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Oriented single crystal photocathodes: A route to high-quality electron pulses

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DE-FG52-09NA29451

UED: Resolution Limit

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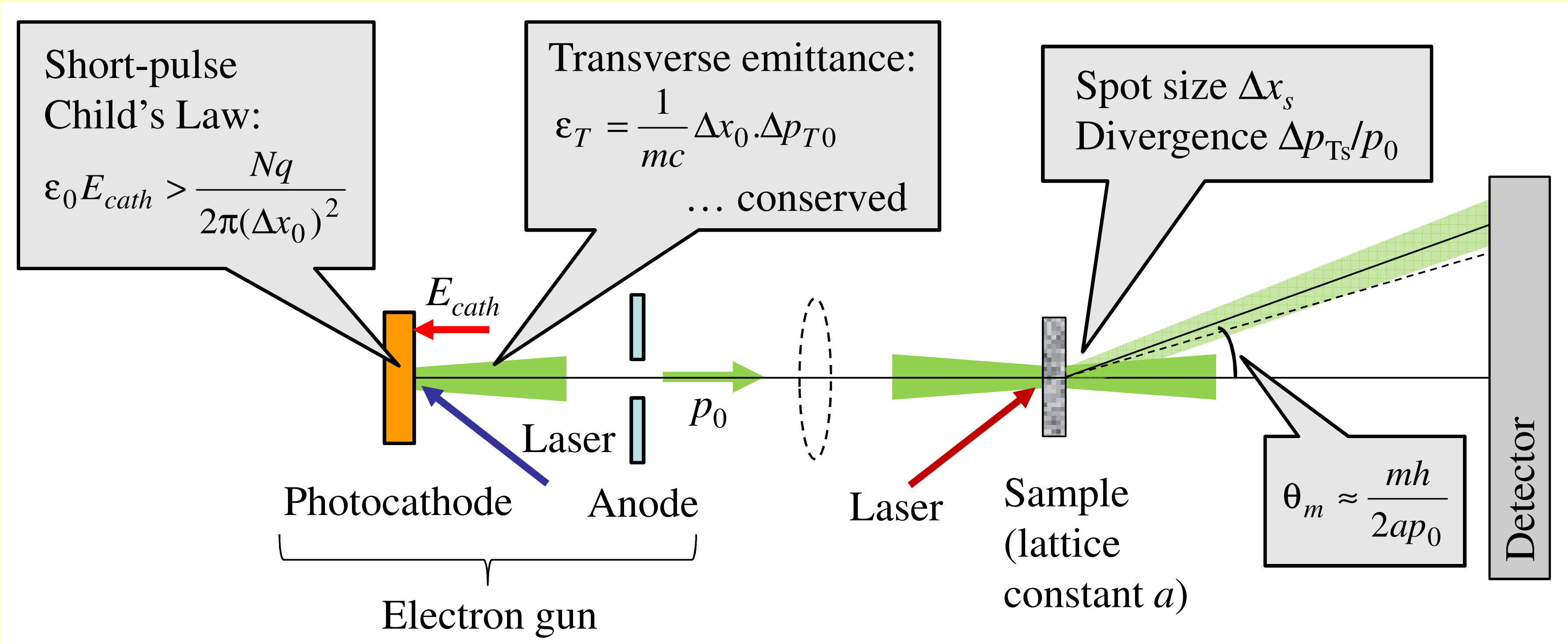
– Non-relativistic regime



$$\Rightarrow \text{Observable} \quad \frac{\delta a}{a} \geq \frac{\xi}{mh}\left(\frac{a}{\Delta x_s}\right)\sqrt{\frac{2Nq}{\pi\varepsilon_0 E_{cath}}}\,.\Delta p_{T0} \;\; ; \;\; \text{with} \;\; \Delta p_{T0} = \sqrt{\frac{m_0(\hbar\omega - \phi)}{3}} \quad ??$$

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

Photoemission Theory I

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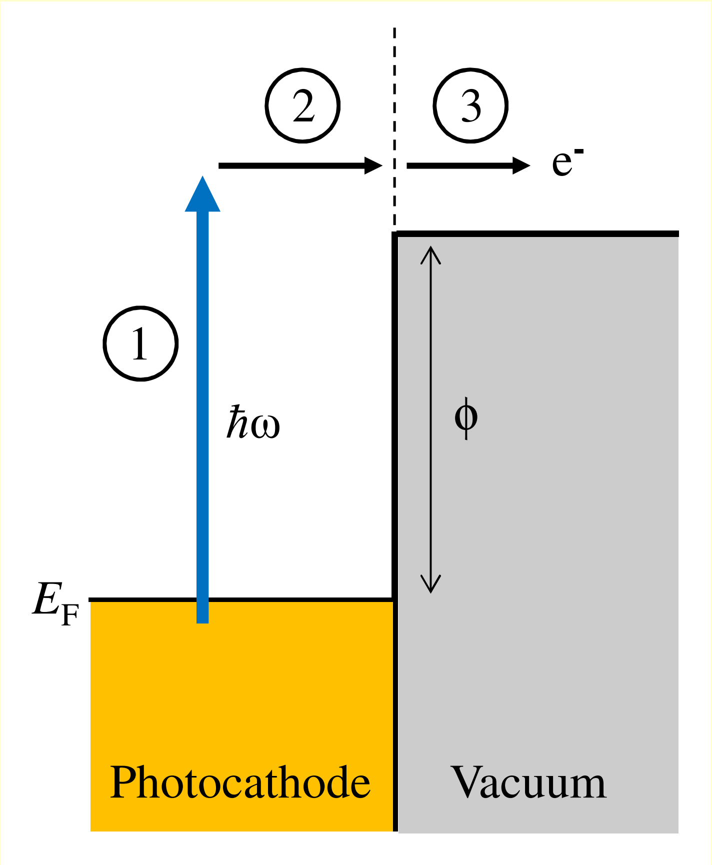
– The semi-classical three-step 'Spicer' model



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 (η_{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

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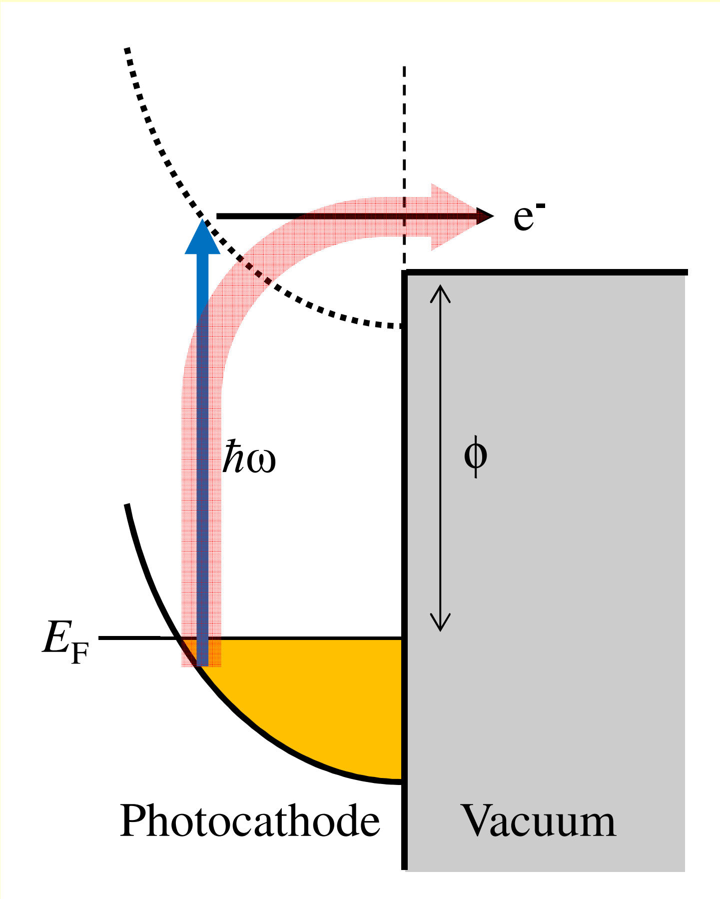
– The 'quantum mechanical' one-step model



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

- Low η_{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)

Experiment: Solenoid Scan

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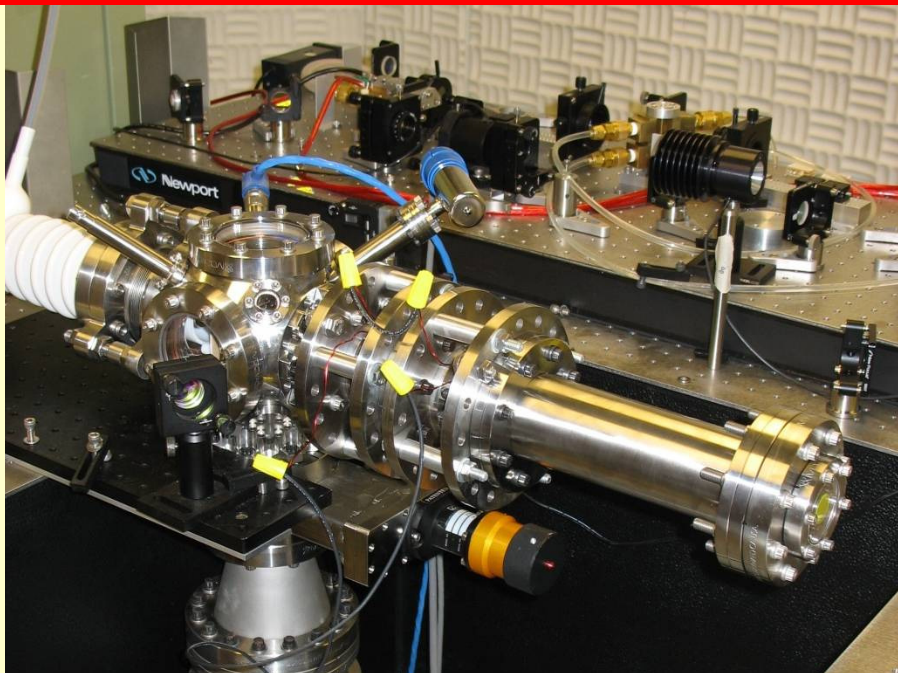
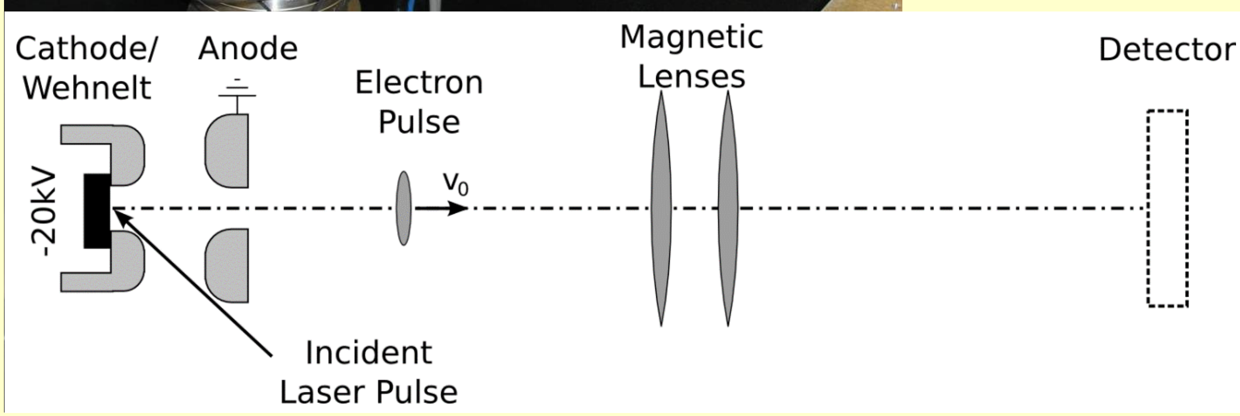


- 2W, 250fs, 63MHz , diode-pumped Yb:KGW laser
  - ~4ps at 261nm ($\hbar\omega$ = 4.75eV)
- YAG scintillator optically coupled to CCD camera
  - Beam size vs. magnetic coil (lens) current measured
  - Analytical Gaussian (AG) pulse propagation model to extract Δp_{T0}



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

Results: Polycrystalline Cr

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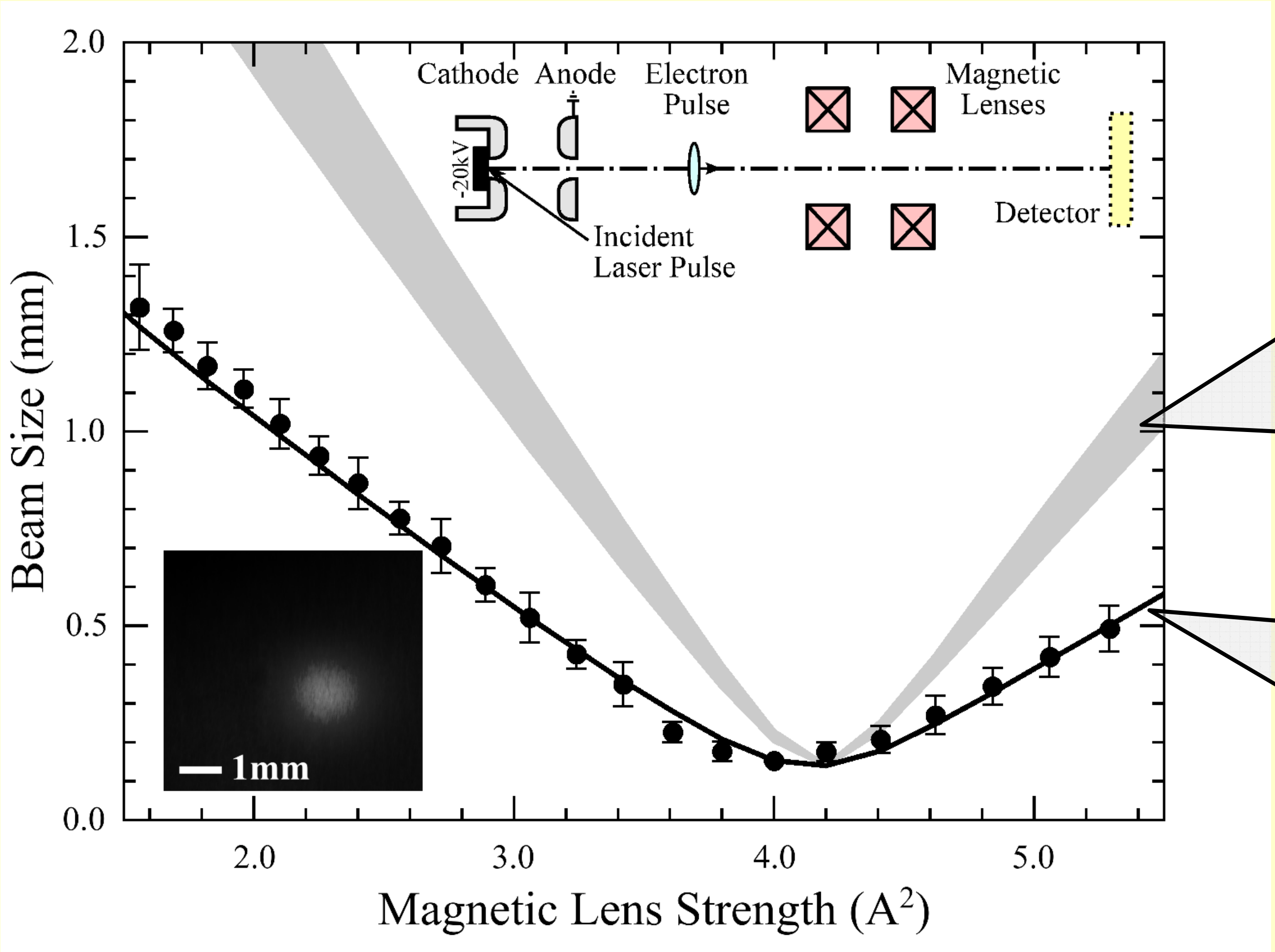
– Solenoid scan measurement



Range for Δp_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

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

Results: Metals

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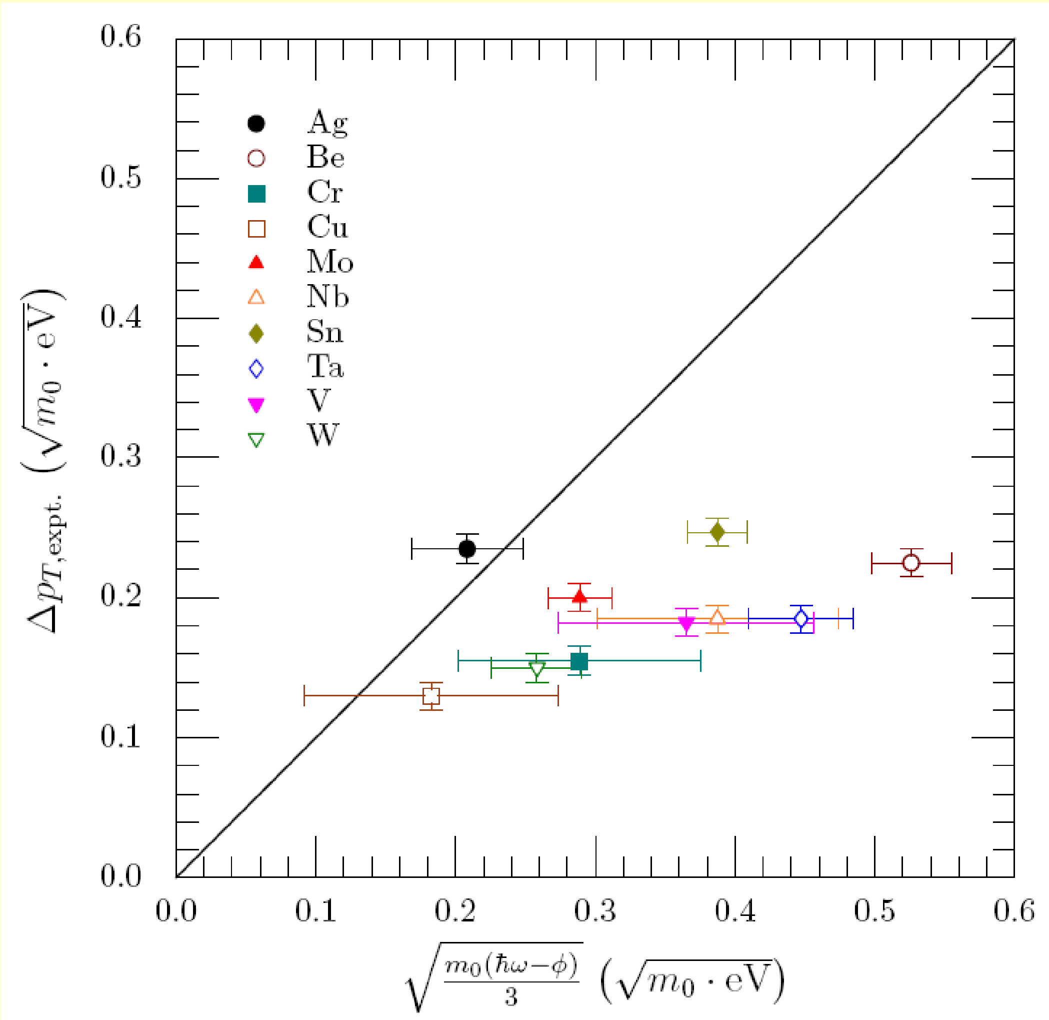
– Ten ***polycrystalline*** metal photocathodes



- Only Ag and Cu (noble metals) consistent with

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

for others

$$\Delta p_{T,\mathrm{expt}} < \sqrt{\frac{m_0(\hbar\omega - \phi)}{3}}$$

$\Rightarrow$ Band structure effects?
– e.g. $m^* < m_0$

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

Band Structure Effects

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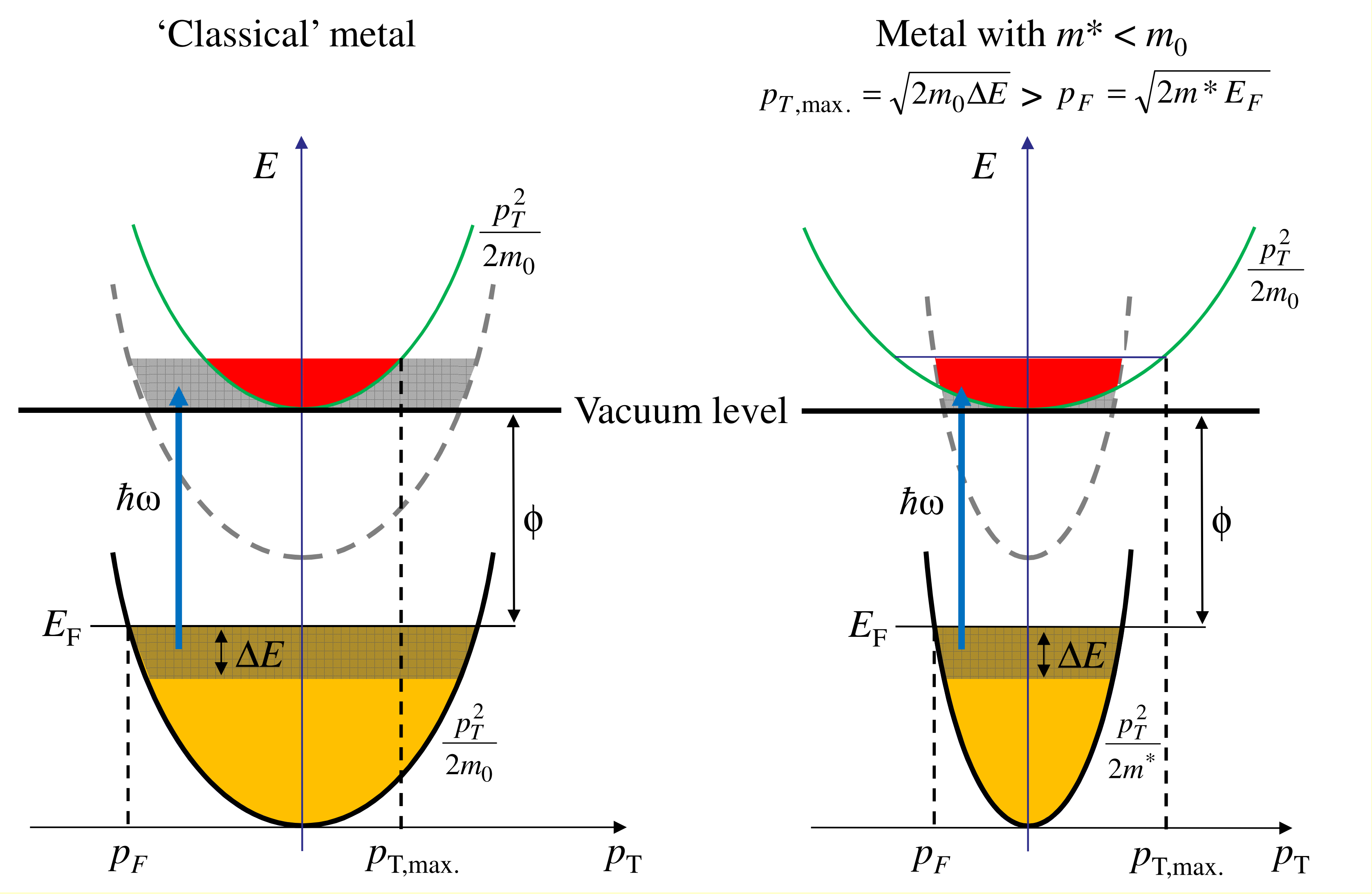
– Transverse momentum p_T conserved in photoemission



Fermi Surfaces

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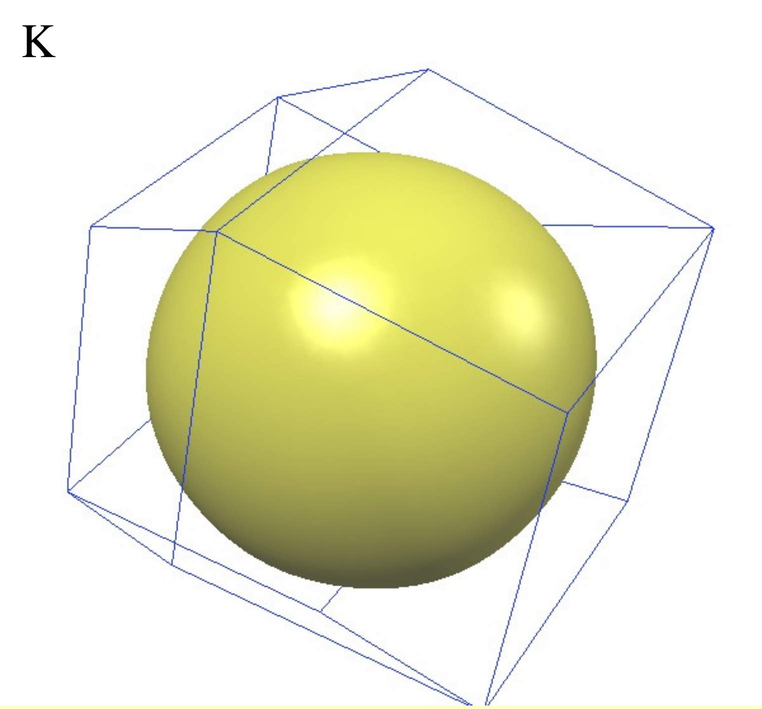
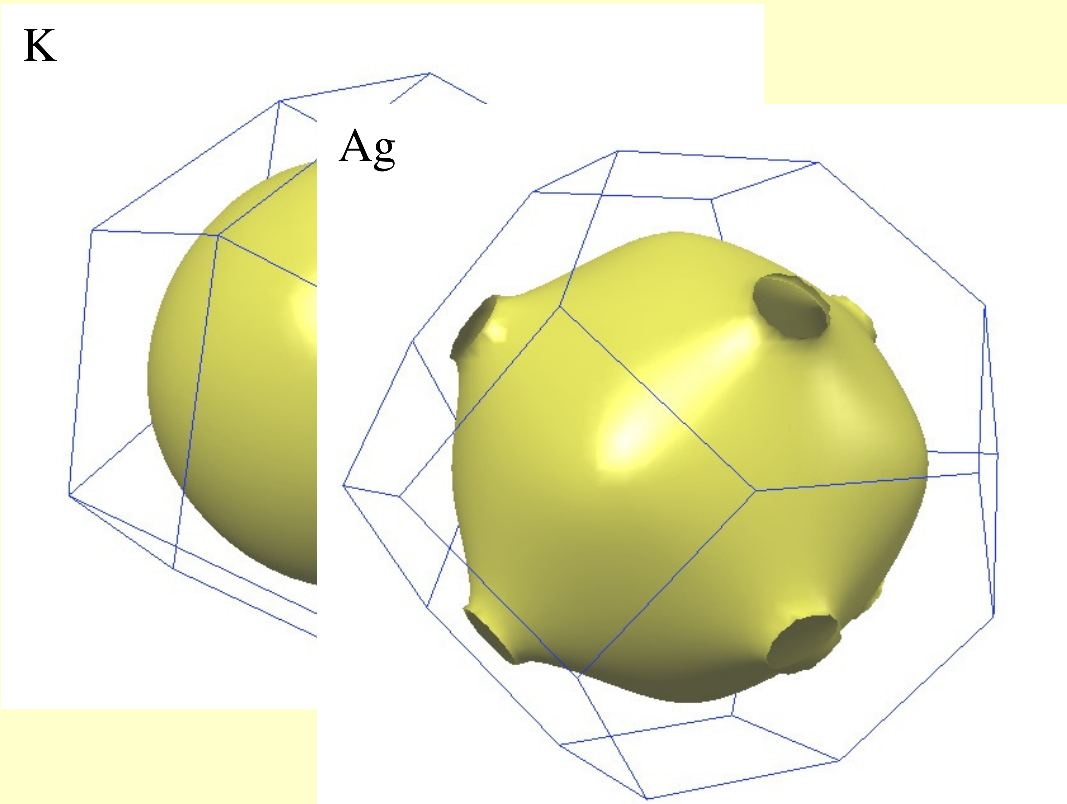


www.phys.ufl.edu/fermisurface/

Fermi Surfaces



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Fermi Surfaces

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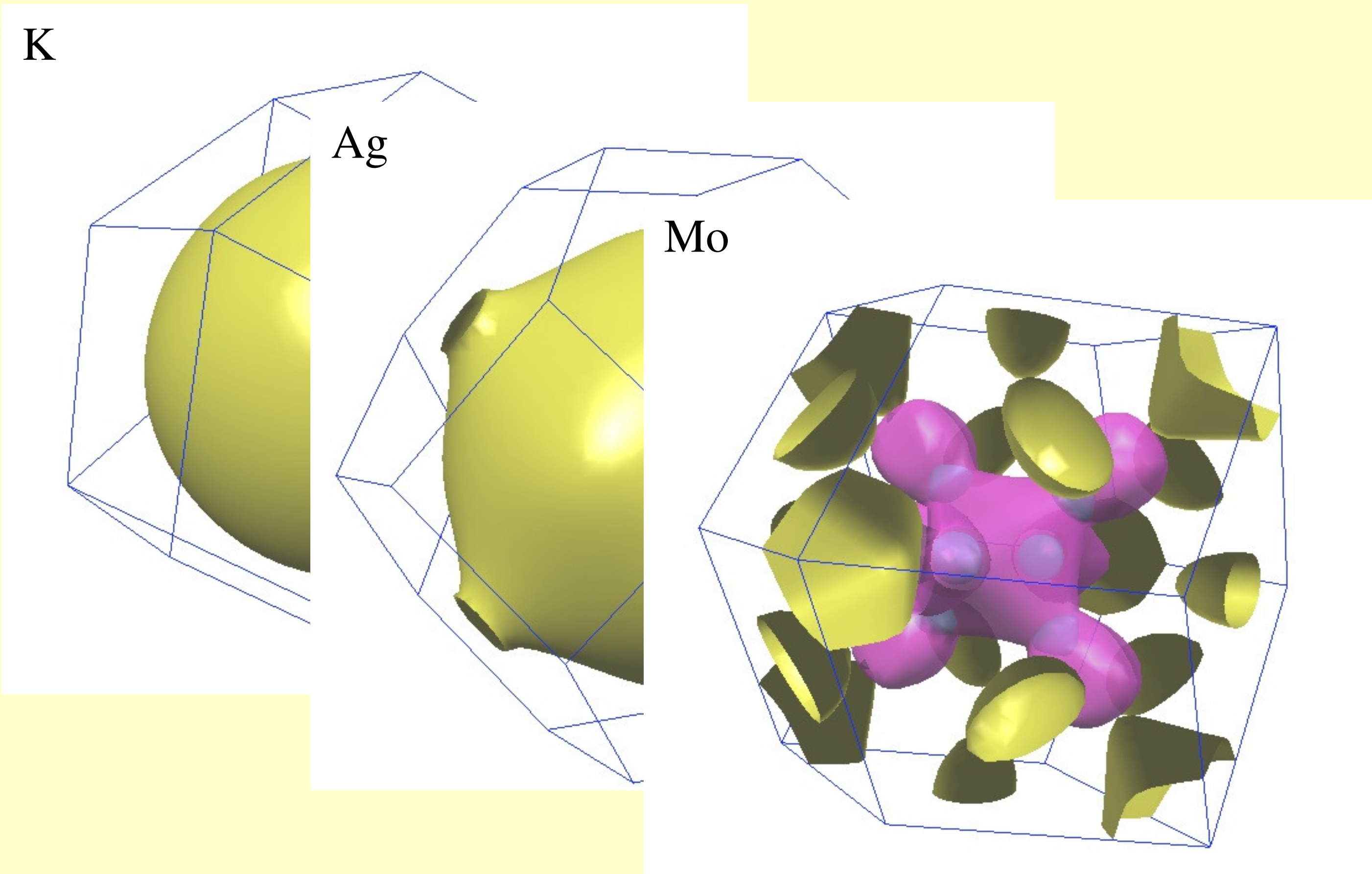

www.phys.ufl.edu/fermisurface/

Fermi Surfaces

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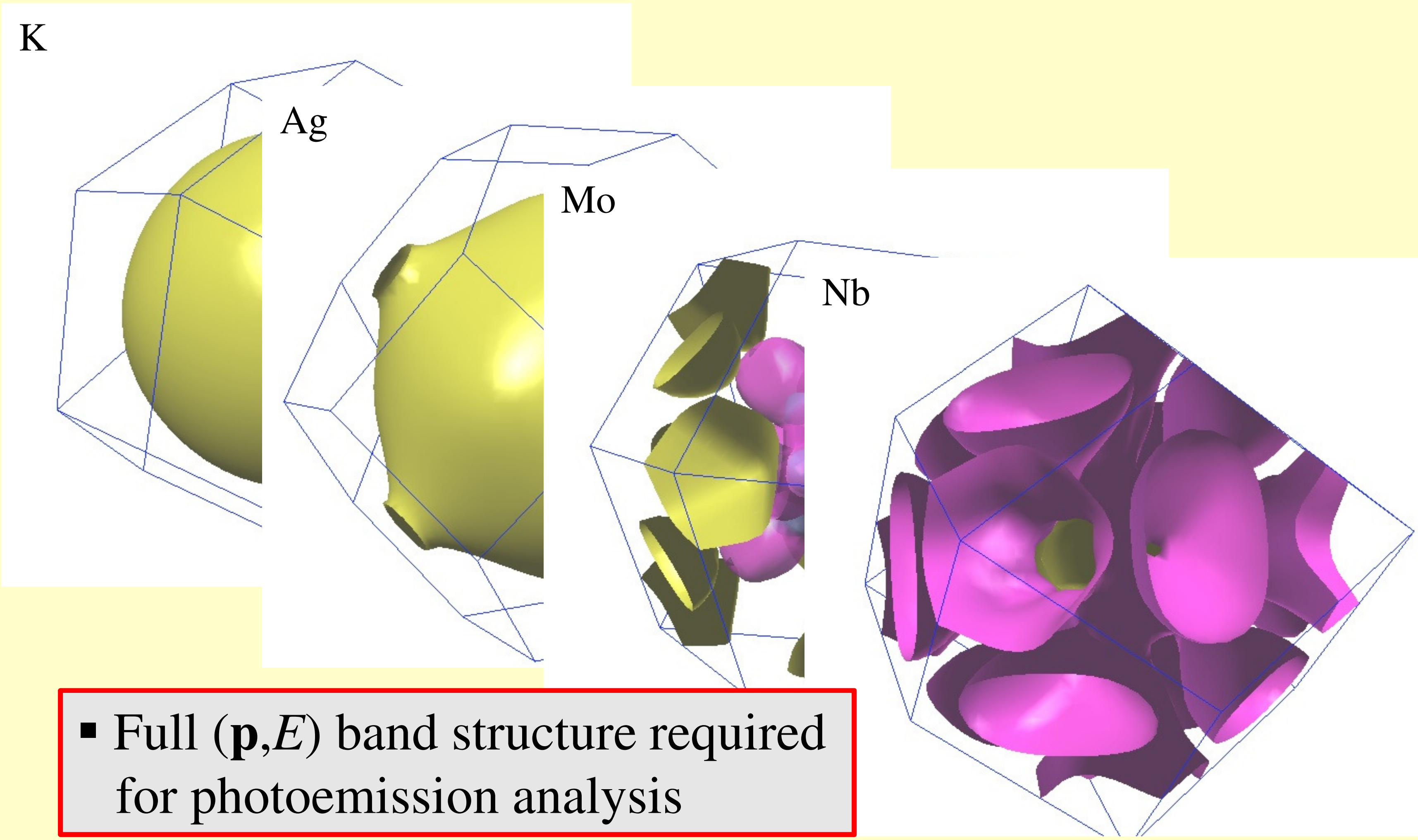

- Full (**p**,*E*) band structure required for photoemission analysis

www.phys.ufl.edu/fermisurface/

Work Function Anisotropy

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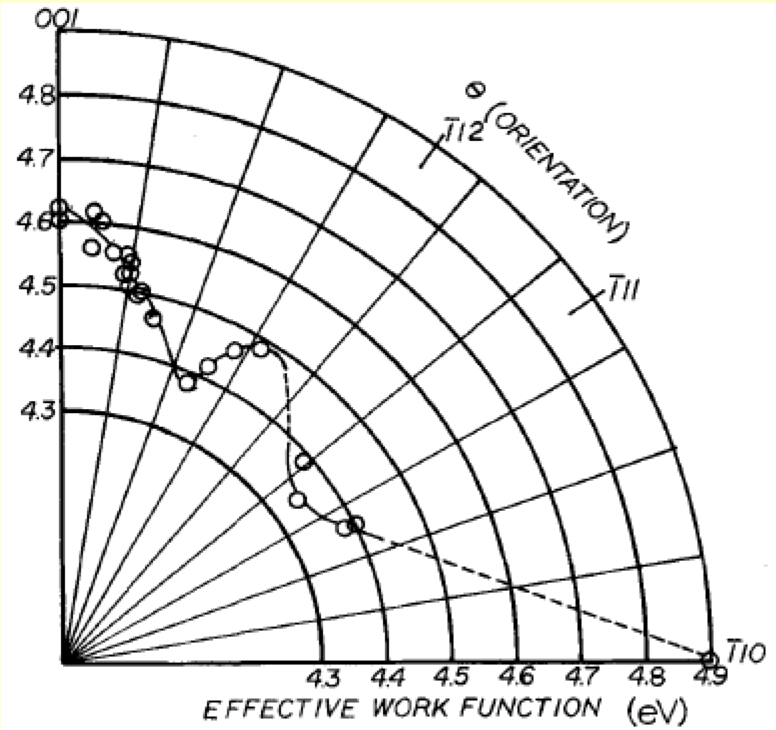
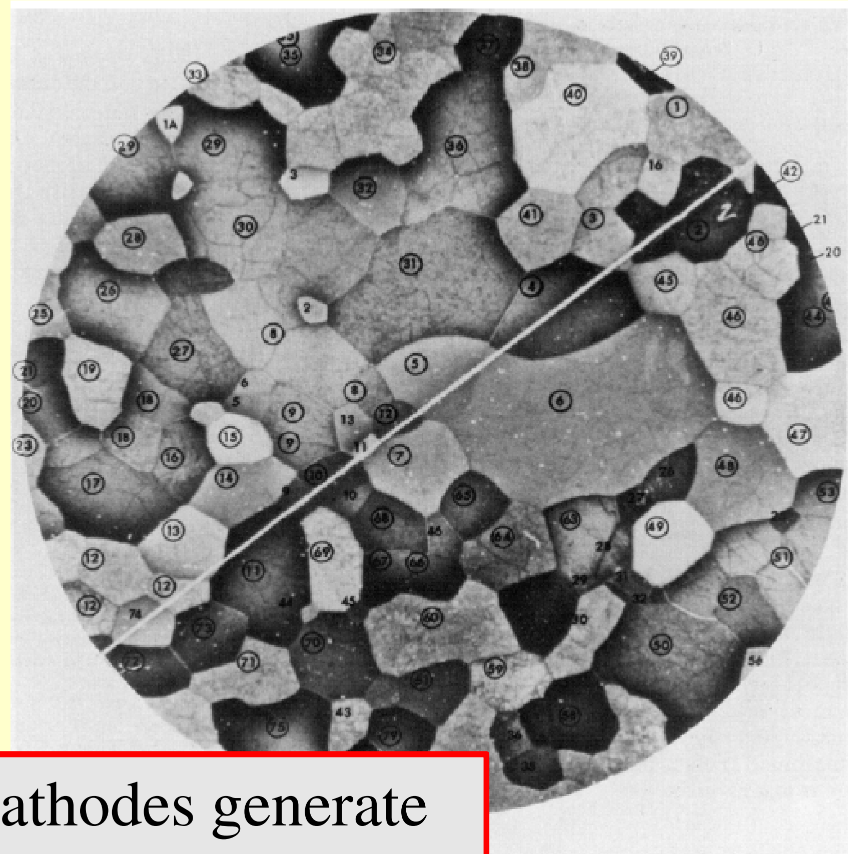
– Example: $\phi_{(ijk)}$ for Mo by electron emission microscopy





- 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 $\phi_{(ijk)}$

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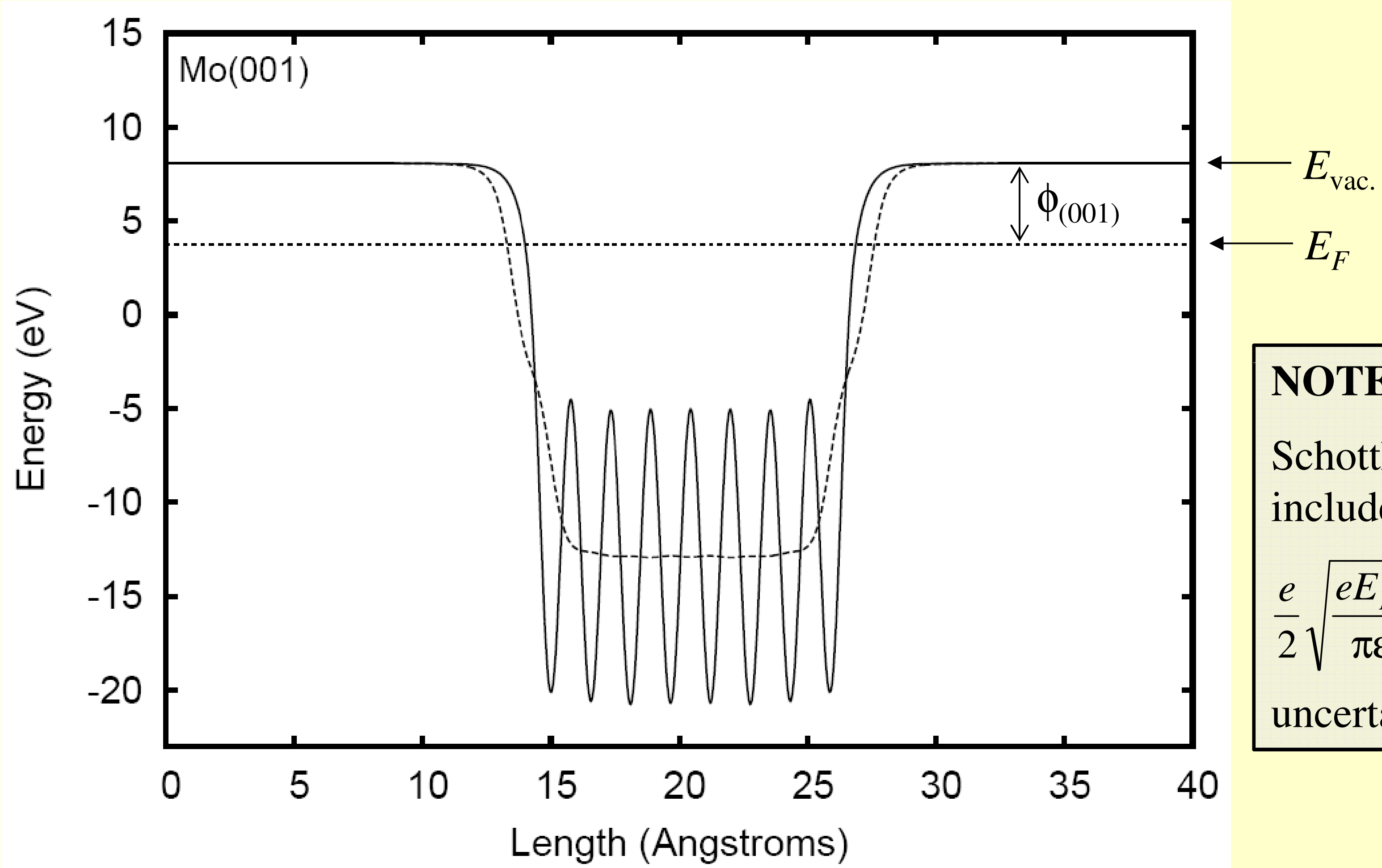
– Example: $\phi_{(001)} = 4.53(\pm 0.05)$eV for Mo



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

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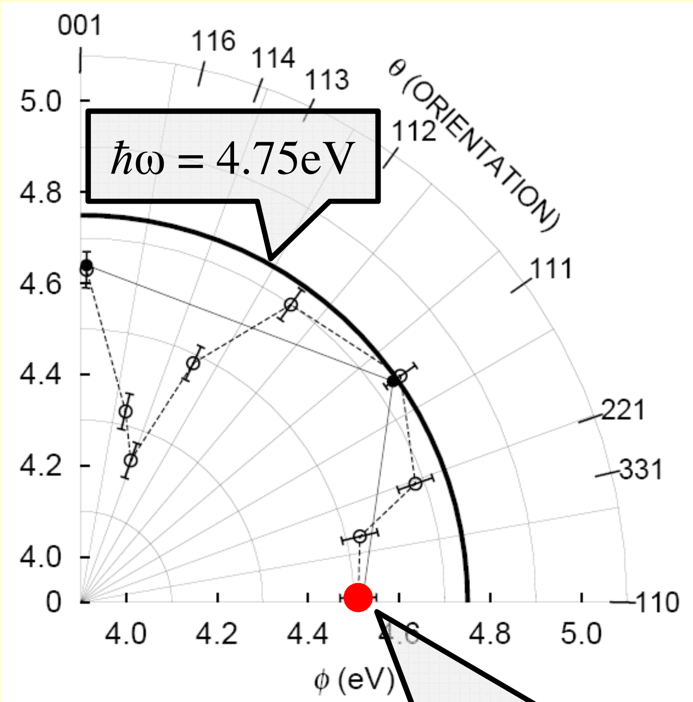
– fcc crystal lattice



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

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

… ±50meV error in $\phi_{(ijk)}$

Photoemission Simulation: Ag

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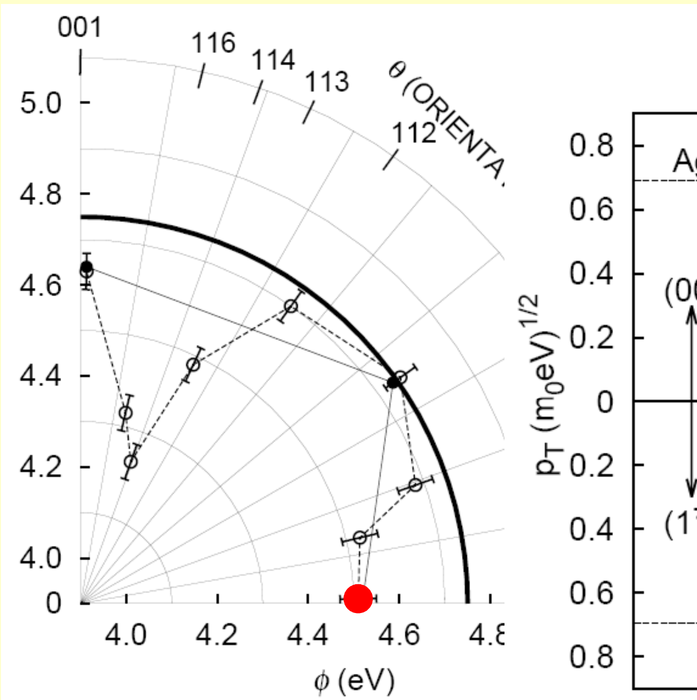
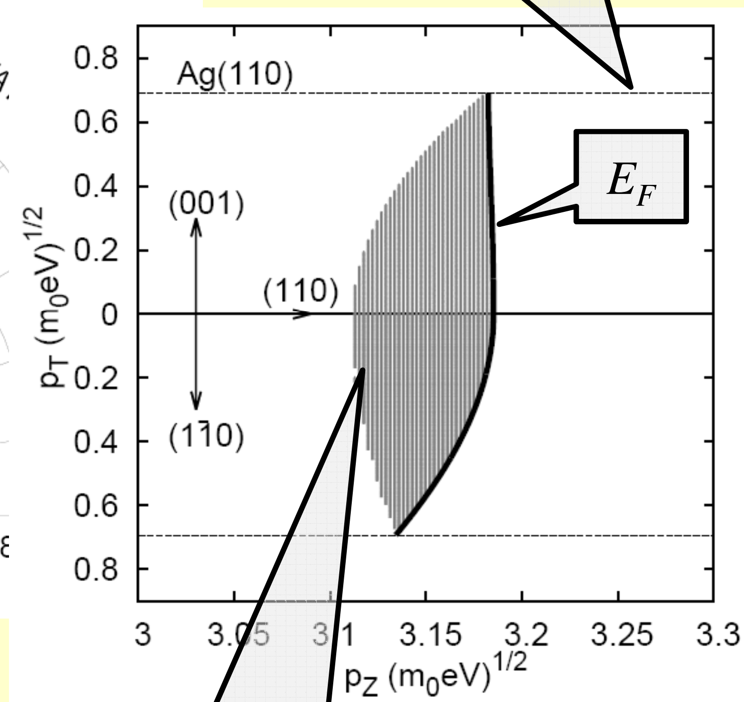
– fcc crystal lattice



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



E_F

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

Photoemission Simulation: Ag

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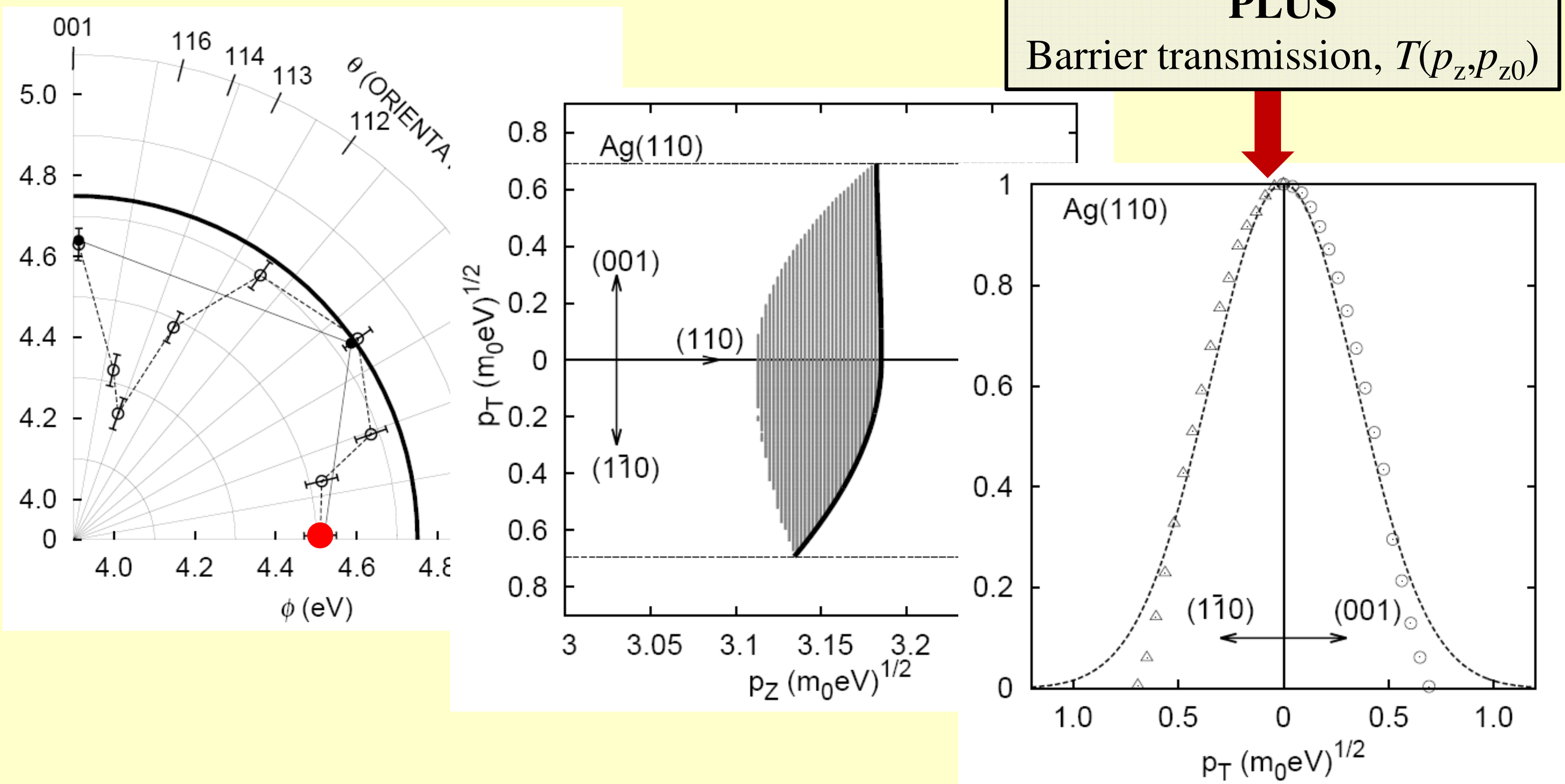
– fcc crystal lattice

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

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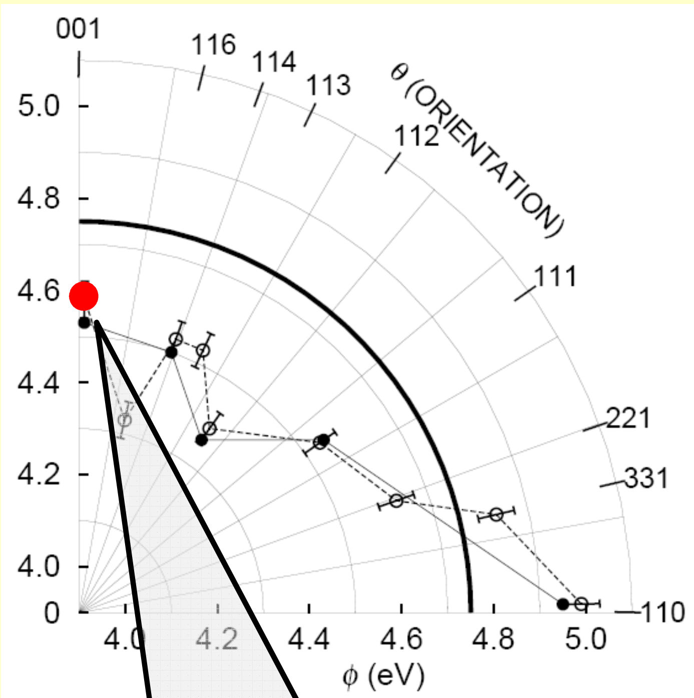
– bcc crystal lattice



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

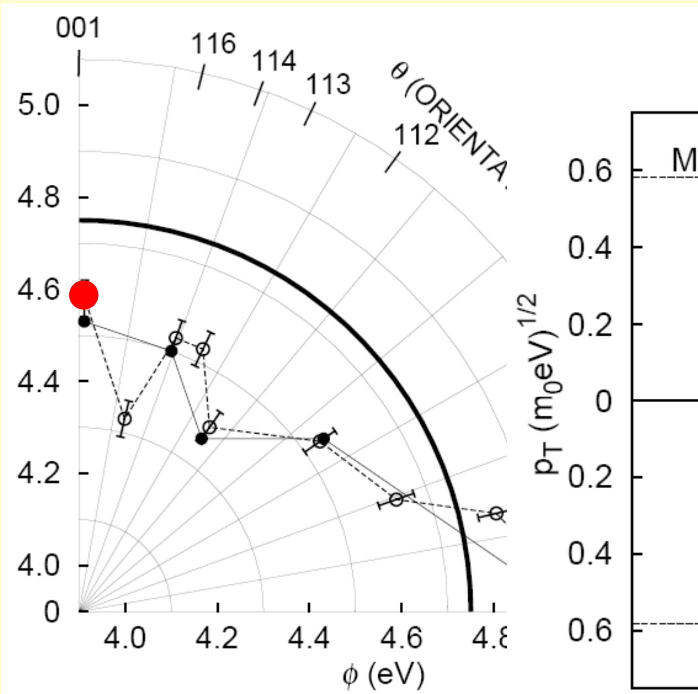
$\Delta E = \hbar\omega - \phi_{(001)} = 0.22\text{eV}$

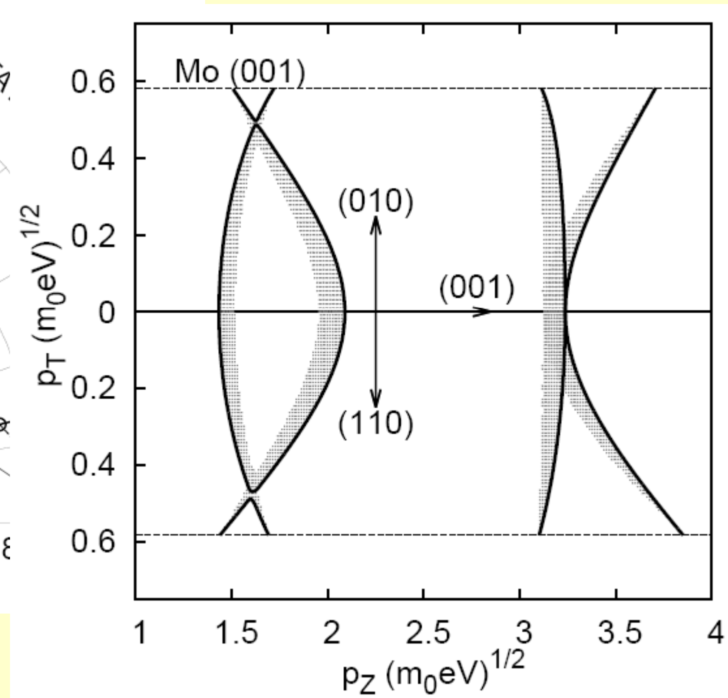
… ±50meV error in $\phi_{(ijk)}$

Photoemission Simulation: Mo

UIC

– bcc crystal lattice





Both electron- and hole-like states contribute to photoemission

Photoemission Simulation: Mo

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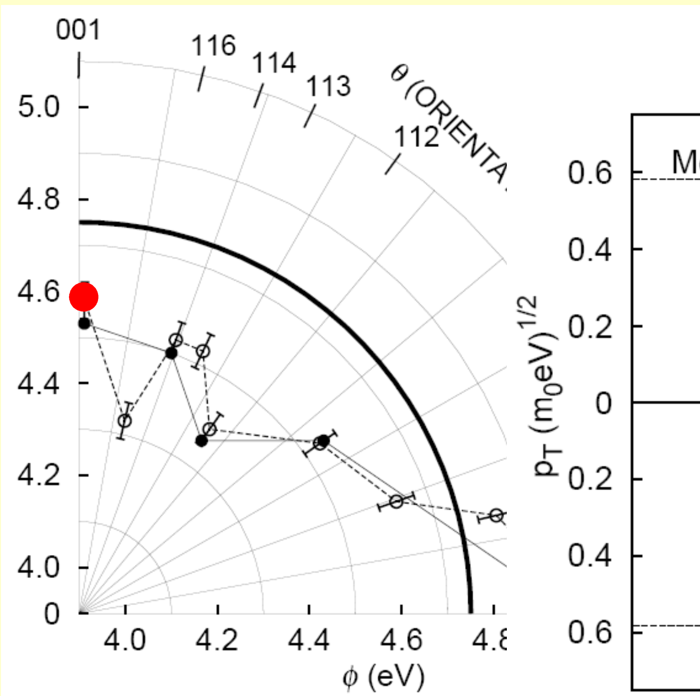
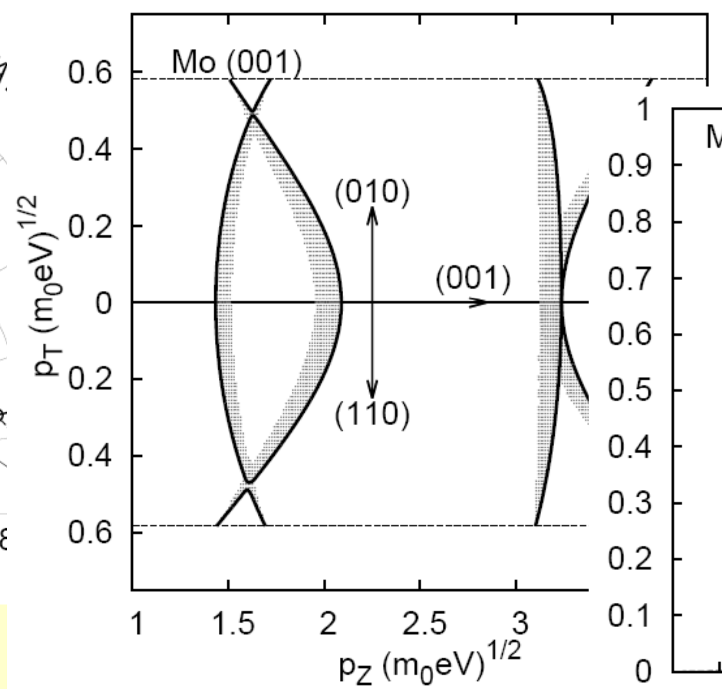
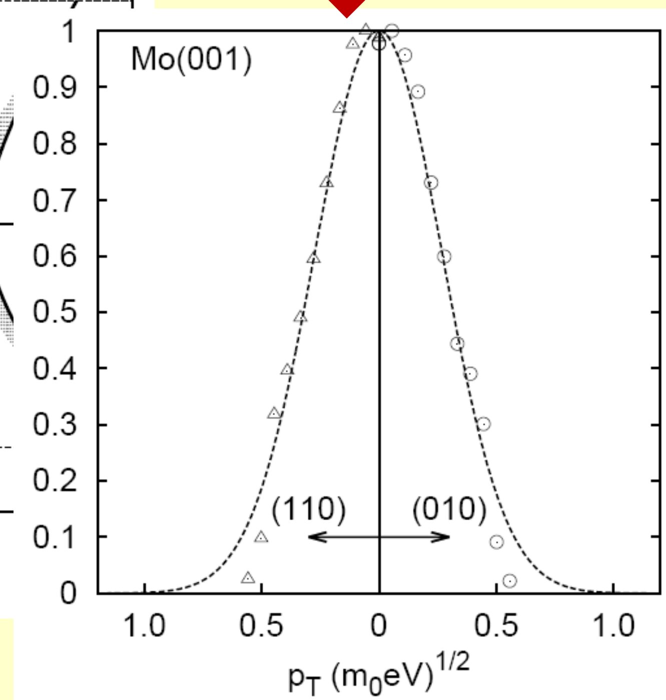
– bcc crystal lattice





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



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

Photoemission Simulation: Nb

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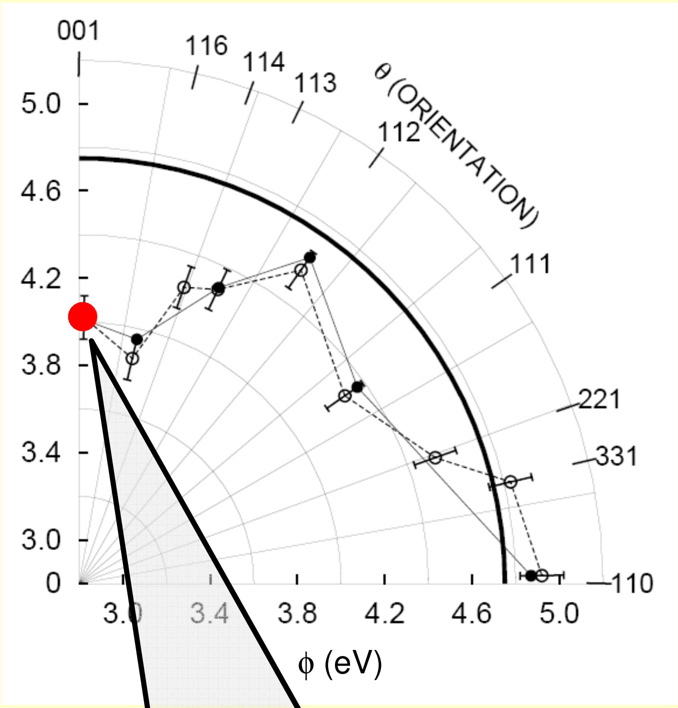
– bcc crystal lattice



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

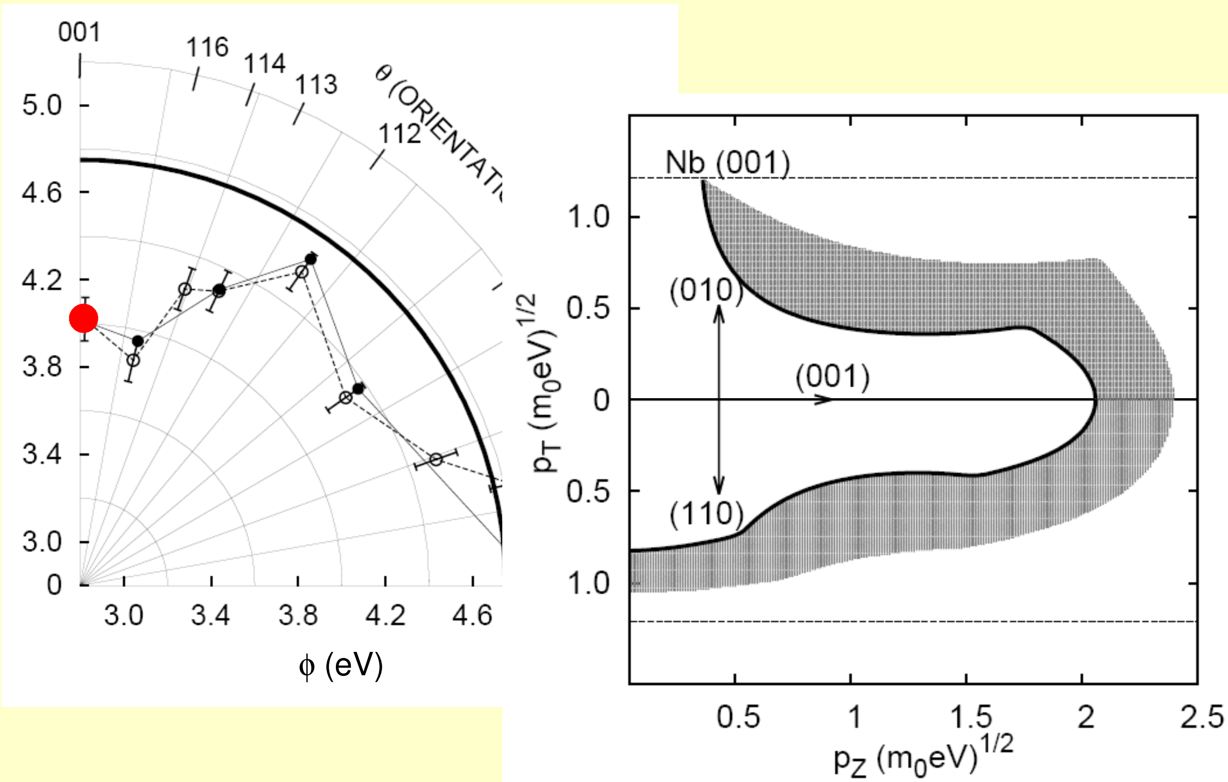
$\Delta E = \hbar\omega - \phi_{(001)} = 0.73\text{eV}$

… ±50meV error in $\phi_{(ijk)}$

Photoemission Simulation: Nb

UIC

– bcc crystal lattice



Only hole-like states contribute to photoemission

Photoemission Simulation: Nb

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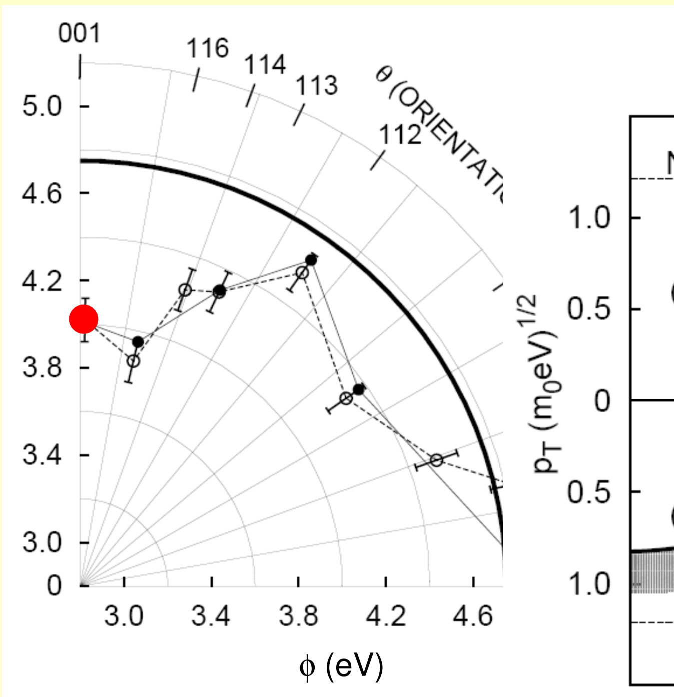
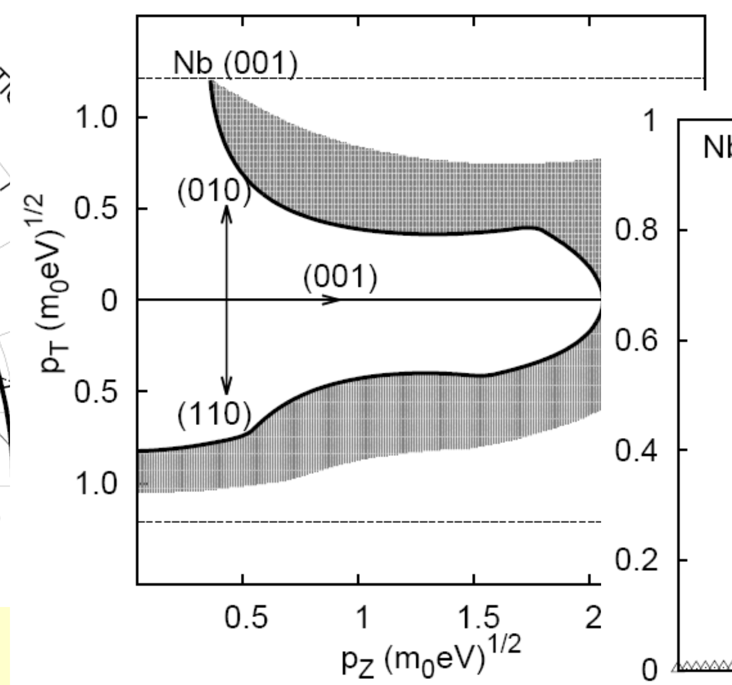
– bcc crystal lattice

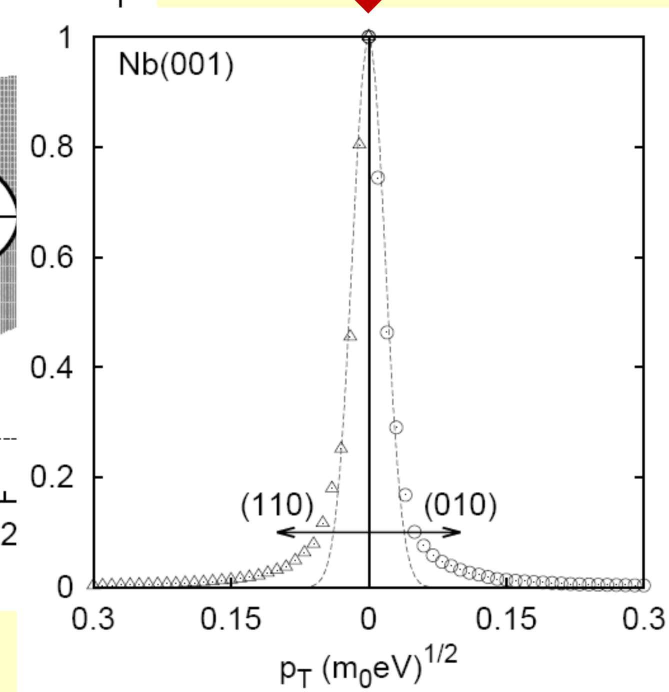




E, $\mathbf{p}_T$ conservation

PLUS

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



Spatially-averaged

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

Experiment vs. DFT Analysis

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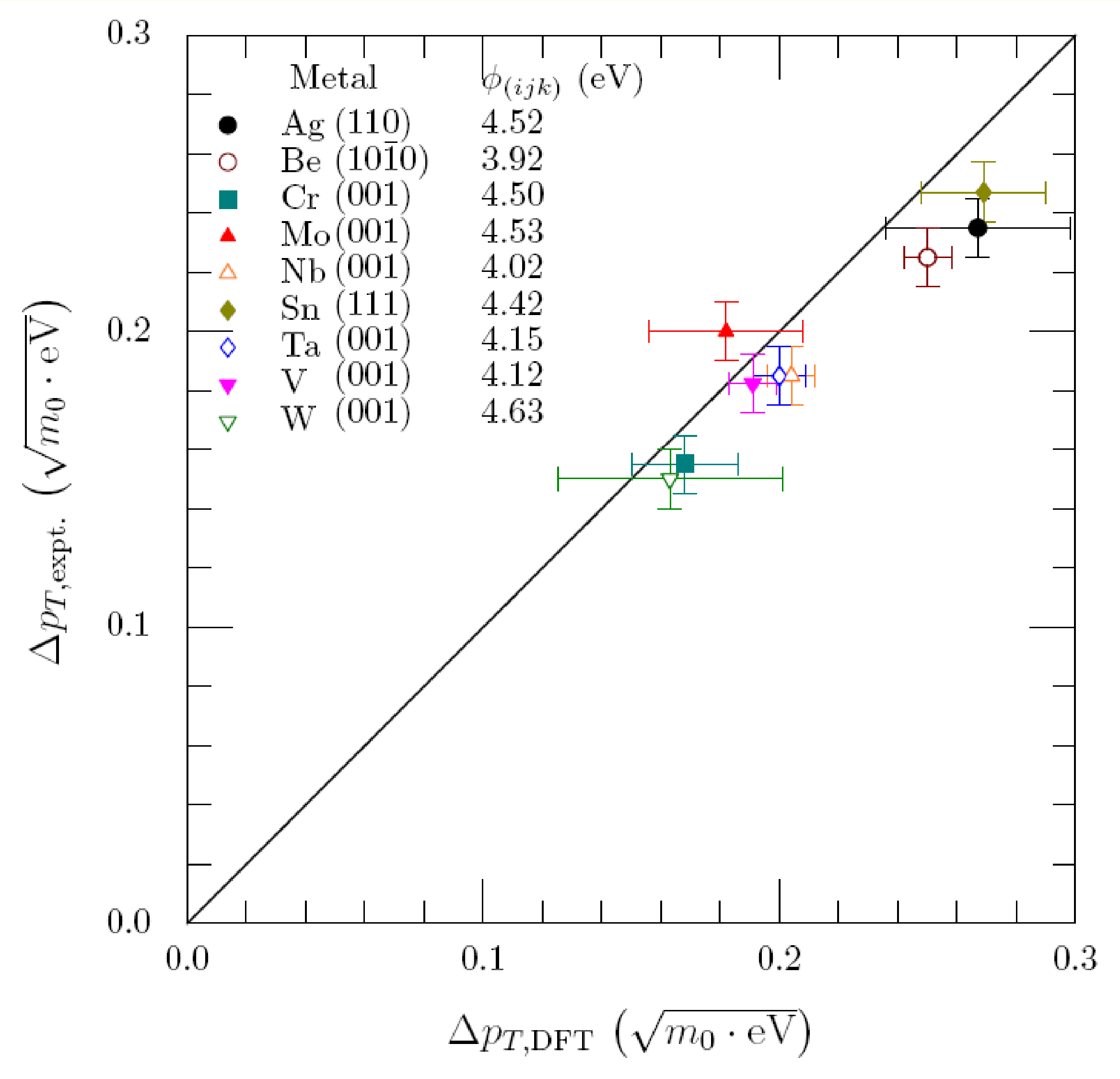

NOTE:

- Polycrystalline vs. single-crystal comparison
  - – Other crystal faces with smaller $\Delta E = \hbar\omega - \phi_{(ijk)}$ contribute lower Δp_{T0}
- DFT analysis at $T_e \rightarrow 0$K
  Experiment at 300K

Temperature dependence: T_e

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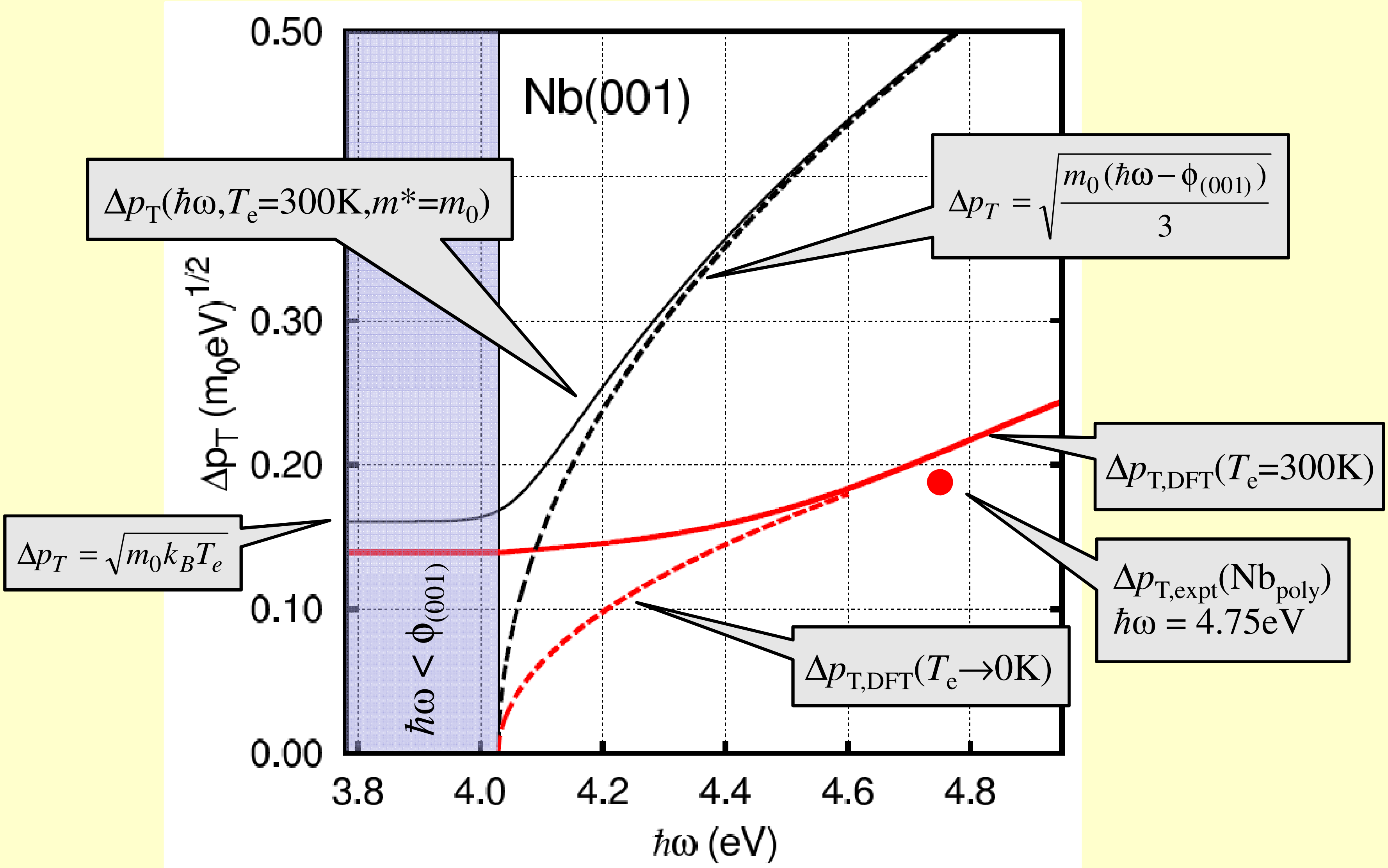

T. Vecchione et al., TUPSO83, *Proc. of FEL 2013*, pp. 424-426.

Summary

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- Photocathodes for UED
  - ***Single-crystals*** for homogeneous electron beam generation ($\hbar\omega > \phi_{(ijk)}$)
    $\Rightarrow$ Higher η_{PE} (?) and higher conductivity (σ and κ)
  - ***Virtual*** excited state emission $\Rightarrow$ Instantaneous response
  - Emission from low m^* states: $p_F > p_{T,max} \Rightarrow$ Lower Δp_{T0}
  - 'Hole-like' emission states are preferred:
    Even lower Δp_{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*** Δp_{T0} solid-state photocathodes:
    Δp_{T0} approaching cold atom electron sources $\Leftarrow$ TID issues?

Finally …

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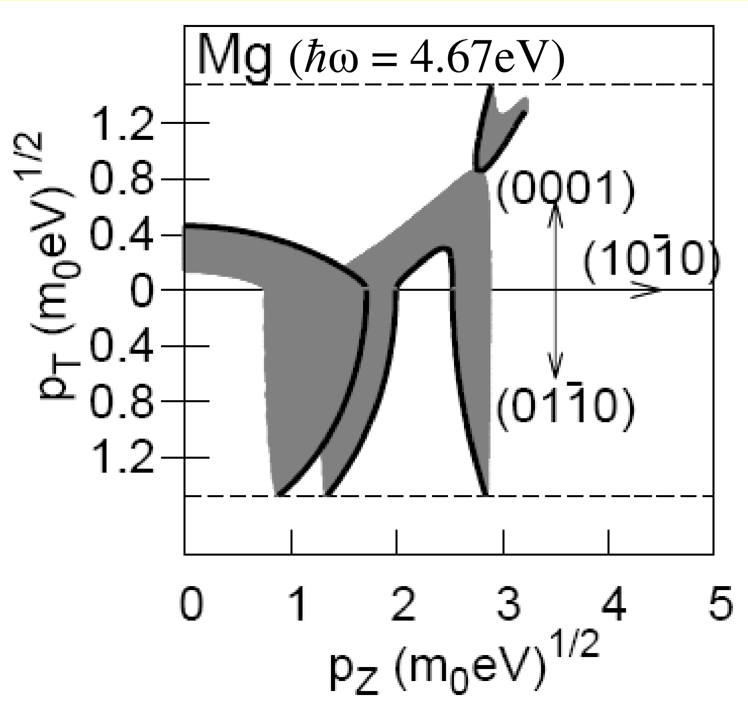
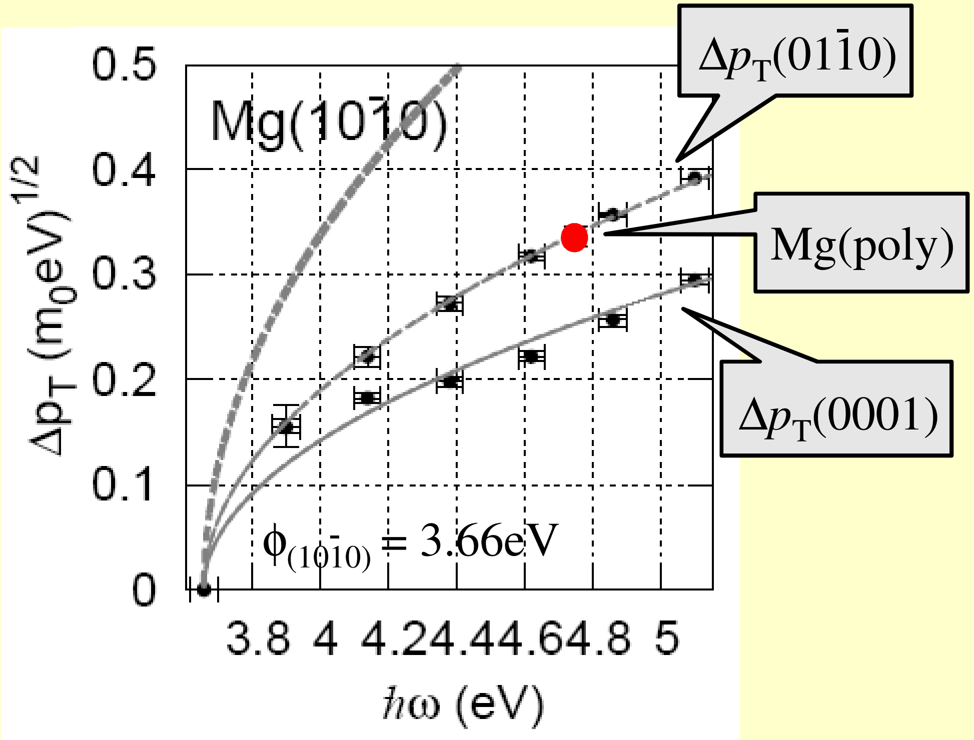
Calculating Δp_T(ijk) for ***all*** elemental metals

– 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.