An off-lattice kinetic Monte Carlo method for the investigation of grain boundary kinetic processes

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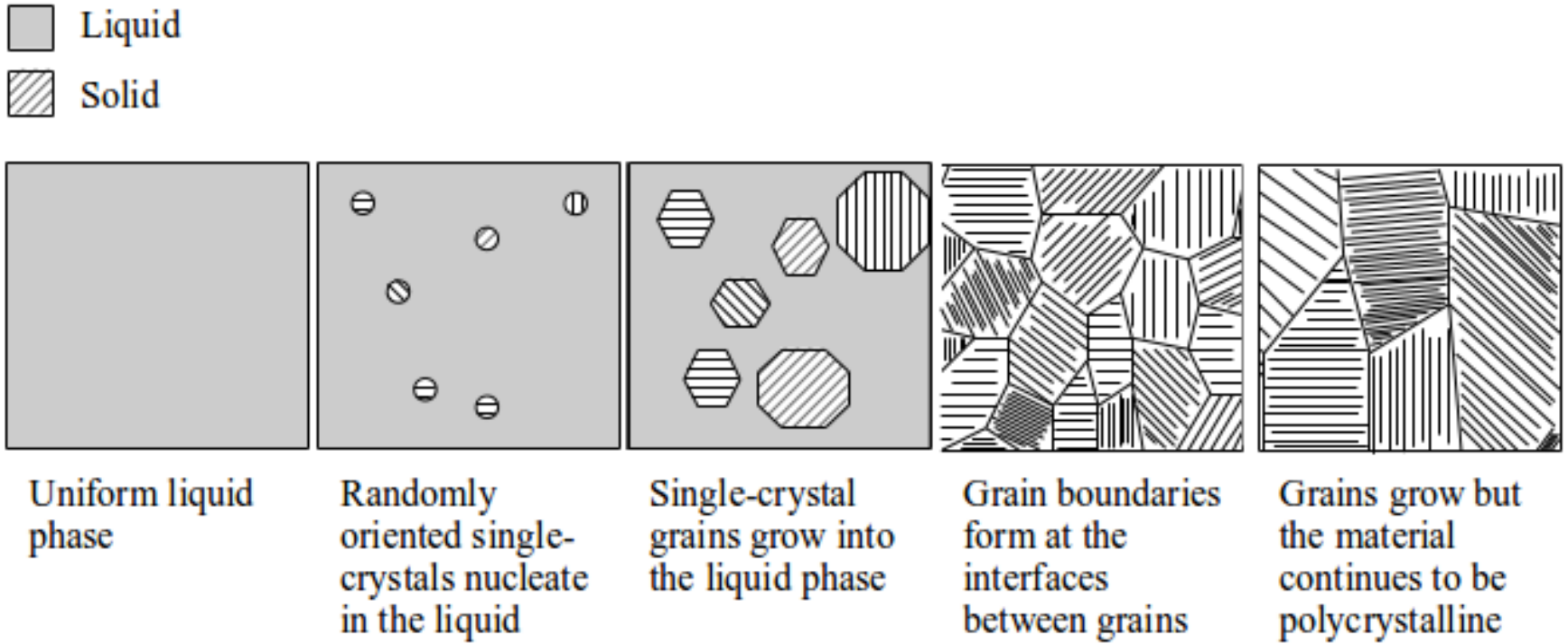
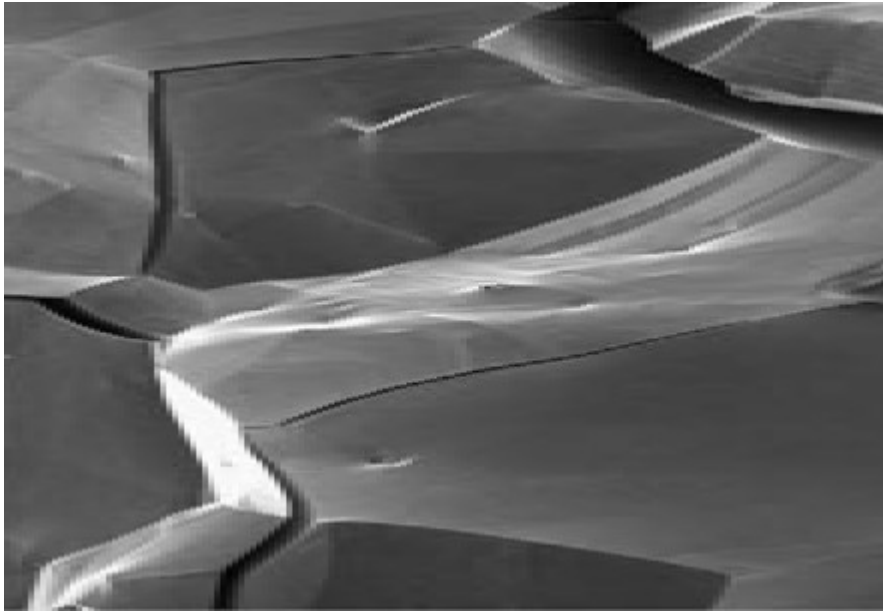
Grain boundaries (GBs) are microscale defects ubiquitous in engineering materials



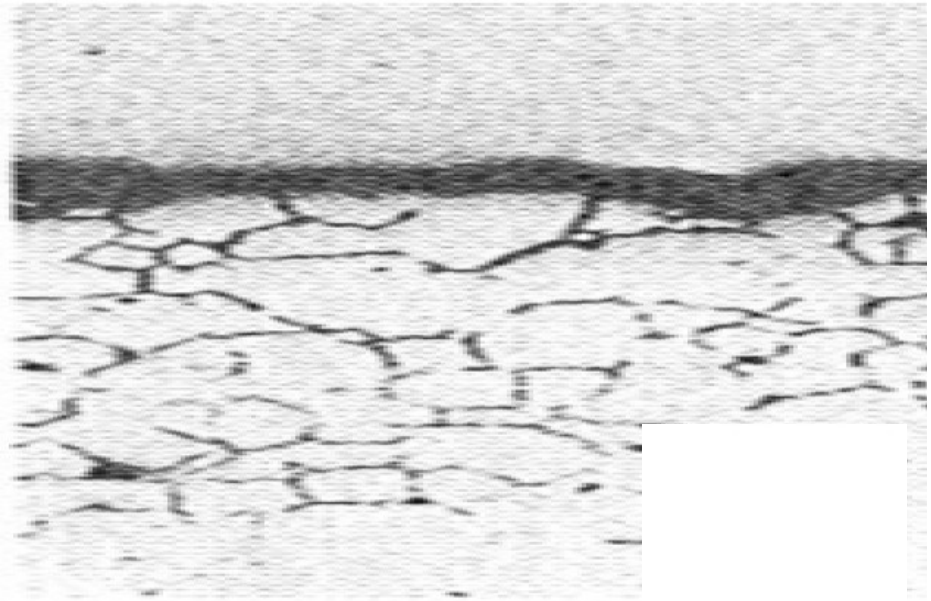
Grain boundaries mediate failure in materials

Intergranular fracture in Nickel



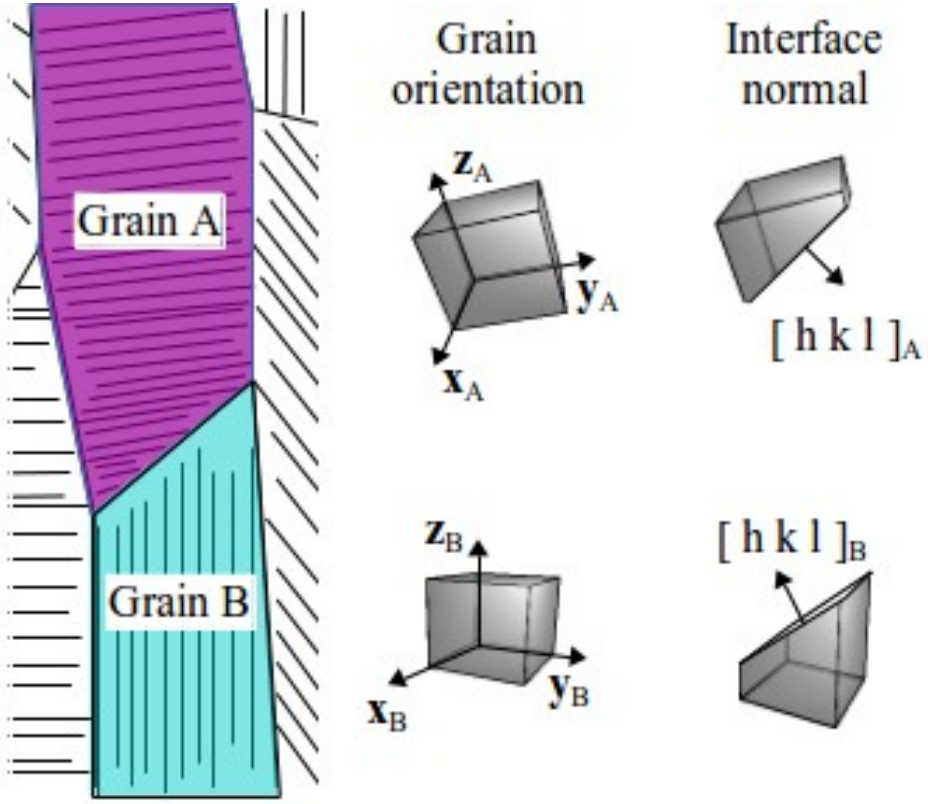
Bechtle, et al., *Acta Mater.* (2009)

Intergranular Corrosion in Fe-35Ni-25Cr



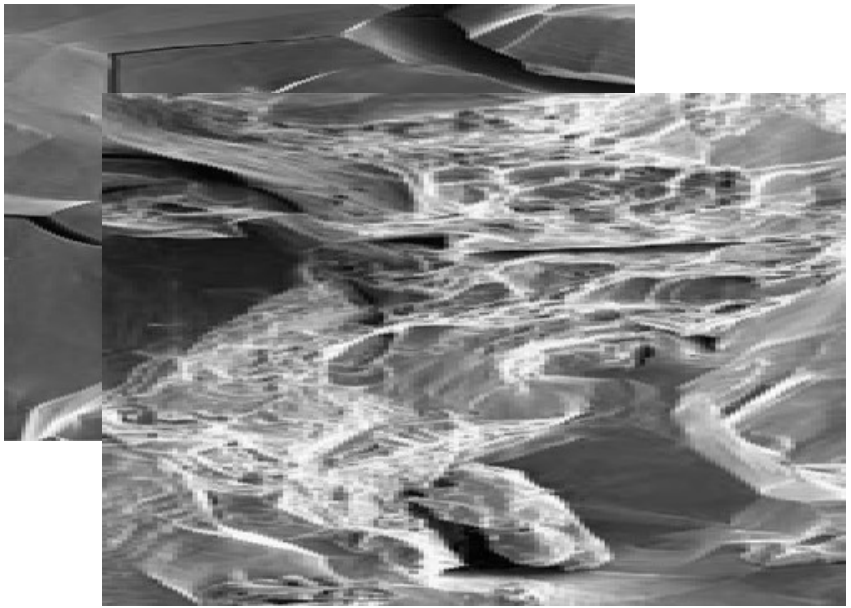
Palumbo, et al., *JOM* (1998)

Grain boundary orientation matters



GB engineering exploits differences in properties between GBs with different orientation

Intergranular fracture in Nickle



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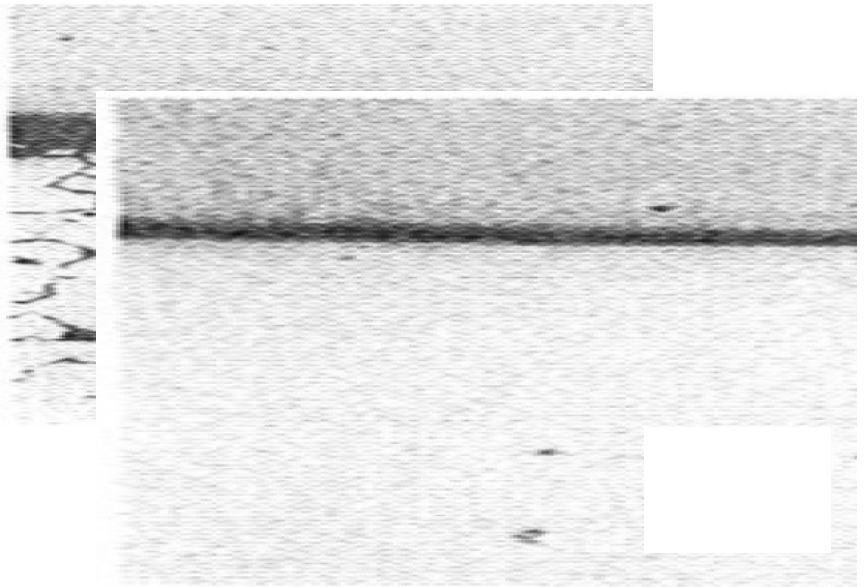
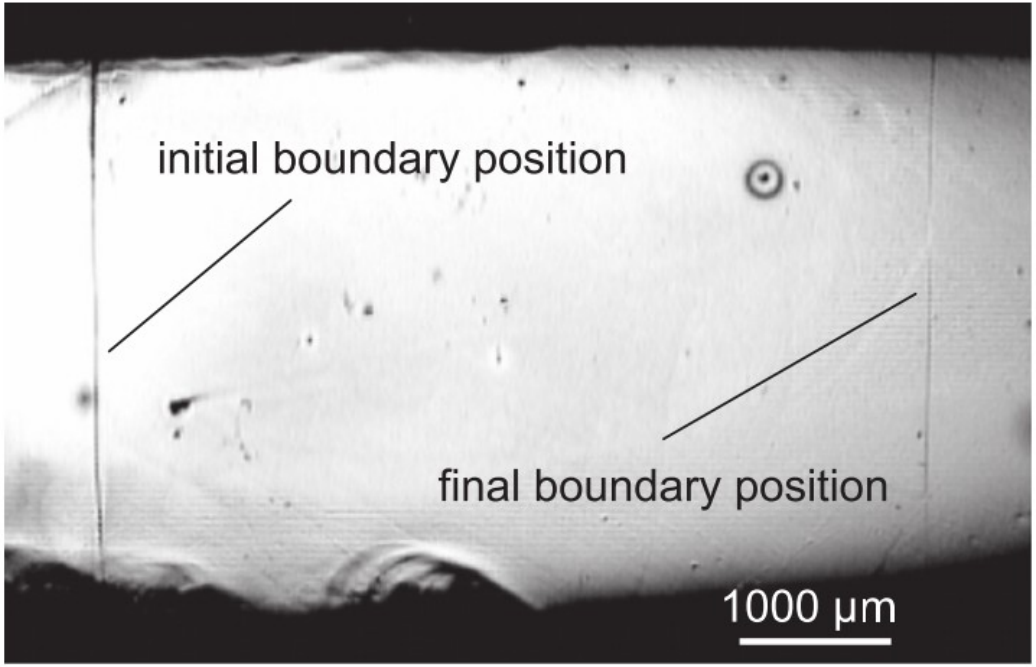
Intergranular Corrosion in Fe-35Ni-25Cr



Palumbo, et al., *JOM* (1998)

We need to know which GBs will provide favorable properties

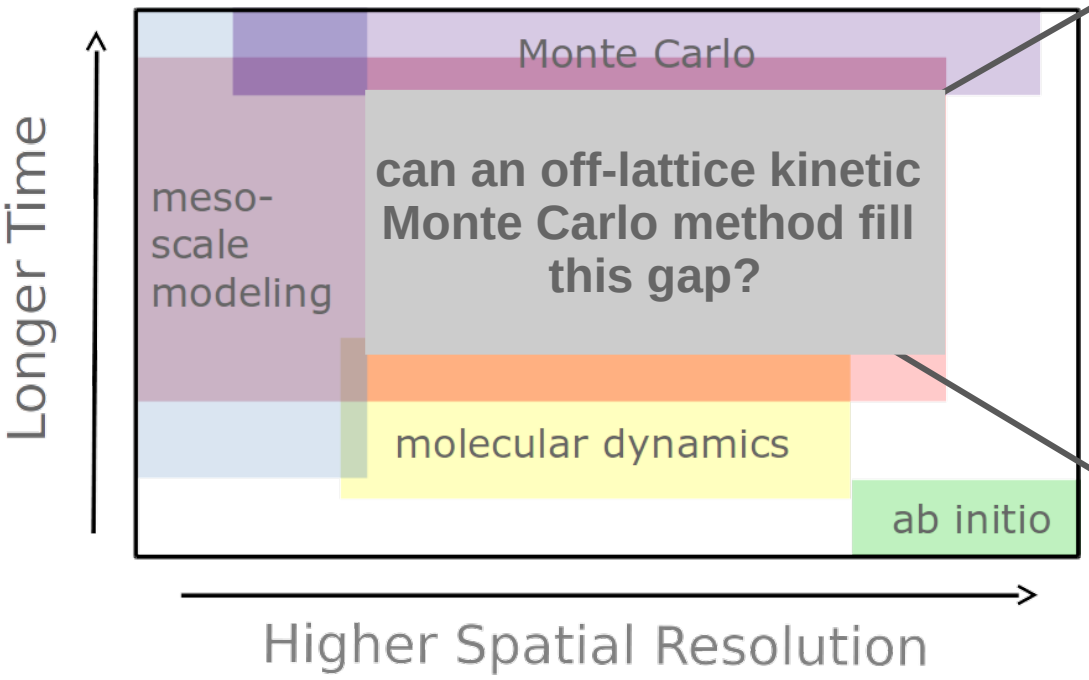


Günster, et al., *Acta Mater.* (2013)

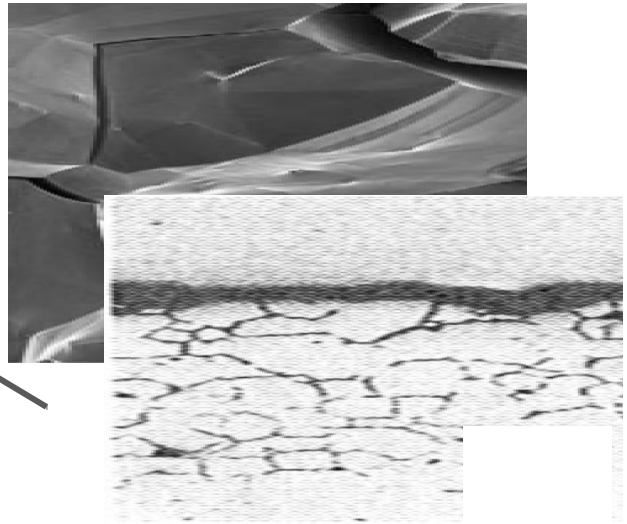
Conventionally, this is done with time-consuming experiments on bicrystals—
not a high-throughput method

...so, let's use computers!

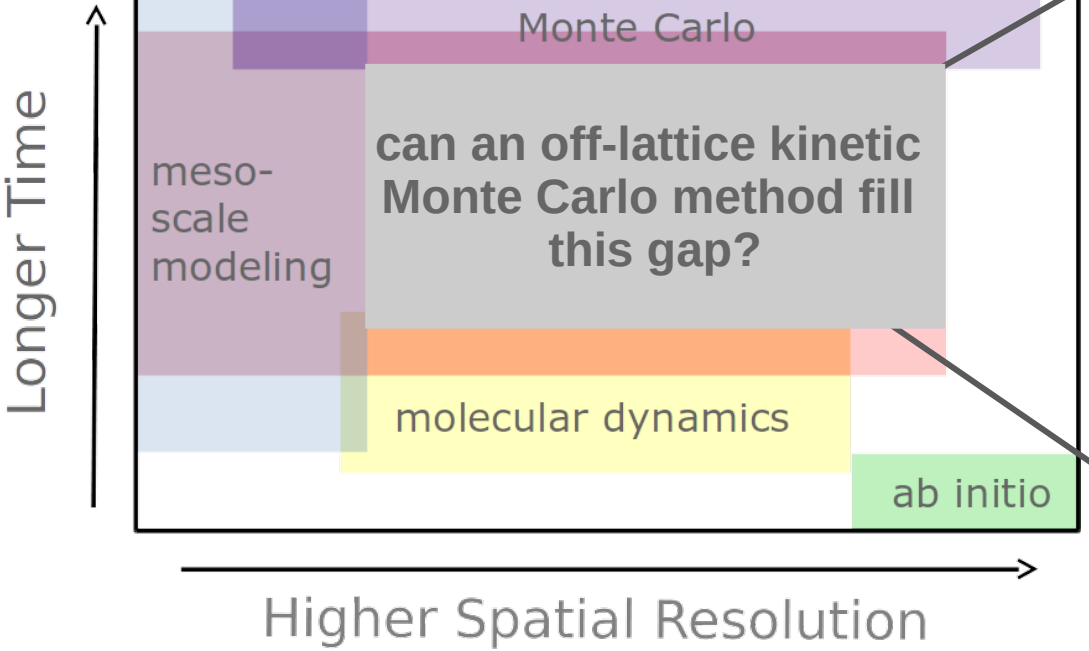
The computational materials science toolbox



Grain boundary kinetics live in the no-man's-land region of the map



The computational materials science toolbox

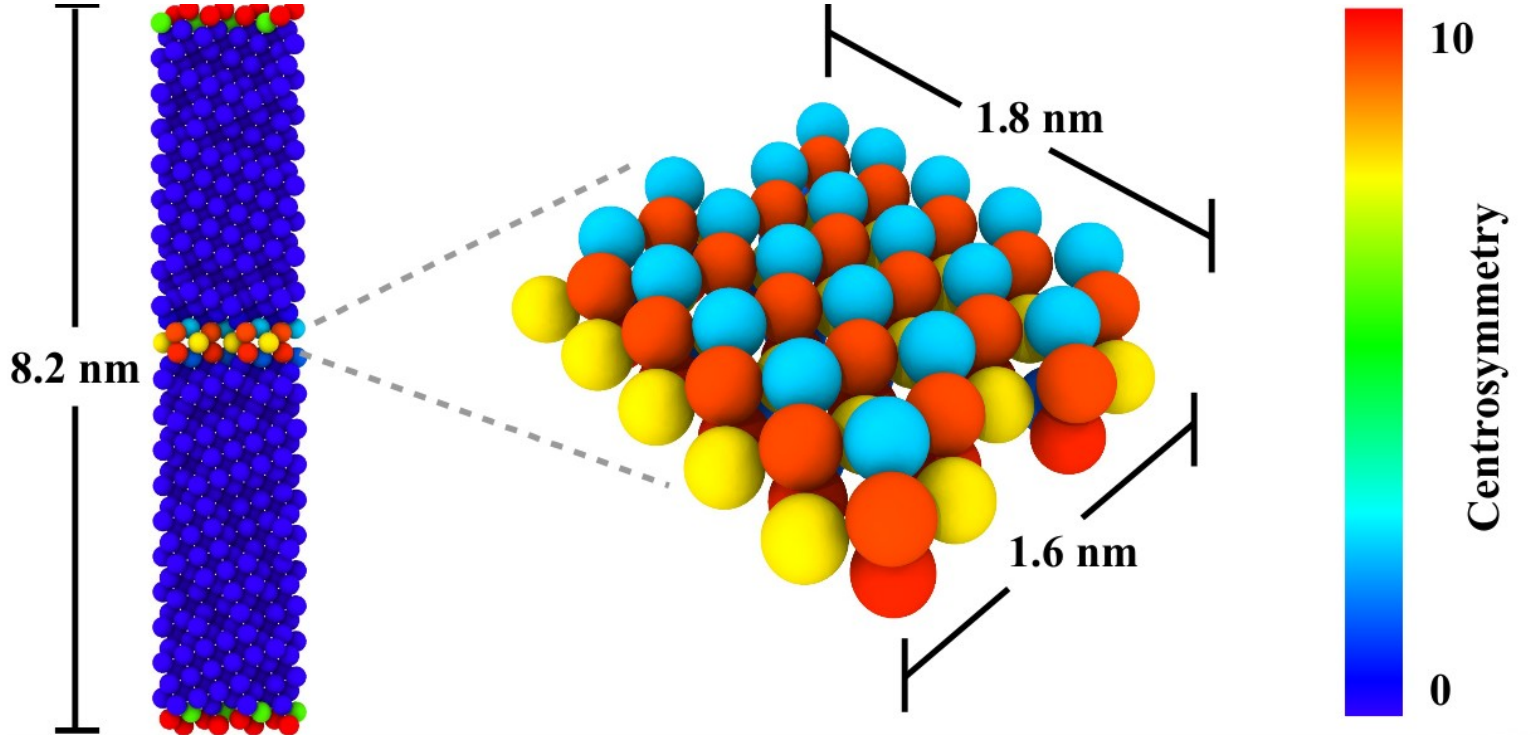


The stochastic nature of KMC simulations is **ideal for parallelization** and allows access to much longer timescales, especially at low temperatures

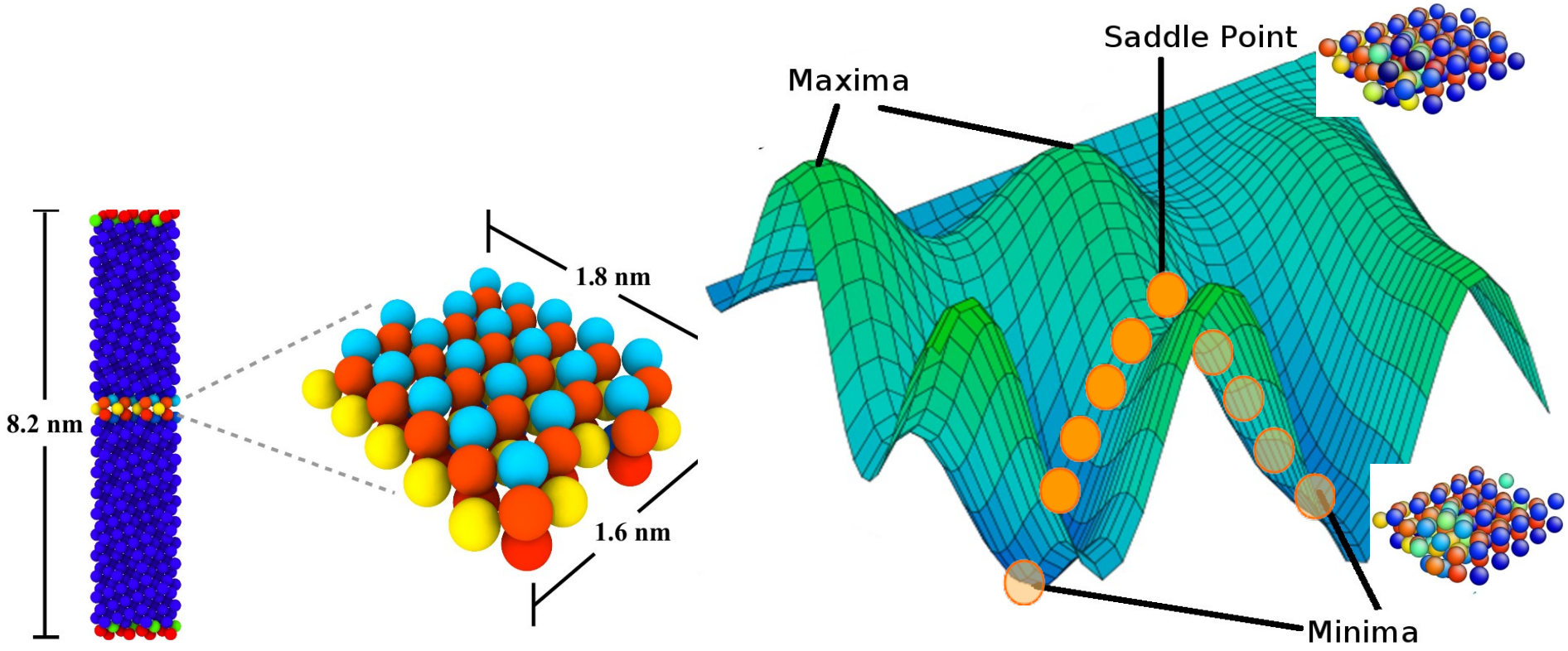
$$f\left(R_N(T), \mathrm{rand}(0,1]\right)$$

An example system

Σ5 (2 1 0) GB in Cu

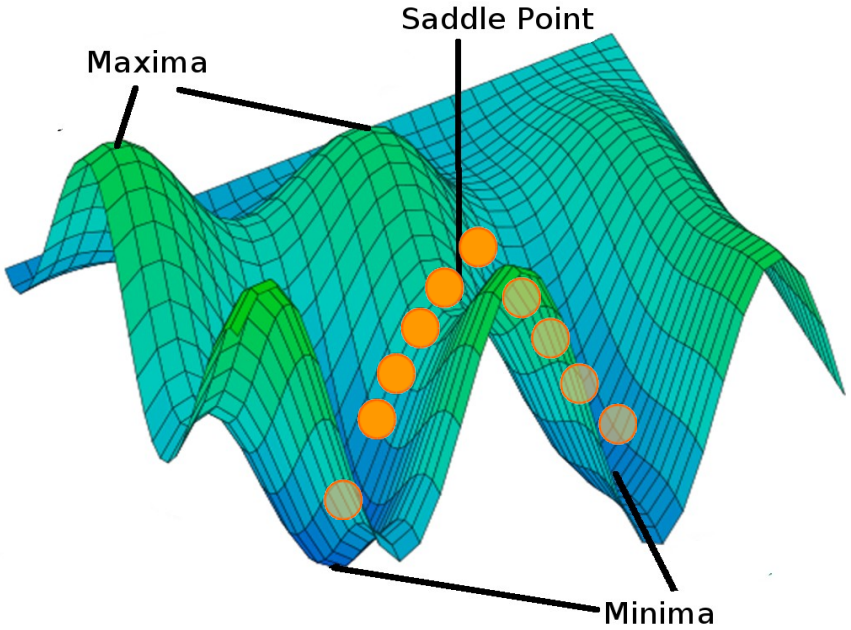


An example energy landscape



Why it's hard

$$R_N = \sum_{i=1}^{N} \sigma_i \nu_i^* \exp \frac{-E_{a_i}}{kT}$$



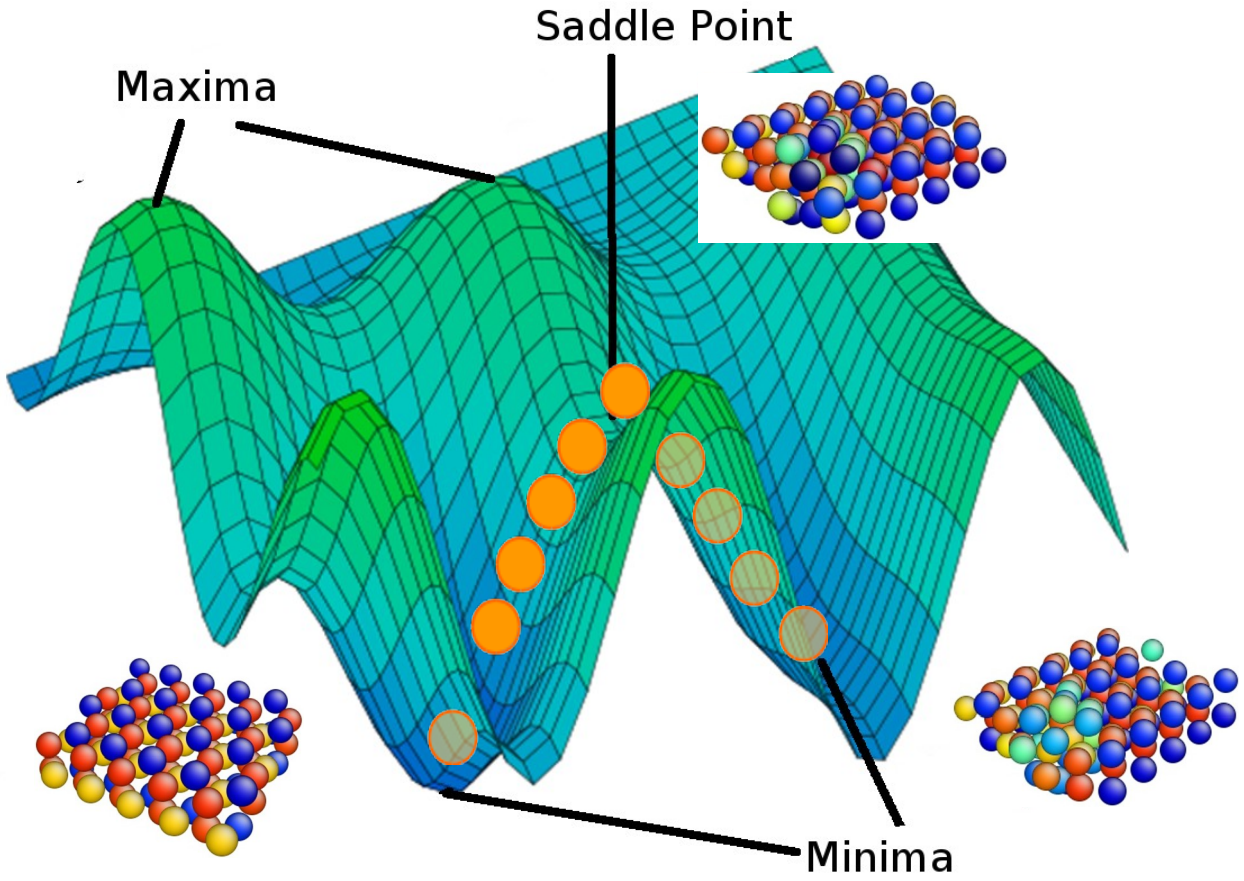
1. Have to find individual saddle points
2. Have to find them all, which means…

   have to be able to compare them,
   tell them apart, catalog them, etc.

Activation-Relaxation Technique

ART is an eigenvector following method. It finds saddle points by following the lowest curvature of the Hessian Matrix.

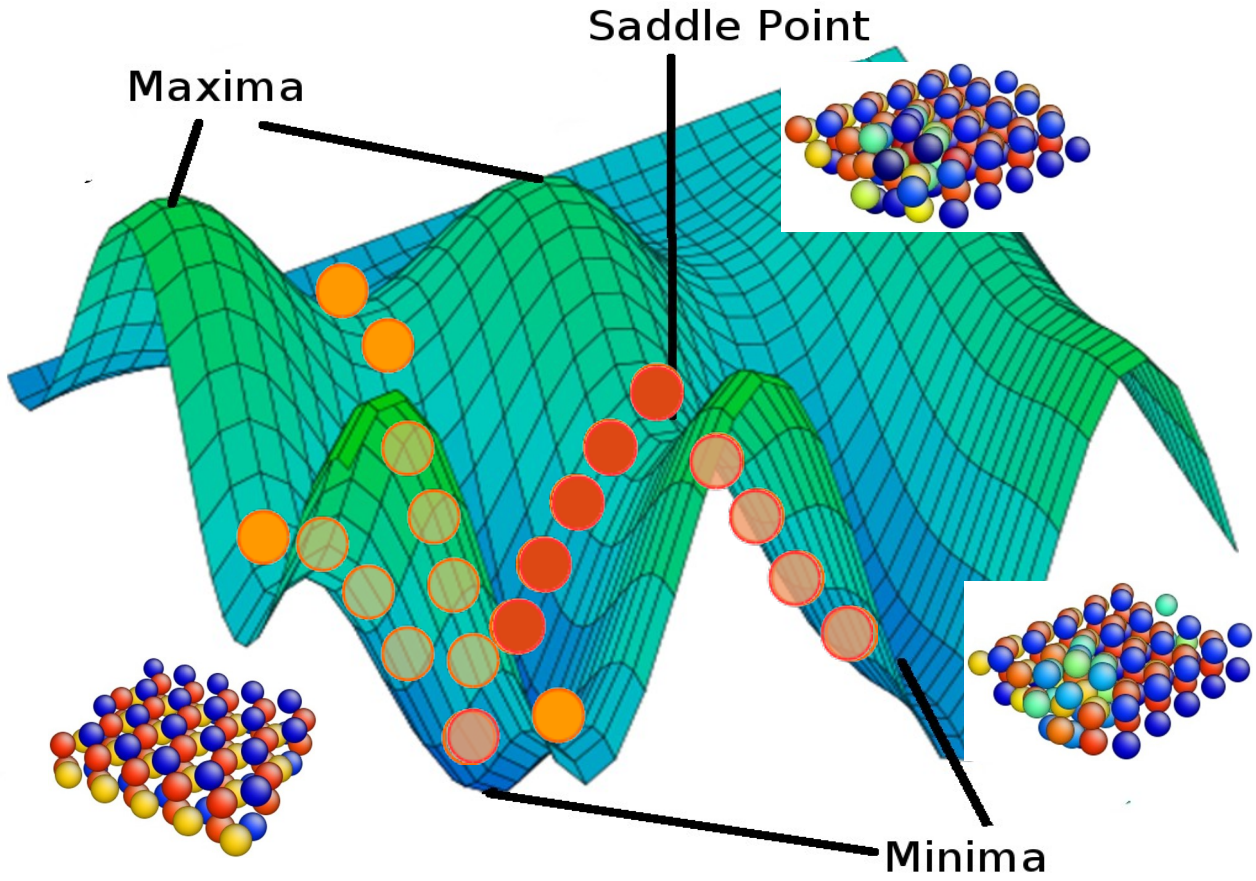
Mousseau, et al., *Phys. Rev. E* (1998)



Performing complete searches

By repeating random perturbations, it is possible to explore the energy landscape.

State of the art is essentially based on arbitrary number of searches, and then calling it “complete”

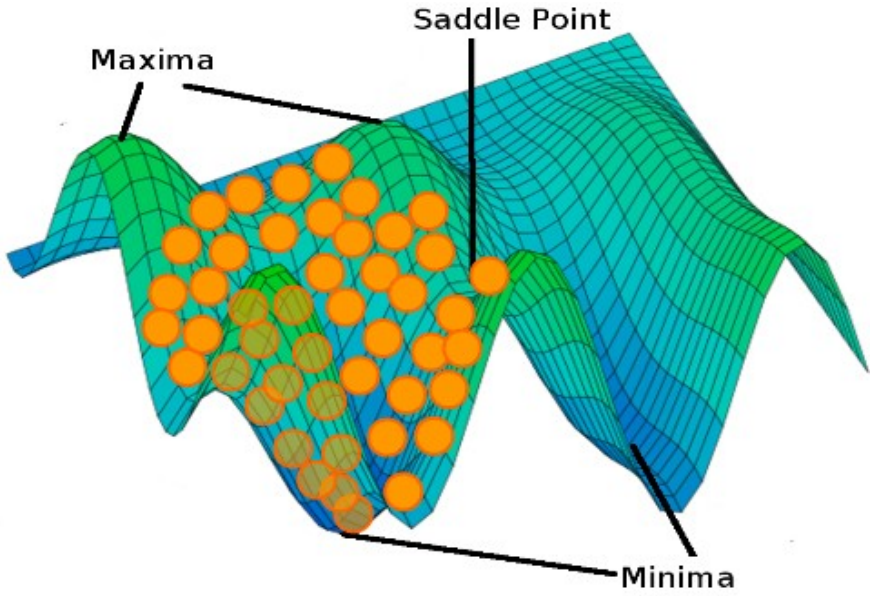


Framework

For performing complete searches- *we need to systematically search the energy landscape*.

Convention is to search randomly;

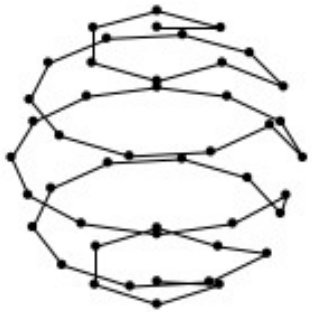
We search deterministically instead.



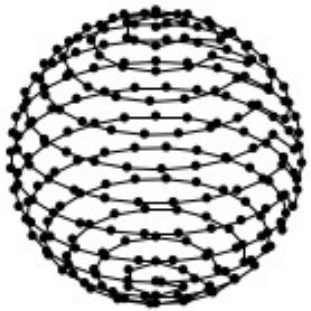
Deterministically searching

How many perturbations required to fully explore the accessible kinetic events?

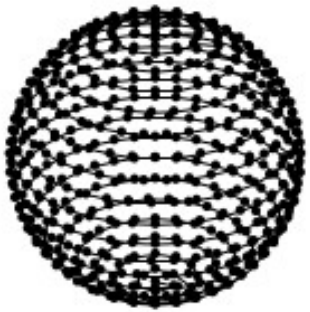
Search space is formally (N x 3) dimensional, but searches are effected by perturbing individual atoms through small physical displacements.



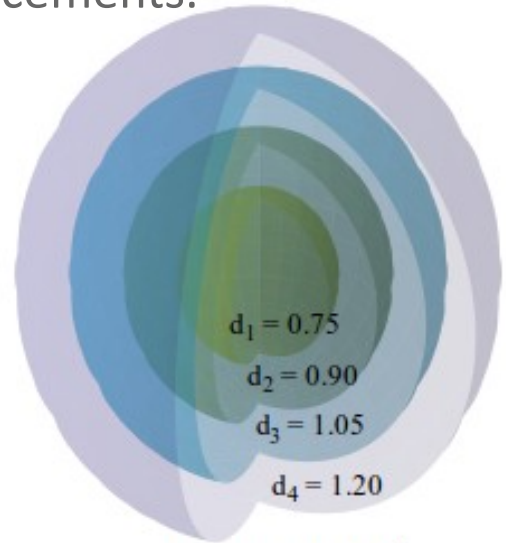
$\rho = 46$



$\rho = 248$

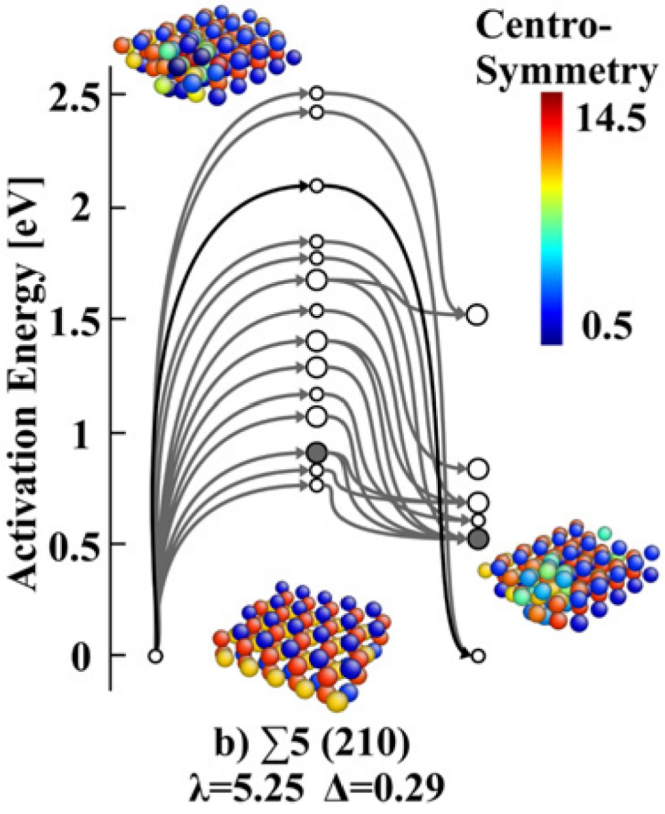


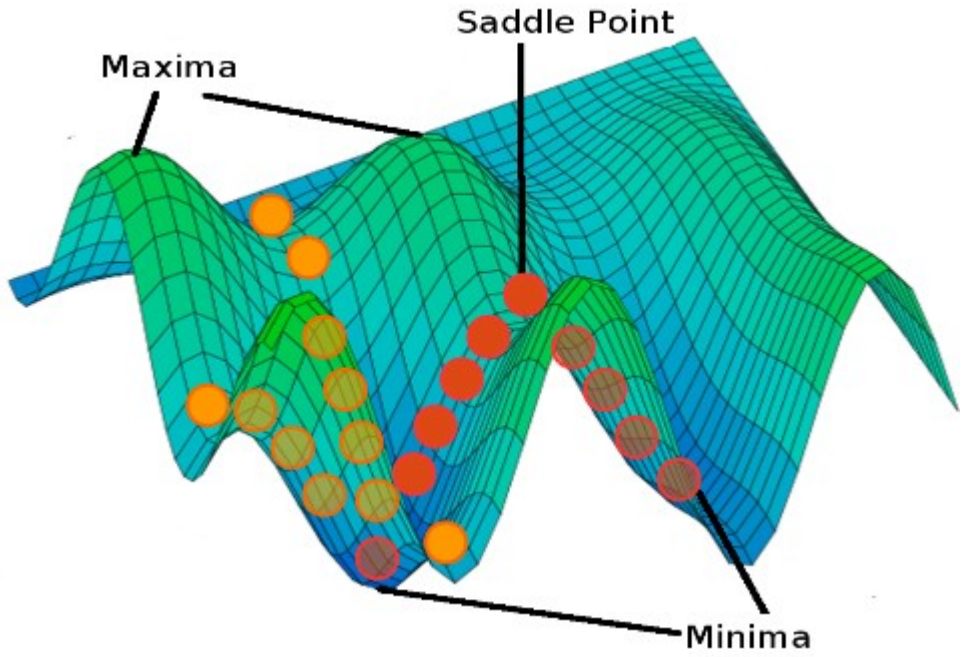
$\rho = 562$



$d = 4, \delta = 0.15$ Å

How to compare events? How even to define events?





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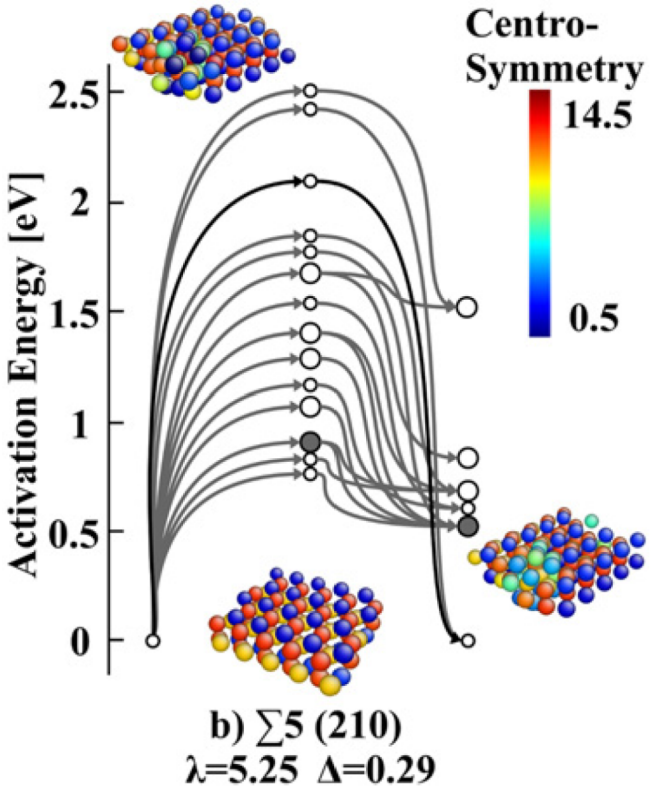
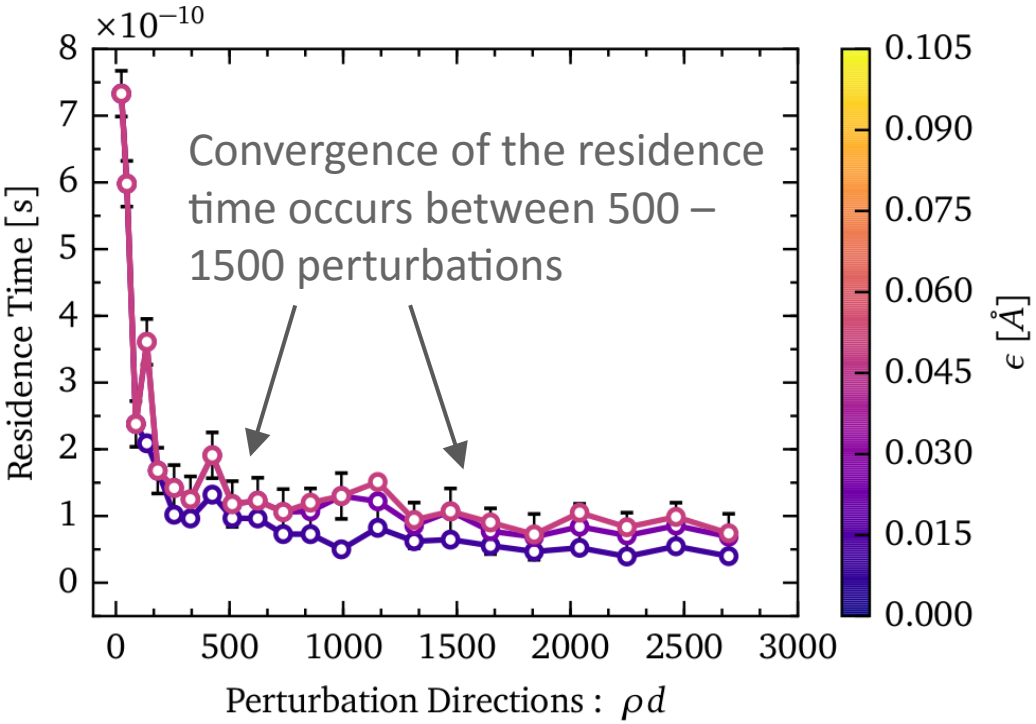


Energies alone say nothing

Topology must be captured, but using cutoffs, neighbors, etc., causes real problems

We favor a (data-intesive) approach of using all displacement vectors, as they can be mapped directly to position uncertainties in energy minimization in the first place

Demonstrated convergence

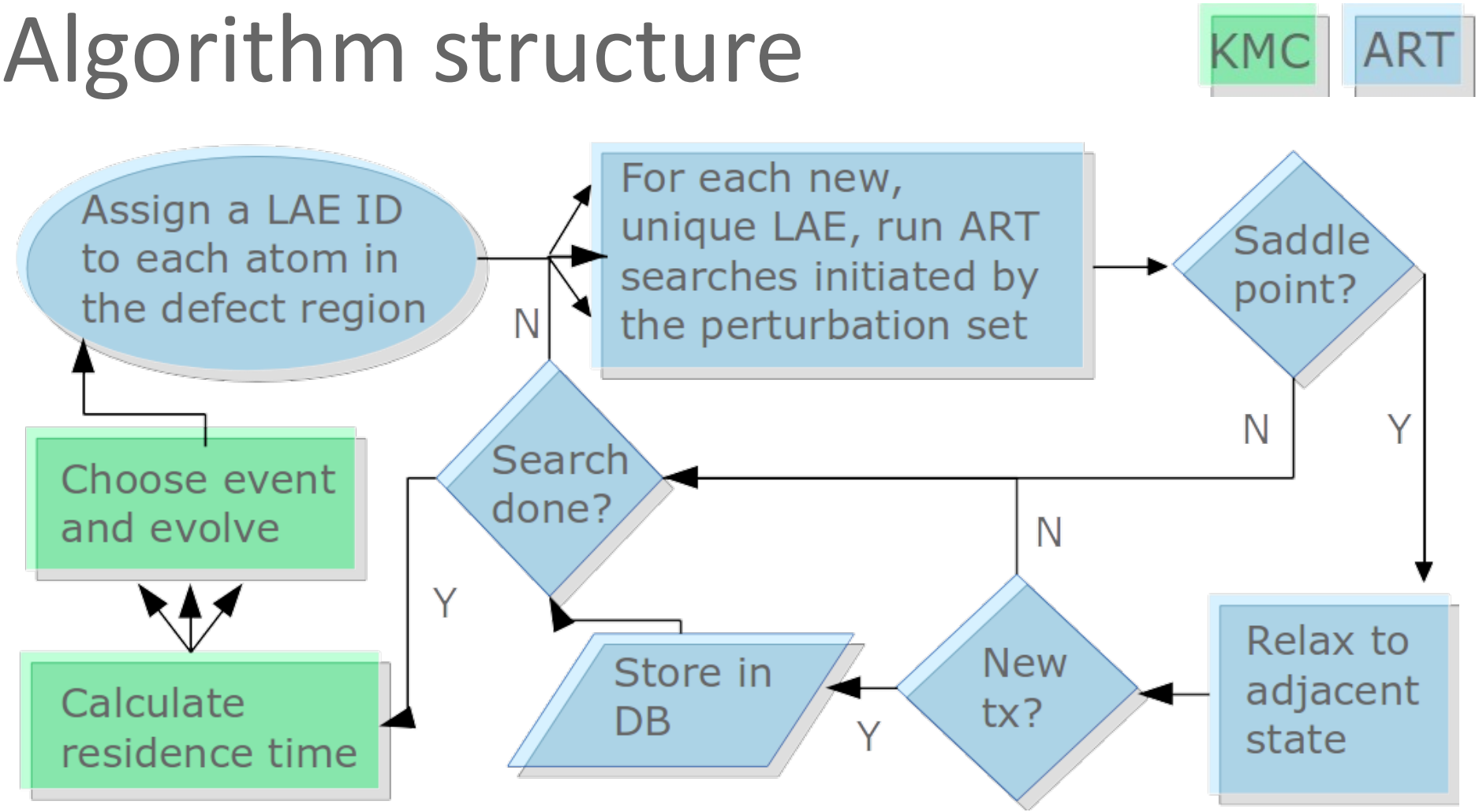


Convergence, pegged to built in uncertainties of energy minimization: *the uncertainties are no worse than those we accept already in using the potential and standard conjugate gradient minimization*

We are not aware of prior efforts to explicitly achieve converged residence times in an off-lattice KMC algorithm.

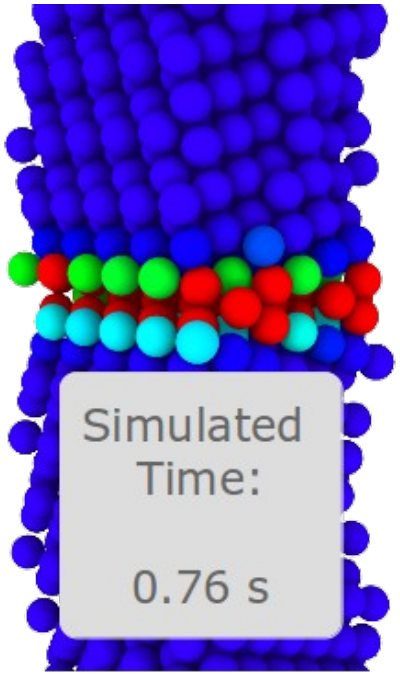
This GB is indeed simple enough to fully map, and therefore to KMC.

Algorithm structure

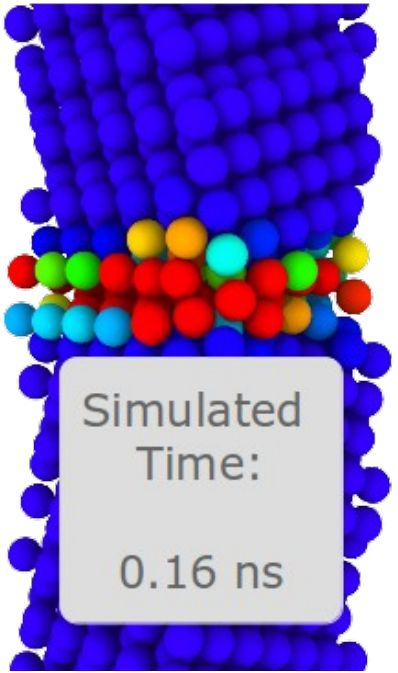


KMC Simulations

298 K

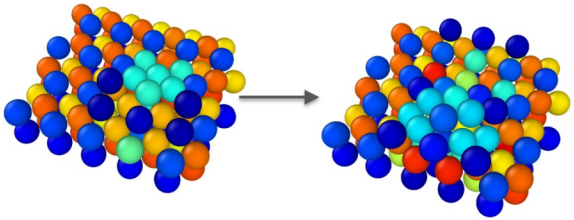


1173 K

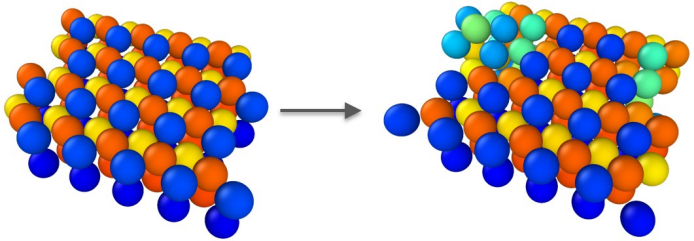


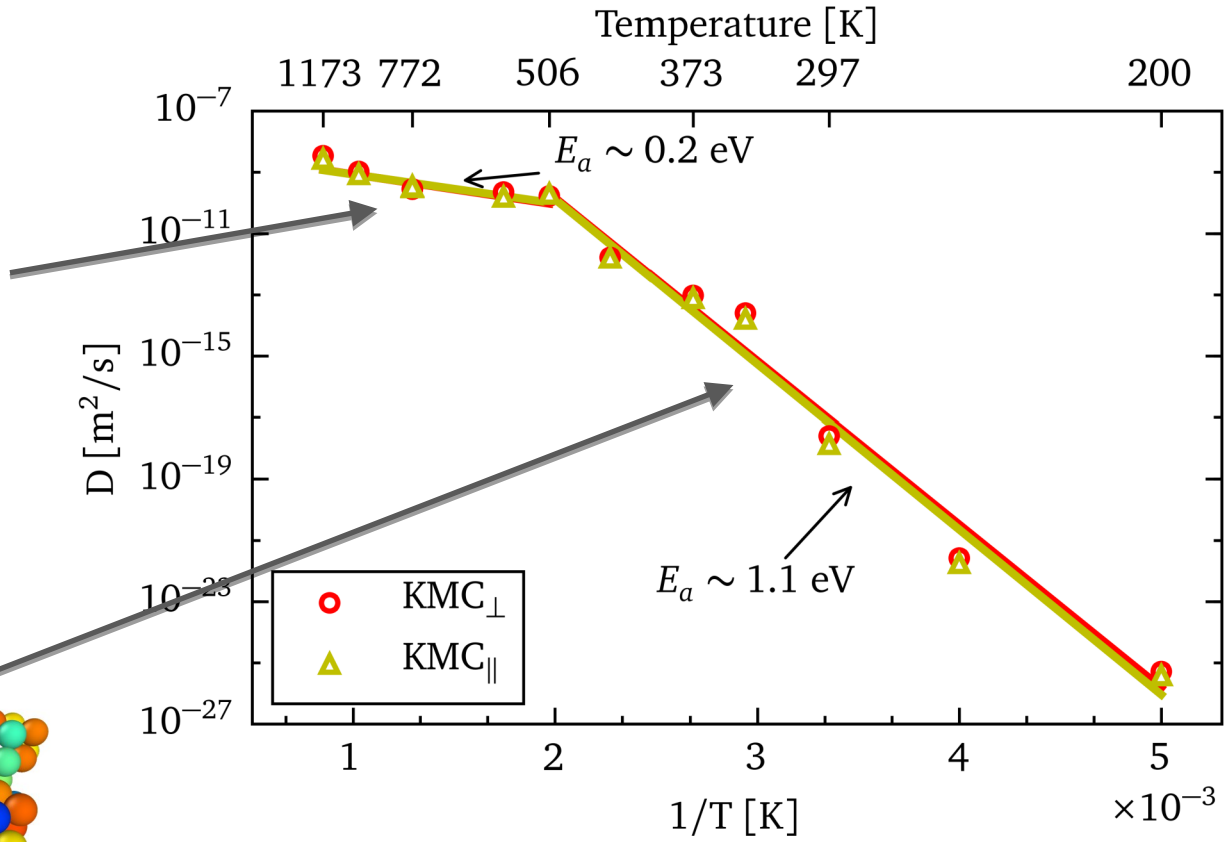
Two classes of events

Low energy events that take place in a disordered GB, and which transport atoms



High energy events that disrupt the order of a ground state GB

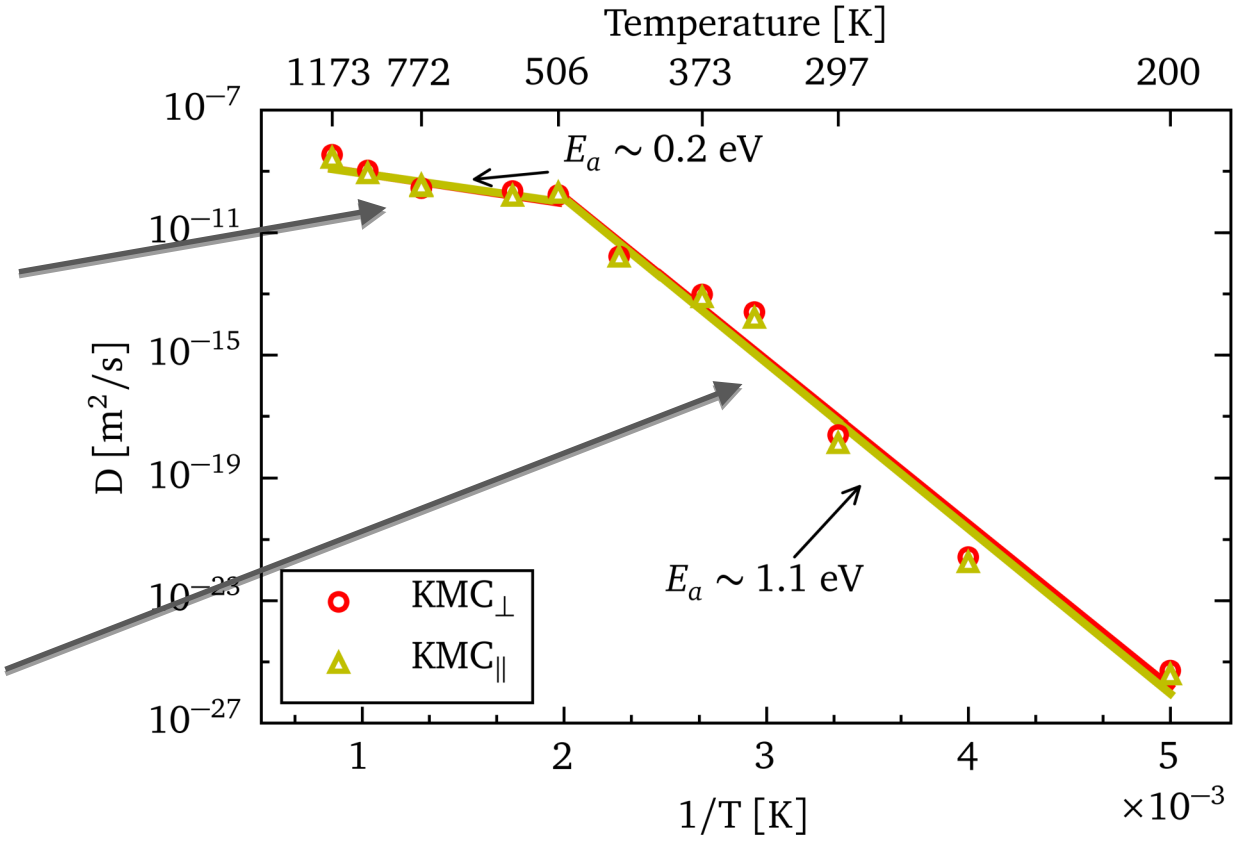




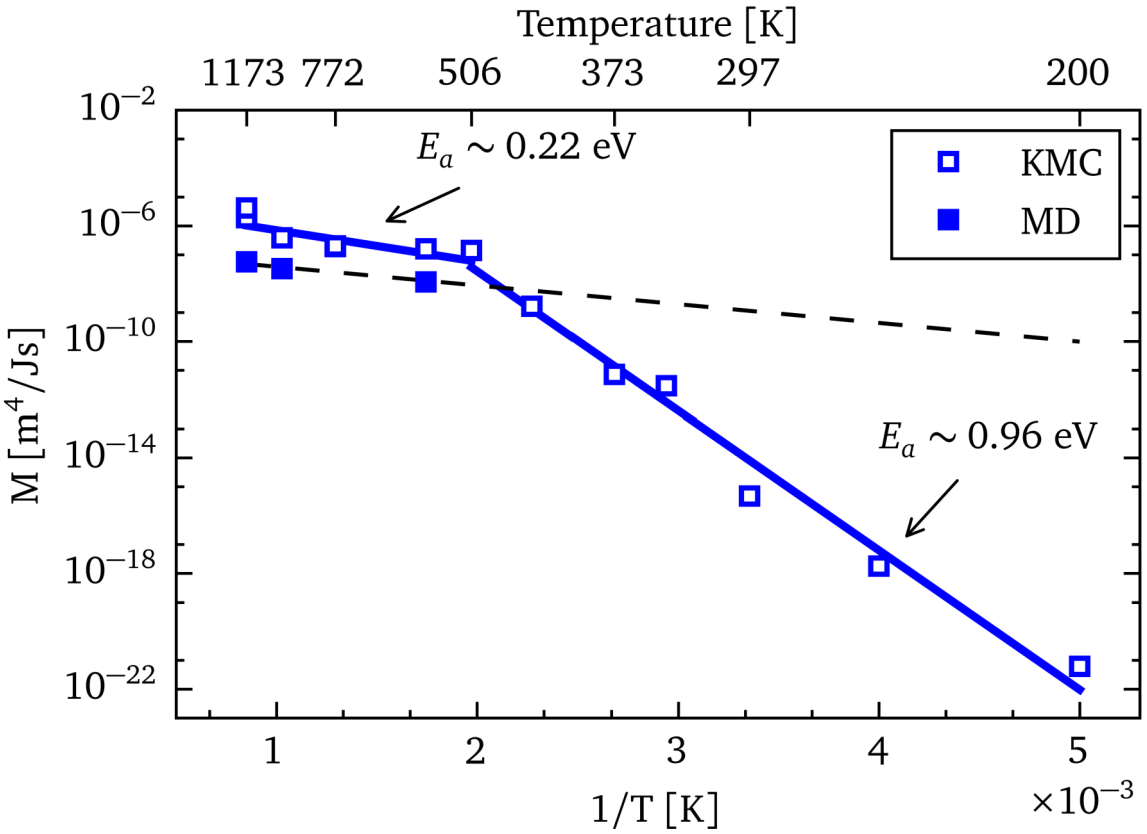
Two classes of events

At high temperatures we can do MD directly, and we get the same activation energy

At low temperatures we can't do the relevant MD: we wait forever and never access a disordered configuration



GB mobility (fluctuation-based measurement)



Again, at low temperatures, highly uncertain MD simulations miss the 2-regime behavior; At high temperatures, similar behavior is predicted.

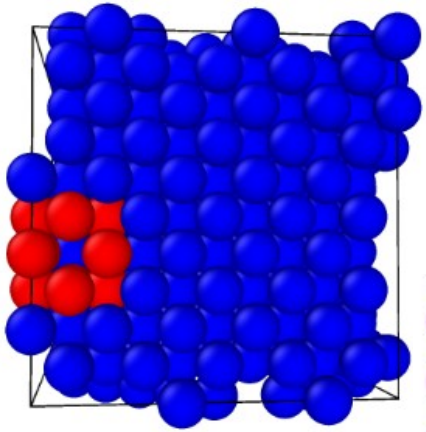
Conclusions:

With some effort, it is possible to probe the energy landscape “fully”, i.e., to an apparently acceptable level of uncertainty

full off-lattice KMC appears preliminarily viable for GBs

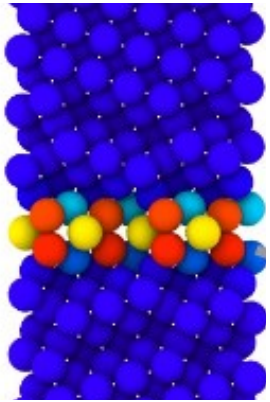
KMC captures the slow process of GB disordering as a rate limiting step for diffusion and migration at low temperatures.

vacancy



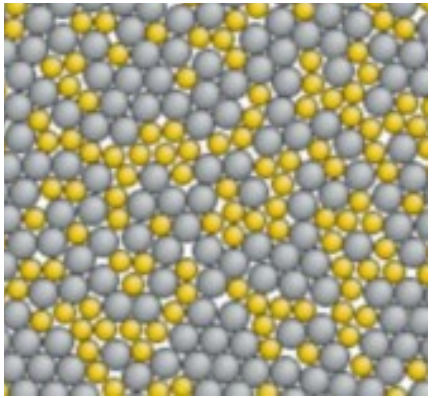
Too Easy

grain boundary



Just right?

amorphous system



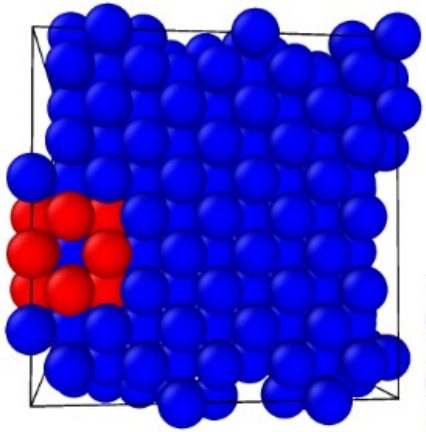
Too Hard

Implications:

We now have a tool that can utilize HPC to characterize GB kinetic behavior.

We are working to build a database of GB properties with the goal of developing GB strucutre-property relationships and ultimately pursuing the next era of GB engineering.

vacancy



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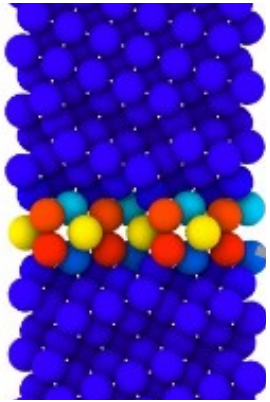

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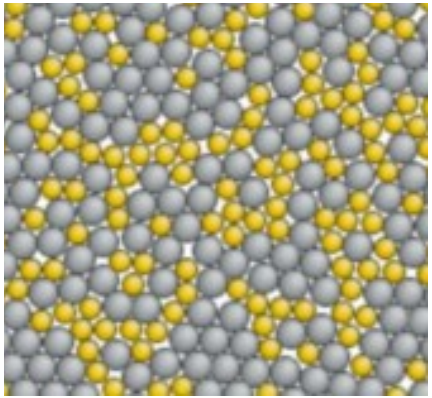

Too Hard

Acknowledgements

odge Office of the Dean for Graduate Education

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freedom to innovate

NSF

Thank You --- Questions?