

Characterizing Errors and Uncertainties in NEMD Simulations of Phonon Transport

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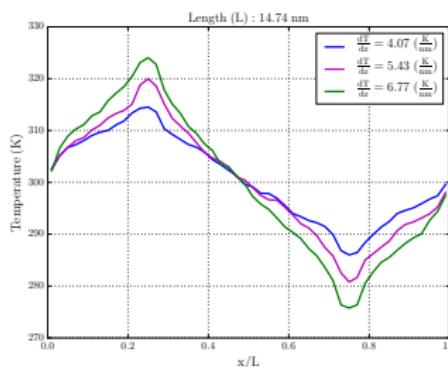
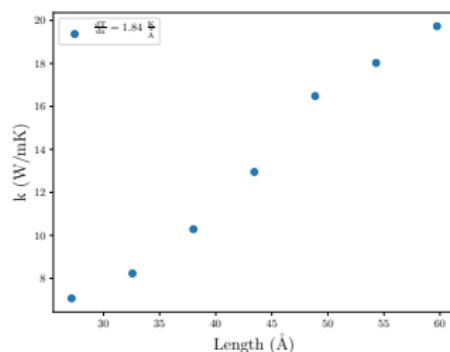
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- System subjected to a temperature gradient using thermostats.
- **Steady-state** thermal energy exchange between the thermostats is used in Fourier's law to estimate bulk thermal conductivity (κ).
- Estimates are **uncertain** and severely **under-predicted**.
 - ▶ Simulation length-scales \ll phonon mean free path.
 - ▶ Thermostats reduce correlation b/w vibration modes.

SOURCES OF UNCERTAINTY



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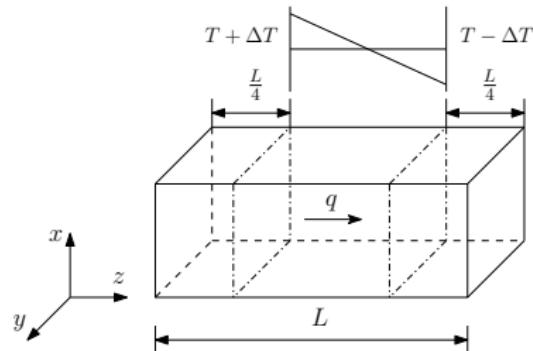
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- Calibrate the *important* parameters in a Bayesian setting.

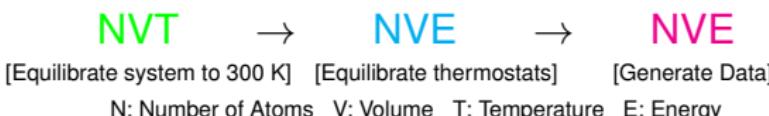
PROBLEM SET-UP

Lattice Constant, a (\AA)	5.43
Width, Height (\AA)	$22a$
Δt (ps)	0.0005
Boundary Condition	Periodic
Lattice Structure	Diamond
Inter-atomic Potential	Stillinger-Weber



PROBLEM SET-UP

- STAGES:



- OBSERVABLE: Avg. energy exchange b/w thermostats (q)
- QoI: Bulk thermal conductivity (κ)

$$\kappa = \frac{q}{\left| \frac{dT}{dz} \right|}$$

- Appropriate selection of width, height, and simulation-time was ensured.

RESPONSE SURFACE: DISCREPANCY

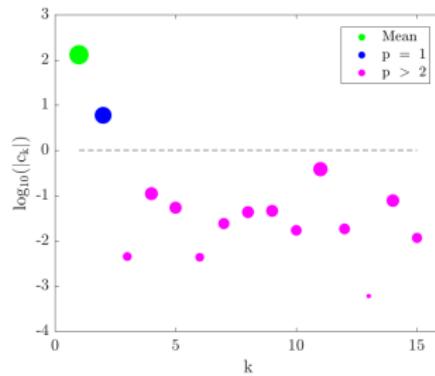
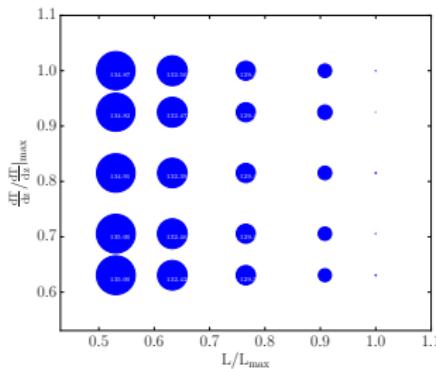
- PC representation of the discrepancy, ϵ_d :

$$\epsilon_d \approx \epsilon_d^{\text{PCE}} = \sum_{k \in \mathcal{I}} c_k(T) \Psi_k(\xi(\theta))$$

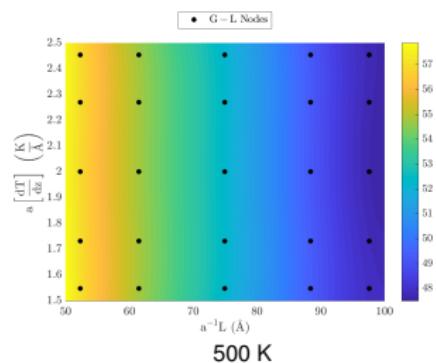
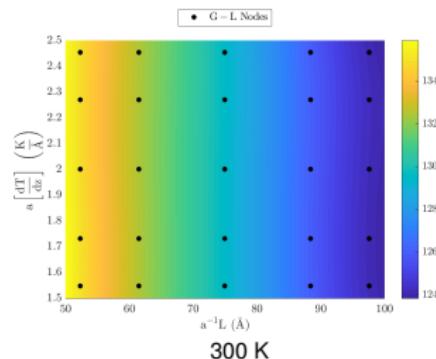
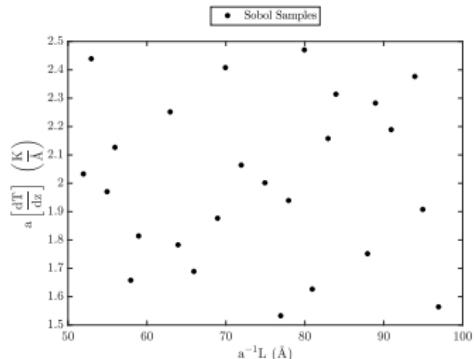
- Uncertain parameters, $\theta: \{L, \frac{dT}{dz}\}$

$$L \sim \mathcal{U}[50a, 100a] (\text{\AA})$$

$$\frac{dT}{dz} \sim \mathcal{U}\left[\frac{1.5}{a}, \frac{2.5}{a}\right] \left(\frac{K}{\text{\AA}}\right)$$

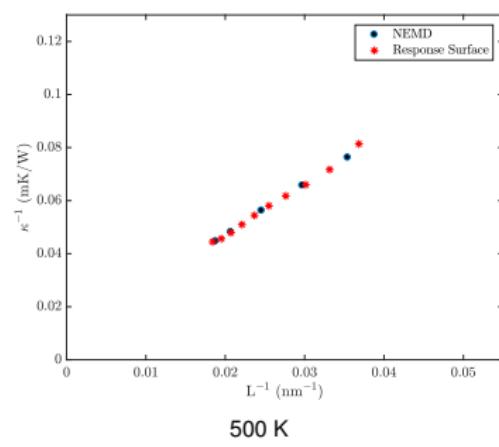
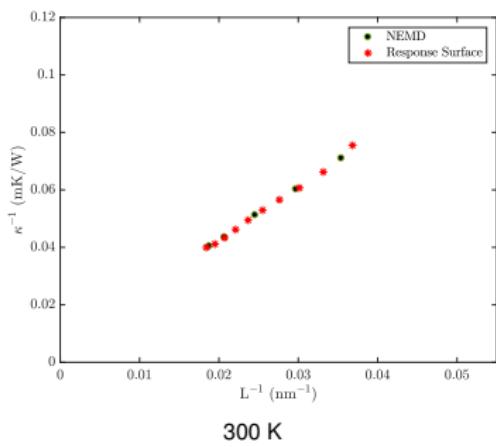


VERIFICATION



$$\varepsilon_{L-2} = \frac{\left[\sum_j (\epsilon_{d,j}^{\text{MD}} - \epsilon_{d,j}^{\text{PCE}})^2 \right]^{\frac{1}{2}}}{\left[\sum_j (\epsilon_{d,j}^{\text{MD}})^2 \right]^{\frac{1}{2}}} \sim \mathcal{O}(10^{-3})$$

DIRECT METHOD



- y-intercept of the straight line fit to the above plot provides an estimate of the bulk thermal conductivity.

THE STILLINGER-WEBER POTENTIAL

FUNCTIONAL FORM

$$\Phi(1, \dots, N) = \sum_{i,j(i < j)} v_{ij}^{(2)}(A, B, p, q, \alpha) + \sum_{i,j,k(i < j < k)} v_{ij}^{(3)}(\lambda, \gamma)$$

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SHORTCOMINGS

- Inadequate functional representation.
- Estimates based on a limited search.
- Do not account for measurement error and MD noise.
- Parametric uncertainties were not considered.

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MITIGATION STRATEGIES

- Sensitivity Analysis of the parameters.
- Forward propagation of the uncertainty.
- Bayesian calibration of the key parameters.

SENSITIVITY ANALYSIS

CHALLENGES

- Global sensitivity analysis is computationally intractable.
- Perturbing potential parameters → loss of structural integrity.

SENSITIVITY ANALYSIS

- Derivative-based sensitivity measures (DGSM) are estimated to identify *unimportant* parameters.

$$\mu_i = \mathbb{E} \left[\left(\frac{\partial G(\theta)}{\partial \theta_i} \right)^2 \right]$$

 \rightarrow

$$\hat{c}_i \mu_i = \frac{c_i \mu_i}{\sum_i c_i \mu_i}$$

 \rightarrow

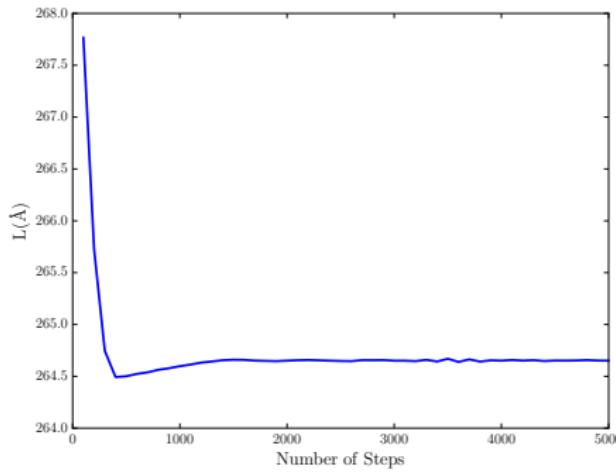
$$\mathcal{T}_i \leq \frac{c_i \mu_i}{V} (\propto \hat{c}_i \mu_i)$$

- DGSM estimates are used to compute upper bound on Sobol total effect index (\mathcal{T}_i).

SENSITIVITY ANALYSIS

- Add an NPT ensemble prior to NVT and run it for a sufficiently long time.

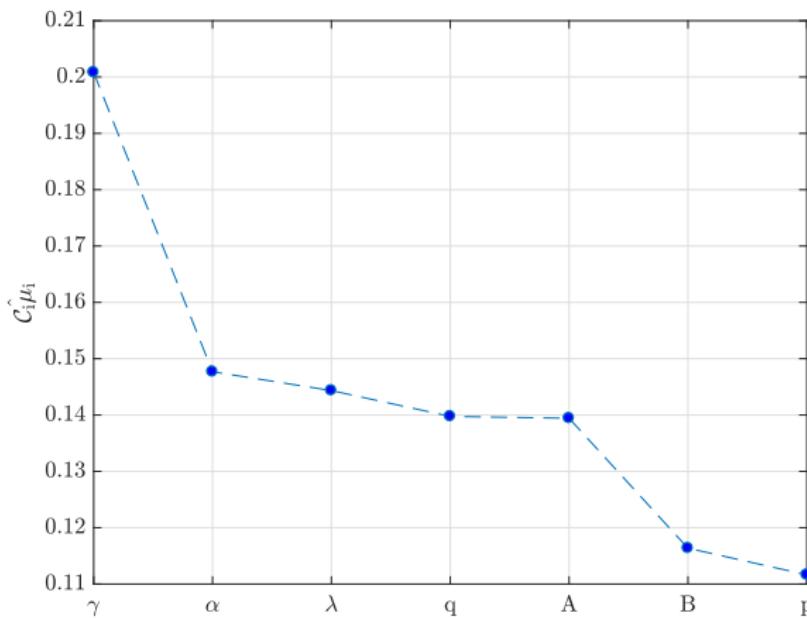
NPT → NVT → NVE → NVE
[Relax system length] [Equilibrate system to 300 K] [Equilibrate thermostats] [Generate Data]
N: Number of Atoms P: Pressure V: Volume T: Temperature E: Energy



PARAMETER SCREENING

- 1 Generate n_1 points in \mathbb{R}^d .;
% d: Number of parameters;
- 2 Perturb each point along the d directions to obtain a set of $n_1(d+1)$ points.;
- 3 Compute μ_i using model evaluations at the $n_1(d+1)$;
- 4 Determine initial ranks, \mathcal{R}^{old} based on $\hat{\mathcal{C}_i\mu_i}$ values for θ_i ;
- 5 Set $k = 1$;
- 6 **repeat**
- 7 Generate n_k new points in \mathbb{R}^d .;
- 8 Perturb each point along the d directions to obtain a set of $n_k(d+1)$ points.;
- 9 Compute and store model evaluations at the $n_k(d+1)$ points.;
- 10 Compute μ_i using prior model evaluations at $(d+1)(n_1 + \sum_j^k n_j)$ points.;
- 11 Determine new ranks, \mathcal{R}^{new} based on updated $\hat{\mathcal{C}_i\mu_i}$ values.;
- 12 Compute $max_pdev = \max\left(\frac{|\mu_{i,k} - \mu_{i,k-1}|}{\mu_{i,k-1}}\right)$.;
% max(pdev: Maximum percentage deviation in μ_i between successive iterations.);
- 13 Set $k = k + 1$;
- 14 **until** ($\mathcal{R}^{new} \neq \mathcal{R}^{old}$ **or** $max_pdev > \tau$);

PARAMETER SCREENING



REDUCED ORDER SURROGATE

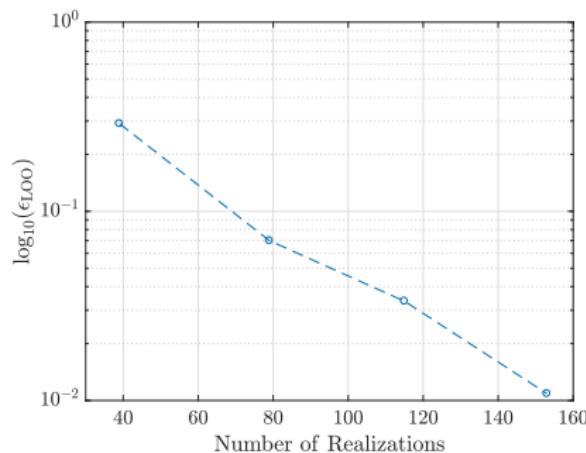
- PCE for κ :

$$\kappa = \sum_{s \in \mathcal{A}} c_s(T) \Psi_s(\xi)$$

$$\xi : \{\xi_1(A), \xi_2(q), \xi_3(\alpha), \xi_4(\lambda), \xi_5(\gamma)\}$$

- Regularized least-squares minimization problem:

$$\hat{c} = \arg \min_{c \in \mathbb{R}^{|\mathcal{A}|}} \mathbb{E} \left[\left(c^T \Psi(\xi(\theta)) - \kappa_{\theta}^{\text{MD}} \right)^2 \right] + \lambda \|c\|_1$$

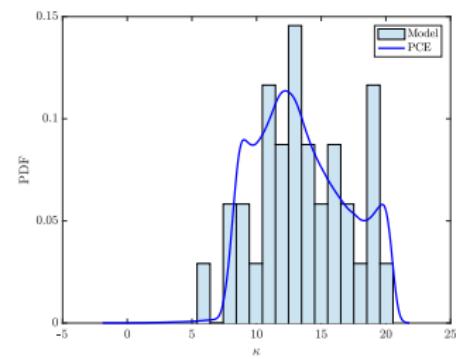


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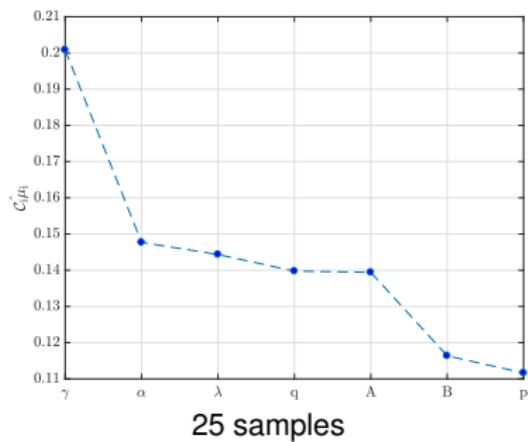
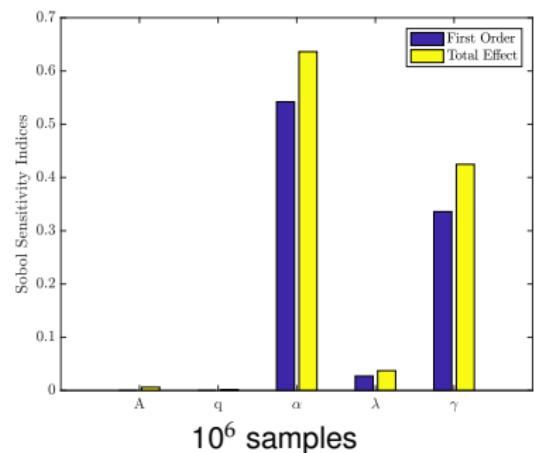
L-2 NORM

$$\epsilon_{L-2} = \frac{\left[\sum_{i=1}^{N=25} \left(\mathcal{M}(\boldsymbol{\theta}_{7D}^{(i)}) - \mathcal{M}^{PCE}(\boldsymbol{\theta}_{5D}^{(i)}) \right)^2 \right]^{\frac{1}{2}}}{\left[\sum_{i=1}^N \left(\mathcal{M}(\boldsymbol{\theta}_{7D}^{(i)}) \right)^2 \right]^{\frac{1}{2}}} \rightarrow \mathcal{O}(10^{-2})$$

PROBABILISTIC



GLOBAL SENSITIVITY ANALYSIS



BAYESIAN CALIBRATION

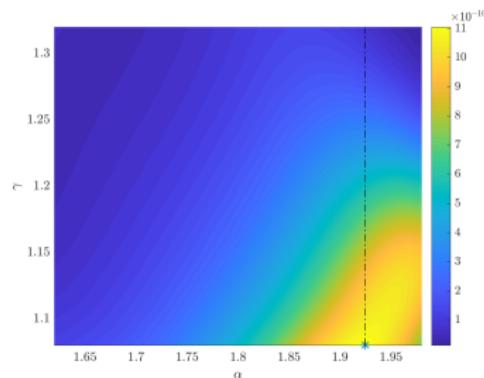
- Bayes' rule:

$$\mathcal{P}(X|Y) \propto \mathcal{P}(Y|X)\mathcal{P}(X)$$

$$X : \{\alpha, \gamma\}$$

- Likelihood, $\mathcal{P}(Y|X)$:

$$\mathcal{P}(Y|X) = \frac{1}{\sqrt{2\pi\sigma^2}} \exp \left[-\frac{(\kappa_E - \kappa_{MD})^2}{2\sigma^2} \right]$$



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- Calibration should be performed in a Bayesian setting to account for all possible errors and uncertainties.
- Strategies presented can be easily extended to other systems and potentials.