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ORIGINAL PAPER

Fractal Structure Favoring Superconductivity at High Temperatures in a Stack of Membranes Near a Strain Quantum Critical Point

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Abstract The statistical physics of the 3D ordered oxygen interstitials has been measured in La_2CuO_{4+y} using an advanced tool, scanning x-ray diffraction with focused synchrotron radiation. The observed fractal scale invariant distribution is found in a cuprate in the proximity to a stripes critical point in the 3D Aeppli-Bianconi phase diagram of cuprates, where T_c is function of both hole doping and superlattice misfit strain. Therefore high-temperature superconductivity is favored by complex fractal systems while on the contrary standard low temperature superconductivity is favored in simple periodic crystals. This work shows that the fractal structural distribution in a stack of membranes favors the macroscopic quantum coherent condensate at high temperature. This result opens new perspectives for the understanding the relationship between emergent scale-free distribution in living matter and possible quantum coherent phenomena able to resist to the attacks of temperature decoherence effects.

Keywords High-temperature superconductivity · Multiscale phase separation · Criticality · Scanning microdiffraction · Interstitial oxygen ions mobility · Scale-free distribution · Quantum entanglement

1 Introduction

Understanding and controlling complexity characterized by multiscale phase separation [1, 2] and criticality near first order quantum phase transitions [3] are needed for developing new smart materials [4]. Complexity appears in colossal magneto resistance (CMR) [5, 6] and high-temperature superconductors (HTS) [2, 7–10] complex functional materials. HTS materials are made of stacks of active superconducting layers intercalated by spacer layers. The nanoscale electronic phase separation in the superconducting layers is related to the distribution of dopants (defects, interstitials, substitutions) in the spacer layers, therefore the quantum functional properties of the Fermi liquid can be controlled by counterions. Little is known on out-of-plane disorder or self-organization in HTS and the relation between the structure and high-T_c superconductivity remains unsolved after 24 years of investigations. A key limitation is due to experimental probes of the k-space as ARPES and neutron diffraction, which do not provide spatial resolved information, and local probes like STM, μSR and EXAFS, which do not provide k-space information. Recently novel HTS investigation by scanning synchrotron radiation x-ray micro-diffraction (μXRD) that combines high resolution in the k-space and micron-scale spatial resolution has been reported [11]. Therefore it is a new emerging technique to investigate micron-scale phase fluctuations [12, 13]. We present further compelling evidence for volume power-law distribution of the ordered interstitial oxygen (i-O) microscale domains in La_2CuO_{4+y}. The increase of the critical temperature in La_2CuO_{4+y} from $T_{c1} = 32$ K to $T_{c2} = 40$ K is shown to be correlated with structural criticality. The power-law distribution of the ordered domains of oxygen interstitials (i-O) in La_2CuO_{4+y} has been measured by using advanced spatial imaging with scanning x-ray diffraction with focused synchrotron radiation beam [12, 13]. The use of an advanced tool for imaging the lattice homogeneity via mapping superstructure satellites and of statistical physics methods for data analysis have provided the

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measure of the distribution of ordered domains. Imaging the order of intrinsic inhomogeneity in doped multilayers is opening new perspectives for the control of complexity to develop functional materials. These unexpected results provide a new scenario for the control of high-T_c phase related with criticality in multilayered materials and open new perspectives for controlling the superconducting phase by interstitial oxygen dopant ordering in the spacer layers.

These phenomena have been studied before in complex fluids, such as liquid crystals, soft matter, biological systems, but recently also in colossal magneto resistance [1] and ferroelectric materials [4]. The coupling among the order parameters for various ground states in high-temperature superconductors and local constrains versus long range interactions can lead to multiscale phase separation and diverse combinations of the order parameters with a hierarchy of structures from nano to micro length-scales. Unforeseeable technological applications can develop as for such smart materials [4].

Recently a large number of experiments have shown an intrinsic heterogeneity in hole-doped HTS cuprates where two electronic components coexist: a first itinerant component observed at nodal arcs of the Fermi surface measured by angular resolved photoemission (ARPES) and a more localized second one, characterized by a large pseudogap like in manganites [14].

All known HTS (cuprates, diborides and pnictides) are multilayered materials made of stacks of active superconducting layers intercalated by spacer layers. Changing the elastic misfit strain between the different layers results in dramatic change on T_c [15–17] in hole-doped cuprates, where it induces both a compressive microstrain within the CuO_2 layers and a tensile microstrain within the spacer layers that increases the mobility of dopant interstitial oxygen interstitial ions [18]. Several experiments show that T_c is enhanced when randomly distributed dopant atoms form an ordered array in the spacer layers [19–24].

La_2CuO_{4+y} is the ideal system to investigate multiscale phase separation in the CuO_2 plane, since the i-O are mobile in the intercalated spacer layers La_2O_{2+y} having a large tensile microstrain because of the largest misfit strain between cuprates [15–17, 25–27]. Therefore the order-disorder of localized holes in the CuO_2 plane can be tracked by the order-disorder of the i-O in the spacer layers. In the optimum doping range $0.1 < y < 0.12$ a single $T_c = 40$ K superconducting phase appears with a 3D ordering of i-O [24]. A phase with randomly distributed i-O can be produced by rapid quench from $T > 350$ K to low temperature. In the quenched phase T_c is reduced by about 8 degrees [24]. The scenario that the critical temperature increases by ordering of mobile i-O in the spacer layers is supported by the Mohottola et al. [28] experiment in $La_{1-x}Sr_xCuO_{4+y}$. The coexistence of ordered and disordered i-O domains, an intrinsic phase separation regime [29–31] and coexistence of a striped phase with the superconducting order below T_c [32, 33] show the presence of phase separation in the optimum doping regime. Nevertheless, no information is available on the relation between the structure and HTS phase.

We report the imaging of microscale phase separation in $La_2CuO_{4.1}$ with different oxygen self-organizations and different critical temperatures providing an answer to long standing open problem on the correlation between the ordering of oxygen interstitials [34] and localized holes with the superconducting long range order.

2 Experimental Results

Imaging of the microscale phase separation regime at optimum doping in $La_2CuO_{4.1}$ has been obtained by scanning μXRD technique. The experiment has been performed at the beam-line (ID13) at ESRF taking advantage of the novel focusing methods of the x-ray beam emitted by high brilliance synchrotron radiation x-ray sources. This is an unique method for mapping ordering heterogeneity in single crystals in the micron scale [12, 13]. We have investigated single crystals with $y = 0.1$ interstitial oxygen concentration with typical millimeter size as used for experimental methods probing the k-space as the ARPES or x-ray or neutron diffraction. The x-ray beam is focused on a 1 μm^2 spot on the sample surface and the relative diffraction pattern has been recorded by a CCD detector. The main μXRD reflections of the single crystal of the orthorhombic Fmmm space group show no splitting or spatial spot-to-spot change. Satellite peaks, associated with a 3D nearly commensurate superstructure of the ordered i-O dopants [28], appear near the main crystal reflections displaced by the wavevector $\mathbf{q}_2$ with components $\pm\Delta l = 0.5c^*$, $\pm\Delta k = 0.25b^*$, $\pm\Delta h = 0.09a^*$ with a strong second and third harmonic components.

The crystalline sample is cleaved with surface normal to the crystalline c axis. The crystal is mounted in a xy mechanical translator for scanning parallel to the crystallographic (a, b) plane. The experimental set up allows the xy translation of the sample with 22 μm steps in the x direction and 5 μm steps in the y direction, scanning a 350×600 μm^2 sample area. The integrated intensity of the satellite superstructure peaks recorded by a CCD detector at each microscale spot probes the square root of the volume of the ordered i-O domains in a μm^2 spot area. Data have been normalized by recording the ratio of the Q2 superstructure satellites integrated intensity on the tail of the main crystalline reflections, typically the (006) reflection, near the satellite spot.

The distribution of the volume of the ordered i-O domains, measured by the distribution of the reflections due

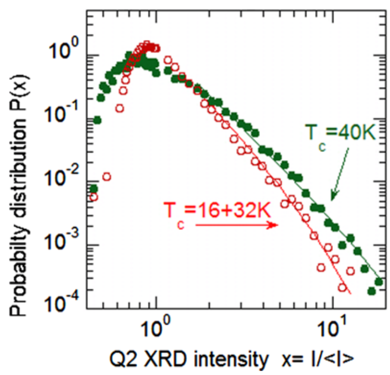

Fig. 1 The probability distribution, $P(x)$, of the intensity $I(\mathrm{Q2})$ of the reflections due to x-ray diffraction (XRD) Q2 satellites of the main crystalline reflections due to the Q2 superstrucrture produced by the ordering of the oxygen interstitials and normalized to the background I0: $x = I(\mathrm{Q2})/I_0$ extracted from the mapping using space scanning micro-X-ray diffraction (μXRD). The intensity distribution of a low T_c and a high-T_c samples are shown. The scale-free distributions, characteristic of fractal structures, are fitted by a power law with exponent $\alpha = 2.6 \pm 0.2$ independently of the sample critical temperature, and a variable cut-off that increases for the $T_c = 16 + 32$ K samples to the $T_c = 40$ K samples

to the superstructure, is shown in Fig. 1 for a typical high-T_c and a typical low T_c sample. The figure shows the difference between the intensity distribution in the two samples related with a different T_c. The difference between the two probability distributions of the two samples is quantified by fitting the data using a power law $P(x) = x^{-\alpha}\exp(-x/x_0)$. The outcome is that the power-law exponent, $\alpha = 2.6$ remains constant while the cut-off, x_0 changes from is less than 10 for the low $T_c = 16 + 32$ K materials and greater than 10 for the high-T_c (40 K) materials.

Panel (a) of Fig. 2 shows the 3D Aeppli-Bianconi phase diagram of cuprate superconductors, where the color plot of T_c as a function of both hole doping and superlattice misfit strain is shown. The super-oxygenated $La_2CuO_{4.1}$ is indicated by the open circle. Panel (b) of Fig. 2 shows the image of distribution of the oxygen ordered domains by scanning μXRD of a $T_c = 40$ K sample. The spatial color plot of normalized superstructure reflections intensity is plotted as a function of the position of the incident x-ray beam on the sample surface in the x, y plane is shown. The scanning images show few spots having intense satellite μXRD reflections with a typical fractal pattern. The Q2 satellite reflec-

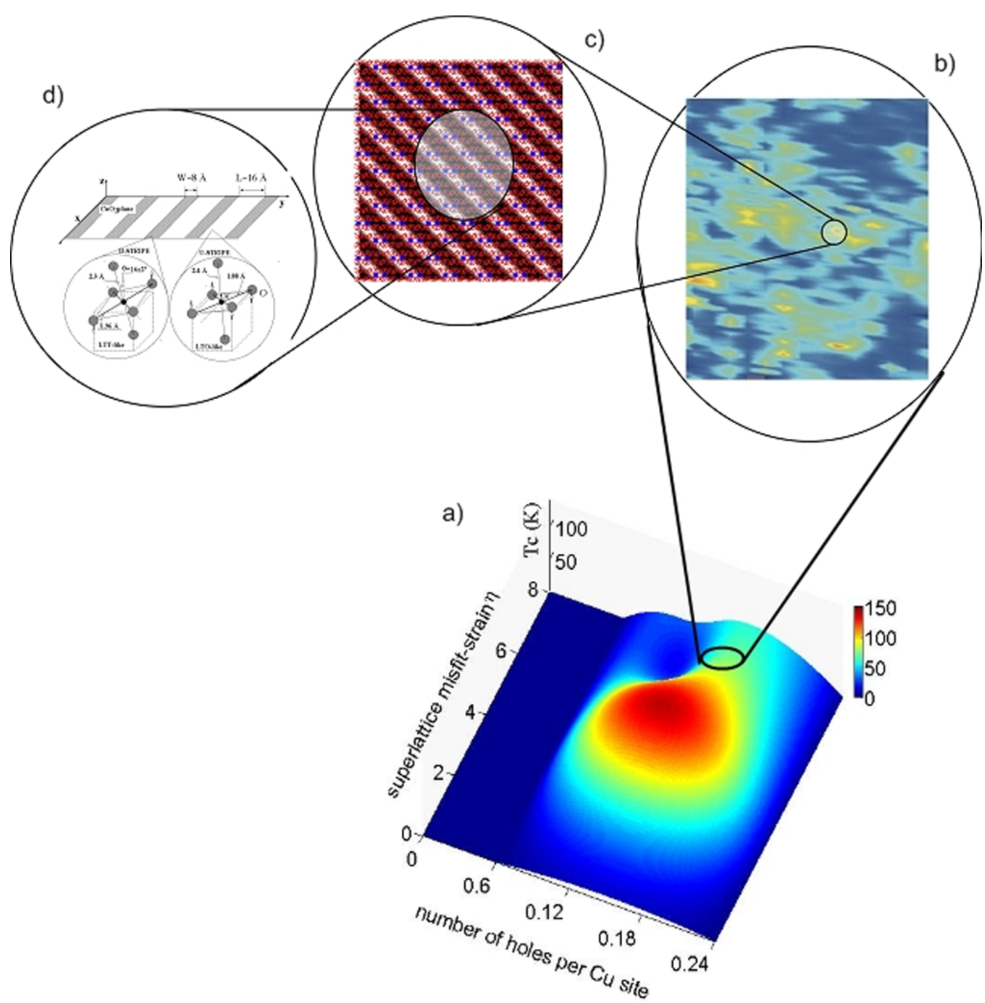

Fig. 2 Panel (**a**) The 3D universal phase diagram of the superconducting critical temperature of cuprates. The values of color plot of the superconducting transition temperature T_c go from 0 (*black*) to 135 K (*white*). The superlattice misfit strain η (related with the microstrain or internal chemical pressure) and the number of holes for Cu site δ. The strain critical point for the formation of a static stripe phase is indicated by the empty circle. Panel (**b**) 2D μXRD color plot of a $T_c = 40$ K sample of La_2CuO_4. Panel (**c**) The ordering of i-O in the bc-plane of the Fmmm crystal structure forming the patches with Q2 superstructure. Panel (**d**) The striped lattice modulation in the CuO_2 plane due to pseudo-Jahn–Teller polaron ordering

tions intensity probe the ordering of the interstitial oxygen ions at the (1/4, 1/4, 1/4) site of the unit cell in the center of the rocksalt layers with a period of about 2, 4 and 11 unit cells along the c, b and a axes as shown in panel (c) of Fig. 2. The physics of multiscale phase separation occurring in the HTS oxides is pictorially represented by the zoom in panel (d) of Fig 2, showing the nanoscale striped lattice modulation in the active superconducting CuO_2 plane, due to ordering of the pseudo-Jahn-Teller polaron measured by EXAFS, a fast (having a measuring time scale of 10^{-15} sec) and local probe.

3 Discussion

The observed presence of the phase separation implies the competition of local lattice distortions and elastic fields in the proximity a structural phase transition driven by the misfit strain field. In the misfit strain critical point scenario $La_2CuO_{4.1}$ is close to the highest values of misfit strain between the bcc CuO_2 and the spacer layers, in fact the system is close to the structural phase transition where the spacer structure changes for rocksalt to fluorite structure, called the $T \rightarrow T'$ phase transition in perovskites at constant doping. In fact the oxygen interstitial occupy the oxygen site in the fluorite structure that can be considered a nucleation of the T' phase. The results shown in Fig. 1 and Fig. 2 clearly indicates a critical opalescence for the ordered i-O domains, providing evidence for the proximity to a critical or multicritical point localized in the 3D phase diagram of cuprates driven by the elastic field.

The fact that the superconducting critical temperature increases by increasing the cut-off of the power-law distribution seen in Fig. 1 can be interpreted as evidence that T_c increases at a *critical distance* from the critical point. The ordering of i-O phase is related with the phase separation and growth of the Q2 domains. The hole doping in the CuO_2 plane can be estimated to be 0.111 holes per Cu site, i.e., near the 1/8 striped phase of ordered localized holes in the CuO_2 plane made of $(\text{i-O}^{-1})_2$ pairs as predicted by Lee and Hoffman [34]. This phase forms strings of 11 lattice units in the a direction, a superlattice of stripes in the b direction and with a stage 2 structure in the c direction. Therefore in this scenario an electronic glassy phase with higher doping of 0.15 hole per Cu site at optimum doping coexists with a striped phase near the 1/8 doping. The scale invariant distribution of the 1/8 domains could be related to quantum fluctuation near the strain quantum critical point for the formation of stripes at the critical 4% compressive microstrain of the CuO_2 two-dimensional lattice. Therefore is possible that the power-law distribution of i-O microscale phase separation in the spacer layers is the result of a photographic process of quantum critical fluctuations in the CuO_2 superconducting layers. The present results support the scenario proposed by Zaanen et al. [3], Dagotto [1], and Bishop et al. [2] where the randomness, the proximity to a structural phase transition and the local lattice constrains associated with the polaron stripes [29, 30] drive the formation of a collective striped phase of i-O ordered microdomains with a power-law distribution.

Several experiments have indicated phase separation in cuprates to be favorable for high-T_c superconductivity before this discovery and several theoretical models have been proposed [29–31, 35–40]. The present results are close to the prediction of the superstripes phase made of bubbles of the 1/8 phase embedded in the glassy phase in the proximity of the stripes phase described in 2000 [41, 42]. In this scenario the elastic field of the superlattice misfit strain plays a key role as is possible in pnictides [43] in proximity to a structural phase transition.

In conclusion we have given an insight into the criticality of the interstitial oxygen self-organization in the La_2CuO_{4+y} using the scanning μXRD technique supporting the critical point located exactly in the 3D phase diagram of cuprates. The cut-off of the power-law distribution in the scanning μXRD mapping provides a measure of the distance from the critical point supporting a scenario where intrinsic multiscale phase separation is a key ingredient for establishing a superconducting phase that resists to the attacks of high temperature.

In order to understand the quantum mechanism that allows a quantum condensate to resist the decoherence attacks of high temperature it has been proposed that it is based on the quantum configuration interaction between open and closed channels proposed by Ettore Majorana in 1929 [44]. In many-body physics of BCS condensates this quantum interference is at the basis of shape resonances in superconducting gaps in multiband or multigap superconductors [45–49]. In this scenario the exchange-like interband coupling controlling the pair transfer from a first condensate to a second condensate increases the stability of the quantum condensate increasing the critical temperature. In Fig. 3 we show a possible pair transfer mechanism between a first superconducting phase in a percolating network of stripes near 1/8 doping and a second superconducting phase at optimum doping in the glassy phase. The phase separation produces electronic states in different portions of the material reducing the possible mixing between the two set of states due to impurity scattering that suppress the multigap superconducting phase. The different structure of the intermixed fractal phases produces two condensates with different topological symmetry producing a particular case of topological superconductors [50, 51] and a particular type of fractal superconductor in a complex disordered phase [52].

We have seen scale-free structural organization in the proximity of a critical point, where shape resonances in superconducting gaps are likely to appear id the electronic

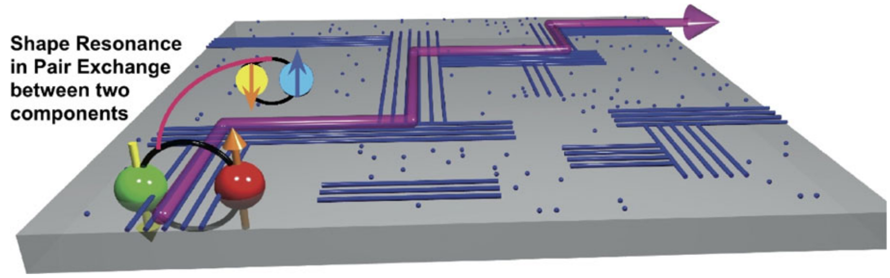

Fig. 3 Pictorial view of the shape resonance mechanism proposed to be the key mechanism moving superconductivity from low to high temperature in the phase separation regime between first pairs in a percolating network of oxygen ordered stripes and second pairs in the disordered glassy phase

bands are finely tuned by chemical doping, randomness and misfit strain.

The present findings of complexity in high-temperature superconductors allow us to speculate about the role quantum phenomena in living systems that is the most interesting case of complex condensed matter made of simple ingredients that show emergence of life.

The idea that the coherent state of living matter emerges in the proximity of a critical point in the so-called scenario of biological order at the edge of chaos is attracting high interest in the scientific community [53, 54]. The scale-free distribution, similar to that we have observed in the present μXRD experiment, appears in the networks of protein-protein interaction in the cell [55] and for growing neuron networks in the brain [56], so one could rise the question if also the living cell is near some criticality. It is known that quantum critical fluctuations are mostly probably the source of entanglement in several condensed systems [57–59] and could be the cause of the fractals observed in La_2CuO_{4+y} [60].

Fractals are ubiquitous in the living world for some unknown reason, and there should be a function for them. One hypothesis is that scale-free organization can be suitable for quantum resonances to appear in the proximity of quantum criticalities. The quantum shape resonances have been found to increase the critical temperature in multigap superconductors and the similar Feshbach resonances to rise the critical temperature of boson superfluid condensates in ultra cold gases. The quantum shape resonances, in the association and dissociation processes of selected biological molecules, have been proposed to drive the coherence in living matter being able to avoid the decoherence effects due to temperature [61]. The idea that quantum coherence may play some role in the enormous robustness of living matter has recently taken more strength by the evidence for wavelike energy transfer thought quantum coherence in photosynthetic systems found by the Fleming's group [62] and in the entanglement of the avian compass [63]. The scientific investigation of the entanglement in many-body complex systems is therefore not only a special problem in physics of quantum information [58], superconductivity, and superfluids, but it is a new feature emerging also in biological systems that need to be explored [64].

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