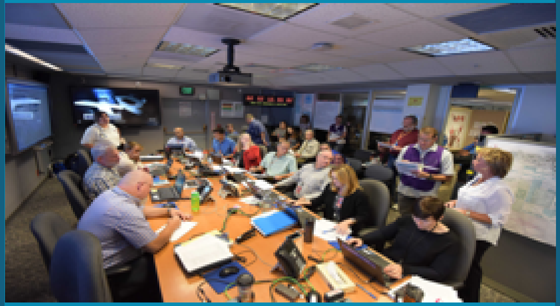
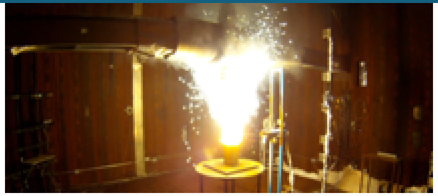
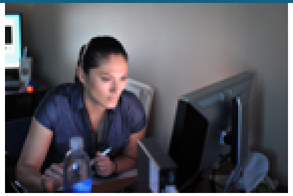
SAND2022-13560C

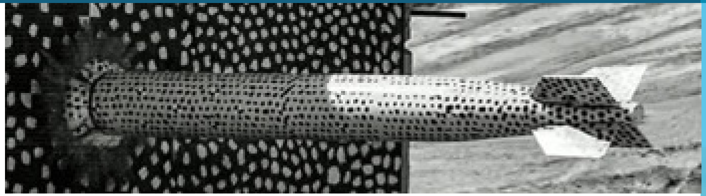
Sandia National Laboratories

Molecular Dynamics Modeling of Hydrogen and Nitrogen Implantation in Tungsten Using Machine Learned Interatomic Potentials









Mary Alice Cusentino, Megan J. McCarthy, Ember L. Sikorski, Mitchell A. Wood, and Aidan P. Thompson

10th International Conference on Multiscale Materials Modeling

October 4, 2022

U.S. DEPARTMENT OF ENERGY

NNSA

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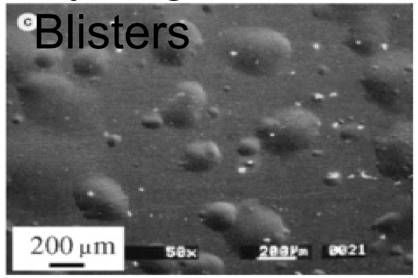
Materials for Fusion Energy

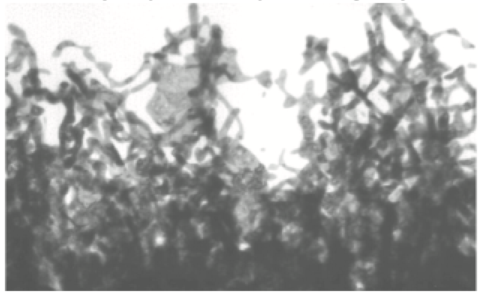
- Difficult to develop materials to handle extreme conditions within tokamak
- Large heat loads of 10-20 MW/m^3
- High particles fluxes of $\sim 10^{24}\ m^{-2}s^{-1}$ of mixed ion species (H/He/Be/N etc.)

Hydrogen Blisters



Ye, et al. J. Nucl. Mater. 313-316, 72-76 (2003)

Helium Fuzz Growth

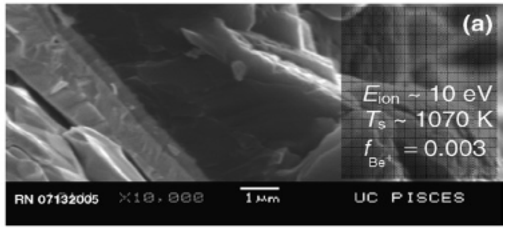


Kajita, et al. J. Nucl. Mater, 418, (2011) 152-158

W-Be Intermetallics

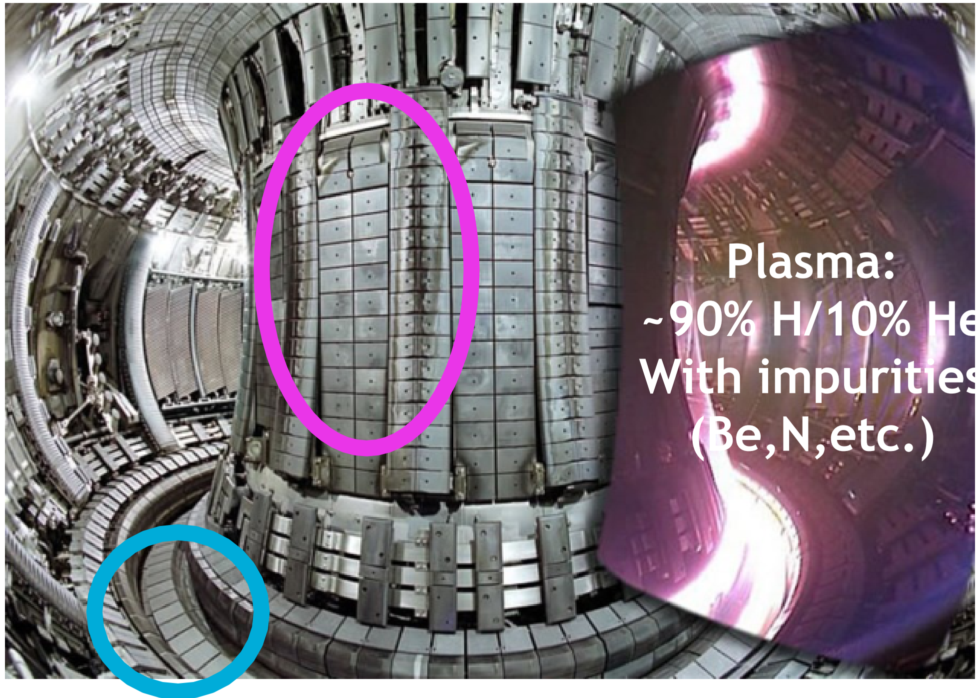
$Be_{12}W$ Be deposits (surface)

W Sub-strate



Baldwin, et. al. J. Nucl. Mater. 363-365 (2007) 1179-1183

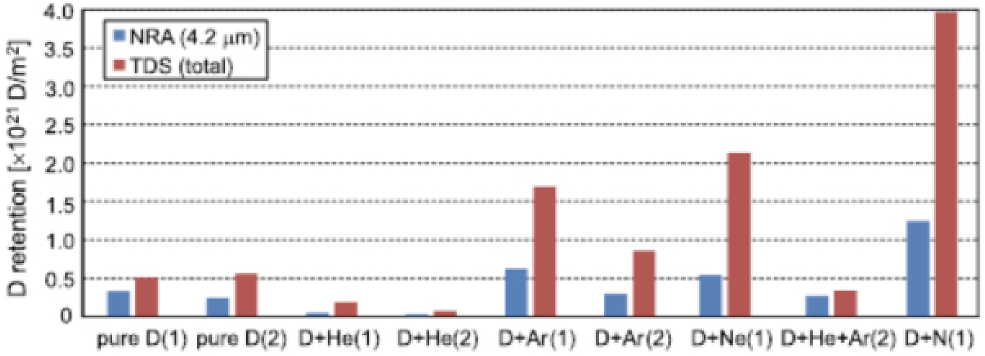
Beryllium First Wall



Tungsten Divertor

iter.org

Effect of Plasma Impurities on Hydrogen Retention



Kreter, et al. Nucl. Fus.. 59, 086029 (2019)

Materials for Fusion Energy

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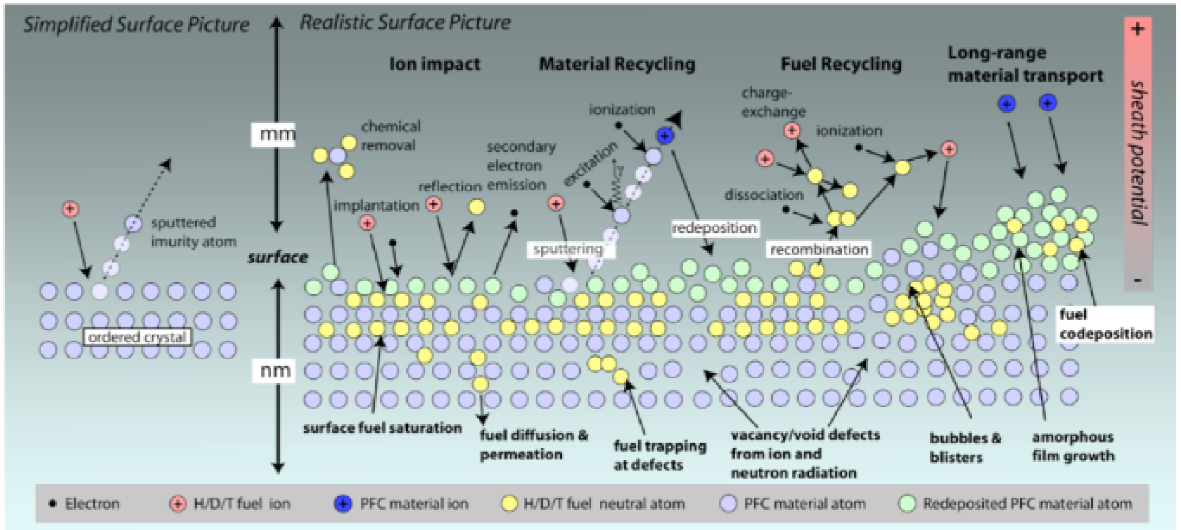
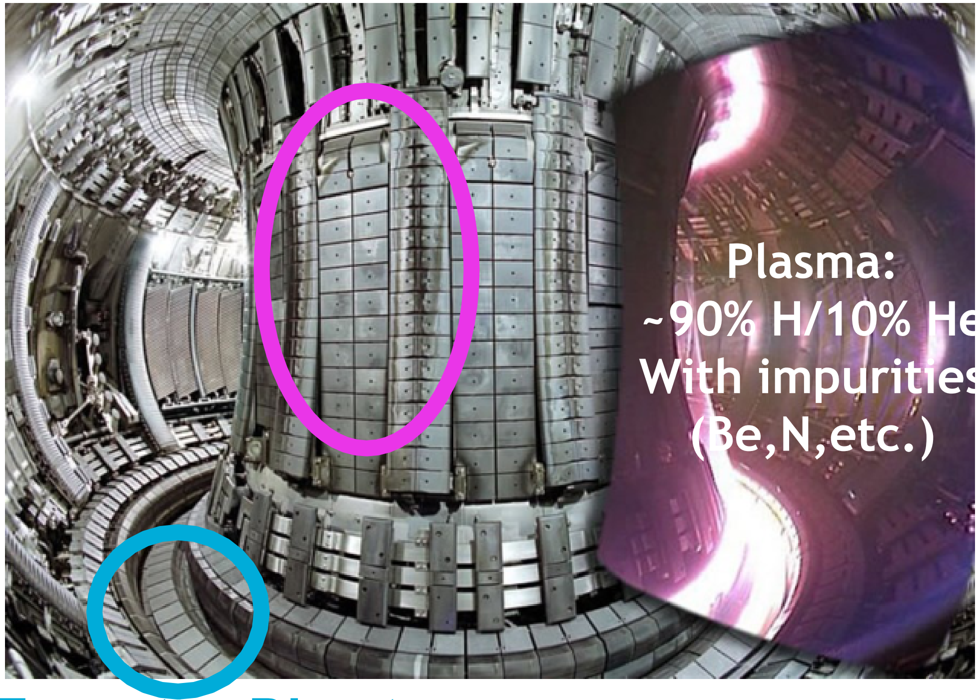


Wirth, et al. MRS Bulletin 36 (2011) 216-222

Beryllium First Wall



Tungsten Divertor

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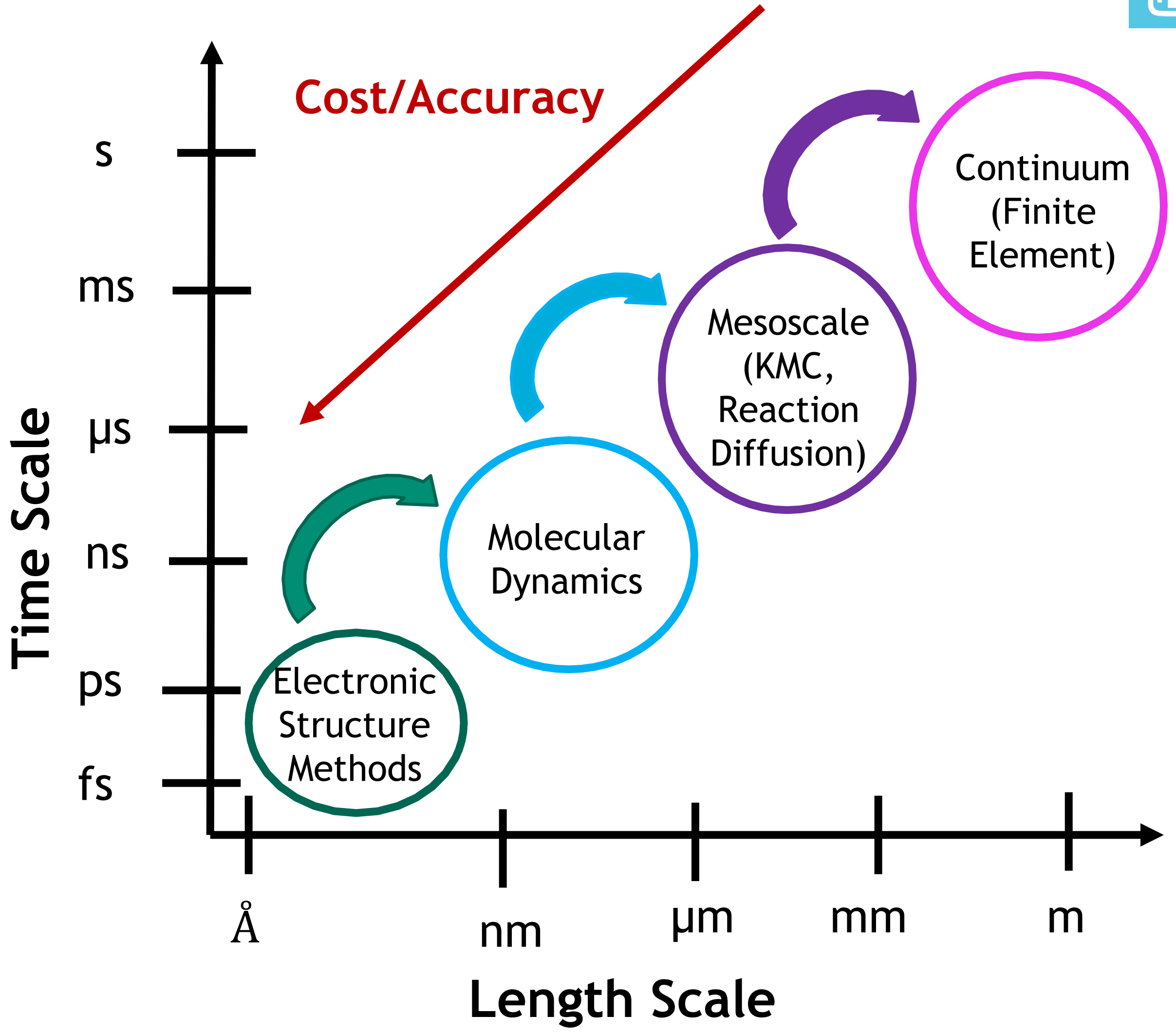
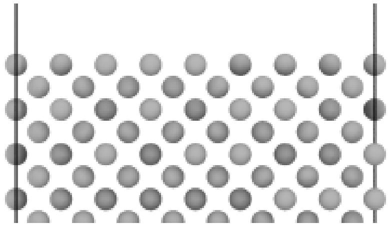
- Many complex processes that occur at the plasma/material interface that can lead to material degradation

Multiscale Modeling of Materials

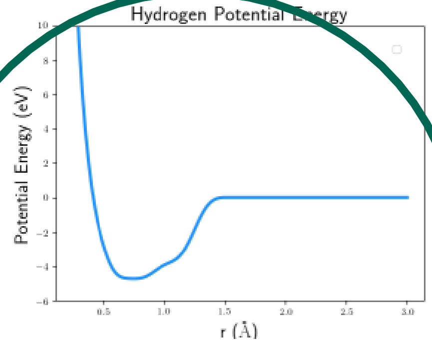


Each simulation technique can provide information to the next scale up

Multiscale Modeling of Materials



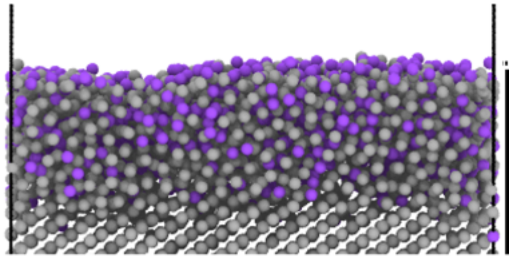
Initial Atom Positions and Velocities

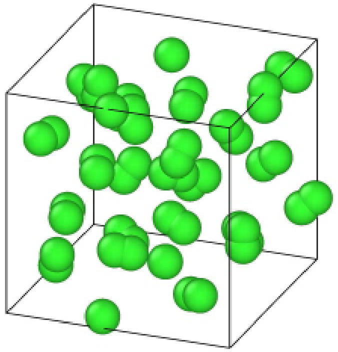


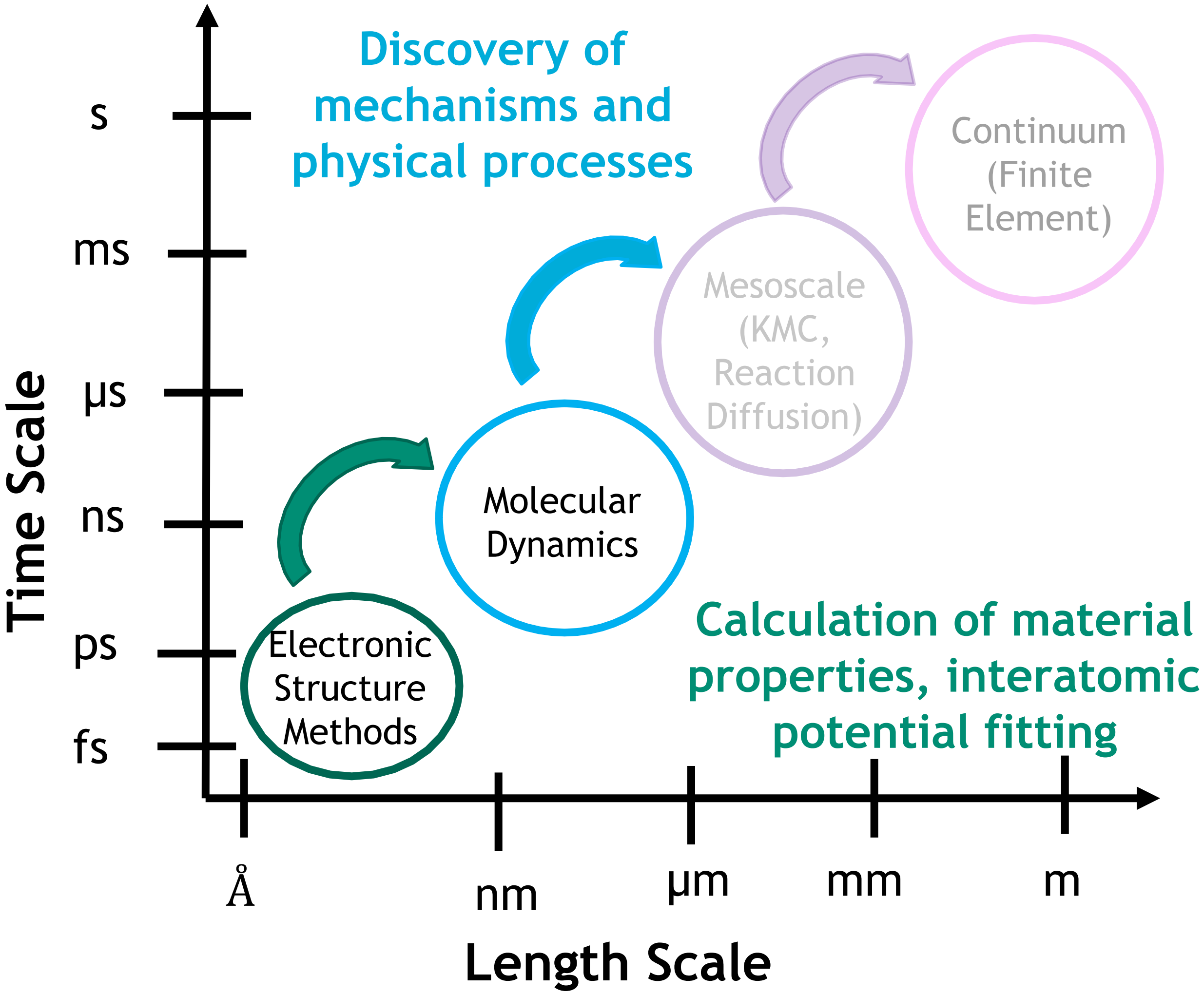
Interatomic Potential

Molecular Dynamics (LAMMPS)

Evolution of Material System Over Time



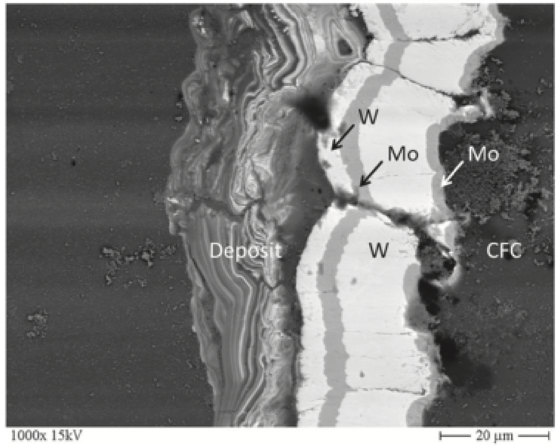




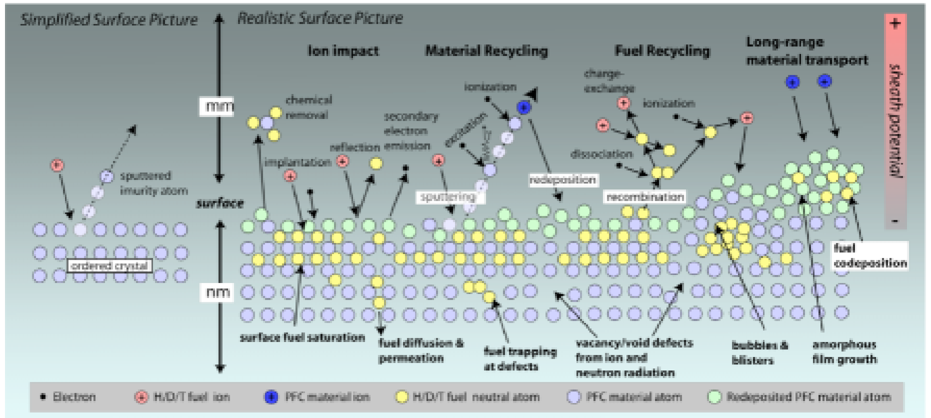
Each simulation technique can provide information to the next scale up

Why do we need ML-IAPs?

We want to model very complex physics at the plasma-material interface.



M Mayer *et al* 2016 *Phys. Scr.* **2016** 014051



Wirth, et al. MRS Bulletin 36 (2011)

Electronic Structure Methods

- Highly accurate
- Can model a lot of relevant physics
- Very expensive, $O(N^3)$ scaling, ~100 atoms

Classical Potentials

- Lots of functional forms that are good for many different materials
- Scales well
- Accuracy highly dependent on potential and application
- Functional form limits type of physics that can be modeled

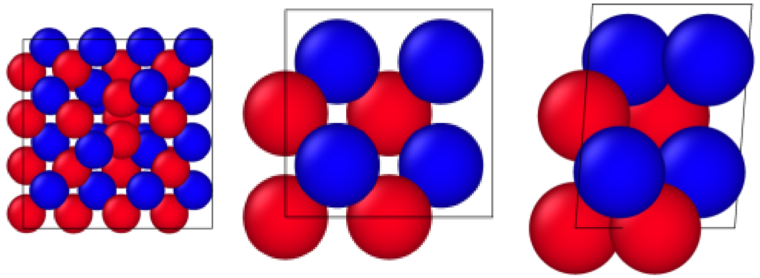
Machine Learned Interatomic Potentials

- Trained to electronic structure data for increased accuracy
- Flexible, not limited by inherent physics of model
- Quantum accuracy but MD scalability
- Need good training data for accurate model

What Makes a Machine Learned Interatomic Potential?

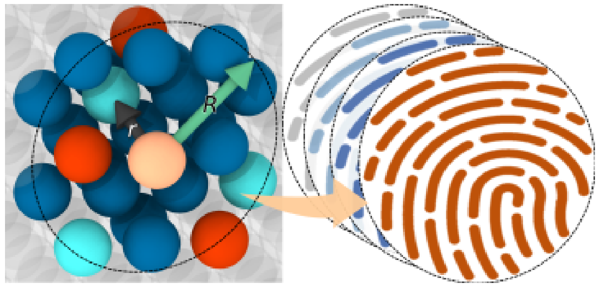
Training Data

- Generated using quantum methods
- Can include:
  - Energies
  - Forces
  - Stresses
- Variety of atomic configurations
  - Bulk structures, liquids, surfaces, defects, etc.



Descriptor

- Describes the local atomic environment
- Requirements
  - Rotation/Translation/. Permutation invariant
  - Equivariant forces
  - Smooth differentiable
  - Extensible
- Some Examples
  - Bispectrum, SOAP, ACE, Moment Tensors, etc.



Regression Method

- Linear regression
- Kernel ridge regression
- Gaussian process
- Non-linear optimization
- Neural Networks

SNAP

- Energies, forces, and stresses from DFT
- Bispectrum component descriptors
- Linear regression

SNAP Definition and Work Flow

Model Form

- Energy of atom i expressed as a basis expansion over K components of the bispectrum (B_k^i)

$$E_{SNAP}^i = \beta_0 + \sum_{k=1}^{K} \beta_k (B_k^i - B_{k0}^i)$$

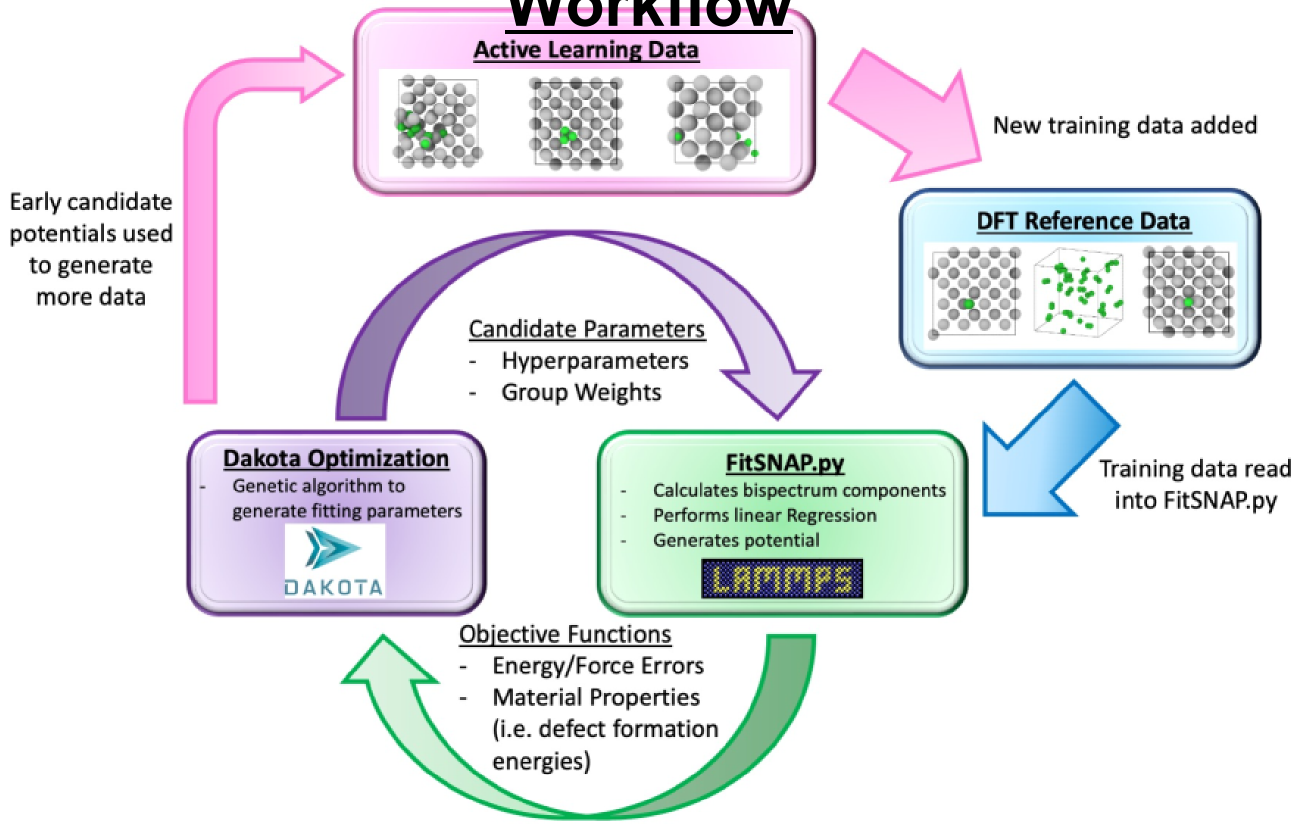
Regression Method

- β vector fully describes a SNAP potential
- Decouples MD speed from training set size

$$\min(||\mathbf{w} \cdot D\boldsymbol{\beta} - T||^2 - \gamma_n \, ||\boldsymbol{\beta}||^n)$$

Weights   Set of Descriptors   DFT Training

SNAP Development Workflow

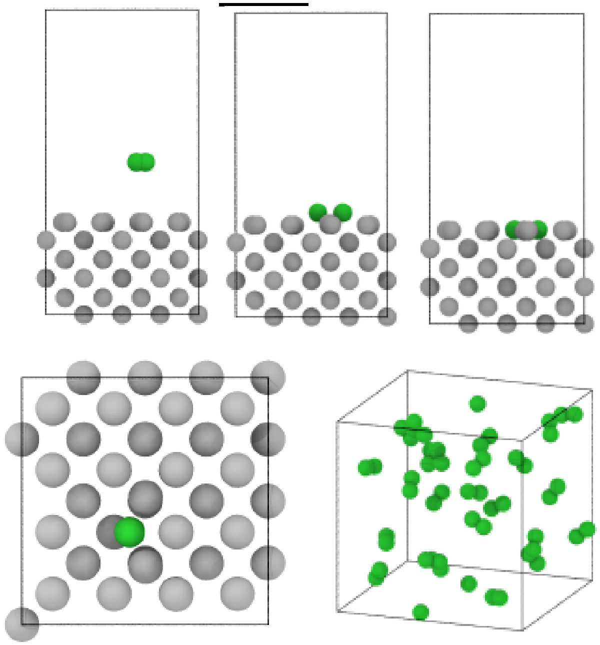


Code available: https://github.com/FitSNAP/FitSNAP

M. A. Wood, M.A. Cusentino, B.D. Wirth and A.P. Thompson, Phys. Rev. B 99, 184305

SNAP Development for W-H

Training Data



Fitted Properties

- Energy/Force errors
- H defect formation energies
- H dimer/trimer binding curves

Defect Formation Energies

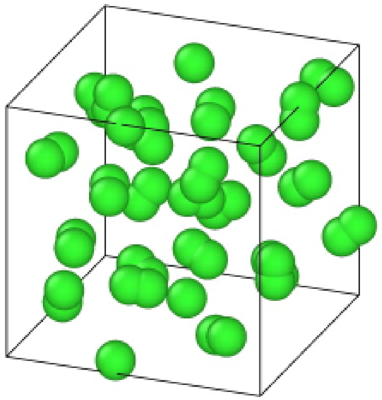
Bulk Defects	DFT (eV)	SNAP (eV)
E_f^{Tet}(eV)	0.88	0.91
E_f^{Oct}(eV)	1.26	1.24
E_f^{Sub}(eV)	4.08	4.42
E_f^{H2}(eV)	-4.74	-4.77

Adsorption Energies

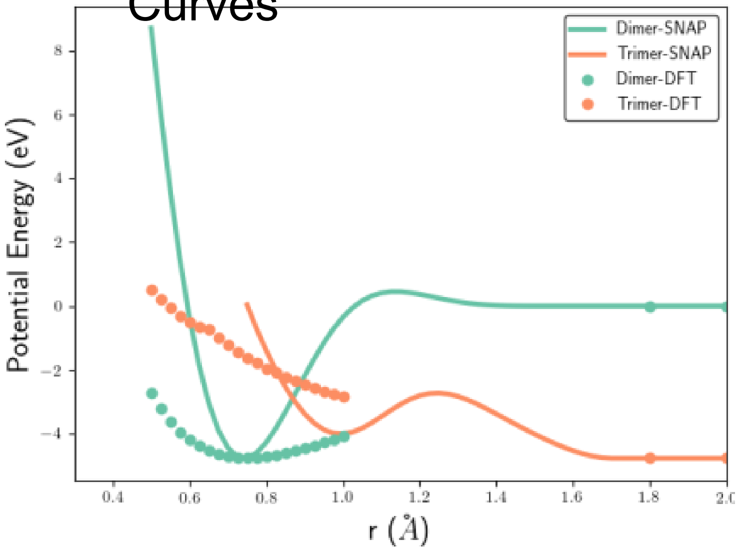
H Ads.	DFT (eV)	SNAP (eV)
(100) Ads. Site	Bridge	Bridge
(100) Ads. Energy	-0.96	-2.41
(100) H_2 Ads. Energy	-0.80	-4.99
(110) Ads. Site	Hollow	Hollow
(110) Ads. Energy	-0.75	-2.64
(111) Ads. Site	Bridge	Hollow
(111) Ads. Energy	-0.59	-3.34

Initial Results

H_2 at 1000 K



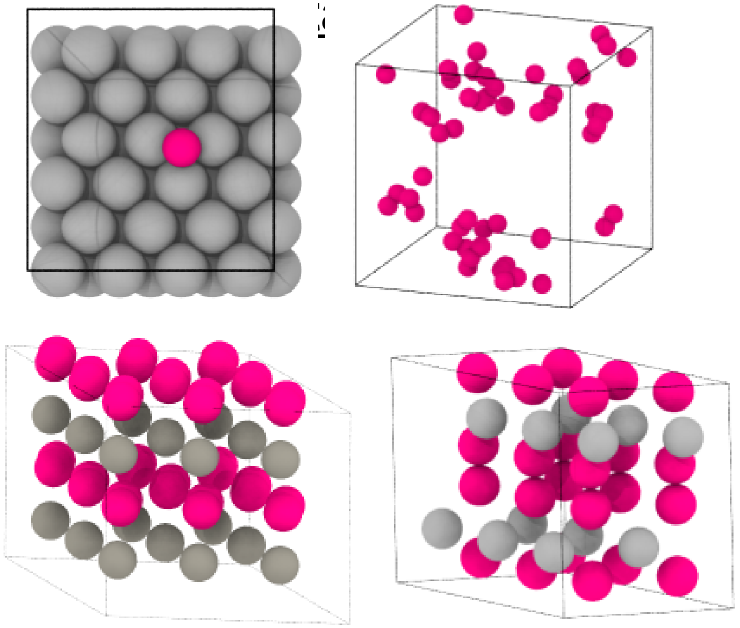
Hydrogen Binding Curves



W: Grey H: Green N: Pink

SNP Development for W-N

Training



Fitted Properties

- Energy/Force errors
- N defect formation energies
- N adsorption energies
- N dimer/trimer binding curves
- W_xN_y cohesive energies

Defect Formation Energies

Bulk Defects	DFT (eV)	SNAP (eV)
	1.85	1.89
	1.11	1.09
	4.72	2.90
	-9.79	-9.47

Surface Adsorption

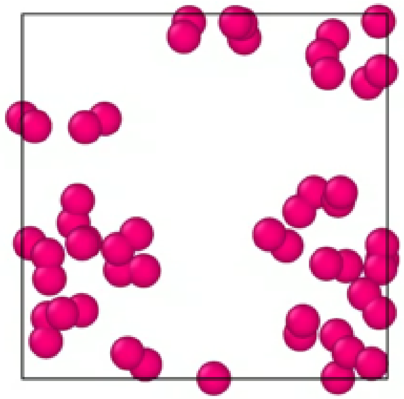
	DFT (eV)	SNAP (eV)
(100) Ads. Site	Hollow	Hollow
(100) Ads. Energy	-3.52	-4.33
(110) Ads. Site	Hollow	Hollow
(110) Ads. Energy	-3.59	-2.97

W_xN_y Formation

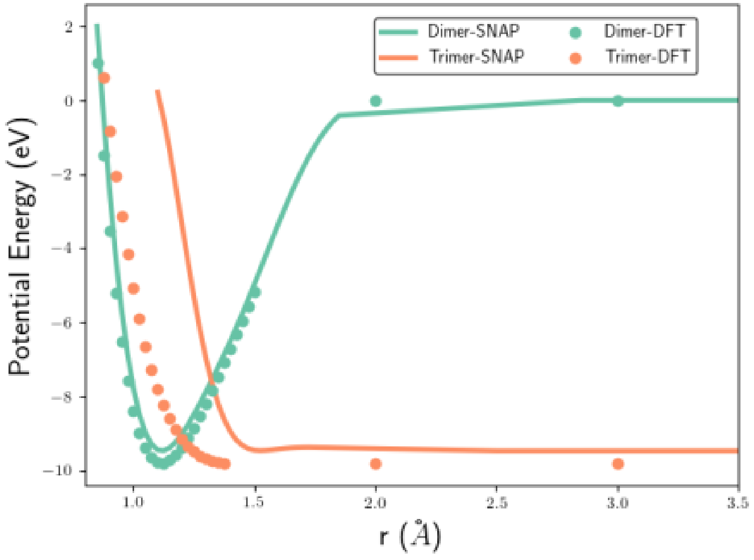
	DFT (eV)	SNAP (eV)
WN_2- P62mmc	-1.82	-1.82
WN_2- P6m2	-0.91	-1.75
WN - NiAs	-0.84	-0.74
WN - WC	-0.23	-1.51
W_2N	-0.03	3.29

Initial Results

N_2 at 1000 K



Nitrogen Binding



W: Grey H: Green N: Pink

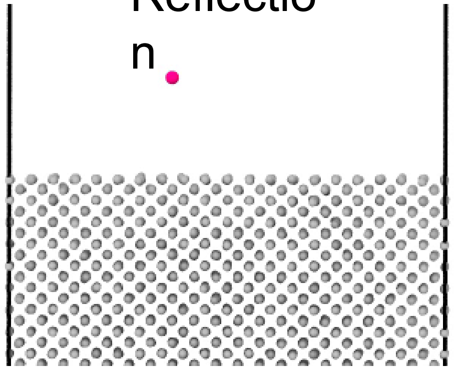
Single Ion Implantations in Tungsten

W: Grey H: Green N: Pink

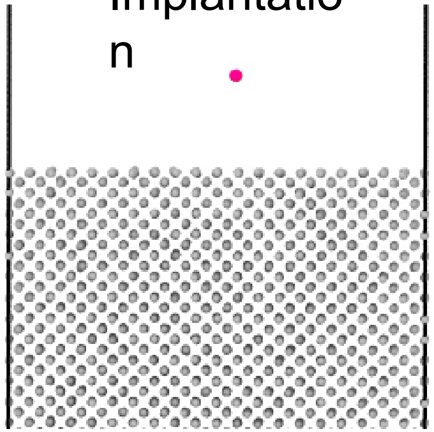
Single ion implantations to assess depth profile

- 10 eV, 75 eV, and 100 eV hydrogen or nitrogen for 1,000 separate implantations
- 1000 K for (100) tungsten surface
- Simulations performed for 2 ps and depth is recorded

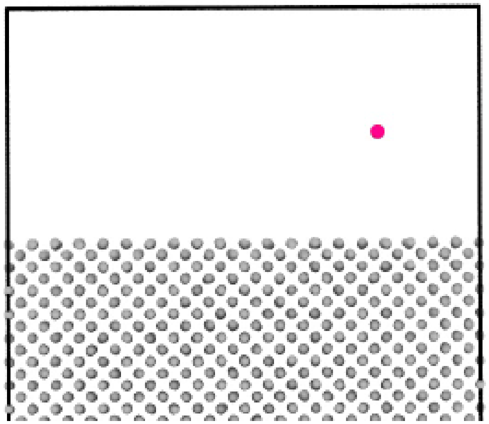
Reflection



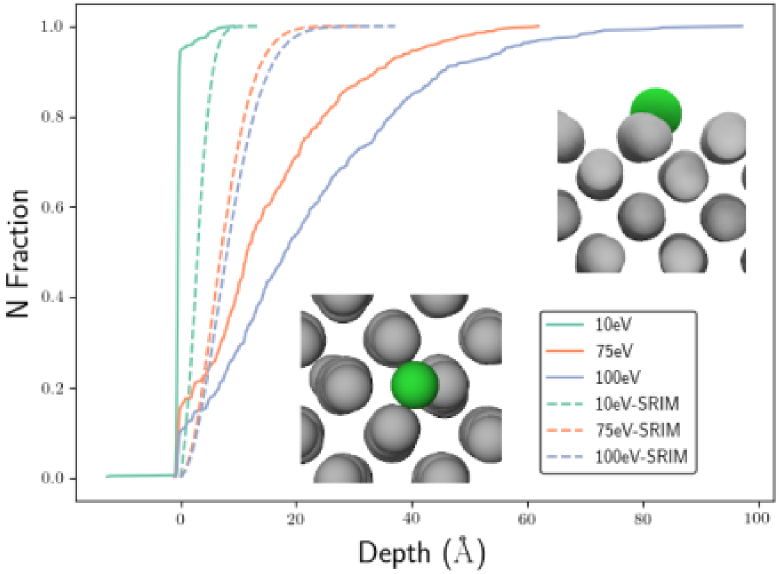
Implantation



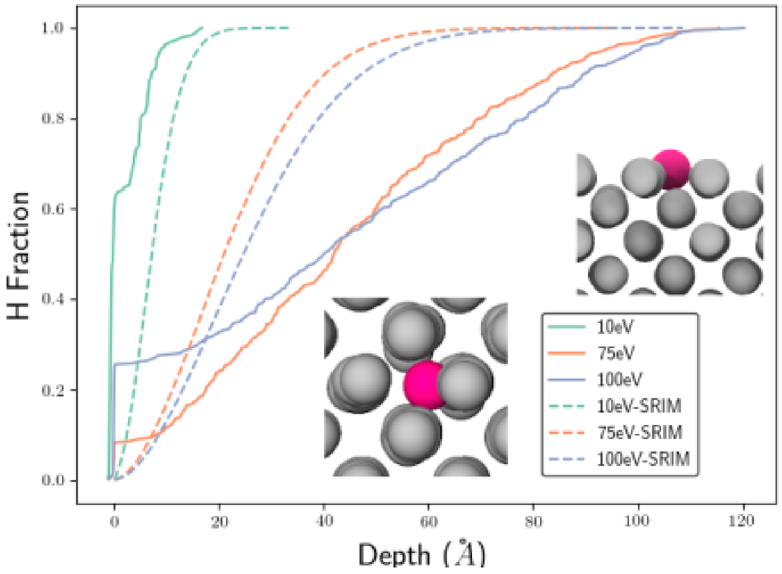
Surface Adsorption



Hydrogen Depth Profile



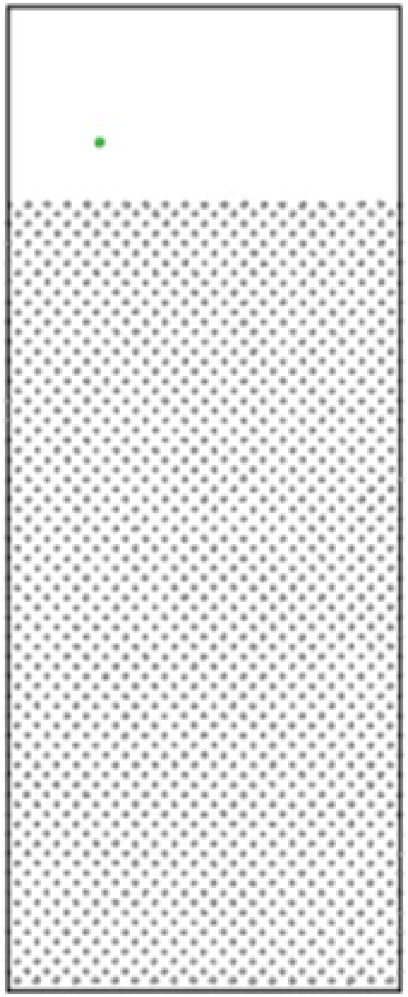
Nitrogen Depth Profile

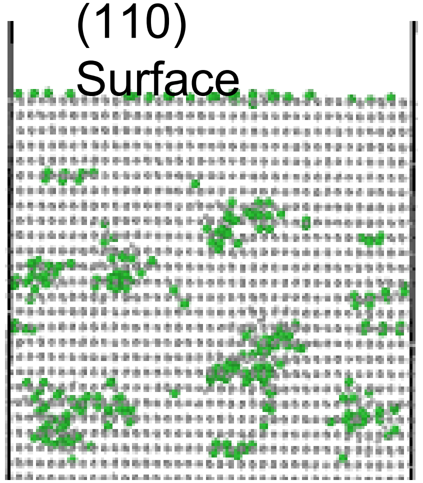


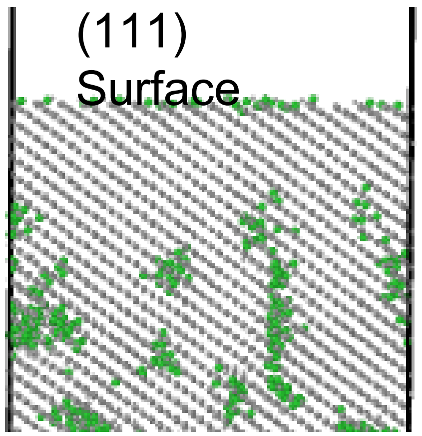
Cumulative Implantation Simulations - Hydrogen

W: Grey H: Green

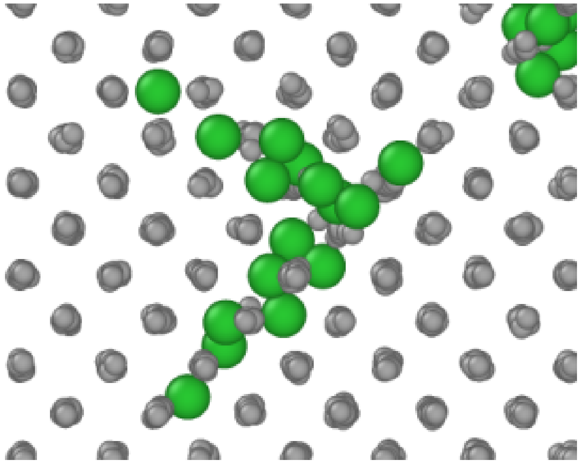
Hydrogen onto (100) Tungsten

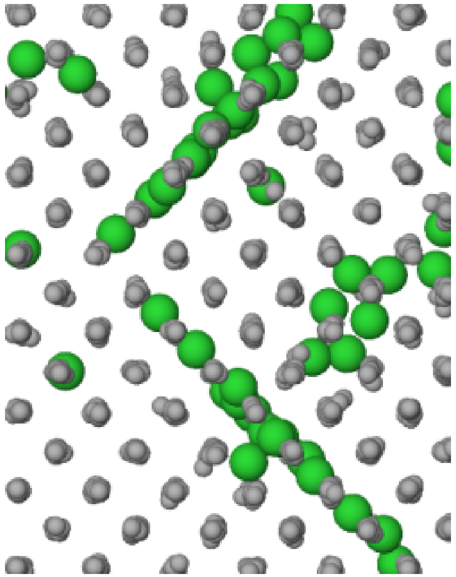




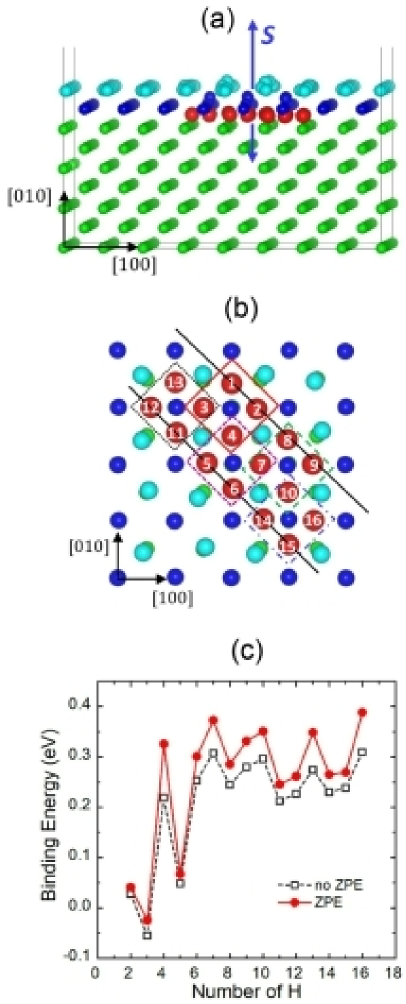


Formation of 2D Hydrogen Platelets





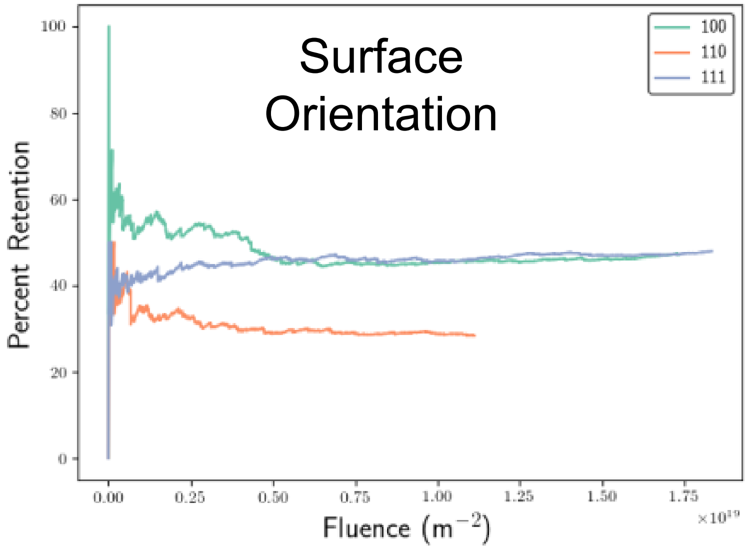
2D Hydrogen Clusters Predicted by DFT

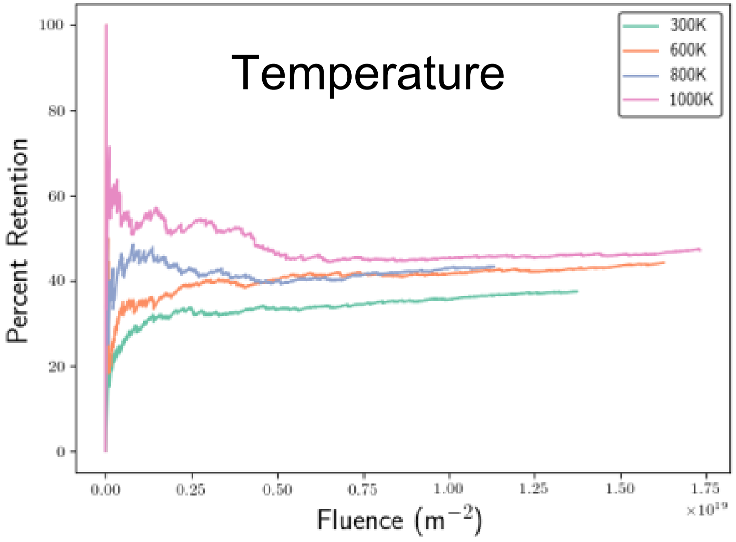


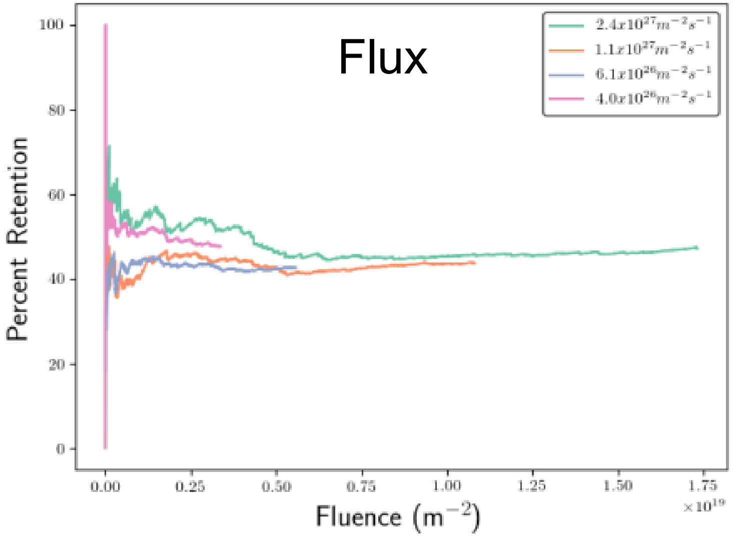
Yang and Wirth, J. Appl. Phys. 125, 16510 (2019)

Hydrogen Retention in Tungsten

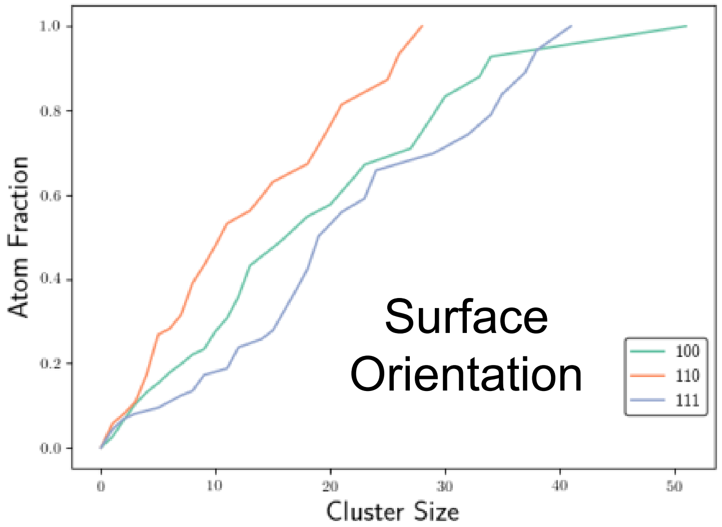
Hydrogen Retention

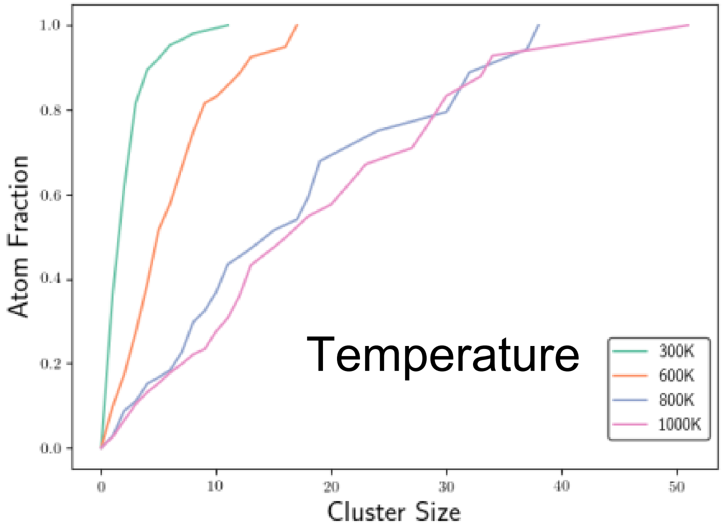


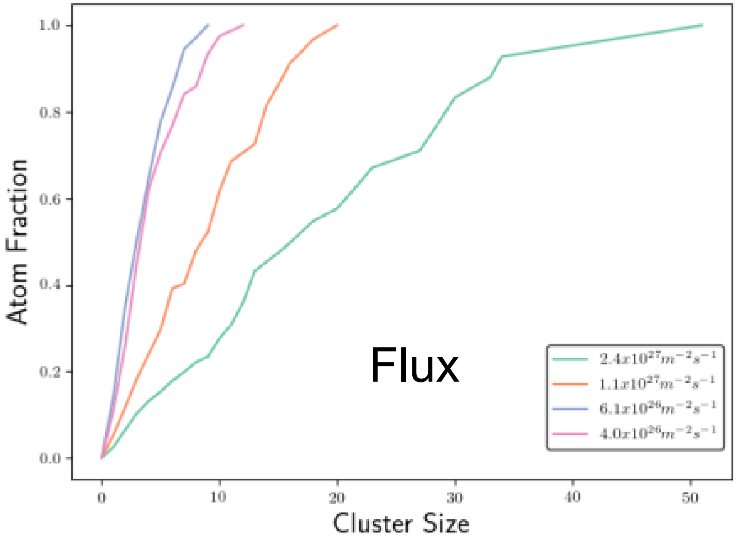




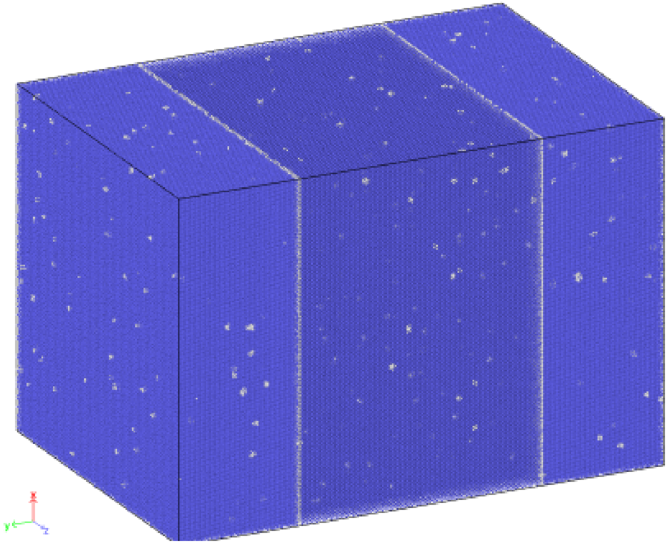
Cluster Distributions



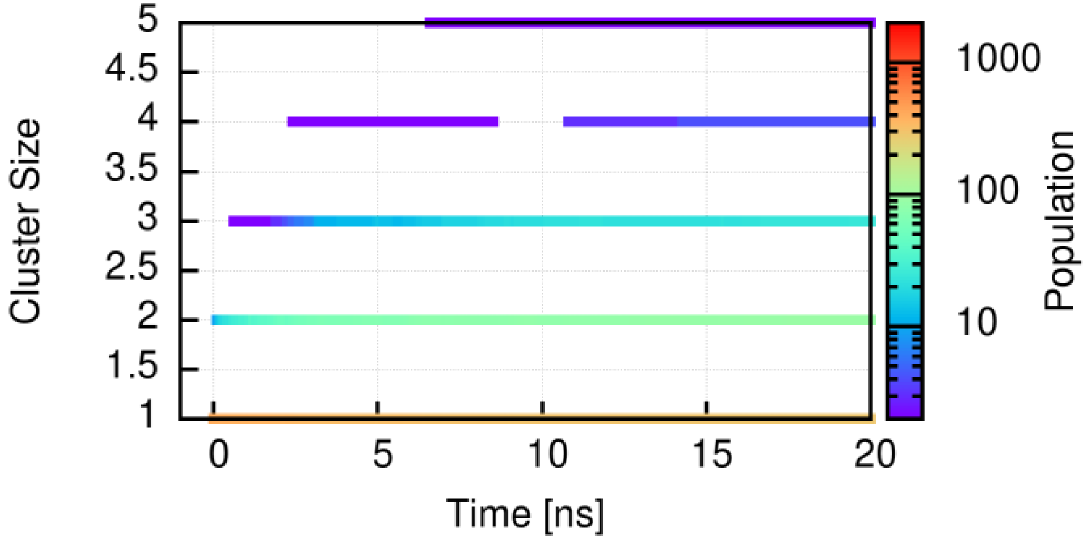




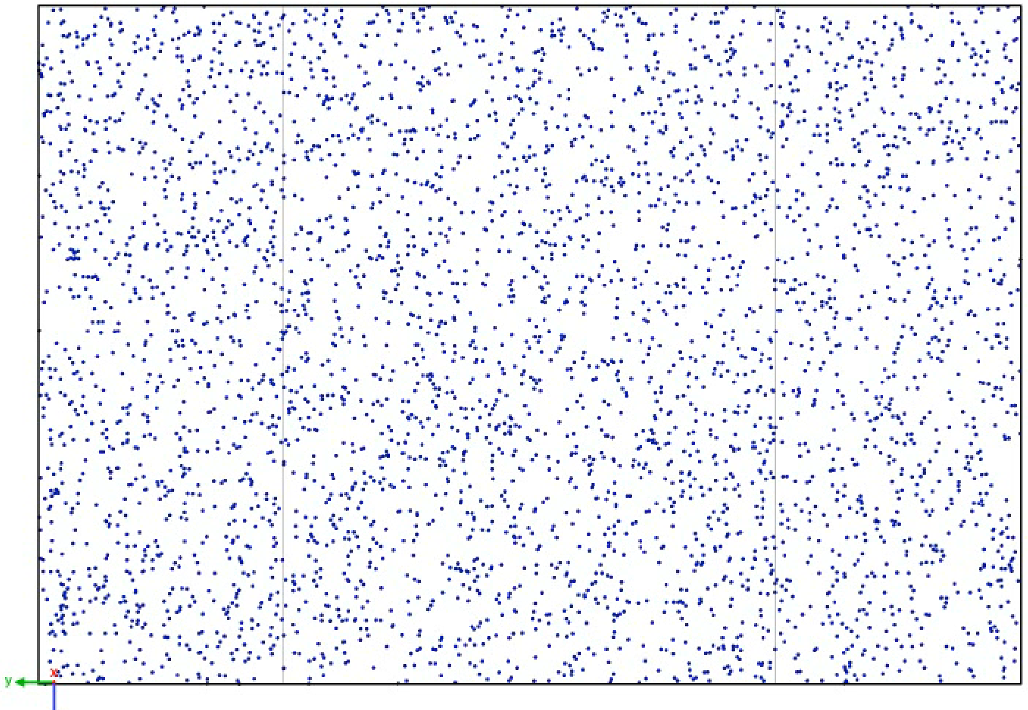
Large-Scale Simulations of Hydrogen Trapping At Grain

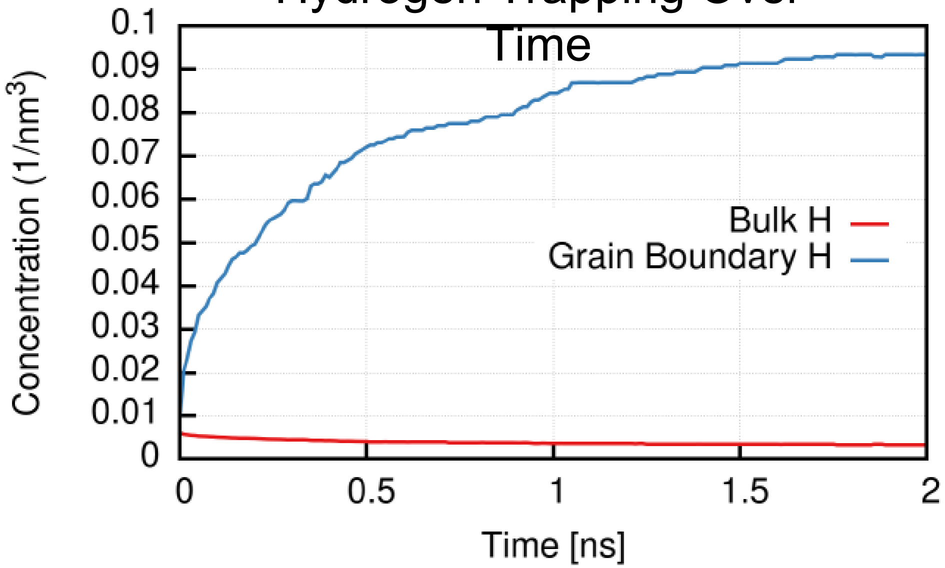


Modeling 0.01% H concentration in twin grain boundary



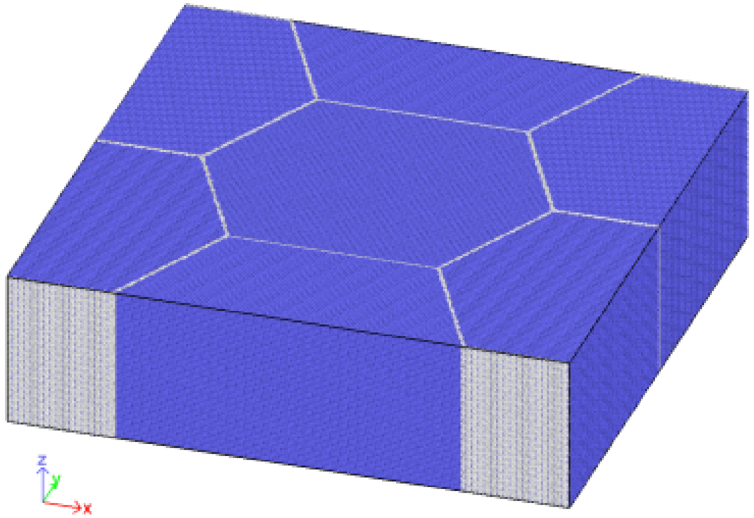
4 ns of hydrogen diffusion



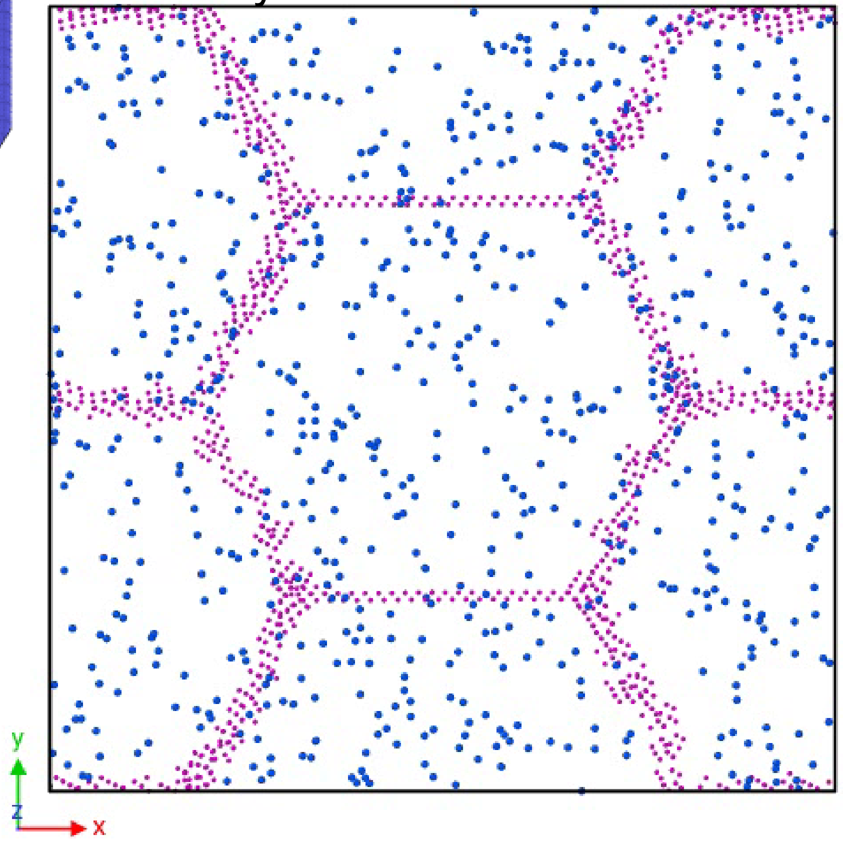


Large-Scale Simulations of Hydrogen Trapping At Grain Boundaries

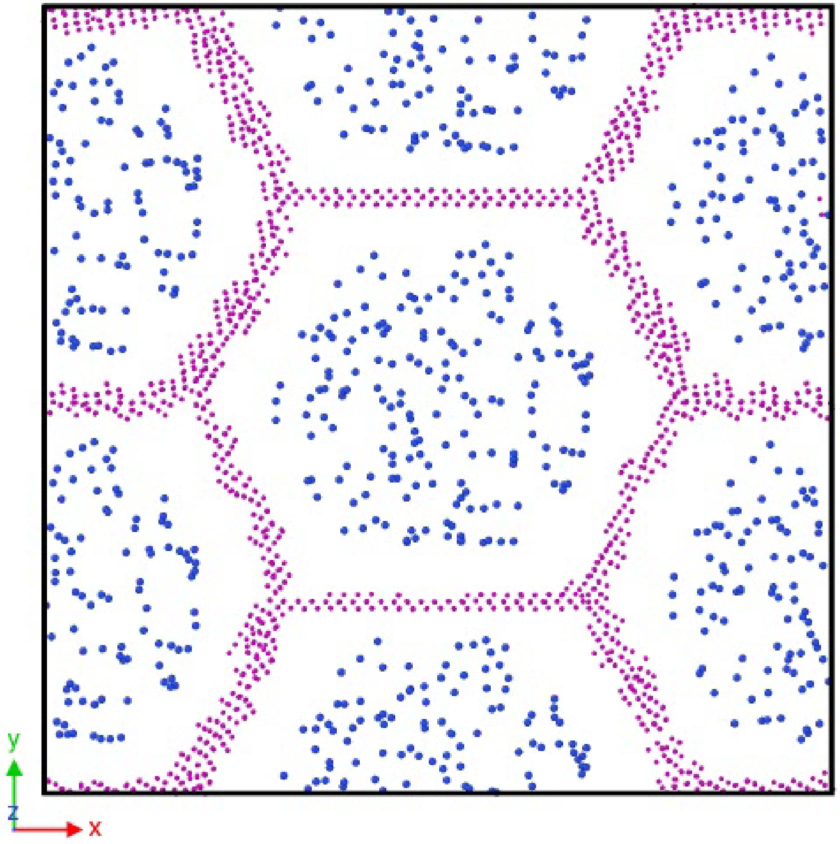
Polycrystalline Tungsten



Hydrogen Seeded Everywhere



Hydrogen Seeded in Grain

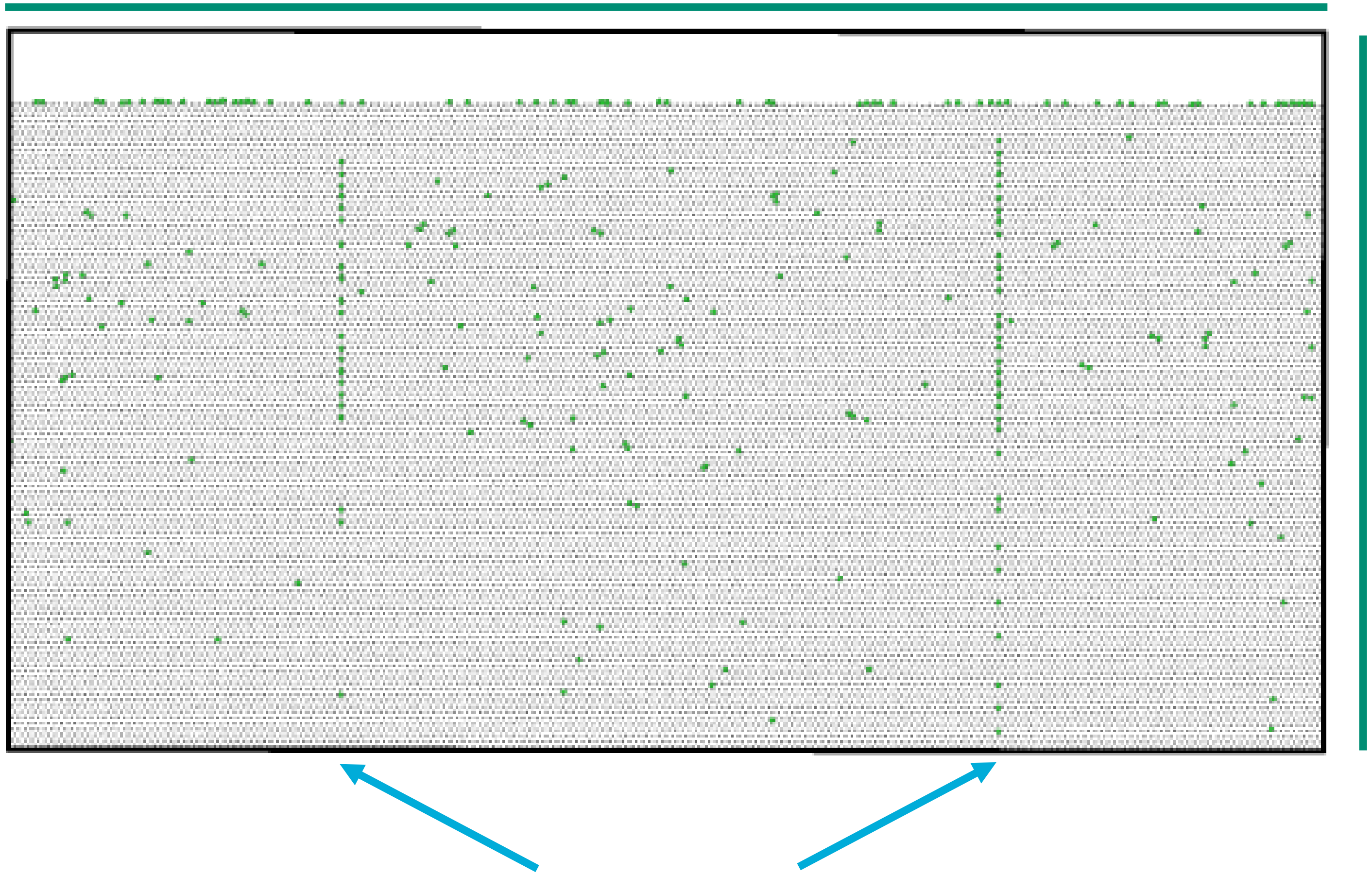


Competitions of hydrogen trapping at the GB or in bulk?

Large-Scale Simulations of Hydrogen Trapping At Grain Boundaries

Hydrogen Implantation in Twin GB at 12 ns

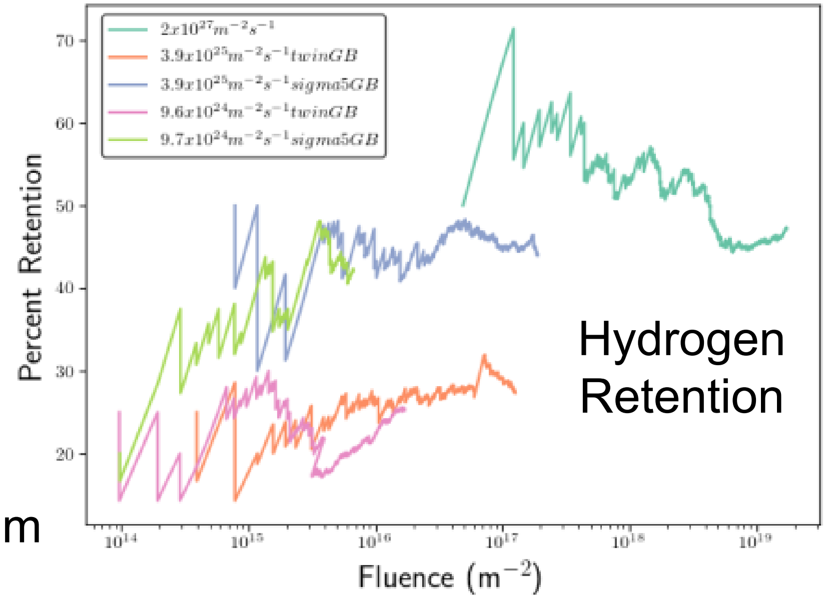
50 nm

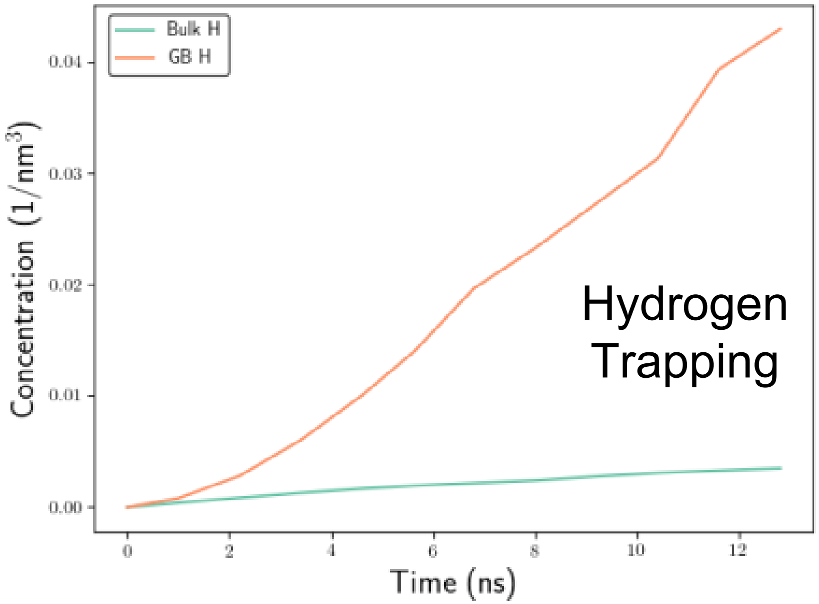


25 nm

Twin Grain Boundary

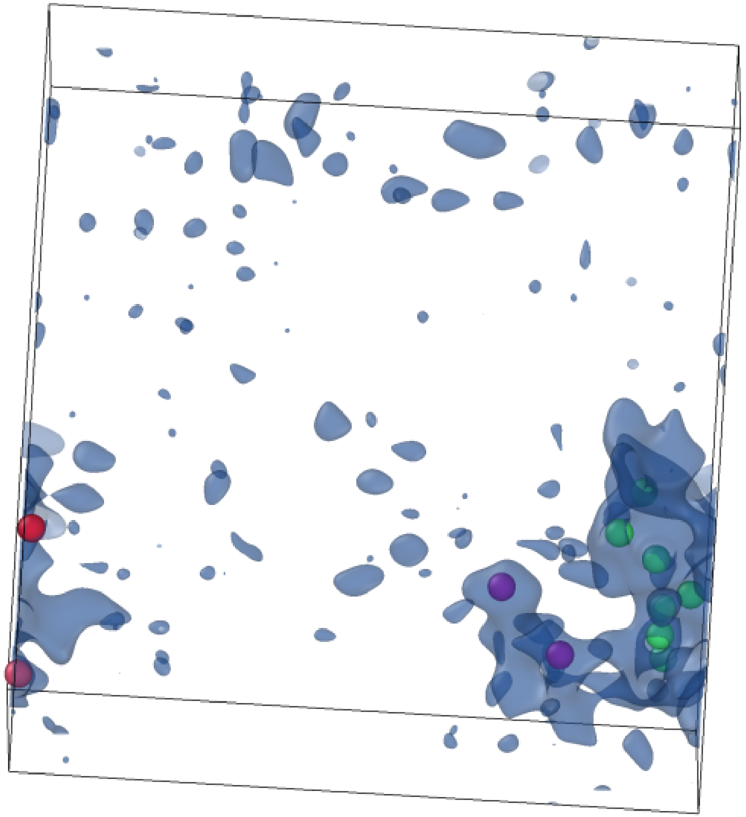
W: Grey H: Green





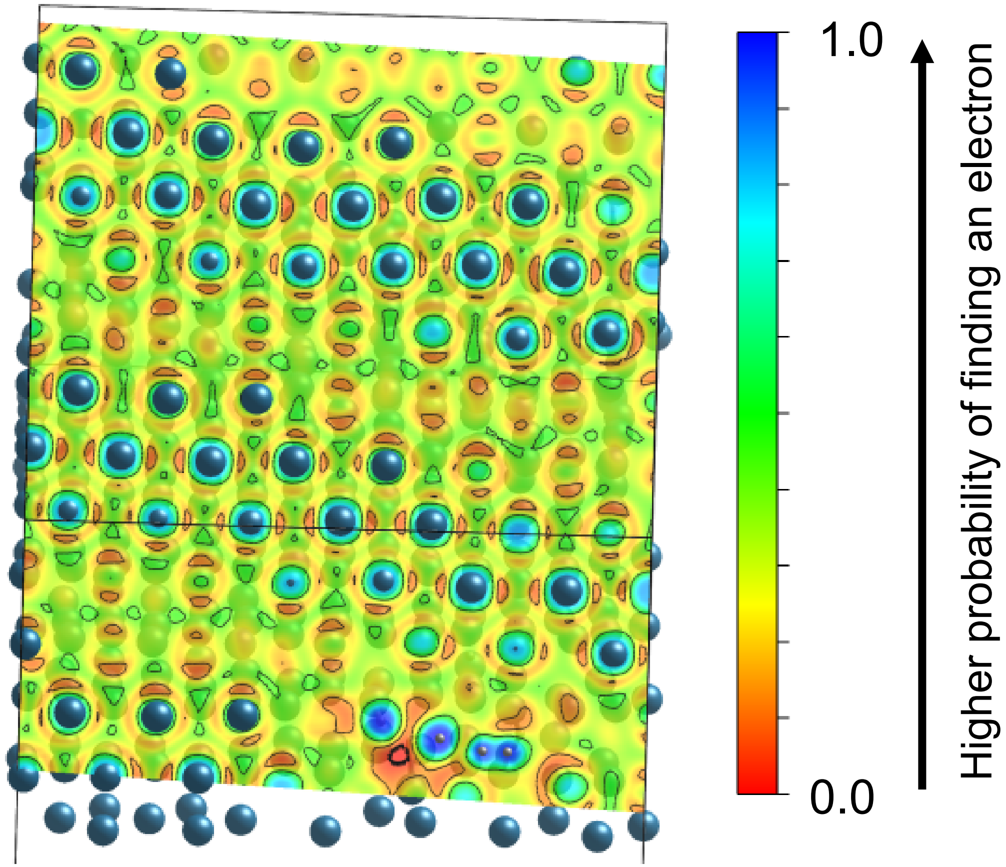
Does DFT Support Platelet Formation?

Partial Charge Distribution -5 to E_F
(**energy resolved** electron visualization)



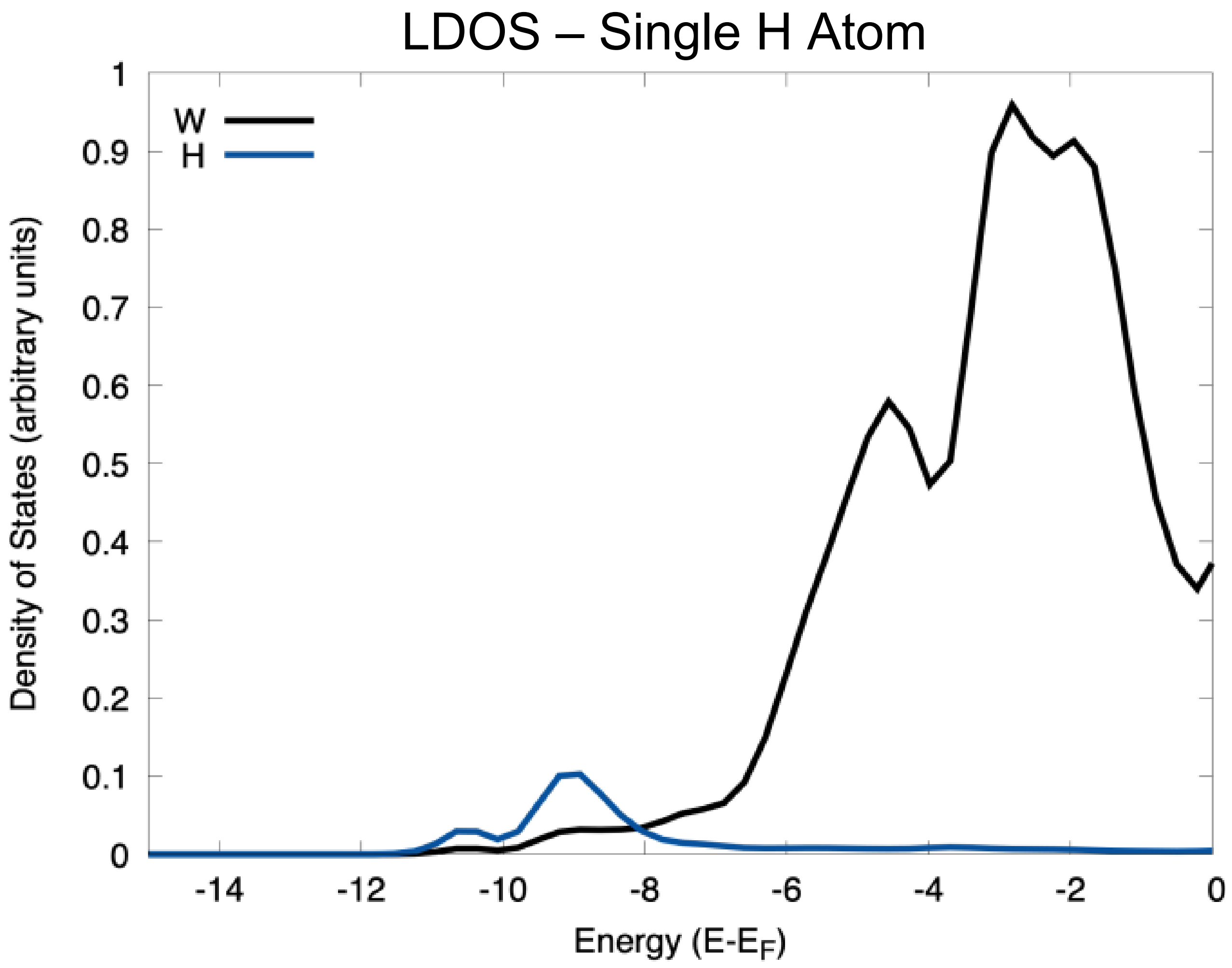
7x7x7 W supercell with 11H; 123 fs

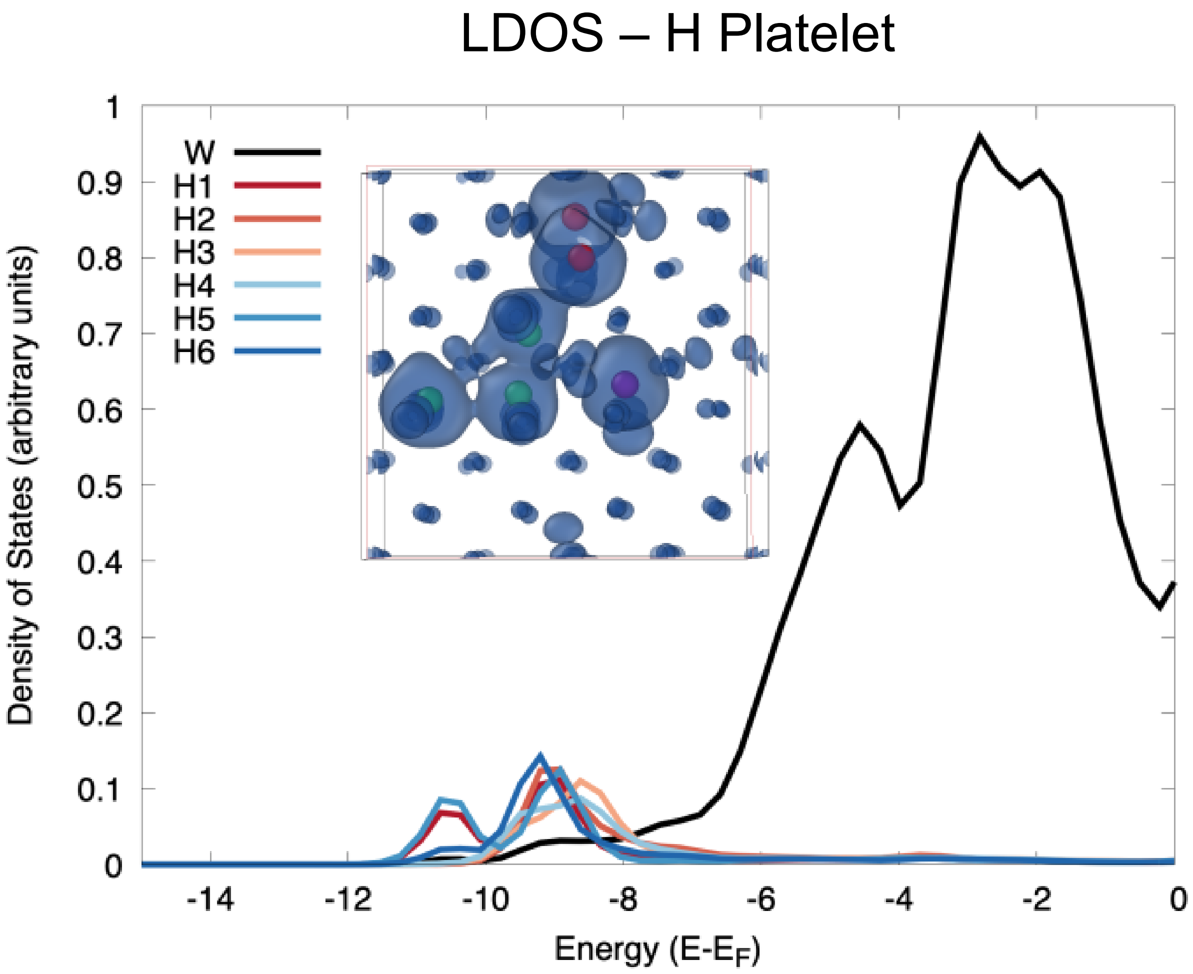
Electron Localization Function
(visualization of **all calculated electrons**)



Hydrogen atoms can share there electrons out to at least 4.2 Å

Does DFT Support Platelet Formation?



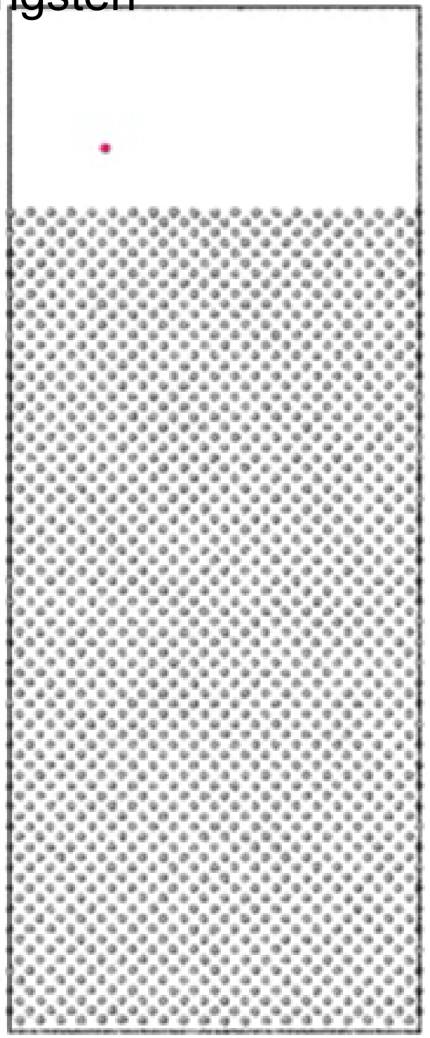


Additional hydrogen atoms continue to accumulate electrons in region near platelet

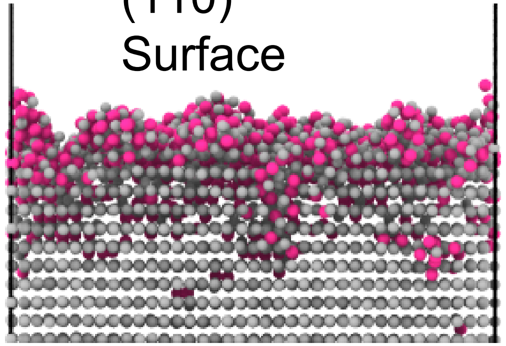
Implantation simulations

W: Grey N: Pink

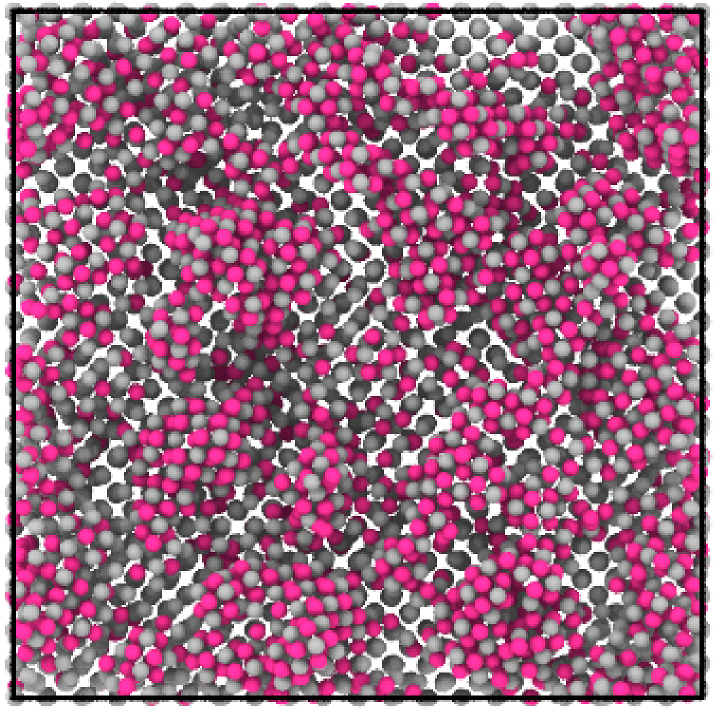
Nitrogen onto (100) Tungsten

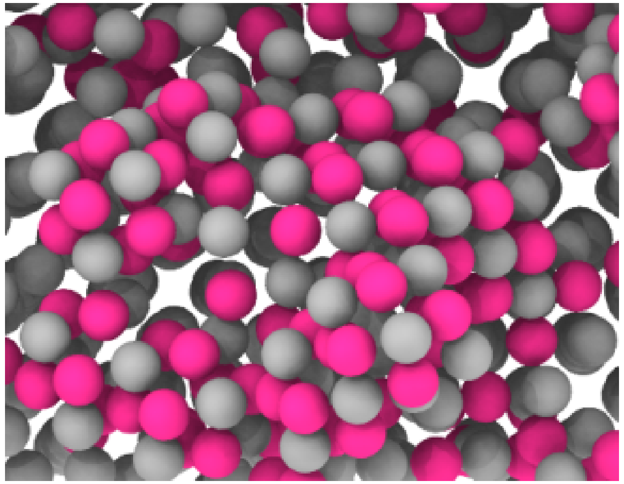


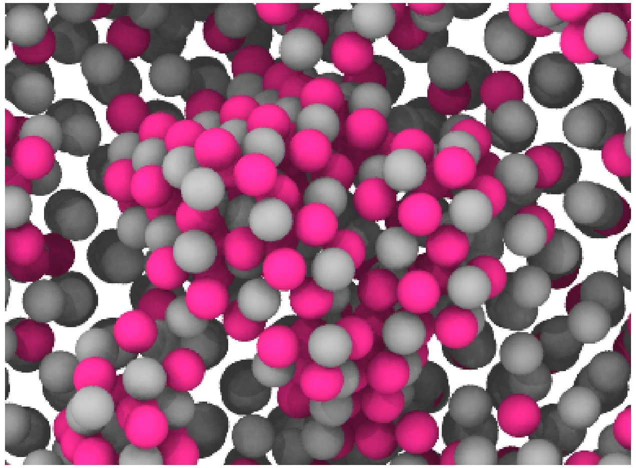
(110) Surface



Ordered Structures Emerging on Surface



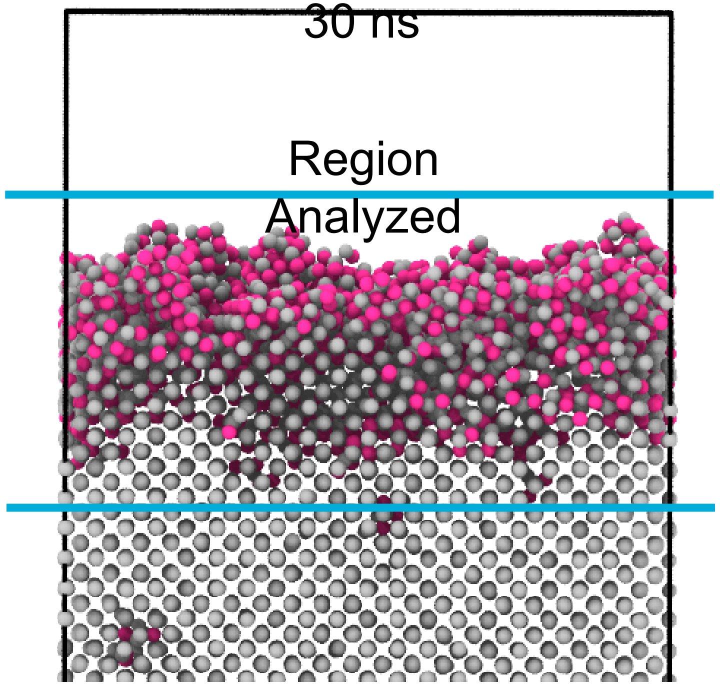




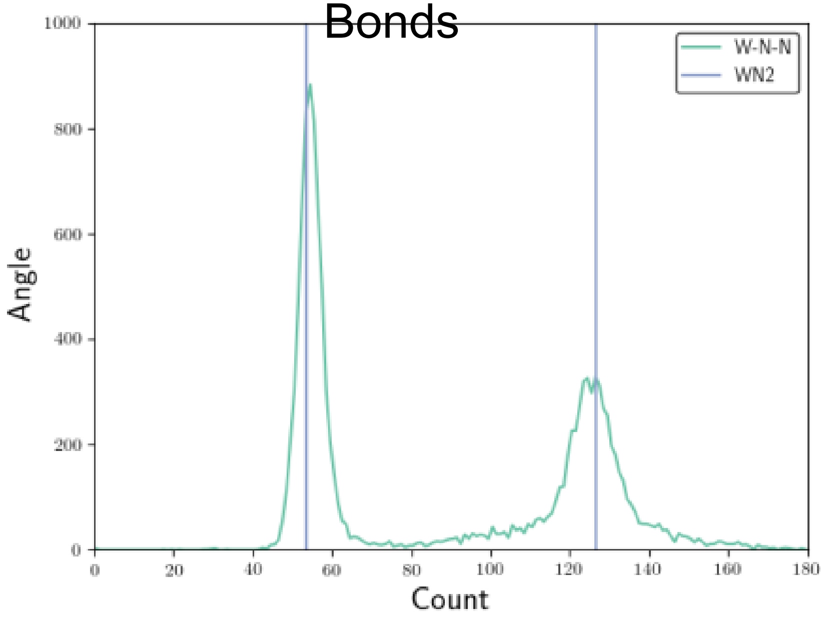
Near-Surface Tungsten Nitride Growth

W: Grey N: Pink

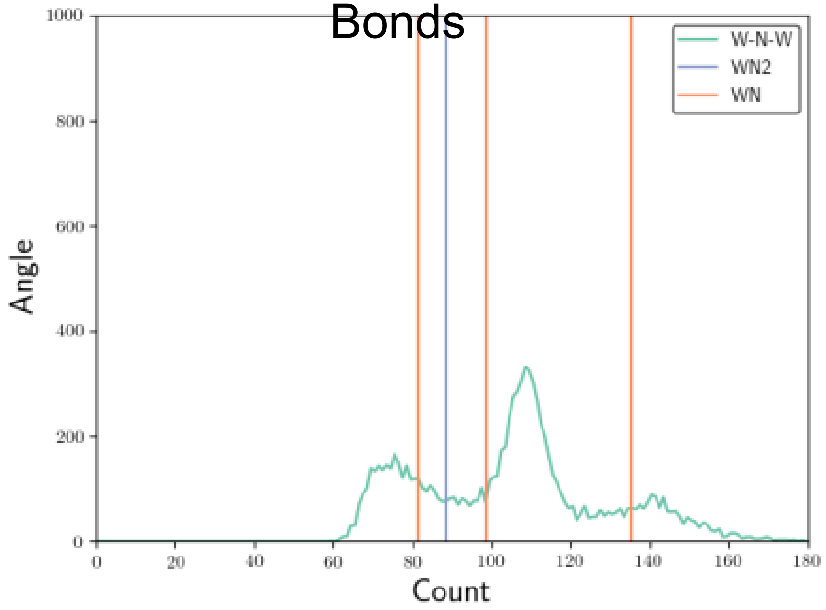
N Implantation (100) Surface 30 ns



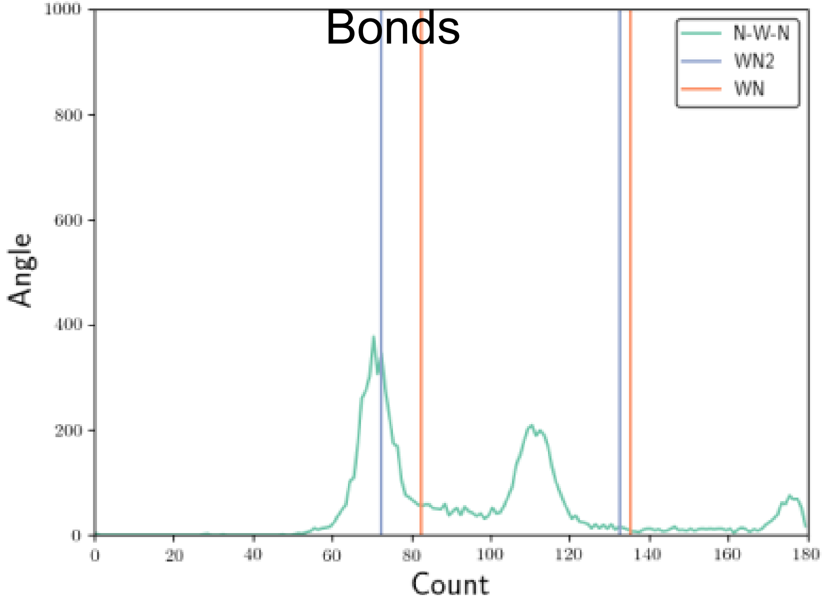
W-N-N Bonds



W-N-W Bonds



N-W-N Bonds



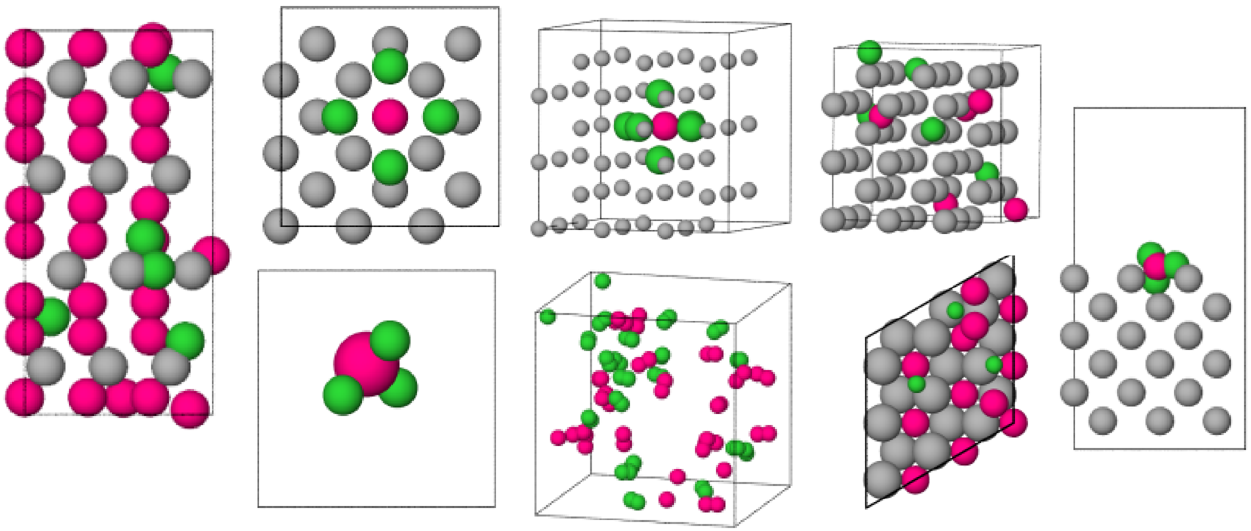
- Bond angle analysis to determine structure in near-surface region
- Analyzed from depth of 10 Å and above

Future SNAP Work: W-Be-H and W-N-H

W: Grey H: Green N: Pink Be: Purple

W-N-H SNAP

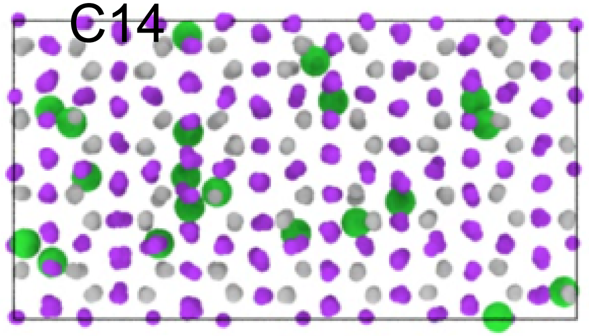
- Interest in studying hydrogen trapping in tungsten nitrides and potential ammonia formation on the surface
- Currently generating training data for W-N-H
  - H/N on surfaces and in bulk, ammonia, ordered and DFT-MD structures
- Focus on H-N surface interactions

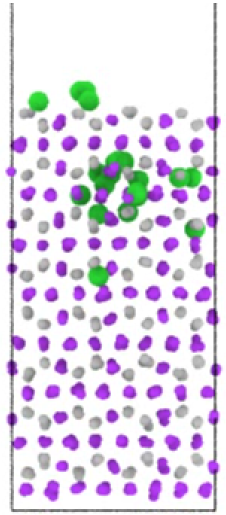


W-Be-H SNAP

- Expand previous W-Be SNAP potential to include hydrogen
- Initial training data and fits have been performed
- Initial potentials are stable, especially in bulk, but H surface behavior is poor i.e. H quickly evacuates Be surfaces
- Generated new training data based on performance of earlier potentials

H Stable in bulk C14

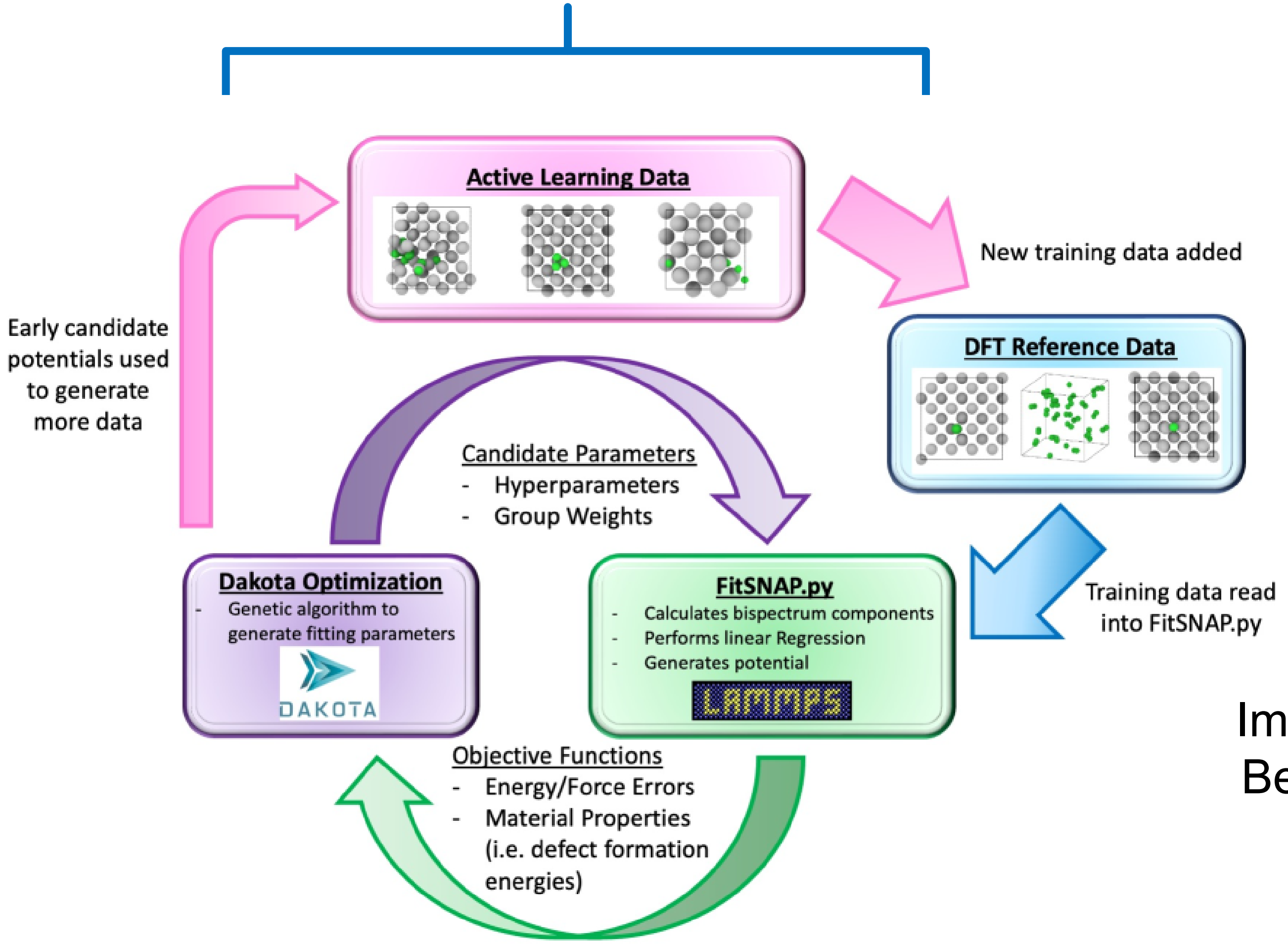




H immediately leaves C14 surface

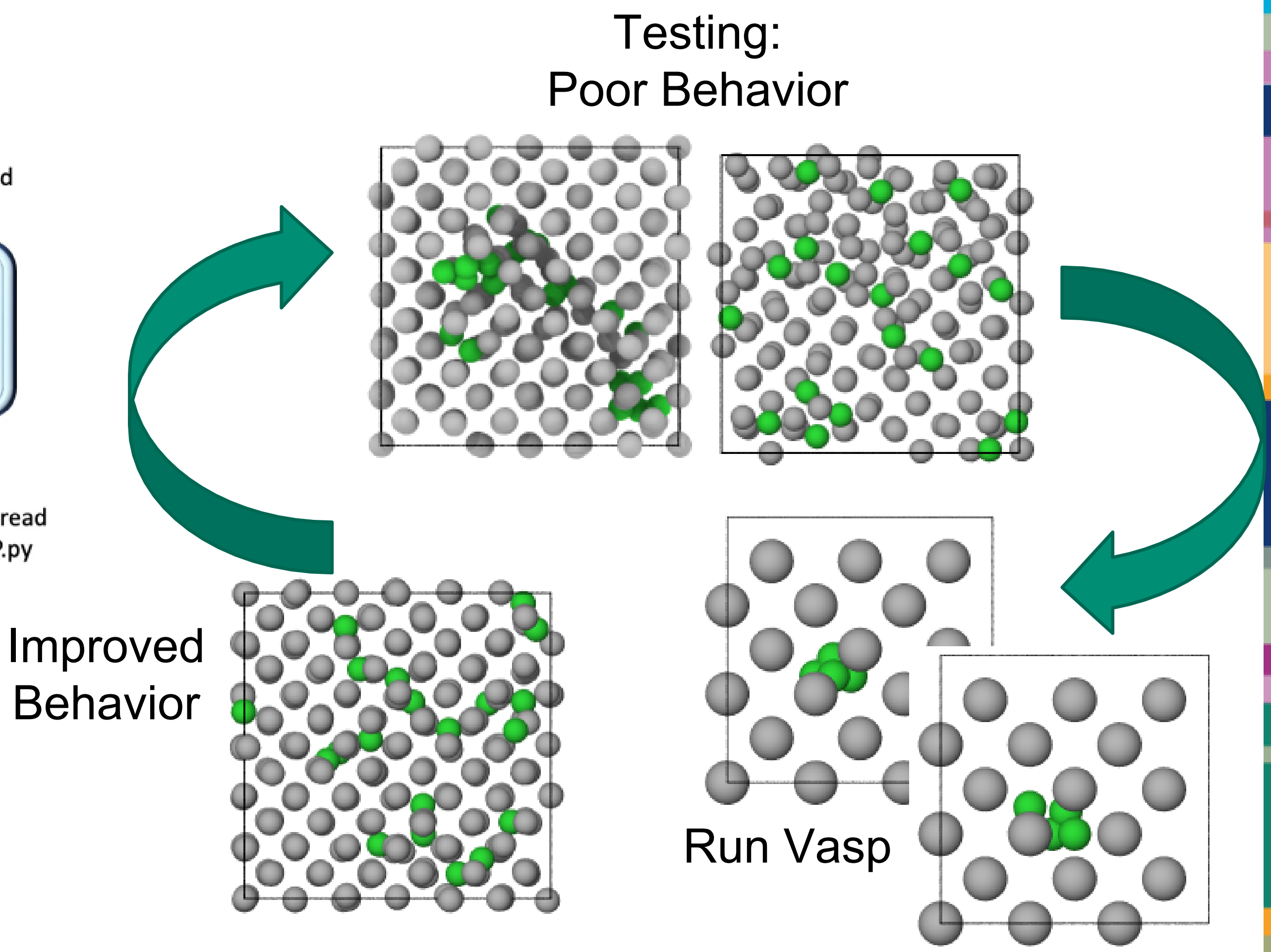
Improving Potentials with Active Learning

Most Effort Here



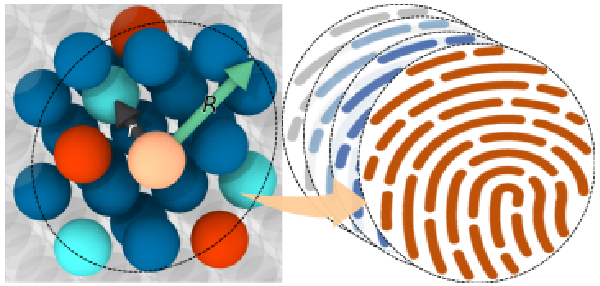
FitSNAP Workflow

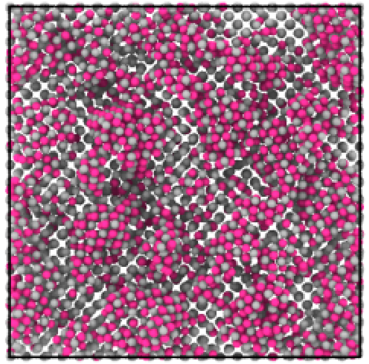
Introduce Additional Training Data
“By Hand” Active Learning

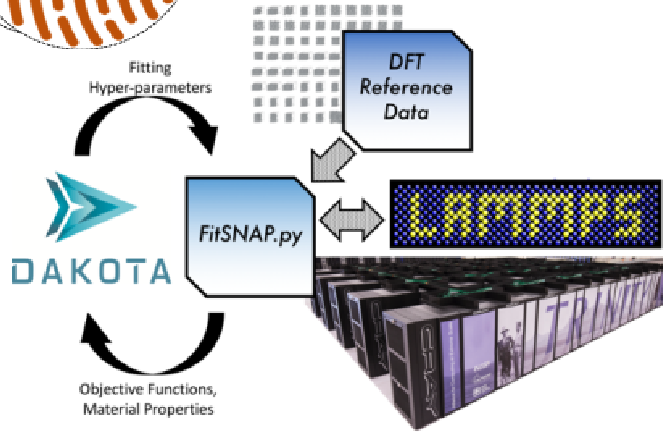


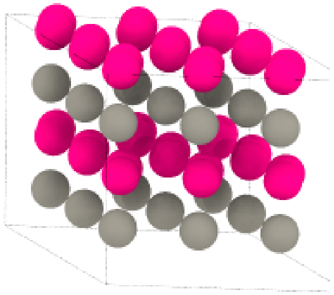
Summary and Future Work

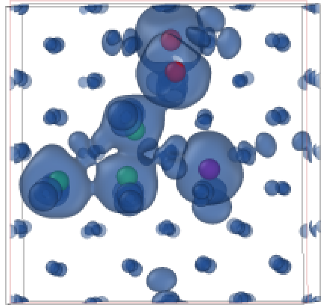
- Understanding material degradation in PFMs is critical for designing viable fusion reactors
- Atomistic modeling plays a key role in understanding relevant physical mechanisms for material degradation at the divertor
- We have developed a variety of SNAP potentials for studying PMIs in tungsten and these potentials have been used to for large-scale MD simulations
- <u>Future Work:</u>
  - Development of W-Be-H potentials to study H retention in mixed W-Be surfaces
  - Development of W-N-H to study H trapping and ammonia formation on W-N surfaces

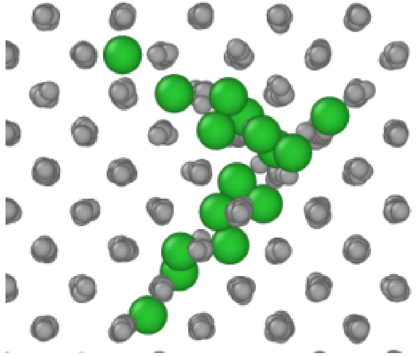


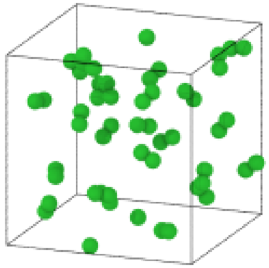


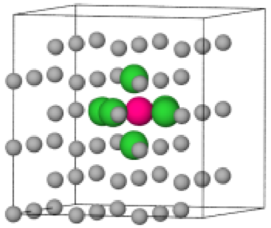












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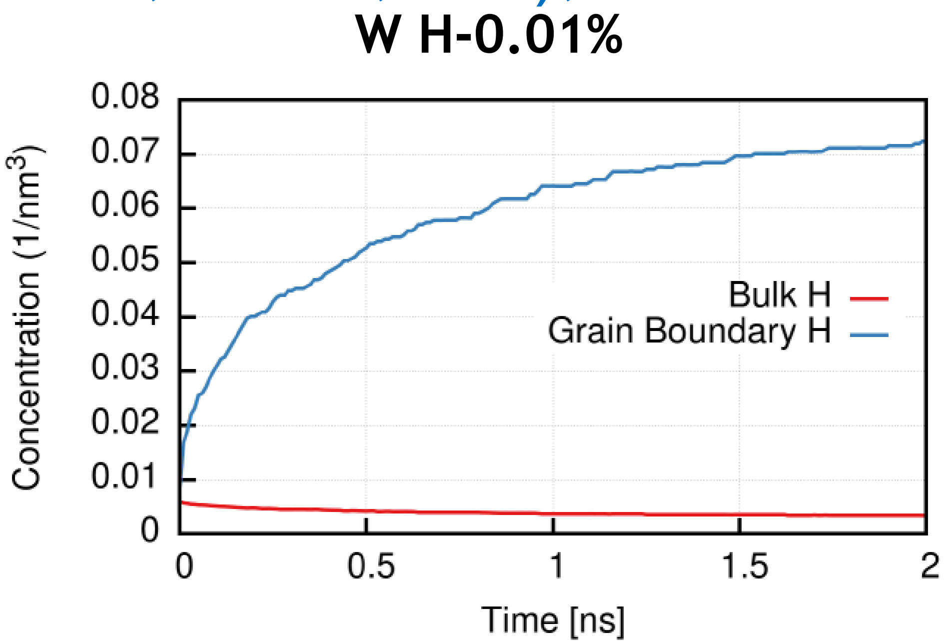
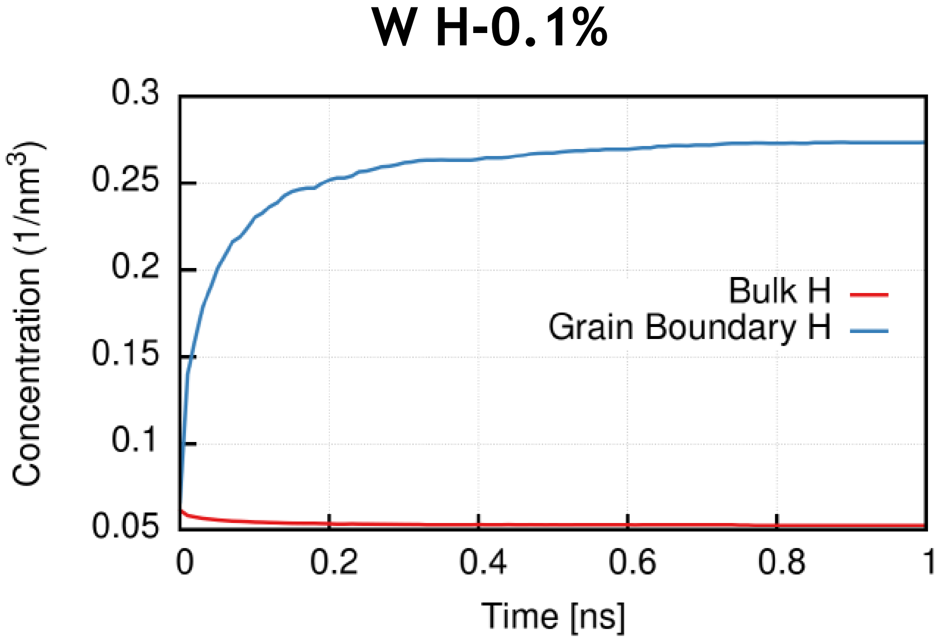
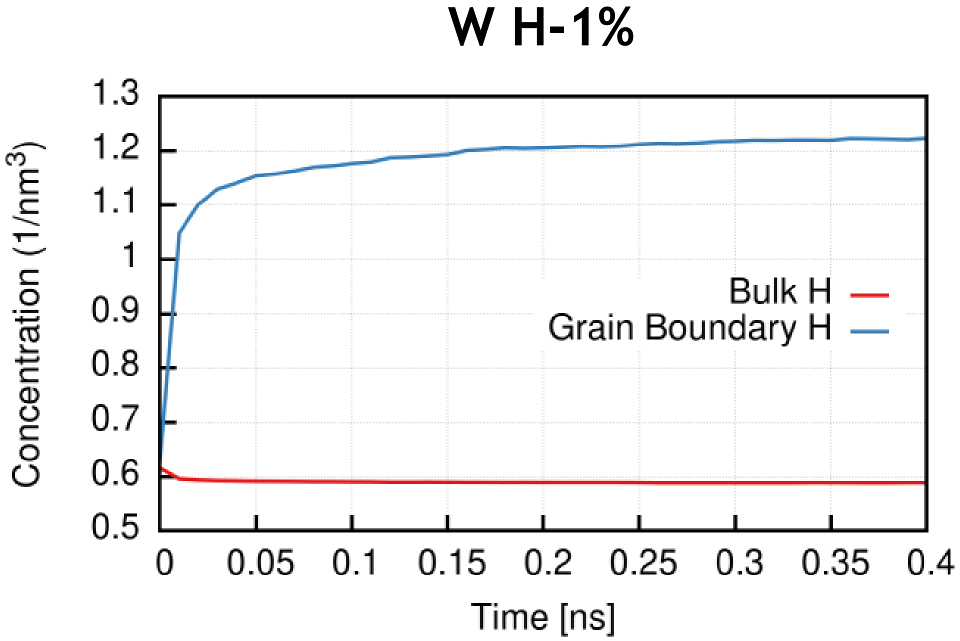
Backup Slides

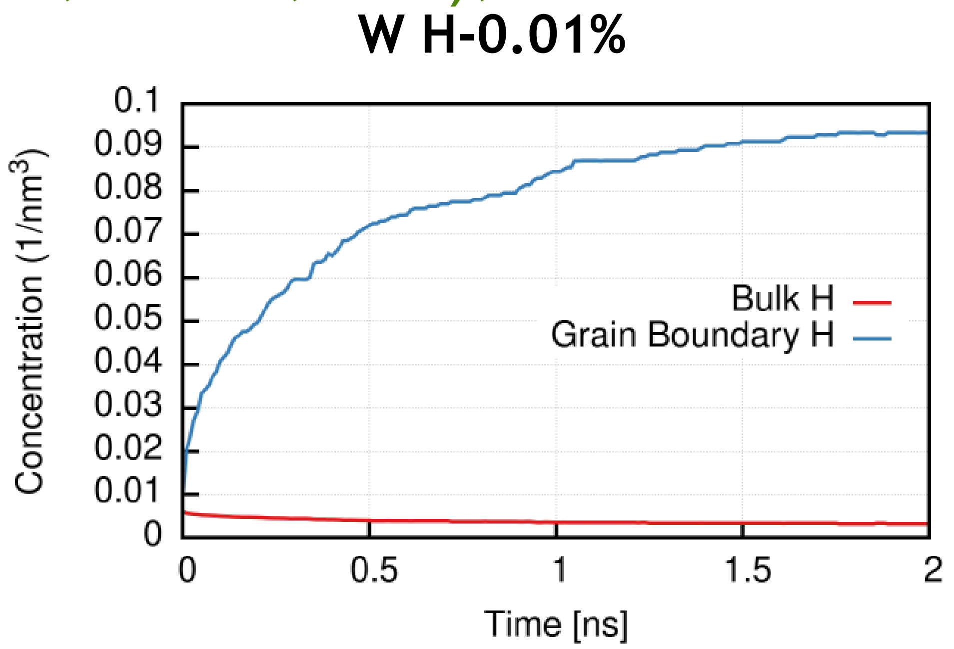
Twin GB (H = 0.01%, 0.1%, 1%), H-concentration (MW plot)

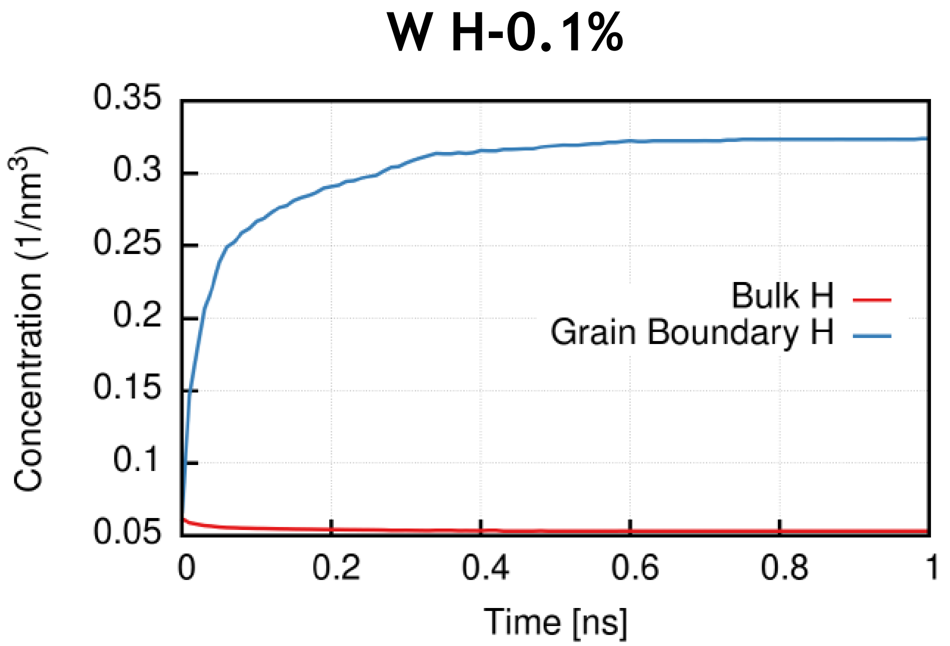


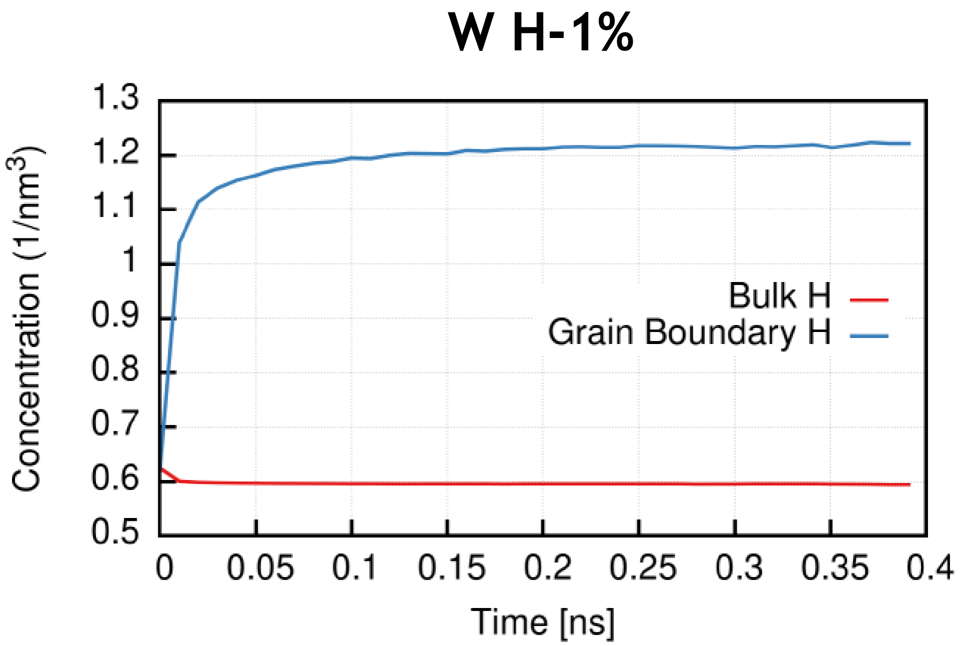




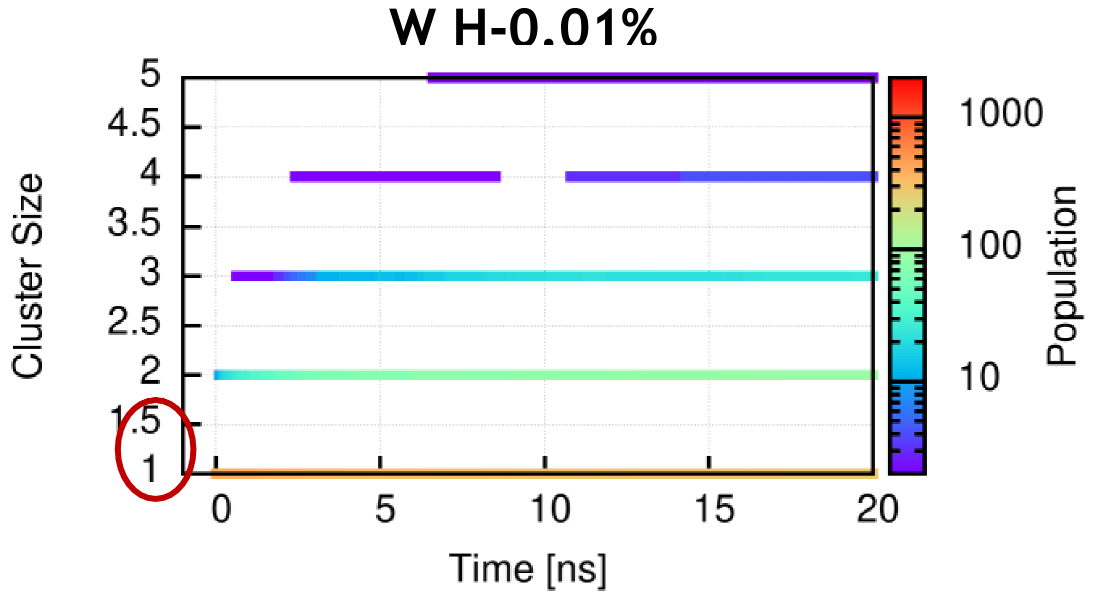
Σ5 GB (H = 0.01%, 0.1%, 1%), H-concentration (MW plot)

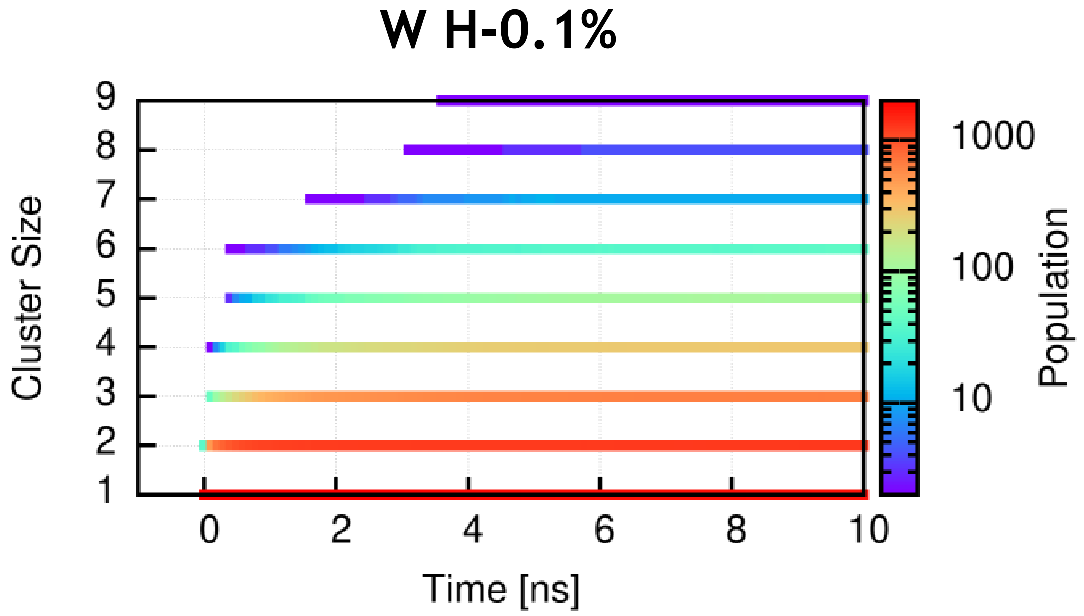


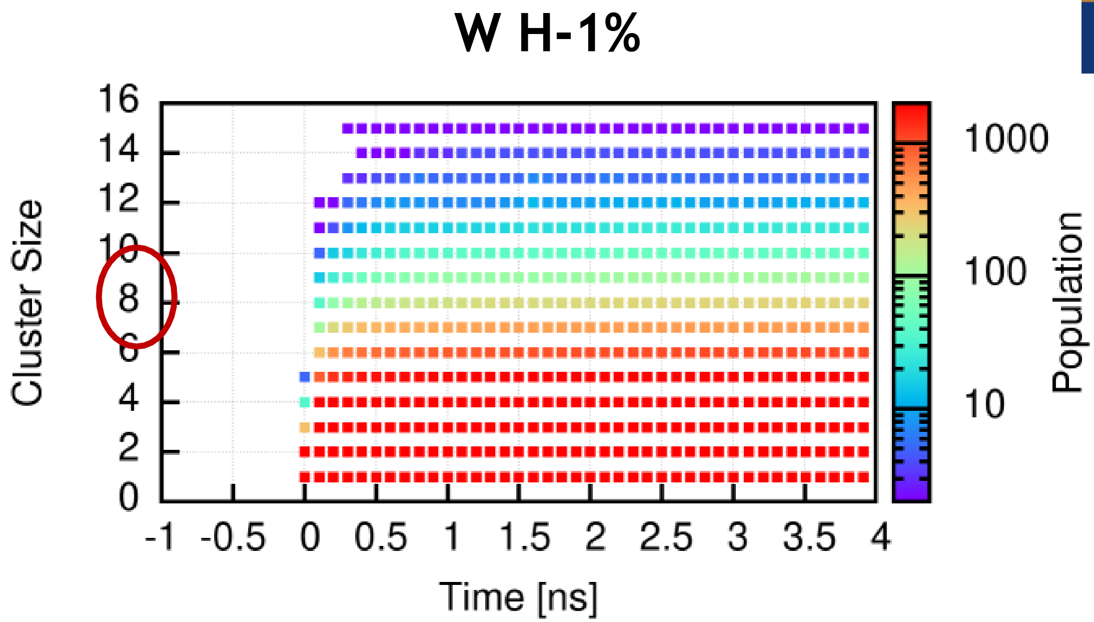




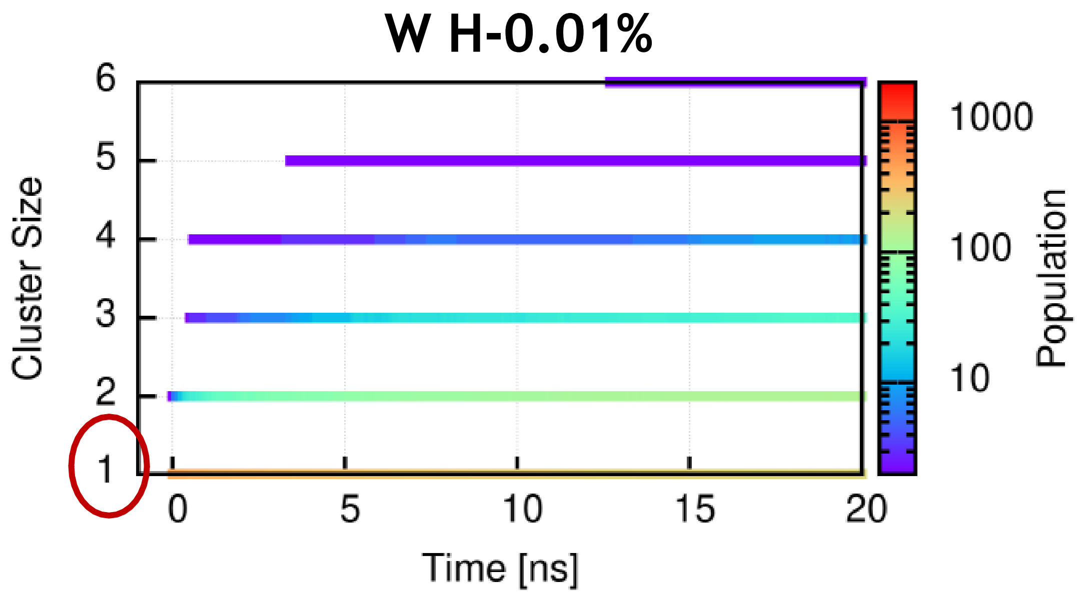
Twin GB (H = 0.01%, 0.1%, 1%), cluster information

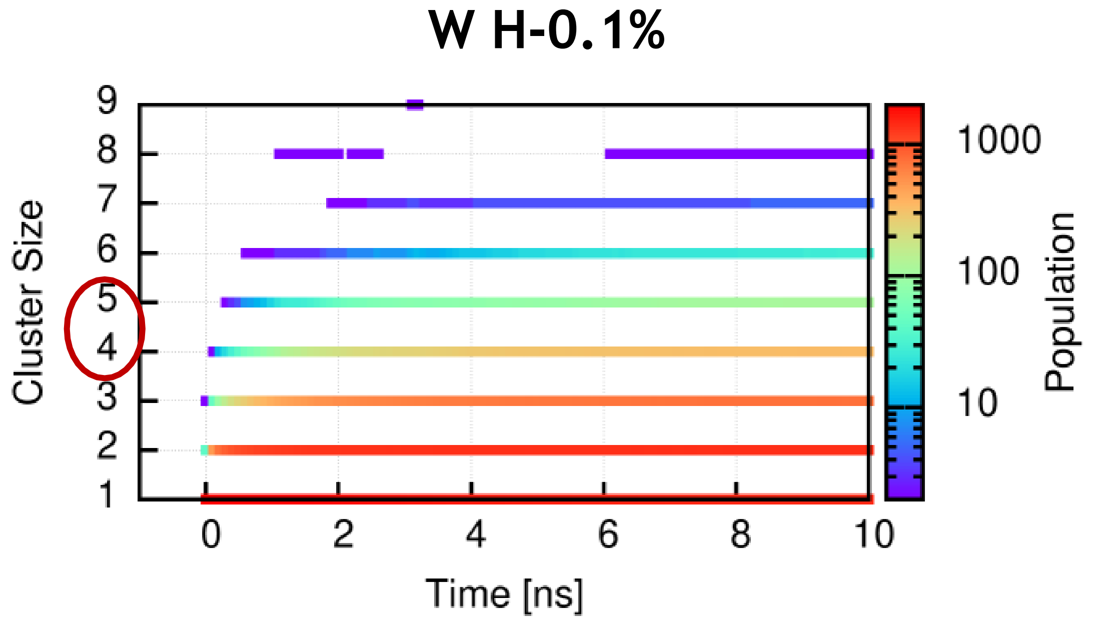


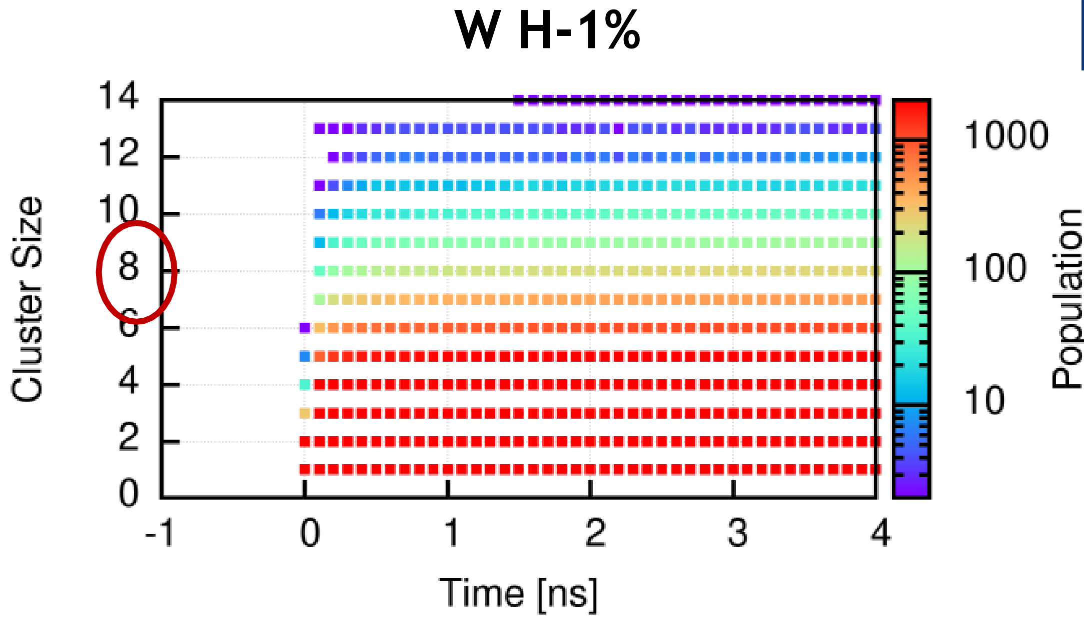




Σ5 GB (H = 0.01%, 0.1%, 1%), cluster information



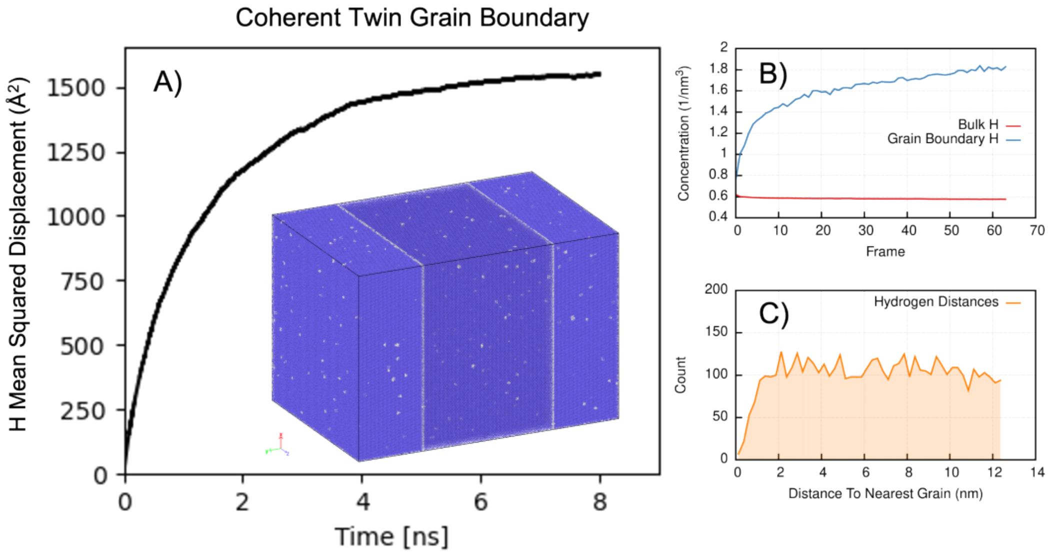




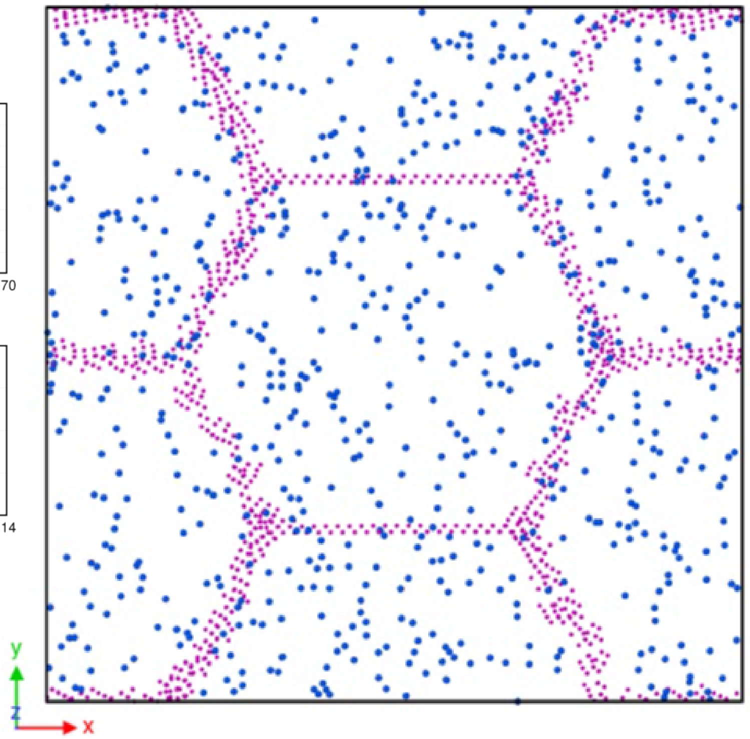
Large-Scale W-H Simulations on Summit

Modeling different GB configurations to investigate trapping at platelets vs. GBs

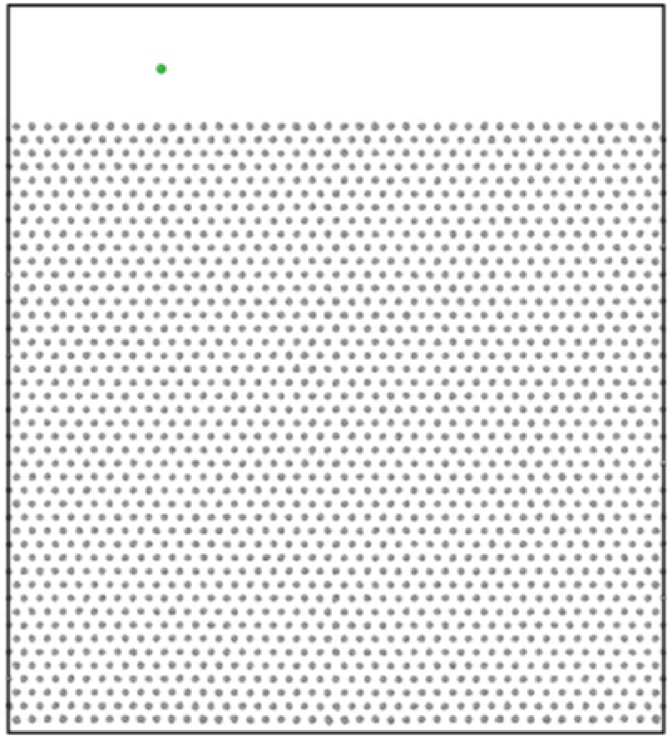
H Distribution with Twin GB



Polycrystalline W with H Seeding



H Implantation with Twin GB



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