ARPES (and FEL)

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band structure, final and intermediate states, spectra?
Compare: energy-momentum conservation (top) with electron distribution curve (bottom)
Some examples of ARPES

- i) GaAs(110), (TiSe$_2$)  
  angular distributions
- ii) S/GaAs (001) – (2x6)  
  spectral series
- iii) Cu (111), Al(100)  
  accuracy
- iv) TiSe$_2$, NbSe$_2$  
  inelastic effects
One-Step Theory of Photoemission

outgoing photoelectron:
- LEED state
- bulk vacuum
- valence band state
- surface state

$E_f$ and $E_i$ are the Fermi level and initial state energy, respectively.
True final states Fourier decomposed (blue)

TiSe$_2$ bandstructure (black) in surface perpendicular direction

close to parabolic band, backfolded by $2\pi/c \ast n$
Side bands for higher $\Gamma$ points

GaAs (110)
Photocurrent reflects integrated product of initial with final state.
Sensitivity of photocurrent on emitting region

middle and outer region separated by a sphere of 0.3 Å radius from nucleus

detects:
interstitial at low photon energy
core region at high energy
Associate emitting region

GaAs(110)
$E_{in} = -4 \text{ eV}$
$E_{fin} = 15 \text{ eV}$
energy surfaces

photocurrent

$E_F$  $E_F+300\text{meV}$

experiment  theory
ARPES from S/GaAs(001)-2x6
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- $S_{\text{dimer}}$
- $SGa_{\text{backb}}$
- $GaAs_{\text{backb}}$
- $S_{p_z}$

Difference of EDC's for 0 and 90 azimuth

Peaks in difference due to anisotropy

$S_{\text{db}}$

S px,py orbitals

dimer bond, dangling bond
One-step photocurrent by APW: LEED

transmitted w.

interface

reflected w.

repeated slab solved for a complete set of states for the interface

Phys. Rev. B 70, 245322 (2004), Krasovskii
Potential and wavefunction for surface system

Phys. Rev. B 70, 245322 (2004), Krasovskii
Photoemission from the surface state $S_1$ on Cu (111)

$V_i = 2 \text{ eV}$

$E_f \rightarrow E_f + 2.5 \text{ eV}$

$\text{Im } k_{S_i} = 0.15 \ \text{Å}^{-1}$

$E_{S_i} - E_F = -0.55 \text{ eV}$

experiment
Louie et al. 1980
Energy Accuracy of ARPES

Total current spectroscopy

normal incidence TCS on TiTe$_2$

EXPERIMENT
(Strocov 2003)

THEORY
(E.K.)

total current

e
Inelastic Effects in Target Current Spectroscopy of NbSe$_2$

Normal Incidence Spectrum

$\nu(E)$ compared to $\int_0^\infty \text{Im}[-1/\epsilon(\omega)] \, d\omega$

Optical Properties of NbSe$_2$

Determining the optical potential from the sharpness of $dT/dE$ spectral structures.

FEL photoemission

• free electron laser (VUV-FEL, at DESY Hamburg) ➔

source of high photon intensity
  ➔ higher order terms of electromagnetic field, golden rule to be extended

lateral focus
  ➔ lateral spatial inhomogeneities, keeps with traditional golden rule
source of high photon intensity:

- quantum kinetics a la Kadanoff-Baym (M. Bonitz)
- time dependent DFT (E. Pehlke)
- higher order perturbation in $H_1(t)$ (WS)

\[
\varphi(t) = \varphi(0) - \frac{i}{\hbar} \int_0^t dt' V(t')\varphi(t') \\
V(t) = \exp\left(\frac{i}{\hbar} H_0 t\right)H_1(t)\exp\left(-\frac{i}{\hbar} H_0 t\right),
\]

\[
H_1(t) = -\frac{e}{2m} (pA + Ap) + \frac{e^2}{2m} A^2
\]

\[
A(r, t) = E \sin(qr - \omega t) / \omega
\]
photoemission in higher order

energy scheme

intensity

multiphoton transition

intermediate states
FEL photoemission

´´golden rule´´ with next order perturbation:

photocurrent \( \text{in dipole approximation (} \bullet \text{ large)} \)

\[
\begin{align*}
  j & \approx \frac{\pi}{4\hbar} \sum_i \left| \langle \varphi_f | A_0 \cdot p | \varphi_i \rangle \right|^2 \delta (E_f - E_i - \hbar \omega) + \\
  & \quad + \frac{\pi}{16\hbar} \sum_i \left( \sum_n P\left( \frac{\langle \varphi_f | A_0 \cdot p | n \rangle \langle n | A_0 \cdot p | \varphi_i \rangle}{E_f - E_n - \hbar \omega} \right) \right)^2 \delta (E_f - E_i - 2\hbar \omega)
\end{align*}
\]

\( P(\ldots) \), principal part \( |n\rangle \), intermediate states

\[
  j = j_{\text{gr}} (E_f, \hbar \omega) + j_{\text{ngr}} (E_f, \hbar \omega)
\]
extrinsic effects: renormalize outgoing LEED state by $A_0 p$

$G^{r/a}(E_f)$, $G^{a}(E_f)$, $\varphi_f$ and similarly $G^{<}(E_f - 2\hbar \omega)$, $G^{r/a}(E_f \pm \hbar \omega)$

via time independent perturbation or scattering theory
FEL photoemission

beyond 2(3…)-photon transitions:

What is the initial state, $\varphi_i$?

- time dependent DFT calculations: following electrons from ground state to the excited plasma and to photoemission (E. Pehlke)

How does the excited many-body system evolve?

- quantum kinetic equations under excitation by electromagnetic radiation (M. Bonitz)