Photocathode Physics for Photoinjectors Workshop
October 12-14, 2010 Brookhaven National Laboratory (Instrumentation Division, Bldg. 535B, Room A-122)

Modeling of Photoemission: Metals & PEA Semiconductors

Kevin L. Jensen
Code 6843, ESTD
Naval Research Laboratory
Washington, DC 20375-5347
kl.jensen@nrl.navy.mil

Acknowledgments:

- J. Yater, J. Shaw (NRL); E. Montgomery, D. Feldman, P. O'Shea (UMD); J. Smedley (BNL), J. Lewellen (NPS), D. Nguyen (LANL), D. Dowell (SLAC), John Petillo (SAIC), N. Moody (LANL)

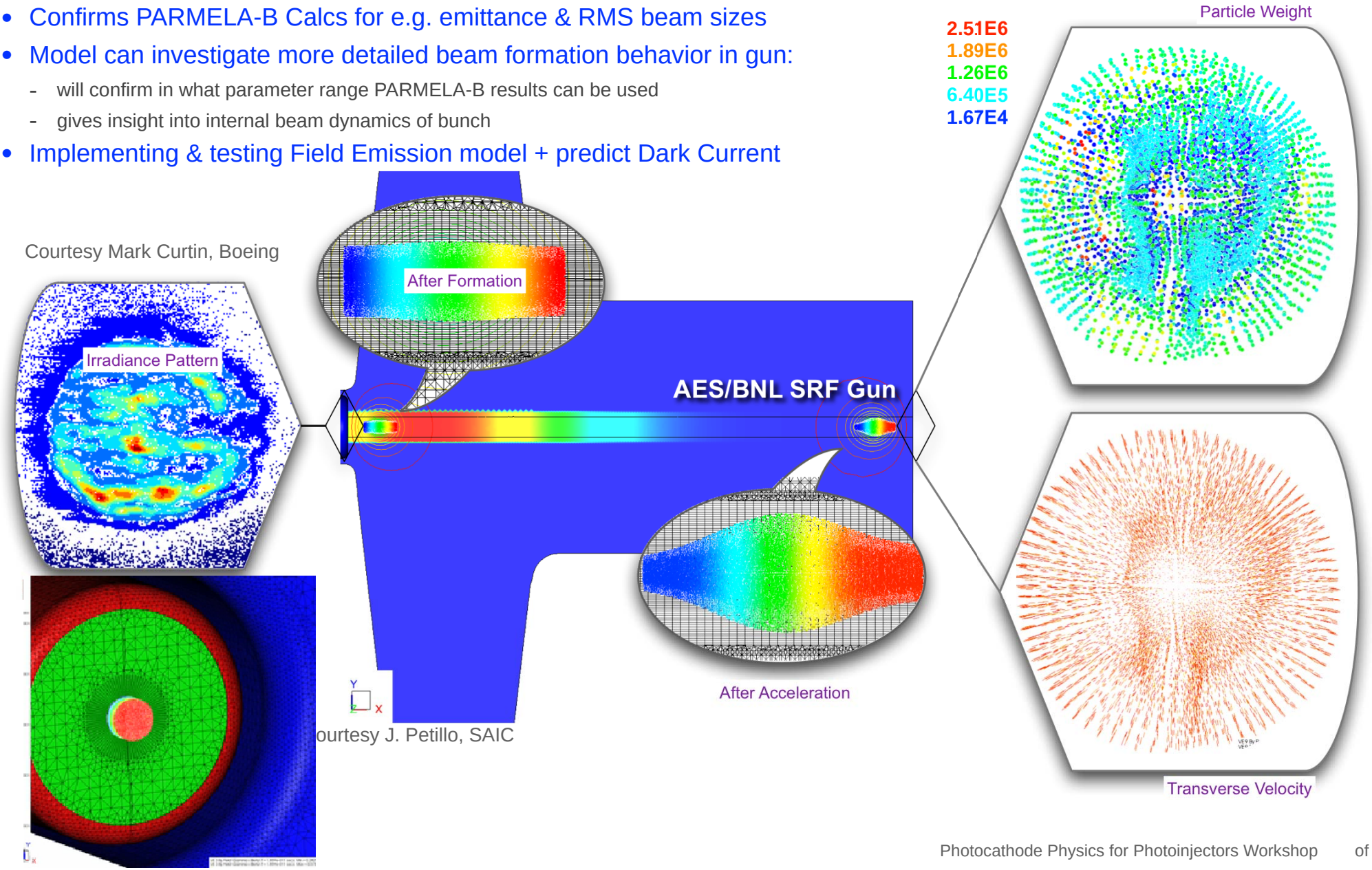
We gratefully acknowledge support from:

- Joint Technology Office, Office of Naval Research

PHOTOEMISSION MODELS IN BEAM CODE

SIMULATION: How Spatial Irradiance Pattern Affects Beam Quality Created from Photoemission Process

- Photoemission Model in NRL/SAIC MICHELLE Code; Applied to AES/BNL SRF Gun
- Confirms PARMELA-B Calcs for e.g. emittance & RMS beam sizes
- Model can investigate more detailed beam formation behavior in gun:
  - will confirm in what parameter range PARMELA-B results can be used
  - gives insight into internal beam dynamics of bunch
- Implementing & testing Field Emission model + predict Dark Current



THE LANGUAGE OF PHOTOEMISSION

Three Components of Photoemission Process

ABSORPTION of light in bulk material and photo-excitation of e-

- laser intensity I_o and frequency ω
- reflectivity R(ω)
- laser penetration depth δ(ω)

TRANSPORT of photo-excited e- to surface w/ scattering

- electron energy E
- scattering rates (relaxation times) τ

EMISSION probability

- Metal: Chemical Potential μ, Work Function Φ
  (work function measured from Fermi level)
- Semiconductor: barrier height E_a, band gap E_g
  (Electron affinity measured from conduction band minimum)

Modified Fowler-Dubridge Model for Metals

$$QE_{MFD} = \left(1 - R(\omega)\right) F_\lambda\left(\delta,\tau\right) P_{FD}\left(\hbar\omega\right)$$

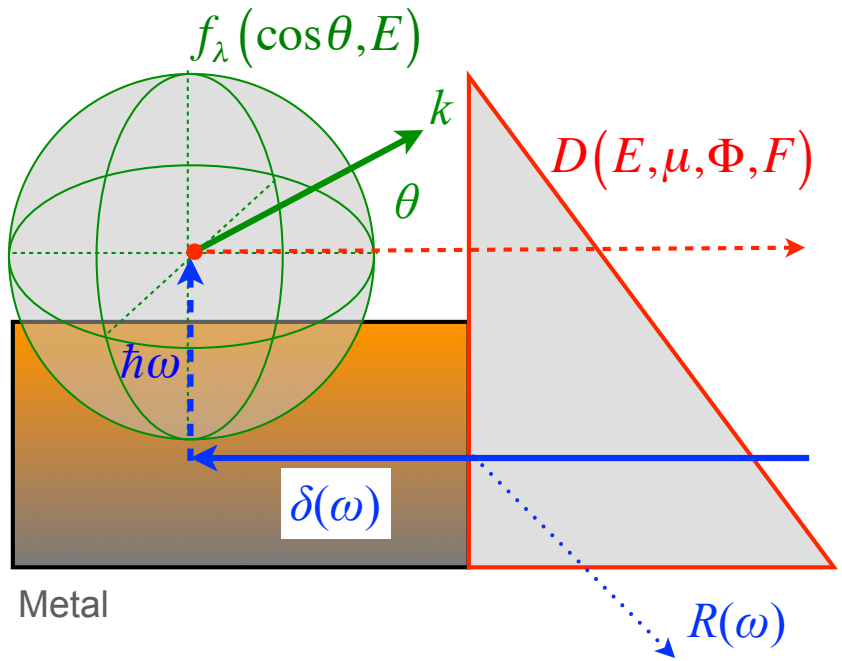
$$P_{FD}\left(\hbar\omega\right) \propto \left(\hbar\omega - \phi\right)^2$$

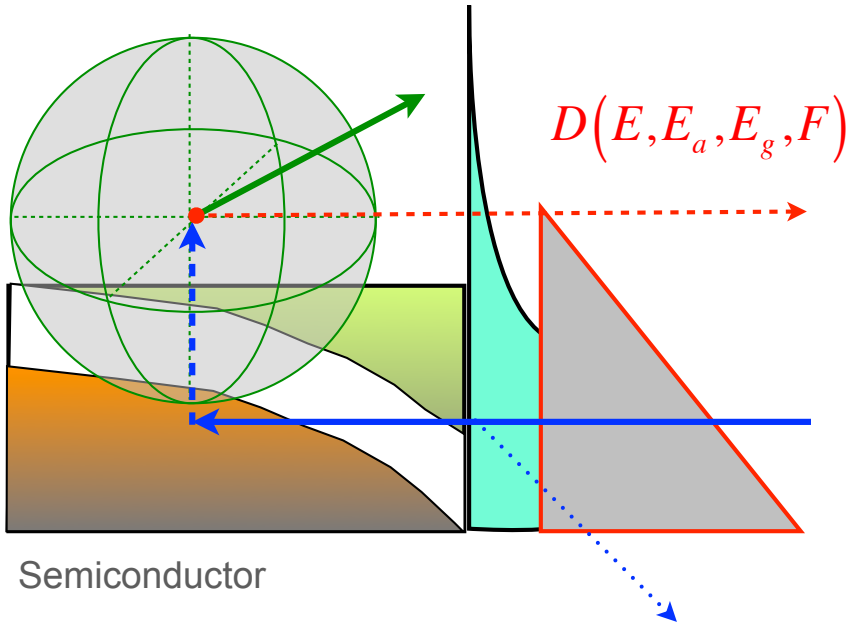
Three-Step Model of Spicer (Semiconductors)

Shown - model developed for semiconductors

- s: quasi-empirical, argued to be 3/2
- B: (Escape)x(Transport) = B* exp(-βx)
- g: absorption factor "over" + "under" barrier terms

$$QE_{spicer} \approx \frac{B}{1 + g\left\{\hbar\omega - \left(E_g + E_a\right)\right\}^{-s}}$$





DISTRIBUTION FUNCTIONS & CURRENT DENSITY

- f(x,k,t) Is Distribution Function Where x & k Are Conjugate Coordinates
- # Of Particles In Phase Space Volume Does Not Change: Taylor Expansion Gives BOLTZMANN'S EQ.
- Integrations Over Momentum Give MOMENTS And CONTINUITY EQ.
- In Quantum Mechanics, Density And Current Become Operators

velocity acceleration

$$\frac{f\left(x+dx,k+dk,t+dt\right)-f(x,k,t)}{dt}\Rightarrow\left\{\frac{\partial}{\partial t}+\frac{\hbar k}{m}\frac{\partial}{\partial x}+\frac{F}{\hbar}\frac{\partial}{\partial k}\right\}f(x,k,t)=0$$

Integral = 0 if f symmetrical

$$\frac{\partial}{\partial t}\rho\left(x,t\right)=\frac{\partial}{\partial t}\left[\frac{1}{2\pi}\int_{-\infty}^{\infty}f(x,k,t)\,dk\right]=-\frac{\partial}{\partial x}\left[\frac{1}{2\pi}\int_{-\infty}^{\infty}\left(\frac{\hbar k}{m}\right)f(x,k,t)\,dk\right]=-\frac{\partial}{\partial x}J\left(x,t\right)$$

Number Density — 0th Moment — 1st Moment — Current Density

"pure state" form

$$\partial_t\hat{\rho}(t)=\frac{i}{\hbar}\left[\hat{H},\hat{\rho}(t)\right]=-\frac{\hbar}{2m}\frac{\partial}{\partial\hat{x}}\left\{\hat{k},\hat{\rho}(t)\right\}=-\frac{\partial}{\partial\hat{x}}\hat{j}\left(t\right)\Rightarrow j_k(x,t)=\frac{\hbar}{2m}\left\langle x\left|\left\{\hat{\rho}(t),\hat{k}\right\}\right|x\right\rangle=\frac{\hbar}{2mi}\left\{\psi_k^{\dagger}\partial_x\psi_k-\psi_k\partial_x\psi_k^{\dagger}\right\}$$

Mixed state form

$$\hat{\rho}(t)=\sum f_{FD}\left(E_k\right)\left|\psi_k(t)\right\rangle\left\langle\psi_k(t)\right|\Rightarrow\rho\left(x\right)=\left(2\pi\right)^{-3}\int dk\int d\mathbf{k}_{\perp}f_{FD}\left(E(\mathbf{k})\right)\left|\psi_k\left(x\right)\right|^2$$

Transmission Probability
ratio of transmitted to incident pure state current density

$$D\left(k\right)\equiv\frac{j_k(k)\big|_{transmitted}}{j_k(k)\big|_{incident}}$$

Energy
parabolic in momentum k

$$E\left(k\right)\equiv\frac{\hbar^2}{2m}\left(k^2+\mathbf{k}_{\perp}^2\right)$$

Supply Function
Fermi Distribution integrated over transverse momentum

$$f\left(k\right)=\left(2\pi\right)^{-2}\int f_{FD}\left(E\left(k,\mathbf{k}_{\perp}\right)\right)d\mathbf{k}_{\perp}$$

Tsu-Esaki-like formula

Velocity — Supply Function

$$J(F,T)=\frac{1}{2\pi}\int_0^{\infty}\frac{\hbar k}{m}D\left(k\right)f\left(k\right)dk$$

Transmission Probability

Current density:
1st Moment of distribution D(k)f(k)

EQUATIONS OF ELECTRON EMISSION

$$J(F,T)=\frac{1}{2\pi\hbar}\int_0^\infty D(E)f(E)dE$$

THERMAL
Low Field, High Temperature

Richardson-Laue-Dushman Eq.

$$D(E)=\Theta\left[E-(\mu+\Phi)\right]$$

$$f(E)=\frac{mk_BT}{\pi\hbar^2}\exp\left[(\mu-E)/k_BT\right]$$

$$J_{RLD}(T)=\left(\frac{mk_B^2}{2\pi^2\hbar^3}\right)T^2\exp\left[-\Phi/k_BT\right]$$

$T=1273$ K

$\Phi=2.1$ eV

$J_{RLD}\approx 1$ A/cm^2

Thermionic cathodes run at lower T levels than possible to preserve lifetime

FIELD
High Field, Low Temperature

Fowler Nordheim Eq.

$$D(E)\approx\exp\left[-(B/F)-C(\mu-E)\right]$$

$$f(E)=\frac{m}{\pi\hbar^2}(\mu-E)\Theta(\mu-E)$$

$$J_{FN}(F)=\frac{F^2}{16\pi^2\hbar\Phi}\exp(-B/F)$$

$B=\frac{4}{3\hbar}\sqrt{2m\Phi^3}$; $C=\frac{2}{\hbar F}\sqrt{2m\Phi}$

$F=7$ GV/m

$\Phi=4.5$ eV

$J_{RLD}\approx$ 1.5x10^5 A/cm^2

Field emission occurs over (5 nm)^2 emission areas

PHOTO
Low Field & Temp, Photon

Fowler-Dubridge Relation

$$D(E)=\Theta\left[E+\hbar\omega-(\mu+\Phi)\right]$$

$$f(E)=\frac{m}{\pi\hbar^2}(\mu-E)\Theta(\mu-E)$$

$$J_{FD}(\hbar\omega)=\frac{m}{4\pi^2\hbar^3}(\hbar\omega-\Phi)^2$$

$\hbar\omega=4.66$ eV

$\Phi=4.5$ eV, $\mu=7$ eV

$$QE_{FD}\approx\frac{J_{over}}{J_{all}}=\frac{(\hbar\omega-\Phi)^2}{\mu^2}$$

$=0.1\%$

Losses due to scattering lowers estimate

More? See K.L. Jensen. J. Appl. Phys. 102, 024911 (2007).

MOMENTS-BASED QE EVALUATION

"Moments" defined by generalizing "distribution function" D(E) f(E)

QE = ratio of two moments: Actual vs. All Possible

- Actual: f_λ and $D(E) < 1$; All: f_λ and $D(E) = 1$
- Coefficient to account for Reflection
- Caution: Simple parabolic energy model used for Density of States (DOS): not actual - causes differences with, e.g., Pb & other metals

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

$$QE = \left\{1-R(\omega)\right\}\frac{M_1(actual)}{2M_1(all)}$$

This is absorption

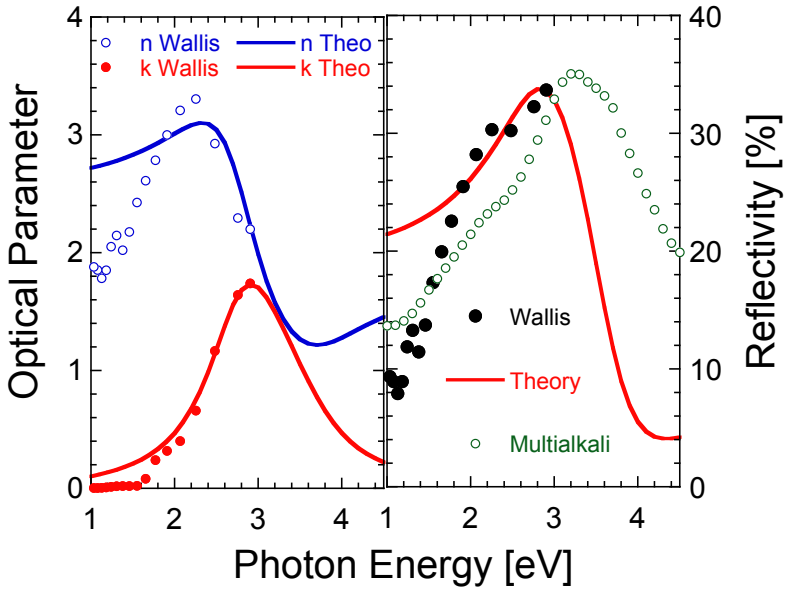
This is forward momentum (cos would be sin for emittance calc)

This is transmission probability for surface barrier

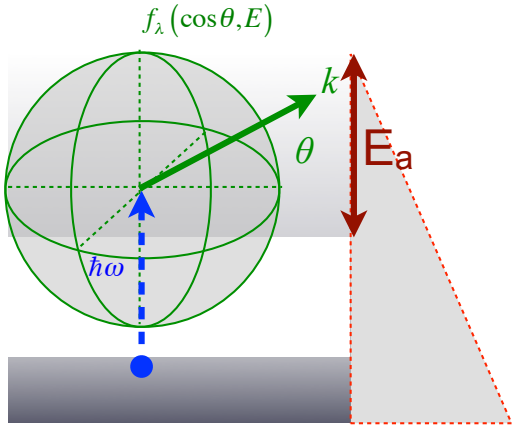
This is scattering loss factor for bulk transport

This is initial and final state occupation factors (metals) or bandgap constraint (semi)

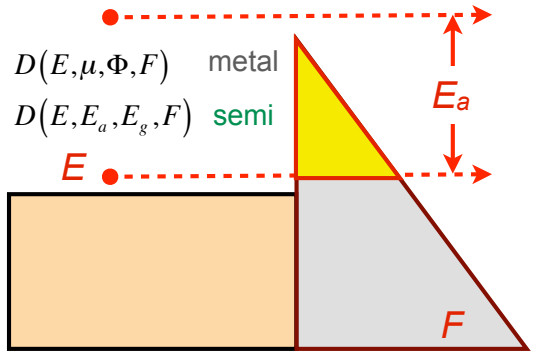
Reflection (topic 1)



Scattering (topic 3)



Transmission (topic 2)

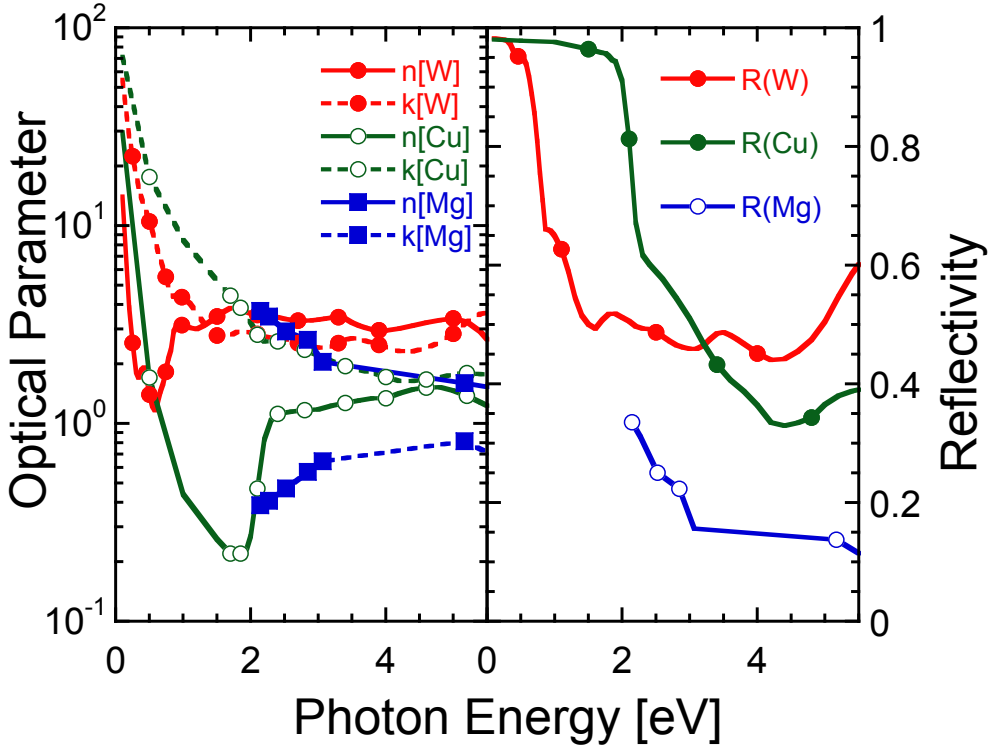


REFLECTIVITY AND PENETRATION

For **METALS**, spline fitting of readily available n, k data works well

- k = extinction coefficient
- n = index of refraction
- Off-normal reflectivity related to normal values

$$\frac{\varepsilon}{\varepsilon_0} = (n - ik)^2 \Rightarrow \begin{cases} R(\omega) = \dfrac{(n-1)^2 + k^2}{(n+1)^2 + k^2} \\ \delta(\omega) = \dfrac{\lambda}{4\pi k} = \dfrac{c}{2k\omega} \end{cases}$$

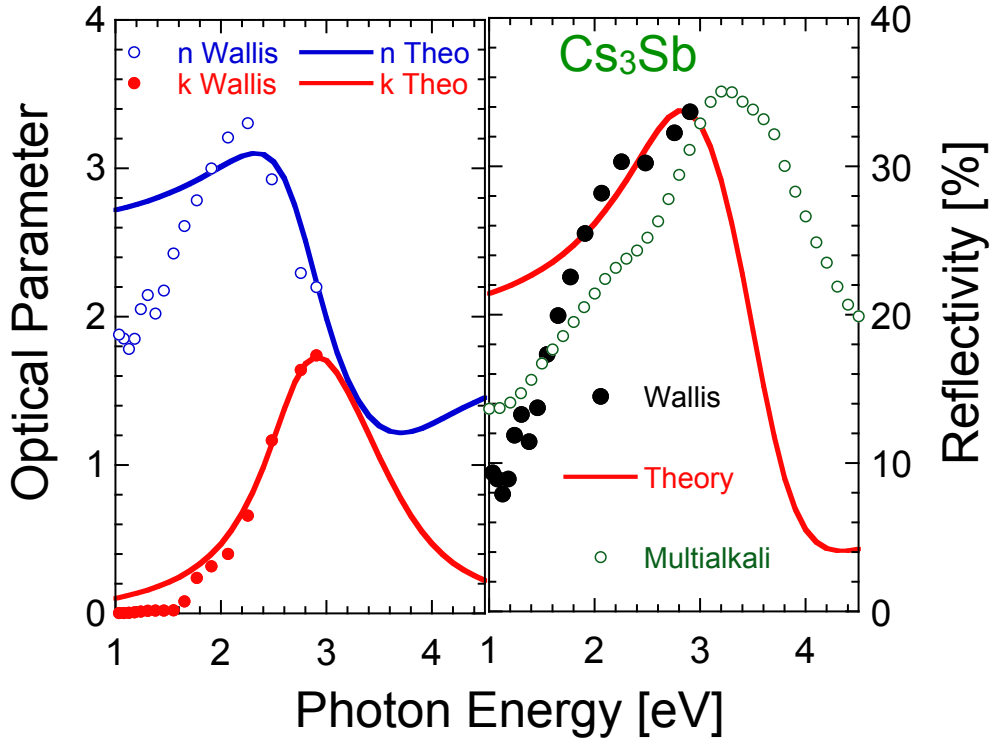


For **SEMICONDUCTORS**... A Drude-Lorentz model makes up for incomplete n,k data

- K_o, K_∞ = static & high freq. dielectric const
- γ_o = damping term
- ω_T = transverse optical phonon
- Some semiconductors may require multiple ω_T

$$n^2 - k^2 \Rightarrow K_\infty + (K_o - K_\infty)\frac{\omega_T^2\left(\omega_T^2 - \omega^2\right)}{\left(\omega^2 - \omega_T^2\right)^2 + \left(\gamma_o\omega_T\omega\right)^2}$$

$$2nk \Rightarrow (K_o - K_\infty)\frac{\gamma_o\omega\omega_T^3}{\left(\omega^2 - \omega_T^2\right)^2 + \left(\gamma_o\omega_T\omega\right)^2}$$



TRANSMISSION PROBABILITY: A SIMPLE MODEL

Match wave function and first derivative

- at x = 0

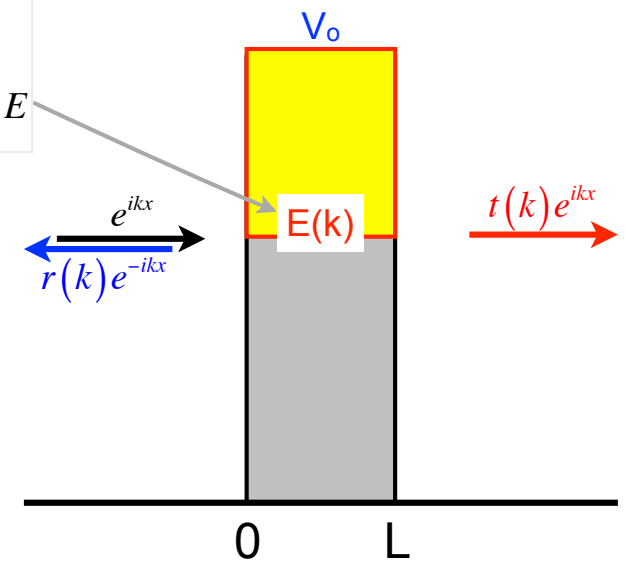
$$\begin{pmatrix} 1 & 1 \\ ik & -ik \end{pmatrix}\begin{pmatrix} 1 \\ r(k) \end{pmatrix}=\begin{pmatrix} 1 & 1 \\ \kappa & -\kappa \end{pmatrix}\begin{pmatrix} a \\ b \end{pmatrix}$$

- at x = L

$$\begin{pmatrix} e^{\kappa L} & e^{-\kappa L} \\ \kappa e^{\kappa L} & -\kappa e^{-\kappa L} \end{pmatrix}\begin{pmatrix} a \\ b \end{pmatrix}=\begin{pmatrix} e^{ikL} & e^{-ikL} \\ ike^{ikL} & -ike^{-ikL} \end{pmatrix}\begin{pmatrix} t(k) \\ 0 \end{pmatrix}$$

$$\frac{\hbar^2 k^2}{2m} = E$$

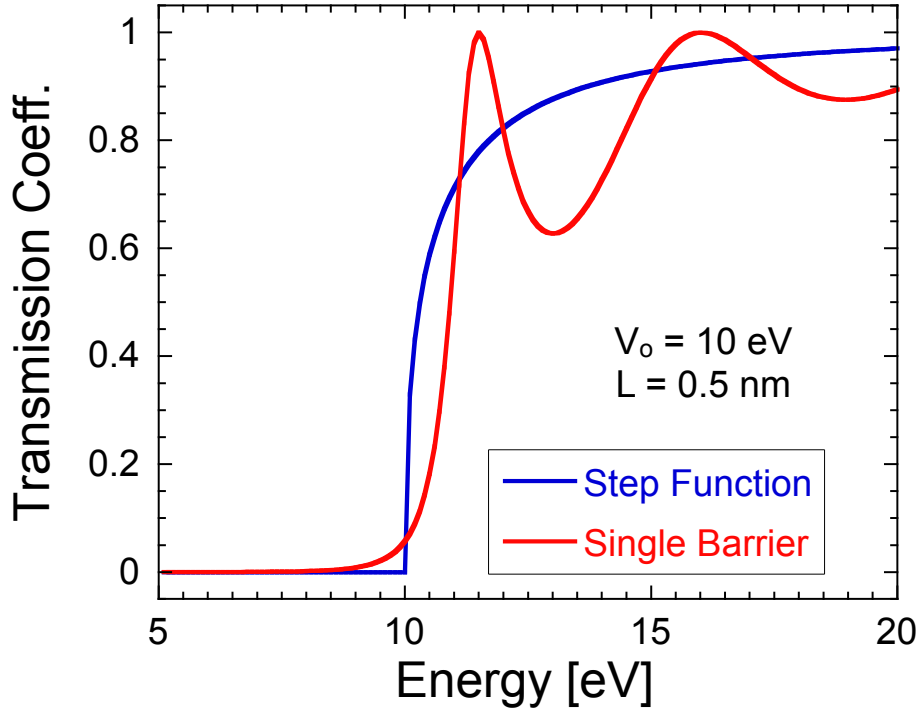
$$\frac{\hbar^2 \kappa^2}{2m} = V_o - E$$



Transmission Probability (E<Vo)

$$t(k)=\frac{2\kappa k}{2\kappa k\cosh(\kappa L)+i\left(\kappa^2-k^2\right)\sinh(\kappa L)}e^{-ikL}$$

$$D(k)=\left|t(k)\right|^2=\frac{(2\kappa k)^2}{(2\kappa k)^2+\left(\kappa^2+k^2\right)\sinh^2(\kappa L)}\approx\left(\frac{2k}{\kappa}\right)^2 e^{-2\kappa L}$$



"Area Under Curve" Interpretation

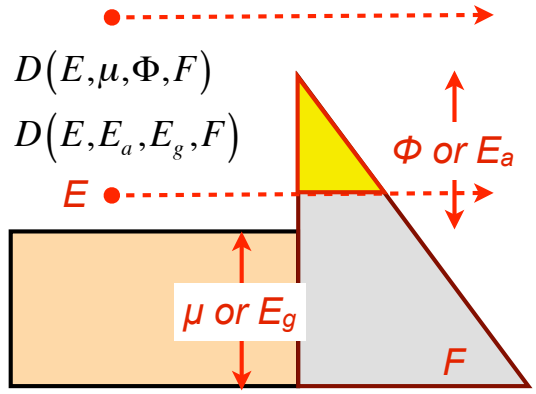
$$2\kappa L=\frac{2L}{\hbar}\sqrt{2m\left(V_o-E\right)}=2\int_{x_{\min}}^{x_{\max}}k(x)\,dx$$

This is 2 x "area" (yellow) under potential max V_o but above E(k)

EMISSION PROBABILITY FOR TRIANGULAR BARRIER

Barrier with applied Field: D(E) is NOT Step Function

- Triangular Barrier from field emission theory
  useful for semiconductors with small electron affinities
- Exact Solution = Airy Functions
- Approximation: JWKB Method
  - D(E) calculation shows a hyperbolic tangent behavior
  - Integrals with tanh act like step + Dirac Delta function correction
    as long as tanh varies more rapidly than remainder of integrand



tanh-func step delta-func

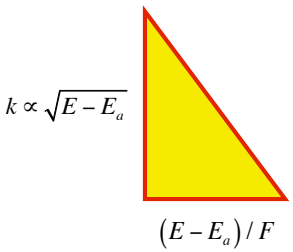
$$\int_0^\infty \frac{g(x)}{1+e^{(x_o-x)/\varepsilon}}dx \approx \int_0^{x_o} g(x)dx - \frac{(\pi\varepsilon)^2}{6}\partial_x g(x_o)$$

For F = 20 qMV/m, term is $(0.01\ \text{eV})^2$

$$|E-E_a|_+ \approx \left[(E-E_a)^2 + \frac{4}{25}\left(\frac{\hbar^2 F^2}{2m}\right)^{2/3}\right]^{1/2}$$

$$\theta(E<E_a) = 2\int k(x)dx = \frac{2\hbar^2}{3mF}\left[\frac{2m}{\hbar^2}(E_a-E)\right]^{3/2}$$

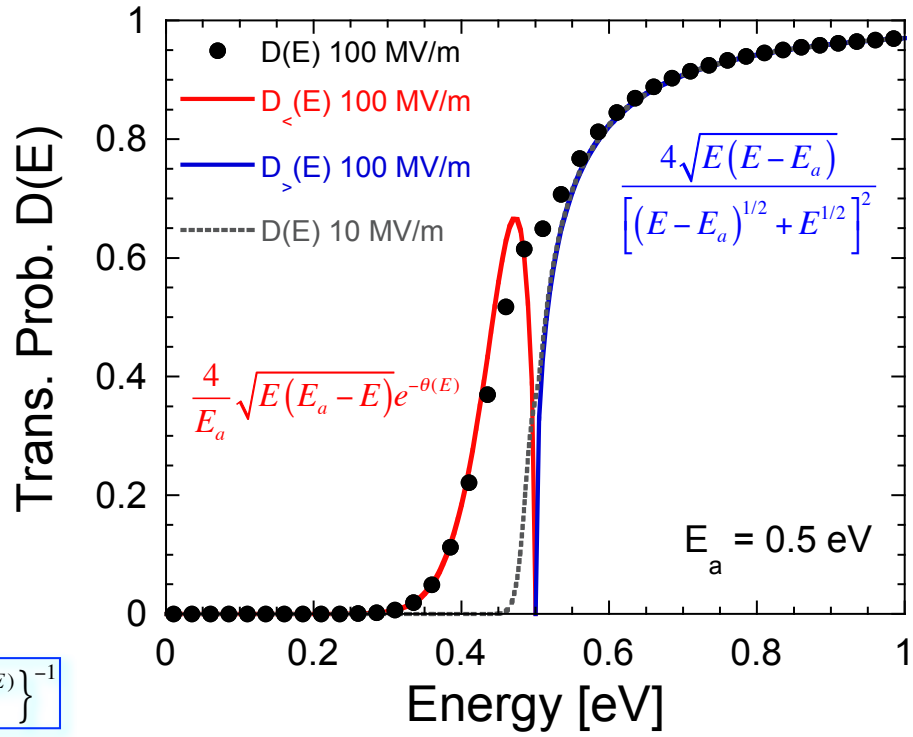
JWKB "area under curve"



$$D_\Delta(E) = \frac{4\sqrt{E|E-E_a|_+}}{2\sqrt{E|E-E_a|_+} + \left(|E-E_a|_+ + E\right)e^{\theta(E)}}$$

For Image Charge (metals): append v(y) to θ(E), replace E_a with Φ and use Kemble approximation

$$D(E) = \left\{1+e^{\theta(E)}\right\}^{-1}$$



SCATTERING AND TRANSPORT FACTOR

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

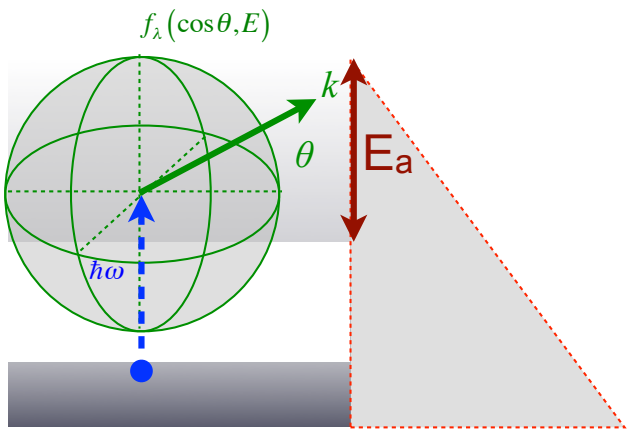
Assume Any Scattering Event Is Fatal To Emission ("Fatal Approximation")

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

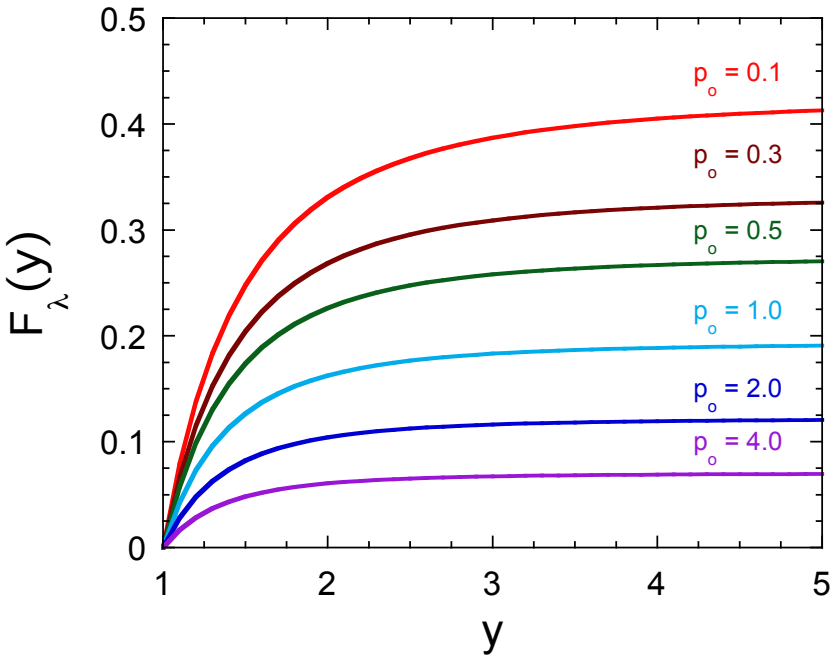
Example: Cu-like
- δ = 12.6 nm
- v/c = 0.675%
- τ = 2.6 fs
- ➡ p ≈ 2.38

Weighted Scattering Fraction (e.g. MFD 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 $E \equiv E_a y^2$
- **IF** τ scales as 1/k, then p is constant $p \approx p_o$

SCATTERING MECHANISMS

Electron-Electron (metals)

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

q_o is screening factor

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

Acoustic (metals&semi)

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

W_: Bloch-Grünnisen

Non-polar Optical (semi)

$$\tau_{nop}^{emission} \approx \frac{\sqrt{2}\pi\rho\hbar^3\omega_0}{D_o^2 m^{3/2}} \left\{ \left(n(\hbar\omega_0) + 1 \right) \sqrt{E - \hbar\omega_0} \right\}^{-1}$$

phonon emission

$$\tau_{nop}^{absorption} \approx \frac{\sqrt{2}\pi\rho\hbar^3\omega_0}{D_o^2 m^{3/2}} \left\{ n(\hbar\omega_0) \sqrt{E + \hbar\omega_0} \right\}^{-1}$$

absorption

$$n(\hbar\omega_0) = 1 / \left\{ \exp(\hbar\omega_0 / k_B T) - 1 \right\}$$

Bose-Einstein (phonon) distribution

Polar Optical (semi)

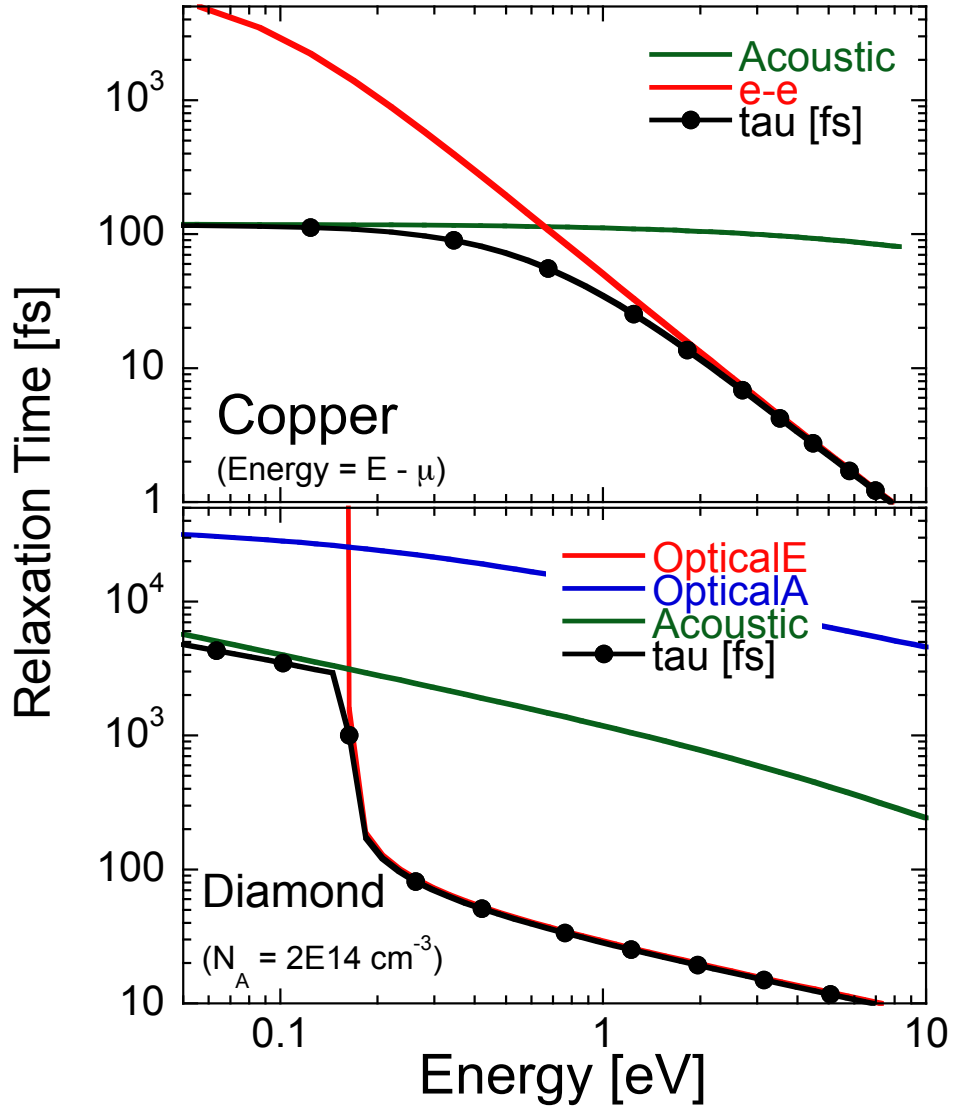
$$\tau_{pop}(\hbar\omega_q, E) = \left\{ 2\omega_q \left(\frac{1}{K_o} - \frac{1}{K_\infty} \right) \left(2n(\beta\hbar\omega_q) + 1 \right) \eta\left(\frac{E}{E_g} \right) \right\}^{-1}$$

e & a combined

$$\eta(u) = \frac{16u^2 + 18u + 3}{3(1+2u)\sqrt{u(u+1)}}$$

Ionized & Neutral Impurity (semi)

$$\tau_{ii}(E) \approx \frac{\alpha_{fs} m^2 c}{4\pi\hbar^2} \left(\frac{K_o^2}{N_i} \right) \frac{k_B T}{\ln(2)} \frac{\sqrt{E(E+E_g)}}{E_g^2}; \tau_{ni} = \frac{\alpha_{fs} m c^2}{20\hbar^2 K_o N_n}$$



General
- β = $1/k_BT$ (T_o = RT)
- E = Electron energy
- k_B = Boltzmann's constant
- a_o = Bohr Radius
- α_{fs} = Fine structure constant
- m = e– mass (eff. or rest)

Specific
- θ = Debye Temperature
- Ξ = Deformation Potential
- D_o = Energy per atomic displacement
- ρ = Mass density
- v_s = sound velocity
- ω_x = phonon frequencies
- hk = Momentum $2\pi(2mE)^{1/2}$
- K_o, K_∞ = Dielectric constants
- E = Electron energy
- N_i, N_n = neutral &ionized impurity conc
- F = external field

MOMENTS-BASED QE: METALS

Simplification of Defining Integral

(note: E refers to electron energy *prior* to photon absorption)

- Numerator (Actual): f_λ & D < 1
- Denominator (All): f_λ & D = 1, Φ = 0
- Fermi functions act like theta functions on limits of integration

$$QE = \left(1 - R(\omega)\right) \frac{\int_{\mu-(\hbar\omega-\phi)}^{\mu} E\,dE \int_0^1 x\,dx D\left\{(E+\hbar\omega)x^2\right\} f_\lambda\left[x, p(E)\right]}{2\int_{\mu-\hbar\omega}^{\mu} E\left[\int_0^1 x\,dx\right] dE}$$

Leading Order Approximation

- Treat D as step function in energy
- Define $\varphi(E) \equiv \left[(\mu+\phi)/(E+\hbar\omega)\right]^{1/2}$

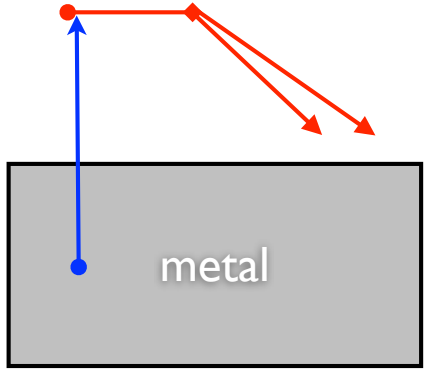
$$QE_0 = \frac{\left(1-R(\omega)\right)}{\hbar\omega\left(2\mu-\hbar\omega\right)} \int_{\mu-(\hbar\omega-\phi)}^{\mu} \frac{\left\{1-\varphi(E)^3\right\}}{1+p\left(E,\hbar\omega\right)} E\,dE$$

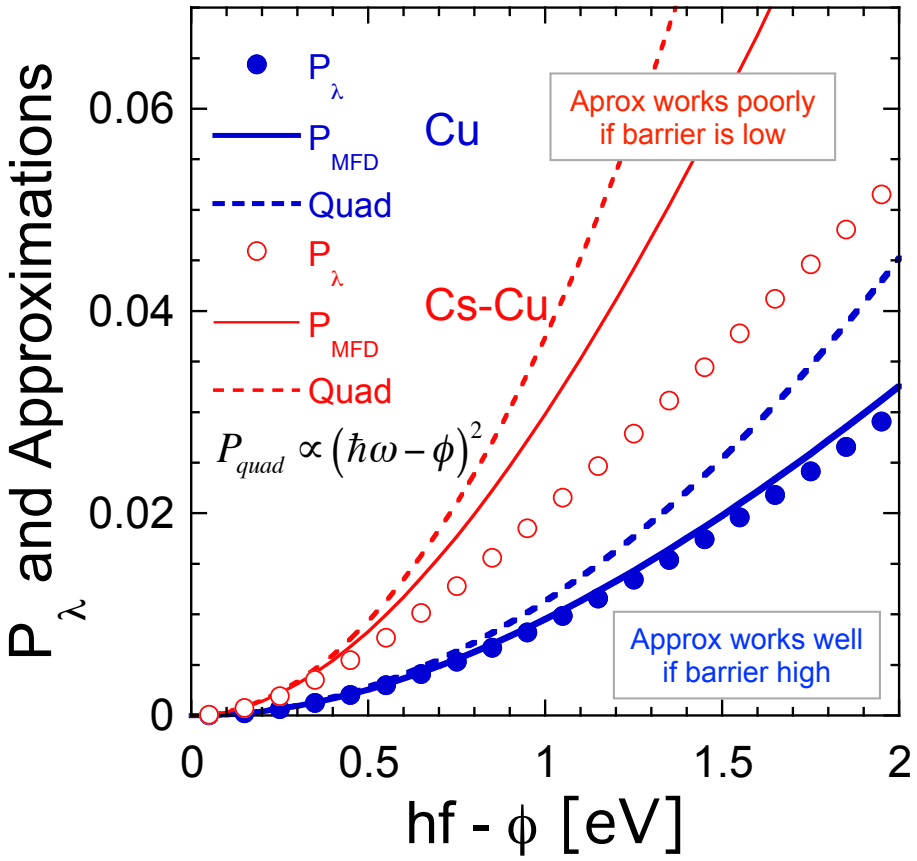
Relationship to Modified Fowler Dubridge (MFD) Approximation

- Ignore E dep. of relaxation time
- Evaluate scattering factor term p at Fermi Level
- Let $1-\varphi(E) \ll 1$

$$QE_{MFD} = \frac{\left(1-R(\omega)\right)}{1+p\left(\mu,\hbar\omega\right)} \frac{3\mu(\mu+\phi)}{4\phi(2\mu-\phi)} \left(\frac{\hbar\omega-\phi}{\hbar\omega+\mu}\right)^2 \Leftrightarrow \left(1-R(\omega)\right) F_\lambda P_{MFD}$$

e-e scattering fast in metals & quickly reduces E of photoexcited e⁻. Both final states must be > μ





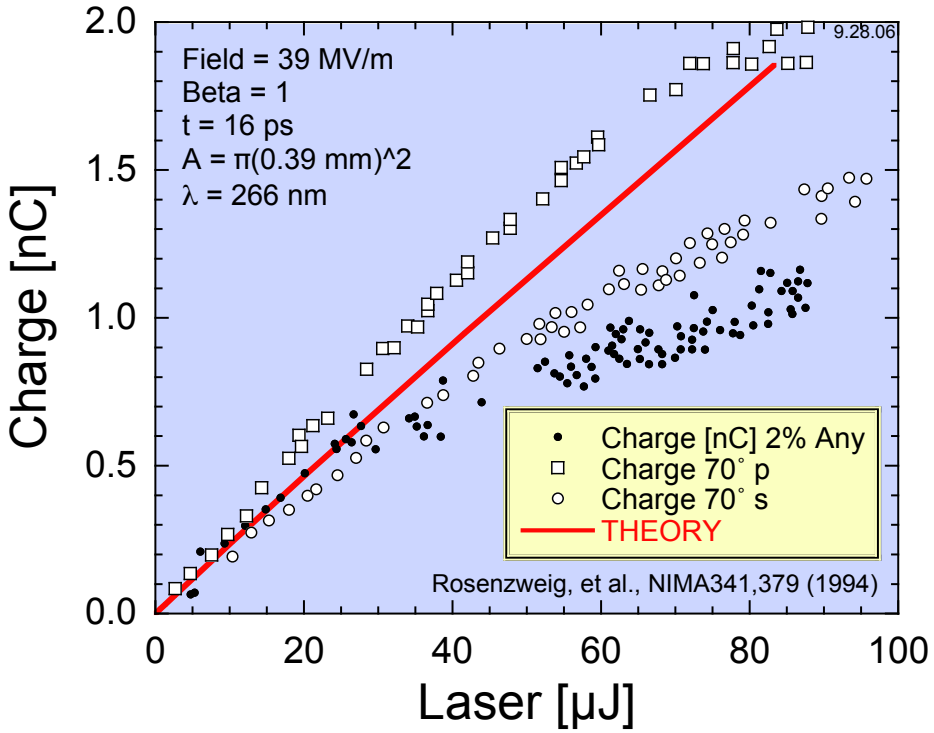
METAL THEORY VS EXPERIMENT

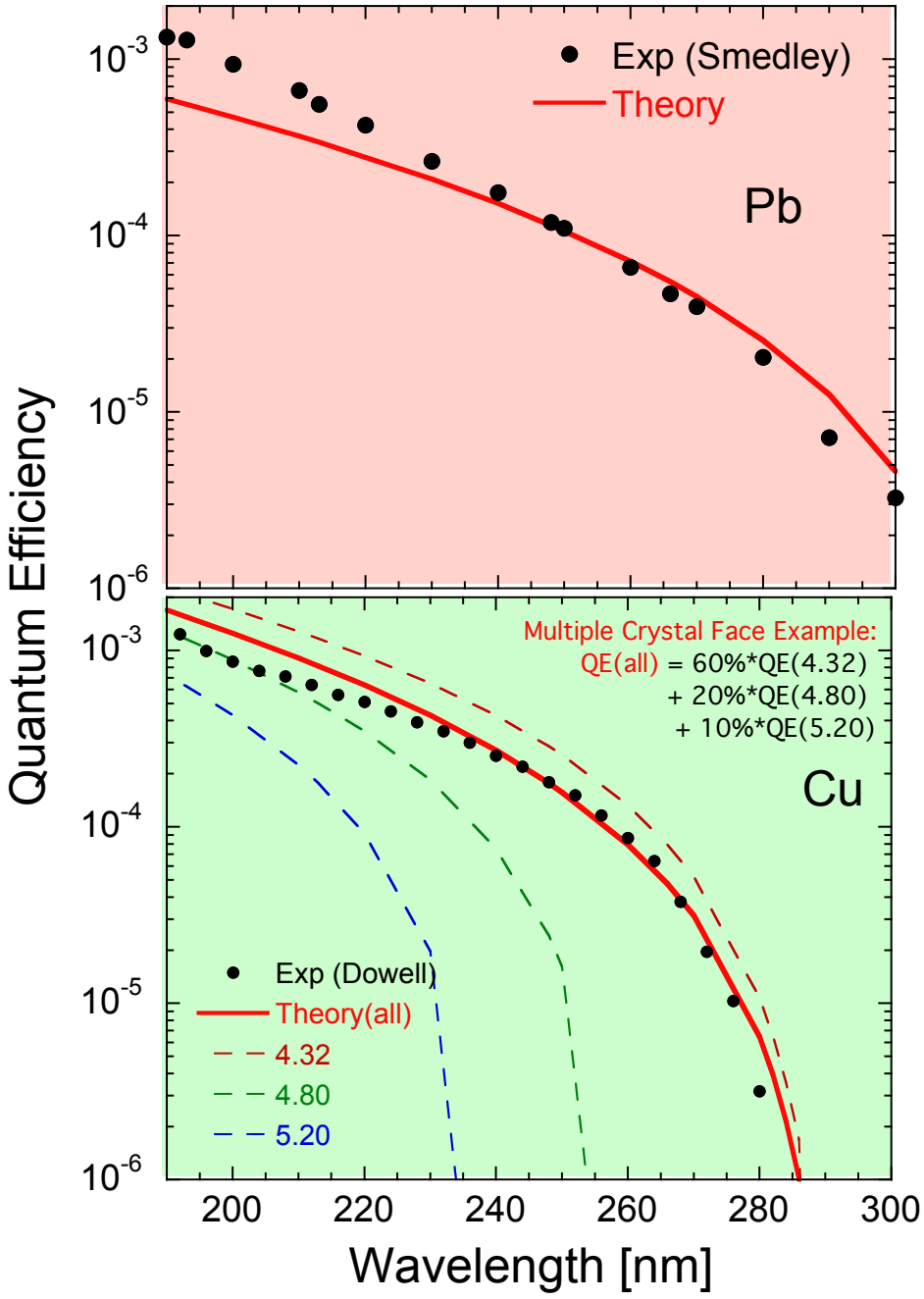
Theory vs Exp for Cu:

- Lower Left: data courtesy of GraphClick
  Parameters conform to
  Rosenzweig, et al, NIMA341, 379 (1994)
- Lower Right: data courtesy of D. Dowell
  Parameters conform to
  D.H. Dowell, et al. PRSTAB9, 063502 (2006).

Theory vs Exp for Pb:

- Upper Right: data courtesy of J. Smedley
  Data file: Solid lead.xls, "lamp Clean III" (2005)





MOMENTS-BASED QE: SEMICONDUCTORS

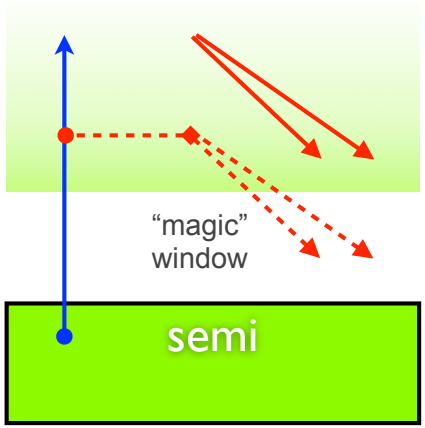
Simplification of Defining Integral

(note: E refers to electron energy *after* to photon absorption)

- Numerator (Actual): f_λ & D < 1
- Denominator (All): f_λ & D = 1, E_a = 0
- Fermi functions act like theta functions on limits of integration

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

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



Leading Order Approximation

- Ignore E dep. of relaxation time

$$F_\lambda(x) = \int_{1/x}^{1} s f_\lambda\left(s, E_a x^2\right) ds$$

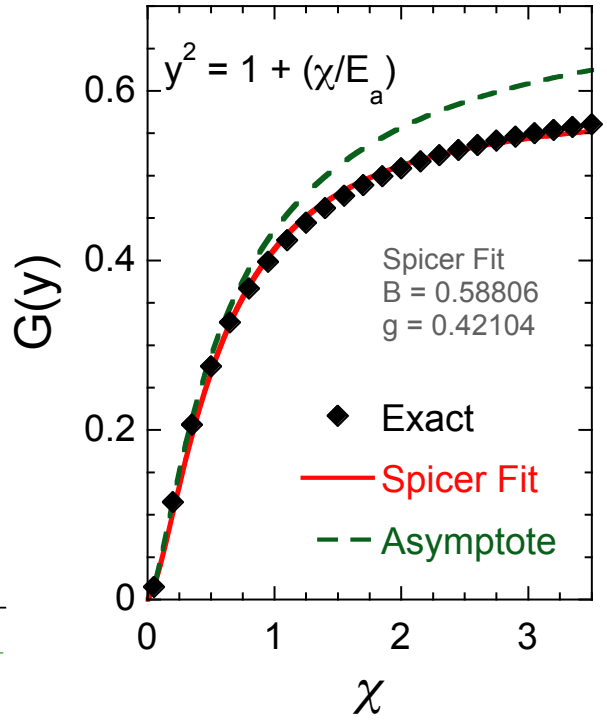
$$G_\lambda(y) = \frac{8}{y^4}\int_1^y x^3 F_\lambda(x)\,dx$$

$$QE_0 = \frac{1}{2}\left(1 - R(\omega)\right) G_\lambda\left(\sqrt{1 + \frac{\chi}{E_a}}\right)$$

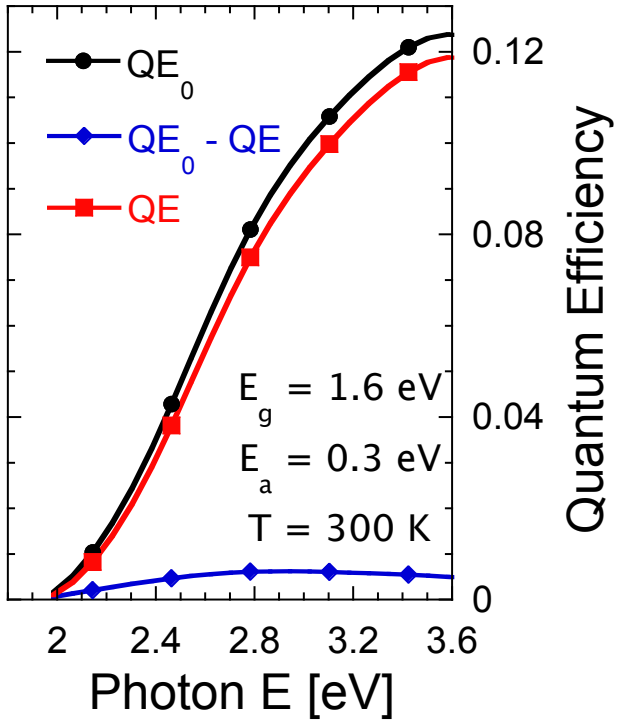
Relationship to the Spicer 3-Step Model

$$\lim_{\chi \to 0} QE_0 \approx \frac{\left(1 - R(\omega)\right)}{2\left(p_o + 1\right)\left(1 + \frac{E_a}{\chi}\right)^2} \Leftrightarrow QE_{spicer} \approx \frac{B}{1 + \frac{g}{\chi^{3/2}}}$$

Spicer Parametric Fit Is Good



QE_0 Works Well - Good For PIC



ALPHA SEMICONDUCTOR MODEL

An Alpha Semiconductor Model (i.e., A Generic Semiconductor) Can Provide Parameters (e.g., Effective Mass) If Such Quantities Are Unknown / Ill-defined / Unavailable

Also, Gives Forms Of Polar Optical And Ionized Impurity Scattering That Are Related To, But Different Than, Small Electron Energy Representations Found In Transport / Scattering Tomes

Alpha Semiconductor Model Restricts Upper Limit On Electron Velocity In Semiconductor To Half Of Product Of Fine Structure Constant With Speed Of Light; Implies Relationship Between Band Gap Energy And Electron Effective Mass

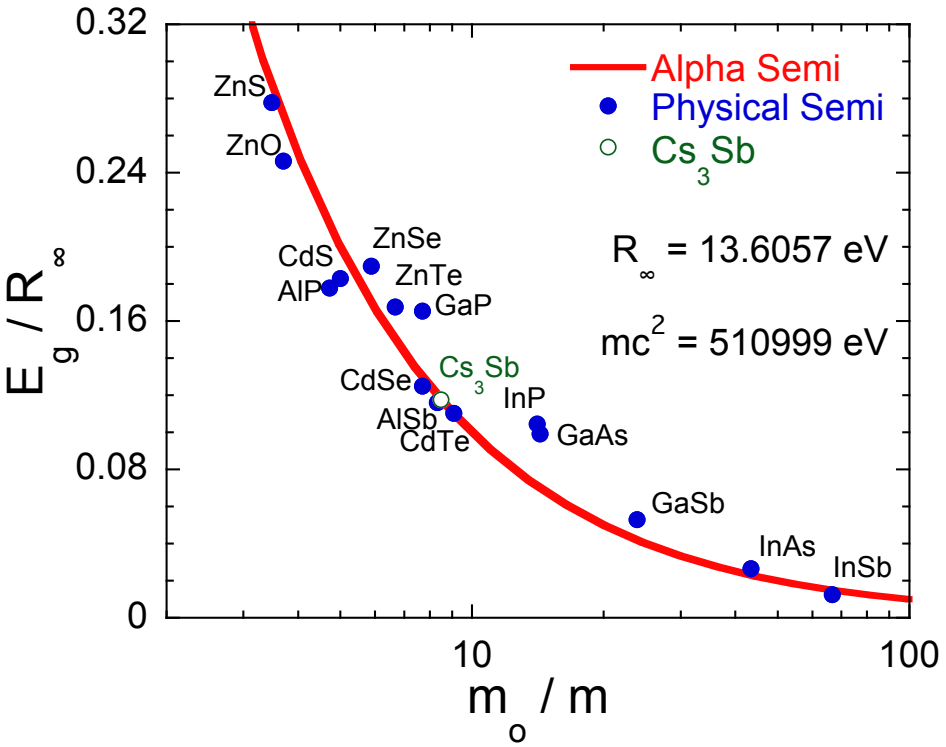
$$\frac{E_g}{R_\infty} = \frac{m}{m_o}$$

$$\frac{1}{\tau_{pop}\left(\hbar\omega_q, E\right)} = 2\omega_q\left(\frac{1}{K_o} - \frac{1}{K_\infty}\right)\left(2n\left(\beta\hbar\omega_q\right)+1\right)\eta\left(\frac{E}{E_g}\right)$$

$$\eta(u) = \frac{16u^2+18u+3}{3(1+2u)\sqrt{u(u+1)}}$$

For u small, $\eta(u) \approx 1/u^{1/2}$

$$\frac{1}{\tau_{ii}(E)} \approx \frac{4\pi\hbar^2}{\alpha_{fs} m^2 c}\left(\frac{N_i}{K_o^2}\right)\frac{\ln(2)}{k_B T}\frac{E_g^2}{\sqrt{E\left(E+E_g\right)}}$$



General

- β = $1/k_BT$ (T_o = Room T)
- k_B = Boltzmann's constant
- K_o, K_∞ = Static & High Freq. Dielectric Const.
- α_{fs} = Fine structure constant
- m = electron mass (eff.)
- E = Electron energy
- N_i = ionized impurity concentration
- n(x) = Bose-Einstein Distribution $1/[e^x - 1]$

Scattering for Cs_3Sb: Typical Values at RT in fs

- Polar Optical on the order of 26.6
- Ionized Impurity on the order of 4395.0
- Acoustic Phonon on the order of 694.0

K.L. Jensen, B.L. Jensen, E.J. Montgomery, D.W. Feldman, P.G. O'Shea, and N. Moody, J. Appl. Phys. 104, 044907 (2008).

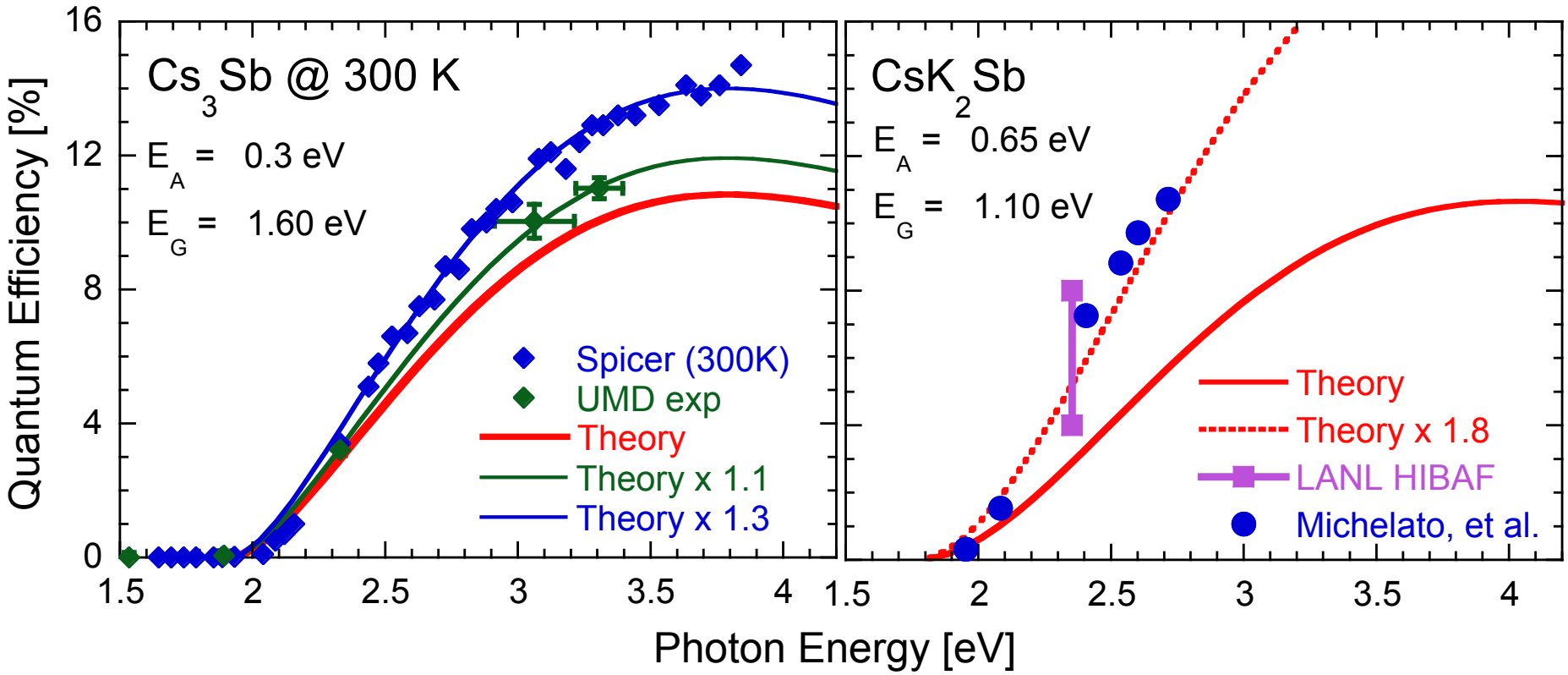
SEMICONDUCTOR THEORY VS EXPERIMENT

Theory vs Exp for Cs_3Sb:

- Theory: E_a and E_g from literature, other parameters from Drude-Lorentz Model for evaluation of dielectric constant and Alpha Semiconductor model for evaluation of electron effective m from E_g
- Experiment: data from UMD; 300K line from Fig. 8, W.E. Spicer, Phys. Rev. 112, 114 (1958).

Theory vs Exp for CsK_2Sb:

- E_a and E_g from literature, other parameters from Cs_3Sb
- Experiment: data from Fig. 1, P. Michelato, et al., Proc IEEE PAC 2, 1049 (1995) and P. G. O'Shea (private communication) - see D.W. Feldman, et al., NIMA 304, 224 (1991)



EMITTANCE EVALUATION BY MOMENTS APPROACH

Transverse Moments Evaluation for Metals & Semiconductors

Moments Approach can be used to calculate the weighted averages used in calculation of emittance

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

Important: Cons. of **k** across surface for k_x & k_y of wave function demands appending photon energy to E in moment term
Dowell, Schmerge, PRSTAB 12, 074201 (2009).

This is transverse momentum (sin would be cos for J calc)

This is transmission probability for surface barrier

This is scattering loss factor for bulk transport

This is initial & final state occupation factor

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

<...> in ε are weighted over f(x,k) of emitted electrons, and therefore already in language of Moments

$$\langle O(x,k_x)\rangle \equiv \int d\mathbf{r}\,d\mathbf{k}\,O(x,k_x)\,f(\mathbf{r},\mathbf{k})$$

THERMAL EMISSION

No photons	$\hbar\omega = 0$
Uniform emission	$2\langle x^2\rangle = \langle\rho^2\rangle = \rho_c^2$
Richardson Approx.	$D(k) = \Theta(E(k)-\mu-\phi)$
No Scattering	$f_\lambda(x,p) = 1$
Maxwell-Boltzmann f(x,k)	$D(k)f(k) \propto \exp\{-(E(k)-\mu)/k_BT\}$
No "Final state" issues	$1-f_{FD}(E) \Rightarrow 1$

$$\varepsilon_{n,rms}(thermal) = \frac{\hbar}{mc}\sqrt{\langle x^2\rangle\langle k_x^2\rangle} = \frac{\hbar}{mc}\left(\frac{\rho_c}{2}\right)\left(\frac{M_2}{2M_0}\right)^{1/2} = \frac{\rho_c}{2}\left(\frac{k_BT}{mc^2}\right)^{1/2}$$

PHOTO-EMISSION

Photons	$\hbar\omega > 0$
Uniform emission	$2\langle x^2\rangle = \langle\rho^2\rangle = \rho_c^2$
JWKB Approx.	$D(k) = D_{JWKB}(E,F)$
Scattering	$f_\lambda(x,p) = x/(x+p(E))$
Schottky Lowering	$\phi = \Phi - \sqrt{q^2F/4\pi\varepsilon_0}$ leading order (metal)

$$\varepsilon_{n,rms}(photo) = \frac{\hbar}{mc}\left(\frac{\rho_c}{2}\right)\left(\frac{M_2}{2M_0}\right)^{1/2} \approx \frac{\rho_c}{2}\left[\frac{(\hbar\omega-\phi)}{3mc^2}\right]^{1/2}$$

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

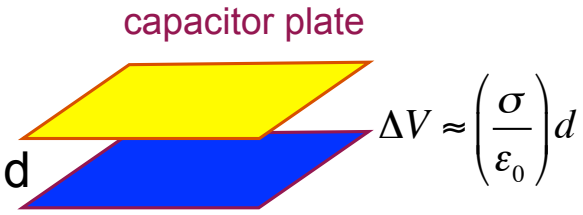
Semiconductors larger ε due to p small, but D also has impact

WORK FUNCTION AND CRYSTAL FACE

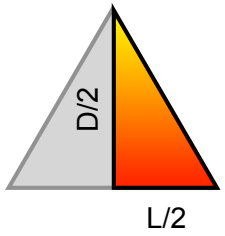
Arrangement of surface atoms matter:

- Smoluchowski : Charge migration on atoms = dipole

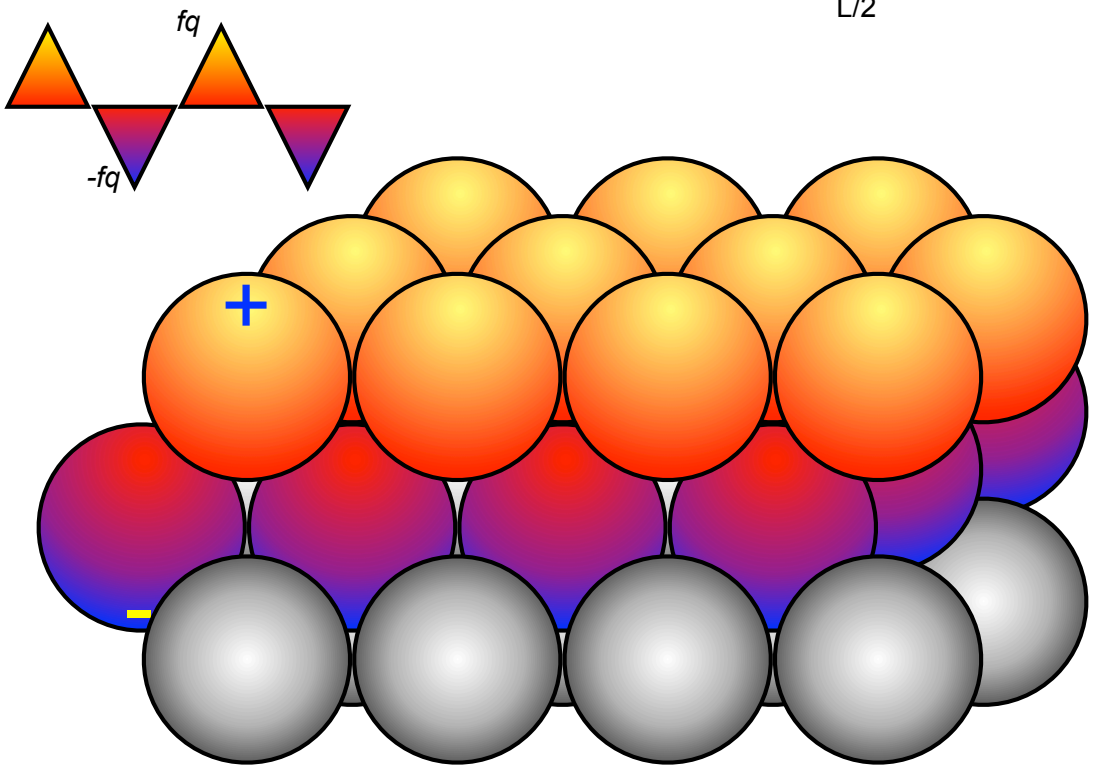
  R. Smoluchowski. "Anisotropy of the Electronic Work Function of Metals." Phys. Rev. 60, 661 (1941)
  (present representation is brutally simplified representation of an elegant study)

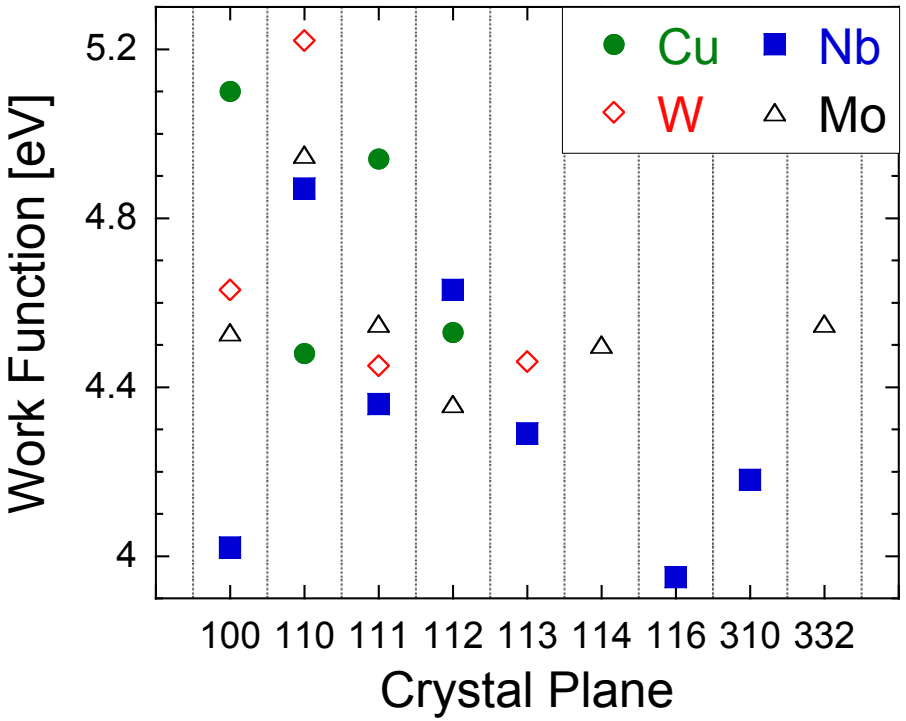


$$\Delta\Phi \approx \left(\frac{q^2}{\varepsilon_0}\right)\frac{fD}{L^2}$$



- f - fractional charge per atom
- D - distance between top and image charge layers
- L - distance between *fq* charges
- f is a fraction e.g. 1/200; L & D ≈ atom diameter.
- ΔΦ ≈0.4 eV typical for differing crystal faces





GYFTOPOULOS-LEVINE THEORY FOR COATINGS

Φ(θ) Due To Dipoles & Electronegativity

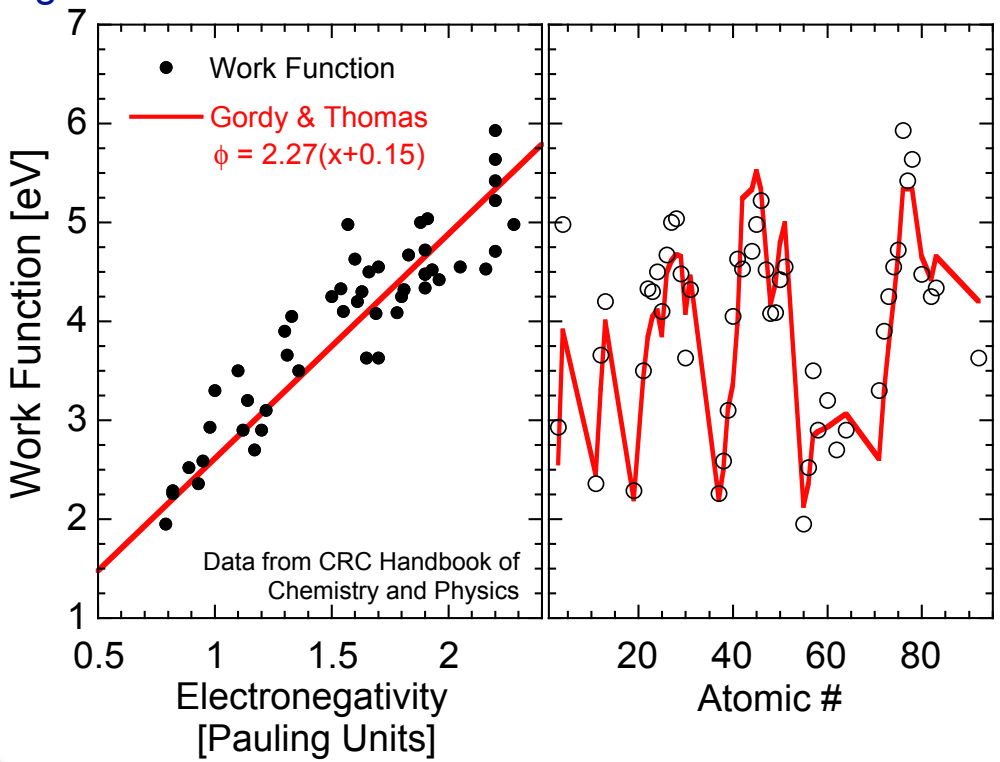
GL Theory Predicts Φ(θ) Due To Partial Monolayer Using Hard-sphere Model Of Atoms (Covalent Radii)

Definition Of Terms

- ϕ_c, ϕ_w Work Function (Monolayer & Bulk)
- r_C, r_W Covalent Radii (Monolayer & Bulk)
- θ Fractional Coverage Factor
- $W(\theta)$ **Electronegativity Barrier**
- $d(\theta)$ Dipole Moment Of Adsorbed Atom

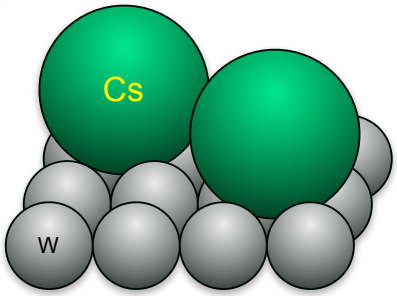
$$\Phi(\theta) = W(\theta) + d(\theta)$$

- Electronegativity - tendency of atoms to attract e-
- Gordy & Thomas: linear approx relating Φ to electronegativity



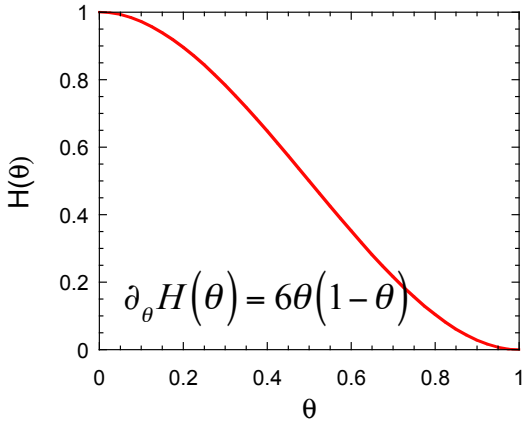
Electronegativity Barrier Term:

$$W(\theta) = \phi_c + (\phi_w - \phi_c) H(\theta)$$
$$H(\theta) = (1 + 2\theta)(1 - \theta)^2$$



H(θ) is simplest polynomial satisfying

$W(0) = \phi_f$	equal to electronegativity of adsorbate
$\partial_\theta W(0) = \phi_f$	subtraction of few atoms doesn't change that
$W(1) = \phi_m$	equal to electronegativity of bulk
$\partial_\theta W(1) = \phi_m$	addition of few atoms doesn't change that



E. P. Gyftopoulos, J. D. Levine, J. Appl. Phys. 33, 67 (1962); J. D. Levine, E. P. Gyftopoulos, Surf. Sci 1, 171 (1964); ibid, p225; ibid p349

DIPOLE TERM

Pauling (paraphrased):

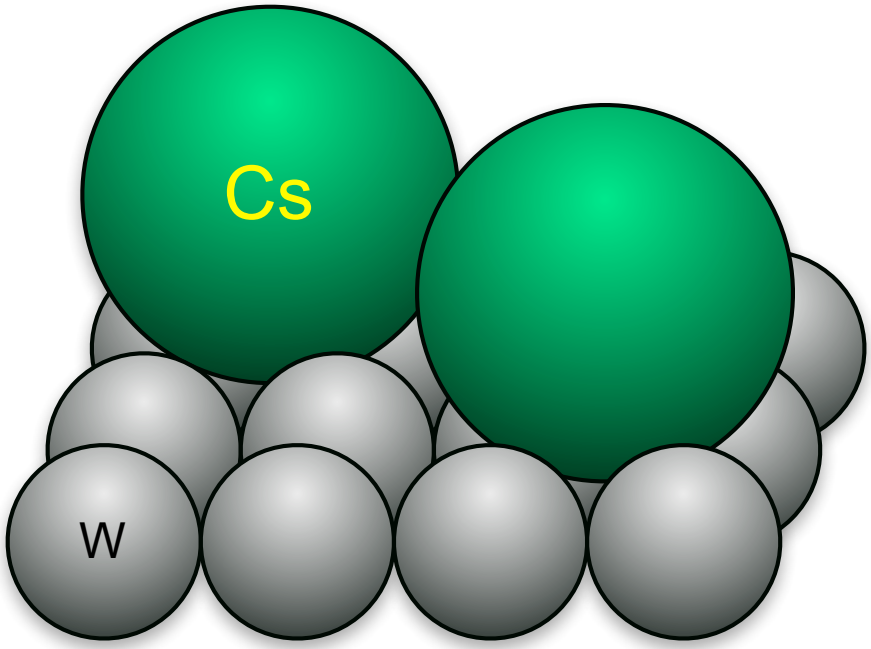
- "Dipole moment of molecule A-B proportional to difference in electronegativities ($\varphi_A - \varphi_B$)"
- Assume true for site of 4 hard sphere atoms in rectangular array with absorbed atom at apex.
- Dipole moment per atom = M(θ)
- **Covalent radii of atom X = r_x**

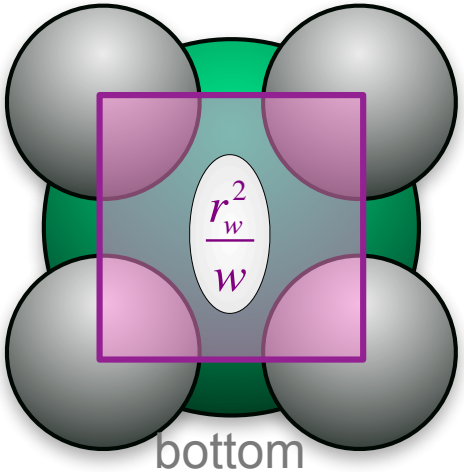
$$M_o = 4\varepsilon_o r_o^2 \cos(\beta)(\phi_w - \phi_c)$$

$$r_o = \sqrt{\frac{k}{2.27\varepsilon_o}} = 0.43653 \text{ nm}$$

constant length scale relating electronegativity and work functions

$$M(\theta) \propto W(\theta) - W(1) \Rightarrow M(\theta) = M_o H(\theta)$$

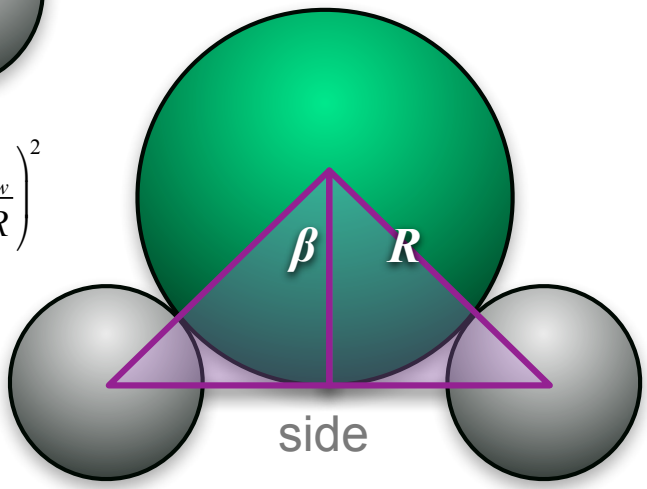




- f = # Cs atoms / unit area
- w = # W atoms / unit area

$$\frac{w}{f}\left(\frac{r_c}{r_w}\right)^2 = \begin{cases} 4 & \text{Cs on W, Mo, Ta…} \\ 2 & \text{Ba on Sr, Th, W…} \end{cases}$$

$$\sin^2\beta = \frac{2}{w}\left(\frac{r_w}{r_f + r_w}\right)^2 = \frac{2}{w}\left(\frac{r_w}{R}\right)^2$$



GYFTOPOULOS LEVINE

Depolarization Effect

- due to other dipoles
- M → M_e ("effective" dipole)
- Depolarizing field E(θ)
- Polarizabity α

$$M_e(\theta) = M(\theta) - \alpha \mathrm{E}(\theta)$$

$$\mathrm{E}(\theta) = \frac{9}{4\pi\varepsilon_0}\left(\frac{f}{4r_c^2}\theta\right)^{3/2} M_e(\theta)$$

$$\alpha = 4\pi\varepsilon_o n r_c^3$$

n = multiple electrons effect

- *n* = 1.00 for Alkali metals (e.g., Cs)
- *n* = 1.65 for Alkaline earth (e.g., Ba)
- Origin: two electrons tends to shield each other from nucleus, so n < 2

Crystal face effect: $f / N_o^{1/2}$ = constant

- N_o = 1 for [100] plane
- N_o = 2 for [110] plane
- N_o = 3 for [B] plane (bumpy)

DIPOLE: product of

- effective dipole moment
- surface density of coating atoms
- coverage factor θ

$$d(\theta) = -M_e(\theta)\left(\frac{f}{4r_c^2}\right)\frac{\theta}{\varepsilon_o} = -\frac{M(\theta)}{1+\alpha\frac{9}{4\pi\varepsilon_0}\left(\frac{f}{4r_c^2}\theta\right)^{3/2}}\left(\frac{f}{4\varepsilon_o r_c^2}\theta\right)$$

COMBINE: get Φ as a function of θ

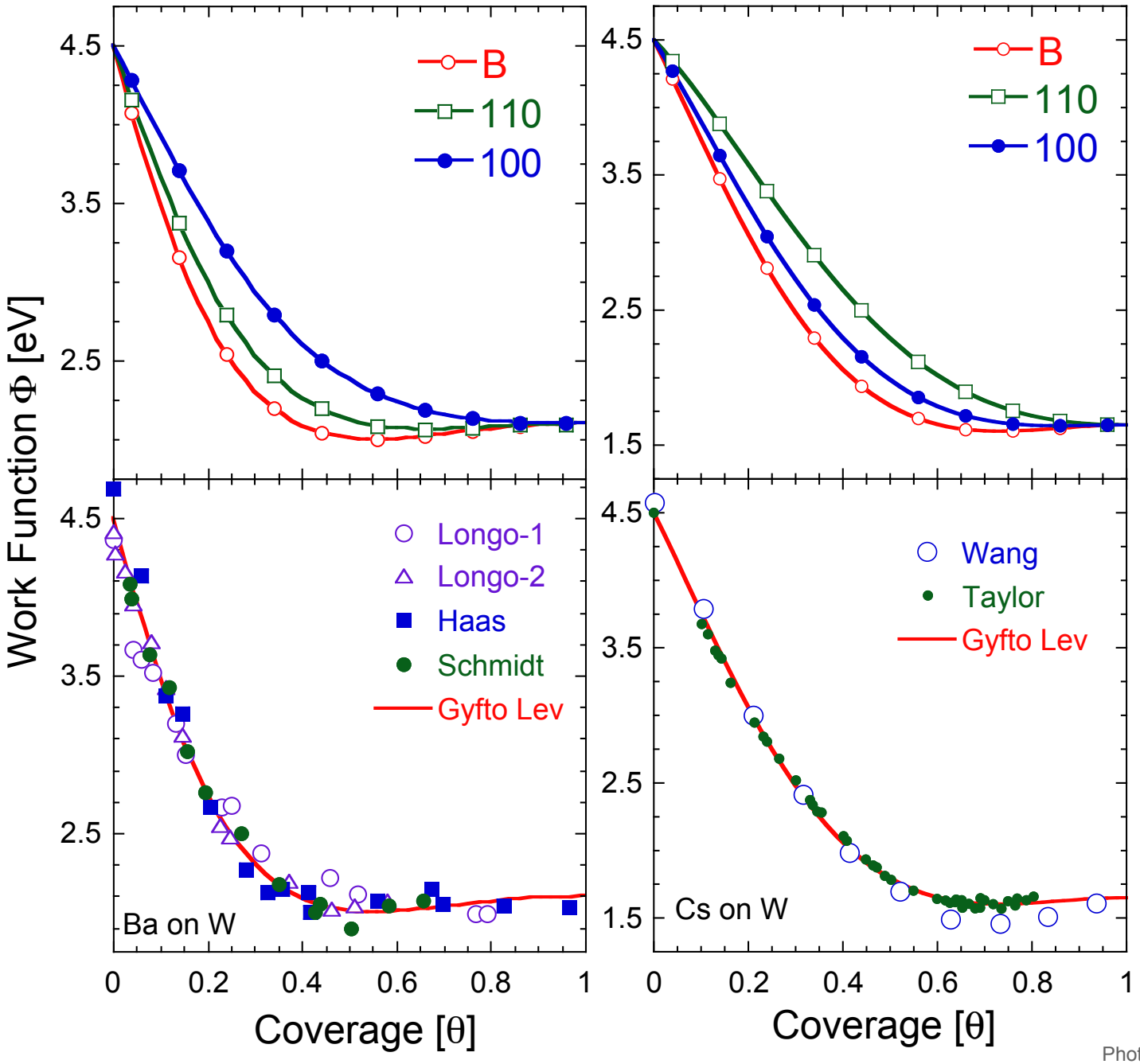
(The Modified Gyftopoulos-Levine Model)

$$\Phi(\theta) = \phi_f - (\phi_f - \phi_m)\left[(2\theta+1)(1-\theta)^2\right]\{1-G(\theta)\}$$

$$G(\theta) = \frac{\left(\frac{r_o}{r_C}\right)^2\left(1-\frac{2}{w}\left(\frac{r_W}{R}\right)^2\right)}{\left(1+n\left(\frac{r_C}{R}\right)^3\right)\left(1+\frac{9n}{8}(f\theta)^{3/2}\right)} f\theta$$

f is only "unknown" and even then, it is tightly constrained

LEAST SQUARES AND DATA FROM LITERATURE



Same f-factor and least-squares analysis of experimental data brings different data sets into excellent agreement

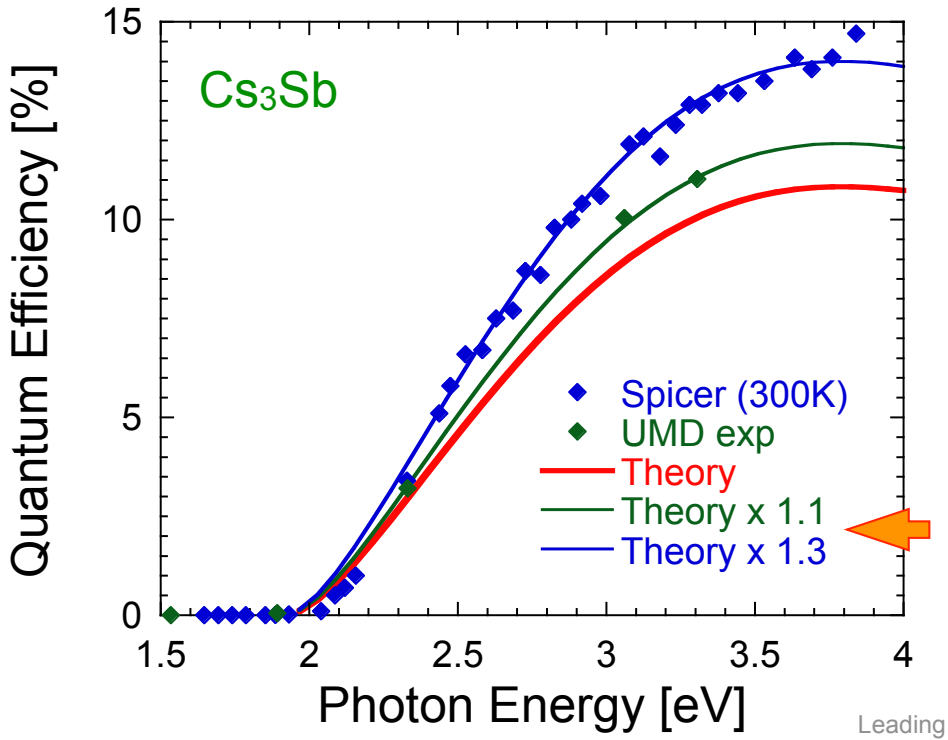
Impact of changing f: as f gets larger, a larger dip occurs in work function

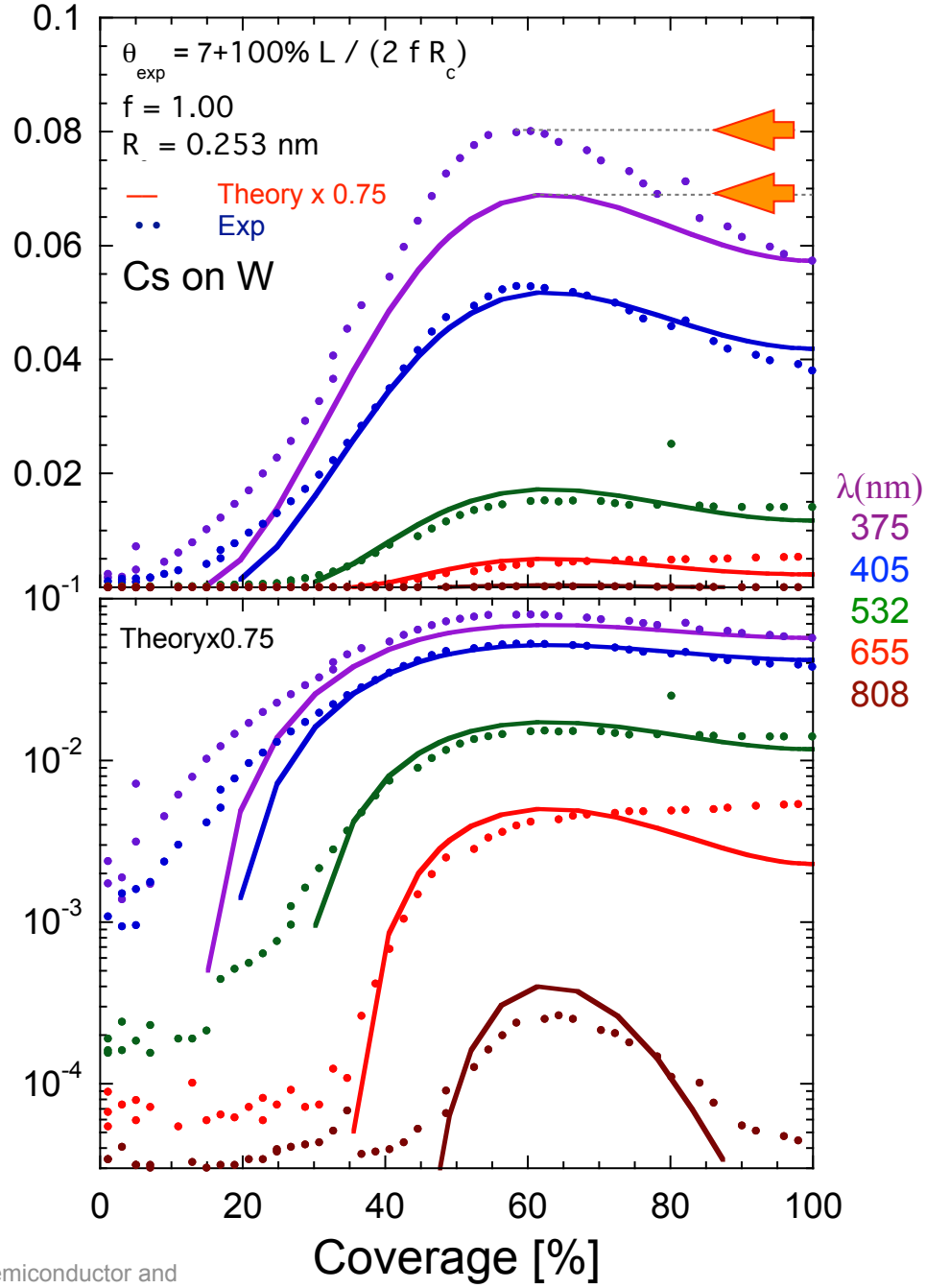
Wang data - Cs only (not Cs-O)

LIMITS OF "FATAL" APPROXIMATION IN SCATTERING

$$f_\lambda\left(\cos\theta, \delta/l\right) = \frac{\int_0^\infty e^{-(x/\delta)-(x/l\cos\theta)}\,dx}{\int_0^\infty e^{-(x/\delta)}\,dx} = \frac{\cos\theta}{\cos\theta + \dfrac{\delta(\hbar\omega)}{l(E)}}$$

- "Fatal" approximation: ***l(E)*** is interpreted as product of velocity and scattering time - a collision removes electron from emission distribution
- **QUESTION:** Does "Fatal" Approx cause slight observed differences between theory and experiment... like these?





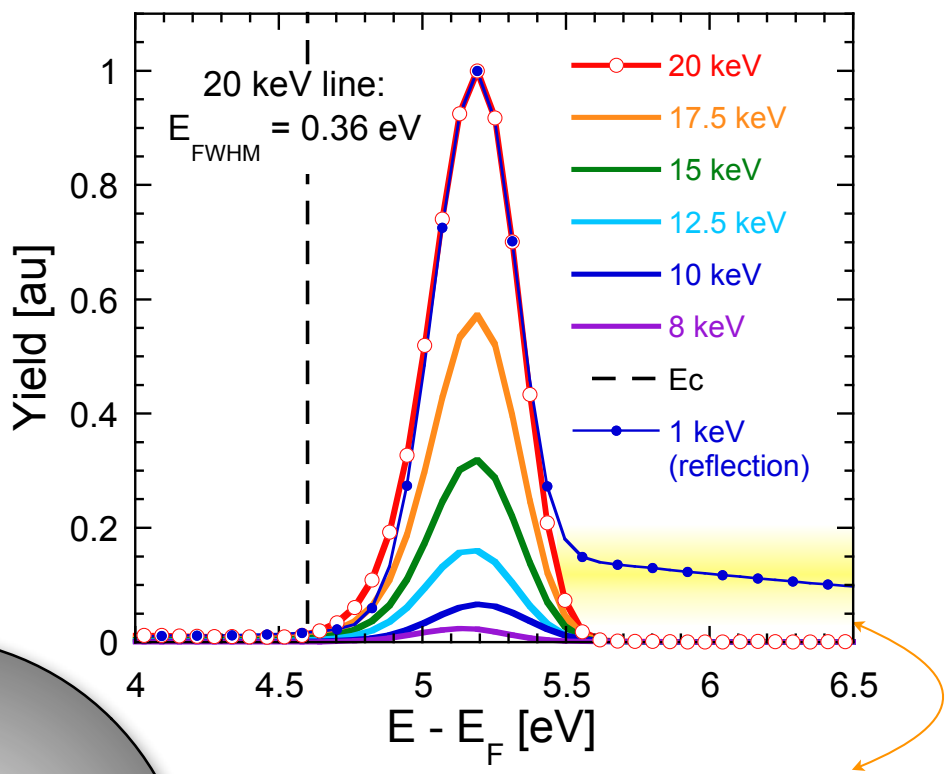
Leading Order Semiconductor and Metal Emission Models in MICHELLE

SCATTERING HAPPENS.

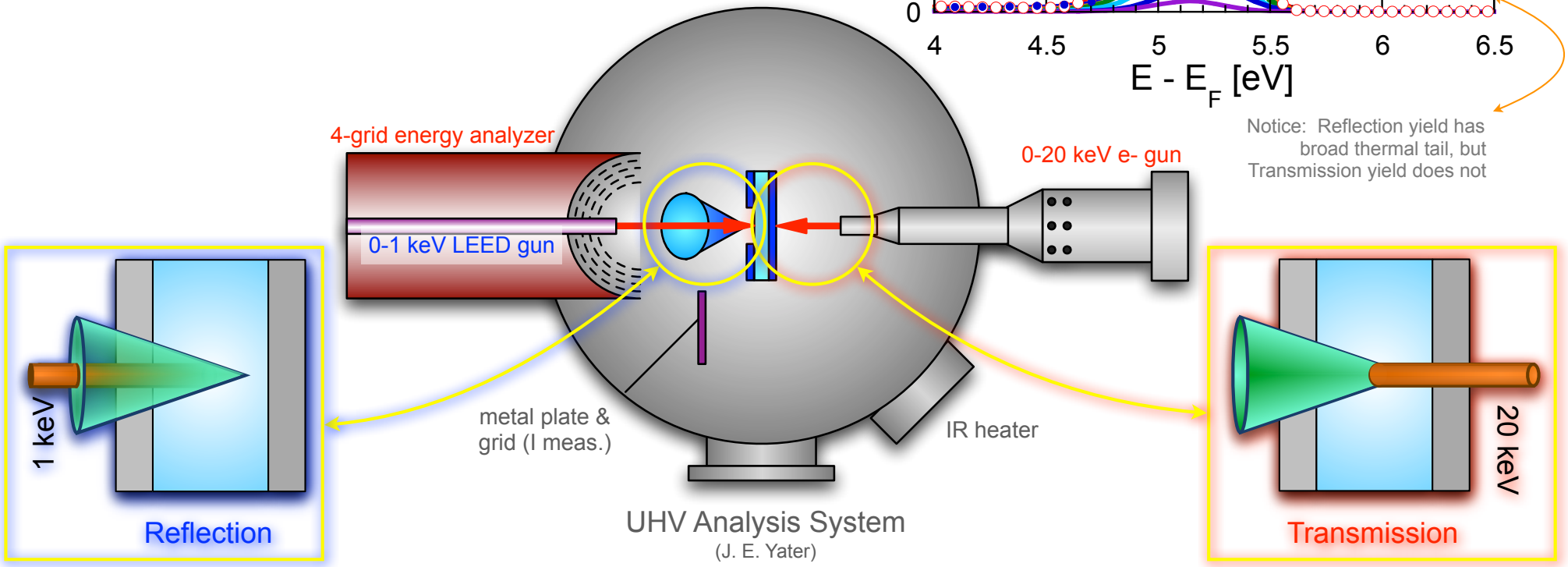
Experimental Measurements on Diamond Film

Yater, Shaw, Pate, Butler, Myers, Fegelyson

- Reflection & Transmission yields from thin single crystal diamond film of **8.3 μm**; No bias across diamond film
- Penetration Range = 15.4 nm (1 keV); 3 μm (20 keV)
  (theoretical estimate using Bethe energy loss equation)
- Identical peak position and width
  - Confirms transmission from conduction band
- No high-energy tail in transmission peak
  - Completely thermalized distribution
- Change of Peak Height with Incident Energy:
  acts as though a zero-field decay time δt = 50 ps exists such that electron lifetime goes as P(t) α exp(-t/δt)



Notice: Reflection yield has broad thermal tail, but Transmission yield does not



UHV Analysis System
(J. E. Yater)

"FATAL" ABSTRACTION

Scattering Will Not Be Ignored.
Find Its Impact Using Monte Carlo:

- r = random #
- Flight Duration - Probability Of Scattering

$$t_r = -\tau \ln(r)$$

$1/\tau$ = Sum Of All $1/\tau_j$

- Scattering Events:

$$1 > P_n \propto \sum_{j=1}^{n} \tau_j^{-1}$$

τ_j are various scattering mechanisms, like e-e, acoustic, optical phonon, etc.

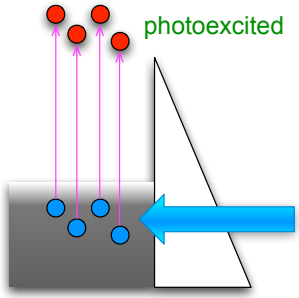
  - Random # r compared to P_n: value sets scattering mechanism;
  - if r > all P_n, then "free flight" (Also called "Self-scattering")

- Determine New Velocity Vector (Mag & Dir)
- After Scattering, Particle Trajectory Determined By Solution To 3D Eq Of Motion
- Repeat For N Particles ("Synchronous Ensemble")
- When Time Exceeds Preset Time Interval, Write Out Data (Positions And Velocities)

Photoemission:

electrons start with energy > barrier height

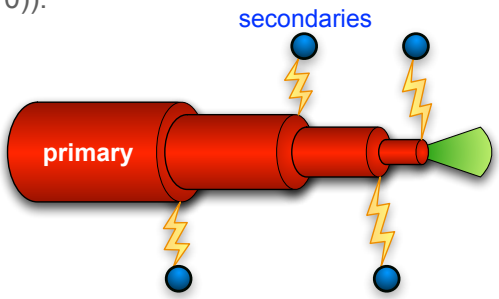
$$E = \mu + \phi + r(\hbar\omega - \phi)$$



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

Secondary Emission

(Jensen et al,, JAP108, 044509 (2010)):
primary with energy E_o generates N_p Secondaries



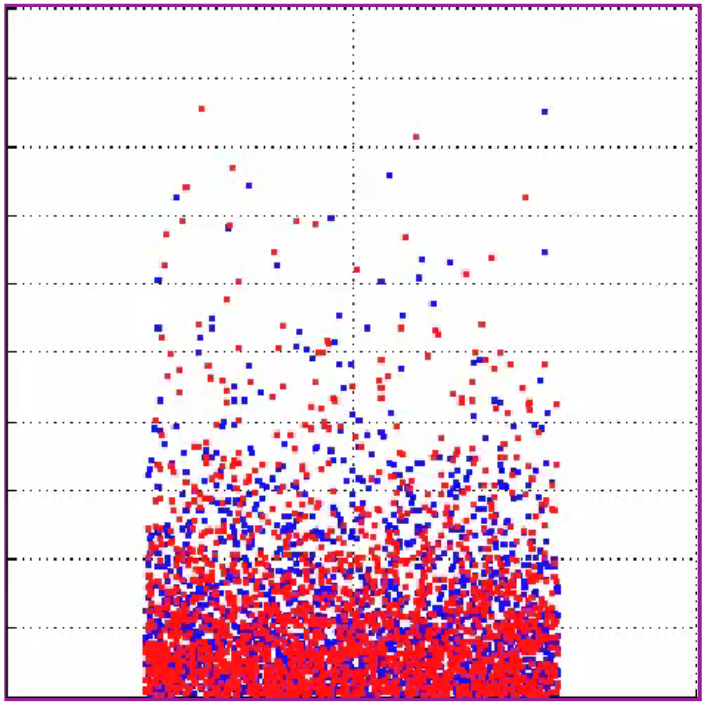
- $N_p = E_o/\Delta E$
- velocity perpendicular to incident
  K. Murata, D. Kyser
  Adv. Elect. El. Phys 69, 175 (1987)
- spaced as $z_{j+1} - z_j \approx \Delta E \left(\partial_z E\right)^{-1}\Big|_{z=z_j}$
- all N_p secondaries appear to be generated simultaneously to a good approximation
- Primary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

COPPER: SIMULATION

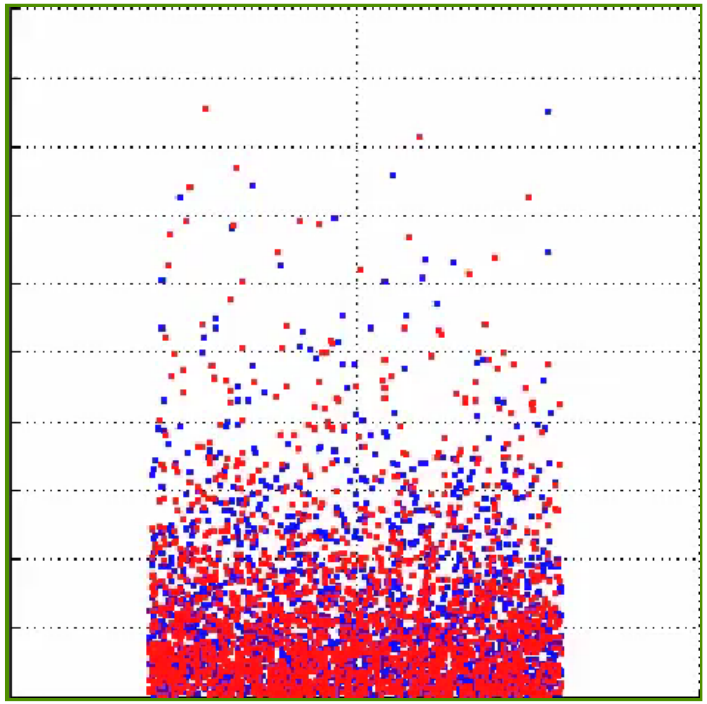
- Start with 4000 electrons
- Laser penetration depth = 12 nm, incident from bottom of frame;
- λ = 266 nm, **Φ = 4.5 eV,** F = 50 MV/m, T = 300 K
- Time Step = 0.2 fs / frame, 100 frames
  Sim Region 20 nm x 20 nm
- When electron energy drops below barrier height $E < \mu + \Phi - (4QF)^{1/2}$, electron is removed from simulation visualization

- Left: "Fatal Scattering" Approx
- Right:
  - Acoustic changes electron direction
  - e-e bleeds away energy by creating 2nd electron & sharing KE, final direction is along one of 6 axes wrt incident direction
- Red electrons travel towards surface (down)
  Blue electrons travel away from surface (up)

caveat: pseudo-Monte Carlo



Approx: Scattering is fatal



Approx: Scattering redirects & shares energy

If initial velocities of two colliding e- are p_z & q_z, where +z is direction of incident electron p_z and q_z is velocity of any allowed second electron, then:

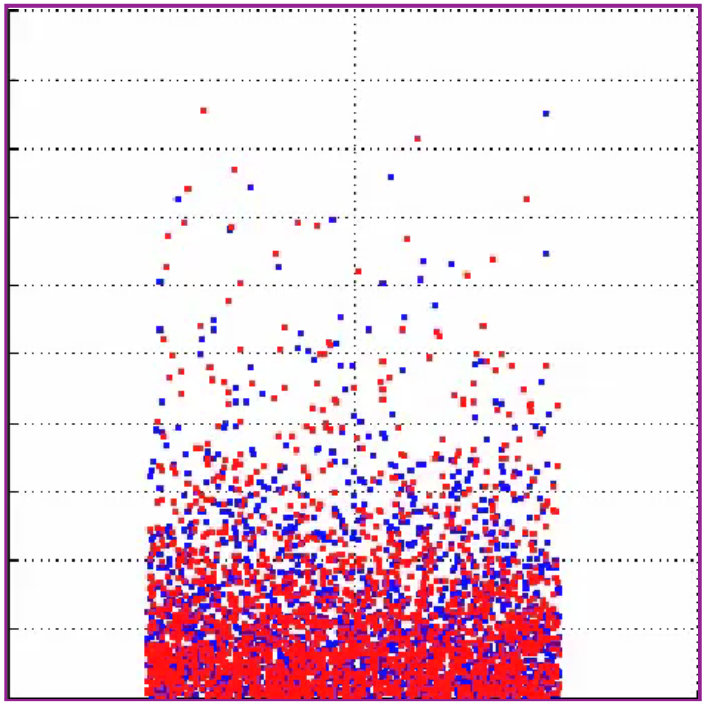
- $p' = (p_z\, q_z)^{1/2}$
  $q' = (p_z^2 + q_z^2 - p_z q_z)^{1/2}$
  and $\theta = 0$ (π reverses role)
  with 33% Probability
- $p' = p_z$
  $q' = q_z$
  and $\theta = \pi/2$
  with 67% Probability

CESIUM ON COPPER: SIMULATION

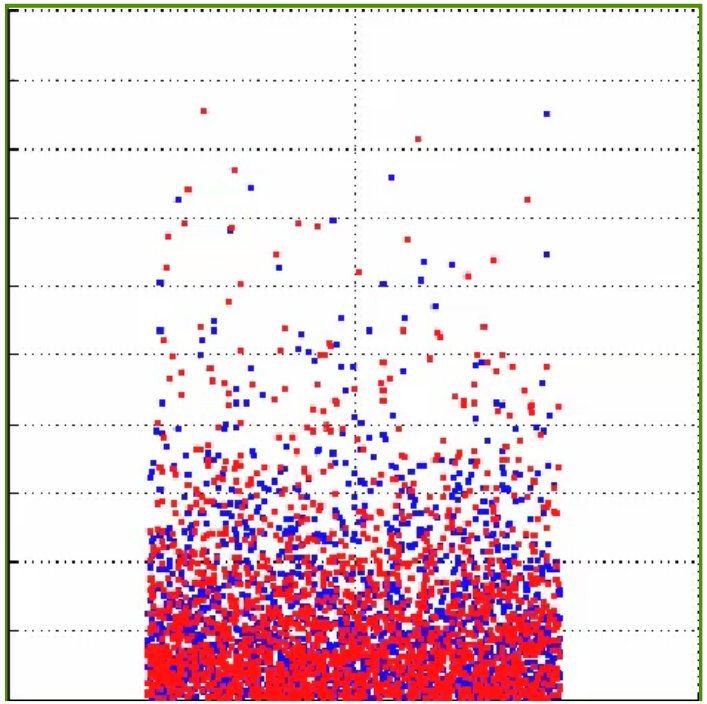
- Start with 4000 electrons
- Laser penetration depth = 12 nm, incident from bottom of frame;
- λ = 266 nm, **Φ = 1.6 eV,** F = 50 MV/m, T = 300 K
- Time Step = 0.2 fs / frame, 100 frames
  Sim Region 20 nm x 20 nm
- When electron energy drops below barrier height $E < \mu + \Phi - (4QF)^{1/2}$, electron is removed from simulation visualization

- Left: "Fatal Scattering" Approx
- Right:
  - Acoustic changes electron direction
  - e-e bleeds away energy by creating 2nd electron & sharing KE, final direction is along one of 6 axes wrt incident direction
- Red electrons travel towards surface (down)
  Blue electrons travel away from surface (up)

caveat: pseudo-Monte Carlo



Approx: Scattering is fatal



Approx: Scattering redirects & shares energy

If initial velocities of two colliding e- are p_z & q_z, where +z is direction of incident electron p_z and q_z is velocity of any allowed second electron, then:

- $p' = (p_z\, q_z)^{1/2}$
  $q' = (p_z^2 + q_z^2 - p_z q_z)^{1/2}$
  and $\theta = 0$ (π reverses role)
  with 33% Probability
- $p' = p_z$
  $q' = q_z$
  and $\theta = \pi/2$
  with 67% Probability

CS AND CU: EMITTED CHARGE

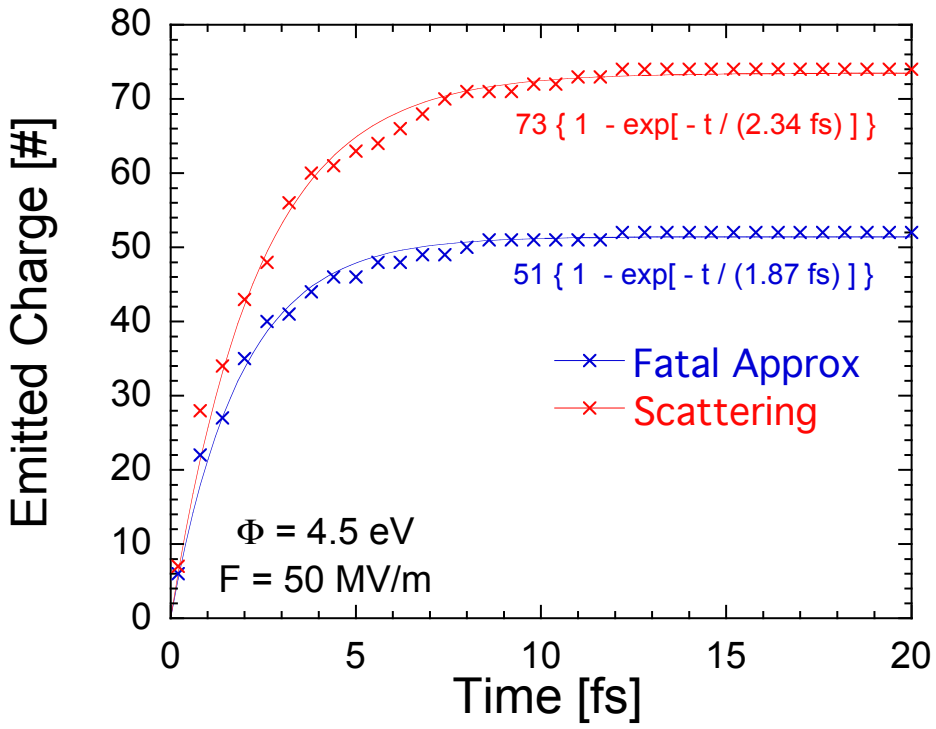
After e-e scattering, primary e shares energy with scattered e. If photon energy is...

- ...well above barrier: more likely both electrons have energy E > barrier.
- ...near barrier: more likely sharing energy results in neither electron having E > barrier
- ...in between: may be primary (more energetic) electron still has E > barrier after scattering

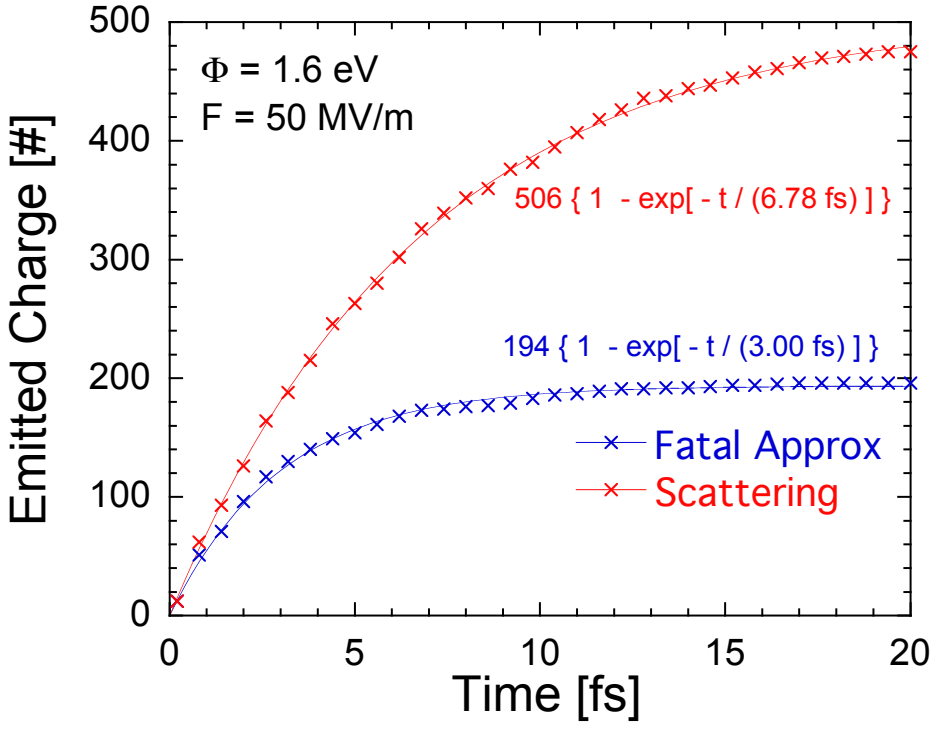
Therefore, expect that the larger (Photon Energy - Barrier Height above Fermi Level) is, then:

- Time constant of emitted charge Q(t) will be longer
- Ratio of Q_o(Scattering) / Q_o(Fatal) will be larger

$$Q(t) \approx Q_o \left\{1 - \exp\left(-t / \Delta t\right)\right\}$$



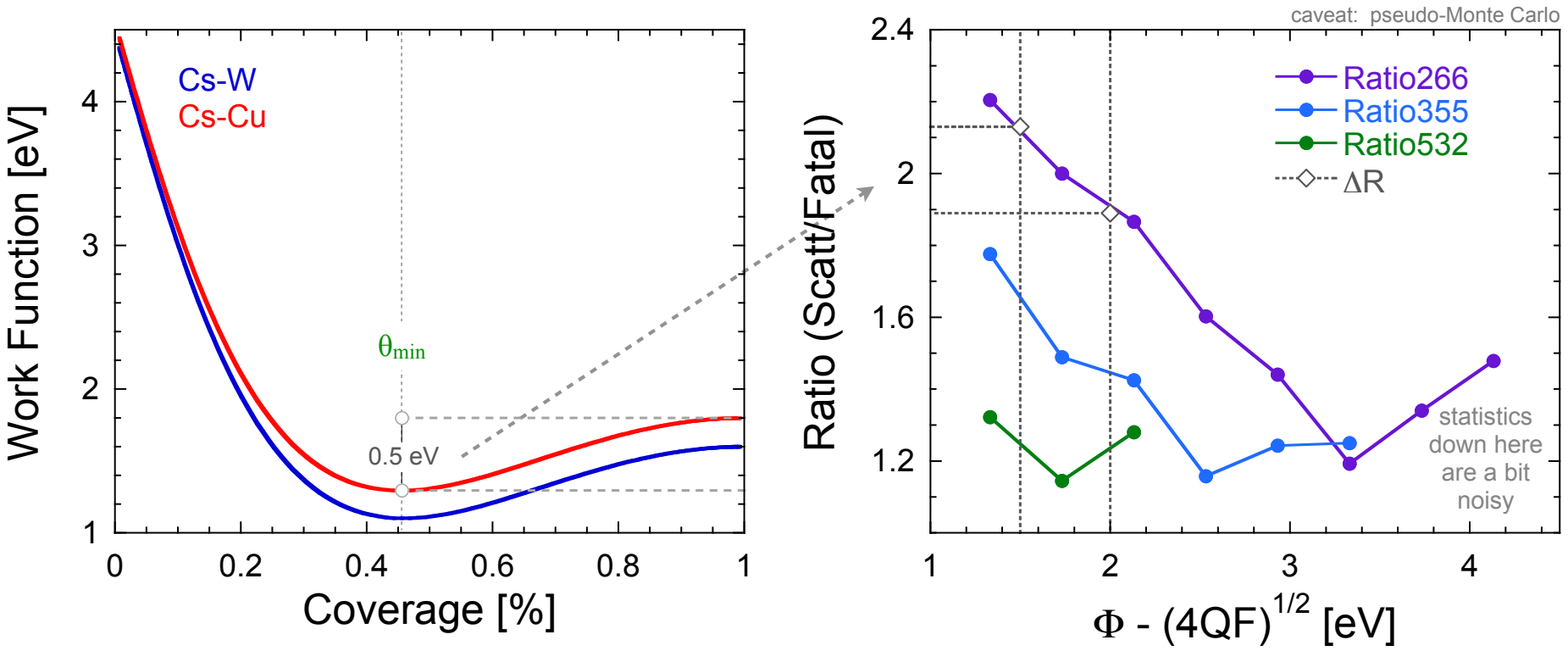
Cu



Cs-Cu

WORK FUNCTION VARIATION

- Gyftopoulos-Levine Theory: $\Phi(\theta_{min}) - \Phi(1) \approx 0.5$ eV
  - Non-fatal approximation will increase peak to monolayer difference in QE for higher energy photons.
  - Shorter λ QE will increase overall compared to longer λ QE
- **Conclusion**: some peak to monolayer difference between Theory & Experiment due to pessimism of "Fatal" Approximation
  (previously attributed solely to 2D surface density factor *f*)



SUMMARY

Approximation or Feature	"Standard" Models	Caveat
Reflectivity and Laser Penetration Depth	Metals: tabulated optical properties Semiconductors: tabulated properties or Drude-Lorentz Model of Optical constants	No approximation to tabulated data, but sometimes limited in energy range; DL approach ≈ ok if enough terms
Scattering Factor and Bulk Transport of electrons	Fatal Approximation (any scattering event removes electron from emitted distribution); Constant or convenient relaxation time τ; Semiconductors: α-semiconductor model for some parameters	FA better when $hf \approx \mu + \Phi$ or E_g+E_a; otherwise, neglects e- that scatter *into* emission cone (not just *out* of it): τ has complex E & T dependence; SM tends to under-predict QE
Transmission Probability	D(E) is step function in energy (metals - FD distribution also treated as step function in energy)	D(E) better represented as tanh function: distribution in energy of emitted e- can be larger than implied by step function D(E); SM tends to over-predict QE
Uniformity of emitter surface / emission	Φ or E_a does not vary; T is constant; surface is uniform and flat; no local field enhancement effects	Scattering terms sensitive to T; Emission barrier varies significantly over different crystal faces + surface coverage with Cs + field enhancement from local sites; All affect QE differently

Insofar as emittance can be evaluated using Moments, analogous comments apply