High-temperature superconductivity (SC) is a great surprise in quantum materials and its mechanism remains a puzzle. In the past 36 years, studies on cuprate superconductors primarily have been focused on the underdoped and optimally doped regions — close to the Mott-insulating state of the phase diagram [1, 2]. It has been a longstanding challenge to understand how the versatile phenomena exhibited in these materials, such as the pseudogap (PG) and strange metal states, together with a plethora of exotic electronic orders, coexist and compete with superconductivity [2]. On the contrary, the overdoped region is generally considered to be a conventional Fermi liquid (FL) and less affected by the doped-Mott-insulator scenario [1]. However, the doping and temperature dependencies of the superfluid density are incompatible with the standard BCS description, suggesting phase fluctuations in the overdoped region [3]. These fluctuations are characterized by photoemission as preformed Cooper pairs well above $T_c$ [4] and magnetic fluctuations persistent in the extremely overdoped regime [5]. Moreover, ferromagnetism has been discovered in overdoped (Bi,Pb)$_2$Sr$_2$CuO$_{6+\delta}$ (Bi2201) [6] and beyond the superconducting dome in La$_{2-x}$Sr$_x$CuO$_4$ [7]. These discoveries suggested that strong electronic correlations remain present in the overdoped cuprates.

Charge orders are ubiquitous in underdoped cuprates [2, 8–16] and have enigmatic interactions with SC [10, 11] and PG [12]; however, there is still no consensus on the underlying mechanism with on-going debates including the real-space electronic correlation scenario [14, 16] versus the momentum-space instability scenario [12]. The latter has been challenged by one resonant X-ray scattering study that revealed charge order in heavily overdoped Bi2201 whose Fermi surface lacks the nesting features [17]. The CO provides a route by which to test how close the overdoped systems are to a FL, and also quantify the underlying fluctuations. Therefore, it is of great importance to assess the universality of charge ordering in overdoped cuprates, which will not only have the potential to elucidate the mechanism of CO instabilities but also provide a valuable perspective to understand the complex phase diagram coming from the overdoped regime.

In this Letter, we report the existence of charge order in heavily overdoped La$_{1.55}$Sr$_{0.45}$CuO$_4$ thin films beyond the superconducting dome (sample growth and characterizations are described in Supplementary Sec.1). The energy resolution of oxygen K-edge resonant inelastic X-ray scattering (RIXS) is $\sim 30$ meV at 41A Taiwan photon source. Figure 1(a) displays the schematic electronic structure of hole doped cuprates and illustrates the resonant absorption as well as the scattering processes at oxygen $K$-edge ($1s \rightarrow 2p$). The Zhang-Rice singlet (ZRS), originating from the hybridization between oxygen ligands and Cu $3d_{x^2-y^2}$ orbitals, manifests as a pre-edge peak in the x-ray absorption spectra (XAS) [18]. Figure 1(b) exemplifies the observed excitations in a metallic LSCO with $x = 0.45$ (resistivity shown in the inset of Fig. 1(b)), including the elastic peak, phonons ($\sim 0.05$ eV), charge excitations ($\sim 0.6$ eV) [20], and oxygen orbital excitations ($> 1.5$ eV). Given the relevance to CO [10, 21, 22], we focus on the elastic peak and low-energy
FIG. 1: Observation of charge order by RIXS in overdoped metallic La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.45$). (a) Schematic plot of RIXS process at O K-edge. (b) A typical RIXS spectrum at $q_{||} = (0.12, 0)$, displaying the elastic peak, phonon, charge and orbital excitations. Inset: resistance curve displaying the metallic nature of the sample. (c) Integrated intensity of elastic peaks for positive and negative (H, 0), and (H, H) directions, using $\sigma$ polarization. Red and blue curves are Lorentzian peak fits to the data with a polynomial background (gray dashed lines). (d) Polarization measurements with $\sigma$- and $\pi$-polarized light, collected at 33 K and 250 K. (e) Reciprocal-space image. The Blue shaded region is the accessible momentum-transfer range of O K-edge. The green and blue lines indicate the momentum cuts, and red dots indicate the observed CO peaks. (f) XAS spectra near the Zhang-Rice Singlet (ZRS) absorption peak with $\sigma$-polarization at normal incidence. Incident energy dependence of the integrated intensity of elastic peak and orbital excitation, normalized to the value at the ZRS peak.

The elastic scattering displays a prominent peak at the planar wavevector $q_{||} = (0.165, 0)$. The full width at half-maximum (FWHM) of the peak is $\sim 0.017 \pm 0.002$ r.l.u. with a correlation length of $\sim 70.8$ Å. This feature is symmetric along both (H, 0) and (-H, 0) directions but absent along diagonal (H, H) directions (Fig. 1(c)) (see RIXS map in Supplementary Fig. S5). Its intensity is more pronounced for $\sigma$-polarization than for $\pi$-polarization (Fig. 1(d)), suggesting a predominant $d_{x^2-y^2}$ character of dopants forming the charge order. And this peak is nearly unchanged at temperatures as high as 250 K (Fig. 1(d)), which is similar to the CO in overdoped Bi2201 [17]. Moreover, this temperature-independent behavior is in line with the short-range high-temperature charge fluctuations (CDF) in the underdoped regions [24]. We also checked the resonant behaviour of this peak to reveal whether it originates from a modulation of the valence electrons. Figure 1(f) shows the intensity of the integrated elastic peak and orbital excitation as a function of incident photon energy compared with XAS. The orbital excitation follows the XAS spectrum, while the integrated elastic peak exhibits a clear resonance with a maximum slightly below the ZRS pre-peak. That is because XAS and scattering signals correspond to the imaginary and real parts of the atomic form factors, respectively. This trend is closely analogous to the behaviour of COs in underdoped cuprates [9, 22]. We have further excluded this peak as a trivial superstructure from hard x-ray scattering measurements (see Supplementary Fig. S4). Thus, we will refer to this feature as a CO peak below. Notably, here the CO wavevector $q_{CO} = (0.165, 0)$ is much smaller than that at underdoped LSCO samples — typically $q_{CO} \sim (0.23, 0)$ [16, 25, 26]. It nevertheless is close to the value in overdoped Bi2201 with $q_{CO} \sim (0.13, 0)$ [17], suggesting that charge instability with relatively long wavelength may prevail in heavily overdoped cuprates.

To investigate whether this CO signal originates from CuO$_2$ planes, we also perform Cu L-edge resonant energy integrated x-ray scattering (REIXS) measurements at Helmholtz-Zentrum Berlin. As shown in Fig. 2(a), we have measured the CO peak along (0, -K) direction (blue cut in Fig. 1(e)) at both Cu L-edge and O K-edge and found it identical at the two edges, suggesting the strong hybridization between the Cu-3d and the O-2p orbitals. This result also demonstrates that the CO is bidirectional along H- and K-directions. Moreover, the CO peak measured by REIXS overlaps very well with the energy-integrated RIXS result (see Supplementary...
FIG. 2: Cu L$_3$-edge and O K-edge REIXS studies of charge order in La$_{1.55}$Sr$_{0.45}$CuO$_4$. (a) Observation of charge order along (0, -K) direction at both Cu L$_3$-edge and O K-edge, offset is applied for clarity. (b) Polarization dependence of the charge order peak, collected at 930 eV and $L = 1.1$ r.l.u. (c) Detuning measurements near the Cu L$_3$-edge after self-absorption correction (see Supplementary). (d) L-dependence of charge order within the accessible range of $[1.1, 1.8]$ r.l.u. at 930 eV, collected with $\sigma$-polarization. (e) $L$-dependence of charge order within the accessible range of $[1.1, 1.8]$ r.l.u. at 930 eV, collected with $\sigma$-polarization.

mentary Fig. S7), proving the consistency of the two experimental techniques. We again observe that the CO peak is stronger at $\sigma$-polarization than $\pi$-polarization at Cu L-edge (Fig. 2(b)) in favor of a charge origin [10]. Figure 2(c) shows the incident energy dependence of CO peak across Cu L$_3$-edge, which does not display a clear resonant behavior due to a relatively poor energy resolution of $\sim 1.3$ eV of REIXS measurement. By selecting the incident x-ray energy at 930 eV with a prominent CO peak, we further investigate the L dependence of CO (Fig. 2(d)). We observe that the CO peak maximizes at $L = 1.1$ r.l.u. with smaller L value inaccessible, which is close to an integer L value. This is in sharp contrast to the behavior of CO in underdoped LSCO [26], which has a maximum at half-integer $L$ due to the modulation of stripes along the c direction [27]. The different L behaviour may relate to the disappearance of spin glass behavior beyond the critical doping of the pseudogap phase ($x_c \sim 0.19$) in LSCO [28], which is favoured by charge-stripe ordering. It is worth noting that CO in underdoped YBa$_2$Cu$_3$O$_{6.67}$ can be enhanced by suppressing SC under magnetic field [29] or optical pump [30], which also shows a maximum at integer L values. The similar L-dependent behaviors of COs induced by the high magnetic field, optical pump and metallic regime suggest that they may have the same origin and be a common feature in the normal state.

To unveil the origin of CO it is crucial to investigate its collective excitations and the interplay between CO and phonons. In underdoped cuprates, it is widely observed that CO coexists with phonon intensity anomaly near the characteristic wavevector, accompanied by different magnitudes of phonon energy softening [16, 21–23, 25, 31]. These have been explained either by enhanced electron-phonon (e-ph) coupling [25] or interference between collective charge fluctuations and phonons [21, 22, 31]. Exploiting the high energy-resolution of O K-edge RIXS, we can probe CO and low-energy excitations at the same time. The RIXS map with better statistics and resolution ($\sim 25$ meV) are visualized in Fig. 3(a). Notably, two phonon branches display pronounced enhancements of intensity near $q_{\text{CO}}$, while the phonon energy softening is negligible here. To quantify the $q_{||}$ dependence of the phonons, we fit the inelastic part of RIXS spectra and reveal three features at $\sim 14$ meV, $\sim 45$ meV and $\sim 75$ meV (See Supplementary Fig. S6). They can be assigned to the acoustic, bond-buckling and bond-stretching phonon modes, respectively, in accord with a recent RIXS study on the optimally doped LSCO [23]. Our intensity distribution curves show that
FIG. 4: Doping dependence of charge order in overdoped La$_{2-x}$Sr$_x$CuO$_4$ and the extended phase diagram. (a, b, c) CO peak profiles measured by Cu L-edge REIXS in LSCO with $x = 0.35, 0.45, 0.6$, respectively. The offset is applied for clarity. $L$ is fixed at 1.1 r.l.u. The peak is nearly temperature independent up to 300 K. (d) The extended CO phase diagram of cuprates. It shows superconducting dome defined by $T_c$, antiferromagnetism (AFM) defined by $T_N$ [32], pseudogap (PG) determined from the Nernst coefficient [33], underdoped charge order and charge fluctuation [16, 24, 26] and overdoped charge order. (e) The doping dependence of the CO wave vector in LSCO and nesting vector obtained from Lindhard function.

We then discuss the possible origins of this re-entrant overdoped CO. First, its emergence at the extremely overdoped region implies that it does not correlate with the pseudogap phase that ends at $x_c ~ 0.19$ [33]. We can also exclude the impact of the van Hove singularity, which was argued to cause the CDW phase in Bi2201 [17], since the Lifshitz transition occurs at much lower doping for LSCO ($x ~ 0.2$) [26]. Figure 4(e) shows the doping dependence of $q_{CO}$ in LSCO [15, 16]. In the underdoped region, the CO wavevector increases with doping but close to 0.25 r.l.u. due to the proximity of spin and charge instabilities [34], while at the overdoped region it is likely to pin to a commensurate vector of $q_{CO} ~ 0.166$ r.l.u.. The Fermi-surface (FS) instabilities induced by the Coulomb interactions may lead to charge fluctuations in the sense of perturbation theory [35]. Accordingly, we have calculated the Lindhard function for LSCO and tracked the doping dependence of the FS nesting vector along the $(H,0)$ direction. The nesting vector shows a non-monotonic behavior with a dip at $p ~ 0.3$ due to the Lifshitz transition from hole-like FS to electron-like FS (see Supplementary Fig. S8). This cannot account for the nearly doping-independent wavevector at (0.166, 0), suggesting that the FS instability is an unlikely route to explain the CO in the overdoped regime.

To investigate the generality of this CO, we study the doping dependence of the CO on another batch of overdoped LSCO thin films ($x = 0.35, 0.45, 0.6$) (Fig. 4). These films have a smaller thickness of 30 unit cells than the above-studied LSCO ($x = 0.45$) with 50 unit cells. We observe that the CO appears at $x = 0.35$ and gets more pronounced at $x = 0.45$ and 0.6, which all persist from 10 K up to 300 K, as shown in Fig. 4, (a) to (c). We fit the REIXS spectrum with a Lorentz function for the peak and a Lorentz background from the specular tail, and the extracted peaks are shown in shaded areas. With smaller thickness, we observe that the CO peak displays a similar wavevector at $\sim 0.166$ r.l.u. but its correlation length becomes much shorter ($\sim 3$ lattice units). This excludes the origin of CO driven from the substrate, while the thickness dependence remains to be understood. The observation of CO in extremely overdoped cuprates provides fresh insight for understanding the phase diagram (Fig. 4(d)). The presence of CO and AFM phases on the two sides of the superconducting dome is enlightening, suggesting that the unconventional superconductivity of cuprates can be regarded as an emergent phase out of either AFM or CO. This is consistent with the coexistence of short-ranged spin fluctuations and CO in the superconducting regime. Previous x-ray scattering studies report that the CO maximized at half-integer $L$ values would disappear at $x \sim 0.25$ in LSCO [16, 26]. Here, the re-entrant CO exhibits distinct temperature and $L$ dependences, as well as different in-plane wave vectors, suggesting different interactions in play compared to CO in the underdoped region.

The nearly doping-independent wavevector at (0.166, 0) suggests that the FS instability is an unlikely route to explain the CO in the overdoped regime. We consider nonperturbatively strong correlations of electrons, which have been found to persist in the
We thank M. Grill, E. Huang, H. Yao, J. Zhang, X. J. Zhou, Y. Li, F. Wang, N. L. Wang, J. Feng, Z. Y. Weng for enlightening discussions. Y. Peng acknowledges the financial support from the National Natural Science Foundation of China (Grant No. 11974029) and the Ministry of Science and Technology of China (Grant No. 2019YFA0308401). Y. Xie acknowledges the financial support from the National Natural Science Foundation of China (Grant No. 12074334). I. E. and Y. W. acknowledge support from the National Science Foundation (NSF) award DMR-2038011. The RIXS experimental data were collected at beamline 41A of the National Synchrotron Radiation Center (NSRRC) in Hsinchu 30076, Taiwan. The REIXS experimental data were collected at the UE46-PGM1 beamline of Bessy-II (Helmholtz-Zentrum Berlin, Germany).

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