Imaging the effect of electron lattice interactions on high-$T_c$ superconductivity in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$

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Abstract. One of key challenges in high-$T_c$ superconductivity research is to identify a predominant mechanism controlling superconductivity at atomic scale. Using the spectroscopic imaging scanning tunnelling microscopy technique we find a $\sim 9 \pm 1\%$ sinusoidal variation in local energy gap $\Delta$ triggered by the crystal “supermodulation” revealing a strong non-random out-of-plane effect on superconductivity[2], substitutional isotope effect on the $d^2I/dV^2$ spectrum reveals 6\% reduction of mode energy $\Omega$ indicating the involvement of the lattice vibrations that couple strongly to the quasiparticle states of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ [3]. We also find that the heterogeneous electronic states coupling to the lattice only occur above a certain energy which separates the spatially homogeneous and heterogeneous excitations[4].

1. Introduction

The superconductive copper oxide materials exist in a variety of complex crystal forms. Introduction of about 16\% holes into the insulating CuO$_2$ crystal plane by chemical doping generates the highest critical temperature $T_c$ superconductors known. However, the maximum $T_c$ varies widely between crystals sharing the same basic in-plane electronic structure. These variations cannot be due to the doping dependence of CuO$_2$ in-plane electronic structure of cuprates. Identification of the predominant out-of-plane influence may be pivotal to finding a route to higher $T_c$ cuprate superconductors.

Changes of inter-atomic distances within the unit cell should, in theory, have major effects on both the underlying electronic structure and the superconducting state[5,6]. To investigate how the geometry of a CuO$_2$ pyramid affects the superconductivity locally, and ideal experiment would be to continuously perturb the dimensions of a single unit cell (Fig. 1b) and measure the resulting changes in superconducting properties within that same unit cell. In fact, significant variations in cell dimensions and geometry occur naturally at the nanoscale in the $\text{Bi}_2\text{Sr}_2\text{Ca}_{n-1}\text{Cu}_n\text{O}_{4+2n}$ ($n=1,2,3$) family.

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These variations take the form of a bulk incommensurate periodic modulation[7-12] perturbing the atoms from their mean positions[13] as shown schematically in Fig 1b. This crystal ‘supermodulation’ is believed to originate from a misfit between the preferred bond lengths of the perovskite and rock-salt layers of the crystal. In each of the 14 layers of the Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi-2212) unit cell, atoms are displaced from their mean locations (i.e. where they would be in an idealized average unit cell) by up to 0.4Å, following a functional form repeating on average every 26Å (~4.8 unit cells) along the $a$-axis.

These distortions are represented schematically in Fig. 1b. Here we exploit the associated modulations of inter-atomic distances (whose largest fractional change is to the Cu-O apical distance) to explore directly associated changes to the superconducting state.

As well as the impact of varying the inter-atomic distances within individual crystal unit-cells, we also discuss atomic scale electron-lattice interactions and their influence on the electronic states. So far, no consensus exists on the electron pairing mechanism of high-$T_c$ superconductivity. One reason is that the extensive studies of bosonic modes and related electronic self-energy changes have not resulted in the unambiguous identification of a boson mediating the pairing.

Here we use spectroscopic imaging scanning tunnelling microscopy (SI-STM) technique to study these atomic-scale lattice effects on high temperature superconductor Bi-2212.

![Figure 1](image)

**Figure 1** a) Topographic image of Bi-2212. b) crystal structure of Bi-2212 with schematic image of crystal ‘supermodulation’.

2. Experimental

We use floating-zone grown single crystals cleaved between the BiO planes in cryogenic ultra-high vacuum and immediately inserted into the STM head at 4.2K. The CuO$_2$ plane is ~5Å beneath the BiO surface and separated from the STM tip by insulating BiO and SrO layers. Figure 1a shows a 482.5Å square topographic image of the BiO surface and a 10nm square image in the inset, revealing the distorted grid-like arrangement of Bi atoms; the Cu atoms lie ~5Å below. The effects of the crystal supermodulation at the BiO layer can be seen in Figure 1a as a surface corrugation.

To explore the CuO$_2$ electronic local-density-of-states LDOS($r,E$) at each point, we use STM-based imaging of the differential conductance $g(r,V)=dI/dV(r,V)$. If spatial variations of the tunneling matrix elements do not predominate, this results in a spatial image of LDOS($r,E=eV$) $\propto g(r,V)$.

3. Impact on superconductivity of varying the interatomic distances in unit cell dimension

Many theoretical models of high temperature superconductivity focus only on the doping dependence of the CuO$_2$ plane electronic structure. But such models are insufficient to explain the strong variations in superconducting critical temperature $T_c$ among cuprates which have identical hole-density but are
crystallographically different. A key challenge has been to identify a predominant out-of-plane influence controlling the superconductivity— with much attention focusing on the distance $d_A$ between the apical oxygen and the planar copper atom. Here we report direct determination of how variations of inter-atomic distances within individual crystalline unit cells, affect the superconducting energy-gap maximum $\Delta$ of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8+\delta$. In this material quasi-periodic variations of unit cell geometry occur in the form of a bulk crystalline ‘supermodulation’. Within each supermodulation period, we find a $\sim 9\pm 1\%$ sinusoidal variation in local $\Delta$ that is anti-correlated with the associated $d_A$ variations. Furthermore, we show that phenomenological consistency would exist between these effects and the random $\Delta$ variations found near dopant atoms if the primary effect of the interstitial dopant atom is to displace the apical oxygen so as to diminish $d_A$ or tilt the CuO$_5$ pyramid.

To make these measurements, we developed a technique which accurately tracks the local phase of the crystal supermodulation, extracting the value of the supermodulation phase at every location—a supermodulation phase map $\phi(r)$. With it, we correctly parameterize the dimensions of the unit cell at every $r$ (See ref. 2). The latter can be achieved because the actual bond length changes in each unit cell are determined from $\phi(r)$ using knowledge from X-ray crystallography. Such studies have established[7-11] that the Cu-O$_\text{apical}$ bond length $d_A$ varies with $\phi$ (as defined here) by as much as 12%, peak-to-peak. If we consider only the first harmonic in atomic displacements within the most widely accepted refinements of the crystal supermodulation, this occurs because the amplitude of the $c$-axis supermodulation is greater in the CuO$_2$ layer than in the adjacent SrO layer containing the apical oxygen. Within this simplified picture the apical oxygen distance $d_A$ is thus minimal at $\phi=0^\circ$, and maximal around $\phi=180^\circ$. A small number of studies present a different crystal refinement[9]. Nevertheless changes in $d_A$ represent the largest fractional change of any bond-length which the unit cell and occur with a primary periodicity of the supermodulation plus small additional departures occurring in the second harmonic[8].
Using this crystal modulation phase-map technique, we next determine $\phi(r)$ and $\Delta(r)$ simultaneously in each field of view. Each pixel is then labeled by its local value of $\phi$ and $\Delta$. Next we generate two-dimensional histograms showing the frequency with which each pair of $\phi, \Delta$ values occurs (Fig.2a). Fig.2b represent the mean gap energy of each of these distributions, plotted again versus $\phi$. This analysis reveals that the superconducting energy gap varies significantly, moving sinusoidally with the unit cell dimension as labeled by $\phi$. The data in Figure 2 demonstrate a direct atomic-scale influence of the unit cell geometry on the local superconducting state of a cuprate. In all samples studied over a wide range of doping, the $\Delta$ vary in the same fashion with supermodulation phase, with gap maxima in the vicinity of $\phi=0^\circ$ and minima near $\phi=180^\circ$. The measured functions $\Delta(\phi)$ were well fit by the single harmonic function $\Delta(\phi)=\Delta(1+A\cos(\phi+\alpha))$, as shown in the Fig2b. Using such fits, the mean peak-peak range $2A$ was found to be $9\pm2\%$, with no apparent dependence on doped hole-density (Fig.2d).

However, a potential cause of the periodic gap modulations reported here could merely be that the dopant density is modulated with same period as the supermodulation. To examine this point, high energy $\sim1\text{V}$ d$I$/d$V$-maps (from which the locations $O(r)$ of the dopant-atom-induced impurity states are identified[14].) were measured in the same field of view as the gap maps, while the topography of the surface was simultaneously recorded. The relation between $O(r)$ and the supermodulation phase $\phi(r)$ was then analyzed by determining the probability of finding a dopant-induced impurity state at each value of $\phi$. In Fig. 2c we show that the dopant density is somewhat correlated with $\phi$ but with peaks at both $\phi=0^\circ$ and $\phi=180^\circ$. Since $\Delta(\phi)$ has only a single peak around $\phi=0^\circ$, Fig.2c is inconsistent with periodic dopant-density variations being the primary cause of the gap modulation at $q_{SM}$.

4. Visualizing the effect of electron-lattice interactions

Electron-pair formation is essential to superconductivity. For conventional superconductors, tunnelling spectroscopy established that such pairing is mediated by bosonic modes (phonon); peaks in the second derivative of tunnel current $d^2I/dV^2(E=eV)$ corresponded to each phonon[15,16,17]. For high-$T_c$ superconductivity, however, no boson mediating electron pairing has been identified. One explanation could be that pairing, related electron-boson interactions (EBI), are heterogeneous at the atomic-scale[18]; complete EBI characterization would then have been impossible without atomic resolution probes. But recent breakthroughs in STM-based $d^2I/dV^2(E)$ spectroscopy now allow direct access to atomic scale bosonic modes[19]. Here the EBI effects are hypothesized to produce signatures at $E_{\text{gap}}+\Omega$ analogous to the strong coupled superconductivity theory which is initially predicted by Eliashberg[20].

The $dI/dV(r,E=eV)$ and $d^2I/dV^2(r,E=eV)$ are simultaneously imaged with atomic resolution and register. From the former, the gapmap $\Delta(r)$ is derived(Fig.3b). From the latter, the energies $\Pi(r)$ at which $d^2/dV^2(r,E)$-peaks occur are measured and the boson interaction-energy maps $\Omega(r)=\Pi(r)-\Delta(r)$.

![Figure 3](image-url)

**Figure 3.** a) typical $dI/dV$ spectra sorted by the gap and their corresponding $d^2I/dV^2$ spectra. b) gapmap $\Delta(r)$. c) omega map $\Omega(r)$.
are then calculated. A typical $\Omega(r)$ is shown in Fig.3c and reveals immediately that boson energies $\Omega(r)$ are heterogeneous at the ~2nm scale with $40\text{meV}<\Omega(r)<65\text{meV}$.

Next we study these $d^2I/dV^2$-signature of EBI at a sequence of different hole densities per CuO$_2$ $p$: $p \sim 0.12 \rightarrow p \sim 0.24$. In Fig. 4a we show that the average superconducting energy gap $\Delta$ decreases from $\sim 60\text{meV} \rightarrow 20\text{meV}$ with increasing doping as expected. In strong contrast, Fig 4b shows that, although changes occur in spatial correlations of $\Omega(r)$, no change is detectable in the average boson energy $\overline{\Omega}$. In fact, histograms of $\Delta$ and $\Omega$ measured on 5 samples at different dopings (Fig. 4b) reveal that, while the distributions of $\Delta$ evolve rapidly with doping, those of $\Omega$ appear unchanged: $\overline{\Omega} = 52\pm 1\text{meV}$ for all dopings.

Thus, $\overline{\Omega}$ are quite independent of doped hole-density. Which boson could exhibit such highly doping-independent EBI characteristics? The ‘resonant’ spin-1 magnetic excitation mode[21] appears inconsistent because its energy is 43 meV in Bi-2212 but, more importantly, is believed to be strongly doping dependent. The incommensurate, dispersive, spin density wave modes[22,23] also appear inconsistent because of their characteristic strong energy- or doping-dependences. By contrast, because energies of lattice-vibration modes change little with doping[24] they are logical candidates for the boson detected by $d^2I/dV^2$-imaging.

To test the hypothesis that the boson we detect by $d^2I/dV^2$-imaging are lattice vibration modes, we prepared crystals in which the normal $^{16}\text{O}$ was completely substituted by $^{18}\text{O}$ (as verified by frequency shifts detected in Raman spectroscopy). Figure 4c provides the quantitative comparisons between the distributions of $\Omega(r)$ and $\Delta(r)$ in samples containing the two different oxygen isotopes. For each sample, we take $\Delta(r)$ and $\Omega(r)$ and construct a two-dimensional histogram of the frequency of occurrence of spectra with a given pair of values ($\Delta$, $\Omega$). Each histogram is peaked along the vertical axis at the most common gap energy $\overline{\Delta}$ and along the horizontal axis at the most common boson energy $\overline{\Omega}$. Comparison between $^{16}\text{O}$ $\Delta\Omega$-histogram(blue) and the $^{18}\text{O}$ $\Delta\Omega$ -histogram (red) reveals immediately that, for any particular value of $\Delta$, the value of $\overline{\Omega}$ for $^{18}\text{O}$ shifts downwards by approximately 3.5$\pm 1$ meV compared to that of $^{16}\text{O}$. Finally, Fig. 4d shows the distribution of boson energy $\Omega(r)$ in two different samples containing complete substitutions of the two oxygen isotopes: $^{16}\text{O}$ in blue and $^{18}\text{O}$ in red. We find that the shift of $\overline{\Omega}$ upon substitution of $^{16}\text{O}$ by $^{18}\text{O}$ is $-3.7\pm 0.8\text{meV}$.

**Figure 4.** a) $\Delta(r)$- and b) $\Omega(r)$-histogram. c) $\Delta\Omega$-2D histogram for $^{16}\text{O}$ and $^{18}\text{O}$. d) $\Omega$ histograms for $^{16}\text{O}$ and $^{18}\text{O}$.
These results are found equally true for both filled \( E=-(\Delta+\Omega) \) and empty \( E=+(\Delta+\Omega) \) states, as expected for EBI in strong coupled superconductivity theory[25]. Consequently, substituted of \(^{18}\text{O} \) for \(^{16}\text{O} \) reduces the mean boson energy scale \( \Omega \) of EBI by \( 6\%(-1-\sqrt{(16/18)}) \) – precisely as expected for lattice vibrational modes involving the O atom.

5. Electronic homogeneity and heterogeneity

Finally, we focus on the feature in \( g(r,V) \) appearing as the “kink” which have been reported ubiquitously[14,26,27,28,29] in cuprate STM data. In general, these kinks are weak perturbations to local density of states near optimal doping, becoming more clear within nanoscale regions increasing in number as \( p \) is strongly diminished[26,27]. In Figure 5a we show representative \( \Delta \)-sorted spectra. Notice that it is for \( \Delta<50\text{meV} \) the kinks become more obvious. Each kink is identified by finding the point of inflection as the minimum in the next derivative \( d^2I/dV^2 \) as shown in Fig. 5b; its energy is labeled \( \Delta_0(r) \). Simultaneous \( \Delta(r) \) and kink-energy \( \Delta_0(r) \) maps can then be derived and are shown in Fig5c,d. By imaging \( \Delta_0(r) \) for all dopings, we find that the excitations are always divided into two categories: \( E<\Delta_0 \) excitations are homogeneous in \( r \)-space and well defined \( d \)-wave quasiparticle eigenstates in \( k \)-space[30,31], while for \( E>\Delta_0 \) they are heterogeneous[14,26,27,28,29] and ill-defined in \( k \)-space. Thus \( <\Delta_0> \) represents the average energy scale separating spatially homogeneous from heterogeneous excitations.

![Figure 5](image)

**Figure 5.** a) \( \Delta \)-sorted averaged \( dI/dV \) spectra. b) representative \( d^2I/dV^2 \) spectra. c) \( \Delta \)-map. d) \( \Delta_0 \)-map and its histogram.

6. Conclusions/Summary

First, in these studies we reveal a direct link between the dimensions of individual unit cells and the local gap maximum in a high temperature superconductor. The gap maxima \( \Delta \) are modulated sinusoidally by the changes in unit cell dimensions of Bi-2212, with the gap maxima occurring in association with the minima of Cu-O apical distance (when only the first harmonic of the crystal refinement is considered). The range of the gap modulation is found to be \( \sim9\% \) of the mean gap maximum independent of doping. Overall our data confirm directly at atomic scale the theoretical
concept of a strong influence of unit cell geometry on cuprate electronic structure and superconductivity[5,6,18].

Second, the $d^2I/dV^2$ –imaging studies of atomic scale EBI reveal an intense nanoscale disorder in the EBI energies $\Omega(r)$. Changing the hole-density has minimal effects on both the average mode energies $\bar{\Omega}$, indicating a mode unrelated to electronic/magnetic structure. Decisively, substitution of $^{18}$O for $^{16}$O throughout reduces $\bar{\Omega}$ by approximately $6\%\sim\sqrt{16/18}$, the expected isotope effect on a lattice vibrational mode[19]. Thus spatially disordered modes involving lattice vibrations that couple to the antinodal quasiparticle states of Bi-2212. Moreover, the mode energies are always anticorrelated with superconducting energy gap disorder.

Finally, we find that the excitations are always divided into two categories; homogeneous ($E<\Delta_0$) and heterogeneous ($E>\Delta_0$) excitations in $r$-space. Modulation of the gap maxima $\Delta$ due to the changes in unit cell dimension and an intense nanoscale disorder in the EBI energies $\bar{\Omega}$ as well as disorder in $\Delta$ are linked to the heterogeneous electronic excitations above $\Delta_0$. Further research is required to fully understand the interplay between electron-lattice interactions and superconductivity.

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