Phonon-Induced Emergent Charge Order in Cuprates

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Charge-density modulations are inevitably accompanied by a distortion of the crystalline lattice. Electronic or phononic origins of charge order are therefore difficult to distinguish. Here we propose a mechanism which traces the charge-density wave formation in underdoped cuprates to the incipient softening of a bond-buckling phonon. The momentum dependence of its coupling to the electrons in the copper oxygen planes favourably selects the incommensurate and axial ordering wavevector. But, it requires strong electronic correlations via their cuprate specific renormalization of the weight and the dispersion of quasiparticles to enable a unique enhancement of the charge susceptibility near the $B_{1g}$-phonon selected wavevector. The frequency of the $B_{1g}$ phonon softens by a few percent, and a lattice instability with concomitant finite range charge-density wave correlations will form locally, if nucleated by materials’ defects or dopant disorder.

The discovery of charge-density wave (CDW) order [1] in underdoped cuprates raised the question of whether it is intimately related to pseudogap physics [2–7], and thereby yet another signature of strong electronic correlations. Charge modulations with a moderate correlation length are detected by x-ray techniques [8–12] and scanning tunneling microscopy (STM) [9, 10, 13] at incommensurate wave-vectors near $q_{CO} = 0.3$ reciprocal lattice units, oriented along the crystalline axes with either uniaxial or biaxial character. The CDW wavevector $q_{CO}$ continuously drops with increasing hole doping in YBa$_2$Cu$_3$O$_{6+x}$ (YBCO) and Bi$_2$Sr$_2$-La$_x$CuO$_{6+x}$ (Bi2201) [1, 9, 12, 14]. Both, the axial orientation of $q_{CO}$ and its doping variation proved difficult to reconcile with purely electronic model calculations [15, 16]. Weak-coupling theories generically predict that $q_{CO}$ lies on the Brillouin zone (BZ) diagonal [5, 17, 18] unless the CDW instability is preceded by a Fermi surface reconstruction to form hole pockets [7]. In the continuing search for the origin of charge order in cuprates, we employ further experimental facts. One is the discovery of charge order in overdoped Bi2201 with an unreconstructed Fermi surface [19]. This result poses a question as to whether the CDW is actually tied to the pseudogap phenomena.

A second experimental hint relates to the softening or broadening of phonon modes at the CDW wavevector [20–22] and to giant phonon anomalies near the CDW instability, which all point to a strong electron-phonon (el-ph) coupling [23]. While the phonon dispersion will necessarily react to a charge-density modulation, phonons may also play a key role in CDW formation, for example selecting the ordering wavevector in CDW-susceptible materials [24, 25]. In cuprates, the frequencies of the phonons decrease only weakly, and a continuous softening to zero frequency is likely precluded by quenched disorder [26].

A third hint comes from the atomic displacement pattern that accompanies the CDW. In charge-ordered YBCO, x-ray diffraction[27] found that the planar oxygen atoms have the largest displacements; they shift out-of-plane with an out-of-phase pattern (Fig. 1) that closely resembles the normal mode of the $B_{1g}$ bond-buckling phonon [28].

FIG. 1. A representation of the out-of-plane oxygen vibrations ($B_{1g}$ pattern) in a copper-oxide bilayer. The arrows represent the out-of-phase motions of the oxygen atoms.

Motivated by these experimental facts, we obtain the Landau free energy for a $B_{1g}$-phonon-mediated CDW. We start from a microscopic model for the CuO$_2$ planes, and find that the structure of the el-ph coupling matrix element $g(\mathbf{q}; \mathbf{k})$ depends strongly on the orbital content of the Fermi surface. In particular, the Cu-4s orbital is crucial; with it, $g(\mathbf{q}; \mathbf{k})$ is maximum at phonon momenta $\mathbf{q}^*$ that are axial (rather than diagonal) and track the doping dependence of the CDW wavevector. The special role of $\mathbf{q}^*$ is imperceptible in the free energy when calculated with bare electron dispersions; however, with electronic correlations, modeled by a renormalization of the band dispersion and quasiparticle spectral weight, an axial wavevector close to $\mathbf{q}^*$ emerges as the dominant wavevector in the CDW susceptibility. The $B_{1g}$ mode is, by itself, too weak to induce true long-range order; but the inevitable presence of disorder will induce short-range CDW correlations at a wavevector selected by the
mode. This leads us to propose a scenario, in which a phonon-based mechanism is enabled by strong electronic correlations, and the incommensurate wavevector of the concomitant charge correlations is dictated by the momentum-space structure of the el-ph coupling matrix element.

We start by modeling a single CuO$_2$ plane in YBCO including copper 4s and 3d$_{z^2}$ - $\gamma^2$ as well as oxygen p$_x$ and p$_y$ orbitals. An effective three-band model is obtained in terms of the d- and p- orbitals by downfolding the original four-band model to [7, 29]

$$H_{kin} = \sum_{k,\sigma} \Psi_{k,\sigma}^\dagger \begin{pmatrix} \varepsilon_d & 2t_{pd}s x & -2t_{pd}s y \\ 2t_{pd}s x & \tilde{\varepsilon}_x(k) & 4t_{pp}s z s y \\ -2t_{pd}s y & 4t_{pp}s z s y & \tilde{\varepsilon}_y(k) \end{pmatrix} \Psi_{k,\sigma},$$

(1)

with the three-spinor $\Psi_{k,\sigma} = (d_{k,\sigma}^x, d_{k,\sigma}^y, p_{k,\sigma}^d, p_{k,\sigma}^s)$ and $s_{x,y} = \sin(k_{x,y})/2$. $\varepsilon_d$ denotes the onsite energy of the d orbital, $t_{pd}$ and $t_{ps}$ are the hopping amplitudes between p- and d- and p- and s- orbitals, respectively. In the downfolding procedure, the hopping processes via the copper 4s orbital renormalize the oxygen energies $\varepsilon_p$ and generate indirect hopping $4t_{pp}$ between oxygen orbitals:

$$\tilde{\varepsilon}_{x,y} = \varepsilon_p + 4t_{pp}s_{x,y}^2; \quad t_{pp} = t_{pp}^d + t_{pp}^s; \quad t_{pp}^d = \frac{t_{ps}^2}{\varepsilon_p - \varepsilon_s},$$

(2)

where $\varepsilon_p$ is the Fermi energy and $t_{pp}^d$ a small direct hopping amplitude. We choose parameter values in order to achieve a close resemblance to the Fermi surfaces of bilayer cuprates [7]. The value of $t_{pd} = 1.6$ eV is taken from Ref. [29]. The Hamiltonian parameters used below are $\varepsilon_d - \varepsilon_p = 0.9$ eV, $t_{pp}^d = 0$ eV and $t_{pp}^s = -1.0$ eV. We diagonalize $H_{kin}$ and focus only on the partially filled anti-bonding band; the irrelevant spin index is subsequently suppressed.

The out-of-plane $B_{1g}$ vibrations of the oxygen atoms, as depicted in Fig. 1, naturally couple linearly to their local electric field $E_z$ [30]. At this point, we neglect the motion of the almost four times heavier copper atoms. Consequently, we start from the ansatz

$$H_{el-ph} = eE_z \sum_{n} \left[ \delta u_{zn} p_{zn} + \delta u_{yn} p_{yn} \right],$$

(3)

where $\delta u_{zn}$ are the out-of-plane displacements of the oxygen atoms in unit cell $n$. We project the el-ph Hamiltonian in Eq. 3, onto the anti-bonding band and obtain the effective Hamiltonian $H = H_{el-ph} + H_{ph}$ with

$$H_{el-ph} = \sum_k \tilde{\varepsilon}_k c_k^\dagger c_k + \sum_{k,q} \tilde{g}(q; k) c_{k+q}^\dagger c_k \left( a_q + a_{-q}^\dagger \right),$$

(4)

where $c_k^\dagger$ is the creation operator for the anti-bonding electrons with dispersion $\varepsilon_k$; $a_q$ annihilates a $B_{1g}$ phonon mode, and $H_{ph} = \hbar \Omega_p \sum_q \Omega_{q_n} a_q^\dagger a_q$ (see Ref. [30] and Supplementary Materials). The momentum-dependent el-ph coupling is written $\tilde{g}(q; k) = \gamma \tilde{g}(q; k)$ [31], where $\gamma$ is the coupling strength and

$$\tilde{g}(q; k) = \left[ e^{\imath q \cdot \phi_x(k')} \phi_x(k) + e^{\imath q \cdot \phi_y(k')} \phi_y(k) \right],$$

(5)

with $k' = k + q$. The eigenfunctions $\phi_x,y$ signify the orbital contents of the oxygen p$_x,y$ orbitals in the anti-bonding band. Similarly, the eigenvectors $e^{\imath q \cdot \phi}$ correspond to the normal mode of the out-of-plane displacements of the two oxygen atoms in the CuO$_2$ unit cell. The overall strength of the el-ph coupling is $\gamma = eE_z \sqrt{\hbar/2m\Omega_P}$, where $m$ is the mass of the oxygen atom and $\Omega_P \sim 40$ meV is the frequency of the dispersionless $B_{1g}$ mode. Adopting the electric-field value $eE_z = 3.56$ eV/Å from Ref. [32] leads to $\gamma = 0.22$ eV. The eigenvectors for the $B_{1g}$ mode are $e^{\imath q \cdot \phi}$, where the normalization factor $M_q = \sqrt{\cos^2(q_x/2) + \cos^2(q_y/2)}$.

*Earlier theoretical work [28, 33] on the $B_{1g}$ phonon in optimally doped Bi-2212, argued that for an antinodal fermion state, $|\tilde{g}(q; k)|^2$ is strongest for an axial scattering wavevector $q$ to the nearby antinodal final state. This value of $q$ is considerably smaller than $q_{CO}$. Instead we find, when the Cu-4s orbital is properly included via the finite indirect hopping $t_{pp}^d$ in Eq. 2, the anisotropic structure of $\tilde{g}(q; k)$ changes qualitatively. The maximum of the coupling $|\tilde{g}(q; k)|^2$ now occurs for larger axial wavevectors $q^*$, that connect initial ($k^*_i$) and final ($k^*_f + q^*$) Fermi surface states near the nodal points (see*}

![FIG. 2. (a) Plot of the el-ph coupling $|\tilde{g}(q; k)|^2$ for phonon wavevectors $q$ that connect the initial $k^*_i$ and the scattered $k^*_f + q$ state, both on the Fermi surface at 10% hole doping. The strongest coupling occurs at the axial wavevector $q^*$. (b) The maximum value of $|\tilde{g}(q; k)|^2$ with respect to all initial momenta $k_i$ for axial wavevectors $q = (q_x, 0)$. The global maximum is achieved at $q^*$ for the initial state at $k^*_i$. (c) Spectral function $A^v(k, \varepsilon_F)$ for the oxygen $p_x$-orbital electron on the Fermi surface. (d) The variation of $A^v(k, \varepsilon_F)$ along the Fermi surface parametrized by the angle $\psi_k$ indicated in panel (c).](image-url)
Fig. 2a). The resulting $q^*$ is quantitatively close to experimental values of $q_{CO}$.

Fixing an initial state $k_i$ on the Fermi surface, we evaluate the maxima of $|g(q^*;k_i)|^2$ with respect to $q$ where $k_i + q$ is the final state on the Fermi surface using Nelder-Mead [34] gradient approximation. We repeat this procedure as we vary the initial state $k_i$ along the Fermi surface branch in the first BZ quadrant and thereby identify the global maximum which we denote as $|g(q^*;k^*)|^2$. A plot of $|g(q^*;k^*)|^2$ versus phonon wavevector $q$ is shown in Fig. 2a. The strongest scattering occurs at the axial $q^*$ indicated by the white arrow.

In order to determine what is special about $k^*_i$ and $q^*$, we show the oxygen $p_y$-orbital resolved spectral function $A^y(k,\epsilon_F)$ in Fig. 2c. The highest spectral weight is obtained for $k = k^*_i$. In Fig. 2d we parametrize the position on the Fermi surface by the angle $\psi_k = \tan^{-1}(k_y/k_x)$. This panel indicates that it is the oxygen orbital content on the Fermi surface that determines $k^*_i$ and $q^*$. The variation of the oxygen content is specifically controlled by the indirect hopping processes via the Cu-4s orbital.

We next calculate the doping evolution of $q^*$ and $k^*_i$ and collect the results in Fig. 3. The magnitude of $q^*$ comes close to the observed CDW ordering wavevector 0.3 (r.l.u.) and decreases with hole doping in a similar fashion as detected in x-ray experiments [1, 9–12, 19]. It is therefore tempting to suspect a close connection. To pursue this idea, we return to the el-ph Hamiltonian Eq. 4 and assume a static mean-field lattice distortion

$$\sqrt{\frac{\hbar}{2m\Omega_P}} \left( a_q + a_0^\dagger \right) = \xi_q,$$

with the four possible axial wavevectors $\pm q^*, \pm q^*_{0}$ oriented either along the $x$- or the equivalent $y$- direction.

We perform a linked-cluster expansion for the free energy to low orders in $\Delta_q = eE_0 \xi_q$ (see Supplementary Material and Refs. [35–37]):

$$\mathcal{F} = \mathcal{F}_0 + \sum_{q = q^*,0} \left[ \frac{|\Delta_q|^2}{4\gamma^2} (\hbar\Omega_P - 2\chi_q^{(g)}) + |\Delta_q|^4 \chi_q^{(4)} \right] + |\Delta_q^*|^2 |\Delta_q^*|^2 \chi_q^{(4)}.$$  

(7)

The static susceptibility $\chi_q^{(g)}$ is defined as

$$\chi_q^{(g)} = -2\gamma^2 \sum_k |g(q;k)|^2 \left[ \frac{f(\epsilon_k) - f(\epsilon_k+q)}{\epsilon_k - \epsilon_{k+q}} \right],$$  

(8)

where $f(\epsilon)$ denotes the Fermi distribution function. Upon cooling from high temperature, a lattice instability occurs at the $q$ value for which the coefficient $(\hbar\Omega_P - 2\chi_q^{(g)})$ in the quadratic term first vanishes. This instability will necessarily produce an incommensurate charge modulation at the same $q$. In previous theoretical work, such a criterion was successfully employed to identify the correct CDW wavevectors for weakly correlated tellurides [36]. Upon further cooling, the fourth-order coefficients decide between uniaxial and biaxial charge order. Because the magnitudes of the coefficients, and even the sign of $\chi_q^{(4)}$, depend sensitively on specific parameter choices, it is difficult to make universal statements about the behavior of the quartic terms.

Returning to the quadratic term, we define for comparison the Lindhard function, $\chi_q^{(L)}$, obtained by setting $\tilde{g}(q;k) = 1$ in Eq. 8; as shown in Fig. 4(a), its weight is concentrated near the $(\pi, \pi)$ point without any prominent wavevector related to nesting. Yet, as demonstrated in Refs. [24, 25], the momentum dependence of the el-ph coupling can by itself select the wavevector for a lattice instability and the concomitant charge order. Indeed, if the structure of $\tilde{g}(q;k)$ is incorporated as in Eq. 8, the momentum-space weight of $\chi_q^{(g)}$ redistributes [Fig. 4(b)]. But, a clear instability wavevector still cannot be seen; in particular, the axial wavevector $q^*$ for the global maximum of $|g(q;k)|^2$ remains invisible.

The above analysis for the el-ph Hamiltonian Eq. 4 apparently fails to find an instability at the axial candidate wavevector $q^*$. This naturally compromises the attempt to carry over a strategy, successful for weakly correlated materials, to strongly correlated cuprates. Yet, including phonons in a multi-orbital model with strong electronic correlations is a demanding task. We therefore proceed with a trial ansatz to include correlation physics on a phenomenological level.

The prevailing issue in the physics of underdoped cuprates is the conundrum of the pseudogap [38]. The Fermi surface appears truncated to Fermi arcs centered around the BZ diagonals (the nodal regions) and it is obliterated near the BZ boundaries (the antinodal regions). In essence, well defined quasiparticles exist for near-nodal directions, while they are wiped out towards the antinodes. This motivates a simple ansatz for the
Green function of the anti-bonding band quasiparticles, similar in spirit to Ref. [35]:

$$G(k, \omega) = \frac{Z_k}{\omega - \varepsilon_k + i\delta} + G_{\text{incoh}},$$  \hfill (9a)

where $Z_k$ is the quasiparticle weight tailored to continuous decrease from 1 at the nodal point to zero at the BZ faces. $\psi_{\text{max/min}}$ denotes the largest/smallest angle $\psi_k$ for the Fermi surface momenta $(k_F, \pi)$ and $(\pi, k_F)$, respectively. The k-dependence in Eq. 9b is analogously carried over to the other segments of the Fermi surfaces.

The ansatz in Eq. 9a, 9b leads to the renormalized static susceptibility

$$\chi^{(z)}_q \approx -2\gamma^2 \sum_k Z_k Z^*_k |\bar{g}(q;k)|^2 \left[ f(\varepsilon_k) - f(\varepsilon_{k+q}) \right],$$  \hfill (10)

where the contributions from the incoherent part of $G(k, \omega)$ are neglected (see Supplementary Material).

We examine the impact of the quasiparticle weight factors on the susceptibility $\chi^{(z)}_q$ in Fig. 5a. The highly anisotropic variation of $Z_k$ reduces the susceptibility around the BZ diagonal and creates new structures along the axes including the global maximum at axial wavevectors $q_{CO}^{(2)}$, and $q_{CO}^{(1)}$, which are larger than, but close to, $q^+$ or $q^-$ for which the el-ph coupling is strongest.

In a second step we incorporate—besides the anisotropic weight factors $Z_k$—also the correlation induced renormalization of the band dispersion by replacing $\varepsilon_k \Rightarrow \varepsilon^R_k$ in Eq. 9a and hence also in the square bracket of Eq. 10. For this purpose, we adopt a phenomenological fit to the measured ARPES dispersion applied previously to data for optimally doped Bi-2212 [39]. Compared to the bare dispersion, $\varepsilon^R_k$ has an almost three times narrower bandwidth and the nodal Fermi velocity $v_F$ is similarly reduced, while the shape of the Fermi surface remains almost preserved. The susceptibility $\chi^{(z)}_q$ with the renormalized $\varepsilon^R_k$ is shown in Fig. 5b. We observe that the axial peaks are retained at wavevectors

$$q_{CO}^{(2)}$$ with $q^+ < q_{CO}^{(2)} < q_{CO}^{(1)}$, whereas the peak at the $(\pi, \pi)$ point loses its strength. $q_{CO}^{(2)}$ is about 16% larger than $q^+$ and follows the same doping dependence as $q^+$. Furthermore, the strength of the peak at $q_{CO}^{(2)}$ has tripled. This increase is naturally tied to the downward renormalization of $v_F$. So, we conclude that it apparently requires both a correlation-induced quasiparticle renormalization, and a specific momentum dependence of the el-ph coupling to generate the axial and incommensurate candidate wavevector $q_{CO}^{(2)}$ for the anticipated lattice and concomitant CDW instability.

Without aiming at a quantitative description we nevertheless translate the obtained result into an estimate. The absolute units in Fig. 5b indicate that $\lambda^{(z)}_{q_{CO}^{(2)}} \ll \hbar \Omega_F$, and we are therefore far from meeting the mean-field instability criterion. Nonetheless, the el-ph coupling inevitably also alters the frequency of the participating phonons. Based on Ref. [40], the renormalized $B_{1g}$ phonon frequency $\tilde{\omega}_q$ follows from

$$\tilde{\omega}_q^2 = \Omega_F^2 (1 - \lambda_q), \quad \lambda_q = \frac{2\lambda^{(z)}_{q_{CO}^{(2)}}}{\hbar \Omega_F},$$  \hfill (11)

where $\lambda^{(z)}_{q_{CO}^{(2)}}$ is the real part of the susceptibility at the bare phonon frequency $\Omega_F$. At $110K$ and $q = q_{CO}^{(2)}$, the dynamical susceptibility is almost twice as large as its static zero-frequency value, from which Eq. 11 gives $\tilde{\omega}_q = 0.97 \Omega_F$, i.e. a softening of 3%. Upon cooling $\lambda^{(z)}_{q_{CO}^{(2)}}$ increases and the softening reaches ~5% at around 30K. For reference we mention that in Ref. [20] a softening of the $B_{1g}$ phonon frequency in YBa$_2$Cu$_3$O$_7$ of about 6% was measured at $3K$ for $q = (0,0,3)$.

Several factors may act to enhance the el-ph coupling in underdoped cuprates. We recall that a dispersion $\varepsilon^R_k$ for an optimally doped material was used for the evaluation of $\chi^{(z)}_q$. On the underdoped side, the nodal Fermi velocity in Bi-2212 drops by as much as 50% [41]. Such a drop necessarily enhances $\chi^{(z)}_{q_{CO}^{(2)}}$ further and the corresponding phonon softening is estimated to rise to about 10% reflecting enhanced CDW correlations at wavevec-
tors $q^{(2)}_{CO}$ for underdoped materials. Furthermore, strong correlations in the underdoped cuprates decrease the copper and increase the oxygen orbital content on the Fermi surface, thereby enlarging the susceptibility at $q^{(2)}_{CO}$.

These estimates indicate that the lattice driven mechanism is too weak to generate a long range ordered CDW state. Still, the charge order in underdoped cuprates is in fact short ranged with moderate planar correlation lengths [8, 11, 12], and is most likely nucleated by defects [26]. A strong magnetic field [42, 43] or uniaxial pressure [21] is needed to enhance the planar correlation length and to even achieve 3D CDW order. The magnetic field suppress superconductivity, which otherwise stops the charge correlations from growing upon cooling.

Our proposed mechanism therefore appears compatible with experimental observations. The momentum-dependence of the $el$-$ph$ coupling to the $B_{1g}$ bond-buckling phonon and the specific the variation of the oxygen orbital content on the Fermi surface select an incommensurate, axial wavevector $q^*$ for which the lattice is most susceptible to deform. But only in conjunction with the strong and anisotropic renormalization of the correlated electrons in the copper oxygen planes does the corresponding susceptibility develop the required enhancement near $q^*$ to move the $el$-$ph$ systems at least towards a charge ordered state with an axial wavevector $q^{(2)}_{CO}$ near $q^*$. It remains to be explored whether a phonon based mechanism carries over to explain also the CDW systems at least $\ast$. A strong magnetic field [42, 43] or uniaxial pressure [21] is needed to enhance the planar correlation length and to even achieve 3D CDW order. The magnetic field suppress superconductivity, which otherwise stops the charge correlations from growing upon cooling.

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Supplementary Material: – Phonon-Induced Emergent Charge Order in Cuprates

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Anti-bonding Band: Dispersion and eigenfunctions

Diagonalizing the downfolded Hamiltonian \( H_{\text{kin}} \) (Eq. (1) in the main text), we obtain energy for the anti-bonding band as

\[
\varepsilon_k = \frac{\varepsilon_d + \varepsilon_x + \varepsilon_y}{3} + \left( A_k + \sqrt{A_k^2 + B_k^2} \right)^{\frac{3}{2}} - \left( A_k - \sqrt{A_k^2 + B_k^2} \right)^{\frac{3}{2}},
\]

where the parameters \( A_k \) and \( B_k \) are defined as

\[
\begin{align*}
A_k &= \frac{\varepsilon_d}{6} \left( t_x^2 + t_y^2 - 2t^2 \right) + \frac{\varepsilon_x}{6} \left( t_x^2 + t_y^2 - 2t^2 \right) + \frac{\varepsilon_y}{6} \left( t_x^2 + t_y^2 - 2t^2 \right) + t' x y + \varepsilon_{\text{AF}} \varepsilon_{\text{AF}} + \varepsilon_{\text{AF}} \varepsilon_{\text{AF}} + \varepsilon_{\text{AF}} \varepsilon_{\text{AF}} + \frac{2\varepsilon_{\text{AF}} \varepsilon_{\text{AF}}}{9} \\
B_k &= -\frac{\varepsilon_x^2 + \varepsilon_y^2 - \varepsilon_x \varepsilon_y - \varepsilon_x \varepsilon_y}{9} - \frac{1}{3} \left( t_x^2 + t_y^2 + t^2 \right),
\end{align*}
\]

where \( t_x = 2t_p d s_x, t_y = 2t_p d s_y \) and \( t' = \frac{1}{4} t_p p x s_y y \). The projections of \( d, p_x \) and \( p_y \) orbitals onto the anti-bonding band follow from the eigenfunction for the anti-bonding band as

\[
\phi_d(k) = \frac{1}{N_k} \left[ (\varepsilon_k - \varepsilon_x)(\varepsilon_k - \varepsilon_y) - t'^2 \right], \quad \phi_x(k) = \frac{1}{N_k} \left[ (\varepsilon_k - \varepsilon_x)x - t' y \right], \quad \phi_y(k) = \frac{1}{N_k} \left[ (\varepsilon_k - \varepsilon_x)y - t' x \right],
\]

\[
N_k = \sqrt{\left[ (\varepsilon_k - \varepsilon_x)(\varepsilon_k - \varepsilon_y) - t'^2 \right]^2 + \left[ (\varepsilon_k - \varepsilon_x)x - t' y \right]^2 + \left[ (\varepsilon_k - \varepsilon_x)y - t' x \right]^2}.
\]

We notice that \((A_k^2 + B_k^2)\) is negative for all \( k \) in the Brillouin zone. Hence, we can write Eq. 1 as

\[
\varepsilon_k = \frac{\varepsilon_d + \varepsilon_x + \varepsilon_y}{3} + 2\text{Re} \left( A_k + \sqrt{A_k^2 + B_k^2} \right)^{\frac{3}{2}},
\]

therefore the anti-bonding band energy is real. A simplified version of Eq. (3) and Eq. (4) was obtained in Refs. [1, 2].

We show the results for the most general three-band Hamiltonian model.

Lattice dynamical model: \( B_{1g} \) oxygen vibration

In this section, we analyze the eigenmodes for the out-of-plane \( B_{1g} \) oxygen vibrations in a \( \text{CuO}_2 \) plane. We assume a three-atom unit cell of a copper and two oxygen atoms \( \text{O}(2,3) \). The \( B_{1g} \) dispersion can be modeled by two spring constants: the first, \( K \), is between O atoms and nearest-neighbour copper sites (Fig. 1); the second, \( K' \), represents a dispersionless coupling between the O atoms and the rest of the lattice. Labeling the positions of Cu, O(2) and O(3) by \( \text{u}_1(\text{R}), \text{u}_2(\text{R}) \) and \( \text{u}_3(\text{R}) \), we write the coupled equations of atomic motion as \( (a \text{ is the lattice constant}) \)

\[
M \ddot{\text{u}}_1(\text{R}_{mn}) = -K \left( \text{u}_1(\text{R}_{mn}) - \text{u}_2(\text{R}_{mn} + \frac{a}{2} \hat{i}) \right) - K \left( \text{u}_1(\text{R}_{mn}) - \text{u}_2(\text{R}_{mn} - \frac{a}{2} \hat{j}) \right)
\]

\[
m \ddot{\text{u}}_2(\text{R}_{mn} + \frac{a}{2} \hat{i}) = -K \left( \text{u}_2(\text{R}_{mn} + \frac{a}{2} \hat{i}) - \text{u}_1(\text{R}_{mn}) \right) - K \left( \text{u}_2(\text{R}_{mn} + \frac{a}{2} \hat{i}) - \text{u}_1(\text{R}_{mn} + a \hat{j}) \right) - K' \left( \text{u}_2(\text{R}_{mn} + \frac{a}{2} \hat{i}) \right)
\]

\[
m \ddot{\text{u}}_3(\text{R}_{mn} + \frac{a}{2} \hat{j}) = -K \left( \text{u}_3(\text{R}_{mn} + \frac{a}{2} \hat{j}) - \text{u}_1(\text{R}_{mn}) \right) - K \left( \text{u}_3(\text{R}_{mn} + \frac{a}{2} \hat{j}) - \text{u}_1(\text{R}_{mn} + a \hat{j}) \right) - K' \left( \text{u}_3(\text{R}_{mn} + \frac{a}{2} \hat{j}) \right)
\]
FIG. 1. (a) The three-atom unit cell with two oxygen atoms O(2) and O(3) coupled to copper via a uniform force constant $K$. The out-of-plane motions of the oxygen atoms are constrained by a uniform tension with force constant $K'$.

where $M$ and $m$ are the masses of copper and oxygen atoms respectively.

$$
\begin{bmatrix}
\frac{4K}{M} & -\frac{2K}{\sqrt{M}m} \cos \left(\frac{q_x}{2}\right) & -\frac{2K}{\sqrt{M}m} \cos \left(\frac{q_y}{2}\right) \\
-\frac{2K}{\sqrt{M}m} \cos \left(\frac{q_x}{2}\right) & 0 & \frac{2K}{m} \sqrt{\frac{M}{m}} \\
-\frac{2K}{\sqrt{M}m} \cos \left(\frac{q_y}{2}\right) & \frac{2K}{m} \sqrt{\frac{M}{m}} & 0
\end{bmatrix}
\begin{bmatrix}
u_{1q} \\ u_{2q} \\ u_{3q}
\end{bmatrix}
= \omega^2
\begin{bmatrix}
u_{1q} \\ u_{2q} \\ u_{3q}
\end{bmatrix}.
$$

(6)

$$
\omega_1^2(q) = \frac{2K}{m}; \quad \omega_2^2(q) = 0; \quad \omega_3^2(q) = \frac{2K}{m}
$$

(7)

$$
\eta_1 = \begin{bmatrix}
0 \\
-\cos \left(\frac{q_x}{2}\right) \\
\cos \left(\frac{q_y}{2}\right)
\end{bmatrix}, \quad \eta_2 = \begin{bmatrix}
C_q \\
\cos \left(\frac{q_x}{2}\right) \\
\cos \left(\frac{q_y}{2}\right)
\end{bmatrix}, \quad \eta_3 = \begin{bmatrix}
-C_q \\
\cos \left(\frac{q_x}{2}\right) \\
\cos \left(\frac{q_y}{2}\right)
\end{bmatrix}
$$

where the variable $C_q$ is defined as

$$
C_q = \frac{2K \sqrt{mM} \left(M - 2m\right) + \sqrt{\left(16K^2 m^3 M + 4K^2 M^3 m + 8K^2 m^2 M^2 (\cos q_x + \cos q_y)\right)}}{4KMm}.
$$

(8)

We now notice that $M = 63.546u$ and $m = 15.9u$, and thus make the simplifying assumption $M \gg m$. In this limit, the normalized $B_{1g}$ eigendisplacement from Eq. (7) is

$$
e^x_q = -\hat{z} \frac{\cos \left(\frac{q_x}{2}\right)}{\sqrt{\cos^2 \left(\frac{q_x}{2}\right) + \cos^2 \left(\frac{q_y}{2}\right)}}, \quad e^y_q = \hat{z} \frac{\cos \left(\frac{q_y}{2}\right)}{\sqrt{\cos^2 \left(\frac{q_x}{2}\right) + \cos^2 \left(\frac{q_y}{2}\right)}}.
$$

(9)

Our dynamical phonon model is specifically tailored to obtain the eigendisplacements for the $B_{1g}$ vibrations.

**Linked cluster Expansion: Free energy**

In this section, we outline the derivation of the mean-field Landau Free energy (Eq. (7) in the main text). The mean-field $el$-$ph$ Hamiltonian for the static mean-field lattice distortion $\xi_q$ follows from (Eq. (4) in the main text) as

$$
H_{MF} = \sum_{k} \varepsilon_k c_k^\dagger c_k + \sum_{k} \sum_{q = \pm q, \pm q^*} \gamma \frac{2m\Omega_P}{h} \tilde{g}(q; k) \xi_q c_k^\dagger c_{k+q} + \frac{m\Omega_P^2}{2} \sum_{q = \pm q, \pm q^*} |\xi_q|^2.
$$

(10)
Following Eq. (10), the mean-field $\xi_q$ is evaluated by minimizing the Landau free energy of the coupled $el$-$ph$ system. Utilizing the linked cluster theorem, the latter is obtained as

$$F = F_0 + \frac{1}{2} \sum_{q} n \Omega_q^2 |\xi_q|^2 - \sum_{i=1}^{\infty} U_i,$$  \hspace{1cm} (11)$$

where $F_0$ is the free energy of the electronic system resulting from the Hamiltonian $H_0$. The second term in Eq. (11) follows from the phonon Hamiltonian $H_{Ph}$. Finally, the linked cluster coefficients $U_i$ are obtained from the $el$-$ph$ Hamiltonian $H_{EP}$ as (see Ref. [3] for a detailed discussion about linked cluster expansion)

$$U_i = \frac{(-1)^i}{i!} \int_0^\beta \int_0^\beta \int_0^\beta \langle T_\tau H_{EP}(\tau_1)H_{EP}(\tau_2)\ldots H_{EP}(\tau_i) \rangle_{\text{connected}}. \hspace{1cm} (12)$$

The notation $\langle \cdot \rangle_{\text{connected}}$ in Eq. (12) refers to all the distinct connected diagrams in the perturbative expansion of the mean-field Hamiltonian in Eq. (10). Restricting the choice of the ansatz wavevector $q$ to only fundamental wavevectors $\pm q^\ast, \pm \bar{q}$, we expand the last term in Eq. (11) up to fourth order to obtain the coefficients

$$U_2 = \frac{1}{2\beta} \sum_k \sum_{q=\pm q^\ast, \pm \bar{q}} \sum_{i\omega_n} |\Delta(q)\tilde{g}(q; k)|^2 G^0(k + q, i\omega_n)G^0(k, i\omega_n), \hspace{1cm} (13a)$$

$$U_4 = \frac{1}{4\beta} \sum_{k_1, k_4} \sum_{q_1 = \pm q^\ast, \pm \bar{q}^\ast} \sum_{i\omega_n} \left[ \prod_{i=1}^4 \Delta(q_i)\tilde{g}(q_i; k_i)G^0(k_i + q_i, i\omega_n) \right] \hspace{1cm} (13b)$$

where we have rewritten the $el$-$ph$ Hamiltonian $H_{EP}$ as

$$H_{EP} = \sum_k \sum_{q=\pm q^\ast, \pm \bar{q}^\ast} \Delta(q)\tilde{g}(q; k)c_{k+q}^\dagger c_k, \hspace{1cm} (14)$$

The quantity $\Delta(q)$ in Eq. (14) was defined in Eq. (7) in the main text. The primed product in Eq. (13b) implies momentum conservation at the vertices of Fig. 2(b), which in turn restricts the choices of the phonon scattering momenta $q_i$'s as

$$k_1 + q_1 = k_2, \quad k_2 + q_2 = k_3, \quad k_3 + q_3 = k_4, \quad k_4 + q_4 = k_1. \hspace{1cm} (15)$$

Adding each term in Eq. (15), we obtain the following constraint on the momenta $q_i$'s (in addition to the restriction $q_i \in (\pm q^\ast, \pm \bar{q}^\ast)$) as

$$q_1 + q_2 + q_3 + q_4 = 0. \hspace{1cm} (16)$$

Similar constraints on the phonon scattering momenta $q_i$'s also arise for the odd order linked cluster terms. However, such conditions can only be satisfied for even order terms as $q_i \in (\pm q^\ast, \pm \bar{q}^\ast)$ and consequently all the odd order cluster terms are zero.
Performing the frequency summation in Eq. (13), we obtain the fourth-order coefficient \( \chi^{(4)}_q \) as (Eq. (7) in the main text)

\[
\chi^{(4)}_q = -\sum_k \frac{1}{2} \frac{(\hat{g}(q; k))^4}{(\varepsilon_k - \varepsilon_k')^2} \left( \frac{2f(\varepsilon_k') - f(\varepsilon_k)}{\varepsilon_k - \varepsilon_k'} + (f(\varepsilon_k') + f(\varepsilon_k')) \right) - \sum_k (\hat{g}(q; k))^2(\hat{g}(q; k + q))^2. \tag{17}
\]

The final fourth-order coefficient \( \chi^{(4)}_{q'\cdot q'} \) is obtained as follows

\[
\chi^{(4)}_{q'\cdot q'} = -\sum_k (\hat{g}(q'; k))^2(\hat{g}(q'; k))^2 \left[ \frac{f(\varepsilon_k + q') - f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} + \frac{\beta f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} \right] - \sum_k (\hat{g}(q'; k))^2(\hat{g}(q'; k)) \left[ \frac{f(\varepsilon_k + q') - f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} - \frac{\beta f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} \right] - \sum_k (\hat{g}(q'; k))^2(\hat{g}(q'; k)) \left[ \frac{f(\varepsilon_k + q') - f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} - \frac{\beta f(\varepsilon_k)}{(\varepsilon_k + q' - \varepsilon_k')^2} \right] \tag{18}
\]

The fourth-order coefficient \( \chi^{(4)}_{q'\cdot q'} \) decides between the possible uniaxial or biaxial character of the emergent charge order. However, both its magnitude and the sign depend sensitively on the parameters specific to the system. Hence, one cannot make any general statements about the character of the pattern of the resulting charge modulation.

**Renormalized charge susceptibility: Pseudogap phase**

In this section, we derive the renormalized charge susceptibility as described in main text in Eq. (11). We obtain the spectral function from Eq. (9) (in the main text) as [4]

\[
A(k, \omega) = -\frac{1}{\pi} \text{Im}[G(k, \omega)] = Z_k \delta(\omega - \varepsilon_k) + \frac{1 - Z_k}{2W} F(\omega), \tag{19}
\]

where the last term is our ansatz for \( \text{Im}[G_{mc}] \). \( W \) in Eq. 19 is the band-width of the anti-bonding band. The function \( F(\omega) \) is defined as

\[
F(\omega) = \begin{cases} 
1; & -W \leq \omega \leq W \\
0; & \text{else} 
\end{cases} \tag{20}
\]
FIG. 3. The renormalized quasiparticle weight ansatz $Z_k$ along the Fermi surface at 10% doping. The nodal points are maximally weighted whereas at the Brillouin zone faces the weight drops to zero.

Utilizing the spectral representation of the Green’s function we obtain the renormalized static charge susceptibility as

$$\chi^R_q = -2 \sum_k |\tilde{g}(q; k)|^2 \left[ Z_k Z_{k'} f(\varepsilon_k) - f(\varepsilon_{k'}) + \frac{Z_k(1 - Z_{k'})}{2W} \int_{-W}^{W} d\omega' \frac{f(\varepsilon_k) - f(\omega')}{i\omega_m + \varepsilon_k - \omega'} \right] (21)$$

We now define the renormalized charge susceptibility $\chi^{(gZ)}_q$ as following

$$\chi^{(gZ)}_q = -2\gamma^2 \sum_k Z_k Z_{k'} |\tilde{g}(q; k)|^2 \left[ \frac{f(\varepsilon_k) - f(\varepsilon_{k'})}{\varepsilon_k - \varepsilon_{k'}} \right], (22)$$

where the last three terms in Eq. 21 have been neglected. We evaluate these terms at zero temperature and notice that all of them scale with the band-width $W$ as $(\log W)/W$. Hence, these terms are quantitatively negligible and the renormalized charge susceptibility is defined as in Eq. (10) in the main text.

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