Bulk charge stripe order competing with superconductivity in La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.12$)

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We present a volume-sensitive high-energy x-ray diffraction study of the underdoped cuprate high temperature superconductor La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.12$, $T_c = 27$ K) in applied magnetic field. Bulk short-range charge stripe order with propagation vector $q_{ch} = (0.231, 0, 0.5)$ is demonstrated to exist below $T_{ch} = 85(10)\ K$ and shown to compete with superconductivity. We argue that bulk charge ordering arises from fluctuating stripes that become pinned near boundaries between orthorhombic twin domains.

A major question in the field of cuprate high-temperature superconductivity concerns the nature of the normal state from which superconductivity emerges in the underdoped region of the phase diagram. Here the opening of a pseudogap manifests itself in the electronic spectrum at temperatures significantly higher than the superconducting transition temperature $T_c$. One school of thought ascribes the pseudogap phenomenon to CDW order [5–12] and the charge stripe order observed in the underdoped region of the phase diagram. Here the opening of a pseudogap manifests itself in the electronic spectrum at temperatures significantly higher than the superconducting transition temperature $T_c$. One school of thought ascribes the pseudogap phenomenon to CDW order [5–12] and the charge stripe order observed in the underdoped region of the phase diagram.

In this Letter, we present direct hard x-ray diffraction evidence for short-range charge stripe order and its competition with superconductivity in La$_{2-x}$Sr$_x$CuO$_4$ ($x = 0.12$). The stripe order is characterized by propagation vector $q_{ch} = (0.231, 0, 1/2)$ and sets in below $T_{ch} = 85(10)\ K$. We argue that stripe order exists essentially in the bulk of the sample volume but that it is pinned by orthorhombic twin domain boundaries. Our results demonstrate that charge stripe order does not require an average LTT structure and that, once established, it displays exactly the same magnetic field- and temperature dependence as the CDW order in YBCO at...
FIG. 1. Momentum scans through the charge stripe order satellite peak $Q_{ch} = (−28, 0, 8.5)$ in LSCO ($x = 0.12$). (a-d) H-scans for temperatures and magnetic fields as indicated. (e) Out-of-plane momentum dependence at 3.8 K and 10 T applied along the $c$-axis. The data in (e) were obtained by scanning along $(0.231, 0, L)$ and subtracting a background estimate evaluated from the average of scans along $(0.271, 0, L)$ and $(0.191, 0, L)$. Regions dominated by powderlines have been removed. All solid lines are fits to Gaussian line shapes.

a comparable doping level and LBCO at concentrations away from $x = 1/8$\cite{20}.

Our bulk-sensitive hard x-ray (100 keV) diffraction experiments were carried out at the BW5 beamline at DESY, Hamburg, operated in triple-axis mode with Ge-gradient Si(111) monochromator and analyzer crystals and a $2 \times 2$ mm aperture to collimate the incident beam. The sample was a 0.8 mm thick platelet with surface normal approximately 45 degrees away from the crystallographic $a$ and $c$ axes. It was mounted inside a 10 T horizontal field cryomagnet. In transmission geometry, this setup allows access to momentum transfers ($Q$) along the crystallographic $c$-axis causes a significant enhancement of the $Q_{ch}$ peak intensity compared to zero field (Fig. 1(a)). Just below $T_c$, at 25 K (Fig. 1(b)), a peak remains in zero field. This peak is, in fact, more intense than at base temperature, and it is only weakly enhanced by the magnetic field. At both 50 K and 70 K (Figs. 1(c) and (d), respectively) the 0 T and 10 T data are identical within errors. In the former case, a broadened profile centered at $Q_{ch}$ can be discerned while at 70 K, any remnant signal centered at $Q_{ch}$ is very weak. Fig. 1(c) illustrates the dependence of the charge order signal on the out-of-plane momentum direction. Prominent broad peaks are observed near half-integer $L$.

We emphasize that the components of the charge and spin propagation vectors $q_{ch} = (−0.231, 0, 1/2)$ and $q_{m} = (0.383, 1/2, 0)$\cite{21, 22} parallel to the CuO$_2$ planes obey the relation $q_{ch}^D = (1/2, 1/2) \pm 1/2 q_{m}^D$ common to charge-stripe ordered materials\cite{13, 15, 17}. Thus, we conclude that the observed $Q_{ch}$ peak in LSCO ($x = 0.12$) reflects the lattice response to charge stripe ordering in orthorhombic LSCO (See Ref. 31 for further support of stripe order in our crystal).

Fig. 2(a) illustrates the temperature and field-dependence of the charge stripe order signal in more detail. To efficiently probe these dependencies we subtracted a background estimate determined as the average of the intensities at $(−0.191, 0, 8.5)$ and $(−0.231, 0, 8.5)$ from the peak intensity collected at $(−0.231, 0, 8.5)$ (closed circles). This procedure is justified by the fact that the background in Figs. 1(a)-(d) is linear to a good approximation. Analysis of full scans (open diamonds in Fig. 2(a)) yield essentially identical results. For $\mu_0 H = 0$ T, the charge stripe peak emerges gradually below an onset temperature which by extrapolation we estimate to be $T_{onset} \simeq 50$ K (grey line in Fig. 2(a)). Upon cooling the peak becomes more intense until $T \simeq T_{c}$. Strikingly, for $T < T_{c}$, a distinct reduction of the intensity is seen, clearly reflecting the competition between charge order and bulk superconductivity. This intensity suppression strongly resembles the CDW response in YBCO\cite{5, 6, 19} and the charge stripe response in LBCO away from $p = 1/8$\cite{20}.

Application of a magnetic field along the $c$-axis sup-
presses superconductivity and, for temperatures smaller than $T_c$ only, enhances the charge stripe order intensity. This last point is brought out most clearly in Fig. 2(b) where we plot the difference between peak intensities measured in finite field and zero field. The enhancement of the charge stripe peak intensity is clear already at 2 T and continues to the highest field probed. The field-dependence at base temperature, extracted from fits to raw data from Ref. [23]). Both quantities have been normalized to their value at base temperature.

The characteristic temperature and field-dependencies of the satellite peak intensity (Figs. 2(a) and 3(a)) appear to be universal signatures of microscopically coexisting but competing superconducting and charge order parameters [5, 23], irrespective of the stripe or density wave nature of the latter. A further hallmark of charge ordered cuprates is broader peaks along the c-axis than along in-plane directions. We can quantify this anisotropy for LSCO ($x = 0.12$) by fitting Gaussian lineshapes to data such as those shown in Fig. 2(a) and extracting correlation lengths, $\xi_c$ and $\xi_a$, as the inverse half-width-at-half-maximum [32]. While this always yielded $\xi_c \sim 6.3 \AA$, implying that the charge stripe order is uncorrelated beyond nearest neighbor CuO$_2$ layers, the in-plane correlation length, $\xi_a$, was found to vary between different experiments. The data in Figs. 2(a)-(d) yield a charge stripe correlation length $\xi_a = 53 \AA$ at 3.8 K and zero field, while a subsequent experiment performed under identical conditions gave $\xi_a = 38 \AA$. This suggests that the charge stripe order is not completely homogeneous in our sample. As will be discussed below, this seems to have a physical reason, and is not related to the crystal’s (high) quality. The key observation, however, is that the evolution of the in-plane correlation length with temperature and magnetic field did not vary between experiments even if the absolute values did. Figs. 2(b) and 3(a) show, respectively, the temperature dependence of the peak width and the field-dependence of the same quantity, normalized to its zero-field, base temperature value. Upon cooling, the peak gradually sharpens up, reaching a minimum below $T_c$ and displaying a small upturn at the lowest temperatures. Similar behaviour has been reported for YBCO [6, 8]. As a function of magnetic field along the c-axis, a ~10% sharpening, reflecting a slight increase of the charge order correlation length, was observed, while an in-plane field leaves the peak width unchanged.

Finally, we turn to the relation between charge order and the crystal structure of LSCO. Fig. 3(a) shows the temperature dependence of a weak signal observed at (300). Peaks of the type $(H \parallel 0)$ with $H = \text{odd}$ are structurally forbidden in the LTO phase but permitted when
and into parts of the sample displaying the average LTO
der must extend significantly beyond the pinning centers
intensity ratio of [40]. On the other hand, to explain the relatively large
of charge order by pinning low-energy fluctuating stripes
with local LTT or LTLO structures cause the emergence
that structural defects in the form of domain boundaries
Consideration of the known stripe-stabilizing potential
reflections (See Ref. [39] for a atomic level explanation).
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orthorhombic twin domain boundaries. From transmis-
scaling with the (100) peak intensity.
LBCO, but not as small as expected in the case of a linear
[20] (Fig. 4(c)). This shows that the charge stripe peak
the peak in Fig. 4(a) is not the signature of an average
the symmetry is lowered to LTT or LTLO [13, 15, 17, 36].
(iii) The integrated intensity of the (100) peak is weaker
LTO
= 255 K [23] of our sample.
(iii) The integrated intensity of the (100) peak is weaker
by a factor of 42 compared to the intrinsic LTT phase of
LSCO at \( p = 1/8 \) (Fig. 4(b)), measured under identical
conditions [10] and normalized by flux and sample vol-
ume, while the 0 T and 10 T charge order peaks LSCO
are weaker only by factors of 4.2 and 2.6, respectively,
than the fully saturated stripe order in LBCO, \( p = 1/8 \)
(Fig. 4(c)). This shows that the charge stripe peak
in LSCO is substantially smaller than the same peak in
LBCO, but not as small as expected in the case of a linear
scaling with the (100) peak intensity.

These considerations led us to conclude that the (100)
and (300) peaks must be related to x-rays diffraeted by
orthorhombic twin domain boundaries. From transmission
electron microscopy studies of samples with compo-
sitions similar to ours [37, 38] it is well known that these
boundaries between LTO twin domains exhibit LTT-type
reflections (See Ref. [32] for a atomic level explanation).
Consideration of the known stripe-stabilizing potential
of LTT or LTLO structures [13, 15, 17] then suggests that
structural defects in the form of domain boundaries
with local LTT or LTLO structures cause the emergence
of charge order by pinning low-energy fluctuating stripes
[40]. On the other hand, to explain the relatively large
intensity ratio of \( q_{ch} \) to (300) peaks, charge stripe order
must extend significantly beyond the pinning centers
and into parts of the sample displaying the average LTO
structure.

This scenario implies that stripe order in orthorhombic
LSCO most likely depends not only on doping, reflecting
a bare charge order susceptibility, and potential com-
mensurability effects, but also on sample-specific details
such as the orthorhombic twin domain structure. Charge
stripe inhomogeneity and glassiness [22, 41] are obvious
possible consequences. The above considerations may
explain why, in a crystal with \( x = 0.10 \) that shows no evidence
for LTT-type reflections, we also found no charge
order. Furthermore, potential differences of the domain
structure of the surface and bulk of a single crystal may
explain why the x-ray study in Ref. [21] found charge
stripe ordering near the surface, but not in the bulk,
of a sample of nominally the same composition as ours.
Inde-
surface regions with local LTT/LTLO structure may
be implicated in observations of shadow-bands in angle
resolved photoemission studies of LSCO [26, 27, 42, 43].
We also note that in samples with average LTT struc-
ture, such as LBCO, the direction of stripes alternate
between the a and b axes, causing charge peaks to cen-
ter at half-integer \( L \) [13, 14]. There is no a priori reason
for this to occur in the LTO structure. The fact that the
peaks in Fig. 4(c) do occur at approximately half-integer
\( L \) therefore strengthens the case for stripe pinning near
the twin domain boundaries with local LTT/LTLO structure.

Our direct detection of charge order in the bulk
of LSCO brings understanding to a host of experiments of
the past two decades (See e.g. Refs. [44–49]), which
suggested that the weak dip in the superconducting
Tc-dome of LSCO [50] may be related to the much more
pronounced 1/8-anomaly in LBCO [51] and to charge stripe
order, but fell short of providing direct evidence. Simi-
larly, in the light of our results, giant phonon anomalies
observed in superconducting LSCO near \( q_{ch} \), and indi-
rectly associated with collective charge excitations (See
e.g. Ref. [52]), can now be directly related to incipient
charge stripe order.

In conclusion, this Letter has demonstrated the exis-
tence of charge stripe order competing with superconduc-
tivity in the bulk of \( La_{2-x}Sr_xCuO_4 \) (\( x = 0.12 \)). Our crys-
tal structure analysis and intensity comparison implies
that the charge stripe order is pinned by tetragonal twin
domain boundaries of the orthorhombic parent phase but
extends far beyond these and into the orthorhombic bulk.

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The propagation vector $q_{\parallel}$ is defined in the first Brillouin zone, while $Q_{\parallel}$ denotes a general satellite peak position. The two are related by $Q_{\parallel} = G \pm q_{\parallel}$, where $G$ is a reciprocal lattice vector.