Simulation, Measurement and Analysis of Photoemission from Dispenser Cathodes, Metals and Coated Materials

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Host:
Carlos Hernandez Garcia
Jefferson Lab
12000 Jefferson Ave MS6A
Newport News, VA 23606

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HISTORY

The ability to predict QE of pure metals / materials is hard.

- Fowler DuBridge Theory:
  - ⇒ R. H. Fowler, Phys. Rev. 38, 45 (1931);
  - ⇒ L. A. DuBridge, Phys. Rev. 43, 727 (1933).

If coatings are involved, it is far harder. Consider:

- A. H. Sommer,
  "The Element of Luck in Research - Photocathodes 1930 to 1980"
  (Gaede-Langmuir Award), J. Vac. Sci. Technol. A1, 119 (1983).

  "Six photocathode materials were developed during the period from 1930 to 1963 to provide the spectral response and other characteristics needed for such applications as photometry, television, scintillation counters, and night vision devices. The history and the essential properties of these materials are reviewed and it is shown that all the cathodes resulted from **lucky accidents** and **not from the application of scientific insight.** The period of empirical innovation came to an end in the late 1960's when negative electron affinity (NEA) materials became the first photocathodes that were developed on a strictly scientifc basis."

What is involved? Is a predictive model possible?

INTRODUCTION

Photocathodes:

- Sources for Electron Beams, From Free Electron Lasers (FELs) to Accelerator Applications Due to the High Quality Electron Beams
- Ideal Photocathode Has High QE at Longest Possible Wavelength, Capable of in Situ Repair or Rehabilitation, Demonstrates Good Lifetime

To meet particular needs of a megawatt (MW) class FEL, a photocathode…

- …should produce 1 nC of charge in a 10-50 ps pulse every ns (100 A peak and 1 A average current) in 10-50 MV/m and 0.01 mTorr for several seconds.

Even if such a photocathode were available…

- Making predictions of performance is complex: Useful models must account for cathode surface conditions and material properties, as well as drive laser parameters.
  - ⇒ surface conditions (coating, field enhancement, reflectivity),
  - ⇒ laser parameters (duration, intensity, wavelength), and
  - ⇒material characteristics (reflectivity, laser penetration depth, scattering)
- Focus: dispenser photocathodes, but also discuss other photocathodes

PRESENT PROGRAM: Develop and validate with experiment a predictive and quantitative theory of photoemission & quantum efficiency.

PHOTOCATHODES

DRIVE LASER

- Reliability <=> System Reliability: UV Unsuitable for Hi-duty
- Non-linear Crystals Decrease λ by 2-4; Efficiency Very Low for UV
- Conversion by 2 From IR to Green ok: Seek High QE Photocathode in Visible

PHOTOCATHODE

- Bulk & Surface of Complex Materials Produced by Empirical Techniques; Short Lifetime, Complex Replacement Process.
- Cathode Selection Influences Drive Laser Chosen (e.g., λ, spot bandwith, laser energy, QE)

METALLIC:

- Hi ave power, drive laser w/ 5 - 500 μJ/pulse req.
- Rugged but require UV, have lower QE (≤ 0.01%).
- For low duty factor, low rep rate UV pulses
- Fast response time (fs-structure On Laser Appears on Beam)

DIRECT BAND-GAP P-TYPE SEMICONDUCTORS:

- Highest QE photocathodes
  - ⇒ alkali antimonides (Cs_3Sb, K_2CsSb); visible, PEA, RF gun
  - ⇒ alkali tellurides (Cs_2Te, KCsTe) UV, PEA, RF gun
  - ⇒ Bulk III-V wCs + oxidant (O or F); IR - visible, NEA, DC guns
- Emission time is long (10-20 ps) for NEA sources: insufficiently responsive for pulse shaping.
- ALL chemically reactive: Easily poisoned by H_20 & $C0_2$ (Protection at expense of QE); "Harmless" H_2 & CH_4 damage by ion back bombardment (greater issue for DC guns)

EMISSION NON-UNIFORMITY

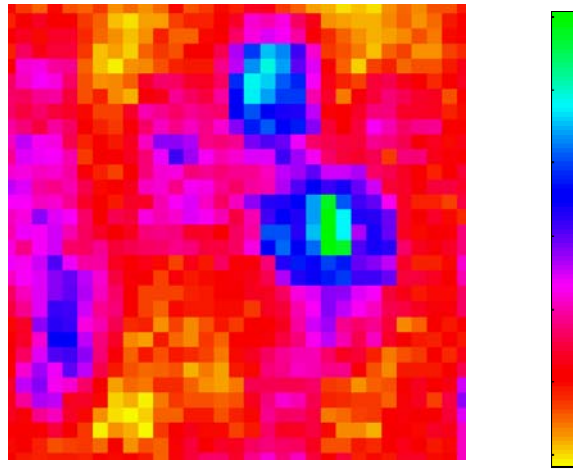
Environmental Conditions Can

- Erode low work function coatings
- Deposit material that degrades performance
- Damage the surface (ion bombardment)

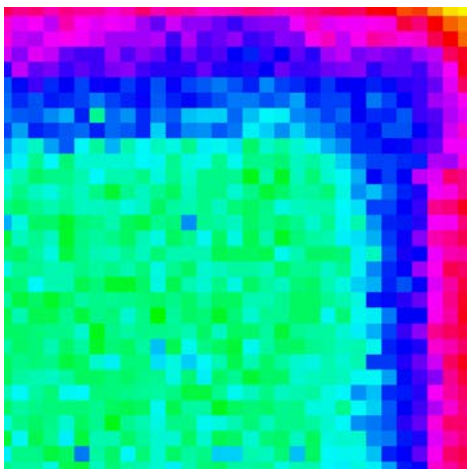
Re-cleaning / Reconditioning does not necessarily restore original performance

- QE scans of **LEUTL Photoinjector Mg Cathode**
  Courtesy of John W. Lewellen, Argonne National Lab
- Details: images from APS photoinjector.
  Blue = 2xYellow; pixels =10 micron^2; image = (300 pixels)^2
  Operation: 6 Hz for 30 days (1.55E7 pulses total); macropulse = 1.5 μs

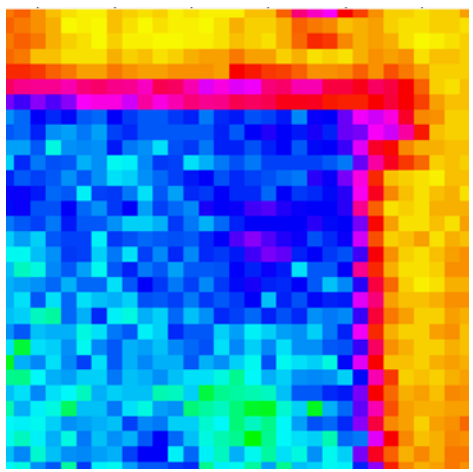
31 Oct 01 – before 1st cleaning



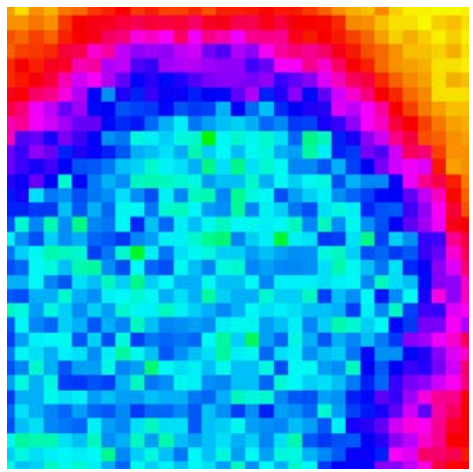
5 Nov 2001 - after 1st cleaning



4 Dec 2001 - after 1st cleaning



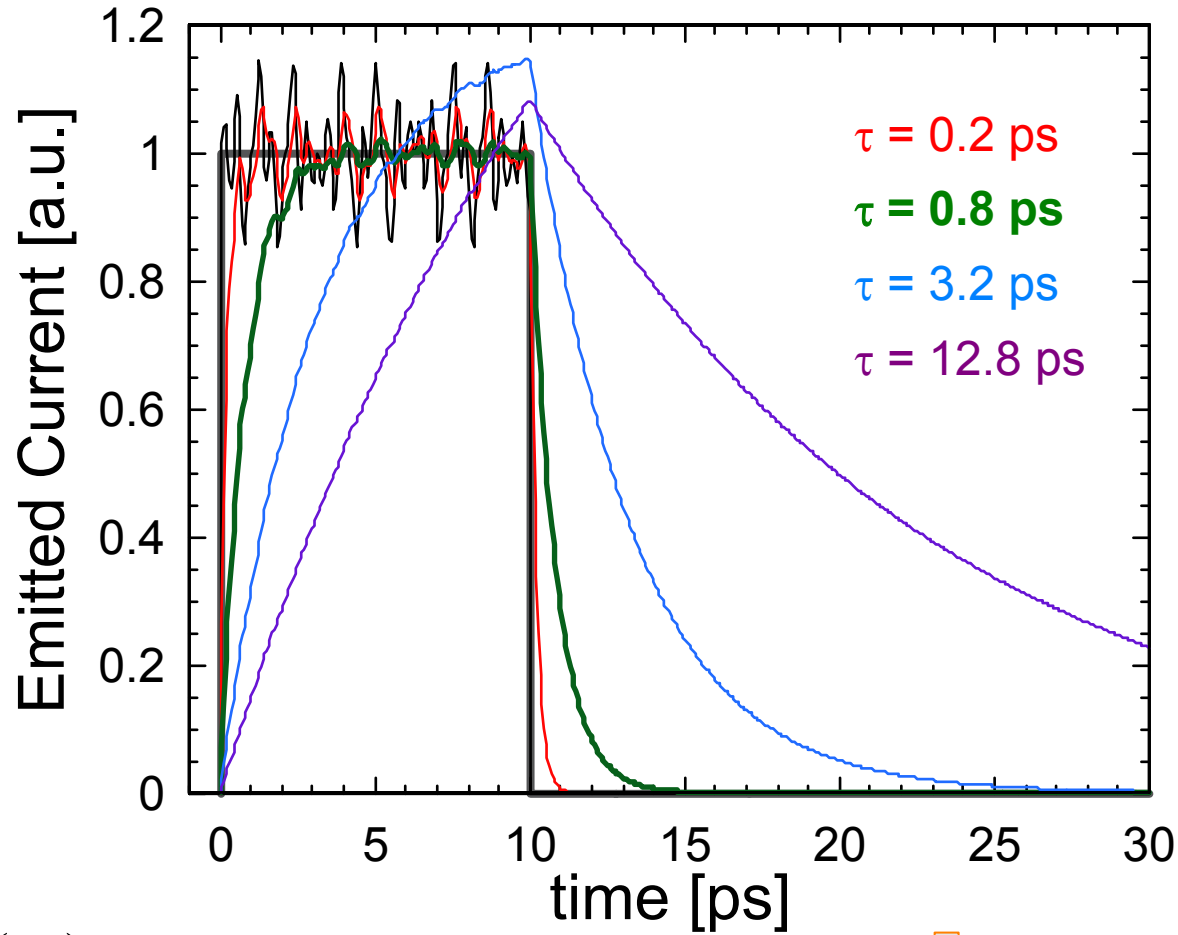
10 Dec 2001 - after 2nd cleaning



PHOTOCATHODE RESPONSE TIME

Pulse Shaping

- Optimal Shape for emittance: beer-can (disk-like) profile
- Laser Fluctuations occur (esp. for higher harmonics of drive laser)
- Fast response: laser hash reproduced
- Slow response: beer-can profile degraded
- Optimal: 1 ps response time



Mathematical Model ($\omega_n = 2\pi n/T$)

$$I_\lambda(t) = I_o\theta(t)\theta(T-t)\sum_{n=0}^{N} c_n \cos(\omega_n t)$$

$$I_e(t) = \frac{QE}{\tau}\int_{-\infty}^{t} I_\lambda(s)\exp\left[-\left(\frac{t-s}{\tau}\right)\right] \propto \sum_{n=0}^{N}\frac{c_n}{1+(\omega_n\tau)^2}\begin{cases}\left[\left(\cos(\omega_n t)+\omega_n\tau\sin(\omega_n t)\right)e^{T/\tau}-1\right]e^{-t/\tau} & t<T \\ \left(e^{T/\tau}-1\right)e^{-t/\tau} & t\geq T\end{cases}$$

FN AND RLD DOMAINS

DOMAINS

- RLD: Corrupted When Tunneling Near Barrier Maximum Is Non-negligible

$$F < \left(\frac{\sqrt{2m}}{10\hbar} k_B T\right)^{4/3} Q^{1/3}$$

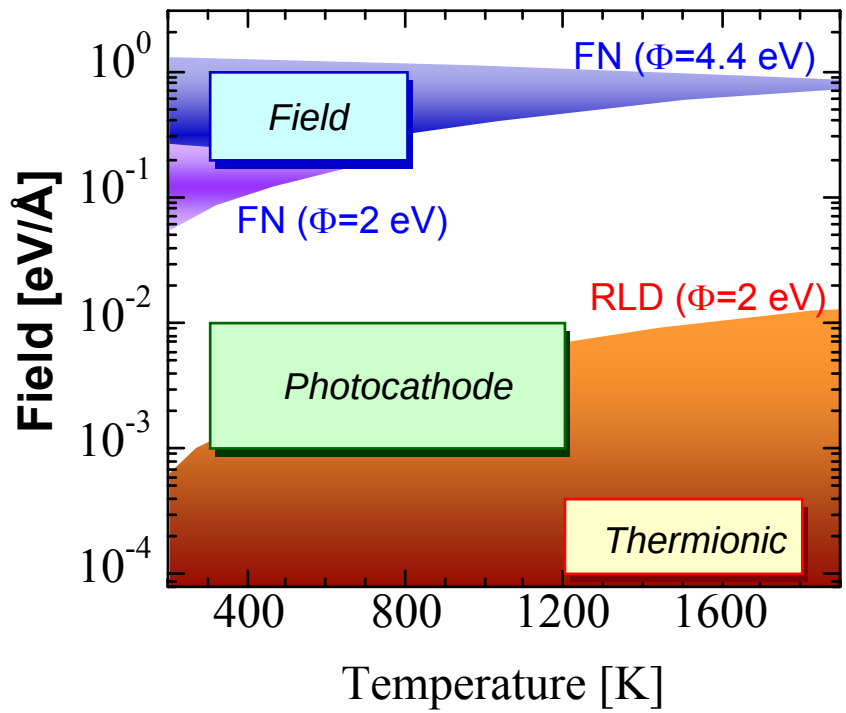
- FN: Corrupted When Barrier Maximum Is Too Close to Fermi Level or Slope of ln($T(E)$) > ln($f(E)$)

  ⇒ Maximum Field: $\beta\phi > 6$

$$F < \frac{1}{4Q}\left(\Phi - 6k_B T\right)^2$$

  ⇒ Minimum Field: $c_{fn} < 2\beta$

$$F > \frac{4}{\hbar}\sqrt{\left(2m\Phi\right)} k_B T$$



For high intensity lasers incident on photocathodes, emission is NOT field OR thermal OR photo, but it is **ALL** of these processes acting in concert.

QUANTUM EFFICIENCY (3-D)

Quantum Efficiency is ratio of total # of emitted electrons with total # of incident photons

$$QE = \frac{\frac{1}{q}\int_{-\infty}^{\infty}\int_{0}^{\infty} J\left[F(\rho), T(\rho,t)\right] 2\pi\rho d\rho dt}{\frac{1}{\hbar\omega}\int_{-\infty}^{\infty}\int_{0}^{\infty} I(\rho,t) 2\pi\rho d\rho dt}$$

Emitted Current *J(F,T)*

- Photoemission component

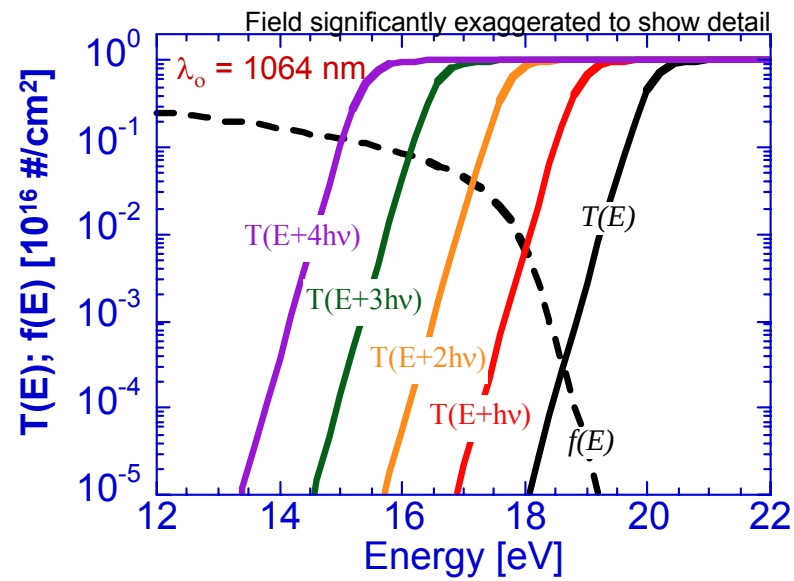
$$\frac{q}{\hbar\omega}(1-R) f_\lambda I(\rho) \frac{\int_0^\infty D(E+\hbar\omega;\rho) f(E) dE}{\int_0^\infty f(E) dE}$$

- Thermal-field component (limit: RLD)

$$\frac{q}{2\pi\hbar}\int_0^\infty D(E) f(E) dE$$

$\Rightarrow$ Richardson Eq. (High T, Low F)

$\Rightarrow$ Fowler Nordheim (High F, Low T)



To estimate local time-dependent current density as a function of local temperature and field, we use:

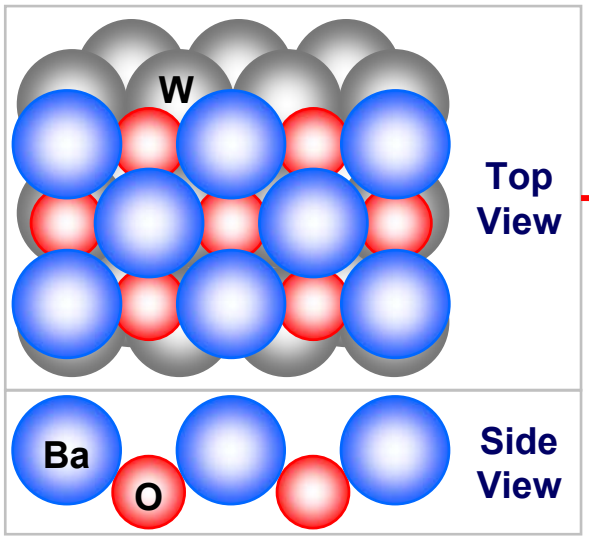
$$J(T,F,\Phi) = f_\lambda \frac{q}{\hbar\omega}(1-R) I_\lambda(t) \frac{U\left[\beta(\hbar\omega-\phi)\right]}{U\left[\beta\mu\right]} + A_{RLD} T^2 \exp\left[-\beta\phi\right]$$

$$\beta\phi = \left[\Phi - \sqrt{4QF}\right] / k_B T_e$$

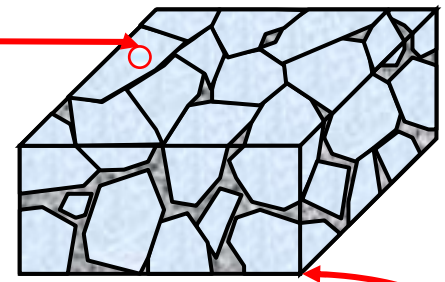
"Fowler factor"

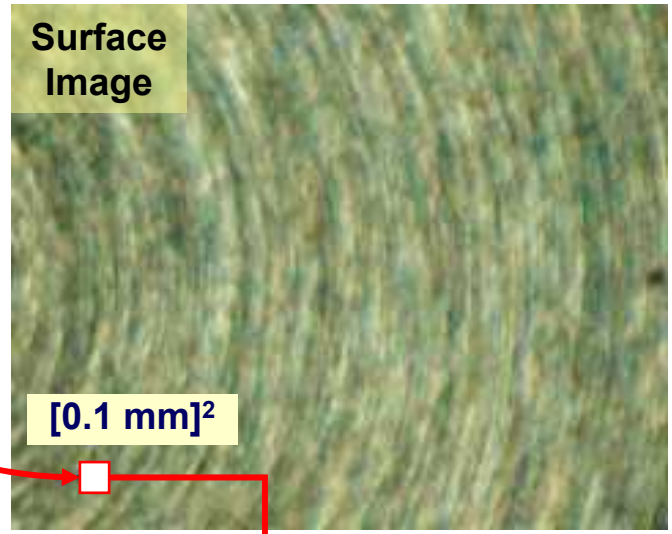
$$U(x) = \int_{-\infty}^{x} \ln\left(1+e^y\right) dy = \begin{cases} e^x\left(1-be^{ax}\right) & (x \le 0) \\ \frac{1}{2}x^2 + \frac{\pi^2}{6} - e^{-x}\left(1-be^{-ax}\right) & (x > 0) \end{cases}$$

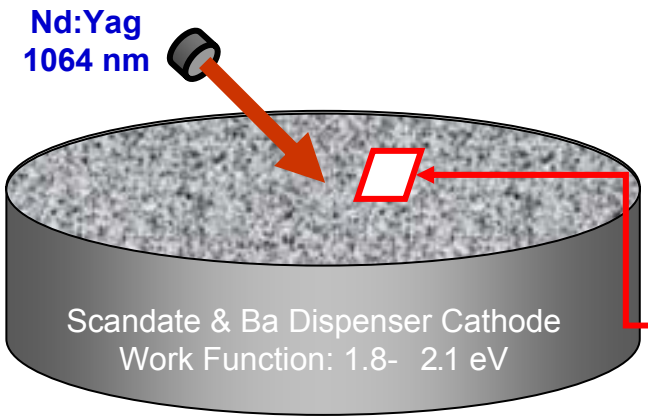
DISPENSER CATHODE AS PHOTOCATHODE

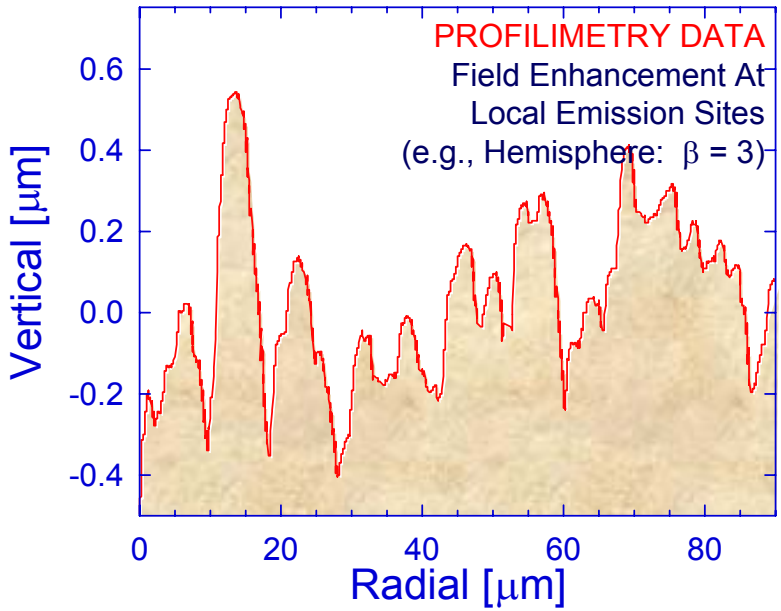


Interpore ≈ 6 μm; Grain Size≈ 4.5 μm; Pore Diam. ≈ 3 μm









DISPENSER CATHODES

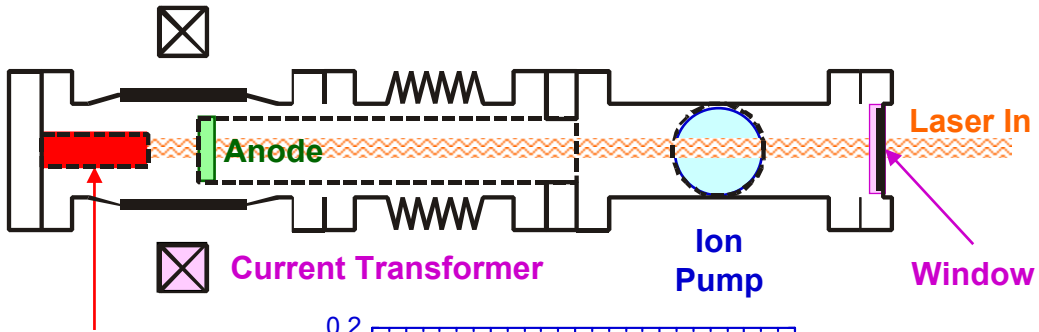
- Used in radar & communications
- Porous tungsten matrix w/ impregnates which diffuse to surface
- Emitting region constantly renewed (self rejuvenating *in situ*)
- Robust and long lived, can operate at elevated temperatures

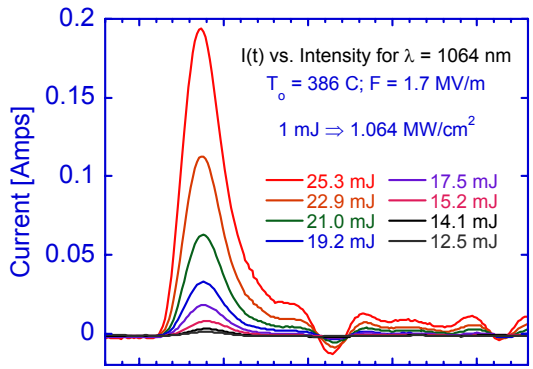
UMD EXPERIMENTAL PHOTOCATHODE PROGRAM

EXPERIMENAL PROGRAM to develop & test robust photocathodes capable of O(ps)-pulses with O(nC) charge, suitable for high duty factor DC and RF guns. A dispenser photocathode that can be self-annealed or repaired, that operates with a visible drive-laser, and at modestly elevated temperatures, is focus.

EXPERIMENT:

- Cathodes from SPECTRAMAT CORP.
  - ⇒ B (sintered tungsten matrix impregnated with barium calcium aluminate)
  - ⇒ Scandate (similar to a B cathode with the addition of scandium oxide impregnate)
  - ⇒ M (B cathode w/ thin coating of osmium
- Field (cathode- anode) varied from 0 25 MV/m
- Q switched Nd:YAG laser: Gaussian pulses of FWHM 4.5 ns focused to spot with FWHM area of approximately 0.3 cm^2
- Cathodes contain integral heater to activate surface by raising T to 1200 C for hour, and maintaining a temperature of several hundred C above RT: lifetime ≈30 hours @ 10E 8Torr.
- After QE depreciation, performance restored by raising temperature to 700C for several minutes.





UMD dispenser cathode data

RAW EXP. DATA (Dispenser Cathode)

EXP. VARIATION PARAMETERS

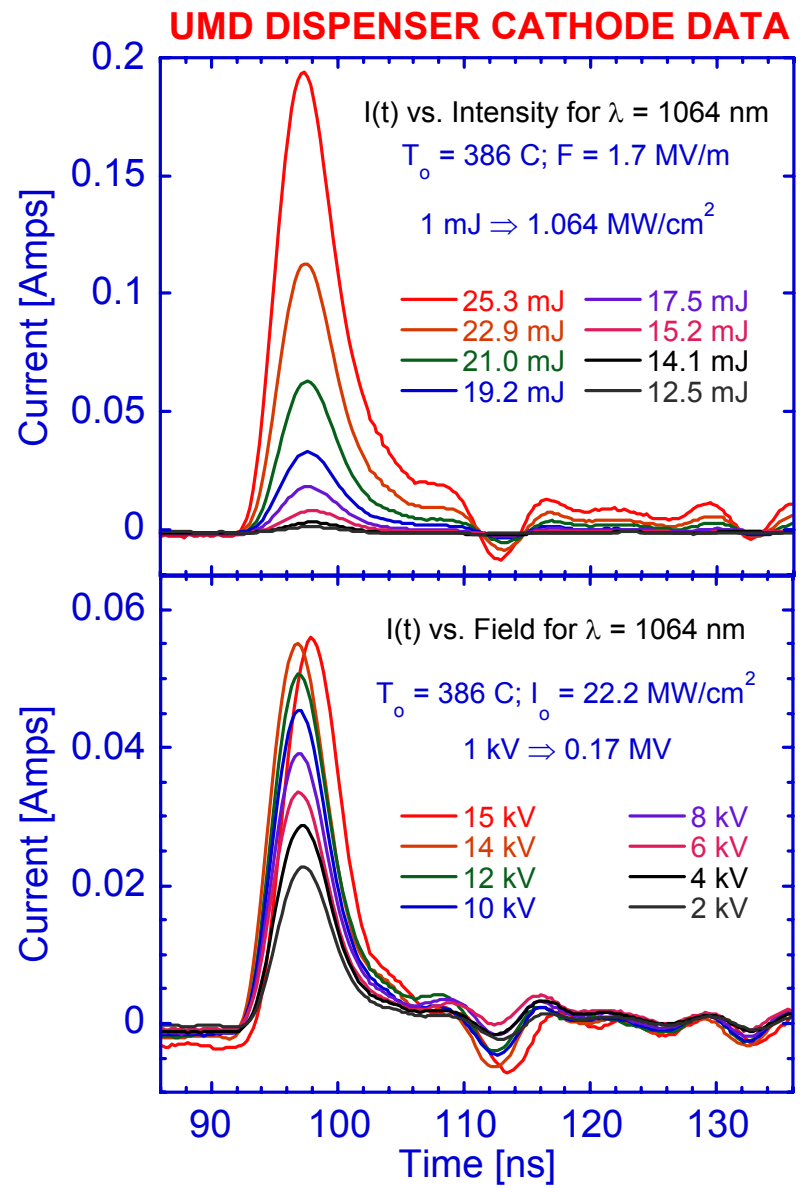
- Anode V_a [kV] 0 -15
- Bulk T_o [K] 297-1040
- Laser Energy [mJ] 4.87 - 22

MATERIAL (Tungsten +Coatings)

- Chemical Potential 18.08 eV
- Laser Wavelength 1064 nm
- Coatings (monolayer) 1.8 - 2.1 eV
- Illumination Radius 0.525 cm - 0.125 cm

THEORY: COMPLICATIONS TO 1-D MODEL

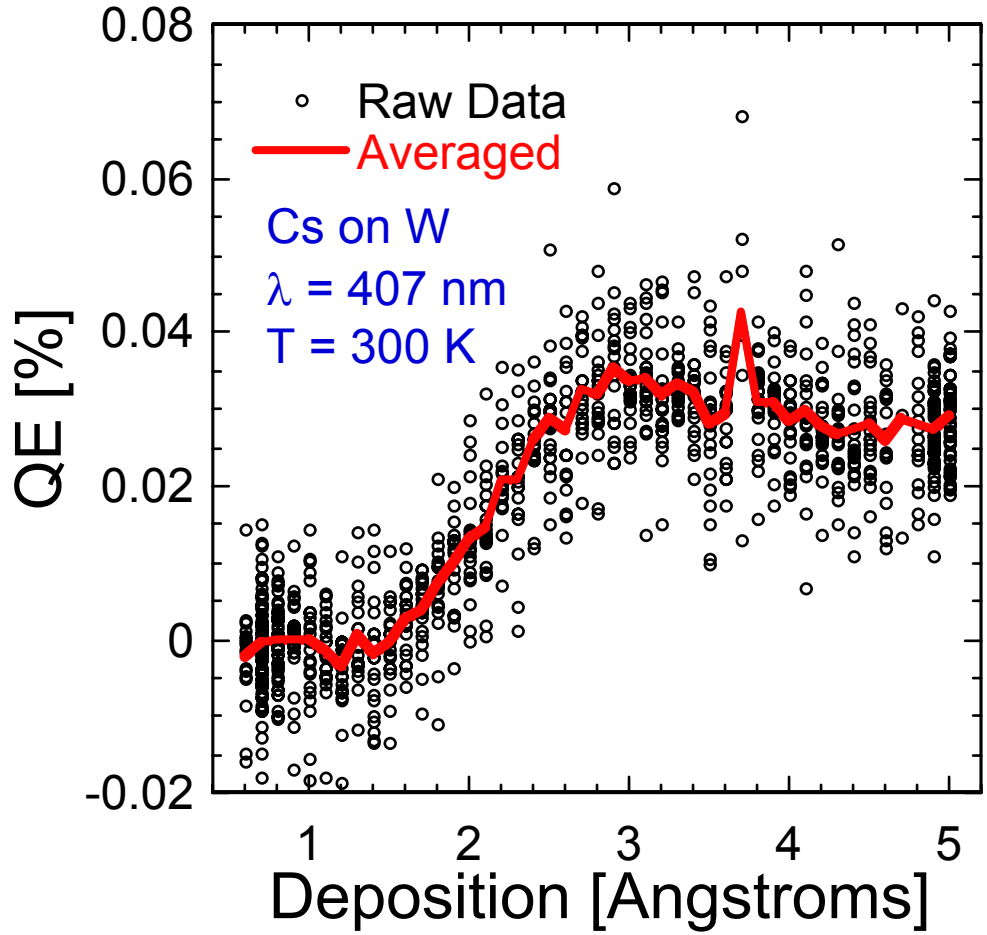
- Laser intensity is Gaussian in cylindrical coordinate ρ: FWHM area = 0.3 cm^2
- Field Variation across surface:
  Cathode = 1.27 cm Diameter.
  Anode: Tube w/ 1.27 cm ID / 2.54 cm OD
  Anode-cathode Separation = 0.4 Cm.
  1 kV Anode = 0.17 Mv/m @ center
- Electron Temperature Greatest Where Laser Strongest (Center of Beam)
- Emitted Charge [nC] based on integration of Gaussian fit to numerical data… but: what is base-line current? What about circuit ringing?
- Work function variation with coverage
- Reflectivity & absorption depth depend on λ



RAW EXP. DATA (Cs evaporation on W)

- Cesium Evaporated Onto Cleaned Tungsten Surface
- Deposition Thickness Is Linearly Related to Coverage Factor
  - ⇒ Proportionality factor ≈ Atomic Diameter
- Noisy Data - several measurements per x-coordinate
  - ⇒ "Averaged" Points:

$$\left\langle QE\left(x_i\right)\right\rangle = \frac{1}{N_i}\sum_{k=1}^{N_i} QE\left(x_i(t_k)\right)$$



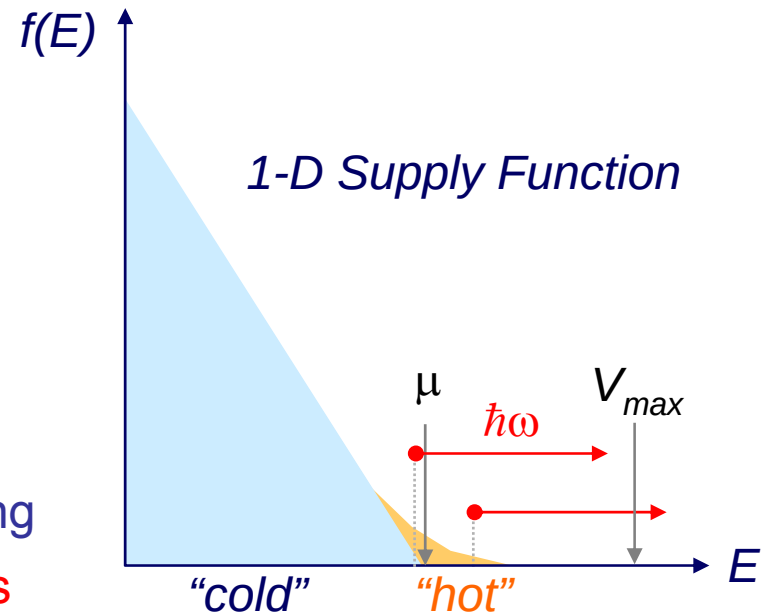
Cesium:

- Atomic Radius: 2.6 Angstroms
- Covalent Radius: 2.25 Angstroms

LASER HEATING & PHOTO-EMISSION

Laser Energy Transferred via:

- Photon Energy Transferred Into Electronic Excitations
- Hot Electrons Come Into Thermal Equilibrium With Other Electrons Via Electron-Electron Scattering.
- Hot Thermal FD Electron Distribution Comes Into Thermal Equilibrium With the Lattice Via Electron-Phonon Scattering
- For Long Duration Laser Pulses, Photons Encounter "Hot" Electron Distribution From Which Photoemission is Enhanced



"Ultrashort Laser-induced Electron Photoemission: a Method to Characterize Metallic Photocathodes" N A Papadogiannis, S D Moustaizis, J. Phys. D: Appl. Phys. 34, 499 (2001):

- "The **duration of the laser pulse** (450 fs) **is relatively long compared to the electron–electron scattering time** for typical electron temperatures…"
- "Thus, the **electrons thermalize rapidly** acquiring a Fermi–Dirac distribution and the refereed electron– electron and electron–phonon scattering times concern the thermalized electrons.
- "...a hot electron gas (a few thousand kelvin) requires about 0.5–2 ps (depending on the experimental conditions) to relax again to its equilibrium state.

LASER HEATING OF ELECTRON GAS

Differential Eqs. Relating Electron (T_e) to Lattice Temperature (T_i)

Electron & Lattice Specific Heat

$$C_e \frac{\partial}{\partial t} T_e = \frac{\partial}{\partial z}\left(\kappa(T_e, T_i)\frac{\partial}{\partial z} T_e\right) - g\left(T_e - T_i\right) + G\left(z,t\right)$$

$$C_i \frac{\partial}{\partial t} T_i = g\left(T_e - T_i\right)$$

Power transfer by electrons to lattice
285.1 GW / K cm^3 (W @ RT)

Laser Energy Absorbed

Thermal Conductivity

$$\kappa\left(T_e, T_i\right) = \frac{2\mu}{3m}\tau\left(T_e, T_i\right) C_e\left(T_e\right)$$

Relaxation Time

$$\tau\left(T_e, T_i\right) = \left[A_{ee} T_e^2 + B_{ep} T_i\right]^{-1}$$

$$A_{ee} = \frac{A_o k_B^2}{h\mu}$$ electron - electron scattering

$$B_{ep} = \frac{2\pi\lambda_o k_B}{h}$$ electron - lattice scattering

A_o and λ_o = dimensionless parameters dictated by photo - cathode material

Deposited Laser Energy

$$G(z,t) = \left(1 - R\right) I_\lambda(t)\left(\frac{e^{-z/\delta}}{\delta}\right)\left[1 - \frac{U\left[\beta\left(h\omega - \phi\right)\right]}{U\left[\beta\mu\right]}\right]$$

Reflection — Penetration — Absorbed Energy

Incident Laser Power [W/cm^2]

Variation in Energy Density with Temperature

$$C\left(T\right) = \frac{\partial}{\partial T} E$$

$$C_e\left(T_e\right) = \frac{\gamma\, T_e}{\left(1 + \frac{7}{40}\left(\frac{\pi}{\beta_e \mu}\right)^2\right)}; \quad C_i\left(T_i\right) = \frac{3Nk_B}{\left(1 + \frac{1}{20}\left(\frac{T_D}{T_i}\right)^2\right)}$$

Electrons

Phonons
T_D = Debye Temp

TIME SIMULATIONS

Matrix form of coupled differential equations for electron & lattice temperature

$$\left(C_e + J - D\right)\Big|_{t+\Delta t} \bullet T_e\left(t+\Delta t\right) = \left(C_e - J + D\right)\Big|_{t} \bullet T_e\left(t\right) + 2J \bullet T_e(t) + \frac{1}{2}\int G(t)dt + T_{bc}$$

Electrons

$$\left(D + H\right)\Big|_{t+\Delta t} \bullet T_i\left(t+\Delta t\right) = \left(D + H\right)\Big|_{t} \bullet T_i\left(t\right) + H \bullet \left(T_e(t+\Delta t) + T_e(t)\right)$$

Lattice

Finite Difference Algorithm (2nd Order Accurate)

Non-linearity in Temperature:

- Finite Difference Multi-point Algorithm Necessary for broad range of time scales

Boundary T conditions are nontrivial and depend on history

- RHS: reflect / LHS: absorb
- Temp BC Given by Macro-Time Results and Held Fixed

Matrices Require $C_x(T_x)$ at Future Time Steps

- Prediction / correction Scheme: Guess j+1 - solve - use results in next iteration
- Repeat for several iterations

$$\left[C_e\right]_{i,j} = \frac{1}{2\Delta t}\left\{C_e\left[T\left(t+\Delta t\right)\right] + C_e\left[T\left(t+\Delta t\right)\right]\right\}\delta_{ij}$$

$$\left[C_i\right]_{i,j} = \frac{1}{2\Delta t}\left\{C_i\left[T\left(t+\Delta t\right)\right] + C_i\left[T\left(t+\Delta t\right)\right]\right\}\delta_{ij}$$

$$\left[H\right]_{i,j} = \frac{1}{2}g\delta_{ij}; \qquad \left[J\right]_{i,j} = \frac{g\left[C_i\right]_{i,j}}{2\left[C_i\right]_{i,j} + g}\delta_{ij}$$

$$\begin{aligned}\left[D(t)\right]_{i,j} = {} & \frac{1}{4\Delta x^2}\left\{\left[\kappa\left(T_e,T_i\right)\right]_{j-1} + \left[\kappa\left(T_e,T_i\right)\right]_{j}\right\}\delta_{i,j-1} \\ & - \frac{1}{2\Delta x^2}\left\{\left[\kappa\left(T_e,T_i\right)\right]_{j-1} + 2\left[\kappa\left(T_e,T_i\right)\right]_{j} + \left[\kappa\left(T_e,T_i\right)\right]_{j+1}\right\}\delta_{i,j} \\ & + \frac{1}{4\Delta x^2}\left\{\left[\kappa\left(T_e,T_i\right)\right]_{j-1} + \left[\kappa\left(T_e,T_i\right)\right]_{j}\right\}\delta_{i,j+1}\end{aligned}$$

Multi-point Algorithm (Preserves stability)

SPECIFICATION OF SCATTERING TERMS

Heat Transfer in Solids Due to Free Electrons & Phonons

"SUM OF PARTIAL RESISTIVITIES":
Total resistance to current flow is sum of each kind of resistance; resistance is inversely related to scattering rate: (Matthiessen's Law)

$$\tau^{-1} = \tau_{ee}^{-1} + \tau_{ep}^{-1}$$

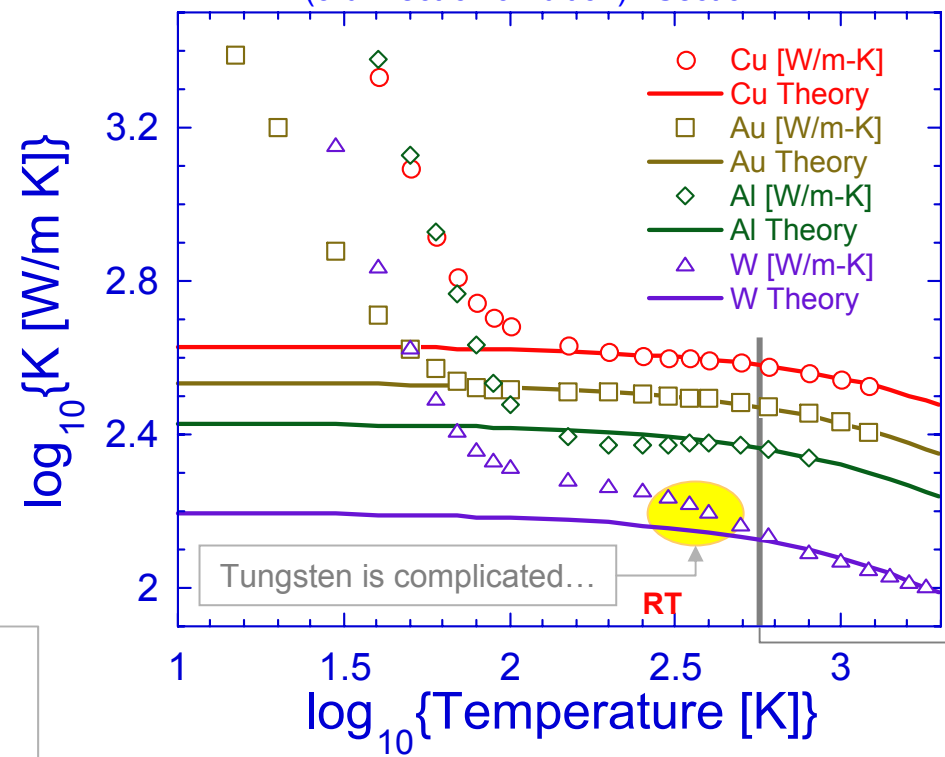
$$\tau_{ee}(T_e) = \frac{1}{A_{ee}T_e^2} \Leftrightarrow \frac{h\mu}{A_o}\left(\frac{1}{k_B T_e}\right)^2$$

$$\tau_{ph}(T_i) = \frac{1}{B_{ep}T_i} \Leftrightarrow \frac{h}{2\pi\lambda_o}\left(\frac{1}{k_B T_i}\right)$$

HEAT CONDUCTIVITY
(Kinetic Theory of Gases)

$$\kappa(T_e,T_i) = \frac{1}{3}C_e(T_e)v_F l = \frac{2\mu}{3m}C_e(T_e)\tau(T_e,T_i)$$

Data from CRC Handbook of Chemistry and Physics (3rd Electronic Edition): Section 12



$$B_{ep} + A_{ee}T \approx \frac{\gamma\mu}{3m\kappa(T,T)}$$

Parameter	Au	W	Cu	Al
A_{ee} [10^7 K^{-2} s^{-1}]	3.553	57.86	4.044	19.77
B_{ep} [10^{11} K^{-1} s^{-1}]	1.299	18.41	1.859	6.886

DETERMINATION OF R[%] & δ

Algorithm:

- Spline-fit experimental optical data (e.g., CRC, AIP Handbook) for index of refraction (n), damping constant (k)
- Designate incident angle = θ
- Use Equations to determine Reflectance R[%] and penetration depth of laser for given wavelength

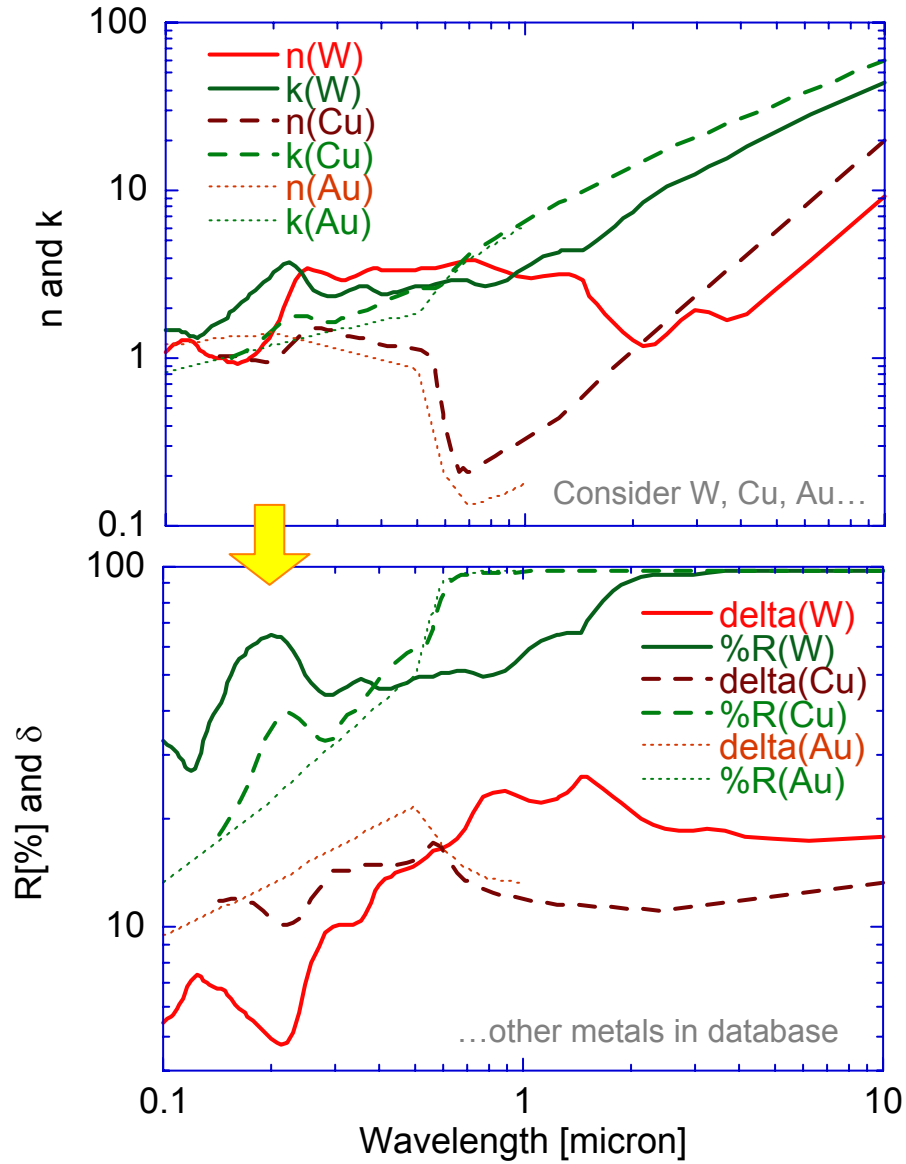
$$R_s = \frac{a^2 + b^2 - 2a\cos\theta + \cos^2\theta}{a^2 + b^2 + 2a\cos\theta + \cos^2\theta}$$

$$R_p = R_s \frac{a^2 + b^2 - 2a\sin\theta\tan\theta + \sin^2\theta\tan^2\theta}{a^2 + b^2 + 2a\sin\theta\tan\theta + \sin^2\theta\tan^2\theta}$$

$$2a^2 = \left[\left(n^2 - k^2 - \sin^2\theta\right)^2 + (2nk)^2\right]^{1/2} + \left(n^2 - k^2 - \sin^2\theta\right)$$

$$2b^2 = \left[\left(n^2 - k^2 - \sin^2\theta\right)^2 + (2nk)^2\right]^{1/2} - \left(n^2 - k^2 - \sin^2\theta\right)$$

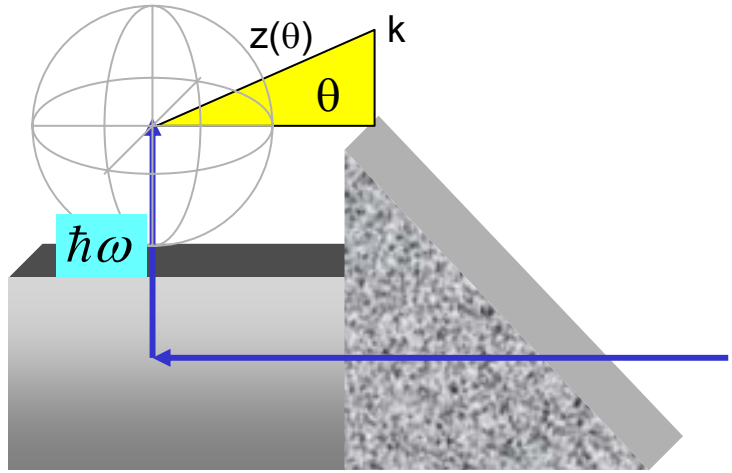
$$R = \frac{1}{2}\left(R_s + R_p\right); \quad \delta = \frac{\lambda}{4\pi k}$$



POST-ABSORPTION SCATTERING FACTOR

Factor (f_λ) governing proportion of electrons emitted after absorbing a photon:

- Photon absorbed by an electron at depth x
- Electron Energy augmented by photon, but direction of propagation distributed over sphere
- Probability of escape depends upon electron path length to surface and probability of collision (assume any collision prevents escape)
  - ⇒ path to surface & scattering length $z(\theta)=\frac{x}{\cos(\theta)};\quad l(k)=\frac{\hbar k\tau}{m}$
- To leading order, k integral can be ignored



$$f_\lambda=\frac{\int_{k_o}^{\infty} f(k)dk\int_0^{\pi/2} d\theta\int_0^{\infty}\exp\left(-\frac{x}{\delta}-\frac{z(\theta)}{l(k_\lambda)}\right)dx}{\int_{k_o}^{\infty} f(k)dk\int_0^{\pi} d\theta\int_0^{\infty}\exp\left(-\frac{x}{\delta}\right)dx}$$

$$k_\lambda=\frac{1}{\hbar}\sqrt{2m\left(E(k)+\hbar\omega\right)}$$

k_o: minimum momentum of electron that can escape after photo-absorption

$$f_\lambda\approx\frac{1}{2}G\left(\frac{m\delta}{\hbar k_o\tau}\right)$$

Average probability of escape

argument < 1

$$G\left[\cos(y)\right]=1-\frac{2}{\pi}\cot(y)\ln\left(\frac{1+\sin(y)}{\cos(y)}\right)$$

argument > 1

$$G\left[\sec(y)\right]=1-\frac{2}{\pi}\left(\frac{y}{\sin(y)}\right)$$

COUPLING OF LATTICE / ELECTRON TEMPERATURE

Transfer Of Electron Energy To Lattice:
For T > T_D (400 K For W), C_i = Constant:

$$\frac{\partial}{\partial t}T_i(t) = \frac{g}{C_i}(T_e - T_i) \equiv \alpha(T_e - T_i)$$

$$g = \frac{\pi^2}{6} m v_s^2 B_{ep} \rho$$

Electron density [#/cm^3]

For Gaussian $T_e(t)$

$$T_e(t) = T_{bulk} + (T_e(0) - T_{bulk})\exp\left[-(t/\Delta t)^2\right]$$

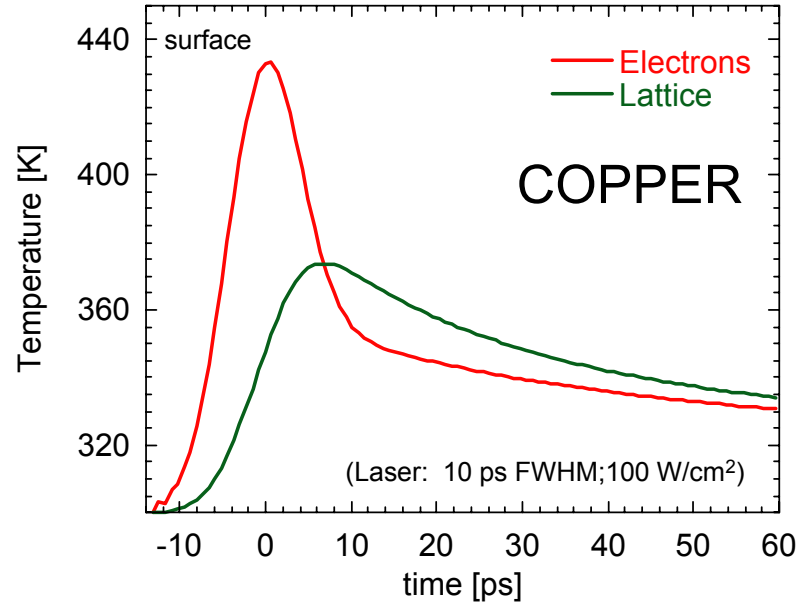
$$T_i(t) = T_{bulk} + \alpha \int_{-\infty}^{t} e^{-\alpha(t-s)}(T_e(s) - T_{bulk})ds$$

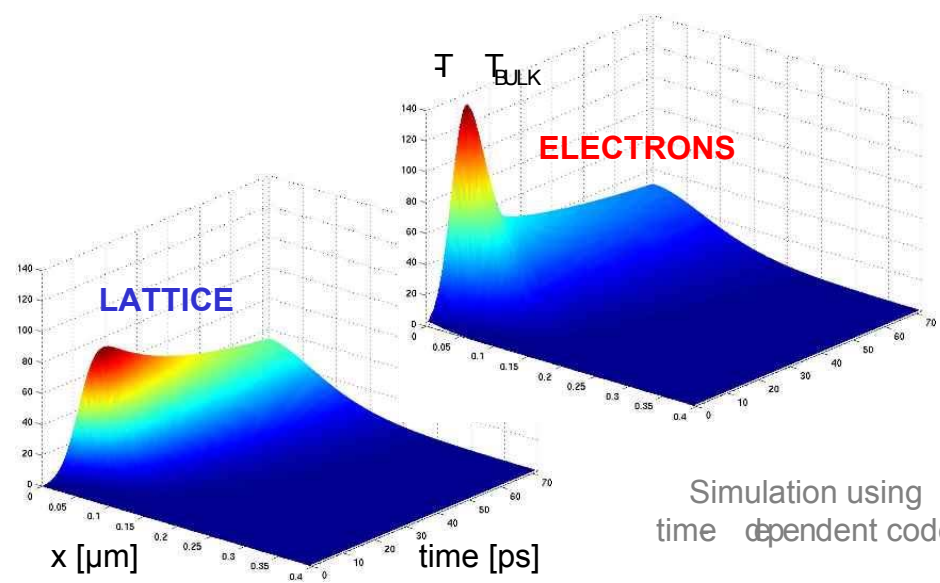
Near Maximum:

$$T_i(0) \approx T_e(0) - (\alpha\Delta t)^{-2}(T_e(0) - T_{bulk})$$

For $\Delta t \geq 10/\alpha$, $T_e(t)$ and $T_i(t)$ and equivalent to within 1%

Ex: Copper: $\Delta t \geq 59.70$ ps
Gold: $\Delta t \geq 209.5$ ps
Tungsten: $\Delta t \geq 0.95$ ps





Simulation using time dependent code

GYFTOPOULOS-LEVINE THEORY

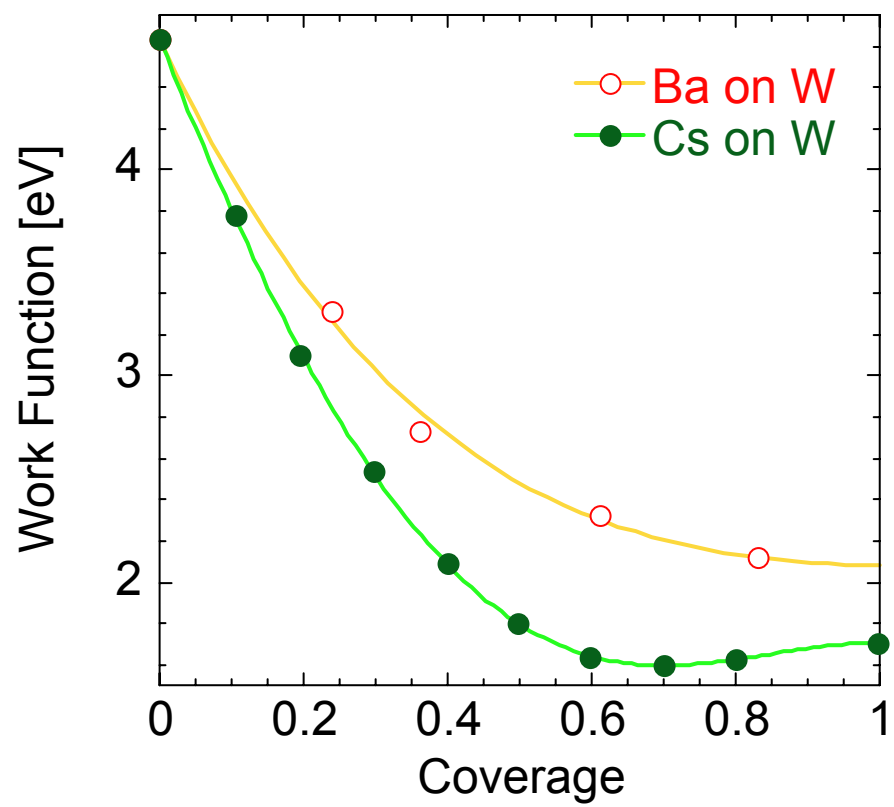
Coverings (e.g., Ba, Cs) on bulk (e.g., W) induces a change in Work Function $\Phi(\theta)$ by presence of dipoles and differences in electronegativity

GL Theory* predicts $\Phi(\theta)$ due to partial monolayer using hard-sphere model of atoms (covalent radii)

Definition of terms

ϕ_f, ϕ_m	Work function (monolayer & bulk)
r_b, r_w	Covalent radii (monolayer & bulk)
θ	Fractional coverage factor
$W(\theta)$	Electronegativity Barrier
$d(\theta)$	Dipole Moment of Adsorbed Atom

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



Ba data: G. A. Haas, A. Shih, C. Marrian, Appl. Surf. Sci. 16, 139 (1983).

Cs data: J. B. Taylor, I. Langmuir, Phys. Rev. 44, 423 (1933).

* 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

ELECTRONEGATIVITY BARRIER

$$W(\theta) = \phi_f + (\phi_m - \phi_f) H(\theta)$$

$$H(\theta) = \sum_{n=0}^{3} C_n \theta^n$$

H(θ) = <u>simplest</u> polynomial satisfing:

- W(0) = ϕ_f : the work function is equal to electronegativity ϕ_f of bulk → $C_0 = 1$
- ∂_θ W(0) =0: …and the addition of a few atoms doesn't change that. → $C_1 = 0$
- W(1) = ϕ_m: the work function is equal to electronegativity ϕ_m of adsorbate → $C_2 + C_3 = 1$
- ∂_θ W(1) =0 …and the subtraction of a few atoms doesn't change that. → $2C_2 + 3C_3 = 0$

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

DIPOLE TERM

Pauling (paraphrased):

"Dipole moment of molecule A-B proportional to difference in electronegativities ($\phi_A - \phi_B$)"

Assume true for site composed of 4 substrate (hard sphere) atoms in rectangular array with absorbed atom at apex.

Dipole moment per atom = $M(\theta)$

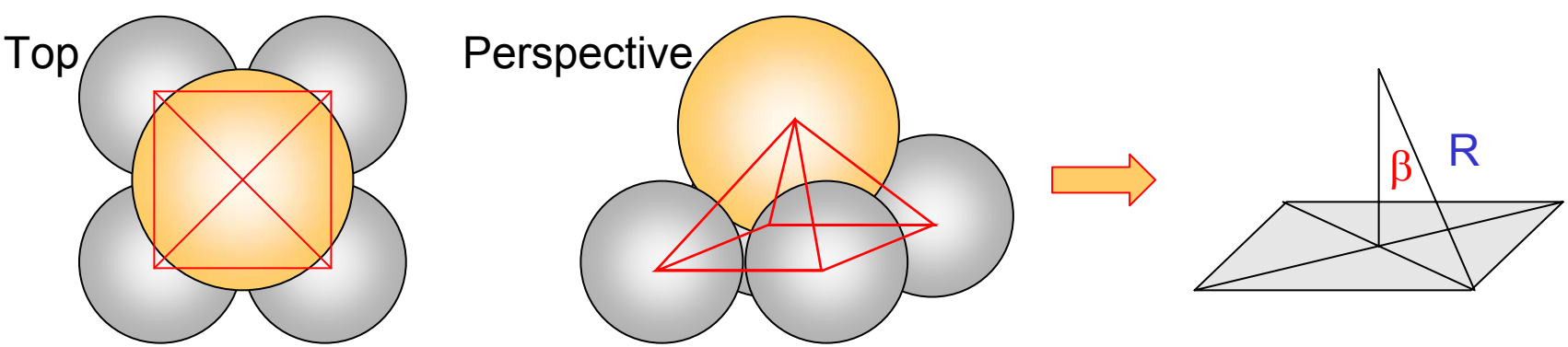
$$\cos(\beta) = \sqrt{1 - \frac{1}{2\gamma_m R^2}}$$

γ_m is number of substrate atoms per unit area

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

$$M_o = 4\varepsilon_o r_o^2 \cos(\beta)(\phi_W - \phi_B)$$

$$r_o = 4.3653 \text{ Angstroms}$$



DEPOLARIZATION EFFECT

Correction for "depolarizing effect" due to other adsorbed atoms (other dipoles) turns M into M_e ("effective" dipole moment")

Depolarizing field $E(\theta)$

Dipole moment of adsorbed atom:

$$M(\theta) \Rightarrow M_e(\theta) = M(\theta) - E(\theta)$$

$$E(\theta) = \frac{9}{4\pi\varepsilon_0}(\gamma_f \theta)^{3/2} M_e(\theta)$$

$$M_e(\theta) = \frac{M(\theta)}{1 + \frac{9\alpha}{4\pi\varepsilon_0}(\gamma_f \theta)^{3/2}}$$

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

γ_f is number of adsorbate atoms per unit area

Polarizability (α)

- n = 1.00 for alkali metals,
  1.65 for alkaline-earth
- r_b = covalent radius of adsorbate
- r_w = covalent radius of bulk

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

WORK FUNCTION IN TERMS OF H & G

Atoms per unit area: re-express in terms of the covalent radii and introduced dimensionless factors "f" and "w" which act as (dimensionless) "atoms per cell"

$$\gamma_m \equiv \frac{w}{(2r_m)^2}; \quad \gamma_f \equiv \frac{f}{(2r_f)^2}$$

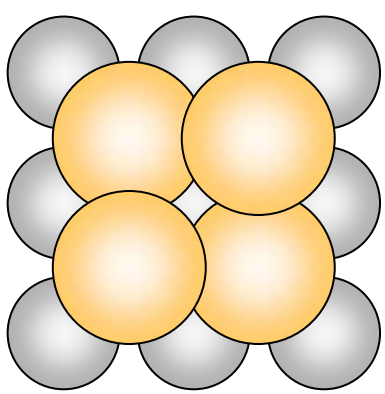
Values of f & w will depend on exposed crystal face. G&L argue that surface is "bumpy [B]":

$$\gamma_m[110] = \sqrt{2}\gamma_m[100]$$

$$\gamma_m[B] = \sqrt{3}\gamma_m[100]$$

$\gamma_f : \gamma_m = 1:4$ for Cs on W, Mo, Ta

$\gamma_f : \gamma_m = 1:2$ for Ba, Sr, Th on W, etc.

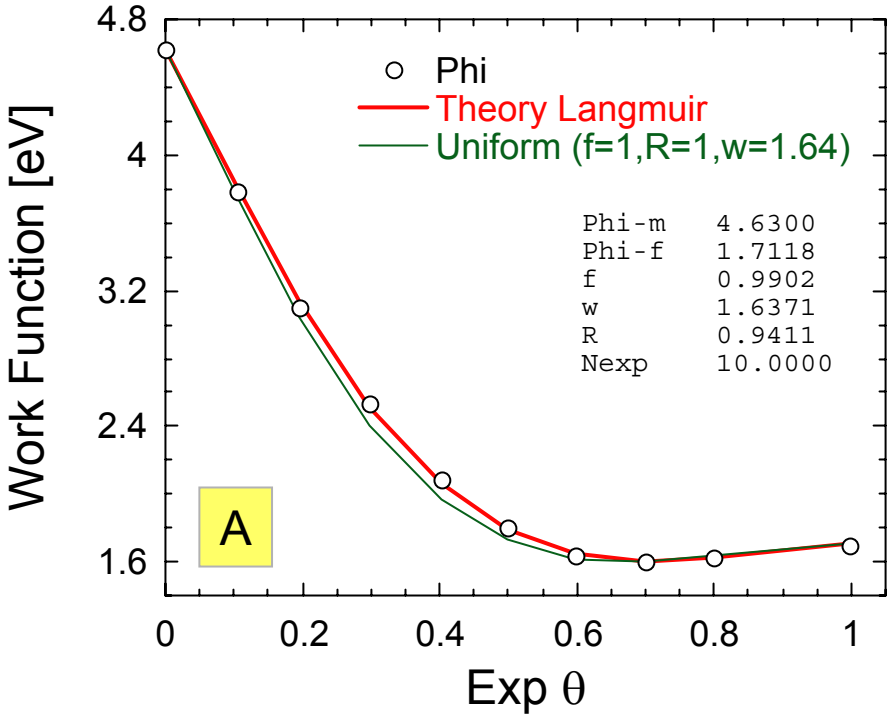


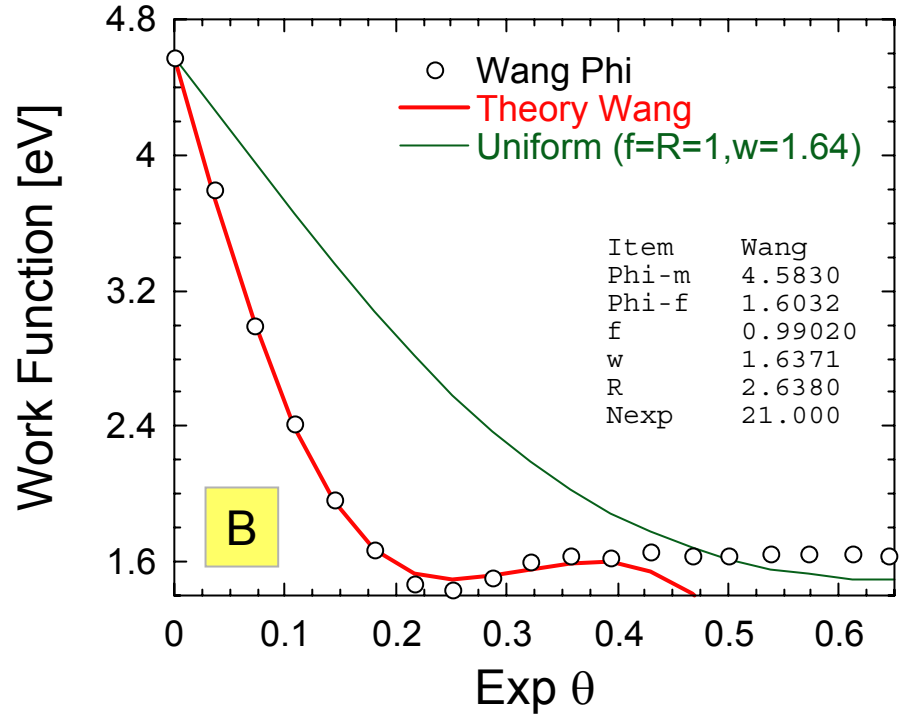
Put the pieces together to obtain a parametric representation of Gyftopoulos-Levine Theory

$$\Phi(\theta) = \phi_f - (\phi_f - \phi_m)\theta^2(3-2\theta)\{1-G(\theta)\}$$

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

RESULT OF ANALYSIS: Cs on W





LEAST SQUARES ANALYSIS:

- Set value of f = 0.9902,
  Bulk Work function = intercept value of experimental data
- Constrain *w* for Cs on W so ratio of coverage factors = 4
- Perform Least Squares Minimization to find optimal Scale Factor *R* and ϕ_f

A — Chen-Show Wang, "High photoemission efficiency of submonolayer cesium-covered surfaces", J. Appl. Phys. 44, 1477 (1977), Figure 1

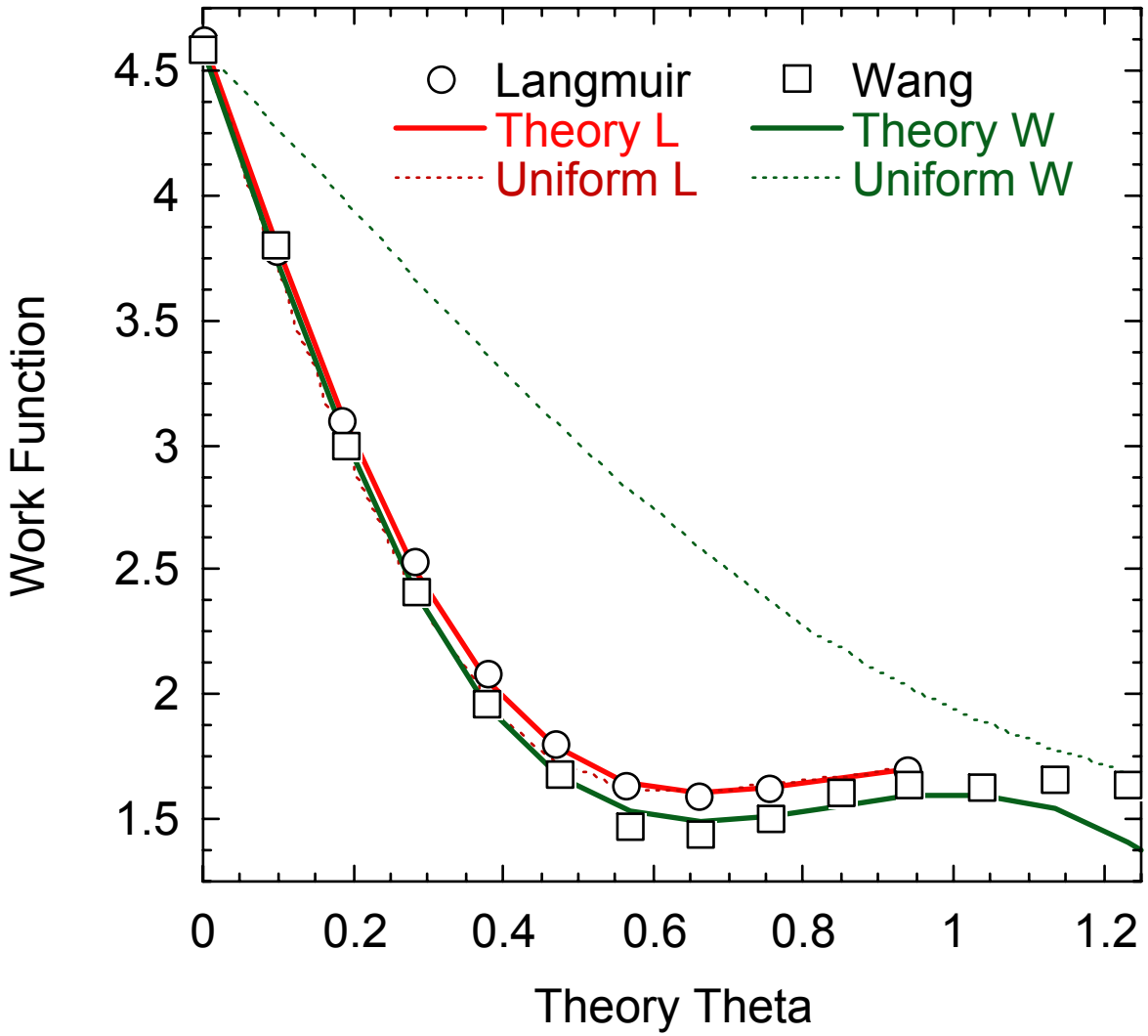
B — G. A. Haas & R. E. Thomas, "Thermionic Emission and Work Function," Chapter 2 from Techniques of Metals Research, Vol. VI, Part 1 (R. F. Bunshah, Ed.) (John Wiley & Sons, 1972), based on Taylor and Langmuir, Phys Rev. 44, No. 6, p423

RECAST EXP IN TERMS OF THEORETICAL θ

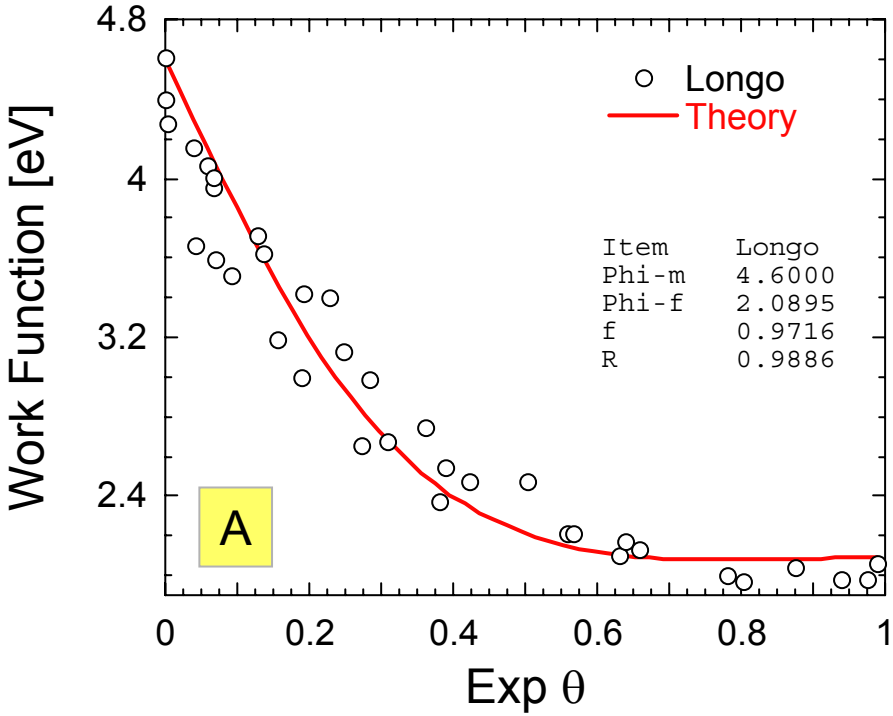
"Correct" the Experimental Coverage Factor to Force Experiment To Agree With Gyftopoulos-Levine Theory

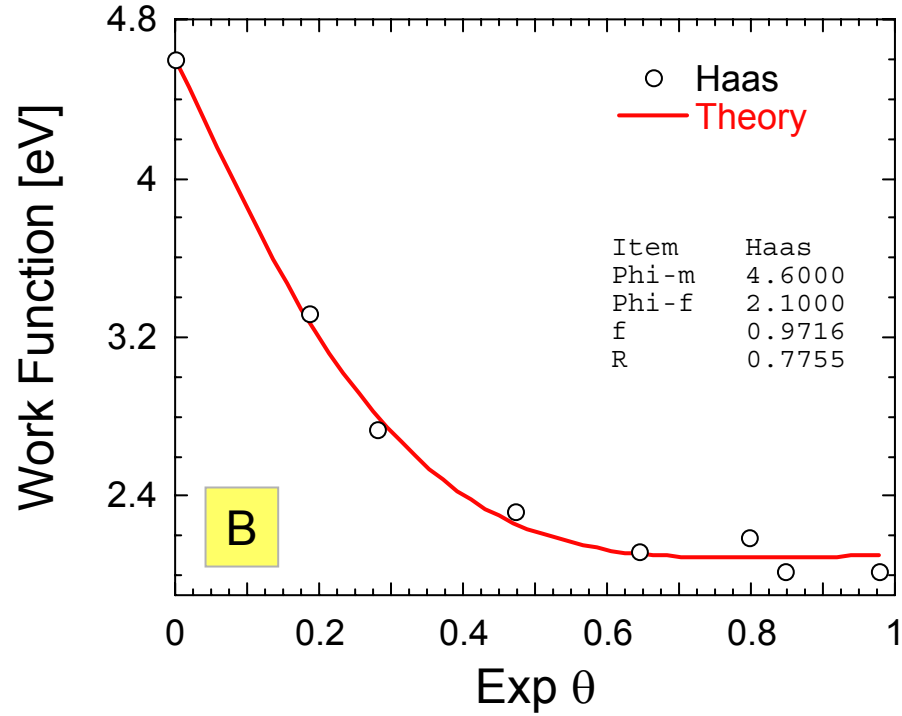
Consequence: Agreement Between the Two Data Sets (Taylor / Langmuir vs. Wang) Becomes Very Good

Agreement With Theory Based on Reasonable Parameters (Uniform: f = 1, R = 1, Surf Ratio = 4) Is Excellent.



RESULT OF ANALYSIS: BaO on W





LEAST SQUARES ANALYSIS:

- Set value of f = 0.9716,
  Bulk Work function = 4.6 eV
- Constrain *w* for Ba on W so ratio of coverage factors = 2
- Perform Least Squares Minimization to find optimal Scale Factor *R* and ϕ_f

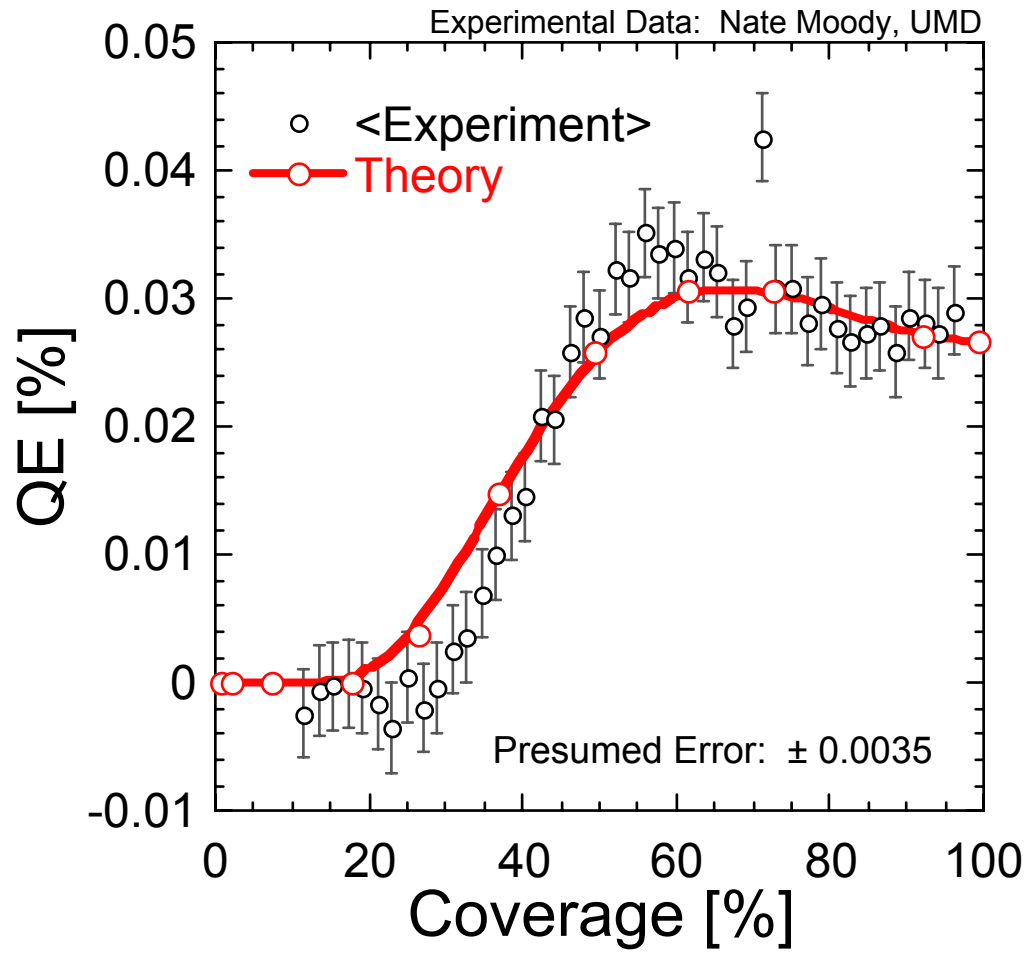
A R. T. Longo, E. A. Adler, L. R. Falce, "Dispenser Cathode Life Prediction Model," IEEE IEDM 84, 12.2 (1984).

B G. A. Haas, A. Shih, C. R. K. Marrian, "Interatomic Auger Analysis of the Oxidation of Thin Ba Films", Appl. Surf. Sci. 16, 139 (1983).

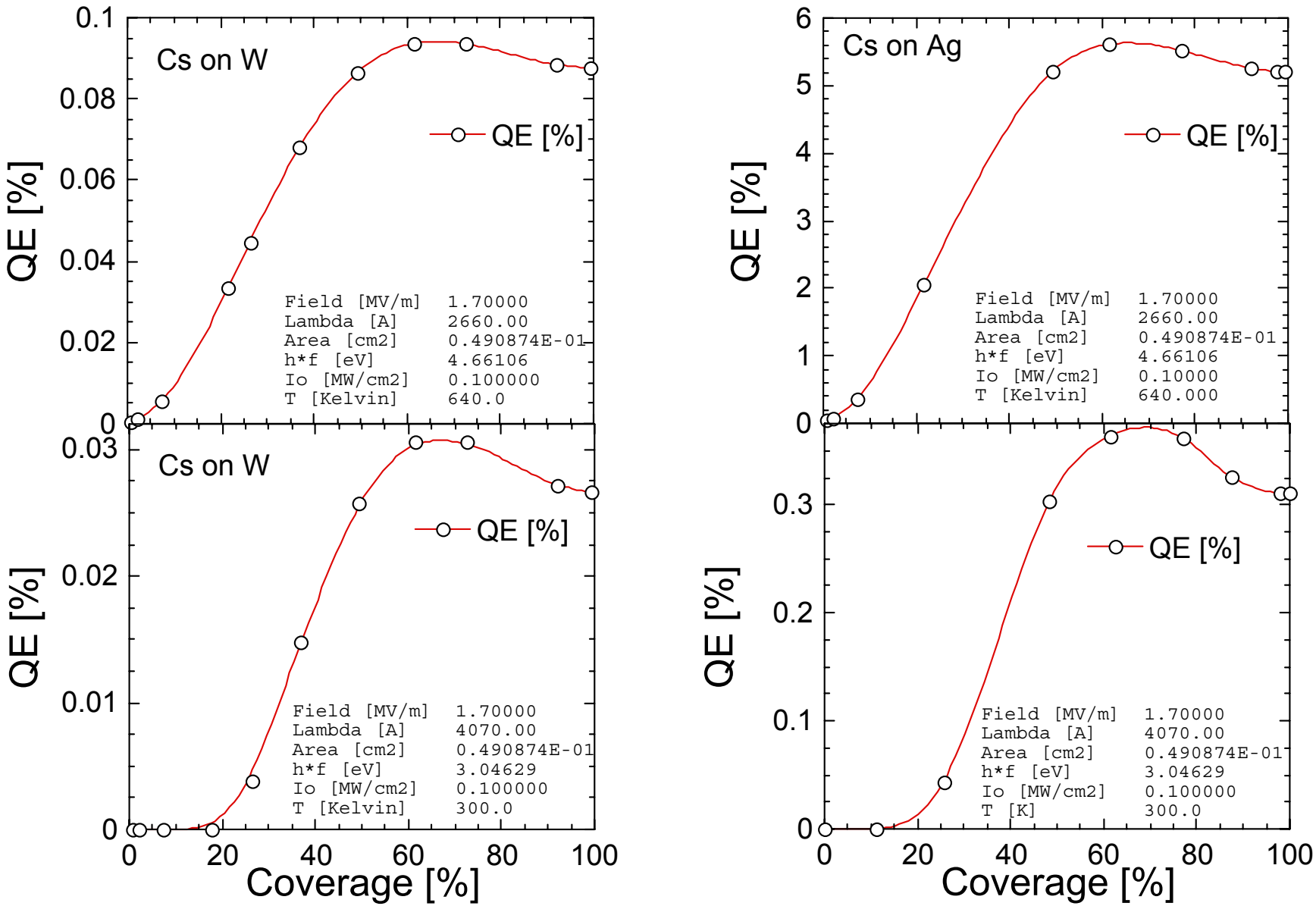
QE OF Cs ON W: EXP. VS. THEORY

Assumptions and Conditions:

- Coverage Is Uniform
- Scale factor between Coverage (theory) and Deposition thickness (exp) taken as Atomic diameter:
  - ⇒ Scale = 100%/(5.2 Angstroms)
- Compare averaged experimental data to theoretical calculation
- Field and Laser intensity low enough so that Schottky barrier lowering, field enhancement, and heating are negligible.



QE OF Cs ON W, Ag: Predictions / Comparisons



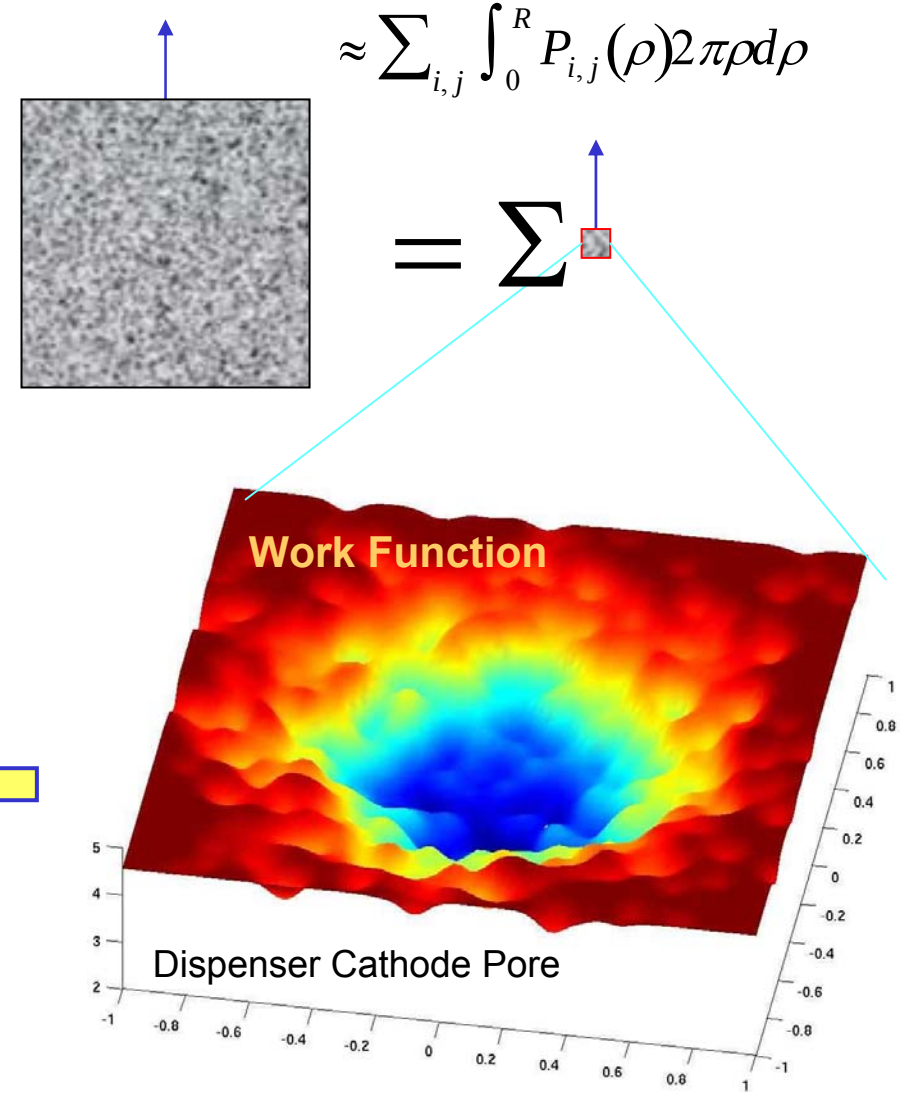
ACCOMMODATING SURFACE VARIATION

Variation can be geometric, adsorbate-induced, and/or coverage dependent:

- Let *P* = property dependent on surface (e.g., work function) and macro variables F and T (e.g., field, temperature)
- Define surface by regions indexed by (i,j)
- Macroscopic surface = sum over micro patches

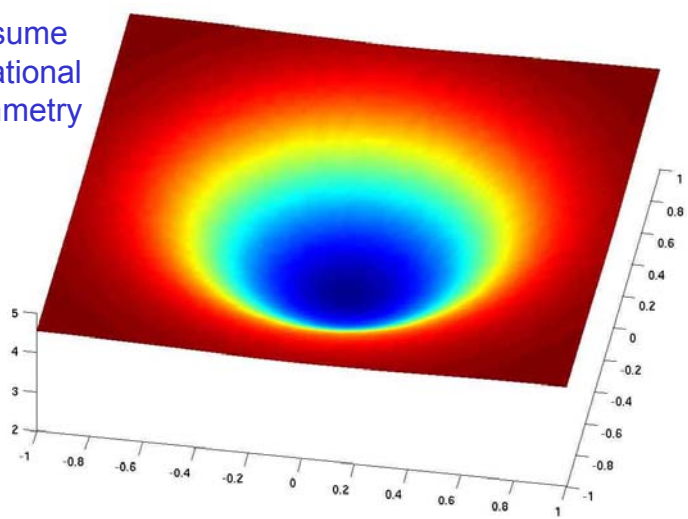
$$\int_{\Omega} P(F,T;x,y)\,d\Omega = \sum_{i,j} \int_{\Omega_{i,j}} P(F,T;x,y)\,d\Omega_{i,j}$$

$$\approx \sum_{i,j} \int_0^R P_{i,j}(\rho)\,2\pi\rho\,d\rho$$



$$P_{i,j}(\rho) = \frac{1}{2\pi}\int_0^{2\pi} P\left(F_{i,j},T_{i,j};x_i+\rho\cos\phi,y_j+\rho\sin\phi\right)d\phi$$

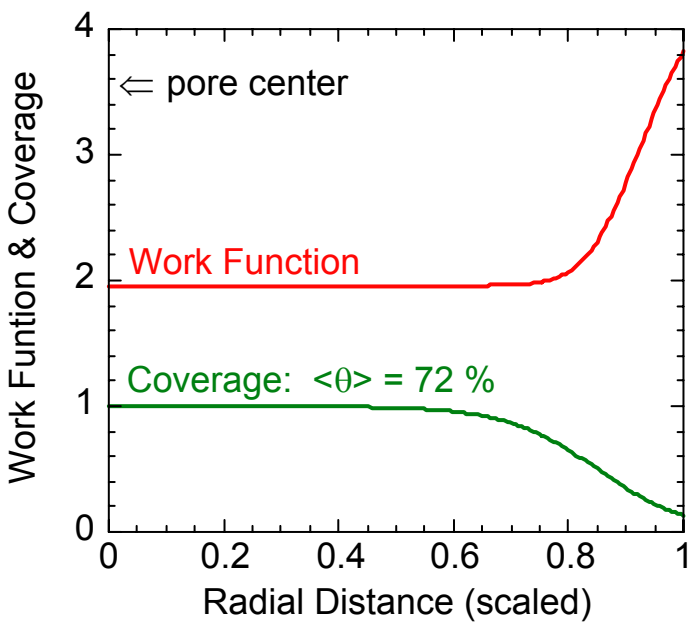
Assume Rotational Symmetry

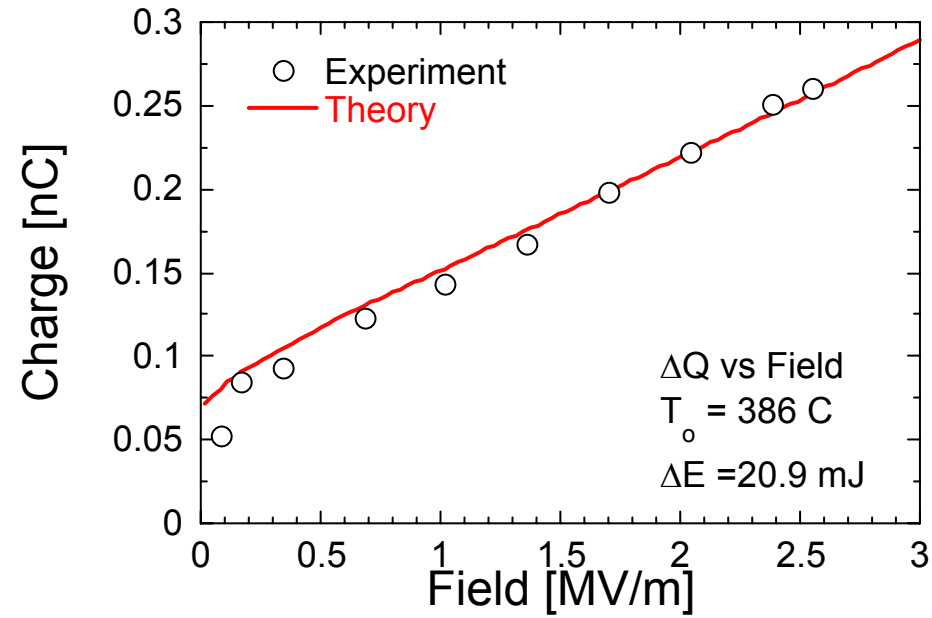


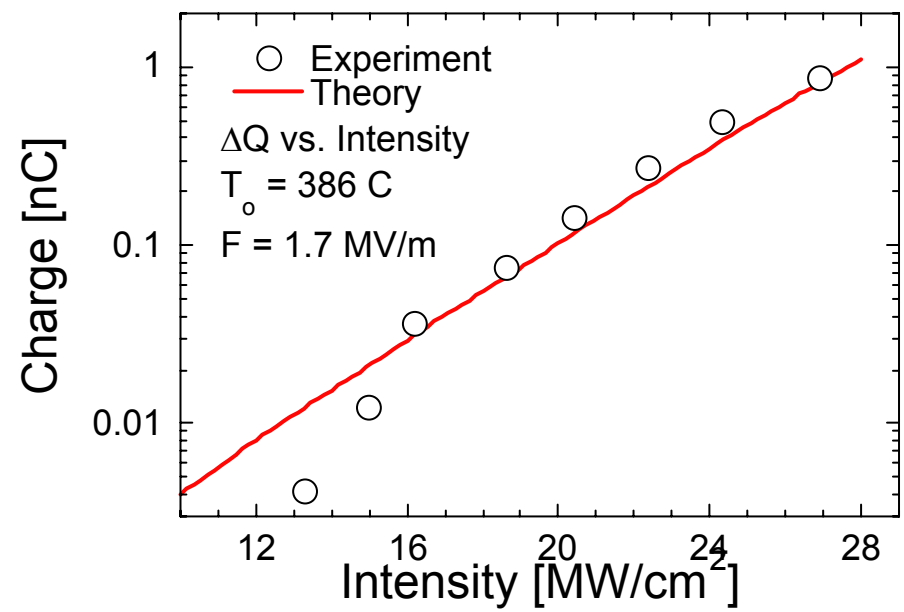
EXP - SIMULATION: EMITTED CHARGE

Exp. Data 9-24-03 C K data (scandate) for 1064 nm: 4.5 ns pulses over ≈ 0.2 cm^2 areas

- Exp: slight changes in conditions due to time between measurements, separation between illuminated regions
- Theory: <u>same</u> input data set used for both simulations (opposed to adjusting parameters to obtain best fit for *each*)
- Coverage factors lead to following work function variation over the pore region:







EXP - SIM: QE (Dispenser Photocathodes)

Sample Data set: (B-type)

BULK: Tungsten (library with user def Rlambda)
COATING: Barium (library with user def Mono Phi)
---
implicit temp-dependent quantities evaluated at RT

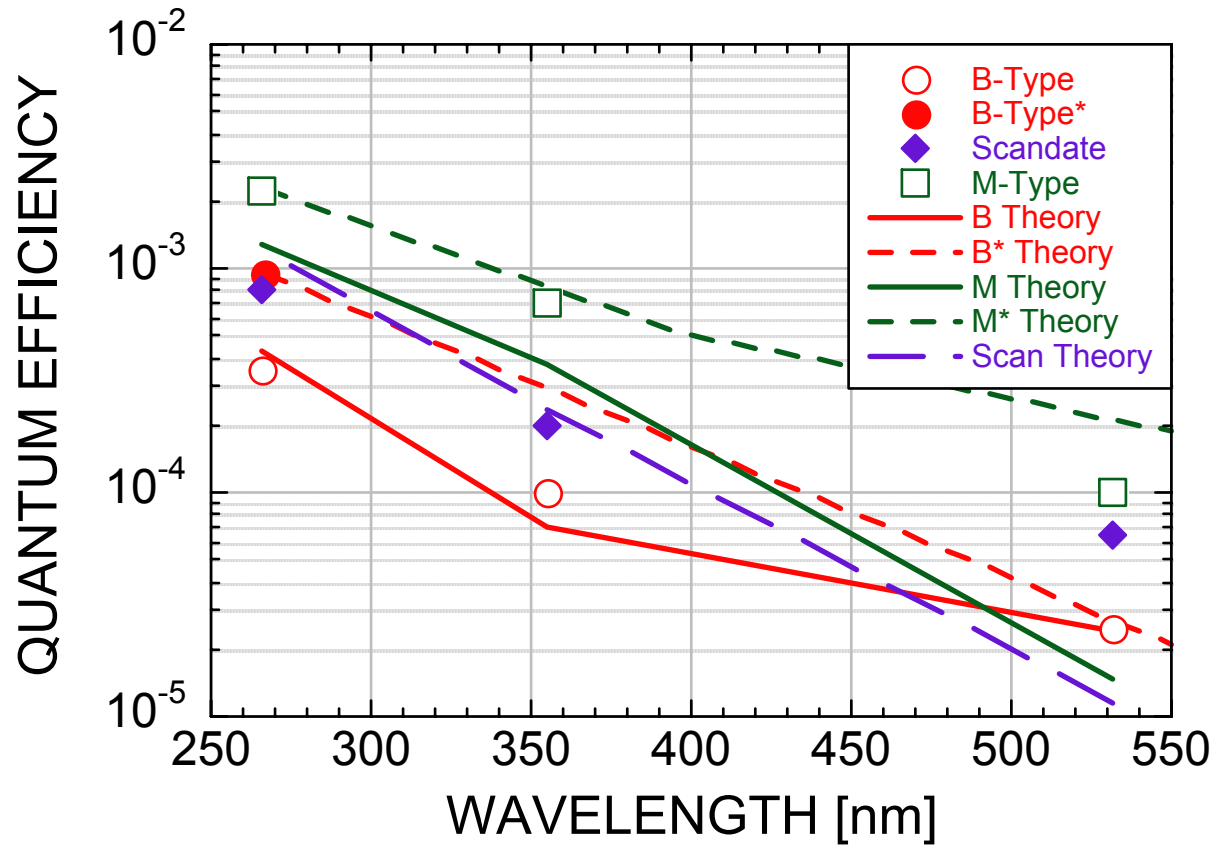
Wavelength [μm]	266.000
Field [MV/m]	3.33333
RInts [MW/cm2]	242.663
Aee [1/sK^2]	0.578593E+09
Bep [1/sK]	0.370169E+13
TD [K]	400.020
Gelion [GW/K cm3]	51842.1
RKappa [W/K cm]	0.746498
Ce [J/K cm3]	0.409377E-01
Ci [J/K cm3]	2.38578
Tau [ps]	0.860155E-03
Elambda [eV]	4.66106

SUBROUTINE ReflectionNK
Wavelength = 0.266000 microns

Angle of incidence =	30.0000Degrees
Reflectivity =	46.1135%
Index of refraction =	3.34361
Extinction Coeff. =	2.44659
Penetration depth =	8.65188nm

SUBROUTINE EVOLVE:

TBC-e [K] =	300.000
TBC-i [K] =	300.000
delt [ps] =	21.0200
Electron: Max val =	719.462
Lattice:Max val =	719.448
ScatFacMax	0.316875E-01
<theta> [%]	71.8834



QE Measured at UMD and in literature for various dispenser cathodes: B-type, M-type, and Scandate

- B-Type: B. Leblond, Nucl. Inst. Meth. Phys. Res. A317, 365 (1992)
- B*-Type: C. Travier, et al., Proc. of 1995 PAC, Vol. 2, p945
- Scandate and M-Type: Measured at U. Maryland
- B and B* Theory: Leblond and Travier parameters, respectively
- M & M* Theory: UMD parameters & UMD + Hi field (50 MV/m)
- Scandate Theory: UMD & Slide 18 parameters, coverage (7%)

EXP - SIM: QE (Flat Metal)

Sample Data set: (Copper)

BULK: Cu (library values throughout)
COATING: Cu (library values throughout)

---

implicit temp-dependent quantities evaluated at RT

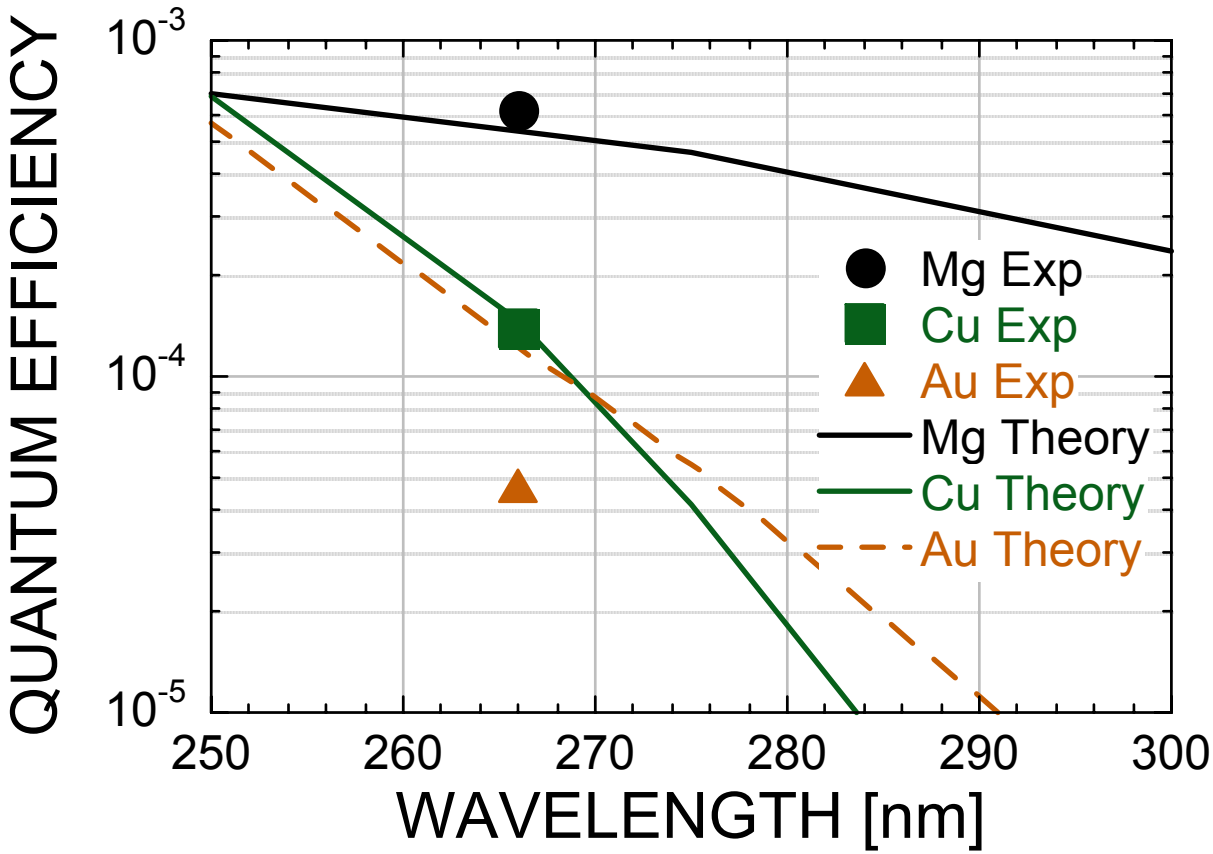
Wavelength [μm]	266.000
Field [MV/m]	5.00
RInts [MW/cm2]	2013.10
Aee [1/sK^2]	0.404536E+08
Bep [1/sK]	0.185989E+12
TD [K]	343.011
Gelion [GW/K cm3]	531.106
RKappa [W/K cm]	4.01838
Ce [J/K cm3]	0.02910
Ci [J/K cm3]	3.28686
Tau [ps]	0.01682
Elambda [eV]	4.66106

SUBROUTINE ReflectionNK
Wavelength = 0.266000 microns

Angle of incidence =	0.0000 Degrees
Reflectivity =	33.6528%
Index of refraction =	1.52728
Extinction Coeff. =	1.67948
Penetration depth =	12.6036 nm

SUBROUTINE EVOLVE:

TBC-e [K] =	300.000
TBC-i [K] =	300.000
delt [ps] =	6.00560
Electron: Max val =	1183.37
Lattice:Max val =	953.414
ScatFacMax	0.15513
<theta> [%]	100%



QE Values for various metals (Gold, Copper, Magnesum)

- T. Srinivasan-Rao, J. Fischer, T. Tsang, ”Photoemission studies on metals using picosecond ultraviolet laser pulses", J. Appl. Phys. 69, 3291, (1990)
- Theory: All parameters taken from published literature for generic metals (e.g., AIP Handbook, 3rd Edition, CRC Tables)…
- …except for field enhancement: Mg = 7.0, Cu = 2.5, Au = 1.0
- Possibility of adsorbate contamination ignored

THE FN, RLD, & WKB APPROXIMATIONS

Field Emission {Fowler Nordheim (FN)} and
Thermal Emission {Richardson-Laue-Dushman (RLD)} Eqs. Dependent upon $T_{wkb}(E)$

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

Fowler Nordheim

$$T(E) \approx \exp\left[-\left(\left(b_{fn}/F\right) + c_{fn}(\mu - E)\right)\right]$$

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

$$J_{FN}(F) = a_{fn} F^2 \exp\left(-b_{fn}/F\right)$$

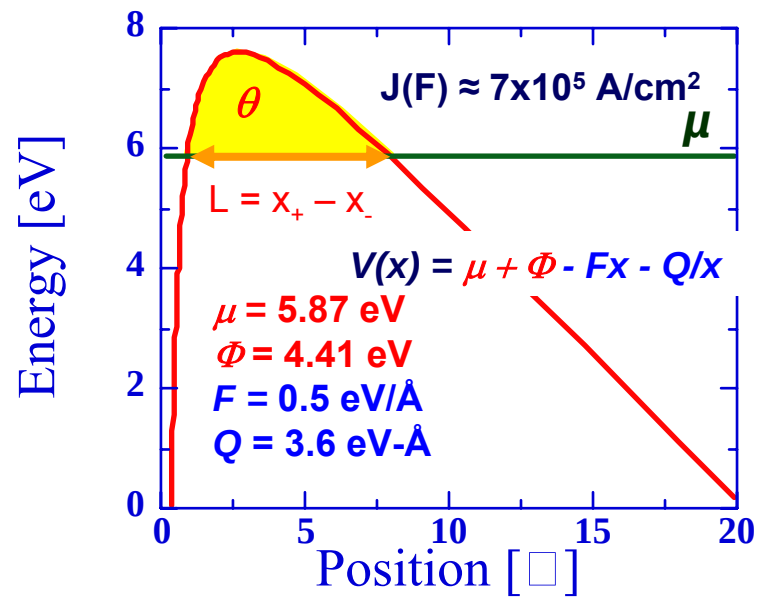
Richardson

$$T(E) = \Theta\left[E - (\mu + \phi)\right]$$

$$f(E) = \frac{m}{\pi h^2}\exp\left[\beta(\mu - E)\right]$$

$$J_{RLD}(F) = A_{RLD} T^2 \exp\left[-\phi / k_B T\right]$$

For Photoemission, FN and RLD asymptotic limits are inadequate



Require T(E) over full range of E

$$T(E) = C(E) / \left\{1 + \exp\left[2\theta(E)\right]\right\}$$

$$\theta(E) = \frac{2}{h}\sqrt{2mFL^3}\, R\left(\frac{x_-}{L}\right)$$

$$R(s) = \int_0^{\pi/2} \frac{\cos^2(\varphi)\sin^2(\varphi)}{\sqrt{s + \sin^2(\varphi)}} d\varphi$$

THERMAL-FIELD ASSISTED PHOTOCURRENT

- Supply Function ($\beta = 1/k_B T_e$)

$$f(E) = \frac{m}{\pi\beta h^2} \ln\left\{1 + \exp\left[\beta(\mu - E)\right]\right\}$$

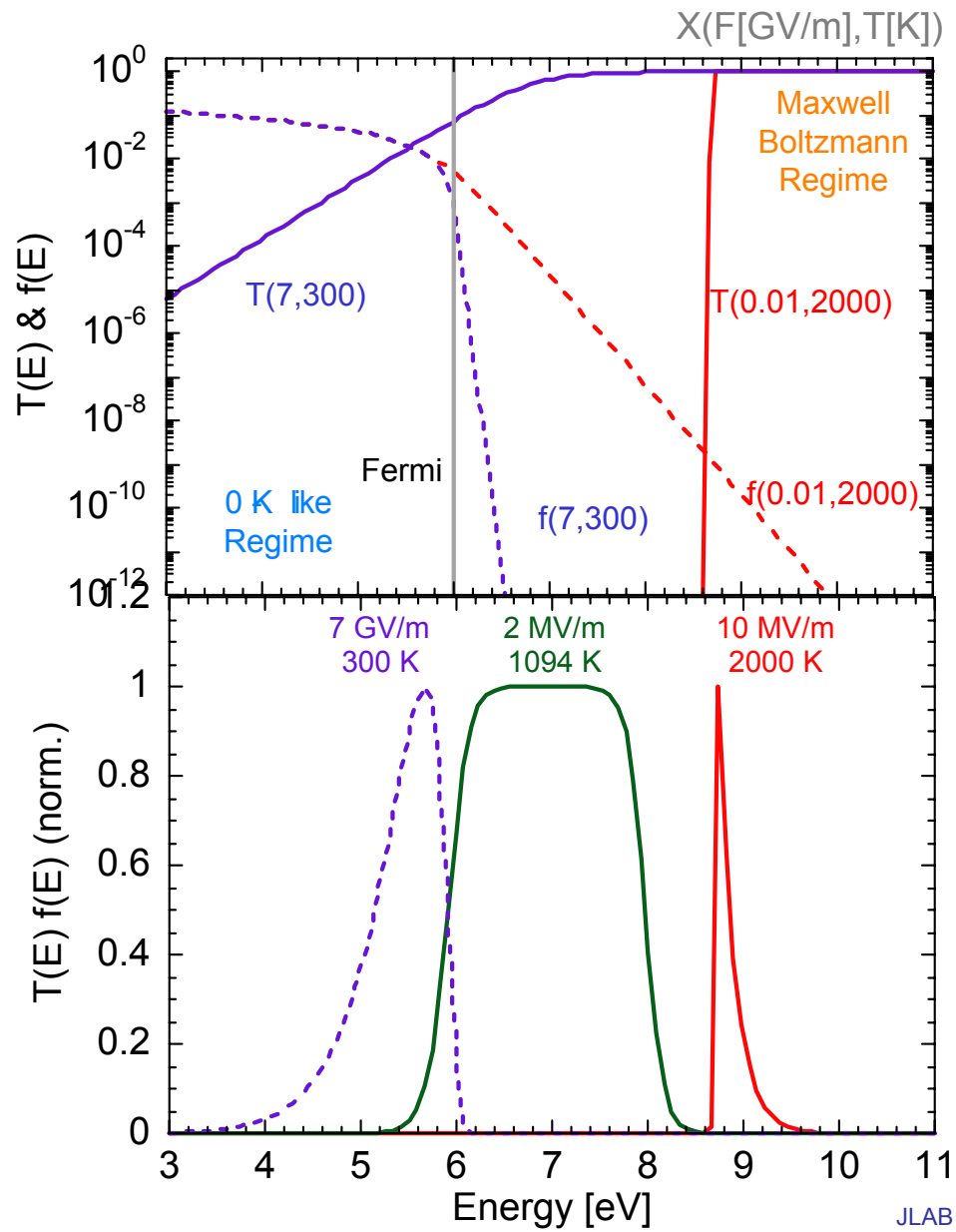
- Transmission Coefficient T(E): (b = slope of -ln[T(E)])

$$T(E) \approx T_o \left\{1 + \exp\left[b(E - E_c)\right]\right\}^{-1}$$

Field $\quad E_c = \mu + \frac{b_{fn}}{F c_{fn}}$

Thermal $\quad E_c = \mu + \Phi - \sqrt{4QF}$

- When β » b: Fowler-Nordheim Eq.
- When b » β: Richardson-Laue-Dushman Eq.
- When b ≈ β : No *simple* analytic form
- Photocurrent: changes T(E) behavior



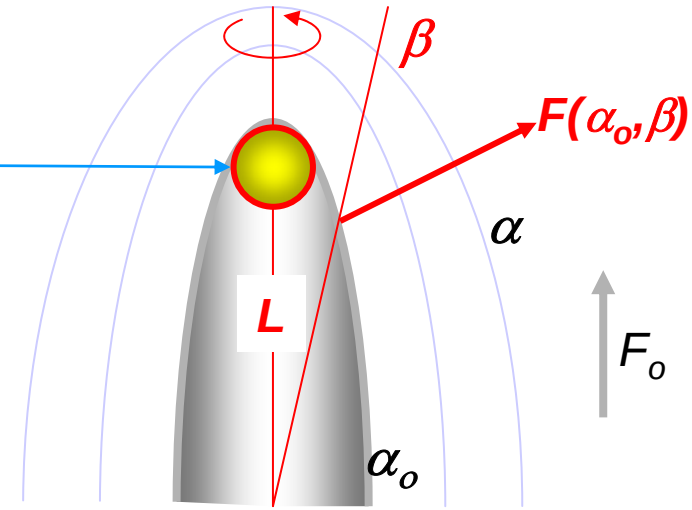
ELLIPSOIDAL MODEL OF NEEDLE / WIRE

Potential and Field Variation Along Emitter Surface Can be Obtained from Prolate Spheroidal Coordinate System

$$z = L\cosh(\alpha)\cos(\beta)$$
$$\rho = L\sinh(\alpha)\sin(\beta)$$

tip radius a_s

$$a_s = L\frac{\sinh^2(\alpha_o)}{\cosh(\alpha_o)}$$



Gradient to Evaluate $F(\alpha,\beta)$

$$\hat{\alpha}\cdot\vec{\nabla} = \frac{1}{\sqrt{(\partial_\alpha x)^2 + (\partial_\alpha y)^2 + (\partial_\alpha z)^2}}\frac{\partial}{\partial\alpha}$$
$$= \frac{1}{L}\left[\sin^2(\beta) + \sinh^2(\alpha)\right]^{-1/2}\frac{\partial}{\partial\alpha}$$

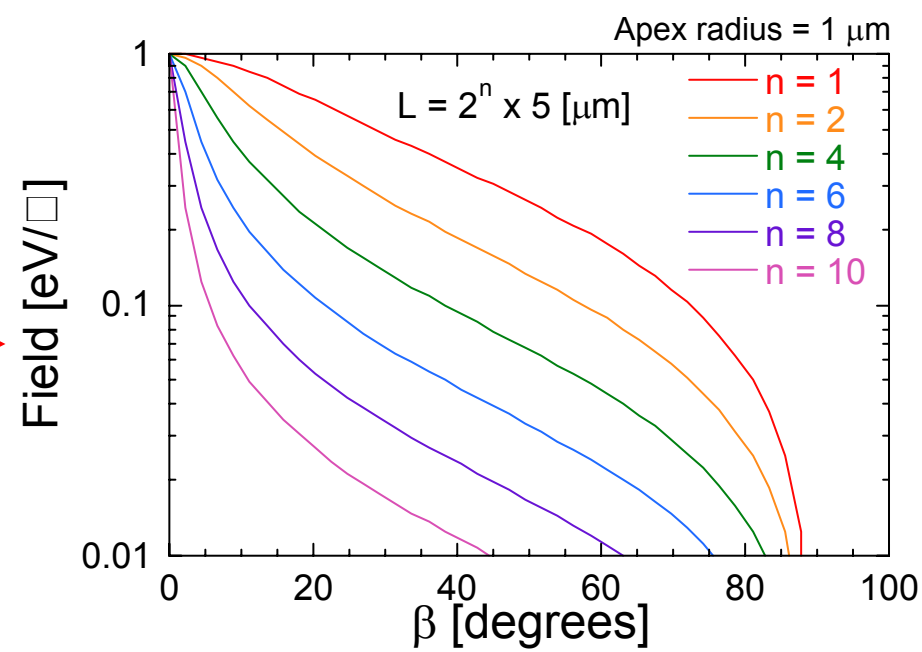
Potential in Ellipsoidal Coordinates

$Q_n(x)$ = Legendre Polynomial of 2nd Kind

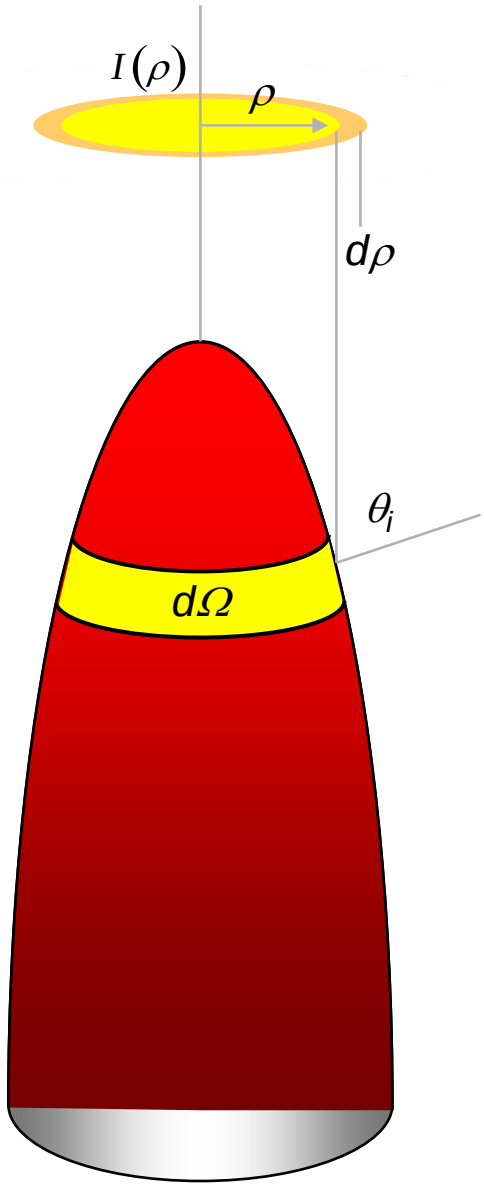
$$V(\alpha,\beta) = V_o - F_o z\left\{\frac{Q_1(\cosh(\alpha))}{Q_1(\cosh(\alpha_o))} - \frac{\cosh(\alpha)}{\cosh(\alpha_o)}\right\}$$

Field Along Surface of Emitter

$$F(\alpha_o,\beta) = \frac{\sinh(\alpha_o)\cos(\beta)}{\sqrt{\sinh^2(\alpha_o) + \sin^2(\beta)}}F_{tip}$$
$$F_{tip} = -\frac{F_o}{\sinh^2(\alpha_o)Q_1(\cosh(\alpha_o))}$$



OTHER FACTORS



FACTORS AFFECTING EMISSION CURRENT

$$QE \propto \frac{\text{total emitted charge}}{\text{total incident energy}} \approx \frac{\sqrt{\pi}\Delta t_q \int_\Omega J(T,F,\hbar\omega)d\Omega}{\sqrt{\pi}\Delta t_\omega \int_\Omega I(R,\Delta\rho,\hbar\omega)d\Omega}$$

- Differential surface area illuminated

$$d\Omega = 2\pi\rho\sqrt{d\rho^2 + dz^2}$$

prolate spheroidal analysis

- Intensity on differential element

$$I_{surf}(\rho) = [1 - R(\theta)]I(\rho)\frac{2\pi\rho d\rho}{d\Omega}$$

index of ref & penetration

- Variation in illumination intensity

$$I(\rho) = I_o(\lambda)\exp\left[-(\rho/\Delta\rho)^2\right]$$

dictated by experiment
(weak variation for small tips)

- Angular variation of reflection coefficient R: determination of incidence angle

$$\tan(\theta) = -\frac{dz}{d\rho} = \frac{\tan(\beta)}{\tan(\alpha_o)}$$

prolate spheroidal analysis

- Electron Gas Temperature

$$T(\rho,\lambda) = T_{bulk} + CI_{surf}(\rho)^p \exp\left\{-\frac{1}{2}\left(\frac{t-t_o}{\Delta t_T}\right)^2\right\}$$

laser - material interaction & time dependent model

TEMPERATURE VS INTENSITY (time dep)

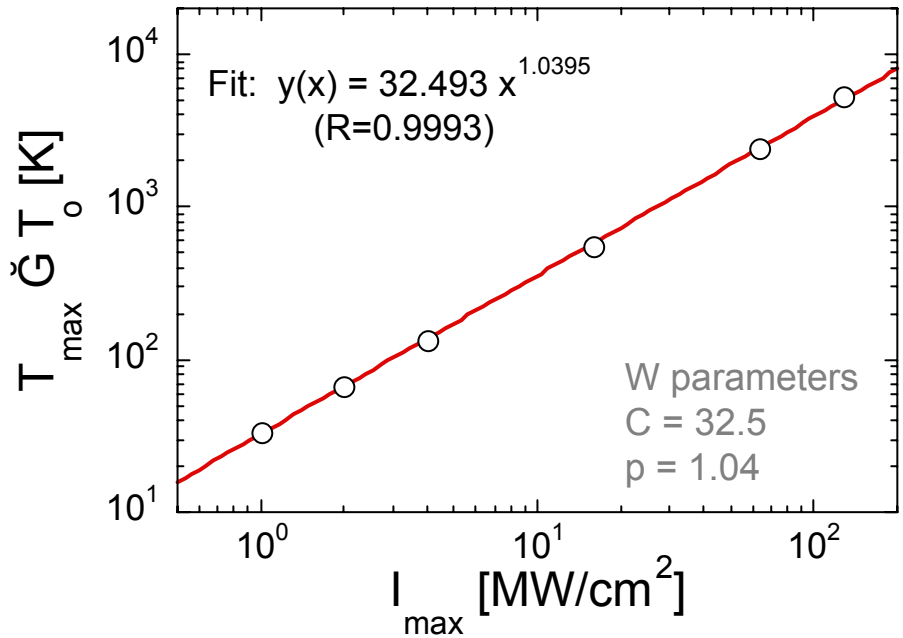
Leading Order Behavior of T(t)

- At high T, $C_e(T) = \gamma T$: Suggests to leading order that dependence of T^2 mimics I(t) (but off-set by t_o)

$$C_e \frac{\partial}{\partial t} T_e = \frac{\partial}{\partial z}\left(\kappa \frac{\partial}{\partial z} T_e\right) - g\left(T_e - T_i\right) + G\left(z,t\right)$$

$$\frac{\partial}{\partial t} T_e^2 \propto I\left(t - t_o\right) \Rightarrow \frac{T(t) - T_{\min}}{T_{\max} - T_{\min}} = \exp\left[-\frac{1}{2}\left(\frac{t - t_o}{\Delta t}\right)^2\right]$$

- Maximum Temperature is linearly (almost) related to intensity:



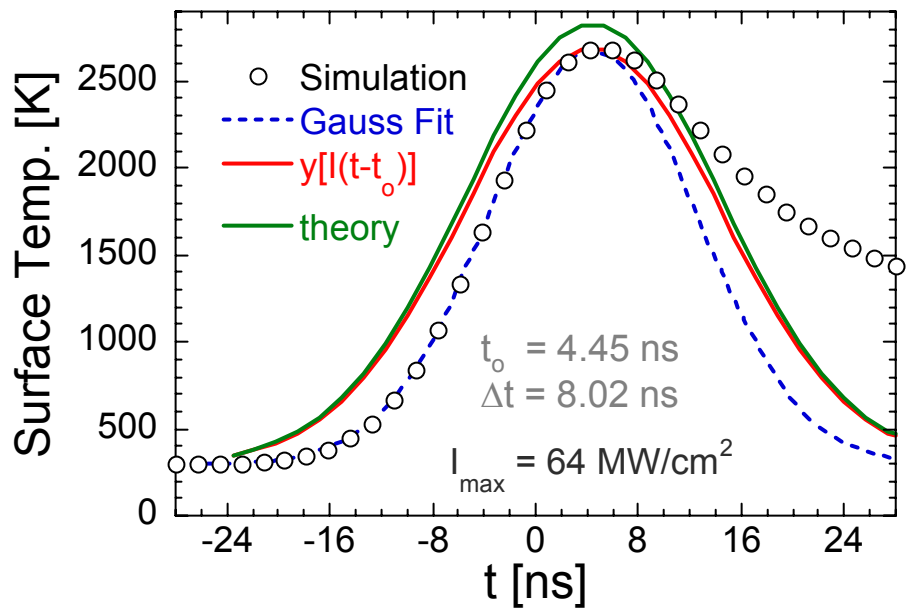
Graphical Analysis to find T_{max}:

$$\text{Gauss Fit} = T_O + \Delta T \exp\left\{-\frac{1}{2}\left(\frac{t - t_o}{\Delta t}\right)^2\right\}$$

$$y\left[I(t - t_o)\right] = T_o + \Delta T \left(\frac{I\left(t - t_o\right)}{I_{\max}}\right)^{1/2}$$

use this => $$\text{theory} = T_o + C I_{\max}^p \left[\frac{I(t - t_o)}{I_{\max}}\right]^{1/2}$$

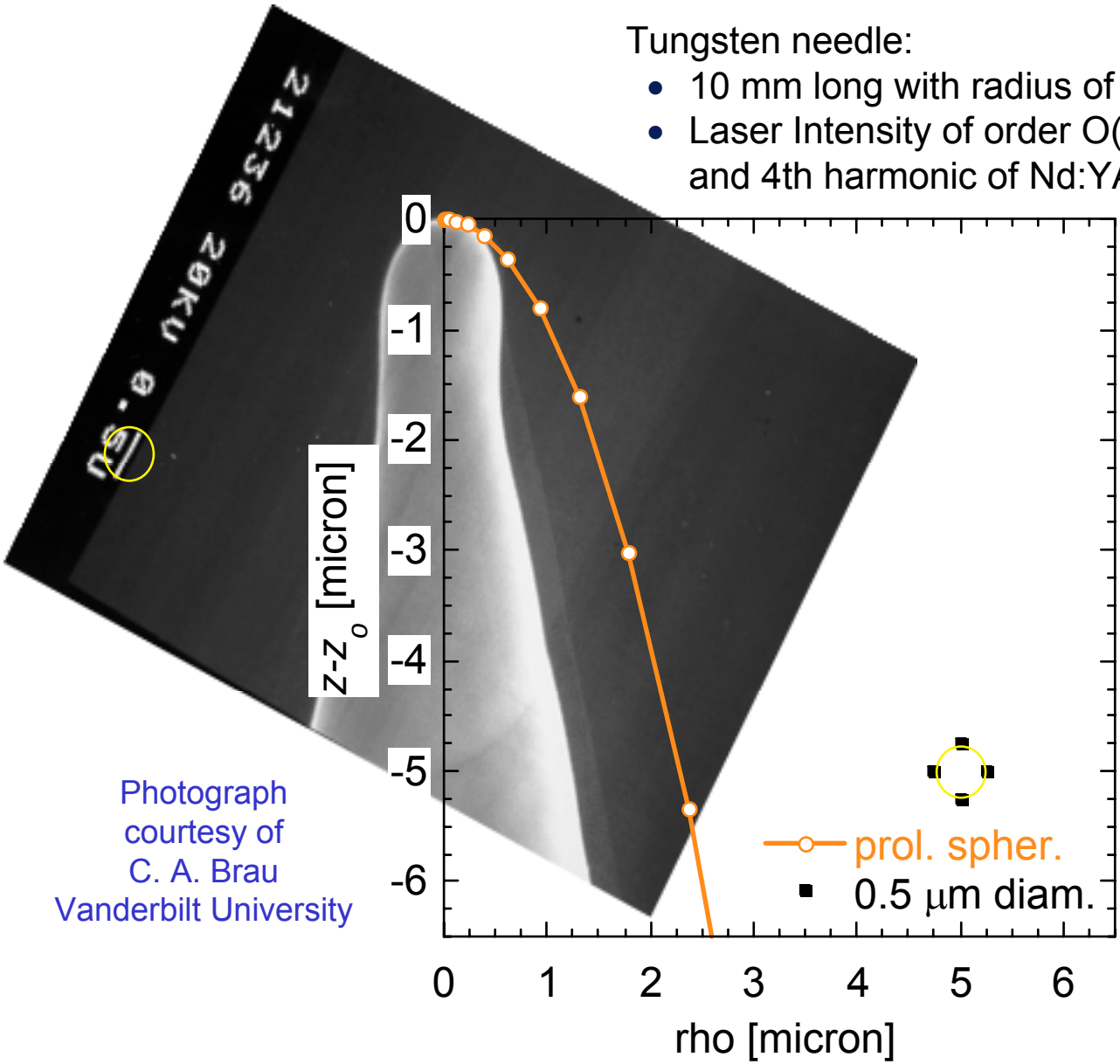
ΔT is evaluated for I_o = 64 MW/cm^2



FIELD-ASSISTED PHOTOEMISSION FROM W

Tungsten needle:

- 10 mm long with radius of curvature at apex = O(1 μm)
- Laser Intensity of order O(100 MW/cm^2) over O(10 ns) and 4th harmonic of Nd:YAG (λ = 266 nm)



Photograph courtesy of C. A. Brau Vanderbilt University

Other Factors:

- Cathode to anode separation ≈ 35 mm
- Max Anode ≈ 33 kV (F_o = 0.94 MV/m)
- Match between prolate spheroidal approx. & actual tip is reasonable
- Constraints of side walls, temperature at apex, etc. result in best estimate of a_s = 0.53 μm

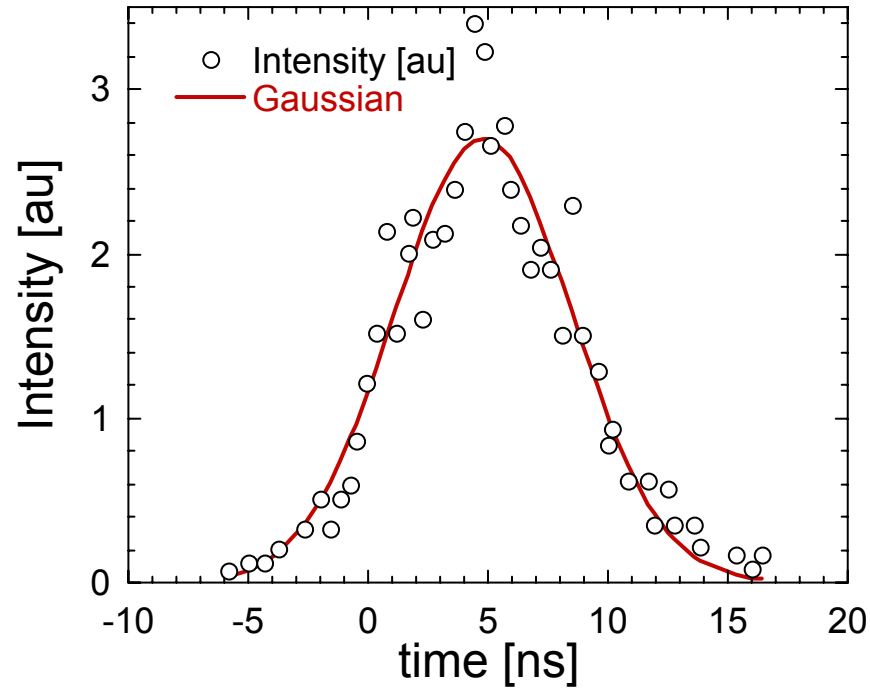
LASER PARAMETERS (EXP)

Experiments on Field Assisted Photoemission on W needle:
C. Hernandez-Garcia, C. A. Brau (Vanderbilt University)

Relevant Publications by Garcia & Brau in Nuclear Instruments and Methods in Physics Research:

- NIMA 429 (1999) 257-263
  Photoelectric Field Emission From Needle Cathodes
- NIMA 475 (2001) 559–563
  Electron Beams Formed by Photoelectric Field Emission
- NIMA 483 (2002) 273–276
  Pulsed Photoelectric Field Emission From Needle Cathodes

See Also:
T. Inoue, S. Miyamoto, S. Amano, M. Yatsuzuka, T. Mochizuki, Jpn. J. Appl. Phys. 41, 7402-7406 (2002) "Enhanced Quantum Efficiency of Photocathode under High Electric Field"



Laser intensity vs time, from Fig. 1 of NIMA483
Wavelength = 266 nm; Current Max = 112 mA.

Gaussian Fit:

$$I(t) = I_o \exp\left[-\left(\frac{t-t_o}{\Delta t}\right)^2\right]$$

$I_o = 2.7047$ au
$t_o = 4.8036$ ns
$\Delta t = 5.2239$ ns

COMPARISON

Reference Point:

- V(ref) = 17.0 kV
- F(ref) = 0.199 GV/m

Simulation: Macro Q(ref):

- Q(266) = 0.528374 %
- Q(355) = 1.74e-03 %

Exp: Macro Q(ref) @ 266:

- Current at Peak = 0.112 A
- Intensity = 32 MW/cm^2
- Gaussian Laser spot 50-100 microns (1/e) (depending on λ): let $\Delta\rho$ = 25 microns (radius)

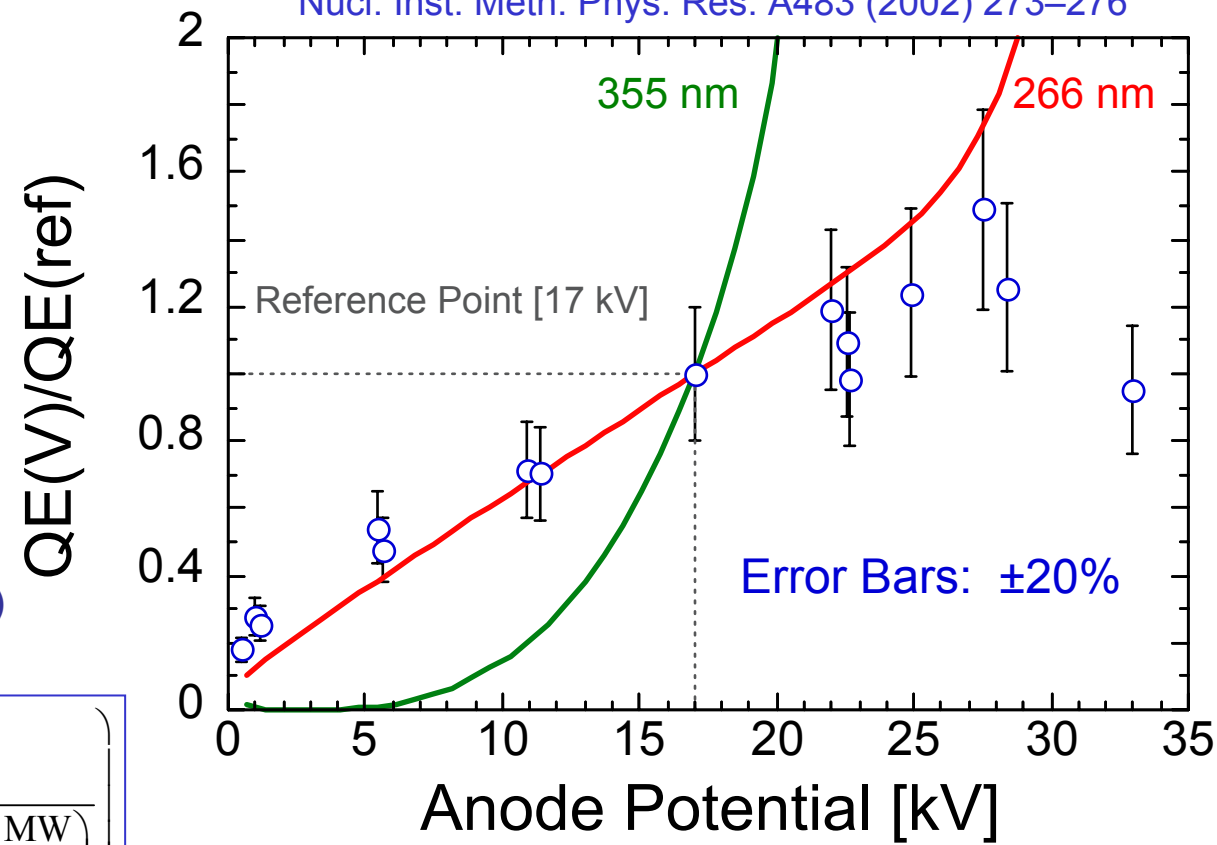
Macro QE Estimation

$$QE[\%] = \frac{200\pi hc}{q(0.266\mu m)}\left(\frac{0.112\,\text{Amp}}{\pi(25\mu m)^2\left(32.2\,\frac{\text{MW}}{\text{cm}^2}\right)}\right)$$

$$= 0.0826\ \%$$

Laser Illuminated W Needle Simulation And Experimental Data†

†C. Hernandez-Garcia, C. A. Brau
Nucl. Inst. Meth. Phys. Res. A483 (2002) 273–276



355 comparison used same R, scat fac.,penetration depth, etc. as 266 and is therefore only qualitative

SUMMARY

Photoinjectors are, or are becoming, important electron sources for…

- Synchrotron Light Sources
- Free Electron Lasers
- X-ray Sources, etc.

…due to high quality beams that can be achieved: Generating a High Quality Beam Right From the Start Is Important

Collaborative Exp. / Theory (NRL & UMD) Research Program:

- (EXP) investigate photocathode technologies and behavior of dispenser cathodes as photoemitters in particular; quantify emitter surface and photoemissive properties (quantum efficiency, emission, etc.); custom design photocathodes
- (THEORY) photoemission theory & photocathode code treating (i) emitter surface (ii) emitted distribution & beam characteristics (iii) factors which modify surface

IMPORTANCE OF PRESENT PHOTOCATHODE PROGRAM

- Megawatt-class FEL's need development of long-lived, robust, in situ repairable, low emittance, high rep rate photocathodes: Such cathodes are presently unavailable.
- To develop such cathodes, a time-dependent model, validated and coupled to experiment, is necessary to predict and characterize temporal response, quantum efficiency, behavior, etc