

Supplementary Material:
Emergent Charge Order from Correlated Electron-Phonon Physics in Cuprates

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SUPPLEMENTARY NOTE 1: Anti-bonding Band: Dispersion and eigenfunctions

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

$$\varepsilon_{\mathbf{k}} = \frac{\varepsilon_d + \tilde{\varepsilon}_x + \tilde{\varepsilon}_y}{3} + \left(A_{\mathbf{k}} + \sqrt{A_{\mathbf{k}}^2 + B_{\mathbf{k}}^3} \right)^{\frac{1}{3}} + \left(A_{\mathbf{k}} - \sqrt{A_{\mathbf{k}}^2 + B_{\mathbf{k}}^3} \right)^{\frac{1}{3}}, \quad (1)$$

where the parameters $A_{\mathbf{k}}$ & $B_{\mathbf{k}}$ are defined as

$$\begin{aligned} A_{\mathbf{k}} &= \frac{\varepsilon_d}{6} (t_x^2 + t_y^2 - 2t'^2) + \frac{\tilde{\varepsilon}_x}{6} (t'^2 + t_x^2 - 2t_y^2) + \frac{\tilde{\varepsilon}_y}{6} (t'^2 + t_y^2 - 2t_x^2) - t' t_x t_y \\ &\quad + \frac{\varepsilon_d^3 + \tilde{\varepsilon}_x^3 + \tilde{\varepsilon}_y^3}{27} - \frac{\varepsilon_d^2 \tilde{\varepsilon}_x + \tilde{\varepsilon}_x^2 \varepsilon_d + \tilde{\varepsilon}_x^2 \tilde{\varepsilon}_y + \tilde{\varepsilon}_y^2 \tilde{\varepsilon}_x + \varepsilon_d^2 \tilde{\varepsilon}_y + \tilde{\varepsilon}_y^2 \varepsilon_d}{18} + \frac{2\varepsilon_d \tilde{\varepsilon}_x \tilde{\varepsilon}_y}{9} \\ B_{\mathbf{k}} &= -\frac{\varepsilon_d^2 + \tilde{\varepsilon}_x^2 + \tilde{\varepsilon}_y^2 - \varepsilon_d \tilde{\varepsilon}_x - \tilde{\varepsilon}_x \tilde{\varepsilon}_y - \tilde{\varepsilon}_y \varepsilon_d}{9} - \frac{1}{3} (t_x^2 + t_y^2 + t'^2), \end{aligned} \quad (2)$$

where $t_x = 2t_{pd} s_x$, $t_y = 2t_{pd} s_y$ and $t' = 4\tilde{t}_{pp} s_x s_y$. 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

$$\begin{aligned} \phi_d(\mathbf{k}) &= \frac{1}{N_{\mathbf{k}}} [(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_x)(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_y) - t'^2], \quad \phi_x(\mathbf{k}) = \frac{1}{N_{\mathbf{k}}} [(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_y)t_x - t't_y], \quad \phi_y(\mathbf{k}) = -\frac{1}{N_{\mathbf{k}}} [(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_x)t_y - t't_x] \\ N_{\mathbf{k}} &= \sqrt{[(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_x)(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_y) - t'^2]^2 + [(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_y)t_x - t't_y]^2 + [(\varepsilon_{\mathbf{k}} - \tilde{\varepsilon}_x)t_y - t't_x]^2}. \end{aligned} \quad (3)$$

We notice that $(A_{\mathbf{k}}^2 + B_{\mathbf{k}}^3)$ is negative for all $\mathbf{k}$ in the Brillouin zone. Hence, we can write Eq. 1 as

$$\varepsilon_{\mathbf{k}} = \frac{\varepsilon_d + \tilde{\varepsilon}_x + \tilde{\varepsilon}_y}{3} + 2\text{Re} \left[\left(A_{\mathbf{k}} + \sqrt{A_{\mathbf{k}}^2 + B_{\mathbf{k}}^3} \right)^{\frac{1}{3}} \right], \quad (4)$$

therefore the anti-bonding band energy is real. A simplified version of Eq. (3) and Eq. (4) was obtained in Supplementary References [1, 2]. We show the results for the most general three-band Hamiltonian model.

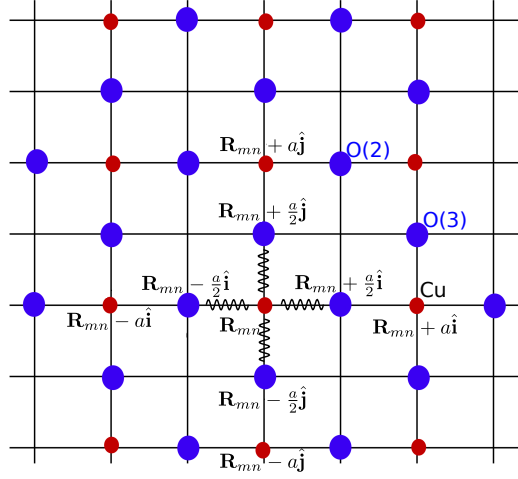
SUPPLEMENTARY NOTE 2: Lattice dynamical model: B_{1g} oxygen vibration

In this section, we analyze the eigenmodes for the out-of-plane B_{1g} oxygen vibrations in a CuO_2 plane. We assume a three-atom unit cell of a copper and two oxygen atoms 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 (Supplementary Figure 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 $\mathbf{u}_1(\mathbf{R})$, $\mathbf{u}_2(\mathbf{R})$ and $\mathbf{u}_3(\mathbf{R})$, we write the coupled equations of atomic motion as (a is the lattice constant)

$$\begin{aligned} M\ddot{\mathbf{u}}_1(\mathbf{R}_{mn}) &= -K \left(\mathbf{u}_1(\mathbf{R}_{mn}) - \mathbf{u}_2(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{i}}) \right) - K \left(\mathbf{u}_1(\mathbf{R}_{mn}) - \mathbf{u}_2(\mathbf{R}_{mn} - \frac{a}{2}\hat{\mathbf{i}}) \right) \\ &\quad - K \left(\mathbf{u}_1(\mathbf{R}_{mn}) - \mathbf{u}_2(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{j}}) \right) - K \left(\mathbf{u}_1(\mathbf{R}_{mn}) - \mathbf{u}_2(\mathbf{R}_{mn} - \frac{a}{2}\hat{\mathbf{j}}) \right) \\ m\ddot{\mathbf{u}}_2(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{i}}) &= -K \left(\mathbf{u}_2(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{i}}) - \mathbf{u}_1(\mathbf{R}_{mn}) \right) - K \left(\mathbf{u}_2(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{i}}) - \mathbf{u}_1(\mathbf{R}_{mn} + a\hat{\mathbf{i}}) \right) - K' u_2^z(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{i}}) \hat{\mathbf{k}} \\ m\ddot{\mathbf{u}}_3(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{j}}) &= -K \left(\mathbf{u}_3(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{j}}) - \mathbf{u}_1(\mathbf{R}_{mn}) \right) - K \left(\mathbf{u}_3(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{j}}) - \mathbf{u}_1(\mathbf{R}_{mn} + a\hat{\mathbf{j}}) \right) - K' u_3^z(\mathbf{R}_{mn} + \frac{a}{2}\hat{\mathbf{j}}) \hat{\mathbf{k}} \end{aligned} \quad (5)$$

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

$$\begin{bmatrix} \frac{4K}{M} & -\frac{2K}{\sqrt{Mm}} \cos(\frac{q_x}{2}) & -\frac{2K}{\sqrt{Mm}} \cos(\frac{q_y}{2}) \\ -\frac{2K}{\sqrt{Mm}} \cos(\frac{q_x}{2}) & \frac{2K+K'}{m} & 0 \\ -\frac{2K}{\sqrt{Mm}} \cos(\frac{q_y}{2}) & 0 & \frac{2K+K'}{m} \end{bmatrix} \begin{bmatrix} \mathbf{u}_{1\mathbf{q}}^z \\ \mathbf{u}_{2\mathbf{q}}^z \\ \mathbf{u}_{3\mathbf{q}}^z \end{bmatrix} = \omega^2 \begin{bmatrix} \mathbf{u}_{1\mathbf{q}}^z \\ \mathbf{u}_{2\mathbf{q}}^z \\ \mathbf{u}_{3\mathbf{q}}^z \end{bmatrix}. \quad (6)$$



Supplementary Figure 1: **A pictorial illustration of planar CuO_2 lattice structure.** (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' .

$$\omega_1^2(\mathbf{q}) = \frac{2K}{m}; \quad \omega_2^2(\mathbf{q}) = 0; \quad \omega_3^2(\mathbf{q}) = \frac{2K}{m} \quad (7)$$

$$\eta_1 = \begin{bmatrix} 0 \\ -\cos(\frac{q_y}{2}) \\ \cos(\frac{q_x}{2}) \end{bmatrix}, \quad \eta_2 = \begin{bmatrix} C_{\mathbf{q}} \\ \cos(\frac{q_x}{2}) \\ \cos(\frac{q_y}{2}) \end{bmatrix}, \quad \eta_3 = \begin{bmatrix} -C_{\mathbf{q}} \\ \cos(\frac{q_x}{2}) \\ \cos(\frac{q_y}{2}) \end{bmatrix}$$

where the variable $C_{\mathbf{q}}$ is defined as

$$C_{\mathbf{q}} = \frac{[2K\sqrt{mM}(M-2m) + \sqrt{\{16K^2m^3M + 4K^2M^3m + 8K^2m^2M^2(\cos q_x + \cos q_y)\}}]}{4KMm}. \quad (8)$$

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

$$e_{\mathbf{q}}^x = -\hat{z} \frac{\cos(\frac{q_y}{2})}{\sqrt{\cos^2(\frac{q_x}{2}) + \cos^2(\frac{q_y}{2})}}; \quad e_{\mathbf{q}}^y = \hat{z} \frac{\cos(\frac{q_x}{2})}{\sqrt{\cos^2(\frac{q_x}{2}) + \cos^2(\frac{q_y}{2})}}. \quad (9)$$

Our dynamical phonon model is specifically tailored to obtain the eigendisplacements for the B_{1g} vibrations. We finally, provide the variation of the global maximum of the momentum-dependent $el-ph$ coupling $|\tilde{g}(\mathbf{q}; \mathbf{k})|^2$ in Supplementary Figure 4

SUPPLEMENTARY NOTE 3: 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 $\xi_{\mathbf{q}}$ follows from (Eq. (4) in the main text) as

$$H_{\text{MF}} = \underbrace{\sum_{\mathbf{k}} \varepsilon_{\mathbf{k}} c_{\mathbf{k}}^\dagger c_{\mathbf{k}}}_{H_0} + \underbrace{\sum_{\mathbf{k}} \sum_{\mathbf{q}=\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*} \gamma \sqrt{\frac{2m\Omega_P}{\hbar}} \tilde{g}(\mathbf{q}; \mathbf{k}) \xi_{\mathbf{q}} c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}}_{H_{\text{EP}}} + \underbrace{\frac{m\Omega_P^2}{2} \sum_{\mathbf{q}=\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*} |\xi_{\mathbf{q}}|^2}_{H_{\text{Ph}}}. \quad (10)$$

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

$$\mathcal{F} = \mathcal{F}_0 + \frac{1}{2} \sum_{\mathbf{q}=\mathbf{q}^*, \bar{\mathbf{q}}^*} m\Omega_P^2 |\xi_{\mathbf{q}}|^2 - \sum_{l=1}^{\infty} U_l, \quad (11)$$

where $\mathcal{F}_0$ 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_l are obtained from the *el-ph* Hamiltonian H_{EP} as (see Supplementary Reference [3] for a detailed discussion about linked cluster expansion)

$$U_l = \frac{(-1)^l}{l\beta} \int_0^\beta d\tau_1 \int_0^\beta d\tau_2 \dots \int_0^\beta d\tau_l \langle T_\tau H_{\text{EP}}(\tau_1) H_{\text{EP}}(\tau_2) \dots H_{\text{EP}}(\tau_l) \rangle_{\text{connected}}. \quad (12)$$

The notation $\langle \dots \rangle_{\text{connected}}$ 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 $\mathbf{q}$ to only fundamental wavevectors $\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*$, we expand the last term in Eq. (11) up to fourth order to obtain the coefficients

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

$$U_4 = \frac{1}{4\beta} \sum_{\mathbf{k}_i} \sum_{\mathbf{q}_i=\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*} \sum_{i\omega_n} \left[\prod_{i=1}^4 \Delta(\mathbf{q}_i) \tilde{g}(\mathbf{q}_i; \mathbf{k}_i) G^0(\mathbf{k}_i + \mathbf{q}_i, i\omega_n) \right] \quad (13b)$$

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

$$H_{\text{EP}} = \sum_{\mathbf{k}} \sum_{\mathbf{q}=\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*} \Delta(\mathbf{q}) \tilde{g}(\mathbf{q}; \mathbf{k}) c_{\mathbf{k}+\mathbf{q}}^\dagger c_{\mathbf{k}}, \quad (14)$$

The quantity $\Delta(\mathbf{q})$ 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 Supplementary Figure 2a,b, which in turn restricts the choices of the phonon scattering momenta $\mathbf{q}_i$'s as

$$\mathbf{k}_1 + \mathbf{q}_1 = \mathbf{k}_2, \mathbf{k}_2 + \mathbf{q}_2 = \mathbf{k}_3, \mathbf{k}_3 + \mathbf{q}_3 = \mathbf{k}_4, \mathbf{k}_4 + \mathbf{q}_4 = \mathbf{k}_1. \quad (15)$$

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

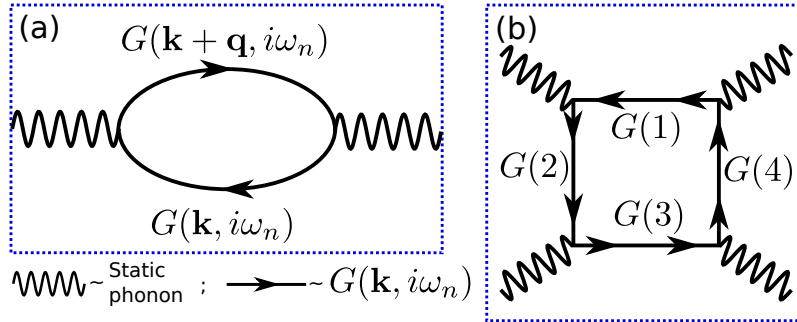
$$\mathbf{q}_1 + \mathbf{q}_2 + \mathbf{q}_3 + \mathbf{q}_4 = 0. \quad (16)$$

Similar constraints on the phonon scattering momenta $\mathbf{q}_i$'s also arise for the odd order linked cluster terms. However, such conditions can only be satisfied for even order terms as $\mathbf{q}_i \in (\pm\mathbf{q}^*, \pm\bar{\mathbf{q}}^*)$ and consequently all the odd order cluster terms are zero.

Performing the frequency summation in Eq. (13), we obtain the fourth-order coefficient $\chi_{\mathbf{q}}^{(4)}$ as (Eq. (7) in the main text)

$$\chi_{\mathbf{q}}^{(4)} = - \sum_{\mathbf{k}} \frac{1}{2} \frac{(\tilde{g}(\mathbf{q}; \mathbf{k}))^4}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})^2} \left(\frac{2\{f(\varepsilon_{\mathbf{k}'} - f(\varepsilon_{\mathbf{k}})\}}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}} + (f'(\varepsilon_{\mathbf{k}}) + f'(\varepsilon_{\mathbf{k}'})) \right) - \sum_{\mathbf{k}} (\tilde{g}(\mathbf{q}; \mathbf{k}))^2 (\tilde{g}(\mathbf{q}; \mathbf{k} + \mathbf{q}))^2. \quad (17)$$

$$\left[\frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}'})}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}''})(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})^2} - \frac{\beta f(\varepsilon_{\mathbf{k}'}) (f(\varepsilon_{\mathbf{k}'} - 1))}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}''})(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'})} - \frac{f(\varepsilon_{\mathbf{k}''}) - f(\varepsilon_{\mathbf{k}'})}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}''})(\varepsilon_{\mathbf{k}''} - \varepsilon_{\mathbf{k}'})^2} + \frac{\beta f(\varepsilon_{\mathbf{k}'}) (f(\varepsilon_{\mathbf{k}'} - 1))}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}''})(\varepsilon_{\mathbf{k}''} - \varepsilon_{\mathbf{k}'})} \right].$$



Supplementary Figure 2: **Feynmann diagrams for the linked cluster expansion.** (a) The second order Feynman diagram for the linked cluster term U_2 as in Eq. 13a. The phonon frequency is set at zero. (b) The fourth order Feynman diagram for the linked cluster term U_4 as in Eq. 13b with static phonons. All possible combinations of the momenta, constrained by the momentum conservation at the vertices, are considered.

The final fourth-order coefficient $\chi_{\mathbf{q}^*; \bar{\mathbf{q}}^*}^{(4)}$ is obtained as follows

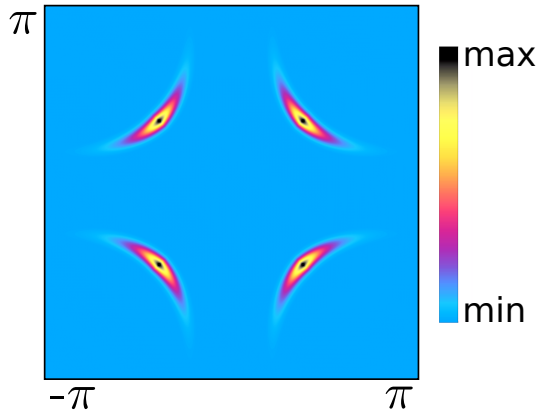
$$\begin{aligned} \chi_{\mathbf{q}^*; \bar{\mathbf{q}}^*}^{(4)} = & - \sum_{\mathbf{k}} (\tilde{g}(\mathbf{q}^*; \mathbf{k}))^2 (\tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k}))^2 \left[\frac{f(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*}) - f(\varepsilon_{\mathbf{k}})}{(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}})^2} - \frac{\beta f(\varepsilon_{\mathbf{k}})(f(\varepsilon_{\mathbf{k}}) - 1)}{(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}})} \right. \\ & - \frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}})}{(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})^2} + \left. \frac{\beta f(\varepsilon_{\mathbf{k}})(f(\varepsilon_{\mathbf{k}}) - 1)}{(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})} \right] - \sum_{\mathbf{k}} (\tilde{g}(\mathbf{q}^*; \mathbf{k}))^2 (\tilde{g}(-\bar{\mathbf{q}}^*; \mathbf{k}))^2 \cdot \\ & \left[\frac{f(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*}) - f(\varepsilon_{\mathbf{k}})}{(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}})^2} - \frac{\beta f(\varepsilon_{\mathbf{k}})(f(\varepsilon_{\mathbf{k}}) - 1)}{(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}})} - \frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}})}{(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})^2} \right. \\ & + \left. \frac{\beta f(\varepsilon_{\mathbf{k}})(f(\varepsilon_{\mathbf{k}}) - 1)}{(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})} \right] - \sum_{\mathbf{k}} (\tilde{g}(\mathbf{q}^*; \mathbf{k}))^2 (\tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k} + \mathbf{q}^*))^2 \left[\frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*})}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})^2} - \right. \\ & \frac{\beta f(\varepsilon_{\mathbf{k}+\mathbf{q}^*})(f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - 1)}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})} - \frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})^2} + \left. \frac{\beta f(\varepsilon_{\mathbf{k}+\mathbf{q}^*})(f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - 1)}{(\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})(\varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*})} \right] \\ & - 2 \sum_{\mathbf{k}} \frac{\tilde{g}(\mathbf{q}^*; \mathbf{k}) \tilde{g}(\mathbf{q}^*; \mathbf{k} + \bar{\mathbf{q}}^*) \tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k} + \mathbf{q}^*) \tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k})}{(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})(\varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*})} \left[\frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*}} - \frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*}} \right. \\ & - \left. \frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*+\bar{\mathbf{q}}^*}} + \frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\bar{\mathbf{q}}^*}} \right] - 2 \sum_{\mathbf{k}} \frac{\tilde{g}(\mathbf{q}^*; \mathbf{k}) \tilde{g}(\mathbf{q}^*; \mathbf{k} - \bar{\mathbf{q}}^*) \tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k} - \mathbf{q}^*) \tilde{g}(\bar{\mathbf{q}}^*; \mathbf{k} + \mathbf{q}^* - \bar{\mathbf{q}}^*)}{(\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}})(\varepsilon_{\mathbf{k}+\mathbf{q}^*-\bar{\mathbf{q}}^*} - \varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*})} \\ & \left[\frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*-\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}+\mathbf{q}^*-\bar{\mathbf{q}}^*}} - \frac{f(\varepsilon_{\mathbf{k}+\mathbf{q}^*}) - f(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}+\mathbf{q}^*} - \varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*}} - \frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}+\mathbf{q}^*-\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}+\mathbf{q}^*-\bar{\mathbf{q}}^*}} + \frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*})}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}-\bar{\mathbf{q}}^*}} \right]. \end{aligned} \quad (18)$$

The fourth-order coefficient $\chi_{\mathbf{q}^*; \bar{\mathbf{q}}^*}^{(4)}$ 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.

SUPPLEMENTARY NOTE 4: 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 (see also Supplementary Reference [4])

$$A(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im}[G(\mathbf{k}, \omega)] = Z_{\mathbf{k}} \delta(\omega - \varepsilon_{\mathbf{k}}) + \frac{1 - Z_{\mathbf{k}}}{2W} F(\omega), \quad (19)$$



Supplementary Figure 3: **A pictorial illustration of the quasiparticle weight distribution of the Fermi surface.** The renormalized quasiparticle weight ansatz $Z_{\mathbf{k}}$ along the Fermi surface at 10 % doping. The nodal points are maximally weighted whereas at the Brillouin zone faces the weight drops to zero.

where the last term is our ansatz for $\text{Im}[G_{\text{inc}}]$. A pictorial illustration of $Z_{\mathbf{k}}$ is provided in Supplementary Figure 3. W in Eq. 19 is the band-width of the anti-bonding band. The function $F(\omega)$ is defined as

$$F(\omega) = \begin{cases} 1; & -W \leq \omega \leq W \\ 0; & \text{else} \end{cases} \quad (20)$$

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

$$\chi_{\mathbf{q}}^{\text{R}} = -2 \sum_{\mathbf{k}} |\mathbf{g}(\mathbf{q}; \mathbf{k})|^2 \left[Z_{\mathbf{k}} Z_{\mathbf{k}'} \frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}'})}{i\omega_m + \varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}} + \frac{Z_{\mathbf{k}}(1 - Z_{\mathbf{k}'})}{2W} \int_{-W}^W d\omega' \frac{f(\varepsilon_{\mathbf{k}}) - f(\omega')}{i\omega_m + \varepsilon_{\mathbf{k}} - \omega'} \right. \\ \left. + \frac{Z_{\mathbf{k}'}(1 - Z_{\mathbf{k}})}{2W} \int_{-W}^W d\omega \frac{f(\omega) - f(\varepsilon_{\mathbf{k}'})}{i\omega_m + \omega - \varepsilon_{\mathbf{k}'}} + \frac{(1 - Z_{\mathbf{k}'})(1 - Z_{\mathbf{k}})}{4W^2} \int_{-W}^W d\omega' d\omega \frac{f(\omega) - f(\omega')}{i\omega_m + \omega - \omega'} \right]_{i\omega_m \mapsto \epsilon(=0) + i\delta}. \quad (21)$$

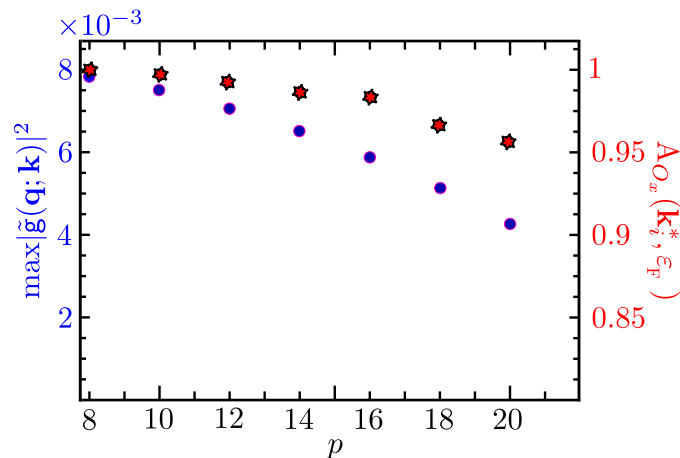
We now define the renormalized charge susceptibility $\chi_{\mathbf{q}}^{(\text{gZ})}$ as following

$$\chi_{\mathbf{q}}^{(\text{gZ})} = -2\gamma^2 \sum_{\mathbf{k}} Z_{\mathbf{k}} Z_{\mathbf{k}'} |\tilde{\mathbf{g}}(\mathbf{q}; \mathbf{k})|^2 \left[\frac{f(\varepsilon_{\mathbf{k}}) - f(\varepsilon_{\mathbf{k}'})}{\varepsilon_{\mathbf{k}} - \varepsilon_{\mathbf{k}'}} \right], \quad (22)$$

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.

SUPPLEMENTARY NOTE 5: Doping dependence of the *el-ph* coupling

In this section, we provide quantitative support for our reasoning on the expected dome-shaped region of the CDW in the phase diagram. First, we notice that the relevant initial electron momentum $\mathbf{k}_i^*$, for which the *el-ph* coupling $\mathbf{g}(\mathbf{q}, \mathbf{k}_i^*)$ attains its global maximum at $\mathbf{q}^*$, varies by almost 5% with increasing the hole doping concentration (see Fig. 3 in the main text) – it moves closer to the nodal points on the Fermi surface. Additionally, the oxygen orbital content on the Fermi surfaces drops with hole doping as shown in Supplementary Figure 4. The combined effects of the oxygen orbital content and phonon eigenvectors (see Eq. 5 in the main text) leads to an almost 50% decrease of the squared *el-ph* matrix element $|\mathbf{g}(\mathbf{q}^*; \mathbf{k}_i^*)|^2$ between $p = 8\%$ and $p = 20\%$.



Supplementary Figure 4: **Doping dependence of the electron-phonon coupling and oxygen orbital content.** The variation of the global maximum of the dimensionless *el-ph* coupling $|\tilde{\mathbf{g}}(\mathbf{q}; \mathbf{k})|^2$ (filled circles) and the maximum oxygen spectral weight on the Fermi surface at doping dependent initial momentum $\mathbf{k}_i^*$ (filled diamonds) as a function of hole doping concentration p . Please note that we have normalized the maximum oxygen spectral weight at 8% hole doping to 1, in comparison to the units for Fig. 2d in the main text.

SUPPLEMENTARY REFERENCES

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