

**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  $\text{CuO}_2$  planes in cuprates. Contrary to the previously proposed  $\theta_{II}$  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.

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**I. INTRODUCTION**

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  $T_{\text{co}} < T^*$  and superconductivity at  $T_c < T_{\text{co}}$ . In addition, broken time-reversal symmetry has been associated with  $T^*$  and a Kerr rotation is measured below a temperature  $T_{\text{Kerr}}$  with  $T_{\text{Kerr}} \sim 0.75T^*$  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 [1]; this finding challenges the viewpoint that the pseudogap arises as a correlation induced phenomenon in a symmetry unbroken paramagnetic phase. Below  $T^*$  spin-polarized neutron scattering experiments detected weak magnetic moments [2–5]. These moments appear to preserve the translational symmetry of the lattice, and led to the proposal of intra-unit cell loop currents (LCs) [6]. However, the so-called  $\theta_{II}$  LC phase by itself has difficulty explaining the partial gapping of charge excitations [7]. While variational methods favored the existence of LC phases in finite clusters [8,9], alternative numerically exact analyses reported no evidence for the  $\theta_{II}$  phase [10,11].

The Kerr effect [12,13] that sets in below  $T_{\text{Kerr}}$  is further evidence for time-reversal symmetry breaking, but also requires that mirror symmetries be broken [14]. Throughout much of the cuprate phase diagram,  $T_{\text{Kerr}}$  and  $T_{\text{co}}$  [15–23] are close, which has motivated further proposals in which fluctuating charge- [14,24] or pair-density wave states [25,26] 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 [27–30].

The ICO phase involves predominantly a charge transfer between oxygen orbitals in the  $\text{CuO}_2$  planes [20–22,31,32].

This challenges notions of immutable  $\text{CuO}_2$  bands, and points to the necessity to employ multiorbital models for the ICO phase [33–35]. Here, we support this reasoning and show that orbital resolved intra-unit cell physics is important throughout the pseudogap regime.

In this work we report the results of an unbiased calculation for a three-band model of  $\text{CuO}_2$  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  $\theta_{II}$  phase, but shares its ordering wave vector  $\mathbf{Q} = (\pi, \pi)$  with the earlier phenomenological proposal of LCs in the  $d$ -density wave (DDW) state [36,37]. 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 [34] is subleading to the  $\pi$ LC instability. Yet, the presence of the staggered order favors a subsequent instability towards ICO with axial wave vectors connecting the tips of the hole pockets [35], consistent with experiments. The charge modulation of the latter necessarily breaks mirror symmetries and will hence allow for a polar Kerr signal [14]. This scenario is offered as a proposal for the cascade of phase transitions in the pseudogap regime of underdoped cuprates.

**II. HAMILTONIAN**

The unit cell of a single  $\text{CuO}_2$  plane is shown in Fig. 2, along with the choice of orbital phases and the corresponding signs of the hopping terms.

The noninteracting part of the three band model is given by

$$\hat{H}_0 = \sum_{i\alpha\sigma} \epsilon_{i\alpha} \hat{n}_{i\alpha\sigma} + \sum_{i\alpha j\beta\sigma} t_{i\alpha j\beta} \hat{c}_{i\alpha\sigma}^\dagger \hat{c}_{j\beta\sigma}, \quad (1)$$

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,  $t_{i\alpha j\beta}$  is the tunneling matrix element between orbital

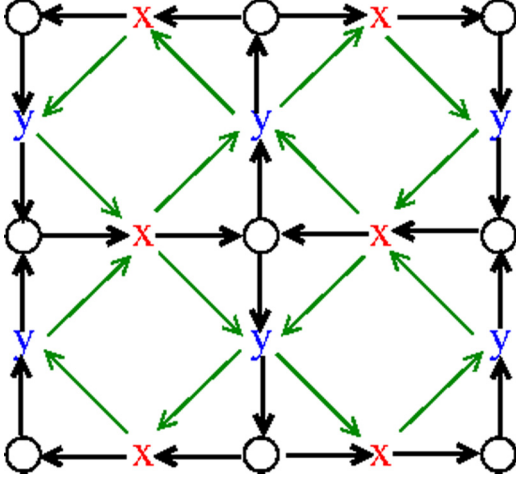


FIG. 1. (Color online) Staggered pattern of spontaneous loop currents. Open circle, “x” and “y” symbols denote  $\text{Cu}d_{x^2-y^2}$ ,  $\text{O}p_x$ , and  $\text{O}p_y$  orbitals, respectively. Currents along  $p-d$  bonds (black arrows) are about three times stronger than those in  $p-p$  bonds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

$i\alpha$  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:

$$\hat{H}_0 = \sum_{\mathbf{k}} \Psi_{\mathbf{k}}^\dagger \mathbf{H}_0(\mathbf{k}) \Psi_{\mathbf{k}}, \quad (2)$$

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

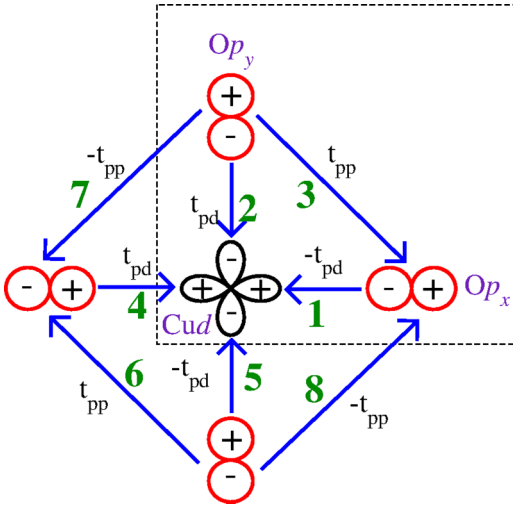


FIG. 2. (Color online) Unit cell of a  $\text{CuO}_2$  plane (dashed box). The orbital phase convention is indicated by the sign of the hopping matrix elements. Numbers in green enumerate the inequivalent bonds, and the direction of the blue arrows indicates a positive sign of the current flow for the current operator definitions given in Appendix A.

$\mathbf{k}$  and orbital  $\alpha$ . Explicitly,

$$\hat{c}_{\mathbf{k}\alpha} = \frac{1}{\sqrt{N}} \sum_i e^{-i\mathbf{k}\cdot\mathbf{R}_{i\alpha}} \hat{c}_{i\alpha}, \quad (3)$$

$$\hat{c}_{\mathbf{k}\alpha}^\dagger = \frac{1}{\sqrt{N}} \sum_i e^{i\mathbf{k}\cdot\mathbf{R}_{i\alpha}} \hat{c}_{i\alpha}^\dagger, \quad (4)$$

where  $N$  is the number of unit cells in the system, and  $\mathbf{R}_{i\alpha}$  is the position vector of the  $\alpha$ 'th orbital in  $i$ 'th unit cell.  $H_0(\mathbf{k})$  is readily obtained by plugging Eqs. (3) and (4) into Eq. (1):

$$\mathbf{H}_0(\mathbf{k}) = \begin{pmatrix} \epsilon_d & -2it_{pd}s_x & 2it_{pd}s_y \\ 2it_{pd}s_x & \epsilon_x & 4t_{pp}s_x s_y \\ -2it_{pd} & 4t_{pp}s_x s_y & \epsilon_y \end{pmatrix}, \quad (5)$$

where  $s_x = \sin(k_x/2)$  and  $s_y = \sin(k_y/2)$ . A more convenient form of  $\mathbf{H}_0$  is obtained after the following gauge transformation:

$$\hat{c}_{\mathbf{k}x} \rightarrow i\hat{c}_{\mathbf{k}x}, \quad (6)$$

$$\hat{c}_{\mathbf{k}x}^\dagger \rightarrow -i\hat{c}_{\mathbf{k}x}^\dagger, \quad (7)$$

$$\hat{c}_{\mathbf{k}y} \rightarrow i\hat{c}_{\mathbf{k}y}, \quad (8)$$

$$\hat{c}_{\mathbf{k}y}^\dagger \rightarrow -i\hat{c}_{\mathbf{k}y}^\dagger. \quad (9)$$

Hence, the final form of  $\mathbf{H}_0$  is obtained:

$$\mathbf{H}_0(\mathbf{k}) = \begin{pmatrix} \epsilon_d & 2t_{pd}s_x & -2t_{pd}s_y \\ 2t_{pd}s_x & \epsilon_p & 4t_{pp}s_x s_y \\ -2t_{pd}s_y & 4t_{pp}s_x s_y & \epsilon_p \end{pmatrix}. \quad (10)$$

We set  $t_{pd} = 1$  so that it defines the unit of energy.

The interacting part of the Hamiltonian includes the intra- ( $U_\alpha$ ) and interorbital ( $V_{i\alpha,j\beta}$ ) Coulomb interactions,

$$\hat{H}' = \sum_{i\alpha\sigma,j\beta\sigma'} \left[ \delta_{i\alpha,j\beta}(1 - \delta_{\sigma,\sigma'})U_\alpha + \frac{V_{i\alpha,j\beta}}{2} \right] \hat{n}_{i\alpha\sigma} \hat{n}_{j\beta\sigma'}, \quad (11)$$

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 [38] with a typical parameter set [39].

### III. INTERACTING CURRENT SUSCEPTIBILITY

The current operator associated with the bond between sites  $i\alpha$  and  $j\beta$  is  $\hat{J}_{i\alpha,j\beta} = -it_{i\alpha,j\beta}(\hat{c}_{i\alpha}^\dagger c_{j\beta} - \hat{c}_{j\beta}^\dagger c_{i\alpha})$  where  $t_{i\alpha,j\beta}$  is the corresponding hopping matrix element. If  $\langle \hat{J}_{i\alpha,j\beta} \rangle > 0$  then current flows from  $j\beta$  to  $i\alpha$ . In momentum space, the current operator along bond  $m$  is given by

$$\hat{J}_m(\mathbf{q}) = -i \sum_{\mathbf{k}} [h_{\alpha\beta}^m(\mathbf{k}, \mathbf{q}) \hat{c}_{\mathbf{k}\alpha}^\dagger \hat{c}_{\mathbf{k}+\mathbf{q}\beta} - h_{\beta\alpha}^m(\mathbf{k}, \mathbf{q}) \hat{c}_{\mathbf{k}\beta}^\dagger \hat{c}_{\mathbf{k}+\mathbf{q}\alpha}], \quad (12)$$

where  $\alpha, \beta$  are the orbitals associated with the bond and the matrix elements of the current operators  $h_{\alpha\beta}^m(\mathbf{k}, \mathbf{q})$  are listed in Table I. As shown in Fig. 2, there are eight distinct bonds on

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.

$\ell$	$\theta\theta'$	$h_{\theta\theta'}^\ell(\mathbf{k}, \mathbf{q})$
1	dx	$t_{pd}e^{i(q_x+k_x)/2}$
1	xd	$-t_{pd}e^{-ik_x/2}$
2	dy	$-t_{pd}e^{i(q_y+k_y)/2}$
2	yd	$t_{pd}e^{-ik_y/2}$
3	xy	$-it_{pp}e^{i(q_y-k_x+k_y)/2}$
3	yx	$-it_{pp}e^{i(q_x+k_x-k_y)/2}$
4	dx	$-t_{pd}e^{-i(q_x+k_x)/2}$
4	xd	$t_{pd}e^{ik_x/2}$
5	dy	$t_{pd}e^{-i(q_y+k_y)/2}$
5	yd	$-t_{pd}e^{ik_y/2}$
6	xy	$-it_{pp}e^{-i(q_y+k_x-k_y)/2}$
6	yx	$-it_{pp}e^{-i(q_x+k_x-k_y)/2}$
7	xy	$it_{pp}e^{i(q_y+k_x+k_y)/2}$
7	yx	$it_{pp}e^{-i(q_x+k_x+k_y)/2}$
8	xy	$it_{pp}e^{-i(q_y+k_x+k_y)/2}$
8	yx	$it_{pp}e^{i(q_x+k_x+k_y)/2}$

the CuO<sub>2</sub> lattice. Accordingly the current susceptibility,

$$\chi_{mn}^J(\mathbf{q}, i\omega_\ell) = \int_0^\beta d\tau e^{i\omega_\ell\tau} \langle \hat{J}_m(\mathbf{q}, \tau) \hat{J}_n(-\mathbf{q}, 0) \rangle, \quad (13)$$

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

$$\chi_{mn}^J = \chi_{\alpha\beta\alpha'\beta'}^{mn} - \chi_{\alpha\beta\beta'\alpha'}^{mn} - \chi_{\beta\alpha\alpha'\beta'}^{mn} + \chi_{\beta\alpha\beta'\alpha'}^{mn}, \quad (14)$$

where

$$\begin{aligned} \chi_{\theta\theta'\gamma\gamma'}^{mn}(\mathbf{q}, i\omega_\ell) &= \frac{-1}{N} \sum_{\mathbf{k}\mathbf{k}'} h_{\theta\theta'}^m(\mathbf{k}, \mathbf{q}) h_{\gamma\gamma'}^n(\mathbf{k}', -\mathbf{q}) \\ &\times \int_0^\beta d\tau e^{i\omega_\ell\tau} \langle \hat{c}_{\mathbf{k}\theta}^\dagger(\tau) c_{\mathbf{k}+\mathbf{q}\theta'}(\tau) \\ &\times \hat{c}_{\mathbf{k}'\gamma'}^\dagger(0) c_{\mathbf{k}'-\mathbf{q}\gamma'}(0) \rangle. \end{aligned} \quad (15)$$

Previously, we investigated charge instabilities in the same three-band model with nonlocal interactions using a generalized random phase approximation (gRPA) [34,40]. While methods like QMC or cluster DMFT are at first glance more desirable as they are designed to handle strong local correlations, they are less accurate in treating nonlocal interactions, and are also limited in momentum-space resolution. Although it neglects strong correlation physics, the gRPA has the advantage that it treats local and nonlocal interactions on the same footing, and is unbiased with respect to wave vector and to the unit cell-resolved current pattern. Within gRPA, the two-particle vertex function includes both exchange and direct interaction diagrams and hence also generates combinations of both (see Fig. 3), while Green functions remain unrenormalized.

Following Ref. [34], we project the interactions onto a set of 19 basis functions  $g_{\alpha\beta}^i(\mathbf{k})$  in orbital and momentum space, leading to a  $19 \times 19$  matrix equation for the effective

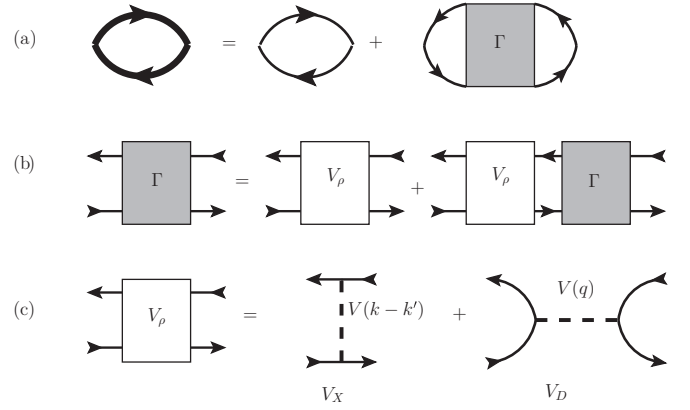


FIG. 3. Diagrammatic structure of gRPA. (a) Interacting susceptibility, (b) vertex function, (c) effective interaction. Reprinted with permission from [34]. ©American Physical Society.

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. [34]. Closing  $\tilde{\Gamma}^{ij}(q)$  on the left and right with current vertex functions  $A_{\alpha\alpha'}^{i,\eta m}(q)$  yields the susceptibility,

$$\chi_{\alpha\alpha'\beta\beta'}^{mn}(q) = \chi_{\alpha\alpha'\beta\beta'}^{0,mn}(q) - \sum_{ij} A_{\alpha\alpha'}^{i,Lm}(q) \tilde{\Gamma}^{ij}(q) A_{\beta\beta'}^{j,Rn}(q), \quad (16)$$

where

$$\chi_{\alpha\alpha'\beta\beta'}^{0,\ell_1\ell_2}(q) = \frac{1}{N} \sum_{\mathbf{k}\mu\nu} h_{\alpha\alpha'}^{\ell_1}(\mathbf{k}, \mathbf{q}) M_{\mu\nu\mathbf{k}\mathbf{q}}^{\alpha'\beta\beta'\alpha} F_{\mathbf{k}\mathbf{q}}^{\nu\mu}(\omega) h_{\beta\beta'}^{\ell_2}(\mathbf{k} - \mathbf{q}, \mathbf{q}),$$

$$A_{\alpha\alpha'}^{i,L\ell} = \frac{1}{N} \sum_{\mathbf{k}\mu\nu\theta\theta'} h_{\alpha\alpha'}^\ell(\mathbf{k}, \mathbf{q}) M_{\mu\nu\mathbf{k}\mathbf{q}}^{\alpha'\theta\theta'\alpha} F_{\mathbf{k}\mathbf{q}}^{\nu\mu}(\omega) g_{\theta\theta'}^i(\mathbf{k}), \quad (17)$$

$$A_{\alpha\alpha'}^{i,R\ell} = \frac{1}{N} \sum_{\mathbf{k}\mu\nu\theta\theta'} h_{\alpha\alpha'}^\ell(\mathbf{k} - \mathbf{q}, \mathbf{q}) M_{\mu\nu\mathbf{k}\mathbf{q}}^{\alpha'\theta\theta'\alpha} F_{\mathbf{k}\mathbf{q}}^{\nu\mu}(\omega) g_{\theta\theta'}^i(\mathbf{k}),$$

$$M_{\mu\nu\mathbf{k}\mathbf{q}}^{\gamma'\theta\theta'\gamma} = S_{\gamma'v}(\mathbf{k}) S_{\theta v}^*(\mathbf{k}) S_{\theta'\mu}(\mathbf{k} + \mathbf{q}) S_{\gamma\mu}^*(\mathbf{k} + \mathbf{q}), \quad (18)$$

$$F_{\mathbf{k}\mathbf{q}}^{\nu\mu}(\omega) = \frac{f(E_{\mathbf{k}}^\nu) - f(E_{\mathbf{k}+\mathbf{q}}^\mu)}{\omega + E_{\mathbf{k}}^\nu - E_{\mathbf{k}+\mathbf{q}}^\mu + i\delta}, \quad (19)$$

$\ell$  denotes bond indices,  $h_{\alpha\alpha'}^\ell(\mathbf{k}, \mathbf{q})$  are matrix elements of the current operators which are explicitly defined in Table I,  $S_{\alpha v}(\mathbf{k})$  is the  $\alpha$ th element of the  $v$ th eigenvector of  $\mathbf{H}_0(\mathbf{k})$ ,  $S_{\alpha v}^*(\mathbf{k})$  is its complex conjugate,  $E_{\mathbf{k}}^\nu$  are the eigenvalues,  $f(E)$  is the Fermi function, and  $i$  and  $\delta$  in Eq. (19) are the complex constant and a small broadening parameter, respectively. The bare current susceptibility  $\chi_{\alpha\alpha'\beta\beta'}^{0,mn}(q)$  and the functions  $A_{\alpha\alpha'}^{i,L\ell}(q)$  and  $A_{\alpha\alpha'}^{i,R\ell}(q)$  differ from Ref. [34] as they contain current operators.

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  $\chi_{mn}^J(\mathbf{q}, \omega = 0)$ .

#### IV. RESULTS

Figure 4 shows typical results for the current susceptibility. The inset shows that the matrix element  $\chi_{11}^J(\mathbf{q}, \omega = 0)$ ,

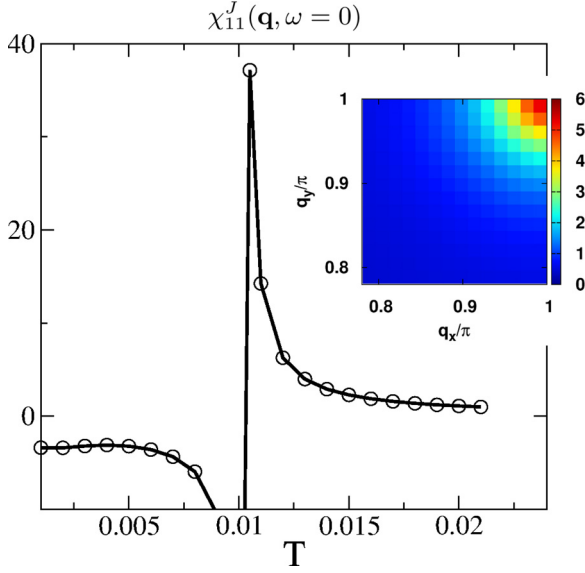


FIG. 4. (Color online) Temperature evolution of  $\chi'_{11}(\mathbf{Q}, \omega = 0)$  for  $\mathbf{Q} = (\pi, \pi)$ . (Inset)  $\chi'_{11}(\mathbf{q}, \omega = 0)$  at  $T = 0.011$ . The parameter values are  $V_{pd} = 2.2$ ,  $V_{pp} = 1$ , and the hole density is  $p = 0.13$ .

corresponding to currents along the  $d - p_x$  bonds, becomes strongly peaked at  $\mathbf{q} = \mathbf{Q} \equiv (\pi, \pi)$  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. The ordering wave vector  $\mathbf{Q}$  of the  $\pi$ LC phase is the same as in the DDW scenario [36], and should be contrasted with the  $\theta_{II}$  phase [6], for which  $\mathbf{q} = \mathbf{0}$ .

The example result in Fig. 4 was obtained for  $V_{pd} = 2.2$  and  $V_{pp} = 1$ . However, the instability towards a  $\pi$ LC phase persists when  $V_{pp} = 0$ , and hence is driven by the Coulomb repulsion  $V_{pd}$  between copper and oxygen orbitals. In fact, also the local interactions  $U_d$  and  $U_p$  have no effect on the  $\pi$ LC instability. As we have explicitly verified, the staggered current instability originates from the exchange (ladder only) diagrams.

As previously established [35], an ICO with a predominant charge redistribution between  $O_{p_x}$  and  $O_{p_y}$  orbitals can be generated by the Coulomb repulsion  $V_{pp}$  between  $O_{p_x}$  and  $O_{p_y}$  orbitals. Based on the interorbital distances, we expect  $V_{pd} > V_{pp}$ , which implies that loop currents emerge at higher temperatures than ICO. Indeed, for our parameter values, the critical temperature for the  $\pi$ LC instability is about twice as large as the critical temperature for ICO.

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  $\hat{J}_1(\mathbf{q}), \dots, \hat{J}_8(\mathbf{q})$ , 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  $t_{pp}/t_{pd}$ . The wave vector  $\mathbf{Q}$  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. This pattern is similar to the previously proposed current-carrying phases of either the DDW [36] or the staggered flux-phase type [41–43], but differs in having two circulating current loops one of which involves oxygen orbitals only.

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  $\hat{n}_a \hat{n}_b = (\hat{J}_{ab} \hat{J}_{ab}^* / t_{ab}^2 + \hat{n}_a + \hat{n}_b) / 2$  that is true for any pair of orbitals  $a$  and  $b$  [44]. Hence, the nonlocal density-density interactions  $\frac{1}{2} \sum_{i\alpha, j\beta} V_{i\alpha, j\beta} \hat{n}_{i\alpha} \hat{n}_{j\beta}$  can be decoupled by introducing the current amplitudes  $z_{i\alpha, j\beta} = \langle \hat{J}_{i\alpha, j\beta} \rangle = \pm z_{\alpha\beta}$ . The mean-field version of the interorbital Coulomb interactions thus reads

$$\hat{H}'_{\text{MF}} = \tilde{\epsilon}_p (\hat{n}_x + \hat{n}_y) + \tilde{\epsilon}_d \hat{n}_d - \sum_{\langle i\alpha, j\beta \rangle} \frac{V_{i\alpha, j\beta}}{2t_{i\alpha, j\beta}^2} \hat{J}_{i\alpha, j\beta} z_{i\alpha, j\beta}, \quad (20)$$

where  $\tilde{\epsilon}_p = V_{pd} + 2V_{pp}$  and  $\tilde{\epsilon}_d = 2V_{pd}$  renormalize the orbital energies. The intraorbital interactions lead to additional Hartree shifts of the orbital energies; these are assumed to be already included in  $\epsilon_d$  and  $\epsilon_p$ . We obtain the mean-field Hamiltonian  $\hat{H}_{\text{MF}} = \hat{H}_0 + \hat{H}'_{\text{MF}}$ , with  $\hat{H}_{\text{MF}} = \sum_{\mathbf{k}} \bar{\Psi}_{\mathbf{k}} \mathbf{H}_{\text{MF}}(\mathbf{k}) \bar{\Psi}_{\mathbf{k}}$ ,  $\bar{\Psi}_{\mathbf{k}} = (\Psi_{\mathbf{k}}, \Psi_{\mathbf{k}+\mathbf{Q}})^T$ , and

$$\mathbf{H}_{\text{MF}}(\mathbf{k}) = \begin{bmatrix} \mathbf{H}_0(\mathbf{k}) & \mathbf{H}_1(\mathbf{k}, \mathbf{Q}) \\ \mathbf{H}_1^\dagger(\mathbf{k}, \mathbf{Q}) & \mathbf{H}_0(\mathbf{k} + \mathbf{Q}) \end{bmatrix}, \quad (21)$$

$$\mathbf{H}_1(\mathbf{k}, \mathbf{Q}) = \begin{bmatrix} 0 & iR_{pd}s'_x & iR_{pd}s'_y \\ R_{pd}c'_x & 0 & +R_{pp}c'_x s'_y \\ R_{pd}c'_y & -R_{pp}s'_x c'_y & 0 \end{bmatrix}, \quad (22)$$

where  $R_{pd} = z_{pd} V_{pd} / t_{pd}$ ,  $R_{pp} = 2z_{pp} V_{pp} / t_{pp}$ ,  $s'_x = \sin[(k_x + Q_x)/2]$ ,  $c'_x = \cos[(k_x + Q_x)/2]$ , and  $s'_y, c'_y$  are defined accordingly, and the orbital energies in  $\mathbf{H}_0(\mathbf{k})$  are shifted by  $\tilde{\epsilon}_d$  and  $\tilde{\epsilon}_p$ .

We show the reconstructed ( $z_{pd} = 0.04$ ,  $z_{pp} = z_{pd}/3$ ) and normal ( $z_{pd} = z_{pp} = 0$ ) Fermi surfaces and densities of states in Fig. 5. For very small  $z_{pd}$  and  $z_{pp}$ , hole pockets, as well

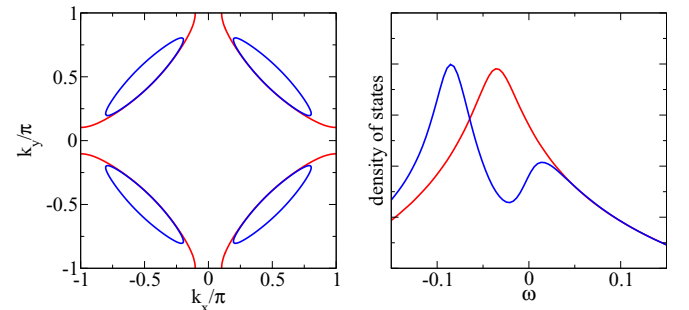


FIG. 5. (Color online) (Left) Normal (red) and reconstructed (blue) Fermi surfaces. The loop current strengths are set to  $z_{pd} = 0.04$  and  $z_{pp} = z_{pd}/3$ . The hole density here is  $p = 0.10$ . (Right) Density of states near the Fermi energy.

as small electron pockets near  $(\pi, 0)$  and symmetry-related points, are formed. The shapes of the pockets depend on the strength of the loop currents, the hole filling, and the curvature of the unfolded Fermi surface. With increasing  $z_{pd}$  and  $z_{pp}$ , the electron pockets rapidly disappear, and only the hole pockets persist (Fig. 5). If one takes  $t_{pd}$  from band structure calculations,  $z_{pd} = 0.04$  corresponds to a  $p-d$  current of  $16 \mu\text{A}$ ; empirical bandwidths are 3 times smaller [45], giving a current of  $5 \mu\text{A}$  and a plaquette magnetic moment in the range of  $0.05\mu_B$  to  $0.09\mu_B$  (see Appendix B).

We propose that the pseudogap appearing at  $T^*$  arises from the  $\pi\text{LC}$  shown in Fig. 1. Physical properties of staggered currents have been discussed previously [36,41,46,47], and we focus here on aspects related to the recent discoveries of charge order and time-reversal symmetry breaking. 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 leads to a pseudogap in the density of states, as shown in Fig. 5 for the  $\pi\text{LC}$  phase. In our calculations  $z_{pd}$  is about one-third of the peak-to-peak pseudogap; experimental pseudogaps of  $\sim 100 \text{ meV}$  [48] therefore suggest  $z_{pd} \sim 0.033 \text{ eV}$ , corresponding to a  $p-d$  current of  $\sim 7 \mu\text{A}$ , consistent with the estimate above.

## V. DISCUSSION

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 wave vector  $\mathbf{q}^*$  of the charge ordered phase are resolved, if the charge order emerges from a preexisting pseudogap phase, rather than causing it. In Ref. [35], a spin-density wave (SDW) with ordering wave vector  $\mathbf{Q} = (\pi, \pi)$  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  $\pi\text{LC}$  phase instead as a viable alternative phase out of which a charge-density wave will form with a  $\mathbf{q}^*$  that connects adjacent hole pockets. In the coexistence with charge order, the Fermi surface of the  $\pi\text{LC}$  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 [49,50].

The  $\pi\text{LC}$  phase shares neutron-scattering signatures with the DDW state, specifically an elastic magnetic peak centered at  $\mathbf{Q} = (\pi, \pi)$  and the opening of a spin excitation gap. Soon after the original proposal of the DDW state it was argued [51] that the neutron-scattering data obtained by Mook *et al.* in underdoped YBCO [52] are consistent with the expected features of a DDW state. Other subsequent neutron scattering measurements on oxygen ordered ortho-II YBCO instead [53] 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  $\pi\text{LC}$  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  $T_{\text{Kerr}}$  requires the onset of a

further transition that eliminates these symmetries [14]. This could naturally occur, for example, with the appearance of incommensurate charge order at  $T_{\text{co}}$ . The experimental doping dependencies of  $T_{\text{Kerr}}$  [12] and  $T_{\text{co}}$  [54,55] 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 [24]. 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  $\mathbf{q}^*$  [56]. In this scenario,  $T_{\text{Kerr}}$  may signal the onset of a charge-nematic phase, in which only the discrete rotational symmetry is broken, while  $T_{\text{co}}$  may mark the pinning of the charge modulation by disorder [56].

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  $\text{CuO}_2$  planes: The local Coulomb repulsion on the Cu  $d$  orbital 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 nonlocal interaction  $V_{pd}$  can cause an orbital current instability, while the nonlocal interaction  $V_{pp}$  is responsible for the charge redistribution between  $O_{px}$  and  $O_{py}$  orbitals and incommensurate charge order.  $V_{pp}$  and  $V_{pd}$  weaken the spin-fluctuation mediated pairing interaction, which suggests a possible reason why  $T_c < T_{\text{co}}$  below optimal doping. The physics of underdoped cuprates therefore appears to reflect the mutual competition and/or coexistence of these ordering tendencies.

## VI. CONCLUDING REMARKS

Within gRPA, we find no trace of a  $\mathbf{q} = 0$  current instability that has been proposed to exist in the three-band model. Recently, Weber *et al.* [9] concluded that the charge-transfer energy,  $\epsilon_d - \epsilon_p$ , is one of the key parameters for this instability. However, over an exhaustive range of this parameter, our calculations did not trace any instability or a sizable enhancement of the  $\mathbf{q} = 0$  current susceptibility. Intra-unit cell magnetism has been inferred from spin-flip neutron scattering experiments [3,5]. At this stage, our calculations offer no explanation for this observation.

The close connection between axial charge order with a  $d$ -wave form factor [31] and oxygen orbitals makes the three-band model a necessary starting point for the microscopic theory. In this context, the focus has so far been on spin fluctuations, and hence on the local Coulomb interactions  $U_d$  and  $U_p$ . Our findings, however, point to the important role of nonlocal interactions. The development of accurate numerical tools capable of handling multi-orbital models with intermediate to strong and nonlocal interactions is, therefore, on demand.

We finally note that, for the typical parameter values used in this work, an SDW phase will, within gRPA, in fact set in at a higher temperature than the  $\pi\text{LC}$  phase. This SDW is driven by the local Coulomb interaction  $U_d$  on the Cu sites, and it is known that correlation effects beyond the approximations discussed here suppress this SDW. Indeed, the feedback of

spin fluctuations renormalizes the electronic structure in a way that reduces the tendency towards antiferromagnetism. We have neglected this aspect here, since we are primarily interested in the charge degree of freedom in the underdoped part of the cuprate phase diagram where pseudogap and charge order occurs. Furthermore, we confirmed that the local interactions  $U_d$  and  $U_p$ , the key parameters which control antiferromagnetism and spin fluctuations, do not have any effect on the emergence of the  $\pi$ LC phase.

It remains yet to be explored how self-energy corrections in our present approach will influence the instability towards spontaneous loop currents. This is on the agenda for further work on loop currents in the three-orbital model for cuprates.

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### APPENDIX A: CURRENT OPERATORS

The current operator is conventionally defined as

$$\hat{J}_{ij} = -it_{ij}(\hat{c}_i^\dagger \hat{c}_j - \hat{c}_j^\dagger \hat{c}_i). \quad (\text{A1})$$

At this point, however, it is important to take into account that we are working with a Hamiltonian which is gauge transformed according to Eqs. (6)–(9). In the transformed operator basis, the current operators in real space take the following form:

$$J_{id,jx} = t_{id,jx}(\hat{c}_{id}^\dagger \hat{c}_{jx} + \hat{c}_{jx}^\dagger \hat{c}_{id}), \quad (\text{A2})$$

$$J_{ix,jd} = -t_{ix,jd}(\hat{c}_{id}^\dagger \hat{c}_{jx} + \hat{c}_{jx}^\dagger \hat{c}_{id}), \quad (\text{A3})$$

$$J_{id,jy} = t_{id,jy}(\hat{c}_{id}^\dagger \hat{c}_{jy} + \hat{c}_{jy}^\dagger \hat{c}_{id}), \quad (\text{A4})$$

$$J_{iy,jd} = -t_{iy,jd}(\hat{c}_{id}^\dagger \hat{c}_{jy} + \hat{c}_{jy}^\dagger \hat{c}_{id}), \quad (\text{A5})$$

$$J_{ix,jy} = -it_{ix,jy}(\hat{c}_{ix}^\dagger \hat{c}_{jy} - \hat{c}_{jy}^\dagger \hat{c}_{ix}), \quad (\text{A6})$$

$$J_{iy,jx} = -it_{iy,jx}(\hat{c}_{iy}^\dagger \hat{c}_{jx} - \hat{c}_{jx}^\dagger \hat{c}_{iy}). \quad (\text{A7})$$

### APPENDIX B: ESTIMATING THE RESULTING MAGNETIC MOMENT

Here, we perform a simple estimate of the magnetic moment associated with the  $\pi$ LC loop currents.

We start by expressing the currents in absolute units. The unit of the current operators is  $e[t_{ij}]/[\hbar] = C(eV)/(eV \cdot s) = C/s$ . For  $z_{pd} = 0.04t_{pd}$ , the current along  $p-d$  bonds is  $I_{pd} = z_{pd} \times (e/\hbar) = 0.04t_{pd} \times (2.4 \times 10^{-4})A = 9.6t_{pd} \mu A$ , where  $t_{pd}$  is measured in eV. Band structure calculations suggest  $t_{pd} \approx 1.6$  eV, yielding  $I_{pd} = 16 \mu A$ ; experimental bandwidths, however, are typically a factor of 3 smaller than predicted by band structure calculations suggesting  $I_{pd} \sim 5 \mu A$ . This is comparable to other estimates of loop current amplitudes: Within a cluster calculation of multiorbital  $t-J$  model, the upper bound of  $\theta_{II}$ -like loop currents was previously estimated to be between  $5 \mu A$  and  $15 \mu A$  for different parameter sets [10]; similarly, in the single band DDW studies, the staggered loop currents were estimated to be  $\sim 7 \mu A$  by assuming a DDW gap value of  $\sim 0.03$  eV [36].

Next, we calculate the magnetic moments of loop currents using the formula  $M = I\eta$  where  $I$  is the current, and  $\eta$  is the area enclosed by the loop. This is indeed a crude calculation, however, we believe this should yield a qualitatively correct number. The total magnetic moment in a given plaquette has two contributions:  $I_b\eta_b$  and  $I_g\eta_g$  due to two independent current loops (shown in black and green in Fig. 1). We set  $I_b = 5 \mu A$ , and since it circulates around the whole plaquette,  $\eta_b = a^2$ . As given by the eigenvectors of  $\chi^J(q)$ ,  $I_g \approx I_{pd}/3$ , and it circulates around an area of  $\eta_g = a^2/2$ . Thus, the magnetic moment is calculated as

$$M = I_b\eta_b + I_g\eta_g \quad (\text{B1})$$

$$= I_{pd} \times a^2 + I_{pd}/3 \times a^2/2 \quad (\text{B2})$$

$$= 7I_{pd}a^2/6 \quad (\text{B3})$$

$$= 7(5\mu A)(3.85 \times 10^{-10}m)^2/6 \quad (\text{B4})$$

$$\approx 0.09\mu_B. \quad (\text{B5})$$

However, since  $I_{pd}$  is shared by the neighboring plaquettes, the effective moment for individual plaquettes might be reduced to  $\sim 0.05\mu_B$ .

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