A re-examination of the electronic structure of $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $Bi_2Sr_2Cu_1O_{6+\delta}$ - An electron-like Fermi Surface and the absence of flat bands at $E_F$.

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We present a re-examination of the electronic structure and Fermi Surface (FS) of Bi-Sr-Ca-Cu-O (BSCCO) as obtained from angle-resolved photoemission experiments. By applying a stricter set of FS crossing criteria as well as by varying the incident photon energy outside the usual range, we have found very different behavior from that previously observed. In particular we have found an electron-like FS centered around the $\Gamma$ point, and the flat bands at $E_F$ near the $\bar{M}$ point of the zone are absent. These results are robust over a large range of dopings and from single to double layer samples.

Angle-Resolved Photoemission Spectroscopy (ARPES) has emerged as one of the most powerful tools for unearthing the electronic structure and physics of the high temperature superconductors (HTSC) and other correlated electron systems since it allows one to directly probe the $\vec{k}$ space information of the electronic structure [1]. Major discoveries obtained from ARPES experiments on the HTSC's have included the observation of flat bands (an extended van Hove singularity) at or very near the Fermi level [2,3,4], a superconducting gap with d-wave symmetry [6,7], and an anomalous pseudogap above $T_c$ [8–10]. Such advances are all predicated on a thorough knowledge of the normal-state FS topology of these superconductors, which has been almost universally accepted to be a hole-like pocket centered around the Brilloin zone corners ((\$\pi, \pi\$) or X, Y points) [2,3,4]. In this Letter we argue that this most fundamental assumption of the FS topology is incorrect or at least greatly oversimplified. This finding should have a major impact on many previous studies and theories, including all those concentrating on the flat bands at $E_F$, the superconducting gap, and on the normal state pseudogap.

The measurements we report on here were taken on high quality single crystals of the BiSrCaCuO (BSCCO) family of cuprate superconductors. These are nearly ideal materials for surface sensitive ARPES studies because of the beautiful cleaved surfaces that are obtainable. Thus the ARPES information is usually interpreted as being representative of the bulk physical properties of BSCCO.

To date ARPES data from BSCCO have been mostly limited to a narrow photon energy range between 19 and 25eV. This has been largely a matter of convenience as well as the general expectation that the physics observed at other photon energy ranges would be essentially unchanged. We have measured BSCCO at photon energies well outside this range and have found very different physics from that observed between 19 and 25eV.

Most of the data presented in this paper was taken with a photon energy of 33eV, at which we find qualitatively different but much clearer behavior than that observed between 19 and 25eV. Namely, the data shows with high clarity the existence of an electron-pocket Fermi surface as well as the absence of flat bands at $E_F$.

The experiments were mostly performed at the Synchrotron Radiation Center in Wisconsin with a few backup experiments performed at the Stanford Synchrotron Radiation Laboratory in California. At both labs we used VSW 50mm hemispherical energy analyzers mounted on two-axis goniometers. The total experimental energy resolution was about 50meV FWHM and the angular resolution was $\pm 1^\circ$. All data shown in this paper was taken at or near 100K, comfortably in the normal state. Most of the $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) samples for this study came from CRIEPI, with a portion coming from ETL. The $Bi_2Sr_2Ca_1O_{6+\delta}$ (Bi2201) sample studied came from the University of Tokyo.

Fig. 1(a) shows Ding et al's version of the generally accepted hole-like FS topology of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), from one of the most complete and heavily referenced data sets available [1]. The thick lines represent the FS due to the main $CuO_2$ band. The thin lines are FS replicas due to the superstructure modulation with a $Q = (0.2\pi,0.2\pi)$. Support for such a topology is displayed in Fig. 1(c) and (d). Here we have Energy Distribution Curves (EDCs) from Ding taken at hv=19 eV along the high symmetry cuts $\Gamma - M$ and $\bar{M} - X$, where $\Gamma = (0,0)$ and $\bar{M} = (\pi,0)$. From these two panels, the authors claim there is no main band FS crossing along $\Gamma - \bar{M}$ but that there is one along $\bar{M} - X$, as evidenced by the relatively rapid loss of peak weight in this region.

Figure 1(e) presents a small portion of our new normal-state ARPES data at 33eV obtained from an overdoped Bi2212 sample along the high symmetry cut $\Gamma - M - Z$ ($Z = (2\pi,0)$). The energy scale for these spectra is the
same as in Ding’s data in panels (c) and (d). Our peaks are sharper and better resolved than Ding’s, showing the very high quality of the data set. Clear dispersion is observed with the peak reaching $E_F$ near $\sim 18 \pi$ (red curve). At this same k-value, the peak rapidly loses weight, indicating a FS crossing. The peak reappears in the second zone and disperses back towards higher binding energy (BE). Although only a single cut, this data already indicates that there is a fundamental difference between our data at the new photon energy of $\sim 33eV$, and the previous data of Ding et al. Our data indicates main-band FS crossings along $\Gamma - M - Z$, while they are not expected according to the accepted hole-like FS topology. In addition, the flat bands at $E_F$ at $\Gamma$ (extended van-Hove singularity) observed near $20eV$ have been replaced at $33eV$ by a strongly dispersive band which crosses $E_F$.

To analyze the data in more detail we have made $\vec{k}$-space plots of the integrated spectral intensity $n(\vec{k})$ as well as the weight right at $E_F$ which we term $E_F(\vec{k})$. We first illustrate how $n(\vec{k})$ and $E_F(\vec{k})$ are expected to behave for a simple band-like state crossing the FS. Fig. 2(a) shows the zero-temperature $n(\vec{k})$ and $E_F(\vec{k})$ for a non-interacting system. In this case there is only weight at $E = E_F$ when $\vec{k} = \vec{k}_F$. In Fig. 2(b) we introduce interactions, which within the Fermi Liquid theory framework will reduce both the weight of the delta function peak in $E_F(\vec{k})$ and the step in $n(\vec{k})$ to the value $Z$ (quasiparticle weight). Finite experimental energy and momentum resolution will broaden both curves, as illustrated in Fig. 2(c). Finally, polarization and matrix element effects will slowly alter $n(\vec{k})$, as shown in Fig. 2(d). It is clear from the above plots that at a true FS crossing the following criteria should be obeyed: (1) $E_F(\vec{k})$ should be maximal. (2) $n(\vec{k})$ should be at 50% of it’s maximal value, or equivalently at the maximal gradient point. Also, at $E_F$ we expect (3) the peak dispersion to extrapolate to zero energy, and (4) the midpoint of the leading edge of the spectrum to be at or even beyond (on the unoccupied side of) $E_F$.

Panels (e)-(i) of figure 2 show real $n(\vec{k})$ and $E_F(\vec{k})$ plots from our new data on the BSCCO family. To our knowledge, this is the first time that $n(\vec{k})$ and $E_F(\vec{k})$ plots have been analyzed together, which turns out to be a powerful new tool to obtain FS crossings. To determine $n(\vec{k})$ we integrated the ARPES spectral weight from $-500meV$ to $+100meV$ so as to span the full energy width of the peak, and for $E_F(\vec{k})$ we integrated over a $50meV$ wide window centered at $E_F$. All plots were normalized so that the maximum weight along $\Gamma - M - Z$ was set to 1. According to criteria (1) and (2), a FS crossing should occur when $n(\vec{k})$ loses half of its maximum value (excluding the background) and $E_F(\vec{k})$ simultaneously peaks. These points are indicated in the figure by the dashed green lines. Panel (e) shows $n(\vec{k})$ and $E_F(\vec{k})$ obtained from the raw data of Fig. 1 (e). As expected, this way of analyzing the data (criteria 1 and 2) gives identical FS crossings as obtained by studying the peak dispersion (criteria 3 and 4). Importantly, the drastic drop in $n(\vec{k})$ at $\bar{M}$ to a value comparable to that at $\Gamma$ or $Z$ indicates that this crossing must be due to the main band and not due to the crossing of a weak superstructure band. Also, the drop in $n(\vec{k})$ at the $Z$ point to a level equivalent to that at the $\Gamma$ point can not be explained by photon polarization or orbital symmetry arguments. Although not central to this paper, this observation includes new physics which warrants much further experimental and theoretical attention.

Panels (f) and (g) show data taken at $hv=22eV$ from a similar sample used to make the $33eV$ data of panel (e). Our raw $22eV$ data looks qualitatively similar to Ding’s data of figure 1(b) or to other previously published data including the presence of flat bands at $E_F$ near $\bar{M}$. The data of panels (f) and (g) show that the peak in $E_F(\vec{k})$ and the 50% point of $n(\vec{k})$ do not coincide, but rather the peaks of each coincide. This behavior is extremely unusual. Furthermore, the data of panel (f) is highly asymmetric about $\bar{M}$ (in the 2d approximation $\Gamma$ would be equivalent to $Z$ and $\bar{M}$ would be a real high symmetry point), in contrast to the much more symmetric behavior observed at $33eV$. This strange behavior makes us question the nature of the states probed near $\bar{M}$ at photon energies near 22 eV.

The interesting behavior of the data at $33eV$ calls for a complete mapping of the FS topology at this new photon energy. We have taken many cuts over the Brillouin zone on a slightly overdoped Bi2212 sample (different from that used in figures (1) and (2)). Within experimental resolution, each of the FS crossing criteria gave identical crossing locations for each cut. The crossing points indicated from these cuts are shown in Fig. 3. We have connected these points with thick lines indicating the main FS (stronger ARPES peaks) and thin lines indicating the superstructure-derived FS (ARPES peaks of about 30% the intensity of the main peaks). The superstructure FS’s are seen to be replicas of the main FS’s, but shifted by $\pm 0.2(\pi,\pi)$. We note that the FS topology measured here is closed around the $\Gamma$ point, i.e. it is electron-like. This is manifestly different from the “accepted” hole-like FS topology of Bi2212.

Despite this discrepancy, there is data in the literature that confirms our new FS topology. Figure 3 shows an overlay of our new FS with an $E_F$ intensity plot from an optimally doped Bi2212 crystal at 33 eV measured by Saini et al. The $E_F$ intensity is analogous to our $E_F(\vec{k})$, i.e. the maximum intensity locations should correspond to FS crossings. The overlay shows a striking similarity between these two independently obtained results. However, instead of interpreting their data as an electron-like FS, Saini et al argued that it was still representative of a hole-like FS centered around X and Y.
They attributed the loss of weight at $\tilde{M}$ to a pseudogap in the spectral function, and further claimed that the high intensity cusps orthogonal to the $\Gamma - Y$ direction were due to the formation of one-dimensional charge stripes. Our experiments and analysis attribute the weight loss around $\tilde{M}$ to a FS crossing. Indeed our data of figure 1(e) shows no evidence for a pseudogap in these samples (the signature of a pseudogap in ARPES is that at $k = k_F$ the leading edge of the spectrum is depressed from $E_F$). Figure 1(e) shows that the midpoint of our leading edge is fully up to or even past $E_F$. Our data also indicates that the periodic arrangement of high intensity cusps are due to the superstructure bands. Meser et al have also argued that superstructure bands and not stripes are important for understanding Saini’s data [1]. However, in their picture the data is analyzed starting from the standard hole-like FS and so they are not capable of explaining the near-complete weight loss observed at $\tilde{M}$.

Figs. 2(h) and (i) show that the new behavior detailed here is robust as a function of sample type. These plots show $n(\vec{k})$ and $E_F(\vec{k})$ along $\Gamma - \tilde{M} - Z$ at hv’s near 33 eV for a heavily underdoped Bi2212 sample and for an overdoped sample of the single-layer compound Bi2201. In both cases the data indicates a main band crossing between $\Gamma$ and $M$, indicating that the FS topology should also be electron-like for these samples (closed around $\Gamma$) (the crossing is more washed out for the underdoped sample, but appears to be qualitatively similar in other respects). This indicates that the new physics shown here is not peculiar to one doping level or sample type. Rather, we argue that it is a product of the new photon energy range used for our measurements.

Changing the photon energy in an ARPES experiment can have a number of effects. For a fixed $k_\parallel$, the most obvious effect is that $k_\perp$ will change since the magnitude of the total momentum must change. In this way, variations in the electronic structure vs. $k_\perp$ may be mapped out, i.e. we can map out the full three-dimensional electronic structure. We have taken data at many more photon energies (not shown here) to check whether the Fermi surface topology oscillates as a function of $k_\perp$. We did not observe any clear effects with a periodicity in $k_\perp$ of $2\pi/c$ where $c$ is the c-axis lattice constant. Thus we conclude that the differences in the data as a function of photon energy are not naturally linkable to a coherent three-dimensionality of the band structure. This is consistent with the huge in-plane vs. out-of-plane transport anisotropy of these materials.

Figure 4a shows the generally accepted $E$ vs. $\vec{k}$ relation for near-optimal BSCCO, with a key feature being the large $k$-space region near $\tilde{M}$ which has flat bands just below $E_F$. Our figure 2g indicates that the crossing along $\Gamma - Y$ is not robust, so we conclude that figure 4a is not a good representation of the physics. Rather, we propose the scenario in figures 4b and 4c. The main band dispersion shown by the red lines is as we have measured at 33eV, and is electron-like with a small saddle-point at $\tilde{M}$ above $E_F$. Additionally, there is a large region of flat bands at $\tilde{M}$ which is observed at 22eV but not 33eV. At 22 eV these states act to mask the true crossing behavior of the bands observed at 33eV, giving the impression of a hole-like FS. An important goal for future studies will be to elucidate the origin of these additional states, including whether or not they are intrinsic to the basic electronic structure of BSCCO. Possible origins include quantum confinement due to stripe formation [13], a contribution from the BiO states [20,21], indirect transitions [22], photoelectron diffraction effects [23], final state (i.e. matrix element) effects [24], or possibly a new type of correlated electron state due for instance to magnetism.

Considering that electron transport should be dominated by the dispersive states we have observed at 33eV, we are left with the puzzling result that the FS topology looks electron-like, while Hall effect measurements indicate that the carriers should be hole-like [24]. A similar disagreement has been reported for the n-type superconductor $N_{d_{2-x}}Ce_xCuO_4$ - Hall effect measurements have indicated an electron-like FS [28] while ARPES results have indicated a hole-like FS [25]. These results indicate a non-simple relationship between the Hall resistance and the electronic structure in these materials. This again highlights the importance of physics beyond the band structure (i.e. correlation effects) in the cuprates.

In conclusion, by invoking a more complete set of FS crossing criteria and by going to a non-traditional photon energy range we have discovered an electron-like FS topology and the absence of flat bands at $E_F$ in the BSCCO family of hole-doped superconductors. The data is robust as a function of doping and is clearer than the previous data which was interpreted as indicating a hole-like topology. The main dissimilarity in the data sets occurs near $\tilde{M}$, which is a critical location in the Brillouin zone - it is where both the superconducting gap and normal state pseudogap were found to reach maximum amplitude [10]. Clearly this calls for more experimental and theoretical works considering this new FS topology as well as the effects of varying photon energy.

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FIG. 1. Normal-state ARPES from Bi2212. (a,b) One quarter of the Brillouin zone showing (a) the hole-like FS extrapolated by Ding et al. [11] and (b) the electron-like FS determined here. Dark lines are the main FS pieces while the light lines represent superstructure-derived FS replicas. (c,d) Raw data from Ding taken at $h\nu = 19$ eV along directions indicated in panel (a). (e) A portion of our new data taken at $h\nu = 33$ eV, with each $\vec{k}$-space point listed as a fraction ($x\pi$) of the $\Gamma - \bar{M}$ distance.

FIG. 2. (a-d): Schematics of the expected behavior of $n(\vec{k})$ and $E_F(\vec{k})$ for a band crossing the FS. (a) Non-interacting system at zero temperature. (b) Interacting Fermi Liquid with a reduced quasiparticle weight $Z$. (c) Same as (b) but with finite energy and momentum resolution. (d) Same as (c) but including polarization and matrix element effects. Panels (e)-(i) are $n(\vec{k})$ (red solid line) and $E_F(\vec{k})$ (blue dashed line) at different photon energies from BSCCO samples with varying number of $CuO_2$ layers and doping levels (indicated on panels). Green dashed lines indicate a main-band FS crossing. Superstructure-derived crossings are labeled s.s..

FIG. 3. FS crossing points (white) from 33eV data on slightly overdoped Bi2212. The thick black lines through the points indicate the main FS and the thin black lines indicate the superstructure-derived FS. Our data is overlayed on a color-scale plot of ARPES intensity at $E_F$ from an optimally doped Bi2212 sample, as measured at 33eV by Saini et al [12]. The highest intensity regions are yellow and the lowest intensity regions are black.

FIG. 4. Schematic diagrams of the $E$ vs. $\vec{k}$ relations for near-optimal BSCCO. (a) The old picture of the extended saddle point below $E_F$. (b,c) New dispersion relation showing a non-extended saddle point above $E_F$. We hypothesize an additional set of non-dispersive states below $E_F$ which are visible at 22eV but not 33eV, and which sometimes mask the true crossing behavior.
Chuang et al. Fig 1
Chuang et al. Fig 2
Chuang et al, Fig 3
Chuang et al. Fig 4