Inhomogeneity of charge–density–wave order and quenched disorder in a high-T_c superconductor

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It has recently been established that the high-transition-temperature (high-T_c) superconducting state coexists with short-range charge-density-wave order4–11 and quenched disorder12,13 arising from dopants and strain14–17. This complex, multiscale phase separation4–8,18–21 invites the development of theories of high-temperature superconductivity that include complexity22–25. The nature of the spatial interplay between charge and dopant order that provides a basis for nanoscale phase separation remains a key open question, because experiments have yet to probe the unknown spatial distribution at both the nanoscale and mesoscale (between atomic and macroscopic scale). Here we report micro X-ray diffraction imaging of the spatial distribution of both short-range charge-density-wave ‘puddles’ (domains with only a few wavelengths) and quenched disorder in HgBa_2CuO_4+y, the single-layer cuprate with the highest T_c, 95 kelvin (refs 26–28). We found that the charge-density-wave puddles, like the steam bubbles in boiling water, have a fat-tailed size distribution that is typical of self-organization near a critical point19,20. However, the quenched disorder, which arises from oxygen interstitials, has a distribution that is contrary to the usually assumed random, uncorrelated distribution12,13. The interstitial-oxygen-rich domains are spatially anticorrelated with the charge-density-wave domains, because higher doping does not favour the stripy charge-density-wave puddles, leading to a complex emergent geometry of the spatial landscape for superconductivity.

Although it is known that the incommensurate charge-density-wave (CDW) order in cuprates (copper oxides) is made of ordered, stripy, nanoscale ‘puddles’ with an average of only 3–4 oscillations, information about the size distribution and spatial organization of these puddles has so far not been available. We present experiments that demonstrate that CDW puddles have a complex spatial distribution and coexist with, but are spatially anticorrelated to, quenched disorder in HgBa_2CuO_4+y (Hg1201). The sample we studied is a layered perovskite at optimum doping with oxygen interstitials (y = 0.12), tetragonal symmetry (P4/mmm) and a low misfit strain14–16. The X-ray diffraction (XRD) measurements (see Methods) show diffuse CDW satellites (secondary peaks surrounding a main peak) at q_{CDW} = (0.23a^*, 0.16c^*) in the b^* = 0 plane and q_{CDW} = (0.23b^*, 0.16c^*) in the a^* = 0 plane (where a^*, b^* and c^* are the reciprocal lattice units) around specific Bragg peaks, such as (108), below the onset temperature T_{CDW} = 240 K (see Fig. 1a). The component of the momentum transfer q_{CDW} in the CuO_2 plane (0.23c^*) in this case is smaller than it is in the underdoped case (0.28a^*)5. The temperature evolution of the CDW-peak profile along a^* (in the h direction; Fig. 1b) shows a smeared, glassy-like evolution for temperatures below T_{CDW}. The CDW-peak intensity reaches a maximum at T = 100 K, followed by a drop associated with the onset of superconductivity at T = T_c. We investigated the isotropic character of the CDW in the a–b plane using azimuthal scans, as shown in Fig. 1c.

Our main result is the discovery of the statistical spatial distribution of the CDW-puddle size and density throughout the sample, which shows an emergent complex network geometry for the superconducting phase. We performed scanning micro X-ray diffraction (SµXRD) measurements (see Methods) to extend the imaging of spatial inhomogeneity previously obtained by scanning tunnelling microscopy (STM)7,9 from the surface to the bulk of the sample and from nanoscale to mesoscale spatial inhomogeneity. Clear evidence of the inhomogeneous spatial distribution of the CDW is provided by the observation of very different CDW-peak profiles collected at different illuminated sample spots (see Fig. 1d) corresponding to spots with ‘large’ and ‘small’ puddles.

We investigated the temperature dependence of CDW domains by recording the CDW-peak intensity and its full-width at half-maximum (FWHM) during cooling from 280 K to 85 K. We collected the data in two different places on the sample corresponding to ‘large’ and ‘small’ CDW puddles. Figure 1e, f shows the temperature dependence of population (intensity), the number of oscillations h_{CDW}/\Delta h_{CDW} (where h_{CDW} and \Delta h_{CDW} are the position and the FWHM of the CDW peak profile in units of a^*, respectively) and in-plane puddle size \xi_a (along the a axis) in large (red filled circles) and small (black filled squares) CDW puddles. The broad phase transition appears to be arrested, as indicated by the size of the CDW puddles \xi_a = 1/\Delta h_{CDW}, which does not diverge below T_{CDW}. This behaviour is typical of low-dimensional systems with quenched disorder. A map representing the spatial organization of the CDW-puddle size is shown in Fig. 1g. The probability density function (PDF) of the in-plane CDW-puddle size \xi_a is shown in Fig. 1h.

The PDF has a long fat tail that extends over an order of magnitude, and is fitted by PDF(\xi_a) = 1/\xi_a^{2+\alpha} \exp\left(-\xi_a/\xi_c\right), where \xi_{CDW} = 2.8 \pm 0.1 is the critical exponent of the puddle-size power-law distribution and \xi_c > 40 nm. Although we can determine that the average size of CDW puddles is 4.3 nm (in agreement with previous work), PDF(\xi_a) has a non-Gaussian shape and rare, larger puddles reaching sizes of 40 nm are detected. Our finding of a fat-tailed distribution for the CDW-puddle size is in agreement with previous results obtained by STM5. Such structures, where spontaneous breaking of both translational symmetry (CDW electronic crystalline phase) and gauge symmetry (superconductivity) coexist, have been called superstripes15. The distribution of the CDW puddles we have found introduces a substantial topological change to the available space for superconductivity.
superconductivity: the current running from a point A to a point B of the material can take different paths (see Fig. 1i) that are not topologically equivalent thus forming an emergent complex hyperbolic geometry.

To investigate the interplay between the CDW puddles and the quenched disorder, we studied the spatial distribution of oxygen defects. The quenched lattice disorder is due to oxygen interstitials (Oi), which form Oi atomic stripes in the HgO layers, in agreement with previous experiments. HgBa2CuO4+δ (ref. 28), like YBa2Cu3O6+δ (refs 15 and 16), shows Tc variations, owing to the effect of the spatial organization of Oi on superconductivity. The average Oi self-organization was detected by high-energy XRD (see Methods). Figure 2a shows the (0 < h < 5, 0 < k < 4) portion of reciprocal space, where there is strong evidence of diffuse streaks running along the a* and b* directions and crossing all the Bragg peaks. Our high-energy XRD data confirm the formation of Oi stripes intercalated between the CuO2 planes, both in the (100) and (010) directions.

The spatial distribution of the intensity of the streaks was obtained by SQUID (see Methods). We measured the reciprocal a*-c* plane (or b*-c* plane) around the (006) Bragg peak in reflection geometry. The Oi stripes in Hg1201 run along the a* (b*) direction with no correlation along the c* direction; therefore, they also lead to streaks on the a*-c* plane. A schematic of Oi atomic stripes is shown in Fig. 2b. In Fig. 2c we show the spatial map of the streak intensity. The picture shows rich (bright yellow) and poor (dark black) regions of Oi stripes. The PDF of Oi-rich regions in Fig. 2d is fitted by PDF(1) = (1/Ip)−αOi exp(1/Ip), where Ip is the average intensity, αOi = 2.0 ± 0.1 is the critical exponent and Ip > 20.

In Fig. 3 we present results on the spatial interplay between CDW-rich regions and Oi-rich regions. We calculated the ‘difference map’ (see Methods) between CDW peaks and Oi diffuse streaks. The poor CDW regions on the CuO2 basal plane correspond to Oi-rich regions on the HgO layers, as illustrated in Fig. 3a. The CDW puddles and Oi-rich regions give rise to the positive and negative peaks, respectively, in the surface plot shown in Fig. 3b. The spatial anticorrelation is evident from the scatter plot of Oi intensity versus CDW intensity (Fig. 3c). As Oi intensity increases, the CDW intensity decreases, and vice versa. This is consistent with the fact that excess Oi means higher doping, and high doping does not favour stripy, underdoped short-range CDW order. Figure 3d shows the two maps obtained via the segmentation of the difference map, and provides a direct image of how doping-poor (CDW-rich regions are shown in red) and doping-rich (Oi-rich regions are shown in blue) phases are arranged in different regions of the material. Figure 3e illustrates the nanoscale configuration of CDW-puddles (red spots) in the CuO2 plane using Gaussian fits.

The CDW peak intensity as a function of temperature, at the two different places on the sample corresponding to large (red filled circles, right axis) and small (black filled squares, left axis) CDW puddles. The dashed line corresponds to T = Tc and the dotted line to T = TCDW. Evolution of the number of CDW oscillations (hCDW/ΔHCDW) inside a CDW puddle and the CDW domain size along the a axis (ξa) (g). Spatial map (g) and probability density function (h) of the CDW-puddle size. Scale bar in g, 10 μm. A schematic of non-equivalent paths, running in the interface space between CDW puddles, connecting point A to point B in the emergent complex non-Euclidean spatial geometry for the superconducting current.
the experimental distribution of CDW size; this distribution generates 'holes' in the space available for the free electrons (light blue area). This space is topologically interesting: there are an infinite number of ways for a current path to connect a point A to a point B around the CDW puddles, which are not only distinguished by the number of times a path goes around a single hole, but also by the way the path passes through the pattern of CDW puddles. The complex space that emerges from the mesoscopic phase separation, both in the spacer layers and in the CuO$_2$ plane, substantially changes (1) the dielectric constant that controls the long-range Coulomb interaction that is relevant for phase separation near a Lifshitz transition, (2) the dielectric constant that is relevant to electron-electron interaction in the

Figure 2 | Correlated quenched disorder due to O$_i$ atomic stripes in Hg$_{1201}$. a, A portion of the h–k diffraction pattern. Resolution-limited streaks connect the Bragg peaks, owing to the formation of O$_i$ stripes in the HgO$_y$ spacer layers. b, Schematic representation of the atomic O$_i$ stripes. c, $\Sigma$XRD map of a region of a showing the relative O$_i$ streak intensity $I_{O_i}$. The bright (dark) spots correspond to sample regions with a high (low) density of O$_i$ atomic stripes, called O$_i$-rich (poor) regions. Scale bar, 10 $\mu$m. d, Probability density function calculated from the O$_i$-streaks intensity map.

Figure 3 | Spatial anticorrelation between CDW-rich and O$_i$-rich regions. a, The CDW-rich regions (red) on the CuO$_2$ planes and O$_i$-rich regions (blue) on the HgO$_y$ layers. b, Surface plot of the difference map (see Methods) between the CDW-peak and O$_i$-streak intensity. The positive (green to red) values indicate the CDW-rich regions and the negative (green to blue) values correspond to O$_i$-rich regions. Scale bar, 5 $\mu$m. c, Scatter plot of O$_i$ versus CDW intensity demonstrating the negative correlation between CDW-puddle and O$_i$-stripe populations. d, Segmentations of the difference map in b highlighting the network of CDW-rich domains (left panel) and O$_i$-rich regions (right panel). Scale bar, 10 $\mu$m. e, A schematic of the nanoscale texture formed by CDW-rich regions (red spots) and the 'charge-O$_i$-rich' region (light blue area), which define an interface space and loci of the superconductivity with a complex non-Euclidean geometry.
pairing and (3) the geometrical and topological properties of the space that is available for the overall phase coherence of the macroscopic quantum condensate that is made up of multiple condensates at the nanoscale with a single critical temperature\textsuperscript{20,21}.

This work offers new insight into the complexity of nanoscale phase-separation phenomena in high-temperature superconductors. More generally, our results deal with the effects of quenched disorder in phase transitions. A phase transition that would be first order in the clean limit gets smeared into a continuous-looking transition in the presence of a random, Gaussian distributed, quenched disorder. Even in the ‘ideal’ single-layer cuprate superconductor HgBa\textsubscript{2}CuO\textsubscript{4+δ} at optimum doping (\(T_c = 95\) K), the CDW order self-organizes into puddles, forming an inhomogeneous landscape with an emergent complex network geometry. Our results provide further evidence for the universality of mesoscale phase separation even in the most optimized superconducting cuprates, which implies that the superconductivity will be non-uniform throughout what is a granular medium.

Online Content Methods, along with any additional Extended Data display items and Source Data, are available in the online version of the paper; references unique to these sections appear only in the online paper.

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Author Information Reprints and permissions information is available at www.nature.com/reprints. The authors declare no competing financial interests. Readers are welcome to comment on the online version of the paper. Correspondence and requests for materials should be addressed to A.B. (antonio.bianconi@ricmass.eu).
METHODS

Sample preparation and characterization. The HgBa2CuO4 + y (Hg1201) crystal with y = 0.12, grown at ETH”, has a sharp superconducting transition at $T_c = 95$ K. The crystal structure has $P4/mmm$ symmetry with lattice parameters $a = b = 0.387480(5)$ nm and $c = 0.95078(2)$ nm at $T = 100$ K (numbers in parentheses indicate the standard deviation of the last digit).

XRD measurements using the XRD1 beamline. To identify the CDW order in a single Hg1201 crystal we used XRD using the XRD1 beamline at the Elettra synchrotron radiation facility in Trieste, Italy, tuning the photon energy between 13 keV and 16 keV with a beam size of 200 × 200 μm². Only selected reflections show clear CDW satellites, in agreement with ref. 2. We focused on the CDW satellite located at $q_{CDW} = (0.23, 0, 0.16)$ around the (108) Bragg reflection, which appeared as the sample was cooled below 240 K. Typical diffraction patterns collected at 85 K, 105 K and 280 K are shown in Fig. 1a. To get a direct view of the temperature dependence of the CDW-satellite reflection for $T = 280–85$ K, a two-dimensional colour plot of the CDW-peak profile along the $a^*$ direction as a function of temperature is shown in Fig. 1b.

High-energy XRD measurements using the BW5 beamline. High-energy XRD measurements were collected using the BW5 beamline at DESY, Hamburg, Germany, using a transmission geometry and an X-ray energy of 100 KeV. High-energy XRD measurements using the BW5 beamline.

SpXRD measurements using the ID13 beamline. SpXRD experiments were performed in reflection geometry using the ID13 beamline at ESRF, Grenoble, France. We applied incident X-ray energy of 13 KeV. By moving the sample under a 1-μm focused beam with an x–y translator, we scanned a sample area of 65 × 80 μm², collecting 5,200 different diffraction patterns at $T = 100$ K. For each scanned point of the sample, the $q_{CDW}$-peak profile was extracted; the FWHMs along the $a^*(b^*)$ and $c^*$ directions were evaluated to obtain the domain size of the charge-ordered regions along the $a(b)$ and $c$ crystallographic axes.

Two quite different CDW-peak profiles in the same crystal measured along the $a^*(b^*)$ direction in the $a^*–c^*(b^*–c^*)$ plane are shown in Fig. 1d. Here we show two typical profiles collected at two different spatial locations in the same crystal corresponding to large (red circles) and small (black squares) puddles. The continuous lines are the Gaussian fits to the data. The different amplitudes and FWHMs (0.033 ± 0.003)$a^*$ and (0.089 ± 0.002)$a^*$ in the upper and lower panels of Fig. 1d, respectively, errors indicate standard deviation) of the two peaks, which correspond to large and small CDW puddles, provide evidence of a strong inhomogeneity in the CDW spatial distribution. The peak profiles do appear the same along the $a^*$ and $b^*$ directions, confirming the peak isotropy in the basal plane of the tetragonal lattice. The intensity of the CDW satellites as a function of temperature, measured at two different locations on the sample corresponding to large (red) and small (black) CDW puddles are shown in Fig. 1e. The vertical lines represent the superconducting temperature $T_c$ and the CDW onset temperature $T_{CDW}$. The order–disorder transition is very broad, which indicates the role of the quenched disorder owing to the presence of defects. Moreover, the CDW intensity shows a clear drop around $T_c$ that appears to depend on the CDW puddle size. The temperature dependence of the number of CDW oscillations inside a single puddle ($n_{CDW} / n_{CDW}^*$) and the domain size of a single puddle along the $a(b)$ axis ($\xi_{a(b)}$) are shown in Fig. 1f. ($\Delta n_{CDW} / n_{CDW}^*$ and $n_{CDW}^*$ are the FWHM and the location along $a^*$ of the CDW peak; the domain size along the $a$ axis ($\xi_{a}$) is given by the correlation length $\xi_{a}$). The inhomogeneity of the CDW distribution is depicted in the 65 × 80 μm² XRD map of the (nanoscale) size of CDW domains in Fig. 1g. This figure shows loci of large (red–yellow area) and small (blue area) CDW puddles. The scale bar corresponds to 10 μm. Using the ID13 microfocus beamline at ESRF, we can also detect the spatial distribution of the quenched disorder. Figure 2c shows the SpXRD map of the integrated intensity of the streaks of Oi stripes. The bright (dark) spots correspond to sample regions with a high (low) density of Oi atomic stripes, called Oi-rich (poor) regions. The scale bar is 10 μm. Figure 2d shows the PDF of the Oi-streak intensity that was obtained from the SpXRD map. This plot shows the probability distribution of the Oi-rich regions. The experimental set-up allows us to investigate the spatial interplay between CDW puddles in the CuO2 plane and Oi-rich domains in the HgO layers, shown in Fig. 3. We measured the ‘difference map’ ($I_{CDW} – I_{Oi}$), where $I_{CDW}$ and $I_{Oi}$ are the intensities of the $q_{CDW}$ peak and the Oi diffuse streaks, respectively, normalized to [0, 1]. The surface plot of this difference map is shown in Fig. 3b. The positive (green to red) peaks indicate CDW–puddle–rich regions and the negative (green to blue) peaks indicate Oi-rich regions. The spatial anticorrelation between CDW puddles and Oi-rich regions is obtained by segmentation of the difference map. We use this segmentation to visualize the phase separation owing to the network of CDW-rich domains, which correspond to ‘charge poor’ domains in the CuO2 planes (left panel of Fig. 3d), and Oi-rich regions in the HgO layers, which correspond to ‘charge rich’ portions of the CuO2 plane (right panel of Fig. 3d).

Code availability. The code we used for statistical analysis of the SpXRD data is not currently available (G.C., A.R. and A.B., manuscript in preparation).