Functional Nanoscale Phase Separation and Intertwined Order in Quantum Complex Materials

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Abstract: Nanoscale phase separation (NPS), characterized by particular types of correlated disorders, plays an important role in the functionality of high-temperature superconductors (HTS). Our results show that multiscale heterogeneity is an essential ingredient of quantum functionality in complex materials. Here, the interactions developing between different structural units cause dynamical spatiotemporal conformations with correlated disorder; thus, visualizing conformational landscapes is fundamental for understanding the physical properties of complex matter and requires advanced methodologies based on high-precision X-ray measurements. We discuss the connections between the dynamical correlated disorder at nanoscale and the functionality in oxygen-doped perovskite superconducting materials.

Keywords: topology; complex materials; spatial statistics

1. Introduction

Nanoscale phase separation (NPS) has been considered to be detrimental for high-temperature superconductivity. However, high-precision X-ray measurements in solid-state physics [1–5] have recently provided experimental validation for the alternative paradigm, where lattice heterogeneity from the atomic limit to the micron scale plays a key role [6–12]. NPS shows that intertwined and interlocked nanoscale lattice structures form heterostructures at the atomic limit [13]. This induces the emergence of novel functionalities, such as high-temperature superconductivity, in complex quantum materials as organics [13–20], doped perovskites [21–39], iron-based superconductors [40–44], charge–density–wave materials [45–52], manganites showing colossal magnetoresistance [53–57], doped diborides [58–60], smart structures and biological systems [61–66], perovskite oxide interfaces [67–69], and magnetic materials [70,71].

Today, at the beginning of the twenties of the XXI century, the development of advanced experimental X-ray methods has provided evidence that quantum functionalities (e.g., colossal magnetoresistance and high-temperature superconductivity) are controlled by the correlated lattice disorder from atomic scales to micron scales. This multiscale lattice heterogeneity has been quite difficult to unveil by conventional experimental approaches, requiring high spatial resolution probes. Nowadays, advanced, latest generation synchrotron sources and new X-ray optics have allowed the design of new scanning techniques, based on focused X-ray beams.

In this way, several crystallographic structures, coexisting on micron scales and nanoscales, have been visualized in high-temperature superconductors (HTS). Strain and doping drive these systems at critical points for phase separation, with the coexistence of different crystallographic phases existing in different nanoregions of the crystal. While in the literature on HTS, the focus of theoretical research has been addressed to electronic
phase separation and inhomogeneity—due to competition or intertwining between superconductivity and magnetism—the main result of scanning X-ray diffraction has been to provide compelling experimental evidence that, at nanoscale, different puddles show a substantial lattice structural difference, which necessarily implies electronic and magnetic phase separation. In fact, looking at the nanoscale, we have observed different nanoregions with different lattice structures, showing different electronic and magnetic ordered phases, with different topologies that influence the Fermi surfaces, the pseudogap energy, and even the superconducting critical temperature. We would like to underline archetypal cases of intrinsic phase separation in $A_xFe_{2-y}Se_2$, which are originated by the coexistence of an insulating magnetic phase and a paramagnetic metallic phase with an in-plane compressed and expanded lattice [40–44]. A second clear case of phase separation has been found in $YBa_2Cu_3O_{6+y}$, with $y < 0.5$, where superconductivity ($y > 0.33$) percolates in a network of oxygen-ordered nanoregions ($y = 0.5$), interspersed with oxygen-depleted domains ($y = 0$) [37].

Here, we have chosen to discuss the relationship between phase separation at nanoscale and the emerging macroscopic properties in two high-temperature superconducting quantum materials, such as $La_2CuO_{4.1}$ [38] and $HgBa_2CuO_{4.12}$ [7], doped by mobile oxygen interstitial ions (O-i).

2. Results

2.1. Correlated Disorder and Phase Separation in $La_2CuO_{4+y}$

$La_2CuO_{4+y}$ is one of the simplest compounds, where $y$ oxygen interstitial ions (O-i) in the rocksalt $[La_2O_{2.5}]$ are intercalated by $[CuO_2]$ layers. Thanks to the high synchrotron photon flux, we have measured the satellite peaks, which are associated with super-cells, due to oxygen interstitial O-i dopants and charge–density waves (CDW) arrangement (see Figure 1a). CDWs incommensurate diffuse satellites with wave vector $q_{CDW} = 0.21b^* + 0.29c^*$, coexist with O-i satellites displaced by $q_{O-i} = 0.25b^* + 0.5c^*$. Oxygen ions, O-i, and CDW superstructures are characterized by a different staging, that is, their $c^*$ component; indeed, O-i and CDW have staging $c^* = 0.5$ and $c^* = 0.29$, respectively, as indicated by the white arrows in Figure 1a.

A common and intriguing feature of quantum materials is that their emerging properties can be easily manipulated by weak external stimuli, such as temperature gradient, strain, and photon illumination. Due to the mobility of oxygen interstitials (O-i), our systems present structural conformational landscapes. The conformations have been manipulated by continuous X-ray photo illumination, as shown in Figure 1b, where we bring the sample from a structural conformation with disordered O-i to a different conformation with ordered O-i, after illumination. Different conformations, characterized by different order degrees of O-i [72,73], can be obtained by combining X-ray flux intensity with thermal cycling, as shown in Figure 1c. Here the (full symbols) X-ray low flux and (empty symbols) high flux on the sample, correspond to $0.5 \times 10^{14}$ photons s$^{-1}$ cm$^{-2}$ and $5.0 \times 10^{14}$ photons s$^{-1}$ cm$^{-2}$, respectively. The temperature change rate has been $1$ K min$^{-1}$ in both low-flux and high-flux illumination. Each point on the two hysteresis corresponds with a specific structural conformation, with a specific degree of order (or disorder) of O-i.

The different O-i ordered domains are inhomogeneously distributed in space, as seen by $S\mu$XRD. The maps in Figure 2a,b show two different O-i distributions, obtained with two different thermal treatments; the first one (a) shows the maximum critical temperature ($T_c = 40$ K) and the second (b) presents a macroscopic phase separation with two critical temperatures, $T_c = 16$ K and $32$ K [6].
We have characterized this inhomogeneity by using spatial statistics tools, described in [6]. In both samples, the probability density function of O-i satellite intensity follows an exponentially truncated power–law distribution, given by $P(x) = x^{-\alpha} \exp(-x/x_0)$, where $\alpha$ is the critical exponent and $x_0$ is the cut-off. The critical exponent, $\alpha$, results to be 2.6, both in the high-$T_c$ (a) and low-$T_c$ (b) conformations, indicating the fractal nature of O-i arrangement. On the other hand, the cut-off, $x_0$, is larger for the high-$T_c = 40$ K sample, as can be seen in Figure 2c. This means that a larger extent of O-i satellite intensity with fractal geometry favors a higher $T_c$ [38].

We have found that the spatial inhomogeneity is characterized by a strong spatial anticorrelation between the O-i-rich and CDW-rich regions. In the oxygen-rich regions, the system is in the over-doped metallic phase, while in the oxygen-poor region in the underdoped phase, we measure a high density of CDW. Figure 3a shows the map of
difference between the intensity of O-i and CDW diffraction satellites. The O-i are located in the 1/4,1/4,1/4 site, and form commensurate stripes in the La$_2$O$_2$ structure, intertwined with incommensurate lattice CDW domains [39]. Indeed, we observed micron size puddles of blue metallic oxygen-rich regions, separated by red CDW-rich zones, by a filamentary percolating interface, indicated by white space in Figure 3b.

![Figure 3](image_url)

(a) Surface plot of the map difference between the normalized intensities of both CDW and O-i satellites in La$_2$CuO$_{4+y}$ [38]. CDW puddles are dominant in red regions, while the O-i stripes are dominant in the blue zones. The lattice modulations corresponding to CDWs and O-i superstructures are depicted. (b) 2D map difference highlighting the interface (white region) between CDW-rich and O-i-rich regions. The white bars correspond to 5 µm.

2.2. Correlated Disorder and Phase Separation in HgBa$_2$CuO$_{4+y}$

Structural and electronic inhomogeneity has been studied in the high-temperature perovskite superconductor, with tetragonal crystal symmetry HgBa$_2$CuO$_{4+y}$ with a single CuO$_2$ plane. In this compound, the oxygen interstitial ions (O-i) form atomic stripes in the spacer layer [HgO$_x$Ba$_{2-y}$O$_y$] between [CuO$_2$] planes. We have measured the diffuse scattering associated with both CDW and oxygen interstitial arrangement in the lattice. Diffuse CDW satellites have been detected at $q_{\text{CDW}} = 0.23a^* + 0.16c^*$ and $q_{\text{O-i}} = 0.23b^* + 0.16c^*$ (where $a^*$, $b^*$, and $c^*$ are the lattice units in the reciprocal space) around the (108) Bragg peak, below the onset temperature $T_{\text{CDW}} = 240$ K [7]. Resolution-limited streaks connecting the Bragg peaks, due to atomic O-i stripes in the HgO$_x$ spacer layers, have been measured in agreement with previous experiments [6,32].

In Figure 4a,b we show the maps of the integrated intensity of CDW peak and oxygen O-i diffuse streaks, respectively [7,9], as seen by SuXRD measurements. Both O-i and CDW maps are spatially inhomogeneous, and their PDF is well modeled by a power-law behavior also in this case, as shown in Figure 4c. The critical exponents in the two intensity distributions are $1.8 \pm 0.1$ and $2.2 \pm 0.1$ for the O-i and CDW, respectively [7].

A map of the spatial organization of the in-plane CDW-puddle size, $\xi_\parallel$, is shown in Figure 4d. Although the average size of CDW puddles is 4.3 nm (in agreement with previous works), its probability density function, shown in Figure 4e, has a fat-tail, fitted by a power-law curve with critical exponent of $2.8 \pm 0.1$. This means that rare and larger puddles up to 40 nm are measured, and their distribution provides a complex topology for the flowing of superconductivity currents [7,9].
As in the La$_2$CuO$_{4+y}$ superconductor, the spatial inhomogeneity shows a negative correlation between O-i and CDW. This is well depicted in the ‘difference map’ between CDW peaks and O-i-diffuse streaks in Figure 5a. The poor CDW regions on the CuO$_2$ basal plane correspond to O-i-rich regions on the HgO$_y$ layers. The O-i atomic striped domains are here intertwined with incommensurate lattice CDW. The percolating filamentary interface between O-i-rich and O-i-poor regions is indicated by white space in Figure 5b.

Figure 5. (a) Surface plot of the map difference between the normalized intensities of both CDW satellite and O-i streaks in HgBa$_2$CuO$_{4.12}$ [7]. CDW puddles are dominant in red regions and the O-i stripes are dominant in the blue zones. The bar corresponds to 5 µm. (b) Pictorial view of the spatial anticorrelation between CDW puddles in the CuO$_2$ plane and the O-i-rich domains in the HgO$_y$ layers. Here, the interface space between CDW and O-i has been highlighted by the white region. The bar corresponds to 5 µm.
3. Discussion

The anomalous phase diagram of HTS cuprates is believed, nowadays, to be closely related to the phase separation at multiple scale length. This feature is particularly pronounced in those compounds doped by mobile oxygen interstitials. In this context, the present work reports evidence of nanoscale phase separation in the cases of two different oxygen-doped layered perovskites, La$_2$CuO$_{4+y}$ and HgBa$_2$CuO$_{4+y}$. At optimum doping level, specific charge ordering and segregation lead to the formation of metallic and insulating domains with oxygen-rich and oxygen-poor regions, respectively. These different zones have been measured as different superlattices in X-ray diffraction and their inhomogeneous spatial distribution has been visualized by scanning micro X-ray diffraction. The results provide relevant evidence for the universality of phase separation, where dopants’ O-i-rich domains are intertwined with CDW domains, even in the most optimized superconducting cuprates.

We have shown that the coexisting phases can be easily manipulated (e.g., by X-ray illumination) on controlled small areas, drawing a way to extend the functionality of the investigated materials in the direction of information storage [74,75].

4. Materials and Methods

The La$_2$CuO$_{4+y}$ (LCO) single crystal with $y = 0.1$ has orthorhombic $Fmmm$ space group symmetry, with lattice parameters $a = (5.386 \pm 0.004)$ Å, $b = (5.345 \pm 0.008)$ Å, and $c = (13.205 \pm 0.031)$ Å at room temperature. The HgBa$_2$CuO$_{4+y}$ (Hg1201) single crystal, with $y = 0.12$, has a sharp superconducting transition at $T_c = 95$ K. The crystal structure has tetragonal $P4/mmm$ space group symmetry with lattice parameters $a = b = 0.387480(5)$ nm and $c = 0.95078(2)$ nm at $T = 100$ K. Diffraction measurements on both single crystals of LCO and Hg1201 were performed on the ID13 beamline at ESRF, as described in [38] and [7], respectively. Data analysis has been performed by using customized and homemade written MATLAB routines [6].

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