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Unveiling Unequivocal Charge Stripe Order in a Prototypical Cuprate Superconductor

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In the cuprates, high-temperature superconductivity, spin-density-wave order, and charge-density-wave (CDW) order are intertwined, and symmetry determination is challenging due to domain formation. We investigated the CDW in the prototypical cuprate $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ via x-ray diffraction employing uniaxial pressure as a domain-selective stimulus to establish the unidirectional nature of the CDW unambiguously. A fivefold enhancement of the CDW amplitude is found when homogeneous superconductivity is partially suppressed by magnetic field. This field-induced state provides an ideal search environment for a putative pair-density-wave state.

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A hallmark of correlated quantum matter is a “zoo” of complex phases in close proximity. It is striking that despite uniquely distinct underlying microscopic interactions and the expected differences in electronic complexity, the resulting phase diagrams are remarkably similar across a large range of material classes. Notably, unconventional superconductivity is almost conclusively found in the vicinity of magnetic quantum instabilities surrounded by a host of competing electronic phases. In the cuprates, high-temperature superconductivity emerges close to a spin-density-wave instability that is accompanied by a mysterious pseudogap state [1], strange metallic behavior [2], electronic nematic phases [3,4], and multiple charge-density-wave (CDW) orders [5,6]. Similar to unconventional superconductivity, a majority of these electronic companion states have been identified in all classes of cuprates as well as in entirely different material families [7] such as the iron pnictides [8], ruthenates [9], and heavy fermion materials [10]. This poses the general question of whether a fundamental principle exists according to which this complex arrangement of correlated metallic quantum states is organized.

A long-standing proposal is that the entire problem derives from a primary order (a mother state) [11], from which all other phases descend via breaking of relevant symmetries. Pair-density waves (PDWs)—spatially modulated superconducting states (see Fig. 1) evidenced by surface tunneling, transport, and far-infrared reflectivity [12–15]—are contenders for such a mother state [11].

However, direct corroboration from diffraction experiments is missing. In Fig. 1(a), we illustrate a commonly assumed relationship between a unidirectional PDW and CDW order. Progress on this topic has been held back by the difficulty to determine the CDW symmetry. Because of its bulk sensitivity to periodic charge modulations such as the ones imposed by the atomic lattice, x-ray diffraction is an ideal probe for charge ordering. In the prototypical cuprate $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ without external stimuli, the CDW order has been interpreted both in terms of unidirectional and bidirectional structures [16–19]. Recent resonant x-ray scattering experiments in combination with uniaxial pressure have been interpreted as evidence in favor of a unidirectional CDW structure [20,21].

For the La-based cuprates, the CDW phase is widely believed to consist of charge stripes [22–27], although a checkerboard-type magnetic vortex structure producing identical spin and charge diffraction patterns has been proposed [28]. As we illustrate in Figs. 1(b) and 1(c), the observed diffraction pattern is consistent with both biaxial (“checkerboard”) and unidirectional (“stripe”) CDW order. This is because for a fourfold symmetric system, stripes generically form domains [Fig. 1(c)], resulting in diffraction peaks in both directions just like the (single domain) checkerboard order.

Here we combine a novel uniaxial pressure cell with high-resolution synchrotron x-ray diffraction at high magnetic fields to unambiguously discern the symmetry of the CDW in the prototypical cuprate $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ (LSCO) and

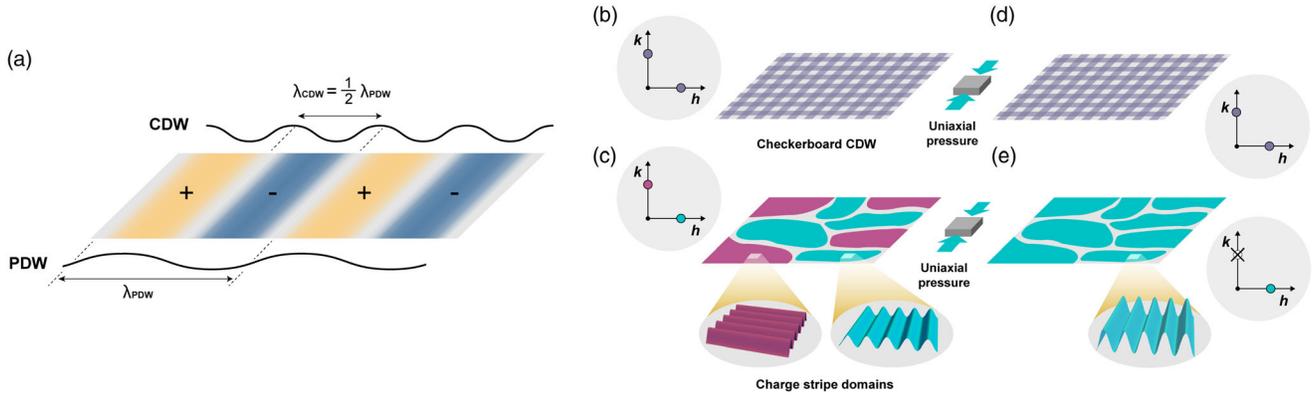


FIG. 1. Intertwined nature of pair-density-wave (PDW) and charge-density-wave (CDW) order and their relation with x-ray diffraction (XRD). (a) A schematic representation of a unidirectional PDW state (yellow and blue stripes) in real space is shown together with the intertwined CDW state (white stripes). The PDW order parameter changes sign in alternate domains with a period λ_{PDW} , and vanishes at the domain walls. In contrast, the local electronic density of states is enhanced at the domain walls resulting in an interwoven CDW pattern with a period $\lambda_{\text{CDW}} = \frac{1}{2} \lambda_{\text{PDW}}$. This illustrates that knowledge of the symmetry of the CDW is vital to identify PDW order. However, due to the fourfold symmetry of $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$, both checkerboard order in (b) as well as degenerate domains of uniaxial stripes [purple and cyan patches in (c)] are allowed. Notably, they result in identical diffraction patterns as illustrated in the circular insets of the respective panels. Note that for panel (c), we have color coded the XRD Bragg reflections in the inset with the color corresponding to the associated stripe domain. Here we use uniaxial pressure to break the fourfold symmetry. As shown in (d) this does not substantially alter the checkerboard XRD intensities. In contrast, it removes the degeneracy of uniaxial CDW stripe order, resulting in a monodomain state (e). As a result, the Bragg reflection associated with the extinct purple domain entirely vanishes as indicated with a black cross in the inset.

its competition with high-temperature superconductivity. Notably, the response of checkerboard and stripe charge order to uniaxial pressure is markedly different [see Figs. 1(d) and 1(e)]. We demonstrate that uniaxial pressure along the copper-oxygen bond provides an external stimulus that lifts the stripe order degeneracy revealing that the CDW is represented by stripes. A symmetry related PDW mother state—if observable with diffraction—is therefore likely to be unidirectional in nature. Via application of high magnetic fields to suppress partially the concomitant homogeneous high-temperature superconductivity, we show that stripe CDW is enhanced fivefold. Because of the strongly intertwined nature of spatially modulated superconductivity with CDW order [cf. Fig. 1(a)], this environment characterized by simultaneous suppression of homogeneous superconductivity and substantial enhancement of CDW order is an ideal setting to identify—by diffraction—the putative PDW state.

High-energy (100 keV) x-ray diffraction experiments were carried out at the P21.1 beamline at PETRA III (DESY) on floating-zone-grown $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ single crystals ($T_c = 27$ K) [29]. The orthorhombic ($Bmab$) structure ($T < 250$ K) generates a spontaneous strain diagonally to the charge-density modulations. Our uniaxial pressure device (see Fig. 1 in Supplemental Material [30]) is compatible with both a standard displax cryostat and a 10 T cryomagnet. The cryomagnet sample environment restricts the accessible momentum space to the scattering plane spanned by the a and c axes. With this configuration, pressure is applied perpendicularly to the scattering plane (along the b -axis direction) while magnetic field points

along the c axis. A zero-field four-circle setup, by contrast, allows access to both the a - c and b - c scattering planes. In this fashion, tensile and compressive strain are accessed directly by lattice parameter measurements [20] (Fig. 2 in Supplemental Material [30]). Uniaxial pressure along a Cu—O bond direction detwines the orthorhombic domains (Figs. 3 and 4 in Supplemental Material [30]). Note that conventional detwining by pressing along the Cu-Cu direction does not by itself impact on charge order in LSCO [35,36]. Probing the charge order reflections $(\delta, 0, \ell)$ and $(0, \delta, \ell)$ with $\delta \sim 0.235$ and $\ell = 8.5, 12.5$ with or without an analyzer yields consistent results (Fig. 5 in Supplemental Material [30]). Amplitude, position and width of the charge order reflections are extracted from fits to a Voigt function. Correlation lengths are defined by the inverse half width at half maximum.

In $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$, CDW order manifests itself by satellite reflections at $\mathbf{Q} = \boldsymbol{\tau} + \mathbf{q}_i$. Here $\boldsymbol{\tau}$ represents fundamental lattice Bragg peaks and for La-based cuprates, $\mathbf{q}_1 = (\delta, 0, 0.5)$ and $\mathbf{q}_2 = (0, \delta, 0.5)$, with $\delta \approx 1/4$ reciprocal lattice units [22,23,25,26] using lattice parameters $a \approx 3.8 \geq b$ and $c \approx 13.2$ Å. In-plane pressure along the b axis produces a compressive (C_i^b) strain. In turn, tensile strains propagate along the orthogonal in-plane (T_i^a) and out-of-plane (T_c^c) directions [Figs. 2(a) and 2(b)]. Along these high-symmetry crystallographic axes, compressive or tensile strain is defined as $\epsilon_j = (u_j - u_j^0)/u_j^0$, where u_j is the lattice parameter along one of the three directions C_i^b , T_i^a , or T_c^c , and u_j^0 refers to the zero-pressure lattice constants.

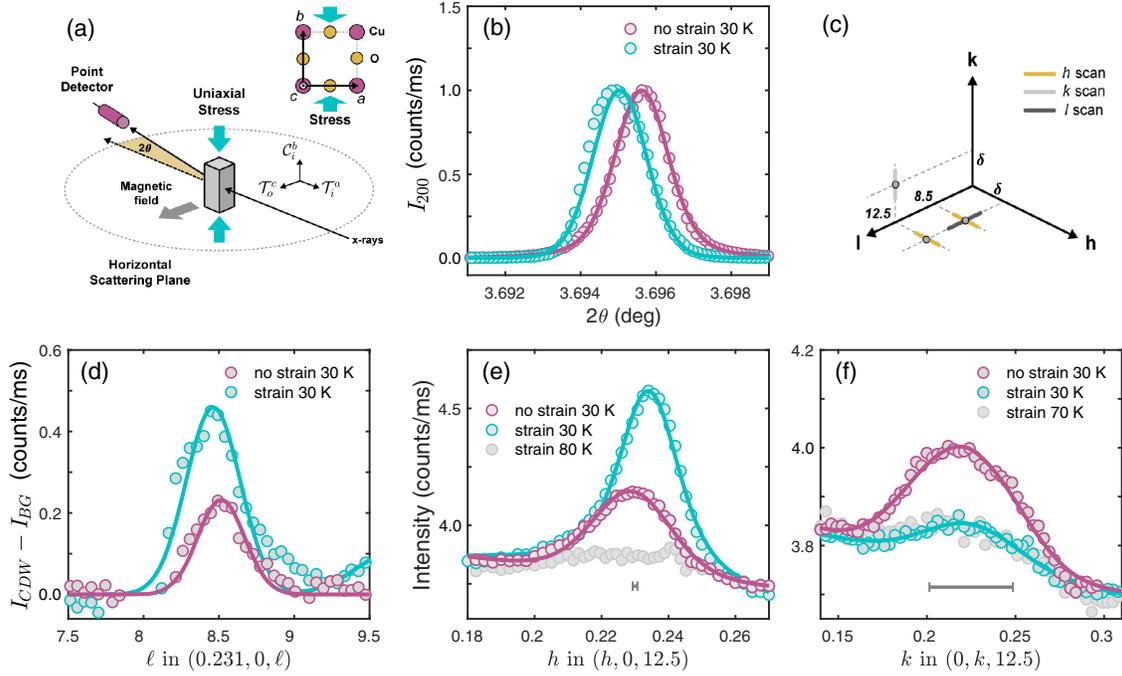


FIG. 2. Effect of uniaxial pressure on the charge-density wave in $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$. (a) Schematic of the experimental scattering geometry. Uniaxial pressure and magnetic field are applied along the b and c axes, respectively. (b) Demonstration of how uniaxial pressure along the b axis enhances the a -axis lattice parameter—smaller scattering angle 2θ implies larger Cu-O-Cu distance. (c) Schematic illustration of the studied part of reciprocal space. Yellow, gray, and dark gray lines indicate, respectively, h , k , and ℓ scans through CDW reflections. Panels (d)–(f) show diffraction intensity of h , k , and ℓ scans through CDW ordering vectors without (purple) and with (cyan) application of uniaxial strain, respectively. For the ℓ scan (d), covering two Brillouin zones (BZs), background subtraction was performed. The h and k scans cover only a small fraction of a BZ and, hence, no subtraction is required. Error bars in (b) and (d)–(f), set by counting statistics, are smaller than the used symbols. Horizontal bars in (e) and (f) indicate the instrumental momentum resolution, and solid lines are fits to a Voigt profile.

Starting in the normal state ($T = 30$ K), uniaxial b -axis pressure induces an approximately twofold enhancement of the charge order reflections found at $(\delta, 0, \ell)$ with $\ell = 8.5, 12.5$ [Figs. 2(c)–2(e)]. Along the C_i^b direction, by contrast, the charge order reflection $(0, \delta, 12.5)$ is completely suppressed [Fig. 2(f)]. The uniaxial-pressure-enhanced CDW order along the T_i^a axis displays a temperature dependence that within a twofold scaling factor is identical to that found under ambient pressure [see Fig. 3(e) and also Fig. 8 in Supplemental Material [30]]. Upon cooling into the superconducting state ($T < T_c$), the charge order is partially suppressed, as commonly found in the cuprates [37].

We find that a c -axis magnetic field has no impact along the C_i^b direction. The CDW order remains completely suppressed even in a 10 T magnetic field [Figs. 3(a) and 3(c)]. By contrast, along the T_i^a direction the CDW diffraction amplitude displays a strong magnetic field effect inside the superconducting state. The stripe order peak is enhanced by another factor of ~ 2.5 upon application of 10 T [Figs. 3(b) and 3(d)]. The in-plane correlation length (T_i^a direction) is only marginally improved with the application of strain. Application of magnetic field induces a more significant increase of the correlation length that reaches $\xi_a = 70$ Å [Fig. 3(f)]. Neither uniaxial pressure nor

magnetic field influences the out-of-plane correlation length (see Fig. 9 in Supplemental Material [30]).

The enhanced CDW diffraction intensity is insensitive to the applied stress magnitude. We find that a strain of $\epsilon_a = 0.01\%$ is enough to trigger the stripe order structure [Fig. 4(a)]. The twofold enhancement remains up to the largest applied strain $\epsilon_a \sim 0.03\%$. Finally, we find that the incommensurability δ is marginally larger in the detwinned stripe ordered phase [Fig. 4(b)].

Although uniaxial pressure may—in principle—change the space group of the crystal structure, such structural changes must be subtle. The previous uniaxial pressure work implicitly assumed the crystal space group remains unchanged [21]. As shown in Fig. 4 in the Supplemental Material [30], the $(2,2,0)$ Bragg reflection remains after the detwinning process, consistent with average $Bmab$ crystal structure. We note that LSCO displays $Bmab$ forbidden Bragg peaks assigned to monoclinic distortions that already exist in absence of uniaxial pressure [38–40]. The primary uniaxial pressure effect on the stripe order is therefore most likely of electronic nature. Mild modification of orthorhombic distortions may nevertheless impact on the incommensurability [41]. Uniaxial pressure may also influence the low-energy electronic structure. The band structure of

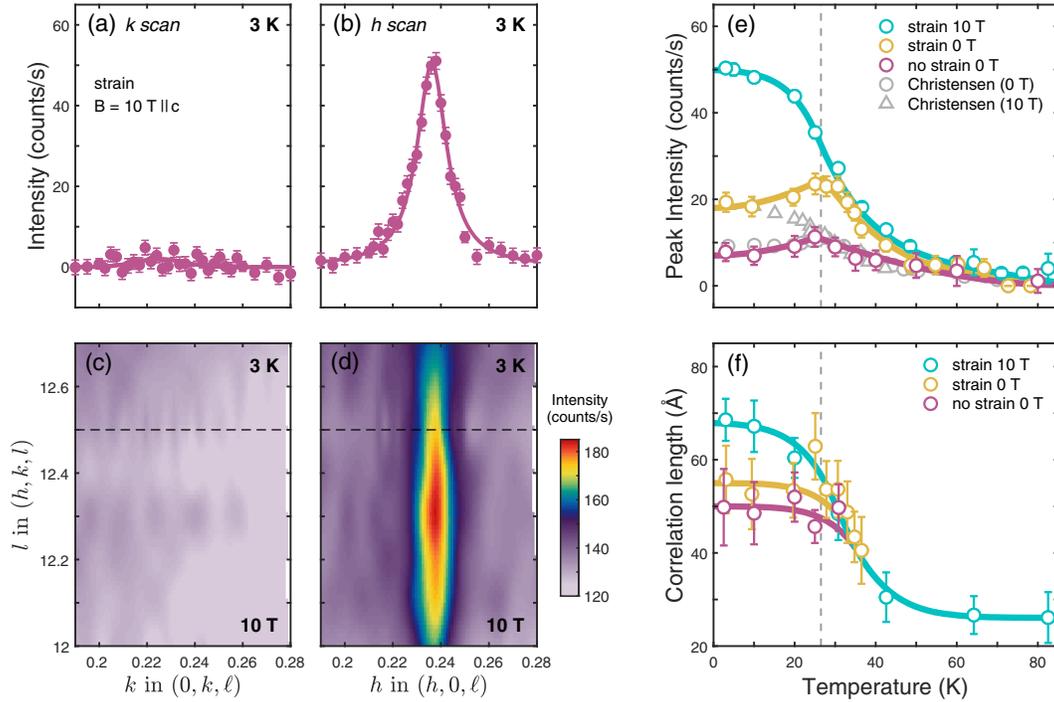


FIG. 3. Magnetic field and temperature dependence of strain-induced charge stripe order. (a),(b) Momentum h and k scans recorded in a c -axis magnetic field of 10 T and with compressive strain along the b -axis direction for $\ell = 12.5$. Absence of charge order along the direction of applied strain reveals the unidirectional nature of the charge-density wave. Error bars are set by counting statistics. (c),(d) Diffraction intensity, displayed in false color scale, around, respectively, $(0, \delta, 12.5)$ and $(\delta, 0, 12.5)$ recorded with temperature and magnetic field as indicated. (e),(f) CDW peak amplitude and correlation length as a function of temperature for magnetic field and strain conditions as indicated. Gray data points are adopted from Ref. [24]. Error bars reflect standard deviations of the fits. Gray dashed line indicates the superconducting transition temperature $T_c = 27 \text{ K}$.

LSCO has a van Hove singularity in the vicinity of the Fermi level [42]. Uniaxial pressure acts to push this singularity closer to the Fermi level (and eventually across) along the T_i^b direction [43,44]. This increase of density of states at the Fermi level may be involved in the stripe order domain lifting.

A charge-density modulation $\delta\rho(r)$ as discussed above is described by [41,45]

$$\delta\rho(r) = \text{Re}(\Phi_x e^{iq_1 \cdot r}) + \text{Re}(\Phi_y e^{iq_2 \cdot r}), \quad (1)$$

where \mathbf{r} is a spatial coordinate vector. The amplitudes $|\Phi_x|$ and $|\Phi_y|$ are nonzero for biaxial structures, whereas stripe order refers to the case with only one nonzero amplitude. Note that for diffraction experiments, domains of stripe order are virtually indistinguishable from a biaxial ordering. The effect of uniaxial pressure on a single domain is captured phenomenologically by a simple Landau free energy,

$$F = \alpha(T)(|\Phi_x|^2 + |\Phi_y|^2) + \frac{\beta}{2}(|\Phi_x|^2 + |\Phi_y|^2)^2 - \gamma|\Phi_x|^2|\Phi_y|^2 - \epsilon(|\Phi_x|^2 - |\Phi_y|^2), \quad (2)$$

where we assume homogeneous order and $\alpha(T)$, β , and γ are phenomenological parameters [41,45]. A sign change in $\alpha(T)$ models the temperature-driven symmetry breaking. Without strain, $\epsilon = 0$, $\gamma < 0$ leads to a stripe order with two degenerate solutions and $\gamma > 0$ leads to biaxial order with $|\Phi_x| = |\Phi_y|$. The effect of strain, in other words $\epsilon \neq 0$, in the two cases then differs significantly: While for stripe order a small but finite strain will lift the degeneracy and, hence, align all domains, the ratio of the magnitudes of the two components of the biaxial order changes continuously with strain. Consequently, the amplitudes in diffraction experiments are doubling for the detwinned stripe order domain. Only for fine-tuned situations with a near degeneracy of stripe and biaxial order or for huge pressures a similar doubling occurs for the biaxial case (see Supplemental Material [30]). Note that the Landau free energy above cannot account for the magnetic field effect inside the superconducting state, which we interpret as evidence for a competing interaction between these two orders. To describe this competition, it requires adding the superconducting order parameter and a competing interaction term to the Landau free-energy model [46].

Our study reveals charge stripe order in LSCO in its purest form. Notably, we demonstrate that stripe order that is coupled with unconventional superconductivity is an

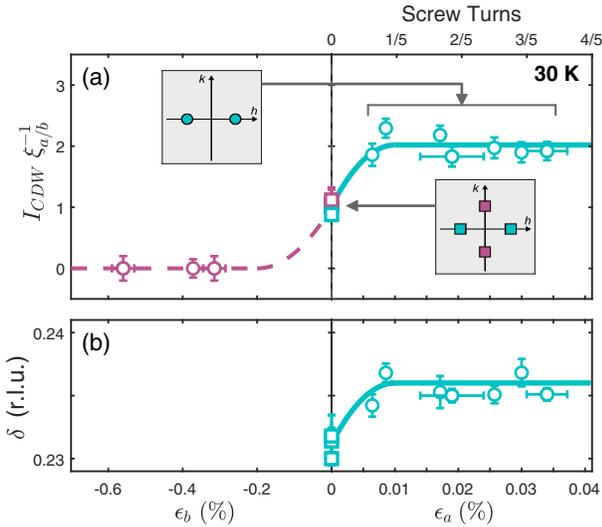


FIG. 4. Strain-induced stripe charge order detwinning. (a) Integrated charge-density wave intensity as a function of strain $\epsilon_a = \Delta a/a$ and $\epsilon_b = \Delta b/b$ along, respectively, the a (tensile strain \mathcal{T}_i^a direction) and b (compressive strain \mathcal{C}_i^b direction) axis. Insets illustrate the observed diffraction patterns consistent with stripe order (left) and biaxial or domains of stripe order (right), respectively. (b) Charge-density-wave incommensurability δ as a function of strain.

intrinsic electronic property of underdoped cuprates. Static charge stripe ordering coupled to superconductivity is a key condition to realize an exotic PDW state [11]. Depending on the CDW symmetry, different flavors of PDW order have been postulated. Our results are not direct evidence of a PDW state, but narrow down theoretical possibilities. Detwinned charge stripe order provides an experimental recipe to uncover subtle signatures of a PDW state, that manifest by an electronic modulation commensurate to the stripe order (see Fig. 1). In fact, resonant x-ray scattering in combination with uniaxial pressure is a promising experimental route to search for the proposed PDW mother state.

In summary, we have carried out a high-energy x-ray diffraction study of charge order structure in $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$ upon application of uniaxial pressure and magnetic field. Twinned stripes and checkerboard order exhibit virtually identical diffraction patterns. The two charge order structures, however, react very differently to application of strain along the copper-oxygen bond direction. Checkerboard order is only marginally impacted by uniaxial pressure. We show that in $\text{La}_{1.88}\text{Sr}_{0.12}\text{CuO}_4$, charge order consists of twinned stripe domains that are detwinned by uniaxial pressure. Application of magnetic field suppresses partially superconductivity and further enhances the stripe order. The high-field stripe order phase may be an ideal environment for a—yet-to-be-observed—pair-density-wave state.

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