Machine learning for Cu surface kinetic Monte Carlo

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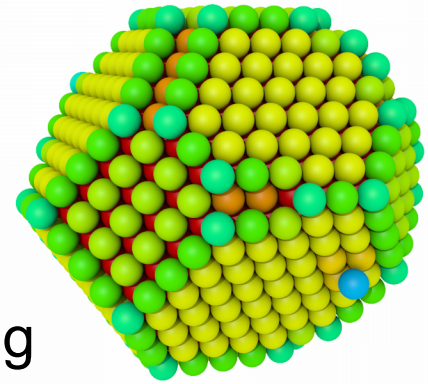
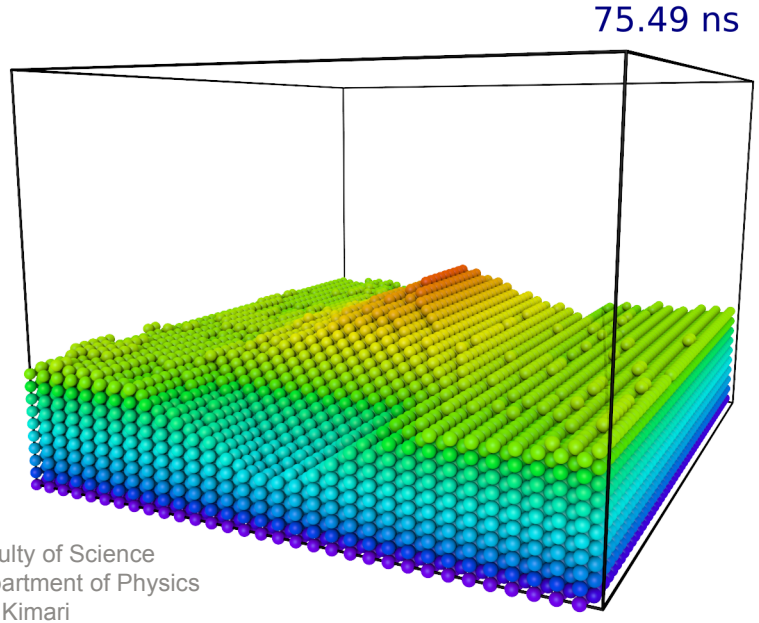
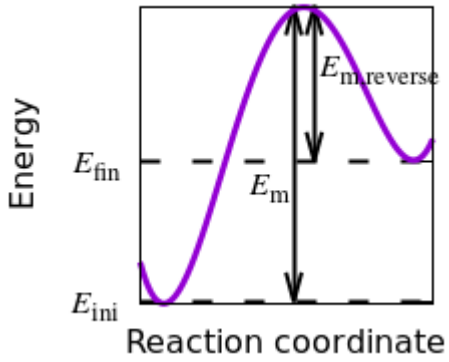
Outline



- Motivation
- Kinetic Monte Carlo + machine learning
- Machine learning accuracy
- Simulation results
- Conclusions

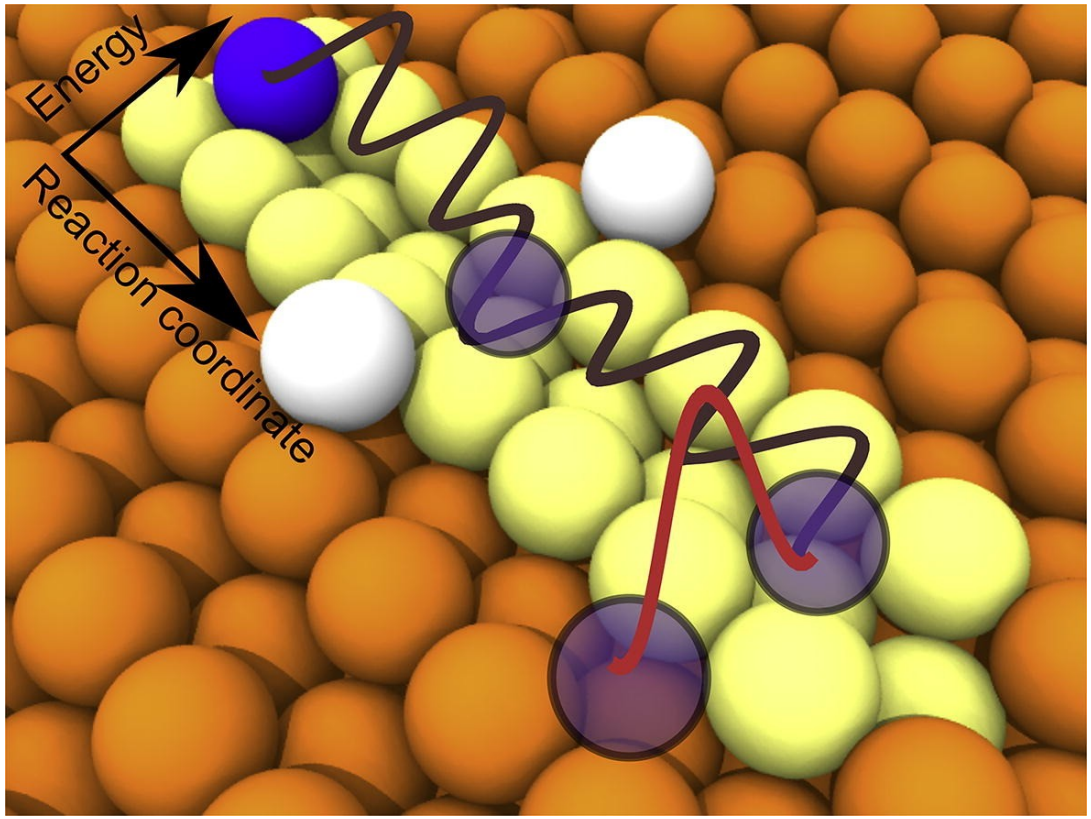


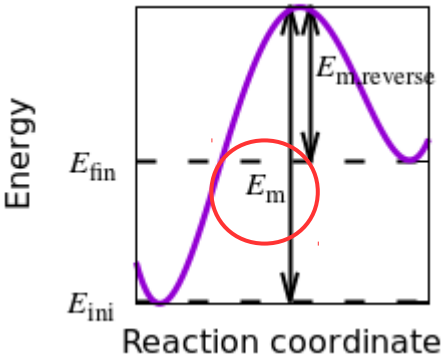


Motivation

- Surface diffusion is one of the possible contributors to the breakdown phenomenon
- Kinetic Monte Carlo (KMC) is the best tool for modelling diffusion in solids
- We have a working KMC model for surfaces, but it's not extensible to multiple elements (Cu + something else)
- Good to check if approximations are valid
  - Some information on the local atomic environment is disregarded in the current model

Kinetic Monte Carlo





Reaction rate:

$$\Gamma \propto \exp\left(-E_m / kT\right)$$

E. Baibuz et al. *Computational Materials Science* **146** (2018): 287-302

Why do we need machine learning?

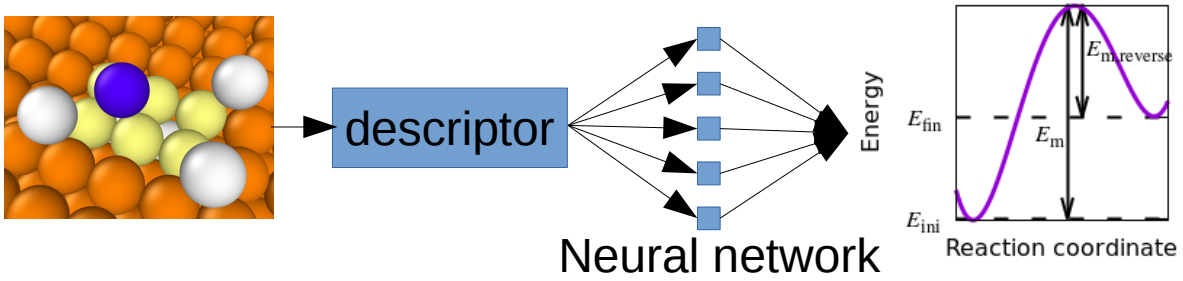
- We need a large number of accurate migration energy barriers for a good model of diffusion
- In principle, everything can be calculated (nudged elastic band), but it is too expensive computation-wise
- Solution:
  - either reduce the amount of barriers needed by some approximations, or
  - find uncalculated barriers using **some clever method**

Methods

- We consider the **relevant** local atomic environment of the jump to 2nd nearest neighbour sites
- The mapping of local atomic environment to migration barrier is a 26-dimensional function

$$E_m = f(s_0, s_1, s_2, \ldots, s_{25}) \qquad s_i = \{0, 1\}$$

- **Machine learning regression is used for interpolation/extrapolation of migration barriers, based on a training set that can be calculated**

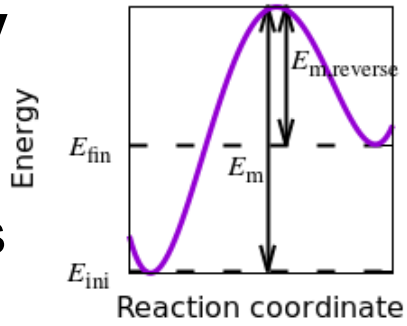


Methods

- Database of migration energy barriers calculated with the **LAMMPS** [1] implementation of the nudged elastic band (**NEB**) [2,3] method
  - Classical **MC/MD-CEM potential** [4]
- Machine learning regression with the **FANN library** [5]
- Kinetic Monte Carlo simulations with the **Kimocs** [6] atomistic KMC program
- Further details in will be in Kimari et al. (in preparation)

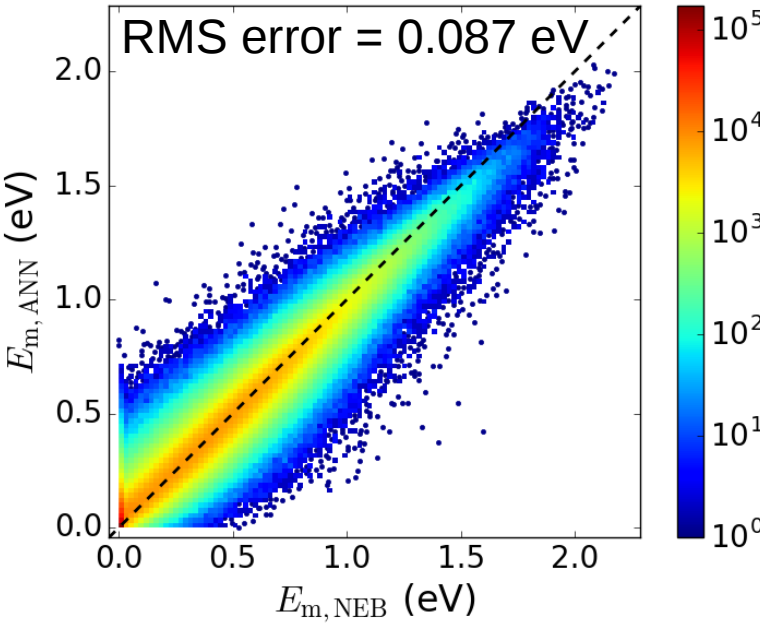
[1] S. Plimpton. *Journal of computational physics* **117**.1 (1995): 1–19.
[2] Mills, Jónsson, *Physical review letters* **72** (1994): 1124
[3] Mills et al. *Surface Science* **324** (1995): 305–337
[4] Stave et al. *The Journal of Chemical Physics* **93** (1990): 4413–4426
[5] S. Nissen. Tech. rep. Department of Computer Science University of Copenhagen (DIKU), 2003. url: http://fann.sf.net.
[6] V. Jansson et al. *Nanotechnology* **27**.26 (2016): 265708.

Machine learning accuracy

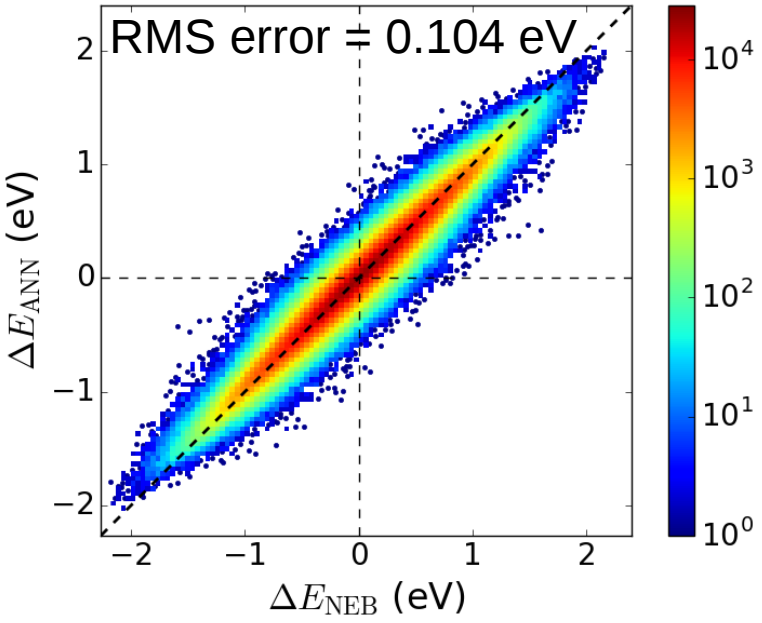


- Correlation of true vs. ML-predicted values
- $\Delta E = E_{\mathrm{fin}} - E_{\mathrm{ini}}$

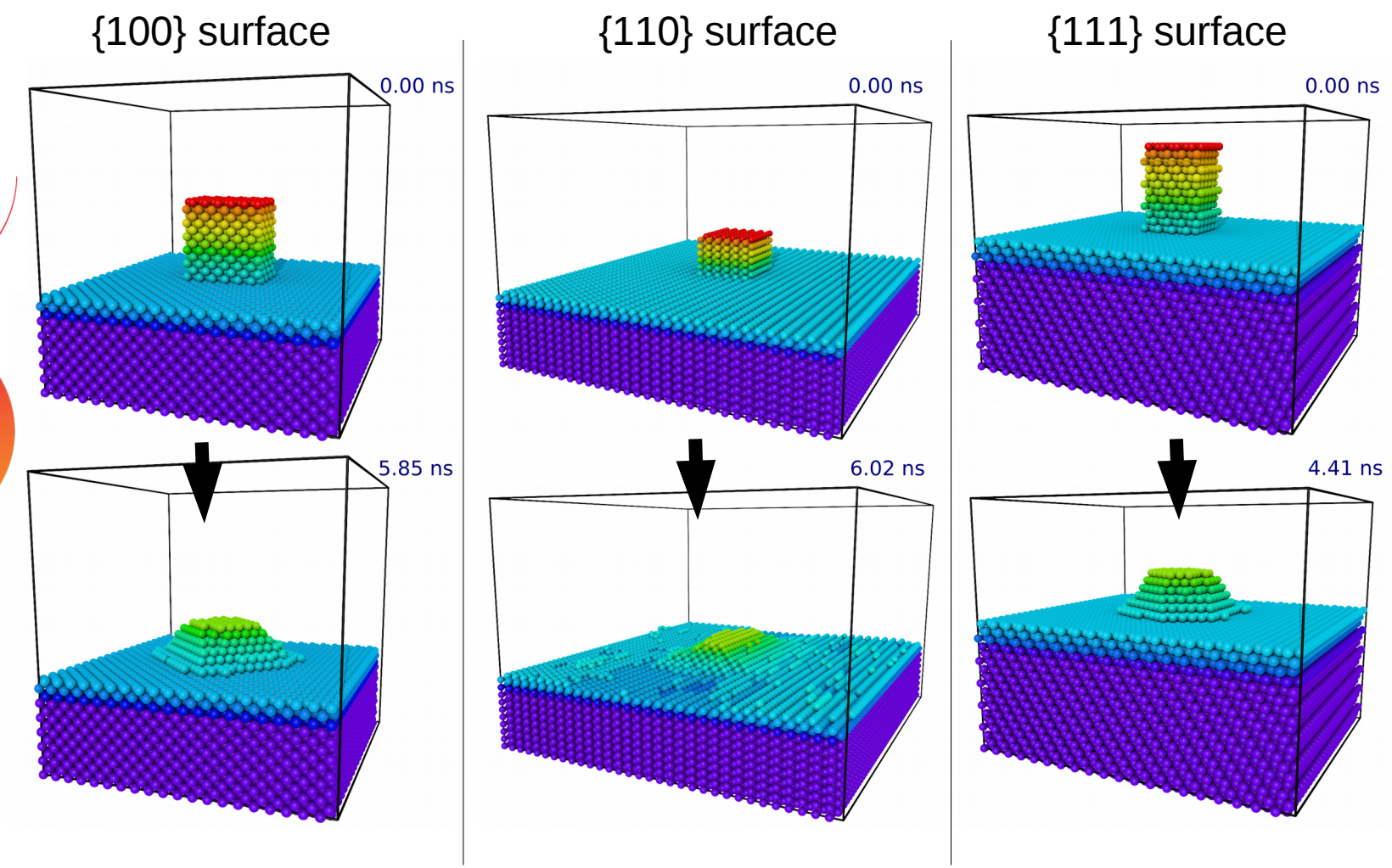
"Kinetics"



"Thermodynamics"

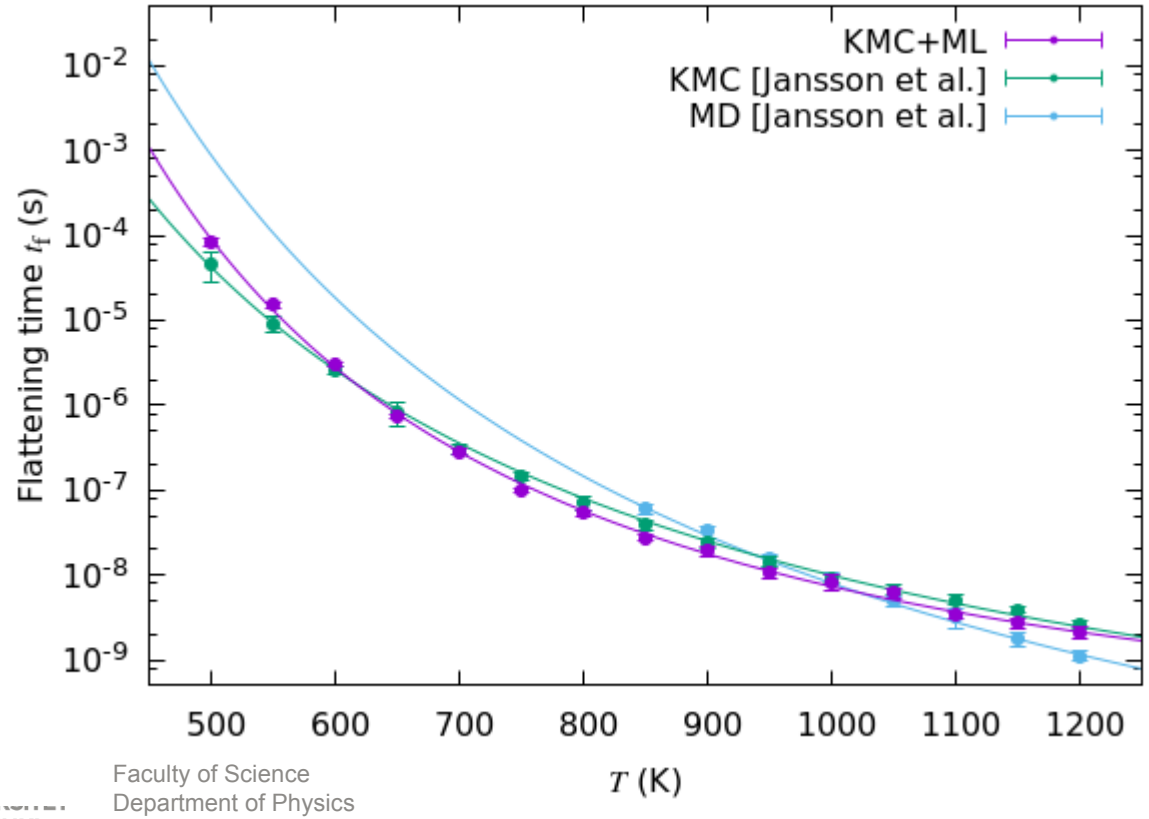


Cuboid nanotip flattening



Finalizing the parameterization

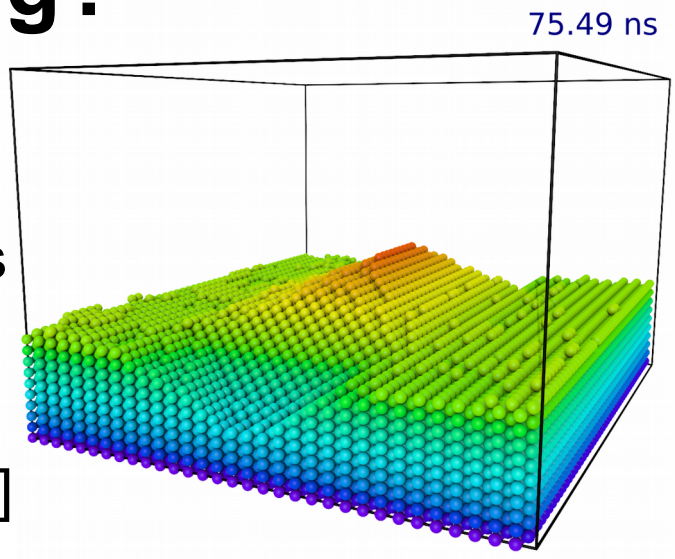
- Fixing the KMC time scale can be done by fitting to molecular dynamics simulations of nanotip flattening

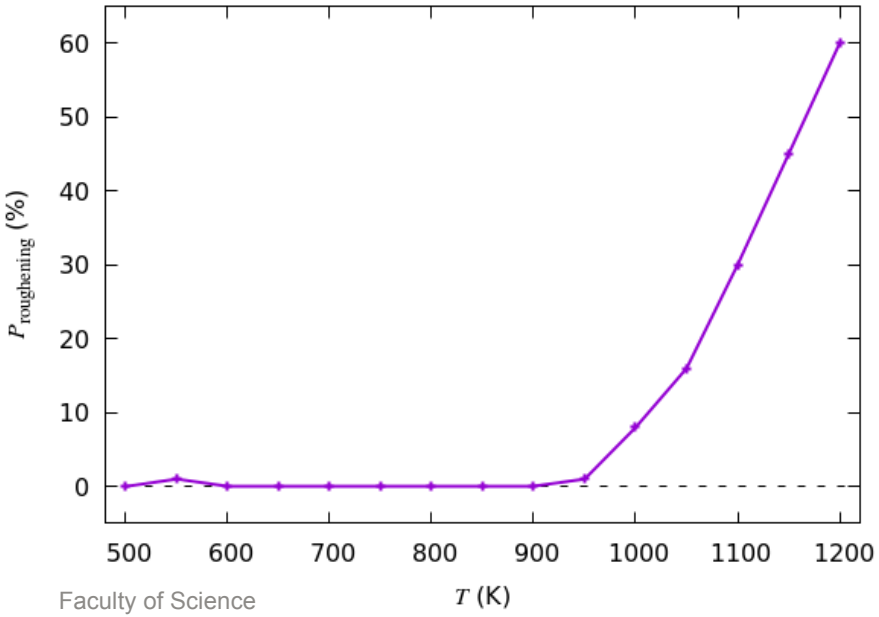


$$\Gamma \propto \exp\left(-E_m / kT\right)$$

Faceting = roughening?

- Sometimes the {110} surface destabilises before the tip flattens
- Happens surprisingly close to the known Cu {110} roughening temperature at 900–1000 K! [7–9]

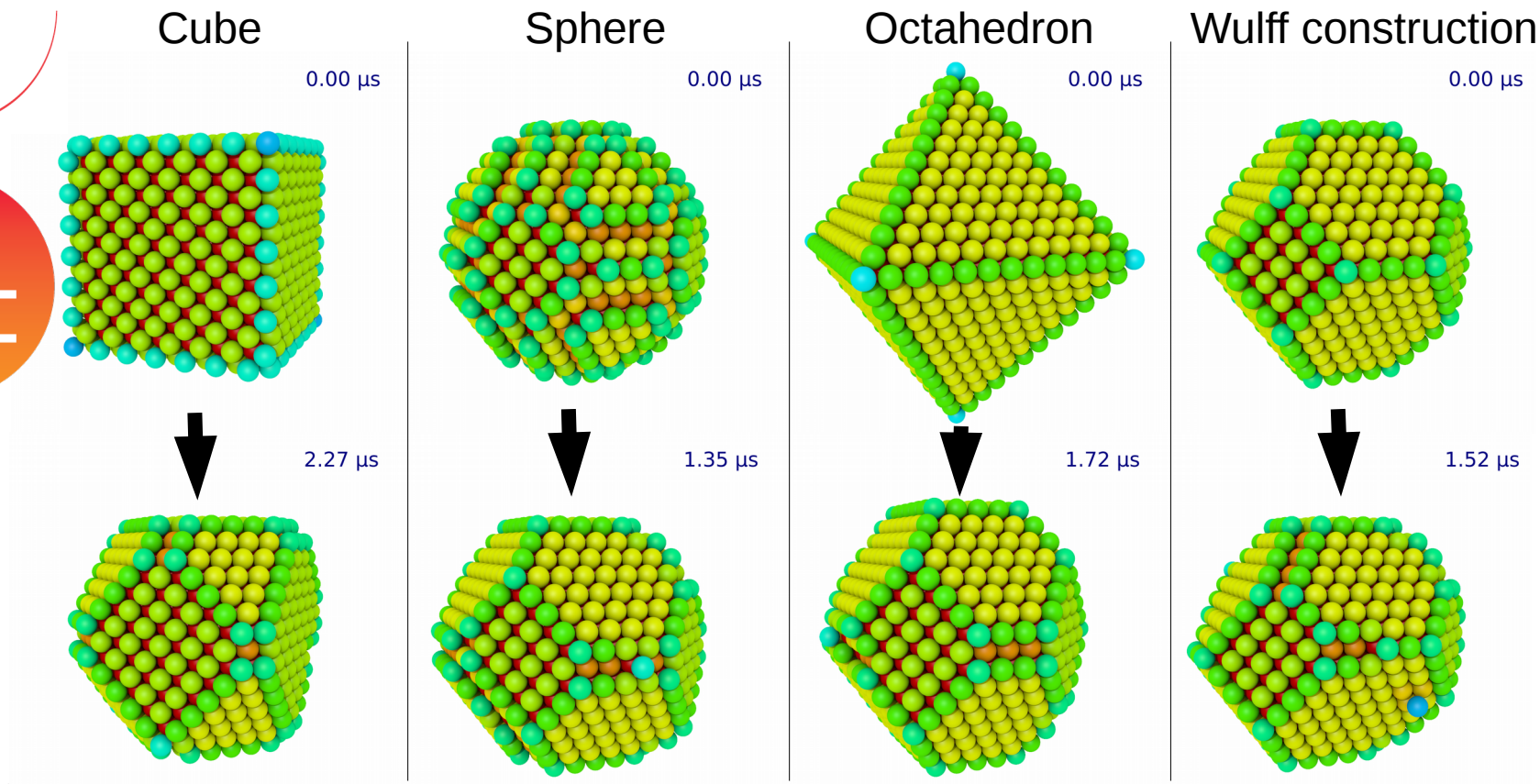




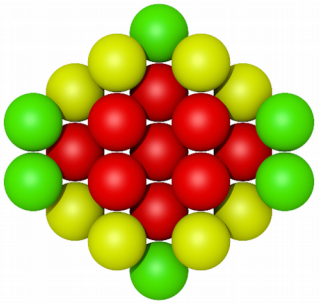
[7] Mochrie. *Physical review letters* **59**.3 (1987): 304.
[8] P. Zeppenfeld et al. *Physical review letters* **62**.1 (1989): 63.
[9] H. Häkkinen et al. *Physical review letters* **70**.16 (1993): 2451.

Nanoparticles → surface energies

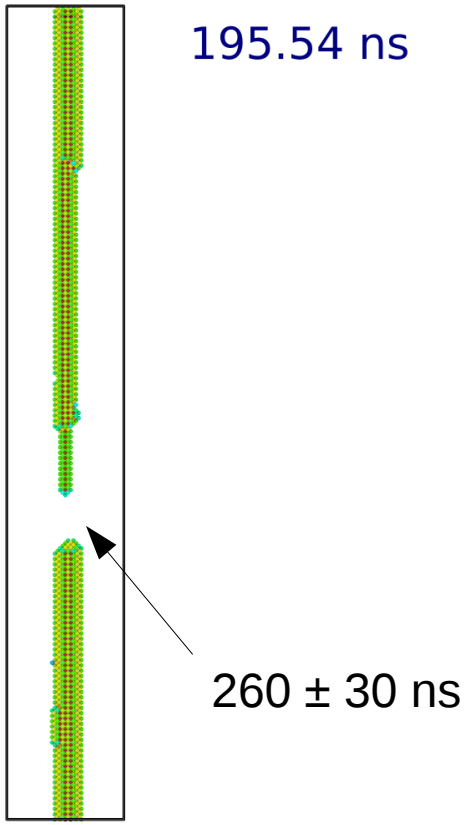
- Relaxing nanoparticle shapes at 900 K

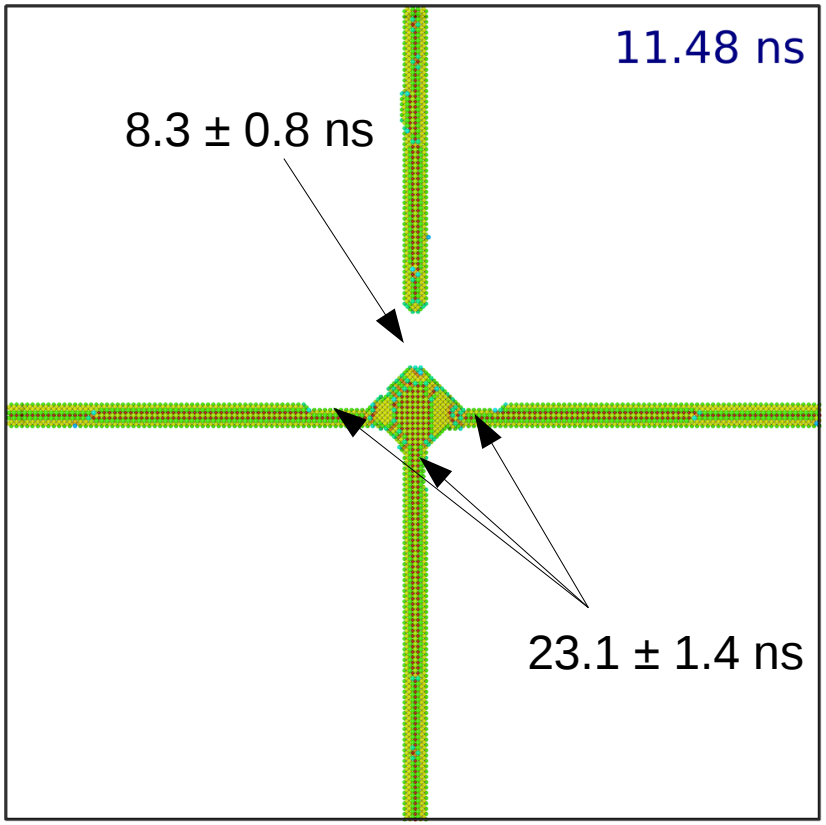


Nanowire fragmentation



- Long nanowires break up by diffusion, 1000 K



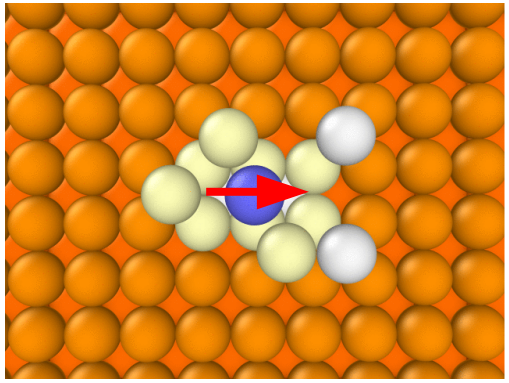


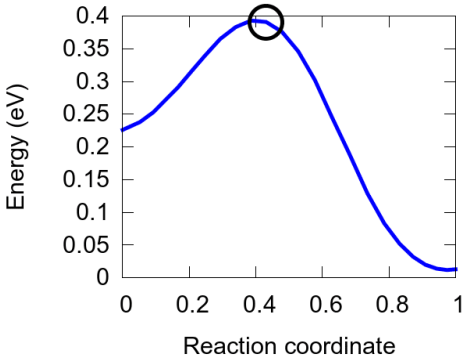
Conclusions

- The machine learning parameterized kinetic Monte Carlo Cu surface model performs at a satisfactory level
- The thermodynamics of the Cu system can be taught to the artificial neural network implicitly
- Destabilisation of the {110} surface observed very near to the known roughening temperature of this surface
- Outlook
  - Extending the model to 3rd and 4th nearest neighbours for potential accuracy gain
  - Adding more elements in addition to Cu
  - Starting the simulations with electric field

Nudged elastic band calculations

- We need the attempt frequencies ν and the migration energies E_m
- Approximation: same attempt frequency for all events
- The barriers are highly dependent on the environment of the jump → the nudged elastic band (NEB) method





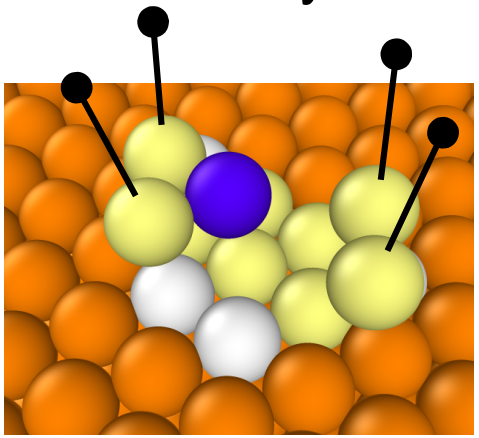
LAMMPS:
S. Plimpton, *Journal of computational physics* **117** (1995): 1–19

NEB:
Mills, Jónsson, *Physical review letters* **72** (1994): 1124
Mills et al. *Surface Science* **324** (1995): 305–337

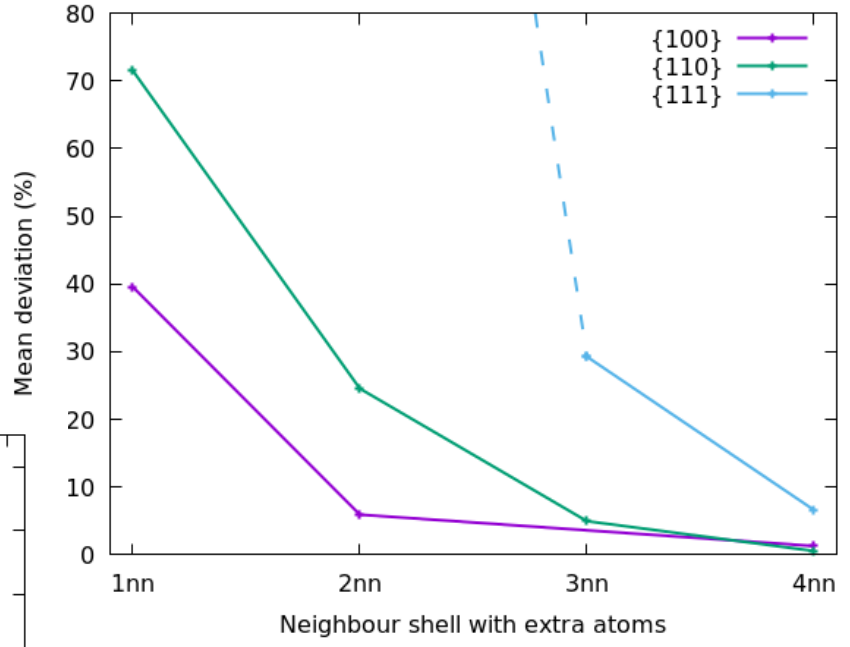
MC/MD-CEM potential:
Stave et al. *The Journal of Chemical Physics* **93** (1990): 4413–4426

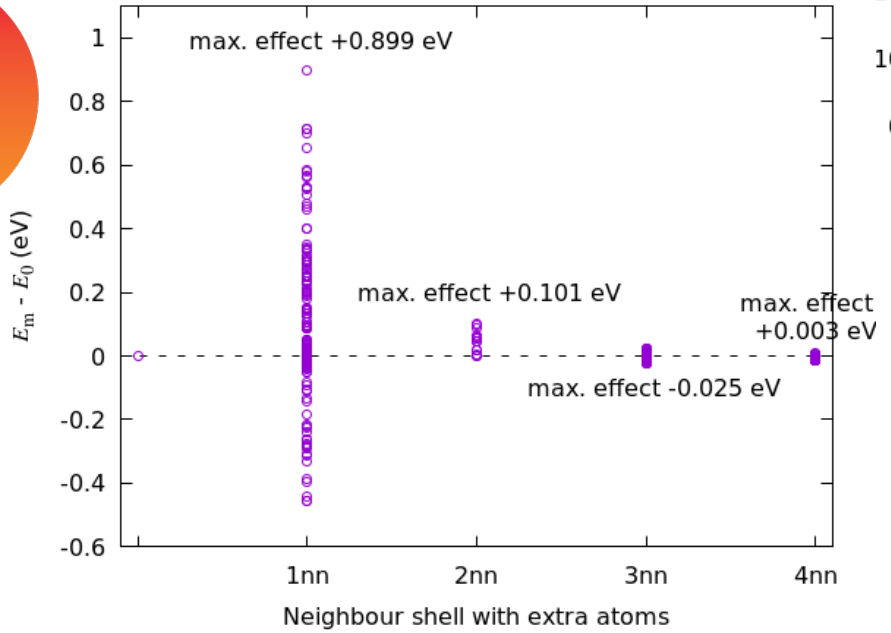
Tethering; problems on the {111}

- Atoms tend to move around pretty easily on the surface nudged elastic band
- If the environment changes, we *cannot use the barrier* obtained from the calculation
- Solution by S. Vigonski: thether all atoms loosely in their initial lattice positions
- Consequence: the barriers are slightly distorted
- The basic jump on the {111} surface is overestimated by a factor of 3… and the machine learning model adds another factor of 2



2nn approximation: error of 0.025 eV





Data efficiency