ at $T \to 0$ only slower than linearly [14,23].

As a partial summary of our results, we comment on the relation to the experiment on $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$ [5,6]. The spin chain is in this case is assumed to be a random mixture of two different values $J_i = 280$ K, 580 K, which correspond roughly to our $\delta J \simeq 0.6$ (fixing the same effective width) and $J = 430$ K. Taking these values, our results for $\Gamma(T)$ as well as $1/\tau_0(T)$ agree well with experiment. In particular we note that at lowest $T \ll J$ our calculated $\Gamma \sim 0.5$ for $\delta J = 0.6$ matches the measured one. There appears to be the discrepancy of a steeper increase of measured $\Gamma(T)$ towards the limiting $\Gamma = 1$ coinciding with observed very narrow PDF(τ) which remains of finite width in our results even for $T \to \infty$ as seen in Fig. 2. As far as calculated $1/\tau_0(T)$ vs NMR experiment is concerned, we note that taken into account, the normalization of average J disordered system reveals at $T \to 0$ smaller $1/\tau_0$ than a pure one consistent with the experiment [6]. In agreement with the experimental analysis is also the strong T variation of $1/\tau_0$ at low T in a disordered system in contrast to a pure one.

Our results on the local spin relaxation $S_{nn}(\omega)$ and in particular its T dependence cannot be directly explained within the framework of existing theoretical studies and scaling approaches to RHCs [2,3,9]. Our study clearly shows the qualitative difference in the behavior of the XX chain and the isotropic RHC. While in the former model mapped on NI electrons, T does not play any significant role on PDF(s) as seen in the inset of Fig. 3(b), the $\Delta = 1$ case shows strong variation with $T \ll J$. It is plausible that the difference comes from the interaction and many-body character involved in the isotropic RHC. To account for that we design, in the following, a simple qualitative argument.

The behavior of $S_{nn}(\omega \sim 0)$ at low T is dominated by transitions between low-lying singlet and triplet states which become in a RHC nearly degenerate following the scaling arguments with effective coupling $\tilde{J} \to 0$ for more distant spins and reflected in diverging $\chi^0(T \to 0)$ [2–4,14]. Such transitions are relevant at the $\omega \to 0$ behavior as presented in Fig. 1. Moreover, local $S_{nn}(\omega \sim 0)$ exhibit a large spread due to the variations in the local environment. Let us for simplicity consider the symmetric Heisenberg model on four sites (with open boundary conditions) with a stronger central bond $J_2 \gg J_1 = J_3$ and $J = (J_1 + J_2 + J_3)/3$. It is then straightforward to show that the lowest singlet-triplet splitting is strongly reduced, i.e., $\Delta E \propto \eta^2 J$ where $\eta = J_1/J_2$. Within the same model one can also evaluate the ratio between two different amplitudes of $S_{nn}(\omega \sim \Delta E) = A_{nn}\delta(\omega - \Delta E)$, on sites $n = 1, 2$ neighboring the weak and strong bond,

$$\frac{1}{W} = \frac{A_{22}}{A_{11}} = \frac{|\langle \Psi_t | S_2^z | \Psi_s \rangle|^2}{|\langle \Psi_t | S_1^z | \Psi_s \rangle|^2} \sim \eta^2. \tag{4}$$

The relation shows that the span between the largest and smallest amplitudes increases as $W \propto 1/\eta^2 \propto 1/\Delta E$. Continuing in the same manner the scaling procedure for a long AFM RHC [2,3] the smallest effective coupling between further spins $\tilde{J}$ vanishes at $T = 0$ and $\Delta E \propto \tilde{J} \to 0$, so that one expects $W \to \infty$ for $T \to 0$. On the other hand, for $T > 0$ the scaling should be cut off at $\tilde{J} \sim T$ at least for $\Delta = 1$, finally leading to the strong $W(T)$ dependence ($W \propto 1/T$).

In summary, we have reproduced qualitatively main experimental NMR results on a mixed system $BaCu_2(Si_{0.5}Ge_{0.5})_2O_7$ including anomalously wide distribution of relaxation rates, together with T dependencies of experimental parameters ($1/\tau_0$, Γ) and provided a microscopic explanation with the help of the random-singlet framework. Our qualitative conclusions on the RHC do not change by changing S^z_{tot} (adding finite field in the fermionic language) or even reducing $\Delta < 1$ provided that $\Delta > 0$ (see Supplemental Material [14]). We also comment on the striking difference between static and dynamic quantities and observed deviations from stretched exponential phenomenology.

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