Anomalous Spin Dynamics in Doped Quantum Antiferromagnets

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Finite-temperature spin dynamics in the planar t-J model are studied using a method based on the Lanczos diagonalization of small systems. The dynamical spin structure factor at moderate dopings shows the coexistence of free-fermion-like and spin-fluctuation time scales. At $T < J$, low-frequency and static susceptibilities show a pronounced T dependence, supporting a scenario related to the marginal Fermi liquid one, for the explanation of neutron-scattering and NMR relaxation experiments in cuprates. Calculated NMR relaxation rates reasonably reproduce experimental ones.

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The understanding of spin and charge dynamics in strongly correlated systems, as realized in cuprates on doping the reference antiferromagnetic (AFM) insulator, still represents a challenge for theoreticians. The low-frequency spin dynamics and static spin response in the undoped and the doped AFM state in cuprates have been recently extensively studied by neutron scattering [1] and in a number of NMR and NQR experiments [2,3]. They established that normal-state spin dynamics differs qualitatively from the one expected for Landau Fermi liquids. Generically, the NMR and NQR spin-lattice relaxation time T_1 is nearly T and doping independent in the normal state $T > T_c$ [3], in contrast to the Korringa law $T_1^{-1} \propto T$ in normal metals. Also, in the same regime the low-ω dynamical susceptibility in doped systems appears to be consistent with $\chi''(\omega) \propto \omega/T$ [1].

Well understood so far are only undoped cuprates, which behave in all respects as isotropic quantum AFM's, with long range order at $T = 0$. For doped systems NMR and NQR data on the spin dynamics have been interpreted within the phenomenological model of AFM correlated spins [4,5]. Here the T dependence is attributed to the variation of the AFM correlation length $\xi(T)$ [4]. At low hole doping, the dynamics has been mapped on the related quantum-critical (QC) scaling regime of the nonlinear sigma model where $\xi \propto 1/T$ [6]. An alternative scenario for the low-ω, low-T behavior has been given in terms of an anomalous T dependence [not directly related to $\xi(T)$], introduced within the marginal Fermi liquid hypothesis [7].

Among the microscopic models the most helpful for the discussion of spin dynamics in strongly correlated metals is the t-J model [8]

$$H = -t \sum_{\langle ij \rangle s} (c_{js}^{\dagger} c_{is} + \text{H.c.}) + J \sum_{\langle ij \rangle} (\vec{S}_i \cdot \vec{S}_j - \tfrac{1}{4} n_i n_j), \quad (1)$$

where $c_{is}^{\dagger}$ (c_{is}) are projected fermionic operators, taking into account that the double occupancy of sites is not allowed. In spite of its simple form the model proved to be very difficult to analyze, analytically [8] as well as numerically [9]. The most reliable results related to the spin dynamics have been obtained by exact diagonalization studies [9–11] of small systems, and by means of the high-temperature series expansion [12,13]. These methods, however, have not been able so far to get results for the low-ω and dc spin response in the most challenging parameter regime $J < t$ and at intermediate $0 < T < J$.

Recently, the present authors introduced a new numerical method, based on the Lanczos diagonalization of small systems combined with random sampling over basis states [14], which allows the study of dynamical and dc response functions at $T > 0$. The method has already been applied to the evaluation of the optical and dc conductivity in the t-J model on the square lattice [15].

In this paper we present results, obtained by the same method, for the dynamical spin structure factor (we later choose units with $\hbar = k_B = 1$),

$$S(\vec{q}, \omega) = \text{Re} \int_0^{\infty} dt\, e^{i\omega t} \langle S_{\vec{q}}^z(t) S_{-\vec{q}}^z(0) \rangle, \quad (2)$$

and the related dynamical susceptibility $\chi(\vec{q}, \omega)$,

$$\chi''(\vec{q}, \omega) = (1 - e^{-\beta\omega}) S(\vec{q}, \omega). \quad (3)$$

Since the numerical requirements are the same as for the conductivity problem, we refer to the description of technical details in Refs. [14,15]. We study a planar system of $N = 4 \times 4 = 16$ sites with $J/t = 0.3$ and variable doping, i.e., with $N_h = 0-10$ holes. Typically we use up to $M = 120$ Lanczos steps and sampling over $N_0 \sim 300$ states. The method has been tested with the full-diagonalization results for $N = 10$ [10]. In comparison with the latter results our discrete spectra obtained for $N = 16$ are much more dense, so that even minimal additional broadening (with a characteristic width $\eta \ll J$) yields smooth macroscopiclike spectra. Generally, the results become unrealistic at low temperatures $T < T^*$, where finite-size effects start to introduce size-dependent features. T^* is related to the average level distance in the low-energy sector. The latter is smallest (for fixed N) in the intermediate-doping regime $0.12 < c_h < 0.5$,

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where we reach $T^* \sim 0.1t$. The low-energy sector becomes more sparse both in the low-electron $c_h > 0.5$ and in the pure AFM regime $c_h < 0.12$, leading to an increase of T^*. The onset of finite-size effects is monitored by the appearance of unphysical structures in the high-frequency spectra, by the dependence on smoothing width η, etc. We choose further on predominantly $\eta = 0.07t$.

We present in Fig. 1 $S(\vec{Q}, \omega)$ spectra at the AFM wave vector $\vec{Q} = (\pi, \pi)$ and fixed $T = 0.2t < J$, but various hole concentrations c_h. As noted above, moderate doping results are reliable in this $T < J$ regime, while low-doping spectra already exhibit finite-size effects, e.g., gaps appearing in spectra, etc. The most interesting feature in Fig. 1 is the qualitative change of the spectra on doping. Whereas at $c_h < 0.12$ the spectra are dominated by a single central peak with the width $\omega \sim 2J$ due to the AFM fluctuations, in the intermediate regime $0.12 < c_h < 0.5$ *a high-frequency component with* $\omega \sim t$ *emerges, coexisting with the remaining low-frequency fluctuations.* It is plausible to attribute the high-ω dynamics to the free-fermion-like component of the correlated system, in particular, since it appears to be quite independent of J (provided that $J < t$) and tends to exhaust the spectra in the overdoped cases $c_h > 0.5$. Although this observation is not unexpected [10,16], the coexistence of spin-fluctuation and free-fermion time scales at "optimum" doping has been established here for the first time. That is, it is harder for other methods, e.g., within the high-T expansion method, to treat coexisting different time scales. The dual character is a crucial property, since the free-fermion part determines to a large extent the static spin correlations $S(\vec{q})$ [and charge density correlations $N(\vec{q})$], interpreted in terms of a quasi Fermi surface [16]. On the other hand, the low-ω spin dynamics dominates dynamical and static spin susceptibilities, $\chi''(\vec{q}, \omega)/\omega$ and $\chi(\vec{q})$, respectively, and hence also the neutron-scattering and the NMR processes.

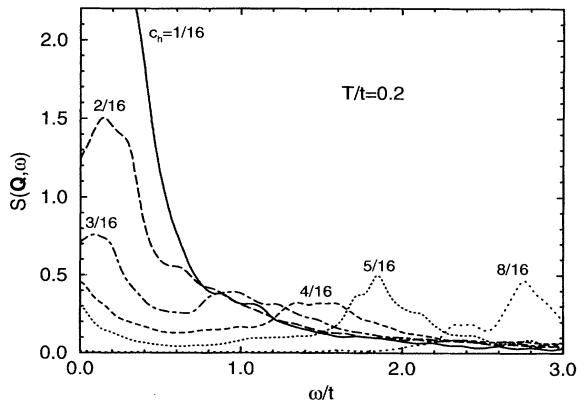

FIG. 1. Dynamical spin structure factor $S(\vec{q} = \vec{Q}, \omega)$ at various hole dopings and fixed $T = 0.2t < J$. Spectra for a planar 16-site system are broadened with $\eta = 0.07t$.

Figure 2 displays dynamical spectra $\chi''(\vec{q}, \omega)/\omega$ for fixed $c_h = 3/16$, but various T and nonequivalent $\vec{q}$. Note that on the 4×4 lattice $\vec{q} = (0, \pi)$ and $\vec{q} = (\pi/2, \pi/2)$ are equivalent. In contrast to Fig. 1, high-ω features are suppressed here. Nevertheless, the free-fermion part is well separated from the low-ω part only for $\vec{q} \sim \vec{Q}$, for which one expects a gap in the response of free fermions with a well-defined Fermi surface. For other $\vec{q}$ (at given low doping) the free-fermion contribution persists at larger $\omega > J$ in the form of a long tail, while in the low-ω regime it merges with the spin contribution. The most striking feature of Fig. 2 is, however, *the strong T dependence of low-*ω *spectra, whereas at the same time the AFM correlation length* ξ *is only weakly T dependent.* Here, we can calculate $\xi(T)$ from the static real space spin correlations $S(\vec{r})$ [the Fourier transform of $S(\vec{q}) = \langle S^z_{\vec{q}} S^z_{-\vec{q}} \rangle$], i.e., $\xi^2 = \sum_i |\vec{r}_i|^2 \exp(i\vec{Q} \cdot \vec{r}_i) S(\vec{r}_i)/4S(\vec{Q})$. In the most interesting regime $c_h = 0.1-0.3$, we find that ξ is short, typically $\xi \sim 1$, governed mainly by correlations at $r_i = 1$. In particular, for $N_h = 2, 3$ ξ increases less than 30% between $T = J$ and $T = J/3$, in agreement with high-T expansion studies [12], as well as with Monte Carlo studies of the Hubbard model [17] and with values $\xi(T = 0)$ obtained via exact diagonalization within the t-J

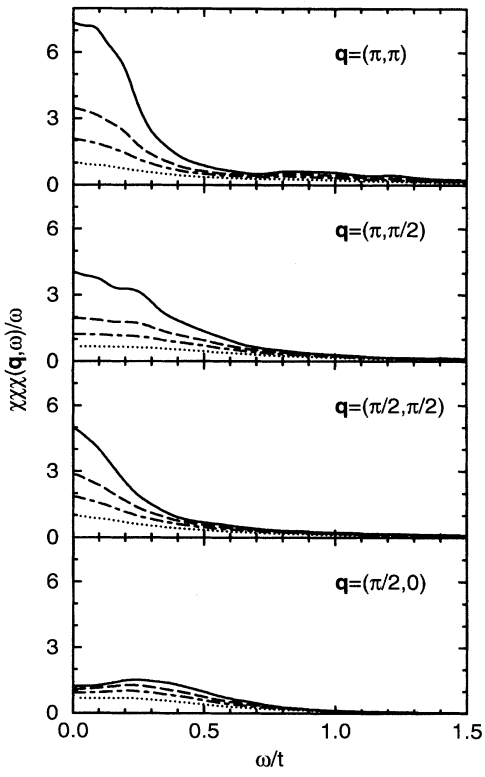

FIG. 2. Dynamical spin susceptibility $\chi''(\vec{q}, \omega)/\omega$ at fixed doping $c_h = 3/16$ for different $\vec{q}$ in the Brillouin zone and various temperatures: $T/t = 0.1$ (full line), 0.2 (dashed line), 0.3 (dash-dotted line), and 0.5 (dotted line).

model [9]. Similar values for ξ can also be extracted considering our static $\chi(\vec{q})$.

The above conclusions on $\chi''(\vec{q},\omega)/\omega$ seem to hold for all $|\vec{q}-\vec{Q}| < \alpha\xi^{-1}$, where $\chi(\vec{q}) \sim \chi(\vec{Q})$. The relevant volume in $\vec{q}$ space clearly increases on doping and exhausts already for $c_h = 3/16, 4/16$ the majority of the Brillouin zone, while within the same doping regime scaling does not hold, e.g., for $\vec{q} = (0, \pi/2)$. The variation at $\omega < J$ is $\chi''(\vec{q},\omega)/\omega \propto 1/T$, or, *equivalently, $S(\vec{q},\omega)$ is nearly T and ω independent* in the same regime, as also observed in the neutron-scattering experiments [1], where $\chi''(\omega) \propto \omega/T$. It should be noted that the universal scaling $\chi''(\omega) = f(\omega/T)$, claimed by several authors [1,17], seems to be close to the requirement of $S(\vec{q},\omega) \sim$ const.

The spectra discussed above have as a direct consequence the T variation of the static $\chi(\vec{q})$ also at $T < J$. We observe a pronounced T dependence, e.g., $\chi(\vec{q}) \propto T^{-1}$ for $c_h = 3/16$, in a wide regime $J/3 < T < t$ for all q within the correlation volume. It should be noted, however, that we are quite restricted in the range of T/J, so that more quantitative conclusions on a possible power-law (or logarithmic) variation with T are not feasible.

We can discuss our results in relation to experimental ones obtained in cuprates via NMR and NQR relaxation. The NQR spin-lattice relaxation time T_1 and the spin-echo decay time T_2 for ^{63}Cu nuclei are related to electronic spin susceptibilities by [3,4]

$$\frac{1}{T_1} = \frac{2T}{g^2\mu_B^2}\frac{1}{N}\sum_{\vec{q}}{}' A_\perp^2(\vec{q})\frac{\chi''(\vec{q},\omega_0)}{\omega_0}, \qquad \omega_0 \to 0,$$

$$\frac{1}{T_2} = \sqrt{\frac{0.69}{8}}\frac{1}{g^2\mu_B^2}\left[\frac{1}{N}\sum_{\vec{q}} A_\parallel^4(\vec{q})\chi^2(\vec{q}) - \left(\frac{1}{N}\sum_{\vec{q}} A_\parallel^2(\vec{q})\chi(\vec{q})\right)^2\right]^{1/2}. \quad (4)$$

Note that in the evaluation of T_1 within a finite system we have to omit the $q = 0$ term in the sum [11] due to the divergent (ill-defined) $\chi''(\vec{q},\omega)/\omega$, $\omega \to 0$, related to the conservation of total S_z. A proper treatment would require an independent evaluation of the $q \sim 0$ spin-diffusion contribution, which, however, seems to be less important at least for the undoped system [11]. To allow a direct comparison with experiments we choose $A_\perp(\vec{q}), A_\parallel(\vec{q})$ as proposed in the literature [4] and $t = 0.4$ eV [8]. Again, $J = 0.3t = 0.12$ eV .

Results for T_1 are presented in Fig. 3(a). For the undoped case our results for T_1 agree with Ref. [11], but the T variation at $T < J$ is already influenced by finite-size effects (due to sparse density of states at low energies). It is remarkable that T_1 appears to be nearly T independent for a broad range of hole concentrations $0.06 < c_h \leq 0.31$ ($c_h = 6/16$ is not included due to more pronounced finite-size effects caused by the closed-shell configuration). Only for the overdoped systems with $c_h \geq 0.5$ does the behavior at $T < t$ approach that of a normal Fermi liquid with $T_1^{-1} \propto T$. Our results are in agreement, even quantitatively without any fitting parameters, with recent remarkable NQR experiments on $La_{2-x}Sr_xCuO_4$ [3], which reveal nearly T- and x-independent T_1 at $T > 300$ K and $x < 0.15$. We, in fact, establish a variation of T_1 with doping, which, however, becomes more pronounced only for $c_h \geq 2/16$. Although not essential, the variation of the calculated T_1^{-1} in the range $c_h = 0-2/16$ could possibly be an artifact either due to finite size effects or due to the omission of the spin-diffusion contribution. We should also stress that for quantitative comparison with cuprates the simplest t-J model cannot be regarded as the complete model. Anyhow, for the optimum doping lower rates T_1^{-1} are expected, consistent with the data for $YBa_2Cu_3O_7$ [2], where, for $T > T_c$, T_1 is again found to be only weakly dependent on T.

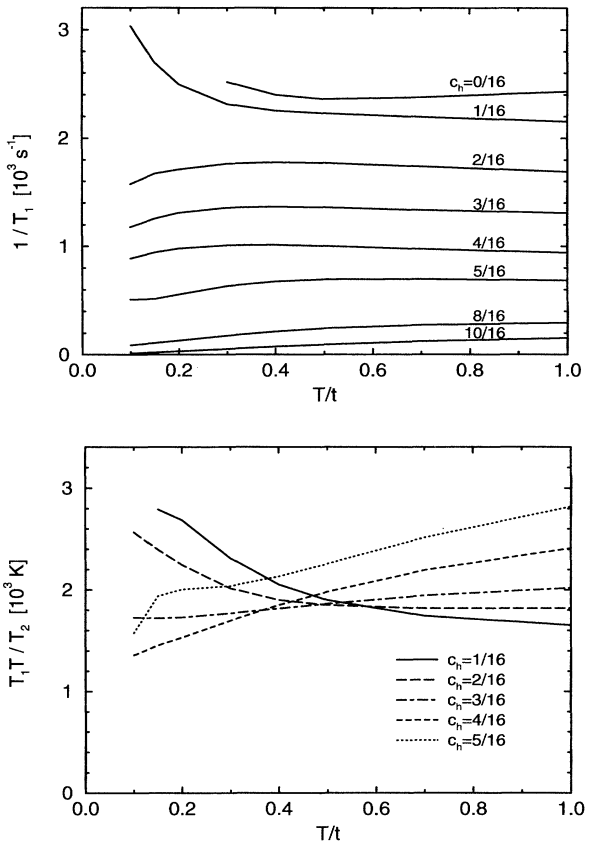

FIG. 3. (a) NQR spin-lattice relaxation rate $1/T_1$ and (b) the ratio T_1T/T_2 both vs T/t for various dopings c_h. Note that here $t = 0.4$ eV $= 4640$ K.

The temperature-independent ratio $R = T_1T/T_2$, as approximately realized in cuprates, has been interpreted as evidence for the QC behavior of the effective spin system [6]. We find quite analogous weak T variation of the ratio within the t-J model, with results presented in Fig. 3(b).

Here the undoped case is omitted due to inaccurate (finite-size dominated) results for $\chi(\vec{Q})$, and, consequently, for T_2, obtained on a small system for $T < J$. The origin of $R(T) \sim$ const is, however, considerably different from the QC scenario, since the $T_2(T)$ dependence does not seem to be connected (in an evident way) with the $\xi(T)$ variation. The results indicate a stronger doping dependence, even at low doping. Quantitatively, the values obtained are in reasonable agreement with experimental ones, e.g., $R \sim 1700$ K at $T = 300$ K for $YBa_2Cu_3O_7$ ($c_h \sim 0.23$), while $R \sim 2400$ K for $YBa_2Cu_3O_{6.63}$ [6].

Essentially different T variation of the T_1 relaxation on Cu and O nuclei, respectively, has been used as evidence for the importance of strong (AFM) correlations and non-Fermi-liquid behavior in doped cuprates. To evaluate the NMR T_1^{-1} for ^{17}O we can again use Eq. (4) with a modified form factor [4], projecting out the AFM fluctuations at $\vec{q} \sim \vec{Q}$. The omitted $q \sim 0$ contribution introduces in this case a larger uncertainty. Nevertheless, for $c_h = 1/16, 2/16$ and $T^* < T < J$ we recover results very well described with the Korringa behavior, i.e., $(^{17}T_1T)^{-1} \sim C$. In particular, at $c_h = 2/16$ we get $C \sim 0.3$, very close to the actual value $C \sim 0.35$ as reported for $YBa_2Cu_3O_7$ [2]. For $c_h \geq 3/16$ deviations from the Korringa law become more pronounced due to very short ξ.

In conclusion, we have presented results for the dynamical spin susceptibility obtained within the t-J model with a new numerical method, for the first time in the challenging regime of low to moderate doping with $J < t$, as well as $T, \omega \lesssim J$. The most interesting finding is the anomalous low-frequency spin dynamics, showing up also in the T dependence of static susceptibilities, which is related to the NMR relaxation times T_1 and T_2, as well as to the neutron scattering experiments. Our results are consistent with nearly T- and ω-independent $S(\vec{q}, \omega)$ [or $\chi''(\vec{q}, \omega) \propto \omega/T$] for $\vec{q}$ around $\vec{Q}$, even in the regime where $\xi \neq \xi(T)$, but still well in the normal state. Such T dependence is close to the concept of the marginal Fermi liquid [7]. A similar concept has also been discussed in [17].

It is tempting to speculate on the origin of the anomalous low-ω spin dynamics. It is quite plausible to relate it to the dramatic increase of the density of low-energy many-body states emerging on doping. The latter show up in substantially enhanced entropy at low T [18], and in the strong carrier scattering as manifested in the resistivity $\rho \propto T$, established recently also within the t-J model [15]. An intuitive picture might be that at low $T < J$ spin clusters with the characteristic size $l \sim \xi(T)$ behave as nearly independent, whereby their interaction is effectively blocked by doped holes, thus leading to a large density of low-ω modes. A more coherent theoretical description is clearly missing so far. Our study shows that the t-J model still remains a promising starting point for these investigations.

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