Molecular Dynamics and Density Functional Simulations of Tungsten Nanostructure Formation by Helium Plasma Irradiation

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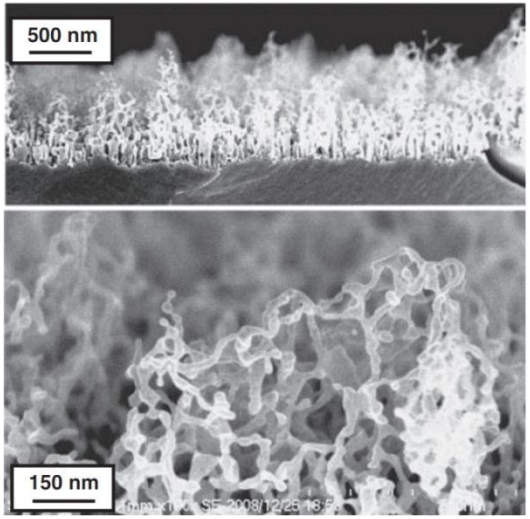
Acknowledgments
N. Yoshida (Kyushu University), Y. Ueda, H. Lee (Osaka University)

Outline

- Introduction
- Four step process of tungsten nanostructure formation
- MD-MC hybrid simulation to represent tungsten nanostructure formation
- Summary

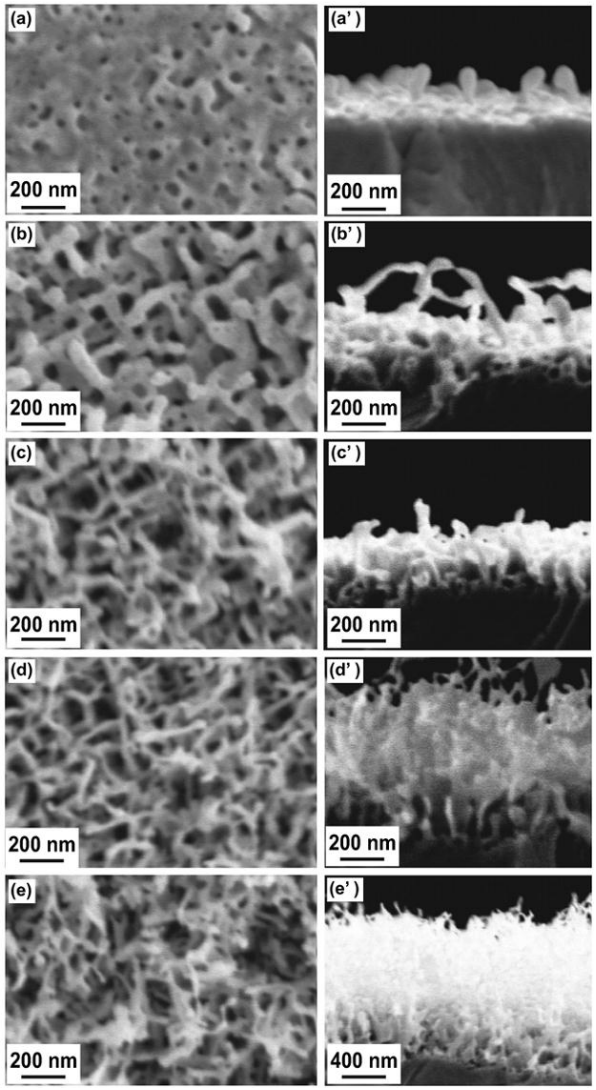
Tungsten nanostructure by He irradiation

By the He irradiation onto tungsten, fuzzy nano-structures of 10-50 nm in diameter grow.

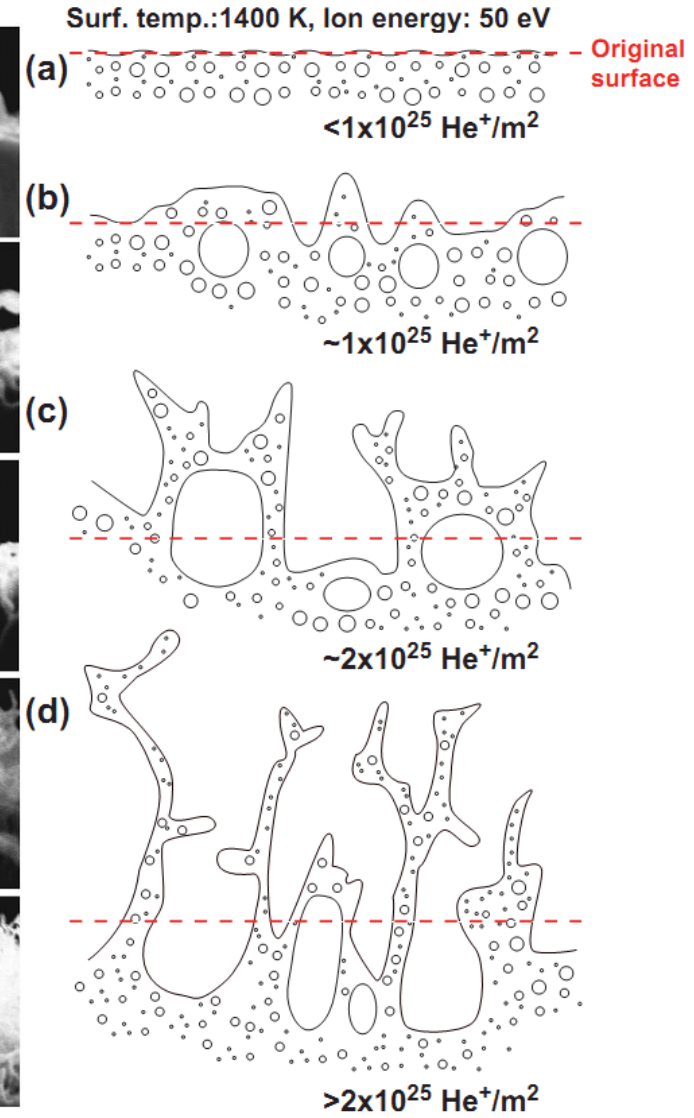


Fuzz structure

*S. Kajita, et al., JJAP. 50 (2011) 08JG01



*S. Kajita, et al., Nucl. Fusion. 49 (2009) 095005.



Story of Fuzz on a W surface

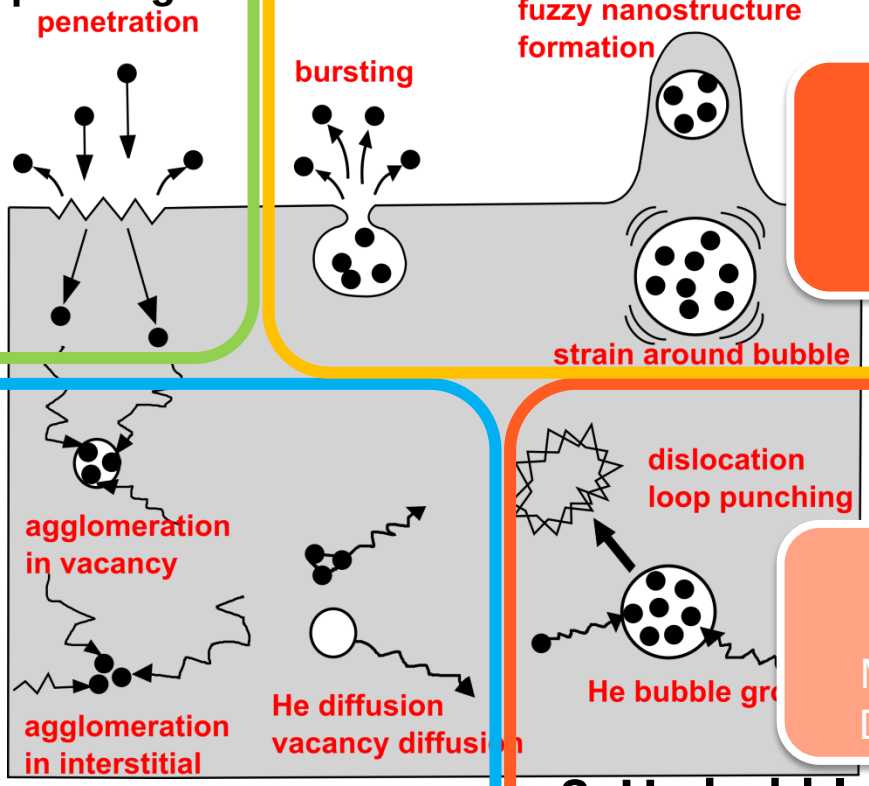
S. Kajita, et al., J.Nucl.Mater.418(2011)152-158

Four-step process of tungsten nanostructure formation

1. Penetration

- Competition of penetration and sputtering
- penetration range depending on incident energy

BCA
Binary collision approximation



4. Fuzzy nanostructure growth

- Bursting on the surface
- Formation of fuzzy structure
- How does He-bubble play a role?

MD-MC
Hybrid simulation

DFT
First-principle calculation

MC
Monte-Carlo

MD
Molecular Dynamics

2. Diffusion & agglomeration

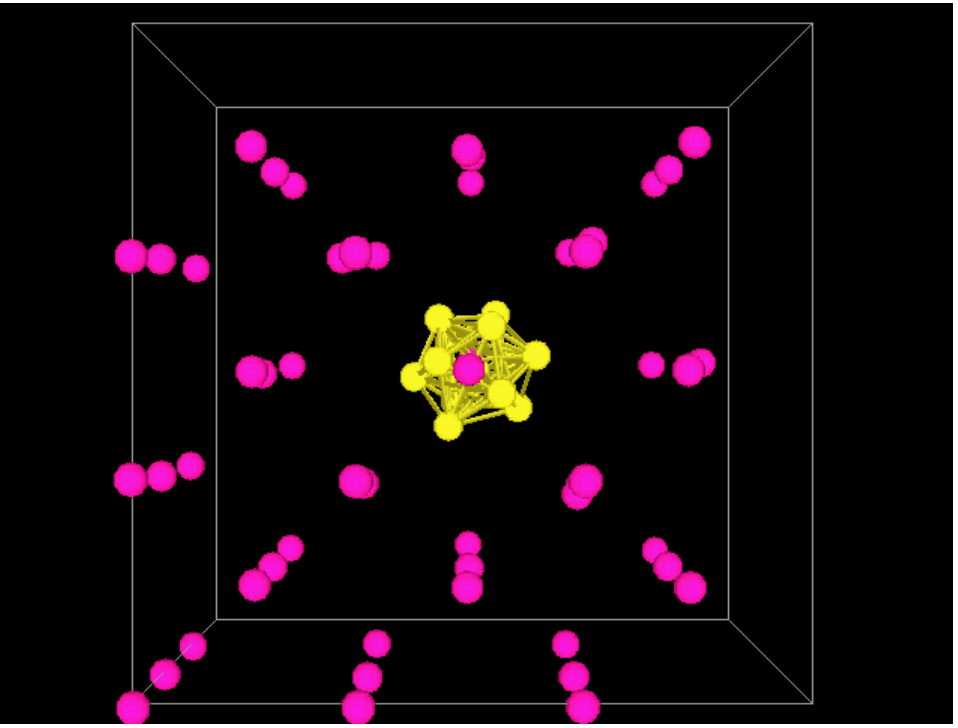
- Agglomeration of He, differing from H
- Diffusion of He, differing from H

3. He bubble growth

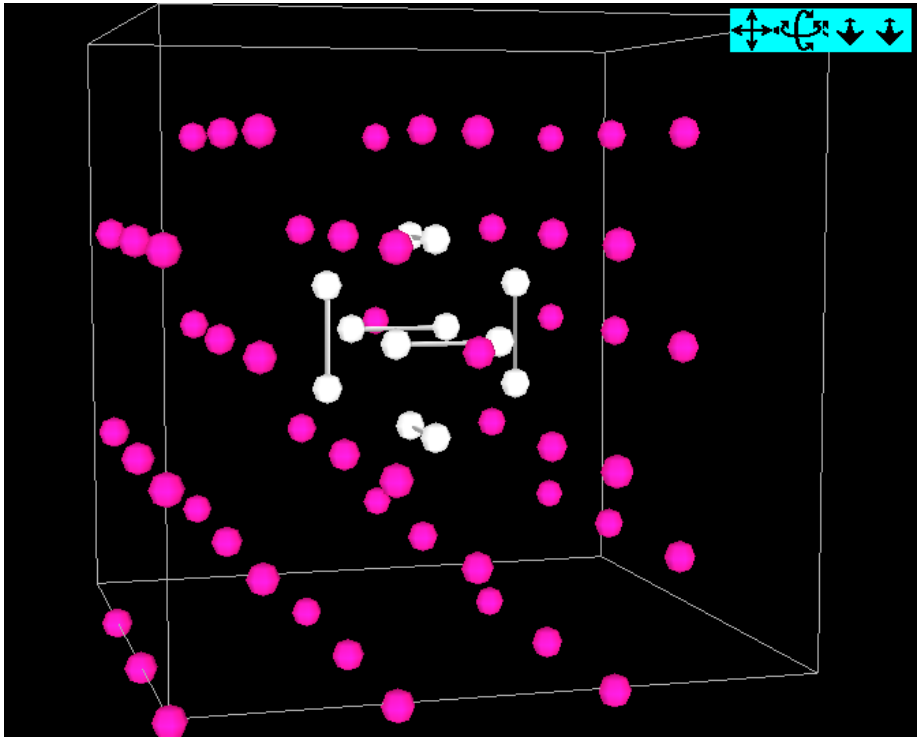
- Growth of He bubble to the size of 1nm or grater.
- Bursting and inter-bubble fracture of He bubble and Loop punching.

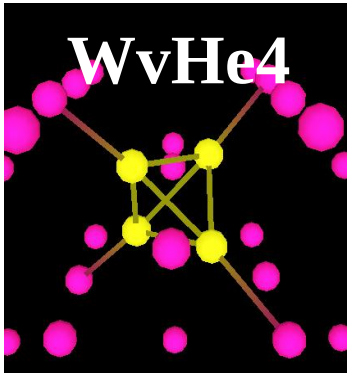
2nd process He cluster in mono-vacancy

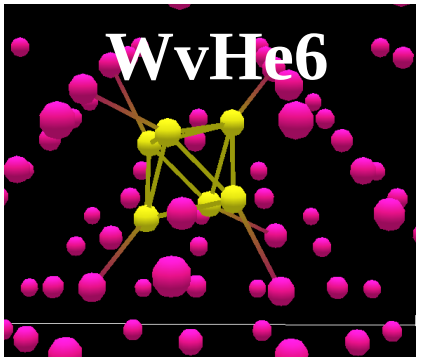
13He in mono-vacancy of W

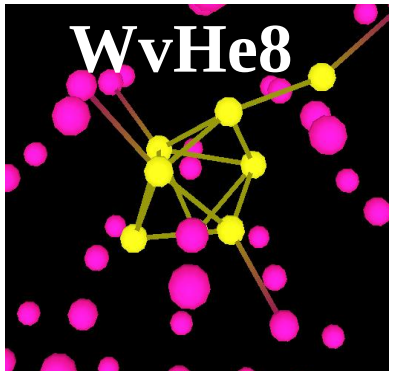


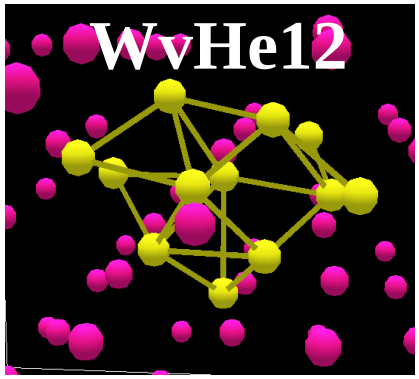
12H in mono-vacancy of W
(but one H locates on unstable position)







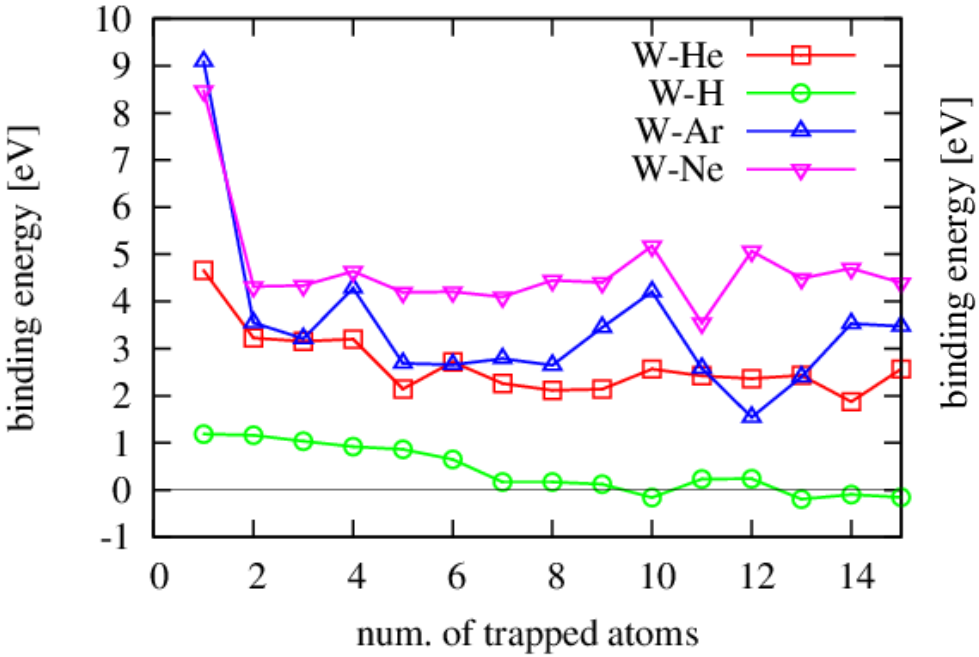




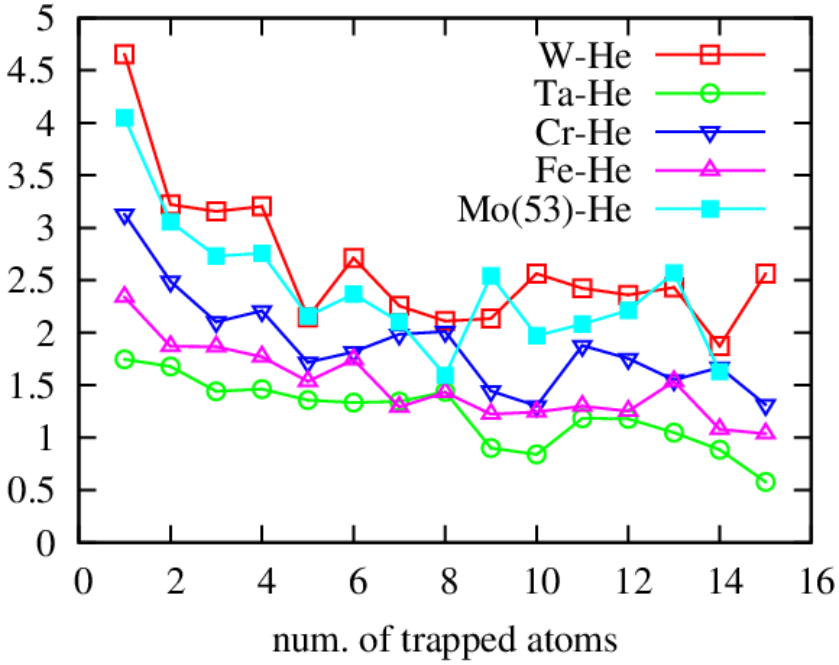
2nd step Nobel Gas Agglomeration is Unlimited

The binding energies of He atoms in a mono-vacancy calculated by OpenMX code based on density functional calculation (DFT).

- Binding energy of Helium in tungsten is always positive.
  → Helium agglomeration is advanced.
  → Hydrogen agglomeration is stopped.
- Helium can agglomerate in also many kinds of metallic materials.



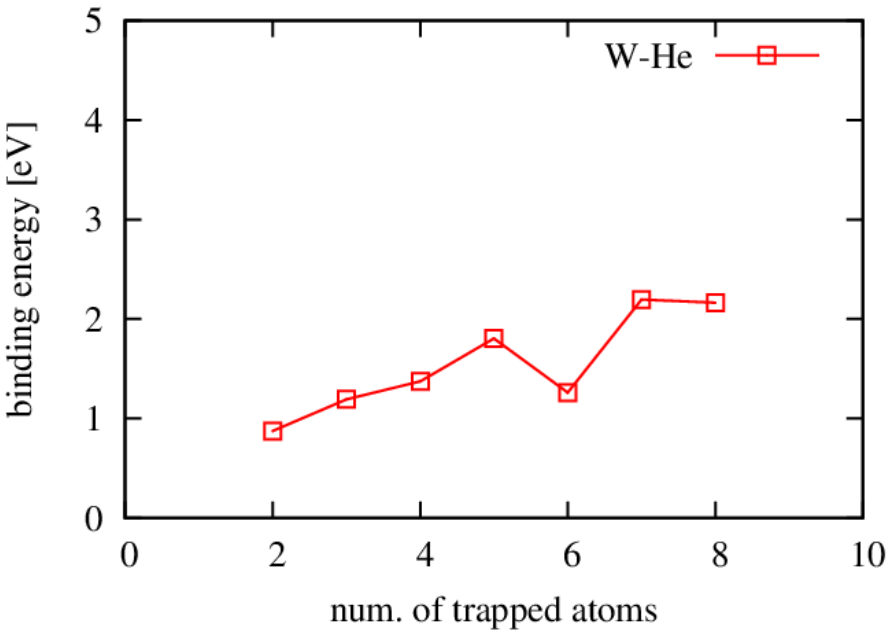
Binding energy of noble gas and hydrogen in tungsten with mono-vacancy



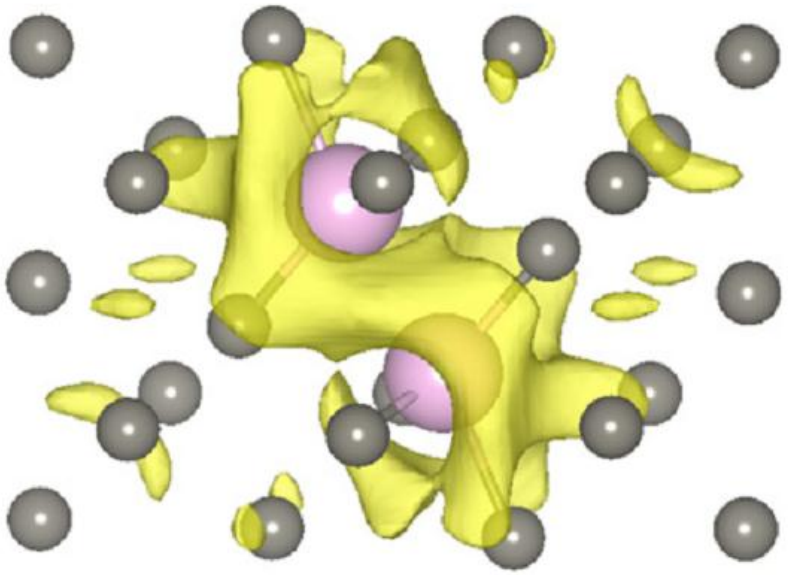
Binding energy of He in metals with mono-vacancy

2nd process Helium can agglomerates without vacancy

- Helium agglomeration is advanced even if it is located at interstitial site



Binding energy of He at tungsten interstitial site*



The lower electron density region**

- Noble gas cluster generates the region in which is lower than that of pure bulk material. The region becomes new stable site for the next noble gas atoms.

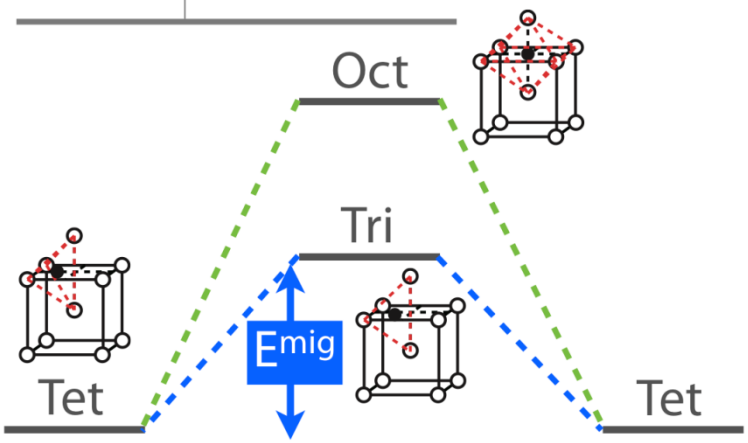
* A. Takayama,et al., PSI2014

T. Tamura, R. Kobayashi, S. Ogata, A. M. Ito, Model. Sim. Mater. Sci. Eng., **22 (2014) 015002.

He diffusion is accelerated by clustering

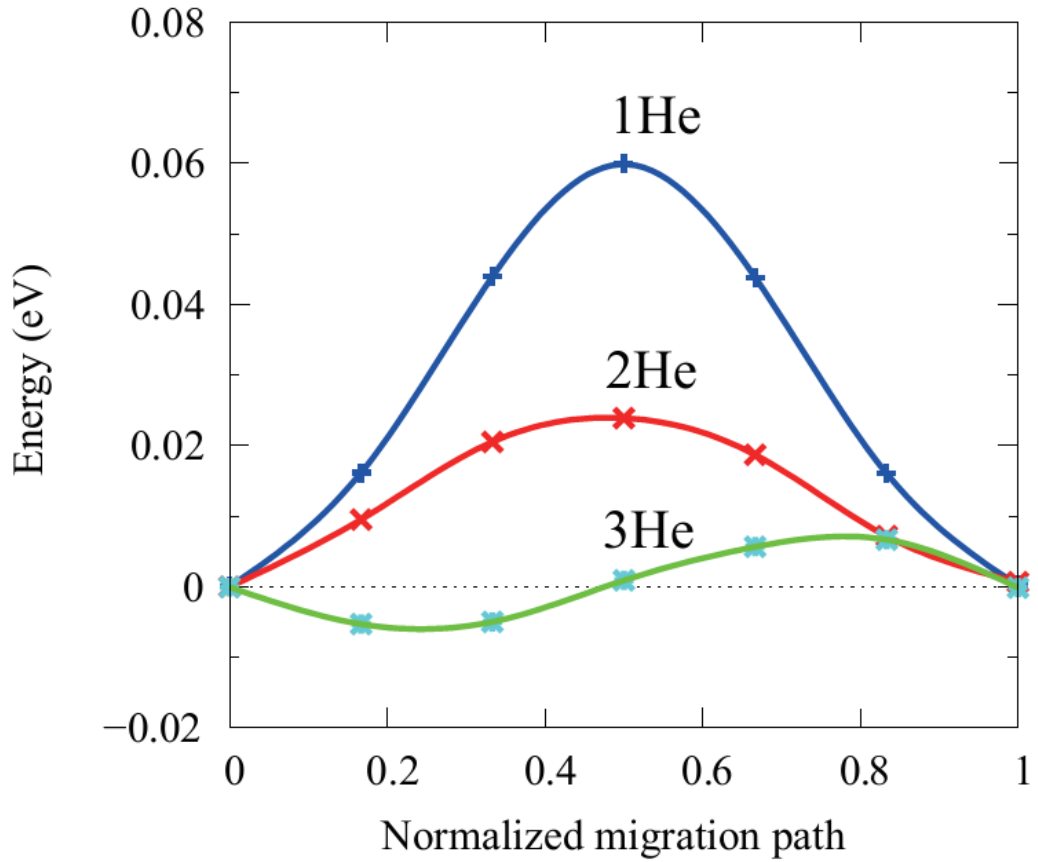
- The migration barrier of helium atom is smaller than that of hydrogen.
- The migration barrier of helium dimer becomes one third of single atom.

	E^{mig} [eV]
H	0.20
He	0.06
Ne	0.17
Ar	**0.19**



$$E^{\mathrm{mig}} = E_{\mathrm{tot}}(\mathrm{Tri}) - E_{\mathrm{tot}}(\mathrm{Tet})$$

Migration barrier of single atom*



Migration barrier of He cluster**

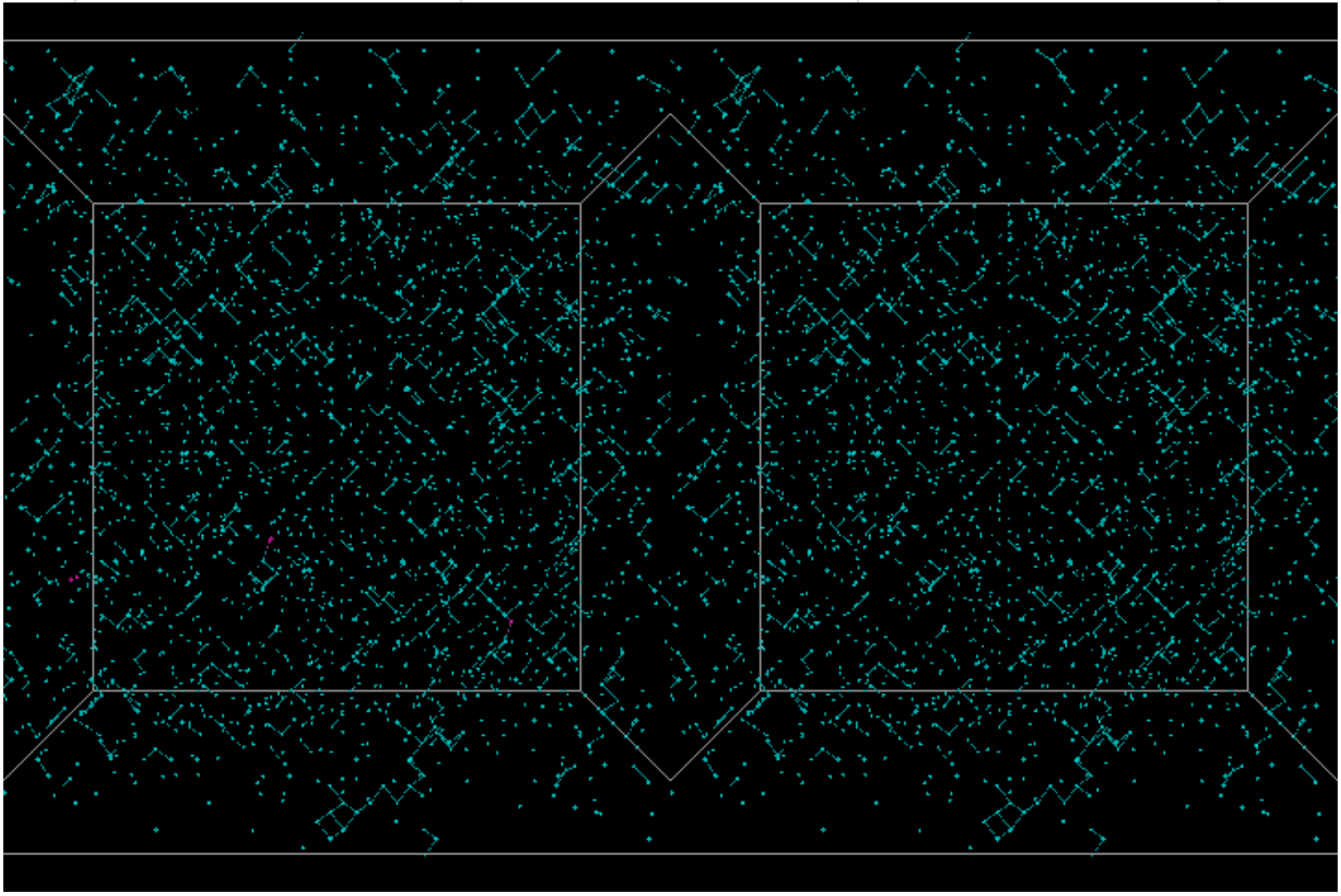
*T. Tamura, R. Kobayashi, S. Ogata, A. M. Ito, Model. Sim. Mater. Sci. Eng., **22** (2014) 015002.
**T. Tamura, PSI2014

MD comparison between He agglomeration with/without vacancy

In some situations, the helium bubbles growth is faster in the system without vacancy rather than the case with vacancy.

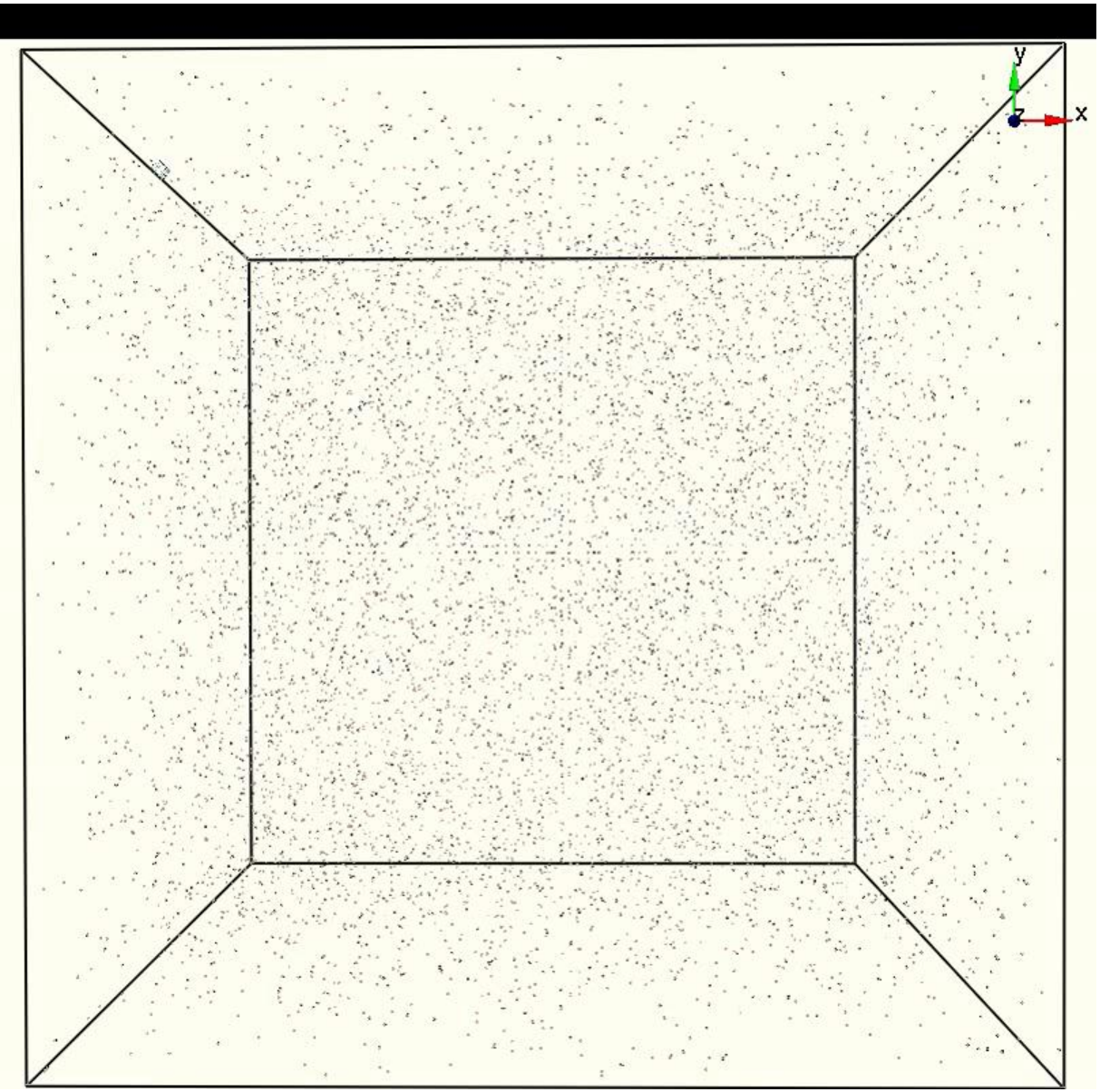
with 1% vacancy

without vacancy



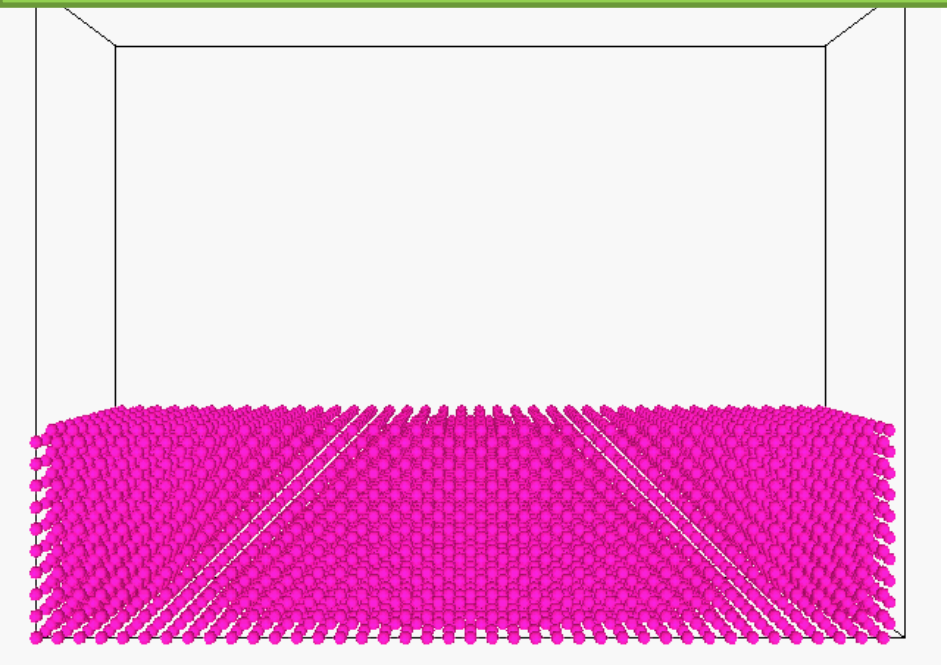
Loop punching observed by MD simulation

R. Kobayashi, et.al. PSI2014



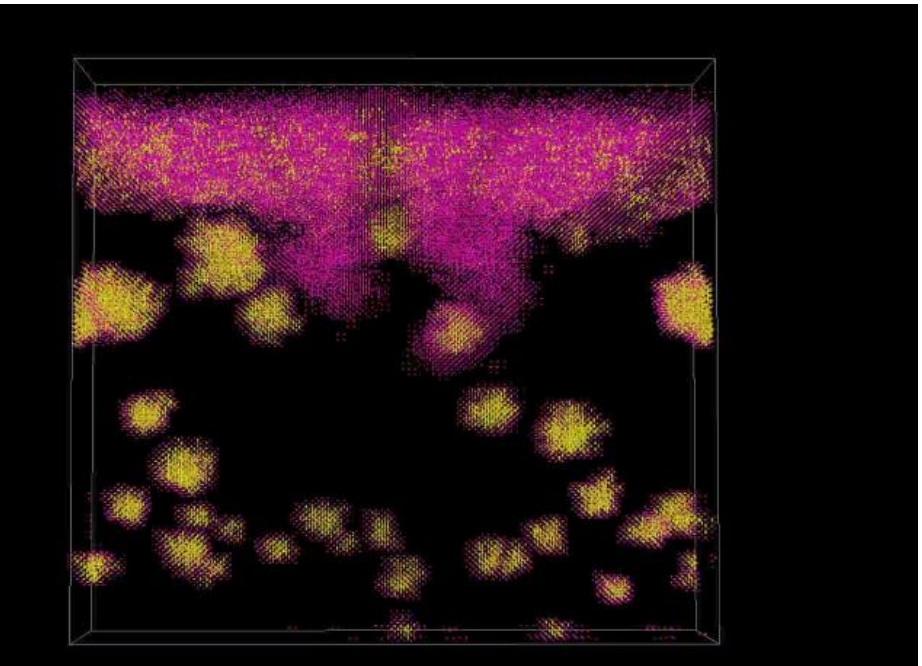
- He agglomeration generates the strain of W materials.
- The strain is released as a dislocation loop.
- The dislocation becomes the site for new He agglomeration.

How to represent the Fuzzy nanostructure formation



○ MD can represent the deformation of surfaces following atomic physical motion.

× MD cannot treat long time scale to represent the diffusion of helium. The surface region is unnaturally Peeled by no-diffused helium.

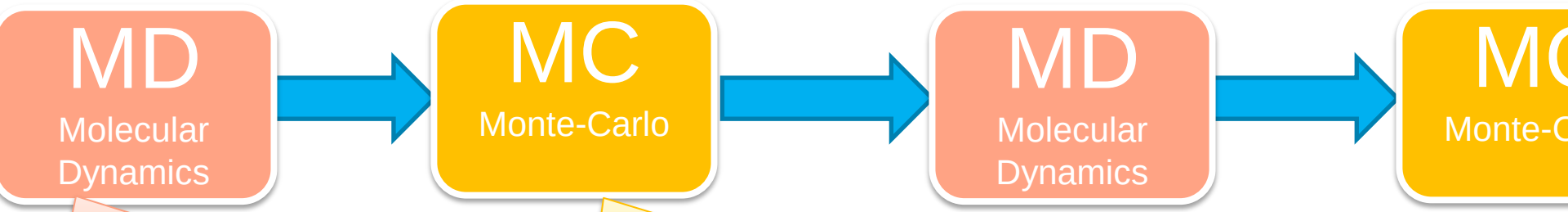


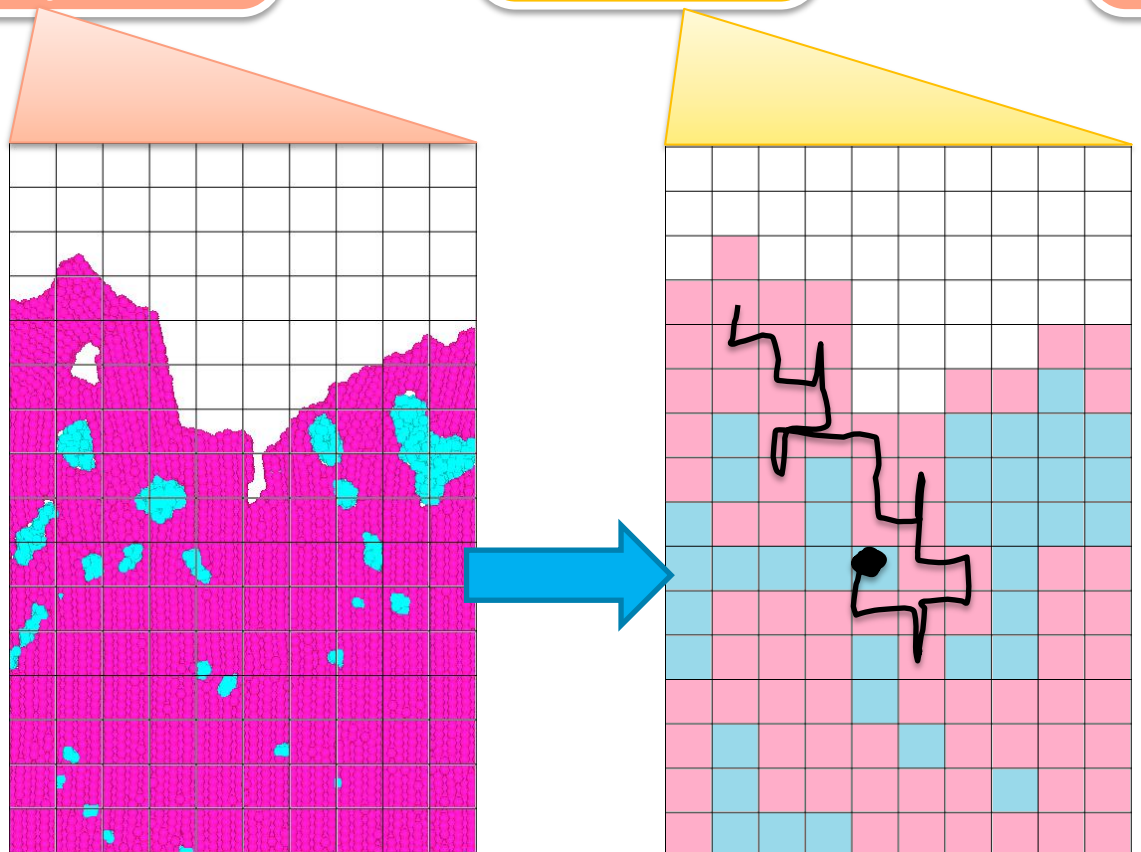
○ MC can treat long time scale to represent the diffusion of helium. And, the growth of a helium bubble can be represented.

× MC cannot treat the deformation of surfaces because MC ‘model’ is created only after we clarify the mechanisms of deformation.

MD-MC hybrid simulation system

- The diffusion of helium atoms is solved by lattice Monte-Carlo.
- The structure deformation of a material is simulated by molecular dynamics.
- This MD-MC cycle is repeated.





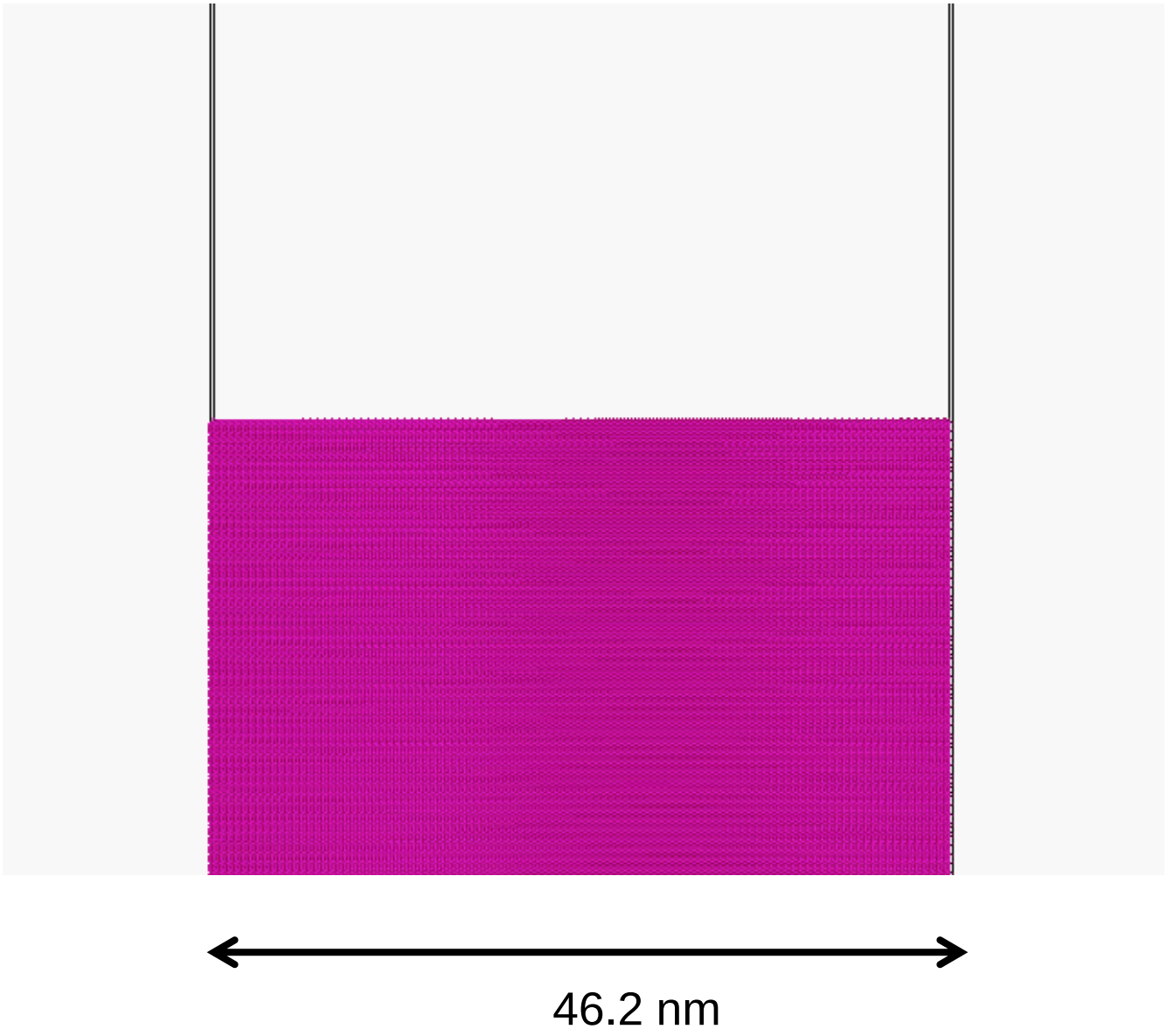
MC phase

- Lattice data is created from the structure in the previous MD phase.
- helium atom is diffused as random walk.

MD phase

- He atoms are doped at the position same to the previous MC phase.

Fuzzy Nanostructure Formation by MD-MC Hybrid Simulation



Temperature:
1500K

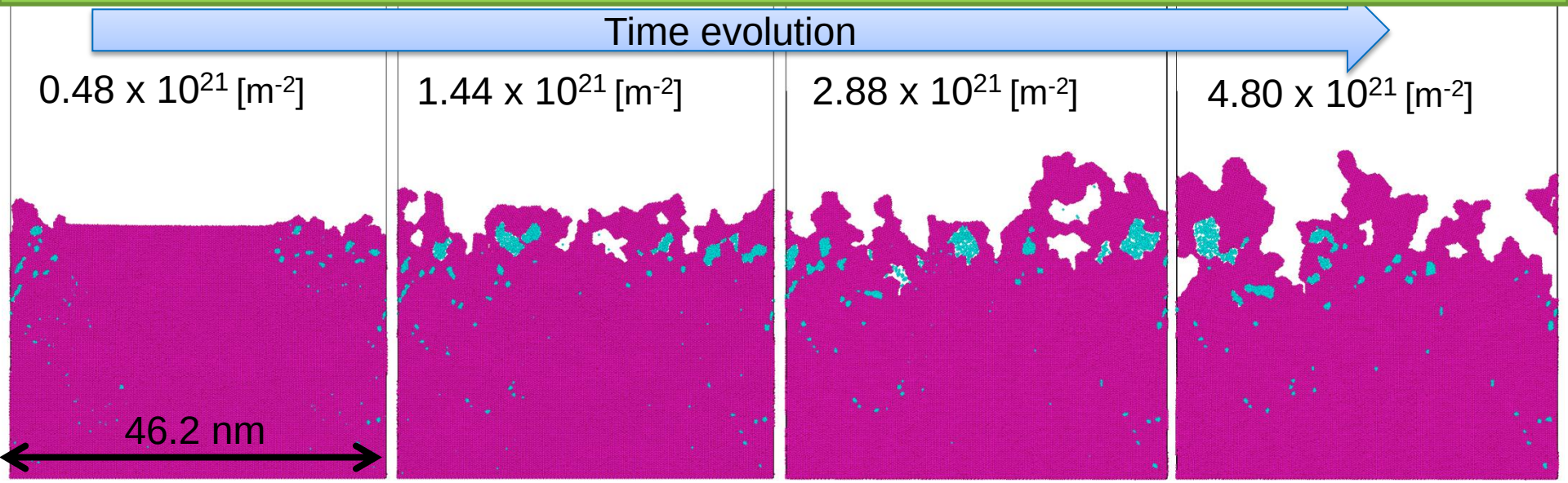
Flux for MC:
1.4×10^{22} $m^{-2}s^{-1}$

Flux for MD:
1.2×10^{30} $m^{-2}s^{-1}$

Diffusion coef.:
1.7×10^{-10} $m^{2}s^{-1}$

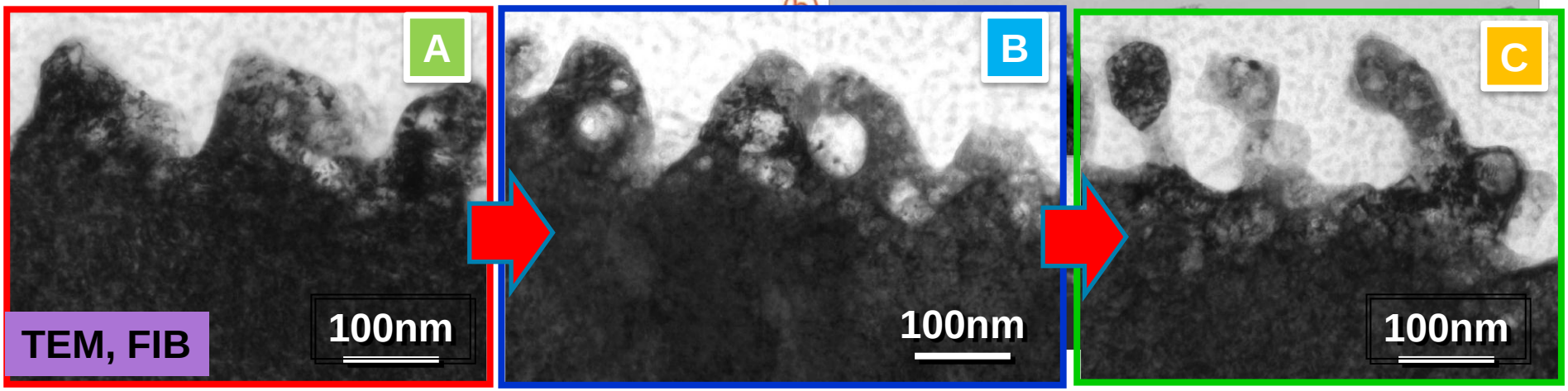
Penetration depth:
10 nm

Key mechanisms of fuzzy structure formation



Key mechanisms

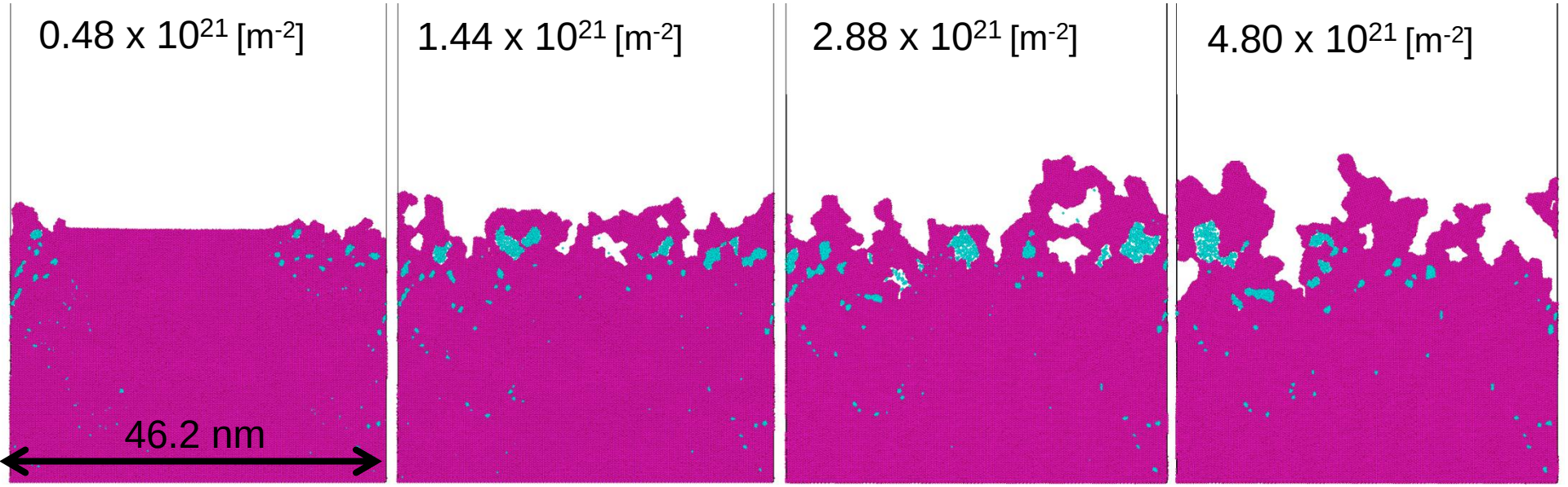
- The bursting of He bubbles
- The flip-up of tungsten surface



Acknowledgement N. Yoshida

*S. Kajita, et.al. APEX 3(2010) 085204

Comparable formation time scale as helium fluence



MD-MC doped He amount: 0.48 x 10^{22}[m^{-2}]

gap of 10^3

x 10 ⇒ 80% of helium are reflected by the surface in penetration process.

x 10^2 ⇒ By ejection from the surface in diffusion process, only 1-2% of helium atoms are retained, which are agreement between experiment* and kinetic MC.

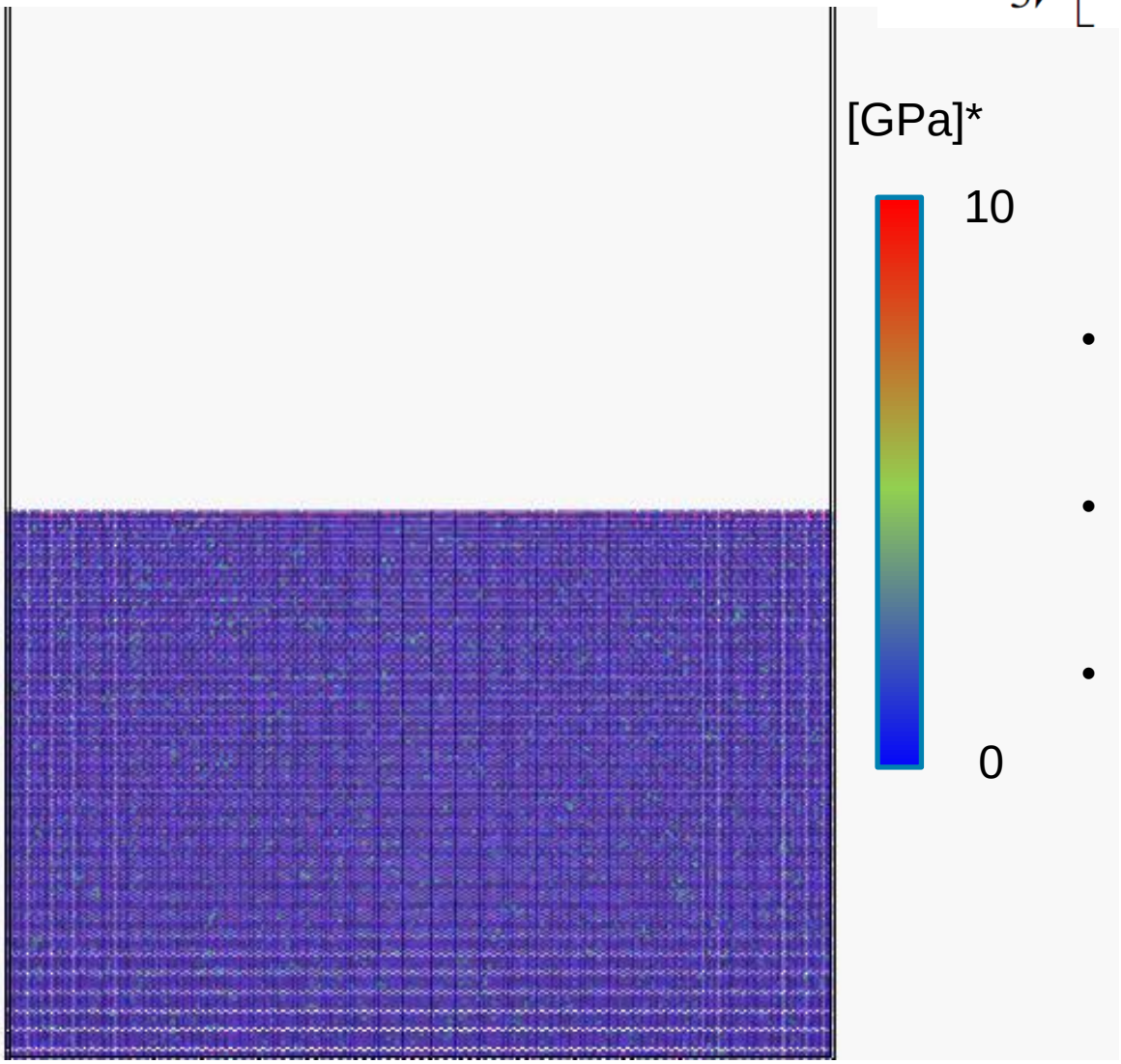
Un-treated processes in MD-MC:

Experimental fluence: 10^{24}~10^{25}[m^{-2}]

*H.T. Lee et.al., Trans. Fusion Sci. & Tech. 63 (2013) 233

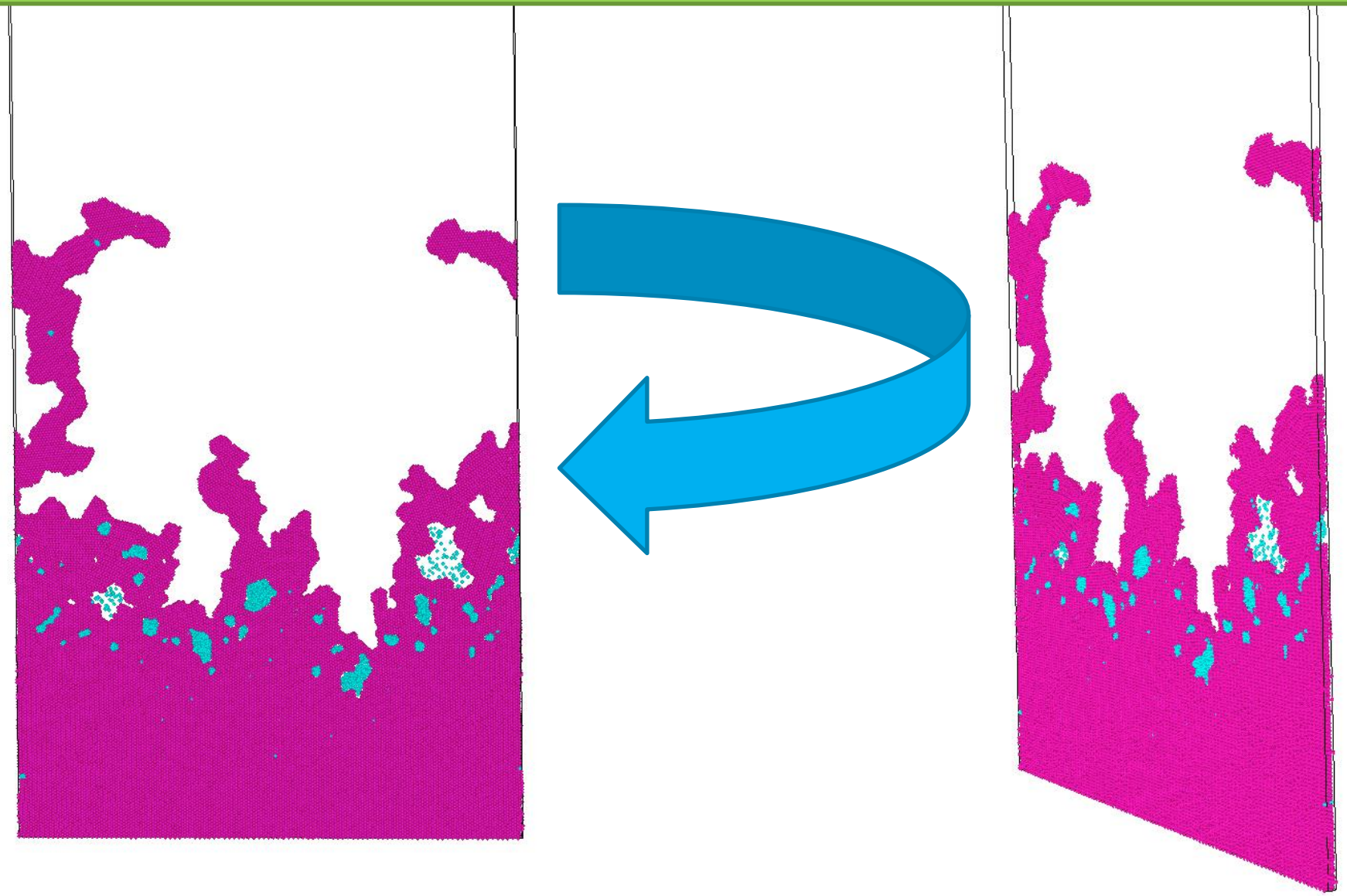
Local stress analysis

Local pressure: $$P(t) = \frac{1}{3V}\left[\sum_{i}^{\vec{r}_i \subset V} \vec{p}_i(t) \cdot \dot{\vec{r}}_i(t) + \sum_{i,k>i}^{\vec{r}_i, \vec{r}_k \subset V} \vec{f}_{ki}(t) \cdot \vec{r}_{ki}(t)\right]$$



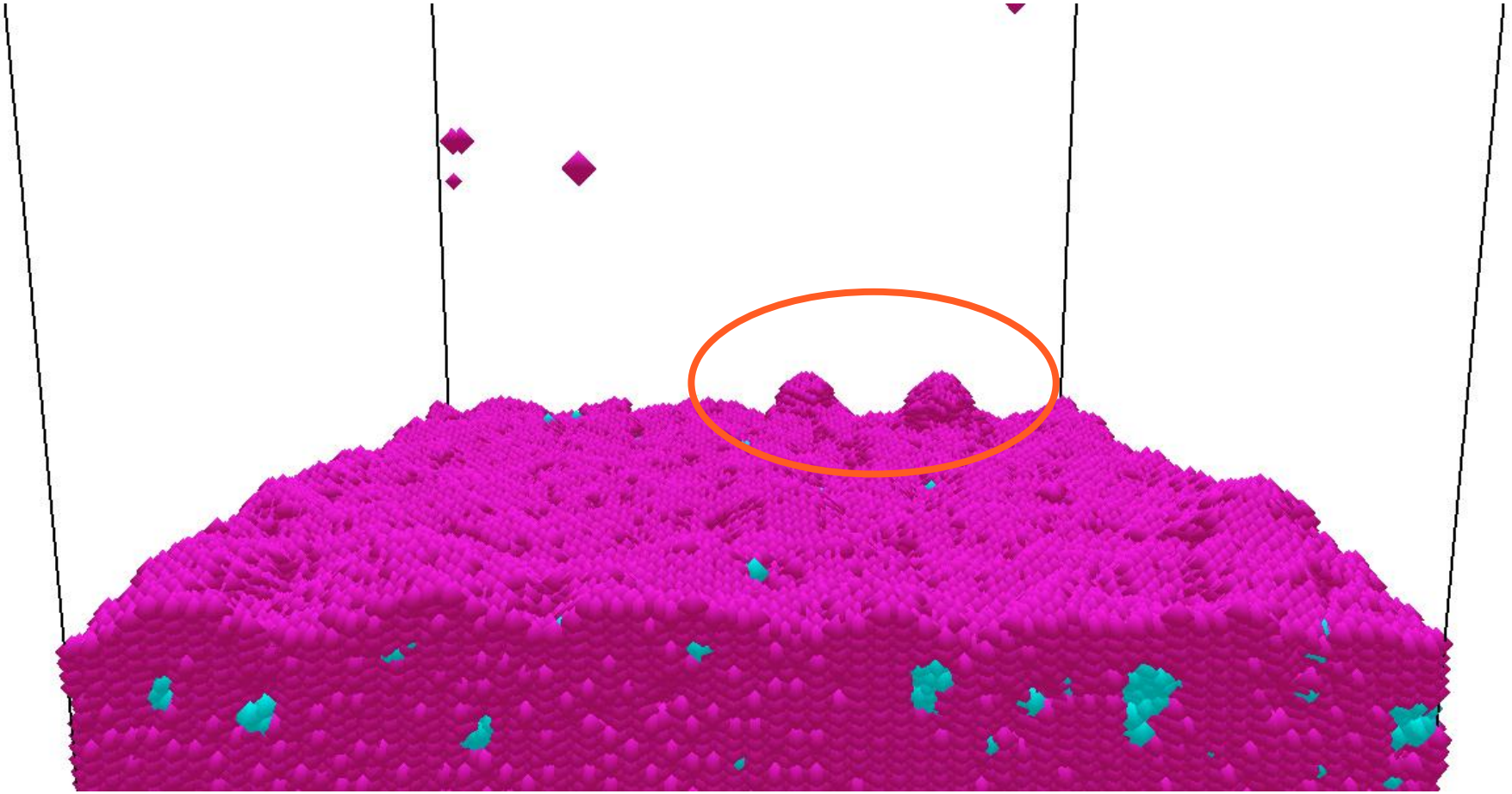
- The pressure in the He bubble is 5 GPa or more.
- The pressure in the tungsten region is lower.
- The bursting occurs when the pressure of a helium bubble reaches 5 GPa.

The present simulation system is psudo-3D space



The present simulation system is three dimensional, but the side in y-direction is thin.
The reason of that is just only to reduce calculation time to do many try & error.

Full 3D simulation has succeeded!!



The full 3D MD-MC hybrid simulation will be compared with experimental fuzzy nanostructure in surface fractal dimension.

Experimental fractal dimention is 2.2-2.6*

*S. Kajita et al., Phys. Lett. A 378 (2014) 2533–2538

Summary

- Fuzzy structure formation is represented by the four step process. The simulation method should be selected step by step:
  1. Penetration: BCA and DFT shows energy window to penetration.
  2. Diffusion and agglomeration: DFT shows that helium agglomeration is unlimited, agglomeration at interstitial site is possible, and the diffusion of helium is much faster than hydrogen.
  3. He-bubble growth : MD can represents the bubble in several nano-meter scale and loop punching.
  4. Fuzzy nanostructure formation : MD-MC hybrid simulation can represent fuzzy nanostructure formation.

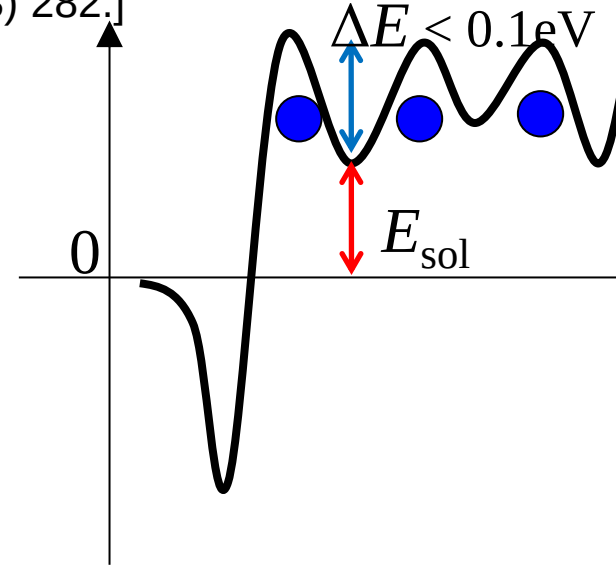
- Fuzzy nanostructure formation can be represented by the MD-MC hybrid simulation.

- Full 3D simulation for fuzzy nanostructure will be reported in the near future.

1. penetration proc. **Energy window for penetration**

- Lower limit of He penetration by DFT[2] well agrees with that measured by NAGDIS[1].
- Energy windows for Ne and Ar are much smaller, relate to Yajima's experiment[M. Yajima, etal, Plasma Sci. & Tech., 15 (2013) 282.]

element	lower limit [eV] (NAGDIS[1])	lower limit [eV] (DFT [2])	higher limit [eV] (BCA [3])
He	6.0	6.29	100
Ne		11.55	30
Ar		14.99	20
H		-2.47	700



[1] lower limit measured by experiment with NAGDIS:
D. Nishijima, M.Y. Ye, N. Ohno and S. Takamura: J. Nucl. Mater. 313-316, 97 (2003), 329-333, 1029 (2004) Proc. 30th EPS ECA 27A (2003) 2, 163.

[2] lower limit as the solution energy calculated by DFT(QMAS)
T. Tamura, R. Kobayashi, S. Ogata, A. M. Ito, Model. Sim. Mater. Sci. Eng., **22** (2014) 015002.

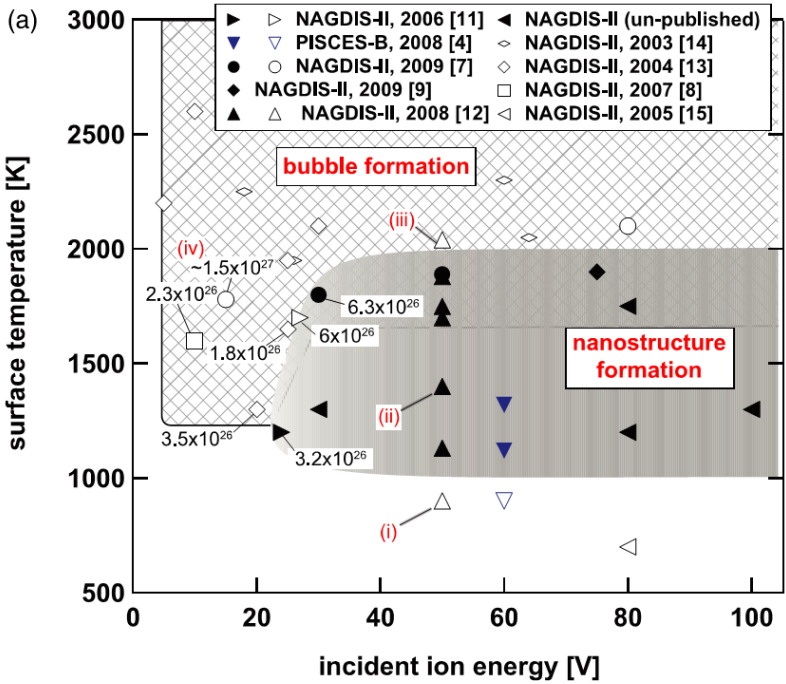
[3] higher limit as the sputtering threshold energy calculated by BCA(ACAT)
S. Saito, etal, J. Nucl. Mater. 438, (2013) S895–S898.

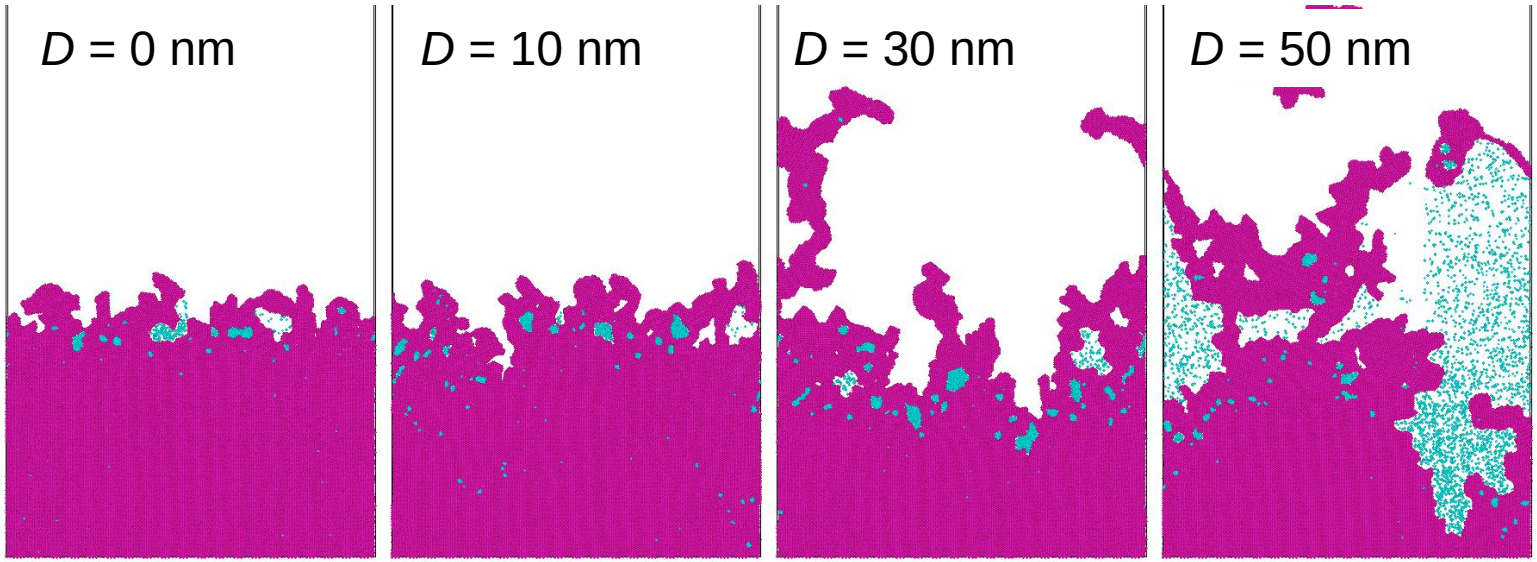
Dependence on penetration depth is confirmed

- Although penetration depth is nano-meter, which is estimated by BCA simulation, the difference of penetration depth in nano-meter is effective to fuzzy nanostructure formation in the MC-MD hybrid simulation.

- We now research the relation between the incident energy threshold to generate fuzzy nano-structure* and this dependence on penetration depth.

*S. Kajita, et al., Nucl. Fusion. 49 (2009) 095005

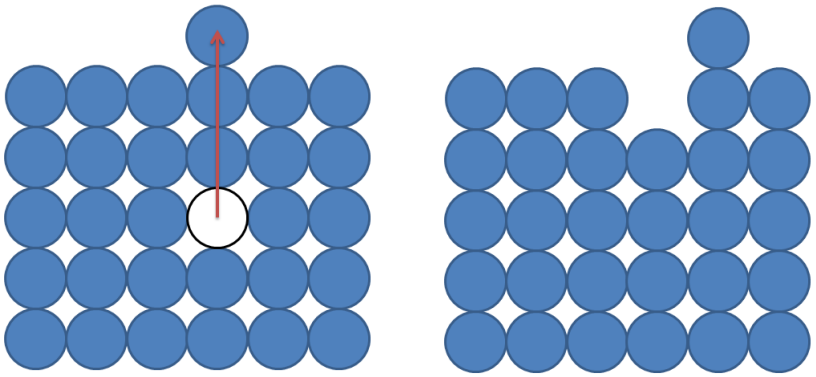




Comparison among penetration depth *D* at the doped He amount of 2.4 x 10^{21} m^{-2}

Growth of thickness can be explained as 1D random walk

The MC-MD hybrid simulation result shows the two works of helium bubbles: “push up” and “bursting”.



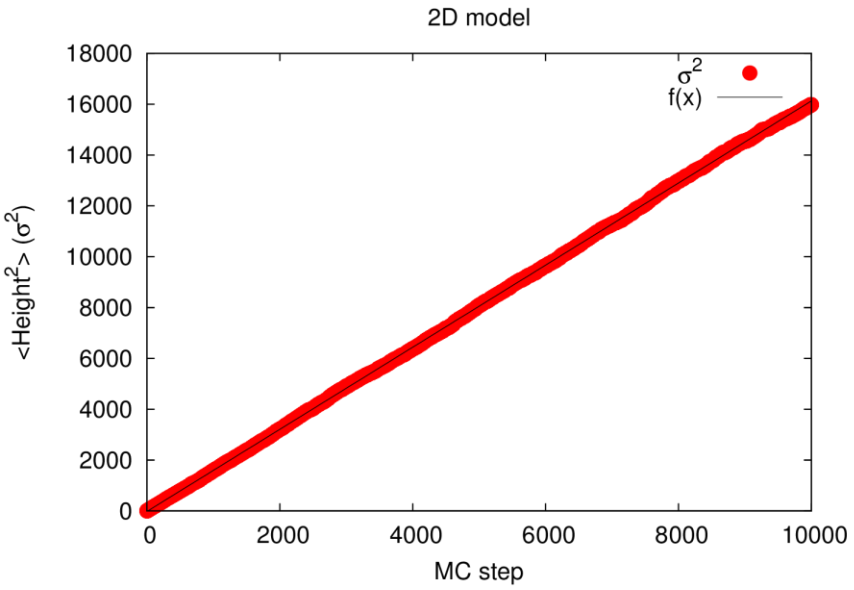
Murashima’s model

Growth of thickness is simplified as the occurrence of “Push-up” or “Bursting” on the surface at random.

The experimental fact that the thickness of fuzzy nanostructure *H* increase with square root of time*:

$$H \sim t^{1/2}$$

is explained as 1D random walk of mean thickness in the model.



Thickness of fuzzy nanostructure as a function of MC step in Murashima model

*M.J.Baldwin Nucl.Fusion48(2008)035001