Theory for Slightly Doped Antiferromagnetic Mott Insulators

T. K. Lee,[1,2] Chang-Ming Ho,[2] and Naoto Nagaosa[3,4]

[1]*Institute of Physics, Academia Sinica, Nankang, Taipei, Taiwan 11529*

[2]*Physics Division, National Center for Theoretical Sciences, P.O. Box 2-131, Hsinchu, Taiwan 300*

[3]*Department of Applied Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan*

[4]*Correlated Electron Research Center, AIST, Tsukuba, Ibaraki 303-0046, Japan*

(Received 18 April 2002; published 10 February 2003)

New trial wave functions, constructed explicitly from the unique Mott insulating state with antiferromagnetic order, are proposed to describe the ground state of a Mott insulator slightly doped with holes or electrons. A rigid band is observed as charged quasiparticles with well-defined momenta being realized in these states. These states have much less superconducting correlations than previously studied ones. Small Fermi patches obtained are consistent with recent experiments on high T_c cuprates doped lightly with holes or electrons.

DOI: 10.1103/PhysRevLett.90.067001 PACS numbers: 74.72.Jt, 75.50.Ee, 79.60.–i

Doping the two-dimensional Mott insulator of the CuO_2 layer is relevant to the physics of high T_c cuprates in the underdoped regime [1,2]. However, recent angle-resolved photoemission spectroscopy (ARPES) results unearth contrasting behaviors between lightly hole-doped $Ca_{2-z}Na_zCuO_2Cl_2$ (Na-CCOC) [3] and electron-doped $Na_{2-x}Ce_xCuO_2$ (NCCO) high T_c cuprates [4]. Although ARPES on the undoped (i.e., $z = x = 0$) insulating state shows an identical energy dispersion of a single hole created below the charge gap, results at a little higher dopings are demonstrated to be different: while a small hole patch (or pocket) is observed to be centered clearly at momentum $(\pi/2, \pi/2)$ in the Na-CCOC at even $z = 0.1$, small electron patches centered at $(\pi, 0)$ and $(0, \pi)$ are observed for $x = 0.04$ in NCCO. The single-hole/electron behavior and its dispersion have been studied using various approaches on the t-t'-t''-J model (see, e.g., [5-9]). However, these studies on doping holes and electrons into the system emphasize the *asymmetry* resulting from the different signs of t' and t'' for the corresponding Hamiltonians. It is unclear whether the same physics is working for these two systems with *different* Hamiltonians. Should one try to construct a different theory when electron-doped cuprates are considered? Furthermore, do the models predict small Fermi surfaces and *quasiparticles?*

There is another puzzle bothering many researchers in this field for many years: So far, most experiments do not support the coexistence of antiferromagnetic long range order (AFLRO) and superconductivity (SC) at low doping. This is far from settled since the absence of coexistence could be due to inhomogeneity [10]. For the t-J type models, it has been shown by several groups [11,12] that at half-filling the projected d-wave superconducting, or the resonating-valence-bond (RVB), state with AFLRO is an excellent wave function (WF). In this state, SC is completely suppressed by the constraint of one particle per site, while the AFLRO survives. These studies also suggest that away from half-filling, the SC revives and the ground state shows both SC and AFLRO [11,13]. However, the recent exact results for the t-t'-t''-J model [14] seem to indicate possible solutions without SC at low doping. Thus there is a need to reexamine this issue.

In this Letter, we propose a theory based on the variational approach to understand these issues. Specific trial wave functions (TWF's) are constructed to describe the ground states of the associated t-t'-t''-J models with lightly doped holes and electrons together. These WF's are generalizations of the single-hole WF first written down by Lee and Shih [6]. In contrast to other TWF's [12], ours are constructed solely from the optimized one at half-filling and include no hopping amplitudes t' and t'' explicitly. However, the important effects of t' and t'' are included. Recent works [15,16] on the t-J model have shown that variational approaches could be made fairly accurate when compared with other techniques. Here we shall focus on the relevant *qualitative* aspects of much importance to understand the physics of cuprates.

At half-filling, the system is described by the Heisenberg Hamiltonian $\mathcal{H}_J = J\sum_{\langle i,j\rangle}(\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4}n_i n_j)$, where $\langle i, j\rangle$ denotes nearest-neighbor (n.n.) sites. Each site is occupied by only one single electron. Following Ref. [6], we assume three mean-field order parameters: the staggered magnetization $m_s = \langle S_A^z\rangle = -\langle S_B^z\rangle$, where the lattice is divided into A and B sublattices, the uniform bond order parameters $\chi = \langle\sum_\sigma c_{i\sigma}^\dagger c_{j\sigma}\rangle$, and d-wave RVB (d-RVB) one $\Delta = \langle c_{j\downarrow}c_{i\uparrow} - c_{j\uparrow}c_{i\downarrow}\rangle$ if i and j are n.n. sites in the x direction and $-\Delta$ for the y direction.

We shall apply the standard variational Monte Carlo (VMC) method [12] that enforces the local constraint exactly. The WF has the form

$$|\Psi_0\rangle = P_d\left[\sum_{\mathbf{k}}(A_{\mathbf{k}}a_{\mathbf{k}\uparrow}^\dagger a_{-\mathbf{k}\downarrow}^\dagger + B_{\mathbf{k}}b_{\mathbf{k}\uparrow}^\dagger b_{-\mathbf{k}\downarrow}^\dagger)\right]^{N_e/2}|0\rangle, \quad (1)$$

where N_e is the total number of electrons and coefficients $A_{\mathbf{k}} = (E_{\mathbf{k}} + \xi_{\mathbf{k}})/\Delta_{\mathbf{k}}$ and $B_{\mathbf{k}} = -(E_{\mathbf{k}} - \xi_{\mathbf{k}})/\Delta_{\mathbf{k}}$ with $E_{\mathbf{k}} = (\xi_{\mathbf{k}}^2 + \Delta_{\mathbf{k}}^2)^{1/2}$. Here $\Delta_{\mathbf{k}} = \frac{3}{4}J\Delta d_{\mathbf{k}}$ with

$d_{\mathbf{k}} = \cos k_x - \cos k_y$. Energy dispersions for the two spin-density-wave (SDW) bands are $\pm\xi_{\mathbf{k}} = \pm[\epsilon_{\mathbf{k}}^2 + (Jm_s)^2]^{1/2}$ with $\epsilon_{\mathbf{k}} = -\frac{3}{4}J\chi(\cos k_x + cos k_y)$. Operators of the lower and upper SDW bands are related to the original electron operators by $a_{\mathbf{k}\sigma} = \alpha_{\mathbf{k}} c_{\mathbf{k}\sigma} + \sigma\beta_{\mathbf{k}} c_{\mathbf{k}+\mathbf{Q}\sigma}$ and $b_{\mathbf{k}\sigma} = -\sigma\beta_{\mathbf{k}} c_{\mathbf{k}\sigma} + \alpha_{\mathbf{k}} c_{\mathbf{k}+\mathbf{Q}\sigma}$, respectively. Here $\mathbf{Q} = (\pi, \pi)$, $\alpha_{\mathbf{k}}^2 = \frac{1}{2}[1 - (\epsilon_{\mathbf{k}}/\xi_{\mathbf{k}})]$, and $\beta_{\mathbf{k}}^2 = \frac{1}{2}[1 + (\epsilon_{\mathbf{k}}/\xi_{\mathbf{k}})]$. The projection operator P_d enforces the constraint of no doubly occupied (or vacant) sites for cases with finite hole (or electron) doping (see below). At half-filling, N_e equals the total number of sites, N_s. Notice that the sum in $|\Psi_0\rangle$ is taken over the sublattice Brillouin zone (SBZ). In this WF there are two variational parameters: Δ/χ and m_s/χ. In the absence of staggered order, m_s, this is exactly the usual d-RVB WF. Without Δ this WF describes the SDW state [12]. This TWF gives energy $-0.332J$ per bond which is within 1% of the best estimate of the ground state energy of the Heisenberg model.

As holes or electrons are doped into the system, we consider the Hamiltonian $\mathcal{H} = \mathcal{H}_{t-t'-t''} + \mathcal{H}_J$ including hoppings between n.n. (t), second n.n. (t'), and third n.n. (t''). Because of the constraint, doping into the half-filled system creates a different kind of *holes*: *empty holes* ($0e$ hole) for hole doping and *two-electron-occupied holes* ($2e$-hole) for electron doping [17]. States in the two cases are in one-to-one correspondence after a particle-hole transformation is made [9]. However, because of the Fermi statistics, the exchange of a single spin with a $2e$ hole has an extra *minus* sign as compared to the $0e$ hole. Hence, the only difference between the hole- and electron-doped t-t'-t''-J model is $t'/t \to -t'/t$ and $t''/t \to -t''/t$ after we change the $c_{i\sigma}$ on the B sublattice sites to $-c_{i\sigma}$ [9]. With all these, we then treat the hole- and electron-doped cases in the same manner as the Lee-Shih WF originally proposed only for a single hole. The VMC results presented below are for $J/t = 0.3$, $t'/t = -(+)0.3$, and $t''/t = +(-)0.2$ in the hole (electron) doped case following Ref. [9].

The TWF for one doped hole with momentum $\mathbf{q}$ and $S_z = 1/2$ is constructed to have $(N_s/2) - 1$ singlet pairs of electrons and a single unpaired electron with momentum $\mathbf{q}$ and $S_z = 1/2$,

$$|\Psi_1\rangle = P_d c_{\mathbf{q}\uparrow}^{\dagger}\left[\sum_{\mathbf{k}}{}'(A_{\mathbf{k}} a_{\mathbf{k}\uparrow}^{\dagger} a_{-\mathbf{k}\downarrow}^{\dagger} + B_{\mathbf{k}} b_{\mathbf{k}\uparrow}^{\dagger} b_{-\mathbf{k}\downarrow}^{\dagger})\right]^{(N_s/2)-1}|0\rangle.$$

The prime on the summation symbol indicates that the momentum $\mathbf{q}$ is excluded from the sum if $\mathbf{q}$ is within the SBZ; otherwise, $\mathbf{q} - \mathbf{Q}$ is excluded. $|\Psi_1\rangle$ is thus explicitly constructed from the optimized half-filled WF $|\Psi_0\rangle$, and it does not contain any information about hoppings, t', t'', or explicitly t, of the doped hole or electron. However, the effect of t is included in the RVB order parameter χ which describes the large quantum fluctuation and spin singlet formation. t' and t'' are compatible with AFLRO; hence there is little effect for them to be included in TWF. But, as shown below, they are important in determining the energy or the dispersion.

The energy dispersion obtained from $|\Psi_1\rangle$ for one doped hole has been shown by Lee and Shih [6] to agree very well with that of several other numerical calculations [18]. For the case of having an extra up-spin electron with momentum $\mathbf{q}$ doped into the half-filled state, the energy dispersion can be calculated with this same WF $|\Psi_1\rangle$ by noting the change of signs of t'/t and t''/t.

The variational energies for one doped electron are shown as black dots in Fig. 1. This result agrees well with that of self-consistent Born approximation (SCBA) [5]. The optimal variational parameters are $(\Delta/\chi, m_s/\chi) = (0.25, 0.125)$. The ground state is at momentum $\mathbf{q} = (\pi, 0)$. The VMC results can be fitted simply by $E_{1\mathbf{k}} = E_{\mathbf{k}} - 2t_{\text{eff}}(\cos k_x + \cos k_y) - 4t'_{\text{eff}} \cos k_x \cos k_y - 2t''_{\text{eff}}[\cos(2k_x) + \cos(2k_y)]$ with parameters described in the caption of Fig. 1. The dispersion thus seems to be simply the combination of the mean-field band at half-filling and the coherent hoppings [6].

To examine further the physical properties of $|\Psi_1\rangle$, we calculated the momentum distribution function (MDF) $\langle n_\sigma^h(\mathbf{k})\rangle$ for the ground state of a single hole with momentum $\mathbf{q} \equiv \mathbf{Q}/2$ and $S_z = 1/2$. Results are shown in Figs. 2(a) and 2(b). Note that the dips or pockets at $\mathbf{Q}/2$ and *antidips* at $-\mathbf{Q}/2$ found in Ref. [7] for the exact results of 32 sites are also clearly seen here. It is quite amazing that $|\Psi_1\rangle$, including no t' and t'', not only produces the correct energy dispersions for a single doped hole or electron it also provides a correct picture about the momentum distribution.

The MDF's $\langle n_\sigma^e(\mathbf{k})\rangle$ for electron-doped systems could also be calculated from $|\Psi_1\rangle$ by using $\langle n_\sigma^e(\mathbf{k})\rangle = 1 - \langle n_{-\sigma}^h(\mathbf{Q} - \mathbf{k})\rangle$. The results for the ground state of a single doped electron with momentum $\mathbf{k} = (\pi, 0)$ and spin $S_z = 1/2$ are shown in Figs. 2(c) and 2(d). There are peaks at $\mathbf{k} = (\pi, 0)$ and an *antipeak* at $(0, \pi)$.

Now we shall generalize the Lee-Shih WF $|\Psi_1\rangle$ to the case of two holes. The simplest possible way is just to take

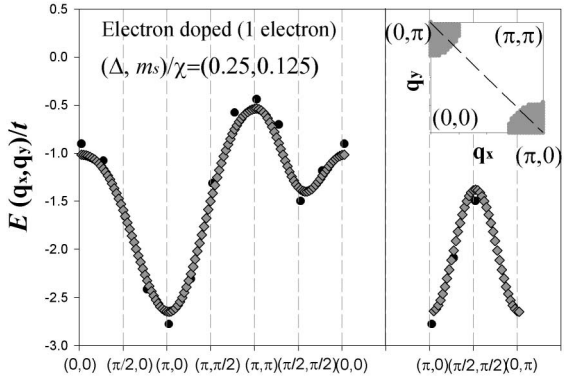

FIG. 1. Energy dispersion of one electron in the t-t'-t''-J model on an 8×8 lattice. Black dots are VMC results by using $|\Psi_1\rangle$. The fitted dispersion $E_{1\mathbf{k}} - E_0$ are plotted as gray diamonds with parameters $\chi = 6.92$, $\Delta = 2.71$, $m_s = 18.84$, $E_0 = 7.43$, $t_{\text{eff}} = 0.06$, $t'_{\text{eff}} = -0.15$, $t''_{\text{eff}} = 0.1$. Inset: patches in one quadrant of BZ by filling the fitted dispersion in the main figure up to about 3% doping.

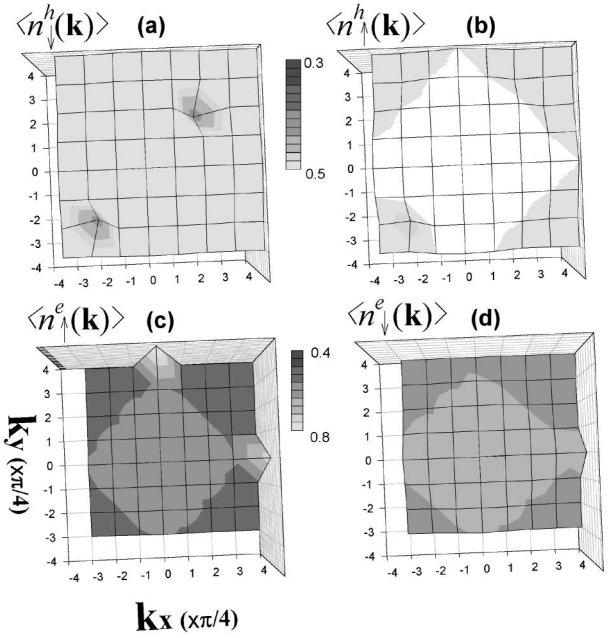

FIG. 2. $\langle n^{h(e)}_\sigma(\mathbf{k})\rangle$ for a single hole, (a) and (b), and electron, (c) and (d), in the 8×8 t-t'-t''-J model. A scale is shown between each set. The darker area indicates smaller values.

out the unpaired spin from $|\Psi_1\rangle$ if we are interested in the state with zero total momentum and $S_z = 0$, which turns out to be the lowest energy state. The TWF for two holes with momenta $\mathbf{q}$ and $-\mathbf{q}$ is

$$|\Psi_2\rangle = P_d\left[\sum_{\mathbf{k}}^{I}(A_{\mathbf{k}}a^\dagger_{\mathbf{k}\uparrow}a^\dagger_{-\mathbf{k}\downarrow} + B_{\mathbf{k}}b^\dagger_{\mathbf{k}\uparrow}b^\dagger_{-\mathbf{k}\downarrow})\right]^{(N_s/2)-1}|0\rangle.$$

Note that the momentum $\mathbf{q}$ is not included in the summation. It is most surprising to find that although $|\Psi_2\rangle$ has zero total momentum irrespective of $\mathbf{q}$, its energy varies with the missing momentum or *the hole momentum* $\mathbf{q}$. The dispersion shown as circles in Fig. 3 is very similar to that of a single electron as shown in Fig. 1. The state with momentum $\mathbf{q} = (\pi, 0)$ has the lowest energy for two electrons. Δ/χ and m_s/χ are the same as $|\Psi_1\rangle$.

The energy dispersion of two holes, shown as diamonds in Fig. 3, has an almost identical form as that of a single hole and the minimum is at $\mathbf{q} = \mathbf{Q}/2$. The lowest energy obtained is $-26.438(3)t$ which is much lower than the variational energy, $-25.72(1)t$, using the TWF applied by Himeda and Ogata [13]. Even if we include t' and t'', the variational energy $-25.763(7)t$ is still much higher than ours [19]. In the inset of Fig. 3 the hopping amplitudes for n.n., second n.n., and third n.n. are shown for one hole and two holes as a function of $\mathbf{q}$. It shows that the values of two holes are almost twice that of one hole. The MDF for this state (not shown) has dips at $\mathbf{Q}/2$ and $-\mathbf{Q}/2$. This is in good agreement with the exact result [14] for the t-t'-t''-J model with two holes in 32 sites.

It is then straightforward to write down the TWF for three holes with momenta $\mathbf{q}$, $\mathbf{q}'$, and $-\mathbf{q}'$ by excluding both $\mathbf{q}$ and $\mathbf{q}'$ from the sum in $|\Psi_1\rangle$. Just like the case with

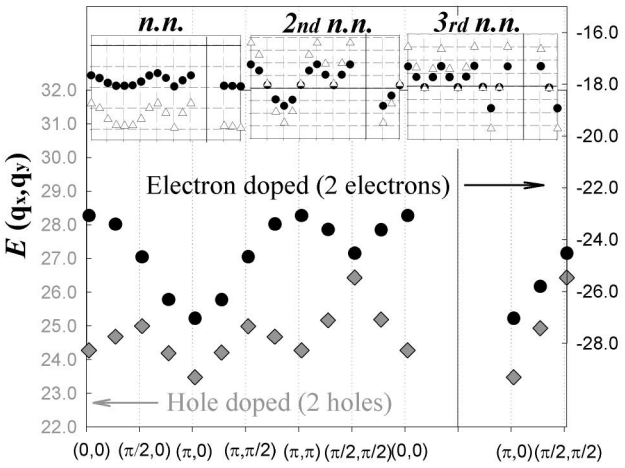

FIG. 3. Energy dispersions of two $2e$ holes (diamond) and two $0e$ holes (circles) in the t-t'-t''-J model obtained by using $|\Psi_2\rangle$ where a total minus sign has been multiplied to the hole doped case. Insets: Comparisons of n.n. (left), second n.n. (middle), and third n.n. (right) hopping amplitudes for 1 (filled circle) and 2 (unfilled triangle) doped charge carriers. Vertical dashed lines in the insets represent the momentum points as in the main figure. The amplitude difference between two horizontal dashed lines is 0.02. Horizontal solid lines in the insets represent zero amplitude.

two holes or two electrons, energy dispersions are now proportional to the sum of the three single-hole energies at momenta $\mathbf{q}$, $\mathbf{q}'$, and $-\mathbf{q}'$.

In Table I, values of staggered magnetization $m = N_s^{-1}\sum_i(-1)^i S_i^z$ for several $0e$ hole and $2e$ hole concentrations are compared. The same variational parameters are used. It is clear that the preference of $\mathbf{Q}/2$ for $0e$ holes causes larger disturbance of the AF order than for the electron-doped case where $2e$ holes with momentum $(\pi, 0)$ shows much less influence on the AF order. This is consistent with previous work [9,20] and experimental results that AF phase is more stable for electron doping than hole doping [21].

So far, based on the t-t'-t''-J model we have proposed a TWF to describe the low energy states of slightly doped antiferromagnetic Mott insulators. Exactly the same TWF's are proposed to account for the behavior of both hole doping and electron doping, after we employed the particle-hole transformation. Different energy dispersions for these two cases are due to the different signs of t'/t and t''/t. Rigid band [22] and quasiparticle behavior are demonstrated for both cases. The theory provides

TABLE I. Staggered magnetization m for 1, 2, and 3 doped holes and electrons in an 8×8 lattice. The parameters used here are $(\Delta/\chi, m_s/\chi) = (0.25, 0.125)$.

Doping number	0	1	2	3
Hole doped	0.365(1)	0.353(1)	0.329(1)	0.285(1)
Electron doped	0.365(1)	0.372(1)	0.348(1)	0.332(4)

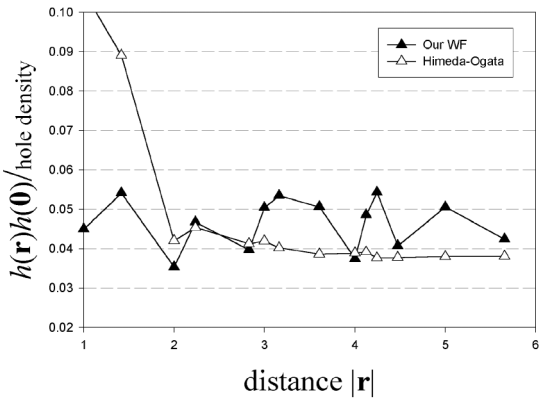

FIG. 4. Hole-hole correlation functions with 4 doped holes in the 8×8 lattice. The result obtained using our WF, $|\Psi_2\rangle$ with momenta $(\pi/2, \pi/2)$ and $(\pi/2, -\pi/2)$ excluded in the sum, is compared with that of WF in Ref. [13] with $\mu = -0.025$.

an explanation of recent ARPES results. In lightly hole-doped cuprates, small Fermi pocket is around $\mathbf{Q}/2$. In electron-doped cuprates, the patch is around $(\pi, 0)$ as shown in the inset of Fig. 1 with doping at about 3%. This result cannot be obtained without t'/t and t''/t.

In addition to having reproduced many numerical results obtained by exact diagonalization, SCBA, etc., for one or two holes or electrons, our TWF's provide several new results. The momentum of each hole (0e hole or 2e hole) is here specifically introduced, and they obey nice energy dispersions. Hence there are only small Fermi surfaces or patches [22]. Most previous WF's use the chemical potential to control the number of holes. Holes are distributed evenly on equal energy surfaces determined by the chemical potential; this leads to large Fermi surfaces. These WF's could be considered as a linear combination of our WF's; thus they have higher variational energies. Another important property of our WF's is that holes are essentially independent of each other as they obey the same energy dispersions (with very little renormalization of parameters). Exactly because this *quasiparticlelike* property is unchanged after doping, our WF has AFLRO but very little superconducting pairing correlations. The presence of a superconducting state certainly will change the excitation spectra. In particular, the d-wave SC, which coexists with AFLRO in some of the previous variational studies, should have low energy excitations along the nodes. This is certainly not seen in our TWF's. In addition, the holes are not attractive to each other in our WF's. In Fig. 4 the hole-hole correlation function for our TWF and the WF used in Ref. [13] are compared. The lack of attraction between holes is consistent with Leung's low energy states obtained exactly for two holes in 32 sites [14]. Long range d-wave pairing correlation [12] for our TWF and that in [13] are, on average, about 0.002 and 0.018, respectively. Thus the d-RVB pairing for spins assumed by our WF's are not in any way implying the pairing of charges when there is AFLRO. The possible superconductivity for larger doping, however, is beyond the scope of this paper, and left for future studies.

We are grateful to Dr. A. Damascelli, Dr. F. Ronning and Professor Z.-X. Shen for sharing their ARPES data prior to publication, and Professor C.-T. Shih for some results and invaluable discussions. T. K. L. is supported by Grant No. NSC91-2112-M-001-011 (R.O.C.) and N. N. is supported by Priority Areas Grants and Grant-in-Aid for COE research from the Ministry of Education, Culture, Sports, Science and Technology of Japan.

[1] P.W. Anderson, in *Frontiers and Borderlines in Many-Particle Physics*, edited by R. A. Broglia and J. R. Schrieffer (North-Holland, Amsterdam, 1988).
[2] P. A. Lee, in *More is Different: Fifty Years of Condensed Matter Physics*, edited by N. P. Ong and R. N. Bhatt (Princeton University Press, Princeton, 2001).
[3] A. Damascelli, Z.-X. Shen, and Z. Hussain (unpublished); F. Ronning *et al.*, Science **282**, 2067 (1998); F. Ronning *et al.* (unpublished).
[4] N. P. Armitage *et al.*, Phys. Rev. Lett. **88**, 257001 (2002).
[5] T. Xiang and J. M. Wheatley, Phys. Rev. B **54**, R12 653 (1996).
[6] T. K. Lee and C. T. Shih, Phys. Rev. B **55**, 5983 (1997).
[7] A. L. Chernyshev, P.W. Leung, and R. J. Gooding, Phys. Rev. B **58**, 13 594 (1998); T. Tohyama and S. Maekawa, Supercond. Sci. Technol. **13**, R17 (2000).
[8] A. Nazarenko *et al.*, Phys. Rev. B **51**, 8676 (1995).
[9] T. Tohyama and S. Maekawa, Phys. Rev. B **49**, 3596 (1994); R. J. Gooding, K. J. E. Vos, and P.W. Leung, *ibid.* **50**, 12 866 (1994).
[10] J. Burgy *et al.*, Phys. Rev. Lett. **87**, 277202 (2001).
[11] T. K. Lee and S. P. Feng, Phys. Rev. B **38**, 11 809 (1988); G. J. Chen *et al.*, Phys. Rev. B **42**, 2662 (1990).
[12] For the introduction and early work of variational WF study, see C. Gros, Ann. Phys. (N.Y.) **189**, 53 (1989); also see, for the more recent, e.g., H. Yokoyama and M. Ogata, J. Phys. Soc. Jpn. **65**, 3615 (1996).
[13] A. Himeda and M. Ogata, Phys. Rev. B **60**, R9935 (1999).
[14] P.W. Leung, Phys. Rev. B **65**, 205101 (2002).
[15] S. Sorella *et al.*, Phys. Rev. Lett. **88**, 117002 (2002).
[16] C. T. Shih, Y. C. Chen, and T. K. Lee, Phys. Rev. B **57**, 627 (1998).
[17] D. G. Clarke, Phys. Rev. B **48**, 7520 (1993).
[18] F. Marsiglio *et al.*, Phys. Rev. B **43**, 10 882 (1991); G. Martinez and P. Horsch, *ibid.* **44**, 317 (1991); Z. Liu and E. Manousakis, *ibid.* **45**, 2425 (1992); P.W. Leung, B. O. Wells, and R. J. Gooding, *ibid.* **56**, 6320 (1997); E. Dagotto *et al.*, Phys. Rev. Lett. **73**, 728 (1994).
[19] Actually, the lowest energy for Himeda-Ogata–type TWF is obtained when its parameters are chosen to replicate our TWF. Details are to be reported elsewhere.
[20] G. Martins *et al.*, Phys. Rev. B **64**, 180513 (2001).
[21] H. Takagi, Y. Tokura, and S. Uchida, Physica (Amsterdam) **162–164C**, 1001 (1989); Phys. Rev. Lett. **62**, 1197 (1989); G. M. Luke *et al.*, Phys. Rev. B **42**, 7981 (1990); K. Yamada *et al.*, J. Phys. Chem. Solids **60**, 1025 (1999).
[22] E. Dagotto *et al.*, Phys. Rev. Lett. **74**, 310 (1995).