Orbital Currents in Extended Hubbard Models of High-T_c Cuprate Superconductors

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Motivated by the recent report of broken time-reversal symmetry and zero momentum magnetic scattering in underdoped cuprates, we investigate under which circumstances orbital currents circulating inside a unit cell might be stabilized in extended Hubbard models that explicitly include oxygen orbitals. Using Gutzwiller projected variational wave functions that treat on an equal footing all instabilities, we show that orbital currents indeed develop on finite clusters and that they are stabilized in the thermodynamic limit if additional interactions, e.g., strong hybridization with apical oxygens, are included in the model.

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Despite intensive efforts in the past 20 years, the physics of high-T_c superconductors remains largely mysterious [1]. This is especially true of the pseudogap phase of underdoped cuprates, for which various explanations have been put forward ranging from preformed superconducting pairs [2] to the existence of orbital currents (OCs) with [3] or without [4,5] broken translational symmetry. The latter case has received considerable attention due, on one hand, to recent neutron experiments [6] indicating the presence of magnetic moments, compatible with the translation-invariant pattern of currents predicted by Simon and Varma [4] and, on the other hand, to Kerr effect measurements [7] showing evidence of time-reversal symmetry breaking. Current (flux) phases have been first proposed for the single-band Hubbard model [2,8–10] but have been found unstable by slave bosons and numerical calculations. Interestingly, OCs were also found to be relevant in a two-band system [11]. In ladder models, where the existence of such phases can be checked in a more controlled way, it was found that somewhat special interactions, more complex than local ones, are needed to stabilize them [12,13]. The resulting phases break the translational symmetry of the lattice, leading to a staggered flux pattern. Similar staggered patterns were advocated as a potential explanation of the pseudogap phase [3] [d-density wave (DDW) phase]. To stabilize flux phases that do not break the translational symmetry, it seems worthwhile to go beyond the single-band Hubbard model and to consider the so-called three-band Hubbard model in which oxygen orbitals are explicitly taken into account. This model has been tested for superconducting instabilities early on, pointing to some differences with the single-band model [14]. Fluctuation exchange analysis on the two-dimensional system showed no sign of instability to staggered flux phases [15]. The one-dimensional (ladder) case has first been investigated at half filling, when the system is essentially an insulator [16]. More recently, a low energy analysis for the three-band model on a ladder has been performed and showed that in a certain range of doping flux phases were indeed stabilized [17]. These phases exhibit a $2k_F$ staggered order parameter, quite natural in one dimension, for the currents, but with a symmetry different from that of the DDW. In two dimensions, a mean-field analysis [4] of the three-band model has suggested the existence of translationally invariant current patterns when the Cu-O nearest-neighbor repulsion is strong enough. This result has not been confirmed by exact diagonalizations [18,19] on small clusters of an effective t-J model. However, the mapping of the three-band Hubbard model on this t-J model can be justified only in the limit of very large oxygen on-site energy, a situation not realized in the cuprates [20]. Moreover, because of the three atoms per unit cell, the exact diagonalizations have been performed for clusters with only a few unit cells, and the relevance of the results for the thermodynamic limit is far from guaranteed, in particular, because the filling that was considered on this small cluster $x = 12.5\%$ is leading to a polarized ground state with finite momentum, which is not representative of the physics on large scales. Therefore, further investigations of the three-band Hubbard model are clearly called for.

In this Letter, we perform a variational Monte Carlo (VMC) investigation of the three-band Hubbard model based on a Gutzwiller projected wave function that allows for the possibility of OCs. This provides a method free from numerical limitations even for large system sizes, for which current instabilities are treated on an equal footing with other instabilities.

We find that, on intermediate system sizes, a flux phase circulating between copper and oxygens is stabilized. This phase has the same symmetry (Θ_2; see Fig. 1) as the phase found in the mean-field solution [4]. Other symmetries or phases that break the translational symmetry are much higher in energy. However, as the system size gets larger,

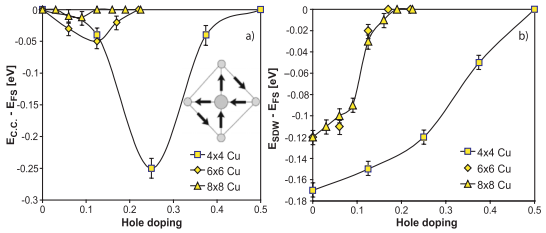

FIG. 1 (color online). Energy of (a) the best orbital current wf and of (b) the best spin-density wave wf. The reference was taken as the projected Fermi sea (full Jastrow projector). Inset of (a): The symmetry of the best current phase found (Θ_2 pattern in the notations of [4]). Although the OC phase is stable on an intermediate size 16 copper lattice, the energy gain is strongly reduced when the size increases. To the accuracy of the calculation, we did not find any energy optimization on a 100 Cu lattice for the OC instability.

the energy gain decreases strongly, making it unclear whether such a phase would survive in the thermodynamic limit. We then propose modifications of the Hamiltonian that take into account apical oxygens or three-body terms and which strongly stabilize such current patterns. The initial three-band model is defined by the Hamiltonian:

$$H = \sum_{(i,j)\sigma} (t_{i,j} c^{\dagger}_{i\sigma} c_{j\sigma} + \text{H.c.}) + \sum_{\alpha=p,d} U_{\alpha} \hat{n}_{\alpha\uparrow} \hat{n}_{\alpha\downarrow} + \Delta_p \sum_{p,\sigma} \hat{n}_{p\sigma} + V_{dp} \sum_{d,p} \hat{n}_d \hat{n}_p, \quad (1)$$

where the sum over couples (i, j) runs over nearest Cu-O neighbor pairs and next-nearest O-O neighbor pairs and c stands for p or d orbitals depending on the site; $t_{i,j}$ stand for the hopping matrix elements of magnitude t_{dp} and t_{pp}, respectively, for Cu-O and O-O matrix elements. Δ_p, U_d, U_p, and V_{dp} denote the atomic energy of the O-p orbitals, the on-site repulsions in the Cu-d and O-p orbitals, and the nearest-neighbor repulsion between Cu-d and O-p orbitals, respectively. A realistic set of parameters found by local-density approximation (LDA) calculations [20–22] is $U_d = 10.5$ eV and $U_p = 4$ eV, $|t_{dp}| = 1.3$ eV and $|t_{pp}| = 0.65$ eV, $\Delta_p = 3.5$ eV, and $V_{dp} = 1.2$ eV, and, unless specified otherwise, these values are used in the following. Note that we work in hole notations. The phase factor that comes from the hybridization of the p-d orbitals gives to the t_{ij} a nonhomogeneous sign. Around each copper atom, there is one Cu-O-Cu plaquette with three minus signs. Moreover, the product of the hopping signs around all of the Cu-O-Cu plaquettes is -1 in hole notations. Interestingly, a simple gauge transformation involving a double copper unit cell leads to $t_{ij} = -|t_{ij}|$.

This model has already been investigated with VMC simulations [23,24], but the wave functions used did not allow for current instabilities. By contrast, the wave function we consider throughout this work is constructed from the ground state of the Hofstadter-like mean-field Hamiltonian:

$$H^{\text{MF}} = \sum_{(i,j)} t_{ij} \chi_{ij} e^{i\theta_{ij}} c^{\dagger}_{i\sigma} c_{j\sigma} + \Delta_p^{\text{var}} \sum_{p\sigma} \hat{n}_{p\sigma} + \sum_i \mathbf{h_i} \mathbf{S_i}, \quad (2)$$

where $\chi_{ij} > 0$ and θ_{ij}, Δ_p^{var}, and $\mathbf{h_i}$ are real variational parameters. The local magnetic field $\mathbf{h_i}$ allows one to consider antiferromagnetism. The 16 variables χ_{ij} and θ_{ij} are independent within one copper unit cell. To deal with staggered order, we multiply a subset of the variational parameters by -1. When $\theta_{ij} \neq 0$, the order parameters are associated with an external flux which leads to the circulation of the holes. In this case the Green function is complex: $\langle c^{\dagger}_i c_j \rangle = |\langle c^{\dagger}_i c_j \rangle| \exp(i\phi_{ij})$. We treat the correlations with a spin and charge Jastrow factor: $\mathcal{J} = \exp(\sum_{i,j=1,N} v^c_{|i-j|} n_i n_j) \exp(\sum_{i,j=1,N} v^S_{|i-j|} S^z_i S^z_j)$, where all $v^c_{|i-j|}$ and $v^S_{|i-j|}$ are considered as free variational parameters. We are mainly interested in the charge current observable. The conservation of the density $\frac{\delta n_i}{\delta t} = 0 = \frac{\hbar e}{c}[H, n_i] = \sum_{\langle i,j \rangle} J_{i,j}$ leads to the current operator on a link: $J_{ij} = \sum_{\sigma} t_{ij} |\langle c^{\dagger}_{i\sigma} c_{j\sigma} \rangle| \sin(\phi_{ij})$. However, the current conservation law is not satisfied by our variational ansatz since it is not an exact eigenstate of (1). The *mean-field current*, defined as $J^{\text{MF}}_{ij} := \sum_{\sigma} t_{ij} |\chi_{ij}| |\langle c^{\dagger}_{i\sigma} c_{j\sigma} \rangle| \sin(\theta_{ij} + \phi_{ij})$, is a conserved quantity, but, in general, J and J_{MF} need not be oriented in the same way. In particular, we find that the O-O currents have different signs for J and J_{MF}. The phase $e^{i\theta_{ij}}$ thus gives the direction of J^{MF} but *not* the direction of J, in general, which has to be computed explicitly. In order to overcome this difficulty and to impose the current conservation at each vertex of the lattice, we apply on the wave function (wf) an additional complex Jastrow factor [25]: $\mathcal{J}_c = \exp(\sum_{i=1,N} i\alpha_i n_i)$. The kinetic energy of the extended wf is $\langle T \rangle = \sum_{\langle i,j,\sigma \rangle} t_{ij} |\langle c^{\dagger}_{i\sigma} c_{j\sigma} \rangle| \cos(\theta_{ij} + \alpha_j - \alpha_i)$. We emphasize that the variables α_i are, in general, not able to cancel the flux θ_{ij}. The minimization of the variational parameters introduced in (1) and in the real Jastrow is performed using a stochastic minimization procedure [26,27] in which the parameters of the uncorrelated part of the wf and the Jastrow parameters are minimized at the same time. This method allows one to deal with a large number of parameters since the gradients are calculated all at the same time during a simulation. The new parameters are then calculated using the obtained gradients, and the procedure is iterated until convergence. At each step, the parameters $\{\alpha_i\}$ of the complex Jastrow are determined by finding the ground state of a classical 2D XY spin model. Once our wave function is optimized, we measure the physical observables, normalized by the number of copper atoms in the lattice. To benchmark our wave function, we have compared its properties with those obtained by exact diagonalizations for 10 holes on a small 8 copper lattice with periodic boundary conditions (25% hole doping), with very encouraging results (see Table I). Details will be presented elsewhere [29].

TABLE I. Variational energies of the different wave functions (wfs) compared with exact diagonalizations (Lanczos, first line) on an 8-copper lattice with 10 holes and $S^z = 0$. We show the total energy (E_{tot}), the kinetic energy of the Cu-O (T_{dp}) and of the O-O links (T_{pp}), the on-site repulsion energy of the d (U_d) and p (U_p) orbitals, and the expectation values of the charge gap operator (Δ_p) and of the Coulomb repulsion between the d and p orbitals (V_{dp}). The results obtained for a simple Jastrow wf (J), by applying one Lanczos step (LS/J) and the fixed node approximation [28] on a simple Jastrow wf (FN/J), are also shown (the calculation of the off-diagonal operators T_{dp} and T_{pp} within the fixed-node approximation is more involved and is beyond the scope of the present study). The comparison shows that our wf captures quite well the low energy physics of the model. The variance $\langle(\frac{H}{N})^2\rangle - (\langle\frac{H}{N}\rangle)^2$ of our best variational ansatz (LS/J) is 0.018.

wf	E_{tot}	T_{dp}	T_{pp}	U_d	U_p	Δ_p	V_{dp}
L	−1.13821	−3.100	−0.796	0.267	0.083	1.775	0.632
J	−1.0775(1)	−3.060	−0.830	0.261	0.081	1.834	0.640
LS/J	−1.1153(1)	−3.140	−0.837	0.265	0.087	1.864	0.644
FN/J	−1.1112(5)			0.269	0.087	1.773	0.631

We now turn to larger lattices, using open boundary conditions to avoid the frustration of the α_i variables. We have performed calculations on lattices ranging from 16 to 64 copper sites. The gains in energy for the best antiferromagnetic [spin-density wave (SDW)] instability and for flux phases are compared in Fig. 1 [30]. Note that these orders are somewhat exclusive: Considering both simultaneously does not lead to any detectable gain in energy. At zero doping we find that the Néel magnetic long-range order is stabilized. By introducing doping, we considered as a first approximation only the $Q = (\pi, \pi)$ pitch vector for the spin-density wave, even though the pitch vector is most likely doping-dependent [31] or other instabilities like stripes can occur [23]. Nevertheless, the long-range correlations contained in the Jastrow factor allow for a decent treatment of the spin correlations. Indeed, for our best variational wave function, the magnetic order parameter $M = \lim_{r\to\infty}\sqrt{\langle S_i^z S_{i+r}^z\rangle}$ is $\approx 60\%$ of the classical value. Using this wf as a guiding function for the fixed node calculations, we find a slightly higher magnetic order (62%). These values compare well with the 60% obtained by quantum Monte Carlo calculations in the one-band Heisenberg model [32]. Note that, in the three-band Hubbard model, the magnetic instability is strongly dependent on the O-O hopping integral t_{pp}. Despite the good agreement for the order parameter, our variational treatment overestimates the magnetic instability, which disappears only for 13% doping, instead of the experimentally observed $x = 2\%$ for the cuprates. VMC tends indeed to overestimate the stability of SDW phases because the alternating magnetization allows one to avoid double occupancy in the uncorrelated part of the wf, which on one side costs kinetic energy but on the other side is better than a pure local Gutzwiller projection that reduces the kinetic energy much more dramatically.

Let us now turn to the current instability. On intermediate system sizes such as the 16 Cu lattice, we find that an OC phase with a symmetry Θ_2 (see the inset in Fig. 1) is stabilized. Our wave function thus shows a tendency to flux phase that does not exist in the corresponding one-band model. However. the gain in energy strongly decreases as the size increases and seems, within the accuracy of our calculation, to vanish in the thermodynamic limit. Taken literally, this suggests that the OC phase is not stable and that the system has only short-range correlations. However, we of course cannot rule out that some fine-tuning of the parameters could stabilize this phase [33] or that the energy gain would be much smaller than our statistical error. Two points should be emphasized. First, regardless of the size of the system, we find consistently that the Θ_2 symmetry is the one with the lowest variational energy. Other symmetries such as the Θ_1 phase or the DDW patterns are unstable within our variational approach in the whole range of doping. Second, varying the Cu-O interaction seems to have no major effect on the stability of the current patterns, in contrast with what one could have expected on the basis of the mean-field solution [4]. As one can check from an analysis of a model with only kinetic energy [29], the relative sign of the transfer integrals around a loop is a crucial parameter for an instability towards long-range current correlations. Indeed, when the sign of t_{pp} is reversed, J and J_{MF} are oriented in the same direction, and the current pattern is stable. For the model (1), the mean-field approximation creates such a change of sign with the correction to the bare t_{pd} coming from the decoupling of the Cu-O repulsion term [4], but the VMC study shows that other terms in the decoupling seem to play an important role as well. Given our VMC calculations for the three-band Hubbard model of CuO_2 planes and the experimental evidence, it is natural to check if other terms not included into this simple version of a multiband model could produce such an effect and stabilize orbital currents. One obvious possibility is to include correlated hopping, as already done successfully for the single-band Hubbard model. This indeed leads to a strong increase of the tendency to develop OCs [29]. Note, however, that the magnitude of these terms is difficult to assess. A second and interesting possibility is to generalize the model by including apical oxygens. Indeed, the relative signs of the hopping around purely oxygen plaquettes allow *a priori* for orbital currents. We have thus repeated the calculation for Cu-O layers including apical oxygens above and below each Cu atom as well as the Cu-$d_{3z^2-r^2}$ orbitals. The bare parameters have been taken from the LDA [34] performed with the crystal structure of the insulating parent compound, and the analysis has been carried out as a function of the distance between the copper and the apical oxygen, modeled by a renormalization parameter η that multiplies the hopping integrals involving the apical oxygens. As

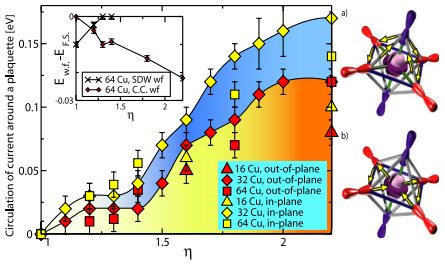

FIG. 2 (color online). Circulation of current around a $p_x - p_z - p_y$ plaquette (triangles) and around a $p_x - d_{3z2-r2} - p_y$ plaquette (squares) measured in our best variational ansatz for the eight-band Hubbard model. The phenomenological parameter η renormalizes the amplitudes of the out-of-plane transfer integrals. The symmetry of the OC pattern is Θ_2 as follows: There are (a) two out-of-plane current loops in the upper pyramid and (b) two current loops in the Cu plane. Finally, the current pattern in the lower pyramid (not shown) is obtained by a horizontal mirror reflection of the upper pyramid. The calculations were done at $x = 0.125$ hole doping, and with periodic boundary conditions. Inset: Energies for both the SDW wf and the circulating current (CC) ones showing the stabilization of the CC phase for $\eta > 1.2$.

expected from the signs of the hopping, a calculation with $x = 0.125$ hole doping confirms that orbital currents involving the apical oxygen or the Cu-$d_{3z^2-r^2}$ indeed develop [35] according to the pattern of Fig. 2. These currents are quite small for the bare values of the parameters, but they acquire significant values when $\eta > 1$, with a steep increase above $\eta = 1.4$ [36]. Interestingly enough, the current circulating in the $p_x - p_z - p_y$ plaquette leads to a tilted moment, which would provide a natural explanation for the out-of-plane moment that was observed in the neutron experiments [6]. Whether the structural changes induced by doping on the position of the apical oxygens reported by some authors can produce the required renormalization of the hopping integrals remains to be seen [37].

After completion of this work, an experimental investigation of mercury compounds [38] has revealed a magnetic signal in the pseudogap phase which is compatible with orbital currents including the apical oxygen, strengthening further the experimental relevance and interest of our proposal. In parallel, a μSR experiment [39] found no evidence for broken time-reversal symmetry in lanthanum compounds, suggesting that orbital currents are sensitive to the details of the electronic structure, also in agreement with the message of the present Letter.

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