DESY, Hamburg, August 10, 2015.

UIC

# Photocathode Physics: Band Structure and Emittance


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DE-FG52-09NA29451

# Space-time resolution

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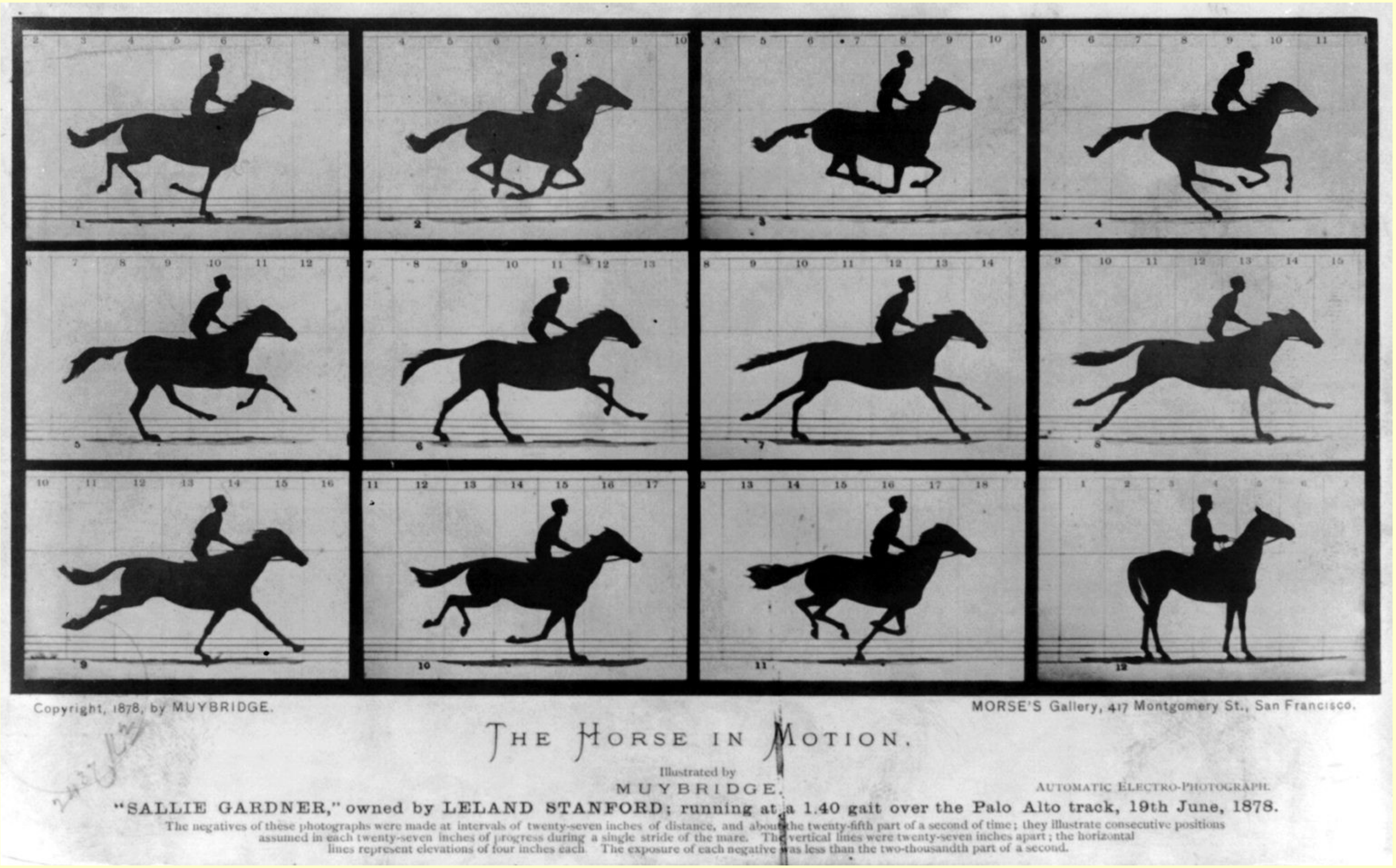


E. Muybridge (photographer, 1878): $\delta t = 0.002$s and $\delta x \sim 10$cm

$\Rightarrow$ Space-time resolution, $\delta x . \delta t \sim 1$ mm.second

# Dynamic TEM

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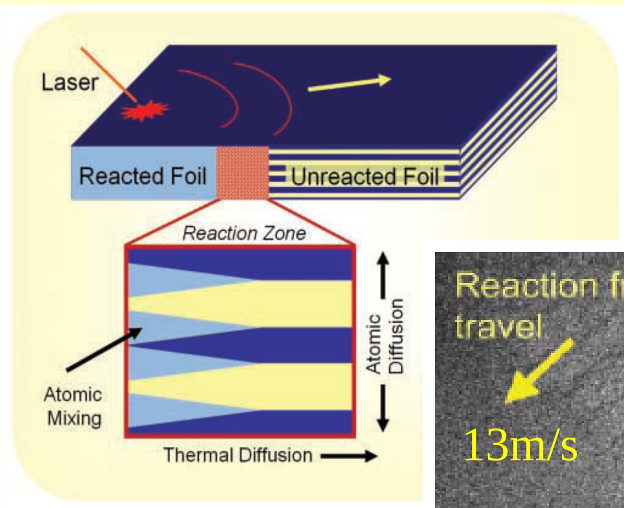
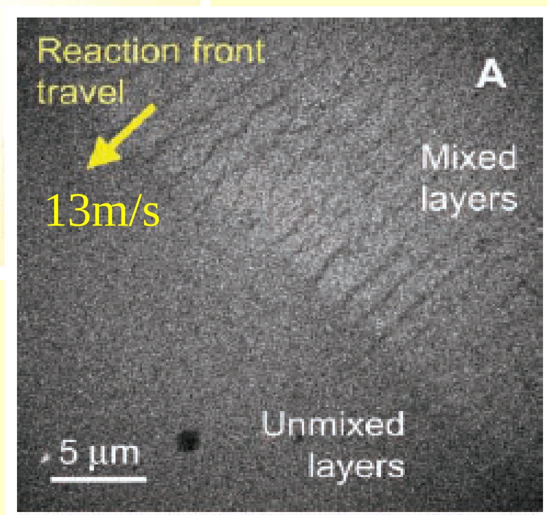
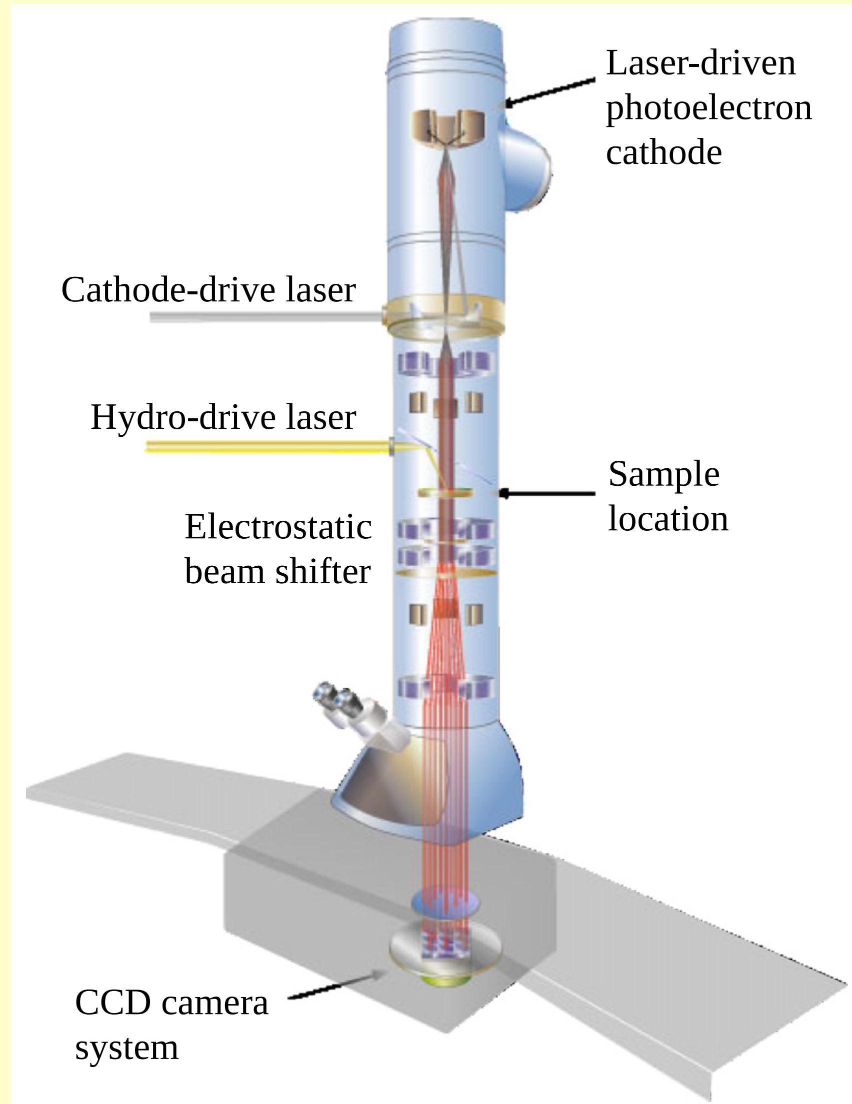
– LLNL DTEM: Single-shot *imaging*

- Laser-initiated reaction in Al/Ni reactive multi-layer foil (RMLF):




Dendritic features NOT found in post-mortem specimens




$\delta x.\delta t \approx$ 10nm.10ns ~ 100 nm.ns

J.S. Kim et al., *Science* **321** (2008) 1472

# Ultrafast X-ray diffraction

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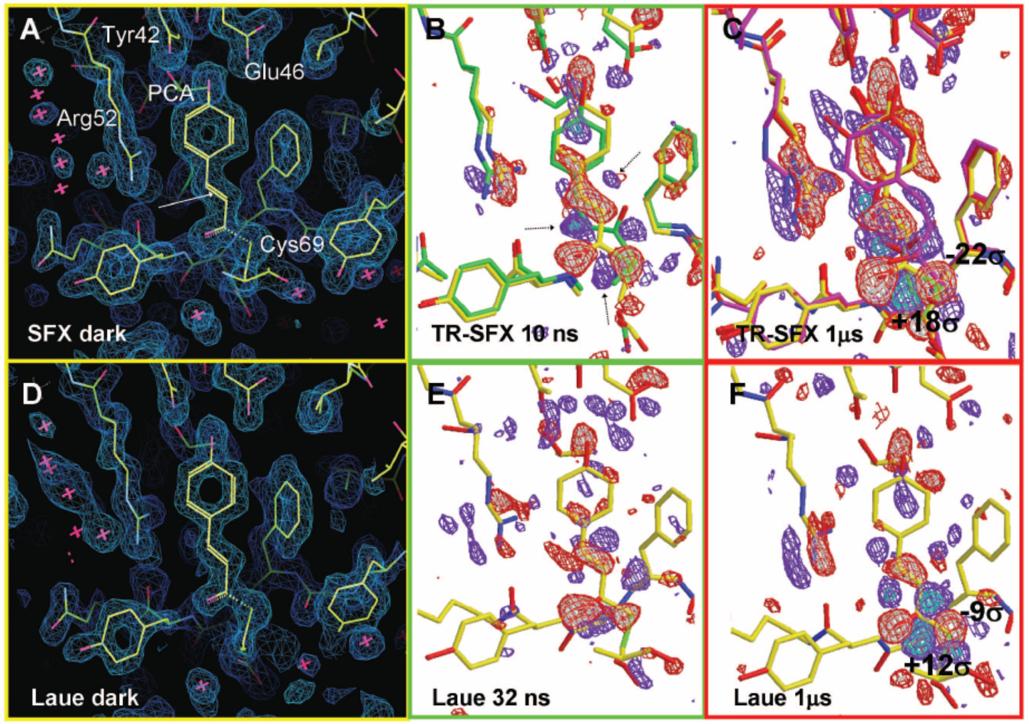
– SLAC X-ray free electron laser (XFEL)

- Reaction intermediates of photoactive yellow protein:




$\delta x.\delta t \approx 1.6Å.100ps > 10^{-2}$ nm.ns

J. Tenboer et al., *Science* **346** (2014) 1242

# Ultrafast electron diffraction (UED)

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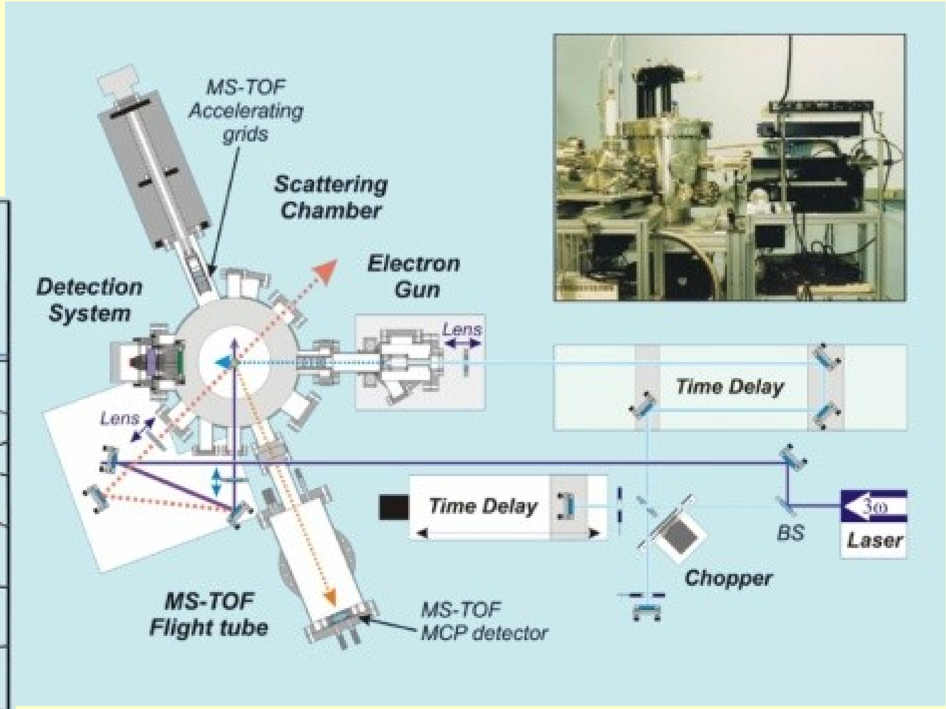
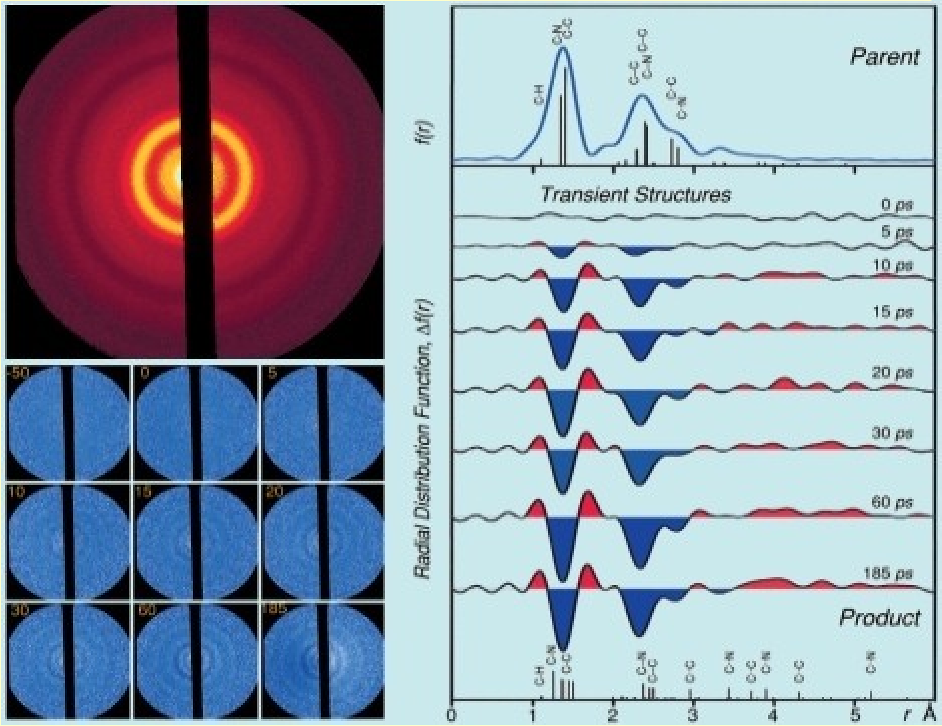
– A. Zewail: 1999 Nobel Prize in Chemistry

- Pump-probe technique:
  - Laser pulse initiates perturbation
  - Time-delayed electron pulse monitors changes




Modern *single-shot* UED systems:
$\delta x . \delta t$ < 1Å.100fs = $10^{-5}$ nm.ns


H. Ihee et al., *Science* **291** (2001) 458
V.A. Lobastov et al., *J. Phys. Chem. A* **105** (2001) 11159

# UED: Spatial Resolution Limit

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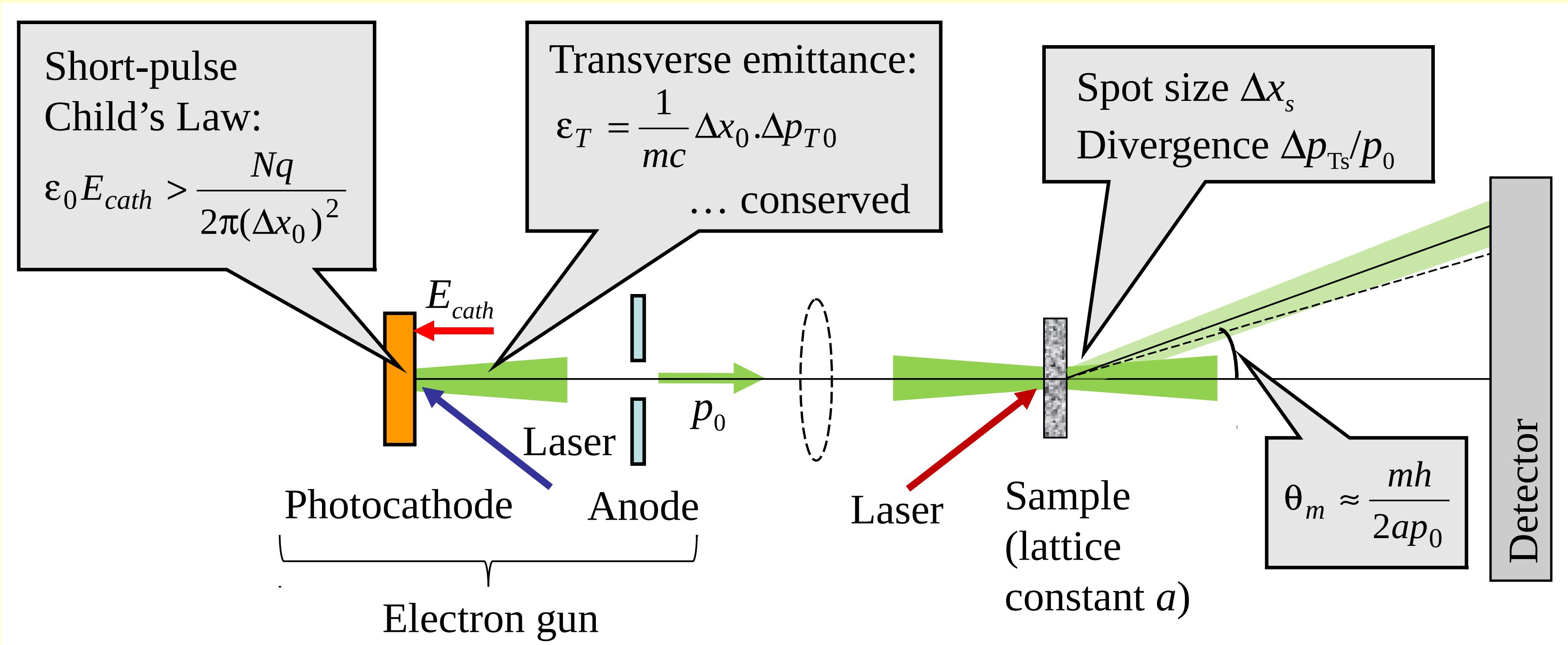
− Non-relativistic regime

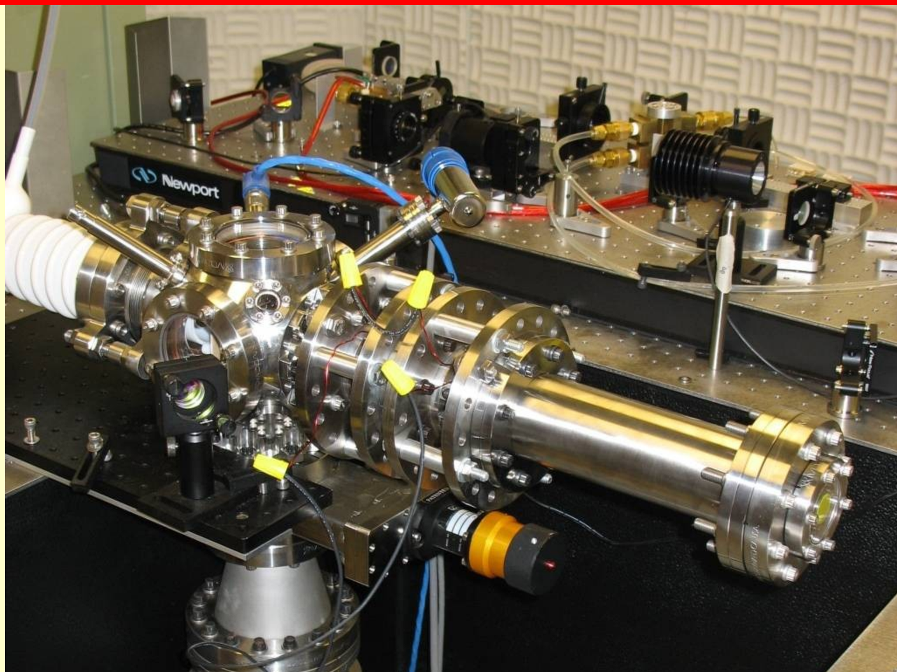



$$\Rightarrow \text{Observable} \quad \frac{\delta a}{a} \geq \frac{\xi}{mh}\left(\frac{a}{\Delta x_s}\right)\sqrt{\frac{2Nq}{\pi\varepsilon_0 E_{cath}}} . \Delta p_{T0} \quad ; \quad \text{with } \Delta p_{T0} = ??$$

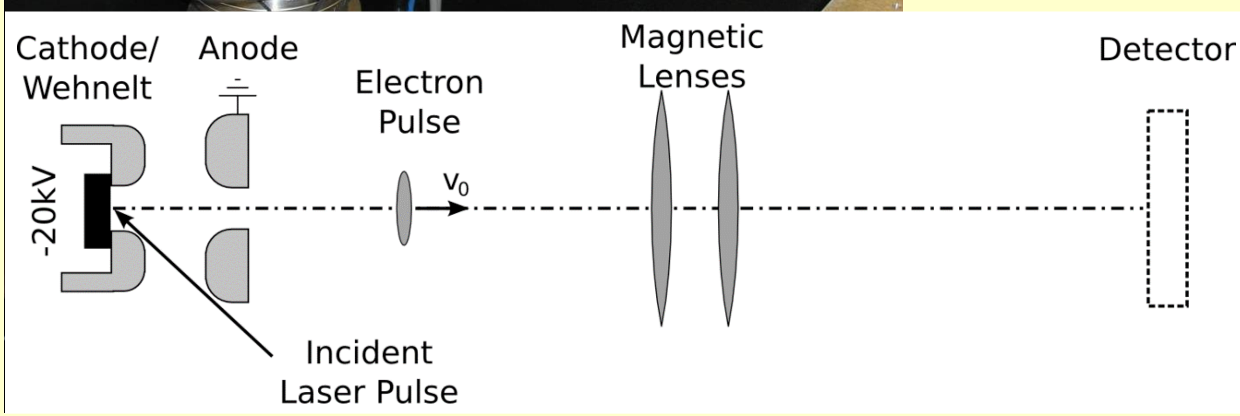
D.H. Dowell & J.F. Schmerge, *Phys. Rev. ST – Acc. & Beams* **12** (2009) 074201
K.L. Jensen et al., *J. Appl. Phys.* **107** (2010) 014903

# $\Delta p_{T0}$ Measurement: Solenoid Scan






- 2W, 250fs, 63MHz , diode-pumped Yb:KGW laser
  - ~4ps at 261nm ($\hbar\omega$ = 4.75eV)
- YAG scintillator optically coupled to CCD camera
  - Beam size vs. magnetic coil (lens) current measured
  - Analytical Gaussian (AG) pulse propagation model to extract $\Delta p_{T0}$




J.A. Berger & W.A. Schroeder, *J. Appl. Phys.* **108** (2010) 124905

# Results: Polycrystalline Cr

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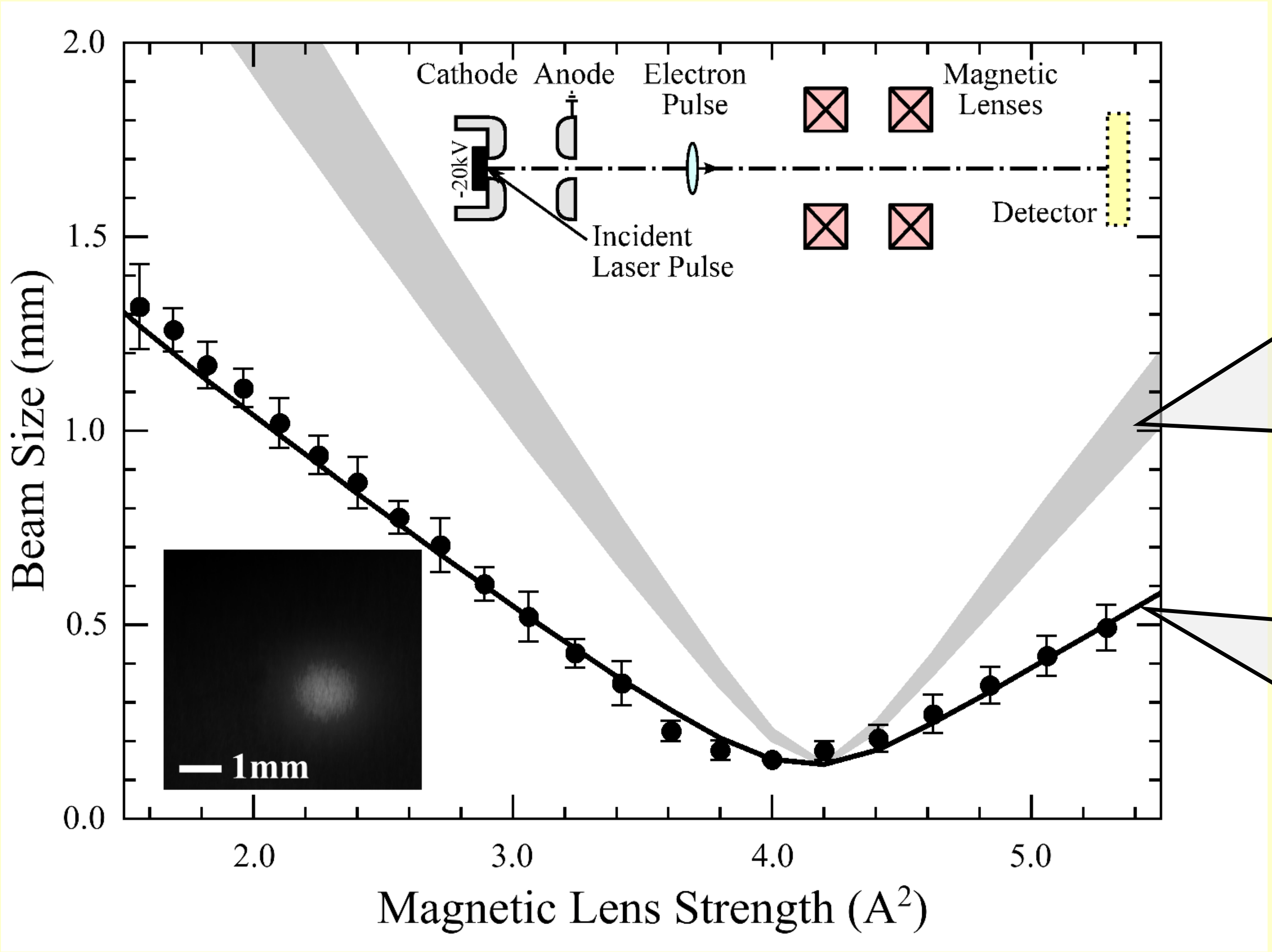
− Solenoid scan measurement




Range for $\Delta p_T$ from

$$\Delta p_{T0} = \sqrt{\frac{m_0(\hbar\omega - \phi)}{3}}$$

… $\phi = 4.50(\pm 0.05)$eV and $\hbar\omega = 4.75$eV

AG model simulation of experiment gives

$\Delta p_{T0} = 0.155(\pm 0.01)$ $(m_0.eV)^{1/2}$


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

# Results: 10 Polycrystalline Metals

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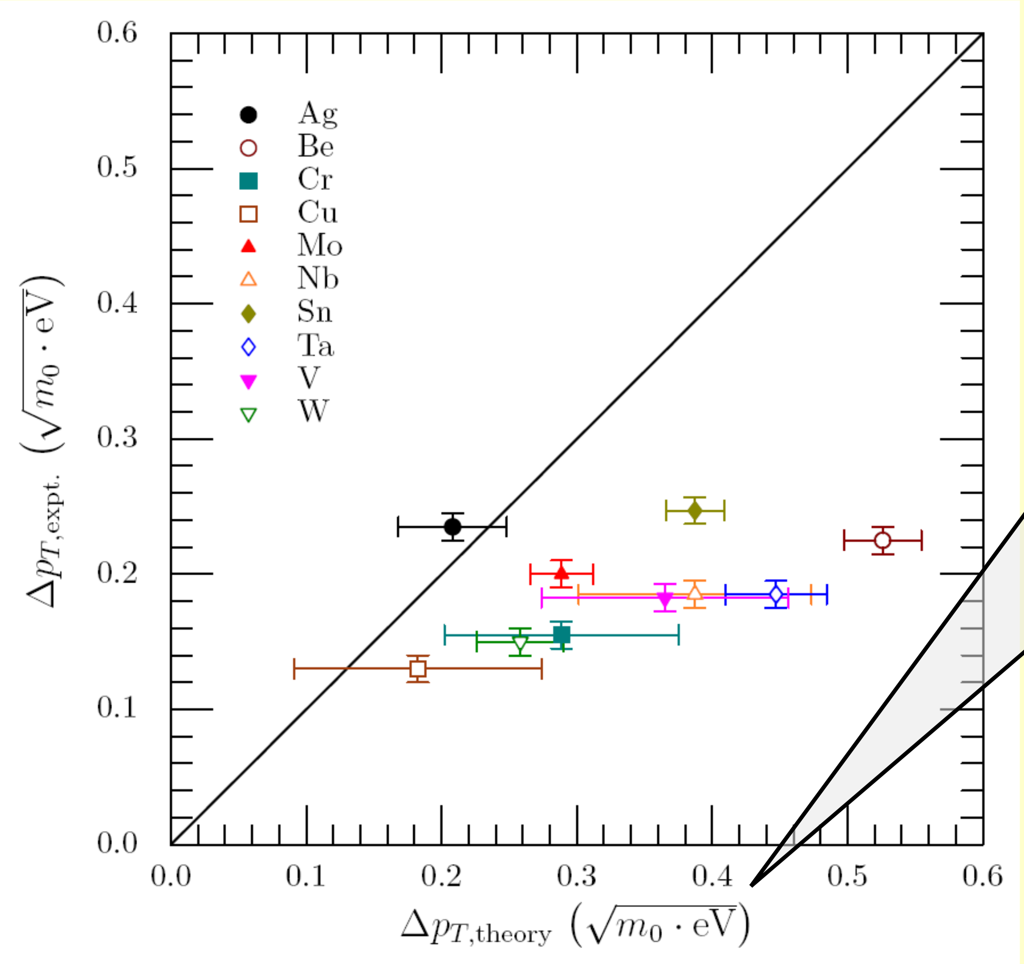
– $\Delta p_T$ from solenoid scan measurements vs. standard theory




Standard expression,

$$\Delta p_{T0} = \sqrt{\frac{m_0(\hbar\omega - \phi)}{3}} \;,$$

assumes:

(i) Isotropic parabolic electron-like bands;

(ii) Constant density of states;

(iii) $m^* = m_0$, the free electron mass.

Disagreement ⇒ Band structure effect ?

B.L. Rickman et al., *Phys. Rev. Lett.* **111** (2013) 237401

# Photoemission Theory I

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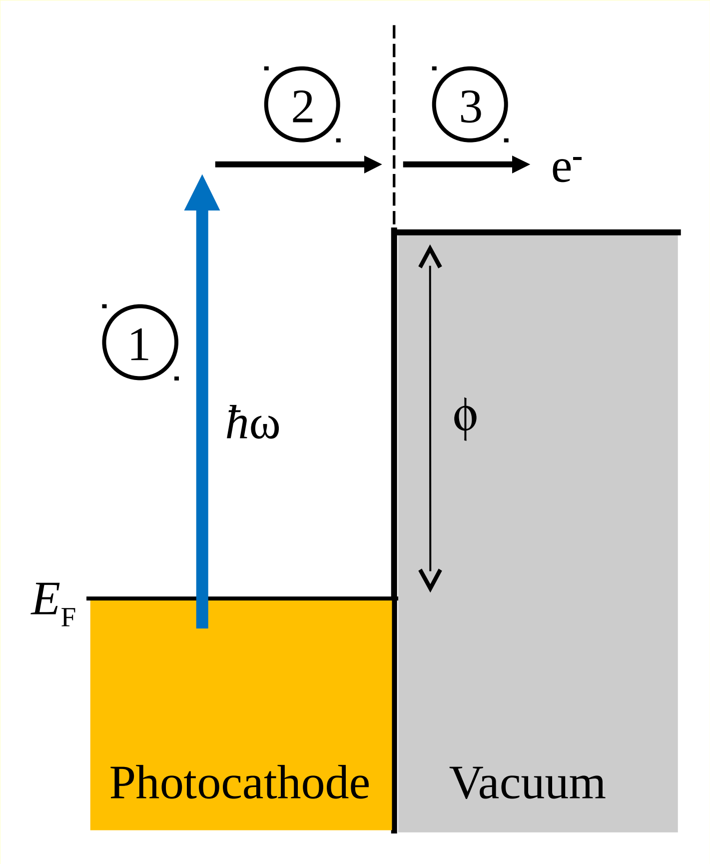
− The semi-classical three-step 'Spicer' model




1. Photoexcitation
2. Transport to surface
3. Emission from surface

- Transport ⇒ ***Real*** electronic band
  ∴ Photoexcitation into upper state near vacuum level

- Emission from upper excited state
  ⇒ High quantum efficiency ($\eta_{PE}$)
  **AND**
  Response time ≈ Lifetime (ps-ns)

∴ **NOT** suitable for UED

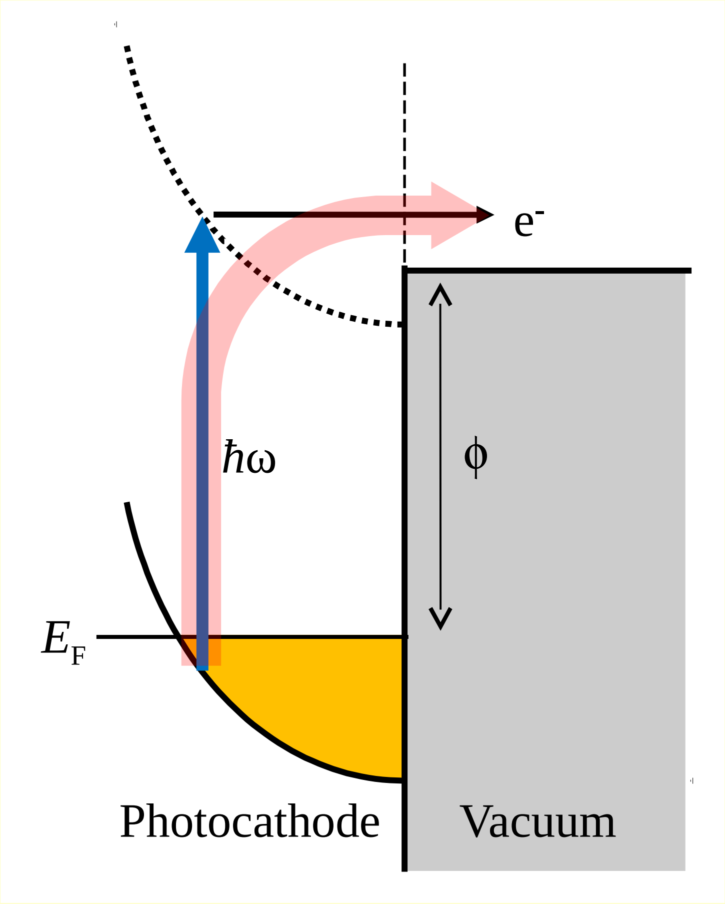
Examples: NEA GaAs, KCsSb, GaSb, diamond, Cu(111)?


C.N. Berglund & W.E. Spicer, *Phys. Rev.* **136**, A1030-A1044 (1964)
P.J. Feibelman & D.E. Eastman, *Phys. Rev. B* **10**, 4932-4947 (1974)

# Photoemission Theory II

UIC

− The 'quantum mechanical' one-step model




Photoexcitation into a ***virtual*** state (excited copy of filled band) emitting into the vacuum in one step

- Low $\eta_{PE}$ ~ $10^{-5}$ to $10^{-7}$
- 'Instantaneous' emission process

Suitable for UED

Examples: Most metals

G.D. Mahan, *Phys. Rev. B* **2**, 4334-4350 (1970)

# Band Structure Effects

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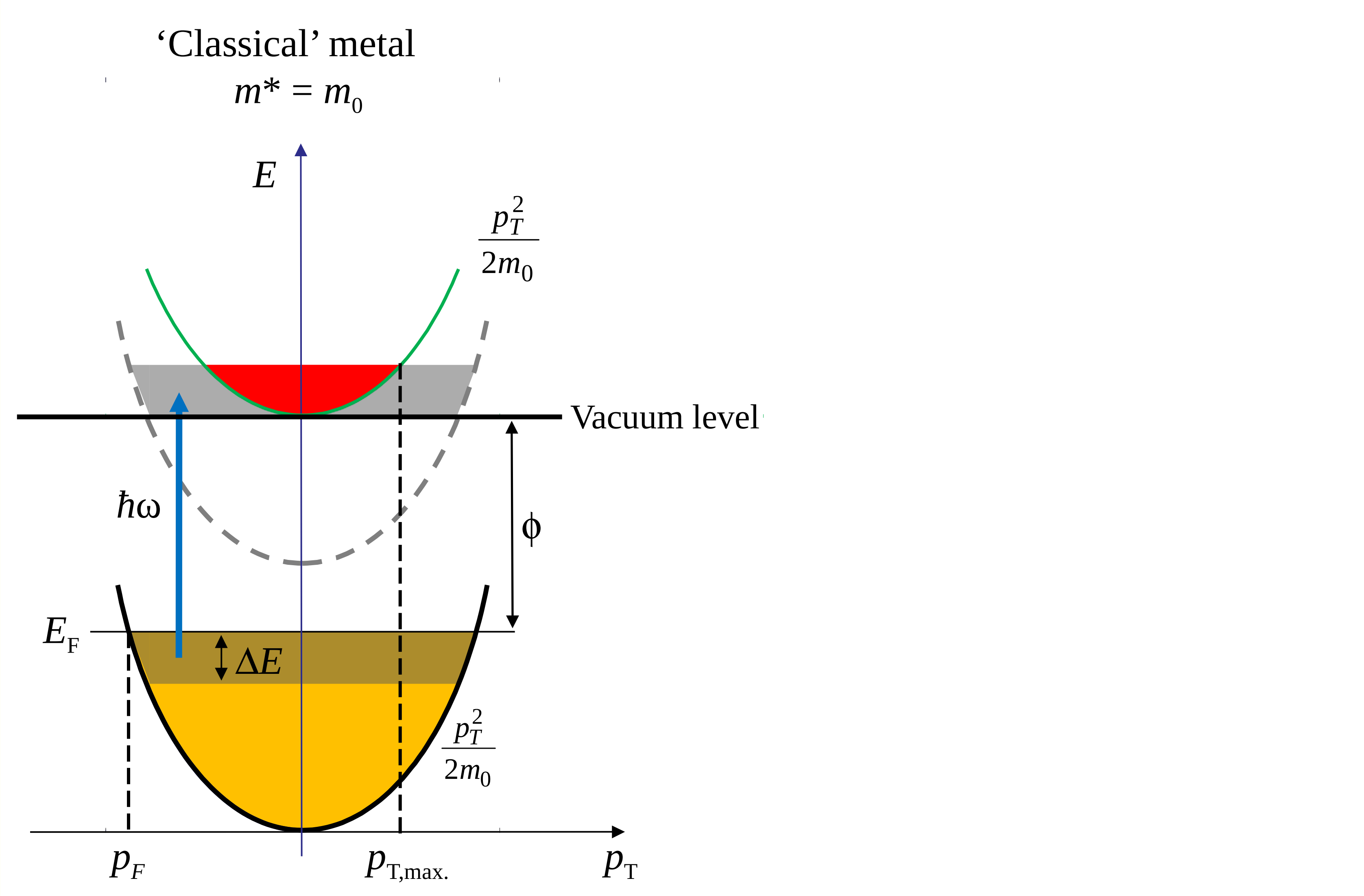
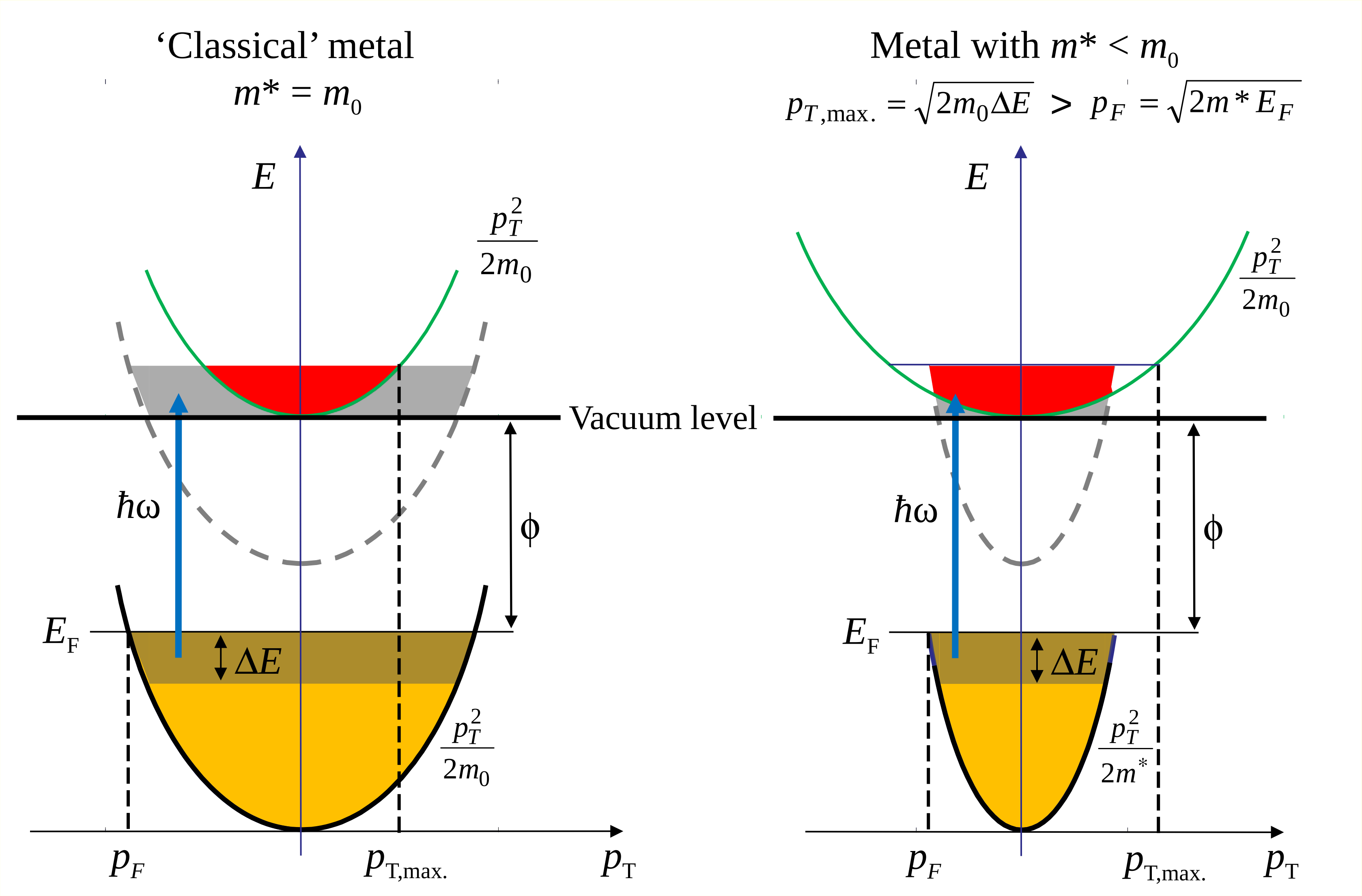
– Transverse momentum $p_T$ conserved in photoemission



# Band Structure Effects

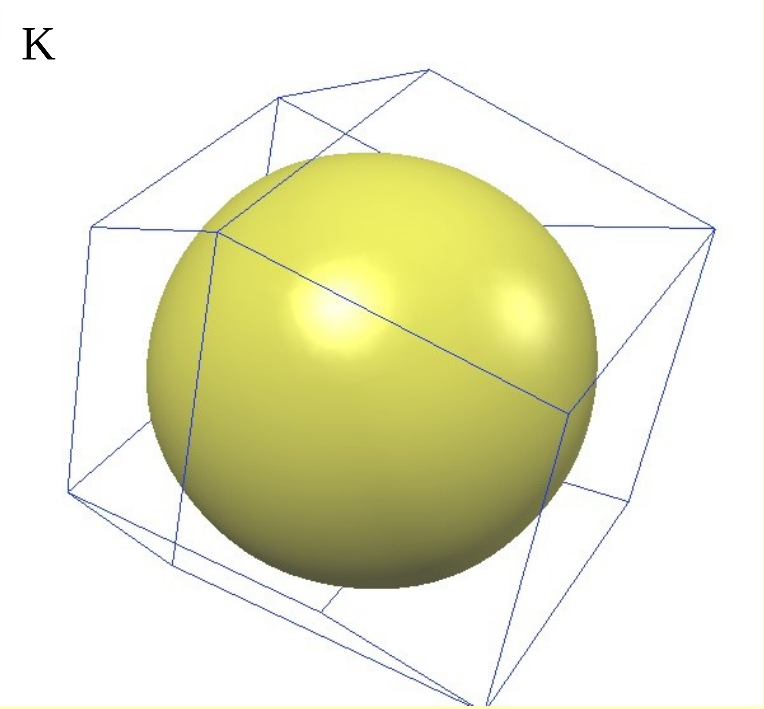
UIC

– Transverse momentum $p_T$ conserved in photoemission



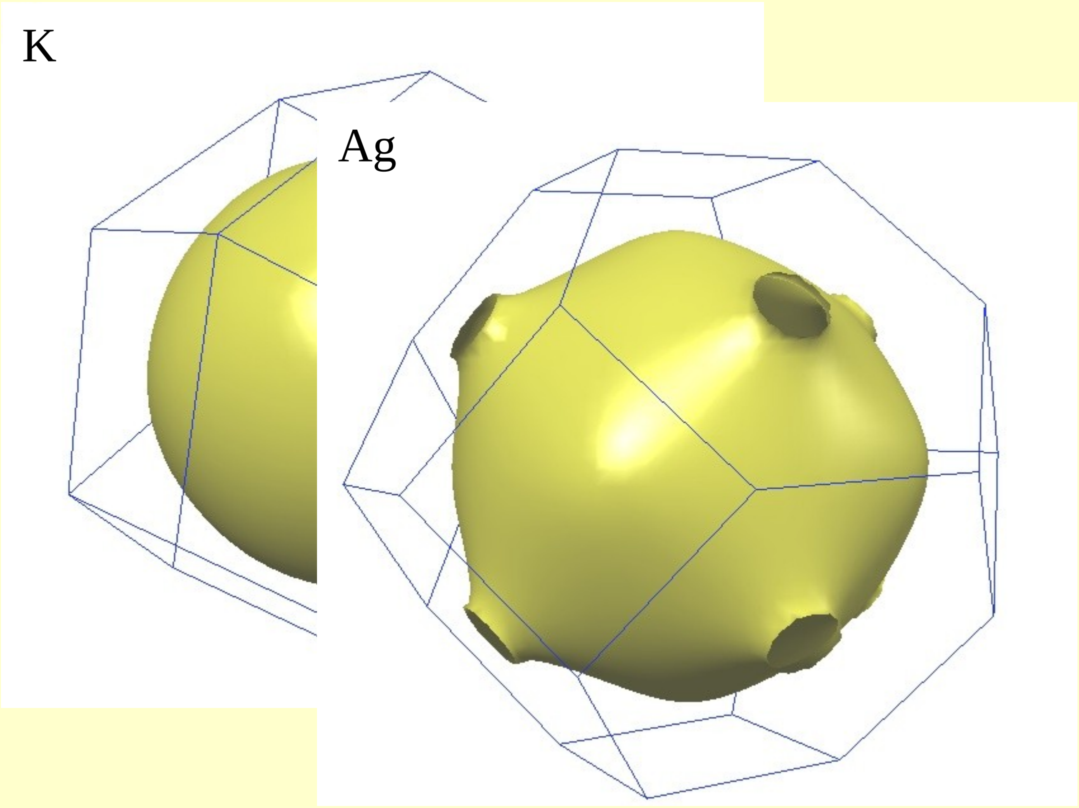
# Fermi Surfaces

UIC



# Fermi Surfaces

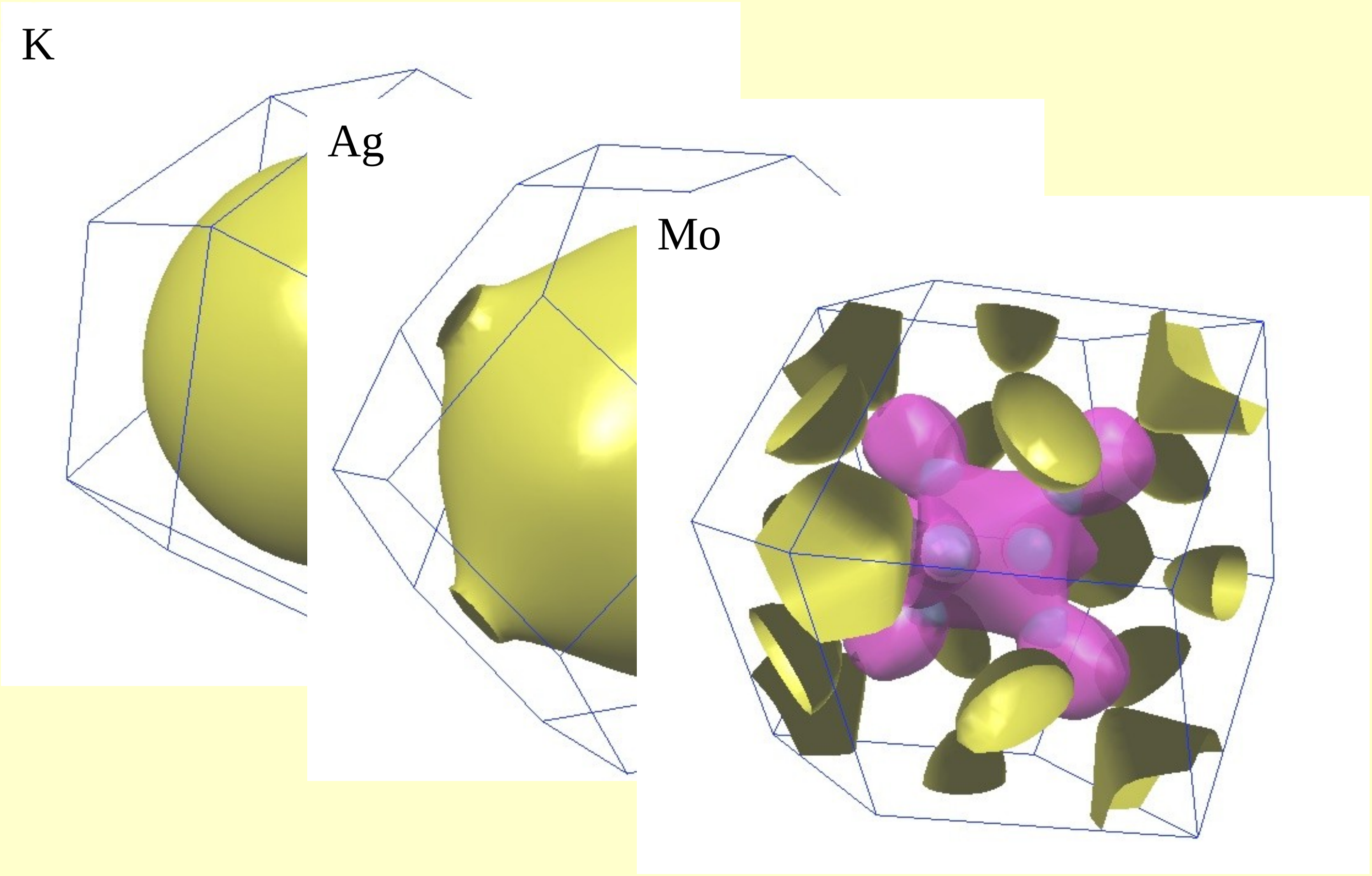





www.phys.ufl.edu/fermisurface/

# Fermi Surfaces

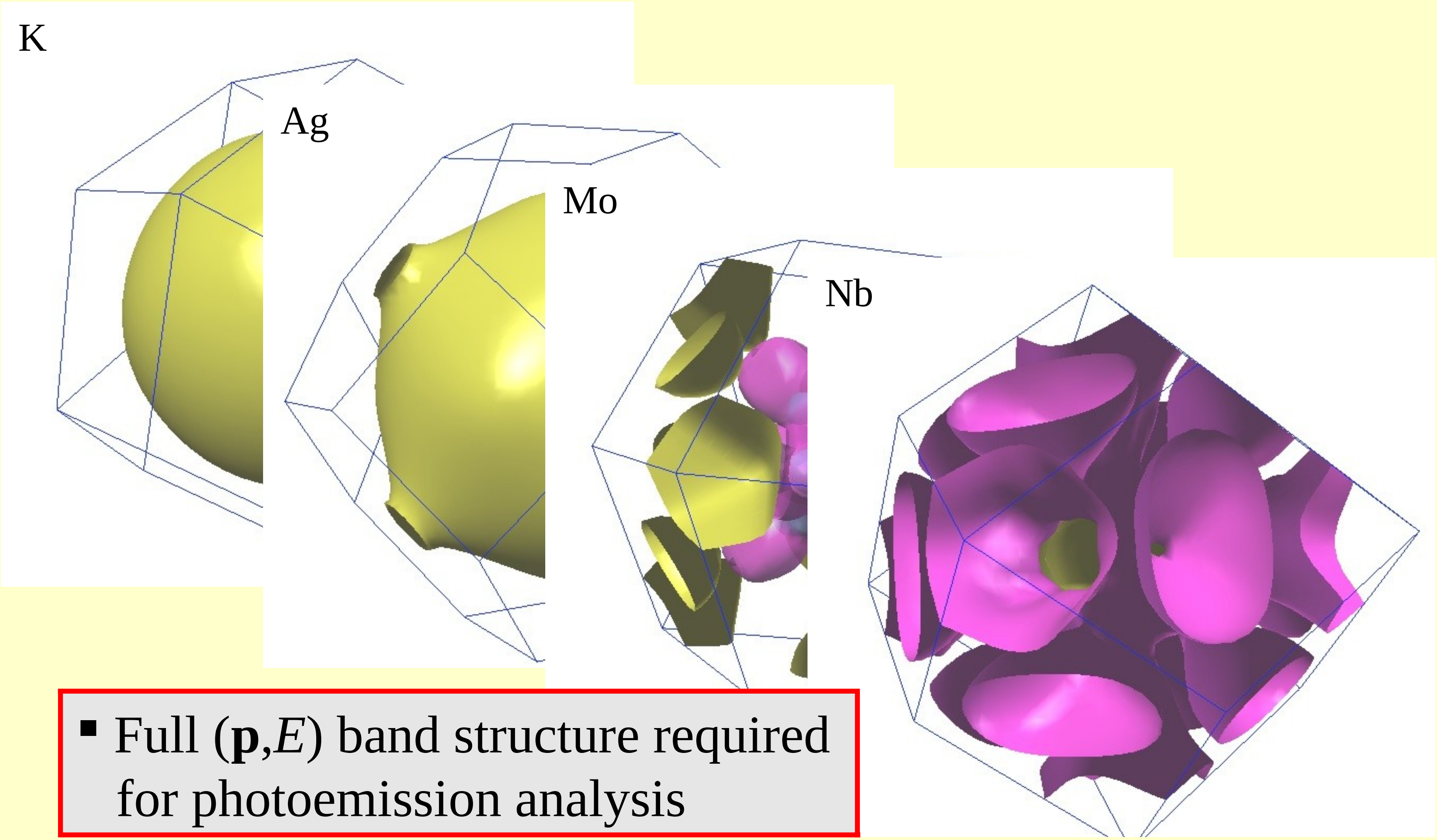
UIC




www.phys.ufl.edu/fermisurface/

# Fermi Surfaces

UIC




- Full (**p**,*E*) band structure required for photoemission analysis

www.phys.ufl.edu/fermisurface/

# Work Function Anisotropy

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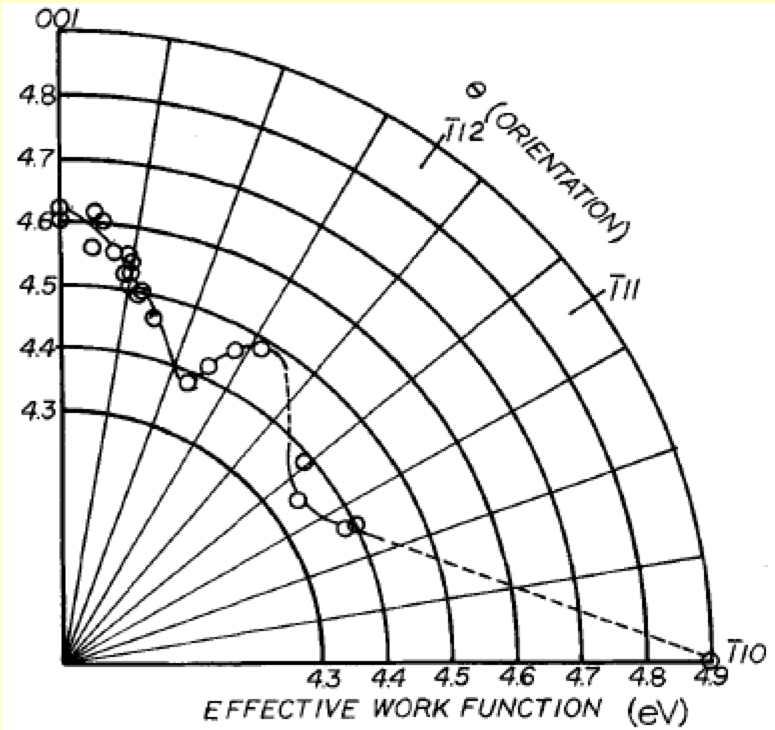
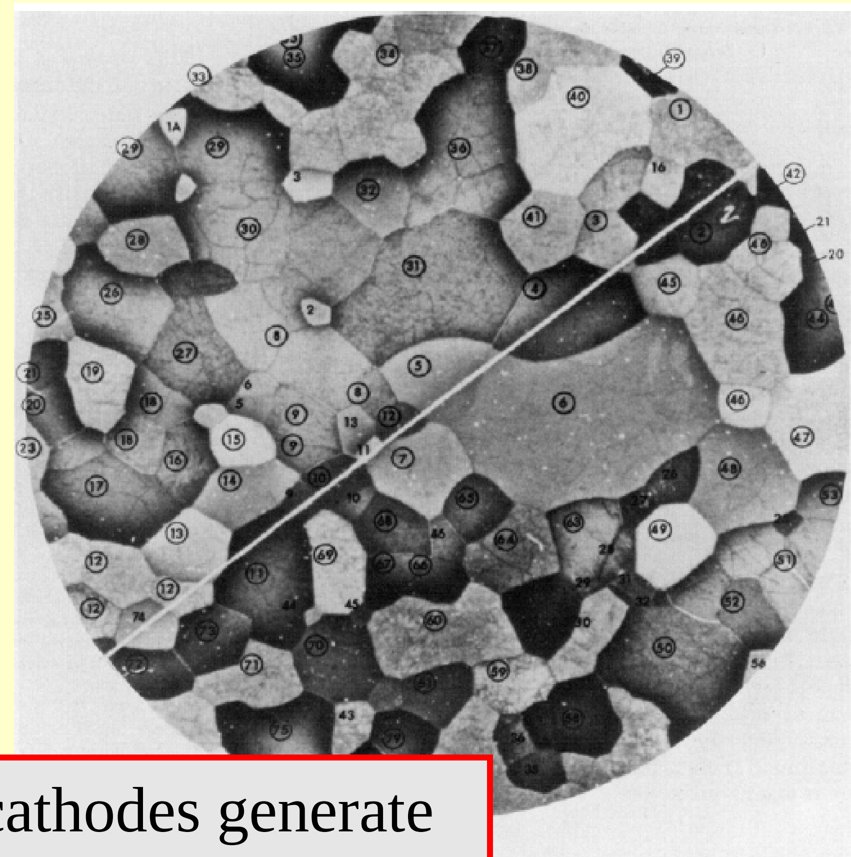
– Example: $\phi_{(ijk)}$ for Mo by electron emission microscopy



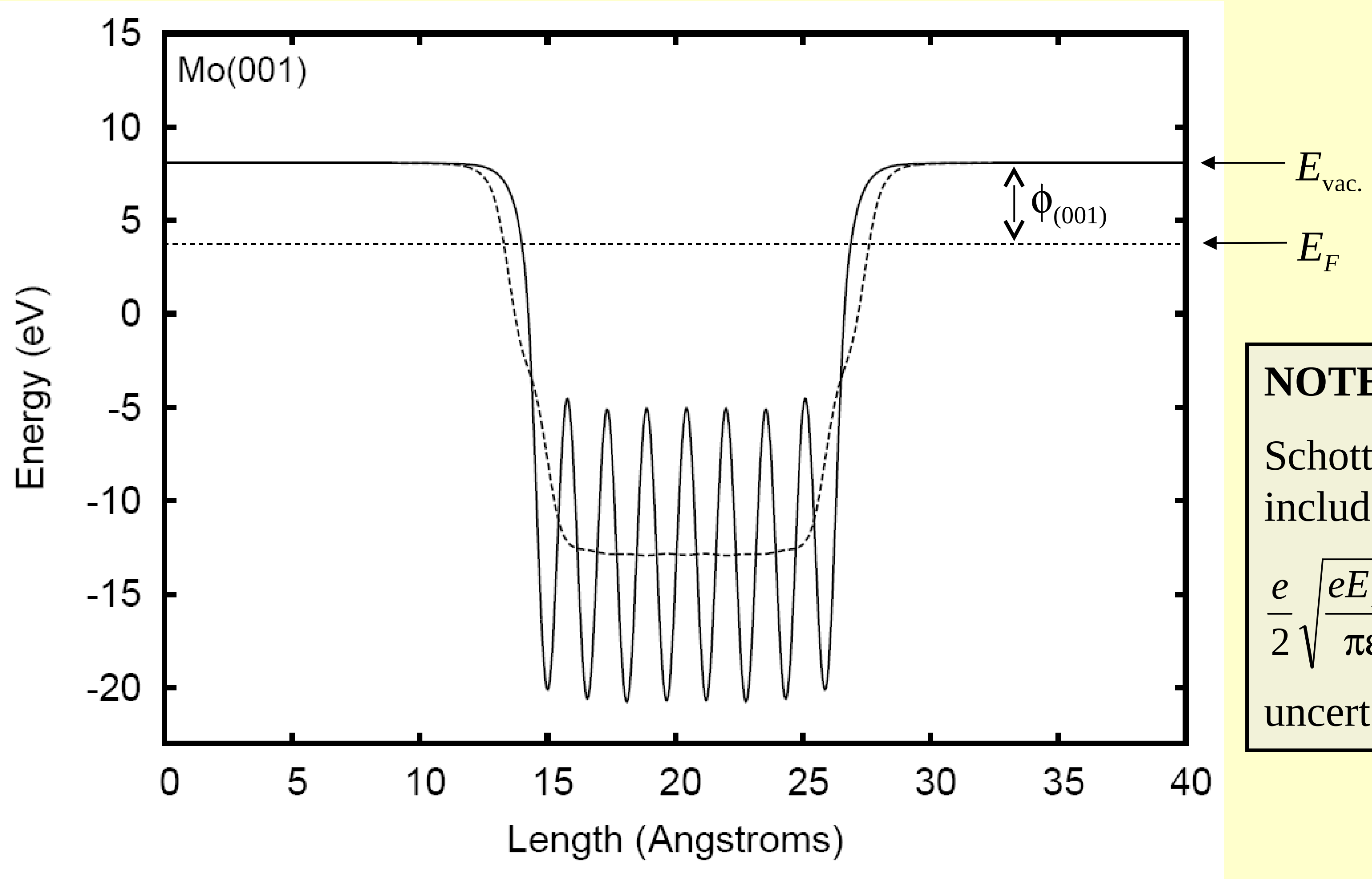
- Polycrystalline metal photocathodes generate *inhomogeneous* electron beams
- Any photoemission analysis ***must*** include $\phi_{(ijk)}$

D. Jacobson & A. Campbell, *Metall. Trans.* **2**, 3063-3066 (1971)

# Thin-slab Evaluation of $\phi_{(ijk)}$

UIC

− Example: $\phi_{(001)} = 4.53(\pm 0.05)$eV for Mo




**NOTE**

Schottky effect *not* included:

$$\frac{e}{2}\sqrt{\frac{eE_{DC}}{\pi\varepsilon_0}} \approx \pm 50\text{meV}$$

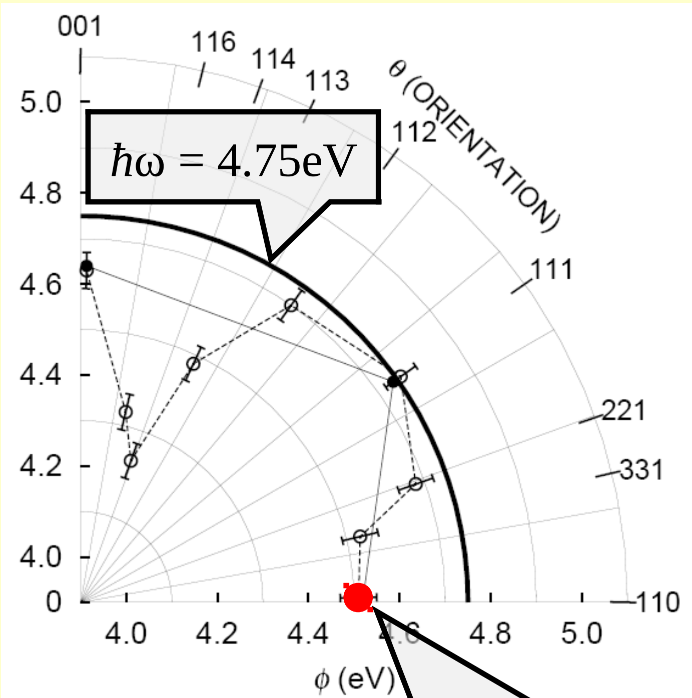
uncertainty in $\phi_{(ijk)}$

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

# Photoemission Simulation: Ag

UIC

− fcc crystal lattice




Lowest index face
with lowest $\phi_{(ijk)}$

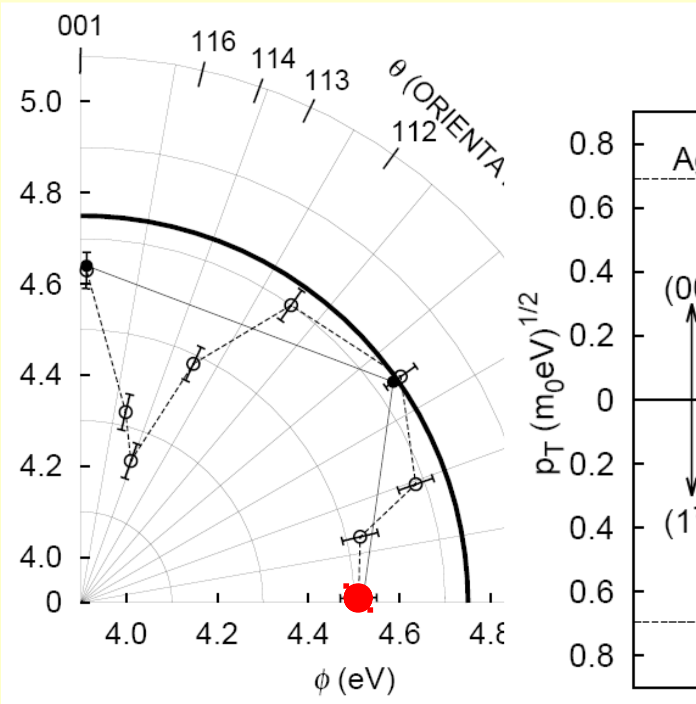
$\Delta E = \hbar\omega - \phi_{(110)} = 0.23\text{eV}$

… ±50meV error in $\phi_{(ijk)}$

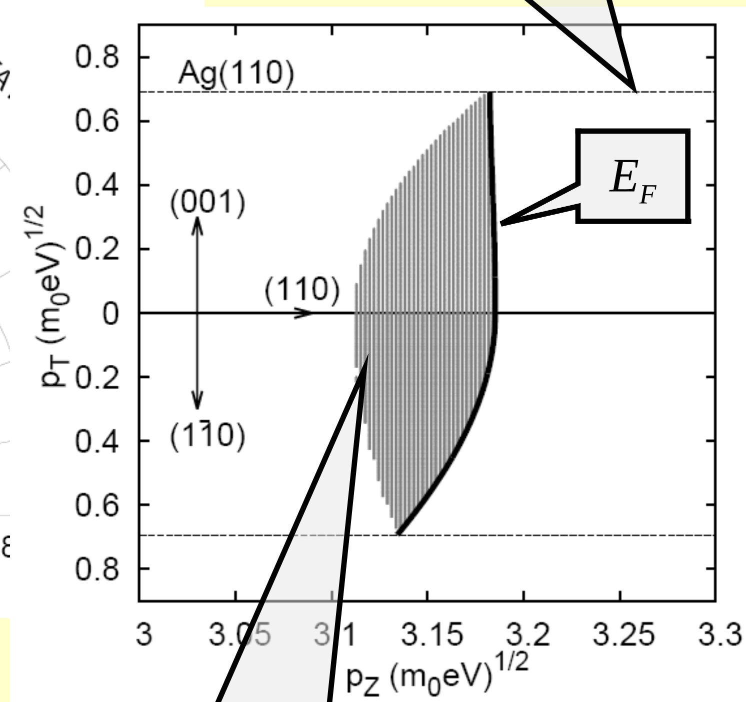
# Photoemission Simulation: Ag

UIC

– fcc crystal lattice




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



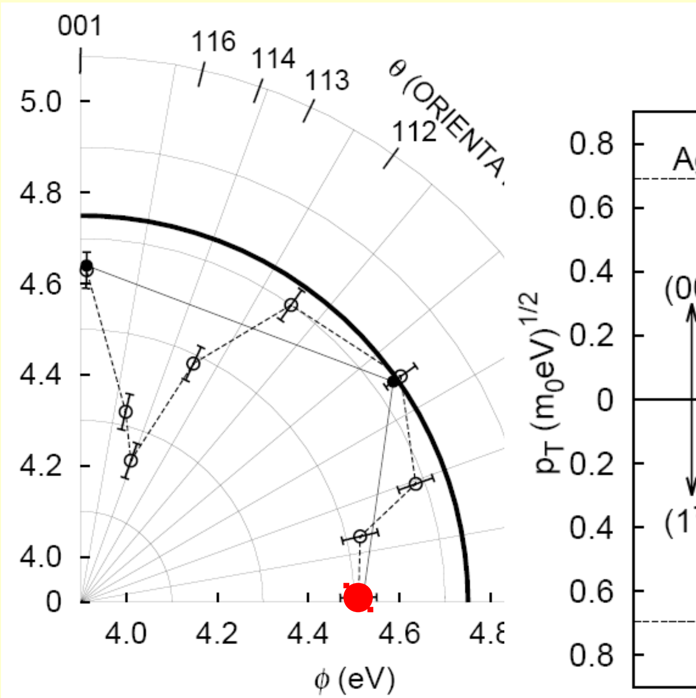

$E_F$

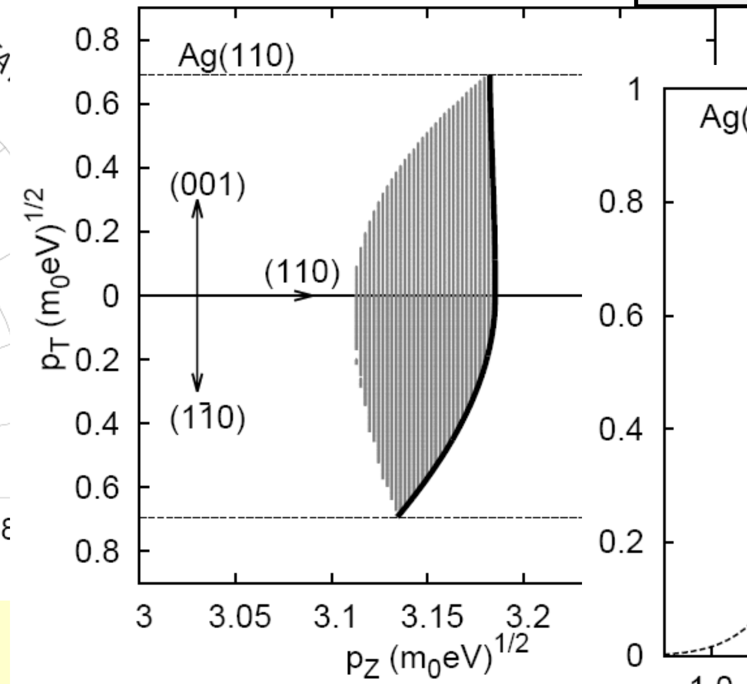
Photoemitting electron-like states for $\hbar\omega = 4.75\text{eV}$ ($\mathbf{p}_T$ and $E$ conserved)

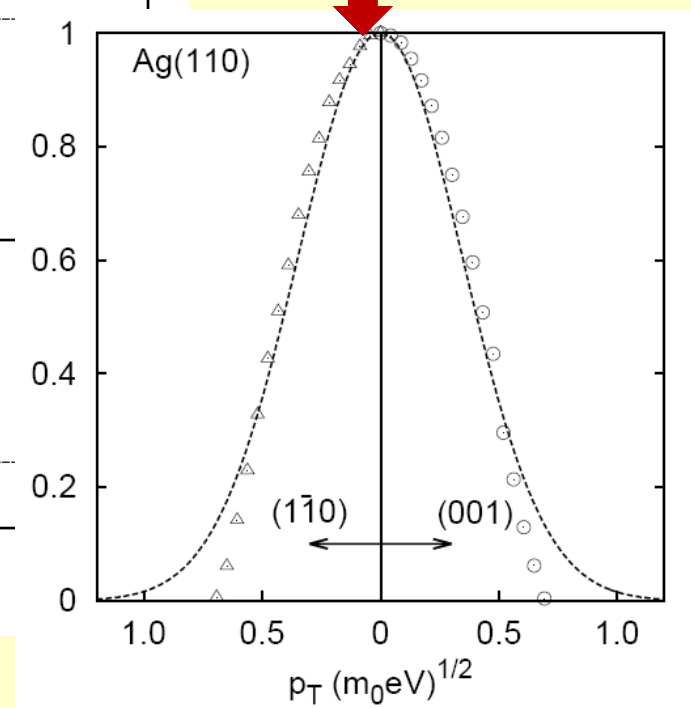
# Photoemission Simulation: Ag

UIC

− fcc crystal lattice




$E$, $\mathbf{p}_T$ conservation

**PLUS**

Barrier transmission, $T(p_z,p_{z0})$

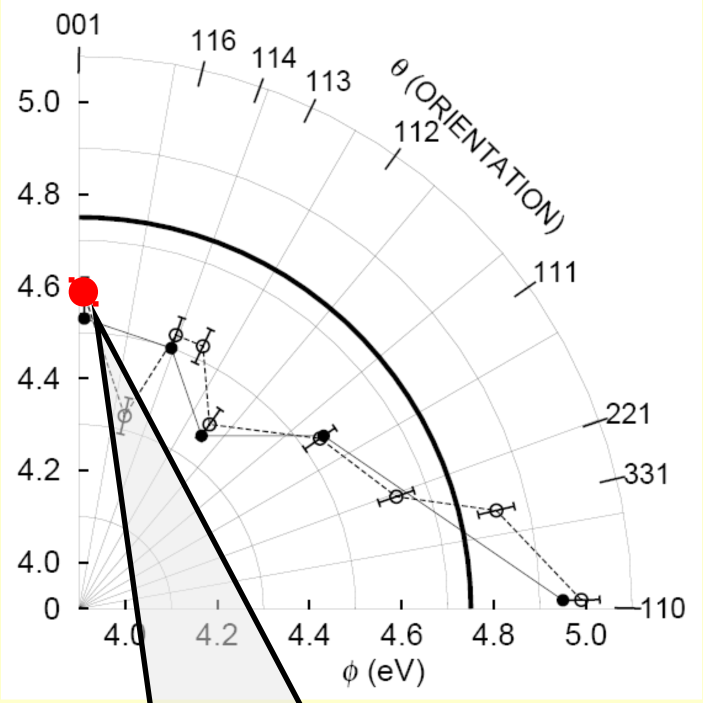
Spatially-averaged

$\Delta p_{T0}$ = 0.267 $(m_0.eV)^{1/2}$

# Photoemission Simulation: Mo

UIC

− bcc crystal lattice




Lowest index face with lowest $\phi_{(ijk)}$

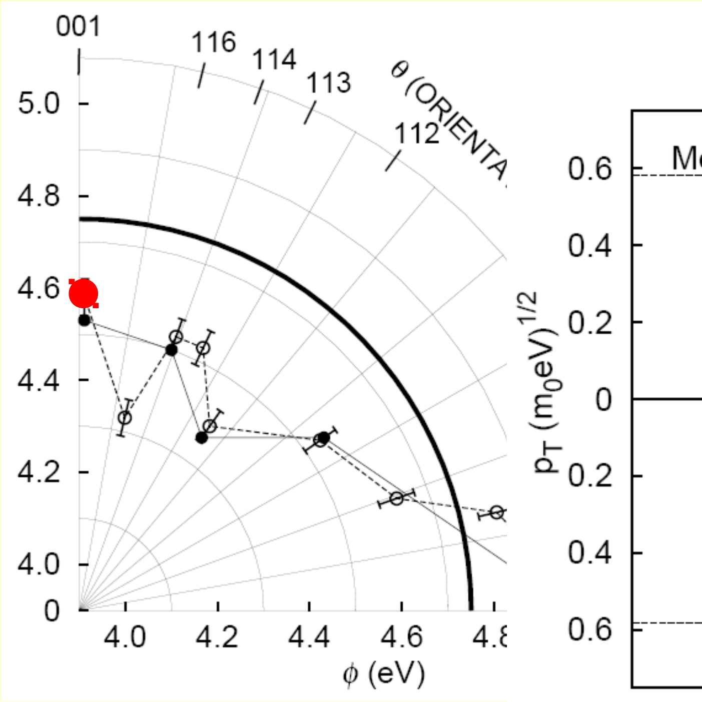
$\Delta E = \hbar\omega - \phi_{(001)} = 0.22\text{eV}$

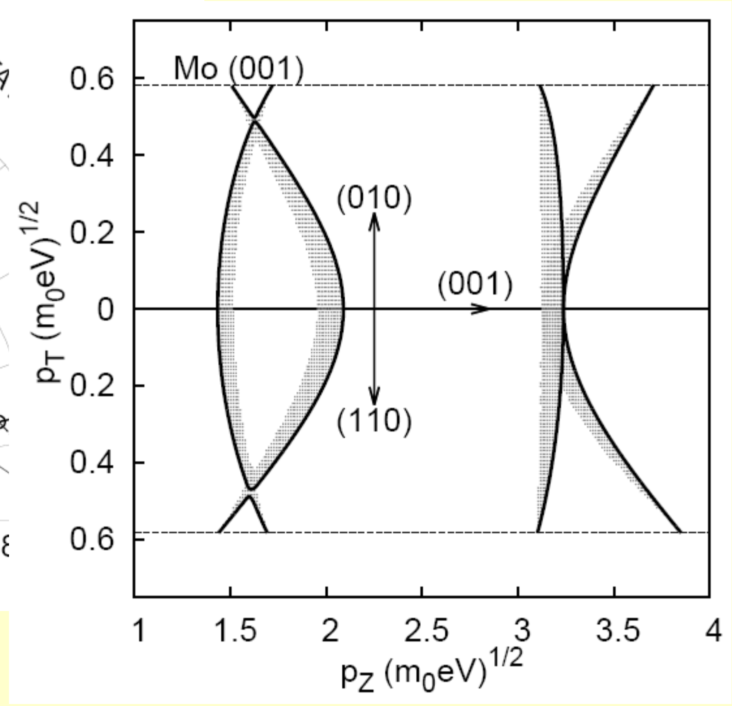
… ±50meV error in $\phi_{(ijk)}$

# Photoemission Simulation: Mo

UIC

− bcc crystal lattice




Both electron- and hole-like states contribute to photoemission

# Photoemission Simulation: Mo

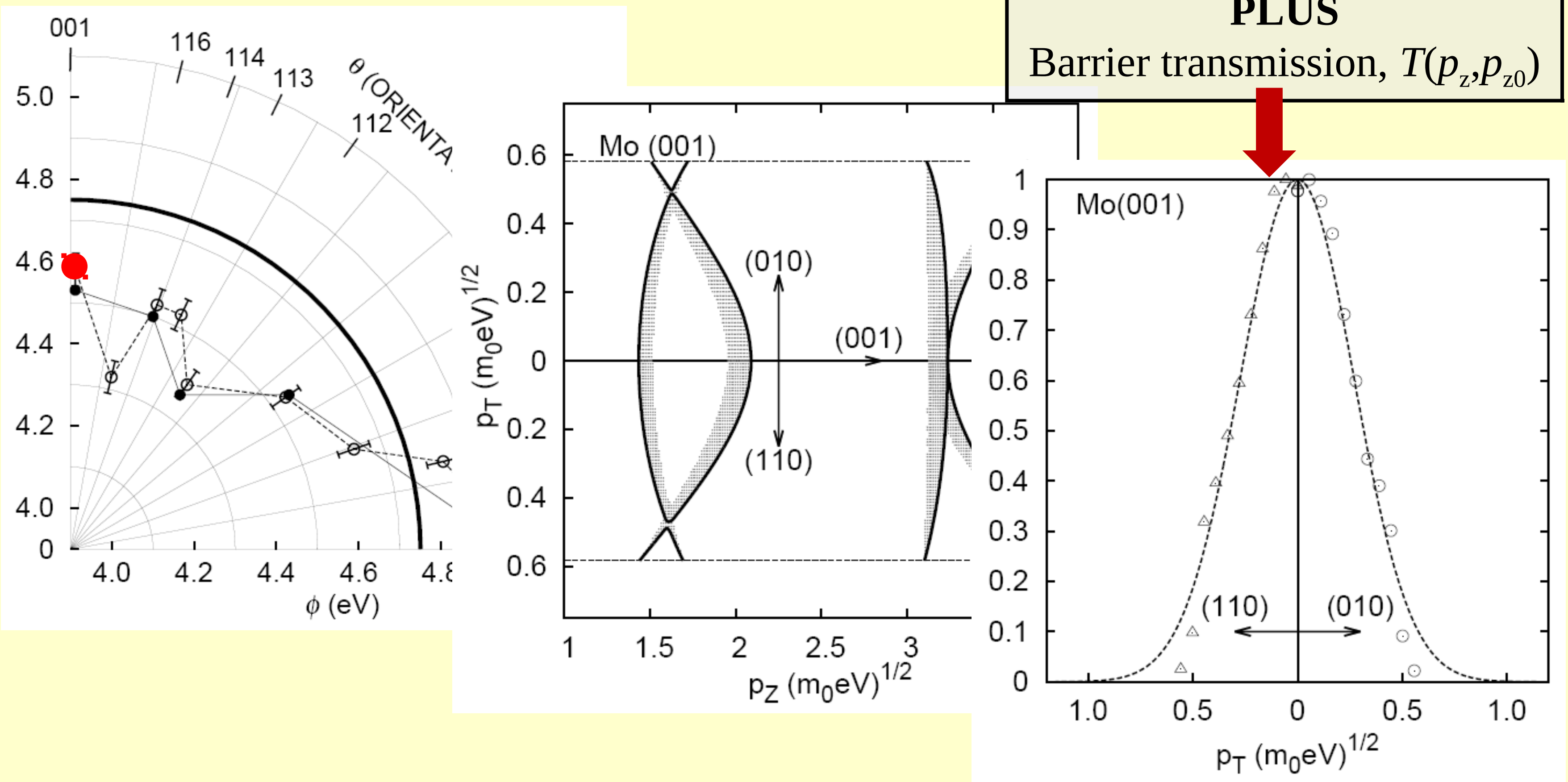
UIC

− bcc crystal lattice

$E$, $\mathbf{p}_T$ conservation

**PLUS**

Barrier transmission, $T(p_z, p_{z0})$




Spatially-averaged

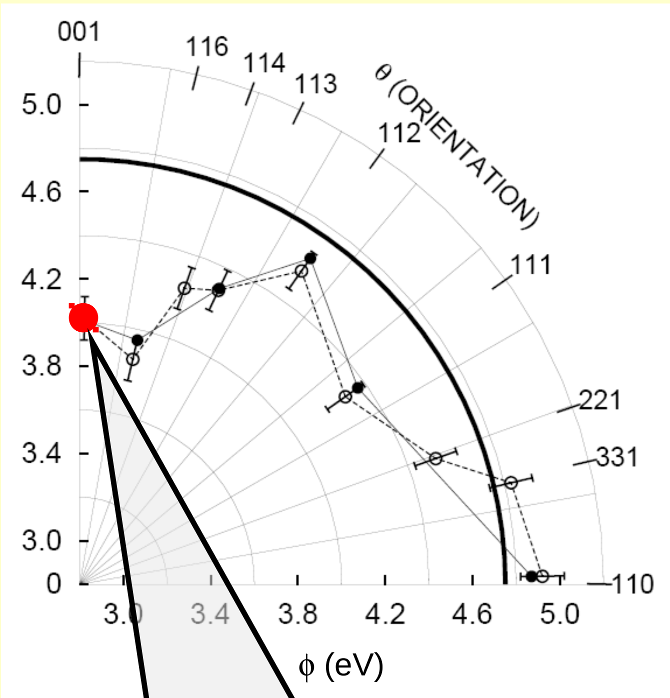
$\Delta p_{T0} = 0.219\ (m_0.eV)^{1/2}$

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

# Photoemission Simulation: Nb

UIC

− bcc crystal lattice




Lowest index face
with lowest $\phi_{(ijk)}$

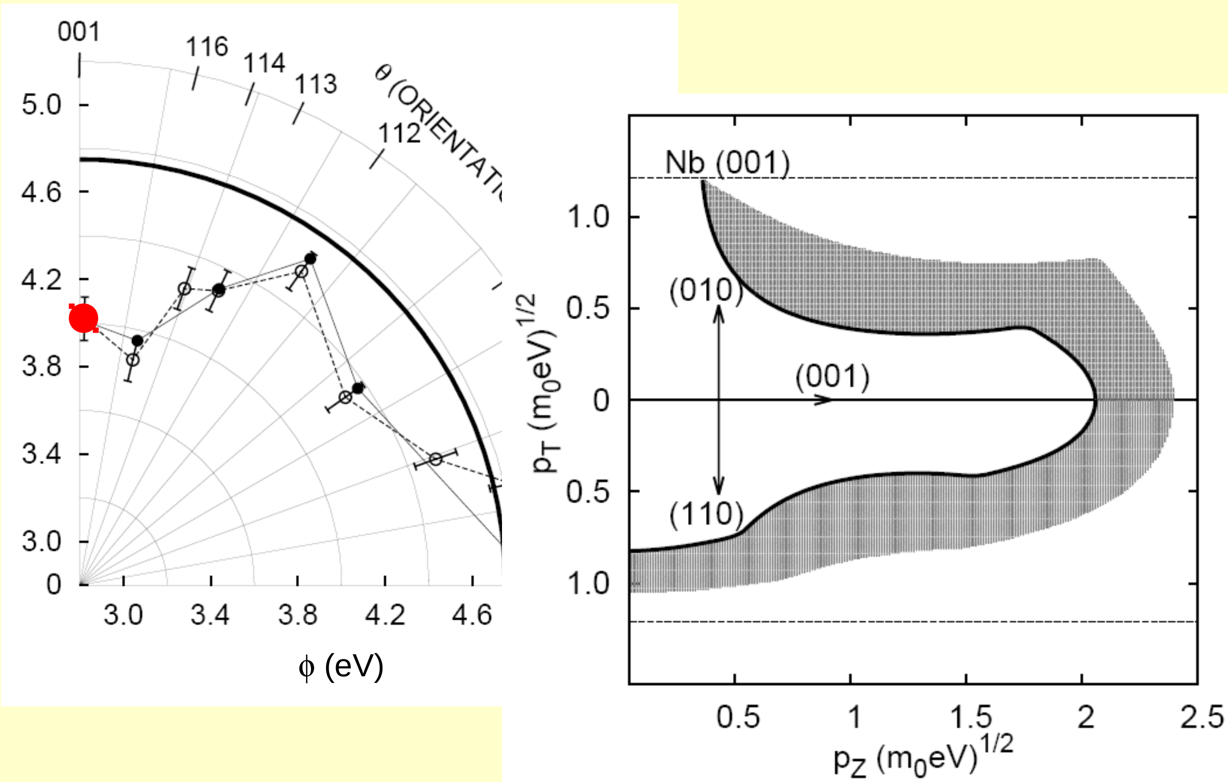
$\Delta E = \hbar\omega - \phi_{(001)} = 0.73\text{eV}$

… ±50meV error in $\phi_{(ijk)}$

# Photoemission Simulation: Nb

UIC

− bcc crystal lattice

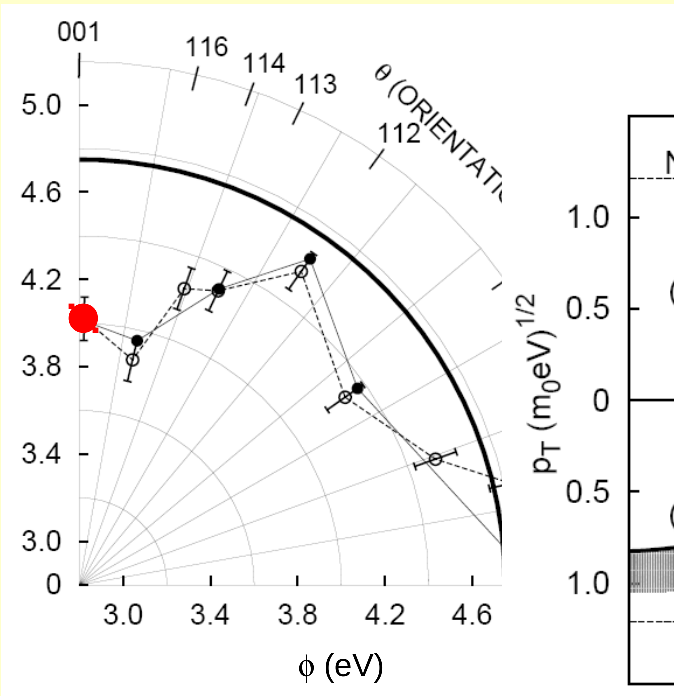


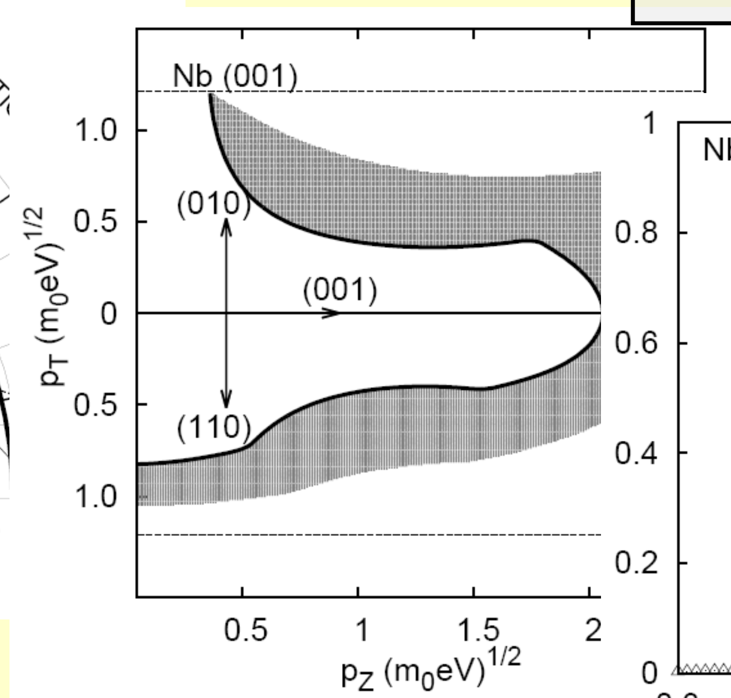

Only hole-like states contribute to photoemission

# Photoemission Simulation: Nb

UIC

− bcc crystal lattice

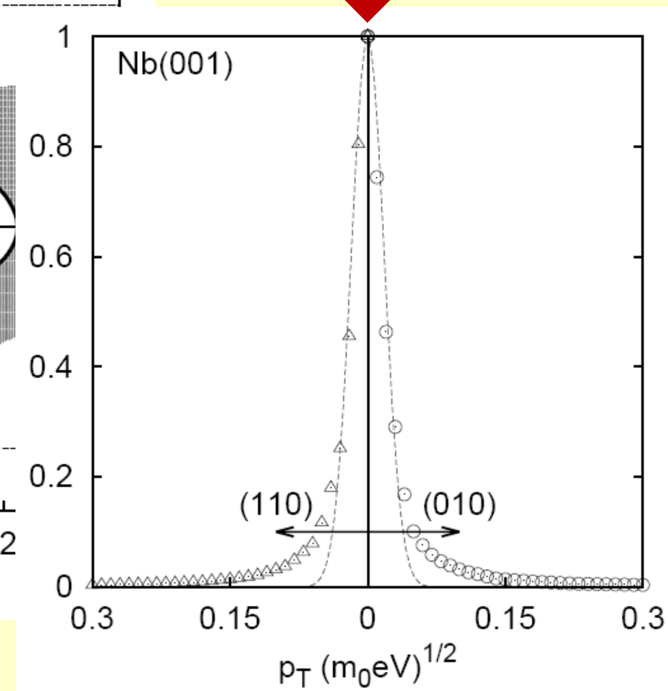



$E$, $\mathbf{p}_T$ conservation

**PLUS**

Barrier transmission, $T(p_z,p_{z0})$




Spatially-averaged

$\Delta p_{T0} = 0.196\ (m_0.eV)^{1/2}$

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

# Experiment vs. DFT Analysis

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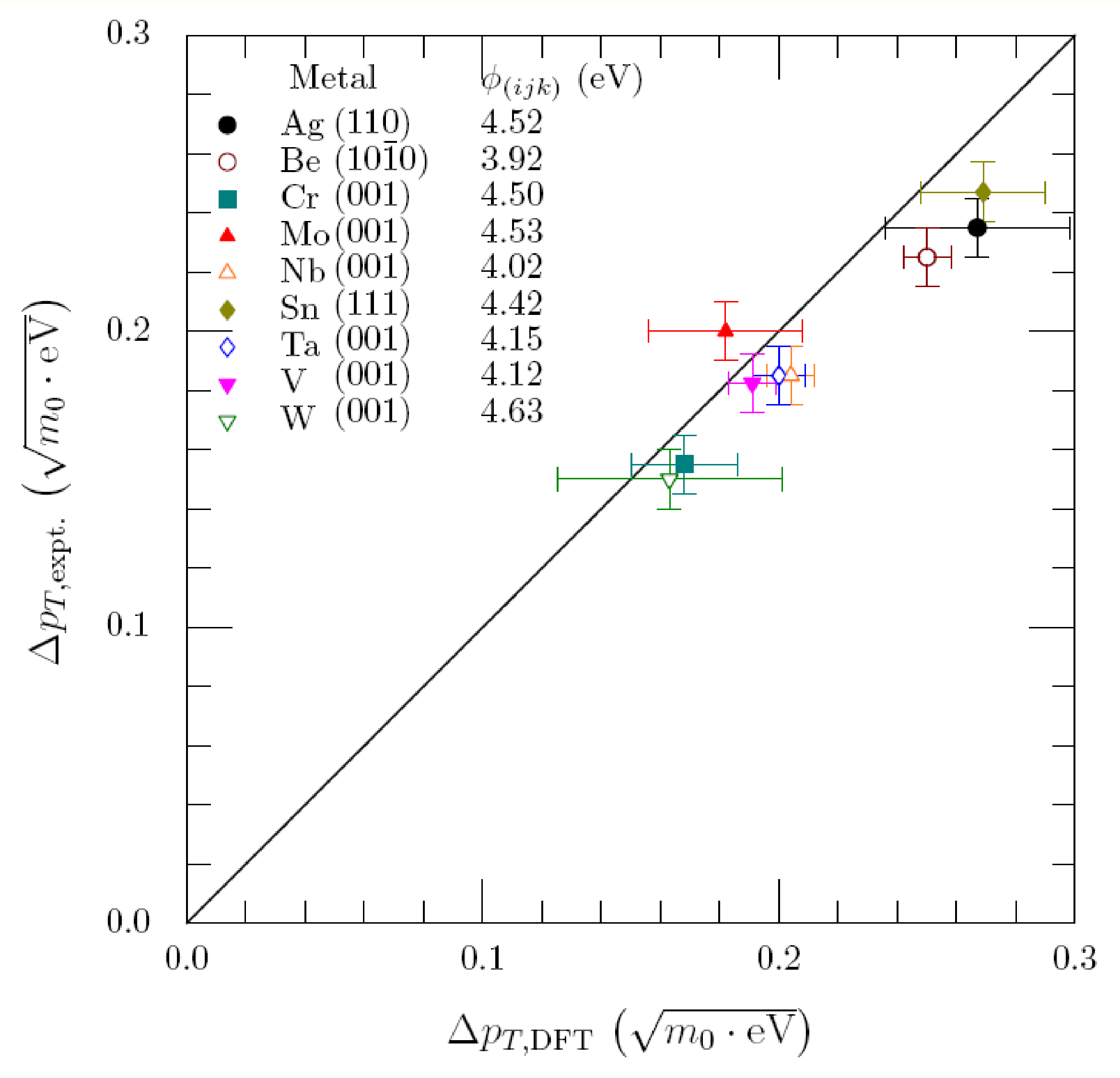



**NOTE:**

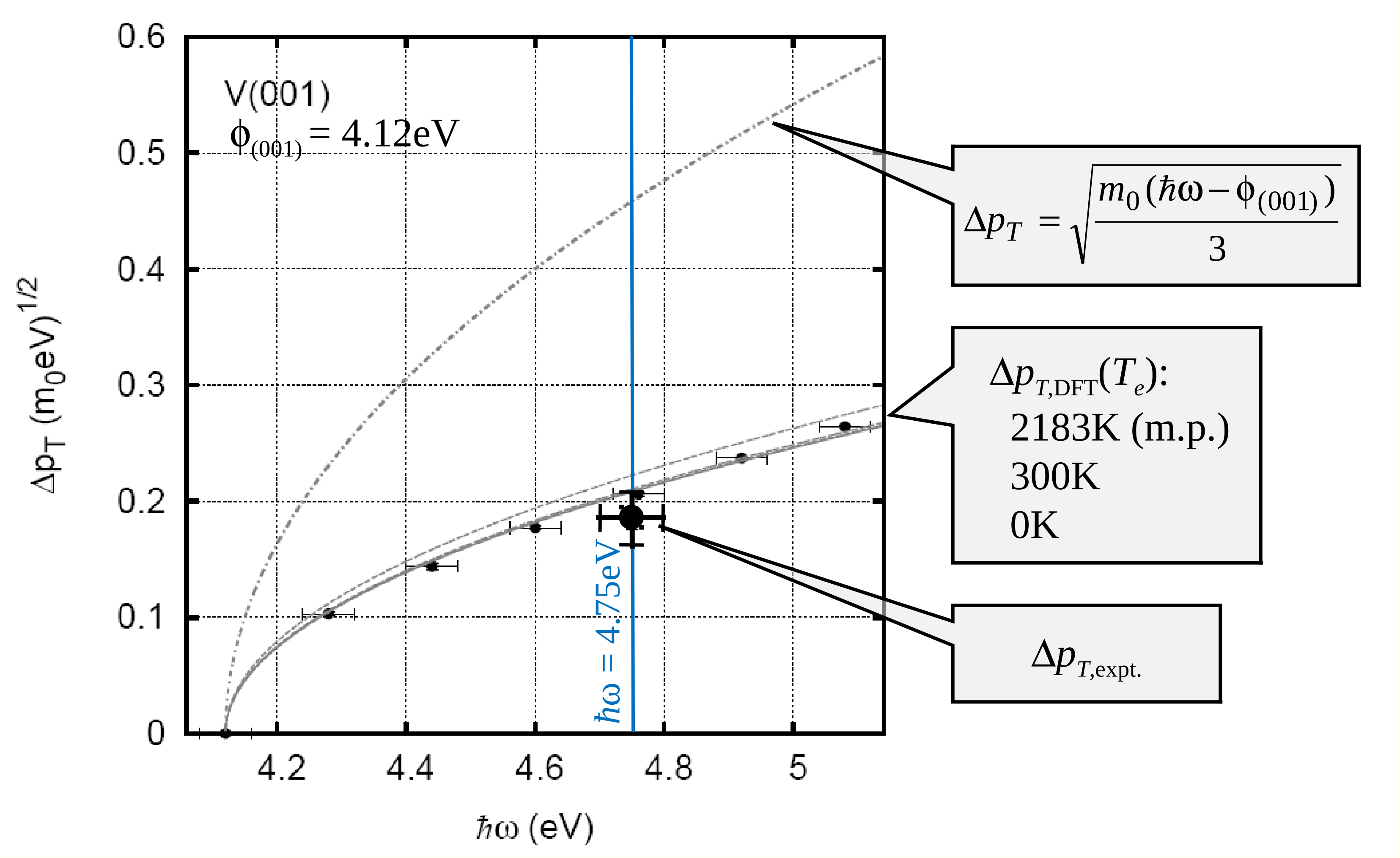
- Polycrystalline vs. single-crystal comparison
  - Other crystal faces with smaller $\Delta E = \hbar\omega - \phi_{(ijk)}$

    contribute lower $\Delta p_{T0}$
- DFT analysis at $T_e \rightarrow 0$K
  Experiment at 300K
- Cu is missing …
- Be is HCP
  $\Rightarrow$ Anisotropic $\Delta p_{T0}$ ?

# $\Delta p_T(T_e)$ for V(001) emission

UIC

– DFT band structure with Fermi-Dirac distribution for electrons



# Electron-like vs. Hole-like States

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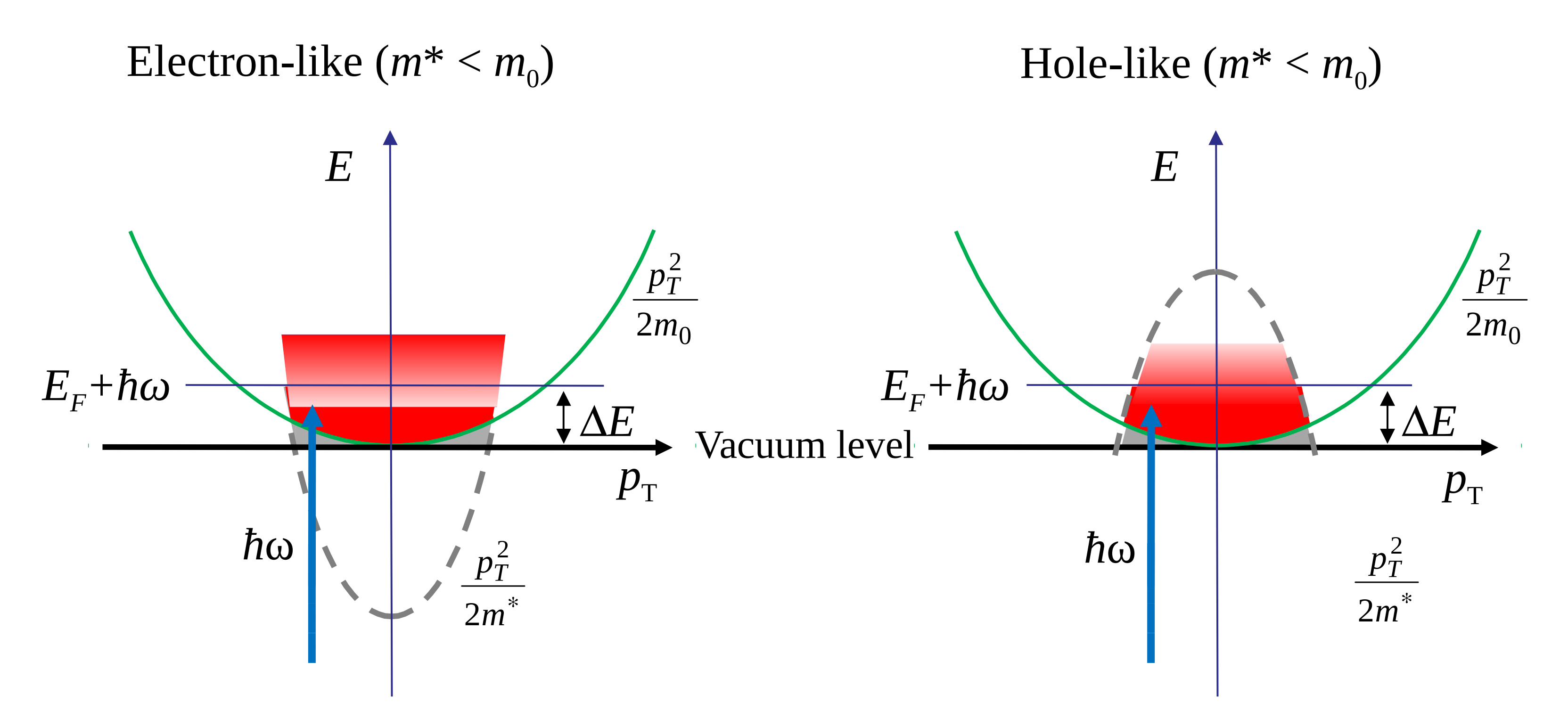
– $\Delta p_{T0}$ and its sensitivity to $T_e$




- Higher $p_T$ electrons at high $E$
- More high $p_T$ states occupied as $T_e$ increases

- Lower $p_T$ electrons at high $E$
- Less high $p_T$ states occupied as $T_e$ increases

∴ Low $m^*$ hole-like states preferred

# Temperature dependence: $T_e$

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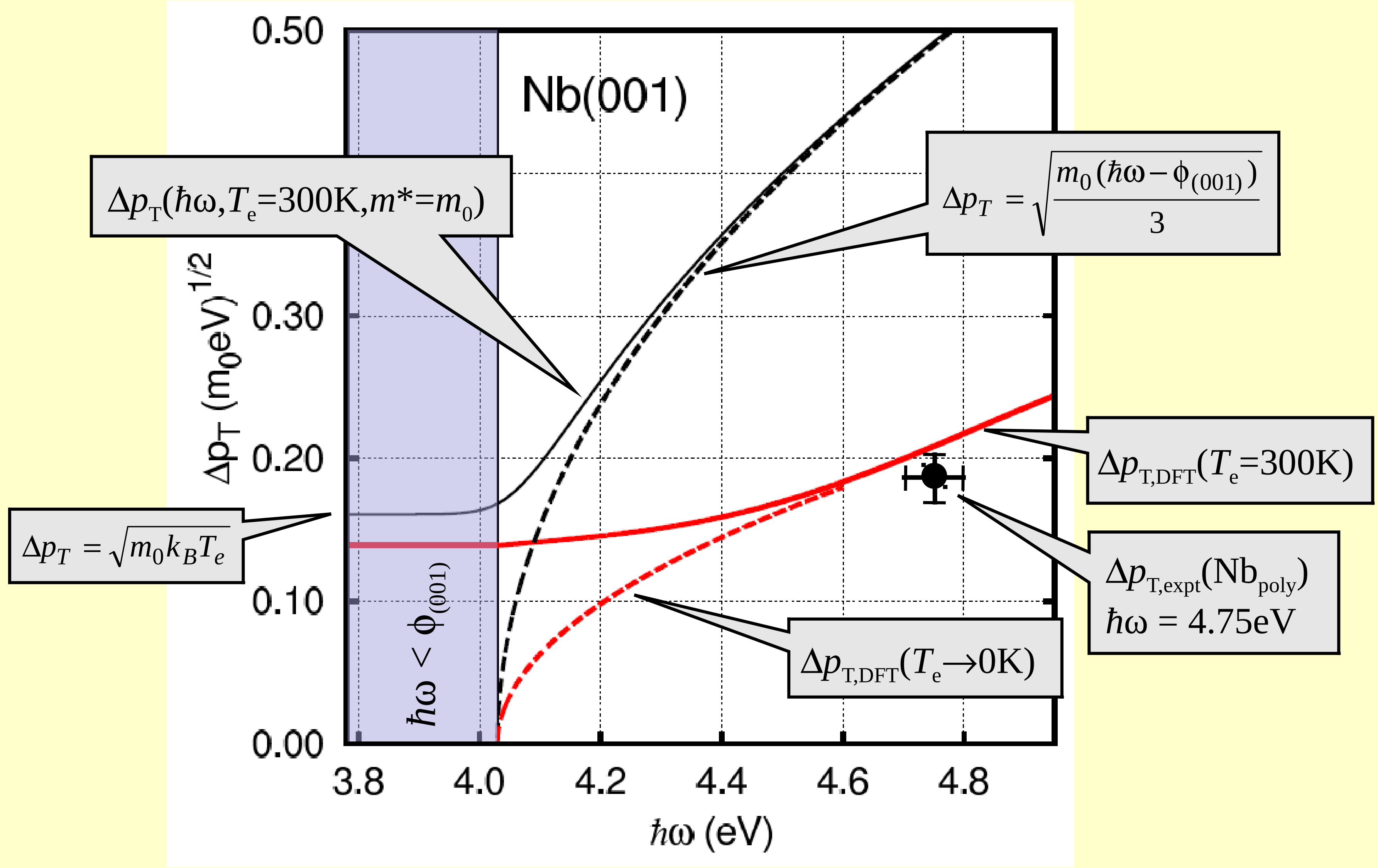


T. Vecchione et al., TUPSO83, *Proc. of FEL 2013*, pp. 424-426.

# Copper Photocathodes

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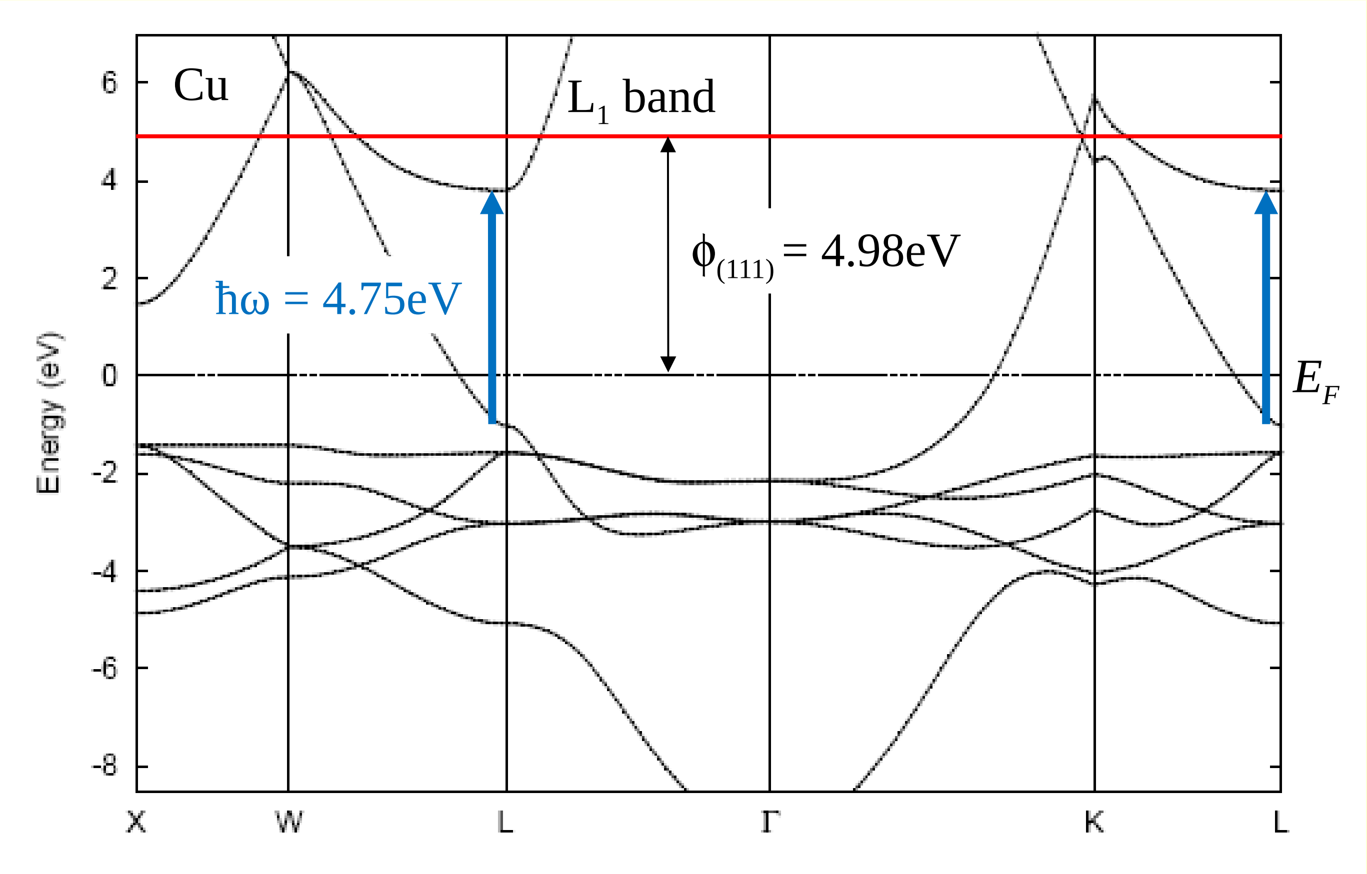
− Possible excited state emission process for Cu(111)




**NOTE:**

Could explain higher than expected QE for polycrystalline Cu photocathodes

R. Courths et al., *J. Phys. F: Met. Phys.* **14**, 1559-1572 (1984)

# HCP Metals: Be

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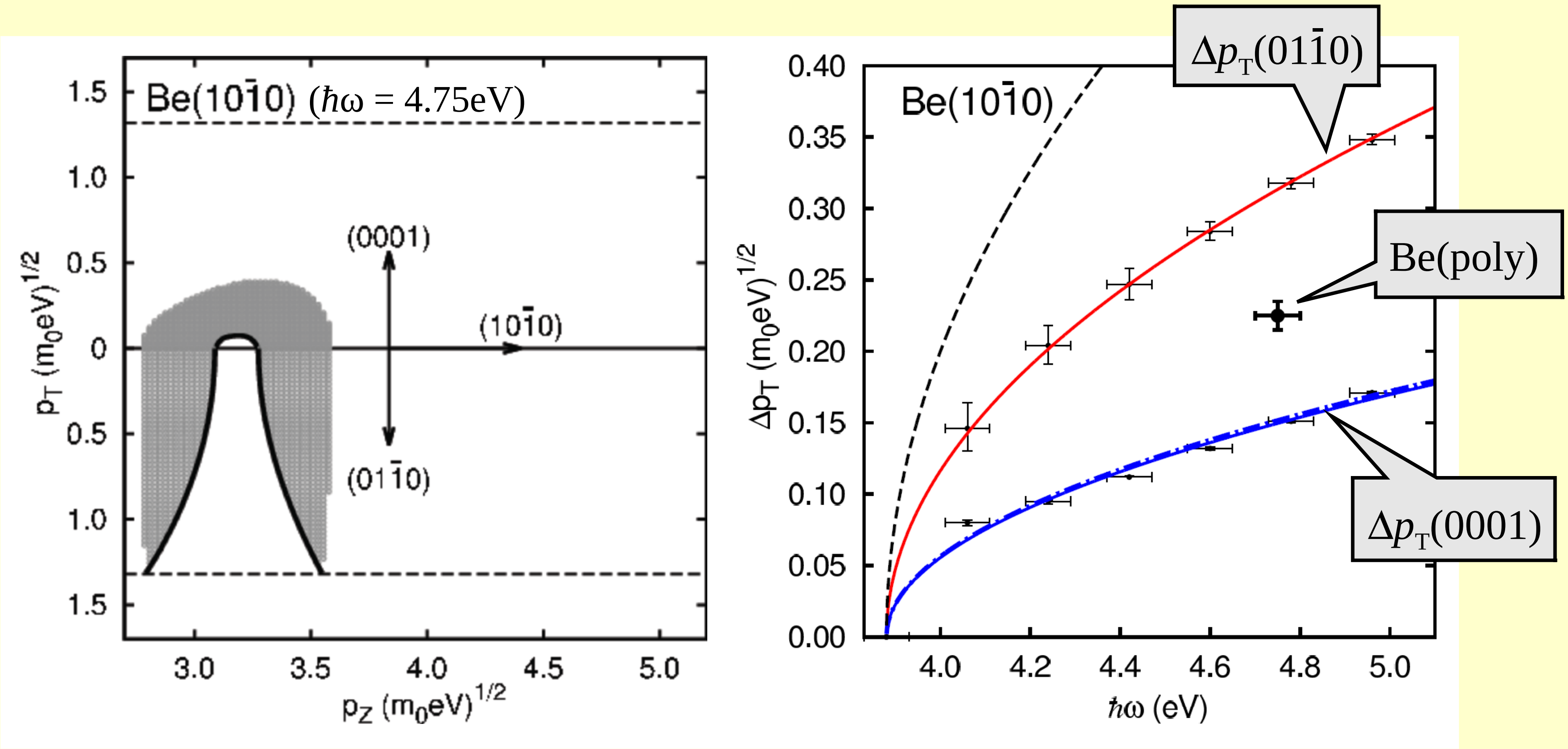
− Intrinsic electron state anisotropy in HCP crystal structure

- $(10\bar{1}0)$-face is most prevalent in polycrystalline HCP metals:




T. Li et al., *Phys. Rev. ST – Accel. & Beams* **18**, 073401 (2015)

# HCP Metals: Mg

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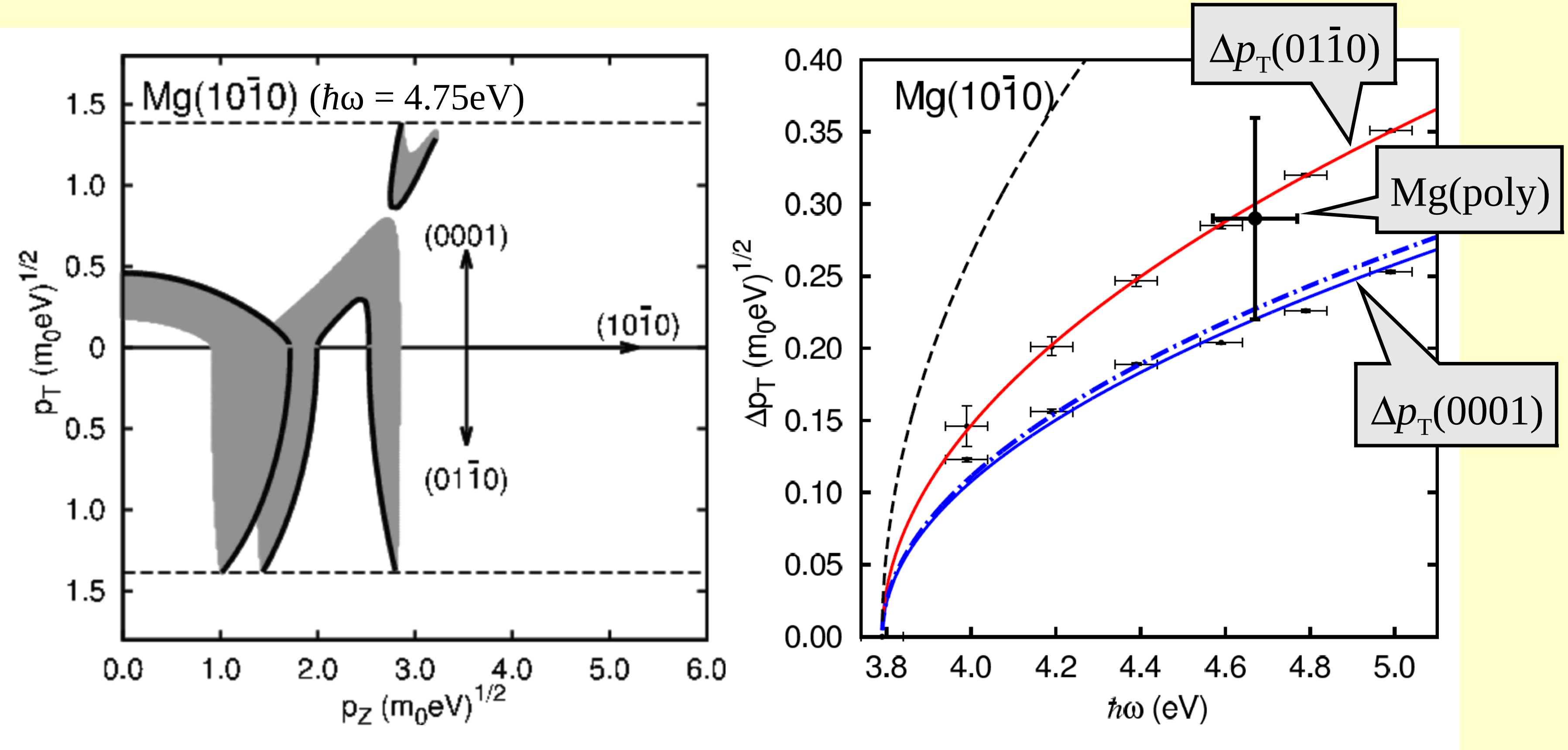
− Mg less anisotropic than Be




X.J. Wang et al., *Proceedings of LINAC2002*, Gyeongju, Korea.

# Summary

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- Photocathodes for UED
  - ***Single-crystals*** for homogeneous electron beam generation ($\hbar\omega > \phi_{(ijk)}$)
    $\Rightarrow$ Higher $\eta_{PE}$ (?) and higher conductivity ($\sigma$ and $\kappa$)
  - Instantaneous response $\Rightarrow$ ***Virtual*** excited state emission
  - Emission from low $m^*$ states: $p_F > p_{T,max}$ $\Rightarrow$ Lower $\Delta p_{T0}$
  - 'Hole-like' emission states are preferred:
    Even lower $\Delta p_{T0}$ and less sensitive to $T_e$ (e.g., laser heating)
  - Robust (e.g., high m.p.) and chemically inert
- Future work
  - Direct comparison with theory: Single-crystal photocathodes
  - Crystalline compounds: $Mo_xNb_{1-x}$, semiconductors, $A_3B$ (e.g., $Nb_3Sb$), …
  - Search for ***ultra-low*** $\Delta p_{T0}$ solid-state photocathodes:
    $\Delta p_{T0}$ approaching cold atom electron sources $\Leftarrow$ TID issues?

# Thank you!

# GaSb band structure

UIC

– Vacuum level at $\phi_{eff}$ = 4.84eV above bulk VB maximum

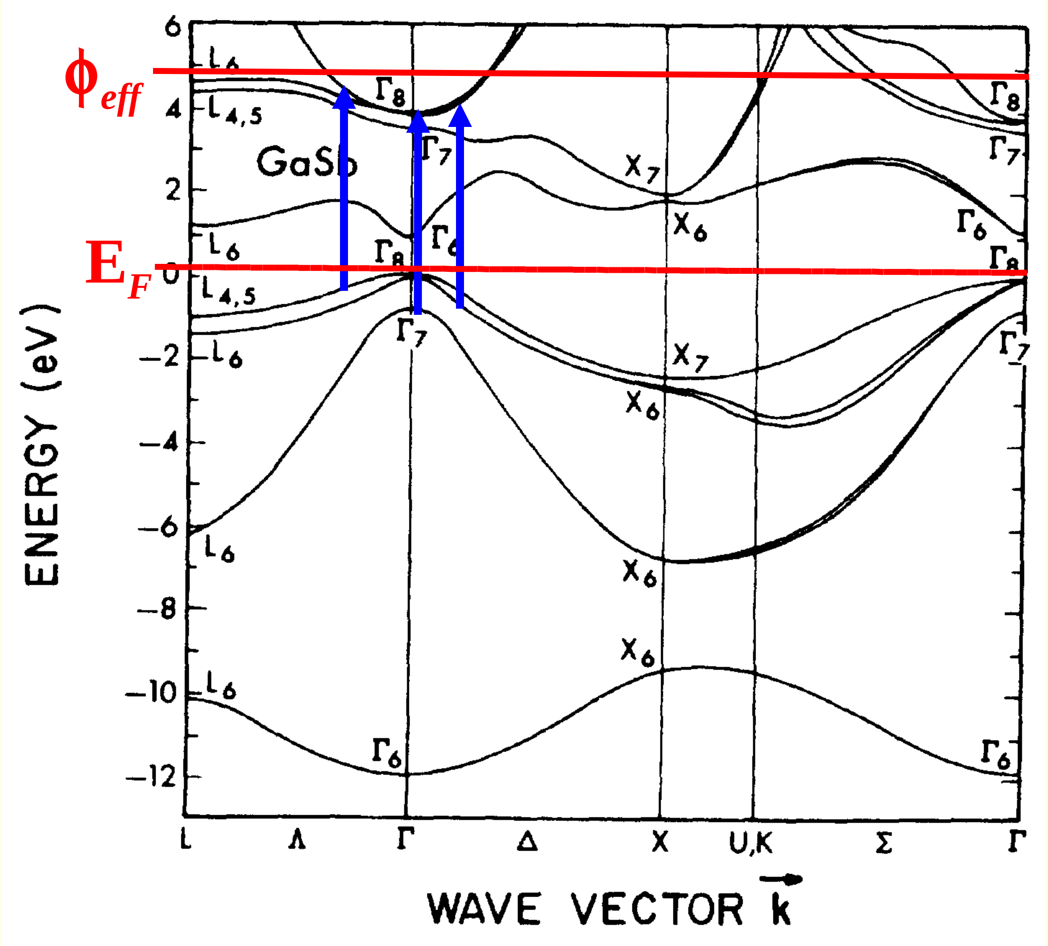
- Strong absorption at 261nm:

  $\alpha = 1.44\text{x}10^{6}\text{cm}^{-1}$

  $\Rightarrow \alpha^{-1} \approx 7\text{nm}$

  … 'metal-like'

- Γ-valley transitions from VB (HH, LH, and SO bands) to upper $\Gamma_8$ conduction band





J.R. Chelikowsky & M.L. Cohen, *Phys. Rev. B* **14** (1976) 556

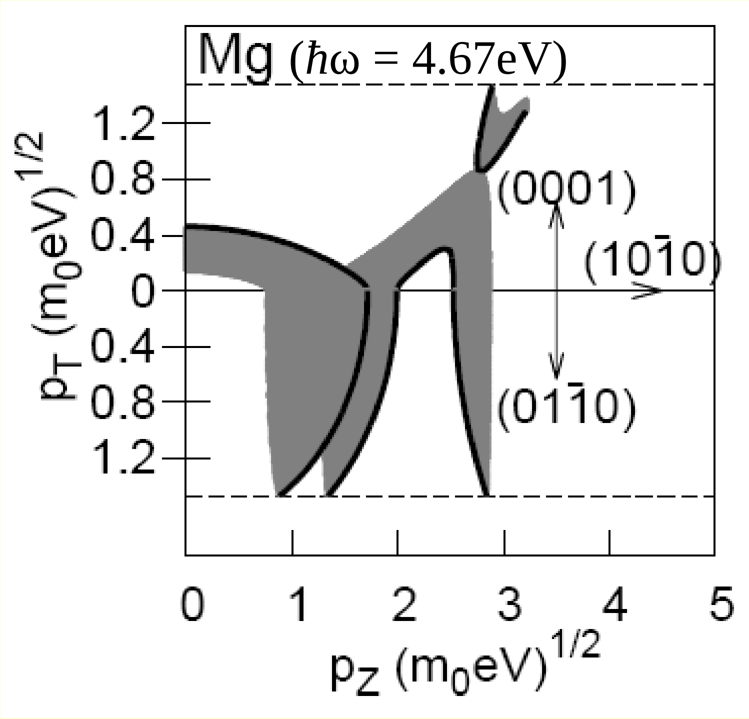
D.E. Aspnes & A.A. Studna, *Phys. Rev. B* 27 (1983) 985

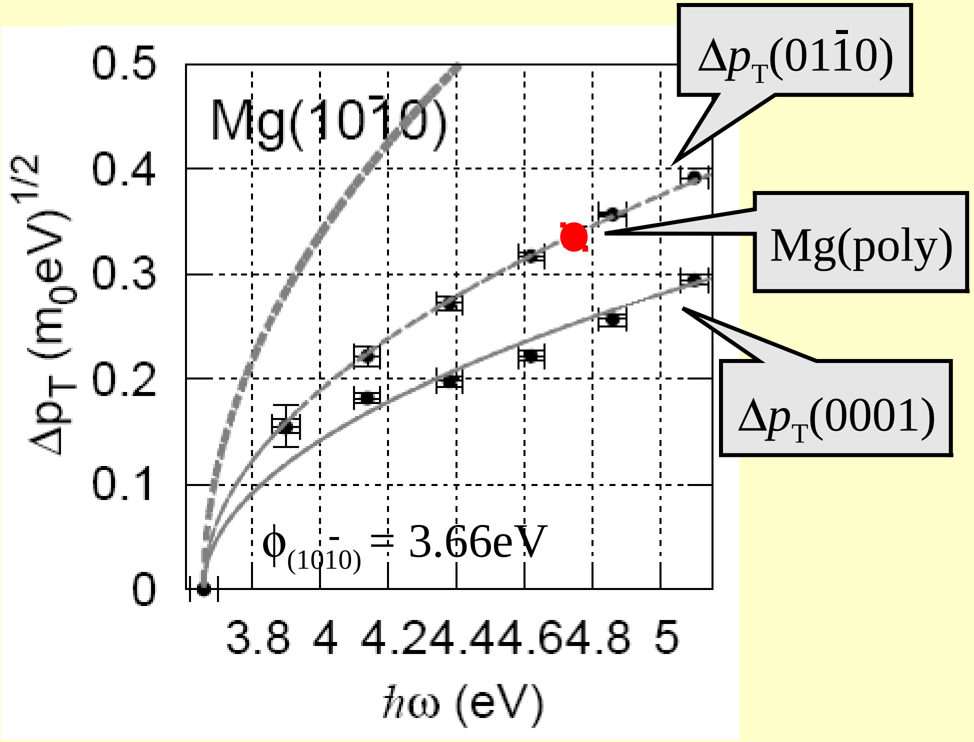
# Finally …

## Calculating $\Delta p_T$(ijk) for ***all*** elemental metals

– Results available on-line at http://people.uic.edu/~tli27/Database.html.

E.g., hcp Mg(10$\bar{1}$0) face emission:




X.J. Wang et al., *Proceedings of LINAC2002*, Gyeongju, Korea.

# Summary

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