

Chapter 2

Angle-Resolved Photoemission Spectroscopy

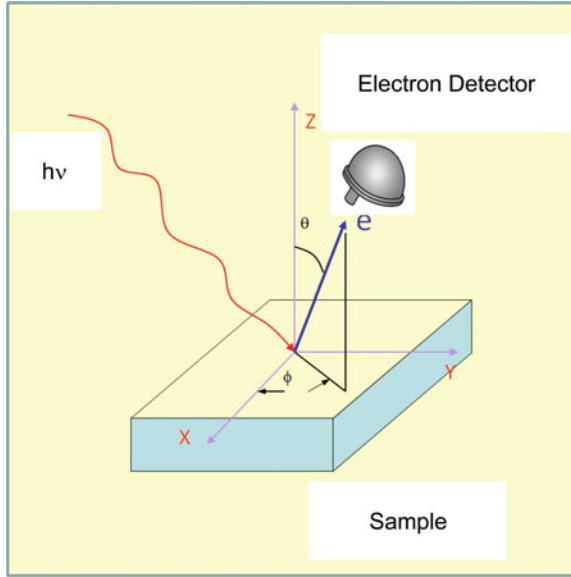
Angle-resolved photoemission spectroscopy (ARPES) is a powerful tool to directly probe the electronic structure of materials, which has been broadly used in the research of condensed matter physics. It plays an important role in unraveling the mechanism of many exotic phenomena in high-temperature superconductors, graphene, topological insulator, and many other advanced materials [1–6]. Generally speaking, ARPES measures the electronic structure of a material via photoelectrons. Shown in Fig. 2.1 is a sketch of an ARPES system containing the light source, sample, and analyzer.

In the history of photoemission, the first breakthrough was the observation of photoelectric effect in 1887 by Hertz [7]. It was then explained by Einstein in 1905 [8] with a simple equation describing the photoelectric effect:

$$E_{\max} = h\nu - \Phi. \quad (2.1)$$

in which $E_{\max}$ is the maximum kinetic energy of the emitted photoelectrons, $h\nu$ is the energy of the incident photon, and Φ is the sample work function. Continuous efforts have been made over a hundred years, which finally enable an accurate measurement of the energy and momentum of the photoelectrons. Spin-resolved photoemission spectroscopy has also been developed, which probes the spin of the electrons. Combining the information of energy, momentum, and spin, one can have a full description of the electronic states in the materials. The light source has also been improved over the decades. Besides the synchrotron radiation and gas discharge lamps, a newly developed laser source has induced significant improvement to the total resolution of the ARPES system by its monochromaticity and narrow line width. The works presented in this book are mainly based on ARPES. In this chapter, a general description of the ARPES technique will be given. In particular, it will explain how the energy and momentum of the electrons are resolved and what physical quantities are deduced from the ARPES spectra.

Fig. 2.1 Schematic of ARPES



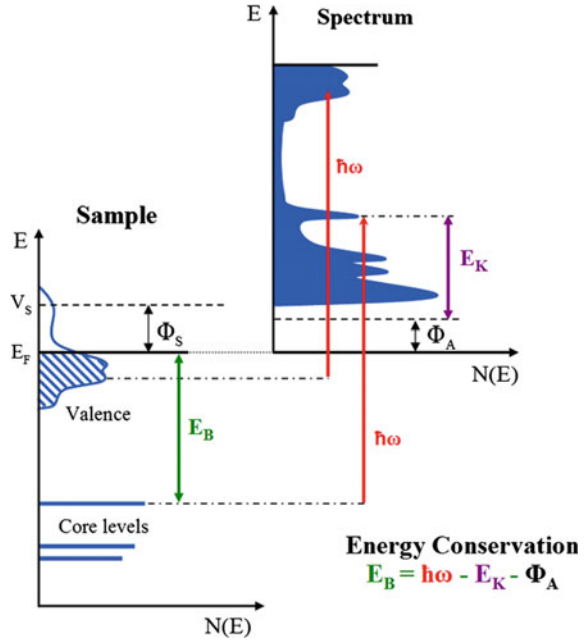
2.1 Energy Resolution of ARPES

Based on Einstein's equation for photoelectric effect, we have:

$$E_{kin} = h\nu - |E_B| - \Phi. \quad (2.2)$$

in which E_{kin} is the kinetic energy of the photoelectrons, $h\nu$ is the energy of the incident photon, Φ is the sample work function, and E_B is the binding energy of the initial states of the electrons in the material. For simplicity, fermi level of the material is always used as an energy reference and binding energy is therefore presented with negative values. If we only consider the energy resolution, then we can simply ignore the emission angles of the photoelectrons and calculate the electron density of states as a function of energy. Figure 2.2 illustrates how the density of states is probed by the photoemission process. According to the energy conservation law, electrons with different binding energy (e.g., valence bands vs. core levels) emit into the vacuum with different kinetic energies after absorbing photons with the same amount of energy. The kinetic energy of the photoelectrons is then detected by the electron analyzer. Shown on the top right of the figure is a typical photoemission spectrum in which the original shallow valence bands and deep core levels are presented as sharp peaks with different kinetic energies. Using Eq. 2.2, we can directly calculate the corresponding binding energy E_B of the original bands or core levels from the measured kinetic energy, which realizes the energy resolution. Nevertheless, according to Eq. 2.2, the correct analysis also depends on the correct determination of work function, which will be discussed in detail below. We also note that accurate analysis of the data

Fig. 2.2 Schematic of photoemission, in which E_F is the Fermi energy, E_B is the binding energy, Φ_S is the work function of the sample, Φ_A is the work function of the analyzer, and E_K is the electron kinetic energy detected by the analyzer [2]



will sometimes involve various factors, for example, the background given by the scattering of secondary electrons.

2.1.1 ARPES Measures the Occupied States

ARPES measures the photoelectrons which are ejected from the initial states to the final states after absorbing the incident photons. In other words, it measures the electronic states occupied by electrons but not the unoccupied states. To measure the unoccupied states, inverse photoemission is needed, in which low-energy electrons are directed at the sample surface and coupled with the unoccupied states. The occupied electrons to be measured by ARPES are governed by Fermi–Dirac distribution function. At zero temperature, if the energy resolution of the system is perfect, then the measured electronic states should show a sudden cutoff in the form of a step function as shown in Fig. 2.2. This is called Fermi cutoff, and the corresponding energy is Fermi energy, an energy reference for the binding energy of the occupied electronic states. In real experiments, the Fermi cutoff is broadened by the temperature and energy resolution of the measurements. On the one hand, we can take advantage of this broadening and calibrate the energy resolution of the system at a given temperature. On the other hand, we can also check the temperature of the sample by the Fermi cutoff broadening when the energy resolution is known.

2.1.2 The Selection of Work Function

Another important term included in Eq. 2.2 is work function. Work function is an energy barrier which prevents the electrons from leaving the materials. The existence of work function at the material surface is natural. Naively, if there is no work function, then electrons with the highest energy in the material can leave the material surface and get into the vacuum without losing any energy, and thus, no stable bulk material can ever exist. The correct selection of work function is very crucial for the ARPES measurement. As for the photoemission process alone, the work function refers to material work function which is the energy an photoelectron needs to pay if it moves from the Fermi level of the material to the vacuum. However, in the real measurement, the free electron-like photoelectrons are measured by the electron analyzer. The electric contact between the sample and the electron analyzer aligns their Fermi levels but not the work functions. Therefore, the measured kinetic energy is relative to the work function of the analyzer, which might be different from the real kinetic energy of the photoelectrons. In other words, if the work function of the material is different from that of the electron analyzer, then the photoelectrons are accelerated or decelerated in the analyzer. Nevertheless, the energy conservation law in Eq. 2.2 is still valid. In order to get the correct binding energy from the measured kinetic energy of the photoelectrons, we need to replace the sample work function with the work function of the electron analyzer.

This work function difference is always very small, and the acceleration or deceleration process is perpendicular to the sample surface and thus does not change the in-plane momentum of the photoelectrons. By considering the work function of the electron analyzer, this effect can be corrected. Nevertheless, we should notice that the real work function which takes place in the photoemission process is the material work function. Therefore, the lower limit of the photoelectron kinetic energy is determined by the material work function. This point has also been clearly illustrated in Fig. 2.2, in which Φ_S is the work function of the material and Φ_A is the work function of the electron analyzer. If $\Phi_S > \Phi_A$, in a certain energy range where the minimum kinetic energy of the photoelectrons E_K determined by Φ_A is larger than zero but $E_K + \Phi_A < \Phi_S$, no spectral weight is obtained in the electron analyzer. This is shown in Fig. 2.2 that an energy cutoff appears at V_S , below which the measured electron density is zero. This is simply because those electrons cannot leave the sample surface in the photoemission process and thus cannot be detected by the analyzer.

2.2 Momentum Resolution of ARPES

In the previous discussion, only energy conservation of the photoemission process was used. We notice that there is also momentum conservation in ARPES measurements. Correct analysis of the momentum conservation can give us the momentum

information of the electrons in the materials and thus realize the momentum resolution. Photoemission is a complicated process which involves the incident photons, photoelectrons, and the detection of photoelectrons. In order to accurately describe it, a so called one-step model is needed [9–12]. In this model, the absorption of photons, ejection of electrons, movement of the photoelectrons, and the detection are all considered as a coherent process and the properties of the bulk material, sample surface, and vacuum level are all included. Although accurate, this model is hard to be understood. Another phenomenology model is also proposed [13, 14], in which the photoemission is divided into three steps: (1) The ejection of electrons from initial states to final states. (2) Travel of the excited electrons from the bulk material to its surface. (3) Escape of the excited electrons from the sample surface into the vacuum. This model is called “three-step model.”

2.2.1 *Momentum Conservation in Step One: Ejection of Electrons from Initial to Final States*

This is the most important step in the three-step model, in which the essential information of the electron interactions is included. When incident light is directed on the sample surface, the electron of the material may absorb a photon and be ejected from the initial state to a final state. In typical ARPES measurements with soft X-rays, the photon energy is from several eV to ~ 100 eV and the corresponding momentum of the photons is negligible comparing to that of the electrons. Therefore, although the energy of the electron is changed by $h\nu$ in the ejection, the momentum of the electron conserves. Shown in Fig. 2.3 is an illustration of this process. After absorbing a photon with an energy of $h\nu$, the electron in the occupied initial state jumps vertically to a final state above Fermi level without changing its momentum. This is called “vertical transition.” In order to quantitatively describe this process, we study the momentum along x, y, and z directions, respectively.

If we label the electron momentum in the initial state as K_i and that of the final state as K_f , then we have:

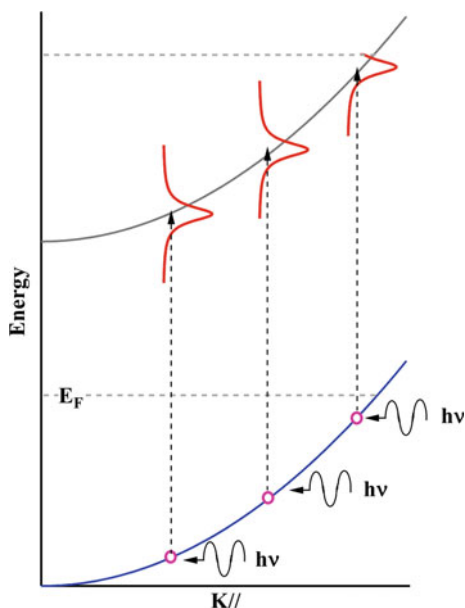
$$K_{f,x} = K_{i,x}; K_{f,y} = K_{i,y}; K_{f,z} = K_{i,z}. \quad (2.3)$$

As a comparison, the equation for the energy conservation in the first step is also listed here:

$$E_f = h\nu + E_B. \quad (2.4)$$

in which E_B is the electron binding energy which is the same as the electron energy in its initial state E_i .

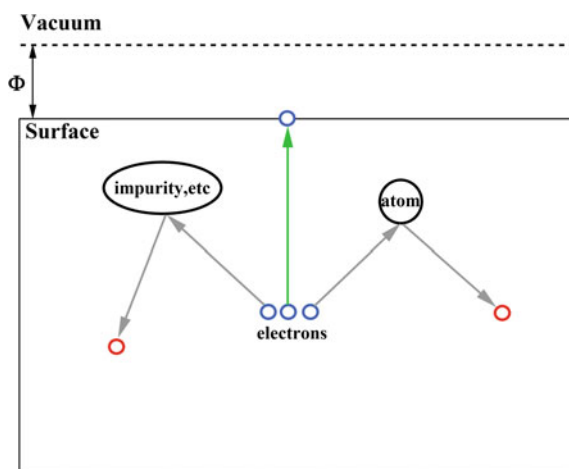
Fig. 2.3 The first step in the “three-step model,” the ejection of electrons from initial states to final states



2.2.2 Momentum Conservation in Step Two: Traveling of the Excited Electrons to Sample Surface

The second step of photoemission is the travel of the excited electrons from bulk material to the sample surface. As shown in Fig. 2.4, many inelastic-scattering processes are involved in this step. For example, the excited electrons might be scattered by

Fig. 2.4 The second step in the “three-step model,” travel of the excited electrons from the bulk material to its surface



other atoms or impurities. Secondary electrons might also be created in the process. However, only the electrons which are not scattered in their travel to the sample surface can contribute to the intrinsic electronic structure we measure. The following two points should be noticed: First, not all the excited electrons move toward the sample surface. Second, the scattered electrons might also travel to the sample surface and be detected, which do not represent the intrinsic electronic states in the material. However, the scattered electrons do not have a particular preference on the emission angle; therefore, they only contribute to some background in the measured photoemission intensity. The intrinsic signal comes from the excited electrons without any scattering. For those electrons, the momentum and energy do not change in step 2. If we define the corresponding momentum as K_{in} and energy as E_{in} , then we have:

$$K_{in,x} = K_{f,x}; K_{in,y} = K_{f,y}; K_{in,z} = K_{f,z}. \quad (2.5)$$

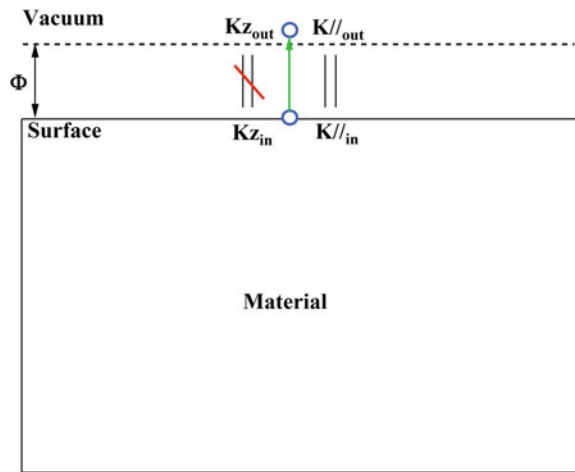
$$E_{in} = E_f. \quad (2.6)$$

2.2.3 Momentum Conservation in Step Three: Escape of the Excited Electrons into the Vacuum

In order to escape from the sample surface into the vacuum, the excited electrons need to go through the surface potential barrier, as shown in Fig. 2.5.

The energy of the potential barrier is the work function Φ of the material. In this step, the in-plane symmetry of the sample is not affected by the photoemission process. In other words, the in-plane momentum of the excited electrons keeps the same. On the other hand, the crystal symmetry is broken along the sample surface

Fig. 2.5 The third step in the “three-step model,” escape of the excited electrons from the sample surface into the vacuum



normal with the appearance of an interface between sample and vacuum. The excited electrons are coupled with the potential barrier perpendicular to the sample surface. Therefore, the momentum conservation law for the excited electrons along the sample surface normal is broken. Of course, the energy of the excited electrons is also changed due to the potential barrier. If we define the momentum of the electrons in the vacuum as K_{out} and the corresponding energy as E_{out} , then:

$$K_{out,x} = K_{in,x}; K_{out,y} = K_{in,y}; K_{out,z} \neq K_{in,z}. \quad (2.7)$$

$$E_{out} = E_{in} - \Phi. \quad (2.8)$$

If the materials are 2D or quasi-2D (for example, cuprate high-temperature superconductors, selected iron-based superconductors, thin films), only the in-plane momenta ($K_{i,x}$, $K_{i,y}$) are concerned. By using Eqs. 2.7, 2.5, and 2.3, we can easily get $K_{i,x}$, $K_{i,y}$ from $K_{out,x}$ and $K_{out,y}$ which are the momenta of the photoelectrons measured by the analyzer. Therefore, the key step is to calculate $K_{out,x}$ and $K_{out,y}$ from E_{out} and the corresponding electron emission angles, both of which are directly measured by the experiment. As for the photoelectron detected by the electron analyzer, the free electron-like plane wave is a sufficient description:

$$E_{out} = \frac{\hbar^2}{2m_e} (K_{out,x}^2 + K_{out,y}^2 + K_{out,z}^2). \quad (2.9)$$

For the electron analyzer with a horizontal slit, the momenta of the photoelectrons can be given as:

$$\begin{aligned} K_{out,x} &= \frac{1}{\hbar} \sqrt{2m_e E_{out}} \sin \Phi; K_{out,y} = \frac{1}{\hbar} \sqrt{2m_e E_{out}} \cos \Phi \sin \Theta; \\ K_{out,z} &= \frac{1}{\hbar} \sqrt{2m_e E_{out}} \cos \Phi \cos \Theta. \end{aligned} \quad (2.10)$$

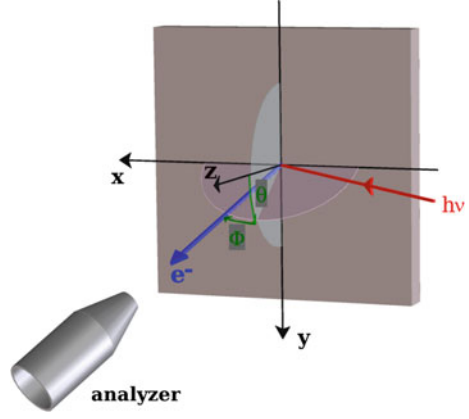
The corresponding emission angles are defined in Fig. 2.6.

Therefore,

$$K_{i,x} = \frac{1}{\hbar} \sqrt{2m_e E_{out}} \sin \Phi; K_{i,y} = \frac{1}{\hbar} \sqrt{2m_e E_{out}} \cos \Phi \sin \Theta. \quad (2.11)$$

As illustrated above, the momenta of the initial states $K_{i,x}$, $K_{i,y}$ can be obtained by the measured electron kinetic energy E_{out} and the corresponding electron emission angles, and thus, the momentum resolution can be achieved by ARPES.

Fig. 2.6 Different angles in the ARPES system. The momenta can be calculated from the angles [2]



2.2.4 Determination of K_z in 3D Systems [2, 15]

ARPES is broadly used to study 2D systems. Due to the breaking of momentum conservation along the sample surface normal, the measurement of K_z in 3D materials is not as straightforward as that of the in-plane momenta. $K_{i,z}$ of the initial state is not given by $K_{out,z}$ directly. However, we are still able to get the information of $K_{i,z}$ under some simple assumptions. Let us first consider the excited electrons after step one in the three-step model. Being ejected to the final states, those electrons can be assumed as quasi-free electrons and thus be described as:

$$E_f = \frac{\hbar^2}{2m_e} (K_{f,x}^2 + K_{f,y}^2 + K_{f,z}^2) + E_0. \quad (2.12)$$

Here, E_0 is called “bottom of the muffin tin” which represents the energy of the parabolic band bottom. Considering Eqs. 2.3, 2.6, and 2.8, we have:

$$E_{out} = \frac{\hbar^2}{2m_e} (K_{i,x}^2 + K_{i,y}^2 + K_{i,z}^2) + E_0 - \Phi. \quad (2.13)$$

Then,

$$K_{i,z} = \sqrt{\frac{2m_e}{\hbar^2} (E_{out} + \Phi - E_0) - K_{i,x}^2 - K_{i,y}^2}. \quad (2.14)$$

If we define $E_0 - \Phi$ as inner potential V_0 , then:

$$K_{i,z} = \sqrt{\frac{2m_e}{\hbar^2} (E_{out} - V_0) - K_{i,x}^2 - K_{i,y}^2}. \quad (2.15)$$

A simple way to determine V_0 is to measure normal emitted electrons whose in-plane momenta $K_{i,x} = K_{i,y} = 0$. By changing the photon energy of the incident beam, we can get different E_{out} values. We also know that $K_{i,z}$ should follow the periodicity of the 3D Brillouin zone. Therefore, we can get V_0 by fitting various attempted $K_{i,z}$ values (obtained with different photon energies) to the Brillouin zone boundary. Finally, the real $K_{i,z}$ can be directly calculated with the obtained V_0 .

2.3 Physical Properties Measured by ARPES [1, 2]

As explained in the previous sections that the energy and momentum resolutions of ARPES are realized by measuring the photoelectrons which are emitted from the material into the vacuum. However, electrons in most materials are correlated. For example, in a strongly correlated superconducting system, the electron–electron and electron–boson interactions are very important. A natural question for ARPES measurement is: How can we get the information of the many-body interactions in the materials by measuring the free electron-like excited photoelectrons. In this section, explanations will be given by introducing the single-particle spectral function.

2.3.1 Transition Probability for the Optical Excitation

Let us consider an optical excitation process in a system with N -electrons. Electrons are excited after absorbing photons, and the N -electron system goes from an initial state Ψ_i^N to a final state Ψ_f^N . The transition probability w_{fi} for this excitation process is given by Fermi's golden rule:

$$w_{fi} = \frac{2\pi}{\hbar} | \langle \Psi_f^N | H_{int} | \Psi_i^N \rangle |^2 \delta(E_f^N - E_i^N - \hbar\nu). \quad (2.16)$$

$E_i^N = E_i^{N-1} - E_B^k$ and $E_f^N = E_f^{N-1} + E_{kin}$ are the energy values for the initial states and final states, respectively. E_B^k is the original binding energy of the photoelectron whose kinetic energy is E_{kin} and momentum is k . The operator describing the interaction between electron and photon is given by

$$H_{int} = \frac{-e}{2mc} (\mathbf{A} \cdot \mathbf{p} + \mathbf{p} \cdot \mathbf{A}) = -\frac{e}{mc} \mathbf{A} \cdot \mathbf{p}, \quad (2.17)$$

under a perturbation theory. Here, $\mathbf{p}$ is the electronic momentum operator, and $\mathbf{A}$ is the electromagnetic vector potential. It seems that the many-body interactions included in these equations are too complicated to be calculated. In order to simplify this problem and obtain the essential physical properties of the materials, reasonable assumptions are needed.

2.3.1.1 Sudden Approximation and Adiabatic Limit

Although based on single-particle picture which is not ideal for the discussion of many-body interactions, the three-step model has achieved great success in practice. Electron transition in the first step of the model contains essential information of the many-body interactions. For simplicity, let us consider the excitation of one electron in the N-electron system. If the transition process is so fast that the excited electron does not interact with the rest N-1 electrons, then the question is simplified and the final state of the N-electron system can be written as:

$$\Psi_f^N = \mathcal{A} \phi_f^k \Psi_f^{N-1}. \quad (2.18)$$

in which $\mathcal{A}$ is an antisymmetric operator, ϕ_f^k is the wave function of the photoelectron, and Ψ_f^{N-1} is the final state wave function of the (N-1)-electron system which can be written as an eigenstate Ψ_m^{N-1} with energy E_m^{N-1} . Then, the total transition probability can be estimated by summing over m. This is called “sudden approximation” in which the photoemission process is assumed to be sudden [1]. This approximation works good for photoelectrons with high kinetic energy. However, if the kinetic energy of the photoelectron is very low that the time it takes to escape is comparable to the system response time, then the transition process cannot be regarded as sudden and the sudden approximation cannot be used any more. This is called “adiabatic limit” [16]. In this case, the final state wave function cannot be simplified in the above way and will be much more complicated. A practical concern is the lower limit of the photon energy by which the “sudden approximation” still works. This question is still under debate. It has been generally accepted that “sudden approximation” works for the light from helium lamp which provides photons with an energy of 21.2 eV. Experiments with a photon energy of 19 eV has also been reported in which “sudden approximation” also works good [17]. Other papers indicate that the “sudden approximation” is still OK with a photon energy of 6.05 eV [18, 19]. Recently, with the advantage of small band width and high photon flux, laser has been used as a new light source for ARPES which greatly improved the data quality. However, the photon energy of laser is relative low (~ 6 – 10 eV) which makes people worry about the solidity of “sudden approximation.” Fortunately, experimental evidence indicates that the “sudden approximation” still works good with laser light source.

2.3.1.2 Transition Probability Under Sudden Approximation

For simplicity, we write the initial state of the N-electron system Ψ_i^N as the combination of a one-electron wave function ϕ_i^k and an (N-1)-electron term Ψ_i^{N-1} :

$$\Psi_i^N = \mathcal{A} \phi_i^k \Psi_i^{N-1}, \quad (2.19)$$

where $\Psi_i^{N-1} = c_k \Psi_i^N$, and c_k is the annihilation operator for an electron with momentum k . Here, Ψ_i^{N-1} is not an eigenstate of the (N-1)-electron system. It is just the remaining part of the N-electron wave function after removing one electron. Therefore, the matrix elements in Eq. 2.16 can be written as:

$$\langle \Psi_f^N | H_{int} | \Psi_i^N \rangle = \langle \phi_f^k | H_{int} | \phi_i^k \rangle \langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle, \quad (2.20)$$

in which $\langle \phi_f^k | H_{int} | \phi_i^k \rangle = M_{f,i}^k$ is called one-electron dipole matrix element. The final state of the (N-1)-electron system Ψ_f^{N-1} is an eigenstate and thus can be replaced by Ψ_m^{N-1} . Then, the photoemission intensity $I(k, E_{kin})$ can be written as:

$$I(k, E_{kin}) = \sum_{f,i} w_{f,i} \propto \sum_{f,i} |M_{f,i}^k|^2 \sum_m |c_{m,i}|^2 \delta(E_{kin} + E_m^{N-1} - E_i^N - h\nu), \quad (2.21)$$

in which $|c_{m,i}|^2 = |\langle \Psi_m^{N-1} | \Psi_i^{N-1} \rangle|^2$ represents the probability that the remaining (N-1)-electron system is at the eigenstate m after removing one electron from the i state. It includes the fundamental information of the transition.

In order to understand the transition probability, let us first consider a non-interacting electron system. In this case, the removal of one electron in the N-electron system does not have any effect on the rest electrons, which means the Ψ_i^{N-1} state is still an eigenstate $\Psi_{m_0}^{N-1}$ of the $(N-1)$ -electron system. Therefore, in the equation of photoemission intensity $I(k, E_{kin})$, only $|c_{m_0,i}|^2 = 1$ (when $m = m_0$) and all the other $c_{m,i}$ terms are zero. Considering the δ function part in $I(k, E_{kin})$, we should expect ARPES spectra with δ function peaks at $E_B^k = -\epsilon_k$. However, in correlated systems, the removal of one electron gives rise to a dramatic change to the rest (N-1)-electron system. Therefore, many terms of $|c_{m,i}|^2 \neq 0$ and Ψ_i^{N-1} will overlap with many of the eigenstates Ψ_m^{N-1} . The corresponding ARPES spectra will not show single delta functions any more. Instead, a main line with several satellites is expected (Fig. 2.7).

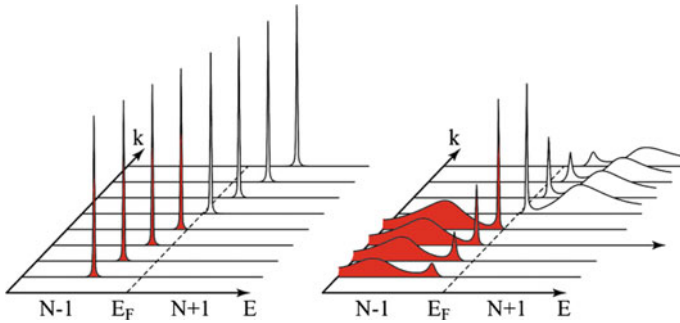


Fig. 2.7 Left panel: ARPES spectra for a non-interacting electron system. Right panel: ARPES spectra for correlated systems. The filled red regions represent the occupied states, whereas the empty regions represent the unoccupied states [2]

2.3.2 Single-Particle Spectral Function

Green's function has been widely used in solid physics to describe many-body effects. The propagation of a single electron can be written as a time-ordered one-electron Green's function $g(t - t')$ which describes the probability amplitude that an electron added at a time of zero keeps in the same state after a time of $|t - t'|$. An energy-momentum representation can be given by performing a Fourier transform:

$$G^{\pm}(k, \omega) = \sum_m \frac{|\langle \Psi_m^{N\pm 1} | c_k^{\pm} | \Psi_i^N \rangle|^2}{\omega - E_m^{N\pm 1} + E_i^N \pm i\eta}, \quad (2.22)$$

in which $G^+(k, \omega)$ and $G^-(k, \omega)$ represent the Green's function for adding and removing a single electron, respectively. The single-particle spectral function can thus be given by the imaginary part of the Green's function:

$$A(k, \omega) = A^+(k, \omega) + A^-(k, \omega) = -(1/\pi) \text{Im} G(k, \omega); \quad (2.23)$$

$$A^{\pm}(k, \omega) = \sum_m |\langle \Psi_m^{N\pm 1} | c_k^{\pm} | \Psi_i^N \rangle|^2 \delta(\omega - E_m^{N\pm 1} + E_i^N). \quad (2.24)$$

By comparing the single-particle spectral function $A^-(k, \omega)$ with the photoemission intensity $I(k, E_{kin})$ (Eq. 2.21), we have:

$$I(k, E_{kin}) \propto \sum_{f,i} |M_{f,i}^k|^2 A^-(k, \omega). \quad (2.25)$$

Therefore, it is clear that the photoemission intensity measured by ARPES is proportional to the single-particle spectral function which explains the reason why the energy gap, Fermi surface, band structures, and many-body effects in the materials can be captured by the ARPES spectra.

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