From Einstein’s Photoelectric Effect to Band Mapping

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PARADIM Summer School

June 16, 2022
Overview of ARPES Lectures & Labs

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
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

We approximate the surface as a square potential barrier.

We assume the electrons outside the sample have energy \( E = \frac{p^2}{2m} = \frac{\hbar^2 k^2}{2m} \)

Can think of an 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

$$E_f(\mathbf{k}) = \frac{\hbar^2 k^2}{2m} - |E_0| = \frac{\hbar^2 (k_\parallel^2 + k_\perp^2)}{2m} - |E_0|$$

because

$$\frac{\hbar^2 k_\parallel^2}{2m} = E_{\text{kin}} \sin^2 \vartheta$$

$$E_f = E_{\text{kin}} + \phi$$

$$V_0 = |E_0| + \phi$$

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

Excitation in the solid

Vacuum

Spectrum

Courtesy of Andrea Damascelli
Schematic of the ARPES process

- Photon source
- Energy analyser
- Sample
- hv
- e⁻

UHV - Ultra High Vacuum
(\rho < 10^{-7} \text{ mbar})
Schematic of the ARPES process

We know the angle ($\theta$) 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 $\theta$ & $E$ 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

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

3D FS (e.g. FS from bulk state)

$$E_k = \frac{\hbar^2}{2m} (k_x^2 + k_y^2 + k_z^2)$$

2D FS (e.g. FS from surface state)

Courtesy of Eli Rotenberg
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 $\Delta E$ we can resolve)  
   - A. 2 only
   - B. 2, 3 & 4
   - C. 1, 2, & 4
   - D. 2 & 4
   - E. All of the above

2. The number of (occupied) states that we can access below $E_F$
   - A. 2 only
   - B. 2, 3 & 4
   - C. 1, 2, & 4
   - D. 2 & 4
   - E. All of the above

3. The number of (unoccupied) states that we can access above $E_F$
   - A. 2 only
   - B. 2, 3 & 4
   - C. 1, 2, & 4
   - D. 2 & 4
   - E. All of the above

4. The intensity of the measured photoemission signal
Concept Question #5

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 $\Delta k$ 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...

<table>
<thead>
<tr>
<th>Photon Source</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>X-ray tubes</strong></td>
<td>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)</td>
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<tr>
<td><strong>Plasma Discharge</strong></td>
<td>Narrow bandwidth, high intensity lamps in VUV (10-100 eV, $\Delta E \sim 0.001$ eV); used for ARPES</td>
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<tr>
<td><strong>Synchrotrons</strong></td>
<td>Complete control over photon beam (energy, polarization, resolution); user facilities</td>
</tr>
<tr>
<td><strong>Lasers</strong></td>
<td>Higher harmonic generation; low energy ($&gt; 10$ eV). Pump-probe, or high resolution</td>
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</tbody>
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**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

<table>
<thead>
<tr>
<th>Photon Energy (eV)</th>
<th>ΔE &amp; Δk</th>
<th>Cross Section</th>
<th>Primarily Used For / Special Capabilities</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 – 100 eV</td>
<td>10^{-3} eV, 10^{-3} Å^{-1}</td>
<td>10 – 1</td>
<td>High-resolution studies of electronic structure &amp; surfaces, Fermi surface &amp; band mapping, low-energy physics</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(Mb / atom)</td>
<td></td>
</tr>
<tr>
<td>100 – 1000 eV</td>
<td>10^{-2} eV, 10^{-2} Å^{-1}</td>
<td>10 – 0.01</td>
<td>Resonant photoemission</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(Mb / atom)</td>
<td>X-ray absorption / magnetic dichroism</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>XPS (elemental chemical analysis)</td>
</tr>
<tr>
<td>1000 – 10,000 eV</td>
<td>10^{-1} eV, 10^{-1} Å^{-1}</td>
<td>10^{-2} – 10^{-4}</td>
<td>Bulk sensitivity</td>
</tr>
<tr>
<td></td>
<td></td>
<td>(Mb / atom)</td>
<td>Elemental &amp; chemical analysis</td>
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<td></td>
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<td>Changing orbital cross-sections</td>
</tr>
</tbody>
</table>
the current state-of-the-art in electron spectroscopy

Evolution of instrumental resolution over time

Heimann, 1977
Kevan, 1987
Paniago, 1995
Nicolay, 2000

\[ \Delta E < 0.0008 \text{ eV} \]

F. Reinert et al., PRB (2001)
T. Kiss et al., PRL 2005

VG Scienta R4000 Analyzer

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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