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Networks of superconducting nano-puddles in 1/8 doped $\text{YBa}_2\text{Cu}_3\text{O}_{6.5+y}$ controlled by thermal manipulation

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Abstract

While it is known that the nature and the arrangement of defects in complex oxides have an impact on the material functionalities, little is known about control of superconductivity by oxygen interstitial organization in cuprates. Here we report direct compelling evidence for the control of T_c by manipulation of the superconducting granular networks of nanoscale puddles, made of ordered oxygen stripes, in a single crystal of $\text{YBa}_2\text{Cu}_3\text{O}_{6.5+y}$ with average formal hole doping p close to 1/8. Upon thermal treatments we were able to switch from a first network of oxygen defect striped puddles with OVIII modulation ($q_{\text{OVIII}}(a^*) = (h + 3/8, k, 0)$ and $q_{\text{OVIII}}(a^*) = (h + 5/8, k, 0)$) to a second network characterized by OXVI modulation ($q_{\text{OXVI}}(a^*) = (h + 7/16, k, 0)$ and $q_{\text{OXVI}}(a^*) = (h + 9/16, k, 0)$)



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(a^*) = ($h + 9/16, k, 0$) and finally to a third network with puddles of OV periodicity ($q_{OV}(a^*) = (4/10, 1, 0)$ and $q_{OV}(a^*) = (6/10, 1, 0)$). We map the microscopic spatial evolution of the out of plane OVIII, OXVI and OV puddle nanosize distribution via scanning micro-diffraction measurements. In particular, we calculated the number of oxygen chains (n) and the charge density (hole concentration p) inside each puddle, analyzing areas of $160 \times 80 \mu\text{m}^2$, and recording 12 800 diffraction patterns to reconstruct each spatial map. The high spatial inhomogeneity shown by all the reconstructed spatial maps reflects the intrinsic granular structure that characterizes cuprates and iron chalcogenides, disclosing the presence of several complex networks of coexisting superconducting domains with different lattice modulations, charge densities and gaps as in the proposed multi-gap scenario called superstripes.

Keywords: phase separation, inhomogeneity, cuprates, high temperature superconductivity, complex materials

1. Introduction

An essential step towards the understanding of modern materials and their implementation in novel nano-electronic devices is the control and manipulation of their microscopic behavior [1–3]. Recently, the interrelationship between spin, charge, and lattice orders in high temperature superconductors (HTSs) has been at the center of a very animated discussion [4–15]. Novel results obtained in $\text{YBa}_2\text{Cu}_3\text{O}_{6+y}$ (YBCO) provide compelling evidence for charge density waves (CDWs), and static magnetic stripes are intertwined and aggregated in nanoscale puddles [16–21]. These domains are spatially separated by superconducting regions composed by ordered lattice stripes [22–26] forming an intrinsically complex lattice of striped puddles called the ‘superstripe’ scenario [13]. In this scenario the local lattice modulations determine multiple subbands crossing the Fermi level and therefore multi-gap superconductivity below the critical temperature [27, 28]. This theoretical proposal has been recently supported by the prediction of the anomalous isotope coefficient at 1/8 doping [29]. Since the nature and distribution of defects becomes the driving force, their control by thermal annealing is of high relevance. Therefore, a primary task for both fundamental physics and novel nano-electronics is the careful visualization of the effects of a thermal treatment on the system, via imaging the quasi-two-dimensional puddles of oxygen chains in YBCO. Unfortunately, due to the lack of proper local bulk-sensitive probes, the microscopic scenario is still not clear and the real-space and real-time observation of thermally induced rearrangements of superconducting micro-regions in HTS is a difficult experimental task.

The development of a technique for imaging the nanoscale intrinsic inhomogeneity of oxygen chain organization is the first step to open new opportunities for their manipulation. Here we explore the nanoscale granular patterns arising in a $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ single crystal with doping close to 1/8 hole content per Cu site in the $\text{Y}(\text{CuO}_2)_2$ bilayer. The $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ crystal is an ideal system for investigation of inhomogeneity due to the short range ordering of oxygen ions [30] at 1/8 doping. Indeed, it exhibits an incommensurate superlattice of (OVIII) chains indicated by the lattice superstructure modulation at $q_{OVIII}(a^*) = (h + 3/8, k, 0)$ and $q_{OVIII}(a^*) = (h + 5/8, k, 0)$. Although the oxygen tendency to form O–Cu–O fragments in the

basal plane has been widely investigated [31, 32], to date there is little information on the spatial distribution of these fragments, and on their inclination towards aggregation in domains.

In the first part of our paper we report the temperature evolution in $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ using standard synchrotron x-ray diffraction of the OVIII puddles of oxygen chains. Using the heat treatment we show the formation of a second network of puddles characterized by OXVI superstructure modulation at $q_{\text{OXVI}}(a^*) = (h + 7/16, k, 0)$ and $q_{\text{OXVI}}(a^*) = (h + 9/16, k, 0)$, and finally a third network of OV puddles characterized by the superstructure wavevectors $q_{\text{OV}}(a^*) = (4/10, 1, 0)$ and $q_{\text{OV}}(a^*) = (6/10, 1, 0)$. The measurements provide us with information about the average order of oxygen chains in the sample. In the following, we investigate the dynamics and spatial distribution of OVIII, OXVI and OV domains upon thermal cycling via micro-x-ray diffraction (μXRD). By scanning microscopic areas, this technique provides mixed information of the reciprocal and real-space the bulk structure inhomogeneities, and it has never been applied on a $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ single crystal before. Finally, we show how through thermal treatment it is possible to control the puddle size distribution, the number of oxygen chains and their charge density. Indeed using the novel experimental method μXRD it has been possible to directly visualize how the thermal treatment affects the intrinsic nanoscale phase heterogeneity in this cuprate superconductor. We relate these changes to the onset variation of the superconducting temperature, that we readily can control over a range of 2 K.

2. Materials and methods

Starting reagents with ultra-high purity (CuO and Y_2O_3 99.999%, BaCO_3 99.997%) have been employed to grow extremely good single crystals of $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ in barium zirconate crucibles by the self-flux technique [33]. Inductively coupled plasma mass spectroscopy indicates a purity of the crystals higher than 99.99 at.%. The oxygen content of the crystal was set to 6.67 by annealing in flowing oxygen at 914 °C, followed by quenching to room temperature under flowing nitrogen gas. The macroscopic oxygen content inhomogeneity was removed by annealing the crystal at 570 °C in a sealed quartz capsule, followed by quenching in an ice–water bath. The crystal was then kept at room temperature to let the short range oxygen order establish. Average structure characterization shows a unit cell described by a $P4/m$ spatial symmetry and lattice dimensions $a = 3.807(11)$ Å, $b = 3.864(12)$ Å and $c = 11.52(2)$ Å, with a unit cell volume of $169.5(8)$ Å³.

Temperature dependent x-ray diffraction measurements were made on the XRD1 beamline at the ELETTRA synchrotron storage ring, Trieste. The beamline is placed on a multipole wiggler insertion device operating under the current ELETTRA conditions of 2 GeV ring energy and 400 mA injection current (see figure S1). The samples were oriented on a kappa diffractometer equipped with a motorized goniometric X – Y stage head and a Mar-Research 165 mm CCD camera as detector. The data were collected in transmission mode with a photon energy of 20 keV ($\lambda = 0.61992$ Å), selected from the source by a double-crystal Si(111) monochromator and using a beam of 200×200 μm^2 . The 2D CCD detector (MAR-Research) was placed at a distance of 70 mm from the sample. Data from a LaB_6 standard were collected for calibration. Measurements were conducted between 300 and 400 K with a temperature step of 2 K for both the heating (300–400 K) and the cooling (400–300 K) cycles. Temperature was varied and controlled by means of a 700 series Oxford Cryosystems cryo-cooler that allows

working in a range of 90–400 K, guaranteeing accuracy better than ± 1 K. All the images measured by single-crystal x-ray diffraction were processed using the FIT2D program jointly with a MATLAB® based software package developed in house. Second, the microscopic behavior of the sample under the thermal treatments has been investigated using scanning μ XRD in reflection geometry (in the a – c plane).

The ID13 beamline of the European Synchrotron Radiation Facility (ESRF) is specialized in the delivery of micro-focused x-ray beams for x-ray diffraction experiments. The photon source, in the range 12–13 keV, is an 18 mm period in-vacuum undulator at the ESRF 6.03 GeV storage ring operated in multi-bunch mode with a current of 200 mA. The optics of the micro-focus beamline include compound refractive lenses, Kirkpatrick Baez (KB) mirrors, crossed Fresnel zone plates or waveguides. The ellipsoidal mirror is the main focusing element, demagnifying the source by a factor of 10 (about 40 μ m in diameter). The focused beam is defined by a pinhole of 5 μ m diameter. The beam is focused by a tapered glass capillary to 1 μ m in diameter. The beamline uses two monochromators positioned in series: the first is a liquid N₂ cooled Si-111 double crystal or Si-111 (bounce); the second is a channel cut monochromator employing a single liquid nitrogen cooled Si crystal. The detector of x-ray diffraction images is a high resolution CCD camera (Mar CCD) with point spread function 0.1 mm, 130 mm entrance window and 16 bit readout placed at a distance of about 90 mm from the sample. To scan the sample area it has been moved using two piezoelectric stages in x – y directions and data have been collected in the θ – 2θ reflection geometry (see figure S2). The huge number of diffraction patterns collected by μ XRD (more than 12 800 for each map) have been processed using a MATLAB® based software package developed in house.

The magnetic behavior versus temperature of our YBCO sample at 1/8 has been characterized by means of the vibrating sample magnetometer (VSM) option in a Physical Property Measurement System (PPMS 6000) from Quantum Design. Here a linear motor vibrates the sample with a frequency of 40 Hz and amplitude of 2 mm at the center of a pick-up coil, and the induced voltage is measured synchronically with the oscillation. A magnetization measurement has been chosen to consist of the averaged value over 40 data points, that is, over a period of 1 s of oscillation. These parameters guarantee a good signal to noise ratio.

3. Results and discussion

The ordering process of the oxygen ions in chain domains has been studied using transmission XRD. The diffraction pattern due to the superlattice reflections was recorded at room temperature and is shown in figure 1(a).

It is possible to distinguish superlattice peaks due to the OVIII (6/16, 1, 0) and (10/16, 1, 0) reflections in the a^* – b^* plane at 300 K, in agreement with previous data [21], which confirms the high quality of our single crystal. We monitored the x-ray diffraction patterns during a thermal cycle from 300 to 400 K, and then back to 300 K, using a slow rate of 0.5 K min^{−1}. The continuous evolution of the superlattice profiles from OVIII to OV through OXVI modulation is shown in figure 1(b). The OVIII (6/16, 1, 0) and (10/16, 1, 0) reflections in the a^* – b^* plane at 300 K evolve into the OXVI (7/16, 1, 0) and (9/16, 1, 0) modulation upon heating to 400 K.

During the subsequent cool-down to 300 K the original OVIII modulation is not restored and a new OXVI phase, identified by the superlattice peaks (7/16, 1, 0) and (9/16, 1, 0), sets in. In figures 1(b)–(e) we report respectively the temperature dependence of the superlattice peak

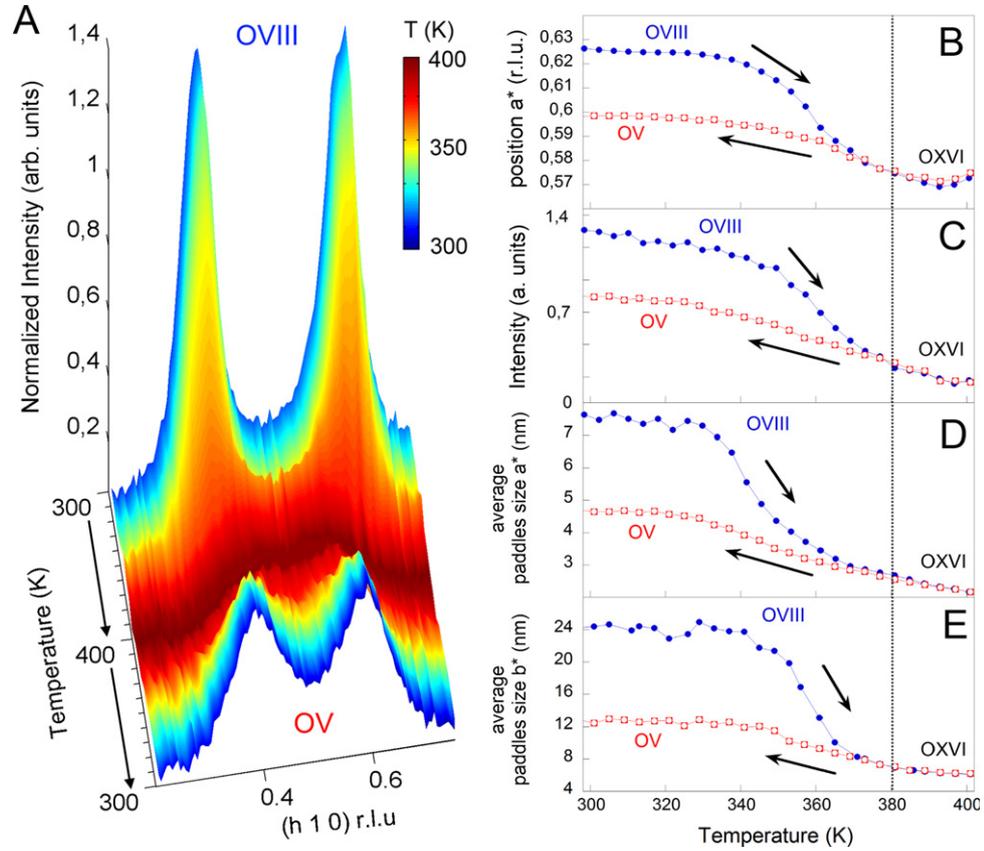


Figure 1. (a) Three dimensional color plot of the thermal evolution of the OVIII oxygen stripe superlattice profile at wavevectors $q_{\text{OVIII}}(a^*) = (3/8, 1, 0)$ and $q_{\text{OVIII}}(a^*) = (5/8, 1, 0)$ into the OV superstructure at wavevector $q_{\text{OV}}(a^*) = (4/10, 1, 0)$ and $q_{\text{OV}}(a^*) = (6/10, 1, 0)$. The temperature ranges from 300 to 400 K, which respectively correspond to dark blue and red colors. The reflections of the superstructures in the $\text{YBa}_2\text{Cu}_3\text{O}_{6+y}$ crystal are measured using a large ($200 \times 200 \mu\text{m}^2$) beam at the ELETTRA storage ring. (b) The thermal cycle of the superlattice position along the a^* crystallographic direction, (c) the intensity, (d) the average puddle size along a^* and (e) along b^* . The arrows show the temporal direction of the experiment. The blue filled circles show the evolution from room temperature to 400 K. The red empty squares show the evolution from 400 K to room temperature. The black dotted line indicates the temperature of 380 K, once crossed the OVIII order can be recovered only by waiting a few days at room temperature.

position, the intensity and the average domain size along a^* and b^* . The average domain size along the in-plane a^* and b^* directions has been calculated by fitting the superlattice reflections with a two-Lorentzian model. In addition, above 380 K the system crosses a OXVI phase with wavevector $q_{\text{mix}} = (h \pm 9/16, k, 0)$. The average size of the OVIII domain along a^* is about 7.5 (5) nm, but approaching the OV it decreases to 4.5(5) nm. The average size is larger along b^* , showing OVIII and OV domains of about 23.5(5) and 11.5(5) nm, respectively. This phenomenology shows that the in plane average domain size of oxygen chains can be controlled and manipulated by tuning the temperature in a quasi-irreversible manner. Leaving the sample under vacuum at room temperature, more than one month is needed for the OV phase to spontaneously drive itself back to a new reconstructed OVIII phase.

This aspect shows relevant analogies with the oxygens and local lattice distortions ordering in $\text{La}_2\text{CuO}_{4+y}$, where the Q2-iO phase (due to interstitial oxygen ordering) and the Q3-LLT (due to local lattice distortion ordering) can be changed by thermal treatments and restored by waiting a long time or by the use of x-ray continuous illumination [34–39].

In order to monitor the microscopic evolution of OVIII domains into OXVI and OV during the thermal manipulation, we used μXRD as already successfully done to investigate phase separation appearing in other cuprates [34–39] and iron-based superconductors [40–43]. We made our measurements using a beam about $1\ \mu\text{m}$ in diameter. The sample has been scanned over an area of $160 \times 80\ \mu\text{m}^2$ collecting 12 800 micro-diffraction patterns showing superlattices at $q_{\text{OVIII}}(a^*) = (3/8, 0, 4)$ and $q_{\text{OVIII}}(a^*) = (5/8, 0, 4)$ due to OVIII puddles of ordered oxygen chains. The superlattice peak profiles have been fitted using a two-Lorentzian model, and the satellite position h_{XY} along a^* and the FWHM_{XY} along a^* and c^* have been extracted for every micro-diffraction pattern at the X – Y position.

From the FWHM_{XY} along c^* we calculated the out of plane domain size for every spot of the scanned map. Figures 2(a)–(c) show the real space map of the out of plane domain size (along c^*), before the heating (OVIII), at 390 K (OXVI) and after the cooling cycle (OV), respectively. These maps show the presence of clear intrinsic nanoscale heterogeneity in this cuprate superconductor, as has been observed in iron chalcogenides [40, 42].

The OVIII probability density functions for the domain size distribution along c^* is relatively sharp, and varies from 7 to 9 nm. Upon heating, the OXVI phase shows instead a very broad distribution, that spans the 1–6 nm range. After the cooling cycle, the size distribution in the OV phase sharpens again, around the decreased average value of 3 nm, therefore indicating a certain recovered order. The overall behavior indicates a sensible response of the granular network to thermal treatment.

Starting from the FWHM_{XY} and the h_{XY} along a^* , measured at each X – Y spatial position, we reconstructed the spatial maps of the number of oxygen chains (n) inside the puddles. Using the expression $n_{XY} = (1 - h_{XY}) / \text{FWHM}_{XY}$, we calculate this quantity for OVIII (figure 3(a)), OXVI (figure 3(b)) and the annealed OV puddles (figure 3(c)).

The number of oxygen chains decreases after the thermal heating cycle from OVIII to OXVI and increases again after the cooling in the OV puddles (but remains lower than in the OVIII phase).

Figures 4(a)–(c) show the spatial map of the charge density (or hole concentration p) inside the puddles of oxygen chains, onto the same area as described in figure 3, before heating (OVIII phase), at 390 K (OXVI phase) and after cool-down (OV phase). The charge density has been calculated considering the difference of the superlattice position along a^* (h_{XY}) with respect to the ortho-II (OII) phase following the relationship $p_{XY} = h_{XY} - 0.5$, where the OII modulation corresponds to a periodicity of a filled CuO chain intercalated by one empty chain. During the thermal annealing process the charge density changed on the microscopic scale and its distribution in the OXVI and OV puddles gets broader, demonstrating the strong granularity of the system. Figure 5(a) shows the spot to spot charge density (p_{XY}) as a function of the number of oxygen chains (n_{XY}), inside the OVIII, OXVI and OV puddles. The behavior has been fitted using an exponential model: $p_{XY} = 1 - \exp\{-[(n_{XY} - n_0)/n_0]\} / \xi_n$. Here n_0 and ξ_n are the minimum and the maximum number of chains present in the average puddle. In order to understand how the microscopic reduction of the effective hole doping affects the superconducting properties, we studied the magnetic response of our sample across the superconducting transition before

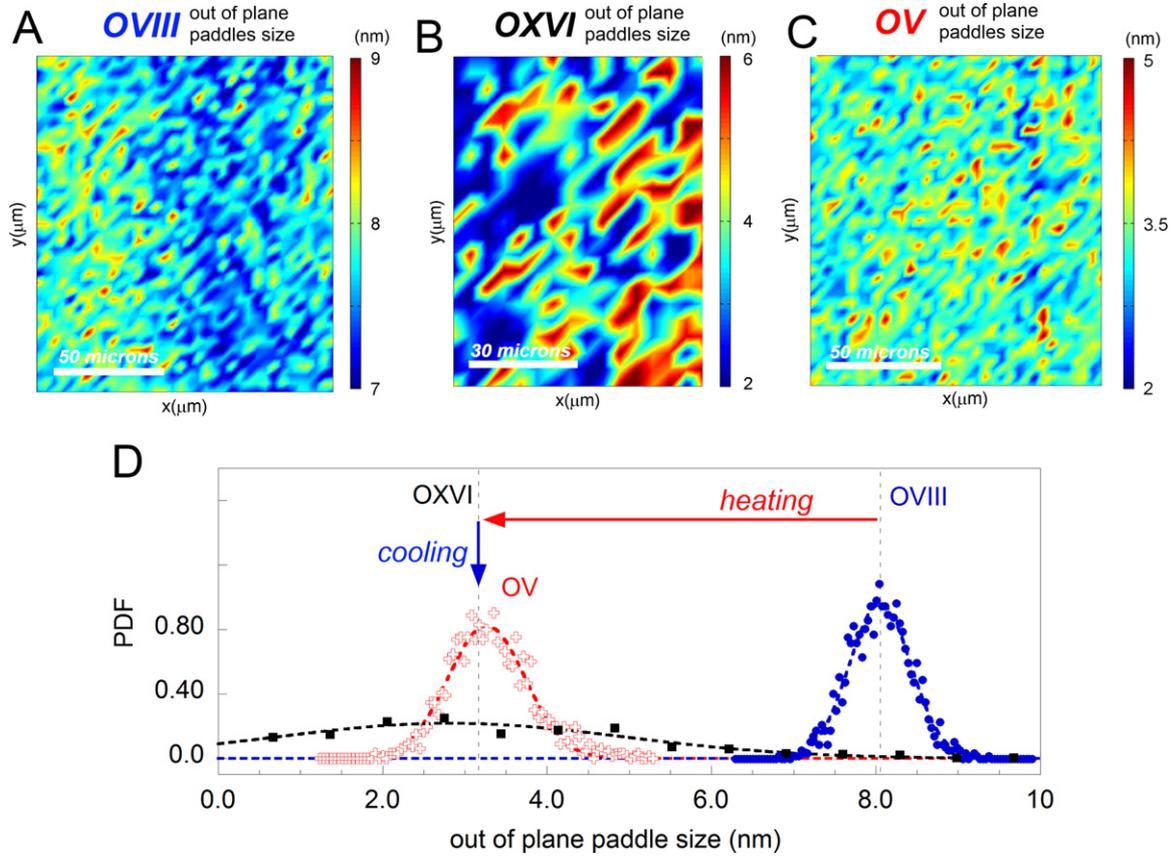


Figure 2. The out of plane puddle sizes (puddle sizes along c^*) before the heating cycle (a), at 390 K (b) and at the end of the cooling cycle (c) in the OVIII, OXVI and OV phases, respectively. (d) The probability density function of the OVIII (blue filled circles), OXVI (black filled squares) and OV (red empty crosses) out of plane puddle sizes. The out of plane puddle sizes decrease after the thermal heating cycle from OVIII to OXVI, where their distribution looks very broad. After the cooling cycle the average out of plane puddle sizes remain the same in the OV phase. On the other hand, the shape of the distribution becomes much sharper, indicating a certain recovered order. Dotted lines are guides for the eyes.

and after the thermal annealing, i.e. in the OVIII and in the OV phase, by means of the VSM option in a PPMS 6000 from Quantum Design. The results are shown in figure 5(b). A first measurement is made before any annealing procedure is carried out (OVIII phase), and shows the onset temperature of the diamagnetic screening (T_c) to be about 66 K. The subsequent measurements are made upon thermal annealing at 380 K (OV phase), with increasing dwell time of 30, 60, 90 and 150 min. In all these cases T_c decreases by about 2 K. This effect is irrespective of the annealing time, and we associate such a reduction with the lower effective charge density we point out in figures 4 and 5(a).

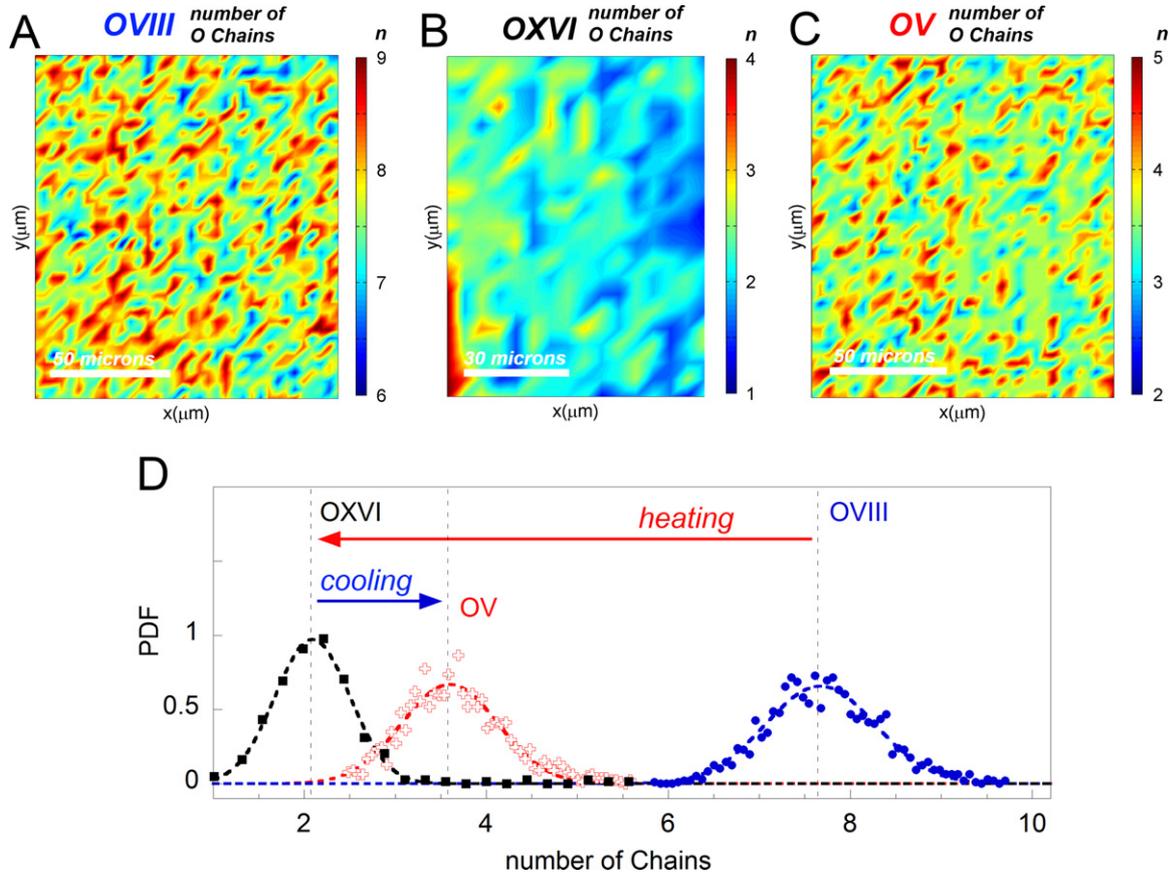


Figure 3. (a)–(c) The spatial map of the number of oxygen chains inside the OVIII, OXVI and OV puddles. (d) Distribution of the number of oxygen chains in the OVIII (blue filled circles), OXVI (black filled squares) and OV (red empty crosses) puddles. The number of oxygen chains decreased after the thermal heating cycle from OVIII to OXVI and increased again after cooling in the OV puddles, but remains lower than in the OVIII phase. Dotted lines are guides for the eyes.

4. Conclusion

In conclusion, we have investigated how thermal treatments allow us to microscopically manipulate and control the functional properties of $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ ($p \approx 1/8$). We used an x-ray diffraction approach, combining standard synchrotron XRD measurements ($200 \times 200 \mu\text{m}^2$ beam size) with scanning μXRD ($1 \times 1 \mu\text{m}^2$) and VSM. Using thermal annealing we induced a continuous phase transition that led to a different final arrangement of Cu–O chains in the sample. In particular we monitored a transition from the OVIII to the OV modulation for the oxygen chain domains, by cycling between 300 and 400 K. The microscopic dynamics of the domains have been investigated by scanning μXRD . We mapped with micrometric resolution the out of plane domain size, the number of oxygen chains and the charge density inside each domain, covering a total area of $160 \times 80 \mu\text{m}^2$. We recorded 12 800 diffraction patterns for each spatial map, showing a high nanoscale inhomogeneity and the presence of a complex network-like organization of competing superconducting puddles that are characterized by different numbers of oxygen chains and charge densities. Reductions in the out of plane domain size, in

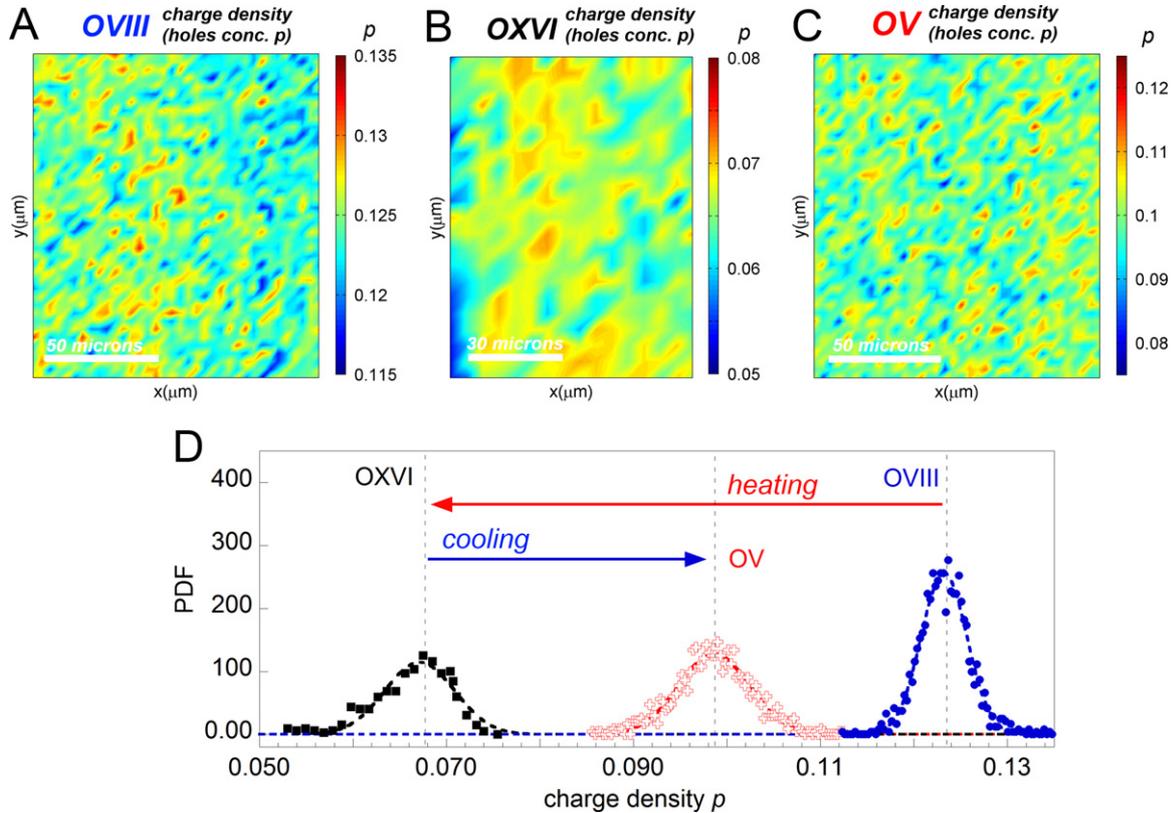


Figure 4. (a)–(c) The spatial map of the charge density (hole concentration p) of the OVIII, OXVI and OV puddles. (d) Distribution of the charge density in the OVIII (blue filled circles), OXVI (black filled squares) and OV (red empty crosses) puddles. During the thermal annealing process the charge density changes and its distribution in the OXVI and OV puddles gets broader.

the number of oxygen chains and in the microscopic distribution of charge density have been observed in the OV phase. These reductions have been connected to a decrease of the number of holes in the active layer. As a consequence, magnetization measurements show that the modification of the network structure of superconducting grains is responsible for a drop of T_c of about 2 K. This can open the way to a possible T_c tuning by microscopic thermal manipulation of the oxygen chain distribution in HTSs. This work shows the presence of a microscale phase separation in $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ with a hole doping close to 1/8, where the lattice misfit strain [43] in these heterostructures at atomic limit and the proximity of the Fermi level to a 2.5 Lifshitz transition near a band edge of the subbands [44, 45, 27–29] induce the observed nanoscale phase separation predicted by the multiband Hubbard model [46]. Finally, in this experiment we observe a superstripe [13] lattice scenario in $\text{YBa}_2\text{Cu}_3\text{O}_{6.67}$ made of different striped nanoscale puddles of locally ordered interstitials with well defined hole doping density. The unique information that our experiment provides on the density distribution of the nanoscale striped puddles shows complex networks of superconducting units, that supports the statistical physics theories of percolative superconductivity in complex networks as an essential feature for understanding the emerging high temperature superconductivity [47–51]. In fact, the reconstructed spatial maps shown here provide compelling evidence for the generic granular

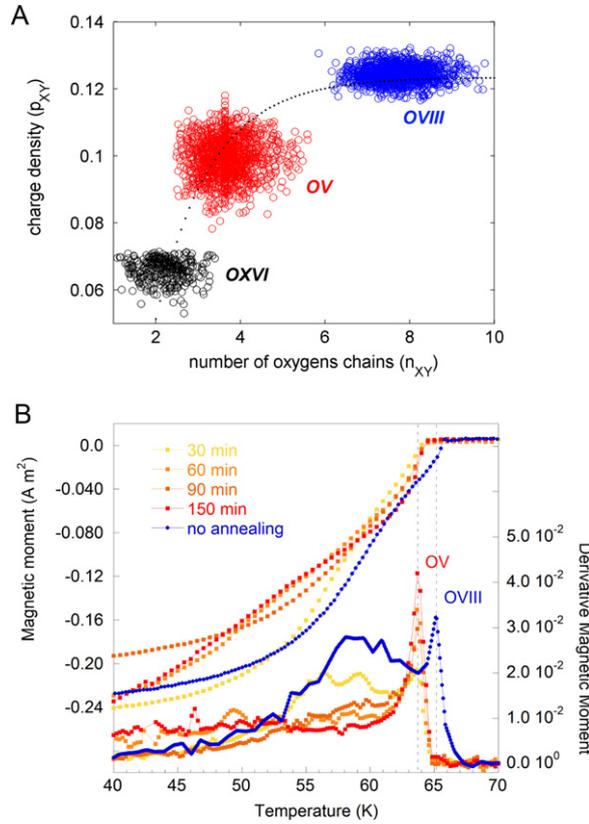


Figure 5. (a) Charge density (hole concentration p) as a function of the number of oxygen chains, inside the OVIII, OXVI and OV puddles. The dotted curve is a fit using a model: $p = 1 - \exp\{-[(n - n_0)/n_0]\}/\xi_n$, where n_0 and ξ_n are the minimum and the maximum number of chains present in the average puddle. (b) Left scale: zero field cooling (ZFC) diamagnetic response of YBCO upon thermal cycles in an external applied field $H=20$ Oe. Blue filled circles: signal before any thermal annealing. Yellow–red squares: the magnetic moment upon sample annealing at 380 K with increasing dwell time of 30, 60, 90 and 150 min. Right scale: numerical derivative of the magnetic moment. Upon thermal cycling the onset of superconducting shielding decreases by about 2 K.

structure that characterizes cuprates and iron chalcogenides. We disclose practical multiple realizations of complex networks of dopant self-organization at the nanoscale with striped puddles characterized by different modulations, local charge densities and superconducting condensates, which share the common features for the emergence of high T_c superconductivity.

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