

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 μ VT.
- 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

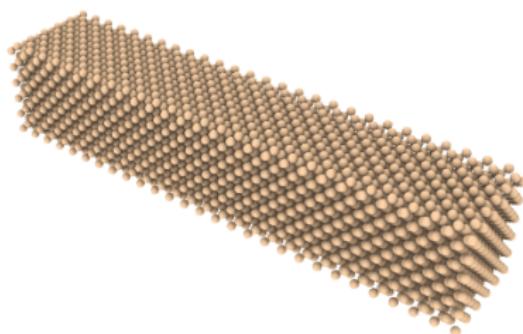
Part I: FORWARD PROBLEM:

- 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.
 - ▶ Dimension reduction using derivative-based sensitivity measures.
 - ▶ Construction of PCE in the reduced space.
 - ▶ Forward Propagation of uncertainty.

Part II: INVERSE PROBLEM:

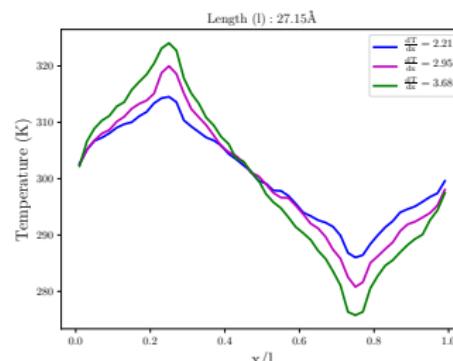
- Calibrate critical parameters associated with the potential function in a Bayesian setting using experimental data.

NEMD ON A SILICON BAR



$L = 50a$, $N \approx 200000$ atoms

Lattice Constant, a (\AA)	5.43
W, H (\AA)	$22a, 22a$
Temperature (K)	300
Δt (ps)	0.0005
BC	Periodic
Structure	Diamond
Potential	Stillinger-Weber



DIRECT METHOD

NVT

→

NVE

→

NVE

[Eqilibrate system to 300 K] [Equilibrate thermostats] [Generate Data]

N: Number of Atoms V: Volume T: Temperature E: Energy

OBSERVABLE: Average energy exchange b/w thermostats (q)

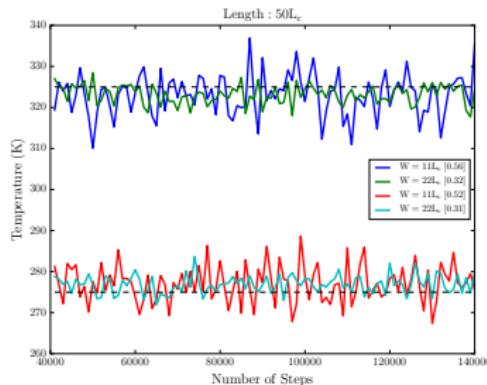
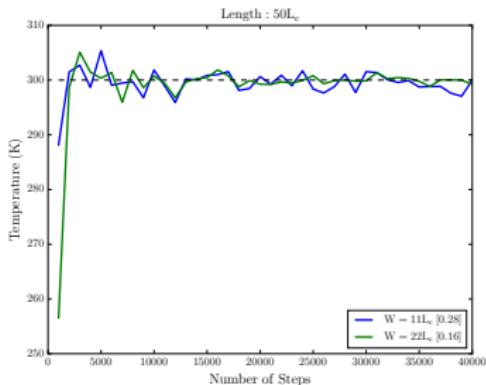
QoI: Bulk thermal conductivity (κ)

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

INITIAL RUNS:

- Determine the time steps needed for equilibration.
- Select a **reasonable** width and height for the Si bar.
 - ▶ Small fluctuations due to changes in height and width.

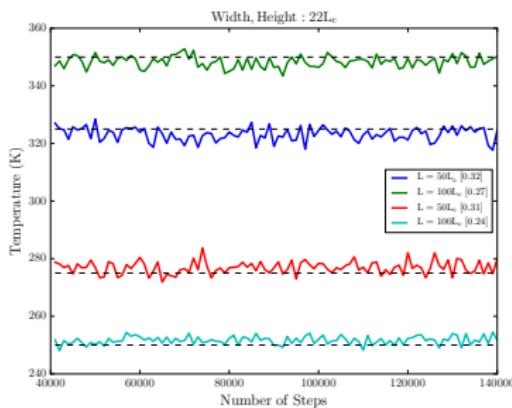
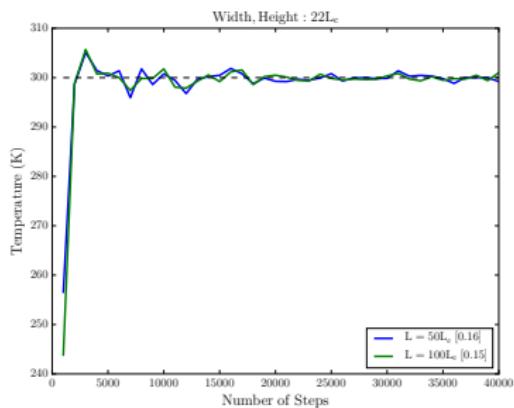
SELECTION OF WIDTH



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

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

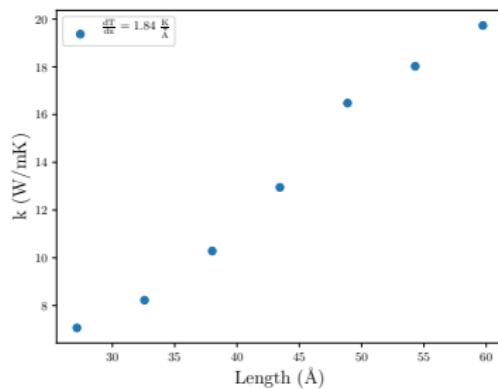
SELECTION OF WIDTH



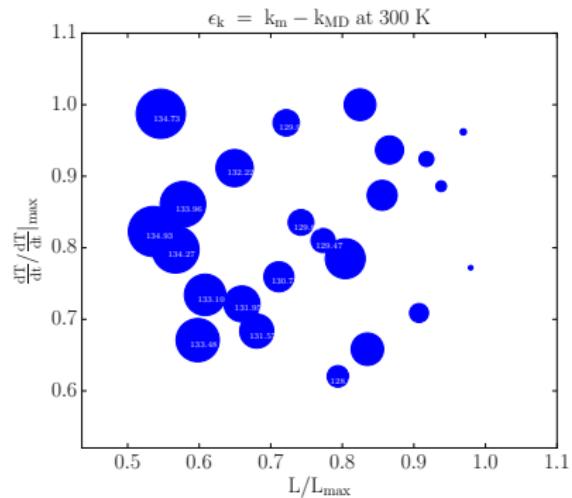
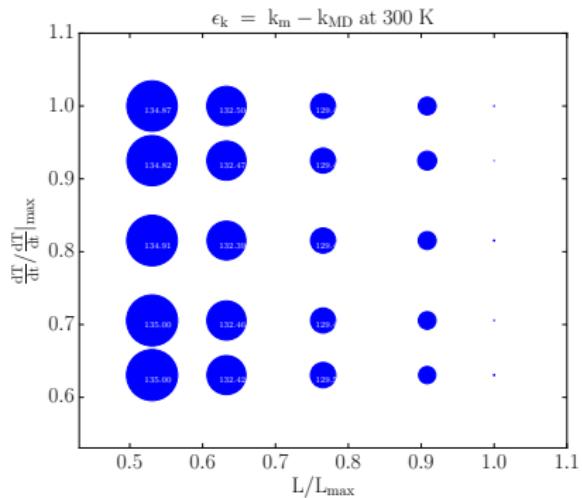
- At $W = 22L_c$, the effect of length on fluctuations seems negligible.

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

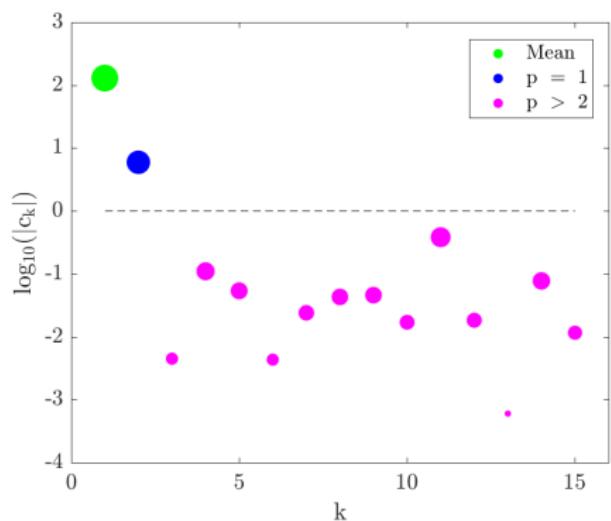


- Model realizations at Gauss-Legendre quadrature nodes are used to construct the PC surrogate.

PC EXPANSION

$$\epsilon (= |\kappa_m - \kappa_{\text{MD}}|) = \sum_j c_j \Psi_j(\xi_1, \xi_2)$$

κ_m : Measured, κ_{MD} : MD Prediction, j : Multi-index

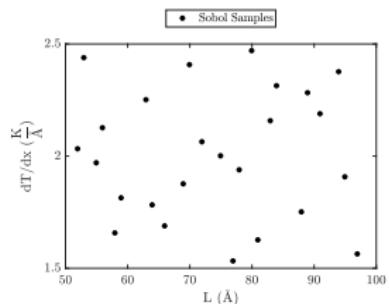


$$L: \mathcal{U}[50L_c, 100L_c] (\text{\AA}) \rightarrow \xi_1 : \mathcal{U}[-1, 1]$$

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

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

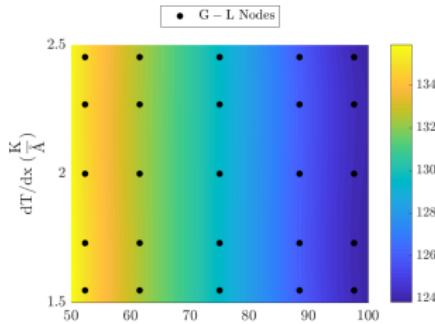
ACCURACY:



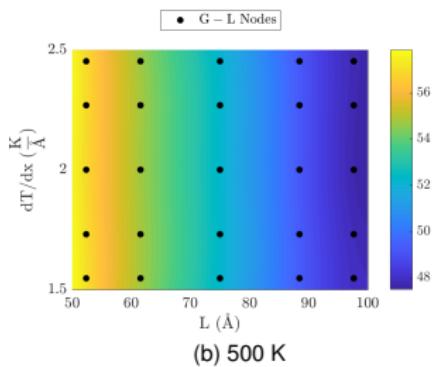
$$\epsilon = \frac{\left[\sum_j (\mathcal{G}_M - \mathcal{G}_{PCE})^2 \right]^{\frac{1}{2}}}{[(\mathcal{G}_M)^2]^{\frac{1}{2}}} \approx 1.8 \times 10^{-3}$$

\mathcal{G}_M : Model Output

\mathcal{G}_{PCE} : PCE Estimate

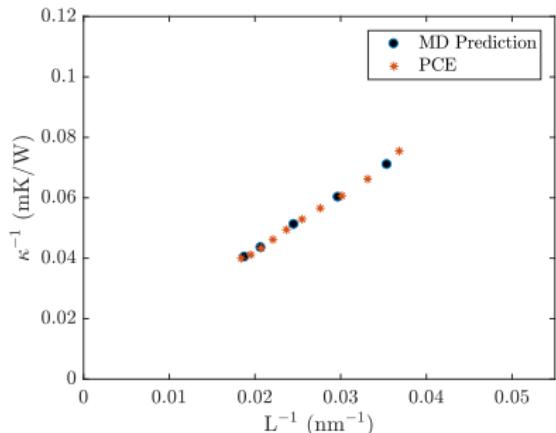


(a) 300 K

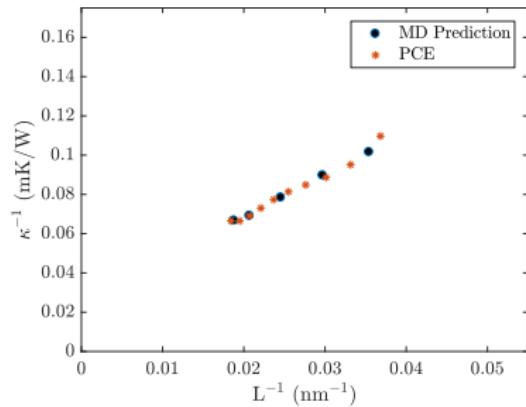


(b) 500 K

BULK THERMAL CONDUCTIVITY



$T = 300 \text{ K}$



$T = 1000 \text{ K}$

UNCERTAINTY IN INTER-ATOMIC POTENTIAL

OBJECTIVES

- Relative importance of potential field parameters using sensitivity analysis.
- Assess variability in thermal conductivity estimates due to perturbations in the potential field (Forward Problem).

$$\Phi(A, B, p, q, a, \lambda, \gamma) \mapsto k$$

- Robust calibration of the potential field parameters in a Bayesian setting.

CHALLENGES

- Global sensitivity analysis and the forward problem are computationally intractable.
- Explore the applicability of a derivative-based sensitivity measure to reduce the dimensionality of the problem.

DERIVATIVE-BASED GLOBAL SENSITIVITY MEASURES

MOTIVATION

- Sensitivity analysis based on Sobol indices is commonly used to determine relative importance of the parameters.
- Sobol sensitivity indices are compute intensive:

$$\mathcal{T}(\theta_i) = \frac{\mathbb{E}_{\boldsymbol{\theta} \sim i} [\mathbb{V}_{\theta_i}(\mathcal{G} | \boldsymbol{\theta}_{\sim i})]}{\mathbb{V}(\mathcal{G})}$$

- Bounds on Sobol indices can be computed easily using DGSM and are shown to converge at a much faster rate.

DERIVATIVE-BASED GLOBAL SENSITIVITY MEASURES

BACKGROUND

- DGSM for Randomly distributed parameters [Sobol and Kucherenko, 2009]:

$$\mu_i = \mathbb{E} \left[\left(\frac{\partial G(\boldsymbol{x})}{\partial x_i} \right)^2 \right]$$

where,

$$\frac{\partial G(\boldsymbol{x}^*)}{\partial x_i} = \lim_{\delta \rightarrow 0} \frac{[G(x_1^*, \dots, x_{i-1}^*, x_i^* + \delta, x_{i+1}^*, \dots, x_d^*) - G(\boldsymbol{x}^*)]}{\delta}$$

- Total number of model realizations required to compute μ_i using N samples is $N(d + 1)$.

DERIVATIVE-BASED GLOBAL SENSITIVITY MEASURES

BACKGROUND

- Upper bound on Sobol Total Effect index (ST_i) [Sobol and Kucherenko, 2009]:

$$ST_i \leq \frac{\mathcal{C}_i \mu_i}{V} (\propto \hat{\mathcal{C}_i \mu_i})$$

$$\hat{\mathcal{C}_i \mu_i} = \frac{\mathcal{C}_i \mu_i}{\sum_i \mathcal{C}_i \mu_i}$$

\mathcal{C} : Poincaré Constant V : Variance

- The Poincaré Constant is specific to a given probability distribution:

$\mathcal{U}[a, b]$	$(b - a)^2 / \pi^2$
$\mathcal{N}(\mu, \sigma^2)$	σ^2

ALGORITHM: PARAMETER SCREENING

```
1 Generate  $n_1$  points in  $\mathbb{R}^d$ ;  
2 Compute  $UB_i$  for parameters  $\theta_i$  using  $n_1$  points;  
    % NF =  $n_1(d+1)$ , NF: Number of model realizations;  
3 Rank Parameters ( $\theta_i$ ) based on  $UB_i$  estimates ( $\mathcal{R}^{old}$ );  
4 set  $k = 1$  % k: Iteration counter;  
5 repeat  
6     Generate  $\beta n_1$  new points in  $\mathbb{R}^d$  ( $\beta n_1 \in \mathbb{Z}$ );  
7     Compute  $UB_i^{new}$  using  $(1+\beta k)n_1$  points;  
    % NF =  $(1 + \beta k)n_1(d+1)$ ;  
8     Rank Parameters based on  $UB_i^{new}$  estimates ( $\mathcal{R}^{new}$ );  
9     if ( $\mathcal{R}^{new} = \mathcal{R}^{old}$ ) then  
10        Compute:  $r_i = \frac{UB_i^{new}}{\max(UB_i^{new})}$ ;  
11        Construct a set  $s = \{\theta_i \ni r_i < \alpha\}$ ;  
12        Exit the loop;  
13    end  
14    set  $k = k + 1$ ;  
15 until  $\mathcal{R}^{new} \neq \mathcal{R}^{old}$ ;  
16 Construct a validation set:  $(\theta_j, \mathcal{M}(\theta_j))$ ,  $j=1, 2, \dots, NF$ ;
```

BACKGROUND
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SET-UP
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ERROR RESPONSE
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SENSITIVITY
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PCE
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LIKELIHOOD
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SUBSPACE
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MD SIMULATION

NPT

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NVT

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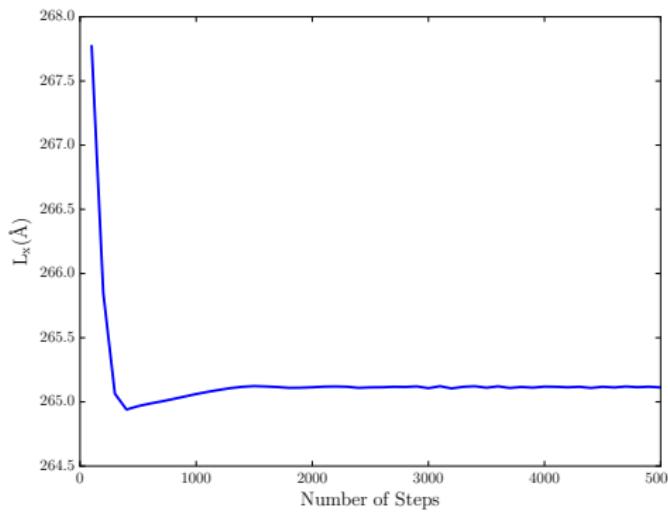
NVE

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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



BACKGROUND
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SENSITIVITY
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PCE
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LIKELIHOOD
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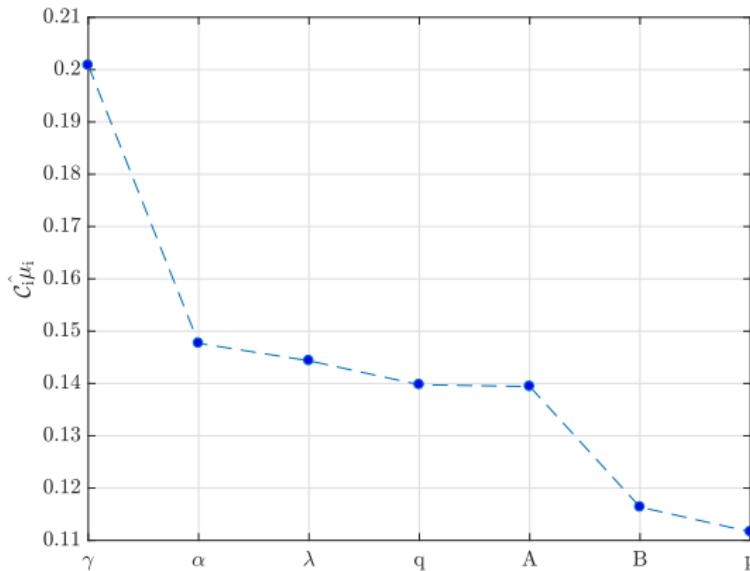
SUBSPACE
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STRUCTURAL INSTABILITY

PARAMETER SCREENING

 $\hat{C_i\mu_i}$: 10 Samples $\hat{C_i\mu_i}$: 15 Samples $\hat{C_i\mu_i}$: 20 Samples $\hat{C_i\mu_i}$: 25 Samples $\hat{C_i\mu_i}$: 30 Samples γ : 0.2531 γ : 0.2001 γ : 0.1933 γ : 0.2075 γ : 0.2024 α : 0.2170 α : 0.1723 α : 0.1593 α : 0.1495 α : 0.1487 A : 0.2014 A : 0.1508 q : 0.1468 q : 0.1493 λ : 0.1424 B : 0.0964 B : 0.1381 λ : 0.1325 λ : 0.1392 q : 0.1404 p : 0.0916 p : 0.1253 A : 0.1261 A : 0.1217 A : 0.1395 q : 0.0741 q : 0.1232 B : 0.1242 B : 0.1175 B : 0.1142 λ : 0.0664 λ : 0.0900 p : 0.1177 p : 0.1153 p : 0.1125

PARAMETER SCREENING



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PCE

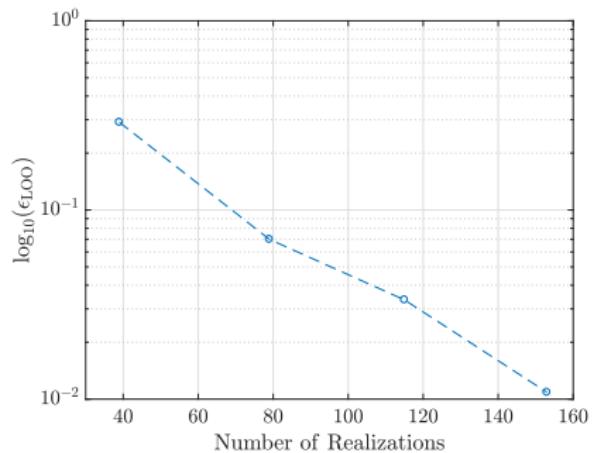
$$\kappa = \sum_{s \in \mathcal{A}} c_s \Psi_s(\xi)$$

\mathcal{A} : Sparse basis

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

$$\Psi_s(\xi) = \psi_{s_1}(\xi_1) \psi_{s_2}(\xi_2) \psi_{s_3}(\xi_3) \psi_{s_4}(\xi_4) \psi_{s_5}(\xi_5)$$

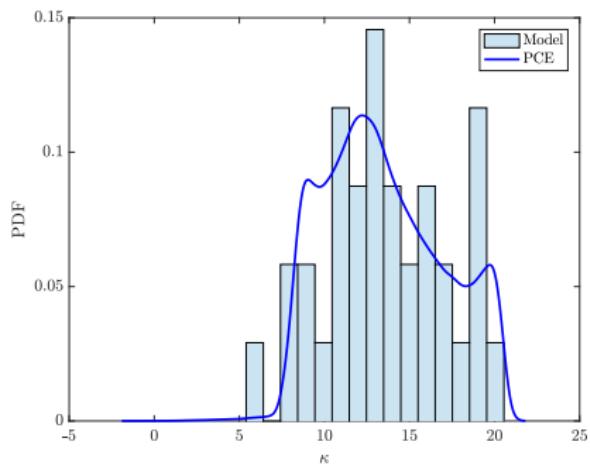
$$\xi_i \sim \mathcal{U}[-1, 1]$$



VERIFICATION

L-2

$$\epsilon_{val} = \frac{\left[\sum_{i=1}^N \left(\mathcal{M}(\mathbf{x}^{(i)}) - \mathcal{M}^{PCE}(\mathbf{x}^{(i)}) \right)^2 \right]^{\frac{1}{2}}}{\left[\sum_{i=1}^N \left(\mathcal{M}(\mathbf{x}^{(i)}) \right)^2 \right]^{\frac{1}{2}}} \\ = 6.88 \times 10^{-2}$$



SENSITIVITY ANALYSIS

$\hat{C}_i \mu_i$: 30 Samples

γ : 0.2024

α : 0.1487

λ : 0.1424

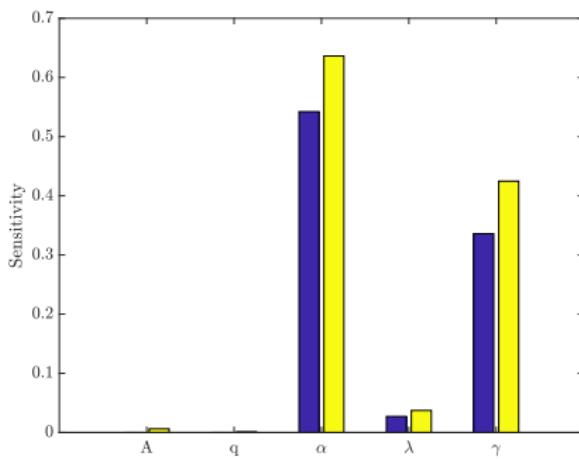
q : 0.1404

A : 0.1395

B : 0.1142

p : 0.1125

SOBOL SENSITIVITY INDICES



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LIKELIHOOD
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SUBSPACE
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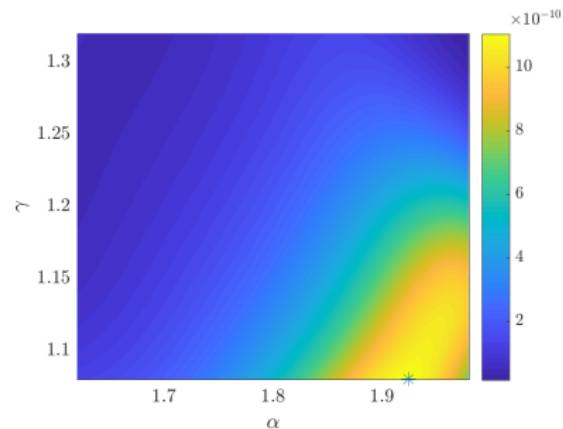
LIKELIHOOD

Bayes' rule:

$$\mathcal{P}(\alpha, \gamma | \kappa) \propto \mathcal{P}(\kappa | \alpha, \gamma) \mathcal{P}(\alpha) \mathcal{P}(\gamma)$$

$$\mathcal{P}(\alpha) \sim \mathcal{U}[1.62, 1.98]$$

$$\mathcal{P}(\gamma) \sim \mathcal{U}[1.08, 1.32]$$



ACTIVE SUBSPACE

Positive Semi-definite Matrix:

$$\mathcal{C} = \int \nabla f(\mathbf{x}) \nabla f(\mathbf{x})^T \rho(\mathbf{x}) d\mathbf{x} = \mathbf{W} \Lambda \mathbf{W}^T$$

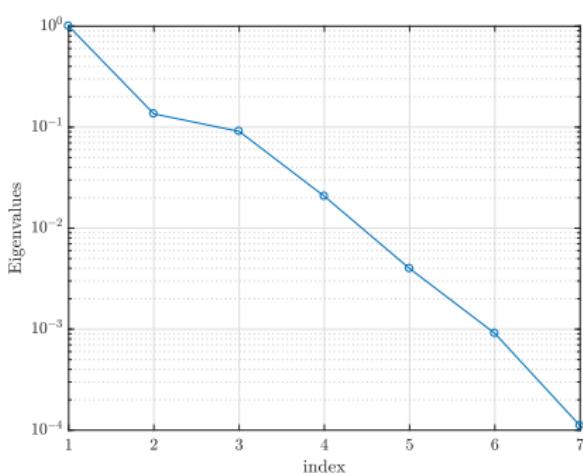
Partition of Eigenpairs:

$$\Lambda = \begin{bmatrix} \Lambda_1 & \\ & \Lambda_2 \end{bmatrix}, \quad \mathbf{W} = [\mathbf{W}_1 \ \mathbf{W}_2]$$

MC Integration:

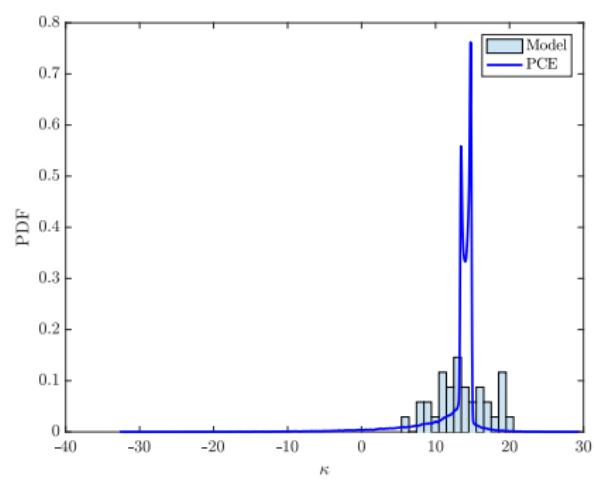
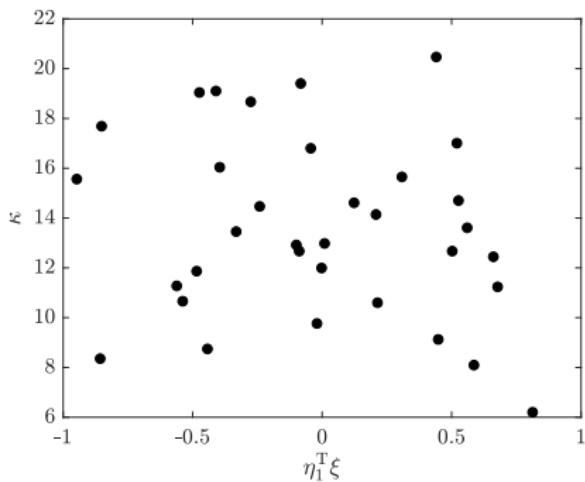
$$\mathcal{C} \approx \hat{\mathcal{C}} = \frac{1}{N} \sum_{i=1}^N \nabla_{\mathbf{x}} f_i \nabla_{\mathbf{x}} f_i^T = \hat{\mathbf{W}} \hat{\Lambda} \hat{\mathbf{W}}^T$$

$$f(\mathbf{x}) \approx g(\mathbf{W}_1^T \mathbf{x})$$



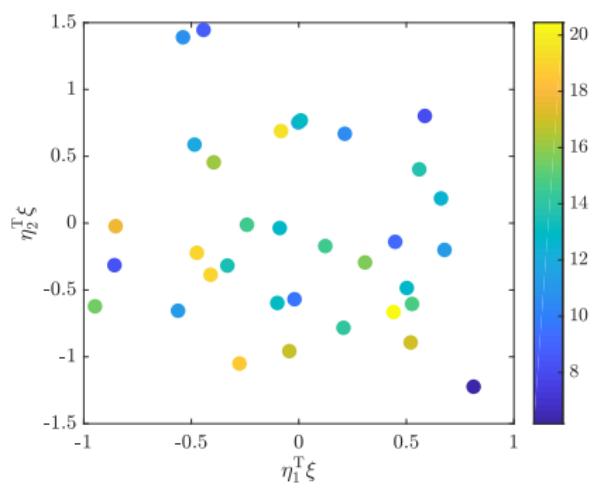
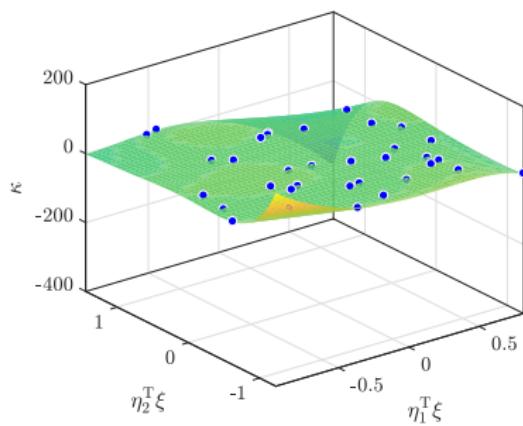
1D ACTIVE SUBSPACE

$$f(x) = g(\eta_1^T \xi) \quad (x \rightarrow \xi \in [-1, 1])$$



2D ACTIVE SUBSPACE

$$f(\mathbf{x}) = g(\eta_1^T \xi, \eta_2^T \xi)$$



3D ACTIVE SUBSPACE

$$f(\boldsymbol{x}) = g(\eta_1^T \boldsymbol{\xi}, \eta_2^T \boldsymbol{\xi}, \eta_3^T \boldsymbol{\xi})$$

