An approach to multiscale modeling plasma surface interactions in tungsten, with a framework for uncertainty quantification

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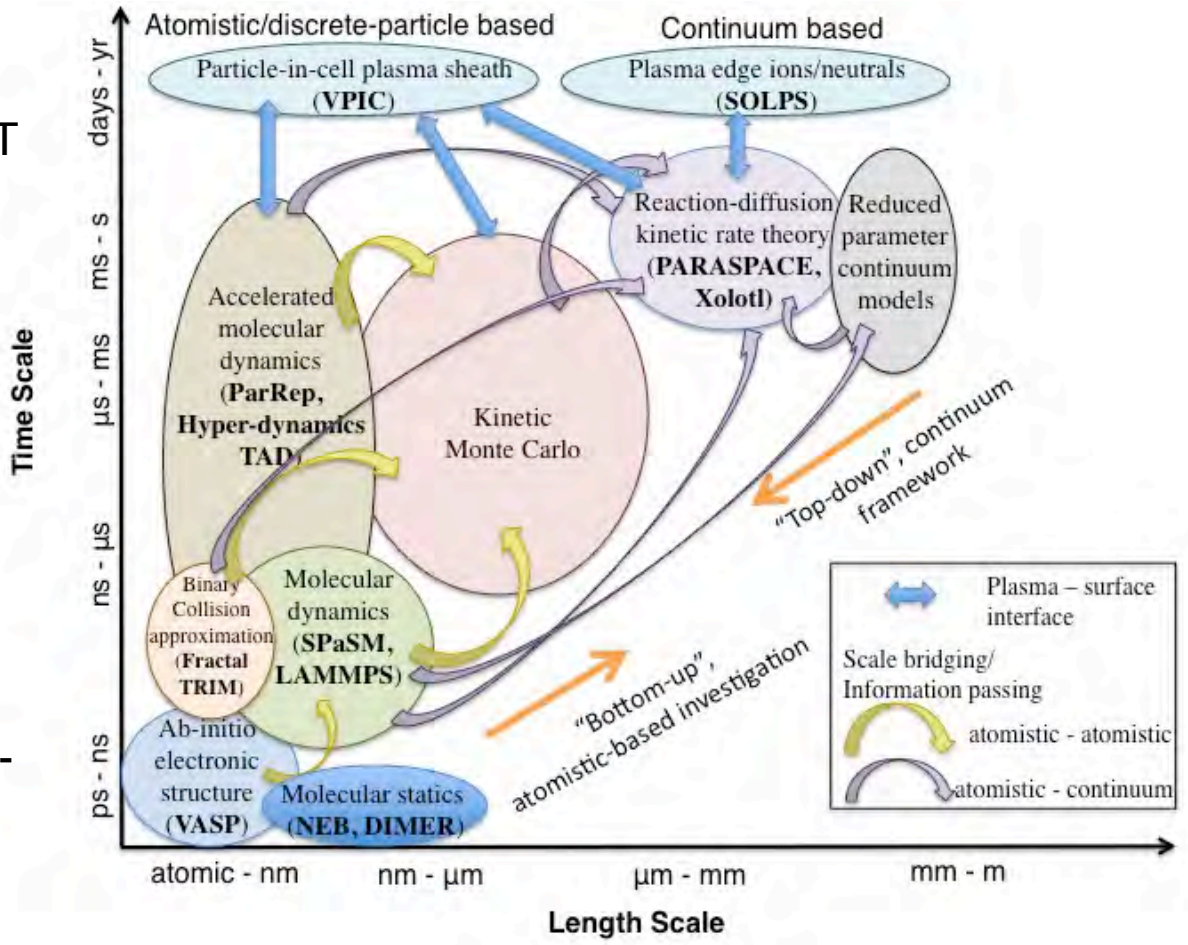
*Today's talk: Update on multiscale integration**

Goal: Discovery science to identify mechanisms/clues to W nanofuzz formation and synergies between He & H exposure that impact H/D/T permeation & retention – and surface mass loss (dust)

Mechanisms of interest:
Sputtering (later), surface adatom formation, He diffusion, bubble formation, growth & rupture

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

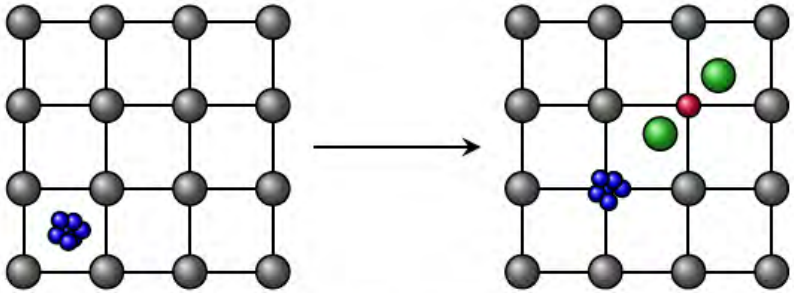


- **Outline:**
  - MD to KMC to cluster dynamics of He accumulation in W: impact of surface orientation
  - Initial results of Bayesian informed UQ assessment of He-vac thermodynamics & approach
  - Initial results investigating H trapping at sub-surface He bubbles (He-H synergies)

* BÐ Wirth, K.D. Hammond, S.I. Krashenninikov, and D. Maroudas, "Challenges and Opportunities of Modeling Plasma Surface Interactions in Tungsten using High Performance Computing, *Journal of Nuclear Materials* **463** (2015) 30.

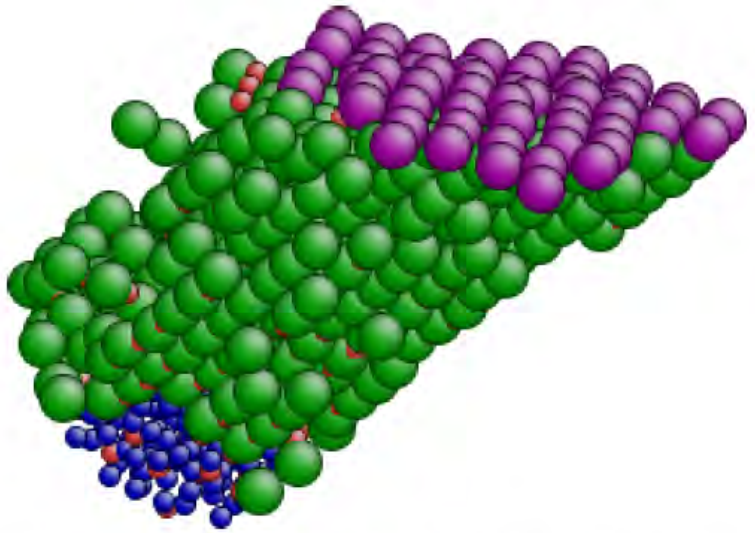
Key MD observations of early stage He bubble evolution

- **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**



"trap mutation" processes

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



"loop-punching" processes

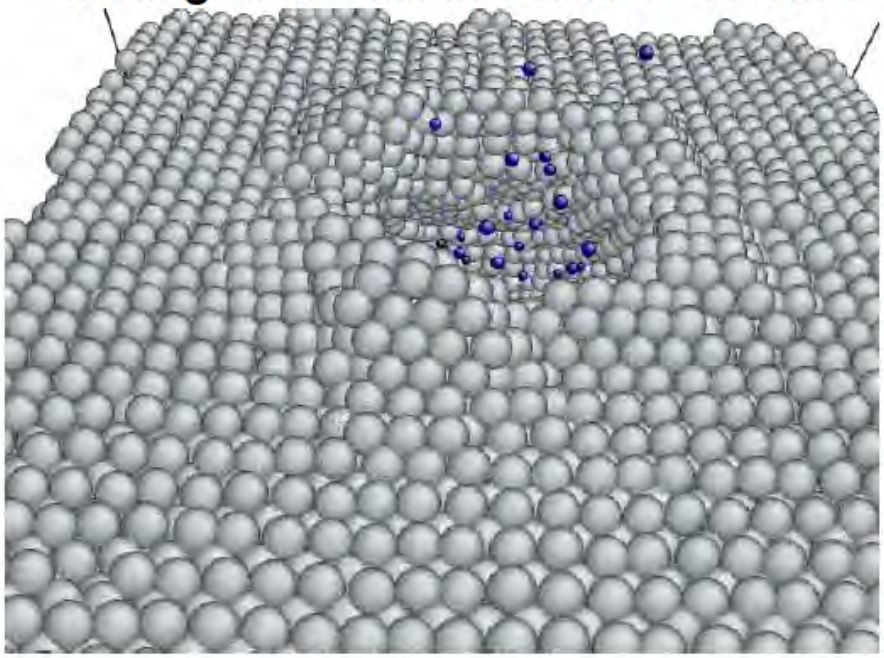
Movie available with F. Sefta, *et al. Nucl. Fusion* **53**: 073015 (2013)

- **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***

A brief word about Molecular Dynamics (MD) calculations

- "Common" MD codes: LAMMPS, SPASM
- Typically run on small clusters (usually because of throughput), especially for 'discovery' science
- Increasingly used for 10^7 atoms & beyond (provide decreased implantation rates)
- Limited by interatomic potentials and achievable timescales

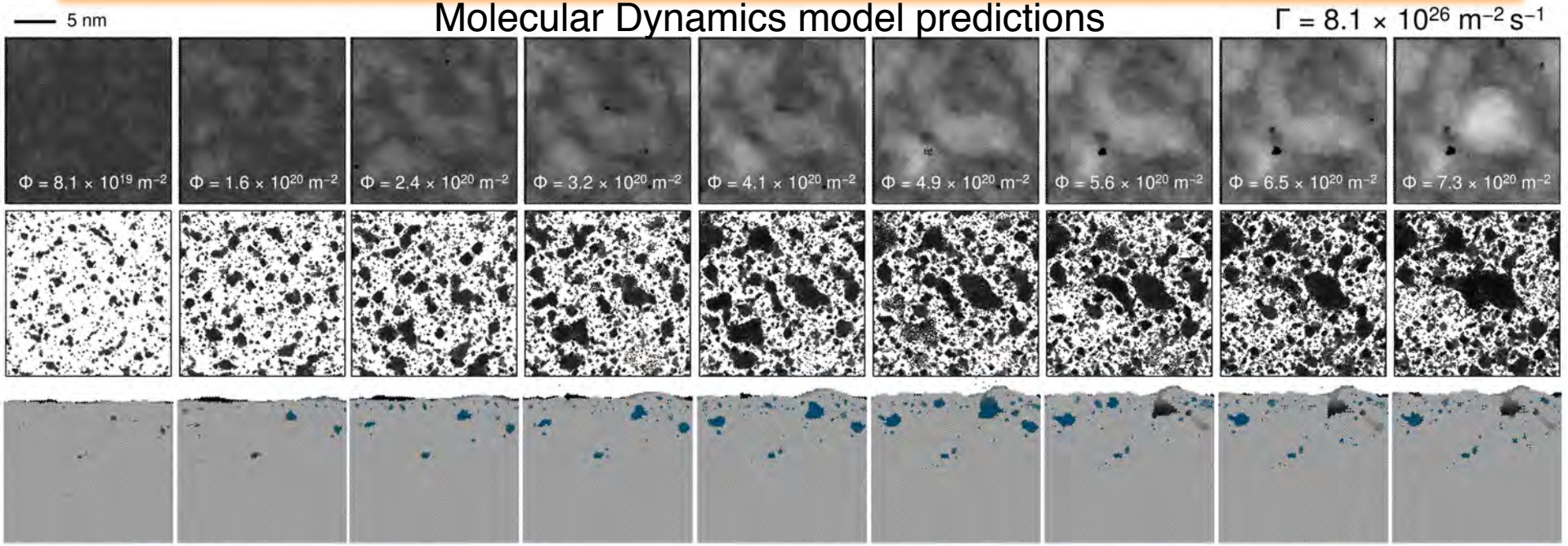
Bursting of He bubble onto W surface



The Time Scale Challenge

- 1 MD time step, $O(10^{-15}$ s) requires 1 ms (10^{-3} s) wallclock time
  - Typical for $O(2\text{x}10^7$ atoms), $O(2\text{x}10^4$ cores) on Mira (ANL)
- Simulating onset of fuzz formation (10^4 s) requires ***O*(300 M years)**
  - Completely unrealistic extrapolation to exascale: "only" ***O*(80k years)**

Tungsten surface response to low-energy He exposure



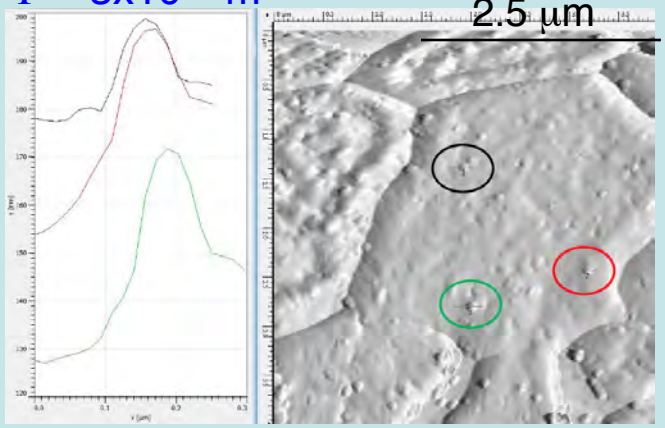
High-flux simulations showing surface growth and helium accumulation below a W(100) surface. Top: View of surface (white = +1.5 nm, black = –2 nm); Middle: helium atoms, top view (black = at surface, white = –15 nm); Bottom: cross-section.

- 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})

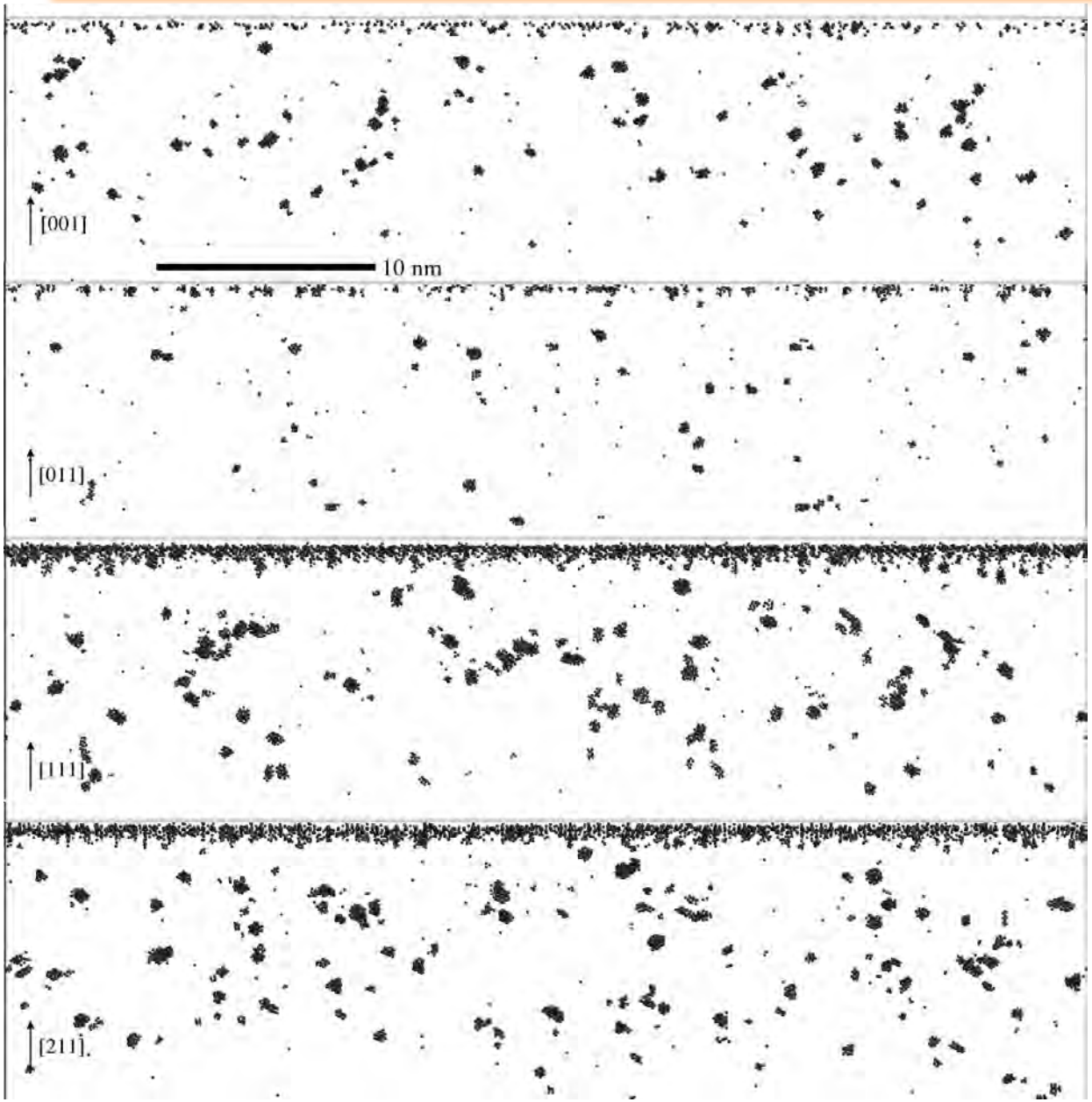
Experiment: $\Gamma = 3\times10^{19}$ $m^{-2}s^{-1}$
$\Phi = 3\times10^{24}$ m^{-2}



* Hammond & Wirth, UTK/ORNL

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

*Impact of surface orientation**



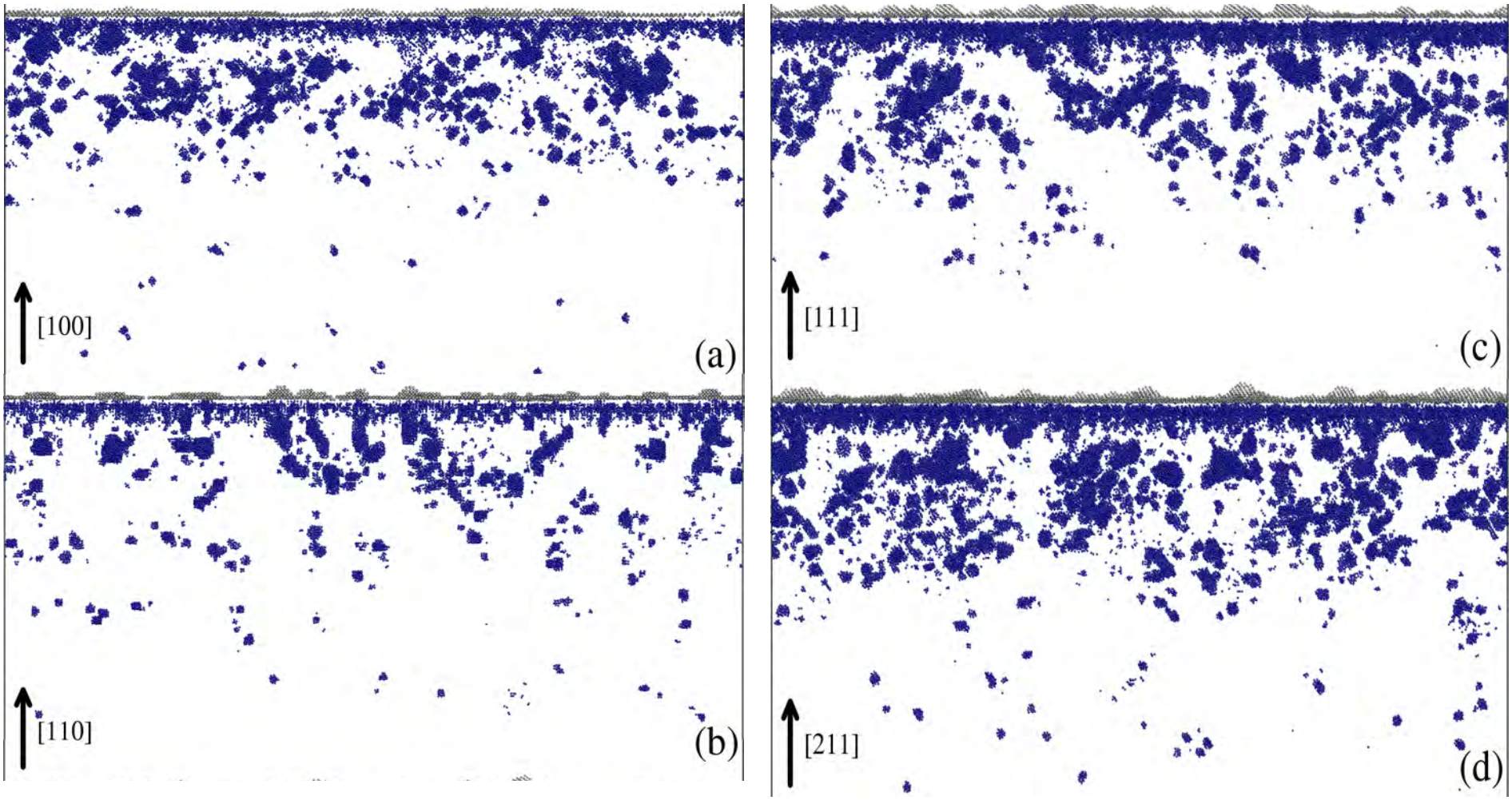
Helium distributions at a fluence of 10^{19} He-m^{-2}

Nominal Flux:
4.0×10^{25} He-m^{-2} s^{-1} of 100 eV He (thermally implanted)
Temperature: 933K

Note presence of concentrated He layer in (111) and (211) cases – surface orientation strongly influences helium retention

* Hammond and Wirth, *JAP* **116** (2014) 143301

*Impact of surface orientation**



Helium distributions at a fluence of 3.3×10^{19} He-m^{-2}

Concentrated near-surface He layer also develops in (100) and (110) surfaces

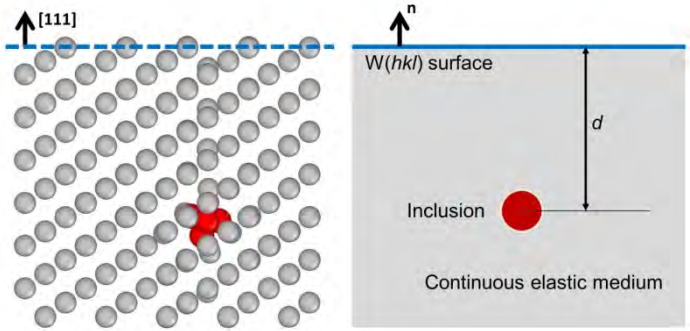
*Hammond (UM), manuscript in preparation

*Interactions of small mobile He clusters with surfaces**

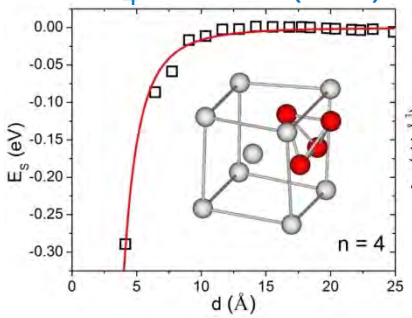
- Small mobile He clusters, from aggregation of implanted helium in tungsten, migrate to the surface by Fickian diffusion and drift due to a thermodynamic driving force for surface segregation originating from the elastic interaction between the cluster and the surface.

$$E_s(d) = -\frac{A_{s,n}}{d^3}$$

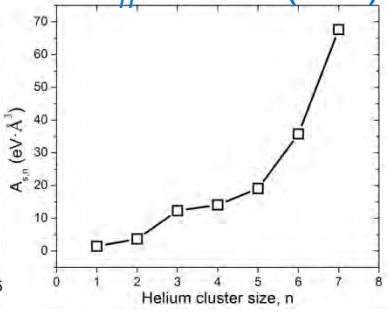
$A_{s,n}$ increases with increasing cluster size n



He_4 near W(211)

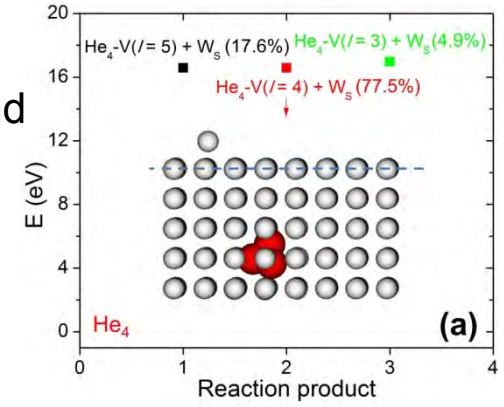


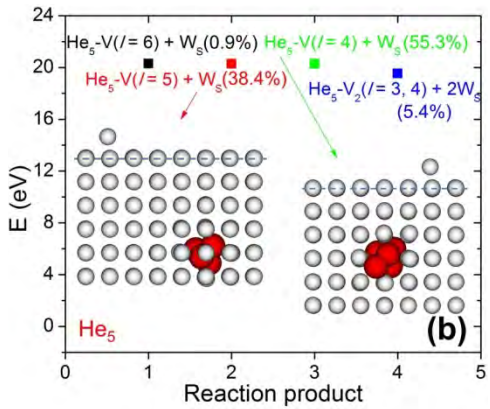
He_n near W(211)

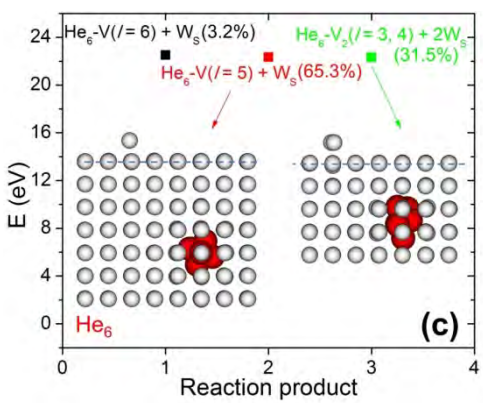


- As the clusters approach the surface, cluster reactions are activated with rates much higher than those in the bulk. The dominant ones are trap mutation (TM) reactions, generating immobile helium-vacancy complexes a few layers below the surface plane and tungsten surface adatoms: $W + He_n \rightarrow He_n\text{-}V_k + k\,W_s$; $k \geq 1$

Examples: He_4, He_5, and He_6 near W(110)







* Hu, Hammond, Wirth, and Maroudas, *J. Appl. Phys.* **115**, 173512 (2014);
Maroudas, Blondel, Hu, Hammond, and Wirth, *J. Phys.*: *Condens. Matter*, subm. (2015).

*Modified trap mutation near surfaces – orientation dependent**

Modified trap mutation (typically happens around He_7 in bulk) influences retention, He depth profile and bubble distributions

Sink	He_n (n = 1)	He_n (n = 2)	He_n (n = 3)	He_n (n = 4)	He_n (n = 5)	He_n (n = 6)	He_n (n = 7)
W(100)	D (100%)	D (19.1%) PD (5.9%) TM (75.0%) 1 W_V	D (1.1%) PD (11.6%) TM (87.3%) 1 W_V	D (2.1%) PD (74.6%) TM (23.3%) 2 W_V	D (4.1%) PD (85.3%) TM (10.6%) 2 W_V	D (2.3%) PD (36.9%) TM (60.8%) 3 W_V	D (3.1%) PD (27.4%) TM (69.5%) 4 W_V
W(110)	D (100%)	D (31.6%) PD (1.3%) TM (67.1%) 1 W_V	D (0.0%) PD (2.0%) TM (98.0%) 1 W_V	D (0.0%) PD (0.0%) TM (100%) 1 W_V	D (0.0%) PD (0.0%) TM (100%) 2 W_V	D (0.0%) PD (0.0%) TM (100%) 2 W_V	D (0.0%) PD (0.0%) TM (100%) 2 W_V
W(111)	D (35.4%) TM (64.6%) 1 W_V	D (1.2%) PD (0.0%) TM (98.8%) 1 W_V	D (1.6%) PD (0.0%) TM (98.4%) 1 W_V	D (0.0%) PD (0.0%) TM (100%) 2 W_V	D (0.0%) PD (0.0%) TM (100%) 2 W_V	D (0.0%) PD (0.0%) TM (100%) 3 W_V	D (0.0%) PD (0.0%) TM (100%) 3 W_V

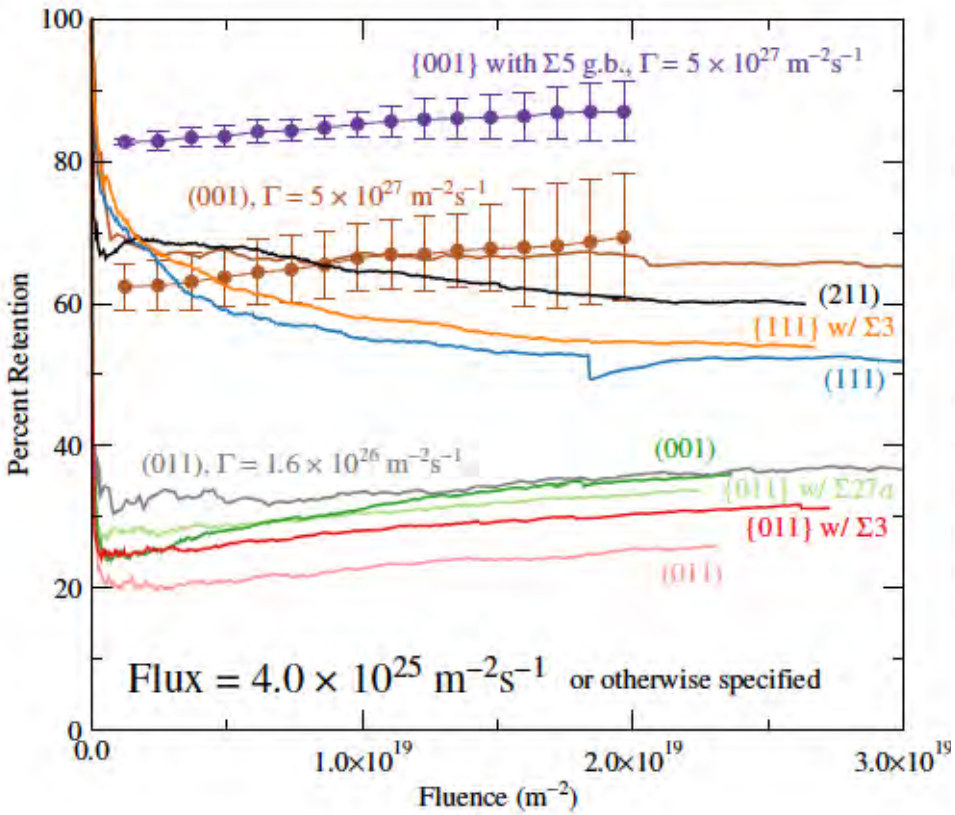
D: He Desorption
PD: Partial Dissociation
TM: Trap Mutation
W_V: Tungsten Vacancy

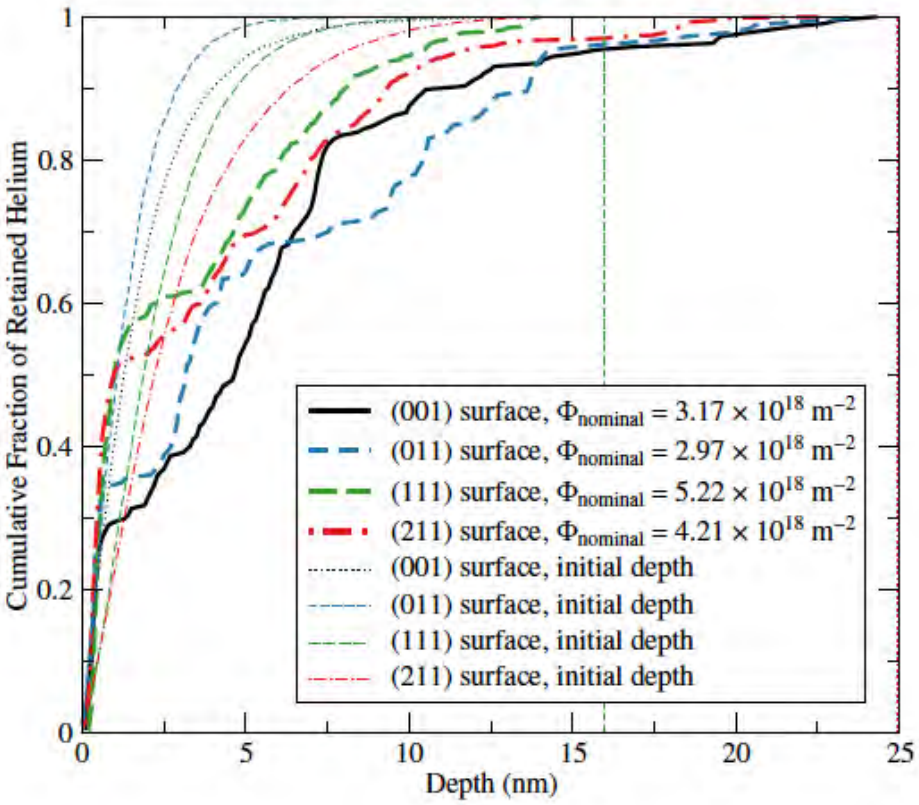
For reactions of He_n clusters with n = 1, 2, and 3 see:

* Hu, Hammond, Wirth, and Maroudas, *Surf. Sci.* **626** (2014) L21-25.

Impact of surface orientation & He flux on retained He

Temperature is 933K



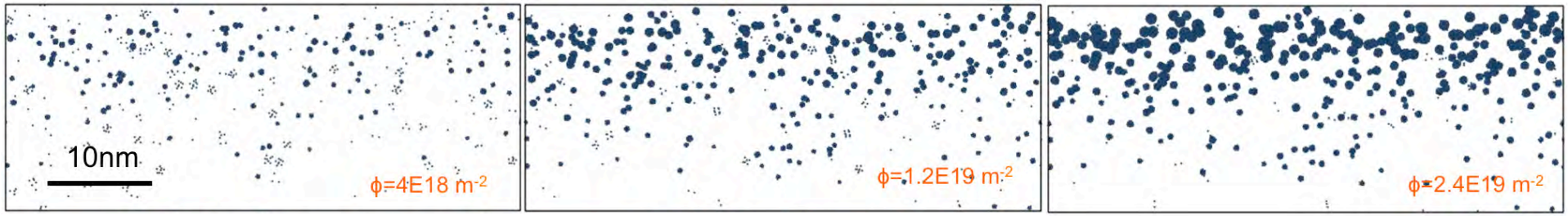


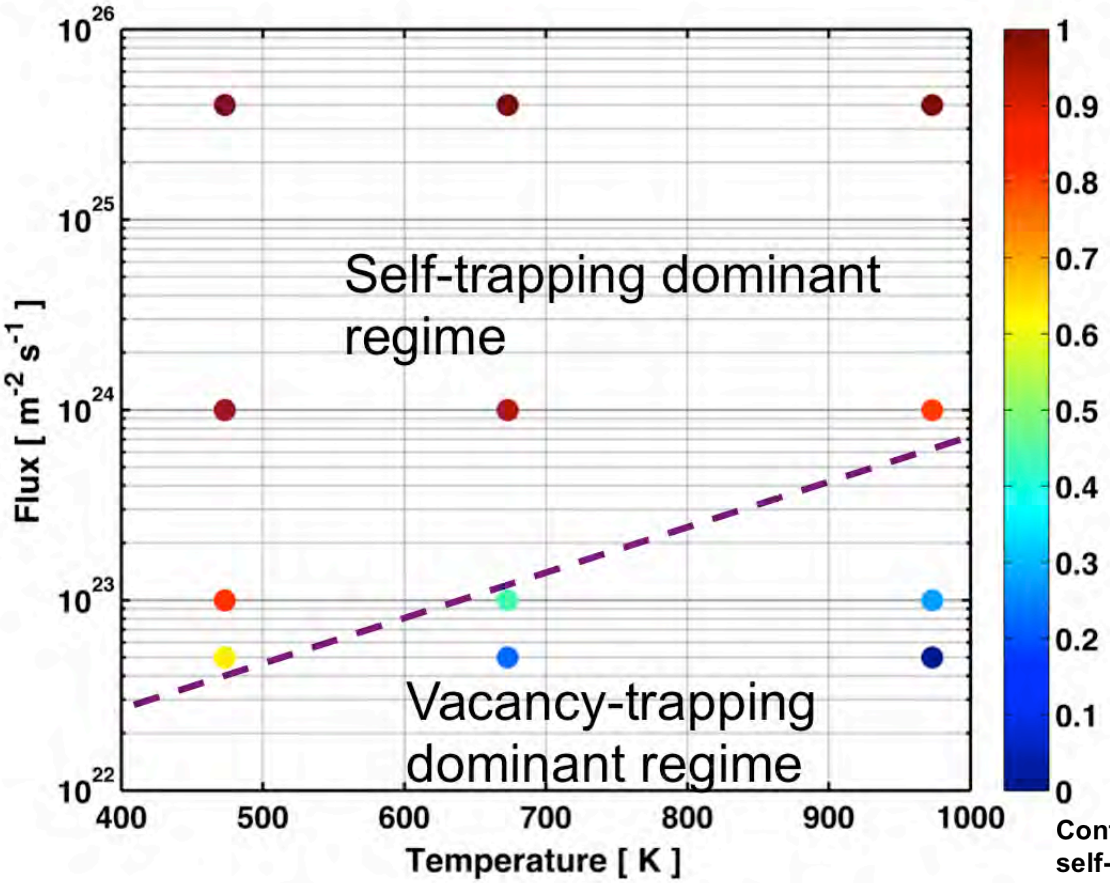
Thin lines: as-implanted.
Thick lines: at that instant.

K. D. Hammond and B. D. Wirth. *J. Appl. Phys.* **116**: 143301 (2014)

KMC simulation of He clustering below W surfaces

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





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).

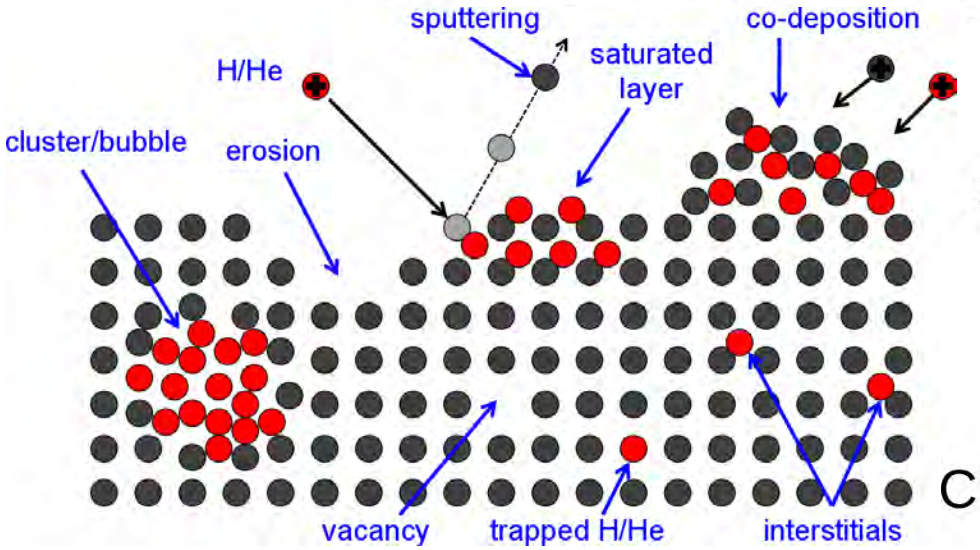
Indicate mechanism boundary of gas bubble nucleation mechanism f(Γ,T)

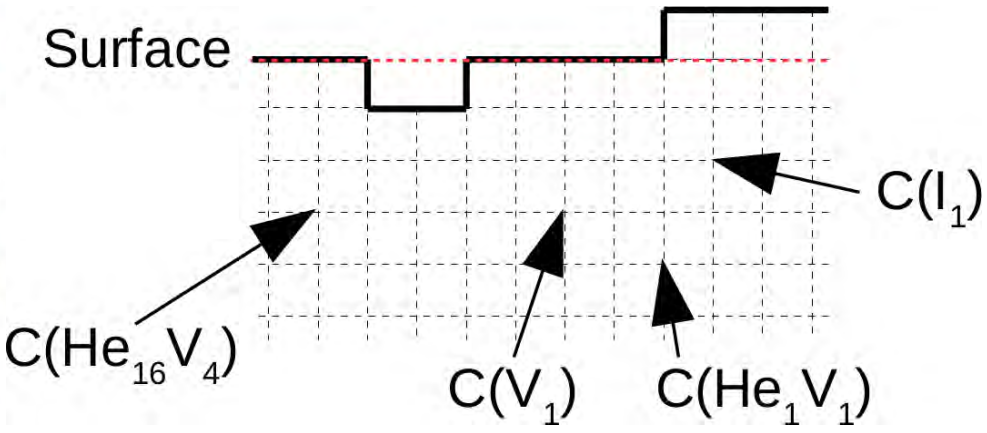
$$\text{Contribution of self-trapping} = \frac{\text{He retention rate with 0 appm vacancy}}{\text{He retention rate with 5 appm vacancy}}$$

*Xolotl-PSI**

- Xolotl (SHO-lottle) is the Aztec god of lightning and death
- Developed from 'scratch' for the SciDAC project, designed for HPC (current and emerging architechtures – multicore, multicore+accelerator) to solve advection – reaction – diffusion cluster dynamics problems within spatially-resolved continuum domain (C++ with MPI and independent modules for physics, solvers and data management)
- 2D and 3D recently implemented
- Model considers continuum concentration of He, vacancies, interstitials and mixed clusters at spatial grid points, solving the coupled advection-reaction-diffusion equations

$$\delta_t \bar{C} = \phi \cdot \rho + D\nabla^2 \bar{C} - \nabla \bar{\nu} C - \bar{Q}(\bar{C})$$





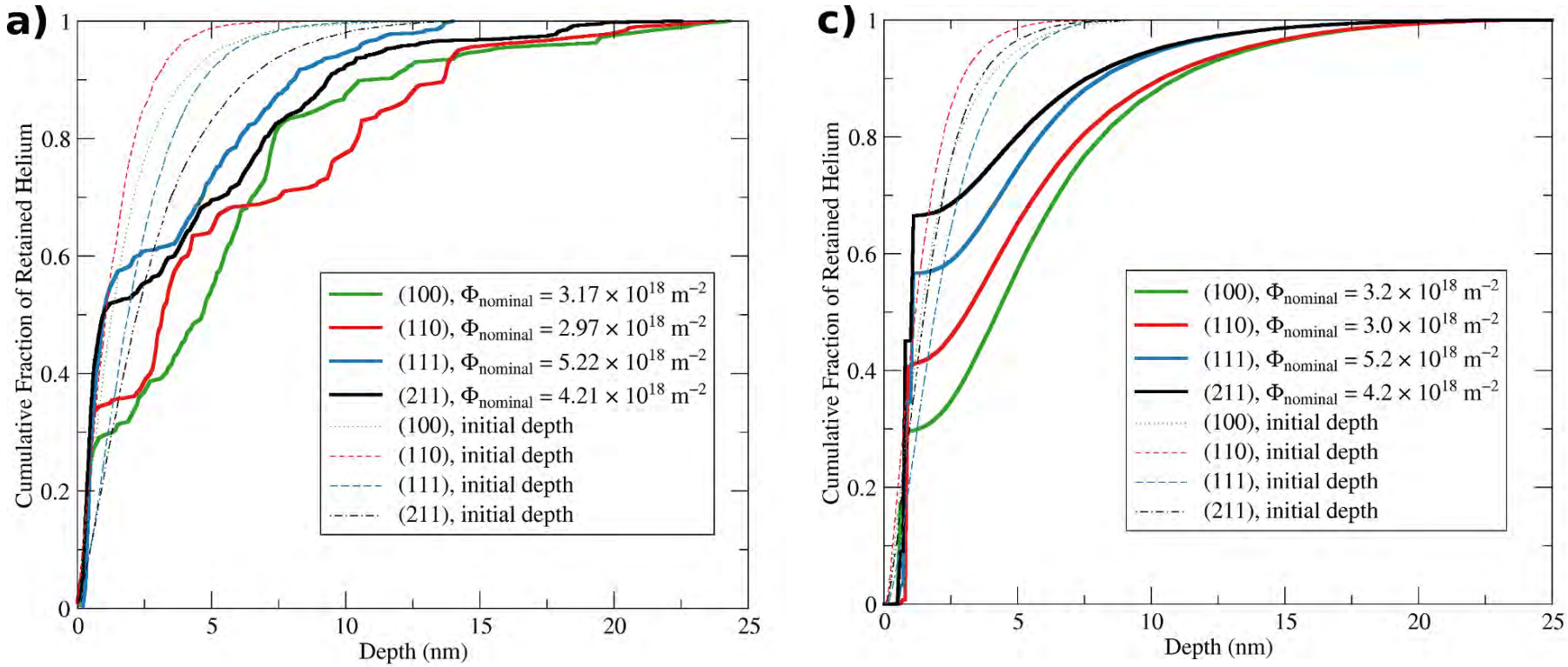
* Available at http://sourceforge.net/projects/xolotl-psi/

Initial results including advection (drift diffusion) & modified trap mutation

$$\frac{\partial C_i}{\partial t} = -\frac{3 D_i A_i}{k_B T}\left(\frac{C_i(x)}{x^4} - \frac{C_i(x+h_x)}{(x+h_x)^4}\right)\frac{1}{h_x}$$

• Also include modification of He_x -> $He_xV_1 + I_1$ in which x depends on proximity to surface (parameterized based on MD simulation probability tables)

MD simulations Γ_{He} **~ 4E25 m^{-2}s^{-1}** **Xolotl simulations**

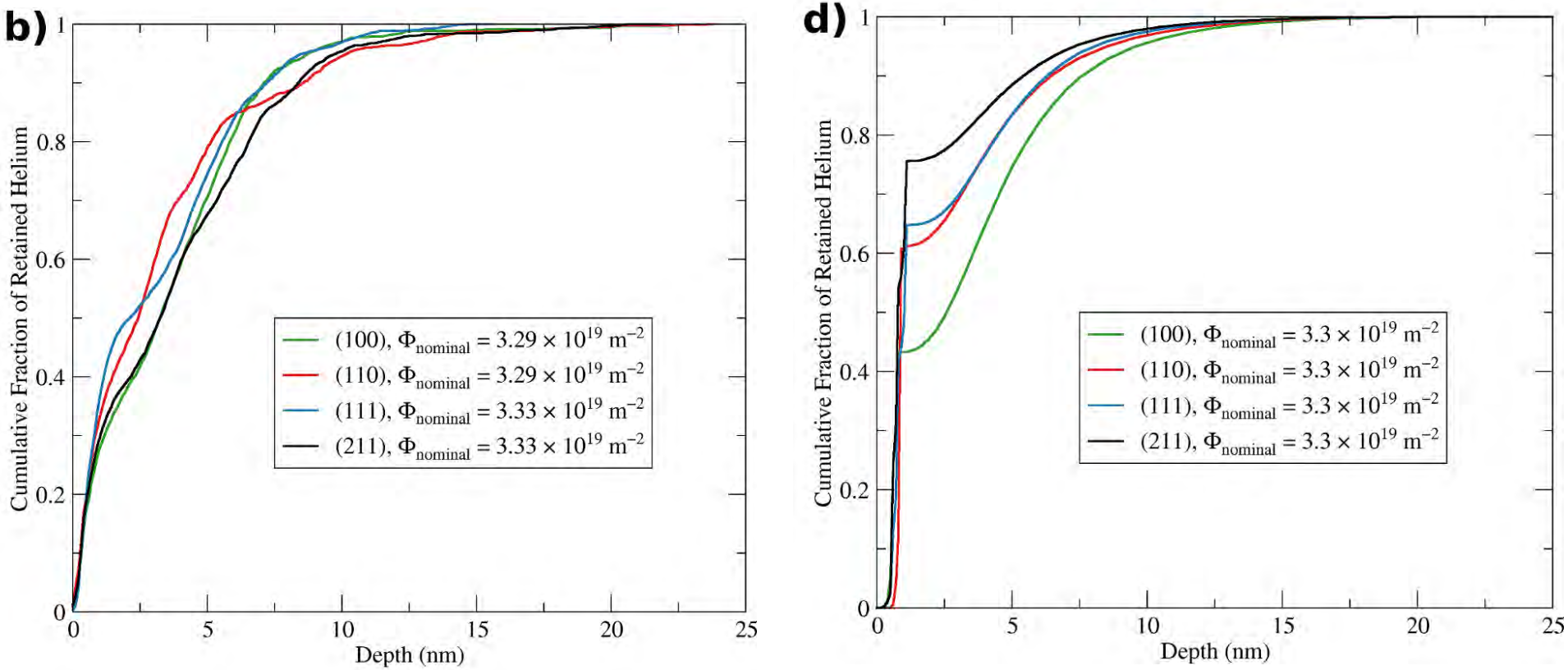


* Maroudas, Blondel, Hu, Hammond, and Wirth, *J. Phys. Cond. Matter.* **submitted**

Initial results including advection (drift diffusion) & modified trap mutation

Good early agreement does not persist at higher fluence: bubble bursting or modifications to the reaction rate constant are suspect. Future detailed comparisons of helium-vacancy cluster size distributions to help resolve this

MD simulations Γ_{He} **~ 4E25 m^{-2}s^{-1}** **Xolotl simulations**

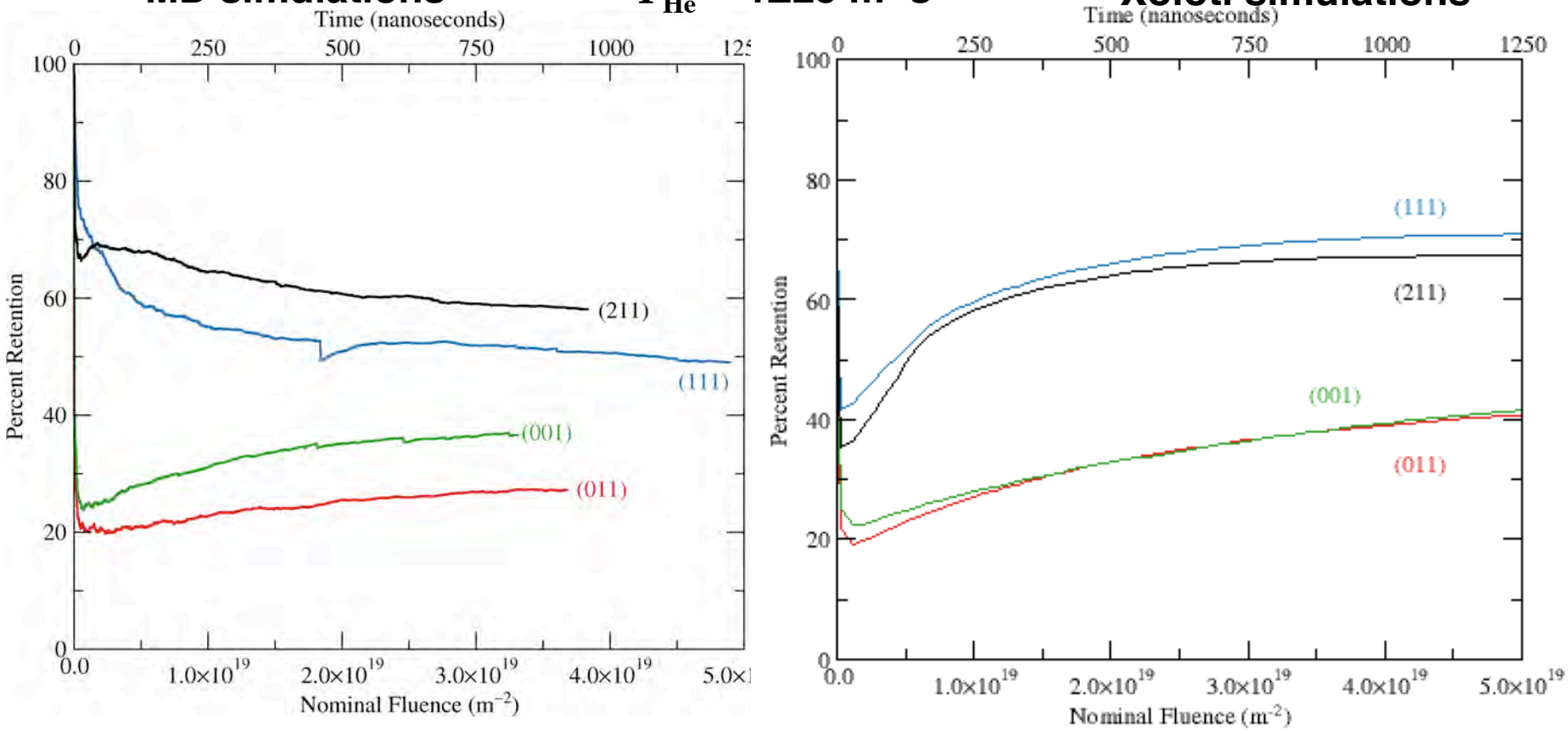


* Maroudas, Blondel, Hu, Hammond, and Wirth, *J. Phys. Cond. Matter.* **submitted**

More detailed Xolotl benchmarking to MD

100 eV He, 933 K

MD simulations Γ_{He} **~ 4E25 m^{-2}s^{-1}** **Xolotl simulations**

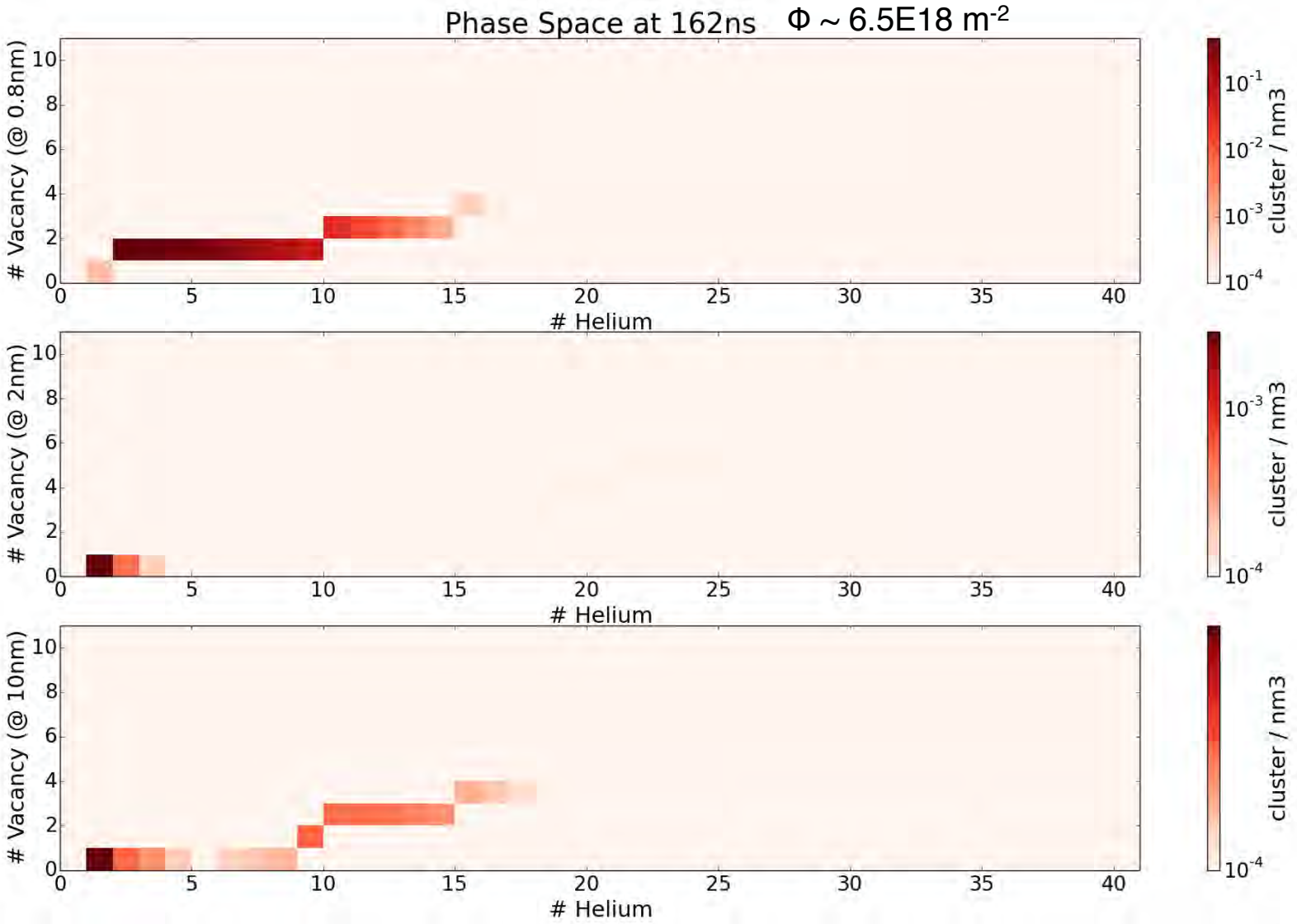


• Xolotl comparison/benchmarking to MD quite promising, but Xolotl is still missing (two) important physics:

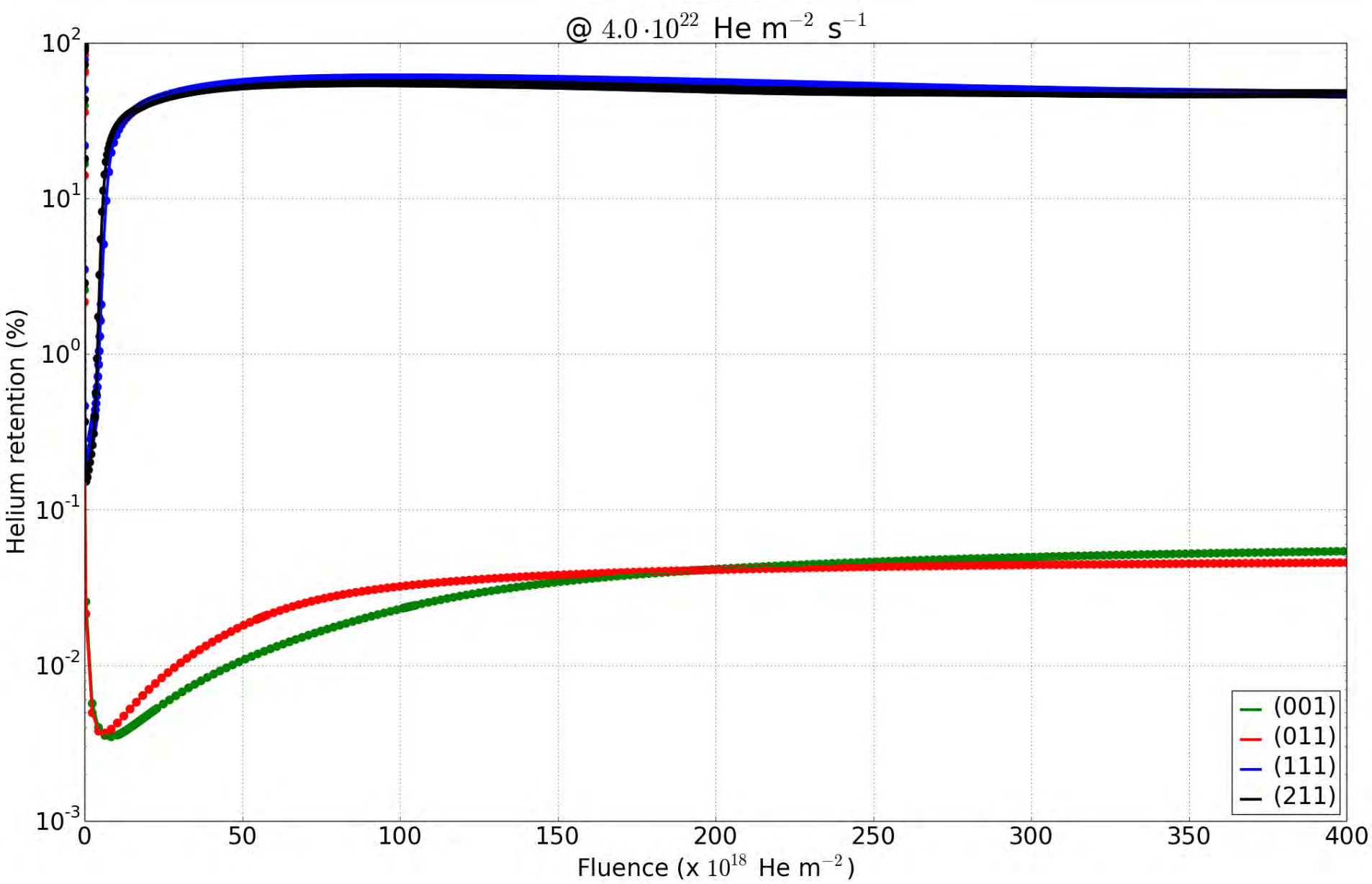
- Bubble bursting
- Modified trap mutation below (211) implemented as (111)
- Bubble coalescence

More detailed Xolotl results: (111) surface at Γ~4E25 $m^{-2}s^{-1}$

Bubble distributions are concentrated just below the surface (where the clusters stay relatively small) and deeper (~ 10 nm)

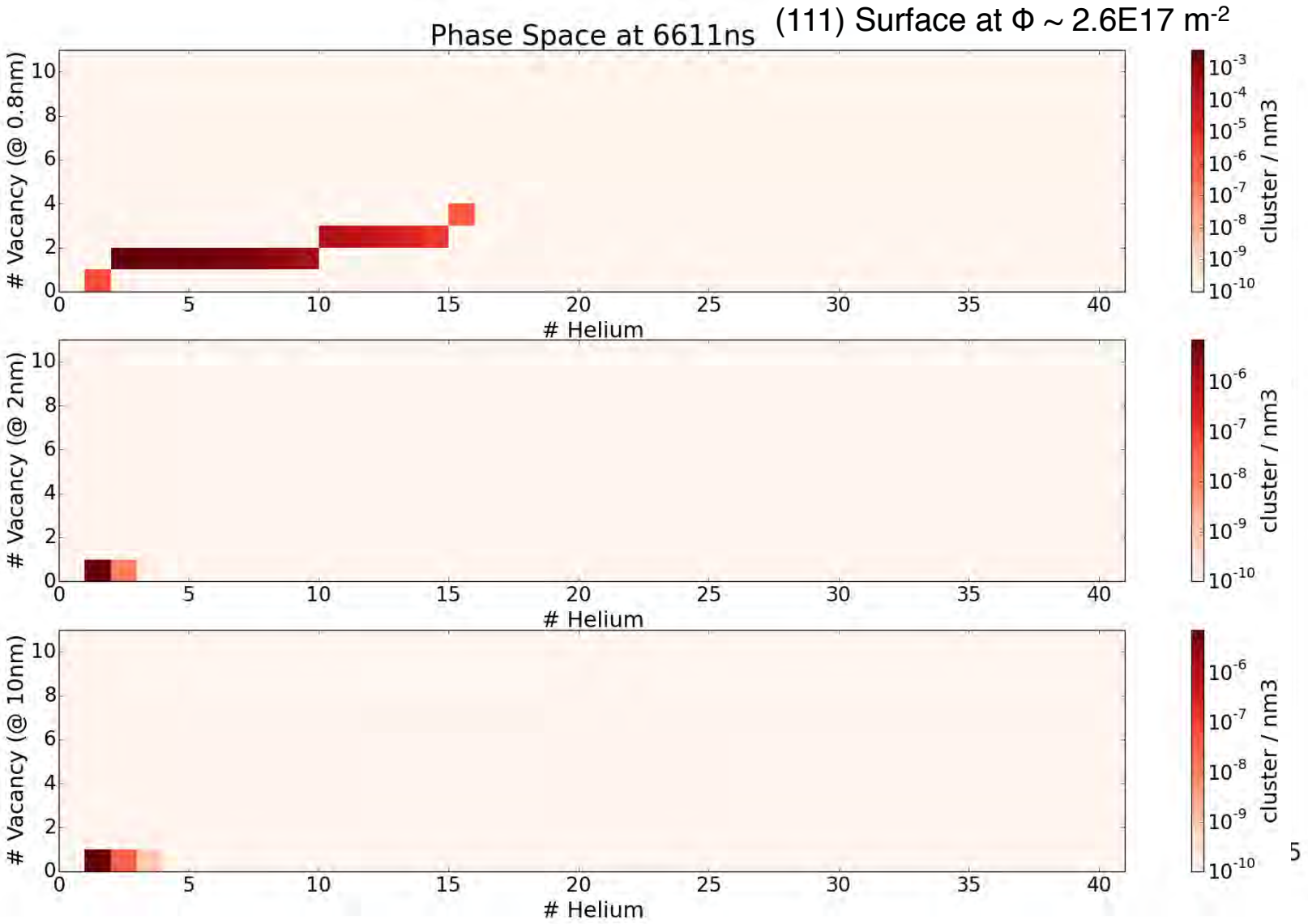


Xolotl results at ITER relevant flux of Γ~4E22 $m^{-2}s^{-1}$



More detailed Xolotl results: (111) surface at Γ~4E22 $m^{-2}s^{-1}$

Now, at lower flux, the bubble distributions are concentrated just below the surface, where nucleation initiates & bubbles begin to grow. Deeper nucleation is delayed

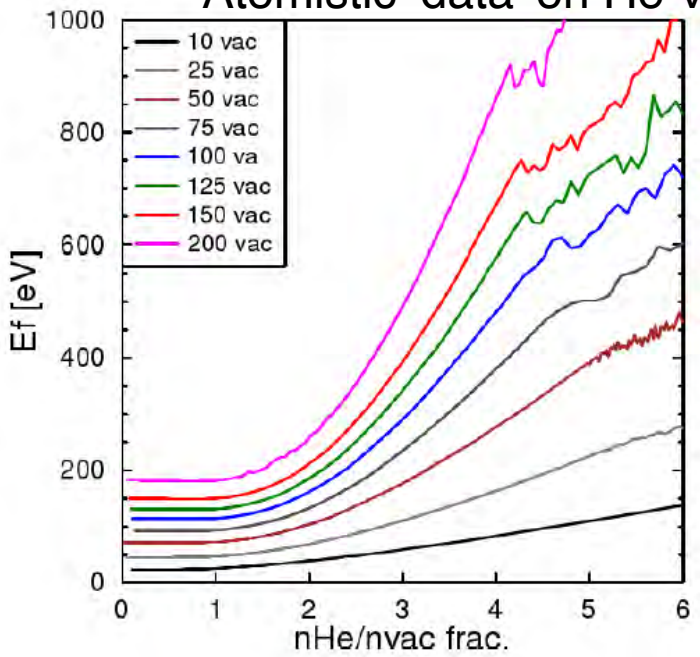


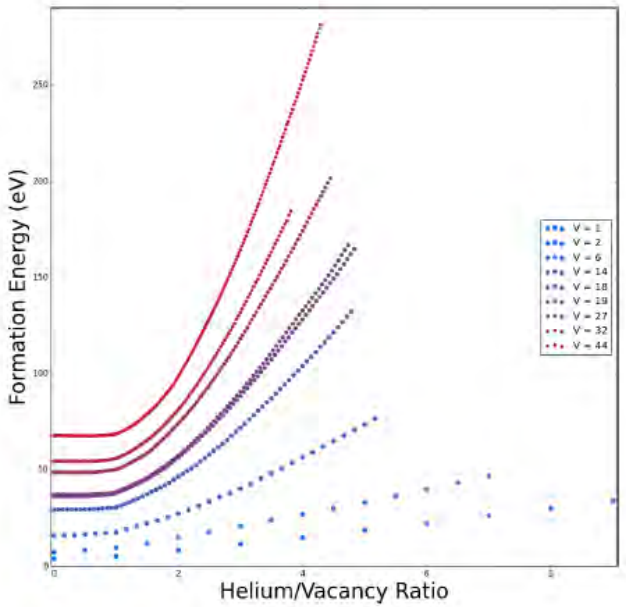
Bayesian informed approach towards UQ

$$P(H|E) \propto P(E|H) \cdot P(H)$$

- the **posterior** $P(H|E)$ (probability of the hypothesis H given the evidence E) that is infered
- the **likelihood** $P(E|H)$ (probability of the evidence E given the hypothesis H)
- and the **prior** $P(H)$ that gathers all the information one had before the evidence E was observed

Atomistic 'data' on He-vacancy formation energies





Approach: Create a model (2D Legendre polynomials) of this data to reduce memory (no look up table) & provide basis for interpolation & extrapolation

Bayesian informed approach towards UQ

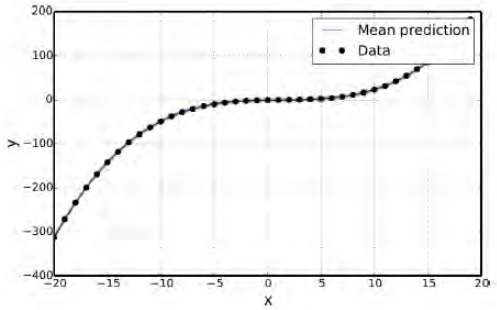
Markov Chain Monte Carlo (MCMC) Method: Metropolis-Hastings algorithm with adaptive proposal distribution.

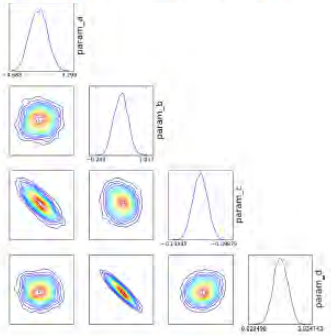
- Metropolis-Hastings: the step Y at t is kept with a probability α

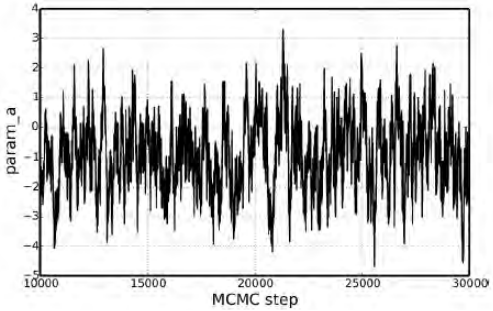
$$\alpha = \min(1, \frac{\pi(Y)}{\pi(X_{t-1})})$$

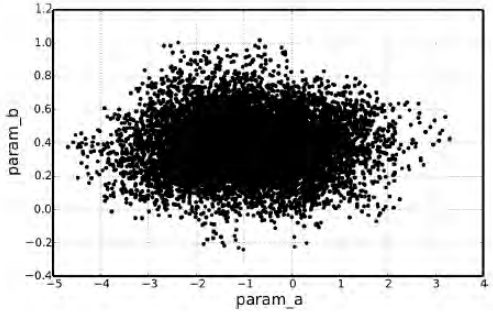
with π the target distribution.

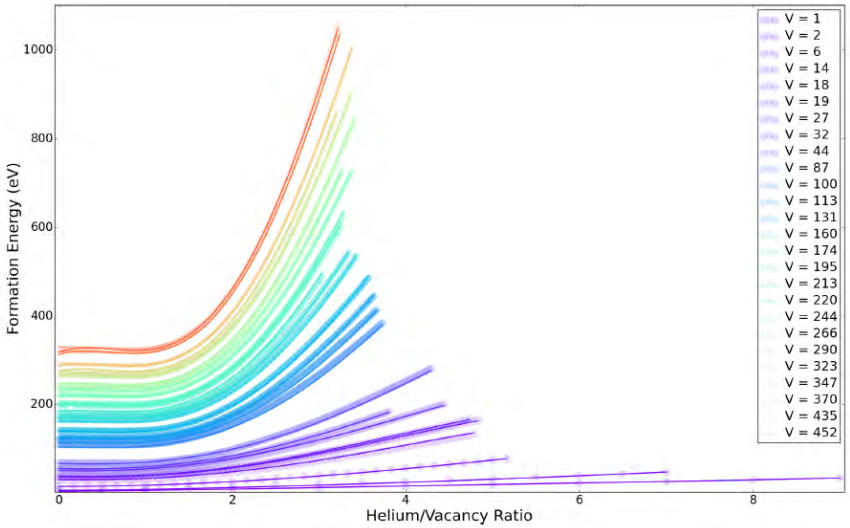
- Adaptive part: the proposal distribution (to go from a step to the next one) is a function of all the previous step.







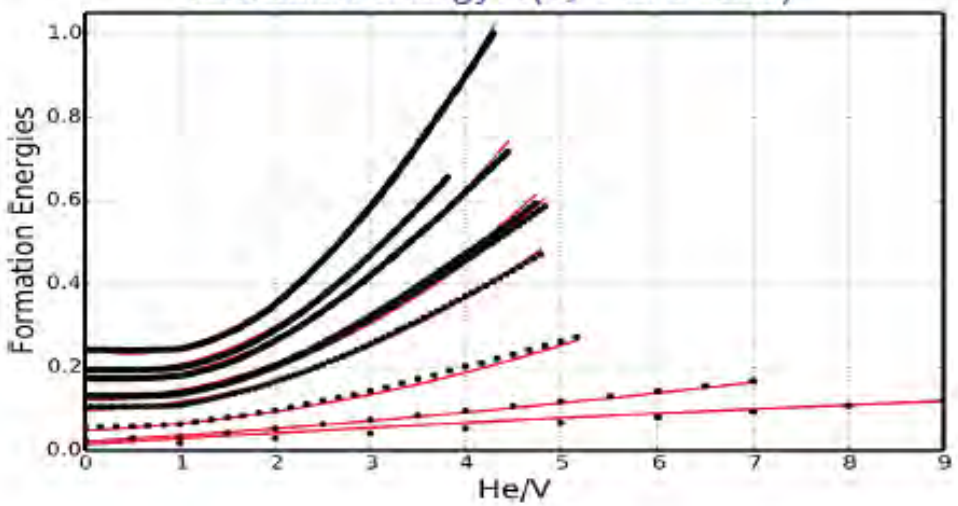




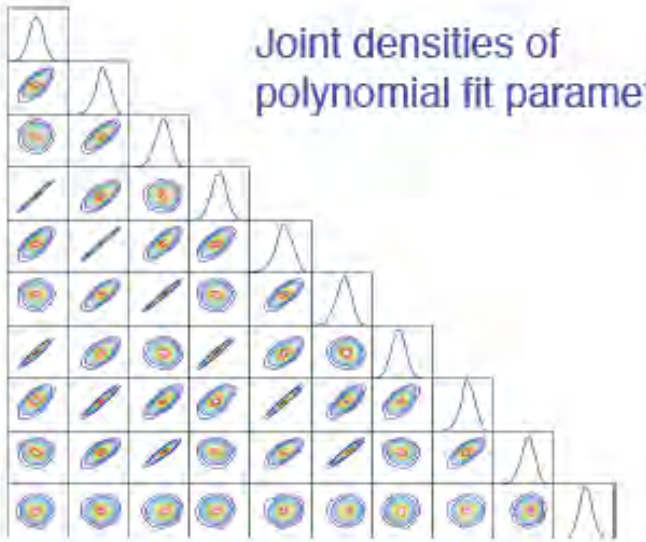
Bayesian informed approach towards UQ

- Close collaboration with SciDAC QUEST, and using UQ toolkit (http://www.sandia.gov/UQToolkit/)
- Performed Bayesian inference, and polynomial chaos expansion to determine the sensitivities of Helium – vacancy cluster formation and binding energies using polynomial expansions to interpolate atomistic database

Atomistic database of cluster formation energy: f(V, He/V ratio)



Joint densities of polynomial fit parameters

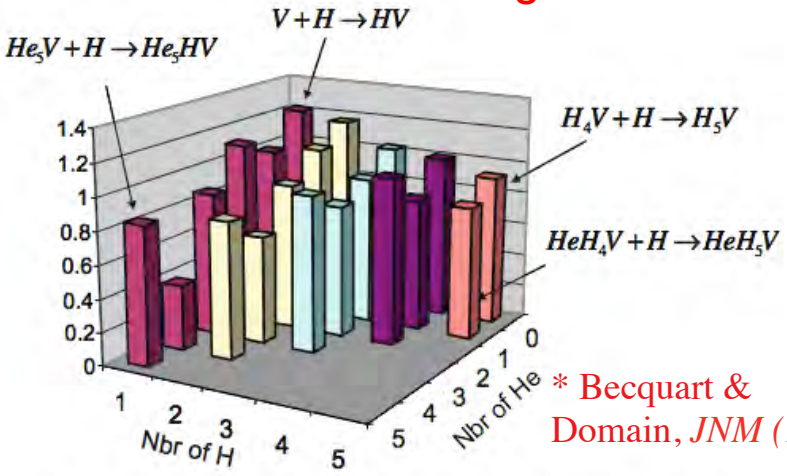


- Initial forward propagation of cluster formation energy variance through Xolotl has been performed -- **Formation energies turn out not to have impact on our QoI**
- Framework in place for Global Sensitivity Analysis
- Ongoing effort to provide detailed UQ analysis of influence of thermodynamic-kinetic cluster energetics impact on Helium retention and bubble distributions

He-H defect interactions in W

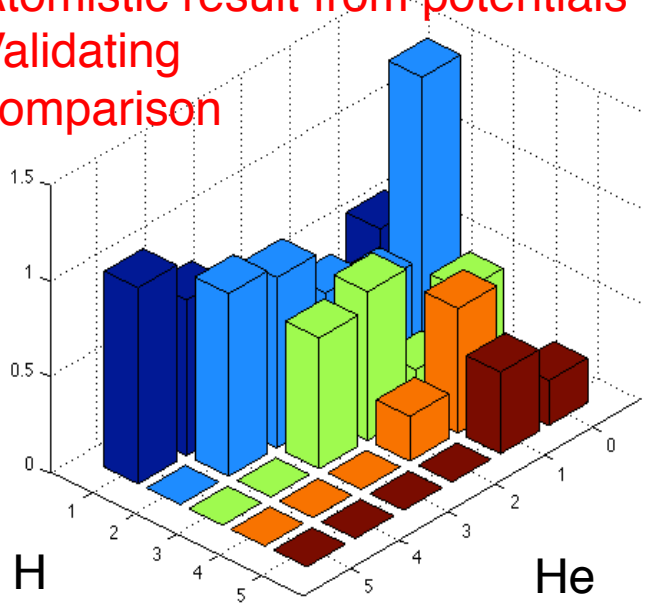
- Interatomic potential(s) derived to describe W-He* and W-He-H** interactions

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



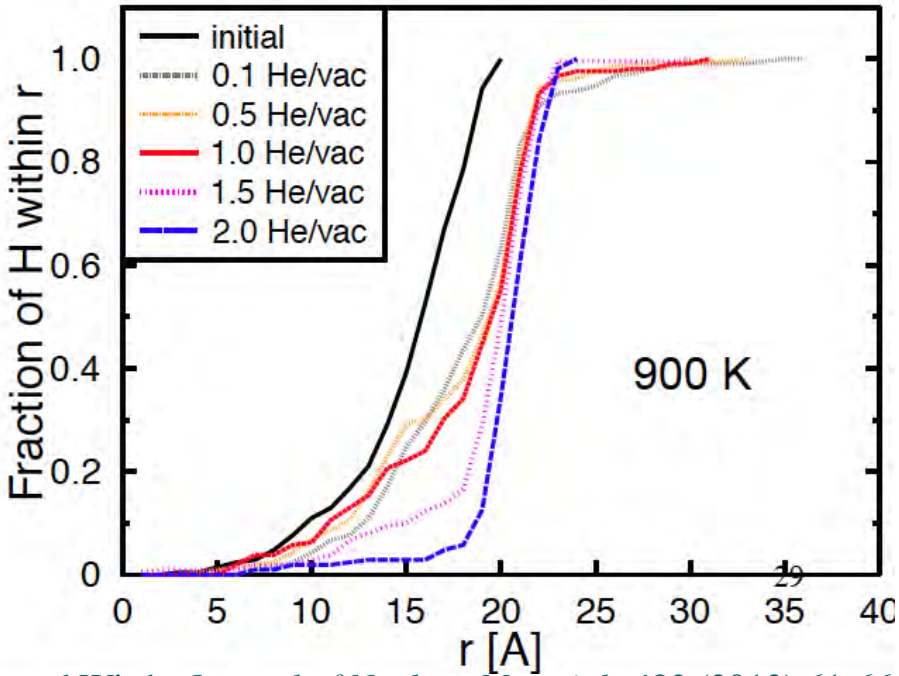
* Becquart & Domain, *JNM (1999)*

Atomistic result from potentials – Validating comparison



Validated potentials used to evaluate H partitioning to He bubbles

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

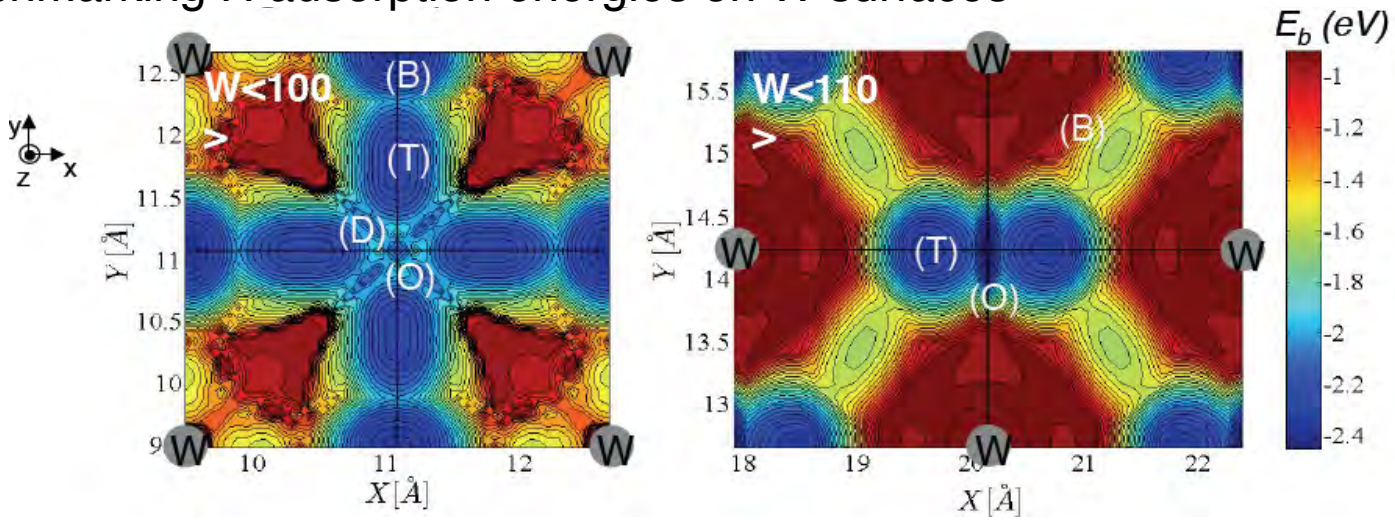


* Juslin and Wirth, *Journal of Nuclear Materials* **432** (2013) 61-66.

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

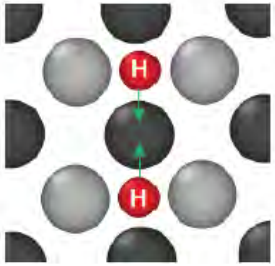
H – W potential validation & modification*

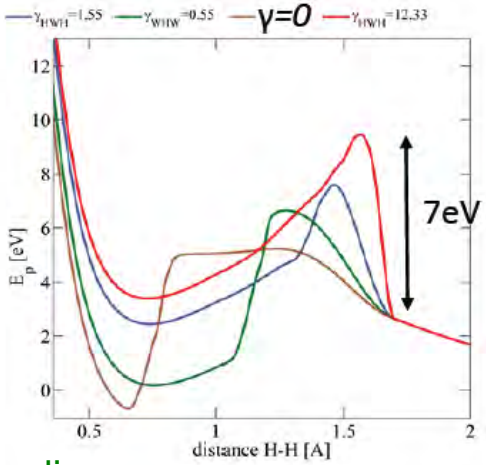
- Benchmarking H adsorption energies on W surfaces

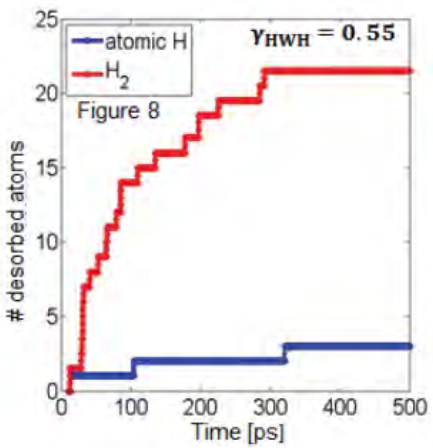


$$E{\downarrow}ads = 2.4eV\ (B),\ 2.3eV\ (T),2.1eV\ (O),\ 2eV\ (D)$$

- 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





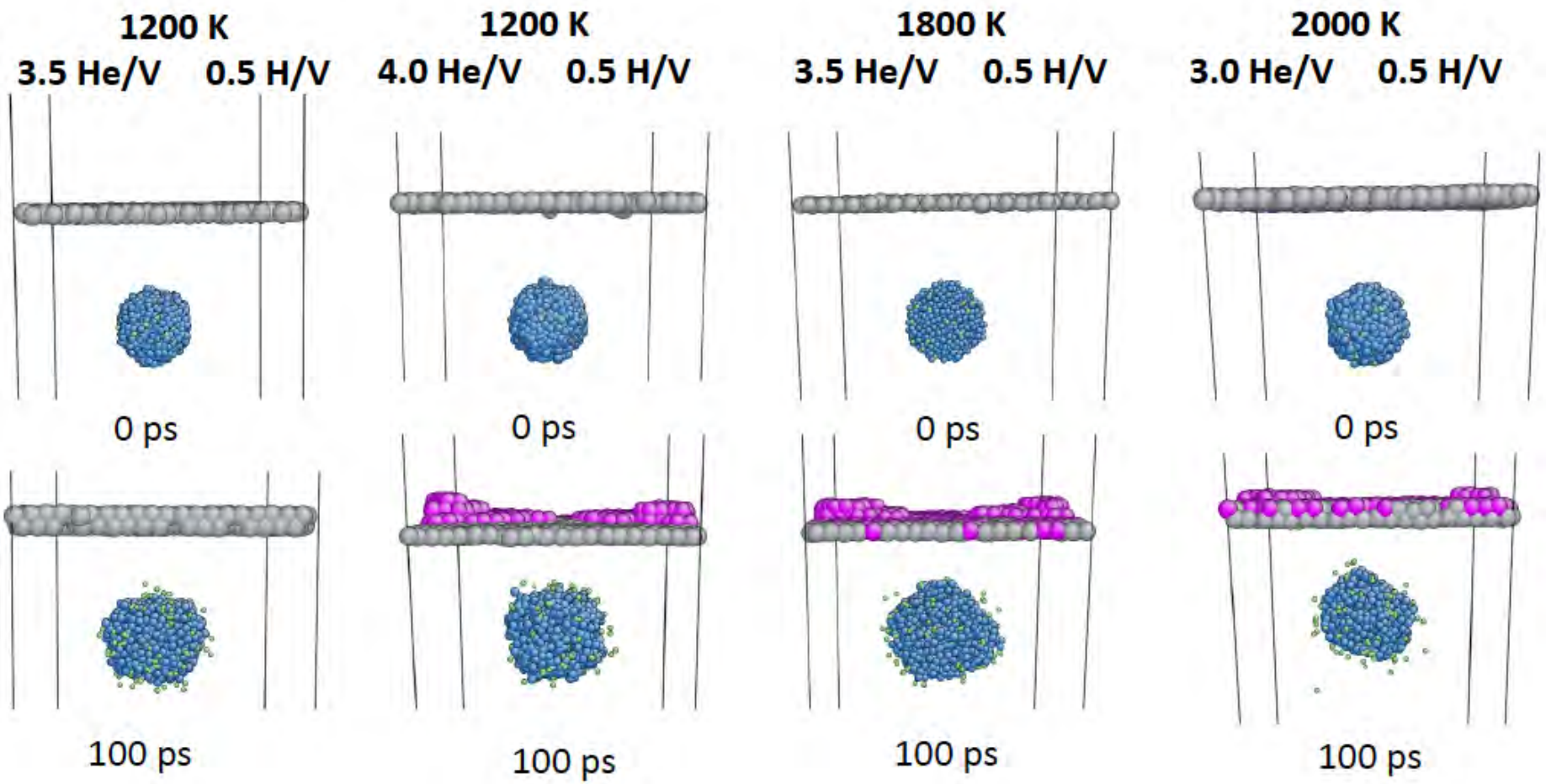


*J. Guterl et al., 2014 PSI proceedings

Preliminary results of He-H synergies & H trapping

- 2 nm diameter, over-pressurized He bubble created 2 nm below W (110) surface
  - populate with He & H (or random H in box), simulated at 1200,1500,1800, 2000 K for 100 ps, then quench to evaluate retention and H partitioning using modified Juslin

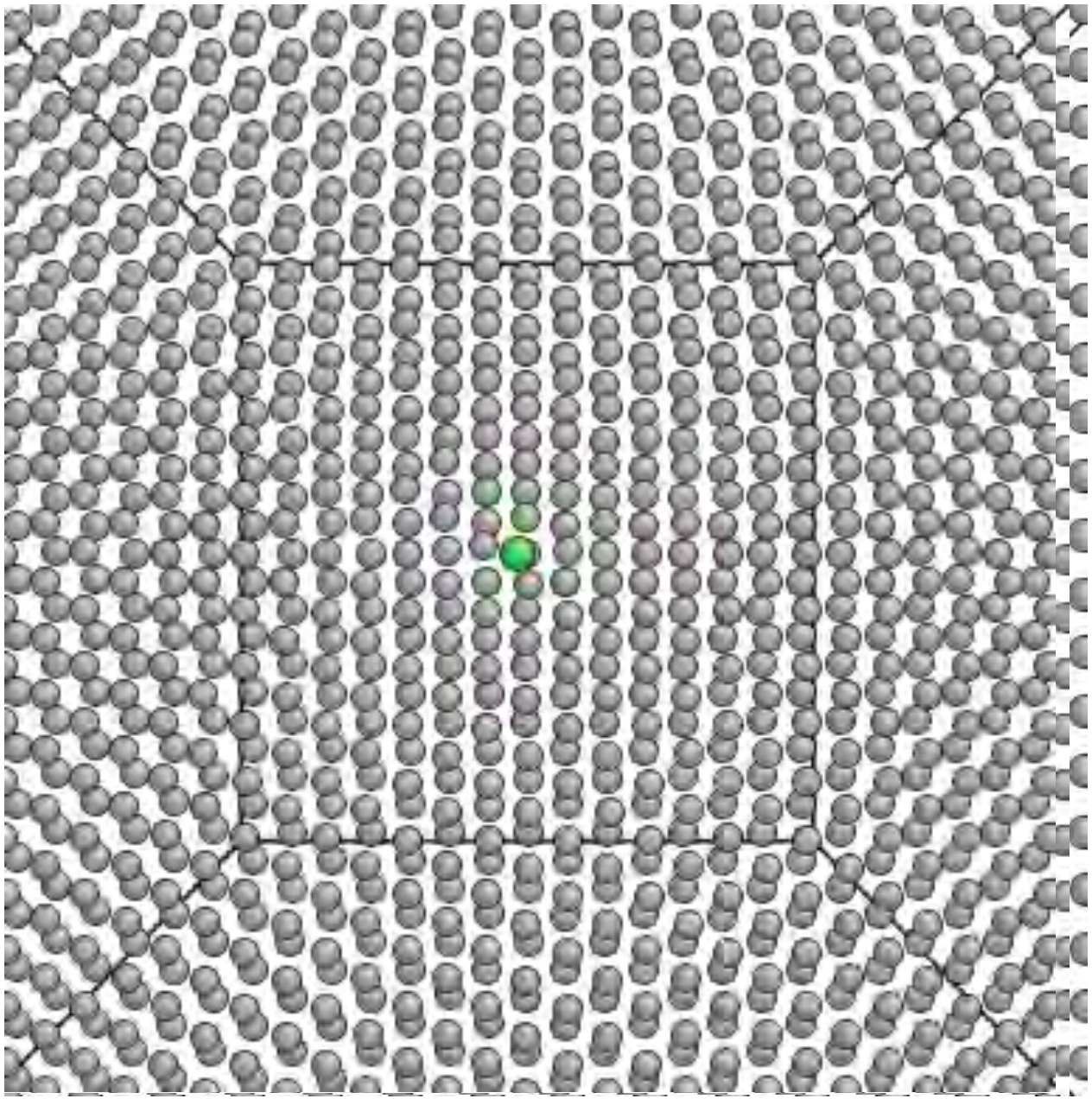
Green: Hydrogen
Blue: Helium
Magenta: Adatoms



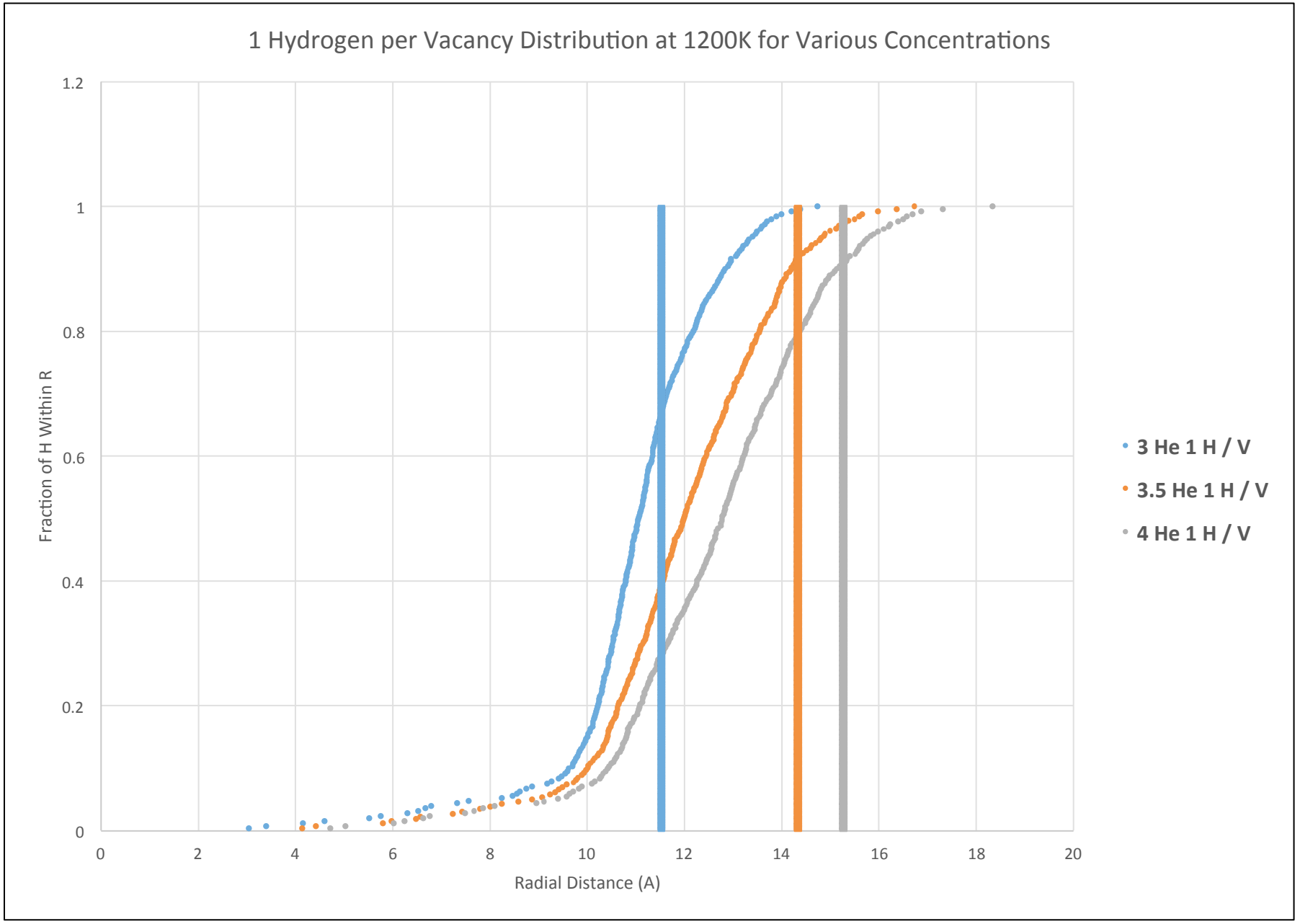
H distributions throughout the bubble

After 100 ps
at 1500K
3 He/Vac
0.5 H/vac (~125 H, ~0.7 at%)

- Hydrogen
- Helium
- Tungsten



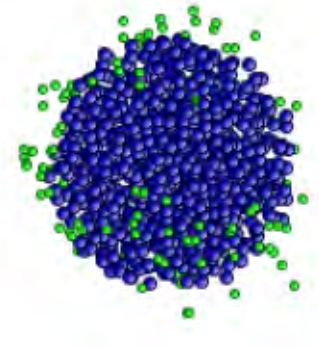
Preliminary results of He-H synergies & H trapping



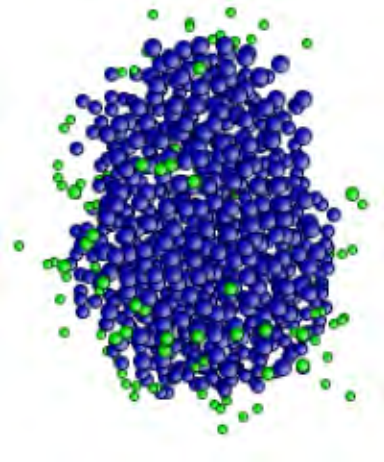
Preliminary results of He-H synergies & H trapping

Gas Atoms Remaining in Cavity at 1200 K				Gas atoms in box		Desorbed from box	
Concentration	**He**	**H**	**H %**	**He**	**H**	**He**	**H**
3 He 0.5 H / V	750	88	70.4	750	125	0	0
3 He 1 H / V	753	167	66.5	753	251	0	0
3.5 He 0.5 H / V	889	119	93.7	889	127	0	0
3.5 He 1 H /V	903	235	91.4	903	257	0	0
4 He 0.5 H / V	996	117	93.6	996	125	0	0
4 He 1 H / V	1008	229	90.9	1008	252	0	0

Initial bubble (3 He/v, 0.5 H/v, 1200K)



Gas Atoms Remaining in Cavity at 2000 K				Gas atoms in box		Desorbed from box	
Concentration	He	H	H %	He	H	He	H
3 He 0.5 H / V	762	118	92.9	762	127	0	0
3 He 1 H / V	768	238	93.0	768	256	0	0
3.5 He 0.5 H / V	875	118	94.4	875	125	0	0
3.5 He 1 H /V	889	250	98.4	889	254	0	0
4 He 0.5 H / V	1004	120	95.2	1004	126	0	0
4 He 1 H / V	994	226	89.7	994	252	0	0

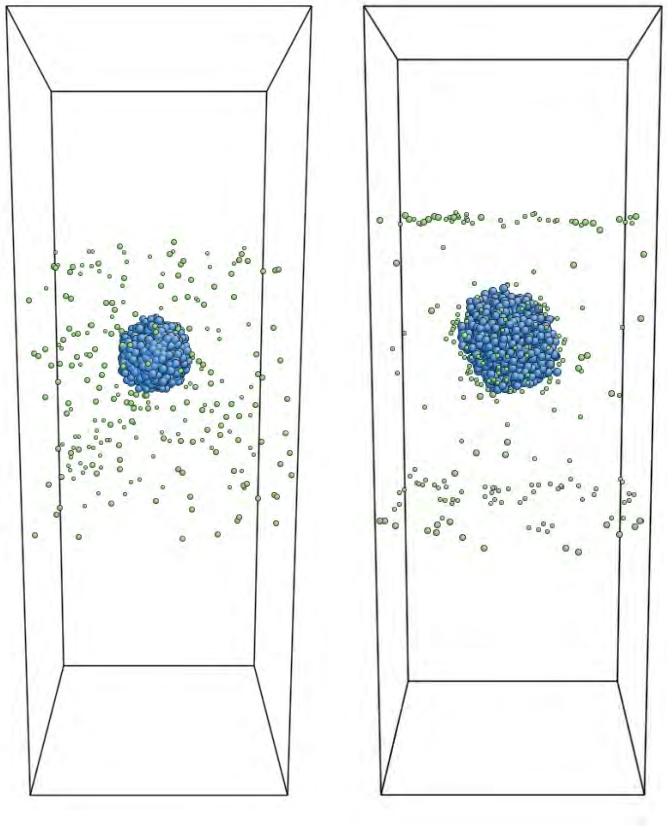


- Significant H content trapped in bubble and periphery, even at 2000K

Have we 'biased' the H to the bubbles?

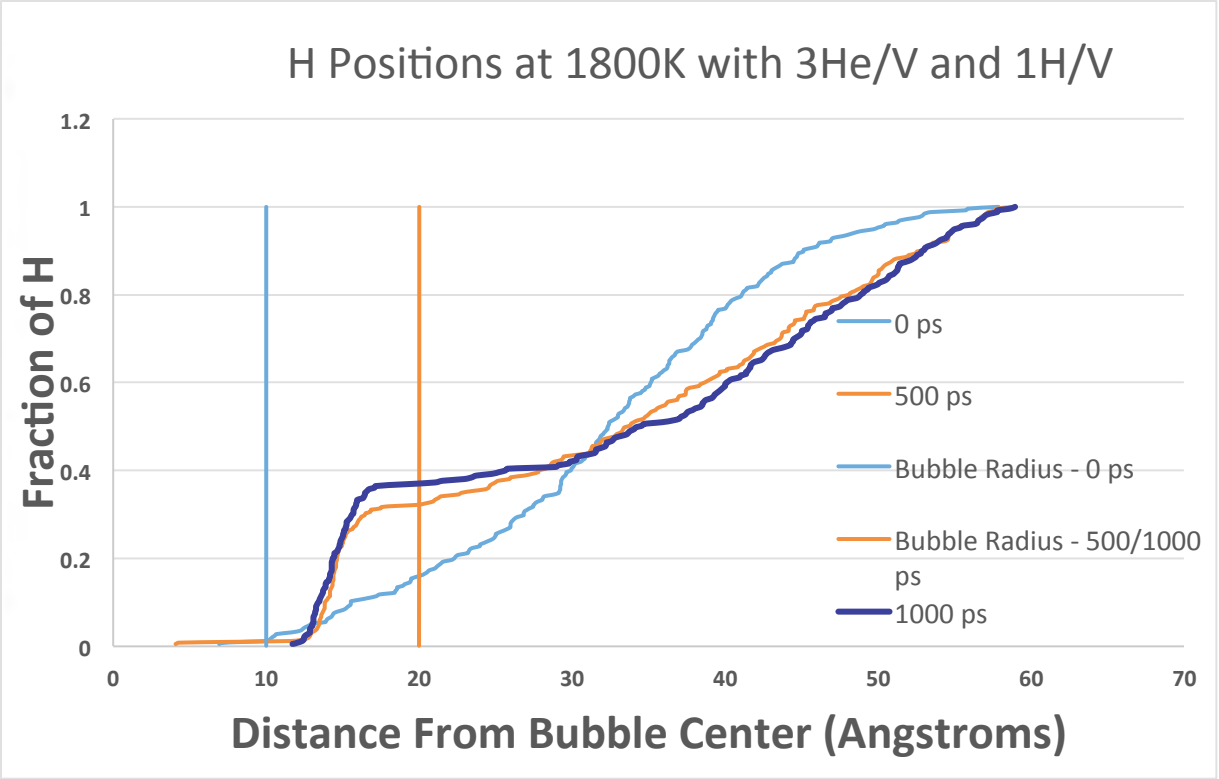
1800 K, 2 nm diameter He bubble with He/V = 3. 255 H atoms (~1.4 at%) randomly distributed throughout simulation cell

He/V =3 H/V = 1



0 ps 1 ns

	0 ps	500 ps
H in simulation	255	255
H in bubble	2	82

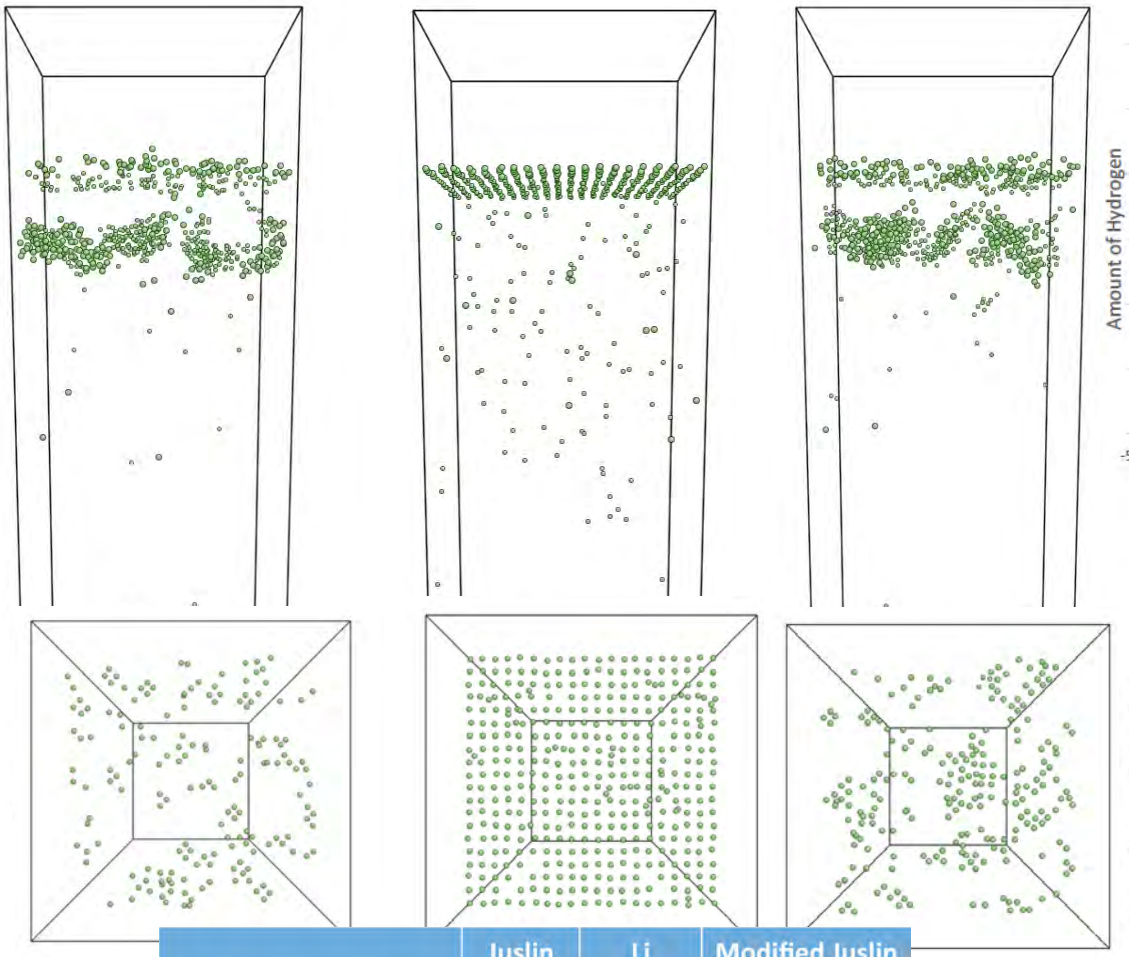


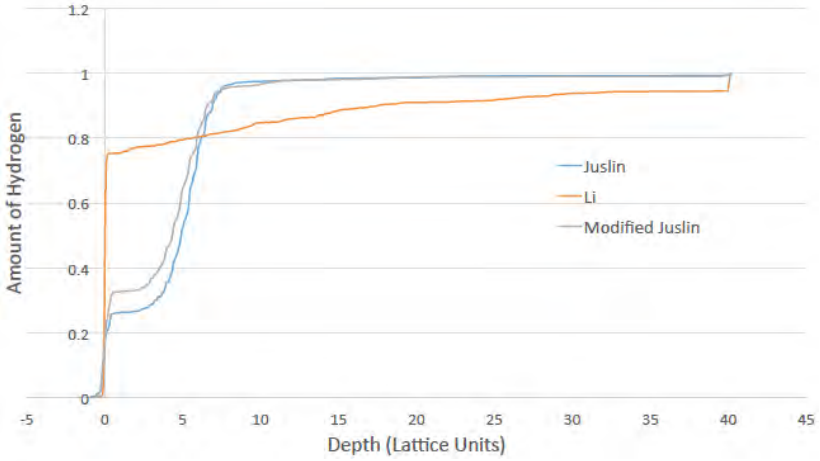
W-H interactions are key uncertainty, MD simulations: Γ=2.5x10^{27} H m^{-2}s^{-1}, 1200K

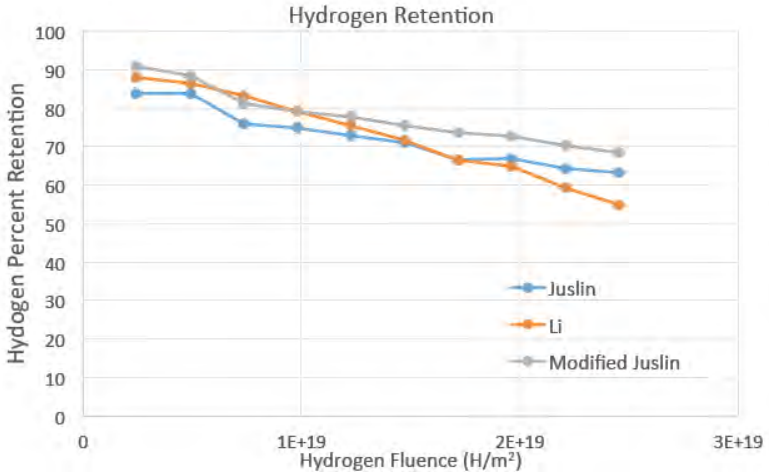
Juslin

Li

Modified Juslin







	Juslin	Li	Modified Juslin
Number of H at Surface	168	412	227
Percent of H at Surface	26.5%	74.9%	33.1%

Summary

• Multiscale materials simulations being used to evaluate He bubble nucleation & He-H synergies

- Results clearly indicate highly mobile He self-traps and small mobile He clusters undergo trap mutation (He_x –> He_xV_y + I_y) that immobilizes clusters leading to nucleation of growing, highly over-pressurized He bubbles. Bubble growth through trap mutation & loop-punching produce substantial surface roughness. Growing bubbles eventually rupture
- Promising results for benchmarking of Xolotl against MD (& KMC, though not shown)
- Strong influence of implantation flux on bubble size distributions as a function of depth – impact of (radiation/thermal) damage still to be resolved

• Initial framework for UQ analysis of the impact of uncertainty in He-vacancy thermo-kinetics on He bubble nucleation, retention and W surface response

• Preliminary investigations of H trapping at He bubbles indicates quite strong trapping, with H preferentially located at bubble periphery, even to high Temperatures beyond typical TDS measurements – validity of the potentials is appropriate question to ask. Future efforts to validate/refine W-H potential by DFT calculations of H(-He)-defect interactions near W surfaces

• Future effort to understand He-H synergies & impact on H/D/T recycling and retention