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ALLOY EXPRESS:

A General and Scalable parallel Hybrid Monte Carlo and Molecular Dynamics Algorithm for Alloy Simulations

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Sandia National Laboratories

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LAMMPS Workshop and Symposium 2025

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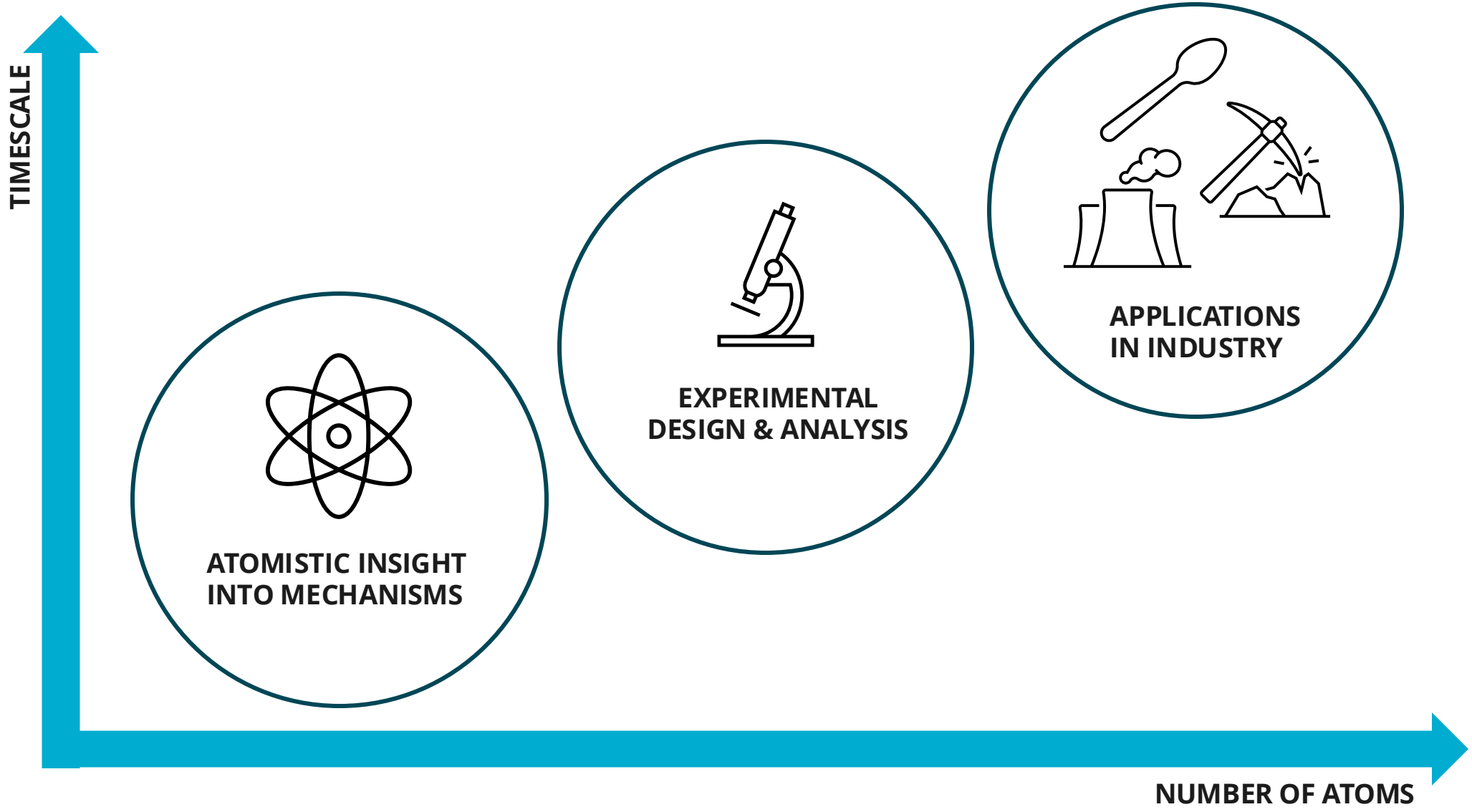
UNDERSTANDING ALLOYS REQUIRES ATOMISTIC MODELS



TRADITIONAL APPROACH: HYBRID MONTE CARLO (MC)/MOLECULAR DYNAMICS (MD)

EXPLORE VARIOUS TRAJECTORIES

RAPIDLY ALTER ALLOY CHEMISTRY

Molecular Dynamics

Numerically solve Newton's equations of motion

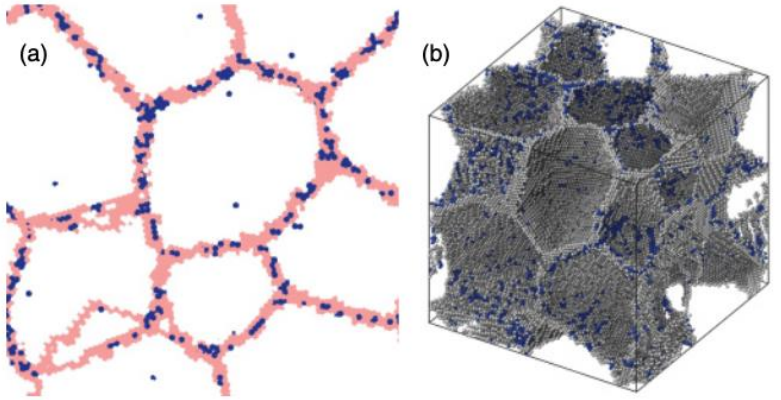
$$F = -\nabla V(\vec{r})$$

Monte Carlo

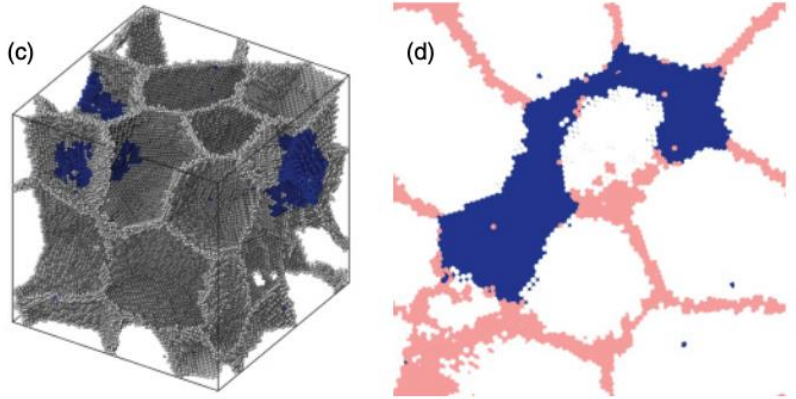
Randomly change atom species with respect to an underlying distribution

LAMMPS

Grain boundary network[1], Fe Cu-5 at.%



Chemical segregation



Phase precipitation

1. Sadigh et al., Phys Rev. B **85**.18 (2012)

TRADITIONAL ENSEMBLE CHOICES FOR MC

	Canonical	Semi-grand canonical	Variance-constrained[1] semi-grand canonical
Usage	Known fixed composition	Changing composition (infinite reservoir per species)	Changing composition, governed by chemical potential difference *dμ*, fixes ensemble-averaged squared concentration
Limitations	Extremely slow evolution in miscibility gaps	Stabilizing a phase requires known chemical potential difference (*dμ),* limited to single stable phase	Performance sensitive to choice of *dμ* Only usable with Embedded Atom Method potentials*
LAMMPS command	`fix atom/swap`	`fix atom/swap semi-grand = yes`	`fix sgcmc`

GOAL: find lowest-energy chemical configuration(s) and/or phase precipitations

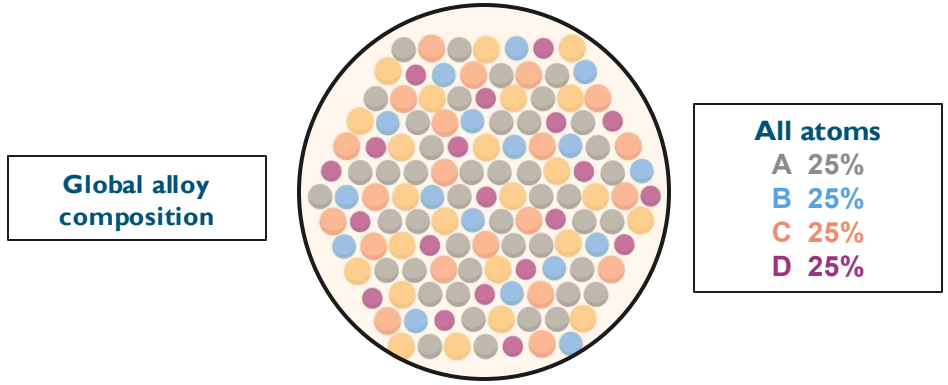
1. Sadigh, Babak, et al. "Salable parallel Monte Carlo algorithm for atomistic simulations of precipitation in alloys." *Physical Review B—Condensed Matter and Materials Physics* 85.18 (2012): 184203.

TRADITIONAL MC SWAPS RELY ON GLOBAL ENERGY: PROHIBITS PARALLELISM

Global Algorithm: Traditional MC

Input: N atoms, S species, E model for atomic interactions

1	$N[\text{rand}] = N_i$	(select random atom N_i)
2	$S[N_i] = s'_i \quad s_i \mathrel{!}= s'_i$	(select **new** random species for N_i)
3	compute $E(N, S) - E(N, S')$	(**GLOBAL** change in energy of swap)
6	**if** **then**	(based on MC ensemble)
7	$S \rightarrow S'$	(accept proposed swap)
8	**else**	
9	$S \rightarrow S$	(reject proposed swap)
10	**end**	



Sadigh et al.[1] Algorithm: parallel MC for **EAM potentials**

Input: N atoms, S species, E model for atomic interactions

1	$N[\text{rand}] = N_i$	(select random atom N_i)
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3	compute $E_i(N_i, S_i) - E_i(N_i, S'_i)$	(**LOCAL** change in **EAM** energy of swap)
6	**if** **then**	(based on MC ensemble)
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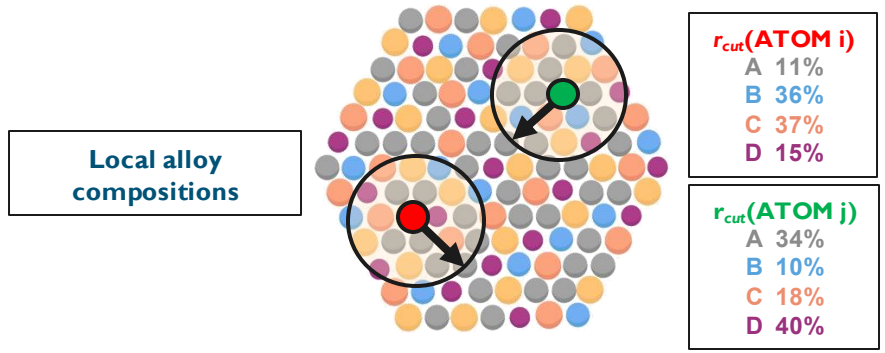
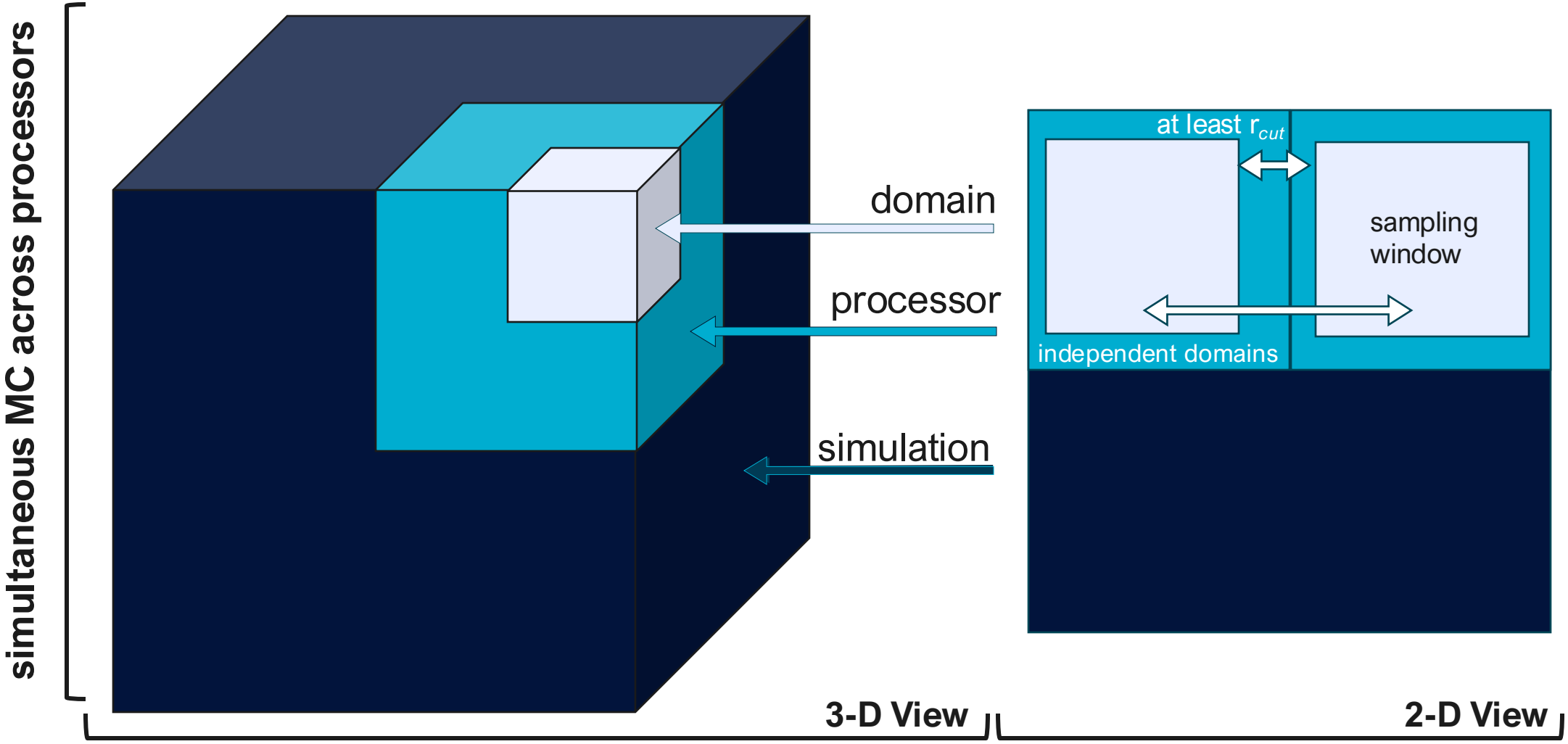


1. Sadigh, Babak, et al. "Scalable parallel Monte Carlo algorithm for atomistic simulations of precipitation in alloys." Physical Review B—Condensed Matter and Materials Physics 85.18 (2012): 184203.

DOMAIN DECOMPOSITION ALLOWS PARALLEL MC SWAPS



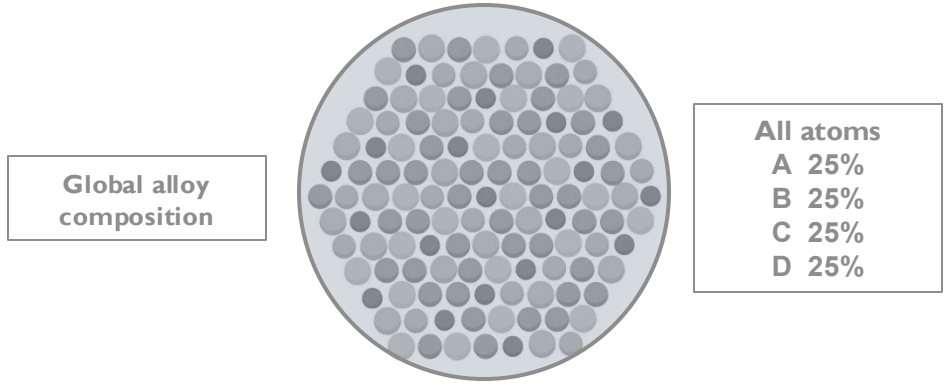
Sadigh, Babak, et al. "Scalable parallel Monte Carlo algorithm for atomistic simulations of precipitation in alloys." *Physical Review B—Condensed Matter and Materials Physics* 85.18 (2012): 184203.

SCALABLE, PARALLEL ALGORITHM FOR HYBRID MC/MD WITH EAM ONLY

Global Algorithm: Traditional MC

Input: N atoms, S species, E model for atomic interactions

```
1  N[rand] = N_i                    (select random atom N_i)
2  S[N_i] = s'_i   s_i != s'_i      (select new random species for N_i)
3  compute E(N, S) - E(N, S')       (GLOBAL change in energy of swap)
6  if        then                   (based on MC ensemble)
7        S→S'                      (accept proposed swap)
8  else
9        S→S                       (reject proposed swap)
10 end
```



Sadigh et al.[1] Algorithm: parallel MC for **EAM potentials**

Input: N atoms, S species, E model for atomic interactions

```
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2  S[N_i] = s'_i   s_i != s'_i      (select new random species for N_i)
3  compute E_i(N_i, S_i) - E_i(N_i, S'_i)   (LOCAL change in EAM energy of swap)
6  if        then                   (based on MC ensemble)
7        S→S'                      (accept proposed swap)
8  else
9        S→S                       (reject proposed swap)
10 end
```

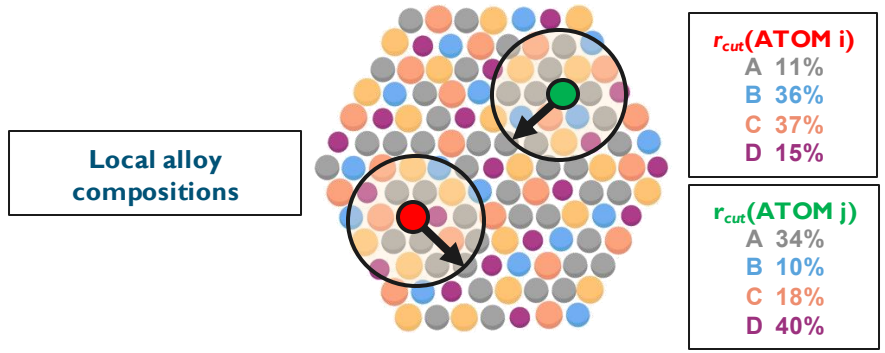

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WHAT IS AN INTERATOMIC POTENTIAL?

Interatomic Potential

$$F = -\nabla V(\vec{r})$$

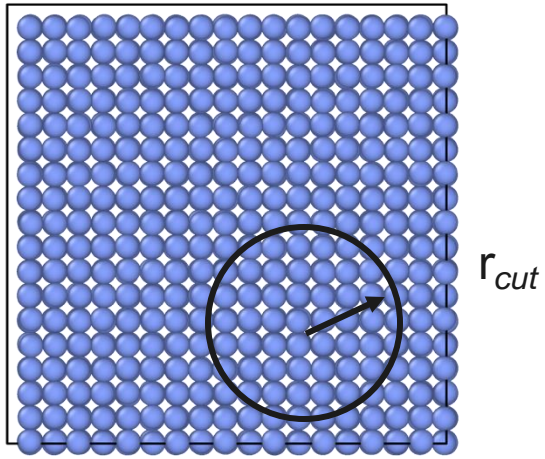
Goal: given a configuration of atoms, predict **energy, force, stress etc.**

EXAMPLE:

Embedded Atom Method (EAM)

$$E_i = F_\alpha\left(\sum_{j\neq i}\rho_\beta(r_{ij})\right) + \frac{1}{2}\sum_{j\neq i}\phi_{\alpha\beta}(r_{ij})$$

embedding energy **pair potential**



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GENERAL, SCALABLE ALGORITHM FOR HYBRID MC/MD

Sadigh et al.[1] Algorithm: parallel MC for **EAM potentials**

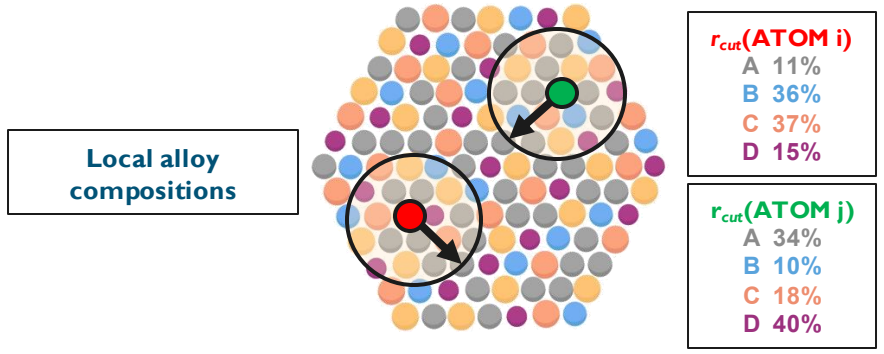
Input: N atoms, S species, E model for atomic interactions

1	$N[\text{rand}] = N_i$	(select random atom N_i)
2	$S[N_i] = s'_i \quad s_i \mathrel{!}= s'_i$	(select **new** random species for N_i)
3	compute $E_i(N_i, S_i) - E_i(N_i, S'_i)$	(**LOCAL** change in **EAM** energy of swap)
6	**if** **then**	(based on MC ensemble)
7	$S \rightarrow S'$	(accept proposed swap)
8	**else**	
9	$S \rightarrow S$	(reject proposed swap)
10	**end**	

General, Local Algorithm: parallel MC for **any potentials**

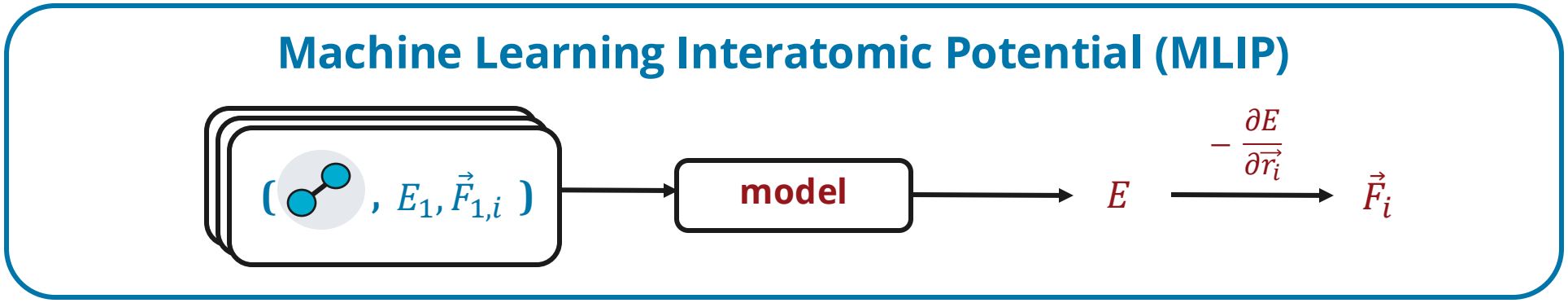
Input: N atoms, S species, E model for atomic interactions

1	$N[\text{rand}] = N_i$	(select random atom N_i)
2	$S[N_i] = s'_i \quad s_i \mathrel{!}= s'_i$	(select **new** random species for N_i)
3	compute $E_i(N_i, S_i) - E_i(N_i, S'_i)$	(**LOCAL** change in energy of swap)
6	**if** **then**	(based on MC ensemble)
7	$S \rightarrow S'$	(accept proposed swap)
8	**else**	
9	$S \rightarrow S$	(reject proposed swap)
10	**end**	



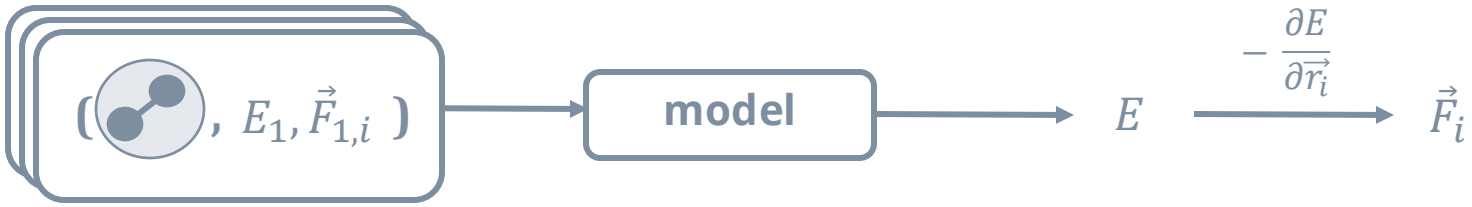
1. Sadigh, Babak, et al. "Scalable parallel Monte Carlo algorithm for atomistic simulations of precipitation in alloys." Physical Review B—Condensed Matter and Materials Physics 85.18 (2012): 184203.

WHAT IS A MACHINE LEARNING INTERATOMIC POTENTIAL (MLIP)?

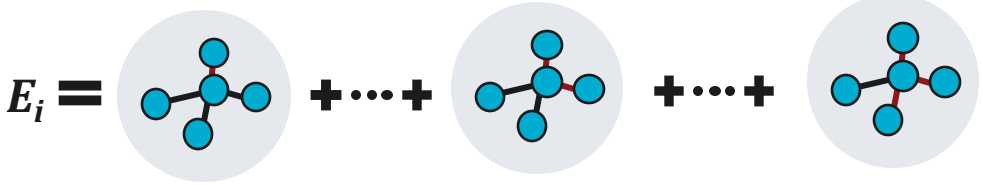


EXTENDED SUPPORT FOR HYBRID MC/MD WITH MLIPS IN LAMMPS

Machine Learning Interatomic Potential (MLIP)



Atomic Cluster Expansion (ACE)



Basis: product of radial functions and spherical harmonics

Drautz, Ralf. "Atomic cluster expansion for accurate and transferable interatomic potentials." *Physical Review B* 99.1 (2019): 014104.

Spectral Neighbor Analysis Potential (SNAP)

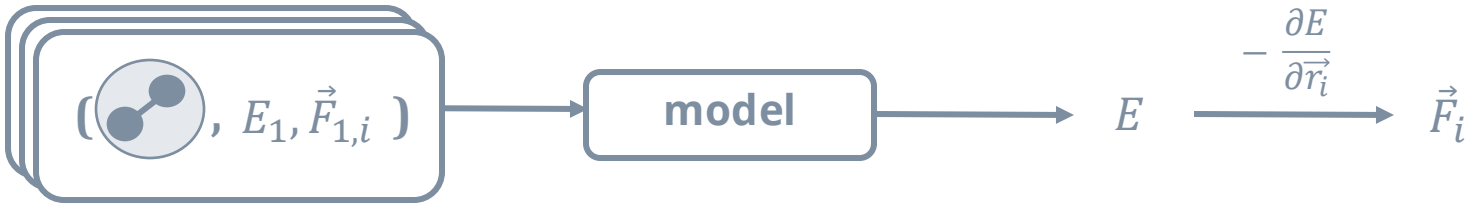
$$E^i_{SNAP}\left(B^i_1, \ldots, B^i_K\right) = \beta^{\mu_i}_0 + \sum_{k=1}^{K} \beta^{\mu_i}_k B^i_k$$

Characterize local environments with **bispectrum components**

Thompson, Aidan P., et al. "Spectral neighbor analysis method for automated generation of quantum-accurate interatomic potentials." *Journal of Computational Physics* 285 (2015): 316-330.

EXTENDED SUPPORT FOR HYBRID MC/MD WITH MLIPS IN LAMMPS

Machine Learning Interatomic Potential (MLIP)



Fast Learning of Atomistic Rare Events (FLARE)

Bayesian inference and active learning with Gaussian process regression

FLARE

Vandermause, Jonathan, et al. *npj Computational Materials* 6.1 (2020): 20. ;Xie, Yu, et al. *npj Computational Materials* 9.1 (2023): 36.

GENERAL, SCALABLE HYBRID MC/MD IN LAMMPS WITH MLIPS

General, Local Algorithm: parallel MC for **any potential**

Input: N atoms, S species, E model for atomic interactions

1	$N[\text{rand}] = N_i$	(select random atom N_i)
2	$S[N_i] = s'_i \quad s_i \mathrel{!}= s'_i$	(select **new** random species for N_i)
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Atomic Cluster Expansion (ACE)

$E_i =$  + ··· + + ··· +

Basis: product of radial functions and spherical harmonics

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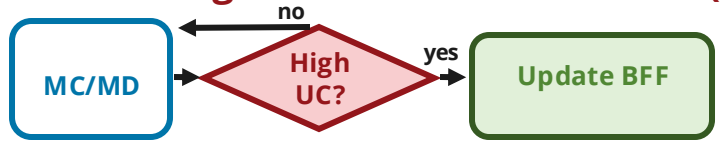
$$E^i_{SNAP}\left(B^i_1, \ldots, B^i_K\right) = \beta^{\mu_i}_0 + \sum_{k=1}^{K} \beta^{\mu_i}_k B^i_k$$

Characterize local environments with **bispectrum components**

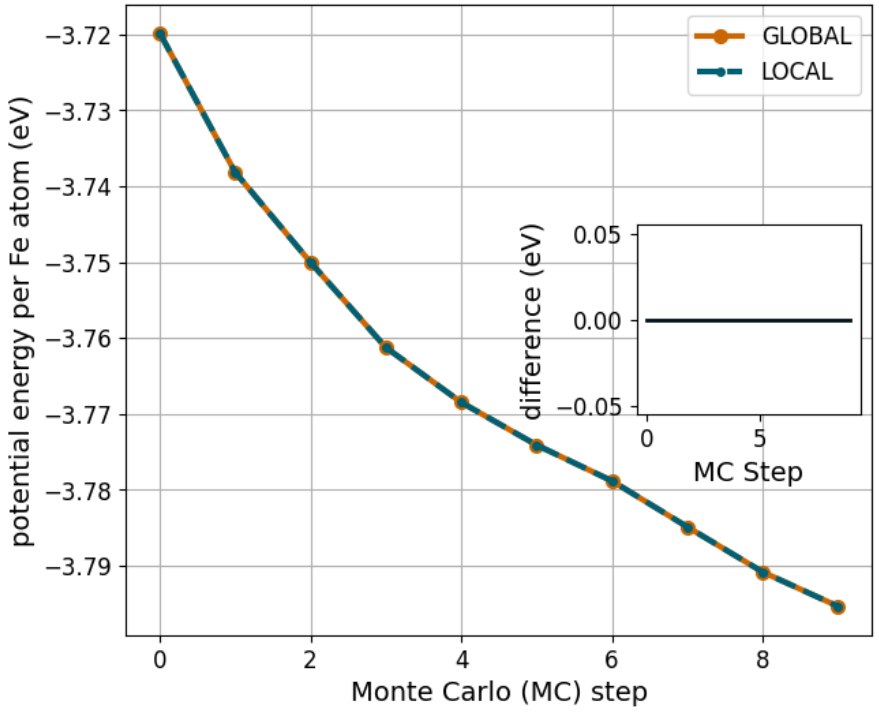
Fast Learning of Atomistic Rare Events (FLARE)



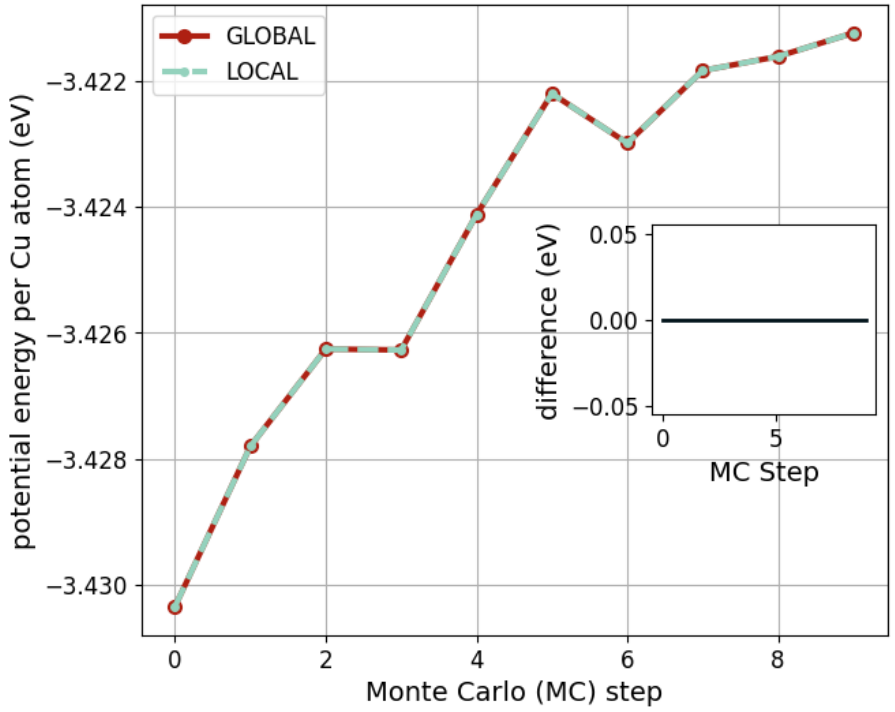
Bayesian inference and active learning with GP regression

LOCAL METHOD EXACTLY REPRODUCES POTENTIAL ENERGY DURING ALLOY SIMULATIONS

Fe atoms, PE

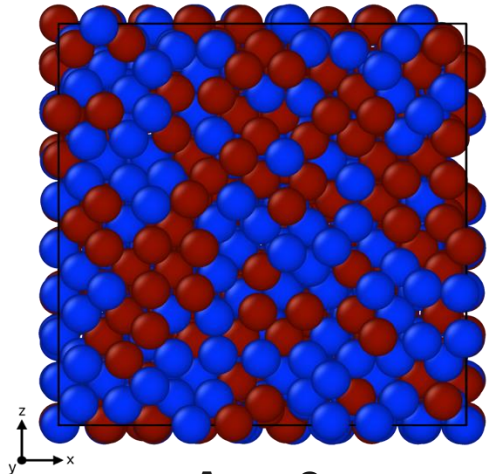


Cu atoms, PE

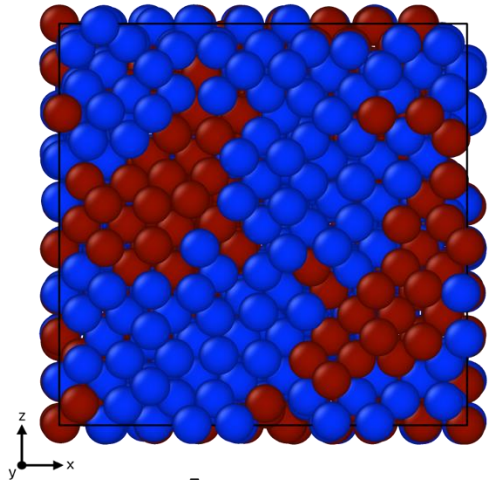


EAM POTENTIAL

FeCu (50/50 at.%)

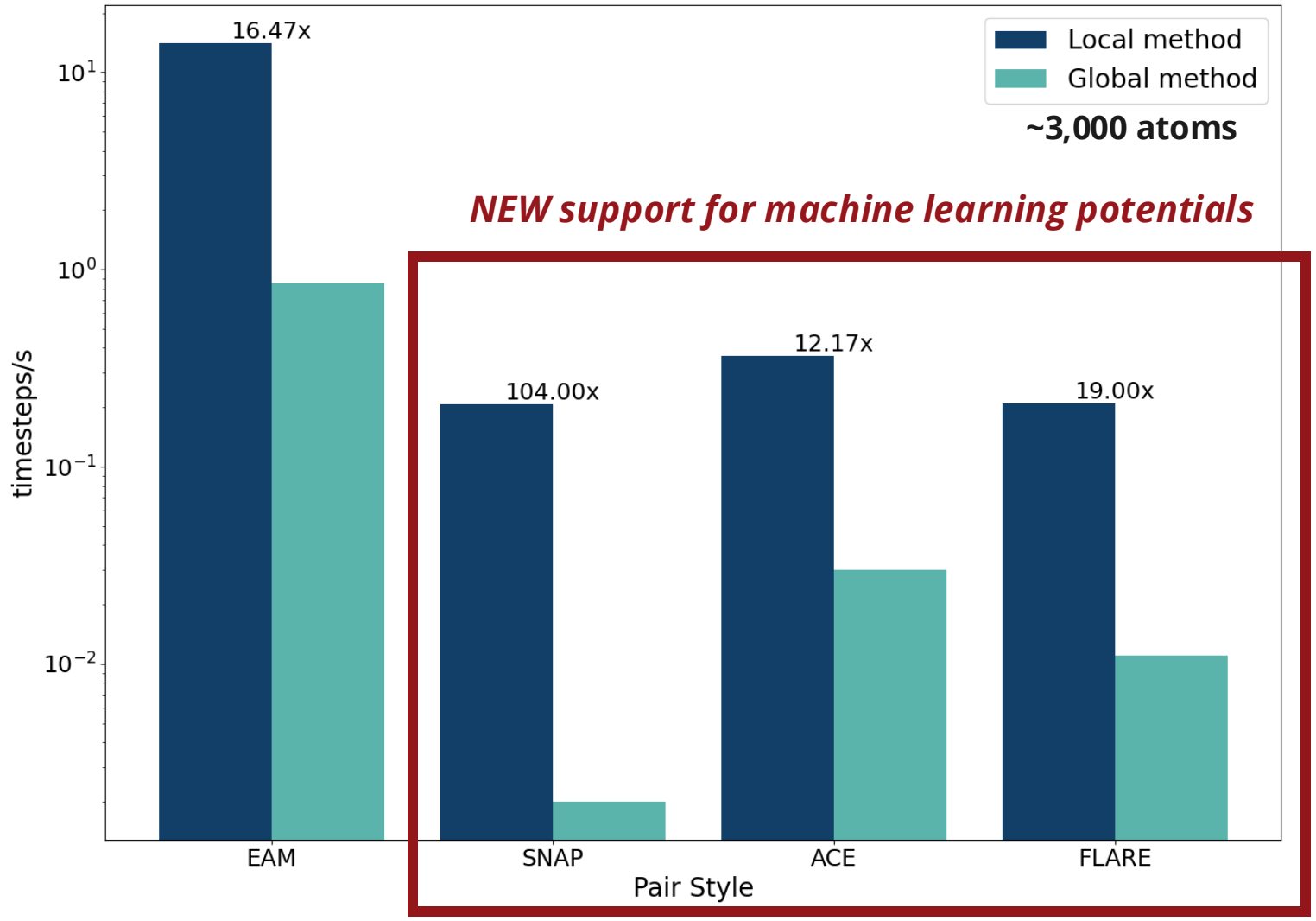


Δt = 0

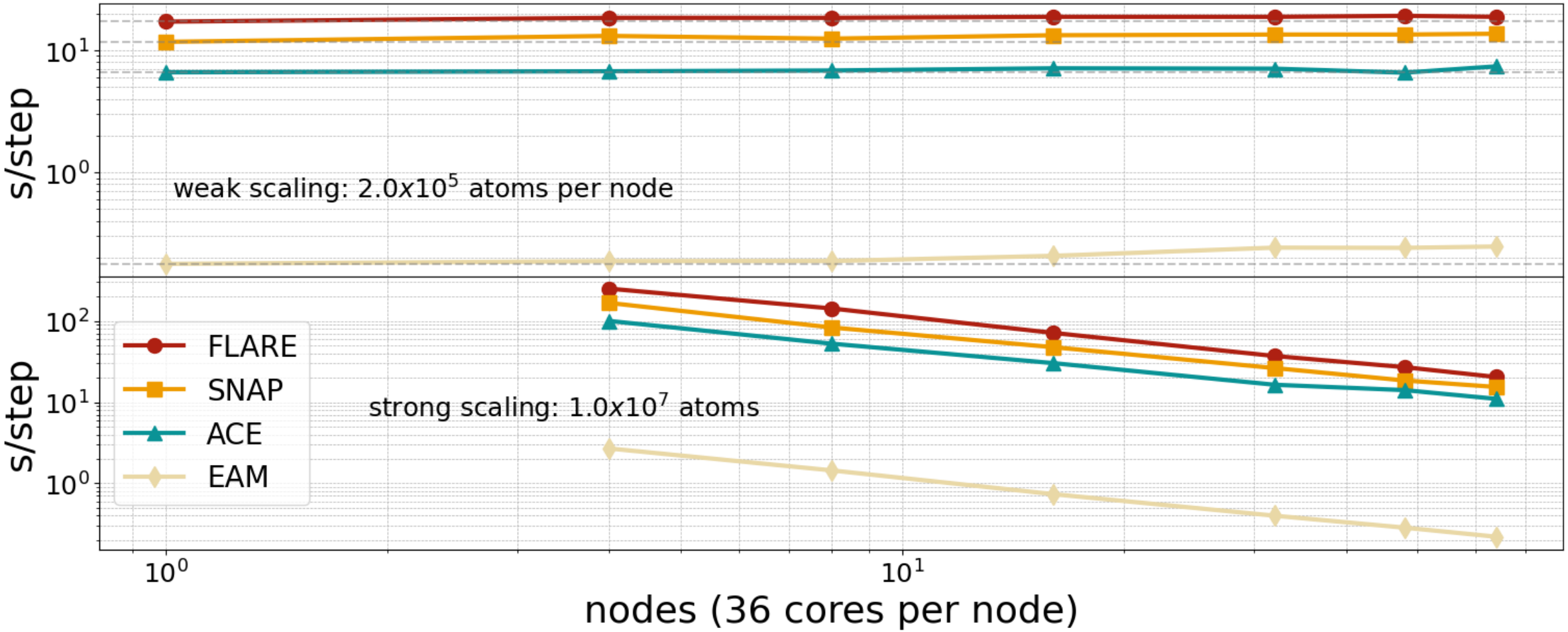


Δt = t_{end}

SPEEDUP: GLOBAL VS. LOCAL METHOD



PERFORMANCE: STRONG AND WEAK SCALING ANALYSIS



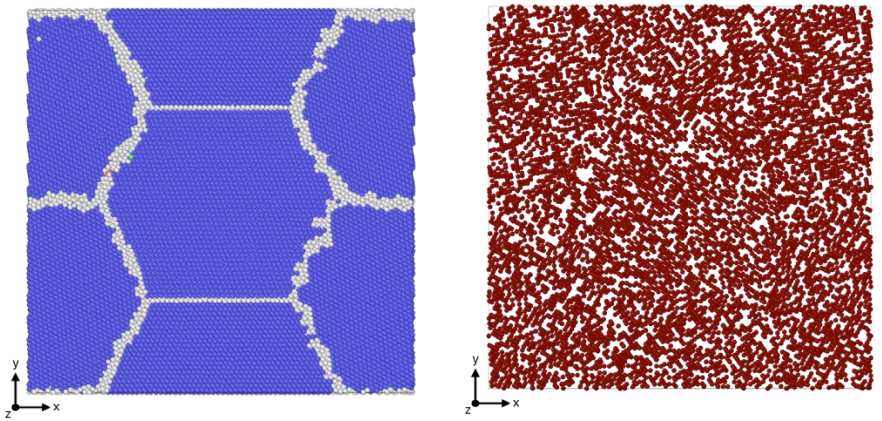
EXAMPLE SIMULATIONS OF ALLOY SPECIES SEGREGATION & PRECIPITATION

EAM/FS: Fe 50 at.% Cu 50 at.%

New code **replicates** original code
Stabilizes mixed crystalline phases quickly

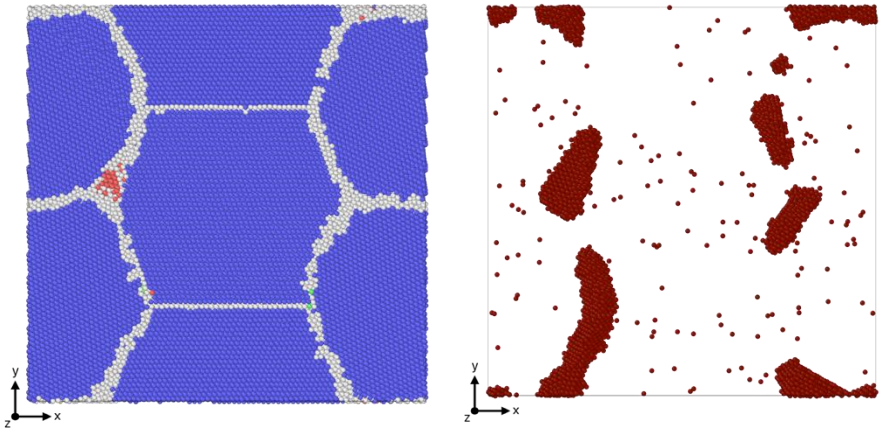
Δt = 0

Random solid solution



Δt = t_{end}

Cu precipitation
(BCC → HCP → FCC)



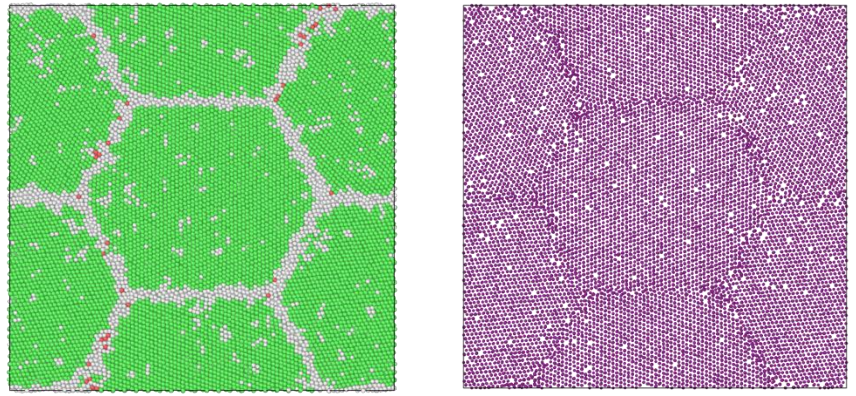
20.1 x 20.1 x 6.1nm; ~205,000 atoms, T_H = 0.5;
Swap 10% of atoms every 200 MD steps

SNAP: Ni 85 at.%, Mo 15 at. %

New code makes this **simulation feasible!** (>100x faster!)
Can now observe Ni-Mo chemical segregation trends

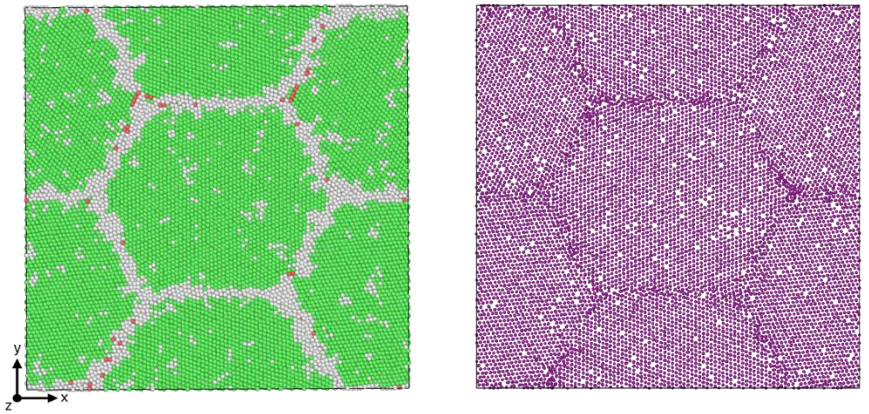
Δt = 0

Random solid solution



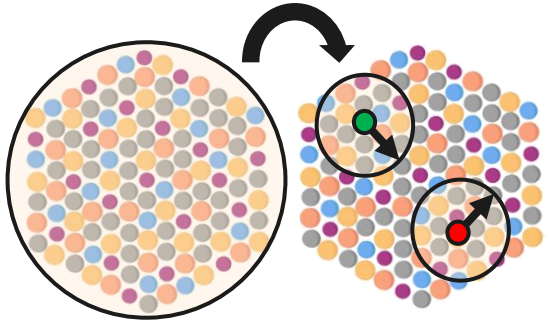
Δt = t_{end}

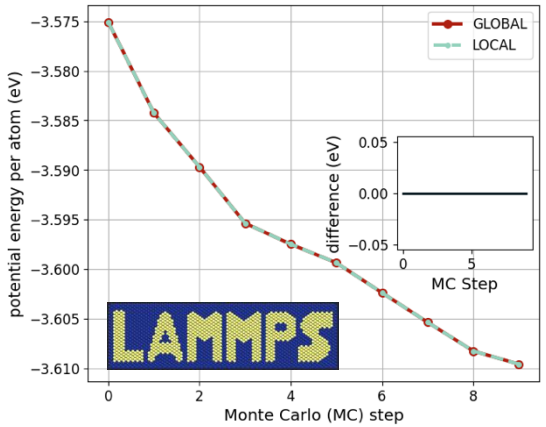
Grain boundary segregation (incipient)

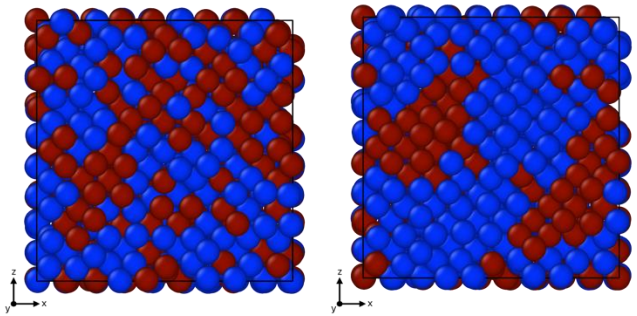


20.6 x 20.6 x 5.1nm; ~183,000 atoms; T_H = 0.85;
Swap 10% of atoms every 200 MD steps

CONCLUSIONS: A GENERAL, SCALABLE HYBRID MC/MD FOR ALLOY SIMULATIONS







<u>General</u>, scalable hybrid MC/MD in LAMMPS

- **Support in LAMMPS for popular MLIPs**
- Can be extended to many more potential forms
- Command: `fix sgcmc atomic/energy yes`

Enables new classes of alloy simulations

- Reproduce exact MC swaps as global method
- Speedups of 10-100x (or ∞!)
- Choice of accuracy of potential

Future plans for development

- Automated determination of $d\mu$ for multi-component systems
- Combine with experimental techniques like TEM

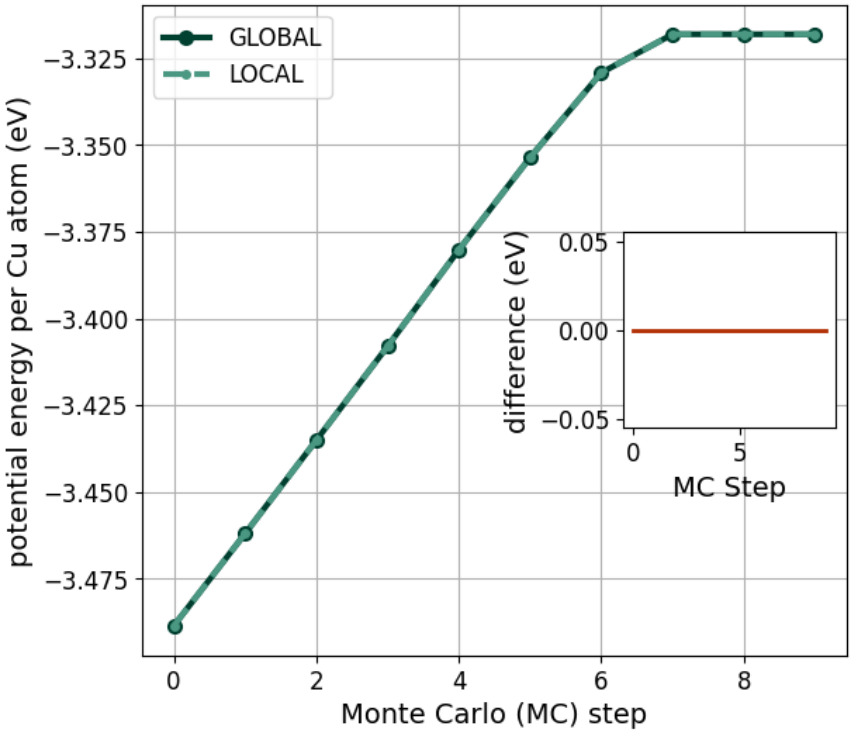
THANK YOU, QUESTIONS?

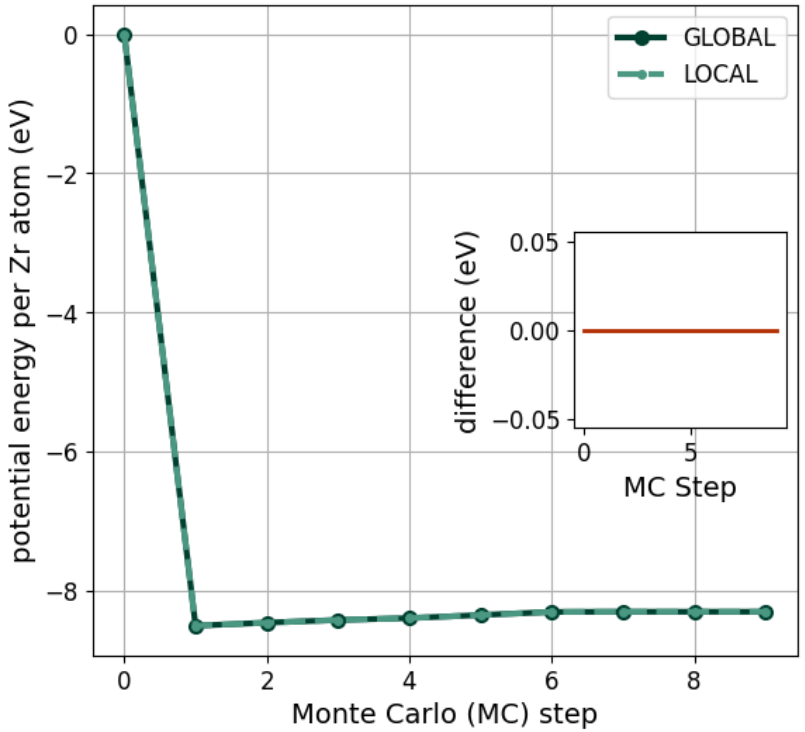
CHOO CHOO

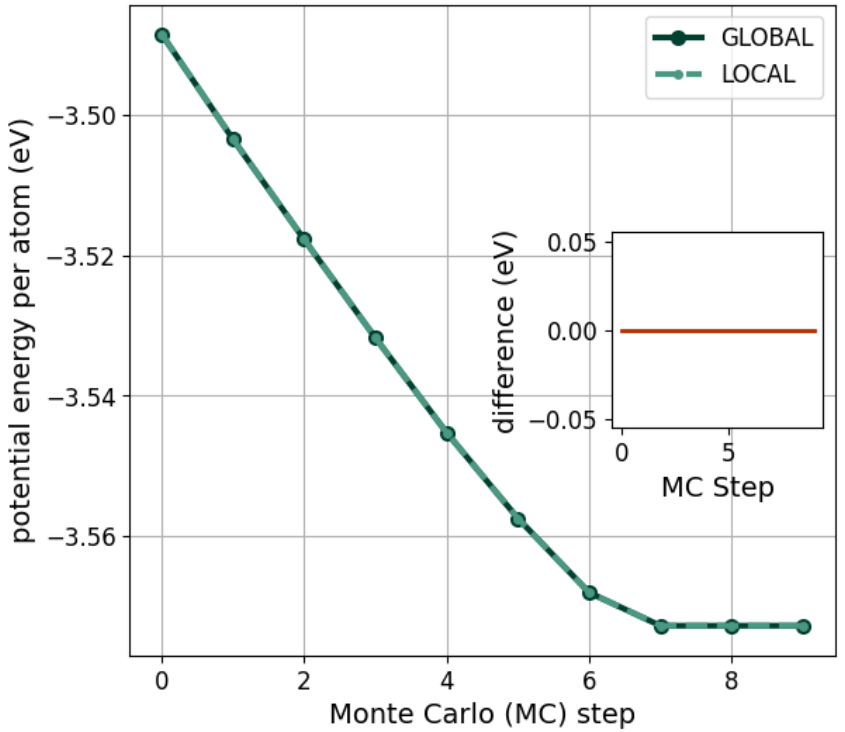
ALL ABOARD ALLOY EXPRESS!

LOCAL METHOD REPRODUCES POTENTIAL ENERGY DURING ALLOY SIMULATIONS

ACE POTENTIAL

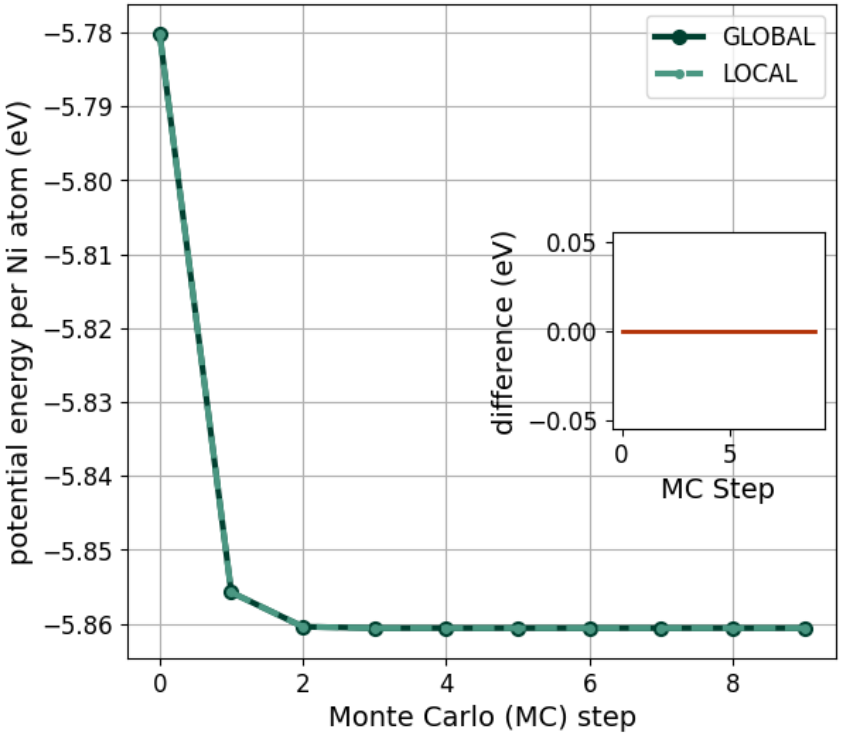


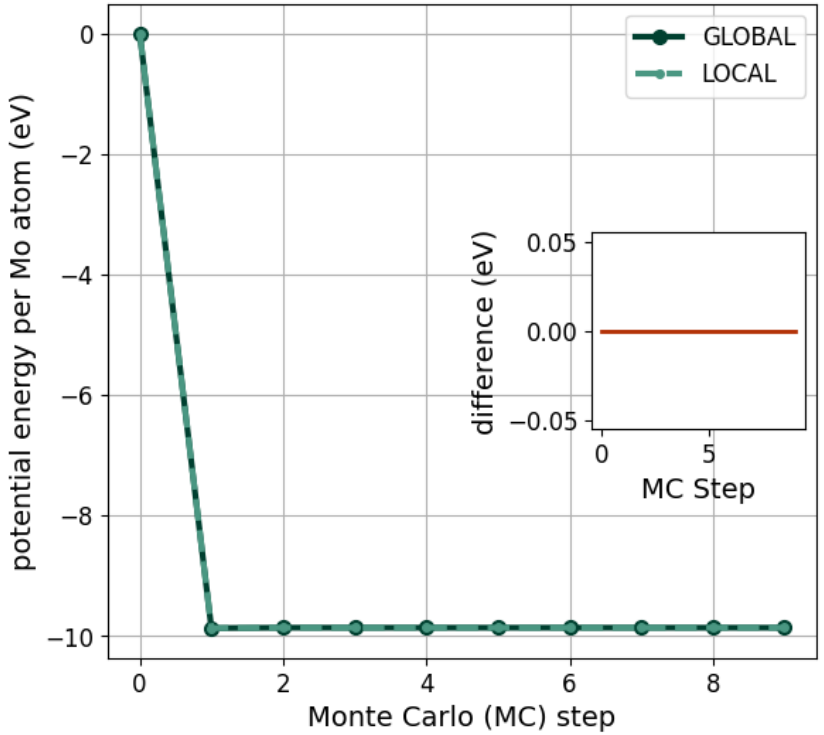


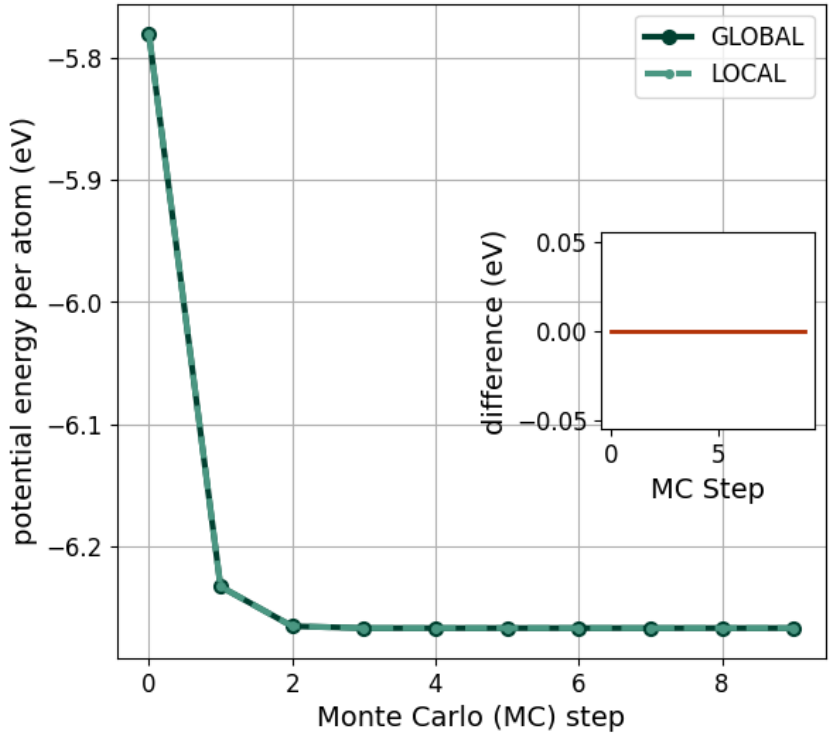


LOCAL METHOD REPRODUCES POTENTIAL ENERGY DURING ALLOY SIMULATIONS

SNAP POTENTIAL







LOCAL METHOD REPRODUCES POTENTIAL ENERGY DURING ALLOY SIMULATIONS

FLARE POTENTIAL