The role of inter-plane interaction
in the electronic structure
of high $T_c$ cuprates

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Abstract

This thesis represents a systematic study of electronic structure of the modulation-free Pb-doped Bi2212 superconducting cuprates using the angle-resolved photoemission spectroscopy (ARPES), which is a leading technique in the experimental investigation of the single particle excitations in solids.

The results presented in this work indicate a very different origin for the observed complex spectral lineshape. Specifically, the peak-dip-hump lineshape can be easily understood in terms of the superposition of spectral features due to bilayer band splitting, namely the splitting of the CuO2 plane derived electronic structure into bonding and antibonding bands due to the interlayer coupling of CuO2 bilayer blocks within the unit cell of Bi2212. By performing experiments at synchrotron beamlines where the energy of the incoming photons can be tuned over a very broad range, the detailed matrix element energy dependence for both bonding and antibonding bands was determined. This gave the opportunity to study the electronic properties of these two bands separately.

For the first time, it was proved that the superconducting gap has the same value and symmetry for both bands. Furthermore, having recognized and sorted out the bilayer splitting effects, it became possible to identify more subtle effects hidden in the details of the ARPES lineshapes. In underdoped samples an "intrinsic" peak-dip-hump structure due to the interaction between electrons and a bosonic mode was observed. Studying the doping, temperature, and momentum dependence of the photoemission spectra it was established that: the mode has a characteristic energy of 38-40 meV and causes strong renormalization of the electronic structure only in the superconducting state; the electron-mode coupling is maximal around the (p,0) point in momentum space and is strongly doping dependent (being greatly enhanced in the underdoped regime). From the above, it was concluded that the bosonic mode must correspond to the sharp magnetic resonance mode observed in inelastic neutron scattering experiments, and that this coupling is relevant to superconductivity and the pairing mechanism in the cuprates.
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Chapter 1

Introduction
The electronic structure of solids determines the variety of many of their physical properties such as electrical, magnetic, optical, thermal behavior and etc. Moreover, the interactions of the electrons, in their turn, cause a lot of outstanding phenomena: metal-insulator transitions, colossal magneto-resistance and superconductivity. These electron correlation effects are especially strong in the so-called low-dimensional transition metal compounds. Thus, the study of the electronic states and electron interactions in such materials is of great importance and promises a deeper understanding of the physics of solids in general.

One of the most fascinating discoveries in solid state physics of the 20th century - the discovery of high-temperature superconductivity is based on low-dimensional copper oxide based compounds. A number of different spectroscopy techniques was used to study empty and occupied electronic states of these materials. But up to now the mechanism of high-temperature superconductivity in cuprates remains unclear. This requires further and more detailed investigations of their electronic structure.

This thesis deals with angle-resolved photoemission studies of high temperature superconductors. This experimental method provides us with the complete information about the occupied band structure and allows to systematically investigate the electronic structure of solids as a function of temperature, doping and momentum.

This thesis is organized as follows: an introduction to the high temperature superconducting cuprates and an overview of the previous photoemission results for Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ are given in the next chapters. The basics of the applied experimental techniques are discussed in chapter 4. The general framework of the experimental data presentation is also introduced in this chapter. The following chapters discuss the main results of this thesis: chapter 5 concerns the effects caused by the presence of bilayer split bands, chapter 6 is devoted to the properties of the quasiparticles in these bands. Finally, a short summary is given in chapter 7.
Chapter 2

Superconducting cuprates
Starting with the discovery of superconductivity in the La-Ba-Cu-O system near 30 K by Bednorz and Müller in 1986 [1], a new era in solid state physics has begun - that of high temperature superconductors (HTSC). As a result, a wide range of new superconducting materials with transition temperatures (T_c) higher than the boiling point of liquid nitrogen was found: starting from a T_c of about 90 K for YBa_2Cu_3O_7−δ (denoted Y123 or YBCO) [2] and Bi_2Sr_2CaCu_2O_{8+δ} (denoted Bi2212 or BSCCO) [3, 4], and ending with the record T_c=134 K for HgBa_2Ca_2Cu_3O_{8+δ} [5, 6] (see Fig. 2.1).

![Graph showing evolution of transition temperature T_c with time.](image)

Figure 2.1: Evolution of the transition temperature T_c with time.

### 2.1 Crystal structure

All these copper oxide based high T_c superconductors have a layered quasi two-dimensional (2D) crystal structure with CuO_2 planes separated from each other by so-called charge reservoir layers as shown in Fig. 2.2. Depending on the number N of CuO_2 planes within the unit cell, the cuprates are classified into single-layer [La_{2−x}Sr_xCuO_4, Bi_2Sr_2CuO_{6+δ}, (Sr,Ca)_2CuO_2Cl_2], bilayer [Bi_2Sr_2CaCu_2O_{8+δ}, YBa_2Cu_3O_{7−δ}],
2.2. Electronic structure

Figure 2.2: The crystal structures of the superconducting compound series of general formula Bi$_2$Sr$_2$Ca$_{n-1}$Cu$_n$O$_y$ with $n = 1$, 2 and 3.

trilayer materials [Bi$_2$Sr$_2$Ca$_2$Cu$_3$O$_{10+\delta}$], et cetera. This structural characteristic affects the superconducting properties: within each family of cuprates, $T_c$ increases with $N$ for $N \leq 3$, and then decreases. For instance, within the TI-based cuprate HTSCs, a maximum $T_c$ of 90, 110, 125 and 110 K is found for $N=1$, 2, 3 and 4, respectively [7, 8]. For a Bi-based cuprate HTSCs (Fig. 2.2), a maximum $T_c$ of 34, 90 and 110 K is found for $N=1$, 2 and 3, respectively [9].

### 2.2 Electronic structure

The key element shared by all such structures is the CuO$_2$ plane with an occupancy of one electron per unit cell forming a "Mott insulator", the parent compound for high $T_c$ superconductors. A Mott insulator is a material with a half filled band in which the conductivity vanishes, even while band theory would predict it to be metallic (for recent review see Ref. [10]).

A Mott insulator is fundamentally different from a conventional (band) insulator where conductivity is blocked by the Pauli exclusion principle. When the
highest occupied band contains two electrons per unit cell, electrons cannot move because all orbitals are filled. Instead, in a Mott insulator charge conduction is blocked by electron-electron Coulomb repulsion. When the highest occupied band contains one electron per unit cell, electron motion requires the creation of a doubly occupied site. If the electron-electron repulsion is strong enough, this band can split in two: the singly occupied lower band (the so-called lower Hubbard band) and the unoccupied upper one (the so-called upper Hubbard band). With one electron per site, the lower band will be full, the electron motion blocked and as a result the system will be an insulator. The amount of charge per unit cell becomes fixed, leaving only the electron spin on each site to fluctuate. This generates a super-exchange interaction, which favors antiparallel alignment of neighboring spins and leads to a long range antiferromagnetic (AF) order at a rather high Néel temperature $T_N$ depending on the material.

In the case of the high-$T_c$ cuprates, a layered two-dimensional crystal structure leads to highly 2D, anisotropic character, with a crystal field splitting of the $Cu3d$ bands as shown in Fig. 2.3.

![Figure 2.3: Formation of the electronic structure of the CuO$_2$ layers in HTSC. Left side: crystal-field splitting of a Cu$3d^9$ level in a regular and distorted octahedron of O ions. Right side: O2$p^6$ level. Middle: bands are formed by hybridization between Cu$3d$ and O2$p$ levels (from Ref. [11]).](image)

The Mott insulating phase of the HTSCs is realized in the Cu $d^9$ configuration, where three $t_{2g}$ bands as well as the $d_{3z^2-r^2}$ band are fully occupied, whereas the $d_{x^2-y^2}$ band is half filled. In this case, the low-energy electronic properties can be described by a single-band Hubbard model.

However, another significant feature of the transition-metal oxides is the strong hybridization of the $Cu3d$ and $O2p$ orbitals. In case of the high-$T_c$ cuprates, the
hybridization is strongest for the \( \sigma \) bonds between in-plane \( Cu3d_{x^2−y^2} \) and in-plane \( O2p_{x,y} \) states. This leads to a three-band extended Hubbard model [12] that includes the \( Cu3d_{x^2−y^2} \) orbital and two \( O2p_{x,y} \) orbitals in the plane. The energy gap is no longer determined by the Coulomb repulsion energy \( U \) as in the Mott-Hubbard (MH) insulator, but by the charge transfer energy \( \Delta = \epsilon_d - \epsilon_p \), which is defined as a difference between the energy of the \( Cu3d \) and \( O2p \) electrons. This kind of insulator, which was clarified by Zaanen, Sawatzky and Allen [13], is called a charge transfer (CT) insulator. The term "Mott insulator" is used here in a broad sense which covers both types. The difference between this two cases is schematically illustrated in Fig. 2.4.

![Diagram](image)

**Figure 2.4**: The Zaanen-Sawatzky-Allen scheme [13] of Mott insulators (left) together with a schematic illustration of energy levels for a Mott-Hubbard insulator (middle) and a charge-transfer insulator generated by the d-site Coulomb interaction effect (from Ref. [10]).

Therefore, undoped (with a half-filled \( Cu3d \) derived band) layered copper oxides are AF insulators and the Fermi level lies in the gap between the \( O2p \) band and UHB.

### 2.3 Hole and electron doping

By substituting different elements in the reservoir layers or by varying their oxygen content one can dope charge carriers (electrons or holes in the case of n- or p-type superconductors, respectively) into the \( CuO_2 \) planes. The electronic structure of charge carrier doped high-\( T_c \) cuprates can be related to the undoped half-filled case, as it shown in Fig. 2.5.
Figure 2.5: Model for the electronic structure of undoped, p-type doped and n-type doped $\text{CuO}_2$ plane, shaded area shows occupied states [11].

In case of the hole-doping the Fermi level is shifted into $O2p$ band (middle panel in Fig. 2.5), while when electrons are added to the undoped system (right panel in Fig. 2.5), the Fermi level will move into the upper Hubbard band ($d^{10}$), resulting in conductive properties. The energy separation between $O2p$ band and the UHB is in order of 2 eV.

Doping creates sites to which electrons can jump without experiencing the Coulomb repulsion energy: this makes the material conducting and, under the right conditions, superconducting. The phenomenological phase diagram of electron- (n-type) and hole-doped (p-type) cuprate HTSCs is presented in Fig. 2.6.

2.4 Phase diagram

Below, only the hole-doped part of the phase diagram will considered (right panel in Fig. 2.6). The antiferromagnetic (AF) region is the best understood region of the phase diagram. At zero dopant concentration cuprates are antiferromagnetic Mott insulators as it was discussed above.
Figure 2.6: Phase diagram of n (left) and p-type (right) superconductors [14].

When the dopant concentration is increased above a critical value (about 0.05), the antiferromagnetic state disappears. This region is the so-called pseudogap or underdoped region of the phase diagram. It is called underdoped because the dopant concentration is less than that one which maximizes the superconducting transition temperature (the so-called optimal doping level). The region where the dopant concentration is larger than the optimal one is called overdoped, correspondingly. The explanation of what a pseudogap is, will be given later.

The superconducting region is characterized by formation of coherent Cooper pairs (which leads to zero resistivity) and opening of a superconducting energy gap (the energy which is needed to split the pairs). However, in HTSCs the pairing symmetry (the symmetry of the wave function describing the Cooper pairs and, correspondingly, the symmetry of the superconducting gap) is different compared to conventional superconductors. It is well established that in conventional low-$T_c$ superconductors (e.g., Pb, Al, Nb, Nb$_3$Sn), the phonon mediated many-body electron-electron interaction leads to spin-singlet pairing with $s$-wave symmetry (see Ref. [15] for Bardeen, Cooper, Schrieffer (BCS) theory overview). On the other hand, the internal structure of the Cooper pairs in the HTSCs was a topic of intense debate for more than fifteen years after the discovery of high-$T_c$ supercon-
ductivity. Recently, a new class of phase-sensitive pairing symmetry experiments (such as SQUID interferometry, tricrystal and tetracrystal magnetometry), as well as a number of other techniques measuring the magnitude of the energy gap, have established predominantly $d$-wave pairing symmetry in a number of high-$T_c$ compounds (for review see Ref. [16]). As a result of a $d$-wave pairing symmetry, the superconducting gap function varies as a cosine function around the Fermi surface with nodes at $k_x = \pm k_y$ (see Fig. 2.7).

![Diagram of conventional and cuprate superconductors]

**Figure 2.7:** Symmetry of the order parameter of conventional (left panel) and cuprate (right panel) superconductors.

The puzzling feature found for underdoped materials is, that the gapped state persist up to a certain temperature $T^*$, which is higher than the superconducting transition temperature $T_c$. Up to now the nature of this so-called pseudogap is under debate [17, 18].

The relevant experimental observations related to the pseudogap properties can be summarized as follows.

- The pseudogap is observed in the underdoped side of the phase diagram and it weakens as optimal doping is approached.

- The pseudogap evolves into the superconducting gap (i.e. the magnitude of the superconducting gap $\Delta(T)$ is not going to zero at $T_c$).

- Both the pseudogap and superconducting gap have a $d$-wave symmetry.
• In the overdoped region of the phase diagram the pseudogap merges with the superconducting gap (i.e. $T^*$ merges with $T_c$).

Moreover these doped cuprate materials exhibit even more unusual properties in their "normal" metallic state (Fig. 2.6), not explainable in terms of conventional metal physics [19]. For example, the observed behavior of cuprates in the overdoped metallic regime can be well explained by a Fermi liquid (FL) theory, which describes electronic excitations in terms of a non-interacting gas of renormalized quasiparticles (see section 4.1.3). But if we consider the part of phase diagram between the underdoped (pseudogap) and overdoped (Fermi-liquid) regions, the applicability of FL theory becomes questionable. While the thermodynamic properties in this region are in fact similar to the FL behavior, all transport properties (such as resistivity, various magnetic relaxation rates, the Hall conductivity and magnetoresistance) and optical conductivity are characterized by unusual power laws as a function of temperature. These unusual properties are the reason why this part of the phase diagram is also called the non-Fermi-liquid region.

All these exceptional properties require new physical concepts to understand the high-$T_c$ cuprates [20]. Theoretical views of the underdoped (pseudogap) region of the phase diagram are just as diverse as the views on the mechanism of high-$T_c$ superconductivity [17]:

• "Pre-formed" pairs. Taking into account the fact, that the symmetry of the pseudogap is the same as of the superconducting state, it was assumed that the pseudogap phase results from the Cooper-pair formation at $T^*$. With decreasing temperature all these "pre-formed" pairs become coherent (occupy the $q=0$ state), which leads to a Bose condensation into the same quantum state at $T_c$ (i.e. superconductivity). Upon heating, phase coherence is lost at $T_c$ and the state just above the transition temperature is a normal Bose liquid of tightly bound pairs. At a much higher temperature $T^*$ the Cooper pairs dissociate.

• Spin-charge separation. Another approach is based on the assumption that spin and charge degrees of freedom associated with the holes become separated. Spin-charge separation creates holons with zero spin (bosons) and spinons which are zero charge $S = 1/2$ fermions. The pairing of spinons (formation of spin-singlets) leads to the formation of a "spin gap". The spin-
singlets are in a \(d\)-wave state, that, according to this model, explains the \(d\)-wave superconducting state at low temperatures. A similar model of the pseudogap has been developed also for dynamical charged stripes separated by insulating antiferromagnetic stripes. Spin and charge are separated as spin resides in the AF stripes, where a spin gap is opening.

- **Antiferromagnetic spin fluctuations.** In the nearly antiferromagnetic Fermi liquid (NAFL) model the dominant interaction between quasiparticles arises from spin fluctuations, as characterized in the dynamical spin susceptibility \(\chi(q, k)\). The strong AF correlations cause a peak in \(\chi(q, k)\) at \(Q = (\pi, \pi)\). This allows to distinguish two classes of quasiparticles: hot quasiparticles which are located near \((\pi, 0)\), connected to each other by \(Q\), and cold quasiparticles which are not strongly connected by AF fluctuations. The cold quasiparticles behave like a strongly coupled Fermi liquid while the hot quasiparticles are anomalous and non-FL like. Below \(T^\ast\) a gap opens in the hot quasiparticle spectrum, creating the pseudogap. Below \(T_c\) the cold quasiparticles become gaped, giving rise to the superconducting transition.

In order to check the validity of proposed theoretical models, a detailed comparison with experiments that probe the electronic properties and the elementary excitations is required. In this context, angle-resolved photoemission spectroscopy (ARPES) plays a major role because it is the most direct method to study the electronic structure of solids. This technique provides information about the single particle spectral function, which can be calculated starting from a microscopic Hamiltonian (see section 4.1.3).

Many of the ARPES results had strong impact on the understanding of the electronic structure of cuprate high temperature superconductors. In the following literature review, I will focus on the most important results obtained by ARPES such as the Fermi surface topology, the discovery of superconducting and pseudo gaps, the "peak-dip-hump" spectral lineshape at the \((\pi, 0)\) point and the recently observed bilayer splitting.
Chapter 3

Photoemission of high $T_c$ cuprates
Since the bilayer $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212) is object of the experimental research presented in this work, the following literature review will consider mainly ARPES results obtained for this compound. It also should be noted, that the vast majority of the published photoemission data were obtained using Bi2212 samples.

3.1 Fermi surface

The Fermi surface (FS) is a surface of constant energy in reciprocal space. It separates the unfilled states from the filled ones at zero temperature. The FS is one of the central concepts in solid state physics, since the electronic excitations across the Fermi surface determine the low energy properties of the material. Therefore, knowing the topology of the Fermi surface is very important.

In this work the following conventional square lattice notation for the 2D Brillouin zone of Bi2212 will be used: $\Gamma - M$ is along the Cu-O bond direction, with $\Gamma = (0,0)$, $M = (\pi,0)$, $X = (\pi,-\pi)$ and $Y = (\pi,\pi)$ in units of $1/a^*$, where $a^* = 3.83 \, \text{Å}$ is the separation between nearest-neighbor planar Cu ions (the orthorhombic $a$ axis is along $\Gamma - X$ and $b$ axis along $\Gamma - Y$).

Figure 3.1: Schematic view of a CuO$_2$ plane with Cu $d_{x^2-y^2}$ and O $p_x$, $p_y$ orbitals (left panel); and the 2D projected Brillouin zone with conventional notations for high-symmetry points, the shaded area represent the irreducible symmetry unit (right panel)
3.1.1 Fermi surface topology

An intensive study of the Fermi surface topology of Bi2212 has been carried out using the ARPES method. An ongoing controversy was present from the very beginning of these investigations [21, 22]. Some experimental results were supporting the electron-like character of the FS in Bi2212, while others, in contrary, reported a hole-like FS (see Fig. 3.2).

![Figure 3.2: Hole-like FS topology (a) versus electron-like topology (b) [23].](image)

This complexity in the interpretation of ARPES experiments arises from the detection of additional features besides those related to the primary electronic structure:

(i) **Shadow bands**, which correspond to a replica of the main FS shifted by the wave vector \((\pi, \pi)\) in the 2D Brillouin zone.

(ii) **Umklapp bands**, which, as usually believed, are originated from the diffraction of the outgoing photoelectrons by the superstructure present in the BiO layers. These bands can be as well a result of the modification of the intrinsic in-plane Cu-O electronic structure due to the incommensurate distortion of the approximate tetragonal symmetry.

The first complete FS mapping for near optimally doped Bi2212 done by Aebi et al. [24] is shown in Fig. 3.3. Panel (a) exhibits a plot of the spectral weight integrated within an energy window of 10 meV around the Fermi level (data were taken with HeI 21.2 eV photon energy and an energy resolution of 30-40 meV). The authors concluded that the FS is a hole like piece centered at \(X\) or \(Y\) points in
Figure 3.3: (a) $k_\parallel$ mapping of the intensity of HeI (21.2 eV) excited photoelectrons collected within an energy window of 10 meV width centered at $E_F$. A logarithmic intensity scale is used to enhance weaker features. The outer circle indicated an emission angle of 90°.  (b) Sketch of (a), emphasizing the fine lines observed in the measurements and distinguishing between the stronger (thick lines) and weaker (dashed lines) sets of lines. (from Ref. [24])

the $k$-space as indicated by the thick lines in Fig. 3.3b. The complications in the intensity map arise from the superposition of "shadow" FSs that are offset from the main FS by $Q = (\pi, \pi)$, as shown in Fig. 3.3b. Note, that antiferromagnetic (AF) ordering of the spins in the CuO$_2$ plane leads to the shift of the "antiferromagnetic" FS of the vector $(\pi, \pi)$. Since the vector $Q$ corresponds exactly to the vector $(\pi, \pi)$, this "shadow" FS can be explained in terms of short range and dynamic AF correlations.

Later on, Ding et al. [25] have investigated the electronic structure and the Fermi surface in the normal state ($T = 95K$) of near-optimal OD Bi2212 ($T_c = 87K$) using incident photon energies of 19 and 22 eV.

They found that the main FS consists of the large hole-like barrels centered around the $X$ and $Y$ points in $k$-space (thick line in Fig. 3.4). Additional FS crossing bands can be modelled by shifting the main band fit by $\pm q$, where $q = (0.21\pi, 0.21\pi)$ is the superlattice (SL) vector arising from the presence of the superstructure in the Bi-O layers (thin curves in Fig. 3.4). This distortion is
characterized by a periodicity of 27 Å along the $b$ axis and results in a unit cell approximately five times larger along this direction. The shadow bands corresponding to the shift of the main FS on vector $Q = (\pi, \pi)$ were also observed. As a result, around $(\pi, 0)$ two main bands, two shadow bands, and four umklapp bands cross the Fermi level.

Recent improvements in energy and angular resolution of ARPES experiments make it possible to resolve all these features in photoemission spectra from Bi2212. Main, shadow and umklapp bands can be absolutely precisely recognized from the series of spectra taken along the $\Gamma - Y$ direction, where they are clearly separated (see Fig. 3.5).

Moreover, Borisenko et al. [27] have shown, that using the superstructure free \( \text{Pb} \)-substituted Bi2212 (see section samples in chapter 4) one can avoid the umklapp bands in photoemission spectra (Fig. 3.6).

While in the case of the pristine Bi2212 the presence of diffraction replicas of the main FS leads to a drastic increase of intensity along a ribbon centered on

![Figure 3.4: Bi2212 \((T_c=87K)\) FS (a) and normal state \((T=95K)\) dispersion (b). The thick lines are the result of a tight binding fit to the main band dispersion. Thin and dashed lines represent \((0.21\pi, 0.21\pi)\) umklapp and shadow bands, respectively. The inset of (b) is a blowup of \(\Gamma - X\). (from Ref. [25])](image-url)
Figure 3.5: Intensity $I(\mathbf{k}, \omega)$ (a) and corresponding ARPES spectra (b) from optimally doped Bi2212 ($T_c = 90K$) taken along $\Gamma - Y$ with 33 eV photons at $T = 40K$. Main, umklapp, second-order umklapp, and shadow bands are denoted as MB, UB, UB(2), and SB, respectively. (from Ref. [26])

the $(0, -\pi) - (\pi, 0)$ line indicated in Fig. 3.6a by grey shading, in the case of the Pb-doped Bi2212 there is no such intensity ribbon, and the intensity profile across the map is practically symmetrical about the $\Gamma - M - Z$ line (see Fig. 3.6b). The main FS presented here reveals hole-like topology, seen here as a cross-section of tube-like forms centered around the $X, Y$ points (the so-called FS "barrels").

A detailed and systematic investigation of the doping dependence of the normal-state ($T = 300K$) Fermi surface topology for the modulation free Pb-doped Bi2212 samples was done by Kordyuk et al. [28]. In contrast with the case of LSCO, where a crossover from a hole- (see Fig. 3.2a) to electron-like (see Fig. 3.2b) FS is clearly evident near optimal doping [29], within the studied doping range (35K around optimal doping) no topological change of the main FS was found. The Fermi surface remains hole-like, centered at the $X, Y$ points (see Fig. 3.7).
Figure 3.6: Normal state \(T = 120\text{K}\) Fermi surface maps of (a) pristine Bi2212 and (b) Pb-doped Bi2212 taken with unpolarized He\(\alpha\) radiation. The guidelines indicate the main FS (thick black solid line); shadow FS (solid red line); first-order umklapp FS (thin solid black line), second-order umklapp FS (thin dashed black line). (from Ref. [27])

Figure 3.7: Normal state Fermi surface for a series of Pb-Bi2212 samples with different doping levels as indicated by the \(T_c\) value of each sample. The sketch shows the FS for the OD69K data set (Ref. [28]).
As hole doping is increased, the main FS "barrels" increase in size as can easily be seen in the decrease of the inter-barrel separation around the $M = (\pi, 0)$ point. The shape of the FS "barrels" changes from being quite rounded at low doping to taking on the form of a square with well-rounded corners at higher doping. In contrast to the previous results of Ding et al. [30], where no shadow band were detected for a strongly underdoped Bi2212 sample ($T_c = 15$K), the strongly pronounced shadow Fermi surface (SFS) was observed for all doping levels.

Furthermore, by fitting the Fermi surface to the experimentally obtained maps (a sketch of the fit result is shown as the yellow line on the right-hand side of Fig. 3.7) one can derive the hole concentration $x$ from the simple relation $x + 1 = 2S_b/S_{BZ}$, where $S_b$ is the area of main FS "barrel" and $S_{BZ}$ is the area of the Brillouin zone. The results obtained from this analysis of the FS area are shown in Fig. 3.8. The solid line shows the widely used empirical parabola for $T_c$ as a function of doping [31].

![Figure 3.8: Symbols: critical temperatures vs the hole concentration $x_{FS}$, calculated directly from the area of the FS's shown in Fig. 3.7. The solid line shows the commonly used empirical relation for $T_c$ vs $x$ (Ref. [28]).](image)

The coordinate pairs ($T_c, x$) directly derived from the experimentally determined FS area also give a parabolic curve (shown as a dotted line in Fig. 3.8), but this curve is downshifted in doping by $\sim 0.05$ towards the underdoped side of the phase diagram. This result can be well understood in terms of the bilayer splitting of the CuO band.

### 3.1.2 Bilayer splitting

The presence of the two CuO$_2$ planes in a unit cell of Bi2212 gives grounds to expect a hybridization of these electronic states into bonding (BB) and antibonding
(AB) bands, the wavefunctions of which are even and odd under reflection in a mirror plane located in the middle of the unit cell parallel to the \( \text{CuO}_2 \) layers, correspondingly.

This band splitting, the so-called \textit{bilayer splitting}, was predicted from a variety of band structure calculations \cite{32, 33}. The general dispersion characteristics of these two bands can be understood in terms of a simple tight-binding model. In a two-layer system like Bi2212, the single-particle hopping between the planes of a bilayer as a function of the in-plane momentum \( \mathbf{k}_\parallel \) can be written as follows \cite{34}:
\[
t_\perp (\mathbf{k}) = -t_z (\cos k_x - \cos k_y)^2.
\]
Therefore, the two bilayer bands are degenerate along the Brillouin zone diagonal \( \Gamma - X \) but become split as one goes along the \( \Gamma - M \) direction. The value of bilayer splitting reaches its maximum at the \( M = (\pi, 0) \) point (see Fig. 3.9).

Figure 3.9: \textit{Band structure of Bi2212 in the vicinity of the Fermi energy \( E_F \) (shown by the horizontal dashed line) along the symmetry lines \( \Gamma - M - X - \Gamma \). Bands noted with }a\textit{ and }b\textit{ are the antibonding and bonding states at the }M\textit{-point, respectively, arising from the }CuO_2\textit{ planes; }c\textit{ and }c'\textit{ are Bi-O related states (Ref. }35\text{).}

In the early work of Dessau \textit{et al.} \cite{22}, two FSs in the \( (\pi, 0) \) region were observed in the normal state \( (T=100\text{K}) \) for pristine Bi2212 \( (T_c = 85\text{K}) \): one electron-like and one hole-like. This was interpreted as a signature of bonding and antibonding bands in a bilayer system. On the other hand, Ding \textit{et al.} \cite{25} claimed from the photoemission studies of the band dispersion the absence of bilayer splitting in the pristine Bi2212 \( (T_c=87\text{K}) \) in both the normal \( (T=95\text{K}) \) and superconducting \( (T=13\text{K}) \) states.
The recent improvement in angular (i.e. momentum) resolution of the electron energy analyzers (for further details see the experimental section in chapter 4) makes it possible to resolve this band splitting in ARPES experiments on Bi2212 and the bilayer splitting was experimentally observed in the heavily overdoped Bi2212 \( (T_c = 65K) \) [36].

In Fig. 3.10a two main FSs are observed (one for the AB and the other for the BB) as well as its corresponding superstructure replicas (AB' and BB'). This FSs give rise to four resolved peaks (Fig. 3.10b), whose dispersion can be followed in the ARPES spectra along the \( \mathbf{k} \)-space cut indicated by the arrow in Fig. 3.10a. A different photon energy dependence of bonding and antibonding bands was observed. At 22.7 eV photon energy (lower right half of Fig. 3.10a), the AB has

![Diagram](image)

**Figure 3.10:** (a) Integrated \( E_F \) intensity map (integration window [-20 meV, 10 meV]) from overdoped Bi2212 \( (T_c=65K) \). The data were measured above \( T_c \) with \( h\nu = 22.7 \) eV (lower-right, \( T = 75 \) K) and \( h\nu = 20 \) eV (upper-left, \( T = 80 \) K). Symbols are the FS crossings estimated directly from the dispersion of the peaks. BB and AB refer to the bonding and antibonding bands, and BB' and AB' to the corresponding superstructure replicas. (b) ARPES spectra along the cut indicated by the arrow in (a) (Ref. [36]).
more weight near $E_F$ than the BB, and this situation is reversed at 20 eV (upper left half of Fig. 3.10a). This strong photon energy dependence of the relative intensities of the AB and BB is consistent with the bilayer splitting, because the antibonding and bonding bands have different symmetry along the $c$-axis (odd and even for AB and BB, respectively) and, therefore, different transition matrix elements. It was found [35, 37], that these matrix element effects cause a nontrivial excitation photon energy dependence of the relative ARPES intensities of the bonding and antibonding states (Fig. 3.11).

Similar results were independently obtained by Chuang et al. [38] for the overdoped Bi2212 ($T_c$=85K). In Fig. 3.12a well separated antibonding (A) and bonding (B) bands can be observed together with the superstructure (S.S.) bands. All together, both studies provide a clear evidence for normal state bilayer splitting in overdoped Bi2212. The presence of bilayer splitting was also confirmed for the slightly overdoped ($T_c$=84K) and overdoped ($T_c$=70K) modulation free Pb-doped Bi2212 [39] as well as for the underdoped ($T_c$=77K) Pb-doped Bi2212 [28].

3.2 Superconducting state properties

All the results discussed in the previous section concern to the normal state electronic properties of the superconducting layered cuprates. However, to understand the mechanism responsible for the occurrence of high temperature superconductivity in these materials a systematic investigation of the electronic structure below the superconducting transition temperature $T_c$ is required.
Figure 3.12: (a) normal state (T=100K) E, k image plot of the ARPES intensity and (b) corresponding MDCs taken on overdoped Bi2212 (Tc=85K) near the (π,0) region (Ref. [38]).

3.2.1 Superconducting gap

One of the most important results obtained in photoemission experiments was the direct observation of the superconducting energy gap [40].

In fact, the ability of ARPES to measure the superconducting gap as a function of crystal momentum provides a well suited opportunity to probe the symmetry of the order parameter. Earlier investigations clearly demonstrated the anisotropic character of the superconducting gap [41, 42].

Fig. 3.14 presents normal (open squares) and superconducting state (solid circles) ARPES spectra from overdoped Bi2212 at two different points A and B in the Brillouin zone. Very clear spectral changes were observed at the A point as the sample is cooled below Tc. The leading edge of the spectrum in the superconducting state is shifted back to higher binding energy, reflecting the opening of the superconducting energy gap. At the same time, a "dip" near -80 meV appears in the spectra. These changes in the spectral lineshape will be discussed later in the next section of this chapter. At the B point, only minor changes with temperature were observed, indicating that the gap is very small and undetectable within the
3.2. Superconducting state properties

experimental uncertainty or equal zero. The energy shift of the midpoint of the leading edge of the superconducting state spectrum gives a gap value of 12 meV at point A and value of 0 to 0.5 meV at point B. These results strongly suggest that the superconducting gap is anisotropic and, in particular, consistent with a d-wave order parameter described within theories where short-ranged antiferromagnetic spin fluctuations mediate pairing in the cuprates (for review see Ref. [43]).

A more detailed investigation of the momentum dependence of the superconducting gap in Bi2212 was done by Ding et al. [44]. The values of the gap were obtained by means of fitting the photoemission spectra taken in the superconducting state (T = 13K) for various points along the main FS in the Y quadrant (see inset in Fig. 3.15) with a resolution broadened BSC spectral function.

![Photoemission spectra](image)

| Figure 3.13: (a) and (b) Photoemission spectra of Bi2212 (Tc = 88K) samples (S1, S2) in the normal (dots) and superconducting (stars) states in comparison with (c) BSC model calculation of the density of states (DOS). The normal state DOS for T = 105K has been simulated by the assumption of the linear decreasing DOS convoluted with a resolution Gaussian and multiplied by the Fermi function. For the DOS in the superconducting state the normal state DOS was multiplied by the BSC function, convoluted with the resolution Gaussian and multiplied by the Fermi function. The superconducting gap Δ was varied to get a satisfactory agreement with the experimentally observed spectrum. The corresponding value of Δ = 30 meV was obtained (Ref. [40]). |
Figure 3.14: Photoemission spectra from a Bi2212 ($T_c=78$K) sample recorded at k-space locations A and B, as illustrated in the inset. The spectral changes above and below $T_c$ are caused by the opening of the superconducting gap (Ref. [42]).

Figure 3.15: Superconducting gap measured at 13K on Bi2212 ($T_c=87$K) plotted versus angle along the normal state Fermi surface (filled circles) together with a d-wave fit to the data (solid curve). The inset shows locations of the corresponding spectra in the Y quadrant of the Brillouin zone as well as the photon polarization direction (Ref. [44]).
The angular variation of the gap value obtained from the fits is in a good agreement with a $d_{x^2-y^2}$ functional form $\Delta(k) = \Delta_0(\cos k_x - \cos k_y)$, which gives zero value of the gap at the FS points corresponding to the $\Gamma - X(Y)$ direction (the so-called nodal points). Moreover, the gap reaches its maximum at the FS points corresponding to the $M - X(Y)$ direction (the so-called antinodal points).

However, in most cases, instead of the well defined cusp seen in Fig. 3.15 an extended area around the nodal region characterized by gapless excitations was found [44, 45].

The doping dependence of the superconducting gap anisotropy is shown in Fig. 3.16. For the best fit the following gap function has been used: $\Delta_k = \Delta_{\max}[B \cos 2\phi + (1 - B) \cos 6\phi]$ with $0 \leq B \leq 1$. The additional $\cos 6\phi$ term corresponds to the next harmonic consistent with $d$-wave symmetry. In general, the symmetry allowed gap function in case of $d_{x^2-y^2}$-wave pairing in the square

![Figure 3.16: Values of the superconducting gap as a function of the Fermi surface angle $\phi$ obtained for a series of Bi2212 samples with varying doping. The solid lines represent the best fit (see text). The dashed line in the panel of an UD75K sample represents the gap function with $B = 1$ (Ref. [45]).](image-url)
lattice has a multi-term form [46]:

\[
\Delta_k^d = \sum_{r_1=1}^{\infty} \sum_{r_2=0}^{\infty} \sum_{r_3=0}^{\infty} a_{r_1 r_2 r_3}^d (\cos(k_x r_1) \cos(k_y r_2) - \cos(k_x r_2) \cos(k_y r_1)) \cos(k_z r_3)
\]

Thus, the first harmonic \(\Delta^{100} \propto (\cos k_x - \cos k_y)\) corresponds to the \(\cos 2\phi\) term in the fit and the second harmonic \(\Delta^{200} \propto (\cos 2k_x - \cos 2k_y)\) to \(\cos 6\phi\), correspondingly. The results of the fit (Fig. 3.16) show that while for the overdoped samples the data sets are consistent with \(B \approx 1\), the parameter \(B\) decreases significantly in the underdoped regime.

Note, that a perfectly defined \(d\)-wave behavior would suggest that one can exclude the presence of two independent gaps with different symmetry of different origin, namely the superconducting gap and the normal-state \(\text{pseudo gap}\). Nevertheless, it should be stressed here that ARPES does not see the phase of the order parameter (wave-function describing the Cooper pair). In other words, angle-resolved photoemission measures the \(k\)-dependence of the magnitude of the gap only.

### 3.2.2 Peak-dip-hump

The other well-known effect observed for Bi2212 in the early photoemission experiments is a dramatic change of the spectral lineshape at the \(M = (\pi, 0)\)-point below the superconducting transition temperature [41].

For temperatures \(T < T_c\), a sharp peak develops at the lowest binding energies followed by a dip and a broader hump, giving rise to the so-called "peak-dip-hump" structure (see Figs. 3.14 and 3.17).

Similar results were obtained previously in tunneling measurements on Bi2212, where "peak-dip-hump"-like tunneling spectra were observed at temperatures below \(T_c\) [47].

The detailed investigation by Norman et al. [48] shows a more complex \(k\)-dependence of this peak-dip-hump lineshape. In Fig. 3.18 the photoemission spectra along the \(\Gamma - M - Z\) direction are shown for an overdoped \((T_c = 87K)\) Bi2212 sample above (a) and below \(T_c\) (b). In the superconducting state almost no dispersion was observed for the low energy peak along a large range in \(k\)-space, while the dispersion of the hump in the superconducting state follows the normal state dispersion.

The simplest explanation of the observed spectral lineshape would be the presence of two bands due to the bilayer splitting discussed above. In this case, the
antibonding band could be responsible for the peak and the bonding band for the hump. However, at the time, there were some experimental observation that seem to contradict this simple picture.

Firstly, if the sharp peak is associated with an antibonding band, then this band should also appear at temperatures above $T_c$. But in a variety of temperature dependent studies the disappearance of the peak above the superconducting temperature was seen [49, 50]. In a strict sense, the peak-dip-hump structure becomes apparent slightly above $T_c$, but the low energy peak achieves considerable spectral weight only below $T_c$ (see Fig. 3.19).

Secondly, if the peak and the hump were from two different bands, then their intensities should have different matrix element dependence. But, as was found by Ding et al. [25], the relative intensities of both features do not depend on the angle of the photon polarization (see Fig. 3.20).

Therefore, it was concluded that the peak-dip-hump lineshape might also be an intrinsic feature. Thus, the dip, the peak and the hump would al bel part of a single spectral function, caused by a strong energy dependence of the superconducting state self-energy (a "strong coupling to bosons"). The hump represents the energy scale at which the spectral function below $T_c$ matches onto that in the normal state (see middle and left column in Fig. 3.18). However, the existence of the dip requires an additional structure in the self-energy below $T_c$. For example, this
Figure 3.18: ARPES spectra in (a) the normal state and (b) the superconducting state along the line $\Gamma - M - Z$, and (c) the superconducting state (13K) along the line $M - Y$ for an overdoped ($T_c=87K$) Bi2212 sample. The inset in (c) shows the assumed Fermi surface (curved line) together with the positions of the presented spectra (open circles) in the Brillouin zone (Ref. [48]).

structure can be understood in terms of electrons interacting with an energetically sharp bosonic mode below $T_c$ [48, 51, 52]. Since the dip structure appears at the $(\pi, 0)$ point, and since the $(\pi, 0)$ point can be mapped onto symmetry equivalent $(\pi, 0)$ points using a $(\pi, \pi)$ vector, it was speculated that this mode could be the resonant spin fluctuation mode observed in inelastic neutron scattering (INS).
experiments on Bi2212 [53]. The characteristics of the magnetic resonance can be summarized as follows [54]: the resonance is narrow in energy, with the energy width smaller than the instrumental resolution (typically less than 10 meV). The resonance lies below a gapped continuum with the mode energy being about 40 meV for optimal doping. The resonance is strongly peaked at the $(\pi, \pi)$ wavevector. The momentum width of the spin fluctuation spectrum is minimal at the resonance energy. With underdoping the intensity at $\vec{Q} = (\pi, \pi)$ increases. On approaching $T_c$ from below, the resonance energy does not change, but its intensity decreases towards $T_c$.

As a result, the "peak-dip-hump" spectral lineshape could be both due to the presence of bilayer split bands or/and interaction with the bosonic mode.

### 3.3 Remaining problems

In previous photoemission studies on Bi2212 it was shown that in a wide range of doping the normal state Fermi surface has a hole-like topology centered around
Figure 3.20: Low temperature ($T = 13$K) photoemission spectra of Bi2212 at the $M$-point for various incident photon angles. The solid (dashed) line is $18^\circ$ ($85^\circ$) from the normal. The inset shows the height of the sharp peak for data normalized to the broad peak at different incident angles (Ref. [25]).

$X(Y)$-point in the Brillouin zone. The complexity of the electronic structure of pristine Bi2212 arises from the co-existence of shadow and superstructure umklapp bands. The Pb substitution in Bi2212 leads to the removal of the superstructure-effects in the photoemission spectra. Two bilayer split bands formed due to the presence of two $CuO$ layers per unit cell were observed in pristine and Pb-doped Bi2212. However, matrix elements as a function of the photon energy and polarization have a strong influence on the ARPES intensity of the bilayer split bands. Therefore, a detailed and systematic examination of this effect is required in order to achieve a complete understanding of the quasiparticle selfenergy in these materials.

In the superconducting state, the presence of a strongly anisotropic superconducting gap was confirmed by ARPES measurements. The observed momentum dependence supports the $d$-wave symmetry of the gap. However, due to the presence of the bilayer splitting a further investigation of the single band gap symmetry is necessary.

The photoemission lineshape in the vicinity of the $(\pi, 0)$ point below the trans-
ition temperature was characterized by a sharp peak, followed at higher energy by a dip and a hump. The origin of this "peak-dip-hump" structure is still not well understood. One of the possible interpretations of an intrinsic "peak-dip-hump" is based on the interaction of quasiparticles with a collective bosonic mode. The detailed inspection of the properties of this mode, such as its energy, momentum, temperature and doping dependence will allow to identify the mode character, and, therefore, help to clarify the origin of superconductivity in high-temperature cuprates.
Chapter 4

Angle-resolved photoemission spectroscopy
In this chapter the experimental method of angle-resolved photoemission spectroscopy (ARPES) \[55\], which has been applied to probe the occupied electronic structure of Bi2212 is presented. In the following pages the principles of ARPES are explained and the equipment used is presented. The closing section of this chapter is devoted to a description of the measurement and data analysis principles.

4.1 Photoemission principles

4.1.1 Basic photoemission principles

The principle of a photoemission experiment is sketched in Fig. 4.1. Photons of energy \( h\nu \) impinge on the sample and create photoelectrons which leave the sample.

![Diagram of ARPES experiment](image)

**Figure 4.1:** Geometry of an ARPES experiment: the emission direction of the photoelectron is specified by the polar \((\Theta)\) and azimuthal \((\phi)\) angles.

By collecting the photoelectrons with an electron energy analyzer characterized
by a finite acceptance angle, one can measure the number of photoelectrons per time interval and solid angle (i.e. their intensity) as a function of their kinetic energy $E_{\text{kin}}$ and a given emission angle.

In this way, the photoelectron momentum $\mathbf{p}$ is completely determined: its modulus is given by $|\mathbf{p}| = \sqrt{2mE_{\text{kin}}}$ and its components parallel and perpendicular to the sample surface are obtained from the polar ($\theta$) and azimuthal ($\phi$) emission angles.

Taking advantage of the total energy and momentum conservation laws, the kinetic energy and momentum of the photoelectron can be related to the binding energy $E_B$ and crystal-momentum $\hbar \mathbf{k}_\parallel$ inside the solid:

$$E_{\text{kin}} = h\nu - \Phi - |E_B|$$

(4.1)

$$\mathbf{p}_\parallel = \hbar \mathbf{k}_\parallel = \sqrt{2mE_{\text{kin}}} \cdot \sin \theta \left( \cos \phi \mathbf{n}_x + \sin \phi \mathbf{n}_y \right)$$

(4.2)

Here $\Phi$ is a work function, reflecting the fact that the photoelectron escaping into the vacuum from the sample needs some additional energy to overcome the surface barrier; $\mathbf{n}_x$ and $\mathbf{n}_y$ are unit vectors which span the sample surface plane (see Fig. 4.1); $\hbar \mathbf{k}_\parallel$ is the component of the electron crystal momentum parallel to the sample surface in the extended zone scheme.

Upon going to large $\theta$ angles one actually probes electrons with $\mathbf{k}$ lying in higher order Brillouin zones. By subtracting the corresponding reciprocal lattice vector $\mathbf{G}$ the reduced electron crystal momentum in the first Brillouin zone can be obtained. It is important to note that the perpendicular component of the wave vector is not conserved across the sample surface due to the lack of translational symmetry along the surface normal. In the case of layered compounds like the copper oxide superconductors, however, which are extremely two dimensional, the electronic dispersion is almost exclusively determined by $\mathbf{k}_\parallel$.

### 4.1.2 Three-step model and sudden approximation

A more rigorous approach to describe the photoemission process is to proceed with the so-called one-step model in which photon absorption, electron removal and electron detection are treated as a single coherent process (overview of one-step model see in [56]). In this case bulk, surface and vacuum have to be included in the Hamiltonian describing the crystal, which implies that not only bulk states have to be considered, but also surface states, and surface resonances.
However, due to the complexity of the one-step model, photoemission data are usually discussed within the *three-step model* [55, 14]. Within this approach, the photoemission process is subdivided into three independent and subsequent steps:

- Optical excitation of the electron in the bulk.
- Travel of the excited electron to the surface.
- Escape of the photoelectron into vacuum.

The total photoemission intensity is given by the product of three independent terms: the total probability for the optical transition, the scattering probability for the travelling electrons, and the probability of the transition through the surface potential barrier. Step (i) contains all the information about the intrinsic electronic structure of the material and will be discussed in detail below. Step (ii) can be described in terms of an effective mean free path, proportional to the probability that the excited electron will reach the surface without scattering (i.e. with no change in energy and momentum). The inelastic scattering processes which determine the surface sensitivity of photoemission, also give rise to a continuous background in the spectra which is usually ignored or subtracted. Step (iii) is represented by a probability of transition through the surface, which is unity if the excited electron has sufficient energy to overcome the work function $\Phi$ and escape into vacuum, and zero otherwise.

To evaluate the photocurrent produced in the photoemission experiment one has to calculate the transition probability $w_{if}$ for an optical excitation between the N-electron ground state $\Psi_i^N$ and a final state $\Psi_f^N$. This can be approximated by Fermi’s golden rule:

$$w_{if} = \frac{2\pi}{\hbar} \left| \langle \Psi_f^N | H_{int} | \Psi_i^N \rangle \right|^2 \cdot \delta(E_f^N - E_i^N - h\nu)$$

(4.3)

where $E_i^N = E_i^{N-1} - E_B^k$ and $E_f^N = E_f^{N-1} + E_{kin}$ are the initial and final-state energies of the N-particle system, $E_B^k$ is the binding energy of the photoelectron with kinetic energy $E_{kin}$ and momentum $\mathbf{k}$. The interaction with the photon can be considered as a perturbation given by:

$$H_{int} = \frac{e}{2mc} (\mathbf{A} \cdot \mathbf{p} + \mathbf{p} \cdot \mathbf{A}) = \frac{e}{mc} \mathbf{A} \cdot \mathbf{p}$$

(4.4)

where $\mathbf{A}$ is the electromagnetic vector potential, $\mathbf{p}$ is the electronic momentum operator. Here the dipole approximation was used together with the commutator
\[ |p, A| = i\hbar \nabla \cdot A. \] Above the Drude plasma frequency, which is true for ultraviolet photons, the light penetrates deeply into the solid, making this term small (since \( A \) is constant over atomic dimensions and therefore \( \nabla \cdot A = 0 \)).

The next assumption is to divide the initial and final state wave function into a photoelectron and remaining \((N - 1)\)-electron term. This, however, is not an easy task, especially in the case of strong electron-electron correlations. One additional difficulty is the process of the relaxation of the system due to the interaction of the remaining electrons with the photoelectron and the photohole. The problem is simplified within the so-called sudden approximation, which is extensively used in many-body calculations of photoemission spectra. In this approximation, the removal of the photoelectron is sudden and relaxation effects on the initial wave function can be ignored.

Therefore the wave within the sudden approximation could be written as:

\[
\Psi_i^N = A \phi_i^k \Psi_i^{N-1} \quad \Psi_f^N = A \phi_f^k \Psi_f^{N-1} \tag{4.5}
\]

where \( \phi_i^k \) and \( \phi_f^k \) are the wave functions of the (photo)electron with momentum \( k \).
before and after optical transition, $\Psi_i^{N-1}$ and $\Psi_i^N$ describe the remaining $(N-1)$-electron system, and $\mathcal{A}$ is an antisymmetric operator that properly antisymmetrizes the $N$-electron wave function so that the Pauli principle is satisfied.

The matrix elements in Eq. 4.3 can be written as:

$$\langle \Psi_j^N \rvert H_{\text{int}} \rvert \Psi_i^N \rangle = \langle \phi_j^k \rvert H_{\text{int}} \rvert \phi_i^k \rangle \langle \Psi_i^{N-1} \rvert \Psi_i^{N-1} \rangle$$  \hspace{1cm} (4.6)

where $M_{f,i}^k \equiv \langle \phi_j^k \rvert H_{\text{int}} \rvert \phi_i^k \rangle$ is the one-electron dipole matrix element and the second term is the $(N - 1)$-electron overlap integral.

The total photoemission intensity measured at a fixed momentum $k$ as a function of kinetic energy $E_{\text{kin}}$ is proportional to:

$$I(k, E_{\text{kin}}) \propto \sum_{f,i} |M_{f,i}^k|^2 \sum_m |c_{m,i}|^2 \delta(E_{\text{kin}} + E_{m}^{N-1} - E_i^{N} - h\nu)$$  \hspace{1cm} (4.7)

where $|c_{m,i}|^2 = |\langle \Psi_{m}^{N-1} \rvert \Psi_i^{N-1} \rangle|^2$ is the probability that the removal of the electron from state $\Psi_i^{N-1}$ will leave the $(N - 1)$-particle system in the excited state $\Psi_{m}^{N-1}$.

In strongly correlated systems many of the $|c_{m,i}|^2$ will be different from zero. Therefore, the ARPES spectra will not consist of single delta functions but will be described by a non-trivial function of both energy and momentum.

### 4.1.3 Spectral function and self-energy

The discussion above is well suited for atoms and molecules. But it cannot similarly well be applied to solids. Therefore, for a many-body system the Green’s function formalism has been developed. The propagation of a single electron is then described by the one-electron Green’s function $G(r,t)$. By taking a Fourier transform, it can be expressed in energy-momentum representation resulting in $G(k,\omega)$. Consequently, the imaginary part of the Green’s function, the so-called one-particle spectral function $A(k,\omega) = -(1/\pi) \text{Im} G(k,\omega)$, represents the probability of adding or removing a particle from the interacting many electron system.

In a noninteracting system the Green’s function is defined by

$$G_0(k,\omega) = \frac{1}{\omega - \varepsilon_k - i\delta}$$  \hspace{1cm} (4.8)

where $\omega$ is the excitation energy in relation to the Fermi level, $\varepsilon_k$ is the energy of the noninteracting electron. The spectral function in this case has a $\delta$-function peak at $\omega = \varepsilon_k$: $A(k,\omega) = \delta(\omega - \varepsilon_k)$.
For an interacting system the Green’s function is similarly defined by

\[ G(k, \omega) = \frac{1}{\omega - \varepsilon_k - \Sigma(k, \omega)} \] (4.9)

where \( \Sigma(k, \omega) = \Sigma'(k, \omega) + i\Sigma''(k, \omega) \) is the self-energy which contains all the information on the interaction effects. In its turn, the spectral function \( A(k, \omega) \) becomes a continuous function and can be written as follows:

\[ A(k, \omega) = \frac{1}{\pi} \frac{\Sigma''(k, \omega)}{[\omega - \varepsilon_k - \Sigma'(k, \omega)]^2 + [\Sigma''(k, \omega)]^2} \] (4.10)

Therefore, \( \Sigma'(k, \omega) \) and \( \Sigma''(k, \omega) \) give the energy shift and the broadening of the noninteracting energy spectrum at \( \omega = \varepsilon_k \), respectively.

For systems of interacting electrons the quasiparticle (QP) concept has been introduced. In this approach, it is assumed that the interacting electron system is not too different from the non-interacting one in the sense that one can map it onto a system of only weakly interacting quasiparticles (the so-called Fermi liquid). The QP’s can then be described as electrons dressed with a variety of excited states.

In the quasiparticle concept, it is assumed that

\[ |\Sigma''(k, \omega)| \ll |\varepsilon_k + \Sigma'(k, \omega)| \] (4.11)

which leads to distinct maxima in the spectral function for the energy \( \varepsilon_k = \varepsilon_k + \Sigma'(k, \omega) \).

If one develops the denominator in Eq. 4.9 in terms of \( \omega - \varepsilon_k \) neglecting higher terms than linear, one gets

\[ G(k, \omega) = \frac{Z_k}{\omega - \varepsilon_k + i\Gamma(k, \omega)} + G_{inc} \] (4.12)

and hence

\[ A(k, \omega) = \frac{1}{\pi} \frac{Z_k \Gamma_k}{(\omega - \varepsilon_k)^2 + \Gamma_k^2} + A_{inc} \] (4.13)

where \( Z_k = (1 - \frac{\partial \Sigma''}{\partial \omega})^{-1} \) is the quasiparticle spectral weight (also called coherence factor), \( \varepsilon_k = \varepsilon_k + \Sigma'(k, \omega) \) is a renormalized energy dispersion and \( \Gamma_k = Z_k \Sigma''(k, \omega) \) is a reciprocal lifetime of the quasiparticles. All contributions to the spectral function which cannot be mapped onto QP are included in the so-called incoherent part of the spectral function \( A_{inc} \).

Thus, using once again the sudden approximation, the intensity measured in an ARPES experiment (Eq. 4.7) can be written in a following form:

\[ I(k, \omega) = I_0(k, \nu, A)f(\omega)A(k, \omega) \] (4.14)
where $\mathbf{k}$ is the electron momentum, $\omega$ is the electron energy with respect to the Fermi level, and $I_0(\mathbf{k}, \nu, \mathbf{A})$ is proportional to the squared one-electron matrix element $|M_{fi}^k|^2$ and, therefore, depends on the electron momentum as well as on the energy and polarization of the photon. The Fermi function $f(\omega) = (e^{-\omega/T} + 1)^{-1}$ reflects the fact that direct photoemission probes only occupied electronic states. It should be noticed that in Eq. 4.14 the presence of an extrinsic background and the broadening due to finite energy and momentum resolution are neglected, which, however, have to be carefully considered when a quantitative analysis of the ARPES spectra is anticipated.

### 4.1.4 Matrix elements and selection rules

As discussed above and summarized by Eq. 4.14, photoemission directly probes the one-particle spectral function $A(\mathbf{k}, \omega)$. However, in extracting quantitative information from the experiment the effect of the matrix element term $I_0(\mathbf{k}, \nu, \mathbf{A}) \propto |M_{fi}^k|^2$, which is responsible for the dependence of ARPES data on photon energy and experimental geometry, has to be taken into account.

First, let us discuss dipole selection rules which arise from very general symmetry constraints imposed on $M_{fi}^k = \langle \phi_f^k | \mathbf{A} \cdot \mathbf{p} | \phi_i^k \rangle$, and which are necessary for the interpretation of ARPES data. Fig. 4.3 shows the photon incident beam along a plane of mirror symmetry of the sample ($\mathcal{M}$), with the detector located in the same mirror plane.

In order to have a non-vanishing photoemission intensity the final state wavefunction $\phi_f^k$ must be even with respect to reflection in $\mathcal{M}$: odd parity final states would be zero everywhere on the mirror plane and, therefore, also on the detector where, in reality, the photoelectron is an even parity plane wave state $e^{i\mathbf{k}\mathbf{r}}$ with momentum lying in the mirror plane.

The dipole transitions are allowed if the entire matrix element has an overall even symmetry. This implies that $|\mathbf{A} \cdot \mathbf{p} | \phi_i^k \rangle$ must be even. Thus, two possibilities arise. (1) For an initial state $\phi_i^k$ which is even with respect to $\mathcal{M}$, the light polarization $\mathbf{A}$ must also be even, i.e. parallel to $\mathcal{M}$. (2) For an initial state which is odd with respect to $\mathcal{M}$, $\mathbf{A}$ must be also odd, i.e. perpendicular to $\mathcal{M}$. Finally, the polarization conditions can be summarized as $[57]$

$$
\langle \phi_f^k | \mathbf{A} \cdot \mathbf{p} | \phi_i^k \rangle \left\{ \begin{array}{ll}
\phi_i^k \text{ even } & \langle + | + \rangle \Rightarrow \mathbf{A} \text{ even }
\phi_i^k \text{ odd } & \langle + | - \rangle \Rightarrow \mathbf{A} \text{ odd }
\end{array} \right.
$$

(4.15)
Figure 4.3: Arrangement of the photon beam and detector in order to make use of the photoemission selection rules.

As an example, let us examine hybridized $Cu3d - O2p$ initial states, as shown in Fig. 4.4, which have $d_{x^2-y^2}$ symmetry at the Cu site.

These states are even under reflection in the plane defined by the $(0,0) - (\pi,0)$ symmetry axis and $z$-axis. Therefore, measurements along the $(0,0) - (\pi,0)$ direction will be dipole-allowed (forbidden) if the polarization vector $A$ is parallel (perpendicular) to this axis. The situation is different for reflection with respect to plane formed by $(0,0) - (\pi,\pi)$ axis. In this case, the dipole transitions will be allowed (forbidden) if the polarization vector $A$ is perpendicular (parallel) to $(0,0) - (\pi,\pi)$ axis.

Hence, the preferable polarization conditions can be selected for photoemission measurements at high symmetry points of the Fermi surface (right panel of Fig. 4.4): for an antinodal point polarization should be parallel to the $(0,0)-(\pi,0)$ direction, for a nodal point polarization should be perpendicular to the $(0,0)-(\pi,\pi)$ direction [58, 59].
4.2 Experimental setup

The experimental setup for ARPES measurements can be described by the flowchart shown on Fig. 4.5. All components shown here have to be operated under ultra high vacuum (UHV) conditions.

![Flowchart of ARPES setup]

**Figure 4.5**: Principle setup of the ARPES experiments.

4.2.1 Photon sources

All experimental results presented below were derived using either a He-discharge lamp or synchrotron radiation (SR) from an undulator as a photon source.

In the Gammadata SCIENTA VUV 5000 He-lamp the radiation from a low pressure He-plasma generated by the Electron Cyclotron Resonance (ECR) technique is used [60].
Figure 4.6: Low pressure He gas in the small cavity is excited by ECR electrons producing high intensity VUV photons.

The ECR resonance is created locally in a small metallic discharge cavity by the application of a strong permanent magnetic field perpendicular to the microwave electric field. This will force the electrons into circular orbits (see Fig. 4.6) with a frequency of 10 GHz. The microwaves are generated by a 10 Ghz klystron amplifier with a power of 250 W. Since each electron is circling around in the cavity, the probability that it will interact with a He atom before being lost to the walls is high, despite the relatively low pressure of typically 50 mTorr. The high power density of the source and the high electron temperature produce an intense photon flux from both neutral and ionized helium.

The UV intensity emitted through an aperture is \(10^{16}\) photons/sr-sec, which is about 500 times higher than that from capillary UV sources. The flux density per unit wavelength interval is comparable to that from undulator beam lines.

Combined with the Gammadata SCIENTA VUV 5040 toroidal grating monochromator, specially designed to collect and focus the VUV light from VUV 5000 lamp, the source gives a pure line radiation. For example, for HeI \(\alpha\) emission, the FWHM is less than 1.5 meV. Other frequently used wave lengths and relative intensities emitted by a Gammadata VUV 5000 source are presented in Tab. 4.1.

In comparison with conventional light sources, such as gas discharge lamps or x-ray anodes, synchrotron radiation has a lot of advantages. Besides offering a
### 4.2. Experimental setup

<table>
<thead>
<tr>
<th>Transition</th>
<th>Short</th>
<th>Energy (eV)</th>
<th>Intensity (%)</th>
<th>FWHM (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$2p^1P \rightarrow 1s^2 ,^1S$</td>
<td>HeI $\alpha$</td>
<td>21.218</td>
<td>$\approx 87$</td>
<td>$&lt; 1.5$</td>
</tr>
<tr>
<td>$3p^1P \rightarrow 1s^2 ,^1S$</td>
<td>HeI $\beta$</td>
<td>23.087</td>
<td>$\approx 8$</td>
<td>$&lt; 1.5$</td>
</tr>
<tr>
<td>$2p^2P \rightarrow 1s^2 ,^2S$</td>
<td>HeII $\alpha$</td>
<td>40.814</td>
<td>$&gt; 5$</td>
<td>$&lt; 2$</td>
</tr>
</tbody>
</table>

Table 4.1: Energies and intensities for the most intense transitions of the VUV 5000 He-lamp.

![Synchrotron radiation from bending magnet (left) and undulator (right).](image)

higher photon flux, it also gives a very broad range of photon energies together with a high degree of linear polarization (for the detailed description of synchrotron radiation technology and its applications see Ref. [61]).

Synchrotron radiation is created when charged particles (electrons or positrons, for example), which travel at relativistic speed ($E = \gamma mc^2$), are accelerated. This is done by means of magnetic fields, which force them to change the direction of their velocity vector to follow a circular (bending magnet) or oscillatory path (undulator). The emitted radiation is concentrated into a forward cone of a small opening angle which is directed tangentially to the travelling path of the particles (Fig. 4.7), a pattern which is drastically different from the dipolar radiation pattern emitted from accelerated charges at non-relativistic speeds. A further important feature of synchrotron radiation is that the radiation emitted in the orbital (oscillation) plane of the charged particles is linearly polarized with the polarization vector in the orbital (oscillation) plane.

Synchrotron radiation from the Berlin electron storage ring facility BESSY has been used for the experiments described in this thesis. The parameters of the undulator beamline are compiled in Tab. 4.2.

Note that in the case of undulator type insertion devices (ID), synchrotron radiation emitted by each period interferes coherently. That gives us a strongly collimated ($\theta = \frac{1}{\gamma \sqrt{n}}$) quasimonochromatic ($\Delta \lambda / \lambda = 1/n$) high intensity ($I \propto n^2$)
light source [64]; here $\gamma = \sqrt{1 - \frac{v^2}{c^2}}$ is a relativistic factor of accelerated electrons and $n$ is a number of undulator periods (see Fig. 4.7).

To select the photons at a fixed but variable energy a plain grating monochromator (PGM) with a motor driven diffraction grating is used and operated in the first diffraction order in which the desired wave length $\lambda$ can be chosen by the Bragg angle $\theta$ according to $2d \sin \theta = \lambda$ with $d$ being the distance between the lines of the grating. This finally gives a resolving power $E/\Delta E$ of about $10^4$ which corresponds to 2 meV resolution for 20 eV photon energy. The monochromator is followed by the UHV experimental chamber equipped with an analyzer and manipulator where the sample is mounted.

### 4.2.2 Electron energy analyzer and detector

In this work Gammadata $SES$ 100 and $SES$ 200 electron energy analyzers with a 2D multichannel detection system [60] have been used for ARPES experiments on two different experimental stations.

In both cases the electron analyzers are electrostatic full hemispherical analyzers of mean radius 100 ($SES$ 100 for experiments with synchrotron radiation) and 200 mm ($SES$ 200 for experiments with a He-lamp).

![Schematic diagram of the hemispherical analyzer and detector system.](image)

**Figure 4.8:** A schematic picture of the hemispherical analyzer and detector system.
The spherical deflection analyzer causes the actual energy dispersion. It consists of two concentric hemispheres (Fig. 4.8). The electron trajectories are bent in the radial electrostatic field between the two concentric hemispheres with a voltage difference between them. For transmission of electrons with pass energy $E_0$ along the path with $R_0 = (R_{in} + R_{out})/2$, the potential of the outer hemisphere has to be $V_{out} = E_0[3 - 2(R_0/R_{out})]$, and that of the inner hemisphere has to be $V_{in} = E_0[3 - 2(R_0/R_{in})]$ and, thus an energy/spatial dispersion is achieved.

The analyzer is equipped with an accelerating/retarding multi-element electrostatic lens system to achieve best focussing on the entrance slit. The lens system can be utilized in several modes of operation in different types of experiments. Double $\mu$-metal shielding ensures optimum performance even in the presence of magnetic fields.

The detector system is responsible for the detection of the electrons and their exact position in two dimensions. This makes it possible to determine their original energy and one additional parameter: either their original spatial position (the so-called transmission mode) or emission direction (angular mode). The detector system consists of a chevron mounted micro-channel plate (MCP) detector with an electroluminescent phosphor screen and CCD-camera (Fig. 4.8). The MCP pair multiplies each incoming electron $10^7$ times and this electron pulse is accelerated to the phosphor screen, where it produces a light flash. This is subsequently detected by the CCD camera. The position of the light flash exactly corresponds to the position of the incoming electron. This unique analyzer + detector system gives an energy and angular resolution of a few meV and 0.2°, respectively.

### 4.2.3 Sample preparation

For ARPES experiments single crystalline samples of $Bi_2Sr_2CaCu_2O_8$ (Bi2212) provided by Helmut Berger from the Institute of Physics of Complex Matter, EPFL, Lausanne, Switzerland were used. The pure and lead doped Bi2212 single crystals were grown by a self-flux method from starting oxides and carbonates melted in an alumina crucible at $\sim 1000^\circ$C. Different doping levels of the measured samples were achieved by changing the oxygen content by means of annealing the samples either in oxygen atmosphere for overdoped samples and in argon or helium atmosphere for underdoped samples. Detailed information about the growth of the single crystals can be found in the review [65] and references therein.

The plate-like crystals with a flat mirror-like surface of a typical size of about
$3 \times 3 \text{ mm}^2$ were mounted on the sample holders with UHV compatible silver-loaded epoxy with the flat side of the crystals (parallel to Cu-O planes) being parallel to the flat side of the sample holder. The sample holder itself was placed into a pocket on a two-axis rotatable cryo-manipulator, which can be operated at temperatures between 30 and 300K (Fig. 4.9).

![Image](image1)

**Figure 4.9: Left:** Typical Bi2212 sample glued on the UHV sample holder. **Right:** Sample holder on rotatable cryo-manipulator.

For the surface sensitive photoemission measurements clean crystal surfaces have to be prepared *in-situ* under UHV conditions. Because of the weak van der Waals forces between Bi-O layers in Bi2212, it is possible to cleave the crystal by stripping off an adhesive tape stuck to the sample surface. The quality of the cleavage planes can be judged from the Low Electron Energy Diffraction (LEED) pattern. An example of a typical LEED pattern of a pure Bi2212 single crystal is shown in left panel Fig. 4.10.

It is well known that in the case of pure Bi2212 the $5 \times 1$ superstructure develops in the Bi-O layer (see for example Ref. [66, 67]). This leads to an additional set of features in photoemission spectra presumably originating from the scattering of the outgoing photoelectrons on the modulated Bi-O layers [26, 27]. The use of the modulation-free lead doped Bi2212 samples allows us to get rid of these superstructure spots (right panels on Fig. 4.10) and related scattering.
4.3 Measurements and data analysis

Usually in ARPES experiments one uses photons with a fixed energy \( h\nu \) and measures the number of photoelectrons per time interval and solid angle depending on their kinetic energy with the emission direction as a parameter.

In our experimental setup, the discrimination between different kinetic energies of the photoelectrons is done by means of an electrostatic analyzer, and the direction of the photoelectron momentum vector is chosen by the azimuthal and polar angles \( \phi \) and \( \theta \) of the rotatable manipulator with respect to the analyzer’s entrance slit.

Using a two-dimensional detector it is possible to collect simultaneously a number of electrons differing in the emission angle along the entrance slit of the analyzer (left panel in Fig. 4.11). This provides us with an additional momentum distribution of the measured photocurrent for a given binding energy. It is thus possible to obtain a 3D set of data: intensity as a function of momentum and energy (right panel in Fig. 4.11).

Therefore, the ARPES experimental data can be presented in different ways. From the measured intensity distribution \( I(k, \omega) \) the so-called energy distribution curve (EDC) can be obtained by fixing the value of momentum (i.e. cutting the data set from Fig. 4.11 parallel to the energy axis). One can also cut the same \( I(k, \omega) \) distribution parallel to the momentum axis. These cuts are termed the

\[ h\nu \]
momentum distribution curves or MDC’s [68]. An MDC reflects the vector nature of momentum and is uniquely defined by a chosen frequency (binding energy) and an arbitrary path in a two-dimensional $\mathbf{k}_\parallel$ space.

The use of a color scale to represent the intensity enables an efficient presentation of the three-dimensional data set (right panel of Fig. 4.11). The result, shown in the upper right panel of Fig. 4.12, is called an energy distribution map (EDM). The EDM can be thought of as an array of EDC’s (or MDC’s) taken along a particular path in the Brillouin zone within a particular range of binding energies.

If we fix a binding energy and plot the intensity with a color-scale in series of MDC’s covering an area in $(k_x, k_y)$ space, then we arrive at a momentum distribution map or MDM, which represents a constant energy surface. The lower right panel of Fig. 4.12 shows such an MDM for binding energy $E_B = 0$, also called a Fermi surface map.

It is wise to note here some of the problems in analyzing EDC’s and contrast them with the MDC’s case. First, note that the EDC lineshape is a complex function of $\omega$ for two reasons. The trivial reason is asymmetry introduced by the Fermi function $f(\omega)$ which cuts off the positive $\omega$ part of the spectral function. The more essential is that the self-energy has non-trivial $\omega$ dependence and this makes the full $A(\mathbf{k}, \omega)$ non-Lorentzian in $\omega$ as seen from Eq. 4.10. Thus, one is usually forced to model the self energy and make fits to the EDC’s. At this point, one is further restricted by the lack of the detailed knowledge of the additive extrinsic background.
4.3. Measurements and data analysis

Figure 4.12: Representing ARPES data in 2D [69].

The MDC's have certain advantages in studying gapless excitations near the Fermi surface. For \( k \) near \( k_F \) we can linearize the bare dispersion \( \epsilon_k \simeq v_F (k - k_F) \), where the \( v_F = \frac{\partial \epsilon_k}{\partial k} \) is the bare Fermi velocity. It is then easy to see from Eqs. 4.10 and 4.14 that the MDC is a Lorentzian:

(a) centered at \( k = k_F + [\omega - \Sigma'(\omega)]/v_F \); with

(b) width (HWHM) \( W_M = |\Sigma''(\omega)|/v_F \).

Thus the MDC has a very simple lineshape, and its peak position gives the renormalized dispersion, while its width is proportional to the imaginary part of the self-energy.
Chapter 5

Bilayer splitting in Bi-2212
5.1 Superconducting gap in the presence of bilayer splitting in underdoped (Pb,Bi)2212

In the light of the recently observed BS, the natural and straightforward question appears: "how does the superconducting gap behave on both Fermi surface sheets?" Could it be that superconductivity in Bi2212 is sheet dependent as, for instance, is the case of $2H - NbSe_2$ [70] or $MgB_2$ [71]. Does the strong correlation between the so-called range parameter (related to the hoping integral $t'$) and $T_c$ max, found recently [72] for nearly all hole-doped one-layer and bonding subband of multilayer HTSC cuprates, really mean that the bonding component plays a more important role in superconductivity than the antibonding? What are the gap symmetry properties for bonding and antibonding bands?

To clarify these questions we determined the superconducting gaps for both Fermi sheets. The main difficulty here is to resolve separately the bonding and antibonding bands. In Fig. 5.1 the clearly visible bilayer splitting is shown on the energy distribution map (EDM) recorded for underdoped ($T_c=77K$) (Pb,Bi)2212 sample along the azimuthal path (see dashed line in the inset of Fig. 5.2). This can possibly lead to the existence of two gaps related to the bonding and antibonding Fermi surfaces (FS). In the lower panels of Fig 5.1 three energy distribution curves (EDC's) are shown, which correspond to cuts located at the numbered white dotted lines of the EDM. Spectra 1 and 3 are taken at $k_F$ vectors determined for $T > T^\ast$. Despite being very close to one another in $k$-space, they have completely different
Figure 5.1: Energy distribution map recorded at $T=40K$ (upper left panel) showing the clearly resolved bilayer splitting. EDC’s from the k-points marked by the white dotted lines on the EDM (lower panels). Comparison of EDC’s 2 and 3 on an expanded energy scale (upper right panel).

lineshapes from which a superconducting gap value can be derived. EDC number 2 is from just in between the gapped sheets of the FS and therefore is not suitable for the determination of the superconducting gap. The upper right panel on Fig. 5.1 compares EDC’s 2 and 3 in detail to illustrate how erroneous the determination of the superconducting gap could be if the precise k-location is uncertain. In this case, the peaks of the EDC’s are separated by $\sim10$ meV and the leading edges by $\sim6$meV. Depending on which method is used for the determination of the gap value
5.1. Superconducting gap in the presence of bilayer splitting in underdoped (Pb,Bi)$_{2212}$

(see below), the error could amount to as much as 50\% of the gap value reported before for this $k$-space region. It is also easy to predict an error which appears as the result of the none-resolved BS due to insufficient angular resolution. Summing up EDC’s from larger $k$-area would inevitably result in an overestimation of the gap values since both peak and leading edge positions of "irrelevant" EDC’s are always at higher binding energies.

In most cases, the excitation conditions which define the distribution of the photocurrent via matrix elements are such that the bilayer splitting is hardly visible. Every change of a given excitation energy and geometry of experiment leads to a particular, as a rule very strong, $k$-dependence of the matrix elements which is, generally speaking, not the same for the bilayer split bands. As it has been shown recently (see chapter 3) the key factor defining relative bonding/antibonding intensity is the energy of the exciting photons. One can exploit these observations and search for suitable excitation conditions by varying the photon energy. This was done here by using tuneable synchrotron radiation. The most convenient region of the $k$-space for such an exercise (in the case of modulation free crystals) is the vicinity of the $(\pi,0)$ point containing the antinodal points of the normal state Fermi surface: here both the superconducting gap and bilayer splitting are expected to be largest.

In Fig. 5.2 azimuthal momentum distribution curves (MDC) are shown for the path indicated in the inset by the double headed arrow. The MDC signal represents integration over 1 degree of polar angle and the first 20 meV in binding energy. The observed variations in the lineshapes with excitation energy are dramatic. If using linearly polarized radiation of $h\nu=21$ eV or $h\nu=38$ eV emissions from the bonding and antibonding bands are comparable, for $h\nu=25$ eV and $h\nu=50$ eV the dominance of antibonding component is obvious. At first sight, it may appear surprising that the MDC’s taken with $h\nu=21$eV and $h\nu=21.2$eV are so different. The two important differences here are polarization and energy. Firstly, for the former, linearly polarized light was used ($E//k$), whereas the latter involved mainly unpolarized radiation from a helium resonance source. Secondly, both theoretical and experimental studies have shown that there is a sharp local minimum in the matrix elements near $\sim 20$ eV [35, 37, 58] and thus in this region, even a small change in energy could be enough to provoke a large variation of the photoemission signal.
Figure 5.2: Azimuthal MDC’s showing how relative intensity of the bonding and antibonding components could be tuned by choosing a suitable excitation energy. The inset illustrates which parts of k-space were probed. The vertical dotted line shows the angular positions of the bonding and antibonding antinodal points. All data were collected for UD77 sample.

At this stage it is necessary to decide which energy would be the most suitable for the determination of the superconducting gaps related to both Fermi surfaces. This choice, in turn, depends on how the gap will be extracted from the spectra. There are two widely-known approaches. The first involves fitting an EDC with
5.1. Superconducting gap in the presence of bilayer splitting in underdoped (Pb,Bi)2212

a model spectral function multiplied by the Fermi function and convoluted with the energy and angular resolution functions [73]. The advantage of this method is that if the fit is successful, one gains access to the absolute value of a gap. The shortcomings of this method include increased error bars due to the fitting procedure itself and, more importantly, a lack of knowledge as regards the precise form of the spectral function, thus making this approach model dependent. The second possibility is the so-called leading edge gap method (LEG), in which the lowest binding energy where the k_F -EDC loses half of its maximal intensity is determined. This approach is attractive in its simplicity and precision (nowadays better than 1 meV), and is by far the most widely applied in the HTSC ARPES literature [14, 56]. The disadvantage is that the LEG is only a good qualitative measure of the gap and does not give its absolute value. Now, both of these gap methods face a real challenge when two separate, gapped features are in close proximity. In this context, the EDC fitting procedure would benefit from having both features of equal intensity (as, for instance, is the case when using linearly polarized 21 eV photons, see Fig. 5.2), whereas the LEG determination would work better when the intensity from the antibonding component dominates, as then the weak bonding component will not strongly influence the position of the antibonding leading edge (see also Fig. 5.1). Since we are interested in a comparative study of the relative sizes and symmetries of the superconducting gaps of the two Fermi surface sheets, the LEG approach is the method of choice and thus the 25 eV data are the most appropriate to answer this question.

After having chosen the excitation energy and the gap determination method, we are still left with the problem which particular EDC should be used to define the LEG (see Fig. 5.1).

By applying a mapping procedure allowing to cover a relatively large k-space region, a new method of presenting information derived from ARPES spectra will be introduced - "the map of gaps" - the momentum distribution of the binding energies of the leading edges. It is the EDM with values of leading edge energy instead of binding energy presented with a color scale.

In Fig. 5.3 such a map is shown as a grey scale image of leading edge binding energy. Four traces of the gapped Fermi surfaces are easily seen, despite the blurring of the bonding components as can be expected from the relative intensities seen in the 25 eV panel of Fig. 5.2. As a next step, the lines formed by joining the "minimum gap loci" [30] were determined , which are then shown superim-
Figure 5.3: Left panel: map of gaps derived from the EDC’s taken with 25 eV photons at \( \mathbf{k} \)-points within the shaded area shown in the inset to Fig. 5.2. Both of the gapped Fermi surface sheets are seen. Right panel: leading edge gaps as a function of the absolute \( \mathbf{k} \)-value. Zero gap corresponds to the binding energy (-7 meV) of the leading edge of the nodal \( \mathbf{k}_F \)- EDC.

posed upon the map of gaps. The LEG values along these minimum gap loci are plotted in the right panel of Fig. 5.3. Thus, as a first major result, these curves unambiguously demonstrate that the superconducting energy gaps on both Fermi surface sheets are identical, indicating no sheet dependence of superconducting order parameter in this case.

Being certain that the gaps are identical, one can take a further step in trying to obtain information about their symmetry. To increase the accuracy of the measurements it is possible to reduce now the influence of one of the bands assuming that both gaps have also the same \( \mathbf{k} \)-dependence in the other part of the BZ, i.e. monotonically decrease when approaching the nodal direction. Looking again at Fig. 5.2 two possible candidates are the photon energies of 21.2 and 50 eV at which emission from the antibonding or bonding bands, respectively, is significantly suppressed. As mentioned before, every experiment using linearly polarized radiation implies particular symmetry conditions which uniquely define the photocurrent distribution. In cuprates, due to the symmetry of the states forming the near-\( E_F \) electronic structure, it is known that favorable conditions for the emission along the \((0,0) - (\pi, \pi)\) direction are such that the vector of polarization should
be perpendicular to this direction whereas for $(0,0) - (\pi,0)$ direction it has to be parallel (see Chapter 4).

In our experimental geometry, when using linearly polarized light from the undulator the polarization vector is always parallel to the studied direction in the reciprocal space and therefore cannot be equally effective for all states at the Fermi level. This means that to be sure not to loose the spectral weight due to symmetry reasons (which is especially important when determining LEG), it is necessary to probe every $k$-point with two mutually perpendicular orientations of the polarization vector which is technically quite difficult. For this reason the data presented here were taken with 21.2 eV radiation from the helium lamp which is mostly unpolarized.

Precise determination of $k$-dependence of the superconducting gap and thus its symmetry requires precise navigation in $k$-space in order to accurately locate a position on the (former) Fermi surface. The common framework in which to discuss the gap symmetry is as a function of the angle $\varphi$ between the $(\pi,\pi) - (\pi,0)$ line and the $(\pi,\pi) - FS$ directions in the reciprocal space, where FS represents the normal state Fermi surface ($\varphi=0^\circ$ for $M = (\pi,0)$-point and $\varphi=45^\circ$ for $\Gamma = (0,0)$-point respectively). This angle $\varphi$ is connected with the standard ARPES azimuthal ($\phi$) and polar ($\theta$) angles via the simple relation:

$$\cos^2\varphi = (1 + ((1 - \eta \sin\theta \cos\phi)/(1 - \eta \sin\theta \sin\phi))^2)^{-1},$$

(5.1)

where $\eta$ is defined by the kinetic energy of the photoelectrons and the lattice constant ($\eta \sim 2.58$ when using 21.2 eV photons).

For this purpose a map of intensity integrated within a 50 meV energy interval is shown in Fig. 5.4(a) which facilitates identification of the $k$-vectors. It should be noted here that not only this map but also an additional Fermi surface map taken at room temperature covering much larger area in $k$-space were needed to assure accurate definition of the angle $\varphi$. Fig. 5.4(b) shows the greyscale "map of gaps". Visual inspection of the map of gaps already clearly points to the anisotropic character of the gap.

One can now use two approaches to construct the $\text{LEG}(\varphi)$ function. One method would be to define the path in the $k$-space using, for example, maxima of MDC's taken perpendicular to the FS and then plot the value of the LEG along this path as a function of $\varphi$. Here the $\text{LEG}(\varphi)$ curve is obtained using the alternative "minimum gap locus" method. The same data set (Fig. 5.4 panel (b)) can be simply re-plotted in other coordinates: LEG value versus Fermi surface
Figure 5.4: a) Angular distribution map of the photoemission intensity integrated within 50 meV of $E_F$ showing the locus of $k_F$-points. b) Map of gaps derived from the EDC’s taken with 21.2 eV photons within the similar $k$-space area. c) Plot of the leading edge gaps as a function of the angle around the Fermi surface. The red symbols are the data from panel (b), dotted lines in both (b) and (c) show the minimum gap locus and corresponding LEG values respectively, the solid line in (c) is the simple $d$-wave gap function $\Delta(\varphi) = \Delta_{\text{max}}|\cos(2\varphi)|$. 
angle (Fig. 5.4 panel (c)). This gives the open circle symbols shown in Fig. 5.4(c). The curve joining the low-gap extremity of these data points represents the true \( \mathbf{k} \)-dependence of the superconducting LEG, and is shown as a dotted line on Fig. 5.4(c). In this way, these extremal LEG values correspond to the points on the "minimum gap locus" dotted line in panel Fig. 5.4(b).

From the comparison shown in Fig. 5.4(c), it is clear that the obtained \( \mathbf{k} \)-dependence of the LEG for these UD77K crystals is quite different from that expected in the case of a simple \( d \)-wave gap function \( \cos(k_x) - \cos(k_y) \). The most striking difference is that the gap behaves much flatter around the node resulting in a "U"-like shape rather than "V"-shape for the LEG(\( \varphi \)) function. Keeping in mind that the LEG is only a qualitative measure of a real gap it is crucial to understand to which extent the LEG(\( \varphi \)) function reproduces the main features of a real gap function. According to the numerical simulations [74] the measured binding energy of the nodal \( \mathbf{k}_F \) -EDC leading edge (-7 meV) corresponds to the gapless spectral function. In other words, the results are consistent with the \( d \)-wave symmetry of the superconducting gap as discussed above.

It was also shown [74] that due to temperature effects the LEG method tracks the real gap with an uncertainty of the order of 2 meV (for the given experimental conditions) when the corresponding absolute values of the real gap are of the same order. This last remark gives finite probability that a nodal "cusp" can be present in the real gap function (although much more flat than is required by the values of the simplest \( d \)-wave function).

The observed partial suppression of the superconducting gap with respect to the simplest \( d \)-wave expectations in underdoped sample is in qualitative agreement with previous ARPES results (see chapter 3). The numerical simulations [74] show that the relation between the LEG and the real gap function is nontrivial and model-dependent. Therefore, quantitative information cannot be extracted here.

However, following the authors of Ref.[45], the observed behavior could be explained by invoking higher harmonics of the gap consistent with \( d \)-wave symmetry: \( \Delta(\varphi) = \Delta_{max}[B\cos(2\varphi) + (1 - B)\cos(6\varphi)] \), with \( 0 \leq B \leq 1 \). The fit of the LEG(\( \varphi \)) function shown in Fig. 5.4(c) near the node using an additional \( \cos(6\varphi) \) term yields the weight of this harmonic of 27% as compared to the weight of the main \( \cos(2\varphi) \) term. It should be noted that the \( \cos(6\varphi) \) term in the Fermi surface harmonics is related to the tight-binding function \( \cos(2k_x) - \cos(2k_y) \), which represents the next nearest neighbor interaction, just as \( \cos(2\varphi) \) is closely related to the nearest
neighbor pairing function $\cos(k_x) - \cos(k_y)$. Therefore, the presence of this higher harmonic is probably connected with an increased strength of the pairing interaction in underdoped compounds due to reduced screening as one approaches the insulator (see also [75], [76]).

## 5.2 ARPES spectral line shape in the $(\pi, 0)$ region

As was shown above, the excitation energy has a strong influence on the photoemission spectral lineshape. In the region of the $(\pi, 0)$ point of the Brillouin zone (BZ), where the bilayer splitting (BS) has the maximal value the most dramatic changes in the spectral line shape were previously observed (see chapter 3). Therefore, the next step in clarifying the influence of this splitting on the ARPES results is to investigate in detail how matrix elements depend on photon excitation energies for both bonding and antibonding bilayer bands.

### 5.2.1 Overdoped (Pb,Bi)2212

To answer this question a collection of superconducting state $(\pi, 0)$ photoemission spectra for overdoped ($T_c = 69$K) (Pb,Bi)2212 was recorded using different excitation energies $h\nu$ (18-65 eV). The black lines in Fig. 5.5 show the experimental data and the red lines represent the results of a fitting procedure which is described below. Upon visual inspection of the raw experimental data in Fig. 5.5, it is evident that the PDH lineshape can no longer be considered to be the result of single band spectral function with a sophisticated self-energy as it was suggested earlier [25]. In spite of the fact that at some excitation energies (e.g. 20 eV) all three components (peak, dip and hump) are present, there are also photon energies at which there is virtually no dip (25 or 42 eV), no hump (50 eV) or even no peak (39 eV). A further, firm conclusion which can be made from inspection of the raw data alone, is that the hump itself cannot be considered as a feature appearing at a fixed binding energy for all $h\nu$ (compare the EDCs for $h\nu$ 20 and 37 eV, for example). For further analysis a fitting procedure has been applied.

Taking into account the bilayer splitting (BS), a two-peak fitting procedure has been applied to analyze changes in the line shape. The results are shown on Fig. 5.5. The measured photocurrent $I(\omega)$ in this case can be written as a sum of two spectral features with different binding energies $\varepsilon_b$ and $\varepsilon_a$ (for bonding and antibonding bands, respectively) and with identical single particle spectral
Figure 5.5: The $\left(\pi, 0\right)$ photoemission spectra from the superconducting state of an overdoped (69K) sample for different excitation energies: the black lines show the experimental data and the color lines represent the results of two-peak fitting procedure which is described in the text.
function $A(\omega)$:

$$I(\omega, h\nu, T) \propto [(M_a(h\nu)A(\omega, \varepsilon_a, T) + M_b(h\nu) \times A(\omega, \varepsilon_b, T)) f(\omega, T) \otimes R_\omega(h\nu) + B(\omega, T)],$$

$$A(\omega, \varepsilon, T) \propto \frac{|\Sigma''(\omega, T)|}{(\omega - \varepsilon)^2 + \Sigma''(\omega, T)^2},$$

where

$$f(\omega, T) = \frac{1}{1 + \exp(-\omega/T)},$$

is a Fermi function, $R_\omega(h\nu)$ is the experimental resolution, and

$$B(\omega, T) \propto (1 + b\omega^2) f(\omega - \Delta_{bg}(T), T + T_b)$$

is a background, which was approximated (assuming it to be $k$-independent) by taking EDC's from the $(\pi/2, \pi/2)$ point in the empirical form with $b = 1 \text{ eV}^{-2}$, $\Delta_{bg}(30 \text{ K}) = 5 \text{ meV}$ and $T_b = 90 \text{ K}$.

In Fermi liquid (FL) theory, the quasiparticle damping near the Fermi surface behaves as $1/\tau \propto \omega^2$ or $(\pi T)^2$, whichever is larger. This gives rise to a $\omega^2(T^2)$ behavior of the imaginary part of the self-energy [77]. However, the fit containing the Fermi liquid self-energy shows a significant disagreement with the experimental EDCs (see Fig. 5.6a). While the superposition of FL-based spectral functions well reproduces the "peak-dip-hump" lineshape and perfectly fit the low energy peak, it does not fit the high energy hump.

Thus, for quantitative analysis a non-Fermi liquid self-energy is required. It was found, that the variety of unusual properties of high-temperature cuprates (see chapter 3) are consistent with an anomalous form for the quasiparticle lifetime $1/\tau \propto \text{max}(\omega, T)$, described by the so-called marginal Fermi liquid (MFL) theory [78]. In the case of the marginal Fermi liquid the renormalized imaginary part of self energy can be written as

$$\Sigma''(\omega, T) = \sqrt{\omega^2 + (\beta T)^2}$$

The parameters $\alpha = 1.1(1)$ and $\beta = 2$ were taken close to the values derived from a similar analysis of the EDC's measured along the $(0, 0) - (\pi, \pi)$ direction [69].

If for simplicity, one assumes the renormalization of the quasiparticle energy by a factor $\lambda$:

$$\Sigma'(\omega) = -\lambda \omega + o(\omega^3)$$
with $(\lambda > 0, \omega > 0)$, then such a renormalization can be handled by a simple scaling procedure: 

$$
\varepsilon/(1 + \lambda) \rightarrow \varepsilon, \quad \Sigma''/(1 + \lambda) \rightarrow \Sigma'', \quad (1 + \lambda)A \rightarrow A.
$$

This resulting marginal Fermi liquid (MFL) theory based fit gives the best agreement with experiment (see Fig. 5.6b). The FWHMs of the resolution Gaussians $R_{\omega}(h\nu)$ were determined for each value of photon excitation energy from measurements of the Fermi edge of polycrystalline gold.

The fitting components are shown beneath each spectrum as thin solid lines: grey lines represent the background, green and blue lines represent the photocurrent from the "bonding" and "antibonding" bands, respectively.

Before going further, three points can be stated:

(i) The position of the renormalized band $\varepsilon$, indicated in each fit as a solid vertical line, does not coincide with maxima in the EDCs, $\omega_m$. This is clearly seen for the 'bonding' peak where $\omega \gg T$ and one can evaluate $\varepsilon = \omega_m \sqrt{1 + \alpha^2}$, or alternatively, for the bare band position one can write:

$$
\varepsilon_{bare} = (1 + \lambda)\varepsilon
$$

(ii) Although the superconducting gap $\Delta$ can be included into the dispersion, with $\varepsilon$ then becoming $\sqrt{\varepsilon^2 + \Delta^2}$, $\Delta$ cannot exceed the $\varepsilon_a$ value for the antibonding band giving rise to the low energy feature. As will be described below, $\varepsilon_a$ is 11 meV, which is close to $\Delta$ derived in the weak-coupling BCS scheme for the given $T_c$. 

Figure 5.6: $h\nu = 20$ eV overdoped Pb-Bi2212 ($T_c = 69$K) photoemission spectra (black line) together with the Fermi liquid (a) and marginal Fermi liquid (b) based fit (red line). Blue and green lines present contributions from the peak and hump, respectively. The grey line shows the experimentally obtained background (see text for details).
Figure 5.7: The intensity prefactors $M_a$, $M_b$, and $M_c$ as functions of excitation energy for a two-peak (upper panel) and a three-peak (lower panel) fitting procedure for an overdoped ($T_c = 69$ K) sample.
(iii) The model upon which the fit is based, and in particular the self-energy function, represents only one choice from myriad possibilities. In the low energy region superconductivity has been assumed having no effect on $\Sigma''$, and no coupling to any low-energy bosons was taken into account. Given that the spectral form of feature 'a' depends upon the low-energy properties of the self-energy function, and that the width of this narrow feature is partially determined by the resolution, it is not possible to extract reliable information on the low-energy behavior of $\Sigma''$ from the fit. On the other hand, from the spectral form of feature 'b', one can obtain reliable information on the high-energy behavior of the self-energy function. The fit indicates that $\Sigma''$ at higher binding energies is linear in $\omega$, not only at the antinodal point along the diagonal of the BZ [69], but also that this behavior is a good approximation for higher binding energies at the $(\pi, 0)$ point. Here it should be emphasized again that the spectra cannot be fitted with comparable accuracy using a conventional Fermi-liquid self-energy $\Sigma'' \propto \omega^2$.

The fitting procedure was done as follows: as a first step, the parameters $\alpha = 1.1(1)$, $\varepsilon_a = 11(1)$ meV, $\varepsilon_b = 154(4)$ meV were determined using the fits of the spectra with $h\nu = 19$, 20 and 21 eV, and then all other spectra were fitted using only two free parameters: $M_a$ and $M_b$. These quantities - which will be directed to in the following as 'matrix elements' - are presented in the upper panel of Fig. 5.7 as functions of $h\nu$. This dependence clearly shows that the global assignment of the peak and hump in the PDH spectra to two different electronic states is fully justified. However, it should be stated that as there was no $h\nu$ for which either $M_a$ or $M_b$ were exactly zero, it cannot be excluded that each individual spectral function has a small, intrinsic PDH-like lineshape.

Turning back to Fig. 5.5, one can easily notice that while at some of the excitation energies (18-30, 50 and 65 eV) the two-band fit is very good, at other energies there are small deviations within the binding energy range from 50 to 100 meV. These deviations are photon energy dependent and thus cannot be explained by any kind of an $h\nu$ independent self-energy. This implies the presence of a third feature in these spectra residing at a binding energy of about 75 meV with about 50 meV FWHM. The question then arises as to whether this third feature is related to either the bonding or antibonding band, or whether it is 'extra'.

In order to clarify this point all the $(\pi, 0)$ EDCs were fitted including three non-background components (Fig.5.8). Such a three-component fit perfectly coincides with the experimental data for all photon energies and gives the following positions
Figure 5.8: The $(\pi, 0)$ photoemission spectra from the superconducting state of an overdoped (69K) sample for different excitation energies: the black lines show the experimental data and the color lines represent the results of three-peak fitting procedure.
for the bands: \( \varepsilon_a = 11(1) \) meV, \( \varepsilon_b = 150(4) \) meV, \( \varepsilon_c = 75(5) \) meV, where the c designates the 3rd feature. The \( h\nu \)-dependence of corresponding matrix elements \( M_a, M_b \) and \( M_c \) are shown in the lower panel of Fig. 5.7.

The fact that the \( M_a(h\nu) \) and \( M_b(h\nu) \) functions exhibit practically the same form for both the 2 or 3-component fits shows that this result is robust with respect to the introduction of the 3rd peak. Consequently, Fig. 5.7 provides two pieces of evidence that exactly these two features are originated from the split CuO band:

1. The \( h\nu \)-average values of \( M_a \) and \( M_b \) are comparable \( (M_b / M_a = 1.2) \) as could well be expected from a pair of bands of the same atomic character split by the c-axis bilayer coupling while the average value for \( M_c \) is much smaller \( (M_b / M_c = 2.9) \);

2. The \( h\nu \)-dependencies of \( M_a \) and \( M_b \) are in a good agreement with recent calculations \[35, 37\] as regards the minima at 20 and 28-30 eV, the maxima at 18 and 23 eV, the \( a \) band minimum at 40 eV, as well as the relative intensities of these bands in the 30-40 eV range.

Further support for this assignment comes from the fact that the energetic separation between the renormalized band positions is about 140 meV, which leads to a difference between the peak maxima in the EDCs of 65-85 meV, in agreement with the normal state splitting observed recently \[38\], or to a difference in the bare band positions of about 300 meV which is consistent with the results of the tight-binding calculations \[79\].

An additional evidence for the fact that 'peak-dip-hump' lineshape is due to the bilayer splitting comes from the temperature dependence of the measured dispersion. The series of EDCs along the \((0, 0) - (\pi, 0)\) direction above and below \( T_c \) are shown in Fig. 5.9. By choosing the spectra with excitation energy \( h\nu = 21.2 \) eV (at which the 3rd feature is weak) it is easy to observe the conventional dispersion of the both 'peak-dip-hump' forming bands. The bonding band mainly determines the position of the room temperature EDC's maxima, whereas the maxima of the low temperature spectra indicate the position of the antibonding band. Both bands have a dispersionless region in vicinity of \((\pi, 0)\) point as it is predicted by band structure calculations (see for example Ref. \[74\]).

Therefore, the temperature, like polarization or variable excitation energy, can also be used to control the relative intensities of the the bonding and antibonding components. At 300 K the actual amplitude of the antibonding peak is much closer to the Fermi level than the bonding one and therefore:
Figure 5.9: The energy distribution maps and series of EDCs of an overdoped (69K) sample, self-normalized to its maximum, along the (0, 0) – (π, 0) direction in the BZ for 300K (left panels) and 39K (right panels).
(I) is more strongly influenced by temperature (see Eq. 5.2 and Eq. 5.3),
(II) is strongly cut by the Fermi function (Eq. 5.4),
(III) has twice smaller matrix element (see Fig. 5.7).

As a result it has noticeably lower amplitude than the bonding peak which
mainly determines the position of the room temperature EDC maxima.

Figure 5.10: The EDCs of an overdoped (69K) sample at (π, 0) point in BZ for
normal (T=300K) and superconducting (T=38K) state: experimental data and a
simulated spectra.

The substitution of the given temperatures into Eq. 5.2 gives a good coincidence
with the experimental spectra. In Fig. 5.10 the experimental data are shown
together with the simulation, were the same set of parameters was used except
the temperature. It should be stressed here, that the bonding band is present in
the low temperature spectra giving rise to the hump in 'peak-dip-hump' spectral
lineshape at (π, 0) point (left panel in Fig. 5.9).

5.2.2 Underdoped (Pb,Bi)2212

A similar photon energy dependence of the (π, 0) spectra lineshape was observed
for an underdoped (Tc = 77K) (Pb,Bi)2212 sample (Fig. 5.11). At first glance, a
comparison of these raw experimental data with analogous data from overdoped
(Pb,Bi)2212 (Fig. 5.5) immediately suggests a similar scenario - the PDH line
shape is strongly excitation energy dependent and therefore cannot be considered
Figure 5.11: The $(\pi, 0)$ photoemission spectra from the superconducting state of an underdoped (77K) sample for different excitation energies: the black symbols show the experimental data and the color lines represent the results of the fitting procedure which is described in the text.
5.2. ARPES spectral line shape in the \((\pi, 0)\) region

Figure 5.12: The intensity prefactors \(M_a\), \(M_b\) and \(M_c\) as functions of excitation energy for the three-peaks fitting procedure for an underdoped \((T_c = 77\ K)\) sample. as originating from a single complex spectral function.

One easily notices the varying relative intensity of the low-energy (peak) and the high-energy (hump) features which could be naturally assumed to be the consequence of the different emission probability (matrix elements) from the separate bands as discussed above. Moreover, the excitation energy dependence of the relative intensities qualitatively agrees with the one observed in the overdoped regime, as one can intuitively expect for the split pair of bands of the same atomic character.

However, a closer inspection of Fig. 5.11 reveals that there is an important and noticeable difference with respect to the data from overdoped crystals. While the overdoped data sets were characterized by the fact that a significant number of spectra exhibited no dip in the lineshape at all, such smooth, 'dip-less' spectra are remarkable by their absence in the underdoped data. Every spectrum in Fig. 5.11 possesses either a dip or a plateau feature (e.g. \(h\nu = 29\) eV and \(h\nu = 50\) eV spectra) which separates the high and low energy parts of the spectral profile.
To go beyond this qualitative description the fit of the \((\pi, 0)\)-spectra with three features (plus a background) was done as for the overdoped case. The only difference in the fitting procedure which was applied here for the underdoped case was an energy shift of the binding energies by the gap value of \(~20\text{meV}\) to account for the substantially larger gap. In Fig. 5.12 the photon energy dependence of the intensity prefactors of each of the three components of the fit, \(M_a\), \(M_b\) and \(M_c\) is plotted.

There is a global agreement between the behavior of the matrix elements of the 'hump' and 'peak' in the underdoped and overdoped samples. This immediately indicates that all arguments regarding the assignment of these features to the bonding and antibonding bands in the overdoped regime are fully applicable here: on the "large scale" the PDH line shape is due to a superposition of these two components. What is really different between Figs. 5.7 and 5.12 is the behavior of the third feature of the fit \((M_c)\). While in the overdoped case \(M_c\) is relatively small and its energy dependence barely tracks that of either of the other two features, in the underdoped case one clearly sees a striking similarity between \(M_a\) and \(M_c\), i.e. between the 'peak' and the third feature. Such a close similarity implies that these two features are components of the same, single spectral feature which possesses a more complex lineshape. Thus, these data provide evidence for a variation of the quasiparticle self-energy in HTSC as a function of doping. Such changes are extremely important to understand the properties of HTSC as they might also allow insight into the mechanism which causes superconductivity. The origin of the observed changes will be clarified in the next chapter.
Chapter 6

Coupling to a bosonic mode
In the previous chapter, it was shown that the emission from the bonding and antibonding bilayer split band is consistently photon energy dependent. This now harbors the intriguing possibility to study the related quasiparticle properties of a particular band by simply choosing the appropriate excitation conditions. The following chapter is devoted to the analysis of intrinsic quasiparticle properties.

6.1 Intrinsic peak-dip-hump

As can be seen from Figs. 5.7 and 5.12, the matrix element for photoelectron emission from the bonding band has a local minimum for both doping levels at $h\nu = 29$ eV and 50 eV. Keeping in mind that the bonding band lies much deeper in energy than the antibonding band, this effect is further multiplied by the strong broadening induced by the frequency-dependent self-energy. Thus, for 29 and 50 eV photon energies, the contribution of the bonding band to the $(\pi,0)$ spectral lineshape is vanishingly small. Consequently, one can consider the spectra measured with these photon energies as originating from the antibonding band and, therefore, examine their lineshape in terms of a single spectral function.

In Fig. 6.1 the corresponding spectra ($h\nu = 29$ eV (a) and $h\nu = 50$ eV (b)) from the underdoped ($T_c = 77$K) sample are compared with the $h\nu = 50$ eV (c) spectrum from the overdoped sample ($T_c = 76$K). The difference between the spectra from the two doping regimes is very clear: both EDCs from the underdoped system possess plateau or dip-like features, whereas the overdoped sample evidently

![Graphs](image)

Figure 6.1: EDCs taken at $(\pi,0)$ using 29 and 50 eV excitation energies at which the contribution of the bonding states to the lineshape is negligible, thus unmasking the 'intrinsic' lineshape of the antibonding component.
6.1. Intrinsic peak-dip-hump

exhibits a single component lineshape. Since the effects of the bilayer splitting can be effectively excluded, the lineshape seen in Figs. 6.1 (a) and (b) must be an intrinsic property of the spectral function of the antibonding Cu-O band in the underdoped regime.

The peak-dip-hump lineshape of a single band was discussed previously in terms of a coupling between the electrons and a collective mode (see chapter 3), whereby anomalies are expected in the electronic spectrum at energies where the probability for boson-mediated scattering of the electrons is maximal (see Fig. 6.2).

The mode energy can be read off from the energetic separation between the peak and the 'dip' (or plateau) in the ARPES lineshape [52]. In this case the mode energy is at about 40 meV.

The next step, then, is to determine whether the observed anomaly is linked to the superconducting state. Firstly, the temperature dependence of the $(\pi, 0)$ spectrum from an underdoped sample was recorded using the photon energy $h\nu = 55$ eV when the spectra represent the antibonding band (left panel of Fig. 6.3).

![Figure 6.2: The qualitative model of the coupling between electrons and a bosonic mode in normal (a) and superconducting (b) state [80, 81]. The dashed line exhibits a bare band dispersion, solid line shows the band dispersion renormalized due to the coupling. The singularity in dispersion occurs at energy $|\omega_0 + \Delta|$, where $\omega_0$ is a collective mode energy and $\Delta$ is an energy of a superconducting gap.](image)


Figure 6.3: Temperature dependence of the peak-dip-hump structure at the \((\pi, 0)\) point in the under- and overdoped cases.

The dip (plateau) disappears approximately at \(T_c\), which indicates its intimate relation with superconductivity. The right panel of the same Figure shows the temperature dependence of the "classic" peak-dip-hump from the overdoped sample recorded using the "traditional" choice of \(h\nu = 21\) eV photons. At this doping level, the peak-dip-hump lineshape persists well above \(T_c\), which is fully consistent with previous experiments [82, 83] and the framework discussed above in which the peak-dip-hump lineshape in the overdoped regime is caused by a superposition of the bonding and antibonding bands (Fig. 6.4a).

Returning to Fig. 6.1, it would be, of course, tempting to conclude that the EDCs imply a strong doping dependence of the mode or of the coupling strength. However, as mentioned before, energetic locations of the bonding and antibonding bands change with doping. The consequence is that the antibonding band then is simply too close to the Fermi level to be strongly influenced by the mode [82], implying in its turn that the mode itself is sharply localized in energy (gray ribbon in Fig. 6.4b).

As a result, the "peak-dip-hump" spectral lineshape is due to the superposition of the bonding and antibonding bands, each of them, in their turn, can have an intrinsic dip feature due to the coupling with the collective mode.
Figure 6.4: The peak-dip-hump line shape at \((\pi, 0)\) point as a superposition of the bonding and antibonding bands (a); each of them can have an intrinsic feature due to the coupling with the mode (b). The depicted band structure was calculated using the tight-binding parameters from Ref. [74], the gray ribbon indicates the mode energy

### 6.2 Doping dependence of the coupling

As was shown above, coupling to a bosonic mode can give rise to the intrinsic peak-dip-hump lineshape in the ARPES spectra in the superconducting state of underdoped high temperature superconductors. The properties of this bosonic mode causing feedback effects in the electronic states can be analyzed by studying the photoemission spectra in a large range of dopant concentrations and temperatures.

#### 6.2.1 Separation of the bonding and antibonding band

In order to be able to analyze the doping dependence of the coupling to the bosonic mode the energy distribution maps (EDMs) taken along \((\pi, \pi) - (\pi, 0) - (\pi, -\pi)\) cuts were measured for overdoped, optimally doped and underdoped Pb-Bi2212 samples (Fig. 6.5).

As discussed above (see Figs. 5.7 and 5.12) and supported by theoretical calculations [35, 37], due to matrix element effects the data taken with \(h\nu = 38\) eV (left column in Fig. 6.5) represent mainly the bonding band with some contributions from the antibonding band, while the data taken with \(h\nu = 50\) (or 55) eV have
Figure 6.5: The energy distribution maps along the \((-\pi, \pi)\) \(-(\pi, \pi)\) direction for over-, optimally and underdoped Pb-Bi2212 taken at \(T=30\) K (upper 3 rows) and \(T=120\) K (fourth row). Left column: photon energy \(h\nu=38\) eV. Middle column: \(h\nu=50\) eV. Right column: subtraction of the latter from the former yielding the spectral weight of the bonding band (see text for details).
almost pure antibonding character. The subtraction of the latter from the \( h\nu = 38 \) eV data represents almost the pure bonding bands. This was performed by scaling the \( h\nu = 50 \) eV spectral weight in such a way that at the \((\pi, 0)\) point no negative intensity occurred (right column in Fig. 6.5).

The collection of EDMs shown in Fig. 6.5 is an important and completely new set of ARPES data. They cover the \((\pi, 0)\) region in a wide doping range starting from the overdoped \( (T_c = 69 \text{K}) \) to the underdoped case \( (T_c = 77\text{K}) \) for photon energies which differently select the two bilayer-split bands. In the superconducting state the data taken with \( h\nu = 38 \) eV (left column) show strong changes upon reducing the dopant concentration. First, for the overdoped regime, the bilayer split bands are resolved giving rise to the 'large scale' \((\pi, 0)\) peak-dip-hump (see previous chapter). On going to the underdoped crystals, a qualitatively different picture emerges. The data from the superconducting state (third row) appear to look very puzzling, the two bands merge into one sharp and dispersionless feature located at \( 20-30 \text{ meV} \) followed by an interval between \( 60 \) and \( 70 \text{ meV} \) in which the spectral weight is strongly suppressed. Such a difference in behavior compared with the overdoped case can be attributed to the larger value of the gap and to an apparent stronger renormalization of the bands in the underdoped regime, both of which hamper the visual resolution of the bilayer splitting in this region of \( \mathbf{k} \)-space. The data collected above \( T_c \) (bottom row) for the same underdoped sample substantially clarify the situation: the picture is now remarkably similar to that of the overdoped case, with the two bilayer-split components being clearly seen.

Using the excitation photon energy dependence of matrix elements together with the subtraction procedure described above, one clearly recognizes that in the whole doping range and even in the UD samples the antibonding and the bonding bands can be well separated (middle and right column in Fig. 6.5). For the bonding band the strong renormalization in the superconducting state causes the sharp bending of the electron dispersion below the superconducting gap - the so-called "kink". This bending of the dispersion could be well understood from a qualitative model in Fig. 6.2. While for the higher binding energies the electron dispersion has mostly unperturbed character, for the energies below the \( |\omega_0 + \Delta| \) the strong renormalization due to the coupling with the mode is occurs. The value of the "kink" can be used for the evaluation of the coupling strength (see section 6.2.2). With underdoping the bonding band moves further and further below the Fermi level and the renormalization strongly increases as indicated by
the appearance of a flat dispersion below the gap energy (right column in Fig. 6.5). The antibonding band has a similar renormalization enhanced with underdoping. For both bands the observed flattening of the dispersion is strongly localized in vicinity of the $(\pi, 0)$ point as one can see most clearly for the antibonding band in Fig. 6.6.

Here the energy distribution maps for an underdoped Pb-Bi2212 sample were plotted along the cuts parallel to $(\pi, -\pi) - (\pi, 0) - (\pi, \pi)$, going along the $\Gamma - M$ direction from the $M = (\pi, 0)$ point towards $\Gamma$-point in the second Brillouin zone ($\Gamma_2 = (2\pi, 0)$) as shown in Fig. 6.6b. The data clearly show that in the superconducting state ($T = 30K$) the two antibonding bands related to the two neighboring barrels of the Fermi surface merge in the one undispersive feature at vicinity of the $(\pi, 0)$-point.

Small changes in the dispersion of the bonding band in the antinodal ($M - X$) direction, comparable with one for the nodal ($\Gamma - X$) \cite{84}, have been observed before in overdoped samples \cite{85}, but this is the first time where a total 'wipe-out’ of the spectral weight at energies some 30-40 meV below the superconducting gap near the $(\pi, 0)$ point has been shown for underdoped HTSCs. It is essential that this dramatic difference in the spectra corresponding to both bonding and antibonding band argues for a strong doping dependence of the feedback effects caused by the bosonic mode and thus for an anomalous enhancement of the coupling strength upon underdoping.
6.2.2 Analysis of the coupling strength

It is tempting to carry out the analysis of the coupling strength in a quantitative sense. For the quantitative analysis, the value of the coupling strength constant $\lambda = -\frac{\partial \Sigma' \omega}{\partial \omega}$ can be evaluated using two methods.

For a steep dispersion, as in the case of overdoped samples (left panel in Fig. 6.7), one can fit momentum distribution curves and obtain a dispersion from the MDC peak positions. Considering the ratio of the bare Fermi velocity value $v_F^b$ to the Fermi velocity $v_F^{sc}$ in the superconducting state, one can calculate the coupling strength constant (or renormalization parameter) $\lambda$.

$$1 + \lambda_{sc} = \frac{v_F^b}{v_F^{sc}}$$

(6.1)

The bare Fermi velocities were taken from a previous analysis of the Fermi velocity in the nodal direction, based on a Kramers-Kronig analysis of the width of the MDC dispersion, including tight-binding bandstructure calculations [74]. The obtained values of about 3eV are close to those derived from the analysis of the anisotropic plasmon dispersion [86] and obtained from LDA bandstructure calculations [34].

![DOS and MDC plots](image)

Figure 6.7: **Left:** the subtracted energy distribution map and the bonding band dispersion obtained from the MDC maxima positions (blue points) together with the resulting fit (red solid line). **Right:** MDC and two-Lorentzian fit for maxima determination (see text for details).

For a flat dispersion, as in the case of underdoped samples (left panel in Fig. 6.8), one can fit the energy distribution curves with a Gaussian and obtain a dispersion from the EDC peak positions.
Figure 6.8: Left: the subtracted energy distribution map and the bonding band dispersion obtained from the EDC maxima positions (blue points) together with the resulting fit (red solid line). Right: EDC and Gaussian fit for maxima determination (see text for details).

Then the dispersion $\omega(k)$ in the vicinity of the gap can be approximated by the BSC-like expression:

$$\omega(k) = \sqrt{\epsilon_k^2 + \Delta^2}$$

(6.2)

where $\omega(k)$ is experimental band energy, $\Delta$ is the superconducting gap and $\epsilon_k$ is the renormalized band dispersion which is given as:

$$\epsilon_k = \frac{\epsilon_k^b}{1 + \lambda}$$

(6.3)

where $\lambda$ is a renormalization parameter, and $\epsilon_k^b$ is a bare dispersion.

In vicinity of the Fermi level ($k \rightarrow k_F$) the bare dispersion $\epsilon_k^b$ can be approximated as following:

$$\epsilon_k^b = \frac{\partial \epsilon_k^b}{\partial k}(k - k_F) = v^b_F(k - k_F)$$

(6.4)

where $v^b_F$ is the Fermi velocity of the bare particles without dressing (see chapter 4) and $k_F$ is the Fermi wave vector. Finally, the band energy in respect to the Fermi level can be written as follows:

$$\omega(k) = \sqrt{\left(\frac{v^b_F(k - k_F)}{1 + \lambda}\right)^2 + \Delta^2}$$

(6.5)

The experimental band energy dispersion obtained as positions of the EDC maxima then can be fitted according to the procedure described above, using a fixed value for the bare Fermi velocity $v^b_F = 3$ eVÅ calculated using the tight-binding parameters from Ref. [74] and varying $\Delta$ and $\lambda$ in the fit.
In the superconducting state \((T_c = 30 \text{K})\) the values for \(\lambda\) were obtained by fits of \(\omega(k)\) given by Eq. 6.5 to the experimental EDC dispersions of the bonding band. This also gives the values of the superconducting gap \(\Delta\) equal to 30, 22 and 16 meV for UD77, OP89 and OD69 samples, correspondingly. The \(\lambda\) values for the same samples were as well derived from the ratio of bare Fermi velocity value \(v_F^b\) to the Fermi velocity \(v_F^{\text{ns}}\) in the superconducting state using Eq. 6.1. Both methods give comparable results as one can see in Fig. 6.12.

In the normal state, \((T = 120 \text{K})\) where steeper dispersions are observed for UD77 and OD81 samples, similar values of \(\lambda \approx 1.25\) were also derived from the ratio of the bare Fermi velocity value \(v_F^b\) to the normal state Fermi velocity \(v_F^{\text{ns}}\), obtained from the linear fit of the experimental MDC dispersion of the bonding band in the vicinity of the Fermi level (Fig. 6.9). These values also plotted in Fig. 6.12.

For the antibonding band reliable values for \(\lambda\) could be only obtained for an underdoped sample since there this band is found well below the Fermi level (Fig. 6.10). In this case the experimental EDC dispersion was fitted according to the described above procedure with \(\Delta\) and \(\lambda\) as free parameters and a bare dispersion calculated using the tight-binding parameters from Ref. [74]:

\[
\omega(k) = \sqrt{\left(\frac{\epsilon_{TB}(k)}{1 + \lambda}\right)^2 + \Delta^2}
\]  

By using this approach and the dataset shown in Fig. 6.6, the coupling strength constant \(\lambda\) of an antibonding band can be evaluated for different Fermi surface

Figure 6.9: Energy distribution maps and the bonding band dispersions derived from MDC maxima positions (blue points) together with the linear fit (solid red line) to obtain \(v_F^{\text{ns}}\).
Figure 6.10: Dispersion of the antibonding band near the antinodal point in an underdoped Pb-Bi-2212 sample at $T=30$K (left) and at $T=120$K (right) as derived from the EDCs (blue points) together with the fit (red solid line) and the bare electron dispersion (black points).

crossing positions $\vec{k}_F$ or radial $\vec{k}$-distances from the $(\pi,0)$ point (see Fig. 6.11). This momentum dependence clearly demonstrates a strongly anisotropic character of the coupling. In agreement with a previously observed results [85], the renormalization of the electronic bands dramatically increases on approaching the $(\pi,0)$ point.

Figure 6.11: Coupling strength constant $\lambda$ of the antibonding band near the $(\pi,0)$ point in the underdoped (Pb,Bi)2212 sample as a function of the radial momentum (blue points) together with a Gaussian fit (red solid line) For the nodal direction a value of the coupling strength constant $\lambda = 1$ was assumed.
6.2.3 Doping dependence of the mass renormalization

Fig. 6.12 summarize the results of the just described analysis of the coupling strength \( \lambda \) near the \((\pi, 0)\) point. With decreasing dopant concentration there is a strong increase of the coupling strength or mass renormalization parameter \( m^*/m = 1 + \lambda \) of about 3 in the overdoped to huge values of about 8 in the underdoped regime. It should be noted here that the increase seems to be continuous and there is no break observed between the over- and the underdoped region. For the antibonding band in the UD77 sample a similar huge mass renormalization has been detected (see Fig. 6.12). The dopant concentration was calculated from the well known empirical equation of \( T_c \) vs. the dopant concentration (see Ref. [31]):

\[
T_c/T_c^{max} = 1 - 82.6(p - 0.16)^2
\]  

where \( T_c^{max} \) is a maximum transition temperature and \( p \) is a hole concentration.

The results for the antibonding and the bonding band in the superconducting state can be also summarized in a different way. With decreasing dopant concentration the coherence factor \( Z=1/(1+\lambda) \) decreases from 0.25 to 0.1. This means that for underdoped samples below \( T_c \) only about 1/10 of the spectral weight near the Fermi level represents coherent states.

Above \( T_c \) we see dispersive states even in the underdoped samples and the renormalization of both bands is strongly reduced. There is also no 'kink' in the MDC dispersion at \( \sim 60 \) meV due to a coupling to a bosonic mode (see Fig. 6.9). The mode is either at much higher energies or there is a continuum which leads to the renormalization. The coupling constant in the normal state as derived from the differences in the slopes of the measured dispersion and the bare particle dispersion is independent of the dopant concentration. These values are also depicted in Fig. 6.12. The comparison of these values with similar \( \lambda \) values at the nodal point [84] indicates that the renormalization in the normal state is rather isotropic along the Fermi surface.

6.2.4 Scattering rate doping dependence

To obtain further information on the renormalization of the charge carriers in the normal state, we have evaluated the MDC width of the antibonding and the bonding bands which multiplied by \( v^b \), is a measure of the imaginary part of the self-energy plus contributions due to the finite energy and momentum resolution
Figure 6.12: The coupling strength parameter \( \lambda \) as a function of dopant concentration. Squares: superconducting state, circles: normal state. Open (solid) symbols: bonding (antibonding) band. The dashed lines are guides to the eye. The horizontal bar represents the experimental error in the dopant concentration.

(see chapter 4). Typical MDC cuts measured with a photon energy of 38 eV are shown in Fig. 6.13 for under- and overdoped samples. These cuts can be well fitted by four Lorentzians, two corresponding to the bonding and the other two corresponding to the antibonding band. For the overdoped and the underdoped samples the derived scattering rates at \( E_F \) are almost the same for both the bonding and the antibonding bands. Values between 80 and 160 meV are derived for the antinodal point at \( T = 120 \) K. These values are not far from values (\( \sim 50-100 \) meV) derived at the same temperature and at the same energy at the nodal point \([87]\). Moreover, at energies smaller than about 100 meV the scattering rates are only slightly dependent on energy (see Fig. 6.13). This is an indication that the modes to which the charge carriers are coupled, at higher energies or form a continuous spectrum.
6.2. Doping dependence of the coupling

The obtained results can be summarized in the following way: in the normal state a rather isotropic mass renormalization of the electronic states without a clear energy scale is observed. The strength of the coupling corresponds to $\lambda \sim 1$. Below $T_c$ and in the underdoped samples, in particular, a much higher strongly anisotropic coupling to a collective bosonic mode could be detected. It is emphasized that the difference in the mass renormalization at the nodal point between the superconducting and the normal state is rather small (less than 5%).

It is difficult to interpret the entirety of these results in terms of a conventional isotropic coupling to phonons. Probably a more promising model would be a coupling to spin fluctuations [88]. Although this scenario has been applied in many previous ARPES studies (see for review Ref. [14]) the present work provides a much more detailed picture.

\[
\Sigma''(\varepsilon) = \begin{cases} 
0.2 & \text{if } \varepsilon < 0.0 \\
0.1 & \text{if } 0.0 < \varepsilon < 0.1 \\
0.0 & \text{if } \varepsilon > 0.1 
\end{cases}
\]

\[
\text{Intensity (arb. units)} = \begin{cases} 
0.2 & \text{if } k_y < 0.1 \\
0.1 & \text{if } 0.1 < k_y < 0.2 \\
0.0 & \text{if } k_y > 0.2 
\end{cases}
\]

Figure 6.13: Upper row: imaginary part of self energy $\Sigma''$ with contributions from the momentum resolution as derived from MDC fits. Second row: MDC fits for OD and UD samples at energies of 15 and 35 meV, respectively.
Above $T_c$ the system has charge carriers which are coupled to a continuum of spin fluctuations. The strength of the coupling is not strongly dopant dependent. Both the antibonding and the bonding band feel a similar coupling as derived from the mass renormalization and from the scattering rates. Previous ARPES studies at the $(\pi, 0)$ point could not resolve the bilayer splitting and could not follow the flat dispersion of bands and therefore came to the conclusion that in the underdoped region above $T_c$ there are only incoherent states. In the present study, renormalized dispersive and possibly coherent states are even detected above $T_c$ in the underdoped samples.

In the spin fluctuation scenario, below $T_c$, the opening of the gap leads via a feed-back process to a magnetic resonance mode at $E_M$ detected by inelastic neutron scattering (INS) to which the charge carriers couple. The strong resonance found in INS experiments \cite{53} has energies $\Omega = 38-40$ meV, which exactly corresponds to the energy of the mode determined from the energetic separation between the peak and the 'dip' in intrinsic peak-dip-hump lineshape at $(\pi,0)$-point, and momentum $Q = (\pi, \pi)$, which corresponds to the distance between two parts of the Fermi surface near the $(0, \pi)$ and $(\pi, 0)$ points in the Brillouin zone, the so-called "hot spots" region (see Fig. 6.14). Also, the energy-integrated spectral weight of the resonance peak is increasing with under-doping.

Figure 6.14: Left: "hot spots" in the Bi2212 Brillouin zone; Middle: energy dependence of the INS resonance; Right: momentum dependence of the INS resonance (from \cite{53}).

Due to the fact that the bilayer splitting of the band could be resolved, a quantitative analysis of the coupling strength could be performed for the bonding band in the entire doping range and for the antibonding band for the underdoped
sample. In a recent theoretical work [89] it was pointed out that according to magnetic susceptibility measurements using inelastic neutron scattering the magnetic resonance mode couples the antibonding band only to the bonding band and vice versa. There is no coupling via the resonance mode within a band. It is remarkable that the coupling of the bonding band to the resonance mode starts when the antibonding band crosses the Fermi level (in the overdoped region). This is a further strong indication that there is a coupling to a mode only via odd susceptibilities and the spin fluctuation scenario is applicable. Moreover, the result that in the underdoped region $\lambda$ is similar for the both bands is understandable, since the Fermi velocities and therefore the density of states and the odd susceptibilities $\chi_{AB}$ and $\chi_{BA}$ should be comparable.
Chapter 7

Summary
The discovery of superconductivity in cuprates 15 years ago was not predicted by any theory. It implies that a completely novel approach is needed to explain the nature of the phenomena. The central question is: how can electrons move through the lattice without being scattered, what helps electrons to co-operate at low temperatures to carry the superconducting current? It is known that in the case of conventional superconductors electrons form pairs in order to overcome the resistance. The pair formation is driven by the coupling between electrons and phonons. In the high $T_c$ cuprates the nature of the force driving the pairing is of an unknown yet or controversial origin. A wealth of information related to this issue has been provided by angular resolved photoemission spectroscopy (ARPES), a leading technique in experimental determination of the single particle excitations in solids.

This thesis shows that the lineshape of these ARPES spectra can be understood in terms of a superposition of spectral features originating from the bilayer split Cu-O bands. By using a tunable excitation photon energy, the matrix element energy dependence was measured in detail for both bonding and antibonding bands. This gives an opportunity to study the electronic properties of these two bands separately. For the first time it was proved that superconducting gaps for both bands have the same value and symmetry. It also became possible to study the *pure* line shape for the different bands. For underdoped samples the *intrinsic* peak-dip-hump line shape, due to coupling with a bosonic mode, was observed. Studying the doping, temperature and momentum dependence of the photoemission spectra, a detailed inventory of the properties of the bosonic mode causing feedback effects in the electronic states was done. Considering the profile of the found 'fingerprints' of the mode such as: its energy is about 40 meV; it only causes strong self-energy effects in the superconducting state; the mode coupling is maximal around $(\pi, 0)$ in momentum space and, finally, its influence is strongly doping dependent, being greatly enhanced in the underdoped regime; it is clear that this mode has most likely to be identified with the sharp magnetic resonance mode observed in inelastic neutron scattering experiments.
Bibliography


[60] additional information about Gammadata Scienta instruments can be found in Internet: www.gammadata.se


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Publication list


Versicherung

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Timur Kim
Erklärung


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