2015 Second Research Coordination Meeting on Plasma-wall Interaction with Irradiated Tungsten and Tungsten Alloys in Fusion Devices

September 8-11, 2015 Seoul, Korea

Modeling and Simulation of Helium and Hydrogen Behaviors in Tungsten

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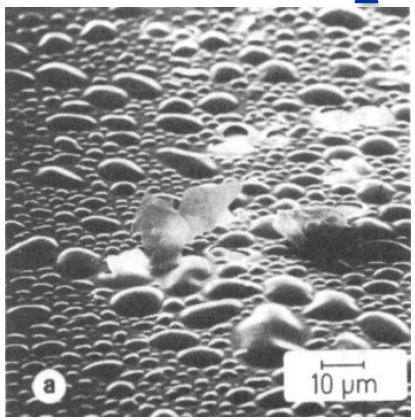
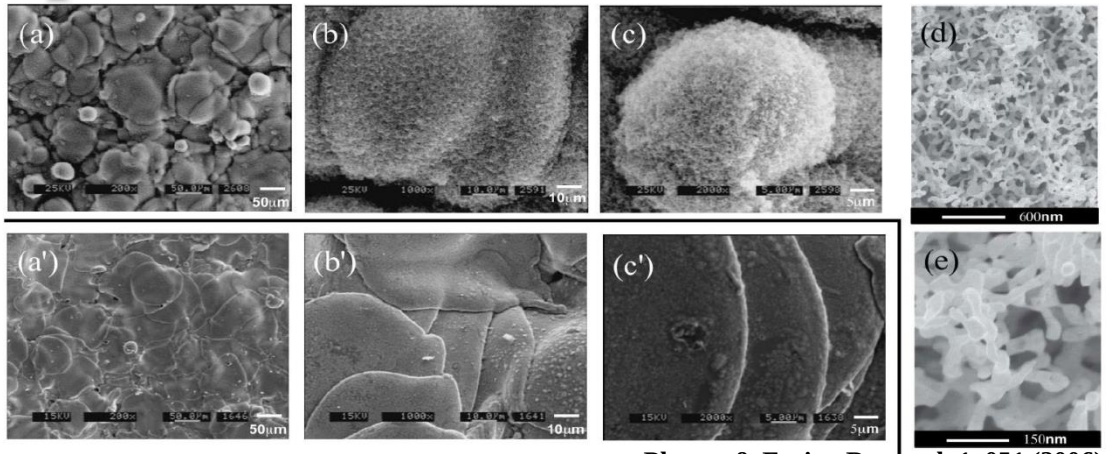
Outline

- Background
- Computational methods
- Main results
- Summary

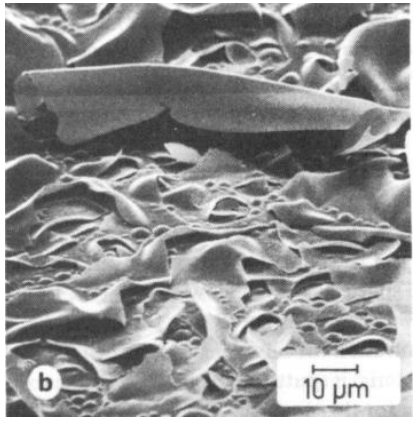
- **Stress tensor: A quantitative indicator of effective volume and stability of helium in metals**
- **Electrophobic interaction: the leading mechanism for helium self-trapping in tungsten**
- **Anisotropic strain enhanced hydrogen solubility in tungsten**

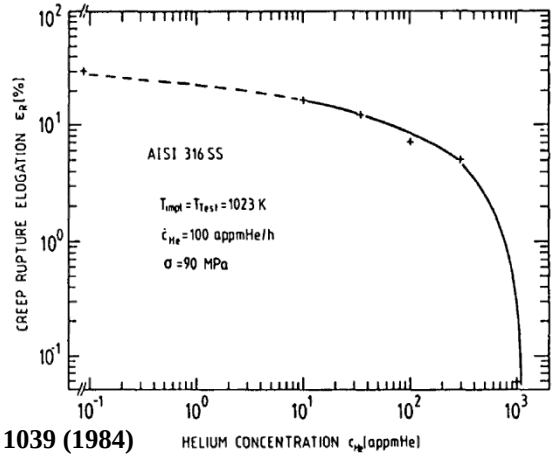
Effects of helium on the microstructure and mechanical properties of metals



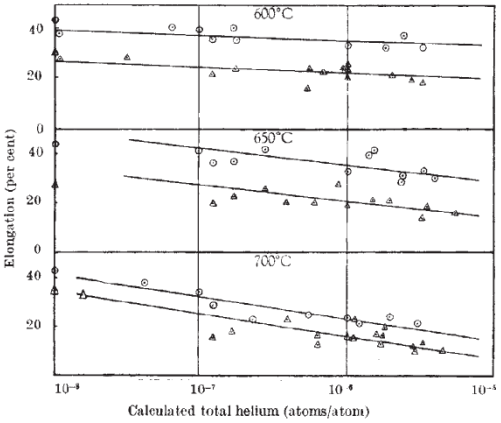


Plasma & Fusion Research 1, 051 (2006)





Nuclear Fusion 24, 1039 (1984)



Nature 211, 291 (1966)

- Helium irradiation has significant effects on the microstructure and mechanical properties of metals.
- The effects of He are directly related to the He-He strong attraction in metals.

Self-trapping of He in metals

- The He-He strong attraction is referred as **self-trapping** of He in metals.

W.D. Wilson, *et al.* Phys. Rev. B 24, 5616 (1981).

- It has been demonstrated that the He-He binding energy is ~1.02 eV in W with the equilibrium distance of ~1.5 Å.

C.S. Becquart, *et al*. Phys. Rev. Lett. 97, 196402 (2006).

Despite large amount of He-related investigations, the physical intrinsic mechanism for He self-trapping is still not fully understood.

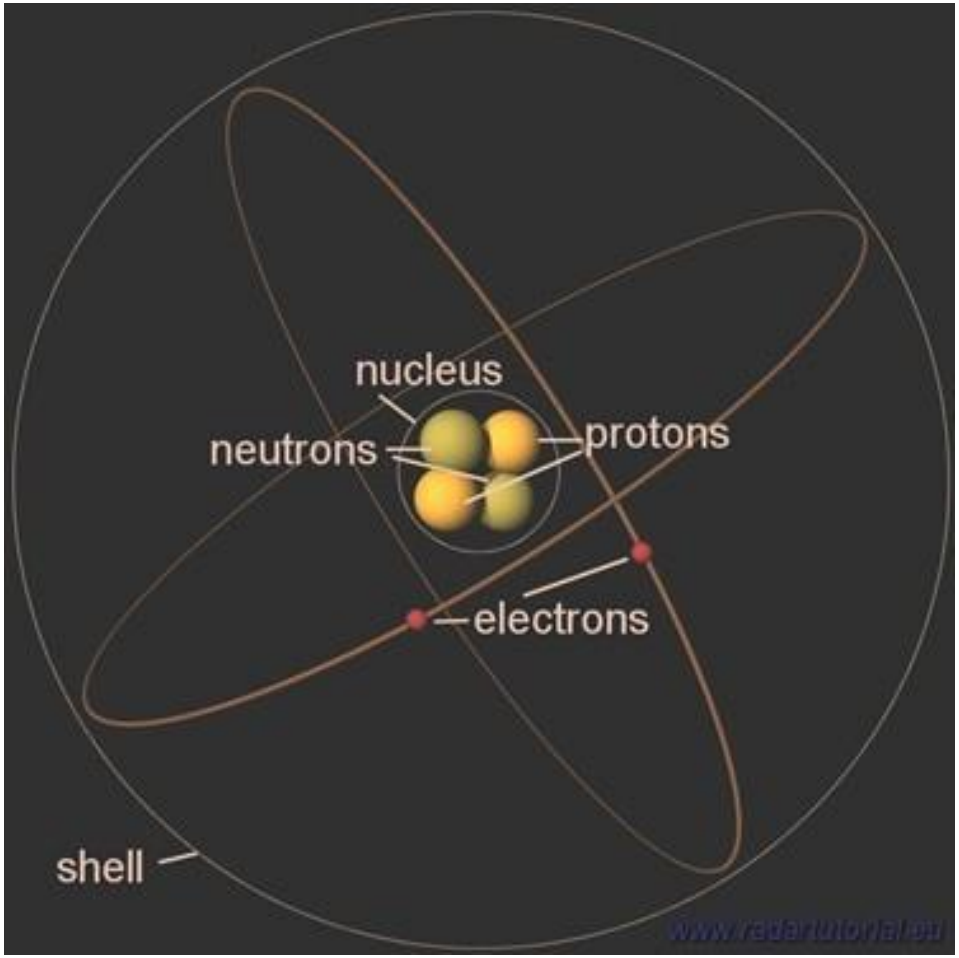
Computational methods

- **A first-principles method, density functional theory**
- **VASP code**
- **PAW-GGA**
- **Ecut: 350 eV**
- **Force on all the atoms : less than 10^{-3}eV/Å**

Part I.

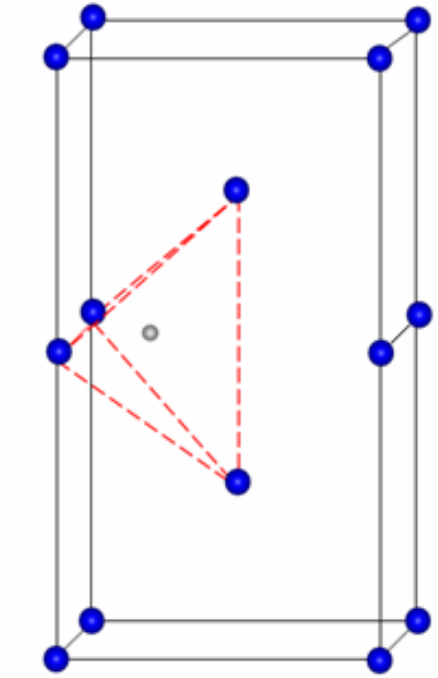
Stress tensor: A quantitative indicator of effective volume and stability of helium in metals

Stability of a single He in W

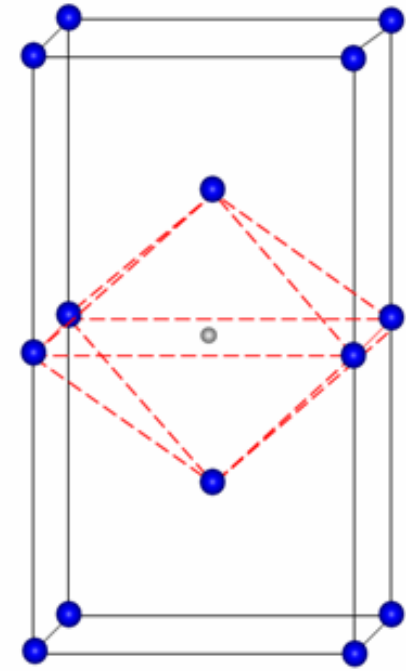


- Helium has a closed-shell electronic structure.
- The stability of He in W should be well characterized by its effective volume.

“Stability” of TIS vs. OIS for He



Tetrahedral interstitial site (TIS)



Octahedral interstitial site (OIS)

- The point-lattice model: *V(OIS)* > *V(TIS)*

Prediction

- Stability of interstitial He: *OIS*

“Stability” of TIS vs. OIS for He

***Solution Energy*:**

$$E_{He}^{s} = E_{NW,He} - NE_{W} - E_{He}$$

$$E_{He}^{s}(TIS) < E_{He}^{s}(OIS)$$

TIS

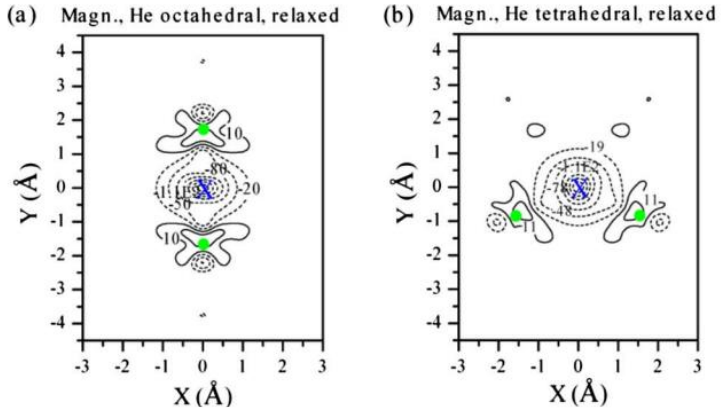
Stability of He in metals

TABLE II. Magnetic moment μ of He defect and its neighboring Fe atoms separated from He by the distance r.

	He	1st Fe		2nd Fe	
	μ (μ_B)	r (Å)	μ (μ_B)	r (Å)	μ (μ_B)
octa, unrelaxed	0.012	1.41	1.67	1.99	2.17
octa, relaxed	0.015	1.66	2.01	2.09	2.24
tetra, unrelaxed	0.007	1.58	1.99	2.54	2.17
tetra, relaxed	0.012	1.76	2.15	2.57	2.21
sub, unrelaxed	0.000	2.45	2.25	2.83	2.08
sub, relaxed	0.000	2.47	2.29	2.85	2.11

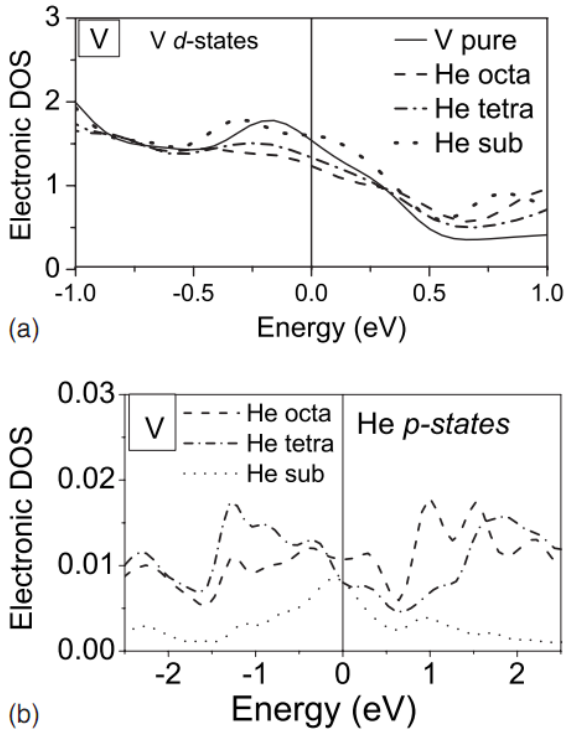
Magnetism, He in Fe

Phys. Rev. Lett. 94, 046403 (2005)



Bonding properties

Phys. Rev. B 80, 054104 (2009)



Hybridization

Phys. Rev. B 78, 134103 (2008)

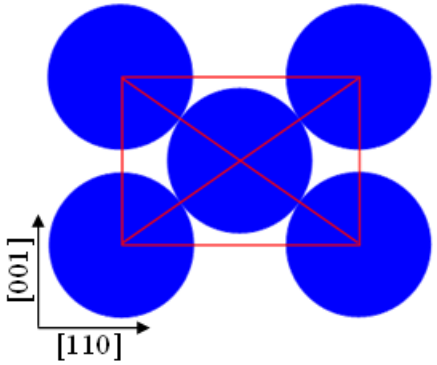
Closed-shell structure of He ?

Lattice model：a qualitative description of the He effective volume

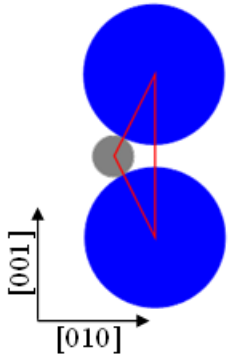
Europhys. Lett. 96, 66001 (2011).

Interstitial He volume based on different models

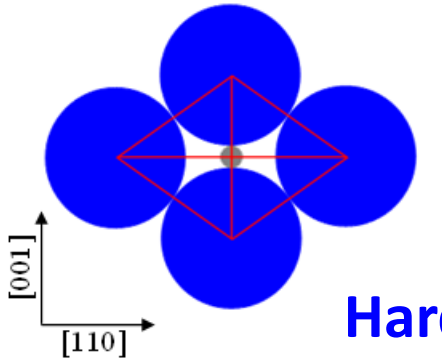
Lattice model	V_{TIS} (a^3)	V_{OIS} (a^3)	V_{TIS}/ V_{OIS}
Point-lattice	8.33%	33.33%	0.25
Voronoi polygon	25.68%	25.11%	1.02
Hard-sphere	0.84%	0.13%	6.46



Bulk



TIS

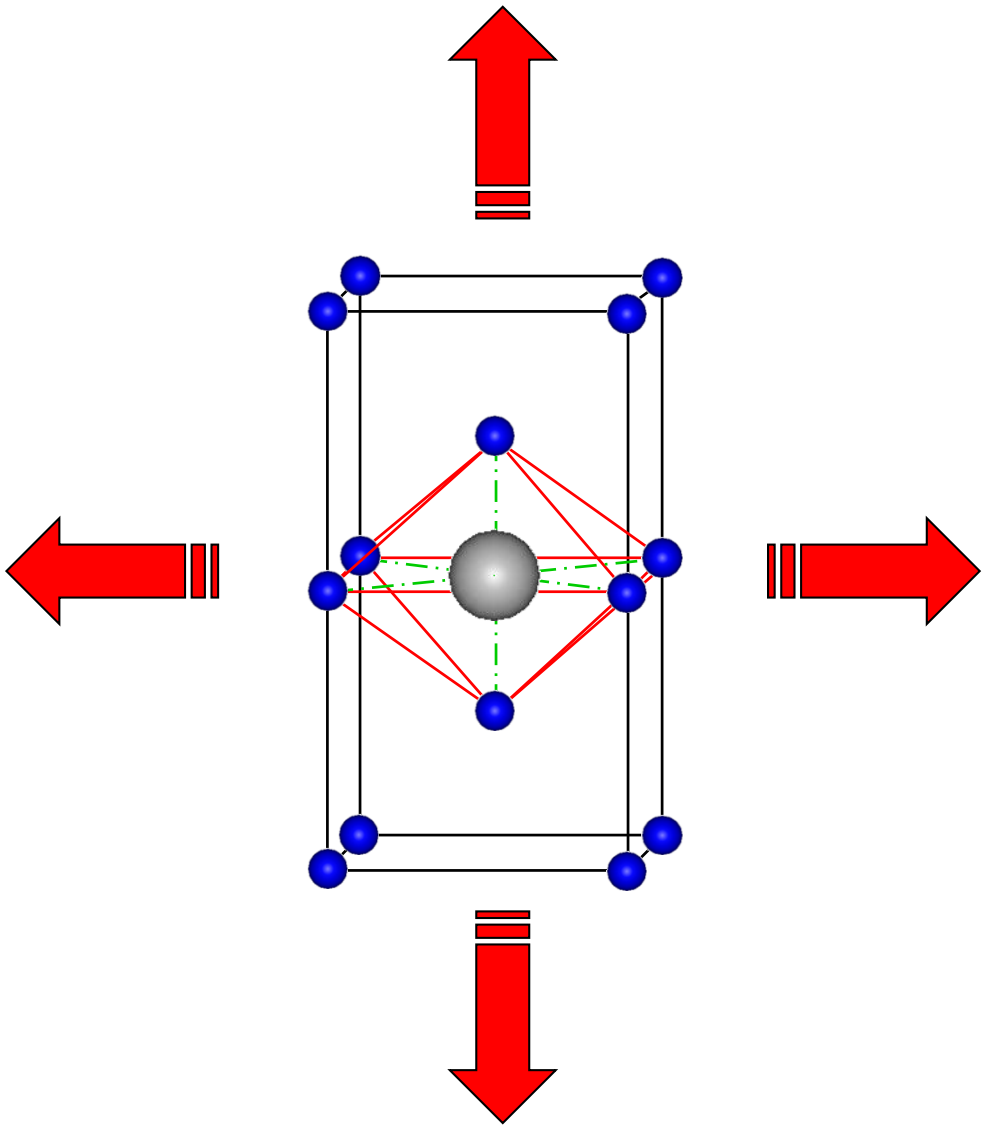


OIS

Hard-sphere

The accuracy of the He effective volume calculation depends on the employed lattice model. Hard-sphere model gives a precise description of the He effective volume compared with other lattice models.

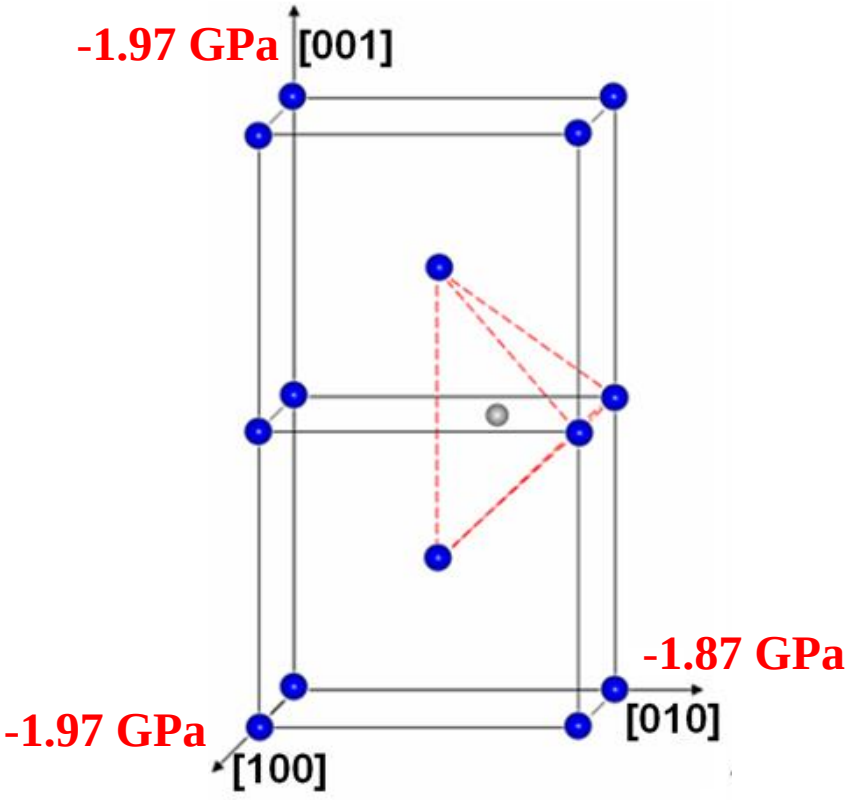
He-induced lattice stress



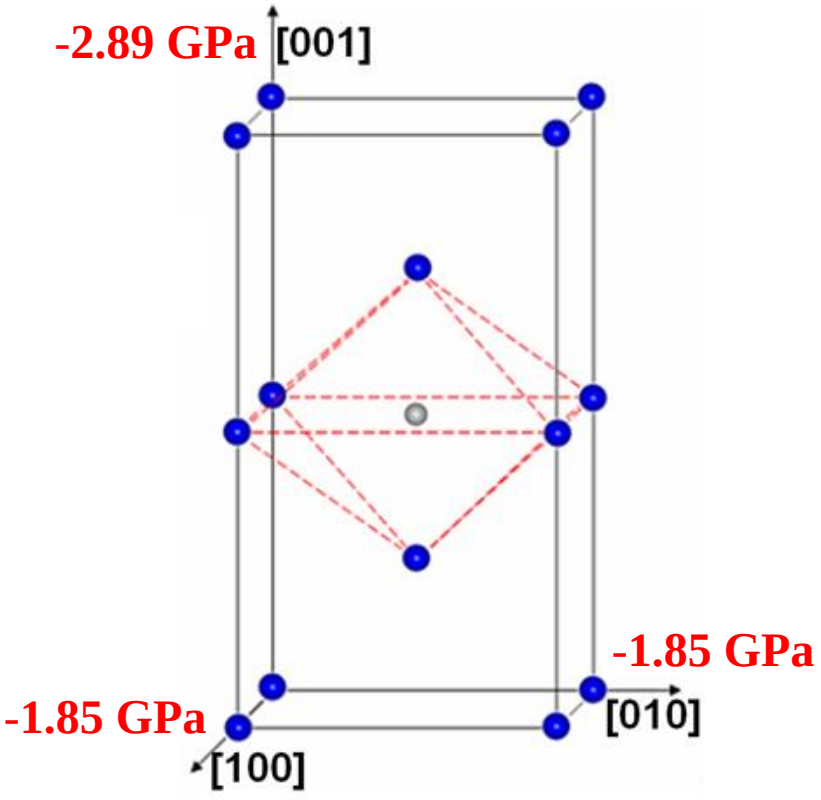
A W lattice is stress free at the equilibrium.

When a He atom is introduced at an interstitial site, it will induce lattice stress.

He-induced lattice stress in W



TIS



OIS

$D_{\text{He-W}}$: 0.559 *a*

0.5 *a*; 0.707 *a*

Stress tensor: A quantitative indicator of effective volume and stability of He in metals

He-induced stress (in GPa) in bcc metals

		W	Mo	Fe	Cr	V
σ_{TIS}	$XX=ZZ$	-1.97	-1.79	-2.54	-2.29	-1.31
	YY	-1.87	-1.76	-2.61	-2.59	-1.79
σ_{OIS}	$XX=YY$	-1.58	-1.48	-2.38	-1.98	-1.59
	ZZ	-2.89	-2.55	-3.24	-3.44	-1.81

Stress tensor could be used to indicate the He effective volume in metals, thus has an influence on the solution of He.

Part II.

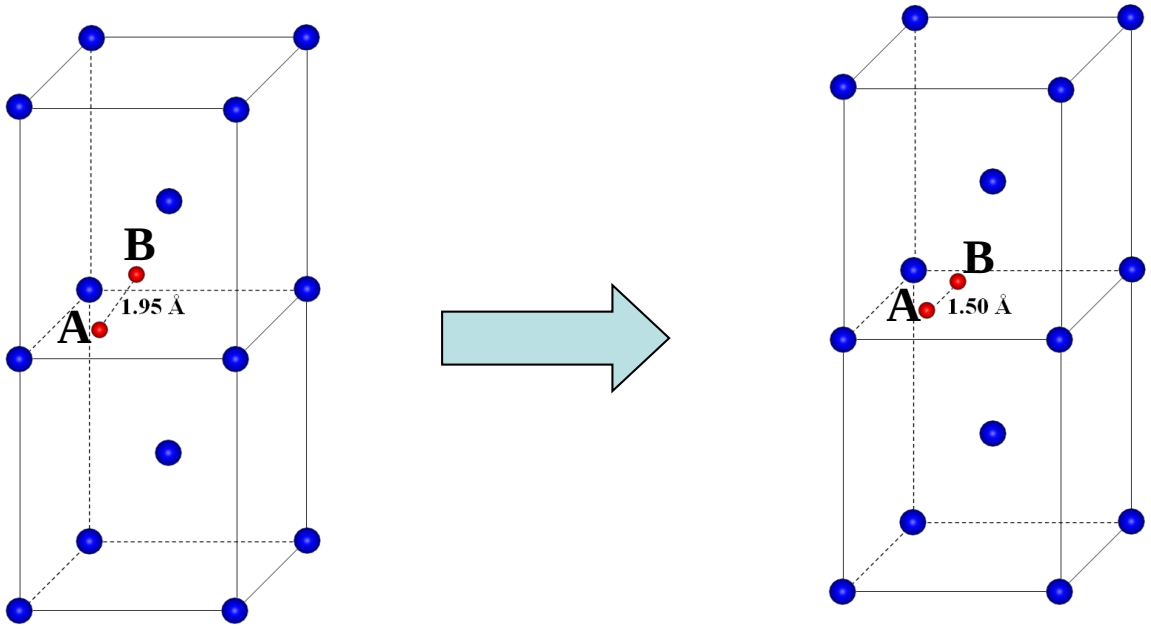
Electrophobic interaction: the leading mechanism for helium self-trapping in tungsten

Solution energy of He in W

	TIS	OIS	Diff
Solution energy	6.17	6.38	0.21
Mechanical contribution	0.98	1.14	0.16
Electronic contribution	5.19	5.24	0.05

The *mechanical contribution* plays a key role in the relative stability of He in W, while the *electronic contribution* should be responsible for the poor solubility of He in W.

He-He interaction in W



$$E^b = 2E^T_{W,1He} - E^T_{W,2He} - E^T_W = 1.02\text{eV}$$

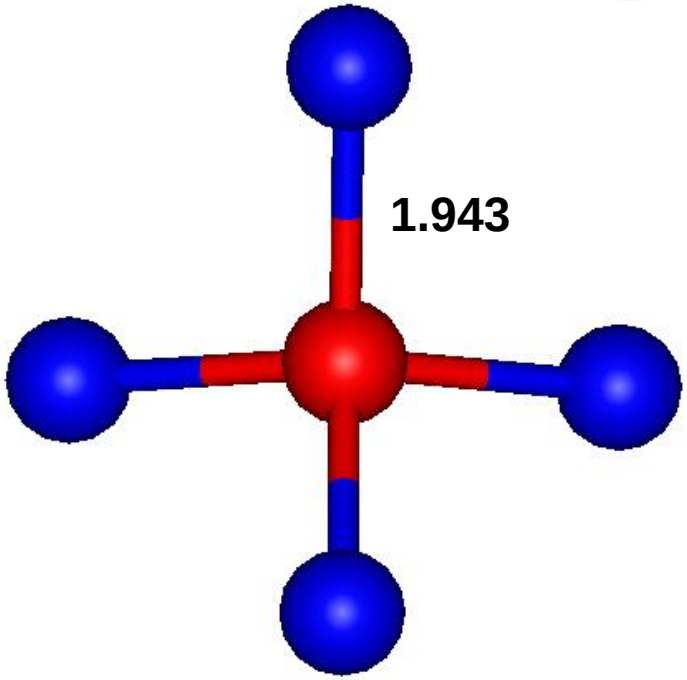
$$= 2E^{sol}_{1He} - E^{sol}_{2He}$$

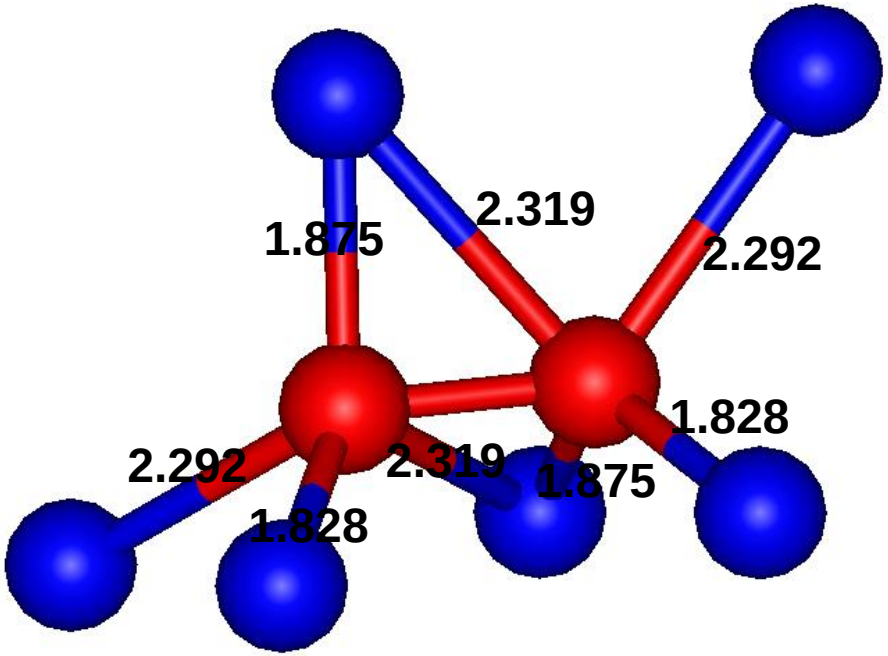
12.34 eV | **11.32 eV**

Mechanical contribution

Electronic contribution

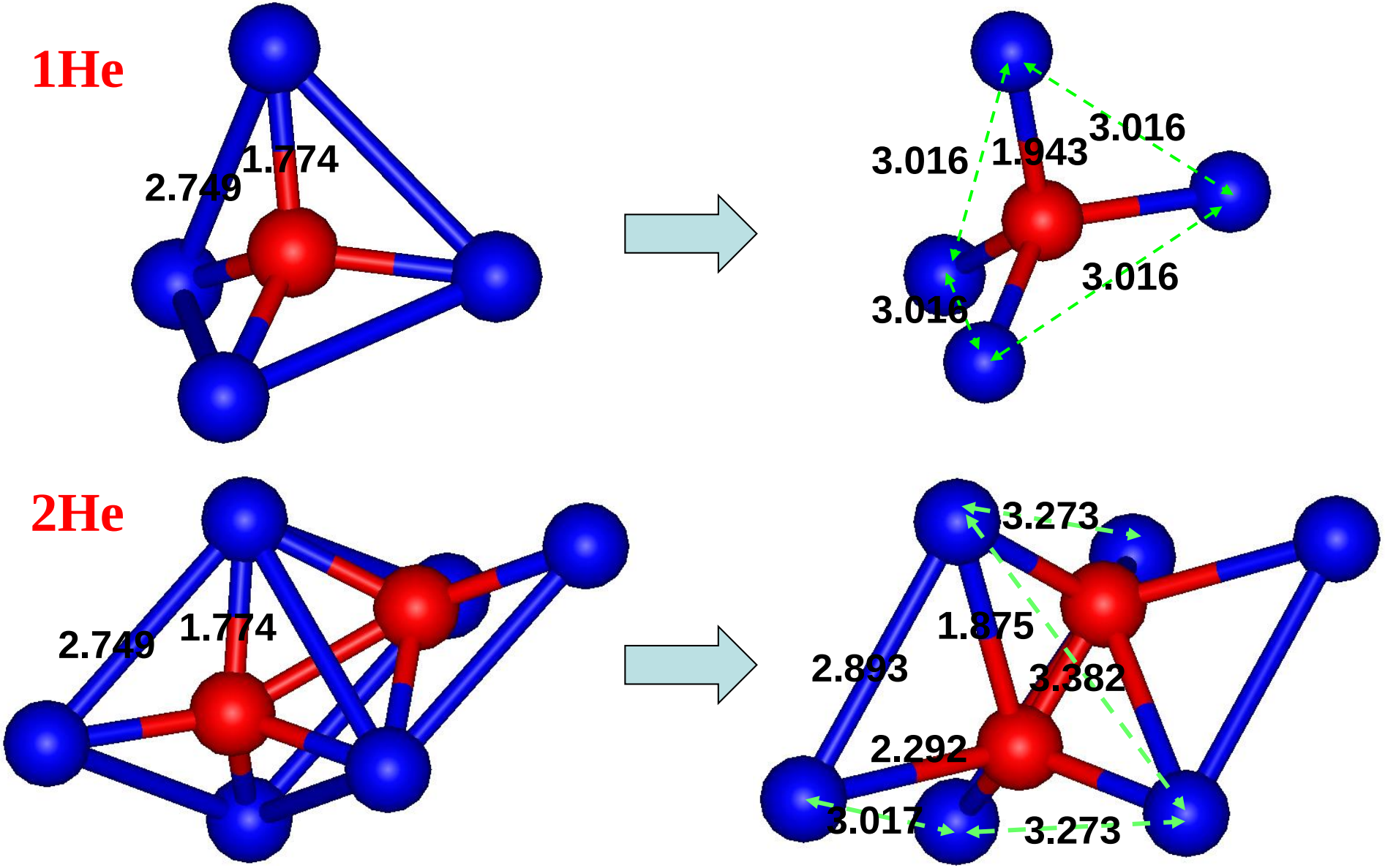
He-He binding energy





	2×1He	2He	Diff
Solution energy	12.34 eV	11.32 eV	-1.02 eV
Mechanical contribution	1.96 eV	3.16 eV	1.20 eV
Electronic contribution	10.38 eV	8.16 eV	-2.22 eV
Electron density	*0.17 e/Å^3*	*0.12 e/Å^3*	

Distortion of W lattice induced by He

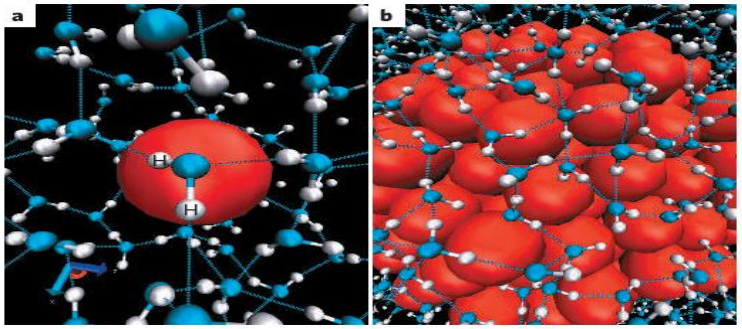


Electrophobic dissolution of He in metals

- **Solutes in water:**

hydrophilic		hydrophobic
Ionic/hydrogen bonds with water	or	Cluster
Sugar and salt		Oil and fat

Oil in water



Nature 437, 640 (2005)

- **Metals → Free electrons**
- **Atom in metals: electropilic or *electrophobic***

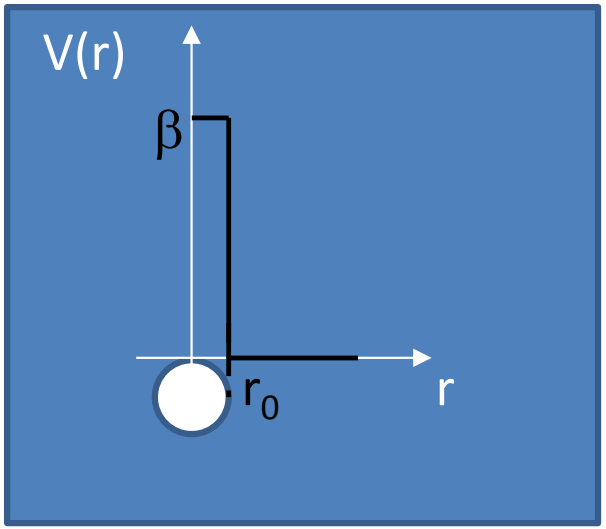
He clustering in metals

The electrophobic interaction provides the leading mechanism for He self-trapping in metals.

Electrophobic interaction between He atoms

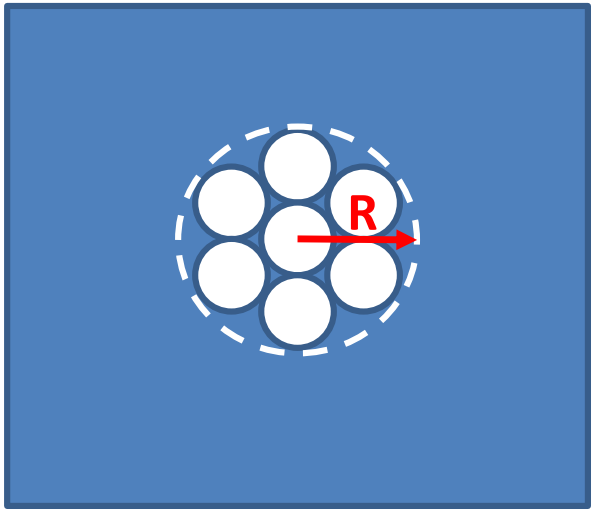
Classical-hard sphere potential :

$$V(r) = \begin{cases} b, & r = r_0 \\ 0, & r > r_0 \end{cases}$$



Total solution energy:

$$N(4\pi r_0^2)\beta$$



Solution energy of the cluster:

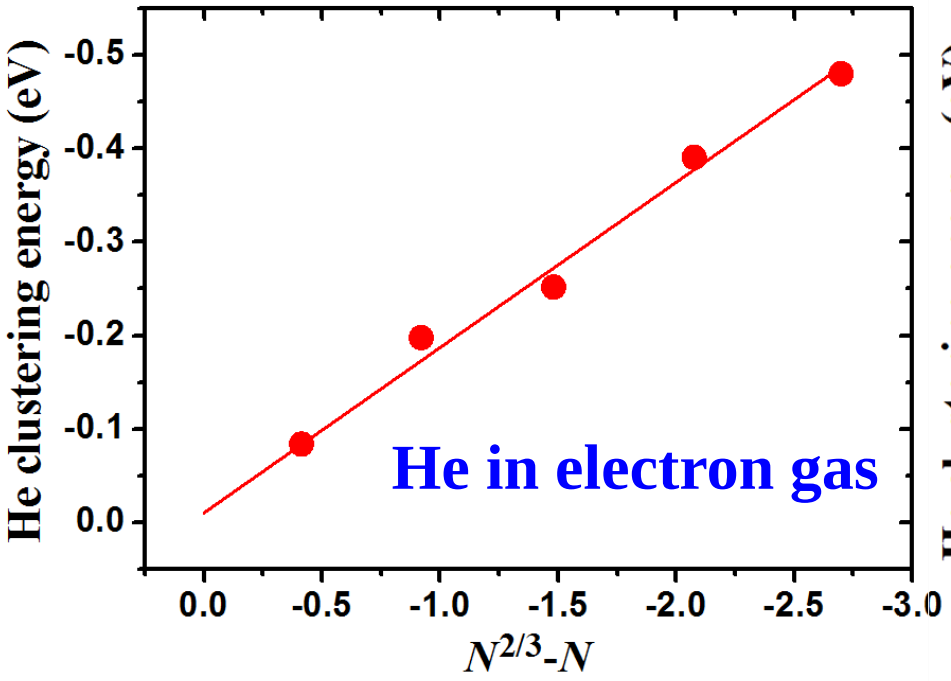
$$(4\pi R^2)\beta \qquad (R^3 \approx N r_0^3)$$

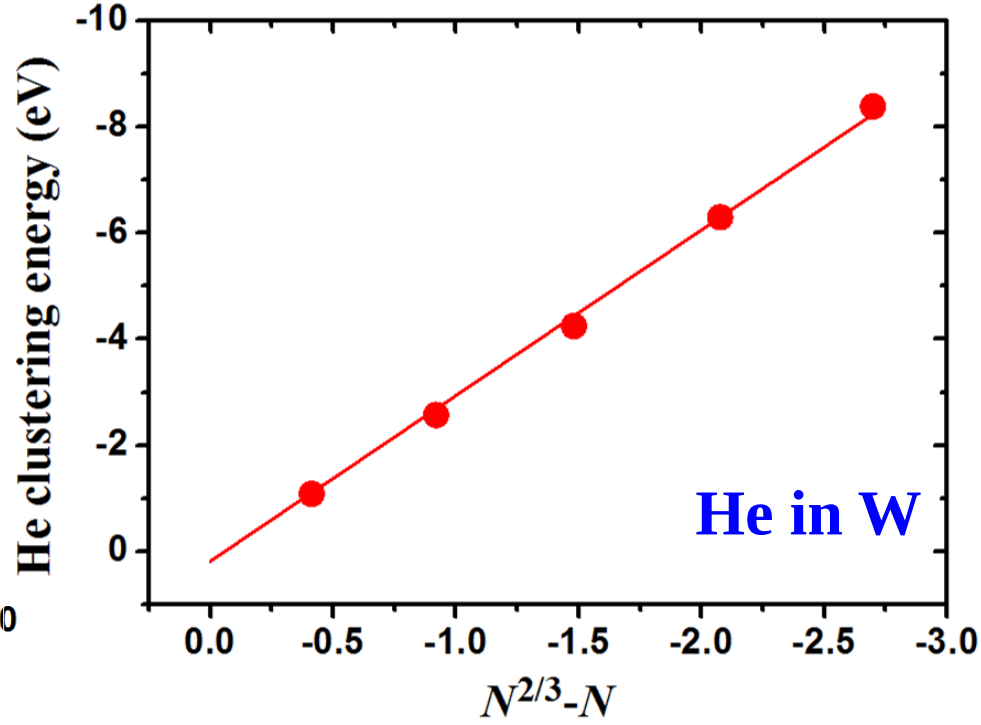
Binding energy of the cluster:

$$E_c \propto (N^{2/3} - N)$$

He clustering energy in electron gas and W

$$E^c \propto (N^{2/3} - N)$$

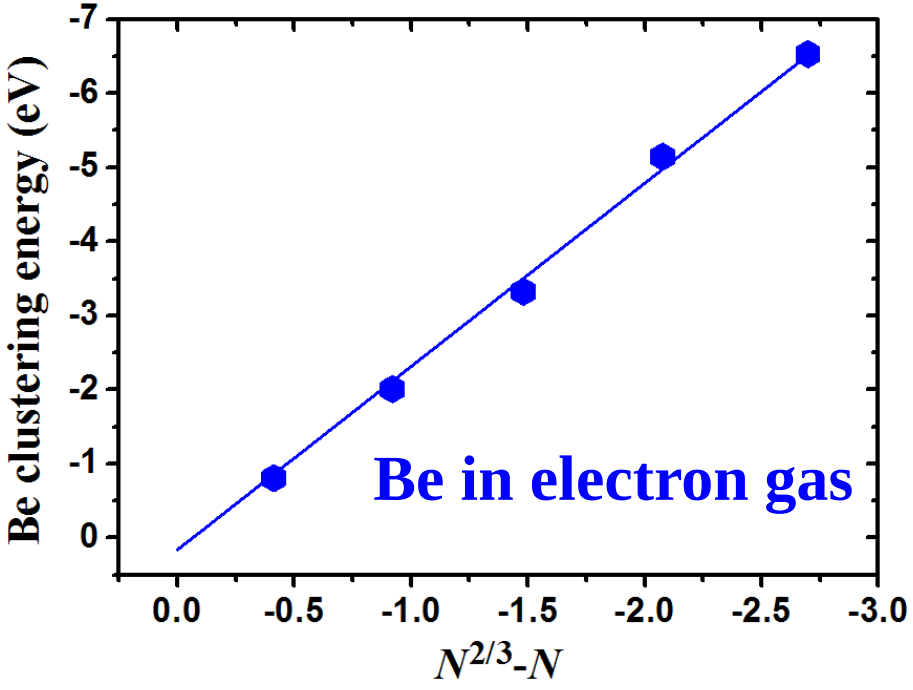


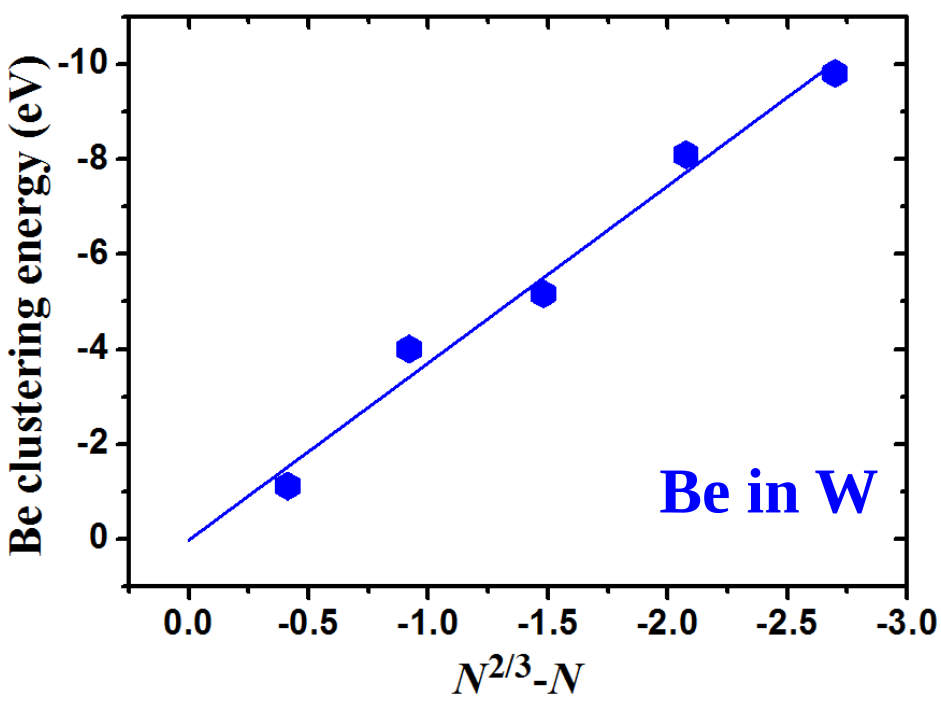


The separated He prefers to get together with high clustering energy in metals due to its electrophobic property.

Be clustering energy in electron gas and W

$$E^c \propto (N^{2/3} - N)$$





The separated Be prefers to get together with high clustering energy in metals due to its electrophobic property.

Part III.

Anisotropic Strain Enhanced Hydrogen Solubility in Tungsten: The Independence on the Sign of Strain

H blistering in W

Application

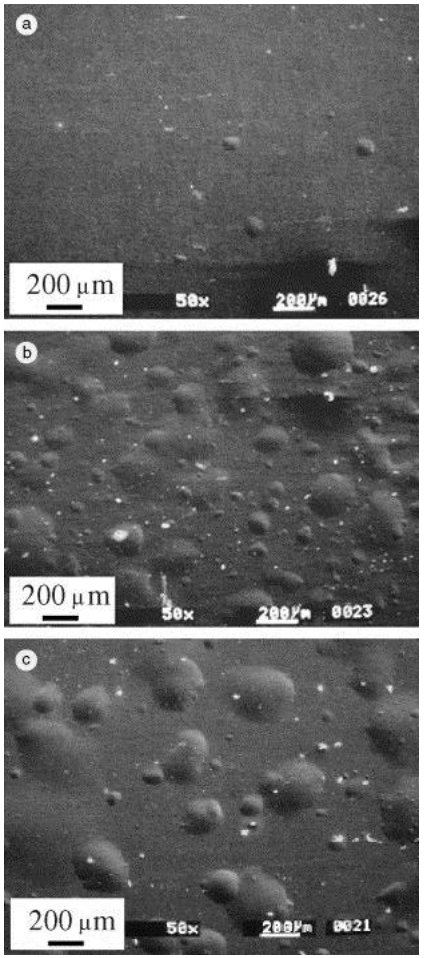
- Tungsten (W) and W-alloys are considered to be one of the most promising candidates for the plasma facing materials in Tokamak.

Advantages

- Low sputtering erosion and good thermal properties including high thermal conductivity and high melting temperature.

Critical issues: *H blistering*

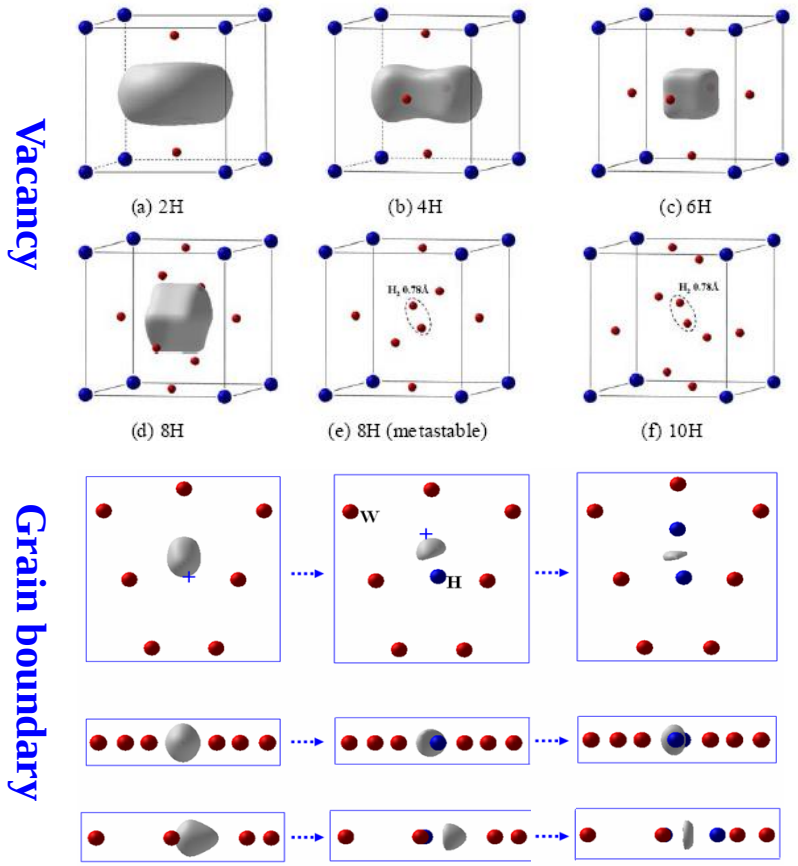
- Higher H concentrations isotope ions on a W single crystal, as well as polycrystalline will give rise to **<u>blistering</u>** at the W surface.



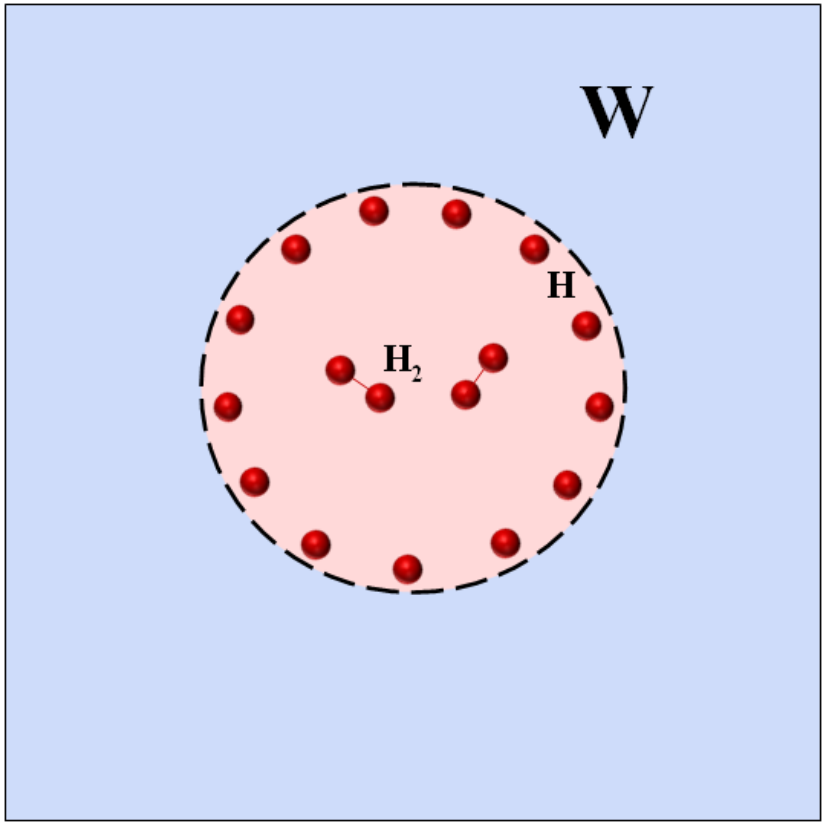
M.Y. Ye, et al., *J. Nucl. Mater.* (2003).

Mechanism for the H bubble formation in W

- Y. -L. Liu and G.-H. Lu *et al*., PRB (2009).
- H. -B. Zhou and G.-H. Lu *et al*., Nucl. Fusion (2010).



Iso-surface of optimal charge for H with different number of trapped H atoms



Vacancy trapping mechanism

● **We have revealed the vacancy trapping mechanism for H bubble formation in W based on the investigations of H dissolution and diffusion in vacancy and grain boundary.**

H bubbles induced strain to the surrounding W lattice

H bubbles

↓

Induce strain to the surrounding W lattice

↓

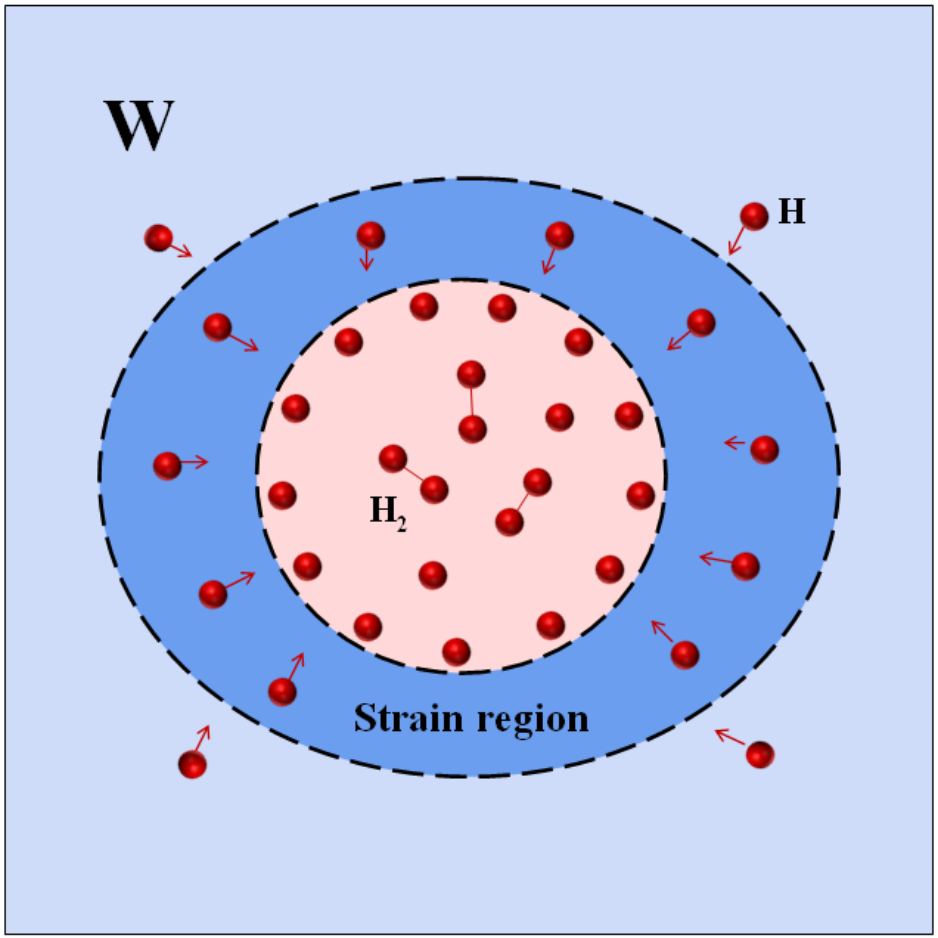
High inner gas pressure

A few GPa to a few tens of GPa

JNM(1998);JNM(1999);NF(2007)

↓

Effect of strain on H solubility

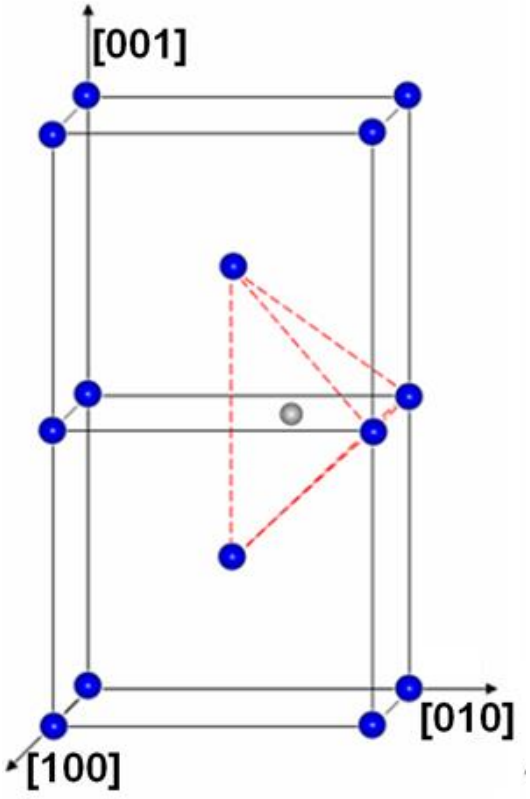


Prediction:

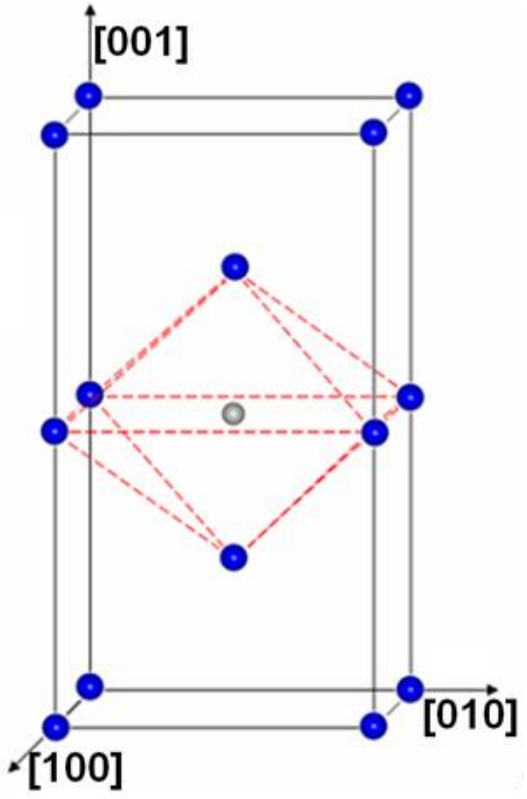
- Strain enhances H solubility → facilitate H bubbles growth
- Strain reduces H solubility → suppress H bubbles growth

?

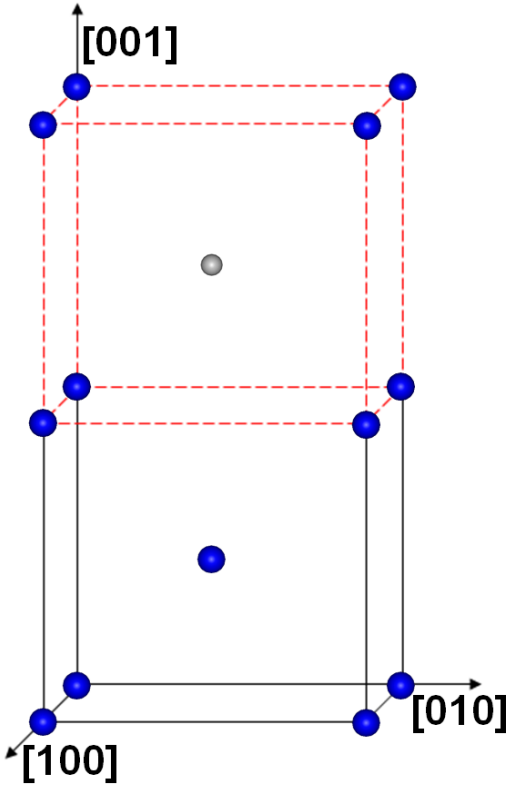
The potential occupation sites



Tetrahedral interstitial site (TIS)



Octahedral interstitial site (OIS)

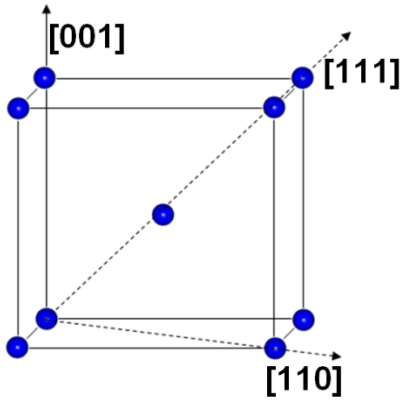


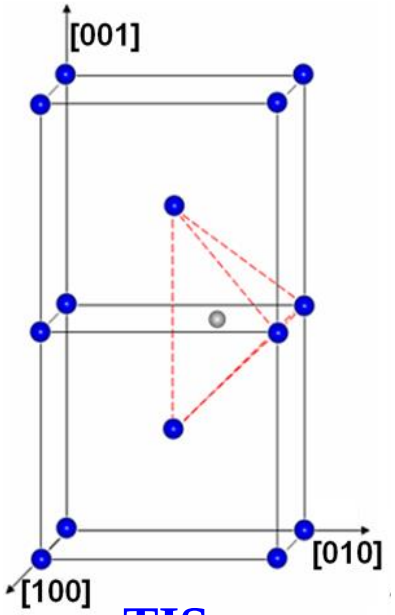
Substitutional site (SS)

E^{sol} : **0.90 eV** **1.28 eV** **2.65 eV**

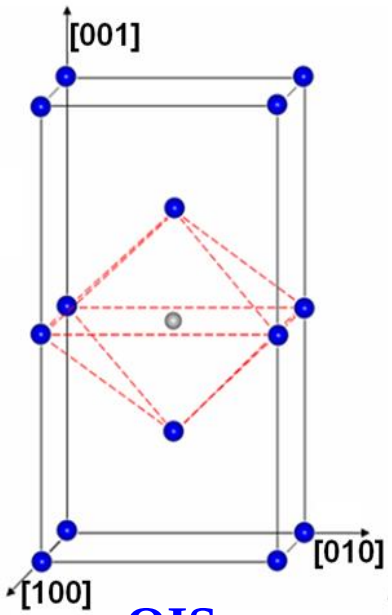
The effect of the triaxial strain on H in W

- Triaxial strain
- Biaxial strain





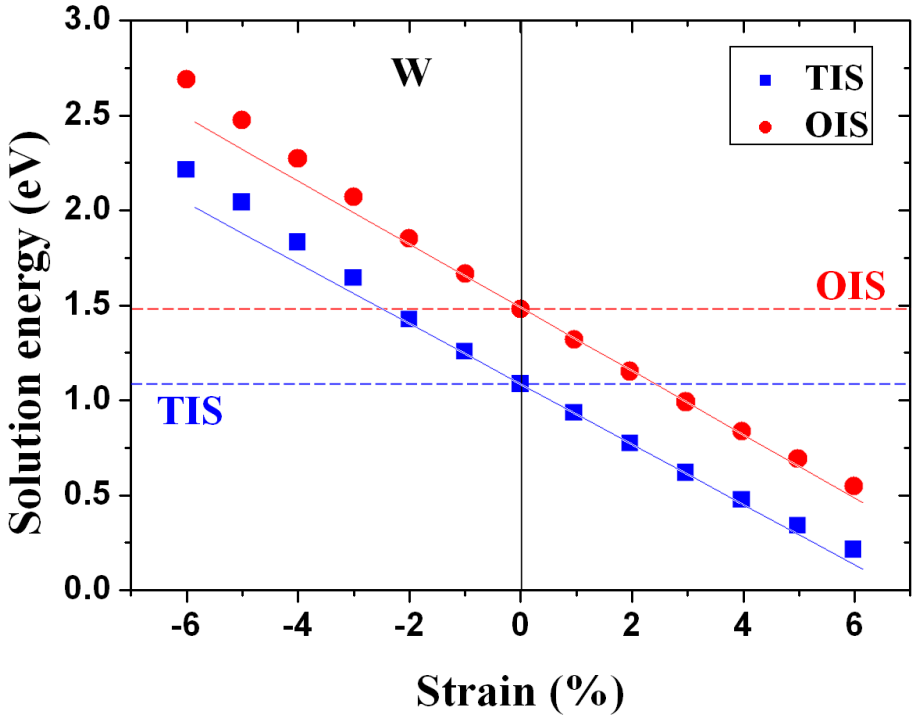
TIS



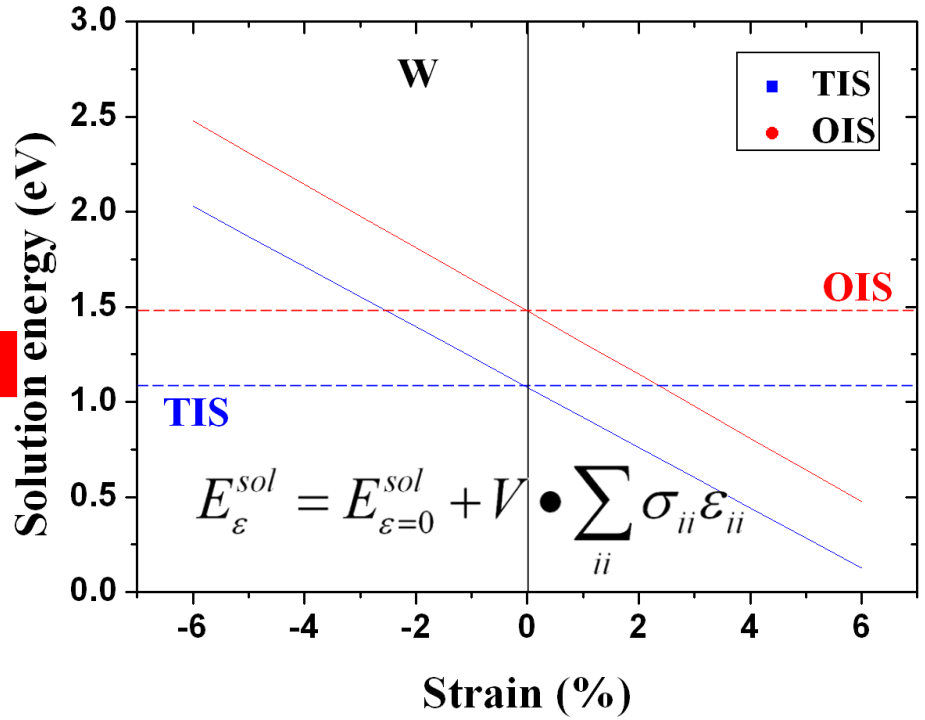
OIS

The potential interstitial sites of H in W under the *triaxial strain*.

Dissolution of H in W under the triaxial strain



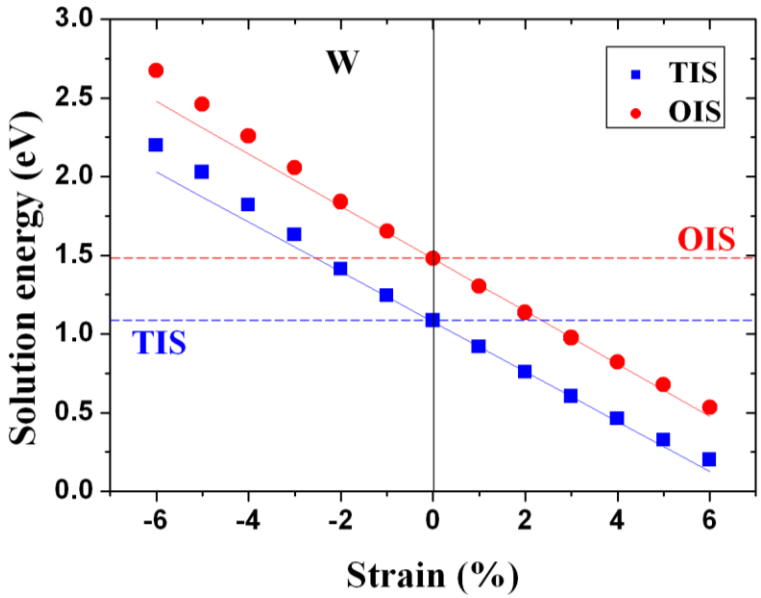
First-principles calculations

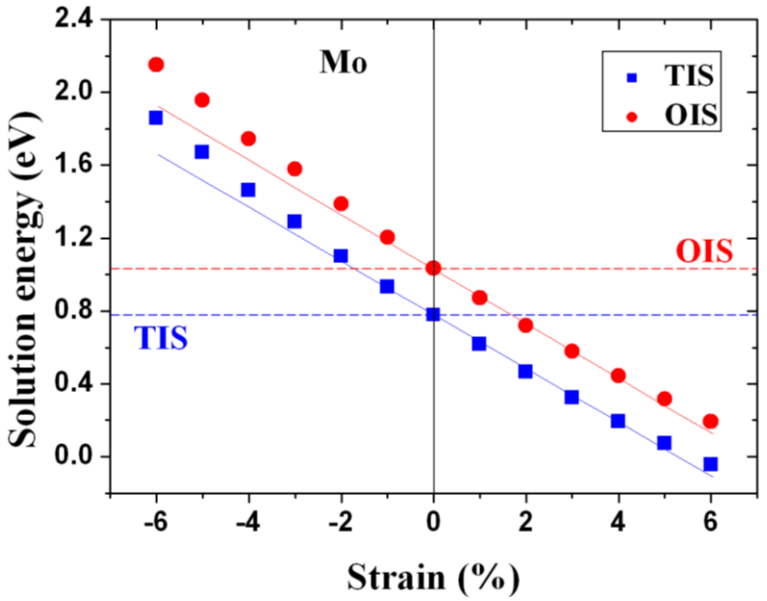


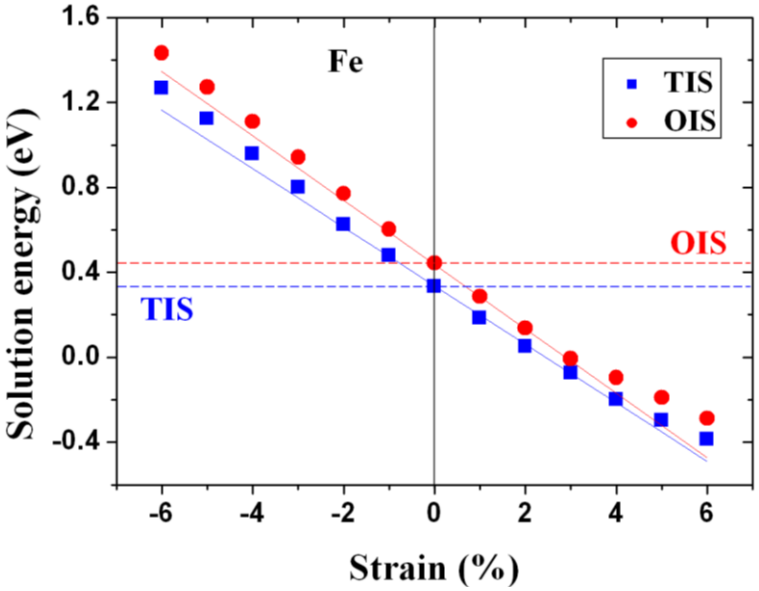
Linear elasticity theory

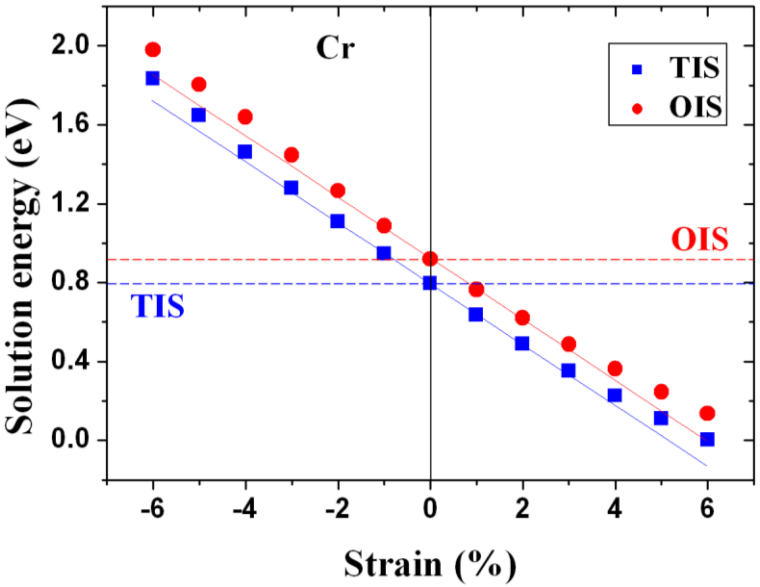
- The solution energies of H at both TIS and OIS will decrease with the increasing tensile strain, but increase with the increasing compressive strain.
- The H solution energy is a linear monotonic function of the triaxial strain.

H in Mo/Fe/Cr under the triaxial strain

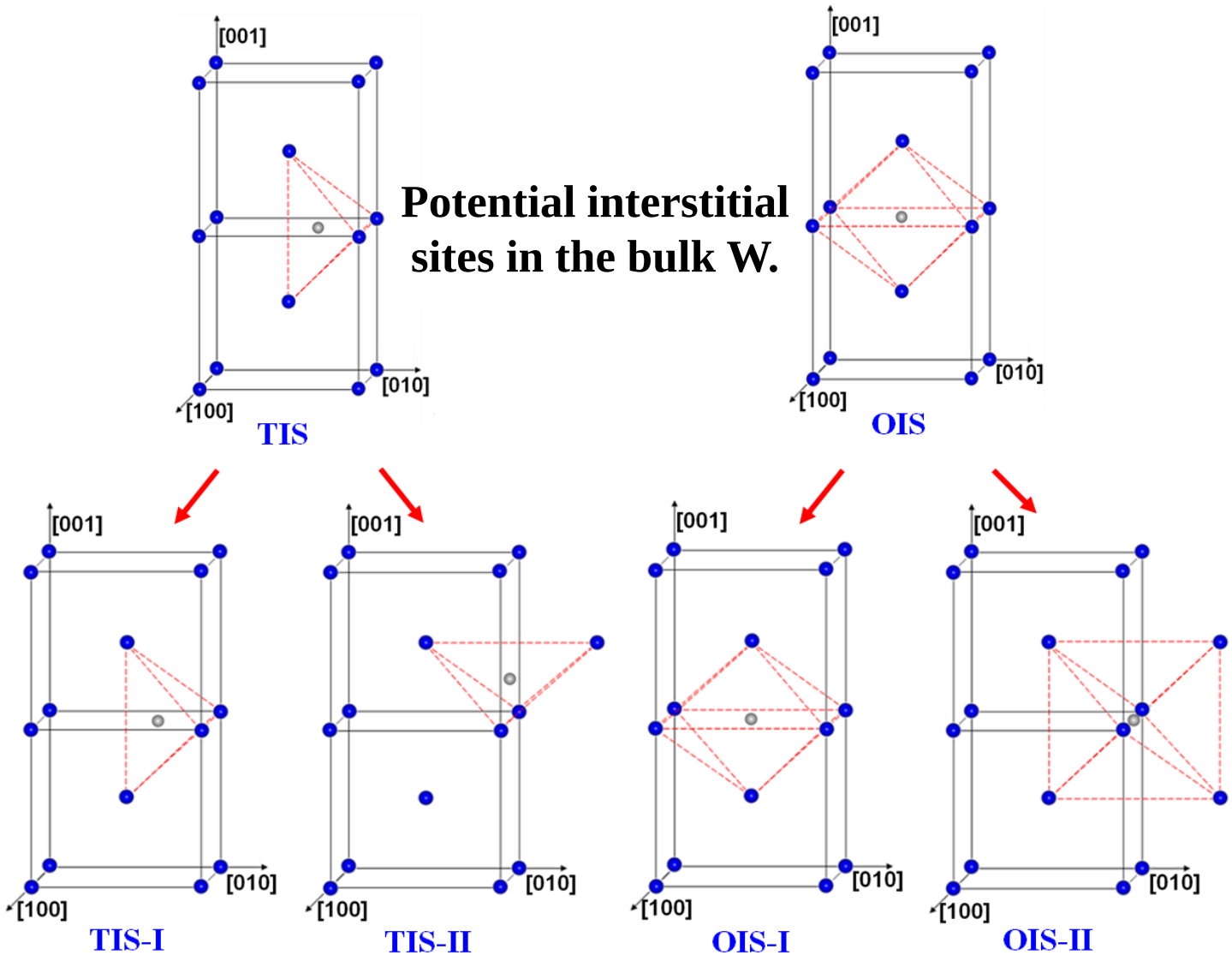






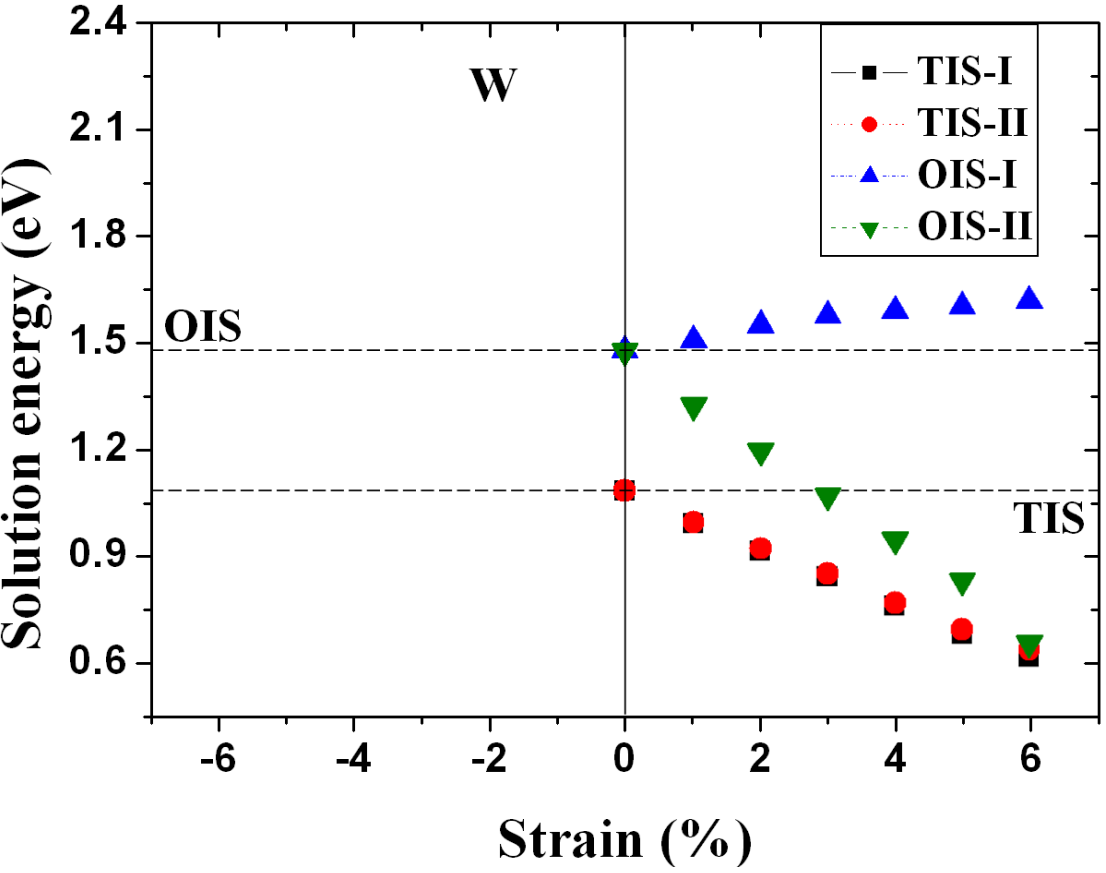


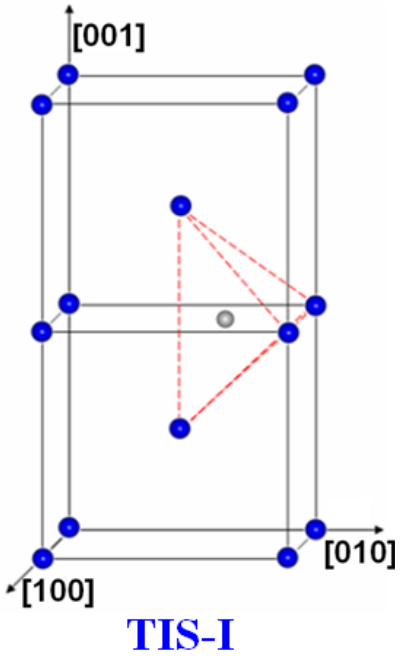
The effect of the biaxial strain on H in W

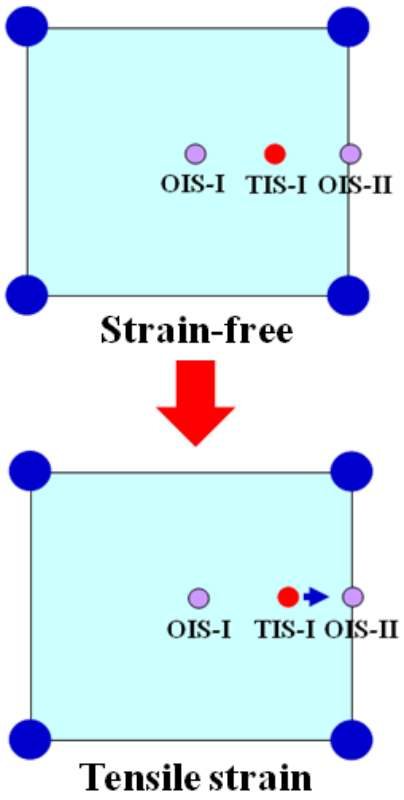


The potential interstitial sites of H in W under the *biaxial strain*.

Dissolution of H in W under the biaxial tensile strain

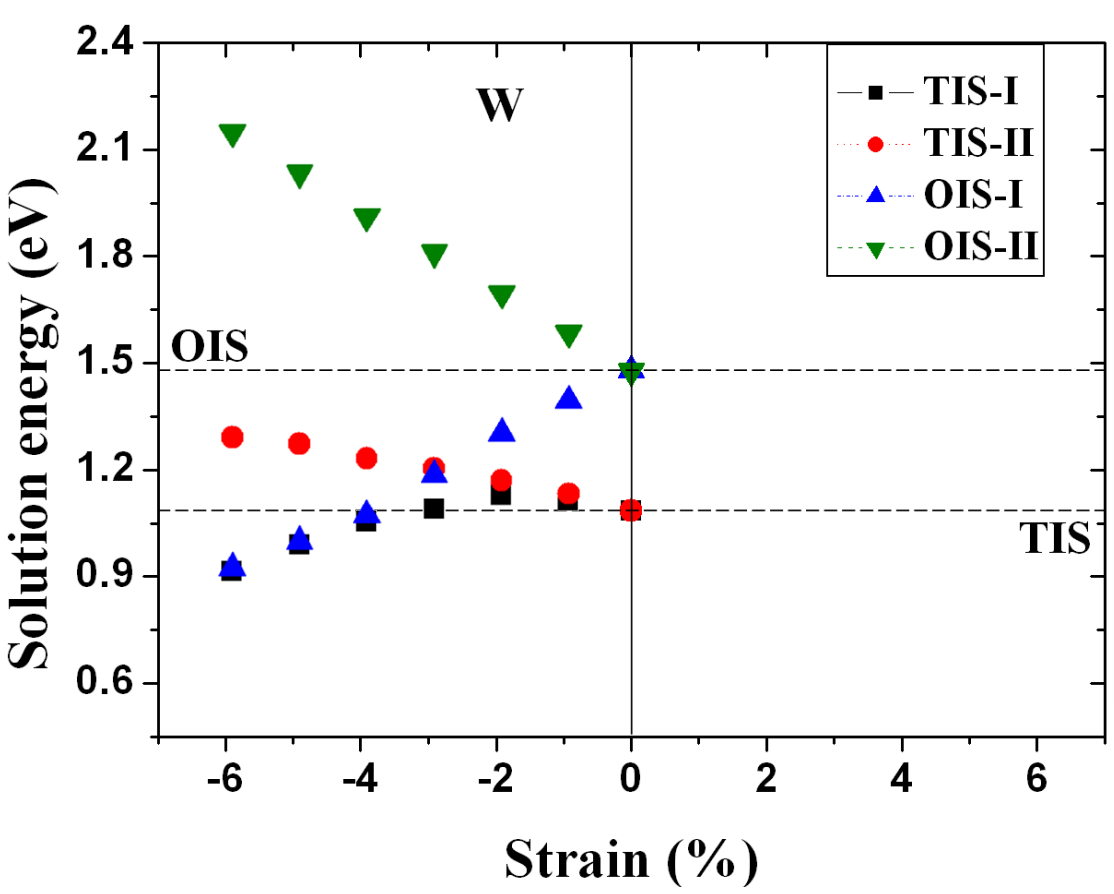


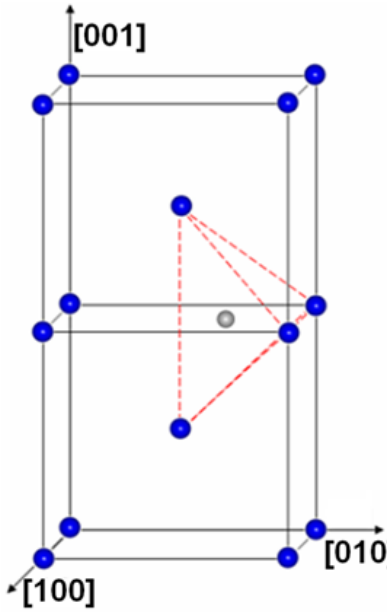


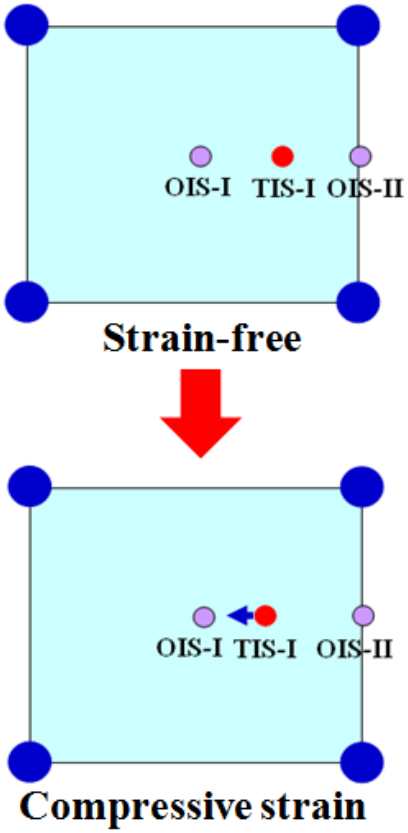


- The solution energies of H at two TIS's and the OIS-II will decrease with the increasing strain, but increase at the OIS-I.
- The energy curves of H at the TIS-I and the OIS-II will merge together.
- Movement of H forced by the biaxial tensile strain.

Dissolution of H in W under the biaxial compressive strain

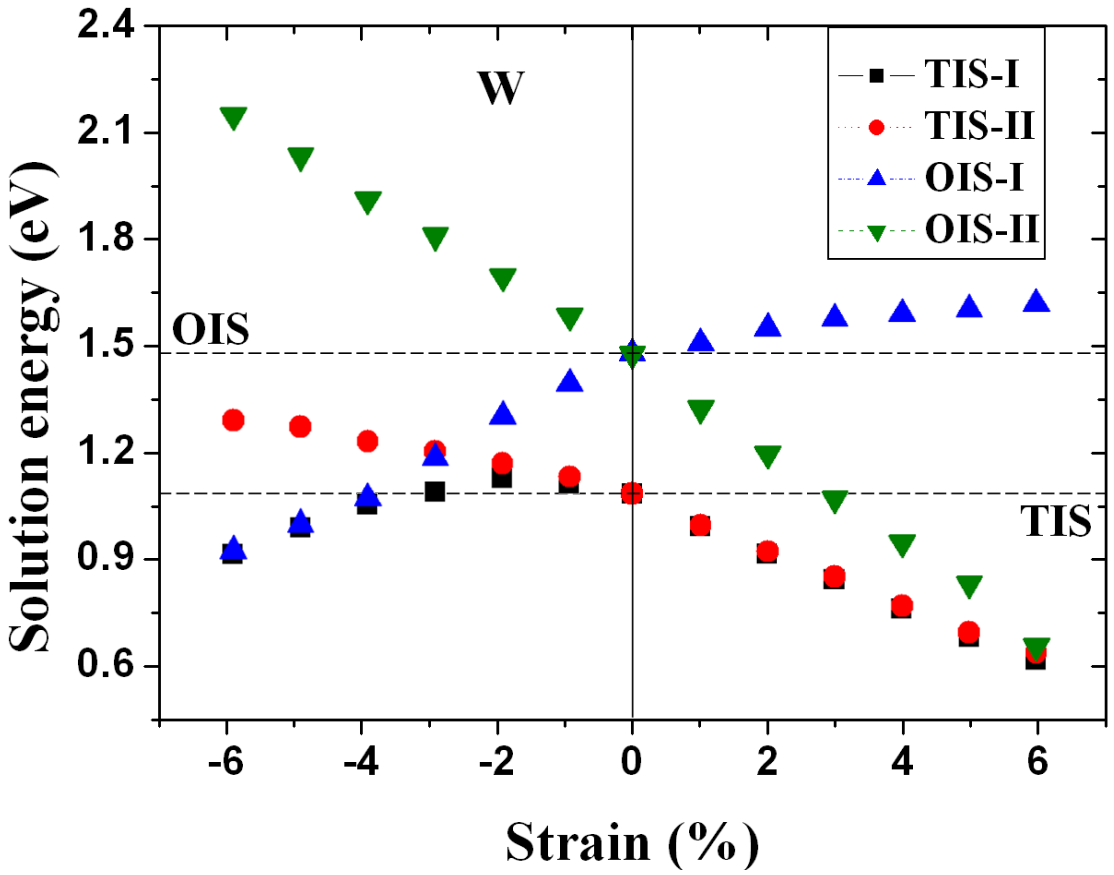


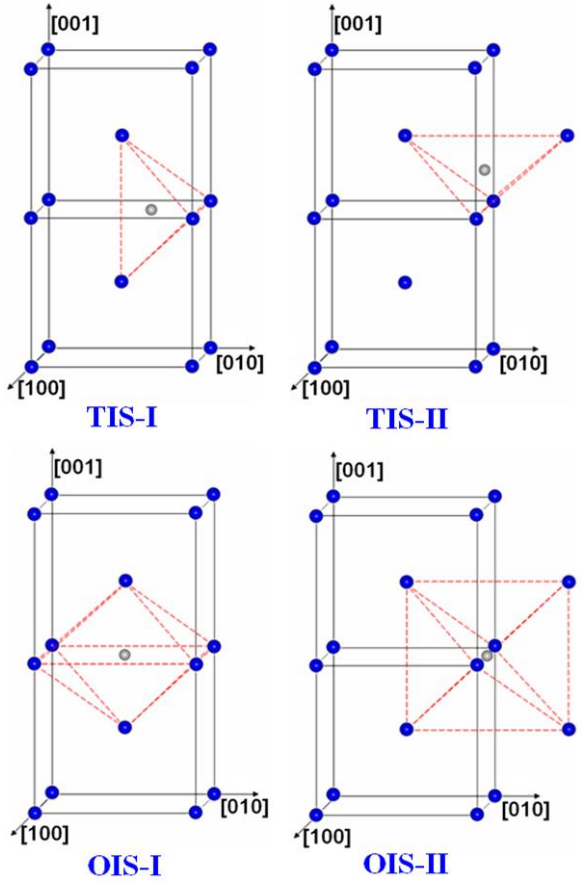




- The solution energies of H at the TIS-I and the OIS-I will decrease with the increasing strain, but increase at the TIS-II and the OIS-II.
- The energy curves of H at the TIS-I and the OIS-I will merge together.
- Movement of H forced by the biaxial compressive strain.

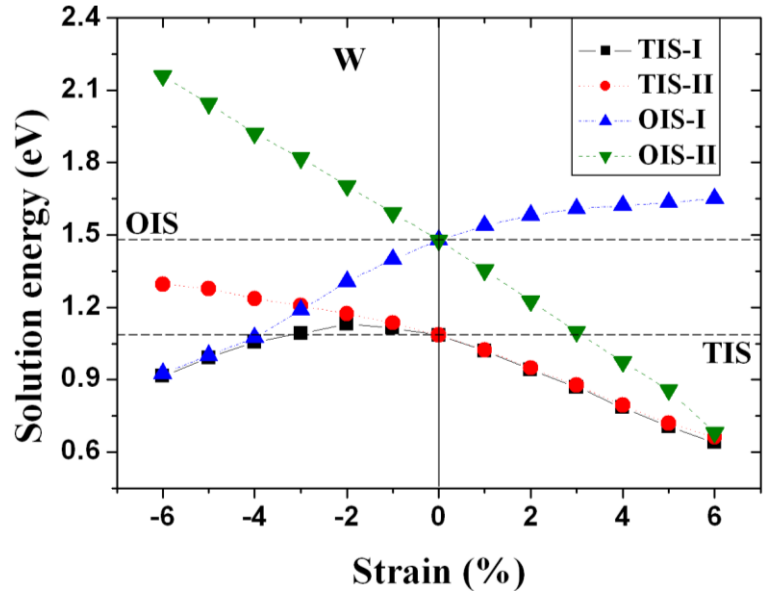
Dissolution of H in W under the biaxial strain

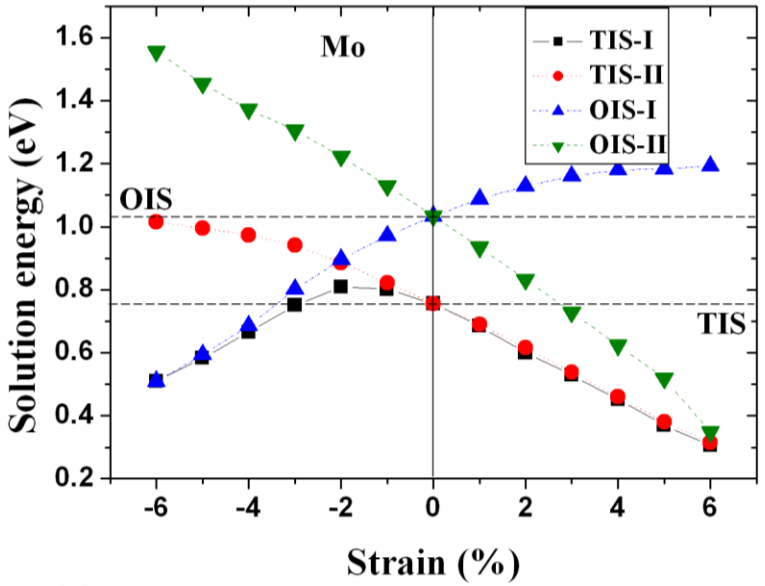


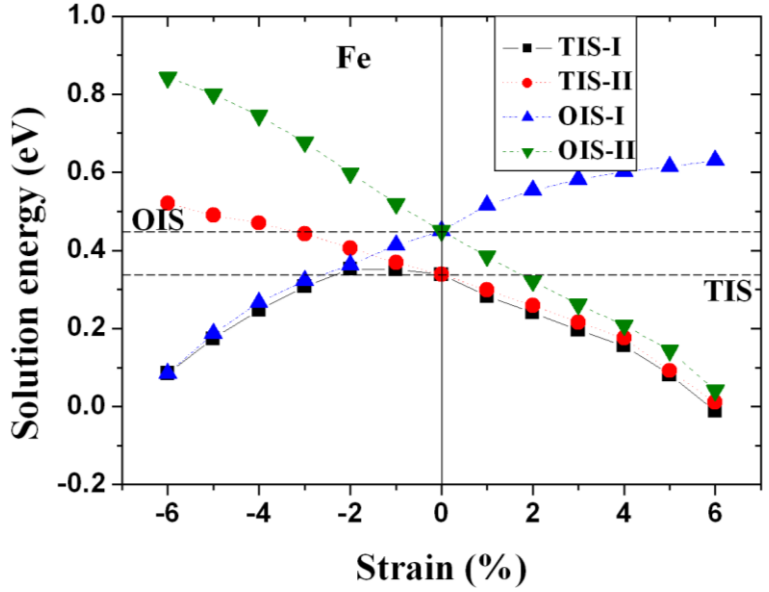


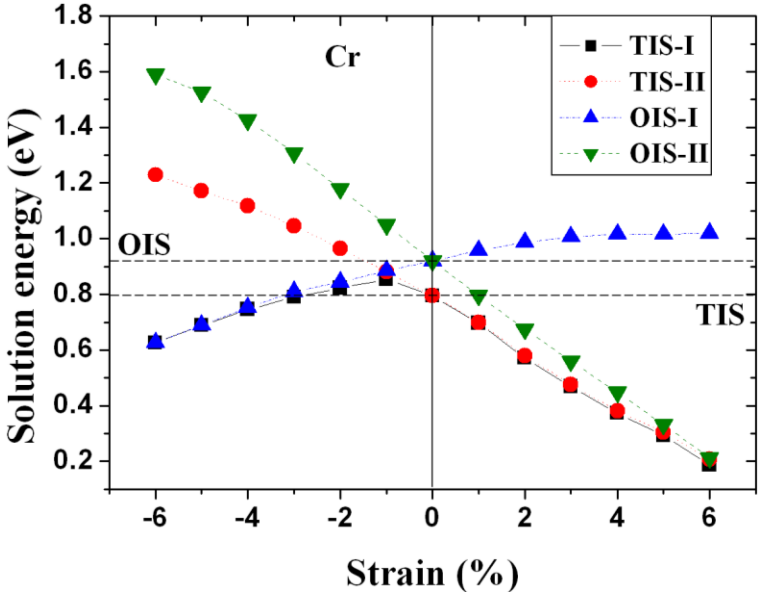
- The solution energies of H at the TIS-II and the two OIS's are normal showing a monotonic dependence on the strain. However, the solution energy of H at the TIS-I shows a non-monotonic dependence on the strain.
- The solution energy of H at the TIS-I "effectively" decreases with the increasing of both signs of strain.

H in Mo/Fe/Cr under the biaxial strain

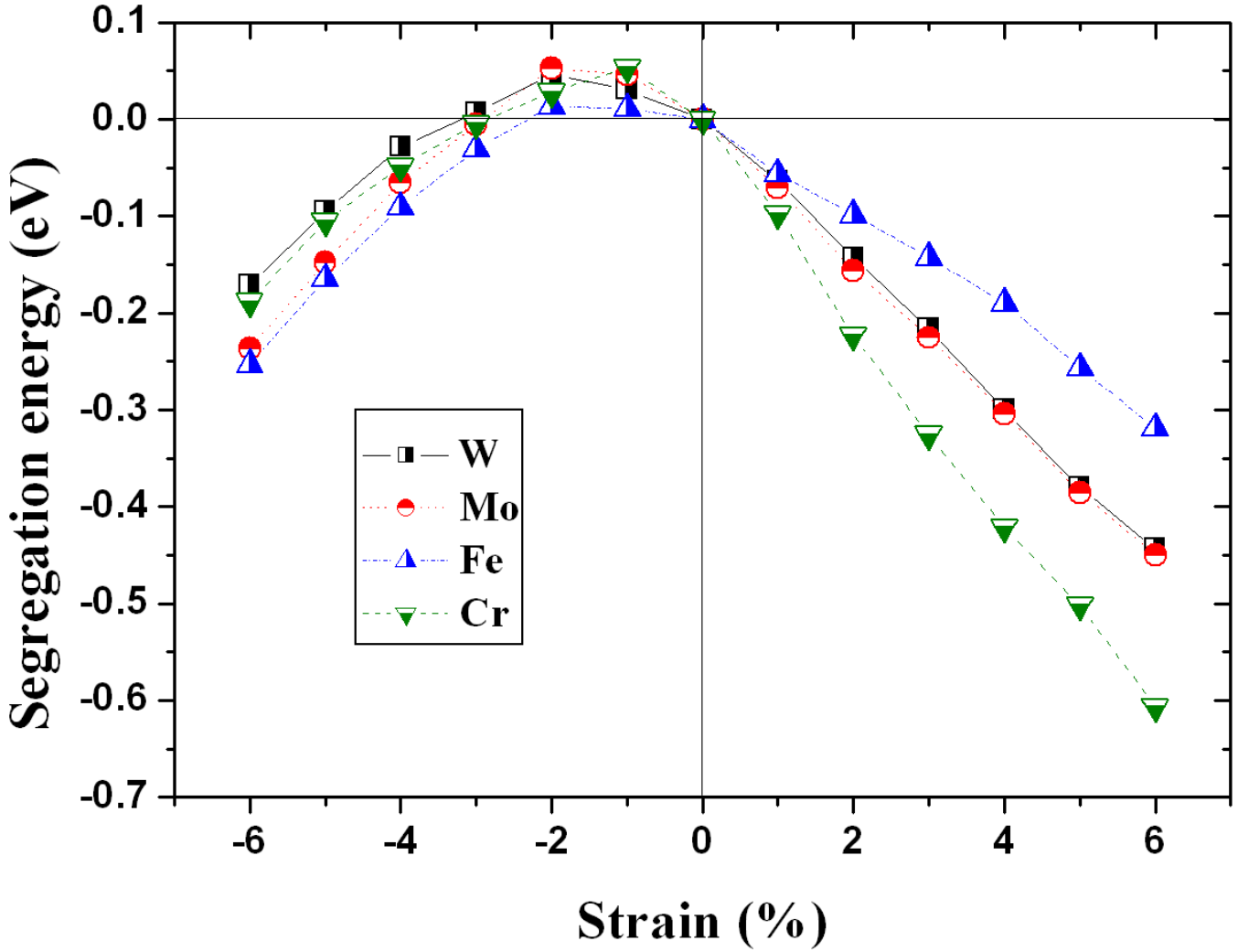






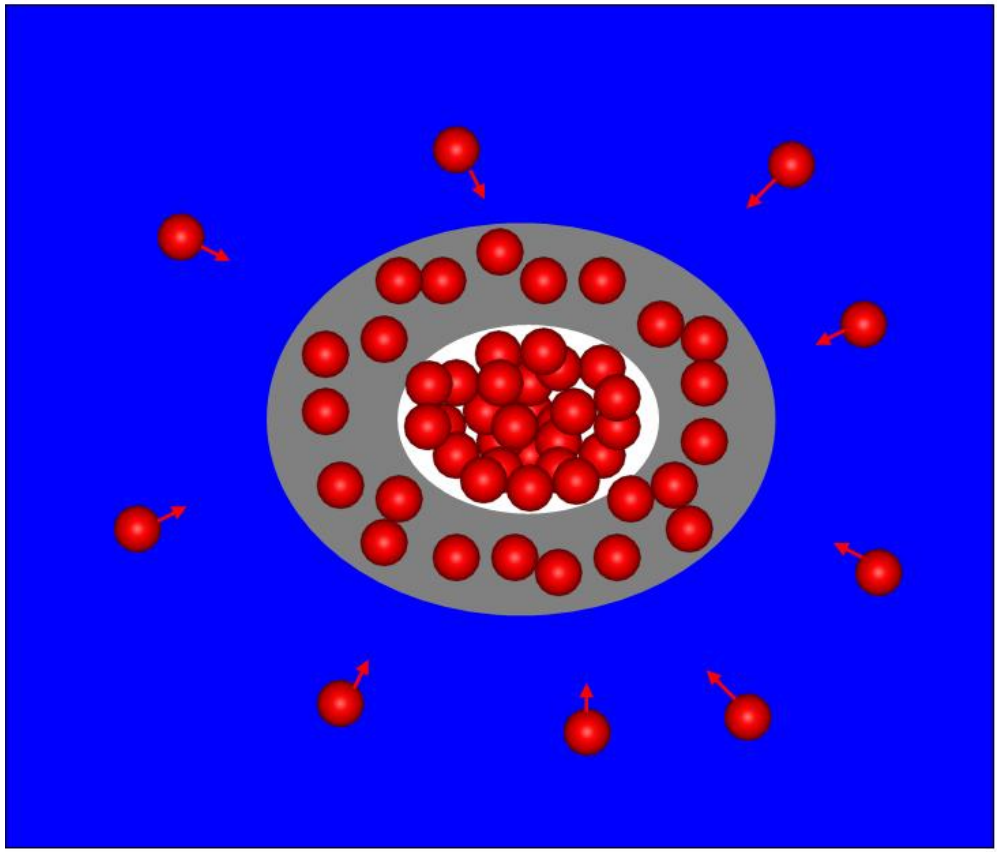


Segregation energy of H in the biaxial strained metals



- The segregation energy indicates that H energetically prefers to segregate in the strained metals instead of the strain-free metals.

Strain-triggered cascading effect on H bubble



H accumulation→ H bubble formation→ Anisotropic strain in W

↓

……←H bubble growth←Enhancing H solubility

Summary

Electrophobic interaction: the leading mechanism for He self-trapping

- The dissolution of He in metals can be popularly considered as electrophobic.
- The electronic effect is largely responsible for the poor solubility of He in the bulk W.
- The electrophobic interaction provides the leading mechanism responsible for the experimentally observed phenomenon of He self-trapping in metals.

Anisotropic strain enhanced H solubility in W

- The H solubility can be enhanced by anisotropic strain in some bcc metals, almost independent of the sign of strain.
- Our finding suggests a cascading effect of H bubble growth in bcc metals.

Thanks for your attention!