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Enhanced cuprate superconductivity from elongated to compressed coordination

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ABSTRACT

In 1986 Professor Müller and Bednorz discovered the historic superconductor of LaBaCuO (La214) that opens the prelude of unprecedented era for high T_c superconductivity research worldwide. The Jahn Teller effects combining with strong interlayer Coulomb interactions lead to the elongated copper oxygen coordination in La214, giving rise to the orbital order scenario that the Cu3dx^2-y^2 locates near Fermi surface while the 3dz^2 is far below without contributions to the conductivity, hence single band model is favorite. We recently discovered a new type of cuprate superconductor Ba_2CuO_{4-y} (Ba214) synthesized at extreme high oxygen pressure that shares a similar crystal structure to the classical La214 but with notably a very rare compressive type octahedron. The compressed octahedron coordination results from the longer in plane CuO bond length than that for apical oxygen bond. The compressed octahedron will lead to a reversed orbital order, i.e. the 3dz^2 will be lifted above 3dx^2-y^2,which is generally considered unfavorable to superconductivity based on the current theories of high T_c cuprates. Ba214 however exhibits bulk superconducting transition with T_c above 70 K at ambient pressure, a surprisingly 80 % enhancement from that for isostructural La214. Here we briefly introduce the unique structure features as well research progress for Ba214, indicating the paradigm shifts roles of 3dz^2 orbital in (I) enhancing cuprate superconductivity, (II) favoring multiband scenario; (III) enabling the *s* wave pairing mechanism.

1. Introduction

$La_{2-x}Ba_xCuO_4$ (La214) is the first copper oxide superconductor discovered by Professor Müller & Bednorz in 1986 [1]. Despite three decades of extensive and in depth research on high T_c cuprate superconductors [2–68], there are still many challenges remained, especially how to further enhance superconducting transition temperature & unveiling the mechanism are selected to be across century puzzles for condensed matter physics. Recently we discovered a new type of single layer cuprate superconductor Ba_2CuO_{4-y} (here briefly Ba214) [18] based on improving the extremely high oxygen pressure synthesis technique to tens of gigapascal (GPa) scale with simultaneously thousands of degree temperature [19]. Coincidentally the crystal structure of Ba214 material is the same as La214 of the first cuprate superconductor discovered by Professor Müller [1,11], i.e. both Ba214 & La214 crystalize into a K_2NiF_4 type principle structure. However in La214 and other cuprates superconductors the strong Jahn Teller effect of Cu^{2+} combining with the strong interlayer Coulomb interaction results in an elongated CuO octahedron along the *c* axis. The elongated coordination will in turn generate the orbital configuration with Cu3dx^2-y^2 far above Cu3dz^2 with the former near Fermi surface. This is the universal scenario for all cuprate superconductors. For the Ba214 on the other hand the larger Ba ion expands the CuO plane, resulting in a rare compressed copper oxygen octahedron configuration. The compressed coordination will generate the unusual reversal orbital configuration with Cu3dz^2 moving above Cu3dx^2-y^2. The feature of the orbital reversal is considered to be unfavorable for superconductivity in the single band model [2,14-16]. Ba214 however exhibits a T_c above 70 K at ambient conditions as evidenced from the measurements of the sample recovered from high oxygen pressure synthesis. The unexpected high Tc superconductivity in Ba214 with compressed coordination thus challenges the traditional understanding of single band *d* wave superconductivity based on quasi two dimensional CuO2 plane, leading to multiband scenario as well as *s* wave paring mechanism perceived by Professor Müller & his colleagues [11,12,17]. The unusual compressed coordination structure and high T_c of Ba214 superconductor hence sparked widespread interests [22–48].

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We here briefly present the features of Ba214 as well as recent progress on the topic.

2. Superconductivity & structure

Ba214 material is synthesized using self oxidizing high oxygen pressure & high temperature technique [18,19]. It is subjected to 18 GPa high oxygen pressure with simultaneously 1000℃. The samples can be recovered to ambient conditions for characterizations of their properties. We demonstrated its superconductivity through magnetization, μSR, specific heat experiments, *etc.* [18]. Its T_c exceeds 73 K more than 80 % higher than that for the isostructural La214 compounds [20]. The superconductivity measurements of Ba214 are shown in Fig. 1. The structure of Ba214 was determined through room temperature x ray diffraction (XRD) and both room temperature & low temperature neutron diffractions. No structural phase transition was observed down to 6 K, while the neutron diffraction refinement results are in good agreement with XRD in terms of lattice parameters. The average CuO bond length for Ba214 can reach a fairly large value of ~2.00 Å in CuO plane because of the expansion from very large Ba ion while ~1.86 Å for the apical oxygen distance obtained from the structural refining results at room temperature. The 2.00 Å in plane CuO bond length of Ba214 is the record long among known hole doped cuprates, while the 1.86 Å of apical distance is the shortest known so far. The CuO bond lengths for typical monolayer cuprates & bilayer cuprates are shown in Fig. 2. Ba214 is the only structure with compressed type CuO octahedral coordination. The x ray absorption spectroscopy studies confirm that the principle electronic structure in Ba214 is resemble to that for La214, i.e. doped Mott insulator [18].

For the elongated octahedron of the rest of cuprates, the two degenerates Cu e_g orbitals will split into higher energy $3dx^2$-y^2 and lower energy $3d3z^2$-r^2 orbital as a result of Jahn Teller distortion plus strong interlayer Coulomb interactions. This orbital order scenario is universal for all cuprates superconductors [2] before Ba214 bulk superconductor was discovered. It is believed that the topmost planar x^2-y^2 orbital contributes to superconductivity while the doped holes mainly reside in the hybrided $3dx^2$-y^2 orbital hybrided with O $2p$ orbital without contribution from z^2 orbital. The cuprate superconductors with elongated copper oxygen coordination exhibit a quasi two dimensional feature and d wave symmetry [2]. However in the compressed version of Ba214 the z^2 orbital is greatly lifted and even flipped with the x^2-y^2 orbital as shown in Fig.3. Hence there will be significant contributions from the z^2 orbital near the Fermi surface. The involvements of z^2 orbital thus show more three dimensional features, multiband like behavior & s wave like paring mechanism similar to iron based superconductivity.

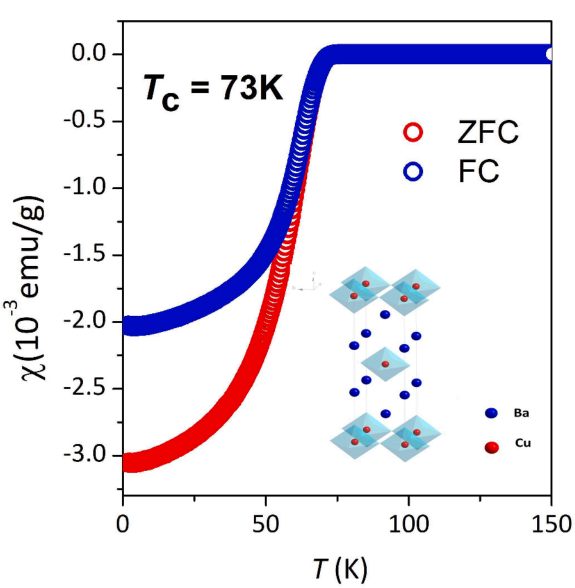

Fig. 1. Ba_2CuO_{4-y} *dc* magnetic susceptibility both of temperature dependence in zero field cooling (ZFC) and field cooling (FC) modes at ambient conditions [18]. The inset is its schematic crystal structure that is isostructural to $La_{2-x}Ba_xCuO_4$ but with a compressed copper oxygen coordination.

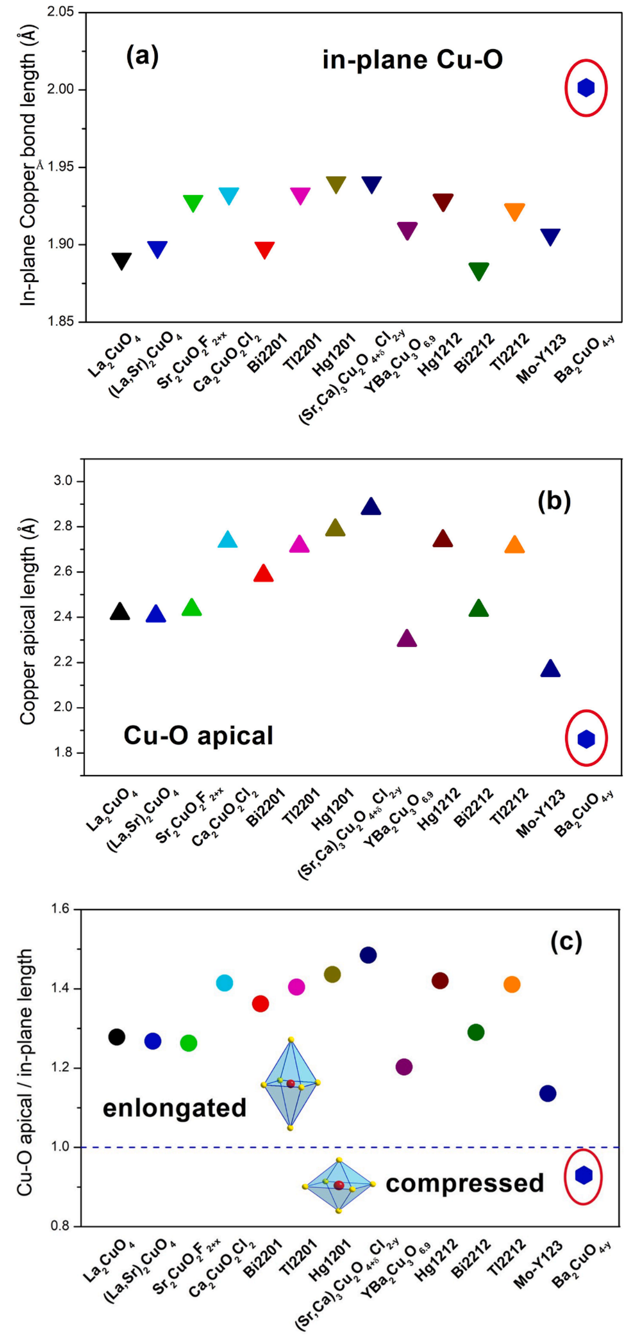

Fig. 2. Bond lengths for typical monolayer & bilayer cuprates superconductors taken from references [59–67] in comparison to the value in Ba214: (a) in plane CuO, (b) apical bond length and (c) the ratio between the apical and in plane bond length.

3. Unconventional pairing mechanisms

The Jahn Teller effect is crucial in the discovery of cuprate superconductors by Professor Müller [1,13]. Professor Müller with his colleagues emphasized in subsequent studies the inconsistencies between a

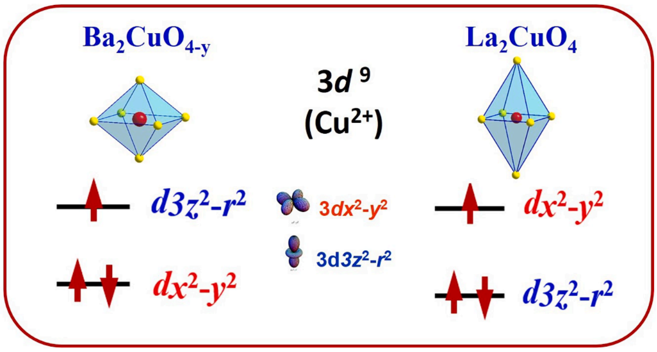

Fig. 3. The distinct $3dz^2$ orbital level of Cu^{2+} ($3d^9$) between the compressed (Ba214) and elongated (La214) octahedron. In Ba214, the $3dz^2$ orbital is expected to be located above the $3dx^2$-y^2 orbital, in sharp contrast to the universal scenario in La214 or other high T_c cuprates with elongated CuO octahedral coordination.

single d wave order parameter and experimental results [11,12,17]. In Ba214 the rare compressed distortion will give rise to the lifted $3dz^2$ orbital towards the Fermi level and thus a significant contribution from the $3dz^2$ orbital to the Fermi surface. This implies that unconventional pairing mechanisms should be taken into account, rather than the single band model composed of only the $3dx^2$-y^2 orbital as usually believed.

Consequently series unconventional models with two band or multiband structure have been proposed, which are sharply distinct from the traditional single band d wave superconductivity. Here we briefly introduce some of them. Maier et al. found two domes of pairing strength using a two orbital (dx^2-y^2, $d3z^2$-r^2) model: one corresponding to the traditional normal amount hole doping regime and the other to the extremely overdoped regime. There is significant pairing strength in both the d wave and $s^{\pm}$ channels in the overdoped regime [22]. Jiang et al. construct a two orbital Hubbard model and found that hole doping the two orbitals Mott insulator leads to orbital dependent correlation and the robust spin and orbital exchange interaction produce a high T_c antiphase d wave superconductor even in the heavily doped regime. Their findings suggest that unconventional cuprates with liberated orbitals with doped two band Mott insulators can be a direction to realize high T_c superconductivity with enhanced transition temperature T_c [23]. Liu et al. suggested that the oxygen vacancies in Ba214 prefer to reside in the planar sites, while charge doping can reduce the energy differences between the low lying magnetic states. The AFM fluctuations in one dimensional CuO chains may play an important role in the superconducting pairing of electrons [24]. Wang et al. proposed an extended t J model on an effective brick wall lattice in Ba_2CuO_{4-y} while a time reversal symmetry broken pairing state can occur [22]. Jiang et al. provided a new model featuring in a Cu $3d$ multiplet structure where the ligand O $2p$ orbitals play an essential role [25]. Li et al. predicted that the O vacancies form a long range order, slicing the CuO plane into 1D chains and two leg ladders [26]. K. Yamazaki et al. proposed a Lieb lattice structure and construct a multiorbital Hubbard model that shows the competition between the s wave and d wave pairing and the coexistence among the intraorbital and interorbital pairings [27]. Ni et al. found that the hole overdoped Ba214 is in the orbital selective Mott phase (OSMP) at half filling, and the typical two orbital feature remains in Ba214. Their results indicate that in hole overdoped Ba214 the OSMP physics and orbital polarization, local magnetic moment, and spin or orbital fluctuations still exist [32].These results suggest an unconventional superconducting mechanism with a multiorbital character and significant s wave superconducting pairing strength. In particular crystal structure model with in plane ordering of oxygen vacancies has been proposed. The crystal structure including the locations of oxygen vacancies in Ba214 plays a crucial role in revealing the source of superconductivity and its mechanisms that needs to be further verified by experiments.

Professor Ghiringhelli et al. probed the electronic structure of Ba214 by combining Cu L_3 x-ray absorption (XAS) and resonant inelastic x ray scattering (RIXS) techniques [37]. The RIXS spectra hint at a significant departure of Ba214 from the common quasi 2D electronic structure of superconducting cuprates with pure dx^2-y^2 symmetry. They proposed that the significant contributions from dz^2 orbital are present in Ba214 superconductor.

It is worth noting that the multiorbital features in Ba214 are also present in the recent discovered nickelate double layered superconductor $La_3Ni_2O_7$ [55–58]. In $La_3Ni_2O_7$, the electronic structures are contributed by both Ni dx^2-y^2 and dz^2 orbitals. The dz^2 orbital between two layers was suggested to be crucial for its superconductivity.

The high T_c in Ba214 comparing with the scenario where only x^2-y^2 orbital takes the role in La214, strongly suggests that dz^2 orbital can play significant roles in further enhancing superconducting transition temperature in cuprates [27].

4. Remarks

Ba214 superconductor with a compressed version of copper oxygen coordination is discovered showing surprisingly 80 % higher Tc than that for isostructural La214 with universal elongated copper oxygen coordination for rest of all high Tc cuprates. The orbital order reversal between $3dx^2$-y^2 over $3dz^2$ in Ba214 caused by compressed coordination is not favorable for superconductivity in main stream theories, calling for new insights as what are the necessary ingredients to further enhance superconductivity of cuprates. Impacts of Ba214 superconductor simply include (I) the significant involvement of $3dz^2$ contribution may be the promising strategies to enhance T_c; (II) the $3dz^2$ orbital will lead to the multiband electronic structure in sharp contrast to the universal single band mode for rest of high Tc cuprate superconductors; (III) the two orbital scenario will in turn generate the paring configuration able to host s wave superconductivity that inspires us to discover more "new buds" on the old tree of cuprate superconductors by walking along the path initiated by Prof.Müller.

Author statements

The paper is invited to the special issue in memorization of Prof. Müller. All authors read the uploaded version. All authors agree with the revised version.

Author contributions

CQJ conceive the research. WML & JFZ performed the research. All authors contributed equally to paper.

Declaration of Competing Interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

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Data availability

The data that has been used is confidential.

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