Atomistic simulation of phonon heat transport across metallic vacuum nanogaps

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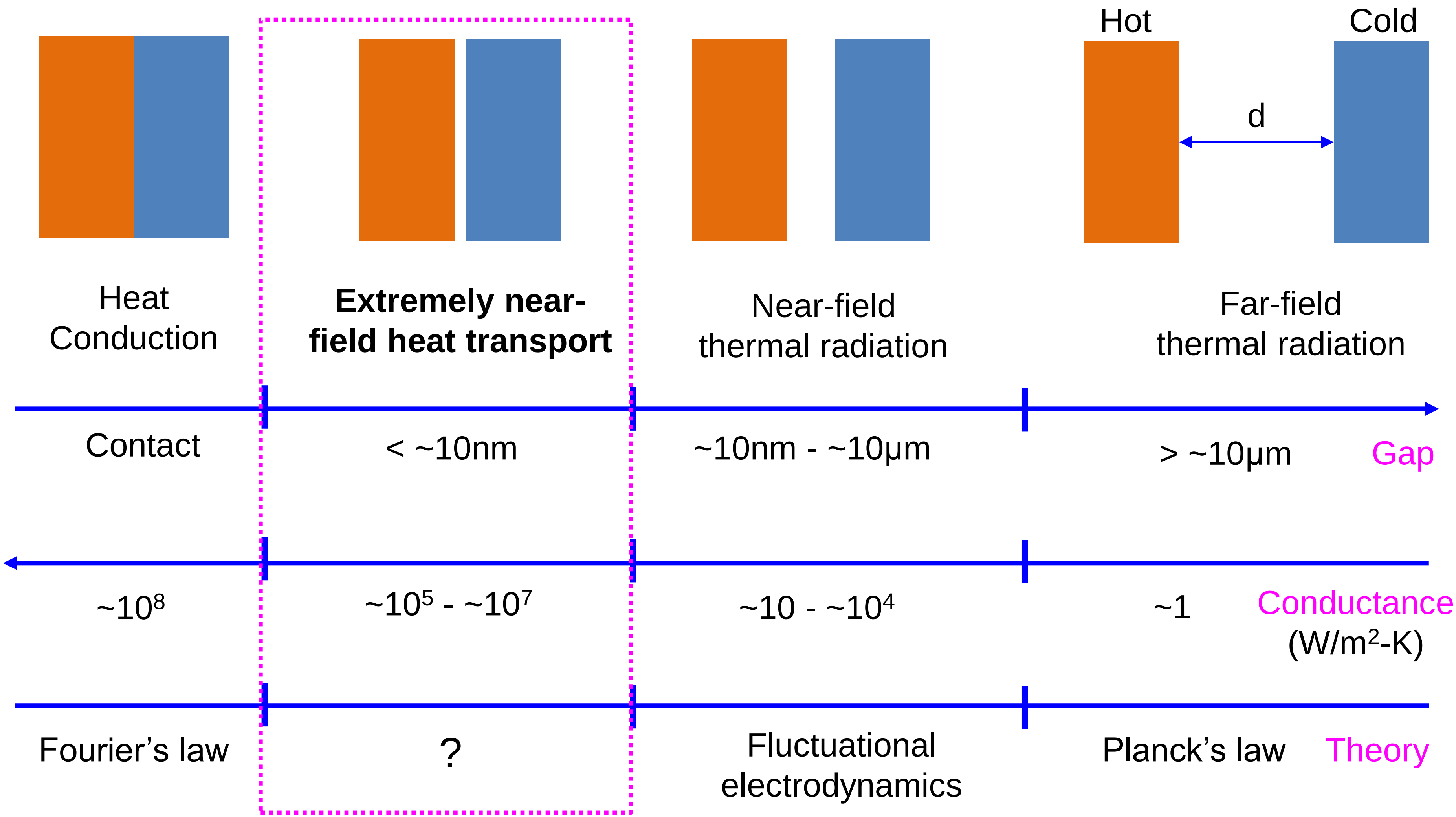
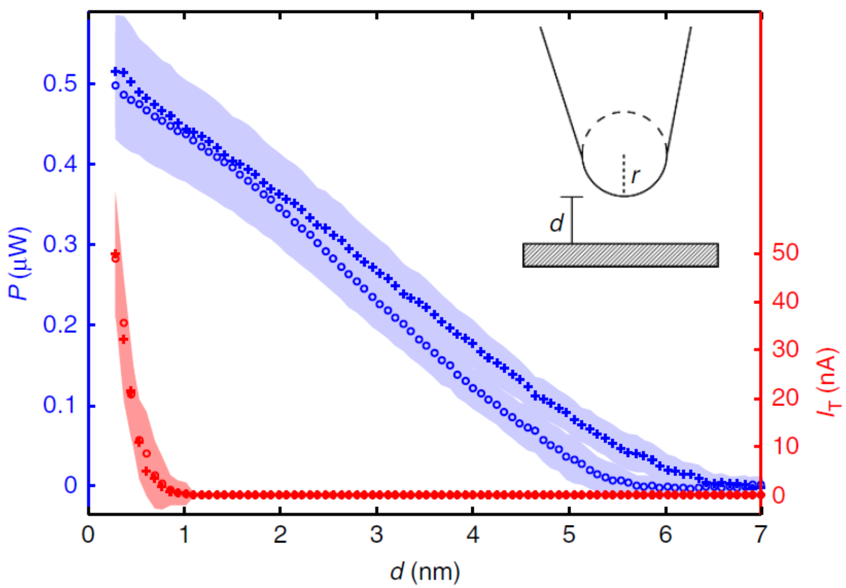
Introduction



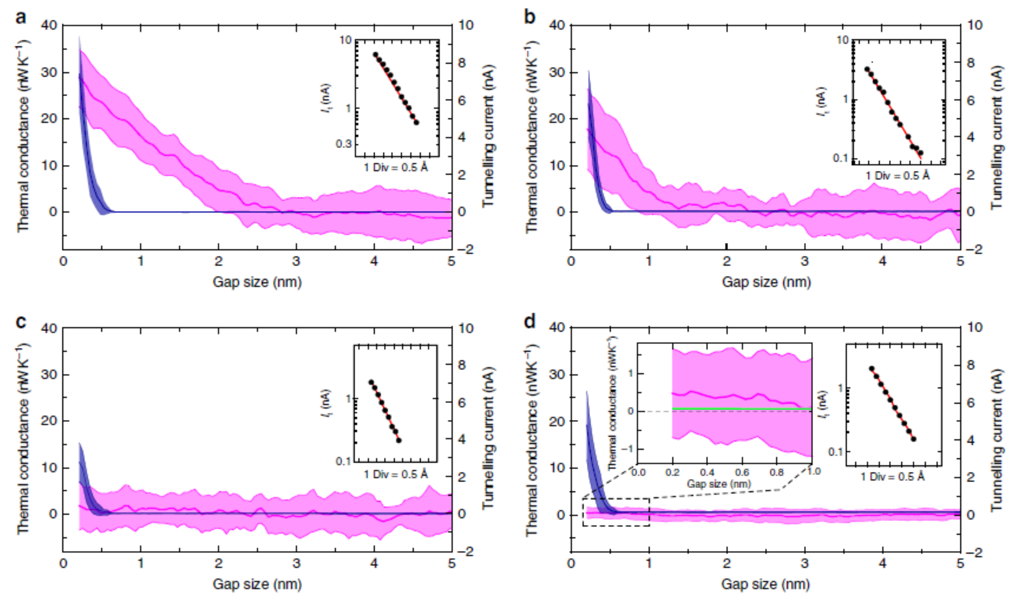
Different regimes of heat transport across metallic nanogaps

Introduction

➢ Extremely near-field experiments: controversies



Kloppstech *et al*. Nat. Commun. 8, 14475(2017)



Cui *et al*. Nat. Commun. 8, 14479(2017)

Conductance ~ 10^6 W/m^2-K

- Contamination-dependent conductance
- The conductance of cleanest sample < the detection resolution !

The understanding of extremely near-field heat transport remains an open question !

Introduction

- Extremely near-field phonon heat transport modeling

Phonon tunneling: one possible mechanism for the high conductance in extremely near-field regime.

- **Harmonic approach**

Landauer's formula + transmission:

Phonon NEGF

Xiong *et al*. PRL, 2014; Tokunaga *et al*. PRB, 2021, 2022

- Usually 1D NEGF
- Usually gap not relaxed

(except: Tokunaga *et al*. PRB, 2022)

Lattice dynamics

Sellan *et al*. PRB, 2012; Alkurdi *et al*. IJHMT, 2020

- Gap not relaxed

Elastic continuum model

Prunnila & Meltaus PRL, 2010; Pendry *et al*. PRB, 2016

➢ Extremely near-field phonon heat transport modeling

Phonon tunneling: one possible mechanism for the high conductance in extremely near-field regime.

- **Harmonic approach**

Fluctuational-electrodynamic model: Volokitin. JETP Lett., 2019; JPCM, 2020

- Acoustic waves (elastic continuum limit)
- Strong effect from electrostatics

- **Anharmonic approach**

Molecular dynamics (***MD***): Chen & Nagayama. IJHMT, 2021

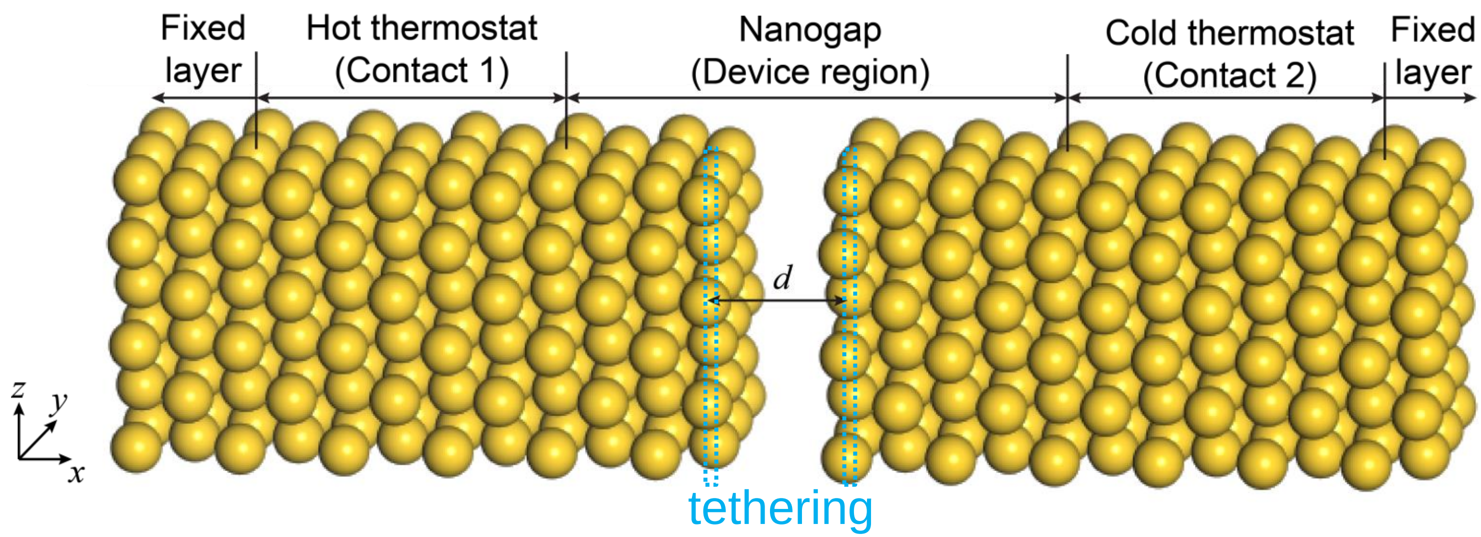
- Stability of the gap system: unknown
- The effect of anharmonicity: unclear

Introduction

- Aim of this work

  - 3D Atomistic modeling by combing MD & NEGF
  - Try to explain the experimental controversies
  - The role of anharmonicity & electrostatics

Physical model and Methodology

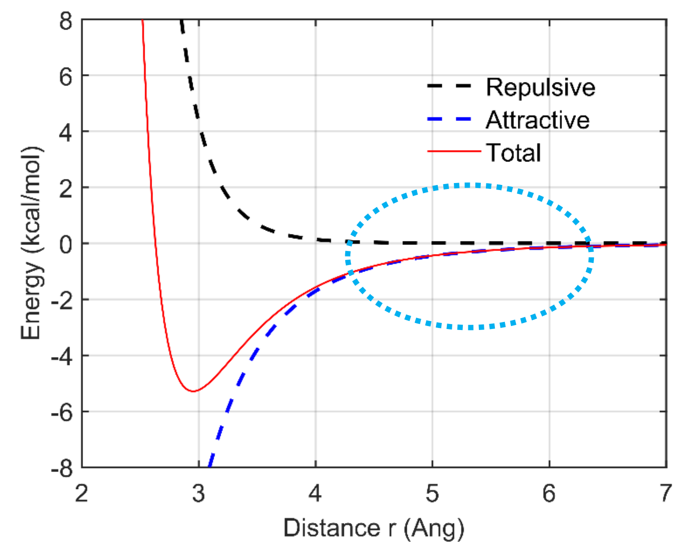


Au-Au (Cu-Cu) nanogap

NEMD

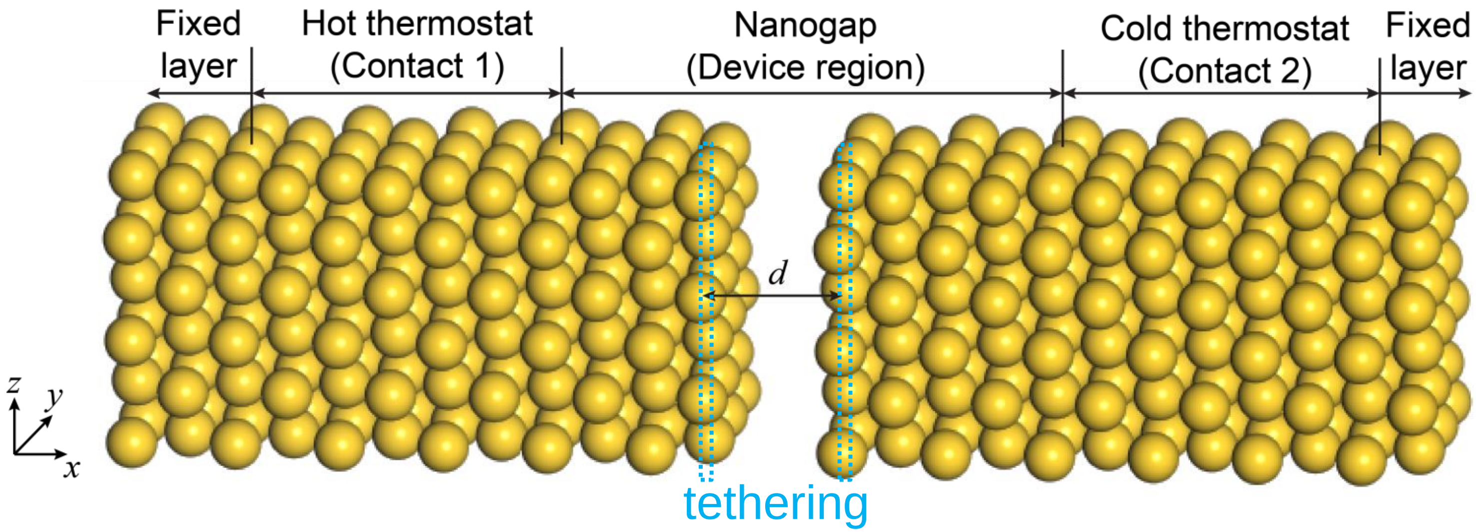
Relaxed structure & Force consts

Harmonic NEGF



Heinz *et al.* JPCC, 2008, L-J potential

Physical model and Methodology



Au-Au (Cu-Cu) nanogap

NEMD

Relaxed structure & Force consts

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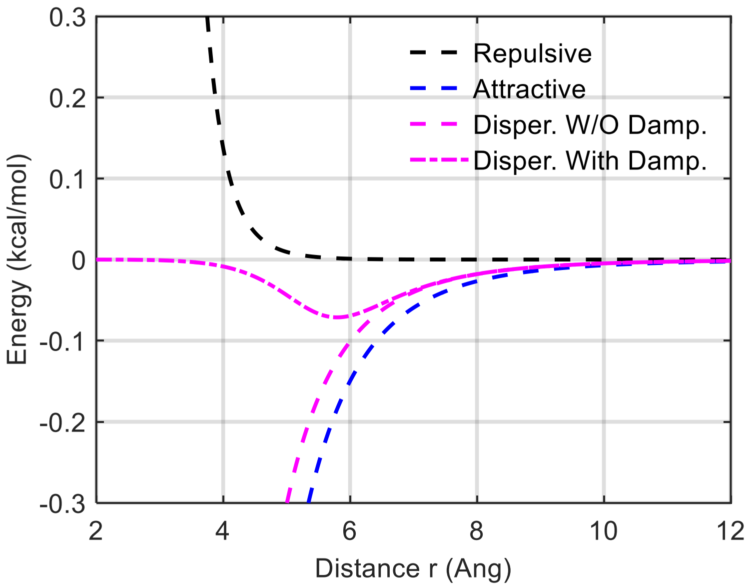
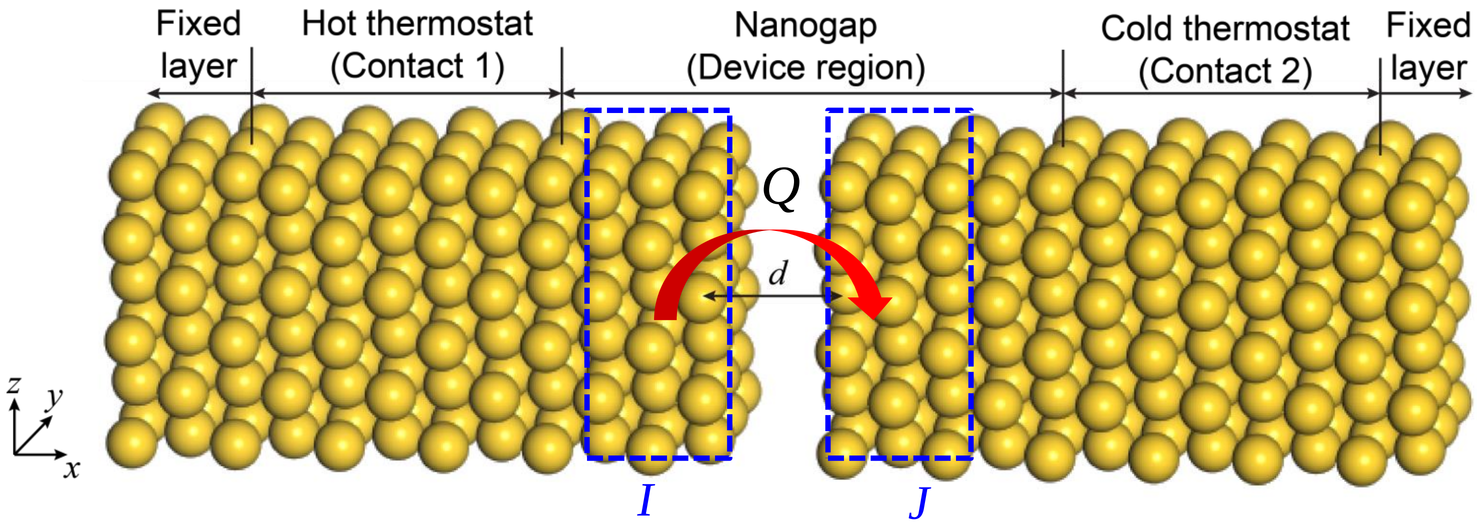


Physical model and Methodology



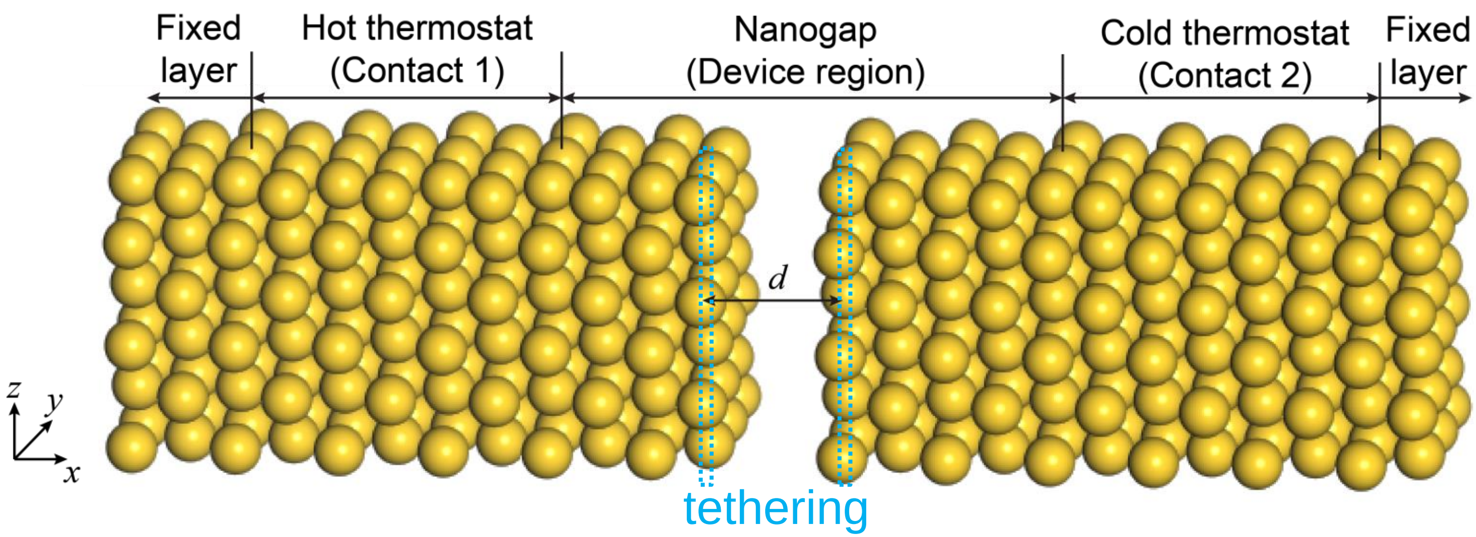
Spectral heat current decomposition in NEMD:

$$Q = \int_0^\infty q(\omega)\frac{d\omega}{2\pi} = \sum_{\substack{i\in I \\ j\in J}} \int_0^\infty q_{i\to j}(\omega)\frac{d\omega}{2\pi} \qquad q_{i\to j}(\omega) = 2\,\mathrm{Re}\left[\tilde{K}_{ji}(\omega)\right]$$

$$\tilde{K}_{ji}(\omega) = \int_{-\infty}^{\infty} K_{ji}(\tau)\exp(i\omega\tau)d\tau \qquad K_{ji}(t_1 - t_2) = \frac{1}{2}\left\langle \mathbf{F}_{ji}(t_1)\cdot\left[\mathbf{v}_j(t_2) + \mathbf{v}_i(t_2)\right]\right\rangle$$

K. Sääskilahti *et al*. PRB 90, 134312 (2014)
K. Sääskilahti *et al*. PRB 91, 115426 (2015)

Physical model and Methodology



1) Retarded Green's function: $$\mathbf{G}^{\mathrm{R}}(\omega;\mathbf{q}_\perp)=\left[\omega^2\mathbf{I}-\tilde{\mathbf{\Phi}}(\mathbf{q}_\perp)-\mathbf{\Sigma}^{\mathrm{R}}(\omega;\mathbf{q}_\perp)\right]^{-1}$$

Dynamic matrix:

$$\tilde{\Phi}^{ij}_{l_x l'_x}(\mathbf{q}_\perp)=\sum_{\Delta\mathbf{R}_\perp}\Phi^{ij}_{ll'}\exp\left(-\mathbf{q}_\perp\cdot\Delta\mathbf{R}_\perp\right)$$

$$\Phi^{ij}_{ll'}=\frac{1}{\sqrt{m_l m_{l'}}}\frac{\partial^2\Phi}{\partial r_l^i\partial r_{l'}^j}$$

transverse wave vector (100 × 100)

2^{nd}-order force constant

2) Transmission: $$\Xi(\omega)=\frac{1}{N}\sum_{\mathbf{q}_\perp}\mathrm{Tr}\left[\mathbf{\Gamma}_1(\omega;\mathbf{q}_\perp)\mathbf{G}^{\mathrm{R}}(\omega;\mathbf{q}_\perp)\mathbf{\Gamma}_2(\omega;\mathbf{q}_\perp)\mathbf{G}^{\mathrm{A}}(\omega;\mathbf{q}_\perp)\right]$$

3) Thermal conductance (classical limit): $$h=\frac{1}{A_c}\int_0^\infty k_B\Xi(\omega)\frac{d\omega}{2\pi}$$

Results and Discussions

➢ Thermal conductance of Au-Au nanogap at 300 K

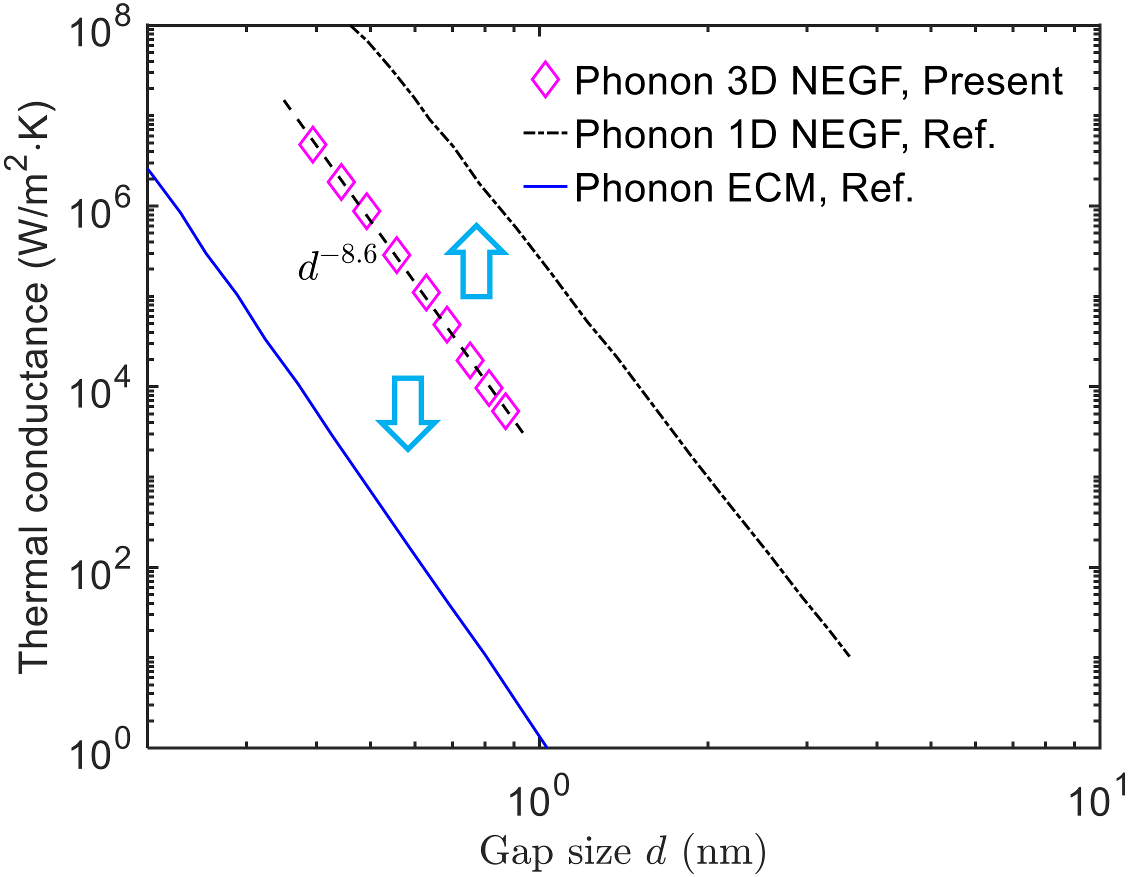


Plate-plate config.

The 1D NEGF / ECM overestimates / underestimates a lot the phonon thermal conductance !

1) 1D NEGF: Tokunaga *et al*. PRB, 2021
2) ECM: Volokitin. JETP Lett., 2019

ECM: elastic continuum model

Results and Discussions

- Thermal conductance of Au-Au nanogap at 300 K

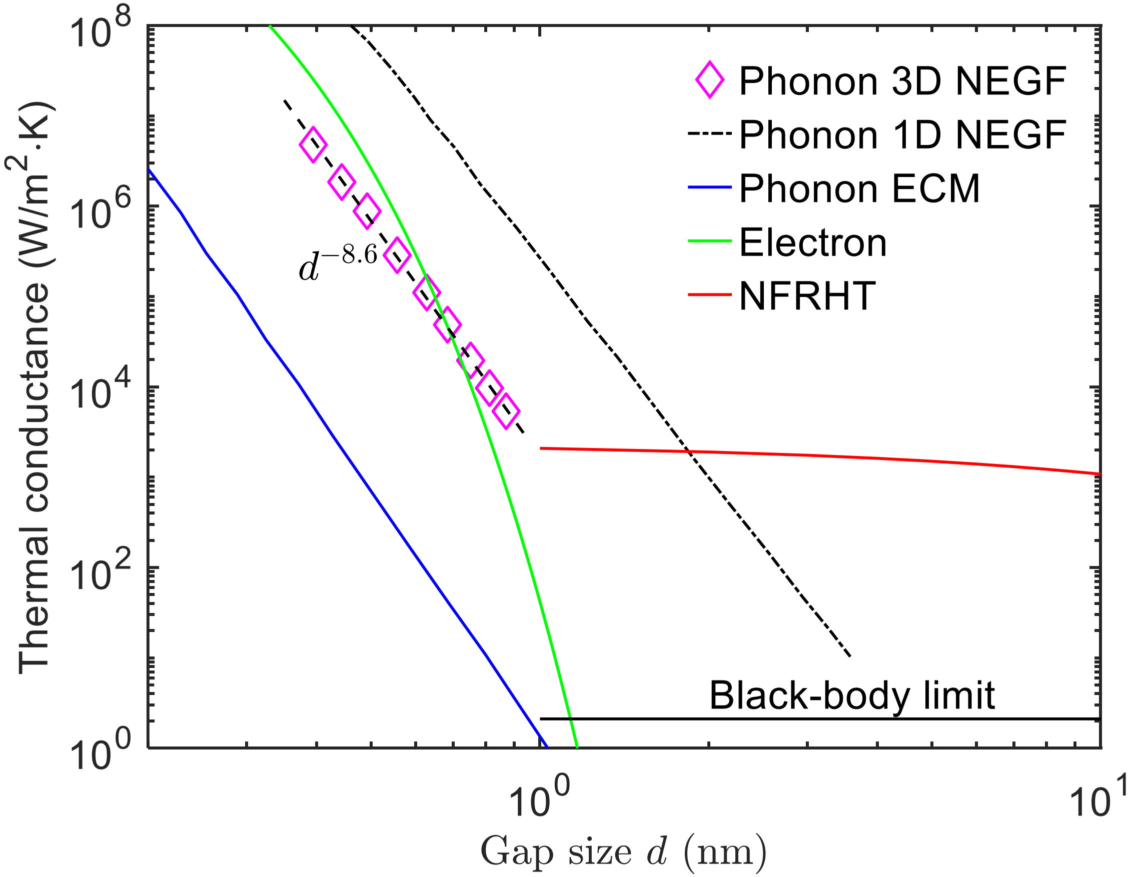
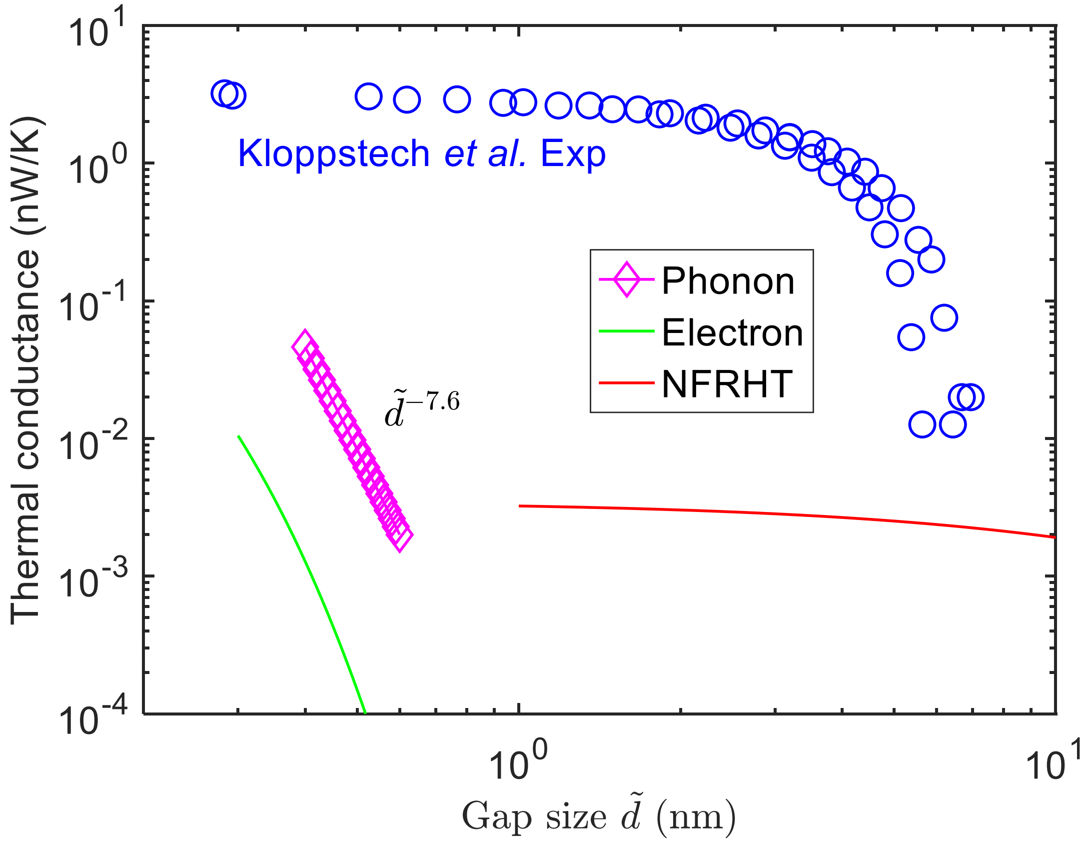


Plate-plate config.

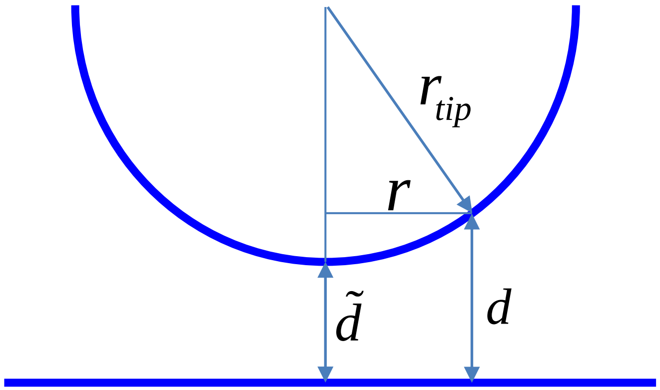
Phonon channel is significant below ~1nm

Results and Discussions

- Thermal conductance of Au-Au nanogap at 300 K



Tip-plate config.



Derjaguin approximation:

$$h(\tilde{d}) = \int_0^{r_{tip}} G(d) 2\pi r dr$$

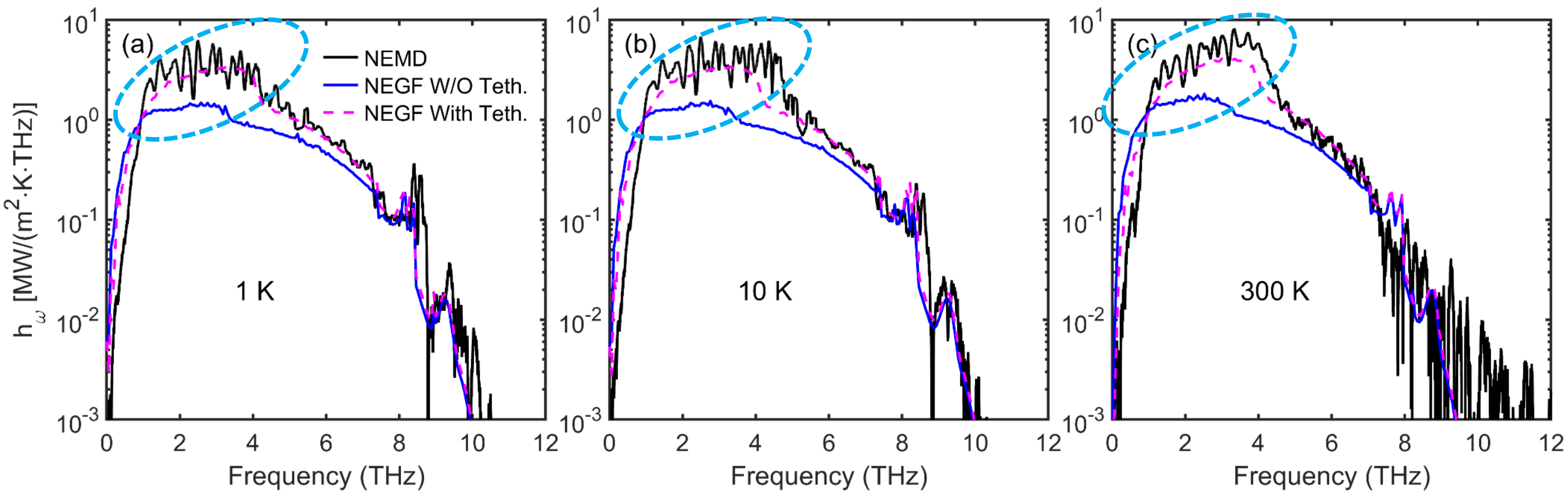
$$d = \tilde{d} + r_{tip} - \sqrt{r_{tip}^2 - r^2}$$

Our modeling result is much lower than Kloppstech *et al*. Exp., but consistent with Cui *et al.* Exp.

Results and Discussions

➢ The role of anharmonicity

Cu-Cu gap (d = 3.45 Å)



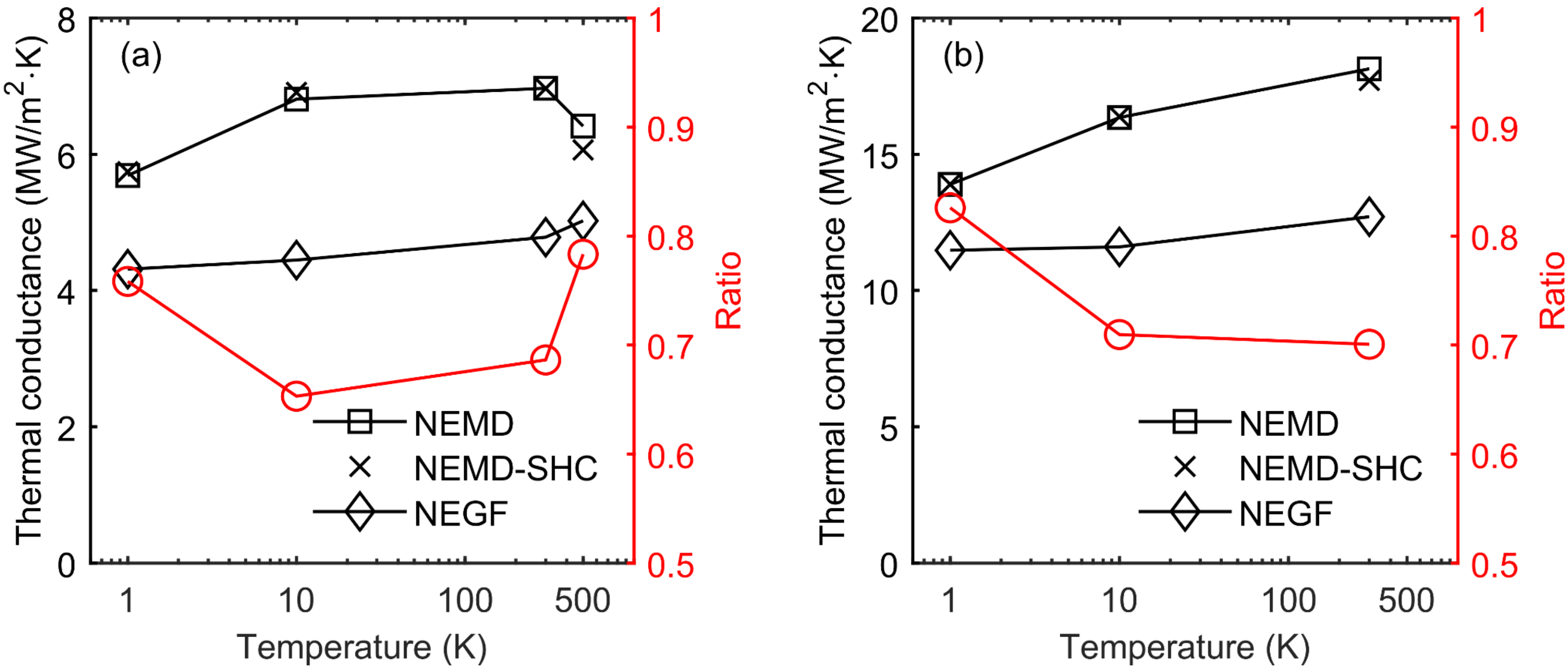
weak → strong

anharmonicity

The difference between NEMD and NEGF ↑

Results and Discussions

- The role of anharmonicity



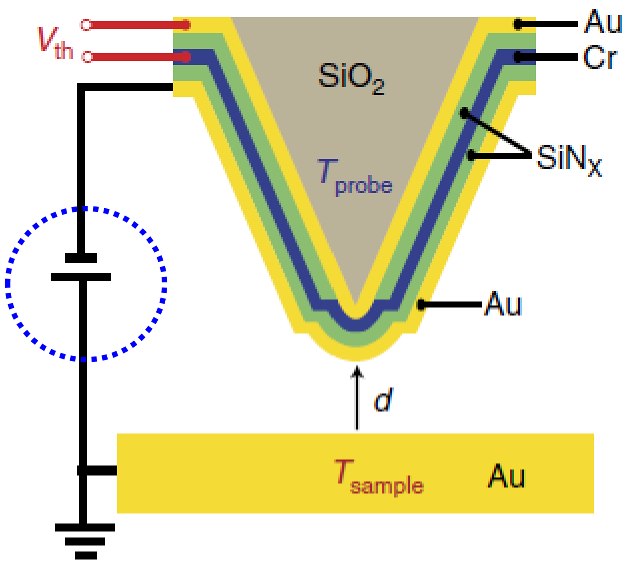
$\gamma_{G,\,Au} \approx 3$ Au-Au gap (d = 3.94 Å)

Cu-Cu gap (d = 3.45 Å) $\gamma_{G,\,Cu} \approx 2$

Anharmonicity contributes to around 20-30%.

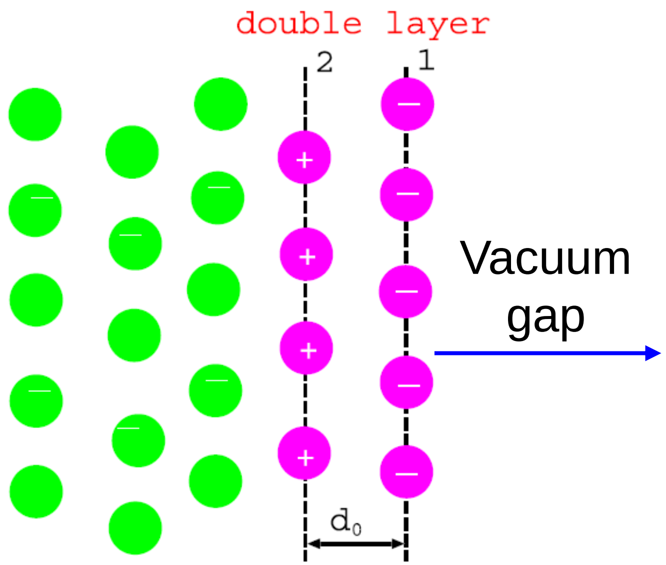
Results and Discussions

- The role of electrostatics



Bias voltage in Exp.

Cui *et al*. Nat. Commun. 8, 14479(2017)



EDL at the interface

Volokitin. PRB **103**, L041403(2021)

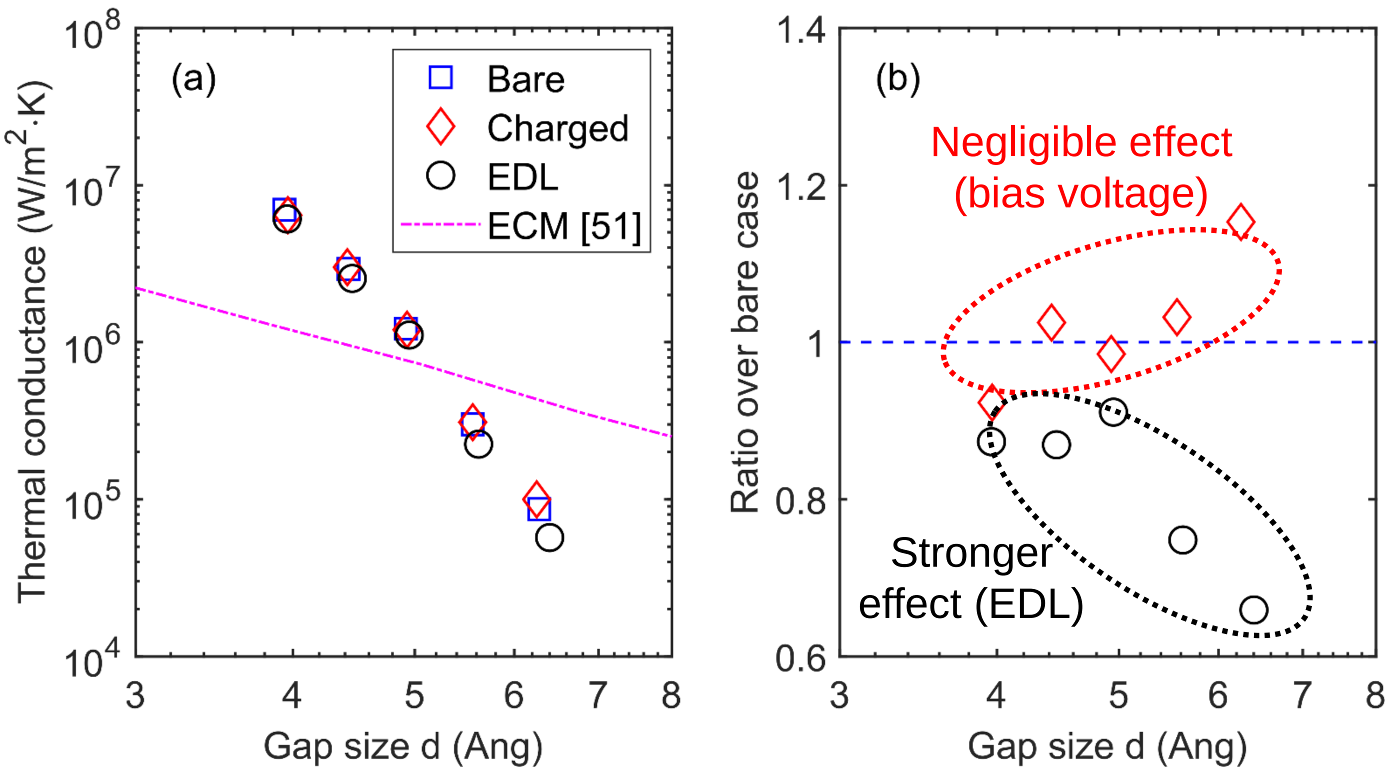
EDL: electrical double layer

Results and Discussions

- The role of electrostatics

Au-Au gap @ 300 K

Kloppstech *et al*. Exp., bias voltage = 600 mV



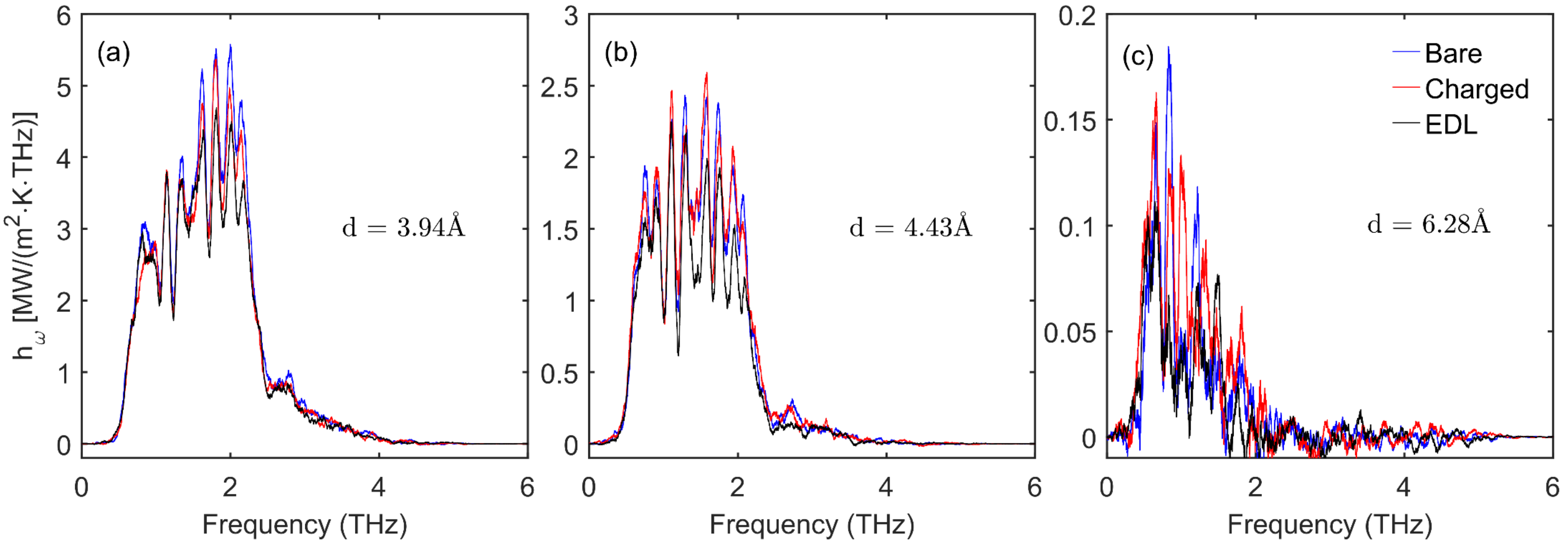
Thermal conductance by NEMD

EDL: electrical double layer

Results and Discussions

➢ The role of electrostatics

Au-Au gap @ 300 K



Spectral thermal conductance by NEMD

EDL affects the spectrum around1-2 THz.

Summary

- ✓ A 3D atomistic simulation framework for studying phonon tunneling across nanogap by combining NEMD and NEGF
- ✓ Our result is consistent with Cui *et al*. exp., while can not explain Kloppstech *et al.* exp. probably due to contaminations
- ✓ The anharmonicity contributes to around 20-30% in phonon heat transport across the nanogap.
- ✓ The bias voltage in experimental setup seems to play a negligible role in phonon heat transport.

Thank you for your attention ^_^

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