

Angle-resolved photoemission at highest resolution- access to electron correlations in solids?

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Abstract

Angle-resolved photoelectron spectroscopy has become one of the most important tools for studying the electronic properties in solids and at surfaces. From the kinematics of the photoemission process the energy versus wave vector relation $E(\vec{k})$ can now be determined routinely provided tunable photon sources are available. Much less attention was focused to the spectroscopic information on electron correlation effects that may be extracted from lineshapes and linewidths, respectively. In fact the determination of an "intrinsic" lineshape is hampered by several effects like broadening by finite spectrometer resolution, electron-phonon interactions and the influence of structural defects in the bulk or at the sample surface. We report on recent progress and discuss the present state of the art from an experimentalists point of view.

1. Introduction

Angle-resolved photoelectron spectroscopy [1-5] and its counterpart, inverse photoelectron spectroscopy [4-8], are among the most important and successful techniques to study the electronic properties of solids and, in particular, of their surfaces. Especially exploiting synchrotron radiation, where the photon energy $\hbar\omega$ may be tuned continuously, photoelectron spectroscopy is used to measure the electronic energy band structure $E(\vec{k})$, i.e. the dependence of the energy eigenvalues E on the wave vector \vec{k} along the various directions of the bulk Brillouin zone. In most valence band investigations reported to date, electron energy distribution curves (EDC) were measured (mostly at ambient

temperature) as a function of photon energy, orientation of the crystal surface, emission angles of the photoelectron with respect to the crystal orientation, and often also the direction of the electron spin. From the kinematics of the experiment, in particular from the conservation laws of energy and electron momentum $\hbar\vec{k}$, a wealth of information can be obtained on the energies E_i of initial and E_f of final states involved in the observed transition and on their dispersion with respect to the components of the electron wave vector parallel (\vec{k}_{\parallel}) and perpendicular (\vec{k}_{\perp}) to the surface. If polarized photon radiation is available, the exploitation of light polarization effects also allows to "switch-on" or "switch-off" particular photoelectron emission lines, and from the dipole selection rules the point-group symmetry of the involved electron wave-functions may be deduced.

In conclusion, angle-resolved valence-band photoelectron spectroscopy has matured to a well understood technique to map $E(\vec{k})$. Given a high-quality single crystal, a worked-out recipe to prepare a well ordered and stoichiometrically correct surface, and given also the availability of a tunable photon source, the determination of an experimental $E(\vec{k})$ is now almost a routine business. Bandmapping, however, represents only the lower level of photoelectron spectroscopy, i.e. the interpretation of peak-positions on the energy scale along the kinematical constraints of the experiment. The second and higher level is to derive and interpret experimental data on lineshapes, linewidths and peak intensities.

In fact the intrinsic spectral function (lineshape) of the photoelectron spectrum reflects excitations at all characteristic energies from the eV scale down to the thermodynamic kT scale. For example, linewidths contain information on the lifetime broadening of the final hole and electron states on electron-hole pair excitations, and conduction-electron screening both in the bulk of the sample and at its surface [4, 5, 9-17]. Moreover, electron-phonon and hole-phonon interactions, crystal-field splittings and e.g. the energy-dependent coupling of localized impurity states to a metallic environment [20] may contribute to the linewidths [13, 18-22]. Despite this richness of information not too much attention was focused to this spectroscopic information in recent years. The reasons are obvious: different experimental parameters like energy resolution, angular resolution and sample quality do often dominate the "experimental" spectral functions, thereby making the extraction of e.g. life-times very difficult or even impossible. The present paper has been written to illuminate this latter problem in some detail, to report on recent experimental progress and to discuss the state of the art as seen with the eyes of experimentalists.

2. The spectral function

Within the one electron picture the kinematics of the photoemission process is well understood, see also Fig. 1 : the photon is absorbed in the single-particle state at E_i and the excited electron, at a final state energy E_f above the vacuum level E_v , moves through the crystal, crosses the surface (thereby conserving \vec{k}_{\parallel}) and its kinetic energy E_{kin} is measured. Energy conservation then requires that $\hbar\omega = E_f - E_i = E_{kin} + \Phi - E_i$. Note that in our convention $E_f > 0$ and $E_i \leq 0$. Momentum conservation in the extended zone scheme then writes $\vec{k}_f - \vec{k}_i = \vec{q}_{ph} + \vec{G}$, where \vec{q}_{ph} is the wave-vector of the photon and \vec{G} represents a vector of the reciprocal bulk lattice. Generally the

photon wave vector is negligible as compared to the size of the first Brillouin zone, $\vec{q}_{ph} \ll 2\pi/a$, where a is the bulk lattice constant. Therefore in the reduced zone scheme momentum conservation in the bulk is generally written as $\vec{k}_f = \vec{k}_i$ ("vertical transition").

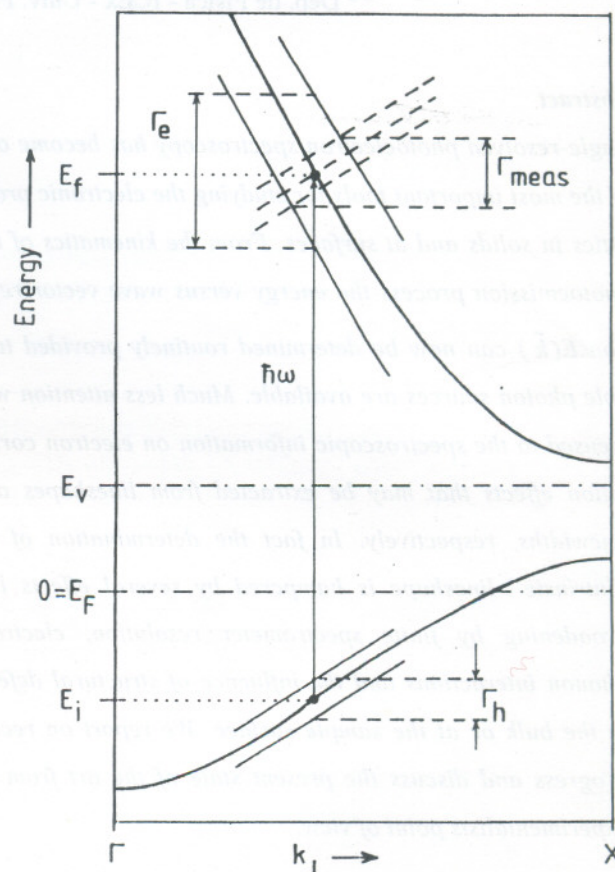


Fig. 1: Schematic energy band diagram defining relevant parameters used in the analysis of angle-resolved photoelectron spectra. For detail see text.

Going beyond this picture electron correlation effects must be included. The creation of a hole in the many-electron system results in an energy difference as compared to the one-electron initial state. This energy difference is usually expressed [4,5] by the real part of the quasi-particle self-energy Σ . The decay of the hole state via several possible excitation channels may be summarized phenomenologically [4,5,9,14,15] in the imaginary part of Σ . Assuming an exponential decay with time of the hole state, the spectral function of the hole is a Lorentzian given by [23, 24]

$$A(E_i, \vec{k}_i) = \frac{1}{\pi} \frac{\text{Im}\{\Sigma\}}{(E_i - E_i^0(\vec{k}_i) - \text{Re}\{\Sigma\})^2 + (\text{Im}\{\Sigma\})^2} \quad (1)$$

where the full width at half maximum $\Gamma_h = \hbar/\tau_h = 2 \cdot \text{Im}\{\Sigma\}$, connects Σ to the hole lifetime τ_h via the uncertainty relation. While $\text{Re}\{\Sigma\}$ is not directly accessible to an experiment, Γ_h may be derived from experimental linewidths, provided other contributions can be safely excluded.

The well known surface sensitivity of photoemission is based on the rather short escape depth of the electron in the final state, typically only a few atomic layers. This is correlated via the uncertainty relation with an uncertainty in $k_{f\perp}$, the final state wave vector perpendicular to the surface. Following Pendry /9, 25/ the distribution of k_f within this "time-reversed LEED state" may be described by

$$L_f = \frac{1}{2\pi} \frac{\Delta k_{f\perp}}{(k_{f\perp} - k_{f\perp}^0)^2 + (\Delta k_{f\perp}/2)^2} \quad (2)$$

If the final state is nearly-free-electron like the width of L_f is dominated by the final state lifetime τ_e via $\Delta k_{f\perp} \approx (\hbar/\tau_e) \cdot \partial k_{\perp} / \partial E_f$. As is evident the finite $\Delta k_{f\perp}$ can be converted into a corresponding width Γ_e which of course also contributes to the experimentally measured linewidth Γ_{meas} , compare Fig. 1 for the details.

In the most simple approximation the excited state electron and the hole left behind are considered to be non-interacting. Then the intensity of a photoemission peak can be expressed within the "one-step-model" formulation /4,5,9/ by the formula

$$I(E_f) \sim |M_{fi}|^2 \cdot L_f \cdot A \cdot \delta(\vec{k}_{i\parallel} - \vec{k}_{f\parallel} + \vec{g}_{\parallel}) \cdot \delta(\vec{k}_i - \vec{k}_f + \vec{G}) \cdot \delta(E_f - E_i - \hbar\omega) \quad (3)$$

where M_{fi} represents the transition matrix element (containing the dipole selection rules), and the δ -functions

account for the various conservation laws. \vec{G} and \vec{g}_{\parallel} are reciprocal lattice vectors in the bulk and at the surface, respectively.

In summary the energy position and the line-shape of the emission line reflect the complex response function of the interacting many-body system, thus giving information beyond the one-electron approximation. In particular actual research on high- T_c superconducting solids and on materials with only one- or two-dimensional electronic properties /14,26-28/ concentrates on electron-electron and electron-phonon interactions. Therefore the experimental determination of the spectral function in photoemission spectra is a matter of considerable current interest. In particular the low-energy excitations in the vicinity of the Fermi level at E_f require a detailed study of the spectral function of the hole. In the following we will therefore discuss the presently available experimental possibilities. For this purpose we started a series of high-resolution measurements on Cu and Ag. Their low-index surfaces are easy to prepare (as compared e.g. to chemically very complicated high- T_c materials), they do not reconstruct, they exhibit rather narrow photoemission lines, and their electronic energy bands (both 3D, and 2D at the surface) are understood in considerable detail /3-8/.

3. Experimental results

3.1 Electron and hole lifetimes

Integration of equation (3) over $dk_{f\perp}$ yields the connection between Γ_h , Γ_e and Γ_{meas} (compare also Fig. 1). This has been discussed in detail by Thiry /29/ and Smith /14, 15/. In the special case of emission normal to the surface, the peak width is given by /14, 15/

$$\Gamma_{\text{meas}} = \frac{\Gamma_h + |v_h^{\perp}/v_e^{\perp}| \cdot \Gamma_e}{1 - |v_h^{\perp}/v_e^{\perp}|} \quad (4)$$

where v_h^{\perp} and v_e^{\perp} are the corresponding band velocities $\hbar v^{\perp} = \partial E / \partial k_{\perp}$ normal to the surface. In equation (4) the wave-vector derivatives of the band structure are evaluated

at the points at which the unbroadened direct transition would occur /24/. In particular we see that $\Gamma_{\text{meas}} = \Gamma_h$ if $v_h^\perp = 0$. This is also seen directly from Fig. 1: with a flat hole band the width Γ_{meas} comes entirely from the hole lifetime and is independent of the momentum broadening in the final state. Although in metals generally $v_h^\perp < v_e^\perp$ (the free-electron like bands get steeper and steeper with increasing E_f) photoemission spectra taken at arbitrary points of the 3D k-space (i.e. $v_h^\perp \neq 0$) are most often completely dominated by Γ_e . To give an example we refer to reference /16/. The reason is that at final state energies typical for angle-resolved photoemission Γ_e is larger than Γ_h by generally an order of magnitude /17,24,30/. Three particular situations allow the sampling of data at $v_h^\perp = 0$: First of course emission from bulk bands at high-symmetry points, e.g. at the center of the bulk Brillouin zone, see /17/ for an example. Second emission from layered systems, like e.g. TiTe_2 or several high- T_c materials, see /26, 28/ for examples. Third photoemission out of occupied surface states for which by definition v^\perp is not at all defined, and their energy bands depend only on k_\parallel . We have therefore studied several surface states on Cu and Ag in great detail to figure out to which extent Γ_h may be extracted from experimental spectra. In the next section, we first discuss the influence of the spectrometer resolution parameters on linewidth and line-shape, respectively.

3.2 Energy-resolution

The energy resolution of our spectrometer can be tuned by the pass energy of the analyzer down to $\Delta E = 7$ meV. Therefore we were really surprised by the data reproduced in Fig. 2. It shows the electron intensity around E_f , indicating a (90% - 10%) width of about 20 meV. However at the sample temperature of $T = 35$ K we expect an intrinsic width as given by the Fermi distribution function of about $4k_B T = 11.7$ meV, which when convoluted with $\Delta E = 7$ meV should result in an experimental width definitely below 20 meV.

The origin of this unexpected broadening around E_f is self-absorption of the HeI-line ($\hbar\omega = 21.2\text{eV}$) within the cold-cathode gas-discharge lamp used by us as the photon source. Although this effect is well-known to atomic physicists /31/ its rather large effect on the effective width $\Delta\hbar\omega$ of the photon energy distribution seems not to be well known to surface physicists using the He-lamp. Therefore we believed it useful to communicate our corresponding results. Without the inconvenience to cool the sample to low T, the effective energy resolution may also be checked by taking photoelectron spectra from gases. A typical electron energy distribution curve as measured in our spectrometer is reproduced in Fig. 3, indicating a full width of 9.3 meV. From this number we extract an effective energy resolution of about 8 meV, after correction for the Doppler broadening of 4.3 meV at room temperature. In fact this number is in agreement with $\Delta E \sim 7$ meV as expected from the spectrometer setting and $\Delta\hbar\omega \cong 2$ meV as expected from atomic data tables /31/. However, to obtain the data of Fig. 3, the He-pressure in the photon source (differentially pumped in two stages using a 1 mm diameter quartz capillary) had to be reduced in order to minimize self-absorption within the lamp. The improvement in $\Delta\hbar\omega$ has however to be paid for by a dramatic loss in photon intensity. This is clearly seen in Fig. 4, where we plotted both the intensity (squares) and effective energy resolution (circles) as observed from the $3p_{3/2}$ emission line of gaseous Argon. As seen from Fig. 4, reduction of the He-pressure by about a factor of 5 may improve the effective resolution from about 16 meV to below 8 meV, but simultaneously reduces the intensity by more than an order of magnitude!

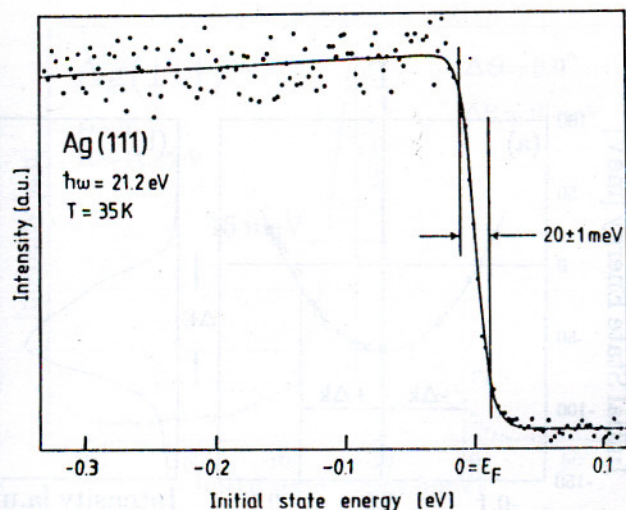


Fig. 2: Photoemission intensity in the vicinity of the Fermi energy E_F . The width (21 meV) of the sharp decrease around E_F reflects a convolution of the Fermi distribution function with the experimental spectrometer resolution function.

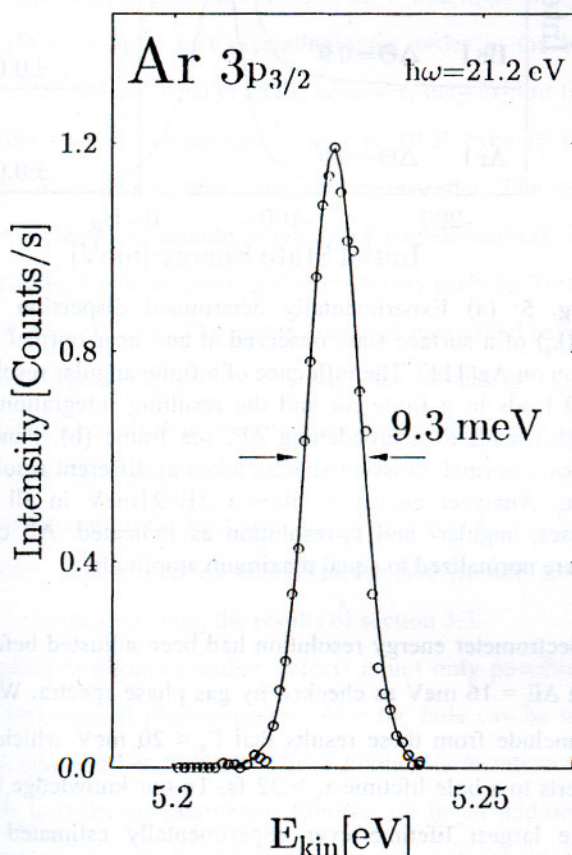


Fig. 3: Electron emission observed from the $3p_{3/2}$ level of gaseous Argon.

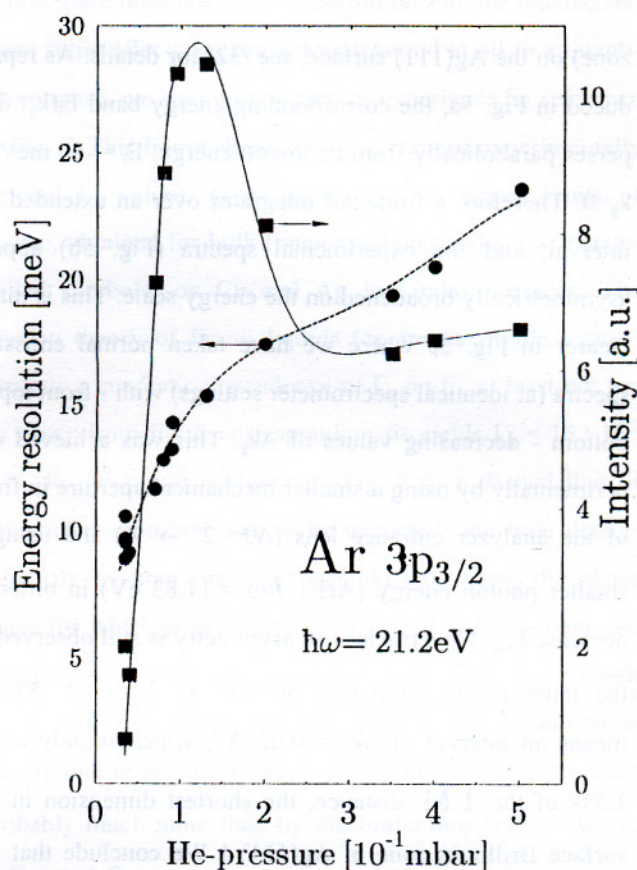


Fig. 4: Dependence of photon intensity (squares, right scale) and experimental energy resolution (circles, left scale) on Helium pressure near the first pumpig stage of a differentially pumped resonance lamp. Note that the electron spectrometer contributes about 7 meV to the total energy resolution. Thus the photon linewidth $\Delta\hbar\omega$ of this photon source contributes only about 2 meV at the lowest He pressures, but more than 20 meV at the highest ones.

3.3 Angular resolution

Surface state dispersions depend only on \vec{k}_{\parallel} , which is given

$$k_{\parallel} = \sin\theta \cdot \left[\left(\frac{2m}{\hbar^2} \right) \cdot E_{kin} \right]^{1/2}. \text{ In the formula } m \text{ is the}$$

free electron rest mass and θ is the polar angle of emission with respect to the surface normal at $\theta=0$. Clearly, any finite $\Delta\theta$ will result also in a finite

$$\Delta k_{\parallel} = \cos\theta \cdot \left[\left(\frac{2m}{\hbar^2} \right) \cdot E_{kin} \right]^{1/2} \cdot \Delta\theta. \text{ The influence of}$$

this effect is demonstrated with data taken from a well-known Shockley-type surface state [4, 5] occurring at and near $\theta=0$ (i.e. around the $\bar{\Gamma}$ -point of the surface Brillouin

zone) on the Ag(111) surface, see /32/ for details. As reproduced in Fig. 5a, the corresponding energy band $E_i(k_{\parallel})$ disperses parabolically from its lowest energy, $E_i = -60$ meV at $k_{\parallel}=0$. Therefore a finite $\Delta\theta$ integrates over an extended k_{\parallel} -interval, and the experimental spectra (Fig. 5b) appear asymmetrically broadened on the energy scale. This is made clearer in Fig. 5c where we have taken normal emission spectra (at identical spectrometer settings) with - from top to bottom - decreasing values of Δk_{\parallel} . This was achieved experimentally by using a smaller mechanical aperture in front of the analyzer entrance lens ($\Delta\theta = 2^\circ \rightarrow 9^\circ$) and using a smaller photon energy (ArI : $\hbar\omega = 11.83$ eV) in order to decrease E_{kin} . Nevertheless an asymmetry is still observed in the latter result. Note that $\Delta\theta = 0.9^\circ$ at $\hbar\omega = 11.83$ eV means an interval of $\Delta k_{\parallel} = 0.02 \text{ \AA}^{-1}$, which is only about 1.5% of the $\bar{\Gamma}\bar{M}$ distance, the shortest dimension in the surface Brillouin zone of Ag(111) ! We conclude that angular resolutions $\Delta\theta < 0.5^\circ$ will be necessary to avoid significant integration over the k_{\parallel} -space. Or in other words: any attempt to derive Γ_h for surface states requires a very detailed consideration of the influence of $\Delta\theta$ on the measured line-shapes.

Fig. 6 reproduces our "best result" obtained at $\Delta\theta=0$. Due to the asymmetry resulting from $\Delta\theta \neq 0$, we considered it reasonable to use exclusively the left wing to estimate its width. We consider the "true" peak to be a Lorentzian, and we use a Gaussian to model the spectrometer resolution function (which turned out to describe the Ar $3p_{3/2}$ spectra perfectly). Their convolution gives a good fit to the left wing to estimate its width. We consider the "true" peak to be a Lorentzian, and we used a Gaussian to model the spectrometer resolution function (which turned out to describe the Ar $3p_{3/2}$ spectra perfectly). Their convolution gives a good fit to the left wing, as indicated by the dashed line shown in Fig. 6. The fit yields a FWHM of 20.5 meV for the Lorentzian and 16.3 meV for the Gaussian. Note that the nominal

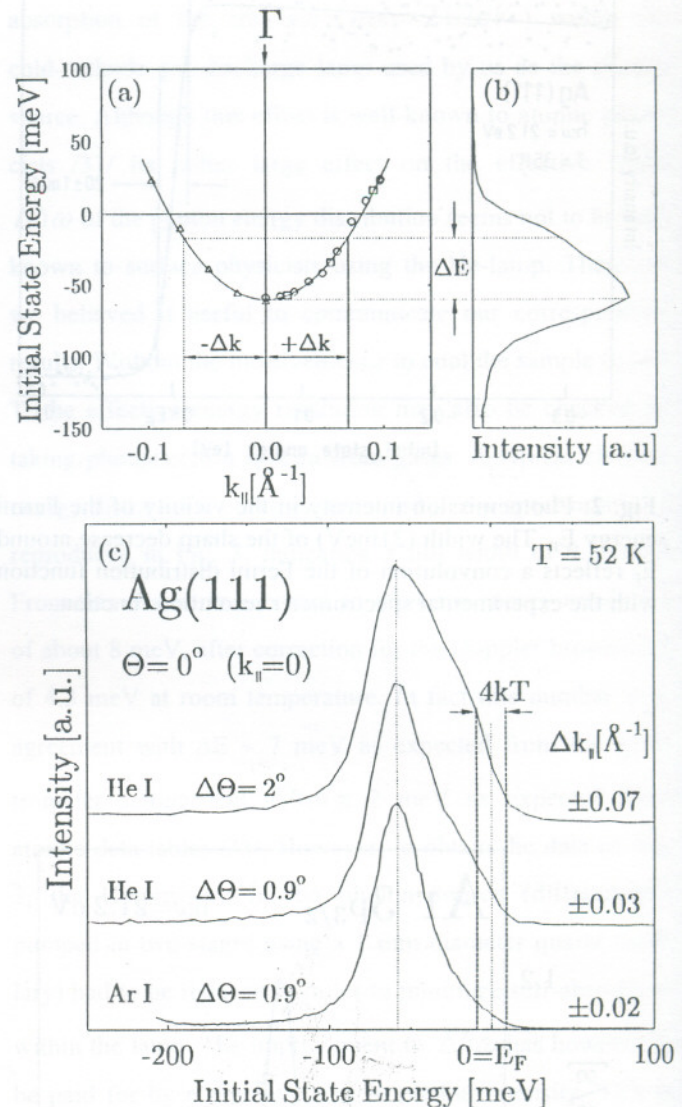


Fig. 5: (a) Experimentally determined dispersion curve $E_i(k_{\parallel})$ of a surface state observed at and near normal emission on Ag(111). The influence of a finite angular resolution $\Delta\theta$ leads to a finite Δk and the resulting integration over $E_i(k_{\parallel})$ leads to a broadening ΔE , see frame (b). Panel (c) shows normal emission spectra taken at different resolution Δk_{\parallel} . Analyzer energy resolution $\Delta E = 21$ meV in all three cases, angular- and k_{\parallel} -resolution as indicated. All curves were normalized to equal maximum amplitude.

spectrometer energy resolution had been adjusted before to be $\Delta E = 16$ meV as checked by gas phase spectra. We can conclude from these results that $\Gamma_h < 20$ meV which converts to a hole lifetime $\tau_h > 32$ fs. To our knowledge this is the largest lifetime ever experimentally estimated from photoemission at solid surfaces. However, we conjecture that Fig. 6 may still contain several sources of broadening.

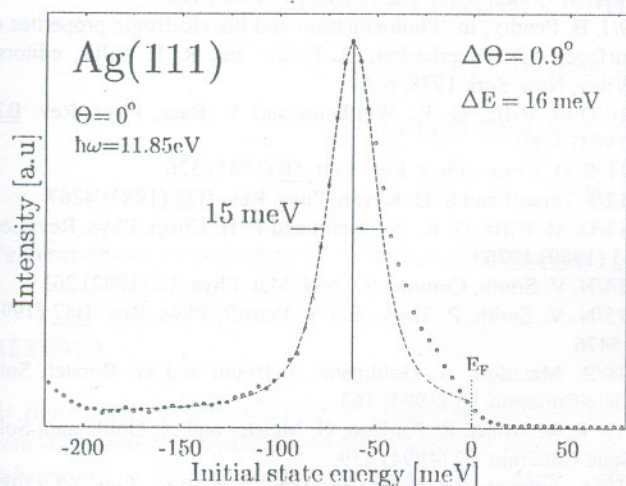


Fig. 6: Normal emission spectrum taken from Ag(111) close to E_F at a sample temperature of $T=56\text{K}$. Spectrometer energy resolution set to $\Delta E=10\text{meV}$. Circles indicate data points. The dashed curve has been fitted only to the left wing of the peak, and represents the convolution of a Gaussian (16.3meV FWHM) with a Lorentzian (20.5meV FWHM).

3.4 Sample quality

The conservation laws expressed in the δ -functions of equation (3) assume perfect crystallographic order in the bulk and at the surface. Real crystals, however, may exhibit bulk disorder (e.g. a mosaic spread of e.g. $\pm 0.2^\circ$ even in very good Cu single crystals) and surface disorder (like steps kinks, defects, or minute amounts of contamination). The latter effect has been discussed already very early by Tersoff and Kevan [11, 12]. Our group has also performed several studies, where the effect of inadequate annealing of the sample was studied in its influence on the line width [33-35]. These data allow the influence of surface disorder to be estimated and, in the best case, be extrapolated to minimum disorder. The effects of bulk disorder like mosaic spread may be estimated using the results of section 3.3.

Coupling to phonons and/or defects is not only possible in the final state of photoemission. Also the hole can be scattered into another hole state by a phonon-hole interaction which is reflected (shortened lifetime τ_h) in an additional width that increases linearly with temperature T [22, 36]. All of these effects must be carefully investigated and discussed before Γ_h can be extracted safely from photoelectron lines.

While space does not allow to go too far into the details, and since some effects are not yet understood at all in a quantitative sense, we sum up our present experience by reference to Fig. 7. This figure shows a compilation of experimentally derived Γ_h values, to be considered as upper limits of course, obtained for bulk (open symbols) and surface states (filled symbols) on Cu and Ag low-index surfaces. The Landau theory of Fermi liquids (as are the noble metals) predicts a quadratic dependence of Γ_h on E_i , at least not too far away from E_F . A corresponding fit yields $\Gamma_h = 15 \cdot 10^{-3} \text{ eV}^{-1} (E_i - E_F)^2$ and is shown in Fig. 7 as a dashed line. It approaches zero at E_F as to be expected: the hole decays primarily by nonradiative Auger-like processes, the phase space for which goes to zero for $E_i \rightarrow 0/4$. In contrast, the surface state widths are almost constant, typically at about $25 \pm 5 \text{ meV}$. We conjecture that near E_F the limitation is mainly due to the insufficient quality of our samples, most probably much more than by the limitations set by $\Delta \hbar \omega$, ΔE and $\Delta \theta$.

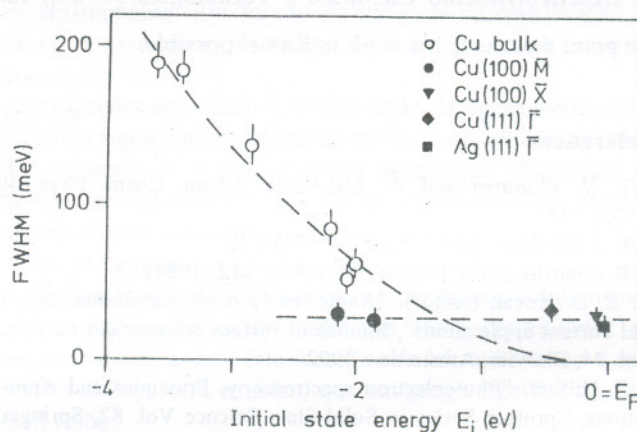


Fig. 7: Photoemission linewidths observed for several transitions on Cu and Ag surfaces. Open symbols: emission out of Cu 3d bulk bands. Filled symbols: emission out of surface states as labeled in the figure.

4. Summary and conclusions

We have discussed to which extent information on line-shapes and line-widths (and of course also on line-intensities, which check for details of the wave-functions involved in a transition) may be derived from photoelectron

spectra taken at very high resolution. As is evident from Fig. 7, values of $\Gamma_h > 30$ meV ($\tau_h < 20$ fs) can be measured reliably provided the influence of spectrometer resolution parameters and of the sample quality is known at a sufficient level of sophistication. Cases of $\Gamma_h < 20$ meV ($\tau_h > 30$ fs) have not yet been resolved, although expected e.g. on surfaces of noble metals at initial state energies near E_F . The present experimental limitation is given most probably by the quality of the available samples, both with respect to bulk and surface. Further progress, besides of course ever improving spectrometers, requires better samples and in particular a more systematic and more quantitative understanding of "all" distortions of the "intrinsic" line shape.

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