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Disappearance of long-range order in doped antiferromagnets: A strong-coupling approach

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The spin fluctuations in slightly doped copper oxide materials are studied within the framework of the t-J model to first order in the doping δ. Due to the motion of the holes, the magnetic properties are modified in the following ways: (i) the system becomes incommensurate; (ii) the spin waves acquire a *finite* lifetime. The latter effect dominates for small δ and is probably responsible for the destruction of long-range order by a few percent doping.

The spin fluctuations in doped copper oxide materials have been intensively investigated both theoretically and experimentally since the discovery of high-T_c superconductors, due to their possible connection to superconductivity. Experimentally, the long-range antiferromagnetic (AF) order, present in the undoped insulators, is quickly destroyed (by approximately 2% doping).[1] Nevertheless, neutron scattering[1] and NMR data[2] suggest that there are still short-range AF fluctuations. With increasing doping, the fluctuations become incommensurate in La_2CuO_4-based materials.[3] Several strong-coupling approaches (typically the large-U Hubbard model or the t-J model) have been proposed. For instance, one can reach a qualitative agreement with the experimental results by assuming that the holes induce a static frustration leading to additional coupling constants.[4] If they are big enough, they will lead to the destruction of long-range order (LRO) by long wavelength fluctuations.[5] This approach is essentially phenomenological, however, and there is little evidence for such a model of static frustration. In particular, starting from the t-J model, and studying the static frustration effectively induced by the holes on the spins, one can get incommensurate order,[6,7] but the doping needed to destroy the LRO is very large ($\sim$60%). In fact, there are good reasons to believe that such a static frustration picture is inadequate. Hopping of doped holes creates a backflow spin current due to strong correlation.[8] The static frustration picture neglects this paramagnetic spin current and violates the spin conservation, resulting in an incorrect Goldstone-mode structure of the spin-wave excitation spectrum.

As we shall show in this Rapid Communication, this paramagnetic spin current is also essential to understand the disappearance of LRO upon doping. Due to their coupling to this paramagnetic spin current, spin waves are scattered by moving holes. This results in a broadening of the spin-wave spectral density. As suggested some time ago by Ramakrishnan,[9] this broadening enhances dramatically the reduction of magnetic order by long-wavelength spin waves, which become the most effective source of destruction of LRO at small doping.

Throughout this paper, we will work with the t-J model

$$H=-t\sum_{NN}(c_{i\sigma}^\dagger c_{j\sigma}+\text{H.c.})+J\sum_{NN}\mathbf{S}_i\cdot\mathbf{S}_j\,, \qquad (1)$$

where, for simplicity, the hopping and Heisenberg interactions are restricted to nearest neighbors. We use Schwinger bosons $b_{i\sigma}^\dagger$ and slave fermions f_i to represent the projected hole operators $c_{i\sigma}^\dagger$:

$$c_{i\sigma}^\dagger=b_{i\sigma}^\dagger f_i,\;\; \mathbf{S}_i=\tfrac{1}{2}\,b_{i\sigma}^\dagger\boldsymbol{\sigma}_{\sigma\sigma'}b_{i\sigma'}\,. \qquad (2)$$

They are subject to the local constraint

$$\sum_\sigma b_{i\sigma}^\dagger b_{i\sigma}+f_i^\dagger f_i=2S\,. \qquad (3)$$

One can justify this choice by noting that in the limit of zero doping, one recovers the two-dimensional (2D) AF Heisenberg model, which is known to be well described by the Schwinger boson mean-field theory.[10,11] Besides, we shall concentrate on the strong-coupling t-J model, i.e., the limit $t/J\gg 1$. It is well known that the t-J model suffers from an instability against charge-density fluctuations leading to phase separation, at least for small t/J.[12,13] Experimentally, there is little evidence for phase separation in the doped copper oxides, except possible chemical phase separation in La_2CuO_{4+x}. Physically, it is clear that the long-range Coulomb interaction, which is neglected in the t-J model, shifts the threshold of long wavelength charge fluctuations to the plasmon frequency, and, therefore, suppresses the charge-density instability.[8] In a realistic model, this interaction should be included. However, it does not play any role in our study of the effect of doping of spin fluctuations. Bearing this in mind, we shall confine our discussion within the t-J model, but we shall neglect the possibility of phase separation since our main concern is not the phase diagram of the pure t-J model.

In this context, LRO appears as a Bose condensation of the Schwinger bosons. Treating the constraint with a Lagrange multiplier, one gets

$$S^*(\delta,t/J)=S-\delta/2-\frac{1}{2N}\sum_{\mathbf{q}\neq 0,\sigma}\langle b_{\mathbf{q}\sigma}^\dagger b_{\mathbf{q}\sigma}\rangle\,, \qquad (4)$$

where $S=\frac{1}{2}$ is the spin, δ is the doping, and S^* is the number of bosons of a given spin that are condensed in the $\mathbf{q}=\mathbf{0}$ state. It can be shown that for $S^*\neq 0$, there is LRO in the spin-spin correlation function with a staggered magnetization equal to S^*. For 2D AF, $\delta=0$, the quantum fluctuations yield $(1/2N)\sum_{\mathbf{q}\neq 0,\sigma}\langle b_{\mathbf{q}\sigma}^\dagger b_{\mathbf{q}\sigma}\rangle\simeq 0.2$. Then, $S^*\simeq 0.3$, and the ground state has long-range order. When the holes are added in, the effect of hole hopping in the static frustration picture is simply to generate incommensurability, and the staggered magnetization S^*

is nearly unchanged[13] (for small doping δ, the change is of order δ^2), in contrast to experiments.

Since holes cannot hop in a perfect AF spin background, the spin background has to be twisted to allow holes to hop and to gain some kinetic energy. The general method to treat such incommensurate magnets is to rotate independently the spin basis at each lattice site so that the reference frame is locally ferromagnetic.[14] The Hamiltonian in the new reference frame is

$$H = H_{\rm even} + H_{\rm odd}\ , \tag{5}$$

$$\begin{aligned} H_{\rm even} = &-\sum_{\rm NN} t_{ij}\cos(\theta_{ij}/2) f_i f_j^\dagger b_i^\dagger b_j + \text{H.c.} \\ &+ J\sum_{\rm NN} \{(\mathbf{S}_i\cdot\mathbf{n}_{ij})(\mathbf{S}_j\cdot\mathbf{n}_{ij}) \\ &+ \cos\theta_{ij}[\mathbf{S}_i\cdot\mathbf{S}_j - (\mathbf{S}_i\cdot\mathbf{n}_{ij})(\mathbf{S}_j\cdot\mathbf{n}_{ij})]\}\ , \end{aligned} \tag{6}$$

$$\begin{aligned} H_{\rm odd} = &-\sum_{\rm NN} t_{ij}\sin(\theta_{ij}/2) f_i f_j^\dagger b_i^\dagger (\mathbf{n}_{ij}\cdot\boldsymbol{\sigma}) b_j + \text{H.c.} \\ &+ J\sum_{\rm NN} \sin\theta_{ij}\mathbf{n}_{ij}\cdot(\mathbf{S}_i\times\mathbf{S}_j)\ , \end{aligned}$$

where $b^\dagger = (b_\uparrow^\dagger, b_\downarrow^\dagger)$, the unit vector $\mathbf{n}_{ij}$ is the axis required to rotate the coordinate system at site j into the coordinate system at site i, and θ_{ij} is the angle of rotations between the two local reference frames. In the static frustration picture, one performs a mean-field factorization of the even part of the Hamiltonian $H_{\rm even}$ and neglect the odd part $H_{\rm odd}$. It is easy to verify that $H_{\rm even}$ does *not* conserve the total spin by calculating their commutator, leading to an incorrect shape of the spin excitation spectrum. The first term of $H_{\rm odd}$ results from the local constraint: a forward hopping hole is associated with a backflow spin current. This term describes a dynamic frustration effect. For $t/J \gg 1$, although its average vanishes, its fluctuation cannot be neglected. In the large S limit, a systematic $1/S$ expansion which conserves spin demonstrated that the correct spin-wave spectrum can be recovered by including the fluctuations of $H_{\rm odd}$.[8] In that case, the spin-wave velocity is renormalized down by hole hopping. In the small doping limit, the leading renormalization comes from the fluctuations of $H_{\rm odd}$, yielding $c = c_0 - \text{const}\times t^2\delta/J$, which signals the instability of LRO for $t^2\delta/J^2 \sim 1$.

In order to study the effect of small doping for the more realistic $S=\frac{1}{2}$ case, we make a mean-field factorization for $H_{\rm even}$ and assume a simple uniform twist: $\mathbf{n}_{ij} = \hat{\mathbf{z}}$, $\theta_{ij} = \mathbf{Q}\cdot(\mathbf{R}_i - \mathbf{R}_j)$. We take into account the local constraint (3) by using a Lagrange multiplier λ and make a saddle-point approximation. Now, $H_{\rm even}$ is quadratic both in Schwinger boson and slave-fermion operators. Since for small doping incommensurability can change the magnetization only by an amount of order δ^2, we can simply approximate the bosonic part of $H_{\rm even}$ by the mean-field AF Hamiltonian to get

$$H_{\rm even} = \sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} f_{\mathbf{k}}^\dagger f_{\mathbf{k}} + \sum_{\mathbf{q},\sigma} [\lambda b_{\mathbf{q}\sigma}^\dagger b_{\mathbf{q}\sigma} + \tfrac{1}{2}\Delta_{\mathbf{q}}(b_{\mathbf{q}\sigma} b_{-\mathbf{q}-\sigma} + \text{H.c.})]\ , \tag{7}$$

where

$$\varepsilon_{\mathbf{k}} = \tilde{t}(\cos k_x + \cos k_y) - \mu\ , \tag{8}$$

$$\Delta_{\mathbf{q}} = \Delta(\cos q_x + \cos q_y)\ , \tag{9}$$

and where λ and Δ have the same value as for $\delta = 0$. In Eq. (8), μ is the chemical potential and is determined by the hole density, while $\tilde{t}$ is the effective fermionic hopping. For small doping, the fermionic band is narrow: When there is long-range order, $\tilde{t} \sim t^2\delta/J$; without long-range order, it becomes of order δ^2. For $H_{\rm odd}$, we only retain the first term, because the second term, of order $\sin\theta_{ij} \sim \delta$, gives a correction to S^* of order δ^2.

$$\begin{aligned} H_{\rm odd} = \frac{1}{N}\sum_{\mathbf{k}_1,\mathbf{k}_2,\mathbf{k}_3,\mathbf{k}_4} &\delta(\mathbf{k}_1 - \mathbf{k}_2 + \mathbf{k}_3 - \mathbf{k}_4) \\ &\times g_{\mathbf{k}_2-\mathbf{k}_3} f_{\mathbf{k}_1}^\dagger f_{\mathbf{k}_2} b_{\mathbf{k}_3}^\dagger \sigma^z b_{\mathbf{k}_4}\ , \end{aligned} \tag{10}$$

where the coupling constant is

$$\begin{aligned} g_{\mathbf{k}} &= -2t[\sin(\tfrac{1}{2}Q_x)\sin k_x + \sin(\tfrac{1}{2}Q_y)\sin k_y] \\ &\approx -2t(\sin k_x + \sin k_y)\ . \end{aligned} \tag{11}$$

Our starting point is the effective Hamiltonian (7) + (10). In a state with LRO, $H_{\rm odd}$ includes terms involving boson condensate $b_{\mathbf{k}=0,\sigma}$. These terms reduce to effective three-particle interactions that describe the processes of spin-wave decay into particle-hole pair. These terms are not responsible for the destruction of LRO in a self-consistent treatment. There are two reasons for this. First, when the critical doping for destruction of LRO is approached, which is the region we are really interested in, this type of interaction becomes vanishingly small because $\langle b_{\mathbf{k}=0,\sigma}\rangle$ goes to zero. Second, for small doping only virtual decaying processes for spin waves are allowed by this type of interactions which lead only to the softening of spin-wave excitations.[8,15] It mainly reduces the spin interaction strength J, which has little effect on the determination of the magnitude of the ordered moment. For example, the order moment is independent of J in pure 2D AF. For these reasons, only the four-particle interactions of $H_{\rm odd}$ which allow real decaying processes for spin excitations will be considered in the following. In the presence of interactions (10), the full Schwinger boson Green's functions can be represented in terms of the self-energy matrix $\Sigma(i\nu_n, \mathbf{q})$,

$$\begin{aligned} \underline{D}_\sigma(i\nu_n,\mathbf{q}) &= \begin{bmatrix} \langle b_{\mathbf{q}\sigma}^\dagger(i\nu_n) b_{\mathbf{q}\sigma}(i\nu_n)\rangle & \langle b_{\mathbf{q}\sigma}^\dagger(i\nu_n) b_{-\mathbf{q}-\sigma}^\dagger(-i\nu_n)\rangle \\ \langle b_{\mathbf{q}\sigma}(i\nu_n) b_{-\mathbf{q}-\sigma}(-i\nu_n)\rangle & \langle b_{-\mathbf{q}-\sigma}(-i\nu_n) b_{-\mathbf{q}-\sigma}^\dagger(-i\nu_n)\rangle \end{bmatrix} \\ &= \begin{bmatrix} i\nu_n - \lambda - \Sigma_{11}^\sigma(i\nu_n,\mathbf{q}) & -\Delta_{\mathbf{q}} - \Sigma_{12}^\sigma(i\nu_n,\mathbf{q}) \\ -\Delta_{\mathbf{q}} - \Sigma_{21}^\sigma(i\nu_n,\mathbf{q}) & -i\nu_n - \lambda - \Sigma_{22}^\sigma(-i\nu_n,-\mathbf{q}) \end{bmatrix}^{-1}\ . \end{aligned} \tag{12}$$

In the absence of magnetic field, $\Sigma_{11}^{\sigma}(i\nu_n,\mathbf{q})=\Sigma_{22}^{\bar\sigma}(i\nu_n,\mathbf{q})$, $\Sigma_{12}^{\sigma}(i\nu_n,\mathbf{q})=\Sigma_{21}^{\bar\sigma}(i\nu_n,\mathbf{q})$, and the self-energies do not depend on spin so that the superscript σ will be omitted. In general, the imaginary parts of the self-energies do not vanish, and the Schwinger boson spectral density is no longer a Dirac function, but is broadened. Now, the important point is that this broadening increases the boson occupation number $n_{\mathbf{q}\sigma}$ for $\mathbf{q}\neq\mathbf{0}$.

Let us show this by using the following simple illustration. In the mean-field AF,

$$n_{\mathbf{q}\sigma}=\frac{1}{2}\left[\frac{\lambda}{\omega_{\mathbf{q}}}-1\right]=-\lambda\int_{-\omega_c}^{\omega_c}d\omega\, n_B(\omega)\delta(\omega^2-\omega_{\mathbf{q}}^2)-\frac{1}{2}\,, \tag{13}$$

where $\omega_{\mathbf{q}}=(\lambda^2-\Delta_{\mathbf{q}}^2)^{1/2}$ is the spin-wave spectrum, $n_B(\omega)=1/(e^{\beta\omega}-1)$ is the Bose distribution function, and ω_c is the maximum spin-wave energy. The first term of (13) $\lambda/\omega_{\mathbf{q}}$ redistributes Schwinger boson spectral weight due to quantum fluctuations. It cancels out in the spectral density sum rule, which is a direct consequence of the Schwinger boson commutation relation. When the spectrum is broadened, let us assume for simplicity that the Dirac function becomes a Lorentzian distribution:

$$\delta(\omega^2-\omega_{\mathbf{q}}^2)\rightarrow\frac{1}{\mathcal{N}}\frac{\Gamma}{(\omega^2-\omega_{\mathbf{q}}^2)^2+\Gamma^2}\,.$$

The normalization factor $\mathcal{N}$ is necessary to ensure the sum rule. If we assume that the self-energies have an expansion in some small parameter ϵ (in our case $\epsilon=t^2\delta/J^2$) and keep only the first-order terms, the broadening Γ is proportional to ϵ. Usually, this kind of expansion does *not* satisfy general sum rules to first order in ϵ and one needs to include contributions of the self-energies of order ϵ^2 to cure it. However, when one calculates a quantity averaged by the spectral density, one can prove that using the spectral density calculated from the first-order self-energies only but with a normalization factor yields a result that is accurate to first order in ϵ, and this is what we do here. We note that the variable involved is ω^2, due to time-reversal symmetry in AF state. This broadening increases the first term of (13). This means that the quantum fluctuations are enhanced. To see it, we can calculate, as an example,

$$\frac{1}{\mathcal{N}}\int_0^{\omega_c}d\omega^2\left[\frac{\omega_{\mathbf{q}}^2}{\omega^2}\right]^{1/2}\frac{\Gamma}{(\omega^2-\omega_{\mathbf{q}}^2)^2+\Gamma^2}\simeq 1+\alpha_{\mathbf{q}}\Gamma/\omega_c^2+\cdots\,,\quad \Gamma\ll 1\,,$$

$$\mathcal{N}=\int_0^{\omega_c}d\omega^2\frac{\Gamma}{(\omega^2-\omega_{\mathbf{q}}^2)^2+\Gamma^2}\,.$$

For our practical problem, $\omega_c\sim O(J+\tilde t)$. Taking $\omega_{\mathbf{q}}^2/\omega_c^2=0.5$, we have $\alpha_{\mathbf{q}}\simeq 0.93$. The enhancement is stronger for smaller $\omega_{\mathbf{q}}$. So, the broadening of the spectral density, which is measured by Γ, leads to an increase of the boson occupation factor, hence, to a decrease of the magnetization S^*. In the actual problem, the broadening Γ depends on the frequency ω and the momentum $\mathbf{q}$, and we must resort to numerical calculations, but the essential physics is the same.

Let us now turn to a more quantitative approach and calculate the self-energies for our model. For small doping, the bosonic self-energies can be expanded in terms of doping δ. We have calculated the RPA self-energies assuming a gapless bare bosonic spectrum (Fig. 1) and retained only the terms linear in δ. The imaginary part of the self-energies is then given by

$$\Sigma''_{\alpha\beta}(\nu,\mathbf{q})=\frac{t^2\delta}{\Delta^2}\tilde\Sigma''_{\alpha\beta}(\nu,\mathbf{q})+O(\delta^2),\quad \alpha,\beta=1,2\,, \tag{14}$$

and the real part is obtained from it by using the Kramers-Kronig relations. Substituting the self-energies calculated this way into (12), we have calculated the renormalized boson occupation numbers $n_{\mathbf{q}}$, from which we get S^* according to

$$S^*(\delta,t/J)=S-\frac{\delta}{2}-\frac{1}{2N}\sum_{\mathbf{q},\sigma}n_{\mathbf{q}\sigma}\,, \tag{15}$$

$$n_{\mathbf{q}\sigma}=\frac{1}{\pi}\int d\nu\, n_B(\nu)\,\mathrm{Im}\langle b_{\mathbf{q}\sigma}^{\dagger}(\nu+i\epsilon)b_{\mathbf{q}\sigma}(\nu+i\epsilon)\rangle\,. \tag{16}$$

As discussed above, this calculation includes all contributions linear in $t^2\delta/J^2$, assuming that $n_{\mathbf{q}\sigma}$ has an expansion.

We mentioned before that $\tilde t$ is small. After taking into account Fermi statistics, the result of the calculation is insensitive to the value of $\tilde t$, so long as $\tilde t/J\ll 1$. So, effectively one can take $\tilde t=0$. Besides, Σ'' depends on δ and t/J only through the ratio $t^2\delta/J^2$ to the first order in δ (actually, it is $t^2\delta/\Delta^2$, $\Delta=1.16J$ for 2D AF), so that the various curves $S^*(\delta,t/J)$ scale with each other. The numerical results are shown in Fig. 2. Extrapolating from the calculated small doping results, the long-range order is destroyed for $t^2\delta/J^2\sim 0.3$. Taking $t/J=3$–4, the required doping is about 1%–2%. So, in the strong-coupling regime $t/J\gg 1$, where the fermions are very mobile, the destruction of LRO is expected to occur well before $\delta\simeq 0.6$, the value predicted in the static frustration picture.

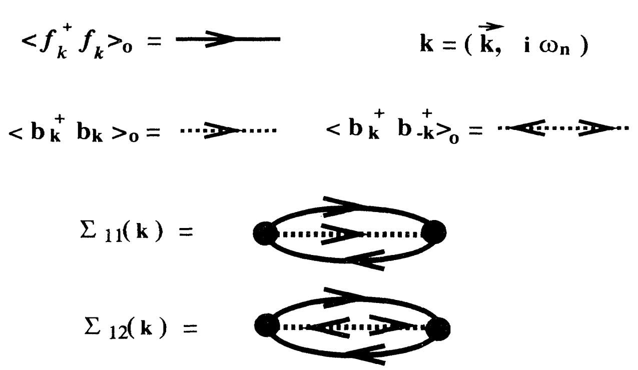

FIG. 1. Bare Green's functions and RPA Schwinger boson self-energies.

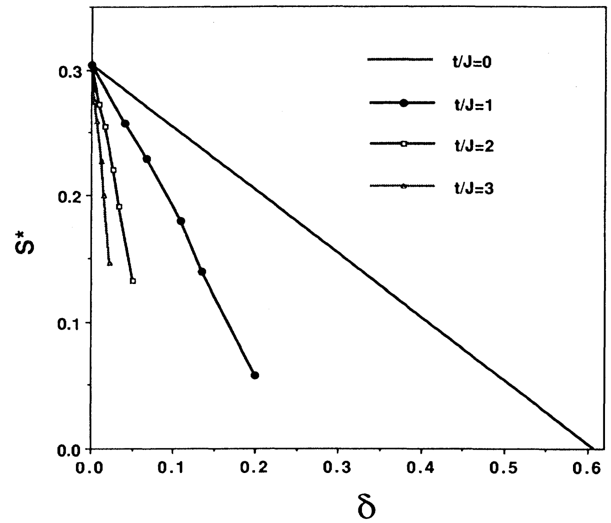

FIG. 2. The staggered magnetization vs doping for various values of parameter t/J.

Beyond the critical doping where LRO is destroyed, we expect that a gap gradually opens up in the Schwinger boson spectrum. For small gap, the spin-spin correlation length will be effectively determined by the imaginary part of the spin excitation frequency. From (14), if we naively assume $\mathrm{Im}\omega_{\mathbf{q}}^2 \sim t^2\delta/J^2$, then for small $\mathrm{Re}\omega_{\mathbf{q}}^2$ (long wavelength) we expect the inverse correlation length ξ^{-1} to be proportional to $\mathrm{Im}\omega_{\mathbf{q}} \sim t\sqrt{\delta}/J$. The scaling $\xi^{-1} \sim \sqrt{\delta}$ is precisely what was found experimentally.[1,16]

To summarize, we have calculated the effect of hole doping in a 2D spin-$\frac{1}{2}$ antiferromagnet to first order in δ. The main effect is a reduction of the ordered moment suggesting that, for large t/J, the destruction of LRO occurs for very small doping. The most direct extension of this work would be to include the incommensurability predicted by various authors.[6,7,17] Let us emphasize again that this effect is of order δ^2 and will not modify qualitatively the main result of the present work. However, the symmetry of the incommensurate state is still controversial: If only H_{even} is taken into account, one finds a pitch vector **Q** along the diagonal $(Q_x = Q_y)$,[7] while the weak-coupling approach, for example, that of Schulz,[17] predicts a pitch vector **Q** along the x and y axis, in agreement with experiments.[16] It would be interesting to see whether, within the present approach, a consistent analysis of the δ^2 contribution, including H_{odd}, reproduces this latter result. Work is in progress along these lines.

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