

Unique phenomena of tungsten associated with fusion reactor: uncertainties of stable hydrogen configuration tapped in tungsten vacancy

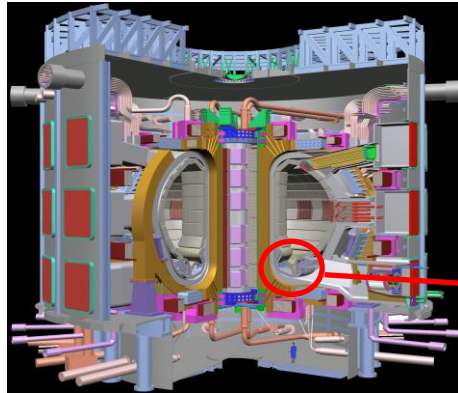
**Kyushu University
Kazuhito Ohsawa**

**Technical Meeting of the International Code Centre Network
Simulation of Plasma-Material Interaction Experiments
IAEA Vienna, 29-31 July 2015**

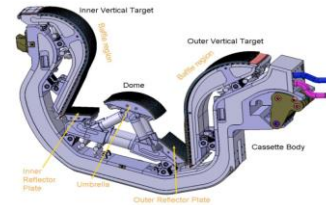
Out line

- 1. Introduction of unique phenomena of W as an example**
- 2. Abnormal H configuration in W mono-vacancy
application of random sampling**
- 3. Zero point energy calculations**

Back Ground of this work



ITER



divertor

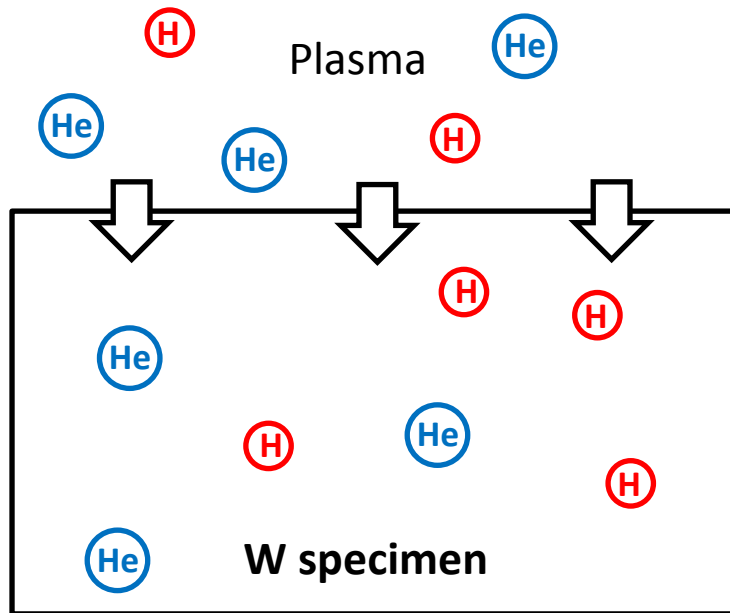
In fusion reactor (ITER), divertor armor is exposed to extremely large fluence of plasma particle , H isotopes D, T , helium (He), and heat flux. In order to protect the sever irradiation, tungsten (W) is considered as a plausible candidate for plasma facing materials in fusion reactor.

Advantage of tungsten as plasma facing materials

1. Highest melting point of all the pure metals 3380 °C
2. Low hydrogen solubility +1.1eV par H
3. Low sputtering erosion
4. High thermal conductivity
5. Low induced radioactivity, compared with molybdenum (Mo)

Some unique properties of tungsten are observed by the studies of fusion reactor materials.

Why do unique phenomena happen in tungsten associated with fusion reactor?
Of course, fusion reactor is an innovative technology.



Schematic view of H and He irradiation to W specimen

H and He compulsorily introduced in W

- 1. Negative formation energy of di-vacancy in tungsten**
- 2. He_2 migrates more quickly than a single He atom at $T < 400\text{K}$**
- 3. Fibreform tungsten covering surface by He irradiation**
- 4. Abnormal H configuration in W mono-vacancy**
- 5. etc.**

1. Negative formation energy of di-vacancy in tungsten

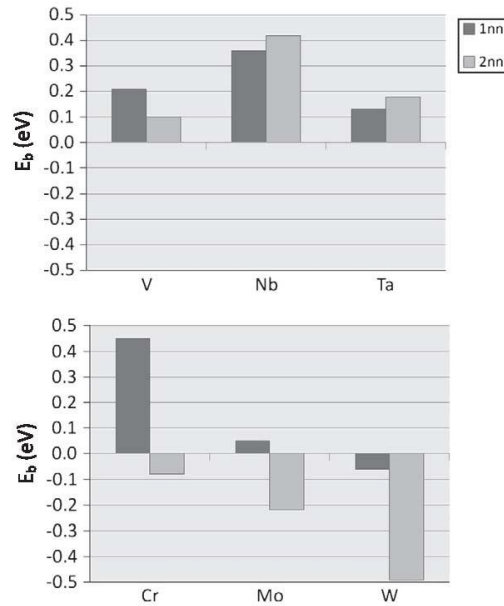


Fig. 5. DFT di-vacancy binding energies at first and second nearest-neighbors: comparison between the results obtained for groups VB (V, Nb, Ta) and VIB (Cr, Mo, W) bcc transition metals using PWSCF and 54-atom cells.

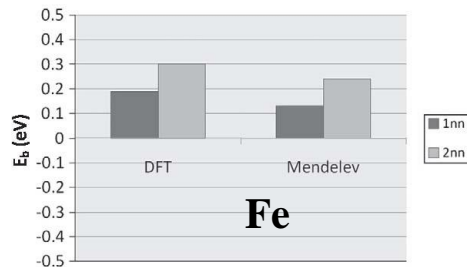


Fig. 4. Binding energies between two vacancies in Fe at first and second-nearest-neighbor positions: comparison between PWSCF results in a 54-atom cell and the Mendelev potential results [16]. The binding energy is defined as positive for an attractive interaction between the two vacancies.

2. He₂ migrates more quickly than a single He atom at T < 400K

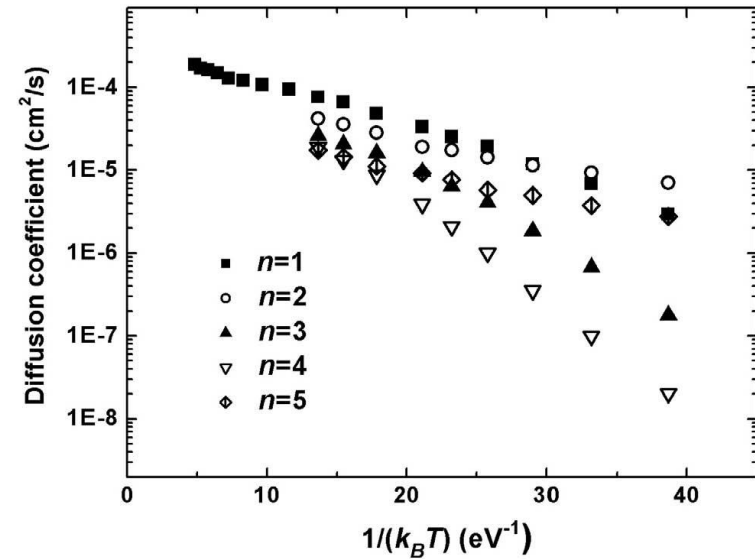


Fig. 8. Diffusion coefficient for He interstitials (He_n, $n = 1-5$) in tungsten.

Y.L. Zhou *et al.*

J. Nucl. Mater. 446 (2014) 49

L. Ventelon *et al.*

J. Nucl. Mater. 425 (2012) 16

3. Fibreform tungsten covering surface by He irradiation

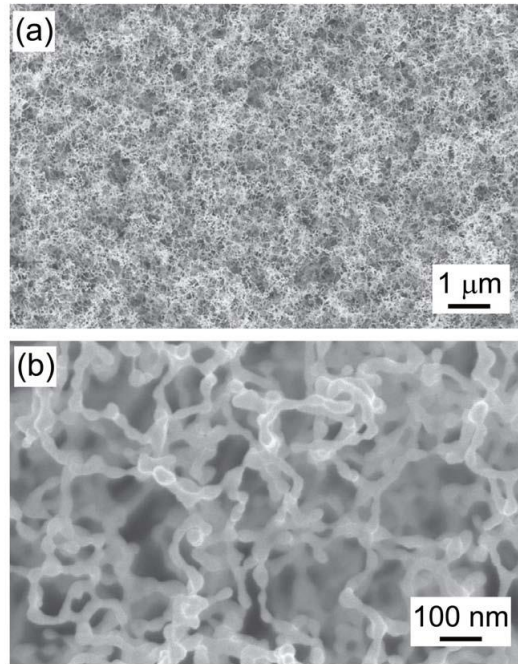


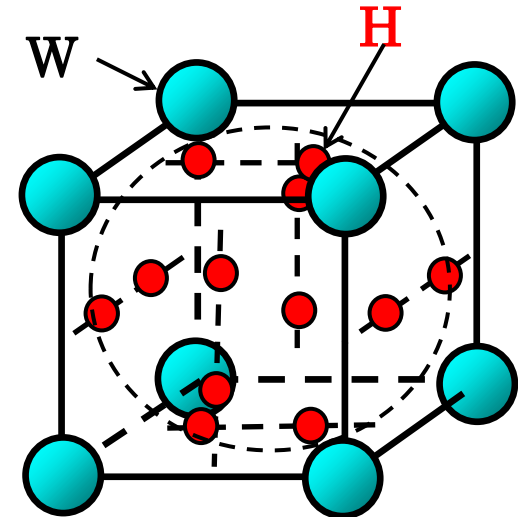
Figure 2. (a) and (b) are SEM micrographs of the electrode surface exposed to the helium plasma in different scale sizes. The surface is covered with network structured fibreform tungsten.

S. Kajita *et. al.*

Nucl. Fusion 49 (2009) 032002

Nucl. Fusion 49 (2009) 095005

4. Abnormal H configuration in W mono-vacancy



K. Ohsawa *et. al.*

Phys. Rev. B 82 (2010) 184117

Phys. Rev. B 85 (2012) 094102

I would like to talk about this topic.

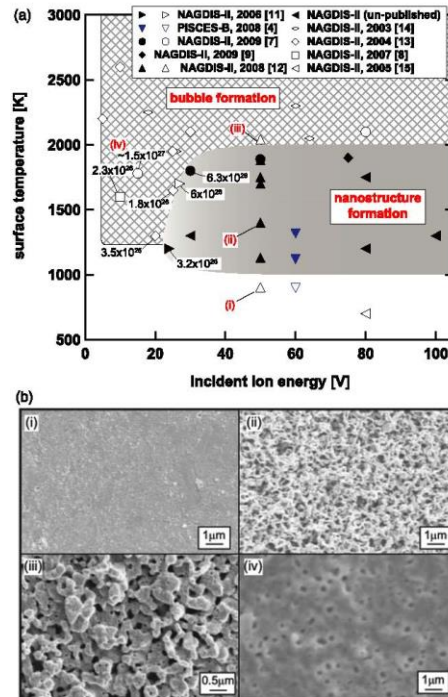


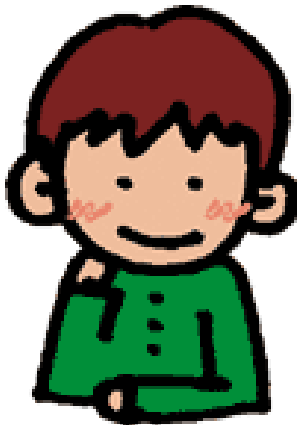
Figure 1. (a) The surface temperature was plotted against the incident ion energy for helium irradiation experiments conducted in the divertor simulator NAGDIS-II [7, 10–15] and PISCES-B [4]. Closed markers represent the cases in which the nanostructure was formed, while open markers represent the cases where the nanostructure was not observed. (b) SEM micrographs under the conditions of (i)–(iv) shown in (a).

Main subject

Abnormal stable configuration of H atoms in W mono-vacancy

How many hydrogen atoms are accommodated in a vacancy in tungsten and other bcc transition metals ?

What is abnormal stable configuration of Hydrogen in vacancy ?



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Ionic compound
 hydrogen storage type $\Delta H < 0$
 anti hydrogen storage type $\Delta H > 0$
 covalent compound

H

Group V , VI, and Fe

BCC metal

BCC metal

Simulation method

First-principle calculation

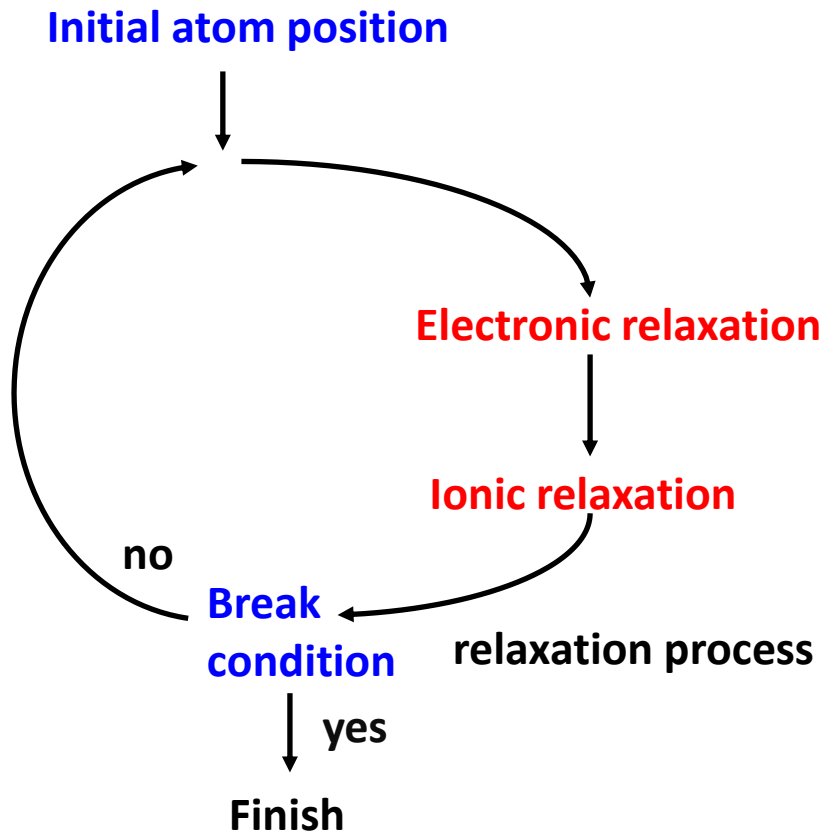
Vienna ab-initio simulation package (VASP)

Perdew-Burke-Ernzerhof (PBE) potential

Super cell size 3x3x3 (54) bcc lattice For W, 5x5x5 (432) lattice is also used

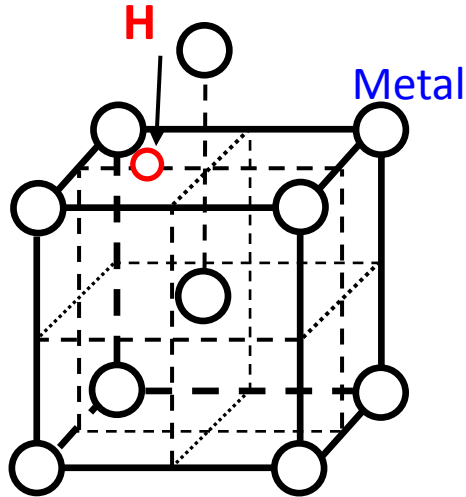
Plane wave cut-off energy is 350 eV

Atomic relaxation are iterated until a break condition is satisfied ($0.003\text{eV}/\text{\AA}$)

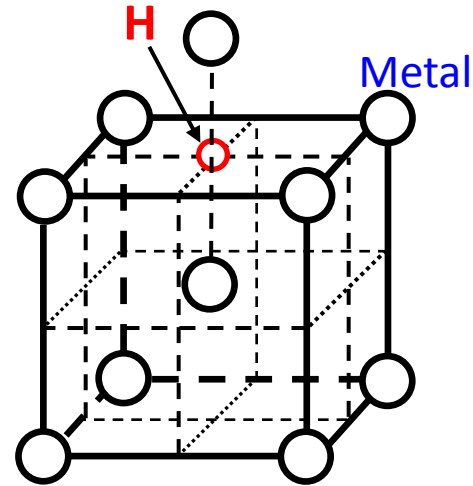


Review of Hydrogen in BCC metals

Interstitial H atom prefers T-site in bulk BCC metals.



**Tetrahedral interstitial site
(T-site)**



**Octahedral interstitial site
(O-site)**

T-site is more favorable for H than O-site.

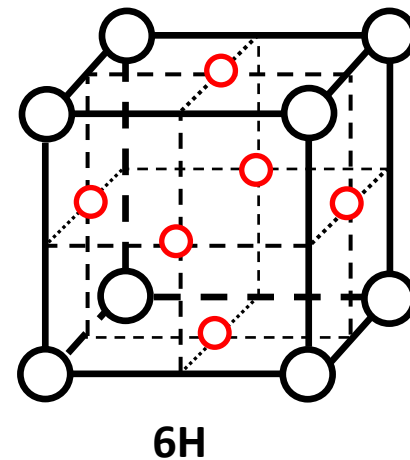
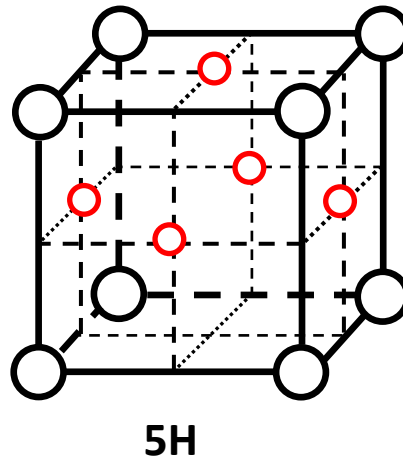
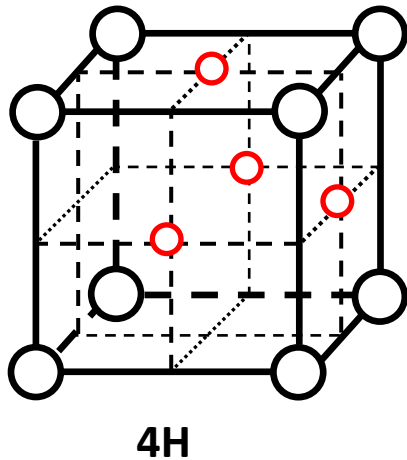
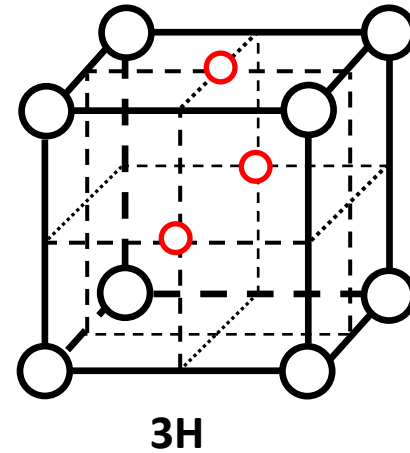
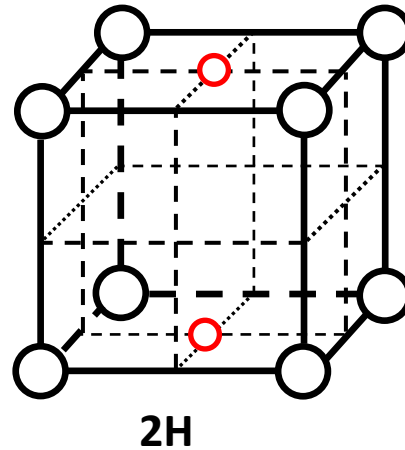
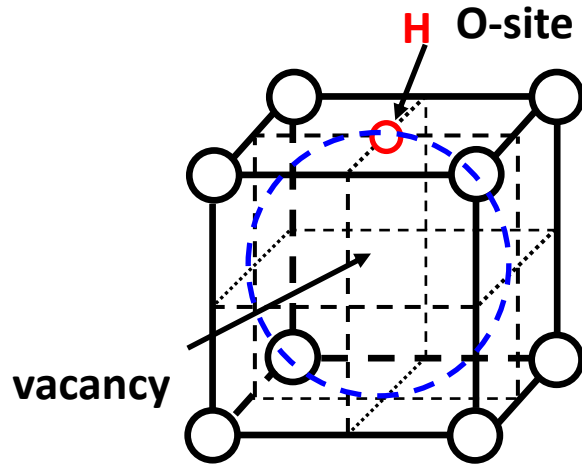
W 0.380eV

Mo 0.265eV

Fe 0.131eV

Nb 0.271

normal stable configuration of H atoms in mono-vacancy in bcc transition metals



Usually, a maximum of 6 H atoms are trapped.

6H's regular tetrahedral configuration

This was established theory but we decided to investigate stable configuration more carefully

Initial H atom positions for ionic relaxation

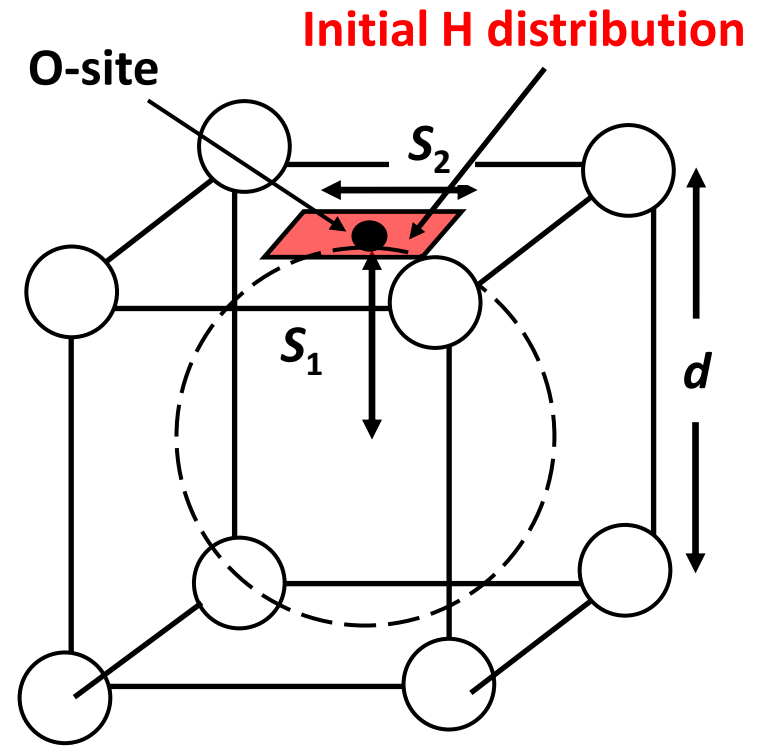
H atoms are located inner surface of vacancy.

Initial configuration for H atoms are randomly generated within a area.

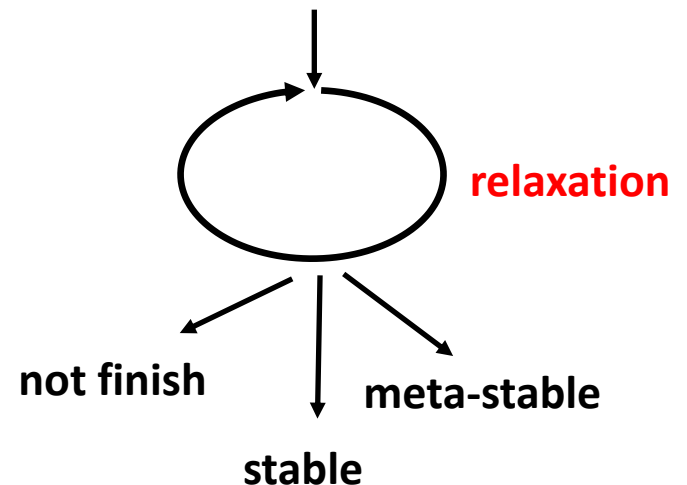
Usually, $S_1=0.4d$, $S_2=0.2d$

The purpose of meeting is **uncertainty quantification**. This is an application of the **random sampling**.

About 30 % initial configurations arrive at true ground-state structure.



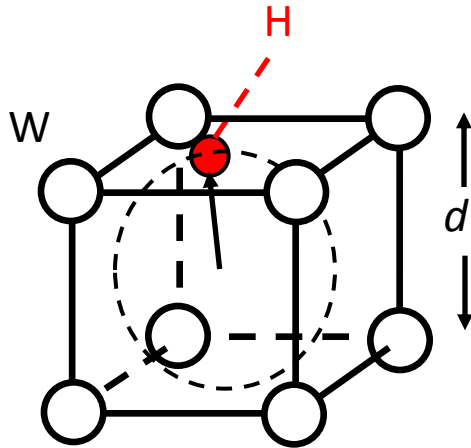
Many Initial atom position



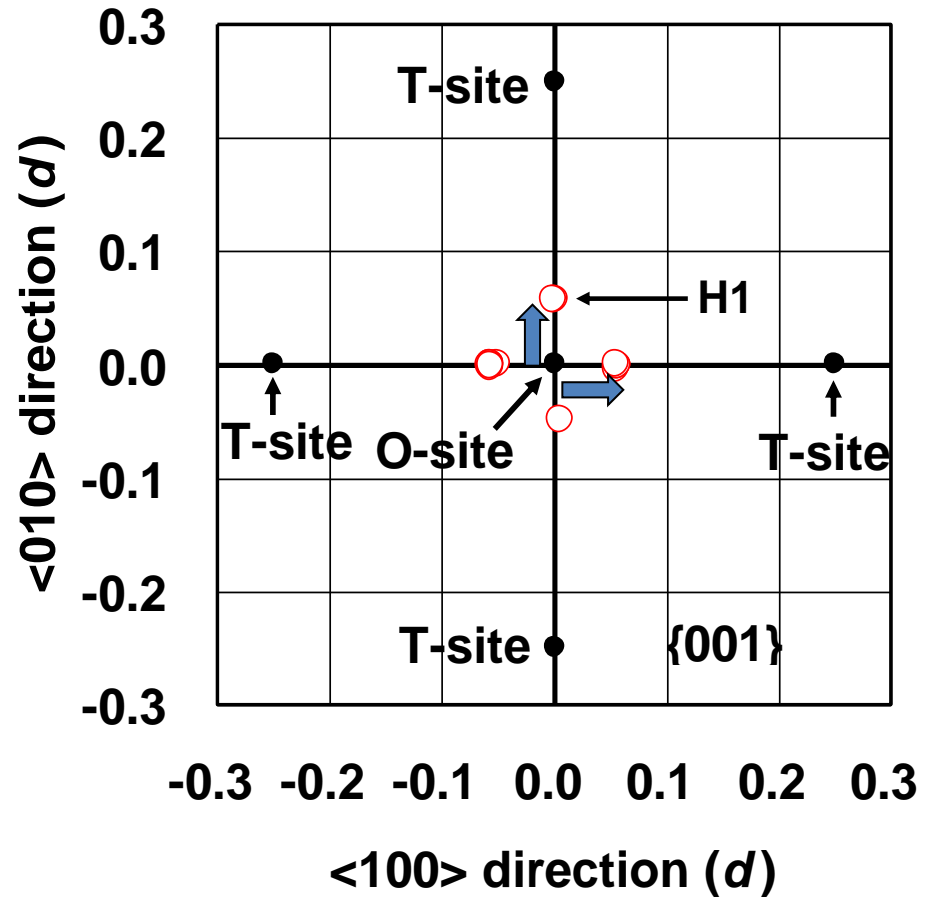
Results of application of random generate initial configuration

(a) Stable configurations of multiple H atoms in W mono-vacancy

【single H atom in monovacancy】

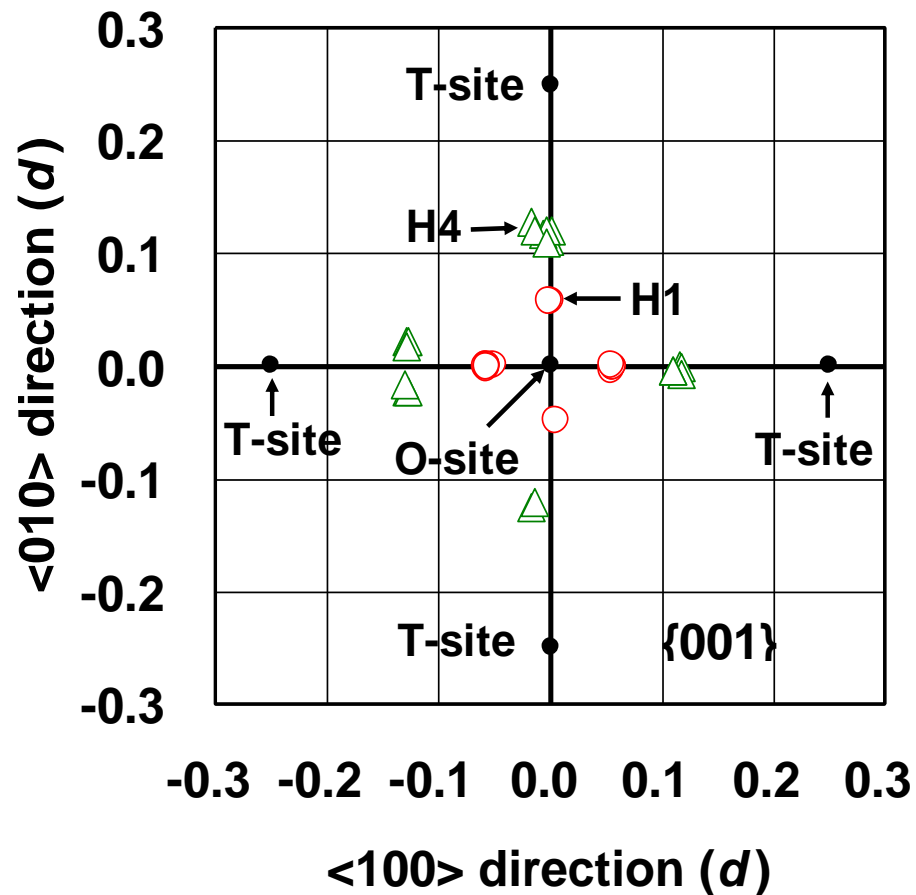
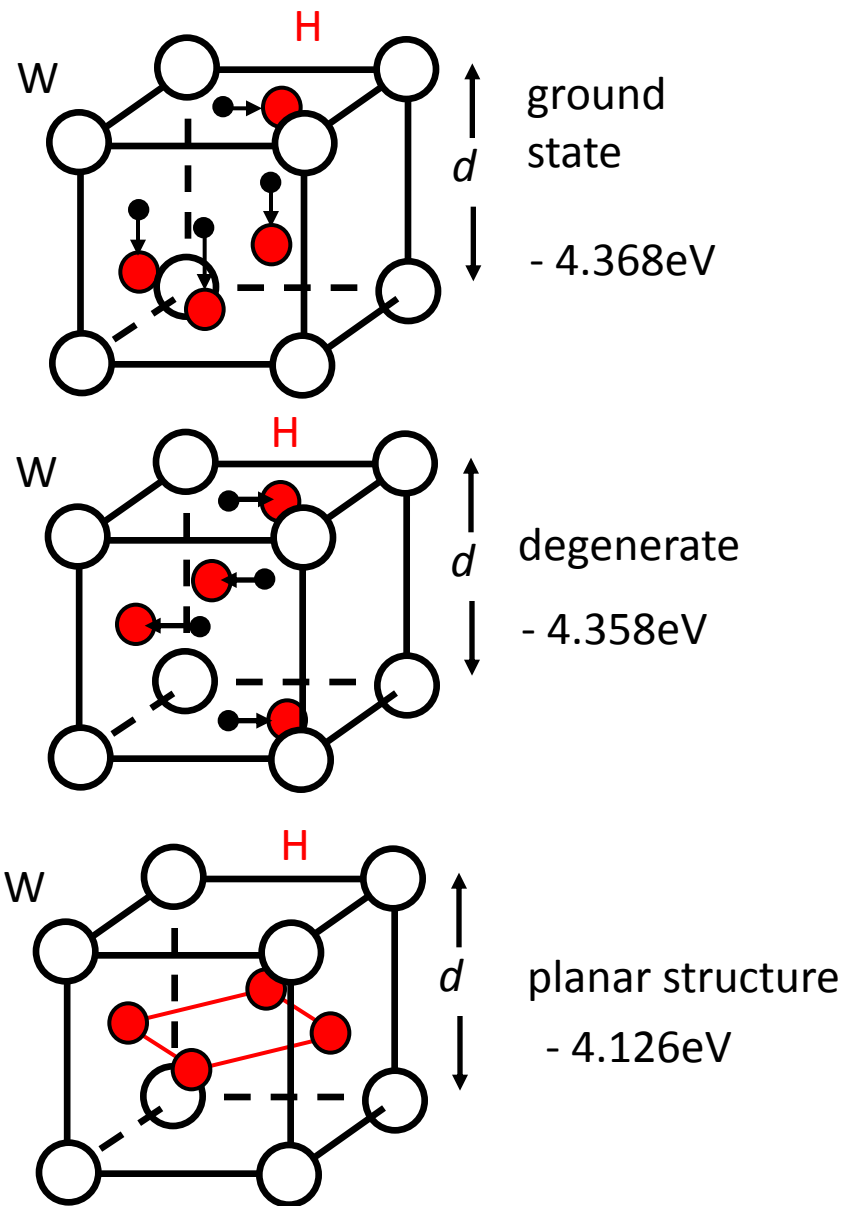


A single H atom is located close to O-site on inner surface of a monovacancy. But

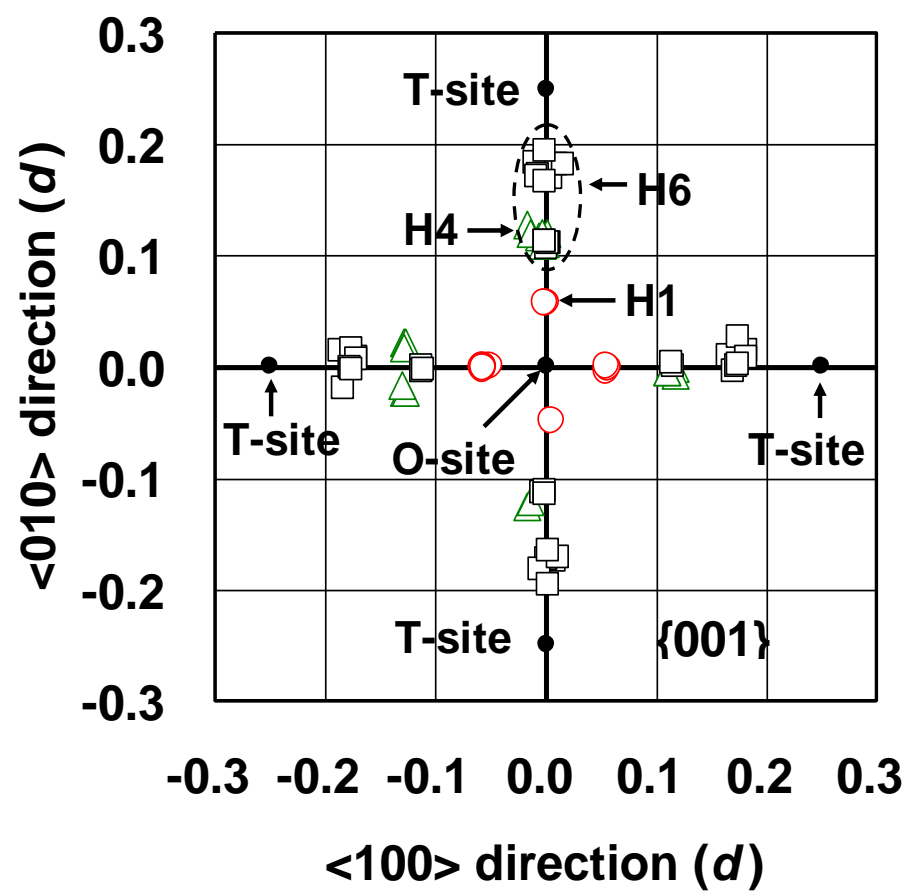
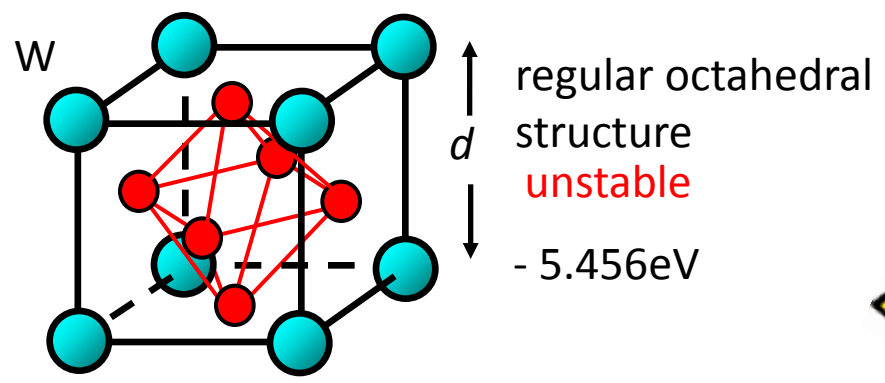
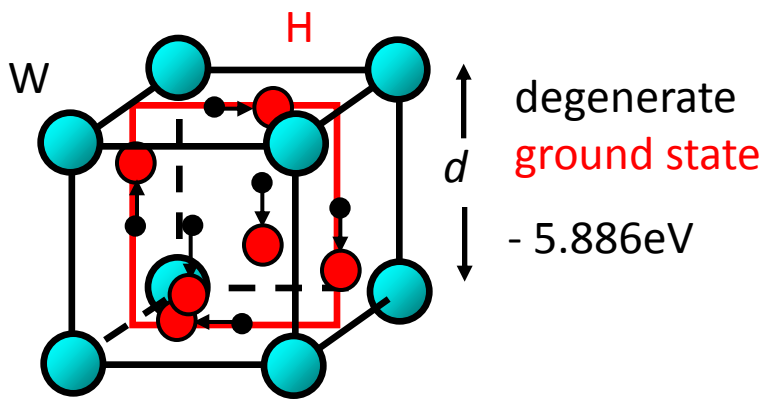
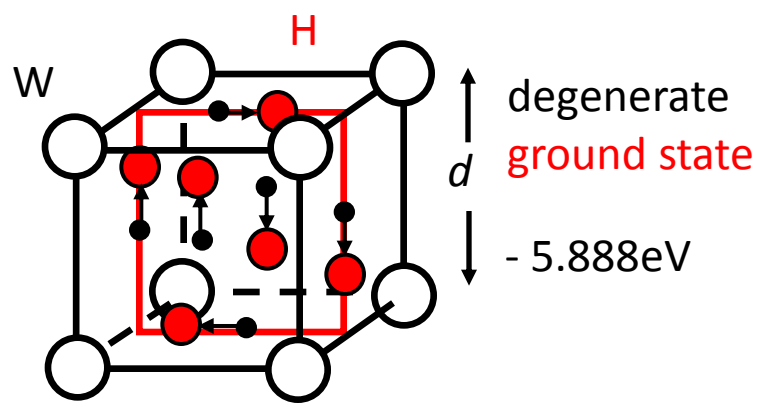


stable positions of H projected onto the {001} plane

【4 H atom in monovacancy】

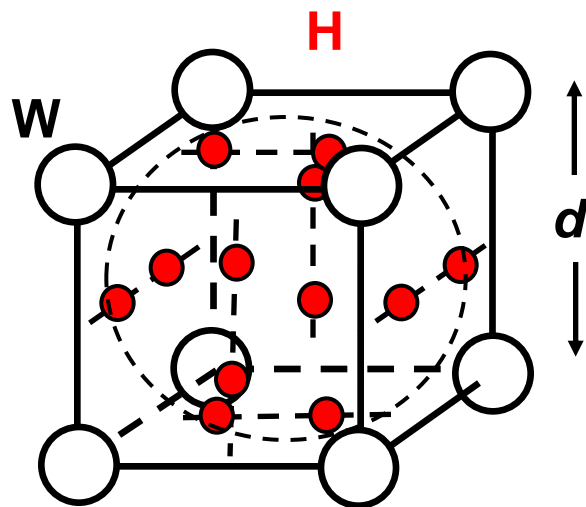


【6 H atom in monovacancy】



Stable positions for H are gradually shifted toward the T-site.

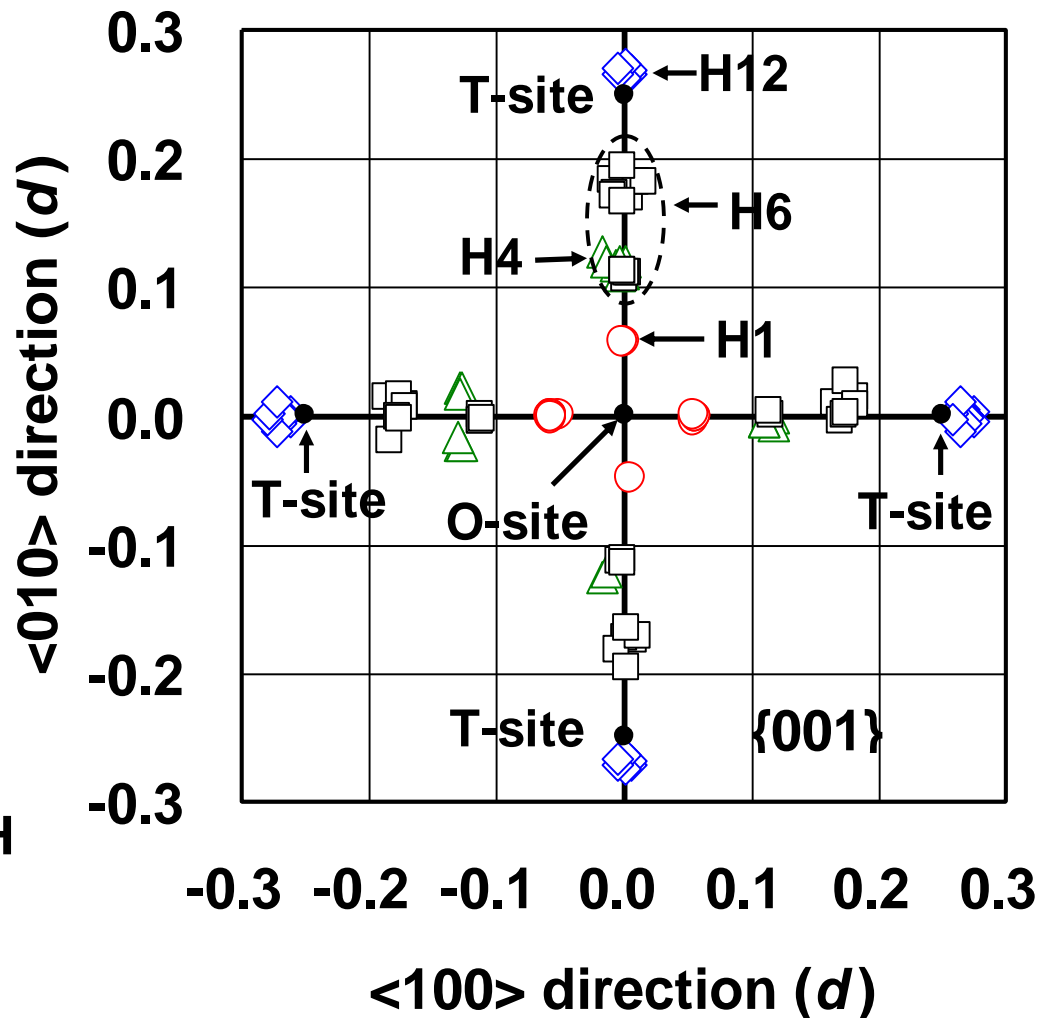
【12 H atom in monovacancy】

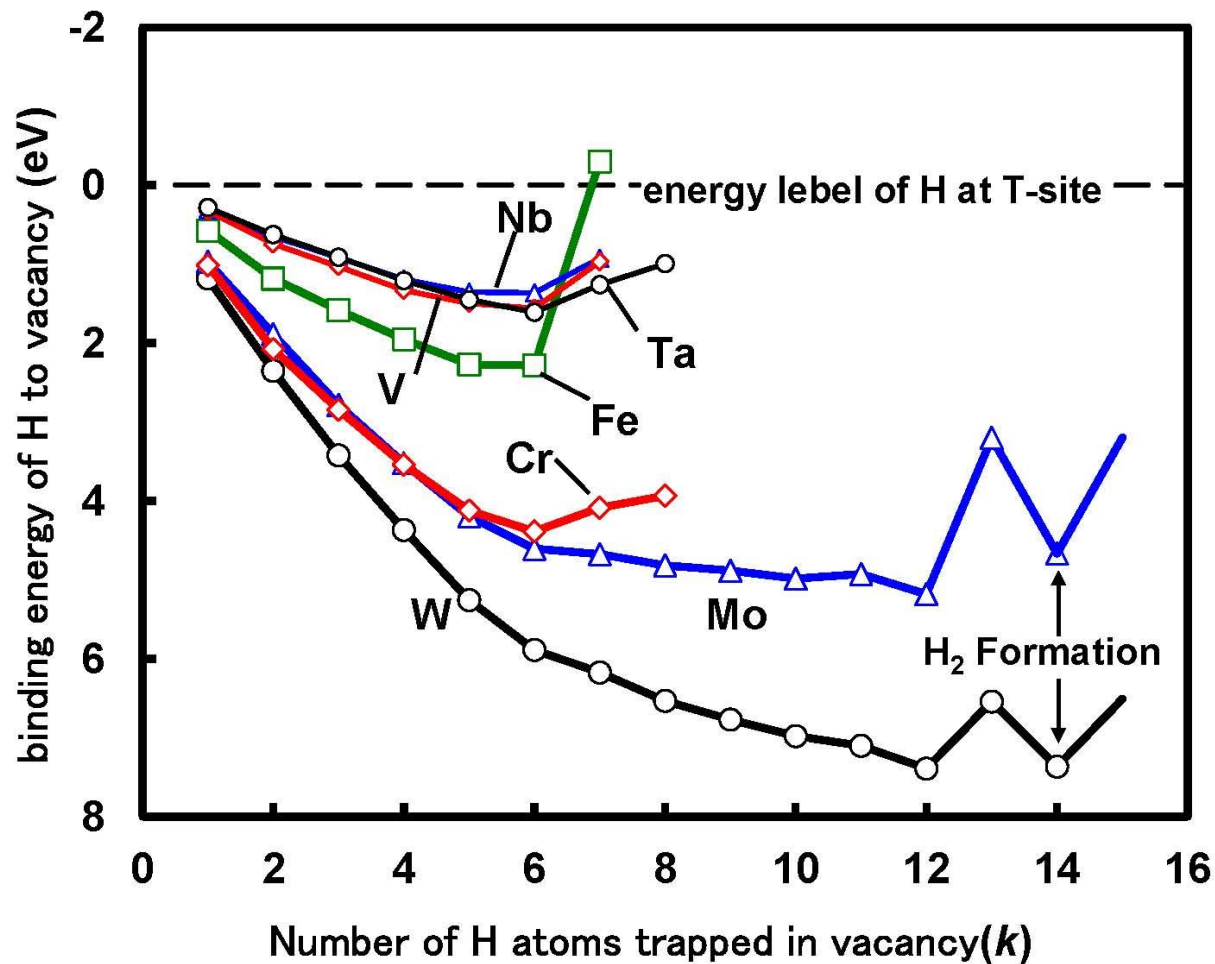


ground state

-7.391eV

Repulsive interaction of H





Finally, we would like to answer the first question.

The total binding energy of multiple H atoms to metal vacancy

$$e_k = E[M_{n-1}V] - E[M_{n-1}VH_k] + k(E[M_{n-1}H^T] - E[M_n])$$

Short summary of the simulations

Group 5 transition metals (**V**, **Nb**, **Ta**) and **Cr** and **Fe**

A maximum of H atoms trapped in mono-vacancy is 6.

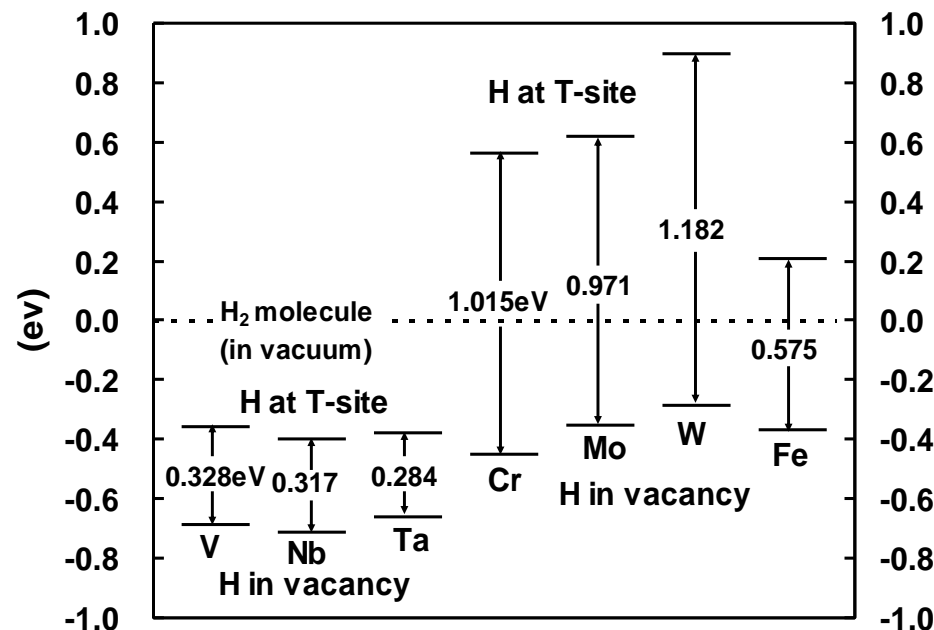
Normal stable configuration of the multiple H atom.

W and **Mo** vacancy can accommodate a maximum of 12 H atoms.

H configurations are abnormal .

New question

Why are so many H trapped in W and Mo vacancy?



H release large energy when trapped in vacancy.

W and Mo have large vacancy.

lattice constant

W 3.16 Å

Mo 3.15 Å

Cr 2.88 Å

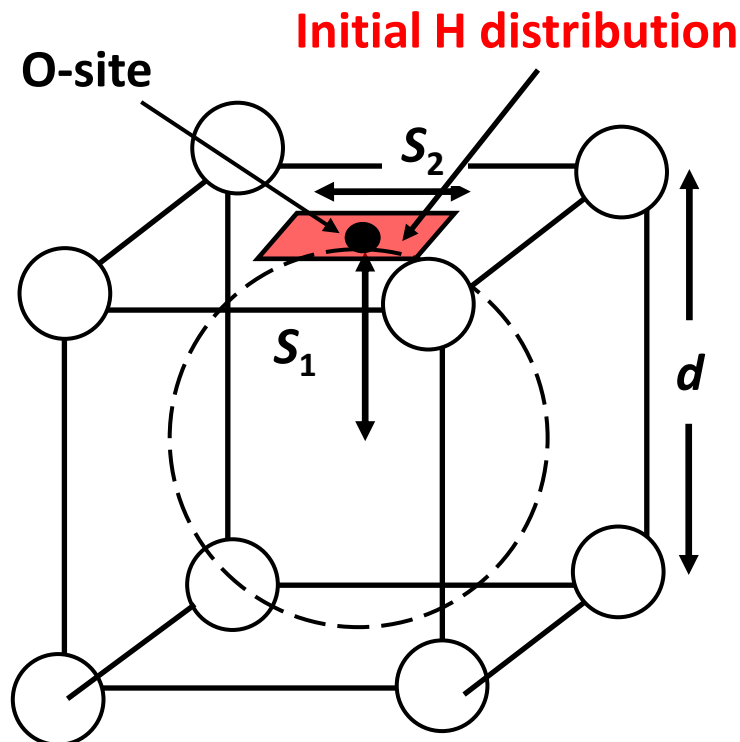
Fe 2.85 Å

What is relation of our works and uncertainty quantification?

Main purpose of this meeting is **uncertainty quantification** (UQ) activities.

Initial positions of H atoms are determined by assuming a random distribution.
This is an important application of random sampling.

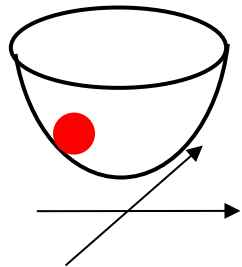
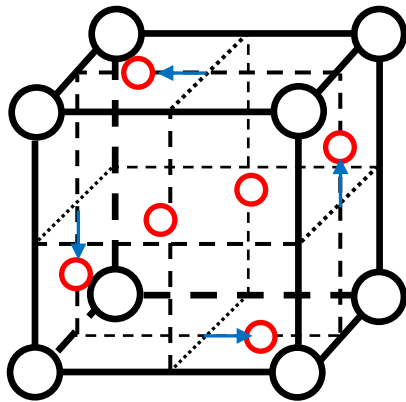
In the present works, we determine the parameter S_1 and S_2 appropriately.
But if we perform more complicated simulations, we have to do same optimization of the method.



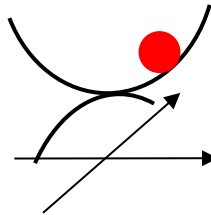
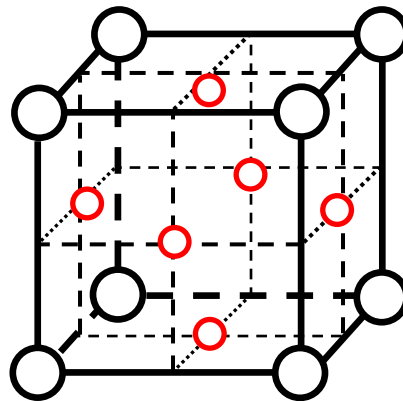
Application: zero point energy calculation



Abnormal H configurations are applied to calculations of zero point energy.



Ground state



Saddle point = unstable

Schematic view of H vibration

We can calculate vibration energy from abnormal H configuration.



Summary and conclusion

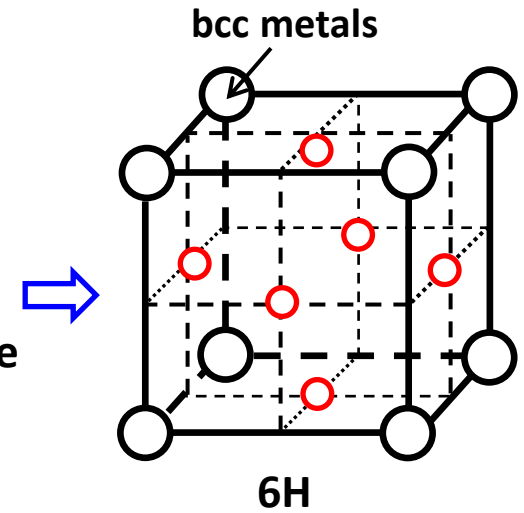
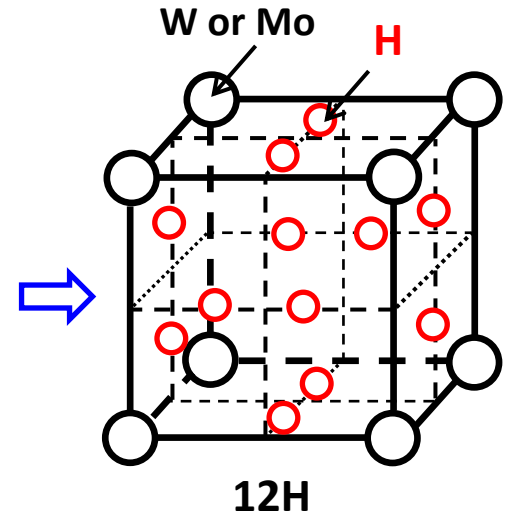
1. The abnormal H atom configurations are found in W and Mo mono-vacancy by first-principle calculations.

2. A maximum of 12 H atoms can be accommodated in the vacancy.

3. The abnormal H configurations can be applied to calculations of zero point energy.

4. H configurations in mono-vacancy of other bcc transition metal (V, Nb, Ta, Cr, Fe) are normal ones.

5. A maximum of 6 H atoms are accommodated in it, which are in good agreement with established theory.



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