Exploring Dependencies of Work Function on Topography and the Application to Micron-scale Field Emission Model for PIC-DSMC Simulations

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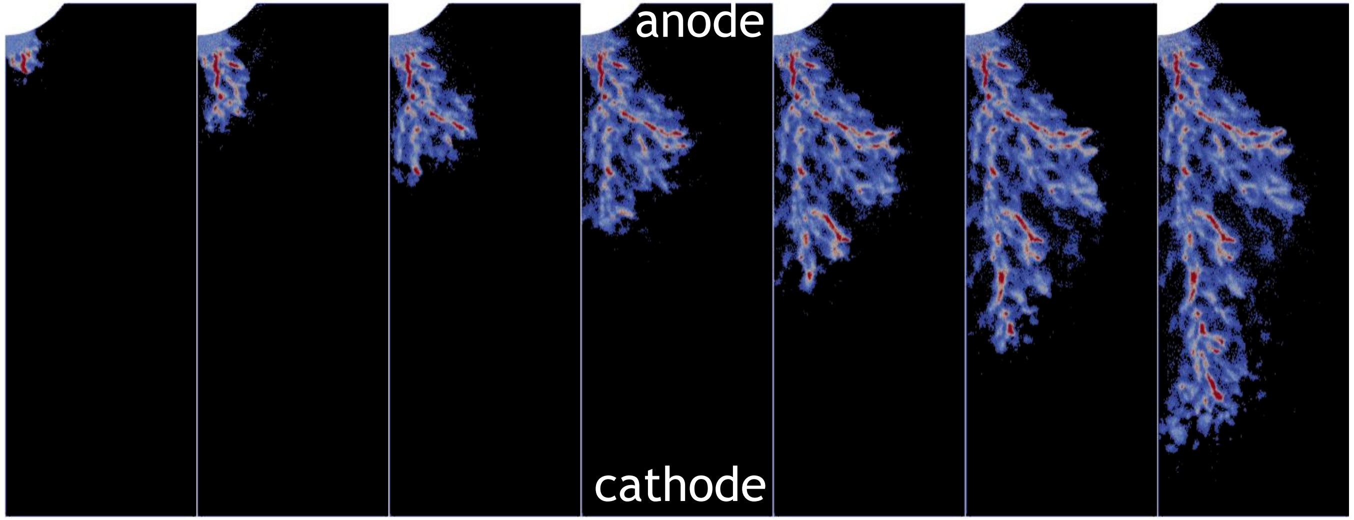
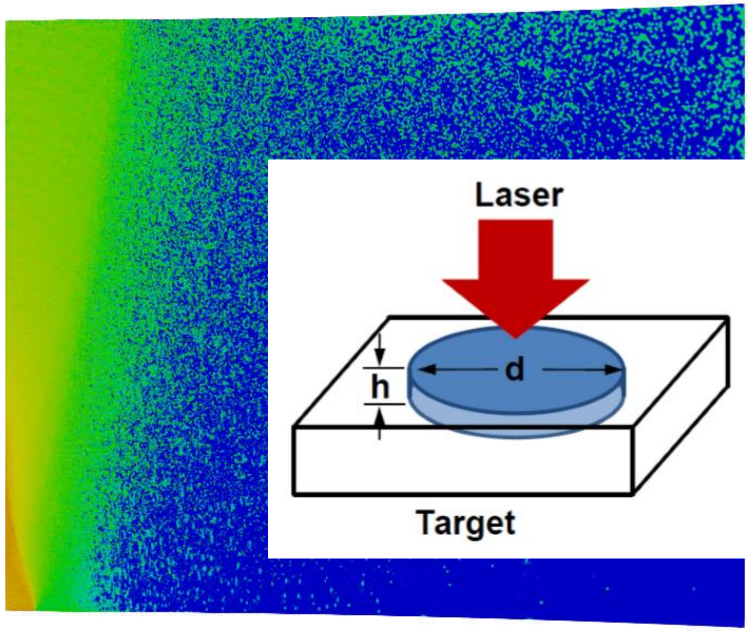
Introduction/Motivation

- We are interested in modeling a variety of discharge situations: from streamers at atmospheric pressure to vacuum arcs
- We have had multiple projects that focused on how plasma-surface interactions drive discharge

3D Streamer evolution (A. Jindal, SAND report 2022):

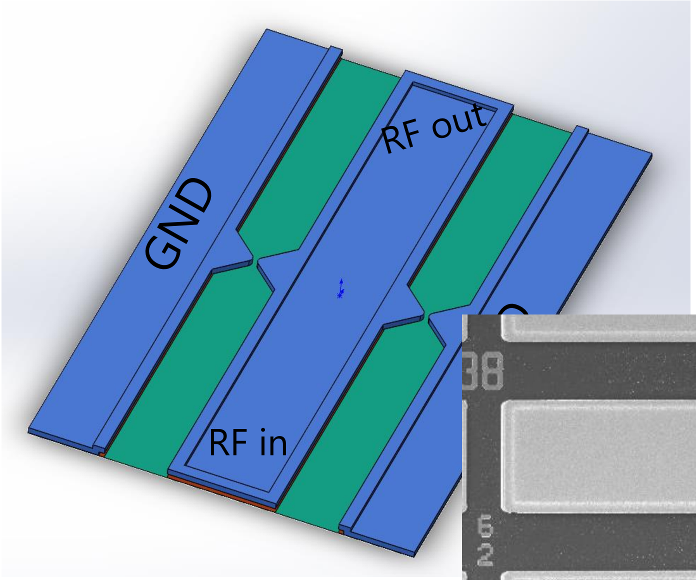


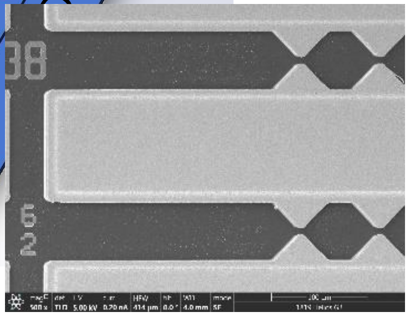
Laser-triggered switch
(A. Fierro, MeVArc 2018):

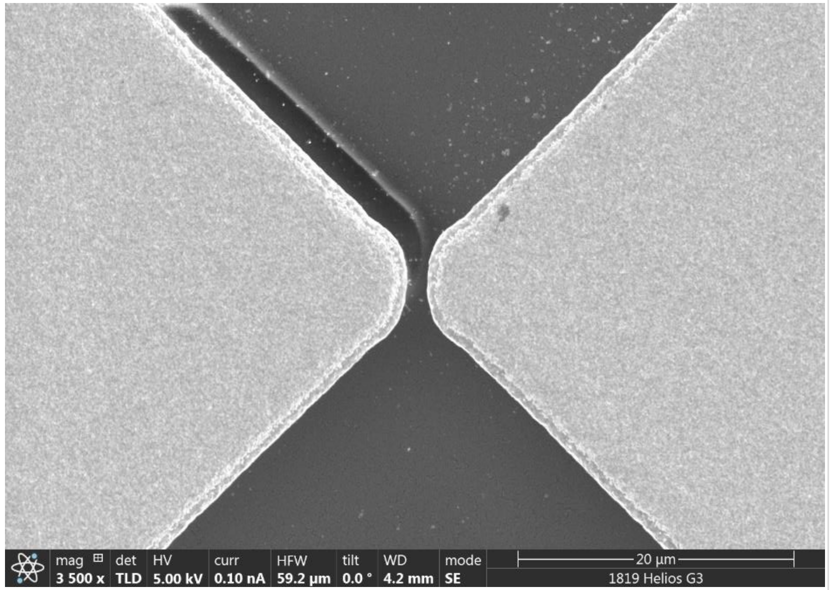


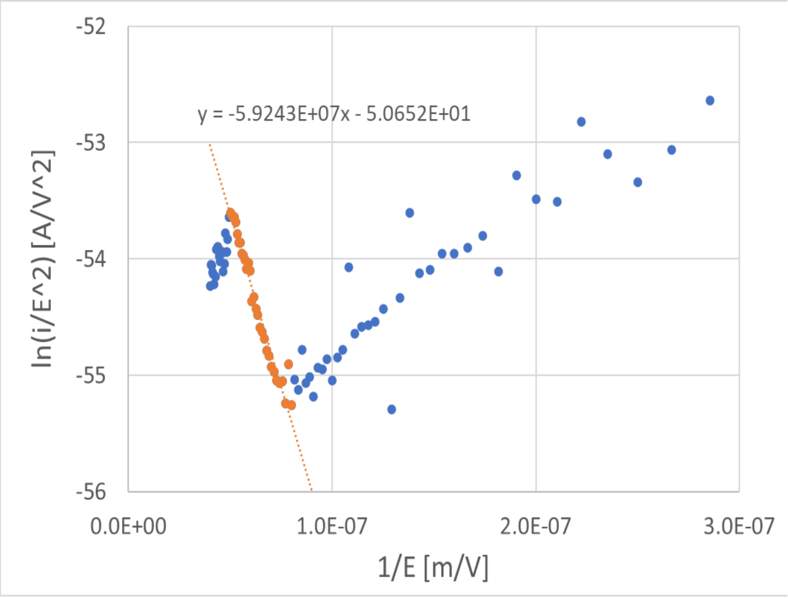
Introduction/Motivation

- We desire predictive PIC-DSMC breakdown simulations, for instance, EMPIRE simulations to predict RF shunt performance:
  - RF micro-discharge project with a goal to shunt signal at very low voltages
  - Fit a β~20 based on fit to data on FN-plots (beyond the enhancement from local macro-curvature)
  - Characterize the as-built electrodes – no sharp features stand out at ~100nm resolutions, so why β~20? Features that are <100nm or that grow as fields are applied?



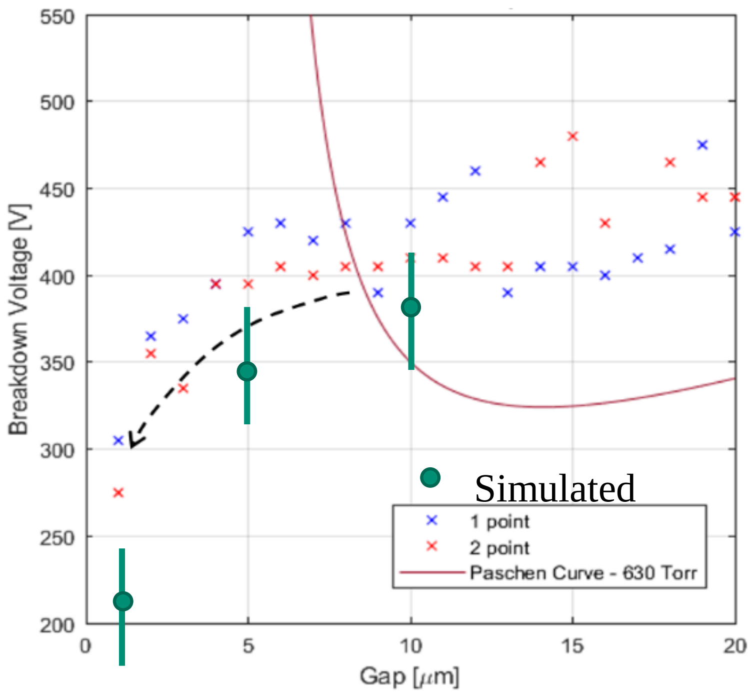




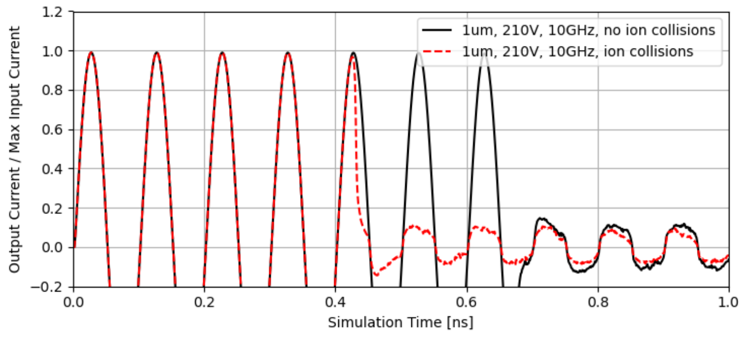


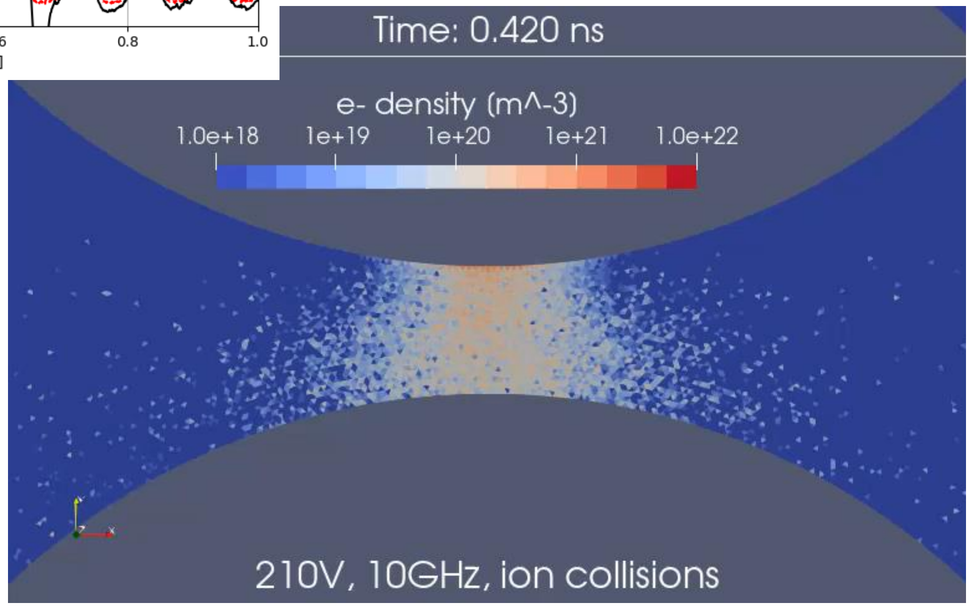
Introduction/Motivation

- Predictive PIC-DSMC breakdown simulations require confidence in the model via rigorous Verification and Validation efforts before a model is considered useful
  - Here predictive means capturing the bounds of discharge behavior due to stochastic variation of as-built real surfaces (variation of contaminants, grain boundaries, dislocations, etc.)



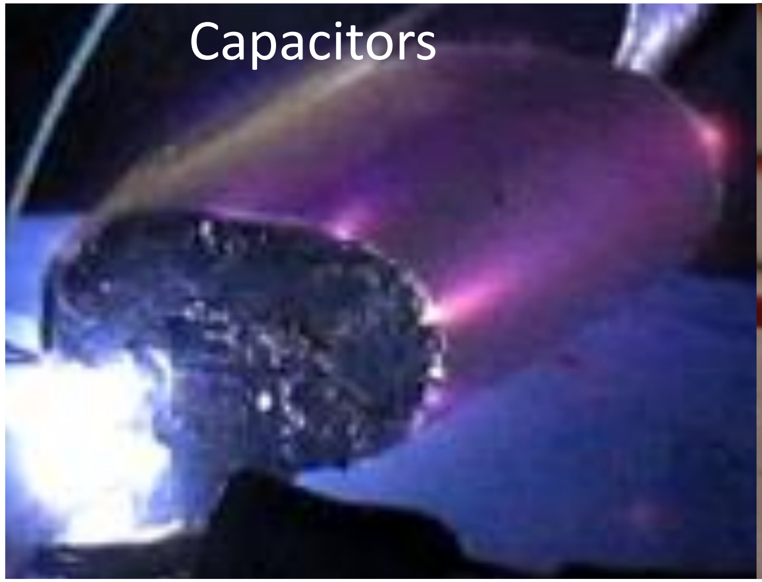
DC breakdown voltages. Field emission is seen to drive discharge for devices < ~5μm in the simulations

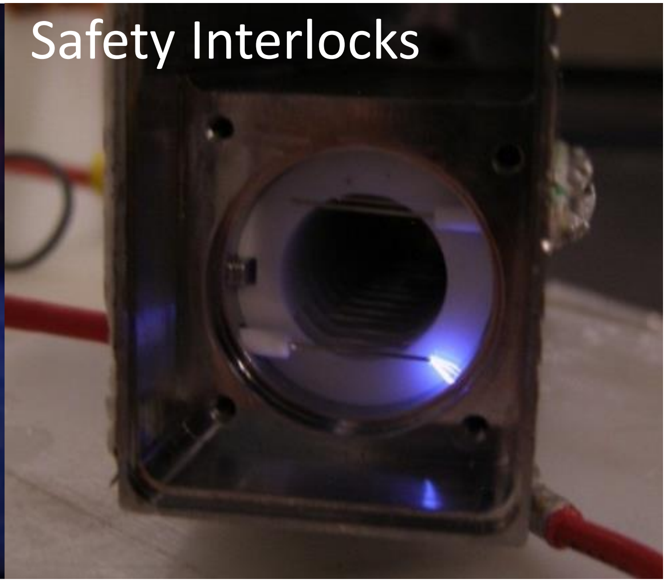




Vacuum Arc Initiation Project

- Vacuum discharge is critical to many modern devices.
  - Critical failure mechanism → Want to avoid
  - Mode of operation → Want to have predictable behavior



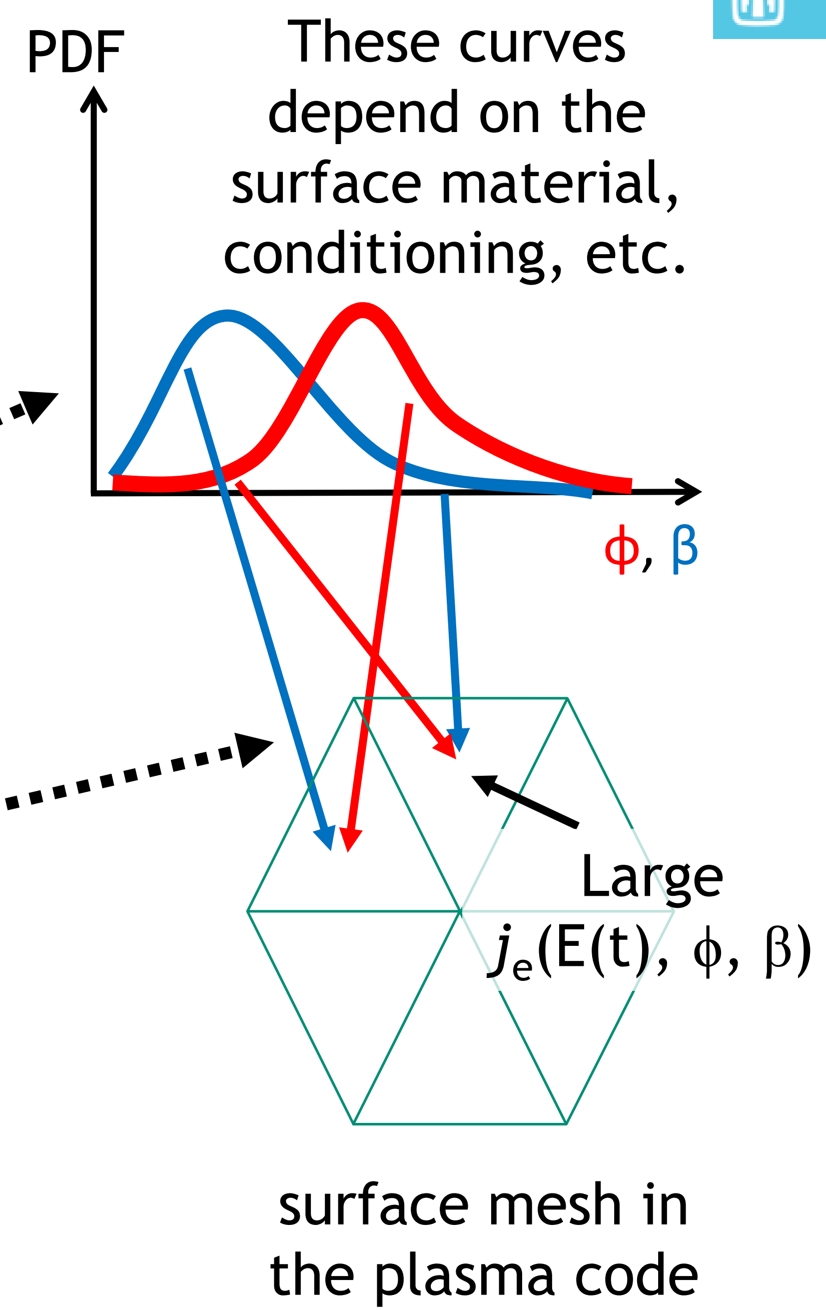


Vacuum Arc Initiation Project

- This project attempted to understand vacuum field emission from well-characterized surfaces to create physics-based models that **bridge ~nm scale features to ~μm-scales** for use in large-scale PIC-DSMC (EMPIRE) breakdown simulations
  - Field emission is necessary precursor to a breakdown event. No field emission → no breakdown.
  - Employ Scanning Tunneling Microscopy and PhotoEmission Electron Microscopy to characterize surface properties (e.g. β and ϕ) 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.
  - Utilize a "meso-scale" (0.1-1.0 μm) model of the surface for PIC-DSMC simulation of breakdown utilizing the plasma code EMPIRE

Overview

- 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

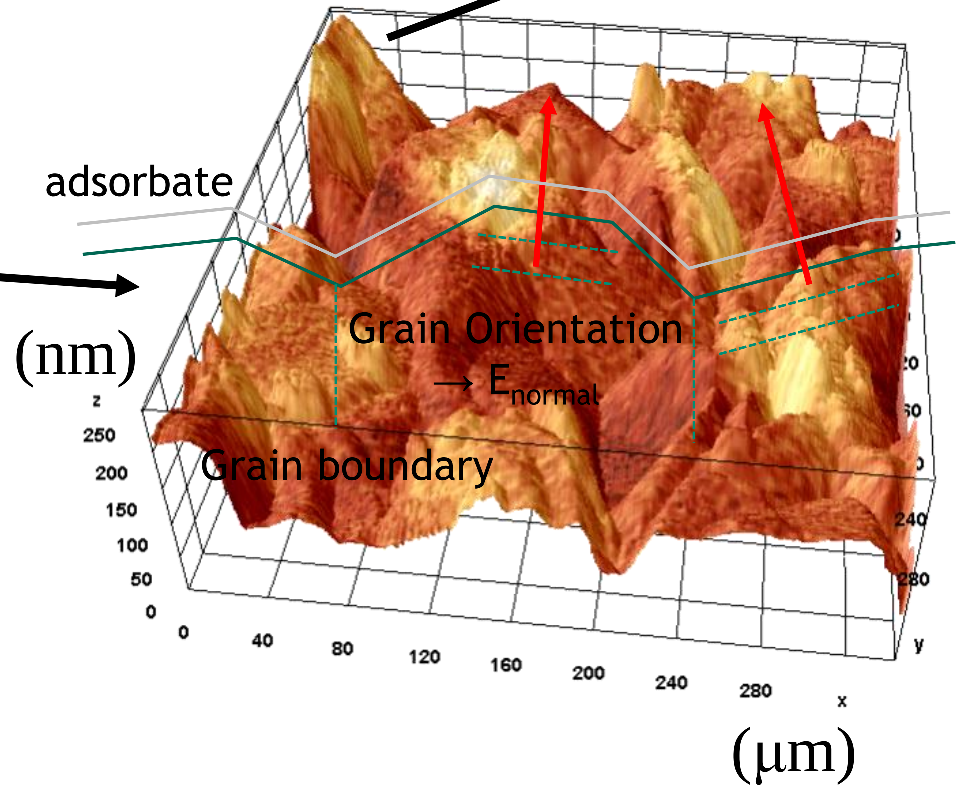


Why local characterization?

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

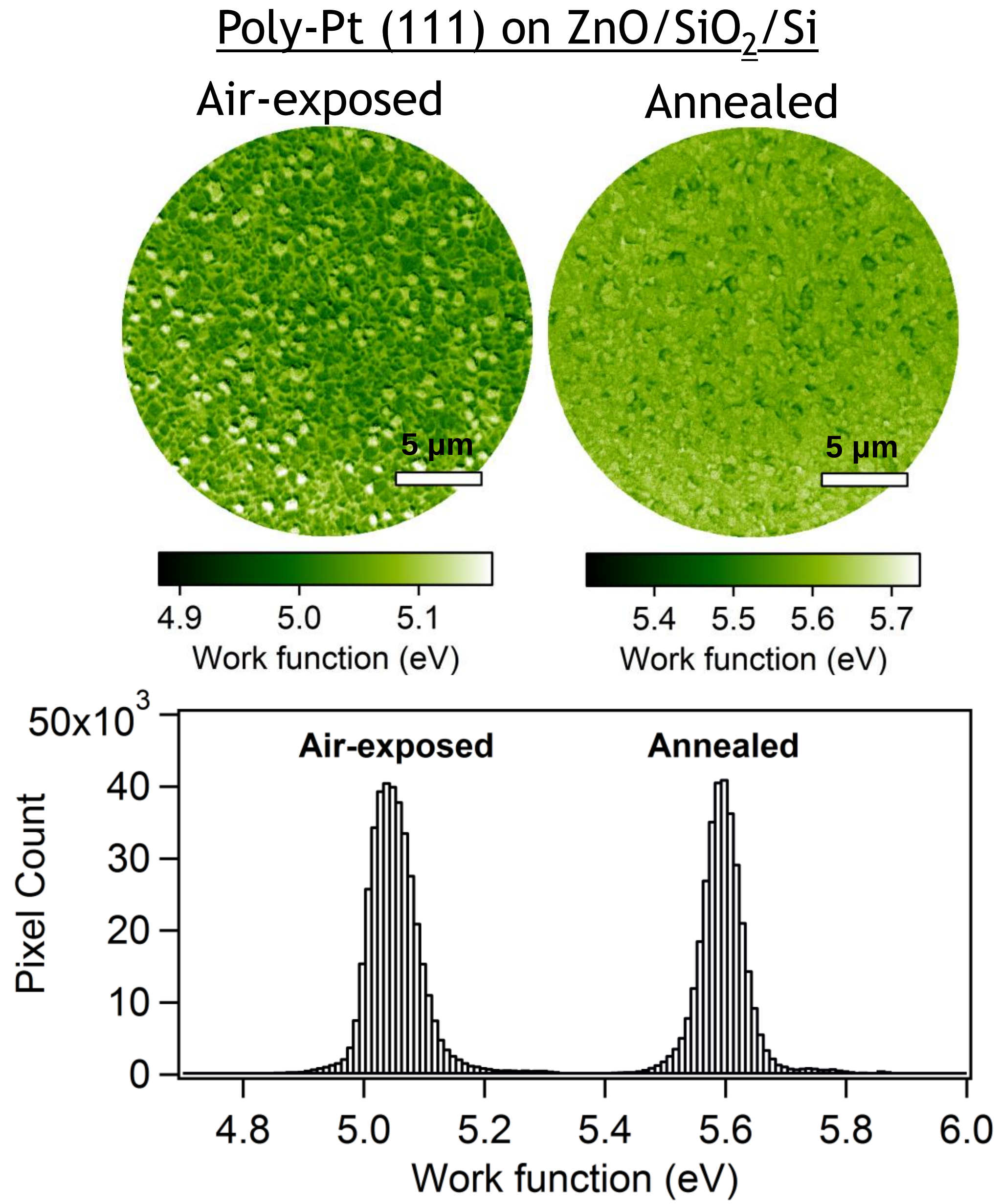
$$i = A_{eff} A_{FN} \frac{(\beta E)^2}{\phi t^2(y)} exp\left[-\frac{B_{FN} v(y) \phi^{3/2}}{\beta E}\right]$$

- We want to locally characterize the surface to eliminate β as a fit parameter
  - Use Scanning Tunneling Microscopy (STM) and Atomic Force Microscopy (AFM) to measure topology (β) at P ~ 10^{-7} mbar
  - Use PhotoEmission Electron Microscopy (PEEM) to measure work function (ϕ) at P ~ 10^{-9} mbar
  - Ultimately want to use measured distributions for ϕ and β to inform macro-scale model for discharge simulations



PEEM Measurement of Work Function Variation

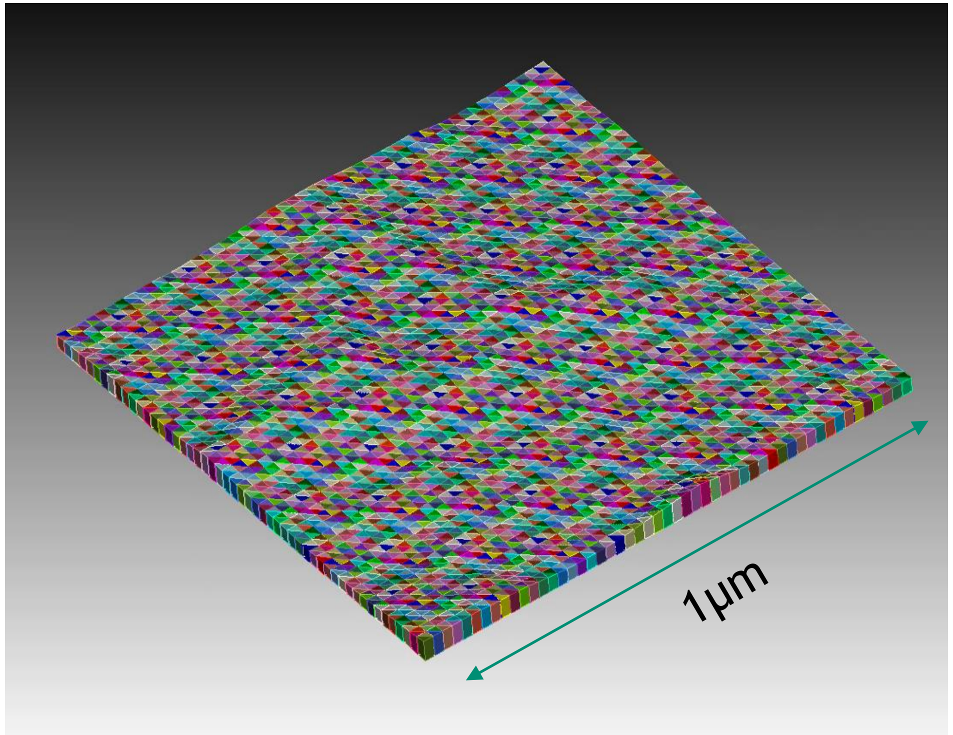
- Measured spatial variation of local work function using PhotoEmission Electron Microscopy (at P ~ 10^{-9} mbar)
  - Variation across given Pt surface relatively small – only a few percent
  - However, ϕ is in the exponential and the tail of the distribution can initiate field emission and eventually breakdown
- Significant (~10%) decrease in the work function due to surface contaminants picked up via exposure to air
- Use the ~10nm-scale PDF's in meso-scale model to set element work functions in PIC-DSMC simulations



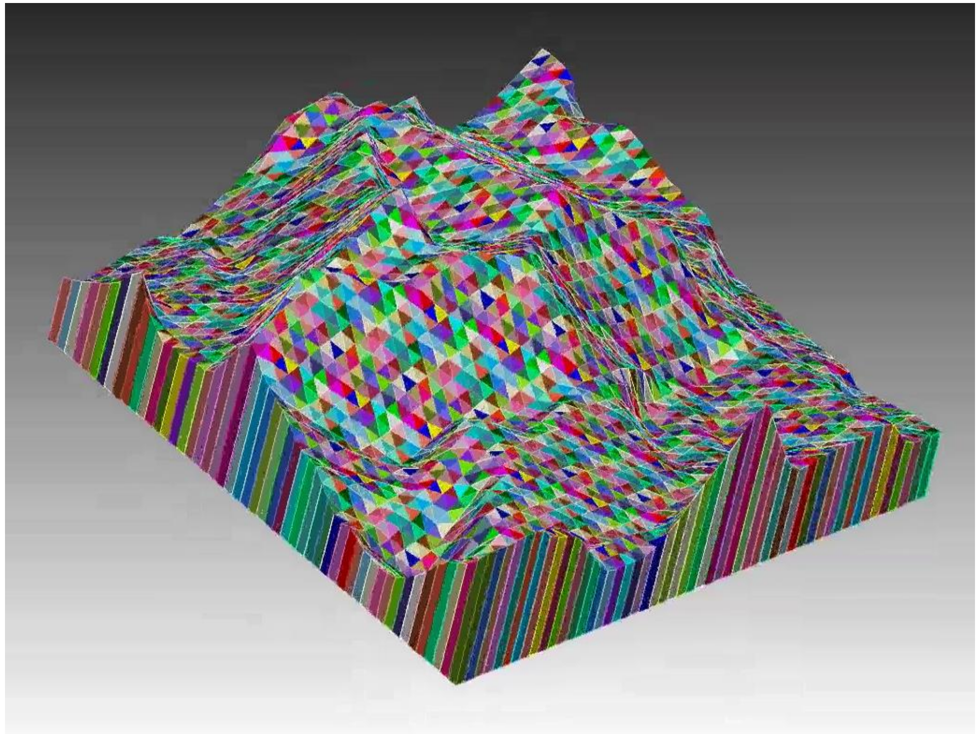
AFM Surface Characterization

- Took the AFM (x,y,z) spatial points (here ~20nm 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 (~ ±15nm)

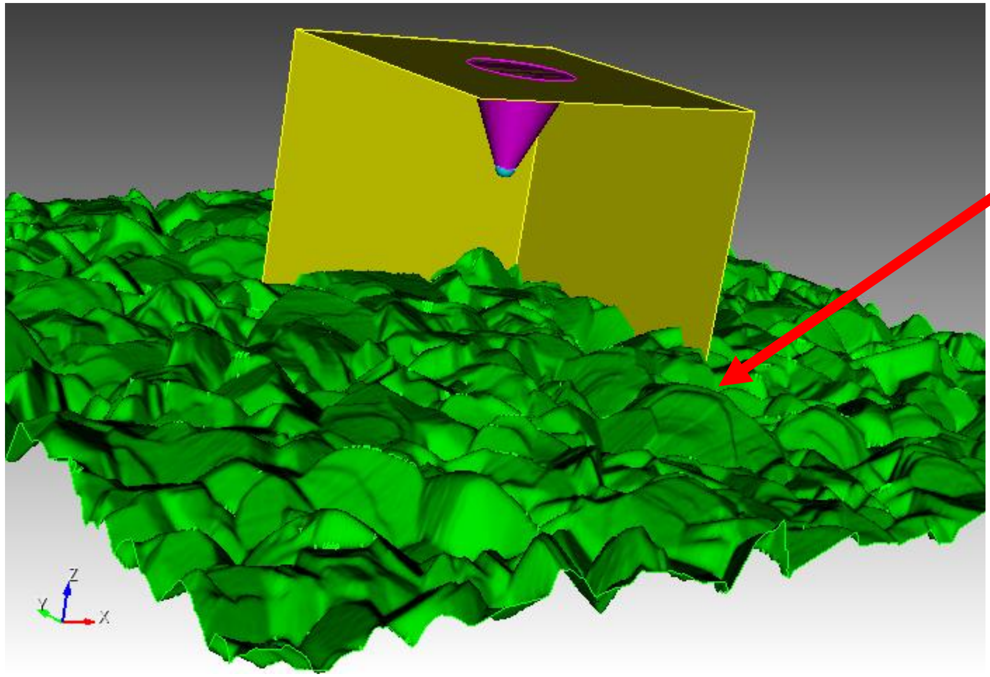


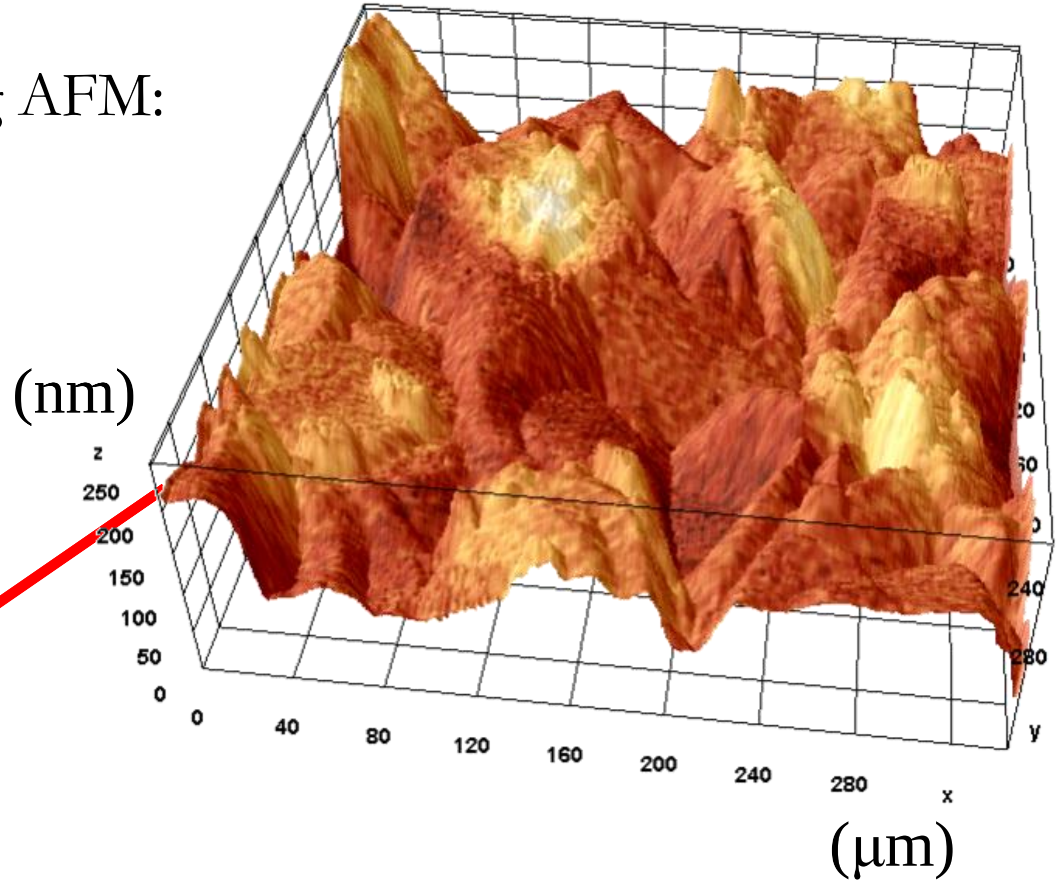
Surface relief increased by 10×



AFM topology → topological atomic-scale β

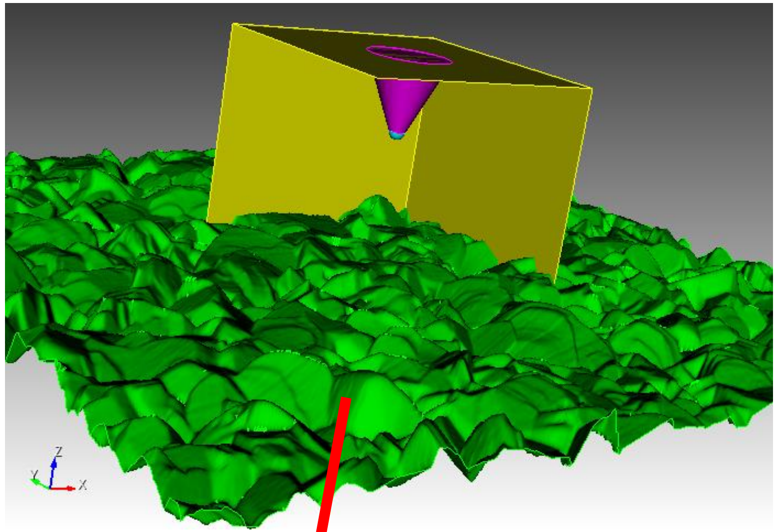
- Measure surface topology before breakdown using AFM:
- Load topology into Cubit and mesh the surface in order to use electrostatic solver
  - Place flat anode ~10μm from as-measured cathode
  - Use ~1 nm elements near cathode to resolve features



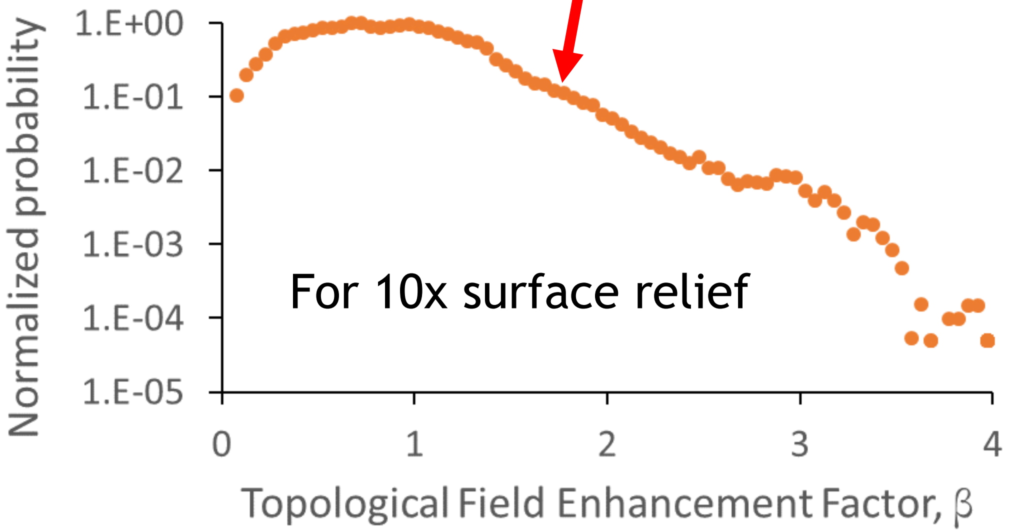


AFM topology → topological atomic-scale β

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



Electrostatic solve



Meso-scale Model for Surface Variations

- We have measured atomic-scale (1-10nm) PDF's of the work function and topological field enhancement factor

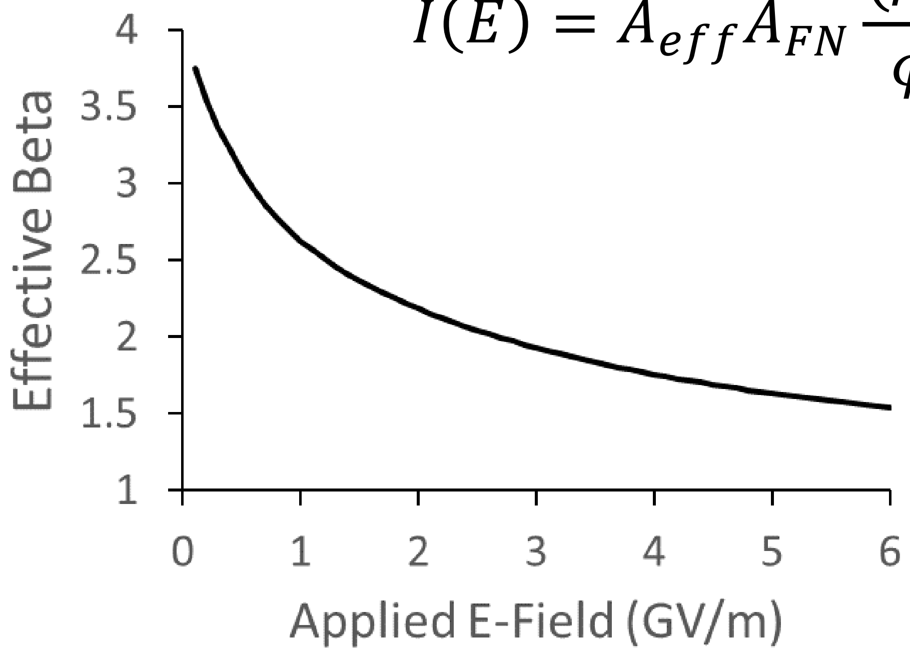
- Must convert these to the meso-scale (0.1-10 μm). Some options:
  1. Just pick the meso-scale β and ϕ from the atomic-scale PDFs
  2. Make an effective β and ϕ to use at the meso-scale
  3. "Brute force" – for each meso-scale element face, pick N local emitters (unique β's and ϕ's)

- The first option 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 meso-scale element's area
  - Other times there will be no tail values picked and no field emission until much higher fields

Meso-scale 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

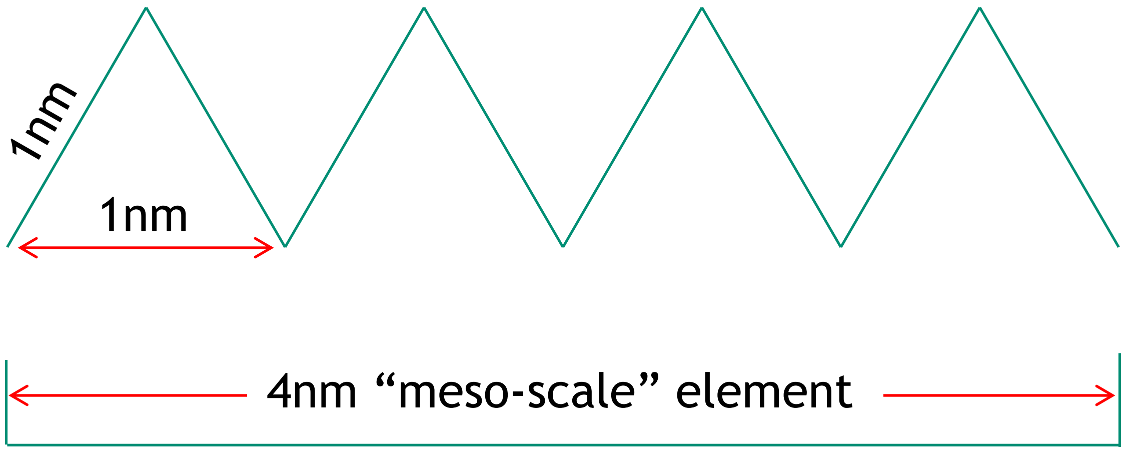
e.g. see: Feng and Verboncoeur, PoP **13**, 073105 (2006)
Jinpu Lin et al., J. Appl. Phys. **121**, 244301 (2017)

Meso-scale Model for Surface Variations

- We are left with "brute force" -- for each meso-scale 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

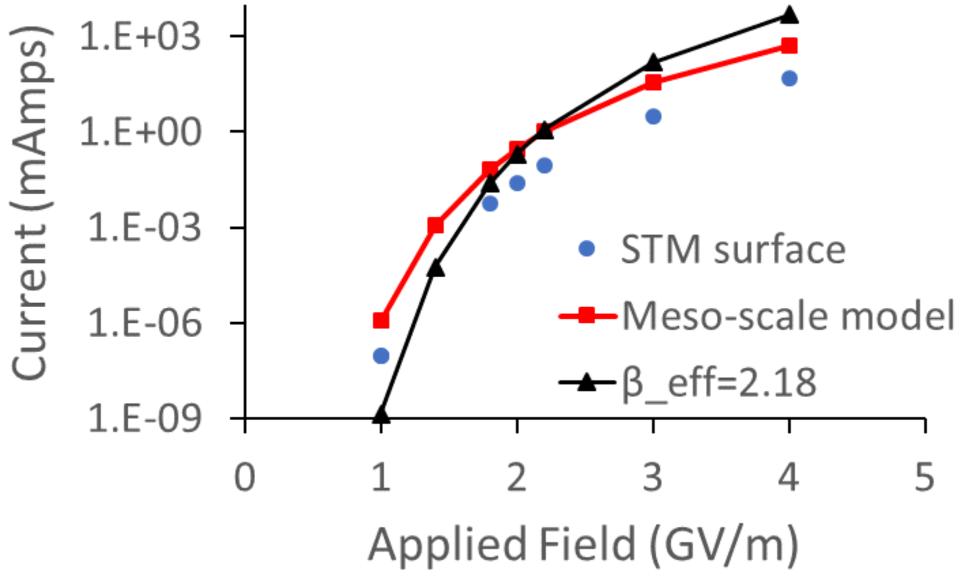
Meso-scale Model for Surface Variations

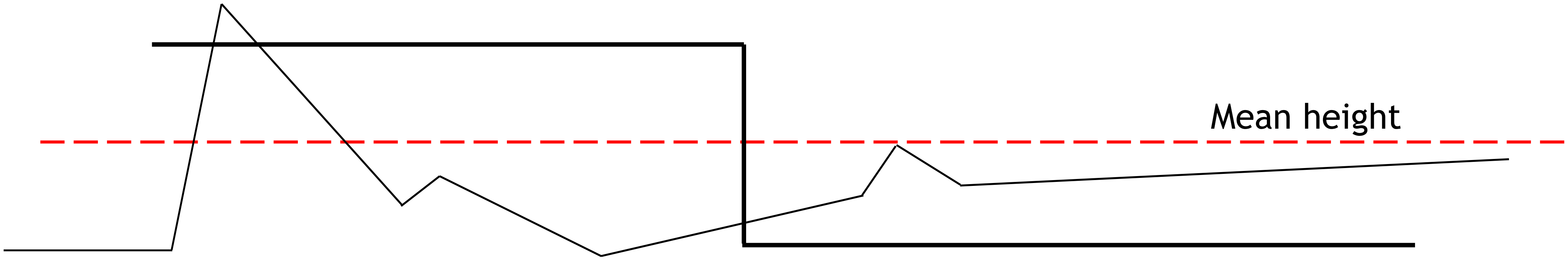
- However, we don't have to store all N local emitters for each surface element face
  - Field emission is highly non-linear and the majority of emitters (β and ϕ) can be neglected
- Store every atomic-scale emitter (β and ϕ) that appreciably contributes to the current
  - A threshold current contribution 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:

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

Meso-scale Field Emission Simulations

- Compare computed global current versus applied field for the resolved STM surface and meso-scale model surface
  - Flat anode placed 10.4µm from the mean STM cathode height
  - Stochastic variation in the meso-scale currents small
- The meso-scale model currents have the same trend as the STM surface, but ~12×i_{STM}.
  - Difference due to non-linear nature of FN current vs. E($V/d_{gap,local}$)
  - Gap distance varies due to local peaks and global tilt in the STM surface data





The Meso-scale Model is Great but…

- We assumed that β and ϕ were independent… however they appear to be correlated due to the local step density where each atomic step acts like a small dipole that can reduce the local work function

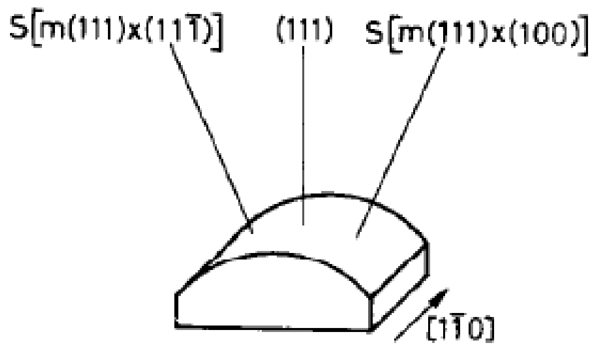
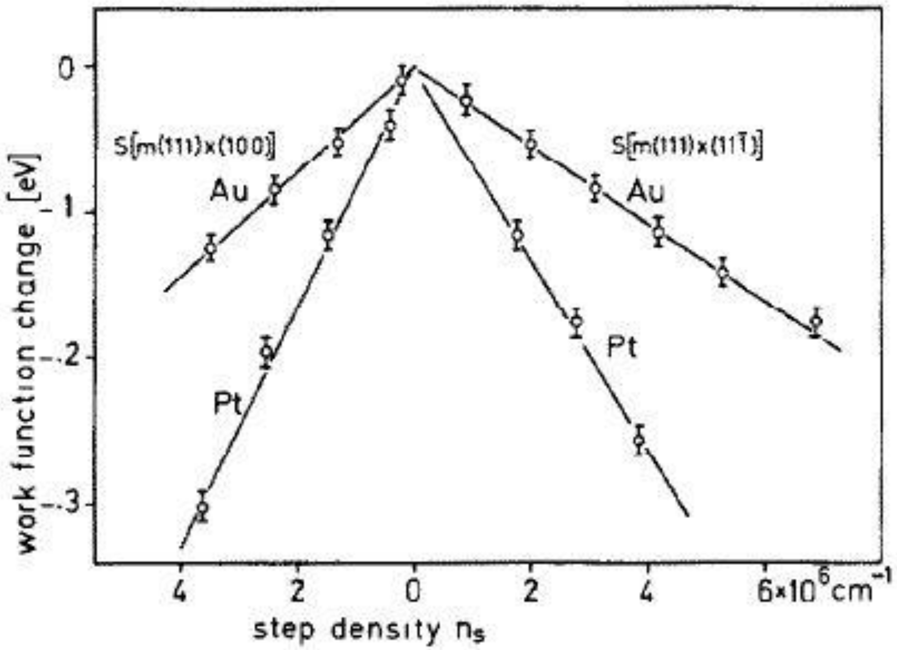


Fig. 1. Schematic representation of curved Pt and Au sample.



K. Besocke, B. Krahlurban, and H. Wagner, *Surf. Sci.* **68, 39 (1977).

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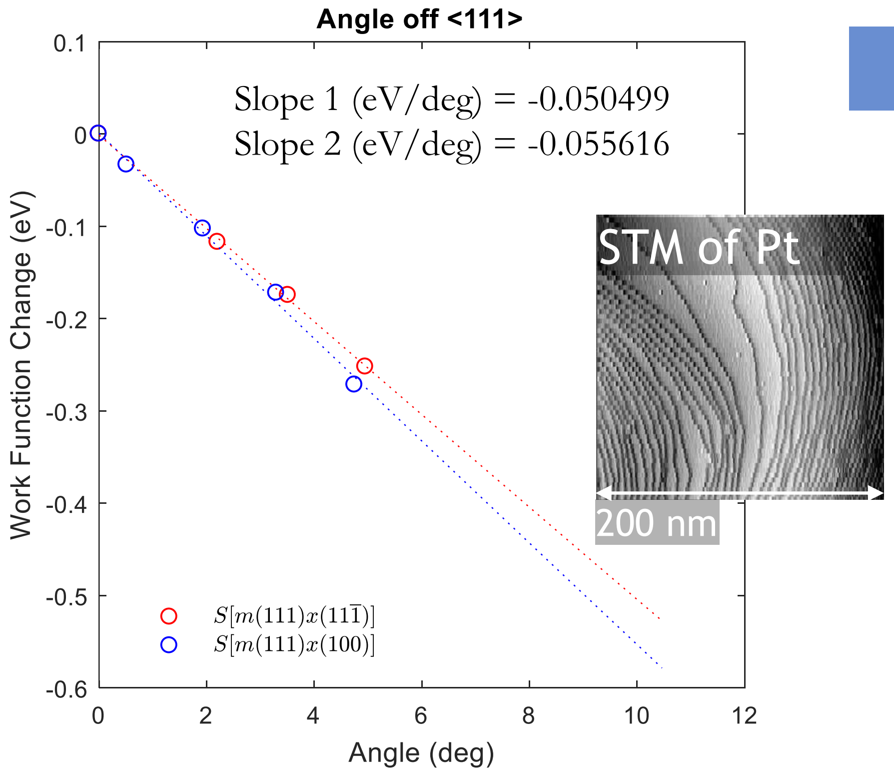
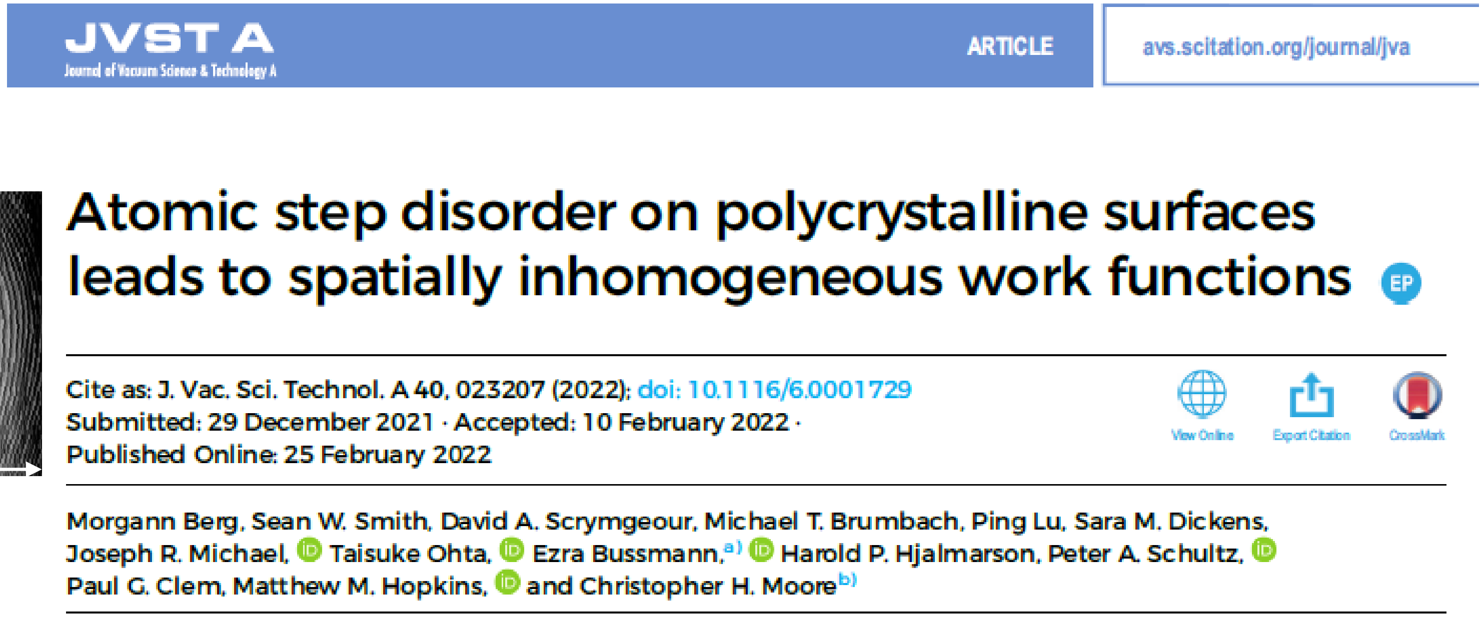


Our measurements on sputtered Pt



JVST A
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Atomic step disorder on polycrystalline surfaces leads to spatially inhomogeneous work functions

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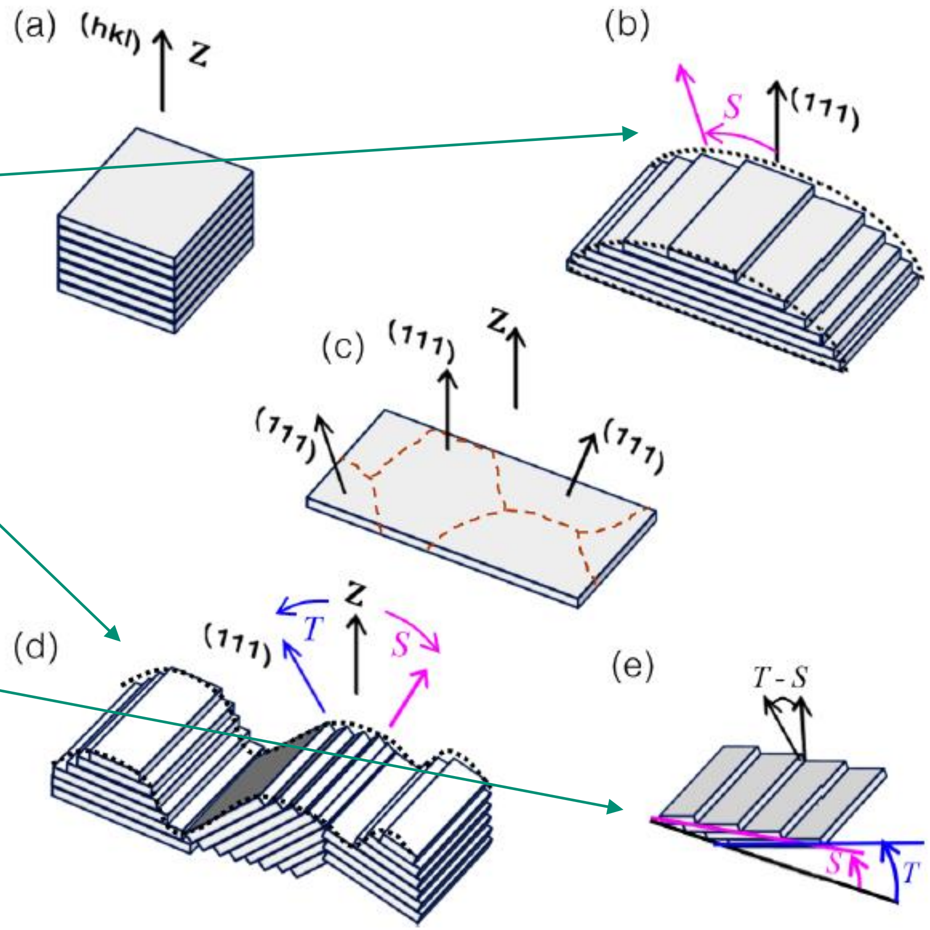
Correlating Local Work Function and Topography

- Here we show the relationship between atomic steps and the crystalline (111) surface.
- Atomic steps result in a local topological slope (S)
- The sputter deposited and annealed Pt grains in our study have small but non-zero grain-to-grain tilt angles (T) which are effectively random
- Tilt and slope both play a role in the dipole (apparent atomic step) density, N_s:

$$N_s = \frac{\sin(T - S)}{a_{111}}$$

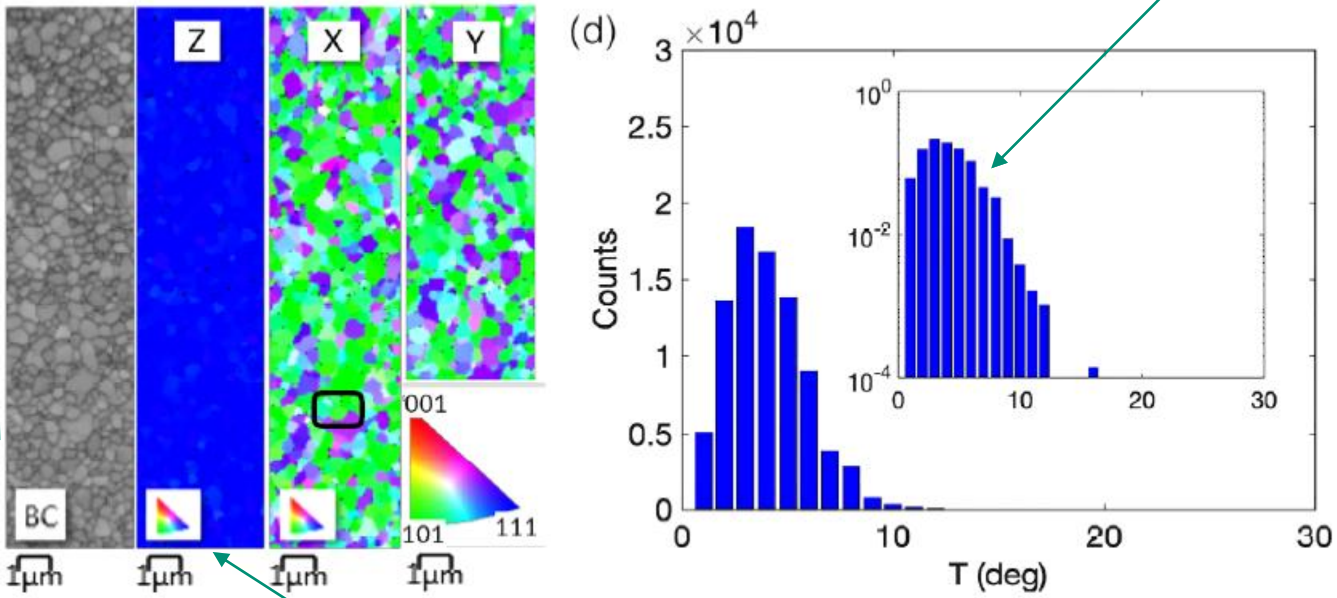


Crystallographic orientation

- Use Electron BackScatter Diffraction (EBSD) to measure ensemble distributions of the grain crystal orientations in the Pt sample.

EBSD local crystallographic orientations

Distribution of grain tilt angles (T) from the EBSD measurements



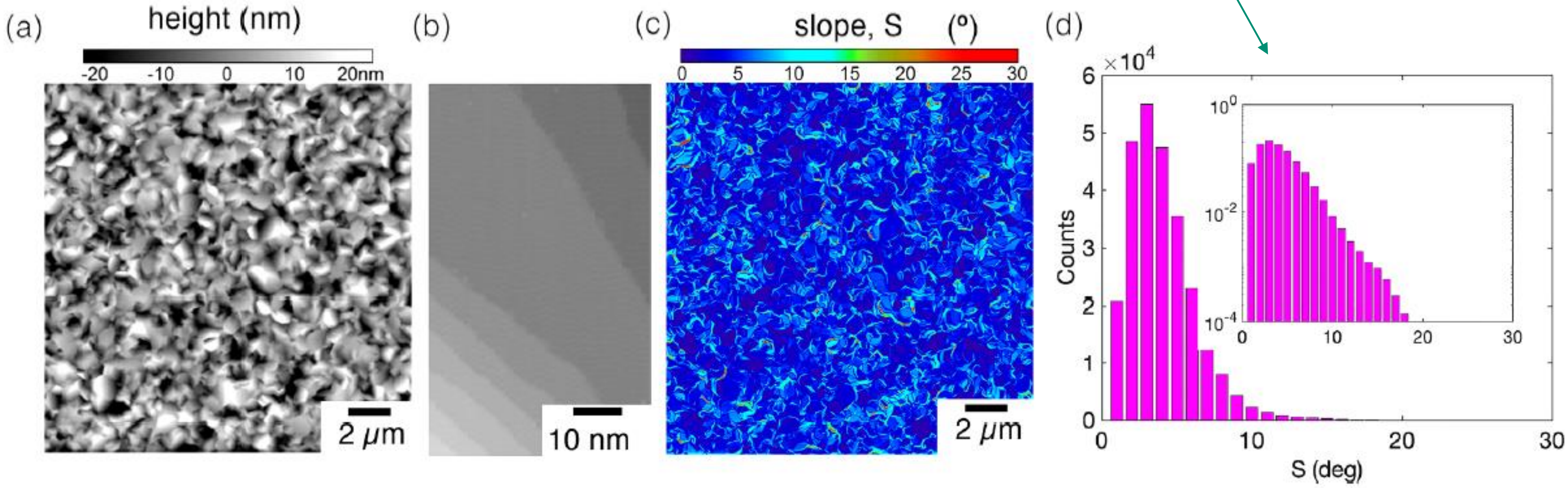
- We will use this distribution to draw local tilt angles for the AFM-resolved field emission simulations

SEM showing grains

Pt grains are predominately oriented with 111 aligned to the global surface normal, Z

Surface Topology and Local Slope

- Use AFM to measure the local height with ~10nm XY resolution
- From this mapping we can plot the local slope and generate a distribution of slopes

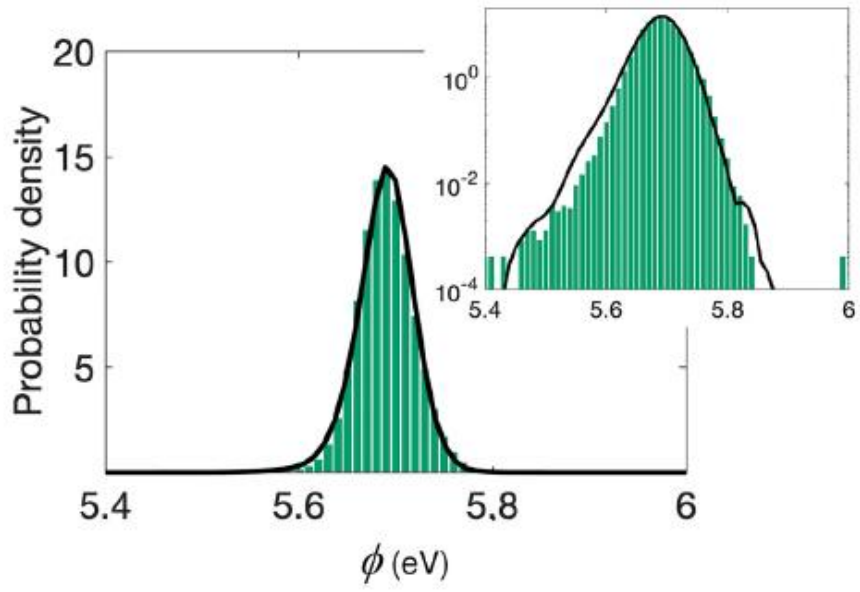


Predicted vs. Measured Local Work Function

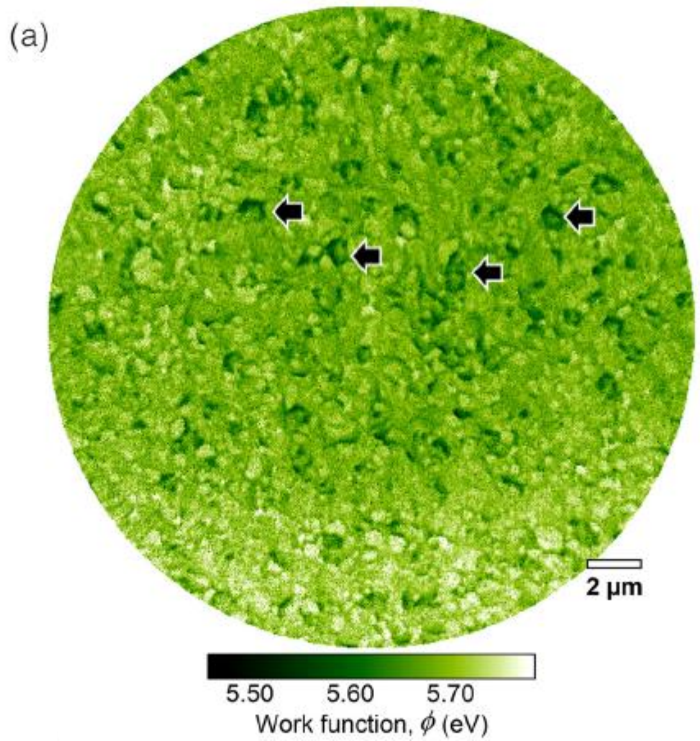
- Generate work function from atomic step density:

$$\phi = \phi_{111} - 300 \times 10^{-18} 4\pi\mu N_s$$

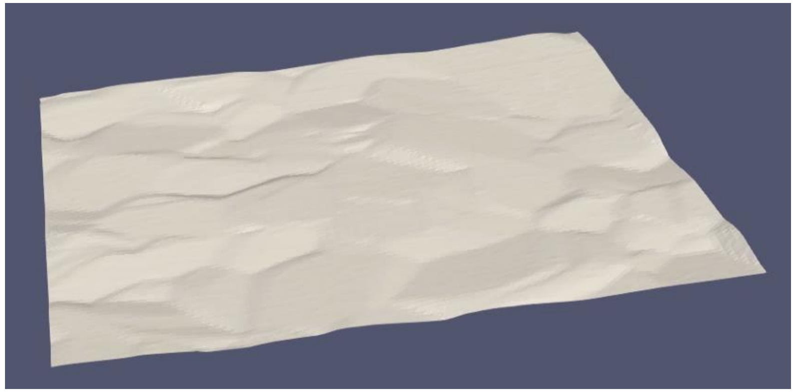
- Assume that tilt and slope are independent and randomly drawn from the two distributions
  - Best fit value to the exp. data: $\mu \approx 4\times10^6$ D/cm
- Predicted: Thick black line; Green bars from PEEM



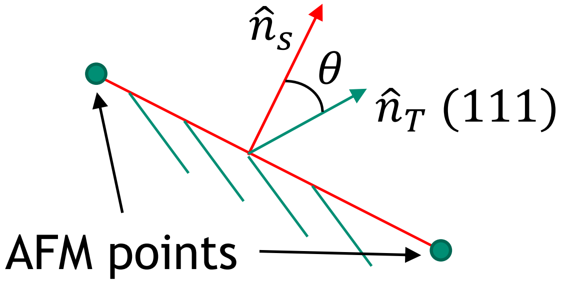
PEEM measurement



EMPIRE PIC-DSMC Simulations



- Mesh 2x2μm AFM-measured surface with tets
  - Use the face normal to get local surface slope
  - Randomly select (111) tilts from the measured distribution

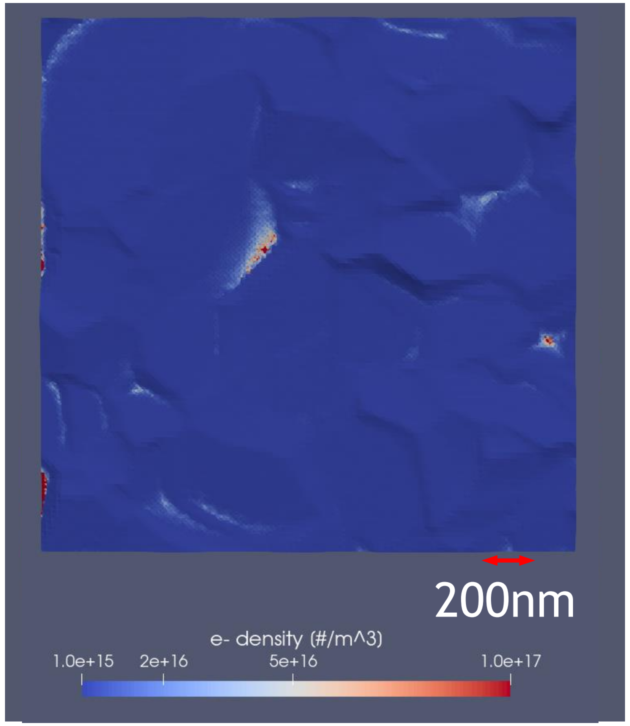


$$\cos\theta = \frac{\hat{n}_T \cdot \hat{n}_s}{|\hat{n}_T||\hat{n}_s|}$$

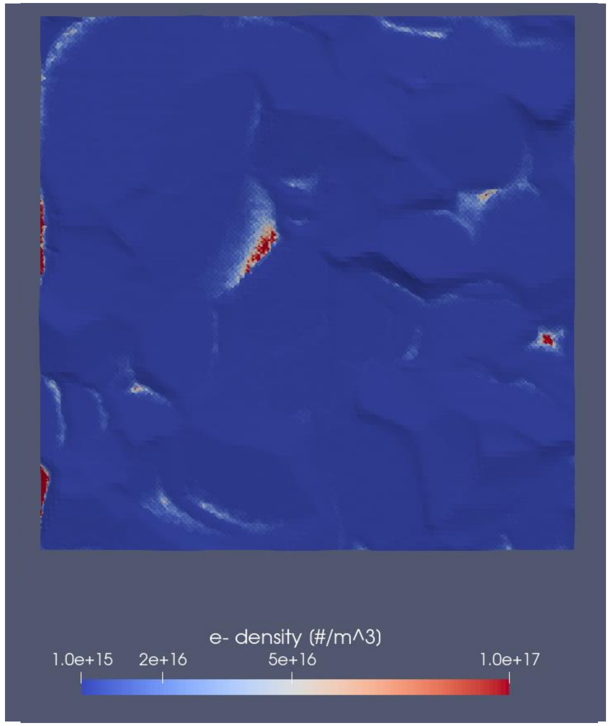
$$N_s = \frac{\sin\theta}{a_{111}}$$

- Simulated Fowler-Nordheim emission with 3GV/m field:

With constant ϕ



With $\phi(N_s)$



- Very little difference, but this was a flat surface with small step densities.
  - The local topology-based work function results in a total current that is ~3x the constant work function.

$$\frac{j_{FN,\phi(N_S)}}{j_{FN,\phi}} \approx e^{\left(\frac{\phi(N_S)}{\phi}\right)^{1.5}} \sim 2.65 \text{ for } \frac{\phi(N_S)}{\phi} = \frac{5.6}{5.7}$$

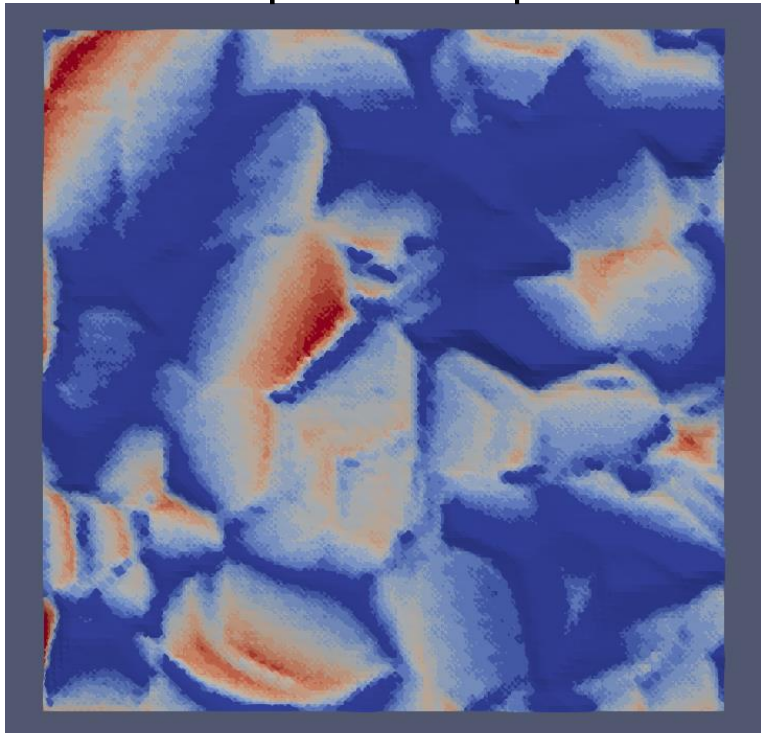
→ Outlier β's & ϕ's drive emission!

Field Emission Variation Due to Gap Size

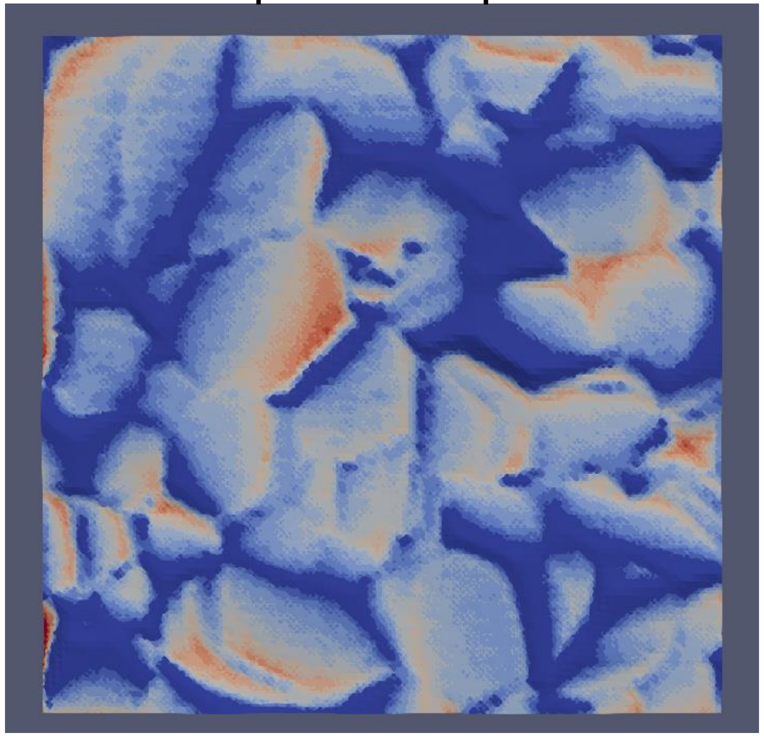
- Word of caution: If surface height variation is not negligible compared to the gap size, there will be substantial variation just due to local gap size changes.

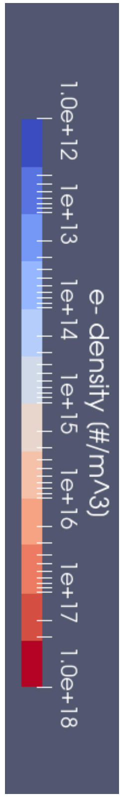
- Our surface had a maximum relief of ± ~0.1µm. Comparing a gap of 0.1µm (with 0.3kV at the anode) and a gap of 10µm (with 30kV anode) we find the smaller gap current is ~4.1x the larger gap current.

Gap size = 0.1µm



Gap size = 10µm





Conclusions

- Investigating surfaces at the atomic scale to characterize features relevant to vacuum field emission → want topology-based β and ϕ
  - Surfaces that we characterized are very flat: β~1 over 100's of μm^2
- By examining field emission at the nanoscale, we have attempted to create a spatially variable, meso-scale physics-based model suitable for predictive (and stochastic) PIC simulation of emission
- Initial model for β-ϕ correlations based on effective dipoles created by the local step density (from measured topology). Still much to do:
  - Look at whether the local crystal tilt is really independent of the local slope
  - How to properly choose model tilt for a grain-sized region
  - Extend this correlation demonstrated on resolved (nm-scale) mesh to the meso-scale model for real problems

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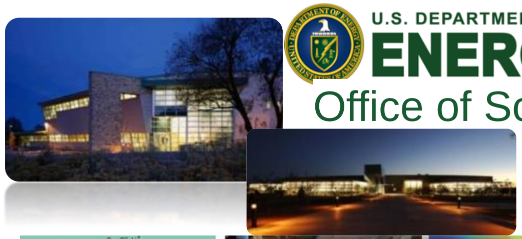

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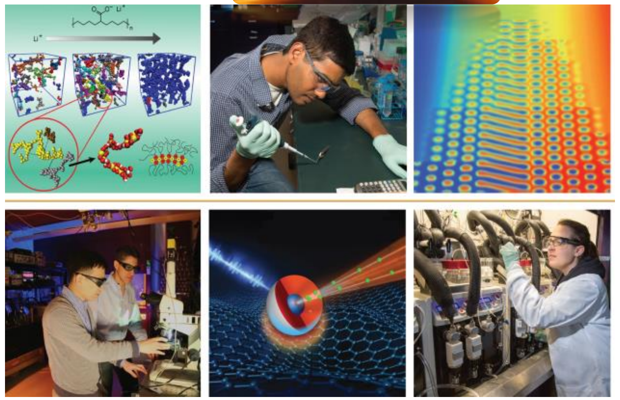

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