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A Fractional Step Method for Coupling Molecular Dynamics and Finite Element Models of Thermal Transport

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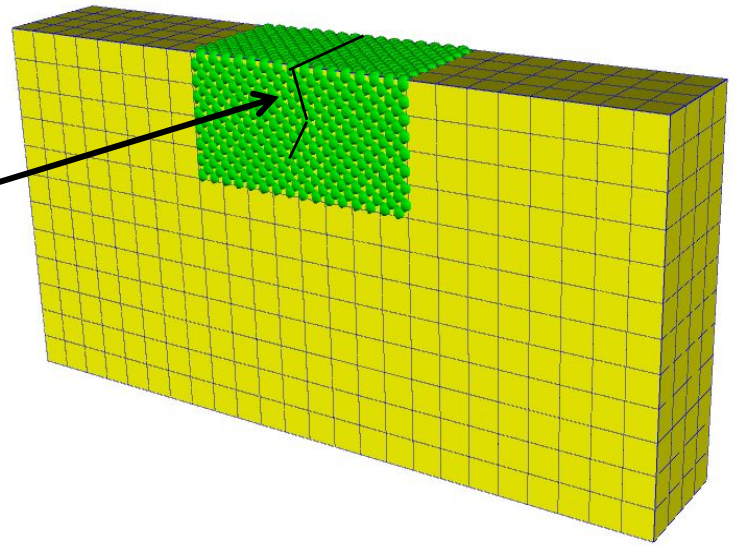
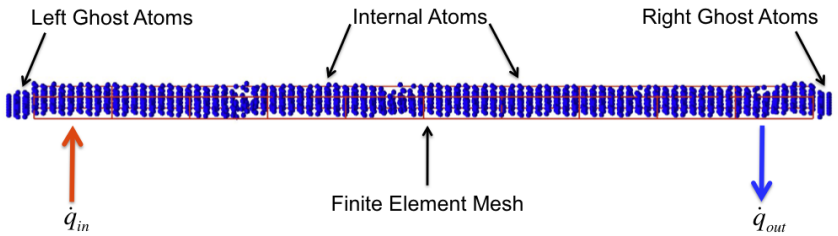
Motivation

- Provide a unified computational framework for finite element (FE) and an molecular dynamics (MD) for problems in which
  1. An atomistic description of material is needed only for a localized region and the dynamical interactions between the FE and MD are important for understanding the system
     - MD cost to simulate entire system atomistically would be prohibitive
     - Dual Statement: FE constitutive models are not of sufficient fidelity for all of the system
     - See Wagner *et al.*
  2. Complex boundary conditions must be applied to an atomic system to understand the physics of interest
     - MD typically does not provide the infrastructure for complex, engineering calculations
     - See Templeton *et al.*
- Specific motivation for improved time integration algorithms
  - Enhanced stability through conservation of energy in the multiscale coupling scheme
  - Enhanced stability by minimizing temperature drift for Dirichlet boundary conditions
  - Ability to consider more complex temperature definitions, e.g. including the potential energy
  - Eulerian frame calculations





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Continuum Heat Equation

- Heat equation with Fourier heat conduction arising from Boltzmann Transport equation for energy conservation:

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

- Finite element discretization leads to a set of ODE's for the nodal temperatures

$$\theta^h\left(\mathbf{x},t\right) = \sum_I N_I\left(\mathbf{x}\right)\theta_I\left(t\right) \implies \frac{\partial}{\partial t}\left(\mathbf{M}\theta\right) = \mathbf{K}\theta$$

$$M_{IJ} = \int_\Omega \rho c_v N_I N_J \, dV$$

$$K_{IJ} = \int_\Omega \kappa \nabla N_I \cdot \nabla N_J \, dV$$

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MD Temperature Definition

- We have to relate the dynamics of atoms to the nodal temperature field

$$E^{MD} = \sum_{\alpha} \frac{1}{2} m_\alpha \left|\mathbf{v}_\alpha\right|^2 + \Phi \implies \begin{aligned} e^k_\alpha &\approx m_\alpha \left|\mathbf{v}'_\alpha\right|^2 / \Delta V_\alpha \\ e^t_\alpha &\approx \left(m_\alpha \left|\mathbf{v}'_\alpha\right|^2 / 2 + \phi'_\alpha\right) / \Delta V_\alpha \end{aligned}$$

- Define restriction operation: MD field → Nodal field
  - E.g. projection, averaging, shape functions…
  - One way: minimize difference between MD and continuum temperature fields

$$\min_{\theta_I} \sum_{\alpha \in \text{atoms}} \left(e_\alpha \Delta V_\alpha - \sum_{I \in \text{nodes}} \rho c_v N_{I\alpha} \theta_I \Delta V_\alpha \right)^2 \Longrightarrow \boxed{\theta_I = \sum_\alpha \hat{N}_{I\alpha} T_\alpha}$$

$$\rho \equiv \frac{m_\alpha}{\Delta V_\alpha}, \quad c_v \equiv \frac{3k_B}{m_\alpha}$$

Dulong-Petit expression for heat capacity of a mono-atomic solid or dense fluid above the Debye temperature

Using row-sum lumping (localization) and atomic quadrature for mass matrix in MD region (thermodynamic consistency)

$$\hat{N}_{I\alpha} = \frac{N_{I\alpha}}{\sum_\beta N_{I\beta}}$$

$$T_\alpha = \frac{1}{3k_B} e_\alpha$$

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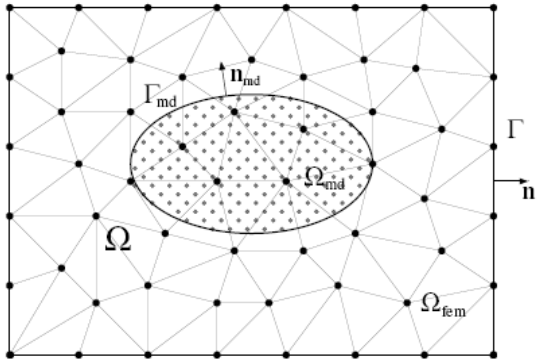
Derivation of Coupled FEM-MD Equations



- Apply Galerkin method to entire domain:
  - Decompose domain:

$$\int_{\Omega} N_I(\mathbf{x}) \frac{\partial}{\partial t}\left(\rho c_v \theta^h(\mathbf{x})\right) dV = \int_{\Omega_{\text{fem}}} N_I(\mathbf{x}) \frac{\partial}{\partial t}\left(\rho c_v \theta^h(\mathbf{x})\right) dV + \sum_{\alpha} \frac{\partial}{\partial t}\left(N_{I\alpha} \rho c_v \theta^h_\alpha \Delta V_\alpha\right)$$

  - Use atomic energy density:

$$\int_{\Omega_{\text{fem}}} N_I(\mathbf{x}) \frac{\partial}{\partial t}\left(\rho c_v \theta^h(\mathbf{x})\right) dV + \sum_{\alpha} \frac{\partial}{\partial t}\left(N_{I\alpha} \rho c_v \theta^h_\alpha \Delta V_\alpha\right) = \int_{\Omega_{\text{fem}}} N_I(\mathbf{x}) \frac{\partial}{\partial t}\left(\rho c_v \theta^h(\mathbf{x})\right) dV + \sum_{\alpha} \frac{\partial}{\partial t}\left(N_{I\alpha} e_\alpha \Delta V_\alpha\right)$$

  - Apply physics:

$$\int_{\Omega_{\text{fem}}} N_I(\mathbf{x}) \frac{\partial}{\partial t}\left(\rho c_v \theta^h(\mathbf{x})\right) dV + \sum_{\alpha} \frac{\partial}{\partial t}\left(N_{I\alpha} \rho c_v \theta^h_\alpha \Delta V_\alpha\right) =$$

$$\int_{\Omega_{\text{fem}}} N_I \nabla \cdot \kappa \nabla \theta^h \, dV + \sum_{\alpha} N_{I\alpha}\left(\mathbf{v}_\alpha \cdot \mathbf{f}_\alpha + \dot{\phi}'_\alpha\right) + \sum_{\alpha} \left(\nabla N_{I\alpha} \cdot \mathbf{v}_\alpha\right) e_\alpha \Delta V_\alpha$$

  - Discretize:

$$\sum_J \frac{\partial}{\partial t}\left(M_{IJ}\theta_J\right) = \sum_J K_{IJ}\theta_J + \sum_{\alpha} N_{I\alpha}\left(\mathbf{v}_\alpha \cdot \mathbf{f}_\alpha + \dot{\phi}'_\alpha\right) + \sum_{\alpha} \left(\nabla N_{I\alpha} \cdot \mathbf{v}_\alpha\right) e_\alpha \Delta V_\alpha$$

Coupling MD Thermostat

- Effects of FEM on MD can be included by prescribing constraints relating the FE and MD dynamics:
  - Temperature constraint

$$\sum_{\alpha} N_{I\alpha}\mathbf{v}_{\alpha}\cdot\mathbf{f}_{\alpha}+\sum_{\alpha} N_{I\alpha}\phi'_{\alpha}+\sum_{\alpha}\left(\nabla N_{I\alpha}\cdot\mathbf{v}_{\alpha}\right)e_{\alpha}\Delta V_{\alpha}-\sum_{J}\frac{\partial}{\partial t}\left(M^{MD}_{IJ}\theta_{J}\right)=0$$

  - Heat flux constraint

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

- Thermostat force exactly cancels the FE boundary flux for the total temperature definition, otherwise there is a partial residual

Governing Equation for the Lagrange Multiplier

- Application of Gauss' principle of least constraint to atomic forces provides an elliptic equation for the Lagrange Multiplier:

$$\sum_{\alpha} N_{I\alpha} K_{\alpha} \sum_{J} N_{J\alpha} \lambda_J = R_I^c$$

- Variable λ is a continuum field defined on the nodes:

$$\lambda\left(\mathbf{x}_{\alpha}\right) = \sum_{I} N_{I\alpha} \lambda_I$$

- Differentiating the constraint w.r.t the new force gives

$$\mathbf{f}_{\alpha} = \mathbf{f}_{\alpha}^{MD} - \frac{m_{\alpha}}{2} \lambda\left(\mathbf{x}_{\alpha}\right) \mathbf{v}_{\alpha}$$

- So atomic degrees of freedom are governed by

$$m_{\alpha} \dot{\mathbf{v}}_{\alpha} = \mathbf{f}_{\alpha}^{MD} - \frac{m_{\alpha}}{2} \sum_{I} N_{I\alpha} \lambda_I \mathbf{v}_{\alpha}$$

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Motivation for the Fractional Step Method

- Fractional step originally developed by Chorin to exactly satisfy elliptic constraints on a a hyperbolic problem
- In this case there are several complicating factors
  - Discrepancy in evolution of kinetic energy in atomic system

$$\Delta K_\alpha = \Delta t \mathbf{v}_\alpha \cdot \mathbf{f}_\alpha + \frac{\Delta t^2}{2} \mathbf{f}_\alpha \cdot \mathbf{f}_\alpha$$

  - No direct evaluation of the time derivative of the potential

$$\frac{\partial \phi_\alpha}{\partial t} = ?$$

  - Drift between change in shape functions and convection

$$\nabla N_{I\alpha} \cdot \mathbf{v}_\alpha \neq \Delta N_{I\alpha}$$

Fractional Step Method for Time Integration

- Gear time integration for FE dynamics: $K_{IJ}\theta_J$
- Velocity-Verlet time integration for MD dynamics: $m_\alpha \dot{\mathbf{v}}_\alpha = \mathbf{f}_\alpha^{MD}$
- Consistent update for MD contribution to FE temperature:

$$\Delta\theta_I = M_{IJ}^{-1} \sum_\alpha N_{I\alpha} e_\alpha \Delta V_\alpha \big|_{t+\Delta t} - M_{IJ}^{-1} \sum_\alpha N_{I\alpha} e_\alpha \Delta V_\alpha \big|_t$$

- Considering the change in nodal energy

$$N_I \Delta E_I = \sum_\alpha N_{I\alpha} \left(\Delta t \mathbf{v}_\alpha \cdot \mathbf{f}_\alpha + \frac{\Delta t^2}{2} m_\alpha^{-1} \mathbf{f}_\alpha \cdot \mathbf{f}_\alpha + \phi'_\alpha \big|_{t+\Delta t} - \phi'_\alpha \big|_t \right)$$

- Enables general temperature definitions to be used
- Eliminates temperature drift between FE and MD systems

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Modified Equation for Exactly Satisfying Constraints

- Second order terms must be account for in the constraint equations

$$\Delta t \sum_{\alpha} N_I^{\alpha} K_{\alpha} \sum_{\mathcal{J}} N_J^{\alpha} \lambda_J - \frac{\Delta t^2}{4} \sum_{\alpha} N_I^{\alpha} K_{\alpha} \left(\sum_{J} N_J^{\alpha} \lambda_J \right) \left(\sum_{K} N_K^{\alpha} \lambda_K \right) = R_I^c$$

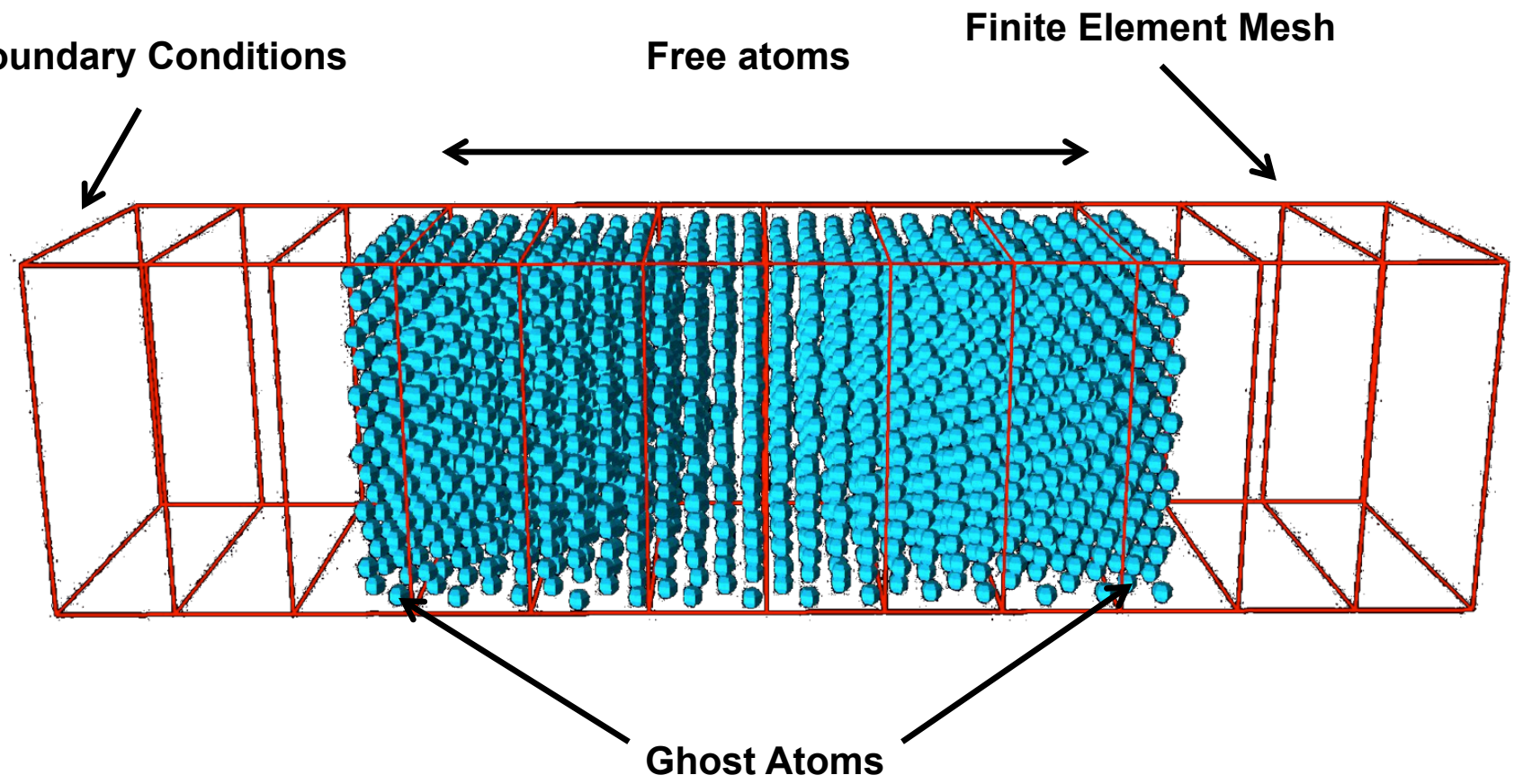
  - Boundary flux when needed is added with the Verlet step

- Solvability criteria gives time step size criterion: $\Delta t \lambda < 1$

- Iterative methods can be use to converge the solution to machine precision for exact constraint enforcement

- Eliminates drift which causes instability with fixed temperature constraints
- Enables exact energy conservation in the coupling algorithm

Demonstration Problems



Lennard-Jones Argon with equilibrium FCC spacing at 20 K

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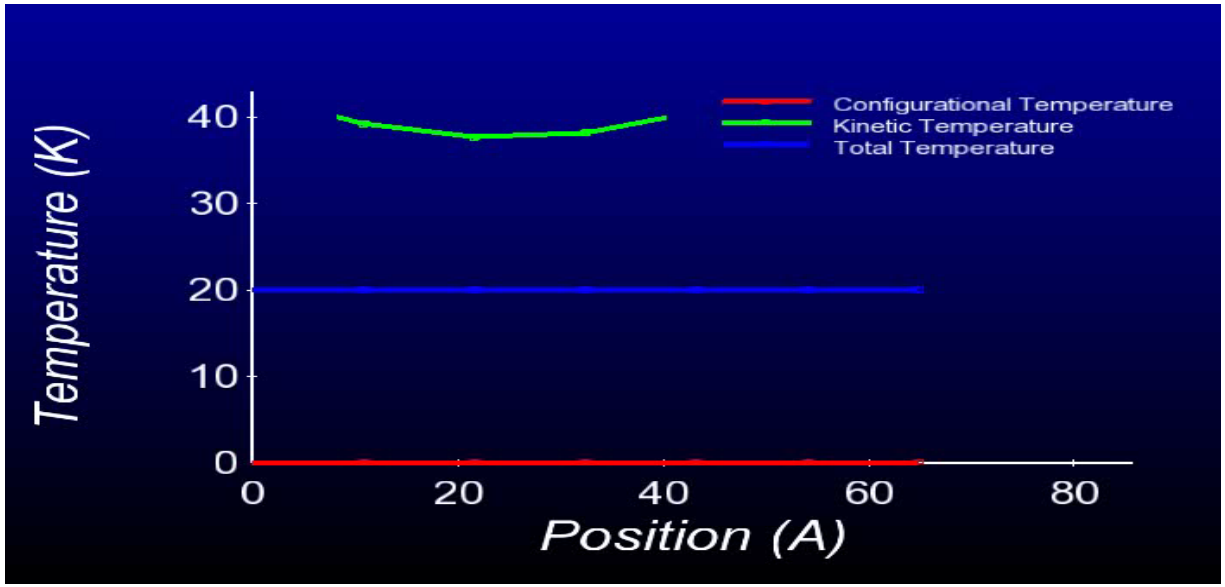
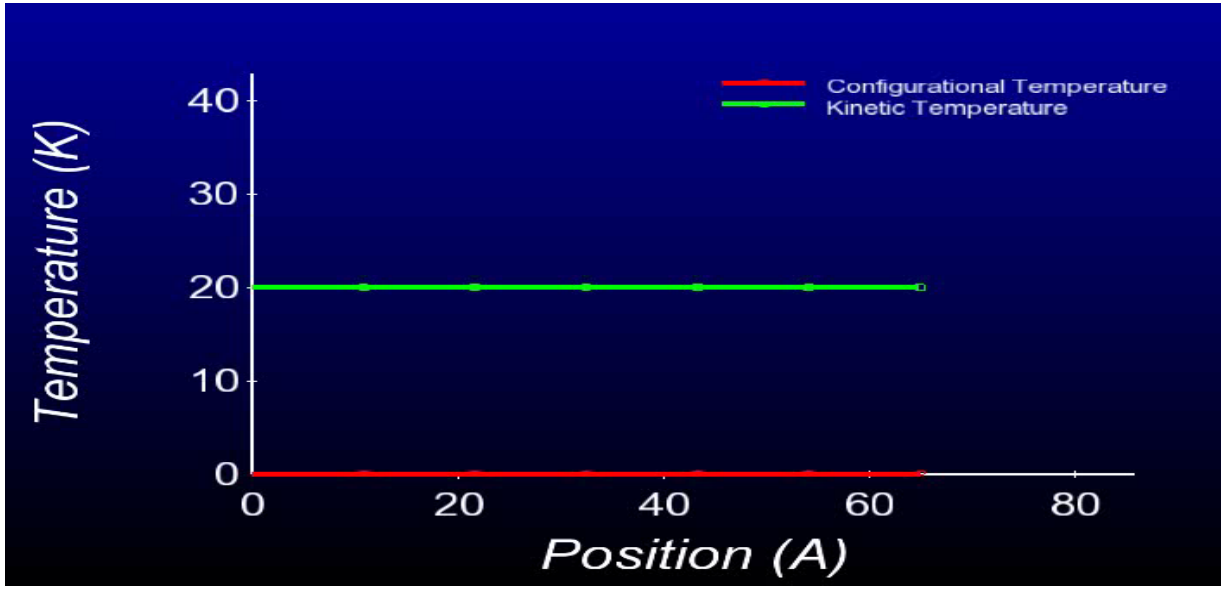
Equilibration to 20 K

Total Temperature



Kinetic Temperature



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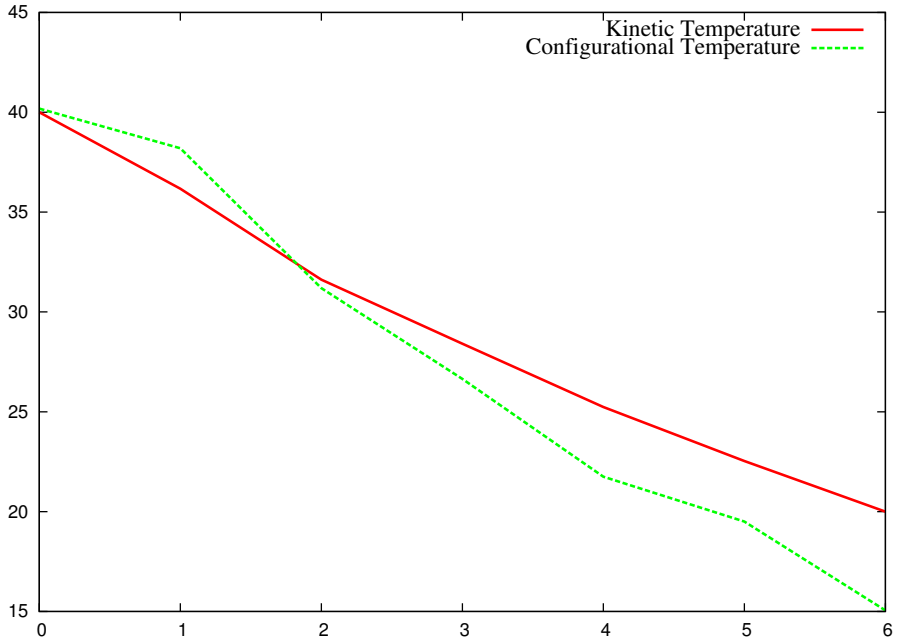
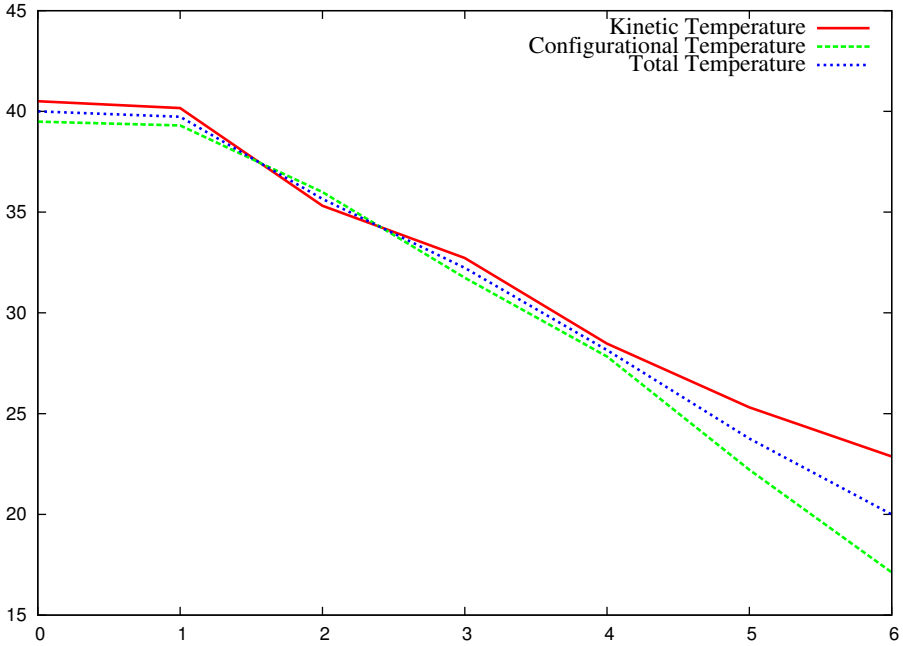
Application of Fixed Temperature BCs



Kinetic Temperature Definition

Total Temperature Definition



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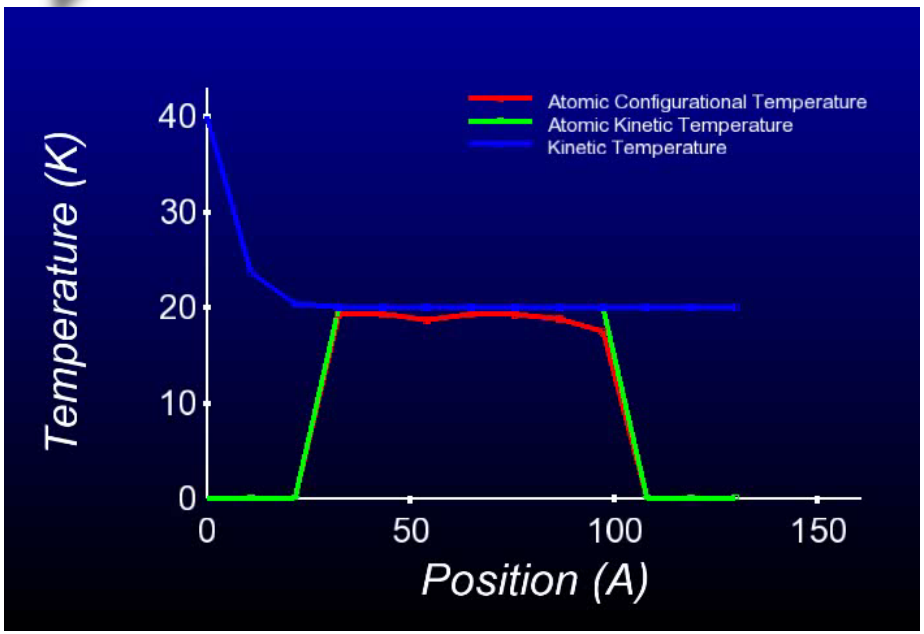
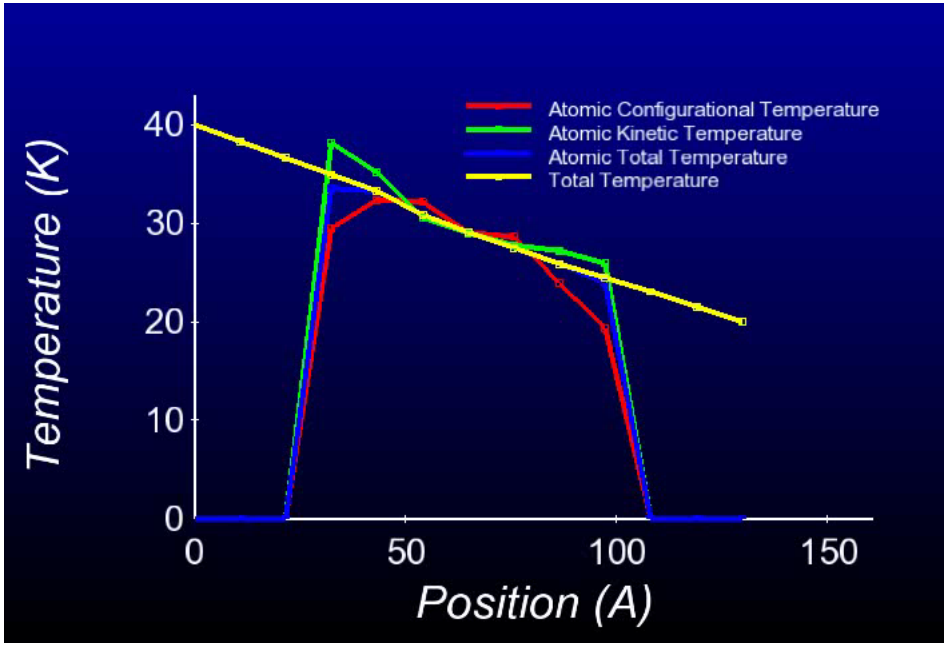
Fluxed-Based Multiscale Coupling





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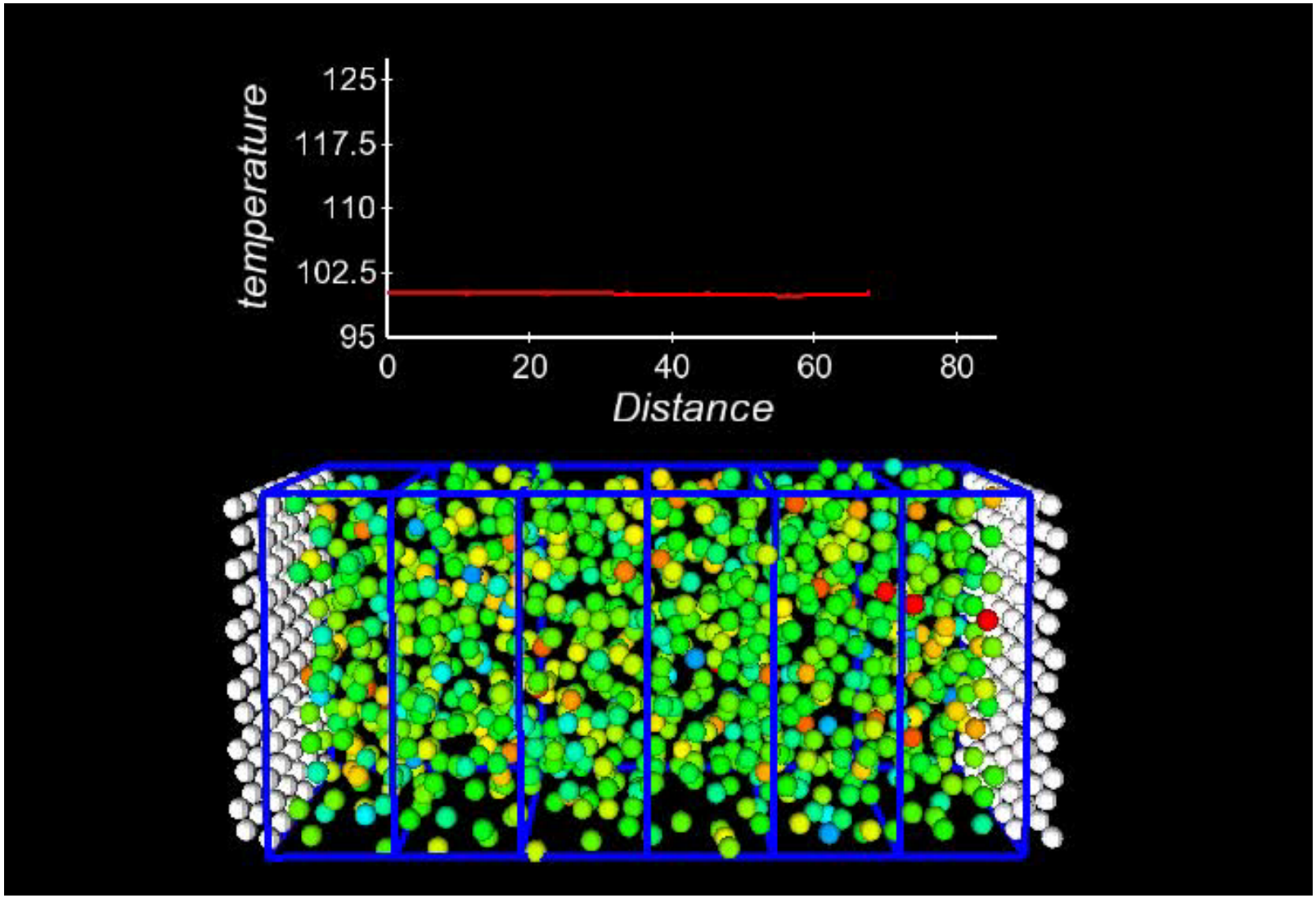
Liquid Argon Temperture BCs



$$\kappa = 1.54 \times 10^{-9}\ \mathrm{AMU/fs^3\ K}$$

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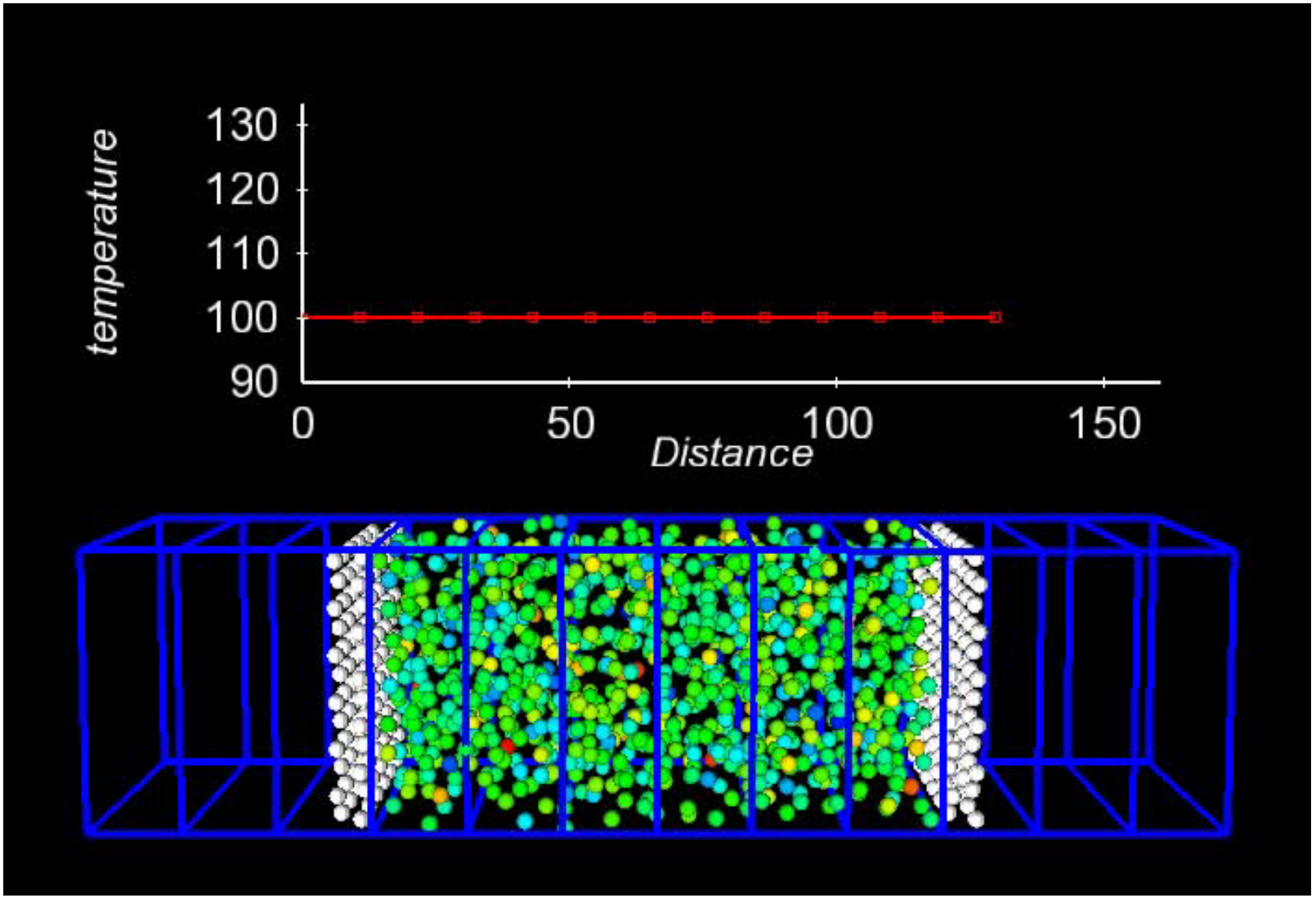
Liquid Argon Flux Coupling



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Conclusions

- A generalized set of equations for coupled MD-FE thermal transport has been derived
  - Appropriate for Lagrangian or Eulerian frames
  - Can use a large class of temperature definitions
- Hyperbolic transport equations were solved consistently with an elliptic constraint using a fractional step method
  - Increases numerical stability by exactly enforcing coupling constraints
- Robustness has been demonstrated in several example problems covering the types of problems of interest
- Future work:
  - Determination of thermal properties of complex systems
  - Assessment of heat propagation through nano-devices
  - Modeling of fluid-solid interfaces