Destruction of long-range antiferromagnetic order by hole doping

F. Carvalho Dias* and I. R. Pimentel
Department of Physics and CFMC, University of Lisbon, 1649 Lisboa, Portugal

R. Orbach
Department of Physics, University of California, Riverside, California 92521

(Received 8 March 1999; revised manuscript received 31 August 1999)

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.

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.[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.[2–5] The critical concentration δ_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.[5] The critical hole concentration δ_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 δ_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.[6] The vanishing of the staggered magnetization as a consequence of doping, has already been studied in the *t-J* model by Gan and Mila,[7] considering the scattering of spins by moving holes, and by Khaliullin and Horsch,[8] considering spin disorder introduced by the incoherent motion of holes.

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

$$H_{t-J} = -t \sum_{\langle i,j\rangle,\sigma} (c^\dagger_{i\sigma} c_{j\sigma} + \text{H.c.}) + J \sum_{\langle i,j\rangle} \left(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j \right), \tag{1}$$

where $\mathbf{S}_i = \frac{1}{2} c^\dagger_{i\alpha} \boldsymbol{\sigma}_{\alpha\beta} c_{i\beta}$ is the electronic spin operator, σ are the Pauli matrices, $n_i = n_{i\uparrow} + n_{i\downarrow}$ and $n_{i\sigma} = c^\dagger_{i\sigma} c_{i\sigma}$. To enforce no double occupancy of sites, we use the slave-fermion Schwinger Boson representation for the electron operators $c_{i\sigma} = f^\dagger_i b_{i\sigma}$, where the slave-fermion operator $f^\dagger_i$ creates a hole and the boson operator $b_{i\sigma}$ accounts for the spin, subject to the local constraint $f^\dagger_i f_i + \Sigma_\sigma b^\dagger_{i\sigma} b_{i\sigma} = 2S$. 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 $b_{j\downarrow} = \sqrt{2S}$, respectively, in the up and down sublattices, and the bosons $b_{i\downarrow} = b_i$ and $b_{j\uparrow} = b_j$ are then spin-wave operators on the Néel background. After Bogoliubov-Valatin transformation on the boson Fourier transform $b_{\mathbf{k}} = v_{\mathbf{k}} \beta^\dagger_{-\mathbf{k}} + u_{\mathbf{k}} \beta_{\mathbf{k}}$, with $u_{\mathbf{k}} = \{[(1-\gamma^2_{\mathbf{k}})^{-1/2} + 1]/2\}^{1/2}$, $v_{\mathbf{k}} = -\text{sgn}(\gamma_{\mathbf{k}})\{[(1-\gamma^2_{\mathbf{k}})^{-1/2} - 1]/2\}^{1/2}$, and $\gamma_{\mathbf{k}} = \frac{1}{2}(\cos k_x + \cos k_y)$, we arrive at the effective Hamiltonian

$$\mathcal{H} = -\frac{1}{\sqrt{N}} \sum_{\mathbf{q},\mathbf{k}} f_{\mathbf{q}} f^\dagger_{\mathbf{q}-\mathbf{k}} [V(\mathbf{q},-\mathbf{k}) \beta_{-\mathbf{k}} + V(\mathbf{q}-\mathbf{k},\mathbf{k}) \beta^\dagger_{\mathbf{k}}] + \sum_{\mathbf{k}} \omega^0_{\mathbf{k}} \beta^\dagger_{\mathbf{k}} \beta_{\mathbf{k}}, \tag{2}$$

having $S = 1/2$ and N sites in each sublattice. In Eq. (2), the first term, with $V(\mathbf{q},\mathbf{k}) = zt(\gamma_{\mathbf{q}} u_{\mathbf{k}} + \gamma_{\mathbf{q}+\mathbf{k}} v_{\mathbf{k}})$, 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 $\omega^0_{\mathbf{k}} = (zJ/2)(1-\gamma^2_{\mathbf{k}})^{1/2}$, z being the lattice coordination number $(z=4)$.

The staggered magnetization is given by

$$M = \langle S^z_\uparrow \rangle - \langle S^z_\downarrow \rangle = 2\langle S^z_\uparrow \rangle, \tag{3}$$

with

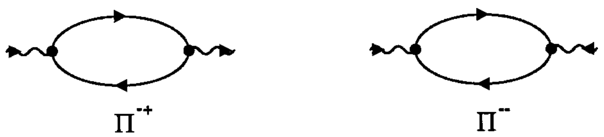

FIG. 1. Spin-wave self-energies in the SCBA.

$$\langle S^z_\uparrow \rangle = \sum_{i \in S(\uparrow)} \langle S^z_i \rangle,$$

where the sum is over the up sublattice. Using the Schwinger boson representation for the spin operator $S^z_i = \frac{1}{2}(c^\dagger_{i\uparrow} c_{i\uparrow} - c^\dagger_{i\downarrow} c_{i\downarrow})$, and the boson condensation associated to the Néel state, one has $S^z_i = (1 - h^\dagger_i h_i)(S - b^\dagger_i b_i)$, which, after Bogoliubov-Valatin transformation, leads to

$$M = (1-\delta)[M_0 - \Delta M], \tag{4}$$

where

$$M_0 = 2\left[NS - \sum_{\mathbf{k}} v^2_{\mathbf{k}} \right] \tag{5}$$

is the staggered magnetization for a pure antiferromagnet, and

$$\Delta M = 2 \sum_{\mathbf{k}} [(u^2_{\mathbf{k}} + v^2_{\mathbf{k}}) \langle \beta^\dagger_{-\mathbf{k}} \beta_{-\mathbf{k}} \rangle + u_{\mathbf{k}} v_{\mathbf{k}} (\langle \beta_{\mathbf{k}} \beta_{-\mathbf{k}} \rangle + \langle \beta^\dagger_{-\mathbf{k}} \beta^\dagger_{\mathbf{k}} \rangle)]. \tag{6}$$

The prefactor in Eq. (4) accounts for the spin dilution due to doping, being negligible for small hole concentrations. In Eq. (5), the order parameter is considerably reduced by quantum fluctuations, to $\simeq 0.6 \times 2NS$. With zero doping the expectation values in Eq. (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 Eq. (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

$$D^{-+}(\mathbf{k}, t-t') = -i \langle \mathcal{T} \beta_{\mathbf{k}}(t) \beta^\dagger_{\mathbf{k}}(t') \rangle,$$

$$D^{+-}(\mathbf{k}, t-t') = -i \langle \mathcal{T} \beta^\dagger_{-\mathbf{k}}(t) \beta_{-\mathbf{k}}(t') \rangle,$$

$$D^{--}(\mathbf{k}, t-t') = -i \langle \mathcal{T} \beta_{\mathbf{k}}(t) \beta_{-\mathbf{k}}(t') \rangle,$$

$$D^{++}(\mathbf{k}, t-t') = -i \langle \mathcal{T} \beta^\dagger_{-\mathbf{k}}(t) \beta^\dagger_{\mathbf{k}}(t') \rangle,$$

where $\langle \ \rangle$ represents an average over the ground state. The spin-wave Green's functions satisfy the Dyson equations: $D^{\mu\nu}(\mathbf{k},\omega) = D^{\mu\nu}_0(\mathbf{k},\omega) + \Sigma_{\gamma\delta} D^{\mu\gamma}_0(\mathbf{k},\omega) \Pi^{\gamma\delta}(\mathbf{k},\omega) D^{\delta\nu}(\mathbf{k},\omega)$, where $\mu,\nu = \pm$. The free Green's functions are $D^{-+}_0(\mathbf{k},\omega) = 1/(\omega - \omega^0_{\mathbf{k}} + i\eta)$, $D^{+-}_0(\mathbf{k},\omega) = 1/(-\omega - \omega^0_{\mathbf{k}} + i\eta)$, $D^{--}_0(\mathbf{k},\omega) = D^{++}_0(\mathbf{k},\omega) = 0$, $(\eta \to 0^+)$, and $\Pi^{\gamma\delta}(\mathbf{k},\omega)$ 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 Fig. 1. These diagrams describe the decay of spin waves into ''particle-hole'' pairs. The spin-wave self-energies can then be written in terms of the hole spectral function, $\rho(\mathbf{q},\omega)$, as

$$\Pi^{\gamma\delta}(\mathbf{k},\omega) = \frac{1}{N} \sum_{\mathbf{q}} U^{\gamma\delta}(\mathbf{k},\mathbf{q}) [Y(\mathbf{q},-\mathbf{k};\omega) + Y(\mathbf{q}-\mathbf{k},\mathbf{k};-\omega)], \tag{7}$$

with

$$Y(\mathbf{q},-\mathbf{k};\omega) = \int_0^{+\infty} d\omega' \int_{-\infty}^0 d\omega'' \frac{\rho(\mathbf{q},\omega')\rho(\mathbf{q}-\mathbf{k},\omega'')}{\omega + \omega'' - \omega' + i\eta},$$

and $U^{--}(\mathbf{k},\mathbf{q}) = U^{++}(\mathbf{k},\mathbf{q}) = V(\mathbf{q},-\mathbf{k}) V(\mathbf{q}-\mathbf{k},\mathbf{k})$, $U^{+-}(\mathbf{k},\mathbf{q}) = V(\mathbf{q}-\mathbf{k},\mathbf{k})^2$, $U^{-+}(\mathbf{k},\mathbf{q}) = V(\mathbf{q},-\mathbf{k})^2$. The relations $\Pi^{-+}(\mathbf{k},\omega) = \Pi^{+-}(-\mathbf{k},-\omega)$ and $\Pi^{--}(\mathbf{k},\omega) = \Pi^{++}(\mathbf{k},\omega)$ are verified, the last implying $D^{--}(\mathbf{k},\omega) = D^{++}(\mathbf{k},\omega)$. The SCBA provides a spectral function for the holes,[9–15] that is composed of a coherent quasiparticle peak and an incoherent continuum, taking the approximate forms, respectively, $\rho^{coh}(\mathbf{q},\omega) = a_0 \delta(\omega - \varepsilon_{\mathbf{q}})$ with $a_0 \simeq (J/t)^{2/3}$, and $\rho^{incoh}(\mathbf{q},\omega) = h\,\theta(|\omega| - zJ/2)\,\theta(2zt + zJ/2 - |\omega|)$ with $h \simeq (1 - a_0)/2zt$, the energies are measured with respect to the Fermi level, and the quasiholes fill up a Fermi surface consisting of pockets, of approximate radius $q_F = \sqrt{\pi\delta}$, located at momenta $\mathbf{q}_i = (\pm\pi/2, \pm\pi/2)$ in the Brillouin zone, the quasiparticle dispersion being, near $\mathbf{q}_i$, written as $\varepsilon_{\mathbf{q}} = \varepsilon_{min} + (\mathbf{q} - \mathbf{q}_i)^2/2m$, with an effective mass $m \simeq 1/J$. The self-energies will then present three contributions, $\Pi^{\gamma\delta}(\mathbf{k},\omega) = \Pi^{\gamma\delta}_{c,c}(\mathbf{k},\omega) + \Pi^{\gamma\delta}_{c,ic}(\mathbf{k},\omega) + \Pi^{\gamma\delta}_{ic,ic}(\mathbf{k},\omega)$, 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 δ.

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

$$\Delta M = -\sum_{\mathbf{k}} \frac{2}{(1-\gamma^2_{\mathbf{k}})^{1/2}} \int_0^{+\infty} \frac{d\omega}{2\pi} [2\,\mathrm{Im}\, D^{+-}(\mathbf{k},\omega) - \gamma_{\mathbf{k}}\, \mathrm{Im}\{D^{++}(\mathbf{k},\omega) + D^{--}(\mathbf{k},\omega)\}]. \tag{8}$$

To lowest order in the hole concentration δ, Eq. (8) gives

$$\Delta M = -\sum_{\mathbf{k}} \frac{2}{(1-\gamma^2_{\mathbf{k}})^{1/2}} \left[-\frac{\gamma_{\mathbf{k}}}{2\omega^0_{\mathbf{k}}} \mathrm{Re}\,\Pi^{--}(\mathbf{k},\omega^0_{\mathbf{k}}) + \int_0^{+\infty} \frac{d\omega}{\pi} \left(\frac{\mathrm{Im}\,\Pi^{-+}(\mathbf{k},\omega)}{(\omega + \omega^0_{\mathbf{k}})^2} + \gamma_{\mathbf{k}} \frac{\mathrm{Im}\,\Pi^{--}(\mathbf{k},\omega)}{\omega^2 - (\omega^0_{\mathbf{k}})^2} \right) \right]. \tag{9}$$

Evaluating Eq. (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

$$\mathrm{Im}\,\Pi^{-\pm}_{c,c}(\mathbf{k},\omega)=zJ\sqrt{\delta}a_0^2\left(\frac{t}{J}\right)^2\frac{1}{\sqrt{\pi}k(1-\gamma_{\mathbf{k}}^2)^{1/2}}F^{-\pm}(\mathbf{k},\omega)\times\{\sqrt{1-s^2(g)}\,\theta(1-|s(g)|)-\sqrt{1-s^2(-g)}\,\theta(1-|s(-g)|)\},$$

with

$$F^{--}(\mathbf{k},\omega)=[\cos(k_xg)+\cos(k_yg)]-\gamma_{\mathbf{k}}[\cos k_x\cos(k_xg)+\cos k_y\cos(k_yg)],$$

$$F^{-+}(\mathbf{k},\omega)=\{(\cos k_x-\cos k_y)[\cos(gk_x)-\cos(gk_y)]/2-(1-\gamma_{\mathbf{k}}^2)^{1/2}[\sin k_x\sin(gk_x)+\sin k_y\sin(gk_y)]-2(1-\gamma_{\mathbf{k}}^2)\},$$

where $s(g)=(1-g)k/2q_F$ and $g=2\omega/Jk^2$, while

$$\mathrm{Re}\,\Pi^{--}_{c,c}(\mathbf{k},\omega_{\mathbf{k}}^0)=-zJ\delta a_0^2\left(\frac{t}{J}\right)^2\frac{\gamma_{\mathbf{k}}k^2}{8[1-\gamma_{\mathbf{k}}^2-(k/2)^4]}\times\frac{(\sin^2k_x+\sin^2k_y)}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}.$$

Regarding the incoherent contributions,

$$\mathrm{Im}\,\Pi^{-\pm}_{c,ic}(\mathbf{k},\omega)+\mathrm{Im}\,\Pi^{-\pm}_{ic,ic}(\mathbf{k},\omega)=\mp zJ\sqrt{\delta}(1-a_0)^2\frac{\pi}{32}\times\left[\left(\frac{\omega}{4J}-1\right)I_1(\omega)+\left(4\frac{t}{J}+1-\frac{\omega}{4J}\right)I_2(\omega)+4\frac{t}{J}\frac{a_0}{(1-a_0)}I_3(\omega)\right]\left[\frac{1}{2\sqrt{\pi}}\frac{k^3}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}\theta(2q_F-k)+\sqrt{\delta}G^{\pm}(\mathbf{k})\frac{(\sin^2k_x+\sin^2k_y)}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}\theta(k-2q_F)\right],$$

with

$$G^{-}(\mathbf{k})=\gamma_{\mathbf{k}},\quad G^{+}(\mathbf{k})=1+(1-\gamma_{\mathbf{k}}^2)^{1/2},$$

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

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

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

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

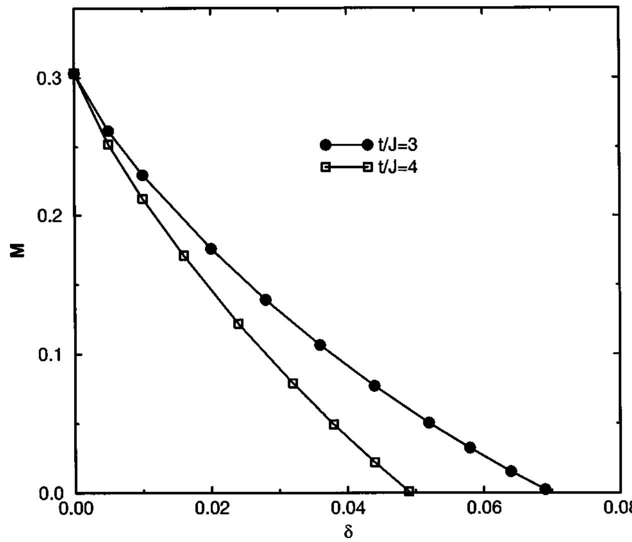

FIG. 2. The staggered magnetization per spin vs hole concentration for different values of t/J.

$$\mathrm{Re}\,\Pi^{--}_{c,ic}(\mathbf{k},\omega_{\mathbf{k}}^0)+\mathrm{Re}\,\Pi^{--}_{ic,ic}(\mathbf{k},\omega_{\mathbf{k}}^0)=zJ\sqrt{\delta}(1-a_0)^2\frac{t}{J}\frac{1}{4}\times\left[\ln 2+\frac{a_0}{1-a_0}\ln\left(1+4\frac{t}{J}\right)\right]\left[\frac{1}{2\sqrt{\pi}}\frac{k^3}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}\times\theta(2q_F-k)+\sqrt{\delta}\gamma_{\mathbf{k}}\frac{(\sin^2k_x+\sin^2k_y)}{(1-\gamma_{\mathbf{k}}^2)^{1/2}}\theta(k-2q_F)\right],$$

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

As a result, we find that the staggered magnetization (4), calculated with Eq. (9), is strongly reduced with doping, vanishing at a small hole concentration, as illustrated in Fig. 2. The reduction of the staggered magnetization is generated by the imaginary part of the self-energies, $\mathrm{Im}\,\Pi^{-\pm}$, which imply broadening of the spin-wave spectral function. The real part of the self-energy, $\mathrm{Re}\,\Pi^{--}$, 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 $\delta_c\simeq0.07$, whereas for $t/J=4$ is $\delta_c\simeq0.05$. The value for δ_c, of a few percent, is consistent with experimental data for the copper oxide high-T_c superconductors. The critical hole concentration δ_c is smaller than the hole concentration leading to the vanishing of the spin-wave velocity (e.g., $\delta_{sw}\simeq0.23$ for $t/J=3$), or the concentration at which all spin waves become overdamped ($\delta^*\simeq0.17$ 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 correlations persist, which allow spin-wave excitations to exist, for length scales less than the magnetic correlation length. This is, in fact, experimentally observed.[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 consistent 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.

We thank T. Imai for bringing Ref. 16 to our attention.

*Electronic address: fdias@alf1.cii.fc.ul.pt

[1] R.J. Birgeneau and G. Shirane, in *Physical Properties of High Temperature Superconductors*, edited by D.M. Ginzberg (World Scientific, Singapore, 1990).
[2] J. Gan, N. Andrei, and P. Coleman, J. Phys.: Condens. Matter **3**, 3537 (1991).
[3] I.R. Pimentel and R. Orbach, Phys. Rev. B **46**, 2920 (1992).
[4] K.W. Becker and U. Muschelknautz, Phys. Rev. B **48**, 13 826 (1993).
[5] I.R. Pimentel, F. Carvalho Dias, L.M. Martelo, and R. Orbach, Phys. Rev. B **60**, 12 329 (1999).
[6] T.V. Ramakrishnan, Physica B **163**, 34 (1990).
[7] J. Gan and F. Mila, Phys. Rev. B **44**, 12 624 (1991).
[8] G. Khaliullin and P. Horsch, Phys. Rev. B **47**, 463 (1993).
[9] C.L. Kane, P.A. Lee, and N. Read, Phys. Rev. B **39**, 6880 (1989).
[10] F. Marsiglio, A.E. Ruckenstein, S. Schmitt-Rink, and C.M. Varma, Phys. Rev. B **43**, 10 882 (1991).
[11] G. Martinez and P. Horsch, Phys. Rev. B **44**, 317 (1991).
[12] Z. Liu and E. Manousakis, Phys. Rev. B **45**, 2425 (1992).
[13] E. Dagotto, Rev. Mod. Phys. **66**, 763 (1994).
[14] N.M. Plakida, V.S. Oudovenko, and V.Yu. Yushanhai, Phys. Rev. B **50**, 6431 (1994).
[15] B. Kyung and S. Mukhin, Phys. Rev. B **55**, 3886 (1997).
[16] K.R Thurber, A.W. Hunt, T. Imai, F.C. Chou, and Y.S. Lee, Phys. Rev. Lett. **79**, 171 (1997).