Deterministic phonon transport and applications in nanoscale heat transfer

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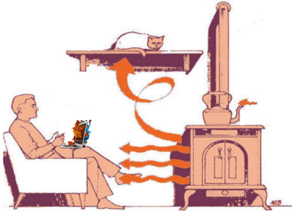
Into to Nuclear Engineering
Tuscon, AZ

Heat transfer

It surrounds us



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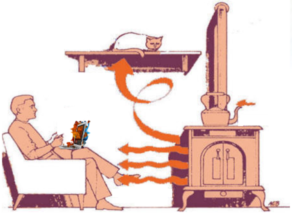

Heat transfer
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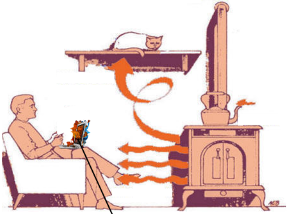
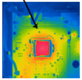
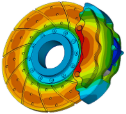
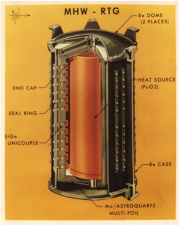
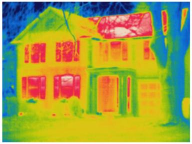
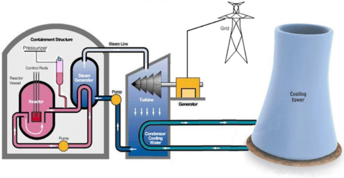
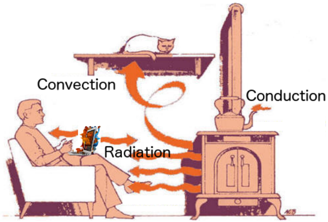



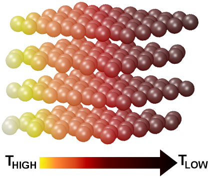



Heat transfer

Mechanisms of conduction





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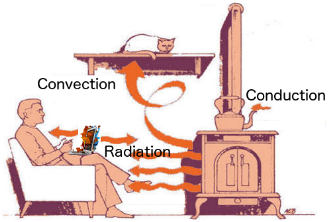

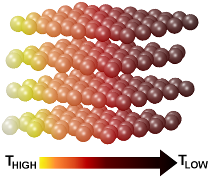

Fourier's law of conduction

$$\mathbf{q}(\mathbf{r}) = -\kappa \nabla T(\mathbf{r}) \qquad (1)$$

Heat transfer

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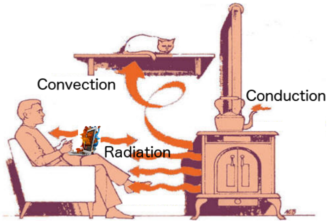

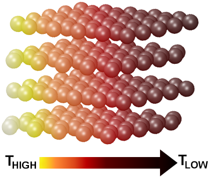

Fourier's law of conduction

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Heat equation

$$\rho C_v \frac{\partial T}{\partial t} - \nabla \cdot (\kappa \nabla T) = \dot{\mathbf{q}}_V \quad (2)$$

Establishing motivation

Conduction at the atomic scale

- Thermal carriers
  - Electrons (metallics)
  - Phonons (insulators)

$$\mathbf{q}(\mathbf{r}) = -\kappa \nabla T(\mathbf{r}) \qquad (3)$$

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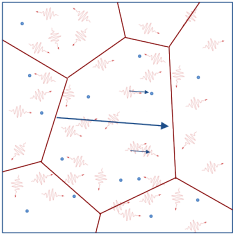

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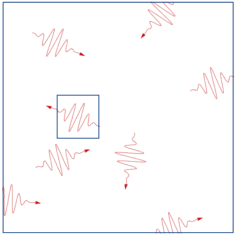

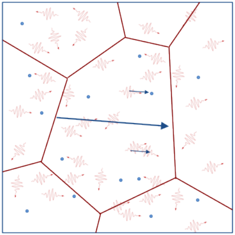

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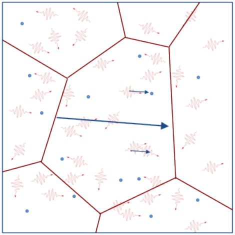

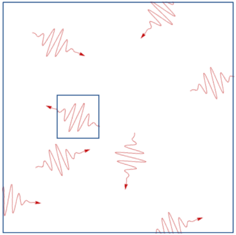

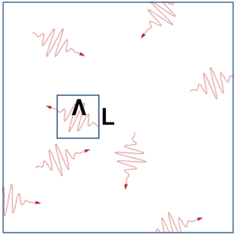

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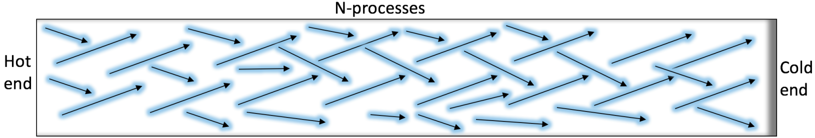
What are phonons?



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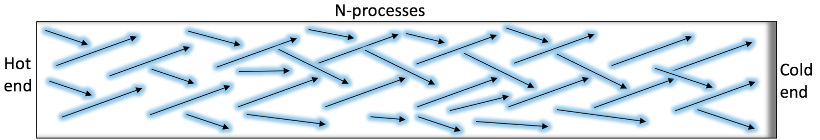
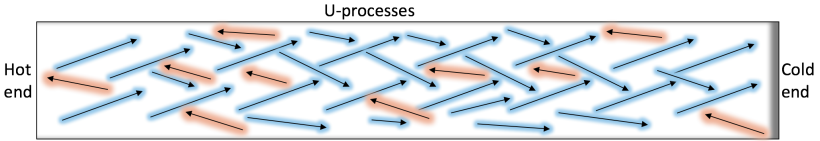
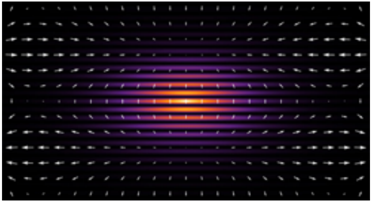




Establishing motivation

What is the importance?

In engineering applications, we use material properties like heat capacity (C_v) in many critical simulations and calculations



$$C_v = \left.\frac{\partial U}{\partial T}\right|_v$$

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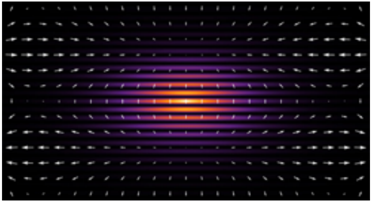

$$C_v = \left.\frac{\partial U}{\partial T}\right|_v$$

Internal energy in an insulator

$$U(\mathbf{r}) = \int_0^\infty \hbar\omega\mathbb{D}(\omega)\langle n_{\mathrm{BE}}\rangle d\omega$$

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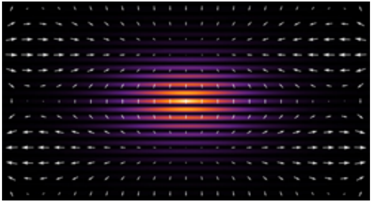

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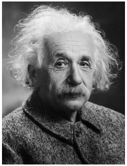
Internal energy in an insulator

$$U(\mathbf{r}) = \int_0^\infty \hbar\omega \mathbb{D}(\omega) \langle n_{\mathrm{BE}} \rangle d\omega$$

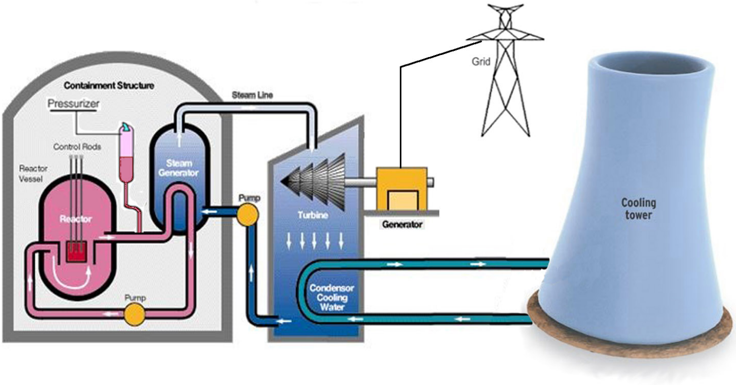
Bose-Einstein statistics

$$\langle n_{\mathrm{BE}} \rangle = \frac{1}{e^{\frac{\hbar\omega}{k_B T(\mathbf{r})}} - 1}$$



Why should you care?
Nuclear engineering applications

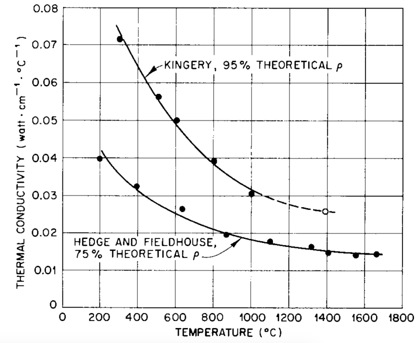
Our (lack of) understanding of materials is the limiting factor in the majority of all engineering disciplines



Establishing motivation

κ in nuclear fuels

- Historically, experiments inform empirical models to determine κ in UO_2
  - Experimental results limited to specific operating temperatures and burnup



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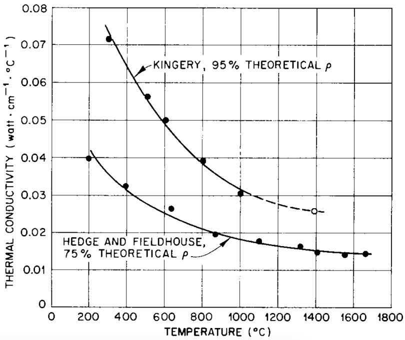

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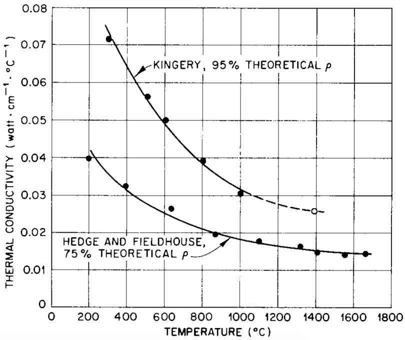

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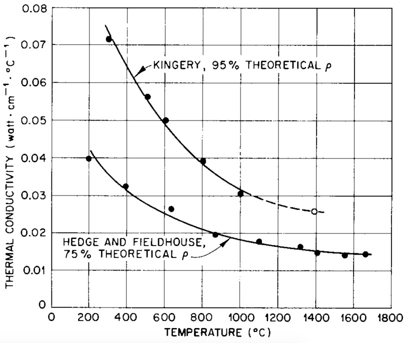
  - Historical approach does not consider evolving microstructure



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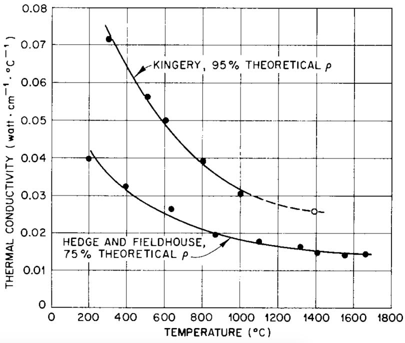
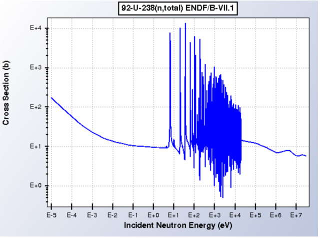
  - Must interpolate between data points to determine κ with associated uncertainty
- κ depends on many factors
  - Historical approach does not consider evolving microstructure
  - Difficult to characterize κ in a dynamic irradiation environment

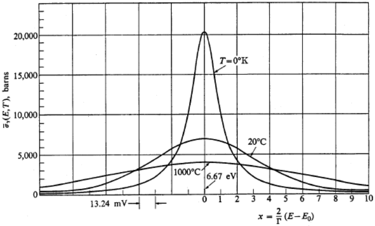


Establishing motivation

Temperature effects in nuclear fuels

- The neutron interaction cross section (σ) is tightly coupled to temperature through *Doppler broadening*
  - Doppler broadening widens the resonances of cross sections; changes spectrum of absorption, scattering or fission

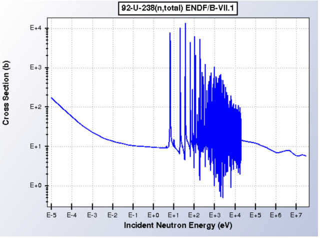


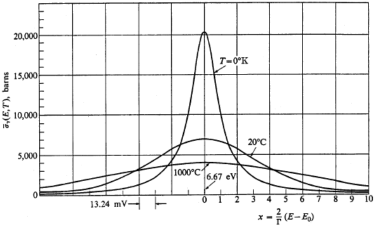


Establishing motivation

Temperature effects in nuclear fuels

- The neutron interaction cross section (σ) is tightly coupled to temperature through *Doppler broadening*
  - Doppler broadening widens the resonances of cross sections; changes spectrum of absorption, scattering or fission
- Quantifying temperature distributions beneficial in characterizing these physics

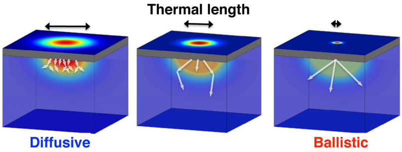




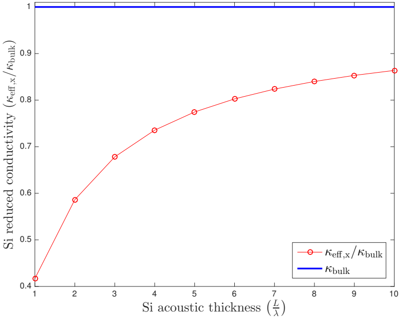
Establishing motivation

Length scale dependence

- Dominant energy carriers in ceramic/insulators are *phonons*



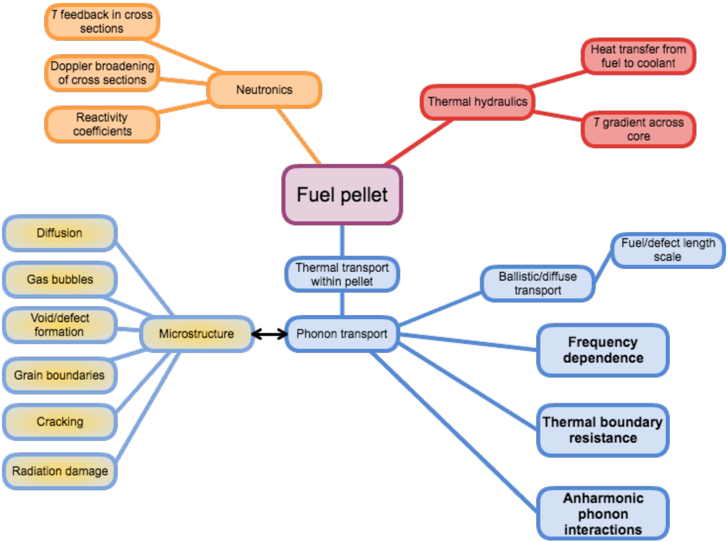
- Acoustic thickness of materials
  - Distance phonons must travel across spatial domain affects contribution to $\mathbf{q}$



- Characterization of bulk κ vs. effective κ
  - Thermal transport behaves differently at reduced length scales
  - Thickness of material vs. Λ

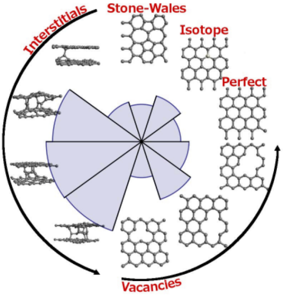
Establishing motivation

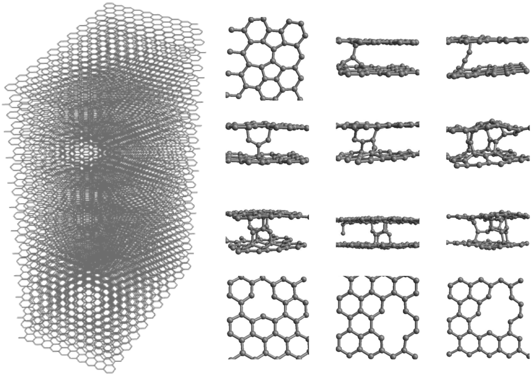
Multiphysics of UO_2



Materials properties in nuclear graphite

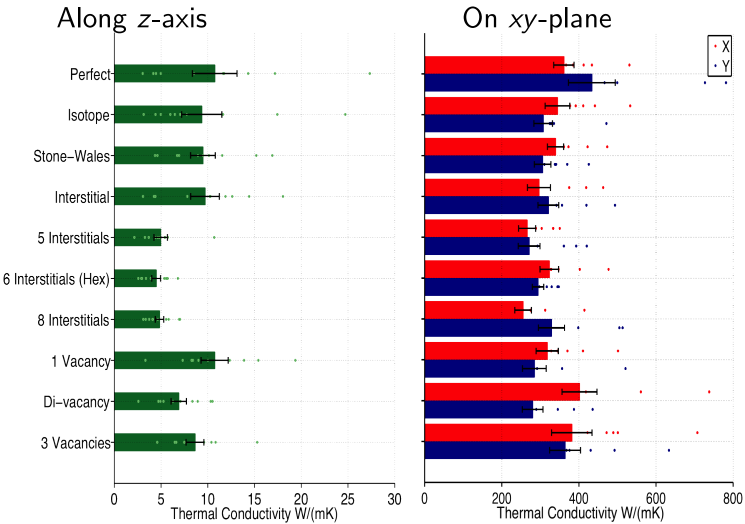
Importance of microstructure on phonon scattering





Materials properties in nuclear graphite

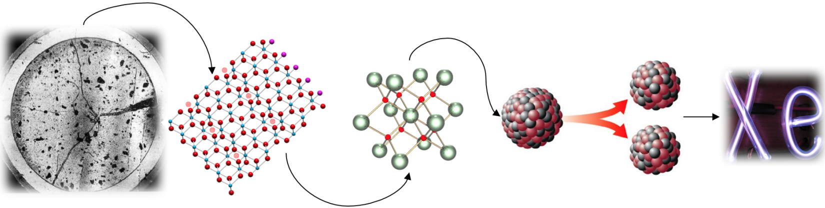
Directional dependence of heat transport



Establishing motivation

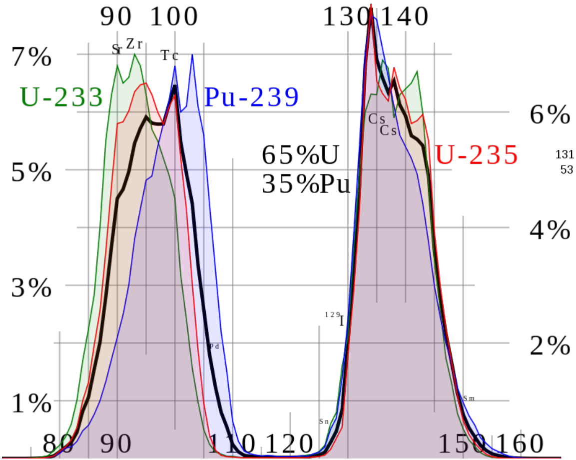
Defect influence on thermal transport in UO_2

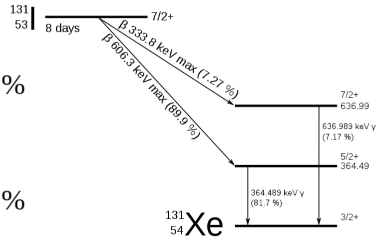
- Presence of defects can severely affect heat transport
  - Grain boundaries
  - Plutonium evolution
  - Fuel cracking
  - Fission products $\rightarrow$ coalescence of xenon forms bubble structures
- Defect presence causes increased phonon scattering, lowering heat flux and κ
- This is a complex, *multiphysics* challenge



Establishing motivation

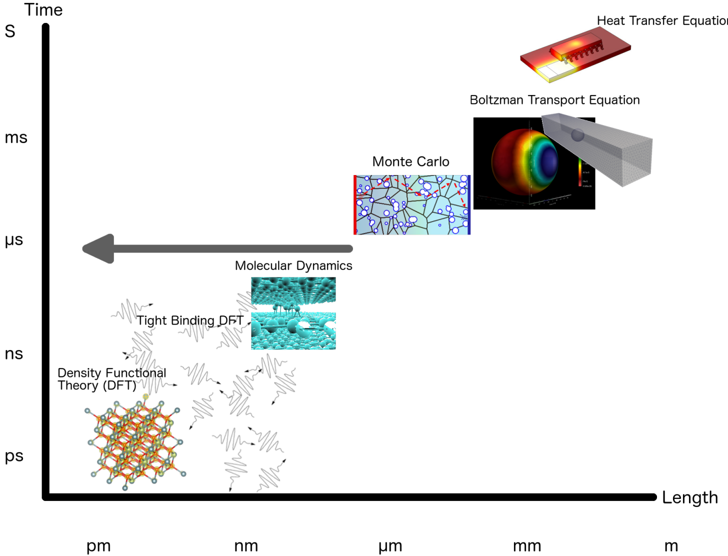
Why xenon?





Establishing motivation

Length and time scales span many orders of dimension



Numerical simulation of phonon transport

Equation framework

Steady state Boltzmann transport equation (BTE) for phonons

$$\left|\mathbf{v}\left(\omega,p\right)\right|\hat{\mathbf{\Omega}}\cdot\nabla f\left(\mathbf{r},\hat{\mathbf{\Omega}},\omega,p\right)=\left.\frac{\partial f\left(\mathbf{r},\hat{\mathbf{\Omega}},\omega,p\right)}{\partial t}\right|_{\text{coll.}}, \qquad (4)$$

applying the single mode relaxation time approximation (RTA)

$$\left.\frac{\partial f}{\partial t}\right|_{\text{coll.}}=\frac{f^{0}\left(\mathbf{r},T,\omega,p\right)-f\left(\mathbf{r},\hat{\mathbf{\Omega}},\omega,p\right)}{\tau\left(\omega,p\right)}. \qquad (5)$$

Multiply phonon state variables into phase space distribution, yielding *angular phonon radiance*

$$\psi\left(\mathbf{r},\hat{\mathbf{\Omega}},\omega,p\right)=\frac{1}{4\pi}f\hbar\omega\left|\mathbf{v}\left(\omega,p\right)\right|\mathbb{D}\left(\omega,p\right).$$

Numerical simulation of phonon transport

Equation framework

- Phonon *equilibrium* radiance, ϕ^0
  - Nonlinear in temperature
  - Function of Bose-Einstein statistical distribution f^0

$$\phi^0\left(\mathbf{r},\omega,T,p\right)=\frac{1}{4\pi}\int_{\omega}\frac{\hbar\omega\left|\mathbf{v}\left(\omega,p\right)\right|\mathbb{D}\left(\omega,p\right)}{\exp\left[\frac{\hbar\omega}{k_B T(\mathbf{r})}\right]-1}d\omega \tag{6}$$

- Mean free path of phonons
  - Average distance between collision events; phonon collisions are numerically treated as purely scattering
  - $\tau\left(\omega,p\right)$ from RTA, lumped relaxation time via Mattheisen's rule

$$\Lambda\left(\omega,p\right)=\left|\mathbf{v}\left(\omega,p\right)\right|\tau\left(\omega,p\right) \tag{7}$$

Numerical simulation of phonon transport

Self-adjoint phonon BTE

- Self-Adjoint Angular Flux (SAAF) formulation of BTE for neutrons developed by Morel and McGee
- We can manipulate BTE for phonons in a similar way
  - Self-adjoint operators are their own adjoint, i.e., $\phi\left(x\right), \psi\left(x\right) \in [a, b]$, $\phi\left(a\right) = \phi\left(b\right) = 0$ and $\psi\left(a\right) = \psi\left(b\right) = 0$

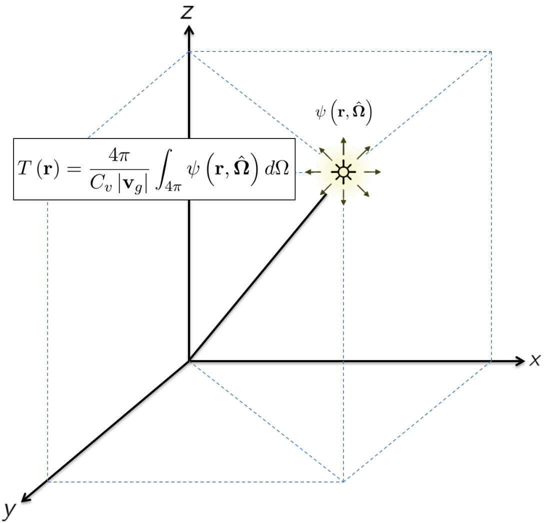
$$\int_a^b \frac{d^2}{dx^2}\psi\left(x\right) \cdot \phi\left(x\right) dx = \int_a^b \psi\left(x\right) \cdot \frac{d^2}{dx^2}\phi\left(x\right) dx \tag{8}$$

- Eq. 9 is the SAAF BTE for phonons

$$\begin{aligned}
-\hat{\mathbf{\Omega}} \cdot \nabla \left[\Lambda\left(\omega, p\right) \hat{\mathbf{\Omega}} \cdot \nabla \psi\left(\mathbf{r}, \hat{\mathbf{\Omega}}, \omega, p\right)\right] + \frac{1}{\Lambda\left(\omega, p\right)} \psi\left(\mathbf{r}, \hat{\mathbf{\Omega}}, \omega, p\right) \\
= \hat{\mathbf{\Omega}} \cdot \nabla \phi^0\left(\mathbf{r}, \omega, p\right) + \frac{1}{\Lambda\left(\omega, p\right)} \phi^0\left(\mathbf{r}, \omega, p\right)
\end{aligned} \tag{9}$$

Numerical simulation of phonon transport

Output quantities



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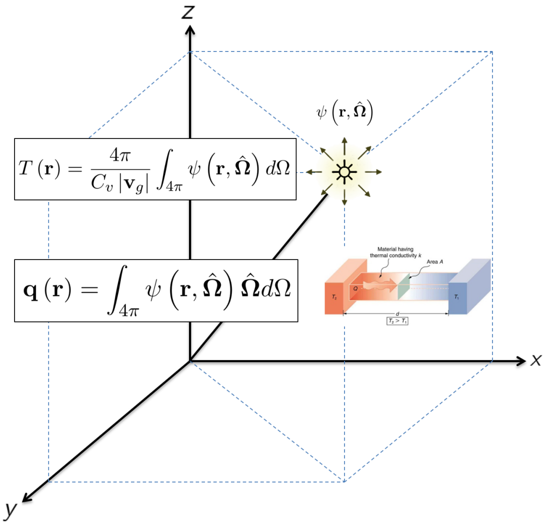


$$T\left(\mathbf{r}\right) = \frac{4\pi}{C_v\left|\mathbf{v}_g\right|}\int_{4\pi}\psi\left(\mathbf{r},\hat{\mathbf{\Omega}}\right)d\Omega$$

$$\mathbf{q}\left(\mathbf{r}\right) = \int_{4\pi}\psi\left(\mathbf{r},\hat{\mathbf{\Omega}}\right)\hat{\mathbf{\Omega}}d\Omega$$

Numerical simulation of phonon transport

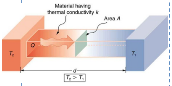
Output quantities

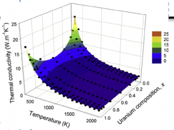
$$\psi\left(\mathbf{r},\hat{\mathbf{\Omega}}\right)$$

$$T\left(\mathbf{r}\right)=\frac{4\pi}{C_v\left|\mathbf{v}_g\right|}\int_{4\pi}\psi\left(\mathbf{r},\hat{\mathbf{\Omega}}\right)d\Omega$$

$$\mathbf{q}\left(\mathbf{r}\right)=\int_{4\pi}\psi\left(\mathbf{r},\hat{\mathbf{\Omega}}\right)\hat{\mathbf{\Omega}}d\Omega$$

$$\left\langle\kappa_{\text{eff},x}\left(\mathbf{r}\right)\right\rangle=-\frac{\int\mathbf{e}_x\cdot\mathbf{q}\left(\mathbf{r}\right)d^3\mathbf{r}}{\int\mathbf{e}_x\cdot\nabla T\left(\mathbf{r}\right)d^3\mathbf{r}}$$





Numerical simulation of phonon transport

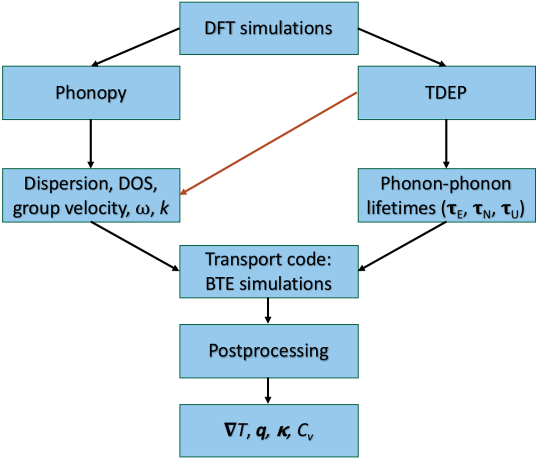
Rattlesnake

Rattlesnake is written in the MOOSE framework and solves the SAAF form of neutron transport equation

- Continuous and discontinuous finite element method spatial discretization with `libMesh`
- Discrete ordinates (S_N) and spherical harmonics (P_N) angular discretization
- Linear solvers, preconditioners from `PETSc`
- Nonlinear diffusion acceleration (NDA) to provide convergence acceleration
- Massively parallelizable

Numerical simulation of phonon transport

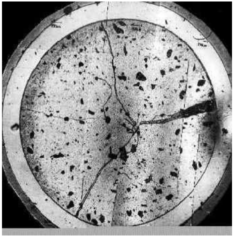
Simulation workflow

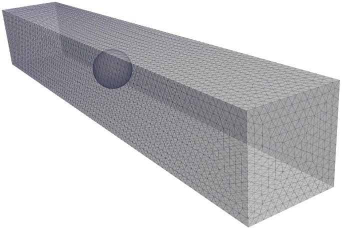


Phonon transport in UO_2

Problem description

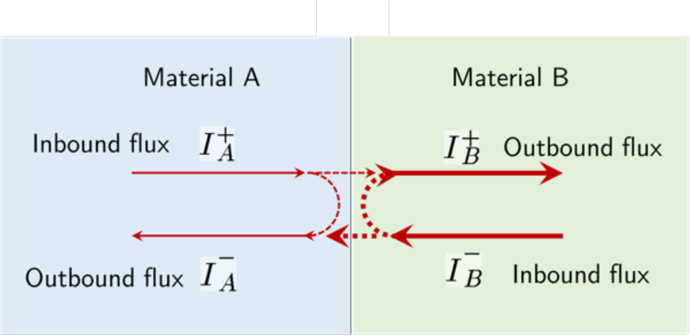
- Simulation of κ in UO_2 with xenon bubble defect
  - Comparison to MD result from Du et al.
  - 3.8 nm x 3.8 nm x 25 nm spatial domain, 30k tetragonal elements
  - S_{20}, $\Delta T = 1$ K, $\epsilon = 10^{-6}$
  - Incident sources on *yz*-planes, reflecting boundaries otherwise
  - Harter JR, Oliveira L, Truszkowska A, Palmer TS, Greaney PA. "Deterministic Phonon Transport Predictions of Thermal Conductivity in Uranium Dioxide With Xenon Impurities." ASME. *J. Heat Transfer*. 2018;**140**(5)





Gray transport in UO_2

Phonon scattering at internal boundaries

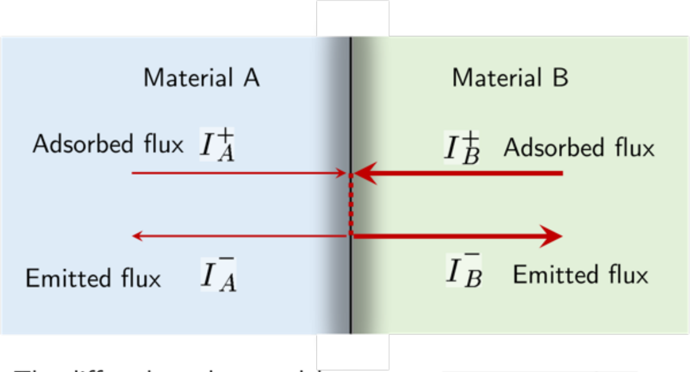


With no temperature gradient detailed balance requires:

$$T_{AB} I_A^o = T_{BA} I_B^o$$

Gray transport in UO_2

Phonon scattering at internal boundaries

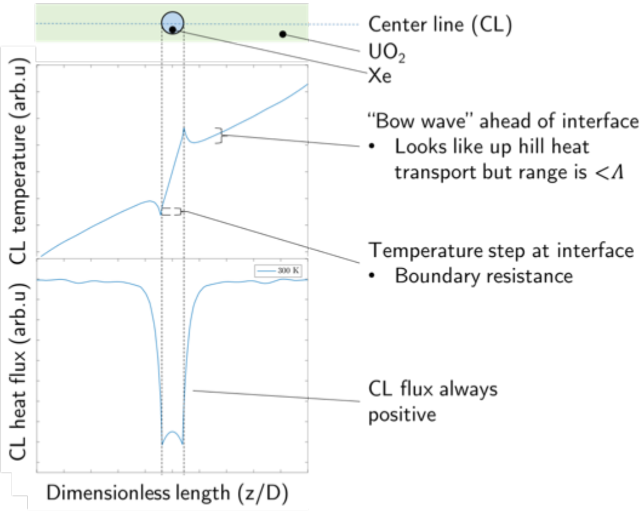


The diffuse boundary model meets this by assuming all flux adsorbed by the boundary and re-emitted:

$$T_{AB} = \frac{I_B^o}{I_A^o + I_B^o}$$

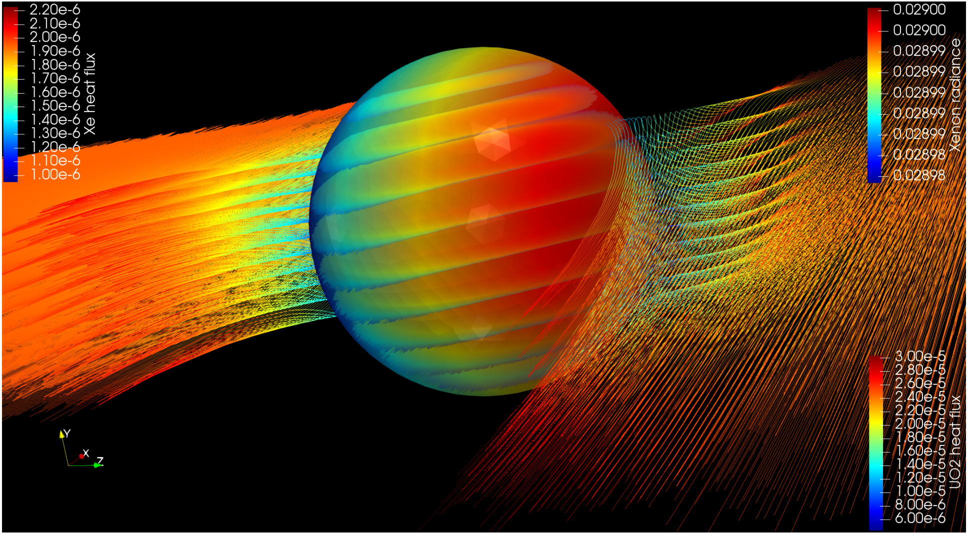
Gray transport in UO_2

Two phenomena at interfaces



Gray transport in UO_2

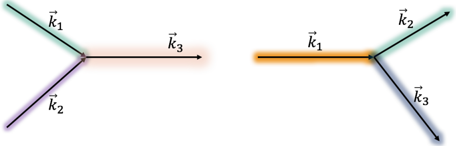
Boundary resistance at UO_2 – Xe interface



Anharmonic effects on heat transport

Phonon strangeness

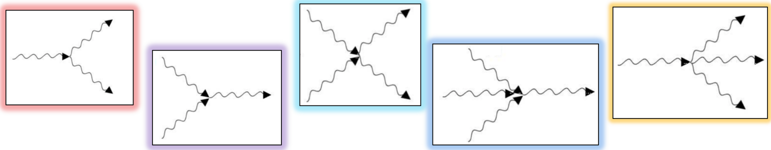
- Anharmonic interactions multiple-phonon scattering events responsible for parts of thermal resistance, can occur at all temperatures
- Conservation of energy (CoE) enforced, but conservation of crystalline momentum (CoM) is not
  - Normal process $\rightarrow$ CoE, CoM
  - Umklapp process $\rightarrow$ CoE
  - Transition probabilities based on perturbations of system energy determine these processes



Anharmonic effects on heat transport

Temperature effects on scattering

- At high temperatures, Umklapp processes predicted to contribute large fraction of thermal resistance
  - However, other processes may contribute as much, or more (Si with impurities)
  - We are interested in UO_2 at operating temperatures, simulating contributions from 3-phonon scattering should be included
  - Effects can change based on *where* scattering happens



Anharmonic effects on heat transport

SMRTA vs. explicit collisions

- In the SMRTA we assumed the form of the collision operator to be

$$\frac{\partial f_{\mathrm{coll.}}}{\partial t} = -\frac{\psi - \phi^0}{\tau} \tag{10}$$

- Nonlinear scattering integrals can describe 3-phonon processes, e.g.,

$$\frac{\partial f_{\mathrm{coll,pp}}}{\partial t} = \int\int \Big\{ \Big[\underbrace{\psi_{\mathbf{k}} \psi_{\mathbf{k}'} \left(1 + \psi_{\mathbf{k}''}\right)}_{\textit{forward}} - \underbrace{\left(1 + \psi_{\mathbf{k}}\right)\left(1 + \psi_{\mathbf{k}'}\right) \psi_{\mathbf{k}''}}_{\textit{reverse}} \Big] \mathbb{L}_{\mathbf{k}\mathbf{k}'}^{\mathbf{k}''} + \Big[\underbrace{\psi_{\mathbf{k}} \left(1 + \psi_{\mathbf{k}'}\right)\left(1 + \psi_{\mathbf{k}''}\right)}_{\textit{forward}} - \underbrace{\left(1 + \psi_{\mathbf{k}}\right) \psi_{\mathbf{k}'} \psi_{\mathbf{k}''}}_{\textit{reverse}} \Big] \mathbb{L}_{\mathbf{k}}^{\mathbf{k}'\mathbf{k}''} \Big\} d\mathbf{k}' d\mathbf{k}''$$

  - How far out of equilibrium are we?

Conclusions

- Simulating heat transport at atomistic scale and bridging gap to engineering scale is the multiphysics nature of complex problems
- Rattlesnake efficiently simulates deterministic multi-frequency phonon transport
  - Intrinsic temperature coupling required to accurately resolve temperatures, heat flux, thermal conductivity

Conclusions
Acknowledgments

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