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Continuum Nanoscale Constitutive Laws with Quantified Uncertainty Extracted from Atomistic Simulations Using Bayesian Inference

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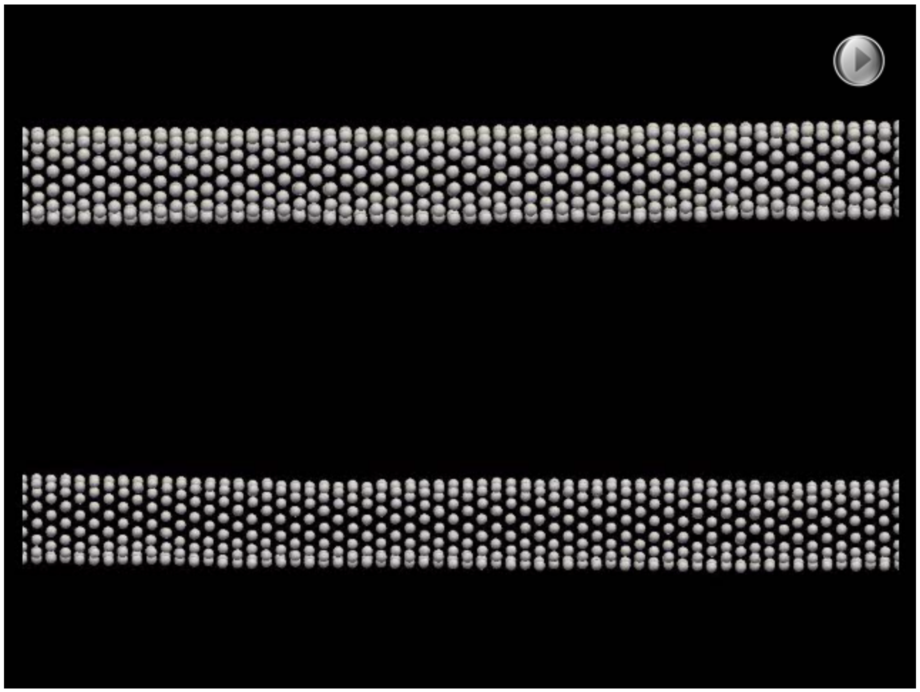
Background and Motivation

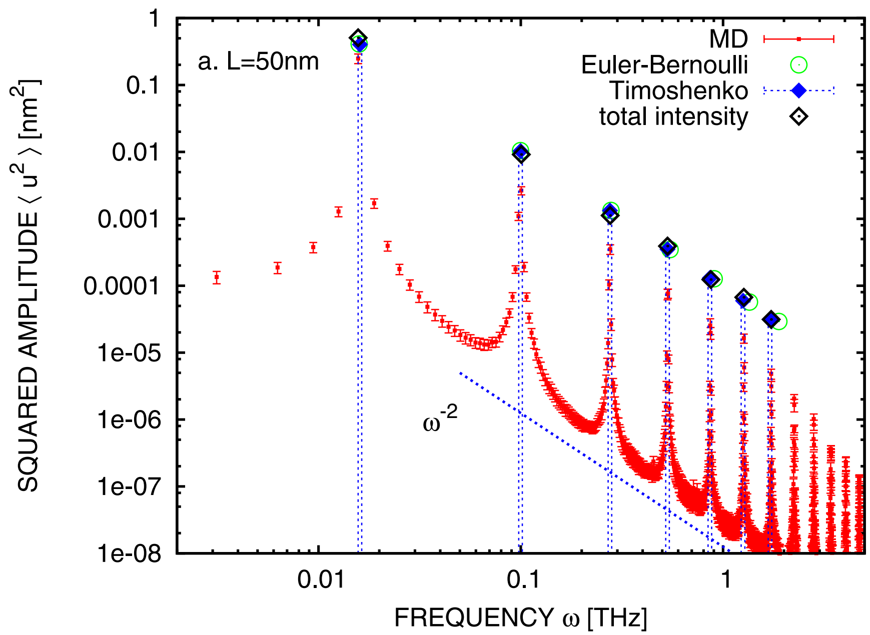
Continuum models can hold at the nanoscale, but….

$$\rho A \frac{\partial^2 y}{\partial t^2} = -\frac{\partial}{\partial x}\left[kAG\left(\phi - \frac{\partial y}{\partial x}\right)\right]$$

$$\rho I \frac{\partial^2 \phi}{\partial t^2} = EI\frac{\partial^2 \phi}{\partial x^2} - kAG\left(\phi - \frac{\partial y}{\partial x}\right)$$



Lee, Meade, Barerra & Templeton, *J. Nanoeng. Nanosys.* 2011



Feng & Jones, *PRB* 2010

See the excellent paper by Govindjee & Sackman (1999) for more

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Background and Motivation

- Physical systems often involve phenomena occurring at scales that are beyond the reach of continuum (*e.g.* applications in nanotechnology and fluid dynamics).
- At such small scales, the continuum constitutive laws (*e.g.* relationship between heat flux and temperature gradient, or between fluid flow velocity and pressure gradient) are proved to be inadequate.
- The unresolved degrees of freedom in the mesoscale and atomistic scales are often approximated by some corrections in the constitutive laws.

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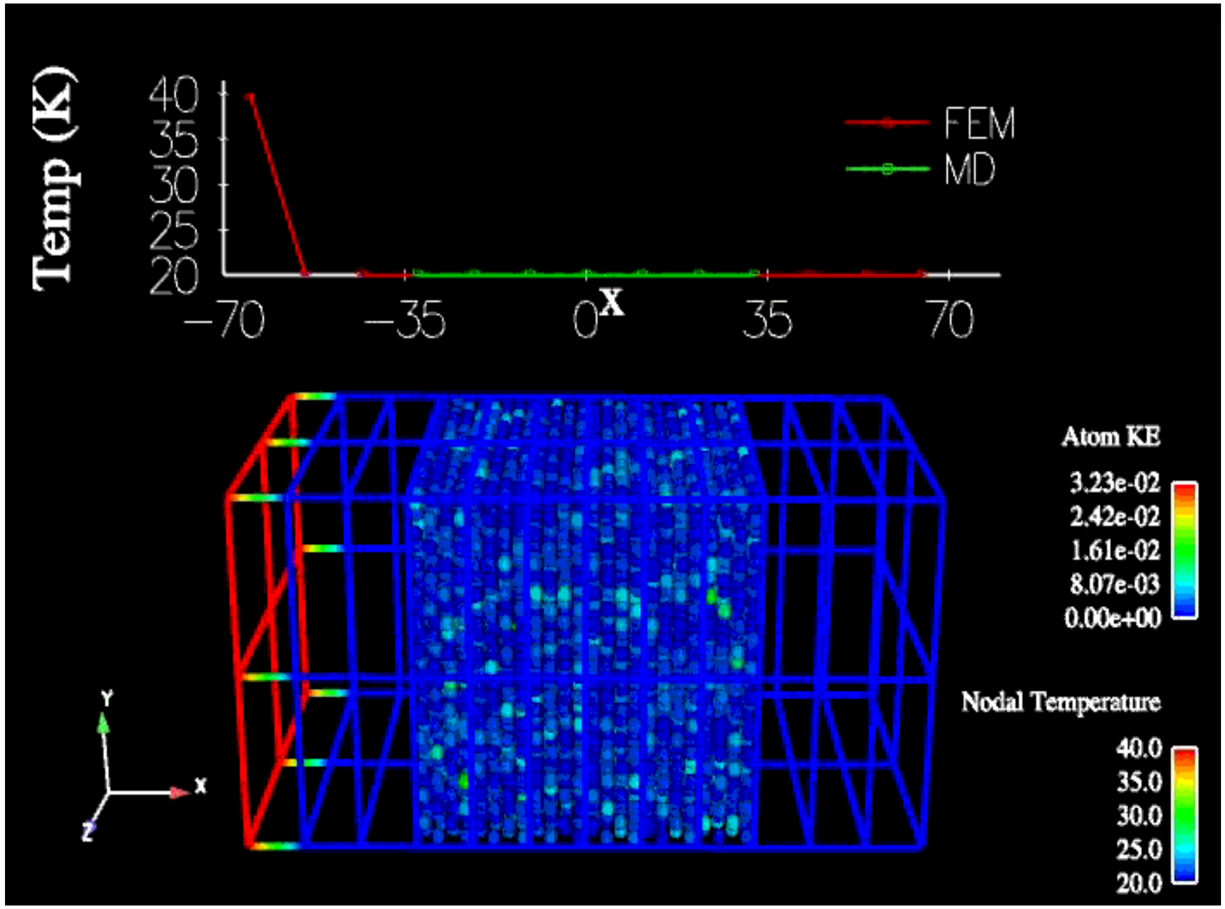
Fourier's law and coarse-grained atomistics follow different dynamics

Possible causes:

- Incorrect conductivity?
- Stochasticity?
- Does Fourier's law even hold at the atomistic scale?



The lack of understanding of these causes is problematic because their effect propagates to the continuum scale.

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Two Questions

1. *Can established continuum constitutive laws be modified to enable predictive simulation at the nanoscale?*

2. *If so, how? If not, is there a framework for deriving such laws?*

Approach

- *Hypothesis: Extraction of continuum scale constitutive laws from atomistic simulations can be accomplished based on uncertainty quantification and statistics.*
  - *How do we use them in continuum simulations?*
  - *What happens when we are not in the large-time limit?*

- We first analyze and study the extraction of a continuum scale constitutive law from molecular dynamics (MD) simulations while accounting for the uncertainty due to finite sample size and time scale arising from physical fluctuations.

- We use Bayesian inference to extract the constitutive relationships from noisy atomistic data, but we need to determine the appropriate mathematical framework for the models.

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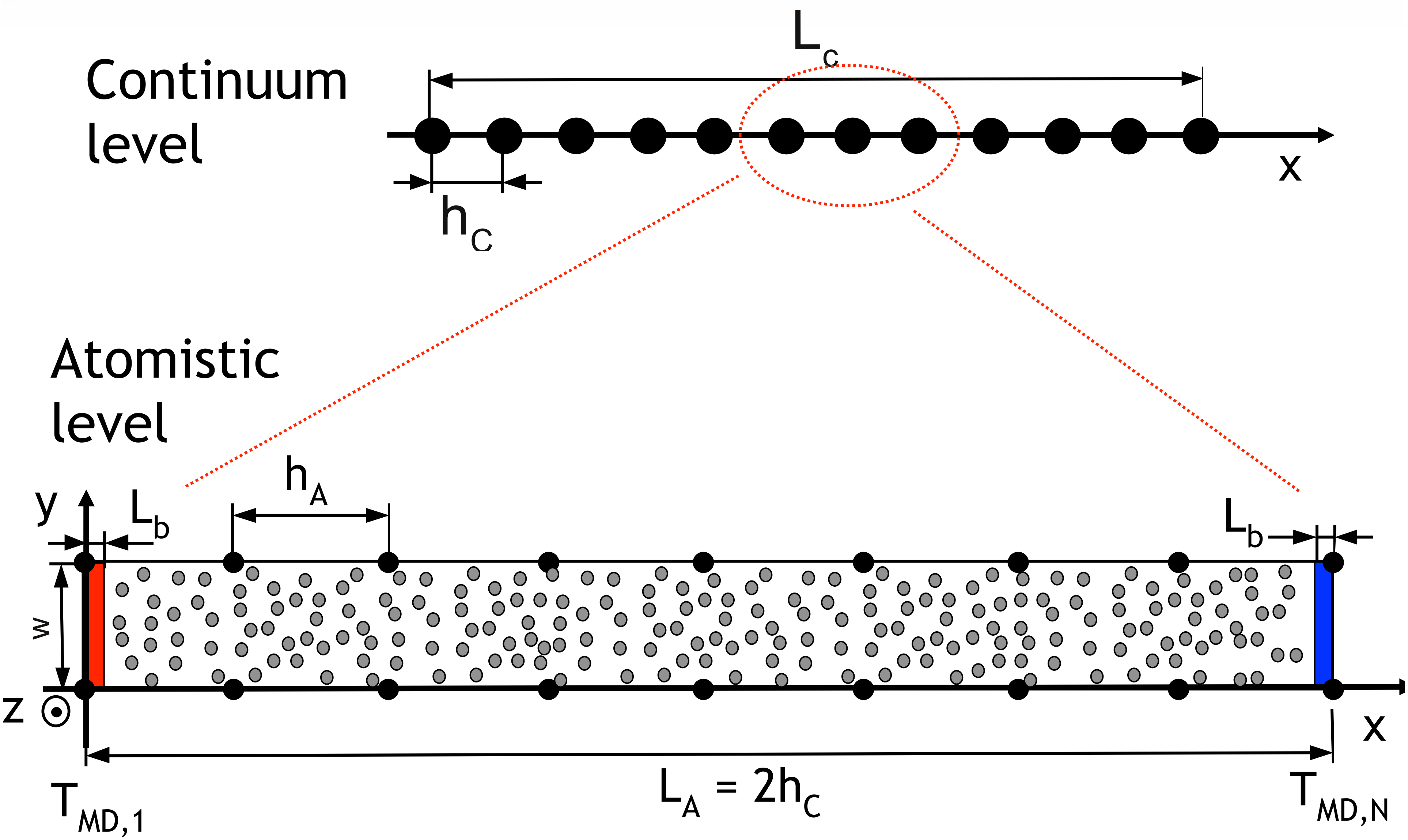
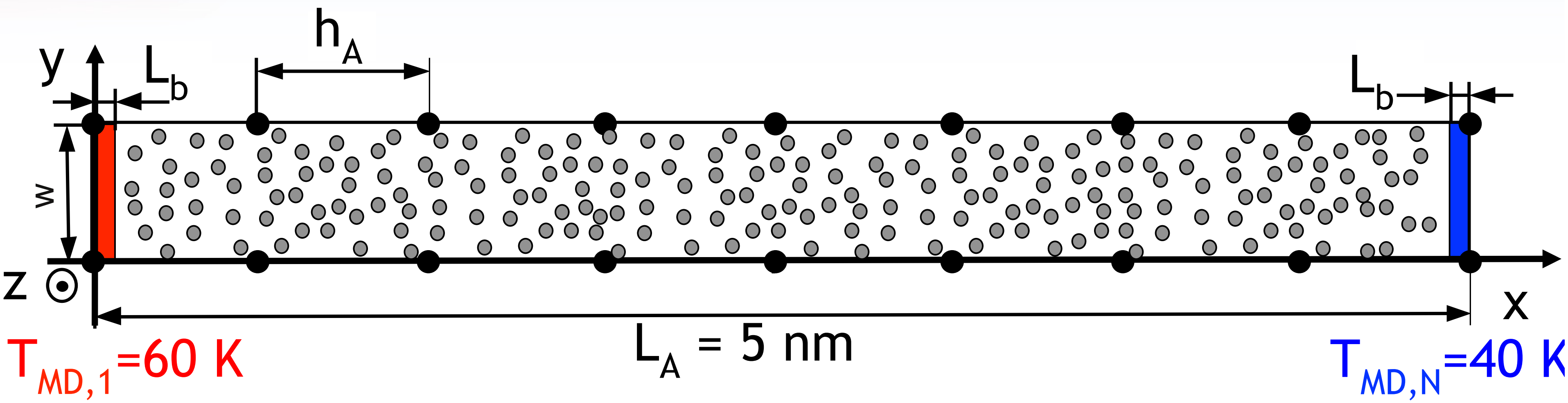
One dimensional heat transfer is used as model problem for study



Atomistic Simulation of the heat transfer in a "1D" bar



- The bar is made of solid Argon.

- Initially the temperature is equal to 20 K all over the bar.
At t=0, we impose the ***Dirichlet boundary conditions*** shown in the figure.

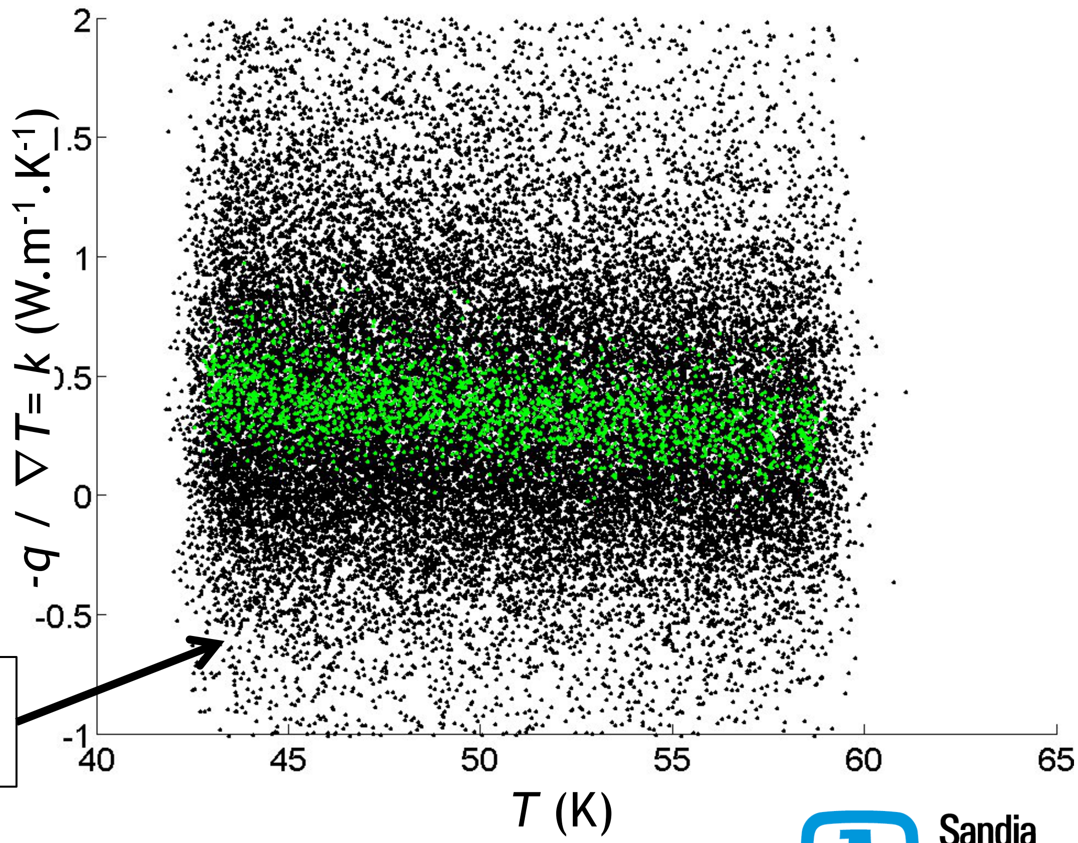
- We extract the temperature *T*, temperature gradient ∇T and flux *q* at the mesh depicted in the above figure using the Hardy formalism.

Templeton, Jones, & Wagner, *MSMSE* 2010.
Zimmerman, Jones, & Templeton. *J. Comp. Phys.* 2010.

Time Averaging and Sampling

We average the data obtained from the atomistic simulations with a time window t_w. The plot below shows short-time averaged values of the thermal conductivity *k* versus temperature.

Black: t_w=64 fs
Green: t_w=1024 fs



At low time scales, the thermal conductivity can go negative!

Bayesian Inference

Let *m* be a hypothesis and *D* observed data.

Posterior — Likelihood — Prior

$$\mathcal{P}(\boldsymbol{m}|\boldsymbol{d}) = \frac{\mathcal{P}(\boldsymbol{d}|\boldsymbol{m})\mathcal{P}(\boldsymbol{m})}{\int_{\Omega} \mathcal{P}(\boldsymbol{d}|\boldsymbol{m})\mathcal{P}(\boldsymbol{m})d\boldsymbol{m}}$$

- The prior expresses the initial knowledge about the hypothesis ***m*** (*e.g.* uniform distribution, expert's knowledge...)

- The likelihood is the probability of observing the data ***d*** given the hypothesis ***m***. It encompasses the forward model of ***m***.

- The denominator is a normalization constant.

- The posterior is the probability of the hypothesis ***m*** given the data ***d*** : offers an enhanced knowledge of ***m***.

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METHOD 1: INFORM A FOURIER CONDUCTIVITY

Bayesian Inference of the Thermal Conductivity

We ASSUME that the thermal conductivity is a linear function of the temperature. We have:

$$k = A - BT, \quad q = -\left(A - BT\right)\nabla T + \sigma\varepsilon$$

We assume a constant parametric noise model

Where $\epsilon \sim N(0,1)$ and, A and B are the parameters represented by PCEs to be inferred along with the hyperparameter σ^2.

The data $\boldsymbol{d}_j^{N_d} = (T_j, \nabla T_j, q_j)$ is obtained from the MD simulations.

We assume a Gaussian likelihood function:

$$L(\boldsymbol{d}|A, B, \sigma^2) = -(2\pi)^{-N_d/2}(\sigma^2)^{-N_d/2}\exp\left(\frac{-\epsilon^T\epsilon}{2\sigma^2}\right)$$

Assuming Jeffrey's prior on σ^2 and improper uniform priors on A and B, the marginal joint posterior (A,B) follows a Student-t distribution and can be derived analytically. The random variable $k=A-BT$ also follows a Student-t distribution.

Rizzi, Salloum, Marzouk, Xu, Falk, Weihs, Fritz, & Knio, *SIAM MMS* 2011.
Marzouk, Najm, & Rahna, *J. Comp. Phys.* 2007.

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Enforcing the positivity of *k*

- The derivation so far does not guarantee that the random variable *k* is positive for all its realizations.

- To enforce the positivity of k, we proceed as follows:

1. Samples are drawn from the obtained Student-t distribution.

2. The samples that do not satisfy the positivity constraints are eliminated.

3. The Rosenblatt transform is used to map the truncated joint posterior (A,B) into two independent uniform random variables.

4. An approximate inverse Rosenblatt transform is used to map the uniform variables into PCEs.

Sargsyan, Debusschere, Najm, & Le Maitre, *SIAM. J. Sci. Comput*. 2010.
Sargsyan, Safta, Debusschere, & Najm, *SIAM. J. Sci. Comput*. 2012.

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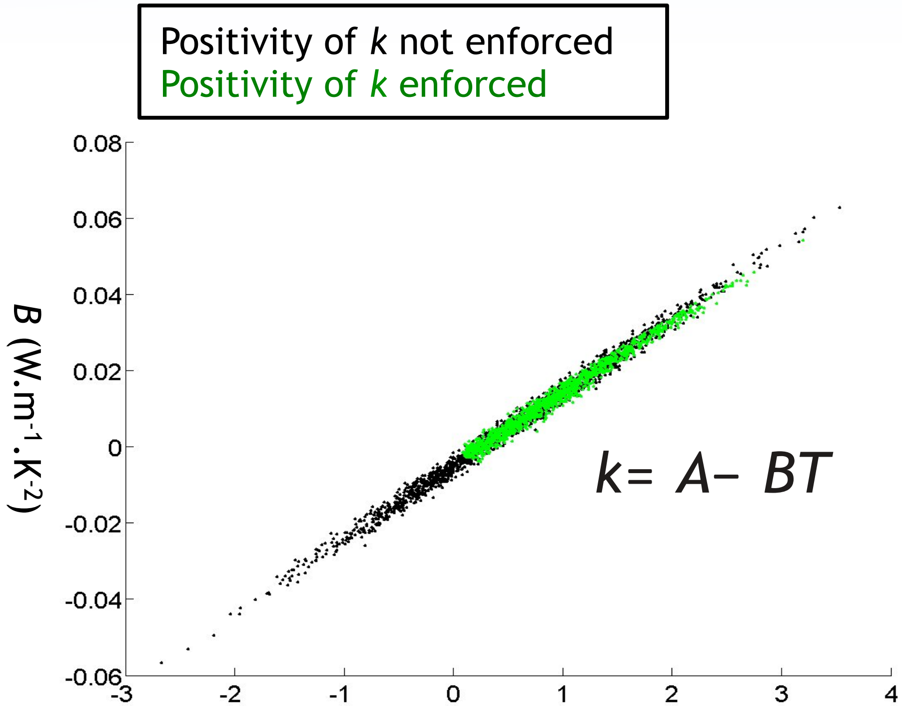
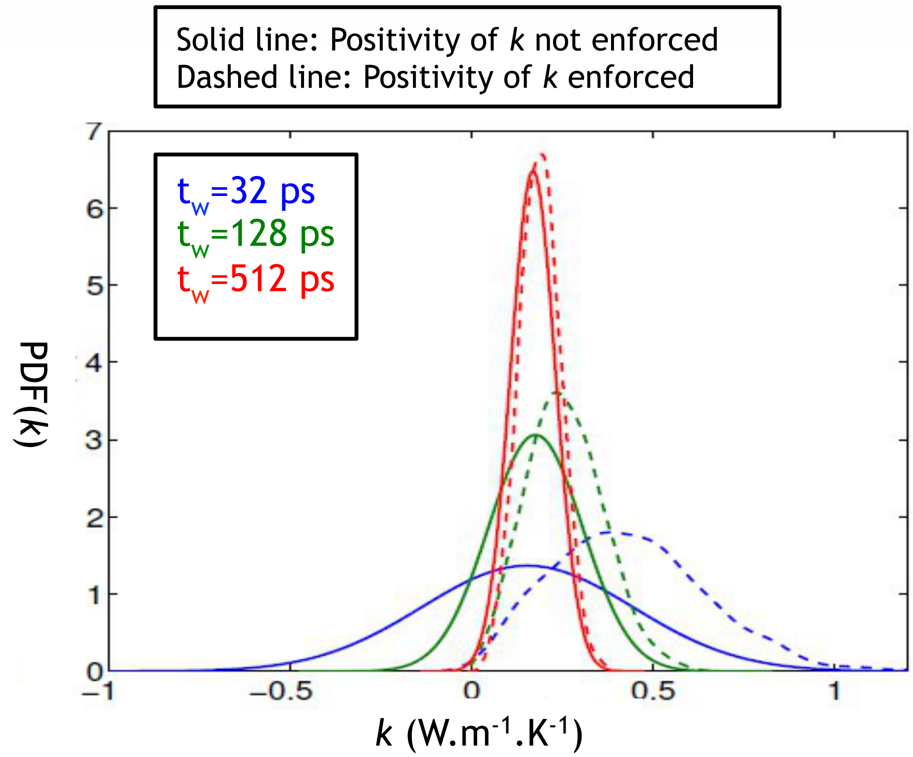
Enforcing the positivity of k





After enforcing the positivity, the mean of k increases which physically implies that more heat is transported in the material when the effect of the backward propagating phonons is eliminated.

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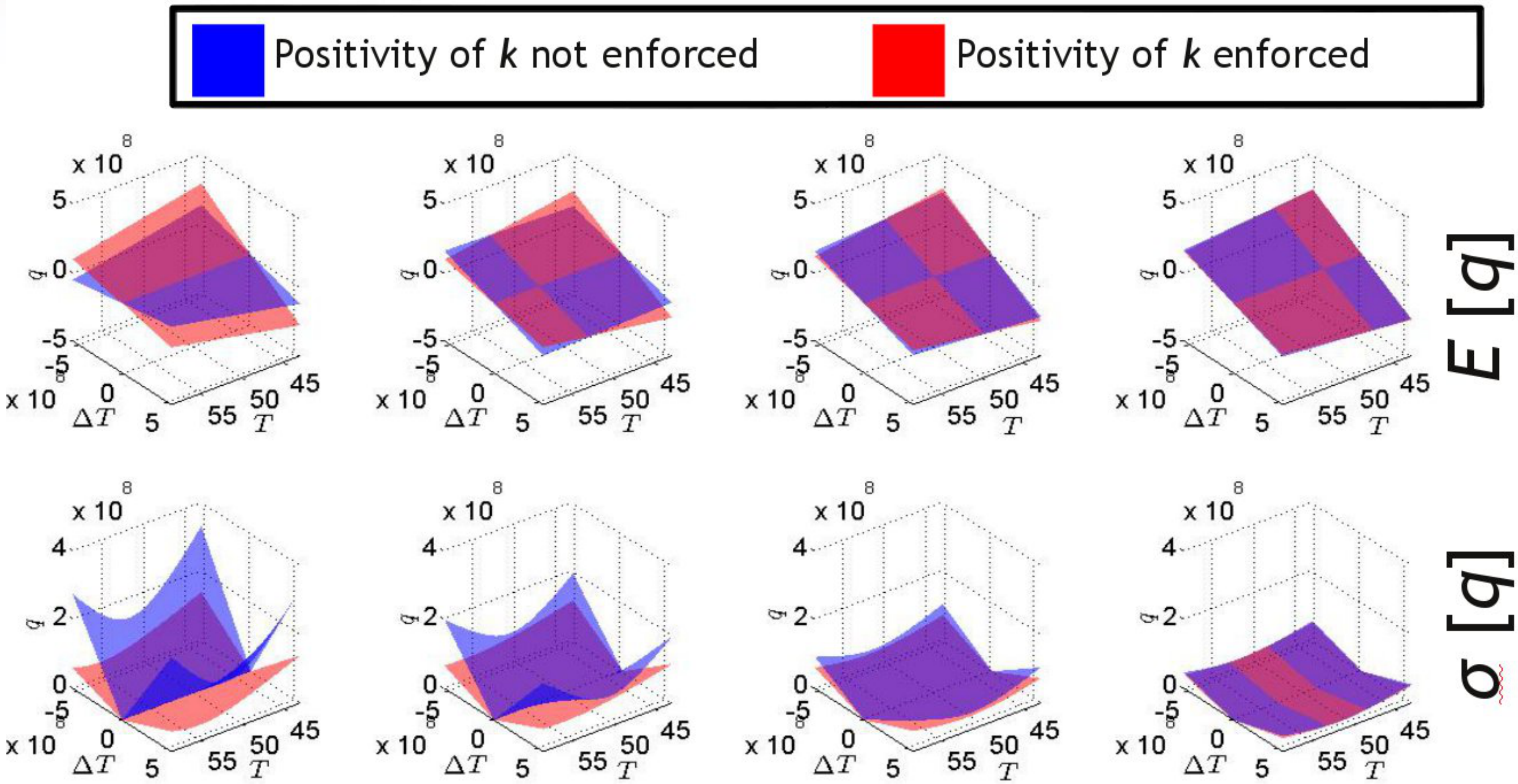
The Constitutive Law



t_w=8 ps t_w=32 ps t_w=128 ps t_w=512 ps

Enforcing the positivity of *k* effectively removes the negative conductivity samples and results in an over-prediction of the heat flux.

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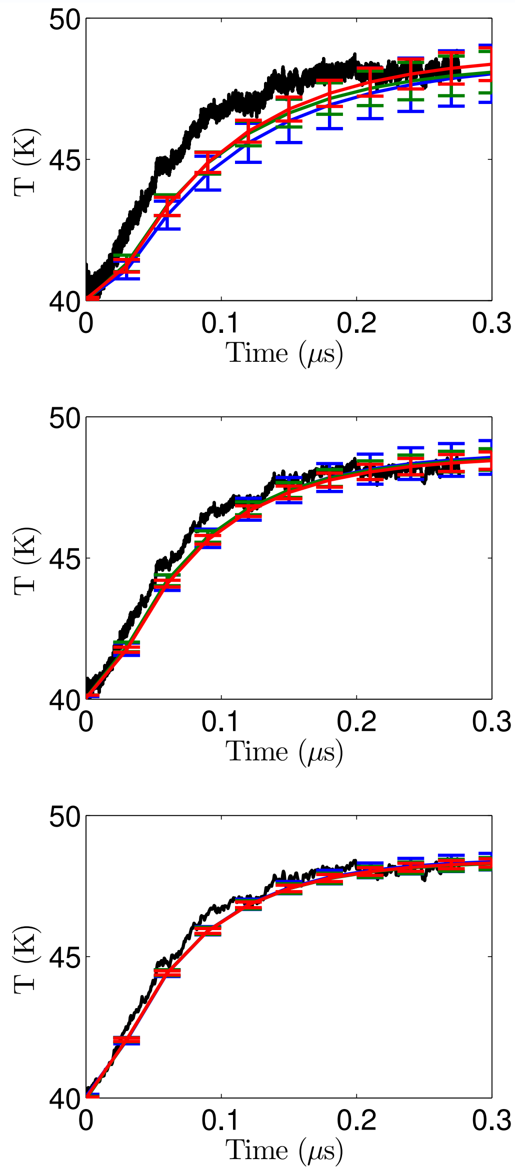
Continuum Simulation

Sample Time Scale

32 ps

128 ps

512 ps



Full MD simulation
Continuum simulation based on 256 samples
Continuum simulation based on 512 samples
Continuum simulation based on 1024 samples

We propagate the obtained constitutive law with its quantified uncertainty in a continuum scale 1D heat transfer simulation.

We compare the obtained mean temperature (smooth profiles) to the one obtained by solving the continuum problem directly using MD (noisy profiles). Very expensive simulation!

A reasonable agreement with this "true solution" is found for t_w>128 ps but the estimator is empirically biased (undesirable).

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METHOD 2: MODEL CONDUCTIVITY AS A GAUSSIAN PROCESS

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Bayesian Inference of a Gaussian Process Heat Flux Model

We ASSUME that the heat flux is a function of the temperature and temperature gradient with the following Gaussian likelihood function:

$$L\left(q \,/\, \beta,\sigma^2,\Psi^2\right) \approx N\left(H\beta,\sigma^2 C\right)$$

Where H and C are derived from data at each training point, $\boldsymbol{d}_j^{N_d} = (T_j, \nabla T_j, q_j)$ and β and σ^2 are inferred from the MD simulations.

We then derive a heat flux model, including model form uncertainty:

$$q = f\left(T,\nabla T\right) + \sqrt{\sigma^2 V}\,\xi$$

Assuming Jeffrey's prior on σ^2 and uniform priors on the coefficients and set roughness parameters by maximizing an appropriate likelihood function, the marginal joint posterior follows a Student-t distribution and can be derived analytically.

Oakley & O'Hagan, *SIAM*. *Biometrika* 2002.
Bastos & O'Hagan, *Technometrics* 2009.

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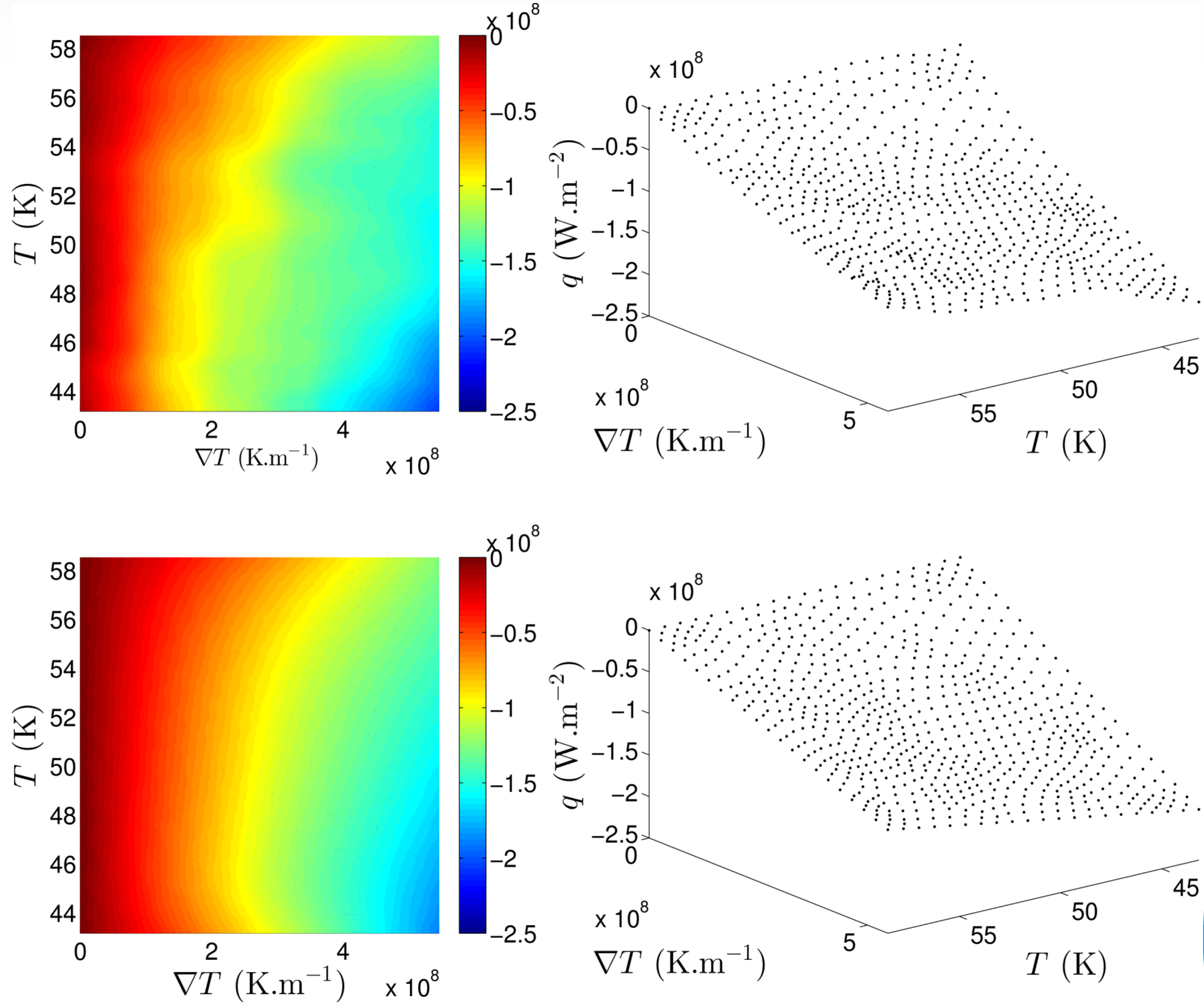
Enforcing Solvability Constraints $\frac{\partial q}{\partial \nabla T} \leq 0 \; \forall \, T, \nabla T$

WARNING: Hack



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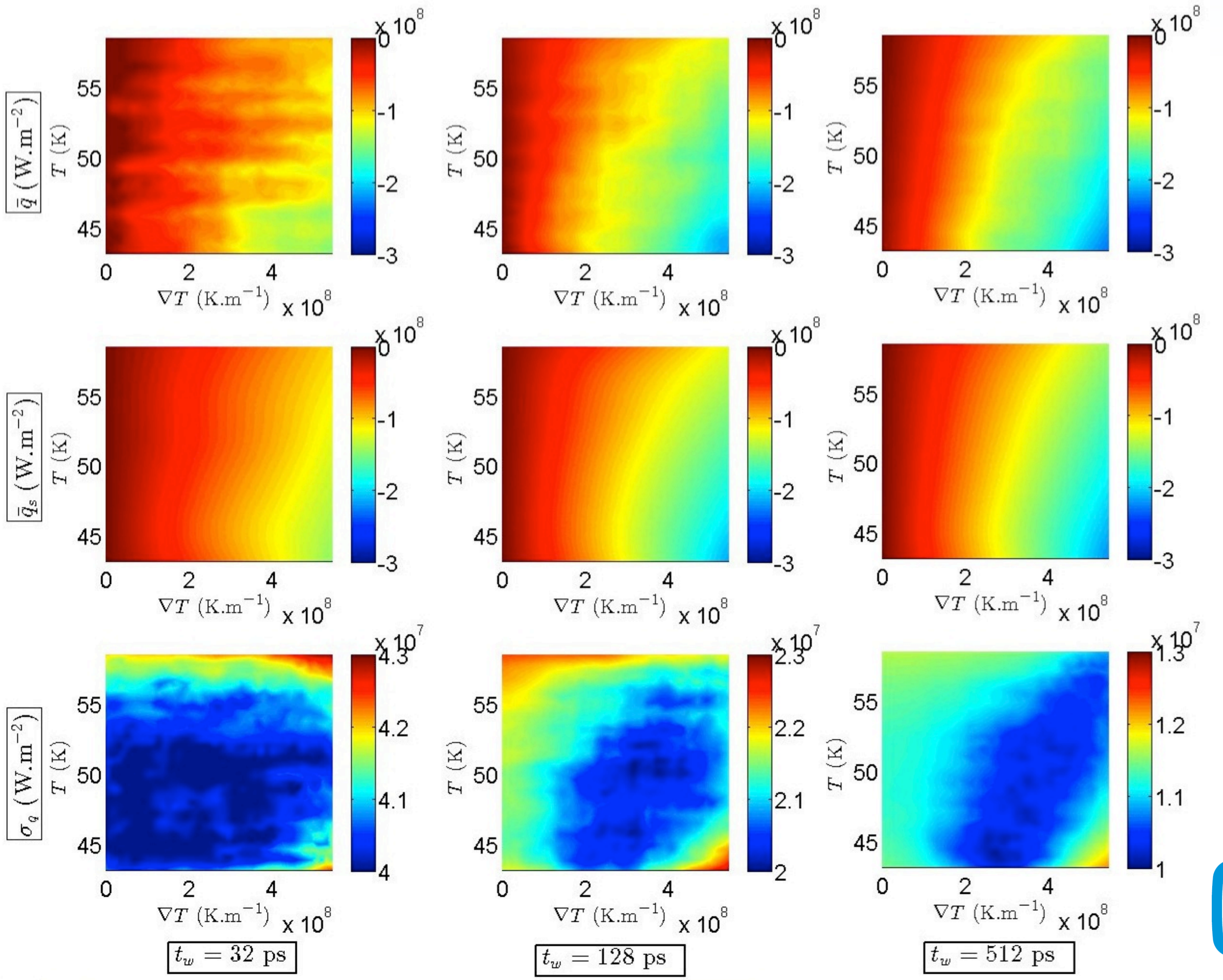
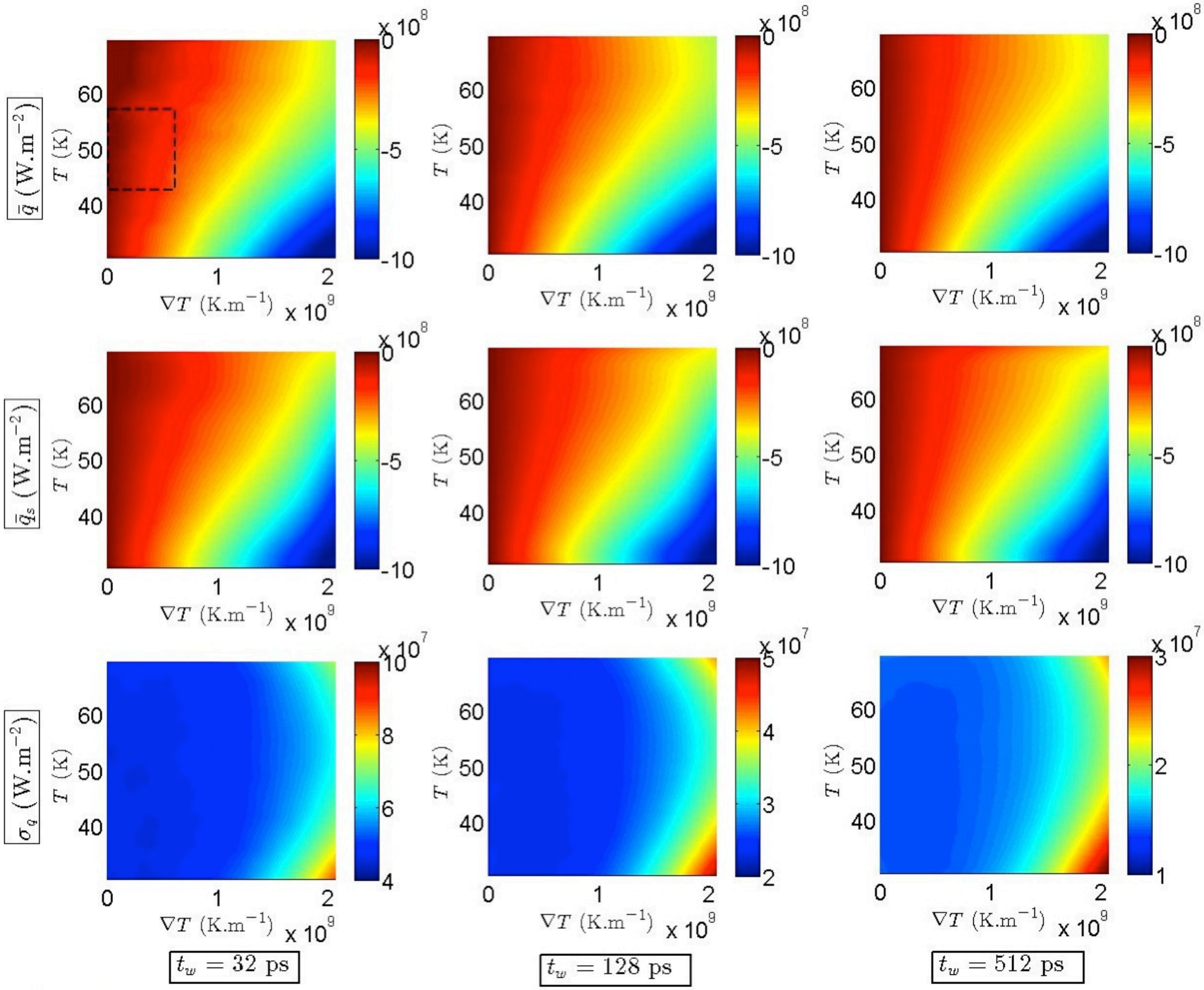
Gaussian Process Results Using Local Sampling



Gaussian Process Results Using Global Sampling



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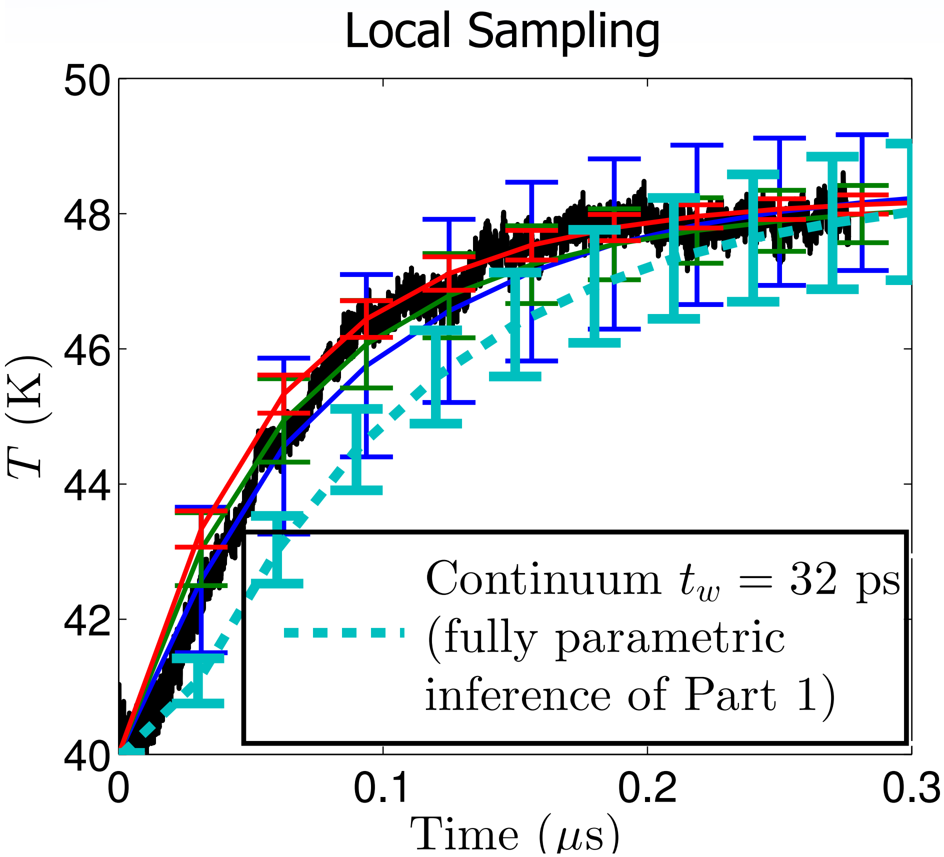
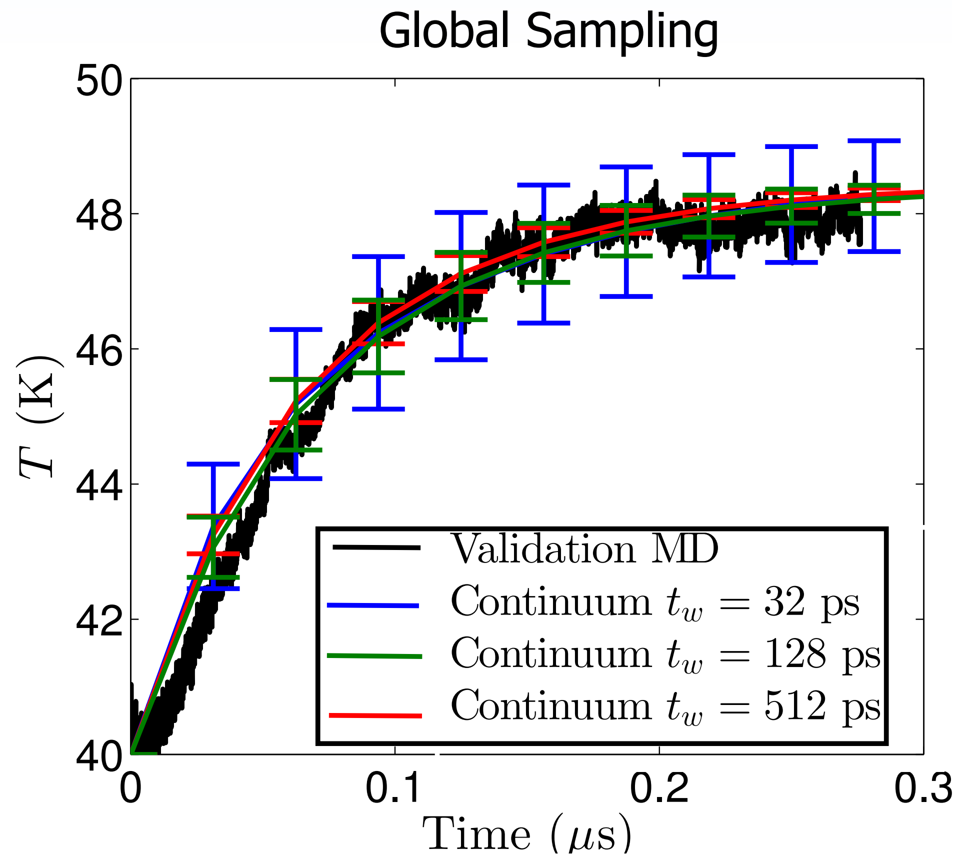
Gaussian Process Results





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Conclusions

- We demonstrated the feasibility of determining unknown continuum constitutive relationships from simulation data in a nanoscale setting using Bayesian inference.
- It was necessary to enforce the appropriate constraints on heat flux forms because diffusion equations have solvability constraints.
- Uncertain constitutive laws were propagated in a continuum scale simulation. The obtained temperature mean is in good agreement with the one obtained from the "true solution" for bigger time scales.
- The method we developed incurs significant computational savings and has promise to to be used in a wide variety of problems in which constitutive relations are needed to efficiently simulate a coarse-grained model.
- Further efforts will be made to account for model errors, dynamically optimize the parameters used in this study and to test its performance in additional problems of physical interest.

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THANK YOU FOR YOUR ATTENTION

Questions???

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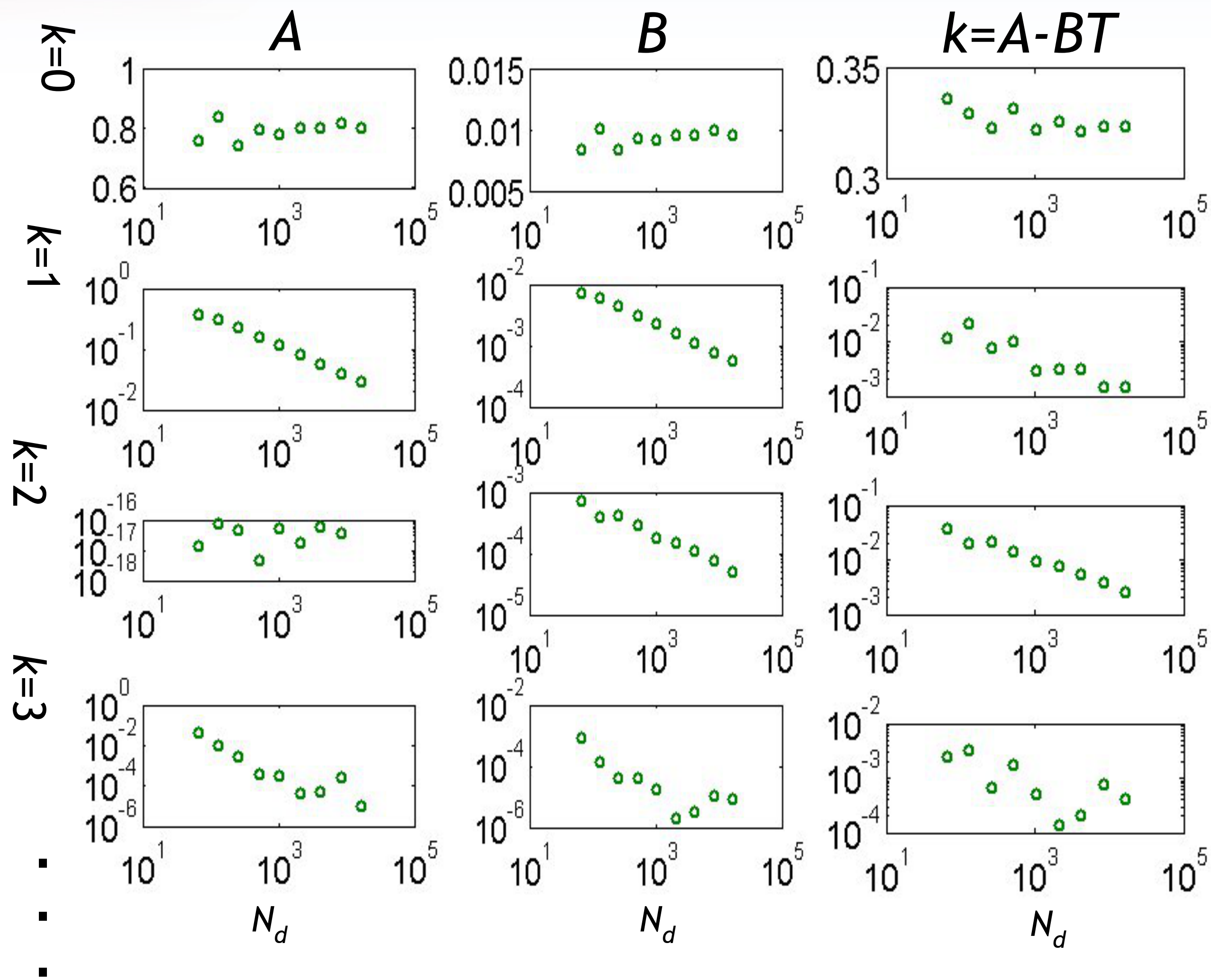
The PC coefficients



T=50 K
t_w=128 ps

$$A = \sum_{k=0}^{P} A^k \Psi^k(\xi)$$

$$B = \sum_{k=0}^{P} B^k \Psi^k(\xi)$$

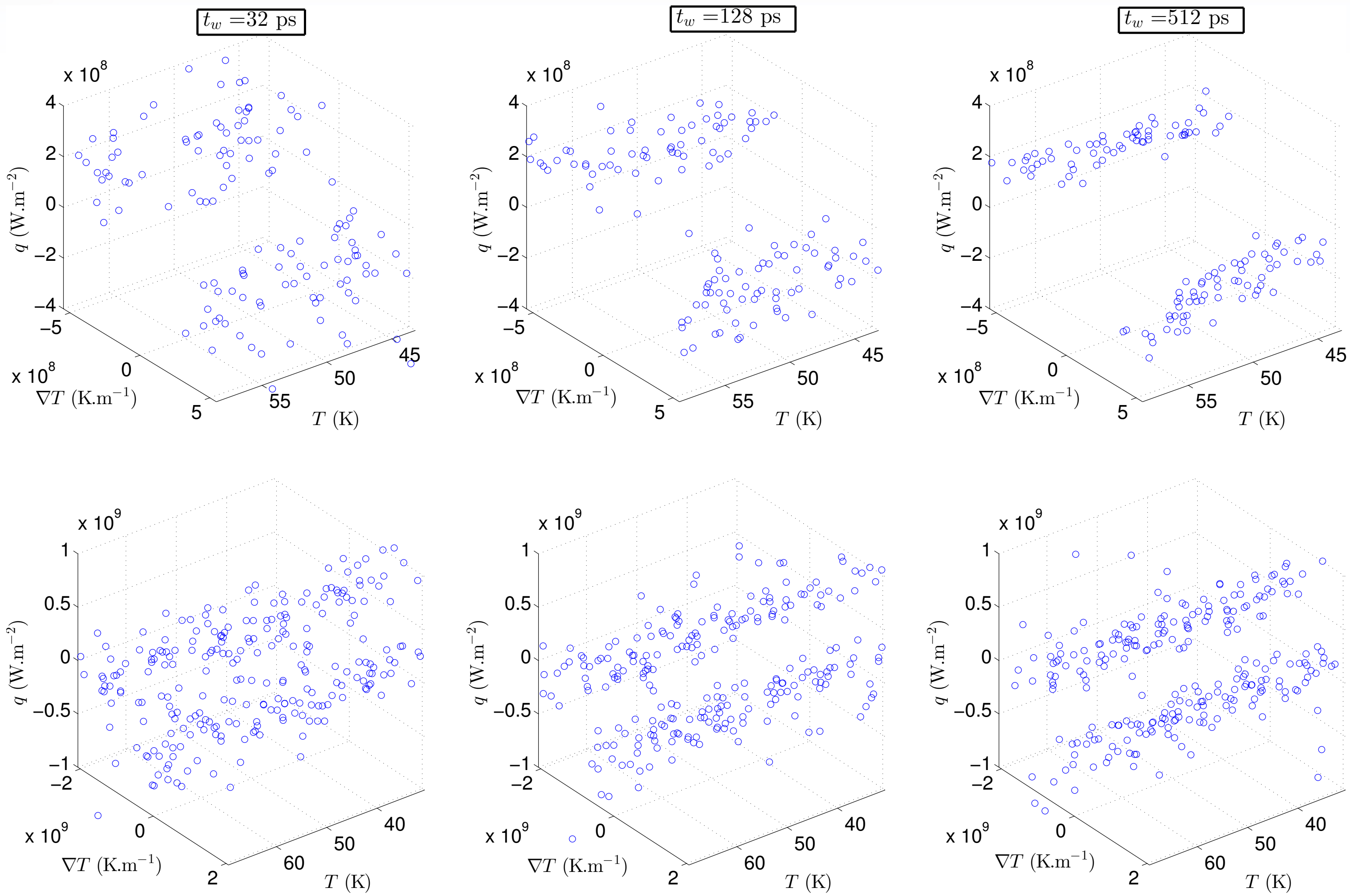
$$k = \sum_{k=0}^{P} (A^k - B^k T) \Psi^k(\xi)$$

Not all PC coefficients follow the central limit theorem

Global vs. Local Sample Locations

Local

Global



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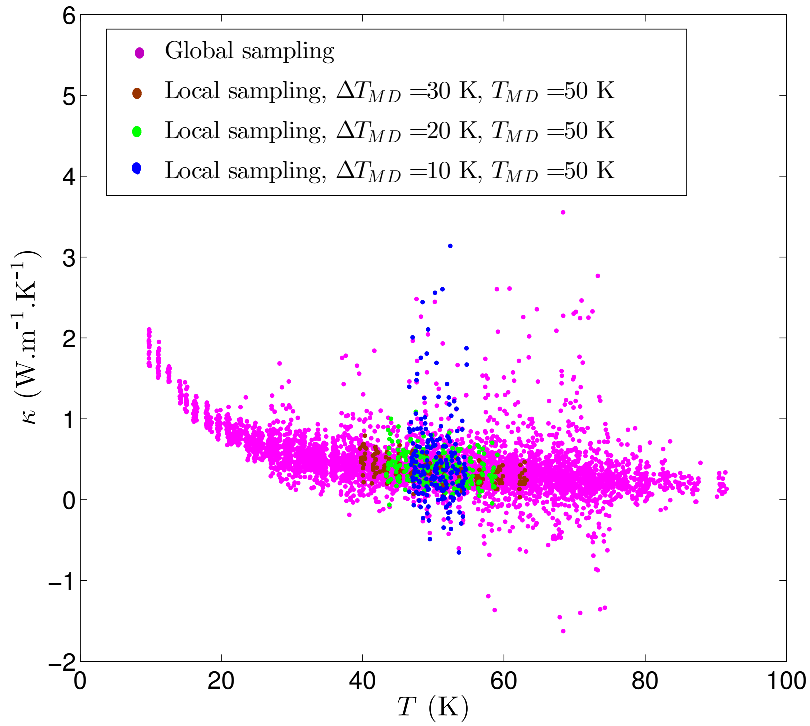
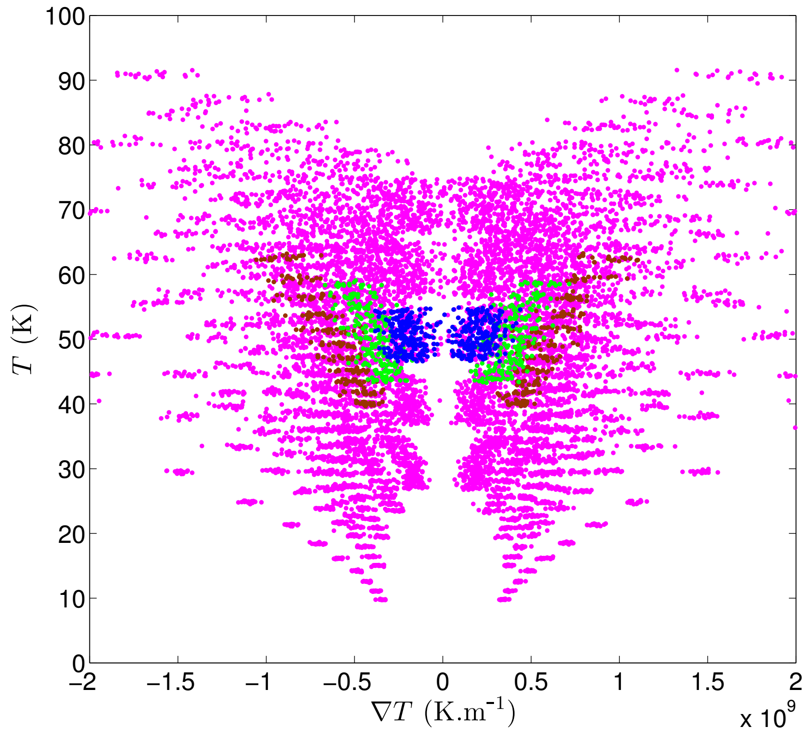
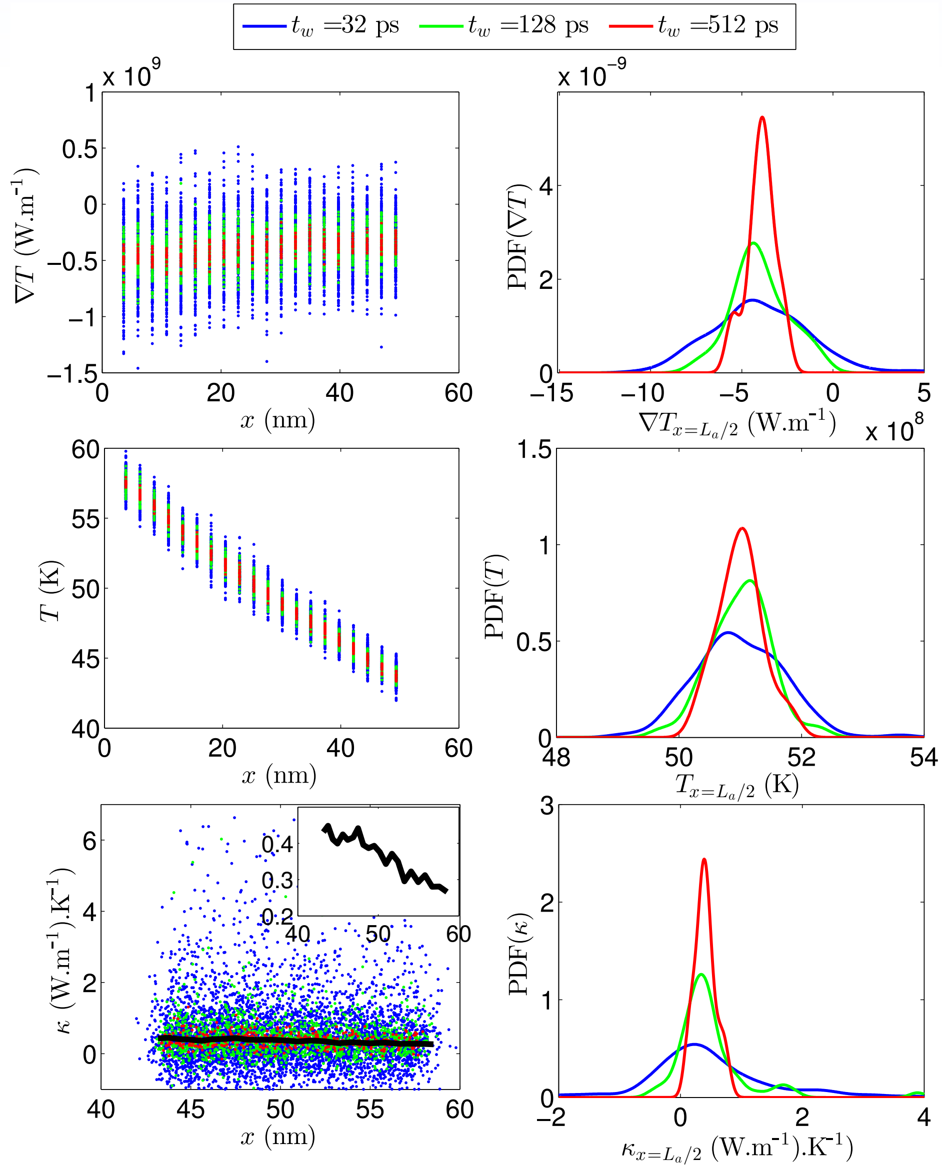
Global vs. Local Data





Raw Atomistic Data



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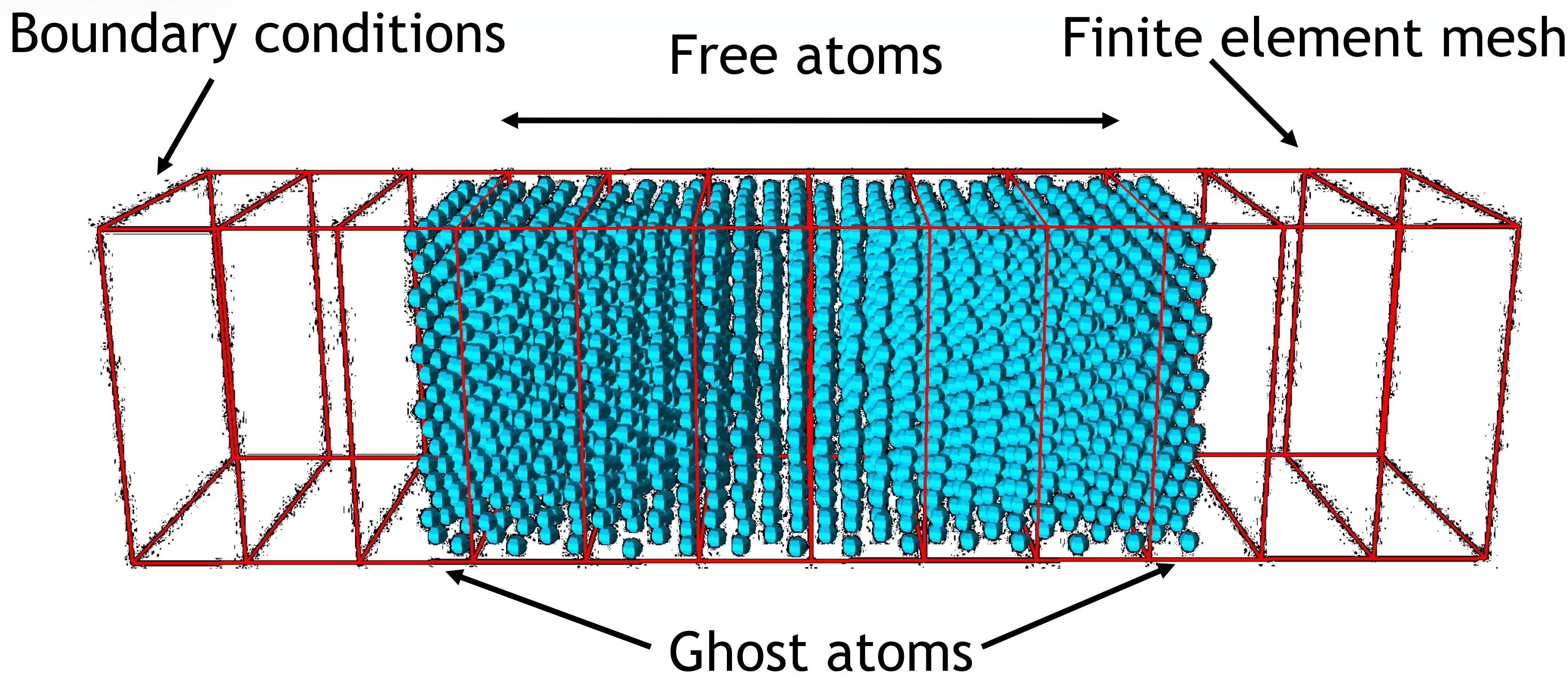
Example: Atomistic-to-Continuum Thermal Coupling



Continuum laws break down at small length scales as the discrete atomistic processes become more pronounced.

Atomistic simulations have a bounding length scale as only a finite amount of atoms can be simulated by a computer.

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Several approaches have made progress at deriving constitutive relations from this perspective

Many existing theories and methods for coupling two distinct mathematical models at different scales.

1. The homogenization methods identify large-scale governing equations through asymptotic analysis, but result in an infinite number of small-scale PDEs. They are restricted to cases of large scale separation.

2. The *computational methods* postulate a single large- and small-scale set of equations. They are based on coarse-graining and they are applicable to cascade scales. However, the lack of a series representation for the solution implies uncertainty is present.

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The Computational Method in Atomistic-to-Continuum Thermal Coupling

- Constant coefficient Fourier's law heat transfer for the continuum:

$$\frac{\partial}{\partial t}\left(\rho c_v \theta\left(\mathbf{x}, t\right)\right) = \nabla \cdot \left(\kappa \nabla \theta\left(\mathbf{x}, t\right)\right)$$

- Finite element projection for atomistic temperature evolution

$$3k_B \sum_J \frac{\partial}{\partial t}\left(N_I N_J \theta_J\right) = \sum_\alpha N_{I\alpha}\left(\mathbf{v}_\alpha \cdot \mathbf{f}_\alpha\right)$$

- Coupling constraints to enforce energy conservation

$$\sum_\alpha N_{I\alpha}\left(\frac{\partial \Phi}{\partial \mathbf{x}_\alpha} \cdot \mathbf{v}_\alpha + \mathbf{v}_\alpha \cdot \mathbf{f}_\alpha\right) + \int_{\Gamma_{MD}} N_{I\alpha} \mathbf{n}_{md} \cdot q^h \, dA = 0$$

* G.J. Wagner, R.E. Jones, J.A. Templeton, M.L. Parks. An atomistic-to-continuum coupling method for heat transfer in solids. Comp. Meth. App. Mech. Eng. 197(41–42):3351–3365, 2008.

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