

Do angle-resolved photoemission experiments reflect the nature of an interacting many-body system?

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Angle-resolved photoemission spectroscopy (ARPES) provides us with the most direct access to the energy and momentum of the occupied electronic states. In this review we want to answer the following basic question: What are we actually measuring in angle resolved photoemission? To answer this question we first introduce the idea of single-particle spectral function. Analyzing the expression of the intensity measured by ARPES we can show that the intensity corresponds to the electron removal part of the spectral function modulated by transition matrix elements. To test our conclusion we present two classic experiments that probe a Fermi liquid and an electron-phonon system.

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I. INTRODUCTION

The great popularity of photoemission in the study of solids can be mainly attributed to the capacity of this technique to yield direct access to the energy and momentum of the occupied electronic states. In this review we shall examine the standard interpretation of angle resolved photoemission (ARPES) data from a theoretical point of view. The first section is devoted to the idea of single-particle spectral function. Then basics of ARPES measurement and an analysis of the measured intensity are introduced. The next section describes an estimation of the self-energy for a Fermi liquid system and a classic ARPES measurement on layered Fermi liquid system $1 - T - \text{TiTe}_2$. We conclude with an analysis of the self-energy of an electron-phonon system and how we can isolate the electron-phonon contribution from other interactions. It followed by a careful experiment done on a nearly free electron like surface state of $\text{Be}(0001)$ that showed an excellent agreement with the calculated line shapes and measured spectra.

II. SPECTRAL REPRESENTATION

One of the most powerful tool available to a many-body physicist is the single-particle spectral function. The basic idea is similar to the spectral decomposition of a time dependent $f(t)$ into the sum of it's components at various frequencies.

$$f(t) = \int_{-\infty}^{\infty} F(\omega) e^{i\omega t} d\omega \quad (1)$$

where $F(\omega)$ gives the spectrum of $f(t)$.

Let us try to evaluate a similar form of decomposition for the propagator $\mathcal{G}(\mathbf{k}, \omega)$. Let $|\psi_n^N\rangle$ and E_n^N be the exact eigenstate and energies of the Hamiltonian H of the interacting N particle system. The single particle propagator or Green function can be defined as

$$\mathcal{G}(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1) = G^+(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1)_{t_2 > t_1} + G^-(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1)_{t_2 \leq t_1}. \quad (2)$$

where $G^-(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1)$ is the probability amplitude that if at time t_1 we remove a particle in $\phi_{\mathbf{k}}$, from the interacting system in it's ground state, then at time t_2 the system will be in it's ground state with an added hole in $\phi_{\mathbf{k}}$.

$G^-(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1)$ can be expressed in terms of creation and annihilation operators.

$$\mathcal{G}(\mathbf{k}_2, \mathbf{k}_1, t_2 - t_1) = -i\Theta(t_2 - t_1) \langle \psi_0 | \hat{c}_{\mathbf{k}_2}(t_2) \hat{c}_{\mathbf{k}_1}^\dagger(t_1) | \psi_0 \rangle + i\Theta(t_1 - t_2) \langle \psi_0 | \hat{c}_{\mathbf{k}_1}^\dagger(t_1) \hat{c}_{\mathbf{k}_2}(t_2) | \psi_0 \rangle. \quad (3)$$

Let $t_1 = 0$ and $t_2 = t$. Then

$$\begin{aligned} G^-(\mathbf{k}, t) &= i\Theta(-t) \\ &\times \sum_n \langle \psi_0^N | \hat{c}_{\mathbf{k}}^\dagger e^{iHt} | \psi_n^{N-1} \rangle \langle \psi_n^{N-1} | \hat{c}_{\mathbf{k}} e^{-iHt} | \psi_0^N \rangle \\ &= i\Theta(-t) \sum_n |\langle \psi_n^{N-1} | c_{\mathbf{k}} | \psi_0^N \rangle|^2 e^{-i(E_0^N - E_n^{N-1})t} \\ &= i\Theta(-t) \sum_n |(c_{\mathbf{k}})_{n0}|^2 e^{-i(E_0^N - E_n^{N-1})t} \end{aligned} \quad (4)$$

Taking Fourier transform of Eq. (4) we find

$$G^-(\mathbf{k}, \omega) = \sum_n |(c_{\mathbf{k}})_{n0}|^2 \frac{1}{\omega - (E_0^N - E_n^{N-1}) - i\delta} \quad (5)$$

For large N , these results can be expressed in terms of chemical potential, $E_0^N - E_n^{N-1} = E_0^N - E_0^{N-1} + E_0^{N-1} -$

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$E_n^{N-1} = \mu^{N-1} - \omega_{n0}^{N-1} = \mu - \omega_{n0}$. This gives

$$G^-(\mathbf{k}, t) = i\Theta(-t) \sum_n |(c_{\mathbf{k}})_{n0}|^2 e^{-i(\mu - \omega_{n0})t} \quad (6)$$

and

$$G^-(\mathbf{k}, \omega) = \sum_n |(c_{\mathbf{k}})_{n0}|^2 \frac{1}{\omega - (\mu - \omega_{n0}) - i\delta} \quad (7)$$

In a system with large volume, the energy levels are so closely spaced that we can go from a sum to an integral by introducing the *single-particle spectral function*

$$A^-(\mathbf{k}, \omega) = \sum_{\omega < \omega_{n0} < \omega + d\omega} |(c_{\mathbf{k}})_{n0}|^2 \quad (8)$$

or, equivalently,

$$A^-(\mathbf{k}, \omega) = \sum_n |(c_{\mathbf{k}})_{n0}|^2 \delta(\omega - \omega_{n0}). \quad (9)$$

This function is defined only for $\omega \geq 0$. It gives the probability that the state $|\psi_0^N\rangle$ with an added hole in state \mathbf{k} is an exact eigenstate of the $(N-1)$ -particle system with energy between ω and $\omega + d\omega$. Substituting Eq. (8) in Eq. (6) and (7), we get

$$G^-(\mathbf{k}, t) = i\Theta(-t) \int_0^\infty d\omega A^-(\mathbf{k}, \omega) e^{-i(\mu - \omega_{n0})t} \quad (10)$$

$$G^-(\mathbf{k}, \omega) = \int_0^\infty d\omega' \frac{A^-(\mathbf{k}, \omega')}{\omega - (\mu - \omega') - i\delta} \quad (11)$$

The *retarded Green function* is defined as

$$G(\mathbf{k}, \omega) = G^+(\mathbf{k}, \omega) + [G^-(\mathbf{k}, \omega)]^*.$$

Hence, in terms of single-particle spectral function

$$G(\mathbf{k}, \omega) = \int_0^\infty d\omega' \frac{A^+(\mathbf{k}, \omega')}{\omega - \omega' - \mu + i\delta} + \frac{A^-(\mathbf{k}, \omega')}{\omega + \omega' - \mu - i\delta} \quad (12)$$

Comparing Eq. (1) with (12) we can see why $A(\mathbf{k}, \omega)$ is also known as spectral density function.

We can make use of the following identity

$$\frac{1}{x + i\delta} = \mathcal{P} \left(\frac{1}{x} \right) - i\pi\delta(x)$$

to get

$$A(\mathbf{k}, \omega) = A^+(\mathbf{k}, \omega) + A^-(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im}G(\mathbf{k}, \omega). \quad (13)$$

The effect of electron-electron correlation to the Green function can be expressed in terms of the electron *proper self energy*

$$\Sigma(\mathbf{k}, \omega) = \text{Re}\Sigma(\mathbf{k}, \omega) + i\text{Im}\Sigma(\mathbf{k}, \omega).$$

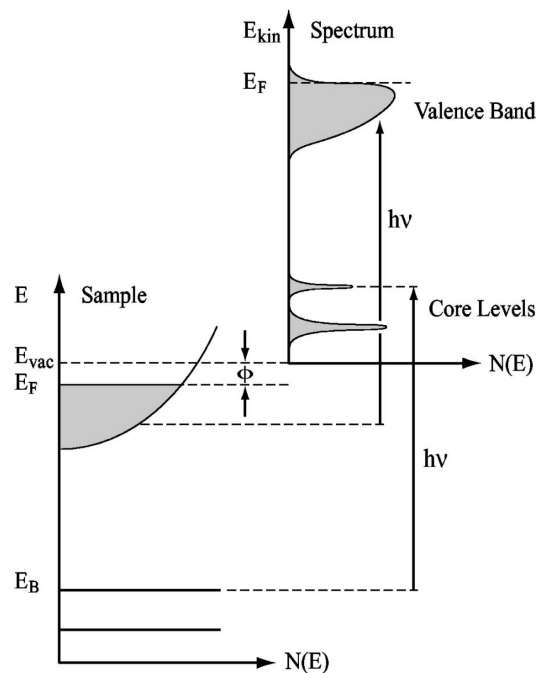


FIG. 1 Energetics of the photoemission process. From (1)

Its real and imaginary parts contain all the information on the energy renormalization and lifetime, respectively, of an electron with band energy $\epsilon_{\mathbf{k}}$ and momentum \mathbf{k} propagating in a many-body system. The Green function can be expressed in terms of self-energy as

$$G(\mathbf{k}, \omega) = \frac{1}{\omega - \epsilon_{\mathbf{k}} - \Sigma(\mathbf{k}, \omega)}. \quad (14)$$

Since $A(\mathbf{k}, \omega) = -\frac{1}{\pi} \text{Im}G(\mathbf{k}, \omega)$,

$$A(\mathbf{k}, \omega) = -\frac{1}{\pi} \frac{\text{Im}\Sigma(\mathbf{k}, \omega)}{[\omega - \epsilon_{\mathbf{k}} - \text{Re}\Sigma(\mathbf{k}, \omega)]^2 + [\text{Im}\Sigma(\mathbf{k}, \omega)]^2}. \quad (15)$$

It is important to notice the profound changes that take place when we go from the sum (7) to the integral (11). The sum (7) has an infinite number of real poles, whereas the integral (11) has a small number of complex poles. We should also note the physical meaning of the corresponding expressions for G^- in the time domain, i.e., (6) and (10). In (6), if there is no interaction, then $(c_{\mathbf{k}})_{n0} = \delta_{kn}$, i.e., $(c_{\mathbf{k}})_{n0}$ is finite only for a single energy level. But with interaction, in typical cases $(c_{\mathbf{k}})_{n0}$ is spread out over a band of energy levels from say n' to n'' , having width $\Delta E = \omega_{n'',0} - \omega_{n',0}$.

III. A GENERAL DESCRIPTION OF ARPES

A. Kinematics of photoemission

When light is incident on a sample, an electron can absorb a photon and escape from the material with a

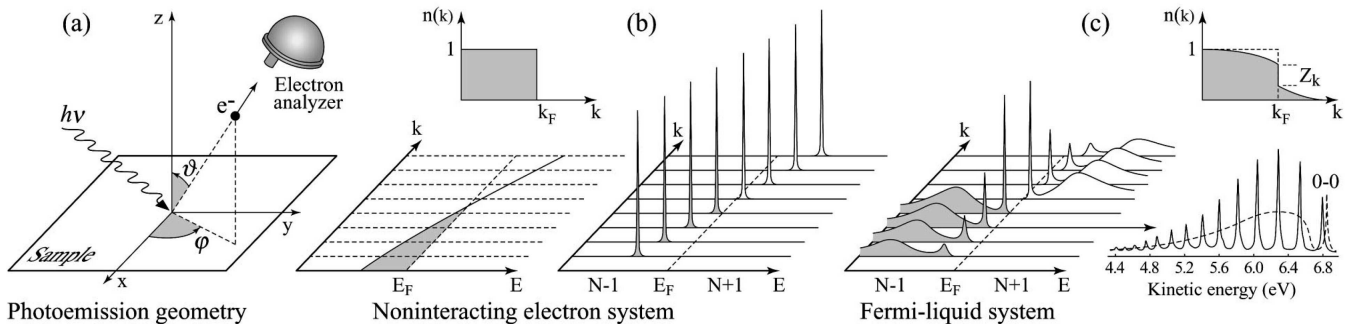


FIG. 2 (a) geometry of an ARPES experiment, (b) momentum-resolved one electron removal and addition spectra for a non-interacting electron system with a single energy band dispersing across E_F , (c) same spectra for an interacting Fermi-liquid system, (d) photoemission spectrum for gaseous system. From (2).

maximum kinetic energy of $h\nu - \phi$ where ν is the frequency of the incident photon and ϕ is the work function of the metal.

The energetics of the photoemission process is sketched in Fig. 1.

In an angle-resolved photoemission experiment, a beam of monochromatic radiation is incident on a sample and, as a result, electrons are emitted by the photoelectric effect and escape into the vacuum in all directions. By collecting photoelectrons with an electron energy analyzer we measure the kinetic energy, E_{kin} of the photoelectrons for a given emission angle. This way photoelectrons momentum \mathbf{p} is also completely determined

$$|\mathbf{p}| = p = \sqrt{2mE_{kin}} \quad (16)$$

and p_{\parallel} and p_{\perp} are determined from the polar (θ) and azimuthal (ϕ) emission angles.

Photon momentum can be neglected at low photon energies typically used in ARPES experiments. Therefore, within the non-interacting electron picture, due to energy and momentum conservation laws, we get the following relations

$$E_{kin} = h\nu - \phi - |E_B| \quad (17)$$

$$p_{\parallel} = \hbar k_{\parallel} = \sqrt{2mE_{kin}} \cdot \sin \theta \quad (18)$$

B. Photoemission intensity

To describe the photoemission process, we can start with how to calculate the transition probability w_{n0} for the optical excitation between the N -electron ground state $|\psi_0^N\rangle$ and one of the possible final state $|\psi_n^N\rangle$. This can be approximated by Fermi's golden rule

$$w_{n0} = \frac{2\pi}{\hbar} |\langle \psi_n^N | H_{int} | \psi_0^N \rangle|^2 \delta(E_n^N - E_0^N - h\nu) \quad (19)$$

where $E_0^N = E_0^{N-1} - E_B^k$ and $E_n^N = E_n^{N-1} + E_{kin}$ 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 of

the the electromagnetic wave, i.e. photon, can be treated as a perturbation given by

$$H_{int} = \frac{e}{2mc} (\mathbf{A} \cdot \mathbf{p} + \mathbf{p} \cdot \mathbf{A}) = \frac{e}{mc} \mathbf{A} \cdot \mathbf{p} \quad (20)$$

where \mathbf{p} is the electronic momentum operator and \mathbf{A} is the electromagnetic vector potential.

The standard model of approximating the photoemission process is known as three-step model. Within this approach, the photoemission process is divided into three independent and sequential steps

- (i) optical excitation of the electron to the bulk,
- (ii) travel of the excited electron to the surface and
- (iii) escape of the photoelectron into vacuum.

The total photoemission intensity is then given by the product of the probabilities of these three independent processes.

In evaluating (i), and therefore, the photoemission intensity in terms of the transition probability w_{n0} , it would be convenient to factorize the wavefunctions in Eq. (19) into photoelectron and $(N-1)$ -electron terms. But doing this is not simple because the system will relax. The problems is simplified within the *sudden approximation* which applies to high kinetic energy electrons. In this limit, the photoemission process is assumed to be sudden, with no post-collisional interaction between the photoelectron and the system left behind. Then the final state ψ_n^N can be written as

$$\psi_n^N = \mathcal{A} \phi_n^{\mathbf{k}} \psi_n^{N-1} \quad (21)$$

where \mathcal{A} is an antisymmetric operator that properly antisymmetrizes the N -electron wavefunction so that Pauli principle is satisfied, $\phi_n^{\mathbf{k}}$ is the wavefunction of the photoelectron with momentum \mathbf{k} , and ψ_n^{N-1} is the final state wavefunction of the $(N-1)$ -electron system left behind which can be chosen as an excited state with energy E_n^{N-1} . The total transition probability is then given by the sum over all possible excited states n .

Let us also write the initial state as the product of a one-electron orbital $\phi_0^{\mathbf{k}}$ and an $(N-1)$ -particle term

$$\psi_0^N = \mathcal{A} \phi_0^{\mathbf{k}} \psi_0^{N-1}. \quad (22)$$

However, ψ_0^{N-1} can be expressed as

$$\psi_0^{N-1} = c_{\mathbf{k}} \psi_0^N.$$

At this point we can write the matrix element of Eq. (19) as

$$\langle \psi_n^N | H_{int} | \psi_0^N \rangle = \langle \phi_n^{\mathbf{k}} | H_{int} | \phi_0^{\mathbf{k}} \rangle \langle \psi_m^{N-1} | \psi_0^{N-1} \rangle \quad (23)$$

where $\langle \phi_n^{\mathbf{k}} | H_{int} | \phi_0^{\mathbf{k}} \rangle \equiv M_{n,0}^{\mathbf{k}}$ is the one-electron dipole matrix element, and the second term is the $(N-1)$ -electron overlap integral. The total photoemission intensity measured as a function of E_{kin} at a momentum \mathbf{k} is

$$I(\mathbf{k}, E_{kin}) = \sum_{n,0} w_{n,0} \quad (24)$$

$$\propto \sum_{n,0} |M_{n,0}^{\mathbf{k}}|^2 \left(\sum_m |c_{m,i}|^2 \delta(E_{kin} + E_m^{N-1} - E_0^n - h\nu) \right)$$

where $|c_{m,i}|^2 = |\langle \psi_m^{N-1} | \psi_0^{N-1} \rangle|^2$ is the probability that the removal of an electron from state 0 will leave the $(N-1)$ -particle system in the excited state m .

Comparing the term inside the parenthesis of Eq. (24) with Eq. (9), we see that we are actually measuring the electron removal part of the spectral function in an ARPES spectrum

$$I(\mathbf{k}, E_{kin}) = \sum_{n,0} |M_{n,0}^{\mathbf{k}}|^2 A^-(\mathbf{k}, \omega) \quad (25)$$

So far we have only discussed about direct photoemission (photon in, electron out). If we repeat the whole calculation starting from $G^+(\mathbf{k}, t)$ we can show that inverse photoemission (electron in, photon out) measures $A^+(\mathbf{k}, \omega)$. Therefore, full $A(\mathbf{k}, \omega)$ can be available from a combination of direct and inverse photoemission. Also $G(\mathbf{k}, t, t')$ is a linear response function to an external perturbation. Therefore, the real and imaginary parts of its Fourier transform $G(\mathbf{k}, \omega)$ have to satisfy causality and hence related by Kramers-Kronig relations. This implies that if we have the full $A^+(\mathbf{k}, \omega)$ available from direct and inverse photoemission, we can calculate $\text{Re}G(\mathbf{k}, \omega)$ and then obtain the full self energy directly from Eq. (14). This really tells us that ARPES should, in principle, be able to contain the information about the interactions in a many-body system.

Now, in our photoemission intensity analysis, we have only considered the $T = 0$ case. In $T \neq 0$ case, we can similarly invoke the sudden approximation and write the intensity measured in an ARPES experiment as

$$I(\mathbf{k}, E_{kin}) = \sum_{n,0} |M_{n,0}^{\mathbf{k}}|^2 A^-(\mathbf{k}, \omega) f(\omega), \quad (26)$$

where $f(\omega)$ is the Fermi function.

IV. FERMI-LIQUID LINE SHAPES MEASURED BY ARPES

We have already shown that apart from the modulation by the matrix elements, ARPES measures essentially

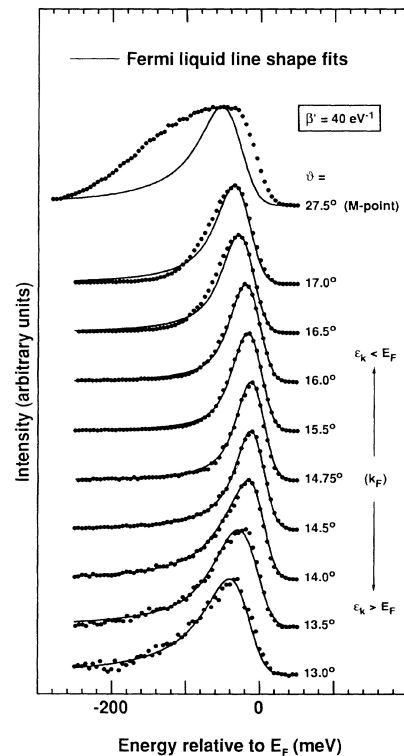


FIG. 3 ARPES spectra of $1-T$ - TiTe_2 taken along the $\Gamma-M$ direction. From (4).

the spectral function. However, in real life systems there are many other factors that can alter ARPES line shape. For example, the lifetime of the photoelectron adds to the total ARPES linewidth. Now, it has been shown that (4) the final state energy width is mixed in with a weight factor of $v_{h\perp}/v_{e\perp}$, where $v_{h\perp}$ and $v_{e\perp}$ are the band velocities, perpendicular to the surface, of the photohole and photoelectron, respectively. Therefore, the effect of the final electron state broadening can be suppressed if $v_{h\perp} \ll v_{e\perp}$ (3). This is why detailed photohole line-shape studies can only be done on surface states or layered systems like high T_c superconductors. Other notable mechanisms that can distort the spectral weight information are scattering and diffraction of the outgoing electrons. Only if all these effects are negligible, or if their energy dependence is small on the scale of the intrinsic linewidths, can the ARPES signal be taken to be representative of the electron removal spectrum.

In the case where the low-lying excitations can be well described by quasiparticles, i.e., in a Fermi liquid, the self-energy near the Fermi energy is well known (5):

$$\Sigma(\mathbf{k}, \omega) = \alpha\omega + i\beta\omega^2, \quad (27)$$

where we have assumed a three dimensional isotropic electron system. It is very important to note that this is an approximation that holds only for energies small compared to the occupied bandwidth ϵ_F . Using this self-

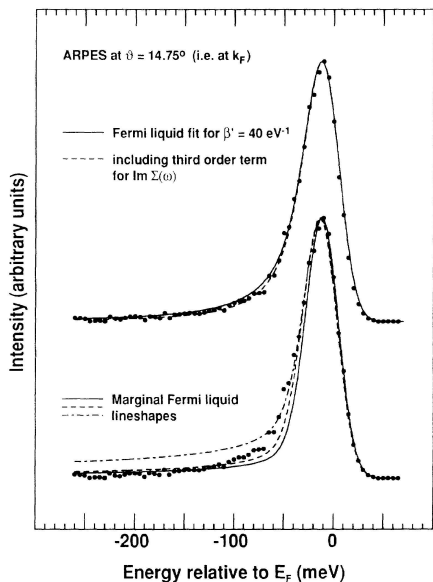


FIG. 4 Upper part: ARPES spectra fitted with Fermi liquid line profile calculated using (28). Lower part: same data fitted with line shapes of the marginal fermi liquid for various model parameters. From (4).

energy the Fermi liquid spectral function is given by

$$A(\mathbf{k}, \omega) \propto \frac{\beta' \omega^2}{(\omega - \epsilon'_{\mathbf{k}})^2 + \beta'^2 \omega^4}, \quad (28)$$

where $\epsilon' = Z_F \epsilon_{\mathbf{k}}$, $\beta' = Z_F \beta$, and Z_F is the quasiparticle weight factor or wave-function renormalization constant on the Fermi surface, given by $(1 - \alpha)^{-1}$. In this approximation, the spectral function depends on momentum \mathbf{k} only through $\epsilon_{\mathbf{k}}$.

The classic experiment to show to what extent ARPES spectra can reflect the nature of a Fermi liquid system has been done by Claessen et. al. (4). The measurements were done on $1 - T - \text{TiTe}_2$ which, according to band structure calculations, is semimetal.

Figure 3 shows the measured ARPES spectra fitted with modelled line shapes by using expression (28) for the Fermi liquid self-energy. To fit the theoretical line profiles to the data they used the parameters β' and $\epsilon'_{\mathbf{k}}$, where β' was kept fixed for all spectra and $\epsilon'_{\mathbf{k}}$ was varied for each spectrum to obtain the correct peak position. Because (27) is an approximation for $\epsilon'_{\mathbf{k}} \ll \epsilon_F$ the best agreement is to be expected for the spectrum at $\theta = 14.75^\circ$ corresponding to the Fermi level crossing, and indeed they found an excellent correspondence. As θ was increased, very good fit was obtained up to 16° . As θ increases after that 28 fails to describe the data due to the eventual breakdown of the self-energy approximation 27 since $\epsilon'_{\mathbf{k}}$ cannot be considered small in comparison to ϵ_F .

Figure 4 shows that the excellent quality of the fits are really unique to the use of Fermi liquid line shapes. Attempts to fit with other line profiles, e.g., that of the

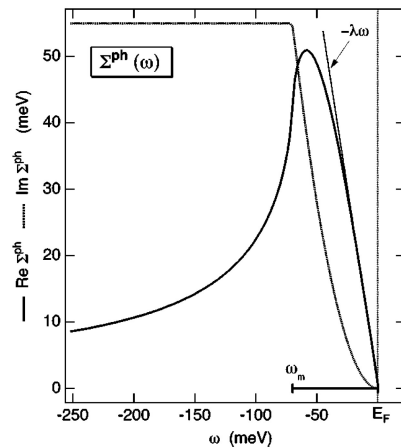


FIG. 5 Plot of the electron-phonon part of the self-energy used in (7).

marginal Fermi liquid (6), failed to reproduce the experimental spectra. The Fermi liquid fits yield for β' a value of $40 \pm 5 \text{ eV}^{-1}$ which is in good agreement with the calculated value.

V. ELECTRON-PHONON COUPLING IN ARPES

To lowest order in the various interactions, the different contributions are simply summed up (5):

$$\Sigma = \Sigma^{ph} + \Sigma^{el-el} + \Sigma^{imp}. \quad (29)$$

For a nearly free electron like surface state of $\text{Be}(0001)$ (7) we may try to isolate the interesting electron-phonon term Σ^{ph} in the following way. First, the impurity term is purely imaginary and nearly constant in the small energy range of interest. Second, the real part of the electron-electron term can be linearized close to the Fermi level on the scale of the electron bandwidth (4), leading to a constant change in the Fermi velocity v_F . In order to implement the imaginary parts, a parameter

$$\Delta(k) = \text{Im} \Sigma^{el-el} + \text{Im} \Sigma^{imp}$$

can be introduced which is used in the analysis as a fitting parameter of the spectral function (7). Eq. (29) leads then to

$$\Sigma(\mathbf{k}, \omega) = \Sigma^{ph}(\mathbf{k}, \omega) + i\Delta(\mathbf{k}). \quad (30)$$

The standard way to express this is the following

$$\Sigma^{ph}(\omega) = \int_{-E_F}^{\infty} d\epsilon \int_0^{\omega_m} d\tilde{\omega} \alpha^2 F(\tilde{\omega}) \frac{1 - f(\epsilon, t) + N(\tilde{\omega}, T)}{\omega - \epsilon - \tilde{\omega} + i\delta^\pm} + \frac{f(\epsilon, t) + N(\tilde{\omega}, T)}{\omega - \epsilon + \tilde{\omega} + i\delta^\pm}, \quad (31)$$

where $f(\epsilon, t)$ and $N(\omega, T)$ are the Fermi-Dirac and Bose-Einstein factors, respectively and δ is an infinitesimal number.

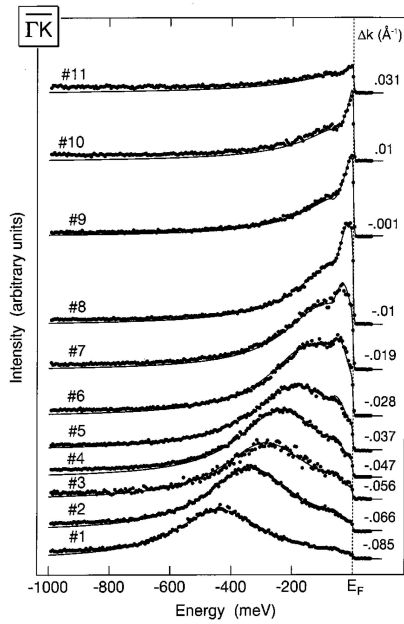


FIG. 6 ARPES of the surface state near E_F in the direction $\Gamma\bar{K}$ (dots), compared to the spectral functions, calculated for the corresponding emission angles (lines). From (7).

Figure 6 shows the estimated line shapes superimposed on the measured ones. The calculation reproduces almost

perfectly the double structure and the intensity ratio between the two main peaks.

VI. CONCLUSION

From the discussion it is evident that ARPES does reflect the signatures of many-body interactions present in the system but the information we are interested in can be mixed with other factors in a complicated way. Therefore one must be very careful in choosing a system to do ARPES on and know how the other factors are contributing to the spectra before extracting any information out.

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