Developing Field Emission Models Employing Nanoscale Surface Characterization

Supported by the Laboratory Directed Research and Development program at Sandia National Laboratories, a multimission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC., a wholly owned subsidiary of Honeywell International, Inc., for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-NA-0003525.

Additional Support: CINT User Facility (DE-AC04-94AL85000)

SAND2021-4704 C

Matthew Hopkins, Ezra Bussmann, Taisuke Ohta, Ashish Jindal, Nicholas Roberds, Christopher Moore

Sandia National Laboratories

13th International Workshop on Breakdown Science and High Gradient Technology (HG2021). April 19-21, 2021.

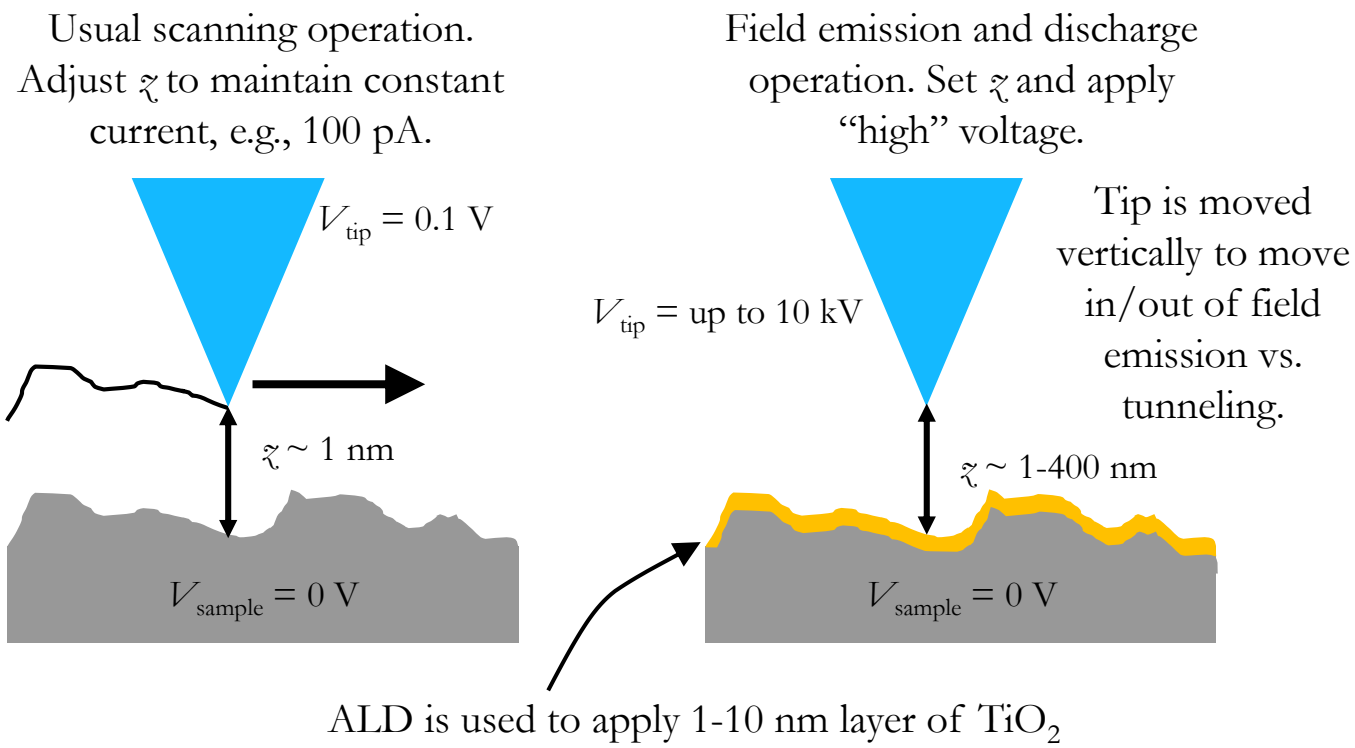
Vacuum Arc Initiation

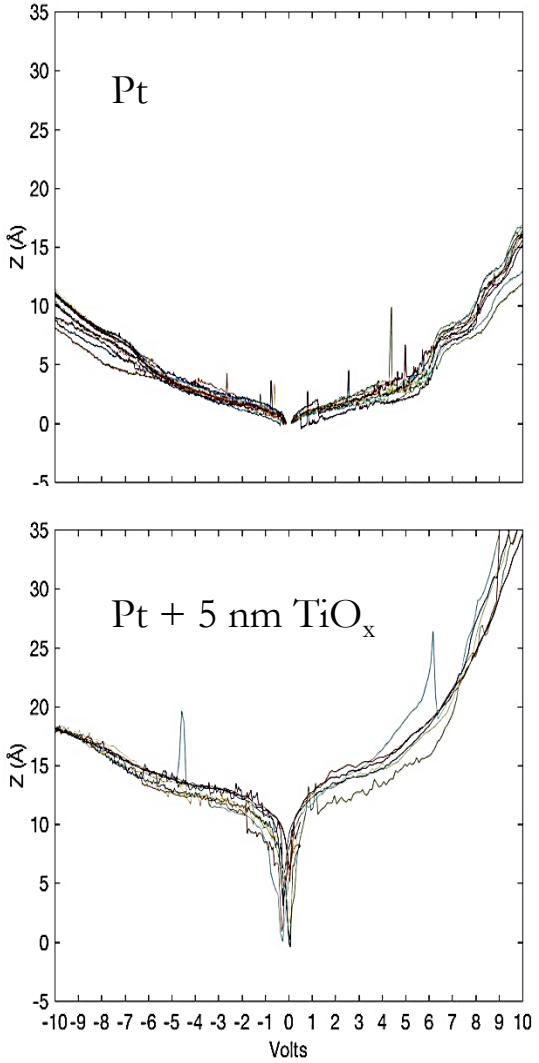
- We are interested in modeling a variety of discharge situations: from streamers at atmospheric pressure to vacuum arcs
- Vacuum discharge is critical to many modern devices.
  - Critical failure mechanism → Want to avoid
  - Mode of operation → Want to have predictable behavior
- We want to understand vacuum field emission from well-characterized surfaces to create physics-based models for use in large-scale PIC-DSMC breakdown simulations
  - Field emission is necessary precursor to a breakdown event. No field emission → no breakdown.
  - Employ Scanning Tunneling Microscopy (STM) and Photoemission Electron Microscopy (PEEM) to characterize surface very locally, and then apply high fields to initiate breakdown. Very locally ~0.1-10 nm.
  - Address the problem of not knowing the state prior to discharge at the location of discharge by characterizing and then discharging.
  - Apply known layers of dielectric (e.g.,TiO_2, MgO) to challenge models and begin investigation of role of surface contaminants and oxide layers.
  - Utilize a "meso-scale" (0.1-1.0 μm) model of the surface for PIC-DSMC simulation of breakdown.

RF vs. DC breakdown
"breakdown rate vs. conditioning" vs. "single shot probability vs. surface state".
Then pulsed DC.

Vacuum Field Emission Measurements via STM

- A pin-to-plane configuration in a scanning-tunneling microscope (STM) is employed. We also incorporated atomic layer deposited (ALD) surface films.



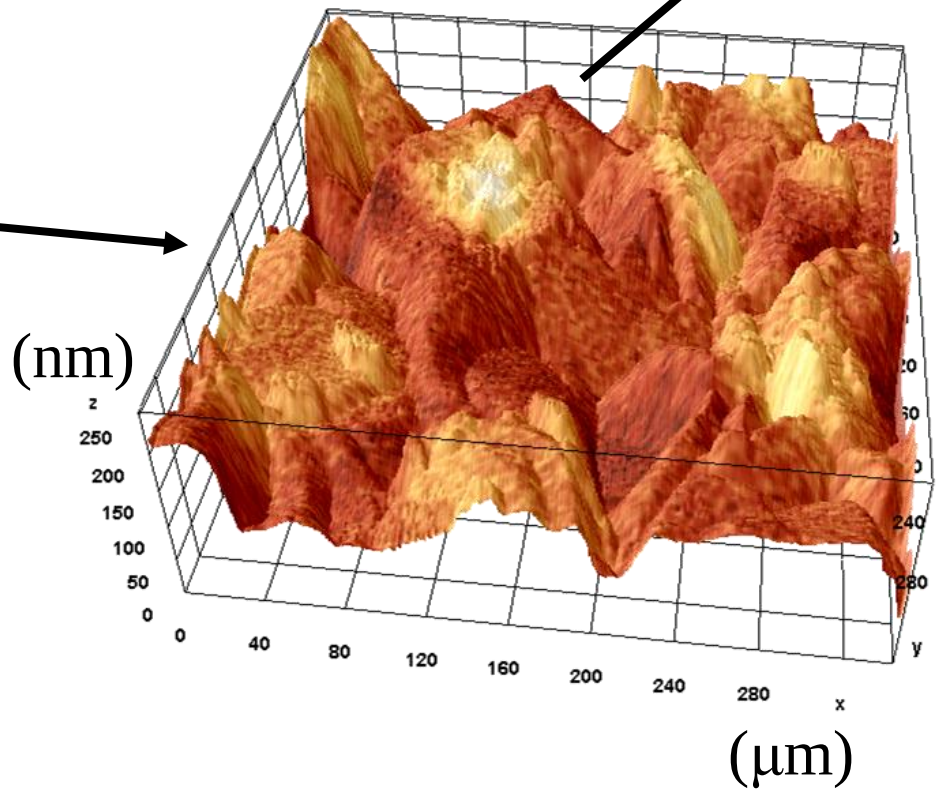


Why Local Characterization?

- Fowler-Nordheim field emission:
  - Typical used in macroscale models to curve-fit measured $j(E)$ from the as-built electrode
  - Can result in β ~10-1000 !!!

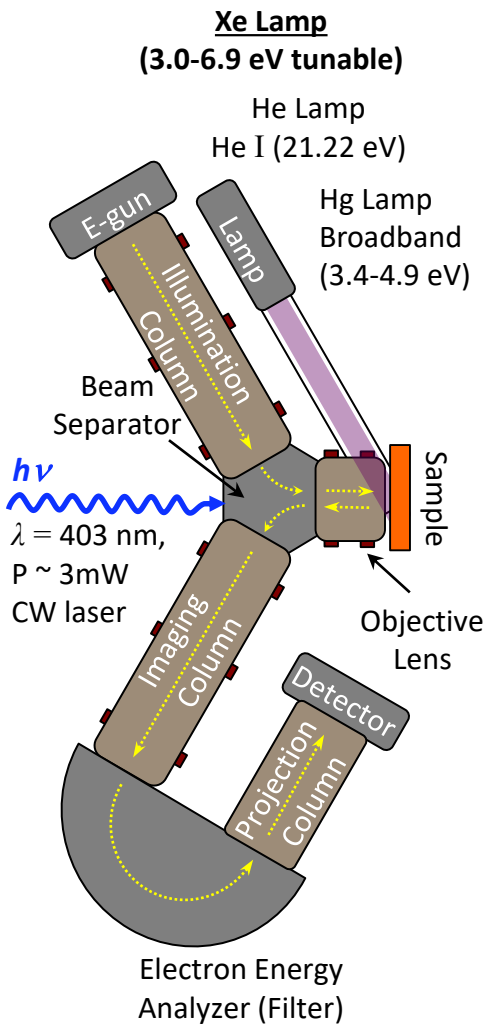
$$i = A_{eff} A_{FN} \frac{(\beta E)^2}{\phi t^2(y)} exp\left[-\frac{B_{FN} v(y) \phi^{3/2}}{\beta E}\right]$$

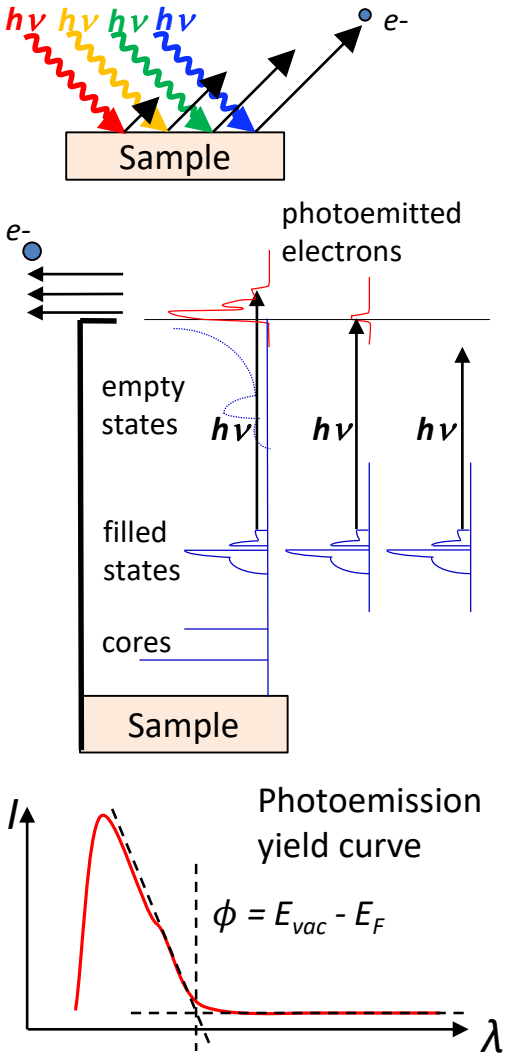
- We want to locally characterize the surface to eliminate β as a fit parameter:
  - Use scanning tunneling microscope (STM) and/or atomic force microscopy (AFM) to measure topology at <= 10 nm resolution.
  - By meshing the microscopy surface and solving E-fields local to that surface, β comes out naturally – no need to "fudge" it.

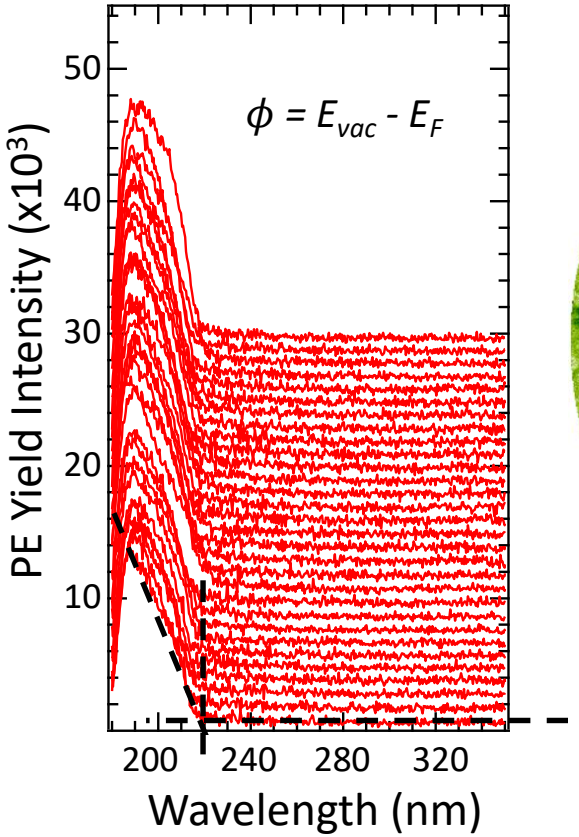


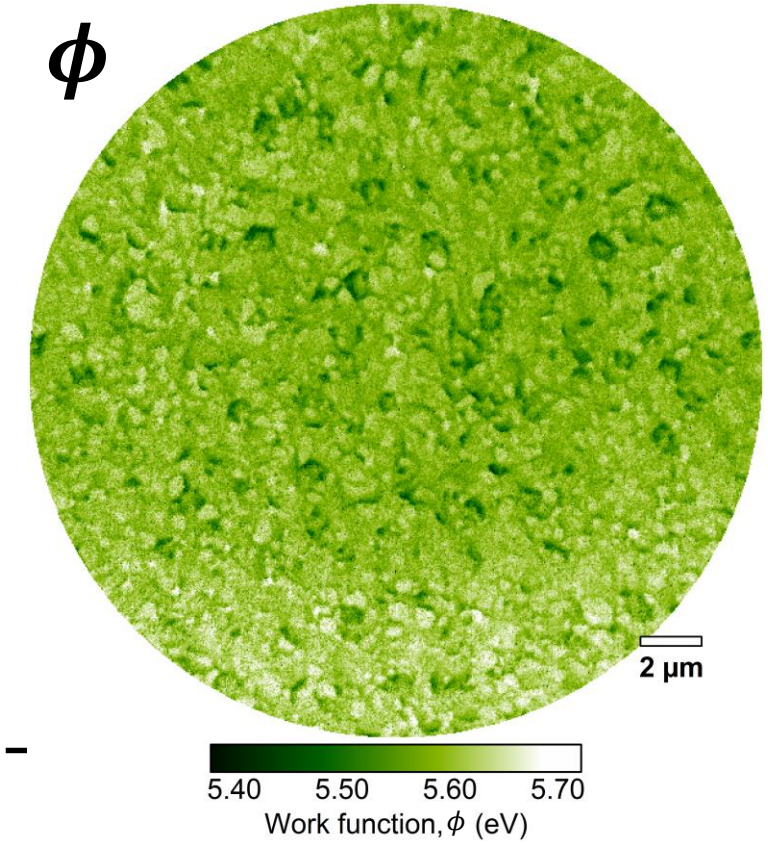
Pixel-resolved ϕ using spectroscopic PEEM

PEY($h\nu$ sweep)







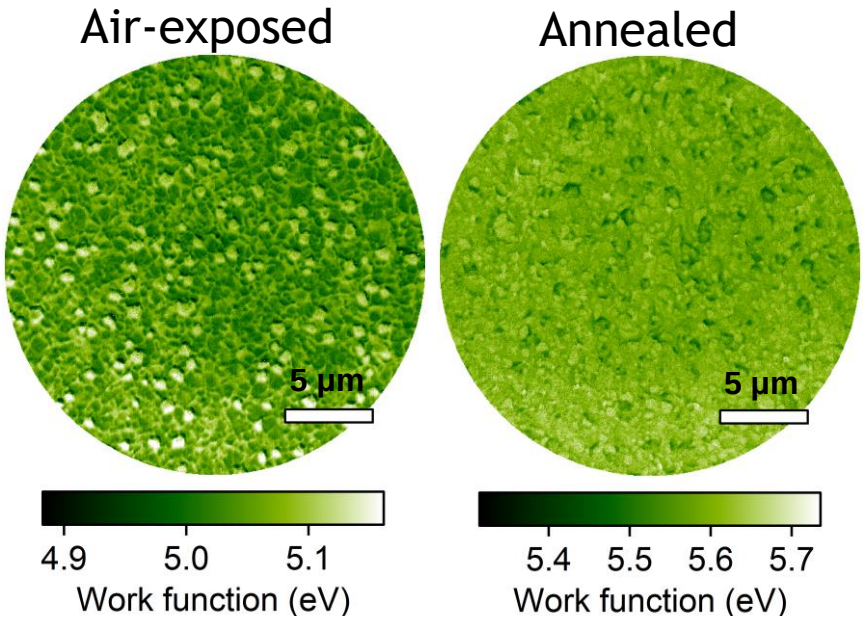


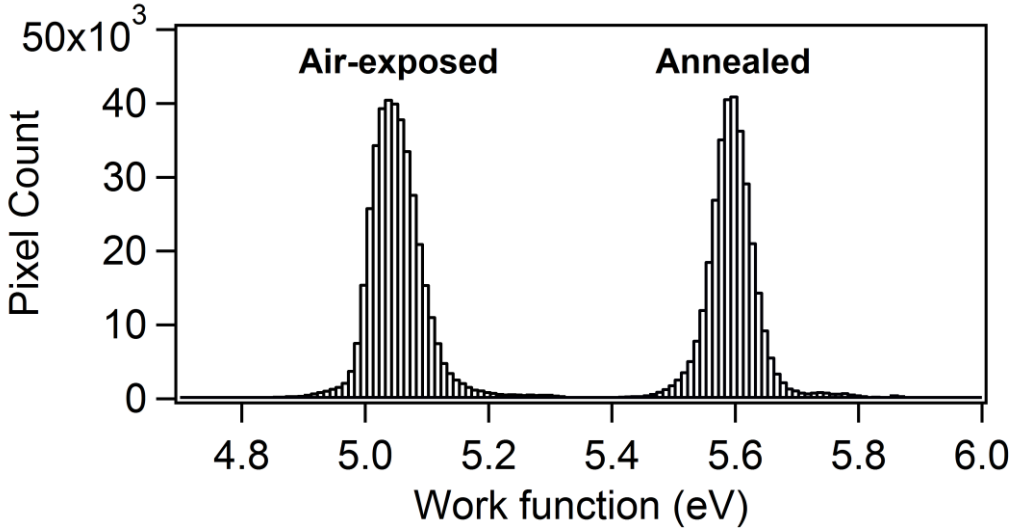
24 µm FOV, 40 nm pixel maps

PEEM Measurement of Work Function Variation

- Measured spatial variation of local work function using photoemission electron microscopy (PEEM)
  - Variation across given Pt surface relatively small – only a few percent
  - However, ϕ is in the exponential and the tail of the distribution can initiate field emission and eventually breakdown
- Significant (~10%) decrease in the work function due to surface contaminants picked up via exposure to air
- Use the ~10 nm-scale PDF's in mesoscale model to set element work functions in PIC-DSMC simulations

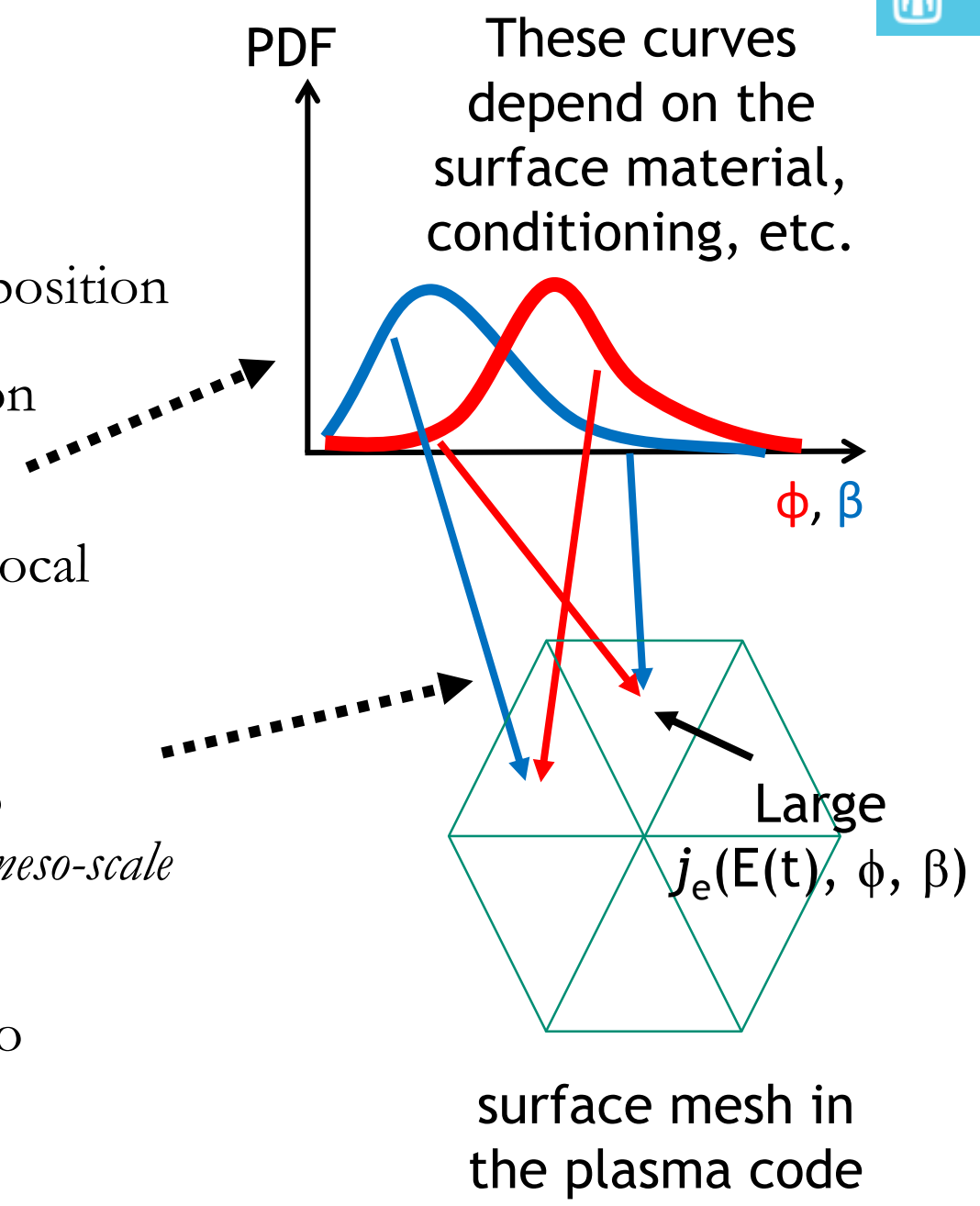
Poly-Pt (111) on $ZnO/SiO_2/Si$





Model Development

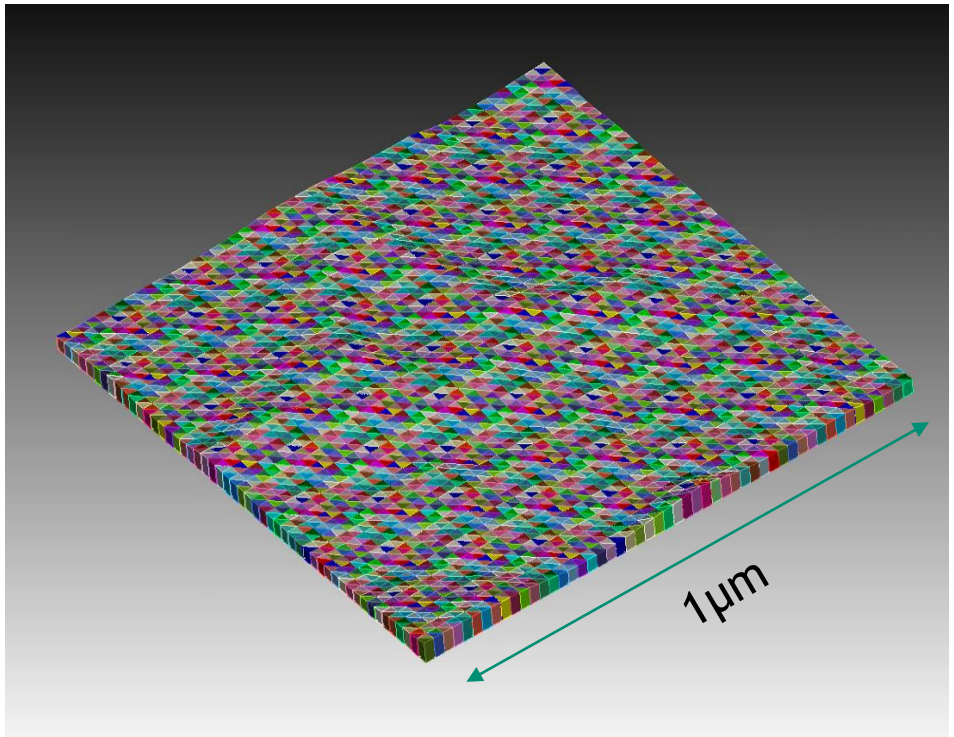
- Create Pt electrode via sputter deposition
- Controllably contaminate Pt via Atomic Layer Deposition
- Measure work function, local topology, and electron emission for sample
- Generate probability density functions (PDF) for local work functions and effective topological field enhancement
- Incorporate measured *atomic-scale* distributions into discharge simulations by populating time-varying *meso-scale* element-based data from the PDFs
- Compare family of plasma discharge simulations to measured breakdown behavior



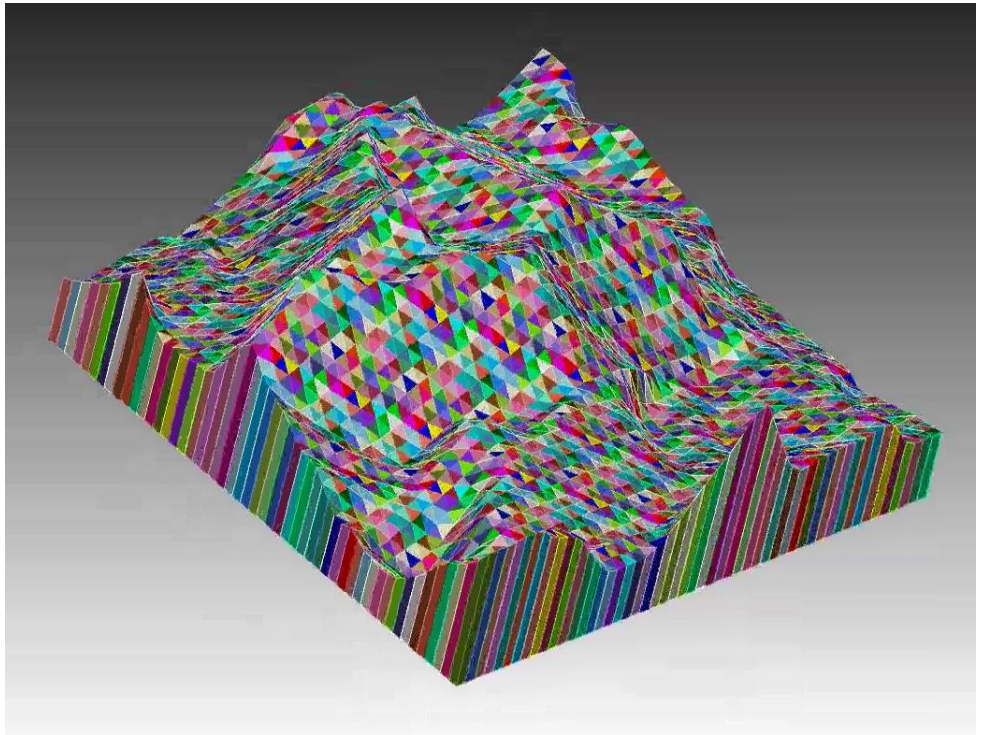
AFM Surface Characterization

- Took the AFM (x,y,z) spatial points (here ~20 nm resolution) and map into Cubit meshing software
- Actual surface has virtually no significant topology – we will see later that β ~1 everywhere
- To demonstrate significant spatial variation of field emission across the surface we also compute results with the surface relief multiplied by 10×

As-measured surface relief

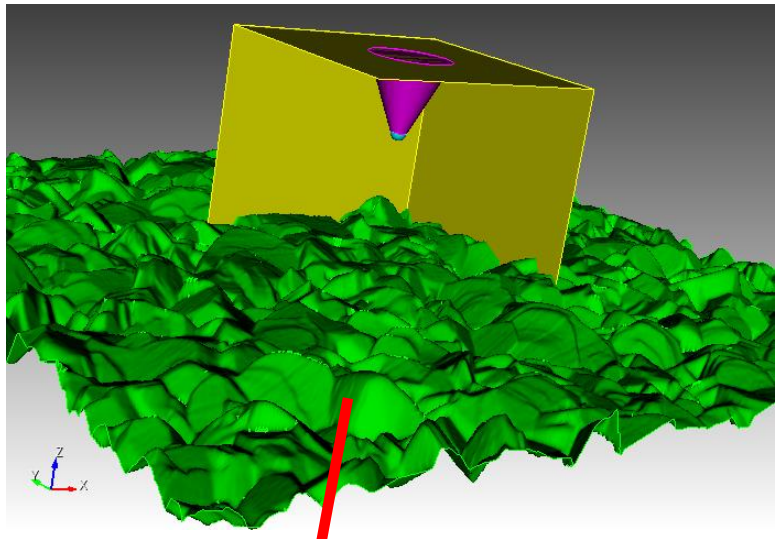


Surface relief increased by 10×

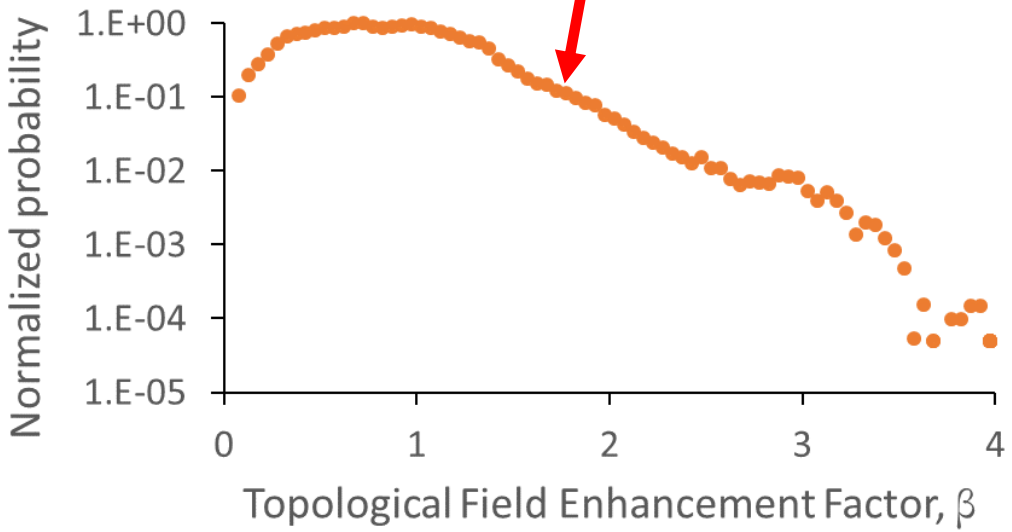


AFM Topology → Topological Atomic-Scale β

- Planar anode ~10 um above surface (ignore tip in pic!)
- Compute E_{norm} and A_{proj} for every element face in the resolved STM mesh
  - < 10 nm elements; ~600K surface faces
- Get projection factor, $f_{proj} = \frac{\sum_{faces} A_{face}}{\sum_{faces} A_{proj,face}}$
  - For present data f_{proj} ~ 1.15
- Create ~20 nm scale PDF of $\beta = \frac{E_{norm}}{E_{applied}}$
- Some elements will have β < 1
  - Globally the surface could be tilted
  - Sides of "sharp" atomic features



Electrostatic solve



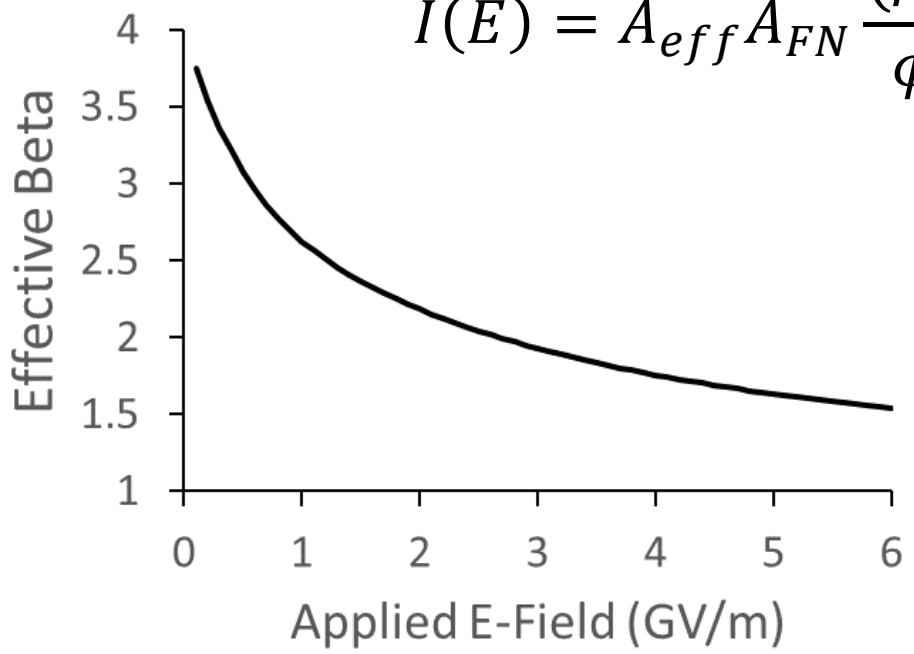
Mesoscale Model for Surface Variations

- We have measured atomic-scale (1-10 nm) PDFs of the work function and topological field enhancement factor
- Must convert these to the mesoscale (0.1-10 μm). Some options:
  1. Just pick the meso-scale β and ϕ from the atomic-scale PDFs
  2. Make an effective β and ϕ to use at the meso-scale
  3. "Brute force" – for each meso-scale element face, pick N local emitters (unique β's and ϕ's)
- Option #1 obviously has artificially large variation for different surface realizations in simulations. We will not consider it further.
  - Sometimes get an extreme tail value and then field emit based on the mesoscale element's area
  - Other times there will be no tail values picked and no field emission until much higher fields
- We will proceed to do #2 and #3 and compare to resolved $\beta = 1$ everywhere.

Mesoscale Model for Surface Variations

- Can we make an effective β (and ϕ) from the data and/or atomic-scale β PDFs?
- Measure/compute the total field emission current versus $E_{applied}$
- Non-linear solve for β_{eff}:

$$I(E) = A_{eff} A_{FN} \frac{(\beta_{eff} E)^2}{\phi t^2(y)} exp\left[-\frac{B_{FN} v(y) \phi^{3/2}}{\beta_{eff} E}\right]$$



→ β_{eff} depends on $E_{applied}$!

- This makes sense: small β regions "turn on" at higher fields and pulls the effective β lower
- The precise functional form depends on the atomic-scale β PDF

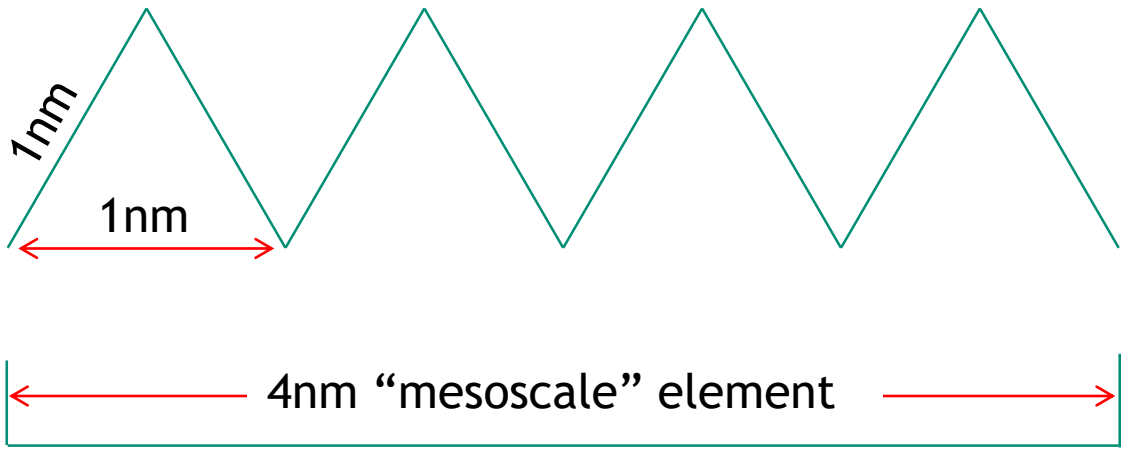
e.g. see: Feng and Verboncoeur, PoP **13**, 073105 (2006)
Jinpu Lin et al., J. Appl. Phys. **121**, 244301 (2017)

Mesoscale Model for Surface Variations

- We are left with "brute force" -- for each mesoscale element face, pick N local emitters (randomly pick unique β's and ϕ's) from the atomic-scale measured distributions:

$$N = \frac{A_{element}}{A_{resolved}} f_{proj}$$

- Must scale the number of local emitters to draw:



8 local faces that the β and ϕ PDF created from

$$f_{proj} = \frac{\sum_{faces} A_{face}}{\sum_{faces} A_{proj,face}} = 2$$

Draw 8 local emitters

Mesoscale Model for Surface Variations

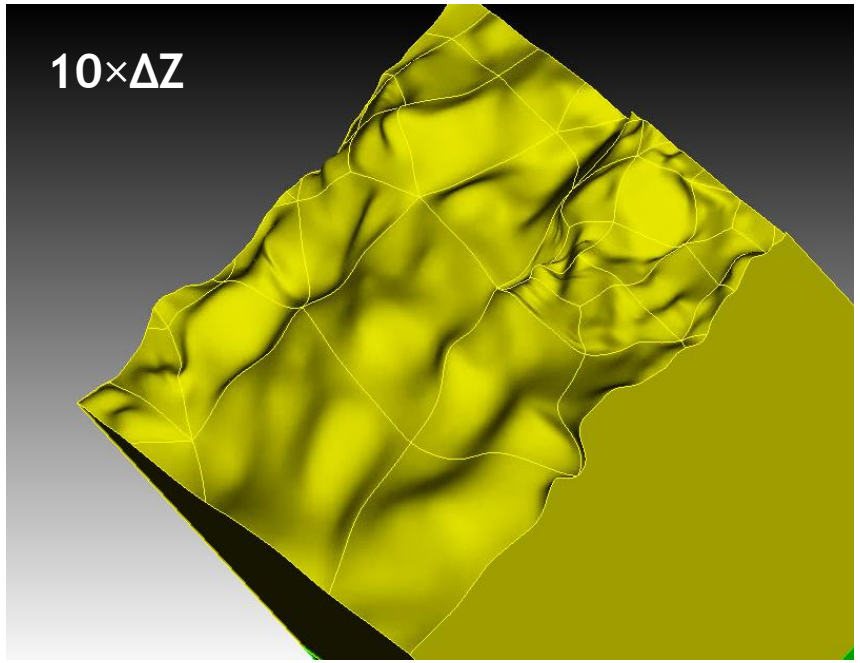
- However, we don't have to store all N local emitters for each surface element face
  - Field emission is highly nonlinear, and most emitters (β and ϕ) can be neglected
- Store every atomic-scale emitter (β and ϕ) that appreciably contributes to the current
  - A threshold current contribution of 0.1% results in storing ~0.01% of the atomic-scale emitters
  - 1 μm^2 element has 10^4–10^6 atomic-scale emitters → store < 1000 emitters.

- PIC field emission algorithm each Δt:
  - Compute E_{norm} on each surface element face
  - Loop over all ~100 atomic-scale emitters:

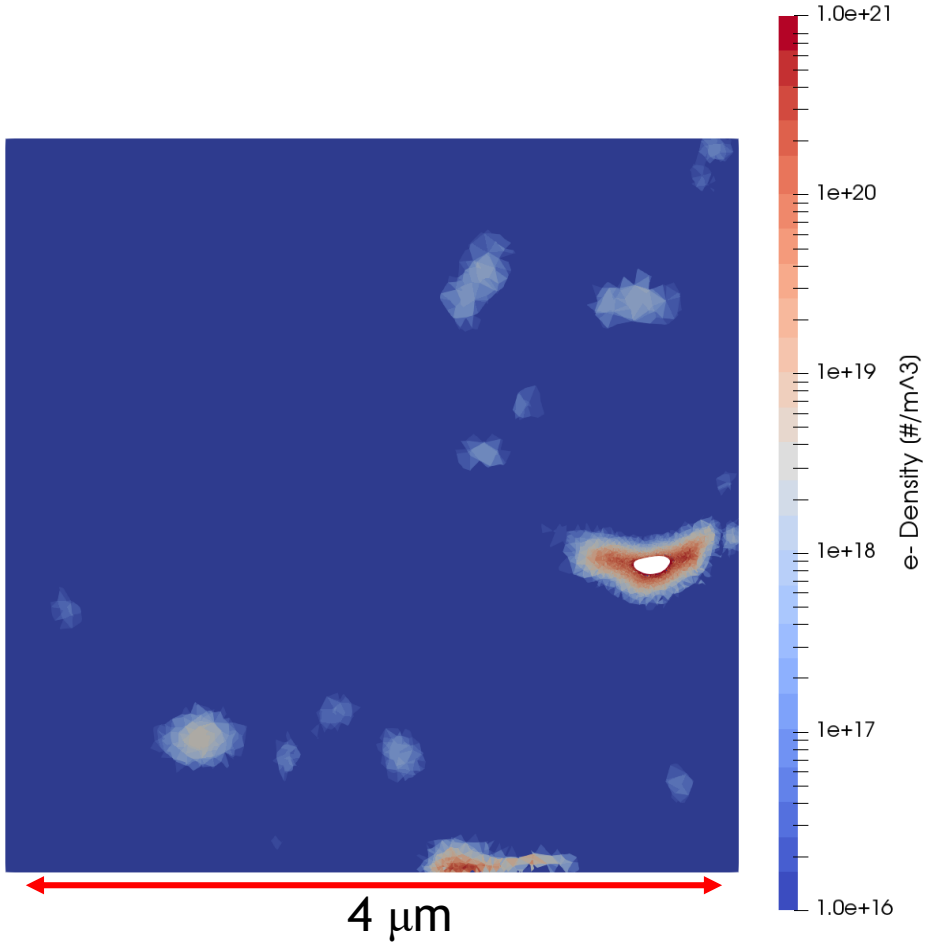
$$I_{face} = \sum_{emitters} A_e A_{FN} \frac{(\beta_e E_{norm})^2}{\phi_e t^2(y)} exp\left[-\frac{B_{FN} v(y) \phi_e^{1.5}}{\beta_e E_{norm}}\right]$$

Simulation of Emission from AFM Surface

- With the resolved ($\Delta x < 10$ nm) mesh, simulate the emission from the AFM surface
  - Show contours of e^- density just above the cathode surface
  - Some clipping of the topology is seen for the largest feature
- See several large-scale features that emit, otherwise very little emission



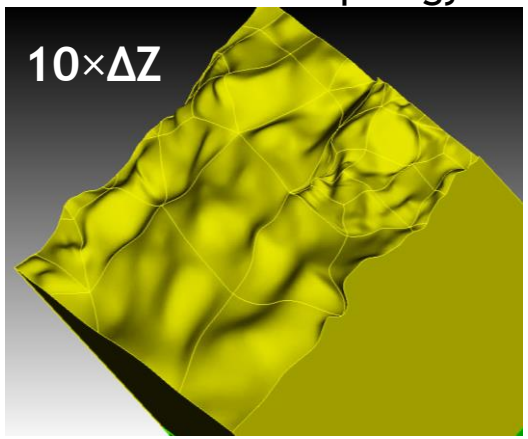
Simulate emission in PIC-code



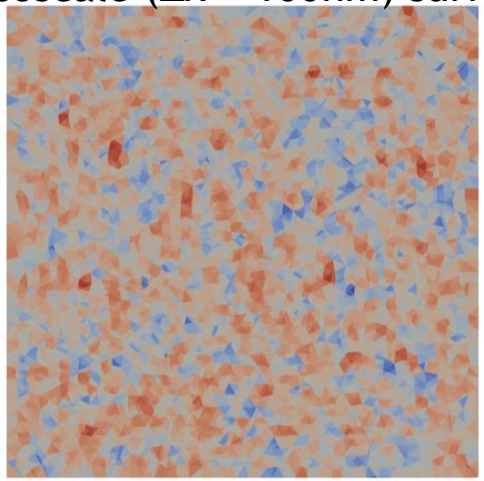
Mesoscale Field Emission Simulations

- Meso-scale model does show stochastic variation in the e- density just above the surface based on the random seed
- Goal is to be able to sample many possible surfaces (e.g., different β's and ϕ's) and compute breakdown probabilities for as-built surfaces
- Contours of electron density just above the cathode show very different spatial variation between the meshed STM surface and the flat, meso-scale surfaces
  - The STM surface was sputtered deposited Pt → large, ~micron-scale features are apparent
  - The current model picks atomic-scale emitter properties (β's and ϕ's) independently for every "meso-scale" surface elements. Clearly not independent for sputtered deposited Pt.

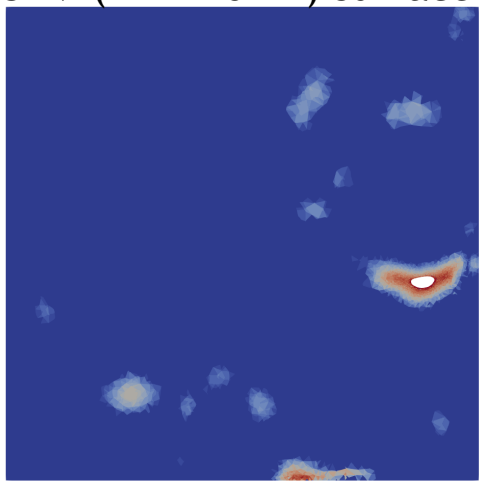
STM surface topology

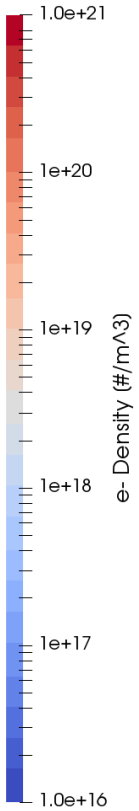


Mesoscale (Δx = 100nm) surface



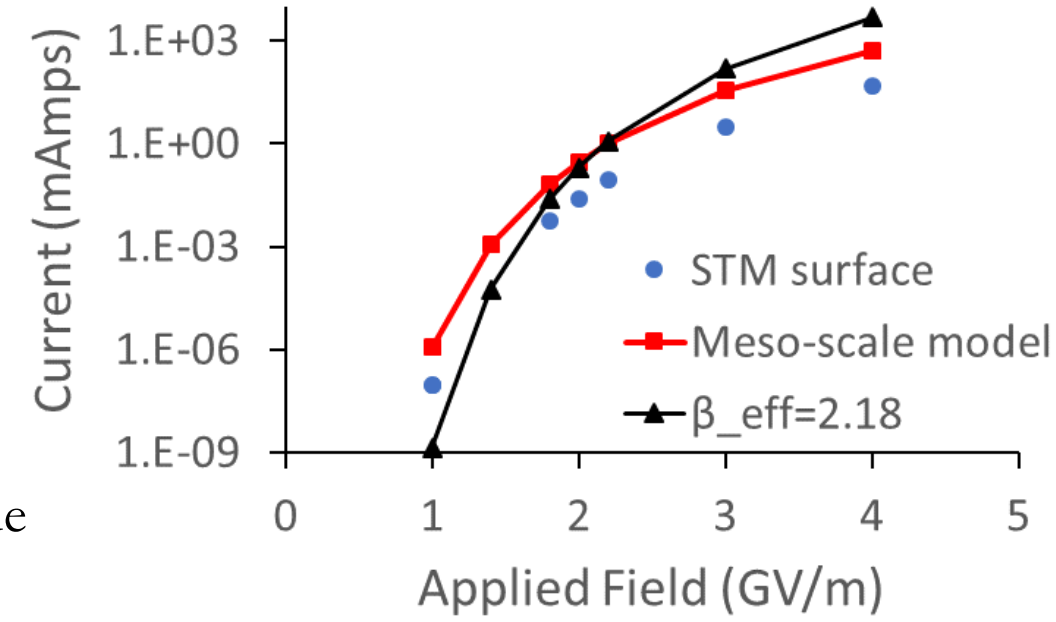
STM (Δx < 10nm) surface

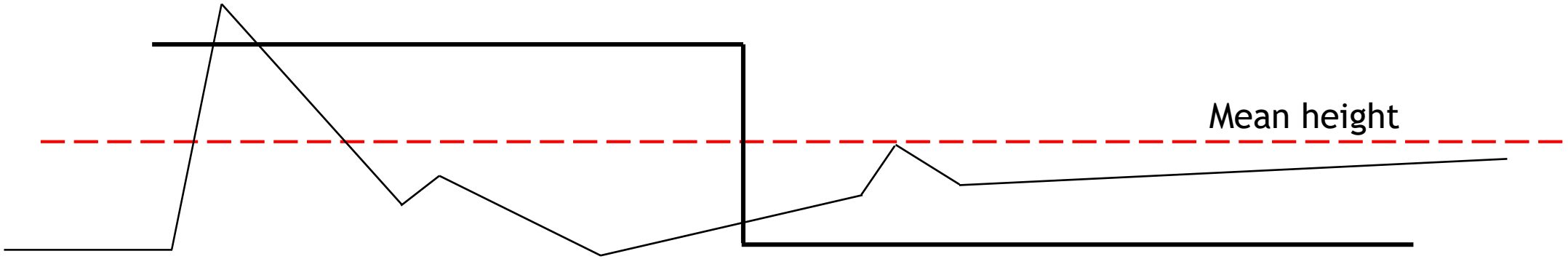




Mesoscale Field Emission Simulations

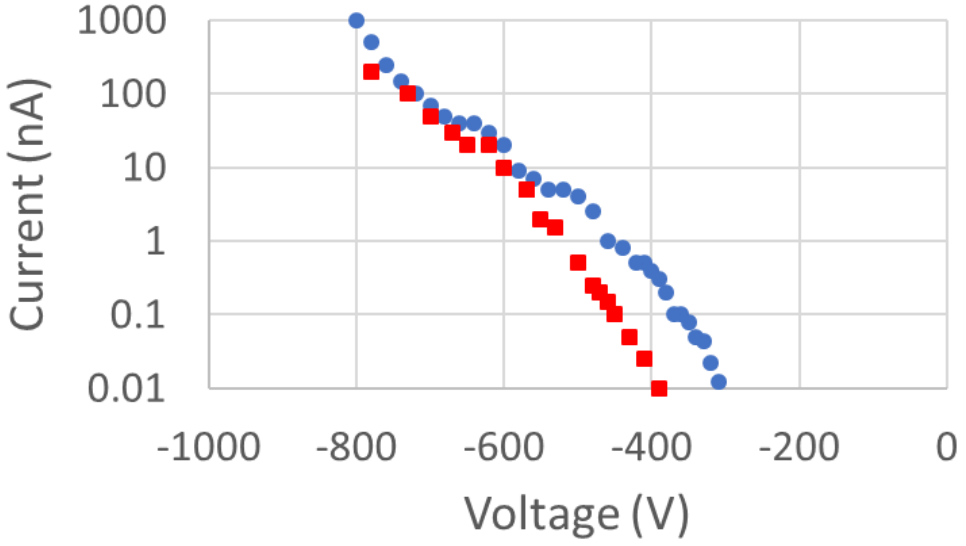
- Compare computed global current versus applied field for the resolved STM surface and meso-scale model surface
  - Stochastic variation in the mesoscale currents small
- The mesoscale model currents have the same trend as the STM surface, but ~12×i_{STM}
  - Difference partially (mostly?) from variation in fields due to changes in gap distance for the STM surface
  - Flat anode placed 10.4μm from the mean STM cathode height





Initial Local STM Breakdown Results

- Took local field emission i-V curves with tip radius < 100 nm at a distance of ~200 nm
- Relatively feature-less surface with small-β within the region of the tip field footprint
- Breakdown at ~4 GV/m!



- This seems to be evidence that, at least for relatively smooth sputter deposited Pt, we do not have small-β atomic-scale features that grow into large-β features which then allow breakdown to occur at ~10 MV/m.
- Perhaps there is a special feature somewhere on a ~1 cm^2 electrode that results in (or can grow to) a large enough β to get breakdown at ~10 MV/m that was not present on our ~10^{-6} cm^2 sampled area.

Conclusions

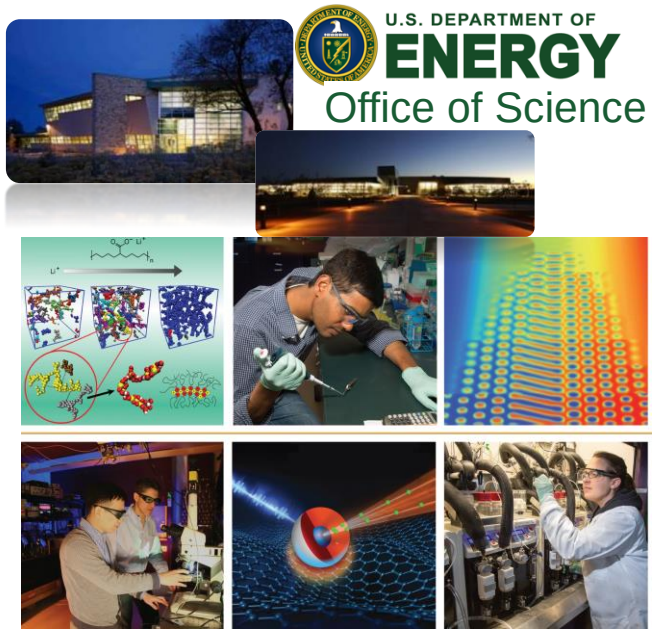
- Investigating surfaces at the atomic scale to characterize features relevant to vacuum field emission.
  - Surfaces that we characterized are extremely flat: $\beta \sim 1$ over 100's of μm^2
  - Want to clarify β-based field emission so β really is only geometry induced field enhancement.
- By examining field emission at the nanoscale, we have attempted to create a mesoscale physics-based model suitable for predictive (and stochastic) PIC simulation of emission
  - Still have a long way to go – working on how to handle the correlation between beta and work function.
- Characterized region, then performed local discharge in STM (spatially constrained surface participation) → Breakdown occurred at ~4 GV/m!
  - Region was flat and uninteresting – the breakdown field is consistent with breakdown from region with a small β

We have a Low Temperature Plasma Research Facility to collaborate with external partners. Please see www.sandia.gov/prf.

Sandia National Laboratories

Center for Integrated Nanotechnologies

U.S. DEPARTMENT OF ENERGY Office of Science



http://cint.lanl.gov