Atomistic modeling of surface evolution under strong electric fields

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NNSA
National Nuclear Security Administration
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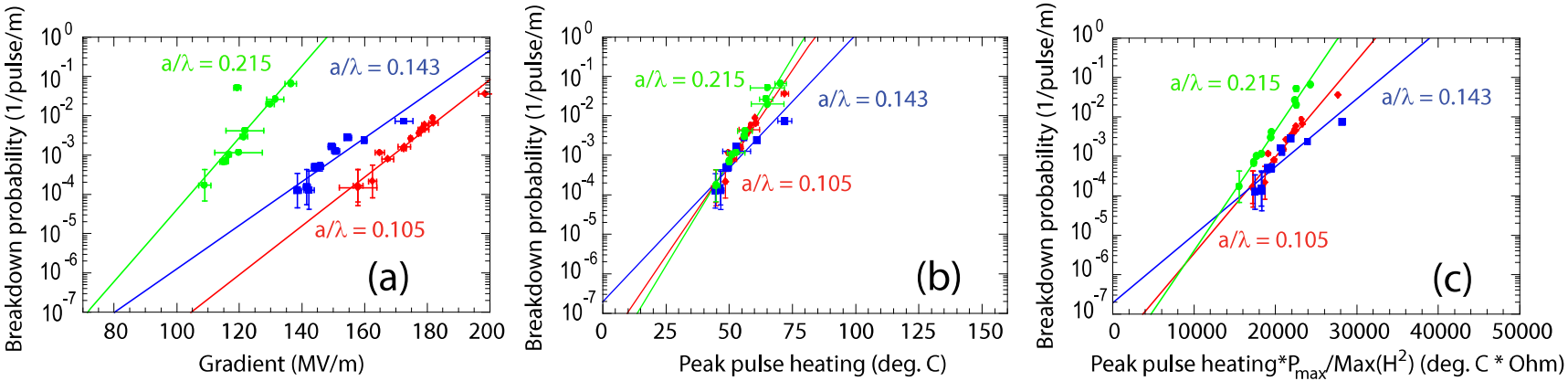
Motivating challenges

- Compact, low-cost, accelerators would benefit from high gradients
- These however lead to increase in RF breakdown
- Incidence of breakdown is well characterized, but microscopic causes are complex:
  - Occurs at fields that are well below what needed of a clean flat Cu surface (~10 GV/m)
  - Implies the formation of precursors that locally enhance the field before breakdown can occur

Key questions

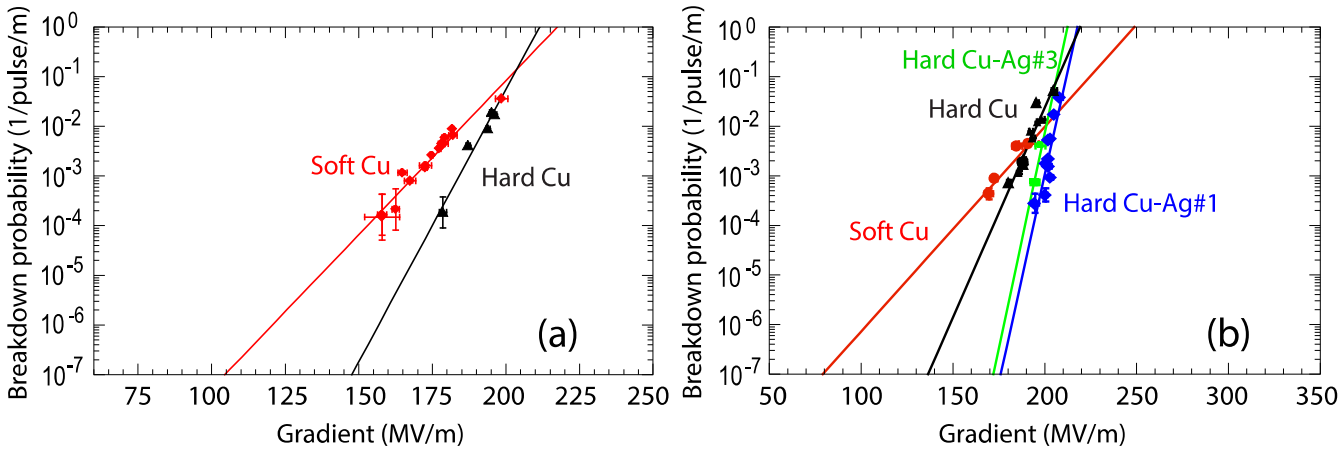
- **What are these precursors and how they form?**
- **What controls their formation rate?**



Dolgashev, Tantawi, Higashi, Spataro, 2010

Key questions

- **How does precursor formation couple with microstructure and composition?**



Simakov, Dolgashev, Tantawi, 2018

Goals

- **Short-term goals:**
  - **Demonstrate that the response of metal surfaces to external E can be … quantum calcula…**
- Long(er)-…
  - Include …
  - Underst… microstr…
  - Inform the design of optimized material solutions

The goal is not to reach engineering scales, but to understand fundamental breakdown mechanisms and inform materials design

Levels of theory

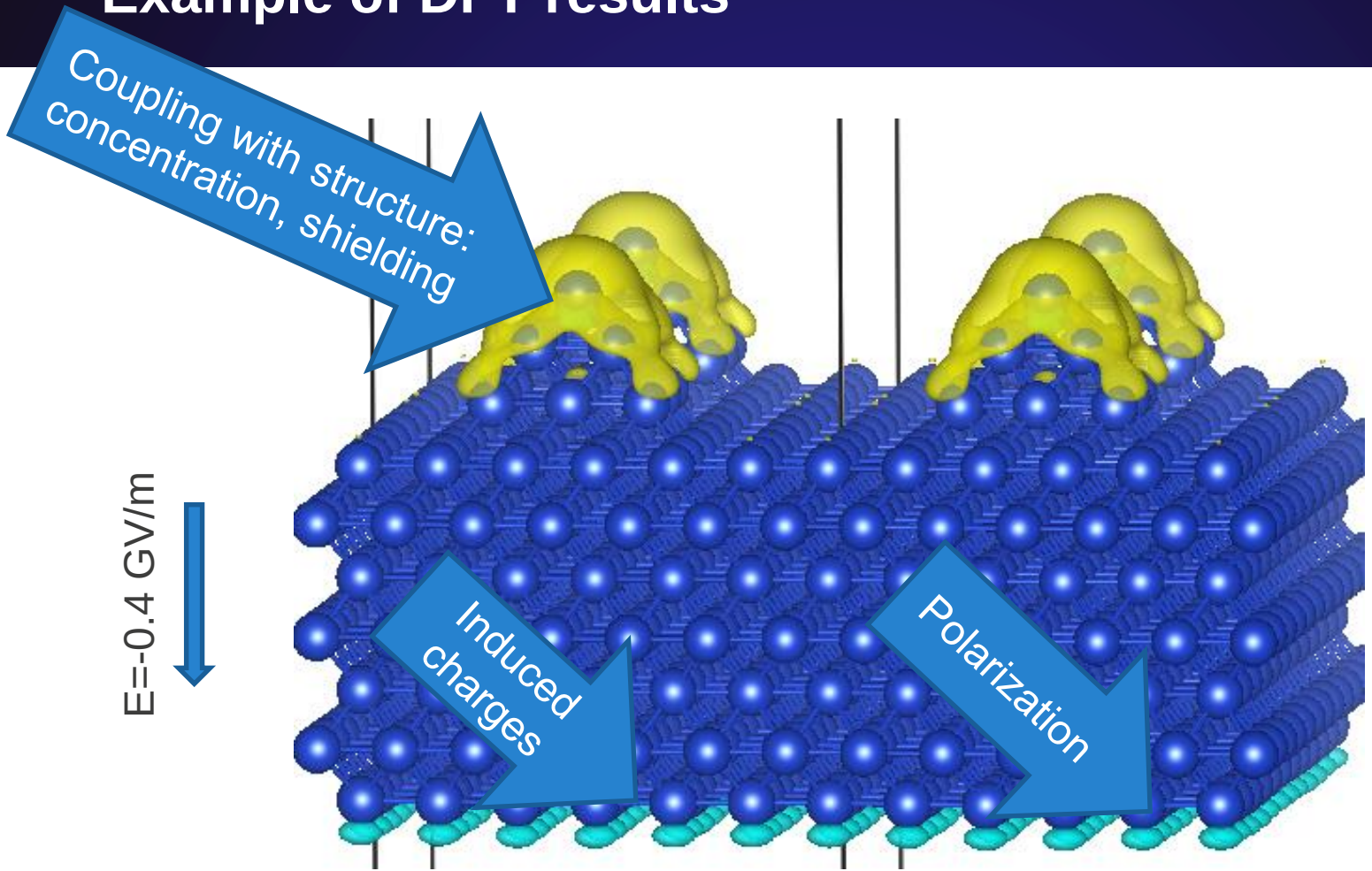
Quantum description:

- Density functional theory (DFT)
- Explicit description of the electron gas

- Strengths:
  - Very accurate (~100 meV/atom)
  - First principles (few parameters, transferable)

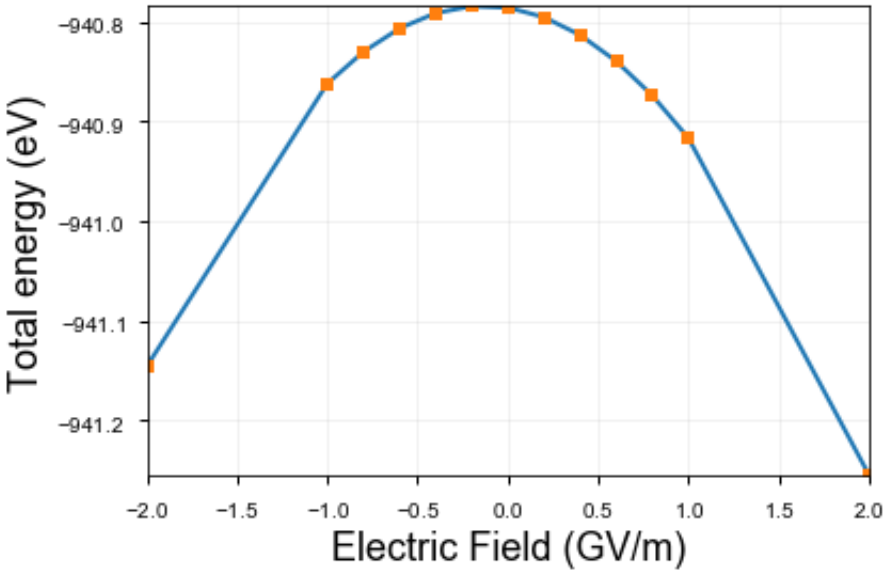
- Weakness:
  - Very expensive (scales as $N_{electrons}^{3}$)
    - Small systems (~few 100 atoms): cannot capture microstructure effects
    - Static or short dynamic simulations (~ps): cannot explicitly simulate surface evolution

Example of DFT results

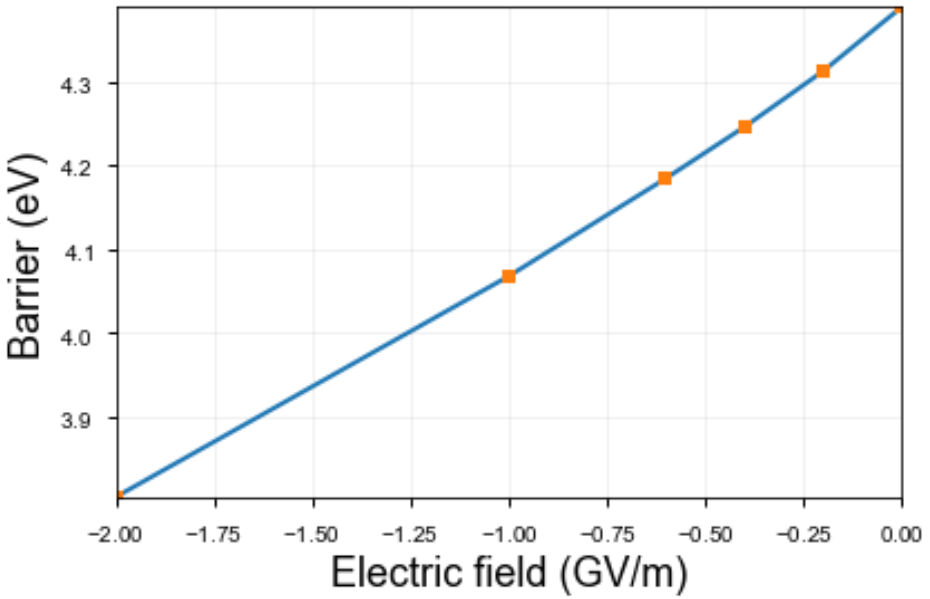


Example of DFT results

Energetics



Electronics



Levels of theory

Classical description:

- Molecular dynamics (integration of classical EOM)

- Strengths:
  - Reasonably accurate empirical description of Cu
  - Relatively fast
    - “Large” systems (>10^6 atoms): can capture microstructural effects
    - “Long” simulations (milliseconds): can capture surface evolution

- Weakness:
  - Scales are very limited compared to engineering scales
  - **How to include field effects?**

Classical MD

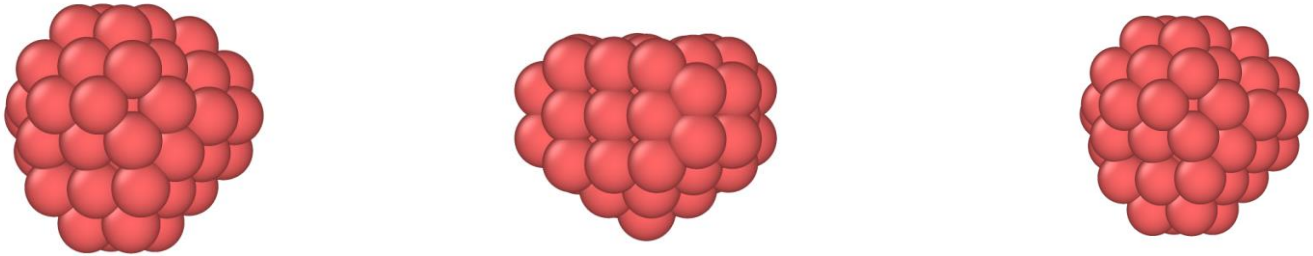
In conventional MD, charges are typically implicit or fixed. In order to capture field effects, we need a **charge equilibration model.**

- Procedure:
  - Choose an empirical Hamiltonian for the electronic energy
  - Parameterize the Hamiltonian by fitting to quantum data
  - Assign charges so as to minimize the electronic Hamiltonian at the instantaneous position of the atoms

Training set

- Bulk atoms are non-informative of fields response. Want as much surface atoms as possible for quantum calculations

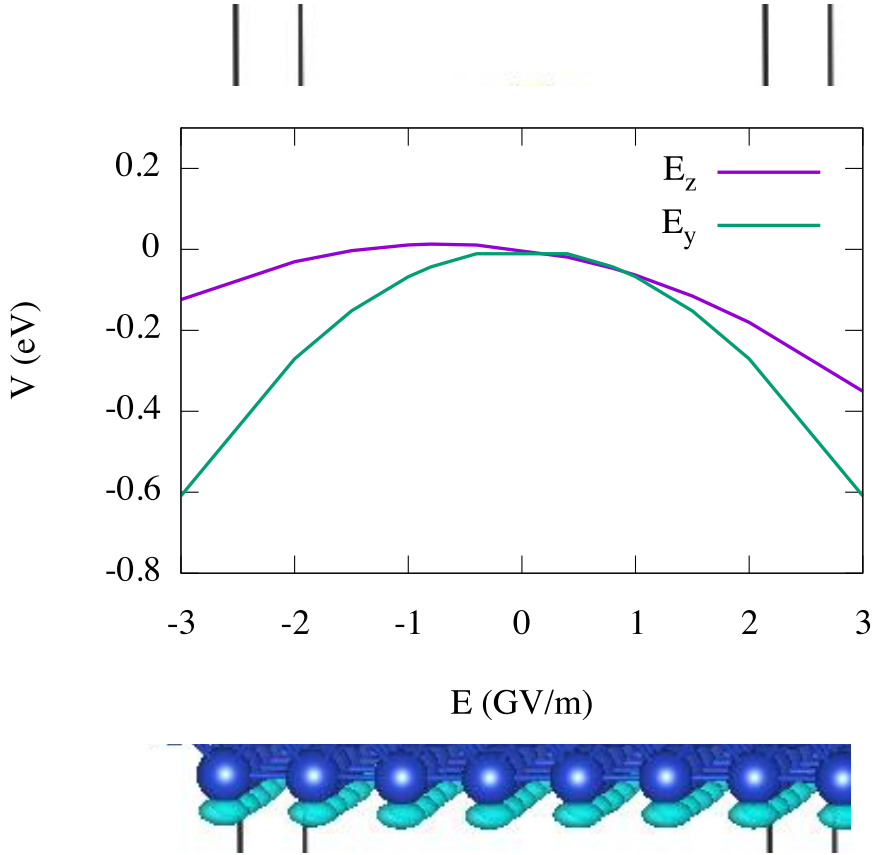
- Characterized 6 Cu small nanoclusters (30-68 atoms) for fields varying between -3 and 3 GV/m using DFT



- Fitted model on energies at highest fields (42 data points), the rest (174 data points) is used as validation

Electronic Hamiltonian

- 3 ingredients are essential:
  - Hardness: onsite charge repulsion
  - Polarizability: excess charges induced in vacuum, not on atoms
  - Intrinsic dipole: breaks the E,-E symmetry.

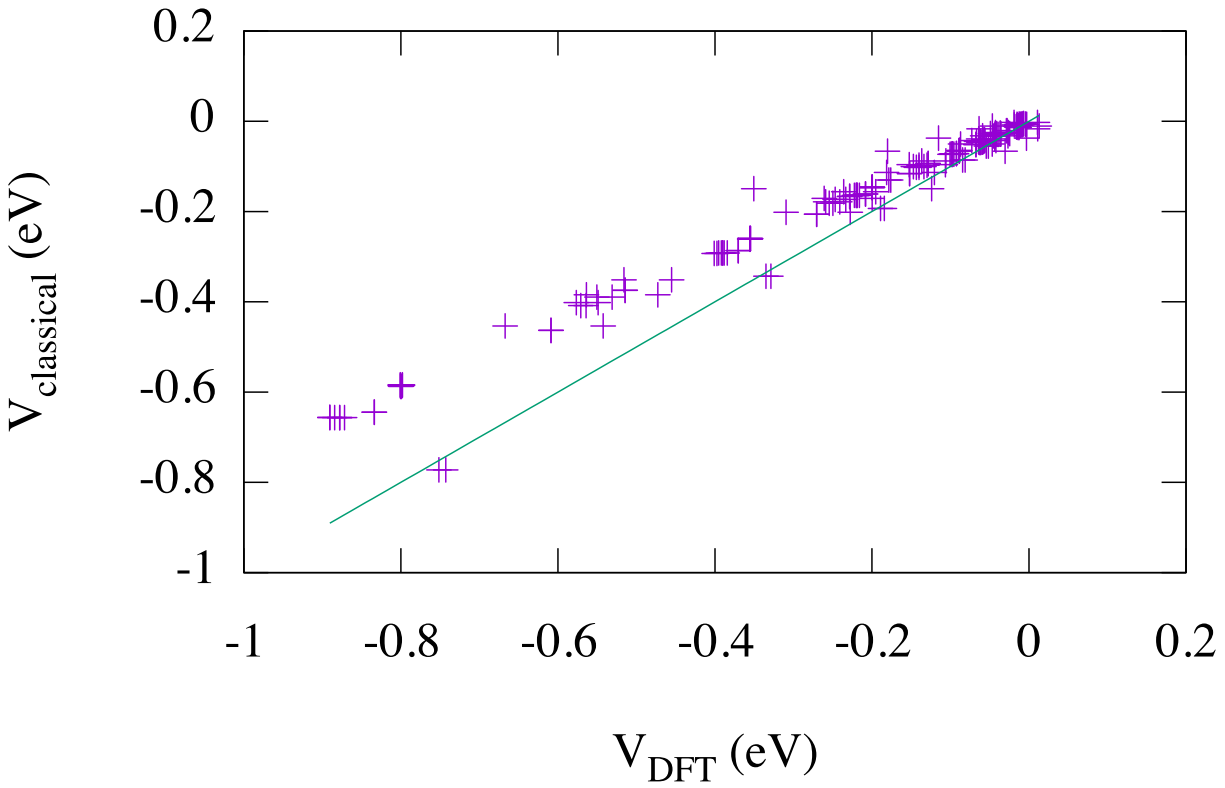


Electronic Hamiltonian

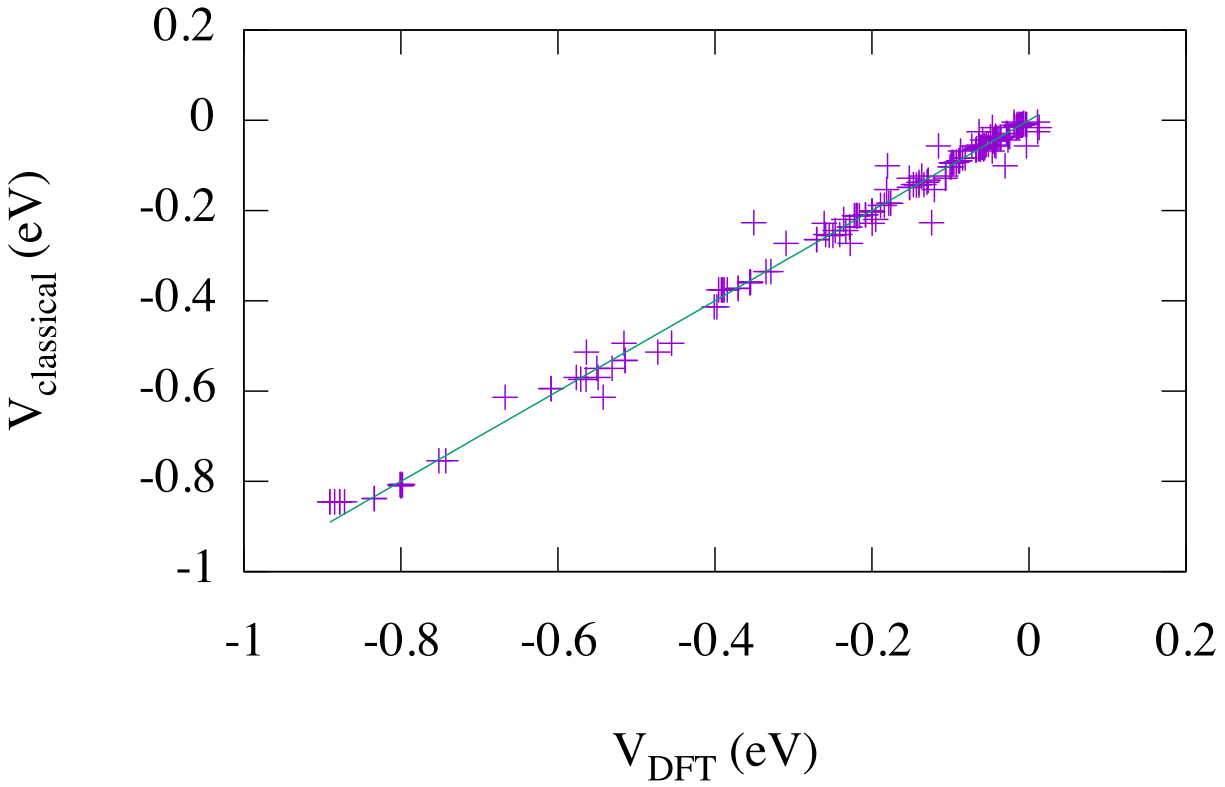
- $V = V_{Coulomb} + V_{Field} + V_{Self}$
- $V_{Self} = \sum_{i=1}^{N} b_i q_i + c q_i^2$ (c penalizes charge accumulation on atoms)
- $b_i = b\ N_{Neighbors,i}$ (electronegativity depends on atomic environment)
- Virtual sites for excess charge located 1A away from the atoms, pointing away from the neighbors.

- **Only 2 adjustable parameters** (b,c)

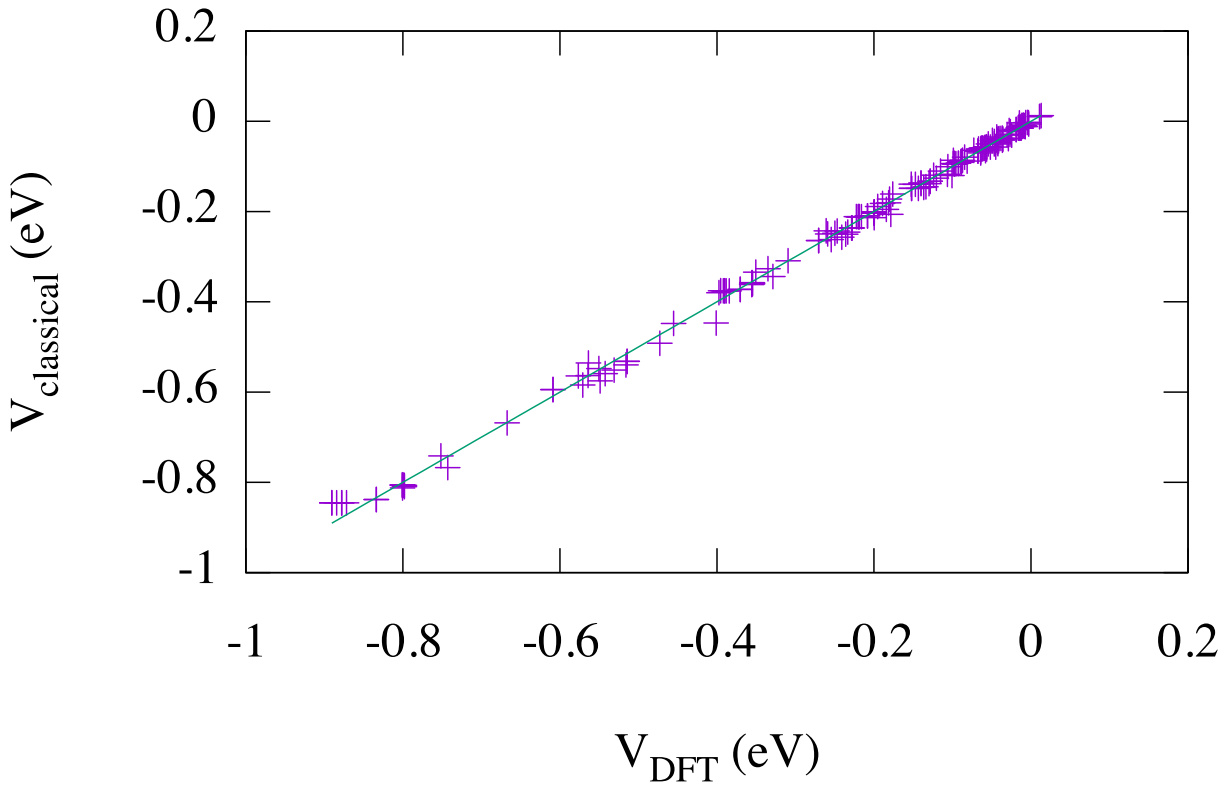
Validation: hardness



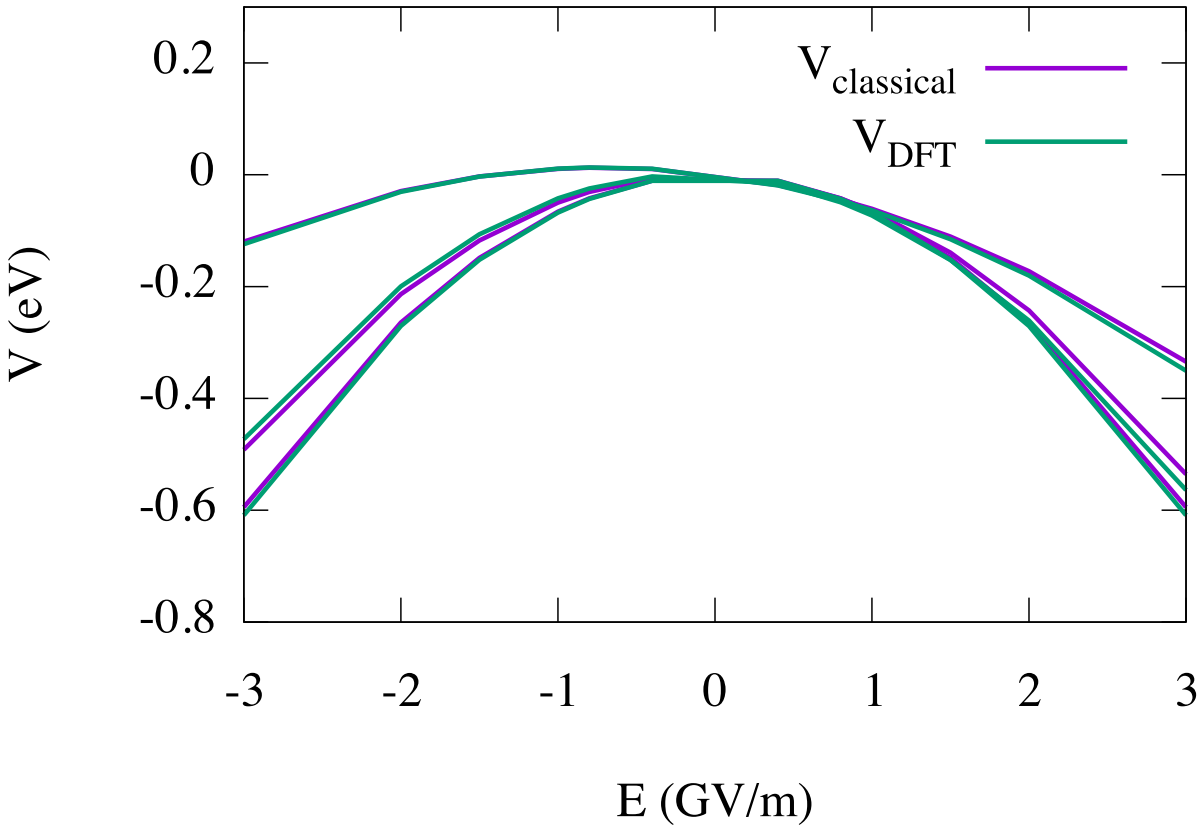
Validation: hardness, polarizability



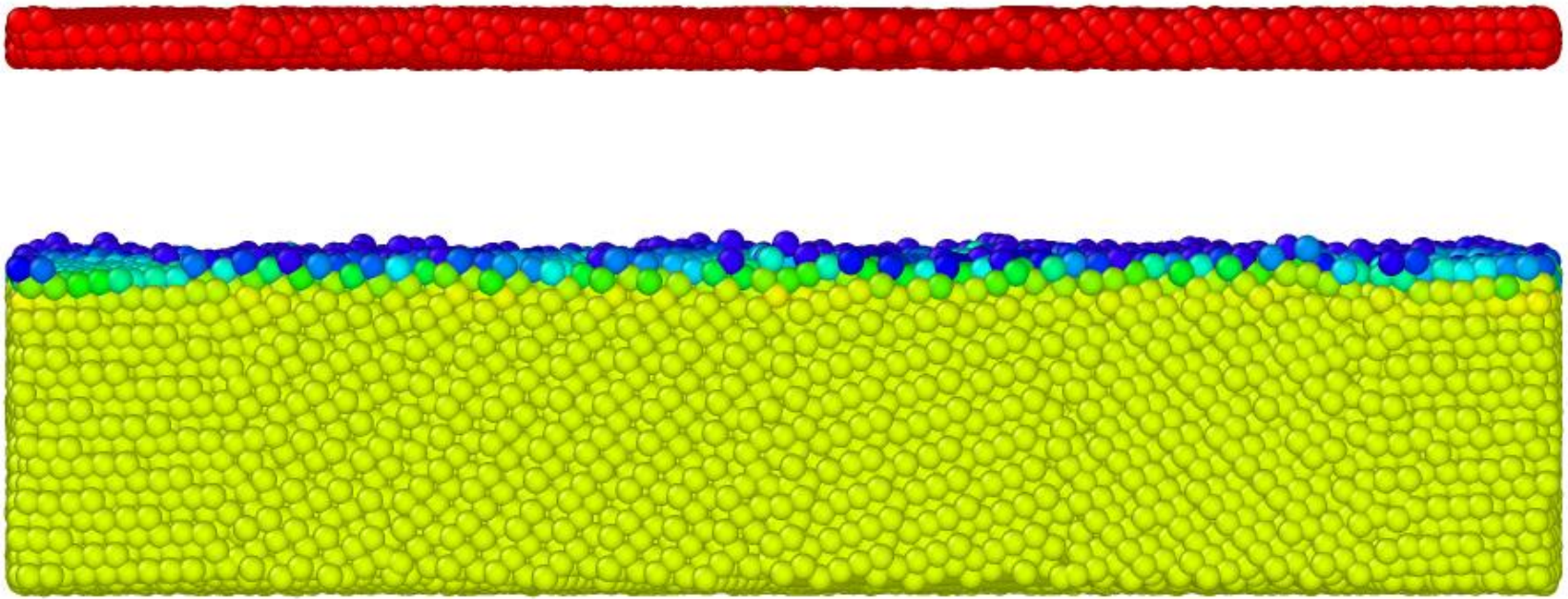
Validation: hardness, polarizability, intrinsic dipoles



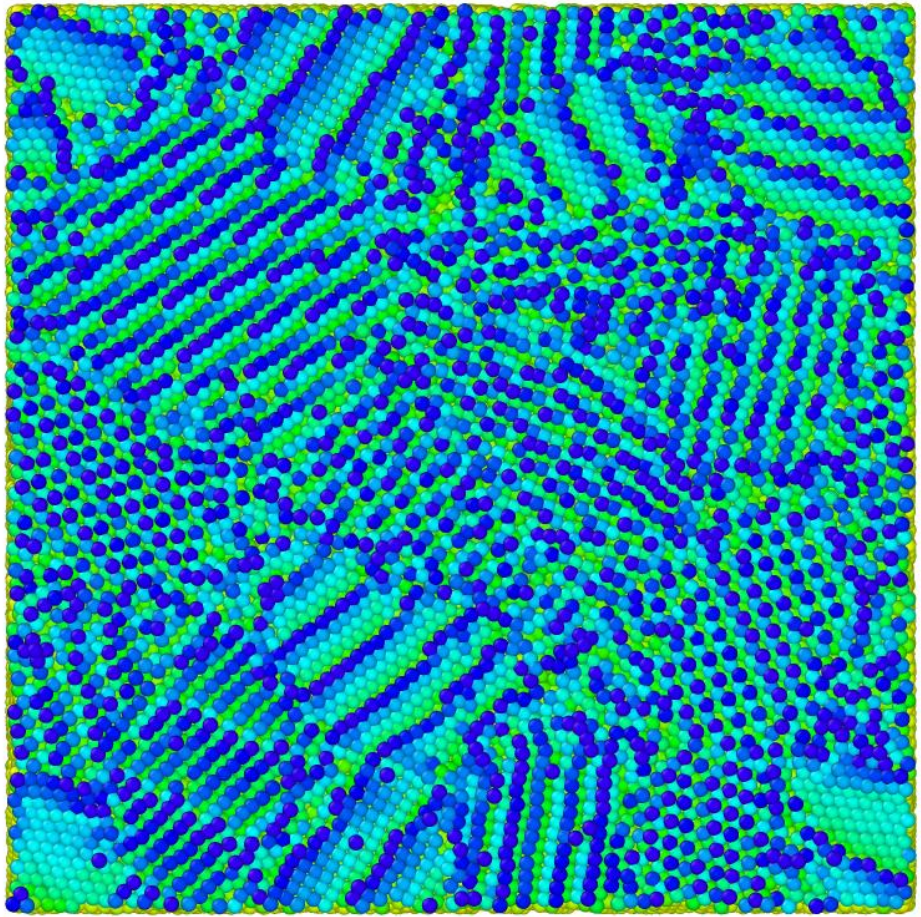
Intrinsic dipole



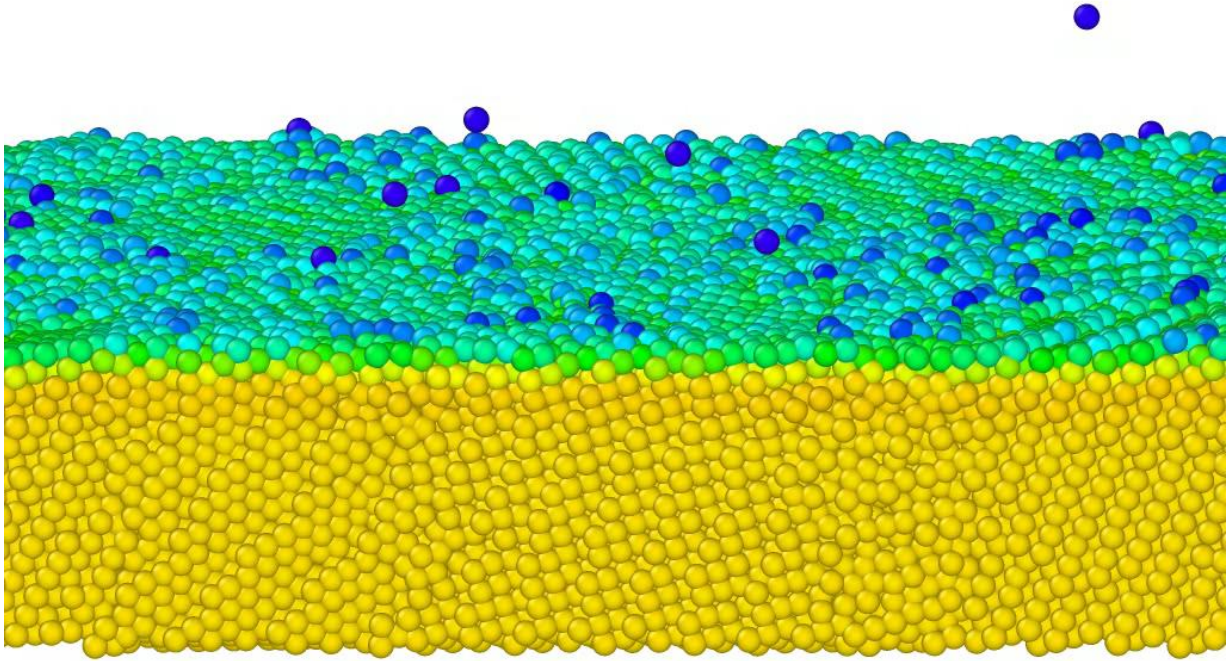
Large-scale MD



Large-scale MD



Large-scale MD



Large-scale MD

We expect a good description of the formation of precursors, but not of the breakdown process itself

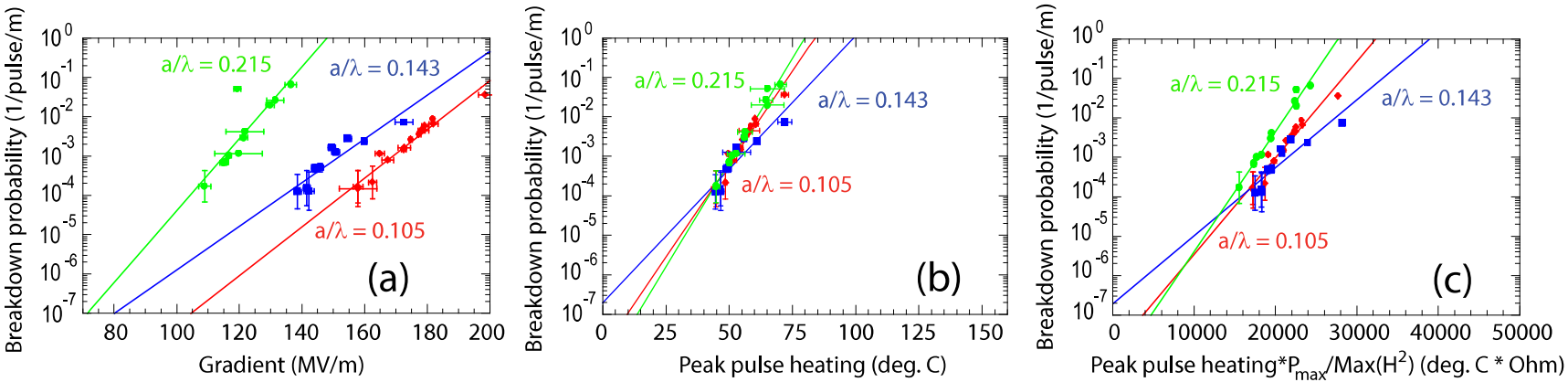
Large-scale MD

- At very high fields (~10 GV/m), nano-pyramids spontaneously form, leading to local field enhancement and evaporation/field emission
- Can this mechanism spontaneously create breakdown precursors? DC models suggest that growth should occur on >h timescales at ~100 MV/m. **>1000h** in pulsed conditions.

- **Might play a role, but not the whole story,** as breakdown affected by bulk mechanical properties
- **This mechanism can amplify short wavelength structures formed by some other mechanism**

Future work: nature of the precursors

- Clear indications that magnetic fields are crucial
- Pulse heating is known to be a factor



Dolgashev, Tantawi, Higashi, Spataro, 2010

- Will explore microstructural changes induced by thermal fatigue and their coupling with E and with the microstructure

Conclusion

- Classical models parameterized from quantum calculations can describe the energetics of metals under high fields
- In critical conditions, we see the spontaneous formation of nano-pyramids and the emission of atoms (i.e., “breakdown”)
- Still have to simulate the growth of the precursors
- Atomistic simulations face a challenge of scales, but can provide unique insights

- **Suggestions are welcome!**