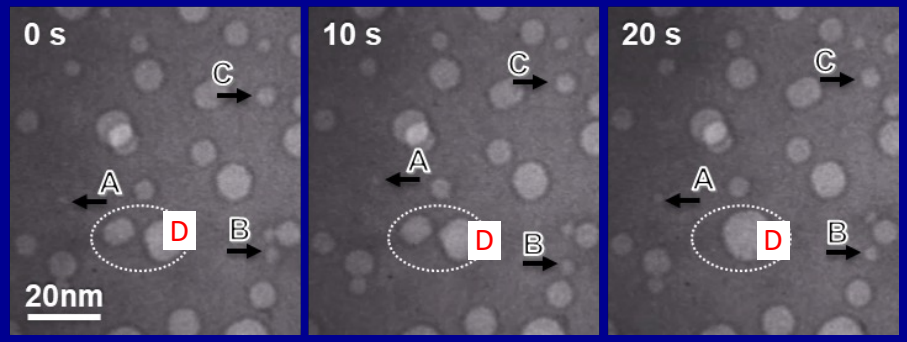
Simulations for dynamic behavior of He bubbles by on-the-fly kinetic Monte Carlo

T. Okita [1], Y. Yamamoto [1], S. Furuta [1], Y. Watanabe [2]

[1] School of Engineering, the University of Tokyo

[2] National Institutes for Quantum Science and Technology

Migration and coalescence of He bubbles



F82H, annealed at 1073 K, following the irradiation with 5 keV-He^+ at 573K [1]

(a) He bubbles diffuse randomly in the absence of interactions between bubbles (i.e. bubble A, B, C).

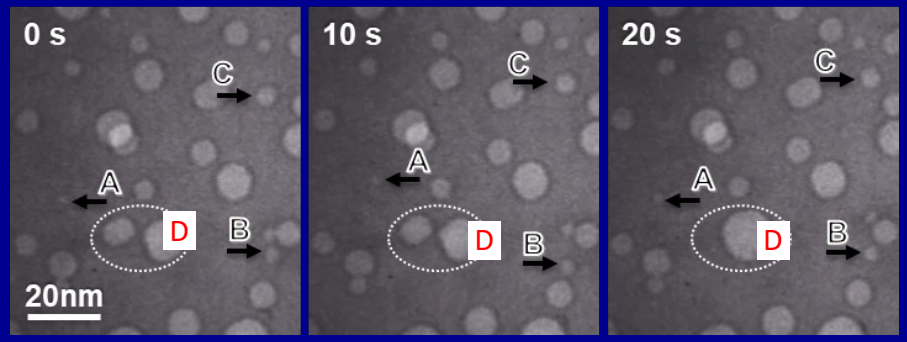
(b) When the distance between bubbles happens to become less than a certain level, they start to approach each other (i.e, bubble D).

(c) They come into contact and coalesce (i.e. bubble D @ 20s).

- He bubbles, namely aggregation of vacancies and He atoms generated through (n, α) nuclear reactions, is a characteristic microstructure of structural materials of fusion reactors.
- Conventional simplified models have investigated the evolution of He bubbles by incorporating only the migration of He atoms and vacancies (i.e. w/o migration of bubbles).
- **Since migrations of He bubbles has a great influence on the evolution of microstructure and resultant changes in mechanical properties, it is necessary to elucidate the mechanism and to construct a model that incorporates the processes in detail.**

[1] M. Miyamoto et al., ICFRM19 (2019)

Migration and coalescence of He bubbles



F82H, annealed at 1073 K, following the irradiation with 5 keV-He^+ at 573K [1]

(a) He bubbles diffuse randomly in the absence of interactions between bubbles (i.e. bubble A, B, C).

(b) When the distance between bubbles happens to become less than a certain level, they start to approach each other (i.e. bubble D).

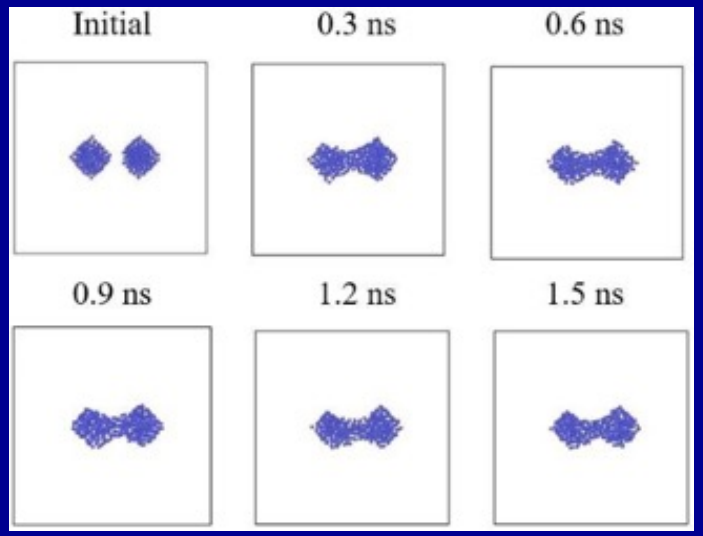
(c) They come into contact and coalesce (i.e. bubble D @ 20s).

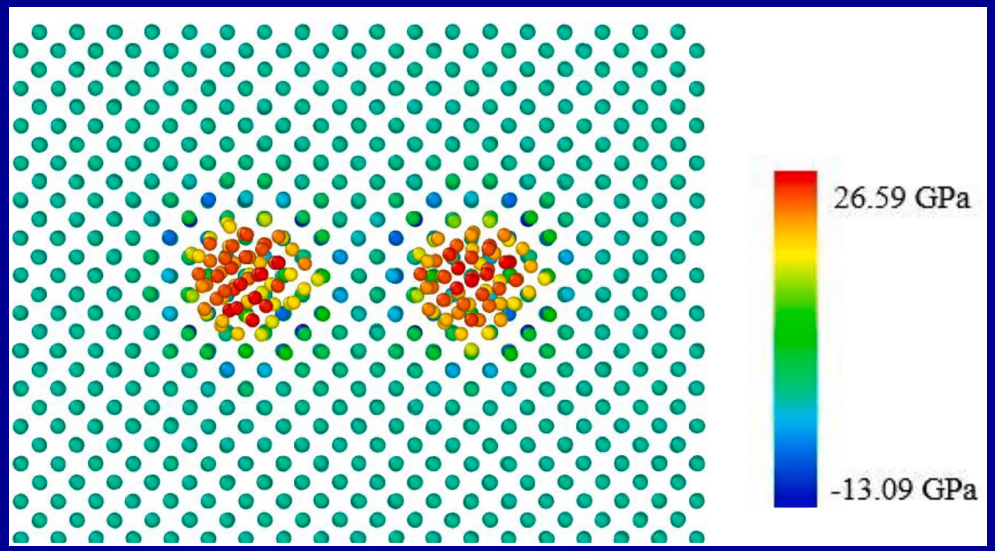
- He bubbles, namely aggregation of vacancies and He atoms generated through (n, α) nuclear reactions, is a characteristic microstructure of structural materials of fusion reactors.
- Conventional simplified models have investigated the evolution of He bubbles by incorporating only the migration of He atoms and vacancies (i.e. w/o migration of bubbles).
- Since migrations of He bubbles has a great influence on the evolution of microstructure and resultant changes in mechanical properties, it is necessary to elucidate the mechanism and to construct a model that incorporates the processes in detail.

[1] M. Miyamoto et al., ICFRM19 (2019)

MD simulations of dynamic behaviors of bubbles in W

Diameter : 1.27 nm
Initial distance : 1.59nm
2100 K, He/v = 3





Contact can occur even on the order of ns, when the distance between bubbles is short.

[2] J. Zhan et al., 2021

Typical timescale for MD simulations

Since MD simulations accurately treat atomistic vibrations that occur in the period of 10^{-13} s, the timestep is normally set to the order of 10^{-15} s.

There is a limit to the timescale that can be reproduced by MD simulations.

Waiting time, t for a phenomenon with activation energy E_a

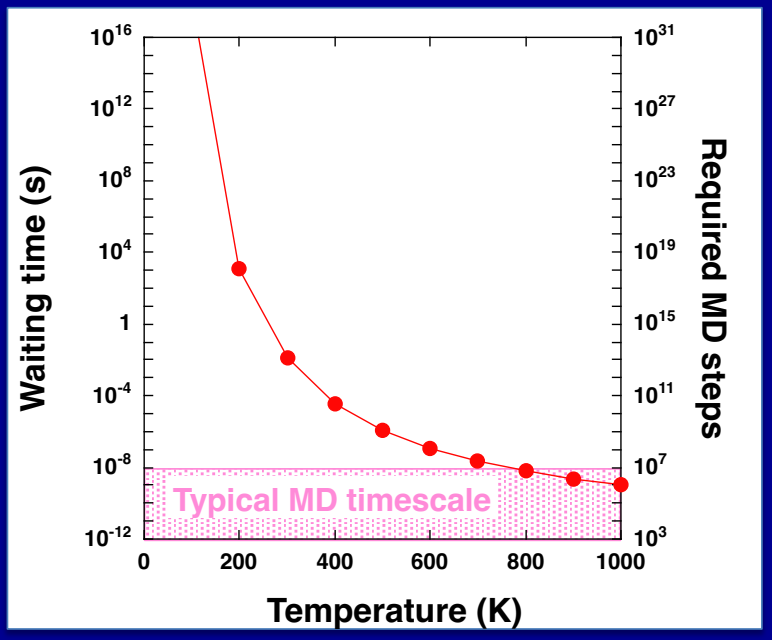
$$t = \nu_0^{-1} \exp\left(\frac{E_a}{kT}\right)$$

ν_0 : frequency factor [1/s]
k : Boltzmann's constant
T : Temperature

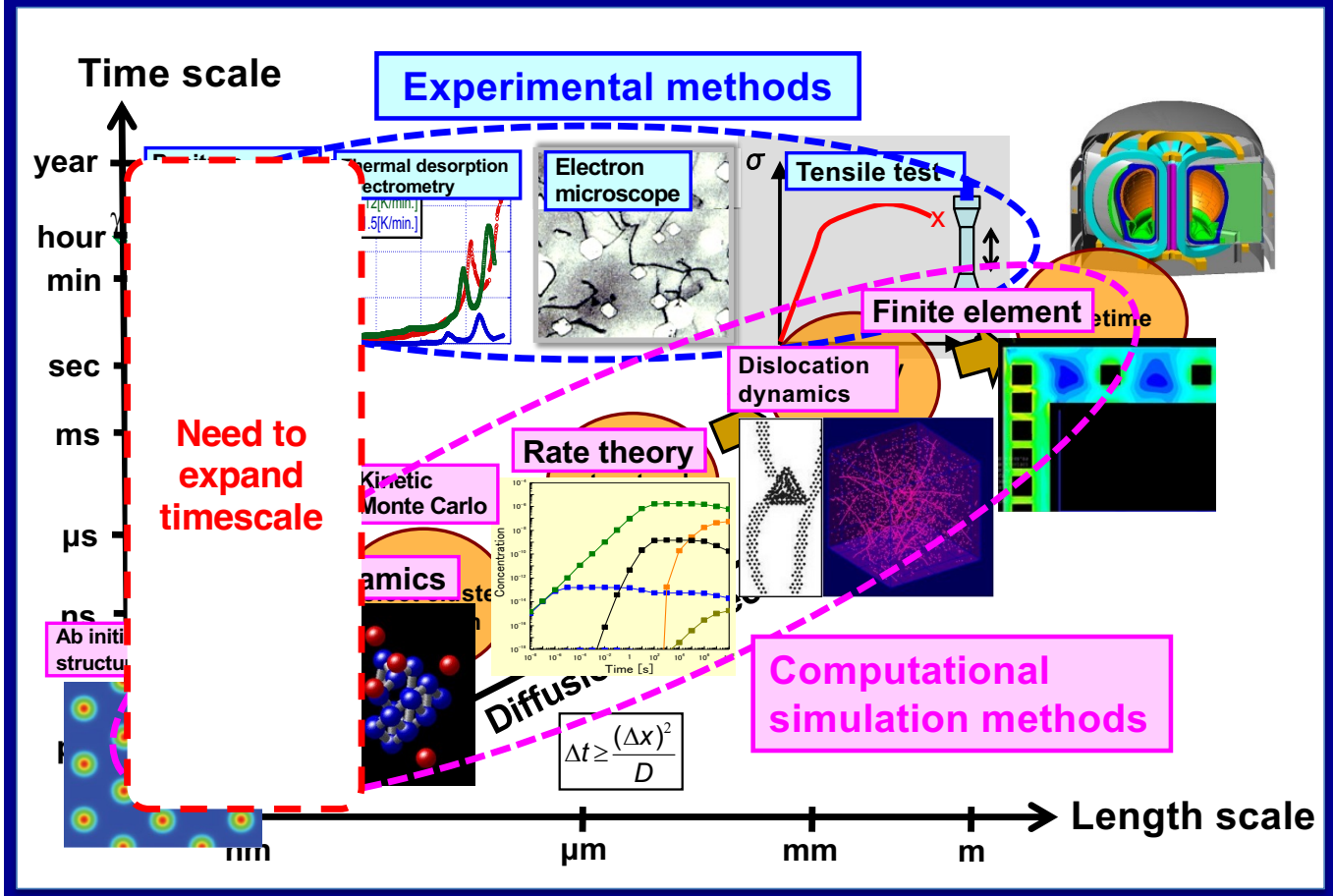
E.g.)
migration energy of a vacancy in Fe :
E_a = ~ 0.6 – 0.7 eV

It is practically difficult to construct a model of the evolution of bubble microstructure by MD simulations alone.

Waiting time as a function of temperature in the case of E_a = 0.6 eV



Molecular simulation of reactor structural materials [3,4]

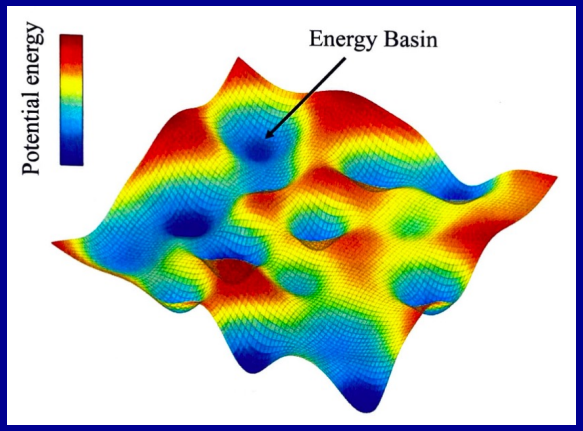


- Changes in properties of structural materials under irradiation are multiscale phenomena in terms of both time and length scale.
- It is indispensable to complementarily utilize experiments and simulations in order to elucidate the phenomena.
- It is important to develop a prediction model based on the understanding of the obtained mechanism.
- **In particular, in order to construct a model of the evolution of bubble microstructure, it is necessary to expand the timescale while maintaining the similar accuracy as MD simulations.**

[3] K. Morishita, JPFR (2008), [4] Y. Watanabe (2020)

Kinetic Monte Carlo (KMC)

– Extension of timescale based on state-to-state dynamics –



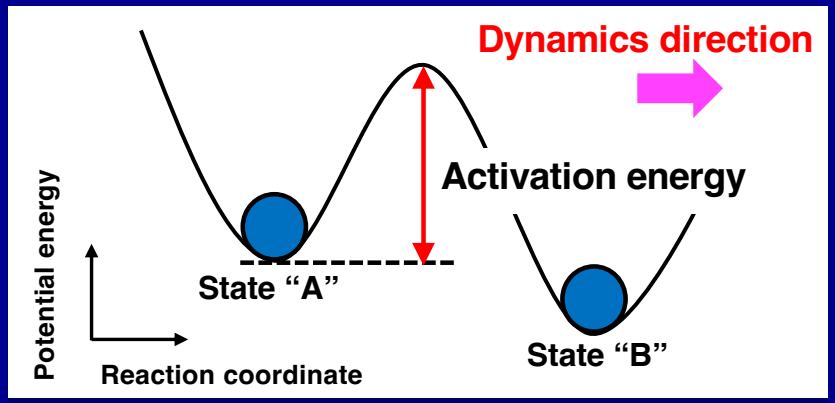
- **Events that can occur in each timestep are summarized in a list, weighted based on the activation energies, and then selected stochastically.**

Event list

- **Event 1 with** E_a^1
- **Event 2 with** E_a^2
- .
- .
- **Event** n **with** E_a^n

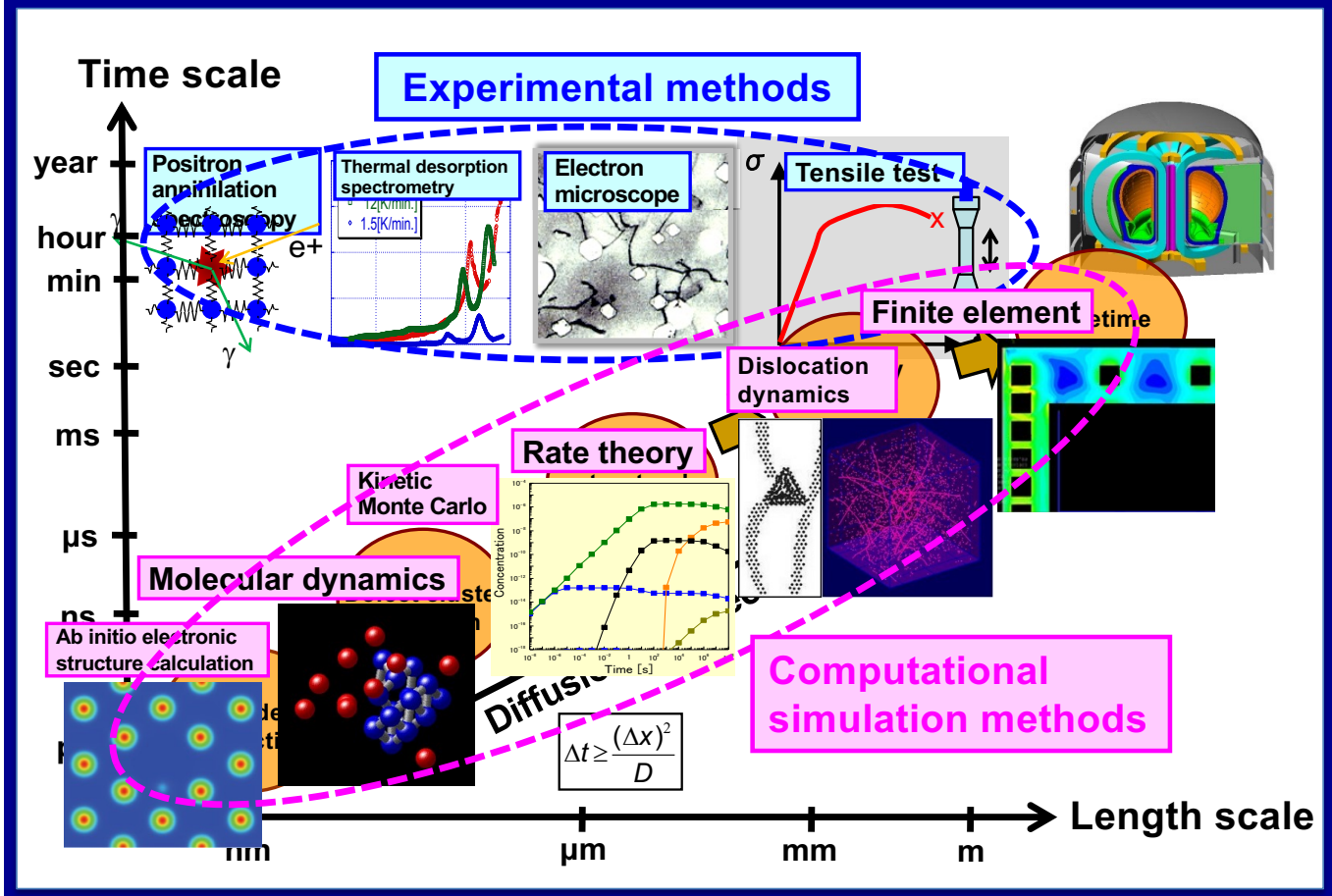
$$v_i = v_0 \exp\left(-\frac{E_a^i}{k_B T}\right)$$

E_a^i : **Activation energy of the event** i **.**



✖ **It is difficult to treat unpredictable events because it is necessary to list all possible processes in advance.**

Objective of this study

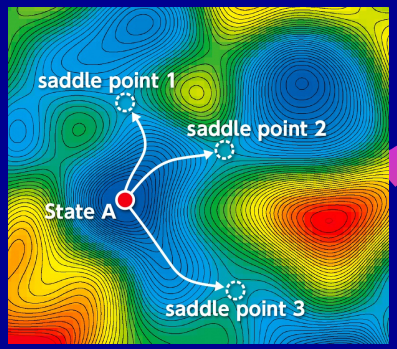


- To develop a computational method that can expand the timescale of MD simulations while maintaining atomistic fidelity of a system that incorporates He bubbles.
- To reproduce a coalescence process with dynamic behaviors of He bubbles, i.e. diffusion of bubbles themselves, using the constructed method.

[3] K. Morishita, JPFR (2008), [4] Y. Watanabe (2020)

On-the-fly KMC (1)

- By introducing an algorithm that searches for the activation processes at each timestep of calculations (on-the-fly), it is a method that can treat complicated behavior without preparing an event list in advance.



Event list

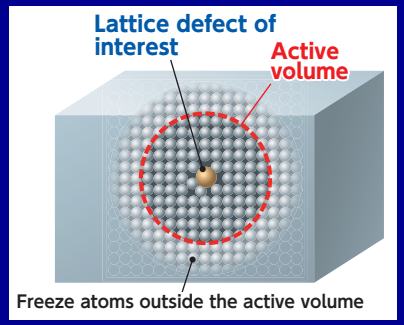
- Event 1 with E_a^1
- Event 2 with E_a^2

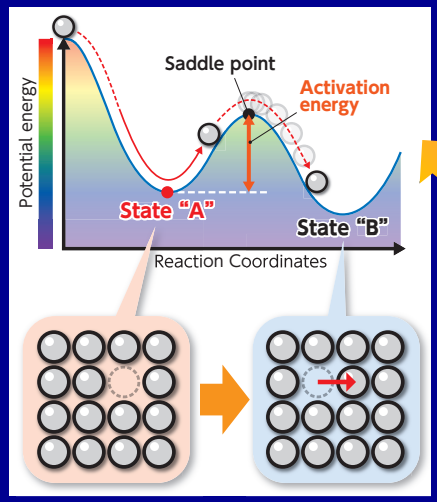
.

.

- Event n with E_a^n

- In the SEAKMC method, which is used in this study, the saddle point search, which occupies more than 90 % of the computational cost, is significantly speeded up by limiting the area (Active volume).





[5] H. Xu et al., 2011

On-the-fly KMC (2)

Challenges for kMC : flicker events

Event list

- Event 1 with E_a = 0.22 [eV]
- Event 2 with E_a = 0.24 [eV]
- Event 3 with E_a = 1.10 [eV]

$$\nu = \nu_0 \exp\left(-\frac{E_a}{kT}\right)$$

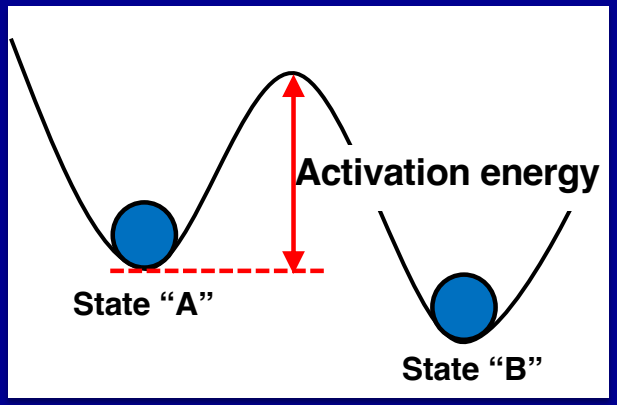
Frequency of occurrence at 300K (ν)

Event list

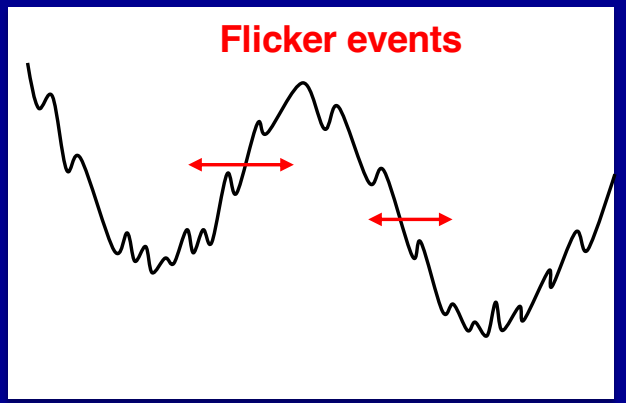
- Event 1 with ν = 2.0×10^{8} [s^{-1}]
- Event 2 with ν = 1.0×10^{8} [s^{-1}]
- Event 3 with ν = 3.3×10^{-7} [s^{-1}]

Probability that Event 3 will be selected (P_3)

$$P_3 = \frac{3.3\times10^{-7}}{2.0\times10^{8} + 1.0\times10^{8} + 3.3\times10^{-7}} \approx 1.1\times10^{-15}$$

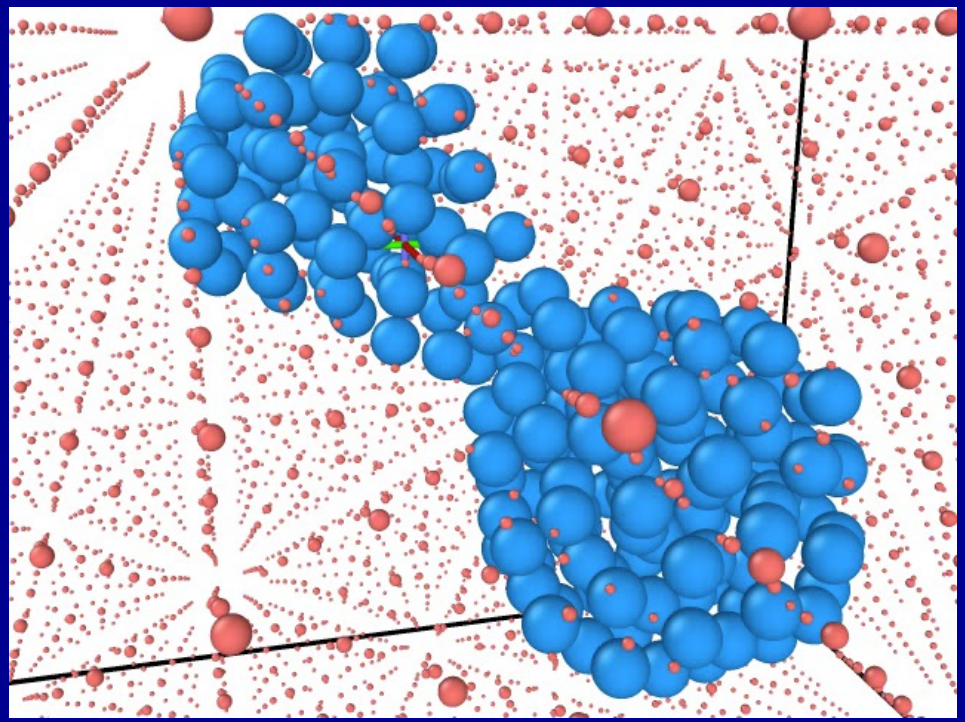


In reality



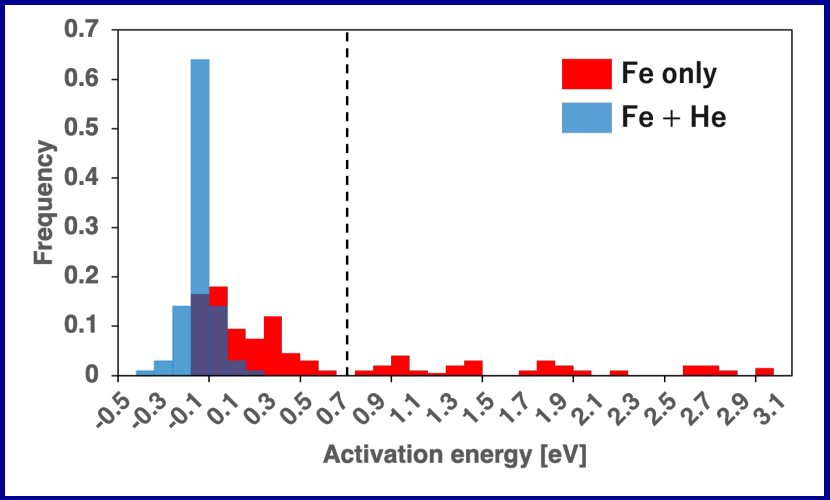
On-the-fly KMC (3)

SEAKMC calculation for contact of two He bubbles in Fe.



✖ It takes many steps to move back and forth between small dents near the energy minimum, and it takes time for the phenomenon of interest to occur. It is a common issue for kMC.

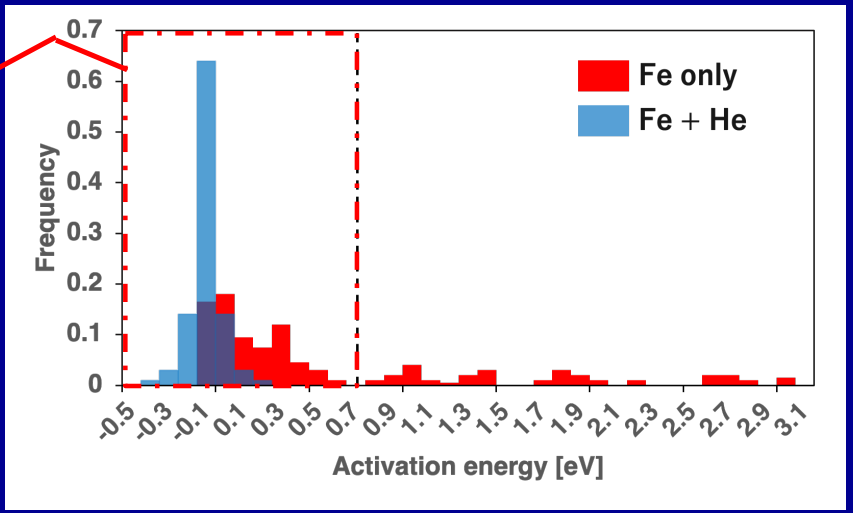
Especially in a system containing light elements such as He atoms, the activation energies of their trivial vibrations are very low, and it is difficult to avoid them.



Dynamic behavior of He bubbles using on-the-fly KMC

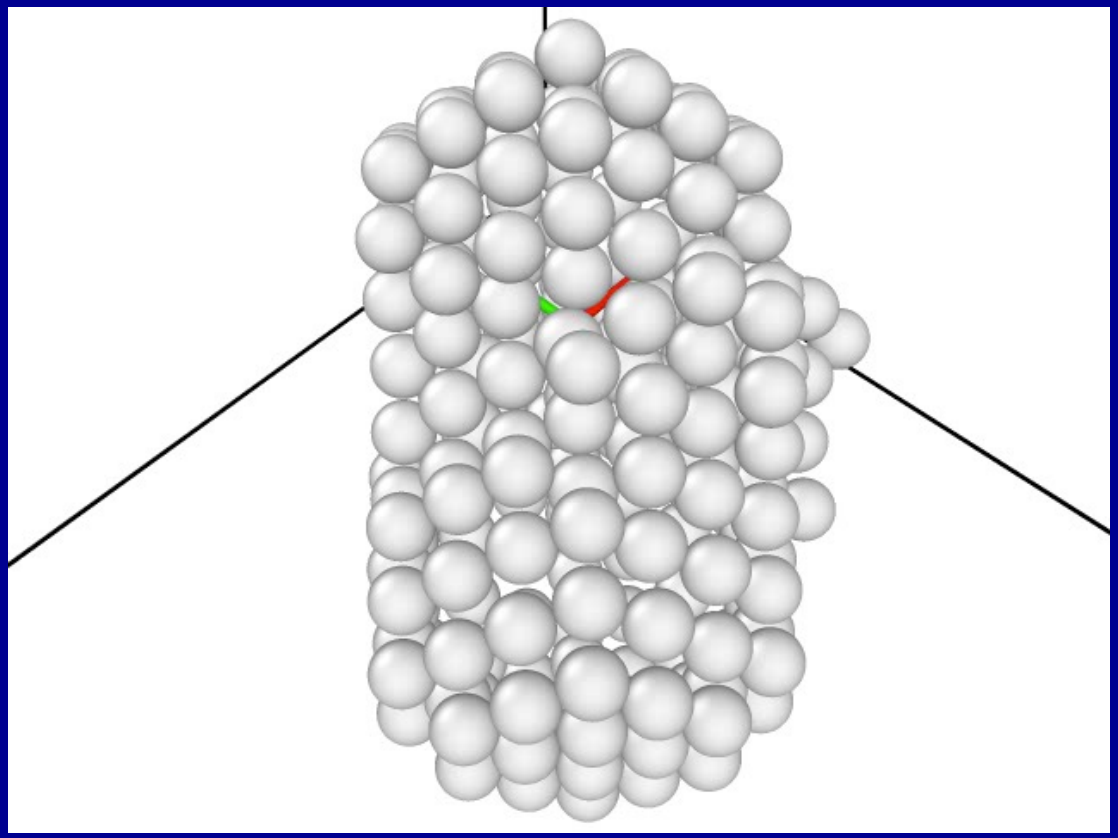
Migration energy of a vacancy in Fe : E_a = ~ 0.6 – 0.7 eV

Events with activation energies of 0.6 eV or less are set so that they are not selected as flicker events.



We have developed a calculation scheme that selects transition destination candidates for saddle point search using only the Fe system, and incorporates the influence of He atoms only during energy calculation and relaxation.

He bubble coalescence by on-the-fly kMC (1)

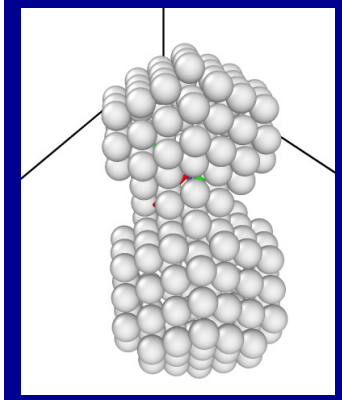


600K, He/v = 2
Diameter : 1nm
Initial distance : 1.34nm

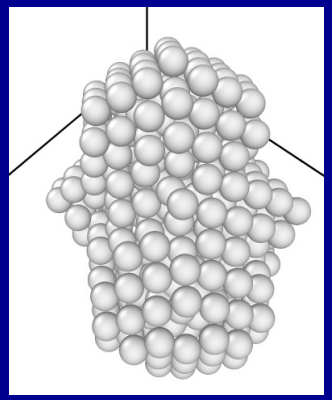
- Using an MD result as an input value, it is possible to reproduce the coalescence process in which two bubbles become an ellipsoid and consequently a sphere through multiple diffusion of vacancies, for the first time.

He bubble coalescence by on-the-fly kMC (2)

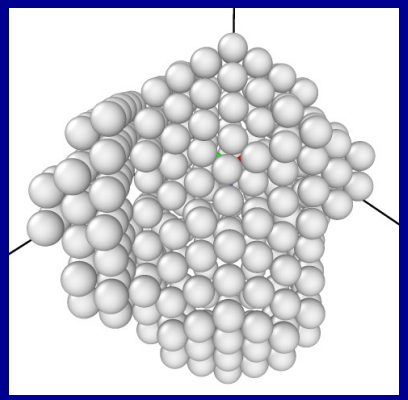
0 s



67 ps



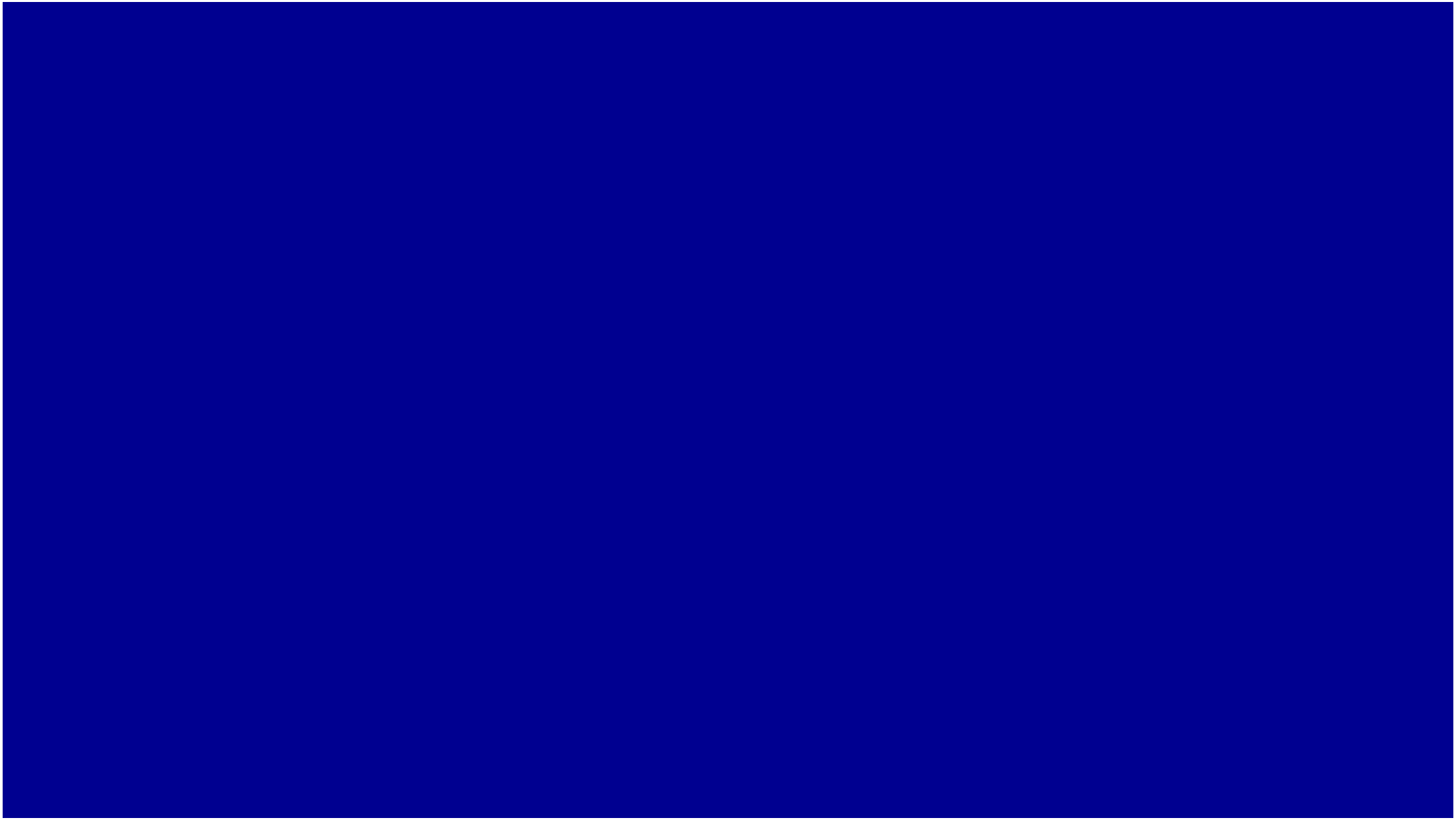
9.64 μs



- We succeeded in reproducing the time evolution up to the order of 10 μs, which is a timescale of 10^5 to 10^6 times longer than that of normal MD simulations, while maintaining atomistic fidelity.
- It was possible to reproduce the process in which multiple diffusions of vacancies occurred so as to reduce the surface area of the bubbles, the dents in the contact part were eliminated and the shape approached elliptic or spherical.
- By using the method that excludes He atoms only for the selection of transition destination candidates for saddle point search, it was also clarified that the existence of He atoms enhances vacancy diffusion by increasing the internal pressure of bubbles.
- The method developed in this study is extremely powerful in reproducing behaviors of He bubbles.

Conclusions

- We analyzed a coalescence process of He bubbles through their dynamic behavior while maintaining atomistic fidelity.
- We have developed a calculation scheme in the on-the-fly kMC that selects transition destination candidates for saddle point search using only the Fe system, and incorporates the influence of He atoms only during energy calculation and relaxation. It enabled reproducing the time progress up to a timescale that is 10^5 to 10^6 times longer than that of conventional MD simulations.
- This method is extremely powerful in reproducing behaviors of He bubbles, and probably the only method that can reproduce the phenomenon of the meso-timescale while maintaining atomistic fidelity.



Calculations

- 16 node, 36 core, 24 hours
- 10 – 20 repeated calculations for each case