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Q-Balls in the Pseudogap Phase of Superconducting $HgBa_2CuO_{4+y}$

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Abstract: Fast and local probes, such as X-ray spectroscopy, X-ray diffraction (XRD), and X-ray microscopy, have provided direct evidence for nanoscale phase separation in high temperature perovskite superconductors composed of (i) free particles coexisting with (ii) Jahn Teller polarons (i.e., charges associated with local lattice distortions) not detected by slow experimental methods probing only delocalized states. Moreover, these experimental probes have shown the formation of a superstripes phase in the pseudogap regime below T* in cuprates. Here, we focus on the anomalous temperature dependence of short range X-ray diffraction CDW reflection satellites with high momentum transfer, probing both charge and lattice fluctuations in superconducting $HgBa_2CuO_{4+y}$ (Hg1201) in the pseudogap regime below T* and above T_c. We report compelling evidence of the anomalous anticorrelation of the coherence volume with the peak maximum amplitude of the CDW XRD satellite by cooling below T*. This anomalous temperature trend of the short-range striped Jahn Teller polaronic CDW puddles is in agreement with predictions of the Q-ball theory of the quark gluon plasma extended to cuprates, providing compelling evidence for non topological soliton puddles of striped condensate of pairs in the pseudogap phase.

Keywords: high temperature superconductivity; synchrotron X-ray diffraction; charge density waves; Q-balls

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1. Introduction

Recently, it has been proposed that the physics of Euclidean Q-balls developed in the frame of the quark-gluon plasma inside a proton in the atomic nucleus could be extended to the physics of complexity in high-temperature superconductivity [1]. In cuprates, short-range charge density waves (CDW) forming a supersolid phase called superstripes [2–4] have been observed by joint experiments of temperature-dependent X-ray diffraction (XRD) and scanning micro-XRD [2]. The superstripes phase [3,4] was also unveiled by X-ray absorption spectroscopy and anomalous diffraction experiments probing the condensation of polaronic charge density waves in striped puddles. Polarons involving localized charges with associated local lattice distortions, proposed by Bednorz and Müller [5], have been confirmed by experiments probing short-range order [6]. The X-ray absorption fine structure (EXAFS) [7,8] in X-ray spectroscopy, probing fast and local bond fluctuations, has been applied to cuprate superconductors, providing information on the polaron anisotropy, shown to be of pseudo-Jahn-Teller type, and on the large polaron size extending over eight copper sites [9–11]. The isotope effect [12] has been observed also at the pseudogap

temperature [13,14] associated with the onset of polaron ordering using X-ray absorption near edge structure (XANES) [15] a fast and local probe of many local body electronic configurations [16] and higher order atomic correlations in a nanoscale atomic cluster around Cu ions [17]. Pseudo-Jahn Teller polarons in cuprates have been confirmed by Goodenough et al. using thermopower experiments [18–22] probing electron-lattice vibronic coupling and heterogeneous charge fluctuations, forming a complex nanoscale phase separation as in colossal magneto-resistance manganite perovskites [23]. Nanoscale phase separation was predicted theoretically [24], and it has been the topic of two important workshops [25,26]. The complex landscape with the coexistence of two electronic components (free and localized charges) was confirmed by several experiments [27]. The experiments pointed toward the coexistence of undistorted stripes of charges in a strongly correlated doped charge-transfer Mott insulator with distorted stripes of polaronic charges. These experimental results provided the basis for the proposal of Bianconi-Perali-Valletta (BPV) theory in 1997 of multigap superconductivity in a striped nanoscale ultrastructure composed of a superlattice of quantum stripes. Here, the multigap superconductivity is generated by quantum size effects forming quantum minibands. The T_c amplification is driven by a Fano resonance due to configuration interaction between the first open pairing scattering channel forming BCS Cooper pairs composed of free particles in the first miniband and a second closed scatting channel forming pairs in the crossover of the *Bose-Einstein* Condensation (BEC) and *Bardeen–Cooper–Schrieffer* (BCS) (called the BEC-BCS crossover) with the formation of bipolarons in the intermediate coupling regime associated with strong electron-lattice interaction in the upper miniband. The coexistence of a first BCS condensate in the first miniband and a second BCS-BEC crossover condensate in the second miniband appears at a topological Lifshitz transition [28]. The discovery of high temperature superconductivity in layered multigap MgB_2 [29] confirmed the theoretical prediction of the amplification of the critical temperature also for a superlattice of quantum wells.

In this complex landscape, the essential ingredient in the mechanism of high-temperature superconductivity is the coexistence of two electronic components: (i) delocalized and weakly interacting itinerant particles, which coexist with (ii) localized and strongly interacting polarons. In perovskite materials, the polarons are formed by doped holes with the associated cloud of phonons and the formation of pairs of polarons. Using extended X-ray absorption fine structure (EXAFS) and X-ray absorption near edge structure (XANES), the probability distribution function of the instantaneous Cu-O bond length of the local lattice geometry has been determined, showing large nanoscale anisotropic pseudo-Jahn-Teller polarons forming instantaneous polaronic short-range CDW, which coexists with a Fermi liquid.

The formation of striped puddles of ordered polarons is driven by elastic attractive polaron-polaron interaction, which is zero when the polarons are in contact and it increases with the polaron-polaron distance. This attractive polaron-polaron force in hole-doped cuprate superconductors is analogous to the strong nuclear force between quarks forming the Q-balls, which competes with the repulsive Coulomb force between charges and determines the charge and the size of the short-range polaronic CDW puddle [30–33].

Using thermoelectric power experiments and thermal conductivity, Goodenough et al. have supported these results by showing vibronic strong electron-phonon coupling with a nonadiabatic regime, large low-symmetry polarons, and the formation of a striped lattice pattern, and they pointed out the similarity of nanoscale phase separation in manganites and cuprates with a striped texture [34–36].

Experiments of Müller, Shengelaya, Keller, Conradson, Mustre de Leon et al., and theory work of Bussmann-Holder, de Gennes, Deutscher, Bishop, Gorkov, Teitelbaum, and Kresin et al. have provided further evidence for the key role of local lattice fluctuations, complexity and charge and lattice heterogeneity in high-temperature superconductors [37–52]. In this paradigm, supported by a large set of different experimental methods and many different authors, the origin of the pseudogap phase below T* and above Tc was assigned to the polaron coherence in high temperature superconducting cuprates with the formation of

isolated condensed nanoscale puddles of local pairs called superstripes [3,4]. EXAFS experiments have unveiled the double-well potential for oxygen vibration in the superconducting perovskite $Ba_{1-x}K_xBiO_3$ with the formation of a Fermi-Bose mixture with stripe-like nanoscale structural phase separation in superconducting in $BaPb_{1-x}Bi_xO_3$ [53–55]. Strong electron-lattice interaction, in particular hot spots in the k-space has been observed also in pressurized sulfur hydrides [56]. This confirms the universal scenario of two-component superconductivity in systems with strong unconventional electron-lattice interaction and ubiquitous unconventional short-range structural fluctuations and percolation, as has been confirmed recently in cuprates [57,58]. Recently, static CDW, long-range CDW, short-range CDW, and dynamic charge fluctuations have been observed by resonant X-ray scattering in cuprates pervading the full phase diagram [59–71] of cuprates and nickelates [72].

The proposed Euclidean Q-ball phase [1] may explain the pseudogap (PG) phase of high-T_c superconductivity in hole-doped high-T_c cuprates that precedes the multigap high-T_c superconducting phase with one particular large gap due to polaronic pairing in a strong coupling regime. This theory is based on a new physical mechanism for binding the fermions into local pairs via exchange with semiclassical density fluctuations of finite amplitude inside the Q-balls. The charge fluctuations inside the Q-balls possess a local minimum of potential energy at finite amplitude and, therefore, provide greater binding energy of fermions into local pairs than usually considered and also exchange infinitesimal spin-waves, charge-density fluctuations, or polaronic charge density waves (CDWs) in the Fröhlich picture. At couplings stronger than some critical values, local pairs percolate between Q-balls, forming a large superconducting cluster. We have used temperature-dependent X-ray diffraction (XRD) to test the theoretical predictions of the Q-ball temperature evolution [1]. We have selected a putative Q-ball reflection at a high momentum transfer [73,74], characterized by the reflection peak amplitude A being much higher than that of static weak CDW, with coherence volume V_{coh} smaller than that of static weak CDW appearing at a small momentum transfer, and the temperature onset at T* being much higher than that of static CDW $T_{CDW,}$ showing an anomalous temperature dependence of both the reflection peak amplitude A and V_{coh}. The measured temperature variation in the pseudogap phase has been found to agree with the prediction of the Q-ball theory [1], allowing us to attribute the nature of the selected short-range CDW to the presence of Q-balls in the pseudogap phase.

2. Results and Discussion

In this work, we focus on the puddles of short-range dynamical charge density fluctuations in the pseudogap phase in a high temperature superconductor, oxygen-doped $HgBa_2CuO_{4+y}$ (Hg1201). Hg1201 has a simple tetragonal average structure [75–87] with an optimum Cu-O bond length of 194 pm. It shows the self organization of dopants composed of atomic stripes of mobile oxygen interstitials (O-i) [81,82] running in both the horizontal (100) and vertical (010) directions in the ab plane. The nanoscale phase separation is composed of first puddles rich in O-i stripes that are anticorrelated with second puddles showing short-range dynamic charge density waves (CDW), which have been visualized by scanning micro-X-ray diffraction [2]. The $HgBa_2CuO_{4+y}$ single crystals have been grown [76] with a final oxygen treatment to establish a y concentration of oxygen interstitials of approximately y = 0.12, showing the superconducting optimum critical temperature Tc of 94 K [75–78]. The crystal structure has been determined by standard X-ray diffraction. The crystal structure has P4/mmm symmetry with lattice parameters a = b = 0.3886 (5) nm and c = 0.9517 (2) nm at T = 100 K (numbers in parentheses indicate the standard deviation of the last digit, in agreement with reference [79].

Short-range dynamic CDW puddles have been investigated by X-ray diffraction measurements performed at the XRD1 beamline of the ELETTRA synchrotron facility in Trieste, Italy. The charge modulation gives rise to clear superlattice reflections [1]. We have identified a particular satellite of a main Bragg diffraction peak assigned to a short-range dynamic CDW order in Hg1201, tuning the photon energy at 17.6 keV with a beam size

of 200 × 200 μm^2. The selected short-range dynamic CDW is located in the k-space at q_{CDW} = (0.23, 0, 0.16) around the (1, 0, 18) Bragg reflection.

We used a liquid nitrogen cryostat whose flux on the sample provided a variable temperature measured with an uncertainty of 1K. The cooling ramp was set with a step of 1K and a thermal waiting time of 10 min between two successive measurements. In order to reduce the temperature uncertainty, we averaged the collected data for every three measurements. The error bar at each temperature corresponds to the standard deviation of each group of three measurements.

To get a direct view of the temperature dependence of the short-range CDW-satellite reflection in the temperature range 85 < T < 280 K, we show in Figure 1 the two-dimensional color plots of the CDW-peak profile along the a* (top panel) and c* (bottom panel) reciprocal lattice directions as a function of temperature. The selected peak appears as the sample was cooled below 240 K, which is close to the onset of the pseudogap phase T*.

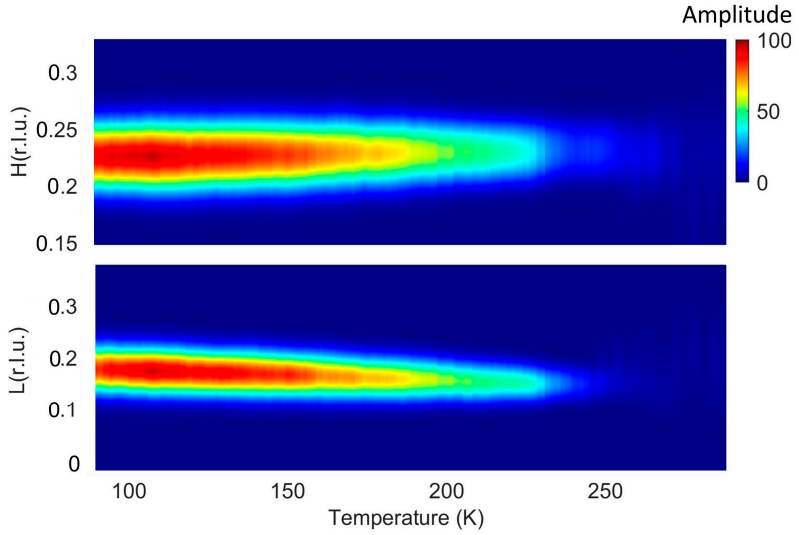

Figure 1. Color plots of temperature evolution of short-range CDW or charge fluctuations XRD profiles along (upper panel) a* and (lower panel) c* directions of reciprocal lattice around the (1, 0, 18) Bragg peak.

The CDW-peak amplitude A at wavevectors q_{CDW}(a*), q_{CDW}(c*), and full widths at half maximum, Δq_{CDW}(a*), and Δq_{CDW}(c*), along both the in-plane a* (H) and out-of-plane c* (L) directions, have been extracted by fitting the CDW profiles with a Gaussian function after background subtraction. The coherence lengths ξ_a and ξ_c have been calculated as $\xi_a = a/\Delta q_{CDW}$(a*) and $\xi_c = b/\Delta q_{CDW}$(c*), where a and c are the crystallographic axes. The short-range dynamic CDW satellite shown in Figure 1 is assumed to be a putative Q-ball, which appears at the pseudogap temperature T*. This temperature is higher than the temperature onset, Tcdw = 159 K, of the long-range static CDW weak reflections. In fact, static long-range CDW satellites are observed by resonant Cu L_3 X-ray scattering at small momentum transfer near the l = 1 main reflection in the approximate range from 0.26 to 0.29 r.l.u. [87]. Therefore, the selected satellite reflection is assigned to a short-range dynamical CDW detected in ref. [69] which is assigned to Q-balls made of pseudo- Jahn Teller polarons [88,89].

The CDW peak amplitude A, indicating the population of CDW puddles, reaches a maximum at T = 100 K, and then undergoes a drop associated with the onset of superconductivity at T = T_c, as shown in the three panels of Figure 2. The in-plane puddle size given by the coherence length ξ_a (along the a-axis) and out-of-plane ξ_c (along the c-axis) of CDW puddles can be inspected in Figure 2a,b, respectively. We observed smaller CDW puddles

along the c-axis. In Figure 2c, we report the coherence volume of the CDW puddles as given by:

$$V_{coh} = \xi_a \cdot \xi_a \cdot \xi_c \quad (1)$$

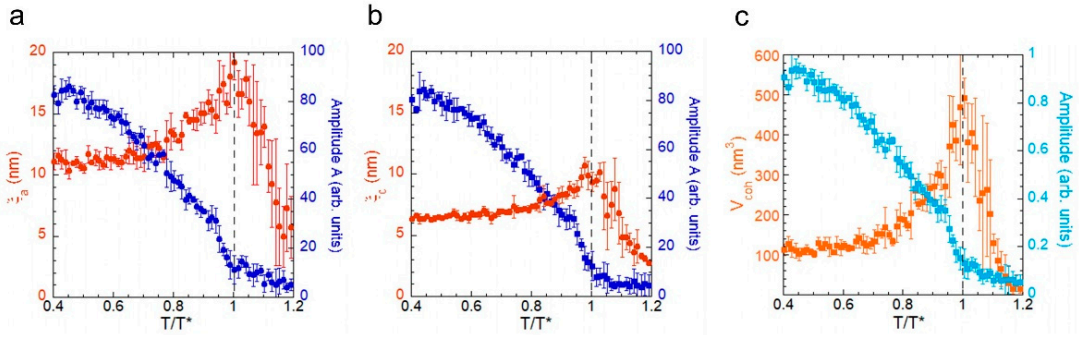

Figure 2. The short-range dynamic CDW-peak amplitude and coherence lengths as a function of the reduced temperature T/T* (**a**) along the a-axis (ξ_a) and (**b**) along the c-axis (ξ_c). T* = 240 K is the onset temperature for CDW in Hg1201. (**c**) Coherence volume, V_{coh}, with the amplitude peak as a function of reduced temperature. The vertical dashed lines indicate the onset of reduced temperature for CDW where T = T*.

Recently, a new theory of the Euclidean Q-ball phase has been proposed [1]. It was demonstrated analytically that the Euclidean action of the strongly correlated electron system may possess stable saddle-point configurations in the form of finite-size puddles (Q-balls) with superconducting density fluctuations coupled to oscillating Matsubara-time fluctuations of charge or spin.

This Q-balls scenario is reminiscent of the famous Q-balls formation in the super-symmetric standard model, where the Noether charge responsible for the baryon number conservation is associated with the U(1) symmetry of the quarks' field [1]. In condensed matter, the Q-Balls can be associated with the short-range charge density wave puddle; thus, we call Q-ball charge according to the Q-ball theory [1,90] as:

$$\text{Q-Ball} = T \cdot A \cdot V_{coh} \quad (2)$$

where T is the temperature, A is the CDW peak amplitude, and V_{coh} represents the coherence volume, given by Equation (1).

In Figure 3a, we show the behavior of the coherence volume V_{coh} as a function of the amplitude normalized to its maximum value. This behavior is well described by a power law $V_{coh} = C\ (A/A_{max})^{-\beta}$ where C is a constant and β is the critical exponent equal to 1.0 as predicted by the theory [90]. In Figure 3b,c, we report the temperature evolution of the product AV_{coh} and the temperute-dependent Q-Ball, respectively. The comparison of the Q-Ball temperature-dependent charge given by $T\ V_{coh}\ A$ measured in this experiment (black dots) with the theoretical curve calculated by Mukhin [90] (red curve) shows a very good agreement below T* = 240 K, which is the critical temperature for the evaporation of the Q-ball.

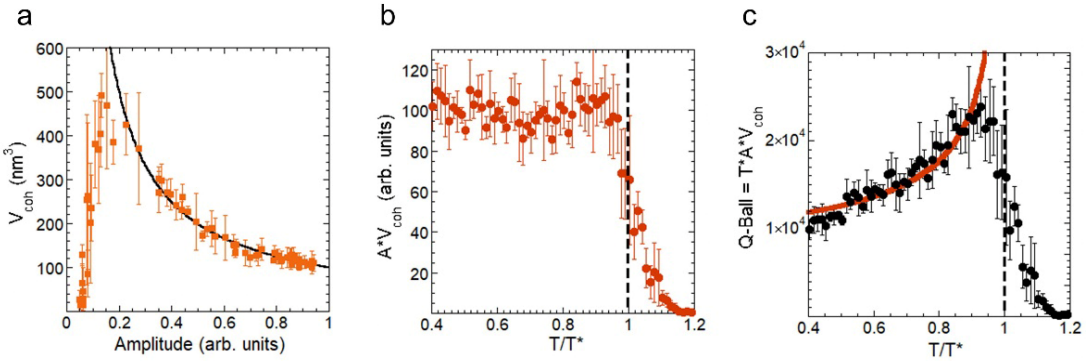

Figure 3. (**a**) Coherence volume, V_{coh}, as a function of the normalized CDW peak amplitude, A/A_{max}. The black line represents the fitting curve following the power-law $V_{coh} = C\ (A/A_{max})^{-1}$. (**b**) Coherence volume, V_{coh}, multiplied by CDW amplitude, A, and (**c**) Q-Ball = T V_{coh} A, as a function of the reduced temperature T/T*, where T* = 240 K. The red line represents the theoretical modeling elaborated by Mukhin [90].

3. Conclusions

We have used synchrotron radiation and X-ray diffraction to measure the short-range dynamic charge density waves (CDW) puddles in the optimum-doped $HgBa_2CuO_{4+y}$ with y = 0.12 and T_c = 94 K. We have found a short-range incommensurate CDW reflection with wavevector q_{CDW} = (0.23, 0, 0.16) around the Bragg peak (1, 0, 18) below T* = 240 K, which is assigned to a dynamic short-range CDW. We have extracted the CDW peak amplitude, wavevector, and coherence length as a function of the temperature from room temperature down to 85 K. The experimental results on the temperature evolution of the dynamic short-range CDW puddles in the superconducting $HgBa_2CuO_{4+y}$ may be interpreted in terms of the Euclidean Q-Balls theory [1,90]. Finally, after decades of experimental and theoretical research on high-temperature copper perovskites, the complexity of these systems is due to a nanoscale phase separation composed of coexisting (i) atomic wires of oxygen interstitials with a scale-free distribution, (ii) static long-range CDW puddles, observed in resonant X-ray scattering at low momentum transfer [87], and (iii) dynamic short-range Q-balls, which have been called charge density fluctuations [69], which show similarity with the case of doped $La_{2-x}Sr_xNiO_4$ where dynamic short-range CDW coexist with quasi-static long-range CDW in different spatial locations [72]. Further and more extensive experimental work investigating the spatial distribution and time evolution of the short-range polaronic CDW puddles is in progress to enforce the proposed analogy between the Q-ball scenario and short-range CDW puddles appearing at the pseudogap temperature, T*, in cuprates.

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