Spectral Intrinsic Emittance Measurements of Mo(001) and PbTe(111) Photocathodes

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Outline

- 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 (η_{PE}) vs. $\hbar\omega$; ϕ extraction
  - Comparison to ‘one-step’ photoemission model
- PbTe(111)
  - Low m^* emitter $\Rightarrow$ ultralow MTE …
  - MTE and η_{PE} vs. $\hbar\omega$
  - Comparison to ‘one-step’ photoemission model
  - Deleterious effects: $\mathbf{E}_{depletion}$, band non-parabolicity, CB emission …
- Summary

Motivation

GOAL:

Realization of robust and practical low intrinsic emittance planar photocathodes

- *Robust* ⇒ Performance at $\leq 10^{-7}$ torr
- *Practical* ⇒ 'Readily' available with $\phi \leq 4.6\,\mathrm{eV}$ ($\rightarrow 3\,\mathrm{eV}$)
- *Low intrinsic emittance* ⇒ Ideally, MTE $\leq 50\,\mathrm{meV}$ ($\rightarrow 10\,\mathrm{meV}$)

REQUIREMENTS:

- Tunable UV radiation source
  - *In situ* ϕ extraction from $\eta_{PE}(\hbar\omega)$; MTE vs. $\hbar\omega$
- Photoemission theory: one-step model
  - $\eta_{PE} = A(\hbar\omega - \phi)^n$; MTE($\hbar\omega$)
- DFT evaluation of photocathode emission states

  ⇒ ONLY ***single-crystal*** photocathodes !

Tunable UV radiation source

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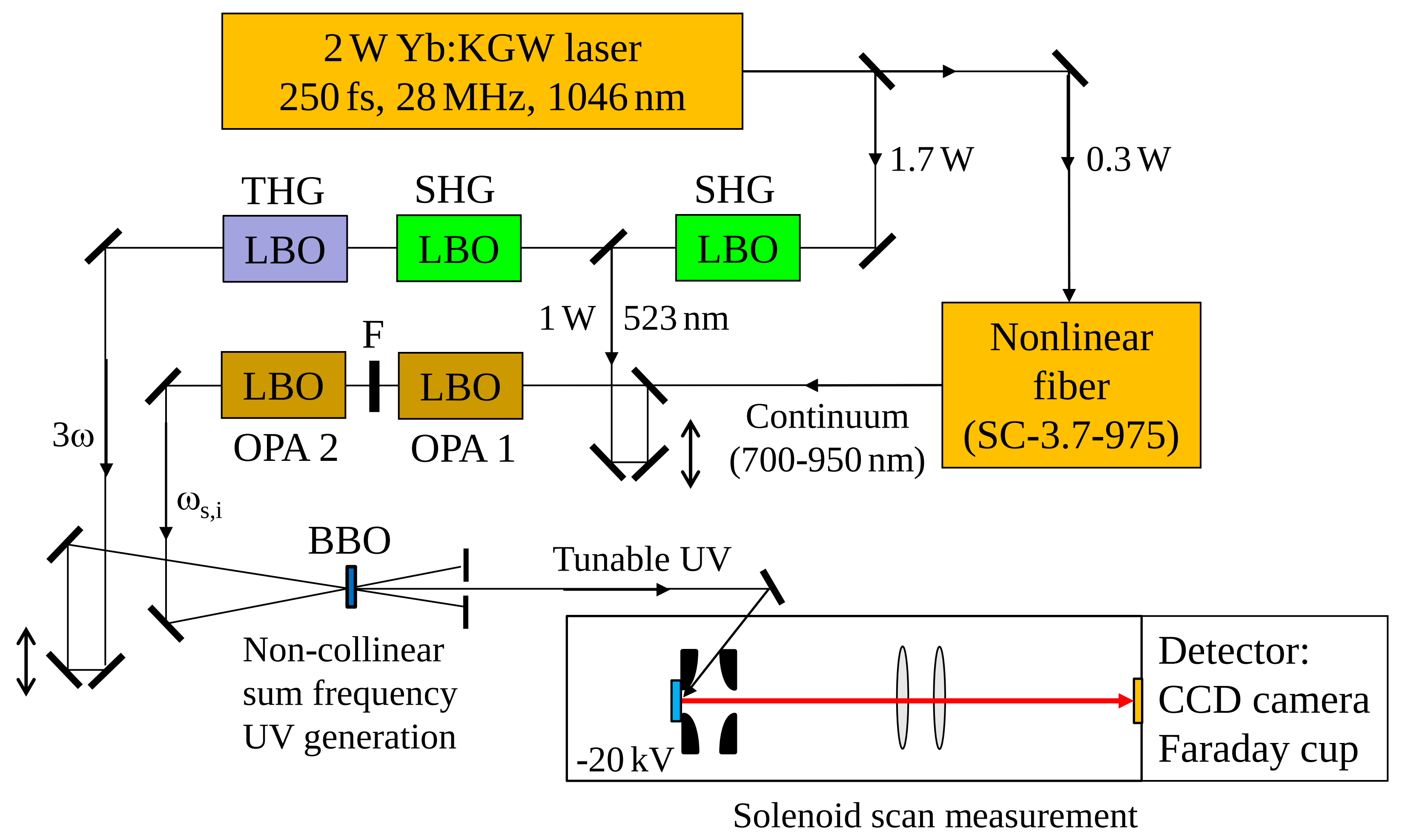
– Laser / NLO based radiation source



Optical parametric amplification

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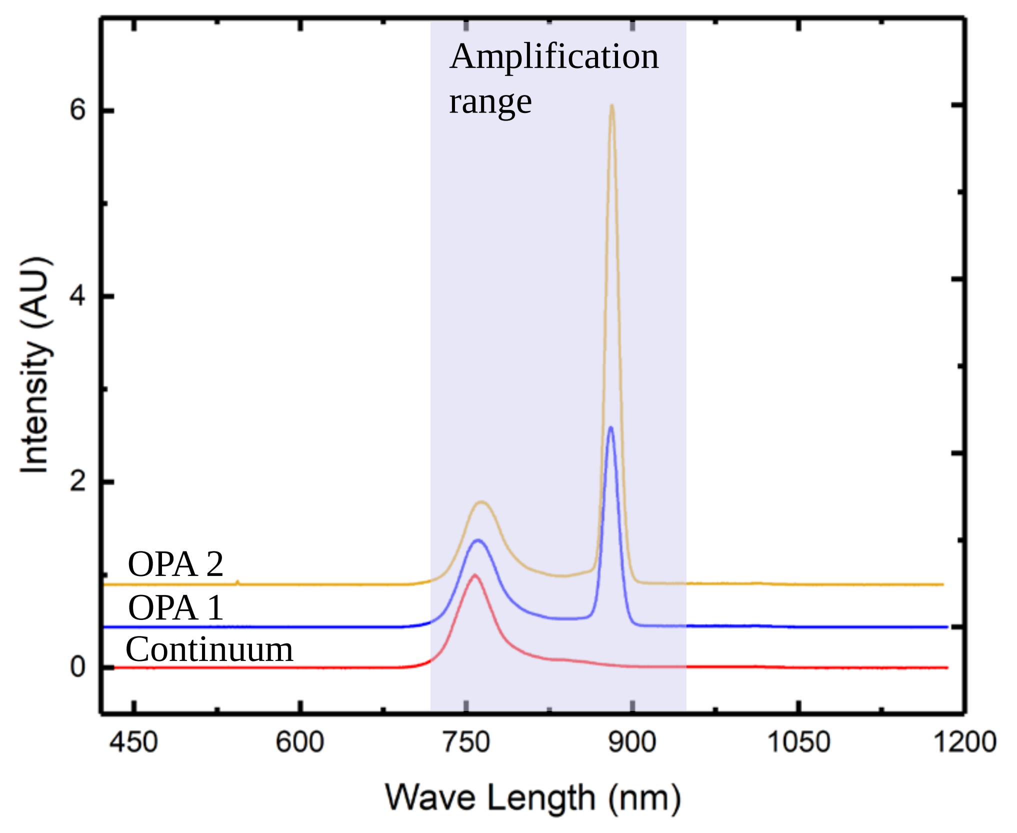
– OPA 1 is temperature tuned (NCPM); OPA 2 is critically phase-matched



- *Signal* wave amplification (715-945 nm)

  $\Rightarrow \hbar\omega_{UV} = 4.87 - 5.29$ eV

- *Idler* wave amplification (1.17-1.95 μm)

  $\Rightarrow \hbar\omega_{UV} = 4.21 - 4.61$ eV

- PLUS: $4\hbar\omega_{laser} = 4.75$ eV

4.2 – 5.3 eV
UV radiation
(~10 μW, ~0.5 ps)

Mo(001): MTE and η_{PE} data

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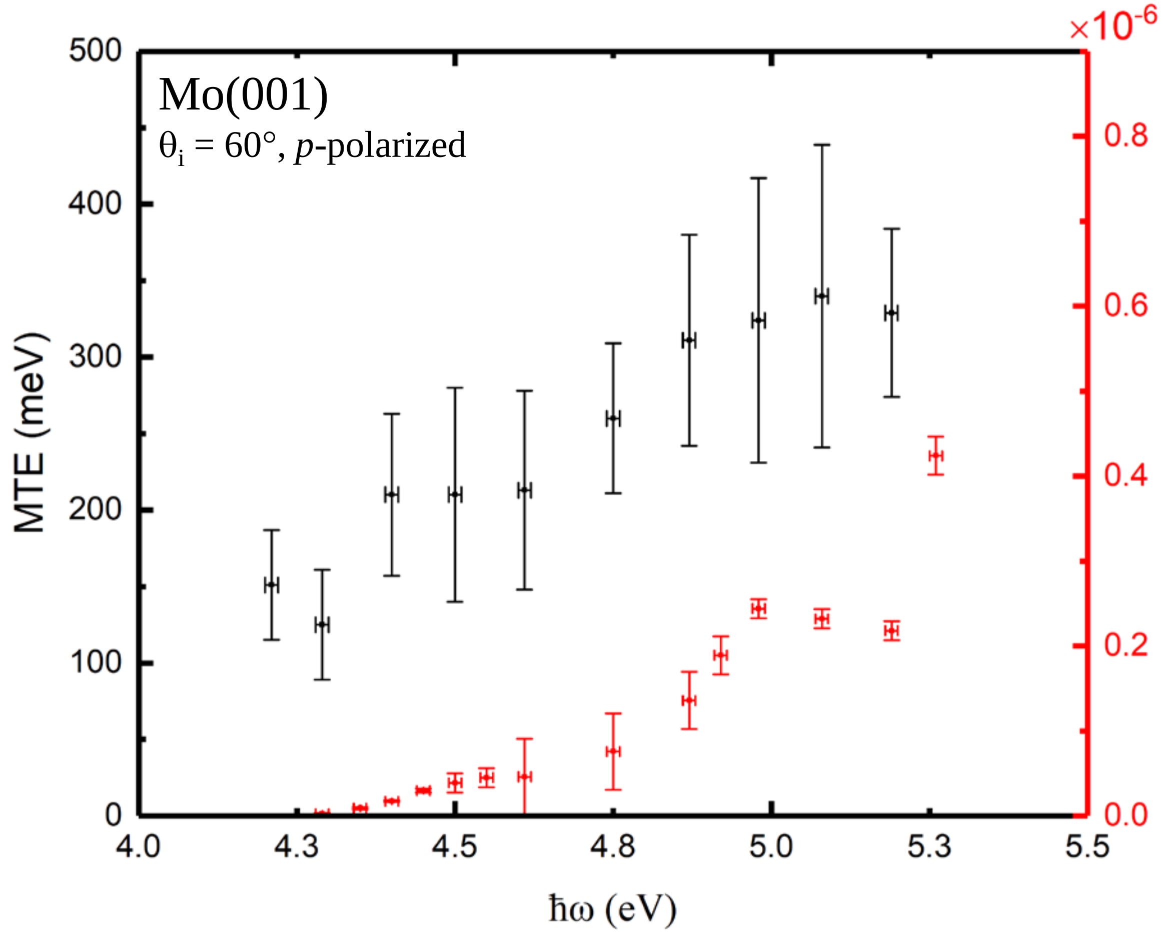

2-step data analysis:

(i) Extract ϕ using $\eta_{PE} = A(\hbar\omega - \phi)^{n}$

(ii) Use ϕ value to compare MTE(ħω) to theory

Metals: ‘One-step’ photoemission

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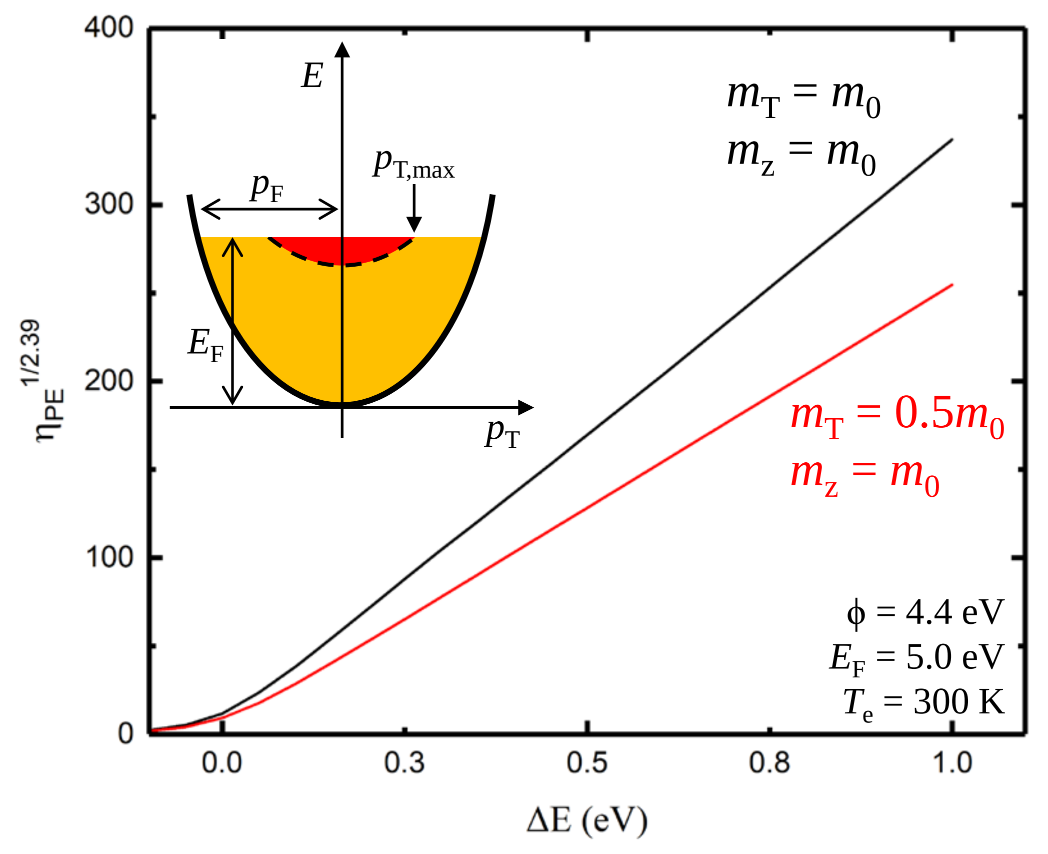
– Photoemission efficiency for ‘electron-like’ metals; $m^*E_F > m_0(\hbar\omega - \phi)$



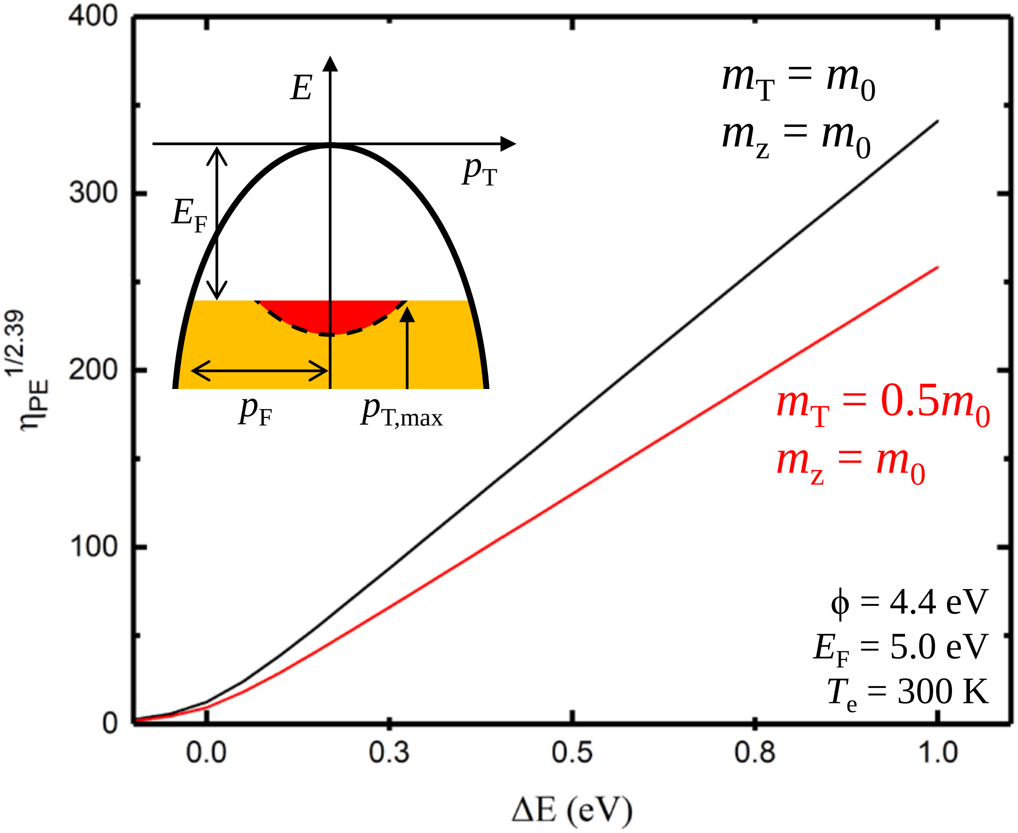
- One-step parabolic band PE model predicts

$$\eta_{PE} = A(\hbar\omega - \phi)^{2.39}$$

- $\eta_{PE}(\hbar\omega)$ power law ***not*** dependent on m^*
- Magnitude of η_{PE} is dependent on m^*
  – Density of states
- Effects of ‘thermal tail’ emission evident around $\Delta E = 0$

Metals: ‘One-step’ photoemission

– Photoemission efficiency for ‘hole-like’ metal; $m^*E_F > m_0(\hbar\omega - \phi)$



- One-step parabolic band PE model again predicts

$$\eta_{PE} = A(\hbar\omega - \phi)^{2.39}$$

- $\eta_{PE}(\hbar\omega)$ power law ***not*** dependent on m^*
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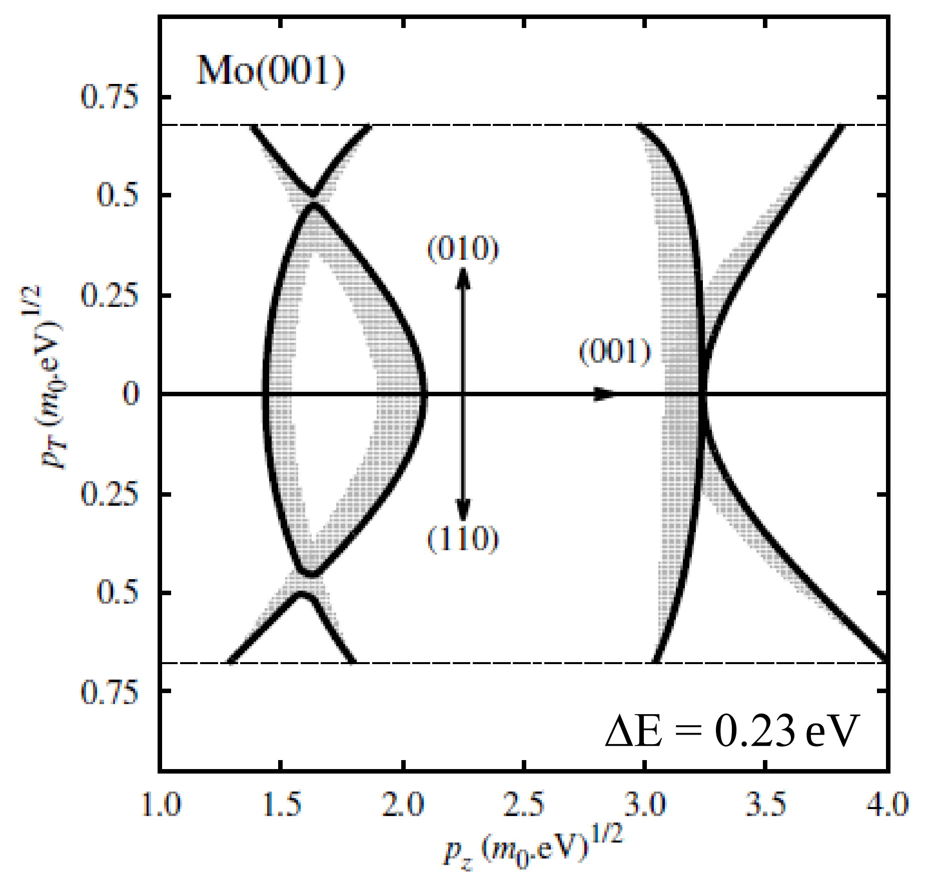
  – Density of states
- Effects of ‘thermal tail’ emission evident around $\Delta E = 0$

Mo(001): Emission bands

– DFT evaluation



- Four emission bands for Mo(001)
  - – 3 ‘electron-like’
  - – 1 ‘hole-like’
- $p_{T,\text{max.}} = \sqrt{2m_0\Delta E} < p_\text{F}$

$\Rightarrow$ Photoemission ***not*** band dispersion limited

T. Li et al., *J. Appl. Phys.* **117**, 134901 (2015)

Mo(001): ϕ extraction

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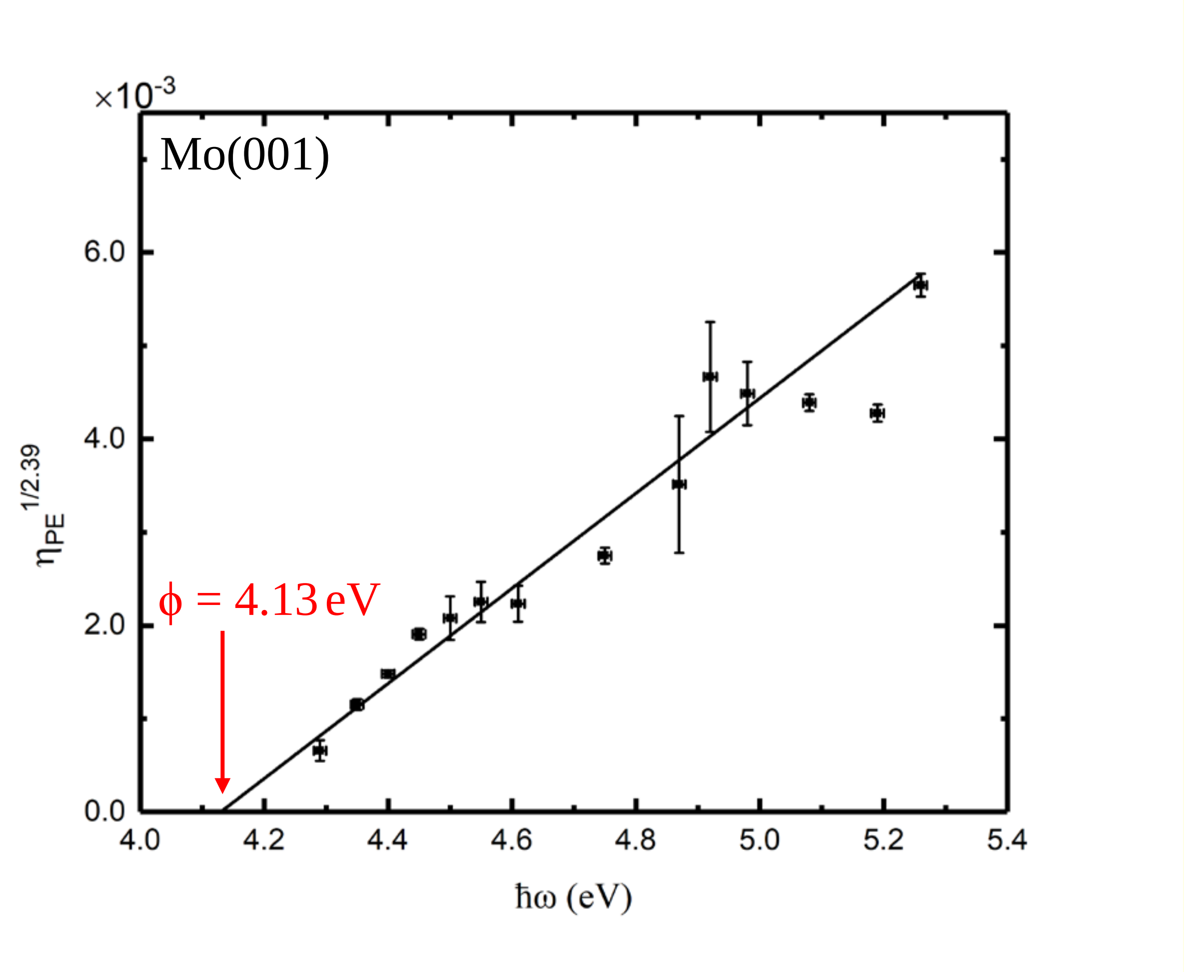

- Theoretical power law ($n = 2.39$) consistent with $\eta_{PE}(\hbar\omega)$
- Extracted value of ϕ = 4.1(±0.1) eV for Mo(001) lower than accepted value of 4.3 eV
  – Photocathode ***not*** atomically flat
    ⇒ $\Delta\phi(x,y)$; $\phi_{(001)}$ a local maximum
    ⇒ Surface roughness (slope effect)
  – Photocathode ***not*** clean at ~10^{-7} torr
    ⇒ Oxidation, etc.

G.W. Gobeli & F.G. Allen, *Phys. Rev.* **137**, A245-A254 (1965)

Mo(001): MTE(ħω)

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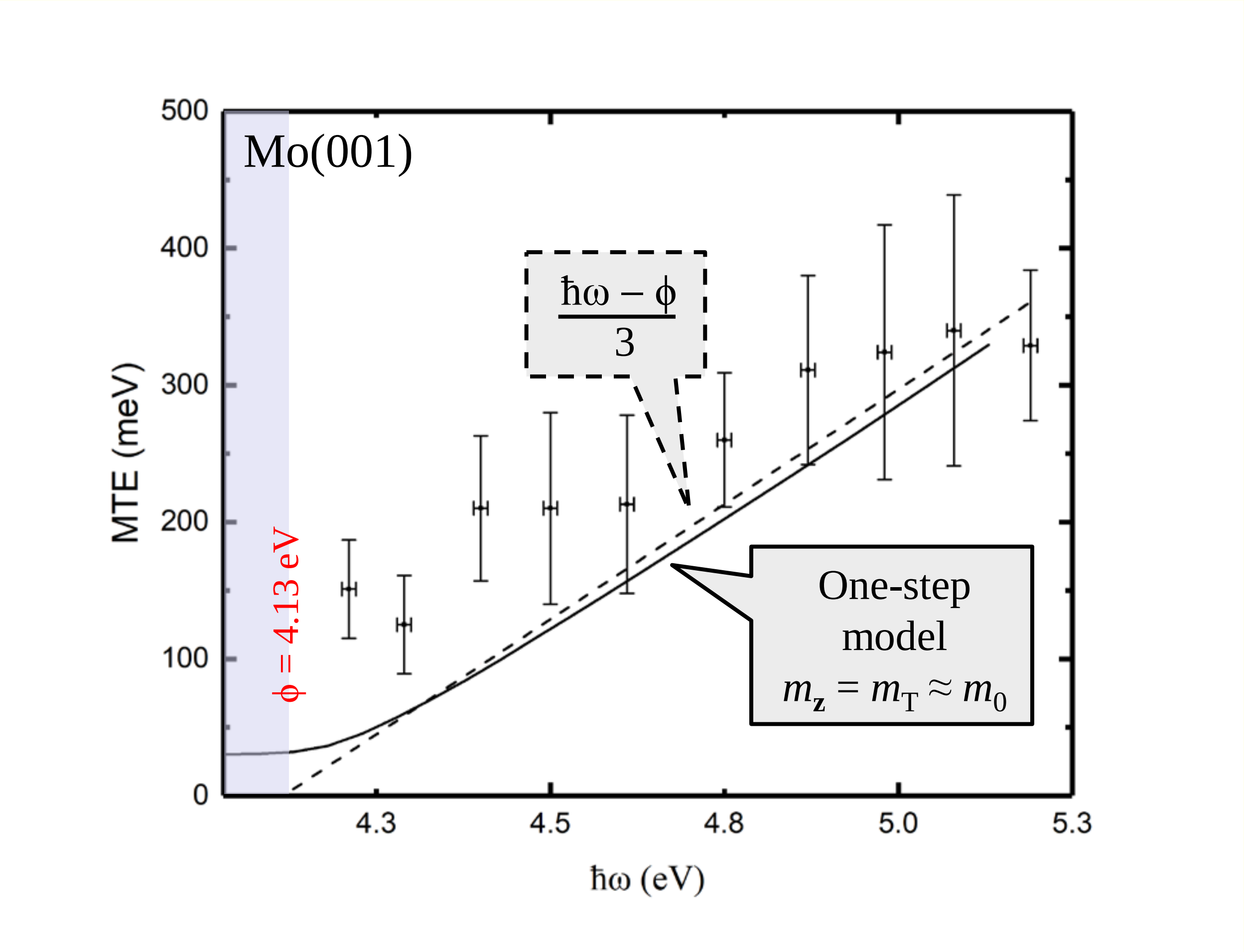
– Experiment (preliminary analysis) vs. theory



- Measurements consistent with theory for $\Delta E > 0.4\,\mathrm{eV}$
- $\mathrm{MTE}_{\mathrm{expt.}} > \mathrm{MTE}_{\mathrm{theory}}$ for $\Delta E < 0.4\,\mathrm{eV}$
  - – Absorbate (e.g. oxide) contributions?
  - – Surface roughness?
  - – Laser beam pointing instability?
  - – …

D.H. Dowell & J.F. Schmerge, *Phys. Rev. ST – Acc. & Beams* **12** (2009) 074201
K.L. Jensen et al., *J. Appl. Phys.* **107** (2010) 014903

PbTe Band Structure

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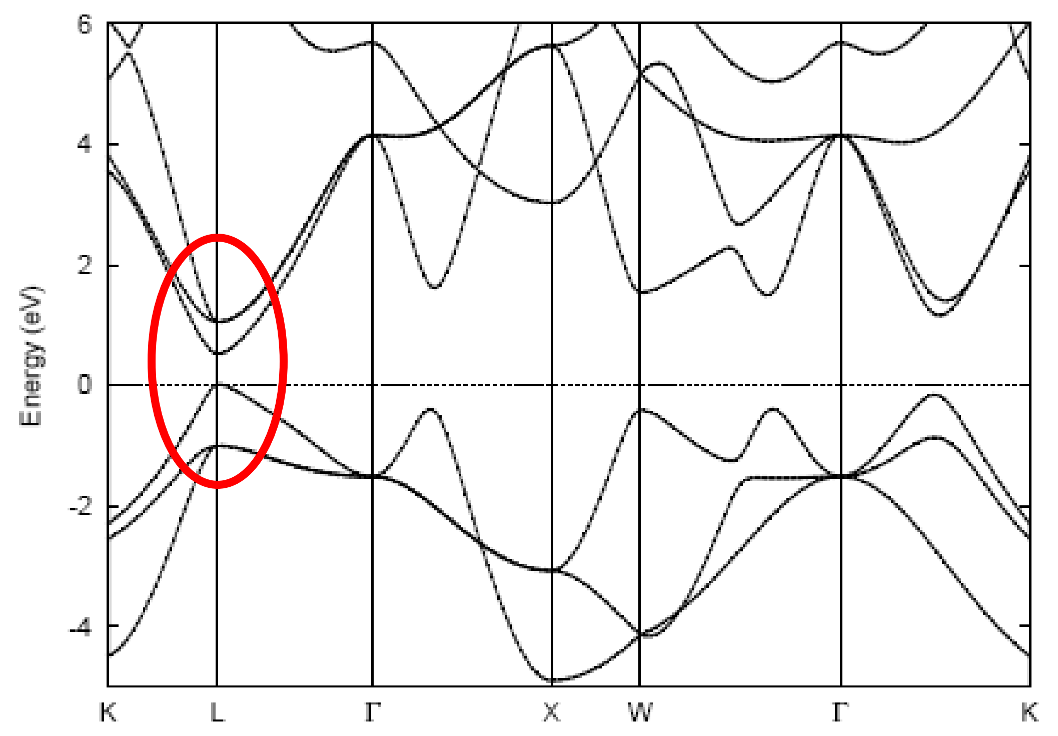
– Evaluation using QUANTUM ESPRESSO

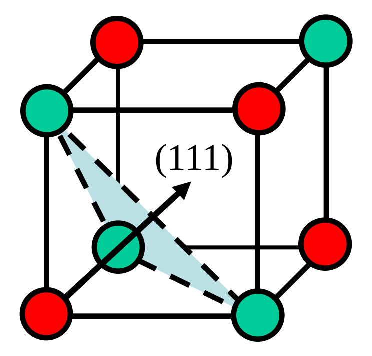


- Lightly *p*-type PbTe crystal

  ⇒ E_F at top of VBM at L point of Brillouin zone

- Very low hole mass ($m^* = 0.022m_0$) transverse to Γ-L direction

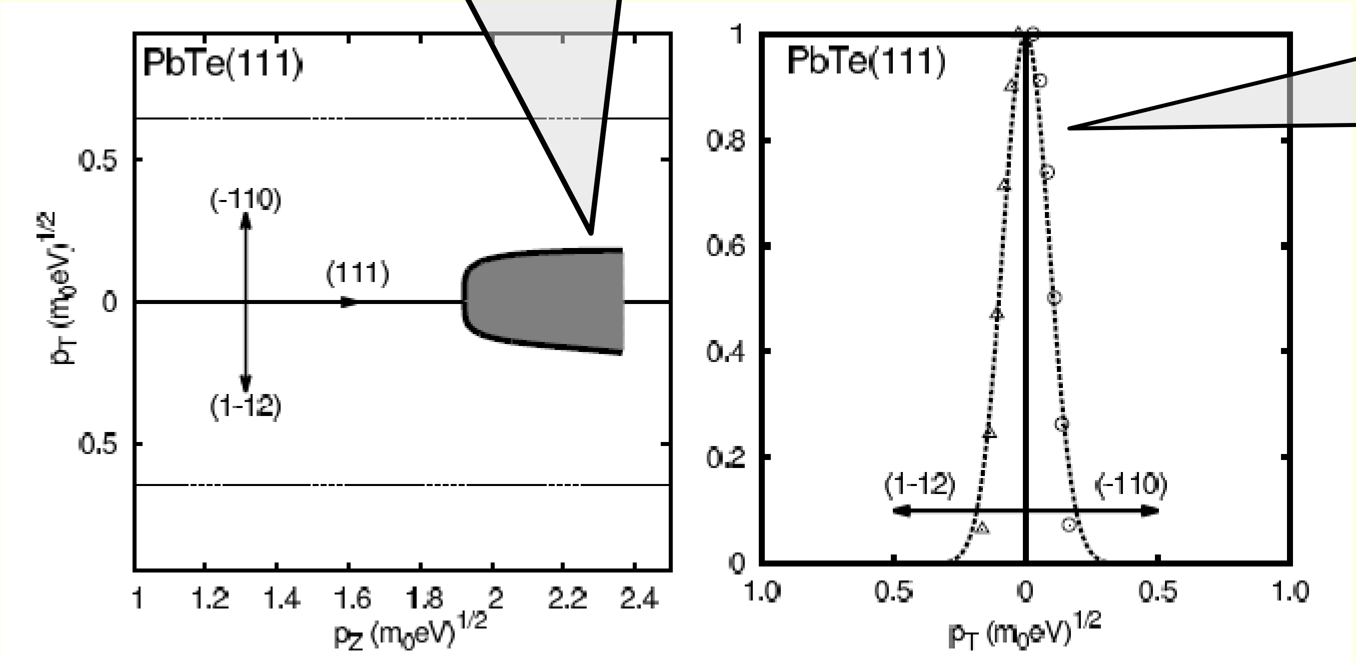
  ⇒ Band restriction on p_T for (111)-face emission



NOTE:
For emission from (111) face,
no CBM exist above 2 eV at L point.

p–PbTe(111): Theoretical MTE

– ‘One-step’ photoemission from L-point VBM with $m^* = 0.022m_0$



Emitting states for $\Delta E = 0.3\,\text{eV}$

E, $\mathbf{p}_T$ conservation
PLUS
Barrier transmission,
$T(p_z,p_{z0})$

DFT-based photoemission prediction:

$\text{MTE} \approx 10\,\text{meV}$ for $\Delta E = 0.3\,\text{eV}$

T. Li et al., *J. Appl. Phys.* **117**, 134901 (2015)

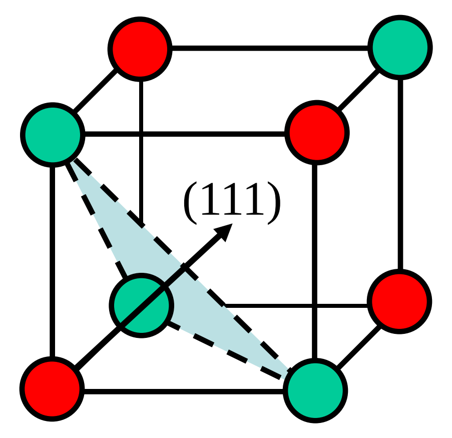
p–PbTe(111): $\phi_{(111)}$

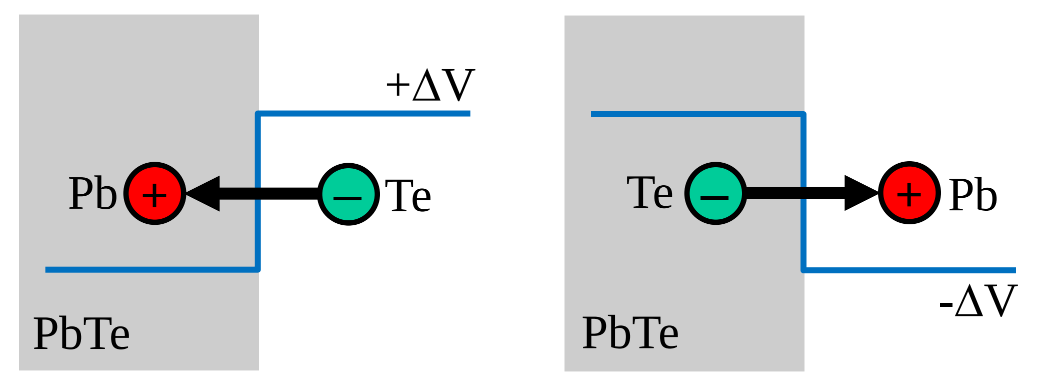
– Evaluation using thin slab technique

- Rock salt (simple cubic) crystal structure of PbTe

  ⇒ Pb or Te terminated regions on (111) surface

  ∴ Different ϕ due to surface dipole orientations





DFT-based thin-slab prediction:

$\phi_{(111),\mathrm{Pb}} \approx 4.21\,\mathrm{eV};\ \phi_{(111),\mathrm{Te}} \approx 4.53\,\mathrm{eV}$

C. J. Fall et al., *Journal of Physics: Condensed Matter* **11**, 2689 (1999)

p-PbTe(111): MTE and η_{PE} data

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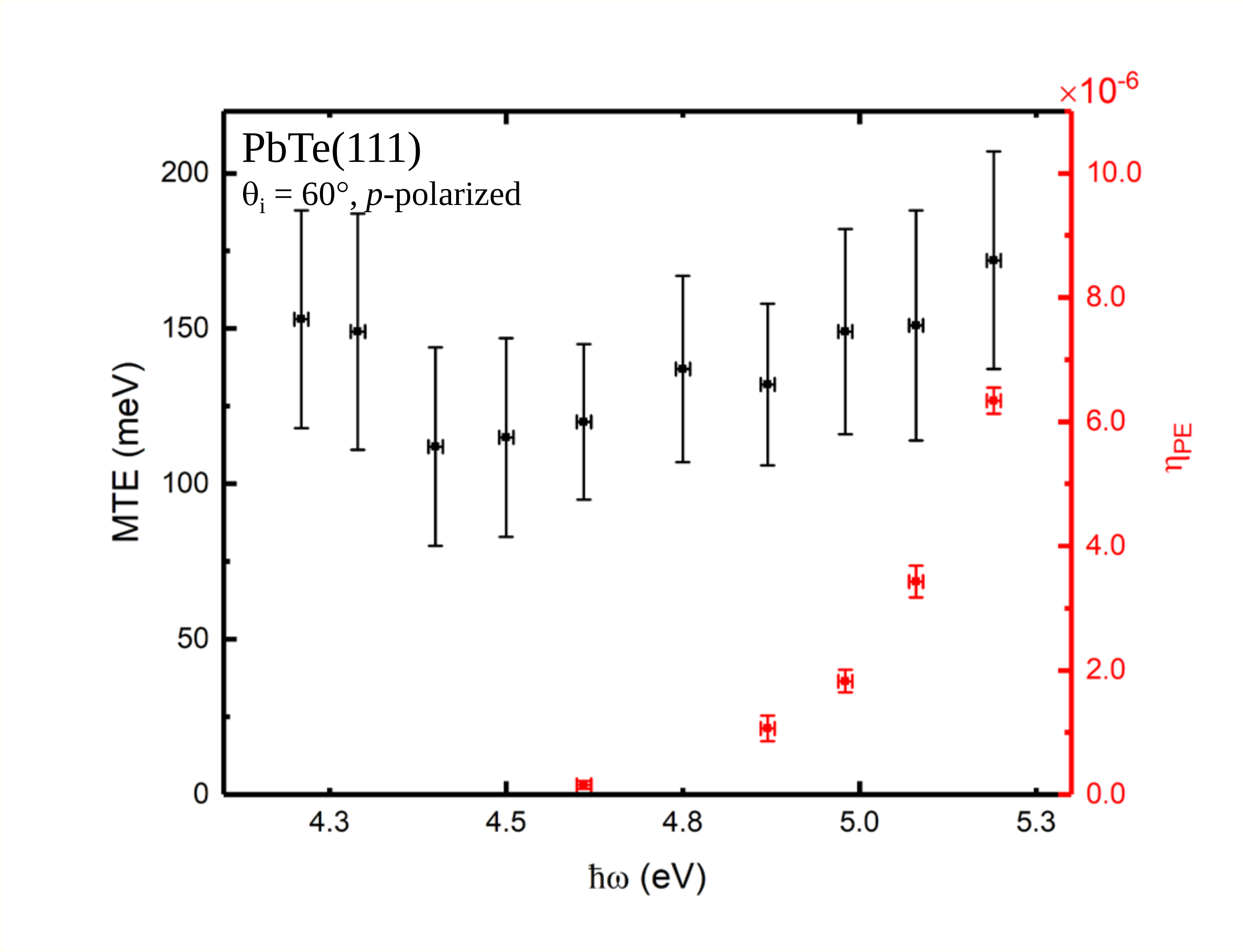

2-step data analysis:

(i) Extract ϕ using $\eta_{PE} = A(\hbar\omega - \phi)^n$

(ii) Use ϕ value to compare MTE($\hbar\omega$) to theory

Semiconductor: One-step model

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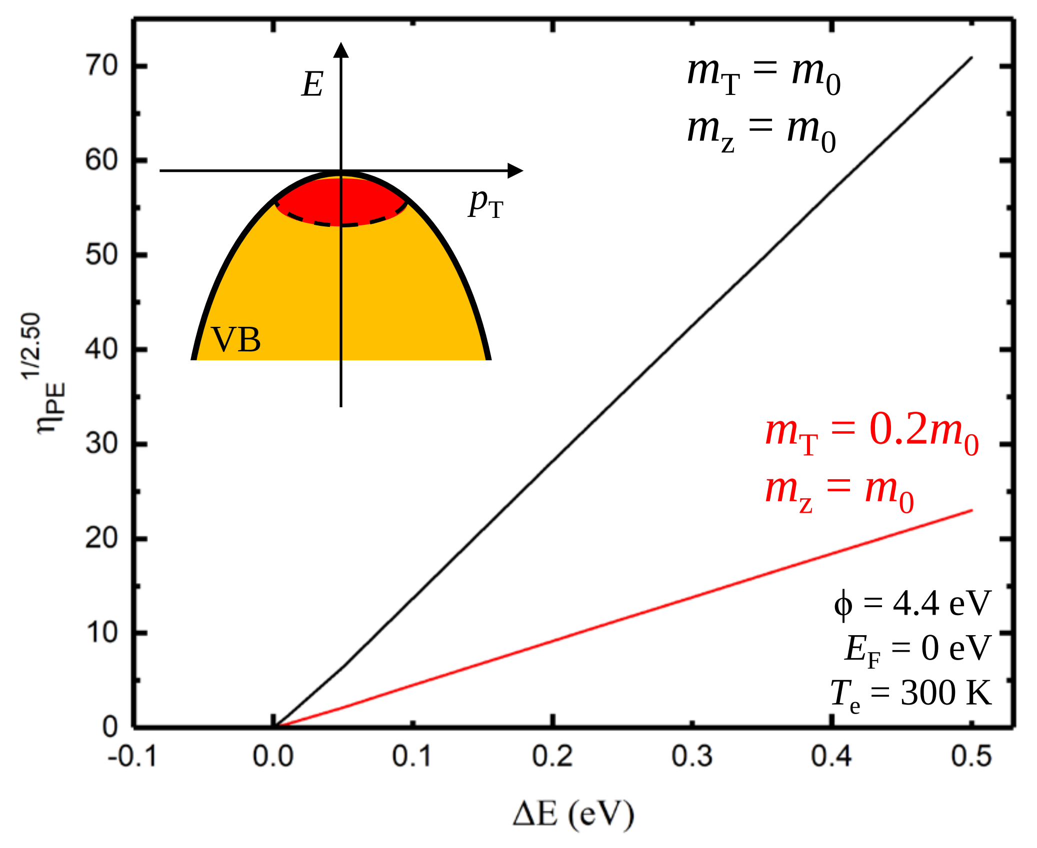
– Photoemission efficiency for a semiconductor: VB emission

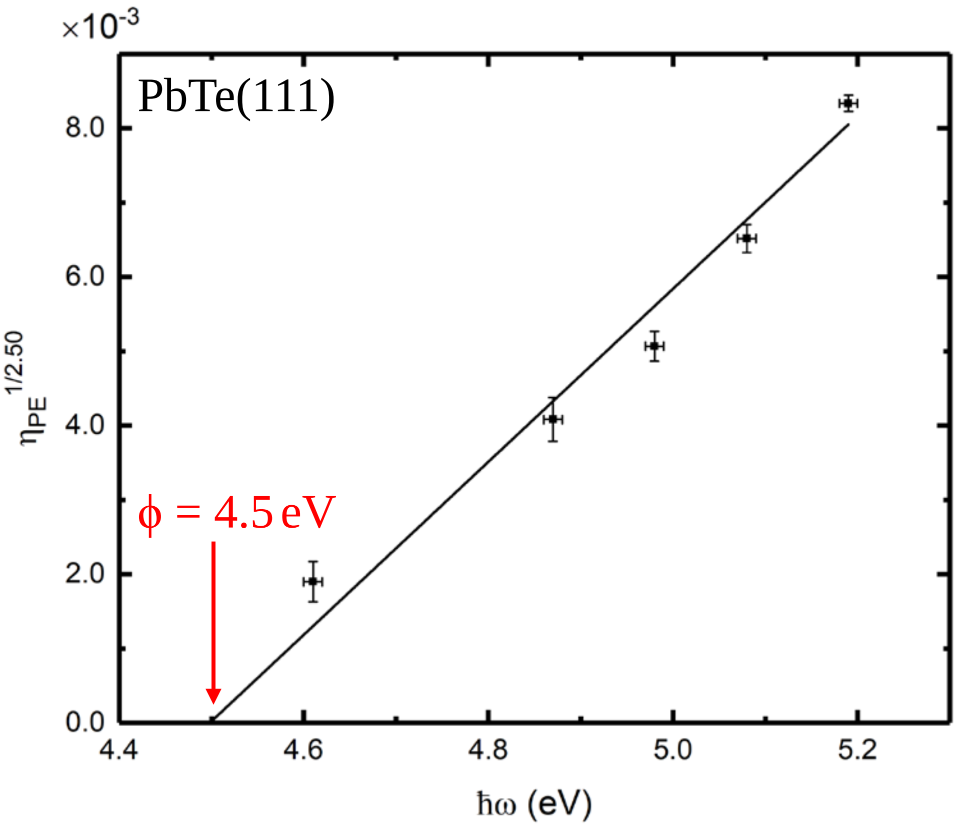


- One-step parabolic band PE model predicts

$$\eta_{PE} = A(\hbar\omega - \phi)^{2.5}$$

- $\eta_{PE}(\hbar\omega)$ power law ***not*** dependent on m^*
- Magnitude of η_{PE} is dependent on m^*
  – Density of states
- No temperature effects around $\Delta E = 0$ due to ***filled*** VB

p-PbTe(111): ϕ extraction



- Theoretical power law ($n = 2.5$) consistent with $\eta_{PE}(\hbar\omega)$

- Extracted value of ϕ = 4.5(±0.1) eV for PbTe(111) consistent with DFT thin-slab evaluated values:

  $\phi_{(111),Pb} \approx 4.21\,\text{eV}$
  $\phi_{(111),Te} \approx 4.53\,\text{eV}$

  – Mono-layer oxidative dipole similar to Te termination …

G.W. Gobeli & F.G. Allen, *Phys. Rev.* **137**, A245-A254 (1965)

p-PbTe(111): MTE(ħω)

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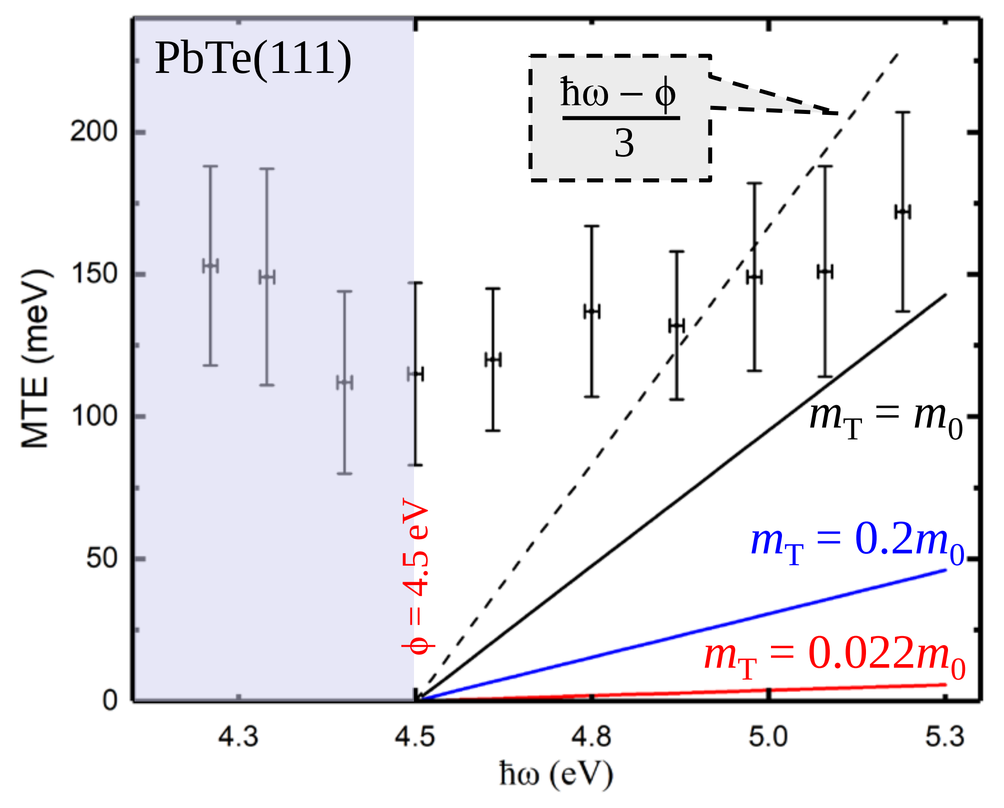
– Experiment (preliminary analysis) vs. theory



- Measurements ***inconsistent*** with theory for low m^* (p_T restricted) emitter
- Two domains:

  (i) Filled VB emitter
  for $\hbar\omega > \phi$
  BUT … larger m^* ?

  (ii) Weak emission
  for $\hbar\omega < \phi$!
  $\Rightarrow$ Additional emitting states

D.H. Dowell & J.F. Schmerge, *Phys. Rev. ST – Acc. & Beams* **12** (2009) 074201
K.L. Jensen et al., *J. Appl. Phys.* **107** (2010) 014903

p-PbTe(111): ħω > ϕ

– $N_A \approx 10^{16}\,\mathrm{cm}^{-3}$, $E_g = 0.34$ eV, $\varepsilon_r \approx 400$, ionic charge $q = 0.2e$, $a = 6.46$ Å

Larger than expected MTE:

(i) Band non-parabolicity
$m^* = 0.022m_0$ only for $\Delta E < 0.1$ eV
– Accounted for in DFT-based simulations

(ii) Band structure distortion due to $\mathbf{E}_{\mathrm{depletion}}$

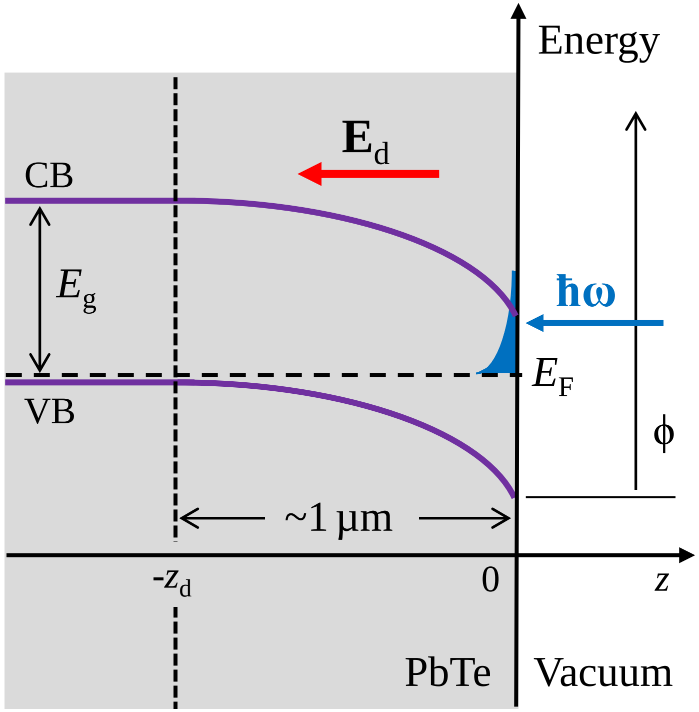
$$E_d = \sqrt{\frac{eN_A E_g}{\varepsilon_0 \varepsilon_r}} \approx 10\,\mathrm{kV/m} \text{ at } z = 0$$

Induced polarization, $P = Nqx = \varepsilon_0(\varepsilon_r\text{-}1)E_d$

$$\Rightarrow \frac{x}{a} \approx \left(\frac{2\varepsilon_0\varepsilon_r a^2}{q}\right)E_d \sim 10^{-3} \qquad \ldots \text{ as } N = \frac{1}{2a^3}$$

(iii) Phonon scattering: Absorption/emission by emitted electrons negates p_T conservation (c.f. indirect VB to CB absorption in Si)

(iv) PbTe is piezoelectric …



… $z_d \gg \alpha^{-1}_{UV} \approx 20$ nm

p-PbTe(111): ħω < ϕ

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Weak ($\eta_{PE} < 10^{-8}$) emission states for ħω < ϕ:

(i) Optically-active surface states

⇒ Filled surface states below E_F

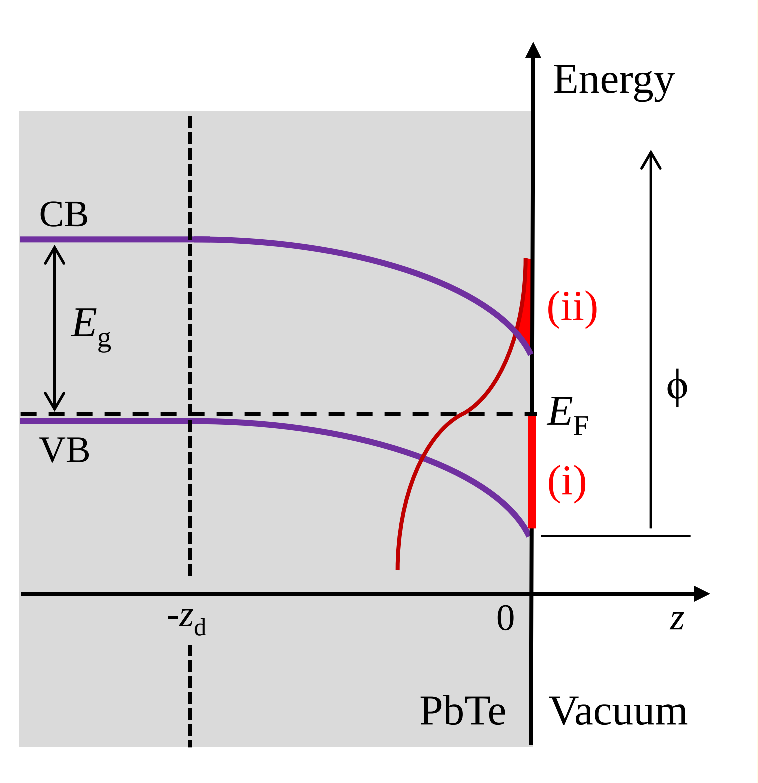
– Expect termination for ħω < 4.2 eV

(ii) CB emission

– E_F pinned near mid-gap at surface

⇒ Population of CB by thermal tail of Fermi-Dirac distribution ($E_g/2 \approx 7k_BT_e$ at 300 K)

PLUS: Optical absorption populating CB (T_e > 300 K ?)



Summary

Investigation of oriented ***single-crystal*** emitters

GOAL: Robust and practical low intrinsic emittance planar photocathodes

- Laser/NLO-based tunable UV radiation source
  - 4.2 - 5.3 eV: $\omega_{UV} = 3\omega + \omega_{s,i}$ [$\omega_{UV} = 2\omega + \omega_{s,i}$ extension $\Rightarrow$ 3.0 - 4.1 eV]
  - MTE($\hbar\omega$) (solenoid scan) and $\eta_{PE}(\hbar\omega)$ (Faraday cup)
- $\eta_{PE} = A(\hbar\omega - \phi)^{\boldsymbol{n}}$
  - Emission band(s) details determine power ***n***
  - $(\eta_{PE})^{1/\boldsymbol{n}}$ vs. $\hbar\omega$ plot $\Rightarrow$ *In situ* ϕ extraction
- MTE($\hbar\omega$) analysis requires $\phi_{\textit{in situ}}$
  - Mo(001): MTE($\hbar\omega$) close to theoretical predictions
    Surface roughness/absorbate state issues for $\Delta E < 0.5$ eV ?
  - PbTe(111): MTE larger than expected for $m^* = 0.022m_0$
    (i) $\hbar\omega > \phi$; band non-parabolicity and distortion ($\mathbf{E}_d$), phonons, …
    (ii) $\hbar\omega < \phi$; surface state and CB emission

Thank You!