



# Vanishing nematic order beyond the pseudogap phase in overdoped cuprate superconductors

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During the last decade, translational and rotational symmetry-breaking phases—density wave order and electronic nematicity—have been established as generic and distinct features of many correlated electron systems, including pnictide and cuprate superconductors. However, in cuprates, the relationship between these electronic symmetry-breaking phases and the enigmatic pseudogap phase remains unclear. Here, we employ resonant X-ray scattering in a cuprate high-temperature superconductor  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO) to navigate the cuprate phase diagram, probing the relationship between electronic nematicity of the Cu 3d orbitals, charge order, and the pseudogap phase as a function of doping. We find evidence for a considerable decrease in electronic nematicity beyond the pseudogap phase, either by raising the temperature through the pseudogap onset temperature  $T^*$  or increasing doping through the pseudogap critical point,  $p^*$ . These results establish a clear link between electronic nematicity, the pseudogap, and its associated quantum criticality in overdoped cuprates. Our findings anticipate that electronic nematicity may play a larger role in understanding the cuprate phase diagram than previously recognized, possibly having a crucial role in the phenomenology of the pseudogap phase.

cuprates | pseudogap | nematicity | quantum criticality | resonant X-ray scattering

There is a growing realization that the essential physics of the cuprate high-temperature superconductors, and perhaps other strongly correlated materials, involves a rich interplay between different electronic symmetry-breaking phases (1–3) like superconductivity, spin or charge density wave (SDW or CDW) order (4–7), antiferromagnetism, electronic nematicity (8–14), and possibly other orders such as pair density wave order (15) or orbital current order (16).

One or more of these orders may also be linked with the existence of a zero-temperature quantum critical point (QCP) in the superconducting state of the cuprates, similar to heavy-fermion, organic, pnictide, and iron-based superconductors (17–19). The significance of the QCP in describing the properties of the cuprates, as a generic organizing principle where quantum fluctuations in the vicinity of the QCP impact a wide swath of the cuprate phase diagram, remains an open question. Evidence for such a QCP and its influence include a linear in temperature resistivity extending to low temperature, strong mass enhancement via quantum oscillation studies (20), and an enhancement in the specific heat (21) in the field induced normal state, with some of the more-direct evidence for a QCP in the cuprates coming from measurements in the material  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO). Moreover, the QCP also appears to be the endpoint of the pseudogap phase (21) that is marked, among other features, by transition of the electronic structure from small Fermi surface that is folded or truncated by the antiferromagnetic zone boundary in the pseudogap phase to a large Fermi surface at higher doping (22, 23) that is consistent with band structure calculations (24). However, in the cuprates, neither the QCP

nor the change in the electronic structure have been definitively associated with a particular symmetry-breaking phase.

In this article, we interrogate the possibility that the cuprates exhibit a connection between electronic nematic order, the pseudogap, and its associated QCP. In the pnictide superconductors, which are similar in many respects to the cuprates, electronic nematic order is more clearly established experimentally, and there have been reports of nematic fluctuations (25), non-Fermi liquid transport (26), and a change in the topology of the Fermi surface associated with a nematic QCP (27). Electronic nematicity refers to a breaking of rotational symmetry of the electronic structure in a manner that is not a straightforward result of crystalline symmetry, such that an additional electronic nematic order parameter beyond the structure would be required to describe the resulting phase. The manifestation of nematic order may therefore depend on the details of the crystal structure of the materials, such as whether the structure is tetragonal or orthorhombic. However, such a state can be difficult to identify in materials that have orthorhombic structures, which would naturally couple to any electronic nematic order and vice versa. Despite these challenges, experimental evidence for electronic nematic order that is distinct from the crystal structure include reports of electronic nematicity from bulk transport (8–10) and magnetometry measurements (11) in  $\text{YBa}_2\text{Cu}_3\text{O}_y$  (YBCO), scanning tunneling microscopy (STM)

## Significance

Understanding the character of the enigmatic pseudogap phase is a fundamental problem in the physics of the cuprate superconductors. Key to this problem is identifying what types of order, if any, are associated with pseudogap. Here, we establish using resonant X-ray scattering that electronic nematicity—a rotational symmetry breaking of the electronic structure—is linked to the onset of the pseudogap phase in the cuprate  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO). We report vanishing of electronic nematicity either by raising the temperature through the onset of the pseudogap phase or by increasing hole doping through the pseudogap critical point, indicating that electronic nematic order is intimately tied to the pseudogap.

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(13, 14, 28) in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Bi2212), inelastic neutron scattering (12) in YBCO, and resonant X-ray scattering (29) in  $(\text{La}, \text{Nd}, \text{Ba}, \text{Sr}, \text{Eu})_2\text{CuO}_4$ . Moreover, STM studies in Bi2212 have reported intraunit cell nematicity disappearing around the pseudogap endpoint (30), which also seems to be a region of enhanced electronic nematic fluctuations (31, 32). In YBCO, there have also been reports of association between nematicity and the pseudogap onset temperature (9, 11).

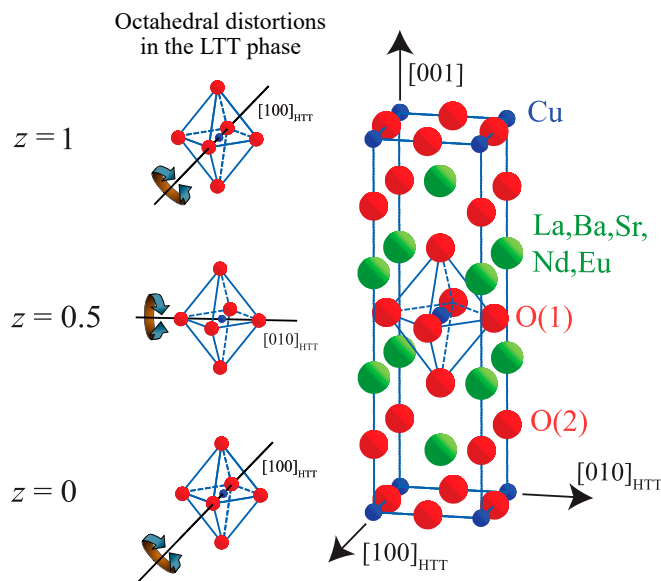
Here, we use resonant X-ray scattering to measure electronic nematic order in the cuprate Nd-LSCO as a function of doping and temperature to explore the relationship of electronic nematicity with the pseudogap phase. While evidence that a quantum critical point governs a wide swath of the phase diagram in hole-doped cuprates and is generic to many material systems remains unclear, investigation of Nd-LSCO provides the opportunity to probe the evolution of electronic nematicity over a wide range of doping in the same material system where some of the most compelling signatures of quantum criticality and electronic structure evolution have been observed. These include a divergence in the heat capacity (21), a change in the electronic structure from angle-dependent magneto-resistance (ADMAR) measurements (24) in the vicinity of the QCP at  $x = 0.23$ , and the onset of the pseudogap (23). Our main result is that we observe a vanishing of the electronic nematic order in Nd-LSCO as hole doping is either increased above  $x = 0.23$ , which has been identified as the QCP doping for this system (21), or when temperature is increased above the pseudogap onset temperature  $T^*$  (23). These observations indicate that electronic nematicity in Nd-LSCO is intimately linked to the pseudogap phase.

### Resonant X-ray Scattering on Nd-LSCO

Our study uses resonant X-ray scattering measurements of the (001) Bragg peak to probe electronic nematicity in Nd-LSCO. Previous work by Achkar et al. (29) showed that the resonant X-ray scattering measurements of the (001) Bragg peak provide sensitivity to electronic nematicity. Understanding how this measurement reveals electronic nematicity requires examination of Nd-LSCO's crystal structure. Nd-LSCO exhibits a structural phase transition from the low-temperature orthorhombic (LTO) to the low-temperature tetragonal phase (LTT) at temperatures between 60 and 90 K, depending on doping (33, 34). The LTO phase is characterized by tilting of the  $\text{CuO}_6$  octahedra along an axis in the  $\text{CuO}_2$  plane but diagonal to the Cu-O bond. In the LTT phase, the axis by which the  $\text{CuO}_6$  octahedra are tilted rotates  $45^\circ$  to be along either the  $a$  or  $b$  axes, such that each individual  $\text{CuO}_2$  layer structurally breaks  $C_4$  rotational symmetry as shown in Fig. 1. The axis of rotation of the octahedra, however, rotates by  $90^\circ$  between neighboring layers producing a crystal structure that is tetragonal, despite having orthorhombic layers. Consequently, probes that average over the (neighboring) layers would yield the same result along the  $a$  and  $b$  axes and may not reveal nematic order.

Resonant X-ray scattering of the (001) Bragg peak, however, provides a direct measure of the difference in the symmetry of the electronic structure between neighboring layers (29, 35). Off resonance or with conventional X-ray diffraction, the (001) Bragg peak is forbidden based on the ionic positions of atoms in the LTT or LTO phases. However, resonant X-ray scattering is sensitive to orbital symmetry, which differs for atoms in neighboring layers in the LTT phase. Moreover, by tuning the photon energy to correspond to different atoms within the unit cell, the electronic symmetry breaking of the  $\text{CuO}_2$  planes—which contain the Cu  $d$  and O  $p$  states that cross the Fermi level  $E_F$  and are most relevant to the low-energy physics of the cuprates—can be differentiated from the  $\text{La}_2\text{O}_2$  spacer layer, which do not have states that cross  $E_F$  but would be sensitive to changes to their orbital symmetry induced purely by structural distortions.

Achkar et al. (29) showed that the temperature dependence of the (001) peak intensity exhibited a dichotomy when the photon energy was tuned to different atoms in the unit cell. An abrupt

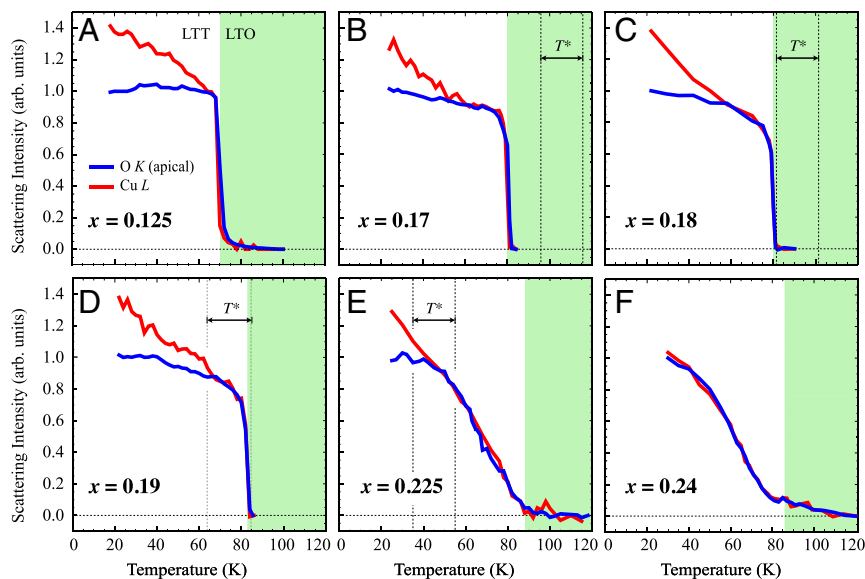


**Fig. 1.** Crystal structure of Nd-LSCO. *Right:* Unit cell of  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  in the high-temperature tetragonal phase (HTT). O(1) and O(2) are in-plane and apical oxygen sites, respectively. In the HTT phase, the axes of the  $\text{CuO}_6$  octahedra are aligned with the axes of the unit cell. The LTO and LTT phases are understood in terms of rotations of the  $\text{CuO}_6$  octahedra. *(Left)* In the LTT phase, octahedra rotate about axes parallel to the Cu-O bonds, with the rotation axis of the octahedra alternating between the  $a$  and  $b$  axes of neighboring planes ( $z = 0$  and  $z = 0.5$ ). This induces  $C_4$  symmetry breaking of the average (or intraunit cell) electronic structure within an individual  $\text{CuO}_2$  plane, referred to here as electronic nematicity.

first-order-like increase at the LTO  $\rightarrow$  LTT transition (consistent with previous measurements of the structural transition) was observed when the photon energy is at the La- $M$  edge ( $\sim 826$  eV) or an energy associated with apical oxygen ( $\sim 532.3$  eV)—atoms in the  $\text{La}_2\text{O}_2$  spacer layer (29, 35). However, a more-gradual temperature dependence was observed when the photon energy is tuned to the Cu- $L$  edge ( $\sim 931.5$  eV) or an energy associated with in-plane O ( $\sim 528.5$  eV)—atoms in the  $\text{CuO}_2$  planes (29). The different temperature dependencies can be understood in terms of a metanematic order, in analogy to a metamagnetic order in magnetism. Here, electronic nematic order in the  $\text{CuO}_2$  planes has a temperature-dependent susceptibility and couples to the lattice order. This provides an additional nematic contribution to the (001) structural Bragg peak. Effectively, this is akin to an internal strain applied to individual  $\text{CuO}_2$  planes by the lattice distortion, with the electronic structure of the  $\text{CuO}_2$  planes (probed by the (001) peak intensity at the Cu- $L$  edge) exhibiting a response that depends on the electronic nematic susceptibility. Importantly, Achkar et al. (29) also showed that this electronic nematic order is coupled to but distinct from charge density wave order. Here, we investigate electronic nematicity as a function of doping (through the pseudogap QCP,  $p^*$ ) and temperature (through the pseudogap onset,  $T^*$ ). Since our measure of electronic nematicity requires the LTT phase, the latter measurement requires a sample with  $T_{\text{LTT}} > T^*$ , which is achieved here at a doping level of  $x = 0.225$ .

### Results

**Doping Evolution of Electronic Nematicity.** In Fig. 2, we show the intensity of the (001) peak as a function of temperature ( $T$ ) at the Cu- $L$  edge (probing the  $\text{CuO}_2$  planes) and the O- $K$  edge at an energy sensitive to apical O (probing the  $\text{La}_2\text{O}_2$  layer). Note, here the data at the different edges is arbitrarily scaled to match in intensity below the LTO  $\rightarrow$  LTT phase transition to highlight the difference in temperature dependence for different doping



**Fig. 2.** Doping and temperature dependence of the (001) Bragg peak intensity in Nd-LSCO. The intensities of the (001) Bragg peak as a function of temperature with photon energy tuned to the apical O-K (blue) and Cu-L (red) edges for  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  with Sr doping levels (A–F):  $x = 0.125, 0.17, 0.18, 0.19, 0.225$  and  $0.24$ . The white and green regions indicate the LTT phase and the LTO phase, respectively. The O-K and Cu-L data are arbitrarily scaled to match in intensity below the LTT phase transition in order to highlight differences in temperature dependence. The dotted lines mark the pseudogap onset temperature,  $T^*$ , in Nd-LSCO based on the high field electrical resistivity measurements from ref. 23. Data on the  $x = 0.125$  sample (A) is reproduced from ref. 29.

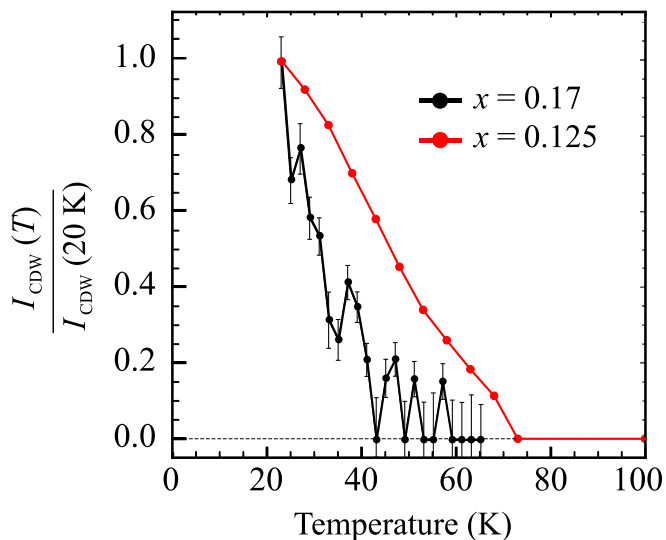
levels. The onset temperature of electronic nematicity,  $T_{\text{EN}}$ , is identified as the temperature where the (001) peak first exhibits a notable deviation between Cu-L and apical O-K edges. In Fig. 24, data at  $x = 0.125$  from ref. 29 is reproduced, which showed a distinct difference in the  $T$  dependence of the (001) peak for measurements at the Cu-L and O-K edges. As the doping is increased in Nd-LSCO, a similar difference in the peak intensity between Cu-L and O-K edges is observed for  $x = 0.17, 0.18, 0.19,$  and  $0.225$  (Fig. 2 A–E). However, for  $x = 0.24$ , as the hole doping is increased across the pseudogap QCP (at  $x \sim 0.23$ ), within the measurement accuracy, we detect no difference in the  $T$  dependence of the (001) peak when measured at the apical O-K and Cu-L edges (Fig. 2F).

Notably we also observe that for  $x = 0.225$ , within the measurement accuracy, we detect no difference in the  $T$  dependence of the (001) peak at the apical O-K and Cu-L edges above the pseudogap onset temperature  $T^*$  but a distinct difference in temperature dependence below  $T^*$  ( $T^*$  is identified in ref. 23 by the resistivity upturn at 50 K in  $x = 0.22$  and 40 K in  $x = 0.23$ ). Our principal conclusion from this result is that static, long-range ordered electronic nematicity in Nd-LSCO is a characteristic of the pseudogap phase, vanishing either upon traversing the QCP with increasing doping or by increasing the temperature above  $T^*$ .

**Doping Evolution of CDW Order.** We also investigated the presence of CDW order in the same series of samples to identify the temperature and doping extent of the CDW phase in the cuprate phase diagram. We observe CDW order in  $x = 0.125$  and  $x = 0.17$  samples, as shown in Fig. 3, but not for  $x = 0.18, 0.19,$  or  $0.24$  above the base temperature ( $\sim 22$  K) of the measurements (SI Appendix, Fig. S4). The onset temperature of CDW order is consistent with the recent Seebeck coefficient measurement (36) in Nd-LSCO. In contrast, SDW order has been observed at low temperature up to a doping as high as  $x = 0.26$  by elastic neutron scattering. However, the onset of SDW order ( $T_{\text{SDW}} = 35$  K in  $x = 0.19$ ) occurs at lower temperatures than our observations of electronic nematicity (37). This shows that the existence of stripe-like CDW or SDW order, that breaks both translational and rotational symmetry, does not appear to be the solely responsible for the

electronic nematicity. Rather, electronic nematicity is either a distinct order or a melted stripe phase that retains rotational symmetry breaking but not the translational symmetry breaking of the CDW or SDW order (38). A schematic phase diagram for Nd-LSCO that summarizes the doping evolution of the LTO  $\rightarrow$  LTT phase transition, the onset of CDW order, and our evidence for long-range ordered electronic nematicity, with onset temperature  $T_{\text{EN}}$ , is depicted in Fig. 4.

Notably, there has been a recent report in the related material LSCO (39), which remains in the LTO phase, that CDW order persists to higher doping levels than in Nd-LSCO; with CDW



**Fig. 3.** Doping and temperature dependence of the CDW peak intensity. The intensities for the CDW peak at Cu-L edge (normalized low-temperature value  $\sim 24$  K) for  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO) with Sr doping levels:  $x = 0.125$  and  $0.17$ . We did not observe a CDW signal at  $x = 0.18, 0.19,$  or  $0.24$  above our base temperature. Data on the  $x = 0.125$  sample is reproduced from ref. 29.

observed up to at least  $p = 0.21$ —which is past the point where the Fermi surface transitions from hole-like to electron-like.

## Discussion

Our observation that electronic nematic order in Nd-LSCO is associated with the pseudogap phase is reinforced by recent measurements in other cuprate families that do not exhibit the LTT structural phase, indicating that an electronic nematic phase terminating at the QCP in overdoped cuprates may be a generic property of the cuprate phase diagram. Specifically in Bi2212, low-temperature STM measurements indicate a disappearance of nematicity at the pseudogap QCP (30), and recent elastoresistance (31) and Raman scattering (32) measurements have shown dynamical electronic nematic fluctuations that are enhanced—in the overdoped region between  $p = 0.20$  and  $0.23$ —and nematic quantum criticality, which is consistent with the putative QCP for Bi2212 at  $p^* \sim 0.22$ .

The questions remain whether electronic nematicity is fundamentally linked to the origins of the pseudogap or is simply stabilized when the pseudogap onsets and what role, if any, quantum nematic fluctuations play in the physics of the cuprates. In the proximity of  $p^*$ , it has also been established that the electronic structure of the cuprates undergoes a Lifshitz transition from a small Fermi surface or Fermi arcs in the pseudogap phase to a large hole like Fermi surface at higher doping in Nd-LSCO (23, 24, 40) and other cuprate families (30, 22, 41). Perhaps most notably, in Bi2212 (30), the Lifshitz transition was shown to occur simultaneous with the disappearance of nematicity, similar to what we observe in Nd-LSCO. This change in the nature of the Fermi surface may provide the conditions for the presence or absence of

electronic nematicity. Or, as suggested by recent theoretical proposals (42), the doping dependence of the electronic nematicity observed here may account for the change in the Hall number around the Lifshitz transition.

It is also possible that the nematicity observed here represents a “vestigial nematicity” that is connected to the pseudogap phase. It has been recently highlighted that in contrast to a CDW order, which can open a gap in the Fermi surface, electronic nematic order is a  $q_{xy} = 0$  order and is not expected to open a gap in the Fermi surface (43, 44). As such, one would not anticipate a natural connection between the pseudogap, which is evidenced by the opening of an antinodal gap, and electronic nematicity. However, a “vestigial nematic” state—derived from disordering a unidirectional CDW but lacking long-range order may open up a gap and is plausibly connected to the pseudogap phase (44). Such vestigial nematic state is consistent with our measurements, where nematicity is present in the pseudogap phase but also at dopings and temperatures where CDW order is not observed.

## Conclusion

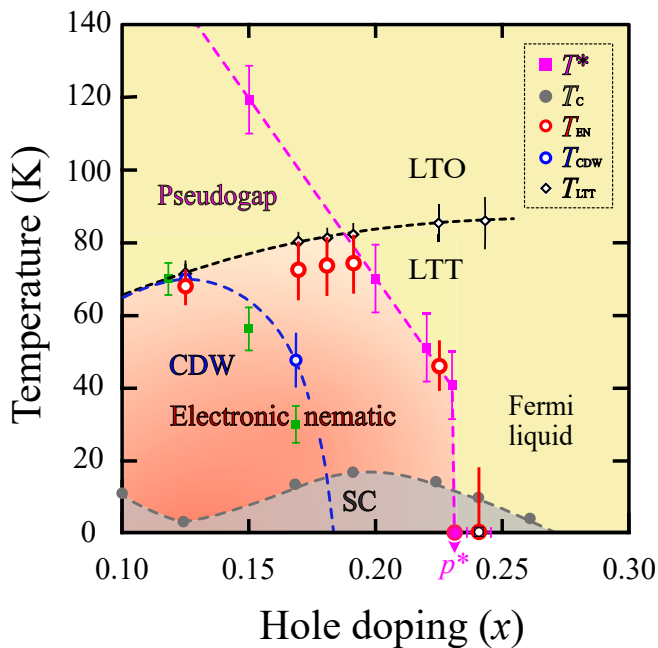
In this study, we report the evidence for vanishing electronic nematic order as the doping is increased beyond the pseudogap QCP at  $x = 0.23$ , that has been identified in Nd-LSCO, or when the temperature is increased above the pseudogap onset,  $T^*$ . This suggests that nematicity is associated with the pseudogap and may possibly be the fluctuating order responsible for the quantum critical phenomena near the pseudogap QCP. Consequently, nematicity may play a larger role in understanding the cuprate phase diagram that has been previously recognized, possibly having a key role in the nature of the pseudogap phase.

## Materials and Methods

Resonant X-ray scattering measurements and X-ray absorption spectroscopy measurements at the Cu-L and O-K edges were performed using the in-vacuum four-circle diffractometer at the Canadian Light Source's Resonant Elastic and Inelastic X-ray Scattering (REIXS) beam line (45). Measurements were performed on single-crystal samples of  $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$  (Nd-LSCO) with  $x = 0.17, 0.18, 0.19, 0.225, \text{ and } 0.24$ . Details of sample growth and characterization of the other samples can be found in ref. 34. Data on the  $x = 0.125$  sample is reproduced from ref. 29. All measurements were performed on the (001) sample surface, with the samples cleaved in air prior to insertion into the vacuum chamber. The vacuum chamber pressure for all measurements was  $\sim 2 \times 10^{-10}$  Torr. The background-subtracted data were fitted to Lorentzian line shapes—raw data, analysis, details, and supporting text are described for the  $x = 0.17$  and  $x = 0.24$  Nd-LSCO single-crystal sample in the *SI Appendix*. The (001) Bragg peak scattering intensity with temperature is obtained in a similar fashion for other doping levels. The crystallographic orientation of the samples was verified in the diffractometer using the (0 0 2) and ( $\pm 1$  0 3) structural Bragg peaks measured with photon energy  $> 2$  keV. Reciprocal lattice units were defined using the lattice constants as  $a = b = 3.787 \text{ \AA}$  and  $c = 13.24 \text{ \AA}$  for Nd-LSCO, where the  $a$  and  $b$  axes are referenced to the high-temperature tetragonal unit cell (34). For  $x > 0.15$ , the LTO to LTT transition is between LTO ( $Bmab$  space group) and LTT ( $P4_2/nm$  space group). For the  $x = 0.125$  sample, the LTO to LTT phase transition is mediated by the LTO2 phase ( $Pccn$  space group), with reduced orthorhombicity relative to LTO and a tilt pattern that alternates between neighboring layers, like the LTT phase (34). The transition depicted in Fig. 2A at 70 K is between the LTO and LTO2 phases but is referred to LTT in Fig. 2A and at  $x = 0.125$  in Fig. 4 for simplicity.

**Data Availability.** All study data are included in the article and/or *SI Appendix*.

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**Fig. 4.** Phase diagram of Nd-LSCO. The doping–temperature phase diagram of Nd-LSCO. The structural phase transition from the LTO to LTT phase,  $T_{\text{LTT}}$  (black diamonds). The onset temperature of the CDW order,  $T_{\text{CDW}}$ , using our resonant X-ray scattering measurement (blue circles) and recent Seebeck coefficient (36) measurement (green squares). The red circles mark the onset of the electronic nematic order in the LTT phase,  $T_{\text{EN}}$ . The superconducting (SC) transition temperature,  $T_{\text{c}}$  (gray circles), has been reported in ref. 34 on the same series of samples as used for our resonant X-ray scattering measurements. The existence of a QCP at  $p^* = 0.23$  has been identified in ref. 21. The pseudogap onset temperature,  $T^*$  (purple squares), as a function of hole doping is measured via the onset of the upturn in electrical resistivity  $\rho(T)$  in ref. 23. The black, purple, blue, and gray dotted lines are a guide to the eye.

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