28th/May/2025 @ IAEA
7th International Workshop on Models and Data for Plasma–Material Interactions in Fusion Devices: MoD-PMI 2025 (26-28 May, 2025)

Applying machine learning potential models to the study of hydrogen in metals: accurate property calculations, complex dynamics simulations, and challenges

Takuji Oda*, Sojeong Yang, Hyukjoon Kwon

(oda@snu.ac.kr)

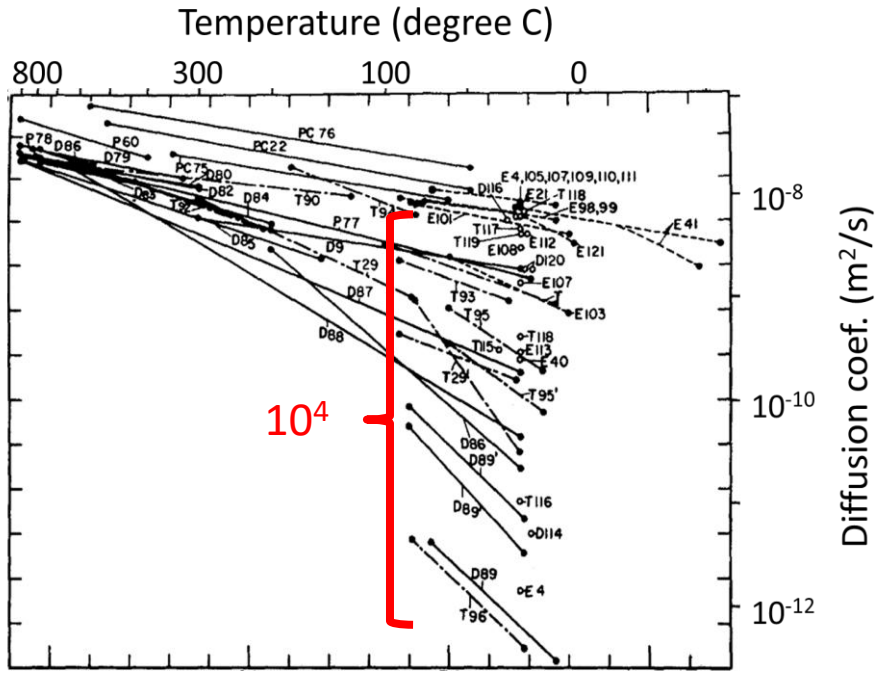
Department of Nuclear Engineering,

Seoul National University

Introduction
- Needs of accurate data of hydrogen isotopes-

- ✓ Efficient use of hydrogen isotopes (HIs), particularly tritium (T), is essential for safe/economic/sustainable operation of fusion reactors.
  - → Accurate data on HIs, such as diffusivity (D) and solubility (S) are needed.
- ✓ Experiment should be the first choice; however, there are intrinsic limitations/difficulties:
  - ✓ H lattice diffusivity at low temperatures.
    - ✓ Tritium data
  - ✓ Sticking coefficient of low-energy hydrogen (<100 eV).

Hydrogen diffusivity in bcc-Fe

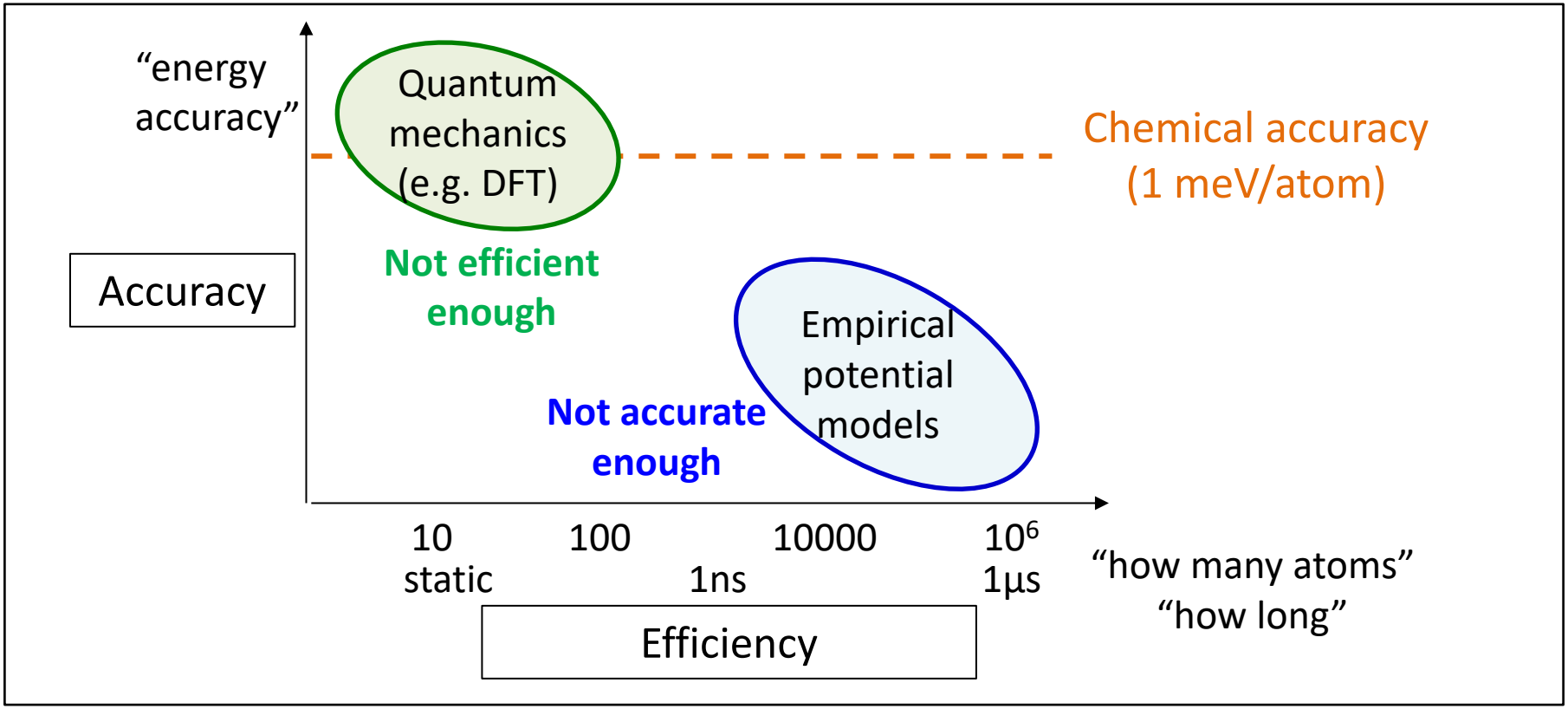


*K. Kiuchi, R.B. McLellan, Acta Mater. 31 (1983) 984.

Introduction
- Limitations of DFT and classical MD-

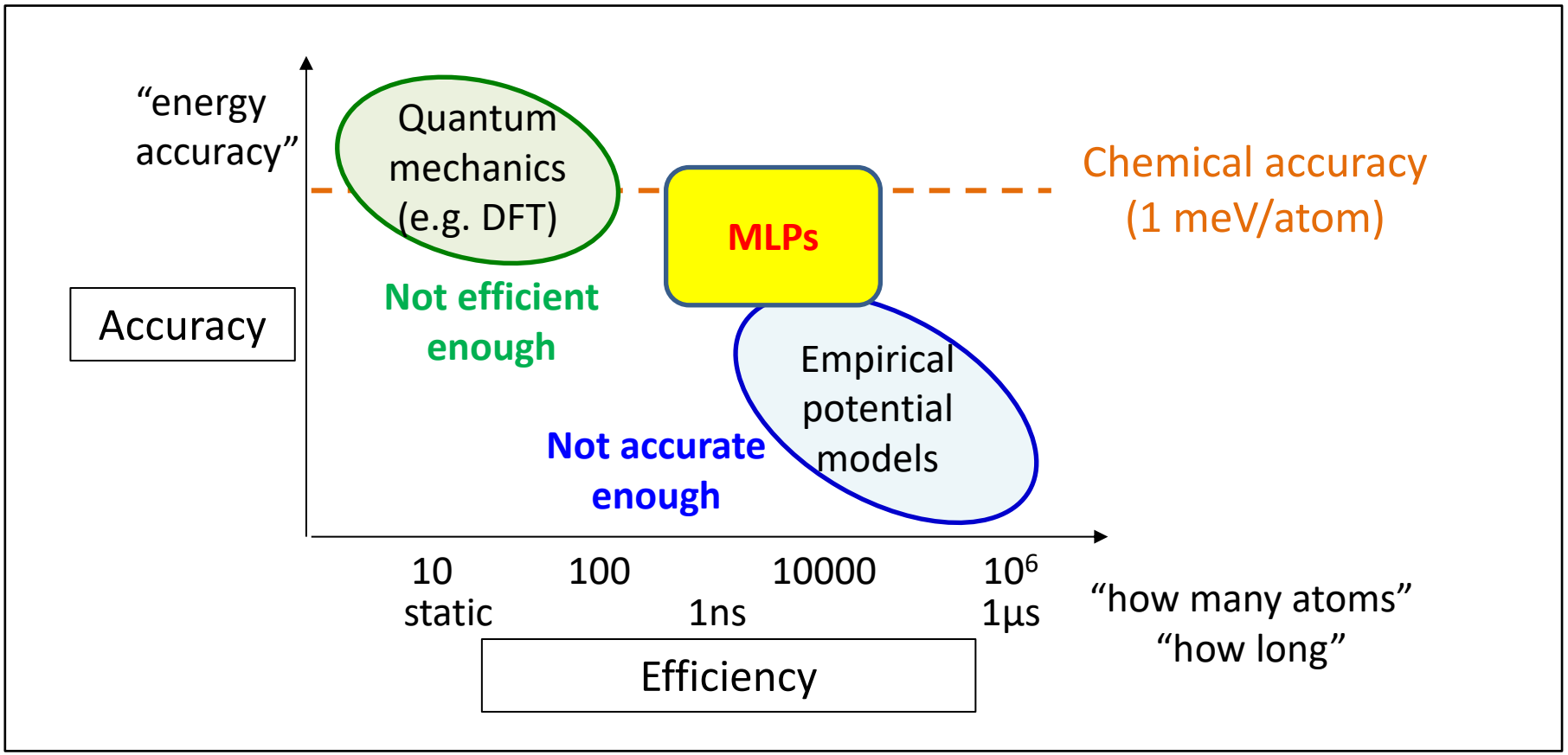
- ✓ **Atomistic simulations can be an alternative.**
  - ✓ **Quantum mechanics** (e.g., DFT) — for high accuracy
  - ✓ **Empirical potential models** (e.g., classical MD) — for computational efficiency

- ✓ **Accuracy–efficiency trade-offs limit applicability, especially in engineering.**



Introduction
- What machine learning potentials can do?-

- ✓ Machine-learning potentials (MLPs), trained on high-accuracy QM data, have substantially improved the accuracy–efficiency trade-off.
- ✓ MD simulations using MLPs (ML-MD) enables **virtual experiments**, minimizing the need for experimental validation.



Contents

< Application of advanced theoretical methods for accurate property calculations >

(Topic-1) Accurate simulations of hydrogen diffusivity in W, including isotope effects.

< Accurate large-scale simulations, i.e., virtual experiments >

(Topic-2) Simulations of PMI-related surface processes in W.

- Classical MD
- Adsorption energy by DFT
- Migration barrier by DFT

→

- "Virtual experiments" on hydrogen implantation
- Evaluation of kinetic parameters

- ✓ We adopt the Moment Tensor Potential (MTP), as it can achieve high computational efficiency with satisfactory accuracy.
- ✓ DFT by VASP; classical MD by LAMMPS; PI methods by PIMD code.

*[Moment Tensor Potential (MTP)] A.V. Shapeev, Multiscale Model. Simul. 14 (2016) 1153.
*[ML potential comparison] Y. Zuo et al., J. Phys. Chem. A 124 (2020) 731.

(1) HI diffusivity calculations
- Path-integral methods for nuclear quantum effects-

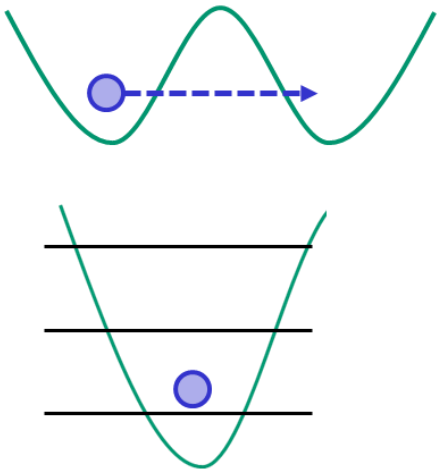
- Classical MD
- DFT + transition state theory

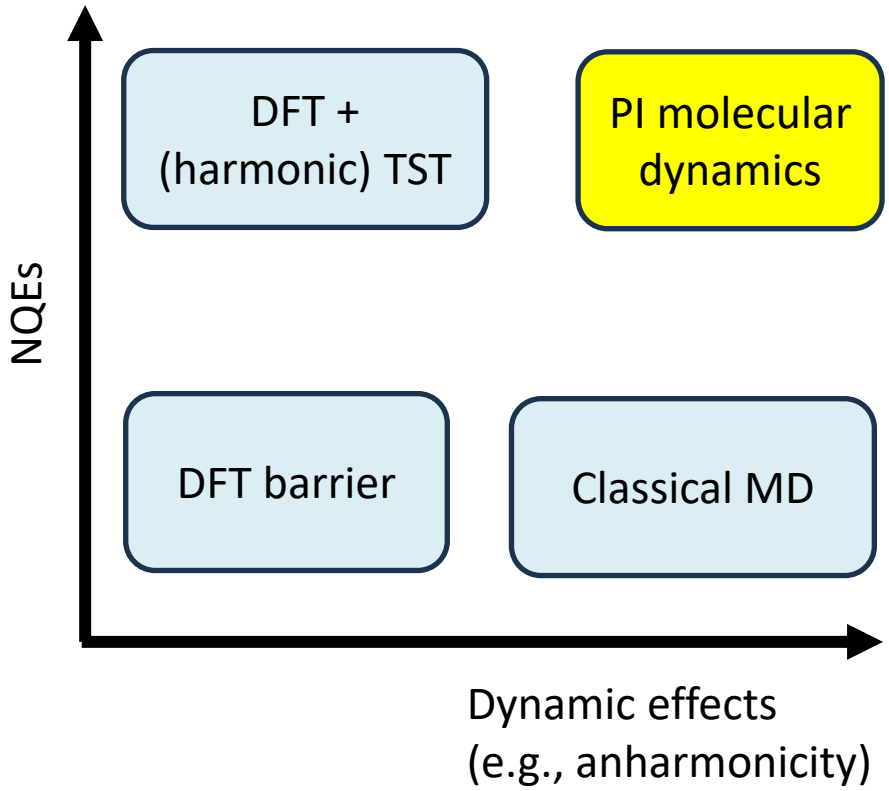
→

- Quantum dynamics

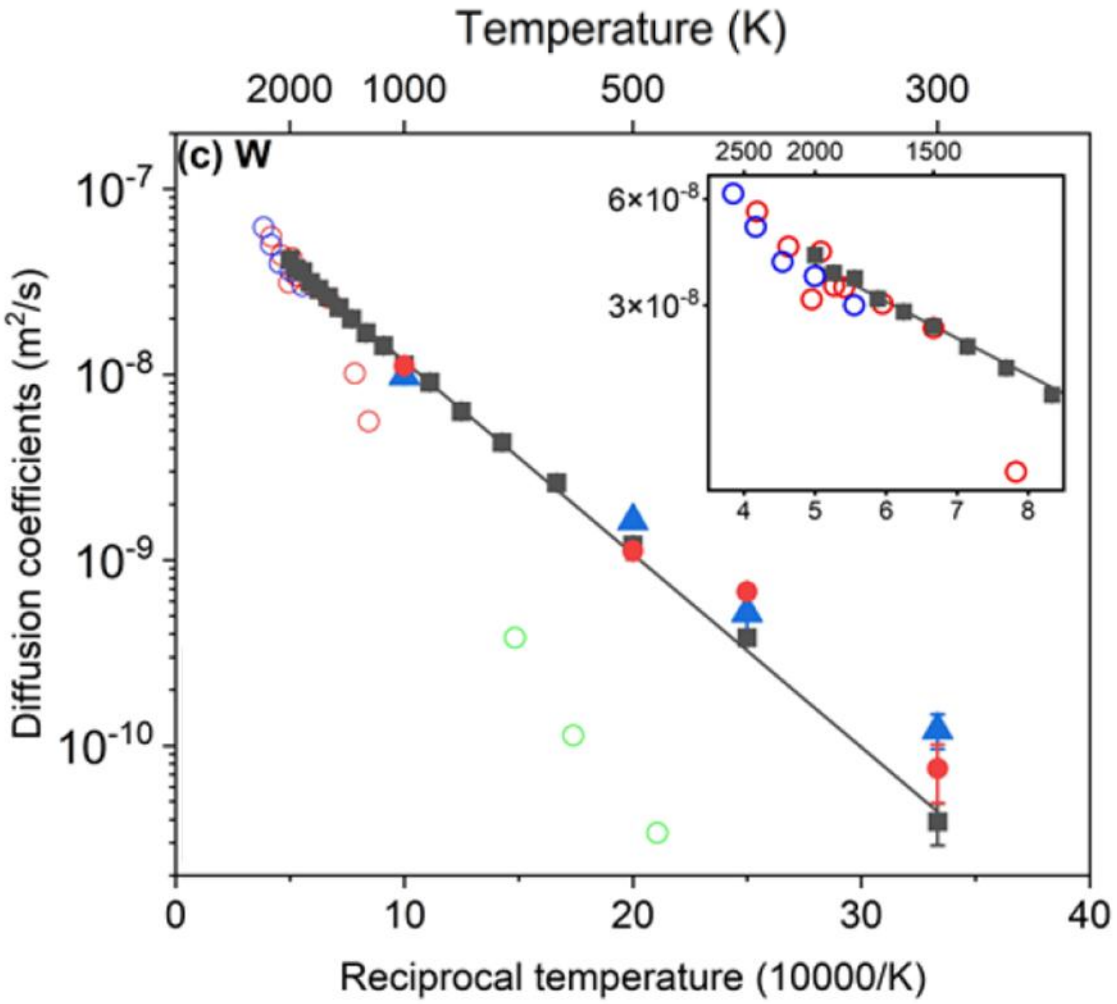
✓ MLPs allow to use path-integral (PI) methods to deal with nuclear quantum effects.
✓ If MLPs are as accurate as experiment, we can achieve accurate properties.

Nuclear Quantum Effects (NQEs)





(1) HI diffusivity calculations
- H diffusivity in bcc-W: comparison with experiments-



CLMD
CMD
RPMD
Frauenfelder
Holzner
Otsuka

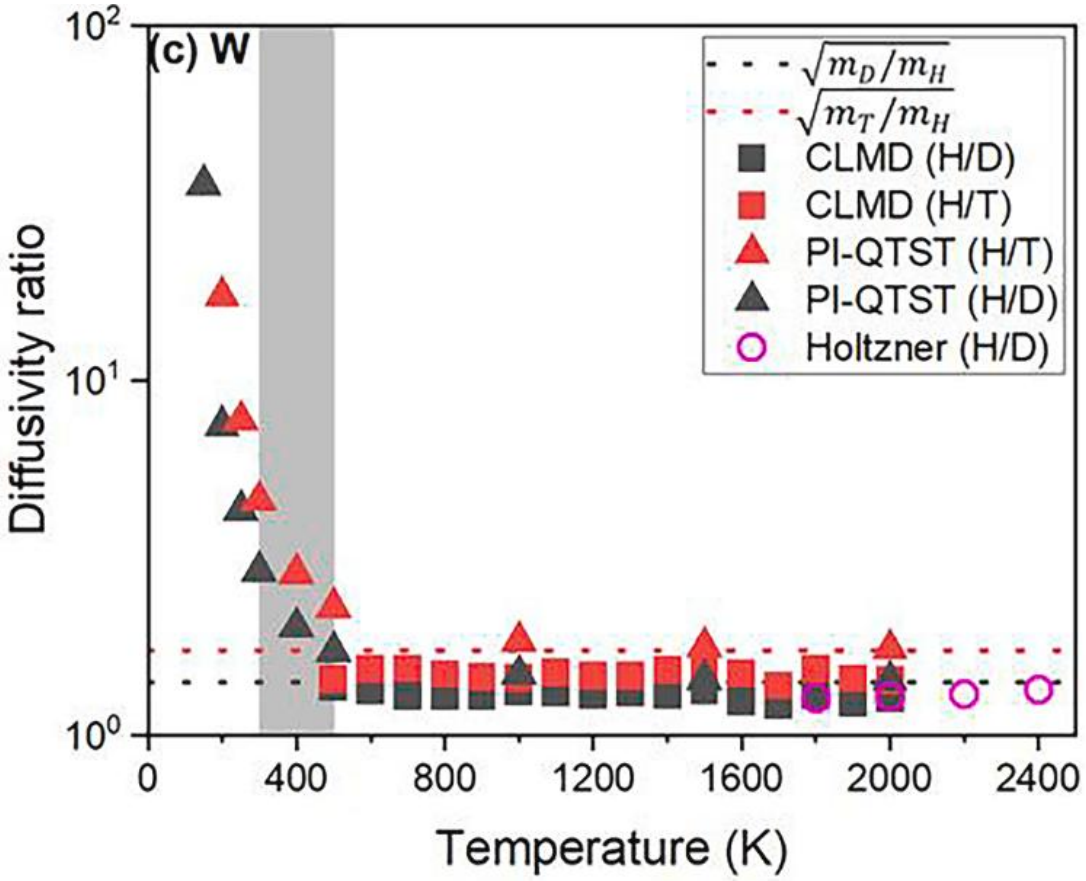
Classical MD
PI methods
Experiment

*[Experiments] Frauenfelder, J. Vac. Sci. Technol. 6 (1969) 388; G. Holzner et al., Phys. Scr. T171 (2020) 014034; T. Otsuka et al., Phys. Scr. T138 (2009) 014052.

*[Simulations] H. Kwon et l., , M. Shiga, H. Kimizuka, T. Oda, Acta Mater. 247 (2023) 118739.

(1) HI diffusivity calculations
- Isotope effects: H/D/T diffusivity in bcc-W -

- ✓ Good agreement with experiment in H/D isotope effect.
  - ✓ Not square root of mass (1.44) but ~1.26 even in the classical regime (i.e., high temp.).
- ✓ Significant deviation at low temperatures (< T_{NQEs}) due to NQEs.



Isotope effects by ML-MD at high temperatures	
D_H/D_D	1.26±0.07
D_H/D_T	1.43±0.09

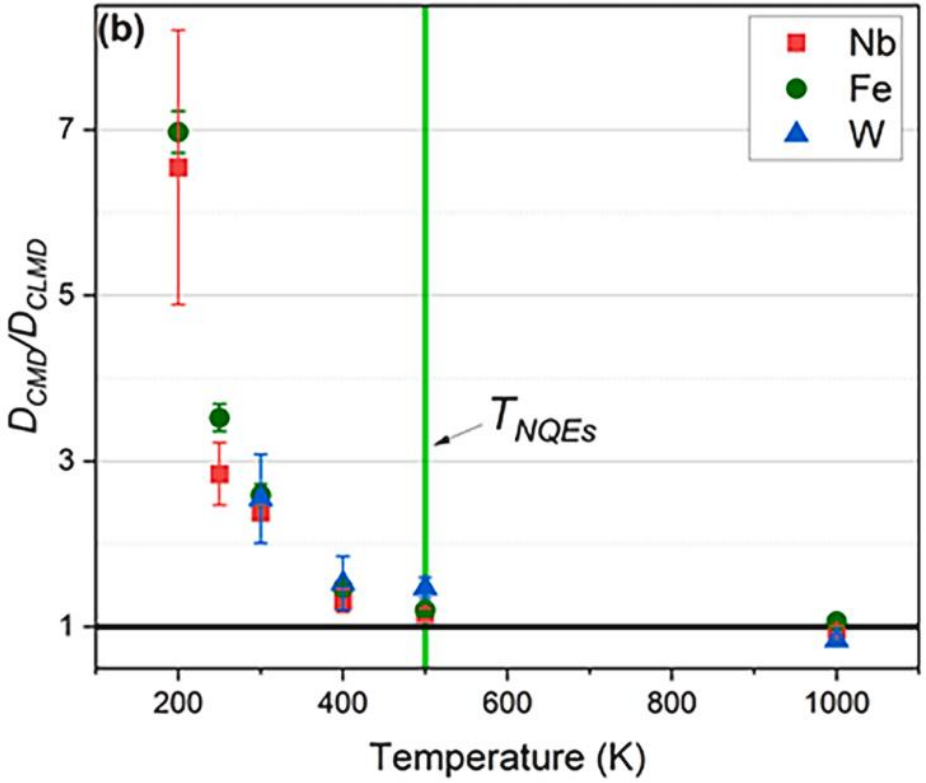
*H. Kwon, M. Shiga, H. Kimizuka, T. Oda, Acta Mater. 247 (2023) 118739.

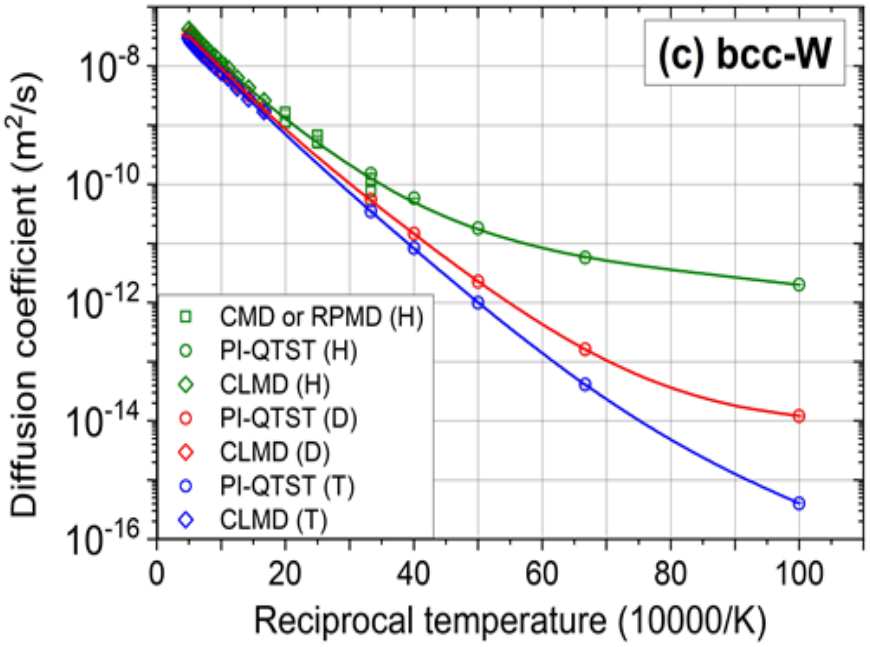
(1) HI diffusivity calculations
- for engineering applications -

- ✓ At high temperatures (> T_{NQEs}), the isotope effect model using inverse isotope masses is fairly accurate (~20% error in bcc).

$$D_T \sim \sqrt{\frac{m_T}{m_H}} D_H = \frac{D_H}{1.73}$$

- ✓ For W and Fe, we have derived equations for H/D/T diffusivity for a wide temperature range.

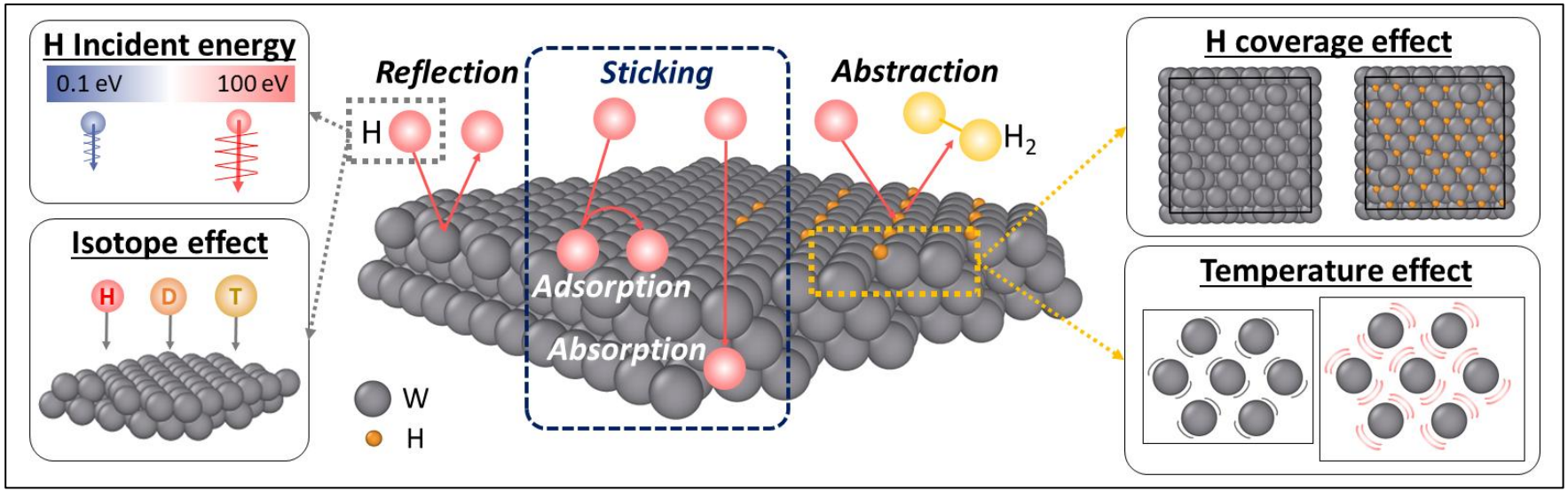




*H. Kwon, M. Shiga, H. Kimizuka, T. Oda, Acta Mater. 247 (2023) 118739.

(2) HI implantation behavior
- MLP extension for surface processes-

- ✓ ML potentials were retrained to simulate the behavior of implanted HI atoms.
  - ✓ H solution/adsorption.
  - ✓ H diffusion on surfaces.
  - ✓ H-V interaction.
  - ✓ H_2 absorption.
  - ✓ Short range collision (<100 eV).



*S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behavior
- Competition between adsorption/absorption/reflection-

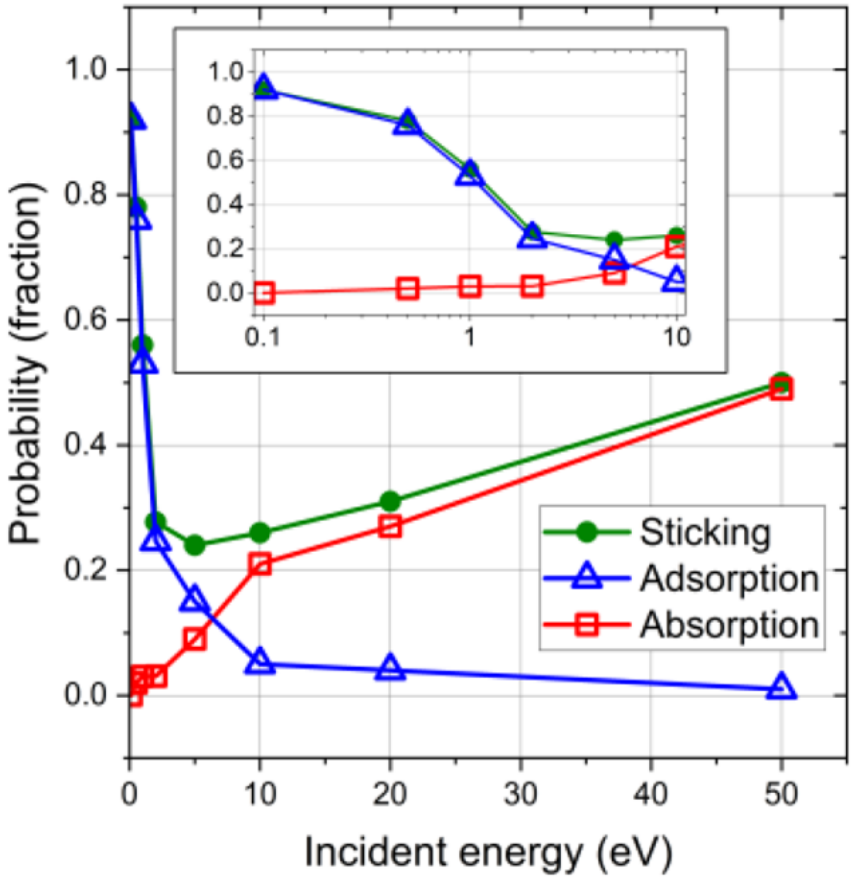
ML-MD of H implantation to W(110) at 0.1-100 eV

- ✓ **At low energies**, an implanted H cannot escape from the surface. -> **adsorption**
- ✓ **At high energies**, an implanted H cannot come back to the surface. -> **absorption**
- ✓ **At intermediate energies** -> **reflection**

➢ Sticking coefficient *S*

$$S \equiv A + P = 1 - R$$

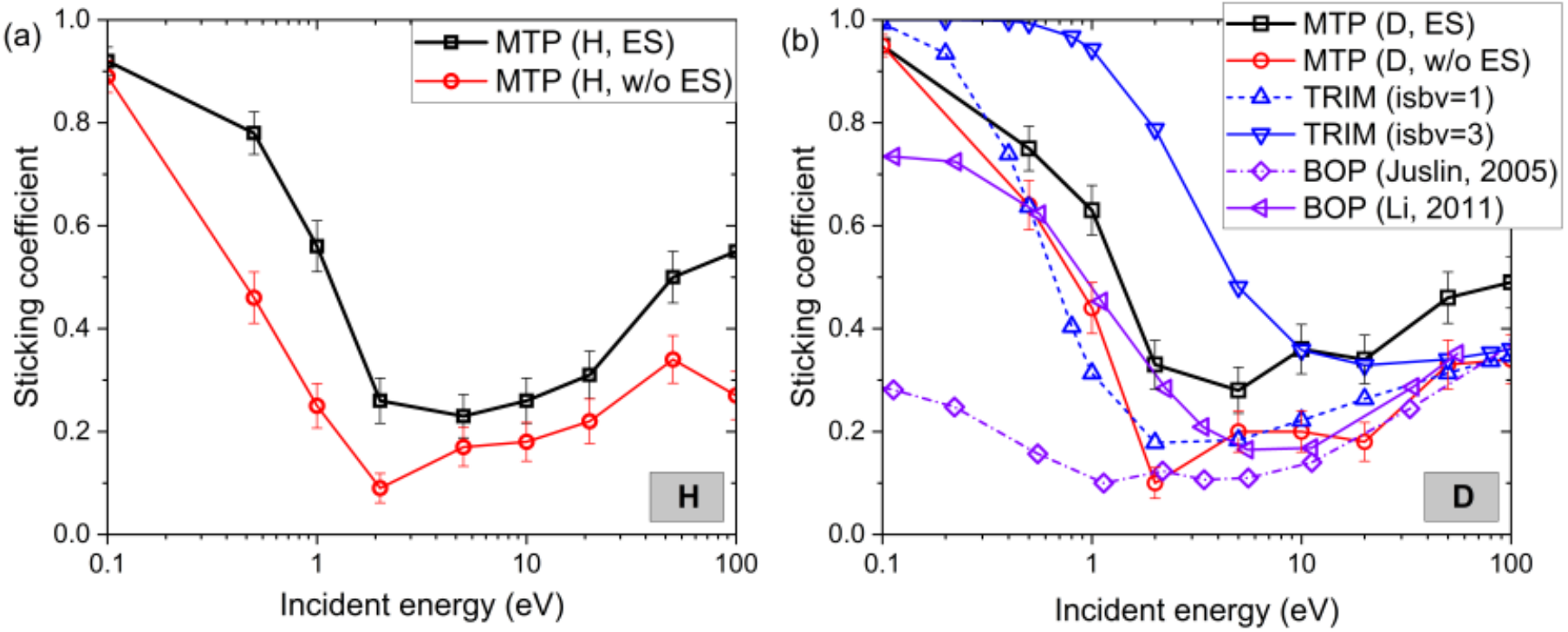
A: adsorption prob.
P: absorption prob.
R: reflection prob.



*S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behavior
- Electronic stopping, comparison with BCA and empirical MD-

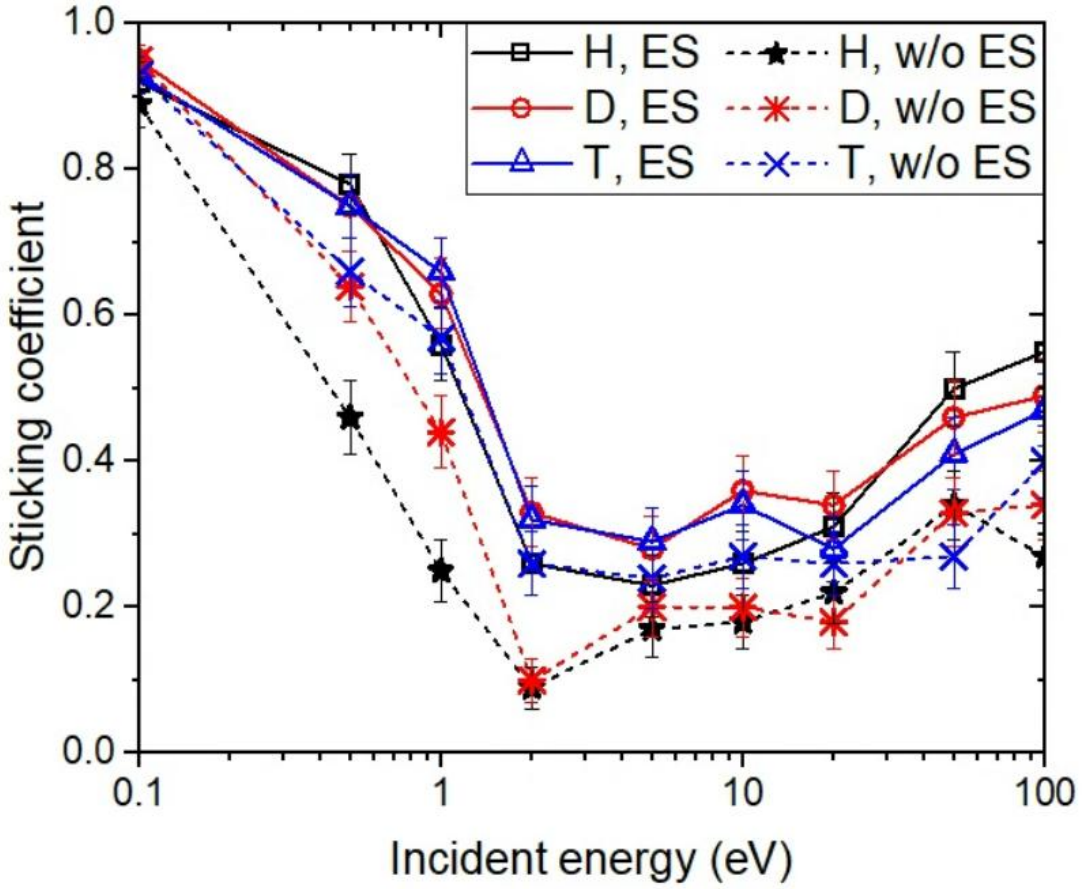
- ✓ Electronic stopping (ES) effects are significant.
- ✓ Classical MD using the bond order potentials (BOPs) are accurate at high incident energies, but not at low.
- ✓ BCA (by SDTrimSP) needs an accurate surface binding energy mode ("isbv").



*S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behavior
- Isotope effects of sticking coefficient-

- ✓ Isotope effects of nuclear process and electronic process are cancelled out, leaving only negligible isotope effects.
  - ✓ Nuclear: Larger energy loss with a heavier isotope.
  - ✓ Electronic: Larger energy loss with a lighter isotope.

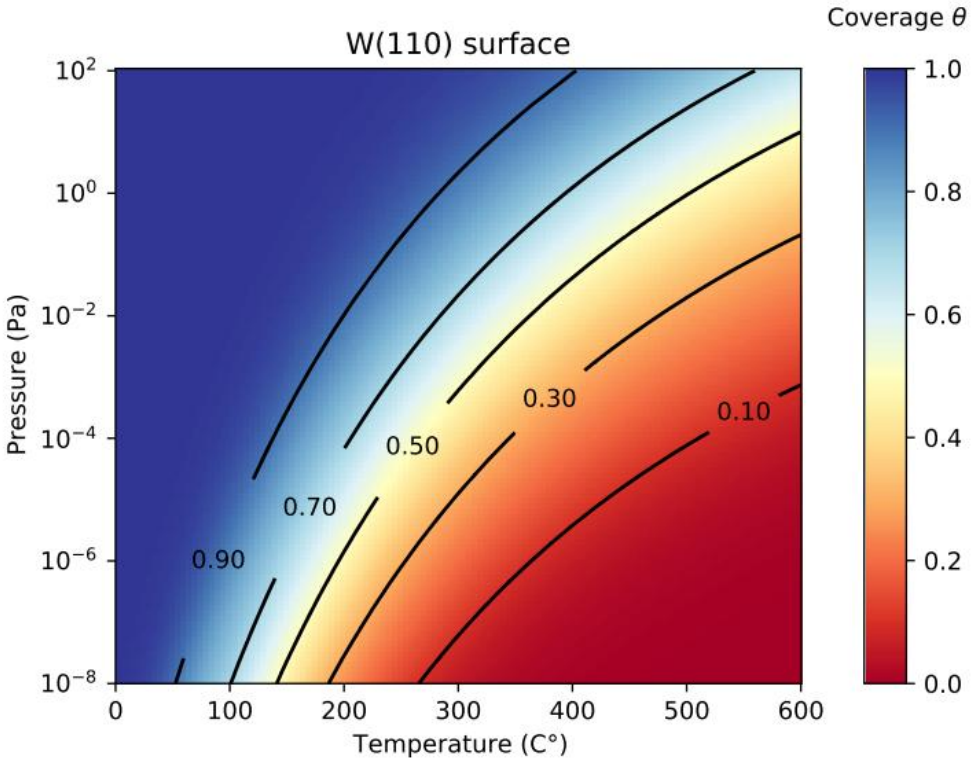


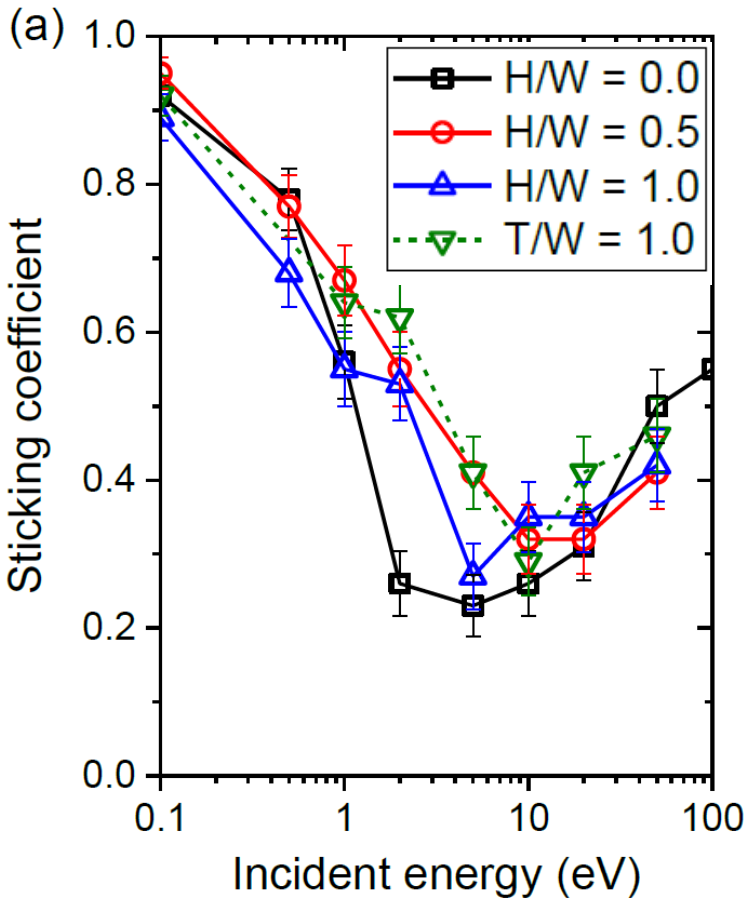
*S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behavior
- Effects of surface coverage (SC)-

ML-MD of surface coverage effects at 0.1-100 eV

- ✓ The surface coverage is likely to reduce SC due to abstraction & blocking.
- ✓ However, SC is less sensitive to the surface coverage; even increases.



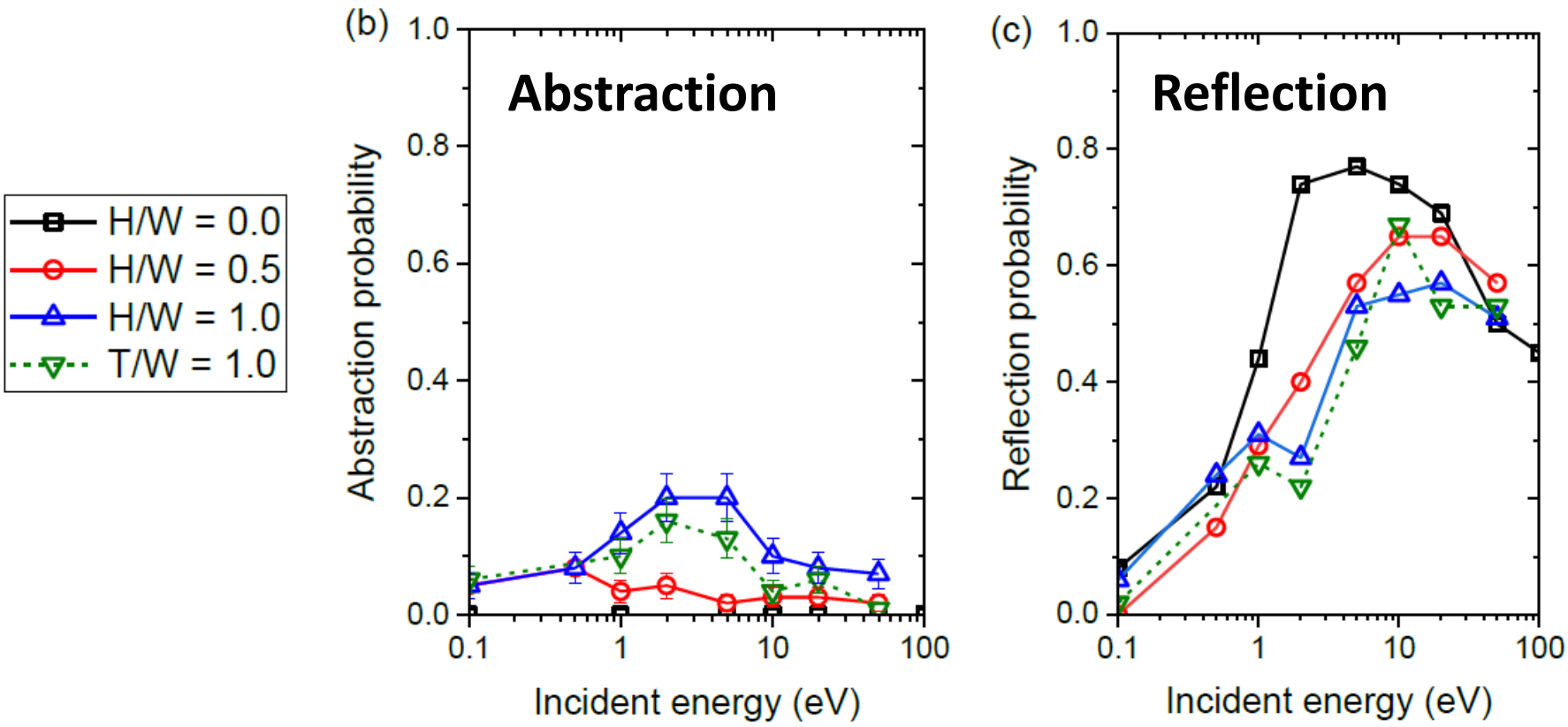


*[left figure] Z.A. Piazza et al., J. Phys. Chem. C 125 (2021) 16086.

*[right figure] S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behaviors by ML-MD
- Effects of surface coverage-

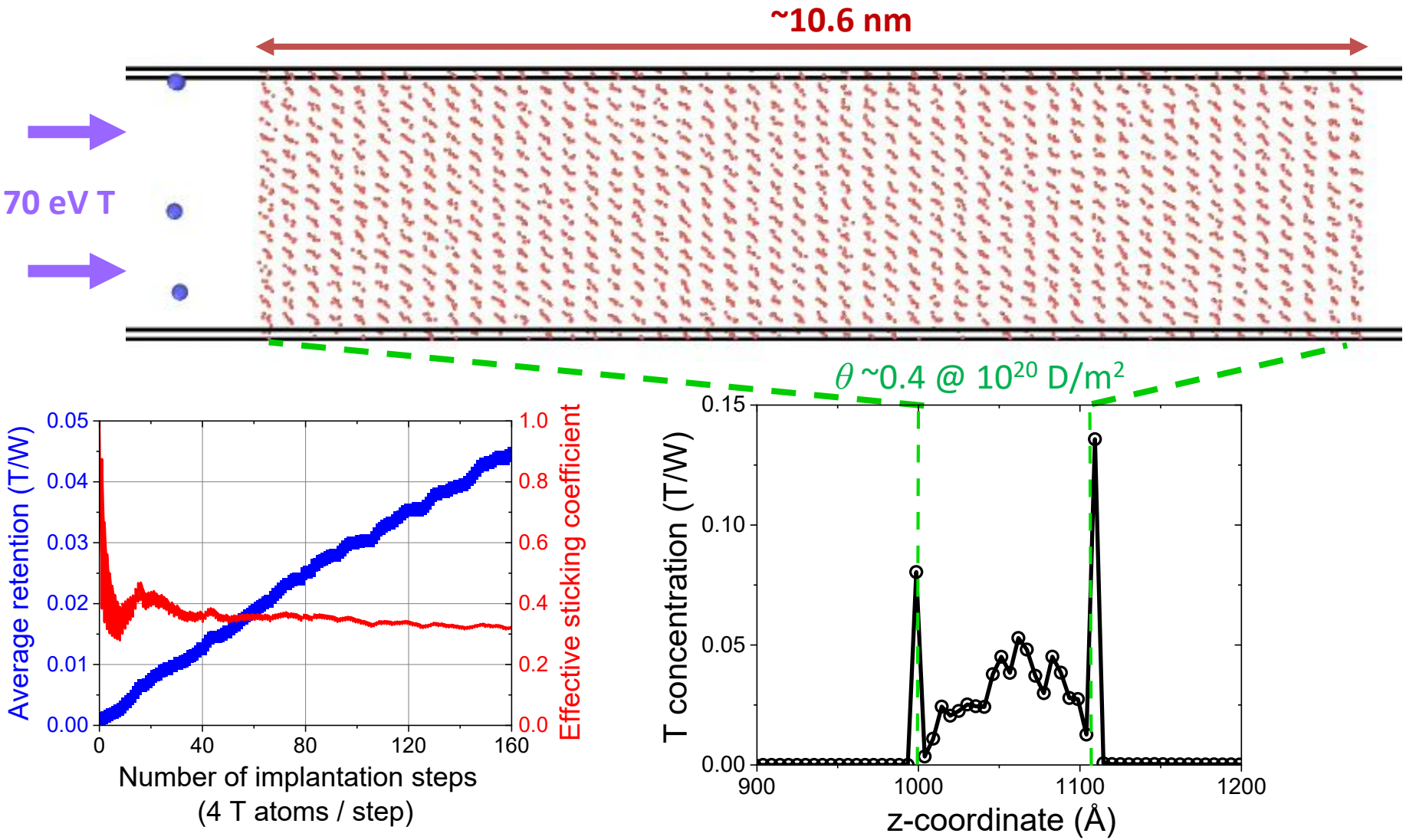
- ✓ Abstraction probability is not very high.
- ✓ Reflection probability decreases due to an additional nuclear stopping with preexisting surface H atoms.



*S. Yang, et al., (2025) [http://dx.doi.org/10.2139/ssrn.5115095]

(2) HI implantation behavior
- “large-scale” implantation simulations-

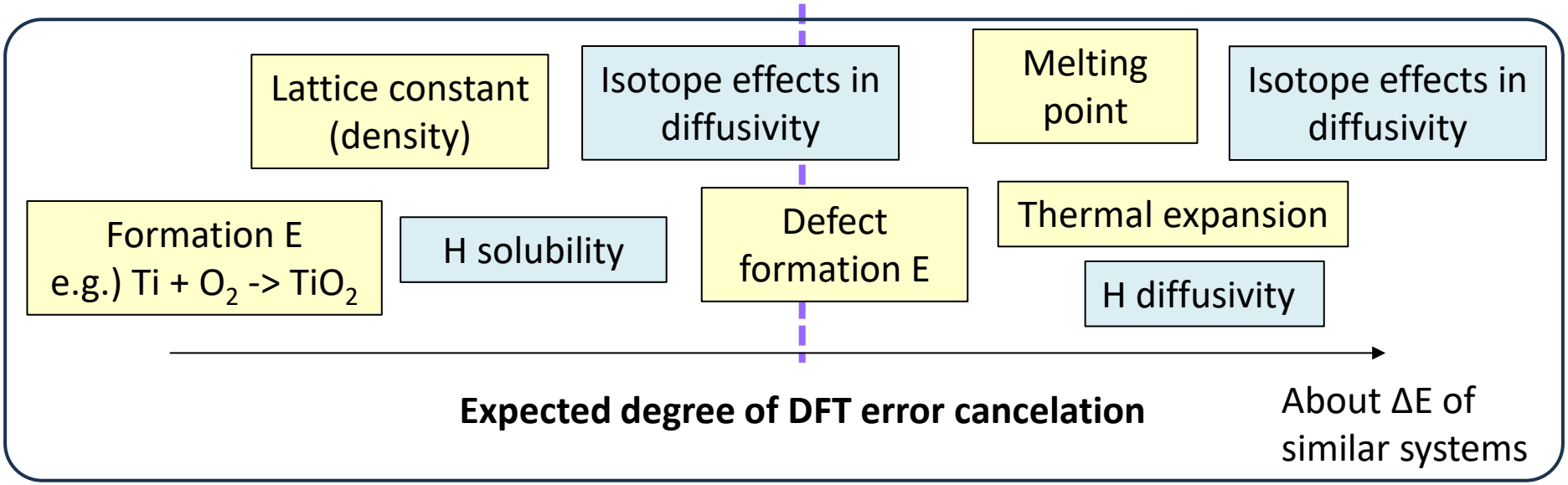
- ✓ 70 eV T implantation to a W (110) surface at 1600 K
- ✓ 12x8x24 supercells of 4608 W atoms (surface area: 6.86 nm^2)



Summary and discussion

ML-MD can evaluate material properties that cannot be directly measured by experiments (H diffusivity, sticking coefficients) at DFT accuracy.

- ✓ (Question) Can we rely on “virtual experiments” using ML-MD?
  - ✓ Depending on DFT accuracy, which is often not equal to experimental accuracy.
  - ✓ In our experiences,
    - ✓ Diffusivity: Overestimation with Mo, V / Underestimation with Fe (a factor of 2-3).
    - ✓ Solubility: Overestimation (up to 0.2 eV) with W, Fe.



- ✓ For simulations of complex phenomena, careful assessment is needed. (but how?)
  - ✓ Preparing benchmark cases for complex phenomena