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Using strain to uncover the interplay between two- and three-dimensional charge density waves in high-temperature superconducting YBa₂Cu₃O_y

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Uniaxial pressure provides an efficient approach to control charge density waves in YBa₂Cu₃O_y. It can enhance the correlation volume of ubiquitous short-range two-dimensional charge-density-wave correlations, and induces a long-range three-dimensional charge density wave, otherwise only accessible at large magnetic fields. Here, we use x-ray diffraction to study the strain dependence of these charge density waves and uncover direct evidence for a form of competition between them. We show that this interplay is qualitatively described by including strain effects in a nonlinear sigma model of competing superconducting and charge-density wave orders. Our analysis suggests that strain stabilizes the 3D charge density waves, and that the two orders compete at the boundaries of these domains. No signatures of discommensurations nor of pair density waves are observed. From a broader perspective, our results underscore the potential of strain tuning as a powerful tool for probing competing orders in quantum materials.

A tendency towards charge ordering in the underdoped hightemperature superconducting cuprates was predicted soon after their discovery¹⁻³. However, it took decades of effort with a variety of experimental methods to demonstrate this phenomenon and its ubiquity across the cuprate superconductors⁴⁻¹². This effort has raised many new questions. One observation that lacks a clear explanation is the fact that the competition between charge order and superconductivity is striking in some cuprate families^{9,10,13}, but is less pronounced in others^{11,12,14}. Another is the very substantial variation in correlation length, which varies over two orders of magnitude amongst the cuprates families – from a few unit cells to several tens of nm – without any obvious relation to the superconducting critical temperature T_c . To address these issues, it has proved useful to focus on materials with low disorder scattering, and to probe them with

¹Institute for Quantum Materials and Technologies, Karlsruhe Institute of Technology, Kaiserstr. 12, D-76131 Karlsruhe, Germany. ²4th Physical Institute - Solids and Nanostructures, University of Göttingen, D-37077 Göttingen, Germany. ³Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel. ⁴Karlsruhe Nano Micro Facility (KNMFi), Karlsruhe Institute of Technology, Kaiserstr. 12, D-76131 Karlsruhe, Germany. ⁵ESRF, The European Synchrotron, 71, avenue des Martyrs, CS 40220, F-38043 Grenoble Cedex 9, France. ⁶Max Planck Institute for Solid State Research, Heisenbergstraße 1, D-70569 Stuttgart, Germany. ⁷Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, D-01187 Dresden, Germany. ⁸School of Physics and Astronomy, University of Birmingham, Birmingham B15 2TT, UK. ⁹Present address: Institute for Materials Research, Tohoku University, Sendai 980-8577, Japan. ¹⁰These authors contributed equally: I. Vinograd, S. M. Souliou. ⁵e-mail: matthieu.letacon@kit.edu external tuning parameters, such as magnetic field¹⁰ or pressure¹⁵⁻¹⁷, that do not introduce disorder.

While hydrostatic pressure has been widely used to investigate unconventional superconductors and correlated electron systems, recent studies have highlighted several benefits of uniaxial stress^{18–21}. In the cuprates, uniaxial stress has been used to probe charge stripes in La-based compounds^{22–24}, and to polarize them^{25,26}. Interestingly and in contrast to the introduction of pair-breaking impurities²⁷ or vortices under high magnetic fields¹⁰, uniaxial stress can also be used to homogeneously suppress T_c^{28} . This allows alternative electronic orders to develop, and the interplay between these orders and the superconductivity to be studied with precision.

Uniaxial compression along the *a* axis (the shortest axis of the orthorhombic structure) has been shown to suppress T_c by up to $30\%^{28}$ in YBa₂Cu₃O_{6.67}. In this compound, this results in an enhancement of the amplitude and correlation lengths of an incommensurate, short-range, 2D-correlated charge density wave (hereafter referred to as the 2D CDW)^{29,30}.

Beyond a threshold stress value, a long-range-ordered, 3Dcorrelated CDW (hereafter referred to as the 3D CDW) emerges^{29,30}. 3D CDW order can also be induced by a magnetic field, though to date, with lower amplitude and shorter correlation lengths than the one induced by uniaxial stress^{8,31-35}. The 2D CDW is biaxial, with components that propagate along both the *a* and *b* axes, while the 3D CDW is uniaxial, propagating along the *b* axis only. On the other hand, the inplane wavelengths of the 2D and 3D CDWs are identical^{29,32} and the analysis of the in-plane correlation lengths revealed that the 2D CDW consists of quasi-independent unidirectional orders³⁶. This has further been confirmed by the observation of substantially different dependencies of amplitude and correlation lengths of its *a* and *b* components on uniaxial stress³⁰. Overall, this suggests that the 2D and 3D CDWs might be intimately related.

A key question remains to be clarified: does the 3D CDW emerge from "patches" of 2D CDWs that lock together, or is it a separate order, formed by different charge carriers, whose periodicity locks to that of the background 2D CDWs? Additionally, there is so far little information on where the 3D CDW exists in strain-temperature space, which is crucial information for understanding its interaction with the superconductivity.

Here, we tackle this issue in underdoped YBa₂Cu₃O_v with synchrotron hard x-ray diffraction under uniaxial stress. We show that the 2D CDW amplitude stops growing when the 3D CDW onsets, and map the boundaries of the 3D CDW phase in a strain-temperature phase diagram. We further report that the strain-induced formation of the 3D CDW and its interplay with the 2D CDW and superconductivity are qualitatively well described by including the effects of strain in an extension of a nonlinear sigma model³⁷ previously used to investigate the magnetic-field-induced formation of the 3D CDW order in YBCO^{38,39}. In this picture, the long-range 3D CDW emerges from the regions between disorder-induced 2D CDW domains (i.e., the regions where superconductivity forms in the absence of strain). The growth of disordered 2D CDW domains is then halted by their increased surface tension due to phase-mismatch at the boundary with phase-ordered 3D CDW. Finally, we have also looked for signs of discommensurations, which would indicate a tendency of the CDW to lock to the lattice⁴⁰, and for pair-density wave (PDW) correlations⁴¹ on top of the 3D CDW, but have found no evidence for either.

Results

Hard x-ray diffraction was performed as a function of temperature on uniaxially pressurized. YBa₂Cu₃O_y (YBCO_y) single crystals, as described in the Methods section. We first discuss the effect of uniaxial compression along the *a* and *b* directions on the 2D CDW, for samples with y = 6.67 (corresponding to a doping level $p \sim 0.125$). As shown in the intensity maps in Fig. 1, the *b* component of the 2D CDW

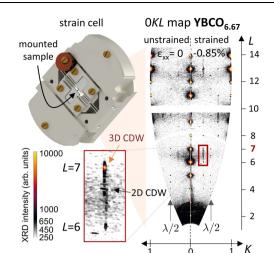


Fig. 1 | Experimental setup and representative results of x-ray scattering under uniaxial stress. Measurements were performed in transmission geometry using a Razorbill CS200T strain cell. The YBCO_{6.67} sample was mounted onto an exchangeable titanium cross. Sections of large (OKL) reciprocal space maps at zero (left) and highest *a* axis compression (right) show the appearance of sharp 3D CDW peaks at integer *L*. Faint 2D CDW intensity in between is present at all strains at half-integer *L* on top of a diffuse phonon background. Grey arrows mark sharp Bragg reflections that appear at k = 0.5 due to higher harmonics with $\lambda/2$ (see Methods section for the wavelength λ). The inset shows an enlarged part of the (OKL) map. To make the 2D CDW more apparent, the colour scale is linear for small and logarithmic for large intensities.

has an extremely broad profile along the [0, 0, L] direction, centered at half-integer L, but is relatively sharp along the [0, K, 0] direction already in the absence of strain. The incommensurate wavevector of the modulations are in perfect agreement with previous reports^{9,10,27,42}. To evaluate the intensity along the K direction, we average the intensity over the range 0.4 < L < 0.6, with results at a few a axis strain values (ε_{xx}) shown in Fig. 2a. For these measurements, the temperature is close to $T_c(\varepsilon_{xx})$. For strains not exceeding the 3D CDW onset strain, the 2D CDW is most intense at $T \approx T_c(\varepsilon_{xx})$. It can be seen in Fig. 2a that the intensity of the b component of the 2D CDW approximately doubles between $\varepsilon_{xx} = 0$ and -0.55% (but the CDW ordering wave vector is only weakly affected by strain). The integrated intensity at a denser set of strains is shown in Fig. 2b, in which this doubling is again visible, along with a reduction in intensity for $\varepsilon_{xx} < -0.55\%$. Also shown in Fig. 2b is the integrated intensity of the b component under compression along the *b* axis ($\varepsilon_{\nu\nu} < 0$). The effect of b axis compression is opposite to that of a axis compression: the intensity shrinks. In Supplementary Fig. 5 we provide evidence confirming the growth of the *a* axis component of the 2D CDW under *b* axis compression, in agreement with previous findings³⁰. Together, these data show that the a- and b-CDWs are quasi-independent order parameters, and emphasize the uniaxial nature of the underlying order parameter^{30,36}.

The decrease in intensity of the 2D CDW for $\varepsilon_{xx} < -0.55\%$ coincides with the onset of the 3D CDW, which manifests itself as a sharp peak at integer *L*, signaling long-range, in-phase correlations along the *c* axis^{32,33}. The 3D CDW intensity is also shown in Fig. 2b. It is worth noting that this onset strain is considerably lower than that estimated in previous x-ray scattering studies^{29,30}, in which the lattice parameter along the direction of compression was not accessible and in which strain could therefore not be determined with sufficient accuracy. It is however closer to the location of an anomaly identified in the stress dependence of T_c in ref. 28. The 3D CDW peak is also seen for y = 6.55 (p = 0.108) (see below), but not yet for y = 6.80 (p = 0.140), where, admittedly, the largest strain achieved in the present work was lower: we reached $\varepsilon_{xx} = -0.45\%$ (see Supplementary Fig. 6).

We turn next to the temperature dependence of the 2D and 3D CDW intensities. Integrated intensities of the 2D and 3D CDWs versus temperature at selected, fixed strains are shown in Fig. 3a for YBCO_{6.67}, and Fig. 3b for YBCO_{6.55}. At both of the selected strains for both

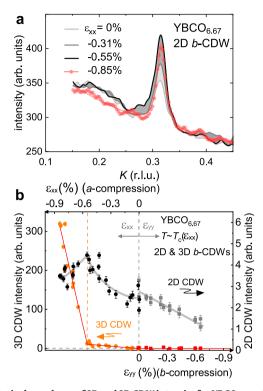


Fig. 2 | **Strain dependence of 2D and 3D CDW intensity for YBCO**_{6.67}. **a** 2D *b*-CDW intensity from *K*-cuts at half-integer *L* values for *a* axis compression. *a* axis compression amplifies the *b*-CDW only up to the onset of 3D ordering. **b** *a* axis and *b* axis compression dependence of the integrated 2D CDW and 3D CDW intensities from *K*-cuts at half-integer *L*=6.5 through the ordering wavevector of the 2D CDW **q**_{2D}. **c**_{DW} (black and grey symbols) and *L*-cuts through the ordering wavevector of the 3D CDW **q**_{3D-CDW} (orange and red symbols). Error bars correspond to standard deviations of Lorentzian (Gaussian) fits. The vertical scales of the integrated intensities (line cuts) at $T - T_c(\varepsilon_{xx})$ correspond to the same total integrated intensity, I_{tot} , based on Supplementary Table 2. The horizontal grey dashed line lies at zero. The orange dashed line marks the onset of long-range 3D CDWs at ε_{xx} =-0.55%.

compositions, the 2D CDW intensity stops increasing (and within experimental accuracy even seems to decrease) when the 3D CDW onsets. At these selected strains, the onset temperature of the 3D CDW exceeds $T_c(\varepsilon_{xx} = 0)$ and so, presuming that *a* axis compression suppresses T_c in YBCO_{6.55} as it does in YBCO_{6.67} (and as inferred from thermal expansion in the zero stress limit⁴³), it substantially exceeds T_c at these strains. Therefore, the end of the 2D CDW growth at these strains is clearly tied to the onset of the 3D CDW, and not to the onset of superconductivity as at $\varepsilon_{xx} \sim 0$.

We summarize our findings in the form of intensity maps of the 3D CDW in strain-temperature space, shown for YBCO_{6.67} and YBCO_{6.55} in Fig. 3c, d, respectively. For YBCO_{6.55} there is no direct measurement of $T_c(\varepsilon_{xx})$, so within the 3D CDW phase we estimate this quantity from the temperature where the 3D CDW signal intensity reaches a maximum, before getting gradually suppressed. This indicates a substantial range of overlap between the 3D CDW and superconductivity. It is, on the one hand, consistent with measurements in high magnetic fields that systematically find the 3D CDW onset field to lie below H_{c2}^{33} , and on the other hand with the 3D CDW signal being observed down to at least 41 K, i.e., well below $T_c(\varepsilon_{xx})$ for YBCO_{6.67} at the highest measured strain of ref. 29 (the present data allow us to re-estimate this value to be $\varepsilon_{xx} = -0.85\%$). For YBCO_{6.55}, the 3D CDW onset lies at a slightly smaller strain, $\varepsilon_{xx} = -0.4\%$, lower than that for YBCO_{6.67}.

We end this section with a description of structural changes induced by the 3D CDW formation which can be quantified, despite experimental limitations on the accessible part of the reciprocal space imposed by the strain cell. Within experimental uncertainty, we observe the main structural changes in the CuO₂-planes as the samples are strained. From structural refinements of YBCO_{6.67} we observe that the clearest response of the average structure (single unit cell) to the 3D CDW onset with increasing strain is a growing anisotropy of the buckling angle of the planar oxygen bonds as shown in Supplementary Fig. 7. Minor changes can be discerned in the parameters of the planar Cu atoms, e.g., in the intra-bilayer distance, but these are less evident than the change observed for the buckling angles of the oxygen bonds. Nevertheless, smaller motions are to be expected for the heavier Cu atoms. This also finds confirmation in the smaller isotropic temperature factors, shown in Supplementary Fig. 8.

Model

In an effort to elucidate the experimental findings we consider a nonlinear sigma model describing competition between

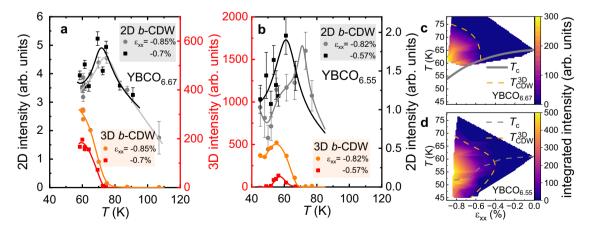


Fig. 3 | Temperature dependence of 2D and 3D CDW intensity for YBCO_{6.67} and YBCO_{6.55}. a Temperature dependence of 3D and 2D CDW intensities for YBCO_{6.67} at *a* axis compressions ε_{xx} = -0.7 % and -0.85 %. b 3D and 2D CDW intensities for YBCO_{6.55} at compressions of ε_{xx} = -0.57 % and -0.82 %. 2D CDW (3D CDW) intensities are determined from *K*-cuts (*L*-cuts). Error bars correspond to standard deviations of Lorentzian fits. Lines are guides to the eye. For a comparison of 2D and

3D intensities, see Supplementary Table 2. **c** Strain-temperature phase diagram of the 3D CDW intensity for YBCO_{6.67} with the T_c curve reproduced from ref. 28. Dashed orange lines mark the boundary of the 3D CDW phase. In the corresponding diagram for YBCO_{6.55} in (**d**) $T_c(\varepsilon_{xx})$ is estimated from the linear extrapolation down to the onset of the 3D phase⁴³. Within the 3D CDW phase $T_c(\varepsilon_{xx})$ is based on the peak in the 3D CDW intensity.

superconductivity and CDW orders^{37–39}. It is formulated in terms of a three-dimensional complex vector whose components correspond to a superconducting order parameter, $\psi_{j\mu}(\mathbf{r})$, and two complex CDW order parameters, $\Phi_{j\mu}^{a,b}(\mathbf{r})$. The latter describe density variations $\delta \rho_{j\mu}(\mathbf{r}) = e^{i\mathbf{Q}_a \cdot \mathbf{r}} \Phi_{j\mu}^a(\mathbf{r}) + e^{i\mathbf{Q}_b \cdot \mathbf{r}} \Phi_{j\mu}^b(\mathbf{r}) + c.c.$, along the *a* and *b* directions with incommensurate wave-vectors $\mathbf{Q}_{a,b}$. The order parameters reside on bilayers that are indexed by *j* and represent the CuO₂ bilayers of YBCO. The index $\mu = 0, 1$ denotes the bottom (top) layer and \mathbf{r} is the inplane position vector. In the following we will coarse grain each of the planes into a square lattice whose lattice constant is the observed CDW wavelength, *i.e.*, about 3 Cu-Cu spacings. The model assumes the existence of some type of local order at every lattice point and incorporates the competition between the different components via the constraints

$$|\psi_{j\mu}|^2 + |\boldsymbol{\Phi}_{j\mu}|^2 = 1, \tag{1}$$

where $\boldsymbol{\Phi}_{j\mu} = (\Phi^a_{j\mu}, \Phi^b_{j\mu})^T$.

1

The vector of order parameters is governed by the Hamiltonian

$$H = \sum_{j} \sum_{\mu=0,1} H_{j\mu} + \frac{\rho_{s}}{2} \sum_{j} \sum_{\mathbf{r}} \left[\tilde{U} \boldsymbol{\Phi}_{j,0}^{\dagger} \boldsymbol{\Phi}_{j,1} + U \boldsymbol{\Phi}_{j,1}^{\dagger} \boldsymbol{\Phi}_{j+1,0} - \tilde{J} \psi_{j,0}^{*} \psi_{j,1} - J \psi_{j,1}^{*} \psi_{j+1,0} + V_{j}^{\dagger} \left(\gamma \boldsymbol{\Phi}_{j,0} + \boldsymbol{\Phi}_{j,1} + \boldsymbol{\Phi}_{j+1,0} + \gamma \boldsymbol{\Phi}_{j+1,1} \right) + \text{c.c.} \right].$$
(2)

Henceforth, the bare superconducting stiffness, ρ_s , is set to 1 and serves as the basic energy scale. We model the Coulomb interaction between CDW fields within a bilayer by a local coupling \hat{U} , and denote the intra-bilayer Josephson tunneling amplitude by J. The (weaker) Coulomb interaction and Josephson coupling between nearest-neighbor planes belonging to consecutive bilayers are denoted by U and I, respectively. In the last term, we include the coupling between the disordered doped oxygens on the chain layers and the CDW fields on the adjacent bilayers. This coupling may originate from local changes in the concentration of doped holes and from the Coulomb interaction between the oxygens and the CDW¹⁴. Hence, we assume that it is reduced by a factor y as one moves from the inner to the outer CuO₂ planes. We model the effective potential induced by the chain layers as a collection of randomly placed discs at \mathbf{r}_{l} , each of radius r_{d} and containing a constant potential with a random phase that couples either to the a or b CDW fields, $V(\mathbf{r}) = V \sum_{l} f(|\mathbf{r} - \mathbf{r}_{l}|) e^{i\theta_{l}} (p_{l}, 1 - p_{l})^{T}$. Here, p takes the values 0,1 with probability 1/2 and $f(\mathbf{r}) = 1 - \Theta(|\mathbf{r}| - r_d)$, where $\Theta(r)$ is the step function.

Within a layer the Hamiltonian reads

$$H_{j\mu} = \frac{\rho_s}{2} \sum_{\mathbf{r}} \left[|\nabla \psi_{j\mu}|^2 + \lambda |\nabla \boldsymbol{\Phi}_{j\mu}|^2 + g |\boldsymbol{\Phi}_{j\mu}|^2 + (\Delta g - \Delta g_s) |\Phi_{j\mu}^a|^2 + \Delta g_s |\Phi_{j\mu}^b|^2 \right],$$
(3)

where ∇ is the discrete gradient, $\lambda \rho_s$ is the CDW stiffness and $g\rho_s$ is the effective CDW mass reflecting the energetic penalty for CDW ordering. The presence of such a penalty ensures that superconductivity prevails over the CDW order at T = 0, at least in the disorder-free regions. The mass anisotropy, encapsulated by the $\Delta g > 0$ term, is included as a result of our assumption that the potential induced by the chain layers favors ordering along the *b* axis. Finally, and most pertinently to the present study, we assume that the application of strain causes an increase in the mass of the CDW component along the direction of the strain, while reducing the mass of the transverse component. Specifically, applying compressive strain in the *a* direction corresponds to $\Delta g_s < 0$.

Theoretical results

Our primary interest lies in the *k*-space (measured from $\mathbf{Q}_{a,b}$) CDW structure factor

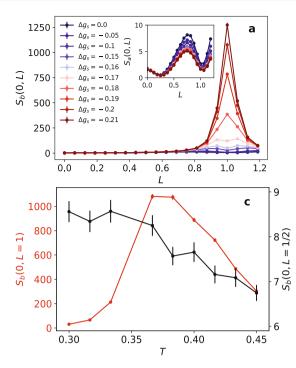
$$S_{\alpha}(\mathbf{q},L) = \frac{1}{N} \sum_{\mathbf{r}\mathbf{r}'} \sum_{jj'} \sum_{\mu\mu'} e^{-i\left[\mathbf{q}\cdot(\mathbf{r}-\mathbf{r}')+2\pi\left(j-j'+\frac{\mu-\mu'}{3}\right)L\right]} \times \langle \Phi_{j\mu}^{\alpha}(\mathbf{r}) \Phi_{j'\mu'}^{*\alpha}(\mathbf{r}') \rangle,$$
(4)

where *N* is the number of lattice points, and the averaging is over both thermal fluctuations and disorder realizations. We have used the fact that in YBCO the CuO_2 planes within a bilayer are separated by approximately 1/3 of the *c* axis lattice constant.

We have calculated $S_{\alpha}(\mathbf{q}, L)$ by Monte Carlo simulations of Eqs. (2),(3)) on a $32 \times 32 \times 32$ (16 bilayers) system, with $\lambda = 1$, g = 1.1, $\Delta g = 0.1$, $\tilde{J} = 0.15$, J = 0.015, $\tilde{U} = 0.85$, U = 0.12, V = 1, and $\gamma = 0.15$. Each data point was averaged over 1000 disorder realizations with 8 disordered regions per bilayer and $r_d = 3$ (The disorder potential in an overlap region between two discs with different disorder orientations was taken to be the sum of the potentials. In the case of two discs with the same orientation the potential in the overlap region was randomly chosen to be one of the two.). Our simulations indicate that the qualitative trends exhibited by the model are largely insensitive to moderate changes in the model parameters. Below we indicate an instance where one can actually improve the agreement with the experimental findings by lowering the disorder strength.

Our core result is presented in Fig. 4a, which depicts the structure factor at a temperature near the superconducting T_c and for various values of Δg_s , emulating the effect of *a* axis strain. For $\Delta g_s = 0$ (absence of strain), both S_a and S_b show broad peaks centered around L = 0.7. This is a result of CDW domains that nucleate due to interaction with the disorder³⁹. Since a disordered region tends to induce the same CDW pattern on both its flanking planes, an out-of-phase arrangement of the CDW order tends to form in the c-direction. The observed skewness away from L = 1/2 is a result of the form factor in Eq. (4). Increasing Δg_s towards negative values increases the energetic cost for nucleating *a*-CDWs and leads to a decrease in the height of the corresponding peak (see inset). An opposite trend is seen for the disorderinduced peak along the b direction. However, a much more dramatic effect emerges in S_h beyond a characteristic value of Δg_s in the form of a large sharp peak centered at L=1. This signal originates from the regions between the disorder-induced CDW domains. It reflects the tipping of the balance between superconductivity and the *b*-CDW order in favor of the latter when the b-CDW nucleation cost is sufficiently reduced. The Coulomb interaction between the strain-induced CDW regions on neighboring bilayers favors an in-phase CDW configuration along the *c*-direction, hence the peak at L = 1. The sharpness of the peak, which corresponds to a correlation length that is of the order of the system size, is a direct result of the fact that these regions are not induced by disorder. The same is true for the in-plane correlation length, as shown in Supplementary Fig. 10.

The interplay between the superconducting and CDW orders is summarized in Fig. 4b, which shows the superconducting T_c and the height of the L = 1 CDW peak as a function of the temperature and Δg_s . Clearly, T_c (calculated by finite size analysis of the superconducting correlation length and renormalized stiffness) is a decreasing function of $|\Delta g_s|$ owing to the enhancement of the competing CDW fluctuations. A large 3D CDW signal appears for $\Delta g_s \leq -0.17$ at a temperature that increases with $|\Delta g_s|$, peaks in the vicinity of T_c of the strained system and then rapidly diminishes at lower temperatures, as is also evident in Fig. 4c. This behavior stands in contrast to that of $S_b(0, L = 1/2)$, which similarly increases when the temperature is lowered towards T_c but then saturates. Nevertheless, Supplementary Fig. 9 demonstrates that a moderate decrease in the disorder strength to V = 0.8 causes also



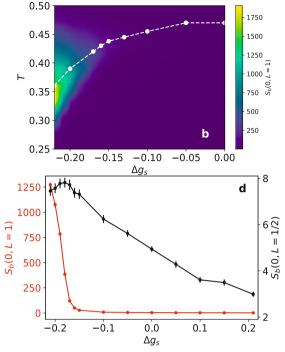


Fig. 4 | **Theoretical CDW structure factors based on a non-linear sigma model. a** The CDW structure factor at the in-plane wave vector of the *b*-CDW peaks as a function of *L* for T = 0.383 and various levels of the change in the CDW-mass, Δg_s , that models the effect of *a* axis compression. The inset depicts the CDW structure factor at the position of the *a*-peak under the same conditions. **b** The intensity of the *L* = 1*b*-CDW peak in the $T \cdot \Delta g_s$ plane. The broken line marks the superconducting

transition temperature. **c** The temperature dependence of the CDW structure factor at the position of the *b*-CDW peaks for L = 1 and L = 1/2 in a system with $\Delta g_{s} = -0.2$. **d** The Δg_{s} dependence of the same quantities at T = 0.383. Lines are guides to the eye. The error bars represent the standard deviation of the estimated quantities calculated using rebinning.

 $S_b(0, L = 1/2)$ to exhibit a peak as a function of temperature, thus recovering the qualitative experimental trend shown in Fig. 3. Supplementary Fig. 9 shows that at the same time the decreased disorder strength does not significantly alter the other aspects of the 2D and 3D CDW signals, and in particular their dependence on strain, which we turn to discuss next.

Figure 4d depicts the Δg_s dependence of the 2D and 3D CDW signals near T_c . There is a clear and sharp onset of the 3D CDW peak at $\Delta g_s \simeq -0.17$, whereas $S_b(0, L = 1)$ essentially vanishes at lower values of *a* axis strain and for all values of *b* axis strain (positive Δg_s). The 2D signal, $S_b(0, L = 1/2)$, on the other hand, diminishes continuously with decreasing *a* axis strain and increasing *b* axis strain. This is because the accompanying increase in the effective mass of the *b*-CDW successively reduces the magnitude of the CDW induced by the disorder. Interestingly, and similarly to the experimental observations, we find a saturation and then a slight downturn of the 2D CDW signal that coincides with the onset of the 3D CDW order. We also note that the model predicts that the in-plane correlation length of the 2D CDW is a very weakly increasing function of the strain, while that of the 3D CDW grows rapidly beyond the onset strain, see Supplementary Fig. 10. Such behaviour is in accord with previous experimental findings²⁹.

Discussion

We start our discussion from the structure of the 3D CDW. Having access to large reciprocal space maps and intense 3D CDW peaks, we can provide strong constraints for deviations from a perfectly sinusoidal charge density modulation in the long-range ordered phase. Previous studies on Bi-based cuprates⁴⁴ have indicated that in this system the CDW is commensurate with periodicity $\lambda = 4a$ over the entire doping range where it appears. More recently, high-field NMR data have been successfully analyzed in a picture where the CDW is locally commensurate with $\lambda = 3b$. In both cases it was argued that sharp phase slips (discommensurations) at the CDW domain boundary could shift the position at which the 3D CDW peak is observed in scattering experiments to an effectively incommensurate value^{40,44}. In this case, one would expect the presence of satellite peaks at 2δ from q_{3D-CDW} (δ being the incommensurability to the commensurate 3D CDW wavevector at k = 1/3)⁴⁰. At T_c , for YBCO_{6.67} we observe no indications for additional satellite peaks along b^* coming from such sharp discommensuration (we cannot fully rule out the presence of spatially extended phase slips). We also note that the K-cut shown in Fig. 5b for YBCO_{6.55} at T = 45 K < T_c does not exhibit a peak at $q_{PDW} = q_{3D-CDW}/2$. Such a peak is expected if the observed CDW is due to a PDW, i.e., a modulated superconducting condensate, at wavevector \mathbf{q}_{PDW} , and if the PDW coexists with uniform superconductivity^{41,45}. Based on our measurements, the PDW signal must be at least 50 times smaller than the signal of the long-range 3D CDW. However, further calculations are needed to estimate the associated periodic lattice displacement and the resulting x-ray intensity to exclude the presence of PDWs.

Next, we briefly discuss our structural results in the context of nematicity, largely associated with the growth of spin and charge-density-wave correlations upon cooling⁴⁶⁻⁵⁰ in the cuprates. In the unstrained orthorhombic unit cell the buckling angles along a and b axes differ only slightly. The increasing trend of the a axis buckling angle under a axis compression (although the error bars are relatively large compared to this subtle effect) is in line with the anisotropic modulation of the copper-oxygen-bonds with significant out-of-plane displacements of the oxygen transverse to the 2D CDW modulation inferred from zero field x-ray diffraction⁵¹, and can be regarded as a structural parameter possibly related to nematicity. The fact that this anisotropy appears maximal in the 3D CDW phase, is also consistent with a larger response of the relative quadrupolar splitting in ¹⁷O-NMR than in ⁶³Cu-NMR when the 3D CDW is induced using high magnetic fields⁴⁰. The buckling anisotropy might therefore be seen as a necessary (but not sufficient) condition for 3D ordering in YBCO, but since the 3D CDW breaks both rotational and translational symmetry

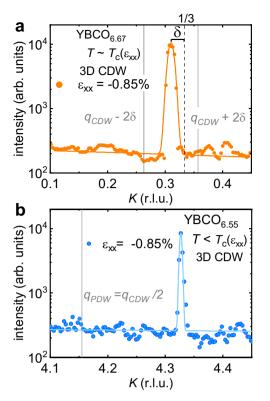


Fig. 5 | *K*-cuts of the strain-induced 3D CDW. a YBCO_{6.67}*K*-cut through the 3D CDW peak at integer *L*, maximal compression and *T* = 60.5 K - *T*_c show an intense peak at incommensurate $K_{CDW} = 0.310(1)$ r.l.u., with δ marking the incommensurability with respect to the closest commensurate value, *K* = 1/3. Vertical grey lines mark where discommensurations would lead to satellite peaks at $K_{CDW} \pm 2\delta$. **b** YBCO_{6.55}*K*-cut through the 3D CDW peak at integer *L*, maximal compression and *T* = 45 K < *T*_c in the superconducting state, where the vertical grey line marks where pair-density waves are expected to produce a peak at $K_{PDW} = K_{CDW}/2 = 0.327(1)/2$ r.l.u. Lines are Gaussian fits on top of a linear background. Note that the width of the peaks in these figures are limited by the experimental resolution (see Supplementary Information).

globally, it should not be considered as nematic⁵². In any event, to better understand the constraints imposed by the crystal structures on the formation of long-range charge order in the cuprates (in particular given the recent reports on the overdoped side^{53,54}), more systematic structural investigations are needed.

We now turn to the nature of the interplay between the CDW orders and superconductivity. On the one hand, a large body of experiments has unambiguously established that the 2D CDW and superconducting orders compete in YBCO^{9,10,42}, as both the CDW peak amplitude and the CDW domain correlation length decrease in the superconducting state. On the other hand, in x-ray scattering, the strongest 3D CDW signals are observed where the homogeneous superconducting phase has been suppressed either by application of a large magnetic field^{32–35} or by uniaxial pressure^{29,30}. In both cases, the maximal achievable strength of the applied perturbations was not sufficient to completely suppress the superconducting state at the lowest temperatures. However, while in the case of strain tuning the low-temperature superconducting phase is homogeneous with no trace of the 3D CDW, the mixed state of the type-II superconductor in a magnetic field contains halos of 3D CDW around vortex cores.

In the experimental data presented above, we have shown that a form of competition can also manifest itself between the 2D and 3D CDW orders, which is best evidenced in the region of the strain-temperature space where superconductivity has been suppressed by strain. Data in unstressed YBCO_{6.67} under applied magnetic field³²⁻³⁵

can also be interpreted in terms of this 2D-3D CDW competition. Specifically, it was shown in ref. 35 that as YBCO_{6.67} is cooled under strong applied field there is a temperature range where the 2D CDW intensity starts to decrease while the 3D CDW keeps increasing. In the same study, a direct conversion of the 2D CDW to the 3D CDW was hypothesized, but the data there do not allow clean disentanglement of the mutual interaction among the 2D CDW, 3D CDW, and superconductivity. Our data clearly indicate that the 2D CDW stops growing and even shrinks at the onset of the 3D CDW, where the superconductivity would have set in, in the absence of strain. In the superconducting state, the evolution of 2D CDWs is complicated by their mutual competition with the 3D CDW and the simultaneous competition with superconductivity.

Finally, we note that the different temperature- and straindependence of the 2D and 3D CDWs can naturally be explained in the framework of the phenomenological model developed above and is rooted in the different mechanisms that are responsible for their respective establishment. The 2D CDW nucleates in domains that are dominated by their coupling to the disorder potential that arises from the doped oxygens. Consequently, these domains continue to host the CDW even when superconductivity sets in within the intervening disorder-free regions. In contrast, the 3D CDW forms in the same intervening regions at large enough strain. By assumption, the strain reduces the energetic cost of the CDW fluctuations, thus allowing them to appear at temperatures that can even exceed the T_c of the unstrained system. In turn, the growth of the 3D CDW correlations contributes, via their competition with superconductivity, to the suppression of T_c . Below T_c the 3D CDW eventually gives way to the superconducting order, which constitutes the ground state of the clean model. If the coupling between the dopants and the CDW does not change appreciably between YBCO_{6.55} and YBCO_{6.67}, then this picture may explain why the 2D CDW is stronger in the latter (see Supplementary Table 2), as it offers more CDW nucleation centers. Nevertheless, the precise interplay between the CDW and the doped oxygens is not fully understood⁵⁵. Within the same picture, the coincident saturation (or shrinkage) of the 2D CDW with the establishment of strong 3D CDW can be traced to phase mismatch between the two types of CDWs. The phase of the 2D CDW within the disordered domains conforms to the local disorder arrangement, whereas the 3D CDW seeks to establish a uniform phase throughout the system. The phase mismatch along the boundaries of the disordered domains gives rise to an effective surface tension, via the CDW elastic term in Eq. (3), that consequently arrests the growth of the 2D CDW domains.

On a broader perspective, we believe that our results highlight the inherent strengths of strain tuning as a powerful approach to investigate and manipulate the intricate interplay between competing orders in quantum materials, hence presenting promising prospects for advancing our understanding of these systems.

Methods

Samples

High-quality single crystals of YBa₂Cu₃O_y with oxygen concentrations $6.55 \le y \le 6.80$ were grown with a flux method and then annealed and detwinned. Their superconducting T_c was measured with SQUID magnetometry⁴². The crystals were then cut and polished to needles with dimensions of $\approx 200 \,\mu\text{m} \times 100 \,\mu\text{m} \times 2 \,\text{mm}$, and were pressurized along their lengths using a Razorbill CS200T piezoelectric-driven uniaxial stress cell mounted in a helium flow cryostat. Information about the samples is summarized in Supplementary Table 1. To enable fast sample exchange, the mounting into the stress cell was done via a flexible titanium support, as displayed in Fig. 1; needles were mounted into these supports in advance, a slow, delicate process, then the supports were mounted into the cell during the beamtime. To achieve higher stresses, the selected samples were further thinned down laterally using a Xenon plasma focused ion beam (PFIB), as described in

Supplementary Note 1. We checked that this procedure did not alter their superconducting properties. The samples were originally cooled under nominal zero-strain conditions – taking into consideration also the thermal response of the device used for the measurements – and then strained at low temperatures.

X-ray diffraction

Hard x-ray diffraction was performed at the ID15B beamline of the European Synchrotron Radiation Facility with a fixed photon energy of 30 keV (λ = 0.413 Å), that enabled us to work in transmission geometry. The scattered photons were collected using a state-of-the-art large area Dectris Eiger2X CdTe 9M hybrid photon-counting detector. Additional experimental details are given in the Supplementary Note 1 and the original data are available under⁵⁶⁻⁵⁸. Rigaku's CrysalisPro software⁵⁹ has been used for data reduction and to obtain reciprocal space maps. Structural refinements of the average unit cell (neglecting oxygen ordering in the chain layer) have been performed using Jana2020⁶⁰. In spite of the large dynamical range of the detector, the very high intensity difference between the CDW features and the main Bragg reflections presents a measurement challenge. To map both features, photon flux was adjusted by varying the undulator gap⁶¹: high photon flux was used to study CDW superstructure peaks, while low photon flux was used to record the lattice Bragg peaks without saturating the detector. Additional details about the structure refinements are given in the Supplementary Note 3.

Strain determination

Strains reported in this paper are derived from the Bragg peak positions, eliminating error from uncertainty in strain transmission to the sample. The determination of lattice parameters was based on integration of about 400 Bragg reflections, a number that was limited by the geometrical constraints imposed by the cryostat and the stress cell, but which is nevertheless sufficient to allow accurate determination of the three lattice constants *a*, *b*, and *c*. Therefore, we also obtain the Poisson's ratios, $v_{ij} \equiv -\varepsilon_{ii}/\varepsilon_{jj}$, shown in Supplementary Fig. 2. Knowledge of the Poisson's ratios is useful both as a reference for other measurements, where the scattering geometry might not allow determination of the longitudinal strain²⁹, and potentially as a thermodynamic probe of possible changes in the electronic structure⁶². Note that throughout this paper we use the engineering definition of strain, in which negative values denote compression.

Data availability

The data reported in this study are available at⁵⁶⁻⁵⁸. The data that support the findings of this study are available from the corresponding author, upon request. Source data are provided with this paper.

Code availability

The code used to generate the Monte Carlo results is included in the Supplementary Information.

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Author contributions

The single crystals used in this study were grown and characterized by Y.L., S.N. under the supervision of Ma. Mi. and B.K.Mi. Me. performed the single crystal XRD and structural refinement at zero strain. The crystal preparation (polishing, FIBing) for the strain cell was done by S.M.S., I.V., H.M. L.N. and K.I. I.V., S.M.S., A.-A.H., T.L., M.F., G.G. carried out the XRD experiment under strain at the ESRF. I.V. and A.-A.H. performed the structural refinement and the fine analysis of the data. Y. C. and D. O. developed the theoretical model and performed the calculations. I.V., S.M.S., D.O., C.W.H. and M.L.T. wrote the paper with input from all coauthors. M.L.T. initiated and supervised the project.

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Competing interests

C.W.H. has 31% ownership of Razorbill Instruments, a company that markets uniaxial pressure cells such as the one used for this study. All other authors declare no competing interests.

Additional information

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