Determination of Interfacial Energy for Solid-State Precipitation

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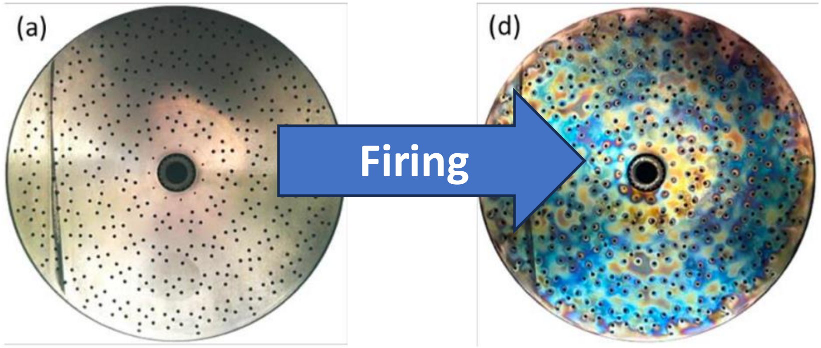
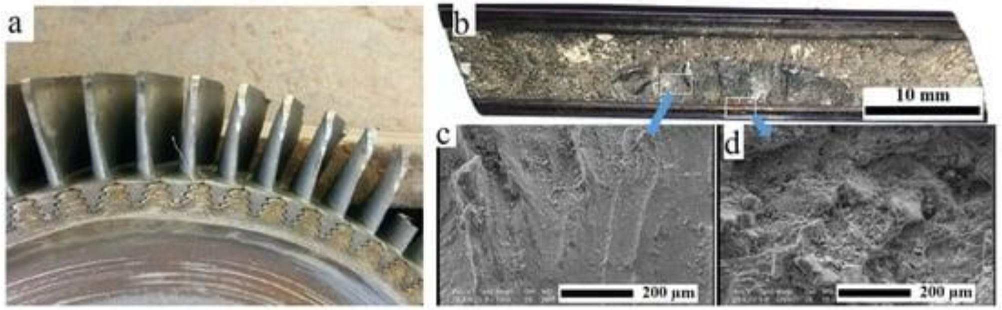
Solid-State Phase Precipitation

- Phase formation of both beneficial and detrimental phases is key for understanding performance of advanced materials
- Beneficial phases improve material capabilities
- Detrimental phases drastically shorten material lifespan
- Research Examples:
  - Improve corrosion resistance through better understanding underlying impact of alloy composition on formation of detrimental phases
  - Tune composition through developing relationship of stress, time, and temperature on detrimental phase formation



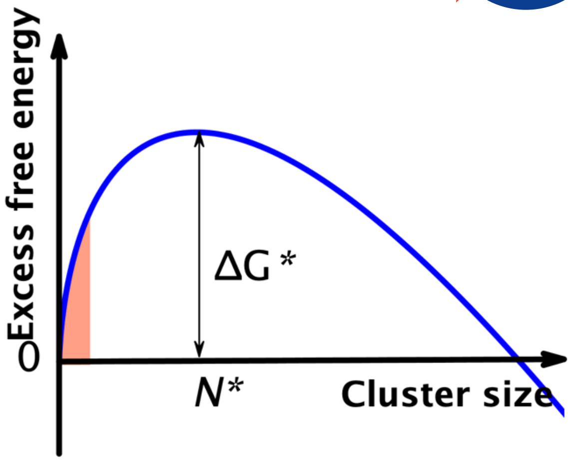
Gradl et.al (2024)



Mousavinia, et.al., *Eng. Fail. Anal.* **2020**, *115*, 104675.

Phase Formation in Solid-State Transformation is Critical for Material Strengthening and Durability

Solid-State Phase Precipitation

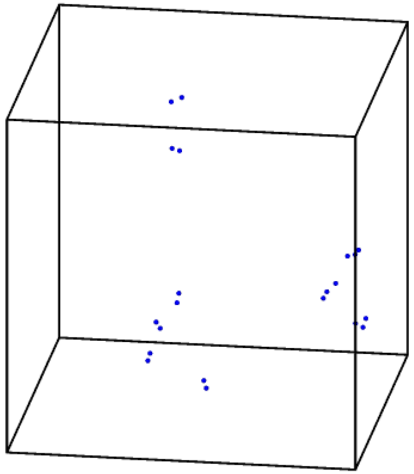


- Precipitation of new phases relies on solid-state diffusion to distribute chemical species
- Majority of time spent in sampling small cluster sizes
- Solved for Liquid-Solid (Persistent Embryo Method)
- Precipitate grows once energy penalty for nucleation is overcome

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

- Calculation of interfacial energy through determining critical nucleus size

$$\frac{3}{2s}|\Delta\mu|\rho^{2/3}{N^*}^{1/3} = \sigma$$



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

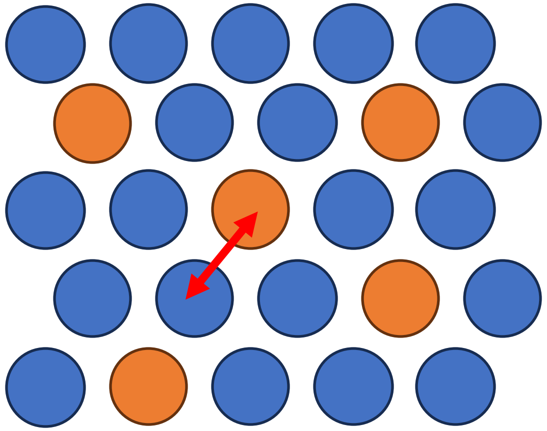
Solid State Diffusion – kMC in MD

Challenge:

- Diffusion processes inaccessible to Molecular Dynamics (MD) simulation due to short timescales
- New phase formation inaccessible to rigid lattice-based kinetic Monte Carlo (kMC) technique
- How can we combine kMC **diffusion** with MD-based **physics**?

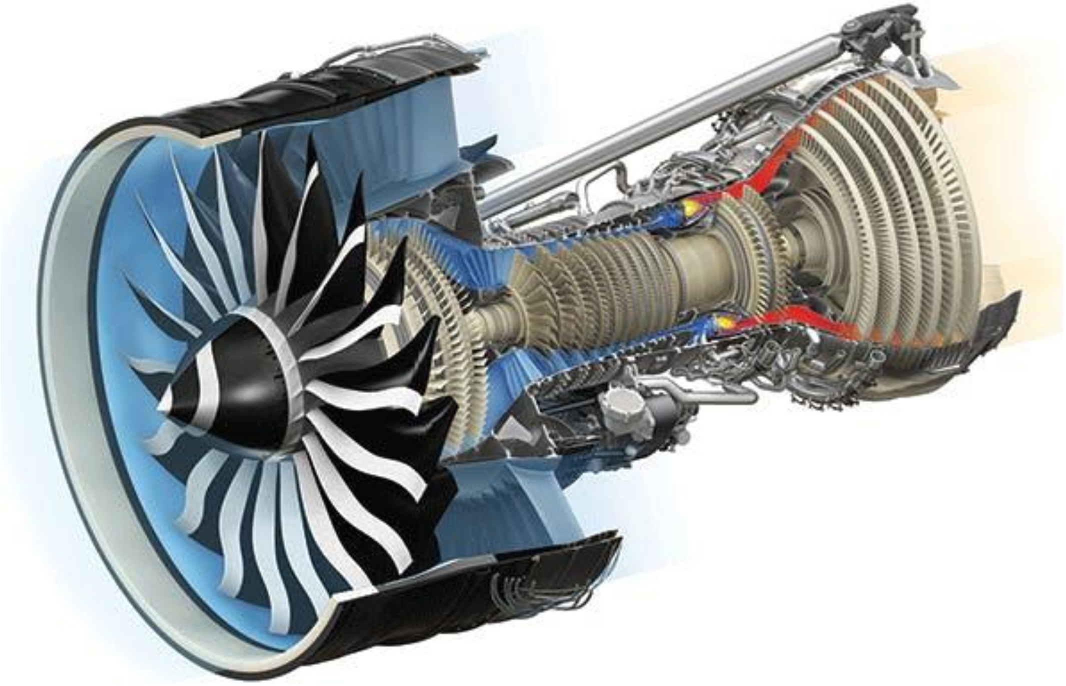
Solution:

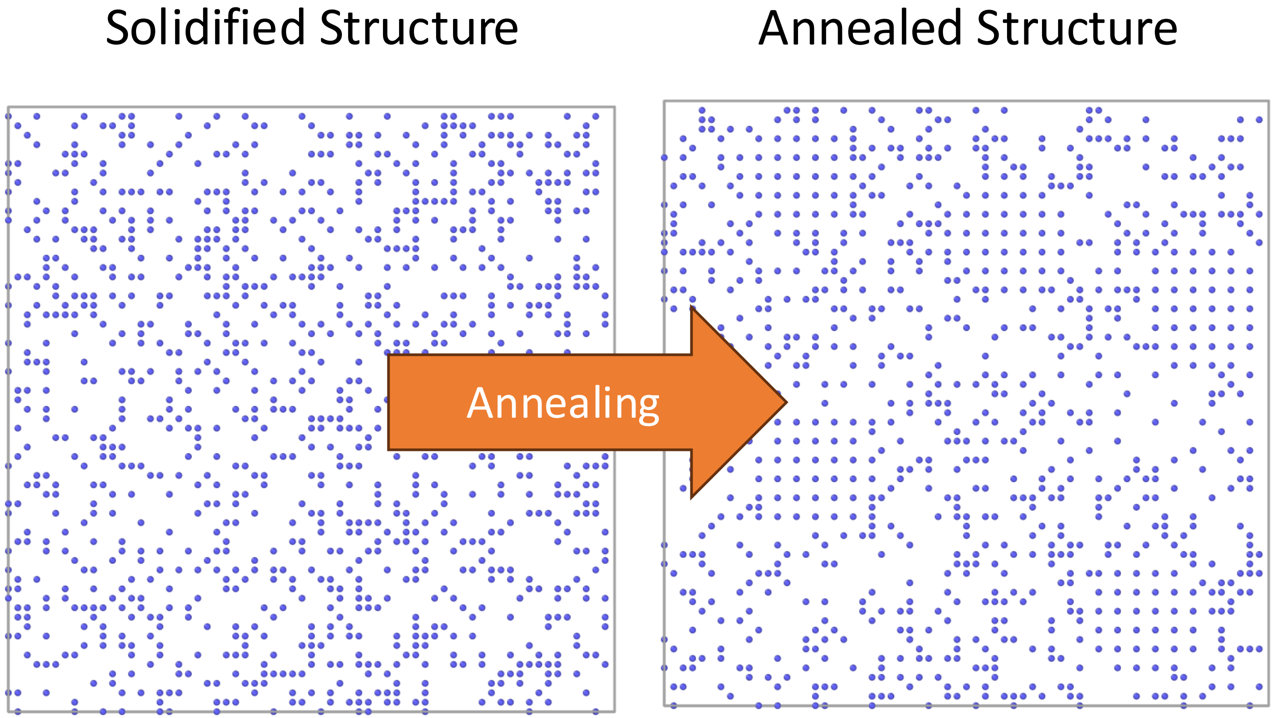
- Only swap nearest-neighbor atoms in kMC-MD LAMMPS simulation
- Select neighbors using Voronoi sampling technique



Case Study: Ni-based Superalloys

- γ' (Ni_3Al – $L1_2$) 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





Showing only Al Atoms

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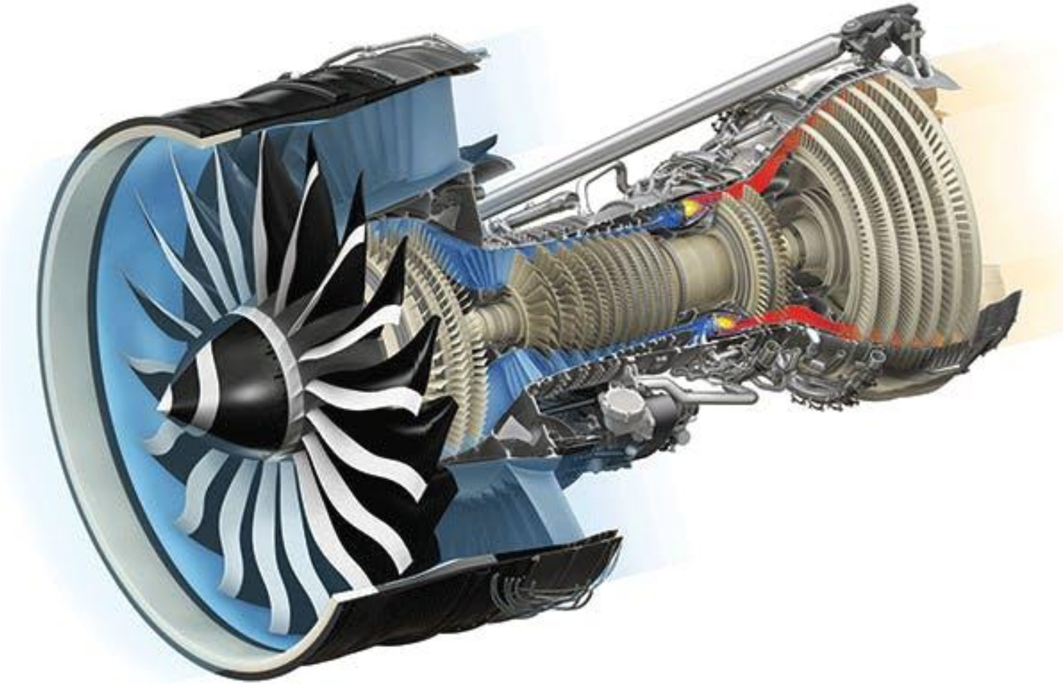
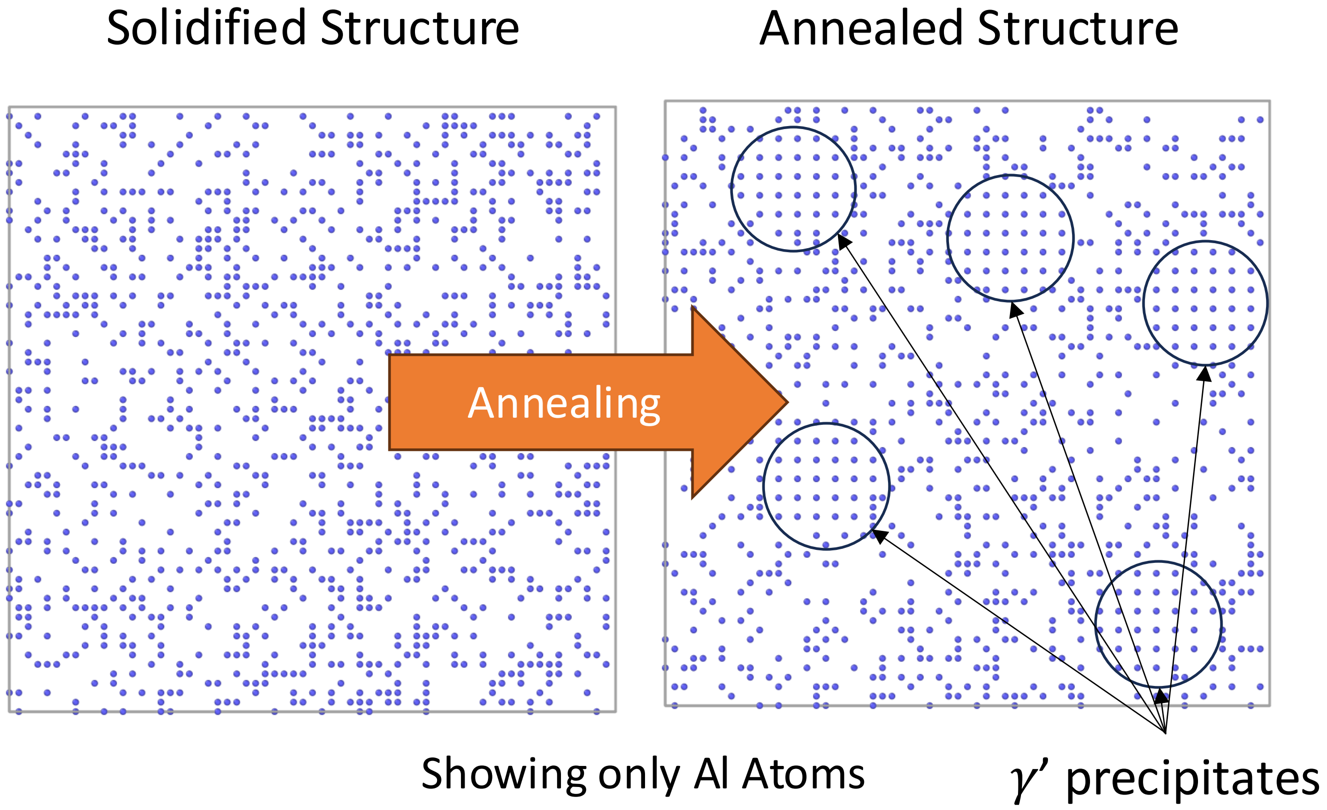




Solid State Diffusion – kMC in MD

Examples:

1) Test kMC-MD diffusion behavior by replicating ideal diffusion profiles
2) Show precipitate growth in Ni-based superalloy
3) Use kMC-MD simulation to calculate interfacial energy from critical nucleus

Example 1) – Verify accelerated diffusion behavior with kMC-MD

kMC-MD Solute Diffusion

Predicted ideal solute diffusion vs. kMC

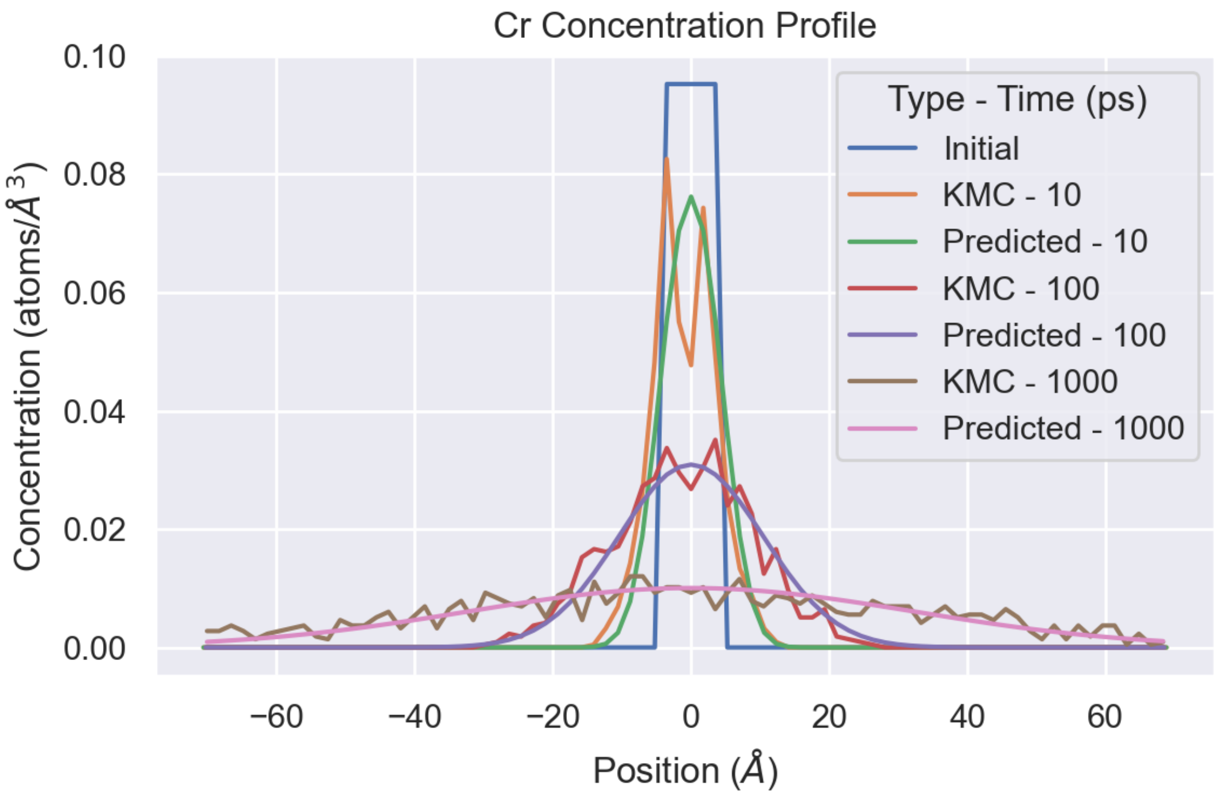
- Initialize fully-periodic cell of Ni with central concentration of Cr
- Simulation size: 3.5 x 3.5 x 14 nm
- 1000K MD/MC Temperature
- Swap rate of 1 atom/fs
- D = 0.001 Å^2/fs

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

w = Initial Pulse Width, D = Diffusivity

x = Position, t = Time

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



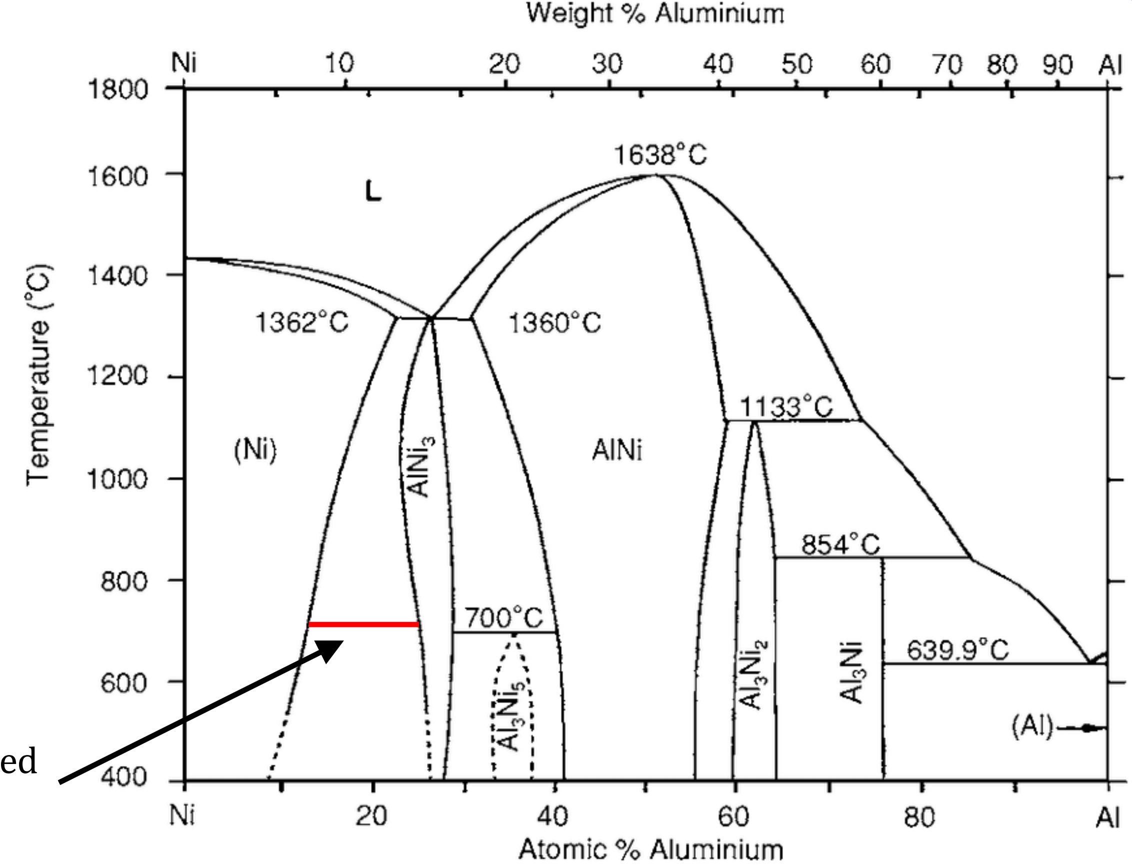
Good agreement between kMC-MD simulation and diffusion equation

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

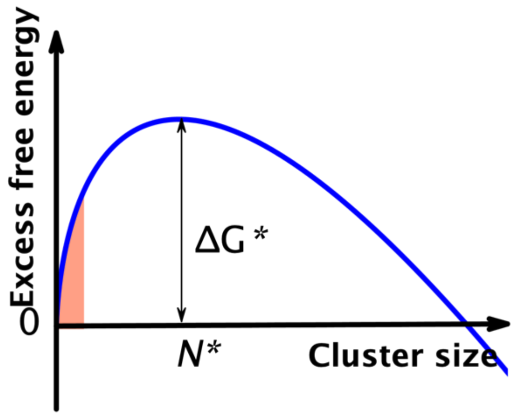
Example 2) – Demonstrate γ’ precipitate growth

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
- ❑ Verify 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



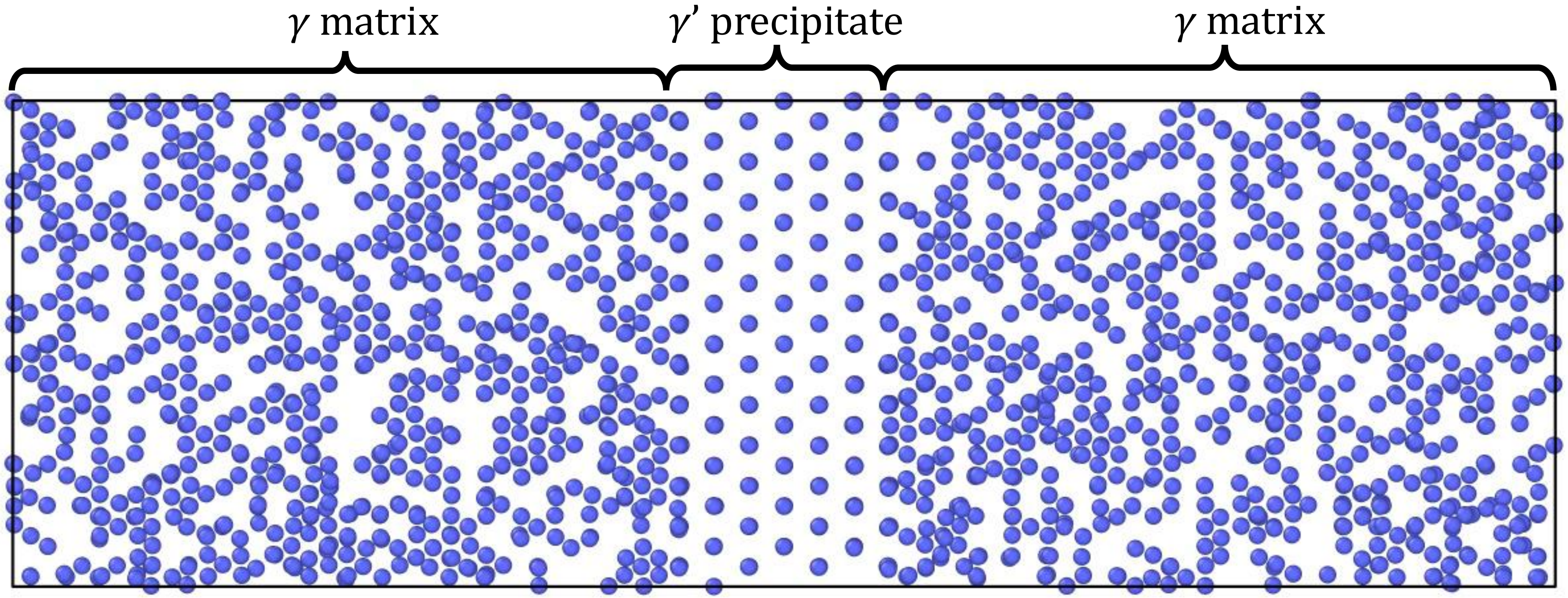
Investigated Condition



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).

kMC-MD γ-γ‘ Phase Growth



- 16% Al in Ni γ matrix
- 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).

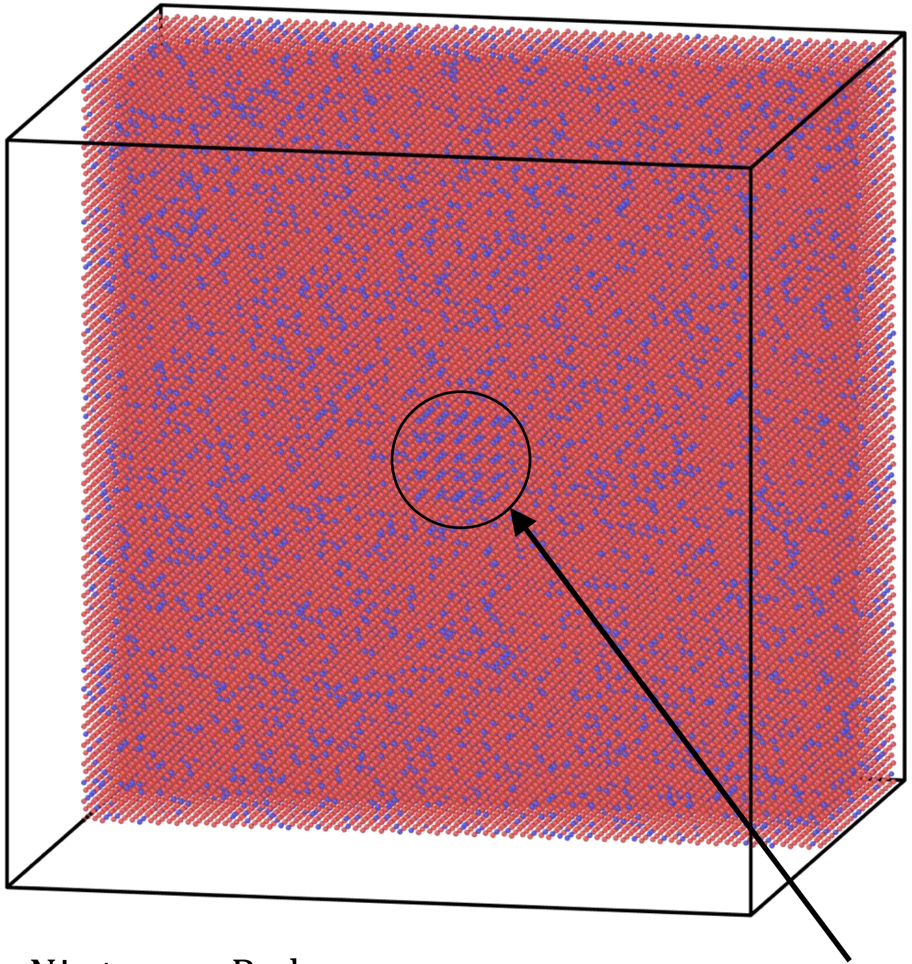
Example 3) – Determine critical nucleus size in kMC-MD simulation and calculate γ-γ’ interfacial energy

Precipitation: Simulation Setup

❑ **Need to identify critical nucleus size N***

- Ni-Al system
  - Initialize spherical γ' 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

Sliced through center of simulation cell



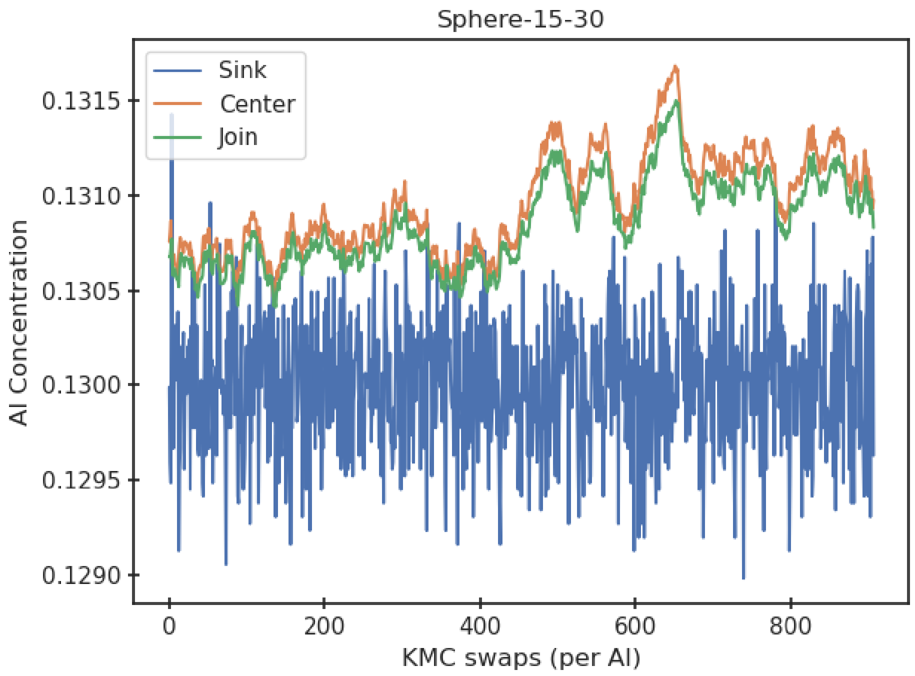
Ni atoms – Red
Al atoms – Blue

Initial Precipitate

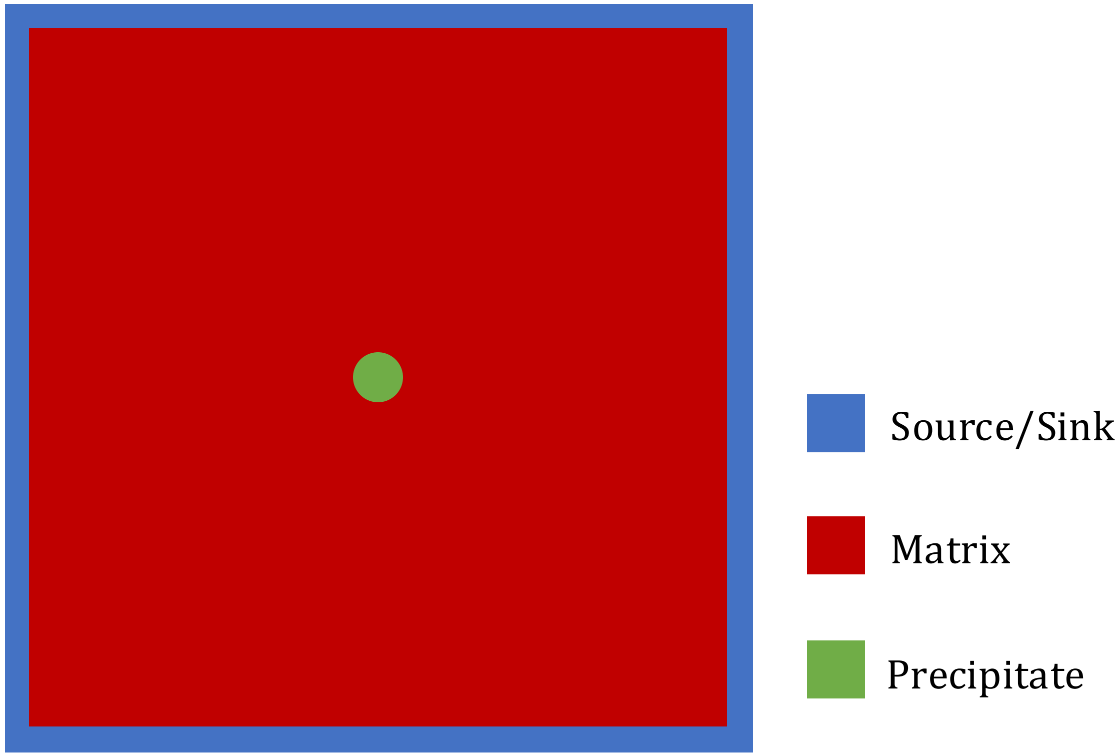
J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

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



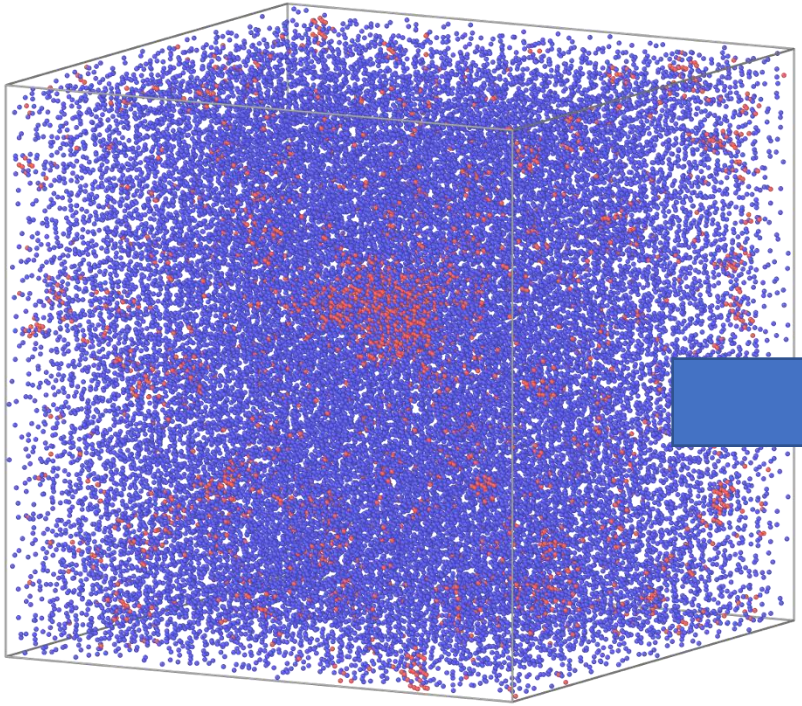
Slice through center of simulation



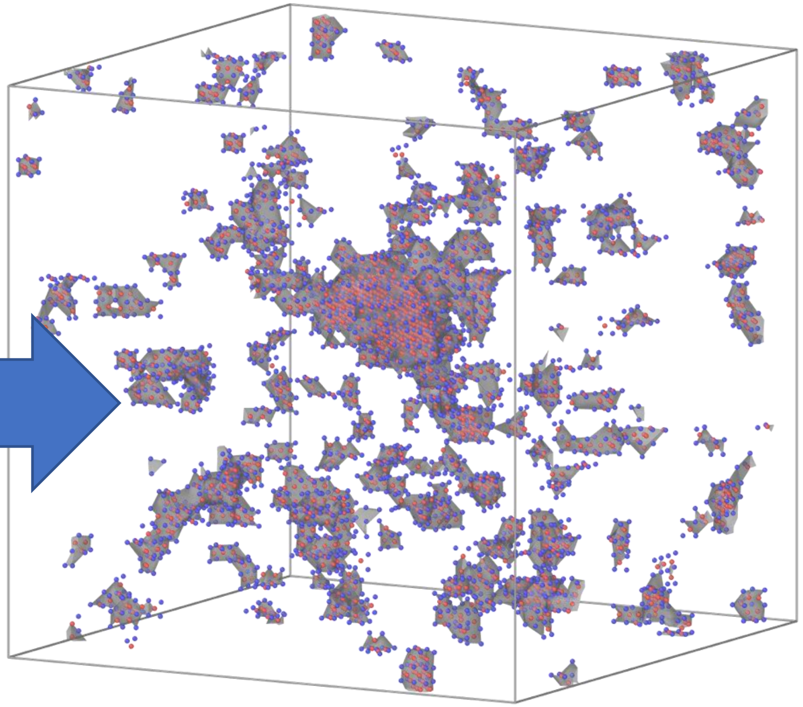
J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

Identifying Precipitate Phase

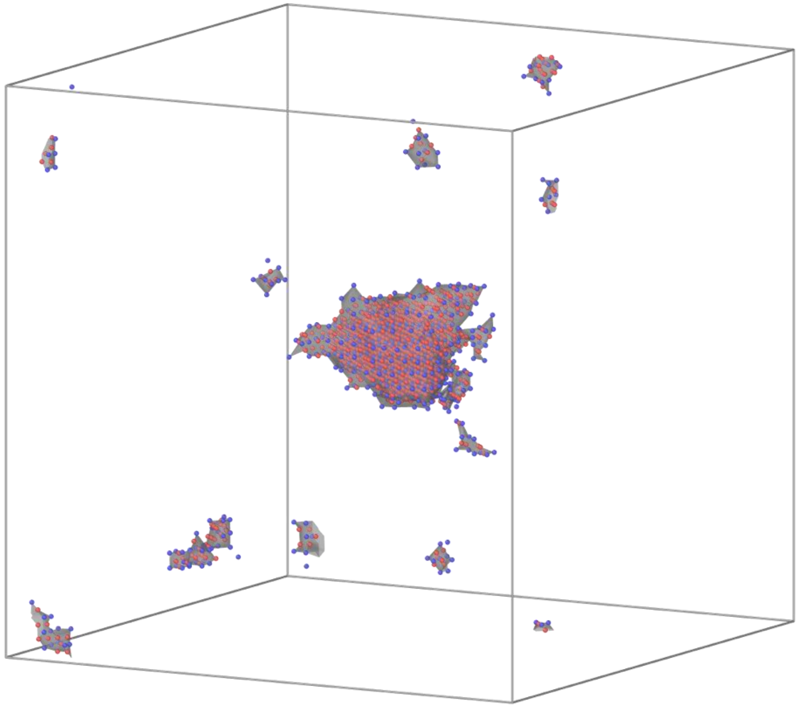
Polyhedral Template Matching (PTM) [Φ]



PTM + Clustering



New Algorithm – Neighbor Shell Concentration



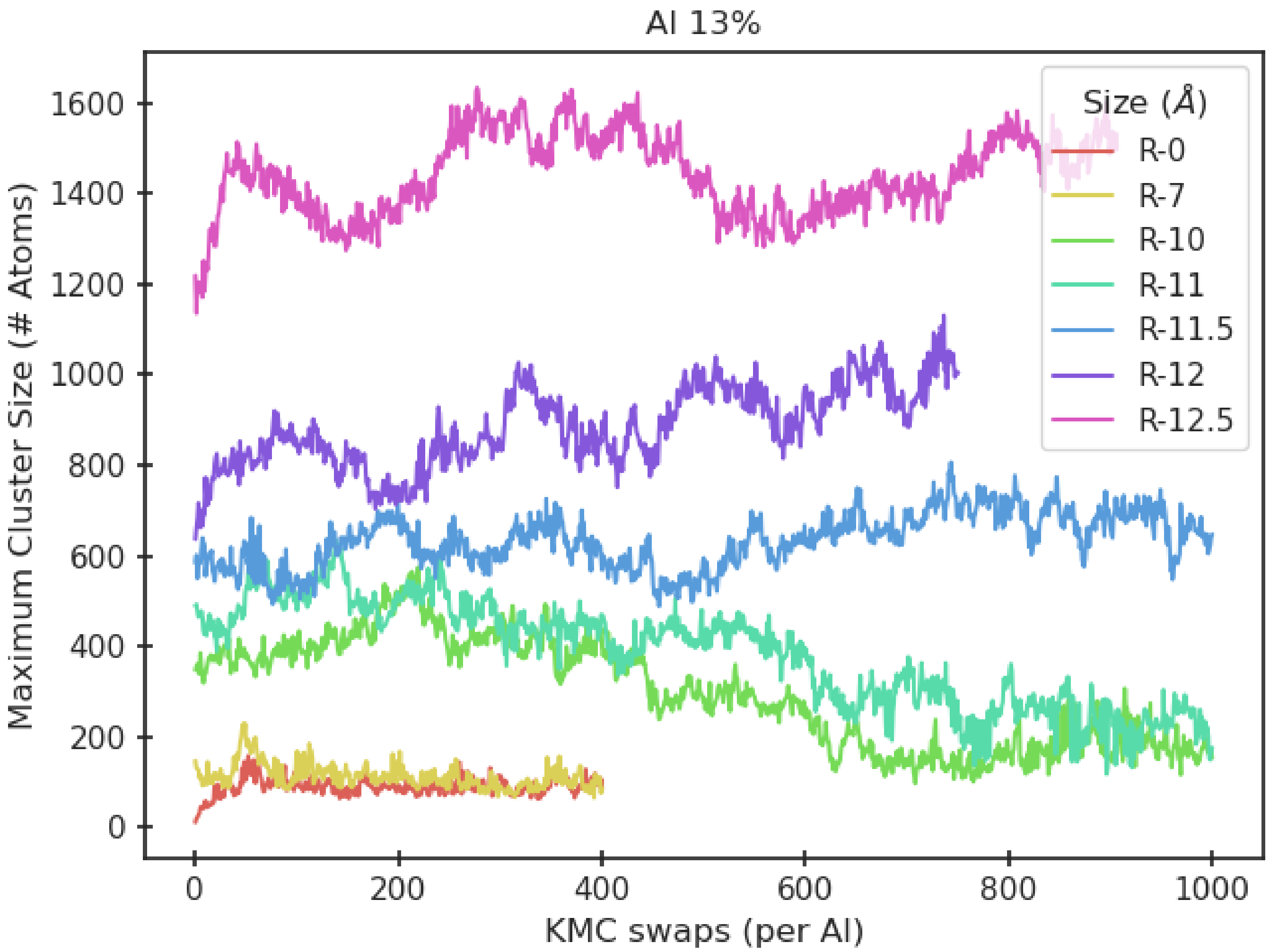
- All three images are the same atomic structure
- PTM implemented in OVITO
- Existing tools cannot select for L_{12} ordered phase with precision

- 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

J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

Determining Critical Nucleus



- 13% Al γ Matrix phase
- 1000K
- Critical nucleus size between 11 and 12 Å initial precipitate radius

J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

Precipitate Comparison

CALPHAD

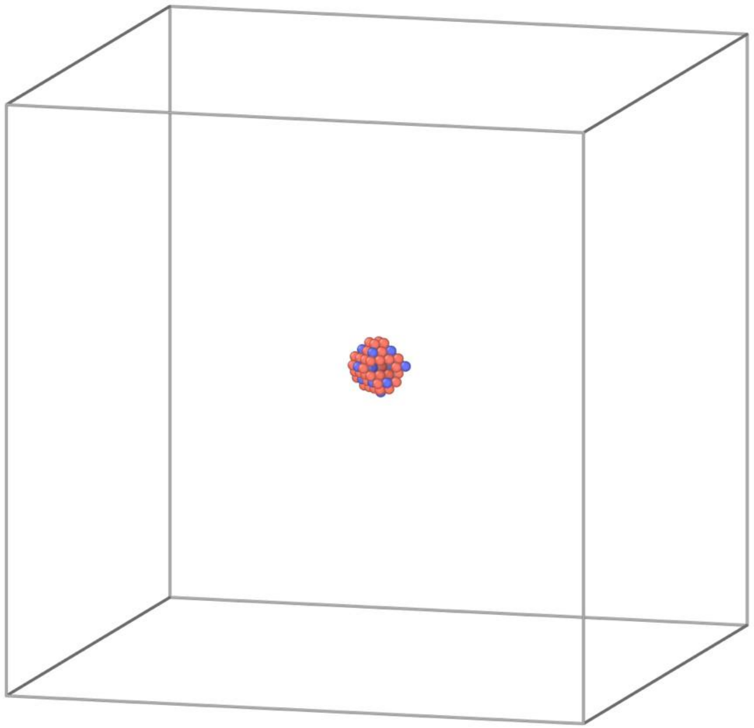
kMC-MD Simulations

$$\frac{3}{2s}|\Delta\mu|\rho^{2/3}N^{*1/3} = \sigma$$

Interfacial Energy

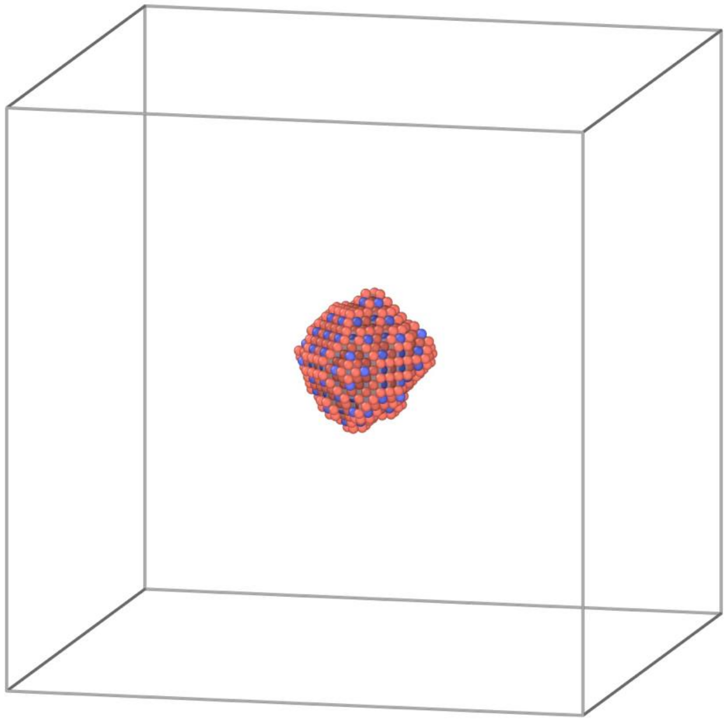
Only Viewing Precipitate Atoms

7 Å

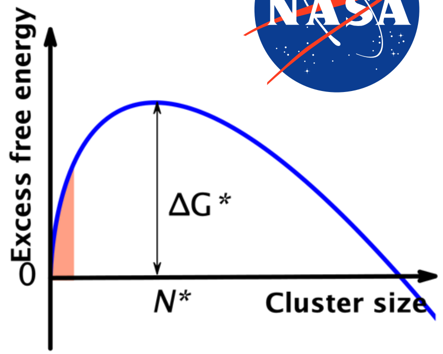


Smaller Precipitate Shrinks

15 Å



Larger Precipitate Grows



Interfacial energy at 1000K

Experiment:

18.2 ± 3.5 mJ/m²

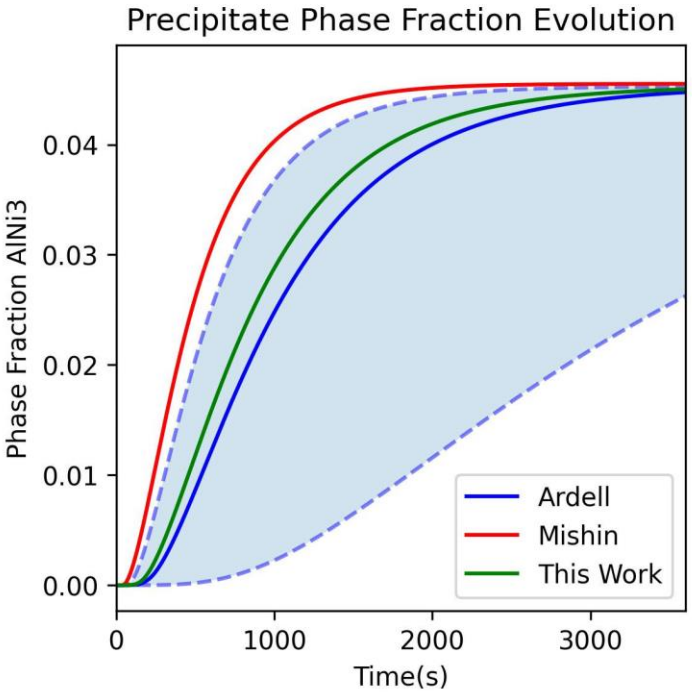
Simulation:

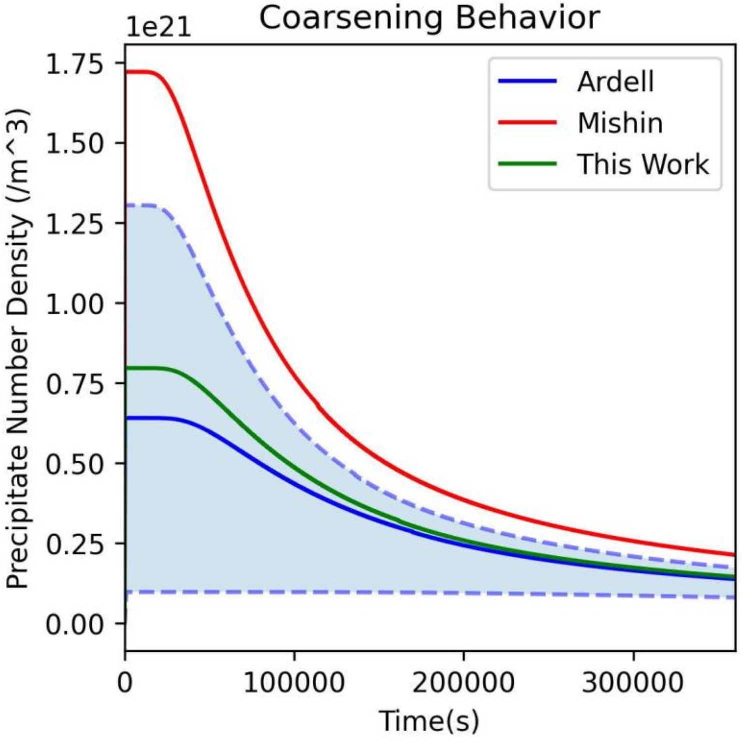
17.4 mJ/m²

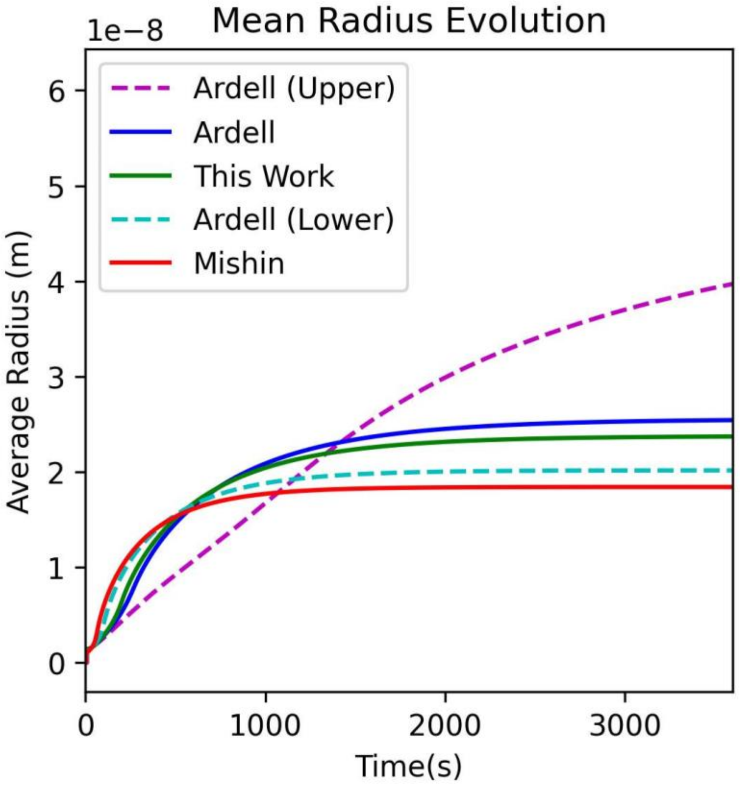
J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

Continuum-Scale Precipitate Predictions

- Once interfacial energy has been determined, experimentally-relevant behaviors can be directly simulated with continuum-scale methods
- Precipitation nucleation and growth simulation in KAWIN demonstrates macroscale kinetic effects of precipitation


Precipitate Phase Fraction Evolution


Coarsening Behavior


Mean Radius Evolution

J. Tavenner et. al, *J. Chem. Phys.* 161, 041102 (2024).

Conclusions

- Our hybrid kMC-MD approach:
  - Enables simulation of substitutional solute diffusion through physics-based Molecular Dynamics framework
  - Allows for simulation of previously untapped atomistic behaviors such as solid-solid phase transformations

- Our MD simulation of precipitation:
  - Improved over previous capillary fluctuation method for determining γ- γ' interfacial energy: 12.9mJ/m^2 @1000K $^{\Phi}$

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

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

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