Instability towards Staggered Loop Currents in the Three-Orbital Model for Cuprate Superconductors

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We present evidence for the existence of a spontaneous instability towards an orbital loop-current phase in a multiorbital Hubbard model for the CuO$_2$ planes in cuprates. Contrary to the previously proposed $\theta_{11}$ phase with intra-unit cell currents, the identified instability is towards a staggered pattern of intertwined current loops. The orbitally resolved current pattern thereby shares its staggered character with the proposal of $d$-density wave order. The current pattern will cause a Fermi surface reconstruction and the opening of a pseudogap. We argue that the pseudogap phase with time-reversal symmetry breaking currents is susceptible to further phase transitions and therefore offers a route to account for axial incommensurate charge order and a polar Kerr effect in underdoped cuprates.

There is now considerable evidence in underdoped cuprate high-temperature superconductors for a cascade of phase transitions, starting at high temperatures with the pseudogap onset at $T^*$, followed by incommensurate charge order (ICO) at $T_{co} < T^*$ and superconductivity at $T_c < T_{co}$. In addition, broken time-reversal symmetry has been associated with $T^*$ and a Kerr rotation is measured below a temperature $T_{Kerr}$ with $T_{Kerr} \sim 0.75T^*$ over wide doping range. At present, there is no unifying theory that explains this intriguing sequence of transitions.

Ultrasound spectroscopy suggests that $T^*$ corresponds to a true thermodynamic phase transition\cite{1}. Below $T^*$ spin-polarized neutron scattering experiments detected weak magnetic moments\cite{2-4}. These moments appear to preserve the translational symmetry of the lattice, and led to the proposal of intra-unit cell loop currents (LCs)\cite{5}. However, the so-called $\theta_{11}$ LC phase by itself has difficulty explaining the partial gapping of charge excitations\cite{6}. While variational methods favored the existence of LC phases in finite clusters\cite{7,8}, alternative numerically exact analyses reported no evidence for the $\theta_{11}$ phase\cite{9,10}.

The Kerr effect\cite{11,12} that sets in below $T_{Kerr}$ is further evidence for time-reversal symmetry breaking, but also requires that mirror symmetries be broken\cite{13}. Throughout much of the cuprate phase diagram, $T_{Kerr}$ and $T_{co}$\cite{14,22} are close, which has motivated further proposals in which fluctuating charge-\cite{15,23} or pair-density wave states\cite{24,25} generate spontaneous current patterns with broken mirror symmetries. These scenarios assume a hierarchy of transitions associated with distinct symmetry breakings needed to form the fully ordered density-wave state. Other proposals follow the common theme that the pseudogap results from the competition between two or more order parameters\cite{26,29}.

FIG. 1: (Color online) Staggered pattern of spontaneous loop currents. Open circle,“x” and “y” symbols denote Cu$d_{x^2-y^2}$, Op$_x$ and Op$_y$ orbitals, respectively. Currents along $p$-$d$ bonds (black arrows) are about three times stronger than those in $p$-$p$ bonds (green arrows).

The ICO phase involves predominantly a charge transfer between oxygen orbitals in the CuO$_2$ planes\cite{19,21,30,31}. This challenges notions of immutable CuO$_2$ bands, and points to the necessity to employ multiorbital models for the ICO phase\cite{32,34}. Here, we support this reasoning and show that orbital resolved intra-unit cell physics is important throughout the pseudogap regime.

In this letter we report the results of an unbiased calculation for a three-band model of CuO$_2$ planes which verifies the existence of an instability towards a staggered pattern of intertwined LCs (Fig. 1). This “$\pi$LC” phase is different from the anticipated $\theta_{11}$ phase, but shares its ordering wavevector $Q = (\pi, \pi)$ with the earlier phenomenological proposal of LCs in the $d$-density wave (DDW) state\cite{35,36}. In the $\pi$LC phase the Fermi surface reconstructs to form hole pockets with a concomitant pseudogap-like structure in the electronic spectrum. For realistic parameters, the ICO reported previously for the same model Hamiltonian\cite{33} is subleading to the $\pi$LC instability. Yet, the presence of staggered order
The Hamiltonian is  $\hat{H}_0 = \sum_{\mathbf{k}\sigma} \Psi_{\mathbf{k}\sigma}^\dagger \mathbf{H}_0(\mathbf{k}) \Psi_{\mathbf{k}\sigma}$, with

$$
\mathbf{H}_0(\mathbf{k}) = \begin{pmatrix}
\epsilon_d & 2t_{pd}s_x & -2t_{pd}s_y \\
2t_{pd}s_x & \epsilon_p & -4t_{pp}s_x s_y \\
-2t_{pd}s_y & -4t_{pp}s_x s_y & \epsilon_p
\end{pmatrix}
$$

where $\Psi_{\mathbf{k}\sigma}^\dagger = [\hat{c}_{\mathbf{k}d\sigma}^\dagger, \hat{c}_{\mathbf{k}p\sigma}^\dagger, \hat{c}_{\mathbf{k}p\sigma}^\dagger]$, and the indices $d, x$ and $y$ stand for Cu, $O_{px}$ and $O_{py}$ orbitals with energies $\epsilon_d$ and $\epsilon_p$, respectively. $s_{x,y} = \sin(k_{x,y}/2)$, and $t_{pd}$ and $t_{pp}$ are the hopping amplitudes between $p-d$ and $p-p$ orbitals. We set $t_{pd} = 1.66eV$ which, in what follows, defines the unit of energy.

The intra- ($U_\alpha$) and inter-orbital ($V_{i\alpha,j\beta}$) Coulomb interactions are

$$
\hat{H}' = \sum_{i\sigma,j\sigma',\alpha} \left[ \delta_{i\alpha,j\beta} (1 - \delta_{\sigma,\sigma'}) U_\alpha + \frac{V_{i\alpha,j\beta}}{2} \right] \hat{n}_{i\sigma} \hat{n}_{j\beta\sigma'},
$$

where $i, j$ are unit cell indices, $\alpha, \beta$ are orbital labels and $V_{i\alpha,j\beta}$ is nonzero for nearest-neighbors only. Throughout, we suppress the spin index $\sigma$, set $t_{pp} = 0.5$, $\epsilon_d - \epsilon_p = 2.5$, $U_d = 9$, $U_p = 3$, $V_{pd} = 2.2$, and $V_{pp} = 1$ unless otherwise stated. The Hamiltonian $\hat{H} = \hat{H}_0 + \hat{H}'$ is thus the conventional three-band model of cuprates\cite{37} with a typical parameter set \cite{38}.

The current operator associated with the bond between sites $i\alpha$ and $j\beta$ is $\hat{J}_{i\alpha,j\beta} = -i t_{i\alpha,j\beta} (\hat{c}_{i\alpha}^\dagger \hat{c}_{j\beta} - \hat{c}_{j\beta}^\dagger \hat{c}_{i\alpha})$ where $t_{i\alpha,j\beta}$ is the corresponding hopping matrix element. If $(\hat{J}_{i\alpha,j\beta}) \neq 0$ then current flows from $j\beta$ to $i\alpha$. In momentum space, the current operator along bond $m$ is given by $\hat{J}_m(\mathbf{\mathbf{k}}) = -i \sum_k h_{m\mathbf{k}\sigma\sigma'}(\mathbf{k},q) \hat{c}_{\mathbf{k}\sigma}^\dagger \hat{c}_{\mathbf{k}+\mathbf{q}\sigma'} - h_{m\mathbf{k}\sigma\sigma'}^\dagger(\mathbf{k},q) \hat{c}_{\mathbf{k}+\mathbf{q}\sigma'}^\dagger \hat{c}_{\mathbf{k}\sigma}$, where $\alpha, \beta$ are the orbitals associated with the bond and the matrix elements of the current operators $h_{m\mathbf{k}\sigma\sigma'}(\mathbf{k},q)$ are listed in the Supplementary Material. As shown in Fig. 2 there are eight distinct bonds on the CuO$_2$ lattice. Accordingly the current susceptibility

$$
\chi_{mn}(\mathbf{q}, i\omega) = \int_0^\beta d\tau e^{i\omega\tau} \langle \hat{J}_m(\mathbf{q},\tau)\hat{J}_n(-\mathbf{q}, 0) \rangle,
$$

is an 8x8 matrix, where $i\omega = 2\pi \ell T$ denotes the Matsubara frequencies. Each matrix element can be decomposed as

$$
\chi_{mn} = \chi_{\alpha\beta\alpha'\beta'} - \chi_{\alpha\beta'\alpha'\beta'} - \chi_{\alpha\beta\alpha'\beta'} + \chi_{\alpha'\beta'\alpha'\beta'}
$$

where

$$
\chi_{\alpha\beta\alpha'\beta'}(\mathbf{q}, i\omega_l) = -\frac{1}{N} \sum_{\mathbf{k}k'} h_{m\mathbf{k}\sigma\sigma'}(\mathbf{k},q) h_{n\mathbf{k}'\sigma\sigma'}^n(\mathbf{k}',-\mathbf{q})
\times \int_0^\beta d\tau e^{i\omega_l\tau} \langle \hat{c}_{\mathbf{k}\sigma}^\dagger(\tau) \hat{c}_{\mathbf{k}+\mathbf{q}\sigma'}(\tau) \rangle \times \hat{c}_{\mathbf{k}'\sigma'}^\dagger(0) \hat{c}_{\mathbf{k}'-\mathbf{q}'\sigma'}(0).
$$

Previously, we investigated charge instabilities in the same three-band model with non-local interactions using a generalized random phase approximation (gRPA)\cite{33,39} that includes both ladder and bubble diagrams. Following Ref. 33, we project the interactions onto a set of 19 basis functions in orbital and momentum space, leading to a 19 x 19 matrix equation for the effective interaction vertex $\tilde{\Gamma}^{ij}(q)$, where $q \equiv (\mathbf{q},\omega)$, and $i, j$ label the basis functions. The basis functions and the interaction vertex are the same as in Ref. 33. Closing $\tilde{\Gamma}^{ij}(q)$ on the left and right with current vertex functions $A_{i\alpha\alpha'}^{mn}(q)$ yields the susceptibility

$$
\chi_{\alpha\alpha'\beta\beta'}(q) = \chi_{\alpha\alpha'\beta\beta'}^{0,mn}(q) - \sum_{ij} \tilde{A}_{i\alpha\alpha'}^{1,lm}(q) \tilde{\Gamma}^{ij}(q) \tilde{A}_{j\beta\beta'}^{Rm}(q).
$$

The bare current susceptibility $\chi_{\alpha\alpha'\beta\beta'}^{0,mn}(q)$ and the functions $A_{i\alpha\alpha'}^{mn}(q)$ differ from Ref. 33 as they contain current operators; they are given in the Supplementary Materials.

We search for the existence of spontaneous currents by following the evolution of the current susceptibility upon cooling. The instability is signalled by a divergence of the momentum-resolved susceptibility at zero frequency.
χ_{\pi\pi}(q,\omega = 0)$. This method is thereby unbiased with respect to the ordering wavevector, and the pattern of the emerging orbital currents.

Figure 3 shows typical results for the current susceptibility. The inset shows that the matrix element $\chi_{\pi\pi}(q,\omega = 0)$, corresponding to currents along the $d_{p_\pi}$ bonds, becomes strongly peaked at $q = Q \equiv (\pi, \pi)$ as the temperature is lowered. This peak indeed diverges upon cooling to the critical temperature near $T = 0.01$ (main panel), which signals an instability towards a current-carrying state. Remarkably, this appears to be the first time that a LC instability has been obtained in a three band calculation that does not rely on mean-field theory. The ordering wavevector $Q$ of the $\pi$LC phase is the same as in the DDW scenario, and should be contrasted with the $\theta_{11}$ phase, for which $q = 0$.

The example result in Fig. 3 was obtained for $V_{pd} = 2.2$ and $V_{pp} = 1$. However, the instability towards a $\pi$LC phase persists when $V_{pp} = 0$, which establishes that it is driven by the Coulomb repulsion $V_{pd}$ between copper and oxygen orbitals. As previously established, an ICO with a predominant charge redistribution between $Op_x$ and $Op_y$ orbitals can be generated by the Coulomb repulsion $V_{pp}$ between $Op_x$ and $Op_y$ orbitals. For the parameter values we chose here, the ICO instability is subleading to the $\pi$LC instability. This observation holds true for a wide doping range below the van Hove filling.

To determine the bond-resolved $\pi$LC pattern, we calculate the eigenvector of the leading eigenvalue of the current susceptibility matrix. In the current operator basis $\hat{J}_1(q), .., \hat{J}_8(q)$, this normalized eigenvector is $[0.48, -0.48, 0.15, 0.48, -0.48, 0.15, 0.15, 0.15]$; this eigenvector reveals the direction and the relative magnitudes of the currents on the eight inequivalent bonds: all bonds are involved in the $\pi$LC instability, and for the selected parameter set the currents along the $p-d$ bonds are about three times stronger than those along the $p-p$ bonds. The relative strength of the $p-d$ and $p-p$ currents varies with the ratio $t_{pd}/t_{dd}$. The wavevector $Q$ of the instability further implies that the pattern alternates between adjacent unit cells. We thus obtain the cartoon shown in Fig. 1 in which two distinct (green and black) and interpenetrating loop currents are evident.

The instability towards spontaneous $\pi$LCs will naturally reconstruct the Fermi surface. To explore this we implement the $\pi$LCs on the mean-field level. The starting point for this calculation is the general identity $\hat{n}_a \hat{n}_b = \langle \hat{J}_{ab}^+ \hat{J}_{ab}^+ \rangle / 2 = \langle \hat{J}_{ab} \hat{J}_{ab} \rangle / 2$ that is true for any pair of orbitals $a$ and $b$. Hence, the non-local density-density interactions $\frac{1}{2} \sum_{ia,j\beta} \langle \hat{J}_{ia,j\beta} \hat{n}_{ia} \hat{n}_{j\beta} \rangle$ can be decoupled by introducing the current amplitudes $z_{ia,j\beta} = \langle \hat{J}_{ia,j\beta} \rangle$. The mean-field version of the interorbital Coulomb interactions thus reads:

$$\hat{H}_{MF}' = \hat{H}_{MF} + \hat{H}_{MF}' = \hat{H}_{MF} + \hat{H}_{MF}' = \hat{H}_{MF}$$

where $\hat{H}_{MF} = \sum_k \overline{\Psi}_k^\dagger H_{MF}(k) \overline{\Psi}_k$, $\overline{\Psi}_k = (\Psi_k, \Psi_{k+\overline{Q}})^T$, and

$$H_{MF}(k) = \begin{pmatrix} H_0(k) & H_1(k, Q) \\ H_1^*(k, Q) & H_0(k + Q) \end{pmatrix}$$

$$H_1(k, Q) = \begin{pmatrix} 0 & -R_{pd} s_x' \rangle \\ -i R_{pd} c_x' & 0 \end{pmatrix}$$

where $R_{pd} = 2 z_{pd} V_{pd} / t_{pd}$, $R_{pp} = 4 z_{pp} V_{pp} / t_{pp}$, $s_x' = \sin((k_x + Q_x)/2)$, $c_x' = \cos((k_x + Q_x)/2)$, and $s_y', c_y'$ are defined accordingly, and the orbital energies in $H_0(k)$ are shifted by $\epsilon_d$ and $\epsilon_p$.

For $z_{pd} = 0.02$ and $z_{pp} = z_{pd}/3$, we obtain the reconstructed Fermi surface and the density of states shown in Fig. 3. The normal Fermi surface in the absence of loop currents is also indicated in red. Initially, for very small values of $z_{pd}$ and $z_{pp}$ hole pockets as well as small electron pockets near $(\pi, 0)$ and the equivalent points in the Brillouin zone are formed. With increasing $z_{pd}$, the electron pockets rapidly disappear, and only the hole pockets persist as shown in Fig. 4. $z_{pd}$ used in Fig. 4 corresponds to $\sim 2 \mu_B$, and the associated per plaquette magnetic moment of the $\pi$LC pattern is estimated to be $\sim 0.03 \mu_B$.
The umklapp scattering from the πLC order furthermore leads to a partial gapping of the Fermi surface from which a pseudogap structure in the density of states originates. The latter is calculated from the momentum integrated spectral function \( A(k, \omega) = -\pi^{-1} \sum_{i=1}^{3} \text{Im} G_{ii}(k, \omega) \) with the Green function matrix \( G(k, \omega) = [(\omega + i\delta + \mu)1 - H_{MF}(k)]^{-1} \) where \( \delta \) is a small broadening parameter, \( 1 \) is the identity matrix, \( \mu \) is the chemical potential, and the superscript “−1” is the matrix inverse, and shown in the right panel of Fig. 4.

We propose that the pseudogap appearing at \( T^\ast \) arises from the πLC shown in Fig. 4. The main distinguishing feature of the pseudogap is the depletion of spectral weight along regions of the Fermi surface near \((\pm \pi, 0)\) and \((0, \pm \pi)\). This is obviously true in the πLC phase, and indeed in any model in which the pseudogap has staggered order. The shape of the Fermi surface pockets depends on the strength of the loop currents, the hole filling, and the curvature of the unfolded Fermi surface.

Within the phase with staggered loop currents subsequent phase transitions are likely to occur. Notably, we showed previously that persistent discrepancies between theory and experiment regarding the ordering wavevector \( q^\ast \) of the charge ordered phase are resolved, if the charge order emerges from a preexisting pseudogap phase, rather than causing it. In Ref. [34], a spin-density wave (SDW) with ordering wavevector \( Q = (\pi, \pi) \) was invoked ad hoc as a proxy for the pseudogap in underdoped cuprates. While the presence of a static SDW is not supported by experiment, we view the πLC phase instead as a viable alternative phase out of which a charge-density wave will form with a \( q^\ast \) that connects adjacent hole pockets. In the coexistence with charge order, the Fermi surface of the πLC phase will further reconstruct. The Fermi surface in the coexistence phase should then serve as the basis to analyze the quantum oscillation experiments which reported evidence for the existence of hole pockets [41] [42].

The πLC phase shares neutron-scattering signatures with the DDW state, specifically an elastic magnetic peak centered at \( Q = (\pi, \pi) \) and the opening of a spin excitation gap. Soon after the original proposal of the DDW state it was argued [43] that the neutron-scattering data obtained by Mook et al. in underdoped YBCO [44] are consistent with the expected features of a DDW state. Other subsequent neutron scattering measurements on oxygen ordered ortho-II YBCO instead [45] reported no evidence for the predicted characteristics of an ordered DDW state. The conflicting results of these experiments have remained unresolved.

An obvious signature of the πLC phase is that it breaks time-reversal symmetry. It does not, however, generate a polar Kerr effect because the pattern in Fig. 1 preserves mirror symmetries. Given this result, an explanation for the observed nonzero Kerr angle at \( T_{Kerr} \) requires the onset of a further transition that eliminates these symmetries. [13] This could naturally occur, for example, with the appearance of incommensurate charge order at \( T_{c0} \). The experimental doping dependences of \( T_{Kerr} \) and \( T_{c0} \) [46] [47] are, however, different; a possible connection between the two is therefore not obvious.

It is nevertheless possible that the different symmetries associated with the charge ordering transition are broken at distinct temperatures. [23] Charge order involves both a continuous broken symmetry associated with the spatial lock-in of the charge modulation, and a discrete broken symmetry associated with the orientation of \( q^\ast \). [18] In this scenario, \( T_{Kerr} \) may signal the onset of a charge-nematic phase, in which only the discrete rotational symmetry is broken, while \( T_{c0} \) may mark the pinning of the charge modulation by disorder. [43]

We are led to trace the origin of the intriguingly complex physics of underdoped cuprates to the distinct phenomena which emerge from the Coulomb interactions in the Cu-d orbitals: the local Coulomb repulsion on the Cu-d orbitals is the source of antiferromagnetism in the undoped compounds and of spin-fluctuation mediated d-wave superconductivity upon doping. From the results of this work we conclude that the non-local interaction \( V_{pd} \) can cause an orbital current instability, while the non-local interaction \( V_{pp} \) is responsible for the charge redistribution between Op_x and Op_y orbitals and incommensurate charge order. \( V_{pp} \) and \( V_{pd} \) weaken the spin-fluctuation mediated pairing interaction, which suggests a possible reason why \( T_c < T_{c0} \) below optimal doping. The physics of underdoped cuprates therefore appears to reflect the mutual competition and/or coexistence of these ordering tendencies.

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The calculations were performed on the Linux Cluster of the LRZ in Garching.

[1] A. Shekhter et al., Nature 498, 75 (2013).
[2] B. Fauqué et al., Phys. Rev. Lett. 96, 197001 (2006).
[3] Y. Li et al., Nature 455, 372 (2008).
[4] Y. Sidis and P. Bourges, J. Phys.: Conf. Ser. 449, 012012 (2013).
[5] C. M. Varma, Phys. Rev. B 73, 155113 (2006).
[6] C. M. Varma, J. Phys. Cond. Mat. 26, 505701 (2014).
[7] C. Weber et al., Phys. Rev. Lett. 102, 017005 (2009).
[8] C. Weber, T. Giamarchi, and C. M. Varma, Phys. Rev. Lett. 112, 117001 (2014).
[9] R. Thomale and M. Greiter, Phys. Rev. B 77, 094511 (2008).
[10] Y. F. Kung et al., Phys. Rev. B 90, 224507 (2014).
[11] J. Xia et al., Phys. Rev. Lett. 100, 127002 (2008).
[12] R.-H. He et al., Science 331, 1579 (2011).
[13] Y. Wang, A. Chubukov, and R. Nandkishore, Phys. Rev. B 90, 205130 (2014).
[14] T. Wu et al., Nature 477, 191 (2011).
[15] G. Ghiringhelli et al., Science 337, 821 (2012).
[16] J. Chang et al., Nature Phys. 8, 871 (2012).
[17] E. H. da Silva Neto et al., Science 343, 393 (2014).
[18] R. Comin et al., Science 343, 390 (2014).
[19] Y. Kohsaka et al., Science 337, 821 (2012).
[20] N. Doiron-Leyraud et al., Nature 447, 565 (2007).
[21] S. E. Sebastian, N. Harrison, and G. Lonzarich, Rep. Prog. Phys. 75, 102501 (2012).
SUPPLEMENTARY MATERIAL

Non-interacting Hamiltonian

The non-interacting part of the three band model is given by

\[ \hat{H}_0 = \sum_{i\alpha\sigma} \epsilon_{i\alpha} \hat{n}_{i\alpha\sigma} + \sum_{i\omega\alpha\beta} t_{i\alpha\beta} \hat{c}_{i\alpha\sigma}^{\dagger} \hat{c}_{j\beta\sigma} \]  

(10)

where \( i \) and \( j \) are unit cell labels, \( \alpha \) and \( \beta \) are orbital labels, \( \sigma \) is the spin label, \( \epsilon_{i\alpha} \) is the orbital energy, \( \hat{n}_{i\alpha\sigma} \) is the number operator, \( t_{i\alpha\beta} \) is the tunneling matrix element between orbital \( i\alpha \) and \( j\beta \), and \( \hat{c}_{i\alpha\sigma} \) and \( \hat{c}_{i\alpha\sigma}^{\dagger} \) are annihilation and creation operators. Below, we suppress the spin labels.

Using the translational invariance, \( \hat{H}_0 \) can be Fourier transformed to reciprocal space:

\[ \hat{H}_0 = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^{\dagger} \hat{H}_0(\mathbf{k}) \Psi_{\mathbf{k}} \]  

(11)

where \( \Psi_{\mathbf{k}}^{\dagger} = [\hat{c}_{\mathbf{k}d}, \hat{c}_{\mathbf{k}x}, \hat{c}_{\mathbf{k}y}] \), and \( \hat{c}_{\mathbf{k}a} \) (\( \hat{c}_{\mathbf{k}a}^{\dagger} \)) is the creation (annihilation) operator for an electron with crystal momentum \( \mathbf{k} \) and orbital \( \alpha \). Explicitly,

\[ \hat{c}_{\mathbf{k}a} = \frac{1}{\sqrt{N}} \sum_{i} e^{-i \mathbf{k} \cdot \mathbf{R}_{\alpha}\mathbf{c}} \hat{c}_{i\alpha} \]  

(12)

\[ \hat{c}_{\mathbf{k}a}^{\dagger} = \frac{1}{\sqrt{N}} \sum_{i} e^{i \mathbf{k} \cdot \mathbf{R}_{\alpha}\mathbf{c}} \hat{c}_{i\alpha}^{\dagger} \]  

(13)

where \( N \) is the number of unit cells in the system, and \( \mathbf{R}_{\alpha}\mathbf{c} \) is the position vector of \( \alpha \)th orbital in \( i \)th unit cell. \( \hat{H}_0(\mathbf{k}) \) is readily obtained by plugging Eqs. (12) and (13) into Eq. (10):

\[ \hat{H}_0(\mathbf{k}) = \begin{pmatrix} \epsilon_{d} & -2t_{pd}s_{x} & 2t_{pd}s_{y} \\ 2t_{pd}s_{x} & \epsilon_{x} & 4t_{pp}s_{x}s_{y} \\ -2t_{pd} & 4t_{pp}s_{x}s_{y} & \epsilon_{y} \end{pmatrix} \]  

(14)

where \( s_{x} = \sin(k_{x}/2) \) and \( s_{y} = \sin(k_{y}/2) \). A more convenient form of \( \hat{H}_0 \) is obtained after the following gauge transformation:

\[ \hat{c}_{\mathbf{k}x} \rightarrow i \hat{c}_{\mathbf{k}x} \]  

(15)

\[ \hat{c}_{\mathbf{k}x}^{\dagger} \rightarrow -i \hat{c}_{\mathbf{k}x}^{\dagger} \]  

(16)

\[ \hat{c}_{\mathbf{k}y} \rightarrow i \hat{c}_{\mathbf{k}y} \]  

(17)

\[ \hat{c}_{\mathbf{k}y}^{\dagger} \rightarrow -i \hat{c}_{\mathbf{k}y}^{\dagger} \]  

(18)

Furthermore, in the matrix elements \( H_{0,23} \) and \( H_{0,32} \), we introduce a minus sign to obtain a realistic Fermi surface. Such a sign change originates from indirect hopping processes via the Cu4s orbital.[49] Hence, the final form of \( \hat{H}_0 \) is obtained:

\[ \hat{H}_0(\mathbf{k}) = \begin{pmatrix} \epsilon_{d} & 2t_{pd}s_{x} & -2t_{pd}s_{y} \\ 2t_{pd}s_{x} & \epsilon_{p} & 4t_{pp}s_{x}s_{y} \\ -2t_{pd}s_{y} & -4t_{pp}s_{x}s_{y} & \epsilon_{p} \end{pmatrix} \]  

(19)

Current operators

The current operator is conventionally defined as

\[ \hat{J}_{ij} = -it_{ij}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} - \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(20)

At this point, however, it is important to take into account that we are working with a Hamiltonian which is gauge transformed according to Equations (15).[1] In the transformed operator basis, the current operators in real space take the following form:

\[ J_{id,jx} = t_{id,jx}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(21)

\[ J_{ix,jd} = -t_{ix,j\sigma}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(22)

\[ J_{id,jy} = t_{id,jy}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(23)

\[ J_{iy,jd} = -t_{iy,j\sigma}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} + \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(24)

\[ J_{ix,jy} = -t_{ix,jy}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} - \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(25)

\[ J_{iy,jy} = -t_{iy,jy}(\hat{c}_{i\sigma}^{\dagger} \hat{c}_{j\sigma} - \hat{c}_{j\sigma}^{\dagger} \hat{c}_{i\sigma}) \]  

(26)

Interacting current susceptibility

The following quantities entering into the calculation of Eq. (16) are explicitly defined as

\[ S_{\alpha\gamma}^{\alpha\omega}(\mathbf{k}, \mathbf{q}) = \frac{1}{N} \sum_{\mathbf{k}_{\mu\nu}} h_{\alpha\omega}(\mathbf{k}, \mathbf{q}) \mu_{\nu \ell}^{\alpha} \nu_{\omega \ell}^{\alpha} F_{\mathbf{k}_{\mu\nu}}(E) h_{\gamma}^{\omega}(\mathbf{k} - \mathbf{q}, \mathbf{q}) \]  

(27)

\[ \epsilon_{\omega\omega}^{\alpha \beta}(\mathbf{k}) = \frac{1}{N} \sum_{\mathbf{k}_{\mu\nu}} h_{\alpha\omega}(\mathbf{k}, \mathbf{q}) \mu_{\nu \ell}^{\alpha} \nu_{\omega \ell}^{\alpha} F_{\mathbf{k}_{\mu\nu}}(E) g_{\gamma}^{\omega}(\mathbf{k}) \]  

(28)

\[ \epsilon_{\omega\omega}^{\alpha \beta}(\mathbf{k}) = \frac{1}{N} \sum_{\mathbf{k}_{\mu\nu}} h_{\alpha\omega}(\mathbf{k}, \mathbf{q}) \mu_{\nu \ell}^{\alpha} \nu_{\omega \ell}^{\alpha} F_{\mathbf{k}_{\mu\nu}}(E) g_{\gamma}^{\omega}(\mathbf{k}) \]  

(29)

\[ \ell \) are bond indexes, \( h_{\alpha\omega}^{\alpha \beta}(\mathbf{k}, \mathbf{q}) \) are matrix elements of the current operators that are explicitly defined in Table 1. \( S_{\alpha\omega}(\mathbf{k}) \) is the structure element of \( \omega \)th eigenvector of \( \hat{H}_0(\mathbf{k}) \); the superscript \( \omega \) is the complex conjugate, \( E_{\alpha}^{\mu} \) is the eigenvalue, and \( g_{\gamma}^{\omega}(\mathbf{k}) \) are 19-dimensional function basis (\( \ell \in [1, 19] \)) in orbital and momentum space which are explicitly defined in Ref. [33].
| $\ell$ | $\theta^\ell$ | $h^\ell_{\theta^\ell}(k, q)$ |
|------|--------------|------------------|
| 1 dx | $t p d e^{i (q_x + k_x)/2}$ | |
| 1 xd | $- t p d e^{- i k x/2}$ | |
| 2 dy | $- t p d e^{i (q_y + k_y)/2}$ | |
| 2 yd | $t p d e^{- i k y/2}$ | |
| 3 xy | $- i t_{pp} e^{i (q_y - k_x + k_y)/2}$ | |
| 3 yx | $- i t_{pp} e^{i (q_x - k_x - k_y)/2}$ | |
| 4 dx | $- t p d e^{- i (q_x + k_x)/2}$ | |
| 4 xd | $t p d e^{i k x/2}$ | |
| 5 dy | $t p d e^{- i (q_y + k_y)/2}$ | |
| 5 yd | $- t p d e^{i k y/2}$ | |
| 6 xy | $- i t_{pp} e^{- i (q_y + k_x - k_y)/2}$ | |
| 6 yx | $- i t_{pp} e^{- i (q_x + k_x - k_y)/2}$ | |
| 7 xy | $i t_{pp} e^{i (q_y + k_x + k_y)/2}$ | |
| 7 yx | $i t_{pp} e^{- i (q_x + k_x + k_y)/2}$ | |
| 8 xy | $i t_{pp} e^{- i (q_y + k_x + k_y)/2}$ | |
| 8 yx | $i t_{pp} e^{i (q_x + k_x + k_y)/2}$ | |

**TABLE I**: Matrix elements of the current operator. $i$ is the imaginary constant. The overall sign of each term results from three factors: the complex constant in the current operator definitions, the sign of the hopping terms, and the gauge transformation.