Characterizing the Uncertainties in Non-Equilibrium MD for Thermal Transport

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BACKGROUND

- Classical MD is used to investigate heat transfer dominated by phonon-phonon interactions in material systems.
  - Commonly applied to study non-metallic systems like C, Si, and Ge.
- Typically conducted under equilibrium conditions characterized by thermodynamic ensembles like NVT, NVE, NPT, and μVT.
- Non-Equilibrium MD involves setting up thermostats in different regions to establish temperature gradients.
  - Thermostatting introduces errors.

WHY MD?

☛ Enables simulation of much larger systems compared to DFT in a reasonable amount of time.

☛ Trends from MD are useful despite possible errors in estimates.

Plan

Part I: Forward Problem:

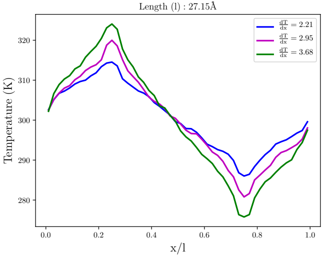
- Investigate error in predictions due to size of the material system and fluctuations in thermal gradient.
  - Construct a response surface for the error.
- Characterize the impact of uncertainty in inter-atomic potential on predictions.
  - Efficient construction of surrogates using NISP: aPSP, Sparse Basis, Active Subspaces
  - Examine sensitivity of estimates on parameters.

Part II: Inverse Problem:

- Calibrate critical parameters associated with the potential function in a Bayesian setting.
  - Exploit the error response surface from Part I.

NEMD ON A SILICON BAR

Lattice Constant (Å)	5.43
W, H (Å)	117.94, 117.94
Temperature (K)	300
Δt (ps)	0.0005
BC	Periodic
Structure	Diamond
Potential	Stillinger-Weber



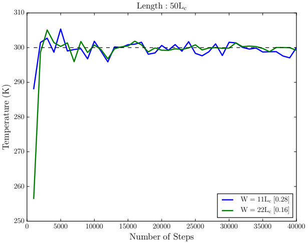
OPTIMIZE FOR EQUILIBRATION

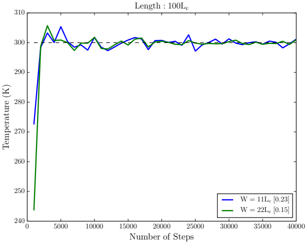
NVT	$\rightarrow$	NVE	$\rightarrow$	NVE
[Eqilibrate system to 300 K]		[Equilibrate thermostats]		[Generate Data]

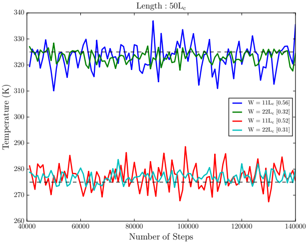
INITIAL RUNS:

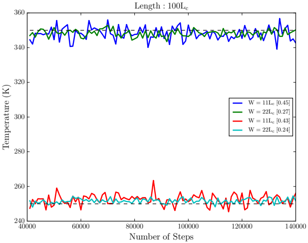
- Determine the time steps needed for equilibration at all stages.
- Select a reasonable width and height for the Si bar.
  - Need to be in a regime where changes in width and height do not impact estimates significantly.

SELECTION OF WIDTH

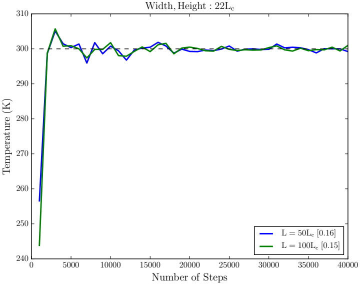


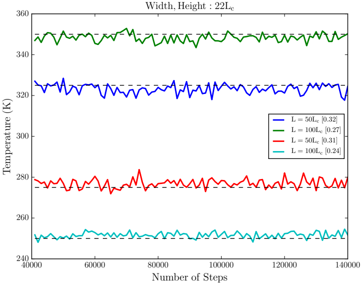






Selection of Width





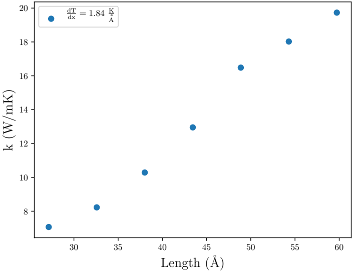
- Norm of the fluctuations (NF) is computed using:

$$NF = \frac{1}{N}\left[\sum_k \left(T_k - T_{\{nvt,nve\}}\right)^2\right]^{\frac{1}{2}}$$

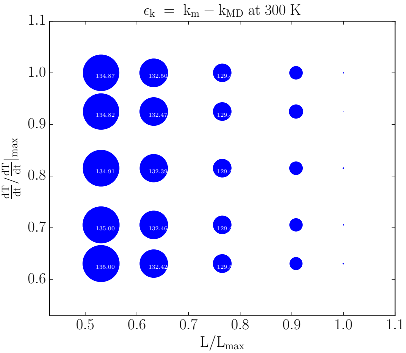
- At $W = 21.72L_c$, the effect of length on fluctuations seems to be minuscule.

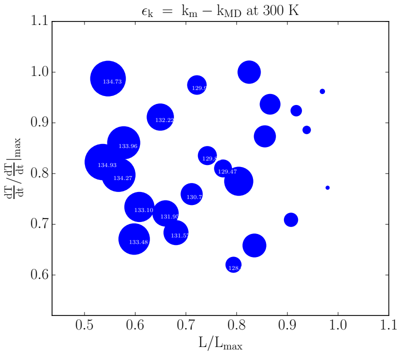
NEED A SURROGATE?

- OBJECTIVE: Forward UQ, Sensitivity Analysis, calibration, Design
- COMPUTATIONAL EFFORT: Simulations are computationally intensive.
- ACCURACY: Can a surrogate represent the observable with reasonable accuracy in the domain of interest?



MODEL REALIZATIONS



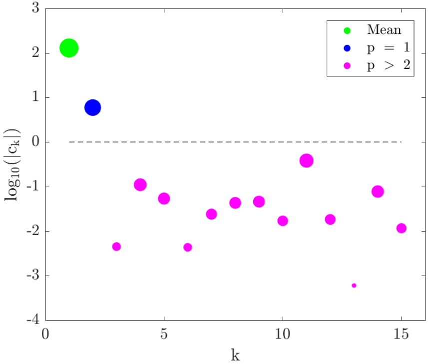


- Model realizations at Gauss-Legendre quadrature nodes are used to construct the PC surrogate.

PC Expansion

$$\boxed{\kappa = \sum_j c_j \Psi_j(\xi_1, \xi_2)}$$

κ: Thermal Conductivity, j: Multi-index



$$L: \mathcal{U}[50L_c, 100L_c]\ (\text{Å}) \quad \rightarrow \quad \xi_1 : \mathcal{U}[-1, 1]$$

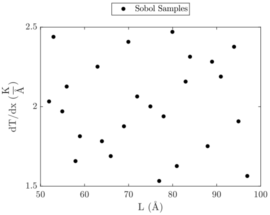
$$\frac{dT}{dx}: \mathcal{U}[1.5/L_c, 2.5/L_c]\ (\tfrac{K}{\text{Å}}) \quad \rightarrow \quad \xi_2 : \mathcal{U}[-1, 1]$$

RESPONSE SURFACE: $\epsilon(L, \frac{dT}{dx})$

$$\epsilon = |\kappa_m - \kappa_{NEMD}|$$

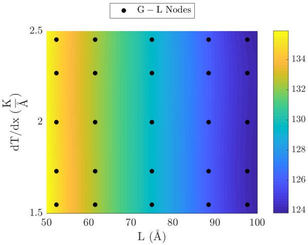
κ_m: Measured Thermal Conductivity
κ_{NEMD}: MD Prediction

ACCURACY:

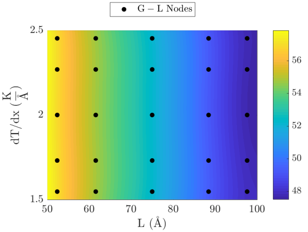


$$\epsilon = \frac{\left[\sum_j (\mathcal{G}_M - \mathcal{G}_{PCE}))^2\right]^{\frac{1}{2}}}{\left[(\mathcal{G}_M)^2\right]^{\frac{1}{2}}} \approx 1.8 \times 10^{-3}$$

$\mathcal{G}_M$: Model Output $\mathcal{G}_{PCE}$: PCE Estimate



(a) 300 K



(b) 500 K

NEXT STEPS

- Dimension reduction through Local Sensivity Analysis.
- Estimate posterior distributions for significant potential function parameters.
- Discover a potential active subspace.