Spatial modulation of midgap states in (001) $La_{1.88}Sr_{0.12}CuO_4$ films: Indications for antiphase ordering of the *d*-wave order parameter

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Using scanning tunneling spectroscopy we have investigated the spatial evolution of the anomalous *c*-axis zero-bias conductance peak, discovered in a previous study by our group, in epitaxial $La_{1.88}Sr_{0.12}CuO_4$ thin films. We found an anisotropic spatial dependence of the corresponding low-energy density of states which complies with the predicted spectral features of an antiphase ordering of the *d*-wave order parameter within the *ab* plane. Such an ordering was recently suggested to account for the 1/8 anomaly in the high-temperature superconductors and the dynamical layer decoupling recently reported to occur in the transport studies of $La_{15/8}Ba_{1/8}CuO_4$.

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I. INTRODUCTION

The family of lanthanum-based high-temperature superconductors, La214, exhibits an anomalous drop of the transition temperature, T_c, at the $x=1/8$ doping level, an effect also known as the 1/8 anomaly. In $La_{2-x}Sr_xCuO_4$ (LSCO) T_c drops at $x=1/8$ by about 30% with respect to the value of the "unperturbed" superconductor dome,[1,2] whereas in $La_{2-x}Ba_xCuO_4$ the effect is much more prominent and T_c reduces to almost zero.[3] At the same doping level, an anomalously small width of the peak momentum, associated with the electronic stripe phase was measured by neutron scattering on LSCO,[4] which in addition, exhibited a commensurate ordering of the stripes with the underlying lattice.[4,5] The coincidence of the two phenomena at $x=1/8$ has led to the conjecture that the stripe order competes with superconductivity, and their strong interaction is responsible for this anomaly.[6,7] This connection, however, has not been fully established yet, neither theoretically nor experimentally.

Recently, the transport properties of the stripe phase were examined in $La_{15/8}Ba_{1/8}CuO_4$ single crystals by Li *et al.*[8] Below the charge- and spin-ordering temperatures, a two-dimensional superconducting Berezinskii-Kosterlitz-Thouless (BKT) (Refs. 9 and 10) transition was identified. The extracted T_{BKT} exceeded the bulk T_c (<4 K), implying that for the $T_c<T<T_{BKT}$ temperature range, superconductivity is confined to two-dimensional planes with negligible interplane coupling, a behavior which was confirmed in a recent photoemission study of $La_{15/8}Ba_{1/8}CuO_4$.[11] Shortly after, Berg *et al.*[12] suggested that an antiphase ordering of the order parameter within each CuO_2 plane accounts for the above two transition temperatures reported by Li and co-workers. Apparently, this type of order-parameter modulation within the *ab* plane suppresses interlayer Josephson coupling while in-plane superconductivity survives.

The local density of states (LDOS) of a phase-biased junction comprising two *d*-wave superconductors was calculated by Tanaka and Kashiwaya[13] who found that for a π-phase difference, zero-energy Andreev bound states (ABSs) form at the interface. Such a junction is analogous to a domain wall (DW) where the order parameter undergoes a π-phase shift (πDW), described in the antiphase ordering model put forward by Berg *et al.*[12] The spatial evolution of the LDOS as a function of distance from a πDW, as well as the corresponding effects of the pairing amplitude, were calculated by Yang *et al.*[14] In the weak-pairing regime of this calculation, $\Delta \ll E_F$, the corresponding zero-bias conductance peak (ZBCP) appears in the LDOS at the vicinity of the πDW. A higher pairing amplitude, on the other hand, is predicted to induce an imbalanced splitting of the ZBCP at small distances from the DW due to the formation of a one-dimensional band of propagating ABSs along the πDW. For both pairing amplitudes studied, the spectral weight shifts from zero energy toward finite energies corresponding to the bulk superconductor-gap edges, as a function of distance from the DW. Away from the πDW, at the center of the domain, the LDOS was predicted to exhibit a suppressed peak at zero energy alongside pronounced peaks at the bulk gap edges. In contrast, for the more realistic scenario of an array of πDWs with the conventional $4a_0$ periodicity of the stripe ordering (where a_0 is the LSCO lattice constant), the calculated LDOS exhibited a gap centered at the chemical potential instead of the ZBCP. It was argued that the overlap of neighboring ABSs couples adjacent πDWs which yields a total quenching of the ZBCP.[14,15]

In spite of the compatibility of the antiphase model[12] with the results of Li *et al.*,[8] the existence of an antiphase ordering has not yet been established experimentally. An indication for the existence of an isolated πDW phase was found in a previous study of our group,[16] albeit at the time this connection was not recognized. There we examined the LDOS near the $x=1/8$ doping level on *c*-axis $La_{1.88}Sr_{0.12}CuO_4$ films, using scanning tunneling spectroscopy (STS). Although a small fraction of the data comprised the expected *c*-axis V-shaped tunneling gap, we ascertained that the dominant spectral feature in the LDOS was a ZBCP. This surprising phenomenon was uniquely found in films with $x=0.12$ and replaced the V-shaped gap found in all other $(001)La_{2-x}Sr_xCuO_4$ samples we examined with $x\neq 0.12$. Noting that the ZBCP is a hallmark feature of *d*-wave superconductivity,[17,18] appearing in tunneling spectra measured on nodal or any in-plane orientated surface [except the antinodal (100) surface], we looked for evidence for extraneous (e.g., faceting induced) in-plane tunneling in our results, but to no avail. Within the sensitivity of our x-ray diffraction measurements, no indication for any in-plane oriented surface was found. On the other hand, sharp Bragg peaks, corresponding to a highly ordered c-axis phase, were detected in all of our films. In addition, facet tunneling effects (i.e., coupling to nodal surfaces residing at the side facets of the c-axis grains) were also ruled out, since our data revealed a spatial continuity of the ZBCP feature *on top* of the c-axis grains. Moreover, a finite ZBCP amplitude was detectable for all temperatures below T_c, contrary to the commonly reported disappearance of the nodal ABSs at temperatures well below T_c.[19,20] Thus, despite the similar spectral signature of our anomalous c-axis ZBCPs to tunneling spectra portraying conventional nodal surface ABSs, we concluded in Ref. 16 that our findings have a different, yet unresolved, origin.

Motivated by the newly proposed antiphase ordering at $x=1/8$,[12] and its possible relation to our previously found c-axis ZBCP,[16] we further pursued our STS study on (001)$La_{1.88}Sr_{0.12}CuO_4$ films in order to examine in detail the spatial evolution of the LDOS and, in particular, of the ZBCP. Our main finding is an anisotropy in the spatial dependence of the ZBCP amplitude. Along lines of sequentially acquired tunneling spectra we found a modulation of the ZBCP amplitude with a modulation length larger than the $4a_0$ interstripe spacing reported for LSCO($x=0.12$).[4] At the same rate of recurrence, lines of constant ZBCP amplitude were measured on the same area but at different directions. Occasionally, the modulated ZBCP was split into two imbalanced peaks. Our diverse spectroscopic features comply well with the antiphase ordering of the superconductor order parameter and its predicted effect on the LDOS (Ref. 14) in the limit of negligible coupling between adjacent πDWs. In addition, our results point to an inhomogeneous distribution of the pairing amplitude at the sample surface.

II. EXPERIMENT

Four 90 nm (001)$La_{1.88}Sr_{0.12}CuO_4$ films were epitaxially grown using the pulsed laser deposition technique on (100)$SrTiO_3$ substrates, as detailed elsewhere.[16] In addition, a nodal-surface-oriented (110)$La_{1.88}Sr_{0.12}CuO_4$ film was grown on (110)$SrTiO_3$ for comparison reasons. Despite our efforts, we were unable to obtain sharp topographic images (i.e., atomic-scale resolution) of the sample's surface due to the notoriously degraded surface of LSCO films[21] and only correlations of the spectra with the gross morphological features, namely, the crystalline structure of the films, were obtained, as demonstrated in our previous report.[16] A typical topographic map showing LSCO crystallites is presented in Fig. 1(a), allowing us to verify that the ZBCP features are not due to any side facet of the c-axis crystallite, and, in particular, not to (110) facets. Although correlations with the fine, atomic-scale, surface morphology of the samples could not be obtained, by acquiring our tunneling spectra along intersecting perpendicular lines, the spatial dependence of the surface DOS was elucidated to some extent. The length of the lines along which the spectra was acquired (at equidistant steps) was limited to a few tens of nanometers, at which point the spectra usually turned featureless (probably due to surface disorder), as demonstrated by the dashed curves in Figs. 1(b) and 1(d). The spectra were acquired with 100 $M\Omega-1$ $G\Omega$ tunneling resistance.

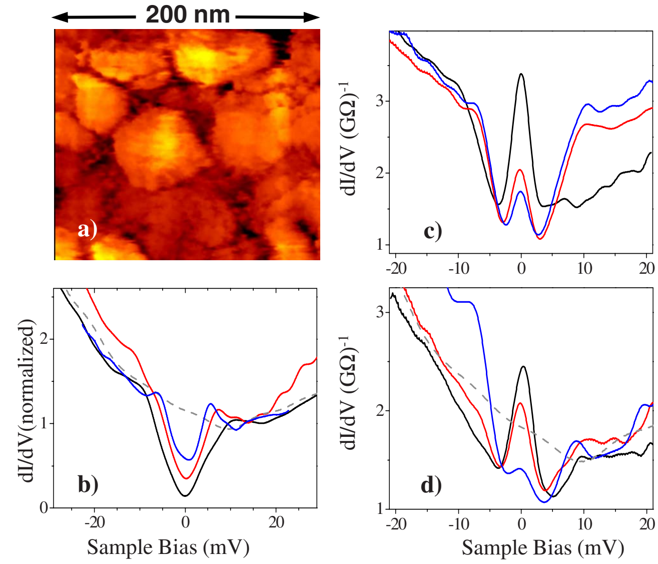

FIG. 1. (Color online) (a) 200×200 nm^2 topographic image of the sample surface showing LSCO crystallites. [(b)–(d)] Typical tunneling spectra of our c-axis LSCO($x=0.12$) samples. (b) V-shaped tunneling gaps expected to be found on c-axis surfaces, however these gaps consisted the minority of our data. The spectra were normalized with respect to the normal tunneling conductance at 13 mV, which exceeds the gap value. [(c) and (d)] The ZBCP, which was the predominant spectral feature of our data. Note that suppressed gaplike features are typically observed alongside the pronounced ZBCPs. The dashed curves in (b) and (d) portray the smearing effect of the surface disorder on the low-bias features. This type of spectra denoted ends or interruptions of the spectra lines.

III. RESULTS AND DISCUSSION

Tunneling into a c-axis surface of a d-wave superconductor yields V-shaped gaps due to the zero-energy nodal excitations. Part of our tunneling spectra featured such a V-shaped gap as presented in Fig. 1(b). Surprisingly, however, these gaps constituted less than 10% of the spectra that exhibited clear superconductor features, which were found on 30–40 % of the sample surface. The majority of the tunneling spectra exhibiting superconducting features, portrayed a ZBCP with gaplike features (GLFs). The variance of the ZBCP amplitude is shown in Figs. 1(c) and 1(d), which present two sets of dI/dV vs V curves (tunneling spectra), each taken along an individual line. Interestingly, it was commonly found that a pronounced peak entailed suppressed GLFs and vice versa [as evident from Figs. 1(c) and 1(d)], reminiscent of the predicted[14] spectral-weight shift as a function of distance from an isolated πDW [see Fig. 1(a) in Ref. 14]. However, no clear correlation was found between the ZBCP and GLF for the intermediate amplitudes, possibly due

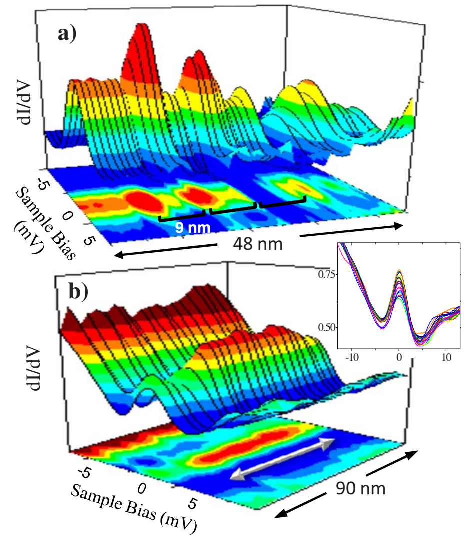

FIG. 2. (Color online) Two spectra lines showing modulated ZBCP amplitude. (a) The shortest modulation length of 3–4 nm is seen for all three ZBCP amplitude maxima. The intermaxima distance is 9 nm for the most pronounced ZBCPs. (b) The longest modulation length we encountered of ~60 nm. Inset: the dI/dV vs V curves taken in the region marked by the double-headed arrow at the base of the figure.

to the inhomogeneity of the superconductor gap, regularly found in the tunneling spectra of cuprates.[21–25] One should recall that the LDOS of a d-wave superconductor should feature a similar ZBCP with GLFs when tunneling into the ab plane (excluding the antinodal direction). In such setups, the GLF to ZBCP magnitude ratio is indicative of the in-plane tunneling angle with respect to the nodal direction. Obviously, this is not the case in our present c-axis films.

Due to the absence of a clear correlation between the midgap and gap-edge states we focus on the spatial evolution of the low-energy states, namely, the evolution of the ZBCP. In Figs. 2 and 3 we plot the dI/dV vs V tunneling spectra as a function of position of their acquisition. The detailed spatial evolution of the LDOS exhibited two distinct types of spatial dependencies. The first type consisted of a modulated ZBCP amplitude with a modulation length ranging from a few nanometers to tens of nanometers. The shortest modulation length we encountered is presented in Fig. 2(a) in which three local maxima of the ZBCP amplitude develop, each having a spatial width of 3–4 nm. About 9 nm part the two most pronounced ZBCPs, and ~18 nm separate the next pronounced peak (of reduced height). In between, and just midway the latter two peaks, one can see the effect of the surface disorder mentioned above, smearing the ZBCPs on a short length scale. The longest modulation scale we found was close to 60 nm and is presented in Fig. 2(b), in which a single wide maximum of the ZBCP amplitude was recorded along the line. The slow variation in the ZBCP magnitude within the segment around the maximal height (marked by a white double-headed arrow in the base of the figure) is demonstrated in the inset. Toward the edges of this line scan, a continuous reduction in the zero-energy spectral weight is noticeable over a length scale of 10–20 nm, an order of magnitude larger than the decay length shown in Fig. 2(a).

The second type of spatial dependence consisted of a constant ZBCP amplitude. Two typical examples are presented in Fig. 3, and evidently, these curves show a minuscule variation in the LDOS along the line. This is clearly illustrated in the inset of Fig. 3(b) where all the dI/dV curves collapse onto a single curve when all the spectra obtained along the entire line are plotted. Typically, the DOS in such cases was robustly fixed for the entire spectrum we examined, and not only at low energies on which the spectra in Fig. 3(a) focus. Figure 3(b) demonstrates the robustness of the LDOS for tunneling spectra taken along a 30-nm-long line, on a different sample. It is important to note that the lines in Figs. 2(a) and 3(a) were measured sequentially at perpendicular and intersecting trajectories.

The parallels of these results with the framework of the aforementioned isolated πDW scenario,[14] are compelling. In the limit of decoupled πDWs, the LDOS were predicted to consist of a ZBCP featuring a spatially dependent amplitude determined by the distance from the DW. Consequently, lines parallel to a πDW should exhibit a constant ZBCP amplitude while any trajectory intersecting a DW at an angle α, will result in a modulated ZBCP amplitude with an α-dependent modulation length, corresponding to the effective distance from the DW. Multiple decoupled πDWs may induce multiple maxima in the ZBCP magnitude along a line crossing them with spacing that depends on the inter-πDW distance and α. The sketch in Fig. 4(a) illustrates two different types of lines within the antiphase ordered plane along which tunneling spectra can be acquired, one running parallel to the πDWs and one crossing them. The spatial evolution of the LDOS in Figs. 4(b) and 4(c) are segments taken from the line scans presented in Figs. 2(a) and 3(a), respectively. Both lines comply well with the two different expected behaviors corresponding to the lines sketched in 4(a). The line connecting A and B in Fig. 4(a) crosses two πDWs (dashed lines), hence, a double-peak structure emerges in the corresponding spectra acquired along such a line, as shown in Fig. 4(b). In contrast, the parallel line connecting C and D should produce a constant ZBCP line, as shown in Fig. 4(c). Note that at a

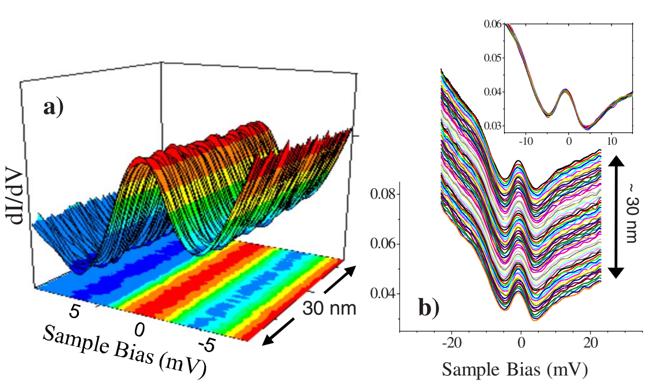

FIG. 3. (Color online) (a) Two spectra lines showing nonmodulated ZBCP amplitude. The scan line in (a) was taken on a line perpendicular and intersecting to that presented in Fig. 2(a). The line in (b) demonstrates that not only the ZBCP but also the GLF remains constant for the entire energy range we examined, as clearly portrayed in the inset where the same spectra plotted without any shift, appear to collapse onto a single dI/dV vs V curve.

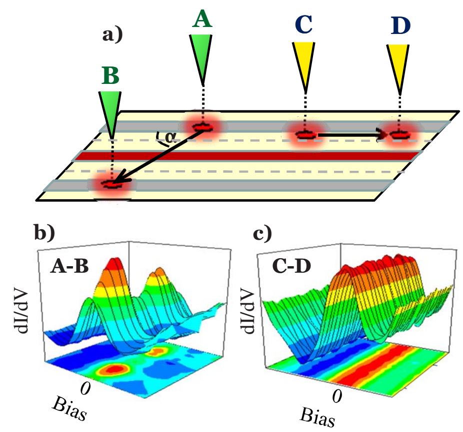

FIG. 4. (Color online) (a) Schematic illustration of the one-dimensional antiphase order. The colored lines represent the domain centers and the πDW is represented by the dashed line. (b) A constant ZBCP is expected for tunneling spectral lines in parallel to the DW like the one connecting points C and D in (a). (c) A modulation of the ZBCP amplitude should occur for any line with $\alpha \neq 0$. In the case of the line connecting the points A and B, a double-peak structure will emerge since it crosses two πDWs.

right angle ($\alpha = \pi/2$), the distance between the most pronounced ZBCPs (intermaxima distance) will be minimal and equal to the local spacing between adjacent πDWs.

Following the above considerations, we attribute our results to a state of decoupled πDWs by assigning an appropriate angle, α, to each line of spectra presented in Figs. 2 and 3. Obviously, $\alpha = 0$ for the lines exhibiting a constant ZBCP in Fig. 3. A small angle will precipitate the slow variation in the LDOS portrayed in Fig. 2(b) since the effective distance from the DW varies slowly along the line. In contrast, the rapid modulation of the LDOS in Fig. 2(a) will occur at large angles. The fact that the spectra presented in Fig. 2(a) were measured along a line intersecting at a right angle the line at which the spectra presented in Fig. 3(a) were acquired, indicates that for Fig. 2(a) $\alpha = \pi/2$, which implies that the observed 9 nm intermaxima interval corresponds to the local distance between the neighboring πDWs. Such distances are consistent with the assumed negligible πDW coupling, since they are almost twice the spatial width of the ABS, which is on the order of ξ_s (~ 5 nm in the case of LSCO). We also note that the modulation scale is larger than the spacing between charge stripes (~ 1.5 nm), an issue that is discussed further below.

A subtle feature of the modulated ZBCP, appearing only occasionally in the spectra lines, was an asymmetric splitting with a varying degree of imbalance between the negative and positive peak heights. The imbalance ranged from a small difference in peak heights to a state where one of the peaks was completely suppressed. The transition from a nearly fully suppressed positive peak, to a nearly fully suppressed negative peak, through a centered (unsplit) ZBCP, could take place even within a distance of a few nanometers, as demonstrated in Fig. 5(a) and highlighted by the three spectra presented in Fig. 5(b). Interestingly, the imbalance of the peaks in Fig. 5(a) meandered continuously between the points indicated by the arrows labeled 1 and 3, and in between (at point 2), where a pronounced unsplit ZBCP was observed, the low-energy spectral weight reached a maxima as clearly depicted in Fig. 5(b). Also note that dI/dV vs V curves featuring a split ZBCP [i.e., the spectra in the region enclosed by arrows 1 and 3 in Fig. 5(a)] exhibited a relatively high spectral weight at low energies compared to the curves at the bottom and top ends of the line, where a suppressed unsplit ZBCP was found.

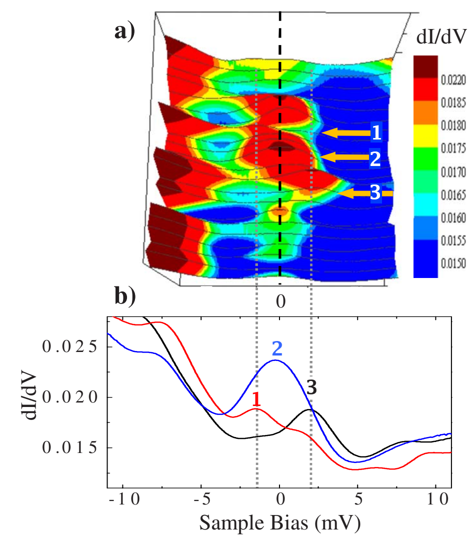

FIG. 5. (Color online) (a) The spatial evolution of a split ZBCP corresponding to a πDW at point 2 in the case of strong local pairing (see text). (b) dI/dV vs V curves exhibiting split ZBCPs and a pronounced centered (unsplit) ZBCP taken at the indicated positions in (a).

A markedly different spatial evolution of a split ZBCP was apparent along the line depicted in Fig. 6(a). Here, a periodic modulation was measured with a period of ~ 4 nm. Unlike the line in Fig. 5(a), the imbalance did not cross the zero energy. Instead, the negative peak was consistently higher than the positive peak throughout the entire line, as shown in Fig. 6(b), in which a selection of dI/dV vs V curves from Fig. 6(a) are plotted.

According to the decoupled πDW scenario[14] (as mentioned in Sec. I), in the case of a moderate pairing strength, an imbalanced split of the ZBCP is expected to occur at the

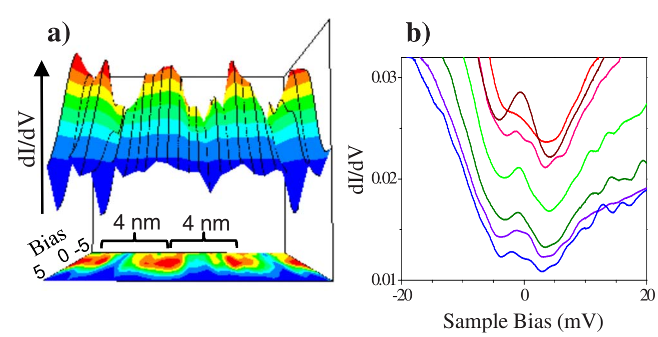

FIG. 6. (Color online) (a) A modulated split ZBCP with a very short modulation period. (b) Selected dI/dV vs V curves taken along the line in (a). The ZBCP appeared to be split with a negative peak consistently higher than the positive peak suggesting that no πDW has been crossed. All curves are vertically shifted for clarity.

vicinity of the DW. At sufficiently long distances from the DW the splitting disappears and a suppressed ZBCP is predicted to be found, as in the case of the weak-pairing limit, as indeed shown in Fig. 5. Consequently, the spatially modulated split ZBCPs presented in Figs. 5 and 6, can be understood within the framework of the decoupled πDW scenario assuming a locally moderate pairing strength (as opposed to the data presented in Figs. 2 and 3, where the data complies with a relatively weak-pairing strength). We note that lines of spectra showing either split or unsplit ZBCPs were found on the same sample, suggesting (according to Ref. 14) that the local pairing amplitude in our samples is spatially inhomogeneous. This is a reasonable conjecture in view of the superconducting-gap inhomogeneities reported in the cuprates[22] which have been considered to reflect an inhomogeneity of the local pairing strength.

Typically, the inversion of the split peak imbalance should take place at the πDW owing to the sign change in the order parameter. This implies that a πDW resides between points 1 and 3 in Fig. 5(a). The suppressed unsplit ZBCP measured at the bottom and top ends of Fig. 5(a) further corroborates this conjecture, since according to the isolated πDW scenario, the splitting occurs only in the vicinity of the DW where, in parallel, the low-energy spectral weight is maximal.

We now turn to discuss the spectra line presented in Fig. 6 that exhibits a periodic modulation of the negative-bias peak height of the split ZBCP, with a period length of ~ 4 nm, close to ξ_s. Such a short period seems to contradict the isolated πDW scenario since at these distances the inevitable overlap of the neighboring ABSs is predicted to quench the ZBCP.[14,15] However, the fact that the negative peak was consistently higher than the positive peak indicates that a πDW was not crossed and the line in Fig. 6(a) is in fact parallel to a πDW. In this case it is possible that the observed modulation is an outcome of the conjectured[14] one-dimensional band formed along the πDW in which the wave function of a charge carrier should oscillate with a period corresponding to the spatial width of an ABS, namely, $\sim \xi_s$.

We have also measured the tunneling spectra of a (110)$La_{1.88}Sr_{0.12}CuO_4$ reference film where ZBCP manifesting the nodal ABSs are the common spectral feature. In this sample only weak spurious variations in the ZBCP and gaplike features amplitude were found, in contrast to the well-defined spatial modulations seen on the (001)$La_{1.88}Sr_{0.12}CuO_4$ film, presented in Fig. 2. Moreover, no split ZBCPs were detected, thus ruling out the possibility of spontaneous splitting due to a subdominant order parameter as has been reported to occur in overdoped (110)$YBa_2Cu_3O_{7-\delta}$.[26,27]

A question that remains is whether the DWs are truly decoupled. Theoretical studies of the antiphase order predicted that π-phase (and consequently πDW) ordering could be unstable since the introduction of additional midgap states increases the energy of the system.[14,28] However, Yang *et al.*[14] find that the energy cost decreases as the domain size increases from the stripe periodicity of 4 unit cells, to 8 and 10 unit cells, although it remained positive for all studied values. Thus according to Ref. 14 it is more likely that an order of decoupled antiphase DWs (due to the large domain size) will form rather than a system of overlapping πDWs once an antiphase ordering sets in. We wish to emphasize however, that the ~ 9 nm spacing (corresponding to 6 stripe unit cells, $24a_0$, in LSCO) between ZBCP maxima observed for a line taken perpendicular to the πDW does not imply that this indeed is the generic spacing between πDWs. It may well be that, due to surface disorder, some πDWs are quenched, yielding spatial variations in the apparent πDW spacing.

Finally, we note that the theoretical works that analyzed arrays of coupled πDWs predicted that a gap will open at the chemical potential rather than a ZBCP. In our previous report[16] we have already disclosed that $\sim 10\%$ of the superconductor related data comprised a V-shaped gap. A gap of ~ 10 meV was extracted by fitting to the theory of tunneling into a d-wave superconductor.[18] It is possible that this gap is truly the c-axis pairing gap found at large enough distances from a πDW so that its effect is negligible. On the other hand, it may well be that the gap is not directly related to the superconducting order parameter and rather originates from the local overlap of a neighboring πDWs. This notion gains support in light of the relatively large variations in the gap features (e.g., width and zero-bias conductance) shown in Fig. 1(b).

IV. SUMMARY AND CONCLUSIONS

Our scanning tunneling spectroscopy data measured on c-axis $La_{1.88}Sr_{0.12}CuO_4$ films exhibit various features that are in accord with the predicted LDOS of the antiphase ordering in the limit of decoupled domain walls. The abundance of the ZBCPs found on our c-axis films is attributed to the formation of ABSs at the πDW and their spatial evolution is a consequence of the distance of the location of measurement from the nearby πDW. The spatial evolution of the imbalanced split ZBCPs, which is another trademark of the πDW model, points to an inhomogeneity of the pairing strength within the sample surface. We conclude that our results are indicative of a decoupled πDWs state that provides possible evidence for the predicted antiphase ordering of the superconductor order parameter at $x=1/8$. This is one of the first experimental indications for an anisotropic ordering of the d-wave order parameter.

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