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

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Workshop on Challenges in Magnetic Fusion Energy

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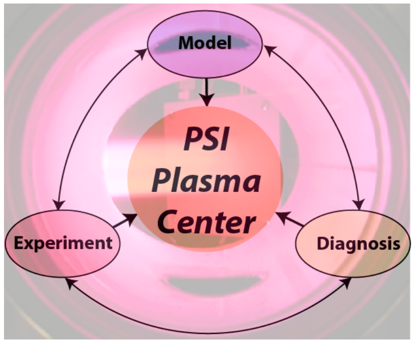

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Presentation Overview

- **Introduction to multiscale phenomena governing the bulk radiation effects in fusion structural materials, the materials dynamics of plasma surface interactions (PSI) and a multiscale modeling approach**
  - Still very much a work in progress – no single, integrated code nor even a well established, validated framework for rigorous multiscale integration of highly non-linear, coupled problems

- **W fuzz formation & MD simulations to address some key mechanisms**
  - Investigating pressure regimes for sub-surface He bubble stability
  - Influence of bubbles on sputtering response
  - Early stage formation of sub-surface gas bubbles

- **Example of spatially-dependent cluster dynamics modeling of He implantation, evolution & desorption in W**

- **Summary and future work**

*Materials issues in Magnetic Fusion Energy (ITER/DEMO)**

- **Magnetic fusion energy presents many materials challenges, including:**
  - High thermal heat fluxes

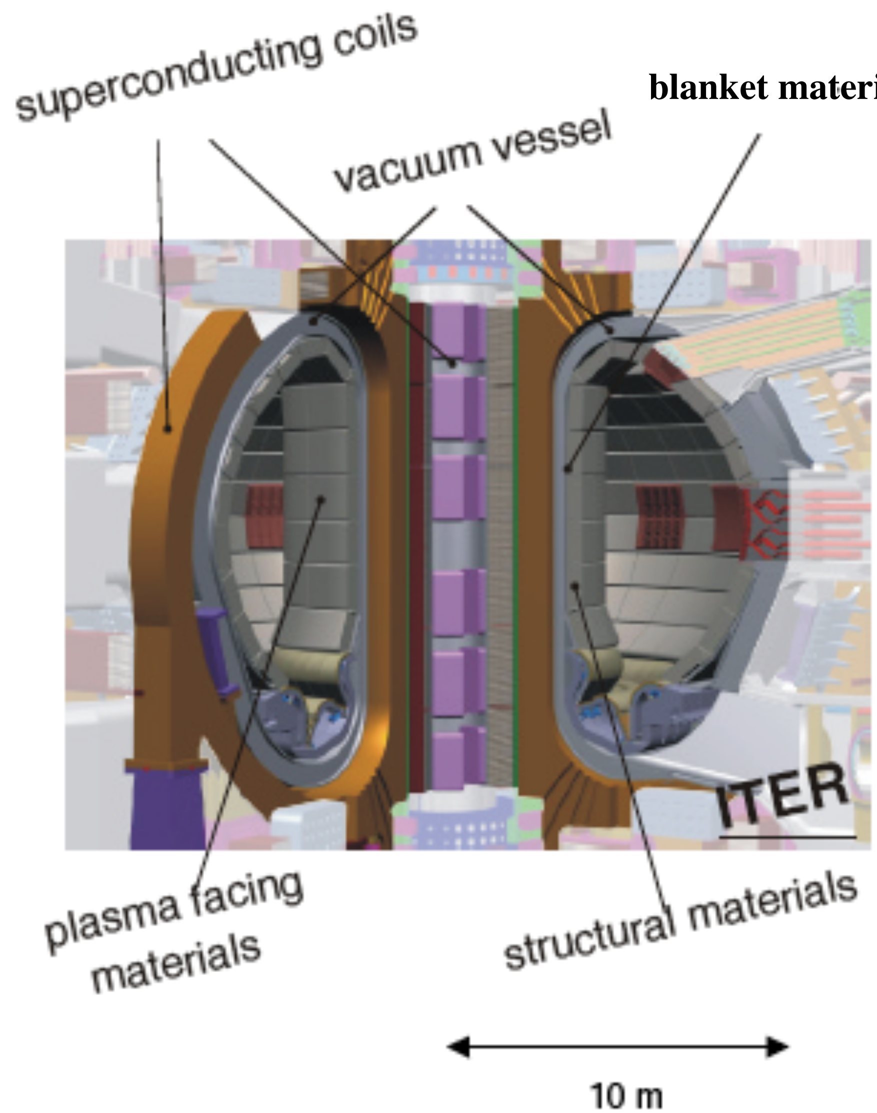
  - Erosion/Sputtering/blistering of plasma facing components

  - Radiation damage

  - Low induced radioactivity

  - Chemical compatibility

  - Joining/Welding

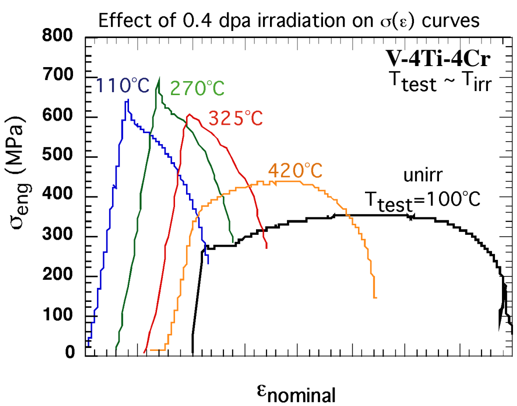


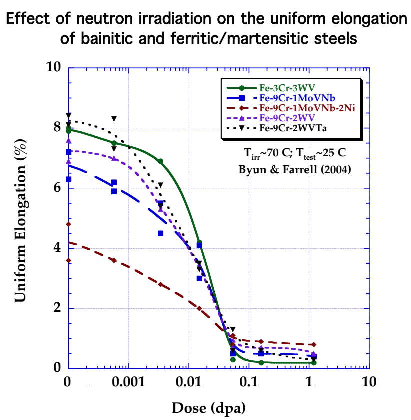
NAE Grand Challenge for Engineering:
2. Provide energy from fusion

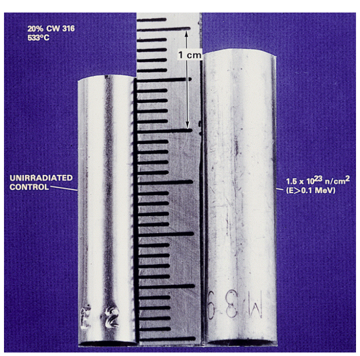
***Ref: H. Bolt, Max-Planck Institute for Plasma Physics, Garching, Germany**

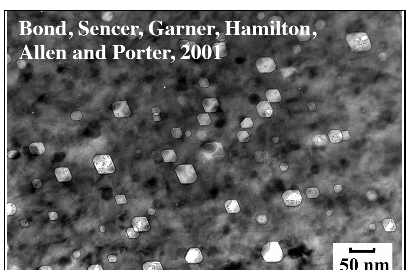
Irradiation effects on structural materials

- Exposure to neutrons degrades the mechanical performance of structural materials and impacts the economics and safety of current & future fission power plants:
  - Irradiation hardening and embrittlement/decreased uniform elongation (< 0.4 T_m)
  - Irradiation (<0.45 T_m) and thermal (>~0.45 T_m) creep
  - Volumetric swelling, dimensional instability & growth (0.3 - 0.6 T_m)
  - High temperature He embrittlement (> 0.5 T_m); Specific to fusion & spallation accelerators
- Additional environmental degradation due to corrosive environments (SCC, uniform/shadow corrosion, CRUD)









304 Stainless steel irradiated in EBR-II, 380°C, ~22 dpa, 1% swelling

Variables

- Structural Materials (Fe-based steels, Vanadium and Ni-based alloys, Refractory metals & alloys, SiC) and composition
- Zr alloy cladding
- Initial microstructure (cold-worked, annealed)
- Irradiation temperature
- Chemical environment & thermal-mechanical loading
- Neutron flux, fluence and energy spectrum
  - materials test reactor irradiations typically at accelerations of 10^2 - 10^4

Synergistic Interactions

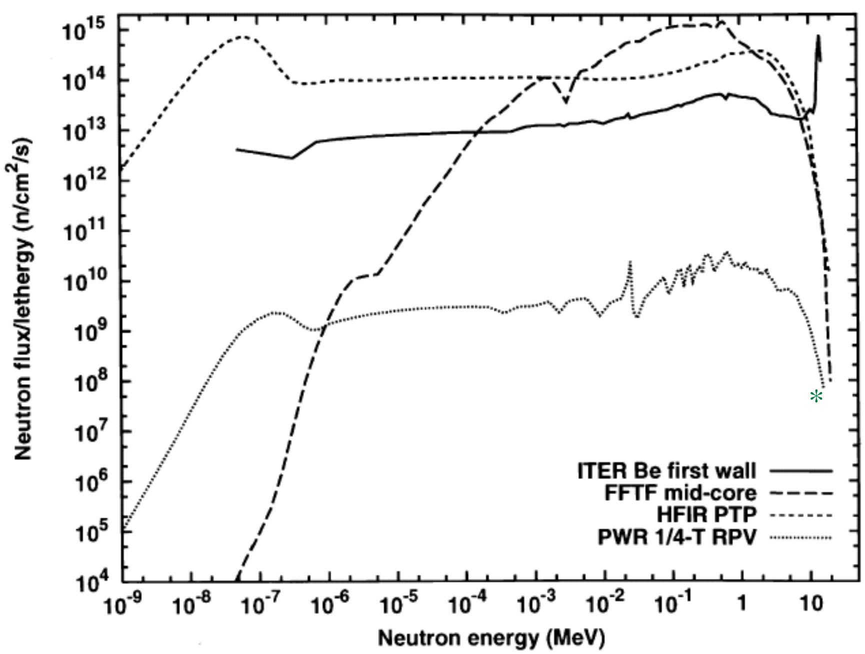
Cause of radiation effects in structural materials

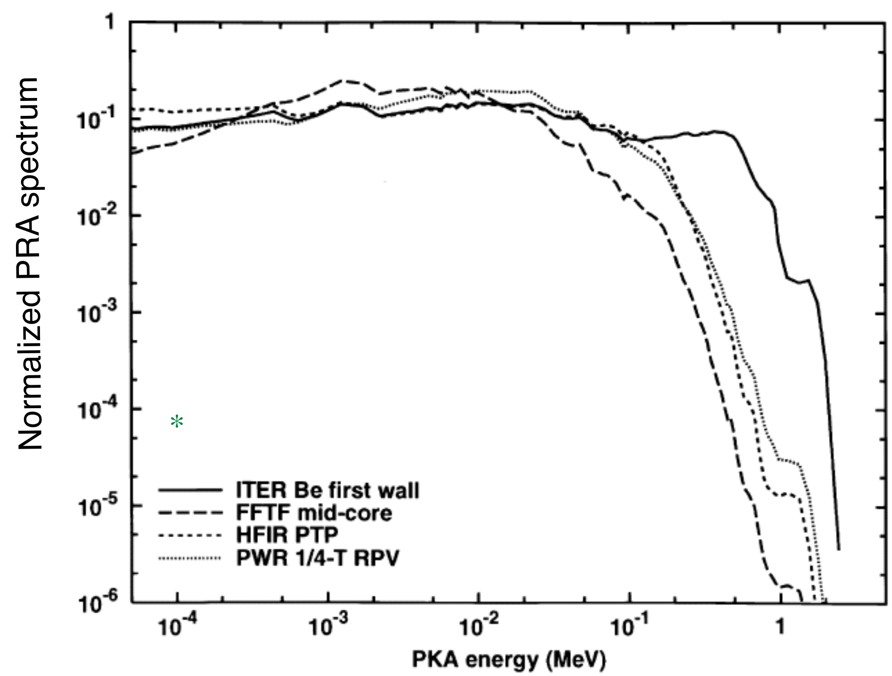
Fast neutrons > ≈ 0.1 MeV major source of *displacement damage*

Fast neutrons > 1 MeV are the major source of *He and H transmutation products*

Average fast fission neutron E ≈ 1-2 MeV, fast fusion neutron energy, E = 14 MeV

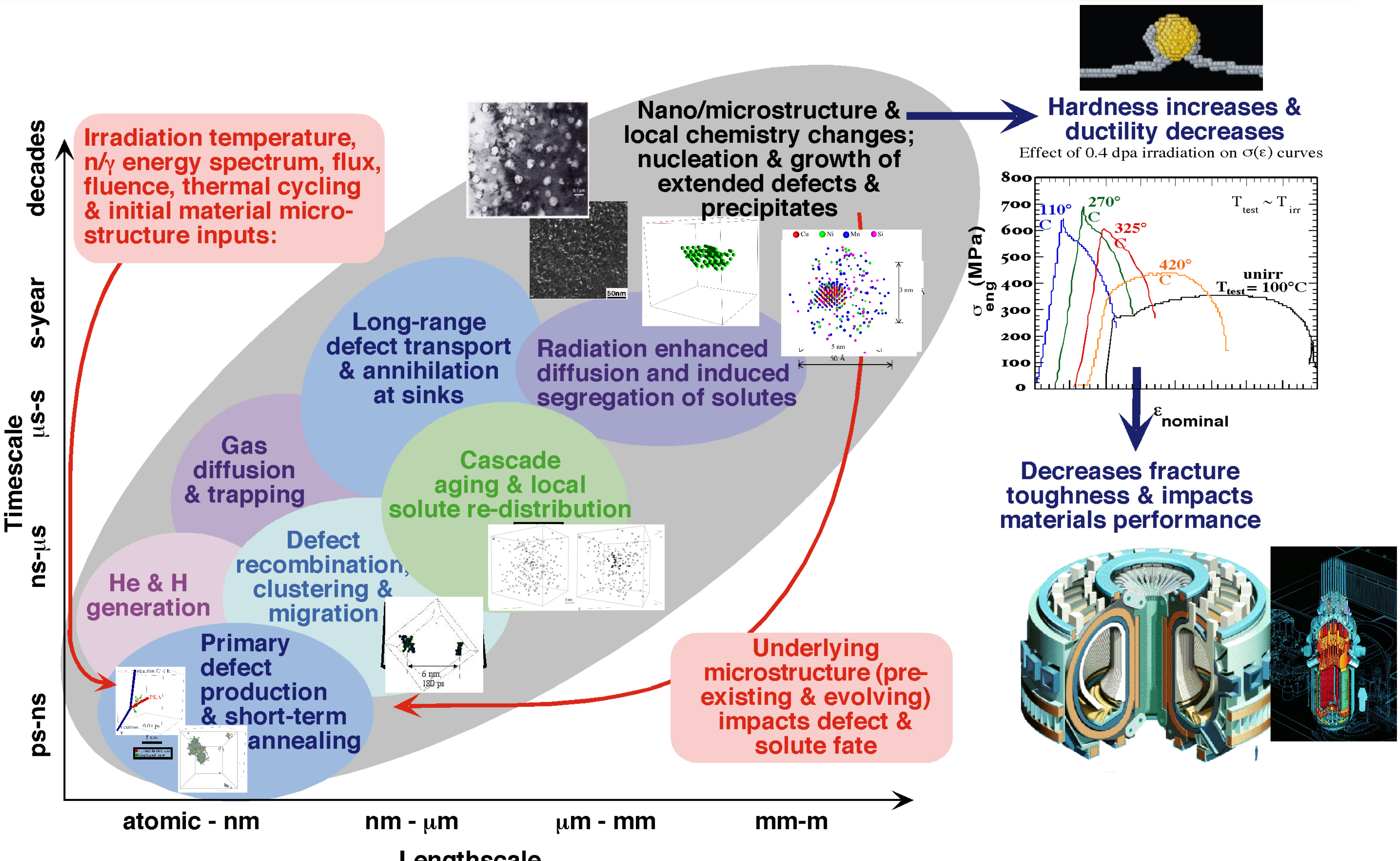
Neutron-nuclear interactions -> primary recoiling atoms (PRA)





Source:* Stoller and Greenwood, *JNM* **271&272 (1999) 57.

Radiation effects on materials is inherently multiscale



Radiation damage produces atomic defects and transmutants at the shortest time and length scales, which evolve over longer scales to produce changes in microstructure and properties through hierarchical and inherently multiscale processes

*Plasma Facing Components/Materials (ITER)**

<u>Key issues</u>

- erosion lifetime and plasma compatibility
- tritium inventory
- thermal transients
- **He blistering**
- heat removal:
- fabrication technology:
- neutron damage:

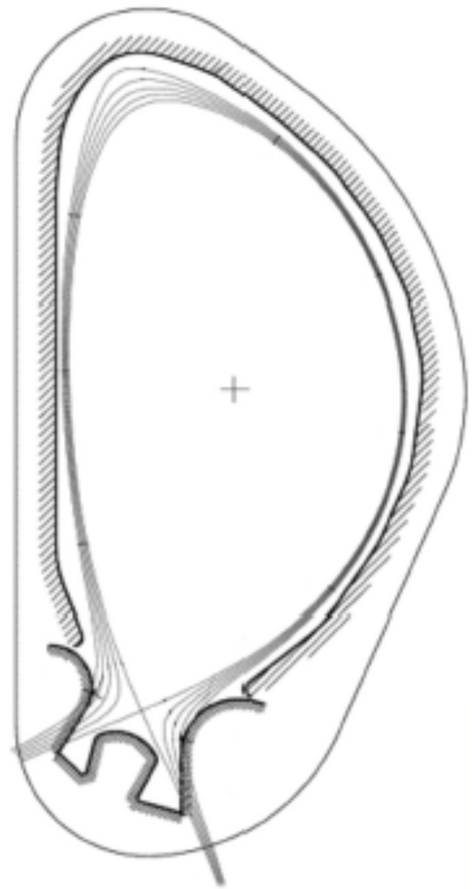
<u>Leading candidate materials</u>

PFC and Divertor:

• Be, W, C

Structural components:

• Fe-Cr steels, V-Cr-Ti, SiC



bulk plasma:

impurity tolerance

W < 2 10^{-5}, reactor < 10^{-4}

Be, C: 10^{-2}

first wall:

modest flux of high energy neutral particles (100s eV),

low energy ions

divertor target:

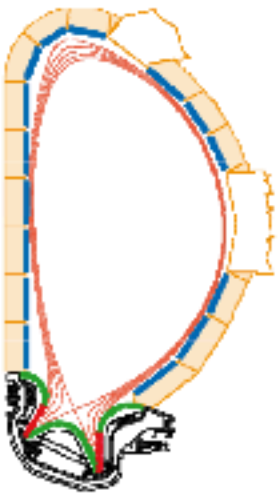
high heat flux 10 (20) MW/m²

<u>transient heat loads</u>:

e.g. ELMs, disruptions

*Ref: H. Bolt, Max-Planck Institute for Plasma Physics, Garching, Germany

*PFC Materials: Surface chemistry evolves as well**



ITER

- First wall on ITER
  - → carbon 55 m^2
  - → tungsten 140 m^2
  - → beryllium 690 m^2
- DEMO first wall / divertor
  - → oxidation-resistant W alloys (e.g. W—Si—Cr)
- Variable local conditions (temperature, fluence, species…)
- Erosion and redeposition, impurities:
  - → mixed phases (e.g. carbides, oxides, alloys)
- Layers on metals influence:
  - → hydrogen inventory: reaction, diffusion, desorption
  - → physical and chemical processes: sputtering, reactions
- Goal: qualitative and quantitative description of fundamental processes
  - → formation and erosion of multi-component layers
  - → influence of layers on hydrogen inventory

→ **Include surface reactions in global integrated PWI model**

Ref: * C. Linsemeier, PSI-19 Invited Presentation (2010).

*Complex, interlinked PSI phenomena**

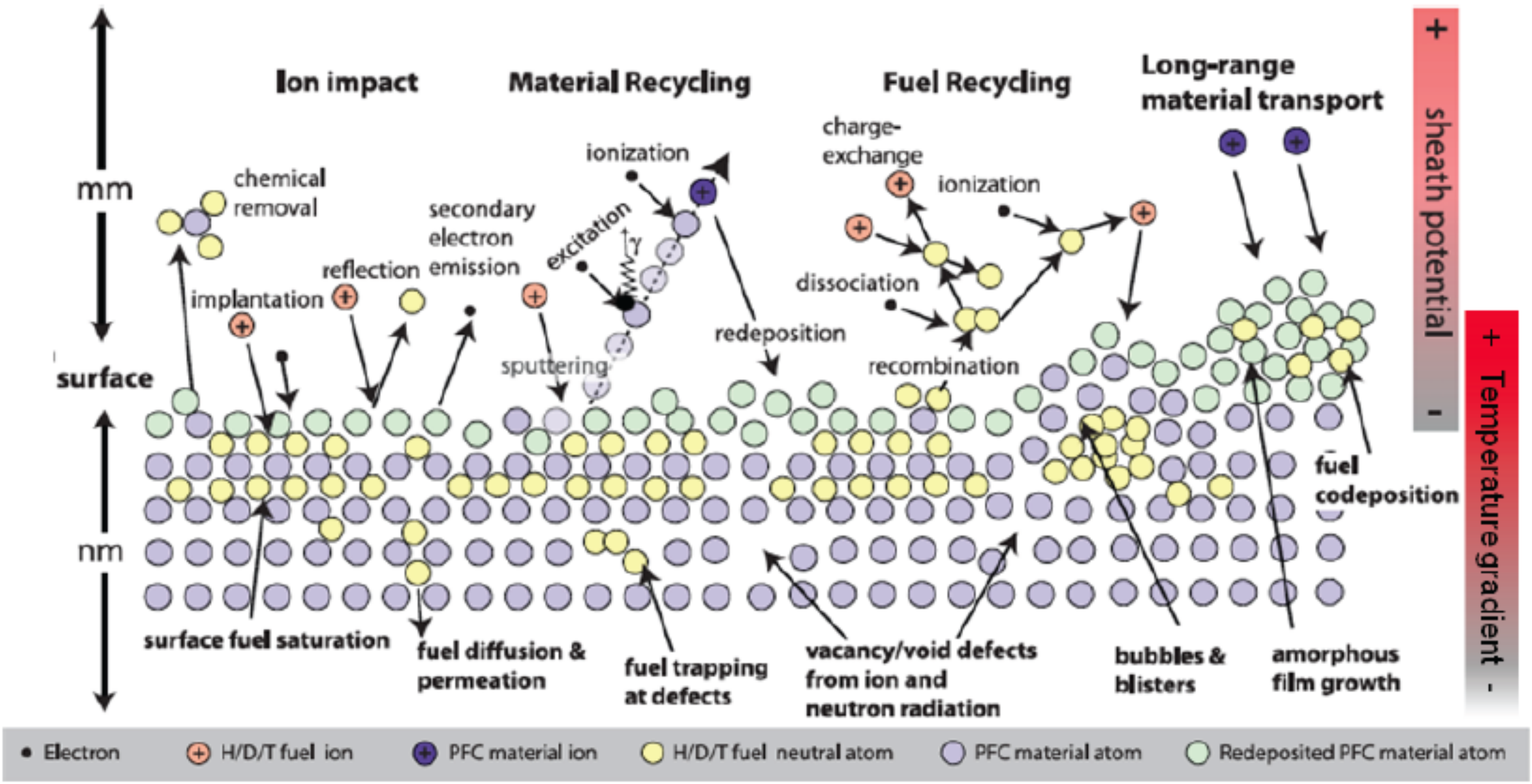


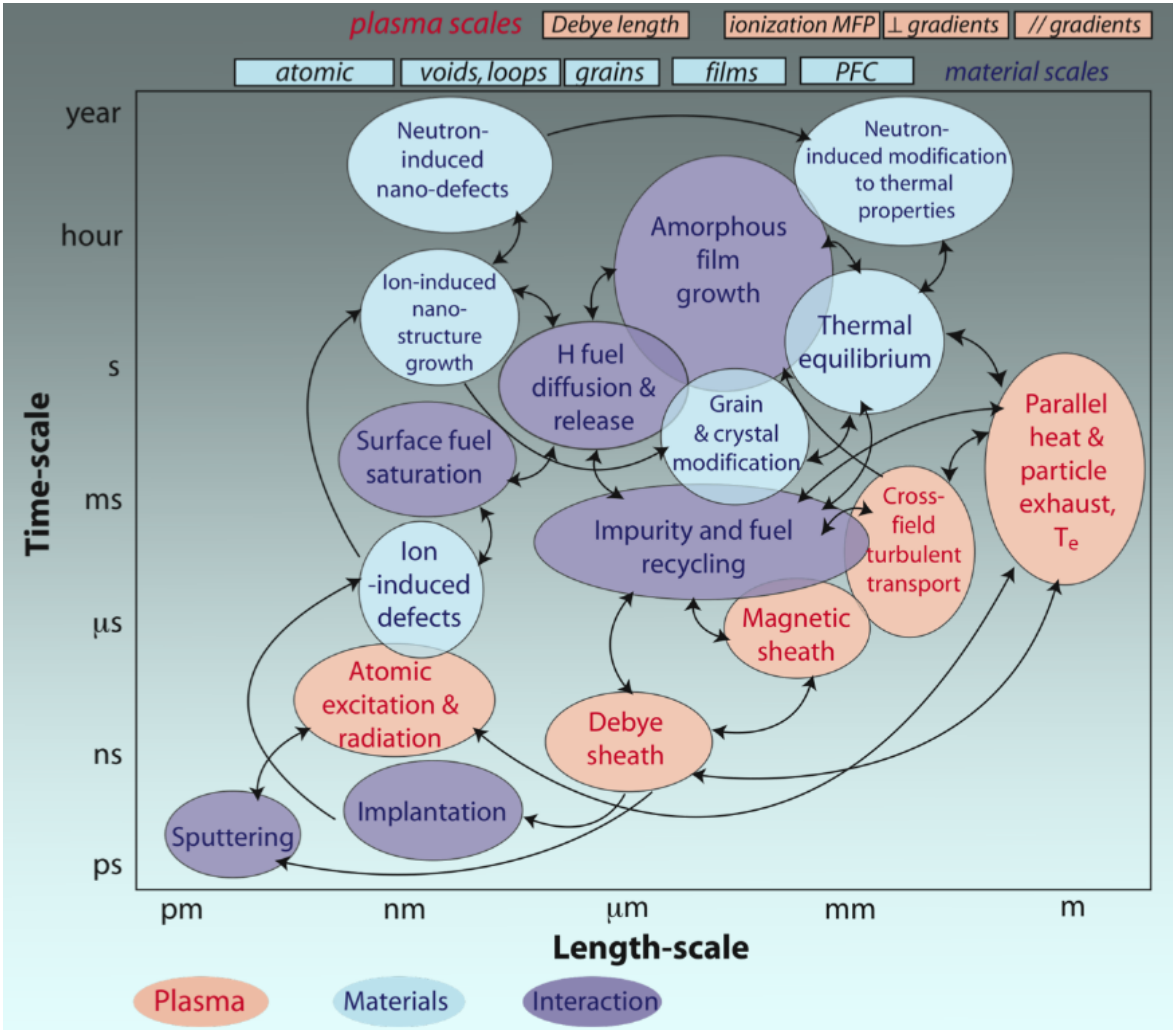
Figure of merit:

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, interlinked Plasma-Surface Interaction phenomena**



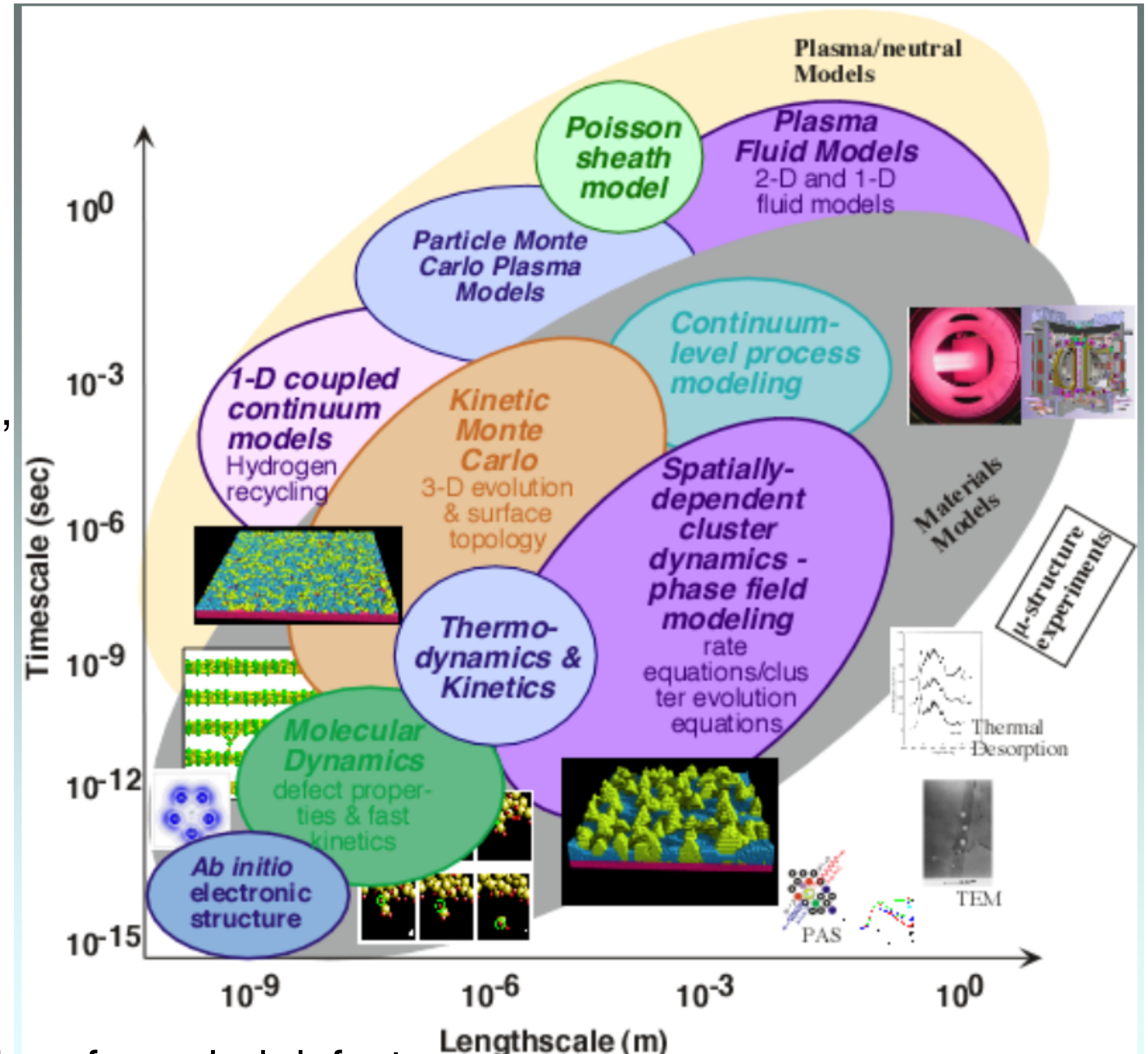
* Wirth, Nordlund, Whyte, and Xu, *Materials Research Society Bulletin* **36** (2011) 216-222

*Multiscale modeling capability – a work in progress**

Goal: Discovery science to obtain clues to W nanofuzz formation mechanism & timescale 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

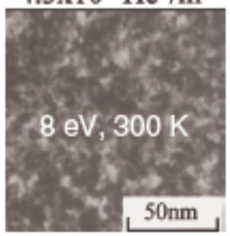


Biggest long-term scientific challenge is understanding the kinetics of coupled defect – impurity evolution with a disparate range of kinetic rates

* Whyte & Wirth, unpublished

W Surface dynamics under combined thermal/particle fluxes

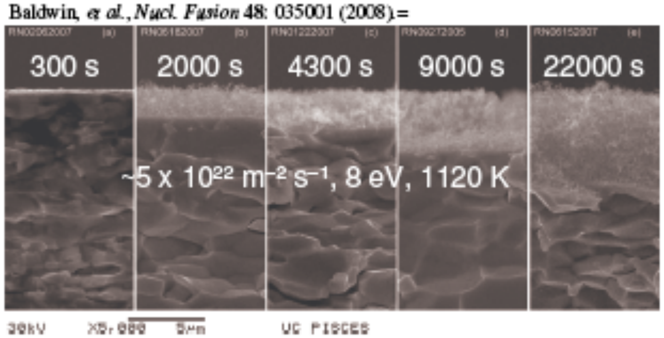
T < 700 K



H. Iwakiri, *et al.*, *J. Nucl. Mater.* 283–287: 1134–1138 (2000).

non-specific damage

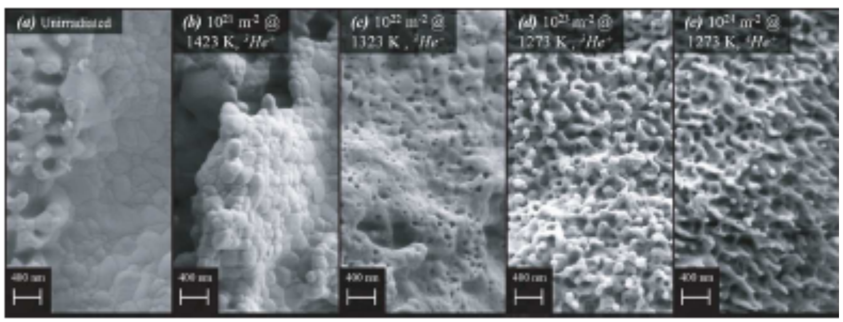
Baldwin, *et al.*, *Nucl. Fusion* 48: 035001 (2008).



900 K < T < 1900 K

fuzz / coral

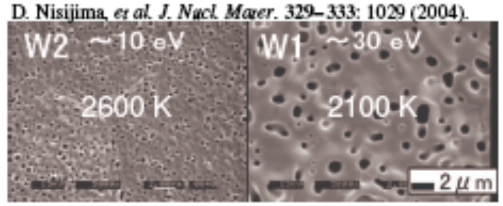
S. J. Zenobia and G. L. Kulcinski. *Phys. Scr.* T138: 014049 (2009).

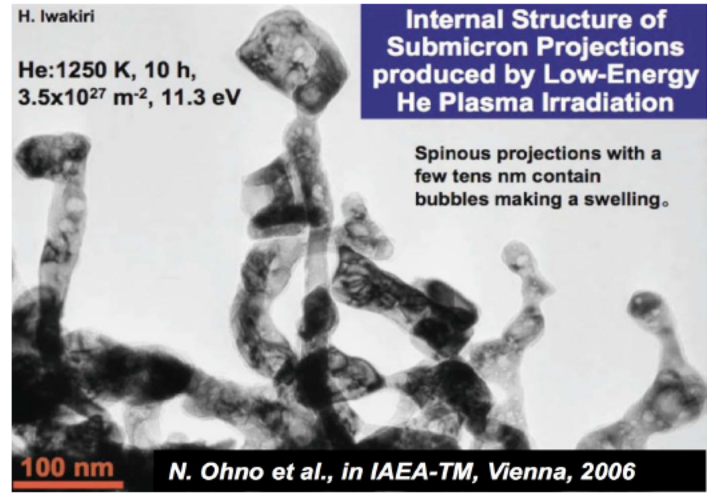


T > 2000 K

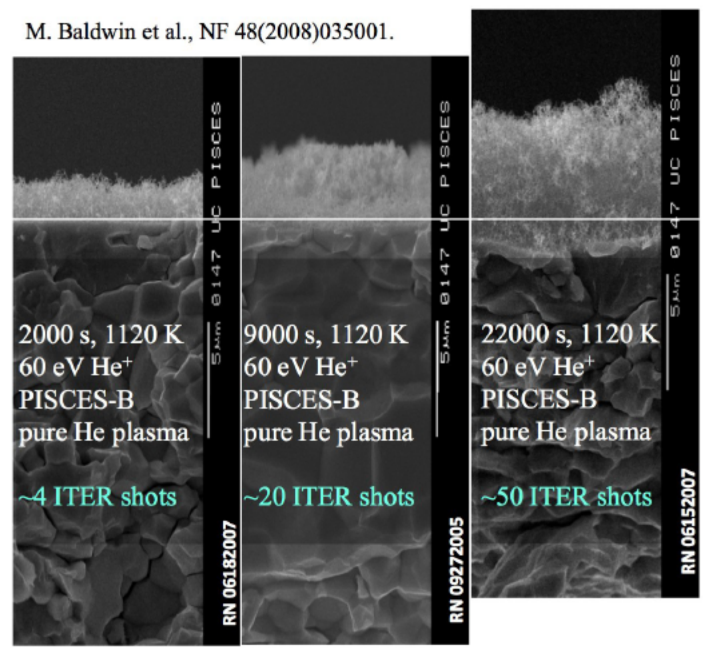
holes

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



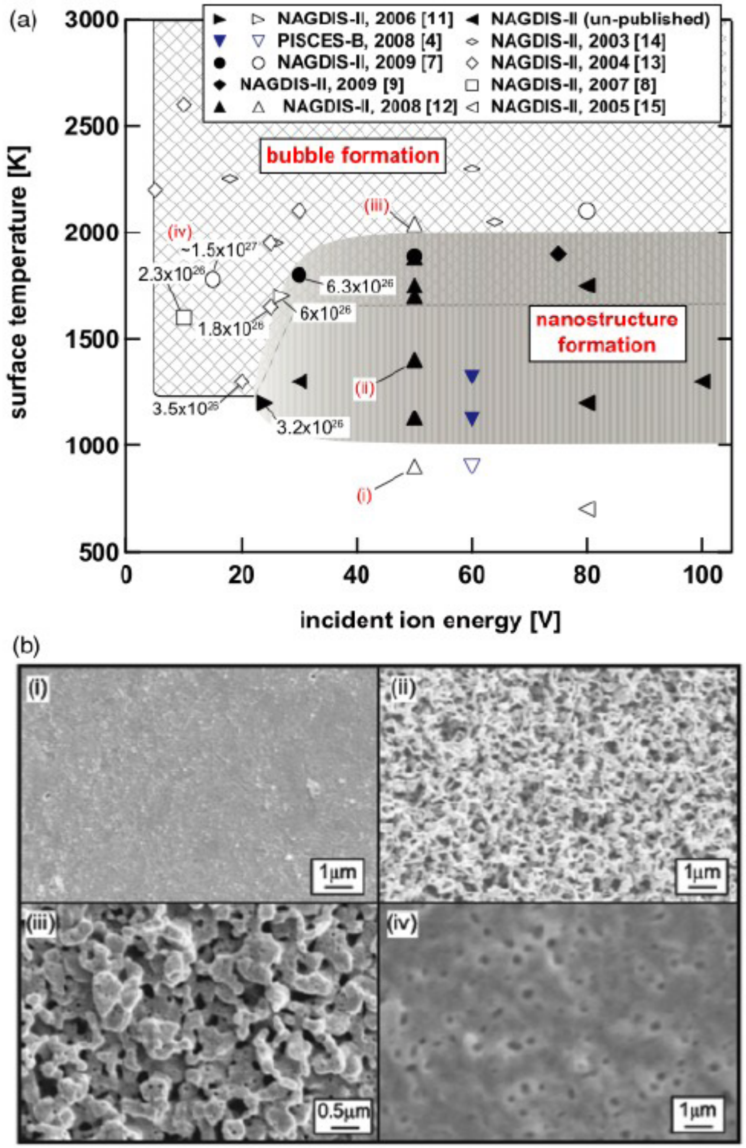


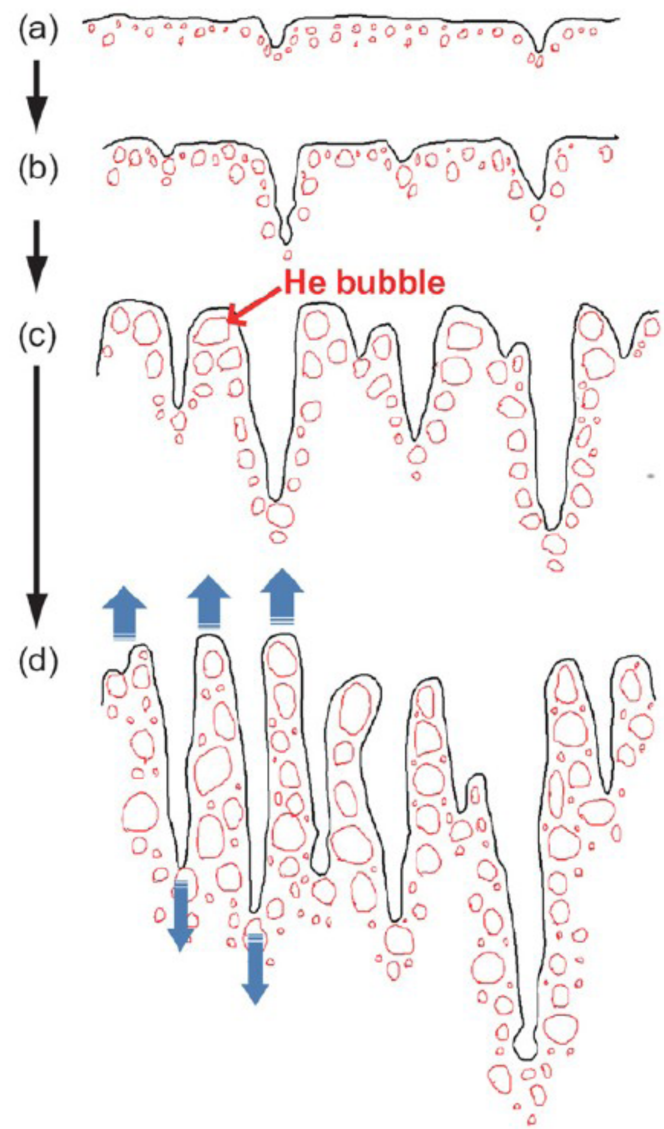
M. Baldwin et al., NF 48(2008)035001.



*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**

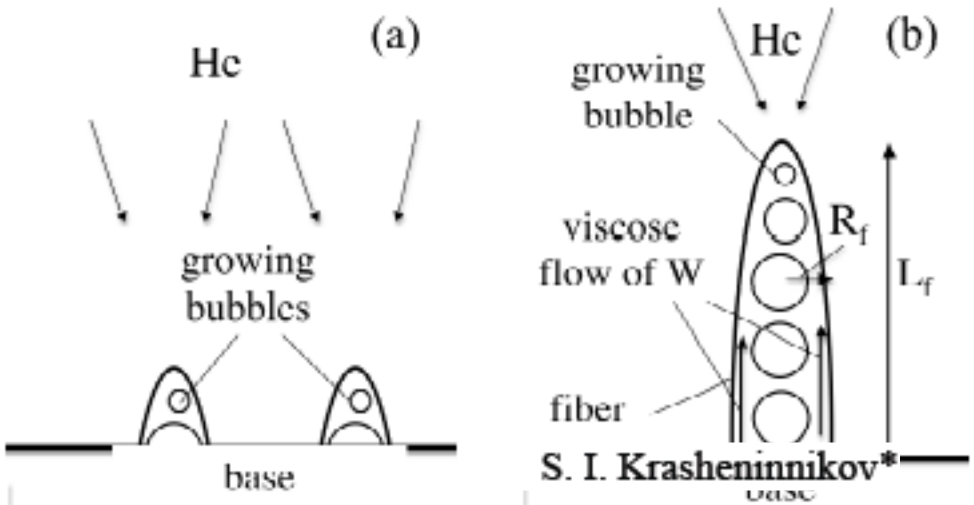


Fig. 2. Schematic views of: (a) initial stage of the fiber growth; (b) developed fiber; (c) viscose flow of W to the tip of the fiber due to the force caused by the pressure of the He in the growing fiber.

$$F_C = P_{He}\pi R_f^2, \quad (2)$$

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

$$P_{He} \sim 2\gamma / R_f, \quad (3)$$

where γ is the tungsten surface tension coefficient.

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

$$\sigma_0 \sim \frac{F_C}{2\pi R_f \delta_f} \sim \frac{2\gamma}{2\delta_f}. \quad (4)$$

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

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

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:

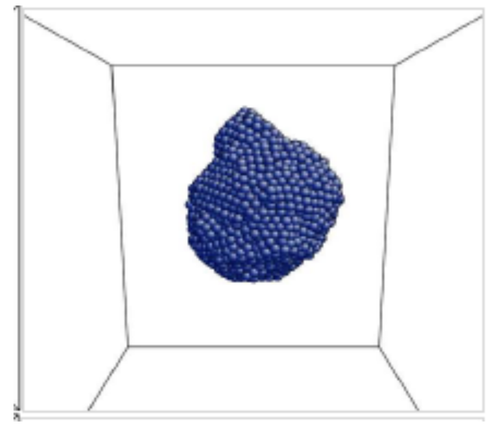
$$L_f(t) = \sqrt{\frac{2\gamma\delta_f}{\mu_W} t}. \quad (6)$$

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

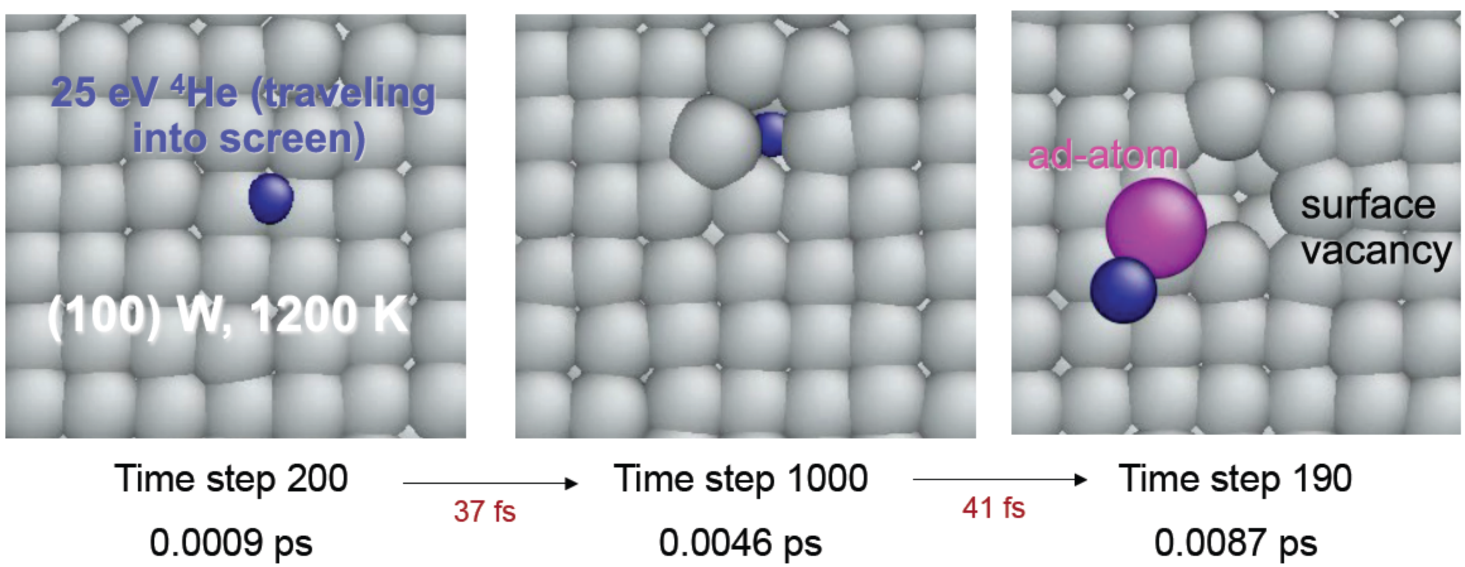
MD simulations: sub-surface He bubbles

- 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

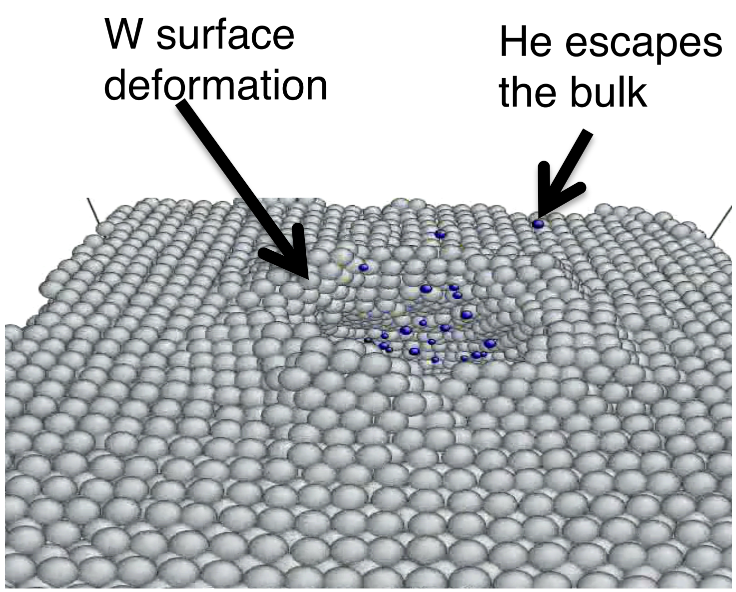


How do these processes influence surface topology evolution, sputtering, etc. & can sub-sputtering threshold He exposure drive surface evolution processes?



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**

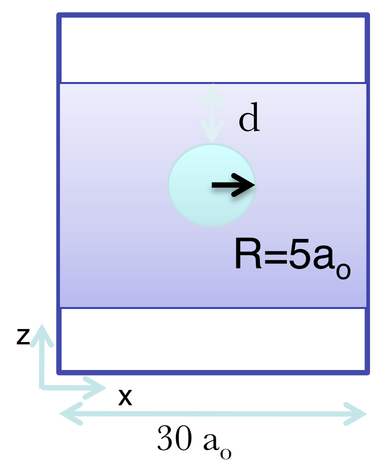


- Bubble bursts
- He escapes from the bulk
- The W "crater" stays in place
  Permanent damage (MD timescales)
- No W erosion

Parameters:

Temperature (K)
500
1200
2000

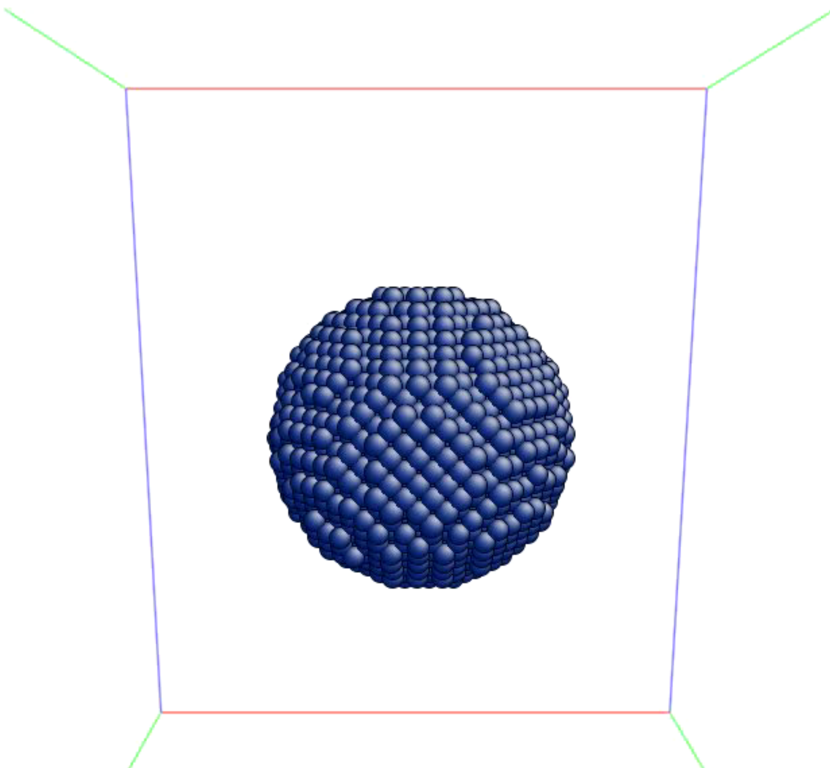
Distance of Bubble to the surface
d=2 a_o
d=3 a_o
d=5 a_o=R
d=10 a_o=2R



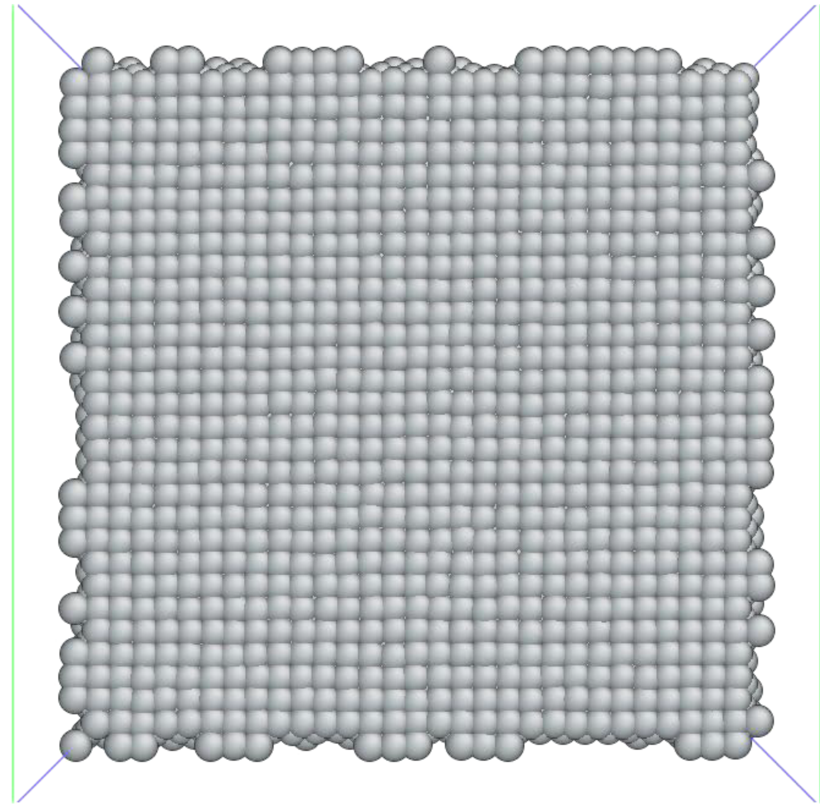
An over-pressurized, sub-surface He bubble

Distance below the surface = 5ao, bubble radius = 5ao

View of bubble evolution: He blue, W invisible



View of surface deformation: W grey, He blue



Pressure evolution of He bubbles

Molecular dynamics simulations to assess He bubble pressure & response of over-pressurized, sub-surface bubbles ($R \approx 1.6$ 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

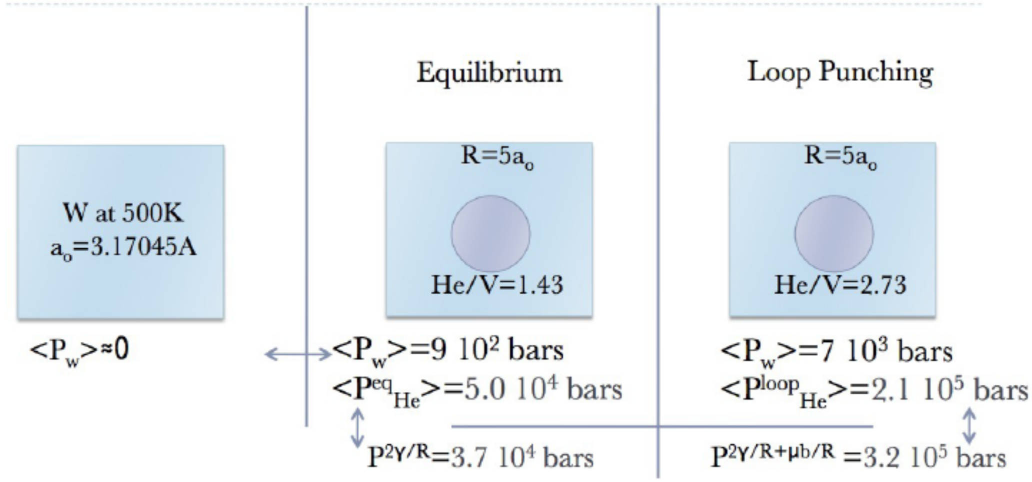
$P_{eq}=\gamma/2R$

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 $P_{loop}=\dfrac{2\gamma}{R}+\dfrac{\mu b}{R}$
- 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 $P_{loop}=\dfrac{2\gamma}{R}+\dfrac{\mu b}{R}$
- Compare P_{bulk} and P_w

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

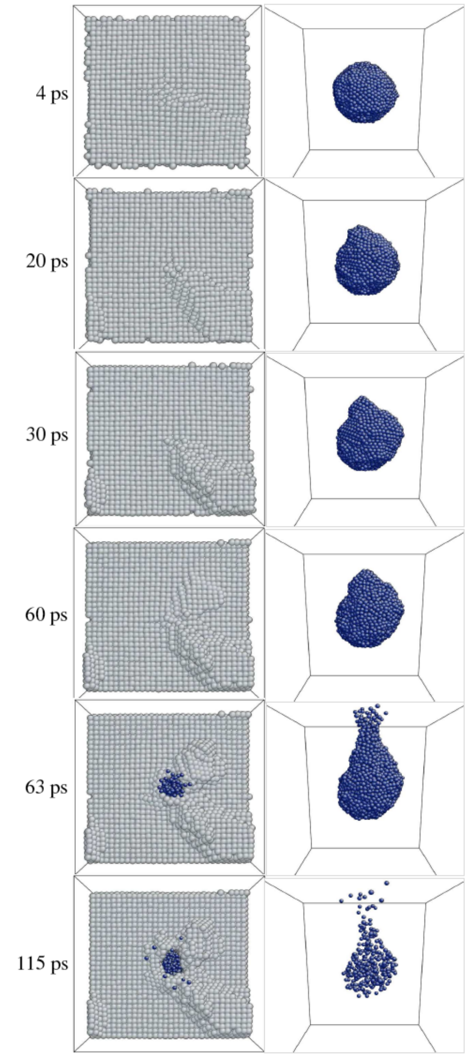
Pressure evolution of He bubbles

- 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

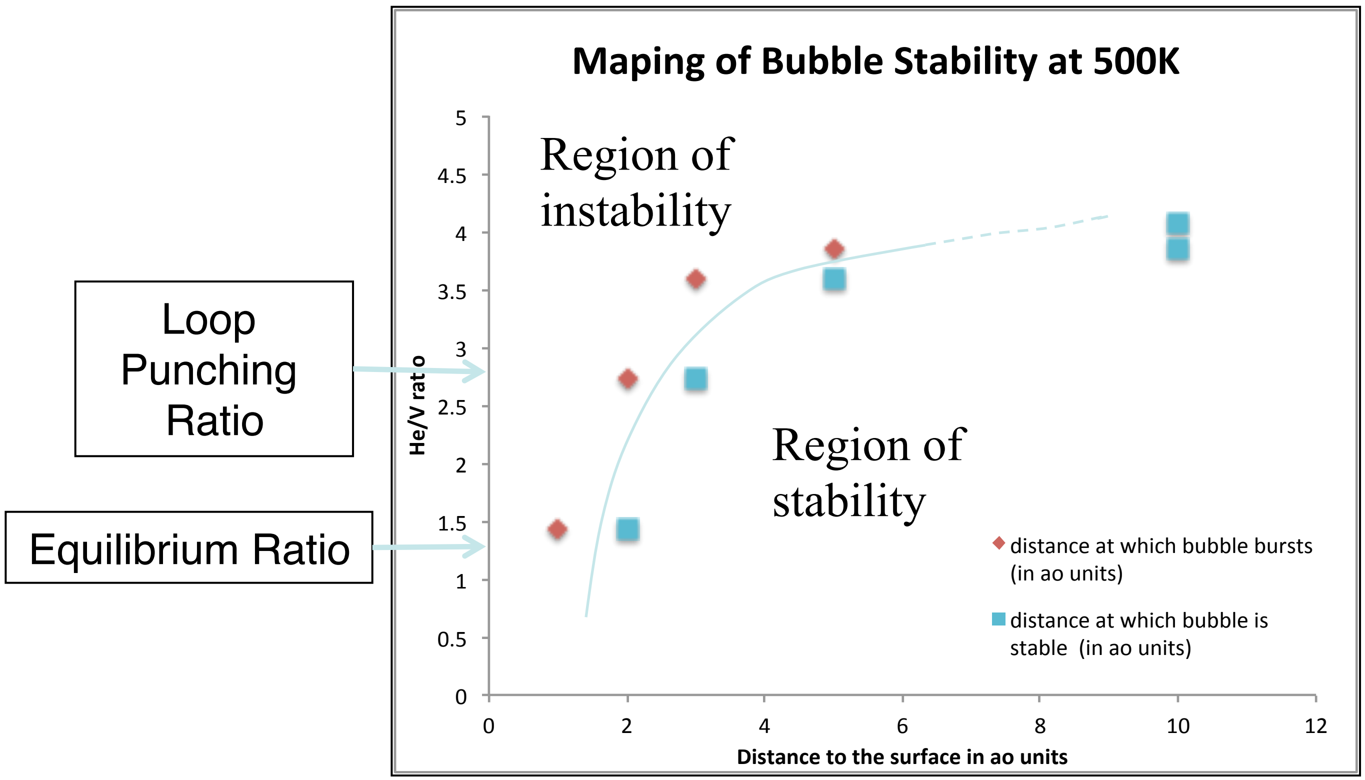
- $d < R \rightarrow$ bubble bursts for He/V < He/V^{loop}
- $d = R = 5\ a_o$:
  Number of He for $P^{2\gamma/R+\mu b/R}$ is 3787 and He/V=3.63
  Number of He for bursting is 4025 and He/V=3.85
  (4025-3727)/3727 = 6% discrepancy in number of heliums between loop punching pressure and bubble bursting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



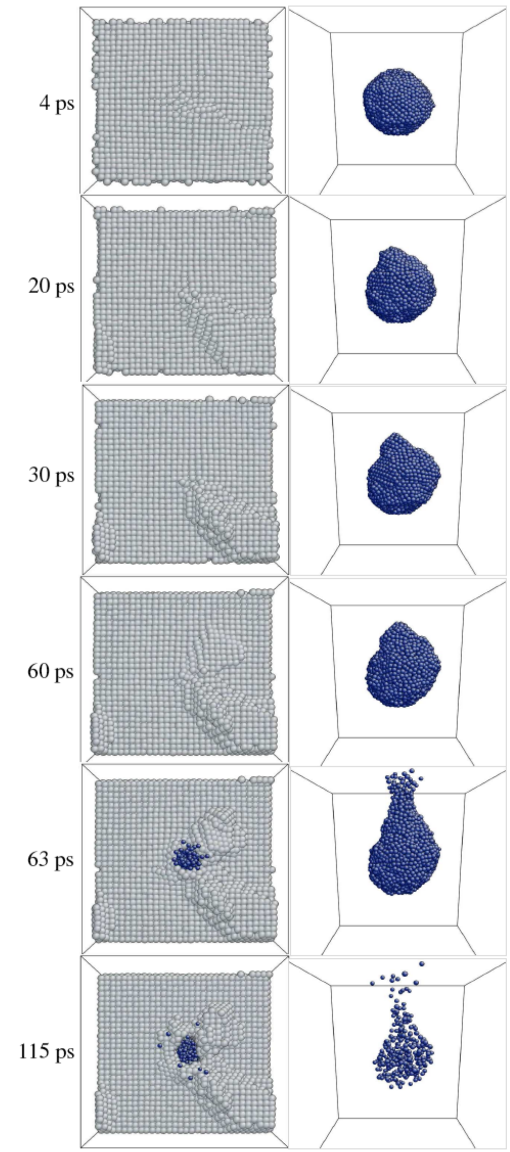
movie

Pressure evolution of He bubbles

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



Snapshots of a bubble rupturing with $d=5a_o$

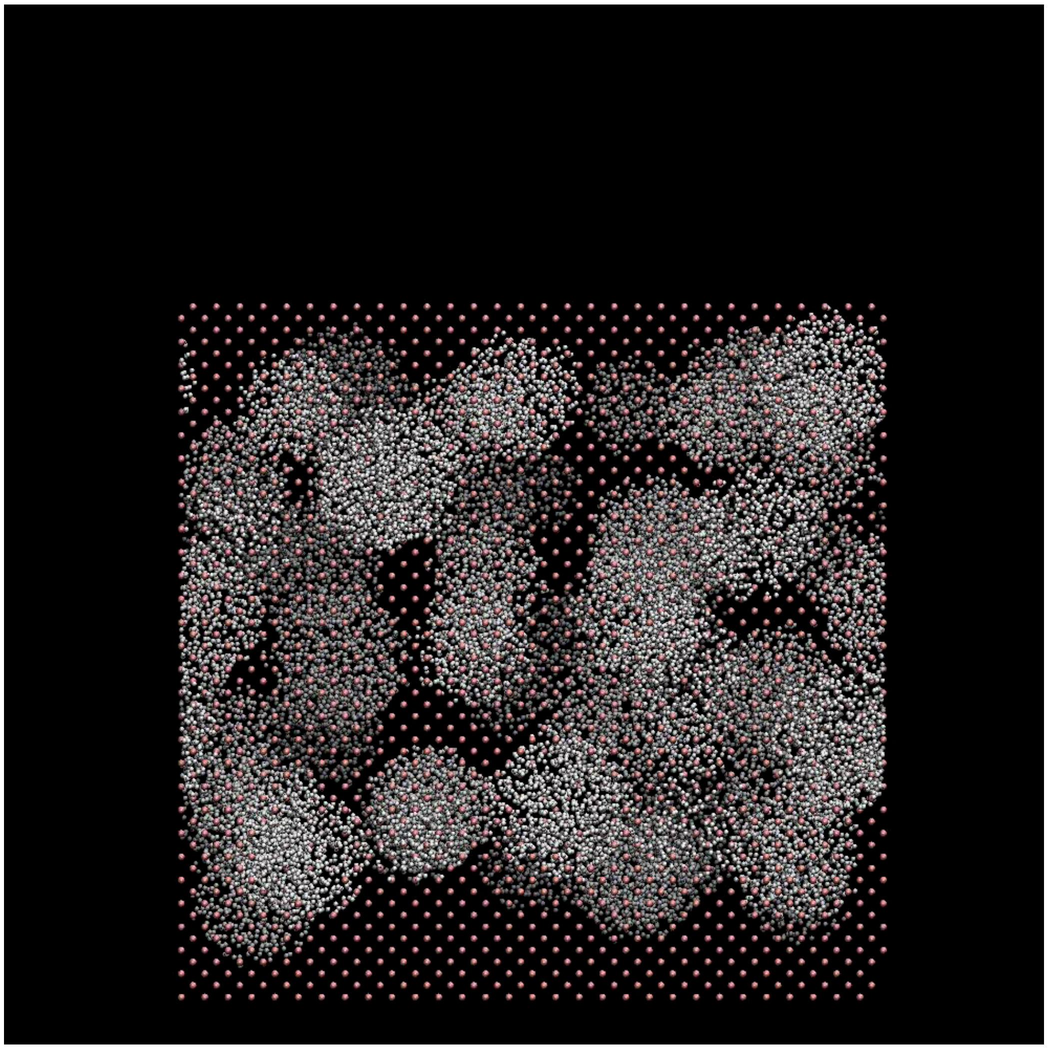


- d<R the bubble bursts for He/V < He/V^{loop}
- $d=R=5a_o$ He/V ratio for bursting corresponds to the loop punching pressure
- $d=2R=10a_o$ Bulk behavior, the bubble is far enough for the surface not to burst

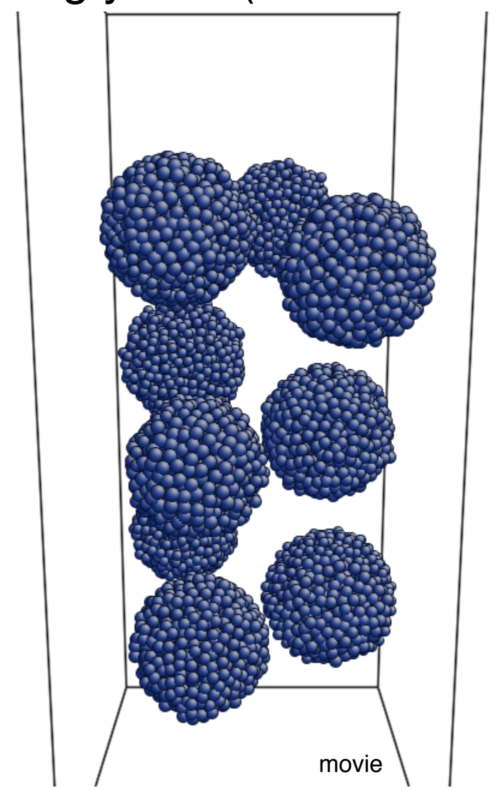
He bubble influence on sputtering & evolution

- How do sub-surface He bubble influence sputtering?
- How do sub-surface He bubbles grow to drive burst phenomena

MD of evolution of He bubble population at 600K
R ~ 0.9 ± 0.5 nm, 2 He/V, d > 0.3 nm

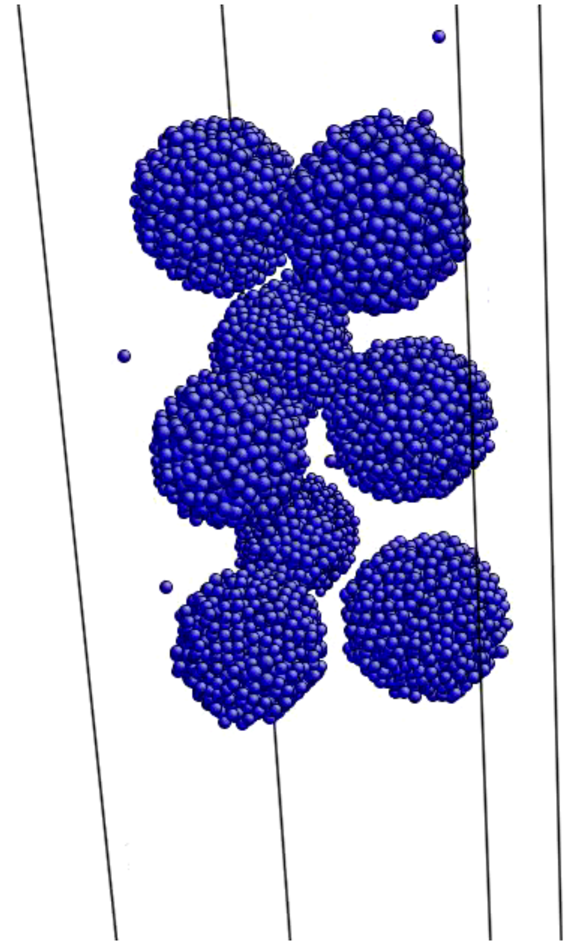


9 bubbles inserted d > 1.6 nm below (100) W surface, 1.2 < R < 1.4 nm, P = 2γ/R
15% He bubble fraction
- Evaluate He induced sputtering yields (300-1000 eV)



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



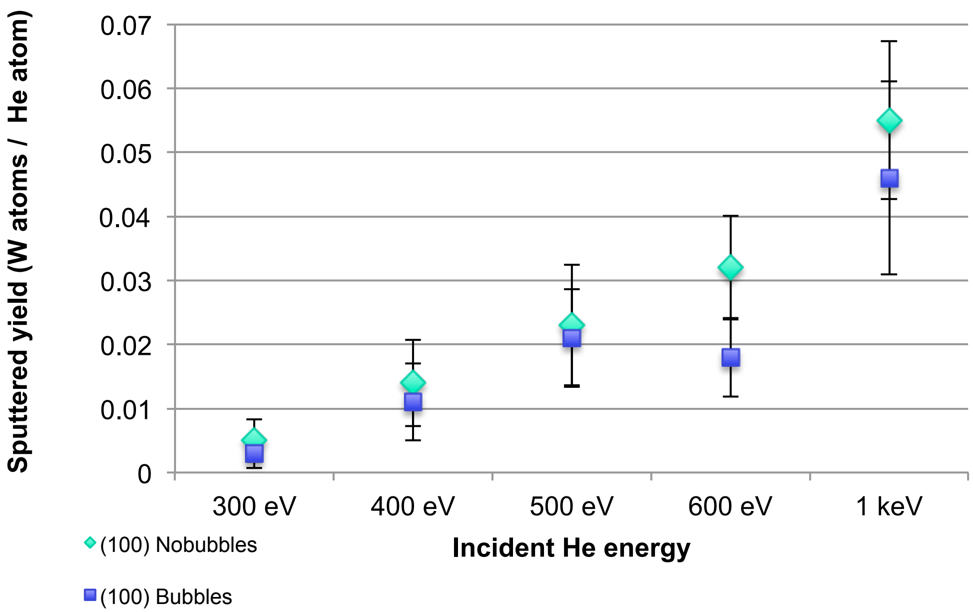
Conditions of the simulation

- 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 ≈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

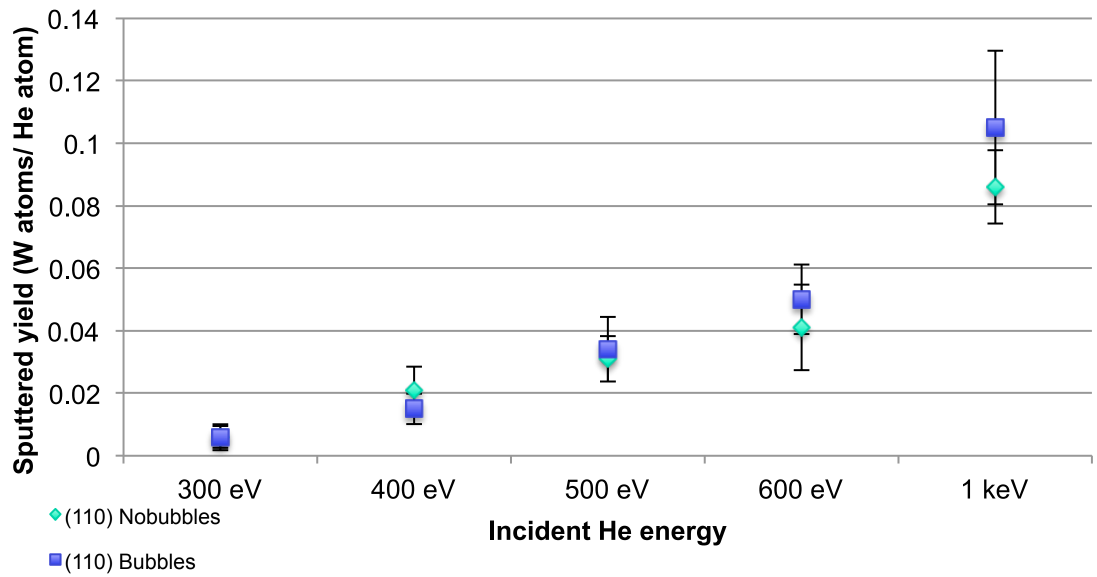
He bubble influence on sputtering

Sputtering of W on the (100) surface

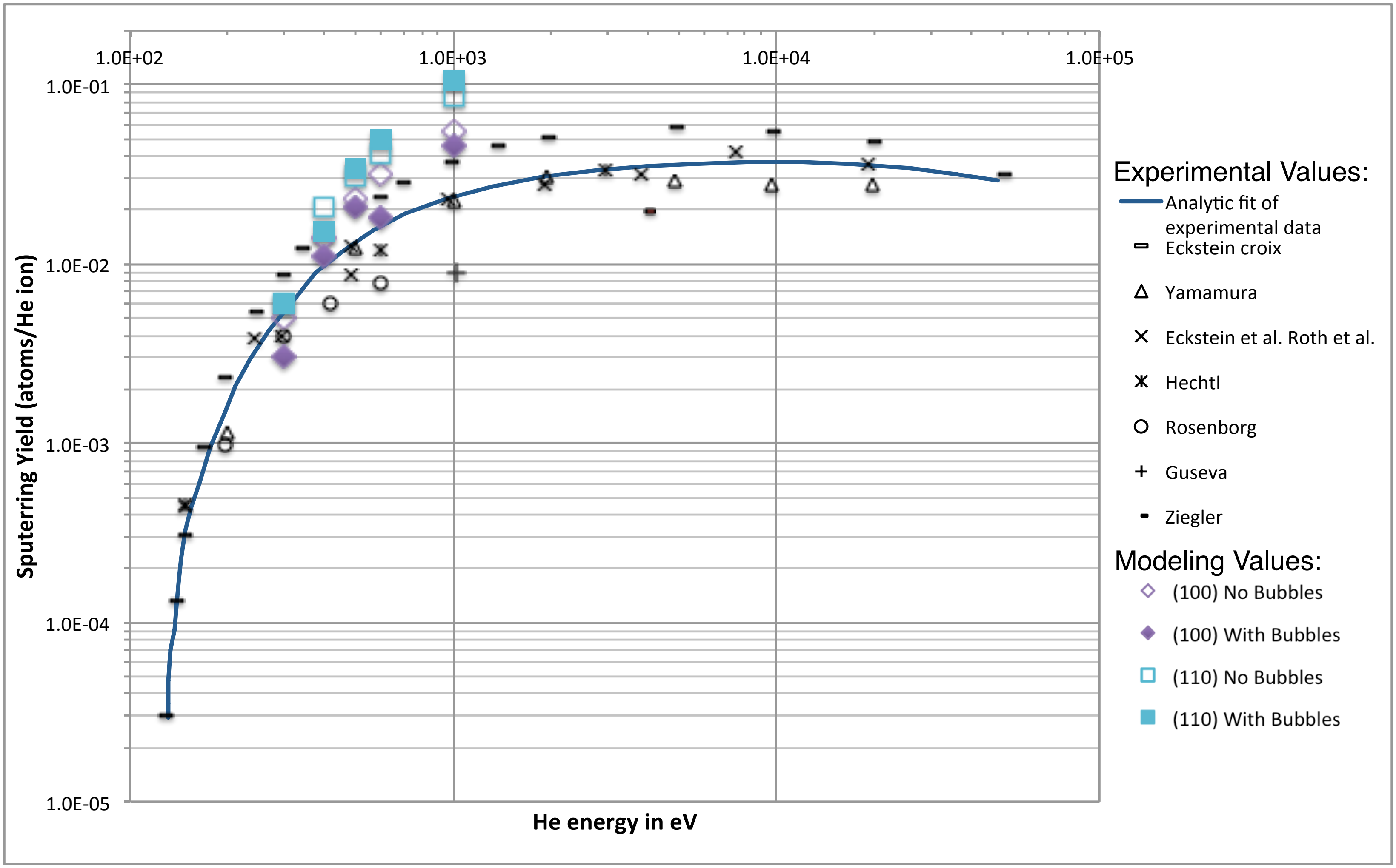


MD indicates no significant difference in sputtering yields due to He ion irradiation

Sputtering of W on the (110) surface



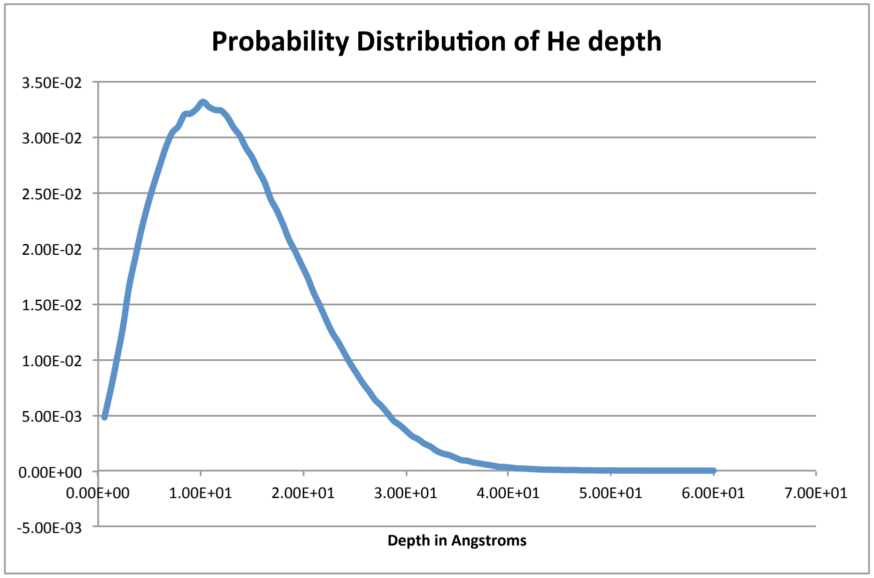
He bubble influence on sputtering

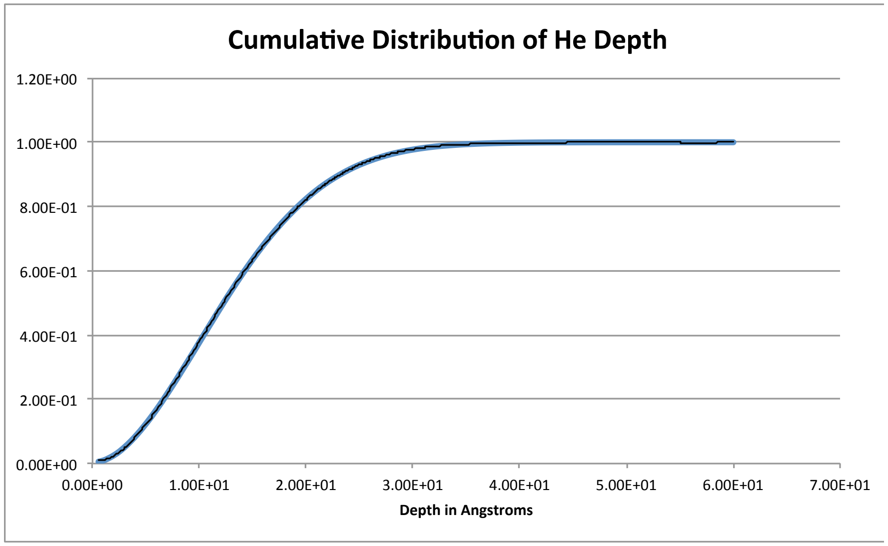


- MD simulations generally consistent with experiment
- Sputtering yields higher on (110) than (100) surfaces, but no significant effect of sub-surface He bubbles

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,

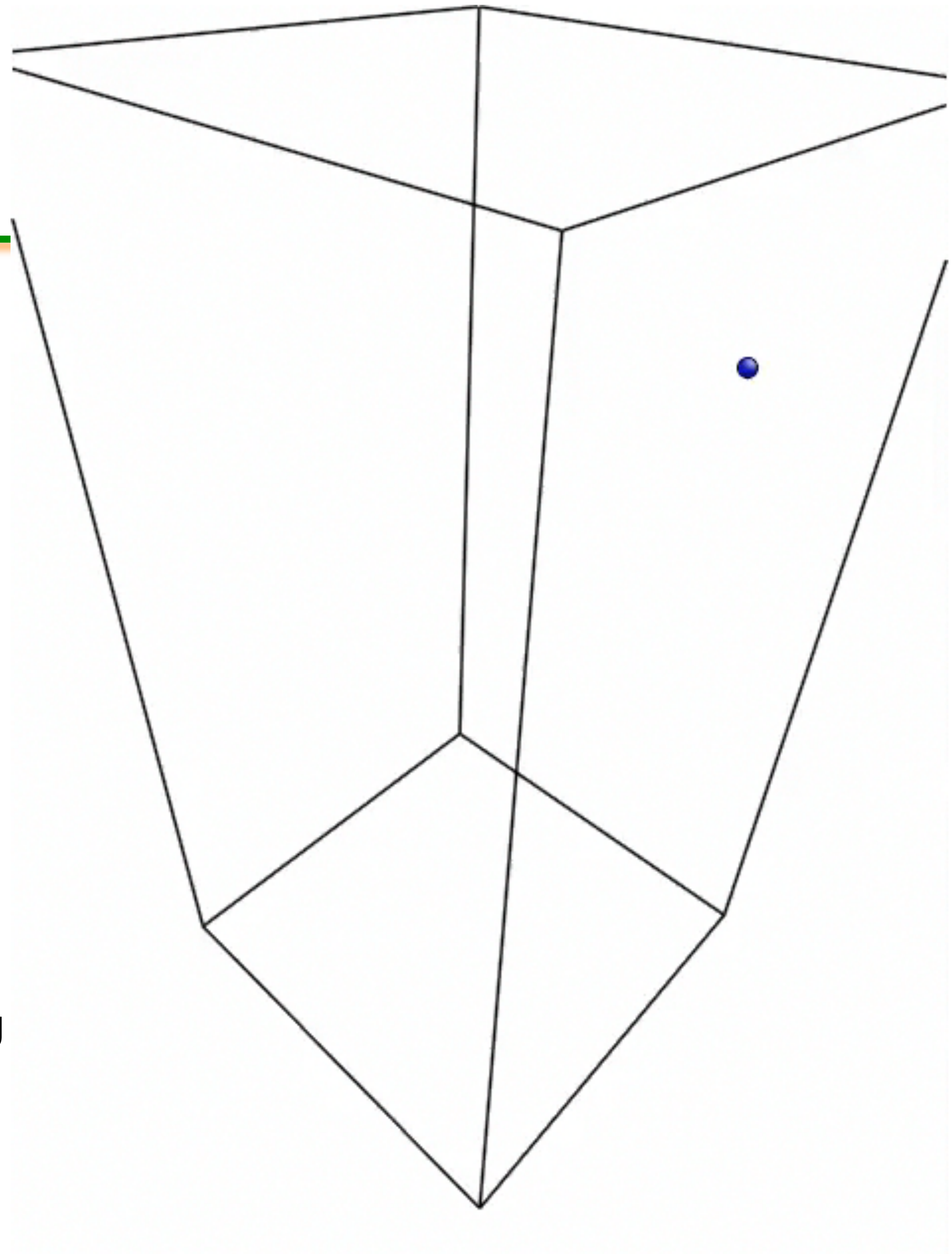
'Thermal' He introduced every 10 ps (very large flux acceleration)

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

~65% of He retained

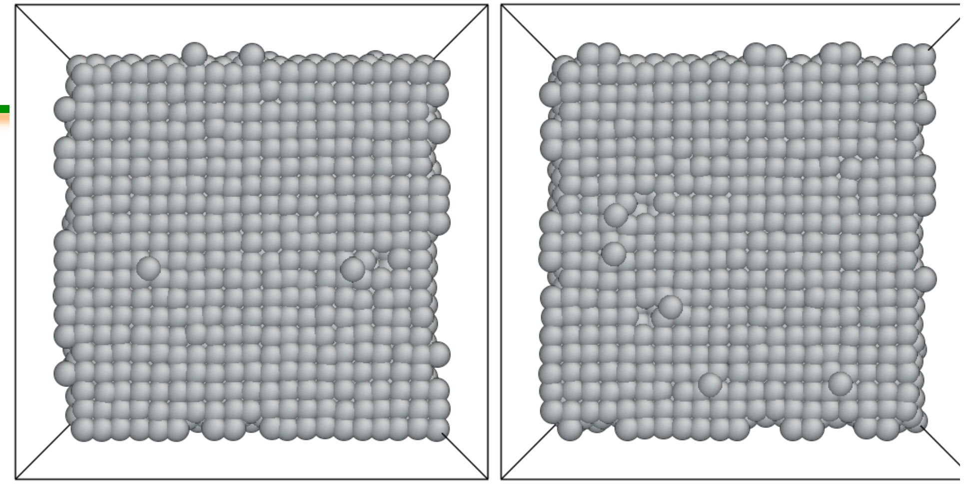
 * Initially small He clusters are mobile and grow through cluster coalescence, until reaching size of 5-8, at which trap mutation occurs

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

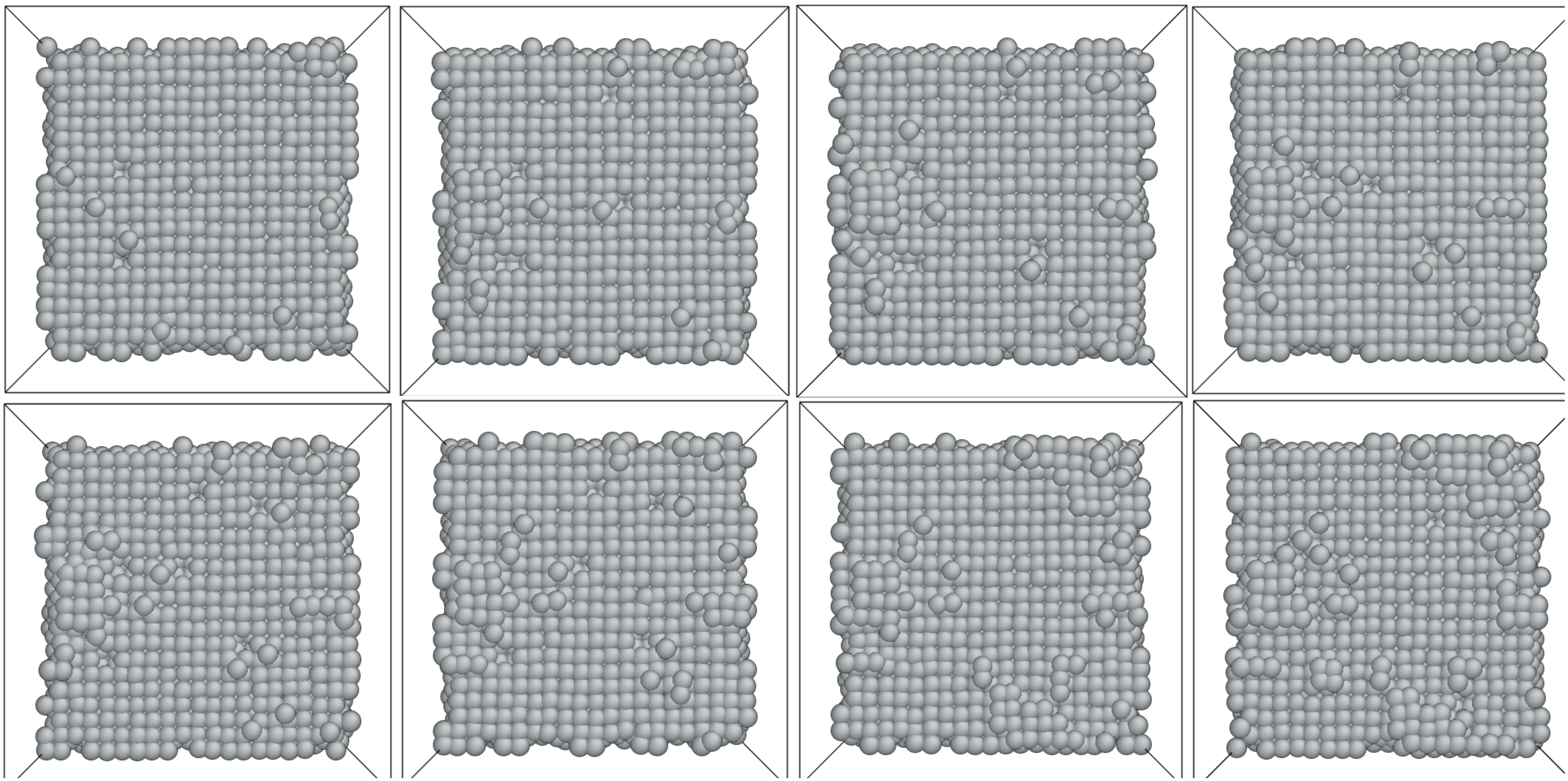


W surface evolution

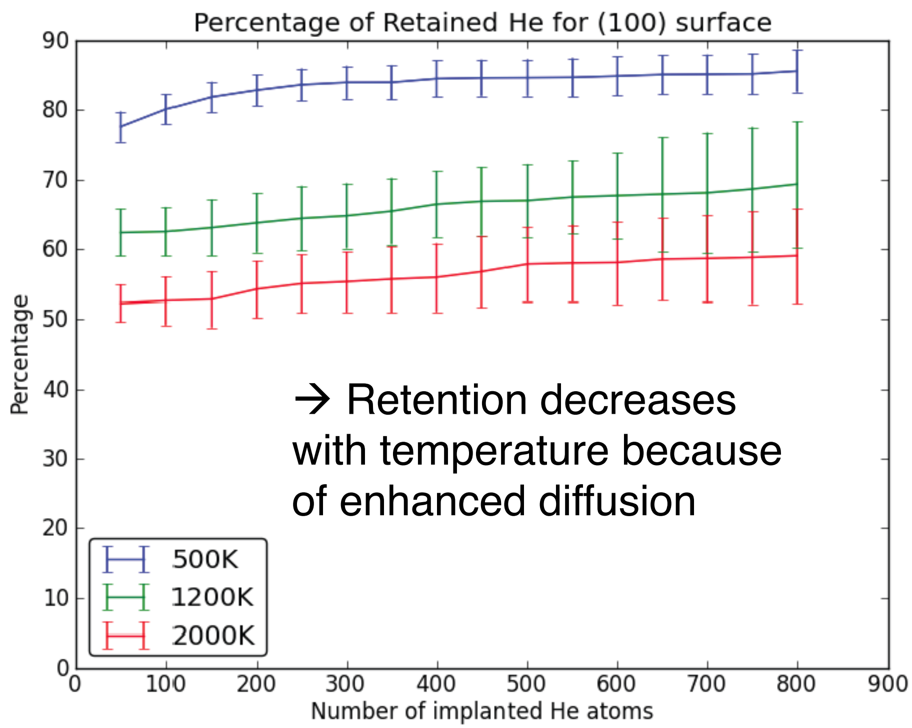
Snapshots of Tungsten Surface during He implantation as a function of time (1200K)

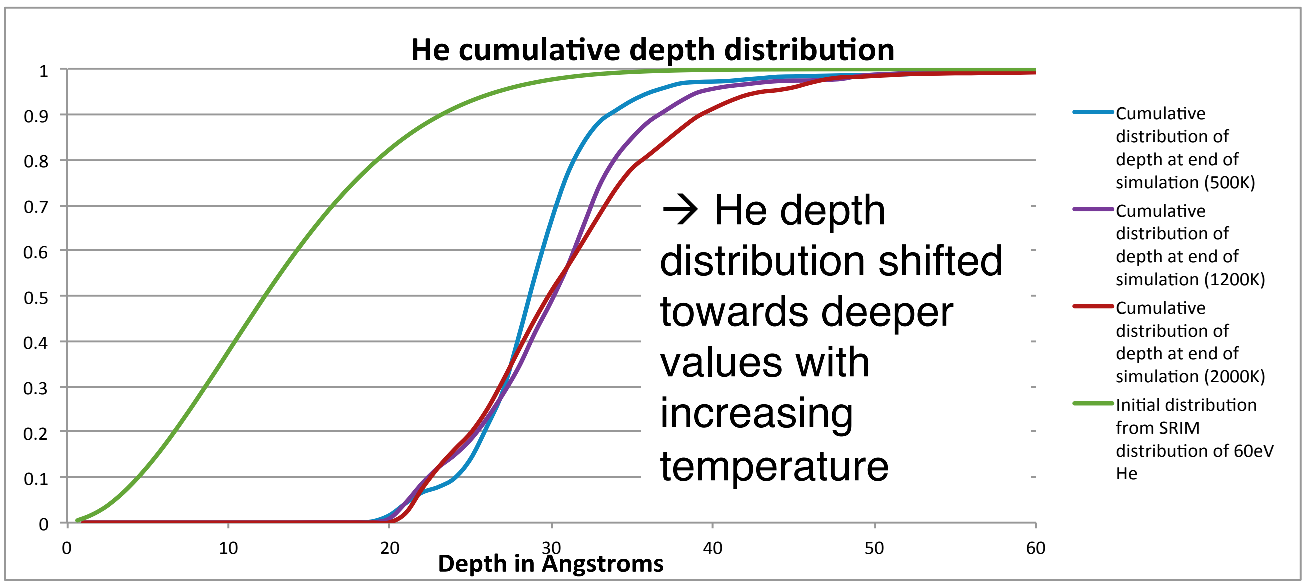


Each snapshot is after 50 extra implanted He Interstitial W atoms due to the He clustering migrate to the surface and accumulate in islands.

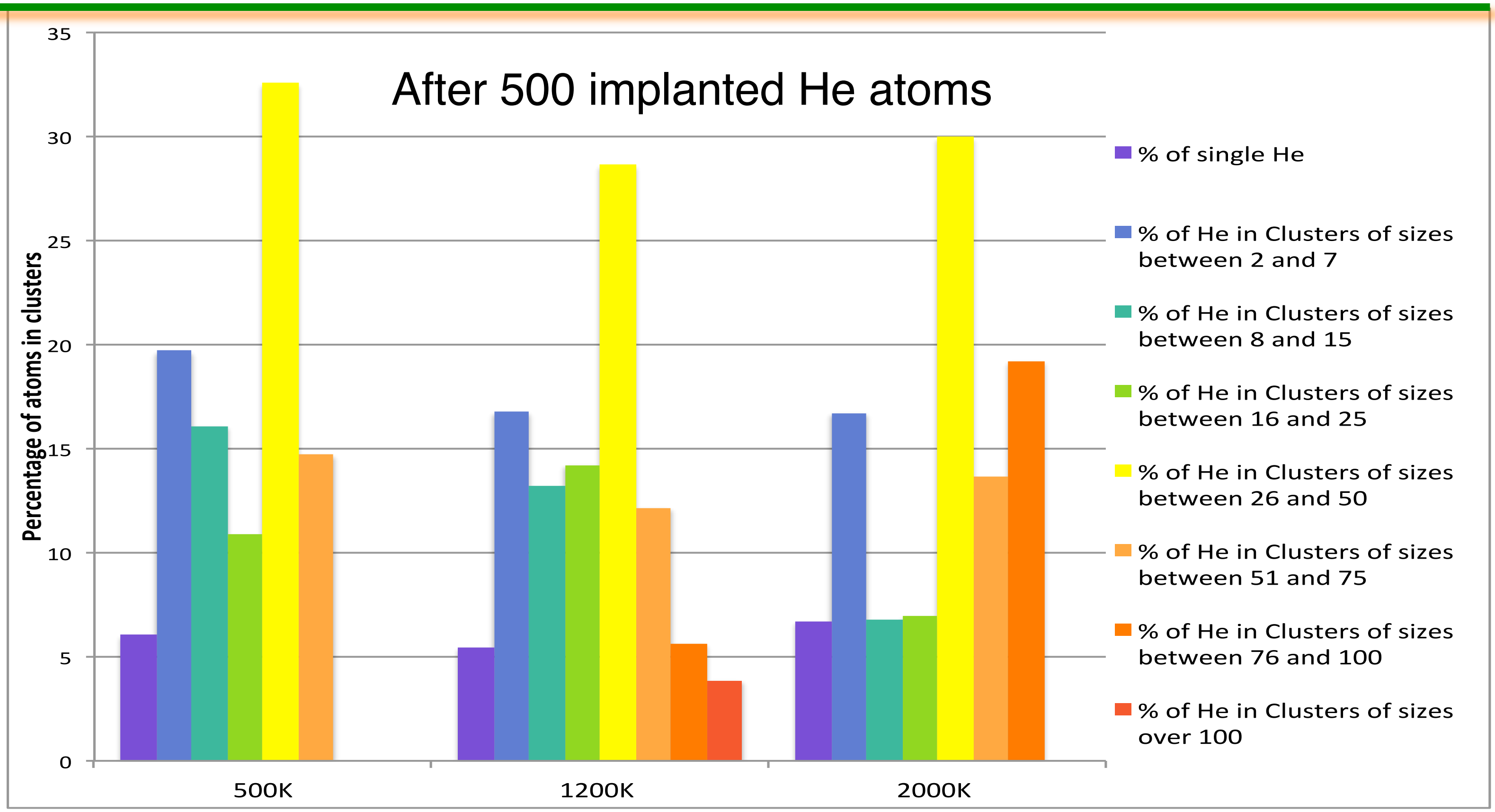


He retention & depth distribution





He cluster distributions



1. All three temperatures have about the same percentage of clusters smaller then 8 He atoms.
2. The higher the temperature, more diffusion promotes more cluster growth:

   → at 500K even though retention is the highest, the clusters aren't very big (no clusters above 75 atoms)

   → On the contrary, at 2000K diffusion is fastest and so there are very few medium sized clusters (between 8 and 25) and much more big clusters (over 50) then at 500K or 1200K

The higher the temperature the smaller the retention:

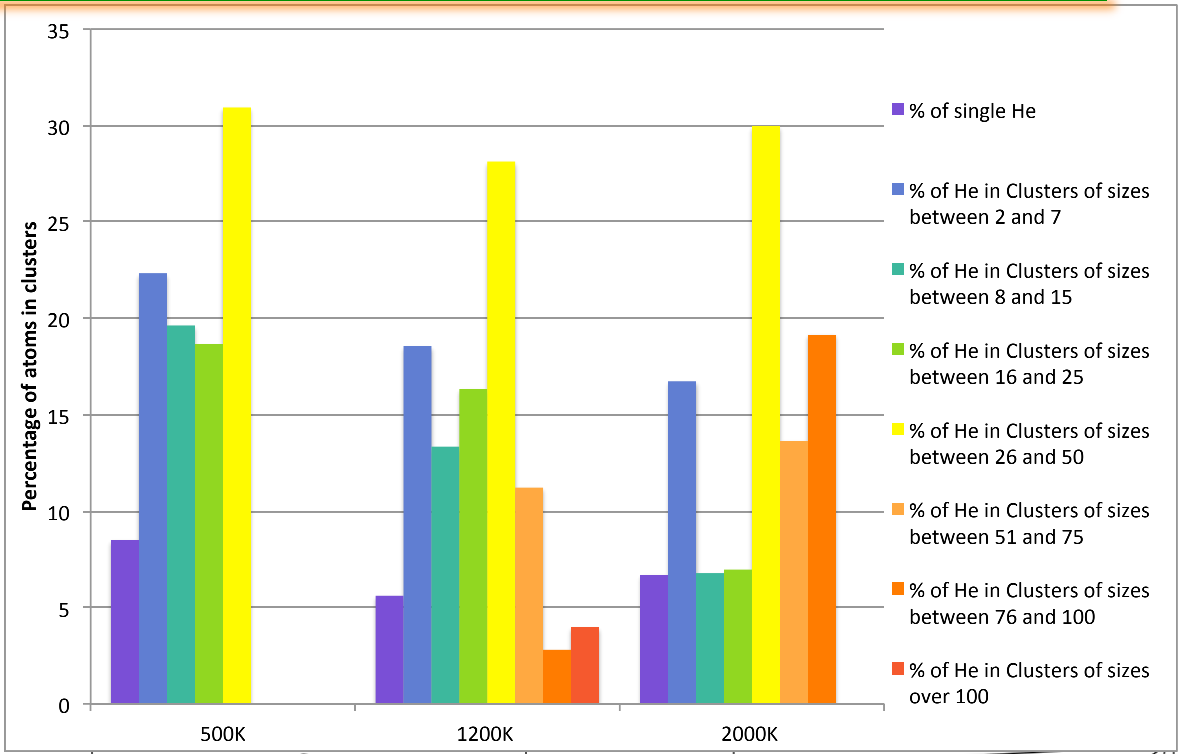
→ Even though at 2000K there are more big clusters then at 1200K, it's at 1200K that we find the biggest clusters (above 100 He atoms)

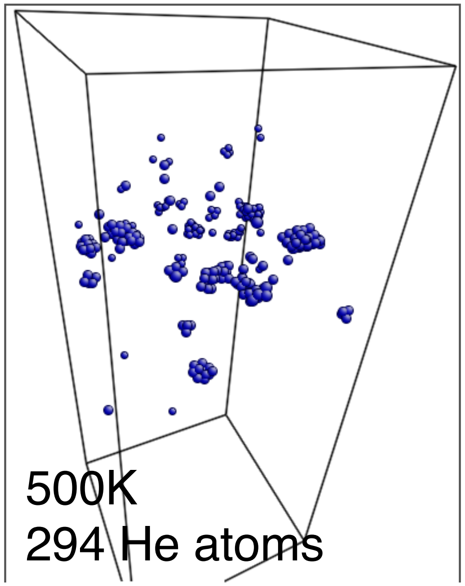
Comparison of He cluster evolution

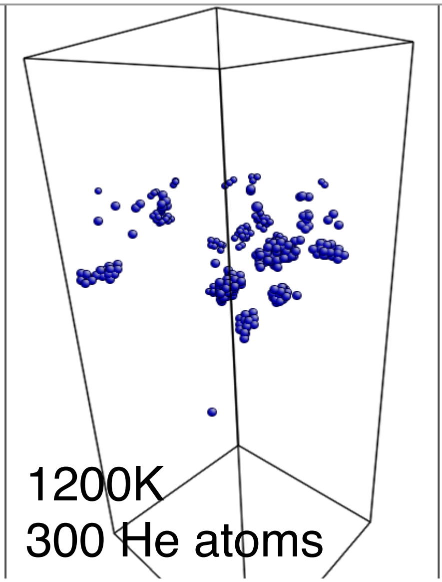
At 500K after 350 implantations (294 He remain)

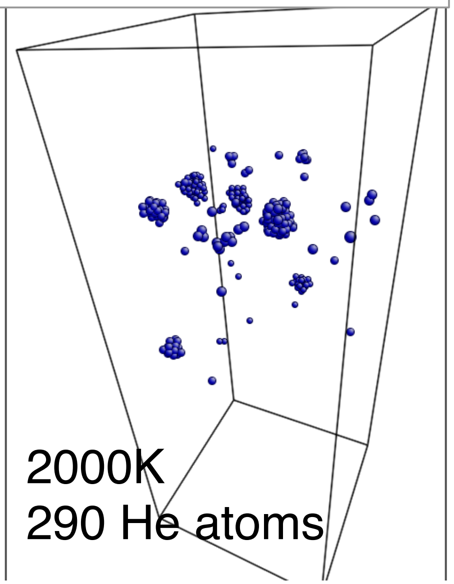
At 1200K after 450 implantations (300 He remain)

At 2000K after 500 implantations (290 He remain)





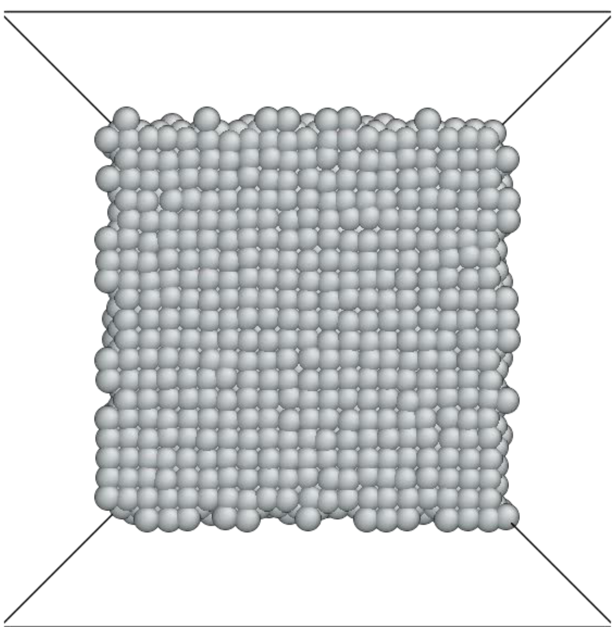


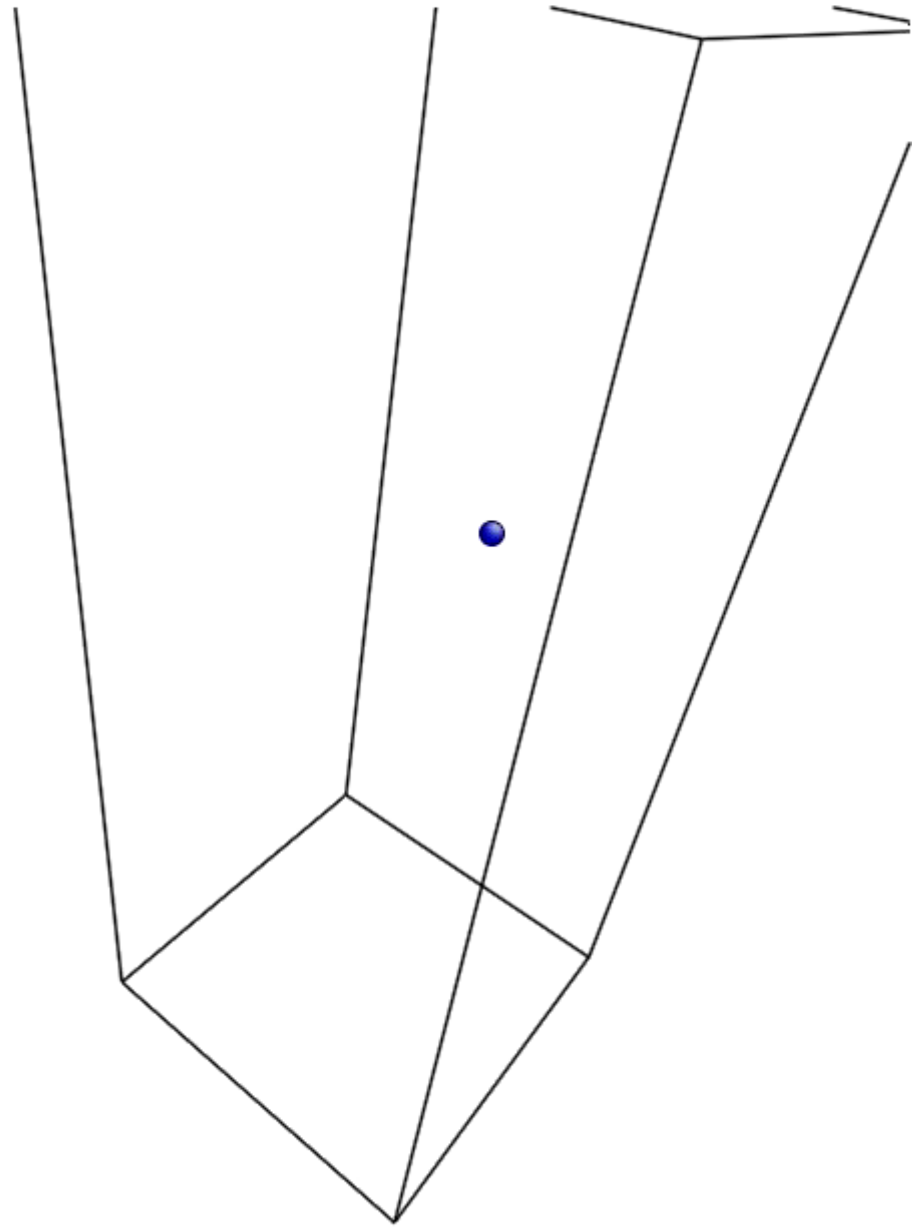


Early stage He bubble evolution below crater

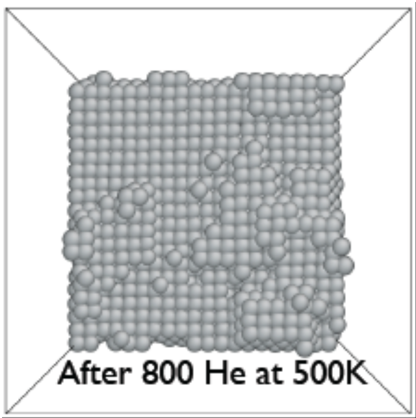
- Similar He clustering behavior below a crater/burst bubble surface, but with reduced He retention

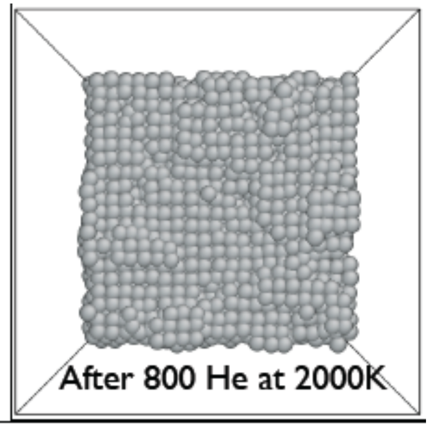
- Additional three dimensional evolution of surface adatoms around crater



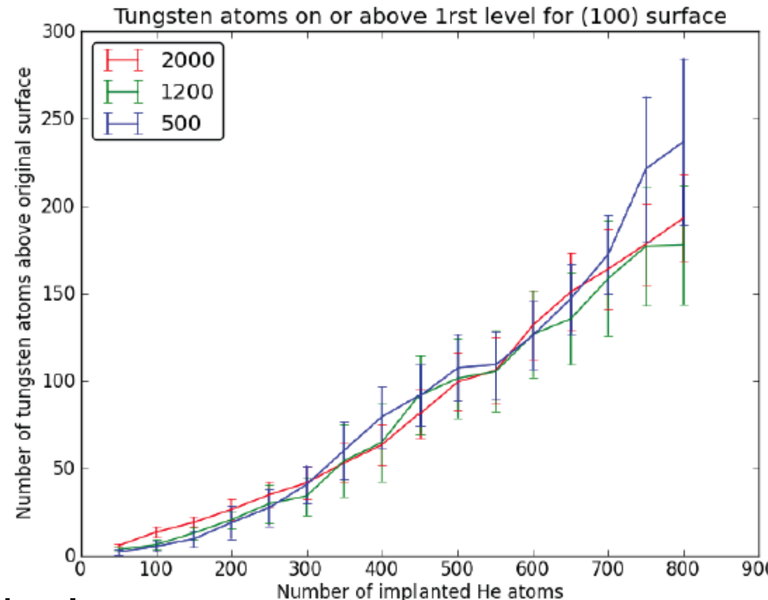


W adatom formation & surface roughness

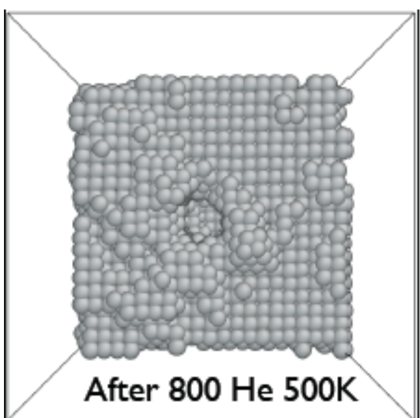


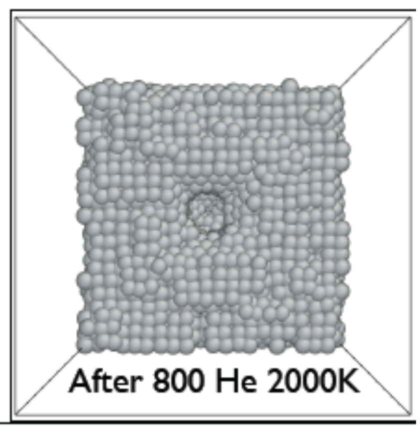


Number of W atoms on or above the initial level is about the same for all three temperatures
The higher T, the less ordered the surface

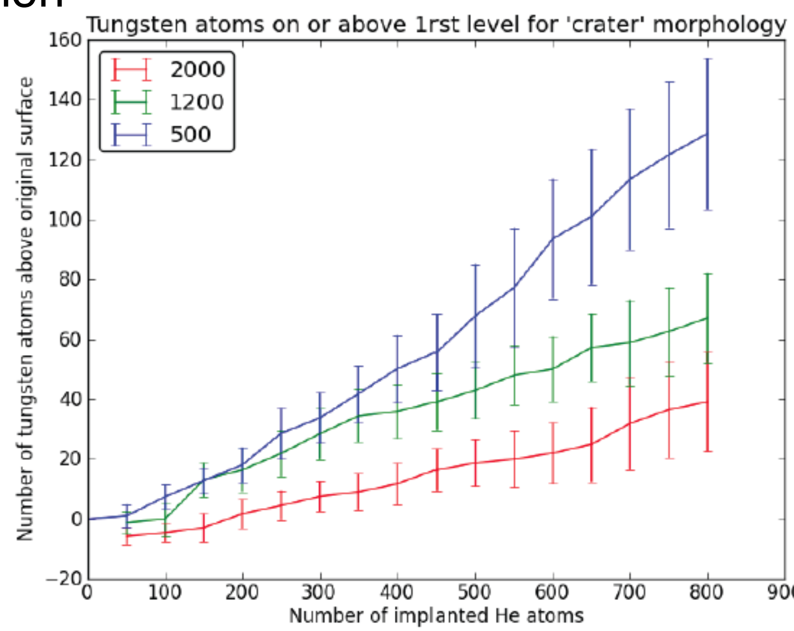


Mechanism of W adatom formation, He 'trap mutation'

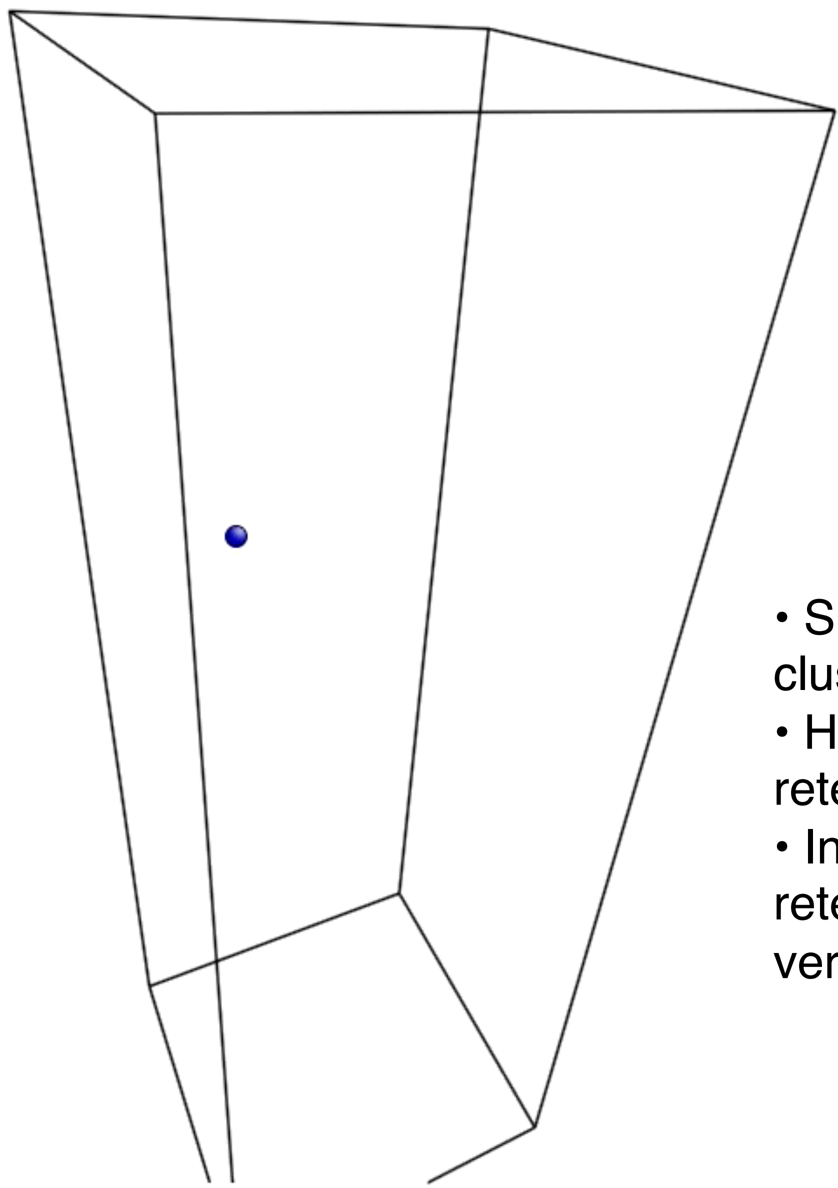


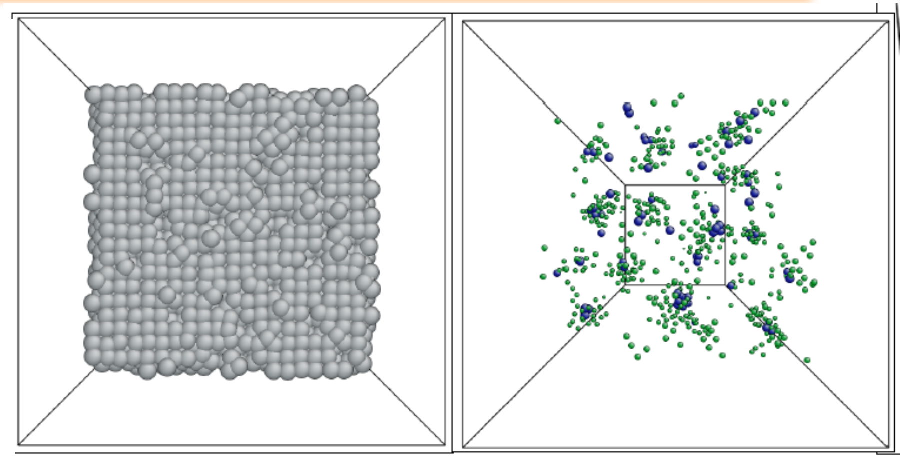


Overall: a lot less pilling up then with the (100) surface
Temperature dependence: at lower T, there's more total W pilling up at surface

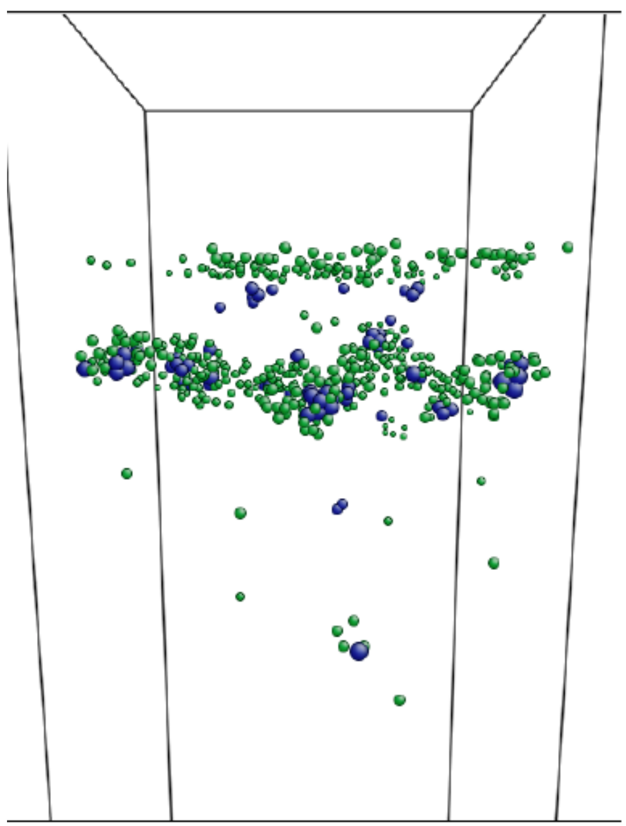


Preliminary simulation with 90%H, 10%He at 1200K





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



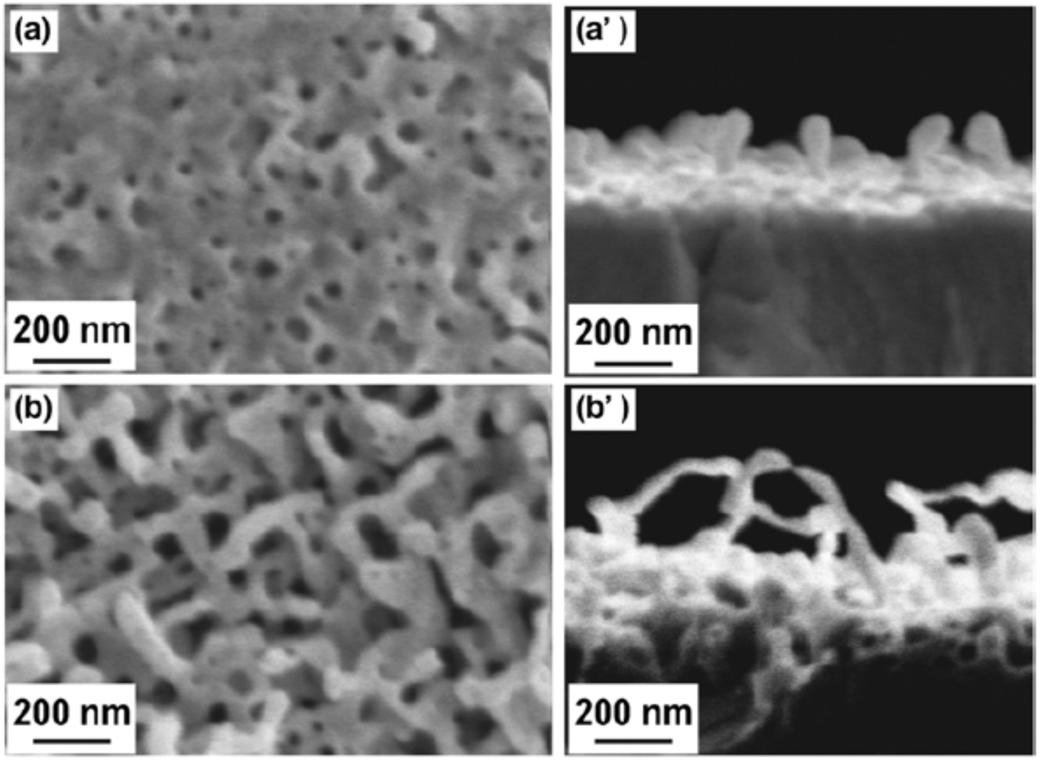
W fuzz formation mechanism(s)

• 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

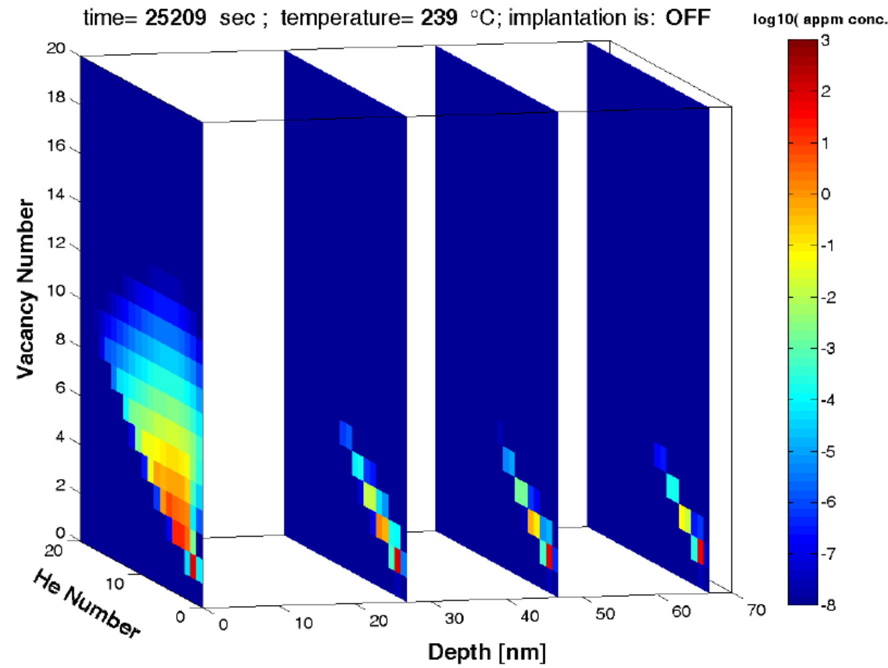
• MD simulations do not indicate any effect of sub-surface He bubbles on W sputtering yields do to He ion irradiation

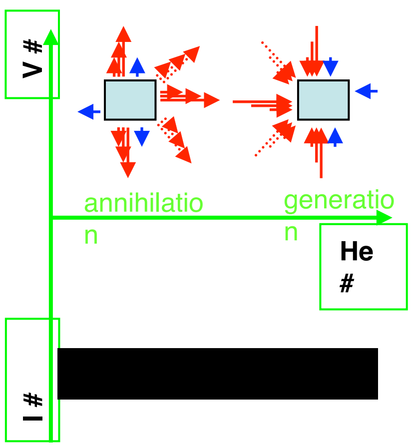


* Kajita, *Nuclear Fusion* **49** (2009) 095005.

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)

$$\frac{\partial[He_i]}{\partial t} = D\frac{\partial^2[He_i]}{\partial x^2} + \mathrm{dissoc.\_rate}(He_mV_n) + \mathrm{He\_kickout\_rate}(HeV) + \mathrm{implan.\_rate} - \mathrm{self\_trap\_rate} - \mathrm{He_i\_trap\_rate}(He_mV_n) - \mathrm{annihilation\ by\ dissociation}$$

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;

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

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

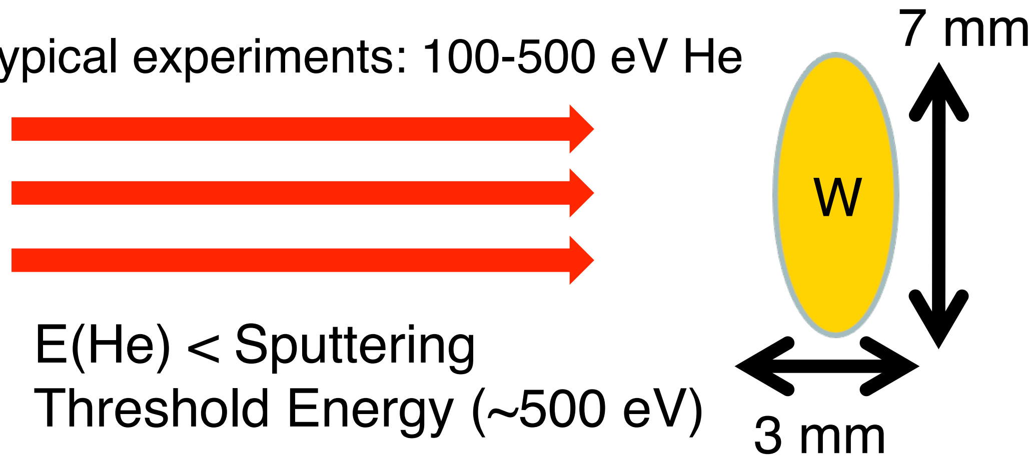
- **Spatial derivative (finite difference)**

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

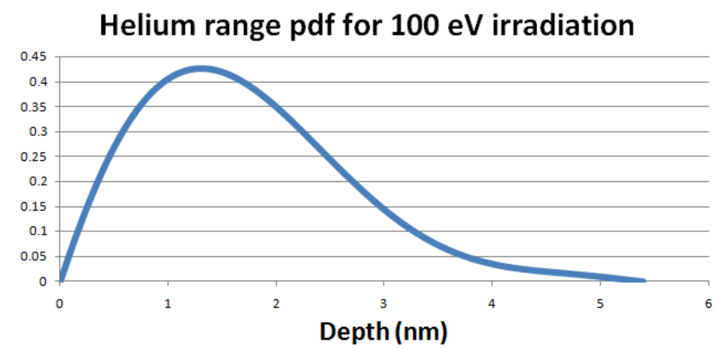
- **Parallel, large sparse-matrix linear solver (PARDISO) using open-MP formalism and backward difference time integration - easily treat systems with 10^7 degrees of freedom**

Low energy He implantation of W

Typical experiments: 100-500 eV He



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



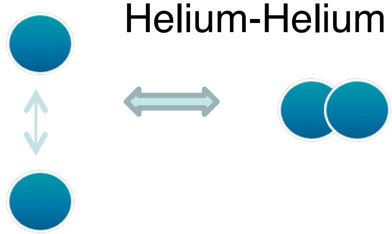
Coupled, 1-dimensional reaction-diffusion model:

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

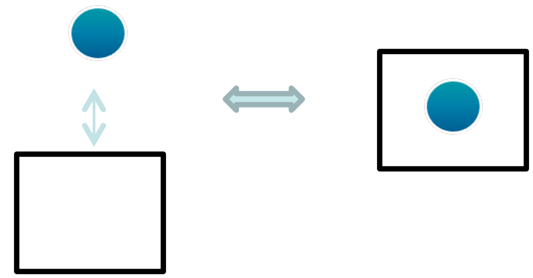
Species considered are Helium, vacancies, interstitials and their clusters, denoted by $He_x V_y I_z$

Low energy He implantation of W

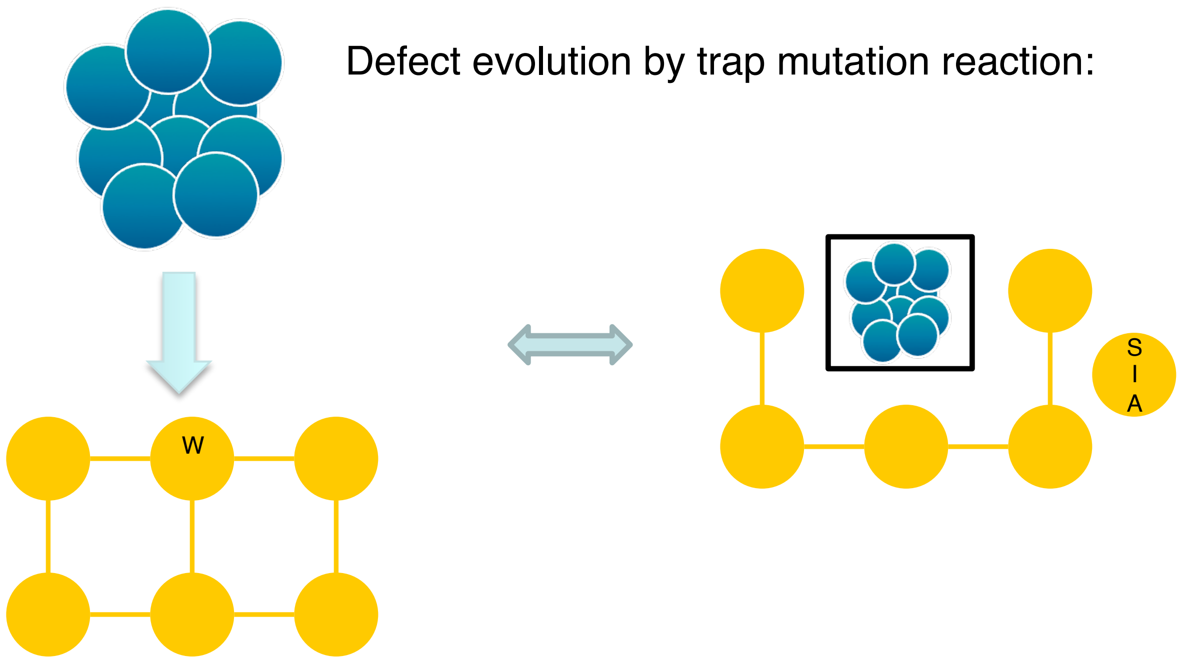
Helium-Helium Interactions



Helium – Vacancy Interactions

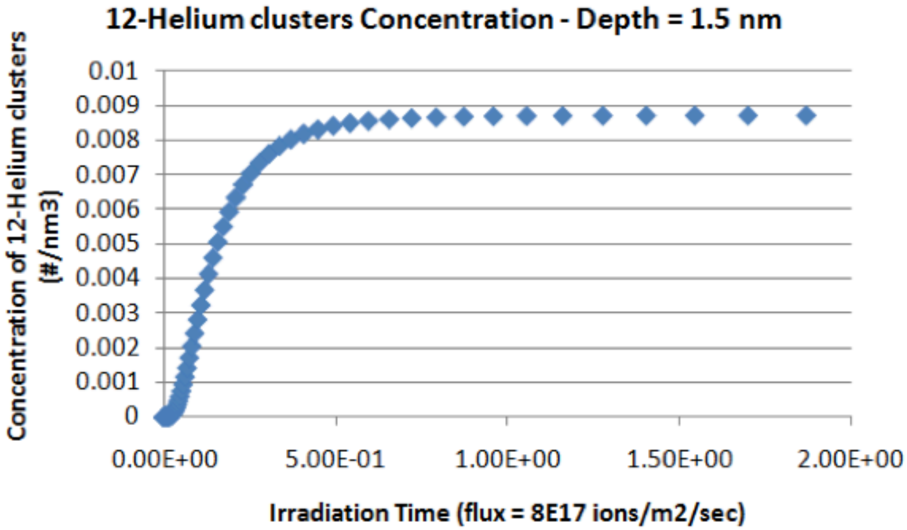


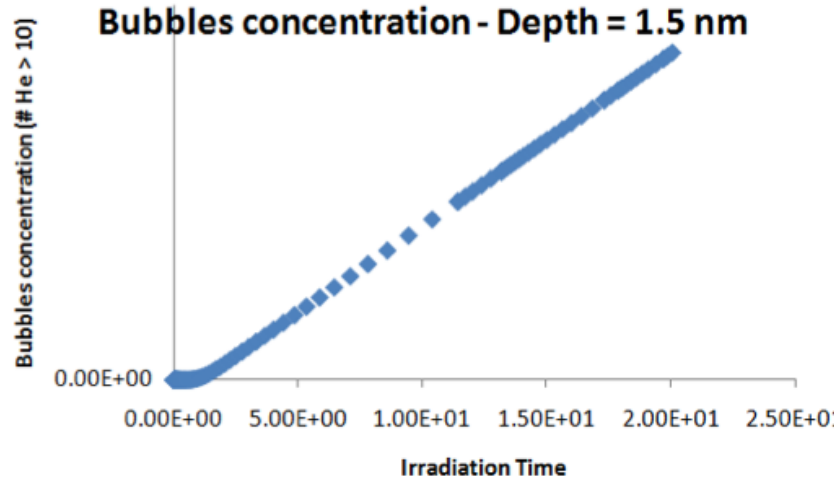
Defect evolution by trap mutation reaction:



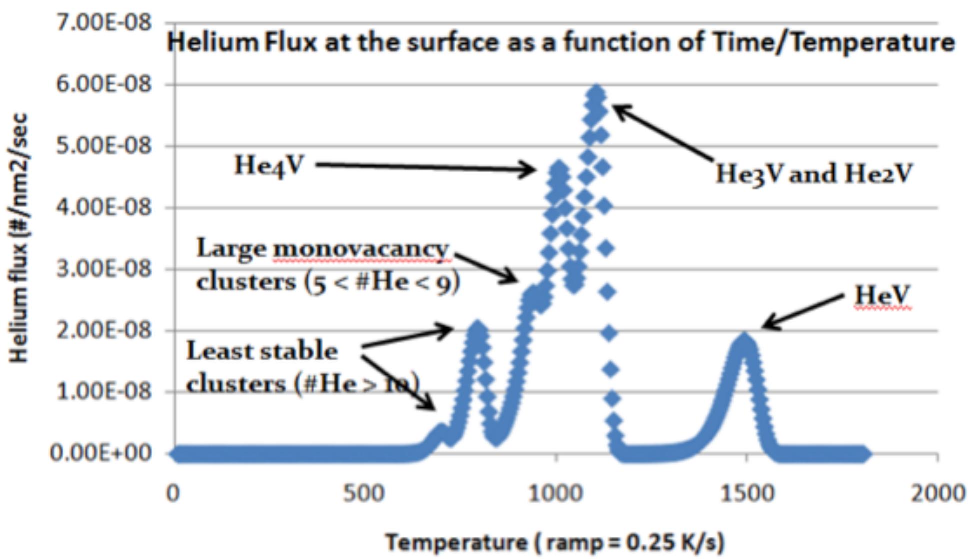
Low energy He implantation of W

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

- Fusion materials performance is an inherently multiscale challenge – significant effort ongoing to utilize multiscale materials modeling and high performance computing – *but this is in the early stages of research and implementation* – lots of effort at different scales, few (none) integrated codes using high-performance computing

- 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

- Future challenges to address the longer-time dynamics (kinetic Monte Carlo & spatially dependent cluster dynamics, or other techniques) of bubble formation and surface evolution. Must eventually extend to multi-elemental surfaces (O, Be, C, …) & couple to scrape-off-layer and edge plasma physics modeling