Evidence for Strongly Interacting Electrons with Collective Modes at \( q(-0.4\pi, 0.4\pi) \) and \( G(\pi, \pi) \) in the Normal Phase of High-\( T_c \) Superconductors


Here we report the \( k \) distribution of the photoelectron spectral weight in an energy window of 50 meV around the chemical potential giving a so-called marginal Fermi surface (MFS) of a representative high-\( T_c \) superconductor, \( \text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8 \). Combining two modes of angle-resolved photoemission (ARPES), namely the constant initial-state angle scanning (ASP) mode and conventional energy distribution curve (EDC) mode, we determined the wavevectors of the collective modes strongly interacting with the conduction electrons: a first one at \( q = (-0.4\pi, 0.4\pi) \) assigned to a charge density wave (CDW) that is second harmonic of the underlying incommensurate and anharmonic lattice fluctuation forming a superlattice of quantum stripes; and a second one at \( G = (\pi, \pi) \), associated with spin density waves (SDW).

1. INTRODUCTION

Anomalous electronic properties of high-\( T_c \) cuprates have been a point of wide discussion in recent years. Angle resolved photoemission (ARPES) is one of the few experimental tools that has been used to take up this task as the technique has the advantage of being resolved both in energy and momentum space [1]. Study of the evolution of anomalous single-particle properties of high-\( T_c \) superconductors could be possible due to availability

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of better quality samples with a wide range of doping and new developments of infrastructure for performing ARPES experiments.

There is growing experimental evidence for the breakdown of the one-electron picture in high-$T_c$ superconductors. The breakdown occurs for a strongly interacting electron gas where the Fermi liquid model of quasi-particles is not valid. In this situation, one expects that the energy indetermination of charge carriers in the conduction band is of the order of the bandwidth, i.e., in the photoemission spectra, the uncertainty in the energy determination is of the same order of magnitude as its energy dispersion. The well-known example of a strongly interacting electron gas, where the Fermi liquid model breaks down, is a one-dimensional conductor where charge current can flow through collective excitations with separation of spin and charge modes.

Recent experiments are uncovering the fact that the doped perovskites are complex materials with segregation of localized and itinerant charge carriers in stripes [2]. It has been found by combined analysis of EXAFS and diffraction studies [3–7] that high-$T_c$ superconductivity coexists with an incommensurate and anharmonic lattice modulation, forming stripes in the CuO$_2$ plane along the diagonal ($-\pi, \pi$) direction. These lattice fluctuations coexist with incommensurate spin fluctuations with slow dynamics in the horizontal direction (the direction of Cu–O–Cu bonds) [8]. Based on the magnetic and charge superstructure peaks observed in insulating doped nickelates and 1/8 doped cuprates, a model of charge and spin fluctuating horizontal stripes has been proposed in inelastic neutron scattering experiments [9].

In the stripe scenario the dimensionality of the system is lower than two due to reduced hopping between the stripes. Therefore, the Fermi liquid picture for a quasi-particle is not valid anymore to describe the electronic structure of these materials, that is close to a one-dimensional Luttinger liquid or a marginal Fermi liquid [10]. To investigate these aspects we have measured the $k$ distribution of the electronic states near the Fermi level in an energy range of 50 meV, which is of the order of expected energy cutoff for the interactions involved in the superconducting pairing mechanism. This is done by an unconventional mode ARPES based on constant initial state angle scanning [11]. We have selected Bi2212 system at the optimum doping as representative material due to its good suitability for such measurements. Because there is not a two-dimensional Fermi surface for this strongly interacting electron gas, we call the measured constant energy contour marginal Fermi surface map. The resulting marginal Fermi surface (MFS) provides a clear identification to the key features related with the collective excitations in the high-$T_c$ superconductors. The measurements are combined with conventional mode of ARPES based on energy distribution curves (EDC) [12].

In this contribution, we discuss two main features of the MFS: (1) asymmetric suppression of spectral weight around the M points and (2) identification of one-dimensional set of electronic states. We argue that the asymmetric suppression of the spectral weight around the $M$ points is due to coupling of electrons with dynamical charge fluctuations along the diagonal direction with a wavevector $Q(-0.4\pi, 0.4\pi)$ whereas the one-dimensional set of states in the $(\pi, 0)$ direction might be at the origin of the dynamical incommensurate spin fluctuations [8].

2. EXPERIMENTAL

The ARPES measurements were carried out at the Laboratoire pour l'Utilisation du Rayonnement Electromagnétique (LURE) (Orsay-France) on the SU6 undulator beamline.
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A Bi2212 stoichiometric crystal ($T_c$ of 91 K with $\Delta T_c \sim 1$ K) of size 6 x 6 x 1 mm grown by floating zone method [13], was used for the measurements. We used the constant initial energy angle-scanning photoemission accompanied with the standard approach, exploiting the high intensity of the synchrotron radiation emitted by an undulator source. The experiments on the well-aligned and clean Bi2212 surface were performed in an ultra-high vacuum (UHV) chamber (base pressure $7 \times 10^{-11}$ mbar) equipped with an angle-resolving hemispherical analyzer and a high-precision manipulator permitting an azimuthal sample rotation ($\phi$) of 360° and polar emission angle relative to the surface normal ($\theta$) of 90° [14]. The angular resolution used for the MFS is 1.5°, whereas for the energy distribution curve (EDC) it is 1°. The spectrometer energy resolution was about 50 meV. The measurement were performed using the linearly polarized synchrotron light with photon energy of 32 eV in the even symmetry. In this geometry, the polarization vector of the synchrotron light, the wave vector of the emitted photoelectron and the surface normal were in the same horizontal plane (where the horizontal plane plays the role of a mirror plane) and the transition from the initial states with even symmetry (Cu 3d$_{xz}$-y$^2$, O 2p$_x$,$y$) is fully allowed [1,15]. The geometry has been used to have maximum emission intensity along the $(\pi,0)$ and $(0,\pi)$ directions. The experiments were repeated with different cleavage in several experimenta runs, and each cleave gave the same MFS map, proving the high quality of the crystal as well as reproducibility of the experimentally observed MFS features.

3. RESULTS

In Fig. 1 we plot the well-known and well-established band dispersion ($E(k)$) curves in the $\Gamma$-X and $\Gamma$-M directions obtained by plotting the energy positions of the dispersing peaks of EDC. Similar band dispersions on the Bi2212 system have been measured by many groups and interpreted in terms of dispersing quasi-particles and fitted with band structure models. The ratio of the width and the binding energy of the peaks (obtained by fitting the EDC curve with an asymmetric Gaussian function and a Fermi step function) is shown as insets. The ratio is close to 1 indicating the breakdown of the single particle picture.

The measured EDC are shown in Fig. 2 as a two-dimensional picture, where the intensity is plotted in a two-dimensional plane ($E$, $k$) [16]. In the picture, it could be seen that the EDCs are quite complex. The dispersing maxima (peaks) disappear for $k < k_m = 0.22\pi/a$ ($a = 5.4$ Å) in the $\Gamma$-X and $k < k_m = 0.2\pi/a$ ($a = 3.8$ Å) in the $\Gamma$-M direction and the energy dispersion for $k_m < k < k_F$ is of the order of 300 meV. Apart from a large peak width that is of the same order of magnitude as the total dispersion, the EDC curves show long tails extending up to about 0.6 eV energy below the chemical potential $E_F$. This kind of tail is observed in the one-dimensional ladder systems and interpreted in term of dispersion of spinons and holons [17]. The shape of these curves indicate directly that the electrons in the conduction band are strongly interacting.

Figure 3 shows the global view of the MFS of the Bi2212 superconductor. We now come directly to the point of missing segments of the MFS around the $M$ points. To have a better view of the suppression of the spectral weight, we enlarged the parts around the $M$ and $M_1$ points, and they are shown in Fig. 3. It is clear that the photointensity is suppressed asymmetrically [16]. The asymmetric topology of the missing segments could be seen in Fig. 4, which shows measured photointensity along azimuthal curvature from $X$ $(\pi,\pi)$ direction to $Y$ $(-\pi,\pi)$ direction across the M$(0,\pi)$ direction on the MFS. From this figure, it
is clear that the suppression of spectral weight around the $M$ points is asymmetric. Moving away from the $M$ direction to the $(-\pi, \pi)$ direction we find no evidence of a peak (the so-called quasi-particle peak), whereas along the $(\pi, \pi)$ direction we clearly observe such a peak, as shown in Fig. 4b.
Fig. 3. (a) Global view of the marginal Fermi surface of a Bi2212 superconductor at the optimum doping, measured by angle-scanning photoemission method at room temperature. The intensity of the emitted photoelectrons (excited from the initial state within an energy window, of order of the spectral resolution, centered at $E_F$) is given in gray scale with maximum corresponding to the white color. The wavevector of the electrons in the CuO$_2$ plane is given in units of $\pi/d$ ($d$ is Cu-O-Cu distance = 3.8 Å). We used the following notations; $\Gamma = (0,0)$, $M_1 = (\pi,0)$, $M = (0,\pi)$, $X = (\pi,\pi)$, $Y = (-\pi,\pi)$, where $\Gamma - M$ is along the Cu-O-Cu bond direction. Enlarged view of the suppression of the spectral weight around the $M$ point (panel b) and $M_1$ point (panel c).
After a careful analysis, we identified two well-defined wavevectors on the MFS connecting the points where a suppression of the spectral weight occurs. These points in the first Brillouin zone are $P_1(0.2\pi,0.8\pi)$, $P_2(0.8\pi,0.2\pi)$, $P_3(-0.8\pi,-0.2\pi)$, and $P_4(-0.2\pi,-0.8\pi)$. They are connected by the vectors $G(\pi,\pi)$ and $q(-0.4\pi,0.4\pi)$, as shown in Fig. 5. These vectors are the wavevectors of collective excitations that suppress the spectral weight of the quasi-particles at $P_n$. We notice that the wavevector $q(-0.4\pi,0.4\pi)$ is diagonal and is the second harmonic of the incommensurate and anharmonic lattice fluctuation; therefore, it can be associated with diagonal charge density waves CDW. However, the vector $G(\pi,\pi)$ is the antiferromagnetic wavevector and it is the wavevector of SDW that have been observed in the superconducting phase [18].

We now turn to the next observation. We identified a new set of electronic states with a one-dimension-like dispersion in the $\Gamma = (0,0)$ to $M_1 = (\pi,0)$, direction crossing the Fermi level at one point $k_F = 0.2 \pm 0.03\pi$ [19]. The observed electronic states are beyond the expected one electron-like Fermi surface, and we could identify these states by combining the angle-scanning mode with the conventional EDC mode. Figure 6 compares the energy distribution curves measured in the $\Gamma \rightarrow M_1(\pi,0)$ (solid line) and $\Gamma \rightarrow M(0,\pi)$ (dotted line) directions at the same $k$ locations. There are dispersing features in both directions;
however, the EDC in the two directions show clear differences in their line shapes. The main spectral band is clearly visible in both directions for polar angles above 5° off the \(\Gamma\) point and disperses toward the Fermi energy.

Although the main band appears quite similar in the two orthogonal directions, the EDC along \(M_1(\pi,0)\) shows a second dispersive spectral feature at lower angles with a smaller intensity. This feature crosses the Fermi level at around \(k_F = 0.2 \pm 0.03\pi\) with a total energy dispersion \(\Delta E \sim 80\) meV. However, we do not see this new set of states in the EDCs measured along the orthogonal \(\Gamma \rightarrow M(0,\pi)\) direction. The direct spectral differences are plotted in Fig. 6 (right panel), showing clearly the new set of electronic states. The second band, appearing only in one direction, was reproducibly observed in different runs performed on different cleaved surfaces ascertaining its intrinsic nature. Thus this result not only shows the anisotropy of the MFS, but also provides a direct evidence for a set of electronic states at

![Diagram](image_url)
the MFS having one-dimensional character along the Cu-O-Cu direction. The differences in the $M_1(\pi,0)$ and $M(0,\pi)$ were further ascertained by measuring photointensity at the MFS using scanning the polar angle along $\Gamma \rightarrow M(0,\pi)$ and $\Gamma \rightarrow M_1(0,\pi)$. The direct measurement of the photointensity scans at the MFS demonstrated that the observed band exists only along the $(\pi,0)$ direction, whereas it is absent along the $(0,\pi)$ direction [19].

4. DISCUSSION AND CONCLUSIONS

Having established experimentally the presence of a new feature on the MFS the question to be asked in on the origin of this feature. There are two points that should be considered here: (1) The energy dispersion of this new set of states ($\sim 80$ meV) is of the order of antiferromagnetic exchange interaction $J$ ($\sim 125$ meV); (2) There are incommensurate spin fluctuations along the Cu-O-Cu direction observed in several inelastic neutron scattering experiments [8,18], and the wavevector of the observed band ($\sim 0.2\pi$) is of the same order of magnitude if charge fluctuation occurs due to these incommensurate spin fluctuations in the CuO$_2$ plane. These facts encourage us to correlate the new states to the incommensurate spin fluctuations in the system.

We now turn to discuss the suppression of spectral weight around the $M$ points. The missing parts of the MFS around $(\pi,0)$ is one of the anomalous features that has been discussed widely during recent years. Preformed pairs without coherence, quasi-particle decay into spinons and holons [20], precursor effect of antiferromagnetic correlations [21] are some of the arguments that have been put forward to understand the suppression of the spectral weight. Pairing of quasi-particles (coherent) with a collective mode (incoherent) [22] could also be a reason for the missing segments around the $(\pi,0)$ points as argued by Shen and Shrieffer. We assigned the suppression of photointensity on the MFS to charge fluctuations associated with stripe ordering in the CuO$_2$ plane along the diagonal direction, i.e., $(\pi, \pi)$ direction. The wavevector of this CDW $q \sim 0.4\pi$ along the diagonal direction. Comparing this wavevector with the charge modulation in CuO$_2$ plane [4], measured by anomalous diffraction ($Q \sim 0.21\pi$) in the same system, we find that it is the second harmonic of the modulation that destroys the MFS around the $M$ points. It should be recalled that, due to strong contribution of second harmonic, the modulation in the Bi2212 system forms striped phase where the potential barrier between the stripes is controlled by the contribution of the second harmonic [4].

In summary, the combination of two modes of ARPES allowed us to identify several anomalous features on the MFS of the Bi2212 superconductor at the optimum doping. We have identified a wavevector around $M$ points along the diagonal direction. It is shown that the asymmetric suppression of spectral weight around the $M$ points is due to a charge ordering in stripes of the CuO$_2$ plane along the diagonal direction with a well-defined wavevector of $(-0.4\pi,0.4\pi)$. We have found a new set of electronic states along the $(\pi,0)$ direction with a small dispersion and small $k_F$ beyond the main MFS. The one-dimensional nature of these states has been confirmed by angle scanning of the MFS as well by recording energy distribution curves. We think that the new set of electronic states along the $(\pi,0)$ direction is related to the low-energy spin fluctuations observed in inelastic neutron scattering experiments. Thus the present results suggest the way to reconcile the magnetic fluctuations along the Cu-O-Cu direction observed in the inelastic magnetic scattering experiments [8,18] and charge ordering along the 45° from the Cu-O-Cu direction observed by EXAFS and
anomalous diffraction studies [3–7]. It should be mentioned that a simultaneous ordering of spin along the Cu-O-Cu direction [23] and charge along the Cu-Cu direction [24] has been found to take place in La2CuO4+x. In conclusion, we show direct evidence of stripes in the electronic structure of high-Tc superconductors. The experimental observations suggest that there are charge stripes along the (−π, π) direction with spin fluctuations along (π, 0) direction.

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11. The conventional method of Fermi surface measurements is based on measurement of energy distribution curves (EDC) in all high-symmetry directions of the Brillouin zone and find the locations of the Fermi surface by following the dispersion of peaks in the EDC (see, e.g., Ref. 1).


