Multiscale Materials Modeling of low-energy He plasma surface interactions with Tungsten

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with significant contributions from
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Oak Ridge National Laboratory

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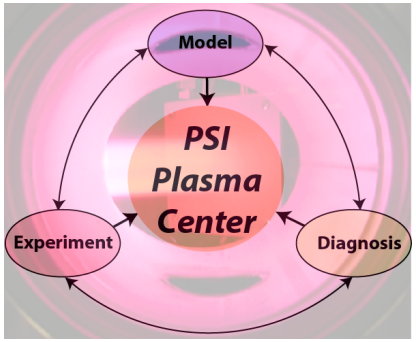

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*Complex, interlinked PSI phenomena**

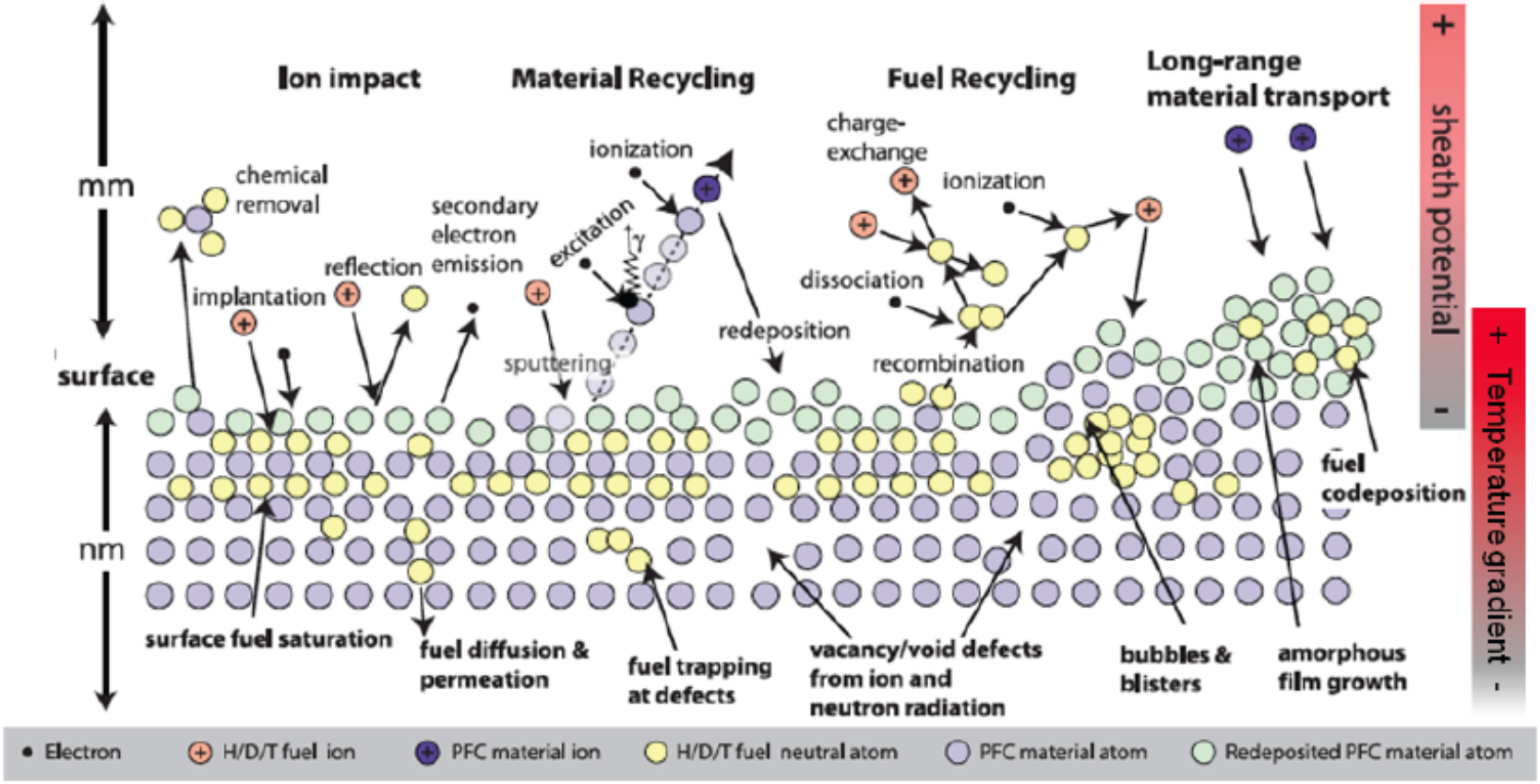


Figure of merit:
Incident plasma ion flux near divertor strikepoint: 10^{24} $m^{-2}s^{-1}$

Steady-state sputtering yield O (10^{-4}) on surface monolayer (10^{19} atoms/m^2) results in sputtering of every atom every 0.1 sec -> every atom sputter >10^8 times/year

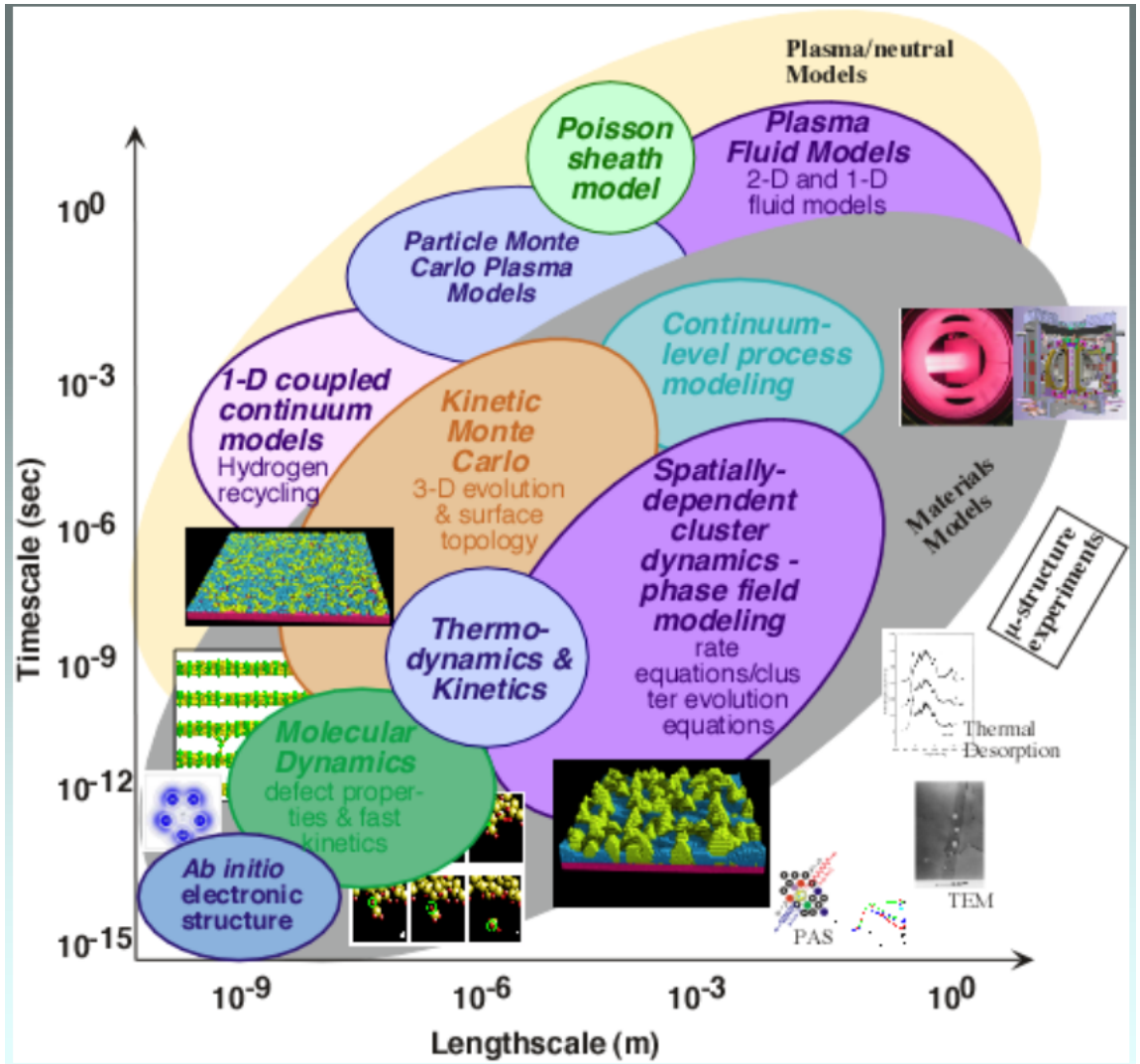
* Wirth, Nordlund, Whyte, and Xu, *Materials Research Society Bulletin* **36** (2011) 216-222

*Multiscale modeling capability – a work in progress**

Goal: Discovery science to identify mechanisms/clues to W nanofuzz formation and synergies between He & H exposure that impact H/D/T permeation & retention

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

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

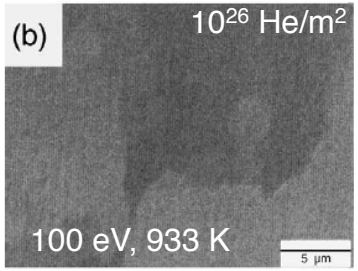


Biggest long-term scientific challenge is understanding the kinetics of coupled defect – impurity evolution with a disparate range of kinetic rates

* Whyte & Wirth, unpublished

W Surface dynamics under combined thermal/particle fluxes

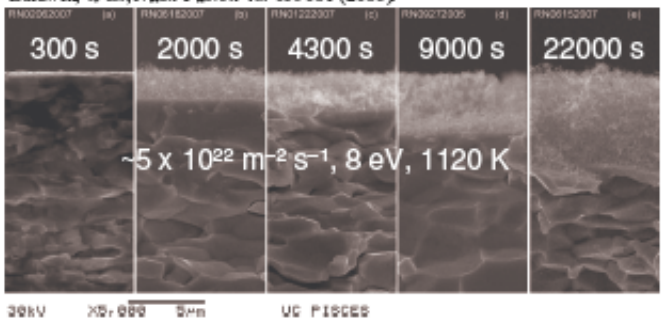
T < 700 K



Tokunaga, Doerner, et. al., *J. Nuc. Mater.* **313-316** (2003) 92

non-specific damage

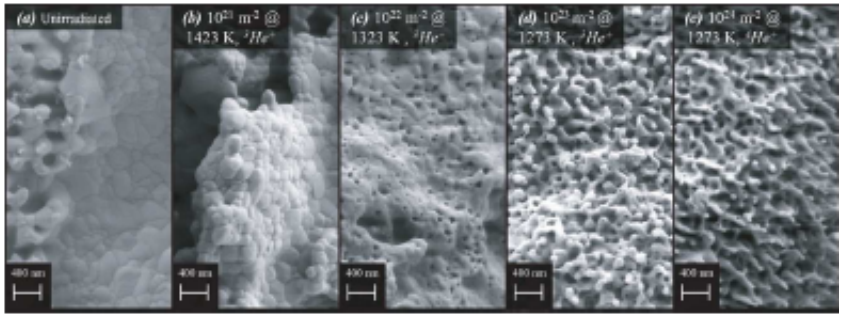
Baldwin, *et al.*, *Nucl. Fusion* 48: 035001 (2008).



900 K < T < 1900 K

fuzz / coral

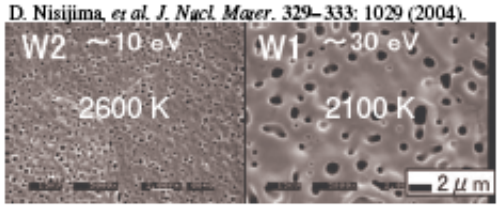
S. J. Zenobia and G. L. Kulcinski. *Phys. Scr.* T138: 014049 (2009).

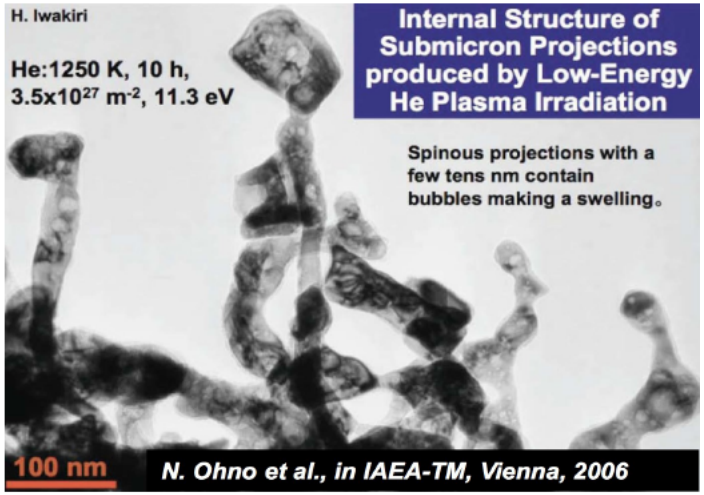


T > 2000 K

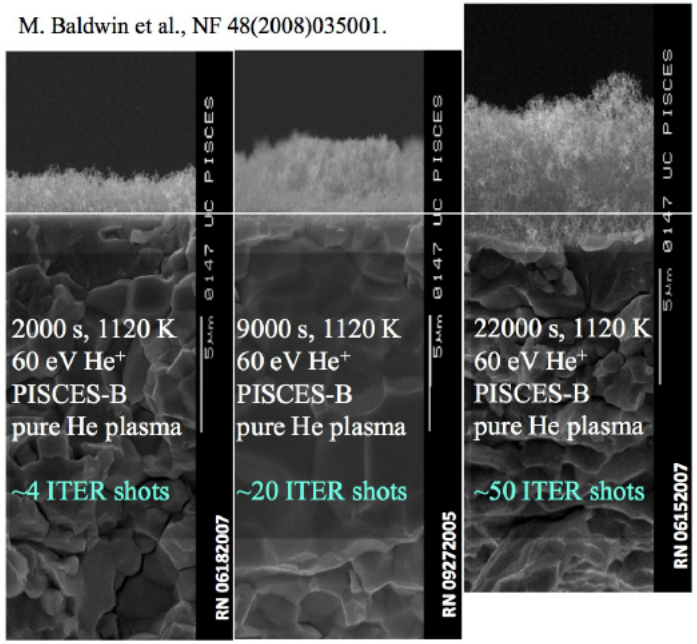
holes

D. Nisijima, *et al. J. Nucl. Mater.* 329–333: 1029 (2004).



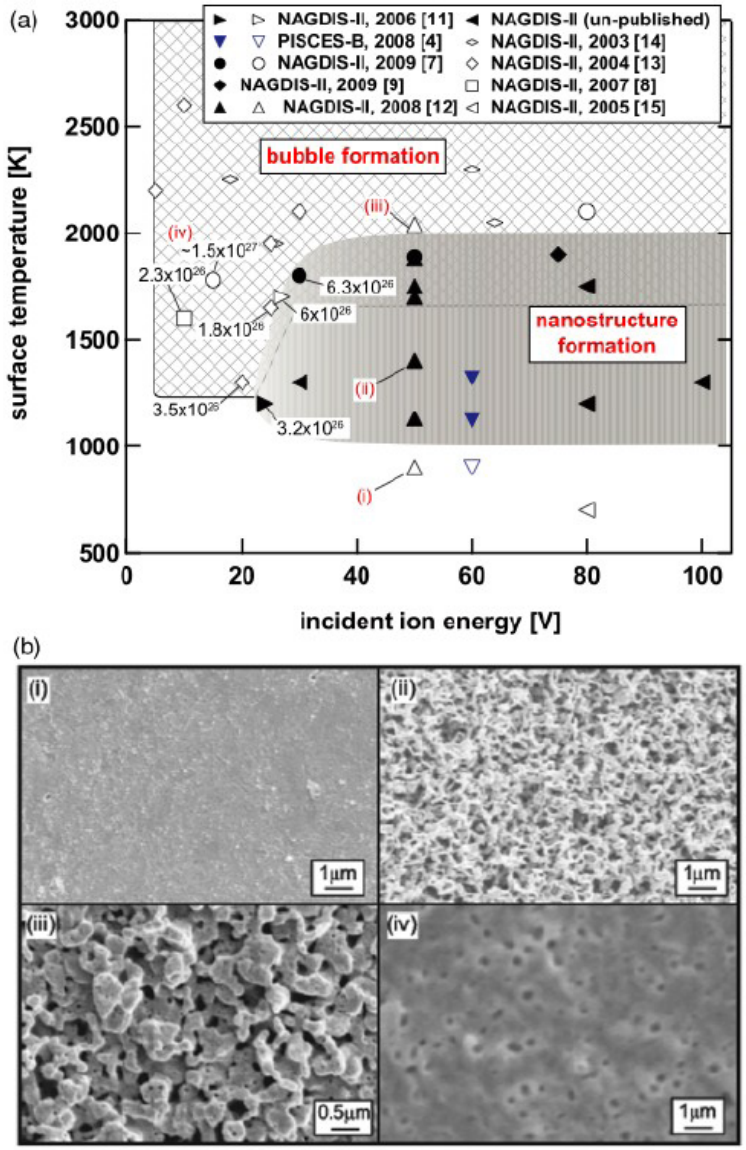


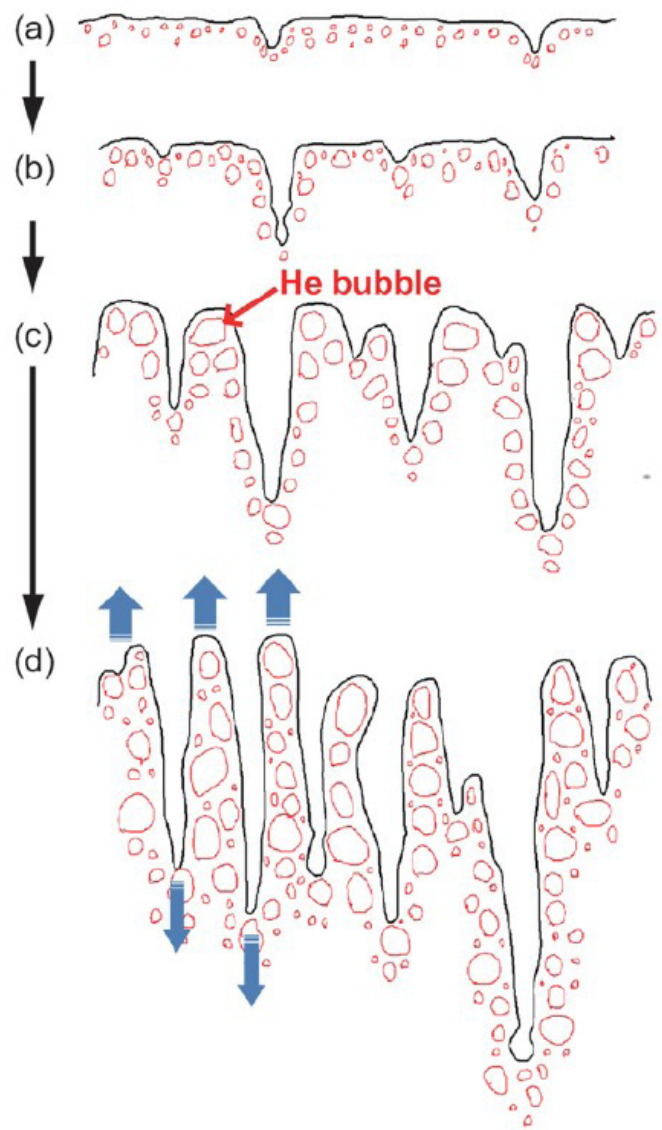
M. Baldwin et al., NF 48(2008)035001.



*Proposed W fuzz formation mechanism**

- **Sub surface He bubbles drive 'finger' instability**





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

*Proposed W fuzz formation mechanism**

- **W 'viscosity' drives transport from below bubble layer driving fuzz growth**

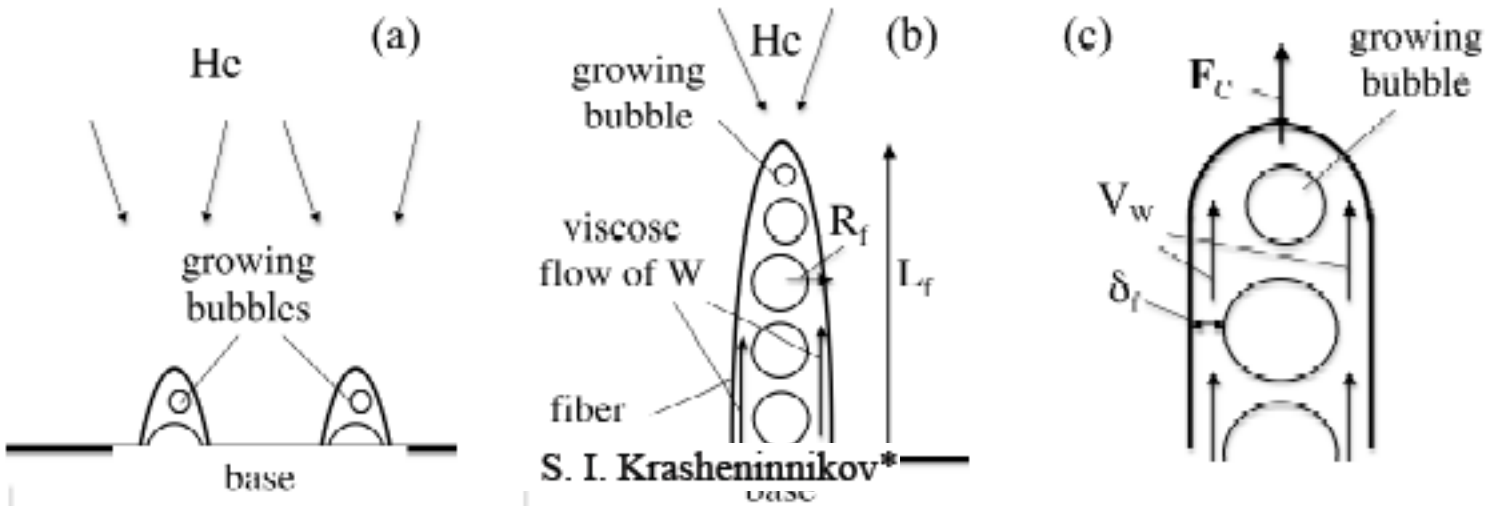


Fig. 2. Schematic views of: (a) initial stage of the fiber growth; (b) developed fiber; (c) viscose flow of W to the tip of the fiber due to the force caused by the pressure of the He in the growing fiber.

$$F_C = P_{He}\pi R_f^2, \tag{2}$$

where P_{He} is the helium pressure in the bubble of radius R_f, which we will assume to be large the thickness of the fiber "skin", δ_f. Helium pressure in the growing bubble can be estimated as

$$P_{He} \sim 2\gamma / R_f, \tag{3}$$

where γ is the tungsten surface tension coefficient.

As a result, the magnitude of the stress in the "skin" can be evaluated as

$$\sigma_0 \sim \frac{F_C}{2\pi R_f \delta_f} \sim \frac{2\gamma}{2\delta_f}. \tag{4}$$

Then substituting expression (4) in Eq. (1) we have

$$\frac{\partial \sigma_{\ell\ell}}{\partial \ell} \sim \frac{\sigma_0}{L_f} = \mu_W \frac{V_W}{\delta_f^2}, \tag{5}$$

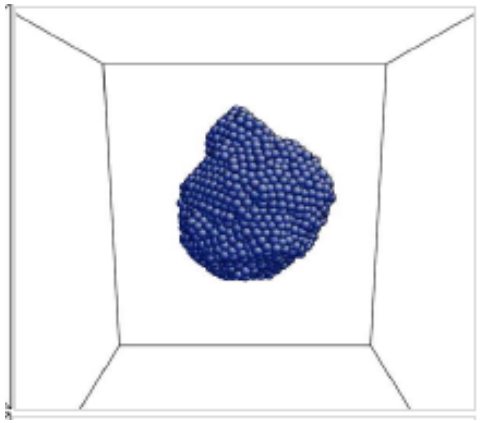
where L_f is the length of the fiber and V_W is the flow velocity of tungsten (see Fig. 2c). Then taking into account that $dL_f/dt = V_W$ from Eq. (5) we find:

$$L_f(t) = \sqrt{\frac{2\gamma\delta_f}{\mu_W} t}. \tag{6}$$

* Krasheninnikov, *Physica Scripta* **T145** (2011) 014040

MD simulations: sub-surface He bubbles

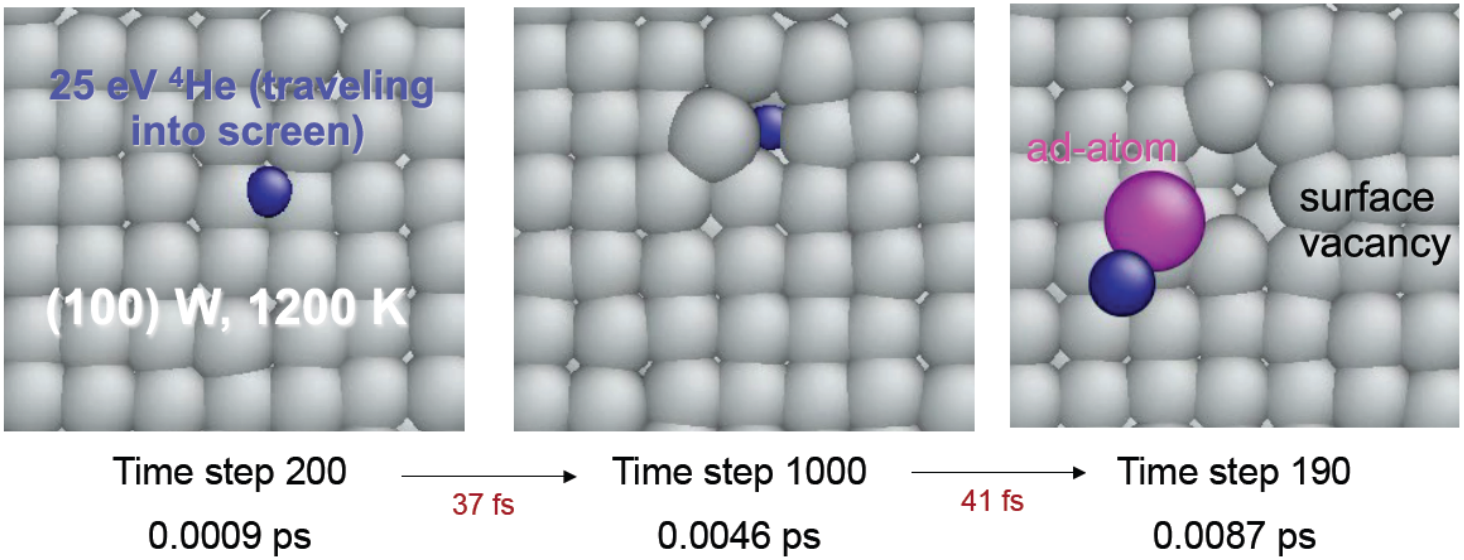
• Evolution of He bubbles below surface: initial nucleation & growth requires a kinetic model (in progress based on learning from MD simulations)



• Evolution of larger He bubbles -> several regimes of interest:

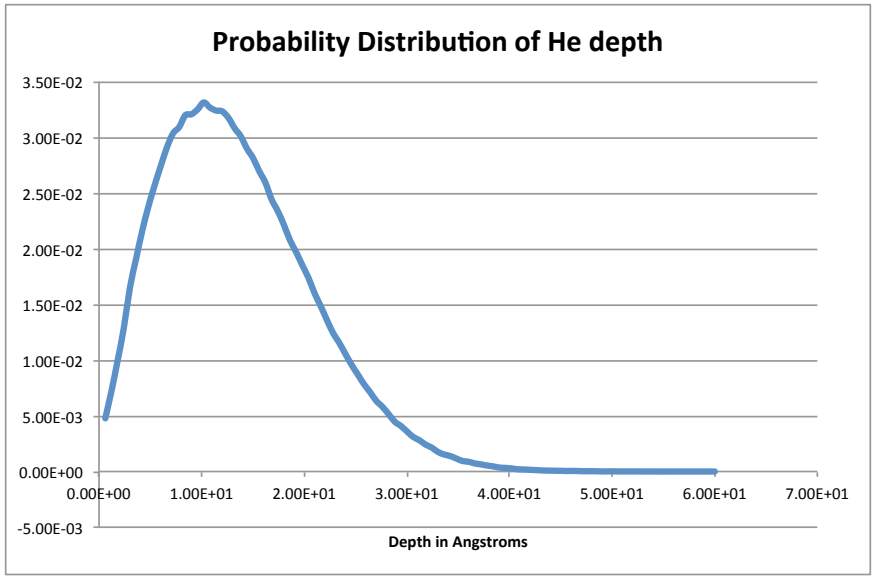
- Equilibrium bubbles (internal gas pressure $P = 2\gamma/R$)
- Over-pressurized bubbles can 'punch loops' ($P = 2\gamma/R + Gb/R$)
- Near-surface, over-pressurized bubbles can rupture

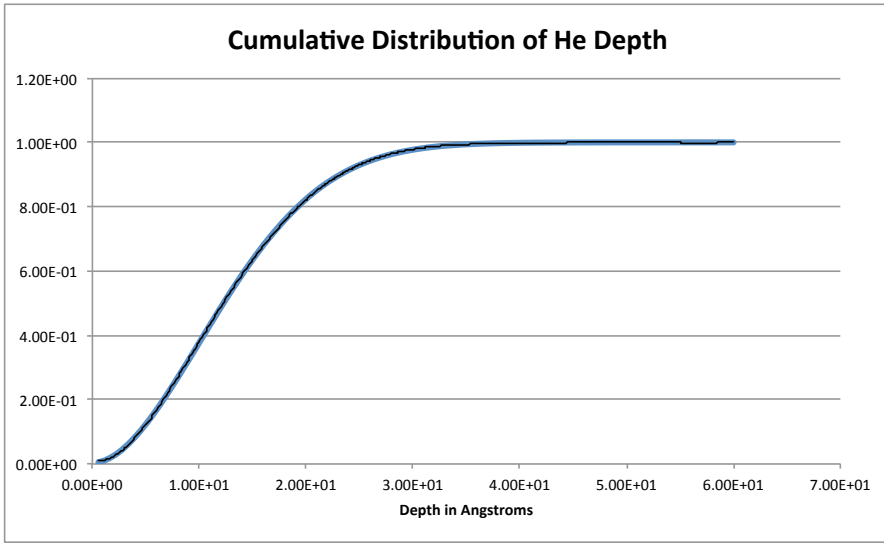
How do these processes influence surface topology evolution, sputtering, etc. & can sub-sputtering threshold He exposure drive surface evolution processes?



Atomistic investigation of early stage He bubble evolution

- **Tungsten with (100) surface**
- **Periodic conditions in the x, y directions and Free Surface in z**
- **Every 10 ps a He atom is added according to the He depth distribution of 60eV He flux calculated using the SRIM program (Stopping and Range of Ions in Matter)**
- **Temperatures of 500K, 1200K and 2000K**
- **10 simulations for each temperature**
- **→ Quantify He depth and cluster size distributions as a function of time (correlated to the number of added He atoms)**





Atomistic investigation of early stage He bubble evolution

He accumulation at 1200 K,

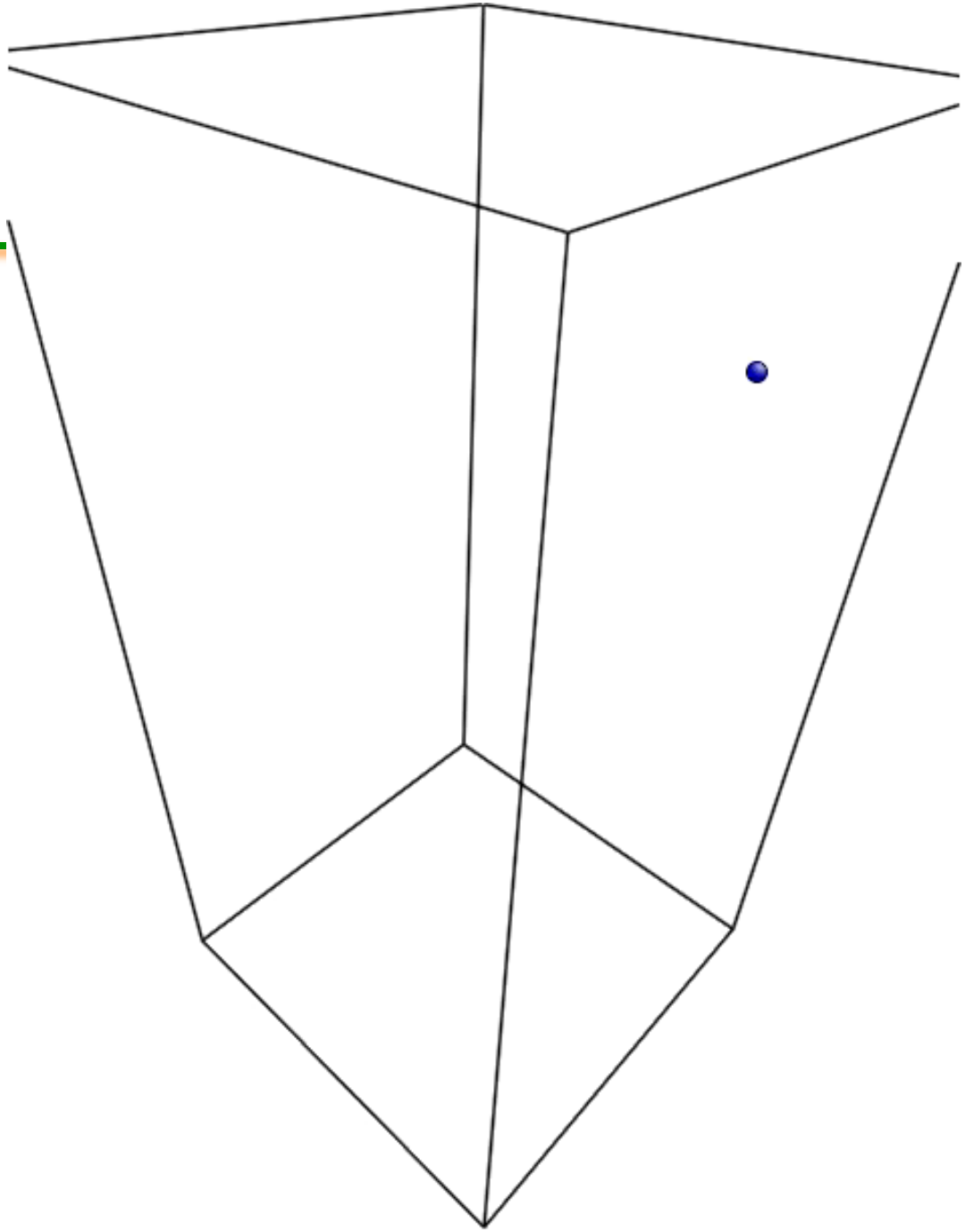
'Thermal' He introduced every 10 ps (very large flux acceleration)

500 He corresponds to ~10^{19} He/m^2

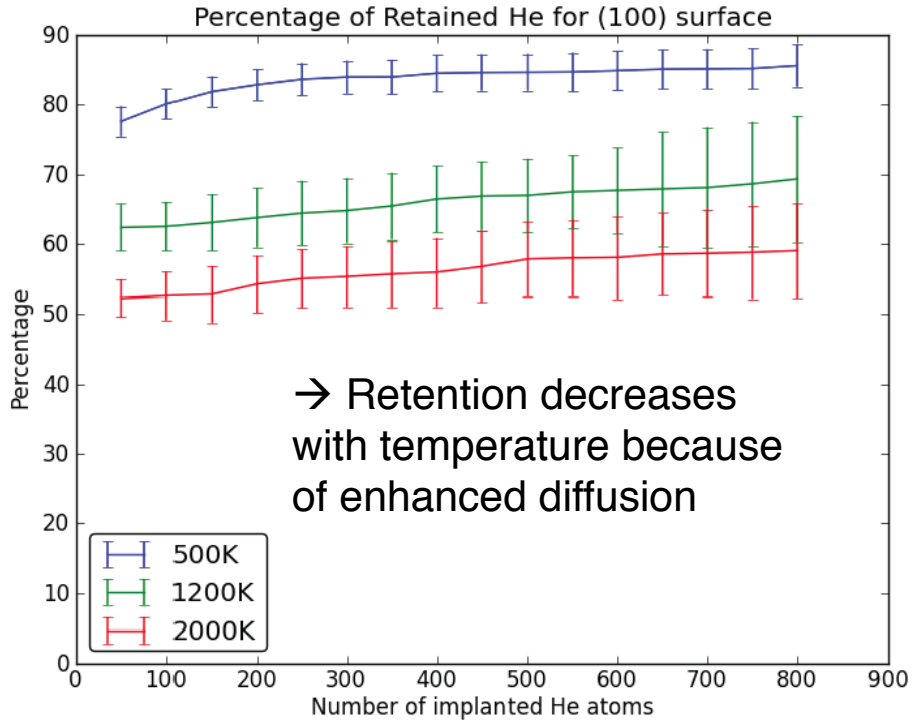
~65% of He retained

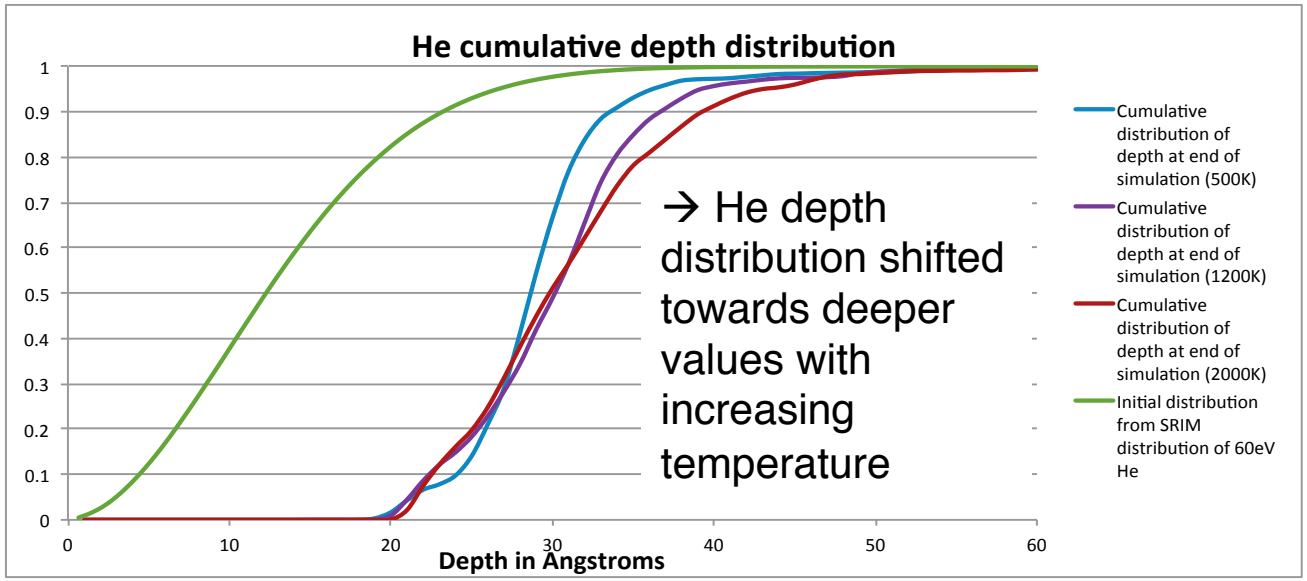
* Initially small He clusters are mobile and grow through cluster coalescence, until reaching size of 5-8, at which trap mutation occurs

* Growth to larger size by absorbing single He and small mobile clusters

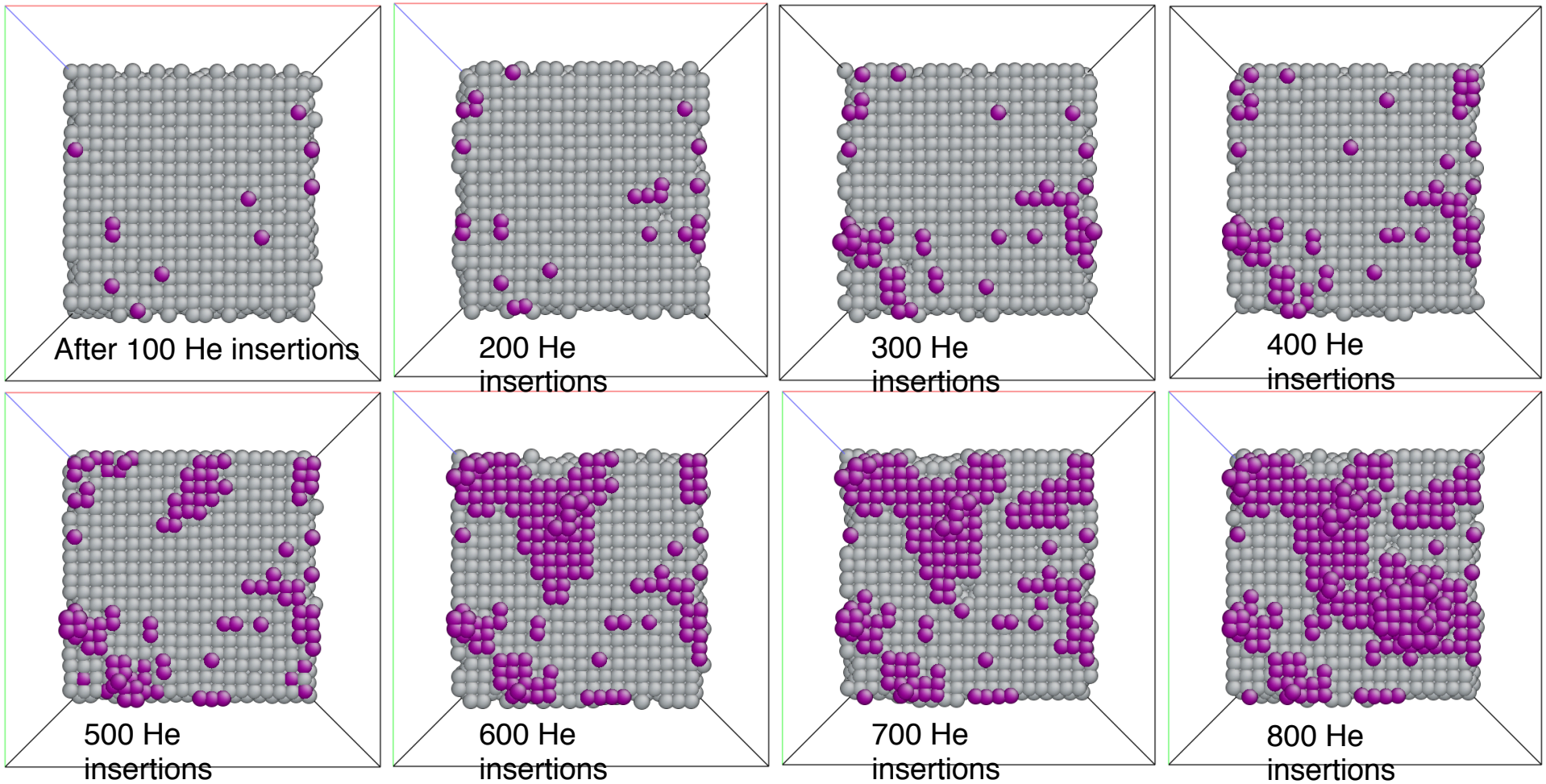


He retention & depth distribution



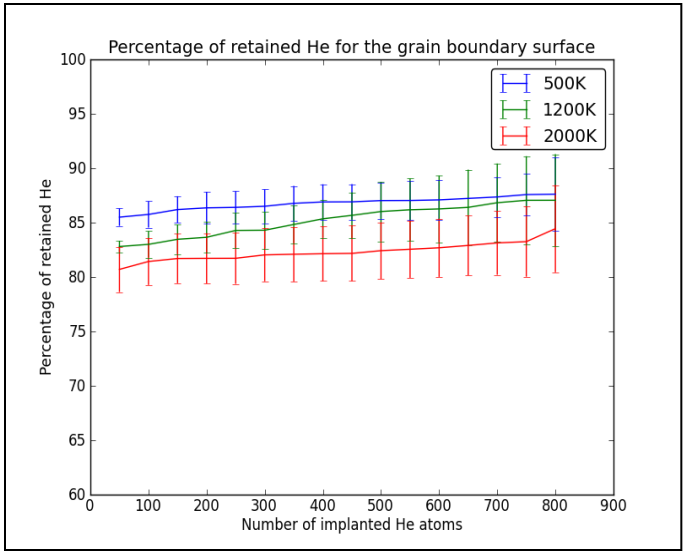


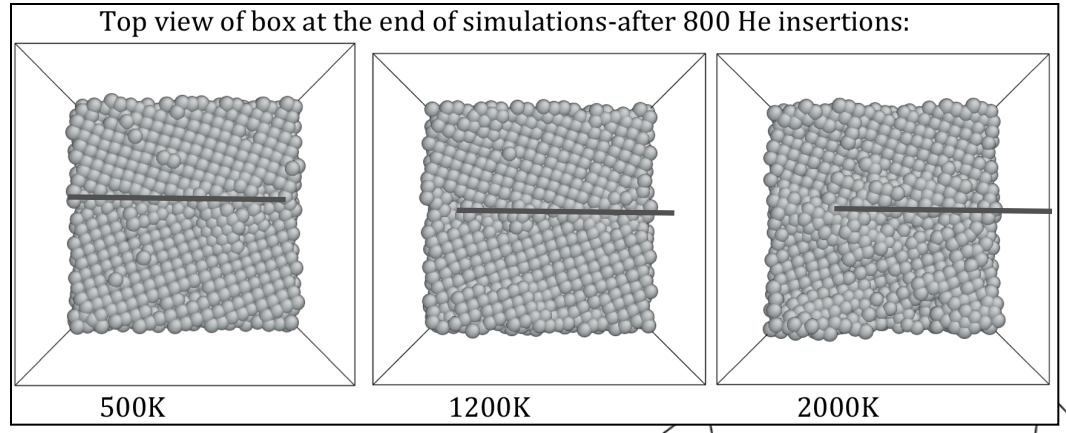
W surface evolution – (100) surface at 500 K

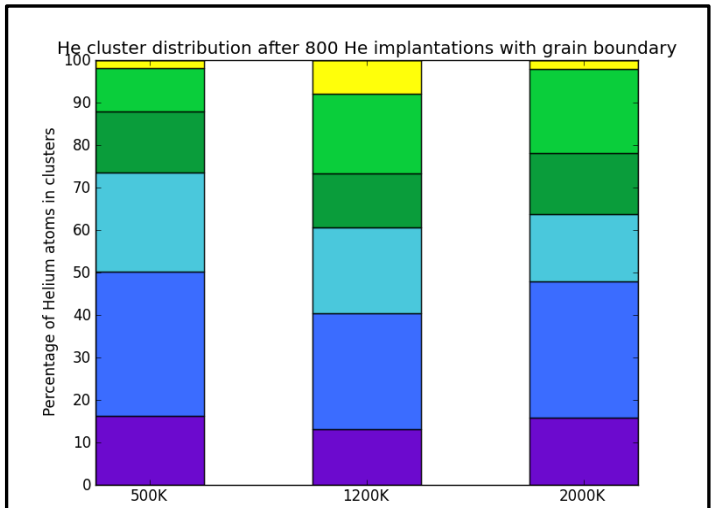


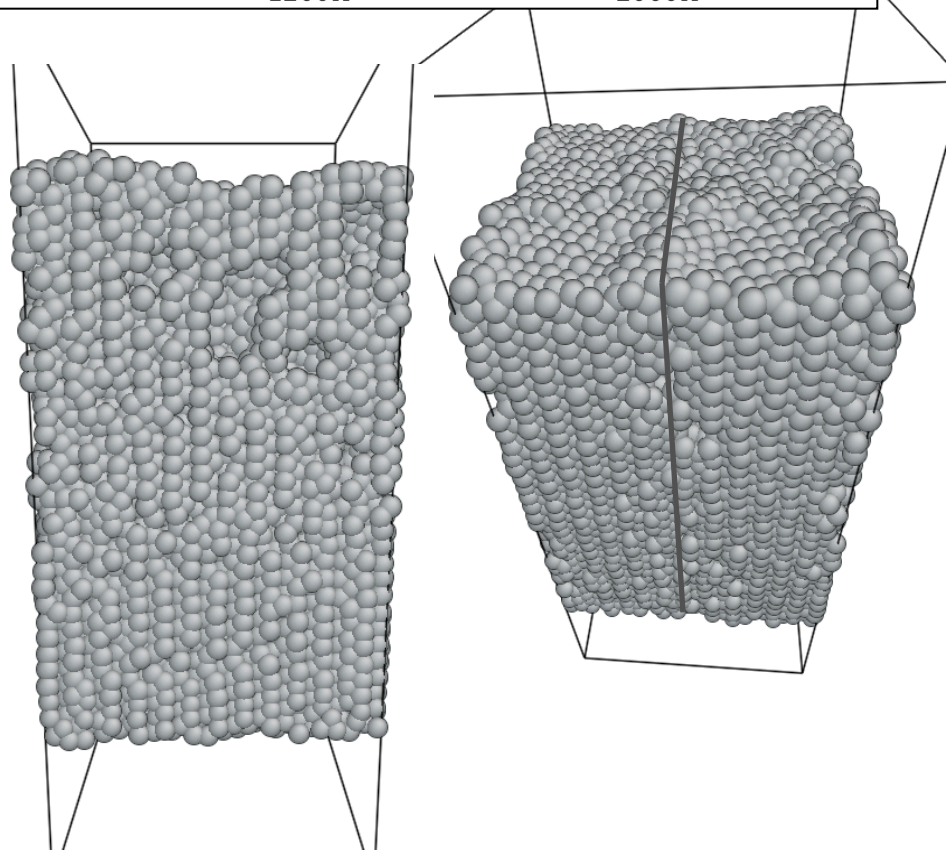
→ Single ad-atom as well as “islands” of ad-atoms accumulation above surface

He Implanted in Tungsten w grain boundary

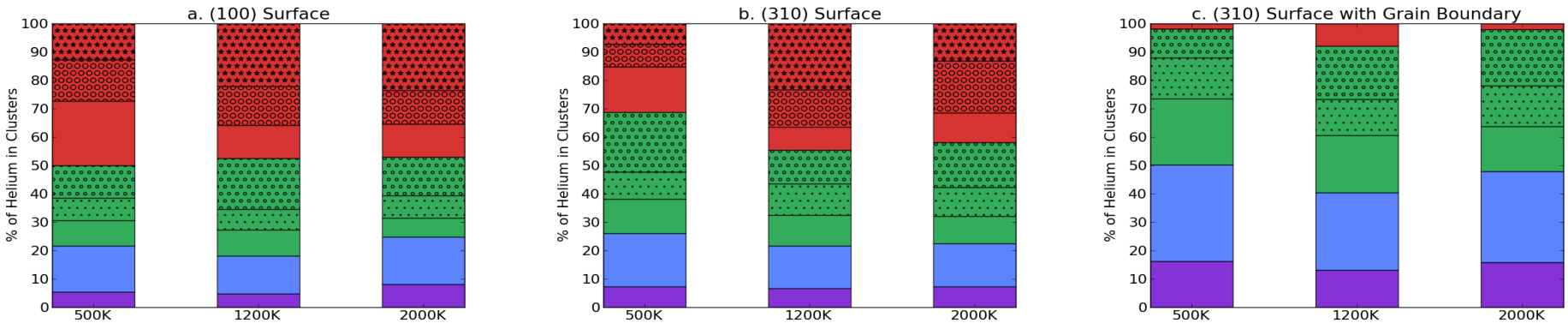








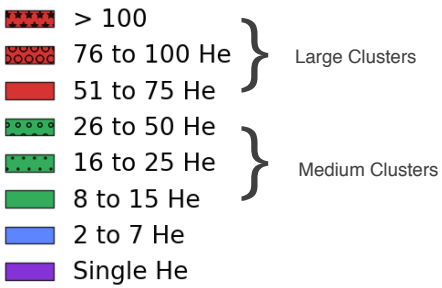
He cluster distribution after 800 He insertions

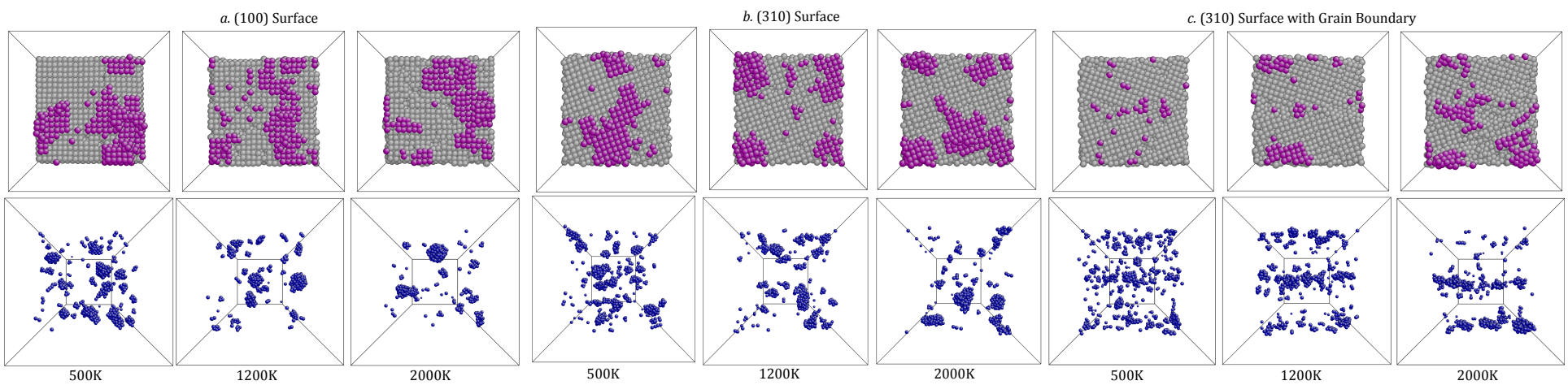


→ Grain boundary plane attracts Helium leading to smaller more numerous clusters than with single crystals

→ Ad-atom islands accumulate, particularly in single crystal cases

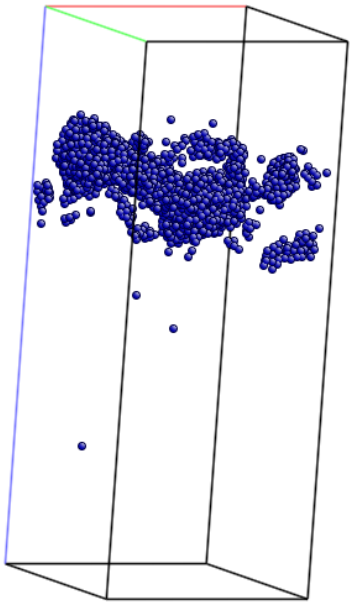
→ Temperature dependence not clear at these timescales

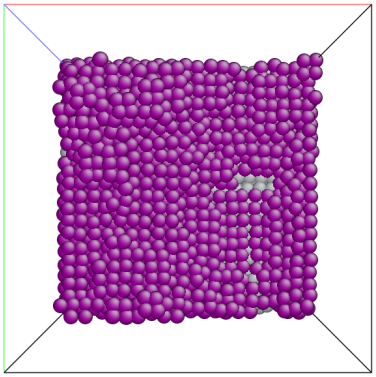


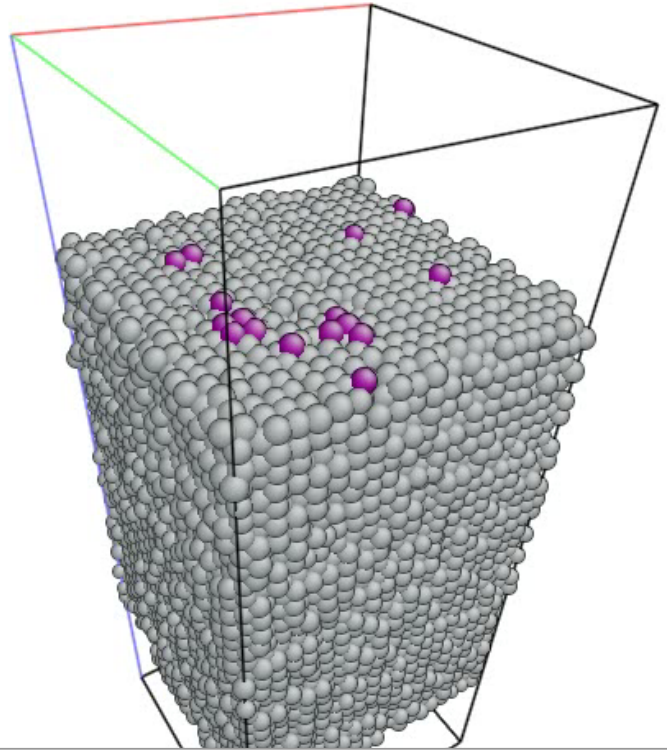


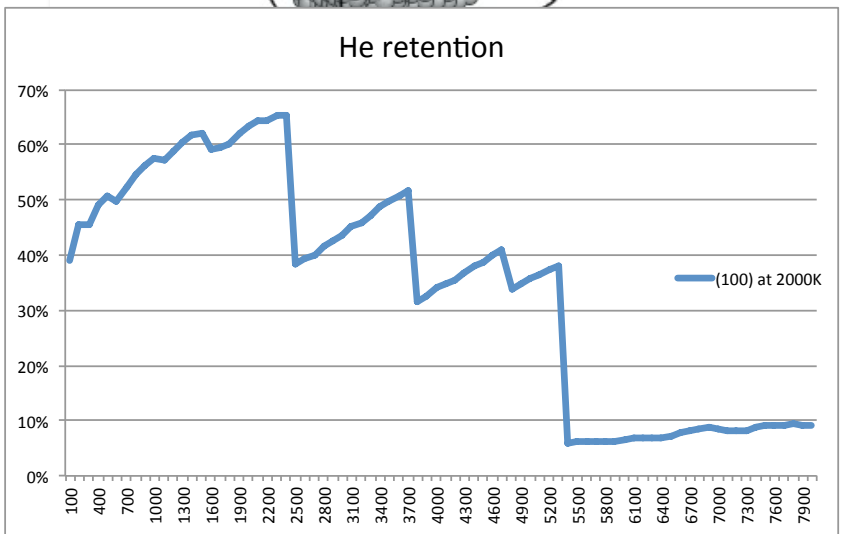
Later stages of bubble growth: 1.(100) Surface at 2000 K

Before bubble burst
(after 5300 He insertions)
→ 38% of inserted He is retained

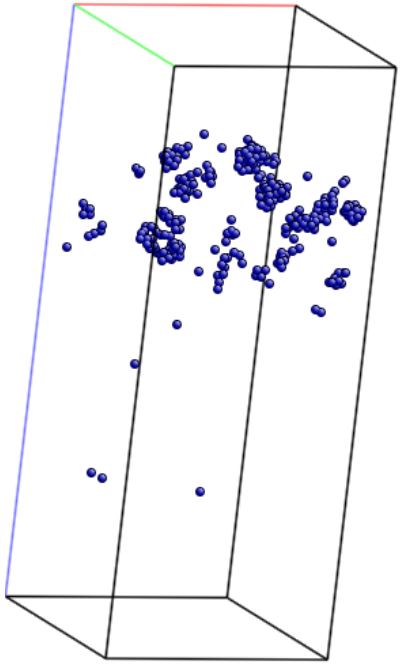


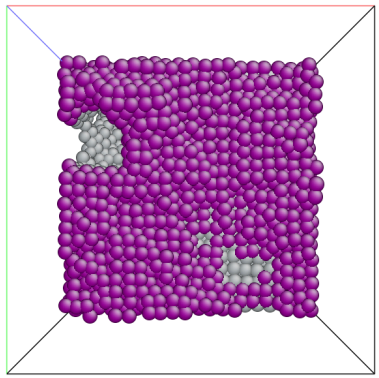




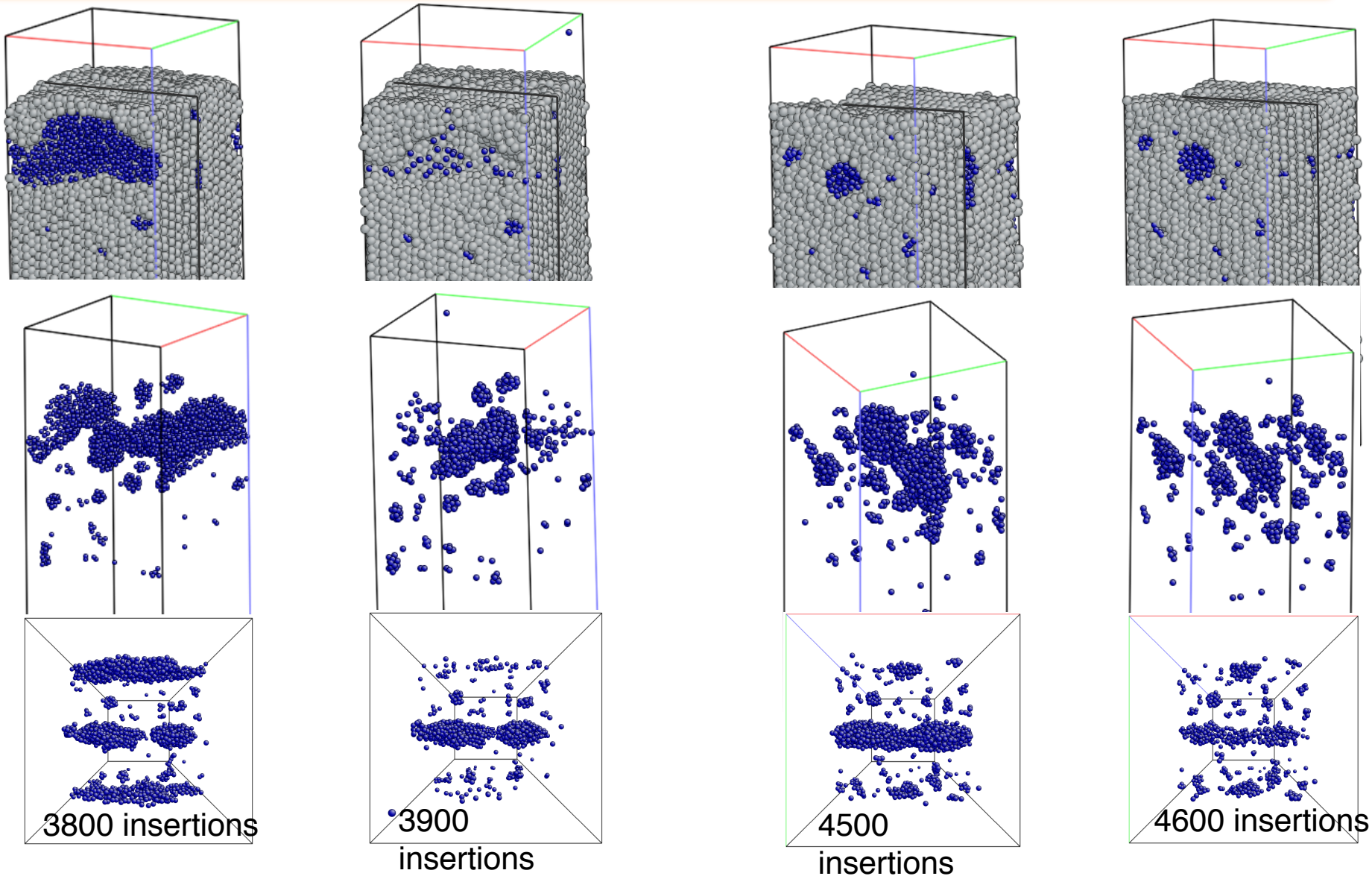


After bubble burst
(after 5400 He insertions)
→ Only 6% He retained





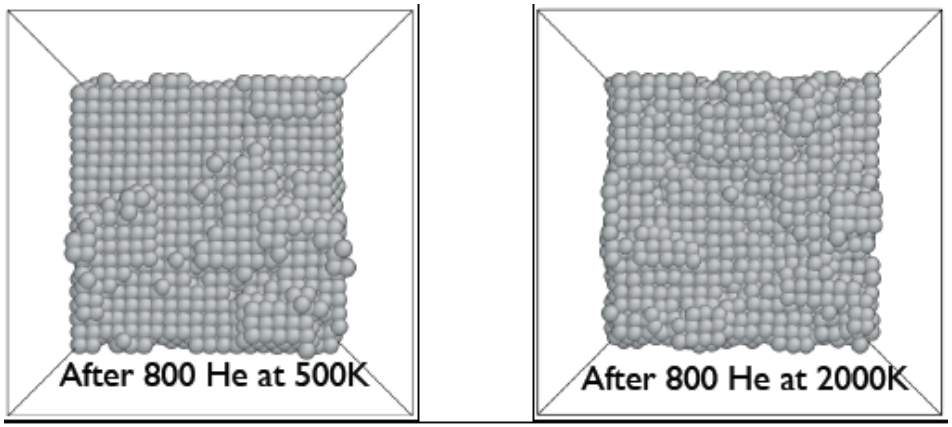
Later stages : 2. Grain Boundary Surface at 2000 K



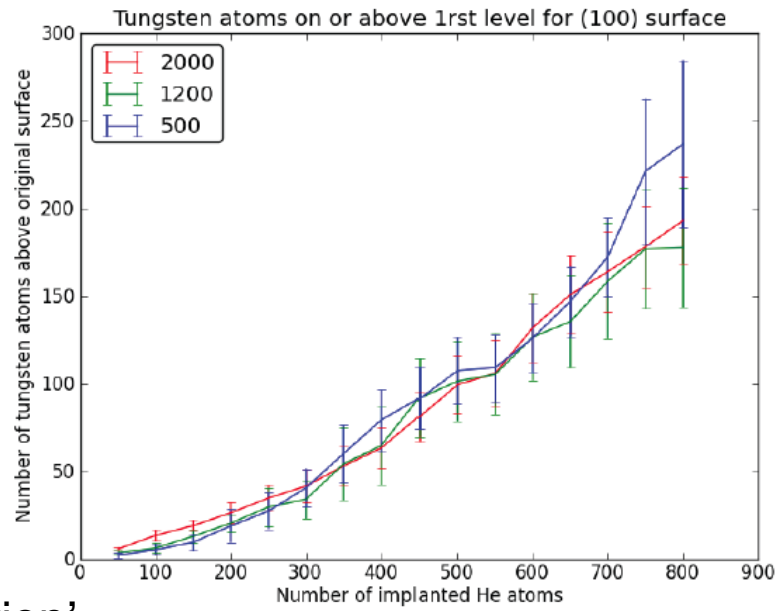
Side grain boundary bubble bursts and empties out

Middle grain boundary bubble bursts and empties out

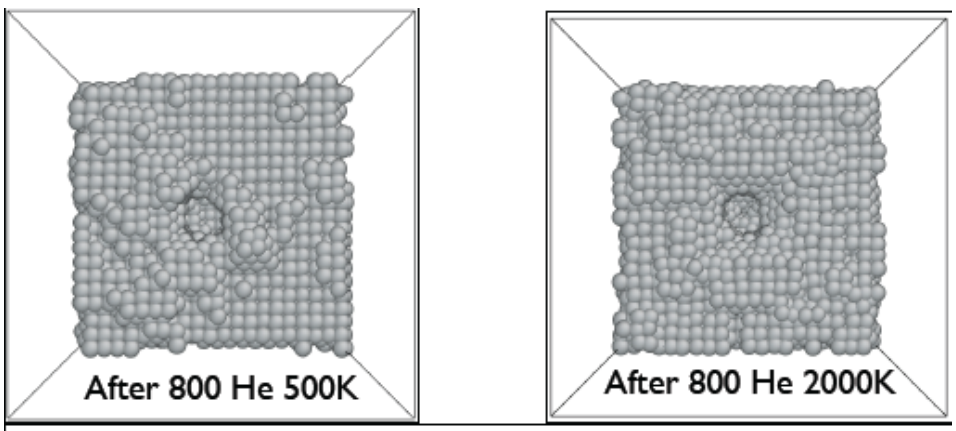
W adatom formation & surface roughness



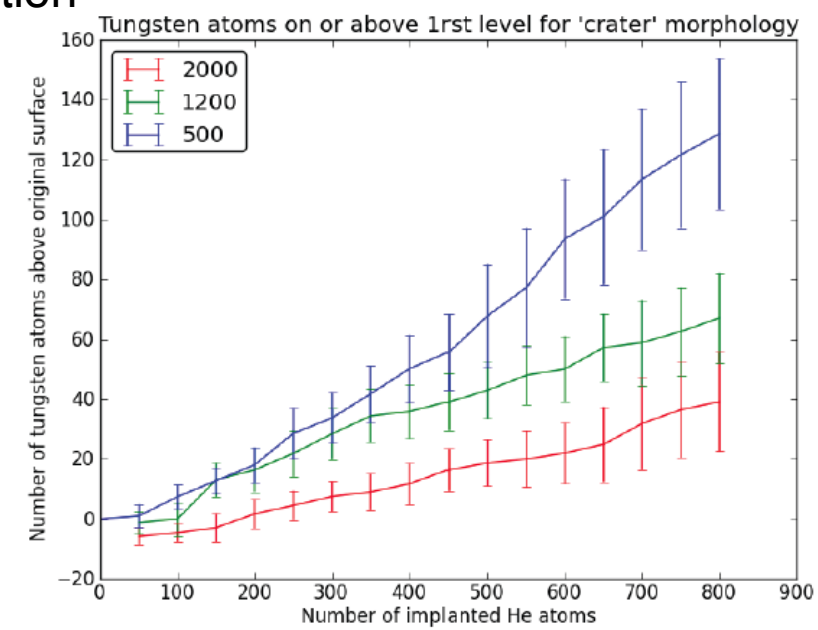
Number of W atoms on or above the initial level is about the same for all three temperatures
The higher T, the less ordered the surface



Mechanism of W adatom formation, He 'trap mutation'

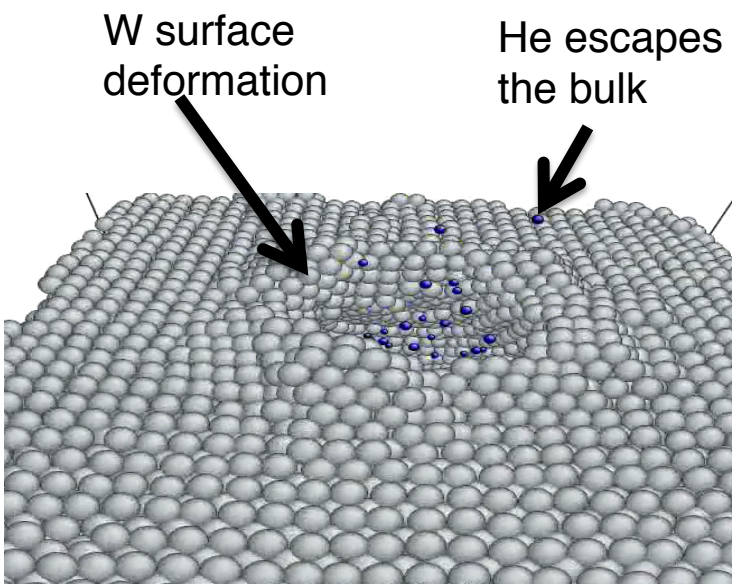


Overall: a lot less pilling up then with the (100) surface
Temperature dependence: at lower T, there's more total W pilling up at surface



Pressure evolution of He bubbles: from equilibrium to burst

- **Objective: Using MD, characterize the He density threshold at which a preexisting bubble pops the surface**

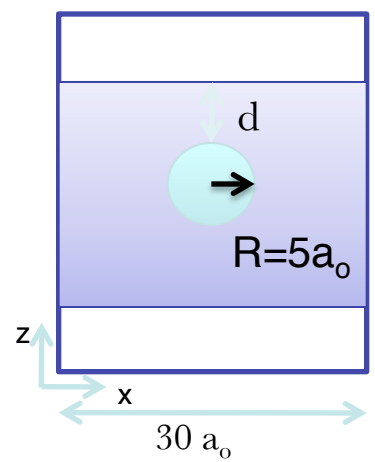


- Bubble bursts
- He escapes from the bulk
- The W "crater" stays in place
  Permanent damage (MD timescales)
- No W erosion

Parameters:

Temperature (K)
500
1200
2000

Distance of Bubble to the surface
d=2 a_o
d=3 a_o
d=5 a_o=R
d=10 a_o=2R



Pressure evolution of He bubbles

Molecular dynamics simulations to assess He bubble pressure & response of over-pressurized, sub-surface bubbles (R ≈ 1.6 nm)

- Equilibrium bubble and loop punching

Ackland-Thetford W-W, Henriksson W-He,
Beck He-He
Future studies with other potentials

Equation of state *(Wolfer 1988)*

R=5 a_0 1044 W vacancies

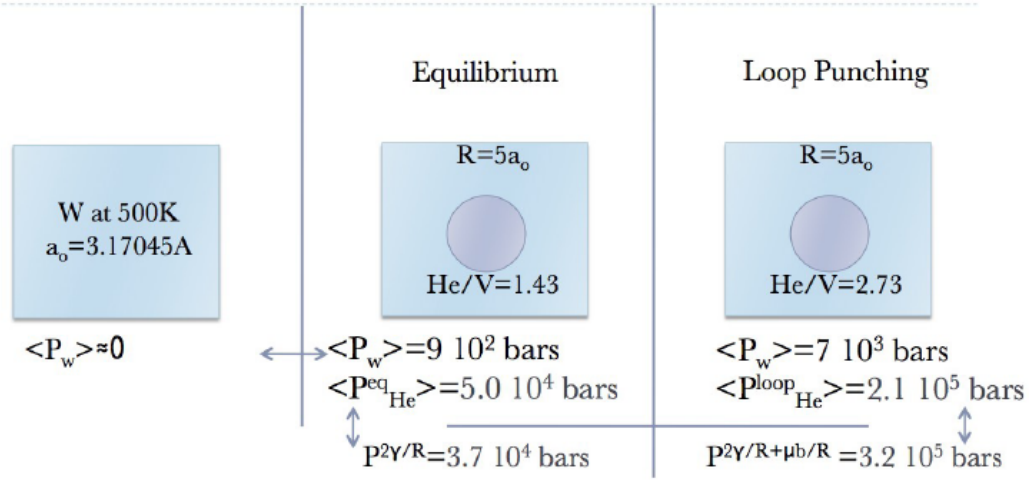
$P_{eq} = \gamma / 2R$

Temperature (K)	500	1200	2000
# He in bubble	1 497	1 234	998
He/V ratio	1.43	1.18	0.969

$P_{loop} = \gamma / 2R + \mu\, b/R$

Temperature (K)	500	1200	2000
# He in bubble	2 847	2 754	2 610
He/V ratio	2.73	2.65	2.53

PBC results at T=500K



- # He is derived from the EOS of He using $P_{eq}=2\gamma/R$ and $P_{loop} = \frac{2\gamma}{R} + \frac{\mu b}{R}$

γ : surface tension
R: radius of bubble
b: burger's vector
μ : shear modulus

- Compute pressure of He in bubble, P_{bubble}, and pressure of W in the bulk P_{bulk}
- Compare P_{bubble} and $P_{eq}=2\gamma/R$ and then with $P_{loop} = \frac{2\gamma}{R} + \frac{\mu b}{R}$
- Compare P_{bulk} and P_w

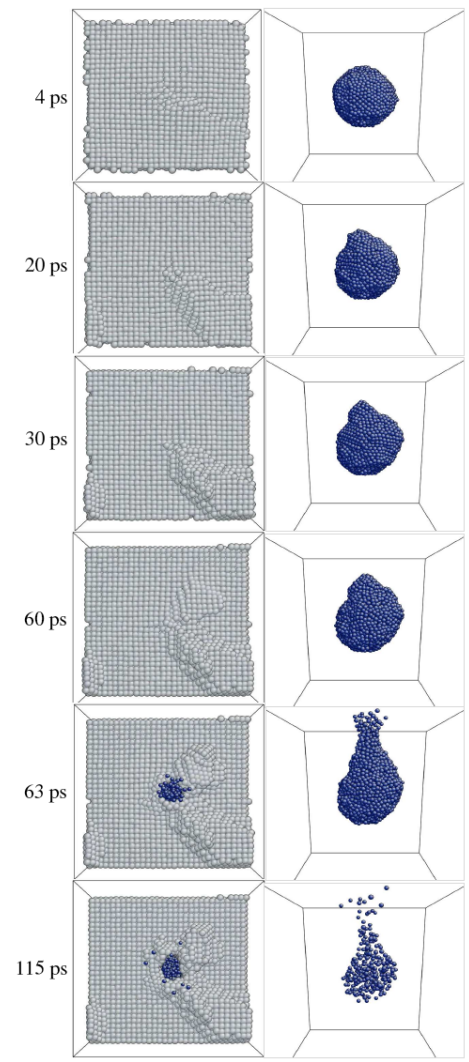
Pressure evolution of He bubbles

- He bubble close to the surface will burst if the pressure is "too high"
  - lead to cratering but no W erosion observed (MD timescales)
- Dependent on distance below surface, size, P, T

Bubble stability as a function of d, depth

He/V	1.43	2.74	3.07	3.85	4.08
$d = a_0$	bursts				
$d = 2\ a_0$	stable	bursts			
$d = 3\ a_0$		stable	bursts		
$d = 5\ a_0$			stable	bursts	
$d = 10\ a_0$				stable	stable

- $d < R \rightarrow$ bubble bursts for He/V < He/V^{loop}
- $d = R = 5\ a_o$:
  Number of He for $P^{2\gamma/R+\mu b/R}$ is 3787 and He/V=3.63
  Number of He for bursting is 4025 and He/V=3.85
  (4025-3727)/3727 = 6% discrepancy in number of heliums between loop punching pressure and bubble bursting
- $d = 2\ R = 10\ a_0 \rightarrow$ bulk behavior (no surface effect)



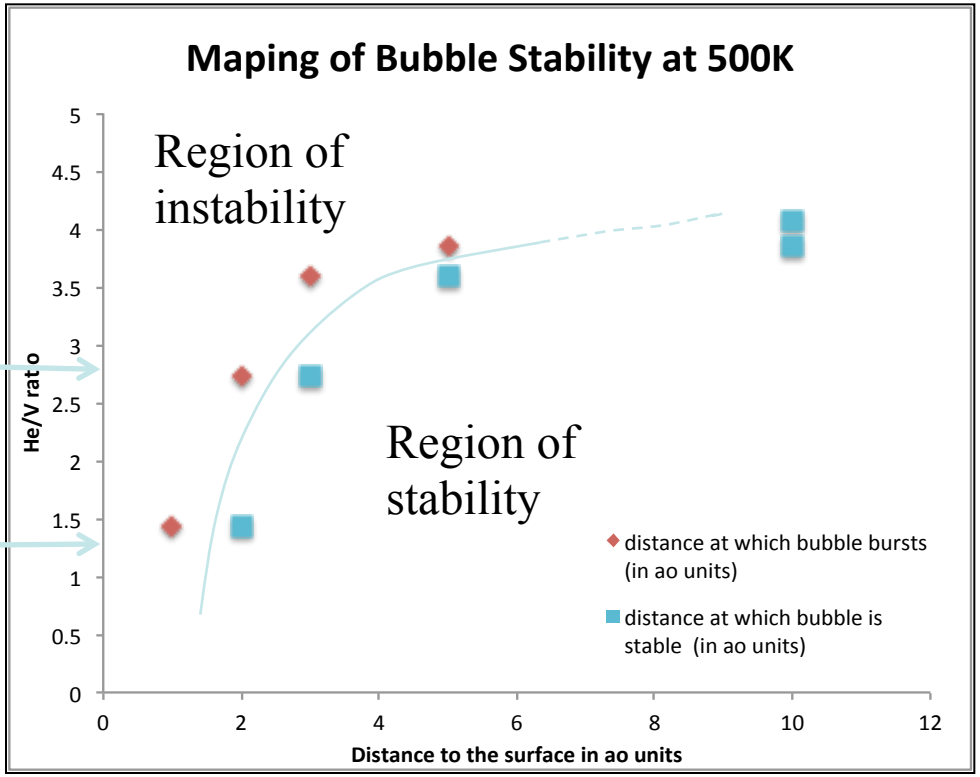
movie

Pressure evolution of He bubbles

T=500K, (100) surface, R=5ao

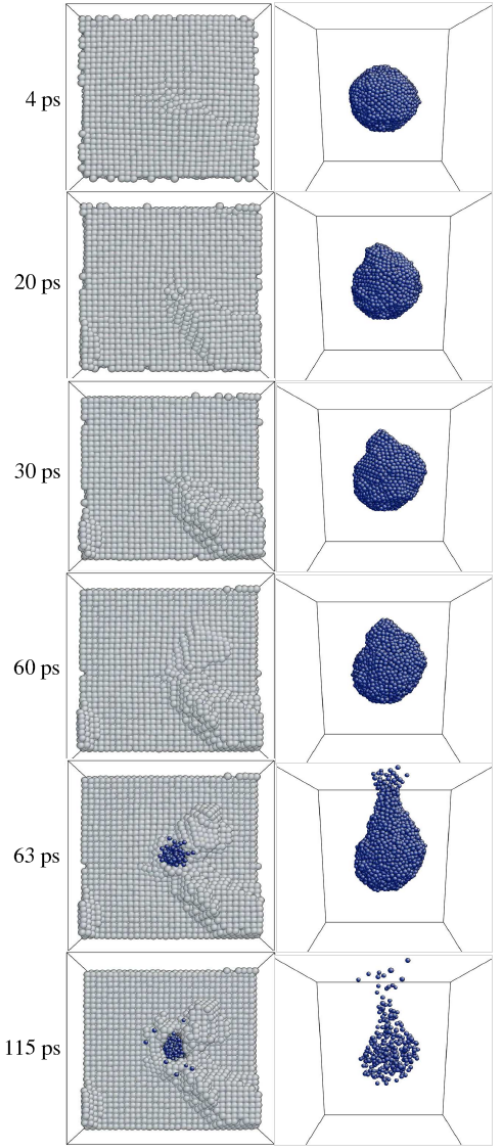
Loop Punching Ratio

Equilibrium Ratio

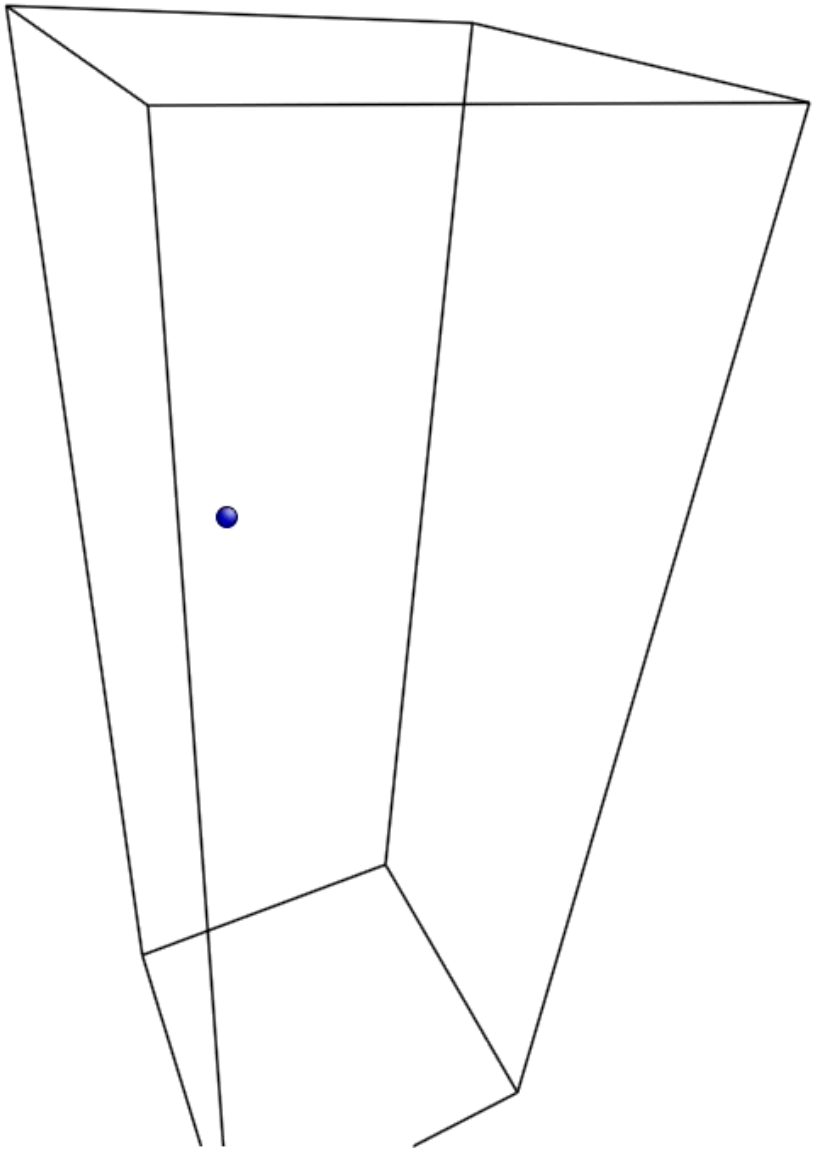


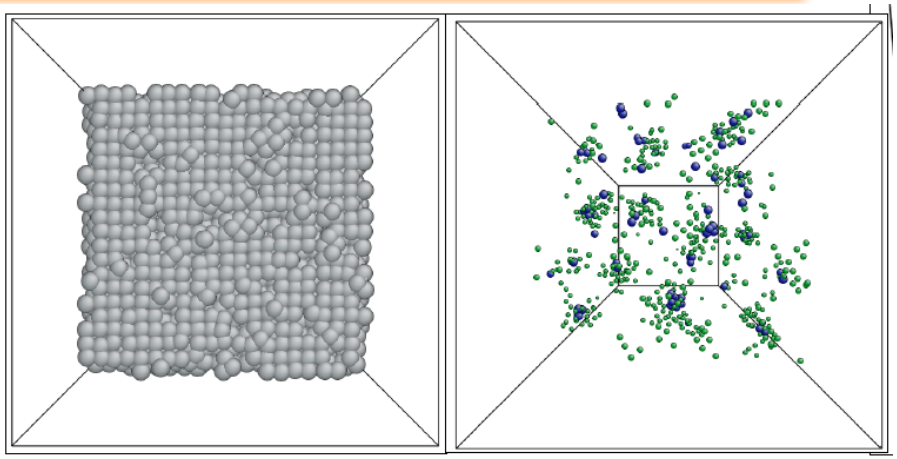
- d<R the bubble bursts for He/V < He/V^{loop}
- d=R=5a_o He/V ratio for bursting corresponds to the loop punching pressure
- d=2R=10a_o Bulk behavior, the bubble is far enough for the surface not to burst

Snapshots of a bubble rupturing with d=5a_o

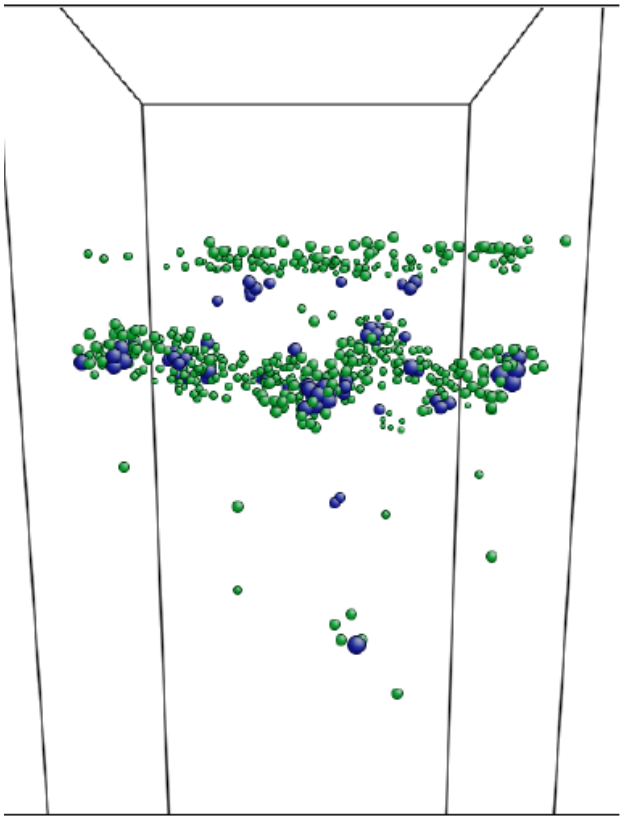


Preliminary simulation with 90%H, 10%He at 1200K

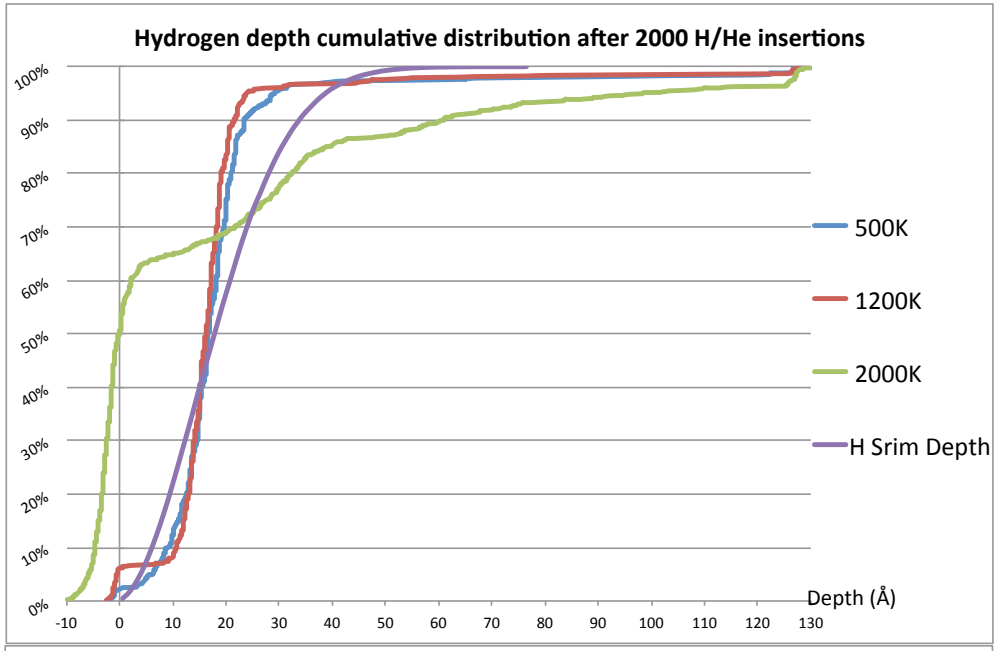


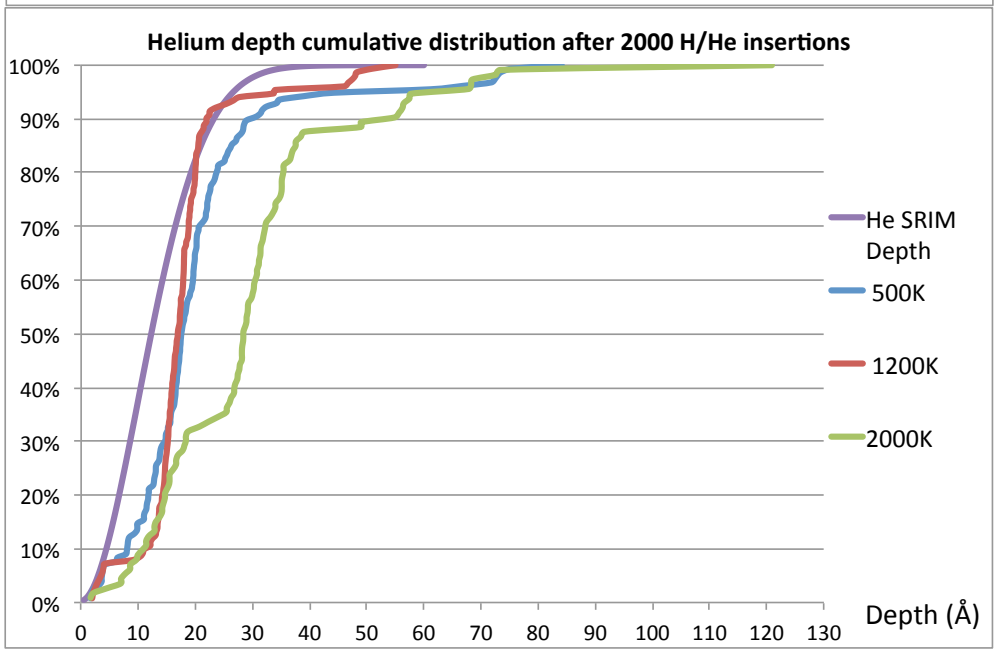


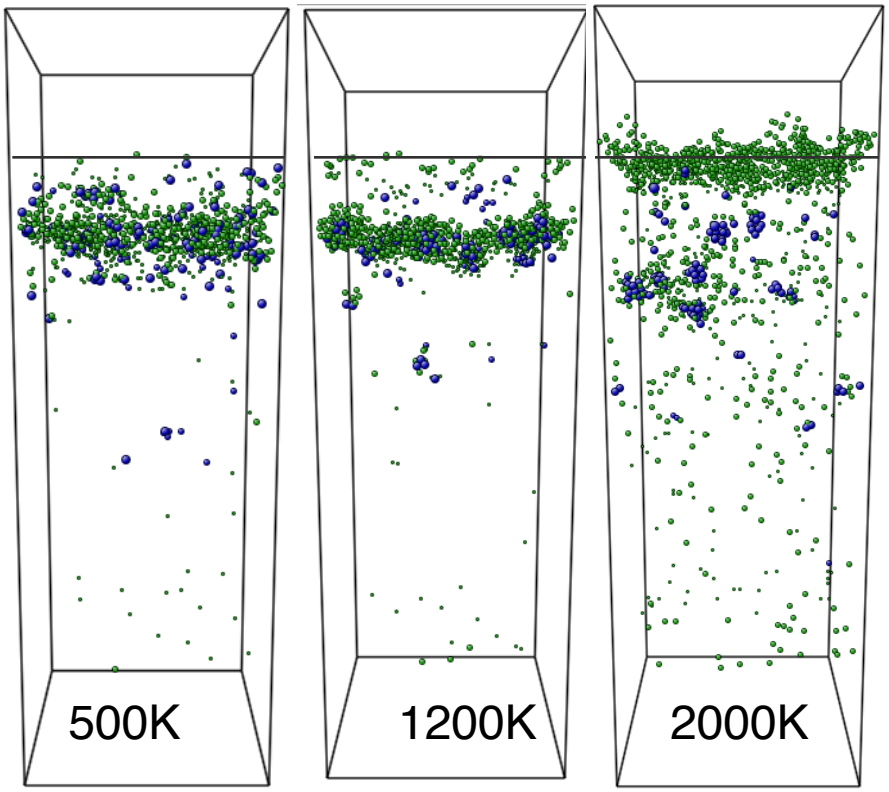
- Significant He/H clustering
- High level of H retention (~68%)
- Increased He retention (~90% versus 68% w/o H)



90%H-10%He Depth distribution after 2000 insertions





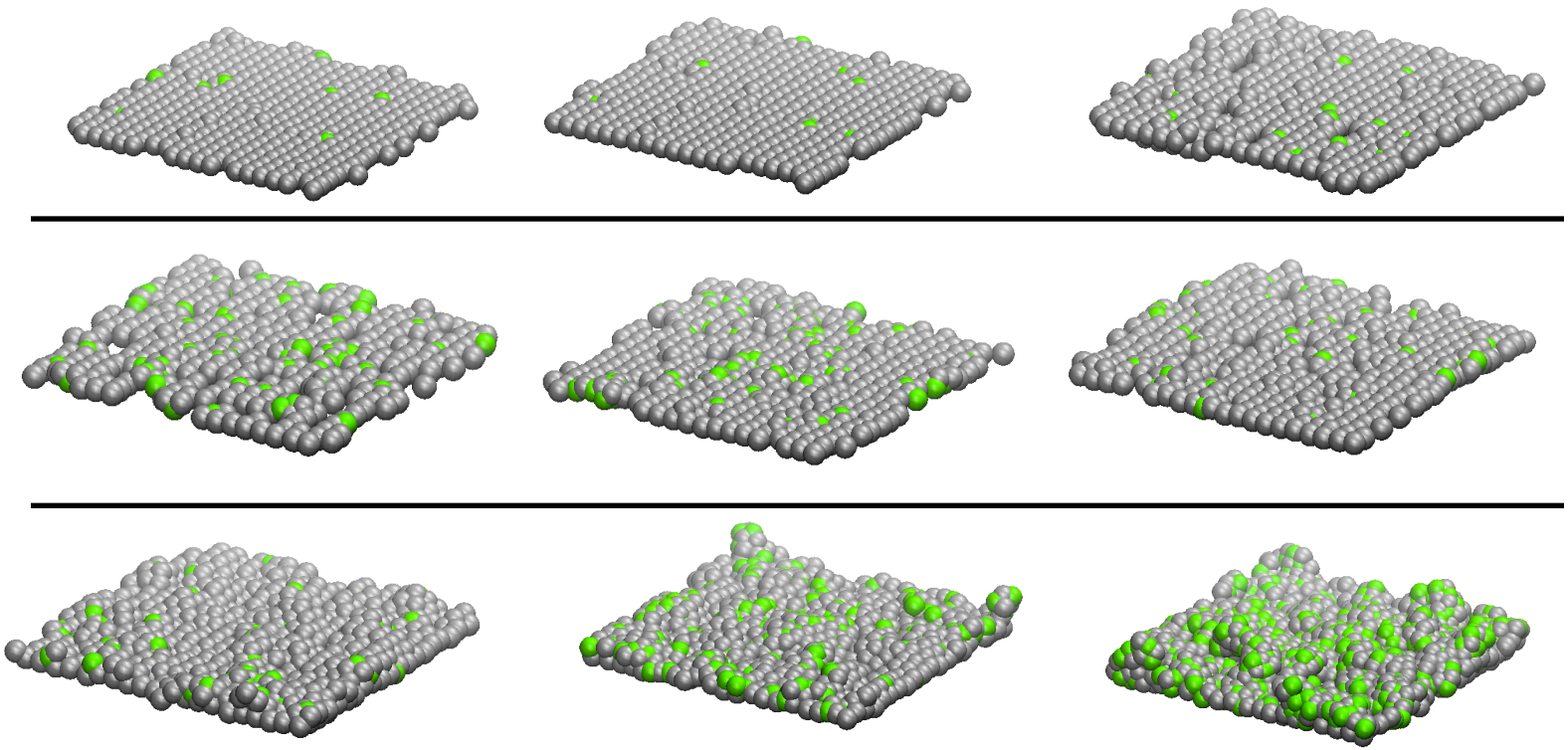


→ Hydrogen saturation layer with temperature dependent depth

→ He clustering, H decorating clusters

Temperature dependence of surface evolution

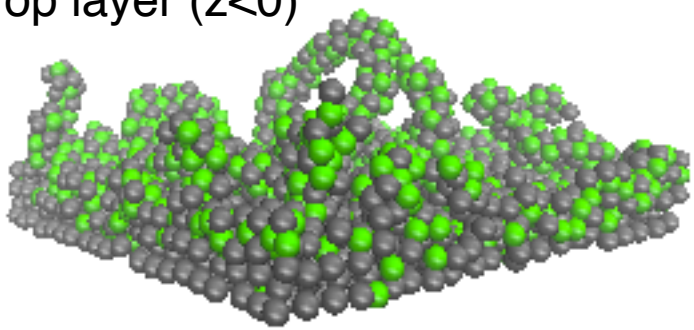
Top layer (atoms above z=0) W in grey and H in green

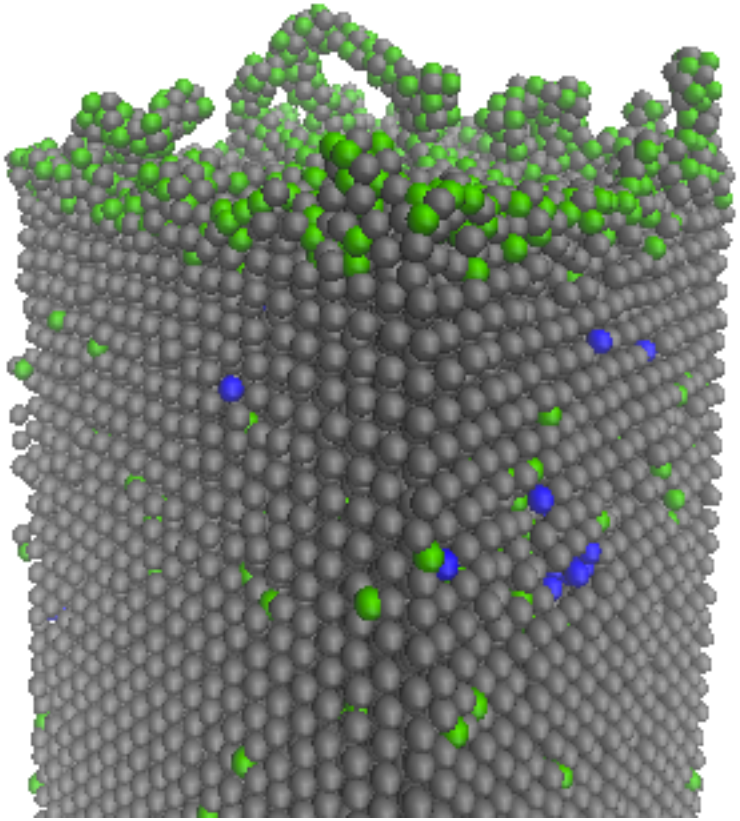


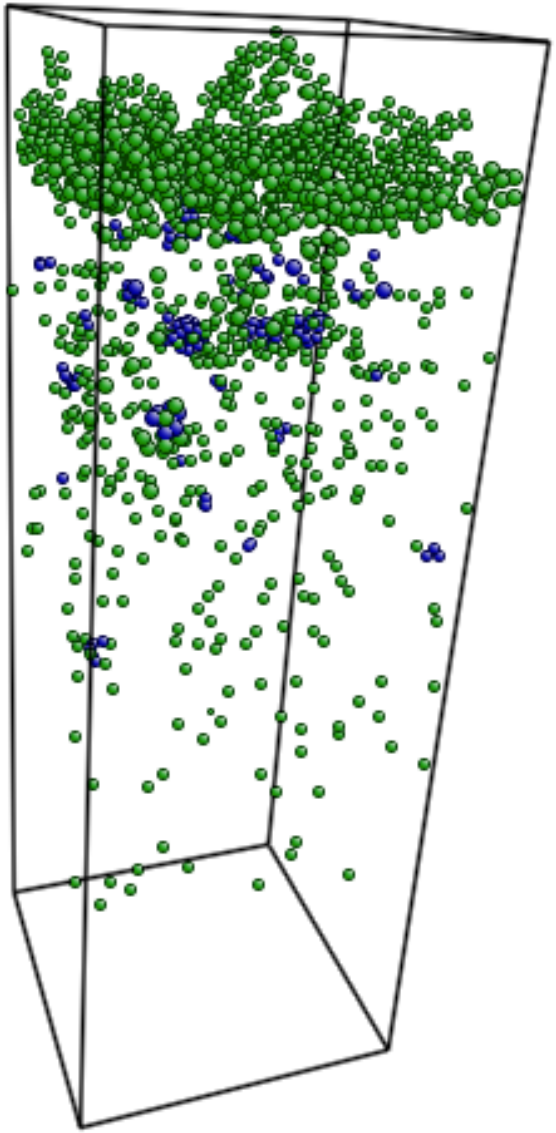
→ Mixed (90% H/10% He) plasma exposure conditions at 2000 K, showing rapid surface topology changes yet no ad-atom "islands"

2000K Surface at 3000 insertions of 90%H/10%He

Top layer (z<0)







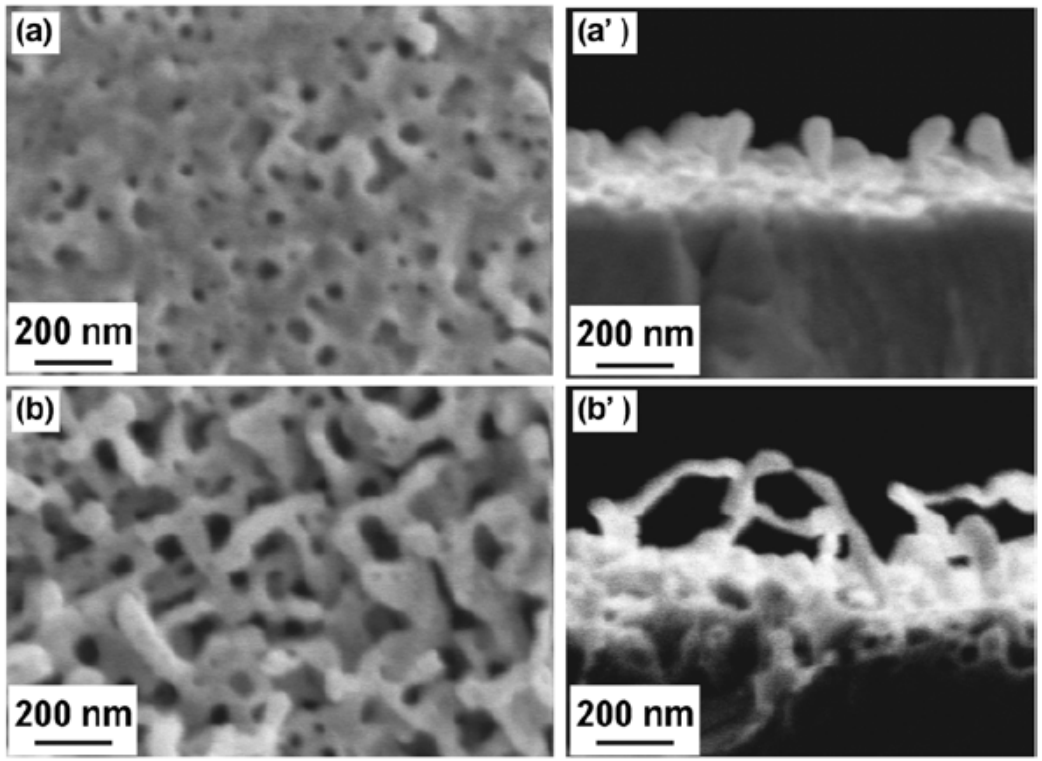
W fuzz formation mechanism(s)

• **Formation mechanism remains unresolved – developing kinetic models 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**

- Key uncertainties: He diffusion through defected surface regions, bubble nucleation versus He absorption at over-pressurized bubbles, influence of temperature/stress gradients

- What happens to displaced W atoms – induce W surface instability

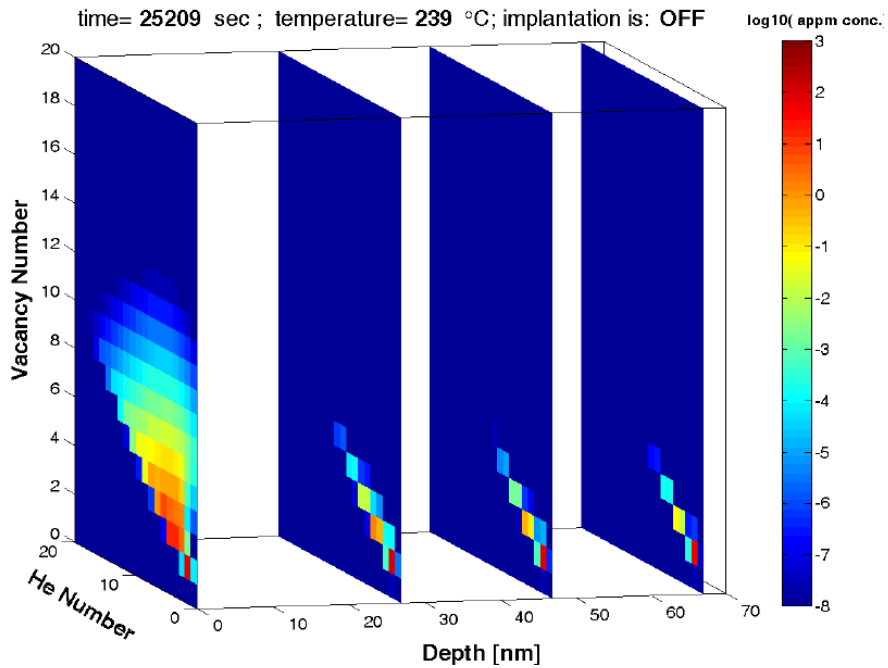
• **MD simulations do not indicate any effect of sub-surface He bubbles on W sputtering yields do to He ion irradiation**

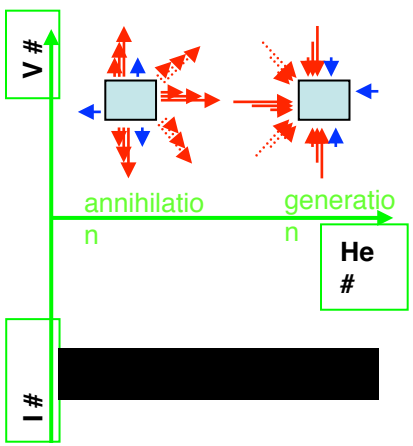


* Kajita, *Nuclear Fusion* **49** (2009) 095005.

Spatially-dependent cluster dynamics model

- *Dimensionality*
  1 spatial dim.: x, non-uniform grids
  1 temporal dim.: t, non-uniform grids
  1.5 phase-space dims: He#, V(I)#
- *What kind of transitions?*
  Any cluster can annihilate (transform to another) or be created (transformed from another) :
  - Capturing: all directions, all step sizes possible, depending on existing mobile species; ***including bubble coalescence***
  - Dissociating: single He, V, I, only





Calculations can involve > 10^7 coupled reaction – diffusion differential equations – utilize parallel solvers (PARDISO)

$$\frac{\partial[He_i]}{\partial t} = D\frac{\partial^2[He_i]}{\partial x^2} + \text{dissoc._rate}(He_m V_n) + \text{He_kickout_rate}(HeV) + \text{implan._rate}$$
$$- \text{self_trap_rate} - He_i\text{_trap_rate}(He_m V_n) - \text{annihilation by dissociation}$$

PARASPACE Model construction

- **How to describe the rates?**
  - capture: C1+C2 → C3;

$$R_{+,1,2} = k_{+,1,2}[C1][C2]; \quad k_{+,1,2} = 4\pi(r_1 + r_2)(D_1 + D_2) \text{ (×Bias, if both interstitial type)}$$

$$r_{(V_n)} = n^{1/3} r_a \qquad r_{(I_n)} = \sqrt{\frac{nV_a}{\pi b}} \qquad D = D_0 \exp(-E_m / k_B T)$$

  - dissociation: C3 → C1+C2;

$$R_- = k_-[C3]; \quad k_- = k_{+,1,2} C_0 \exp(-E_{b,1in3} / k_B T)$$

- **Boundary conditions (BC)**
  black BC, i.e., all concentrations are zero on the surfaces

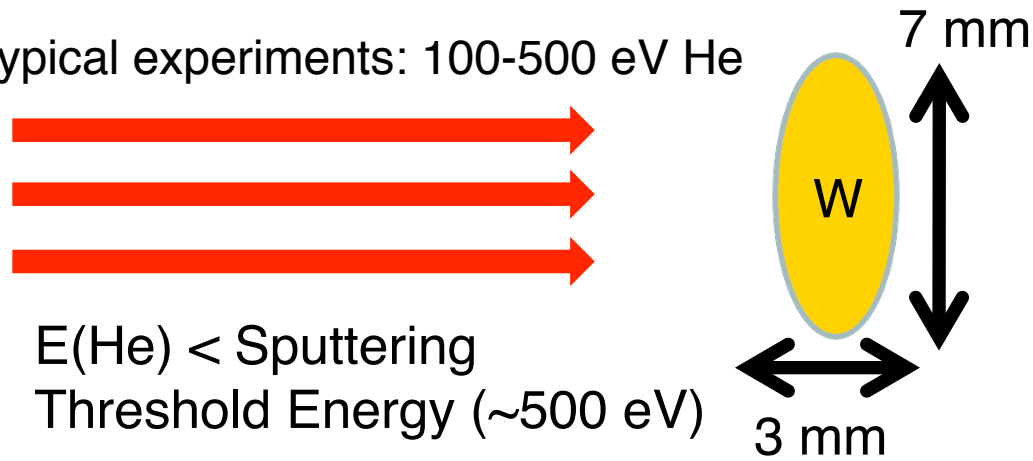
- **Spatial derivative (finite difference)**

$$\frac{\partial^2 C_i^{x_n}}{\partial x^2} = \frac{\frac{(C_i^{x_{n+1}} - C_i^{x_n})}{x_{n+1} - x_n} - \frac{(C_i^{x_n} - C_i^{x_{n-1}})}{x_n - x_{n-1}}}{(x_{n+1} - x_{n-1})/2}$$

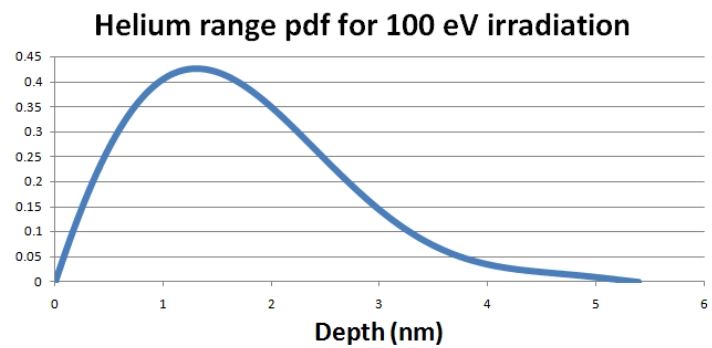
- **Parallel, large sparse-matrix linear solver (PARDISO) using open-MP formalism and backward difference time integration - easily treat systems with 10^7 degrees of freedom**

Low energy He implantation of W

Typical experiments: 100-500 eV He



E(He) < Sputtering Threshold Energy (~500 eV)



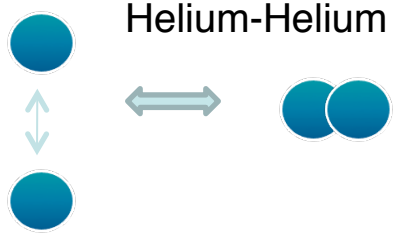
Coupled, 1-dimensional reaction-diffusion model:

$$\frac{\partial C_i}{\partial t}(x,t) = D_i \frac{\partial^2 C_i}{\partial x^2}(x,t) + P_i(x) - Loss\ at\ sinks + Reaction + Dissociation$$

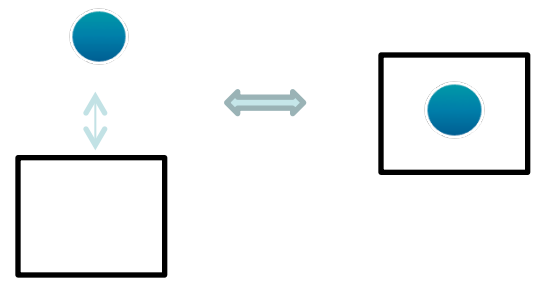
Species considered are Helium, vacancies, interstitials and their clusters, denoted by $He_x V_y I_z$

Low energy He implantation of W

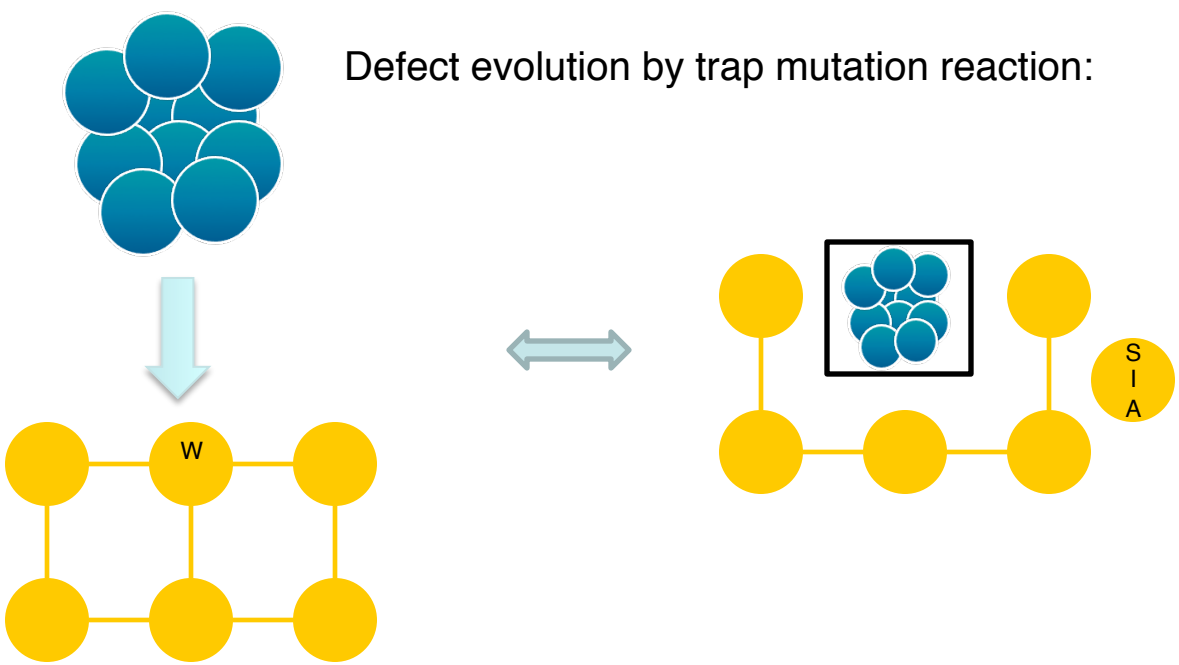
Helium-Helium Interactions



Helium – Vacancy Interactions

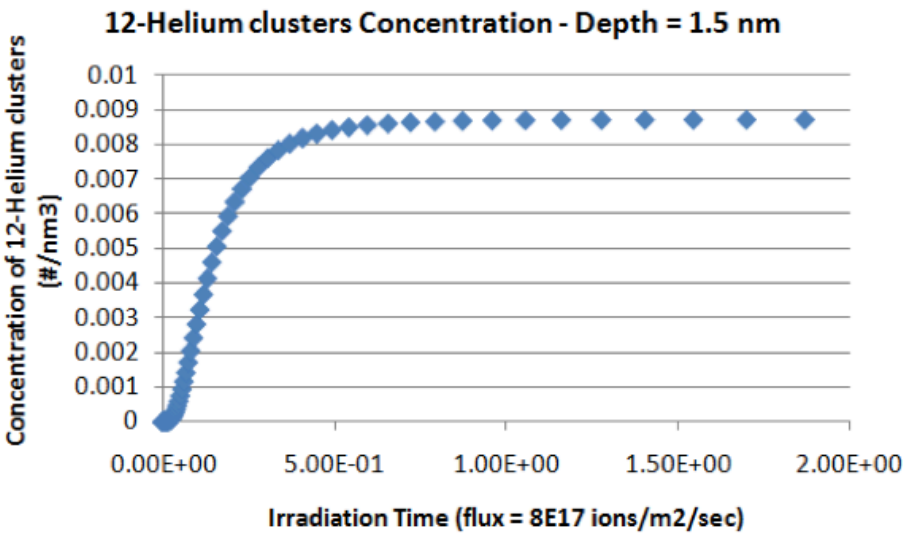


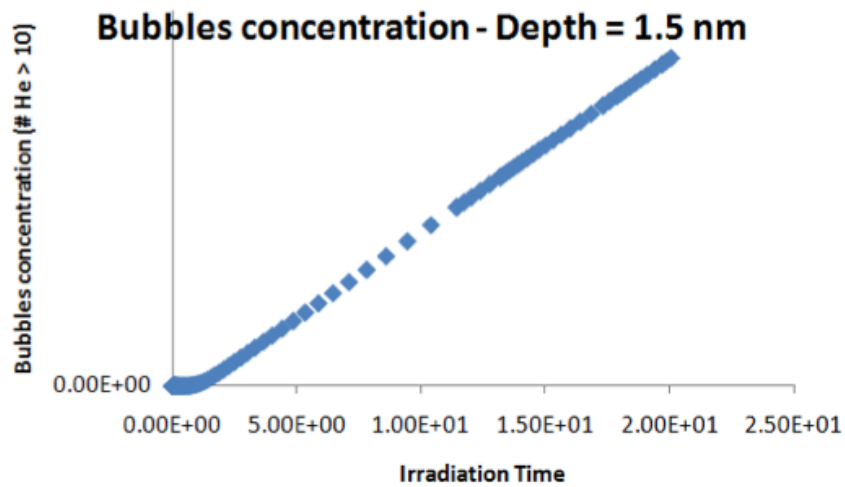
Defect evolution by trap mutation reaction:



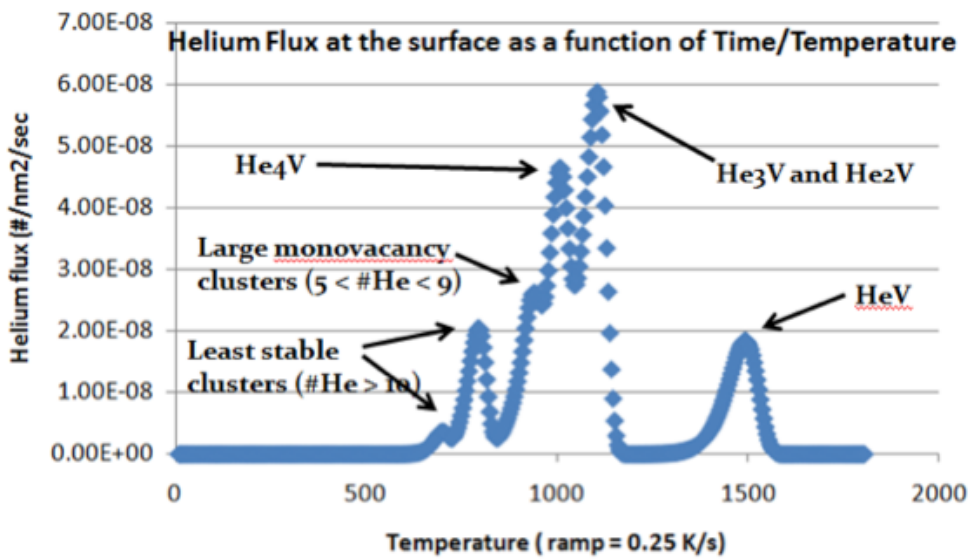
Low energy He implantation of W

Predicted evolution of He clusters below surface during 'irradiation'





Predicted release of He from surface during thermal annealing



Summary of experimentally observed release peaks following 250 eV He into W

Peak	n_{He}	T_{THDS} (K)	Assigned reaction
H	1	1520	$VHe \rightarrow V + He$
G	2	1220	$VHe_2 \rightarrow VHe + He$
F_2	3	1130	$VHe_3 \rightarrow VHe_2 + He$
F_1	4	1080	$VHe_4 \rightarrow VHe_3 + He$
E	5–9	960	$VHe_n \rightarrow VHe_4 + (n-4)He$

A. van Veen, Materials Sci. Forum 15-18, 3 (1987)

Summary & Future Challenges

- **Initial steps towards discovery science to provide mechanistic understanding of W surface dynamics & to integrate with experimental efforts**
  - **Discovery of surface topological changes (ad-atom, loop punching, bursting) & He bubble evolution regimes through MD studies**
  - **Successful initial modeling of longer term, desorption behavior of He following implantation into W**
  - **No influence of sub-surface He bubbles on He-ion sputtering of W (not shown here, Aachen PSI, in press JNM)**

Plasma Surface Interactions (PSI): Bridging from the Surface to the Micron Frontier through Leadership Class Computing

Institution (Lead)	Principal Investigator (Lead)	Additional Personnel
ANL	Tim Tautges (FASTMath)	Emil M. Constantinescu, Jungho Lee, Barry Smith (FASTMath)
GA/DIII-D	Vincent Chan	Adam McLean (LLNL on assignment at DIII-D)
LANL	Xianzhu Tang	Jim Ahrens (SDAV), David Higdon (QUEST), Ollie Lo (SDAV), Danny Perez, Arthur Voter, Blas Uberuaga
ORNL	Brian D Wirth	David E. Bernholdt, Jay Jay Billings, John Canik, Jeremy Meredith (SDAV), Philip C. Roth (SUPER), Roger Stoller, Stanislav Golubov
PNNL	Rick Kurtz	Giridhar Nandipati, Kenny Roche
UCSD	Sergei Krasheninnikov	Roman Smirnov
UIUC	David Ruzic	Davide Curreli, Kyle Lindquist
UMass	Dimitrios Maroudas	

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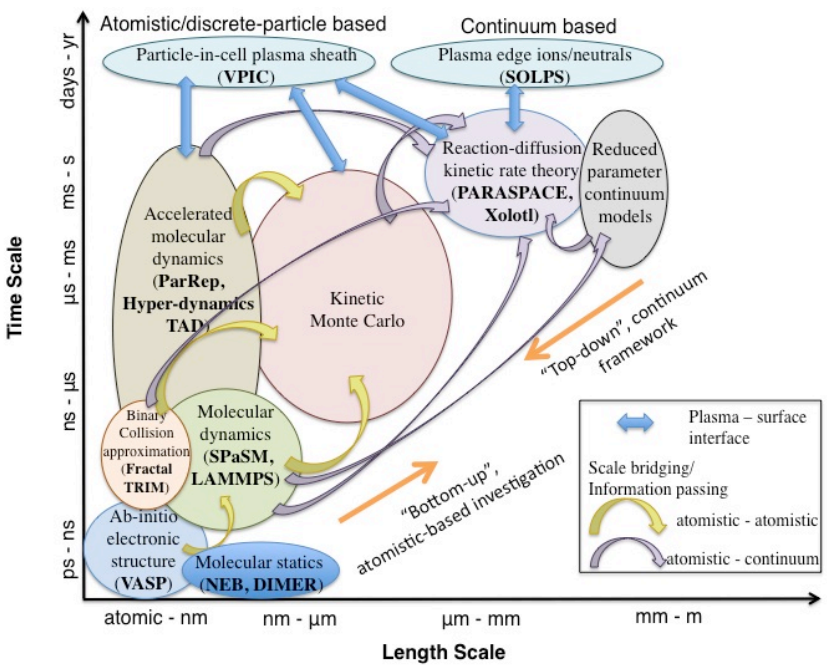
This work is supported by the U.S. Department of Energy, Office of Fusion Energy Sciences and Advanced Scientific Computing Research (ASCR) through the SciDAC-3 program.

Multiscale modeling approach for SciDac

• Integrated team of computer scientists, plasma edge physics and materials scientists to simultaneously address multiscale plasma surface interaction challenges with a 'bottom-up' and 'top-down' approach



Key materials physics questions:

• What physical parameters control near-surface morphology and composition of the re-deposition layer – key phenomena to model include recycling, surface morphology, gas bubble, precipitate and second phase domains (including porosity), and gas fueling/recycling

• What are the effects of high-energy neutron damage on mediating, or exacerbating, near-surface defect evolution and tritium species permeation and retention?

• What is the impact of dilute impurities on surface morphology evolution and plasma contamination and how does mixed material transport in tokamaks impact erosion and impurity generation?

• Feedback effects of evolving structure

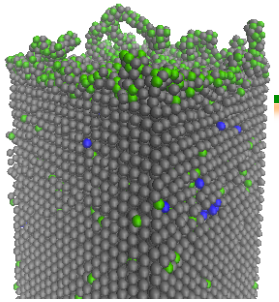
Key computer science questions:

• Scalable algebraic and geometric multigrid methods for reaction-diffusion type problems and more efficient coupled ODE integrators

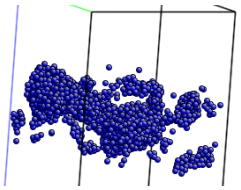
• Forward uncertainty propagation, inverse modeling and parameter estimation, linking information and uncertainties across scales

• Dynamic performance about data collection, abstractions and tools for performance portability, and resilience

• Improvements in visualization/data management from computer simulations



Solid Surface Modeling Roadmap



Low temperature (< ~1000 K) regime of low-energy (~100 eV) He (later mixed He-H) plasma exposure to tungsten, focused on bubble formation, growth & over-pressurization leading to tungsten surface morphology changes

Key Physics Questions:

- Rate effects (AMD, MD and KMC) versus continuum reaction-diffusion & experiment
- Dilute limit approximations in concentrated He bubble populations
- Biased/drift diffusion (elastic strain field interactions that add drift term to diffusional flux)
- Multiscale integration

Dilute limit (?)

(Additional) collective phenomena (mixed species, higher T, etc.) →

Diffusion
drift
aggregation
theory

Visualization/ post-processing

Comparison with data – what observables?

↑↓ Parametric studies

"Integrated bubble evolution modeling" & inter-comparison (bubble dynamics, etc…)

'Individual effects' (e.g., bubble bursting, drift-diffusion)

Continuum models with explicit presence of bubbles that need to be discretized (front-tracking)

Diffuse interface models (e.g., phase field)

Eventual development of model reduction (UQ) to reduce species tracking

Paraspace (initially) - XOLOTL-PSI

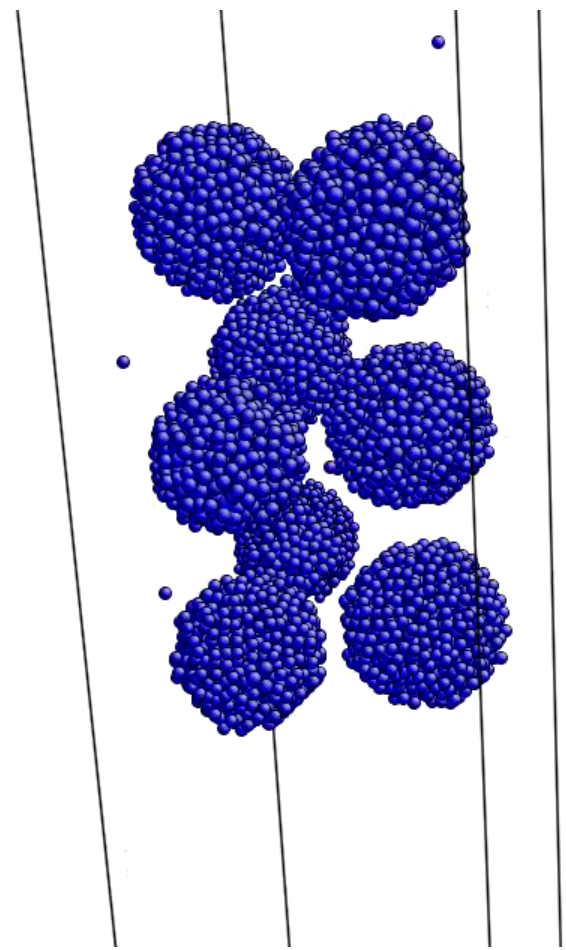
Both input (multiscale linking) & comparisons of predictions

MD/AMD/KMC

Near surface to bulk → matching ←

He bubble influence on sputtering

Sputtering observed in PISCES is generally lower then in ion-based accelerator studies. Hypothesis that bubbles and voids formed in PISCES might be the underlying reason behind this difference.
→ MD simulations to see if this hypothesis is supported by modeling

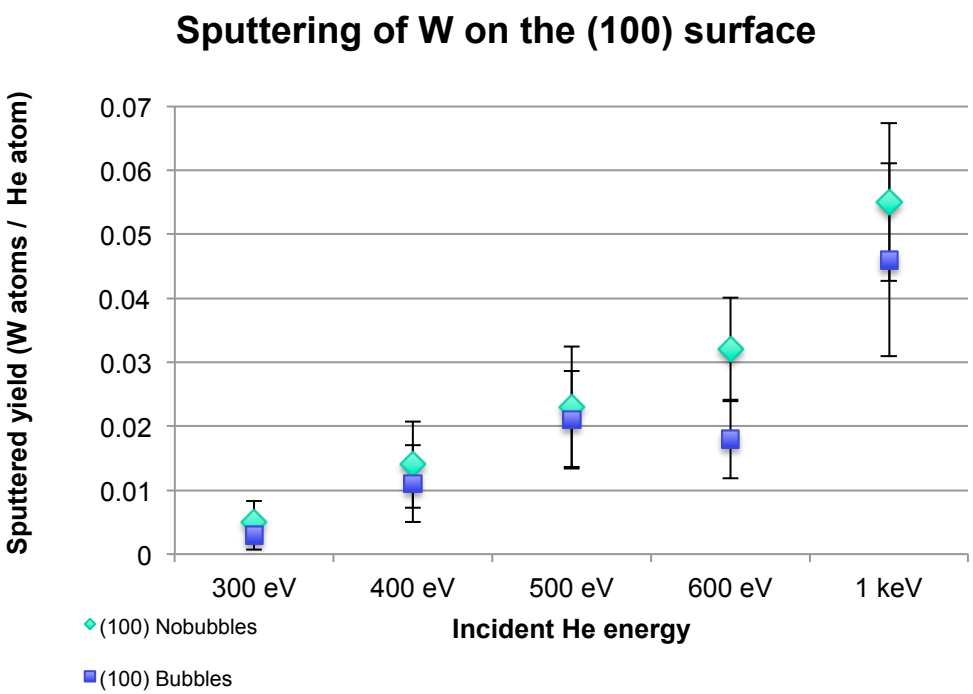


Conditions of the simulation

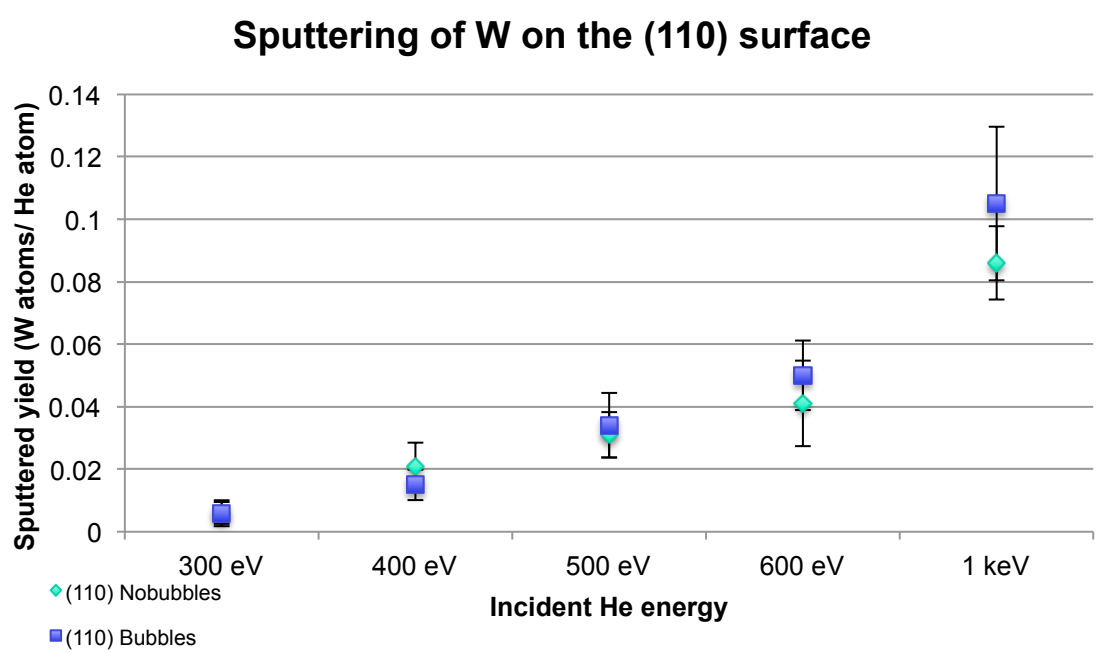
- T=293K
- Tungsten: 2 different W surfaces (100) and (110)
- Bubbles: - fill 15% void fraction in the tungsten
  - placed randomly and R ≈ 1.2 nm
  - equilibrium pressure in the bubbles
  - closest bubbles are 1.5 nm from the surface
- He ions: - 300eV, 400eV, 500eV, 600eV and 1keV
  - flux $\approx 10^{27}$ He/(m^2-s)
  - 30 ps between He atoms
- Runs: averages over 10 runs with 100 incoming He
- Comparison of benchmark, simulations with no bubbles, to simulations with bubbles

< W end – W initial> → number of sputtered W for 100 incoming He atoms

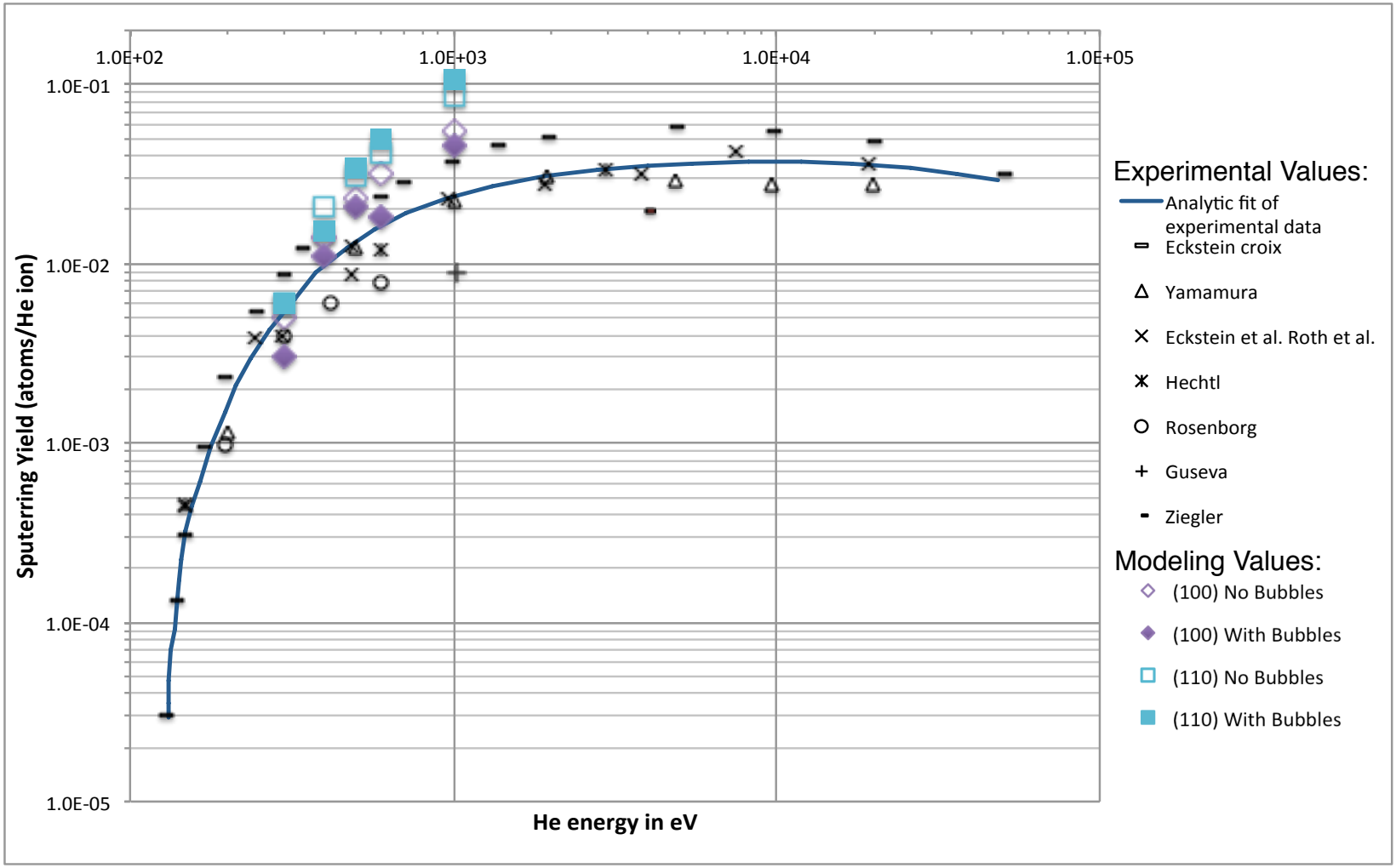
He bubble influence on sputtering



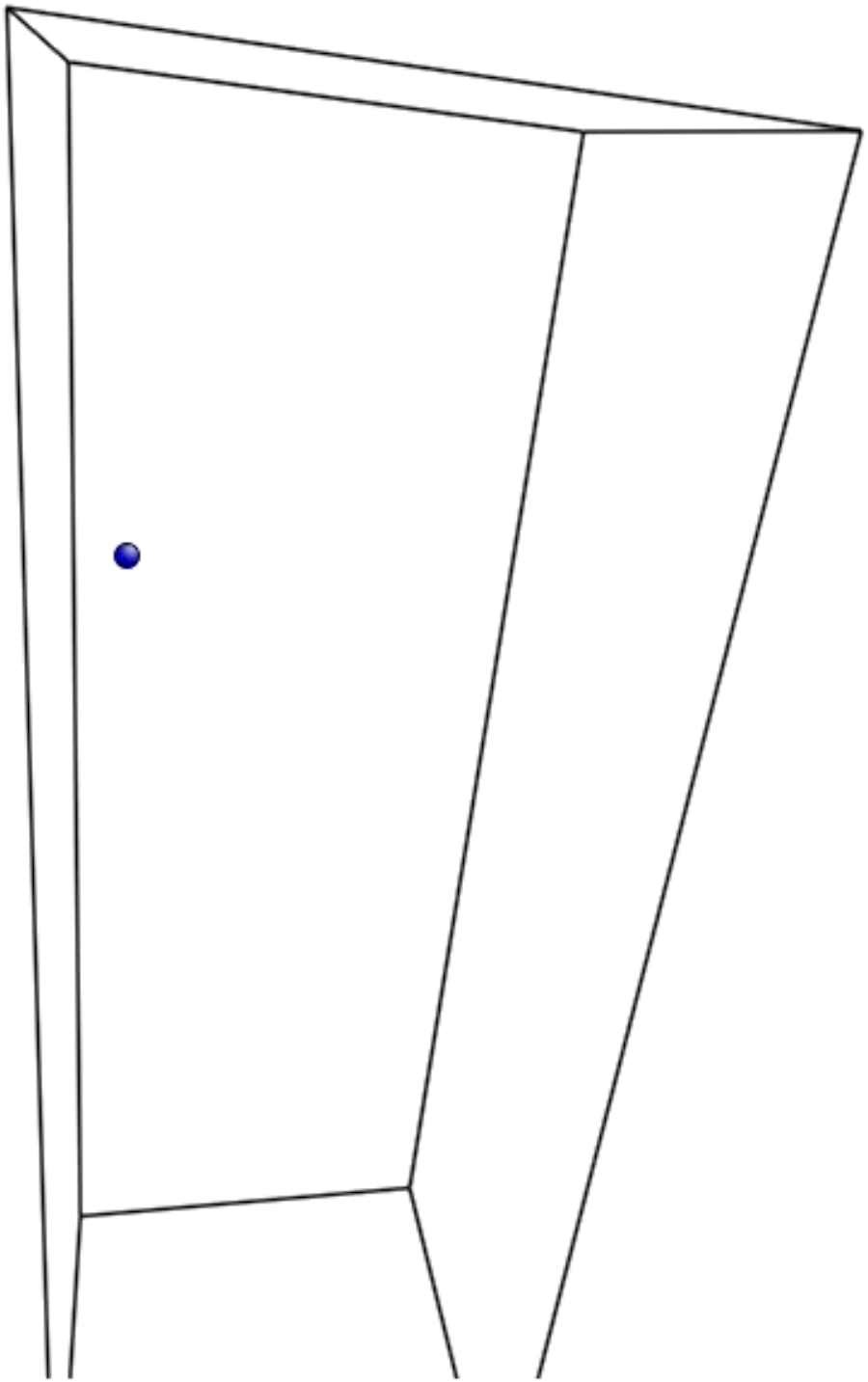
MD indicates no significant difference in sputtering yields due to He ion irradiation



He bubble influence on sputtering



- MD simulations generally consistent with experiment
- Sputtering yields higher on (110) than (100) surfaces, but no significant effect of sub-surface He bubbles

Evaluating He-H exposure conditions

- **Early stage evolution with 90%H-10% He at 1200K**