Applications of a New Atomistic Monte Carlo Method: SEAKMC

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Beyond Molecular Dynamics:
Long Time Atomic-Scale Simulations

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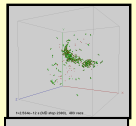
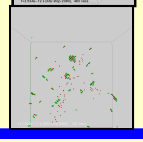
What are we interested in and why?

- simulating radiation damage in materials involves many length and time scales
- simple and complex processes with a broad range of activation energies (time scales), <0.1 eV to ~1 eV
- primary damage event, atomic displacement cascades, occur over ~10 ps
  - relevant short-term evolution up to ~μs
  - influences damage accumulation and property changes up to years
- current EFRC effort to directly measure cascade dynamics using time-resolved x-ray diffuse scattering



20 keV, part 1

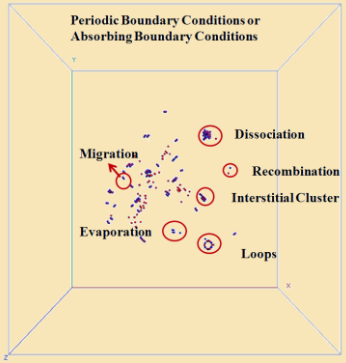


20 keV, part 2

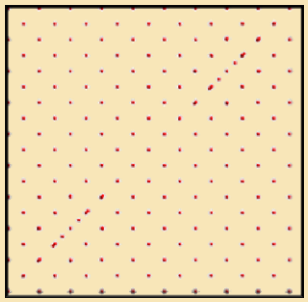
State of the art

Modeling atomistic phenomena for long times is a fundamental problem: a number of methods exist, accuracy-time scale trade-offs

Object Kinetic Monte Carlo (OKMC)

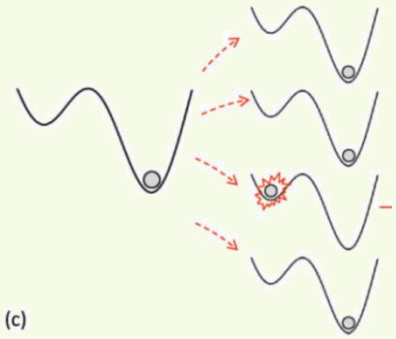


Atomistic Kinetic Monte Carlo (AKMC)

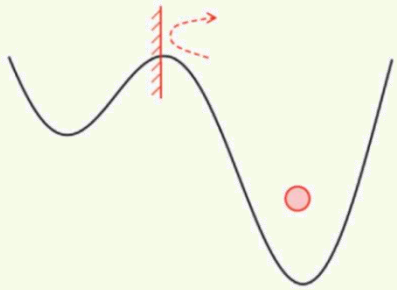


activation-relaxation technique

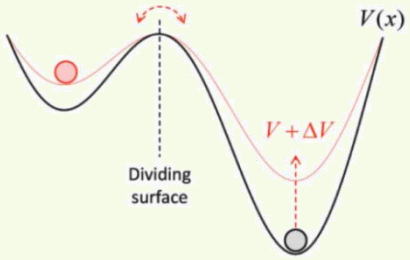
Parallel Replica Dynamics (RPD)



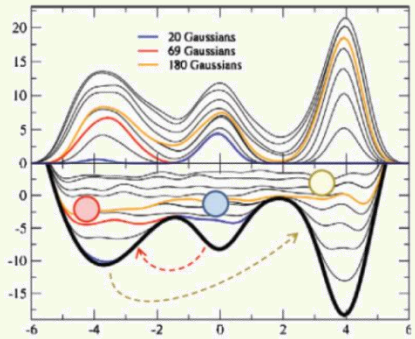
Temperature Accelerated Dynamics (TAD)



Hyperdynamics

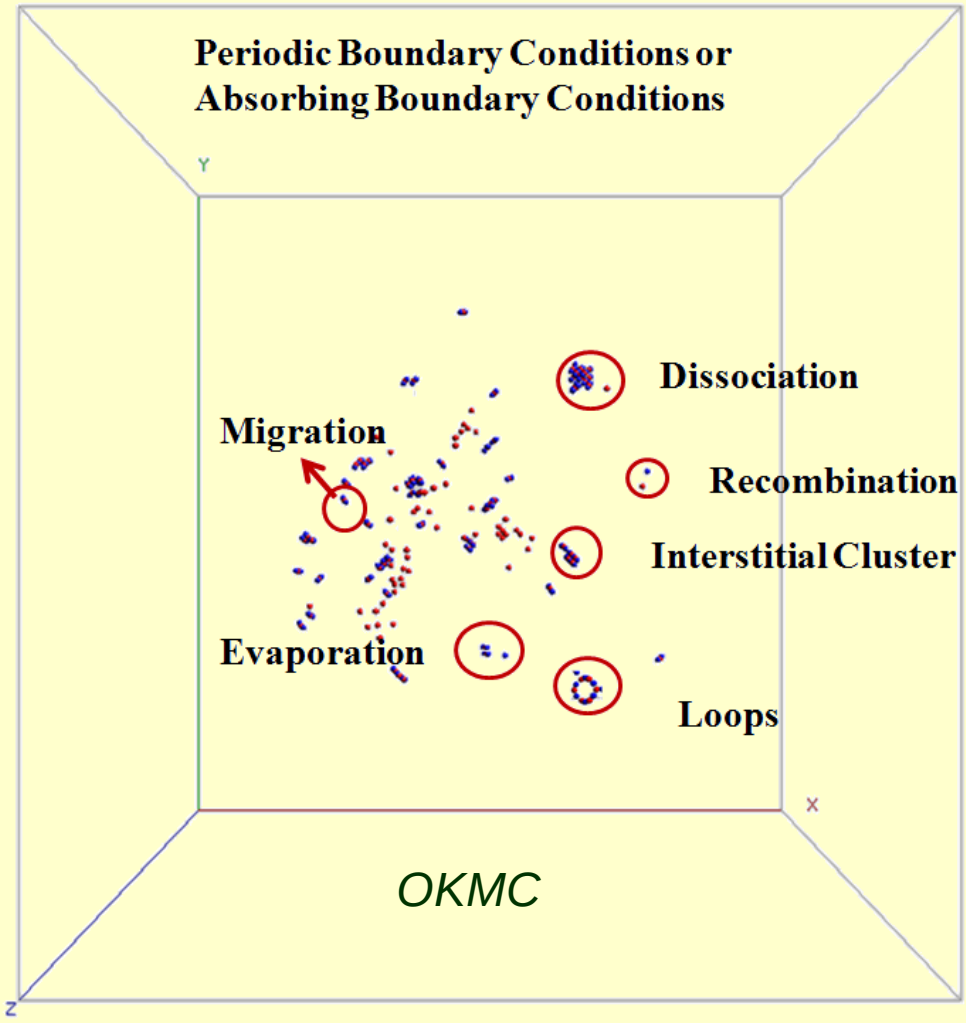


Metadynamics



autonomous basin climbing

Background - OKMC for Long-Term Defect Evolution



H.X. Xu, Y.N. Osetsky, R.E. Stoller, Journal of Nuclear Materials, Accepted

Simulation Setup

- Input from MD cascades
- Simulation box
- Temperature

Diffusion Related

- Diffusion mechanism
  - 3D, 1D, 1D+R
- Migration energy data
  - *ab initio* vs. empirical potential
- Dissociation of interstitials
- Evaporation of vacancies
- Rotation of interstitial clusters
- Reaction Radius

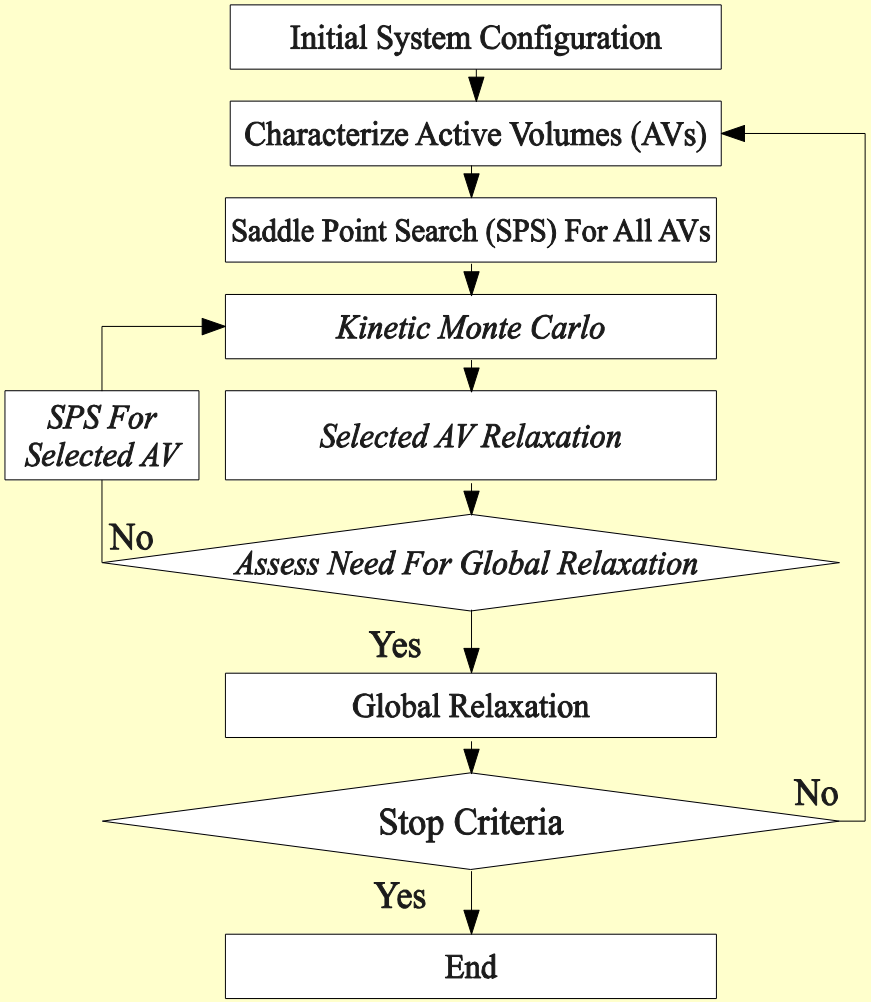
Background - Deficiencies of OKMC

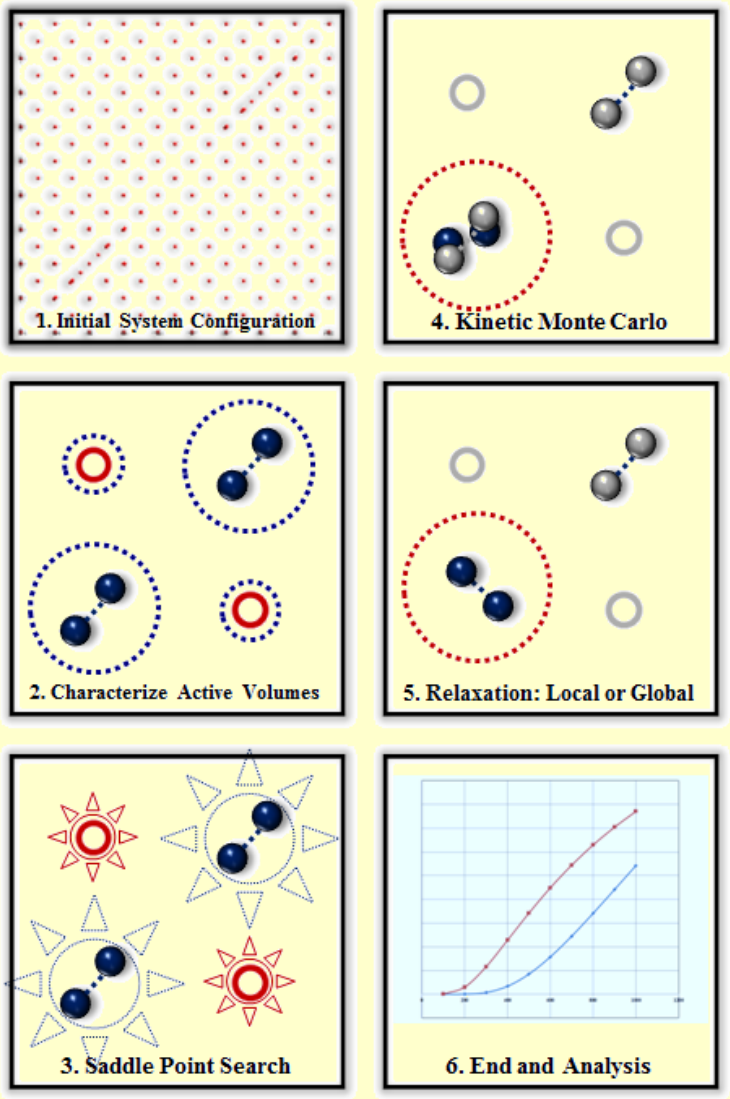
- no atomistic details
- properties of each type of defect is fixed during the simulation
- impossible to determine migration energies for all the possible events
- the simulation results are significantly affected by the defect diffusion mechanisms, which are difficult to predetermine
- artificial defect interactions and interaction radius

- Atomistic details are necessary in order to accurately describe long-term defect evolution
- Most atomistic KMC employs on-lattice approximation, not suitable for interstitial clusters

A general framework including multiple technique is proposed to study long-term defect evolution: both defect diffusion and interaction

Self-Evolving Atomistic Kinetic Monte Carlo (SEAKMC)

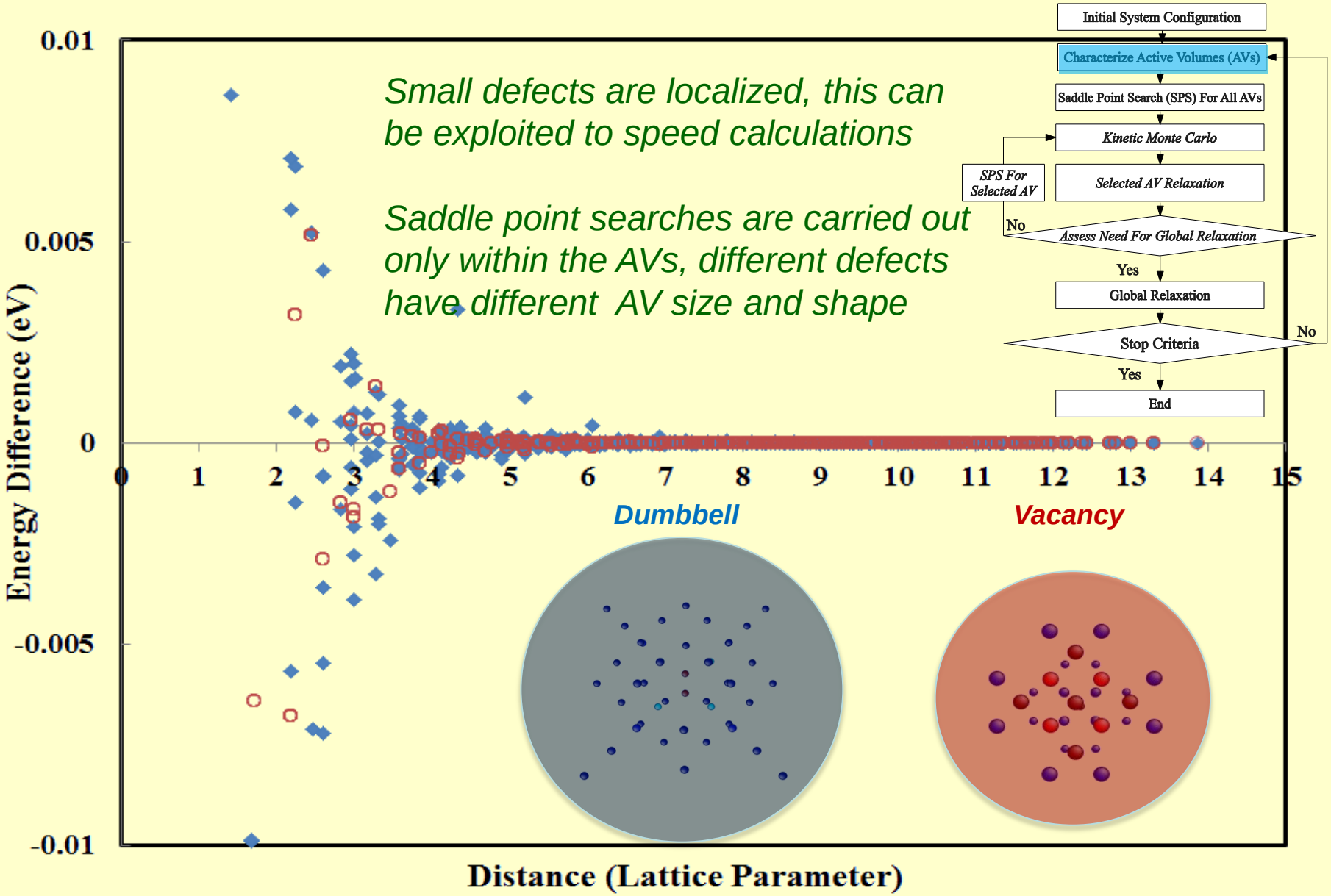




H.X. Xu, Y.N. Osetsky, R.E. Stoller, Physical Review B, Brief Reports, 84, 132103 (2011)

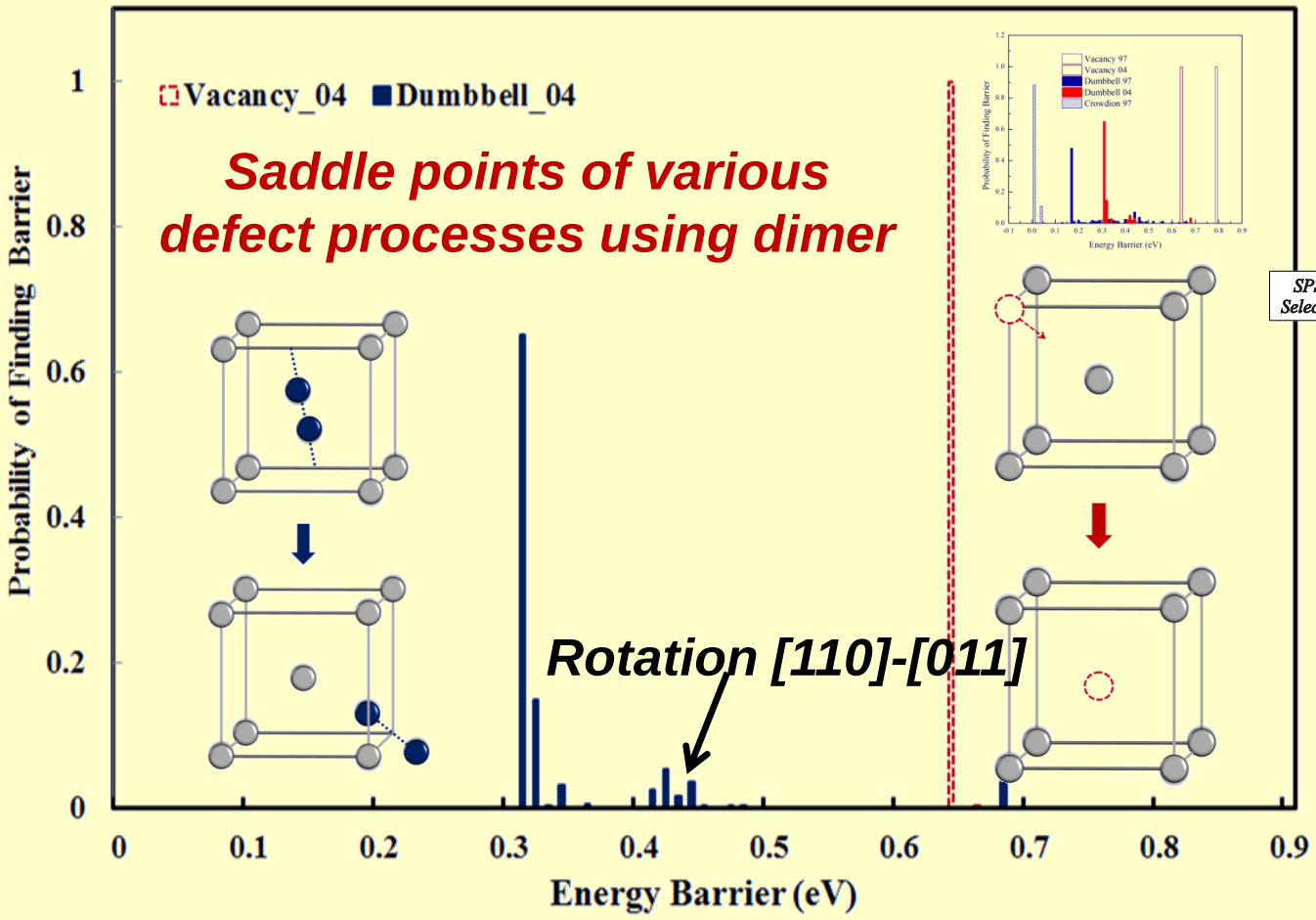
SEAKMC is particularly powerful for large systems with complex defects; accurately includes defect diffusion, defect interactions naturally occur

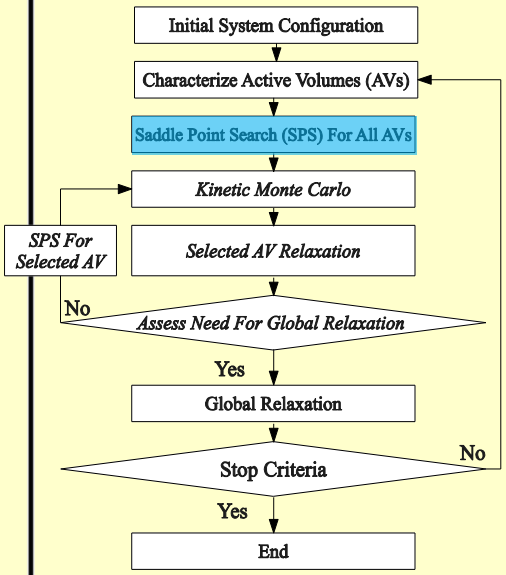
Active Volumes (AVs) – Spatial Localization



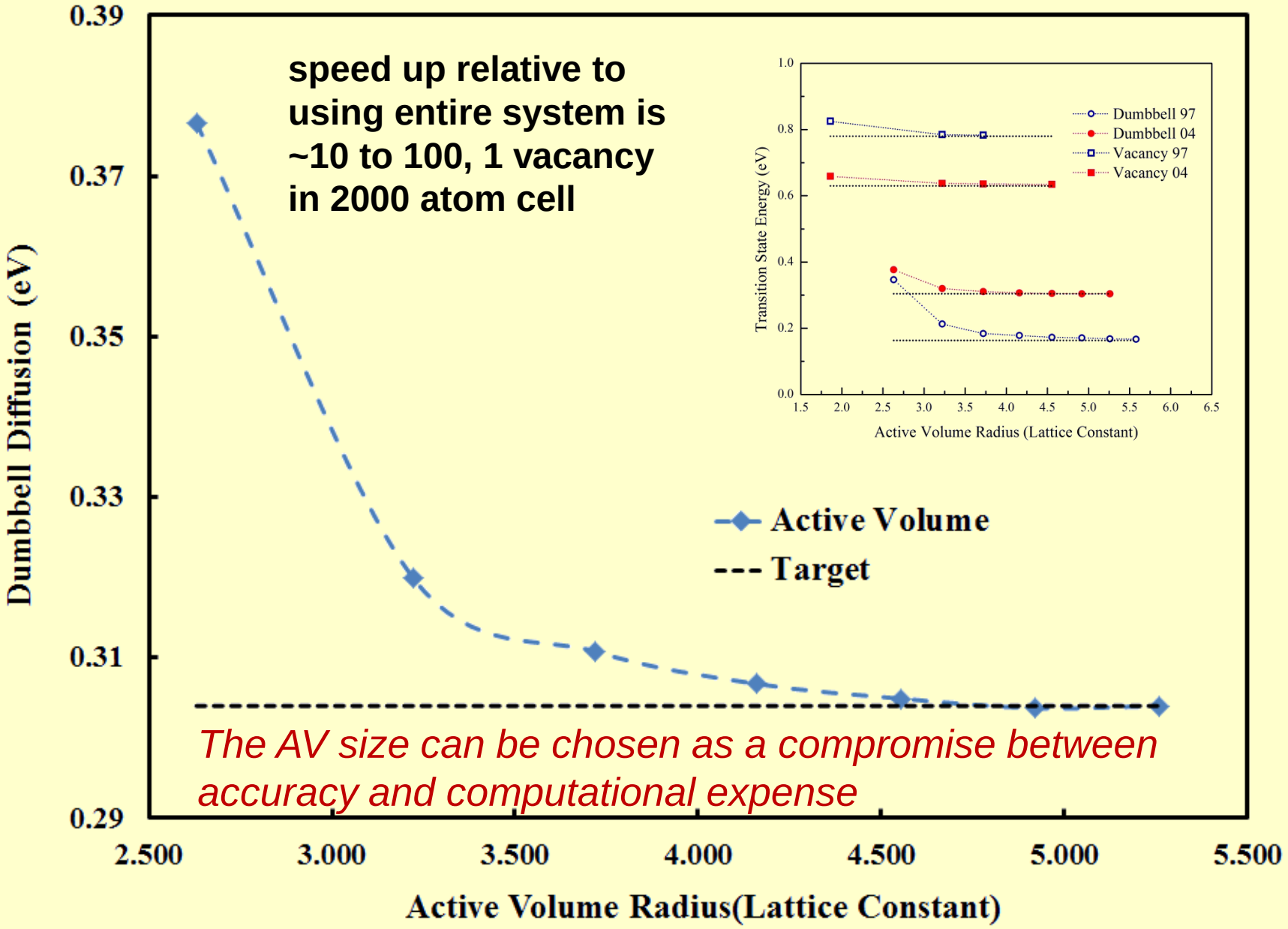
Saddle Point Search Techniques

- *A variety of methods exist: dimer, Lanczos, NEB, ...*
- *SEAKMC developed using dimer, harmonic transition state theory*
- *Find migration barriers on-the-fly*
- *Only need initial configuration; return the saddle point configuration*

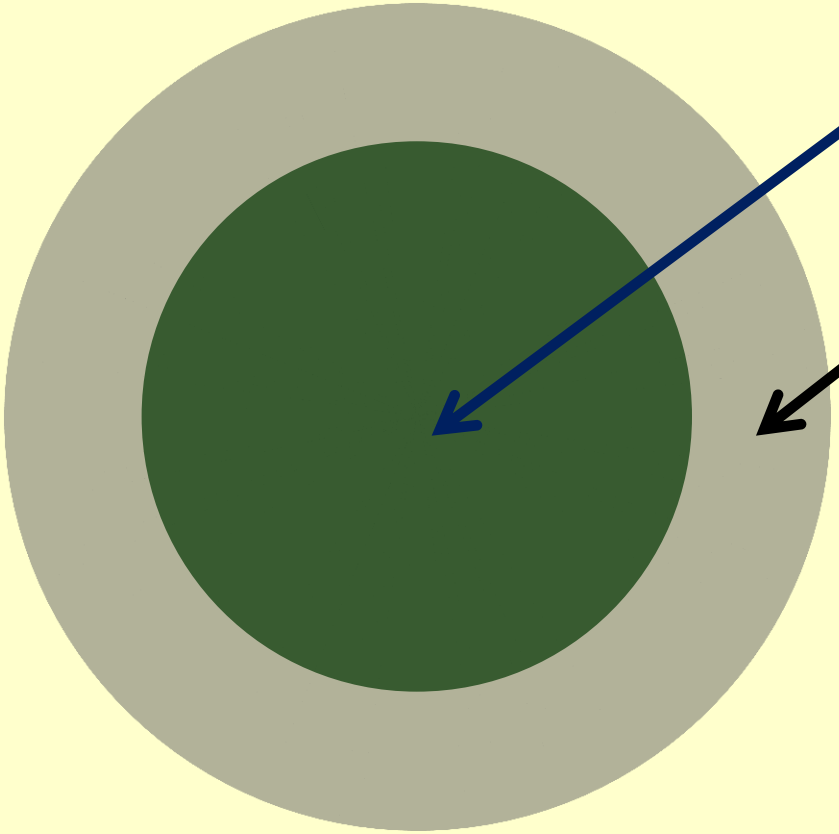




Activation Energy vs. Active Volume Radius



Multi-Step Procedure



Find an approximate saddle point in smaller AV

- Fewer force calculations
- Fewer degree of freedom

Move to larger AV

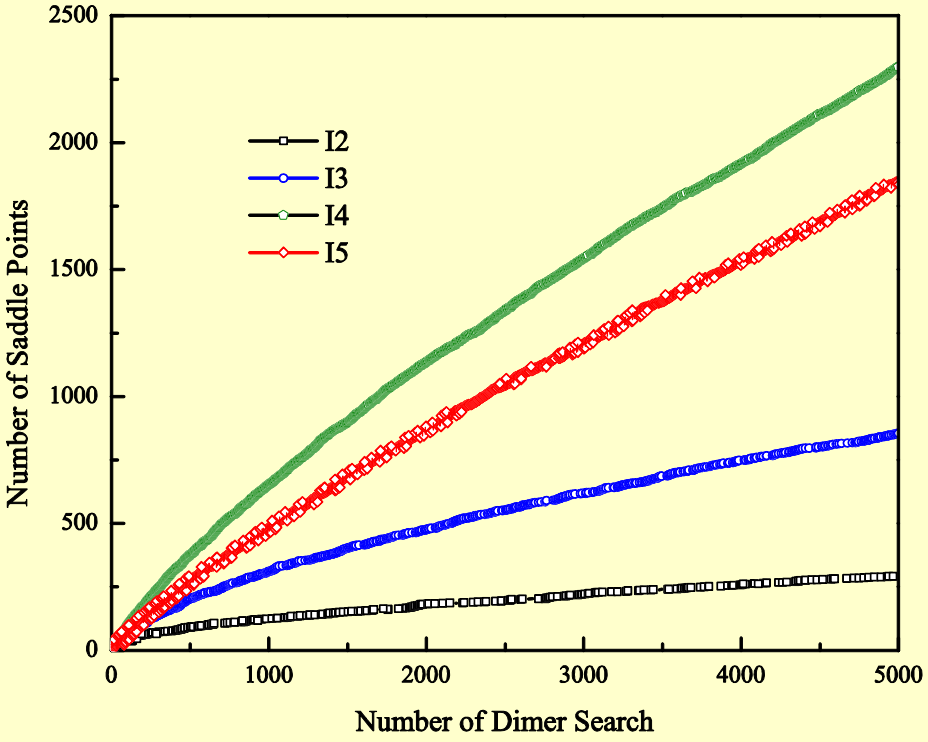
- Find the accurate saddle point
- Converge to saddle point quickly
- Corrects any error from previous step

Shape of the active volume depends on the nature of the defect

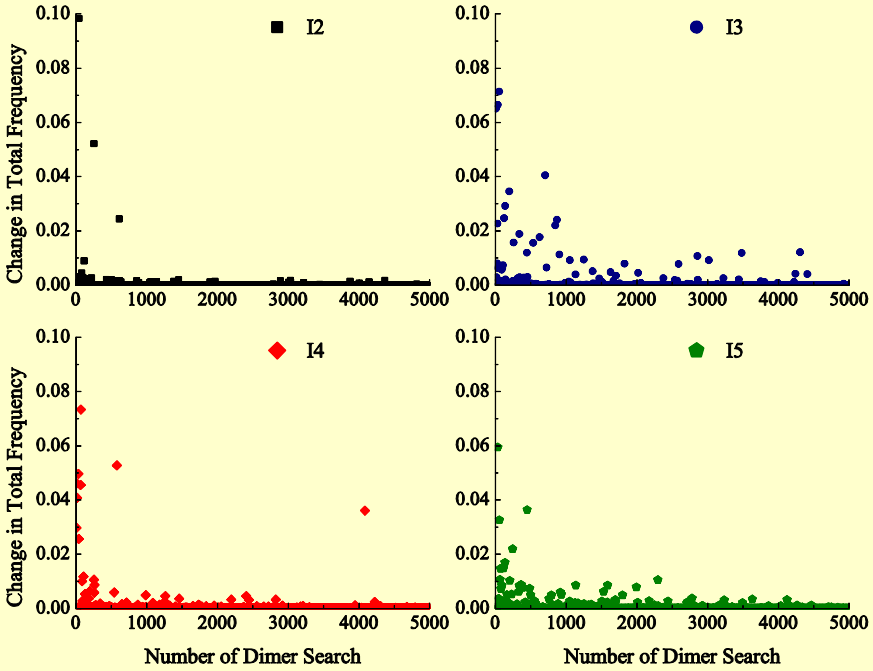
- Point defect: sphere
- Dislocation: cylinder

Relative to using larger AV initially, speed up is ~2 for vacancy diffusion and somewhat greater for interstitial

Issue of many saddlepoints and accuracy



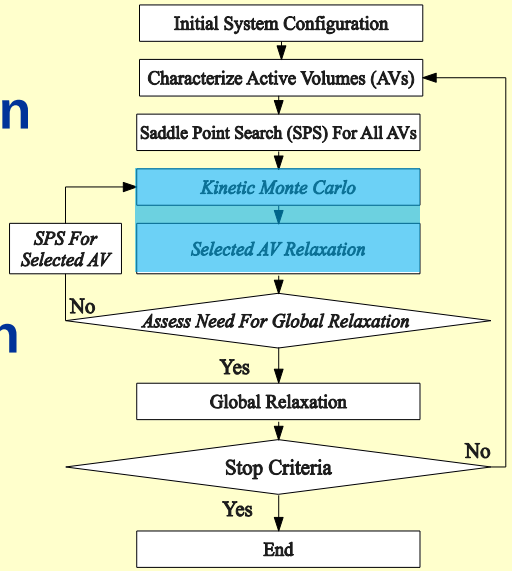
Number of distinct SPs as a function of dimer search for SIA clusters I2-I5. For each case, an initial defect configuration is randomly chosen and 5000 dimer searches are carried out.



Error estimate in the total reaction frequency for SIA clusters I2-I5, interval of five dimer searches is used to compute the rate of change in frequency.

Kinetic Monte Carlo (KMC) and Relaxation

- Randomly choose an event in one AV based on relative probabilities
- Advance time, residence time algorithm
- The events table is updated during the simulation
- Static relaxation moves system over the saddle point to another local minimum
- Conjugate gradient method is used for relaxation
- AVs can merge during relaxation if appropriate, local and/or global relaxation



- New saddle point search only in affected AV, others are "recycled"

Application (benchmarking) of SEAKMC for a few interesting cases

- *Point defect diffusion*
- *Behavior of specific interstitial defects*
- *Cascade annealing in bcc iron*

Defect Diffusion

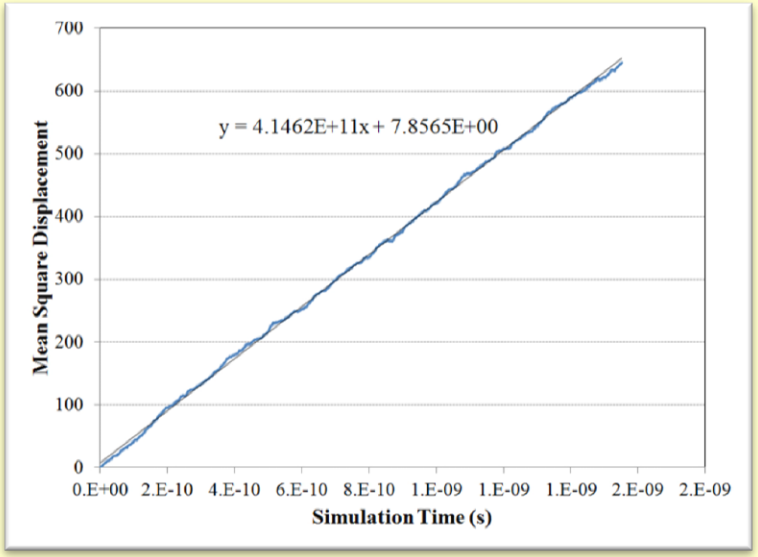
- **Prefactor and Migration Energy**
  - $k^{HTST} = \frac{\prod^{3N} v^{init}}{\prod^{3N-1} v^{sp}} e^{-E_m/k_B T}$

  Vineyard's expression for transition attempt frequency

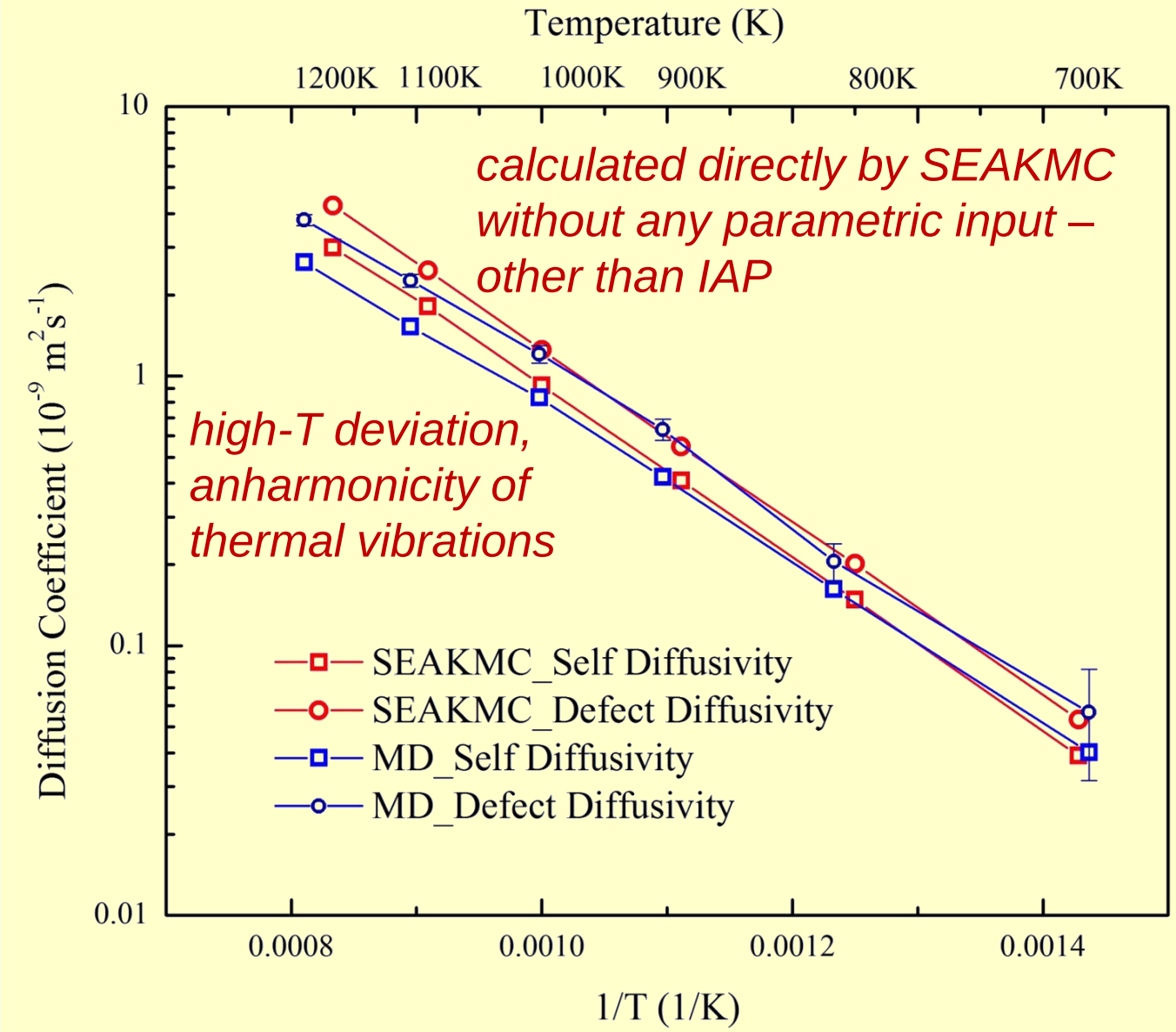
- **Correlation factor: defect and tracer**
  - $f^d = \frac{1+\langle cos\theta \rangle}{1-\langle cos\theta \rangle}$ $f^{tr} = \frac{D_{self}}{D_{defect}}$ *θ is the angle between jumps*

- **Mean square displacement (MSD)**
  - $D_{self} = \langle R_{atoms}^2 \rangle / 2nt$
  - $D_{defect} = \langle R_{defect}^2 \rangle / 2nt$

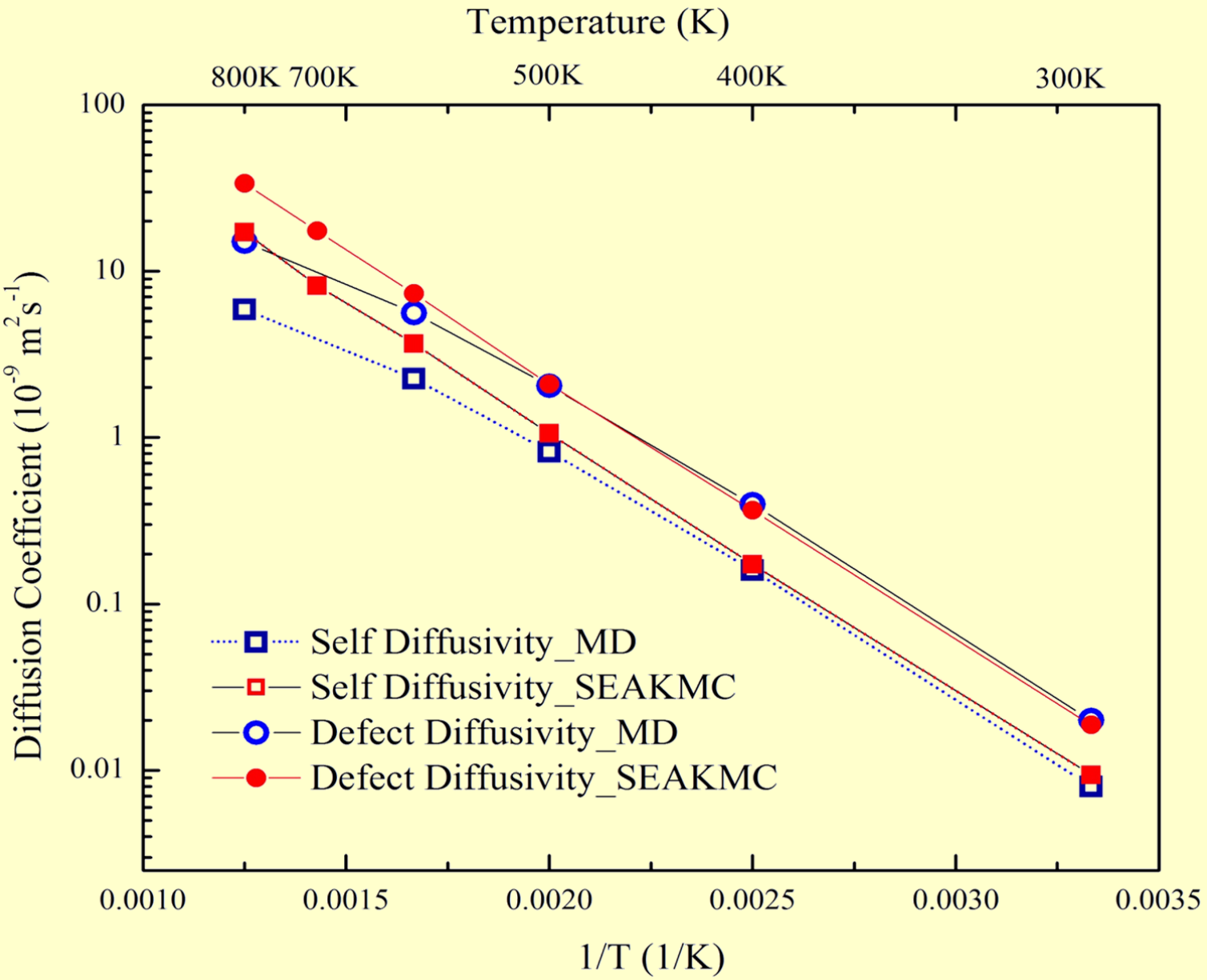
- **Analytic formula**
  - $D_{defect} = f^d \frac{k\Delta^2}{2n}$



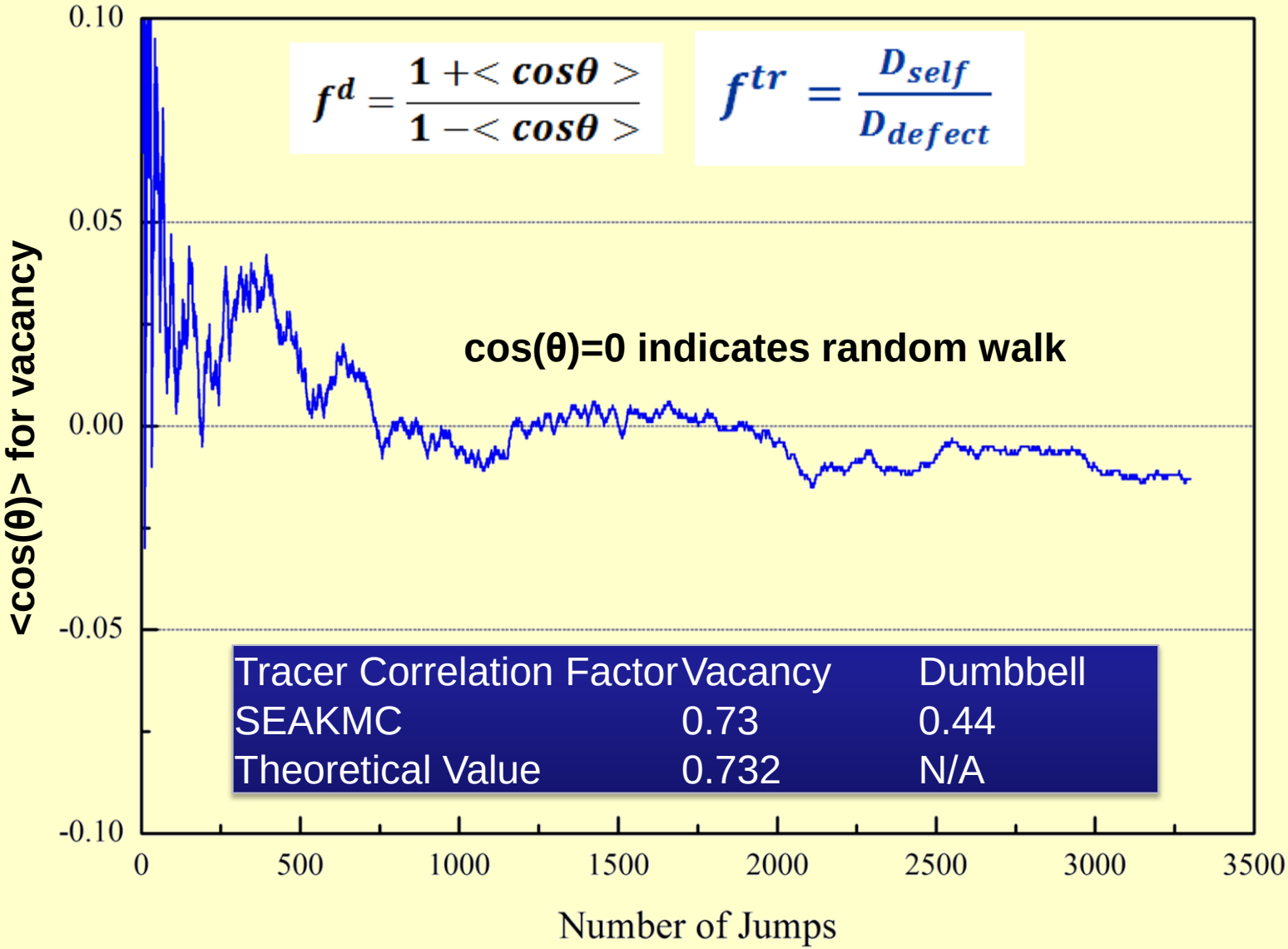
Diffusion coefficient for vacancies



Diffusion coefficient for dumbbell interstitial



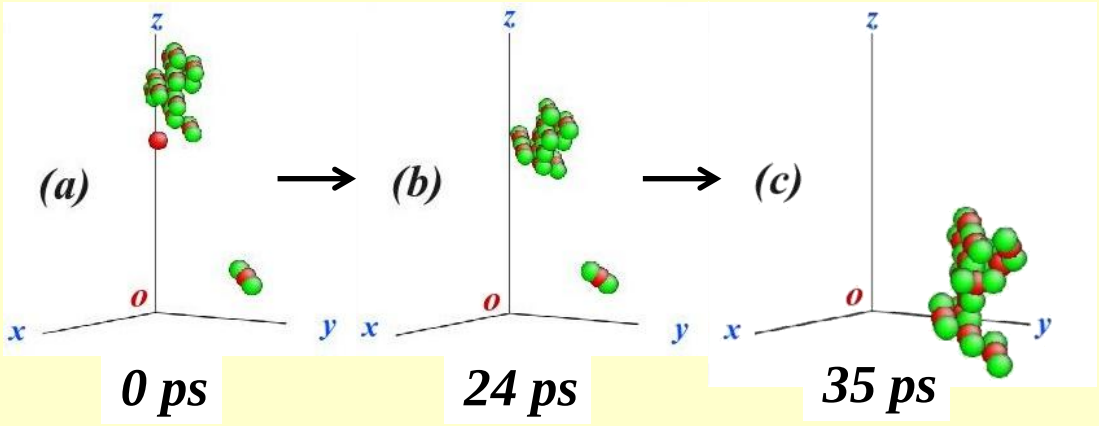
Correlation Factors



Tracer Correlation Factor	Vacancy	Dumbbell
SEAKMC	0.73	0.44
Theoretical Value	0.732	N/A

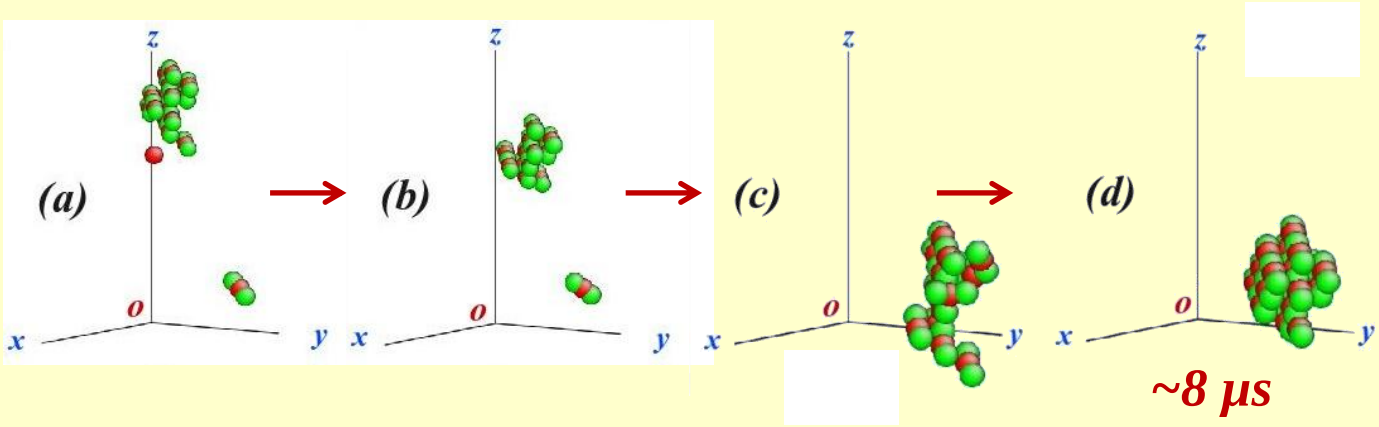
Behavior of Interstitial Defects: Comparison with MD

MD



No further changes observed on MD time scale

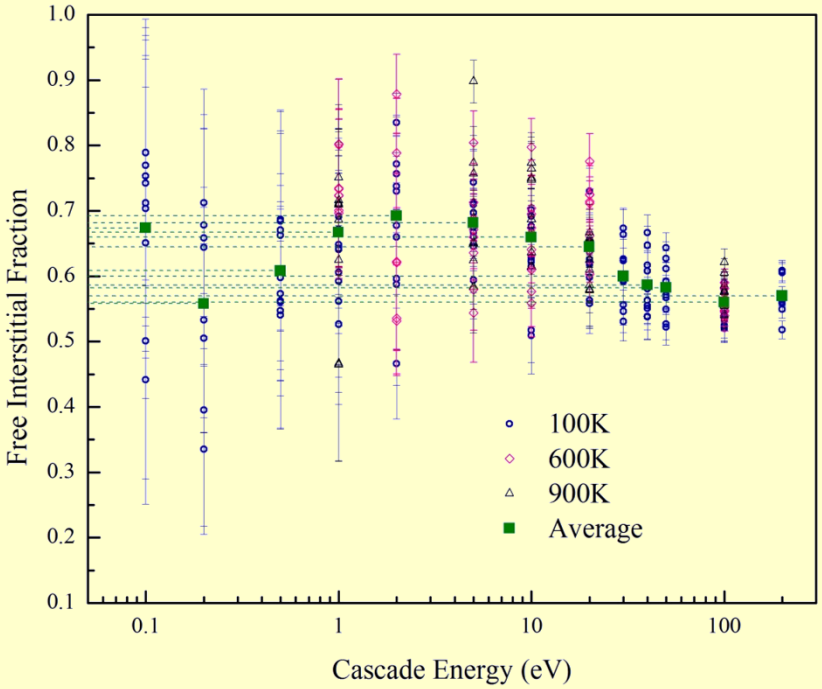
SEAKMC

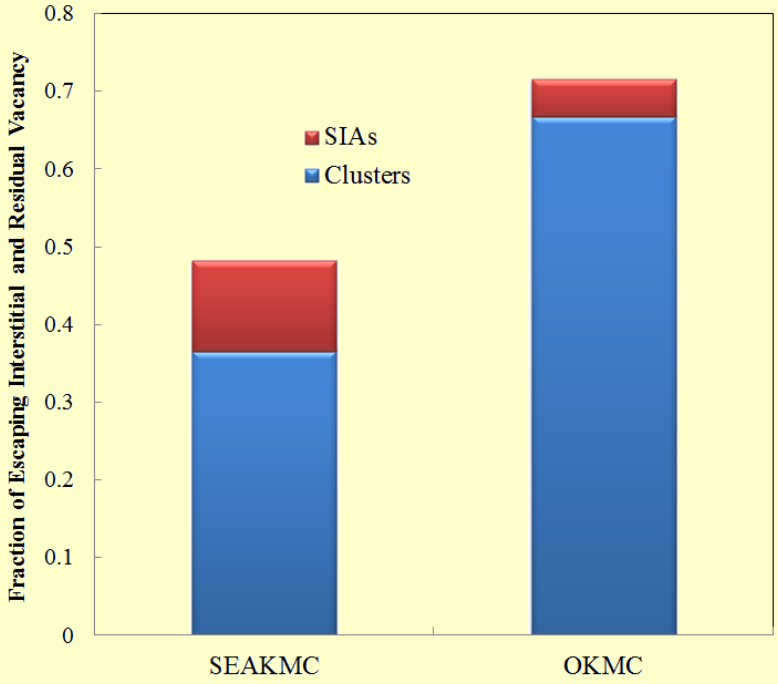


- *New phenomena observed at time scale well beyond MD using SEAKMC*
- *SEAKMC accurately describes defect diffusion; interactions occur naturally*
- *Sessile interstitial clusters created from the interactions of glissile defects, long-time conversion back to glissile observed in SEAKMC*

Cascade Annealing-Comparison of OKMC and SEAKMC

- Initial structure from MD cascade simulations
- Cascade energy is 10 keV
- Potential: Ackland-04
- System size: 128,000 atoms, with absorbing boundary condition
- MD simulation and SEAKMC annealing temperature : 650 K





Parameter-free SEAKMC leads to different estimates of defect survival and yields atomic structure that can be used for direct comparison with x-ray experiments

Potential Applications and Summary

Potential Applications

- Cascade annealing, defect interaction with solutes, dislocations, grain boundaries, and interfaces
- Simulations of formation, motion, and interactions of dislocations on a much longer time scale, i.e. deformation

Summary

The SEAKMC framework for long-term defect evolution was developed, much longer time scale than MD, more accurate than OKMC

- Includes multiple components: active volumes, saddle point searching, kinetic Monte Carlo, and static relaxation
- Can accurately simulate complex defect diffusion and reactions; the defect interactions naturally occur: e.g. the meta-stable sessile interstitial clusters in bcc iron can also be created by the interaction between mobile interstitial defects