No Evidence for Spontaneous Orbital Currents in Numerical Studies of Three-Band Models for the CuO Planes of High Temperature Superconductors

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We have numerically evaluated the current-current correlations for three-band models of the CuO planes in high-T_c superconductors at hole doping $x = 1/8$. The results show no evidence for the orbital current patterns proposed by Varma. If such patterns exist, the associated energy is estimated to be smaller than 5 meV per link even if $\epsilon_p - \epsilon_d = 0$. Assuming that the three-band models are adequate, quantum critical fluctuations of such patterns hence cannot be responsible for phenomena occurring at significantly higher energies, such as superconductivity or the anomalous properties of the strange metal phase.

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The problem of understanding high-T_c superconductivity has been something like a holy grail to the field of condensed matter physics for the past two decades [1]. It has turned out to be an exceedingly difficult problem, with much of the effort invested just deepening the mysteries, but it has also led to a plethora of new developments extending far beyond the field. Many of the experimental techniques used to study the systems, like angle resolved photo emission spectroscopy (ARPES) or scanning tunneling microscopy (STM), have undergone revolutions with regard to resolution and data processing. The theory of superconductivity in high-T_c cuprates has been found many times, but while individuals believe to have the theory, there is no consensus what the theory should be. Many ideas, even though too general to qualify as complete theories of the cuprates, have inspired a vast amount of research in both high-T_c and other areas. Most prominently among them are the notions of a resonating valence bond (RVB) state [2] including the gauge theories of spin liquids [3], and the notion of quantum criticality [4]. There have been, however, a few concise proposals which make falsifiable predictions. Masterpieces among them have been the theory of anyon superconductivity [5], the proposal of kinetic energy savings through interlayer tunneling [6], the SO(5) theory of a common order parameter for superconductivity and magnetism [7], and a more recent proposal that the anomalous properties of the cuprates may be due to quantum critical fluctuations of current patterns formed spontaneously in the CuO planes [8,9]. This last proposal is further investigated in this Letter.

The idea of a spontaneous symmetry breaking through orbital currents was, as with so many major advances in physics, motivated by experiment. The normal state of the cuprates at optimal doping shows a behavior which can be classified as quantum critical and has been rather adequately described by a phenomenological theory called marginal Fermi liquid [10]. This phenomenology suggests a quantum critical point (QCP) at a hole doping level of $x_c \approx 0.19$, an assumption consistent with a significant body of experimental data [11–15]. Critical fluctuations around this point would then be responsible for the anomalous properties of the strange metal phase and provide the pairing force responsible for the superconducting phase which hides the QCP.

Interpreting the phase diagram in these terms, one is immediately led to ask what the phase to the left of the QCP, i.e., for $x < x_c$, might be. The theory would require a spontaneously broken symmetry beyond the global U(1) symmetry broken through superconductivity (which is often erroneously referred to as a broken gauge symmetry [16]). In addition, as the fluctuations are assumed to determine the phase diagram up to temperatures of several hundred Kelvin, the characteristic energy scale of the correlations inducing this symmetry violation must be at least of the same order of magnitude. No definitive evidence of such a broken symmetry has been found up to now, even though several possibilities have been suggested. These include stripes [17], a d-density wave [18], and most recently a checkerboard charge density wave [19].

The general consensus is that the low energy sector of the three-band Hubbard model proposed for the CuO planes [see (1) below] [20] reduces to a one-band $t - t' - J$ model, with parameters $t \approx 0.44$, $t' \approx -0.06$, and $J \approx 0.128$ (energies throughout this Letter are in eV) [21–25]. For the undoped CuO planes, the formal valances are Cu^{2+} and O^{2-}. As the electron configuration of Cu atoms is [Ar] $3d^{10}4s^1$, this implies one hole per unit cell, which will predominantly occupy the $3d_{x^2-y^2}$ orbital. As the on-site potential ϵ_p in the O $2p_x$ and $2p_y$ orbitals relative to the Cu $3d_{x^2-y^2}$ orbital is generally assumed to be of the order of $\epsilon_p = 3.6$ (with $\epsilon_d = 0$), and hence smaller than the on-site Coulomb repulsion $U_d \approx 10.5$ for a second hole in the $3d_{x^2-y^2}$ orbital, it is clear that additional holes doped into the planes will primarily reside on the oxygens. The maximal gain in hybridization energy is achieved by placing the additional hole in a combination of the surrounding O $2p_x$ and $2p_y$ orbitals with the same symmetry as the original hole in the Cu $3d_{x^2-y^2}$ orbital, which requires antisymmetry of the wave function in spin space; i.e., the two holes must form a singlet. This picture is strongly supported by data from NMR [26] and even more directly from spin-resolved photoemission [27]. In the effective one-band $t - t' - J$ model description of the CuO planes, these singlets constitute the "holes" moving in a background of spin $1/2$ particles localized at the Cu sites.

In contrast to this picture, Varma [8,9] has proposed that the additional holes doped in the CuO planes do not hybridize into Zhang-Rice singlets, but give rise to circulating currents on O-Cu-O triangles, which align into a planar pattern as shown in Fig. 1. He assumes that the interatomic Coulomb potential $V_{\rm pd}$ is larger than the on-site potential $\epsilon_{\rm p}$ of the O $2p$ orbitals relative to the Cu $3d_{x^2-y^2}$ orbitals, an assumption which is not consistent with the values generally agreed on [see the list below (1)]. Making additional assumptions, Varma has shown that the circulating current patterns are stabilized in a mean-field solution of the three-band Hubbard model. The orbital current patterns break time-reversal symmetry (T) and the discrete fourfold rotation symmetry on the lattice, but leave translational symmetry intact. The current pattern is assumed to disappear at a doping level of about $x_c \approx 0.19$. The phenomenology of CuO superconductors, including the pseudogap and the marginal Fermi liquid phase, are assumed to result from this symmetry breaking and critical fluctuations around this QCP, as outlined above.

Motivated by this proposal, several experimental groups have looked for signatures of orbital currents or T violation in CuO superconductors. While there is no agreement between different groups regarding the manifestation of T violation in ARPES studies [28,29], a recent neutron scattering experiment by Fauqué *et al.* [30] indicates magnetic order within the unit cells of the CuO planes. Their results are consistent with Varma's proposal and call the validity of the one-band models into question.

In a recent article, Aji and Varma [31] have mapped the four possible directions of the current patterns in each unit cell onto two Ising spins and investigated the critical fluctuations. Within this framework, the coupling between and the transverse fields for these Ising spins decide

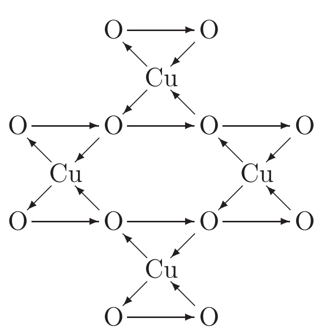

FIG. 1. Orbital current pattern proposed by Varma.

whether or under which circumstances the model displays long-range order in the orbital currents.

We hence undertook to estimate these couplings through numerical studies of finite clusters containing 8 unit cells, i.e., 8 Cu and 16 O sites, and periodic boundary conditions (which do not frustrate but should enhance the correlations). The total number of holes on our cluster was taken $N = 9$ (5 up-spins and 4 down-spins), corresponding to a hole doping of $x = 1/8$. We had hoped that the energy associated with a domain wall, which may be implemented through a twist in the boundary conditions, would provide information regarding the coupling aligning the orbital currents in neighboring plaquettes, while the splitting between the lowest energies for a finite system would provide an estimate for the transverse field. The result of our endeavors, however, is a daunting disappointment: the coupling is zero within the error bars of our numerical experiments.

Let us now report our numerical studies in detail. To begin with, we wish to study the three-band Hubbard Hamiltonian $H = H_t + H_U$ with

$$
\begin{aligned}
H_t = {} & \sum_{i,\sigma} \epsilon_{\rm p} n^{\rm p}_{i,\sigma} - t_{\rm pd} \sum_{\langle i,j\rangle,\sigma} (d^\dagger_{i,\sigma} p_{j,\sigma} + p^\dagger_{j,\sigma} d_{i,\sigma}) \\
& - t_{\rm pp} \sum_{\langle i,j\rangle,\sigma} (p^\dagger_{i,\sigma} p_{j,\sigma} + p^\dagger_{j,\sigma} p_{i,\sigma}) \\
& + V_{\rm pd} \sum_{\langle i,j\rangle,\sigma,\sigma'} n^{\rm d}_{i,\sigma} n^{\rm p}_{j,\sigma'}, \\
H_U = {} & U_{\rm p} \sum_i n^{\rm p}_{i,\uparrow} n^{\rm p}_{i,\downarrow} + U_{\rm d} \sum_i n^{\rm d}_{i,\uparrow} n^{\rm d}_{i,\downarrow},
\end{aligned}
\tag{1}
$$

where $\langle\, ,\rangle$ indicates that the sums extend over pairs of nearest neighbors, while $d_{i,\sigma}$ and $p_{j,\sigma}$ annihilate holes in Cu $3d_{x^2-y^2}$ or O $2p$ orbitals, respectively. Hybertsen *et al.* [23] calculated $t_{\rm pd} = 1.5$, $t_{\rm pp} = 0.65$, $U_{\rm d} = 10.5$, $U_{\rm p} = 4$, $V_{\rm pd} = 1.2$, and $\epsilon_{\rm p} = 3.6$.

In order to be able to diagonalize (1) for 24 sites, we need to truncate the Hilbert space. A first step is to eliminate doubly occupied sites, which yields the effective three-band $t - J$ Hamiltonian

$$
\begin{aligned}
& H_{\rm eff} = P_G H_t P_G + H_J \quad \text{with} \\
& H_J = J_{\rm pd} \sum_{\langle i,j\rangle} (\mathbf{S}^{\rm p}_i \cdot \mathbf{S}^{\rm d}_j - \tfrac{1}{4}) + J_{\rm pp} \sum_{\langle i,j\rangle} (\mathbf{S}^{\rm p}_i \cdot \mathbf{S}^{\rm p}_j - \tfrac{1}{4}),
\end{aligned}
\tag{2}
$$

where

$$
J_{\rm pd} = 2t^2_{\rm pd}\left(\frac{1}{U_{\rm d} - \epsilon_{\rm p}} + \frac{1}{U_{\rm p} + \epsilon_{\rm p}}\right), \qquad J_{\rm pp} = \frac{4t^2_{\rm pp}}{U_{\rm p}},
$$

and the sums in H_J are limited to pairs where both neighbors are occupied by holes. P_G eliminates configurations with more than one hole on a site. The dimension of the $S^z_{\rm tot} = \frac{1}{2}$ subsector is with 164 745 504 just within our capabilities.

Let us now turn to our results for the current-current correlations in the ground state (situated at the M point in

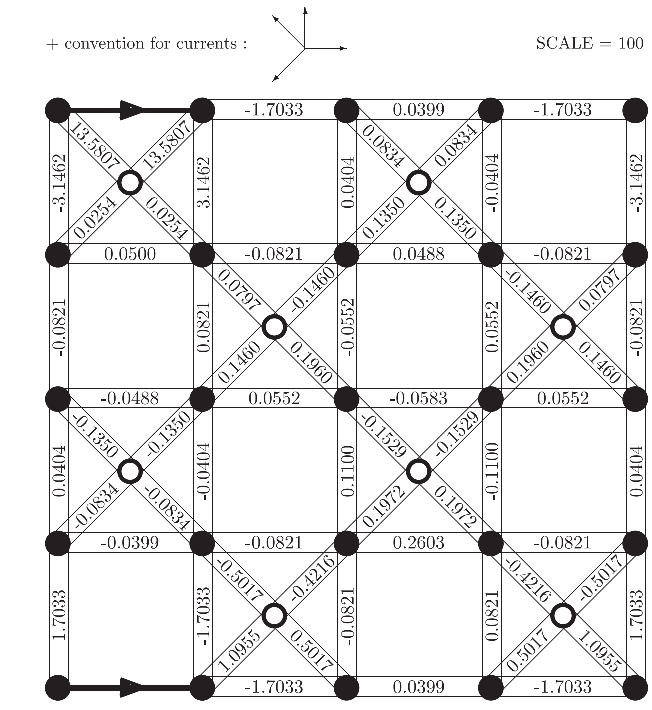

FIG. 2. Current-current correlations $\langle j_{k,k+\hat{x}} j_{l,m} \rangle$ multiplied by 10^2 for the ground state of (2) with $\epsilon_p = 3.6$ on a 24 site cluster (8 Cu = open circles, 16 O = filled circles) with periodic boundary conditions (PBCs). The reference link is indicated in the top and (due to the PBCs) bottom left corner.

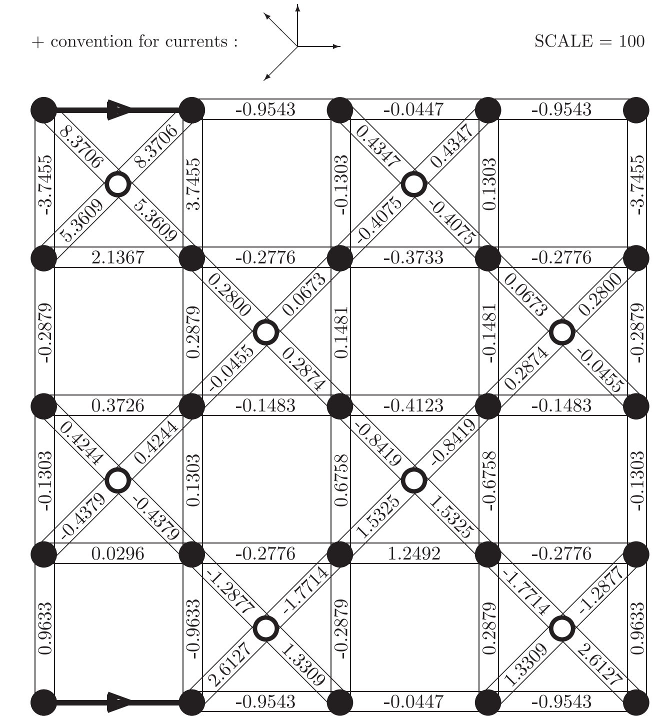

FIG. 3. As in Fig. 2, but with $\epsilon_p = 0$. Except for the vertical links, positive correlations indicate alignment with the current pattern shown in Fig. 1.

the Brillouin zone) for our 24 site cluster with 9 holes and parameters as calculated by Hybertsen, where no orbital current patterns are expected. With the current operator for, e.g., an O-O link given by

$$j_{k,l} = it_{\mathrm{pp}} \sum_{\sigma} (p_{l,\sigma}^{\dagger} p_{k,\sigma} - p_{k,\sigma}^{\dagger} p_{l,\sigma}), \tag{3}$$

the correlations function $\langle j_{k,k+\hat{x}} j_{l,m} \rangle$ with an O-O link as reference link is depicted in Fig. 2. As expected, the correlations fall off rapidly, and there is no indication of order.

The crucial result is that the correlations change continuously and only quantitatively, but not really qualitatively, as ϵ_p is lowered from 3.6 to 1.8, 0.9, 0.4, and finally to $\epsilon_p = 0$, where the result is shown in Fig. 3 (the ground state is now twofold degenerate and situated at the Γ point). This is the situation for which Varma has proposed that the current pattern sketched in Fig. 1 would occur. Figure 3, by contrast, shows no alignment of the currents.

The numerical experiments for the finite cluster can, as a matter of principle, never rule out that a symmetry is violated spontaneously, as this would require an infinite system. We can use our results, however, to put an upper bound on the size of the spontaneous currents, and hence the energy associated with these currents. If a current pattern as sketched in Fig. 1 were to exist, the current correlation $\langle j_{k,k+\hat{x}} j_{l,l+\hat{x}} \rangle$ for links far away from each other in a rotationally invariant ground state should approach $\frac{1}{2}\langle \hat{x}|j_{k,k+\hat{x}}|\hat{x}\rangle^2$, where $|\hat{x}\rangle$ denotes a state with a spontaneous current pointing in $\hat{x}$ direction (the factor $\frac{1}{2}$ arises because by choosing our reference link in x direction, we effectively project onto two of the four possible directions for the current pattern). From the values of $10^2\langle j_{k,k+\hat{x}} j_{l,l+\hat{x}} \rangle$ for the four horizontally connected links in the center of Fig. 3, $+0.3726$, -0.1483, -0.4123, and -0.1483, which should all be positive if a current pattern were present, we estimate $10^2\langle j_{k,k+\hat{x}} \rangle^2 < 0.20$ and hence $\langle \hat{x}|j_{k,k+\hat{x}}|\hat{x}\rangle^2 < 4 \times 10^{-3}$ as an upper bound for a current pattern we are unable to detect through the error bars of our numerical experiment. We now denote $\langle \hat{x}|j_{k,k+\hat{x}}|\hat{x}\rangle$ by j_{pp}.

We roughly estimate the kinetic energy $\varepsilon_{\mathrm{pp}}$ per link associated with a spontaneous current j_{pp} of this magnitude using $j_{\mathrm{pp}} = n_{\mathrm{p}} v$ and $\varepsilon_{\mathrm{pp}} = \frac{1}{2} n_{\mathrm{p}} m v^2$ with $m = 1/2t_{\mathrm{pp}}$, where n_{p} is the hole density on the oxygen sites ($n_{\mathrm{p}} = 0.33$ for the state of Fig. 3), and obtain

$$\varepsilon_{\mathrm{pp}} \approx \frac{j_{\mathrm{pp}}^2}{4t_{\mathrm{pp}} n_{\mathrm{p}}} < 5 \times 10^{-3}.$$

A similar analysis for the CuO links, using data not shown here, yields with $10^2\langle j_{k,k+\hat{x}+\hat{y}} \rangle^2 < 1.0$ and hence $j_{\mathrm{pd}}^2 < 10^{-2}$ (there is no factor $\frac{1}{2}$ in this case) and $n_{\mathrm{d}} = 0.47$ an estimate of

$$\varepsilon_{\mathrm{pd}} \approx \frac{j_{\mathrm{pd}}^2}{4t_{\mathrm{pd}} \sqrt{n_{\mathrm{p}} n_{\mathrm{d}}}} < 4 \times 10^{-3}.$$

We conclude that while we cannot rule out that orbital current patterns exist, we can rule out that they are responsible for the superconductivity, the properties of the pseudogap phase, or the anomalous normal state properties extending up to temperatures of several hundred Kelvin, as the energy associated with the spontaneous loop currents is less than 5 meV per link if such currents exist. Note that this energy is not the condensation energy E_c per unit cell, but a positive contribution to the energy of the current carrying state, which would have to be (more than) offset by other contributions (like the energy gain from aligning the circulating currents according to the pattern Varma proposed) if such a state were realized. We would expect the transition temperature T_c of such a state to be of the order of the effective coupling of the Ising spins introduced by Aji and Varma [31] and hence smaller than 5 meV, while we would expect $E_c \ll T_c$.

In this work, we have assumed that the CuO planes are adequately described by the three-band Hubbard model (1), but we have allowed $\epsilon_{\rm p}$ to be much smaller than generally agreed upon, and based our estimates on the extreme and to our purposes most unfavorable value $\epsilon_{\rm p} = 0$. (Note that the ordered antiferromagnetic phase in undoped cuprates requires a finite $\epsilon_{\rm p}$.) Numerical data not presented here show that our conclusions remain intact if we set $J_{\rm pd} = J_{\rm pp} = 0$ or/and double the value of the repulsion $V_{\rm pd}$, which generates the orbital currents in Varma's mean-field calculation. They also hold for other low energy states for the finite system (e.g., as situated at the M point in the Brillouin zone for $\epsilon_{\rm p} = 0$).

Nonetheless, we should keep in mind that any analysis of a model can only reach a conclusion valid for this model. The question of whether current patterns exist in CuO superconductors can ultimately only be settled by experiment. We consider it likely, however, that an eventual consensus among experiments will confirm our conclusion.

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