Defect Evolution in Structural Materials from SEAKMC Simulations: Opportunities and Challenges

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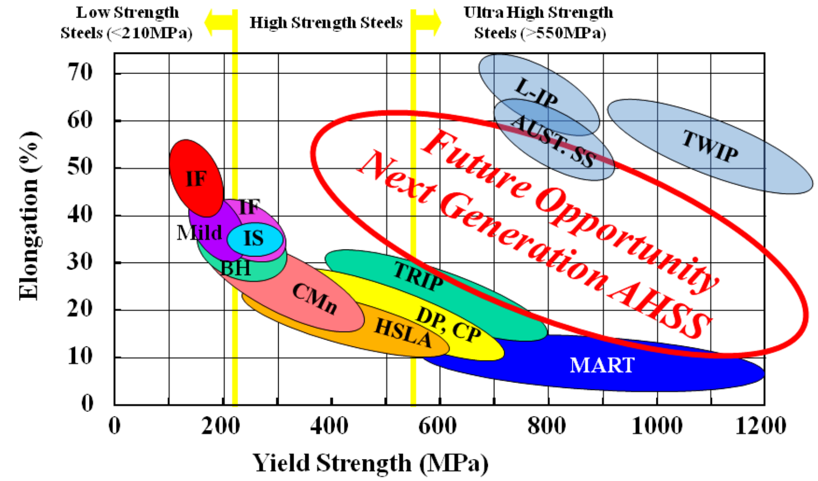
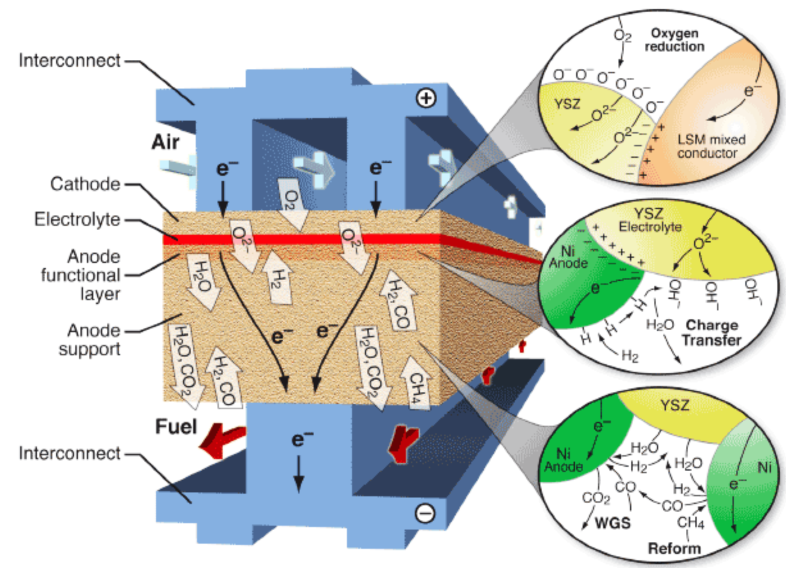
Why do we study defect evolution?

- **Collective effects of defects determine real materials properties**
  - Strength, toughness, conductivity, etc.
  - Underpinnings of our energy technologies
- **Performance limits are rarely the result of insurmountable physical principles**

  Structural alloys exhibit strengths that are typically only 5-10% of theoretical limits
- **Increased understanding of defects will result in new materials with substantially improved properties**

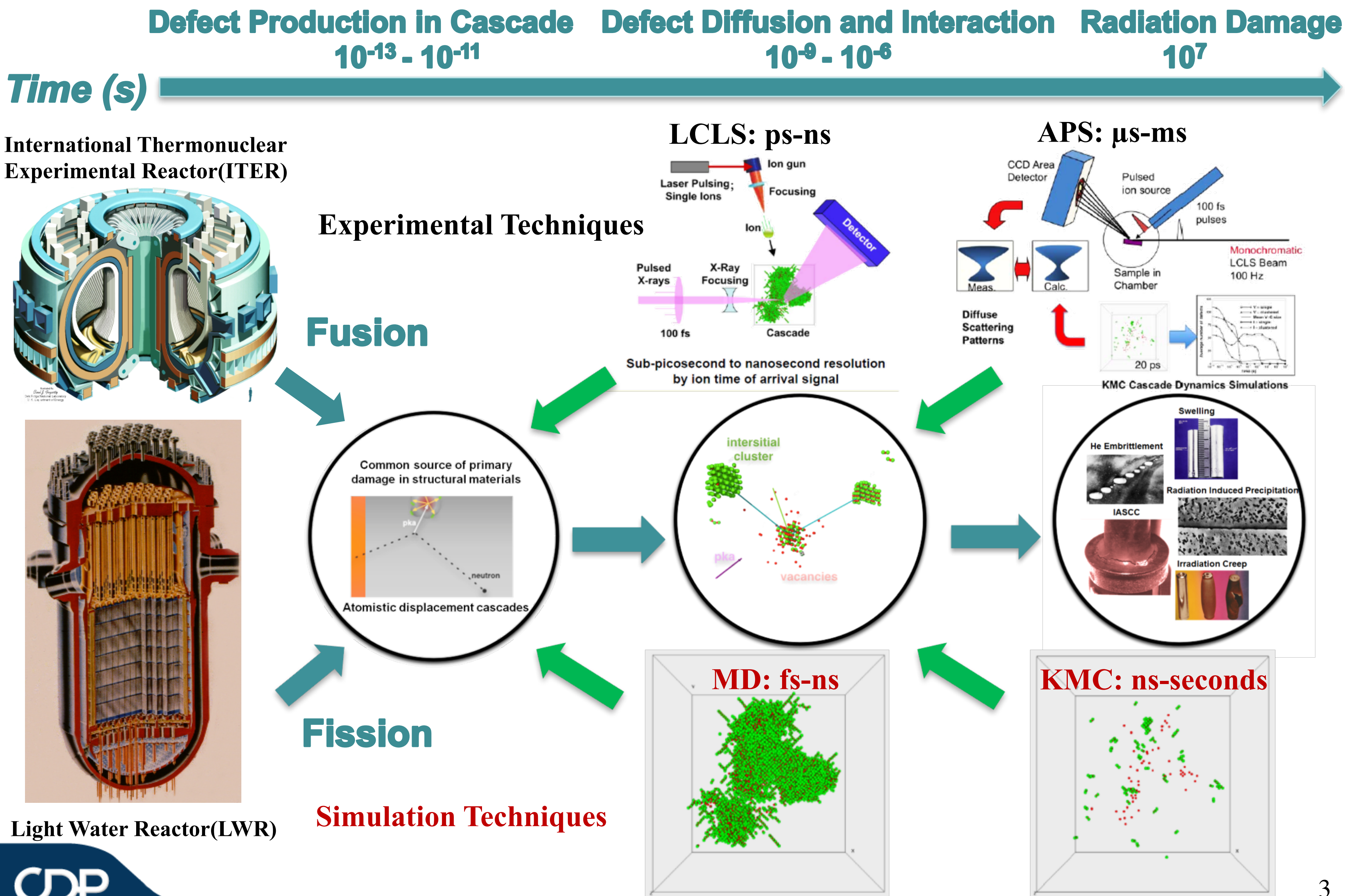




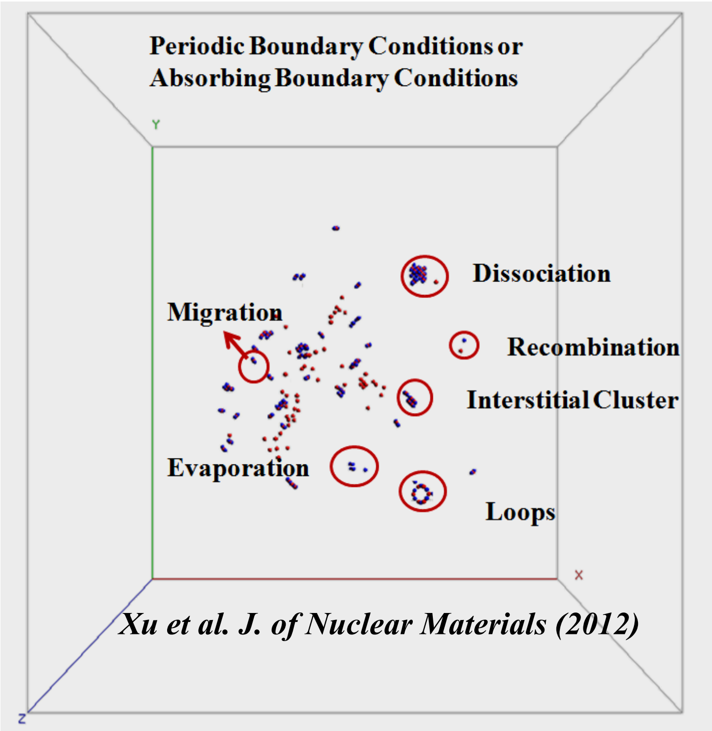
http://www.coloradofuelcellcenter.org/pages/projects/fundamental.html

Defect governs the microstructure evolution and materials properties

Evolution of Cascade Defects



Conventional Object KMC for Long-Term Defect Evolution



Processes Related

- Diffusion mechanism
  - 3D, 1D, or 1D+Rotation
- Migration energy and prefactors
  - *ab initio* or empirical potential
- Dissociation of clusters
- Reaction distance

Deficiencies

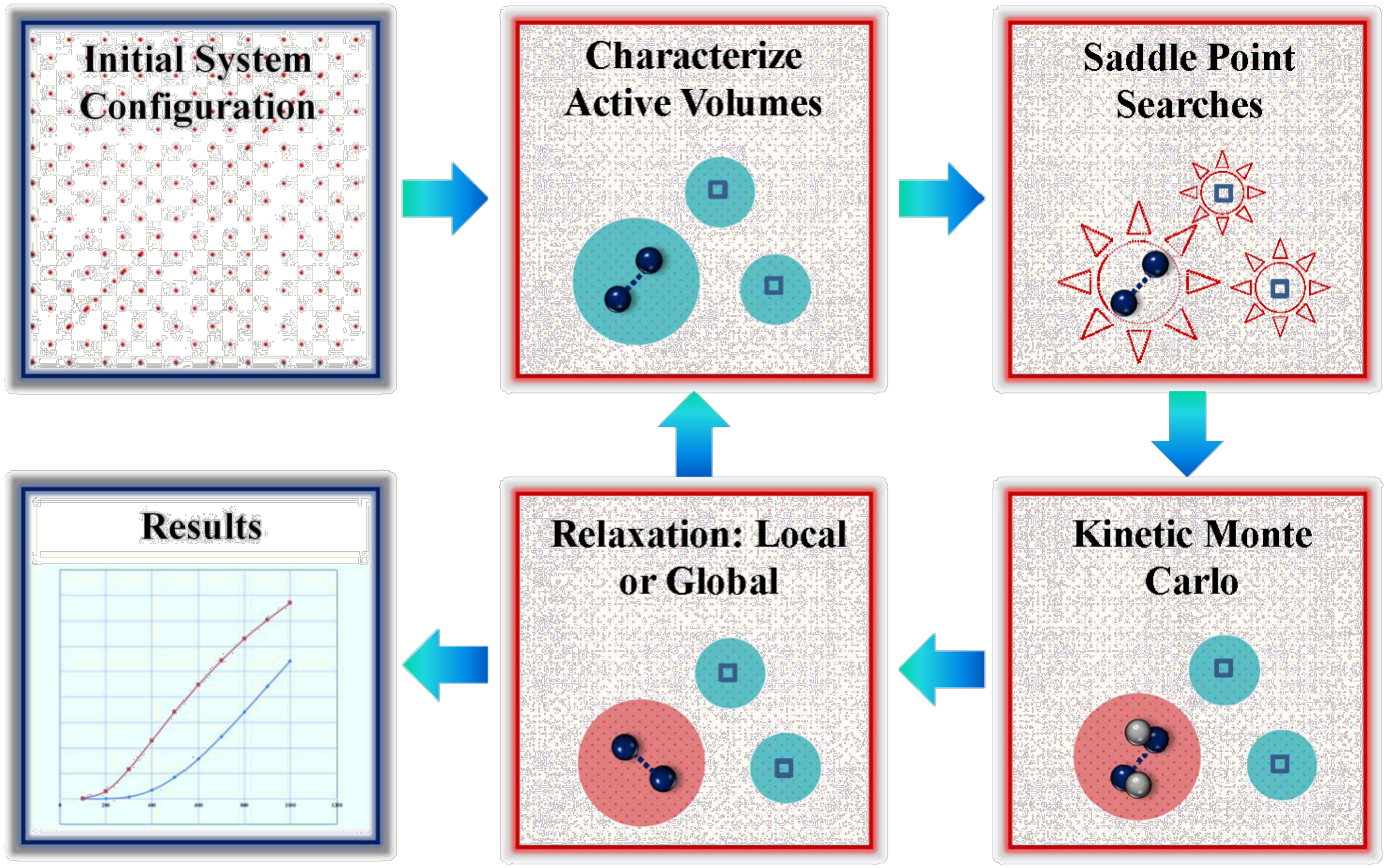
- Difficult to predetermine the diffusion mechanisms and corresponding energy
- Too much simplification: no atomistic configurations of defects
- Does not account for migration mode and energy dependence on configuration
- Simplistic description of defect interactions

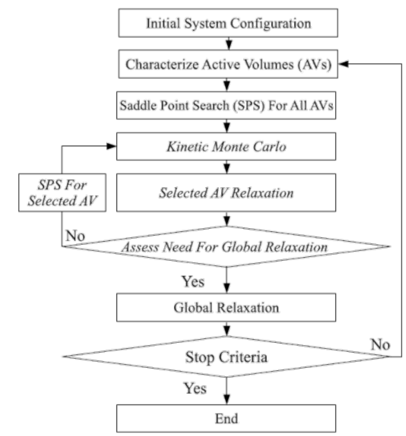
Object KMC is a oversimplified model

Atomistic details are crucial to accurately describe defect evolution

CDP

Self-Evolving Atomistic KMC (SEAKMC) Method

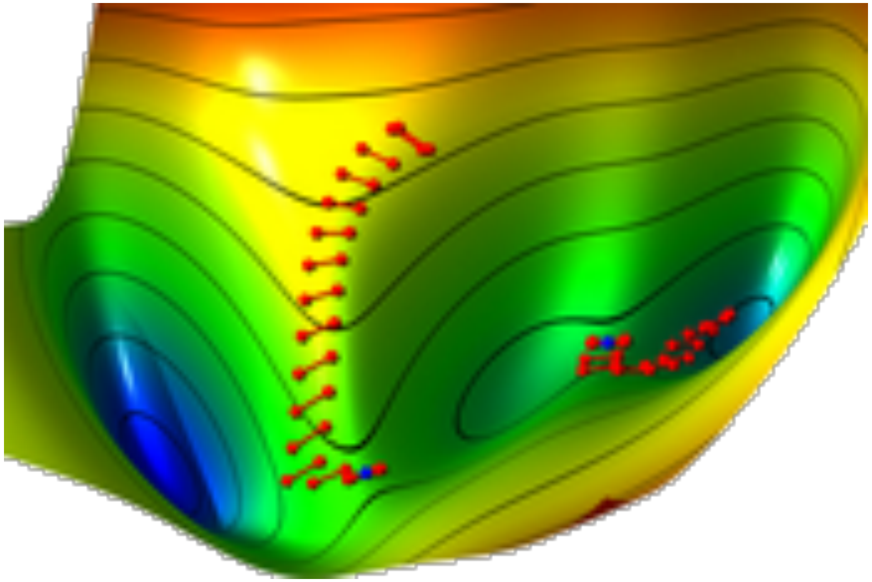




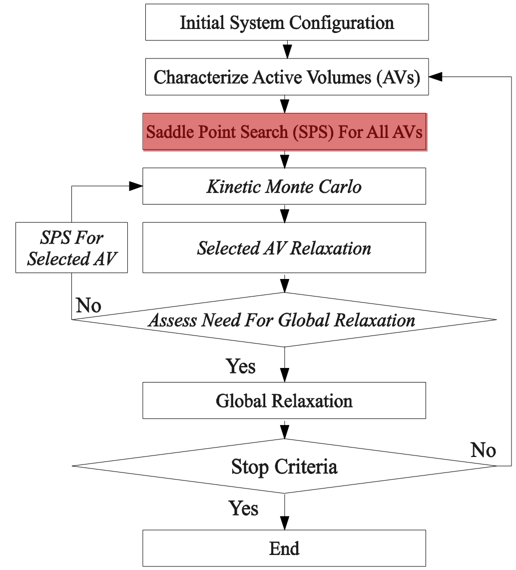
Xu et al. Phys. Rev. B 84, (2011) J. Phys.: Condens. Matter 24 (2012), Computational Materials Science 100, 135(2015) Computational Materials Science 100, 124 (2015)

SEAKMC is a general framework including several techniques, particularly powerful for large systems with complex defects

Saddle Point Search Techniques



http://www.theochem.uni-stuttgart.de/kaestner/dlfind.html



Calculating Hessian

Partitioned Rational Function Optimizer

Without Calculating Hessian

Minimum Mode Following

Dimer

Lanczos

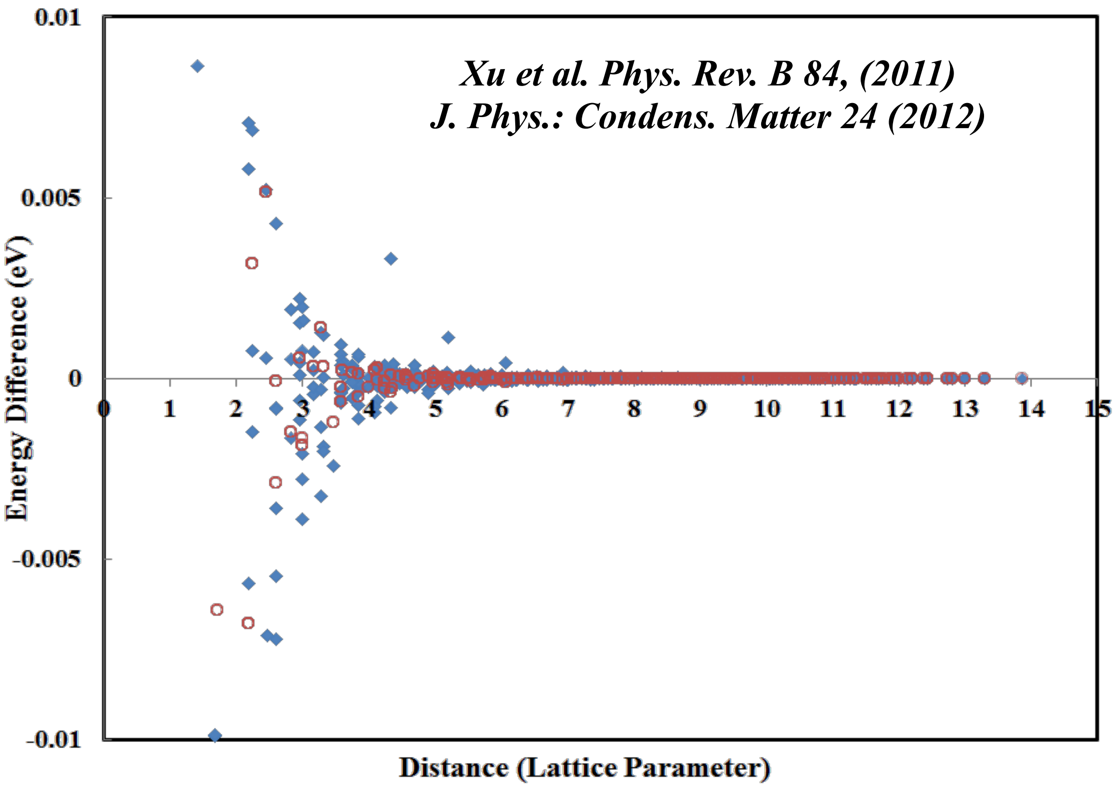
Others

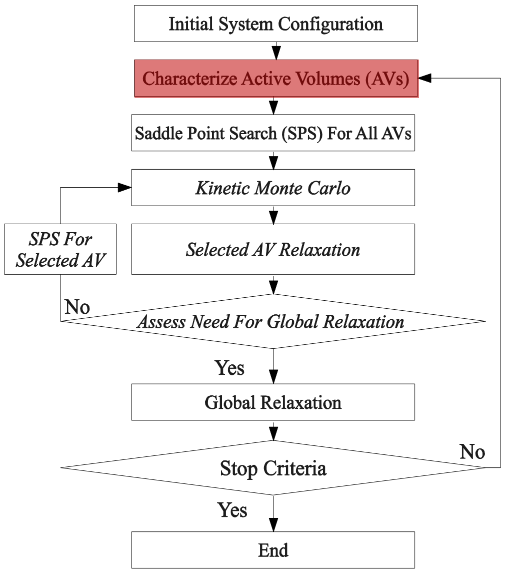
ABC+NEB

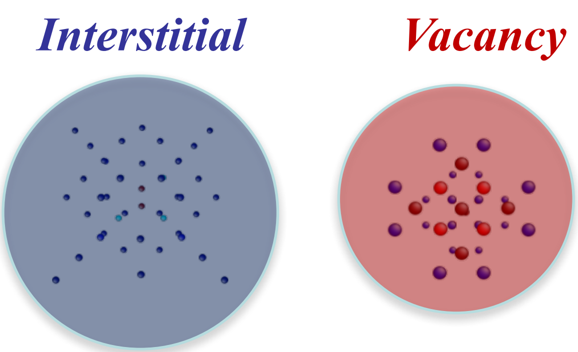
- ***Find migration barriers on-the-fly, based on harmonic transition state theory***
- ***Only need initial configuration; find the saddle point configurations***

Part I: Active Volumes-Concept and Application

Point defects are localized so activate volume can be defined based on energy, stress/strain criteria

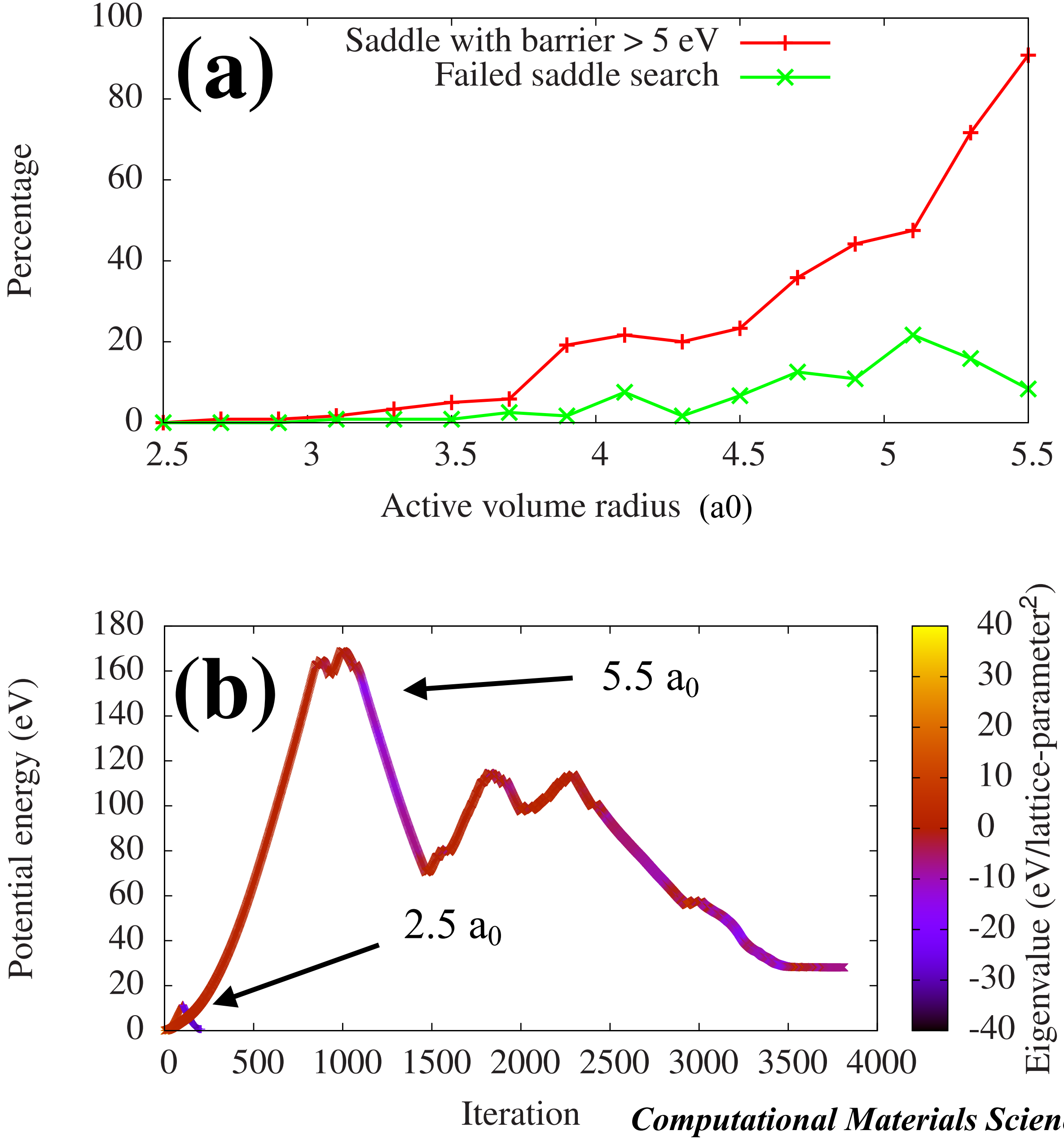






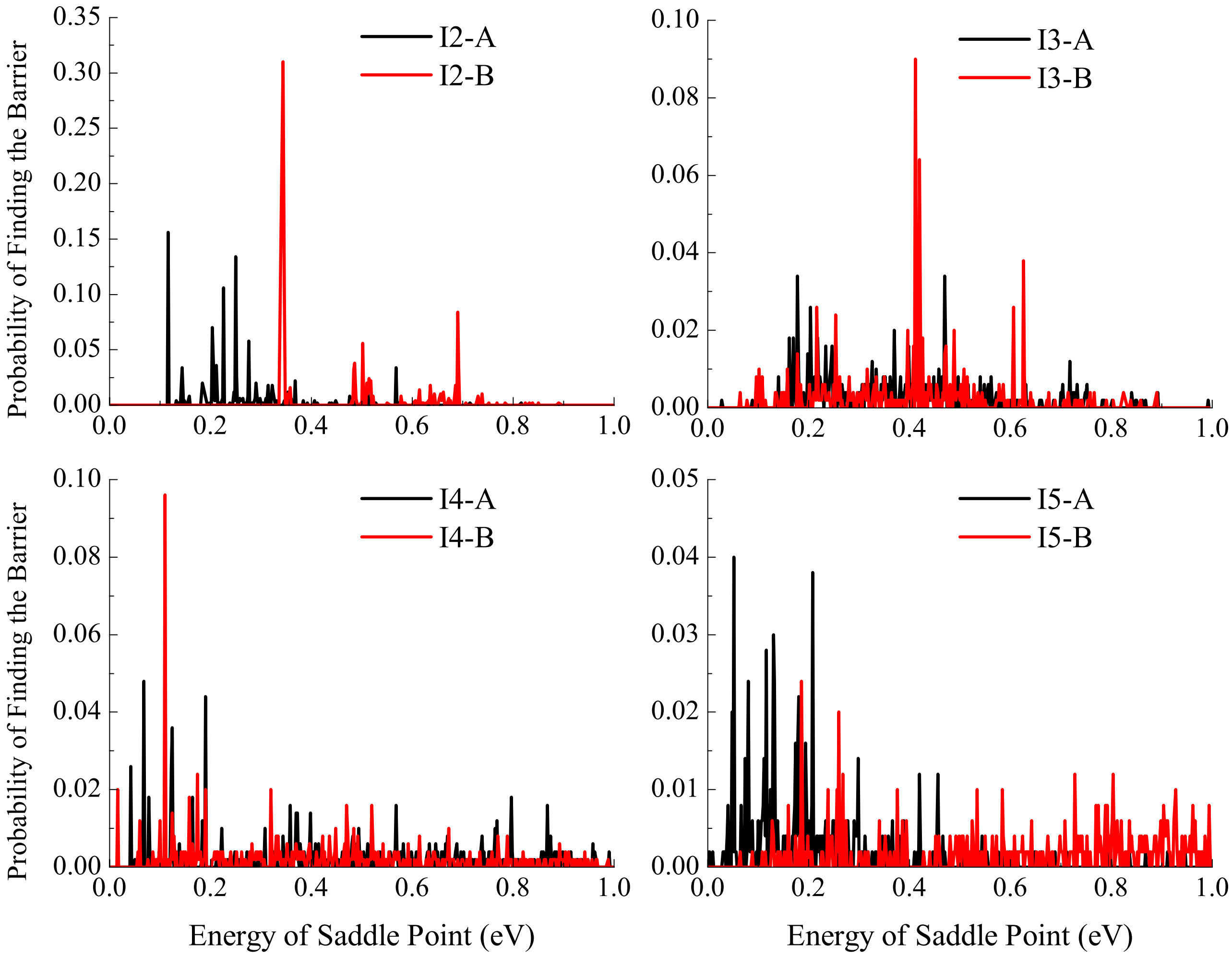
Saddle point searches are only carried out within the active volumes, significantly reduce the computational cost

AV size



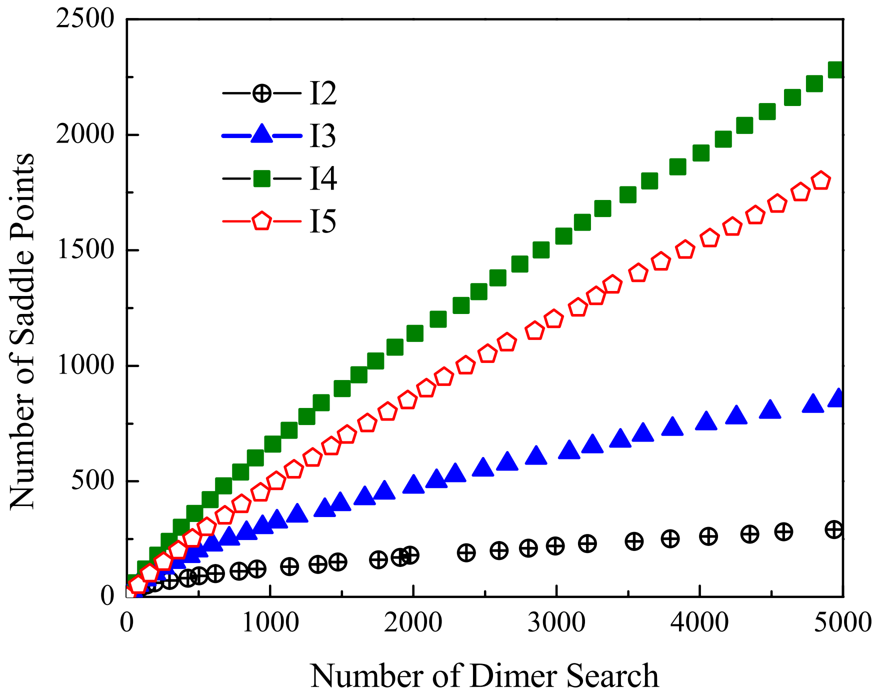
Computational Materials Science 100, p135(2015)

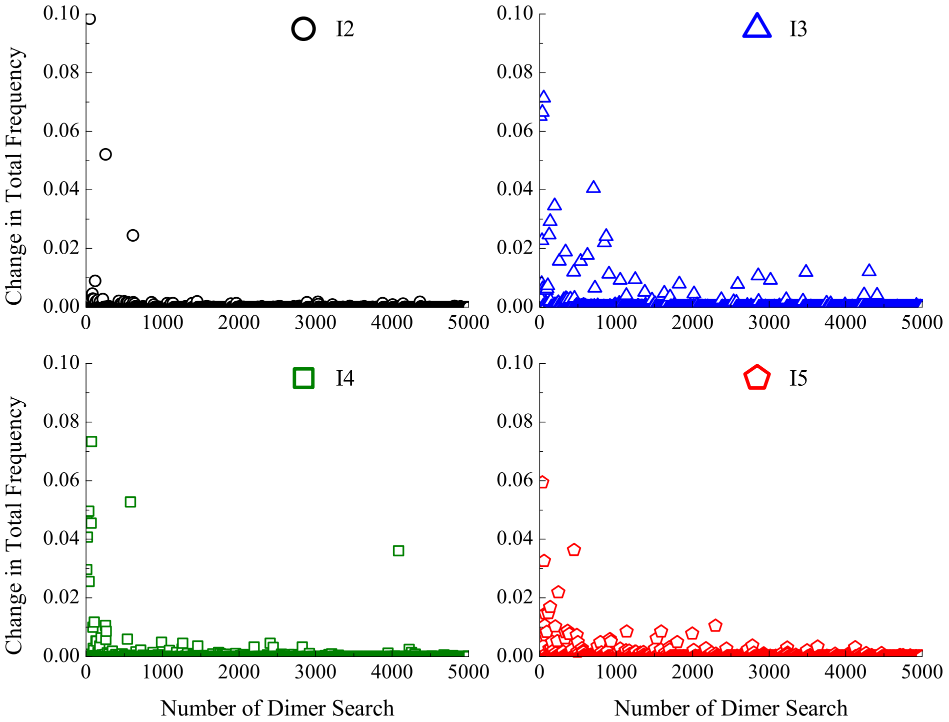
Saddle Point Searches



J. Phys.: Condens. Matter 24 (2012)

Number of Unique Saddle Points





J. Phys.: Condens. Matter 24 (2012)

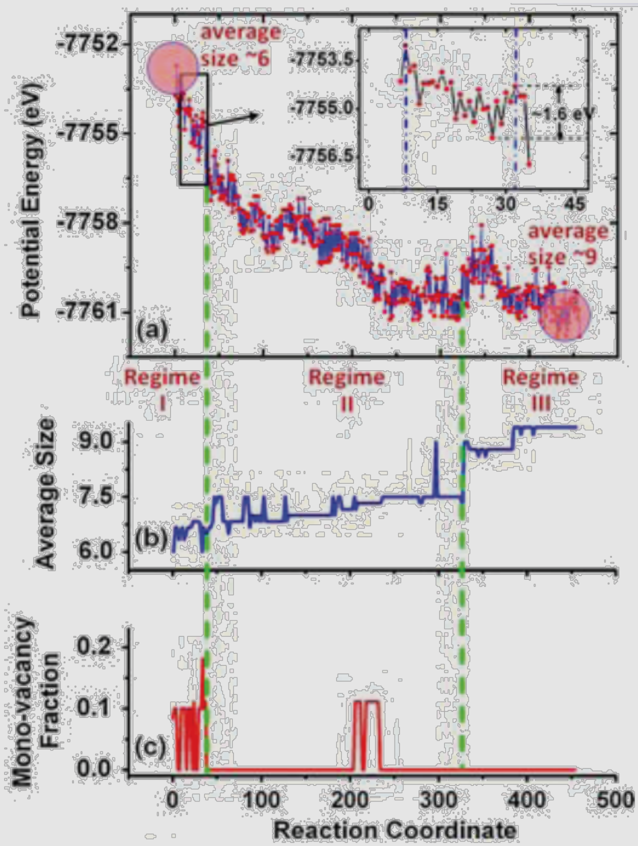
For interstitial clusters (complex defects), the number of unique saddle points does not converge.

Vacancy Clustering in Iron

For vacancy clusters grow beyond 6.5 vacancies (system energy is around -7760 eV)

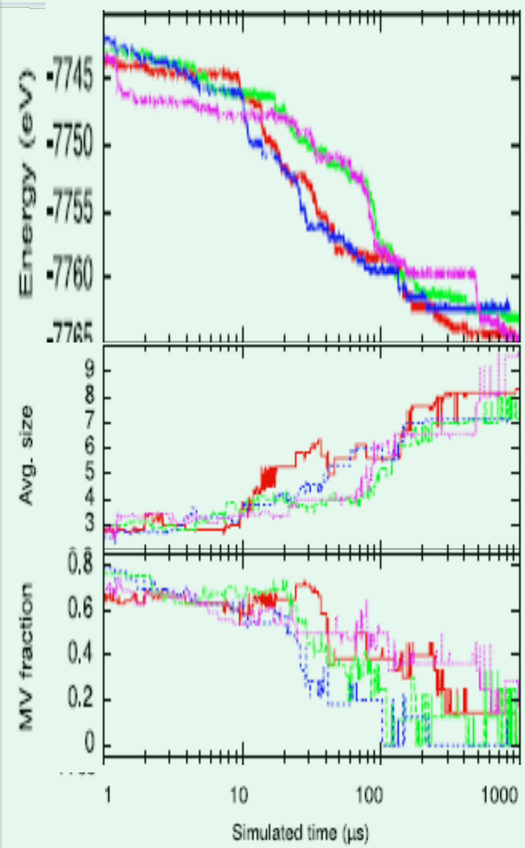
Autonomous Basin Climbing +KMC
20000 seconds at 150 °C

Phys. Rev. Letter 106, (2011) 125501



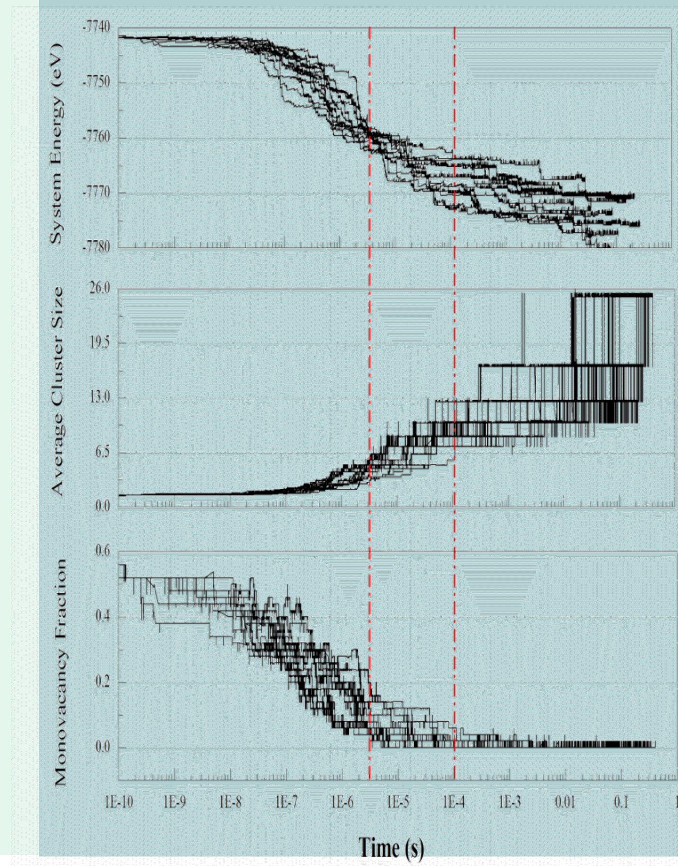
Kinetic Activation Relaxation Technique
0.5 ms at 50 °C

Phys. Rev. Letter 108, (2012) 219601

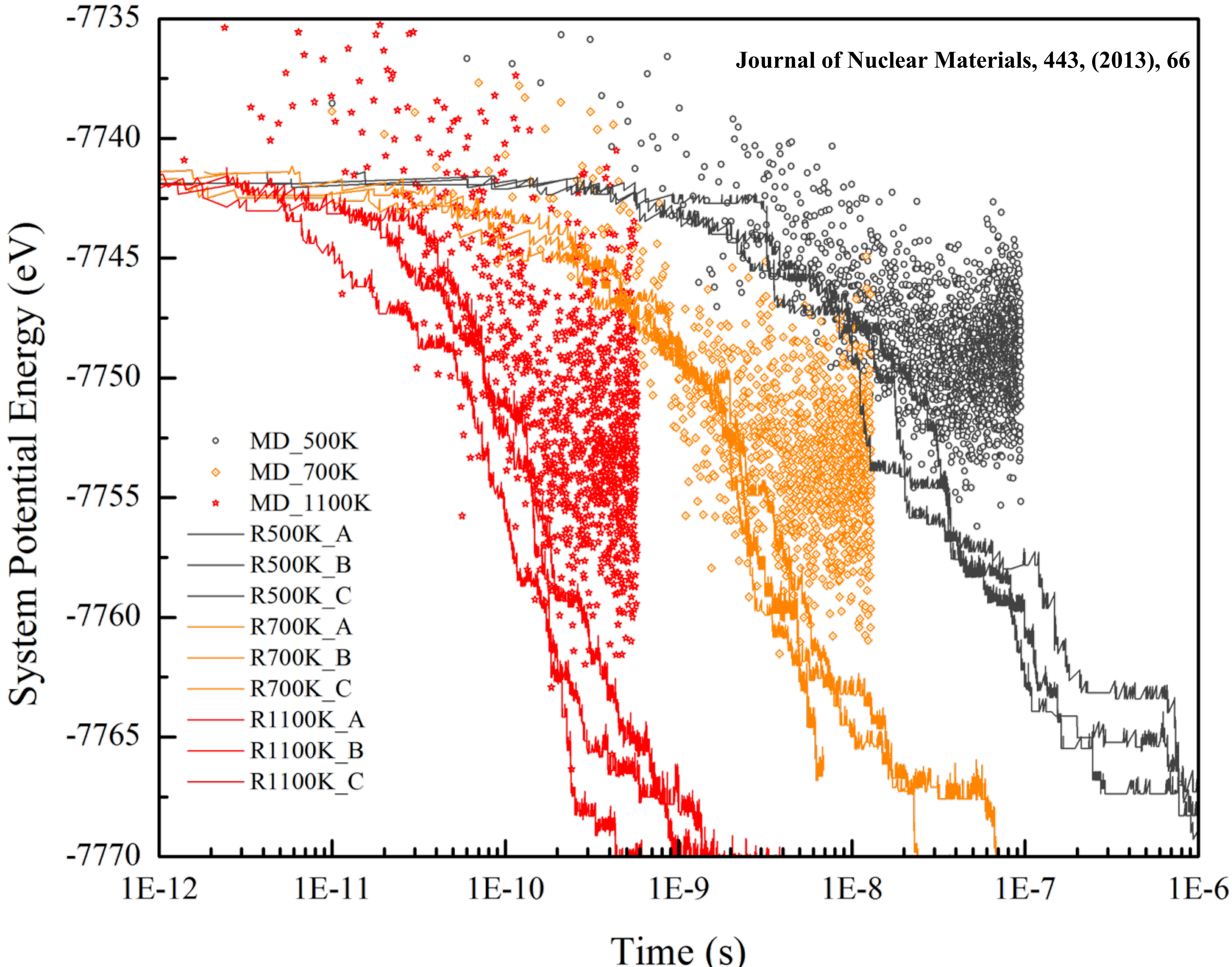


SEAKMC
0.005-0.1 ms at 50 °C

Journal of Nuclear Materials, 443, (2013), 66



Vacancy Clustering in Iron: SEAKMC vs MD



SEAKMC agrees well with MD at high temperatures

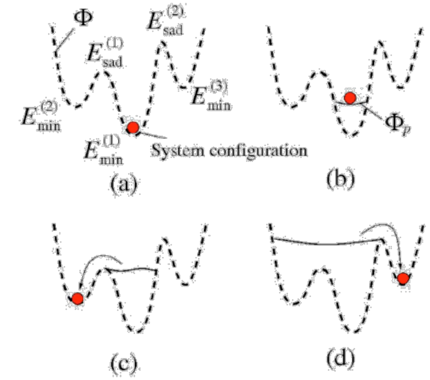
Where does the difference come from?

- ABC+KMC
- One saddle point (of minimum energy barrier) is used per KMC step
- Missing low-energy reaction paths

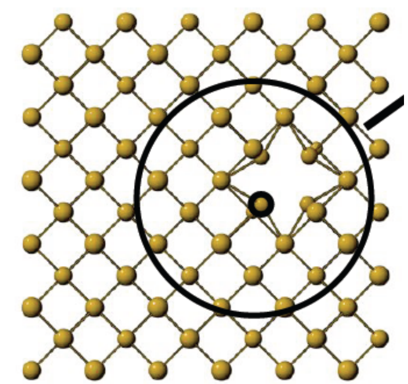
- kART
- Topological classification of configurations based on the 2nd/3rd nearest neighbors
- Basin Filling Methods

- SEAKMC
- Active volumes are sufficiently large
- Nearly complete sampling of saddle points

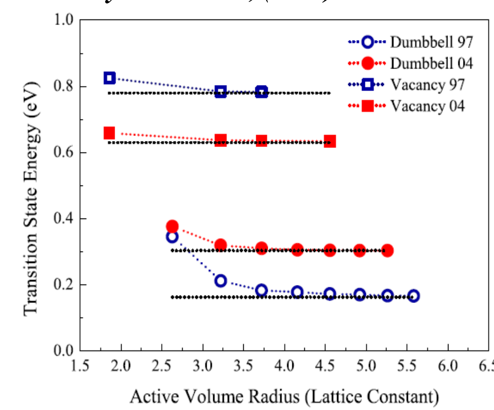
Beland et al, Computational Materials Science 100, p124 (2015)



J. Chem. Phys. 130, (2009) 224504



Phys. Rev. B 78, (2008) 153202



Xu et al. J. Phys.: Condens. Matter (2012)

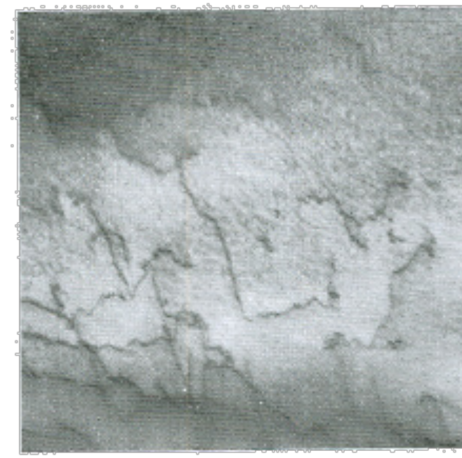
Interstitial Loops -Unique Signature of Radiation Damage

- Eyre 1962: Discovery of interstitial loops in bcc Fe
- Masters 1964: Two population of loops ½ <111> and <100> loops
- ½ <111> and <100> loops have very different mobility
- Eyre-Bullough Mechanism 1965

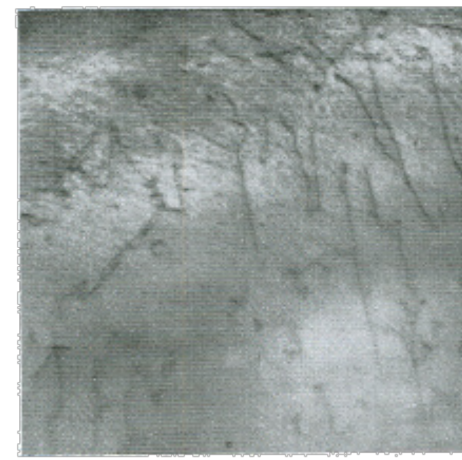
  ½[110]+½ [001]=½ [111]

  ½[110]+½ [1-10]=[010]

- Because of the high stacking fault energy, ½<110> loop was not observed experimentally and spontaneously transform into ½<111> loops in simulations. Therefore, Eyre-Bullough mechanism is not applicable.



(a)



(b)

Dislocation arrangements in α-iron after a dose of 5×10^{19} n cm^{-2} followed by deformation. (a) Dislocations pinned by irradiation obstacles. ×40 000. (b) Loops left by dislocations breaking away from pinning points. ×40 000.

B. Eyre Phil. Mag. (1962)

Recent Atomistic Studies

Collision cascades

- **SIA clusters are formed in cascades, mainly of ½<111> type**

Mutual interaction of dislocation loop

Marian 2002: proposed a modified Eyre-Bullough Mechanism

½[111]+½ [00-1]=½ [110]

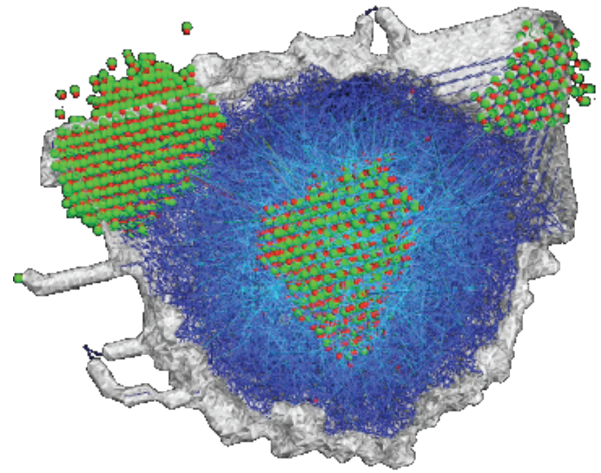
½[110]+½ [1-10]=[010]

Terentyev 2008: MD simulations

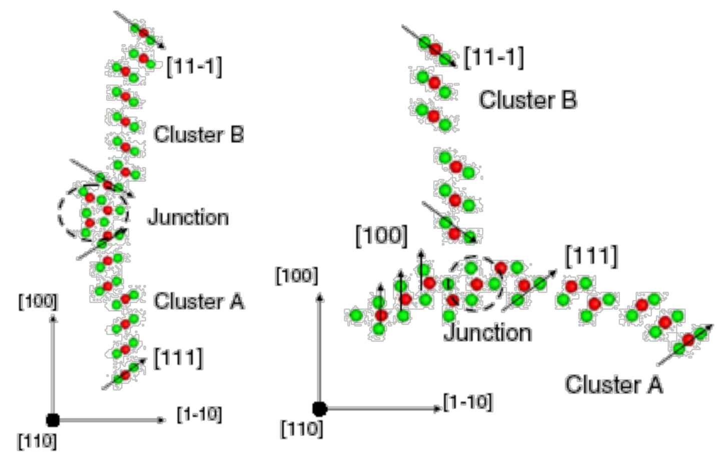
- *Only <111> loops are observed from mutual interactions*

Loop Stability

Dudarev 2008: developed a model to include the temperature dependence of an anisotropic elastic self-energies of dislocations in iron



A. Calder et al. Phil. Mag. (2010)



D. Terentyev et.al., J. Nucl. Mater. (2008)

The <100> loop formation mechanism remains undetermined after fifty years since it was first discovered in 1962

Simulation Setup

- Simulation methods: SEAKMC and molecular dynamics
- Interatomic potentials: A97, A04, M07
- System size: $27a_0 x 27a_0 x 27a_0$
- Temperature: From 0 K to 1200K
- Interaction angle: acute and obtuse
- Interstitial cluster(IC) size: 19,37,61
- Active volume size: 4.5-7.5 a_0

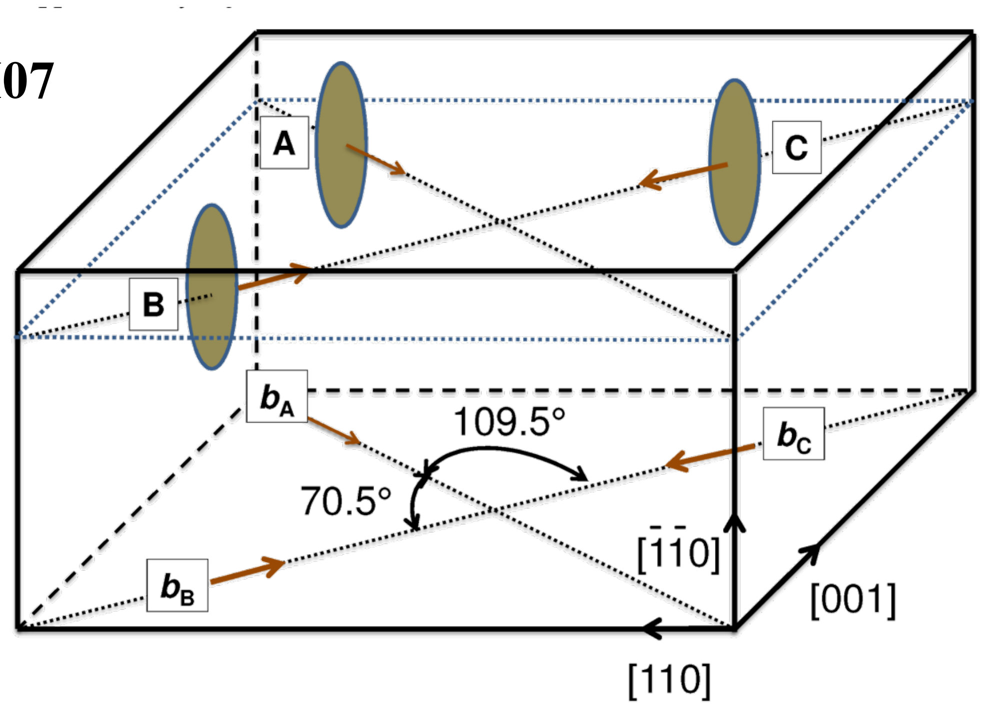
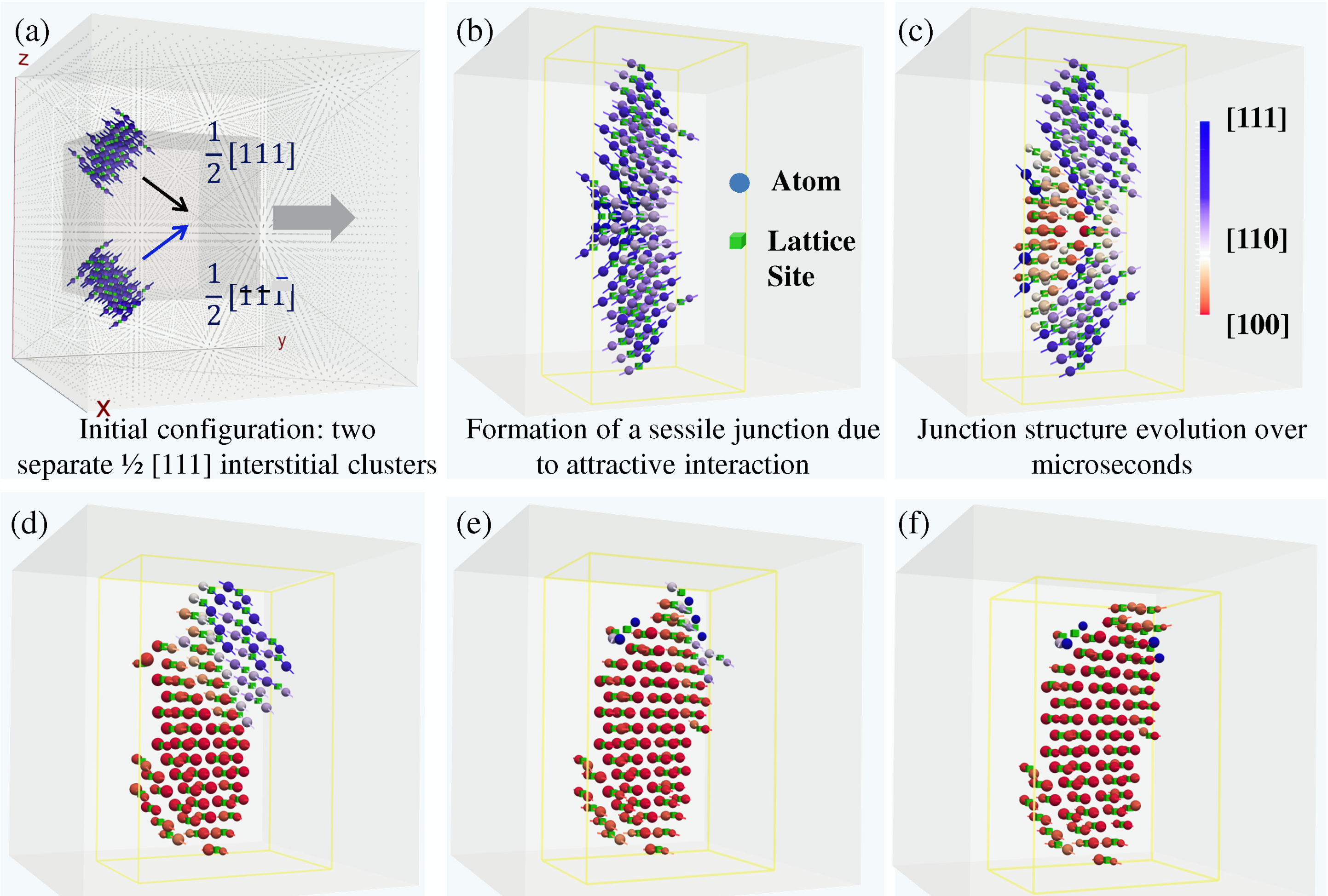


Illustration of <111> type interstitial clusters moving in a {110} plane with their angle of intersection projected onto the corresponding base plane

[100] Loop Formation Process from SEAKMC



(a) Initial configuration: two separate ½ [111] interstitial clusters

(b) Formation of a sessile junction due to attractive interaction

(c) Junction structure evolution over microseconds

(d) Partial [100] orientation formed as configuration evolves

(e) Further propagation of [100] orientation

(f) Complete [100] loop formed with point defects at boundary

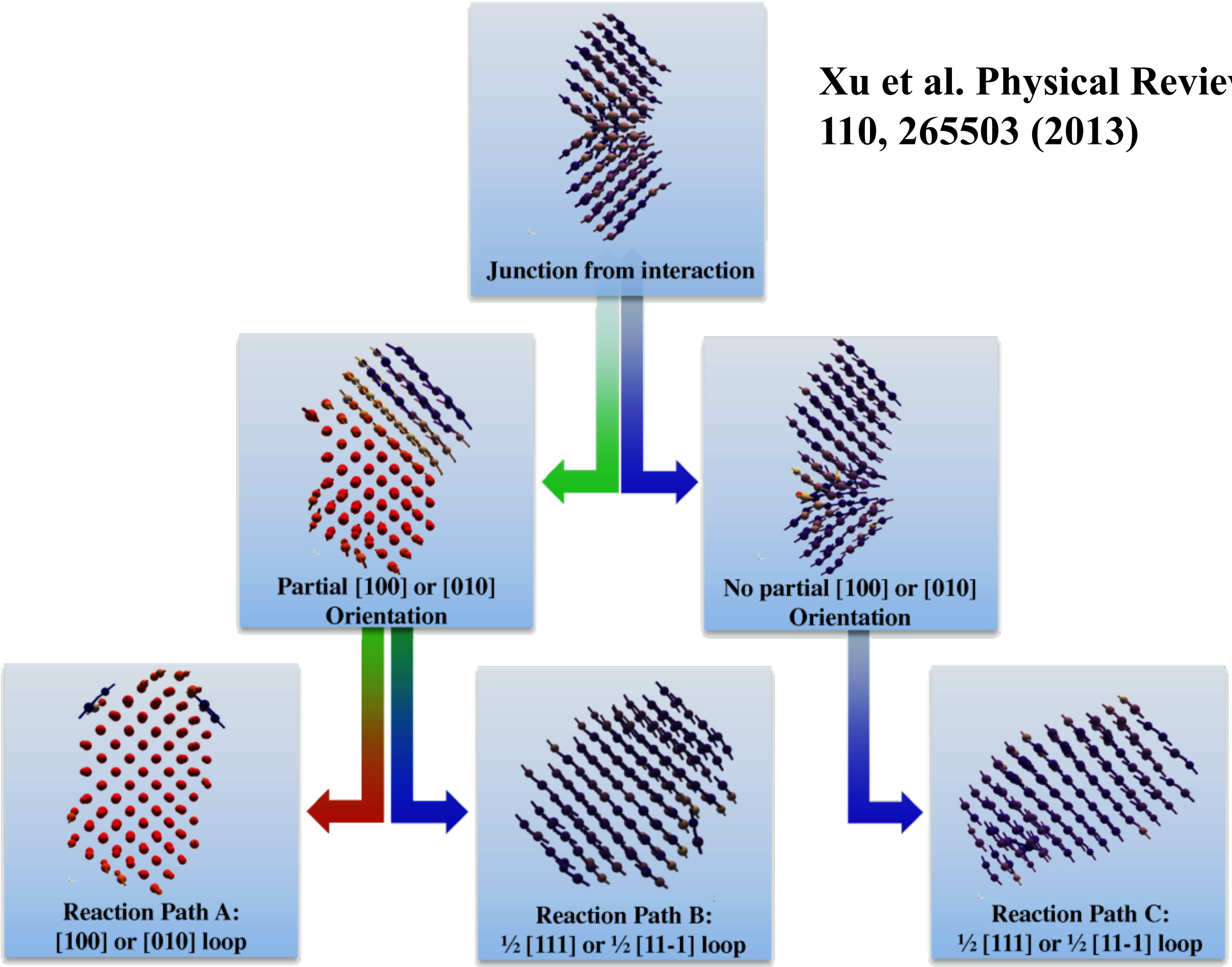
Xu et al. Physical Review Letters, 110, 265503 (2013), Journal of Alloys and Compound (2015)

Never previously observed, distinctly atomistic process

Different from all previous proposed mechanisms

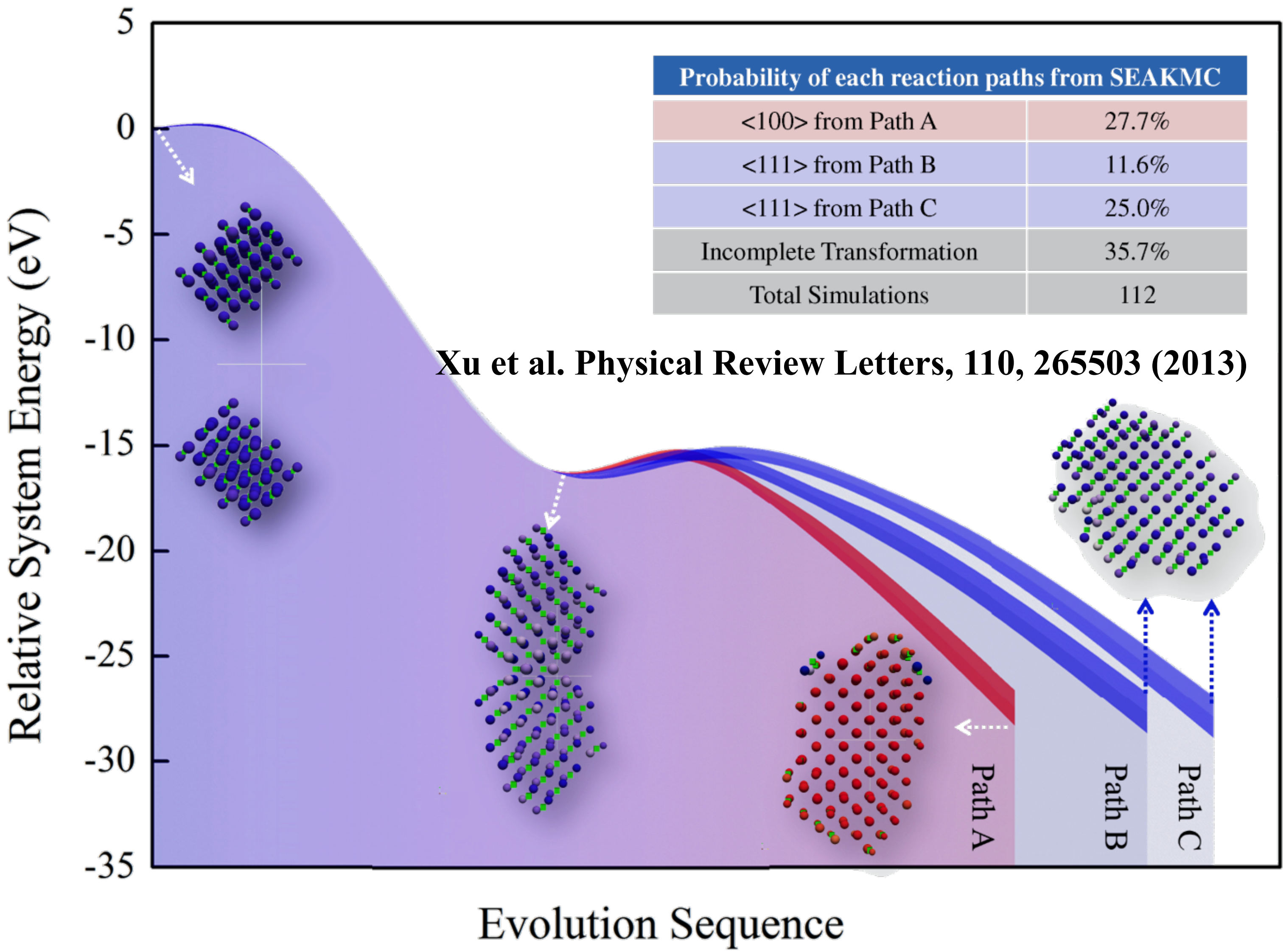
Stochastic Process in Nature

Xu et al. Physical Review Letters, 110, 265503 (2013)



Different configurations may form from the same initial structure

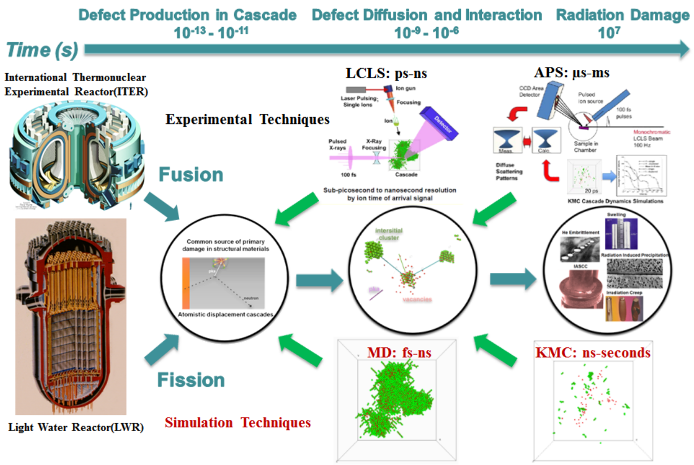
Energetics of the Evolution Process



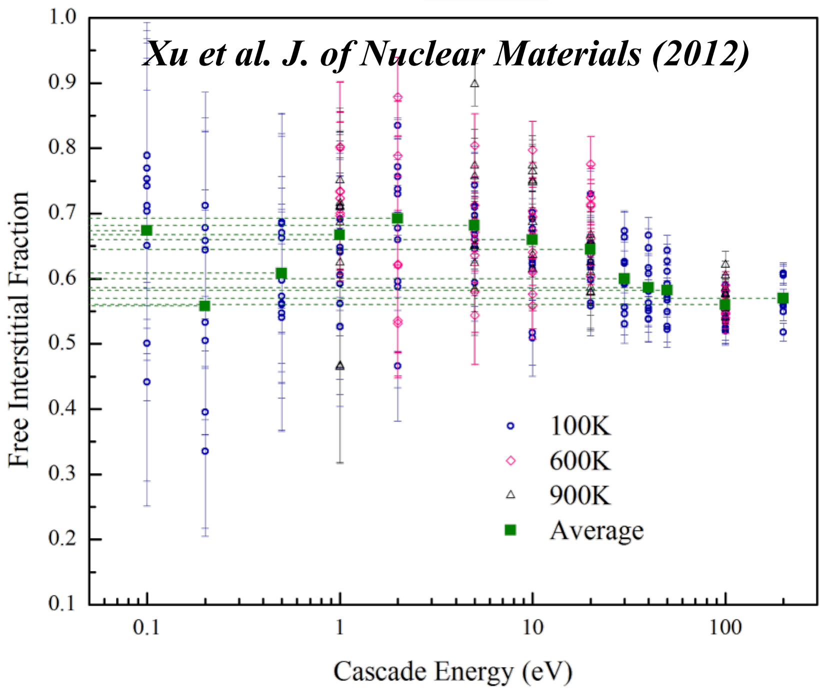
Probability of each reaction paths from SEAKMC	
<100> from Path A	27.7%
<111> from Path B	11.6%
<111> from Path C	25.0%
Incomplete Transformation	35.7%
Total Simulations	112

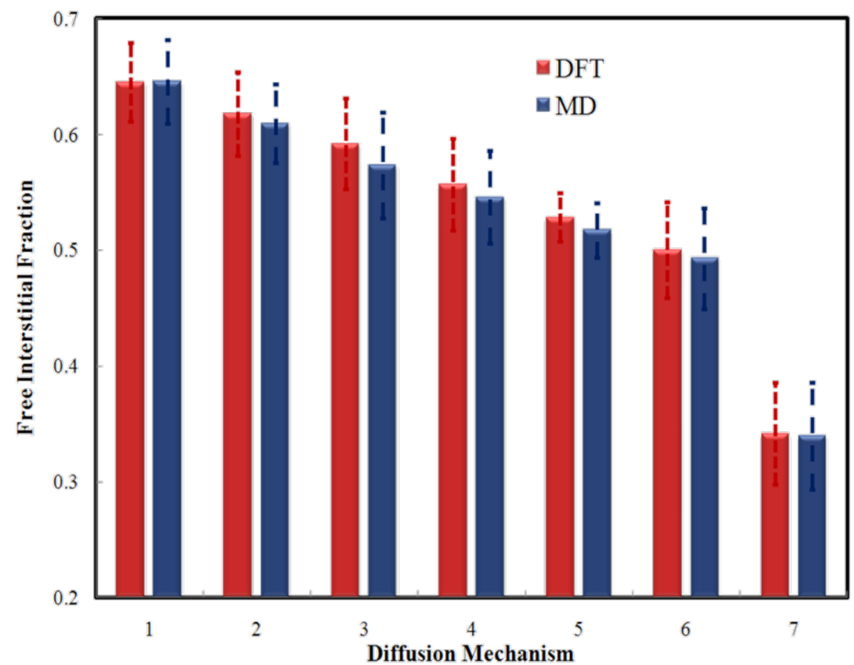
MD employs high temperature to speed up the process, significantly increasing the stability of <111> loop due to the its larger entropy

Cascade Annealing-Comparison of OKMC and SEAKMC



- Initial structure from MD cascade simulations
- Cascade energy is 10 keV and 25 keV
- System size: 128,000 and 250,000 , with an absorbing boundary condition
- Annealing temperature : 650 K

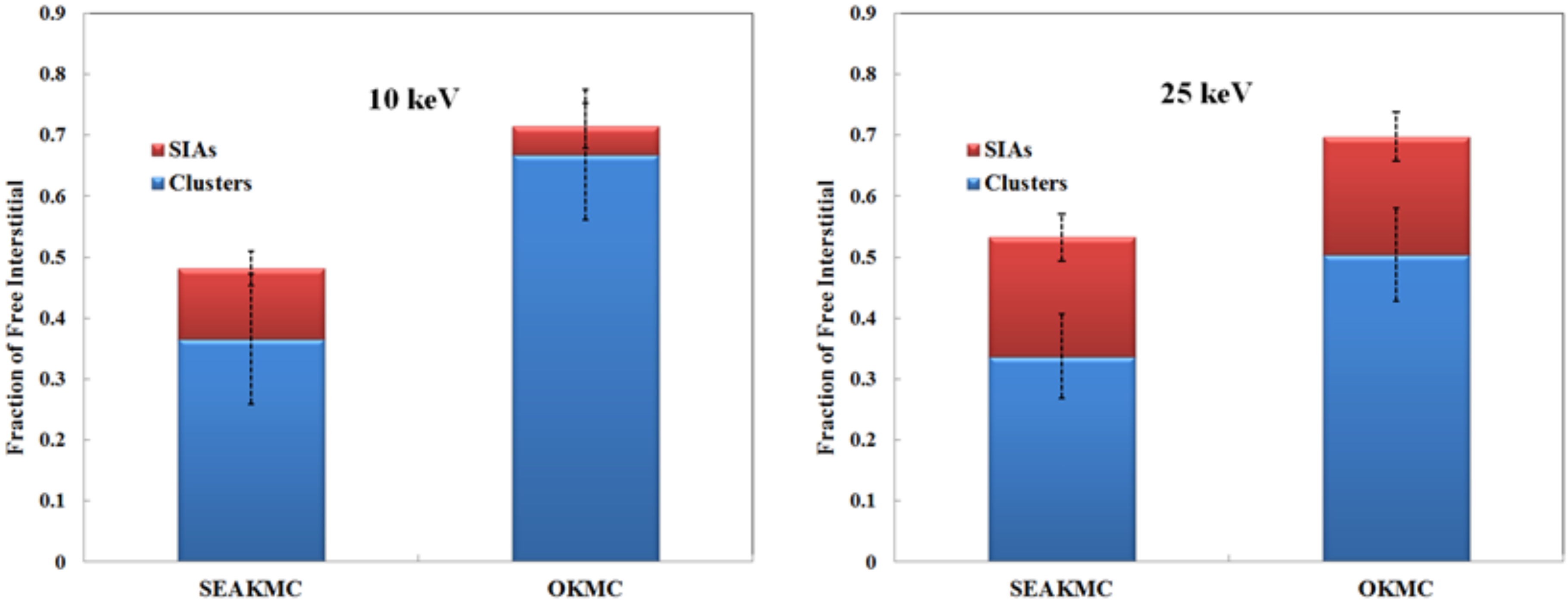




Parameter-free SEAKMC provides different defect survival fractions

Comparison of OKMC and SEAKMC-Continued

Simulation were perform under the same condition using both methods



Parameter-free SEAKMC provides different defect survival fractions

SEAKMC can help OKMC to increase the accuracy of the simulations

Challenges

On-the-fly KMC methods have made significant progresses.

- ***How to efficiently determine saddle points if a large number of atoms is involved in the transition states***

- ***How to ensure sufficient sampling of saddle points for complex defects?***

- ***How to validate the accuracy of on-the-fly KMC methods?***

Thank you very much for your attention!