Theoretical Description of Angle Resolved Photoemission in the X-ray Regime on the Basis of the One-Step Model

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Outline

- Introduction
- Importance of surface contributions
- One-step model of photo-emission
- Reduction of surface sensitivity for high photon energies (HAXPES)
- HAXPES specific issues
  - photon momentum
  - thermal effects
  - non-dipole contributions (not yet)
- Summary
Description of PES via three-step model

three-step model of photo-emission (ARPES)
Berglund and Spicer (1964)

\[ I \sim \tilde{D}(E, \omega) \sum_{nn'} \int d^3k \ T(E, \tilde{k}) \left| \langle n'\tilde{k} | \vec{p} | n\tilde{k} \rangle \right|^2 \]

\[ \times \delta(E - E_{n\tilde{k}} - \omega) \delta(E_{n'\tilde{k}} - E) \Theta(E - E_F) \Theta(E_F + \omega - E) \]
PES calculations based on three-step model

PES of polycrystalline Cu for $\hbar \omega = 8 - 17$ eV

Theory: Janak et al. (1975)  
Expt: Eastman and Grobman

- Central ingredients to three step-model calculated from band structure data; e.g.: $\langle n' \vec{k}|p|n \vec{k} \rangle$

- Life times enter as parameters

$$T(E, \vec{k}) = \frac{\alpha(\omega)l(E,\vec{k})}{1+\alpha(\omega)l(E,\vec{k})}$$

$\tau_{\text{phot}} \rightarrow \alpha(\omega)$

$\tau_{\text{el}} \rightarrow l(E, \vec{k})$

Expt. and Theor. photoemission distribution for $\hbar \omega = 8 - 12$ eV (left) and 13 - 17 eV (right)
Ag/Au(111): surface state emission


radiation source
wave vector $\vec{q}$
polarisation $\lambda$

photo electron detector
wave vector $\vec{k}$
spin state $m_s$

photo-current (Fermi’s golden rule)

$$ j \propto \sum_i \lvert \langle \phi_f | \hat{H}_{rad}^{\vec{q}\lambda} | \phi_i \rangle \rvert^2 \delta(E_f - E_i - \omega) $$

with final state $\phi_f = T_R \phi^{LEED}$ — time reversed LEED state
One-step model of photoemission II

photo current

\[ j \propto \sum_i \langle \phi_f | \hat{\mathbf{H}}_{\text{rad}}^{\vec{q}_\lambda} | \phi_i \rangle \langle \phi_i | \hat{\mathbf{H}}_{\text{rad}}^{\vec{q}_\lambda\dagger} | \phi_f \rangle \delta(E_f - E_i - \omega) \]

\[ \propto \langle \phi_f | \hat{\mathbf{H}}_{\text{rad}}^{\vec{q}_\lambda} | \text{Im} \ G_i \hat{\mathbf{H}}_{\text{rad}}^{\vec{q}_\lambda\dagger} | \phi_f \rangle \]

initial state Green’s function

\[ \text{Im} \ G_i(E) = \sum_i |\phi_i\rangle \langle \phi_i| \delta(E - E_i) \]

final state

\[ \phi_f = \mathcal{T}_R \phi_{\text{LEED}} \]

\[ = \mathcal{T}_R \left[ e^{i\vec{k}_f \vec{r}} + \int d^3r' G(\vec{r}, \vec{r}', E_f) V(\vec{r}') e^{i\vec{k}_f \vec{r}'} \right] \]

e.g. Caroli et al. (1973), Feibelmann and Eastman (1974)
One-step model of photoemission

Implementation via multiple scattering theory
Adaption of LEED formalism (Pendry et al. 1980)

Single layer scattering matrix

\[
M_{\tau \tau' ss'} = \delta_{\tau \tau' ss'} + \frac{8\pi^2}{kk'gg'} \sum_{\kappa\mu} \sum_{\kappa'\mu'} \sum_{\kappa''\mu''} \sum_{ii'} i^{-l} C_{\kappa\mu s} Y_{l}^{\mu - s} (\hat{k}_{g}) e^{-i k_{g} R_{i}}
\]

\[
\times \left(1 - X\right)^{-1} i_{\kappa''\mu''}^{l'} \sum_{i'\kappa'\mu'} Y_{l'}^{\mu' - s'} (\hat{k}_{g'}) e^{i k_{g'} R_{i'}}
\]

In-layer scattering
- UV photoemission: \( X \neq 0 \)
- X-ray photoemission: \( X = 0 \)

Relativistic formulation: e.g. Braun (1996)
Surface sensitivity: \( n \text{MgO/Fe(001)} \) at 1000eV

calculated ARPES intensities \( I(E, \Theta) \)

Fe(001)  

\[
\begin{array}{c}
\text{1 ML MgO/Fe(001)} \\
\text{increasing coverlayer thickness}
\end{array}
\]

\[ \Rightarrow \]

MgO  

\[
\begin{array}{c}
\text{8 ML MgO/Fe(001)} \\
\text{Fe-related features get lost with increasing MgO coverage}
\end{array}
\]
Surface sensitivity for high photon energies

calculated ARPES intensities $I(E, \Theta)$

$E_{\text{phot}} = 1000\text{eV}$

$E_{\text{phot}} = 6000\text{eV}$

8 ML MgO/Fe(001)

Fe-related features recovered for high photon energies

$\Rightarrow$ access to burried interfaces (SPIN HAXPES; Felser et al.)
Photon momentum effects on Ag(001)

Ag(001) photoemission intensities along ΓK with LCP-light at $h\nu=552$ eV

$q_{\text{photon}}$ ignored

$$k_i = (k_{\parallel} + g, \sqrt{2(E - iV_{i1}) - |k_{\parallel} + g|^2})$$

$$k_f = (k_{\parallel} + g, \sqrt{2(E + \omega - iV_{i2}) - |k_{\parallel} + g|^2})$$

$q_{\text{photon}}$ included

$$k_i = (k_{\parallel} - q_{\parallel} + g, \sqrt{2(E - iV_{i1}) - |k_{\parallel} - q_{\parallel} + g|^2})$$

$$k_f = (k_{\parallel} + g, \sqrt{2(E + \omega - iV_{i2}) - |k_{\parallel} + g|^2})$$

Venturini et al. PRB 77, 045126 (2008)
How to minimise photon momentum effects?

A. $q = 0, \Theta = 0$

B. $q \neq 0, \Theta = 0$

C. $q = 0, \Theta = 0.7$

D. $q \neq 0, \Theta = 0.7$
Photon momentum effects on Ag(001)

**Experiment**

![Experiment Graph]

**Theory**

![Theory Graph]

Venturini et al. PRB 77, 045126 (2008)
Thermal effects I

- Thermal vibrations: fundamental limit to band mapping as energy or temperature is raised

- Superposition of direct and Non-direct transitions:

\[ I(E, T) = W(T)I_{DT}(E) + (1 - W(T))I_{NDT}(E) \]

Debye-Waller factor \( W(T) \propto \exp(-\Delta k^2 \langle u^2 \rangle_T) \)

mean-square displacement \( \langle u^2 \rangle_T \)

Shevchik (1977)

- Experimental support:
  White and Fadley (1981)

- \( T \)-dependent scattering matrix \( t \) within LEED-formalism (Duke (1980))

\[ t(T, \vec{k}, \vec{k}') = t(0, \vec{k}, \vec{k}')W(T) \]
Thermal effects II

- Based on Glaubert’s theorem (1955)

\[
\langle e^{i \Delta \vec{k} \cdot \vec{u}} \rangle_T = e^{-\frac{1}{2} \langle (\Delta \vec{k} \cdot \vec{u})^2 \rangle_T}
\]

- Transfer to 1-step model of PES and inclusion of matrix element effects: Larsson and Pendry (1981)

- Forward focusing in VB-XPS: Osterwalder et al. (1990)

- Improved treatment of phonon effects on LEED state - cluster implementation: Zampieri et al. (1996)
W(110): Experiment and theory at 870eV

Plucinski, Minar et al., PRB 78, 035108
W(110): Experiment and theory at 870eV
W(110) at 5954 eV, $T = 30K$

Experiment

(a) Raw

(b) Corr.

Brillouin zone

Theory (T=0K)

Corrected for phonon-induced density-of-states-like intensity

Expt.-Ueda, Kobayashi, SPring8
Data analysis-Papp, Gray, Plucinski, C.Fadley
Theory-Minar, Braun, Ebert

Theory, wide angular scan (T=0K)
W(110) at 5954eV, $T = 100K$

- Effects of Phonons on final LEED state
  - Shevchik (1977): $I(E, T) = W(T)I_{DT}(E) - (1 - W(T))I_{NDT}(E)$
  - Zampieri et al. (1996): Cluster approach, two site scattering
  - Current approach: full multiple scattering of final state
  - XPD limit (?)
Outlook: Electron-phonon interaction

Eliasberg function of Ni

Γ – L Spectral function for Ni (kink)

KKR Phonon calculations: linear response

Self energy \( \Sigma_{el-ph}(E, \vec{k}) = \int 2 \Sigma^E i(E, \omega) \alpha^2 F(\omega, \vec{k}) d\omega \): A. Eiguren, C. Ambrosch-Draxl

include \( \Sigma_{el-ph}(E, \vec{k}) \) into KKR via Dyson Equation + Photoemission

calculations for complex systems and alloys (high-\( T_c \)-materials)
Scattering theory for dislocated atoms

- Vibrations of lattice sites uncorrelated
- Assume dislocations $\Delta \vec{R}_n$ to depend on temperature $T$ with probability $P(\Delta \vec{R}_n, T)$
- Average over dislocations using CPA alloy theory $P(\Delta \vec{R}_n, T) \Leftrightarrow$ concentration
- Combine with PES-theory for alloys (Durham)

CPA averaging $\downarrow$

Leads to proper Green’s function without artefacts (Heiglotz)!
Summary

Extension of the one-step model of photo-emission allow to deal with all aspects of HAXPES

- photon momentum
- thermal effects
- more refined models necessary
- relativistic effects
- matrix elements effects
- (magnetic) dichroism
- non-dipole contributions
- spin-resolution (SPIN-HAXPES)
Munich SPR-KKR package

Forthcoming hands on course

- KKR- hands on course
  - H. Ebert, W. M. Temmerman
  - 24.-26. June, Munich

- see: http://olymp.cup.uni-muenchen.de/ak/ebert