Influence of nanoscale surface modifications to the estimated field enhancement and emission currents

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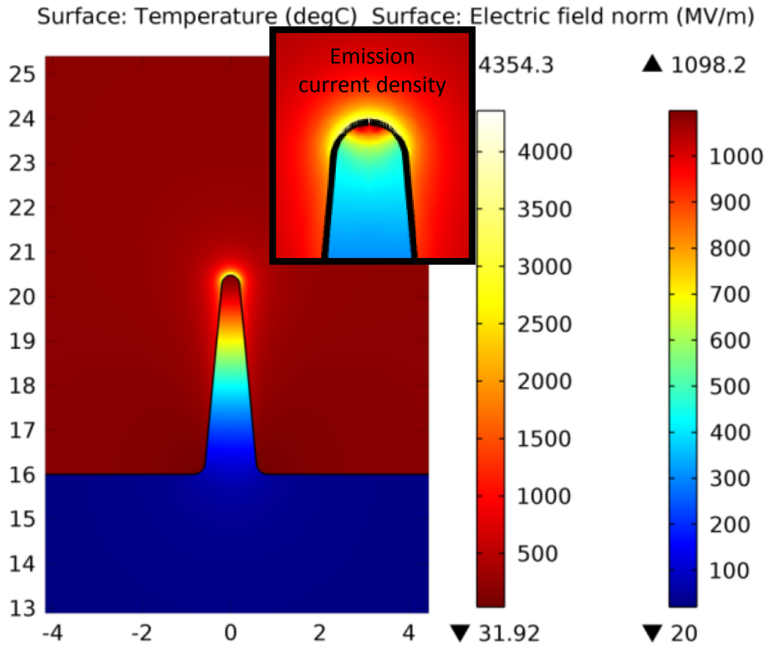
Hypotheses and Aims

- **Possible mechanism responsible of field enhancing surface modifications:**
  - material structure, fatigue and plastic deformations related causes
  - electric field assisted diffusion of surface atoms
  - reduction of work function due to the influence of oxide layers and contaminants on the surface
- **The field enhancement factor may not only be caused by high aspect ratio surface features**
- **Possible other mechanisms are**
  - dynamic, electric field or time dependent changes of surface features,
  - change of work function (due to the oxides)
- **The AIM: Fundamental understanding of processes initiating the field emitters**
- **Checking for the possibilities of surface modifications with least possible assumptions – DFT calculaitons**

Heating and emission currents

Local emission currents – connection to the experiment



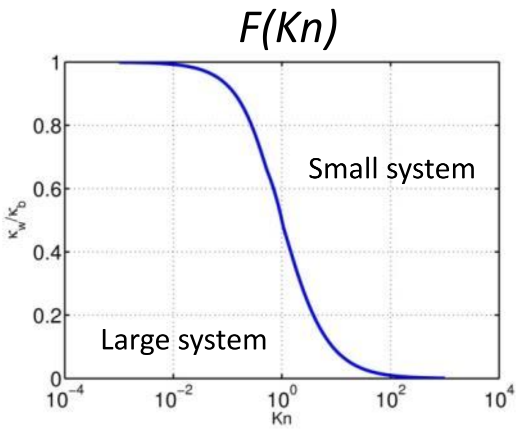
- Heat equation in steady state
- Fully coupled currents and temperature
- Emission currents concentrated to the top of the tip
- **Nottingham effect included in thermal modelling**

Field emitters as nanowires

$$\sigma_w = F(Kn) \cdot \sigma_b$$

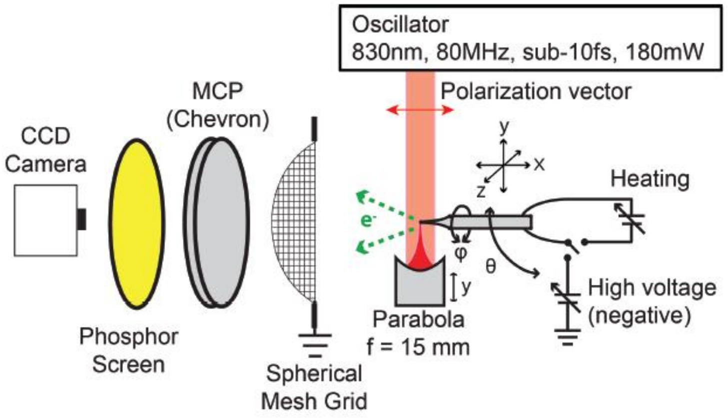
$$\kappa_w = F(Kn) \cdot \kappa_b$$

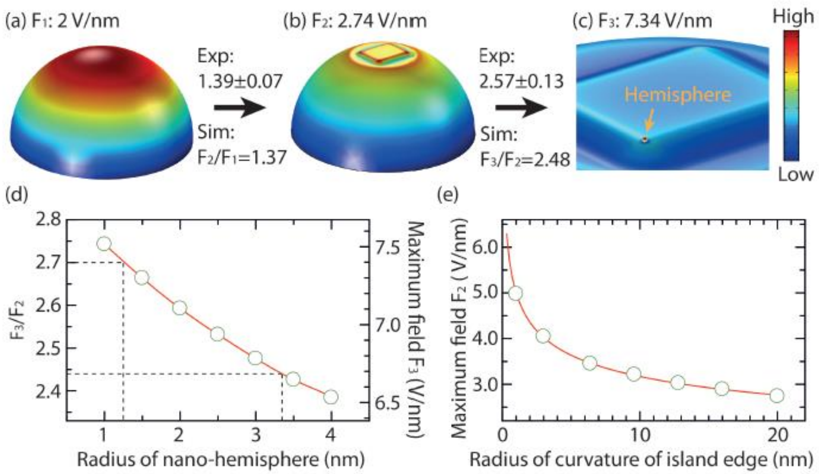
$$Kn = \frac{L_{free}}{d}$$



- Size dependence of electric and thermal conductivity
- Conductivity in nanoscale emitters is significantly decreased (more than 10x for sub-nanometer tip)
- Knudsen number to characterizes nanoscale size effects
- Wiedemann-Franz law for thermal conductivity
- **Optionally, temperature dependence in finite size effects**

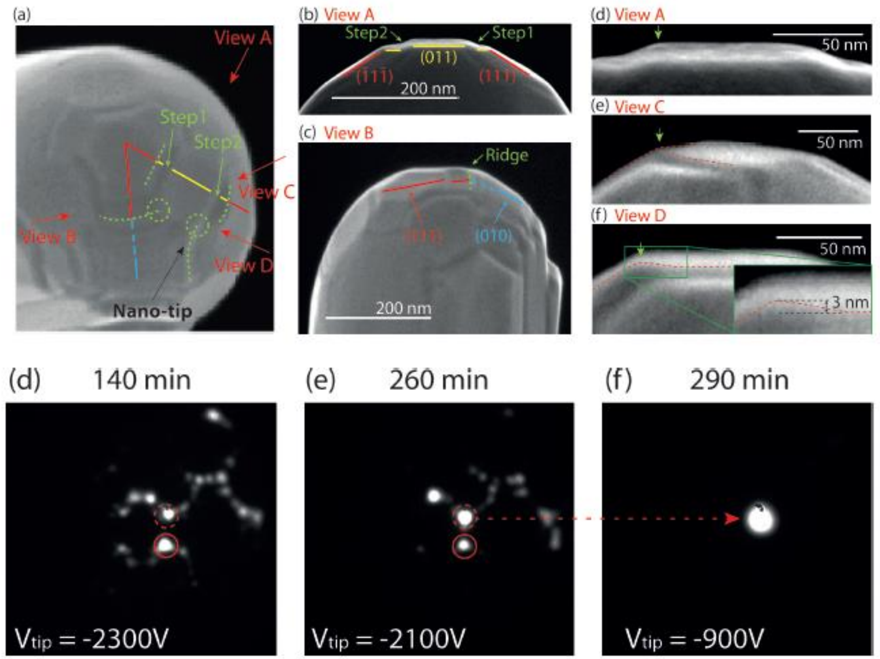
Static surface under el. field





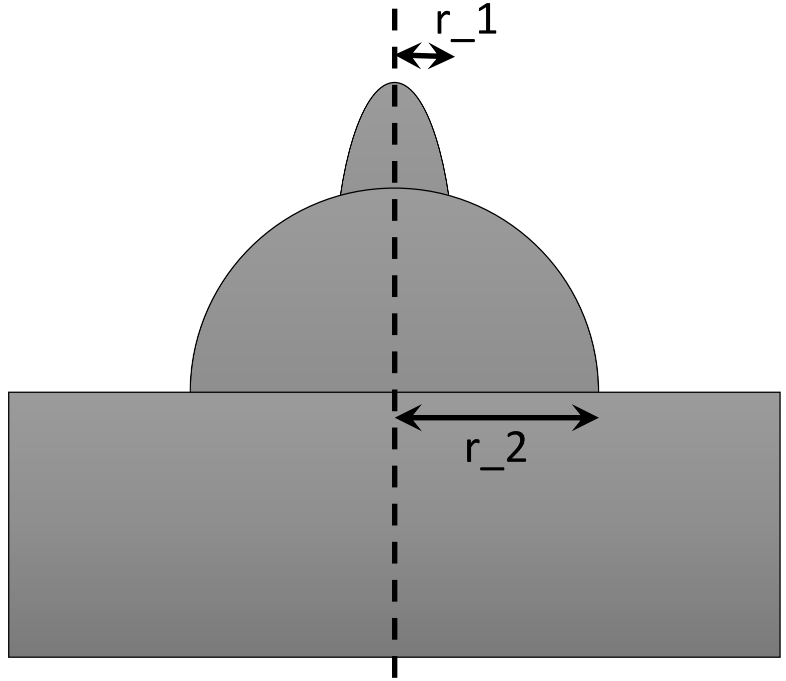
H. Yanagisawa, V. Zadin et al., APL Photonics 1 (2016) 091305

- Field Emission Microscopy experiment
- Collaboration with Dr. Hirofumi Yanagisawa(Max-Planck Institute of Quantum Optics)
- Surface faceting and protrusion formation
- **Possible mechanism for emitter formation**

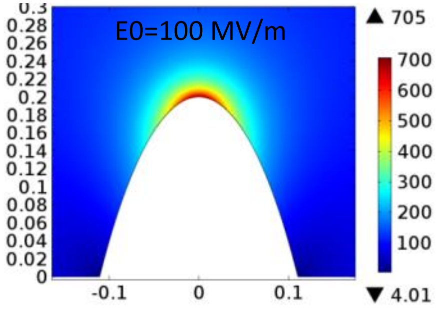


Static behavior of single emitter – sensitivity to surface roughness

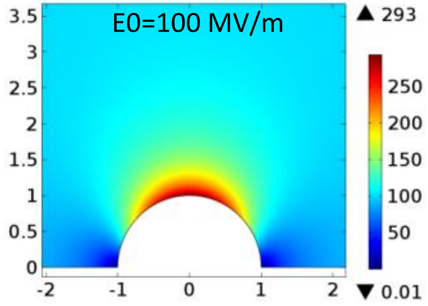
- We can see different surface modifications leading to small β
  - Large β is needed
- Multiplication of field enhancement factors
  - Can explain observed high beta values
- Incorporates surface roughness
- r_1/r_2<0.1 is needed to observe significant influence



Max. enhancement



Reference sim.



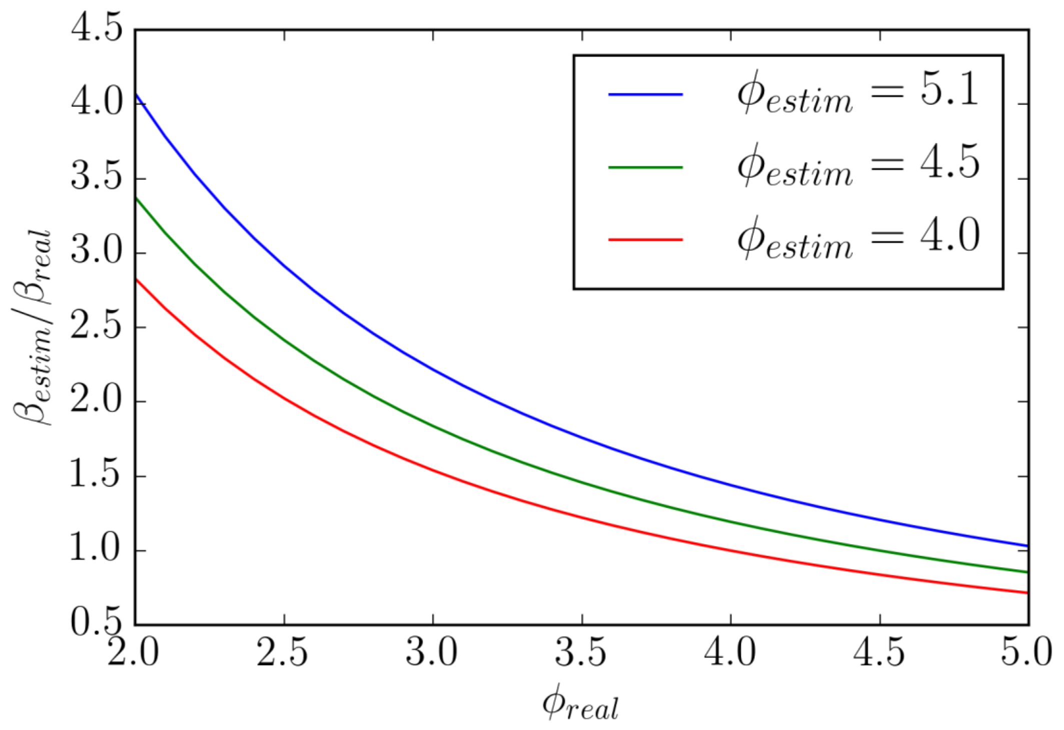
Influence of work function lowering on Fowler-Nordheim plot analysis

The field enhancement factor β is usually found from the slope of FN plot γ by

$$\beta = \frac{-b\phi^{3/2}}{\gamma}$$

The work function is usually assumed to be ϕ = 4.5, but if the real value is different, then the estimated enhancement is

$$\beta_{estim} = \left(\frac{\phi_{estim}}{\phi_{real}}\right)^{3/2} \beta_{real}$$

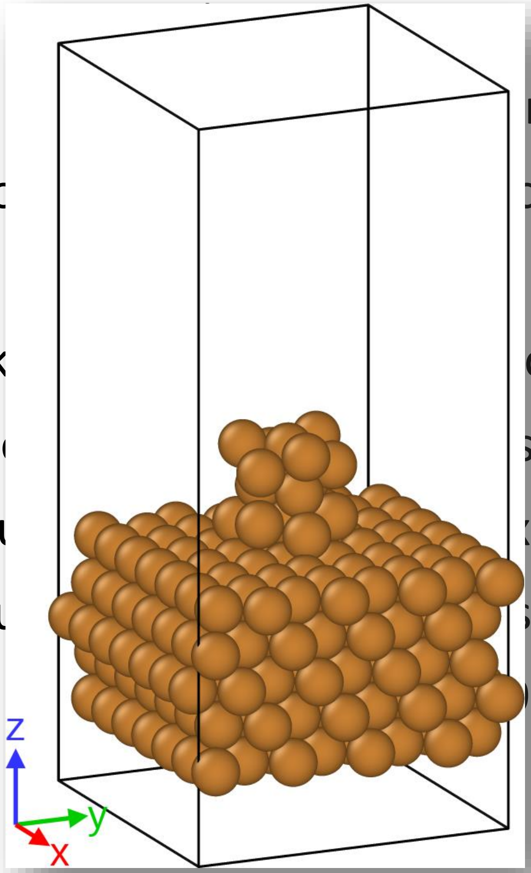


Methodology

DFT calculations

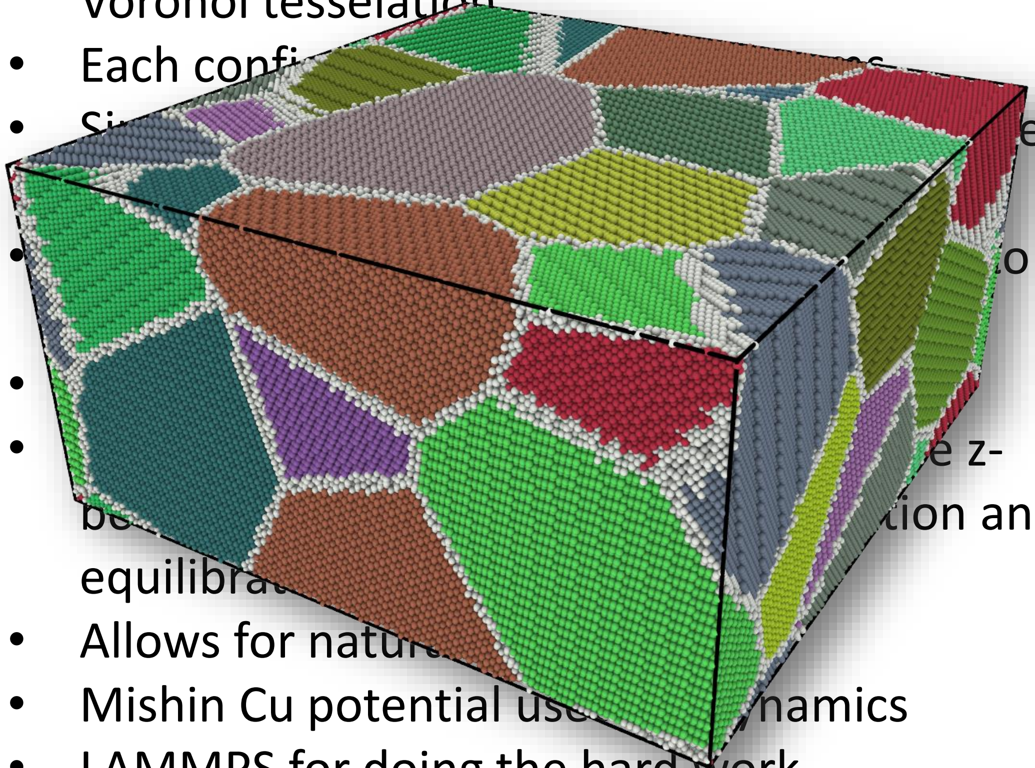
- DFT
- PBE … nctional
- Dipc … c field calculation

- Bulk … oms = 280 total
- Peri … s, vacuum in z direction
- Simu … x40.0 angstroms
- Vacu … s (minus defect)
- 111 … ang^2



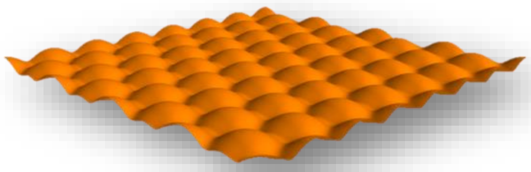
Molecular Dynamics

- Polycrystal model for MD was created with Voronoi tesselation
- Each confi…
- Si… es –
- … o
- …
- … e z-
  be… tion and equilibra…
- Allows for natu…
- Mishin Cu potential use… namics
- LAMMPS for doing the hard work

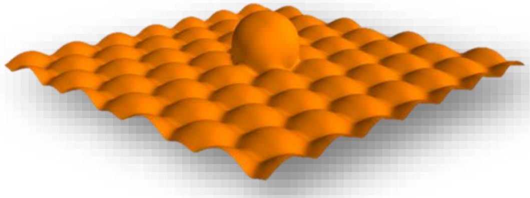


Geometries

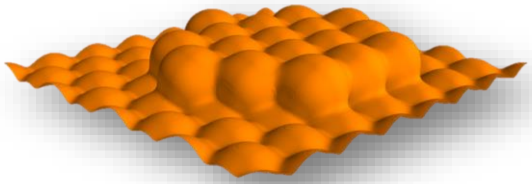
Clean Surface



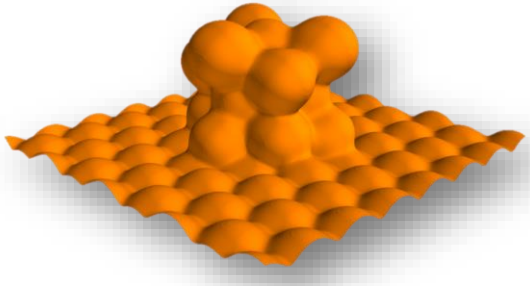
Single



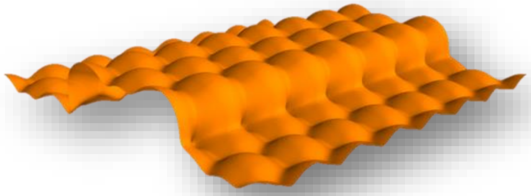
Planar



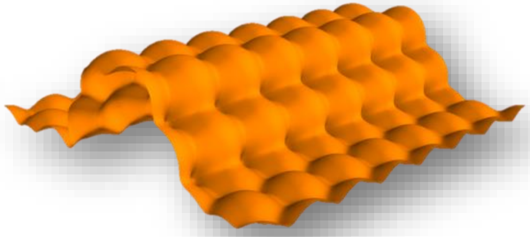
Extrusion 1



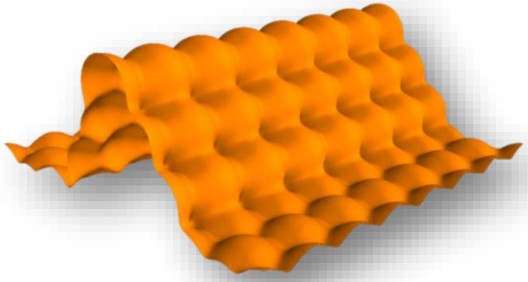
Extrusion 2D 1



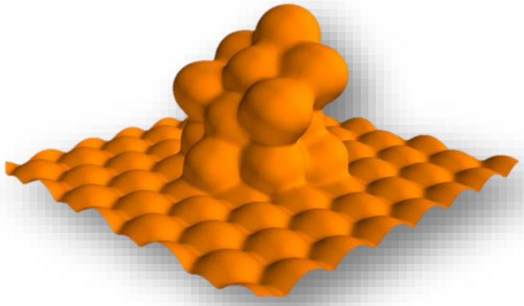
Extrusion 2D 2



Extrusion 2D 3

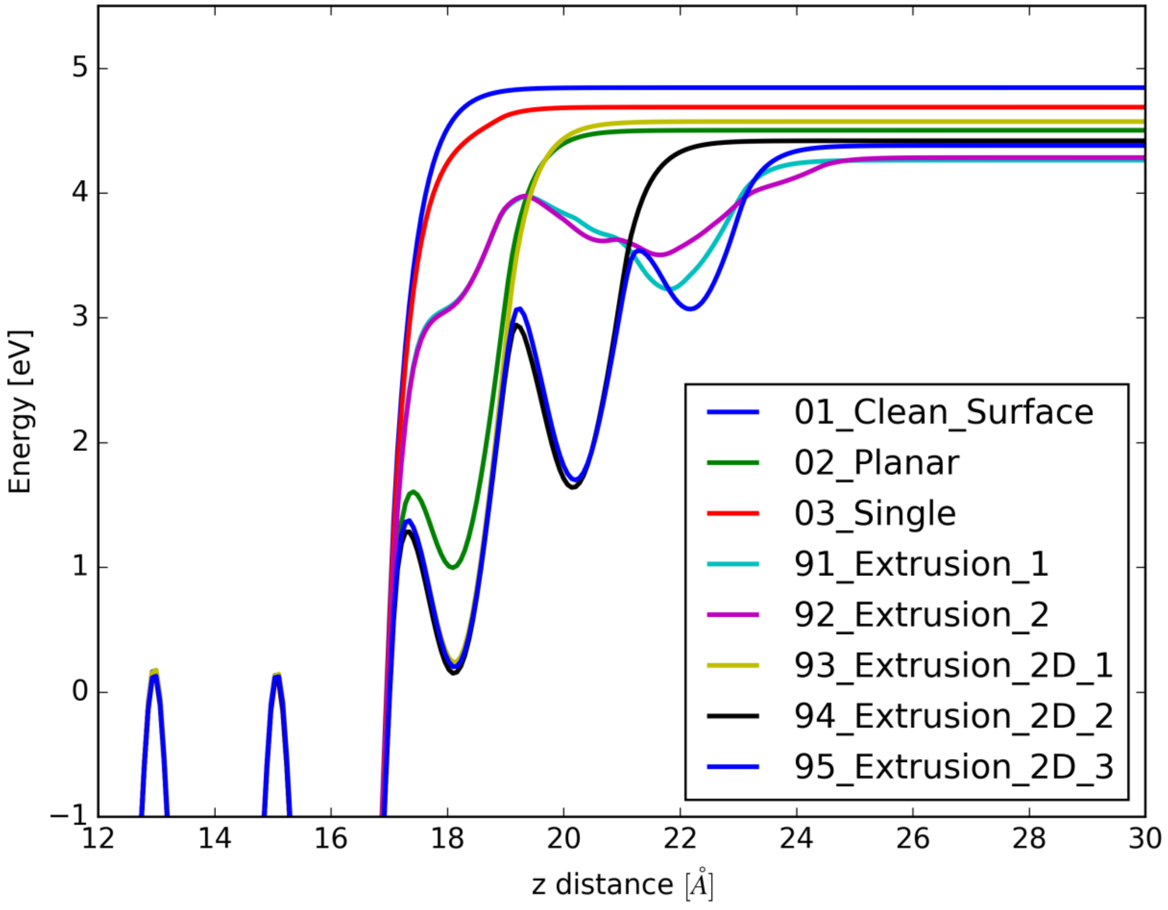


Extrusion 2



Work function decrease due to surface defects

Geometry	WF (eV)	dWF	%
Extrusion 1	**4.26**	**0.58**	**12.0%**
Extrusion 2	4.28	0.56	11.6%
Extrusion 2D 3	4.38	0.46	9.5%
Extrusion 2D 2	4.42	0.42	8.7%
Planar	4.50	0.34	7.0%
Extrusion 2D 1	4.57	0.27	5.6%
Single	4.69	0.15	3.1%
Clean Surface	**4.84**	**0.00**	**0.0%**

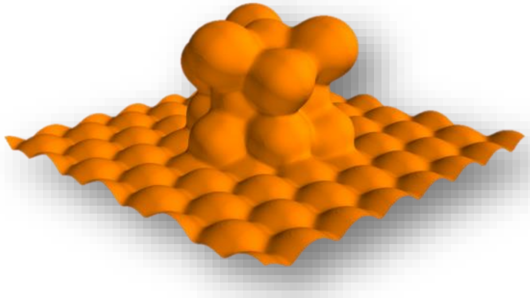


Smooth 111 surface from literature:

- 4.85 (Gartland, P. O., Slagsvold, B. J.: Phys. Rev. B 12 (1975) 4047)
- 4.88 (Kubiak, G. D.: Surf. Sci. 201 (1988) L475.)

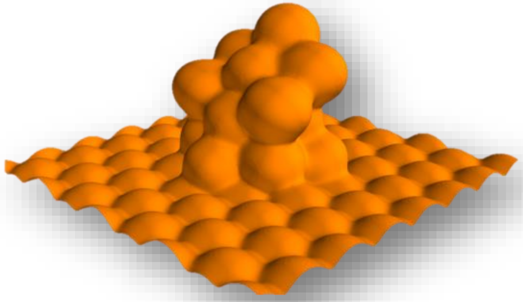
Geometries – arranged by WF

Extrusion 1



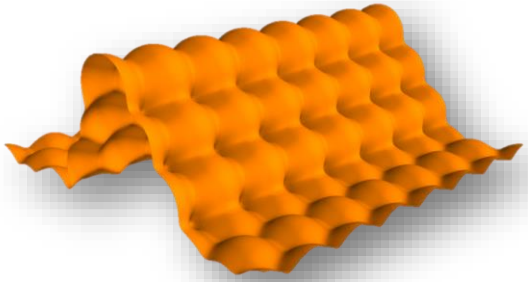
ϕ=4.26

Extrusion 2



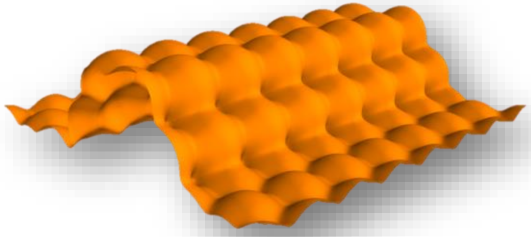
ϕ=4.28

Extrusion 2D 3



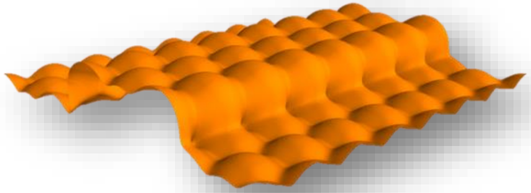
ϕ=4.38

Extrusion 2D 2



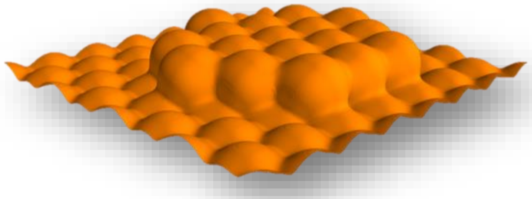
ϕ=4.42

Extrusion 2D 1



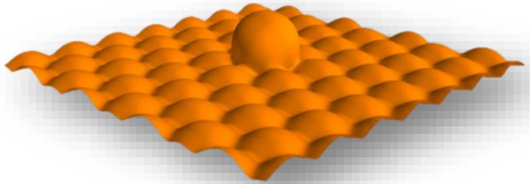
ϕ=4.50

Planar



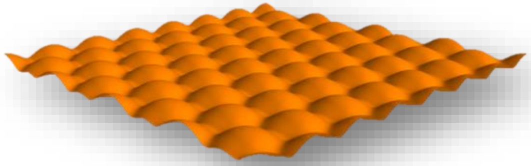
ϕ=4.57

Single



ϕ=4.69

Clean Surface



ϕ=4.84

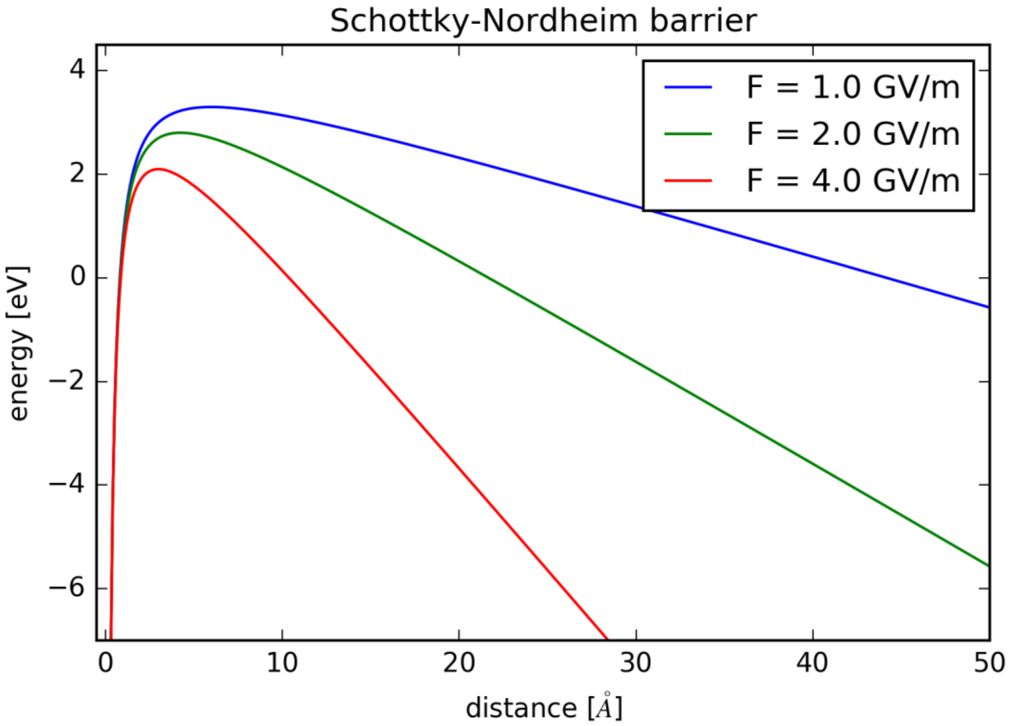
Schottky-Nordheim barrier height

Analytical emission current equations assume the Schottky-Nordheim barrier:

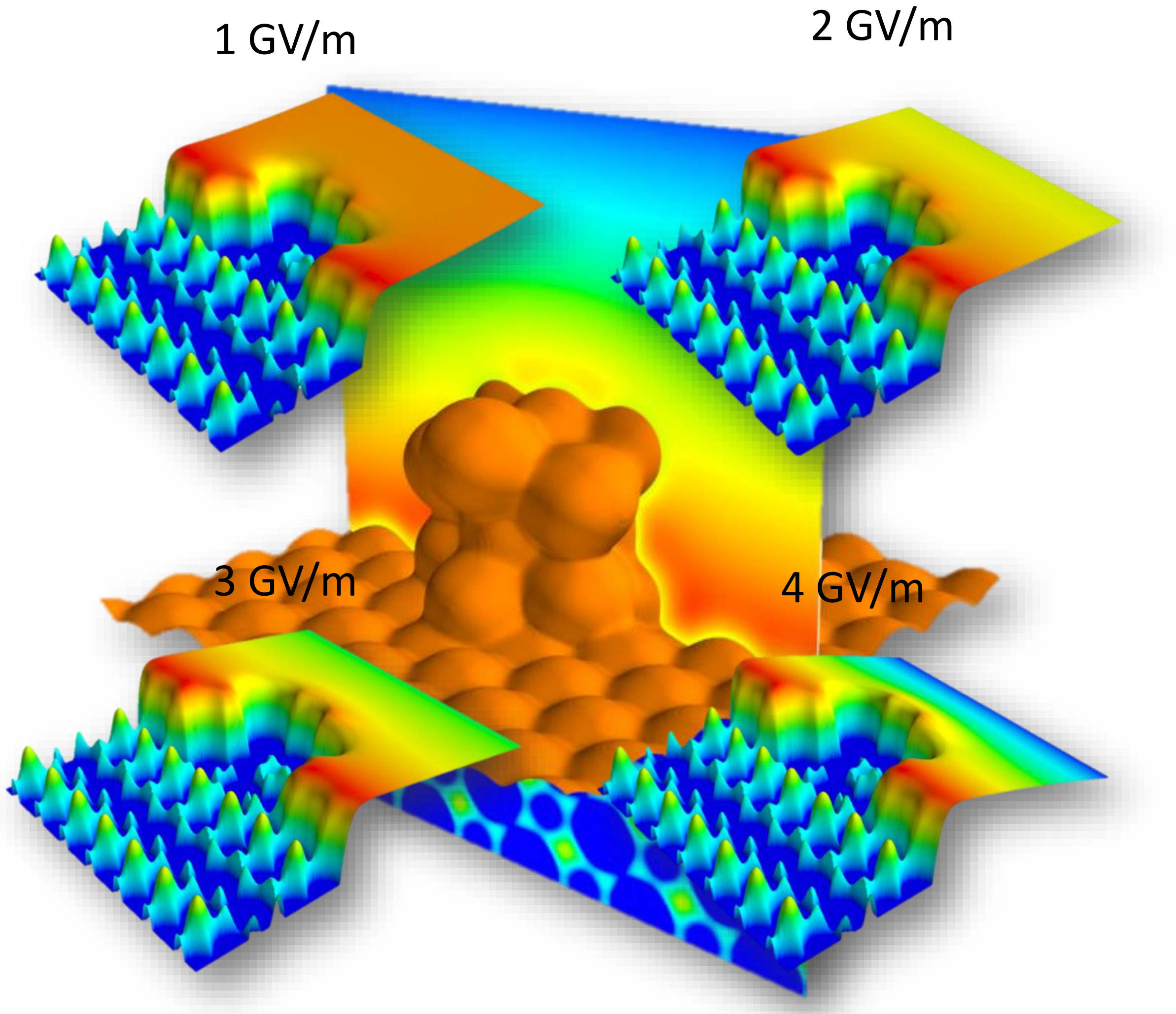
$$V(x) = \phi - qFx - \frac{q^2}{16\pi\epsilon_0 x}$$

Maximum of the barrier depend on the electric field F

$$V_{max} = \phi - \frac{q}{2}\sqrt{\frac{F}{\pi\epsilon_0}}$$



Potential landscape around defects



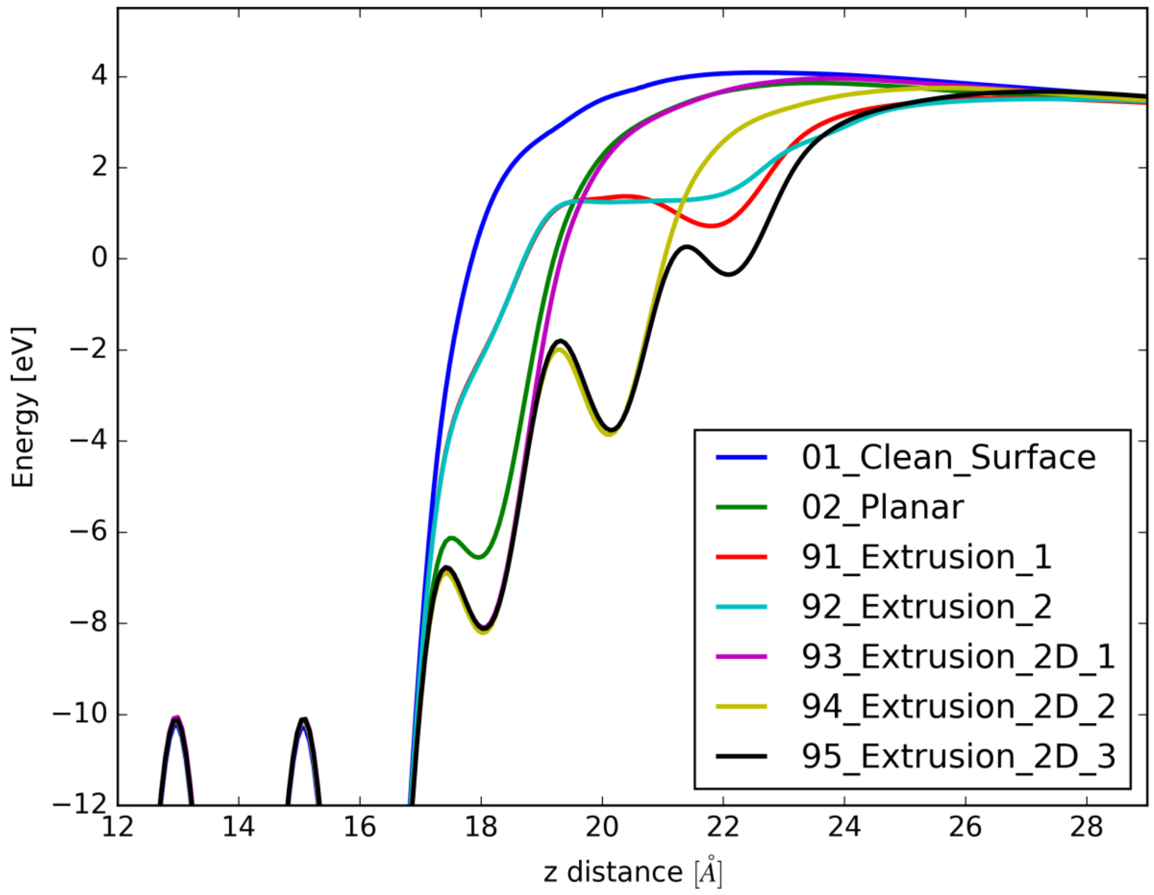
- Potential landscape for different rough surface features with DFT under electric field
    - DFT calculations done for multiple electric field values (potential shown for a geometric slice)
    - Complex potential landscape is formed due to the surface protrusions

- Application of Schottky-Nordheim barrier for such defects questionable
- Difficulties with surface curvature corrected barriers expected as well (GETELEC code)
- Possible solution for work function estimation based on estimation of electron tunneling probabilities

Barrier height dependence on el. field

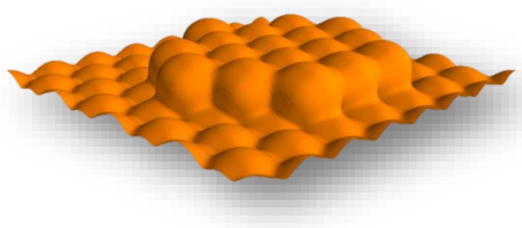
Geometry	BH	WF-BH
Clean Surface	4.08	0.76
Planar	**3.86**	**0.64**
Extrusion 1	3.52	0.74
Extrusion 2	**3.51**	**0.77**
Extrusion 2D 1	3.96	0.61
Extrusion 2D 2	3.75	0.67
Extrusion 2D 3	3.67	0.71

Used El. field is 1 GV/m

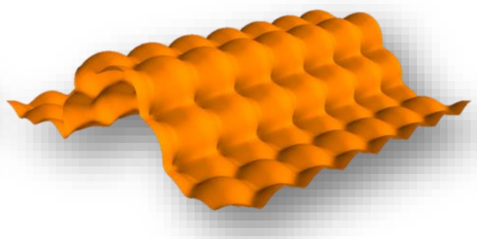


El field influence to work function

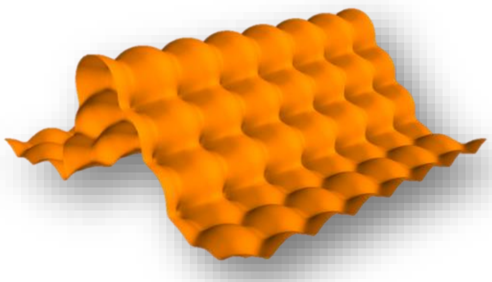
Planar

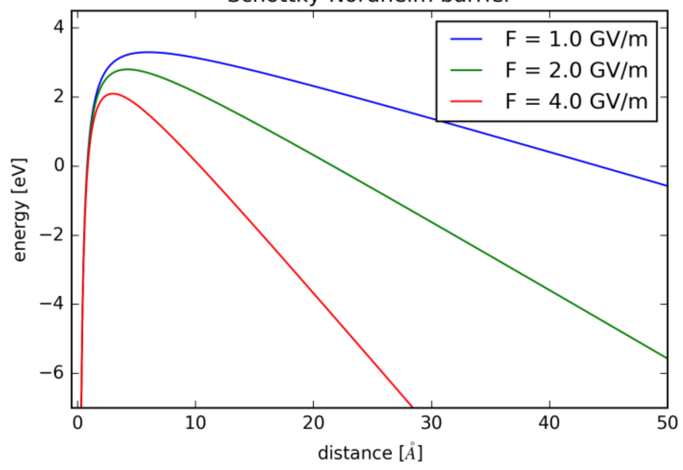


Extrusion 2D 2



Extrusion 2D 3





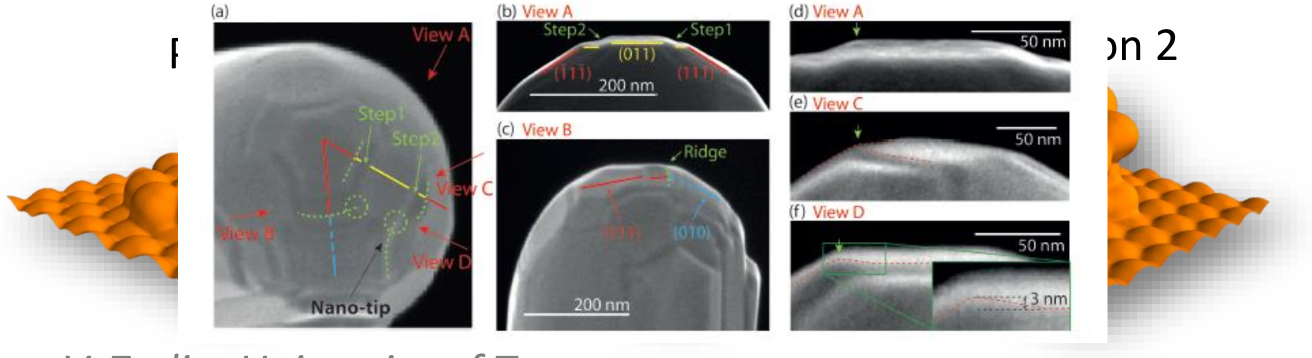
Geometry	WF	BH	WF-BH	(WF-BH)/WF (%)
Clean surface	**4.84**	4.08	0.76	15.7%
Extrusion 2D 1	**4.57**	3.96	0.61	13.3%
Planar	**4.50**	3.86	0.64	14.2%
Extrusion 2D 2	**4.42**	3.75	0.67	15.2%
Extrusion 2D 3	**4.38**	3.67	0.71	16.2%
Extrusion 2	**4.28**	3.51	0.77	18.0%
Extrusion 1	**4.26**	3.52	0.74	17.4%

- Changes in barrier height remain similar to the clean surface
- Difference between WF and BH used to evaluate work function changes due to field
  - Current work function estimation methodology may be too crude
- El. Field influence to the work function expected to remain small

System energies

- **General trend – system energy decreases when** electric field is applied
  - Field makes surface modifications more stable
- Surface with islands preferred over surface with adatoms
  - Good agreement with previous KMC simulations by V. Jansson
  - Effect observed in experiments as well
- **Applied field lowers planar defect energy below clean surface energy**
  - **Surface roughening due to field energetically feasible**

F on 2

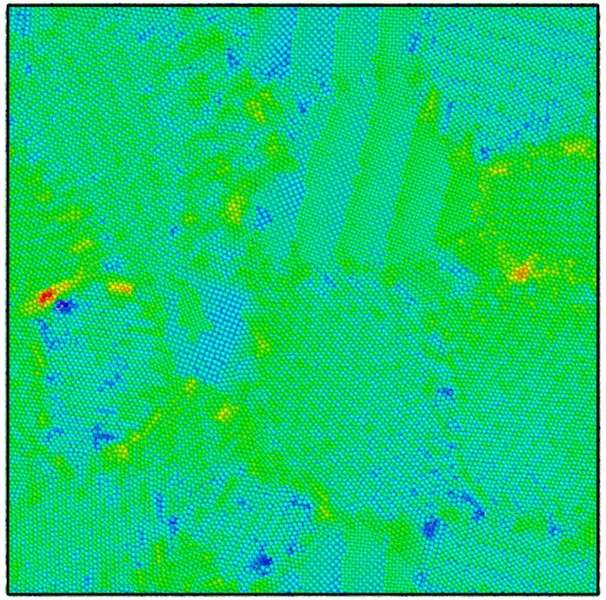


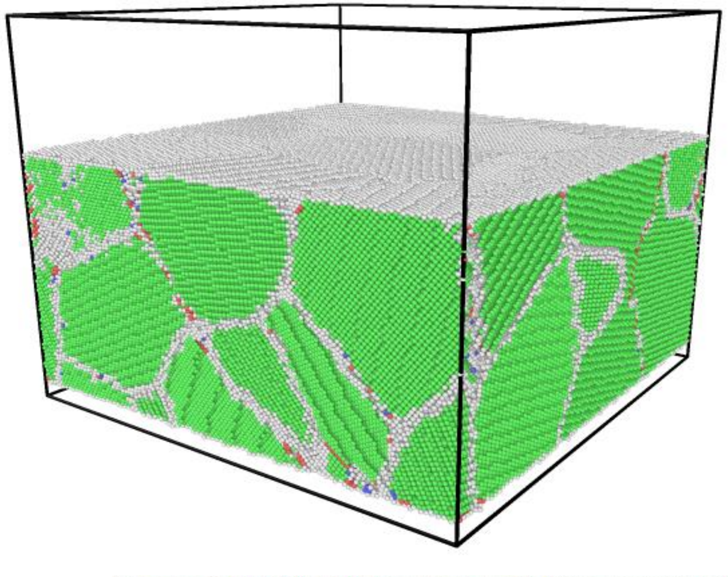
Energy per atom for all geometries (in **meV** = 10^-3 eV)

Field (GV/m)/ Geometry	0	1	2	4
Clean Surface	**0.0**	**-0.36**	**-1.45**	-5.81
Planar	**3.67**	3.46	2.50	**-1.69**
Single	**3.90**	3.61	-	-
Extrusion 1	**24.85**	**24.72**	**23.71**	**13.93**
Extrusion 2	**24.62**	**24.46**	**23.42**	**-**
Extrusion 2D 1	0.70	0.46	-	-
Extrusion 2D 2	5.94	5.72	-	-
Extrusion 2D 3	12.78	12.56	-	-

Polycrystalline copper surface deformations due to applied electric field

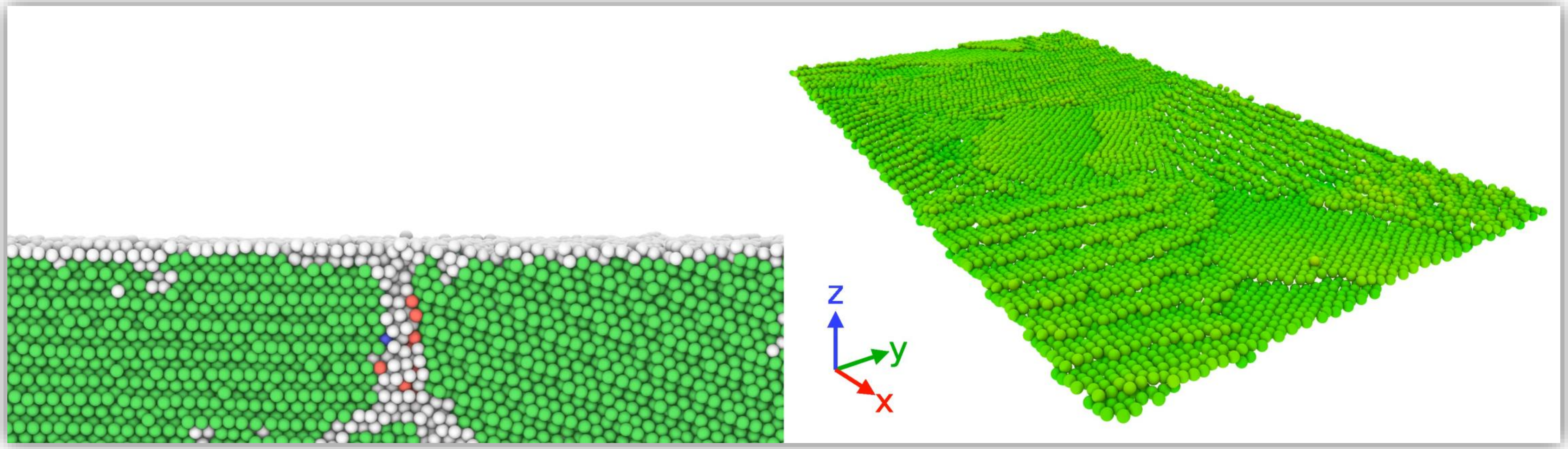
- Mechanical response of polycrystalline surfaces
- Interaction of grain boundaries with a free surface in electric field
- Electric field is modelled as a force on the surface atoms perpendicular to the surface
- Preferential surface diffusion towards the intersections of grain boundaries with the surface





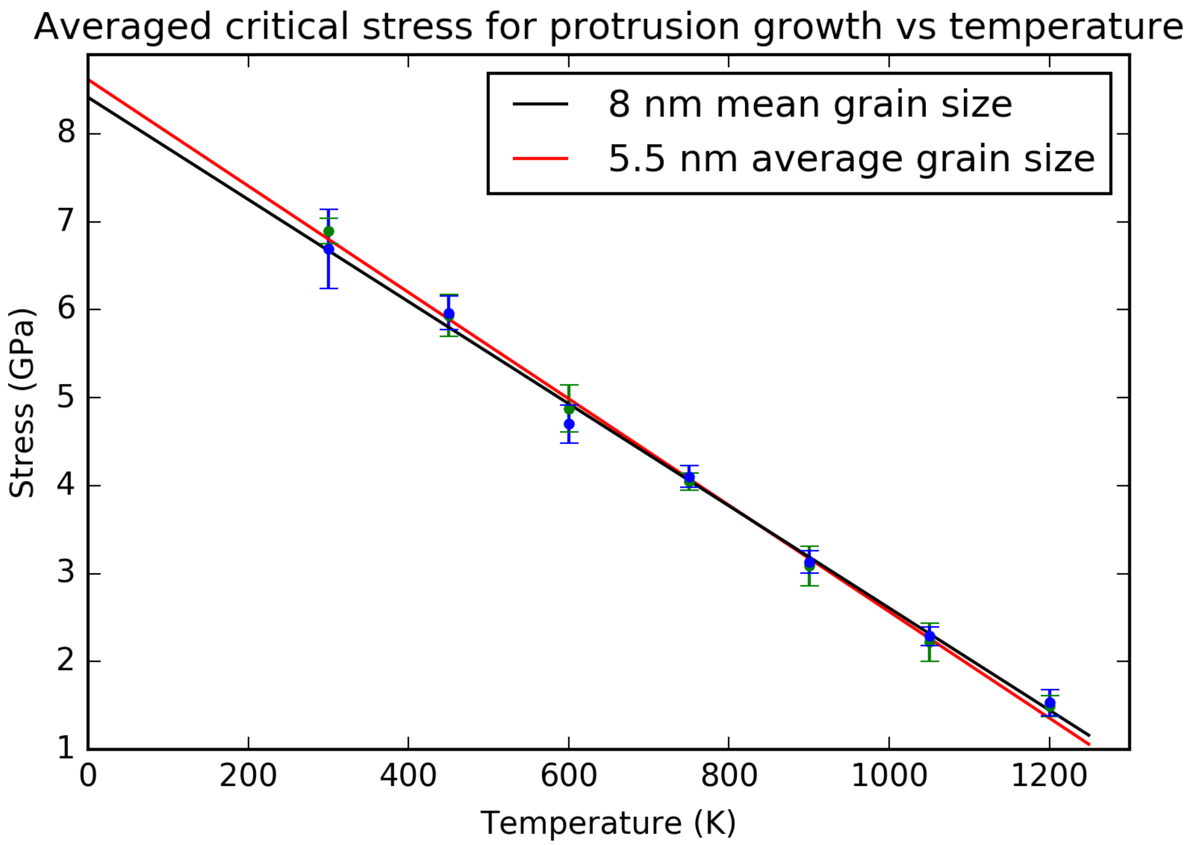
- Surface atoms identified dynamically during the run by coordination analysis
- Systems generated using Voronoi tessellation
- **Green** – fcc atoms,
- **Grey** – surface and grain boundary atoms
- **Red** – stacking faults

Surface protrusion evolution

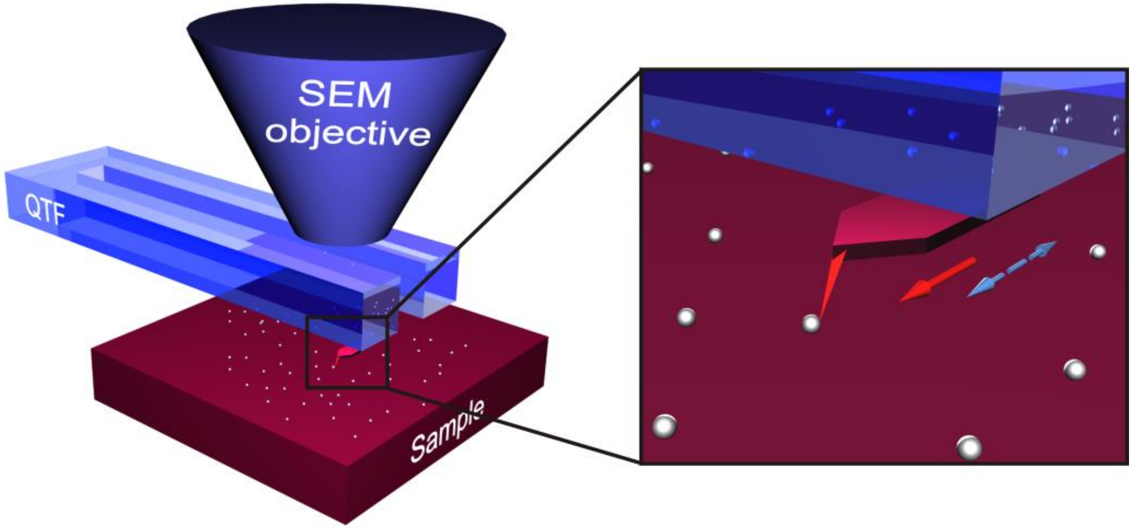


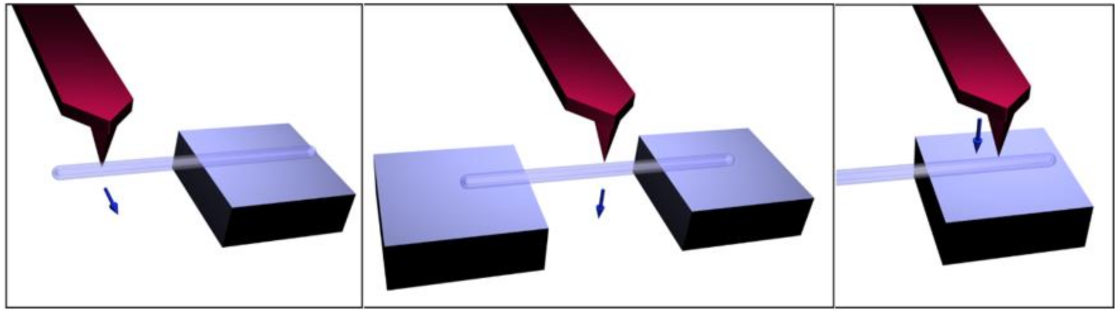
Critical stress dependence on temperature

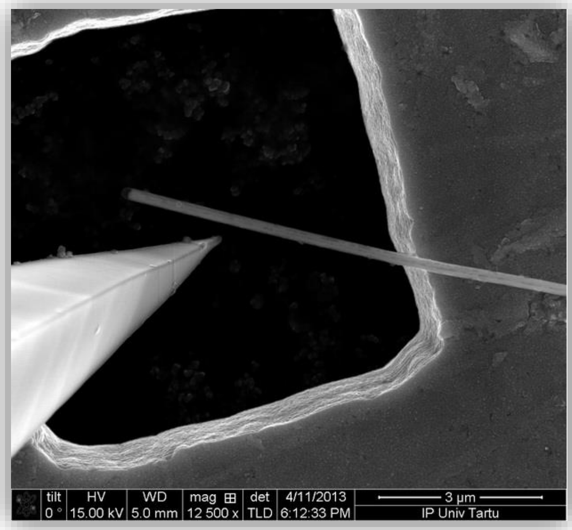
- Necessary stress for nucleation of surface
  protrusions decreases linearly with temperature from 300K to 1200K
- Stress needed for nucleation of dislocation ~4 GPa

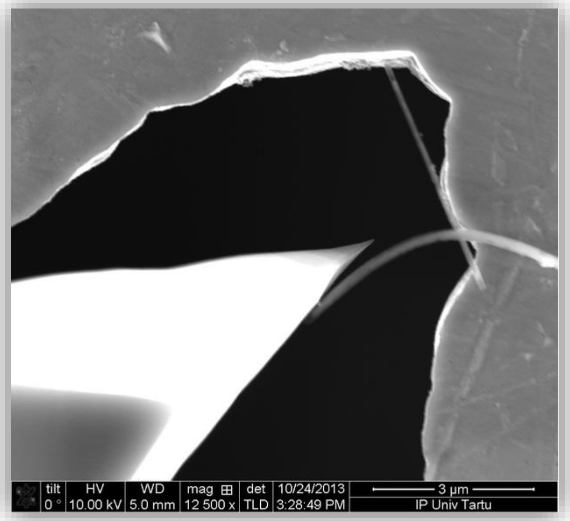


Future plans









Conclusions

- Significant work function decrease due to the surface morphology changes
  - Flat surface 4.84 eV vs. defect 4.26 eV
- Electric field leads to formation of complex potential landscape near defects, application of Schottky-Nordheim barrier complicated
  - Current work function estimation methodology may be too crude
  - Influence of electric field to the work function estimated to be small
- Spontaneous protrusion formations possible
  - From grain boundaries or extension to other disordered material defects or spots
- Future plans: In situ SEM tests with nanowires under electric field
  - Aim to observe electric field induced surface modifications

Thank You for Your attention!

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