Monte Carlo Scattering Corrections to Moments-Based Emission Models

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OUTLINE

Demands on electron sources for high performance FELs include high currents (kA/cm^2 peak, A/cm^2 average) and low emittance.

- Implies constraints on E distribution, temporal characteristics of bunches, Quantum Efficiency (for photoemission)

Monte Carlo based models developed to augment emission modules for the beam simulation particle-in-cell (PIC) code MICHELLE.

- Transport of charge through semiconductor materials followed by emission into vacuum - Augments Moments-based models of QE (photocathodes)
- Scattering determines how bunches evolve under band bending, temporal characteristics, and phase space distribution
- Revised code: faster, 10x more electrons than previous, more accurate

Show how Moments is augmented to include scattered electrons, and what changes are entailed by semiconductor photocathodes.

Examine temperature rise and impact

PHOTOEMISSION PHYSICS

A Moments-Based Method is used to treat Metals & Semiconductor Photocathodes

Simple DOS

$$M_n = (2\pi)^{-3}\left(\frac{2m}{\hbar^2}\right)^{3/2}\int_0^\infty E^{1/2}\,dE\int_0^{\pi/2}\sin\theta\,d\theta\left\{\frac{2m}{\hbar^2}(E+\hbar\omega)\sin^2\theta\right\}^{n/2} D\left\{(E+\hbar\omega)\cos^2\theta\right\} f_\lambda\left[\cos\theta, p(\hbar\omega)\right]\begin{cases} f_{FD}(E)\left(1-f_{FD}(E+\hbar\omega)\right) \\ \Theta\left(\hbar\omega+E-E_g\right)\end{cases}$$

Averages of powers of momentum k are known as "Moments of the distribution function" = $\int k^n f(x,k)\,dk$

Transverse momentum uses sin; J calc would use cos for moment

This is transmission probability for surface barrier

This is scattering loss factor for bulk transport

This is initial & final state occupation factor

Quantum Efficiency: Ratio of Current (k_z^1) Moments

$$QE = \left\{1-R(\omega)\right\}\frac{M_1(k_z)}{2M_1(k_z)\big|_{D=1,f_\lambda=1}} \propto \begin{cases} (\hbar\omega-\phi)^2 & metal \\ (\hbar\omega-E_a-E_g)^v & semi\end{cases}$$

This is absorption

Metals p(E) large & $f_\lambda \approx \cos\theta/p$: therefore, emittance indep. of p.

Semiconductors larger ε due to p small, but D also has impact

Emittance: Ratio of Transverse Energy (k_ρ^2) Moments

$$\varepsilon_{n,rms} = \frac{\hbar}{mc}\sqrt{\left\langle x^2\right\rangle\left\langle k_x^2\right\rangle - \left\langle xk_x\right\rangle^2}$$

$$\Rightarrow \frac{\hbar}{mc}\left(\frac{\rho_c}{2}\right)\sqrt{\frac{M_2(k_\perp)}{2M_0(k_\rho)}} \approx \frac{\rho_c}{2}\left[\frac{(\hbar\omega-\phi)}{3mc^2}\right]^{1/2}$$

$$k_\perp^2 = k_\rho^2 + k_\omega^2$$

leading order (metal)

The "Fatal" Approximation

Commonly made approximation is electron scattering prevents emission

While good for metals w/ big barriers, it is bad when
(a) barriers are small or
(b) extended to semiconductors

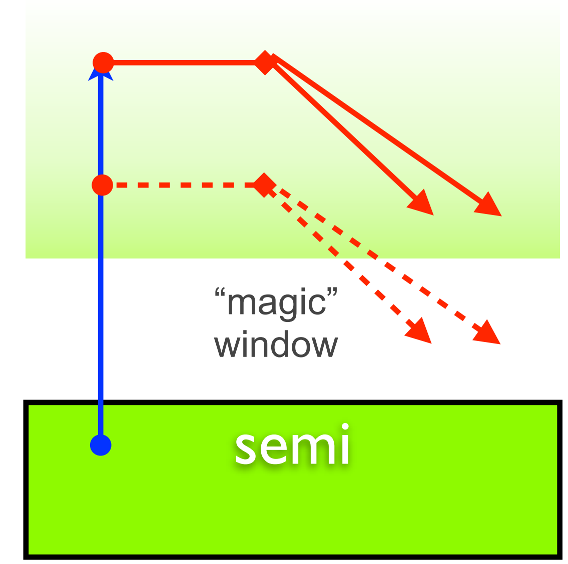
MOMENTS-BASED QE: SEMICONDUCTORS

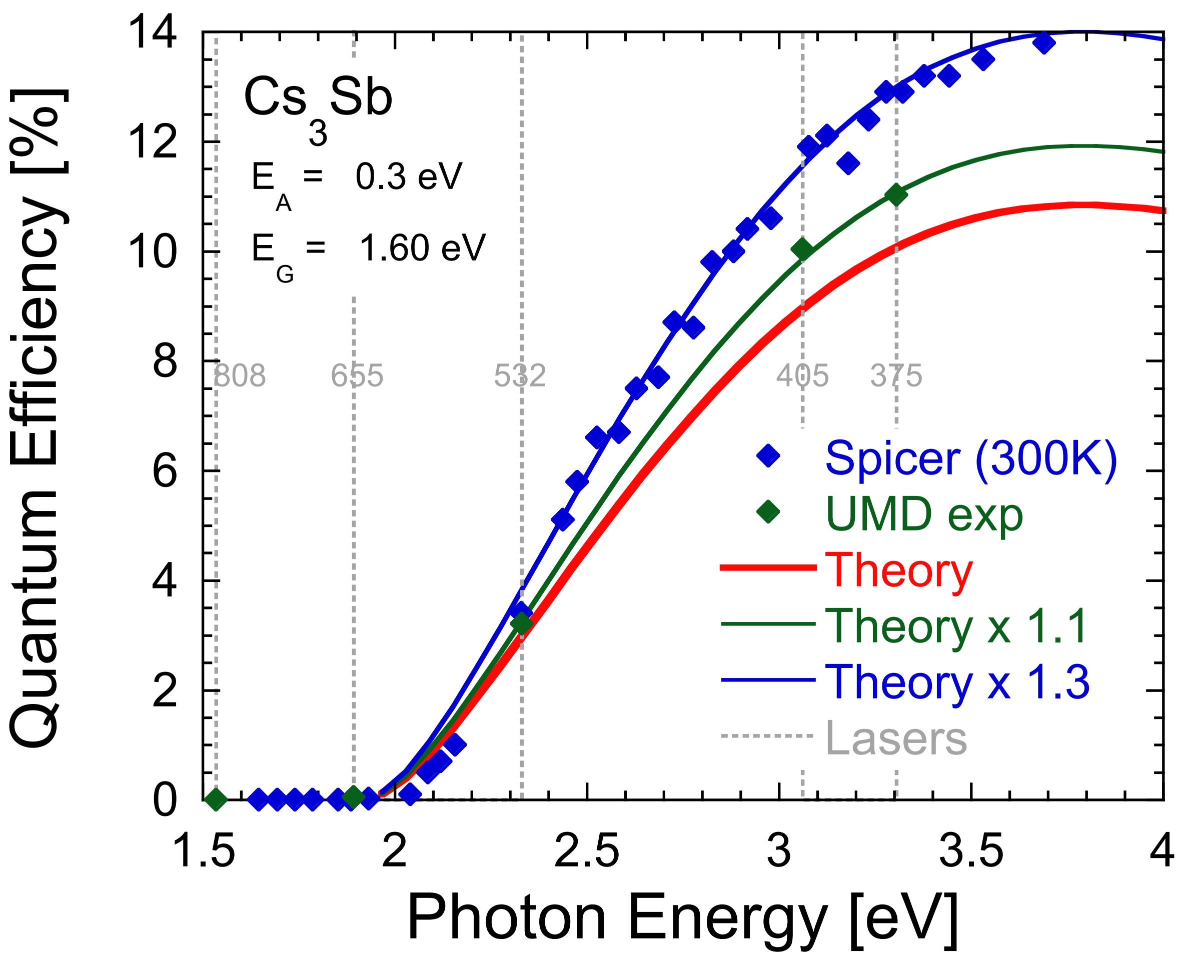
Leading Order Approximation with $\chi = \hbar\omega - \left(E_g + E_a\right)$

$$QE = \left(1 - R(\omega)\right) \frac{\int_{E_a}^{\hbar\omega - E_g} E\, dE \int_{\sqrt{E_a/E}}^{1} x\, dx\, D_\Delta\left[Ex^2\right] f_\lambda(x,E)}{2\int_0^{\hbar\omega - E_g} E\left[\int_0^1 x\, dx\right] dE}$$

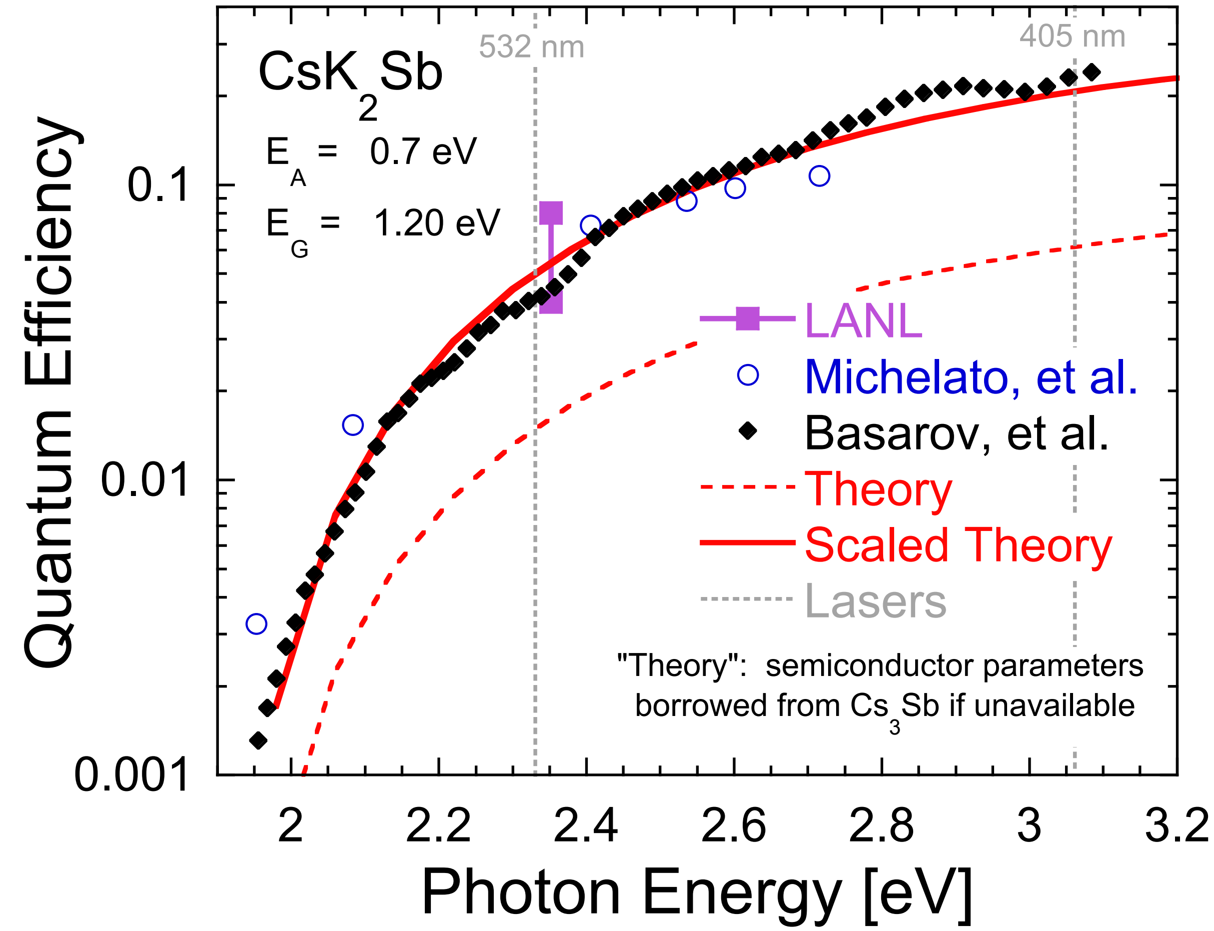
$$\lim_{\chi \to 0} QE \approx \frac{\left(1 - R(\omega)\right)}{2\left(p_o + 1\right)\left(1 + E_a/\chi\right)^2} \Leftrightarrow QE_{spicer} \approx \frac{B}{1 + g\chi^{-3/2}}$$

Semiconductors: e-e scattering not allowed unless final states unoccupied & in conduction band (creates "magic" window)





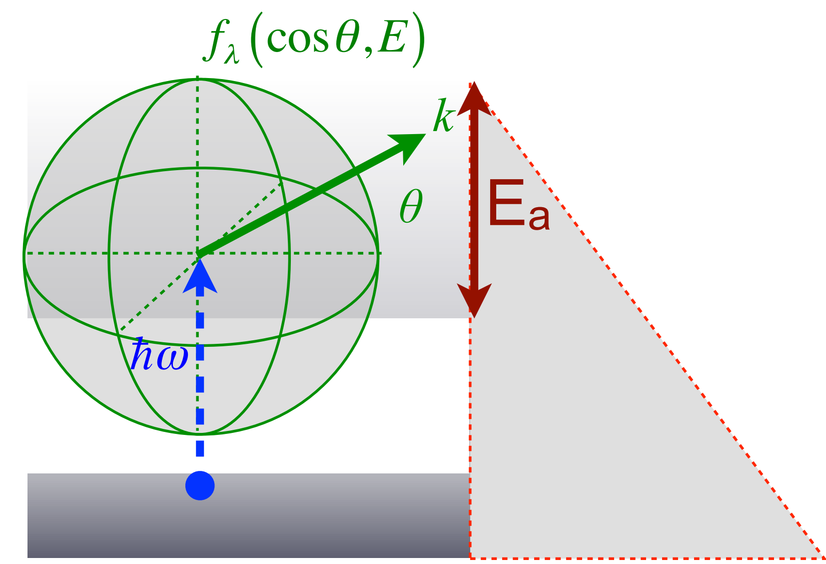
Data courtesy of I. Bazarov, Appl. Phys. Lett. 99, 152110 (2011)



MOMENTS-BASED SCATTERING FACTOR

In Polar Coordinates, Velocity of e- at angle θ to normal

Assume Any Scattering Event Is Fatal To Emission ("Fatal Approximation")



Ratio of penetration depth to distance between scattering events

$$p(E) = \frac{\delta(\hbar\omega)}{l(E)} = \frac{m\delta(\hbar\omega)}{\hbar k(E)\tau(E)}$$

Matthiessen's Rule:

$$\tau_{total}^{-1} = \sum_j \tau_j^{-1}$$

Fraction Of Photoexcited Electrons Surviving Transport Back To Surface

$$f_\lambda(\cos\theta, p) = \frac{\int_0^\infty \exp\left(-\frac{x}{\delta} - \frac{x}{l(E)\cos\theta}\right)dx}{\int_0^\infty \exp\left(-\frac{x}{\delta}\right)dx} = \frac{\cos\theta}{\cos\theta + p(y)}$$

Example: Cs_3Sb-like
- δ = 27 nm
- v/c = 0.8%
- τ = 31 fs
- ➡ p ≈ 0.36

Example: Cu-like
- δ = 12.6 nm
- v/c = 0.675%
- τ = 2.6 fs
- ➡ p ≈ 2.38

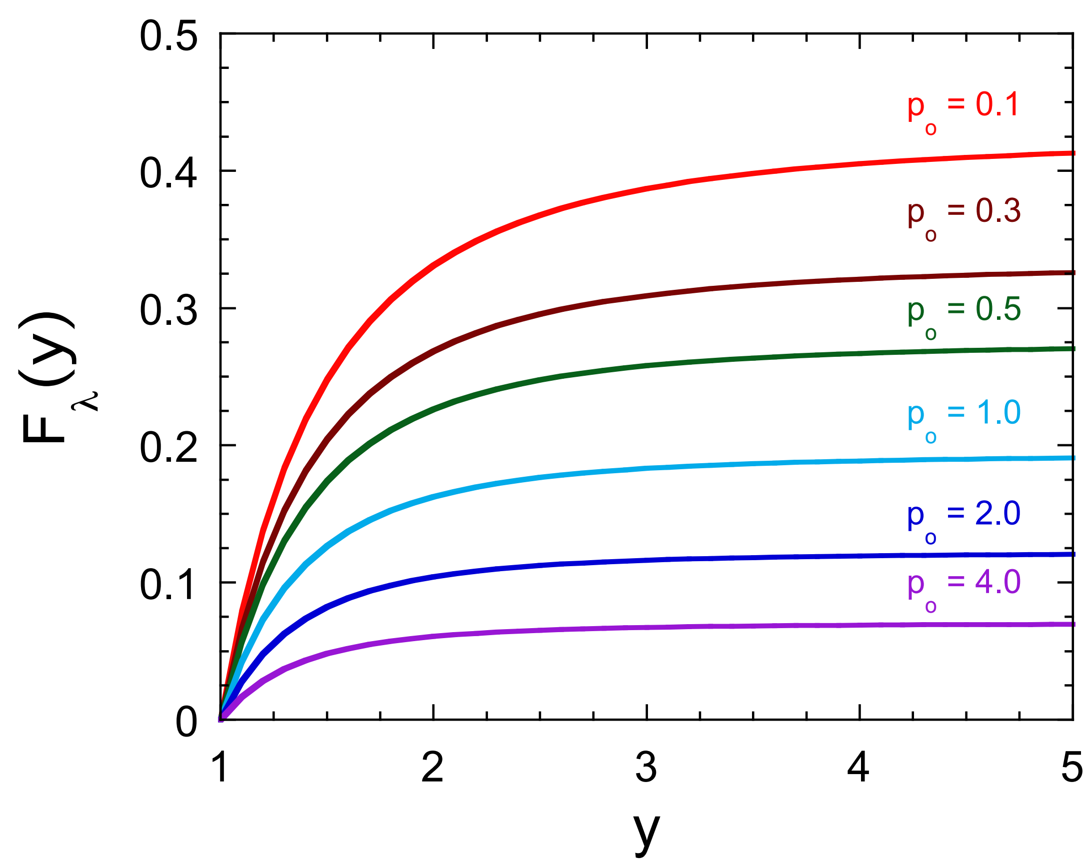
Scattering Fraction As Used In Modified Fowler Dubridge Eq

(1/y Acts As Cosine Of **Escape Cone Angle**)

$$F_\lambda(y) = \int_{1/y}^{1} x f_\lambda(x,p)dx$$

$$= p^2 \ln\left[\frac{y(1+p)}{1+yp}\right] + \frac{1}{2y^2}(1-y)(2yp - y - 1)$$

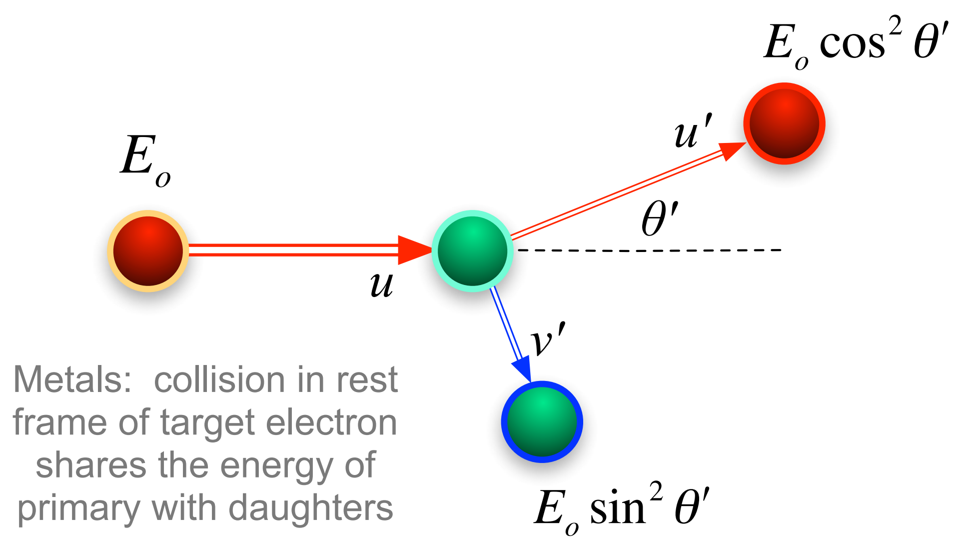
- for semiconductors, measure E w.r.t. E_a $\quad E \equiv E_a y^2$
- IF τ scales as 1/k, then p is constant $\quad p \approx p_o$



MONTE CARLO AND TRANSPORT

Calculation of Emittance (and QE) requires distribution function f(x,k). MC provides it.

- NAIVE EXPECTATIONS:
- In Metals: e-e collisions syphons energy off primary (target electron at Fermi level) quickly so Fatal approximation expected to be good

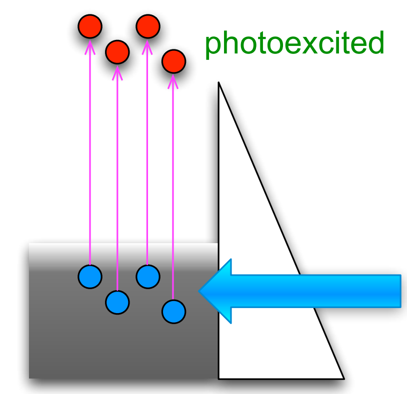


- If barrier small, sharing energy may not kill emission
- In Semiconductors: e-ph take fraction of eV from primary, so Fatal Approximation is suspect

Photoemission (Photocathodes)

electrons start with energy > barrier height

$$E = \mu + \phi + r(\hbar\omega - \phi)$$

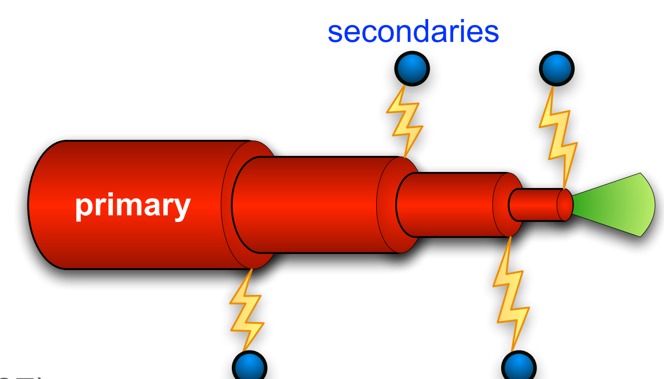


- spaced according to exponential decay of Photon number with depth into metal
  (Optical penetration depth)
- Primary energy loss mechanism: Electron-Electron scattering

Secondary Emission (Diamond Amplifier)

(Jensen et al,, JAP108, 044509 (2010)):

primary with energy E_o
generates N_p Secondaries



- $N_p = E_o/\Delta E$
- velocity perpendicular to incident
  K. Murata, D. Kyser
  Adv. Elect. El. Phys 69, 175 (1987)
- spaced as $z_{j+1} - z_j \approx \Delta E (\partial_z E)^{-1}\big|_{z=z_j}$
- all N_p secondaries appear to be generated simultaneously to a good approximation
- Primary energy loss mechanism: Optical Phonon

SCATTERING CONSIDERATIONS

Electron-Electron Scattering Energy & Direction

Lab Frame

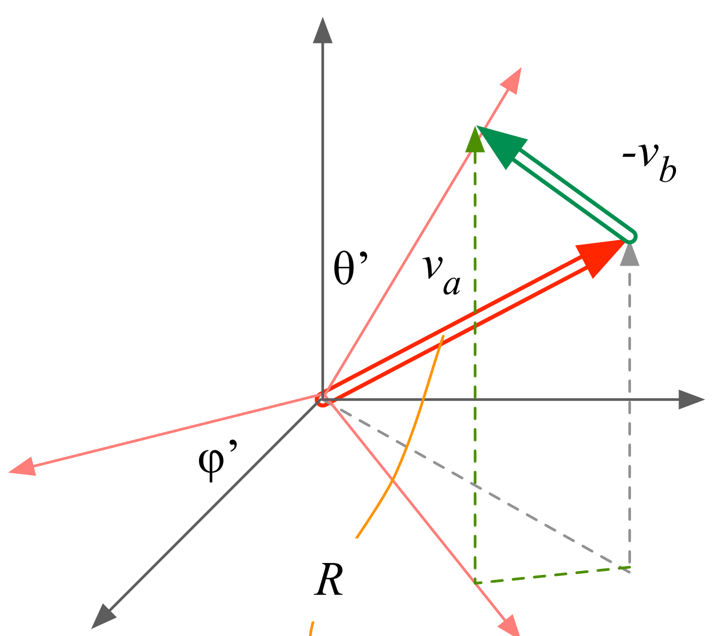
$$E_a + E_b = E_a' + E_b' \quad \text{conservation of Energy}$$

$$\vec{v}_a + \vec{v}_b = \vec{v}_a' + \vec{v}_a' \quad \text{conservation of momentum}$$

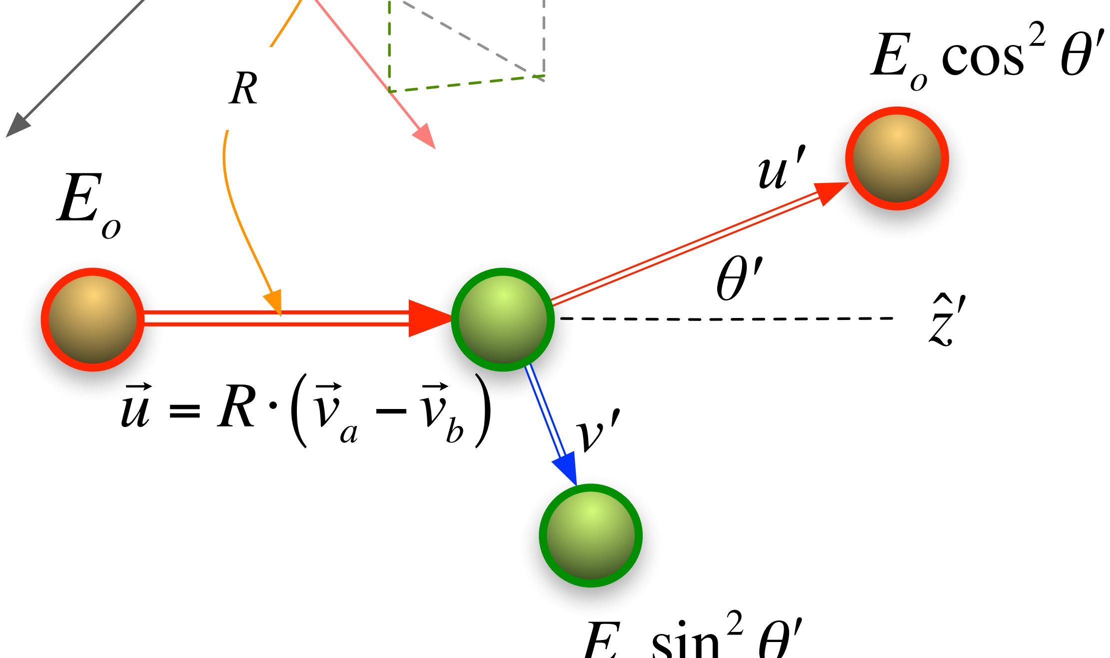
B electron frame

$$\vec{u} = \vec{u}' + \vec{v}' \Rightarrow \begin{cases} \vec{u}' = u_o \cos\theta\left(\hat{x}'\sin\theta + \hat{z}'\cos\theta\right) \\ \vec{v}' = u_o \sin\theta\left(-\hat{x}'\cos\theta + \hat{z}'\sin\theta\right) \end{cases}$$

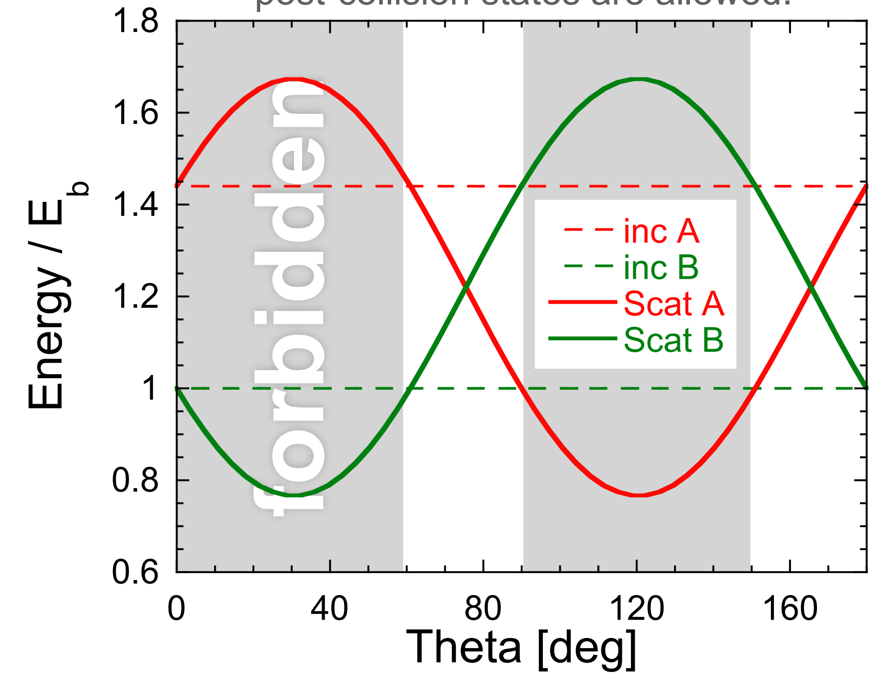
Random Angles: azimuthal φ (R commutes with R(φ))
polar θ (subject to final state constraints)



Incident (photoexcited) electron
Target (Fermi level) electron
Move to rest frame of Target
Rotate so v_a-v_b is along z axis



B electron at Fermi level: final products > Fermi level, therefore, only some θ post-collision states are allowed.



Dominant Scattering Rates (metals)

Electron-Electron

q_o is screening factor

$$\tau_{ee} = \frac{4\hbar K_s^2}{\alpha^2 \pi m c^2 (k_B T)^2}\left[\left(1+\frac{\Delta E}{\pi k_B T}\right)\gamma\left(\frac{2k_F}{q_o}\right)\right]^{-1}$$

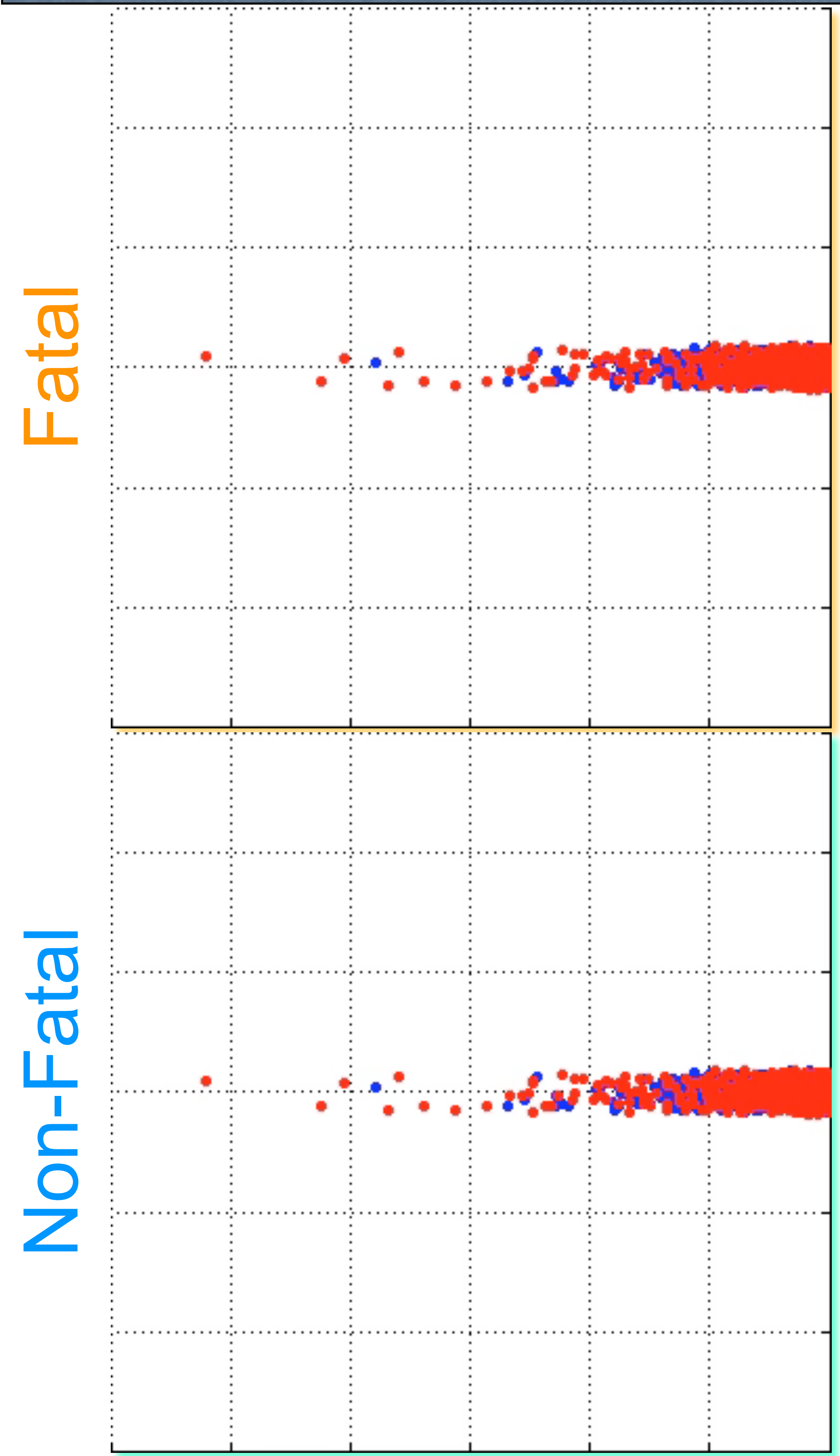
$$\gamma(x) = \frac{x^3}{4}\left(\tan^{-1}x + \frac{x}{1+x^2} - \frac{\tan^{-1}\left(x\sqrt{2+x^2}\right)}{\sqrt{2+x^2}}\right)$$

Acoustic (metals&semi)

$$\tau_{ac} \approx \frac{\pi\hbar^3 \rho v_s^2}{4m\Xi^2 k(E)(k_B T)}\left\{\left(\frac{T}{\Theta}\right)^4 W_-\left(5,\frac{\Theta}{T}\right)\right\}^{-1}$$

W_: Bloch-Grünnisen Function
Collision randomly changes direction, removes phonon energy $k_B\theta_D$ from electron

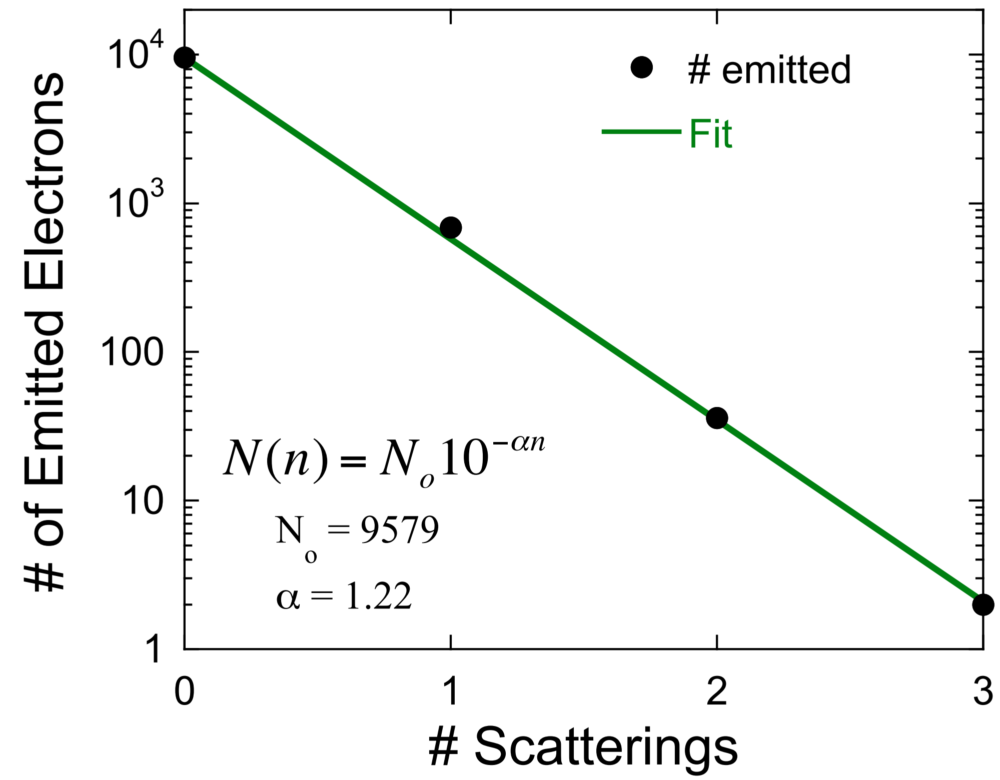
BARE COPPER PHOTOEMISSION @ 266 NM

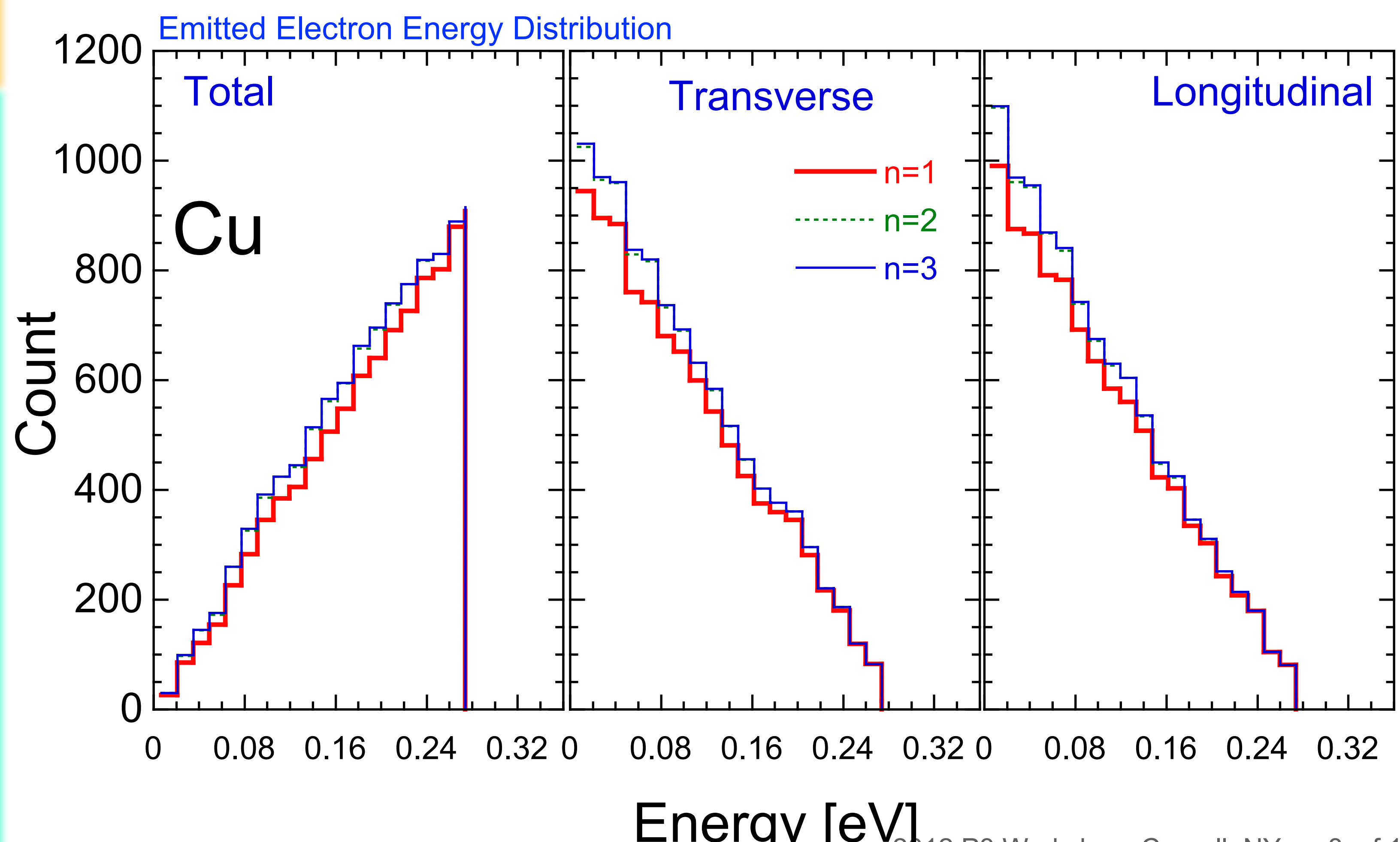


each time step 0.4 fs
40 frames = 16 fs
Width = 120 nm
# initial = 10,000,000

Fatal Approximation works well for metals with large work function

Approximately 90% of electrons removed per scattering event

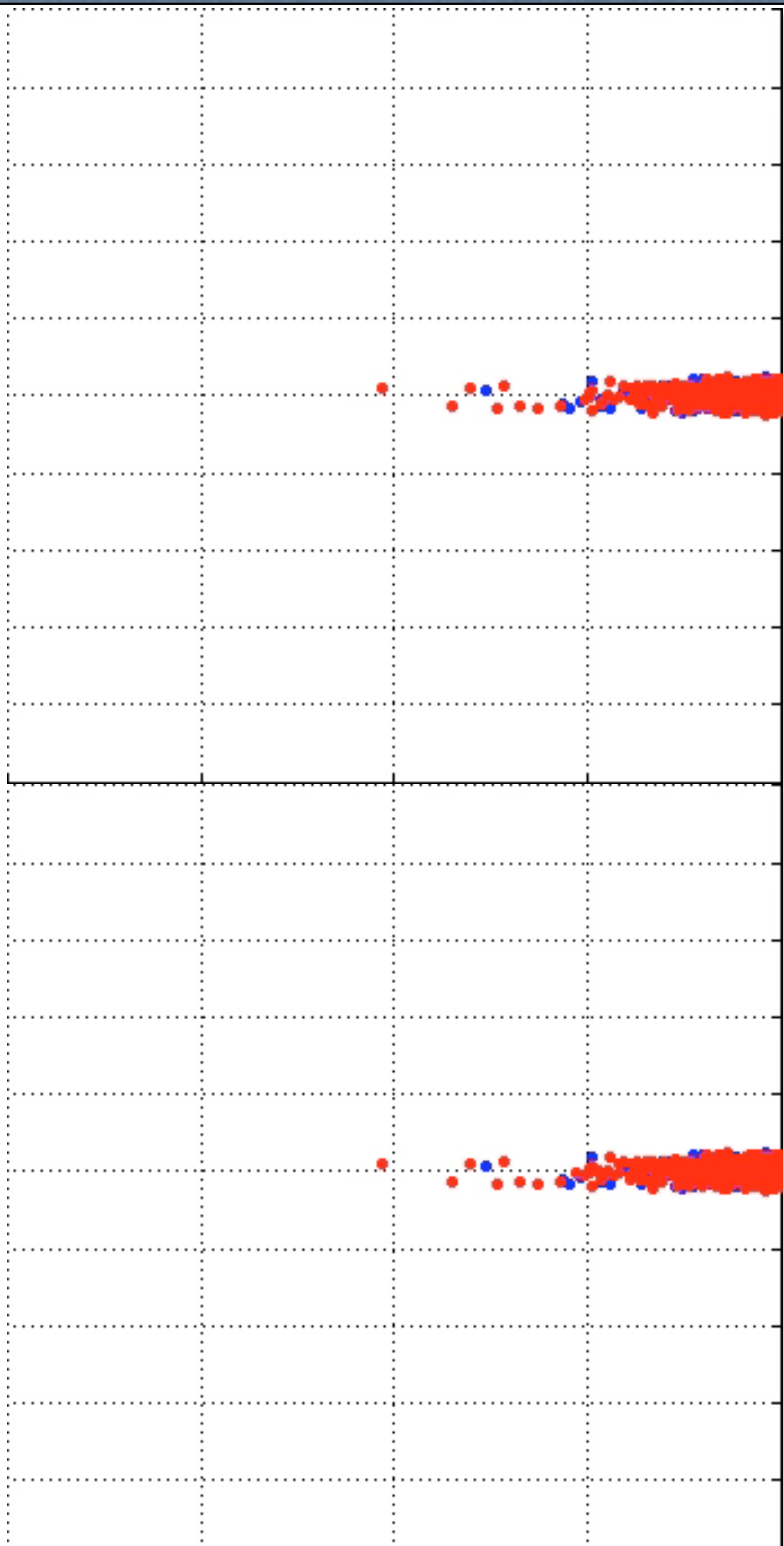




CESIATED COPPER PHOTOEMISSION @ 266 NM

Fatal

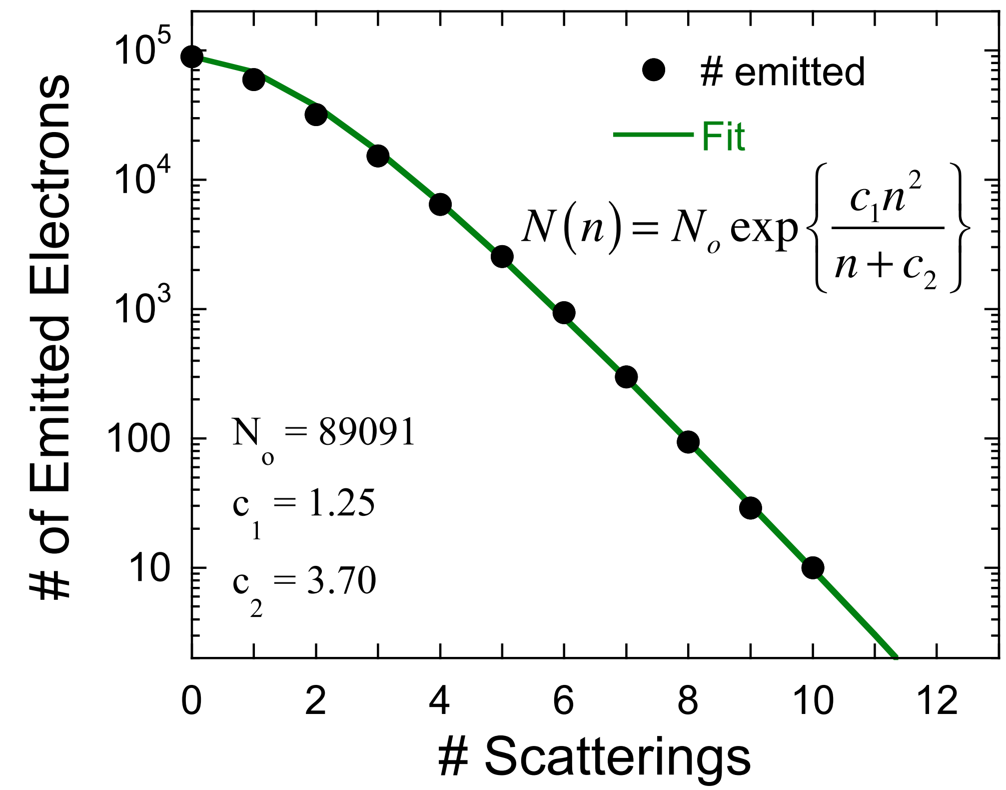
Non-Fatal

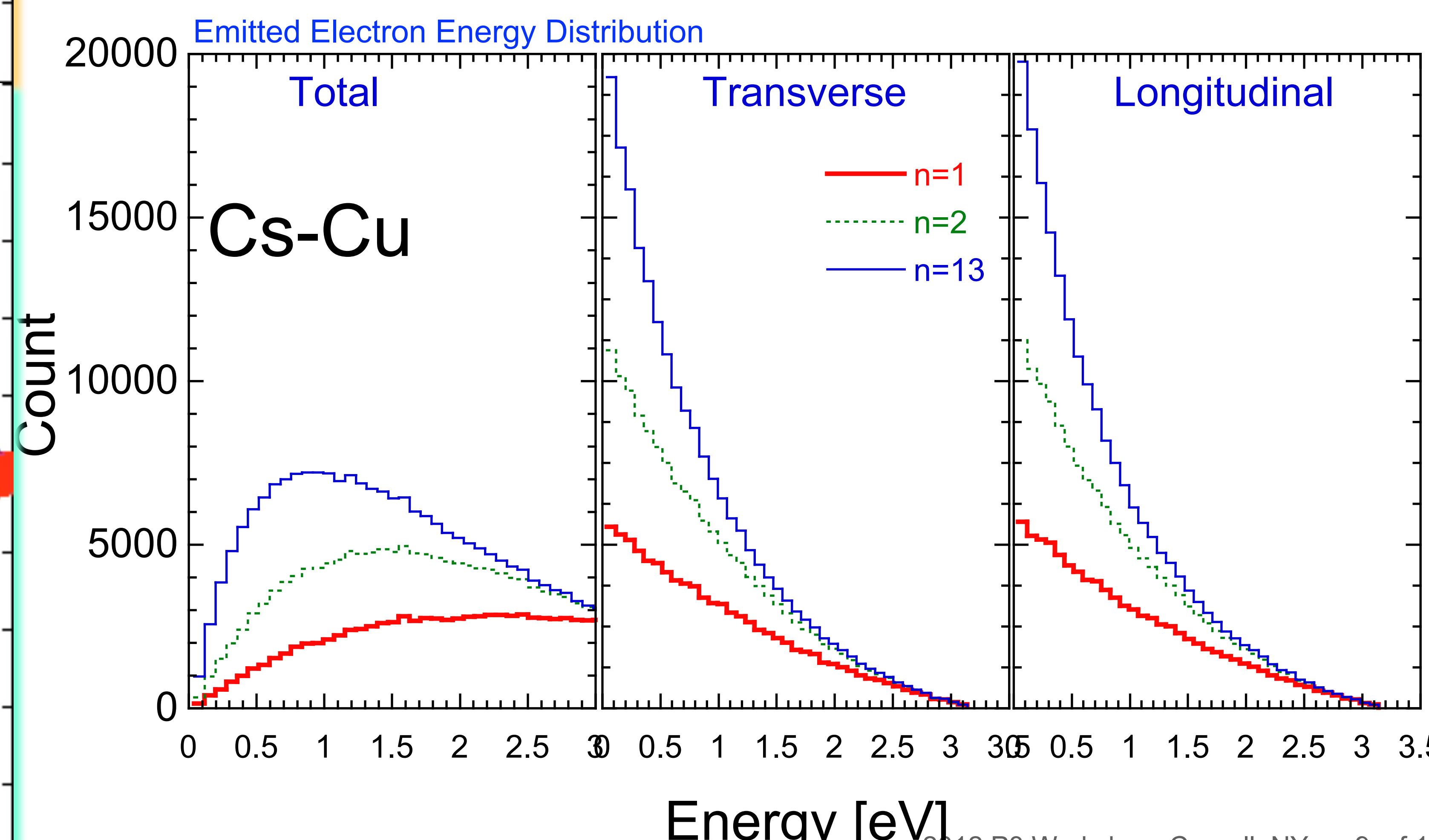


each time step 1.0 fs
60 frames = 60 fs
Width = 200 nm
# Initial = 5,000,000

Fatal Approximation works well for metals with large work function

Approximately 90% of electrons removed after 3 scattering events





MC FIX TO QE MOMENTS - METALS

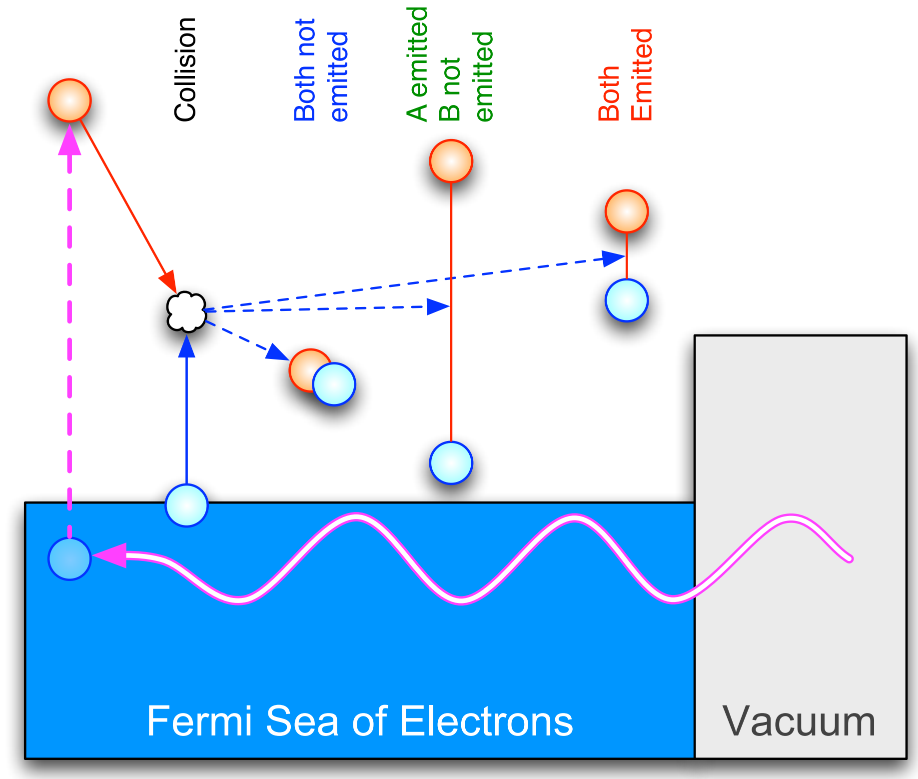
MONTE CARLO modifications to Moments-Based Photoemission depends on size of emission barrier

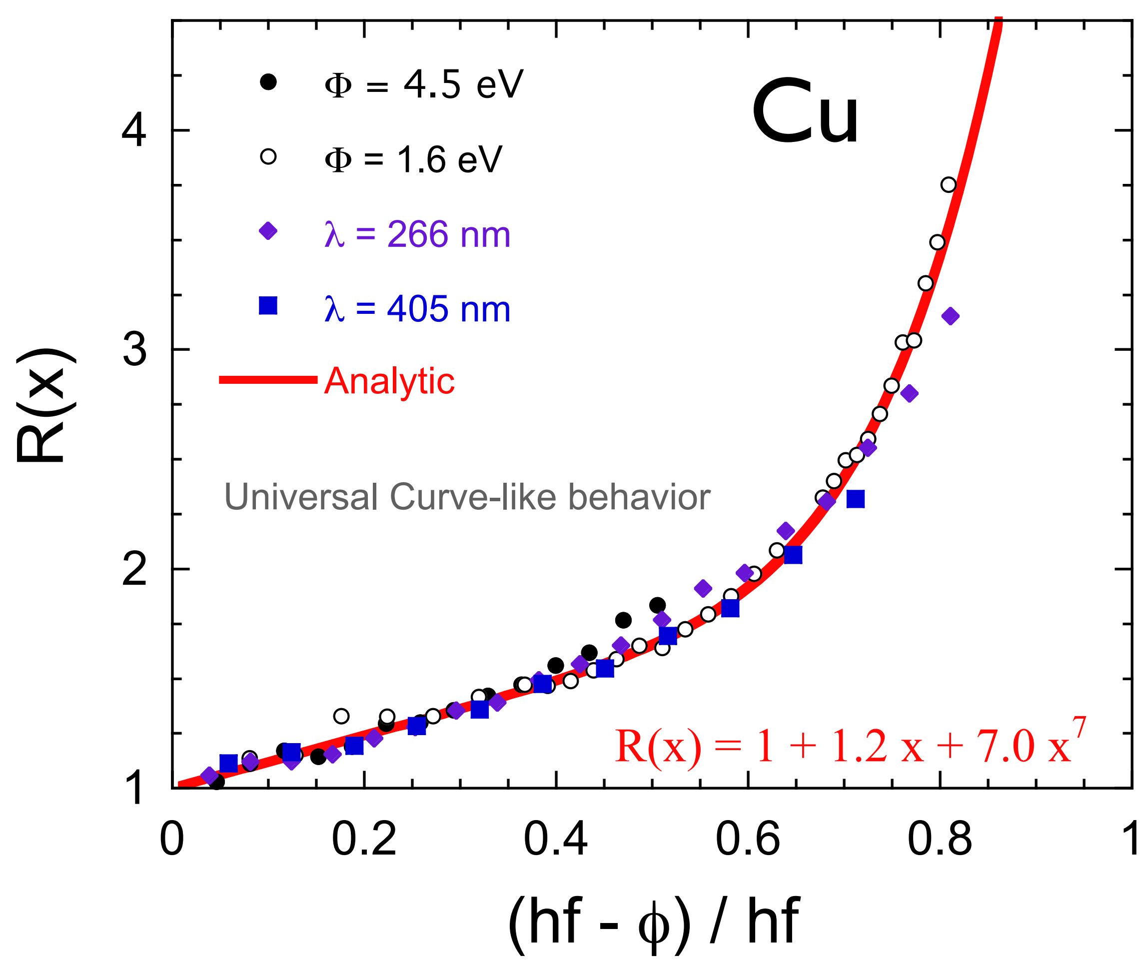
After e-e scattering, primary e shares energy with target e. If...

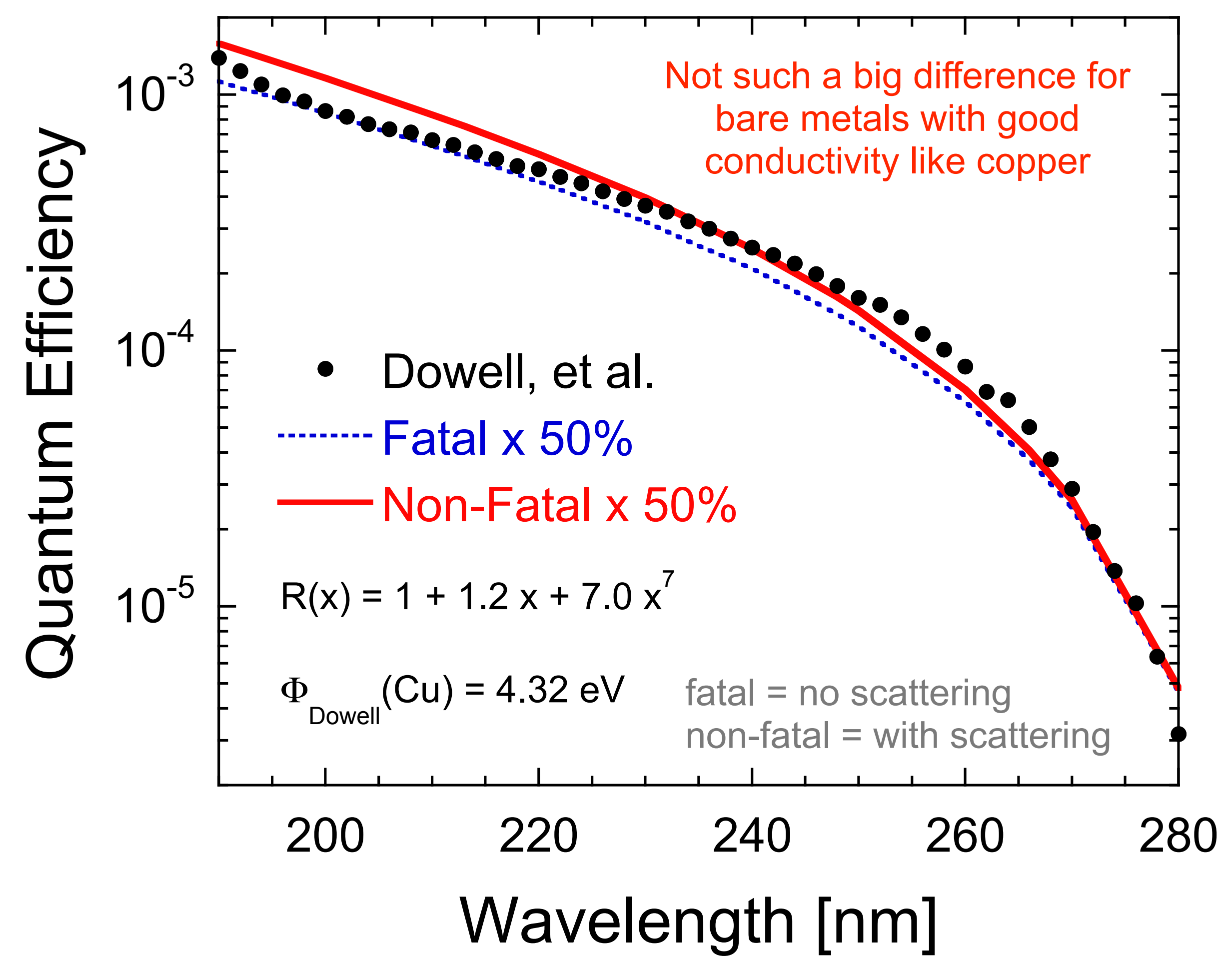
- hf ≈ barrier: shared energy means neither electron emitted
- hf ≈ 1.5 x barrier: more energetic e- may still emit
- hf > 2 x barrier: both electrons can be emitted

To Fix Moments QE:

- Define R = ratio of all e- emitted / all unscattered e- emitted
- Multiply Moments QE by R(x) to estimate QE where
  x = (hf - φ)/hf = ratio of energy above barrier to photon energy







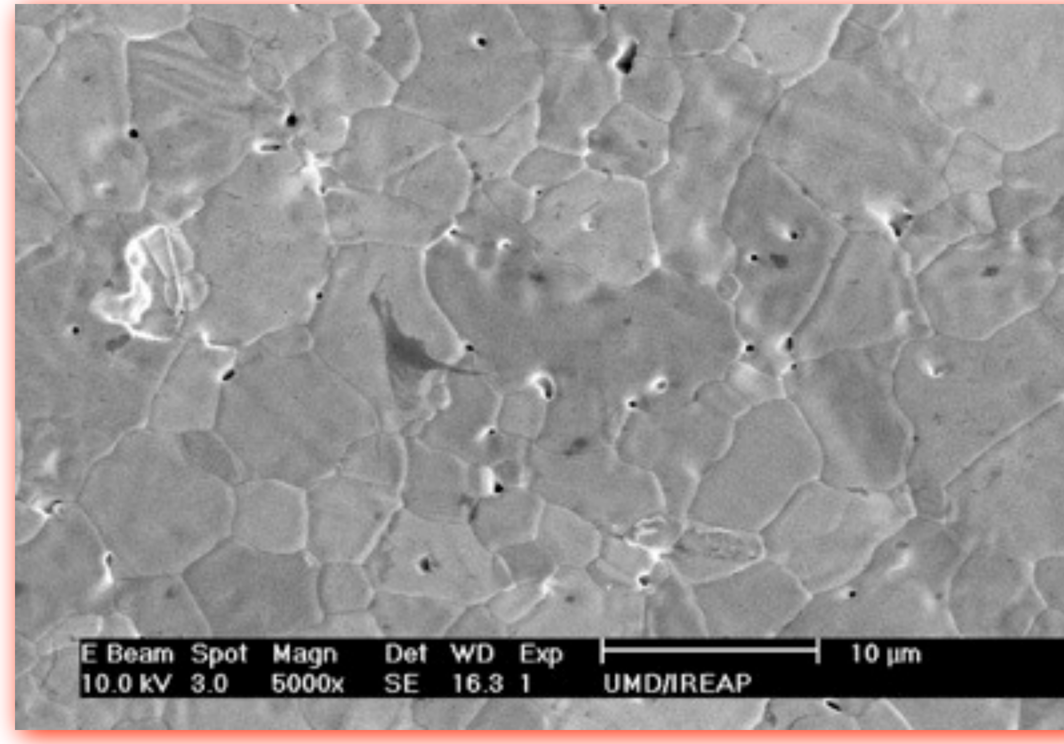
MC FIX TO QE MOMENTS - CESIATED METALS

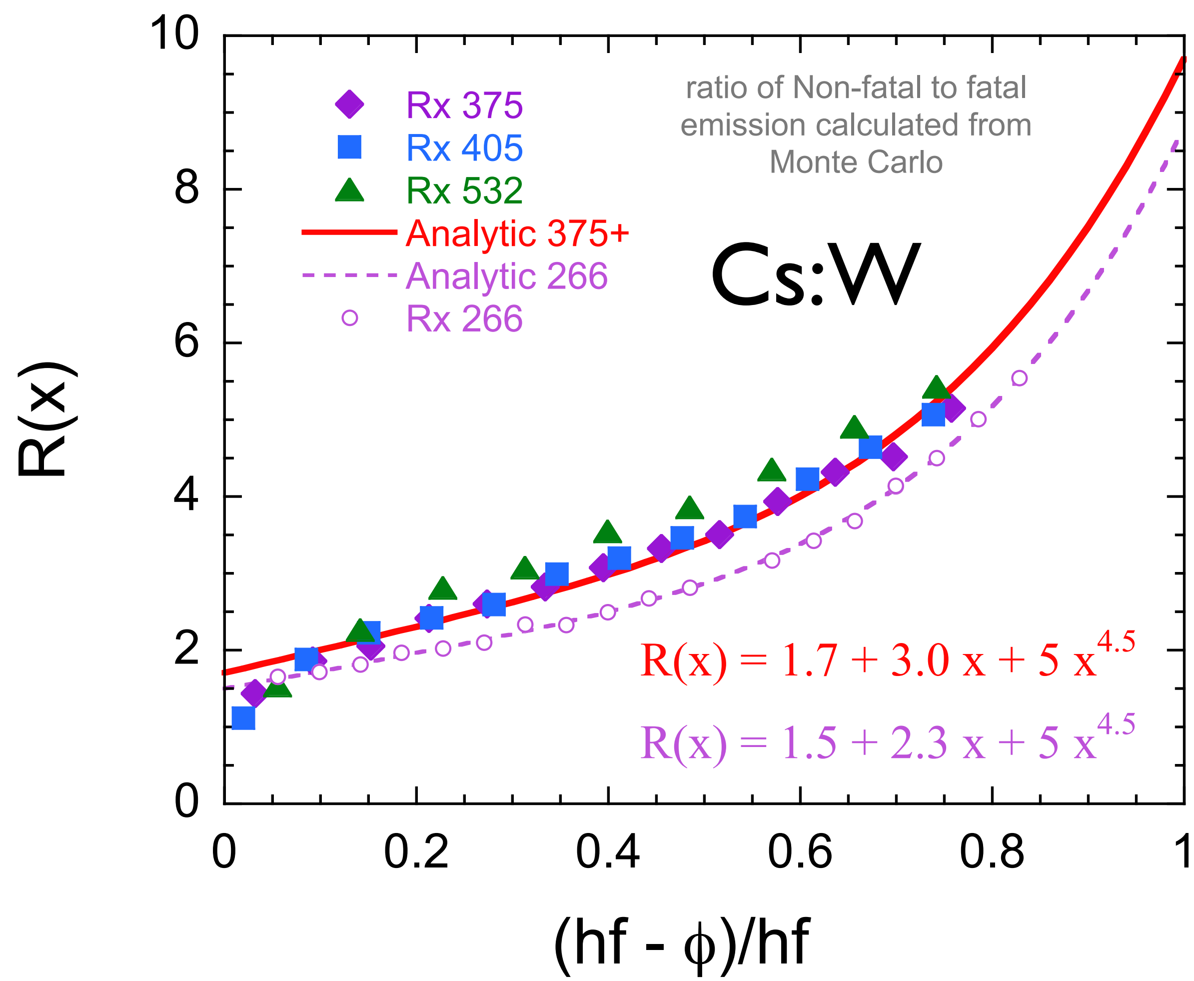
Comparison to Cs-W Surfaces: UMD Dispenser Photocathode

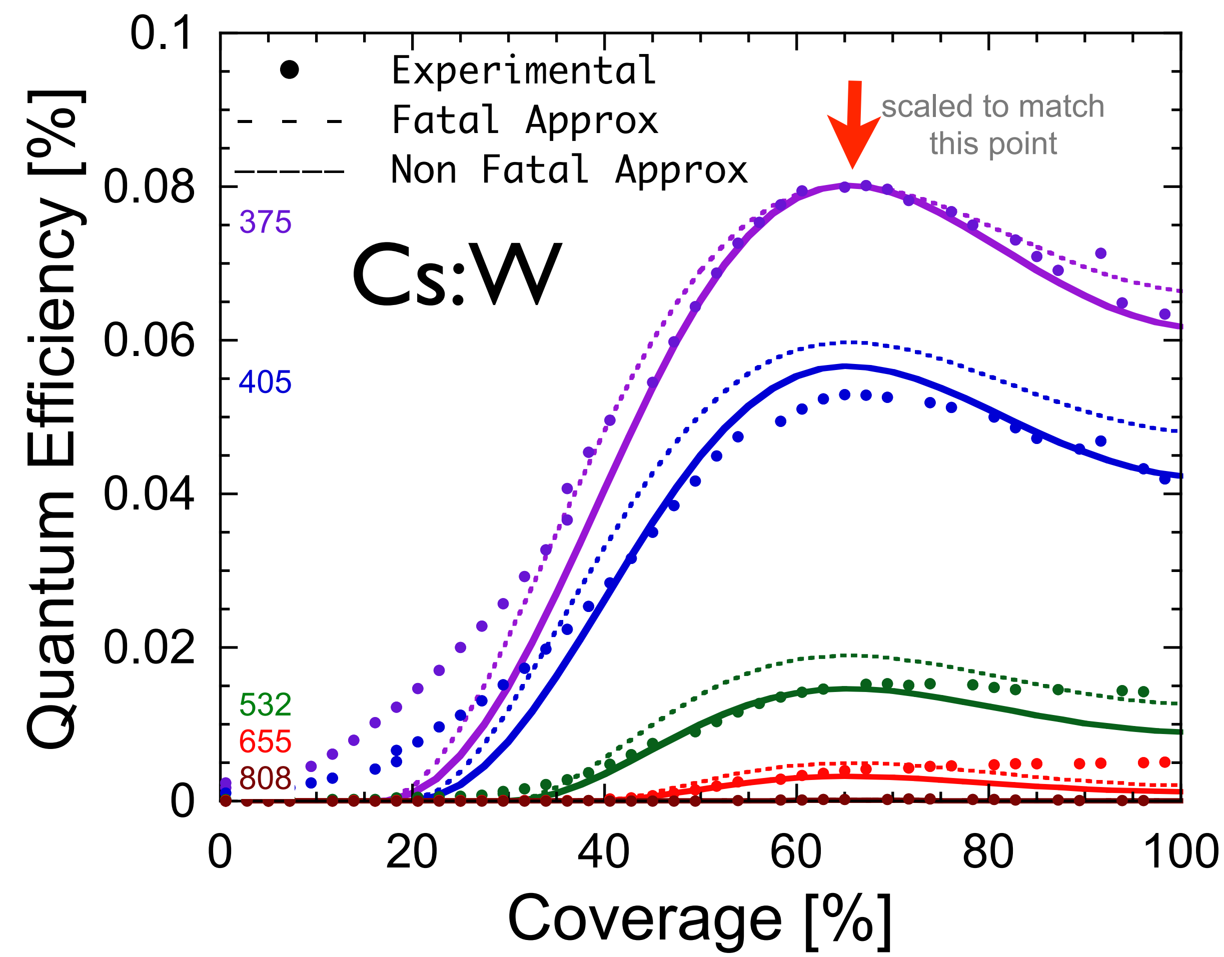
- Contamination present, Multiple crystal faces exposed, and variation in crystal face work function expected
  Photoemission Φ (from Samsonov, "Handbook of Thermionic Properties")
  {011} = 5.85 eV, {111} = 4.39 eV, {110} = 4.56 eV*
- Acoustic scattering more important in W than in Cu

QE(Exp) = (scaling factor) x QE(Theory)

- Moments-based w/o MC: Scaling factor = 0.922
- Moments-based w/ MC: Scaling factor ≈ 0.246
  - accounts for crystal faces
  - Better correspondence to QE peak variation with λ
  - Better overall shape of QE(θ) near respective peaks







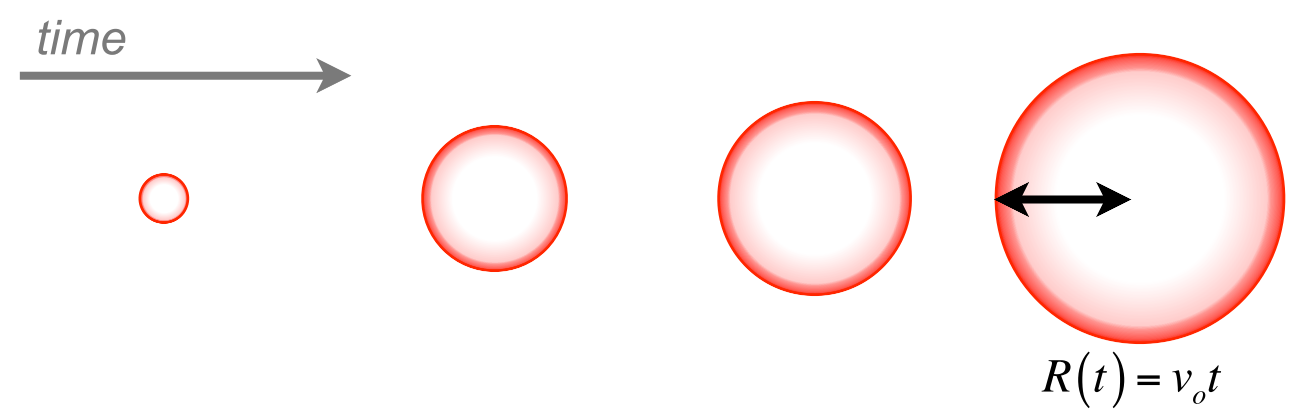
* {110} value is for thermionic emission

“SHELL” VS “SPHERE” EMISSION PROCESSES

- Electrons that do no scatter before emission travel ballistically as expanding SHELL
- Electrons that scatter before emission travel as a diffusively expanding SPHERE
- Scattered electrons change time dependence by adding long time tail
- Simple Current model:
  passage of shell or sphere through surface boundary as a function of time

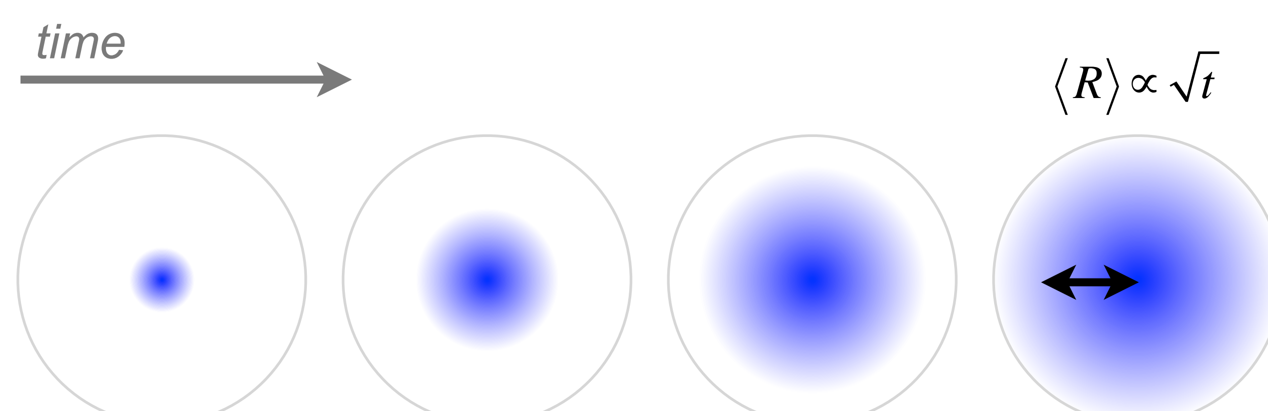
Expanding SHELL

$$Q(t) = \frac{Q_r}{4\pi R(t)^2}\int_0^{2\pi} R\,d\varphi \int_0^{\cos^{-1}(R(0)/R(t))} R\sin\theta\,d\theta = \frac{Q_r}{2}\frac{t}{(t+t_o)}$$



Diffusive SPHERE

$$Q(t) = Q_d \frac{\int_{v_d t}^{\infty} t^{-1/2}\exp\left(-z^2/4Dt\right)dz}{\int_{-\infty}^{\infty} t^{-1/2}\exp\left(-z^2/4Dt\right)dz} = \frac{1}{2}Q_d\left\{1 - Erf\left(\sqrt{\frac{\tau^*}{t}}\right)\right\}$$

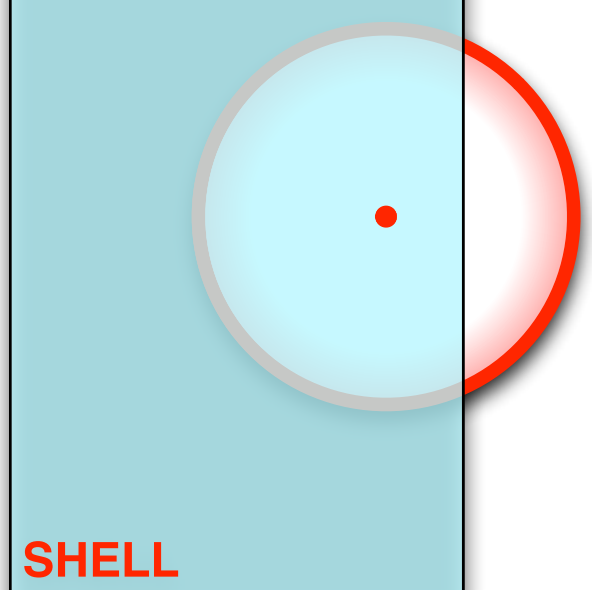


"SHELL" VS "SPHERE" EMISSION CURRENT

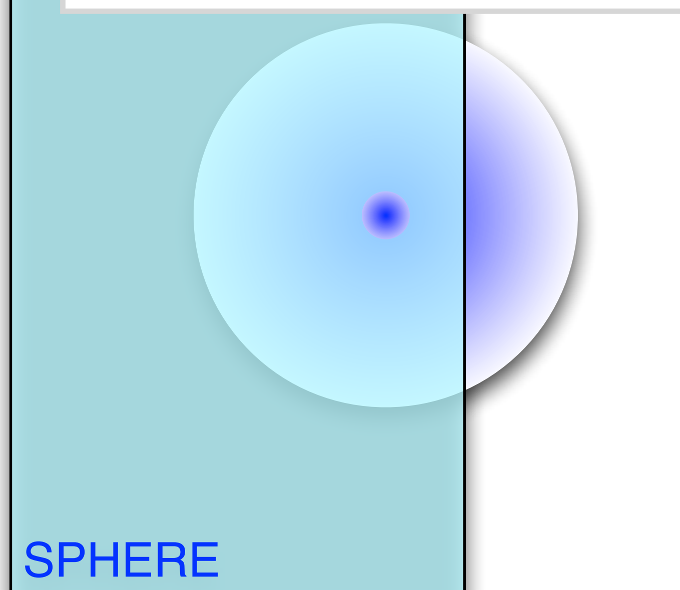
Charge to pass through surface is sum of Shell & Sphere Processes: I(t) = dQ/dt

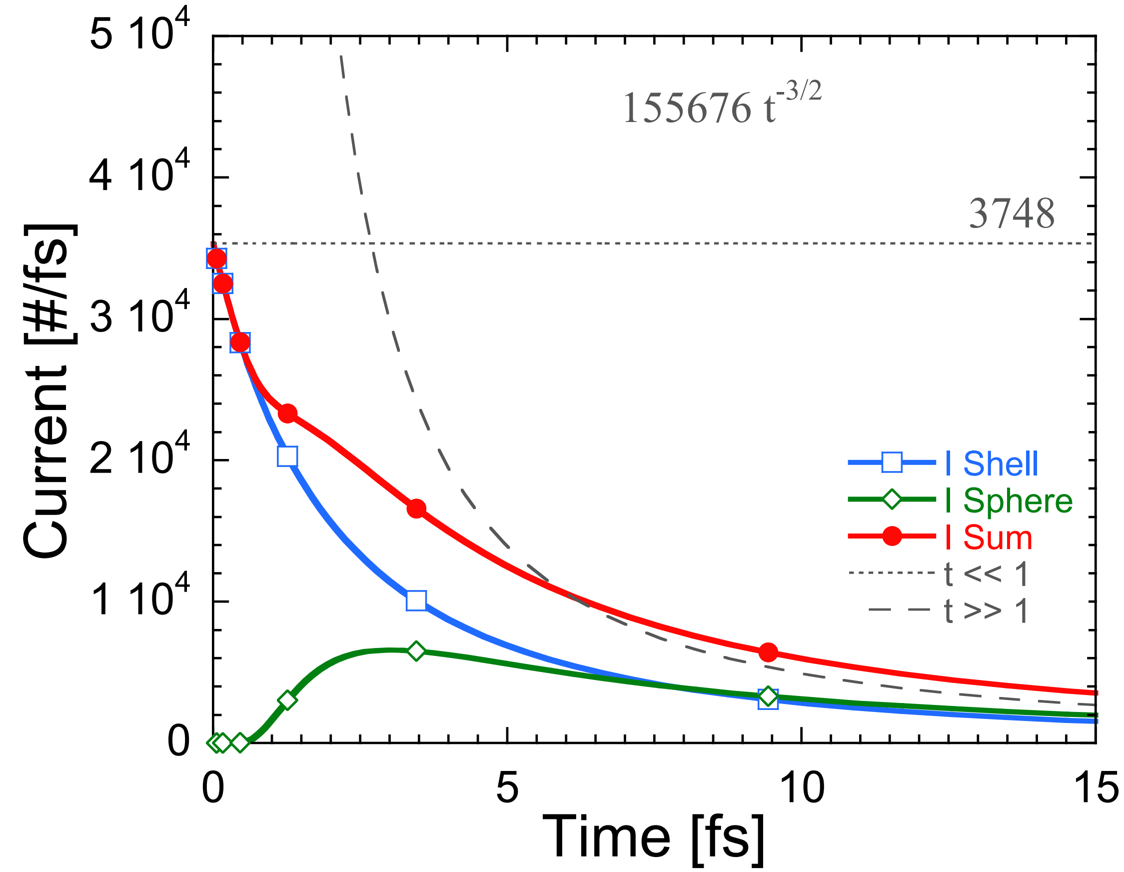
$$I(t) = Q_s \frac{t_o}{2(t+t_o)^2} + Q_d \left(\frac{\tau^*}{4\pi t^3}\right)^{1/2} \exp\left(-\frac{\tau^*}{t}\right)$$

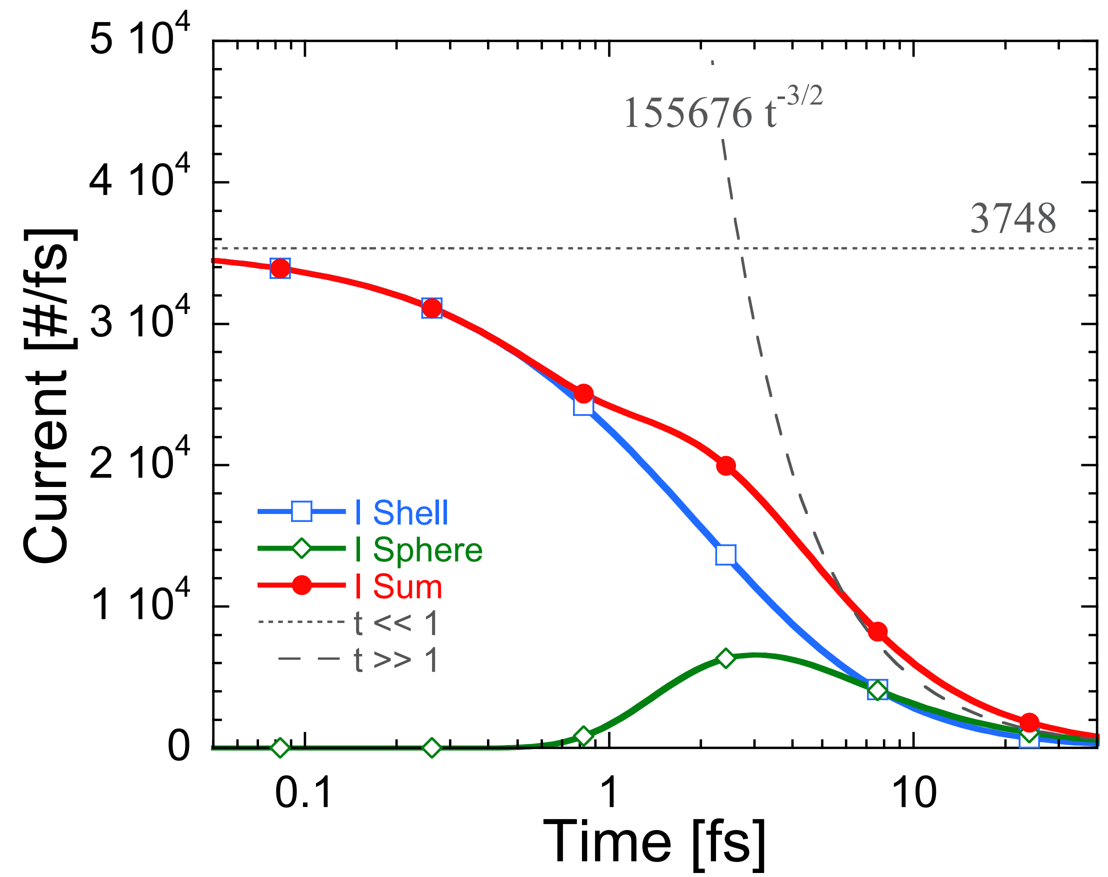
$$Q_{shell}(t) = \frac{Q_r}{2}\frac{t}{(t+t_o)}$$



$$Q_{sphere}(t) = \frac{1}{2} Q_d \left\{1 - Erf\left(\sqrt{\frac{\tau^*}{t}}\right)\right\}$$







EXTENSION TO SEMICONDUCTORS

Scattered electrons contribute more to QE

- Electrons photoexcited much further into semiconductors
- Rate of energy loss due to phonons ($\Delta E \approx$ phonon energy) far slower than e-e collisions ($\Delta E \approx (E-E_F)/2$)

Complication: electrons likely distributed in deposition so that shell model has range of times (t_a to t_b) rather than characteristic time (t_o) as for metals. Affects SHELL model

- EX: GaAs mobility = 8500 cm^2/Volt-sec
  Implies τ = 0.324 ps. Assume $t_a \approx \tau$.
  - Then B(0.42,1.01) = B(0.55,2) = B(1.27,10) ≈ 1/2 ←
  - Full width at half maximum (FWHM) of D(x) is 1.8
- **Long tails persist**: D(10) ≈ 7%, D(38) = 1%

CONCLUDE:

- Semiconductor physics results in longer characteristic times for both the shell and sphere components
- Time response & rise/fall ≈ multiples of scattering time
- Long diffusive time tails due to D(x) and associated with NEA photocathodes mitigated by small but finite positive electron affinities (PEAs) of multialkali antimonide photocathodes

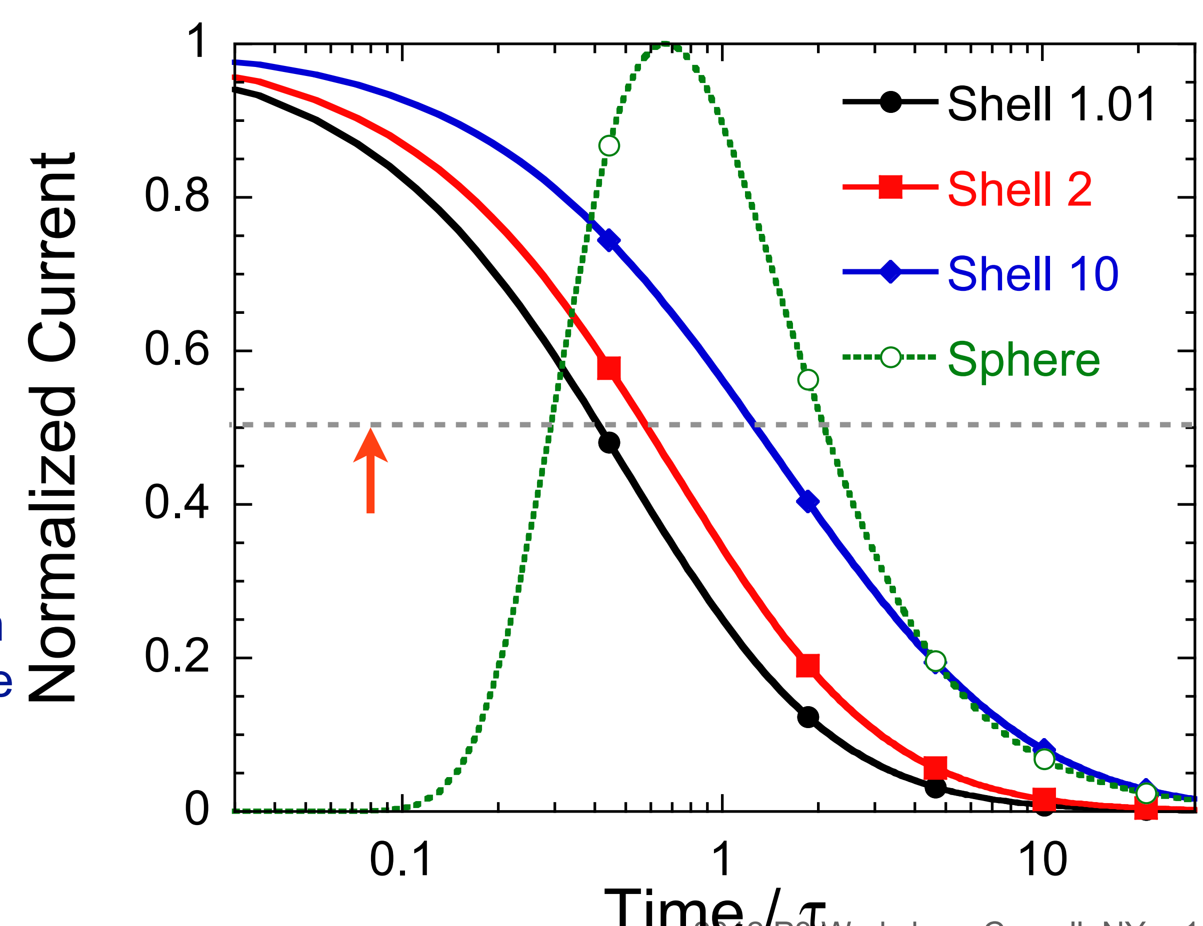
$$I_{shell}(t) = \frac{Q_{shell}}{2(t_b - t_a)} \int_{t_a}^{t_b} \frac{s}{(t+s)^2} ds$$

$$\Rightarrow I_p B\left(\frac{t}{t_a}, \frac{t_b}{t_a}\right)$$

$$I_{sphere}(t) \Rightarrow I_s D\left(\frac{t}{\tau}\right)$$

$$B(x,n) = \frac{1}{\ln(n)} \left\{ \ln\left(\frac{n+x}{1+x}\right) - \frac{x(n-1)}{(1+x)(n+x)} \right\}$$

$$D(x) = \left(\frac{2e}{3x}\right)^{3/2} e^{-1/x}$$



MODELING OF QE FOR SEMICONDUCTORS

Moments-Based Model of QE for **SEMICONDUCTORS**

- R = reflectivity
  D_Δ = Transmission Probability,
  f_λ = scattering factor

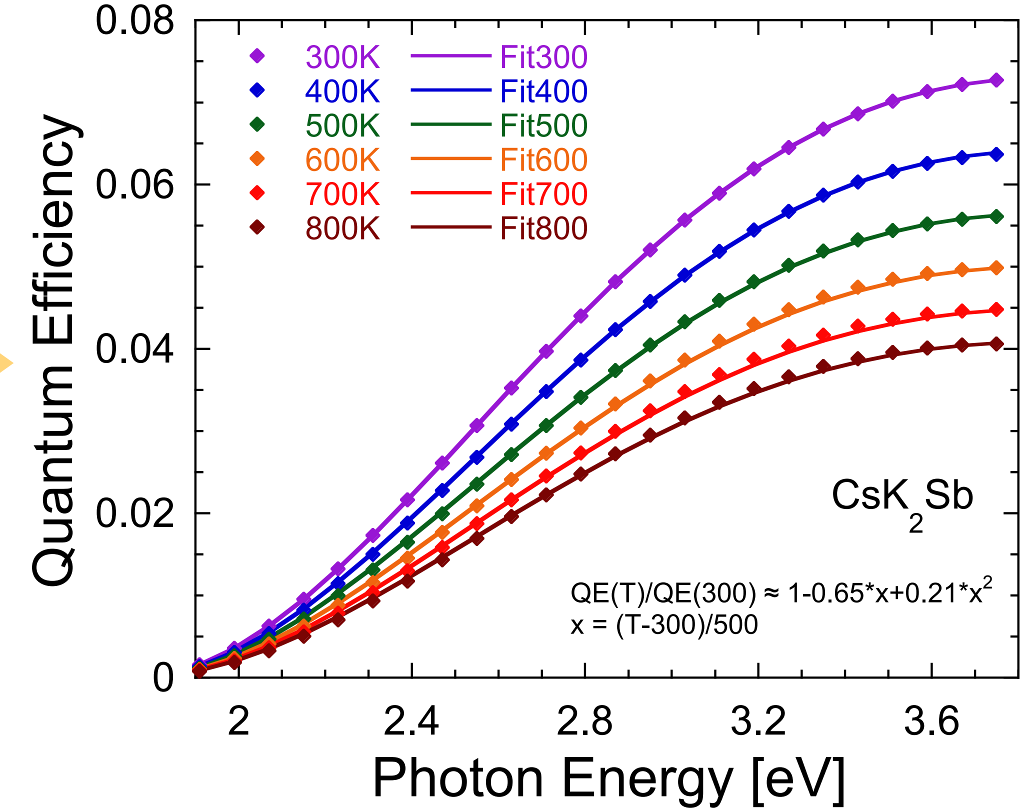
$$QE = \left(1 - R(\omega)\right) \frac{\int_{E_a}^{\hbar\omega - E_g} E\,dE \int_{\sqrt{E_a/E}}^{1} x\,dx D_\Delta\left[Ex^2\right] f_\lambda(x,E)}{2\int_0^{\hbar\omega - E_g} E\left[\int_0^1 x\,dx\right] dE}$$

- Scattering factor: Temperature dependence of QE arises from dependence of scattering rate (inverse relaxation time) on T

factor accounts for fraction of electrons lost due to scattering during emission:
δ= laser penetration depth,
v = electron velocity
τ = scattering time

$$f_\lambda(\cos\theta, p) = \frac{\cos\theta}{\cos\theta + \dfrac{\delta(\hbar\omega)}{v(E)\tau(E)}}$$

Code validated for Cs3Sb Extended to CsK2Sb



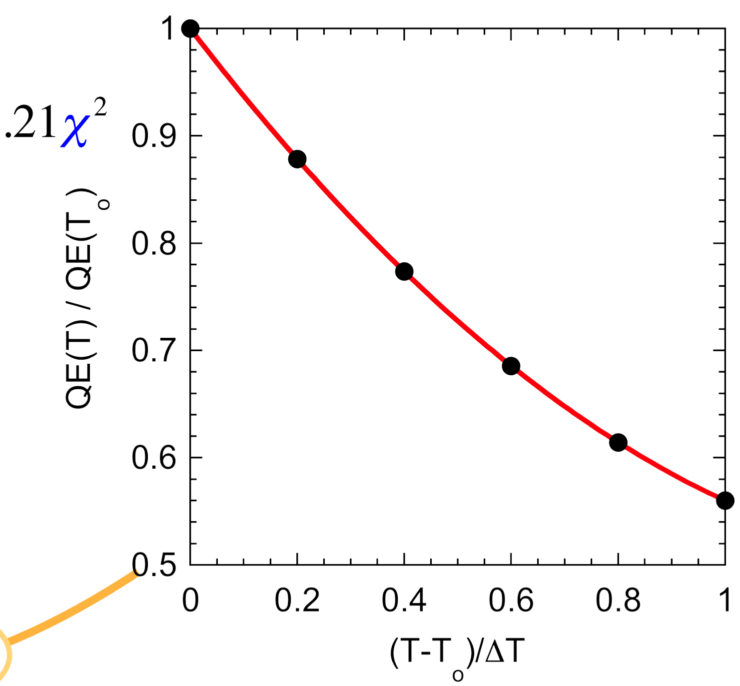
Simulation: QE declines with T because scattering rates increase (i.e., τ(E) decreases)

$$\frac{QE(T)}{QE(T_o)} \approx 1 - 0.65\chi + 0.21\chi^2$$

$$\chi = \frac{T - T_o}{\Delta T}$$

$$T_o = 300K$$

$$\Delta T = 500K$$



Does Cathode heat up during few seconds of illumination?
YES

$$C_v(T)\frac{dT}{dt} = \frac{\Delta E}{\pi r^2 L}$$

$$\Rightarrow T(t) = T_o + 240K\left\{t[\sec]\right\}$$

Assumptions:

- CsK_2Sb is a thin (tens of nm) layer on a Mo disk of thickness L ≈ 0.1 cm w/ radius *r* = 0.5 cm.
- Energy / pulse is 0.115 mJ
- Laser power is 50 W @ 355 nm
- QE(300K) = 2% => ΔQ = 1 nC
- Cv ≈ 2.65 J/K-cm^2 for Moly
- T-rise during a laser pulse is small, but SUM of all pulses add up to a large effect

after 3 sec, QE drops to 60% of initial

SUMMARY

Monte Carlo Corrections to Moments

- Revised scattering operators for electron-electron and acoustic phonon
- Speed improvement, greater accuracy
- Correction factor to Moments Approach:
  R(x) exhibits “universal” behavior - appropriate for PIC simulations

Emitted Electrons and scattering:

- No-Scatter electrons: fast time response - Shell Model
- Scattered electrons: long time tail - Diffusive Sphere Model

Temperature dependence of scattering

- Affects Quantum Efficiency
- Simple model: cathode heats up over time, so QE changes over time

Next: Extend MC techniques to...

- ...QE from semiconductors
  optical phonon replaces e-e as dominant loss mechanism
- ...PIC-ready simulation modules