Nonadiabatic coupling of the dynamical structure to the superconductivity in YSr$_2$Cu$_{2.75}$Mo$_{0.25}$O$_{7.54}$ and Sr$_2$CuO$_{3.3}$

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A crucial issue in cuprates is the extent and mechanism of the coupling of the lattice to the electrons and the superconductivity. Here we report Cu K edge extended X-ray absorption fine structure measurements elucidating the internal quantum tunneling polaron (iqtp) component of the dynamical structure in two heavily overdoped superconducting cuprate compounds, tetragonal YSr$_2$Cu$_{2.75}$Mo$_{0.25}$O$_{7.54}$ with superconducting critical temperature, $T_c$, = 84 K and hole density $p$ = 0.3 to 0.5 per planar Cu, and the tetragonal phase of Sr$_2$CuO$_{3.3}$ with $T_c$ = 95 K and $p$ = 0.6. In YSr$_2$Cu$_{2.75}$Mo$_{0.25}$O$_{7.54}$ changes in the Cu-apical O two-site distribution reflect a sequential renormalization of the double-well potential of this site beginning at $T_c$, with the energy difference between the two minima increasing by ~6 meV between $T_c$ and 52 K. Sr$_2$CuO$_{3.3}$ undergoes a radically larger transformation at $T_c$ > 1 Å displacements of the apical O atoms. The principal feature of the dynamical structure underlying these transformations is the strongly anharmonic oscillation of the apical O atoms in a double-well potential that results in the observation of two distinct O sites whose Cu-O distances indicate different binding modes and valence-charge distributions. The coupling of the superconductivity to the iqtp that originates in this nonadiabatic coupling between the electrons and lattice demonstrates an important role for the dynamical structure whereby pairing occurs even in a system where displacements of the atoms that part of the transition are sufficiently large to alter the Fermi surface. The synchronization and dynamic coherence of the iqtps resulting from the strong interactions within a crystal would be expected to influence this process.

X-ray absorption fine structure | cuprates | superconductivity | dynamical structure | tunneling polarons

More than 30 y after the discovery of unconventional superconductivity in cuprates (1) and subsequently in analogous materials its underlying mechanism and in particular the role of the lattice are still under debate. Proposed microscopic theories range from purely electronic Mott–Hubbard and t-J approaches at one extreme to Bose–Einstein condensates of bipolarons at the other (2–4). Experimentally, however, anomalous isotope effects (5), resonant ultrasound (6), angle-resolved photoemission spectroscopy (7–9), femtosecond optical pump terahertz (10)/megaelectron-volt transmission electron microscopy probe (11), infrared pump (12), and so on have demonstrated that specific phonons not only couple to the superconductivity but correlate directly with the gap energy and may even transiently induce it well above the superconducting critical temperature, $T_c$. Cuprates also exhibit a plethora of superstructures indicative of strong electron–lattice coupling, stripes that have been proposed to stabilize the superconductivity (13), and charge-density waves (14, 15) and the pseudogap (PG) (16) that compete with it. Another possibility is mechanisms that boost $T_c$ from a low value expected within a conventional Bardeen-Cooper-Schrieffer (BCS) scheme. That this question remains unanswered suggests considering more unconventional approaches (4). One candidate is the dynamical structures of cuprates, $S(Q,E)$ or experimentally $S(Q,\tau = 0)$, specifically their internal quantum tunneling polarons (iqtp). An iqtp is a set of atoms oscillating between two structures that possess different geometries, energy levels, and charge distributions (17–19). A chemist would describe these endpoints as separate species, adapting this term that applies more intuitively to solutions to the atoms in crystalline solids. Neutron scattering and X-ray absorption fine structure (XAFS) experiments (20) on Sr$_2$CuO$_{3.3}$ (with superconducting critical temperature, $T_c$, = 84 K) and Sr$_2$CuO$_{3.3}$ ($T_c$ = 95 K) through their superconducting transitions demonstrates that the common factor in superconductivity in cuprates, including those prepared by high-pressure oxygenation, is an internal quantum tunneling polaron in its dynamical structure. In addition, Sr$_2$CuO$_{3.3}$ is the first material to show a comcomitant transformation in this structure involving atom displacements > 1 Å that would be expected to modify its Fermi surface, which would complicate the transition beyond a purely electronic one consisting of the pairing of electrons of opposite momentum across fixed electronic states.

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results from “overdoped” YSr2Cu2.75Mo0.25O7.54 (YSO-Mo) that is isostructural with YBa2Cu3O7 (Fig. 1A and SI Appendix, Fig. S1), Tc = 84 K (26) and hole doping p (excess charge on the planar Cu2 site) = 0.3 to 0.5 (27) and Sr2CuO3.3 (SCO) that is structurally analogous to La2CuO4 (Fig. 1B and SI Appendix, Fig. S1), Tc = 95 K (28, 29), and p = 0.6, both synthesized via high-pressure oxygenation (HPO) (30, 31). In YSCO-Mo the Cu2-apical O (Oap) double-well potential is degenerate in the normal state but renormalizes below Tc with the energy difference between its two minima increasing with decreasing temperature by \( \sim 6 \) meV. SCO is already unique among cuprates in not having intact CuO2 planes (32, 33). Its Cu EXAFS demonstrate that it is unique among superconductors in that its Oap shift by \( >1 \) Å at its superconducting transition, challenging our conception of superconductivity as an electronic transition that is incompatible with structural transformations.

We have recently shown that HPO cuprates are described by their own phase diagram (34). The principal feature of the well-known one for non-HPO cuprates is the superconducting “dome”
that begins at $p \sim 0.06$, peaks at $p \sim 0.16$, and ends at $p \sim 0.27$. Subsequent augmentations with the microstrain in the planes (35) and hole density on the O atoms (36) explain some of the material specificity but do not modify this overall pattern. For HPO compounds the superconductivity may begin at $p < 0.06$ and continues to increase beyond $p = 0.27$ with possible flattening but no reduction in $T_c$. Although we have found that the excess O in YSCO-Mo is mostly taken up by domains enriched in octahedral Mo(VI) substituting in the Cu(1) chains, much of the extra charge resides in the CuO$_2$ planes (27) and some of the carriers constitute a normal Fermi liquid that coexists with the superconductivity (26). The inherent inhomogeneity (37) in YSCO-Mo and SCO was probed by EXAFS, which is arguably the most incisive experimental method for characterizing short-range order and is especially sensitive to its changes. Diffraction patterns originate in the long-range average structure of a material and provide precise information on the symmetry and symmetry-constrained locations of the atoms that dominate the Bragg peaks. In contrast, EXAFS—and pair distribution function (pdf) analysis—are sensitive to local order separate from the crystallographic symmetry. The element selectivity of EXAFS provides further advantages by separating the atom pairs comprising the distribution function. Especially important for this study, EXAFS measures the instantaneous structure factor, $S(Q, t = 0)$, that incorporates the dynamic structure components, $S(Q, E)$, observed with inelastic scattering. EXAFS therefore accesses time and energy scales corresponding to collective dynamical phenomena (25, 38, 39). Dynamical structures such as the iqtp are demonstrated when $S(Q, E/t = 0)$ gives locations for atoms that differ from those (19, 38) obtained from diffraction and elastic scattering measurements (20–22, 40). This complementarity was the basis for the original...
identification of the Cu-Oap two-site distribution (41) and its assignment to the double-well potential of the iqtp (17, 19, 42).

Results

For YSCO-Mo one-fourth of the Cu1 atoms in the CuO3 chain are replaced by Mo (26). The EXAFS of the sample of the e[II][E] orientation of tetragonal YSCO-Mo is composed of the Cu-Oap-Cu2 pairs and the c-axis projections of the Y/Sr pairs. The e[II][a(a)] sample contains the contributions of the CuO2 planes–CuO2 chains (34) and Y/Sr projections in the aα plane (Fig. L4 and SI Appendix, Fig. S1). Crystallographically tetragonal SCO (Fig. 1B and SI Appendix, Fig. S1) is unique in that its a-axis that contains half-filled -Cu-O- chains aligns with the field, its b axis in the bc plane perpendicular to the field contains intact -Cu-O- chains, and its Oap sites are only partly occupied (33). Its EXAFS corroborates that its Cu-Sr sublattice is locally tetragonal as found by diffraction.

The EXAFS spectra, χ(R), are presented as their Fourier transforms, χ(R), where the modulus peaks correspond to the contributions of the neighbor atom shells in an approximate representation of the partial pair distribution. This interpretation is modified by the phase shift that causes the locations of the modulus peaks to be below the actual distances and the finite data range that causes the peaks to be much wider than the actual resolving capability of the analysis. The overlap of the modulus peaks from adjacent neighbor shells therefore does not relate to the actual π/kmax = 0.11 Å resolution in this analysis. Since the EXAFS signal includes both an amplitude and phase, the modulus represents only half of the information with the remainder contained in the real/imaginary component of χ(R). The E|aa (Fig. 1C) and E|c (Fig. 1D) spectra of YSCO-Mo are dominated by the contributions from the nearest-neighbor O, Sr, and Cu (34, 43–46). The increased thermal Debye–Waller factors at higher temperatures cause the small reductions in the amplitudes of the spectral features. Although the EXAFS spectra of SCO show the expected features from the nearest-neighbor O, Sr, and Cu, it differs from that of the La2CuO4 family (37, 47) since the spectra originate in the structures along the a axis and in the bc plane. In contrast to the expected behavior demonstrated by the YSCO-Mo spectra, the E|a spectra of SCO are complicated, showing a large reduction with increases governed more by the Cu-O feature and an anomalous diminution of the Sr modulus amplitude with decreasing temperature (Fig. 1E).

The focus of this paper, however, is the behavior of the E|bc spectra of SCO in the region R = 1.5 to 2.8 Å that contains the contributions of Cu-O pairs at 2.2 to 3 Å. In contrast to the relatively subtle effects on the EXAFS resulting from the partial collapse of the iqtp double-well potential at Tc (20, 23, 42), these spectra have minimal amplitude in this region down to 97 K but at the onset of superconductivity at 95 K a complex pattern of large-amplitude features abruptly appears (Fig. 1F). This indicates the formation throughout the crystal of a structural moiety in the bc plane consisting of O ions displaced over long distances from their positions in the normal state into new ones in the superconducting state. That the crystal structure shows O atoms in symmetry-constrained positions that do not give these distances and no change in the structure across Tc (28, 29) indicates its dynamical origin. Smaller, continual changes divide the remaining spectra into temperature regions (Fig. 1 G, I, and J), demonstrating that the Cu environments undergo additional stepwise transformations.

Curve fitting (SI Appendix, Figs. S2–S70) confirms that the EXAFS gives the crystallographic positions for the cations and the YSCO-Mo plane and chain O atoms (33, 34). It also gives detailed depictions of the anomalous temperature dependence of Oap in YSCO-Mo and the entire O sublattice in SCO. The behavior of the Cu2-Oap in YSCO-Mo is visualized in the EXAFS of this pair isolated by subtracting the contributions of the other neighbors determined by the curve fits. The two-site distribution is evident in the two peaks in the residual χ(R) (Fig. 2A). The R = 1.75 Å peak increases continuously with increasing temperature, with a significant jump at Tc. The trend in position and amplitude of the R = 1.95 Å peak is more complicated, being relatively high at the lowest temperatures, then diminishing until near Tc it melds with the R = 1.75 Å one at a higher amplitude. This behavior reflects the sensitivity of χ(R) to the details of the two-site distribution, which are unraveled by the curve fits to reveal a consistent pattern. The two Cu–O distances and occupancies for the Cu2-Oap pair (Fig. 2 B and C and SI Appendix, Figs. S71–S86) divide into four regions: 1) the normal state for T > 87 K, 2) a fluctuation region from 77 to 85 K in the distances and 77 to 94 K in the numbers of the two O, 3) a first superconducting region from 68 to 77 K, and 4) a second superconducting region for T < 68 K. Although the distances and numbers are all obtained independently, the stepwise shifts in distances and numbers occur at the same temperatures, delineating the boundaries of temperature regions where the two-site distribution and its double-well potential change.

Another quantity showing coupling to the superconductivity is the anharmonicity of the c-oriented Cu2-Sr pair. This is evaluated from the EXAFS from the logarithm of the amplitude divided by the amplitude of the selected baseline spectrum vs. k2 (Fig. 2D). Since the Cu-Sr sublattice is conserved the linear part of this function is the difference in the Gaussian Debye–Waller factor and the curvature and deviation of the intercept from zero reflects the change in the anharmonic component of the pair distribution (48). The conservation of the Cu-Sr distribution in the normal state is demonstrated by these ratios being essentially horizontal lines with extrapolated intercepts of 1 from 85 to 110 K. At Tc, these ratios begin to decrease continuously, at a faster rate and with greater curvature as the temperature becomes lower. This pattern implies that the polarizability of the Sr scales with the superfluid density.

The most remarkable component in the structure of SCO is the entity that forms in the bc plane at Tc to give the spectral features at R = 1.8 to 2.6 Å in the superconducting phase. Curve fits confirm the absence of significant numbers of O neighbors beyond 2.4 Å in the normal state. Subtracting the contributions of the two O neighbors with Cu–O distances <2 Å and the Sr and Cu distances R <3 Å from the relevant portion of the spectrum for the Cu-Oap feature and an anomalous diminution of the Sr modulus amplitude with decreasing temperature (Fig. 1E).

Discussion

Descriptions of the structural transformations in YSCO-Mo and SCO consistent with these results begin with the previously described environments of the relevant O atoms (33, 34) (Fig. 3).
The two-site apical O distribution in YSCO-Mo is simple (Fig. 3A), here showing the positions synchronized along the chain. SC is much more complex. Interpreting the high degree of disorder and the large change at $T_c$, a random distribution of the apical and chain vacancies, there are three types of Cu, all with two b chain but distinguished by having zero, one, or two a chain O neighbors. The square planar geometry of CuII dictates that the $2a+2b$ transformation would have its Oap either vacant or with one or two Oap at $>2.1$ Å and the $2a+2b$ conformation two Cu-Oap distances at $<2$ Å. The $2a+2b$ Cu with its long or short Cu-Oap and distorted environment would then be the Cu(III) required for charge balance. This combination of occupied and vacant Oap sites explains the EXAFS spectra of the normal phase, with the low Cu-Oap amplitude caused by the range of Cu-Oap distances (Fig. 3B). At $T_c$ an Oap adjacent to an Oap-vacancy shifts $\sim$2 Å in the b direction to form a Cu$_2$O$_2$ unit with an asymmetric bridge that gives the Cu–O distances of 2.6 and 2.9 Å (Fig. 3C and D). The other O of the Cu$_2$O$_2$ shifts along c away from this Oap, buckling the original -Cu-O-Cu- chain. This O would also move away from the Cu with the shorter, 2.6 Å Cu–O distance, splitting the original distance into two distances at 1.85 and 2.05 Å. The expansion of the 2.05 Å Cu–O distances at $T_c$ would therefore be a response to the formation of these asymmetrically bridged Cu$_2$O$_2$ units. The expansion of the 1.85 Å distance assigned to a long Cu-Oap (Fig. 3G) would result from the modified charge distribution and the increased homogeneity of the Cu sites, whereby ones with and without Oap become more uniform. The 1.9-Å Cu–O distances are unaffected because they are not involved in this transformation.

Analogous to YBa$_2$Cu$_3$O$_7$, the Cu$_2$O$_2$ two-site distribution in YSCO-Mo is assigned to the iqtp of the dynamical structure. This is corroborated by the single site distribution found for CuI-Oap that shows that the correlated motion of the Cu$_2$O$_2$ pair differs from the CuI-Oap pair. Assignment of the asymmetrically bridged Cu$_2$O$_2$ unit in superconducting SCO to an iqtp is supported by the difference with the crystallographic structure determined by neutrons (28, 29) that also show no change across $T_c$. The dynamical component of the iqtp would be the oscillation of the O atoms between their two equivalent positions across the Cu$_2$O$_2$ midpoint.

Nonadiabatic superconductivity are the first significant finding of this paper. Recent reports by the authors have shown that the common attribute in cuprate superconductivity is not the superconducting “dome” of the phase diagram (34) or Cu$_2$O$_2$ planes (33) or the prolate geometry of the Cu in these planes (50). A remaining factor is the dynamical structure of specific CuO pairs, the iqtps. The identification of iqtps as a ubiquitous property of the O and the Zhang–Rice singlet state that occurs on Oap despite the 3d/2 level’s being lower in energy than the 3d/2,2. However, the relationship of the iqtp to the superfluid quasiparticles is unclear insofar as, pending more complete data, the forms and populations of the double-well potentials do not appear to track the phase diagram with the exception of the softening at $T_c$.

The second remarkable result is the $\sim$2-Å displacement in the dynamic structure of Oap coincident with the superconducting transition. Changes in the structure across $T_c$ in response to charge screening by the superfluid are only a few parts per thousand in the lattice constants of conventional BCS-type superconductors. Percent shifts occur in the Cu$_2$O$_2$ distributions in cuprates. The O displacement in SCO would be the first observation of a rearrangement of the atoms sufficiently large to effect a change in the Fermi surface at the transition. This is a substantial departure from the standard depiction of BCS as a pairing of electrons of opposite momentum across a conserved Fermi surface in which the mediation of this pairing by a soft lattice mode prevents that mode from providing a path for a structural transformation. The caveat in this result is that this transformation occurs in the dynamic and not the static structure. Recent inelastic neutron scattering results in other systems have confirmed that such structural features demonstrate dynamic pair correlations not present in the static structures that originate in processes such as tunneling (39). An outstanding question is the effects of the HPO synthesis that results in compounds displaying a very much different superconducting region in their phase diagram than the “dome” of conventional cuprates prepared with O$_2$ (34, 60). The 84 K $T_c$ of YSCO-Mo, which EXAFS shows conforms very closely to the typical structure of conventional cuprates without substantial disorder, is 25% lower than that of its parent YSr$_2$Cu$_3$O$_6$+$\delta$ despite a measured $p$ of 0.5 (27). It nevertheless possesses the same two-site Cu-Oap distribution found in other cuprates, differing in that the relative occupancies change instead of converging at $T_c$. The 95 K $T_c$ of this phase of Sr$_2$CuO$_3$ is over 2.5 times higher than its La$_{1.5}$Sr$_{1.5}$CuO$_4$ structural analog (32). Its unique Cu charge, structure, magnetism, and very high degree of disorder in its O sublattice (33) could be expected to convey additional unique behaviors, for example the structural transformation we have identified and described here.

These results provide evidence for a direct role in cuprates for nonadiabatic lattice dynamics that manifests itself as anharmonic oscillations of the atoms and charges (61) coupled to the superconductivity. Analogous to, for example, monolayer FeSe on SrTiO$_3$ (62) and Li$_2$CuO$_2$ (63) our analysis demonstrates that collective behaviors of iqtps most likely have an equally significant role in pairing and condensation. For the iqtp, this collective action is easily assigned to the dynamic synchronization of the O oscillations over distances of the order of the superconducting coherence length. The interaction of the oscillators occurs not only through their dipoles but also directly through their neighbors, for example the Sr that bridges the Oap atoms in YSCO-Mo. Oscillating charge and a significant role for the inert cations have not been previously reported or proposed. In addition, the dynamical structure report here is of substantial interest as a mechanism for the collective integration of lattice dynamics and charge that derives from a charge-coupled structural instability.
Fig. 3. Conceptual structures of the dynamical structure transformations. The results from SCO are derived from the EXAFS results. The shaded areas highlight the atoms of the iqlps whose structure will change across $T_c$. (A) Cu2-Oap two-site distribution in YSCO-Mo, organized in pairs with corresponding displacements of the Sr. (B) The $bc$ plane in the normal state of SCO. There are some Oap vacancies, and the $b$-oriented Cu-O chains are fully occupied. As described in the text, the half-vacant O sites in the $a$-oriented Cu-O chains give three types of Cu: 1) zero O along $a$, two O along $b$, two Oap with Cu-Oap $\approx 1.9$ Å; 2) two O along $a$, two O along $b$, 0 to 2 Oap with Cu-Oap $> 2.1$ Å; and 3) Cu(III) with one O along $a$, two O along $b$, and variable Oap. (C) The Cu$_2$O$_2$ unit in SCO whose structure changes across $T_c$. (Left) In the $bc$ plane in the normal state (B) showing the two Cu sites, one of which has an O vacancy in the $a$ direction, and one with an Oap vacancy. In the superconducting state (Center, D) the Oap has shifted toward the Oap vacancy to give the 2.7- and 2.9-Å Cu–O distances and the O in the Cu has moved slightly downward. (Right) The same set of atoms viewed in the $ac$ plane as in E. (D) The $bc$ plane in the superconducting state of SCO, showing the doubly bridged Cu$_2$O$_2$ units. (E) The $ac$ plane in the superconducting state of SCO, showing the three types of Cu.
that does not culminate in static transformation. It is thereby consistent with the original rationale for superconductivity in cuprates, stabilizing the Cooper pairs by increasing the electron–lattice coupling beyond the adiabatic approximation while simultaneously frustrating incipient structural phase transitions (1, 5), but also constitutes a significant extension to high temperature superconductivity.

Materials and Methods

The samples, data acquisition, and analysis are the same as in our recent reports (33, 34) with the addition of having been measured at multiple temperatures and are detailed in SI Appendix. The two compounds were synthesized and samples prepared by magnetic orientation and characterization by standard methods as described in those reports and others that they cite. XAFS measurements were performed at the Stanford Synchrotron Light Source in the continuous scanning transmission mode on end station 2–2. X-ray absorption, normalization, and approximation of the smooth atomic source in the continuous scanning transmission mode on end station 2–2. The spline parameters were also held to narrow ranges within each of the four sets of spectra to ensure the lower than those of the structural features. The spline parameters were also held to narrow ranges within each of the four sets of spectra to ensure the lower than those of the structural features.

Quantitative analysis of the XAFS spectra was performed with the FEFF9 code (64). Debye–Waller factors and $\theta_0$ values were constrained to narrow ranges determined by inspection of the results from unconstrained fits on the spectra from all temperatures. Debye–Waller factors for the separable Cu-neighbors pairs were also evaluated by the logarithm–amplitude ratio method as shown in Fig. 2D, often showing analogous correlations with the superconducting transition. These will be reported later.

Data Availability. In the absence of an appropriate institutional open access data repository, the original data can be obtained by contacting the first author at s33v3n.chnardsdn@icloud.com.

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