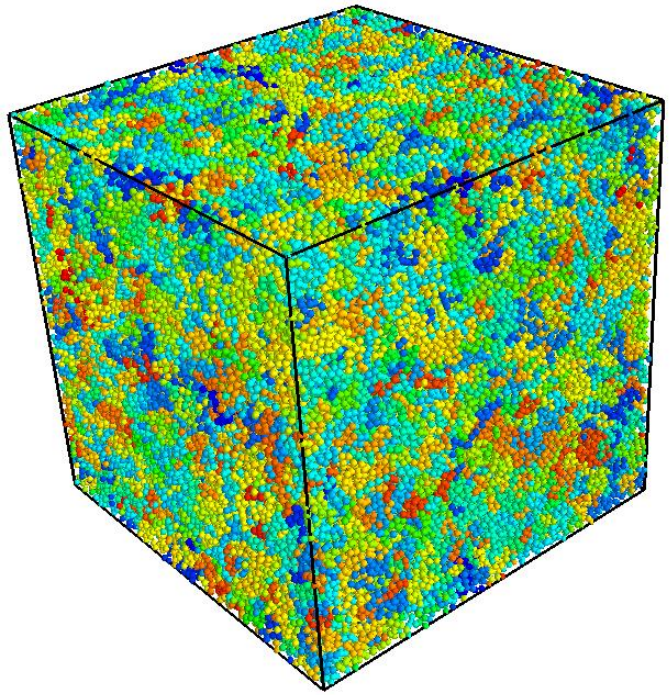
Investigating Primary Nucleation in Polymer Melts Using GPU-Accelerated Wang-Landau Simulations

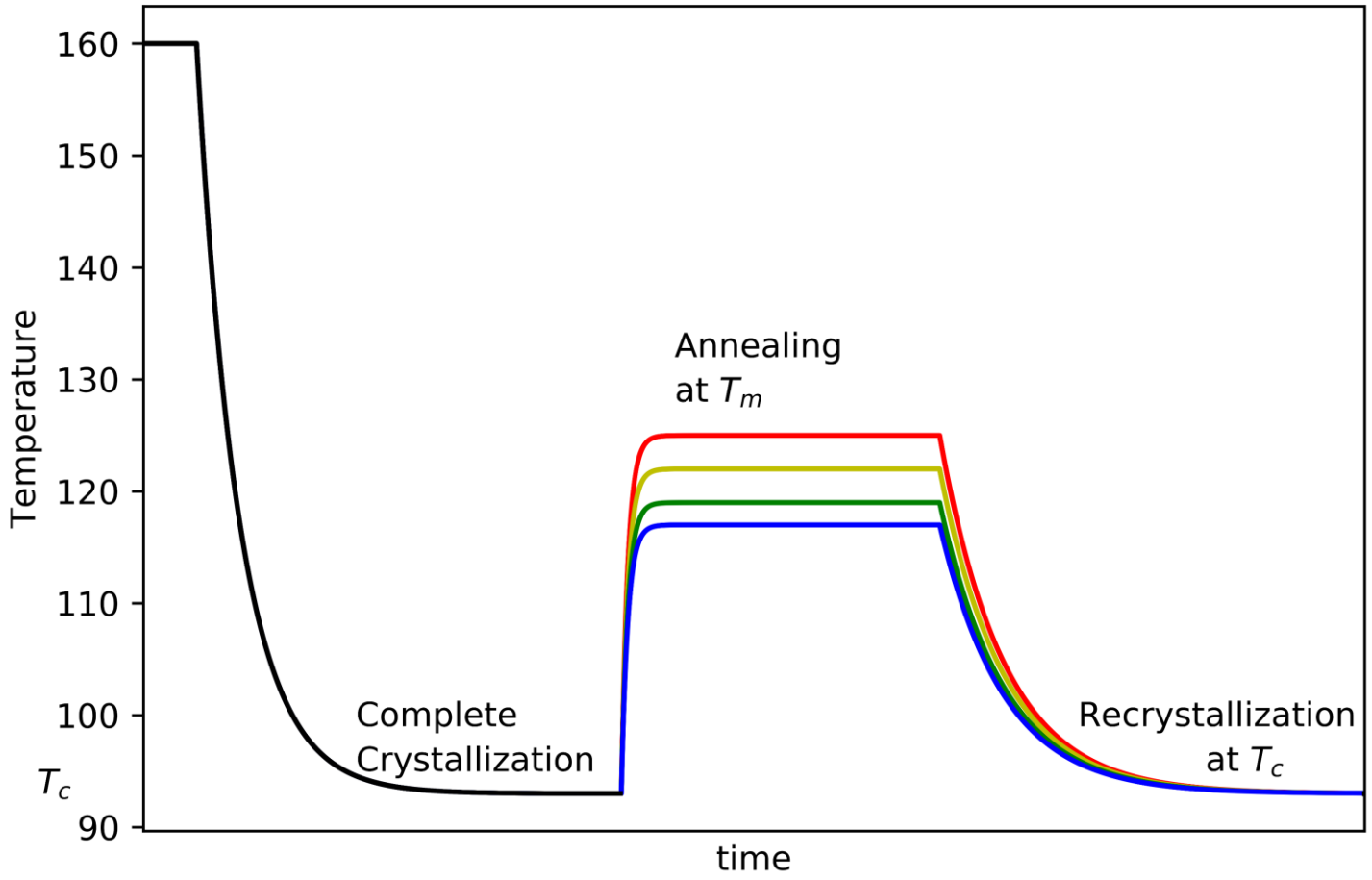
Pierre Kawak, Andrew S. Gibson, Logan S. Brown,
Beverly Delgado, Dakota S. Banks, Douglas R. Tree



16th November 2020

AIChE Annual Meeting

Melt memory is indicative of unusual crystallization behavior



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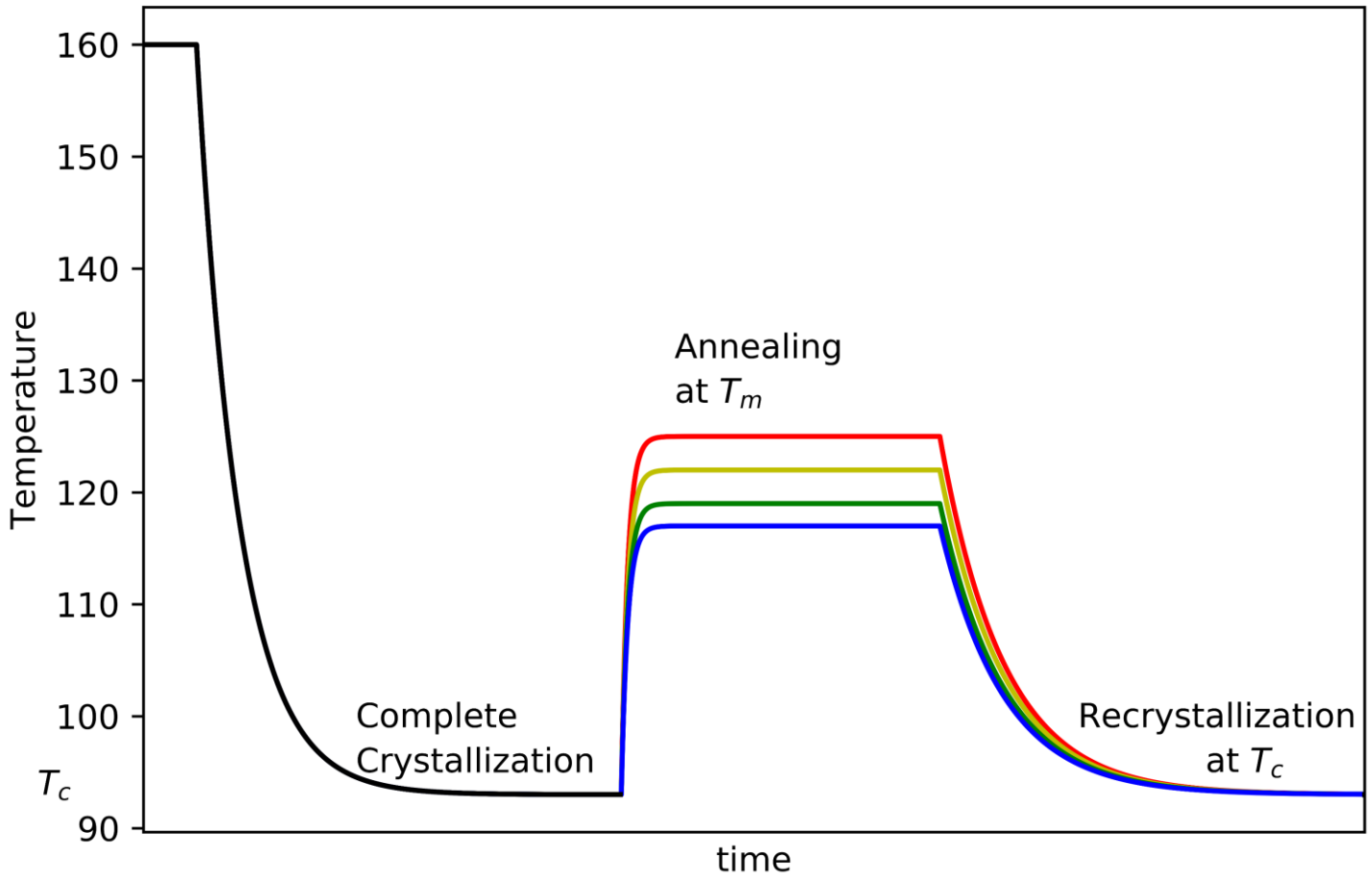

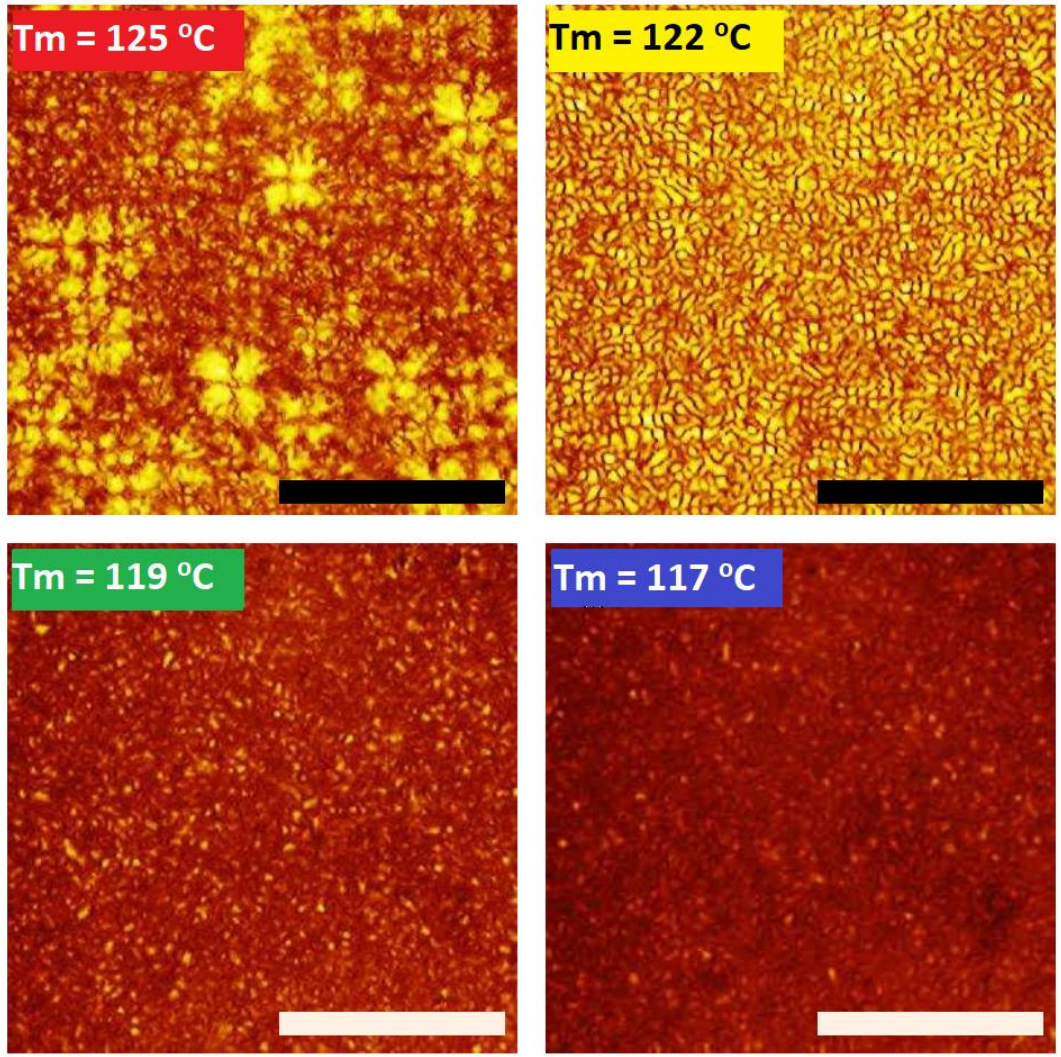

Häfele et al. Eur Phys J E (2005)

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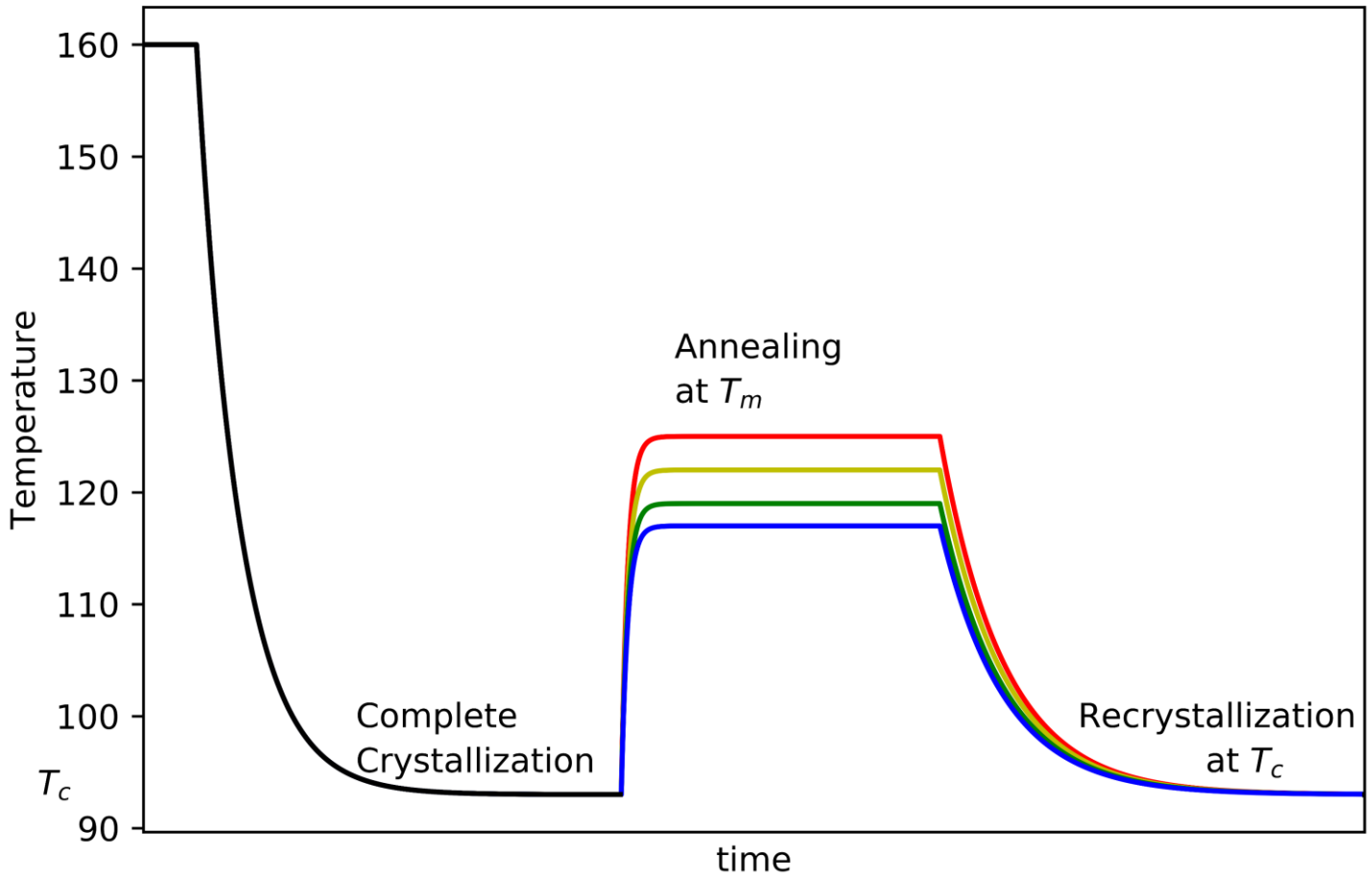

Other Observations:

- Initial larger scale ordering in SAXS and WAXS
- Deviant Crystallization and Recrystallization
- Intermediate Phase Observations
- No Copolymer Effect on Crystallization

Are the previous observations caused by slow kinetics or thermodynamics in the nucleation process?

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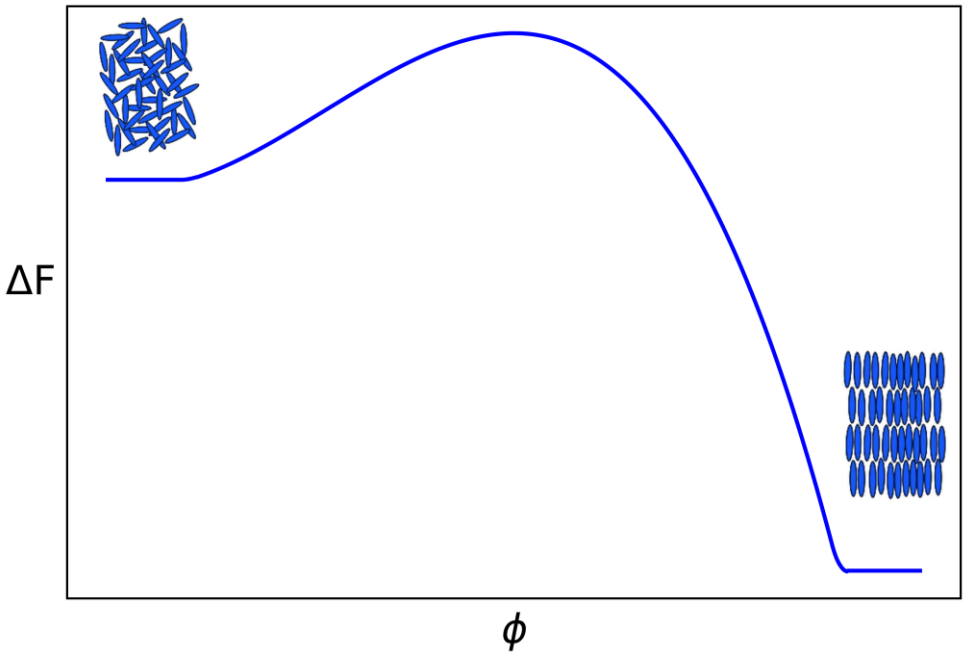
Role of friction vs. thermodynamics



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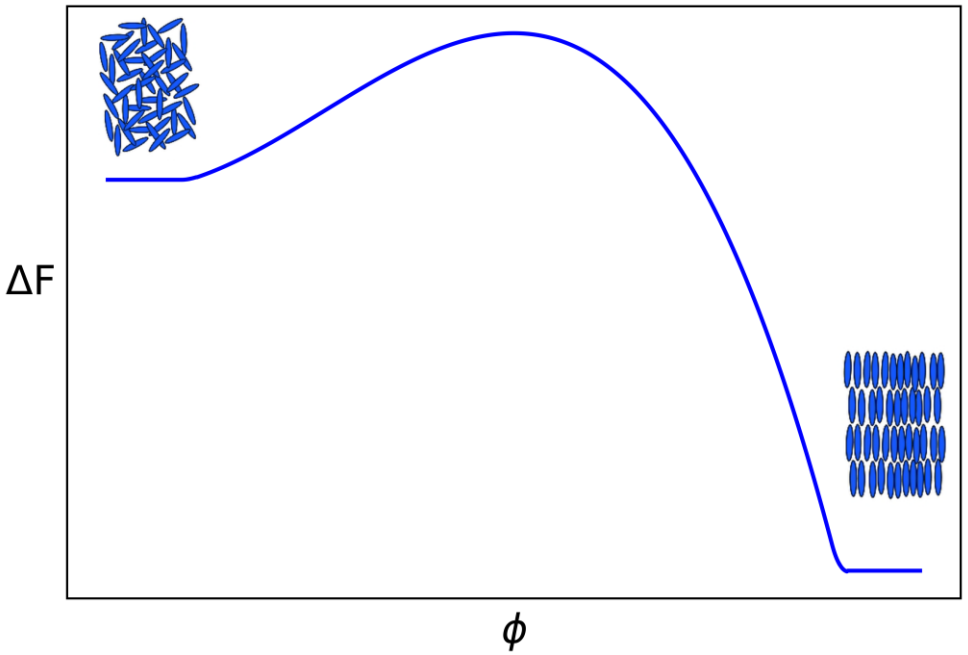
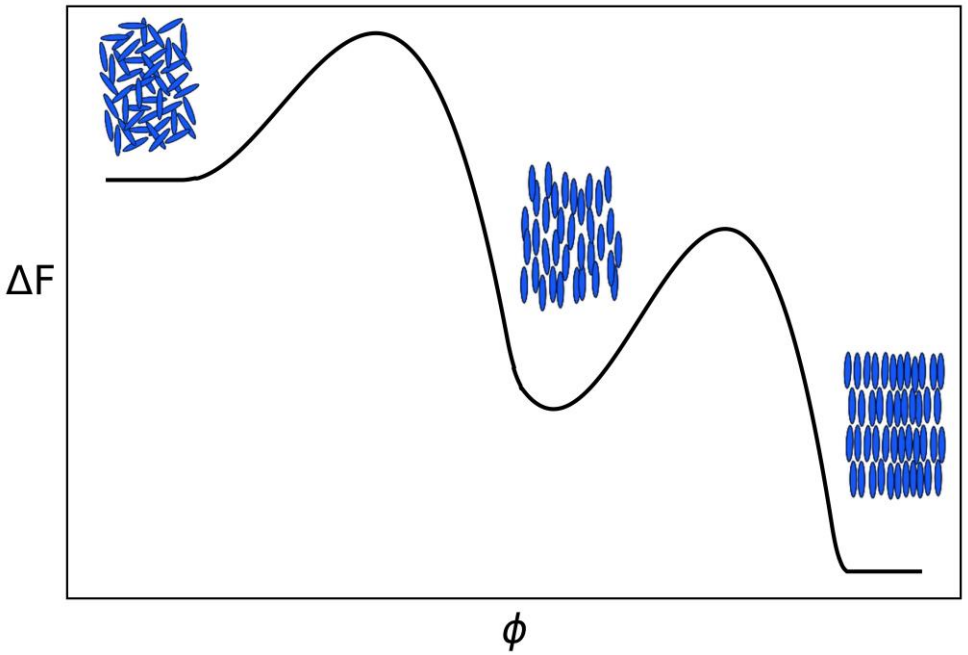
Role of friction vs. thermodynamics

Relative stability of different phases





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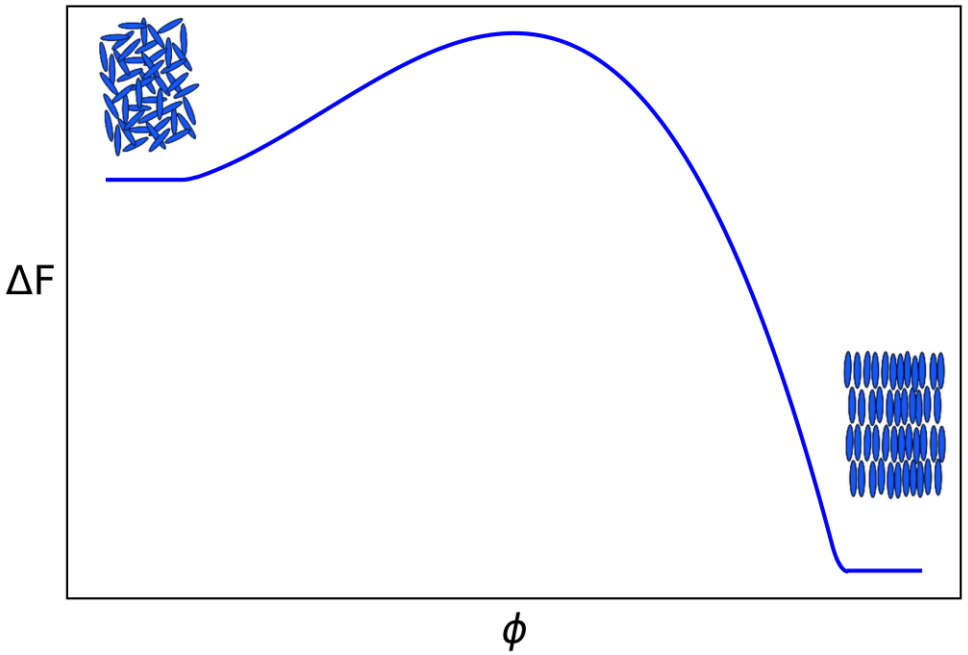

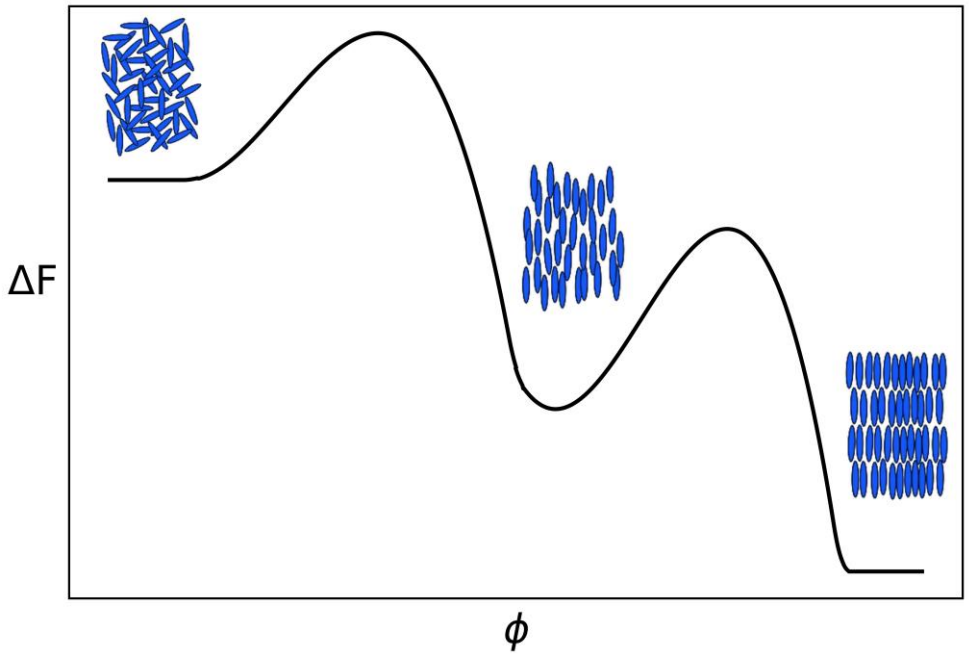

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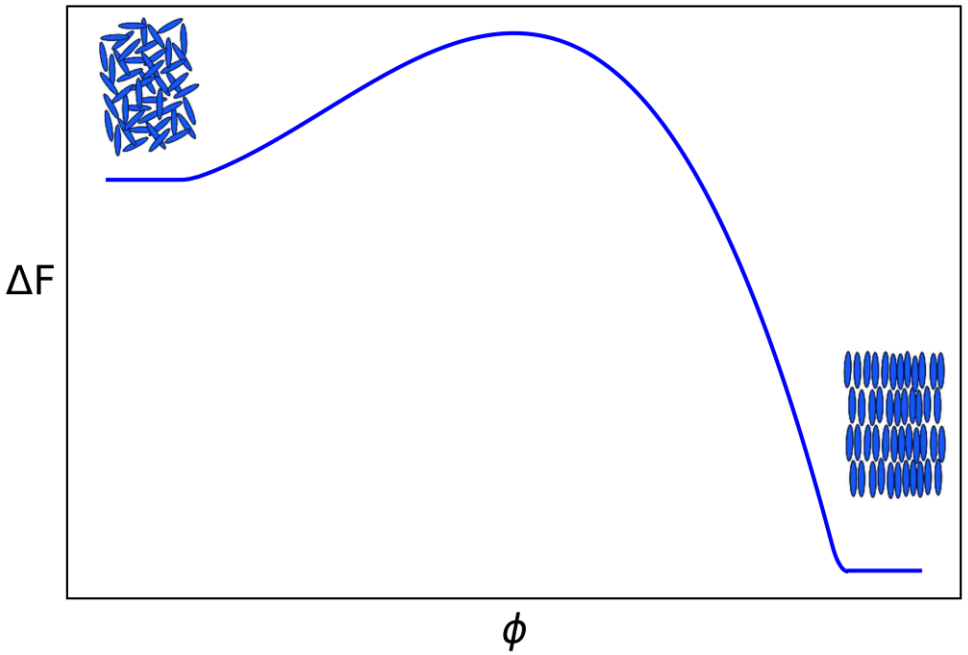
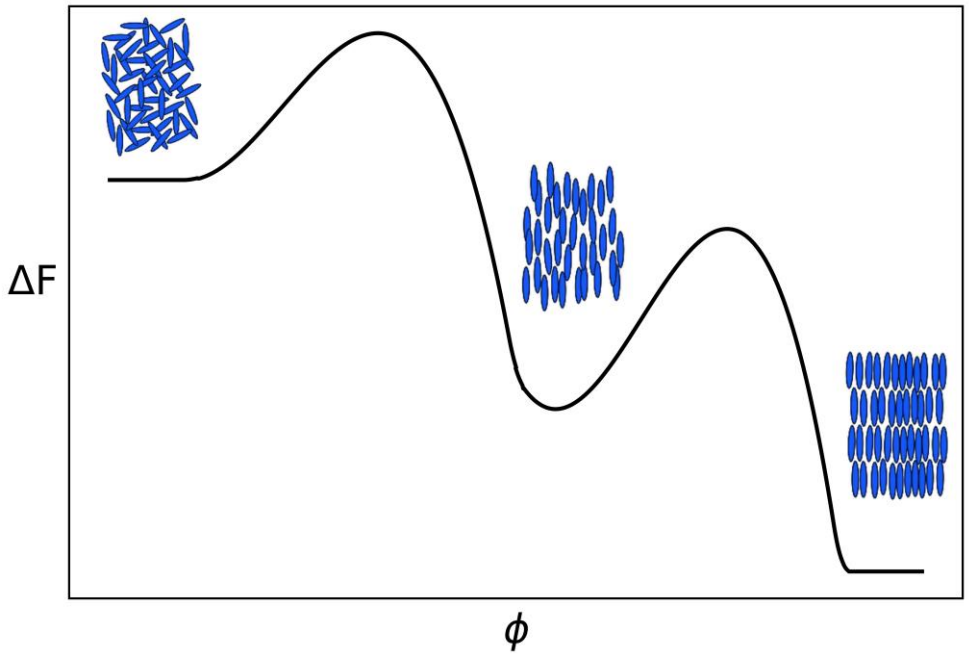
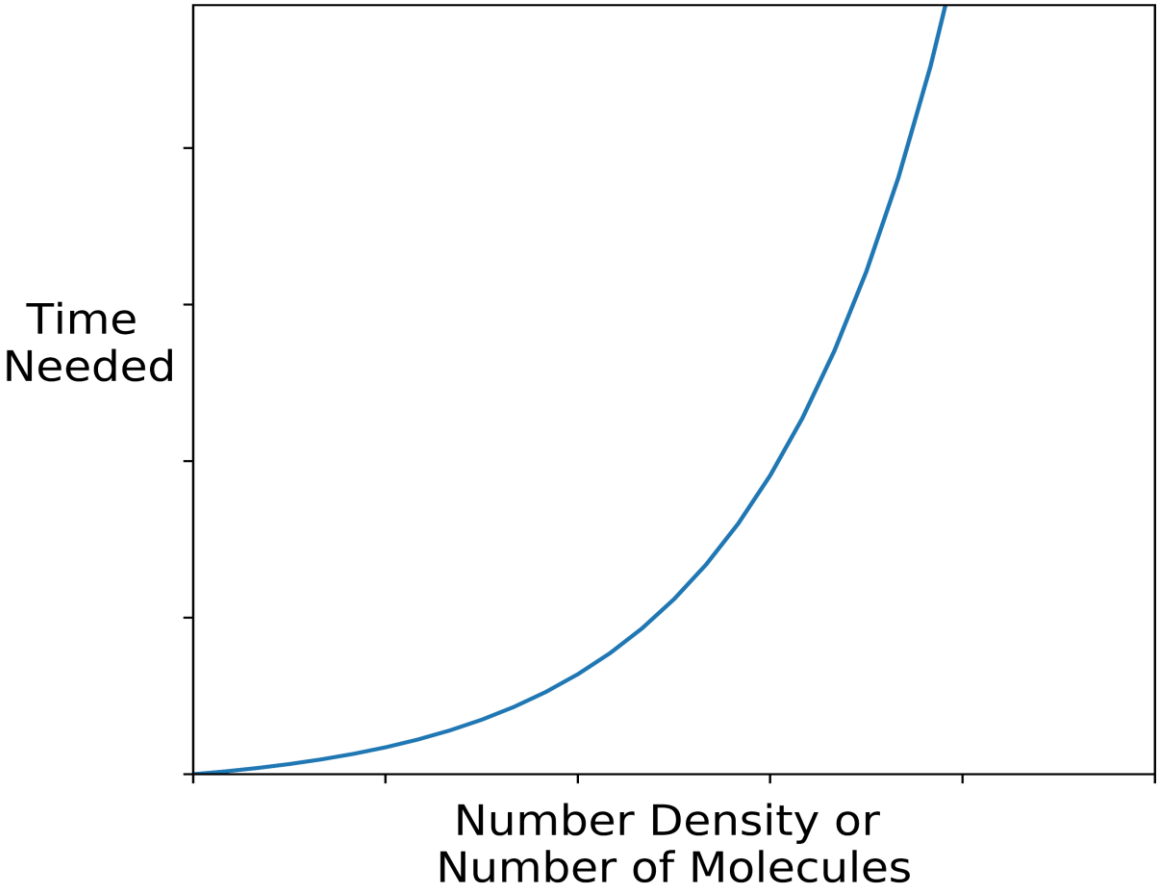




Mission: Build Free Energy Surface (FES)

There are 2 major obstacles to overcome to better understand crystallization

Polymer simulations are expensive



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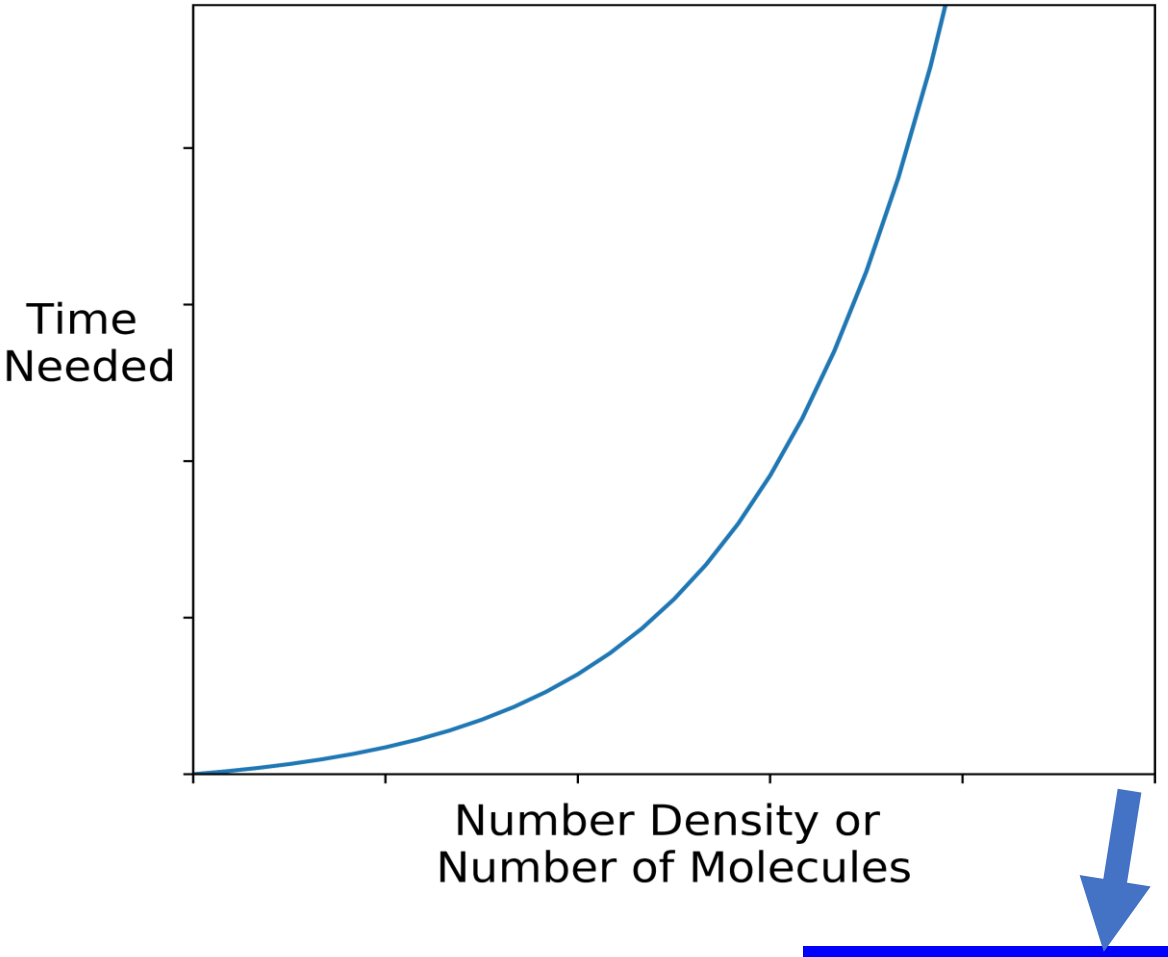

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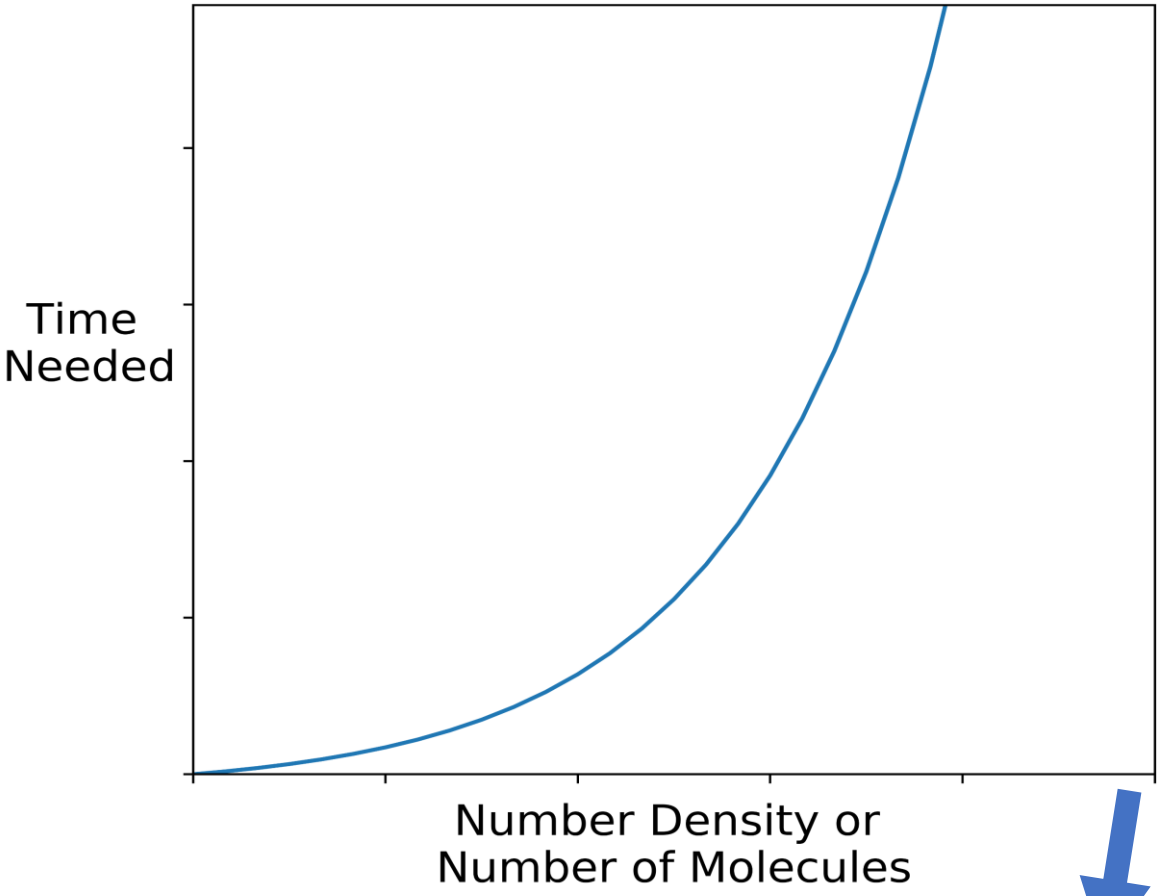

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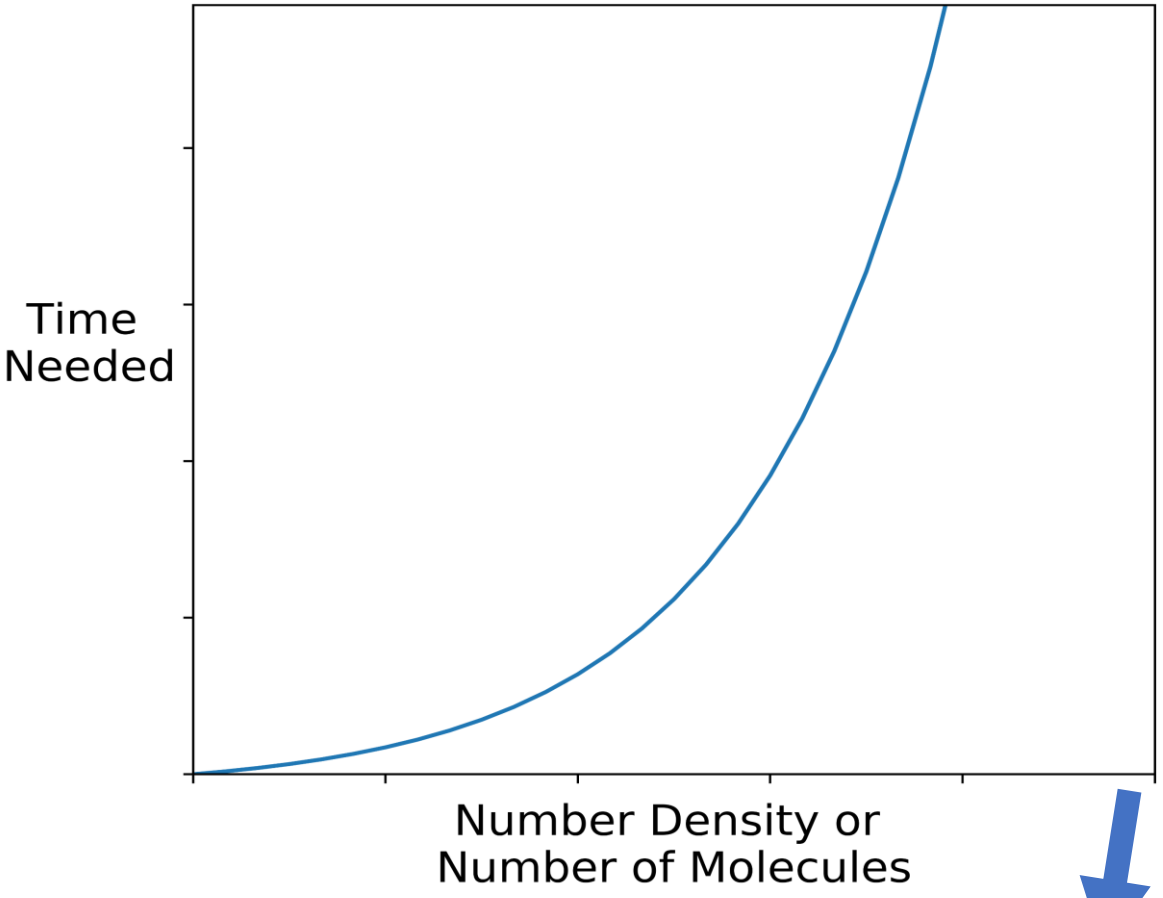

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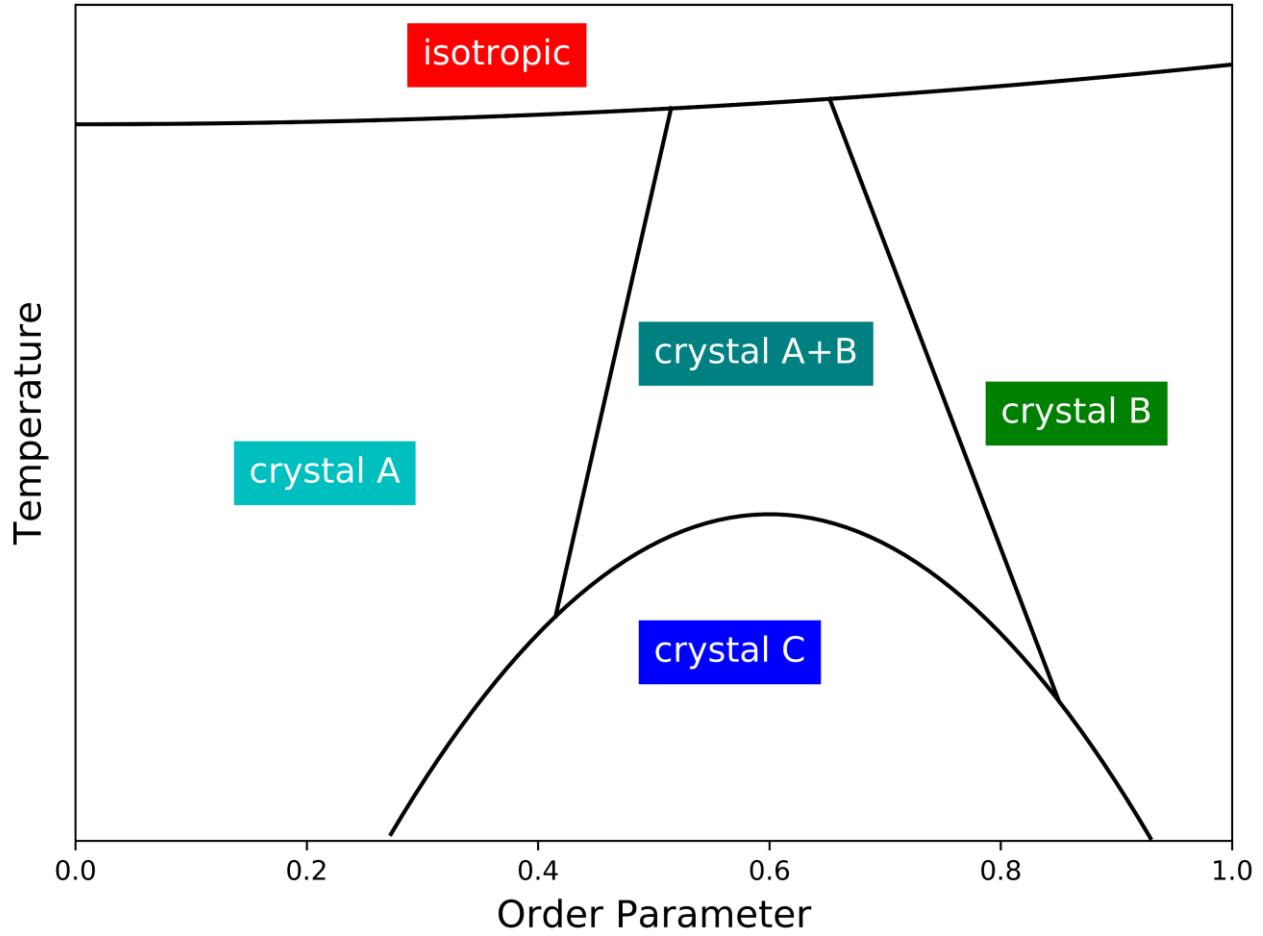

What simulation techniques can build an FES?

Molecular Dynamics (MD)

- **Access to dynamic properties**
- **Readily available (e.g. LAMMPS)**
- **Limited by entanglement dynamics**
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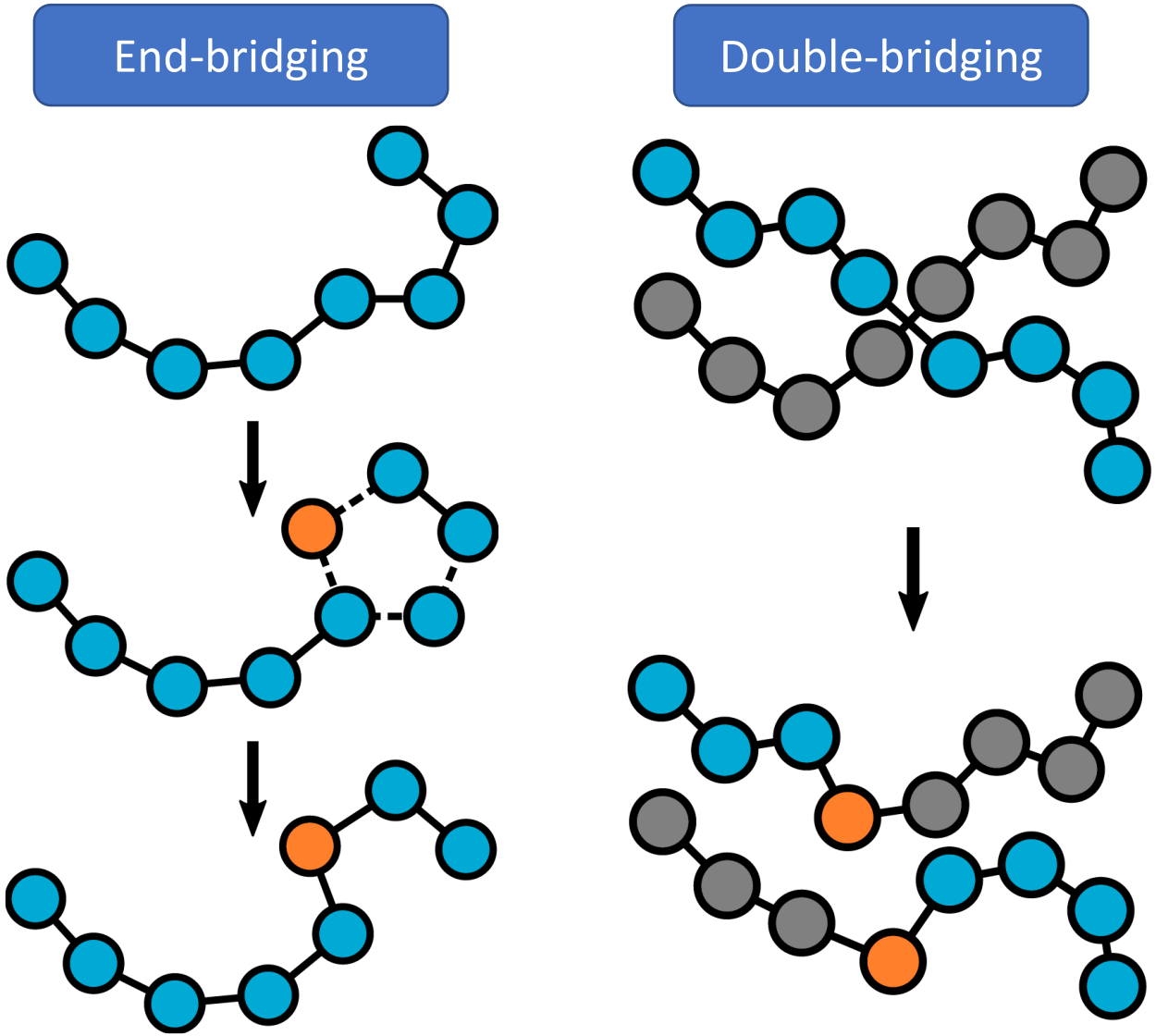
Monte Carlo (MC)

- **Bond-breaking moves**
- **No dynamic considerations**
- **Faster and simpler**

We can use advanced move set and domain decomposition to speed equilibration



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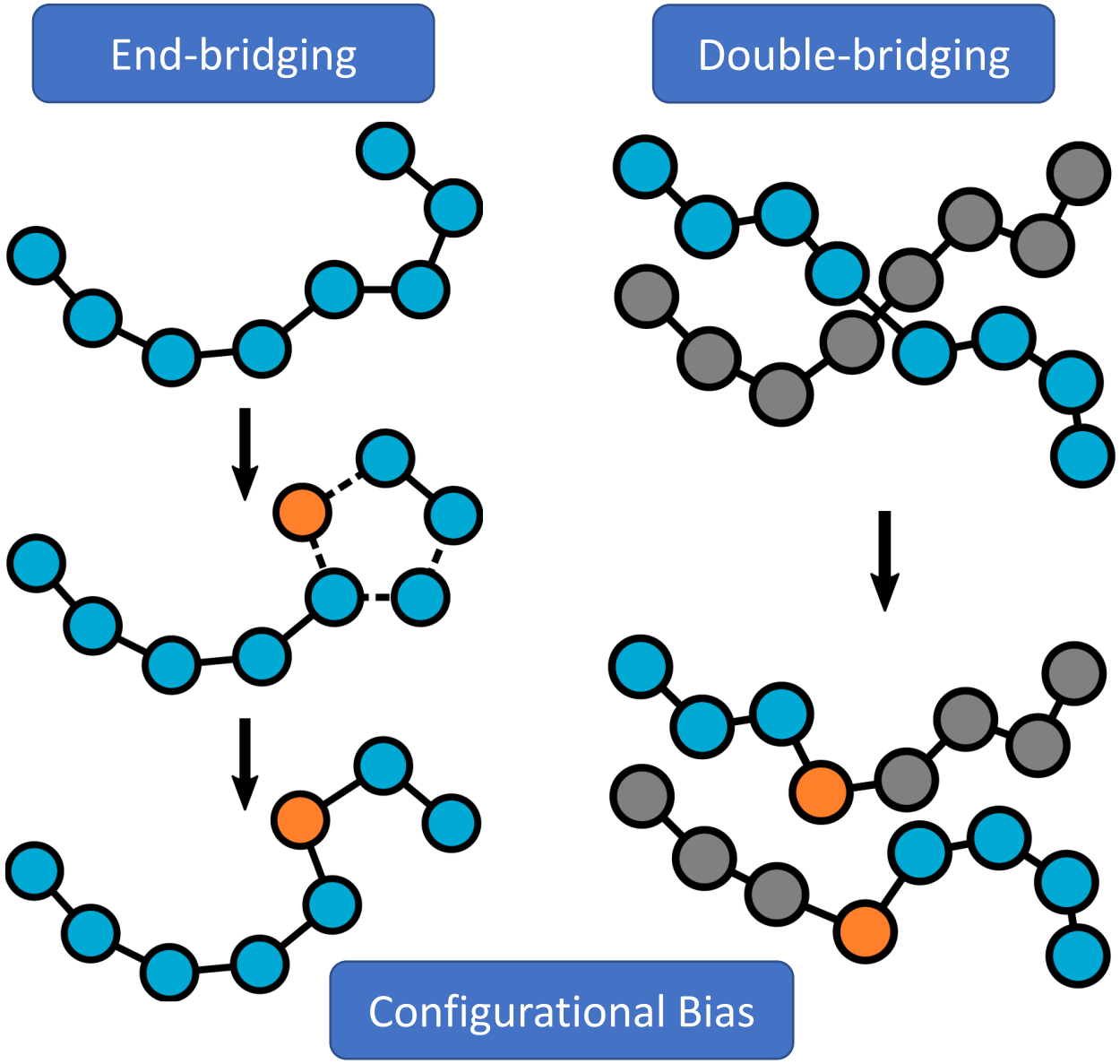

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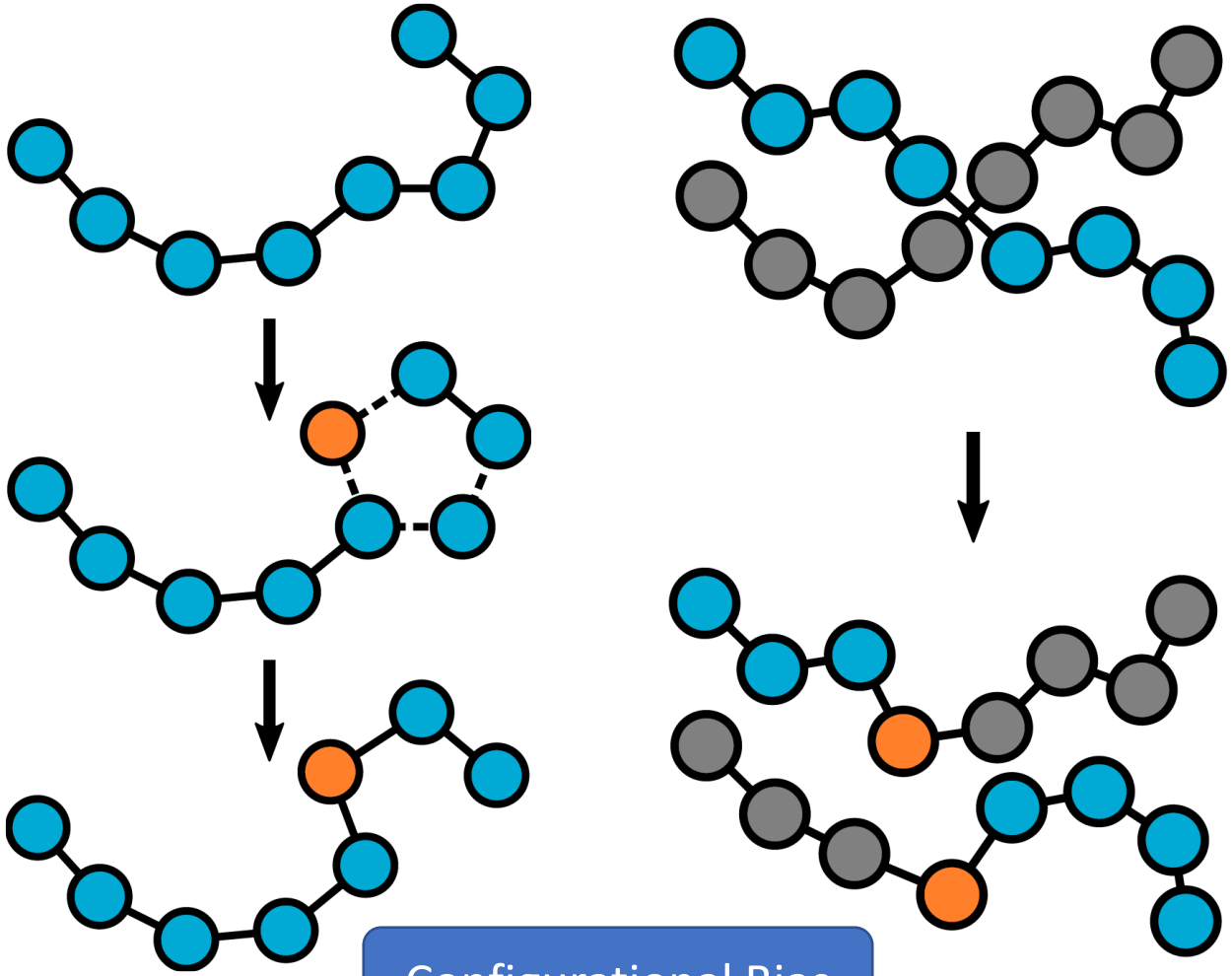
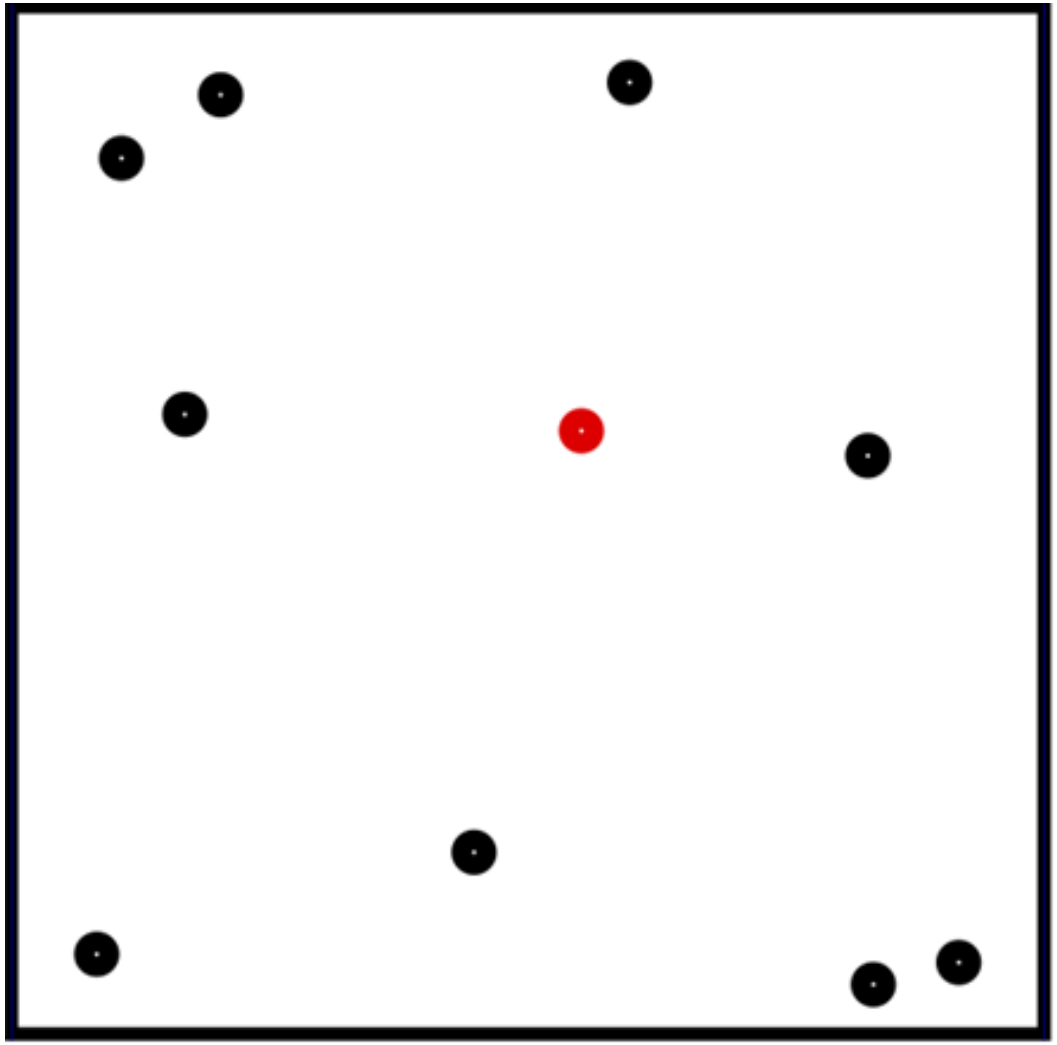
End-bridging

Double-bridging



Configurational Bias



Conventional MC

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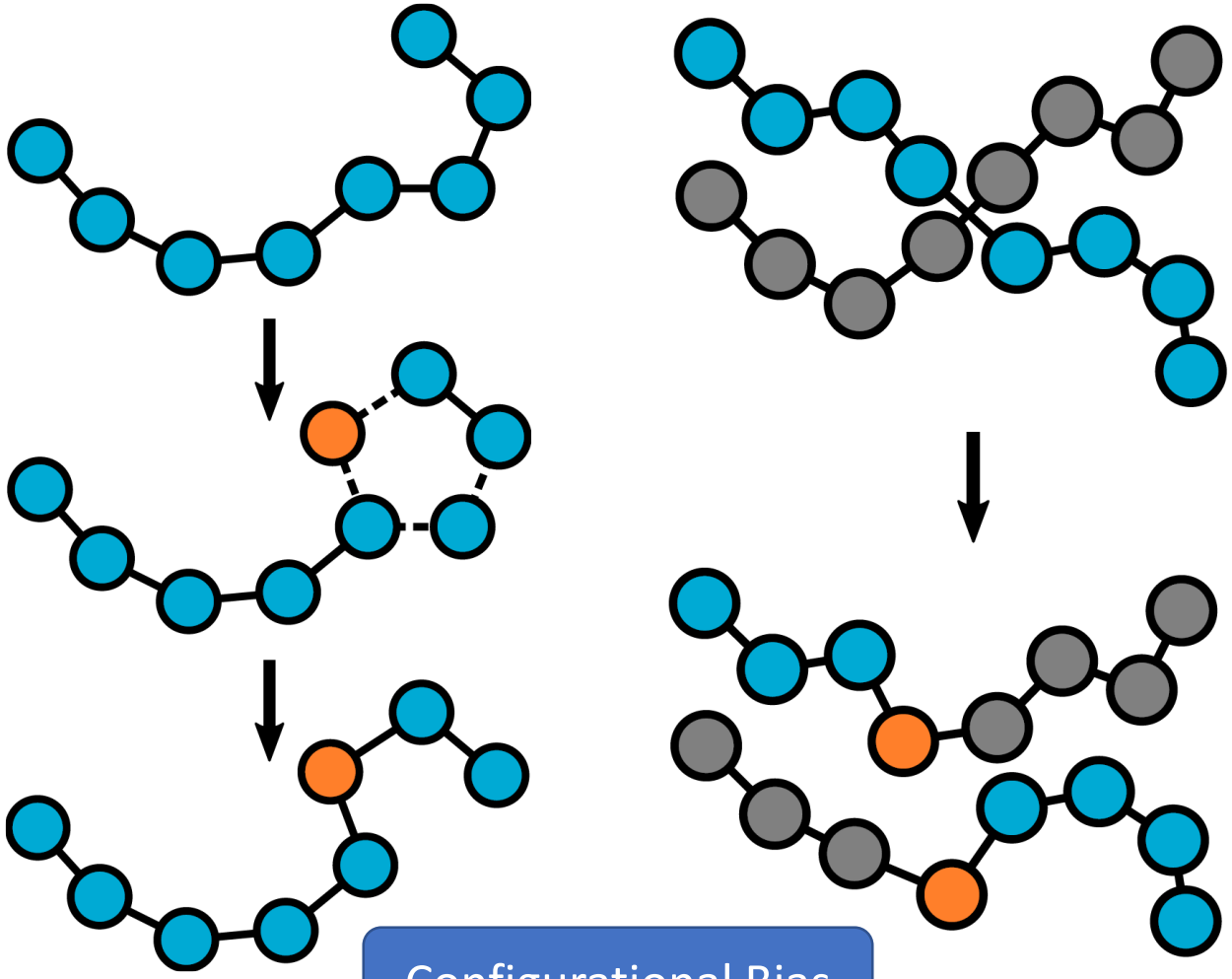

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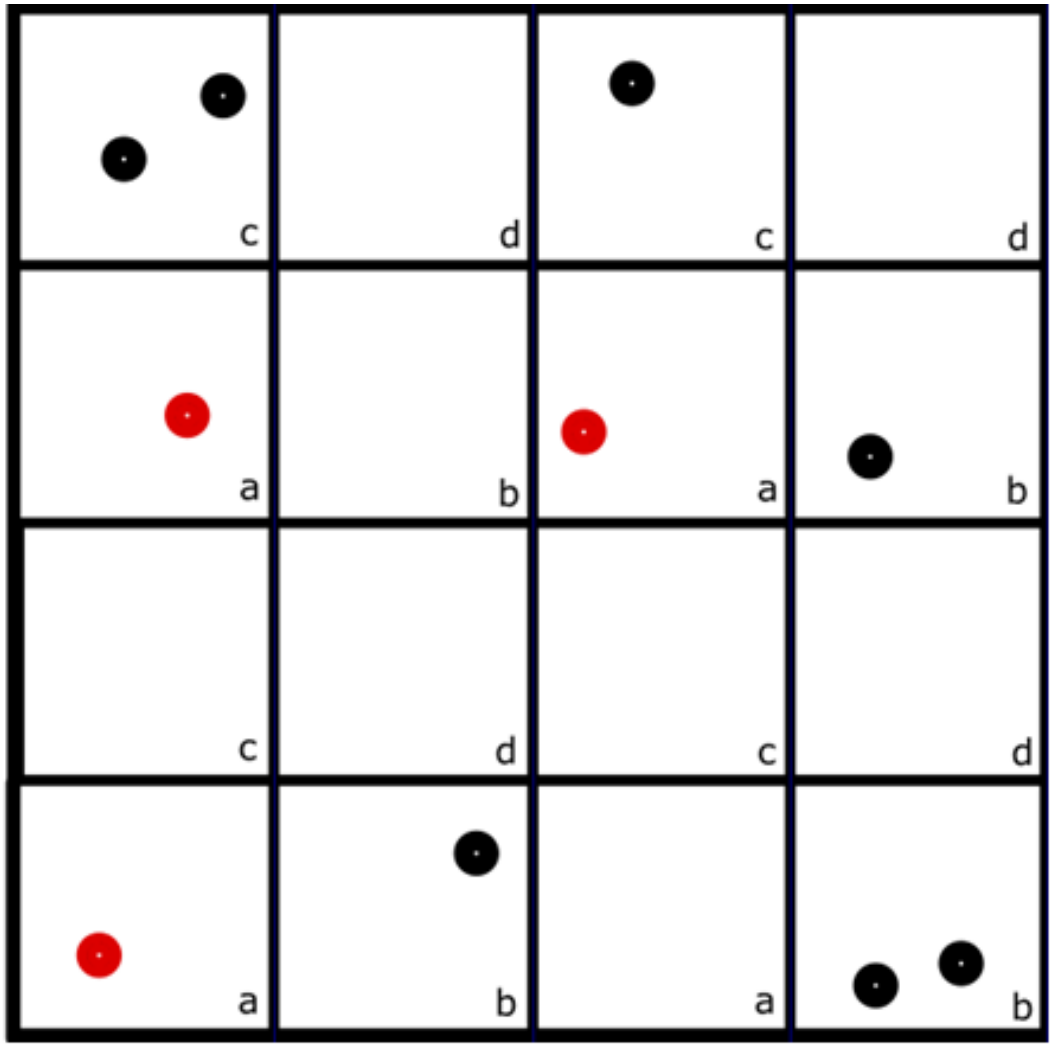

Checkerboard MC

We can use advanced move set and domain decomposition to speed equilibration

Anderson et al. J. Comp. Phys. (2013)

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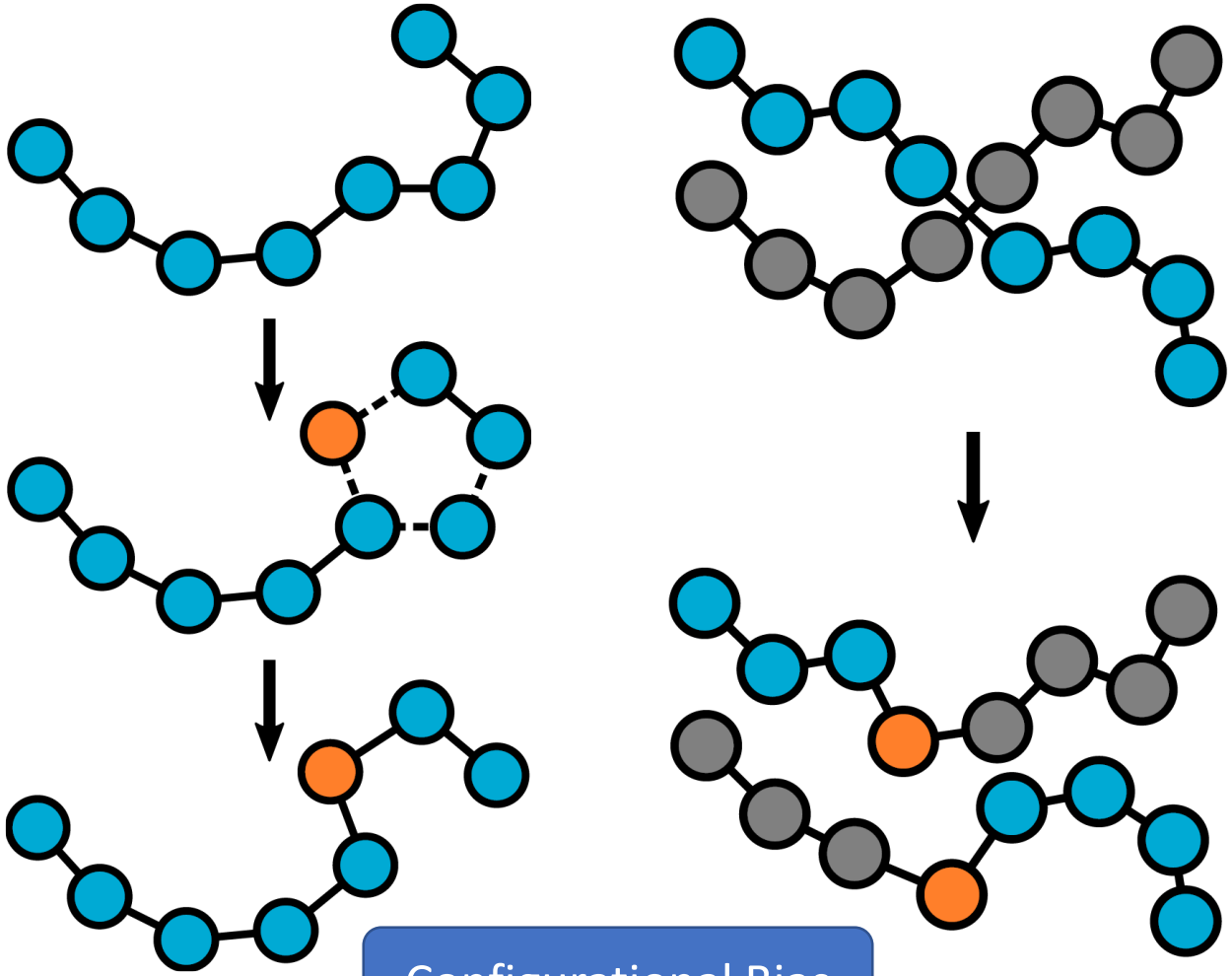

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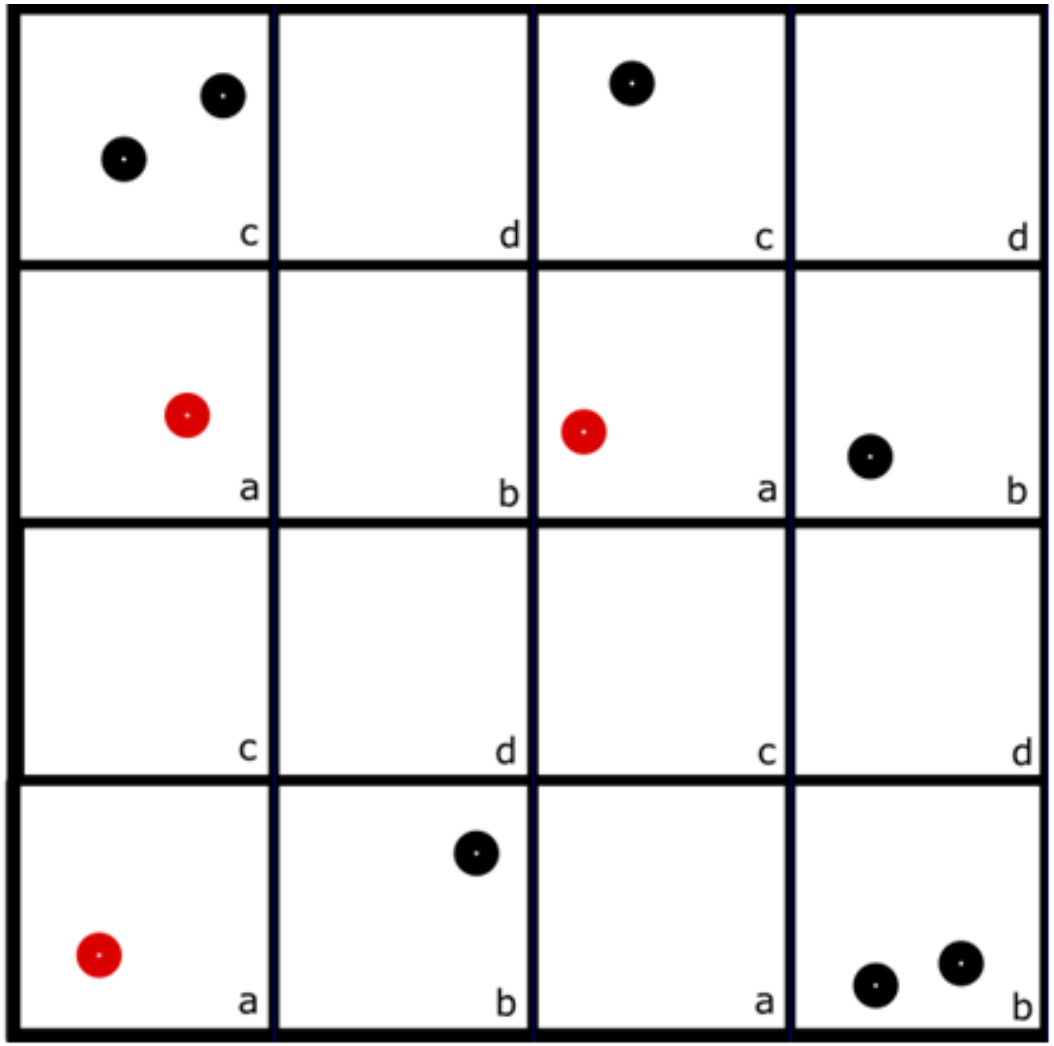
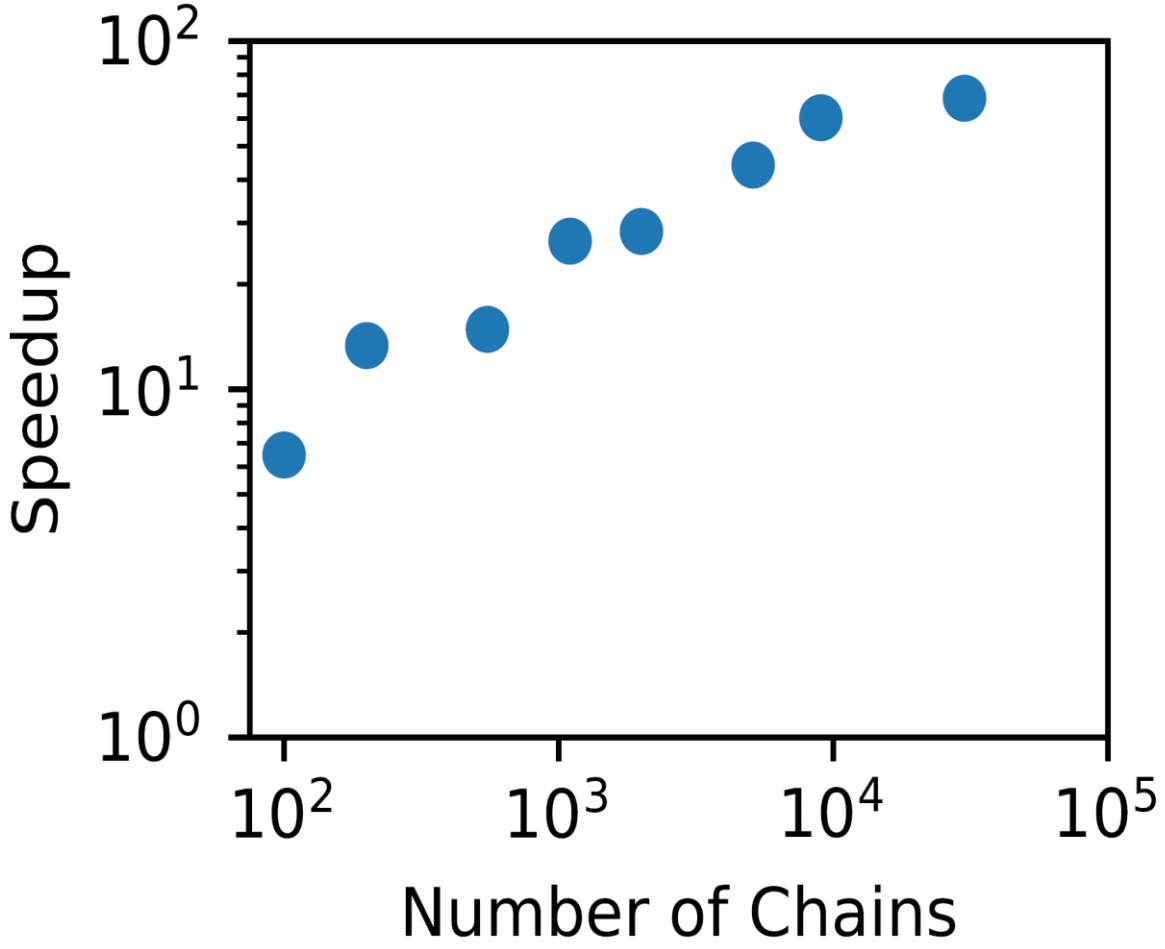


Checkerboard MC

Parallel simulations are 2 orders of magnitude faster



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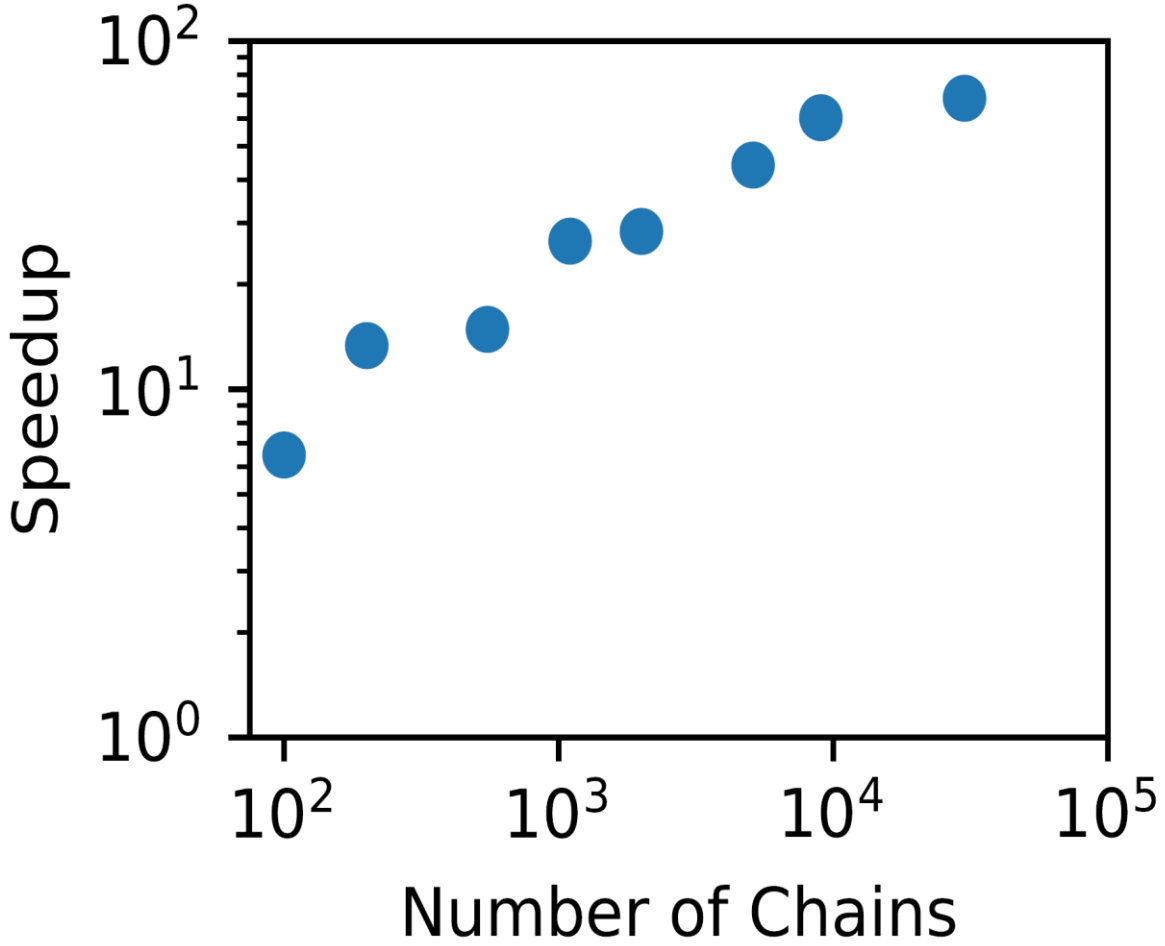

$$Speedup = \frac{time\ per\ move_{serial}}{time\ per\ move_{parallel}}$$

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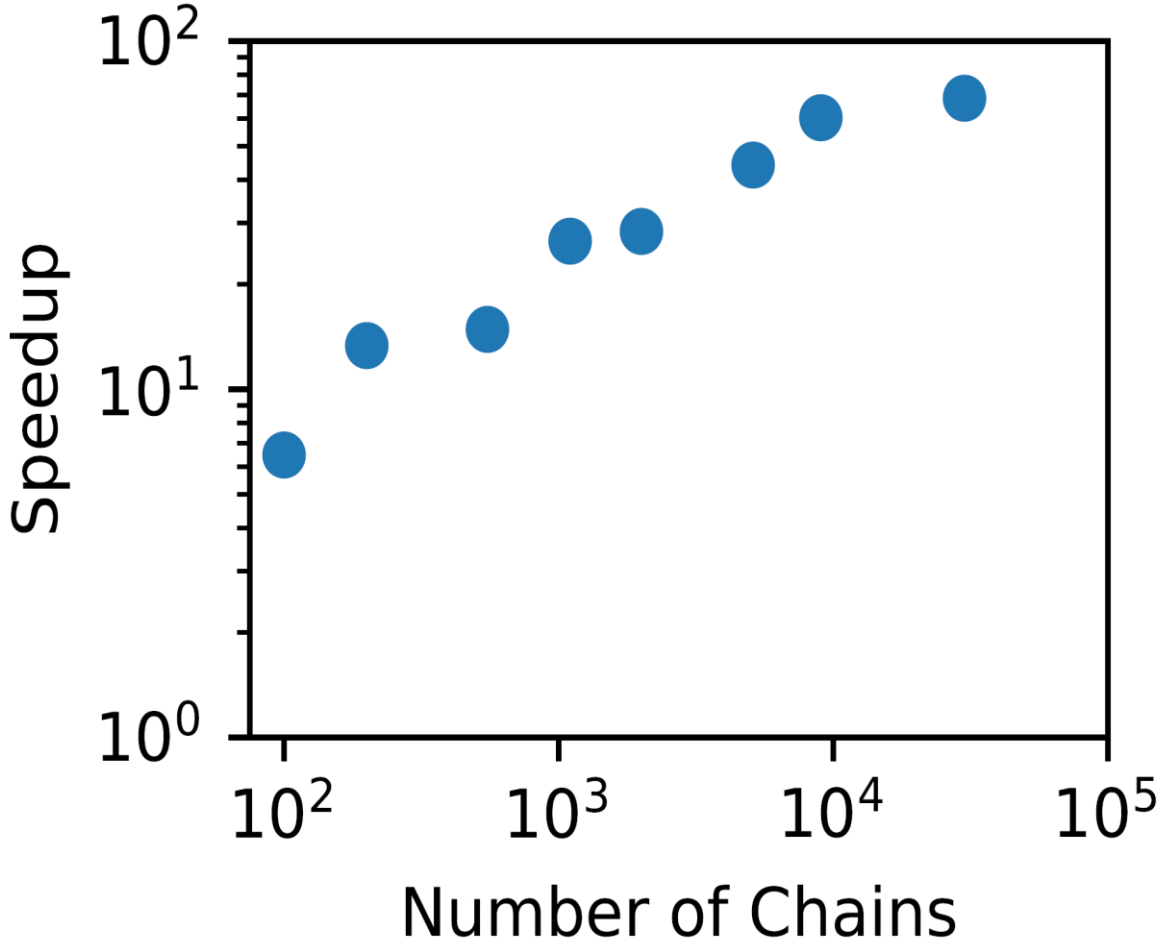

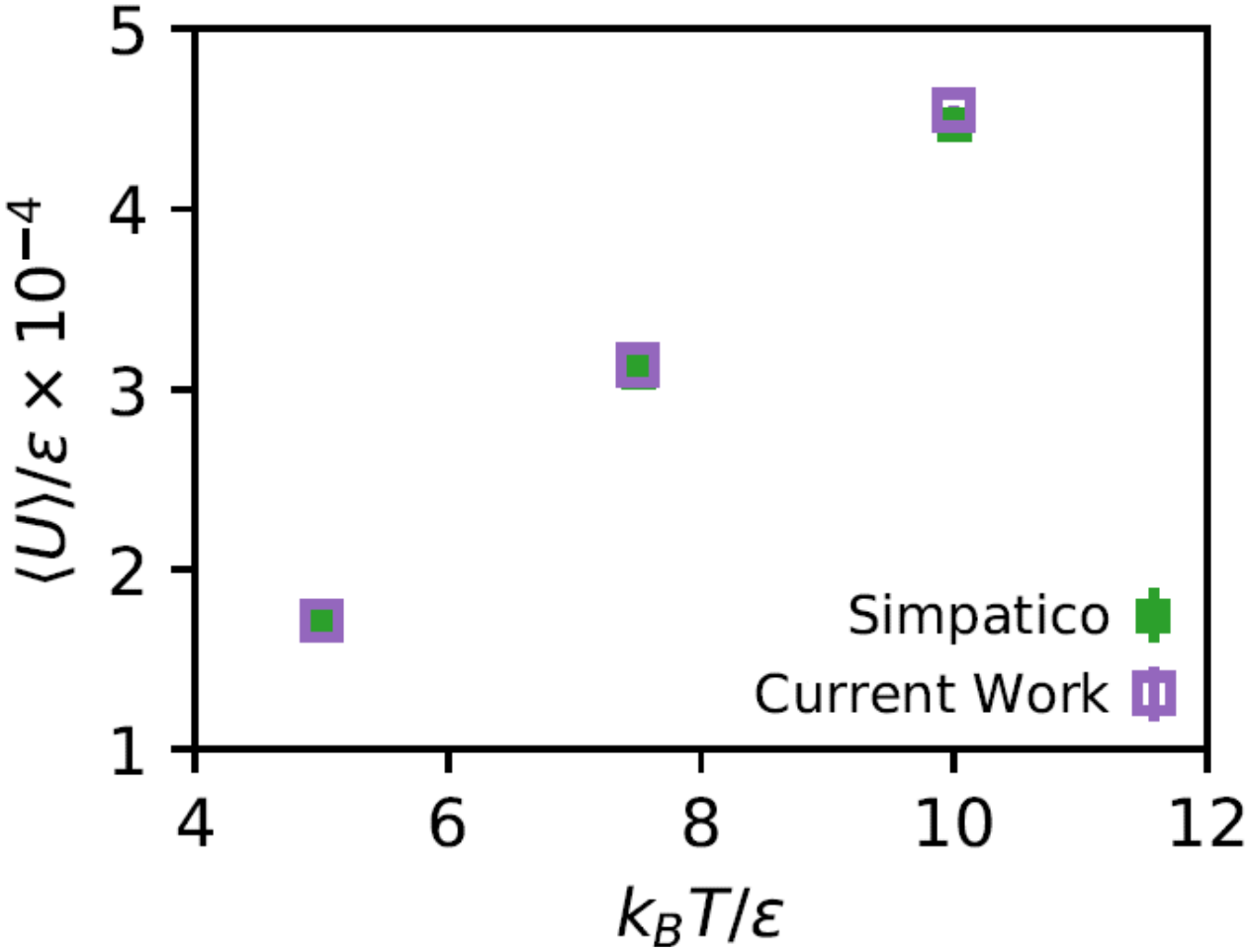

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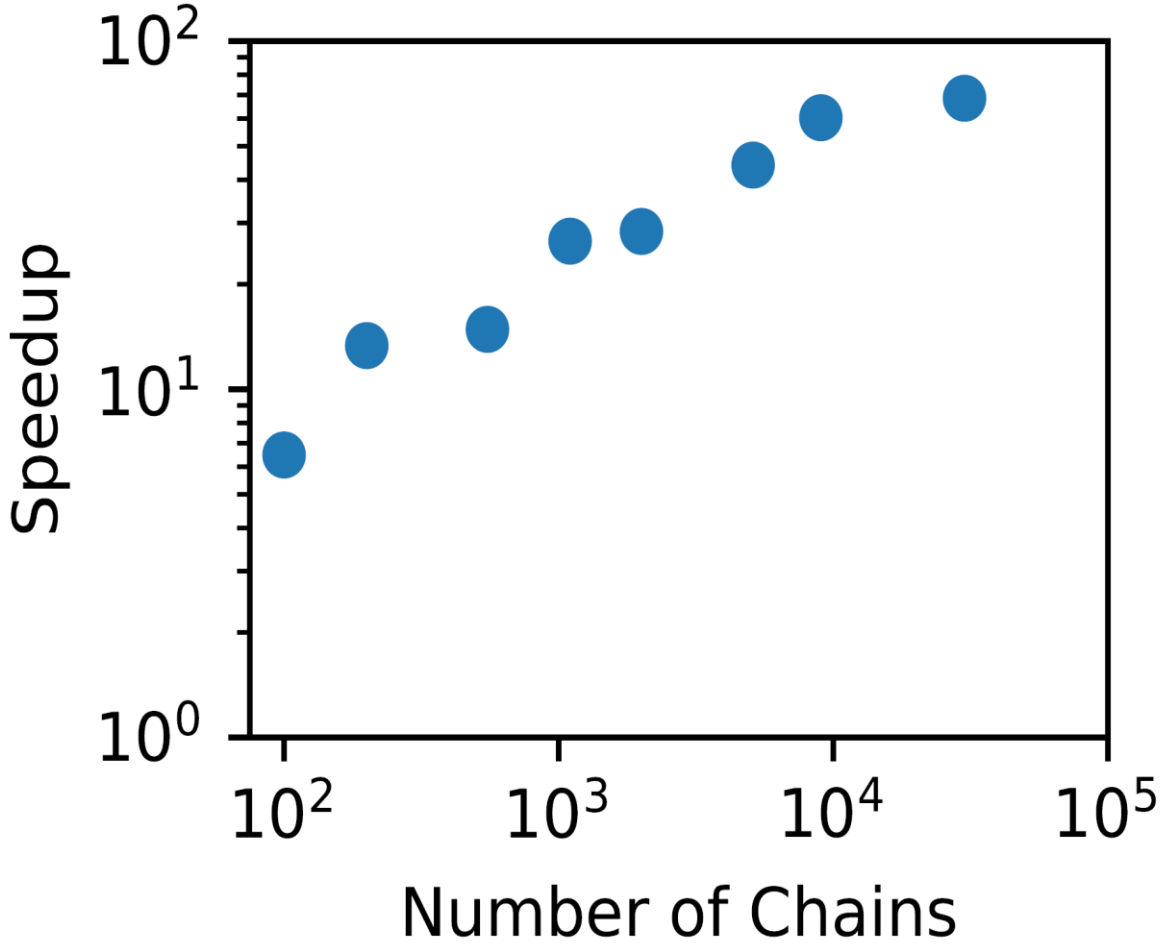
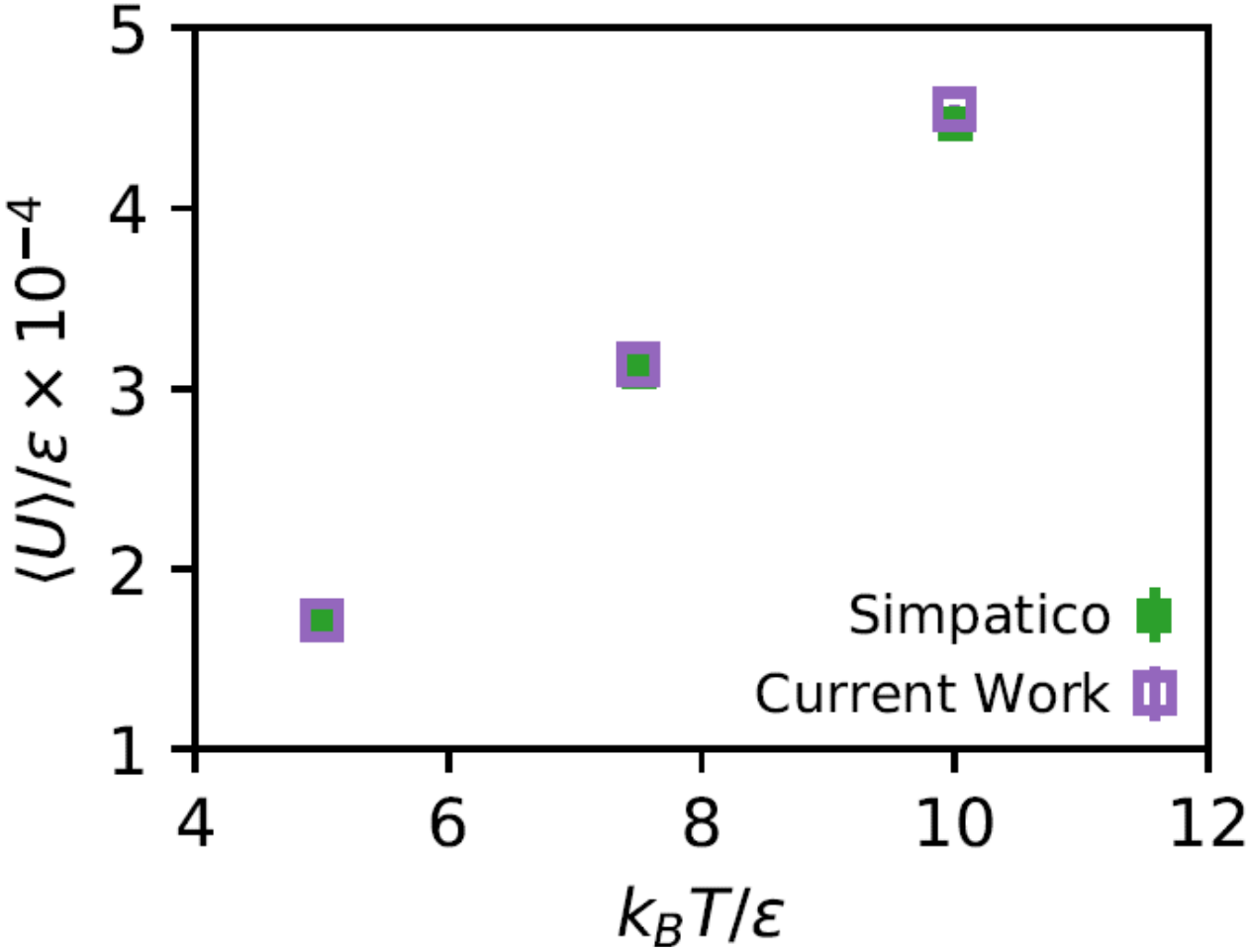


Simpatico [Computer software]. (2010).



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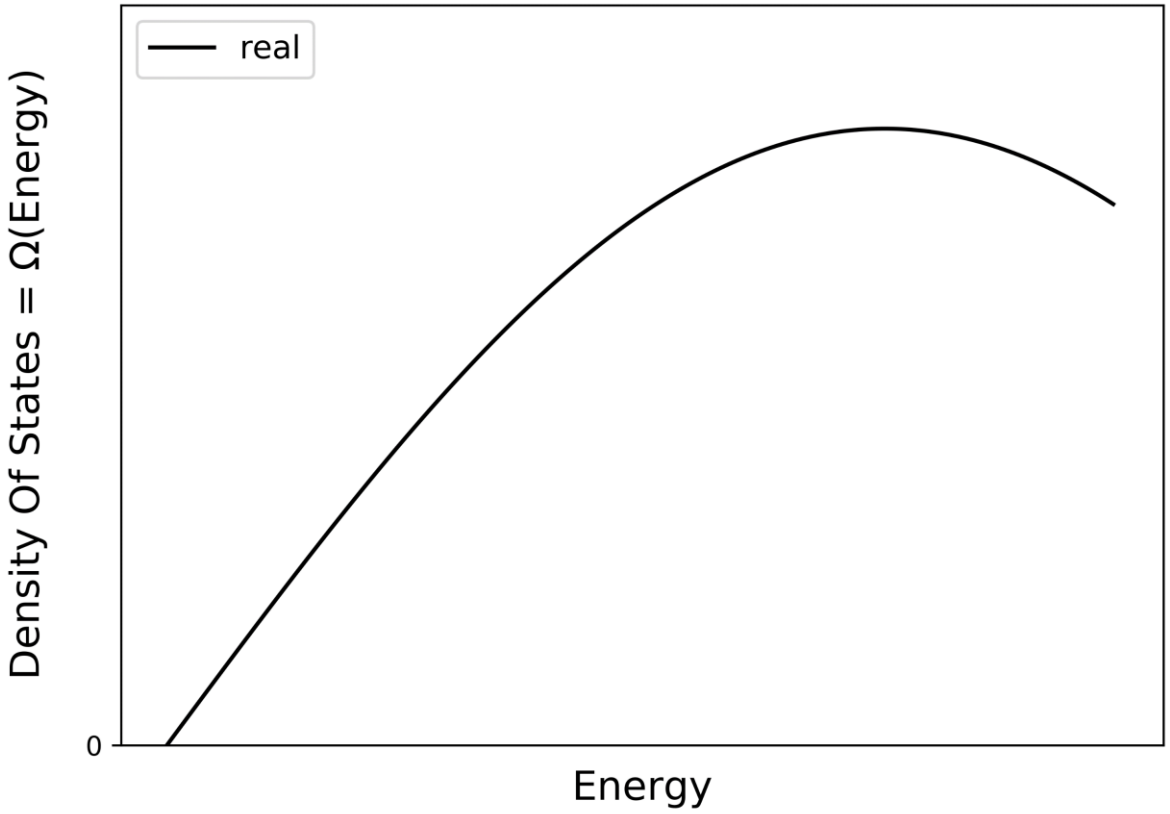

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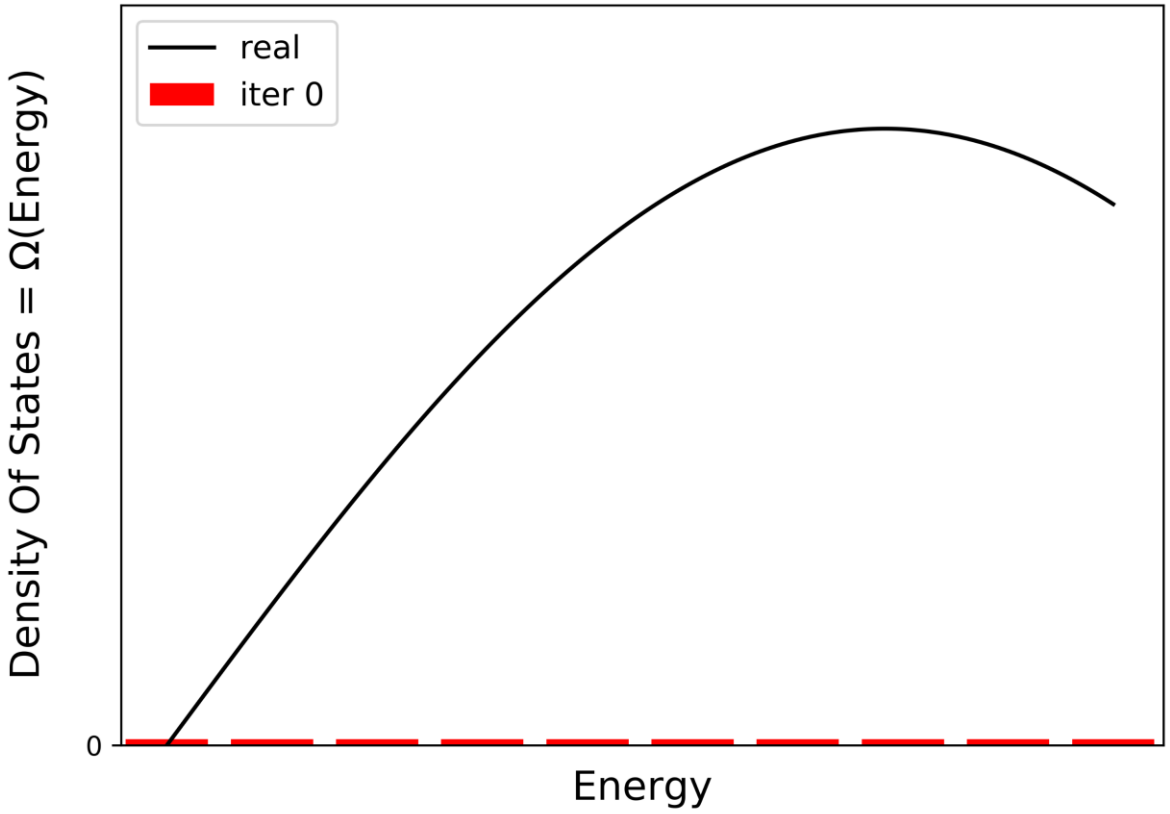

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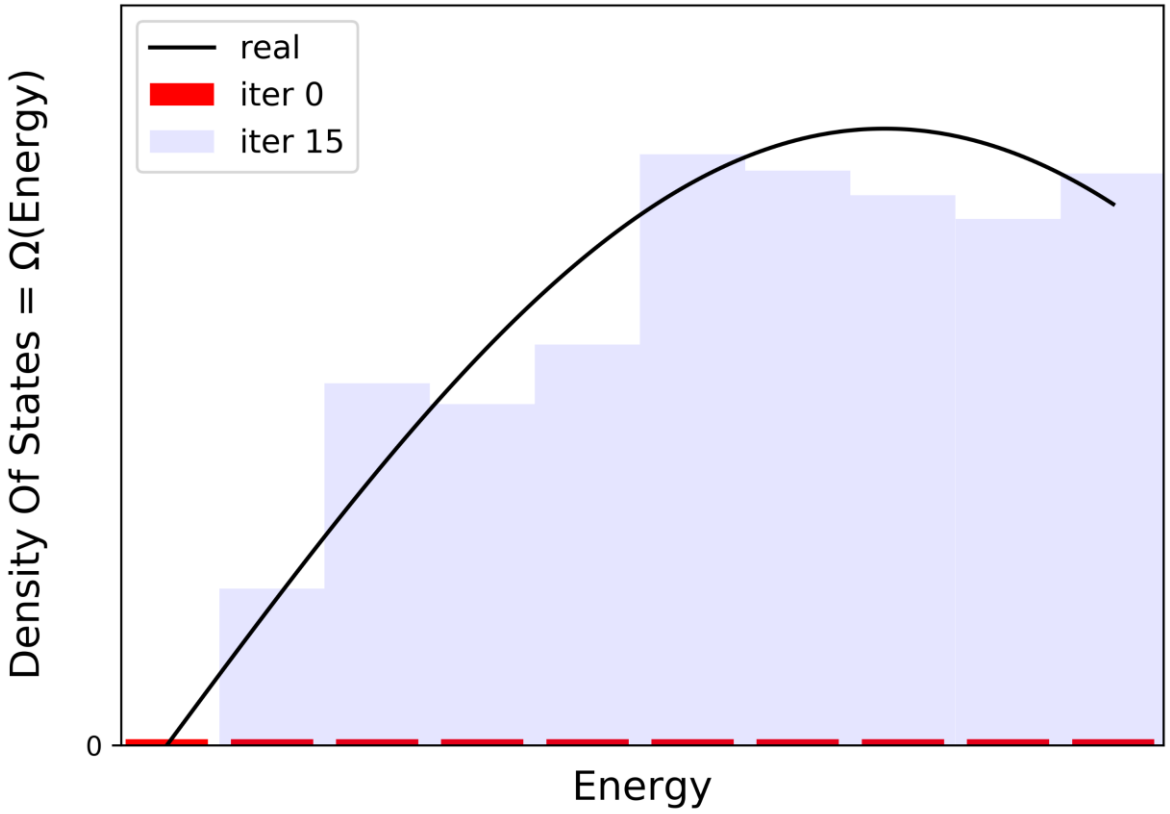

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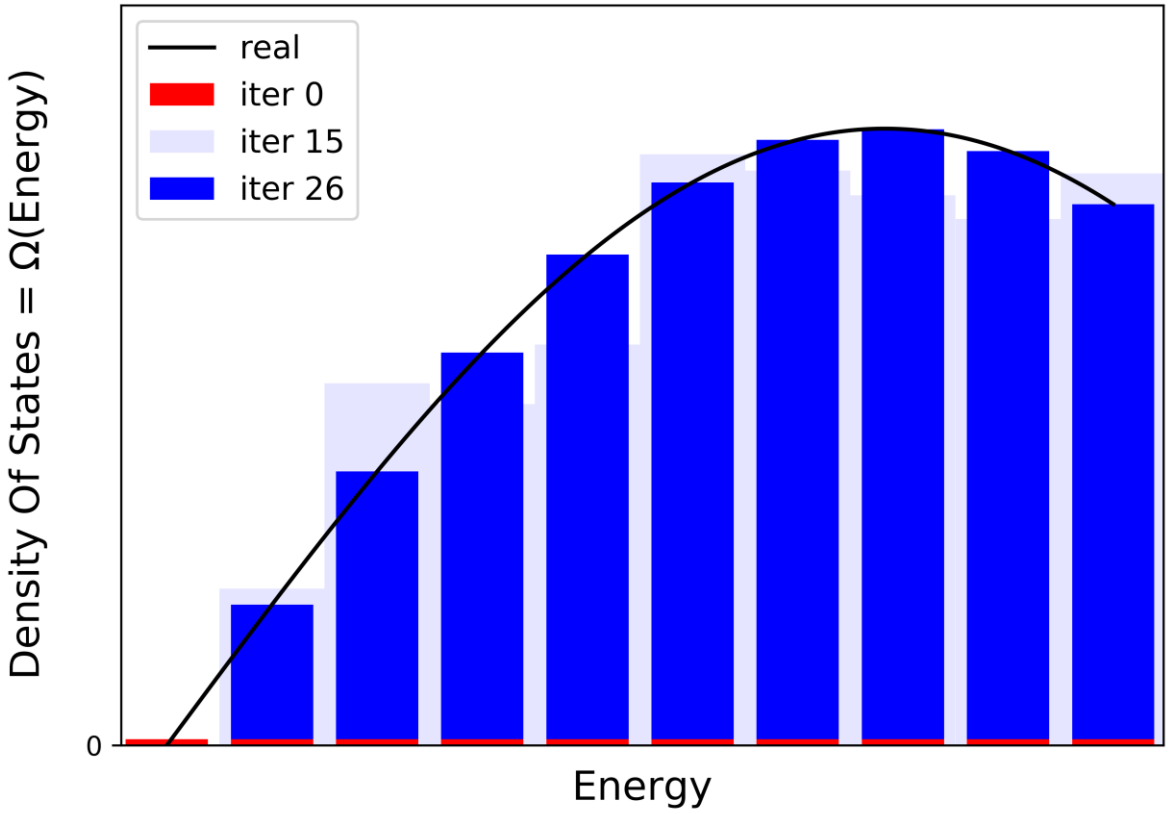

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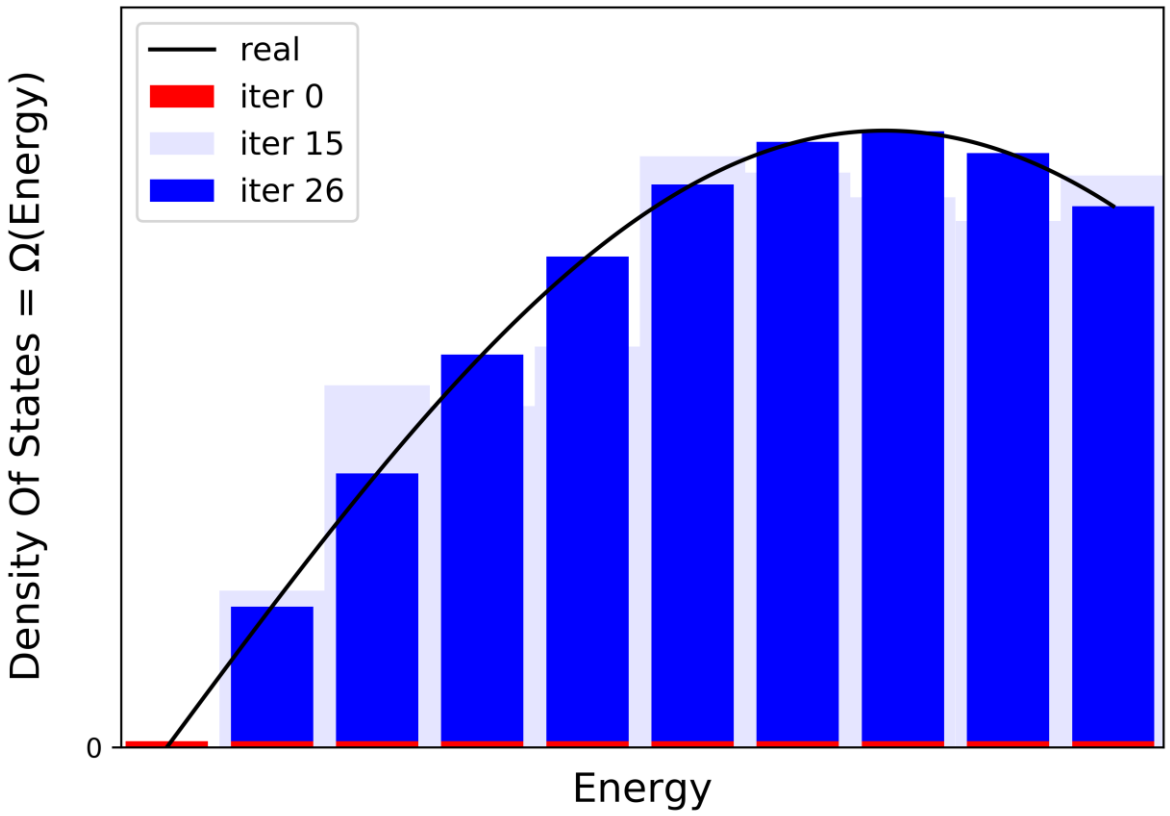

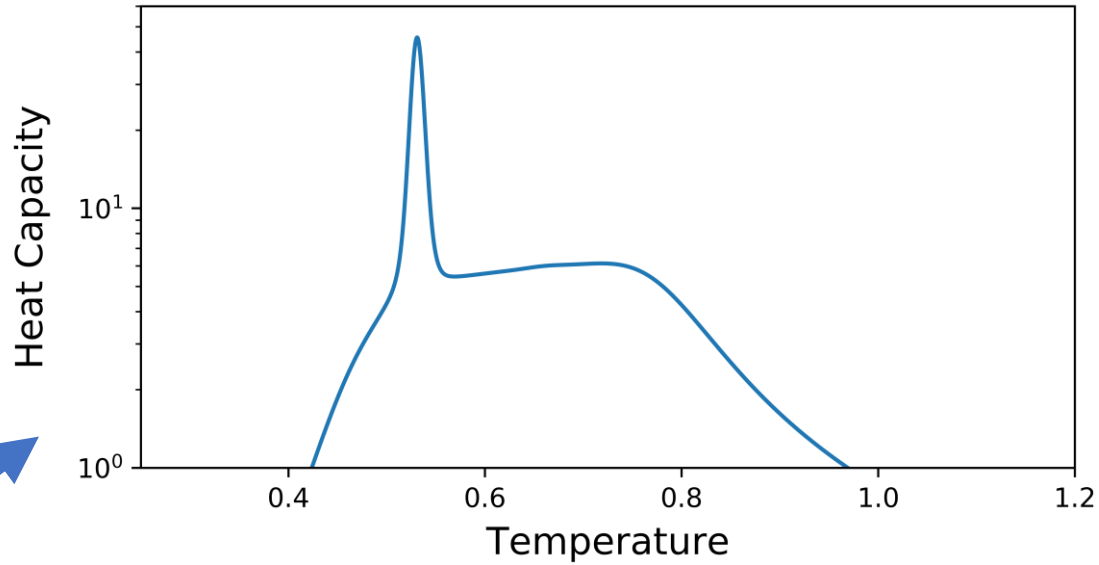

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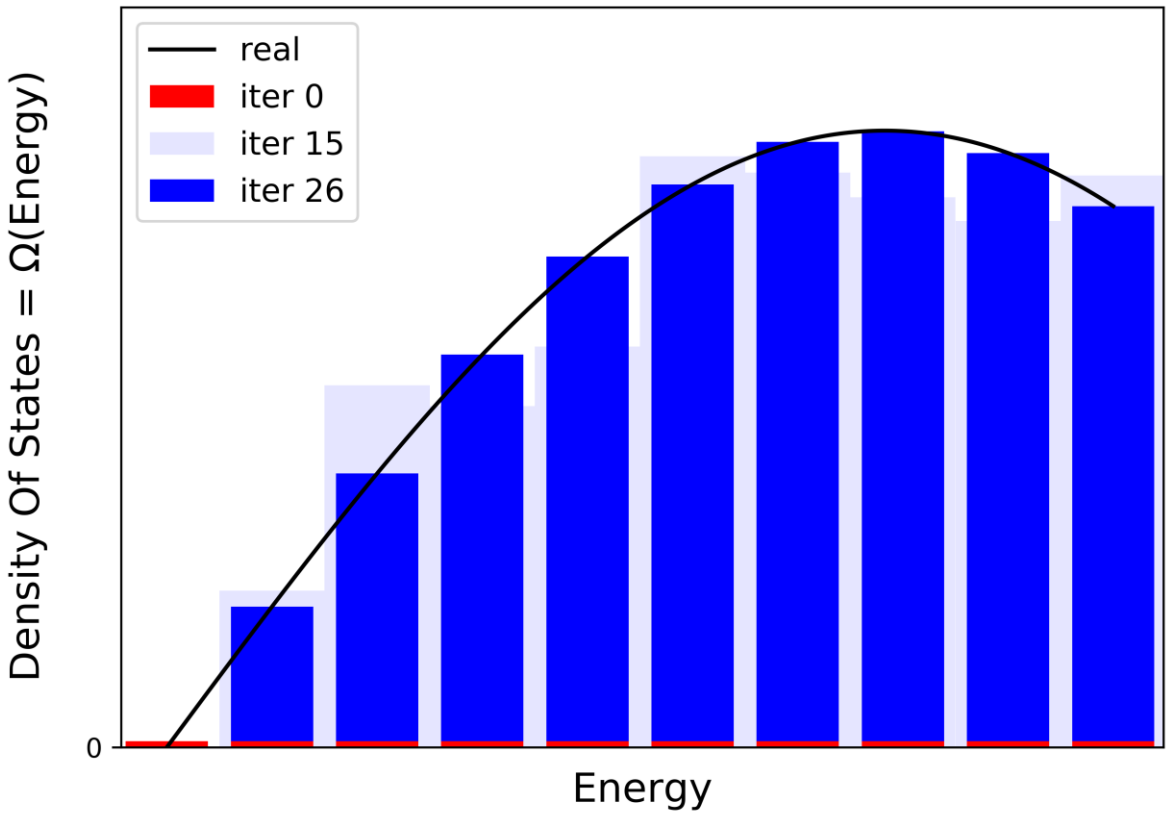

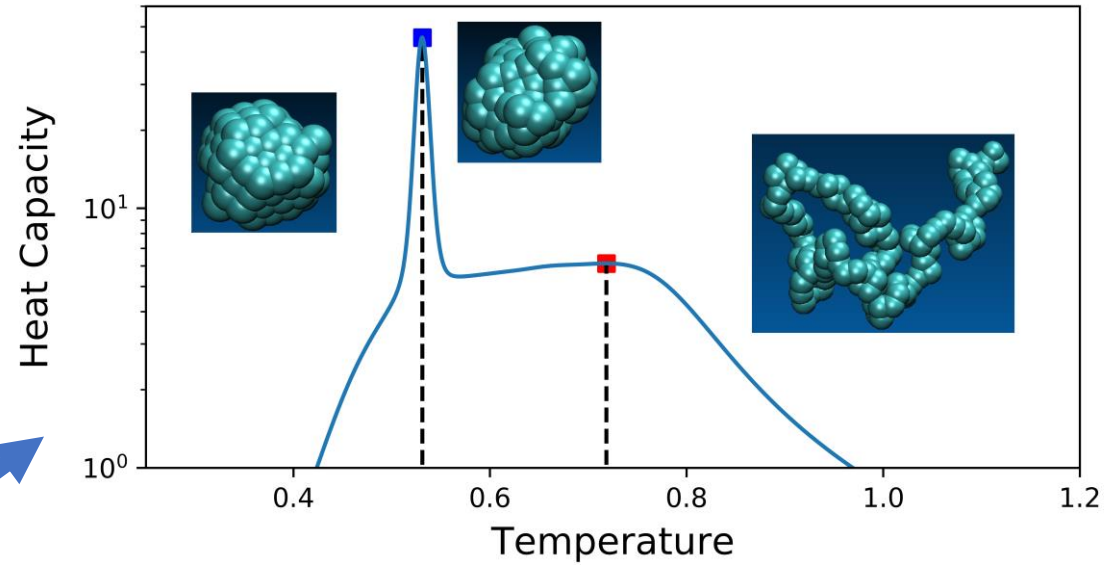

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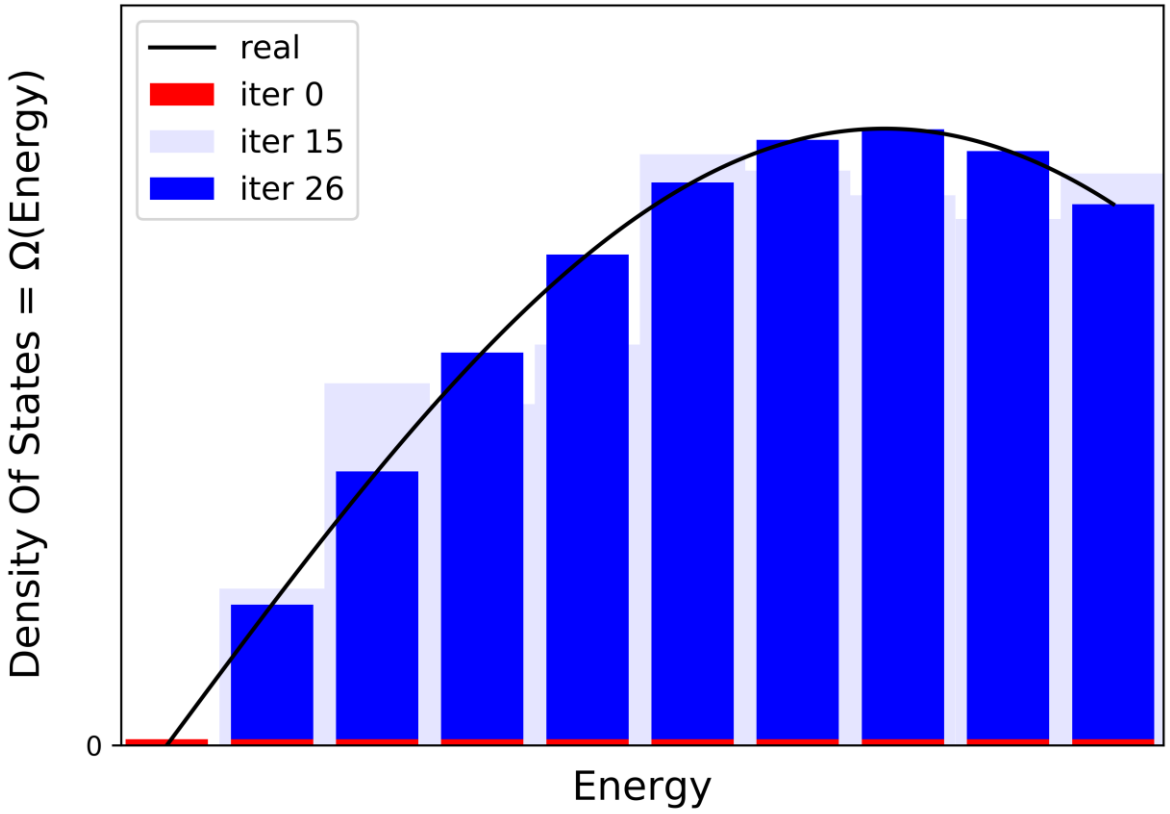

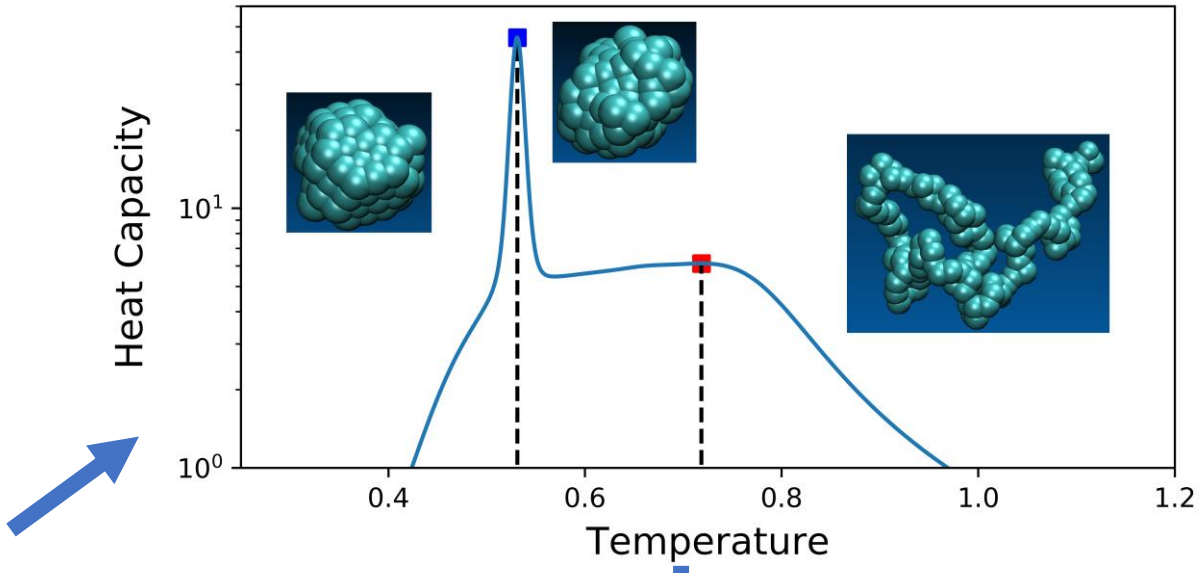

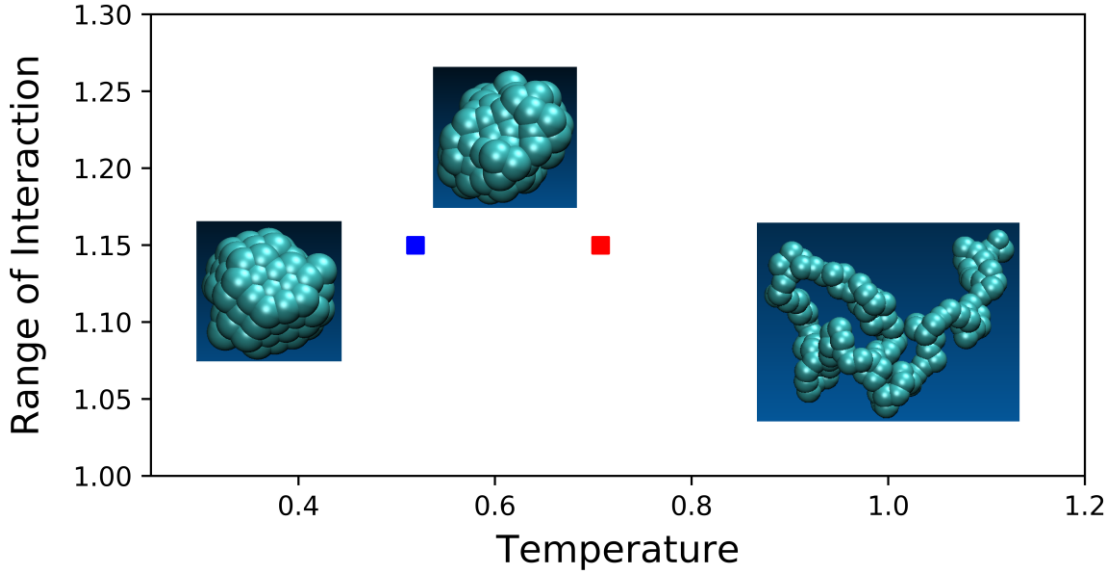

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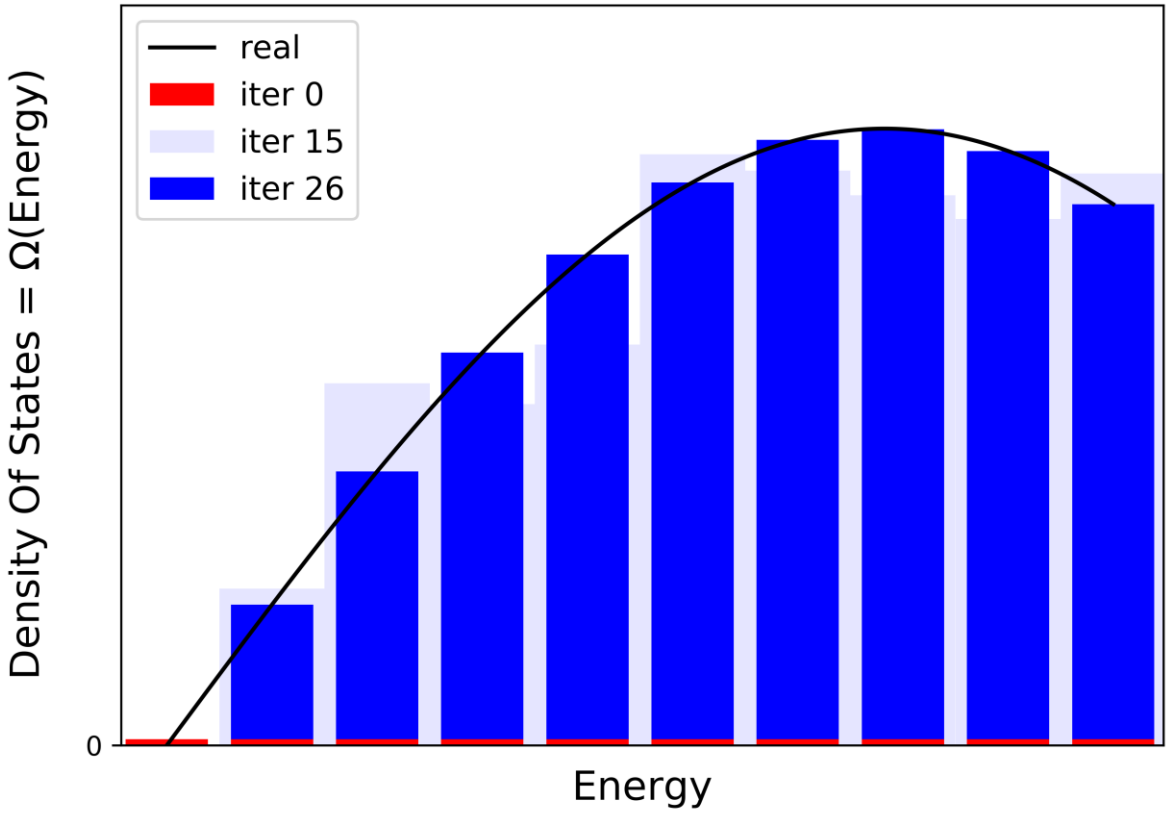
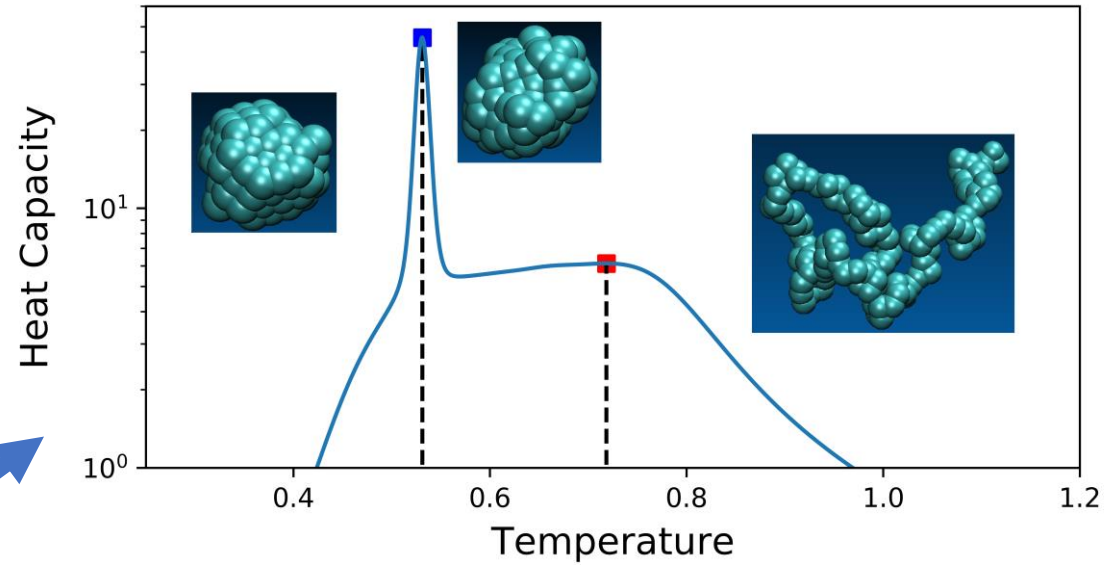
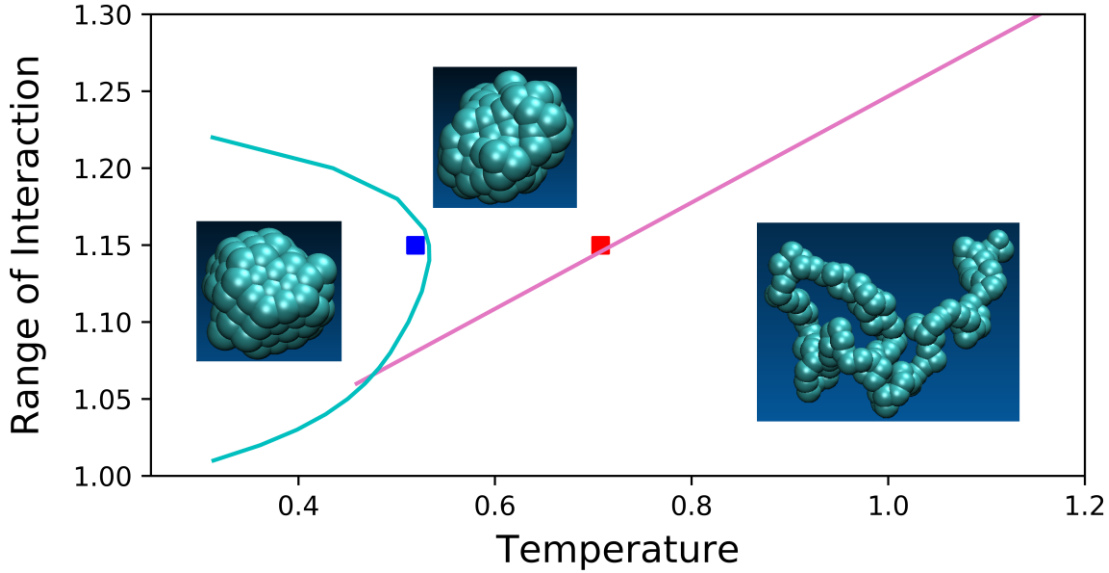
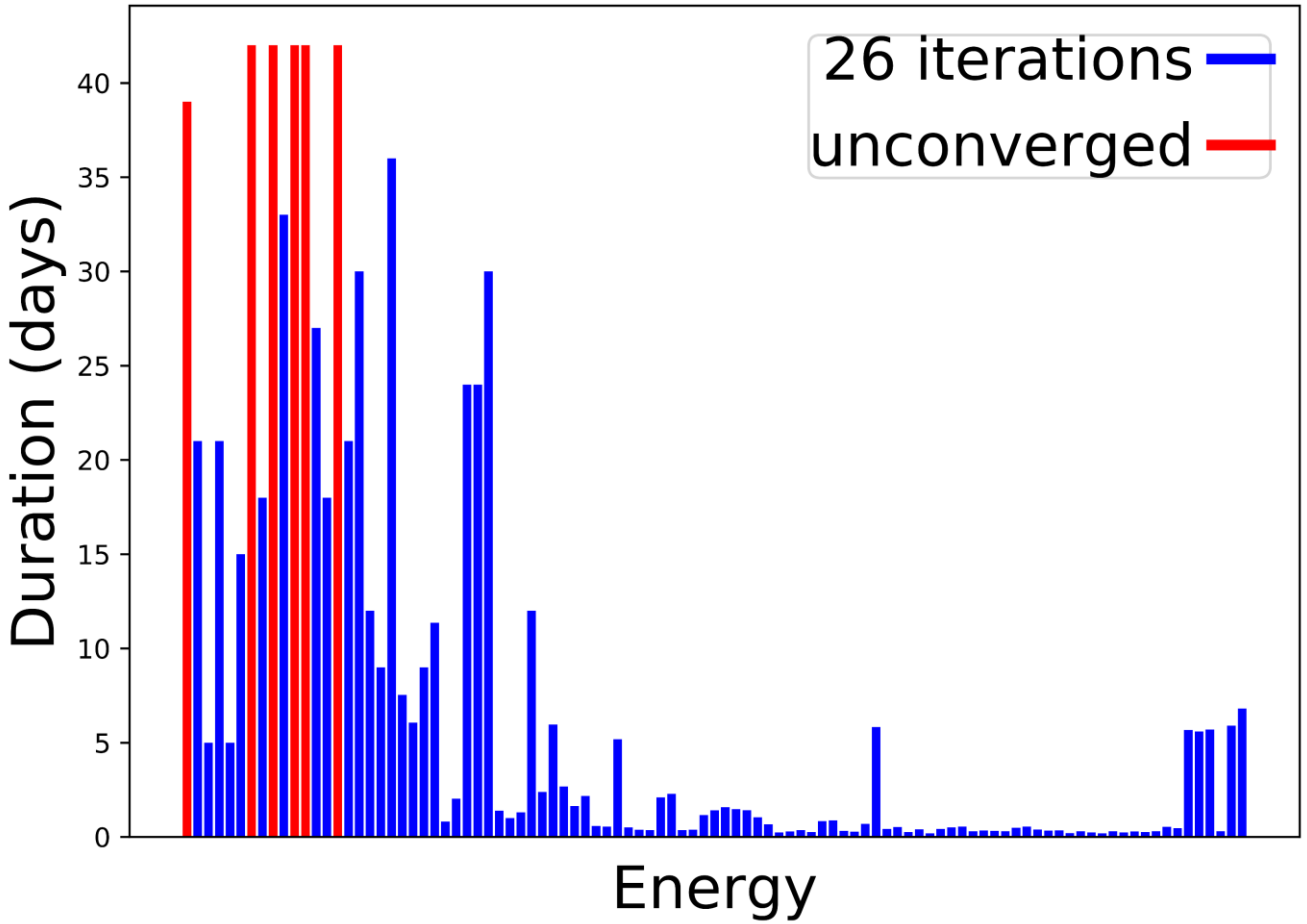






Simulating Wang-Landau for polymer melts (2000 chains, 100 beads) is more difficult than anticipated



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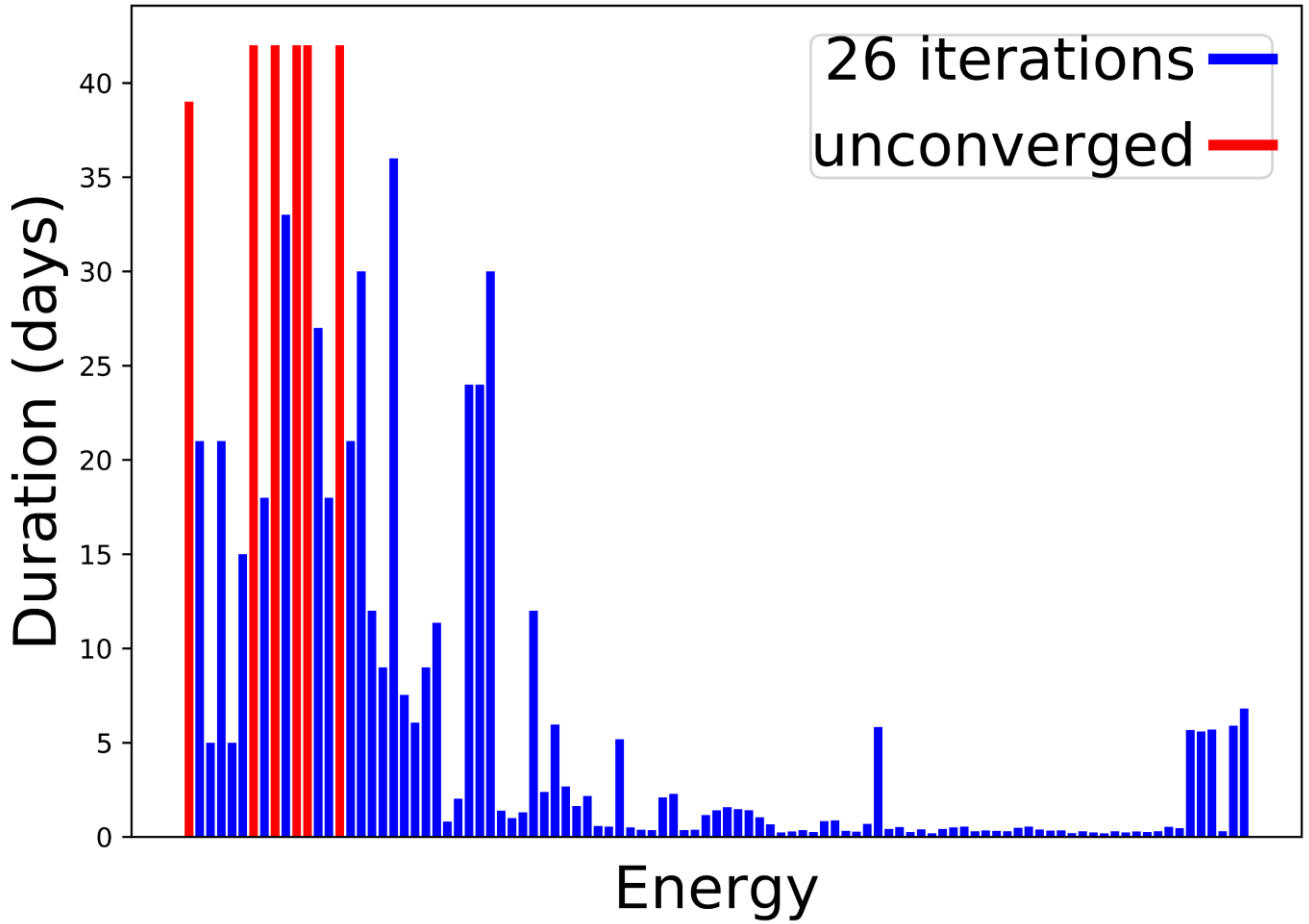

- Shakirov and Paul *Phys Rev E* (2018) DOS spans 5600 orders of magnitude and "…sets the limit…one can study… 1 year."
- Our system spans **190,000 orders of magnitude!**

Conclusion and Future Work

Current Work

- Built a **GPU-accelerated WLMC** algorithm from scratch
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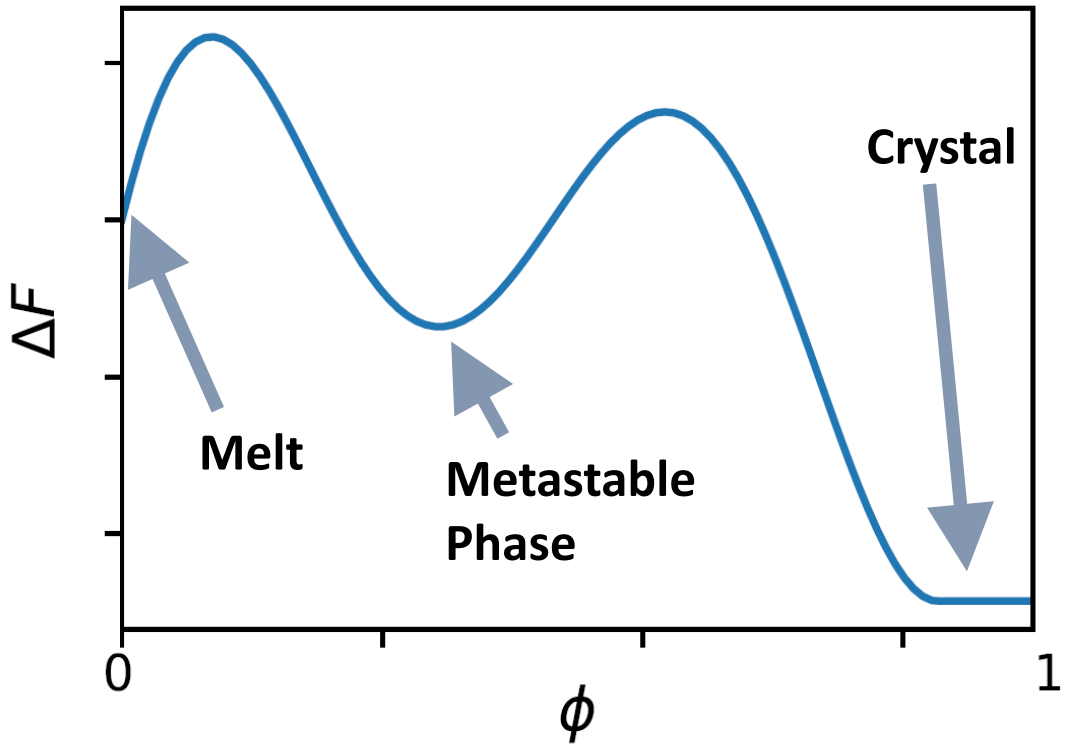

Acknowledgements

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  (PRF# 59244-DN16**)**

- BYU **Board of Trustees**

- BYU **Office of Research Computing**

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