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Coupled Ionic and Electronic Heat Transport at the Nanoscale

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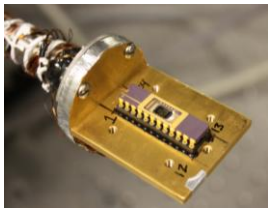
Novel Experimental Work at CINT – Thermal Transport at the Nanoscale

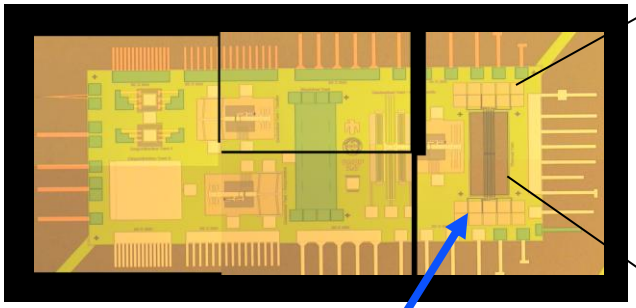
Experimental Goal: Methods to sensitively measure thermal, electrical & thermoelectric properties simultaneously for single nanostructures

In situ TEM to probe structure



J. Huang

Cantilever Array Discovery Platform 2.0

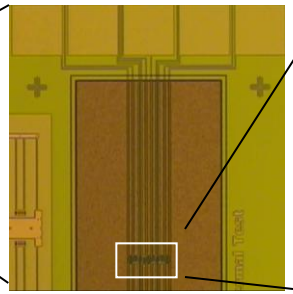


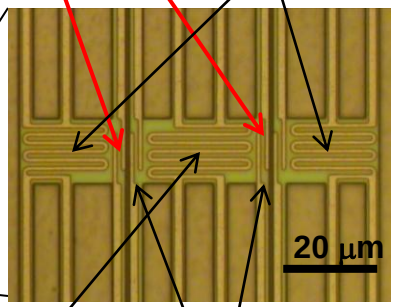
J. Sullivan

NW transport structure

Differential thermal transport

Thermometers

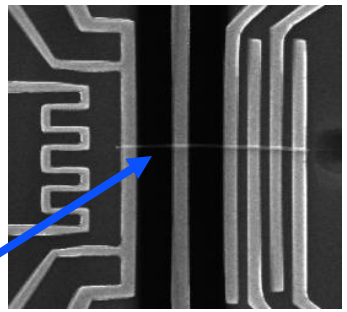




Heater

4-probe transport

Test structure



T. Harris, J. Huang

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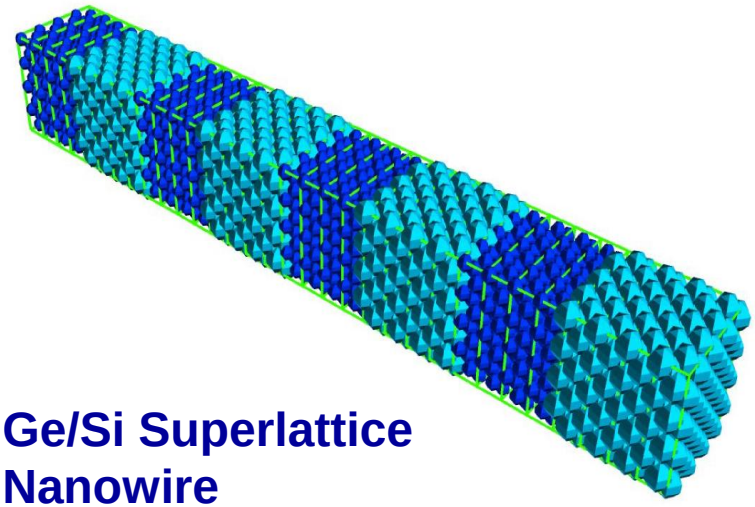
Why Are Electrons Important to Thermal Behavior?

Material	Thermal Conductivity [w/K-m]			Heat Capacity [J/K-kg]			Debye T [K]	e-p coupling [W/K-m^3]
Cu	401	11.2	244	385	394	10.8	345	2.6 X 10^{17}
Si	148	120	0.02	705	888	0.0002	645	1.0 X 10^{11}

- Both phonons and electrons contribute to thermal properties
- At macroscale, electron-phonon equilibrium gives aggregate thermal properties
- This is not necessarily true at the nanoscale!



Ge/Si Superlattice Nanowire

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Two Temperature Model (TTM)

- Second moment of Boltzmann equation
- Two temperatures: phonon θ_p and electron θ_e
- Two coupled, diffusive systems

$$c_p \frac{\partial \theta_p}{\partial t} = \nabla \cdot \left(k_p \nabla \theta_p \right) - g\left(\theta_p - \theta_e \right) + r_p\left(\vec{x}, t \right)$$

$$c_e \frac{\partial \theta_e}{\partial t} = \nabla \cdot \left(k_e \nabla \theta_e \right) - g\left(\theta_e - \theta_p \right) + r_e\left(\vec{x}, t \right)$$

- Where c is heat capacity, k is conductivity, r is a heat source, and g is electron-phonon exchange

Phenomenology

- **The spatially uniform solution gives a time scale**

$$\tau_{ep} = \frac{c_e c_p}{g\left(c_e + c_p\right)} \qquad \theta_e - \theta_p = \left(\theta_e - \theta_p\right)_{t=0} \exp\left(-\frac{t}{\tau_{ep}}\right)$$

- **The steady-state solution gives a length scale**

$$\lambda_{ep} = \sqrt{\frac{k_e k_p}{g\left(k_e + k_p\right)}} \qquad \theta_e - \theta_p = \left(\theta_e - \theta_p\right)_{x=0} \exp\left(-\frac{x}{\lambda_{ep}}\right)$$

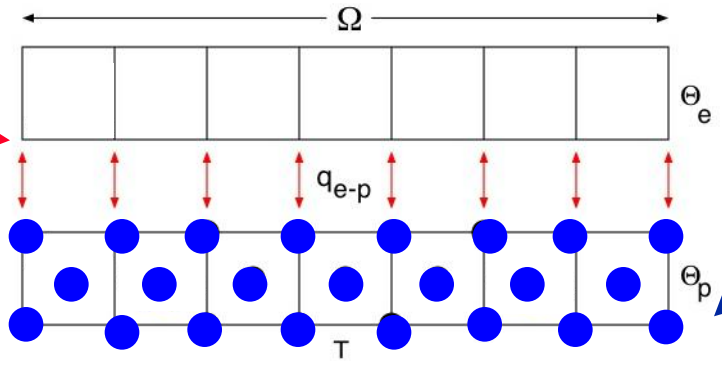
Material	τ_{ep}	λ_{ep}
Cu	0.36 ps	6.41 nm
Si	5.8 ps	494 nm

Below these time and length scales, it is not safe to assume that phonons and electrons are in equilibrium!

Our Modeling Approach: MD for Phonons, PDE for Electrons

PDE Model of Electrons

(Electrical and Thermal Conduction, Thermoelectric Coupling, Joule Heating)

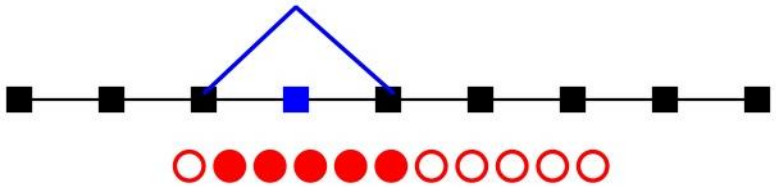


MD Model of Phonons

(Phonon Confinement, Ballistic Transport, Scattering Mechanisms)

- Use finite elements to solve PDE for electronic temperature θ_e
- Obtain local ionic temperature θ_p from MD velocities

$$\theta_p \equiv \sum_{I,\alpha} \frac{m_\alpha}{3k_B} \tilde{N}_{I\alpha} \langle v_\alpha \cdot v_\alpha \rangle$$



- Ionic thermostats enforce Two Temperature Model coupling

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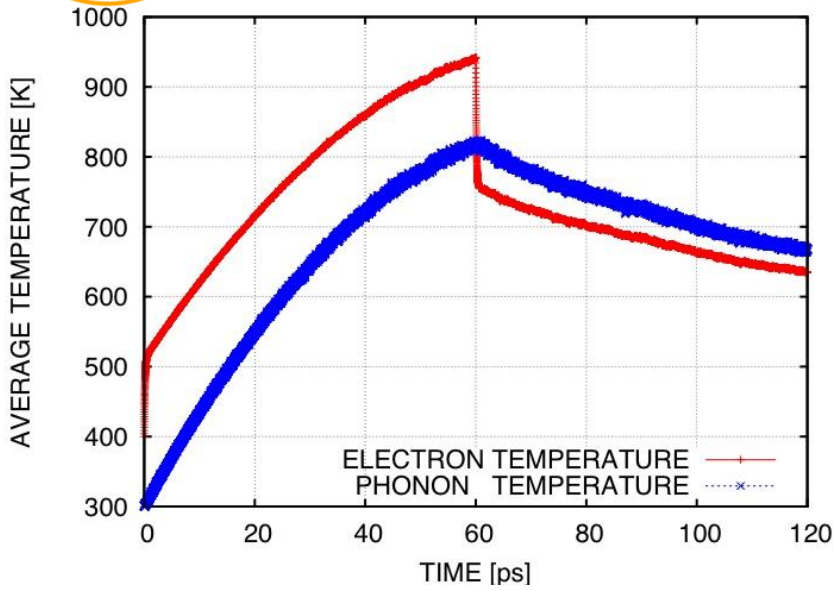
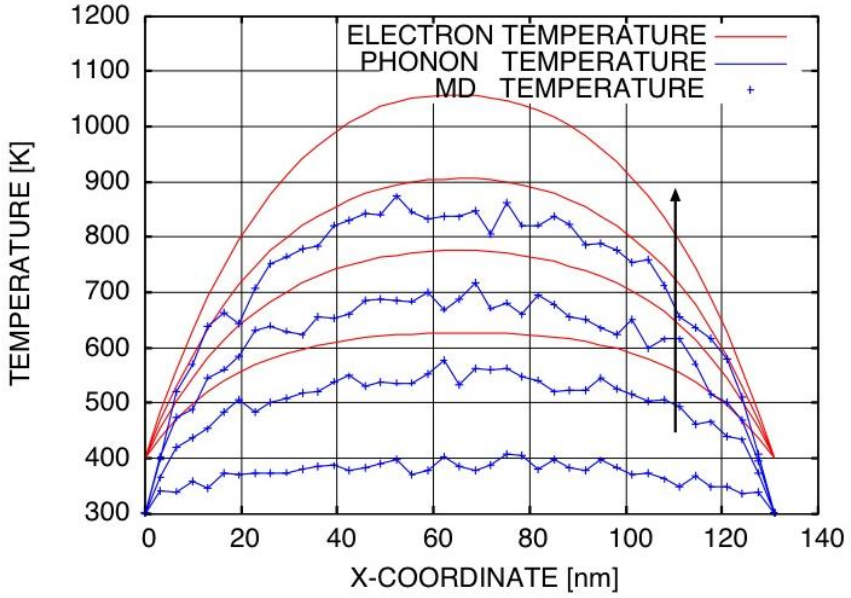
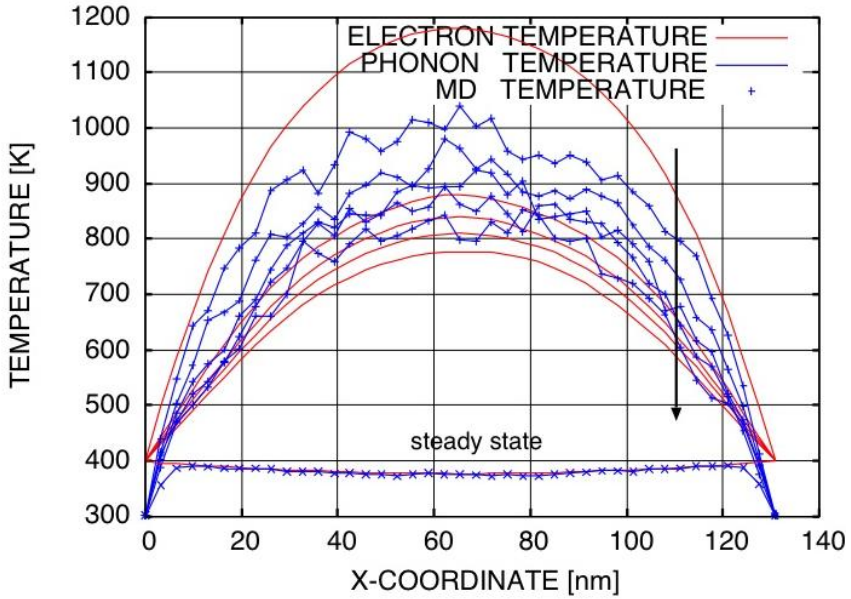
Example 1: Cu Nanowire with Electrons Uniformly Heated by 60 ps Pulse



- **Small electron heat capacity - Rapid electron response**
- **Large phonon heat capacity - Slower phonon response**
- **Different profiles for electron and phonon temperatures**

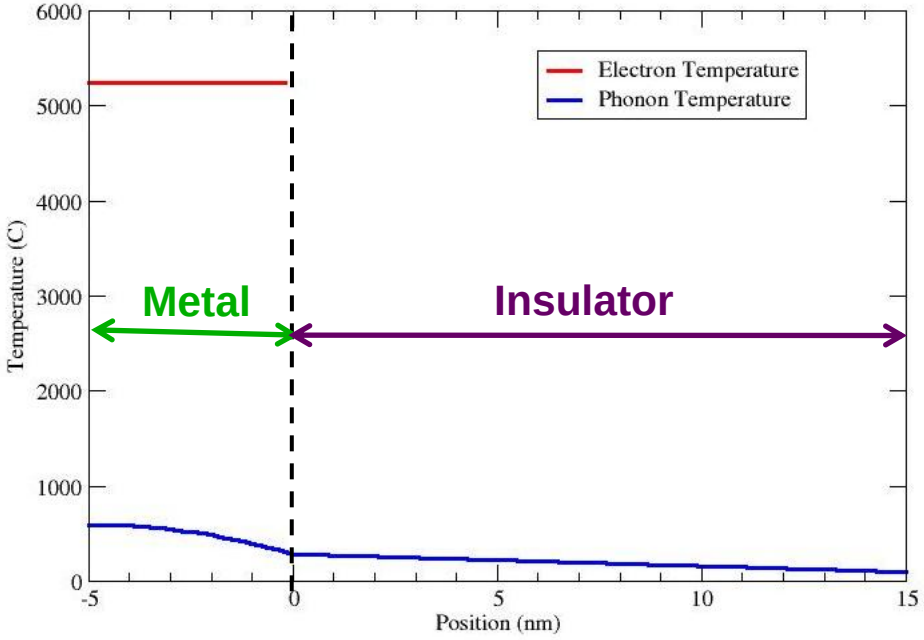




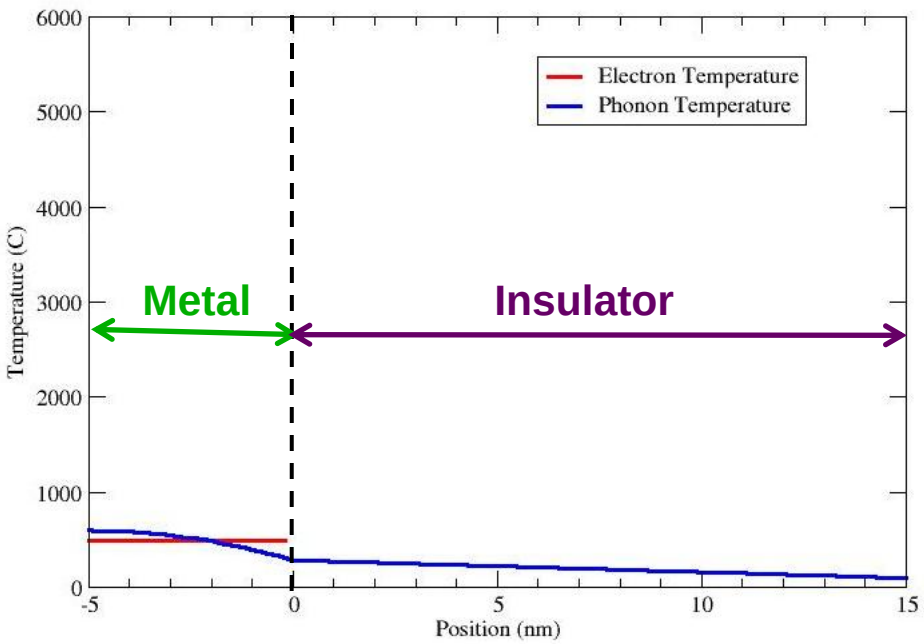
Large Electron-Phonon Temperature Differences in Steady-State Systems

Example 2: A Thin Metal Film on an Insulator

Uniform Heating in Electrons of Metal



Uniform Heating in Phonons of Metal



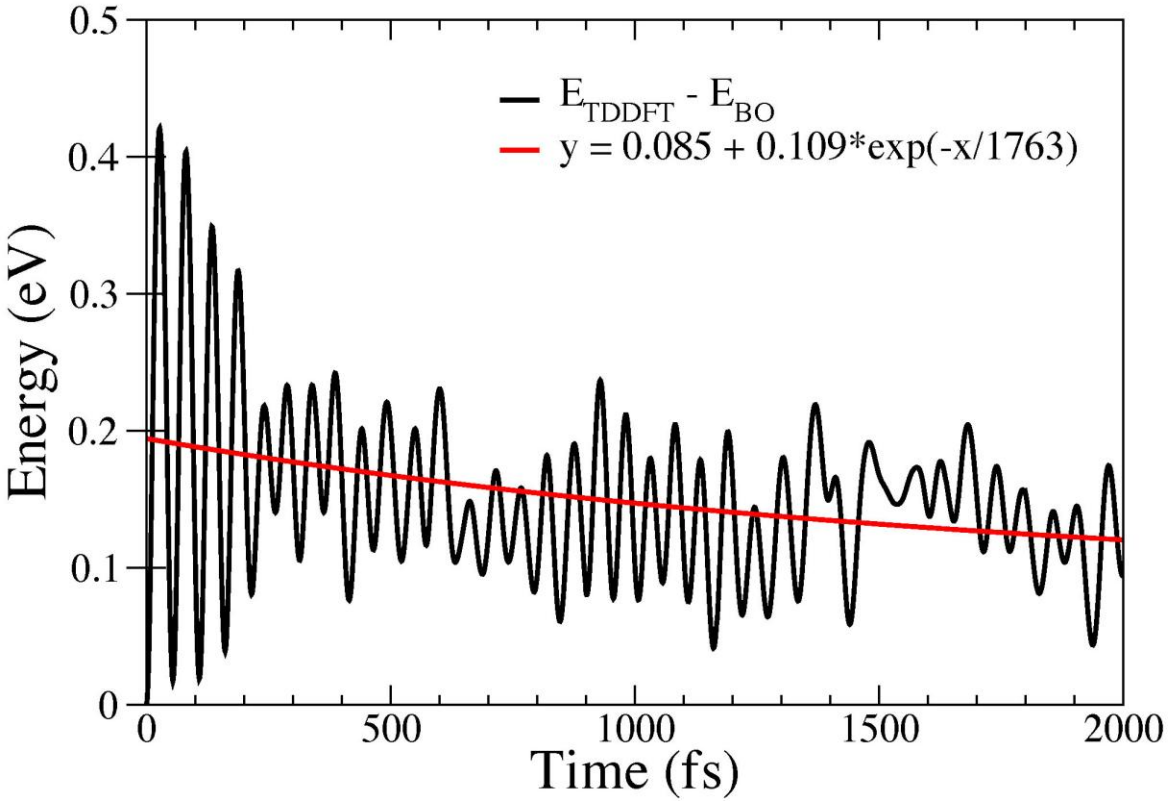
Finding Parameters from First Principles - Time Dependent Density Functional Theory

Unlike DFT, TDDFT allows electrons and ions to exchange energy!

- **Direct simulation of energy transfer between electrons and ions in Al gives time constant τ_{ep}= 1.8 ps**

$$\tau_{ep} = \frac{c_e c_p}{g\left(c_e + c_p\right)}$$

- **Good agreement with 1.5-2.0 ps equilibration time from experiment (Kandyla, Shih, and Mazur, 2007)**



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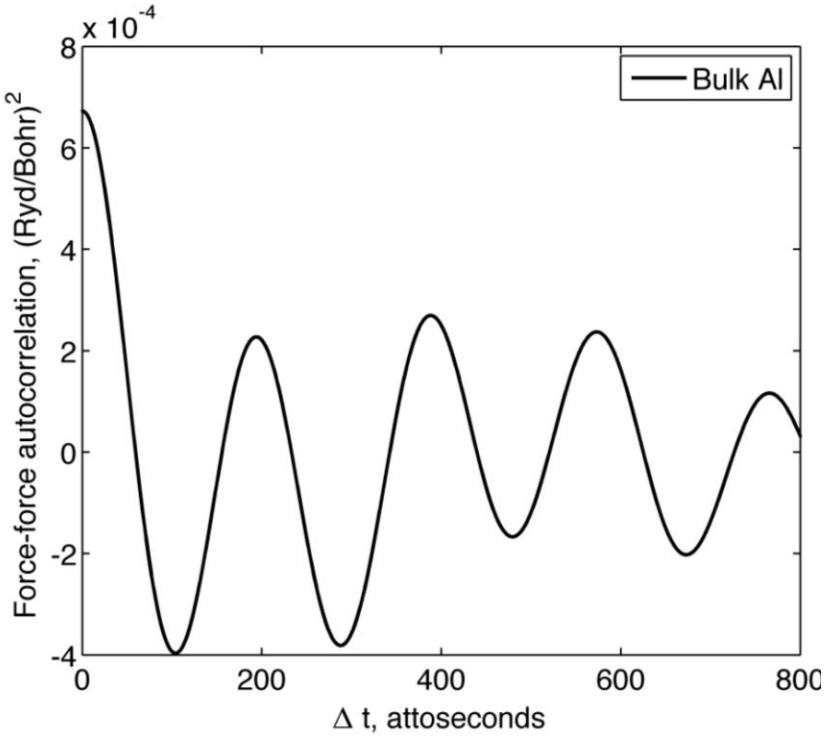
Working on TDDFT-Based Green-Kubo Approach to Improve Efficiency

- Explicit energy flow is computationally expensive
- Green-Kubo approach gives same quantities from steady state fluctuations
- Electron-Phonon coupling obtained from fluctuations in the ionic force
- Can calculate a wide variety of quantities with same approach



Problem: Autocorrelation functions oscillate strongly!

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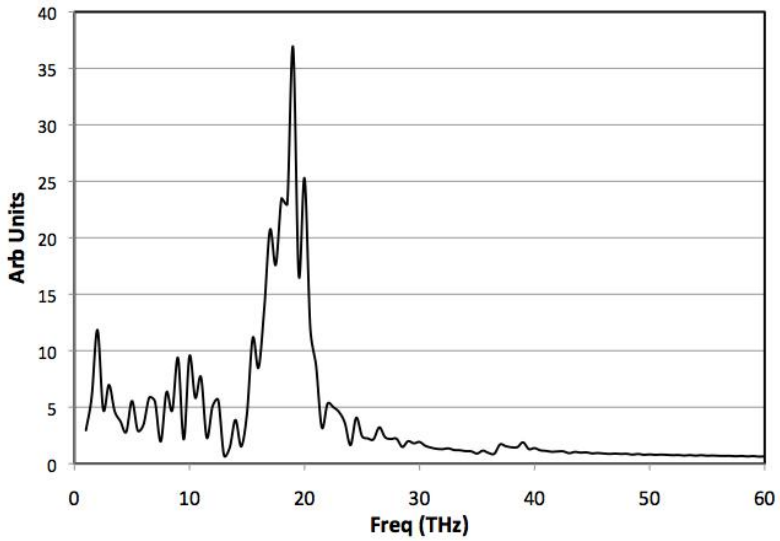
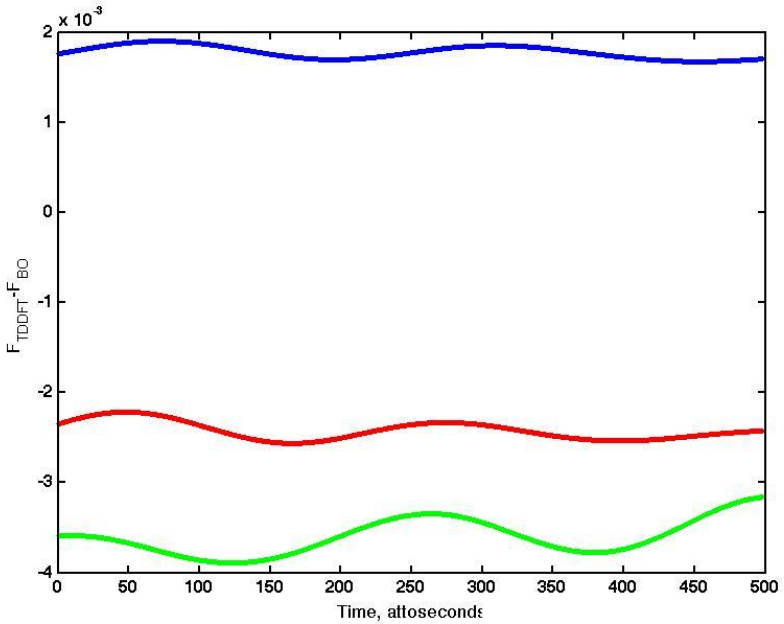
What are the fluctuations in TDDFT?

Low energies: Phonons



High energies: Plasmons

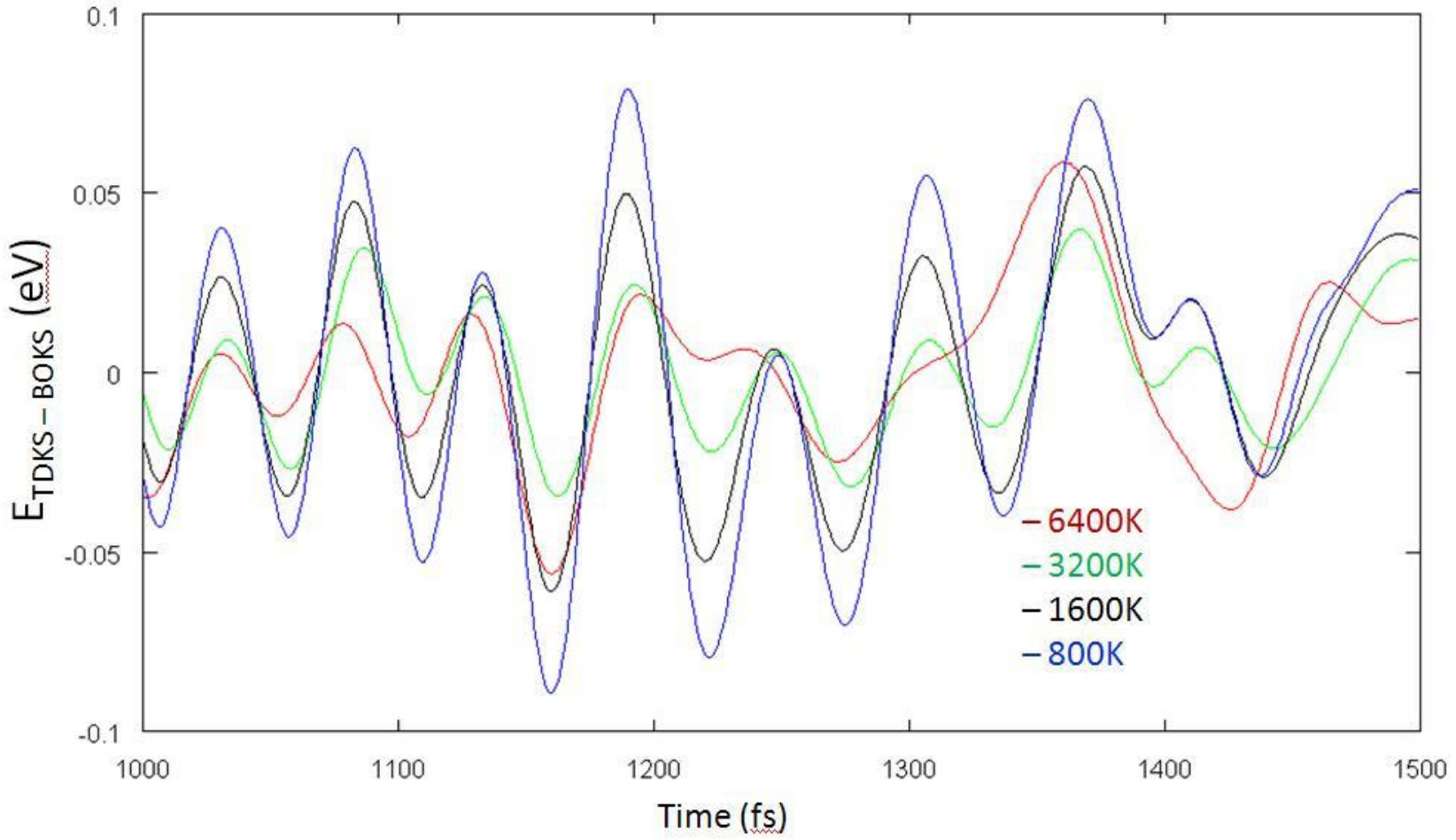


Working to separate electron-hole fluctuations

Conclusions

- It is not safe to assume that the electronic and ionic temperatures are the same at the nanoscale
- Enhanced a MD code with a PDE-based representation of electronic heat conduction
- Developing a promising approach to calculating electronic properties using TDDFT
- Our approach is intrinsically multiphysics and multiscale

Why does E_{TDDFT} - E_{BO} fluctuate with the phonons?



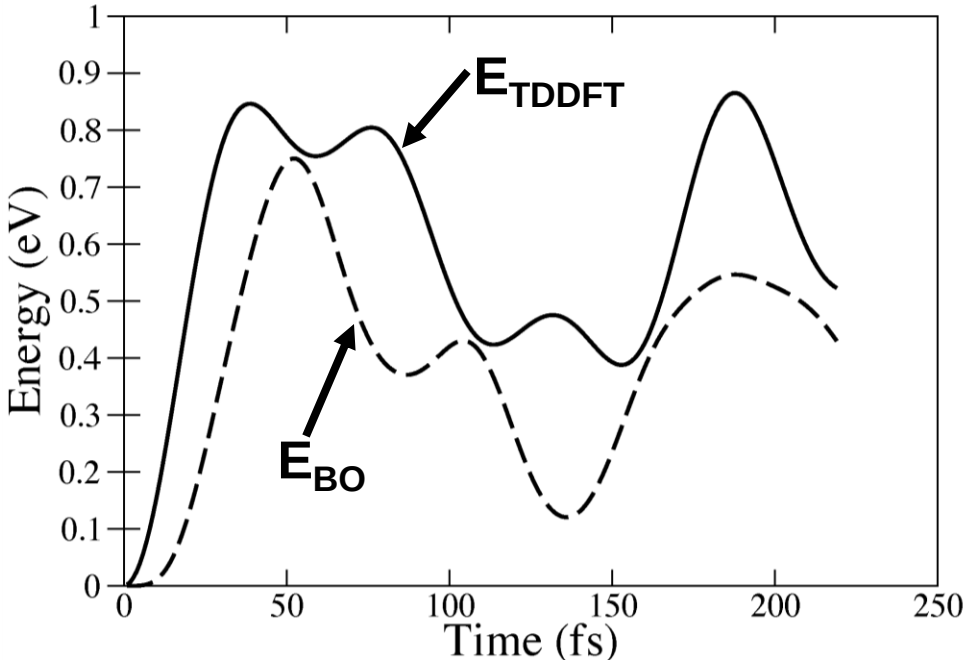
Electronic excitations change the potential surface for ionic motion

Finding Parameters from First Principles - Time Dependent Density Functional Theory

TDDFT allows electrons and ions to exchange energy!

- **Initially, ions in thermal motion and electrons in ground state**
- **TDDFT energy E_{TDDFT} rises above Born-Oppenheimer (ground state) energy E_{BO}**
- **$E_{TDDFT} - E_{BO}$ is instantaneous thermal energy of electrons**

TDDFT Run for 32 Atoms of Al



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Current TDDFT Work

- Understand, model, and remove phonon effects
- OR work with non-moving ions (but what about equilibration?)
- Develop better initialization to start closer to equilibrium
  - Eliminate plasmons and accentuate electron-hole pairs
- Can we get hot ions and cold electrons?

Issues in Modeling Heat Transport at the Nanoscale

- Time-dependent electronic structure (e.g., TDDFT)
  - Captures full electron and ion dynamics, BUT
  - Not feasible for most nanoscale systems
- Partial Differential Equation (PDE) based methods
  - Works well at macroscale, BUT
  - Misses effects of nanoscale structure on phonons (e.g., phonon confinement, ballistic transport, etc.)
- Molecular Dynamics (MD)
  - Explicitly represents phonons and their effects, BUT
  - Physics of electronic transport is absent