

Imaging the Essential Role of Spin Fluctuations in High- T_c Superconductivity

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We have used scanning tunneling spectroscopy to investigate short-length electronic correlations in three-layer $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Bi-2223). We show that the superconducting gap and the energy Ω_{dip} , defined as the difference between the dip minimum and the gap, are both modulated in space following the lattice superstructure and are locally anticorrelated. Based on fits of our data to a microscopic strong-coupling model, we show that Ω_{dip} is an accurate measure of the collective-mode energy in Bi-2223. We conclude that the collective mode responsible for the dip is a local excitation with a doping dependent energy and is most likely the (π, π) spin resonance.

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The presence of phonon signatures in the electron tunneling spectra of classical superconductors [1] and their quantitative explanation by the Eliashberg equations [2,3] stand among the most convincing validations of the BCS phonon-mediated pairing theory [4]. For high- T_c superconductors, the pairing mechanism still remains an intriguing mystery. Several cuprate superconductors present a spectroscopic feature, known as the dip hump [5–7], that resembles the phonon signatures of classical superconductors. There is still no consensus on the origin of the dip hump nor its connection to high- T_c superconductivity. In this work, we report on a scanning tunneling microscopy (STM) study of the three-layer compound $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10+\delta}$ (Bi-2223). We observe that the gap magnitude, a direct measure of the pairing strength, is periodically modulated on a length scale of about five crystal unit cells. This variation follows the superstructure, a periodic modulation of atomic positions naturally present in Bi-based cuprates. By fitting the STM data with a strong-coupling model [8,9], we demonstrate that the dip feature originates from a collective excitation. This allows us to image the collective-mode energy (CME) at the atomic scale and reveal a modulation that also follows the superstructure. The CME and the gap are locally anticorrelated. These findings support that the collective mode probed in our study is related to superconductivity and is most likely the antiferromagnetic spin resonance detected by neutron scattering [10]. Our results, in particular the CME value of 30–40 meV, are in agreement with the spin-fluctuation-mediated pairing scenario [11,12], in which the spin resonance in high- T_c s is a consequence of pairing.

An important issue in understanding high- T_c superconductivity (HTS) is the relevance of the observed inhomogeneous

electronic properties to superconductivity [13]. The relationship between these inhomogeneities and structural [14] or dopant atom [15,16] disorder is actively debated. Bi-based cuprates are suitable materials to study this problem since they naturally present a bulk modulation of atomic positions [17,18] unrelated to dopant inhomogeneities. A recent study on the two-layer compound $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ (Bi-2212) reports a spatial modulation of the gap following the superstructure [14]. This phenomenon calls for an investigation on the sensitivity of other spectroscopic features to variations of the interatomic distances. In particular, since the dip feature can be regarded as the fingerprint of Bogoliubov quasiparticles coupling with a collective excitation [8,9,19], its relation with the superstructure can bear information on the nature of this interaction.

The dip is detected neither above T_c nor within the vortex cores [13], suggesting an intimate connection with the superconducting state. We explore this possibility by mapping the local tunneling conductance of Bi-2223, the Bi-based cuprate with the highest T_c and the strongest dip [13,20]. We find that the local gap $\Delta_p(\mathbf{r})$ presents a one-dimensional modulation which follows the superstructure, being largest at the maxima of the surface corrugation. We observe that the energy separation between the dip and the coherence peak, Ω_{dip} , is also modulated with the same period. Excellent fits of the STM spectra with a model that considers a strong coupling of quasiparticles with a collective mode [8] establish that Ω_{dip} is an accurate measure of the CME. This allows us to reveal that the CME is locally anticorrelated with the gap. As we will discuss, these findings support that the collective mode is the (π, π) spin resonance [10].

Our crystals, free of Bi-2212 intergrowths, were grown by the traveling floating zone method and oxygen annealed [18,21]. We studied underdoped ($T_c = 103 \pm 2$ K), nearly optimally doped ($T_c = 109.5 \pm 0.5$ K), and overdoped ($T_c = 108.5 \pm 0.5$ K) crystals using a home-built UHV scanning-tunneling microscope. The crystals were cleaved at room temperature in a $1\text{--}5 \times 10^{-9}$ mbar atmosphere and immediately cooled down to 5 K.

STM measurements in Bi-2223 image the Bi atoms of the topmost BiO plane exposed by cleaving and the incommensurate modulation of the atomic positions (see Fig. 1). The unit cell vector of this superstructure \mathbf{a}_s is parallel to the \mathbf{a} axis. For the nearly optimally doped sample we estimate $a_s = 27 \pm 2 \text{ \AA} \approx 5a$, in agreement with x-ray diffraction data [18]. The maxima and minima of the surface corrugation are nearly straight lines perpendicular to \mathbf{a}_s . We choose one of the lines of minima as the origin and assign to each point \mathbf{r} a phase φ proportional to the orthogonal distance to this reference.

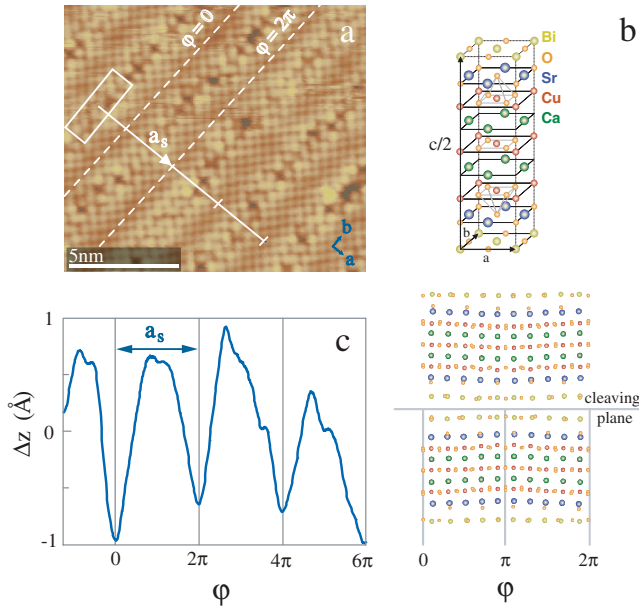


FIG. 1 (color). STM topography and crystal structure of Bi-2223. (a) Surface topography of an underdoped sample ($T_c = 103 \pm 2$ K) acquired at 5 K in constant-current mode (tunneling current set to 0.2 nA and sample bias to 0.4 V). The Bi atoms exposed after cleaving the sample are observed as bright spots. The in-plane unit cell vectors of the ideal crystal structure, \mathbf{a} and \mathbf{b} , and of the superstructure, \mathbf{a}_s , are indicated. Lines of constant phase are depicted [see also (b)]. The so-called missing atom row [32] is observed at $\varphi = (2n + 1)\pi$. (b) Top panel: upper half of the ideal Bi-2223 unit cell. Bottom panel: schematic representation of the periodic modulation of atomic positions along the c axis. The in-plane location is measured as a phase with $\varphi = 2n\pi$ corresponding to minima in the vertical position of the Bi atoms of the layer exposed by cleaving. (c) One-dimensional corrugation of the surface as a function of φ . The profile was obtained by computing the average height within the box depicted in (a) and by displacing this box along the white line.

For every pixel in a spectroscopic map, we assign a gap $\Delta_p(\mathbf{r}) = (\Delta_p^+ - \Delta_p^-)/2$, with Δ_p^+ and Δ_p^- the energy of the positive- and negative-bias coherence peaks in the dI/dV spectrum. We find that $\Delta_p(\mathbf{r})$ is locally correlated with the superstructure: in Figs. 2(b) and 2(d) the bright diagonal stripes corresponding to larger $\Delta_p(\mathbf{r})$ are located at the maxima of the surface corrugation. This phenomenon is observed in the three samples studied. Figure 3(b) shows that the gap distribution at constant φ follows a sinusoidal dependence on φ . Averaging all pixels at a constant φ results in a smooth curve with a peak-to-peak amplitude of $8 \pm 2\%$ of the mean value. This amplitude is similar to the $\approx 9\%$ value reported for Bi-2212 [14].

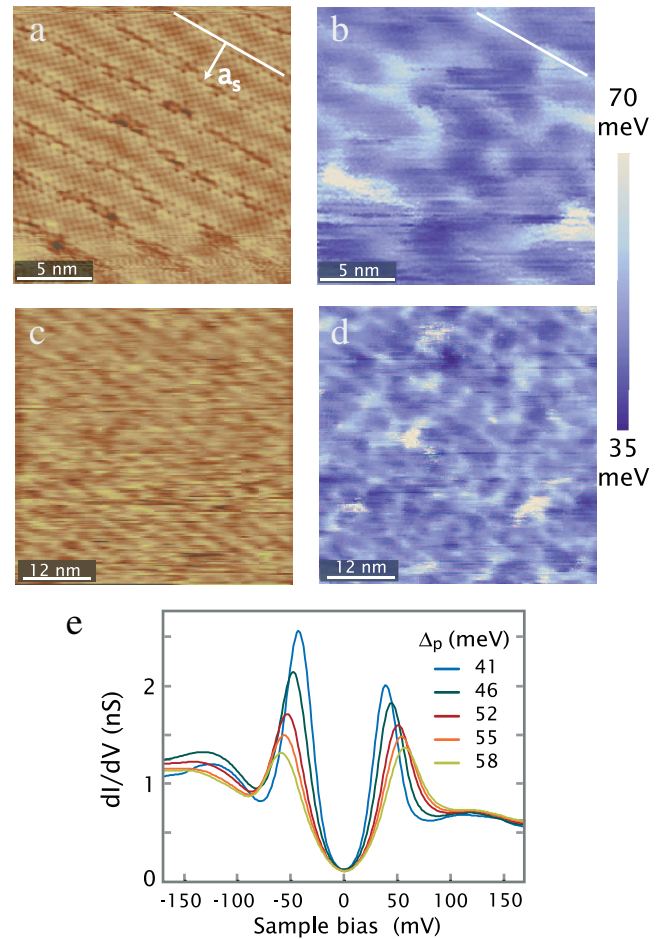


FIG. 2 (color). Local gap modulation and average spectra for nearly optimally doped Bi-2223. (a) Topographic image of a $17 \times 17 \text{ nm}^2$ region and (b) simultaneously measured local gap map $\Delta_p(\mathbf{r})$. The white line is a spatial reference. (c) Topographic image and (d) simultaneous local gap map in a $46 \times 46 \text{ nm}^2$ field of view in a different region of the sample. The color scales in (b) and (d) are identical and bright areas depict larger gap values. (e) Average tunneling conductance spectra obtained from thousands of spectra with the same gap acquired over the field of view of (b). Measurements were performed at 5 K.

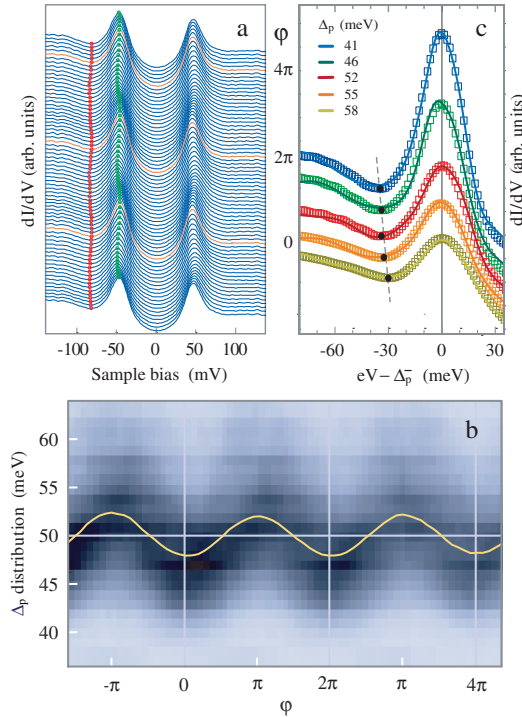


FIG. 3 (color). Spatial modulation of the gap and dip energy in nearly optimally doped Bi-2223. (a) Evolution of the dI/dV spectra with the phase ϕ : every curve results from averaging thousands of spectra with the same ϕ . The green and red dots indicate the coherence peaks and the dip feature for negative bias, respectively. (b) Color-scale representation of Δ_p distributions for spectra with the same ϕ : darker pixels correspond to more frequent gap values. The yellow curve depicts the average gap as a function of ϕ , $\bar{\Delta}_p(\phi)$. (c) Negative-bias part of the Δ_p -averaged spectra shown in Fig. 2(c), offset by Δ_p^- . Open symbols correspond to the experimental data whereas the lines are fits of the spectra with a strong-coupling model (see text). Black dots indicate the energy location of the dip in the experimental spectra and the dotted gray line is a guide to the eye. The spectra in (a) and (c) are vertically offset for clarity.

A first clue on the variation of the dip energy, E_{dip} , with the gap can be obtained from averaging spectra with the same Δ_p . The average spectra shown in Fig. 2(e) indicate that the coherence peaks' height and E_{dip} follow a systematic trend with the gap. Figure 3(c) shows the same spectra but with the energy shifted by Δ_p^- . This representation clearly shows that $\Omega_{\text{dip}} = |E_{\text{dip}} - \Delta_p^-|$ decreases when the gap increases, in agreement with tunneling and photoemission data obtained in Bi-2212 by several groups [9,22,23]. Recent STM results have suggested that Ω_{dip} is constant in Bi-2212 [24]. However, the spectra reported in Ref. [24] have a strongly asymmetric background, and a normalization procedure was used to remove this background before determining Ω_{dip} . In contrast, no background subtraction is necessary in our analysis, and Ω_{dip} is calculated directly from each of the raw dI/dV curves.

Furthermore, according to the data of Figure 3B in Ref. [24], the claim of a constant Ω_{dip} can only be made with an accuracy of roughly 10%, which is on the order of the systematic variation in Ω_{dip} that can be clearly seen in our measurements.

Determining the relationship between the gap and the CME requires a reliable estimation of both quantities from the tunneling spectra. We have fit the average spectra of Fig. 2(e) using a strong-coupling model [8] that considers a tight-binding band structure and the interaction of d -wave Bogoliubov quasiparticles with a (π, π) resonant mode [9,25]. Figure 3(c) shows the excellent agreement between the fits and the experimental data. Contrary to theoretical expectations [25], for all spectra in Fig. 3(c) the CME obtained from the fit falls within 1 meV of $\Omega_{\text{dip}} = |E_{\text{dip}} - \Delta_p^-|$ calculated from the experimental data. This allows us to deduce the CME directly from each of the $\sim 65\,000$ spectra in the spectroscopic maps and thus build $\Omega_{\text{dip}}(\mathbf{r})$ maps directly from raw data.

The $\Omega_{\text{dip}}(\mathbf{r})$ maps exhibit stripes running perpendicular to \mathbf{a}_s [see Fig. 4(b)]. The spatial modulation of $\Omega_{\text{dip}}(\mathbf{r})$ has the same period and direction as the superstructure, which is confirmed by the two peaks located at $\pm 2\pi/a_s$ in the Fourier transform of the map. We note that $\Omega(\mathbf{r})$ maps were

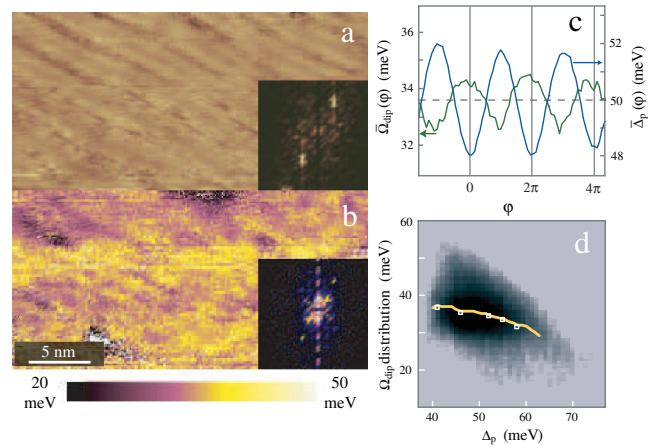


FIG. 4 (color). Spatial modulation of the collective-mode energy (CME) in nearly optimally doped Bi-2223. (a) Topographic image of a $30 \times 15 \text{ nm}^2$ region including the field of view of Fig. 2(a). Inset: square modulus of the Fourier transform of the topography depicting the two peaks at the wave vectors of the superstructure $\pm 2\pi/a_s$. (b) Simultaneously acquired CME map. White pixels indicate spectra where the energy of the dip was not resolved. Inset: Fourier transform of the CME map. (c) Phase dependence of the CME $\bar{\Omega}_{\text{dip}}(\phi)$ (green line) and gap $\bar{\Delta}_p(\phi)$ (blue line) averaged at constant ϕ . The gap and the CME are locally anticorrelated. (d) Color-scale representation of Ω_{dip} distributions for spectra with the same Δ_p . The yellow curve corresponds to the average Ω_{dip} as a function of Δ_p , computed from the geometrical mean of each histogram. The open squares indicate the CME obtained from fitting the average spectra shown in Fig. 3(c).

reported in Ref. [26] for Bi-2212. In these maps, Ω was determined from a peak in d^2I/dV^2 and no correlation with the superstructure was observed. Our fits presented here show that the dip minimum, not the d^2I/dV^2 peak, is the appropriate estimation of Ω . The stripes corresponding to low Ω_{dip} in Fig. 4(b) systematically coincide with large gap values. In order to correlate the φ evolution of the CME and the gap, we define a φ -averaged CME, $\bar{\Omega}_{\text{dip}}(\varphi)$, and compare it with $\bar{\Delta}_p(\varphi)$ in Fig. 4(c). The comparison demonstrates that $\bar{\Omega}_{\text{dip}}(\varphi)$ is anticorrelated with $\bar{\Delta}_p(\varphi)$. The peak-to-peak amplitude of the CME modulation is 6% of the mean value 33.5 meV. This anticorrelation is also evident from the distribution of Ω_{dip} for spectra with the same Δ_p [Fig. 4(d)]. The average Ω_{dip} deduced from the distribution is in excellent agreement with the CME obtained by fitting the average spectra with the same Δ_p .

The evidence presented in this work indicates that the collective mode at the origin of the dip is the (π, π) spin resonance detected by inelastic neutron scattering [27]. First, excellent fits of the STM data over a wide energy range are obtained only if assuming that the collective mode is located at (π, π) [8,9,25]. Second, we obtain a CME in the 30–40 meV range, in agreement with the few neutron data in Bi-based cuprates [10]. Third, the CME estimated from STM spectra decreases on increasing Δ_p [Fig. 4(d)]. In the case of underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, the only compound for which the doping dependence of the spin resonance has been thoroughly characterized, a similar trend is observed [27,28]. This must be contrasted with the almost complete absence of doping dependence found for optical phonons in Bi-2223 [29]. Our fourth piece of evidence is that the CME is modulated over the superstructure length scale of $\sim 5a$, implying that the collective mode must have a similarly short coherence length. Remarkably, the coherence length of the (π, π) resonance is only a few unit cells [27]. This new result, that the CME and the superconducting gap vary coherently over the short length scale of the superstructure, is the key point of our Letter.

It has been reported that the dip feature is neither detected above T_c nor within the vortex cores [13], and that the intensity of the spin resonance is negligible at $T > T_c$ [30]. These observations support that the (π, π) spin resonance has an intimate connection with the superconducting state. Several theories were proposed in order to explain high- T_c superconductivity, but none has yet managed to generate consensus in the community. Our results are in agreement with the spin-fluctuation scenario [31] that predicts the existence of a (π, π) resonance [11] as a consequence of the feedback of pairing on the spin fluctuations [12]. Nevertheless, the findings presented in this Letter challenge the theories that do not account for the short length scale modulation and the local anticorrelation

of the gap and the CME. Predicting this anticorrelation at the local scale is a critical test for any scenario aiming to explain high- T_c superconductivity.

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