Spontaneous Symmetry Breaking by Charge Stripes in the High Pressure Phase of Superconducting $La_{1.875}Ba_{0.125}CuO_4$

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In those cases where charge-stripe order has been observed in cuprates, the crystal structure is such that the average rotational symmetry of the CuO_2 planes is reduced from fourfold to twofold. As a result, one could argue that the reduced lattice symmetry is essential to the existence of stripe order. We use pressure to restore the average fourfold symmetry in a single crystal of $La_{1.875}Ba_{0.125}CuO_4$, and show by x-ray diffraction that charge-stripe order still occurs. Thus, electronically driven stripe order can spontaneously break the lattice symmetry.

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Charge and spin stripe order has been observed in a limited class of cuprate superconductors [1–3]. An enduring question is whether stripe correlations represent a fundamental instability of hole-doped CuO_2 planes [4,5], which could be relevant to the unconventional superconductivity, or whether stripes are the consequence of a particular lattice structure with only twofold symmetry of the planes, in which case they would represent a less interesting state that simply competes with bulk superconductivity. There have been theoretical proposals for dynamic electronic correlations that should intrinsically break the fourfold symmetry of the planes [6–8]. Intriguing observations of anisotropic spin [9] and transport [10,11] properties in underdoped $YBa_2Cu_3O_{6+x}$ have been reported; however, the structural symmetry reduction due to Cu-O chains has motivated alternative explanations [12,13]. The observation of spontaneous symmetry breaking by stripe order in an otherwise square lattice would resolve the significance of stripes. In this work, we use high pressure to tune the crystal structure of $La_{1.875}Ba_{0.125}CuO_4$, restoring fourfold symmetry to the planes, and apply x-ray diffraction to demonstrate that, indeed, charge-stripe order still develops. Our results provide strong evidence that stripe correlations in the cuprates are electronically driven and do not depend on reduced lattice symmetry.

At ambient pressure and room temperature, $La_{1.875}Ba_{0.125}CuO_4$ has the high-temperature tetragonal (HTT) structure, with fourfold symmetric planes. On cooling below T_{HT}, the structure transforms to the low-temperature orthorhombic (LTO) phase, and below T_{LT} one reaches the low-temperature tetragonal (LTT) phase [14]. In each of the latter two phases, the CuO_2 planes have only twofold symmetry due to tilts of the Cu-O octahedra about an in-plane axis. In the LTT phase, the tilt axis is along a Cu-O bond direction, so that orthogonal Cu-O in-plane bonds are inequivalent. Previous diffraction studies have shown that T_{HT} and T_{LT} decrease with pressure [15,16]; T_{HT} reaches zero at a critical pressure p_c.

The in-plane anisotropy of the LTT phase pins charge stripes [1,2,17]. If the crystallographic anisotropy drives the charge order (CO), then we would expect the CO to disappear at p_c. Though experimentally challenging, we can test this possibility by directly monitoring the charge order with *in situ* x-ray diffraction. Another quantity that is sensitive to stripe order is the bulk superconducting transition temperature, T_c, which is strongly depressed when stripe order is optimal [18]. If bond-aligned charge-stripe order can only occur in the LTT phase, then one would expect to see a large jump in T_c when the LTT phase is suppressed. Previous studies have indicated a modest, continuous increase in T_c on suppressing the LTT phase, though T_c remains lower than one might anticipate [15,16,19]. We are not aware of any previous attempts to directly measure charge-stripe order under pressure, although there have been recent high pressure studies of charge-density-wave order in Cr [20] and in RTe_3 (R = La, Ce) [21].

The $La_{1.875}Ba_{0.125}CuO_4$ single crystal studied here and those measured in earlier work [22,23] were cut from the same large crystal. A mosaic spread of 0.008° has been determined at the (110) Bragg reflection in the HTT phase, demonstrating an extremely high sample quality. The high pressure x-ray diffraction experiment was performed on the triple-axis diffractometer at wiggler beam line BW5 at HASYLAB, using a piston-type pressure cell [24]. The calibration of the pressure cells and estimation of pressure uncertainties are described in [24]. To increase the signal to background ratio, a sample with optimized shape, 1.6 mm ø × 1.3 mm, was used. Taking advantage of the large penetration depth of 100-keV photons (λ = 0.124 Å), the bulk properties of the charge-stripe order as well as the crystal structure were studied in transmission geometry, using a 1×1 mm^2 beam size. Scattering vectors $\mathbf{Q} = (h, k, \ell)$ are specified in reciprocal lattice units (r.l.u.) of $(2\pi/a, 2\pi/a, 2\pi/c)$, where $a = 3.78$ Å and $c = 13.2$ Å are the lattice parameters of the HTT unit cell.

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Absolute intensities are normalized to a storage ring current of 100 mA. Charge-stripe peak intensities, I_{CO}, measured in several experimental runs, are normalized with $I_{(206)}(0\ \mathrm{GPa})/I_{(206)}(p)$ of the nearby (206) Bragg reflection. The pressure dependence of the superconducting transition temperature T_c was extracted from measurements of the ac susceptibility, performed in a He-gas cell at pressures up to $p = 0.58$ GPa, and in a diamond anvil cell with He pressure medium up to 14.7 GPa.

The temperature versus pressure phase diagram in Fig. 1 summarizes our results. Both the LTO and LTT phase are suppressed at $p_c = 1.85$ GPa. Below p_c the transition temperature of the CO phase, T_{CO}, and T_{LT} are locked together. Unlike the LTT phase, however, the CO phase continues to exist beyond p_c, with T_{CO} remaining higher than T_c. Within the 2.7 GPa range of the diffraction experiment, T_c increases from 3 to 10 K. The inset of Fig. 1 shows in more detail that, even at 14.7 GPa, T_c reaches only 18 K, which is far below the maximum T_c of ~30 K found in $La_{2-x}Ba_xCuO_4$ [25,26].

To identify the different phases in Fig. 1, we have performed scans in reciprocal space through specific reflections; Fig. 2 presents key results. Figure 2(a) shows that the orthorhombic splitting between the (200) and (020) Bragg reflections (simultaneously present due to twin domains) is clearly resolved. The pressure dependence of the orthorhombic strain, $2(b-a)/(a+b)$, at $T \gtrsim T_{LT}$ is shown in Fig. 2(b), together with the calculated average tilt angle Φ of the CuO_6 octahedra.

That the suppression of the average octahedral tilts at $p = p_c$ also occurs at our base temperature (10 K) is demonstrated by the decay of the (100) superlattice intensity in Fig. 2(c); the (100) is unique to the LTT phase. In sharp contrast, the figure shows that the intensity of the CO superlattice peak $(2 + 2\delta, 0, 5.5)$ decreases only modestly with pressure, remaining substantial at $p \gg p_c$ and indicating that CO survives in the phase with restored fourfold symmetry. (Note that the survival of the CO in the HTT phase has been observed at two pressures, 2.33 and 2.7 GPa, although the temperature dependence was measured only at the second of these.) The temperature-dependent data in Fig. 3(b) demonstrate that T_{CO} also decreases only gradually with pressure. The T_{LT} transition decreases at the same rate (for $p < p_c$), as indicated in Fig. 3(a), but the amplitude of the LTT lattice modulation is strongly suppressed as $p \rightarrow p_c$.

The observation that $T_{CO} = T_{LT}$ for $p < p_c$ is consistent with a unidirectional charge modulation that couples to the

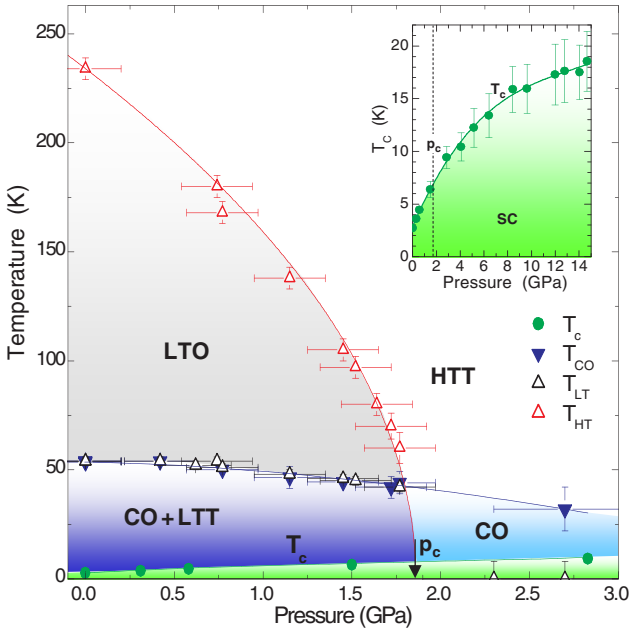

FIG. 1 (color online). Observed temperature versus pressure phase diagram of $La_{1.875}Ba_{0.125}CuO_4$. Indicated are the transition temperatures of the structural phases HTT, LTO, and LTT, the charge-stripe phase (CO), and bulk superconductivity (SC). At ambient pressure we find $T_{HT} = 235$ K, $T_{LT} = 54$ K, $T_{CO} = 54$ K, and $T_c = 3$ K, respectively. The line separating LTO from HTT describes $T_{HT}(p)$ using $T_{HT}(0)[(p_c - p)/p_c]^{0.5}$ with a critical pressure of $p_c = 1.85$. Inset: T_c for pressures up to 14.7 GPa.

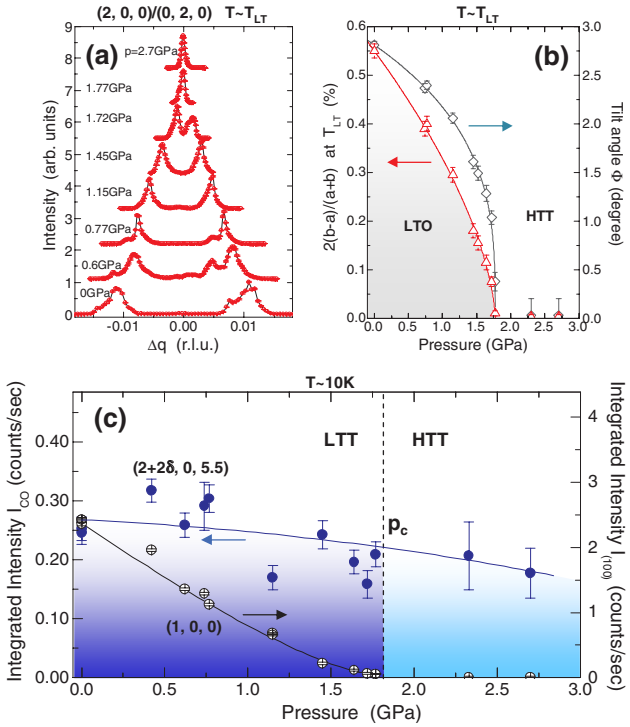

FIG. 2 (color online). Pressure dependence of crystal structure and charge-stripe order. (a) Transverse scans at T_{LT} through the (200) and (020) Bragg reflections, showing the suppression of the orthorhombic splitting with pressure. (b) Orthorhombic strain calculated from the splitting in (a), and average tilt angle Φ, calculated using $\Phi^2 = f(b-a)$ with $f = 380\,(^\circ)^2/\text{Å}$ [15]. (c) Comparison of the integrated intensity from k scans through the (100) LTT peak and the $(2 + 2\delta, 0, 5.5)$ CO peak. The vertical line indicates the critical pressure p_c. Solid lines are guides to the eye.

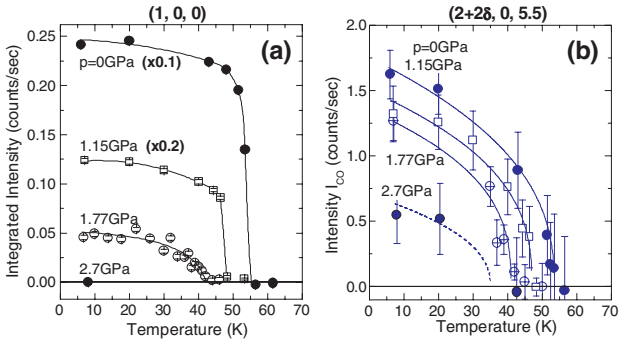

FIG. 3 (color online). Temperature and pressure dependence of LTT phase and charge-stripe order. (a) Integrated intensity from k scans through the (100) LTT peak. (b) Integrated intensity from h scans through the $(2 + 2\delta, 0, 5.5)$ CO peak. For the results at 2.7 GPa, in particular, the intensity versus temperature is confirmed by k scans. Lines are guides to the eye.

in-plane anisotropy of the LTT phase. In fact, without the presence of the stripe order, one might expect T_{LT} to decrease with pressure in proportion to T_{HT}, as $\Phi \rightarrow 0$. With the detection of charge-stripe order in the HTT phase at $p > p_c$, one might expect that some local tilt distortions could be induced. (Previous work at ambient pressure has established that static octahedral tilt disorder is common in the HTT phase of $La_{2-x}Ba_xCuO_4$ [27–29].) We show next that this is, in fact, the case.

Figure 4 shows transverse scans of the $(2 + 2\delta, 0, 5.5)$ CO peak at pressures from below to above p_c. The CO peak has a finite width at ambient pressure [30], with the inferred correlation length ξ decreasing only slightly from 120 Å at 0 GPa to 80 Å at 2.7 GPa. Ideally, we would like to test at $p > p_c$ for possible diffuse scattering at a superlattice position such as (100) that is unique to the LTT phase; unfortunately, those features have very small structure factors. Instead, we have followed the (1.5, 1.5, 2) peak, which is sensitive to octahedral tilts in both the LTO and LTT phases. Transverse scans through that position are indicated in green in Fig. 4. At ambient pressure, the peak is resolution limited, while it has developed a small but finite width at 1.77 GPa ($\xi \sim 500$ Å). Finally, at 2.7 GPa, where there is no long-range order associated with octahedral tilts, the width of the scattering centered at (1.5, 1.5, 2) matches that of the CO peak. The width of the weak, diffuse (1.5, 1.5, 2) peak is found to saturate at its minimum value for $T < T_{\mathrm{CO}}$.

Based on the scattering data, we come to the conclusion that charge stripes in the HTT phase are consistent with a short-range ($\xi \sim 5$ stripe periods) smectic electronic liquid-crystal state [6] that breaks the rotational symmetry and freezes in the high pressure regime due to its coupling to local octahedral tilts. The smooth variation of the stripe order through p_c is also consistent with the gradual rise in superconducting T_c, as indicated in Fig. 1. The depression of the bulk T_c by stripe order at ambient pressure is known to be due to a frustration of interlayer coupling, as there is

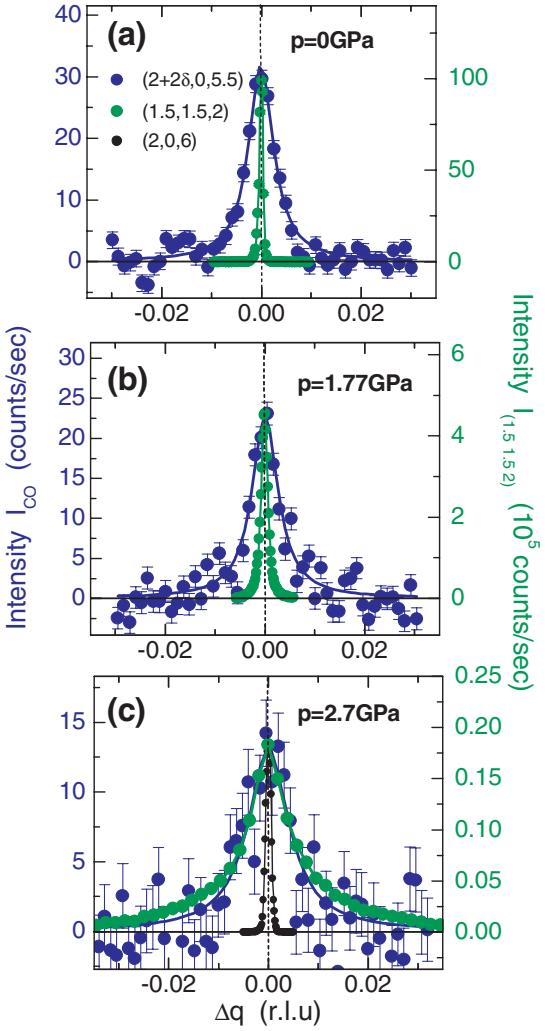

FIG. 4 (color online). Comparison of CO peak and tilt-peak profiles. k scans through the $(2 + 2\delta, 0, 5.5)$ CO peak and transverse (h, k) scans through the (1.5, 1.5, 2) tilt peak at $T \sim 10$ K for representative pressures: (a) $p = 0$, (b) $p \lesssim p_c$, and (c) $p > p_c$. A linear background has been subtracted. Solid lines through the CO peaks are fitted Lorentzians. In (c) the k scan through the (206) Bragg reflection demonstrates the insignificance of pressure induced broadening at $p = 2.7$ GPa. The data in (c) were collected with a pressure cell with thicker walls (stronger absorption), resulting in lower counting statistics.

evidence of two-dimensional superconducting correlations at temperatures as high as 40 K [22,30]. It has been proposed that the frustration could be due to a type of striped superconducting state [31,32], also described as a pair-density-wave (PDW) state [33]. That proposal depends on the 90° rotation of the stripes from one layer to the next that is associated with stripe pinning in the LTT phase [17]. Figure 5 shows scans of the CO intensity over a range of $\mathbf{Q}$ varying perpendicular to the planes. At high pressure we observe the same sinusoidal modulation (with zeros at integer ℓ) as at ambient, demonstrating that in the HTT phase the charge stripes retain the interlayer correlations found in the LTT.

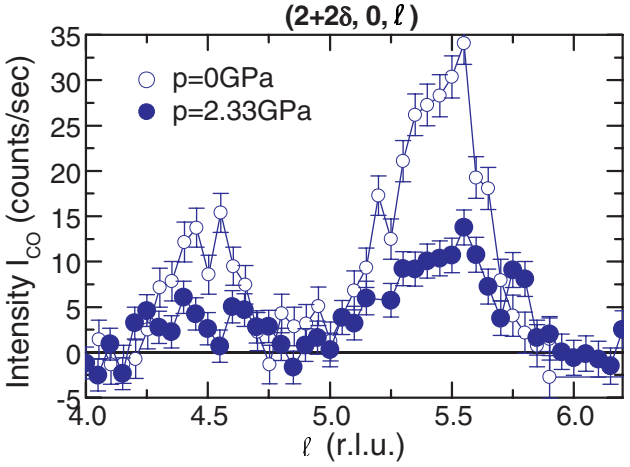

FIG. 5 (color online). Background-subtracted intensity of the CO intensity measured along $\mathbf{Q} = (2 + 2\delta, 0, \ell)$ at base temperature for $p = 0$ GPa (open circles) and $p = 2.33$ GPa (filled circles). In the first case, background was measured along the same $\mathbf{Q}$ at $T = 60$ K; in the second, background was measured at base temperature along a parallel path in $\mathbf{Q}$ displaced by $(0, 0.03, 0)$, corresponding to a displacement of several peak widths in k.

It was noted quite some time ago that the impact of pressure on T_c in $La_{2-x}Ba_xCuO_4$ is very sensitive to x [19]. In particular, T_c rises very slowly with p for $x = \frac{1}{8}$, but quite rapidly for x slightly larger or smaller than $\frac{1}{8}$. Compared to our results, Crawford *et al.* [15] found a more rapid rise in T_c near p_c in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$, which might be due to the small nominal difference in hole doping. Takeshita *et al.* observed an extreme sensitivity of T_c to strain along [110] in $La_{1.64}Eu_{0.2}Sr_{0.16}CuO_4$, but this could be due to inducing a transition from LTT to LTO. The survival of charge-stripe order in the HTT phase of $La_{1.875}Ba_{0.125}CuO_4$, along with extreme sensitivity of $T_c(p)$ to x, may indicate a special stability at $x = \frac{1}{8}$ for the order (such as the proposed PDW state) that decouples the layers.

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