Oral Presentation: 전산재료과학

2022 대한금속 · 재료학회 춘계학술대회

2022. 04. 27. (수) ~ 29. (금)

창원컨벤션센터(CECO)

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Computational analysis on quantum diffusivity of hydrogen in bcc iron: Application of machine learning interatomic potential

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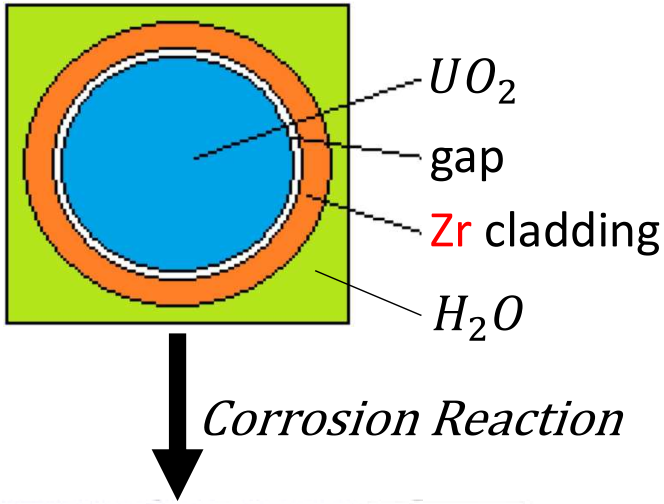
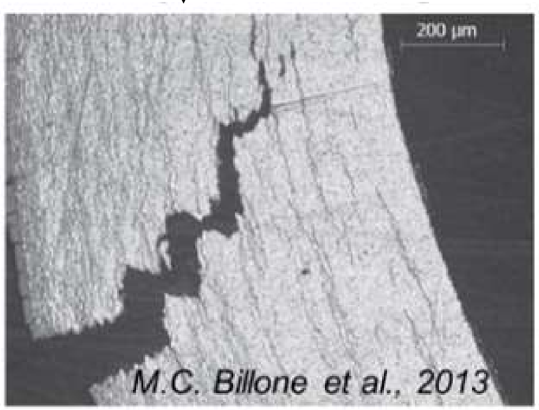
Introduction

■ Hydrogen and nuclear engineering

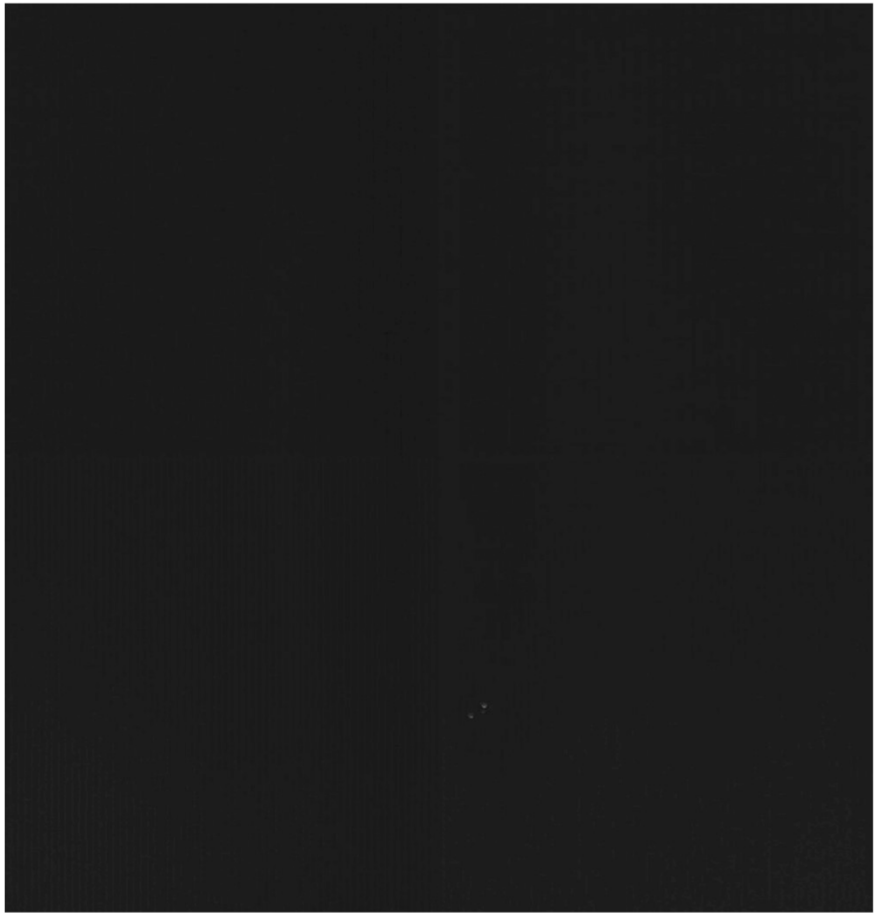
- Nuclear materials are always exposed to hydrogen gas.

Nuclear fuel rod





Corrosion reaction of Zr cladding in nuclear reactor

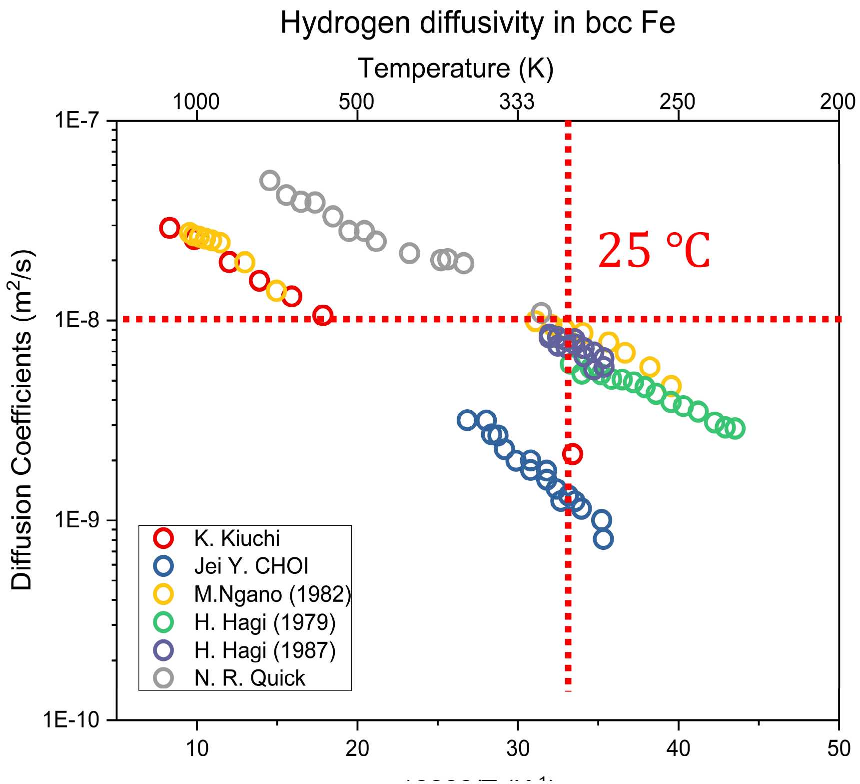


Hydrogen plasma in tokamak

Introduction

■ Hydrogen diffusivity in metals

- Generally, hydrogen diffusivity in metals is so large that their mechanical properties can be degraded.



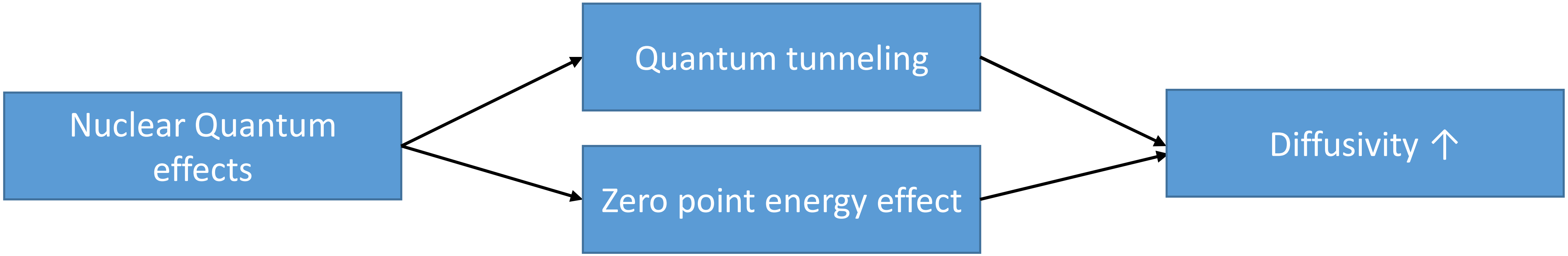
If $D \cong 10^{-8} m^2 s^{-1}$ at room temperature,

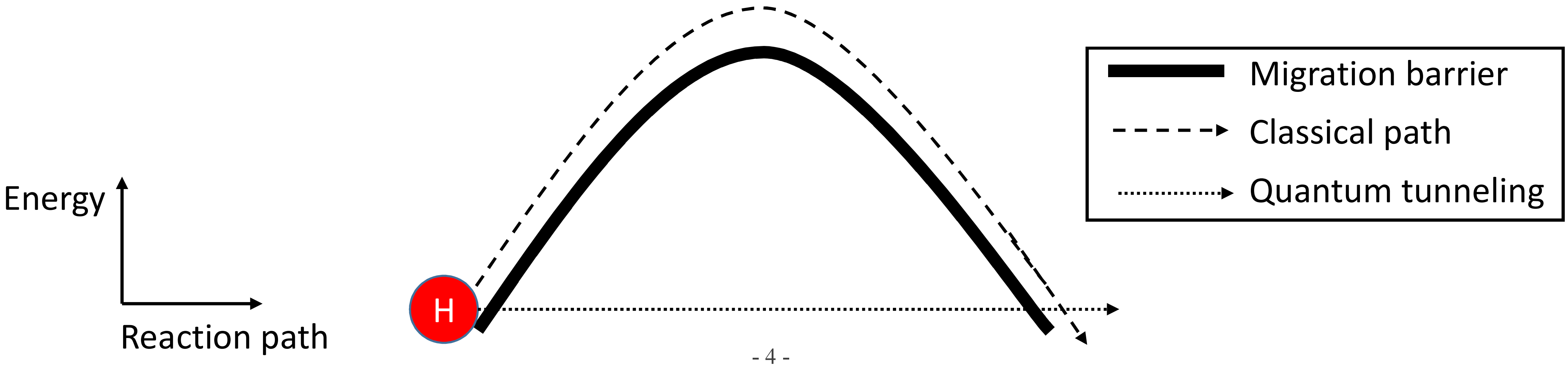
average migration distance per H atom is around **3 cm per day (1 m per month)**

Introduction

■ Nuclear Quantum Effects (NQEs)

- Light nuclei sometimes show considerable quantum effects even at room temperature.





Introduction

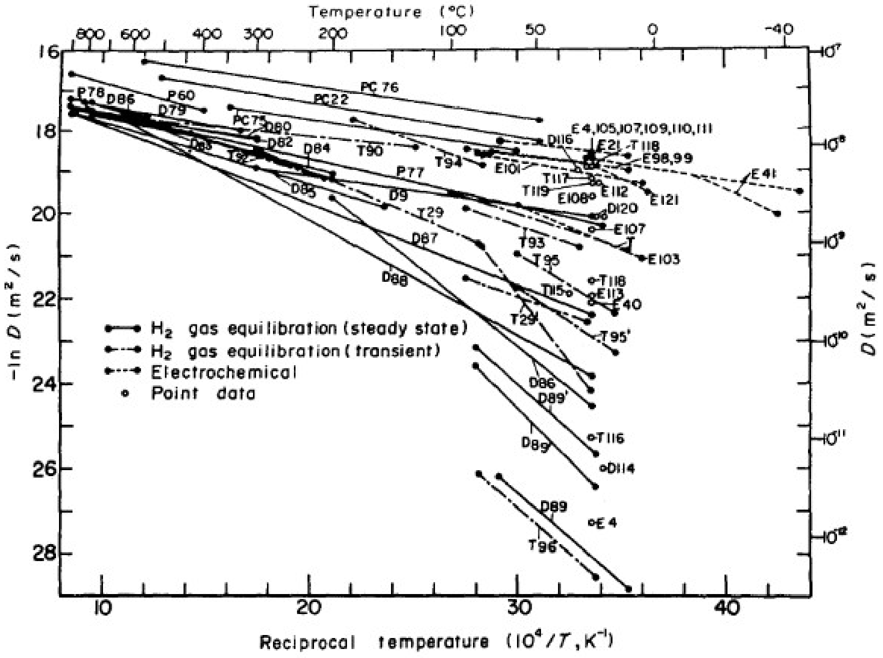
■ Preceding research about H diffusion in metals

Preceding research

Experiments: Too scattered

Computation: path integral theory

Ab-initio path integral molecular dynamics



H diffusivity in bcc iron (Ref. [7])

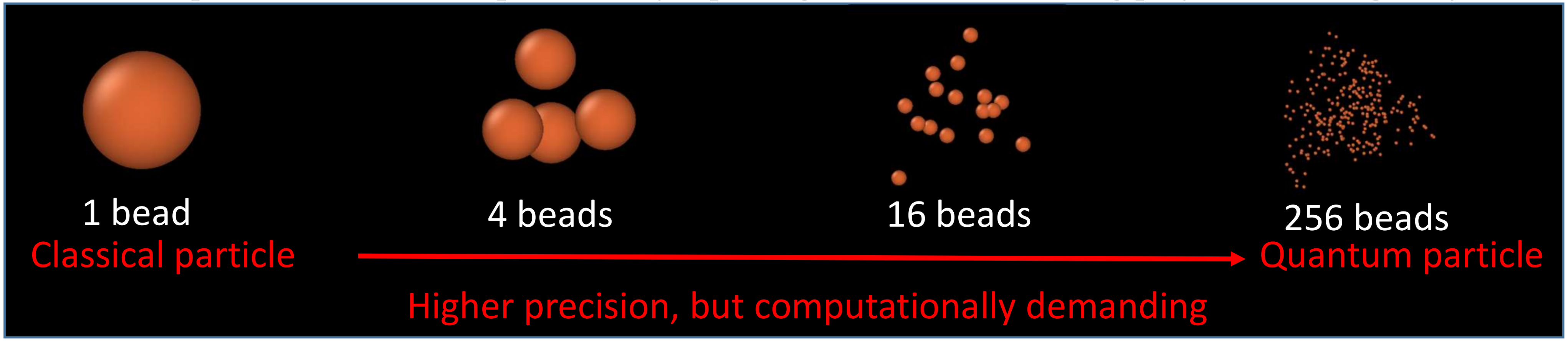
Objective

- Estimate H diffusivity in bcc metals with machine learning potential & path integral theory to reproduce nuclear quantum effects (NQEs).
- bcc Fe (α-Fe) was selected as a test case due to its high resistivity to radiation damage.

Path integral formulation

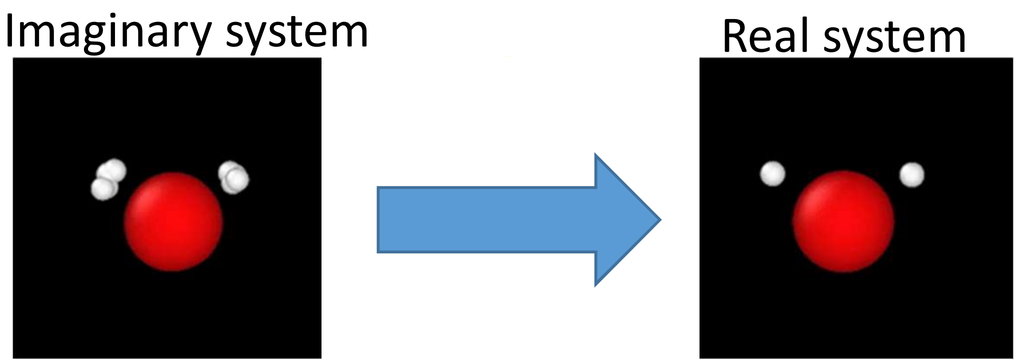
■ Classical isomorphism

- Nuclear quantum effects are reproduced by replacing **a nucleus** with a ring polymer of **imaginary beads**



■ Centroid path integral molecular dynamics (CMD)

- CMD is a approximate quantum dynamics theory for dynamic quantities in real-time.

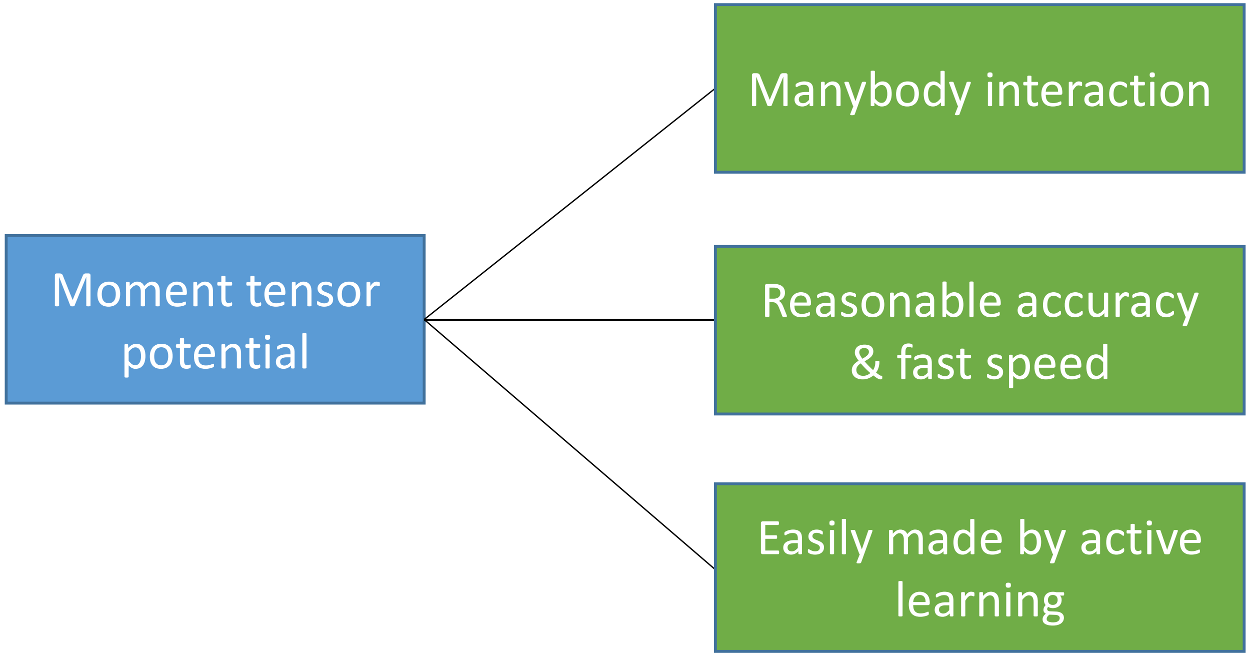


RPMD simulation of a water molecule (H_2O)

Generation of moment tensor potential (MTP)

■ Machine learning potential: Moment tensor potential (MTP)

- MTP was invented by A. V. Shapeev (2016).
- It can be trained to **imitate** a target force field (DFT-PBE) with a set of polynomials

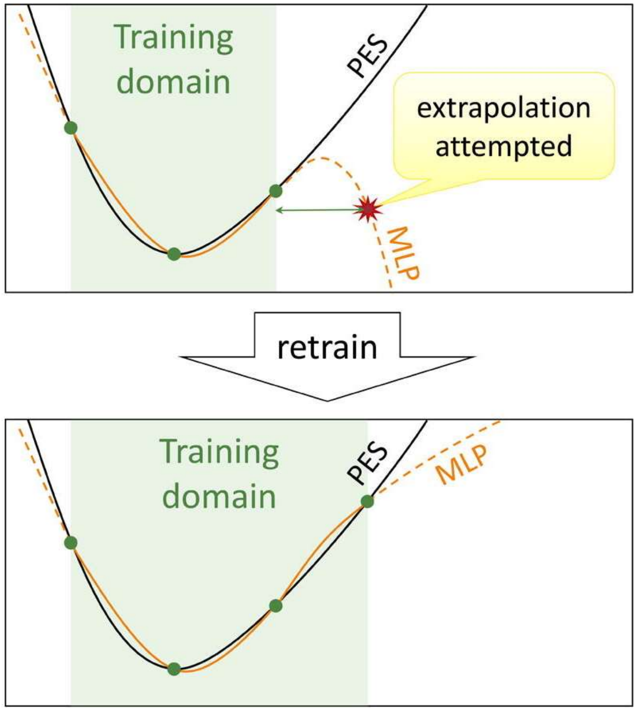


Three major advantages of MTP

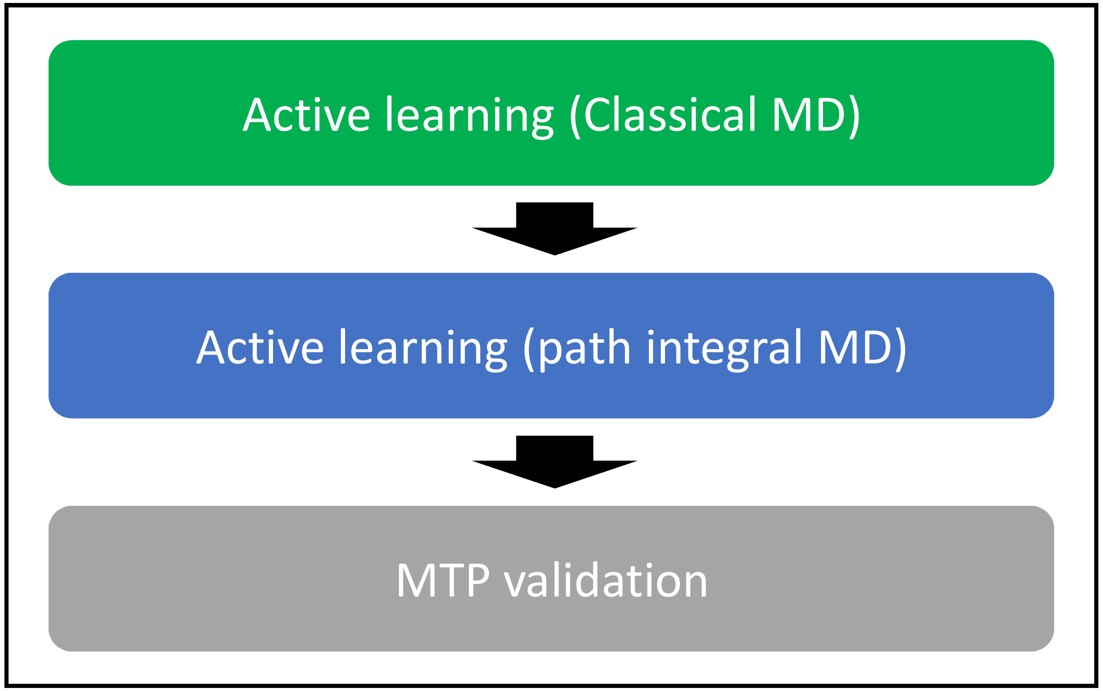
Generation of moment tensor potential (MTP)

Active learning

- The optimal training set for generating MTP was collected from **active learning.**



Active learning



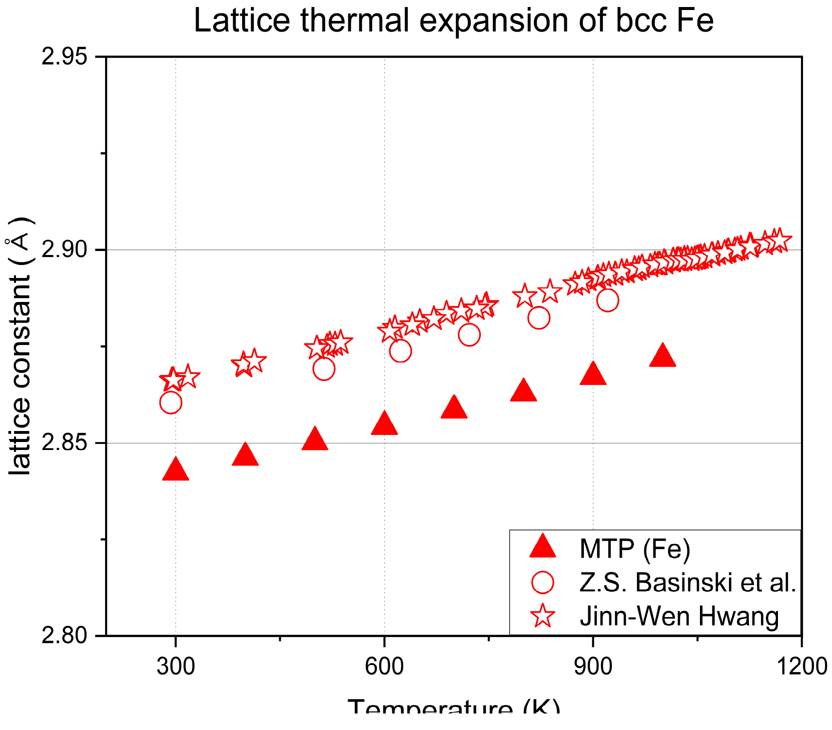
MTP generation & validation scheme

Validation of moment tensor potential (MTP)

Lattice & Elastic constants of bcc Fe without H

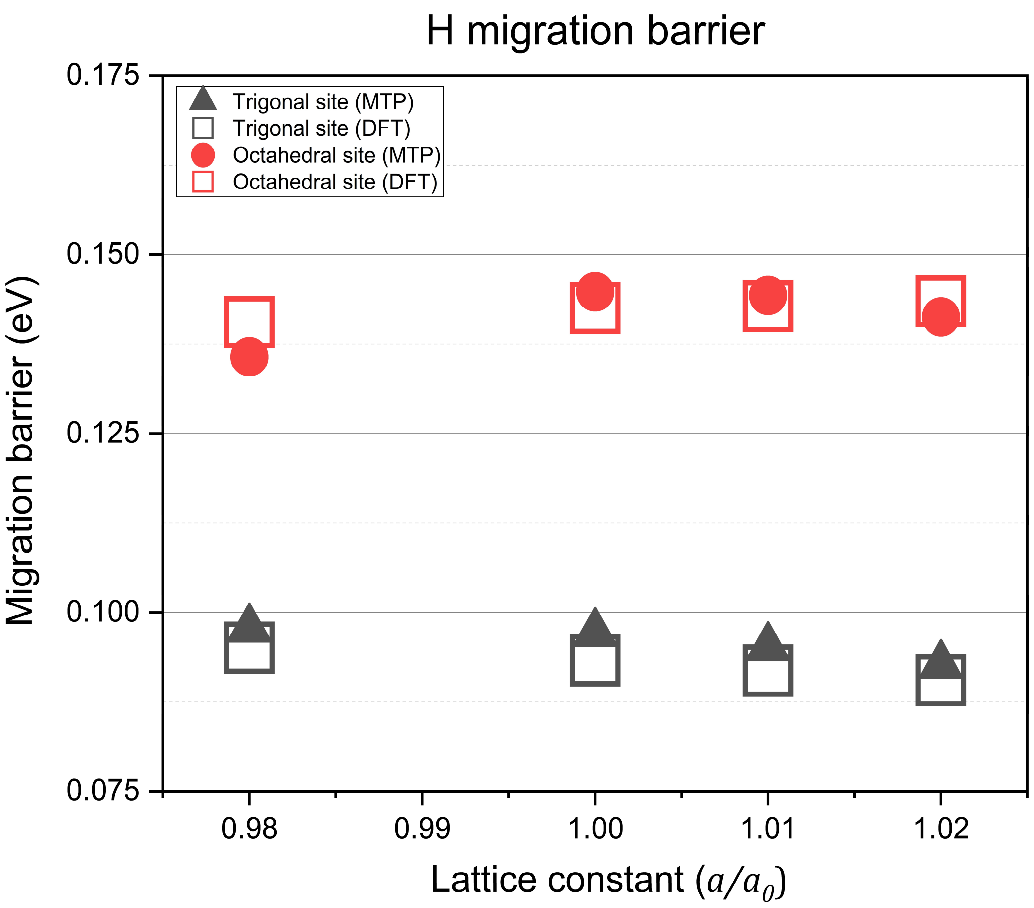
Comparison of MTP, DFT, and experiments	MTP	DFT	Experiments
Lattice constant	2.832 Å	2.832 Å	2.860 Å (293 K)
Elastic constant (C_{11})	258 GPa	278 GPa	240 GPa
Elastic constant (C_{12})	142 GPa	148 GPa	136 GPa
Elastic constant (C_{44})	96 GPa	98 GPa	131 GPa

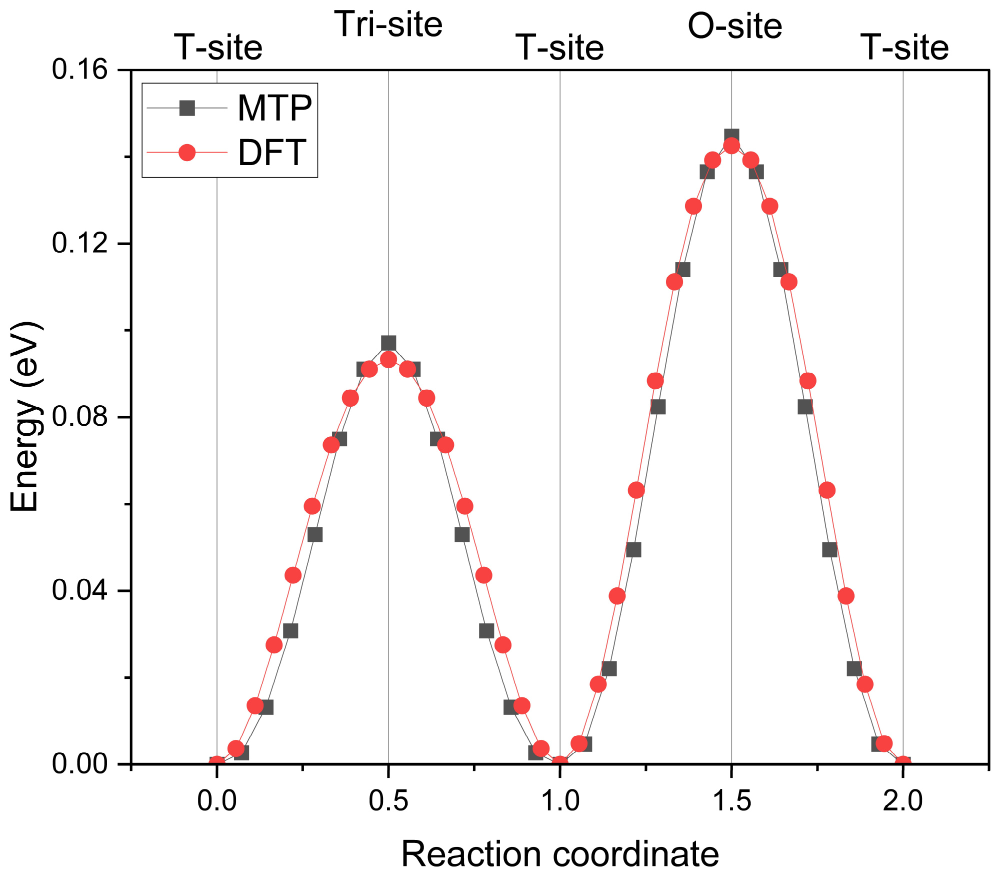
Lattice thermal expansion



Validation of moment tensor potential (MTP)

■ Migration barrier

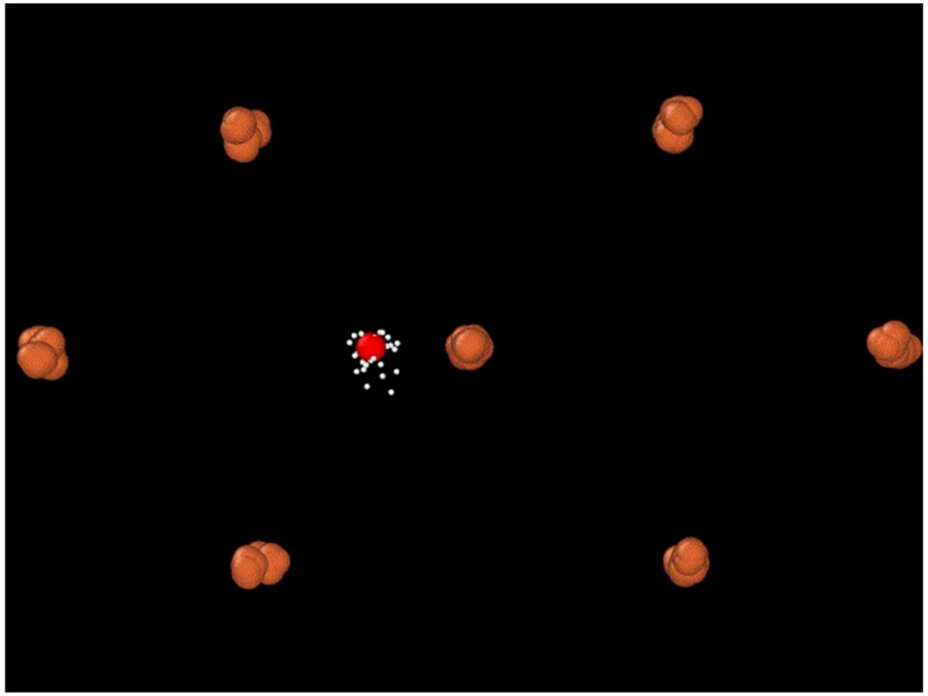




Simulation conditions

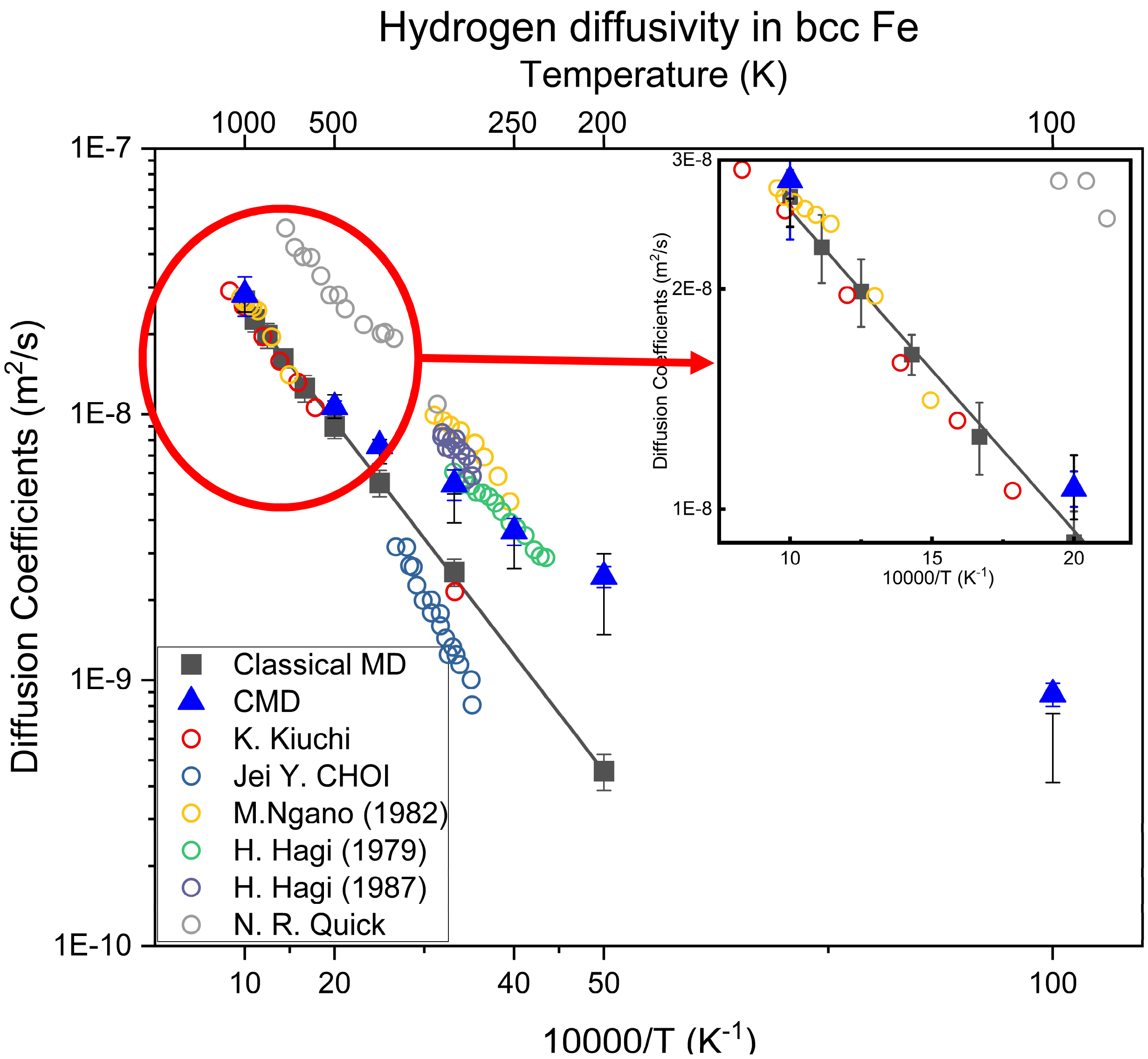
Note that
Classical Physics → classical MD
Quantum Physics → centroid path integral MD (CMD)

Simulation conditions	Experiments
Structure	one H in 4 × 4 × 4 bcc Fe (periodic boundary condition)
Ensemble	NVT
Number of imaginary beads	64
Production time	30 ns



Animation of RPMD
(red ball: centroid position, white beads: imaginary beads)

H diffusivity in bcc Fe



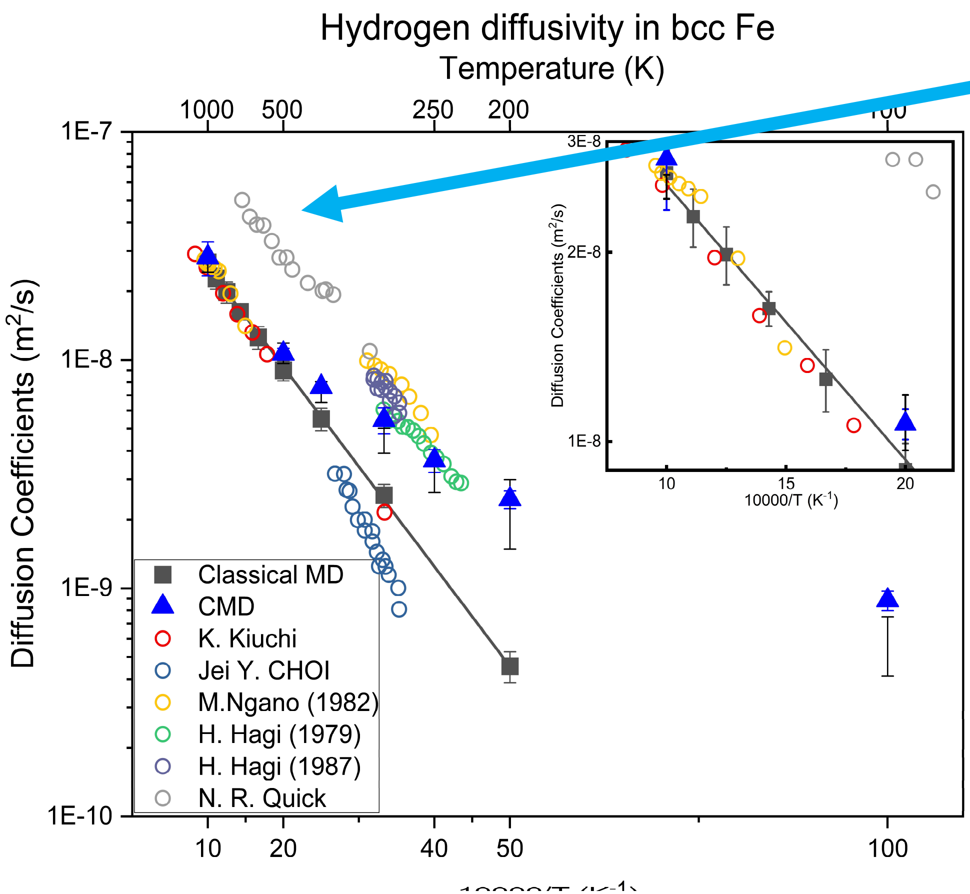
H diffusivity in bcc Fe

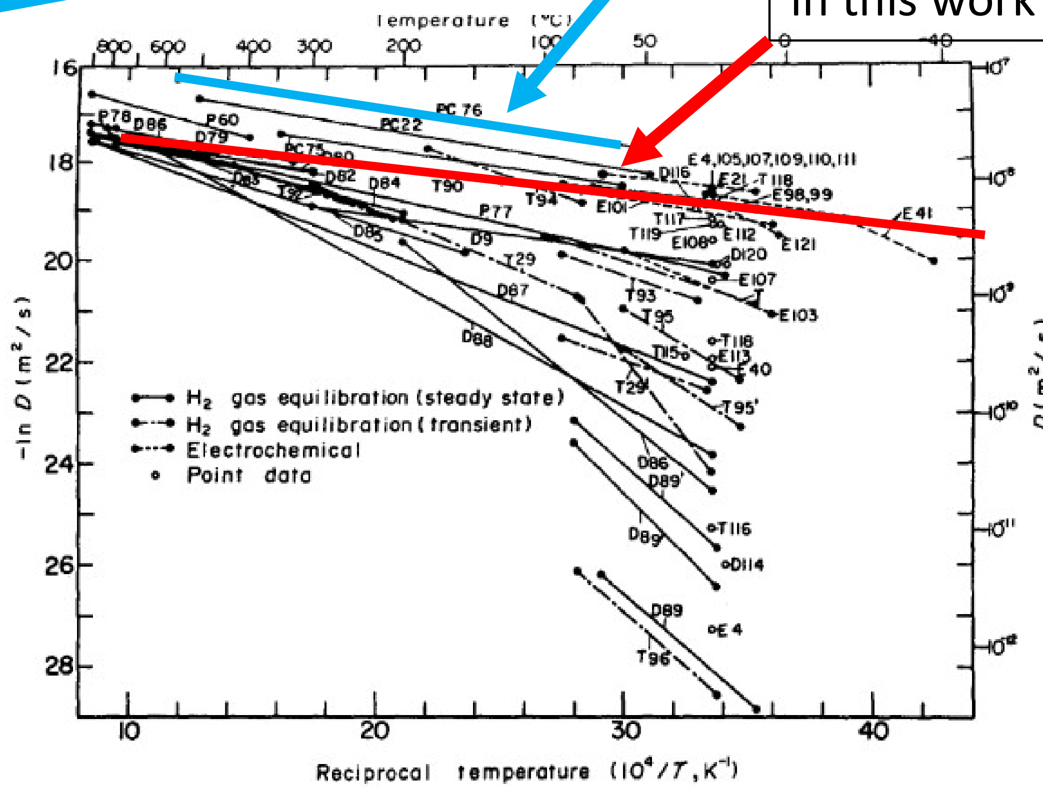
■ Diffusivity at high temperature (Classical region)

- Classical MD showed linear Arrhenius equation as the classical rate theory says.
- Centroid path integral MD (CMD) converged to classical MD over 500 K,
- Classical MD results were comparable to the K. Kiuchi's statistical estimation.

N. R. Quick

Classical MD in this work



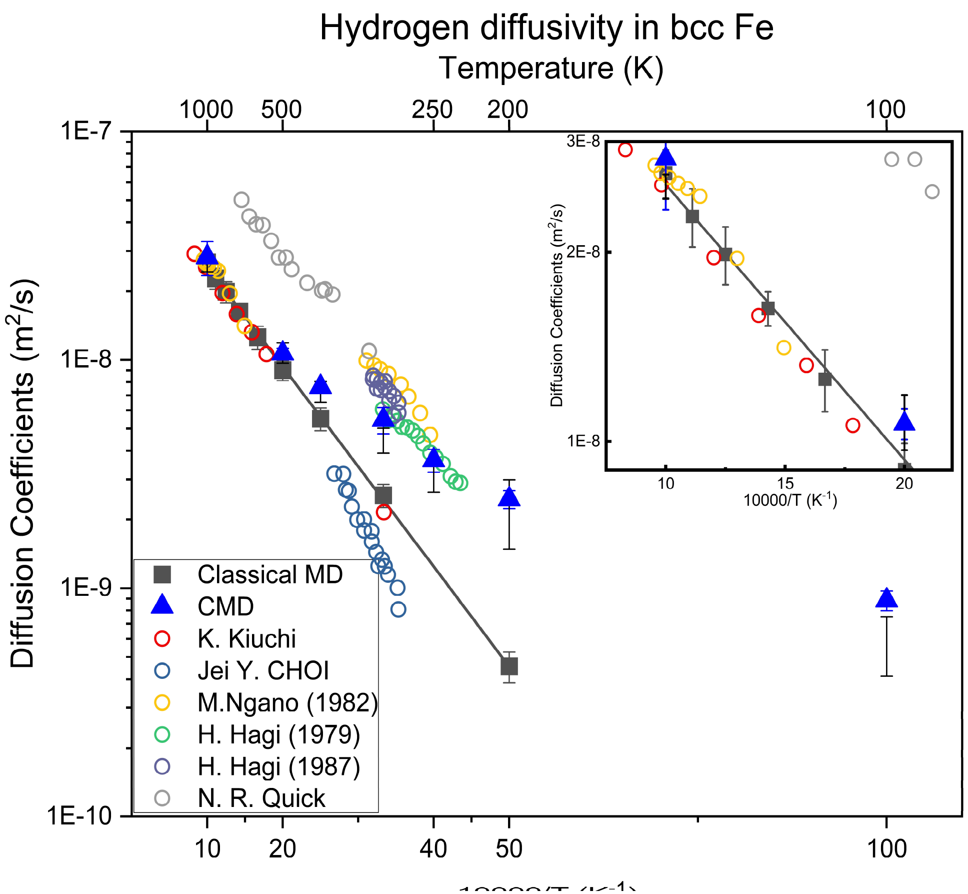


H diffusivity in bcc iron (Ref[7])

H diffusivity in bcc Fe

■ Diffusivity at low temperature (Nuclear Quantum effects)

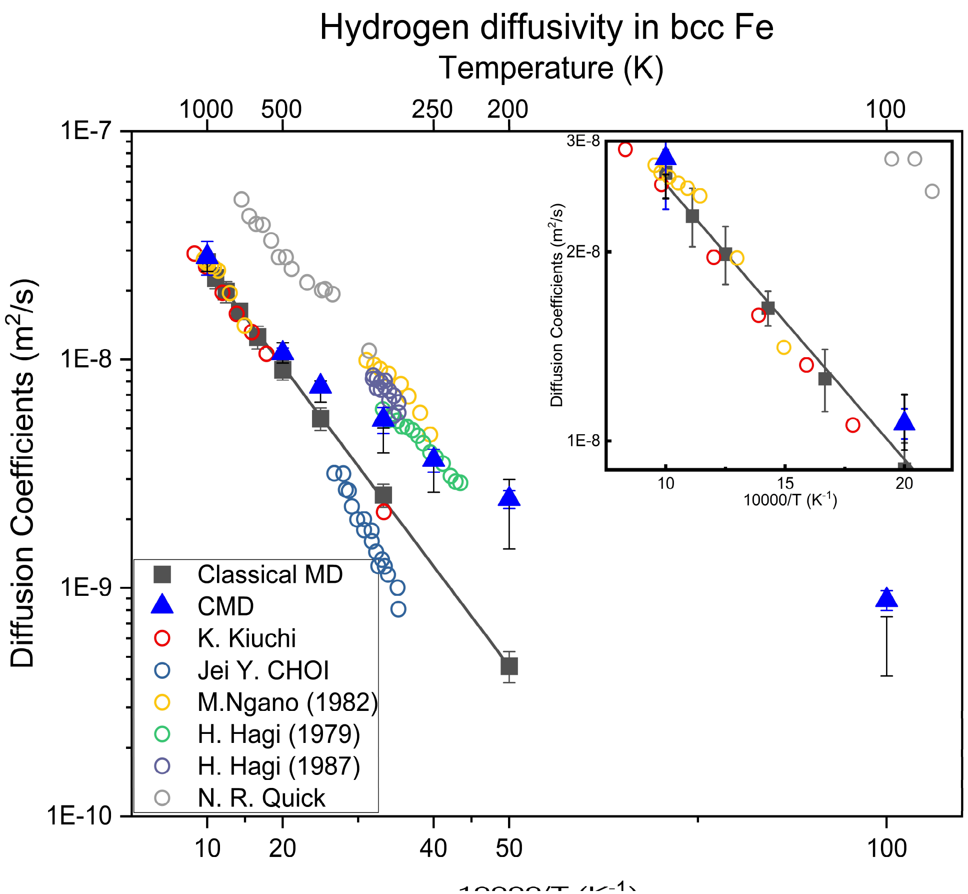
- The diffusivity difference between centroid path integral MD (CMD) and classical MD was visible **below 500 K**.
- Experimental data are too scattered due to trapping effects.

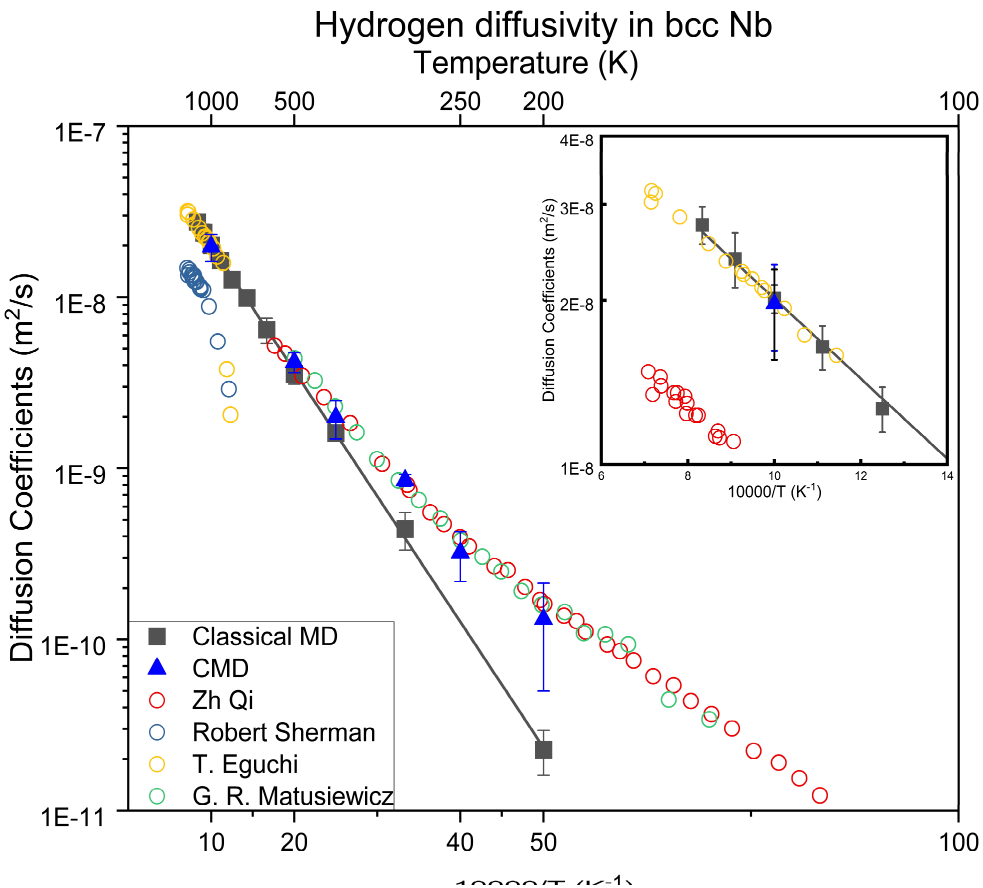


H diffusivity in bcc Fe

■ Diffusivity at low temperature (Nuclear Quantum effects)

- Similar method was applied to Nb, and it showed good agreement to experiments where data were consistent even at low temperature.





Conclusion

■ Machine learning interatomic potential

- Moment tensor potential (MTP) was generated from active learning.
- Accuracy of MTP was validated by comparing key quantities to DFT values.

■ Nuclear quantum effects

- Although experimental data are highly scattered due to trapping effects,

(T > 500 K) both classical MD and CMD were comparable to overall measurements.

(100 K <T < 500 K) CMD of H in bcc Nb showed good agreement with experiments.

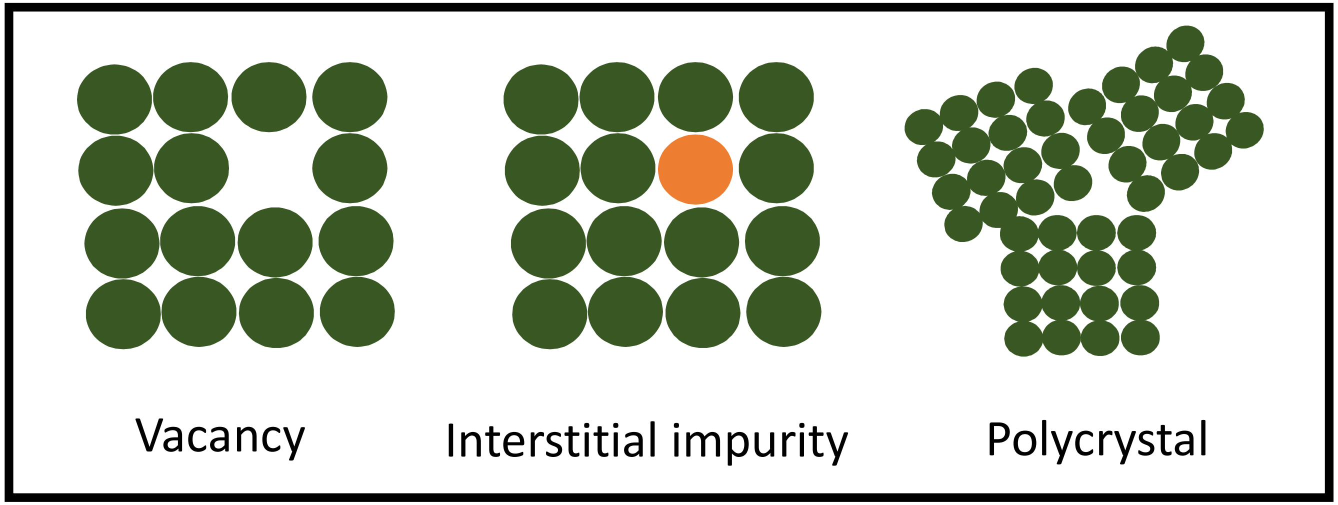
- Classical-Quantum crossover temperature was around 500 K.

    (centroid MD produced about 35 % larger diffusion coefficient than classical MD at 400 K.

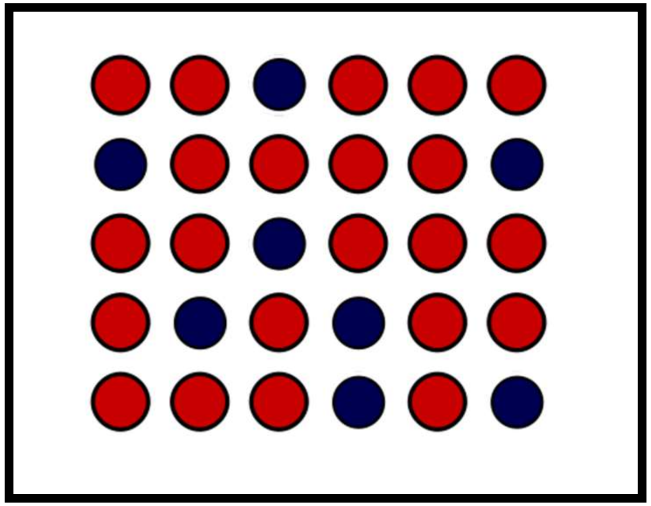
Future work

■ Potential value of machine-learning interatomic potentials

- Although only perfect bcc crystal was considered, our methodology can be further applied to more complex systems with a high level of transferability.
- Practical problems (e.g. hydrogen embrittlement) can be analyzed by this methodology.



Defected systems



Alloy

Acknowledgement

① I would like to express my deepest gratitude
to my supervisor, Prof. Oda for his careful instructions.

② Furthermore, I would like to acknowledge Dr. Shiga and Prof. Kimizuka
for their active discussion about path integral MD.

③ Finally, I am so indebted to the HPCI of Japan
for support of the supercomputer Fugaku.

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