

Density Wave Probes Cuprate Quantum Phase Transition

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In cuprates, the strong correlations in proximity to the antiferromagnetic Mott insulating state give rise to an array of unconventional phenomena beyond high-temperature superconductivity. Developing a complete description of the ground-state evolution is crucial to decoding the complex phase diagram. Here we use the structure of broken translational symmetry, namely, d -form factor charge modulations in $(\text{Bi, Pb})_2(\text{Sr, La})_2\text{CuO}_{6+\delta}$ as a probe of the ground-state reorganization that occurs at the transition from truncated Fermi arcs to a large Fermi surface. We use real space imaging of nanoscale electronic inhomogeneity as a tool to access a range of dopings within each sample, and we definitively validate the spectral gap Δ as a proxy for local hole doping. From the Δ dependence of the charge modulation wave vector, we discover a commensurate-to-incommensurate transition that is coincident with the Fermi-surface transition from arcs to large hole pocket, demonstrating the qualitatively distinct nature of the electronic correlations governing the two sides of this quantum phase transition. Furthermore, the doping dependence of the incommensurate wave vector on the overdoped side is at odds with a simple Fermi-surface-driven instability.

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I. INTRODUCTION

In cuprates, high-temperature superconductivity lies between an undoped antiferromagnetic (AFM) insulator and a metal at high hole doping (p). In proximity to the AFM insulator, the strong electronic correlations give rise to a complex phenomenology, including a large spectral gap Δ that opens above T_c and a k -space structure lacking a conventional Fermi surface (FS) but described by open arcs

[1,2]. Both gap and arcs are widely considered hallmarks of this underdoped region of the phase diagram and have drawn significant attention aiming to uncover their origin(s) [3]. However, at a doping near optimal superconductivity, the Fermi arcs undergo an abrupt transition to a “large” pocket consistent with a conventional area proportional to $1 + p$ [4–7]. A crucial challenge remains to identify the appropriate ground state(s) that underlie the theoretical framework on *both* sides of this transition.

On the overdoped side, long thought to be a Fermi liquid, several recent reports of anomalous behavior call into question the conventional interpretation [4,8–12]. The observations of resistivity linear in temperature [8,9] in Bi-based and La-based families challenge the expectations of standard Fermi-liquid theory, and in MBE-grown $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ compounds, there are reports of mysterious symmetry breaking [11] and anomalous scaling of the superfluid density with critical temperature [10,12], although the latter finding remains controversial [13,14]. Furthermore, resonant inelastic x-ray experiments on Tl-, Y- [15], and La-based [16] compounds revealing spin fluctuations have been interpreted in terms of significant electron correlations, and there are now theoretical proposals [17,18] for how overdoped compounds may retain certain characteristic features of Fermi-liquid-like behavior, while exhibiting

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fractionalization in the presence of strong stripe fluctuations. Additional experiments are necessary to understand the overdoped compounds, and in particular to clarify the extent to which the effects of strong correlations may persist through the FS transition.

In nearly all cuprate families, charge order in the form of disordered charge modulations have been reported in underdoped compounds, with detection terminating at [19] or before [6,20] the doping where the FS transition occurs. In $(\text{Bi, Pb})_2(\text{Sr, La})_2\text{CuO}_{6+\delta}$, Bi2201, however,

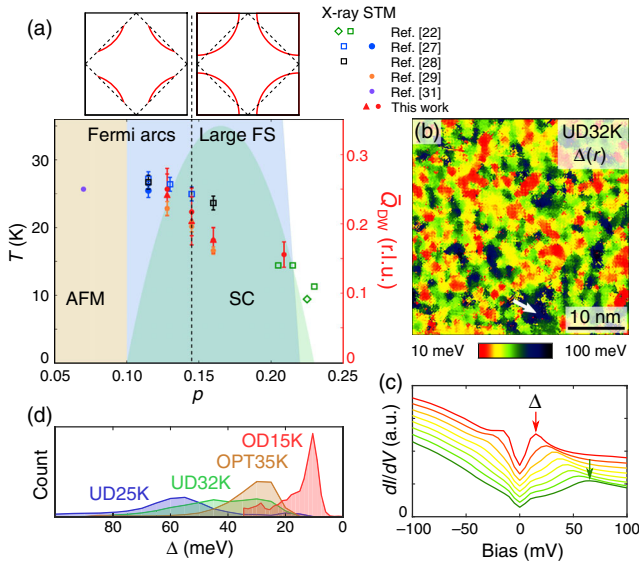


FIG. 1. Using local electronic inhomogeneity and charge modulations to probe the Bi2201 phase diagram. (a) Phase diagram for Bi2201 in the region of the superconducting dome (green shading). With increasing p at low temperature, the appearance of sharp antinodal quasiparticles (insets) indicative of a large normal-state Fermi surface occurs just below optimal doping (dashed line [21]), while the spectral gap persists into overdoped compounds (blue shading, with boundary marking the closing of the gap, as measured by ARPES [25] and NMR [26]). The data points mark existing sample-average measurements of the charge modulation wave vector \bar{Q}_{DW} in Bi2201 from x-ray scattering (green [22], blue [27], and black [28] open symbols) and STM (blue [27], orange [29], and purple [31] filled symbols). Red triangles and circles are the sample-average measurements of the d -form-factor charge modulations in the x and y directions, respectively, from this work, with p determined from Ando’s conversion [32], as described in the Supplemental Material [33]. The green squares and diamond are the annealed and as-grown samples, respectively, from Peng *et al.* [22]. (b) The local spectral gap Δ (shown for UD32K) measured as the local minimum in the second derivative of the empty-state differential conductance spectrum measured at each point. The white arrow marks the same location as in Fig. 2(h) to highlight a region of large Δ contributing to the Fermi-arc QPI in Fig. 2(f). (c) Differential conductance spectra from UD32K averaged over spatial regions binned by Δ and offset vertically for clarity. (d) Distributions of Δ within the UD25K, UD32K, OPT35K, and OD15K samples. The histograms are normalized to have equal areas.

charge modulations extend into the overdoped regime [21,22]. These modulations reflecting an ordering instability of the electronic system therefore serve as a doping-dependent fingerprint of underlying electronic interactions, not just in the underdoped regime, but across the FS transition [Fig. 1(a)]. In the Bi2201 phase diagram, the FS transition occurs just below optimal doping [23], while the spectral gap persists in the presence of the large FS [21,25,26]. Peng *et al.* [22] recently investigated the charge modulation structure across the closing of the gap in the far-overdoped region, but it remains crucial to clearly define how the wave vector \bar{Q}_{DW} evolves across the FS transition [Fig. 1(a)]. Existing measurements in this doping range [27–29] exhibit large scatter and doping coverage insufficient to clearly establish the \bar{Q}_{DW} trend on the underdoped side. Furthermore, with the exception of Ref. [30], the measurements did not take into account the electronic inhomogeneity within these samples, even though the influence of annealing suggests that the bulk-averaged \bar{Q}_{DW} is sensitive to disorder [22]. Specifically, examining \bar{Q}_{DW} in Fig. 1(a), it is unclear (1) if a single doping-dependent incommensurate trend should be drawn through all of the measurements from $p = 0.11$ to 0.23 and (2) how the charge modulation structure in this range relates to the commensurate modulation observed in a lightly doped compound near the insulating state [31].

Here, we use the spatial dependence of the Bi2201 density wave (DW) as a probe of the parent states in both Fermi-arc and large FS regions. We find that the FS transition marks the boundary between two distinct ground states that give rise to commensurate and incommensurate charge modulations, respectively. Furthermore, concurrent mapping of the DW and FS demonstrates that conventional Fermiology is insufficient to explain the overdoped evolution of \bar{Q}_{DW} .

II. INHOMOGENEOUS FERMI-SURFACE TRANSITION

Within a single Bi-based cuprate crystal, an average doping of p holes per unit cell produces a highly inhomogeneous spatial distribution resulting in large variations in the local electronic properties [30,34–39]. In a scanning tunneling microscope, the local electronic density of states is typically measured by the spatially resolved differential conductance $g(\mathbf{r}, E = eV) \equiv dI/dV(\mathbf{r}, V)$, where V is the sample bias and $I(\mathbf{r}, V)$ is the tunneling current. Binning and averaging the local spectra by gap size $\Delta(\mathbf{r})$, as shown in Figs. 1(b) and 1(c), demonstrates the variation of the spectrum over a large field of view. Previous work has shown that smaller Δ corresponds to higher hole concentration, both locally within each sample [34,36,37] and globally from sample to sample [40]. Thus, the same spectrum can be found locally in samples with different global p , and the overlapping Δ distributions from the four samples studied in this work, UD25K, UD32K, OPT35K,

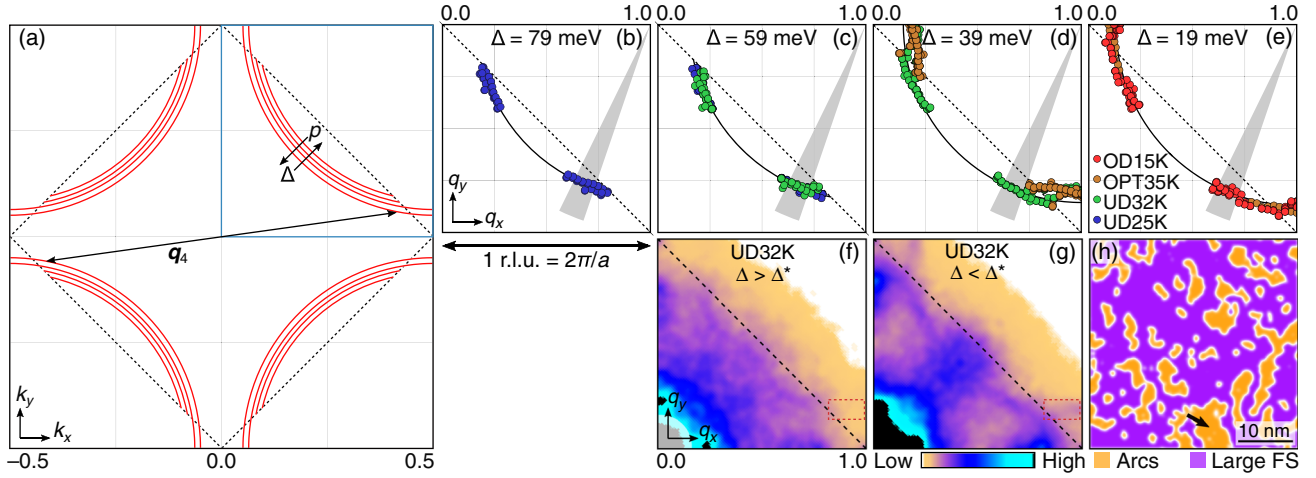


FIG. 2. A continuous doping axis from local electronic inhomogeneity. (a) Fermi-surface evolution with local doping over the range of Δ and p studied here. The double-headed arrow indicates an example \mathbf{q}_4 scattering vector. The blue box marks the quadrant that is shown for the QPI panels. (b)–(e) The QPI evolution with the local gap is shown by data points marking the peak positions extracted from gap-masked $Z(\mathbf{q})$ for all four samples. Refer to Appendix C and the Supplemental Material [33] Fig. S1. On each panel, data are compiled from masked regions with average Δ within a 4 meV range, centered at the indicated value. The black lines are circles determined by the average radius of the near-nodal data within a fixed angular range indicated by gray shading. For $\Delta > \Delta^*$, points are extracted only for regions inside the dashed line, as the intensity of antinodal QPI is negligible. (f) UD32K QPI from masking $Z(\mathbf{r})$ by the yellow regions in (h) with Δ primarily greater than Δ^* . QPI from near-nodal quasiparticles is strong, but antinodal QPI (red dashed box) is not visible. (g) UD32K QPI from masking $Z(\mathbf{r})$ by the purple regions in (h) with Δ primarily less than Δ^* . QPI extends out to $q_{x,y} = 1$ r.l.u. (red dashed box), indicating the presence of antinodal quasiparticles arising out of the antinodal normal-state Fermi surface. The gap-masked $Z(\mathbf{q})$ in (f) and (g) is integrated from 10 to 25 meV, fourfold symmetrized, and smoothed with a Gaussian filter of width 0.015 r.l.u. to reduce the appearance of noise. (h) Spatial division of UD32K into regions with Δ primarily greater than (yellow) or less than (purple) Δ^* (Appendix D). The arrow in (h) marks the same location as in Fig. 1(b) to highlight a region of large Δ contributing to the Fermi-arc QPI. All \mathbf{k} and \mathbf{q} axes are in reciprocal lattice units ($1 \text{ r.l.u.} = 2\pi/a$).

and OD15K [Fig. 1(d)], allow us to move continuously from underdoped (UD) to overdoped (OD) in the phase diagram using spatial masking [30] to hone in on a single local doping within a larger field of view (Appendix B).

To validate the use of local doping to construct the Bi2201 phase diagram, we demonstrate that the evolution of the FS with Δ mimics that of bulk samples with p . We calculate the ratio map $Z(\mathbf{r}, E) \equiv g(\mathbf{r}, E)/g(\mathbf{r}, -E)$, which enhances Bogoliubov quasiparticle interference (QPI), and eliminates artifacts associated with the tip-sample junction setup [41]. The normal-state FS can be inferred from QPI in the superconducting state as follows [42–44]. The QPI signal is dominated by wave vectors connecting regions of high density of states, i.e., extrema in the Bogoliubov dispersion $E(k) = \pm \sqrt{\epsilon_k^2 + |\Delta_k|^2}$, where ϵ_k is the normal-state band dispersion, and Δ_k is the momentum-dependent superconducting gap. For a given angle, extrema in $E(k)$ are given by $\epsilon_k = E_F$ (normal-state Fermi energy); thus, the Bogoliubov QPI dispersion traces out the normal-state FS. In particular, it is well established that the $\mathbf{q}_4(E)$ channel [Fig. 2(a)] traces out $2k_F$ [19,21,44], in excellent agreement with the normal-state FS measured by ARPES [44,45]. The \mathbf{q}_4 wave vectors—extracted as a function of Δ by selecting a range of Δ values in $\Delta(\mathbf{r})$ to mask the $Z(\mathbf{r})$ data (Appendix B)—together describe a single evolution of the momentum-space electronic structure extending across

all samples [Figs. 2(a)–2(e)]. Regions with a small gap [$\Delta = 19$ meV, Fig. 2(e)] exhibit QPI tracing out a large normal-state FS: The Bogoliubov quasiparticles near the antinodes at the edge of the Brillouin zone $k_{x,y} = \pm\pi/a$ [Fig. 2(a)] generate scattering with \mathbf{q}_4 wave vectors that extend out to $q_{x,y} = \pm 2\pi/a$. Moving to larger Δ [Fig. 2(d)], the \mathbf{q}_4 trajectory shrinks, consistent with decreasing hole concentration, and the full evolution of the FS size inferred from the QPI (Fig. S3 in the Supplemental Material [33]) confirms that Δ is well correlated to doping, in agreement with previous observations [30,34,38].

For larger Δ , the absence of observable QPI near $q_{x,y} = \pm 2\pi/a$ (Fig. S1 in the Supplemental Material [33]) is consistent with a normal-state Fermi arc that lacks sharp antinodal (AN) quasiparticles [19,21]. Quantitatively, the intensity of the AN QPI in the UD32K sample [Fig. 4(a)] decreases with increasing Δ before settling at a constant value indistinguishable from the background, indicating the transition occurs at $\Delta \approx 50$ meV, which we label Δ^* . Spatially dividing the data into regions of $\Delta > \Delta^*$ and $< \Delta^*$ [Figs. 2(f) and 2(g)] shows that QPI associated with a large FS and Fermi arcs both exist within the sample, such that one can move on the phase diagram from one side of the transition to the other spatially. This spatial division emphasizes that the electronic structure is determined on length scales similar to the Δ correlation length. The FS p

evolution of the cuprate phase diagram is therefore reproduced locally as a function of Δ , with the transition from Fermi arcs to large FS occurring at $\Delta^* \approx 50$ meV.

III. COMMENSURATE-TO-INCOMMENSURATE TRANSITION

We now determine the doping dependence of the charge modulation wave vector Q_{DW} as a function of Δ to look for signatures of the change in ground state at Δ^* . We examine $D(\mathbf{r})$, the d -form-factor (d FF) component [46] of $\sum_{0 < \varepsilon < E} g(\mathbf{r}, \varepsilon) / \sum_{-E < \varepsilon < 0} g(\mathbf{r}, \varepsilon) \sim I(\mathbf{r}, E) / I(\mathbf{r}, -E)$, where the integration over an energy range larger than the typical Δ for each sample, enhances the DW signal [47]. In all four samples, the amplitude of the Fourier transform $D(\mathbf{q})$ has broad peaks at $(\pm Q_{\text{DW}}, 0)$ and $(0, \pm Q_{\text{DW}})$ near the charge modulation wave vectors that have been observed by previous experiments, indicating clearly that a d FF DW exists in all (Fig. S5 in the Supplemental Material [33]). We employ the demodulation phase residue minimization technique of Mesáros *et al.* [48] to make Q_{DW} measurements robust against the strong disorder apparent from the broad shape of the DW peaks.

Comparing Figs. 3(a)–3(c) to Figs. 3(d)–3(f) reveals that the d FF DW exhibits distinct evolutions for small and large spectral gap regions. From 16 to 48 meV, Q_{DW} increases

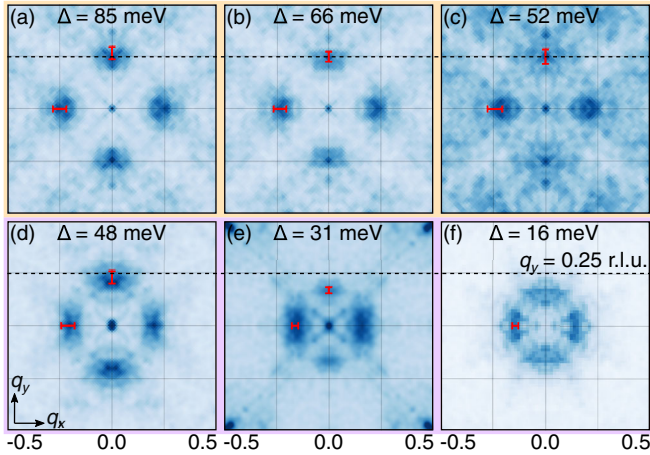


FIG. 3. d -form-factor charge modulations. Examples of gap-masked $D(\mathbf{q})$ where the indicated Δ is the average value within each masked area. The data are symmetrized along the q_x and q_y directions, and Gaussian smoothed with a 0.01 r.l.u. width. (a)–(c) Data with $\Delta > \Delta^*$ (Fermi-arc regime). (d)–(f) Data with $\Delta < \Delta^*$ (large FS regime). The thin gray lines have a spacing of 0.25 r.l.u. to facilitate comparison to the Fermiology in Fig. 2. The red symbols mark Q_{DW} determined for x and y modulation directions, where the bar length indicates \pm the estimated standard deviation of spatial fluctuations within each masked area. No measurement of Q_{DW} is made for q_y modulations in OD15K (f), as the wave vector is not sufficiently well defined. The panels correspond to data from UD25K (a)–(c), UD32K (d), OPT35K (e), and OD15K (f).

from 0.15 to 0.23 r.l.u., matching the change in wave vector that has been measured by resonant x-ray techniques from $p \approx 0.20$ to near-optimal doping [22,27,28] and consistent with an evolving incommensurate wave vector. However, from $\Delta \approx 50$ meV to $\Delta \approx 85$ meV, no significant increase is observed [Figs. 3(a)–3(c)]. The constant value of Q_{DW} near 0.25 r.l.u. suggests a dominant commensurate instability. In fact, the entire $Q_{\text{DW}}(\Delta)$ evolution [Fig. 4(b)] is consistent with a commensurate-to-incommensurate transition at a location indistinguishable from Δ^* . The dashed line showing the expected broadening of the underlying commensurate- (yellow) to-incommensurate (purple) trend describes the data accurately (Supplemental Material [33] Sec. SX). Furthermore, the coincident changes in Fermiology and DW commensurability strongly suggest the presence of a quantum phase transition at Δ^* .

A. Commensurate density wave

The observed wave vector in the Fermi-arc state ($\Delta > \Delta^*$) is consistent with a commensurate four-unit-cell charge modulation. The average \bar{Q}_{DW} from UD25K, which lies almost entirely on the underdoped side of the transition, is 0.25(3) and 0.24(3) r.l.u. for the x and y directions, respectively, where the errors represent the estimated standard deviation of spatial variations across the entire field of view. Resonant x-ray experiments [27] have reported a doping-dependent \bar{Q}_{DW} in this same doping range, down to $p = 0.115$. However, this apparent discrepancy can be understood by considering (1) the FS transition occurs near $p = 0.14$ [21], and beyond this point, we also observe an incommensurate wave vector, (2) the local inhomogeneity could plausibly induce a doping dependence of \bar{Q}_{DW} even in samples with average p below the transition, and (3) the strongly disordered structure of charge modulations limits the precision with which the value of Q_{DW} (and \bar{Q}_{DW}) can be determined unambiguously [48]. For the samples studied here, ignoring local doping variations and taking the average \bar{Q}_{DW} value for each sample hides the kink at the FS transition and produces a trend of decreasing \bar{Q}_{DW} with doping similar to previous reports [Fig. 1(a)]. Furthermore, while our data do not rule out a small doping dependence, recent experiments on Bi2212 [48–50] and very underdoped Bi2201 [31] are also consistent with a $Q = 0.25$ r.l.u. commensurate DW within the Fermi-arc regime arising from proximity to the Mott insulating state.

B. Incommensurate density wave

What is the mechanism for the incommensurate Q_{DW} observed for $\Delta < \Delta^*$? Previous work interpreted the monotonically decreasing Q_{DW} as evidence of a FS instability that follows the growing FS hole pocket [30]. In this picture, there are two natural candidates for Q_{DW} : (1) Q_{AN} that connects nested antinodal segments of the FS and (2) Q_{AFZB} that connects the points where the FS crosses the antiferromagnetic zone boundary (AFZB), the hot spots

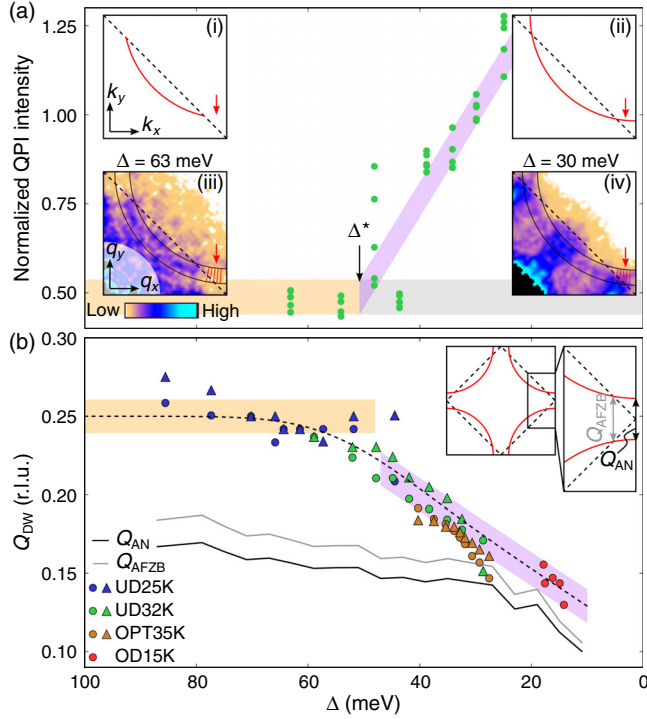


FIG. 4. Simultaneous DW and FS transitions. (a) Intensity of AN QPI in UD32K determined at five locations [red lines in the insets (iii),(iv)] and normalized to the intensity of near-nodal QPI (Appendix E). Δ^* indicates the approximate location where the AN QPI becomes indistinguishable from the background noise. The thick shaded lines are guides to the eye. Insets (i),(ii) show the approximate Fermi-surface structure inferred from two examples of the gap-masked $Z(\mathbf{q})$ (iii),(iv) without and with AN QPI. (b) Wave vector (Q_{DW}) of the $d\text{FF}$ charge modulations in the x (triangles) and y (circles) directions, extracted from gap-masked $D(\mathbf{r})$. Refer to Fig. S9 in the Supplemental Material [33] for the standard deviation of Q_{DW} for each value of Δ . The thick shaded lines indicate the commensurate-to-incommensurate trend underlying the dashed line, which includes the expected effect of Gaussian smoothing ($\sigma = 12$ meV) in Δ due to the resolution of the masking technique (Fig. S10 in the Supplemental Material [33]). The gray and black lines indicate the Fermiology-driven candidate wave vectors, as indicated schematically in the insets and based on the circular Fermi-surface models shown in Figs. 2(a)–2(e).

for (π, π) spin fluctuations. Both $Q_{\text{AN}}(\Delta)$ and $Q_{\text{AFZB}}(\Delta)$ are shown in Fig. 4(b). For the largest p (smallest Δ), Q_{DW} becomes similar to these Fermiology-derived wave vectors. However, upon decreasing p towards the transition, Q_{DW} grows more rapidly than the FS evolves. This unexpected discrepancy between Q_{DW} and Fermiology constitutes our second major finding.

IV. DISCUSSION

Whereas most recent theories for charge modulations in the cuprates have aimed to explain an incommensurate DW in the presence of Fermi arcs, here we discuss (1) a

commensurate DW in the presence of Fermi arcs and (2) an incommensurate DW occurring in the presence of the large FS. This leads to two important distinctions. First, in the Fermi-arc regime, strong interactions are expected, and the associated renormalization can affect a hot-spot (HS) wave vector, Q_{HS} . For an instability of the large FS, however, it is not *a priori* clear that there should be any influence of correlations on such a Q_{HS} . This first distinction reconciles our conclusion with Ref. [27]’s interpretation that \bar{Q}_{DW} is Fermiology driven [51]. We note that additional factors, such as coupling to the lattice, may affect the observed Q_{DW} . Second, theoretical studies have found that the dominant charge-density wave (CDW) instability of the large FS in the presence of exchange interactions has a wave vector along the $(\pm q, \pm q)$ direction rather than $(0, \pm q)$ or $(\pm q, 0)$, and the presence or absence of antinodal states is important in stabilizing the former or latter orientation, respectively [52]. It is therefore significant that the orientation of the charge modulations does not change at Δ^* .

To search for information about the nature of the transition at Δ^* , we consider that generically in CDW systems, a discontinuity or sharp jump in Q_{DW} occurs at a commensurate-to-incommensurate transition [53]. From this work, we cannot distinguish between a continuous Q_{DW} or one with a small jump, as shown by the yellow and purple trends in Fig. 4(b) [see also Fig. S10(j) in the Supplemental Material [33]]. To extract this information, Δ^* and the incommensurate $Q_{\text{DW}}(\Delta)$ would need to be determined with reduced uncertainties. However, if present, a continuous Q_{DW} , which generically requires fine-tuning, would imply a mechanism-derived constraint on the position of Δ^* not explained by existing theoretical models of the Fermi-surface transition.

Ubiquity across underdoped compounds has widely been cited to motivate studying charge modulations as a route to understanding Fermi-arc physics and the mechanism behind high- T_c superconductivity in cuprates. However, the relationship among these three phenomena has remained an open question. The coincidence of the DW and FS transitions observed here establishes an intimate link between the DW and the presence of Fermi arcs, and furthermore suggests that the same interactions which generate the commensurate instability may also be responsible for the arc phenomenology.

In summary, we report three concrete observations: (1) There is a commensurate-to-incommensurate transition of Q_{DW} at a doping consistent with that of the FS transition, (2) on the underdoped side, Q_{DW} is consistent with a commensurate four-unit-cell modulation, and (3) on the overdoped side, the doping dependence of Q_{DW} is stronger than that of the FS size.

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APPENDIX A: STM DATA

Differential conductance maps were collected in scanning tunneling microscopes at 6 K with the following tip-sample junction setup conditions: 100 mV and 100 pA for OD15K, -100 mV and 400 pA for OPT35K, -200 mV and 400 pA for UD32K, and -150 mV and 400 pA for UD25K. The measurements used a lock-in technique with 2, 10, 5, and 5 mV bias modulations, respectively. Data were corrected for artificial distortions due to instrument drift to register the simultaneously recorded topography to a perfect lattice [54].

APPENDIX B: GAP-MASKING TECHNIQUE

The gap map $\Delta(\mathbf{r})$ is calculated by finding the position of the local minimum in the second derivative of the empty-state differential conductance for the spectrum at each pixel \mathbf{r} , in $g(\mathbf{r}, E)$, where Gaussian smoothing in energy reduces errors from noise in the data. The gap masks are then generated by dividing the values of Δ into bins with equal counts. The mask for bin b , $M_b(\mathbf{r})$ has a value of 1 if $\Delta(\mathbf{r})$ is in b , or zero otherwise. In order to reduce periodic structure in the masks arising from the atomic corrugation or the charge modulations [30], a bilateral filter is applied to $\Delta(\mathbf{r})$ before generating the masks.

To obtain clear images in momentum transfer \mathbf{q} space, we use an additive masking technique, where for each bin b , we look for the change in Fourier-transform amplitude when adding b into the field of view. The additive masks therefore include bins summed up to a bin b : $M_{1,b}(\mathbf{r}) = \sum_{j=1}^b M_j(\mathbf{r})$. To avoid introducing artifacts from spatial structure of the masks, we apply Gaussian smoothing to the mask, with a spatial resolution of $1/w$. The filtered masks $\mathcal{M}_{1,b}(\mathbf{r}) = G_w(\mathbf{r}) * M_{1,b}(\mathbf{r})$ can now have any value in the range $[0,1]$, where G_w is the Gaussian filter, and $*$ indicates convolution. This filtering imposes a spatial resolution of the masks and implies a spread in the Δ distribution within each mask (Fig. S10 in the Supplemental Material [33]).

To explain the additive masking technique, we describe how the images in Fig. 3 are generated. The additive masks are applied to the $D(\mathbf{r})$ map in real space: $D_{a,b}(\mathbf{r}) = \mathcal{M}_{a,b}(\mathbf{r})D(\mathbf{r})$. To visualize the charge order peaks from bin b , we take the difference of the absolute values of the Fourier transforms, including and not including bin b . The additive masking can either add up Δ values

from small to large (forward, fw) or in the reverse direction (backward, bw). For these two cases, the gap-masked $D(\mathbf{q})$ is

$$D_b^{\text{fw}}(\mathbf{q}) = D_{1,b}(\mathbf{q}) - D_{1,b-1}(\mathbf{q}), \quad (\text{B1})$$

$$D_b^{\text{bw}}(\mathbf{q}) = D_{b,N}(\mathbf{q}) - D_{b+1,N}(\mathbf{q}), \quad (\text{B2})$$

where N is the total number of bins, and $D_{a,b}(\mathbf{q})$ is a real number, the amplitude of the Fourier transform. Unless otherwise specified, functions of \mathbf{q} refer to amplitudes of Fourier transforms. The panels of Fig. 3 have Gaussian smoothing applied with width 0.01 r.l.u. to reduce the appearance of noise.

The QPI shown in the insets of Fig. 4 are $Z_b^{\text{fw}}(\mathbf{q})$ generated from $Z(\mathbf{r})$ using this same technique. Additional gap-masked QPI from UD32K in Fig. S1 in the Supplemental Material [33] shows the evolution of the Fermi-surface structure across the full range of Δ within the sample. These QPI images have Gaussian smoothing applied with width 0.015 r.l.u. and are fourfold symmetrized.

For the analysis of the Fermi-surface structure, we use overlapping bins, i.e.,

$$Z_b^{\text{fw}}(\mathbf{q}) = Z_{1,b}(\mathbf{q}) - Z_{1,b-n}(\mathbf{q}), \quad (\text{B3})$$

$$Z_b^{\text{bw}}(\mathbf{q}) = Z_{b,N}(\mathbf{q}) - Z_{b+n,N}(\mathbf{q}) \quad (\text{B4})$$

in order to measure the QPI wave vectors at finer-spaced intervals of Δ with more variations in the masks. The integer n determines the overlap between subsequent bins.

The average value of the gap for bin b is then

$$\Delta_b^{\text{fw}} = \frac{\sum_{\mathbf{r}} \Delta(\mathbf{r}) [\mathcal{M}_{1,b}(\mathbf{r}) - \mathcal{M}_{1,b-n}(\mathbf{r})]}{\sum_{\mathbf{r}} [\mathcal{M}_{1,b}(\mathbf{r}) - \mathcal{M}_{1,b-n}(\mathbf{r})]}, \quad (\text{B5})$$

$$\Delta_b^{\text{bw}} = \frac{\sum_{\mathbf{r}} \Delta(\mathbf{r}) [\mathcal{M}_{b,N}(\mathbf{r}) - \mathcal{M}_{b+n,N}(\mathbf{r})]}{\sum_{\mathbf{r}} [\mathcal{M}_{b,N}(\mathbf{r}) - \mathcal{M}_{b+n,N}(\mathbf{r})]}, \quad (\text{B6})$$

where $n = 1$ for distinct as opposed to overlapping bins.

Here, we use $N = 19$ for UD25K, 31 for UD32K, 31 for OPT35K, and 21 for OD15K, with $n=4$ and $w = 0.20$ r.l.u.

For $D(\mathbf{q})$ in Fig. 3, we use $N = 9$ with $n = 1$ for UD25K, UD32K, and OPT35K, and $N = 21$ with $n = 4$ for OD15K. For OD15K, only the data for bins $b = 5, 9, 13, 17,$ and 21 are plotted in Fig. 4(b) to present data from distinct bins, as are used on the other three samples. The mask smoothing parameter w is chosen to include the resolution Λ of determining Q_{DW} (Appendix F): $w = \Lambda = 0.10$ r.l.u. for UD25K, $w = \Lambda = 0.10$ r.l.u. for UD32K, $w = \Lambda = 0.04$ r.l.u. for OPT35K, and $w = (\Lambda^{-2} + 0.20 \text{ r.l.u.}^{-2})^{-1/2}$ with $\Lambda = 0.04$ r.l.u. for OD15K.

APPENDIX C: FERMI-SURFACE STRUCTURE FROM GAP-MASKED QPI

$Z(\mathbf{q})$ is integrated over the low-energy layers to capture the full dispersion of the Bogoliubov quasiparticles from 1.5 to 9 meV for OD15K, 5 to 5 meV for OPT35K, 10 to 25 meV for UD32K, and 5 to 15 meV for UD25K.

For each Δ bin, the QPI wave vectors, as shown in Fig. 2, are extracted from the positions of peaks in one-dimensional cuts through $Z_b^{fw}(\mathbf{q})$ and $Z_b^{bw}(\mathbf{q})$ and are shown in Fig. S1 of the Supplemental Material [33] for UD32K. To quantitatively determine the size of the Fermi surface, the wave vectors from all samples are binned together, as shown in Fig. 2. A circular hole pocket is determined from the average radius of the data in the range $\theta = 0.105\pi$ to 0.145π , where θ is defined in Fig. S1 of the Supplemental Material [33], and the range is selected because this near-nodal QPI is consistently measured across the Fermi-surface transition (unlike the antinodal QPI) and is least influenced by nearby scattering channels or the DW signal. The evolution of the Fermi-surface radius is shown in Fig. S3 of the Supplemental Material [33] and explicitly demonstrates that Δ tracks the local doping.

APPENDIX D: FERMI-ARC AND LARGE-FERMI-SURFACE QPI

To generate Figs. 2(f) and 2(g), we locate the bin b^* , which has the largest average gap value below 49 meV, the average of Δ^* estimates based on forward and backward masked data shown in Fig. 4(a) and Fig. S4(b) of the Supplemental Material [33]. Bins $b \leq b^*$ primarily have $\Delta < \Delta^*$, and $b > b^*$ primarily $\Delta > \Delta^*$. This division is only approximate due to the Gaussian smoothing of the masks (Fig. S10 in the Supplemental Material [33]). To probe QPI from the two regimes divided by Δ^* , Figs. 2(f) and 2(g) show $Z_{b^*+1,N}(\mathbf{q})$, and $Z_{1,b^*}(\mathbf{q})$, respectively. Note that in this case, there is no subtraction after applying the Fourier transform. Figure 2(h) shows $\mathcal{M}_{1,b^*}(\mathbf{r})$, where the color scale interpolates between 0.0 (yellow) and 1.0 (purple). Because the masks used to obtain Figs. 2(f) and 2(g) are related by $\mathcal{M}_{b^*+1,N}(\mathbf{q}) = 1 - \mathcal{M}_{1,b^*}(\mathbf{q})$, the yellow and purple indicate the regions primarily contributing to the Fermi arc and large-Fermi-surface QPI, respectively.

APPENDIX E: INTENSITY OF AN QPI IN UD32K

In order to quantify the disappearance of the AN QPI, which is apparent directly in the data (Fig. S1 in the Supplemental Material [33]), we measure the intensity at cuts spaced at regular angular intervals, as shown in Fig. S4 of the Supplemental Material [33]. The cuts are averaged over a transverse width of 0.07 r.l.u. and a length determined by the QPI radius (twice the FS radius) ± 0.06 r.l.u. To compare QPI intensities from different bins, the intensities of the cuts are normalized by the average intensity of

five cuts closer to the nodal QPI [dashed red lines in Fig. S4(a) of the Supplemental Material [33]]. Figure 4(a) tracks this normalized intensity for five cuts near the antinode [red lines in Fig. 4(a) insets iii and iv].

APPENDIX F: EXTRACTING Q_{DW} FROM dFF CHARGE MODULATION

We follow the procedure described by Mesaros *et al.* [48], where Q_{DW} is determined as the wave vector which minimizes the demodulation residue $R_{\mathbf{Q}} = \sqrt{|R_{\mathbf{Q}}^x|^2 + |R_{\mathbf{Q}}^y|^2}$ over the field of view. In the case of a strongly disordered density wave, this measurement has a more clearly defined interpretation than fitting peaks of the Fourier-transformed data. Both techniques are compared in Sec. SVIII of the Supplemental Material [33]. A detailed explanation of demodulation residue can be found in Ref. [48]. In this section, we extend the technique for application to masked regions of the data's field of view.

$D(\mathbf{r})$ is demodulated by the reference wave vector \mathbf{Q} in

$$\tilde{\Psi}_{\mathbf{Q}}(\mathbf{q}) = \exp\left(\frac{-q^2}{2\Lambda^2}\right)\tilde{\psi}(\mathbf{q} + \mathbf{Q}), \quad (\text{F1})$$

where $\tilde{\psi}(\mathbf{q}) = \tilde{D}(\mathbf{q})$ over a domain which isolates the charge order peak, \tilde{D} is the complex-valued Fourier transform of $D(\mathbf{r})$, and $\tilde{\Psi}_{\mathbf{Q}}$, $\tilde{\psi}$ are complex-valued functions. The Gaussian cutoff imposes a spatial resolution of $1/\Lambda$.

The Q_{DW} measurement proceeds in each bin b by integrating the residue only over the masked region:

$$R_{b,\mathbf{Q}}^\alpha[\Psi] = \int d^2\mathbf{r} \mathcal{M}_b(\mathbf{r}) \text{Re}[\Psi_{\mathbf{Q}}^*(-i\partial_\alpha)\Psi_{\mathbf{Q}}],$$

$$\sigma^2 = \frac{\int d^2\mathbf{r} \mathcal{M}_b(\mathbf{r}) \sum_\alpha |\Psi_{\mathbf{Q}}^*(-i\partial_\alpha)\Psi_{\mathbf{Q}}|^2}{\int d^2\mathbf{r} \mathcal{M}_b(\mathbf{r}) |\Psi_{\mathbf{Q}}|^2}, \quad (\text{F2})$$

where α is either x or y , $\Psi_{\mathbf{Q}}(\mathbf{r})$ is the inverse Fourier transform of $\tilde{\Psi}_{\mathbf{Q}}(\mathbf{q})$, and with $\mathbf{Q} = \mathbf{Q}_{DW}$, σ estimates the standard deviation of spatial fluctuations in the modulation wave vector.

For this analysis, we use $w = 0.0$ as the mask smoothing parameter for UD25K, UD32K, and OPT35K, with $\Lambda = 0.10, 0.10,$ and 0.04 r.l.u., respectively. For OD15K, we use $w = 0.20$ r.l.u. and $\Lambda = 0.04$ r.l.u. The samples with smaller wave vector require a smaller Λ for Q_{DW} to be robust against the choice of Λ .

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