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Perspective

Nanoscale Lattice Heterostructure in High-Tc Superconductors

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Abstract

Low-temperature superconductivity has been known since 1957 to be described by BCS theory for effective single-band metals controlled by the density of states at the Fermi level, very far from band edges, the electron–phonon coupling constant l, and the energy of the boson in the pairing interaction w_0, but BCS has failed to predict high-temperature superconductivity in different materials above about 23 K. High-temperature superconductivity above 35 K, since 1986, has been a matter of materials science, where manipulating the lattice complexity of high-temperature superconducting ceramic oxides (HTSCs) has driven materials scientists to grow new HTSC quantum materials up to 138 K in $HgBa_2Ca_2Cu_3O_8$ (Hg1223) at ambient pressure and near room temperature in pressurized hydrides. This perspective covers the major results of materials scientists over the last 39 years in terms of investigating the role of lattice inhomogeneity detected in these new quantum complex materials. We highlight the nanoscale heterogeneity in these complex materials and elucidate their special role played in the physics of HTSCs. Especially, it is highlighted that the geometry of lattice and charge complex heterogeneity at the nanoscale is essential and intrinsic in the mechanism of rising quantum coherence at high temperatures.

Keywords: high-temperature superconducting ceramic oxides (HTSCs); polarons; Q-balls; polaron tunnelling; Kuramoto synchronization; nanoscale lattice fluctuations; artificial high-Tc quantum superlattices; multigap superconductivity; Fano–Feshbach resonances

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1. Essential Heterogeneities in Hole-Doped High-Tc Cuprate Superconductors

Only a year after their revolutionary work in superconductivity, J. G. Bednorz and K. A. Müller were awarded the Nobel Prize for their "important break-through in the

discovery of superconductivity in ceramic materials" [1]. This wording implies already that the material, namely "ceramics", is an inhomogeneous off-stoichiometric compound. Clear proof of inhomogeneity was rapidly provided by site-selective oxygen isotope experiments on the superconducting transition temperature T_C and phonon frequencies, emphasizing that structurally different oxygen ions in YBaCuO (chain, apex, and plane oxygens) contribute differently to the total oxygen isotope effect [2,3]. In Zürich, oxygen isotope effects were further investigated and a number of unexpected effects on the superconducting properties were discovered [4–10]. The differentiation between constituting structural elements of the cuprates continued with the seminal work by the Bianconi group on $La_{1.85}Sr_{0.15}CuO_4$ [11,12], where stripe-like patterns referring to tetragonal and orthorhombic structural elements were detected. This work prompted intense efforts in the search for heterogeneity and phase separation [13–21] in cuprates and initiated a novel understanding by showing that at least two superconducting order parameters, namely s + d, coexist [22,23]. Experimentally as well as theoretically, several papers were devoted to this subject, and its existence was convincingly demonstrated.

K. A. Müller and J. G. Bednorz began their project based on the idea that polarons in perovskites and the Jahn–Teller effect are at the origin of high-temperature superconductivity [24] (see Figure 1). Local lattice effects were a focus in the following years. Especially, the work of the Bianconi group [11,12] continued, and the coexistence of localized carriers with itinerant ones was established. Importantly, it was shown that the onset temperature T* of stripe formation carries a large oxygen isotope effect [5,10,25], which is also dependent on structurally different elements. Coinciding with this finding were local anomalous lattice effects evident from phonon anomalies and neutron scattering [26]. These results demonstrate that heterogeneity and local lattice effects have been a focus since 1986 and proposed and experimentally validated for almost 40 years.

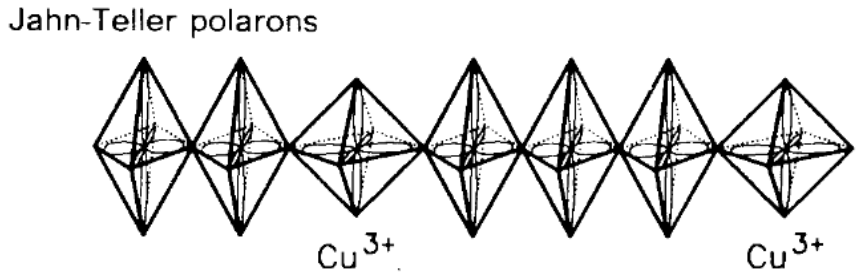


Figure 1. Schematic sketch of the Jahn–Teller polaron (from Ref. [1]).

2. Q-Balls and the Role of Local Nanoscale Lattice Fluctuations in the High-Tc Superconductivity Mechanism in Cuprate Perovskites

The recently proposed mechanism of the pseudogap state and high-Tc superconductivity in cuprates [27–29] is supported by data obtained for $HgBa_2CuO_{4+y}$ via micro-X-ray diffraction using synchrotron radiation techniques [30,31]. The quintessence of the mechanism lies in the idea that nested fermionic states in the vicinity of a van Hove singularity on the Fermi surface of the electron/hole subsystem may cause an instability in the form of local condensation of bosonic spin or charge fluctuations, which break the symmetry of the Matsubara time chirality in Euclidean space–time, forming Q-balls (nontopological solitons), as predicted by Sidney Coleman [32] for quark–gluon plasma in Minkowski space–time. These local collective bosonic fluctuations, arising via the first-order phase transition below the temperature T* [27,28], possess wave vectors that connect the nested fermionic states at the opposite 'antinodal' regions of the Fermi surface, forming superconducting condensates of Cooper/local pairs inside the Q-balls, thus lowering their energy via opening of the energy gap on the nested parts of the Fermi surface; see Figure 2. Hence,

the pseudogap phase is created, as demonstrated in Figure 3. Simultaneously, a 'strange metal' phase appears around the effective coupling strength κ^* above the T_c temperature corresponding to the top of the superconducting (half)dome in Figure 3.

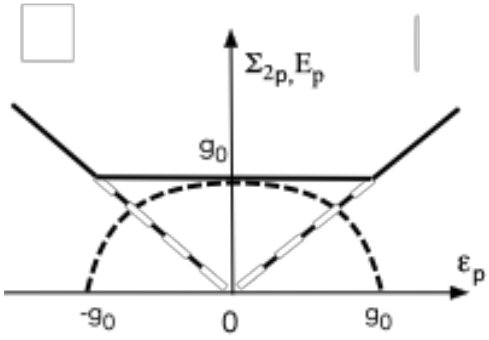


Figure 2. Anomalous self-energy Σ_{2p} (short-dashed line) and fermionic dispersion E_p (solid line) as a function of bare fermionic dispersion ε_p in the vicinity of the Fermi-level $\varepsilon_p = 0$ near the 'nested'/antinodal points of the bare Fermi surface inside a Q-ball with local superconducting (pseudo)gap g_0 (from Ref. [27]).

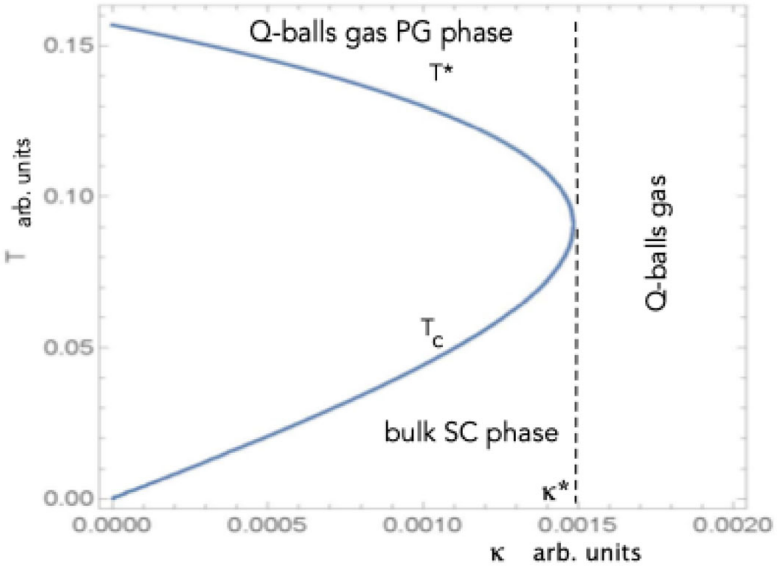


Figure 3. The phase diagram that follows from the Q-ball gas picture, where κ is effective fermion-Q-ball field coupling strength and 'PG' is pseudogap phase (from Ref. [27]).

The amplitude of the bosonic field inside the Q-ball rotates either clockwise or anti-clockwise with Matsubara frequency $\Omega = 2\pi T$ in the Euclidean space–time [27–29]. Finite radius solution for the Q-ball nontopological soliton is allowed due to conservation of the Noether 'charge' Q proportional to the number of condensed spin or charge excitations forming the Q-ball [27,32]. It was found recently [33] that linear temperature dependence of electrical resistivity arises naturally due to scattering of uncondensed fermions on the local Q-ball spin–charge fluctuations, which may explain the famous 'Planckian' behavior of the 'strange metal' phase in high-Tc cuprates observed experimentally [34]. The diamagnetic response of Q-ball gas is also calculated [33] and shows good accord with experimental data by L. Li et al. [35] in the 'strange metal' phase. The phase diagram of high-T_c cuprates with the superconducting dome touching the 'strange metal' area at optimal hole doping is also reproduced by the theory regarding the Q-ball mechanism [27–29,33]. Overall, we believe that these results provide support regarding quantum thermodynamic time crystals, Euclidean Q-balls, as a model of the nanoscale lattice heterostructure driving the superconducting properties of high-Tc cuprates. In particular, anharmonic lattice dynamics related

to Internal Quantum Tunneling Polarons (IQTP)s reviewed in Section 4 below may actually reflect the anharmonicity of the Q-ball field dynamics, which is a prerequisite of Q-ball nontopological soliton formation; see Ref. [32]. Simultaneously, evaluations in Ref. [28] indicate that, in the case of sufficiently strong Van Hove singularity in the density of states in the vicinity of the 'nested' antinodal points or the hot-spot points of the bare Fermi surface, superconducting transition due to the charge redistribution within the Sr-O_{ap} layer in the form of Q-balls would precede bulk SDW/CDW formation.

3. Local Lattice Fluctuations in Cuprates Seen by X-Ray Spectroscopy XANES and EXAFS

X-ray absorption spectroscopy (XANES and EXAFS) at synchrotron radiation has proven to be the most informative for studying the local structure of high-temperature superconductors (HTSs), which was first demonstrated by us on the $BaBiO_3$-based superconducting oxide family two years before the discovery of HTSs in cuprates [36]. Polarized XANES and EXAFS studies at the K-Cu edge of thin copper-based HTS films, irradiated with high-energy helium ions, allowed for the first time to obtain data on changes in the local electronic structure in the Cu-O_2 plane, leading to the loss of superconducting properties [37–41]. A unique experiment on the study of the electronic structure of $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$ films irradiated by He^+ ions using XANES at Cu-L_3 and Ce-$M_{4,5}$ edges was performed at the superconducting synchrotron Super-ACO (LURE, Orsay, France). It provided new information concerning the role of oxygen deficiency in the emergence of superconducting properties of electron-doped cuprates [42,43]. The low-temperature studies of the EXAFS spectra in $La_{2-x}Sr_xCuO_4$ and $Nd_{2-x}Ce_xCuO_{4-\delta}$ revealed low-temperature anharmonicity manifested as anomalous temperature dependences of the Debye–Waller factors of the Cu-O bond in the superconducting Cu-O_2 plane. The observed anomalies were explained by the oxygen ion vibrations in a double-well potential at low temperatures for both hole- and electron-doped cuprates [44–46] (see Figure 4), similar to those previously observed in the family of high-temperature superconductors based on $BaBiO_3$ [47]. The results of low-temperature studies enabled proposing a model of the mechanism of superconductivity in cuprates based on local charge carrier pairing in real space [48] by analogy with the bismuthate family [47]. The achieved understanding of the features of the local structure of cuprates made it possible to determine the correlations between the influence of nano-inclusions that increase the critical current in real 2G MOCVD YBCO tapes and the changes these inclusions introduce into the local structure of the Cu-O_2 plane of HTS-coated conductors [49,50].

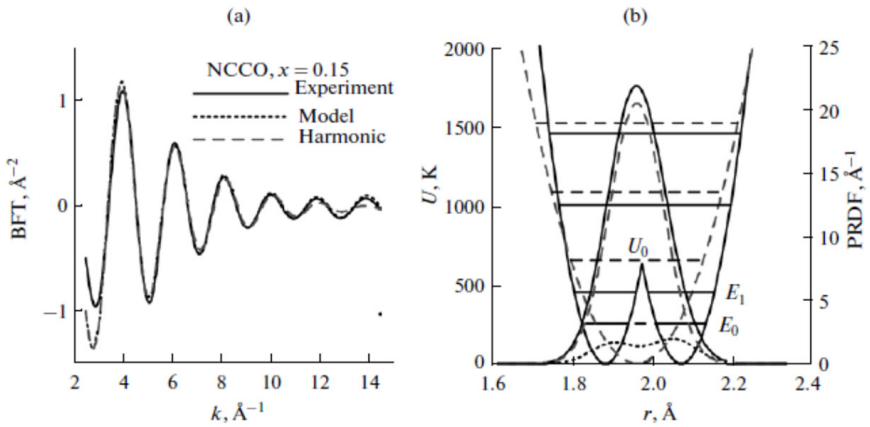


Figure 4. Panel (**a**) illustrates EXAFS oscillations above the Cu *K* edge in $Nd_{1.85}Ce_{0.15}CuO_{4-\delta}$, and panel (**b**) presents the extracted double-well potential for oxygen ion vibration (from Ref. [45]).

In addition, the local non-centrosymmetric structure of $Bi_2Sr_2CaCu_2O_{8+y}$ has been demonstrated by X-ray magnetic circular dichroism at Cu *K*-edge XANES [51].

4. Internal Quantum Tunneling Polarons' Dynamical Structure and Kuramoto Synchronization in Cuprate Superconductors

The microscopic mechanism behind high-temperature superconductivity (HTSC) in cuprate superconductors remains elusive despite decades of experimental and theoretical efforts. However, there is a growing body of evidence suggesting that charge–lattice dynamics associated with apical oxygen (O_{ap}) atoms play an important role. In this context, recent studies combining extended X-ray absorption fine-structure (EXAFS) spectroscopy and exact diagonalization modeling have provided a novel framework for understanding the coupling between local charge inhomogeneities and anharmonic lattice dynamics in terms of Internal Quantum Tunneling Polarons (IQTPs) [52–54]. IQTPs differ from conventional, e.g., Holstein–Hubbard polarons, in that the excess charge is localized on the oxygen instead of the metal ion. This adds charge transfer between neighboring oxygen ions, accompanied by the associated shift in the bond length, to the polaron dynamics. The transit through the lattice is therefore quantum tunneling rather than hopping. Recent EXAFS experiments on heavily overdoped cuprates, such as $YSr_2Cu_{2.75}Mo_{0.25}O_{7.54}$ and $Sr_2CuO_{3.3}$ [55,56], have extended the original ones [57–65] to reveal temperature-dependent transformations in the local structure at the superconducting (SC) transition not only in the Cu-O_{ap} distances and numbers but also in the Cu-Sr anharmonicity. These features are captured in real space via deviations from Gaussian pair distributions in the Fourier-transformed spectra, signaling the presence of soft anharmonic vibrational modes [57–60]. These distortions are not associated with thermal effects; instead, they emerge from the tunneling of the charge and lattice distortion between degenerate Cu-O configurations. The defining characteristic of the IQTP is therefore that the charge and atomic displacement oscillate between sites faster than the polaron's center-of-mass hopping rate, confining the dynamics within a small cluster. The defining signature of an IQTP that differentiates it from a conventional polaron is its presence in the inelastic structure, $S(Q,\omega)$, at the energy corresponding to the tunneling frequency. It is therefore observed in EXAFS that it originates in the instantaneous structure, $S(Q,t = 0)$, but not in the Bragg peaks of the diffraction pattern that are determined by $S(Q, \omega)$, that is, the elastic structure. Facilitated by its element specificity and magnetic orientation of samples, IQTPs are therefore identified in EXAFS as a two-site distribution of a Cu-O_{ap} pair that is not observed in diffraction or elastic pair-distribution functions. These features have been observed over the years in various hole-doped cuprates [61–71]. However, because of the limited number of scientists familiar with inelastic structure and EXAFS as a probe of it, it has often been attributed to static disorder or local lattice inhomogeneities to the exclusion of the importance of dynamic structure and IQTPs. Notwithstanding this opinion by the larger community, we have proceeded with interpreting multi-site Cu-O distributions that are coupled to the SC transition as a signature of collective quantum coherence. Specifically, the six-atom cluster model (see Figure 5) that comprises two Cu-Oap IQTPs coupled via a shared planar oxygen (Opl) and a soft Oap-Sr-Oap bridge exhibits a first-order phase transition triggered by an anharmonic three-phonon interaction mediated by the Sr ion, coupling the two IQTPs, to a phase-synchronized state [52–54], like the one described by the Kuramoto model in networks of coupled oscillators [67–69]. This synchronization causes an expansion of the charge distribution from its location on the apical sites into the CuO_2 plane. This increase in planar electronic density may be a requirement for pair formation. At the same time, a planar IQTP mode develops in the planar oxygen site, suggesting that coherent charge–lattice dynamics are not restricted to the dielectric layer but extend into the superconducting

planes. These findings were further confirmed by mapping the full quantum Hamiltonian of the cluster onto a Kuramoto-like mean-field equation, revealing an emergent order parameter that quantifies the internal synchronization of polarons [52–54].

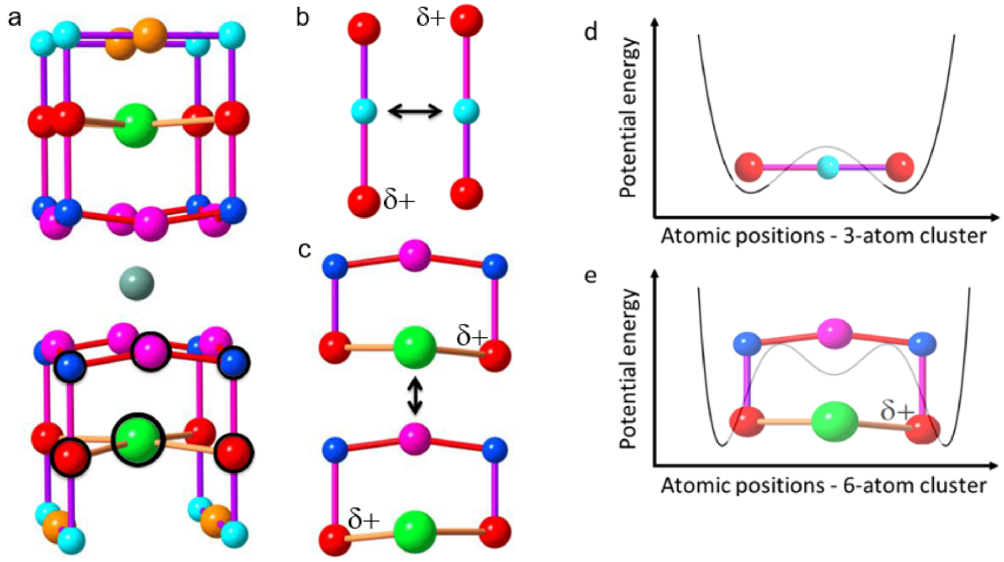


Figure 5. (**a**) The crystal structure with the general features of multilayered cuprates with higher Tc's. (**b**) The original three-atom cluster derived from the two-site distributions [64]. (**c**) The atoms circled in black in (**a**) form the six-atom cluster [52–54]. (**d**) For the three-atom cluster, the potential energy corresponds to a double-well structure [64]. (**e**) For the 6-atom cluster, the potential energy corresponds to a triple-well structure.

These results call into question the so-called "passive" role of the dielectric layer in cuprates. The traditional view of the Sr-O_{ap} layer as limited to a structural spacer or charge reservoir is challenged by evidence showing that anharmonic dynamics and charge redistribution within this layer are strongly correlated with superconducting properties [70,71]. The IQTP unifies diverse observations from EXAFS, Raman, and inelastic neutron scattering, and it offers a microscopic picture of nonadiabatic strongly coupled lattice–charge dynamics. Moreover, the synchronization of IQTPs offers a route to understanding how local lattice dynamics involving apical oxygen can participate in the high-Tc mechanism. An essential next step is to determine whether the coherence inherent to the entire six-atom system extends further, where it may promote the formation of the coherent superfluid. By bridging real-space structural probes with quantum synchronization theory, this approach opens new avenues for interpreting complex behavior in quantum complex matter and motivates the design of new compounds and techniques where local dynamical coherence may be tuned.

5. Polarons Nanoscale Self-Organization in Cuprate Perovkites as Seen by X-Ray Absorption Spectroscopy

The polaronic states are related to the breakdown in quantum complex materials of the Born–Oppenheimer approximation used in standard solid-state physics to separate the motions of free electrons and nuclei. In 1986, Alex Müller and Georg Bednorz proposed that high-temperature superconductivity appears in the complex phase of doped cuprate perovskite oxides by condensation of Jahn–Teller bipolarons [72–75] expected near metal–insulator transitions in strongly correlated electronic systems [5], which have been observed recently [76,77]. The CuO_2 plane in doped cuprates was described as a mixed-valence phase of a transition metal oxide composed of a background of Cu^{2+} ($3d^9$) states and Jahn–Teller polaron associated with hole doping. To underpin the complex phase

behavior of polarons controlled by the interplay between charge carriers and lattice distortions, X-ray absorption spectroscopy techniques, particularly extended X-ray absorption fine-structure (EXAFS) and X-ray absorption near-edge structure (XANES), emerged in the 1980s as powerful tools to directly probe the local lattice environment around copper sites in these materials [11,78–82].

The microscopic lattice evidence for self-organization of pseudo-Jahn–Teller polarons [72,83] associated with the hole hoping formed by Cu($3d^9$)O($2p^5$) singlet configuration called $3d^9\underline{L}$ (where $\underline{L}$ indicates a ligand hole in the oxygen plaquette) formed by mixing of Cu $3d_{x^2-y^2}$ and $3d_{z^2}$ orbitals with b_1 and a_1 orbitals of the oxygen plaquette with the polaronic pseudo-Jahn–Teller local lattice distortions (LLDs) organized at nanoscale was provided by a very fast and local experimental method involving X-ray absorption spectroscopy [83]. Central to polaron detection, two distinct Cu–O planar bond lengths within the CuO_2 planes of $La_{1.85}Sr_{0.15}CuO_4$ were revealed by polarized Cu K-edge EXAFS at low temperatures below approximately 100 K. This splitting signals the coexistence of two types of CuO_6 octahedra: the first one with a small tilt of rectangular CuO_4 planar plaquette (LTO-like) with a long Cu-O_{ap} distance and the second one with a rhombic CuO_4 planar plaquette with a larger tilt angle (LTT-like) and short Cu-O_{ap} distance, corresponding to regions where doped holes locally trap lattice distortions, thus forming D stripes of condensed pseudo-Jahn–Teller polarons (see Figure 4). The probability of (LTT-like) domains appears to be consistent with a substantial fraction (~33%) of the rhombic CuO_4 plaquettes developing local distortions in D stripes and (~66%) LTO-like plaquettes forming U stripes. Such coexistence creates a superlattice of striped phases called a "superstripe" scenario: nanoscale arrays of distorted D and undistorted U lattice stripes. The superstripe scenario is thought to be a real-space manifestation of polaronic charge ordering reported quite independently by Goodenough [84,85] and predicted by Kusmartsev et al.'s theoretical analysis [86]. These local lattice distortions are connected with the emergence of a striped phase, evidenced by complementary diffuse X-ray scattering measurements that reveal superlattice modulations indicative of nanoscale periodic lattice distortions. The Bianconi–Perali–Valletta (BPV) theory of high-Tc superconductivity [87] was developed on the basis of the phenomenological data on the nanoscale geometrical parameters of the observed topology of the stripe phase shown in Figure 6. This theory predicted the Tc amplification by the Fano–Feshbach shape resonance in multigap superconductivity, where nanoscale superconducting units are quantum-confined by intercalated normal units disclosed in the patent [88], initiating the material design of artificial high-Tc superconductors [89–93]. The core of the success of BPV theory [87–93] is the quantitative formulation of the configuration interaction between a first BCS condensate and a second condensate in the crossover BEC–BCS regime in multigap superconductivity. The smoking gun of the success of BPV theory is the anisotropic Fano line shape of the drop of superconducting T_c as a function of temperature, which shows a characteristic minimum at the Lifshitz transition for the appearance of a new Fermi surface i.e., the flat band. On the contrary, the maximum of T_c occurs at a second Lifshitz transition for opening a neck in the new Fermi surface, and the width of the shape resonance is provided by the characteristic energy range of the boson energy in the pairing process.

It has been shown that nanoscale phase separation could be associated with the proximity of the Fermi level to a Lifshitz topological transition for the appearance of a new Fermi surface in a correlated multi-band system [94]. The polaronic stripes in cuprates reflect a charge density wave intertwined with orbital and bond-order waves, a complex electronic–lattice ordering stabilized by strong anisotropic electron–phonon coupling, as detailed in [76,95], which emphasizes that the polaronic CDW phase in cuprates can be associated with a generalized Van der Waals phase separation thermodynamic model

where doped holes behave as interacting extended objects exhibiting directional bonding tendencies around a critical hole doping level of 1/8. Below this doping, the system forms localized Mott insulating puddles coexisting with striped polaronic CDW puddles, while, above this doping range, striped CDW puddles coexist with metallic (Fermi liquid) puddles, yielding a nanoscale phase separation landscape. The model accounts for these phase coexistences as a gas-to-liquid-like transition in a polaronic charge fluid and is supported by X-ray absorption and diffraction data, demonstrating the intricate pattern of puddles and stripes in perovskites. Furthermore, experimental findings show that the CDW onset temperature with its lattice character can possess a large isotope effect [96,97], strongly implicating lattice vibrations in the stabilization of polaronic CDW phases. Additionally, the misfit strain between the active copper oxide layers and spacer layers strongly influences the local lattice distortions and promotes an arrested phase transition, manifesting as a correlated quantum disorder phase of stripe puddles in perovskites.

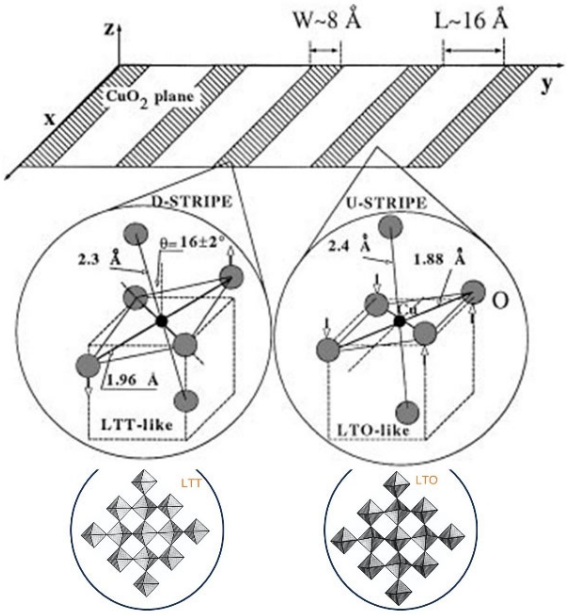


Figure 6. Pictorial view of the nanoscale phase separation of stripes composed of first units, polaronic doped distorted CuO_6 octahedra, left side, with the "LTT-like" structure assigned to the doped distorted D stripes of width W and second units of the undistorted octahedra, right side, of the undoped "LTO-like" undistorted U stripes of width L. The period of the superlattice of quantum stripes is d = L + W = 2.4 nm, and the quantum geometry factor L/d at optimum doping shows the magic value 2/3 = 0.66.

Crucially, EXAFS and XANES are sensitive probes, detecting the amplitude of the periodic lattice distortion and its spatially heterogeneous distribution, thereby providing direct evidence for intrinsic nanoscale electronic and lattice inhomogeneity, which has been unveiled in the superconducting overdoped cuprate [98–102].

The resulting "superstripe" phase exhibits complex multiscale patterning across length scales ranging from the atomic to mesoscale, reflecting a frustrated electronic and lattice phase separation where polaronic and metallic regions coexist. This arrested phase separation scenario helps to explain the broad transition widths and the competition between superconductivity and charge ordering.

Collectively, the combination of EXAFS/XANES results and complementary diffraction studies consolidates the viewpoint that polarons in cuprates manifest as lattice-localized distortions that organize into nanoscale striped CDW puddles, intimately linked to the physics of the Mott insulator-to-metal crossover and high-temperature superconductivity itself. The detailed temperature- and doping-dependent evolution of these polaronic

distortions documented in the referenced studies provides a consistent microscopic foundation for understanding the complexity of cuprate phase diagrams, emphasizing the essential role of lattice degrees of freedom concomitant with strong electron correlations. Recent advances have presented a roadmap to high-temperature superconductivity by tailoring nanoscale lattice heterostructures.

The comprehensive spectroscopic evidence underscores that superconductivity in cuprates emerges from a complex nanoscale electronic–lattice landscape, where polaronic charge density waves and spatial phase separation are fundamental elements, as revealed uniquely by X-ray absorption spectroscopy techniques.

6. Conclusions

High-temperature superconductivity is primarily a subject within materials science [102], focusing on the synthesis of new materials with higher superconducting transition temperatures. The field has captivated researchers because experimental results indicate that quantum entanglement at high temperatures can result in macroscopic superconductivity in particular mesoscopic complex quantum matter. Polaron self-organization has been clearly observed in manganites, nickelates and other compounds [103–110].

The theory of polarons in cuprates was developed by Nobel Prize winners like Goodenough and Heeger [84,85], and by Alexandrov, Mott, Devreese, Salje, Ciuchi, Perali, and Capone [111–152], supported by K. Alex Muller and other experiments [153–165]. In cuprates, fast and local methods have clearly shown the presence of lattice and electronic inhomogeneity, phase separation, and the coexistence of local polaronic and itinerant states [166–171]. These materials exhibit a complex phase of quantum condensed matter showing nanoscale phase separation [12,17,29,33,90,93] and lattice quantum heterogeneity with nanoscale building blocks [172–197]. Moreover, the investigation regarding the fundamental role of local lattice effects in high-Tc superconductivity [198] has stimulated advances in cuprate twistronics for quantum hardware [199–201].

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