High-Temperature Superconductivity in a Hyperbolic Geometry of Complex Matter from Nanoscale to Mesoscopic Scale

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Abstract While it was known that high-temperature superconductivity appears in cuprates showing complex multiscale phase separation due to inhomogeneous charge density wave (CDW) order, the spatial distribution of CDW domains remained an open question for a long time, because of the lack of experimental probes able to visualize their spatial distribution between atomic and macroscopic scale. Recently scanning micro-X-ray diffraction (μXRD) revealed CDW crystalline electronic puddles with a complex fat-tailed spatial distribution of their size. In this work, we have determined and mapped the anisotropy of the CDW puddles in HgBa2CuO4+δ (Hg1201) single crystal. We discuss the emergence of high-temperature superconductivity in the interstitial space with hyperbolic geometry that opens a new paradigm for quantum coherence at high temperature where negative dielectric function and interference between different pathways can help to raise the critical temperature.

Keywords X-ray diffraction · High temperature superconductivity · Hyperbolic geometry

Organization of local lattice fluctuations and charge inhomogeneity determines intrinsic physical properties in new functional materials. This heterogeneity varies from the scale of microns to the atomic level [1–3]. Imaging structural fluctuations and inhomogeneity is an interdisciplinary fundamental issue for understanding function-structure relationship in complex materials and for the design of new functional materials. Nowadays, the improved X-ray optics and imaging techniques make possible to image the bulk structure at nanoscale and mesoscale that is the scale length between the atomic and macroscopic world. In particular, scanning micro-X-ray diffraction (μXRD) using focused beams on the micro-scale is able to probe the k-space order in different spatial locations of the material. This allows to unveil the complex maps showing the multiscale structure in materials. In this way, ultrastructure and phase separation have been visualized in complex and heterogeneous materials for applications in different fields from biomedicine [4–7] to material science [8–11].

In high-temperature superconductors (HTS), the spatial imaging of nanoscale phase separation due to quenched disorder of dopants has been obtained both in cuprates [12–14] and iron-based superconductors [15–17]. Recently, the interest of the scientific community has been focused on the competition of short-range charge density wave (CDW) order and superconductivity [18–21]. Scanning micro-X-ray diffraction has been used for imaging the spatial distribution of short-range charge density wave puddles in La2CuO4+y [20] and recently in HgBa2CuO4+y [21], the single-layer cuprate with the highest Tc, 95 K [21]. In this work, we investigate the anisotropy due to the different size of CDW puddles in the CuO2 plane and out-of-plane directions.

The Hg1201 crystal was grown at ETH [22]. Crystal structure has P4/mmm symmetry with lattice parameters \(a = b = 0.3875(5)\) nm and \(c = 0.9508(2)\) nm at...
We have identified the CDW order by single-crystal X-ray diffraction using a beam size of 200 × 200 μm² and a photon energy of 17 KeV on the XRD1 beamline at ELETTRA synchrotron, Trieste [23]. Only selected reflections show clear CDW satellites, in agreement with Croft [18]. We focused on the CDW satellite $q_{CDW} = (0.23, 0, 0.17)$ around the (1 0 8) Bragg reflection, due to stripe-like modulation propagating along the tetragonal $a(b)$ axis with an average of only three–four oscillations per puddle [20]. The CDW order appears below the onset temperature $T_{CDW} = 240$ K [20]. Figure 1a shows how the CDW satellite peak in a diffraction pattern measured at $T = 100$ K. By inspection of the profiles of the CDW peak, along the $h(k)$ and $l$ directions, shown in Fig. 1b, c respectively, we clearly observe how the peak width is larger along the $l$ direction.

Here, we determine the variation of CDW puddle size both along the in-plane direction and along the out-of-plane direction, point by point by means of μXRD measurements. The sample was kept at $T = 100$ K where the system shows the maximum CDW order [20]. μXRD measurements were performed on the ID13 beamline at ESRF, Grenoble, France. We applied an incident X-ray energy of 13 KeV to measure CDW ordering reflections. Moving the sample under a $1 \times 1$ μm² focused beam with an x-y translator, we scanned a sample area of $50 \times 50$ μm² collecting 2500 different diffraction patterns. For each scanned point of the sample, the $(0.23\ 0.0\ 0.16)$ peak profile has been extracted; the full-width half-maximum along $a^*$ (FWHM($a^*$)) and FWHM($c^*$) along $c^*$ directions have been evaluated to obtain the domain size of the charge ordered puddles both along the $a$ and $c$ crystallographic axes. We got clear evidence of inhomogeneous spatial distribution of CDW, by mapping the in-plane domain size, $\xi_{ab} = a(b)/$FWHM($a^*$), and the out-of-plane domain size, $\xi_c = c/$FWHM($c^*$), as a function of position in the sample.

The map of the CDW domain size, $\xi_{ab}$, and $\xi_c$, is shown in Fig. 2a, b, respectively. It is clear that while the puddle size shows large fluctuations in the CuO₂ planes, the puddle size along the out-of-plane direction appears more uniform. The spatial distributions of puddle size have been characterized by the cumulative probability density function given by $\text{CDF}(x) = \int_{-\infty}^{x} p(\xi) d\xi$ where $p(\xi)$ is the probability density function of the size, $\xi$. In Fig. 3, we show the CDF of the domain size, $\xi_{ab}$, and $\xi_c$. We found a fat-tailed CDW size distribution along the in-plane direction $a(b)$. To investigate whether and how much this upper tail approaches a power-law pattern, we fitted the domain size, $\xi$, with the distribution $p(\xi) = \xi^\alpha$ where $\alpha$ is the maximum likelihood estimate of the scaling exponent for $\xi$ larger than a lower bound [24]. The fat tail of the in-plane CDW domain size distribution follows a power-law behavior: $p(\xi_{ab}) \approx \xi_{ab}^{\alpha_{CDW}}$ where $\alpha_{CDW} = 2.8 \pm 0.2$ is the critical exponent. This particular behavior highlights a
scale-free like distribution of the in-plane CDW domain size in Hg1201 at optimum doping.

From the CDF(x), we can determine that the in-plane “average size” of CDW puddles amounts to 4.3 nm in good agreement with previous work probing only the average size. On the other hand, our space-resolved \( \mu \)XRD measurements indicate a size distribution with fractal-like geometry with rare domains reaching a maximum domain size of 40 nm.

At the same time, the probability distribution of the out-of-plane CDW size appears to be exponential, indicating a normal size distribution of out-of-plane CDW puddles. The average out-of-plane size of the CDW puddles has been found to be about 4 nm.

A view of CDW puddles packing is represented in Fig. 4a. The CDW crystalline puddle arrangement introduces a topological change in the available space for the free electrons. These paths go around the CDW puddles in different ways, creating non-trivial interference patterns for electrons and favoring the emergence of the superconducting phase in this complex space.

The free electrons can travel between two different points, taking different paths that cannot be topologically deformed one into the other. This complex space resulting from the measured power-law distribution of the in-plane puddle size can be mapped to a hyperbolic space [25, 26] as it is shown in Fig. 4b where different possible non-equivalent paths taken by electrons at the Fermi level are shown running in the interface space left by the CDW puddles.

It was already established, using XANES spectroscopy [27–29], that the cuprate superconductors show a heterogeneous copper oxide plane due to polaronic charge density wave formation [30, 31].

The use of scanning micro-X-ray diffraction [20] has provided compelling evidence for the emergence of the hyperbolic space in the mesoscopic space with hyperbolic geometry favoring superconductivity in cuprate superconductors.

The hyperbolic space has recently been found to play a key role in different fields looking for the emergence of order in complex systems ranging from network theory to quantum gravity [32, 33]. The role of a spatial hyperbolic geometry in disordered media is known to be of high relevance in the design of new metamaterials showing a negative dielectric constant [34–37], as well as in the manipulation of complex light in optical metamaterials [38–45]. The hyperbolic space has recently been shown to influence critical phenomena [46]. The search for complex materials with zero or negative dielectric constant has been one of the road maps proposed for the amplification of the superconducting critical temperature to reach room temperature superconductivity [47–53]. The present experiments show a hyperbolic space in the mesoscale of cuprates but it could be present also in pressurized sulfur hydride superconductors [54, 55], iron-based superconductors [56], and diborides [57] which all show local lattice fluctuations. Therefore, further investigations are needed to investigate
the substantial role of non-Euclidean space in the mesoscale range in high-temperature superconductors [49–52].

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