Experimental proof of a structural origin for the shadow Fermi surface in Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

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By combining surprising new results from a full polarization analysis of nodal angle-resolved photoemission data from pristine and modulation-free Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ with structural information from LEED and $ab$ initio one-step photoemission simulations, we prove that the shadow Fermi surface in these systems has structural origin, being due to orthorhombic distortions from tetragonal symmetry present in both surface and bulk. Consequently, one of the longest standing open issues in the fermiology of these widely studied systems finally meets its resolution.

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The Fermi surface and associated low-lying electronic excitations are central to the physical properties of metallic solids. Thus in systems displaying highly anomalous and complex electronic behavior, such as the high-$T_c$ cuprate superconductors, much effort is expended in the study of their 'fermiology' using photoemission (ARPES)\footnote{1}. In this context, although Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (Bi2212) is one of the most intensively studied solids known, significant pieces of the Fermi surface puzzle for these compounds have still managed to remain in the shadows. Fittingly, it is the so-called shadow Fermi surface (SFS) in these systems - together with the main FS (MFS) forming the two primary features of the intrinsic fermiology of Bi2212 - which has eluded a definitive interpretation up to the present.

In this letter, we prove that the SFS in Bi2212 and Pb-doped Bi2212 are intrinsic parts of the occupied electronic states of these systems. By combining new data on the polarization dependence of these states in photoemission with electron diffraction data and one-step $ab$ initio photoemission simulations, we can definitively identify structural distortions from tetragonality that result in the electronic bands giving rise to the SFS. Consequently, the shadow Fermi surface in Bi2212 has lost its enigmatic character and is now needs a new name.

The SFS resembles a copy of the main FS, but shifted by $(\pi,\pi)$\footnote{2} and equivalent vectors in the 2D tetragonal Brillouin zone, and was first observed in 1994\footnote{3}. This feature is an accepted part of all high quality ARPES data from Bi2212 and the related Bi2201 (Bi$_2$Sr$_2$CuO$_{4+\delta}$) high $T_c$ superconductors. Two groups of mechanisms have been proposed for the SFS. In the first, the shadows are magnetic: short range antiferromagnetic (AFM) fluctuations being the culprit\footnote{4}. In the second, the shadows are of a non-magnetic origin\footnote{5}, including the suggestion of back-folding of bands due to departure from the ideal tetragonal structure of Bi2212 and related systems\footnote{6}. Within the AFM scenario, significant renormalization of the shadow band (SB) dispersion with respect to that of the main band (MB) should occur, and the SB intensity should be strongly doping dependent. A recent ARPES study\footnote{7} argued convincingly against the AFM scenario, due to the lack of additional renormalization, doping or temperature dependence of the SB signal in photoemission. The question then arises: rather than simply stating that the AFM scenario is incorrect, can one prove that these primal Fermi surface features in the drisophila of high $T_c$ ARPES investigations are of structural origin? Are they thus anchored in the initial states relevant not only for photoemission, but also for transport and other physical properties?

In order to answer these questions, we carried out ARPES at the SIS beamline of the Swiss Light Source, with typical energy and angular resolution of 34(40) meV and 0.3° for $h\nu = 40(100)$ eV. High quality pristine and Pb-doped Bi2212 untwinned single crystals were grown using floating zone and flux methods, mounted on a 5-axis cryomanipulator allowing polar and azimuthal rotation of the sample in ultra-high vacuum with a precision of 0.1°. Cleavage and measurement took place at T=15K.

In the geometry used, the synchrotron radiation, sample surface normal and the detected photoelectrons all lie in a horizontal plane. The beamline provides linearly polarized radiation (polarized either horizontally [p-polarization] or vertically [s-polarization]) or circularly polarized radiation (denoted $\sigma^+$ for left circular polarization). As a result, for a high symmetry line such as (0,0)-($\pi,\pi$), which is a structural mirror plane in the tetragonal cell\footnote{2}, we can classify the initial states as being either even or odd, and thus expect to detect intensity from an even(odd) initial state only for p(s)-polarization. In this context, $\sigma^+$ polarized light can be considered as a superposition of p- and s-polarization, and consequently we use it to get a view of all features concerned.
FIG. 1: (color online) (a) Sketch of the Bi2212 Fermi surface. EDMs ($h\nu = 100$ eV) measured at location I in (a) using the polarizations indicated. The white lines show MDCs at $E_F$. (e) map of intensity at $E_F\pm 15$ meV from region II, measured with $s$-polarization and $h\nu = 40$ eV.

Fig. 2: (color online) EDMs ($h\nu = 40$ eV) measured at location III in Fig. 1(a), using the polarizations shown. The $E_F$ MDCs are shown as white lines. The inset to panel (a) shows a blow-up of the $E_F$ MDC, including a Lorentzian as a guide to the eye.

FIG. 2: (color online) EDMs ($h\nu = 40$ eV) measured at location III in Fig. 1(a), using the polarizations shown. The $E_F$ MDCs are shown as white lines. The inset to panel (a) shows a blow-up of the $E_F$ MDC, including a Lorentzian as a guide to the eye.

polarization. Surprisingly, the shadow features are essentially unchanged with respect to the $\sigma^+$ case. Switching to $s$-polarization (Fig. 1(e)), we now see that the situation has reversed: only the MB related features are visible, whereas the MB emission is suppressed. Fig. 1(b) illustrates that the 'switching off' of the main band (in fact a DR of the main band, labelled 3) occurs only within $\pm 2^\circ$ of $\Gamma Y$, and also that the SB intensity is maximal exactly along the nodal line. The fact that the MB initial states are odd with respect to the $\Gamma Y$ line, and - unexpectedly - the SB initial states are even will prove very important for the elucidation of the microscopic origin of the SFS.

We now turn to the $\Gamma X$ direction, in which (for pristine Bi2212) the contributions from the main, shadow and replica features are less widely spaced. In fact, the MB and its associated DRs are only separated by 0.07 Å$^{-1}$, which is resolvable in Fig. 2 by using the lower photon energy of 40 eV. Fig. 2(a) is recorded with $\sigma^+$ radiation. On the left is an intense feature related to the MB, and on the right, a weak feature originating from the SB. At $E_F$, the momentum distribution curve (MDC) shows that the left-hand feature can be resolved into two distinct branches, with that closest to $\Gamma$ being the main band and the other being due to main band DRs (see also Fig. 1(a)). After zooming the scale of the MDC (see inset), a similar analysis is possible for the shadow states. Here the component furthest from $\Gamma$ is the SB. The intensity observed in the inset at the left hand side of the Lorentzian is attributed to the SB DRs.

Fig. 2(b) now shows the same $\Gamma X$ cut, but measured with $p$-polarization. Now the intensity from both the MB and the SB is highly suppressed, whereas the DRs of both bands are still observed. This means that along $\Gamma X$, the MB and SB show identical polarization behavior, and originate from initial states which are odd with respect to reflection in $\Gamma X$.

As ever in the Bi-based high $T_c$ superconductors, it
is wise to check the validity of an important result in modulation-free, Pb-doped crystals. Fig. 3(a–c) shows the results for ΓY. Using $\sigma^+$ polarization, we see both the MB and SB. For $\rho[\sigma]$ polarization (panels b[c]) the same behavior as in the pristine case is evident with opposite polarization behavior of the MB (odd initial state) and SB (even initial state).

By now the alert reader will pose the question: why is the symmetry character of the MB and SB alike in one nodal direction, whereas in the orthogonal nodal direction they behave as opposites? The first point here is that the initial state symmetry has to be involved, as for these data recorded along high symmetry lines we are able to rule out an origin arising purely from multiple scattering in the photomission final states. As a second point we recall that the AFM scenario for SFS formation has essentially been disqualified both from earlier data as well as from a similar analysis of the data presented here. Thus, our next step is the suggestion that an orthorhombic structural distortion could lie at the root of things. Not only would this lead to a back-folding of the MB into a smaller, orthorhombic Brillouin zone, there is also experimental evidence for such a structural modification from low energy electron diffraction (LEED), x-ray and neutron diffraction of both Bi2212 and (Pb,Bi)-2212. The trick in the tail is the explanation as to how this would lead to a change in parity of the backfolded (SB) states, but only along one of the two nodal directions present in the basic tetragonal structure.

To facilitate the discussion, we introduce a toy model in order to illustrate the qualitative behavior. We start from a $c(2\times2)$ unit cell in the undistorted tetragonal system, which would be, in-fact, non-primitive and which contains two identical Cu atoms per plane, delivering two identical ZRSs. These states all have odd parity with respect to both ΓY and ΓX and give the MB: this tetragonal structure has no SB. If we now introduce a real orthorhombic distortion, a SB appears. In this sense, the wave function responsible for the SB is the difference between the distorted (orthorhombic) and undistorted (tetragonal) wave functions. We can write the overall wave function of the distorted system as $\phi_D(x, y) = D(x)\phi(x, y)$, where $D(x)$ is the distortion (along one direction in space) and $\phi(x, y)$ the undistorted wave function. If the distortion is small, we can write: $D(x) = 1 + xD'(0) + \ldots$. The total wave function thus becomes $\phi_D(x, y) = \phi(x, y) + xD'(0)\phi(x, y) + \ldots$, now reformulated in terms of the undistorted wave function plus a distortion of strength $D'(0)$. The distortion - i.e. the feature responsible for the SB wave function - is proportional to $x\phi(x, y)$. Crucially, since both the functions $x$ and $\phi$ are odd in $x$, their product will be even. We note that a second order term (or other even numbered terms) in the perturbation would not lead to a parity swap: only the odd terms can do this. This situation is reminiscent of the k-dependent parity swaps observed for states in other systems possessing glide-mirror planes, such as pyrolytic graphite and C on Ni(100).

The toy model, in fact, describes the essence of the surprising and new experimental results presented here. As evident from diffraction results, the distorted structure only has a single mirror plane, parallel to the $xz$-plane: the $yz$-plane being rather a glide-mirror plane (however, both the $xz$- and $yz$-planes are mirror planes of the 'distortion wave function'). The XRD results indicate a significant shift of Bi atoms in the BO layers (by as much as 0.1 Å) in the $x$ direction away from their ideal tetragonal sites, resulting in two Bi sub-lattices. This shift is sketched in Fig. 4(a). Such displacements should and do show up as $c(2\times2)$ superstructure spots in LEED data. We now take the LEED analysis an important step further with the data shown in Fig. 4(b). The spots are indexed according to the orthorhombic $a$ and $b$ axes. Clear superstructure spots (with respect to the tetragonal cell) are visible, as expected. A closer inspection reveals that the $(h=\text{odd}, k=0)$ spots are missing (independent of the electron beam energy). These systematic extinctions point unambiguously to the presence of an orthorhombic distortion giving only a single nodal mirror plane running parallel to $xz$, wholly in keeping with our simple model and Fig. 4(a).

In the final leg of our journey out of the shadows, we test the illustrative picture developed thus far by means of DFT-based one-step photoemission calculations (for the methodology see Ref. 19). Here we are able to deal explicitly with the $k$- and $\omega$-dependence of the matrix elements governing the photoemission intensity. The input of the calculation is modulation-free Bi2212 in the average structure from Ref. 11. Figs. 4(c) and (d) show portions of calculated Fermi surface maps for polarization vectors along $x$ and $y$, respectively. From the calculated intensity distributions, we can see that the MFS displays the expected behavior. It is suppressed along the zone diagonals for $p$-polarization and is strong for $s$-polarization.
Crucially, the SFS behaves differently along \( \Gamma Y \), showing a parity swap along the vertical nodal line. For the \( \Gamma X \) azimuth, the SFS follows the intensity distribution of the \( \pi \)-polarization vector aligned as indicated. The dotted boxes indicate the relevant polarization and the black (red) 1's and 0's mark the presence and suppression of the main shadow features, respectively.

In summary, by proving the microscopic structural origins of the shadow bands in the Bi22212 and Bi2201 families of cuprate superconductors, we have finally been able to close this chapter in the rich and complex tale of the high \( T_c \) superconductors.

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