Doping dependence of the \((\pi, \pi)\) shadow band in La-based cuprates studied by angle-resolved photoemission spectroscopy

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Doping dependence of the \((\pi, \pi)\) shadow band in La-based cuprates studied by angle-resolved photoemission spectroscopy

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Abstract. The $(\pi, \pi)$ shadow band (SB) in the La-based cuprate family (La214) was studied by angle-resolved photoemission spectroscopy over a wide doping range from $x = 0.01$ to $x = 0.25$. Unlike the well-studied case of the Bi-based cuprate family, an overall strong, monotonic doping dependence of the SB intensity at the Fermi level ($E_F$) was observed. In contrast to a previous report for the presence of the SB only close to $x = 1/8$, we found that it exists in a wide doping range, associated with a doping-independent $(\pi, \pi)$ wave vector but a strongly doping-dependent intensity: it is strongest at $x \sim 0.03$ and systematically diminishes as the doping increases until it becomes negligible in the overdoped regime. This SB with the observed doping dependence of intensity can in principle be caused by the antiferromagnetic fluctuations or a particular form of low-temperature orthorhombic lattice distortion known to persist up to $x \sim 0.21$ in the system, with both being weakened with increasing doping. However, a detailed binding-energy-dependent analysis of the SB at $x = 0.07$ does not appear to support the former interpretation, leaving the latter as a more plausible candidate, despite a challenge in quantitatively linking the doping dependences of the SB intensity and the magnitude of the lattice distortion. Our finding highlights the necessity for a careful and global consideration of the inherent structural complications for correctly understanding the cuprate Fermiology and its microscopic implication.

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1. Introduction

A current focus of high-$T_c$ superconductivity research is on the topology of the Fermi surface (FS) of underdoped cuprates in the normal state well above the superconducting transition temperature $T_c$ (see [1] and other papers in the same focus issue). Whether it is a single, large,
hole-like FS or consists of multiple, small, electron- and hole-like FSs, determines whether high-
$T_c$ superconductivity is an instability of a single-band metal [2] or its transformed version due to
some lattice (rotational and/or translational) symmetry breaking [3]. Central to this debate is the
existence and origin of shadow bands (SBs), which potentially represent additional ingredients
in the band structure that goes beyond the single-band picture.

The SB is defined in relation to the main band (MB). It generally has weaker spectral
weight and holds a certain symmetry relation with the MB. Its appearance in angle-resolved
photoemission spectroscopy (ARPES) [4] indicates the existence of some form of super-
modulation (of wave vector $\vec{q}$) in the crystal that scatters the electronic states from the MB at $\vec{k}$
to the SB at $\vec{k} + \vec{q}$. Depending on the form of super-modulation, it usually has a structural [5],
electronic [6] or magnetic [7] origin, as has been demonstrated in various materials. Concerning
high-$T_c$ superconductivity, electronic or magnetic super-modulation, as a result of collective
excitations of the many-body electronic system, has been generally deemed to be more relevant.

In particular, about the SB with $\vec{q} = (\pi, \pi)$ in Bi-based cuprates, consensus was finally
reached after years of debate between scenarios for its magnetic [8] or structural [9, 10] origin.
The lack of additional renormalization in the dispersion and lack of additional momentum
broadening of the SB relative to the MB and the binding energy independence of the intensity
correlation of the SB over the MB (SB/MB) [11], all argue against the magnetic interpretation where
the antiferromagnetic correlations in this hole-doped system are generally recognized to be
dynamic rather than static. This intensity ratio was found to exhibit a doping dependence in the
underdoped regime that is opposite [12] (if finite [13]) to the expectation for the weakening of
antiferromagnetic correlations as doping increases. On the other hand, the structural alternative
was confirmed by a polarization dependence study of untwinned samples, which uncovered the
hidden fourfold symmetry-breaking nature of the SB formation [14]. This SB has thus been
concluded to arise from a unique form of (bulk) orthorhombic distortion of a tetragonal lattice
structure that breaks its original fourfold symmetry and primarily takes place in the BiO planes.

Such a structural interpretation unique to Bi-based cuprates apparently cannot be directly
applied to explain a similar existence of $(\pi, \pi)$ SBs at various doping levels of La214 re-
ported in the literature [15]–[20]. However, in the low-temperature orthorhombic (LTO) phase of
La214 [21, 22] and also locally in the high-temperature tetragonal (HTT) phase above [23], another form of $(\pi, \pi)$ orthorhombic lattice distortion exists, which is caused by a staggered tilting of the CuO$_6$ octahedra about the Cu–O bond diagonal. While the contribution of this structural aspect might be similarly expected to dominate the SB formation in La214, a recent report [20]
on an anomalous enhancement of the SB intensity at $x = 1/8$ argued for its intimate connection
with the coincident static spin and charge stripe orders, despite the fact that these orders are
associated with some incommensurate wave vectors different from $(\pi, \pi)$ [24]–[26]. Such an
anomaly tied to $x = 1/8$, if it turns out to be true, would strongly support the multi-band nature
of the band structure of La214 in the normal state and its origin due to a specific form of lattice
symmetry breaking. However, a considerable intensity of this SB has been reported at other
doping levels [15]–[19], which casts such anomaly into doubt. Before a reliable evaluation of its
potential importance for a unified understanding of the origin of the SB in cuprates can be made,
a careful re-examination of this possible anomaly in La214 has to be performed, which should
be placed in a wider context by means of a more systematic doping dependence study in La214.

In this paper, we present ARPES data at doping levels ranging from $x = 0.01$ to $x = 0.25$
for various members of La214. Unlike the Bi-based cuprate family, a strong, monotonic doping
dependence of the SB intensity at the Fermi level ($E_F$) was observed. Although the SB intensity
fluctuates by a considerable amount from experiment to experiment at a given nominal doping

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level, we found that it is generally stronger at lower dopings and systematically weakens with the increase of doping until it becomes negligible in the overdoped regime. The observed existence of the SB over a wide doping range and its overall weakening of intensity are very different from the previous report for a maximal SB intensity at \( x = 1/8 \) and negligible intensity at other dopings [20]. While such a trend of SB intensity appears to be consistent with decreasing strength of antiferromagnetic fluctuations in the system, a similar lack of additional renormalization in the SB dispersion and of binding energy dependence of the SB/MB intensity ratio is found and exemplified at \( x = 0.07 \), which questions the conventional magnetic interpretation as proposed for the Bi-based case [11, 27]. On the other hand, the observation that the SB becomes virtually undetectable near \( x \sim 0.21 \) is broadly consistent with the structural interpretation based on the aforementioned orthorhombic distortion, which disappears at \( x \sim 0.21 \). However, important discrepancies in their detailed doping dependences are noted, which pose a challenge for establishing their precise connection in a quantitative fashion.

2. Experimental

ARPES measurements were carried out at Beam Line 10.0.1 in Advanced Light Source (ALS) using different versions of the Scienta electron energy analyzer, SES-200, SES-2002 and SES-R4000, over the years (1999 to the present). The photon energies \((h\nu)\) used were 55, 55.5 and 59.5 eV and the polarization of light was fixed in plane and orthogonal to the zone diagonal (nodal) direction during each experiment by virtue of a sample-fixed analyzer-rotating capability. The typical energy resolution was \( \sim 20 \) meV and angular resolution was \( \sim 0.25^\circ \) \((0.015\,\text{Å}^{-1} \text{ in momentum})\). Single crystals grown by the traveling solvent floating zone method were studied for different members of the La214 family, \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) (LSCO or LS, grown by groups in Tokyo Institute of Technology [28, 29], University of Tokyo [30], Electric Power Industry [31] and Chinese Academy of Sciences [32]), \( \text{La}_{2-x}\text{Ba}_x\text{CuO}_4 \) (LBCO or LB, grown by groups in Tohoku University [25, 33, 34]) and 1% Fe-doped LSCO (Fe-LSCO or FeLS, grown by a group in Tohoku University [35]). The \( T_c \) versus doping relations are similar to published results (of LSCO [21], LBCO [34] and Fe-LSCO [36]) but vary slightly among those grown by different groups. Samples were cleaved in vacuum with a typical base pressure better than \( 5 \times 10^{-11} \) Torr and measured at temperature \( \sim 20 \) K.

3. Results and discussion

3.1. Existence of the \((\pi, \pi)\) shadow band at \( x = 0.07 \)

Figure 1(a) shows an example of the typical FS mapping geometry of this study. The spectral intensity in the nodal region of the second Brillouin zone (BZ) is relatively enhanced compared to the one in the first BZ due to the matrix element effects [15, 37]. Despite the suppression of overall intensity and quasi-particle spectral weight (figure 1(e)), the spectra along the nodal cut in the first BZ of LSCO \( x = 0.07 \) show two counter-dispersing bands that have comparable spectral weight (figure 1(c)). Their momenta are approximately symmetric about the \((\pi, \pi)\) BZ boundary (figure 1(d)). We denote hereafter the ones marked in blue and in green by the MB and its \((\pi, \pi)\) SB, respectively. In contrast, the spectra taken at the nodal cut in the second BZ are dominated by a single dispersing band with barely noticeable intensity on its corresponding SB feature (figures 1(b) and (d)). The inequivalence of the SB/MB intensity ratio at equivalent in-plane momenta indicates that the SB and MB have different matrix element effects. This
Figure 1. (a) FS map of LSCO $x = 0.07$ measured by SES-2002 across two BZs ($Z'$–Γ–$Z$) taken with cuts parallel to the bond diagonal (nodal) direction. $h\nu = 55$ eV. The intensity integration window is $E_F \pm 5$ meV. The blue dashed line is the $(\pi, \pi)$ BZ boundary, whose crossing with a given cut is denoted by $k_{AF}$ hereafter. (b, c) ARPES intensity as a function of binding energy and parallel momentum along cuts 1 and 2, the nodal direction in the second and first BZs, respectively. The momentum direction and range as indicated by the red and green arrows correspond to those in (a). (d) Intensity line profiles of cuts 1 and 2 in (a) shown in corresponding colors. Red, blue and green vertical bars indicate the Fermi crossings ($k_F$) of MB in the second BZ and of MB and SB in the first BZ, respectively, which correspond to those in (b) and (c). The bar thickness reflects the uncertainty in determining $k_F$. (e) Energy distribution curves (EDCs) at $k_F$ marked by the same colors as in (b) and (c).

can be more easily understood if the SB reflects an inherent symmetry of initial states (i.e. the SB exists in the band structure), similar to the Bi-based case [14,15], rather than being a trivial replica of the MB due to the final-state photoelectron diffraction.

3.2. Momentum-distribution-curve analysis of the $(\pi, \pi)$ shadow band at $x = 0.07$

To check whether a magnetic interpretation is compatible with the observed $(\pi, \pi)$ SB, we have performed a momentum-distribution-curve (MDC) analysis similar to [11] in figure 2. At $x = 0.07$, commensurate antiferromagnetic fluctuations have a broad maximum centered at the energy transfer position $\sim 25$ meV according to [38]. As discussed in [11] for the Bi-based case, if they are at play in our case, one expects the following observations of the SB

15 Note that our LSCO samples are not detwinned in terms of the orthorhombic distortions, so a similar polarization dependence study to [14] was not performed. In fact, according to that work, the current polarization condition for Cut 2 in figure 1(a) should favor an anti-phase intensity distribution between the MB and SB, which is not found in our experiments, presumably due to the different nature of distortions in La214 and/or the twinned nature of crystals.
Figure 2. (a) MDCs from $E - E_F = -104$ meV (bottom) to 11 meV (top) shown in 5 meV steps for figure 1(c). Red curves are MDC fits with two Lorentzians and a linear background. (b–d) Results of the MDC analysis of the MDC peak dispersion, MDC peak width (full-width at half-maximum (FWHM)) and MDC peak intensity, respectively, of the MB and SB in the first BZ (as shown in (a)), in comparison with the results for the MB in the second BZ (as shown by figure 1(b); error bars not shown are slightly smaller than the former). In (b), the SB dispersion is folded symmetrically about $k_{AF}$ for comparison. Error bars are from the MDC fitting. Inset: results of fitting each peak individually with a Lorentzian and a linear background for the MB and SB in the first BZ. The SB/MB intensity ratio is also shown in (d).

(in comparison to the MB): (i) an energy renormalization in the dispersion due to the fermionic exchange of spin fluctuations at finite energy transfers; (ii) an additional momentum broadening of the order of $0.1 \text{ Å}^{-1}$ due to the short-range nature of the fluctuations; (iii) a binding energy dependence of the SB/MB intensity ratio by the same argument as in (i). Within experimental error bars, neither of the above predictions is supported by the results shown in figures 2(b)–(d), respectively, making the magnetic interpretation a less likely candidate for the cause of the SB.

Note that the existence of a kink-like feature in the SB dispersion at $\sim -25$ meV is an artifact due to the imperfect fitting quality of the two-peak fit, which can be improved by fitting the SB or MB peak feature individually (the inset of figure 2(b)); the MDC peak width of the SB appears to be systematically smaller than that of the MB, which is beyond the error bars, robust
against different MDC fitting schemes and quite commonly seen for other dopings (figure 3(a)). Its trivial cause by the sample misalignment can be ruled out as many cuts, for example, Cut 2 (figure 1(a)), are precisely aligned with the high-symmetry direction.

Another puzzling observation was found when we performed the MDC analysis of the MB in the second BZ based on a single Lorentzian fit. While its peak dispersion and intensity are similar to those of the bands in the first BZ within error bars (figures 2(b) and (d)), the peak width has a different energy dependence and the deviation is more pronounced towards higher binding energy (figure 2(c)). This observation that both bands in the first BZ tend to broaden more radically than that in the second BZ as a function of binding energy is also visible by eye in figures 1(b) and (c). We note the following. Firstly, a similar observation has also been made at all other doping levels on which we could reliably perform a similar MDC analysis. Secondly, if one compares states in different 2D projected BZs, there are additional complications due to the effects of finite $k_z$ dispersion and matrix element. At this stage, we can fairly speculate at least two corresponding possibilities. There could exist a $k_z$-dependent scattering mechanism that causes different broadening of states at different $k_z$. More interestingly, some additional nodal states located away from $E_F$ could be responsible for the additional energy broadening observed in the first BZ but have an unfavorable photoemission matrix element in the second BZ with our experimental settings.

Both the above anomalies in the MDC peak width of the SB and MB and of the bands in the first and second BZs have not been reported previously, to our knowledge. Their origins are currently unclear and probably deserve special attention and future studies.

3.3. Doping dependence of the $(\pi, \pi)$ shadow band

We have compiled data of comparable quality which were similarly taken at different doping levels of various members of La214, as shown in figures A.1–A.3. The $(\pi, \pi)$ SB can be seen for many doping levels, consistent with previous reports [15]–[20]. If we simply focus on the first BZ where its existence is most obvious in the field of view for each FS map, we can see that it is generally more pronounced for low dopings than for high dopings, hinting at its finite doping dependence.

The simultaneous detection of the SB and MB within a single cut measured with identical instrumental settings (polarization, mapping geometry and $h\nu$), e.g. Cut 2 in figure 1(a), offers us a good chance to quantitatively assess the doping dependence of the SB intensity. The SB and MB can have different matrix elements but they are largely doping independent, as doping should not change the characters of states but mainly their weights and relative proportions. We thus normalize the intensity at $E_F$ of the SB by the MB in the same cut and directly compare this SB/MB intensity ratio, which mainly reflects the intrinsic spectral weight of the SB, between different experiments at different dopings. Such a normalization scheme also allows us to bypass the difficulty in the spectral intensity normalization between different momentum cuts, e.g. cuts 1 and 2 in figure 1(a), and to avoid most time-dependent instrumental artifacts due to the beam and/or manipulator instability, etc.

By using a similar two-peak MDC fitting routine, we obtain the doping dependence of this ratio (figure 3(c)), $k_F$ separation and the midpoint between the SB and MB (figure 3(d)). Within error bars, the midpoint is maintained close to $k_{AF}$, suggesting that the wave vector associated with the SB is close to $(\pi, \pi)$ and independent of doping; the $k_F$ separation shows a systematic increase with doping, which is consistent with the increase of Luttinger’s volume of the FS and the corresponding shift of the node position away from $k_{AF}$ [16].
Figure 3. (a, b) Maximum-normalized intensity line profiles along the nodal cut in the first BZ (shown in green in figure A.1(a)) for all FS maps shown in figures A.1–A.3. Green curves are fits with two Lorentzians of identical FWHM and a linear background. Note that the FWHM asymmetry for the MB and SB as discussed in section 3.2 is not addressed here. Identical FWHM were assumed in order to reduce the number of fitting parameters. A two-Lorentzian fit with this constraint removed yielded similar results within error bars for the cases where the SB intensity is strong, but it often gave unphysical results for the SB–MB midpoint for the cases where noise dominates over the SB intensity. (c) The SB/MB intensity ratio at $E_F$ by the fitting. (d) $k_F$ separation (measured from $k_{AF}$, solid symbol) and the midpoint (empty symbol) between the SB and MB by the fitting. Error bars are from the fitting. Color ribbons are guides to the eye. (e) Doping dependence of the HTT–LTO phase transition temperature and $T_c$ for LSCO, reproduced from [23].

We will focus on the SB/MB intensity ratio at $E_F$ in the following. A salient feature of figure 3(c) is that there exists quite a large scattering in this ratio beyond the fitting error bars between different experiments on different samples at a given doping level. Because most instrumental artifacts mentioned above are expected to be eliminated by taking the intensity ratio, this result more likely reflects the uncertainties related to the samples, e.g. the variations
In the surface doping, impurity concentration and/or cleaving quality, etc. It, in turn, highlights
the necessity for a systematic doping dependence study, with particular attention paid to the
reproducibility of results, in order to obtain true insights into this problem. In this regard,
the abrupt jumps observed at \(x = 0.01\) and \(x = 0.11\), each from only one sample, should be
treated cautiously and be subjected to more studies at these two doping levels, before a reliable
conclusion on their intrinsic or extrinsic nature can be reached.

Despite the pronounced scattering of results at a given doping level and the uncertain
anomalies at \(x = 0.01\) and \(x = 0.11\), the intensity ratio shows an overall monotonic doping
dependence. At low dopings, the SB intensity appears very strong, even comparable with the
MB intensity at \(x = 0.03\), and has a strong doping dependence. Both the intensity and its doping
dependence weaken with an increase of doping. At high dopings, the doping dependence, if
finite, seems very weak. Affected by a noise level that is comparable to the weak SB intensity,
the two-peak fitting routine tends to give finite peak intensity value in this regime. Although it is
in principle uncertain with such large error bars (and scattering of results) whether the slope is
zero or finite, we note that the SB can show up on some samples and at some particular doping
levels in this regime, e.g. at \(x \sim 0.15\) in figure 3(b), which is consistent with what was reported
in [17].

Besides, from our data, it is difficult to tell a significant difference in the SB intensity
between \(x = 1/8\) and above. In contrast, it was reported previously that the SB intensity is
enhanced at \(x = 1/8\), compared with both its lower and higher doping sides where virtually no
SB could be identified [20]. At \(x < 1/8\), the discrepancy with our data is even larger: here we
can clearly observe a systematically stronger SB intensity than at \(x \geq 1/8\). We note that the
previous result was based on a study of three doping levels, each with only one sample. In light
of our preceding discussion, one should generally be cautious not to over-interpret the results
obtained in such a way.

The strong doping dependence in the low-doping regime contrasts with the Bi-based
case whose doping dependence is subtle and still controversial (doping dependent [12] or
independent [13]). Together with the weak doping dependence in the high-doping regime,
this unique behavior presumably holds the clue to understanding the origin of the \((\pi, \pi)\)
SB in La214. While the magnetic scenario can likely be ruled out based on the binding
energy dependence results at \(x = 0.07\), we are left with the structural alternative to be
discussed below.

The \((\pi, \pi)\) orthorhombic distortion sets in at the HTT–LTO phase transition which shows
an increased distortion magnitude as temperature decreases [22]. As a result, the distortion
magnitude at a fixed low temperature, which determines how much spectral weight is transferred
from the MB onto the SB, decreases with doping. This scenario generally expects a stronger
SB intensity for a larger distortion magnitude and can thus explain the observed monotonic
decrease of the intensity ratio in a qualitative fashion. It also anticipates no SB to be observable
above \(x \sim 0.21\), which is also consistent with the experiment. However, significantly strong
distortions with a strong doping dependence have been found to persist close to \(x \sim 0.20\). For
example, the phase line runs through the optimal doping almost linearly at \(\sim 200\) K, as shown
in figure 3(e). This suggests a strong orthorhombic distortion at our measurement temperature
and its strong doping variation, which are seemingly at odds with the weak SB intensity and
its weak doping dependence observed at \(x \sim 0.15\) and above. Nevertheless, we should remark
that such a semi-quantitative assessment of the structural perspective for our experimental result
might be premature. The exact relationship between the distortion magnitude and the intrinsic
spectral weight of the SB is yet to be established, which might or might not be quadratic (as expected for a simple final-state photoelectron diffraction). Our observation discussed previously, that the SB more likely arises from the scattering of the initial states, is relevant in this regard.

Although our finding suggests that the \((\pi, \pi)\) SB might not be directly connected with the stripe order as previously suggested [20], it does not preclude the connection of the stripe order with the mysterious normal-state properties of cuprates. In fact, it has been recently shown that the stripe fluctuations, the precursor to the stripe order, set in below the pseudogap temperature \(T^*\) for both the Y-based and La214 families [39]. A latest scanning tunneling spectroscopy study supports a similar conclusion for Bi-based cuprates [40]. Combined with our recent observation of the violation of particle–hole symmetry (as a hallmark of superconductivity) of the pseudogap [3], it is therefore very likely that the normal-state band structure of cuprates is far more complicated than the single-band starting point that has been generally assumed before 2006 [41]. The band structure reconstruction due to the fluctuating stripe order necessarily produces SBs at momentum locations that are determined by, among others, the incommensurate ordering wave vectors. However, because of the short-range and fluctuating nature of the order, these SBs could lack intrinsic spectral weight and appear poorly defined in momentum space, and thus are hard to detect [42, 43]. A structural interpretation for the \((\pi, \pi)\) SB, which is associated with a commensurate wave vector and has an overall comparable spectral intensity to the MB, is hence not inconsistent with such a picture.

4. Summary

We have presented the first comprehensive doping dependence study of the SB in La214 by ARPES. Its corresponding wave vector is close to \((\pi, \pi)\) and independent of doping. The SB at \(x = 0.07\) is shown to present a similar binding energy dependence with the MB in terms of the dispersion, quasi-particle momentum linewidth and spectral weight. Different from the Bi-based family, the intensity of SB at \(E_F\) in La214 exhibits a systematic, monotonic change with doping, from being strong at low dopings to being negligible at high dopings. Essentially similar to the Bi-based case, we find that an interpretation based on a unique form of structural distortion provides a better account of the origin of this SB than the magnetic alternative, although there seems to exist a semi-quantitative discrepancy from the experiment, which warrants further theoretical examinations. Our results are inconsistent with a maximal SB intensity previously reported at \(x = 1/8\) and suggest that the manifestation of the stripe correlations in the band structure could be in a more subtle way.

Acknowledgments

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Figure A.1. FS maps of LSCO at different doping levels. Data were taken with SES-200 analyzers from November 1999 to February 2001. $h\nu = 55.5$ eV. $E_F \pm 10$ meV.

Figure A.2. FS maps of LSCO at different doping levels. Data were taken with SES-2002 analyzers from May 2002 to April 2004. $h\nu = 55$ eV for (e–g) and $h\nu = 59.5$ eV otherwise. $E_F \pm 10$ meV.
Figure A.3. FS maps of LBCO, LSCO and Fe-LSCO at different doping levels. Data were taken with SES-R4000 analyzers since April 2006. $h\nu = 55 \text{ eV}$. $E_F \pm 10 \text{ meV}$. The data of LBCO-1/8 are reproduced from [44], Fe-LSCO $x = 0.25$ from [36]. Additional features, having an overall angular offset from the main one, in the first BZs of Fe-LSCO $x = 0.25$ are from another crystalline domain.

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