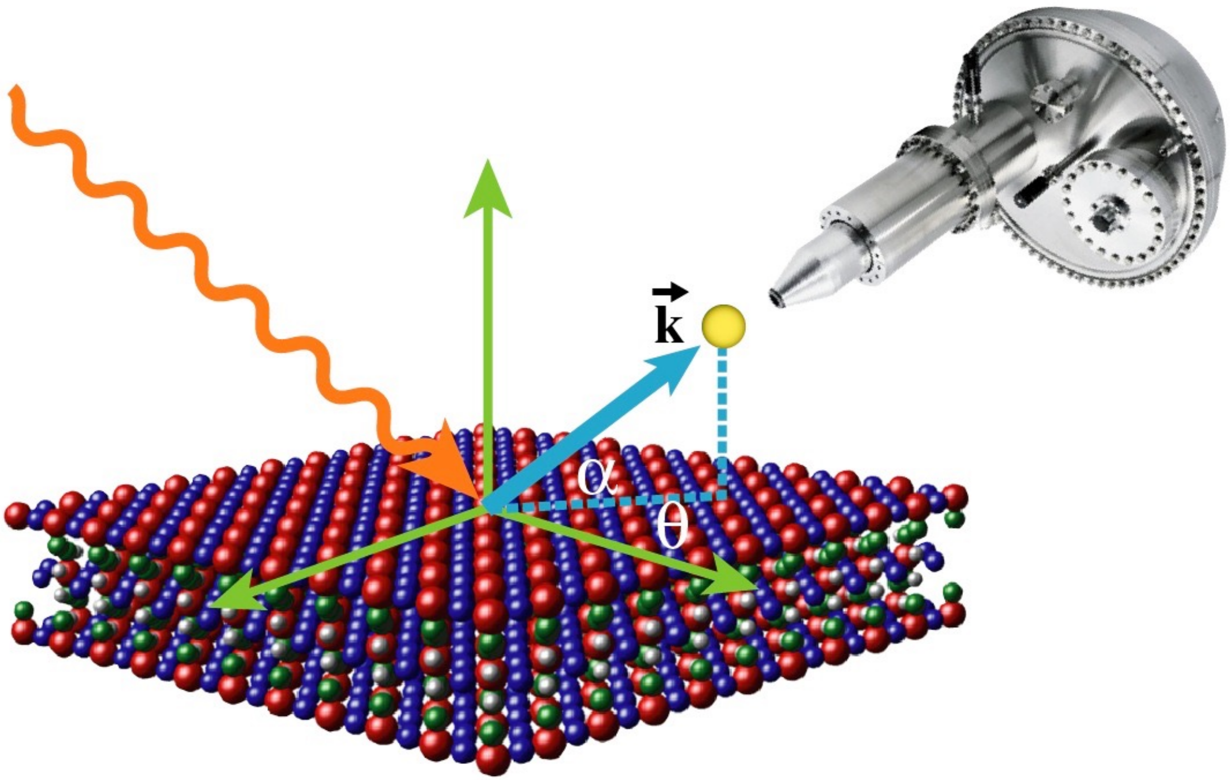
From Einstein's Photoelectric Effect to Band Mapping



Kyle Shen

PARADIM Summer School

June 16, 2022

Overview of ARPES Lectures & Labs

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Thursday, June 16

- Lecture #1 : From Einstein's Photoelectric Effect to Band Mapping
- Lecture #2 : From Particles to Quasiparticles : Understanding & Measuring Interactions by ARPES
- Lab Lecture : Introduction to ARPES Labs (Single Crystals)

Friday, June 17

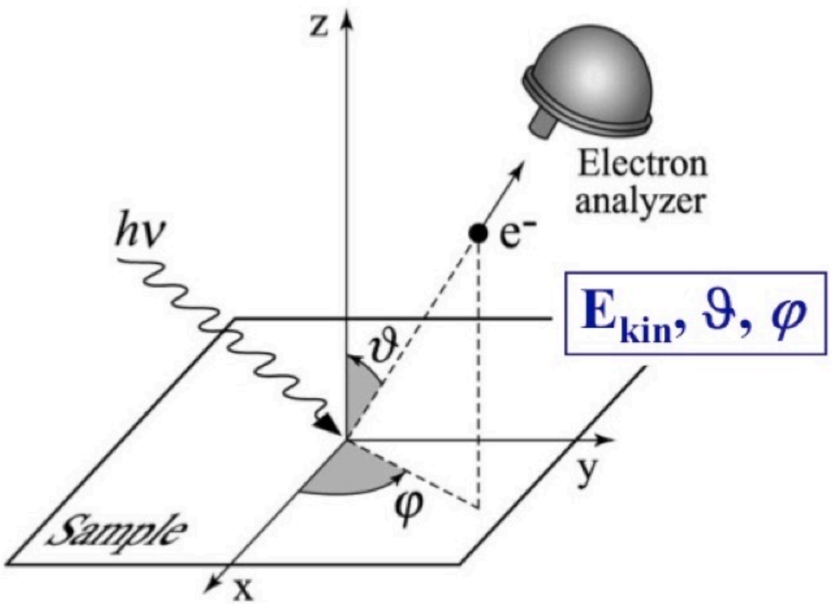
- Lecture #3 : ARPES Studies of Quantum Materials
- Lecture #4 : Frontiers in ARPES
- Lab Lecture : Introduction to ARPES Labs (Thin Films)

ARPES Labs

- Lab #1 : ARPES on single crystals
- Lab #2 : ARPES on thin films

Concept Question #3

In the photoemission process, the in-plane (longitudinal) momentum of the photoelectron can be directly related to the in-plane momentum of the electron when it was inside the solid, due to translational symmetry. For the out-of-plane (perpendicular) component of the photoelectron's momentum :



A. The momentum of the ejected electron is **not** conserved along the perpendicular direction
B. The out-of-plane photoelectron momentum can likewise be directly related to the out-of-plane momentum of the electron when it was in the solid
C. The perpendicular component of the electron's momentum cannot be experimentally determined
D. The perpendicular momentum of the electron inside the crystal is not a good quantum number

Transmission of electrons through the surface

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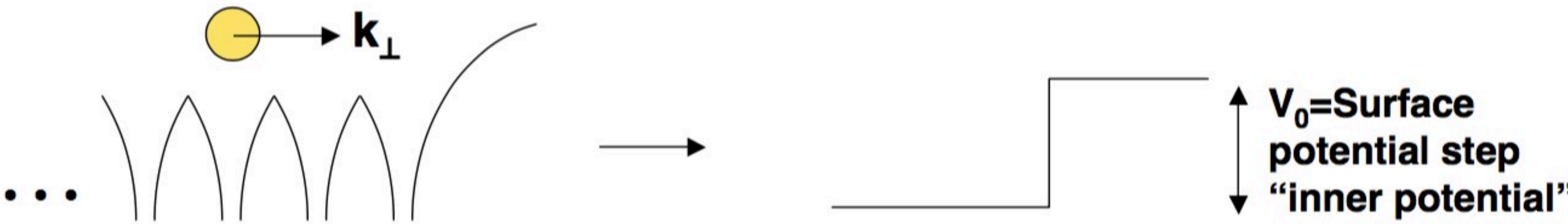
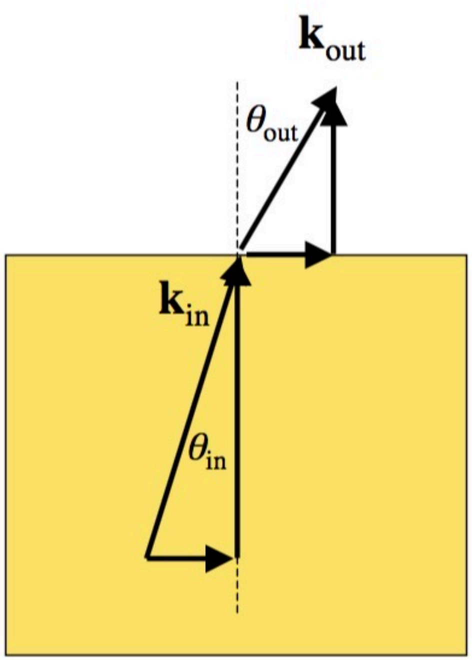
We approximate the surface as a square potential barrier.



We assume the electrons outside the sample have energy $E = p^2/2m = \hbar^2 k^2/2m$



Can think of a analogous “Snell’s Law” for photoemission, where the in-plane momentum is conserved and the out-of-plane momentum changes due to scattering off the potential barrier (work function)

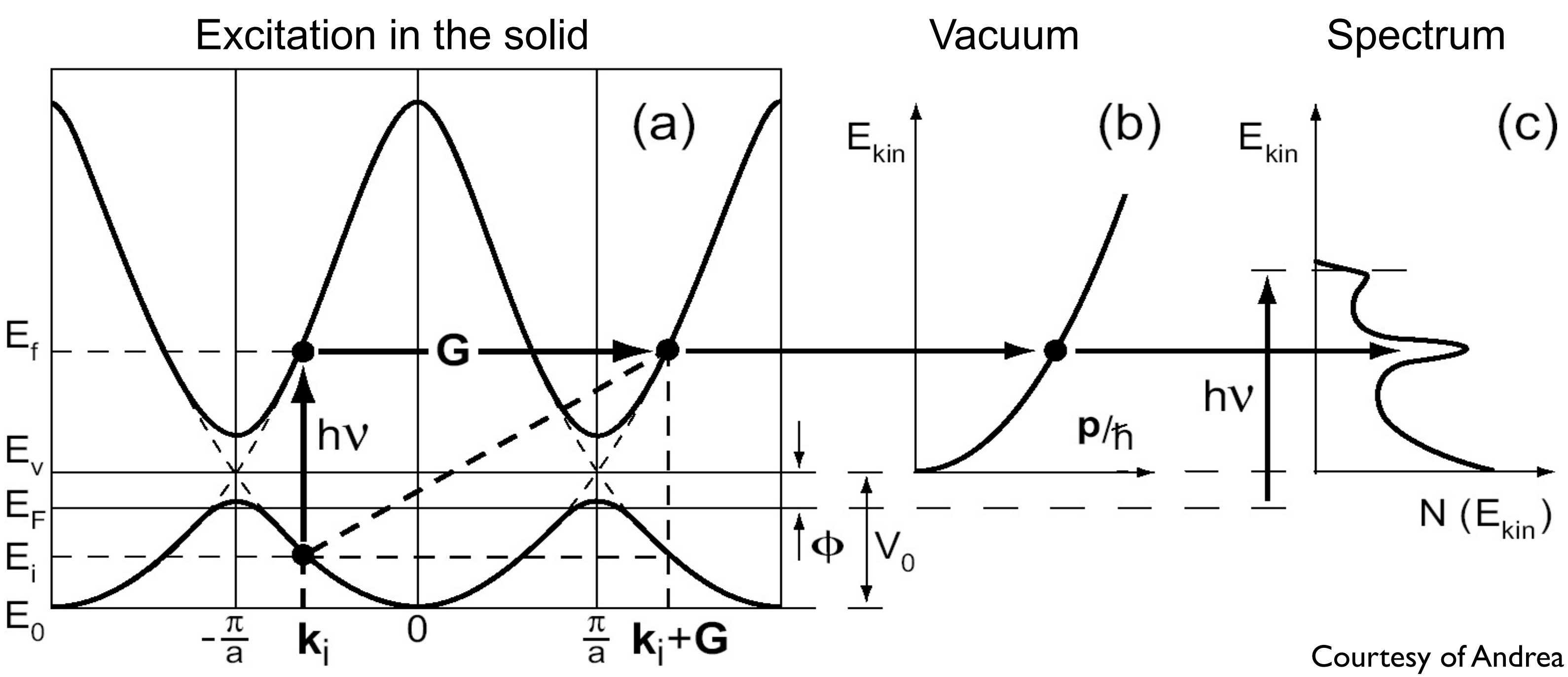
Courtesy of Eli Rotenberg

inner potential and determination of k_z

Free-electron final state $E_f(\mathbf{k}) = \frac{\hbar^2 \mathbf{k}^2}{2m} - |E_0| = \frac{\hbar^2 ({\mathbf{k}_{\|}}^2 + {\mathbf{k}_{\perp}}^2)}{2m} - |E_0|$

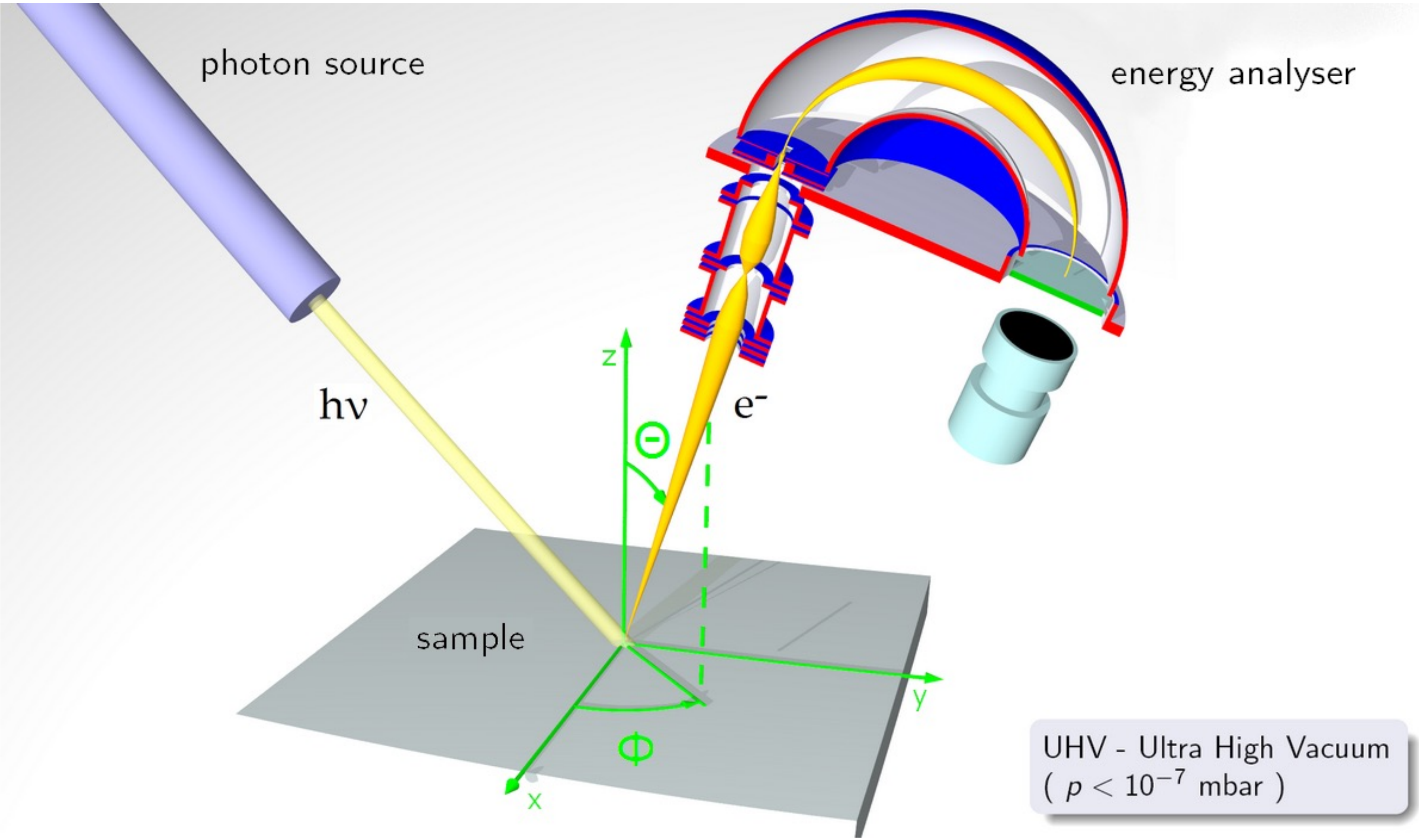
because $\hbar^2 \mathbf{k}_{\|}^2 / 2m = E_{kin} \sin^2 \vartheta$ $\quad E_f = E_{kin} + \phi$ $\quad V_0 = |E_0| + \phi$

$$\mathbf{k}_{\perp} = \frac{1}{\hbar} \sqrt{2m(E_{kin} \cos^2 \vartheta + V_0)}$$

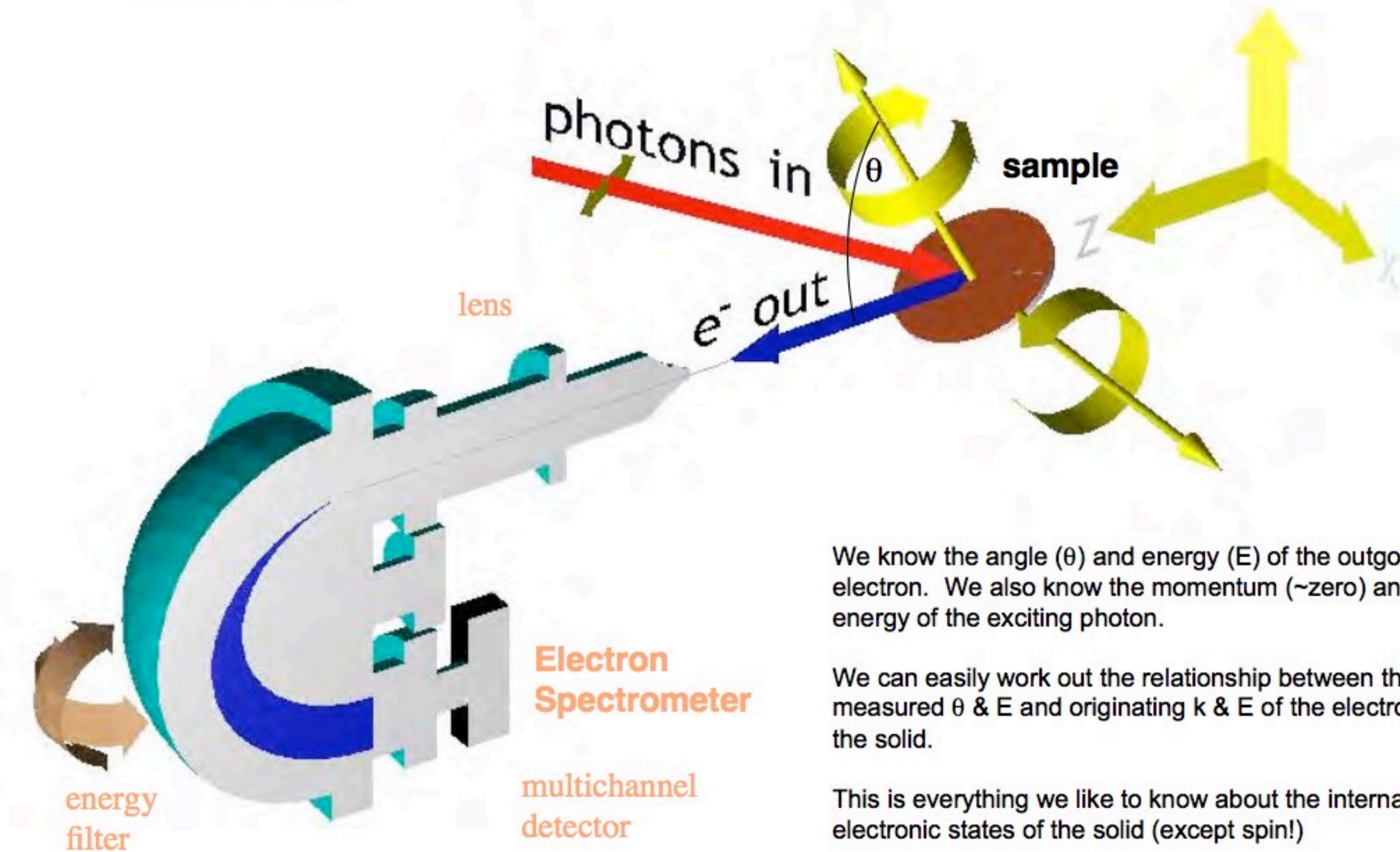


Courtesy of Andrea Damascelli

Schematic of the ARPES process



Schematic of the ARPES process



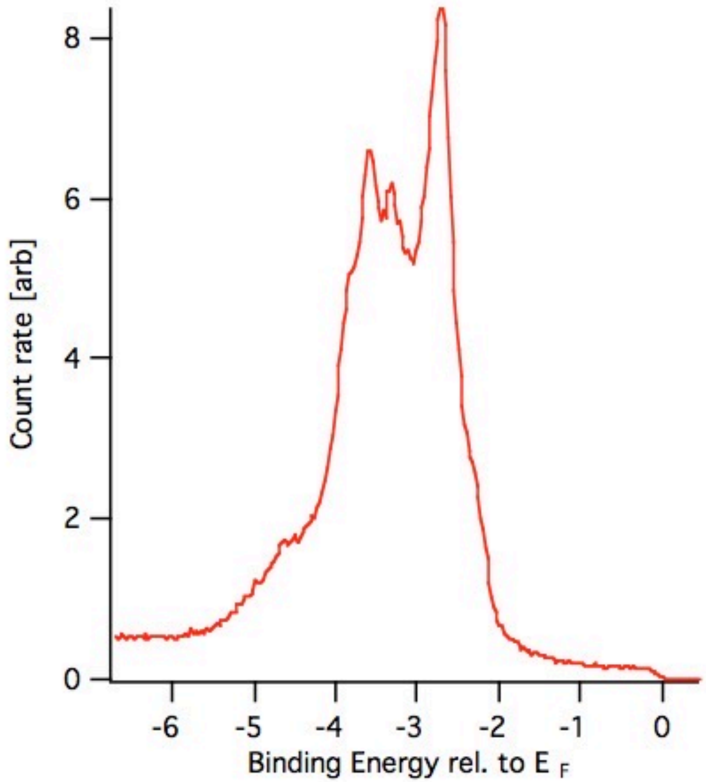
We know the angle (θ) and energy (E) of the outgoing electron. We also know the momentum (~zero) and the energy of the exciting photon.

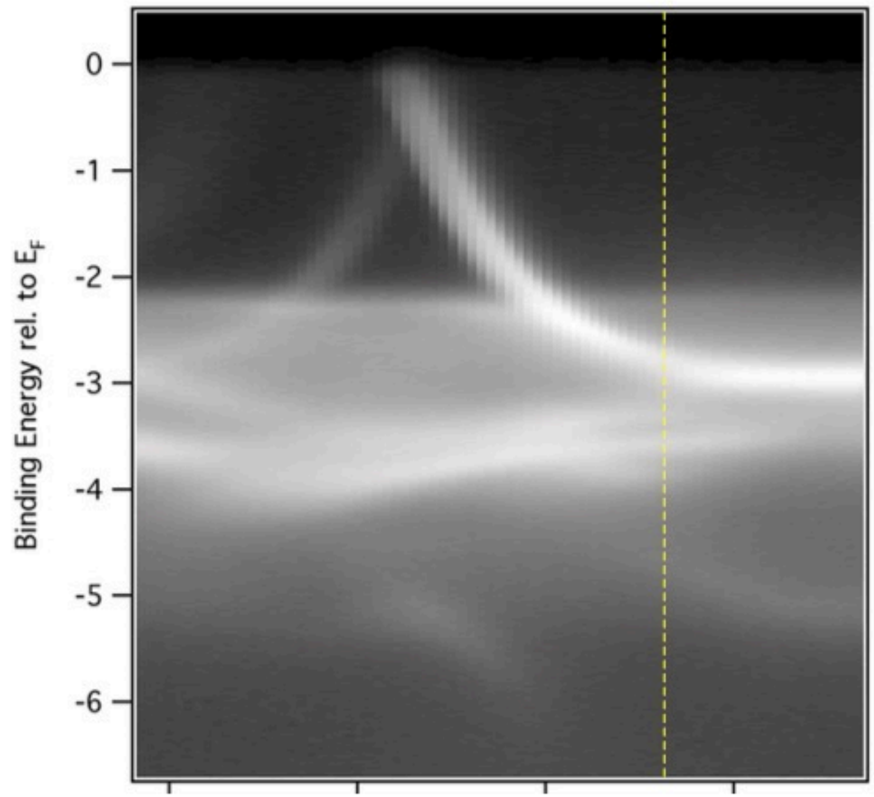
We can easily work out the relationship between the measured θ & E and originating k & E of the electrons in the solid.

This is everything we like to know about the internal electronic states of the solid (except spin!)

Courtesy of Eli Rotenberg

ARPES data acquisition





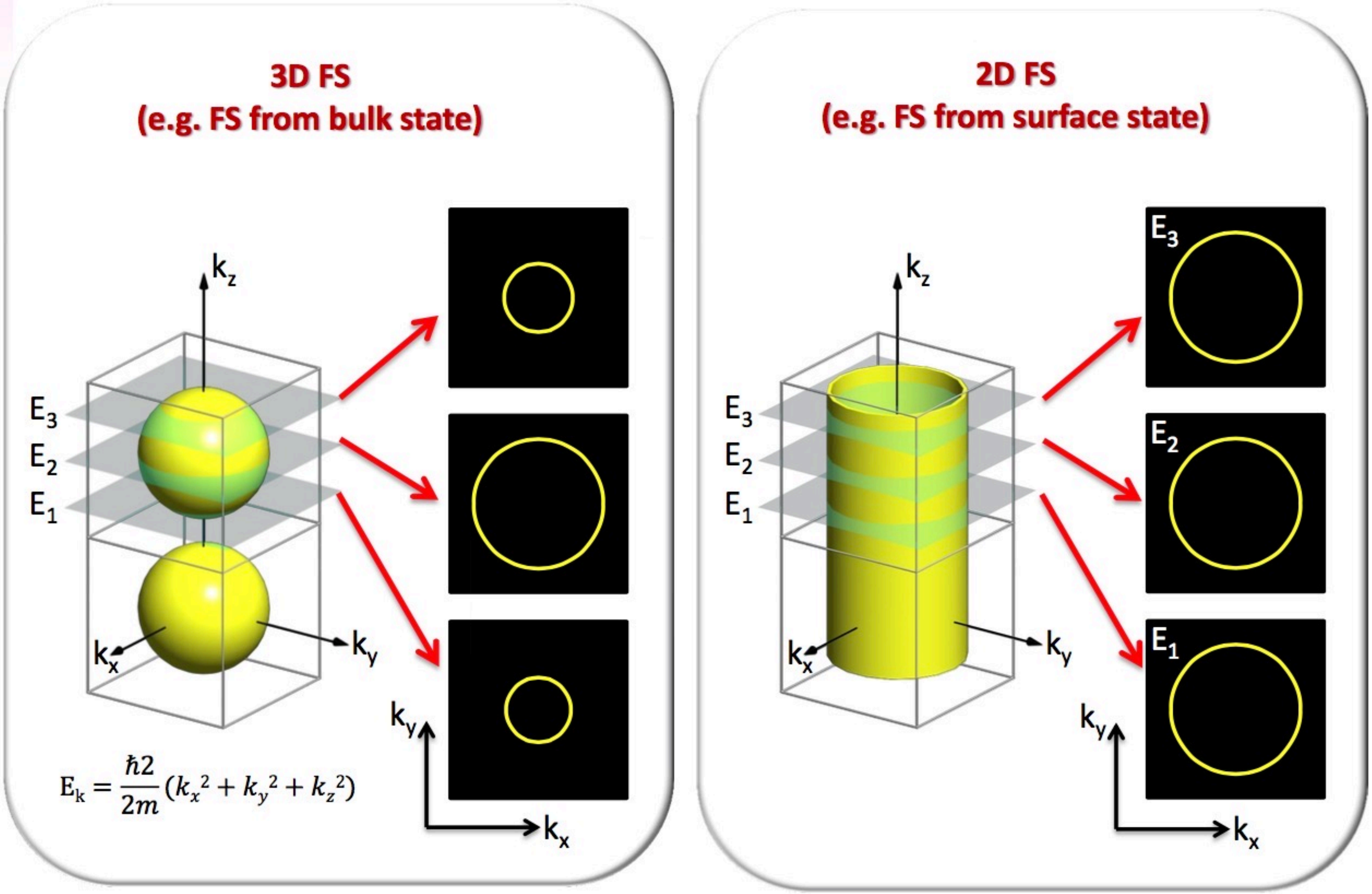
A spectrum at a single momentum k_x

Accumulate spectra as the momentum k_x is scanned

Courtesy of Eli Rotenberg

Three-dimensional electronic structure mapping

$$\mathbf{k}_\perp = \frac{1}{\hbar}\sqrt{2m(E_{kin}\cos^2\vartheta + V_0)}$$



Courtesy of Eli Rotenberg

Concept Question #4

Which of the following is strongly dependent on the value of the photon energy used to perform the experiments? Assume the photon energy is always large enough to eject electrons from the sample.

1. The energy resolution of our measurement (i.e. how fine a feature ΔE we can resolve)
2. The number of (occupied) states that we can access below E_F
3. The number of (unoccupied) states that we can access above E_F
4. The intensity of the measured photoemission signal

A. 2 only

B. 2, 3 & 4

C. 1, 2, & 4

D. 2 & 4

E. All of the above

Concept Question #5

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Which of the following is strongly dependent on the value of the photon energy used to perform the experiments? Assume the photon energy is always large enough to eject electrons from the sample.

1. The momentum resolution of our measurement (i.e. how fine a feature Δk we can resolve)
2. The range in momentum space (span of the Brillouin zone) that we can access
3. The out-of-plane momentum, k_z, that is being accessed

A. None of the above

B. 2 only

C. 2 & 3

D. 3 only

E. All of the above

photon sources for photoemission can be…

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X-ray tubes Most common sources for XPS (Al, Mg anodes), can be used with grating for better energy resolution (1000-10,000 eV, $\Delta E \sim 0.1-1$ eV)

Plasma Discharge Narrow bandwidth, high intensity lamps in VUV (10-100 eV, $\Delta E \sim 0.001$ eV); used for ARPES

Synchrotrons Complete control over photon beam (energy, polarization, resolution); user facilities

Lasers Higher harmonic generation; low energy (> 10 eV). Pump-probe, or high resolution

Photon source must be :

1. Monochromatic
2. High intensity (> 10^9 s^{-1})
3. Energetic ($h\nu > \phi \sim 5$ eV)

photoemission at different photon energy ranges

$h\nu$ (eV)	ΔE & Δk	Cross Section	Primarily Used For / Special Capabilities
10 – 100 eV synchrotrons, plasma lamps, lasers	**10^{-3} eV** **10^{-3} A^{-1}**	**10 - 1** (Mb / atom)	High-resolution studies of electronic structure & surfaces, Fermi surface & band mapping, low-energy physics
100 – 1000 eV synchrotrons	**10^{-2} eV** **10^{-2} A^{-1}**	**10 - 0.01** (Mb / atom)	Resonant photoemission X-ray absorption / magnetic dichroism XPS (elemental chemical analysis)
1000 – 10,000 eV X-ray tubes, synchrotrons	**10^{-1} eV** **10^{-1} A^{-1}**	**10^{-2} – 10^{-4}** (Mb / atom)	Bulk sensitivity Elemental & chemical analysis Changing orbital cross-sections

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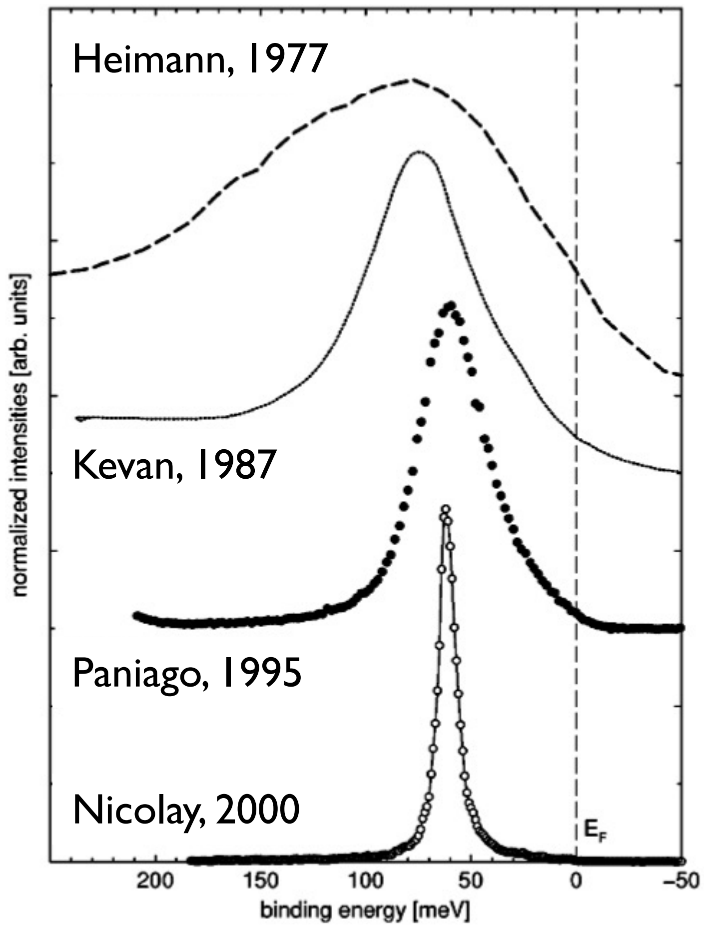
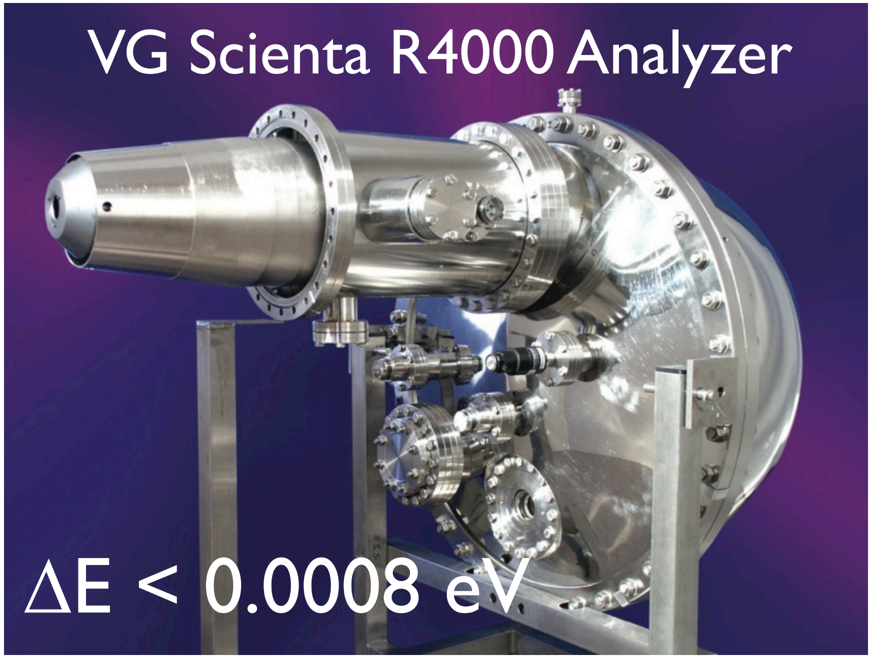
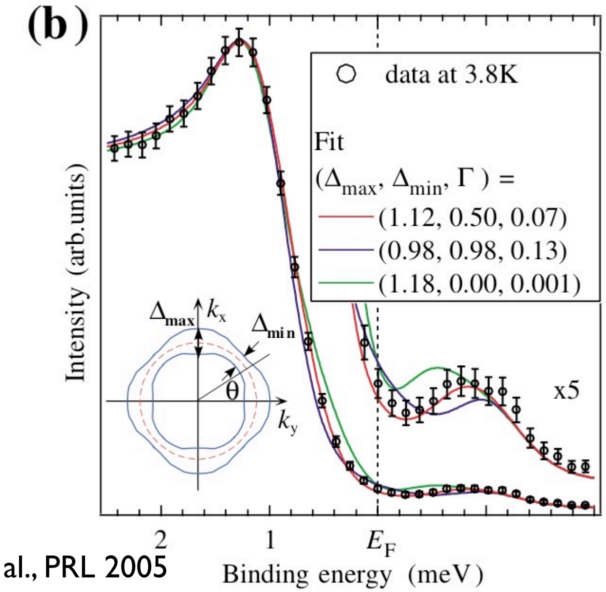
Evolution of instrumental resolution over time



F. Reinert *et al.*, PRB (2001)

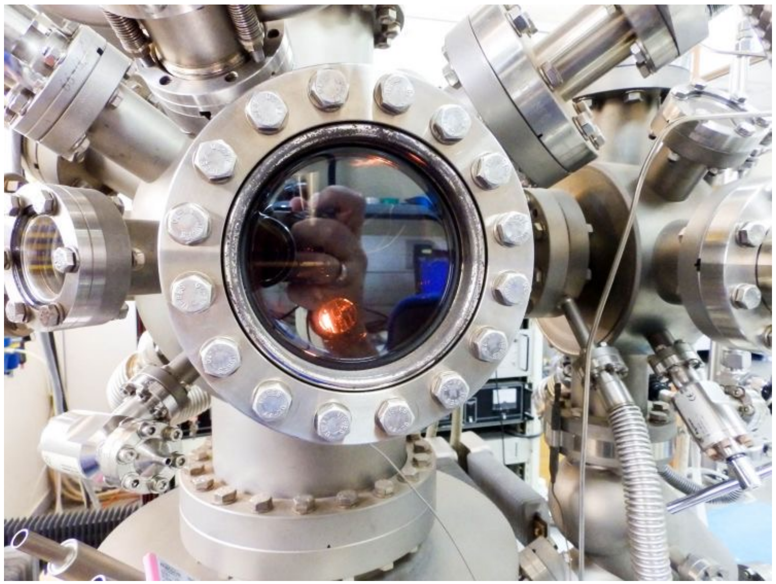




T. Kiss et al., PRL 2005

Concept Question #6

ARPES measurements need to take place in ultrahigh vacuum (10^{-10} torr or better). Which of the following is the **most important** factor which determines the level of vacuum needed to perform experiments?



A. The scattering / absorption of photoelectrons traveling inside the chamber

B. The operation of the electron analyzer

C. The absorption of vacuum ultraviolet (VUV) photons used for photoemitting the electrons

D. The scattering of electrons from adsorbed molecules at the sample's surface

E. All of the above are equally important