Transfer Matrix Methods, Photoemission, and Heterostructures

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Photocathode Physics for Photoinjectors (P3)

Theory and Simulation

Sections Outline

Photocathode Metrics

Need

Particle accelerators and light sources such as x-ray Free Electron Lasers (FEL) make severe demands on e-source in terms of J and ability to shape pulse. Some of the Metrics of performance are at odds with each other[a]

[a] † *e.g.*, QE and $\varepsilon_{n,rms}$. See D.H. Dowell, J.F. Schmerge. "*QE and Thermal Emittance...*" Phys. Rev. ST Accel. Beams, **12(7)**, 074201, 2009.

Five metrics of photocathode performance emerge as particularly important for next generation light sources (such as x-ray FEL's):

1. *Quantum Efficiency* Number of e^- emitted / number of $\hbar\omega$ absorbed→ big is good.
2. *Emittance* Tendency of e-beam to spread as beam propagates→ small is good.
3. *Lifetime* of photocathode before replacement / rejuvenation→ long is good.
4. *Ruggedness* or survivability in photoinjector† → tough is good.
5. *Response Time* of photocathode, affects bunch pulse shaping→ fast is good.

Cesium

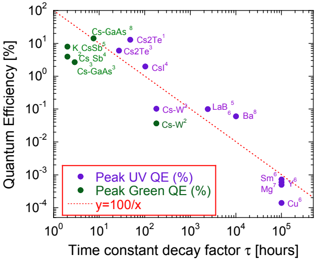
All high QE photocathodes ↔ semiconductors. All metrics concerned with e-transport through bulk material + emission over (or through) surface barrier. Band bending, barriers, and resonances require more than usual Heaviside transmission probability.

Photocathode Materials

Desirable Photocathode Performance: $J_{peak} = 20 - 500$ A/cm^2, $\langle J \rangle \approx 1$ A/cm^2, Minimize emittance

Charge per bunch: 0.1 - 1 nC. Pulse length: 1 - 50 ps. Pulse to pulse: > 5 ns. Area: 0.1 cm^2

- harmonic: change of $\lambda_o = 1.06\ \mu$m to shorter λ using ω-doubling crystals such that $\lambda = \lambda_o / n$.
- Efficiency: conversion efficiency of crystals used for frequency doubling
- Temp Resp: temporal response time, or how long photoexcited electrons take to come out
- Ave P at cath: laser power deposited on cathode
- Ave P Laser: Power of 1064 nm Nd:YAG before conversion
- 8760 hours: one year (metal photocathodes last longer)



Property	Units	K_2CsSb	Cs_2Te	GaAs	Cu	Mg
harmonic	–	2	4	2	4	4
Wavelength	nm	532	266	532	266	266
Efficiency	%	50	10	50	10	10
Starting QE	%	8	5	5	1.4E-2	6.2E-2
Lifetime	hrs	4	> 100	58	> 8760	> 8760
Temp Resp	ps	< 1	< 1	> 50	< 0.05	< 0.05
Vac tol	–	poor	very good	poor	excellent	excellent
Ave P at cath	W/cm^2	29.13	93.22	46.61	33293	7518
Ave P Laser	W/cm^2	58.26	932.20	93.22	332932	75178

The Three Components of Photoemission

Absorption

...of light in bulk material and photo-excitation of e^-

- reflectivity $R(\omega)$ and penetration depth $\delta(\omega)$
- ω-dependent dielectric constant: optical parameters n and k

Transport

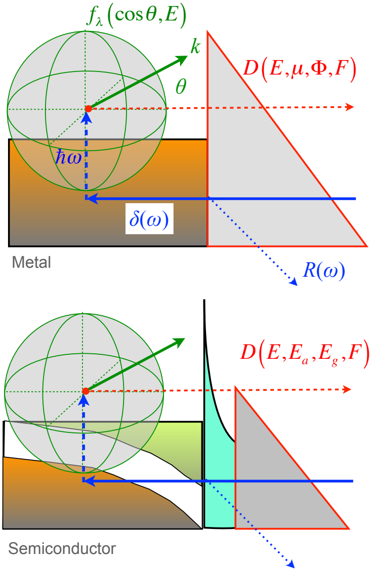
... of photo-excited e^- to surface with scattering

- electron energy E
- scattering rates (relaxation times) $\tau(E)$: scattering factor f_λ

Emission

...probability of transport over/thru barrier

- Metal: Chemical Potential μ Work Function Φ (measured from Fermi level)
  Semiconductor: barrier height E_a (measured from conduction band minimum), band gap E_g
- Escape cone θ_{max} (or k_m in Moments model)



Assumptions Behind The Three Step Model

The Moments Model makes the same five simplifications behind 3-step model of Berglund and Spicer[a]

1. photoexcited e^- isotropically distributed;
2. only inelastic scattering events;
3. inelastic scattering ↔ mean free path, depends only on electron's energy;

$$l(E) = \frac{\hbar k(E)}{m}\tau(E) \quad (1)$$

4. isotropic scattering
5. normal energy $E_x = E\cos^2\theta$ of photoexcited $e^- >$ barrier height V_o with

$$P(E > V_o) = 1 \quad (2)$$

This is the Heaviside Step Function emission probability.

[a] C.N. Berglund, and W.E. Spicer. "*Photoemission Studies of Copper and Silver: Theory*." Physical Review **136(4A)**, A1030, 1964.

Preparing for the Moments Approach

Phase Space distribution approach

- Particles (like e^-) are labeled by their position $\vec{r}$ and their momentum $\hbar\vec{k}$
- Assume conditions $\perp$ to $\vec{J}$ are uniform. The 6D phase space $f(\vec{r},\vec{k})$ can therefore be reduced to a 2D phase space in x and $k_x \equiv k$, or $f(x,k)$.
- The phase space points are conserved: $df/dt = 0$, or

Boltzmann's Transport Equation

$$\frac{df}{dt} = \frac{f(x+dx, k+dk, t+dt) - f(x,k,t)}{dt} \Rightarrow \left\{\frac{\partial}{\partial t} + \frac{\hbar k}{m}\frac{\partial}{\partial x} + \frac{F}{\hbar}\frac{\partial}{\partial k}\right\} f(x,k,t) = 0 \tag{3}$$

Integration over dk results in Continuity Equation; (∂_k-term vanishes on BC)

$$\frac{\partial}{\partial t}\left[\frac{1}{2\pi}\int_{-\infty}^{\infty} f(x,k,t)dk\right] = -\frac{\partial}{\partial x}\left[\frac{1}{2\pi}\int_{-\infty}^{\infty}\left(\frac{\hbar k}{m}\right) f(x,k,t)dk\right] \tag{4}$$

$$\frac{\partial}{\partial t}\rho(x,t) = -\frac{\partial}{\partial x} J(x,t) \tag{5}$$

$\rho(x,t) \leftrightarrow 0^{th}\ k$–moment of $f(x,k)$; $J(x,t) \leftrightarrow 1^{st}\ k$–moment of $f(x,k)$

Defining the Transmission Probability

- In the Quantum Mechanical version of the Continuity Equation, density ρ and current density j become pure-state (particular k) operators; time derivatives are commutators with the Hamiltonian $\hat{H}$

$$\partial_t \hat{\rho}_k(t) = \frac{i}{\hbar}\left[\hat{H}, \hat{\rho}_k(t)\right] = -\frac{\hbar}{2m}\frac{\partial}{\partial \hat{x}}\left\{\hat{k}, \hat{\rho}_k(t)\right\} = -\frac{\partial}{\partial \hat{x}}\hat{j}_k(t) \tag{6}$$

- Pure state current density $j_k(x,t)$: Anti-commutator with $\hat{k}$

$$j_k(x,t) = \frac{\hbar}{2m}\langle x|\left\{\hat{\rho}_k(t), \hat{k}\right\}|x\rangle = \frac{\hbar}{2mi}\left\{\psi_k{}^{\dagger}\partial_x\psi_k - \psi_k\partial_x\psi_k{}^{\dagger}\right\} \tag{7}$$

Transmission probability is ratio of current density for a given k

$$D(k) = \frac{j_{trans}(k)}{j_{incident}(k)} \tag{8}$$

A Simple Barrier Model

Rectangular Barrier of height $V_o = \hbar^2 k_o^2 / 2m$ and width L

$$D_{rec}(k) \equiv \left\{ 1 + \left[\left(\frac{k_o^2}{2k} \right) \frac{\sinh\left(L\sqrt{k_o^2 - k^2} \right)}{\sqrt{k_o^2 - k^2}} \right]^2 \right\}^{-1} \quad (9)$$

- Gamow Factor:

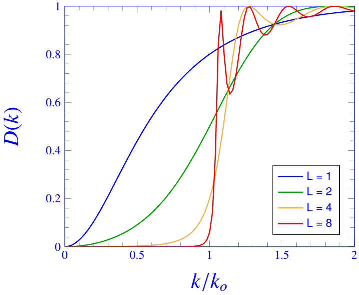
$$\theta(k) = 2L\sqrt{k_o^2 - k^2} \quad (10)$$

$$\rightarrow 2\int_{x_-}^{x_+} k(x)\, dx \quad (11)$$

- Kemble form of D(k):

$$D(k) \approx 1/\left\{1 + \exp\left[\theta(k)\right]\right\} \quad (12)$$

- $D(k > k_o)$ oscillates. Oscillations? Structure? "Simple" is not so simple.



Tsu-Esaki-like Formula for Current Density

A general expression for Current Density composed of

- A velocity: $\hbar k/m$
- A weighting factor or supply function: $f(k)$
- A probability of transmission past the barrier: $D(k)$

$$J(F,T) = \frac{1}{2\pi}\int_0^\infty \frac{\hbar k}{m} D(k) f(k)\, dk \tag{13}$$

Convert: $k \to E(k) = \hbar^2 k^2/2m$ (parabolic relation)

$$J(F,T) = \frac{1}{2\pi\hbar}\int_0^\infty D(E) f(E)\, dE \tag{14}$$

$f(k)$ = supply function, from Fermi-Dirac Distribution (μ = Fermi level; $\beta = 1/k_B T$)

$$f(k) = \frac{2}{2\pi^2}\int_0^\infty \frac{2\pi k_\perp dk_\perp}{1+\exp\left[\beta(E+E_\perp-\mu)\right]} = \frac{m}{\pi\beta\hbar^2}\ln\left[1+e^{\beta(\mu-E)}\right] \tag{15}$$

Sections Outline

Recast QE as Moment of Distribution Function

Moments are integrals of k_j^n with a distribution:

$$M_n\left(\tilde{k}_j\right) = \frac{1}{(2\pi)^3}\int d\vec{k}\left[\tilde{k}_j^n\right]\times[E]\times[T]\times[A]$$

The Parts

- Phase Space Element
- Momentum component
- Absorption (occupation)
- Transport (scattering factor)
- Emission (transmission prob)

$$d\vec{k} \rightarrow \left(\frac{2m}{\hbar^2}\right)^{3/2}\int_0^\infty E^{1/2}dE\int_0^{\pi/2}\sin\theta d\theta$$

$$[\tilde{k}_j^n] \rightarrow \left\{\frac{2m}{\hbar^2}\tilde{E}\cos^2\theta\right\}^{n/2}$$

$$[A] \rightarrow \left\{\begin{array}{c} f_{FD}(E)\,(1-f_{FD}(E+\hbar\omega)) \\ \Theta\left(\hbar\omega+E-E_g\right)\end{array}\right.$$

$$[T] \rightarrow f_\lambda(\cos\theta, p(E))$$

$$[E] \rightarrow D\left\{(E+\hbar\omega)\cos^2\theta\right\}$$

f_{FD} is Fermi-Dirac distribution; E_g is band gap; Θ is Heaviside step function; $\tilde{k}$ and $\tilde{E}$ indicates momentum / energy moment calculated with / without $\hbar\omega$; p is a ratio between the laser penetration depth δ and the mean free path $v\tau$

Transport and the effects of Scattering

The "Fatal" Approximation: Common approximation that scattering prevents emission

- Good for big barriers (metals), less good for small barriers & semiconductors.
- Important factor: Ratio of penetration depth to distance between scattering events

$$p(E) = \frac{\delta(\omega)}{l(E)} = \frac{m\delta(\hbar\omega)}{\hbar k(E)\tau(E)}; \quad \tau_{total}^{-1} = \sum_j \tau_j^{-1} \quad (16)$$

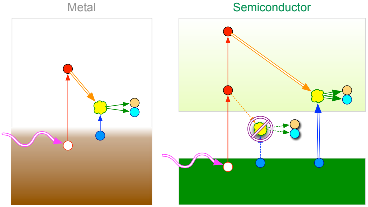
- Fraction of photoexcited electrons surviving transport back to surface

δ governs how far laser light gets *in*; $x/\cos\theta$ governs how far electrons have to go to get *out*

$$f_\lambda(\cos\theta, p) = \frac{\int_0^\infty \exp\left(-\frac{x}{\delta} - \frac{x}{l(E)\cos\theta}\right)dx}{\int_0^\infty \exp\left(-\frac{x}{\delta}\right)dx} = \frac{\cos\theta}{\cos\theta + p(E)} \quad (17)$$

- Final states have to be allowed: semiconductors have a "magic window" where $e-e$ scattering (large energy loss) is forbidden and only $e-p$ (small energy loss) occurs

- Fraction surviving (modifies FD): $F_\lambda(y) = \int_{\cos\theta_m}^1 x f_\lambda(x,p)\,dx$
- Cs_3Sb-like $(e-p)$: $\rightarrow p \approx 0.36$
  $\delta = 27$ nm; $v/c = 0.8$; $\tau = 31$ fs
- Cu-Like $(e-e)$: $\rightarrow p \approx 2.38$
  $\delta = 12.6$ nm; $v/c = 0.675$; $\tau = 2.6$ fs



QE and Emittance In Terms of Moments

Quantum Efficiency depends on k_z

- $k_z = k\cos\theta$ is momentum component directed into barrier

$$QE = \{1 - R(\omega)\} \frac{M_1(k_z)}{2\,M_1(k)|_{D=1,\,f_\lambda=1}} \propto \begin{cases} (\hbar\omega - \phi)^2 & metal \\ \left(\hbar\omega - E_a - E_g\right)^\nu & semi^a \end{cases} \quad (18)$$

normalized Emittance $\varepsilon_{n,rms}$ depends on $k_\perp$

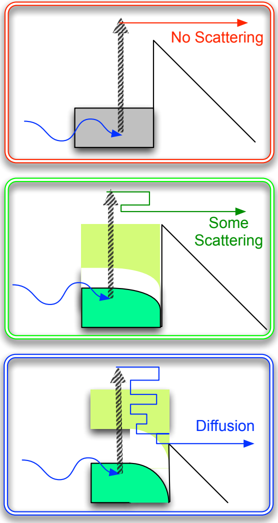
- Emittance $\varepsilon_{n,rms}$ is area in phase space enclosing all (x, k_x) of e^- in a beam.
  $\varepsilon_{n,rms} = (\hbar/mc)\sqrt{\langle x^2\rangle\langle k_x^2\rangle}$ for rotationally symmetric uniform emission areas
  $k_\perp^2 = k_\rho^2 + k_\omega^2$; $\rho_c^2 = 2\langle x^2\rangle$ $k_\perp$ along surface; ρ_c is radius of emission area
- Reproduces Dowell-Schmerge formula[b] for $\varepsilon_{n,rms}$

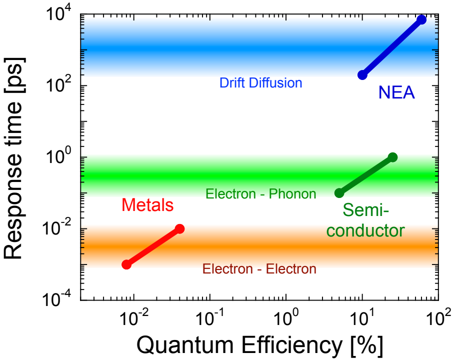
$$\varepsilon_{n,rms} = \frac{\hbar}{mc}\left(\frac{\rho_c}{2}\right)\sqrt{\frac{M_2(k_\perp)}{2M_0(k_\perp)}} \approx \frac{\rho_c}{2}\left[\frac{(\hbar\omega - \phi)}{3mc^2}\right]^{1/2} \quad (19)$$

[a] W.E. Spicer. "*Photoemissive, Photoconductive, and Optical Absorption Studies of Alkali-antimony Compounds.*" Phys. Rev., **112**, 114, 1958
$QE = B/[1 + g(E - V_g)^{-3/2}] \rightarrow \nu = 3/2$. Moments Model recommends differently.

[b] D.H. Dowell, J.F. Schmerge. "*QE and Thermal Emittance....*" Phys. Rev. ST Accel. Beams, **12(7)**, 074201, 2009.

Response Time and Scattering





- Metals, $e - e$ scattering is fatal to emission (shares energy) so only unscattered electrons get to barrier
- Semiconductors, $e - p$ scattering changes E a small amount, so few scatterings do not impede emission
- Negative Electron Affinity: e^- thermalize to bottom of conduction band; can be emitted over an NEA surface

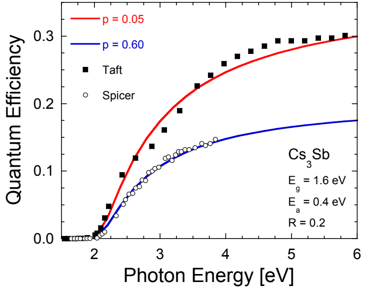
PHOTOEMISSION FROM SEMICONDUCTORS

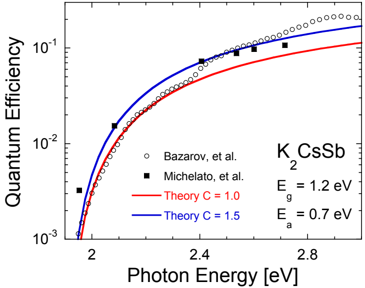
Semiconductor emission barrier is approximately triangular: Use Airy $D_\triangle(E)$:

$$QE = (1 - R(\omega)) \frac{\int_{E_a}^{\hbar\omega - E_g} E dE \int_{\sqrt{E_a/E}}^{1} x dx D_\triangle\left[Ex^2\right] f_\lambda(x, E)}{2\int_0^{\hbar\omega - E_g} E \left[\int_0^1 dx\right] dE} \tag{20}$$

$$D_\triangle(E) \approx \frac{4[E(E-E_a)]^{1/2}}{(E^{1/2} + (E-E_a)^{1/2})^2} \to QE \approx \frac{2Cs^5}{(1+s^2)(1+\sqrt{1+s^2})(s+\sqrt{1+s^2})} \tag{21}$$

where $s^2 \equiv (\hbar\omega - E_g - E_a)/E_a$ and $C \approx n(1-R)/(1+p)$ with n of order unity

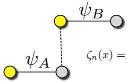




$D_\triangle(E)$: K.L. Jensen, JVSTB21, 1528 (2003); Forbes and Deane, Proc. Roy. Soc. London A (May 18, 2011).

Airy Functions and the Transfer Matrix Approach

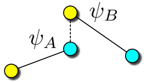
- Plane-wave Transfer Matrix Approach



$$\psi(x) = t(k)e^{ikx} + r(k)e^{-ikx}$$

$$\zeta_n(x) = \begin{pmatrix} \psi_n(x) \\ \partial_x \psi_n(x) \end{pmatrix}; \quad \hat{S}(n) = \begin{pmatrix} e^{ik_n x} & e^{-ik_n x} \\ ik_n e^{ik_n x} & -ik_n e^{-ik_n x} \end{pmatrix}$$

- Airy based Transfer Matrix Approach[a]



$$\psi(x) = t(k)\frac{Zi(c,\omega)}{Zi(c_o,\omega_o)} + r(k)\frac{Zi(-c,\omega)}{Zi(-c_o,\omega_o)}$$

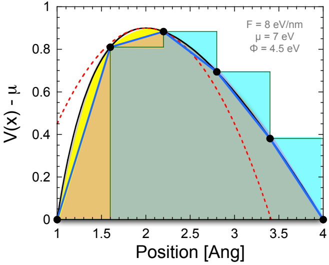
$$Zi(c,\omega) = \frac{1}{\omega^{1/4}}F(c,\omega)\exp\left(\frac{2}{3}c\omega^{3/2}\right)$$

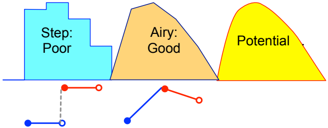
- S-matrix Assembly

$$D(E_k) = \frac{j_{trans}}{j_{inc}} \sim |t(k)|^2$$

$$\zeta_{n-1}(x_n) \equiv \hat{S}(n) \cdot \zeta_n(x_n)$$

$$\begin{pmatrix} 1 \\ r \end{pmatrix} = \left\{ \prod_{n=1}^{N} \hat{S}(n) \right\} \begin{pmatrix} t \\ 0 \end{pmatrix} \Rightarrow t(k) = \left\{ \left[\prod_{n=1}^{N} \hat{S}(n) \right]_{1,1} \right\}^{-1}$$



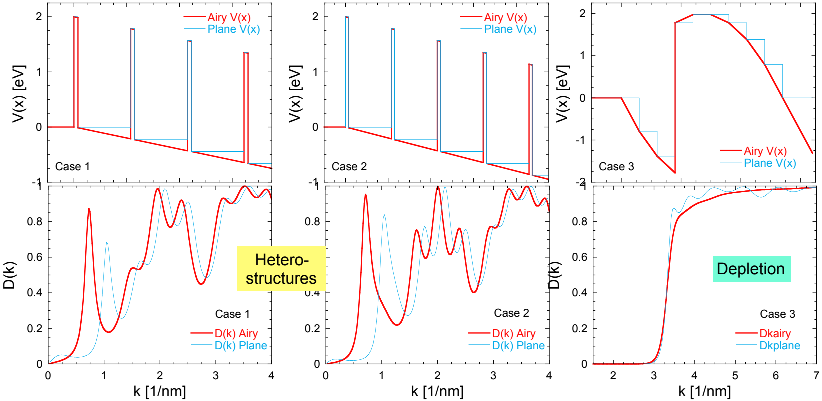


[a] K.L. Jensen. "*A quantum dipole....*" J. Appl. Phys., **111**, 054916, (2012)
K.L. Jensen, *Electron Emission Physics* (Academic Press, San Diego, CA (2007).

Sections Outline

Resonances

Crystal Structure And Current Are Inherently 3D



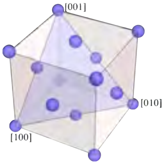
Great flexibility to treat arbitrary V(x), particularly for coatings / Graphene

Use Airy TMA on potentials from DFT / Computational Physics

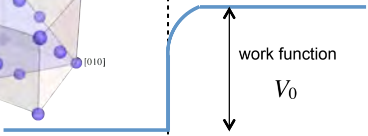
Surfaces: Work Function

Treat Resonances, Multi-photon, And Band Bending / Bound States

periodic crystal lattice



free space

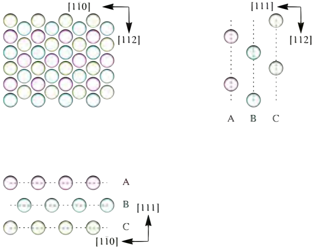


$$u_{j\vec{k}}(\vec{r})\, e^{i\vec{k}\cdot\vec{r}} \rightarrow e^{i\vec{k}'\cdot\vec{r}}$$

$$u_{j\vec{k}}(\vec{r}) = u_{j\vec{k}}(\vec{r}+\vec{a}_n)$$

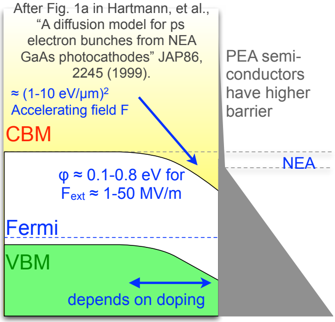
periodic Bloch amplitude

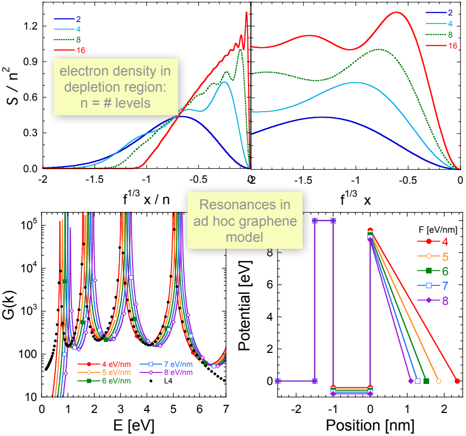
crystal lattice interface



Surfaces: Band Bending

Develop Airy TMA to find D(E)





*Apply **TMA-Zi methods to V(x)** determined by **computational materials** methods*

Summary and Future

- Develop computational model of emission for time-dependent surface conditions; find contributions of scattered electrons; photo-assisted field emission and multi-photon processes
- Incorporate atomistic simulations (e.g. DFT) for $m*$, n and k, $V(x)$, E_G, Density of states, and supply function into nearly free electron Moments based formalism to find QE and $\varepsilon_{n,rms}$ for compositional variations, heterostructures, accumulation regions.
- Extend the atomistic simulations surface potential structure to a full 3D potential representation; develop a scattering representation through the well structure (transverse and parallel components of current at interface)