Material surface modifications due to combined influence of femtosecond laser irradiation and applied electric fields

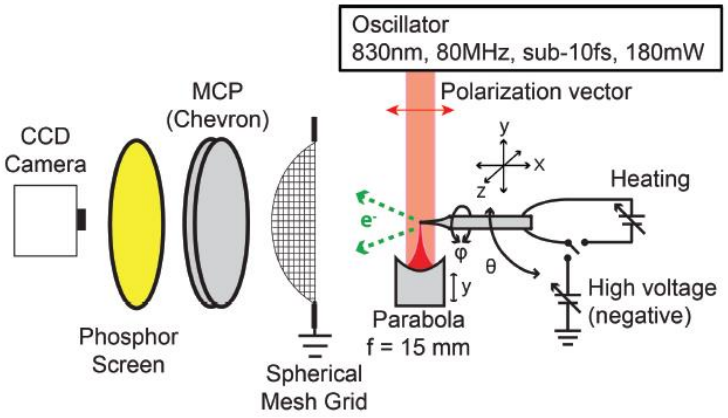
V. Zadin, S. Vigonski, K. Kuppart, R. Aare, A. Aabloo

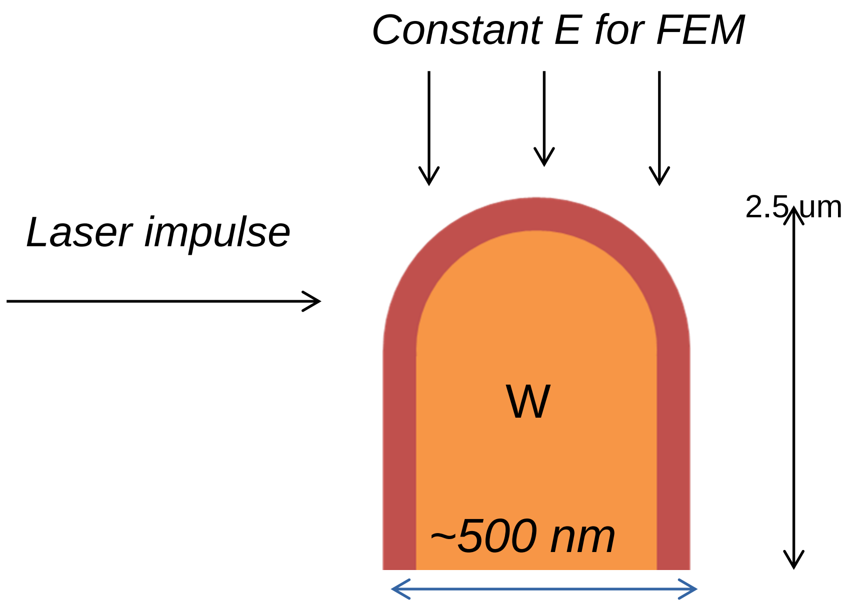
V. Jansson, E. Baibuz, A. Kyritsakis, F. Djurabekova

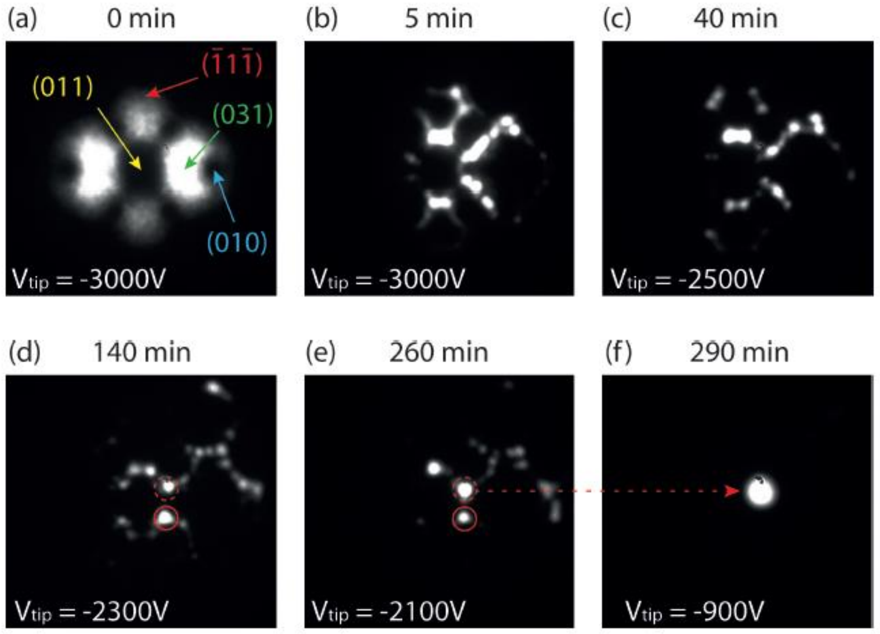
2019

Static surface under el. field

- 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**

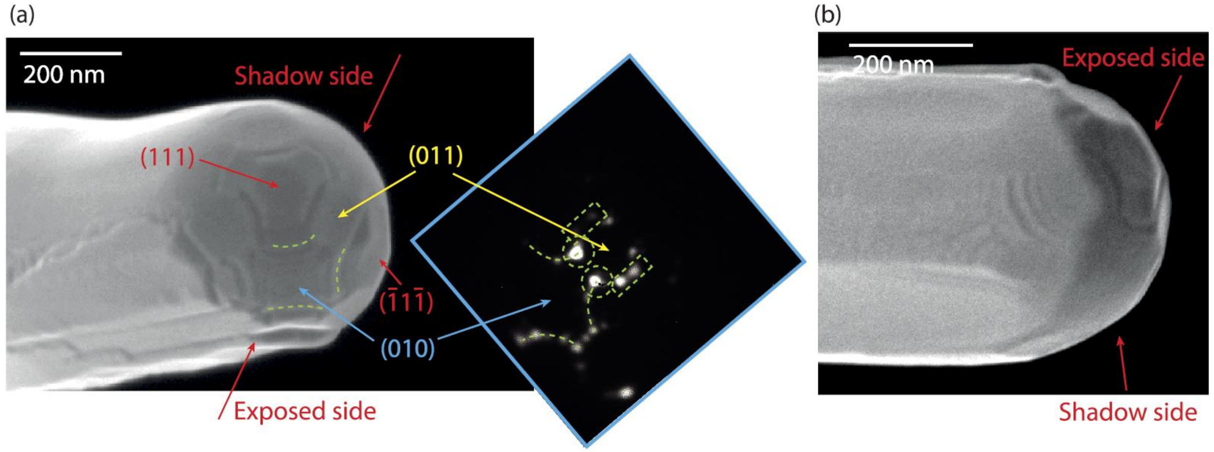


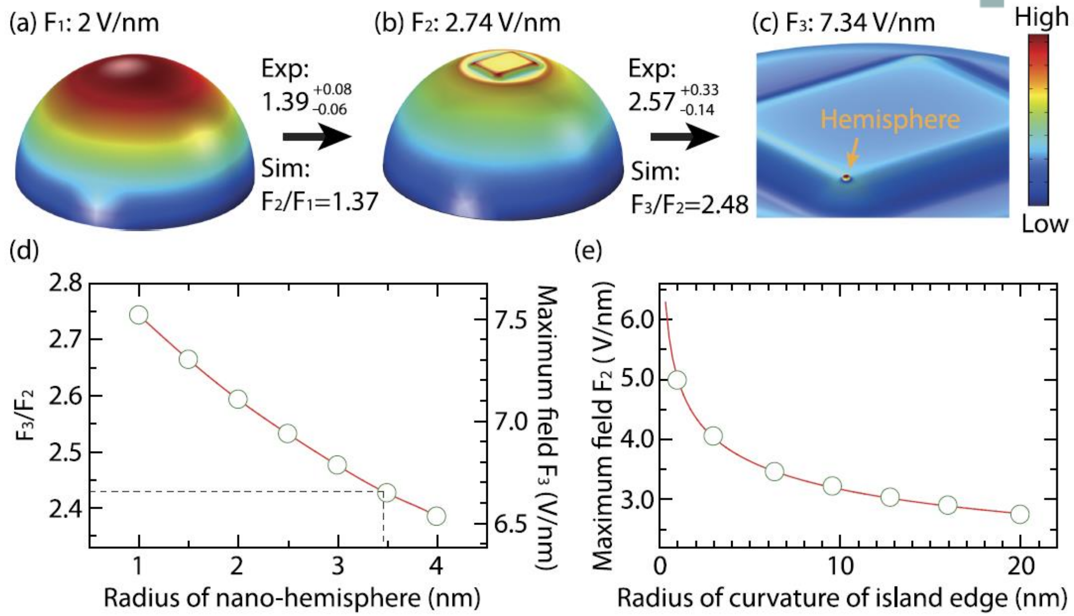




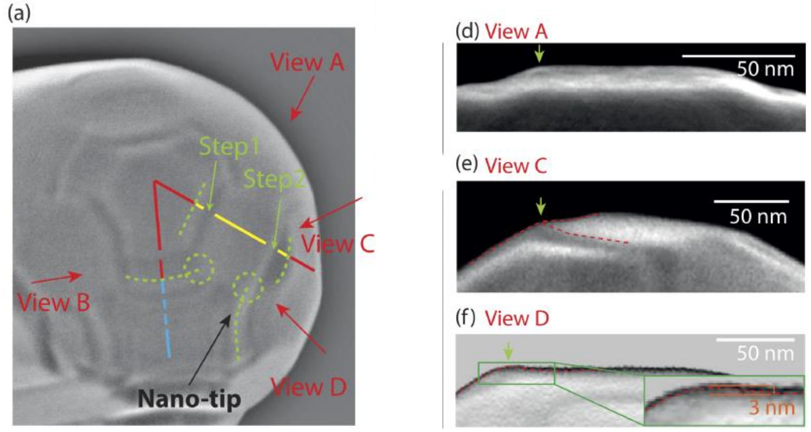
H. Yanagisawa et al., APL Photonics (2016)

The faceting as seen on SEM images





- The faceting may create nanotips along the ridges formed between separate facets.
- After **heating** up to 80% of the melting temperature, **facets disappear**, making the experiment **repeatable.**
- **El. field calculations with FEM agree with tip hypothesis**



H. Yanagisawa et al., APL Photonics (2016)

Fujita *et al.*: Faceting of a single crystal W tip

- Similar experiment setup - results shown are with static T=2300 K
- Lower temperatures behaved differently, omitting D-F.
- **Single crystal tip.**

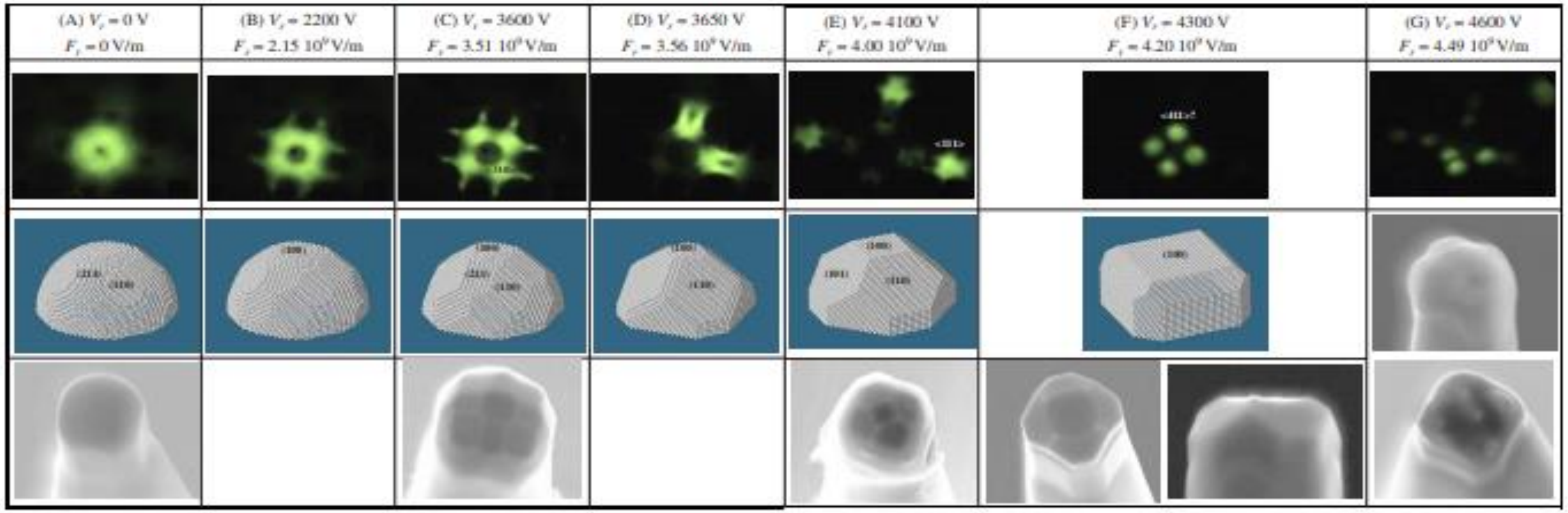
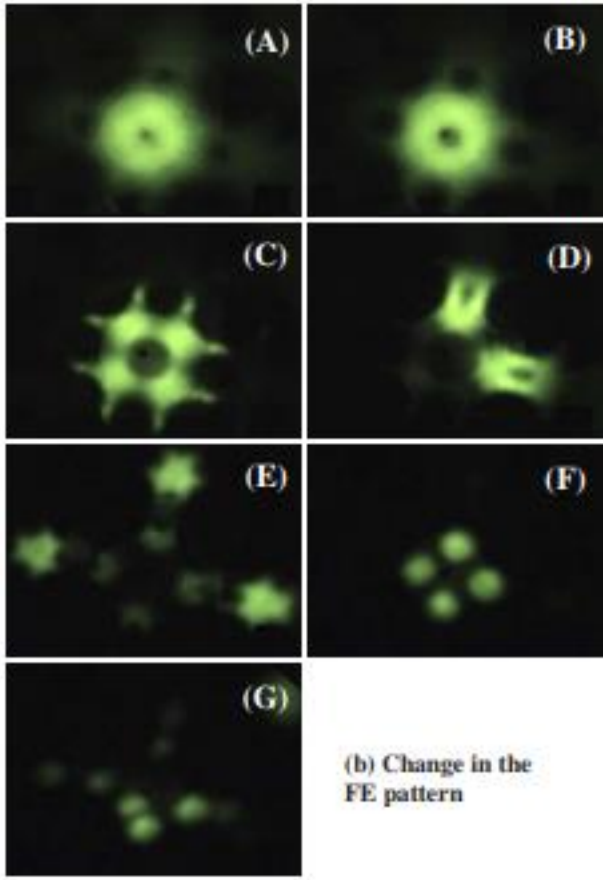
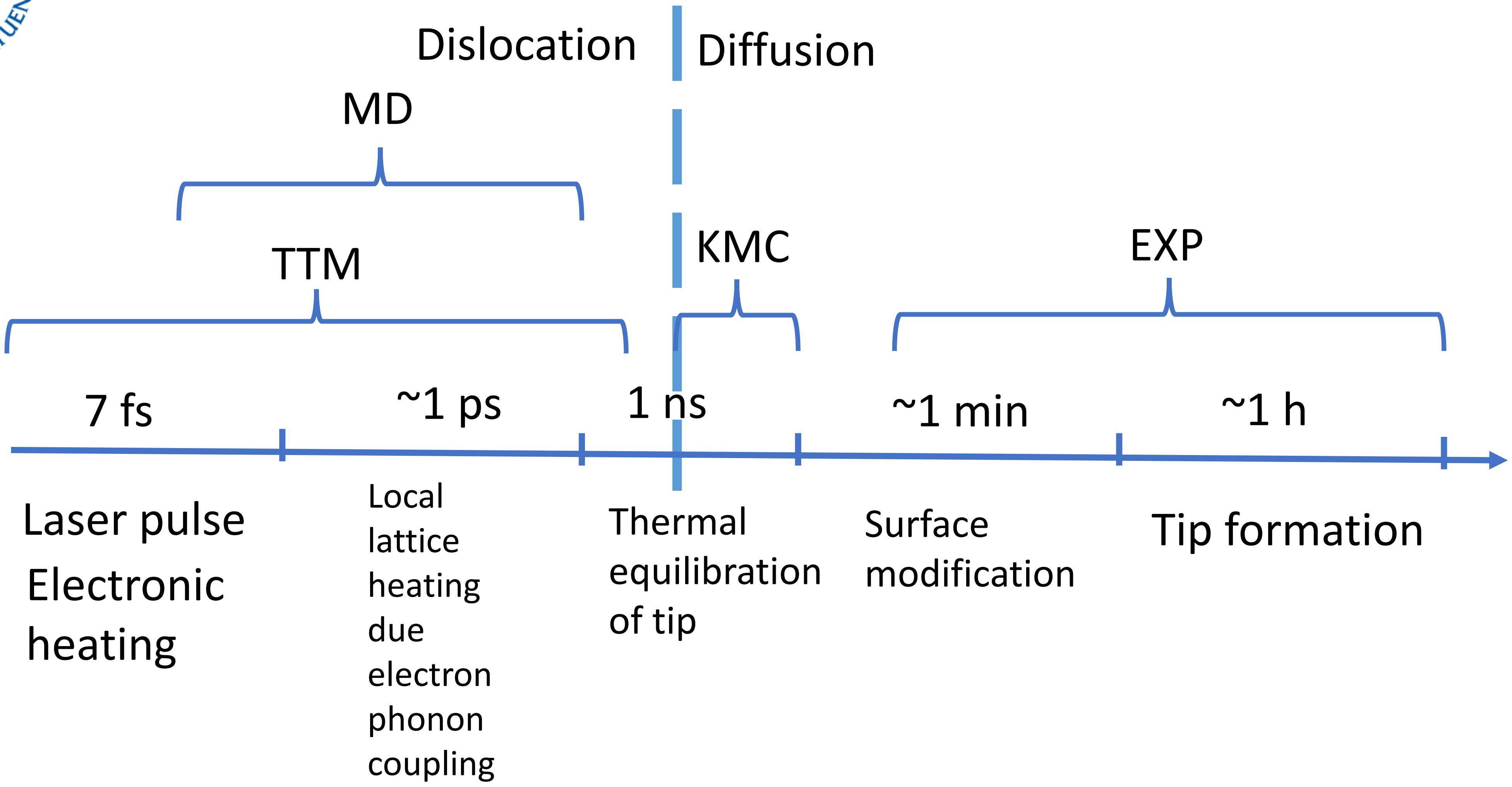


FIG. 5. (Color online) Summary of the evolution of the tip shape in the remolding process at the temperature T_r=2300 K. The FE patterns (second row), the emitter tip shape models (third row), and the SEM images of the tips at selected stages (fourth row) are given in the order of the increasing remolding voltage.



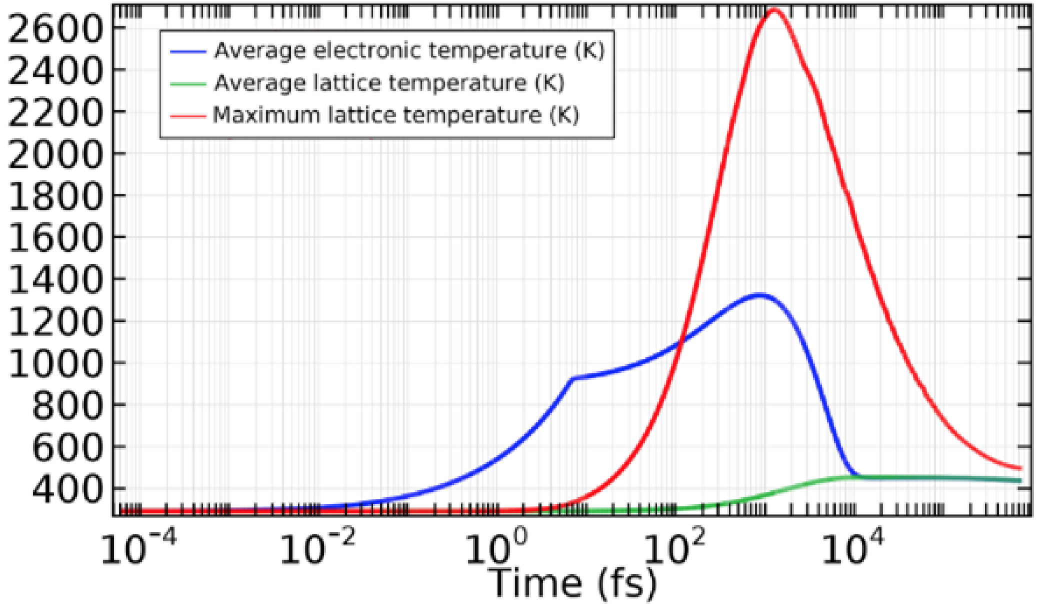
Fujita *et al.*, "Mechanism of surface-tension reduction by electric-field application: Shape changes in single-crystal field emitters under thermal-field treatment", Phys. Rev. **75**, 2007

Reconstruction of process timeline



- All methods will leave gaps in timeline

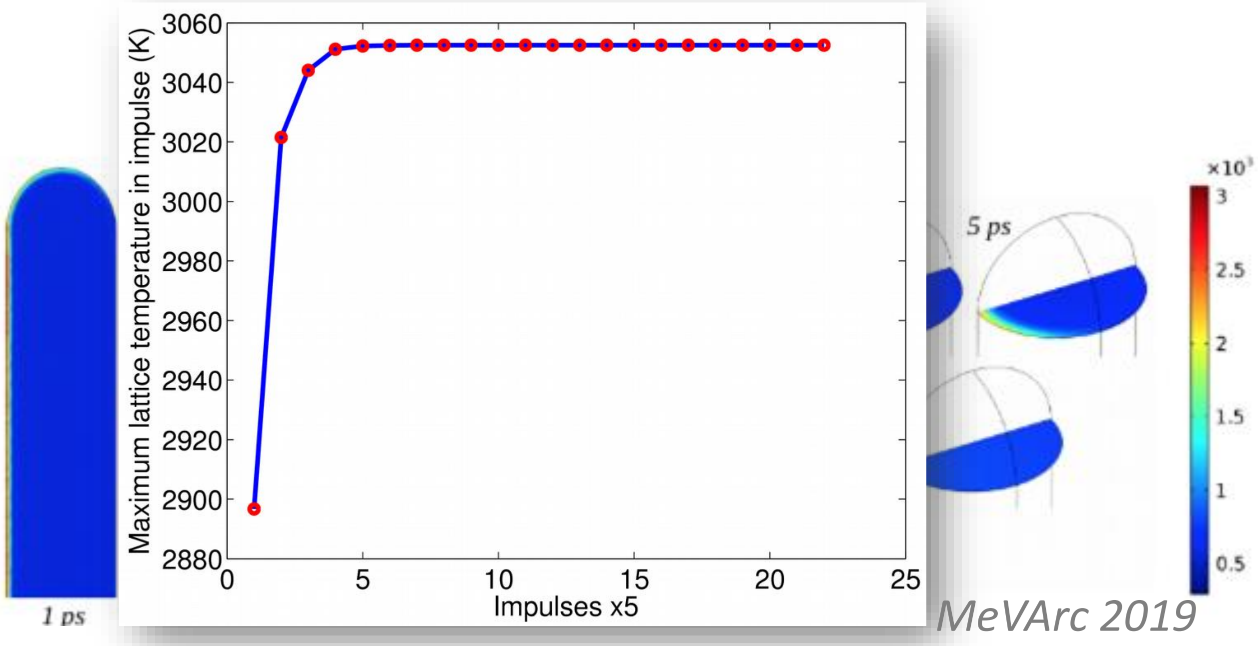
TTM – dynamic heat deposition



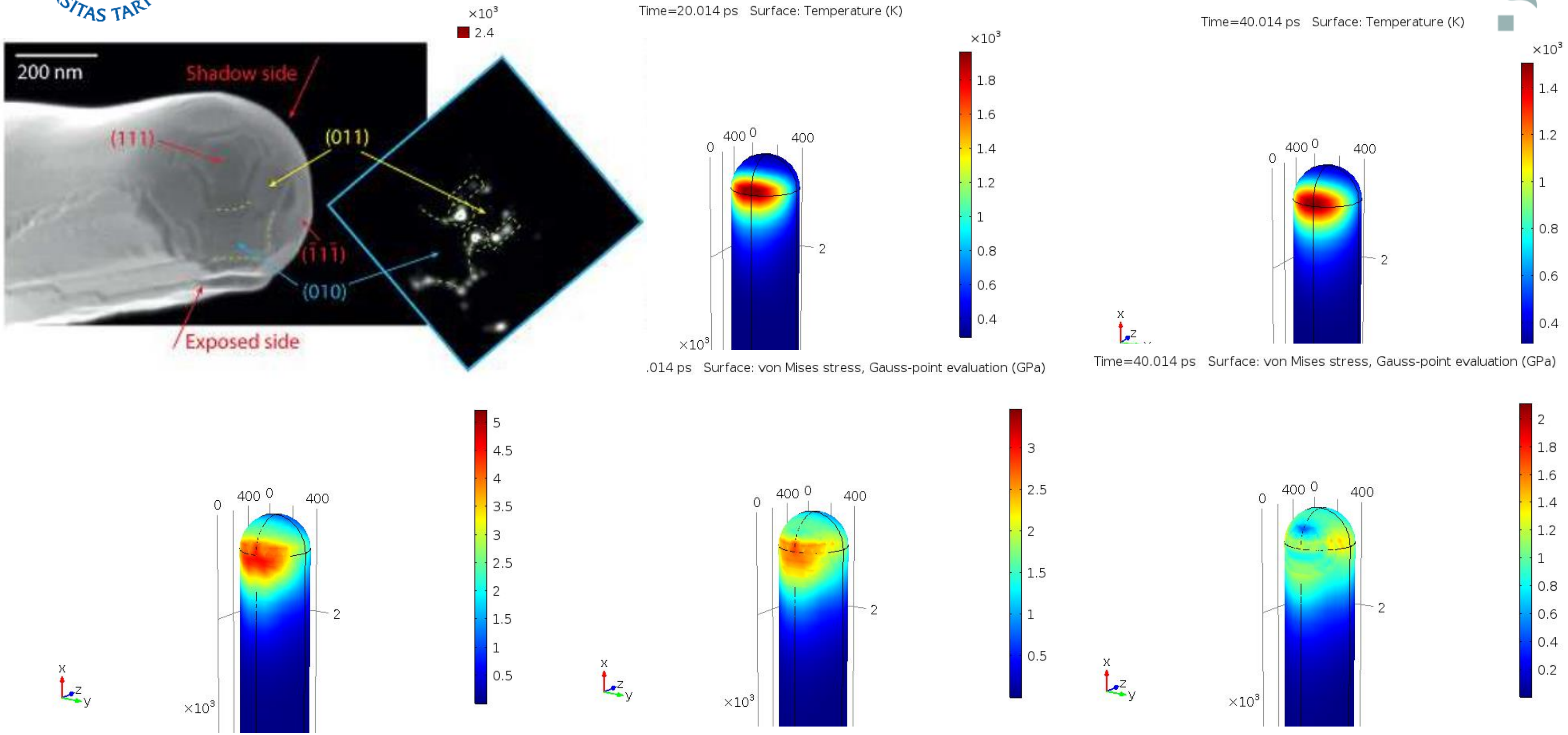
Electronic and lattice temperature behaviour due to

- laser + emission currents
- energy deposition+TTM+resistive heating+Nottingham effect

- The maximum temperature reached is 2689.7 K,
  - occurring 1.26 ps after the laser impulse,
  - just after the average electronic temperature peaks,
- Temperatures above 2500 K exist for 1.6 ps within 5nm of the crystal surface



TTM results – surface maps from laser irradiation

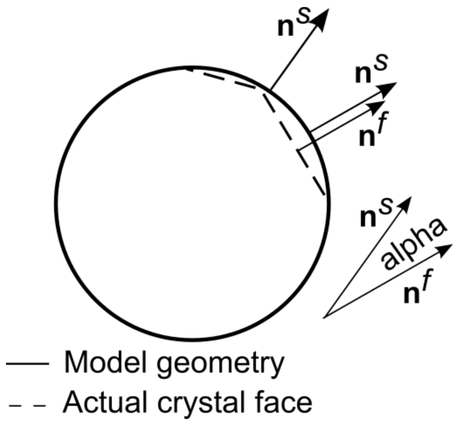


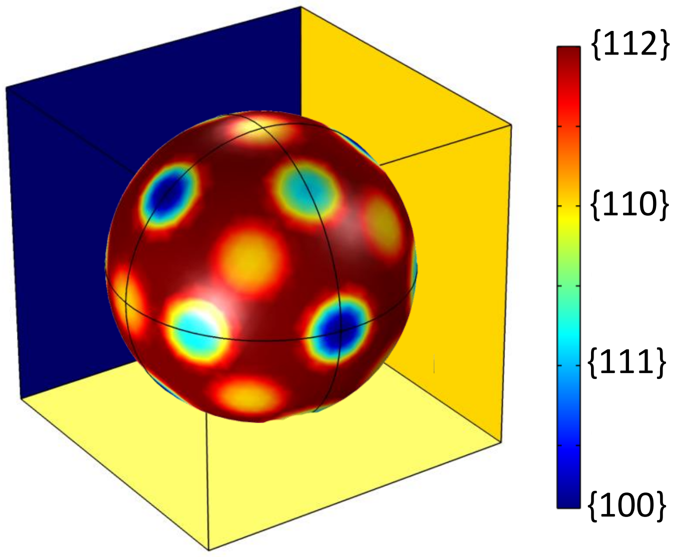
Surface stress using FEM

- Elastic deformation of material, simulation of large strains:
  - Anisotropic bulk material model
  - Surface stess is calculated using thin layer aproximation in boundary layers
  - Surface parameters are isotropic but crystal face dependent
- Crystal faces detected algorithmically:
  - Ability to simulate arbitrary shapes
  - Only crystal orientation is needed to intialize the simulation
- Initial surface stress and elastic parameters of surface from MD simulations
- Fully copled surface and bulk stress model

	Stress	Strain
Bulk model	$S = C : \varepsilon^b$	$\varepsilon = \frac{1}{2}(F^T F - I)$
Surface model	$\tau_{ij} = \tau_{ij}^0 + S_{ijkl}\varepsilon_{kl}^s$	

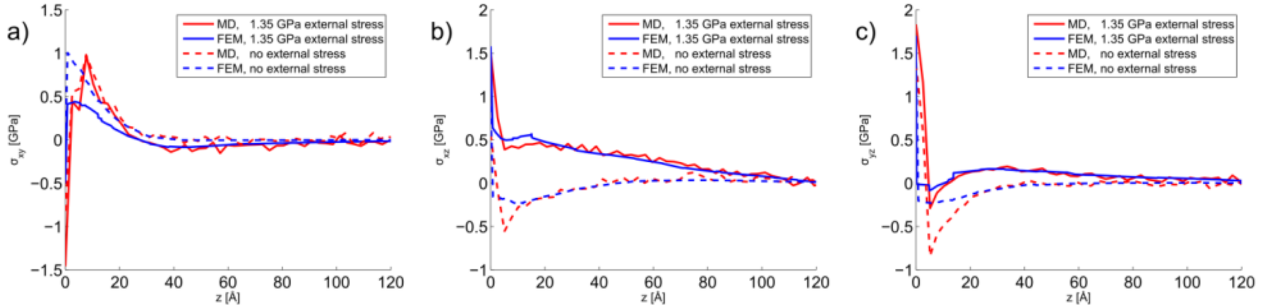
Crystal face detection

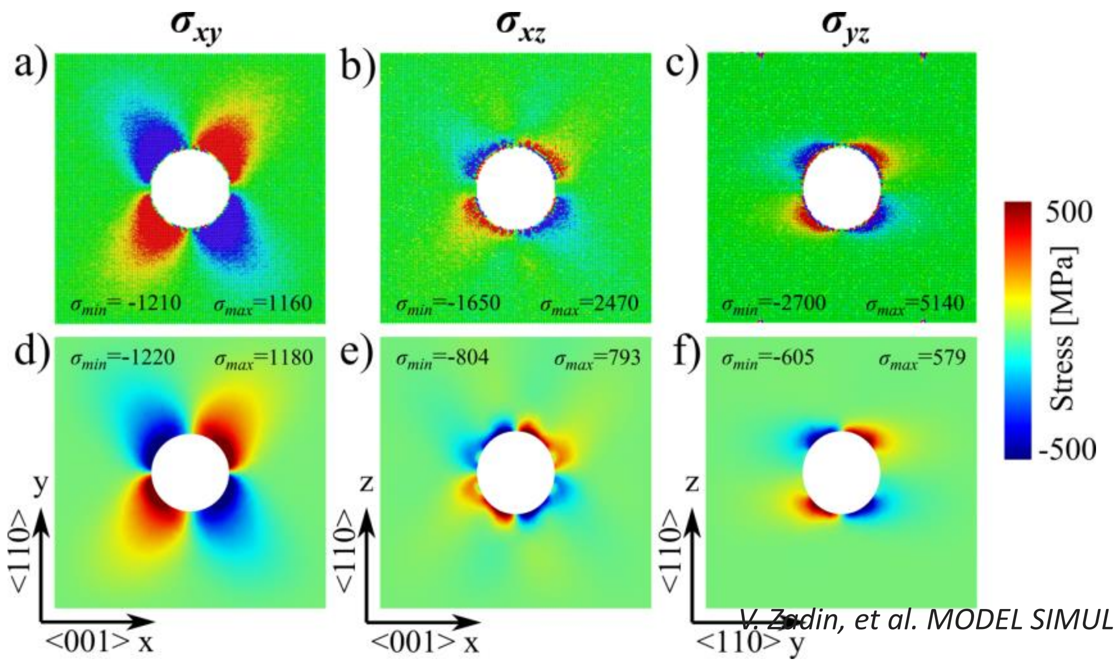




Surface stress effects in nanoscale modelling

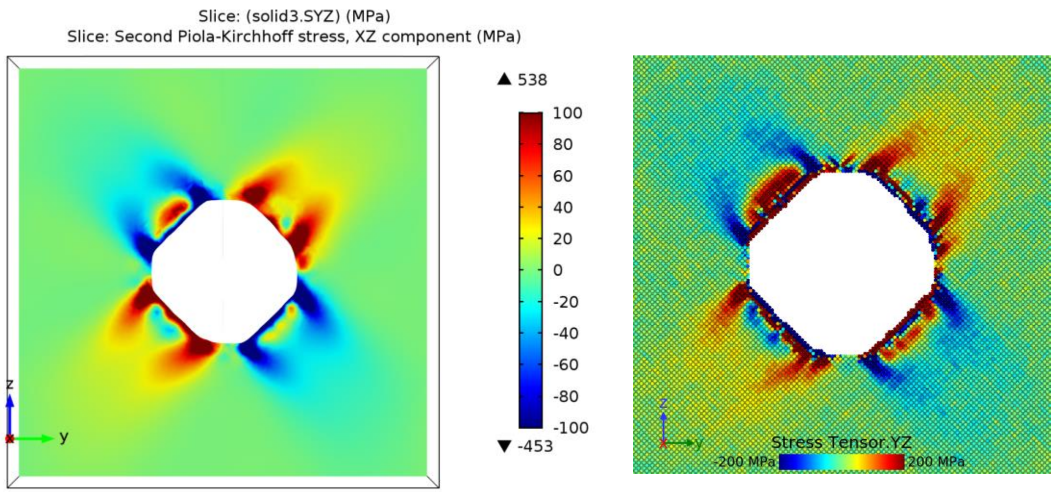
- Anisotropic material model
- Crystal plane dependent surface properties
- The surface effects important in range ~ 6-10 nm
  - Corrections for surface stress (surface tension)
  - Model complexity improved towards nonlocal simulations
  - Strongest/weakest nanostructure estimation
- Plastic deformation
  - Accurate limits to be determined
  - Dependence from grain size, average dislocation length and plastic deformation activation volume
  - More complex model needed to account microstructure effects, dislocation densities etc.





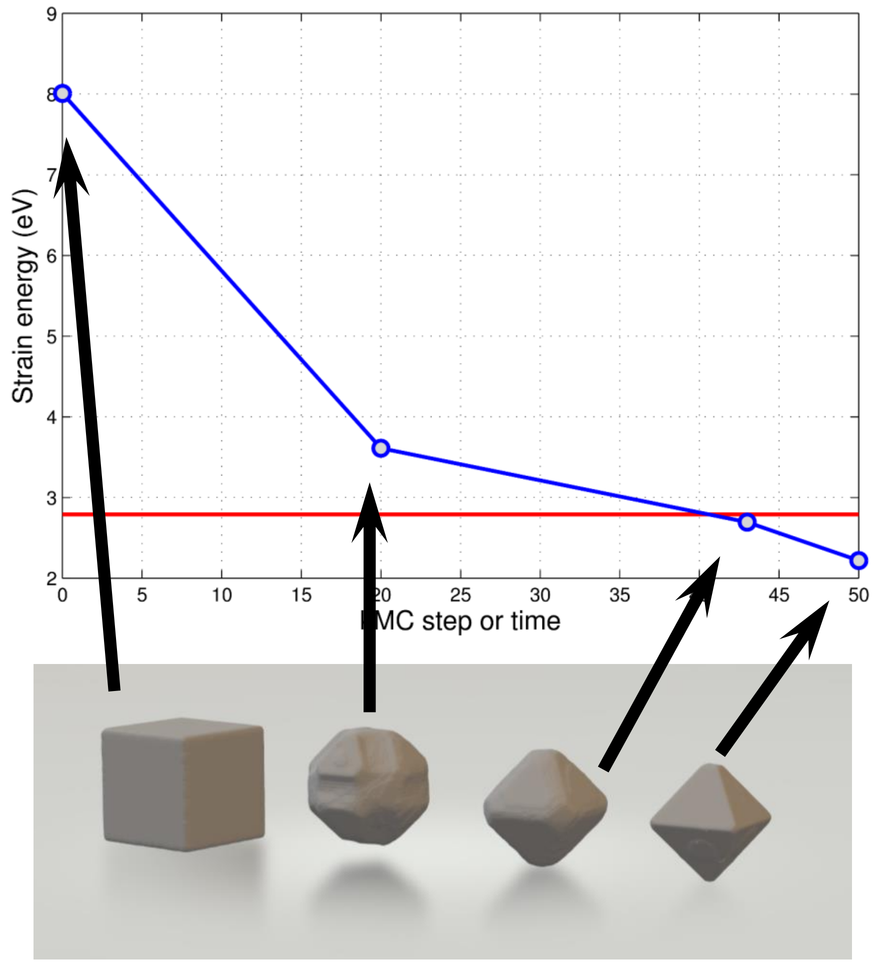
V. Zadin, et al. MODEL SIMUL MATER SC (2018)

Utilization of the surface stress model



Possible to combine TTM + Atomistic stresses + system energy minimization

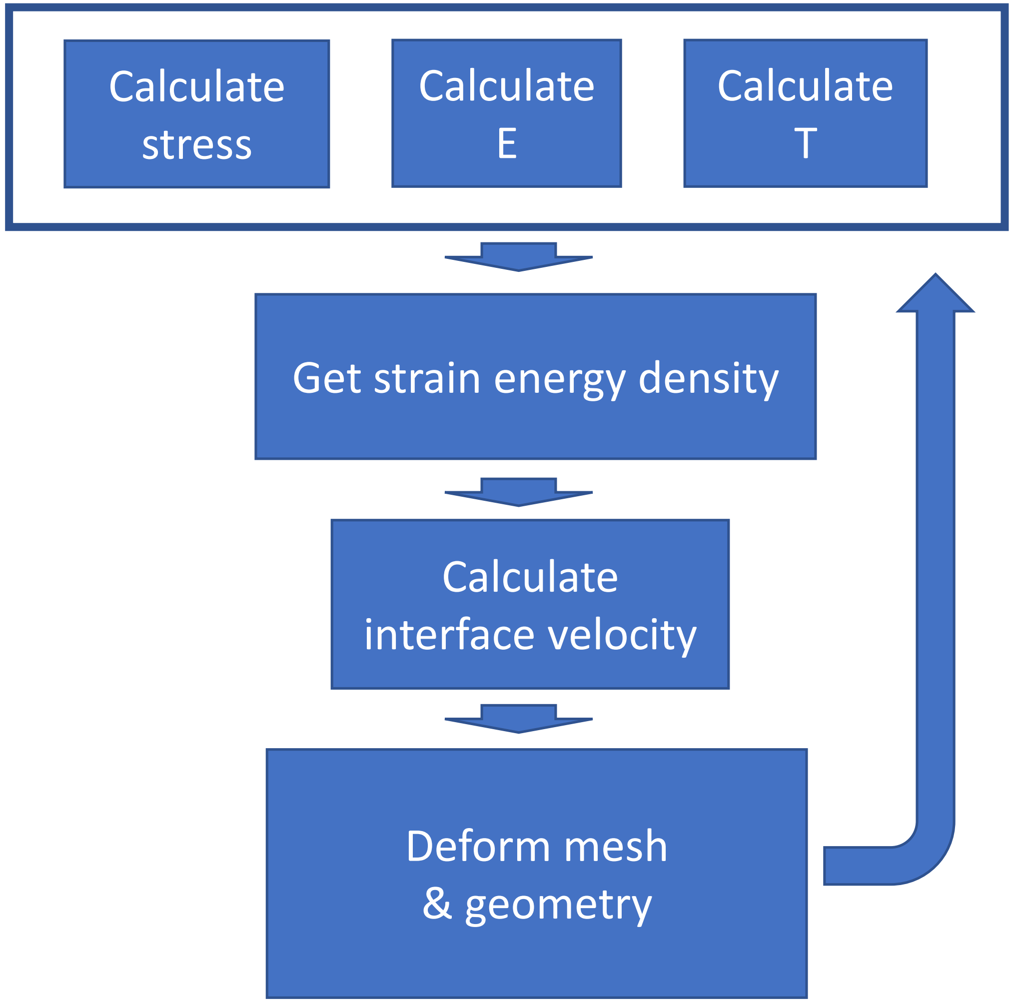
- Similar to KMC without it’s uniform temp. requirement
  - Mechanics always in steady state
    - Also in every TTM time step
- Pseudo time based optimization conducts shape changes



Different void geometries from kMC

Topology optimization

- Main principles in current implementation:
  - Minimize a functional with respect to some constraints
    - Constraints are PDE-s (stress calculations) & volume
- Based on Lagrangian formalism => the geometry is deformed
  - Strain energy density is minimized
  - Euler-Lagrange equation is used to find extrema of the functional
    - As a result gives surface velocity for geometry deformation algorithm
  - ALE type Moving mesh approach is used to deform geometry
  - Volume is constrained by using approach of Lagrange multipliers
    - LM is average surface velocity
      - In theory, volume is conserved, in practice volume conservation is differential & allows drifts
- Optimization runs using pseudo-time for iterative stepping
- Derivatives are analytically determined in background theory … **remeshing is possible !!!!!**



Core equations of the implementation

- Small strain mechanics
  - Deformation relative to current shape, not to initial
- Laplace equation for E
- Steady state heat equation
- ALE for deforming the geometry
  - Laplace mesh smoothing algorithm

The optimization problem:

$$\text{Minimize: } J = \int_{\Omega} \frac{1}{2} \epsilon^T D \epsilon d\Omega$$

$$\nabla \cdot (D\epsilon) = f$$

$$\int_{\Omega} 1 d\Omega = V^*$$

Method of Lagrange multipliers

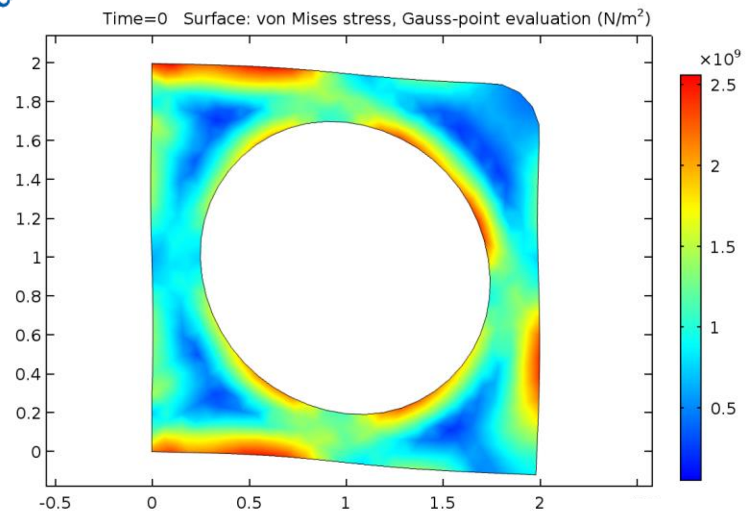
$$V_n = -\left(\frac{1}{2}\epsilon^T D\epsilon + \lambda + \gamma\kappa\right)$$

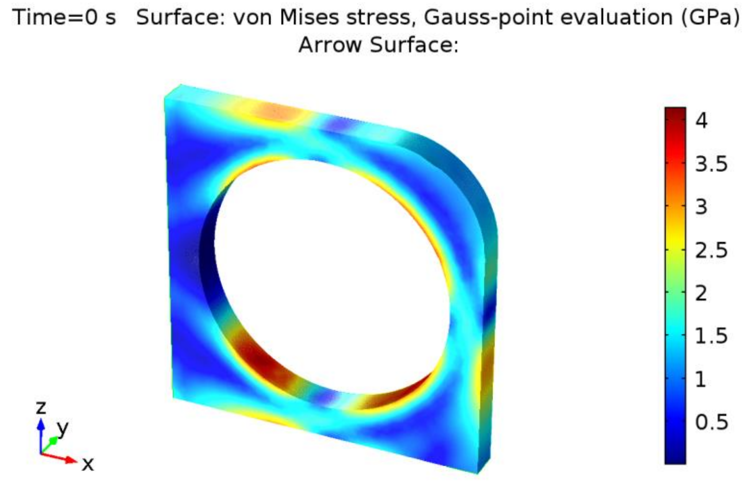
Volume change in pseudo time required to be zero:

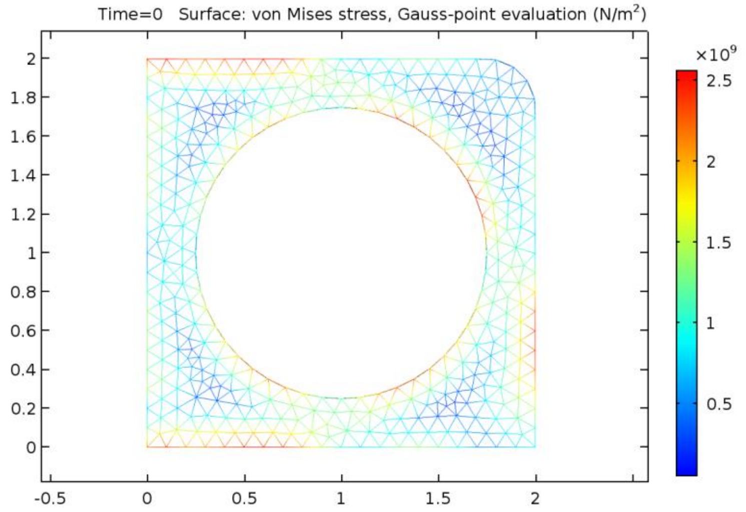
$$\lambda = -\frac{\int_{\Gamma} V_n ds}{\int_{\Gamma} 1 ds}$$

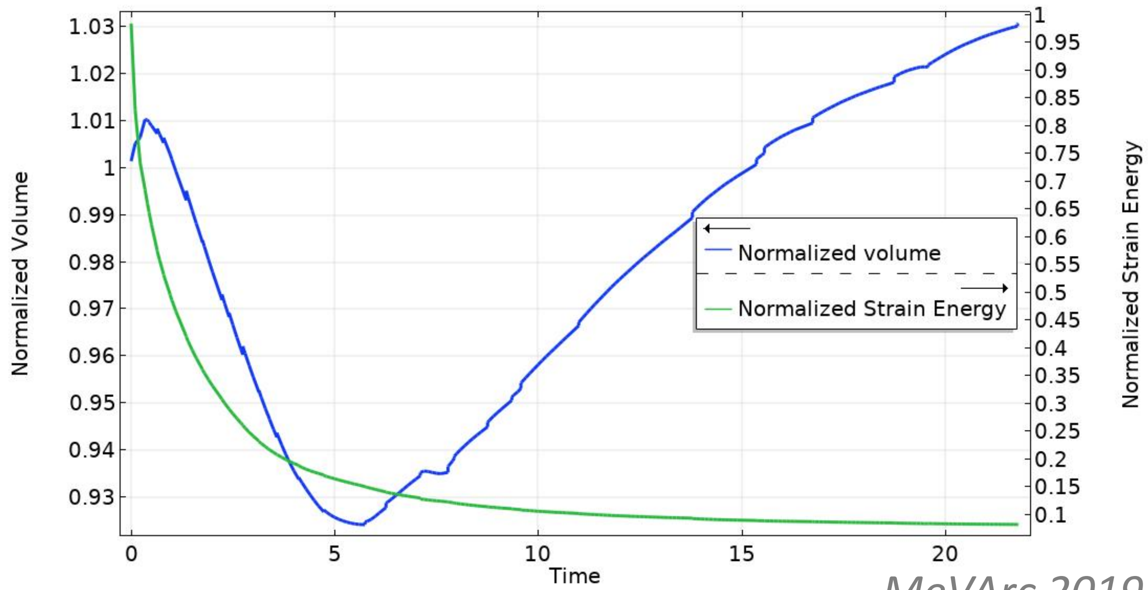
Z. Liu et al. Struct. Multidisc. Optim. (2005)
V. Zadin et. al J. Power Sources (2013)

Benchmark examples

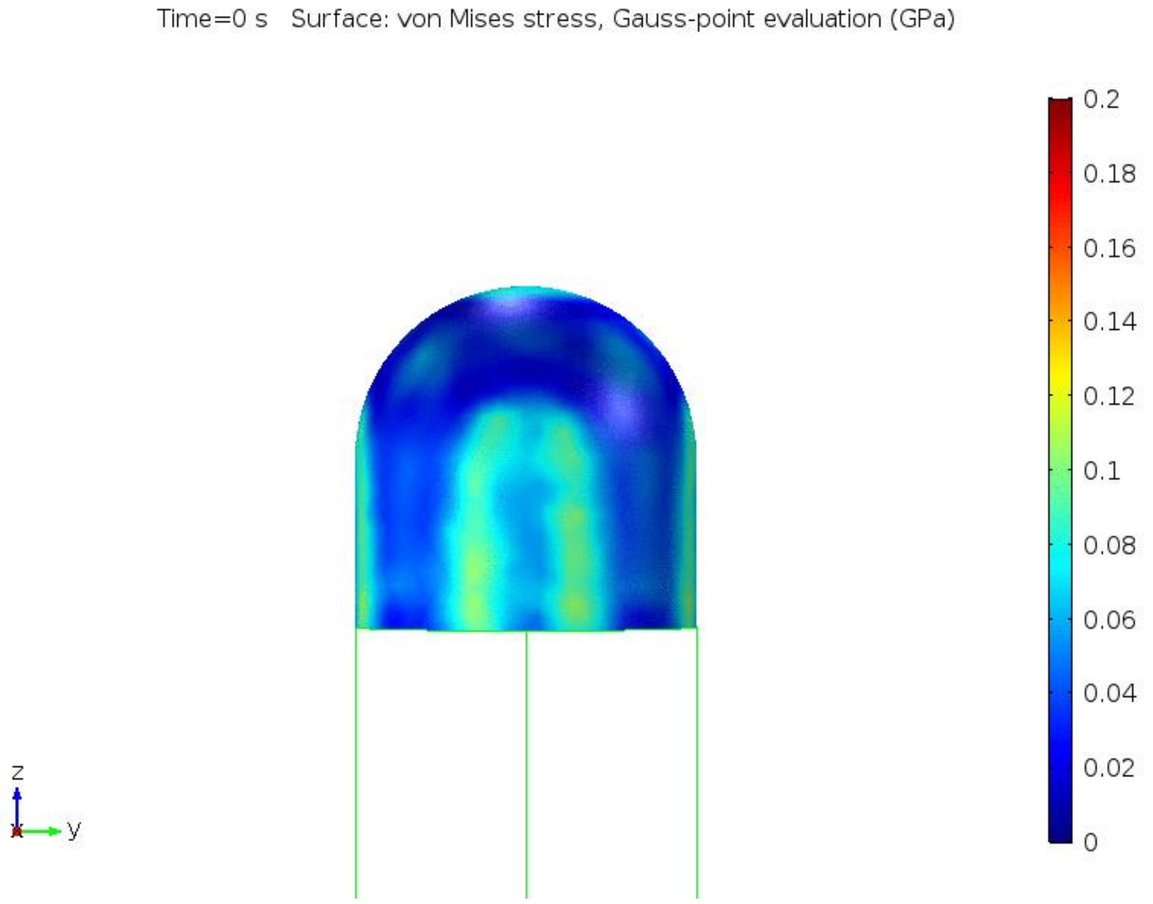


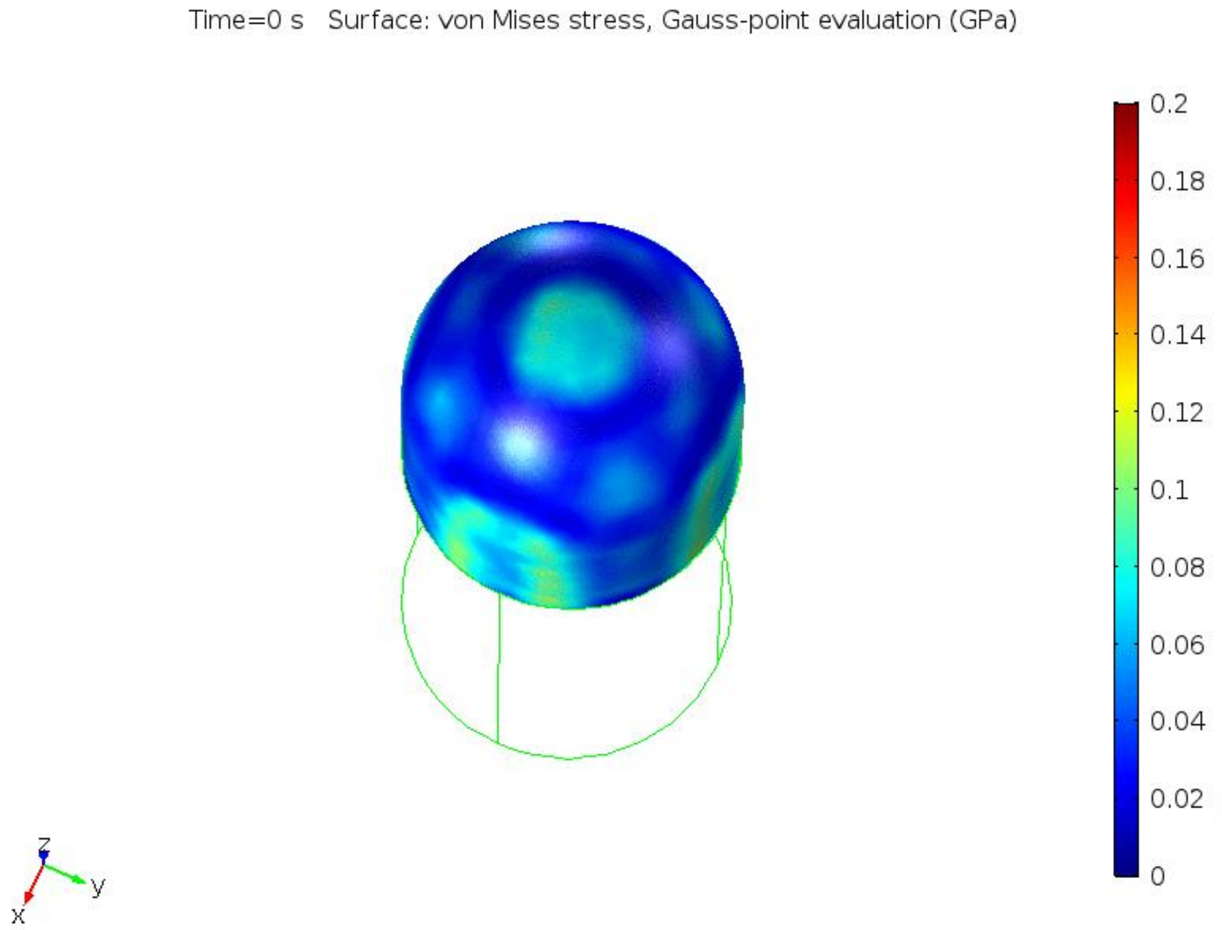




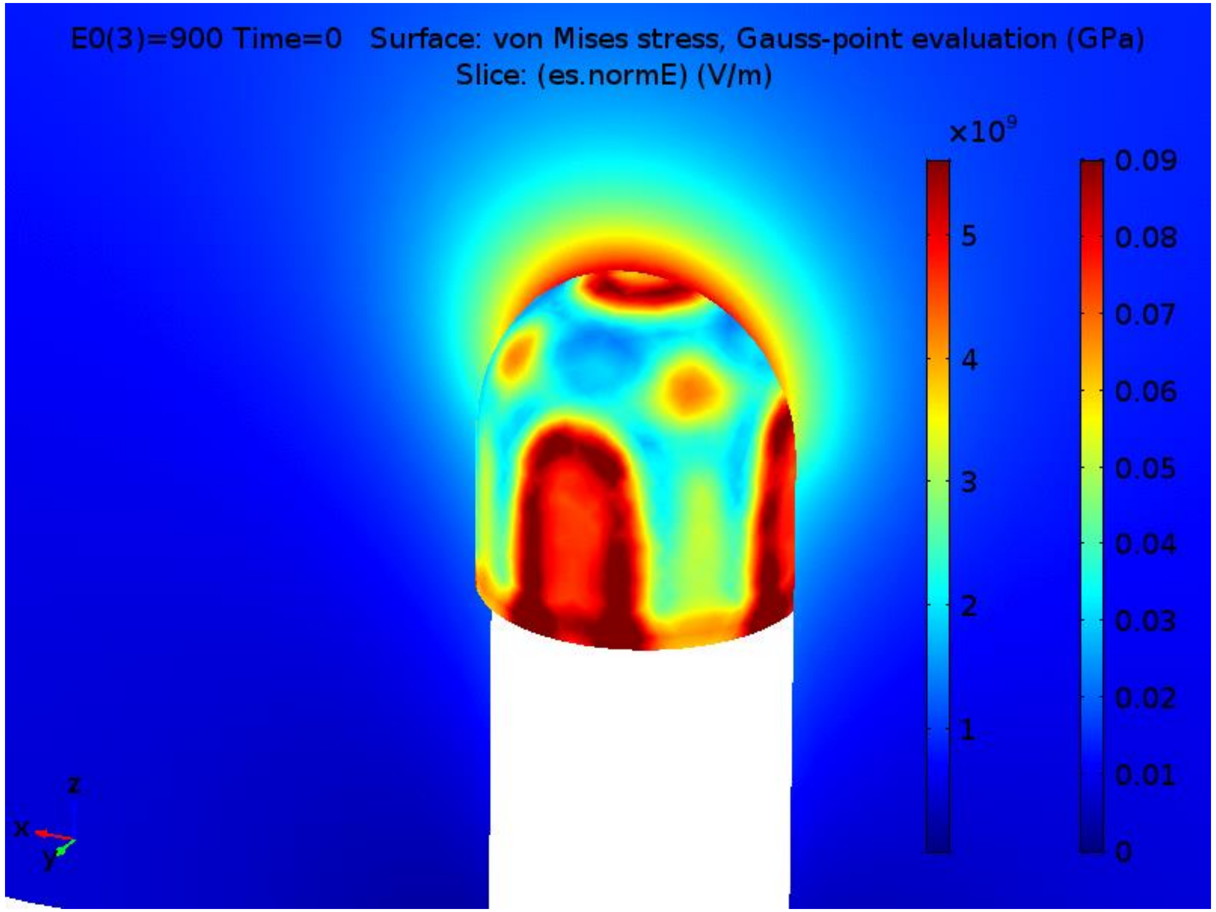


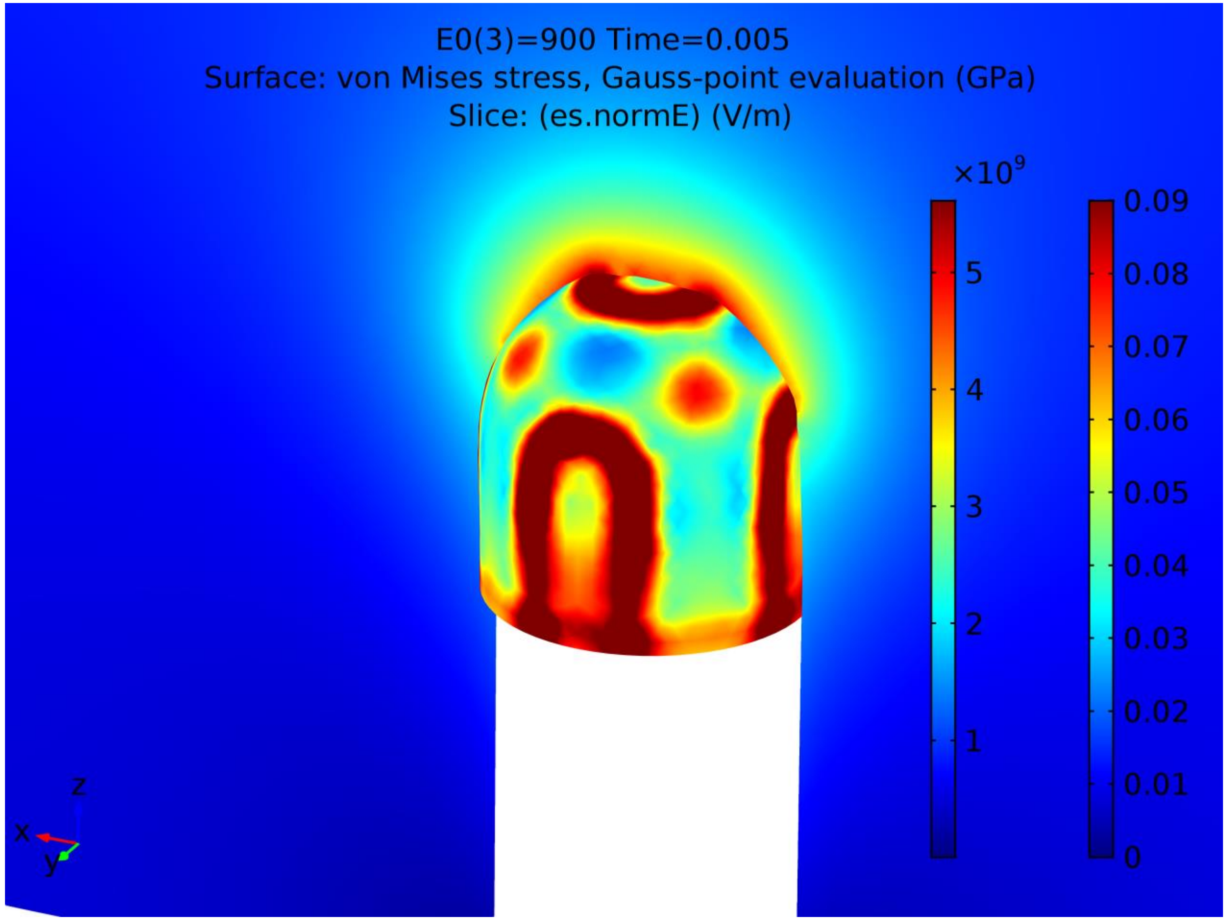
Relaxation to the pyramid in case without field



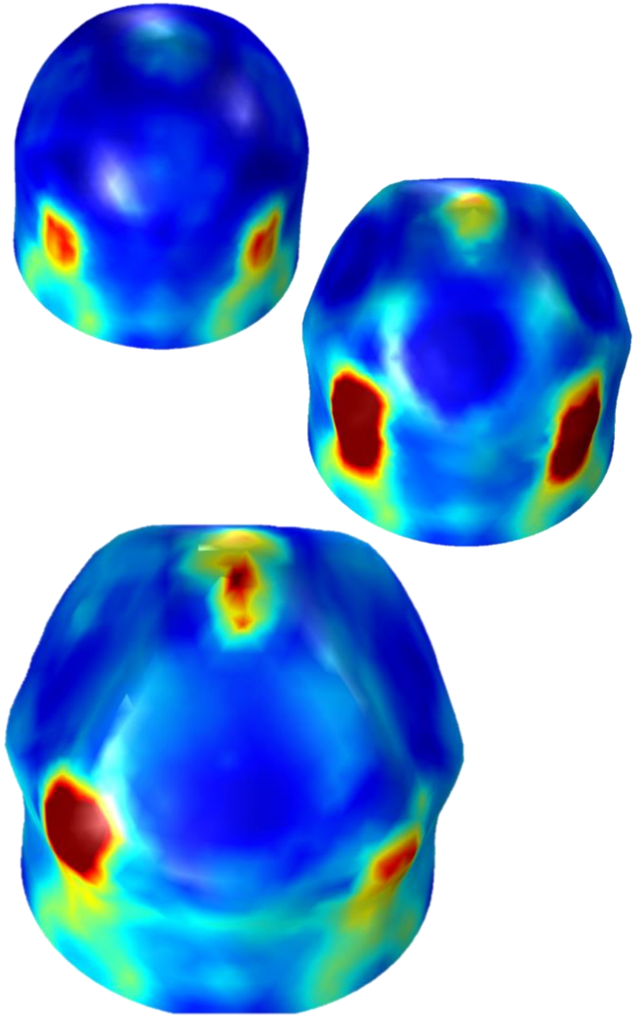


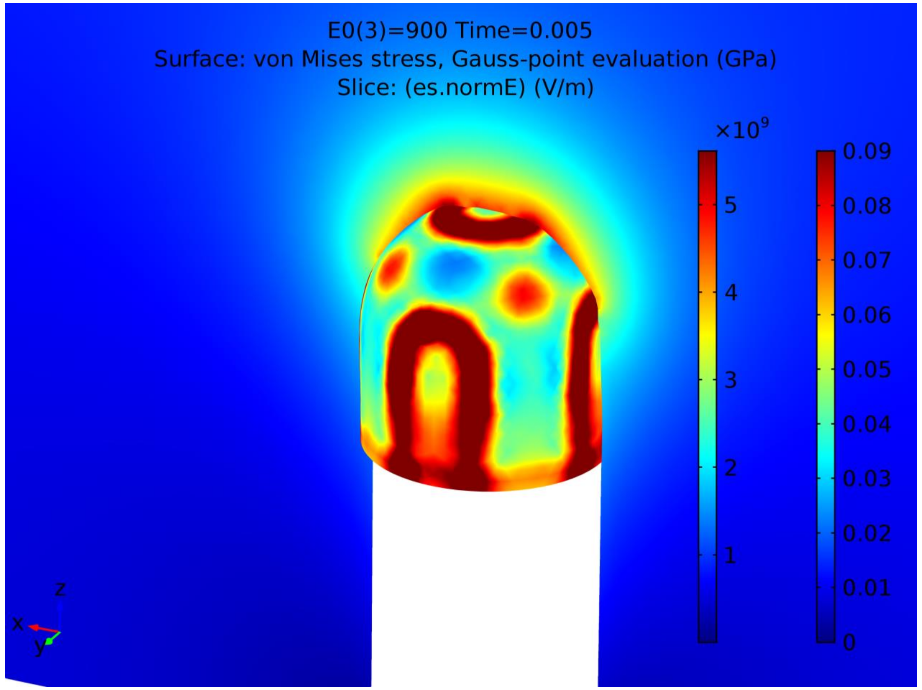
Influence of field – relaxation to faceted structure

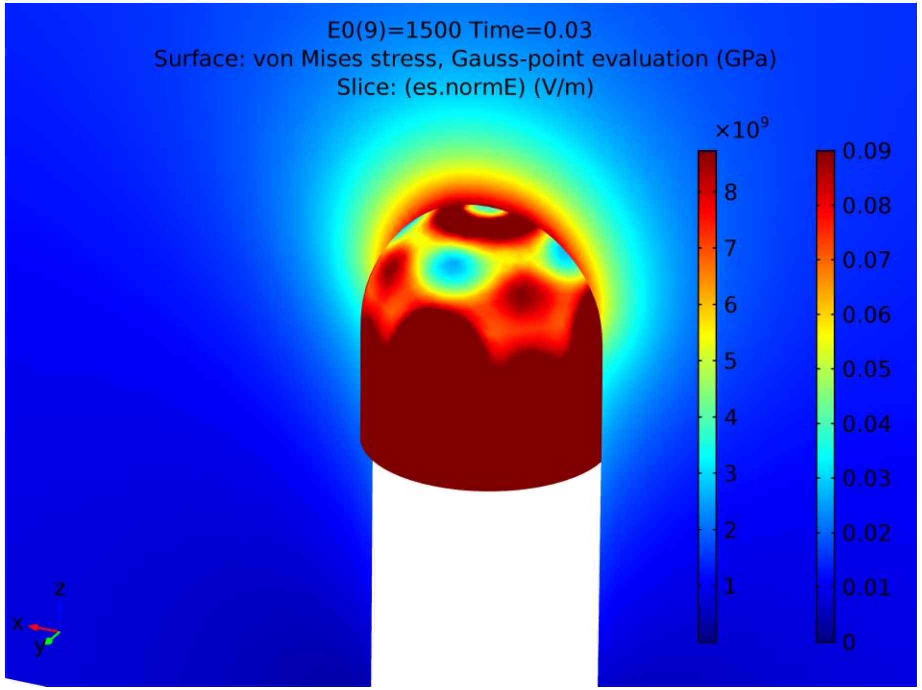




Surface evolution

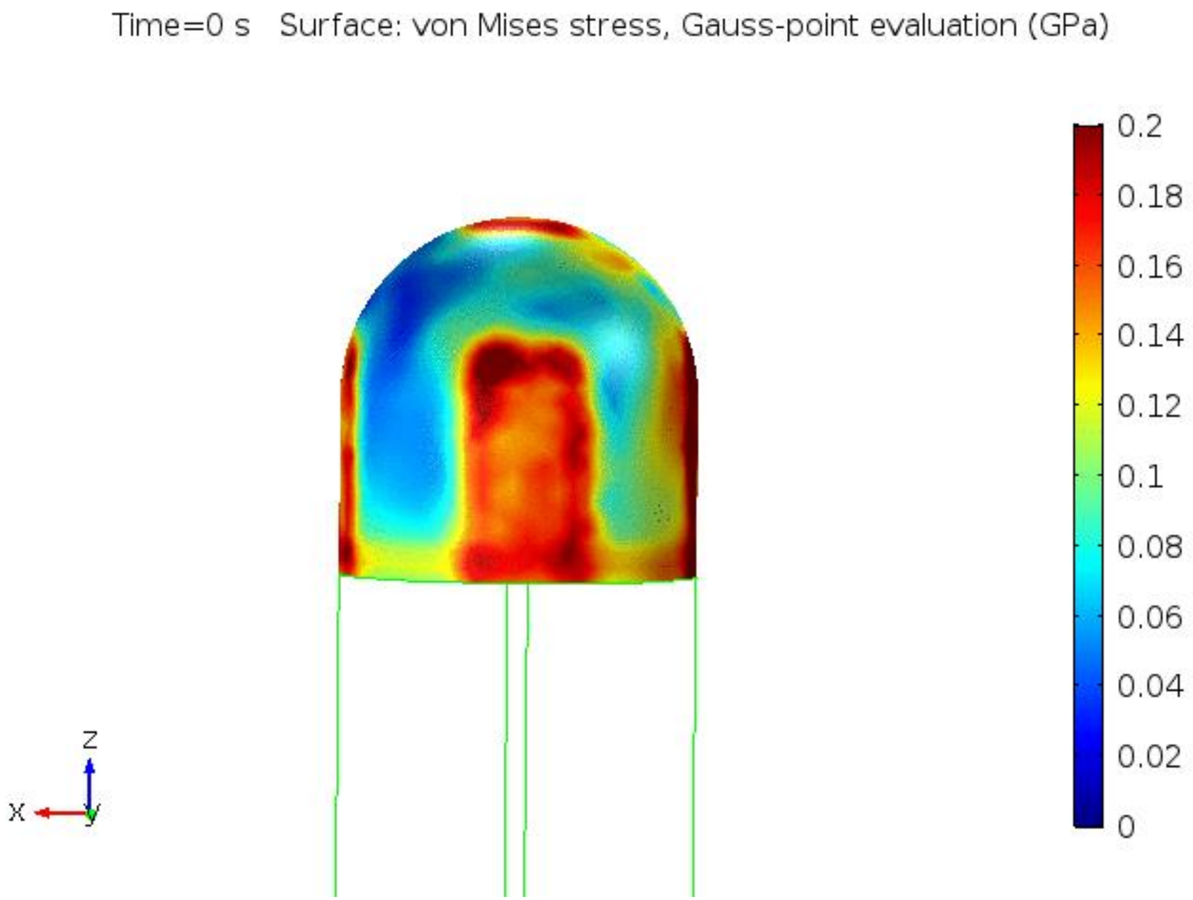




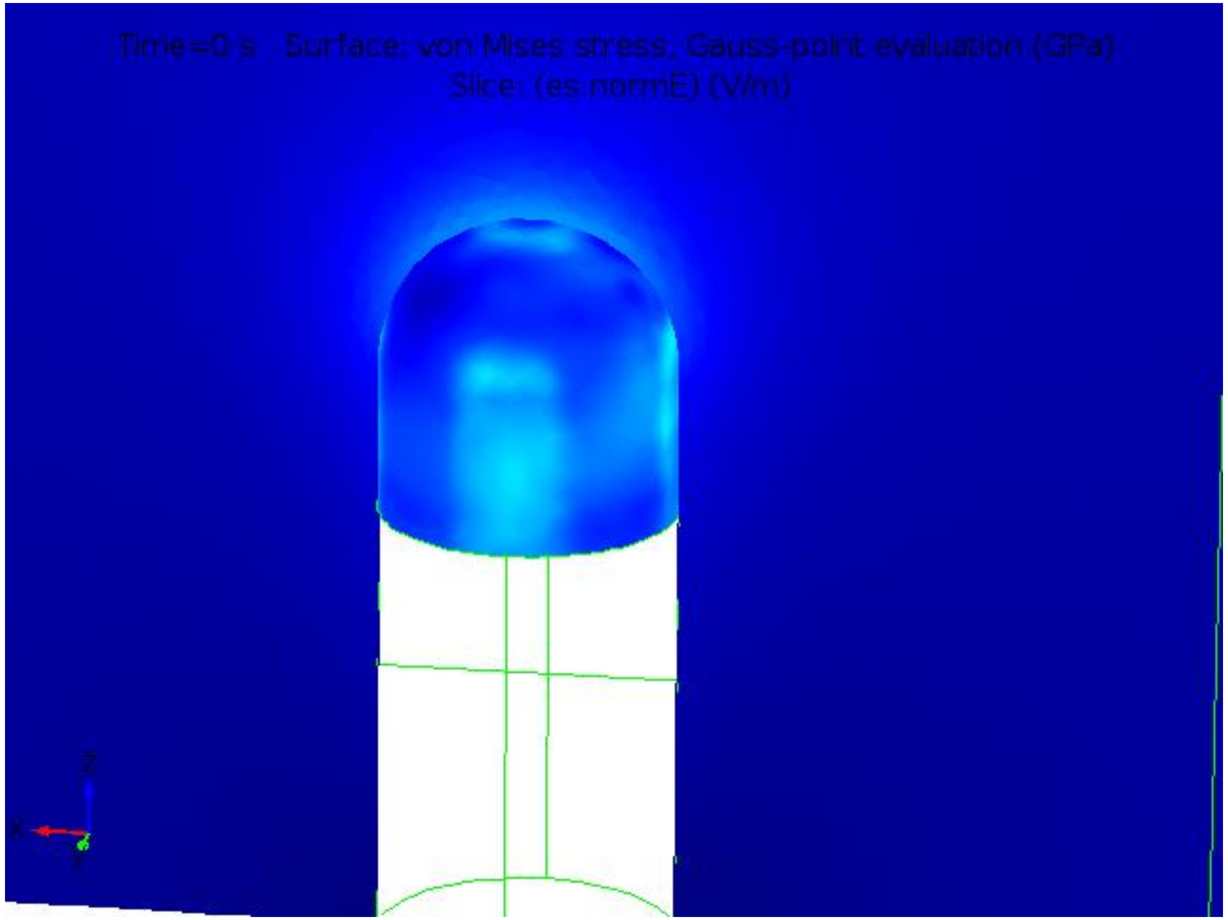


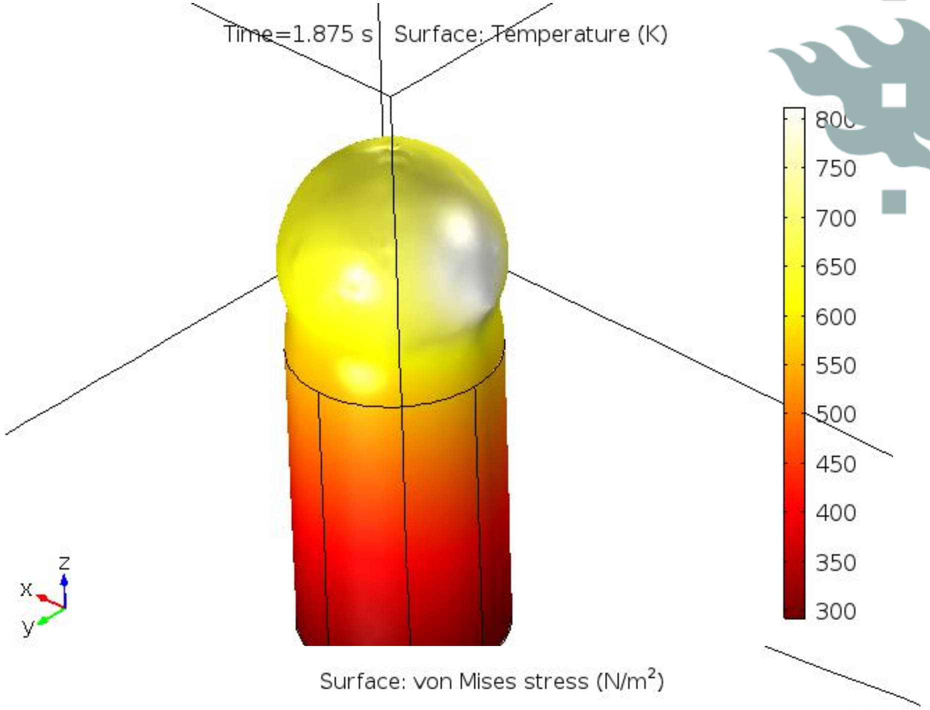
- {1 1 1} surfaces form due to energy minimization without field
  - Correspond to minimal surface energy structures
  - Good agreement with kMC
- Strong field dependence at flattening of the tip in case with the field
  - 700 MV/m still converged towards no-field case
  - 900 MV/m showed very fast flattening
  - 1200 MV/m … 1500 MV/m – shape changes were inhibited

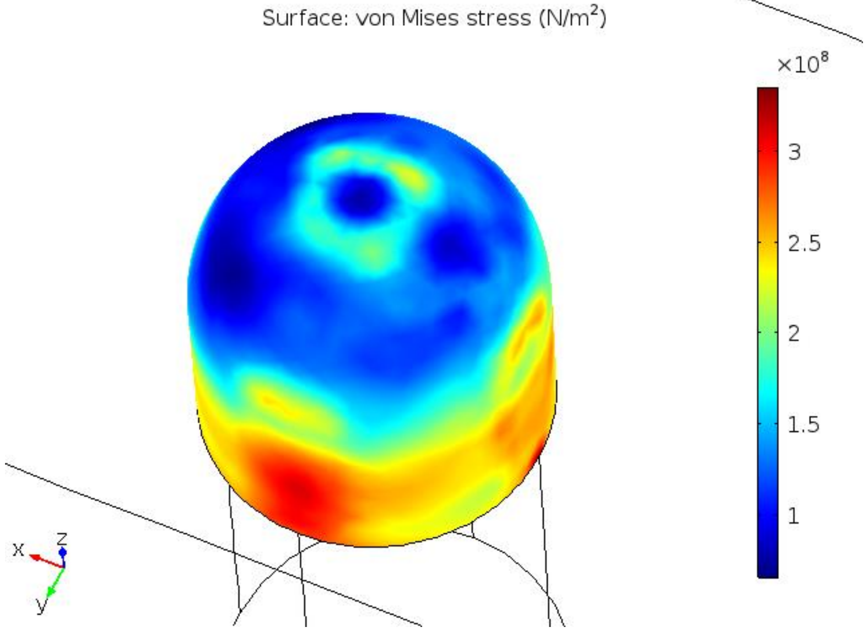
Influence of the temperature – no field



Combined effects

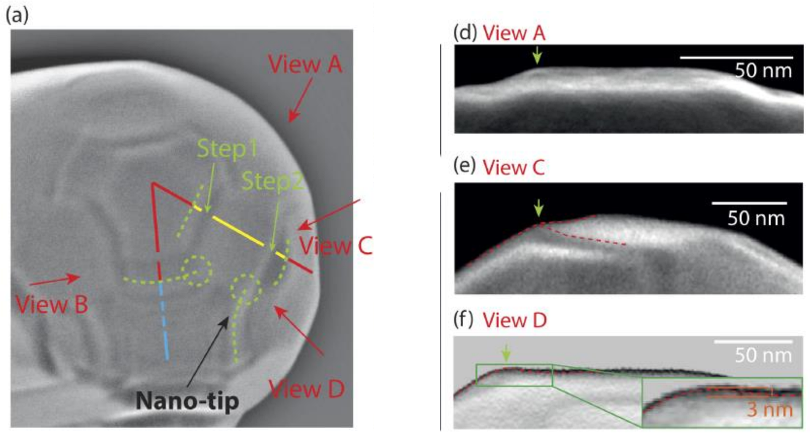


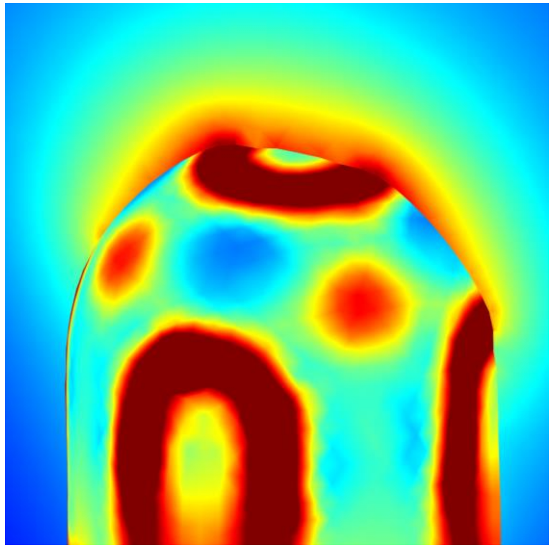




Conclusions

- Polycrystalline tip – thermal stress due to grain misalignment?
- Surface layer melting?
  - Very fast convergence in case of **E** only
  - **T** influence very strong
    - Over estimation of thermal conductivity? Too small tip?
    - Possible tests with artificially reduced thermal conductivity
    - Possible test with surface layer having lower thermal conductivity
- Possible Cu effect (FCC crystal behavior)
- Possible fatigue effect – cycled loading
  - E reduces surface stress effects …. T tends to behave uniformly …..
    - No field, no T may be more correct representation of T effects
    - Perhaps optimization with T tries to converge to spherical shape
    - E and no E interplay provides ridges
      - T acts only as means for speeding up surface diffusion?
- Different behavior in all cases – without field, with field & with heating
  - Interplay of combination of phenomena will lead to selective faceting of the surface
  - Balancing between stresses by electric field, surface and thermal effects are needed





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