Solid-state Precipitation in Molecular Dynamics: kMC-MD Hybrid Simulations

Jacob Tavenner[1], Mikhail I. Mendelev[2], John W. Lawson[2]

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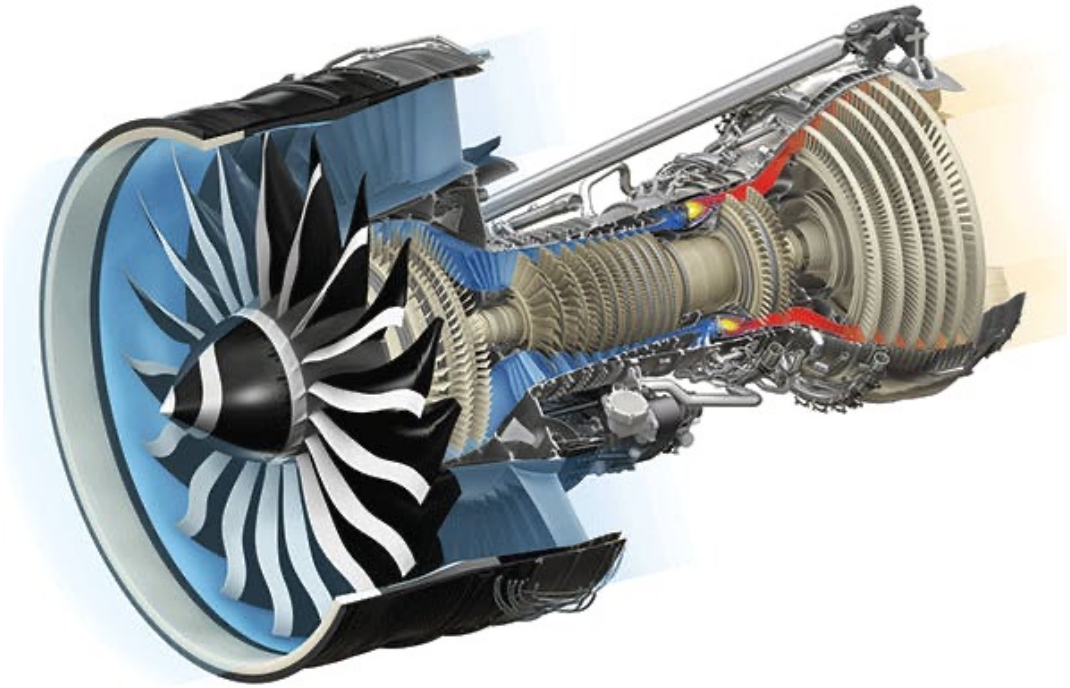
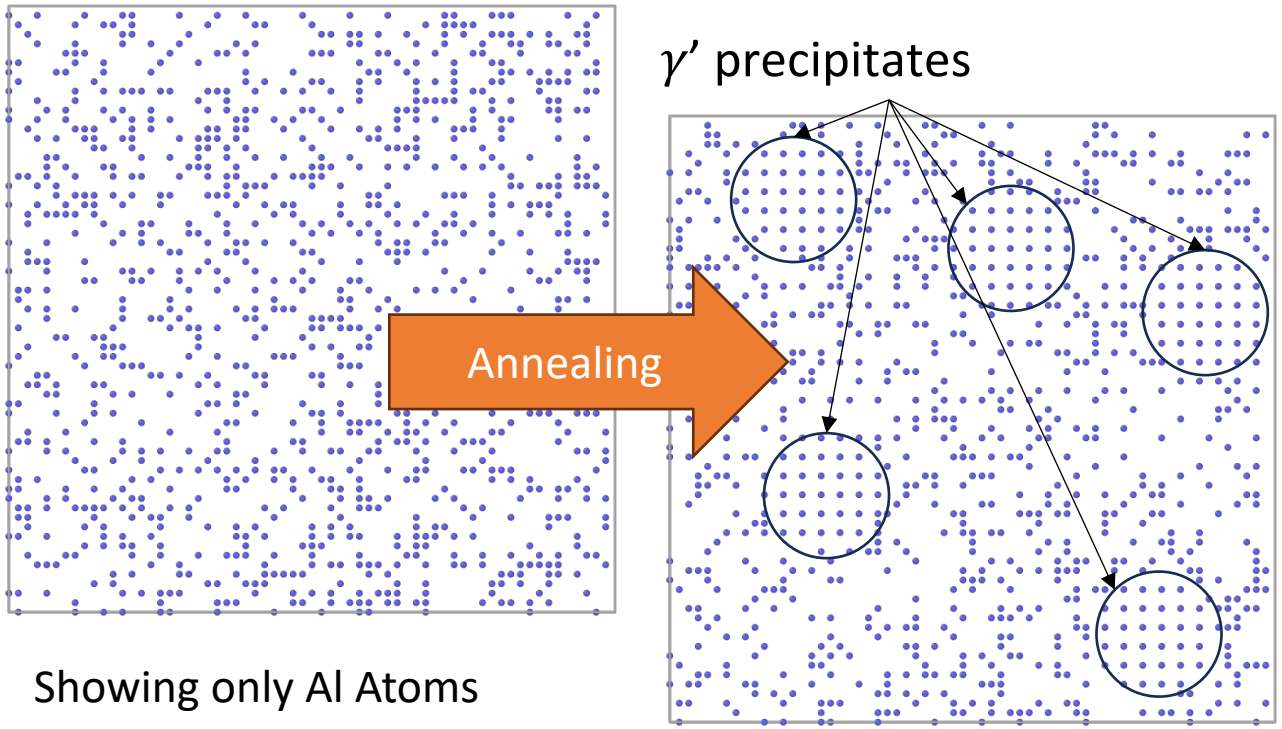
[1]*KBR, Inc., Intelligent Systems Division, NASA Ames Research Center, Moffett Field, CA, USA*

[2]*Intelligent Systems Division, NASA Ames Research Center, Moffett Field, CA, USA*

Ni-based Superalloys

- γ' (Ni_3Al – L_{12}) precipitates are key in controlling creep properties in advanced superalloys
- Determining annealing conditions to produce precipitates of specific size and distribution through experimental trial-and-error
- Fundamental parameters of nucleation difficult to obtain through experiments





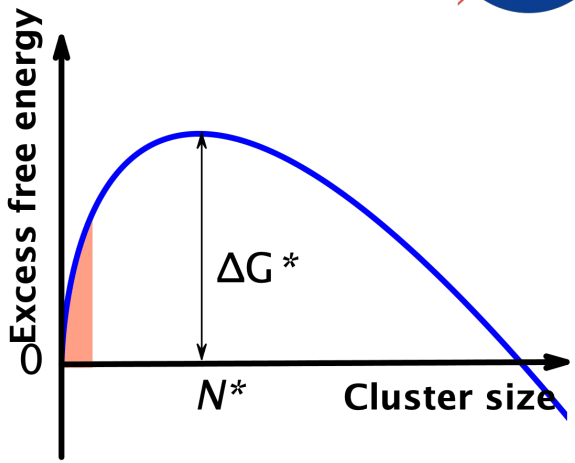
Solid-State Phase Precipitation

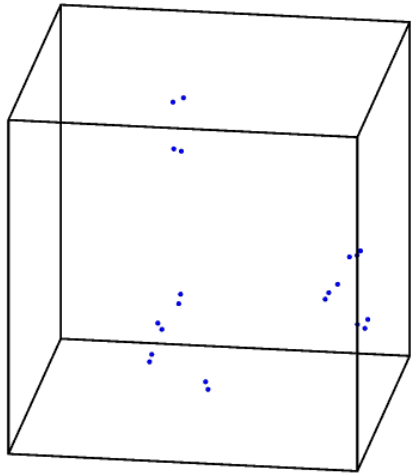
- Precipitation of new phases relies on solid-state diffusion to distribute chemical species evenly
- Majority of time spent in sampling small cluster sizes
- Solved for Liquid-Solid (Persistent Embryo Method)
- Precipitate grows once energy penalty for nucleation is overcome

$$\Delta G = V\Delta\mu + A\sigma$$

- Calculation of interfacial energy through determining critical nucleus size

$$\frac{R^*\Delta G}{2} = \sigma$$





Solid-state Diffusion is too slow for observing precipitation in MD simulation

Solid State Diffusion – kMC in MD

Challenge:

- Inaccessible to MD simulation due to short timescales
- Atom-based diffusion models use lattice-based kMC technique
- How can we combine kMC diffusion with MD-based physics?

Solution:

- Use Voronoi-based neighbor selection to moderate solute movement between nearest-neighbors under Monte-Carlo based sampling, implemented in LAMMPS

Examples:

- Test by illustrating replication of ideal diffusion profiles
- Show precipitate growth in Ni-based superalloy
- Use kMC-MD simulation to calculate interfacial energy from critical nucleus

KMC-MD Solute Diffusion

Predicted ideal solute diffusion vs. KMC

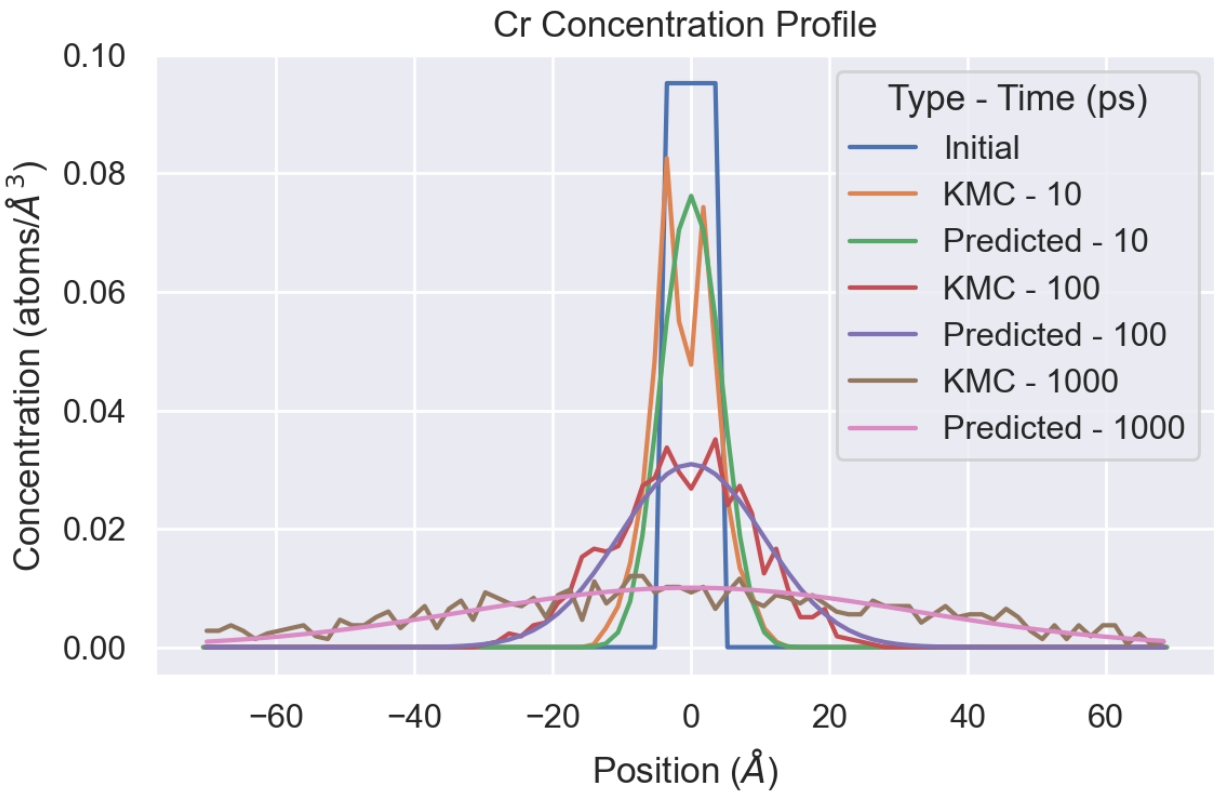
- Initialize fully-periodic cell of Ni with central concentration of Cr
- 3.5 x 3.5 x 14 nm
- 1000K MD/MC Temperature
- Swap rate of 1 atom/fs
- D = 0.001 Å²/fs

$$c(x,t) = \frac{c_0}{2}\left(erf\left(\frac{x+\frac{w}{2}}{\sqrt{4Dt}}\right) - erf\left(\frac{x-\frac{w}{2}}{\sqrt{4Dt}}\right)\right)$$

w = Initial Pulse Width, D = Diffusivity

x = Position, t = Time

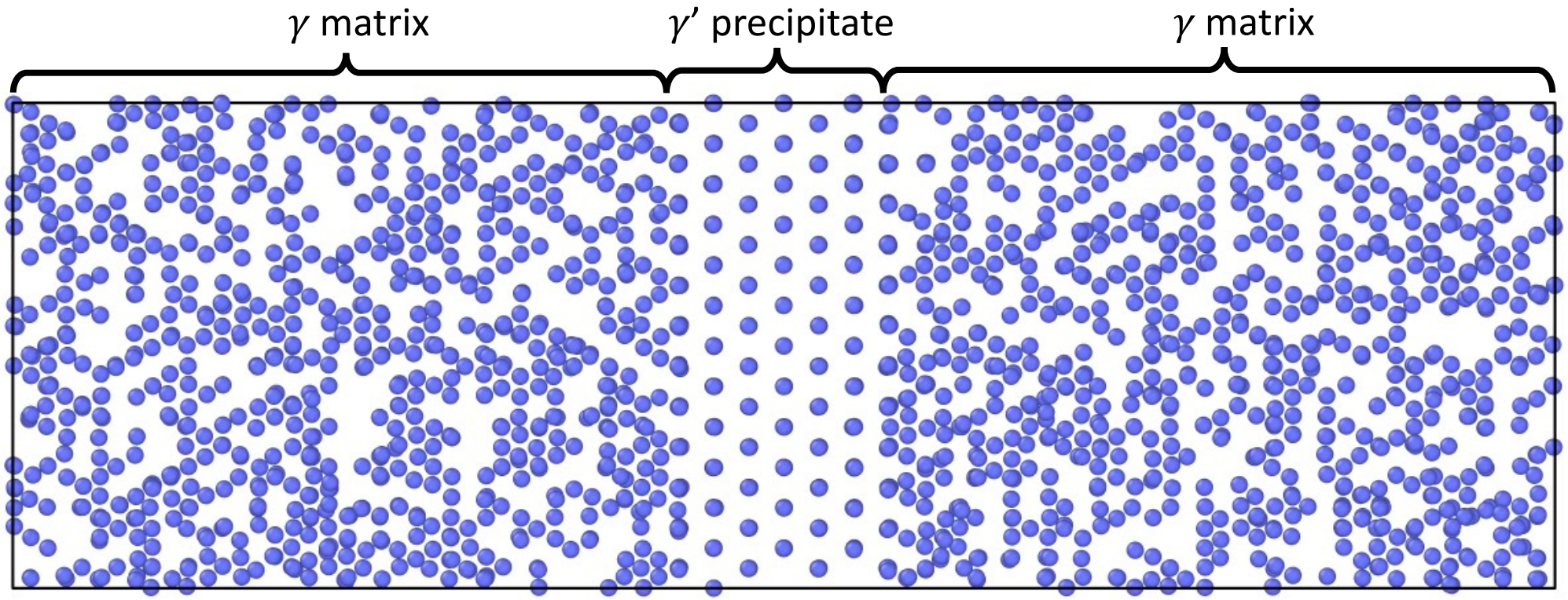
$$D = \frac{r^2}{12}$$



Good agreement between KMC-MD simulation and diffusion equation

J. Tavenner et. al, *Computational Materials Science* 213, 5 (2023).

KMC-MD Precipitate Growth



Swap 1% of Al atoms
every 50 MD steps

Showing only Al Atoms

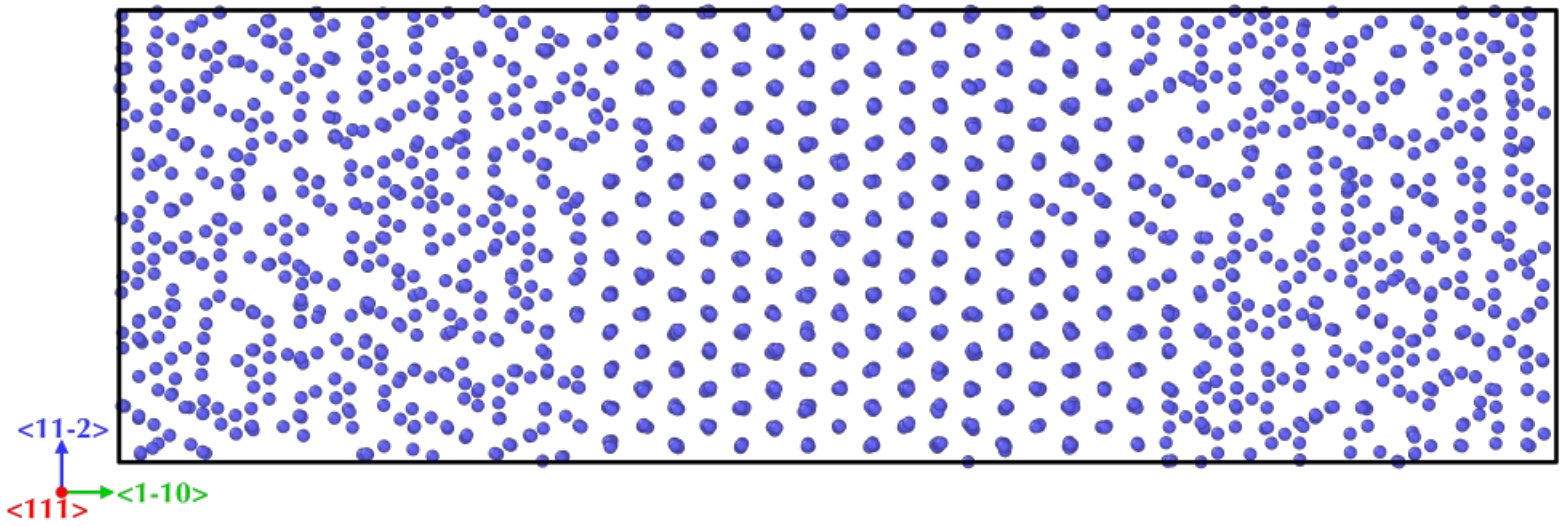
2128 Al Atoms
1000K, 0 Bar NPT

J. Tavenner et. al, *Computational Materials Science* 213, 5 (2023).

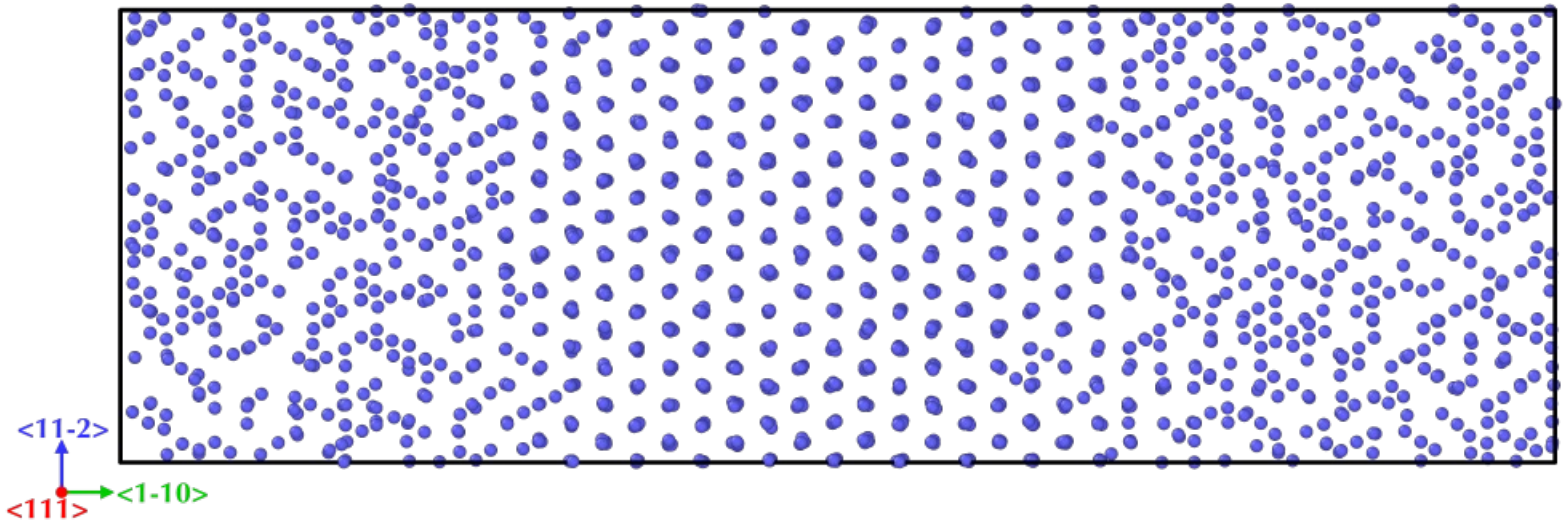
Final Structures – Al atoms

kMC-MD

Swap 1% of Al atoms every 50MD steps



MC-MD

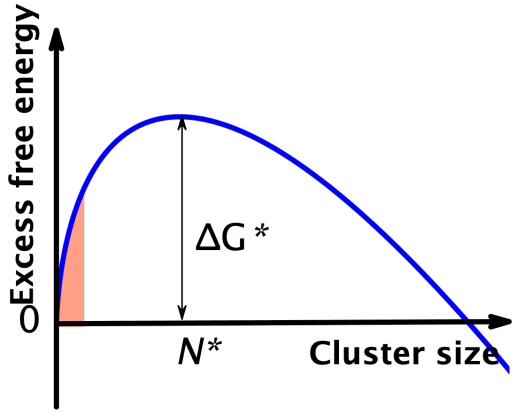


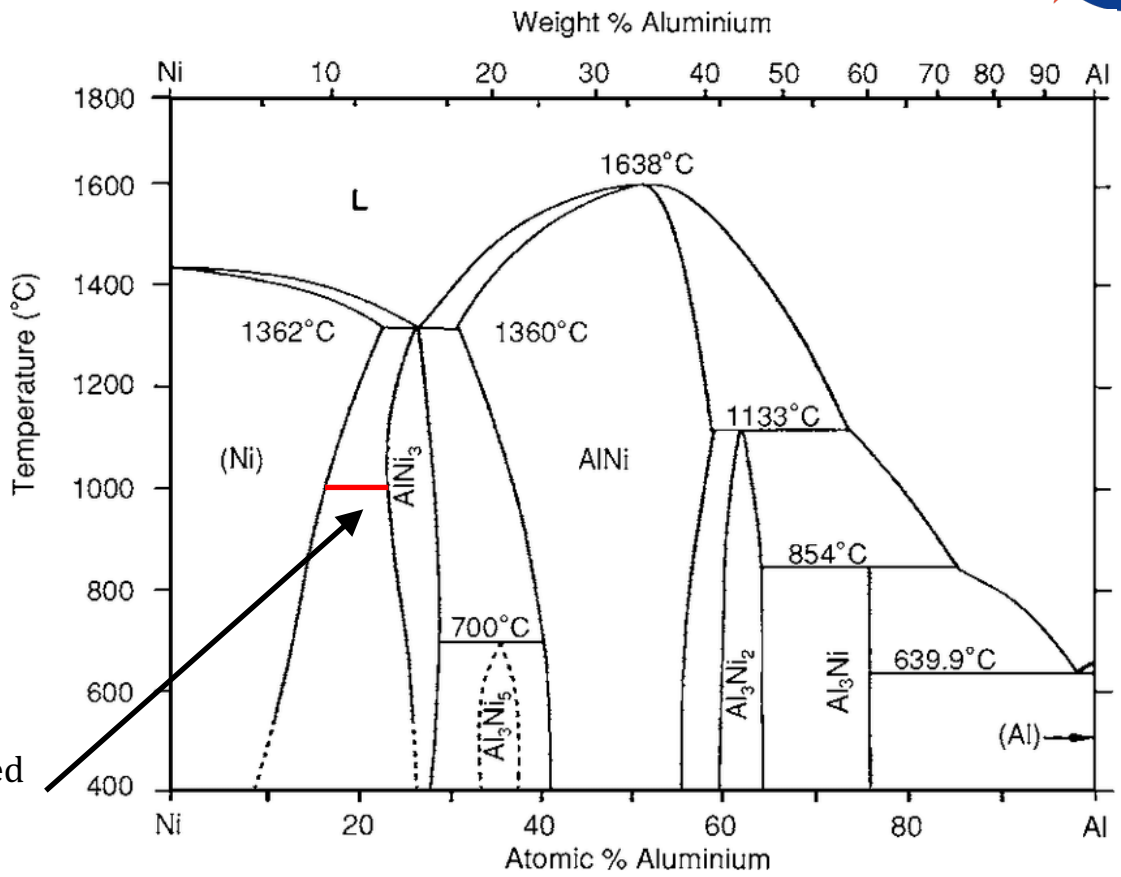
2128 Al Atoms
1000K, 0 Bar NPT

J. Tavenner et. al, *Computational Materials Science* 213, 5 (2023).

MD Simulation of Precipitation

- ❖ kMC-MD algorithm combines the time resolution of kMC and correct physics of MD simulation in LAMMPS.
- ❖ Demonstrated accelerated solute diffusion behavior
- ❖ Verified solid-phase growth behavior using flat interface
- Simulate phase precipitation from a solid solution using novel MD/kMC simulation technique.
- Determine critical nucleus size N*, convert to interfacial energy





Investigated Condition

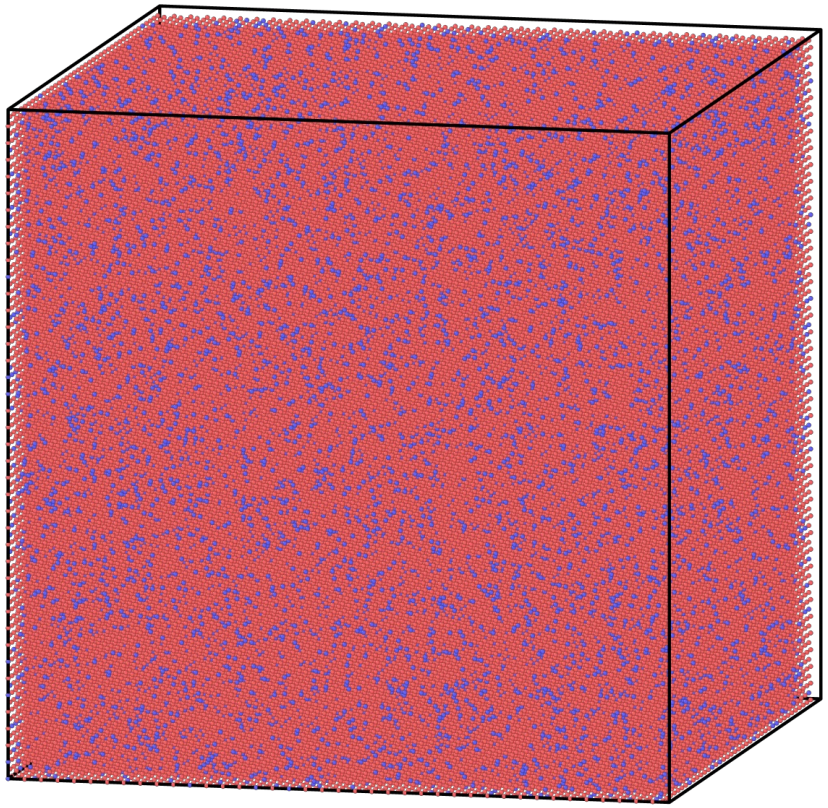
Equilibrium Condition @ 1000K: FCC Ni ~ 12.5% Al
L_{12} Ni-Al ~ 24% Al

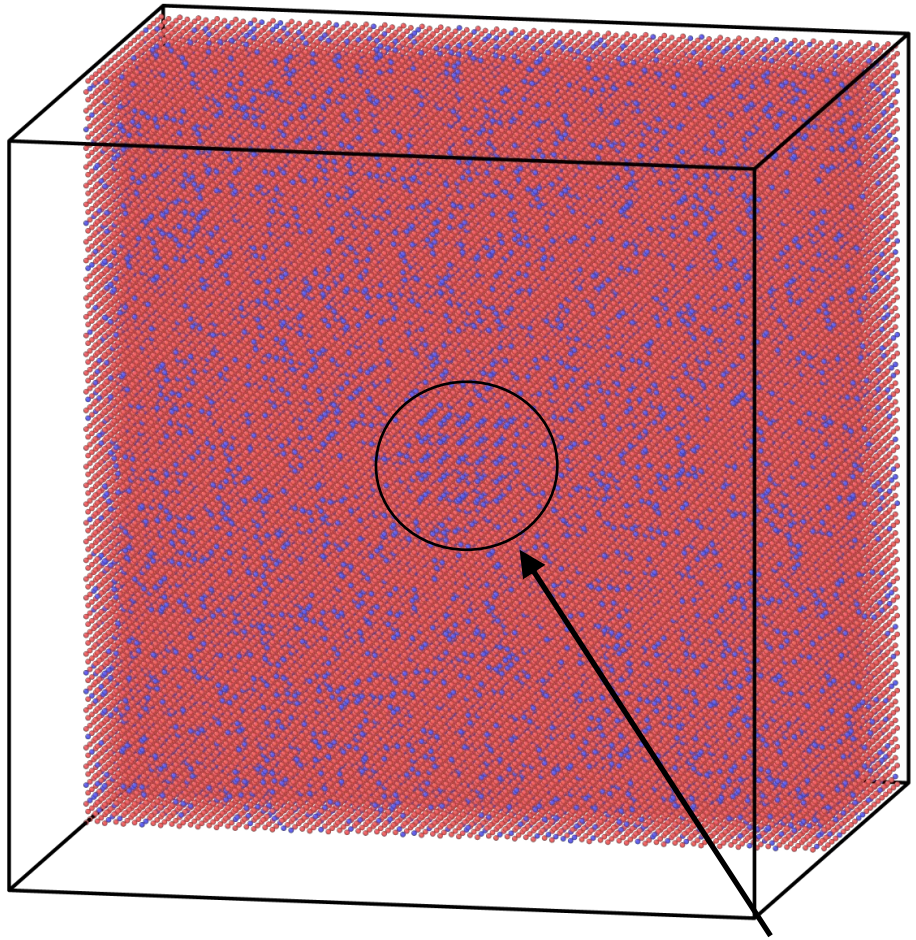
G. Lopez et. al, *Interface Science* 10(1):13-19 (2002).

Simulation Setup

- Need to identify critical nucleus size N*

- Ni-Al system
  - Initialize Cuboidal or Spherical L_{12} Precipitate Nucleus inside FCC Ni
  - Randomly distribute Al in remaining Ni matrix
  - Equilibrium matrix concentration ~12.5%
- Evolve system with KMC/MD
  - Swap Al atoms with neighboring Ni atoms to simulate diffusion process
  - Accelerating diffusion process accelerates precipitate growth

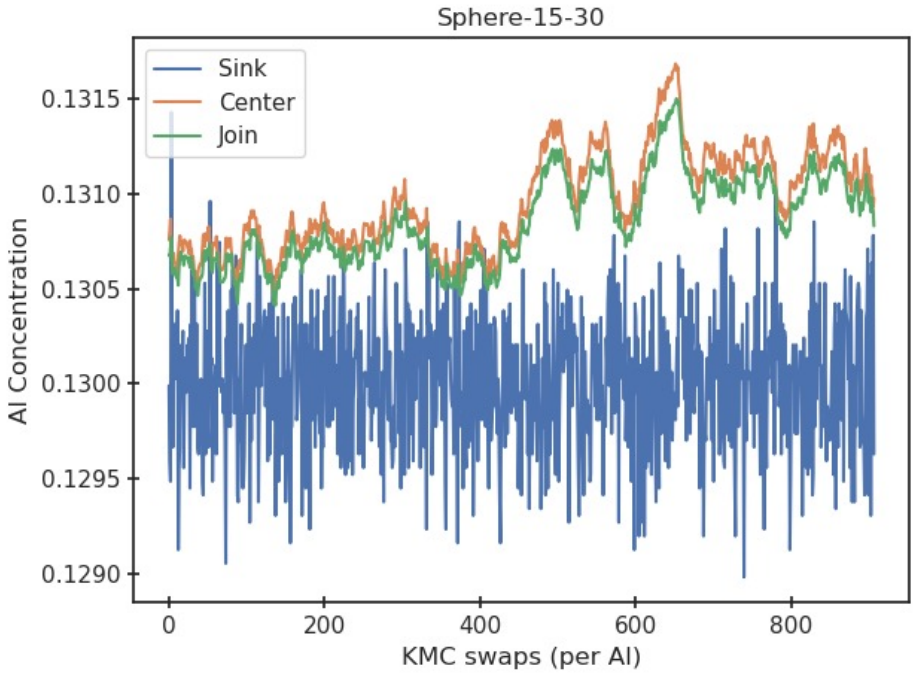




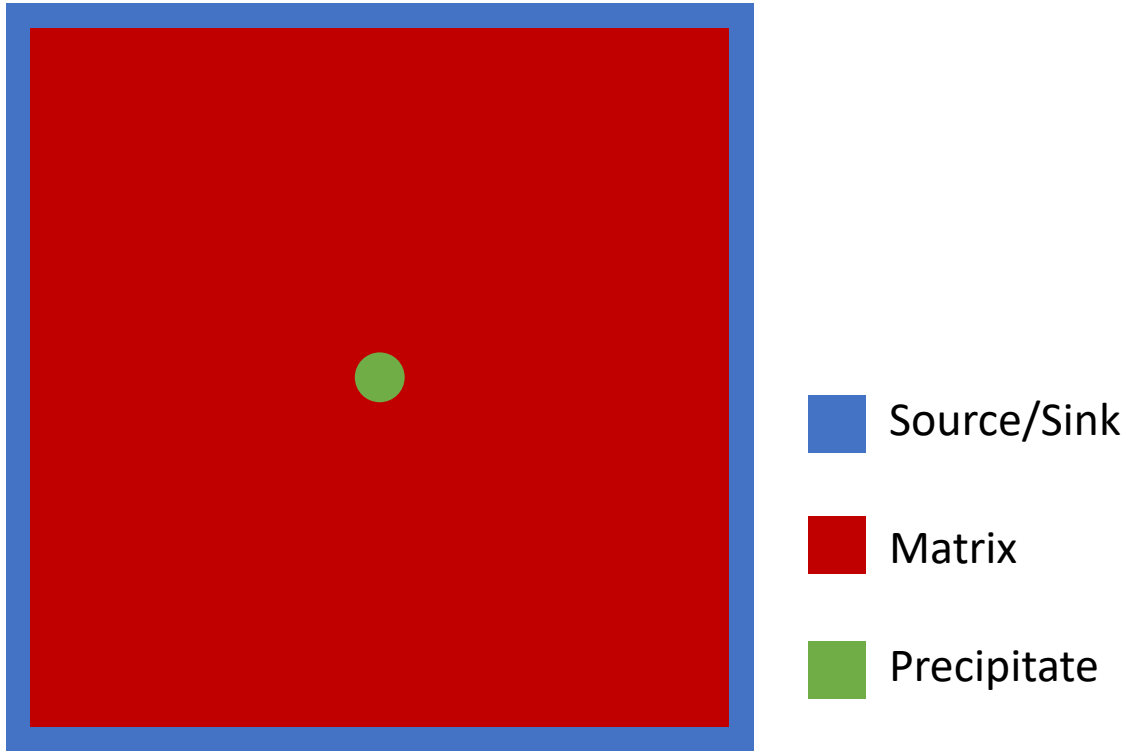
Ni atoms – Red
Al atoms – Blue

Constant Driving-Force MD Simulation

- Developed new procedure in LAMMPS for fixing composition of regional composition to specific concentration, varying the global concentration over the simulation
  - Calculate concentration in sink region
  - Determine difference from target concentration
  - Change required number of atoms to new type to meet target

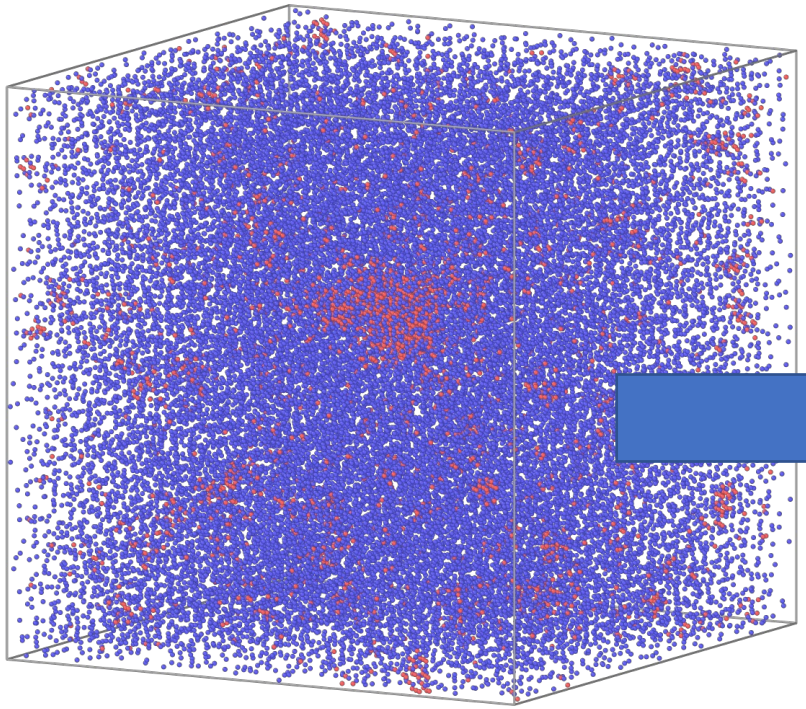


Slice through center of simulation

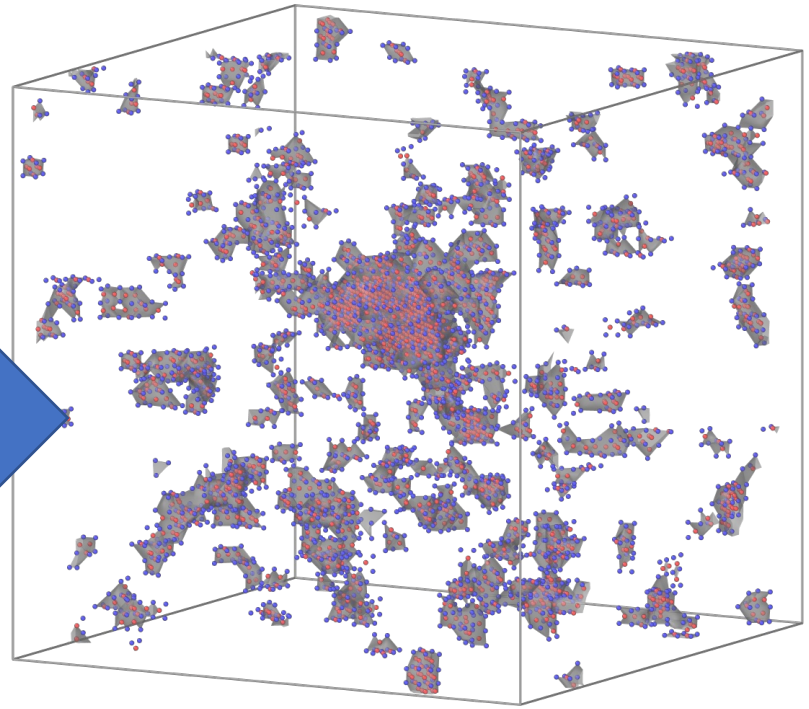


Identifying Precipitate Phase

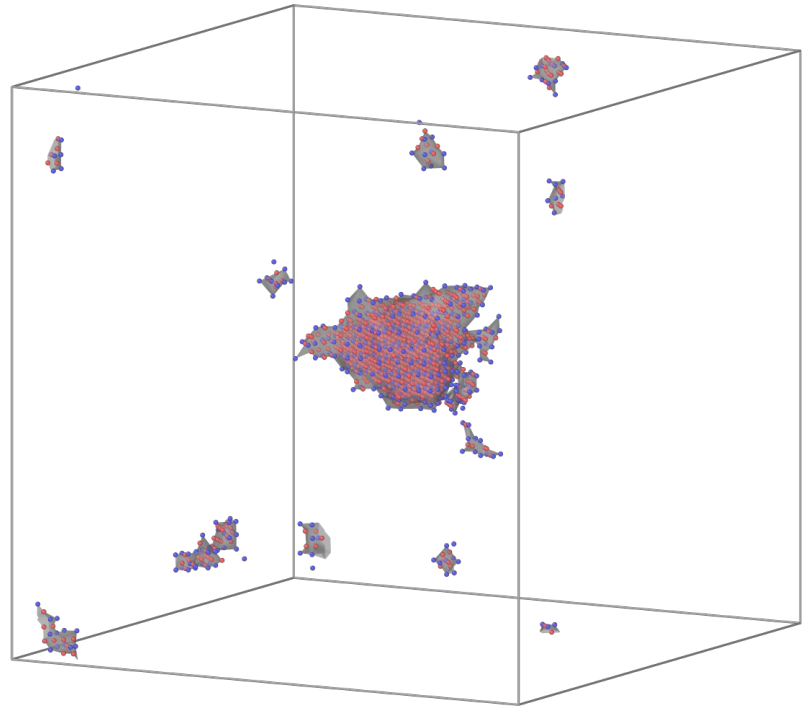
Polyhedral Template Matching (PTM) [Φ]



PTM + Clustering



New Algorithm – Neighbor Shell Concentration

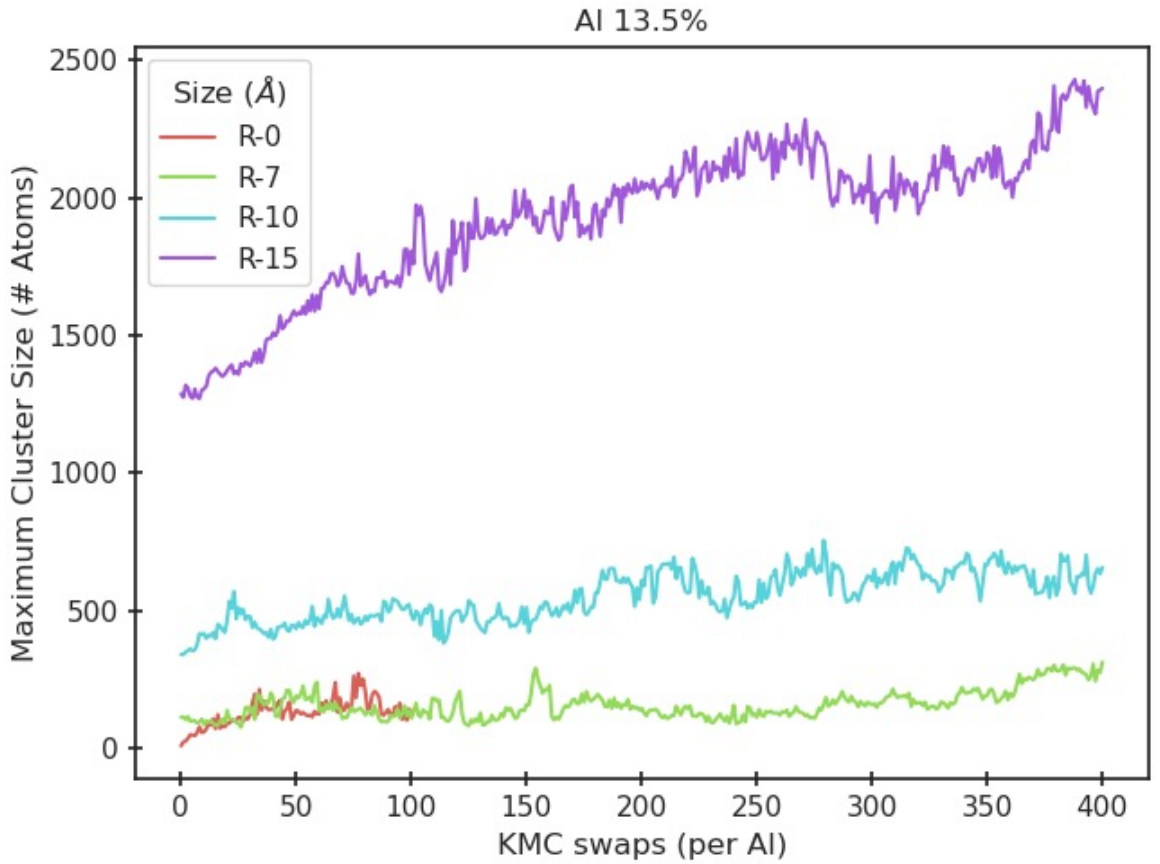


- All images are the same atomic structure
- PTM implemented in OVITO
- Existing tools cannot select for L_{12} ordered phase with precision

- Developed new algorithm for identifying L12 nuclei
- Based on local concentration of neighbor shells

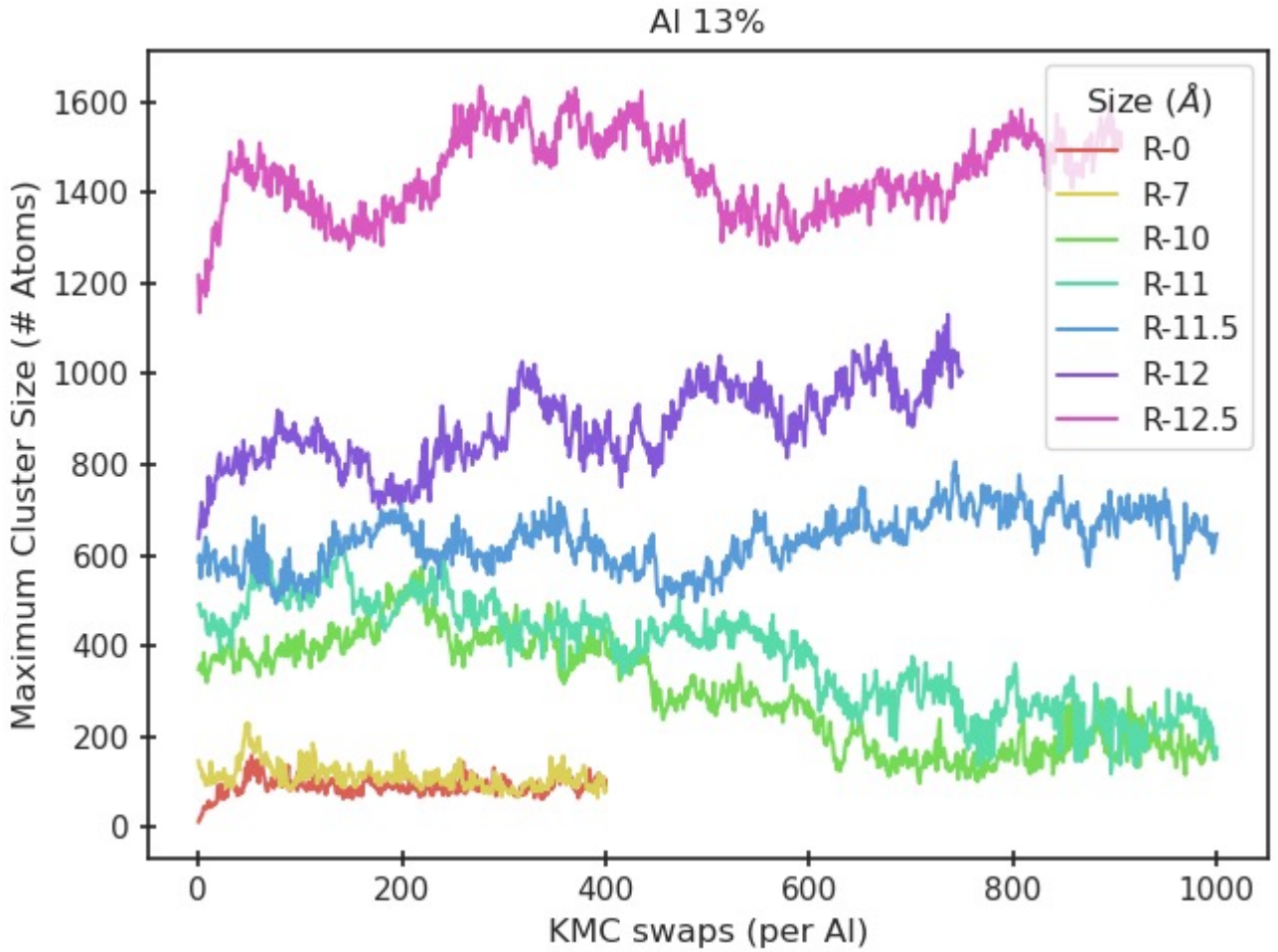
[Φ] Peter Mahler Larsen et. Al, 2016 Modelling Simul. Mater. Sci. Eng. 24 055007

13.5% Al Matrix Phase



- 7 Å initial precipitate no different growth than starting without initial nuclei
- Driving force for precipitation too large

13% Al Matrix Phase



- Constant driving force from source/sink allows for clearer picture of precipitate size change.
- Critical nucleus size between 11 and 12 Å initial precipitate radius

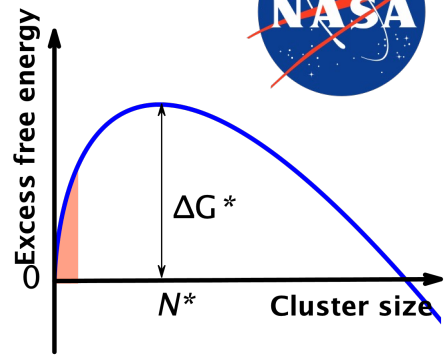
Precipitate Comparison

CALPHAD

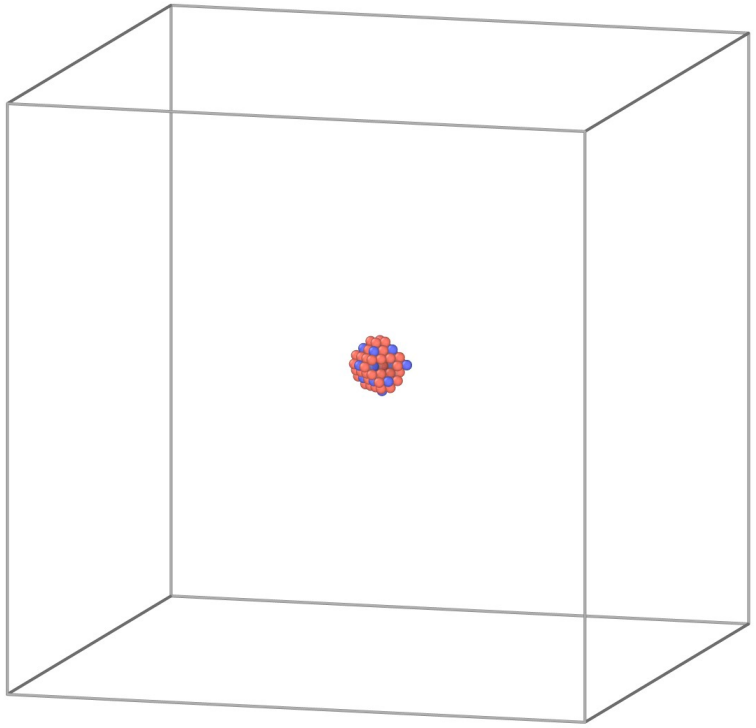
kMC-MD Simulations

$$\frac{\Delta G * R^*}{2} = \sigma$$

Interfacial Energy

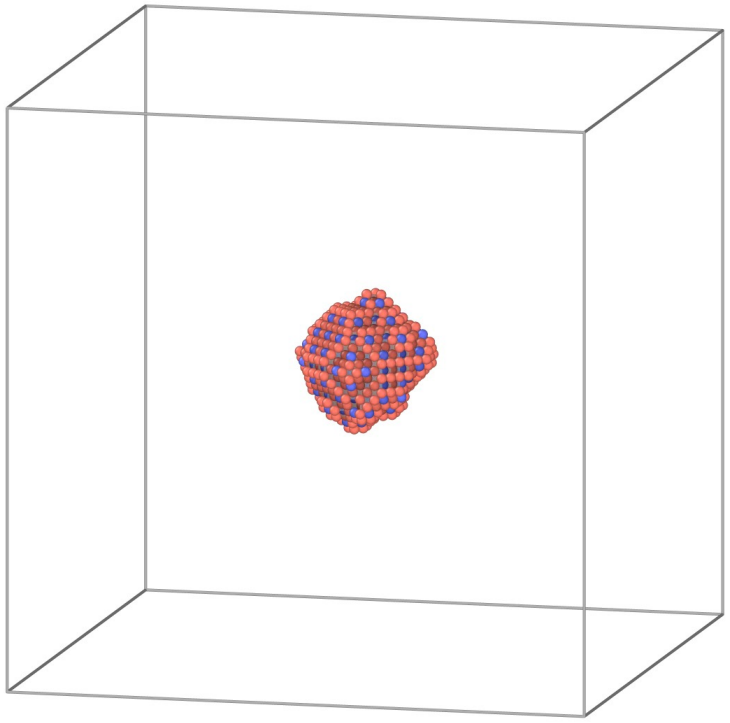


7 Å



Smaller Precipitate Shrinks

15 Å



Larger Precipitate Grows

Only Viewing Precipitate Atoms

Interfacial energy at 1000K

Experiment:

18.2 ± 3.5 mJ/m^2

Simulation:

17.4 mJ/m^2

Conclusions

- Hybrid kinetic Monte Carlo simulation approach enables simulation of substitutional solute diffusion through Molecular Dynamics

- New kMC-MD simulations allow for simulation of previously untapped atomistic behaviors such as solid-solid phase transformations

- Identified γ- γ' interfacial energy of 17.4 mJ/m^2 within 5% of experimental value (18.2 mJ/m^2) at 1000K

- Improved over previous computational method for determining γ- γ' interfacial energy using capillary fluctuations at 12.9mJ/m^2 @1000K $^{\Phi}$

$^{\Phi}$ Y. Mishi, 2014 Modelling Simul. Mater. Sci. Eng. 24. 4

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