Band Structure Interpretation of Mo(100) and W(100) Spectral Emission Properties

W. Andreas Schroeder and Gowri Adhikari

Physics Department
University of Illinois at Chicago

National Science Foundation
PHYS-1535279
Motivation: Mo(001) MTE data

\[ \phi = 3.7 \text{eV} \]

\[ \text{MTE} = \frac{1}{3} \Delta E \]

NOTE:
Deviation from expectations for \( \hbar \omega > 4.6 \text{eV} \) (\( \Delta E > 0.9 \text{eV} \))

Tunable UV radiation source

- Laser / NLO based radiation source

2 W Yb:KGW laser
250 fs, 28 MHz, 1046 nm

Nonlinear fiber (SC-3.7-975)

Detector: CCD camera
Faraday cup

Non-collinear sum frequency UV generation

-20 kV

Solenoid scan measurement
Optical parametric amplification

- OPA 1 is temperature tuned (NCPM); OPA 2 is critically phase-matched

- **Signal** wave amplification (715-945 nm)
  \[ \hbar \omega_{UV} = 4.87 - 5.29 \text{ eV} \]
  \[ = 3.68 - 4.1 \text{ eV} \]

- **Idler** wave amplification (1.17-1.95 µm)
  \[ \hbar \omega_{UV} = 4.21 - 4.61 \text{ eV} \]
  \[ = 3.0 - 3.43 \text{ eV} \]

- PLUS: \( 4\hbar \omega_{laser} = 4.75 \text{ eV} \)
  \( 3\hbar \omega_{laser} = 3.56 \text{ eV} \)

\[ 3.0 - 5.3 \text{ eV} \]

UV radiation

(\(~10 \mu W, \sim 0.5 \text{ ps}\))
One-step photoemission analysis

Exact 1-D quantum solution for triangular barrier
- Above and below threshold emission

Extension for photoemission simulation includes:

i) $p_T$ conservation

ii) Band dispersion;

$$E(p) = \frac{p_T^2}{2m_T} + \frac{p_z^2}{2m_z}$$

iii) Fermi-Dirac population distribution; $T_e$

iv) Bulk local DOS

v) Vacuum density of states

Photoemission theories: MTE

- Ag(100): $\phi = 4.36\text{eV}$, $\varepsilon_F = 5.49\text{eV}$, $T_e = 300\text{K}$, and $m^* = m_0$

Photoemission theories: QE

- Ag(100): $\phi = 4.36\text{eV}$, $\mathcal{E}_F = 5.49\text{eV}$, $T_e = 300\text{K}$, and $m^* = m_0$

\[ \text{QE} = A. (\Delta E)^n \]

Simulation results: $\epsilon_F \sim \Delta E$

- MTE for ‘Ag(100)-like’ photocathode: $\phi = 4.36\text{eV}$, $\epsilon_F = 0.5\text{eV}$, $T_e = 300\text{K}$, and $m^* = m_0$

Three regimes:

(i) $\Delta E \ll \epsilon_F$

... ‘normal’ MTE($\Delta E$) behavior

(ii) $\Delta E \sim \epsilon_F$

... cusp in MTE($\Delta E$) due to ‘E($p$) resonance’

(iii) $\Delta E > \epsilon_F$

... MTE $\sim$ constant as all available states already emitting
Simulation results: $\mathcal{E}_F \sim \Delta E$

- QE for ‘Ag(100)-like’ photocathode: $\phi = 4.36\text{eV}$, $\mathcal{E}_F = 0.5\text{eV}$, $T_e = 300\text{K}$, and $m^* = m_0$

- $\mathcal{E}_F = 0.5\text{eV}$

- Full theory

- $\mathcal{E}_F \neq A.(\Delta E)^n$

- For $\Delta E > \mathcal{E}_F$
  $\mathcal{E} \approx \text{const.}$

  ... as all available states already emitting
Band structure: Mo

Fermi surface:

‘Lens’

Emission direction

www.phys.ufl.edu/fermisurface
Band structure: Mo

- Parabolic band approximation
Mo(100): MTE

- Good agreement for $\Delta E < 0.8\text{eV}$ ($\hbar\omega < 4.5\text{eV}$)
- Other states contribute for $\Delta E > 0.8\text{eV}$
Mo(100): $\phi$ extraction from QE

- Theoretical prediction for low $\Delta E$:

\[ \text{QE} \approx A.(\Delta E)^{2.8} \]

in good agreement with measurements

$\Rightarrow \phi \approx 3.7 \text{ eV}$

Band structure: W

- Full relativistic DFT calculation

Fermi surface:

Band structure: W

- Parabolic band approximation

W(100): MTE

- Excellent agreement with experiment for $\Delta E < 0.6\text{eV}$ ($\hbar\omega < 4.5\text{eV}$)
- Qualitative agreement for $\Delta E > 0.6\text{eV}$

Diagram:
- Full theory (QE weighted)
- 1st band
- 2nd band (preliminary)
**W(100): φ extraction from QE**

- Theoretical prediction for low ΔE:
  \[ \text{QE} \approx A(ΔE)^{2.7} \]
  in good agreement with measurements
  \[ \Rightarrow \phi \approx 3.9 \text{eV} \]

Mo and W oxidation: $\phi$ reduction

- $\sim 10^{-7}$ torr vacuum chamber

- Observed $\Delta\phi \approx -0.8$ eV work function reduction

- Oxide thickness on Mo and W known to be $1.0(\pm0.5)$ ML

<table>
<thead>
<tr>
<th></th>
<th>$\phi$ (eV) (clean surface)</th>
<th>$\phi$ (eV) (measured)</th>
<th>$\Delta\phi$ (eV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mo(100)</td>
<td>4.46(±0.11)</td>
<td>3.7(±0.05)</td>
<td>-0.76</td>
</tr>
<tr>
<td>W(100)</td>
<td>4.70(±0.06)</td>
<td>3.9(±0.05)</td>
<td>-0.8</td>
</tr>
</tbody>
</table>

Negligible effect of $\sim 1$ ML surface oxide on MTE($\hbar\omega$) measurements

... c.f. UV absorption depth $\alpha^{-1} \approx 6$-7nm $\approx 40$ ML

Summary

Band structure interpretation of Mo(100) and W(100) emission
– MTE(ℏω) and QE(ℏω) data using tunable 3.0-5.3eV UV radiation source

- One-step photoemission simulation
  - Extended an exact 1-D triangular barrier quantum transmission solution:
    \( p_T \) conservation, local DOS, \( T_e \), AND vacuum state physics

- W(100) and Mo(001): MTE(ℏω) consistent with analysis
  - Parabolic bands approximation (\( m_z \) and \( m_T \)) to real emission states

- QE = \( A(ℏω−\phi)^n \); power \( n \) is function of emission band(s) details
  - \( (QE)^{1/n} \) vs. \( ℏω \) plot \( \Rightarrow \phi \) extraction

FUTURE WORK:
Direct evaluation of MTE (and absolute QE?) from band structure
– (\( p, E, \) local DOS) from DFT \( \rightarrow \) One-step photoemission simulation
Thank You!
Mo(001): Emission bands

- DFT evaluation

- Four emission bands for Mo(001)
  - 3 ‘electron-like’
  - 1 ‘hole-like’

- $p_{T,\text{max.}} = \sqrt{2m_0\Delta E} < p_F$

$\Delta E = 0.23$ eV

⇒ Photoemission *not* band dispersion limited

Motivation

Tunable UV radiation source
- Laser / Nonlinear optics based ($\hbar\omega = 4.2 - 5.3$ eV)

Mo(001)
- Mean transverse energy (MTE) vs. $\hbar\omega$
- QE ($\eta_{PE}$) vs. $\hbar\omega$; $\phi$ extraction
- Comparison to ‘one-step’ photoemission model

PbTe(111)
- Low $m^*$ emitter $\Rightarrow$ ultralow MTE …
- MTE and $\eta_{PE}$ vs. $\hbar\omega$
- Comparison to ‘one-step’ photoemission model
- Deleterious effects: $E_{\text{depletion}}$, band non-parabolicity, CB emission …

Summary