Destruction of long-range antiferromagnetic order by hole doping

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Abstract

We study the renormalization of the staggered magnetization of a two-dimensional antiferromagnet as a function of hole doping, in the framework of the t-J model. It is shown that the motion of holes generates decay of spin waves into “particle-hole” pairs, which causes the destruction of the long-range magnetic order at a small hole concentration. This effect is mainly determined by the coherent motion of holes. The value obtained for the critical hole concentration, of a few percent, is consistent with experimental data for the doped copper oxide high-$T_c$ superconductors.

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One of the interesting features of the copper oxide high-$T_c$ superconductors is the dramatic reduction, with doping, of the long-range magnetic order of their parent compounds.\textsuperscript{1} The undoped materials, e.g. $La_2CuO_4$, are antiferromagnetic (AF) insulators. Doping, e.g., in $La_{2-\delta}Sr_{\delta}CuO_4$, introduces holes in the spin lattice of the $CuO_2$ planes, and the long-range AF order is destroyed at a small hole concentration, $\delta_c \sim 0.02$. The $CuO_2$ planes are described by a spin-1/2 Heisenberg antiferromagnet on a square lattice, with moving holes that strongly interact with the spin array. The motion of holes generates spin fluctuations that tend to disrupt the AF order. It has been shown that hole motion produces strong effects on the magnetic properties, leading in particular to significant softening and damping of the spin excitations as a function of doping.\textsuperscript{2–5} The critical concentration $\delta_c$ where long-range magnetic order disappears has often been identified with the concentration where the spin wave velocity vanishes. However important damping effects occur, which have to be taken into account. In particular, all spin waves become overdamped at a concentration well below the one for which the spin wave velocity vanishes, suggesting that the long-range AF order may disappear at a smaller concentration.\textsuperscript{5} The critical hole concentration $\delta_c$ is provided by the vanishing of the staggered magnetization order parameter.

In this work we use the t-J model to calculate the doping dependence of the staggered magnetization of a two-dimensional antiferromagnet, and determine the critical hole concentration $\delta_c$. It is shown that the motion of holes generates decay of spin waves into "particle-hole" pairs, leading to broadening of the spin-
wave spectral function. This broadening gives rise to a drastic reduction of the staggered magnetization and the disappearance of the long-range order at low doping, in agreement with experiments. Such a process was suggested some years ago by Ramakrishnan. The vanishing of the staggered magnetization as a consequence of doping, has already been studied in the t-J model by Gan and Mila, considering the scattering of spins by moving holes, and by Khaliullin and Horsch, considering spin disorder introduced by the incoherent motion of holes.

We describe the copper oxide planes with the t-J model,

\[ H_{t-J} = -t \sum_{<i,j>,\sigma} \left( c_i^\dagger c_j + H.c. \right) + J \sum_{<i,j>} \left( \mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j \right), \]  

(1)

where, \( \mathbf{S}_i = \frac{1}{2} c_i^\dagger \sigma_{\alpha\beta} c_i \beta \) is the electronic spin operator, \( \sigma \) are the Pauli matrices, \( n_i = n_{i\uparrow} + n_{i\downarrow} \) and \( n_{i\sigma} = c_i^\dagger c_{i\sigma} \). To enforce no double occupancy of sites, we use the slave-fermion Schwinger Boson representation for the electron operators \( c_{i\sigma} = f_i^\dagger b_{i\sigma} \), where the slave-fermion operator \( f_i^\dagger \) creates a hole and the boson operator \( b_{i\sigma} \) accounts for the spin, subject to the local constraint \( f_i^\dagger f_i + \sum_{\sigma} b_i^\dagger b_{i\sigma} = 2S \). For zero doping, the model (1) reduces to a spin-1/2 Heisenberg antiferromagnet, exhibiting long-range Néel order at zero temperature. The Néel state is represented by a condensate of Bose fields \( b_{i\uparrow} = \sqrt{2S} \) and \( b_{i\downarrow} = \sqrt{2S} \), respectively in the up and down sub-lattices, and the bosons \( b_{i\downarrow} = b_i \) and \( b_{i\uparrow} = b_j \) are then spin-wave operators on the Néel background. After Bogoliubov-Valatin transformation on the boson Fourier transform \( b_k = v_k \beta_k^\dagger + u_k \beta_k \), with \( u_k = \left[ (1 - \gamma_k^2)^{-1/2} + 1 \right]/2 \right)^{1/2} \), \( v_k = \)
\[-\text{sgn}(\gamma_k) \left[ \left( (1 - \gamma_k^2)^{-1/2} - 1 \right) / 2 \right]^{1/2}, \text{ and } \gamma_k = \frac{1}{2} (\cos k_x + \cos k_y), \] we arrive at the effective Hamiltonian

\[
H = -\frac{1}{\sqrt{N}} \sum_{q, k} f_q f_q^\dagger \left[ V(q, -k) \beta_k - V(q - k, k) \beta_k^\dagger \right] + \sum_k \omega_k^0 \beta_k \beta_k^\dagger, \tag{2}
\]

having \( S = 1/2 \) and \( N \) sites in each sub-lattice. In (2), the first term, with \( V(q, k) = z t (\gamma_q u_k + \gamma_{q+k} v_k) \), represents the interaction between holes and spin waves resulting from the motion of holes with emission and absorption of spin waves, and the second term describes spin waves for a pure antiferromagnet, with dispersion \( \omega_k^0 = (zJ/2) \left( 1 - \gamma_k^2 \right)^{1/2}, \) \( z \) being the lattice coordination number \( (z = 4). \)

The staggered magnetization is given by

\[
M = < S^z_i > - < S^z_i > = 2 < S^z_i >, \tag{3}
\]

with

\[
< S^z_i > = \sum_{i \in S(\uparrow)} < S^z_i >,
\]

where the sum is over the up sub-lattice. Using the Schwinger boson representation for the spin operator \( S_i^z = \frac{1}{2}(c^\dagger_i\sigma^\uparrow c_i - c^\dagger_i\sigma^\downarrow c_i) \), and the boson condensation associated to the Néel state, one has \( S_i^z = (1 - b_i^\dagger b_i)(S - b_i^\dagger b_i) \), which, after Bogoliubov-Valatin transformation, leads to

\[
M = (1 - \delta) [M_0 - \Delta M], \tag{4}
\]
where
\[ M_0 = 2 \left[ N S - \sum_k v_k^2 \right] \] (5)
is the staggered magnetization for a pure antiferromagnet, and
\[ \Delta M = 2 \sum_k \left[ (u_k^2 + v_k^2) < \beta_{-k}^\dagger \beta_{-k} > \ight. \\
\left. + u_k v_k ( < \beta_k \beta_{-k} > + < \beta_{-k}^\dagger \beta_{-k}^\dagger > ) \right]. \] (6)
The prefactor in (4) accounts for the spin dilution due to doping, being negligible for small hole concentrations. In (5), the order parameter is considerably reduced by quantum fluctuations, to \( \simeq 0.6 \times 2 N S \). With zero doping the expectation values in (6) vanish and \( \Delta M = 0 \). However, in a doped system the motion of holes generates spin fluctuations, giving rise to nonzero expectation values in (6), even at zero temperature, and then \( \Delta M \neq 0 \).

In order to calculate the staggered magnetization for the doped system, we need the spin-wave Green’s functions, defined as
\[ D_{\mu \nu}^{+ -}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
\[ D_{\mu \nu}^{+ +}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
\[ D_{\mu \nu}^{- -}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
\[ D_{\mu \nu}^{+ -}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
\[ D_{\mu \nu}^{+ +}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
\[ D_{\mu \nu}^{+ -}(k, t - t') = -i \langle T \beta_k(t) \beta_k^\dagger(t') \rangle, \]
where \( \langle \cdot \rangle \) represents an average over the ground state. The spin-wave Green’s functions satisfy the Dyson equations:
\[ D_{\mu \nu}(k, \omega) = D_{0 \mu \nu}(k, \omega) + \sum_{\gamma \delta} D_{0 \mu \gamma}(k, \omega) \Pi_{\gamma \delta}(k, \omega) D_{0 \delta \nu}(k, \omega), \] where \( \mu, \nu = \pm \).

The free Green’s functions are
\[ D_{0 + +}(k, \omega) = 1/(\omega - \omega_k^0 + i \eta), \]
\[ D_{0 - -}(k, \omega) = 1/(\omega - \omega_k^0 - i \eta), \]
\[ D_{0 + -}(k, \omega) = D_{0 - +}(k, \omega) = 0, \] (\( \eta \to 0^+ \)), and \( \Pi_{\gamma \delta}(k, \omega) \) are the self-energies generated by the interaction between holes and spin waves. We calculate the spin-wave self-energies in the self-consistent Born approximation (SCBA), which corresponds to consider only ”bubble” diagrams with dressed
hole propagators, as illustrated in Figure 1. These diagrams describe decay of spin waves into "particle-hole" pairs. The spin-wave self-energies can then be written in terms of the hole spectral function, \( \rho(q, \omega) \), as

\[
\Pi^{\gamma\delta}(k, \omega) = \frac{1}{N} \sum_q U^{\gamma\delta}(k, q) \left[ Y(q, -k; \omega) + Y(q - k, k; -\omega) \right],
\]

with

\[
Y(q, -k; \omega) = \int_{-\infty}^{+\infty} d\omega' \int_{-\infty}^{0} d\omega'' \frac{\theta(q, \omega')\rho(q - k, \omega'')}{\omega + \omega'' - \omega' + i\eta},
\]

and, \( u^{--}(k, q) = u^{++}(k, q) = V(q, -k)V(q - k, k), \) \( u^{+-}(k, q) = V(q - k, k)^2, \) \( u^{-+}(k, q) = V(q, -k)^2. \) The relations \( \Pi^{++}(k, \omega) = \Pi^{+-}(-k, -\omega) \) and \( \Pi^{--}(k, \omega) = \Pi^{++}(k, \omega) \) are verified, the last implying \( D^{--}(k, \omega) = D^{++}(k, \omega). \)

The SCBA provides a spectral function for the holes,\(^9\)\(^{-15}\) that is composed of a coherent quasi-particle peak and an incoherent continuum, taking the approximate forms, respectively, \( \rho^{coh}(q, \omega) = a_0 \delta(\omega - \varepsilon_q) \) with \( a_0 \simeq (J/t)^{2/3} \), and \( \rho^{incoh}(q, \omega) = h\theta(|\omega| - zJ/2)\theta(2zt + zJ/2 - |\omega|) \) with \( h \simeq (1 - a_0)/2zt \), the energies are measured with respect to the Fermi level, and the quasi-holes fill up a Fermi surface consisting of pockets, of approximate radius \( q_F = \sqrt{\pi\delta} \), located at momenta \( q_i = (\pm \pi/2, \pm \pi/2) \) in the Brillouin zone, the quasi-particle dispersion being, near \( q_i \), written as \( \varepsilon_q = \varepsilon_{min} + (q - q_i)^2/2m \), with an effective mass \( m \simeq 1/J \). The self-energies will then present three contributions, \( \Pi^{\gamma\delta}(k, \omega) = \Pi^{\gamma\delta}_{c.c.}(k, \omega) + \Pi^{\gamma\delta}_{c.i.e.}(k, \omega) + \Pi^{\gamma\delta}_{i.c.i.e.}(k, \omega) \), corresponding, respectively, to transitions of holes within the coherent band, between the coherent and incoherent bands, and within the incoherent band. We have calculated the different contributions to lowest order in the hole concentration \( \delta \).
The change in the staggered magnetization induced by the interaction between holes and spin waves (6), is written in terms of the spin-wave Green’s functions, as

$$
\Delta M = -\sum_k \frac{2}{(1-\gamma_k^2)^{1/2}} \int_0^{+\infty} \frac{d\omega}{2\pi} \left[ 2\text{Im}D^{+-}(k,\omega) - \gamma_k \text{Im} \left(D^{++}(k,\omega) + D^{--}(k,\omega)\right) \right].
$$

(8)

To lowest order in the hole concentration $\delta$, (8) gives

$$
\Delta M = -\sum_k \frac{2}{(1-\gamma_k^2)^{1/2}} \left[ -\frac{\gamma_k}{2\omega_k} \text{Re}\Pi^{+-}(k,\omega_k^0) \right.
\left. + \int_0^{+\infty} \frac{d\omega}{\pi} \left( \frac{\text{Im}\Pi^{++}(k,\omega) + \gamma_k \text{Im}\Pi^{--}(k,\omega)}{(\omega + \omega_k^0)^2 + \gamma_k^2 \omega^2} \right) \right].
$$

(9)

Evaluating (9), one finds that the behavior of the staggered magnetization is essentially determined by the coherent motion of holes, and moreover, that it is governed by the imaginary part of the self-energies, i.e., the contributions

$$
\text{Im}\Pi^{\pm \pm}_{c,c}(k,\omega) = zJ\sqrt{\alpha_0^2} \left( \frac{t}{J} \right)^2 \frac{1}{\sqrt{\pi k(1-\gamma_k^2)^{1/2}}} F^{\pm \pm}(k,\omega)
\times \left[ \sqrt{1-s^2(g)}\theta(1-|s(g)|) - \sqrt{1-s^2(-g)}\theta(1-|s(-g)|) \right],
$$

with

$$
F^{--}(k,\omega) = [\cos(k_x g) + \cos(k_y g)] - \gamma_k [\cos k_x \cos(k_x g) + \cos k_y \cos(k_y g)],
$$

$$
F^{+-}(k,\omega) = \left( (\cos k_x - \cos k_y) (\cos(gk_x) - \cos(gk_y)) / 2 \right)
\times \left[ (1-\gamma_k^2)^{1/2} (\sin k_x \sin(gk_x) + \sin k_y \sin(gk_y)) - 2(1-\gamma_k^2) \right],
$$
where \( s(g) = (1 - g)k/2q_F \) and \( g = 2\omega/Jk^2 \), while

\[
\text{Re}\Pi^{-}_{c,c}(k, \omega^0_k) = -zJ\delta a_0^2 \left( \frac{t}{J} \right)^2 \frac{\gamma_k k^2}{8(1 - \gamma_k^2 - (k/2)^4)} \frac{(\sin^2 k_x + \sin^2 k_y)}{(1 - \gamma_k^2)^{1/2}}
\]

Regarding the incoherent contributions,

\[
\text{Im}\Pi^{-\pm}_{c,i,c}(k, \omega) + \text{Im}\Pi^{-\pm}_{i,c,i,c}(k, \omega) = \mp zJ\sqrt{\delta}(1 - a_0)^2 \frac{\pi}{32}
\]

\[
\times \left[ \left( \frac{\omega}{4J} - 1 \right) I_1(\omega) + \left( \frac{t}{J} + 1 - \frac{\omega}{4J} \right) I_2(\omega) + 4 \frac{t}{J} \left( 1 - a_0 \right) I_3(\omega) \right]
\]

\[
\times \left[ \frac{1}{2\sqrt{\pi}} \frac{k^3}{(1 - \gamma_k^2)^{1/2}} \theta(2q_F - k) + \sqrt{\delta} G^\pm(k) \frac{(\sin^2 k_x + \sin^2 k_y)}{(1 - \gamma_k^2)^{1/2}} \theta(k - 2q_F) \right],
\]

with

\[
G^-(k) = \gamma_k, \quad G^+(k) = 1 + (1 - \gamma_k^2)^{1/2};
\]

\[
I_1(\omega) = \theta(\omega/4J - 1)\theta(2t/J + 1 - \omega/4J),
\]

\[
I_2(\omega) = \theta(\omega/4J - 1 - 2t/J)\theta(4t/J + 1 - \omega/4J),
\]

\[
I_3(\omega) = \theta(2t/J + 1/2 - \omega/4J)\theta(\omega/4J - 1/2),
\]

is one to two orders of magnitude smaller than \( \text{Im}\Pi^{-\pm}_{c,c}(k, \omega) \), while

\[
\text{Re}\Pi^{-\pm}_{c,i,c}(k, \omega^0_k) + \text{Re}\Pi^{-\pm}_{i,c,i,c}(k, \omega^0_k) = zJ\sqrt{\delta}(1 - a_0)^2 \frac{t}{J} \frac{1}{4} \left[ \ln 2 + \frac{a_0}{1 - a_0} \ln \left( 1 + 4 \frac{t}{J} \right) \right]
\]

\[
\times \left[ \frac{1}{2\sqrt{\pi}} \frac{k^3}{(1 - \gamma_k^2)^{1/2}} \theta(2q_F - k) + \sqrt{\delta} \gamma_k \frac{(\sin^2 k_x + \sin^2 k_y)}{(1 - \gamma_k^2)^{1/2}} \theta(k - 2q_F) \right],
\]

is of the same order of magnitude as \( \text{Re}\Pi^{-\pm}_{c,c}(k, \omega) \), though smaller.

As a result, we find that the staggered magnetization (4), calculated with (9),

is strongly reduced with doping, vanishing at a small hole concentration, as illustrated in Figure 2. The reduction of the staggered magnetization is generated by the imaginary part of the self-energies, \( \text{Im}\Pi^{-\pm} \), which imply broadening of
the spin-wave spectral function. The real part of the self-energy, \( \text{Re} \Pi \), gives rise to an increase of the staggered magnetization, which however is one order of magnitude smaller than the decrease due to the imaginary part of the self-energies. The increase of the staggered magnetization arising from the real part of the self-energy results from the coherent motion of holes, while the incoherent motion leads to a decrease, though with a smaller amplitude. We find a critical hole concentration that for \( t/J = 3 \) is \( \delta_c \simeq 0.07 \), whereas for \( t/J = 4 \) is \( \delta_c \simeq 0.05 \). The value for \( \delta_c \), of a few percent, is consistent with experimental data for the copper oxide high-\( T_c \) superconductors. The critical hole concentration \( \delta_c \) is smaller than the hole concentration leading to the vanishing of the spin wave velocity (e.g., \( \delta_{sw} \simeq 0.23 \) for \( t/J = 3 \)), or the concentration at which all spin waves become overdamped (\( \delta^* \simeq 0.17 \) also for \( t/J = 3 \)), in the same approach.\(^5\) This is because the staggered magnetization is specially influenced by the strong damping effects induced by hole motion. Khaliullin and Horsch\(^8\) did not consider damping effects, and concluded that the long-range order disappears as a result of the incoherent motion of holes, however having estimated a decrease of the staggered magnetization due to the incoherent motion of holes that is over one order of magnitude larger than the one calculated by us. Gan and Mila\(^7\) studied the effects of damping on the staggered magnetization, though considering the scattering of spins by holes, i.e. a four-particle interaction with "uncondensed" bosons. Our results, giving the vanishing of the magnetization for a hole concentration where the spin wave velocity is still finite, suggest that, even when long-range order has disappeared, strong AF cor-
relations persist, which allow spin wave excitations to exist, for length scales less than the magnetic correlation length. This is in fact experimentally observed.\textsuperscript{1}

In conclusion, we have shown that the staggered magnetization of a two-dimensional antiferromagnet is significantly reduced as a function of doping due to the strong interaction between holes and spin waves. The motion of holes generates decay of spin waves into "particle-hole" pairs, leading to the destruction of the long-range magnetic order at a small hole concentration. This effect is mainly determined by the coherent motion of holes. The calculated critical hole concentration is in agreement with experimental data for the doped copper oxide high-$T_c$ superconductors.

We also note that NMR measurements, reported in Ref. 16, show damping of the low-energy spin excitations in the doped CuO$_2$ planes due to “particle-hole” excitations, which supports the mechanism for destruction of the long-range order presented in this work.

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References:

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\textsuperscript{1}R.J. Birgeneau and G. Shirane, in Physical Properties of High Temperature Superconductors, ed. D.M. Ginzberg (World Scientific, New Jersey, 1990).

\textsuperscript{2}J. Gan, N. Andrei and P. Coleman, J. Phys.: Condens. Matter 3, 3537 (1991).

\textsuperscript{3}I. R. Pimentel and R.Orbach, Phys. Rev. B 46, 2920 (1992).

\textsuperscript{4}K.W. Becker and U. Muschelknautz, Phys. Rev. 48, 13826 (1993).
5I. R. Pimentel, F. Carvalho Dias, L. M. Martelo and R. Orbach (accepted in Phys. Rev. B).

6T. V. Ramakrishnan, Physica B 163, 34 (1990).

7J. Gan and F. Mila, Phys. Rev. B 44, 12624 (1991).

8G. Khaliullin and P. Horsch, Phys. Rev. B 47, 463 (1993).

9C.L. Kane, P.A. Lee, and N. Read, Phys. Rev. B 39, 6880 (1989).

10F. Marsiglio, A. E. Ruckenstein, S. Schmitt-Rink, and C. M. Varma, Phys. Rev. B 43, 10882 (1991).

11G. Martinez and P. Horsch, Phys. Rev. B 44, 317 (1991).

12Z. Liu and E. Manousakis, Phys. Rev. B 45, 2425 (1992).

13E. Dagotto, Rev. Mod. Phys. 60, 763 (1994).

14N.M. Plakida, V.S. Oudovenko and V. Yu. Yushanhai, Phys. Rev. B 50, 6431 (1994).

15B. Kyung and S. Mukhin, Phys. Rev. B 55, 3886 (1997).

16K.R. Thurber, A.W. Hunt, T. Imai, F.C. Chou, and Y.S. Lee, Phys. Rev. Lett. 79, 171 (1997).
Figure Captions:

FIG. 1. Spin-wave self-energies in the SCBA.

FIG. 2. The staggered magnetization per spin vs hole concentration for different values of t/J.
Fig. 1

\[ \Pi^{-+} \]

\[ \Pi^{--} \]
Fig. 2