Does Frustration Describe Doping in Models for High-Temperature Superconductors?

Franco Nori,(1) Eduardo Gagliano,(2) and Silvia Bacci(2)

(1)*Physics Department, University of Michigan, Ann Arbor, Michigan 48109-1120*

(2)*Physics Department, Materials Research Laboratory, and Science and Technology Center for Superconductivity, University of Illinois, Urbana, Illinois 61801*

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We compare predictions of frustrated and doped Hamiltonians by studying the t-J model with one hole and the frustrated Heisenberg J-J' model. In particular, we compare for both systems the dynamic spin-spin structure factor $S(\mathbf{q},\omega)$, and the B_{1g} Raman scattering spectrum $R(\omega)$ at zero temperature. We observe that, for the t-J model, both quantities are in qualitative agreement with experimental measurements while the corresponding ones for the J-J' model are not. These results indicate that doped systems cannot be accurately modeled by a purely spin model and that the latter, including the chiral spin states, might not be relevant to describe measurable quantities of the high-T_c materials.

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The relationship between doping and antiferromagnetism is a central issue in high-temperature superconductors. When undoped, these materials display conventional Néel order which can be described by quantum Heisenberg models [1]. When doped, the long-range antiferromagnetic order is suppressed and superconductivity appears. These facts have been modeled by the t-J Hamiltonian constrained to the subspace with no double occupancy which, in standard notation, is given by

$$\mathcal{H} = -t \sum_{\langle i,j\rangle,\sigma} (1-n_{j-\sigma}) c_{j\sigma}^{\dagger} c_{i\sigma} (1-n_{i-\sigma}) + J\sum_{\langle i,j\rangle} (\mathbf{S}_i\cdot\mathbf{S}_j - \tfrac{1}{4} n_i n_j)\,. \quad (1)$$

This model, which describes the effect of the hole motion on the spin background, has been derived in the large-U limit of a single band Hubbard model. In this case, J favors Néel order, while t tends to disrupt it.

In the past four years, there has been a renewed interest in the study of frustrated magnetic systems [1–3]. The general assumption has been that frustrated spin models might be relevant in describing the high-T_c materials. However, in spite of extensive work on frustrated spin Hamiltonians, their relationship with either doped models or experiments remains unclear. It is the purpose of this work to explore these relationships.

Usually, extensive comparisons between different models, and experiments, are performed in order to assess the validity and relevance of their predictions. No such systematic comparisons have been performed among two of the most widely studied models since the discovery of the cuprous oxides: the t-J and J-J' systems. Recently, a study of this type, in the context of slave-boson generalized flux phases [4], has been presented [5].

Several works have suggested a possible connection between purely spin systems and doped ones. For example, it has been proposed [2] that the effect of doping can be described by including further neighbor couplings, J', into the quantum Heisenberg model,

$$\mathcal{H}_{J\text{-}J'} = J\sum_{\langle i,j\rangle} \mathbf{S}_i\cdot\mathbf{S}_j + J'\sum_{\langle i,k\rangle} \mathbf{S}_i\cdot\mathbf{S}_k\,, \quad (2)$$

the new couplings being proportional to the doping x. Another suggestion invokes an adiabatic continuation between doped and frustrated models. Other proposals have received widespread attention; for instance, the chiral spin states [6] have been derived under the explicit assumption [6] that frustrated spin models *effectively describe* doped systems. In this paper, we will not be concerned with a particular proposed link, but on the general issue of the relevance of frustrated spin Hamiltonians on the modeling of measurable quantities in the high-T_c materials.

The static magnetic structure factor $S(\mathbf{q})$ indicates that the effect of frustration and doping, on the *magnetic* properties, may be considered to be similar [1]. The reason is that in relation to the pure antiferromagnetic Heisenberg *both* of these models suppress the $S(\pi,\pi)$ peak, broadening and shifting this peak to another $\mathbf{q}$ point. The meaning of these results is clear: When we add a large enough perturbation to the spin-$\frac{1}{2}$ Heisenberg Hamiltonian, the ground state is bound to change from the (unperturbed) Néel state to some other (distorted) state. This result is not surprising. However, a more specific question is: How large must these perturbations be in order to destroy the Néel order? The critical values of the frustration parameter $(J'/J)_c$ and of the doping parameter x_c are $(J'/J)_c \sim 0.5$ and $x_c \leq 0.05$. Therefore, $(J'/J)_c \gg x_c$. This presents two problems: (i) While a very *small* amount of doping can destroy the long-range Néel order, a *large* amount of frustration is required to obtain the same result; and (ii) $(J'/J)_c$ is unphysically large since *ab initio* calculations [1] indicate that the physical $J'/J < 0.05$. We will present Raman and neutron spectra for both models in a broad parameter range, including the region where both are losing the antiferromagnetic order. The choice of Raman and neutron scattering is relevant since both have provided valuable information about magnetic properties.

Let us first consider Raman scattering [7]. The laser light incident on a magnetic insulator causes atomic motion and, therefore, a change in the distance between neighboring magnetic ions. This modulates the local exchange coupling and excites short-wavelength, high-energy spin excitations. The polarization dependence of the scattered light allows the magnetic signal to be separated from other contributions. In La_2CuO_4 and $YBa_2Cu_3O_6$, the only excitations between $\sim$0.1 and 1.5 eV are spin excitations because (i) these materials are insulators with a large charge excitation gap ($\sim$1.5 eV), and (ii) phonon excitations have energies below 0.1 eV. In the insulating phase, the Raman intensity, as a function of energy transfer, has a clear peak for certain "allowed" polarizations. The first moments of the spectrum are in good agreement with calculations based on the antiferromagnetic Heisenberg model. In the superconducting cuprates, the peak in the allowed polarizations *broadens* rapidly with doping.

The scattering Hamiltonian [8] which describes the interaction of light with the spin pair is

$$H_R = \sum_{\langle i,j \rangle} (\mathbf{E}_{\rm inc} \cdot \hat{\sigma}_{ij})(\mathbf{E}_{\rm sc} \cdot \hat{\sigma}_{ij}) \mathbf{S}_i \cdot \mathbf{S}_j \,. \tag{3}$$

Here $\mathbf{E}_{\rm inc}$ and $\mathbf{E}_{\rm sc}$ are the incident and scattered electric-field vectors of the photons and $\hat{\sigma}_{ij}$ is a unit vector connecting the spin sites i and j. The Raman scattering intensity $R(\omega)$ at $T=0$ is given by

$$R(\omega) = \int dt\, e^{i\omega t} \langle \psi_0 | H_R(t) H_R | \psi_0 \rangle \,,$$

where $|\psi_0\rangle$ is the ground state. To study $R(\omega)$ numerically we used a Lanczos method adapted to the evaluation of dynamic properties [9]. Using this approach we can evaluate exactly any response function as well as the moments of this distribution. All our results are obtained through exact diagonalization on a 4×4 lattice with periodic boundary conditions.

Depending on the orientation of the incident and scattered electric fields with respect to the crystal directions, different scattering geometries can be analyzed. One of the most studied modes is the B_{1g}. Figure 1 shows the B_{1g} Raman spectrum for the two-dimensional spin-$\frac{1}{2}$ (a) pure Heisenberg Hamiltonian, (b) t-J model with one hole, and (c) frustrated Heisenberg model. Here, we have selected some typical values of the parameters, however, other values of J'/J and t/J, not shown here, have also been considered. The dominant feature observed in the Raman spectra of Fig. 1 is the prominent two-magnon (2 nn spin flip) peak located at $\approx 3J$. The only effect of frustration [see Fig. 1(c)] is to continuously and rapidly *shift* the spectra to lower energies as a function of J' (approximately linearly for $J' \ll J$). This is in clear disagreement with the results for the doped case (either the t-J model or the experiments) since the frustrated case shows no low-frequency structure (below the prominent two-magnon peak), while the doped case exhibits a very rich structure at low energies. For the t-J model, these excitations are string states as well as two-magnon processes in the neighborhood of the hole [10]. In this model, the hole motion is responsible for an entirely new type of state (string type) located at the bottom of the spectrum. Furthermore, in the highly mobile limit, the intensity of the low-frequency part of the spectrum becomes dominant, since the intensity of the two-magnon peak, which is dominant for $t<J$, decreases as t/J increases. So, we conclude that the behavior of the two-magnon peak located at $\approx 3J$ for $J'=x=0$ is different in the two cases: Doping rapidly *suppresses* this peak, while frustrating the system continuously *shifts* the peak to lower energies and increases its intensity up to $J'/J \approx 0.5$. Furthermore, the uniform shift as a function of frustration exhibited by the J-J' model must be contrasted with the very complex structure that appears in the Raman spectrum as a function of doping and t/J. Similar results were also found to be valid for other values of the parameters.

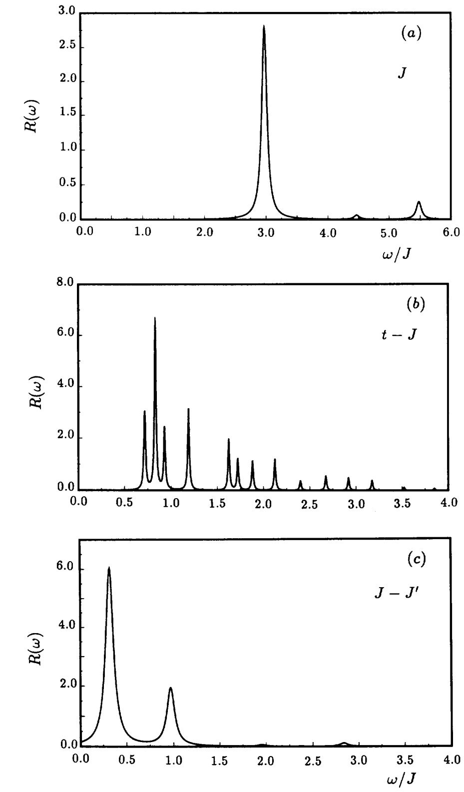

FIG. 1. Raman spectrum for the B_{1g} geometry. (a) Pure Heisenberg model; (b) t-J model with one hole, $t/J=2.5$; (c) frustrated Heisenberg model, $J'/J=0.5$.

The general features of these complex spectra can be better captured by studying their first moments as a function of the parameters J'/J and t/J (in the t-J model the region of experimental interest is $t/J \sim 2.5$–5). In Fig. 2 we show the first two moments for the Raman spectrum of (a) the J-J' model and (b) the t-J model with one hole. The first moment, M_1/J, corresponds to the center of gravity of the spectrum, while the second, M_2/J, measures its width. Let us start with the pure Heisenberg model. Increasing J'/J produces a *uniform* shift of the center of gravity towards zero frequency, for $J'/J \le 0.6$.

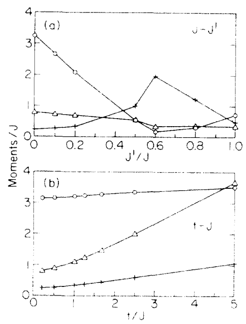

FIG. 2. Moments of the B_{1g} Raman spectrum for (a) the J-J' model, and (b) the t-J model with one hole, as a function of the frustration parameter. The symbols refer to the center of gravity, M_1/J (○), width of the spectrum M_2/J (△), and M_2/M_1 (+).

For $J'/J \geq 0.6$, the center of gravity slowly increases with increasing frustration. The corresponding behavior of M_1/J, as a function of t/J, is different, being in this case a straight line with positive slope. Increasing J'/J (t/J) produces a *decrease* (dramatic *increase*) in the width M_2/J of the Raman spectrum for the J-J' (t-J) model. Clearly, the results between the frustrated and doped models differ substantially. It is important to note that the doped results qualitatively reproduce the experimental measurements in the B_{1g} geometry, while the frustrated Hamiltonian does not.

Let us now consider the neutron scattering [11]. The quantum Heisenberg model has provided a qualitative description of the undoped cuprates. However, the magnetic excitations in the superconducting cuprates have been more difficult to study not only because the magnetic scattering is weaker, in part due to the lack of long-range order, but also due to the scarcity of good samples. Currently, there is no detailed comparison between theory and experiments. Recent data on the $S(\mathbf{q},\omega)$ of a $La_{1.85}Sr_{0.15}CuO_4$ crystal with $\mathbf{q}=h(\pi/a,\pi/a)$ show a clear peak at $(\pi/a,\pi/a)$, suggesting the presence of antiferromagnetic fluctuations, which appear to be incommensurate with a maximum at $h\approx 0.85$. This incommensurate peak might be due to the introduction of frustration. The main effect of doping is to broaden and shift the (π,π) peak. The $T=0$ dynamic spin-spin structure factor, $S(\mathbf{q},\omega)$, is given by

$$S(\mathbf{q},\omega)=\frac{1}{N}\sum_l \sum_m \int dt\, e^{i\omega t - iq(l-m)}\langle \psi_0 | S_l^z(t) S_m^z(0) | \psi_0 \rangle , \tag{4}$$

where $S_l^z(t)$ is the z component of the spin at site l, after time evolution in real time t, and $|\psi_0\rangle$ is the ground-state wave function. In Fig. 3 we show $S(\mathbf{q},\omega)$ for $\mathbf{q}=(\pi,\pi)$ of the spin-$\frac{1}{2}$ (a) pure Heisenberg antiferromagnet, (b) the t-J model, and (c) the J-J' Hamiltonian. Our goal now is to study the effects of frustration and doping on $S(\mathbf{q},\omega)$. We observe that the only effect of increasing J' is to shift all the peaks towards the low-energy region. This occurs for all peaks except the (π,π) which has a shift toward higher frequencies. This is precisely the peak shown in Fig. 3(c). A simple explanation of this behavior comes from the observation that the energy of the triplet state with momentum (π,π) and the ground-state energy are almost parallel straight lines for $J'/J \leq 0.60$. The displacement of this peak appears to be a linear function of the frustration parameter J'/J. On the other hand, the evolution of the structure as a function of (small) doping is very different, the spectrum now being broader and with a pseudoband. This behavior is shown in Fig. 3(b) for $J/t=0.4$. For the $t=0$ static-hole case, only two peaks are present, while the structure becomes much richer for the $t>0$ case. In particular, there is a broadening of the spectrum as one moves over the first Brillouin zone. This is not the case for the pure Heisenberg and frustrated Heisenberg antiferromagnets, since they show a monotonic behavior. The width $M_2^{\mathbf{q}}/J$ of the spectrum for the different models is shown in Fig. 4. We think that the origin of these results is the fast destruction of the antiferromagnetism by the hole doping. In fact, the suppression of the antiferromagnetism is local in the t-J model, while it is global in the J-J' model.

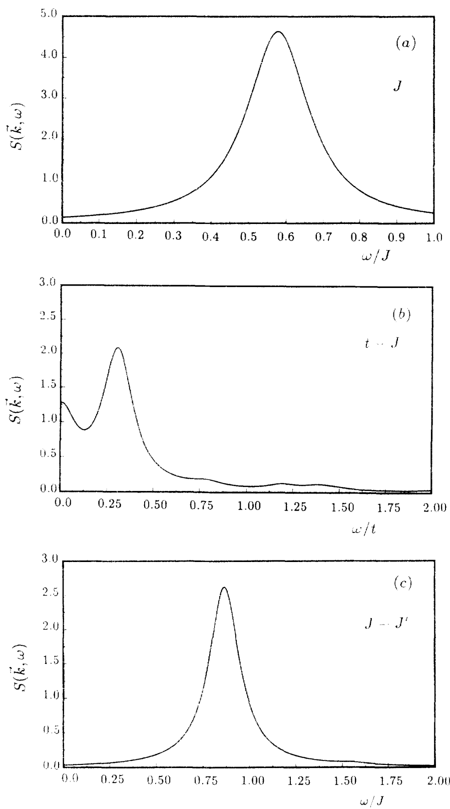

FIG. 3. Dynamic structure factor at $\mathbf{q}=(\pi,\pi)$ for (a) the pure Heisenberg model, (b) the t-J model with one hole, and (c) the frustrated Heisenberg model. The parameters are as in Fig. 1.

In all exact diagonalization studies, finite-size effects

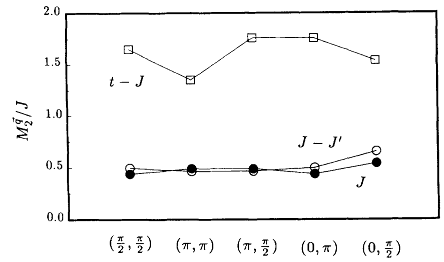

FIG. 4. Width, $M_2^{\mathbf{q}}/J$, of the $S(\mathbf{q},\omega)$ spectrum as a function of $\mathbf{q}$. The parameter values are as in Fig. 1.

are unavoidable. For instance, neutron-scattering results for the insulating case can be understood by low-frequency, long-wavelength magnetic models for which finite-size effects could be expected to play a role. However, since hole motion reduces the antiferromagnetic order present at half filling, there is a suppression of spin excitations at long and intermediate wavelengths for the doped case. Therefore, the finite-size effects present in the Heisenberg model become much smaller for the t-J case [1]. In inelastic light-scattering measurements, one probes high-frequency *very-short-wavelength* spin fluctuations (from energetic spin-pair two-magnon excitations) that are much less dependent on finite-size effects. In fact, results for a 20-site lattice indicate that the same structure persists for the B_{1g} Raman spectrum [12]. In all cases, size effects generate small quantitative corrections while the results, for doped and frustrated models, are qualitatively and drastically different. Finally, *much larger lattices* (up to 30×30) have been studied [5], for *any* value of the doping, by using a completely different approach. Still, the general conclusion remains the same.

In conclusion, the two-dimensional spin frustrated [3,6] (J-J') and electronic (t-J) models have been extensively studied, mostly *separately*, since the discovery of the high-T_c materials. A link between them has been argued [2], while many other proposals, for example, the chiral spin state [6], have assumed it. The connection between these two models has been at the heart of much recent work on magnetic spin Hamiltonians as models for high-temperature superconductors. It has been the purpose of this work to analyze this connection by computing several measurable quantities. More specifically, we have computed and analyzed the B_{1g} Raman spectrum and the dynamic spin-spin structure factor for the t-J model with one hole, the J-J' model, as well as the Heisenberg model, and qualitatively compared these results with the available experimental measurements. Our results indicate that the predictions based on purely frustrated spin systems [3,6] are *not* appropriate, either quantitatively or qualitatively, for modeling the magnetic behavior of the doped systems. These conclusions are consistent with other studies [5] based on a completely different approach (mean field) in much larger lattices, and for *any* value of the doping. On the other hand, experimental observations in the superconducting cuprates can be qualitatively described by the t-J model. Our observations and conclusions, obtained by looking at two *different physical quantities* in several different models, are compelling because of their mutual consistency.

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[12] The two-magnon peak for the 20-site cluster has a relative shift of ≈2% (with respect to the 160-site lattice) to higher frequencies. Its location is $3.045J$ ($2.976J$) for 20 (16) sites.