Overview of SciDAC – PSI: A multiscale – multiphysics approach to simulating tungsten plasma surface interactions from the boundary plasma to the bulk substrate

Rick Kurtz, on behalf of:

Institution	Principal Investigator	Additional Personnel
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GA/DIII-D	Phil Snyder	Rui Ding, Adam McLean (LLNL), Vincent Chan (retired)
LANL	Xianzhu Tang	Jim Ahrens & Li-Ta "Ollie" Lo (SDAV), Sham Bhat (QUEST), Danny Perez, Art Voter, Blas Uberuaga
ORNL*	**Brian Wirth***	**David Bernholdt****, John Canik, Ozgur Cekmer, Ane Lasa, Jeremy Meredith (SDAV), Phil Roth (SUPER), German Samolyuk
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UMass-Amherst	Dimitrios Maroudas	Lin Hu
U Missouri	Karl Hammond	Ian Naeger, Marissa Zirges
UTK	Brian Wirth	Sophie Blondel, Mary Alice Cusentino, Donghua Xu, Tim Younkin

In partnership with:

FASTMATH

QUEST

SDAV Scalable Data Management, Analysis, and Visualization

SUPER INSTITUTE FOR SUSTAINED PERFORMANCE, ENERGY, AND RESILIENCE

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DEPARTMENT OF NUCLEAR ENGINEERING

DIII-D NATIONAL FUSION FACILITY SAN DIEGO

Pacific Northwest NATIONAL LABORATORY

Los Alamos NATIONAL LABORATORY EST. 1943

Argonne NATIONAL LABORATORY

OAK RIDGE National Laboratory

Project web site:
https://collab.mcs.anl.gov/display/PSIscidac/

Presented at the IAEA CRP meeting on Plasma Material Interactions
Vienna, Austria
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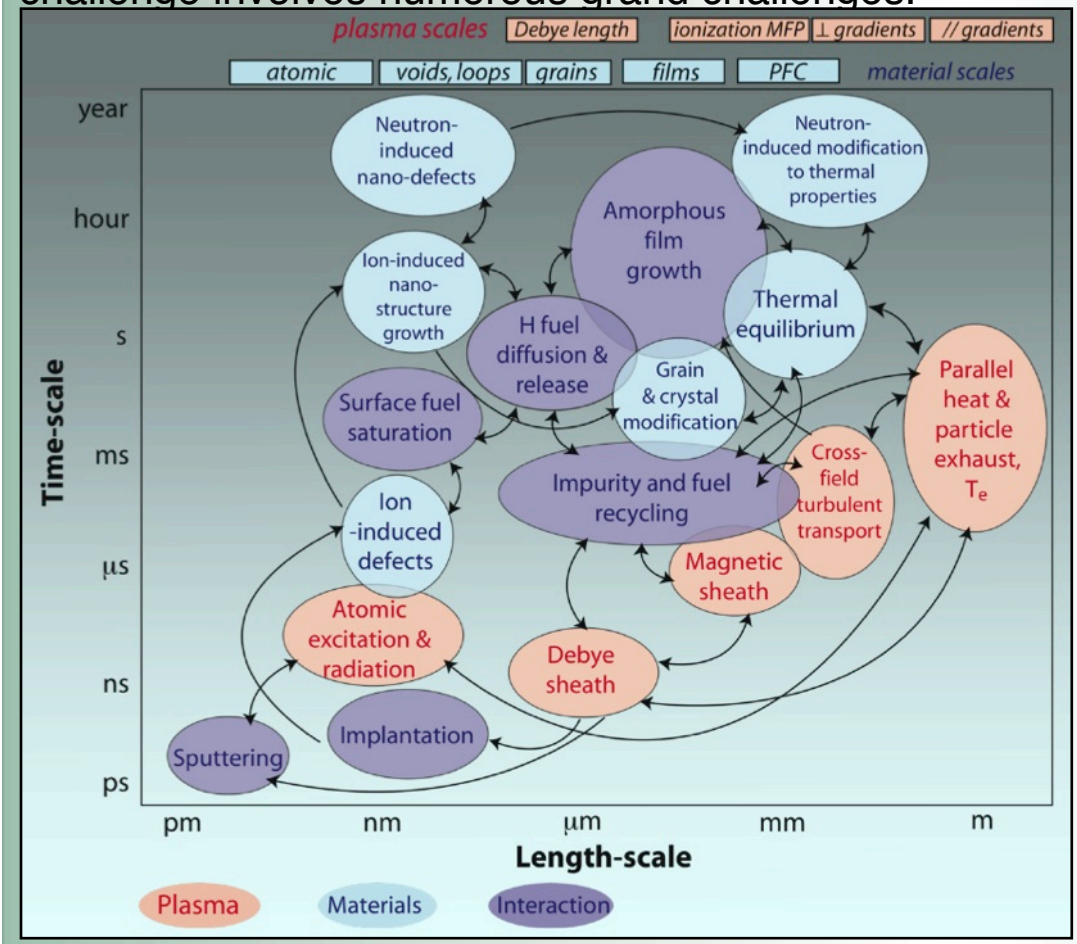
#

This work was supported by the U.S. Department of Energy, Office of Fusion Energy Sciences and and Advanced Scientific Computing Research (ASCR) through the SciDAC-3 program.

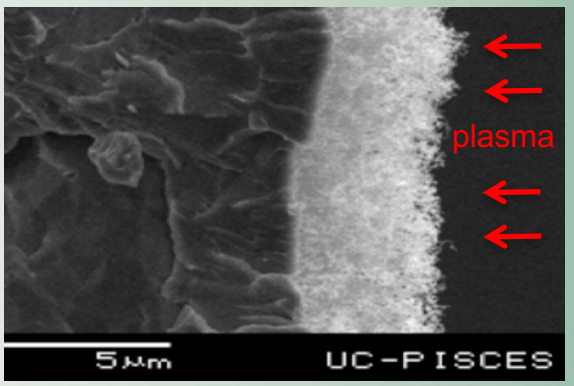
Challenge of the Fusion Nuclear Environment

- Plasma Surface Interactions, Fusion Neutron Transmutation & Damage -

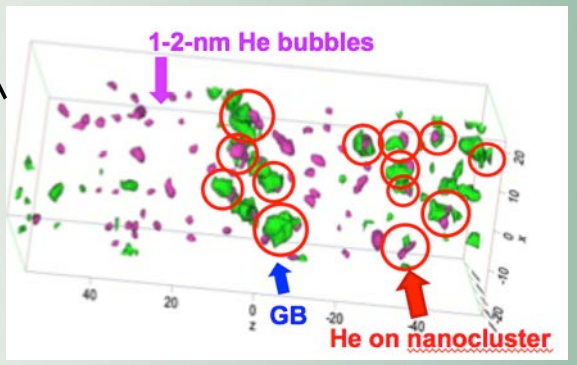
Application of leadership class computing and computational materials science are key tools to accelerate fusion materials development. However, as governing phenomenon span decades in length and time scale; this challenge involves numerous grand challenges.



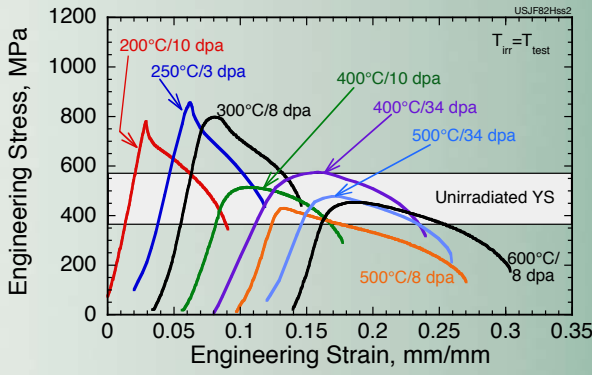
plasma interaction



transmutation helium



cascade hardening



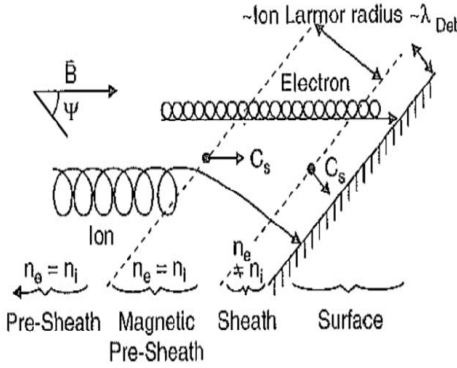
PSI Perspective & Outline

• Objective is to develop PSI simulation capability across three coupled spatial regions:

- **Edge/scrape-off-layer region of the plasma, with sheath effects**

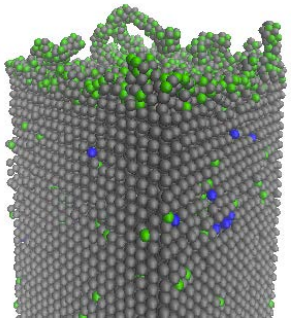
- **Near surface material response to plasma exhaust, with neutron damage and influenced/coupled to plasma sheath**

- **Structural materials response to intense, 14 MeV-peaked neutron spectrum**



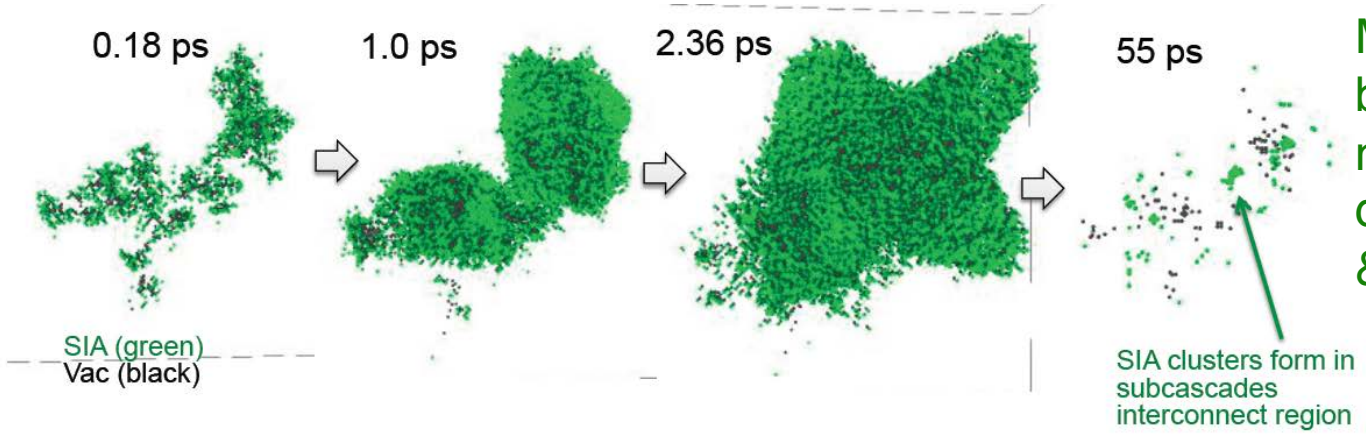
Plasma Edge Scrape Off Layer Sheath – heat & particle flux, recycling, etc.

mm



Material surface - erosion (impurity & dust), T retention, surface evolution

10^2 – 10^3 nm



Material bulk – neutron damage & transmutation

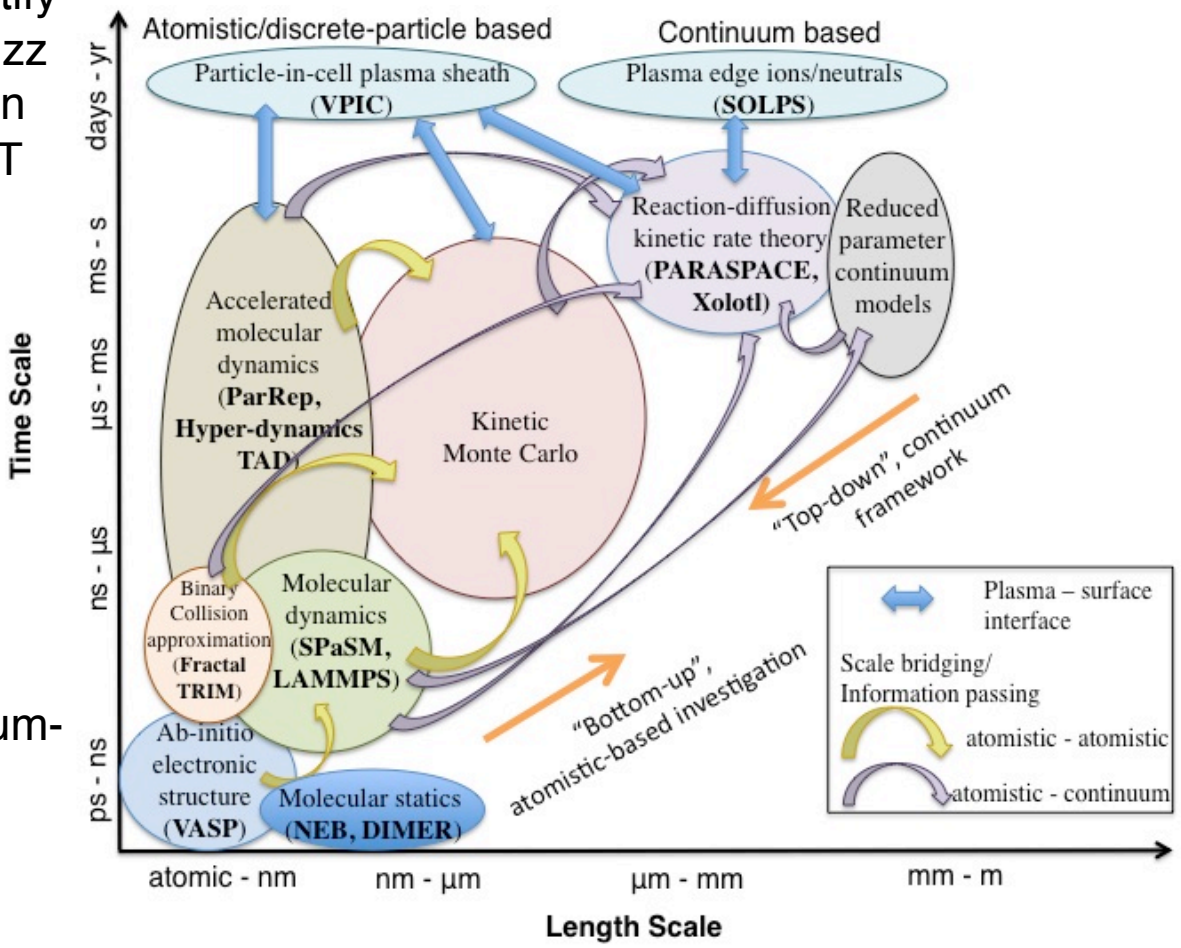
10^{-3} – 10^2 mm

*Multiscale modeling capability – a work in progress**

Goal: Discovery science to identify Mechanisms of W nano-scale fuzz formation and synergies between He & H exposure that impact D/T permeation & retention – and surface mass loss (dust)

Mechanisms of interest: sputtering, surface adatom formation, diffusion, He bubble formation, expansion & rupture

Focus on MD & kinetic modeling approaches, leading to a large-scale continuum-level reaction-diffusion code for plasma materials interactions

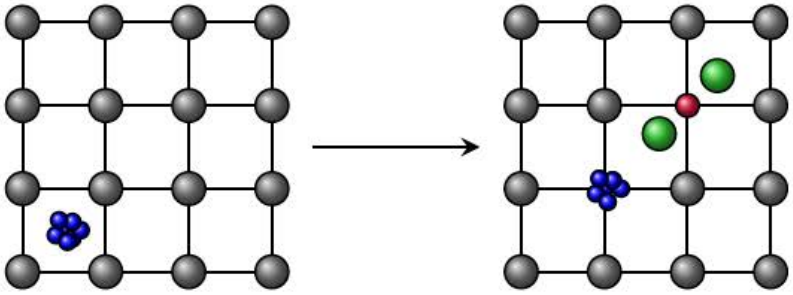


Biggest long-term scientific challenge is understanding the kinetics of coupled defect – impurity evolution with a disparate range of kinetic rates --- this requires algorithmic improvements on both the physics and computing side

* BD Wirth, K.D. Hammond, S.I. Krashenninikov, and D. Maroudas, *Journal of Nuclear Materials* **463** (2015) 30-38.

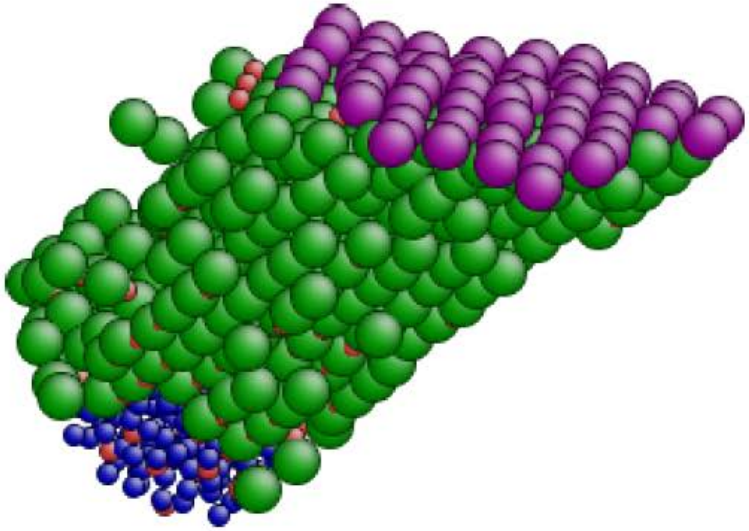
Key MD observations of early stage He bubble evolution

- **Helium insoluble but highly mobile and can self-trap (at high implantation rates) due to strong He-W repulsion to form highly mobile, strongly bound helium clusters – *implantation rate effects are very important***



"trap mutation" processes

Occurs when 6–9 helium atoms coalesce, depending on temperature, after which bubble grows by absorbing smaller clusters.



"loop-punching" processes

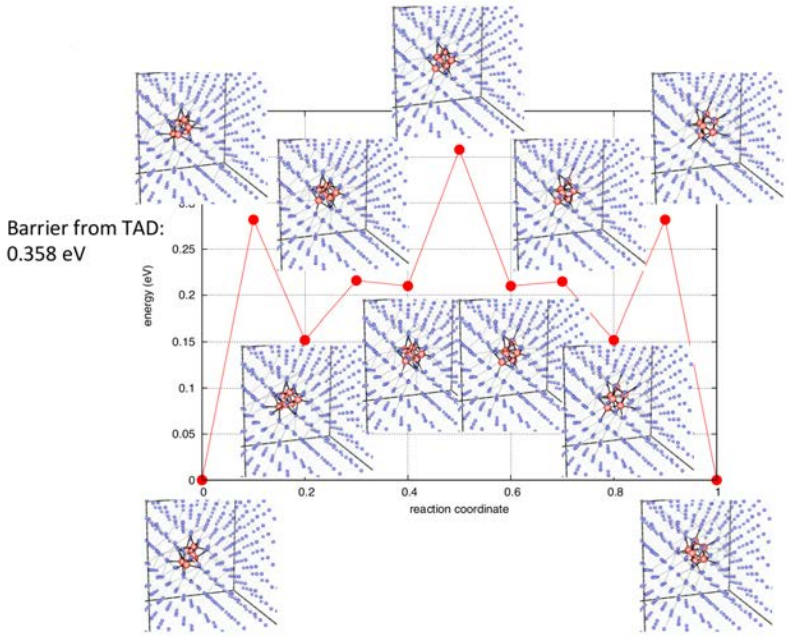
Movie available with F. Sefta, *et al. Nucl. Fusion* **53**: 073015 (2013)

- **Significant surface evolution through tungsten adatom formation, driven by trap mutation and loop-punching as tungsten interstitials rapidly diffuse to surface**
- ***As bubbles continue to grow at very high pressure, eventually rupture***

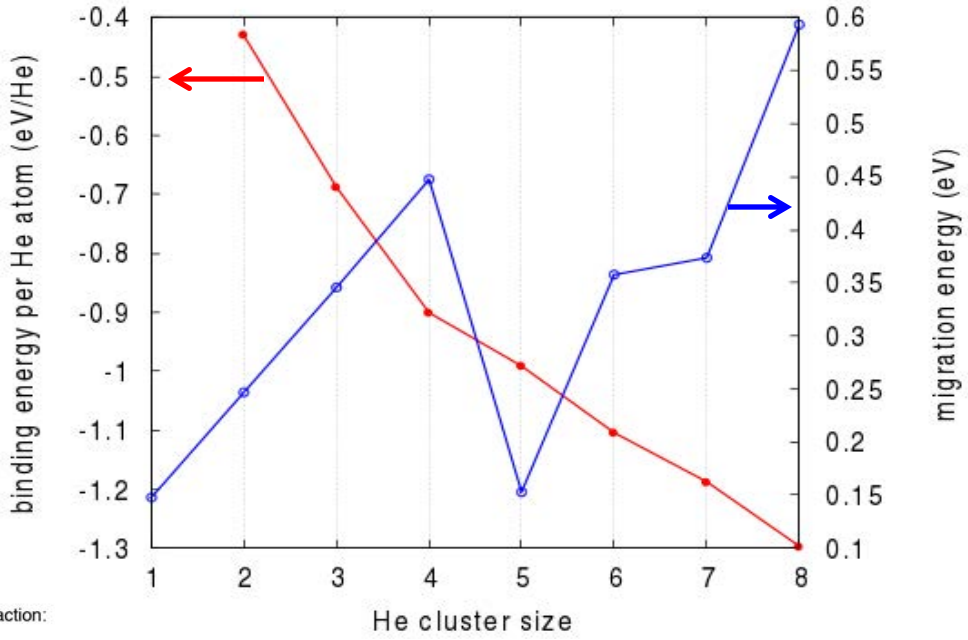
*Thermodynamics & kinetics of small He clusters**

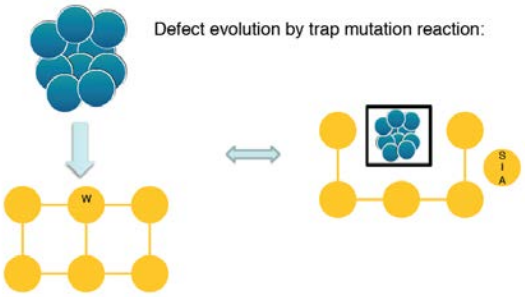
- **Atomistic simulations (AMD, MD, statics) used to identify unit transport/reaction mechanisms**
  - Challenges relate to multitude of pathways with increasing cluster size

4-He cluster migration:



Thermodynamics (binding) & kinetics (migration):





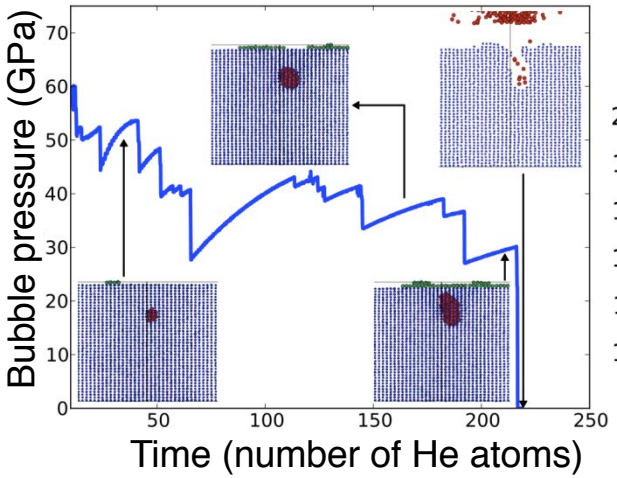
Larger clusters undergo 'trap' mutation which decreases mobility

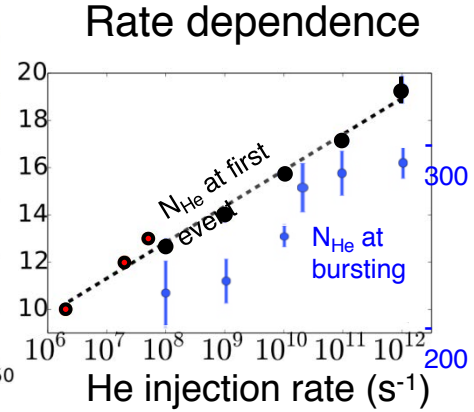
Monotonic increase in binding energy,
Complex, size-dependent kinetics

* Perez, Vogel and Uberuaga, *Phys. Rev B* **90** (2014) 014102.

Accelerated MD simulations of rate effects on near-surface helium bubbles with rates approaching ITER relevance

First simulation of He bubble growth at He-irradiation flux appropriate for fusion first-wall in ITER. The simulations find a qualitatively different growth mode when rates approach experimental values. They reveal rate effects on bubble size, shape, pressure, and surface damage.





Left: Growth/bursting at intermediate (near-crossover) rate (10^9 He/s). Helium pressure is relieved via emission of W interstitials. At slower growth rates, interstitials have time to diffuse over surface of bubble, ultimately emitting more interstitials from top in the form of dislocation loops (crowdion clusters) that travel to surface, leaving adatoms. Right: Dependence of bubble properties on bubble growth rate.

Research

Parallel Replica Dynamics simulations of bubble growth with He injection rate ranging from 10^{12} s^{-1} to $2x10^6$ s^{-1}. Efficient to petascale: utilized 160,000 cores (over half of Titan) at ORNL at 77% efficiency.

Slower growth leads to smaller, more anistropic bubble that grows in a directed way towards surface, producing fewer adatoms during growth and creating less surface damage upon bursting.

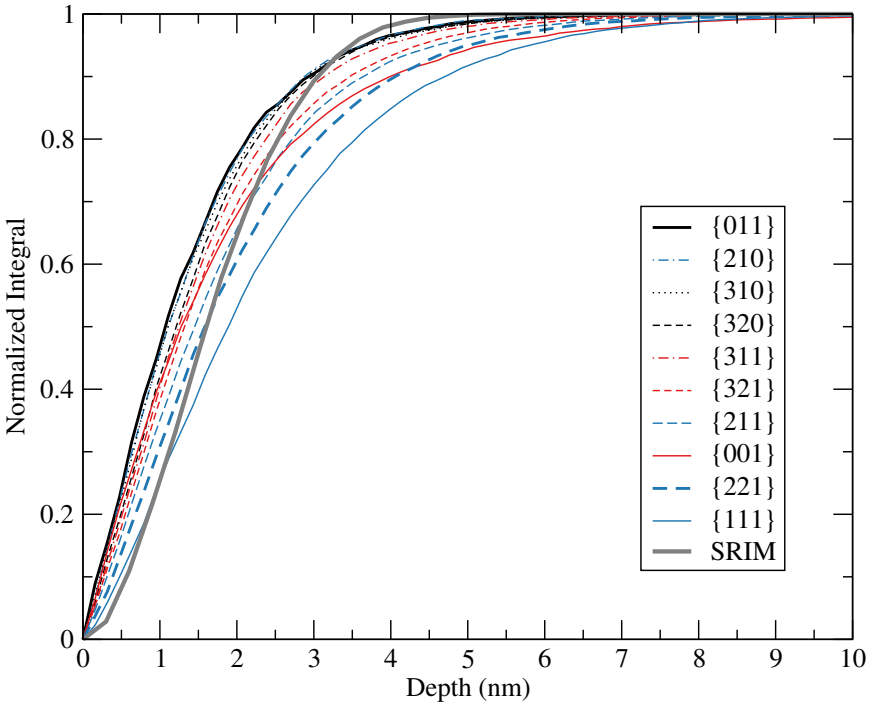
Collaboration with BES program Accelerated Molecular Dynamics (Voter) at LANL.

L. Sandoval, D. Perez, B.P. Uberuaga, and A.F. Voter; *Physical Review Letters* **114** (2015) 105502 (2015). Work performed at LANL, computing at ORNL**. OLCF and DOE Office of Science highlight**

Our approach to large-scale MD simulations

- Develop a database of implanted He range

  - Bombard a “clean” surface with 100 eV helium 20 000 times
  - Calculate, for each crystallographic surface,
    - Fraction of ions that reflected
    - Fraction that embedded but escaped in < 5 ps
    - Depth distributions
  - Advantages
    - Constant (large) time step
    - Don’t simulate ions scattering off the surface
    - > 90% reduction in simulation time



Caveat: ignores electronic stopping

Potential: N. Juslin and B. D. Wirth, *J. Nucl. Mater.* **432**: 61 (2013)

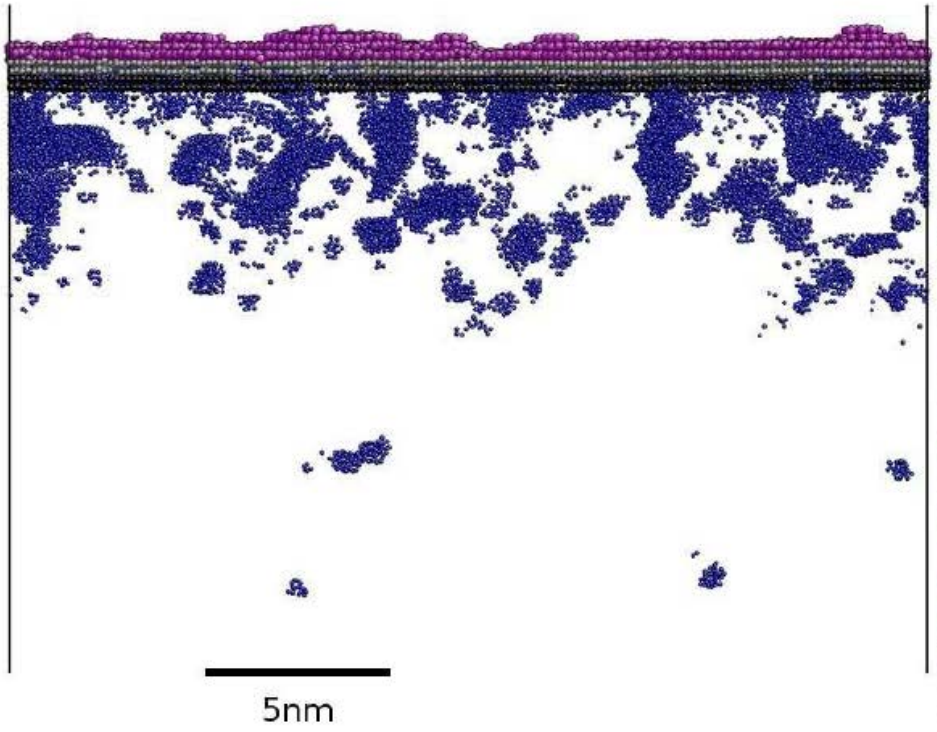
- Periodic conditions in the x, y directions and Free Surface in z (typical simulations are 50x50 nm cross section and 25-50 nm deep (O(20 Million atoms)

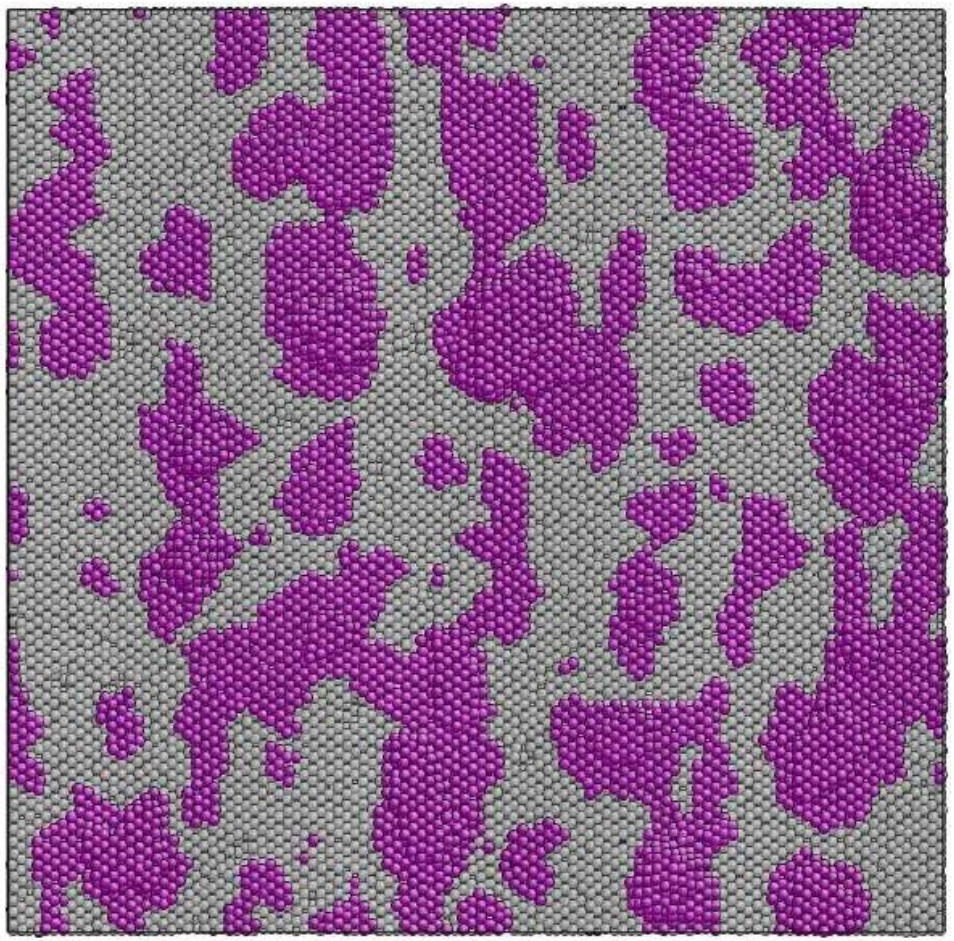
- Every 10 ps a He atom is added according to the He depth distribution from MD (ignores electronic stopping) and reflection, but effective implantation flux of ~4×10^{25} $m^{-2}s^{-1}$

Tungsten surface response to low-energy He exposure

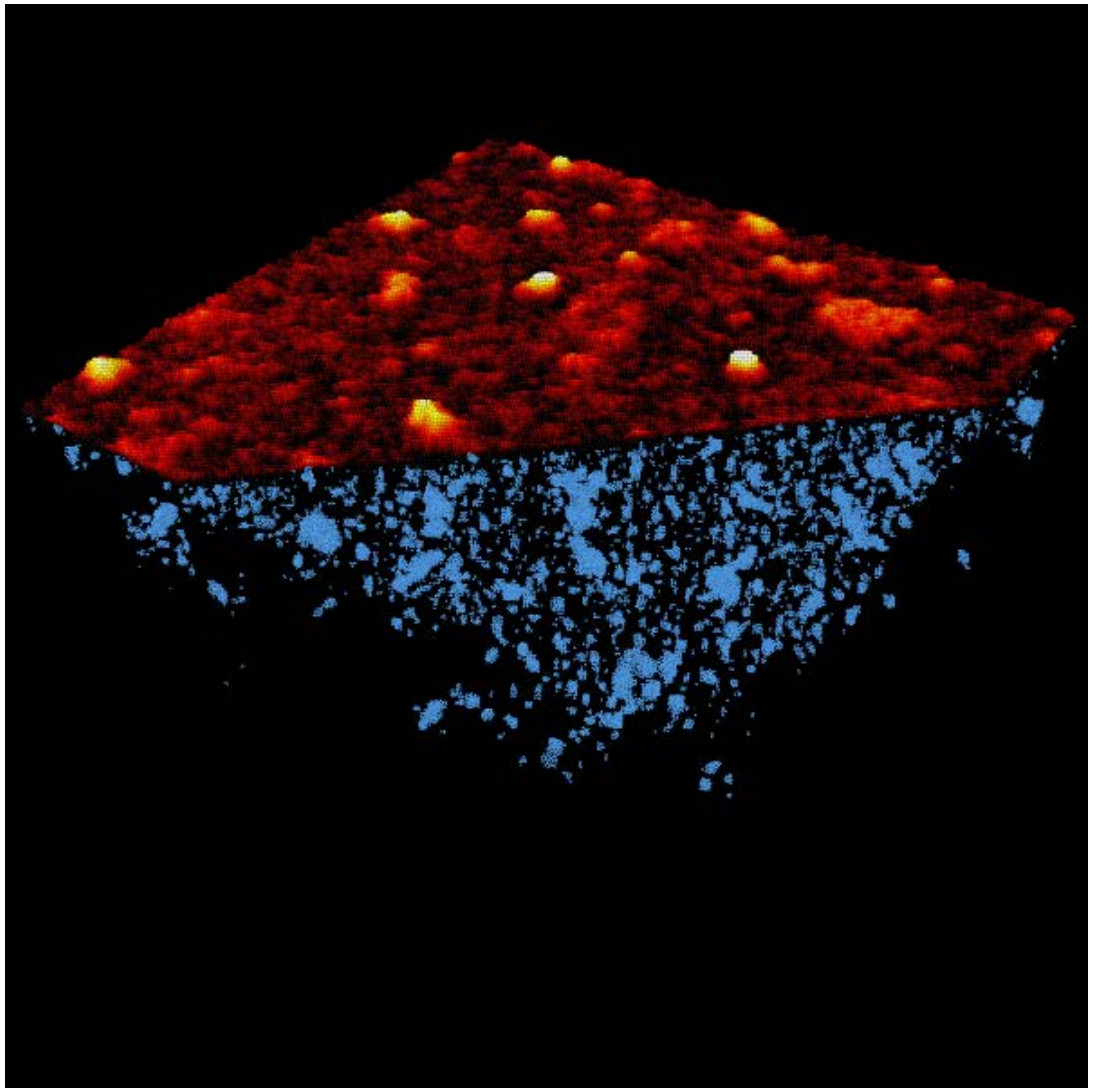
(110) surface, 1.25 μs

$\Gamma = 5.3 \times 10^{26}\ \mathrm{m^{-2}\,s^{-1}};\ \Phi \in [0, 6.8 \times 10^{20}\ \mathrm{m^{-2}}]$





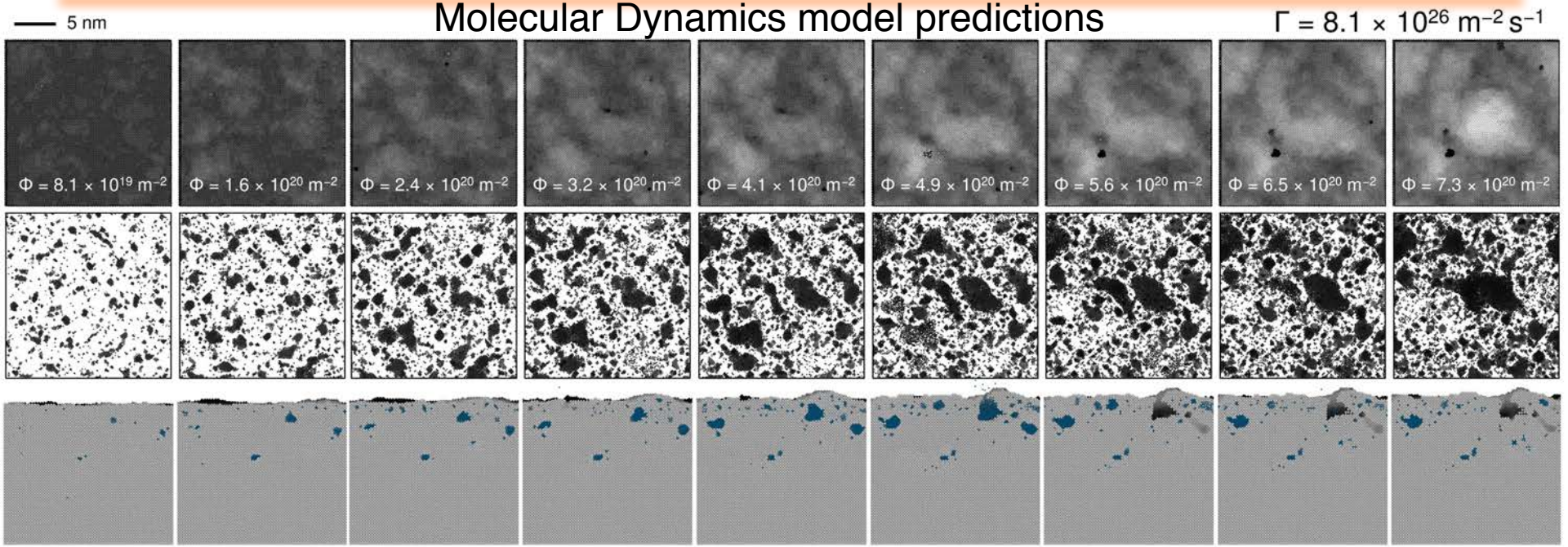
Large-scale MD results (100 eV He -> W)



- Raised portions (red/yellow/white) are “pushed up” by helium bubbles
- Bubbles typically vent non-destructively and may “heal”
- Still too low a fluence to see significant bursting events or tendrils larger than $\approx$ 2 nm

(111) surface, 1.5 μs, $\Gamma = 4 \times 10^{25}$ m^{-2}s^{-1} ($\Phi = 6 \times 10^{19}$ m^{-2})

Tungsten surface response to low-energy He exposure



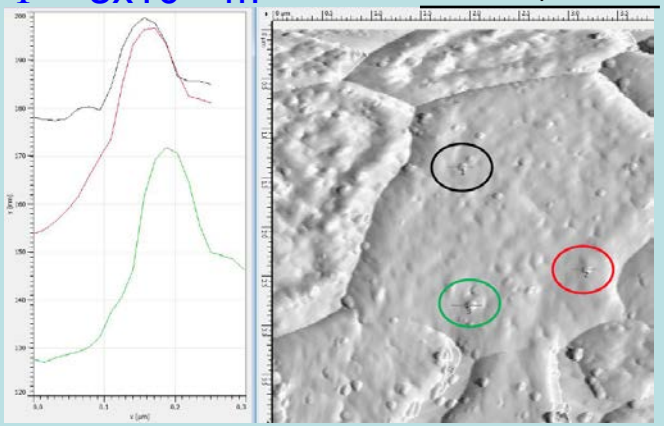
High-flux simulations showing surface growth and helium accumulation below a W(100) surface. Top: View of surface (white = +1.5 nm, black = –2 nm); Middle: helium atoms, top view (black = at surface, white = –15 nm); Bottom: cross-section.

- MD* of 100 eV He implanted into W reveals formation and growth of over-pressurized, sub-surface He bubbles thru self-trapping, trap mutation, loop punching and bubble bursting that evolve tungsten surface (hillocks & craters)

→ Qualitatively consistent with experiments** of W surface evolution following 60 eV He on tungsten

→ Quantitative comparison requires evaluation of rate & scale effects (Γ:MD 10^{26} vs expt 10^{19}; Φ: 10^{20} vs 10^{24})

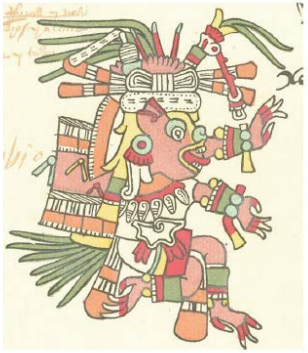
Experiment: $\Gamma = 3\times10^{19}$ m^{-2}s^{-1}
$\Phi = 3\times10^{24}$ m^{-2}



* Hammond & Wirth, UTK/ORNL

** Donovan, Buchenauer, Kolasinksi et al., SNL

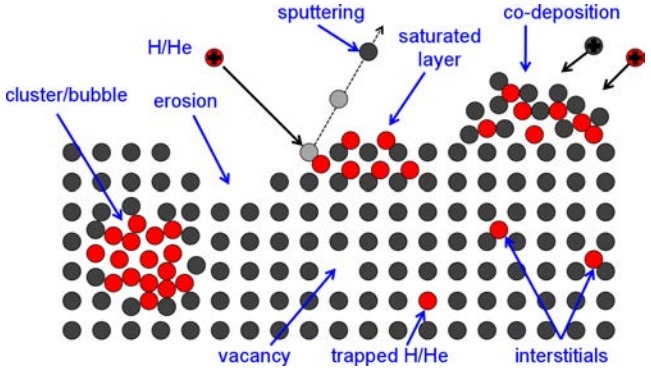
Xolotl (continuum) calculations to reach experimental scale



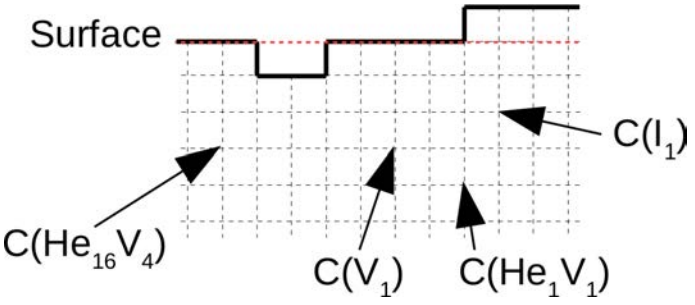
• Xolotl (SHO-lottle) is the Aztec god of lightning and death & is our continuum cluster dynamics code for modeling plasma surface interactions, focused on sub-surface bubble dynamics & surface evolution (to date), will include erosion in near future

• Developed from 'scratch' for the SciDAC project, designed for HPC (current and emerging architectures – multicore, multicore+accelerator)

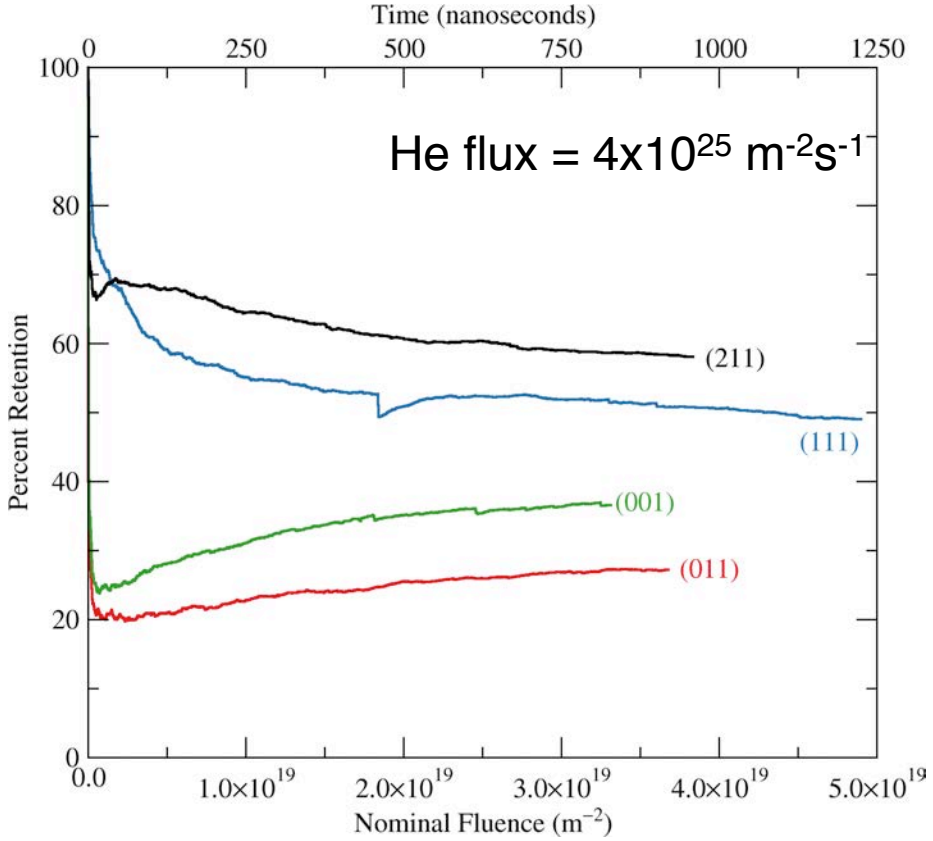
Atomistic picture



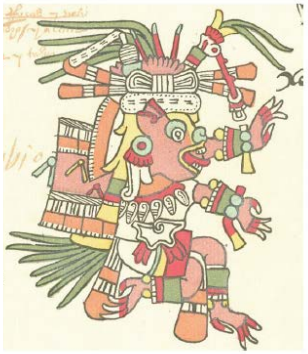
Continuum picture



MD simulations of He retention



Xolotl (continuum) calculations to reach experimental scale



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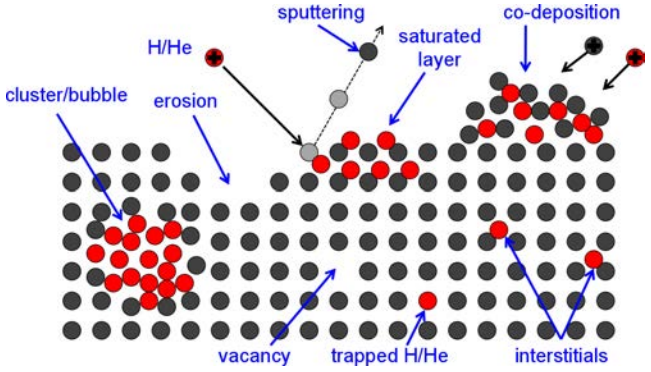
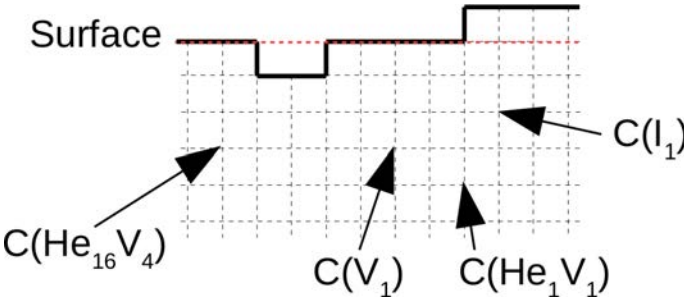
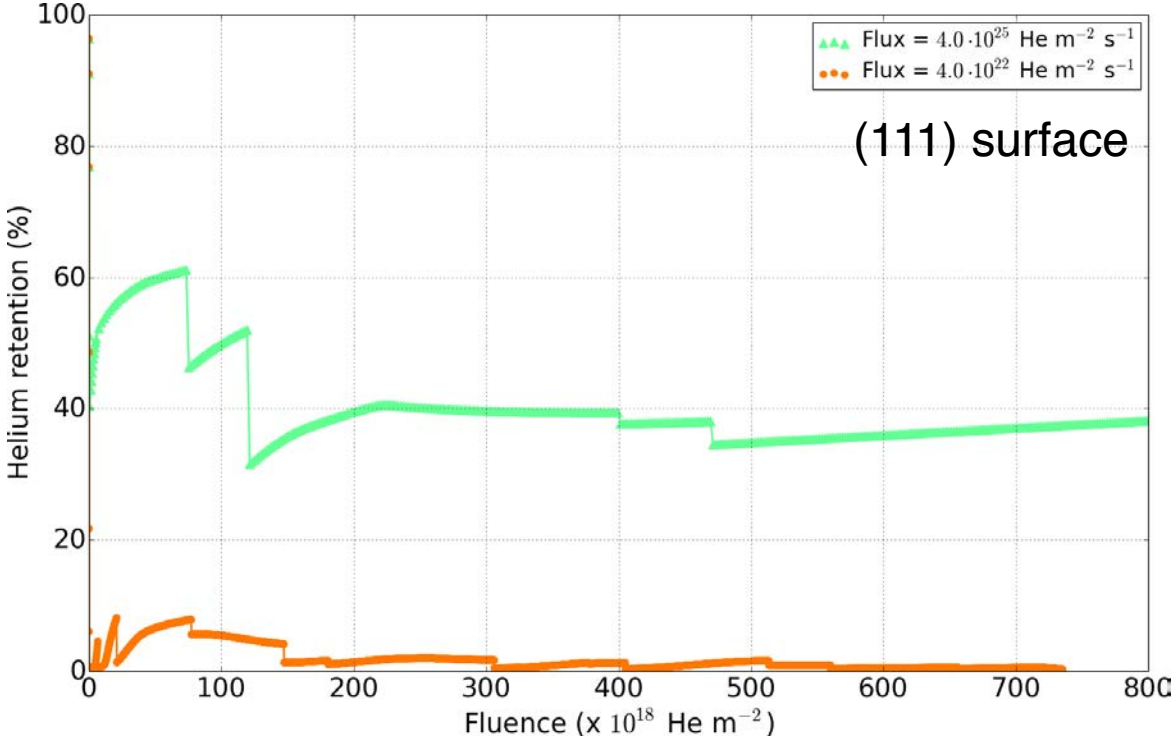


Continuum picture

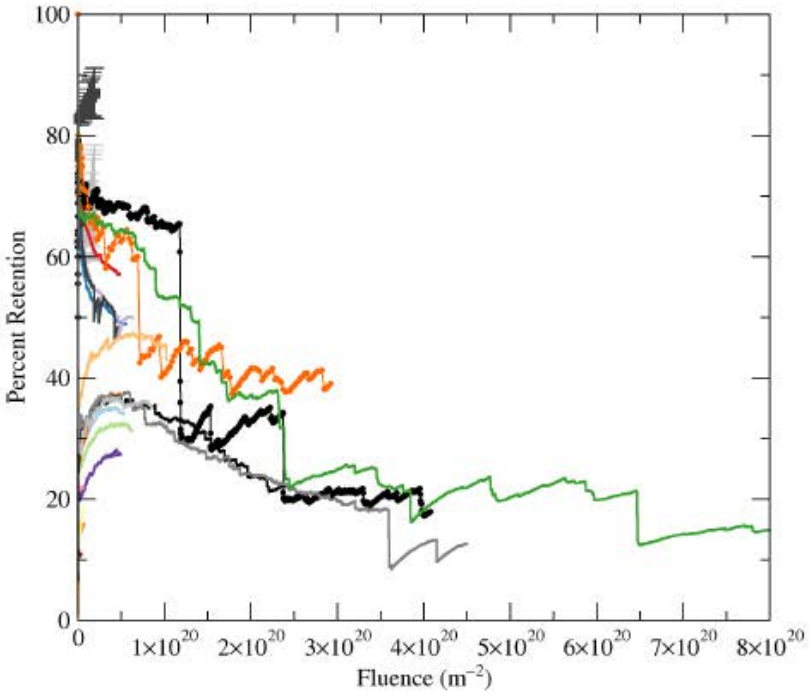


Xolotl predictions



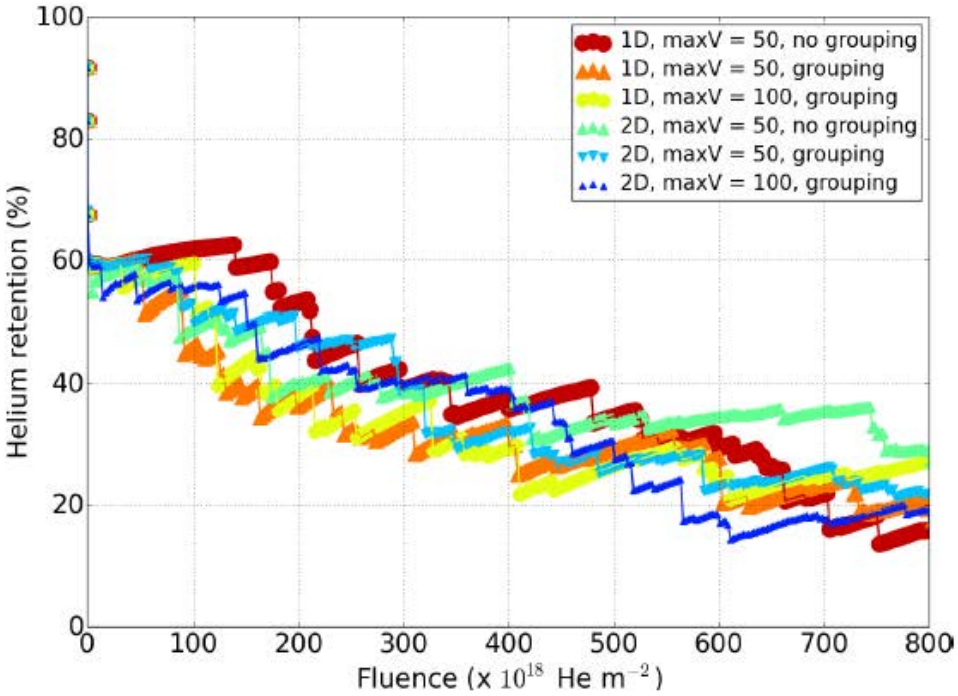
Using MD to 'train' continuum scale (Xolotl)

MD simulations – $\Gamma \sim 5E27\ m^{-2}s^{-1}$



MD

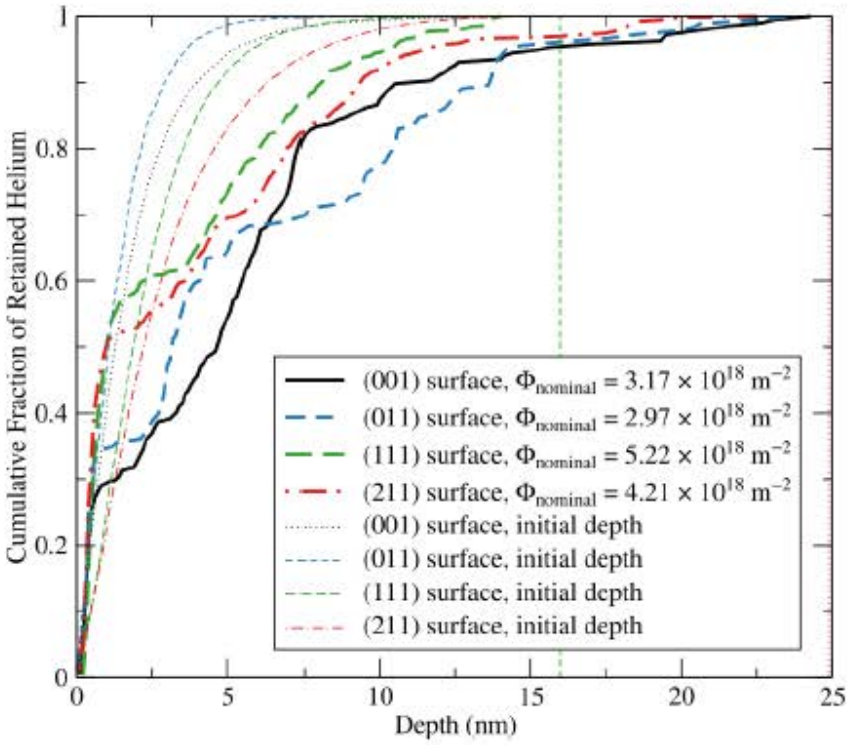
Xolotl simulations



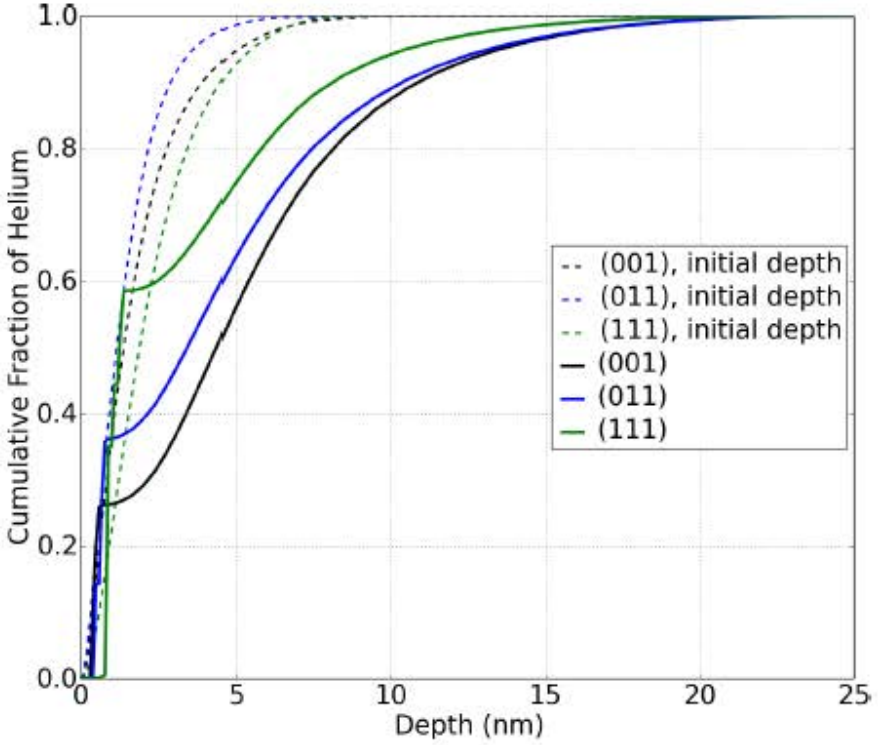
Xolotl

Using MD to 'train' continuum scale (Xolotl)

MD simulations – $\Gamma \sim 4E25\ m^{-2}s^{-1}$

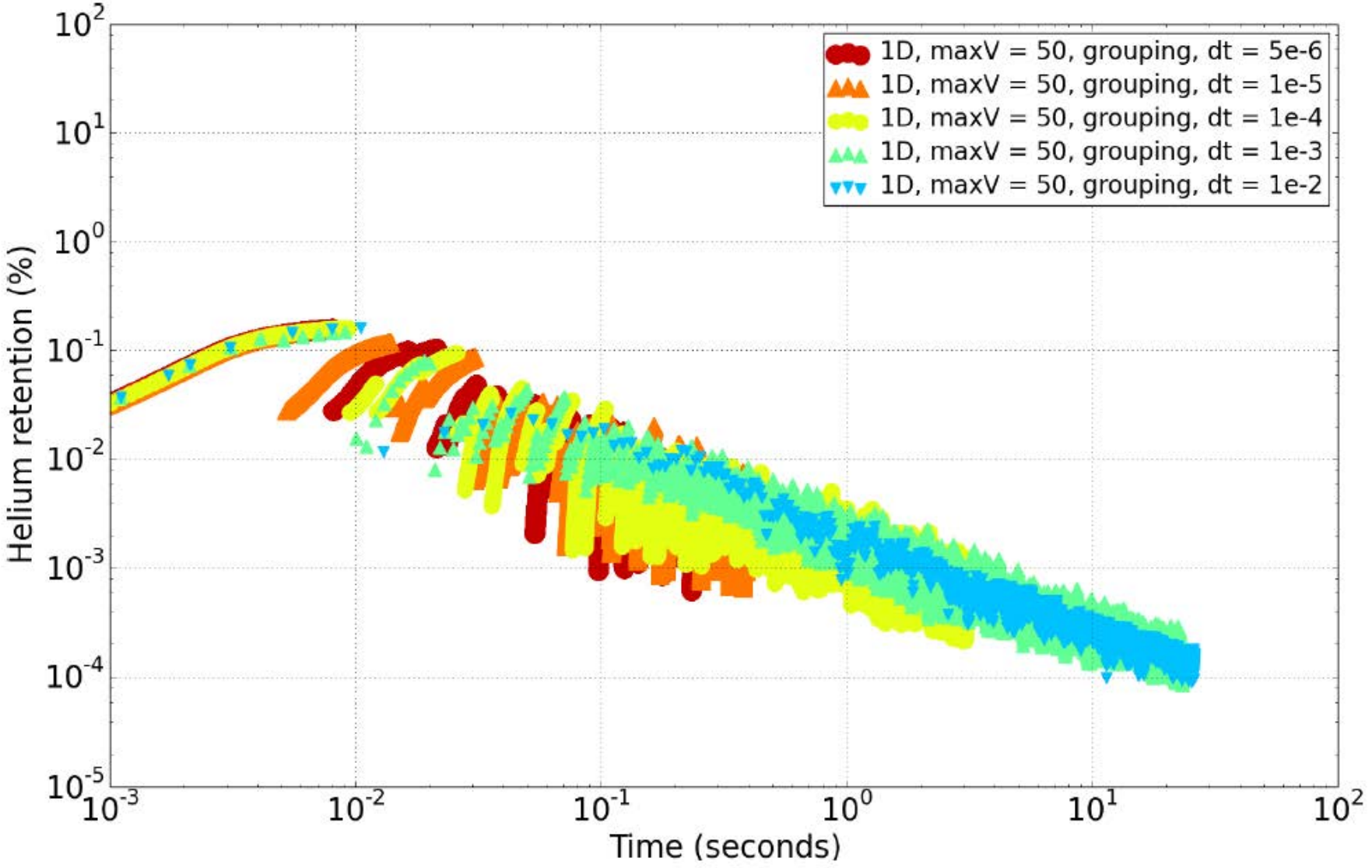


Xolotl simulations



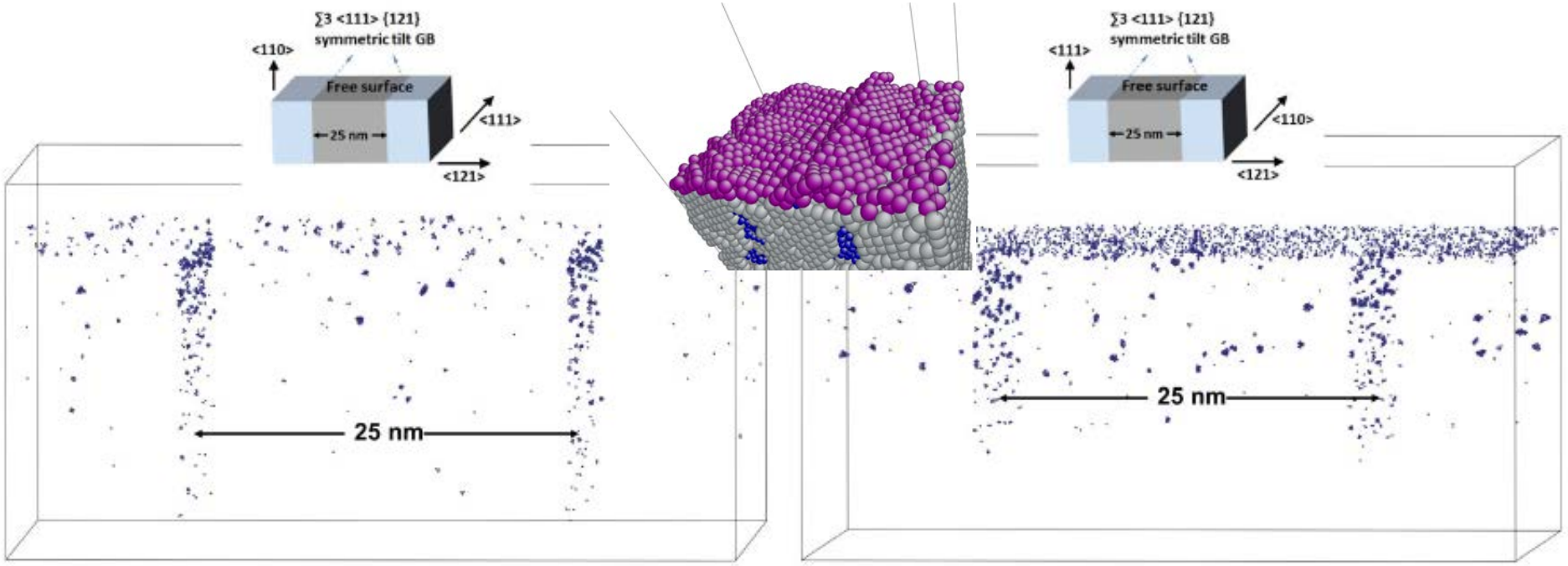
Xolotl now calculates experimental timescales

Flux $= 4 \cdot 10^{22}$, W100.



MD database demonstrates drift and segregation of He

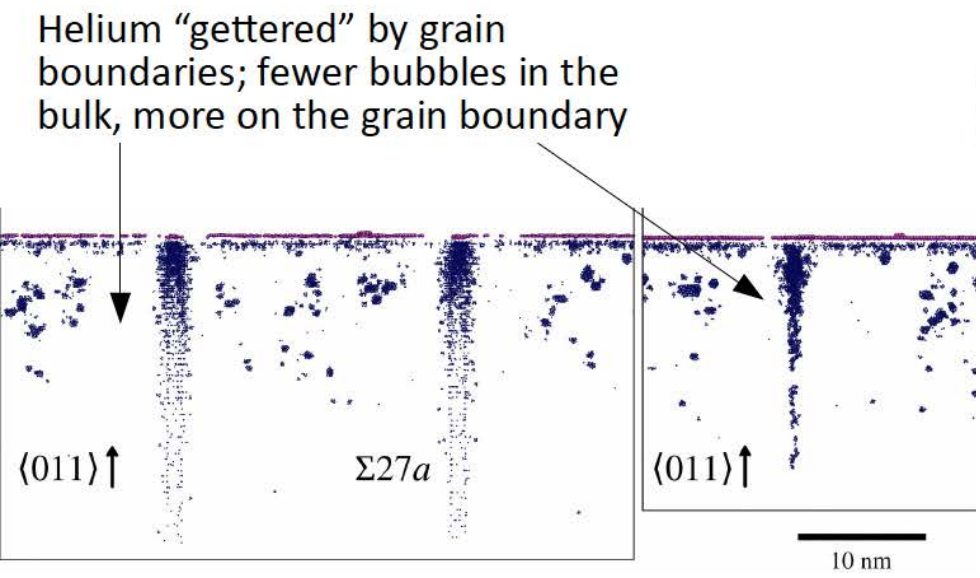
- Large-space-scale molecular-dynamics (MD) simulations (T = 933 K) of evolution of implanted He in model polycrystalline tungsten
- Simulations reveal helium aggregate formation and growth, as well as mobile He_n cluster diffusion, drift, and segregation on W surfaces and GBs
- Example: He distribution near GB intersections with W{110} and W{111} surfaces. Helium distribution determined by sink segregation strength, as well as trap mutation reactions that are activated as the clusters approach the sinks

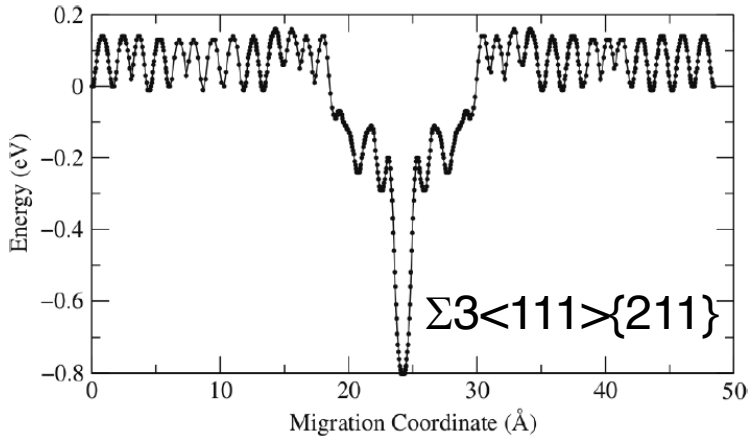


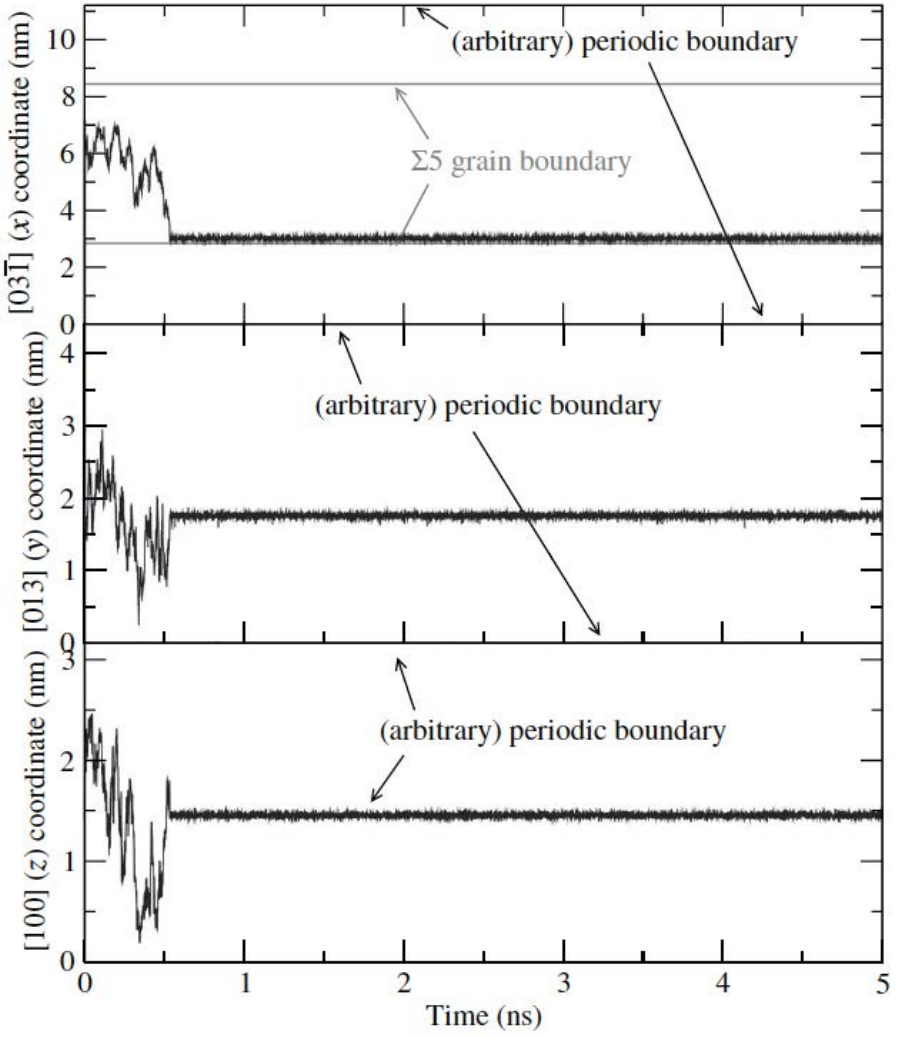
Hu, Hammond, Wirth, and Maroudas, *J. Appl. Phys.* **115**, 173512 (2014).

Significantly reduced He mobility on W grain boundaries

• Atomistic simulations across representative grain boundaries indicate strong trapping of He at grain boundaries, with significantly reduced He diffusivity along the grain boundary compared to the bulk



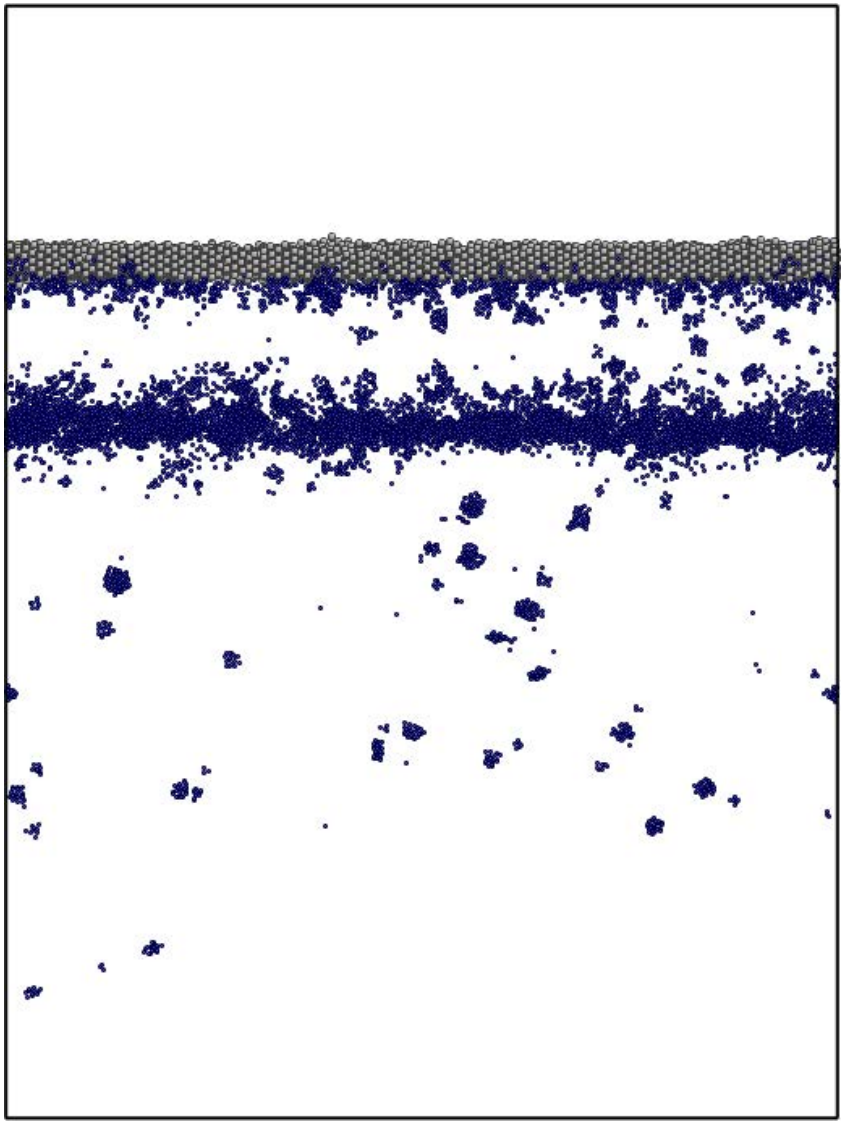




* Hammond, Hu, Maroudas, and Wirth, *Europhysics Letters* **110** (2015) 52002.

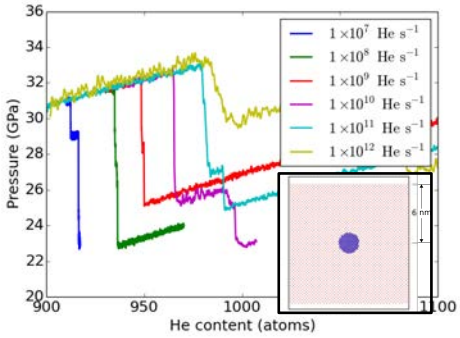
But, He is not completely trapped by W grain boundaries

- Grain boundaries "getter" helium
- We've done several perpendicular to the surface plane
- This one is parallel; helium isn't stopped!



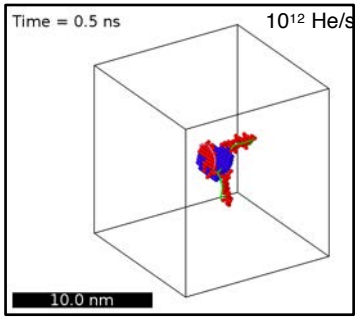
Additional atomistic/Accelerated MD in progress

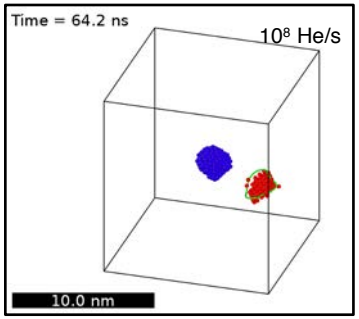
Deep bubble growth



He bubble initially located in a spherical void of 277 vacancies. ~10^5 atoms at 1000 K.

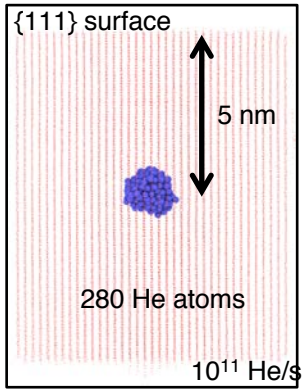
As in the shallow bubble case, slower growth rates favor transitions with lower He content.



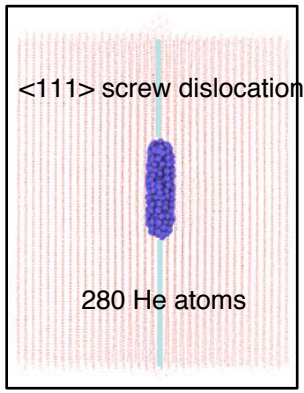


- At the over-driven rates simulated with MD, the tungsten matrix responds differently than at the slower rates representative of experiments.

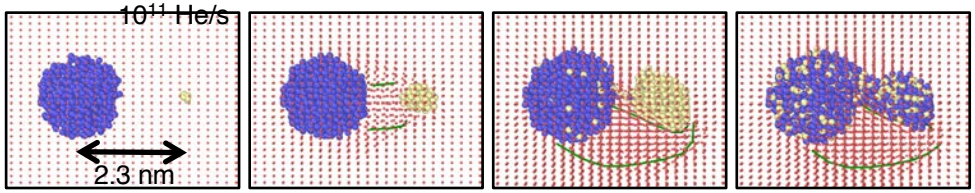
Bubble growth near <111> screw dislocation



He bubble growth process strongly influenced by dislocations, which act as traps. For example, a He bubble nucleated in the at a screw dislocation (right) grows along the core and reaches the surface faster, as compared with the perfect crystal case (left).



Bubble-bubble coalescence



Simulations of bubbles growing in close proximity show a strong directionality of the growth process for the smaller bubble. The coalescence is characterized by the frequent nucleation and growth of connecting dislocations, eventually released from the bubbles as dislocation loops.

Sandoval, Uberuaga, Perez (LANL) unpublished results, in collaboration with BES funding (Voter)

Further Xolotl code development

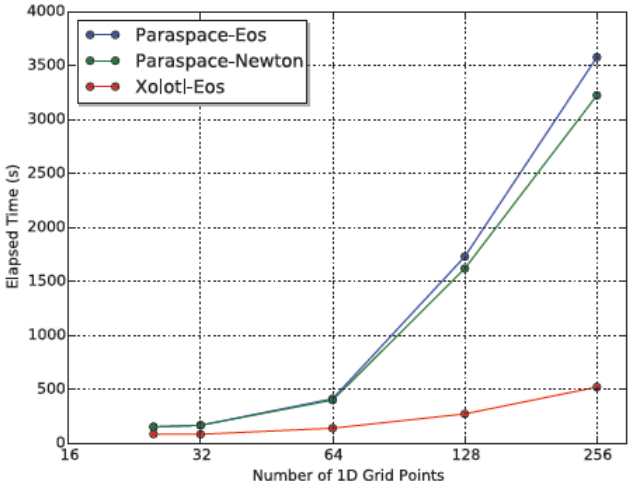
•Verification of Xolotl 1D through cross code comparison against LAMMPS, Paraspace, and KSOME, as well as multiscale integration & benchmarking to large-scale MD

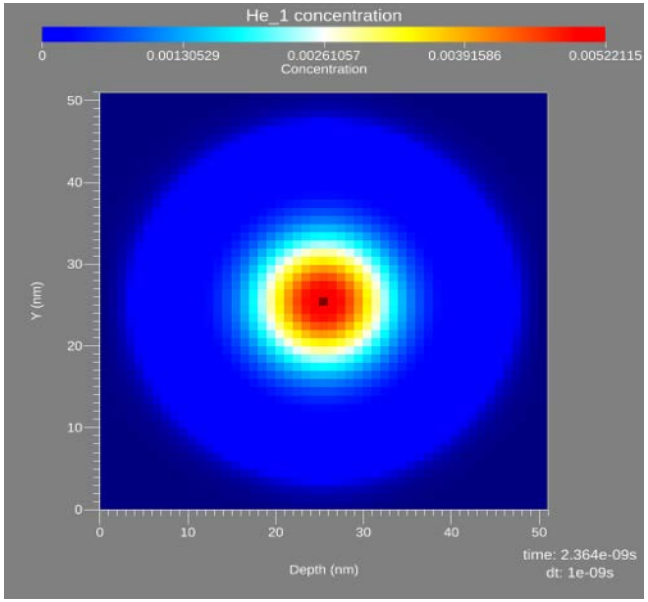
•Performance profiling performed against Paraspace by P. Roth (SUPER)

•Generalization of the system of equations in 2D and 3D, working closely with B. Smith and S. Aithal (FASTMath).

•Significant improvement of the memory usage and performance run-time through strong interactions with B. Smith (FASTMath):

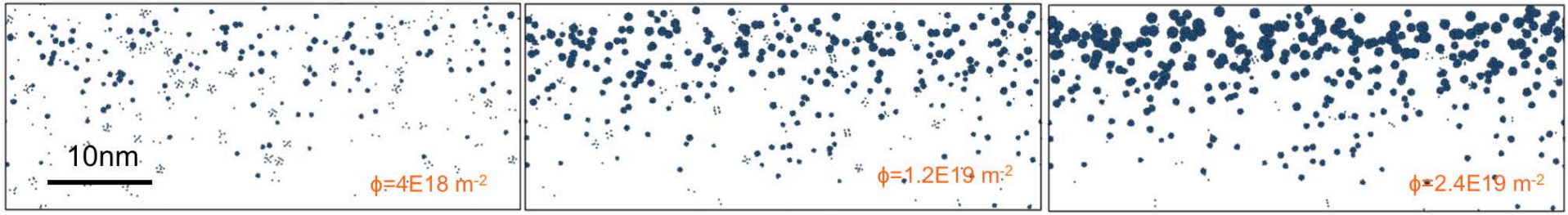
- 4th Order implicit Runge-Kutta ODE integrators with adaptive time steps allows much larger time steps while preserving accuracy
- Composite pre-conditions for linear systems with direct (1d) or multigrid (2 or 3d) solves for the diffusion terms with point-block Gauss-Siedel for reaction solves appears to be optimal and scalable solver for larger problems



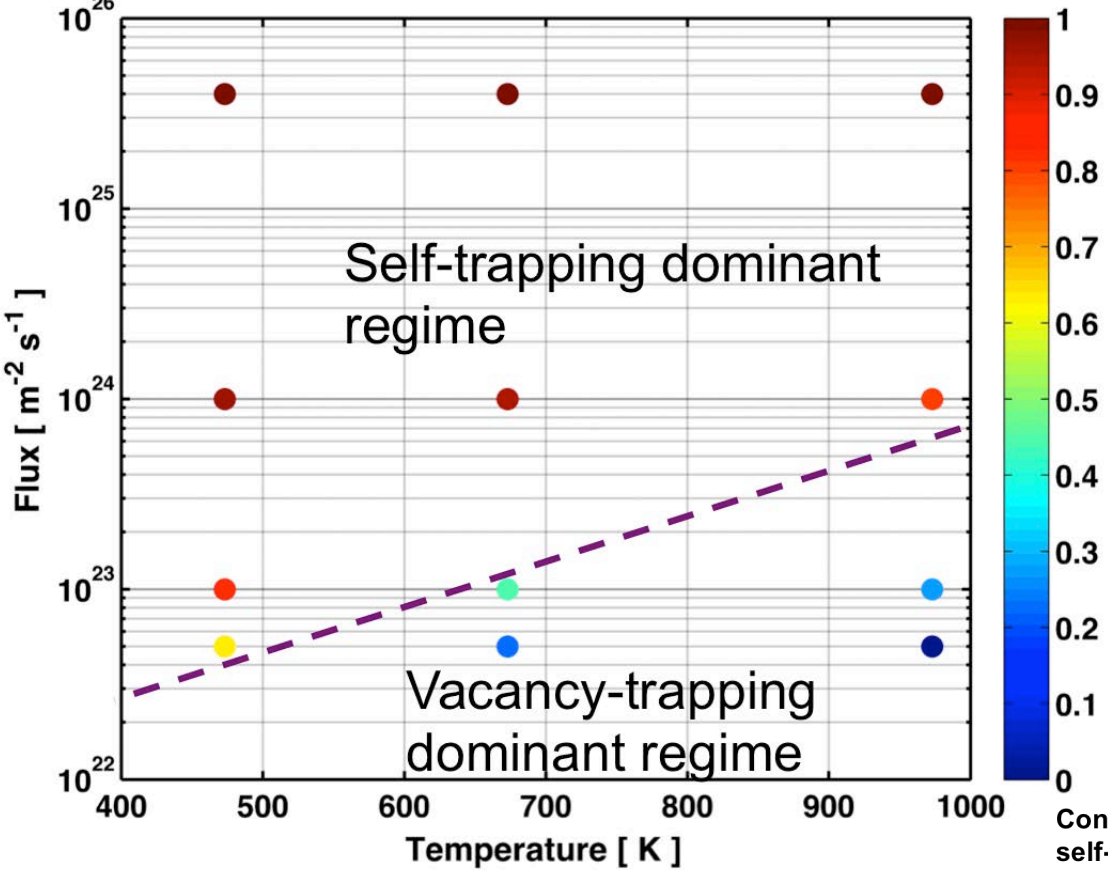


KMC simulation of He clustering below W surfaces

T=973K, Flux (Γ) of 100 eV He at 4E25 He $m^{-2}s^{-1}$



Z. Yang, S. Blondel, K.D. Hammond, B.D. Wirth, *Fus. Sci. Tech.* 71 (2017) 60



Kinetic Monte Carlo (KMC) simulations incorporating atomistic gas diffusion, clustering mechanisms used to extrapolate from ultra-fast MD implantation fluxes to experimentally relevant rates but limited to relatively short times O(seconds).

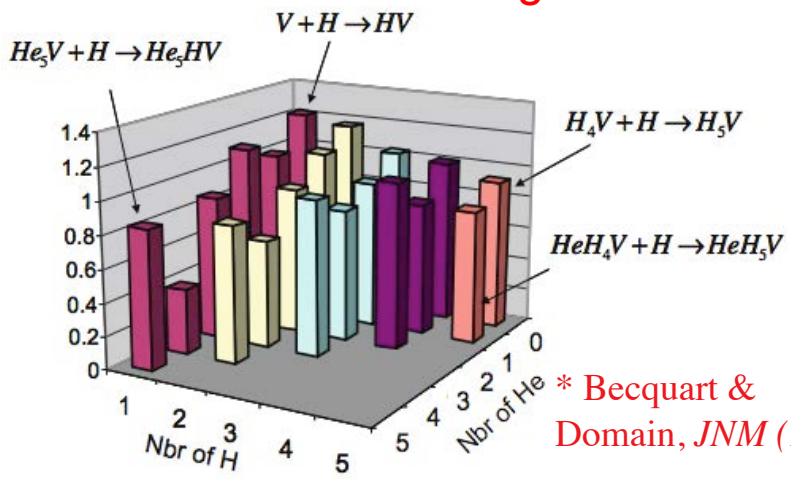
Indicate mechanism boundary of gas bubble nucleation mechanism f(Γ,T)

$$\text{Contribution of self-trapping} = \frac{\text{He retention rate with 0 appm vacancy}}{\text{He retention rate with 5 appm vacancy}}$$

He-H defect interactions in W

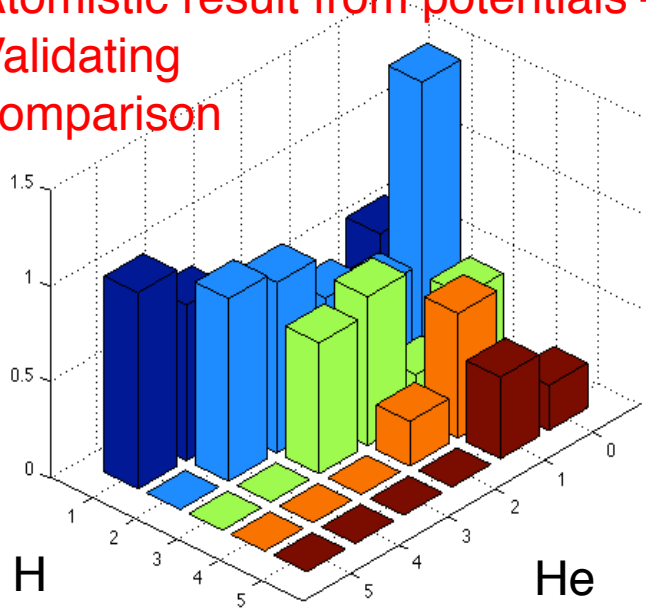
- Interatomic potential(s) derived to describe W-He* and W-He-H** interactions

Ab-initio data of H binding to He-H-V in W*



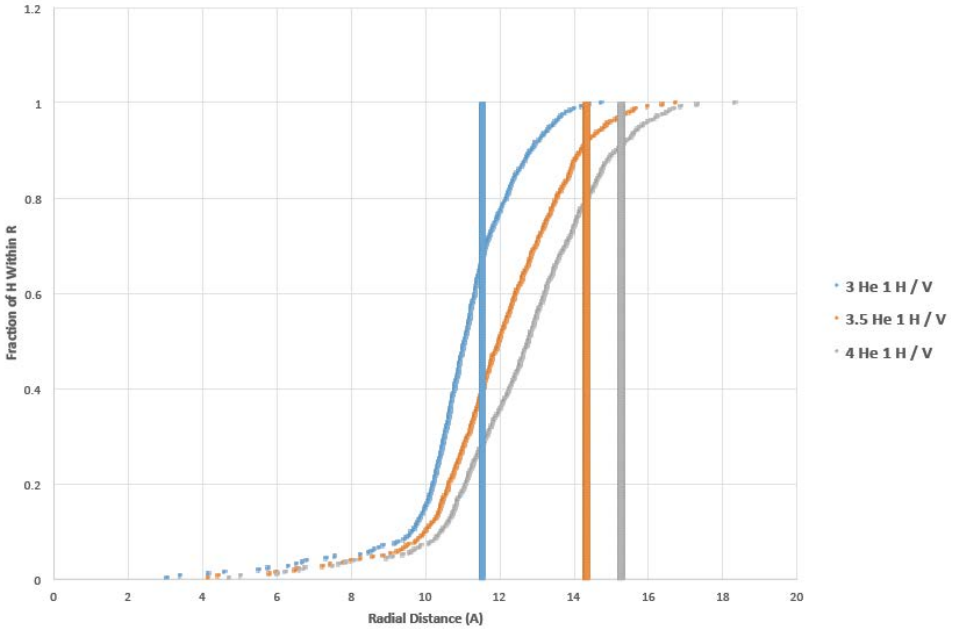
* Becquart & Domain, *JNM (1999)*

Atomistic result from potentials – Validating comparison



Validated potentials used to evaluate H partitioning to sub-surface He bubbles

- He is uniform, but H partitions to the bubble surface
- evaluating H storage capacity as function of bubble size & He pressure

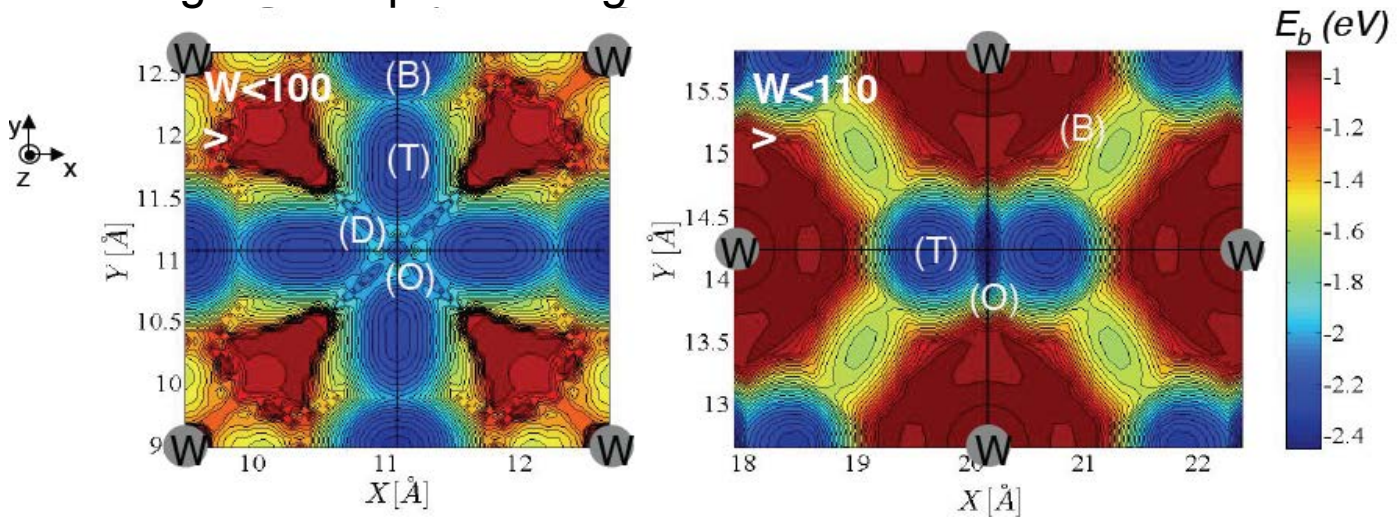


* Juslin and Wirth, *Journal of Nuclear Materials* **432** (2013) 61-66.

** Juslin and Wirth, *Journal of Nuclear Material* **438** (2013) 1221-1223.

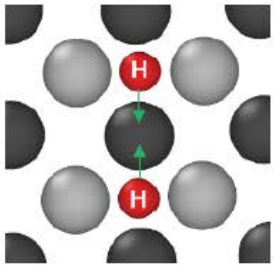
H – W potential validation & modification*

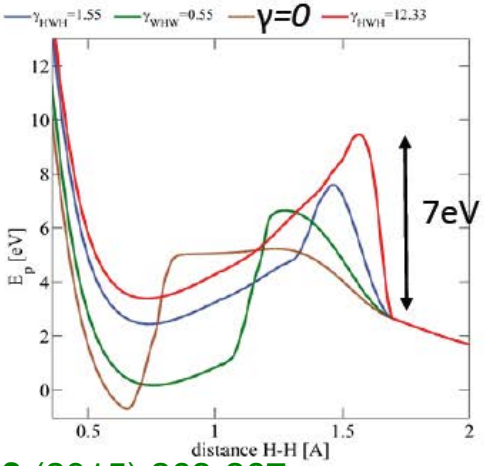
- Benchmarking H adsorption energies on W surfaces

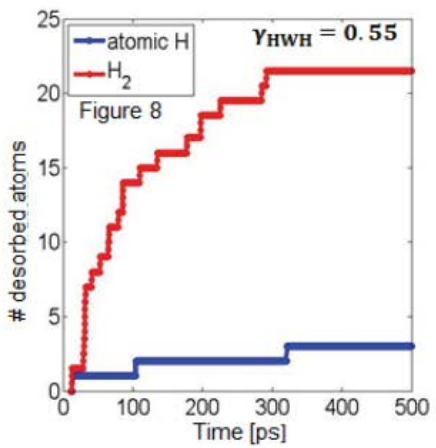


$$E_{ads} = 2.4eV\ (B), 2.3eV\ (T), 2.1eV\ (O), 2eV\ (D)$$

- But MD doesn't indicate H_2 formation and desorption at 2500 K, so 3-body W-H term modified (now called modified Juslin W-H potential), resulting in H_2 desorption







*J. Guterl et al., *J. Nucl. Mater.* **463** (2015) 263-267

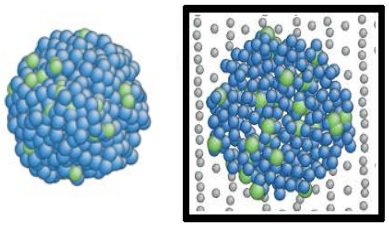
Atomistic modeling of He-H synergies

- MD simulations performed for 2 nm diameter bubble containing high pressure He (3 He/vac) and random distribution of H (0.5 H/vac) at 1800K

- H is observed to rapidly migrate to bubble periphery and remains 'trapped' at the bubble interface

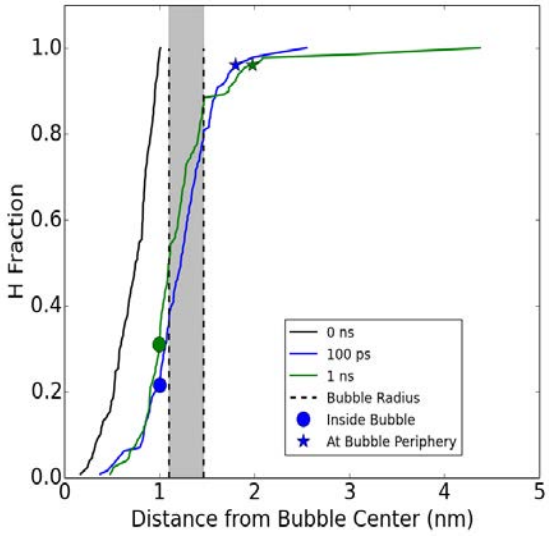
- Raises question about potential for tritium trapping/inventory
  - artifact of interatomic potential
  - short time MD simulations

0 ps

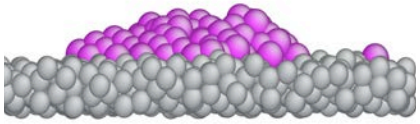


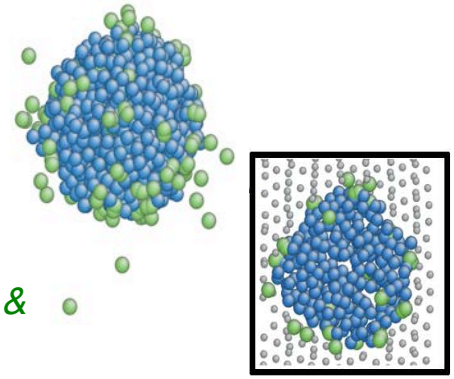
Green: Hydrogen
Blue: Helium
Grey: Surface Tungsten
Magenta: Adatoms

H Distribution for the (111) 1800 K with 3 He/V and 0.5 H/V

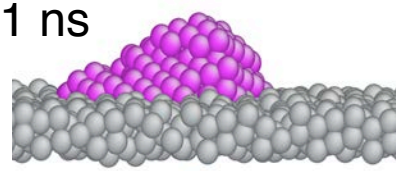


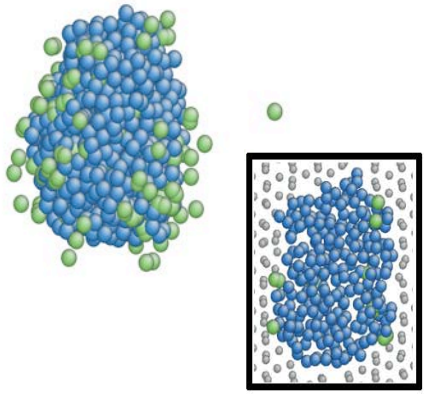
100 ps





1 ns





* Bergstrom, Cusentino and Wirth, *Fusion Science & Technology* ***71*** *(2016) 122-135.*

*Experimental evidence of D/T trapping/retention**

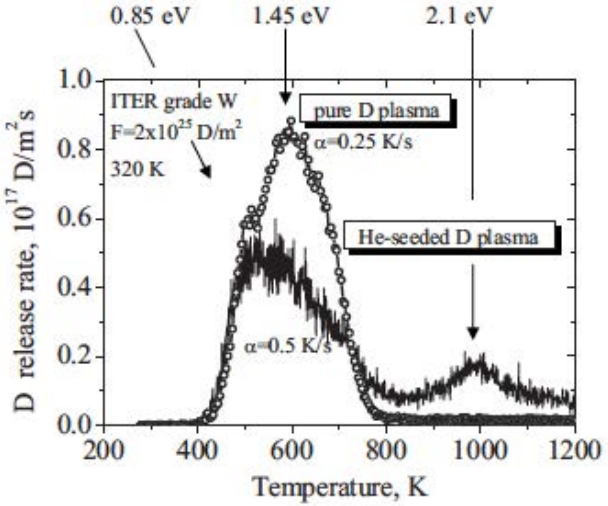
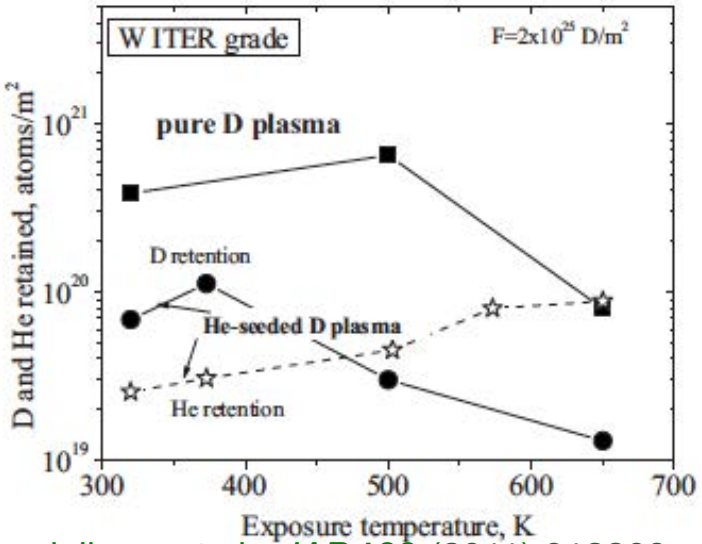
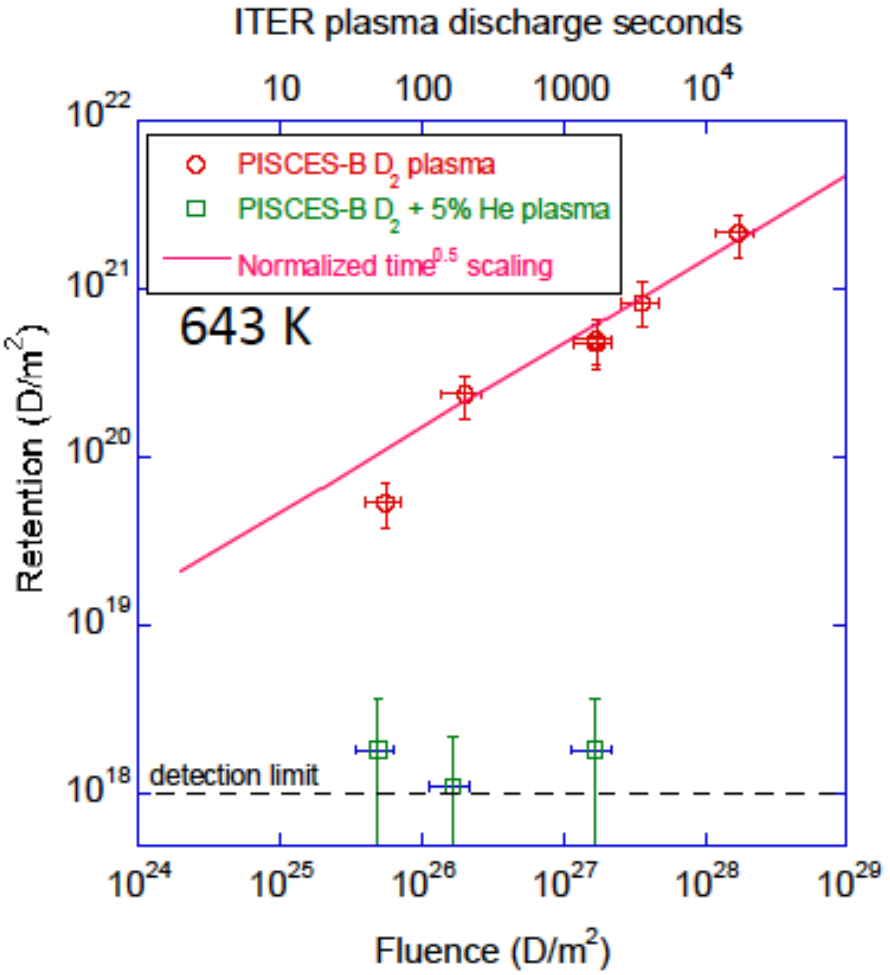


FIG. 7. Thermal desorption spectra of deuterium from polycrystalline W ITER grade exposed to pure and 10% He-seeded deuterium plasma at 320 K up to a fluence of $F=2\times10^{25}$ D/m^2. The temperature ramp is 0.25 K/s for W exposed to pure deuterium plasma and 0.5 K/s for W exposed to 10% He-seeded deuterium plasma. Trapping energies are also shown.



* Ogorodnikova et al., *JAP* **109** (2011) 013309.



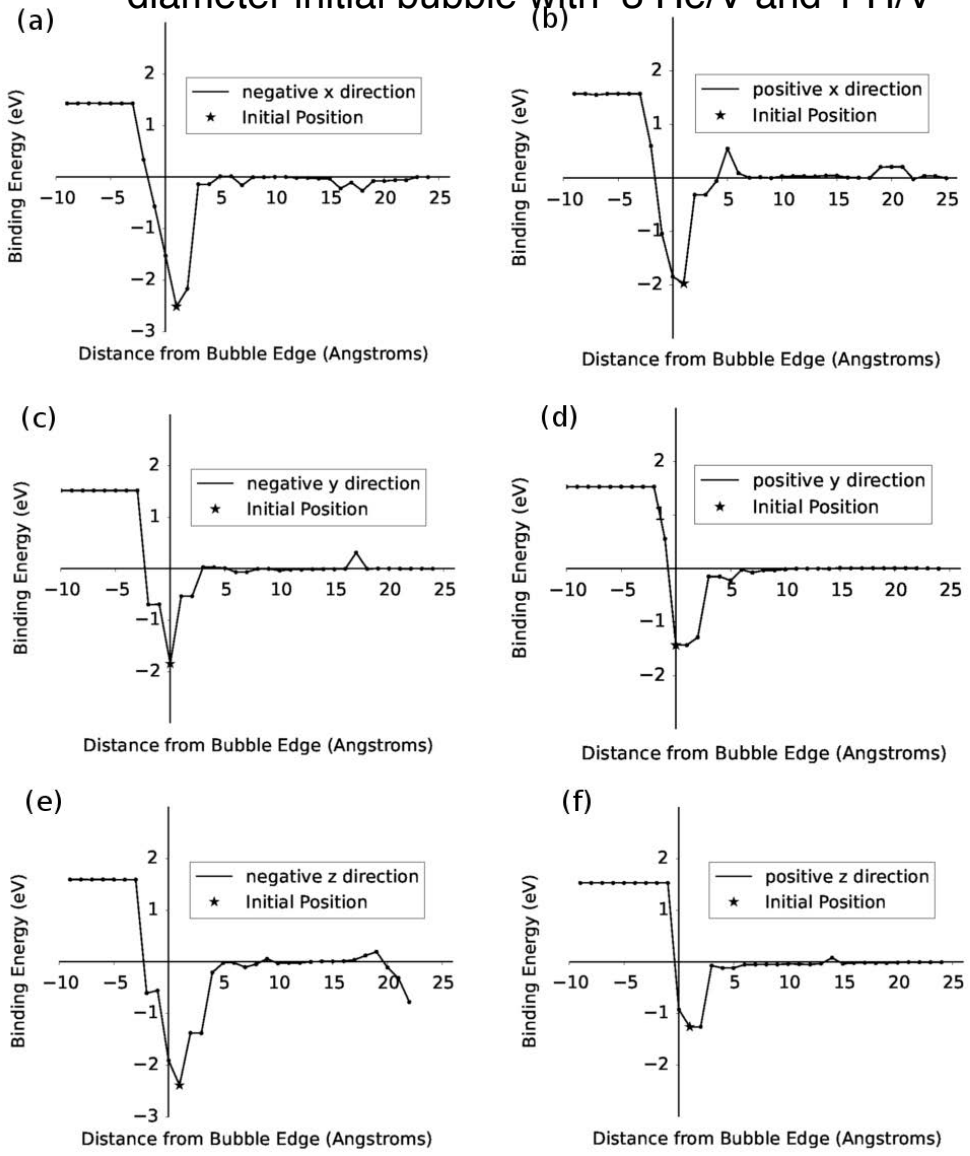
"D not retained in He/D exposed plasma (NRA measurements) following 1000° C"

* Doerner et al., ICFRM19 & PSI2016, personal communication

*Binding energy of H to the He bubbles**

- Simulations quenched to 0 K then hydrogen atoms moved along the [100] direction towards and away from the bubble in 0.1 nm steps with energy minimization
- Plots are normalized such that the energy of hydrogen far from the bubble is 0 on the y axis and the edge of the bubble is 0 on the x axis
- A well of approximately 1.5-2.5 eV that is roughly 1-2 lattice units thick exists near the bubble periphery while there is a high potential energy within the bubble
- A binding energy of ~ 2 eV and an activation energy of ~0.3 eV (migration energy of interstitial H), implies desorption Temperatures > 1000 K ---- the energy needed to overcome this potential well could be as high as ~2.3 eV

H binding energies for (111) surface with 2 nm diameter initial bubble with 3 He/V and 1 H/V



* Bergstrom, Cusentino and Wirth, *Fusion Science & Technology* ***71*** *(2016) 122-135.*

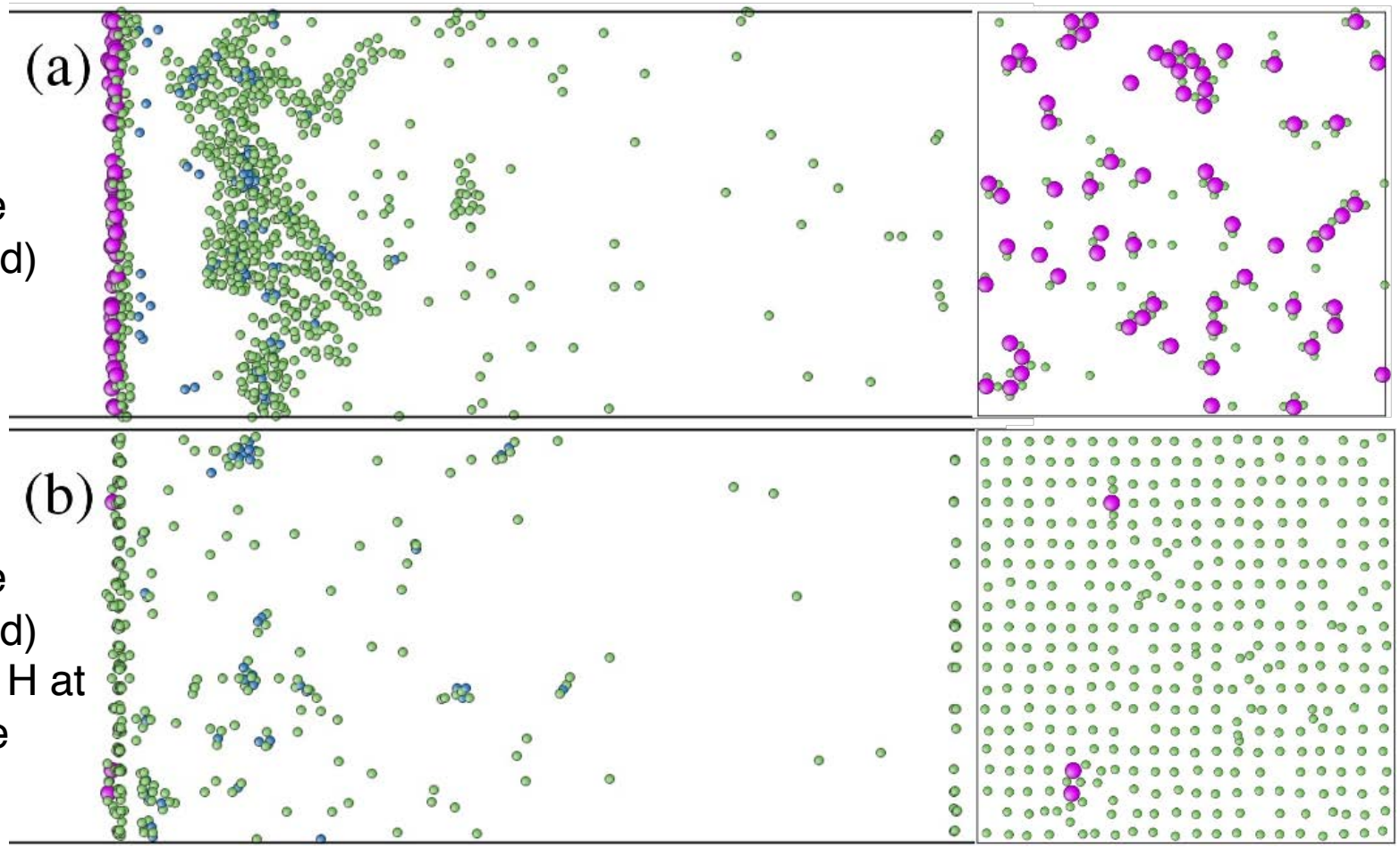
H partitioning to He bubbles

- H/He implantation below W surface at 1200 K (Γ ~4x10^{27} m^{-2}s^{-1}) --- dramatically different H behavior and He/H clustering depending on W-H potential (900H/100He implantation shown below)

Green = H
Blue = He
Purple = W adatoms

Juslin BOP (~85% H & He retained)

Li BOP (~50% H & He retained) atomic H at surface hollow sites

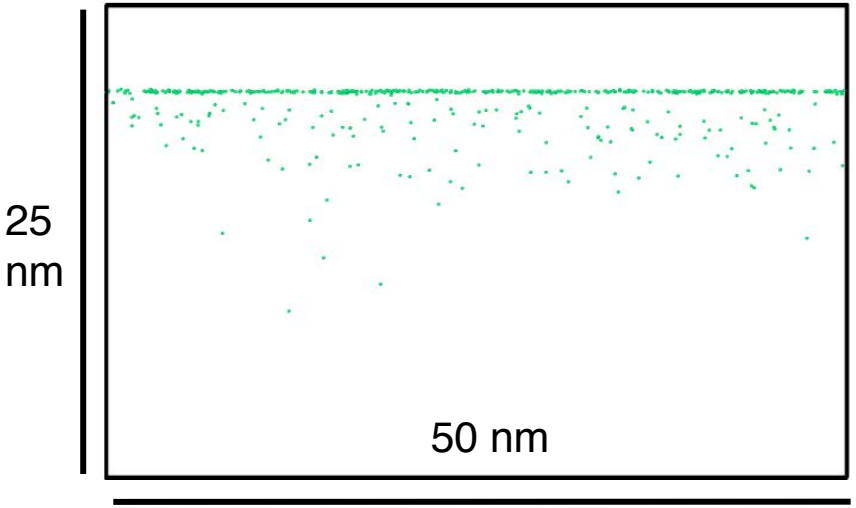


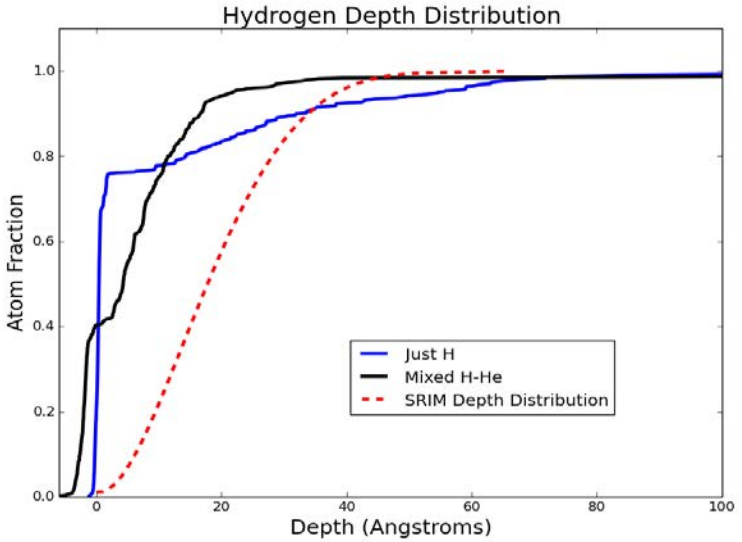
* Cusentino, Hammond, Sefta, Juslin, and Wirth, *Journal of Nuclear Materials* **463** (2015) 347-350.

H partitioning to He bubbles

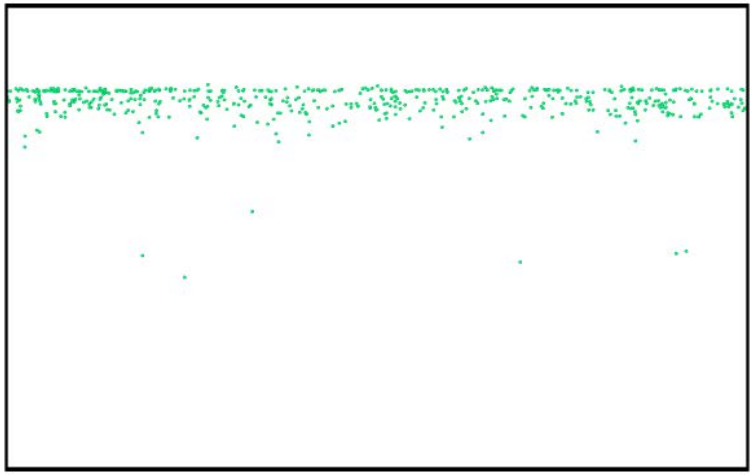
- Introduced H into pre-He implanted Tungsten (MD simulations)

Pure Tungsten

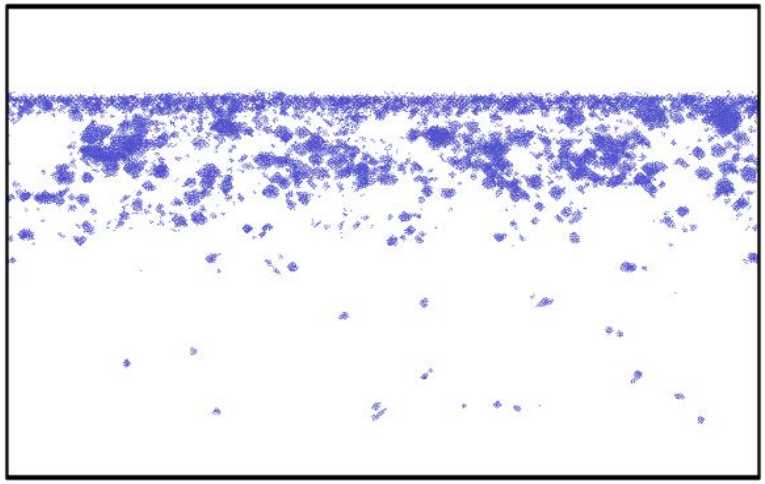




Helium Pre-Irradiated Tungsten



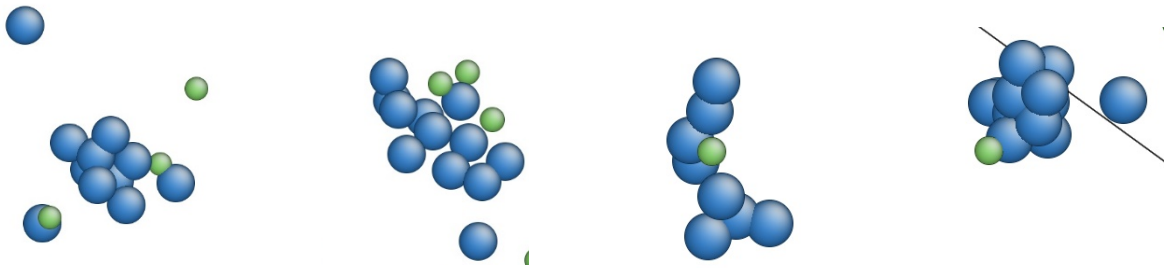
Initial Helium Distribution



Green: Hydrogen **Blue:** Helium

H binding to He clusters (observed clusters from MD)

Snapshots of H – He cluster interactions in MD simulations with pre-implanted He (green is H, blue is He)

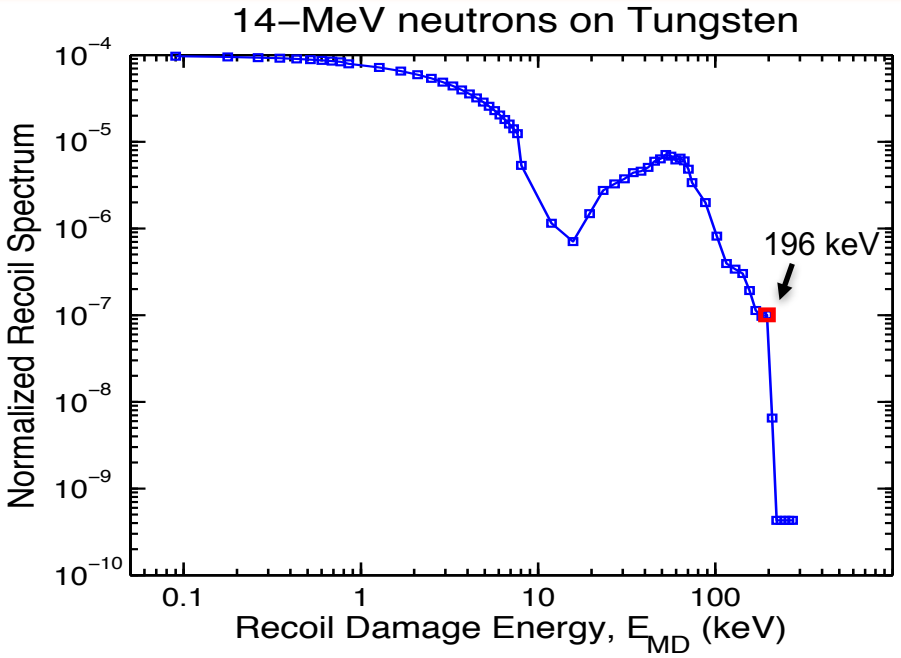


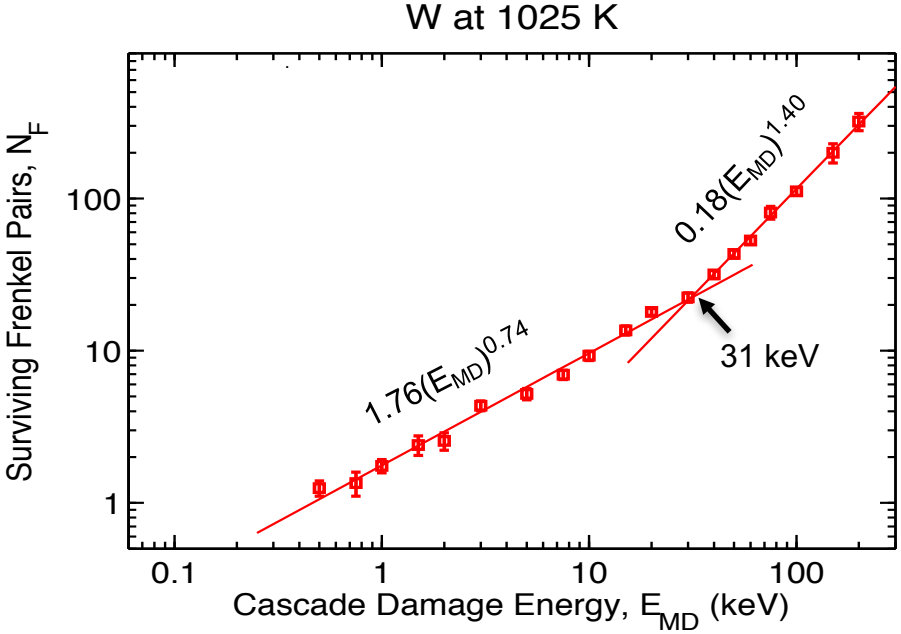
Cluster Size	Binding Energy (eV)
$He_{11}H_1V_2$	2.41
$He_{11}H_3V_3$	0.05
$He_3H_1V_1$	1.63
$He_8H_1V_3$	2.50

- 60% of H is located in near-surface helium cluster saturated layer
- 37.5% of this H is located near a helium cluster/bubble (using a search algorithm & interaction cutoff distance of 0.32 nm)
- Remainder of H is mostly atomic with a few H_2 molecules

Modeling Cascade Damage in Bulk Tungsten

- Spectrum of W PKAs due to 14-MeV neutrons shows a significant number of PKAs up to 280 keV of recoil energy or 196 keV of damage energy (E_{MD})
- Previously, primary defect damage database includes E_{MD} up to 100 keV
- New displacement damage data generated at 150 and 200 keV for 300, 1025, and 2050 K
- Data at 150 and 200 keV follow the trend of defect production curve (N_F) for E_{MD} > 30 keV
- KMC simulations of irradiation damage accumulation due to 14 MeV neutrons are currently underway





* Setyawan, Selby, Juslin, Stoller, Wirth and Kurtz, *J. Phys. Cond. Matter* **27** (2015) 225402.

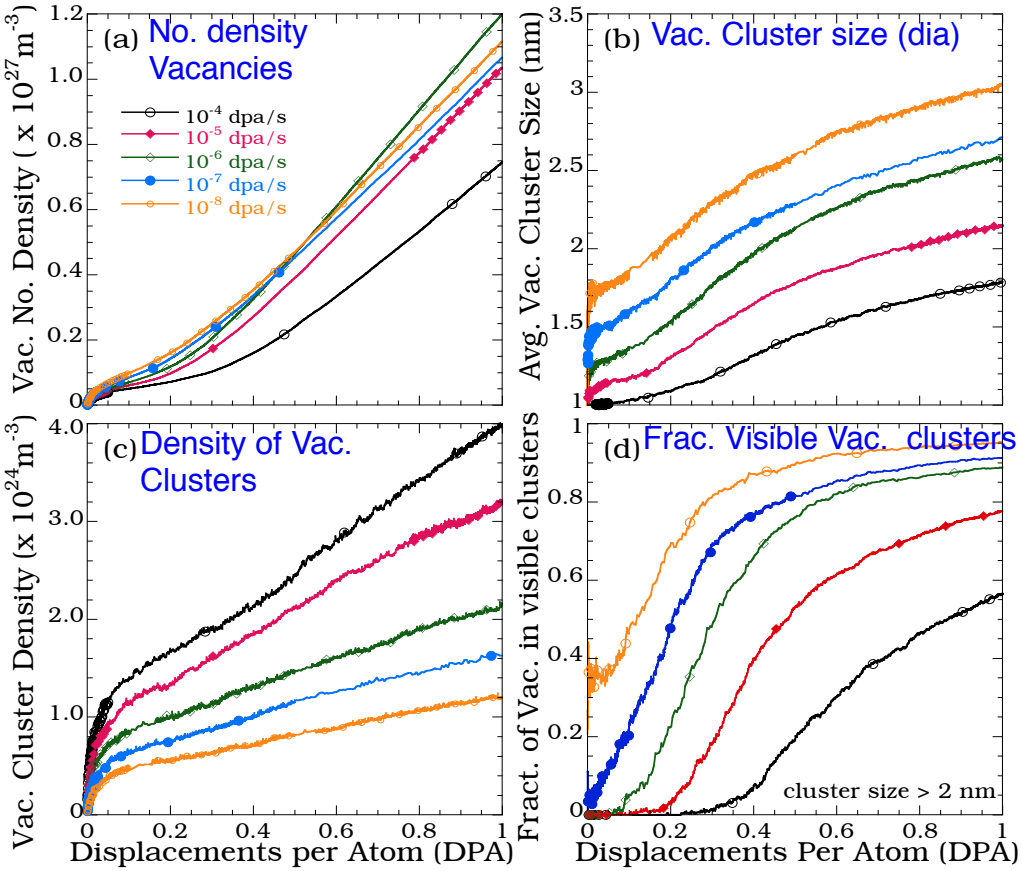
KSOME: Kinetic Simulations of Microstructural Evolution

- KSOME is an object kinetic Monte Carlo code to simulate the evolution in time and space the distribution of lattice defects in crystalline materials accumulated during irradiation
- KSOME is designed to simulate the diffusion, emission, transformation and reaction events of vacancies, interstitials, impurities and their complexes, including any number of combinations of point defects, for bcc and fcc lattices at a given temperature
- Cascade insertion is random, based on a specified cascade production rate and additionally allows for creation of any type of defect based on their production probability
- Interaction of mobile defects (simple absorption) with sinks such as dislocations, grain boundaries and free surfaces
- Defect parameters like type, size, orientation, etc. are used to identify defect diffusion, emission, reaction and transformation events between various types of defects
- Example events:
  - → Rotation or change of direction of 1D-diffusing SIA clusters
  - → Vacancy loop transforming into a spherical cluster or void
  - → Emission can also be associated with a loop punching or trap mutation event
  - → Allows emission of multiple (size or type) defects from a single source
- Simulation of radiation damage according to HFIR PKA spectrum and migration parameters

Dose Dependence of Vacancy Cluster Densities and Sizes

- With increasing dose rate:
  - Number density of vacancies increases
  - Vacancy cluster density decreases
  - Average vacancy cluster size decreases
- Fraction of visible clusters:
  - 10^{-8} dpa/s - saturates at 95% of the vacancy population
  - 10^{-4} dpa/s –reaches 55% of the vacancy population at 1 dpa
  - Visible clusters - 2 nm diameter or about 300 vacancies
- Vacancy cluster sizes at 10^{-8} dpa/s:
  - Grow larger than at higher dose rates due to the greater time between cascade insertions permitting more defect diffusion
  - Di-vacancies are not stable, which suppresses nucleation of new clusters
- No formation of SIA clusters
  - SIAs quickly diffuse to grain boundaries
  - SIAs are more likely to recombine with the increasing population of vacancy clusters

Dose rate has significant effect on void growth

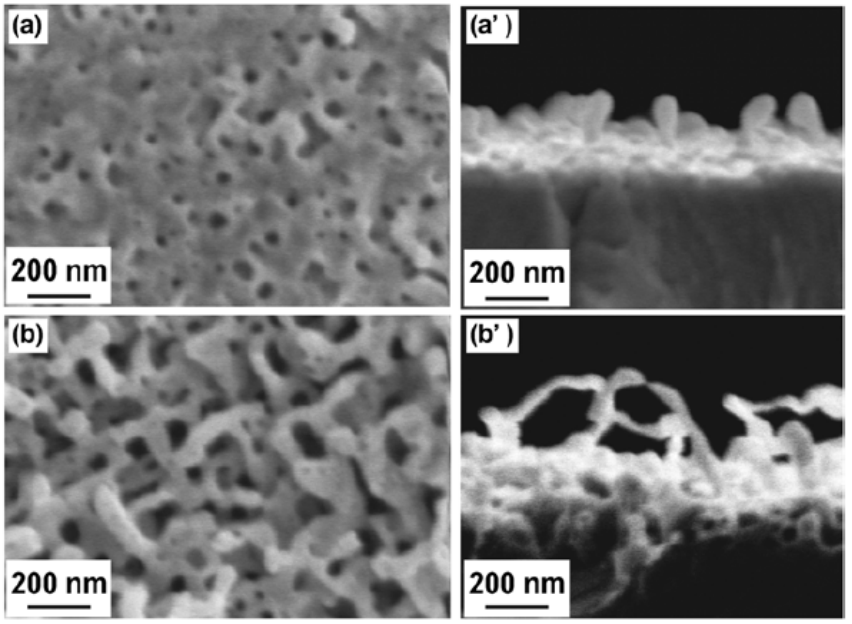


Summary & Future Work

• Fuzz formation mechanism remains to be conclusively determined; but significant hints that helium gas bubble formation, agglomeration/coalescence and bursting phenomena play a key role in driving initial W surface roughening

- MD reveals initial surface roughening by adatom formation (trap mutation) followed by adatom island (bubble expansion by loop punching) and subsequent bubble bursting & indicates H/D/T trapping at He bubbles

•Kinetic models (Xolotl-PSI) now bench-marked to predict He bubble R, N & P as a function of He exposure conditions & models for W defect/loop/surface adatom diffusion to model both bubble formation, evolution & topology changes – framework for coupling to plasma edge is emerging



* Kajita, *Nuclear Fusion* **49** (2009) 095005.

• Key uncertainties: He implantation rate effects; influence of temperature/stress gradients (TRANSIENTS)

- What mechanisms transform W surface instability into tendril filaments of 10-100 nm diameter?

- T storage & retention in He bubbles & W-He-H(+C, Be, impurity) materials