Thickness effect on basal-plane heat transport in graphite thin film

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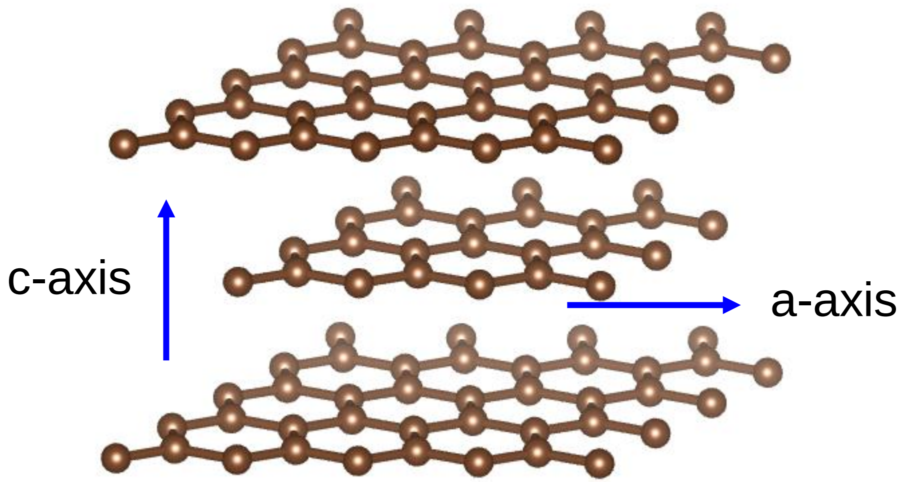
Introduction

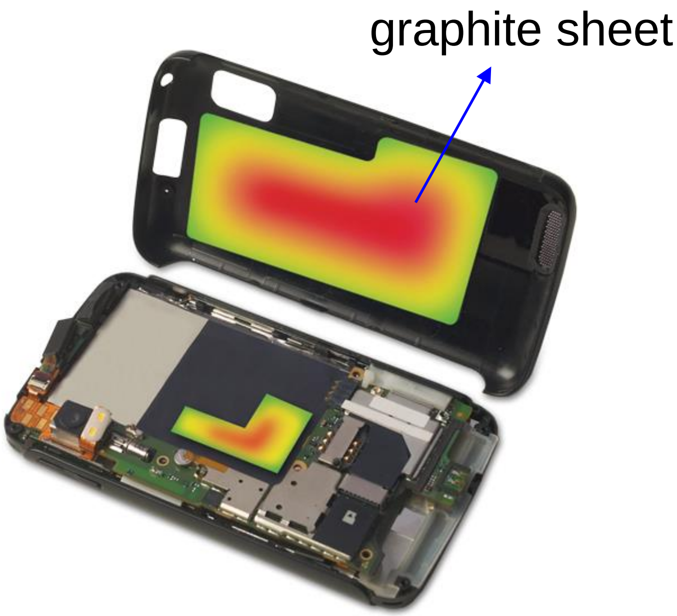


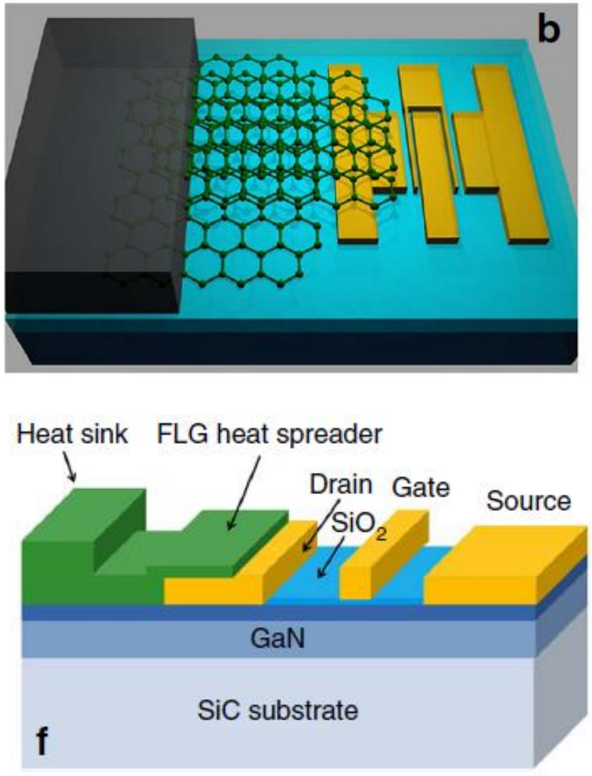
Graphite as a very anisotropic material

Bulk thermal conductivity at 300 K:
- a-axis (basal plane): ~2000 W/m.K
- c-axis: ~6 W/m.K

Introduction

- Graphite thin film as thermal management material

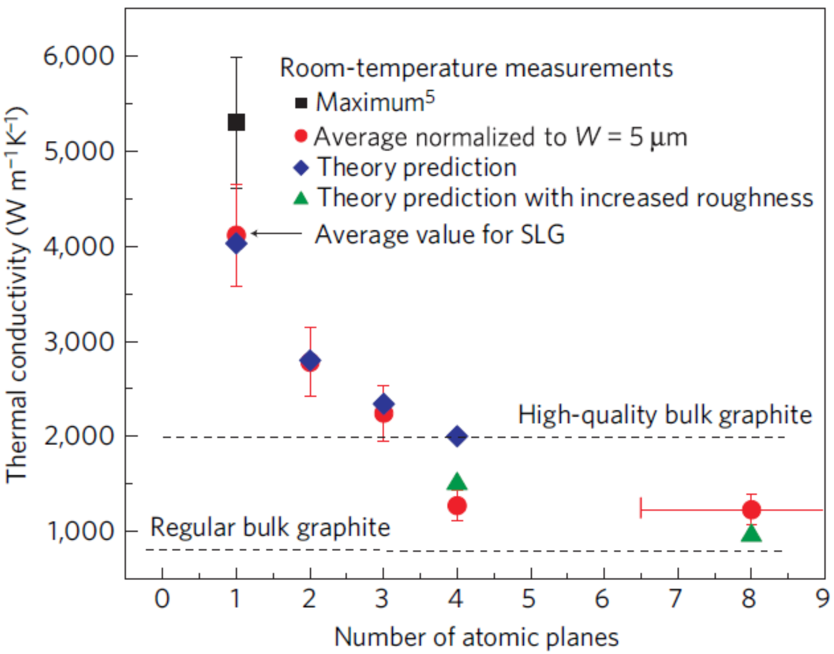




Yan *et al. Nat. Commun.* 3:827 (2012)

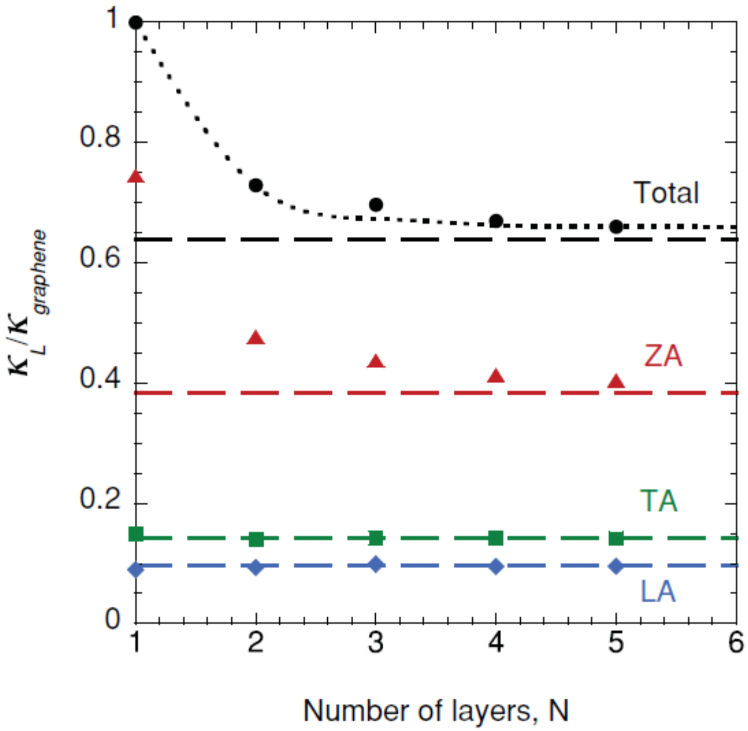
Introduction

- Thickness effect on basal plane heat transport:



Ghosh *et al*. Nat. Mater. 2010

Experimental



Lindsay *et al*. Phys. Rev. B 2011

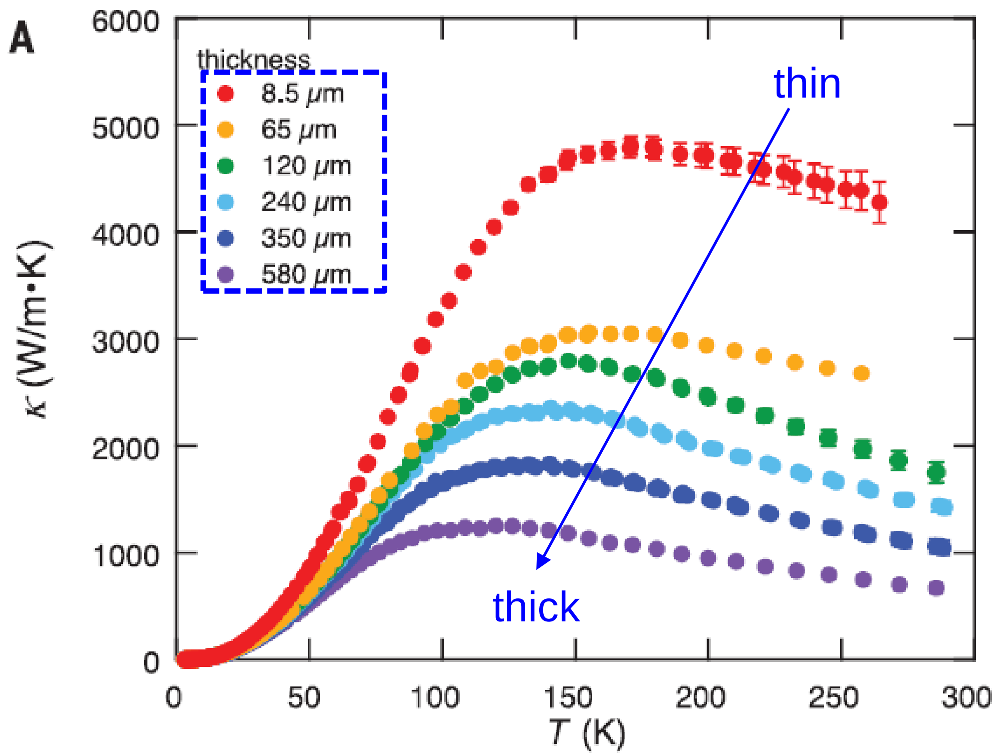
BTE solution with empirical potential

"Coherent regime"

Thermal conductivity converges after ~5 atomic planes.

Introduction

- Thickness effect on basal plane heat transport:



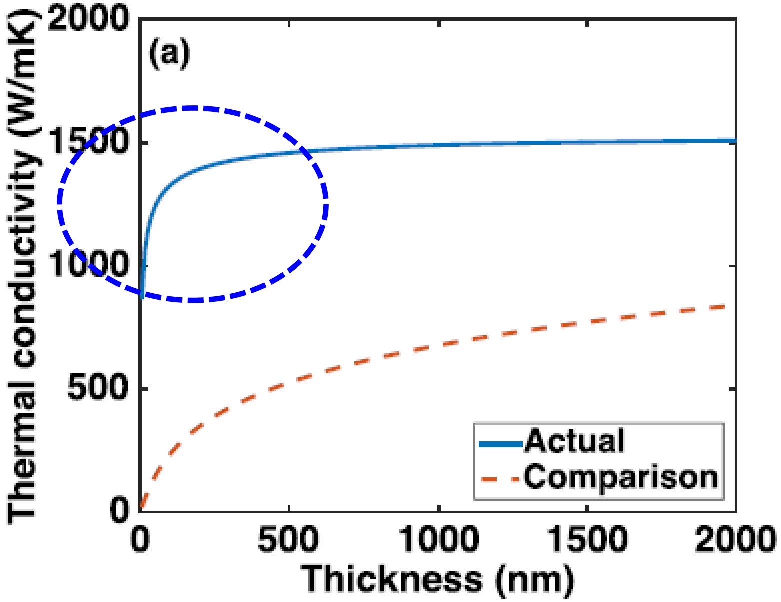
"Incoherent regime"

Machida *et al*. Science, 2020
Experimental

Anomalous thickness dependence of thermal conductivity !

Introduction

- Thickness effect on basal plane heat transport: theoretical modeling



Minnich, APL 2015

BTE-SMRT with empirical relaxation times

Actual: generalized F-S model

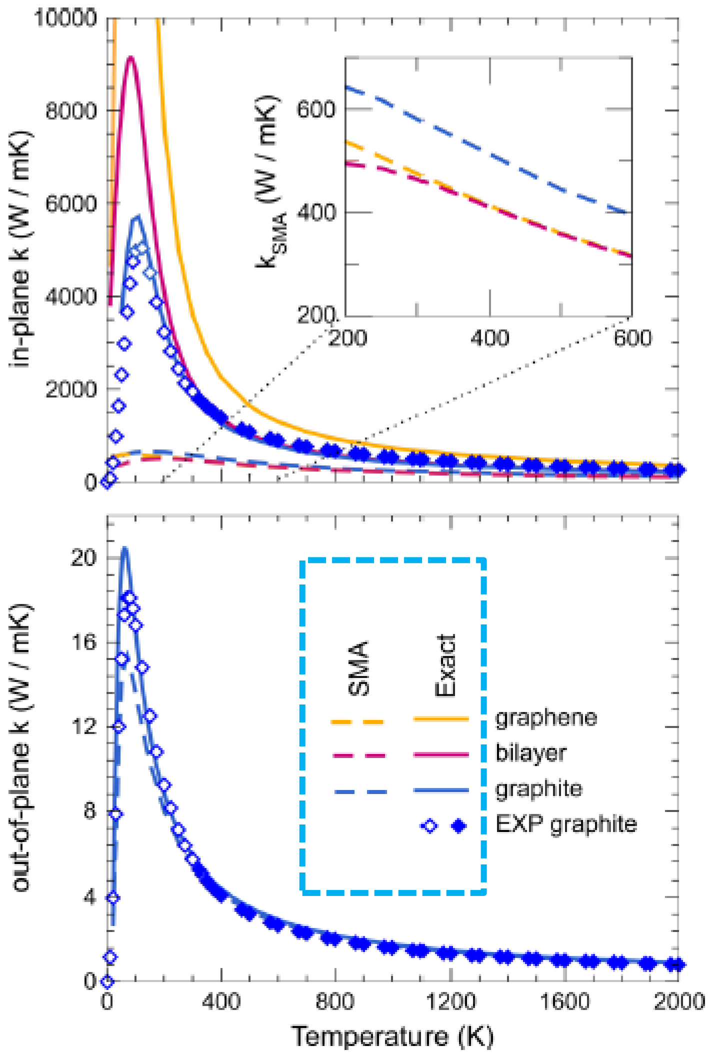
Comparison: $\Lambda_c = \Lambda_{ab}$ (isotropic)

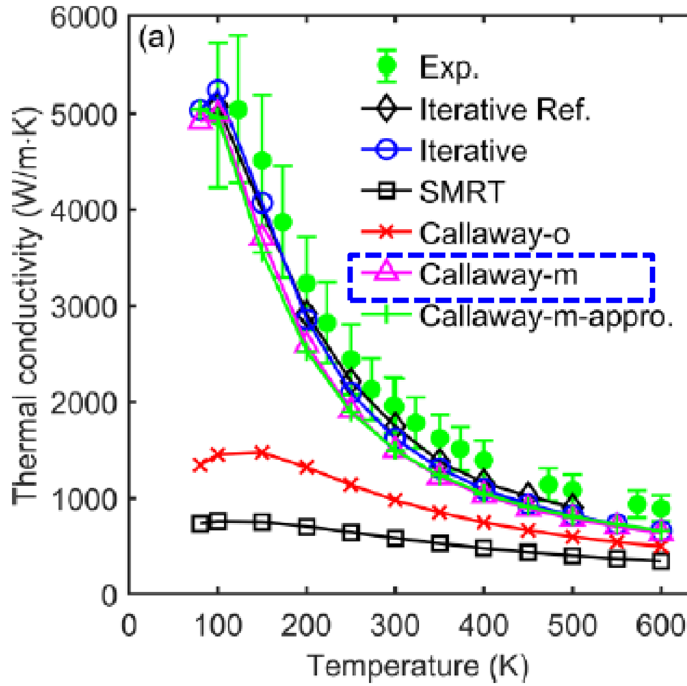
Weak thickness effect on basal plane heat transport due to small c-axis MFP.

- F-S model: Fuchs-Sondheim model
- SMRT: single mode relaxation time approximation

Introduction

- Analysis and motivation





Guo *et al.* Phys. Rev. B **104**, 075450 (2021)

- SMRT underestimates a lot the bulk thermal conductivity of graphite

- Callaway's dual relaxation model works well.

Fugallo *et al. Nano Lett.* 14, 6109−6114 (2014).

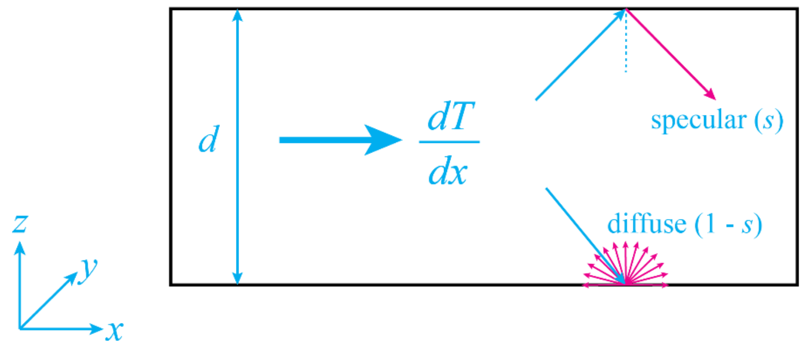
Introduction

- Aim of the present work

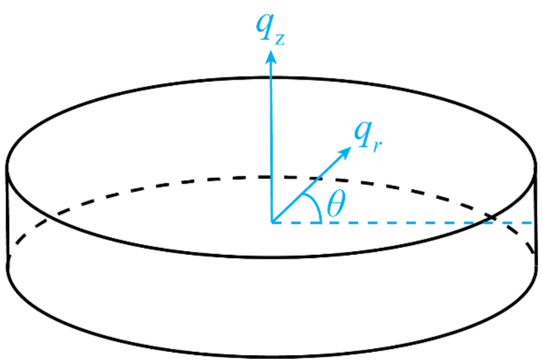
  ✓ Semi-analytical modeling of basal plane heat transport along graphite thin film with finite thickness by BTE-Callaway's model with *ab initio* input.

  ✓ Thickness and surface roughness effect on heat transport in graphite thin film.

Methodology



(a)



(b)

- Phonon BTE under Callaway's dual relaxation model

$$\frac{\partial f}{\partial t} + \mathbf{v}_{\mathrm{g}} \cdot \nabla f = \underbrace{\frac{f_{\mathrm{R}}^{\mathrm{eq}} - f}{\tau_{\mathrm{R}}}}_{\text{SMRT}} + \frac{f_{\mathrm{N}}^{\mathrm{eq}} - f}{\tau_{\mathrm{N}}}$$

$$f_{\mathrm{R}}^{\mathrm{eq}} = \frac{1}{\exp\left(\hbar\omega / k_{\mathrm{B}} T\right) - 1}$$

$$f_{\mathrm{N}}^{\mathrm{eq}} = \frac{1}{\exp\left[\left(\hbar\omega - \hbar\mathbf{q}\cdot\mathbf{u}\right) / k_{\mathrm{B}} T\right] - 1}$$

- Partially specular and partially diffuse B.C.:

$$\begin{cases} z = 0,\ f^{+}\left(0, \theta, q_r, q_z, p\right)\Big|_{v_{gz} > 0} = \left(1 - s\right) f_{\mathrm{R}}^{\mathrm{eq}} + s\, f^{-}\left(0, \theta, q_r, -q_z, p\right)\Big|_{v_{gz} < 0} \\ z = d,\ f^{-}\left(d, \theta, q_r, q_z, p\right)\Big|_{v_{gz} < 0} = \left(1 - s\right) f_{\mathrm{R}}^{\mathrm{eq}} + s\, f^{+}\left(d, \theta, q_r, -q_z, p\right)\Big|_{v_{gz} > 0} \end{cases}$$

Methodology

- The semi-analytical solution

$$
\begin{cases}
g^{+}(z,\theta,q_r,q_z,p)\big|_{v_{gz}>0} = -\tau_{\mathrm{C}} v_{gx}\dfrac{\partial T}{\partial x}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\left[1+\dfrac{(s-1)\exp\left(-z/v_{gz}\tau_{\mathrm{C}}\right)}{1-s\exp\left(-d/v_{gz}\tau_{\mathrm{C}}\right)}\right]+\dfrac{1}{v_{gz}}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\dfrac{Tq_x}{\omega\tau_{\mathrm{N}}}\displaystyle\int_0^z u_x(z')\exp\left(\dfrac{z'-z}{v_{gz}\tau_{\mathrm{C}}}\right)dz' \\
\qquad +\dfrac{1}{v_{gz}}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\dfrac{Tq_x}{\omega\tau_{\mathrm{N}}}\dfrac{s}{1-s^2\exp\left(-2d/v_{gz}\tau_{\mathrm{C}}\right)}\displaystyle\int_0^d u_x(z')\left[s\exp\left(\dfrac{z'-z-2d}{v_{gz}\tau_{\mathrm{C}}}\right)+\exp\left(-\dfrac{z'+z}{v_{gz}\tau_{\mathrm{C}}}\right)\right]dz', \\
g^{-}(z,\theta,q_r,q_z,p)\big|_{v_{gz}<0} = -\tau_{\mathrm{C}} v_{gx}\dfrac{\partial T}{\partial x}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\left[1+\dfrac{(s-1)\exp\left[(d-z)/v_{gz}\tau_{\mathrm{C}}\right]}{1-s\exp\left(d/v_{gz}\tau_{\mathrm{C}}\right)}\right]+\dfrac{1}{v_{gz}}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\dfrac{Tq_x}{\omega\tau_{\mathrm{N}}}\displaystyle\int_d^z u_x(z')\exp\left(\dfrac{z'-z}{v_{gz}\tau_{\mathrm{C}}}\right)dz' \\
\qquad -\dfrac{1}{v_{gz}}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}\dfrac{Tq_x}{\omega\tau_{\mathrm{N}}}\dfrac{s}{1-s^2\exp\left(2d/v_{gz}\tau_{\mathrm{C}}\right)}\displaystyle\int_0^d u_x(z')\left[s\exp\left(\dfrac{z'-z+2d}{v_{gz}\tau_{\mathrm{C}}}\right)+\exp\left(-\dfrac{z'+z-2d}{v_{gz}\tau_{\mathrm{C}}}\right)\right]dz', \\
g^{0}(z,\theta,q_r,q_z,p)\big|_{v_{gz}=0} = -\tau_{\mathrm{C}} v_{gx}\dfrac{\partial T}{\partial x}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}+\tau_{\mathrm{C}}\dfrac{T}{\tau_{\mathrm{N}}}\dfrac{q_x u_x(z)}{\omega}\dfrac{\partial f_{\mathrm{R}}^{\mathrm{eq}}}{\partial T}
\end{cases}
$$

Drift velocity

SMRT solution

$$\boxed{g = f - f_{\mathrm{R}}^{\mathrm{eq}}}$$

$$\frac{1}{\tau_{\mathrm{C}}}=\frac{1}{\tau_{\mathrm{N}}}+\frac{1}{\tau_{\mathrm{R}}}$$

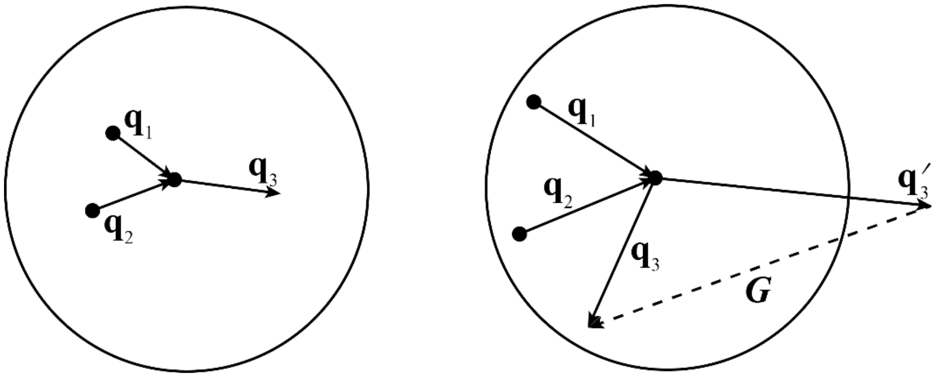
Methodology

- Determination of drift velocity: quasi-momentum conservation

$$\sum_p \int \hbar q_x \frac{f_{\mathrm{N}}^{\mathrm{eq}} - f}{\tau_{\mathrm{N}}} \frac{d\mathbf{q}}{(2\pi)^3} = 0$$

$$\Rightarrow \sum_p \int \hbar q_x \frac{g(z)}{\tau_{\mathrm{N}}} \frac{d\mathbf{q}}{(2\pi)^3} = \left(\mathbf{C}_{\tau_{\mathrm{N}}}^{1}\right)_{xx} T u_x(z)$$

$$\boxed{\mathbf{C}_{\tau_{\mathrm{N}}}^{1} = \sum_p \int \frac{C_{\mathbf{q}p}}{\omega^2} \frac{\mathbf{q}\mathbf{q}}{\tau_{\mathrm{N}}(\mathbf{q}, p, T)} \frac{d\mathbf{q}}{(2\pi)^3}}$$



normal scattering　　resistive scattering

Methodology

- Integral equation of the drift velocity

$$C_3 = \left(\mathbf{C}^1_{\tau_\mathrm{N}}\right)_{xx} - C_2 \qquad \bar{z} = \frac{z}{d},\ Kn_{\mathrm{C}z} = \frac{v_{\mathrm{g}z}\tau_\mathrm{C}}{d},\ \Lambda_{\mathrm{C}r} = v_{\mathrm{g}r}\tau_\mathrm{C},\ \Lambda_{\mathrm{N}z} = v_{\mathrm{g}z}\tau_\mathrm{N}$$

$$\boxed{C_3 Tu_x\left(\bar{z}\right) = F(\bar{z}) + \frac{\Delta q_z}{8\pi^2}\sum_p \sum_{q_z} {}_{v_{\mathrm{g}z}>0} \int_0^{q_{r\max}} \frac{C_{\mathbf{q}p} q_r^3 d}{\omega^2 \tau_\mathrm{N}\Lambda_{\mathrm{N}z}} \int_0^1 Tu_x(\bar{z}')K\left(\bar{z},\bar{z}'\right) d\bar{z}' dq_r}$$

$$F(\bar{z}) = -C_1\frac{\partial T}{\partial x} - \frac{\partial T}{\partial x}\frac{\Delta q_z}{8\pi^2}\sum_p \sum_{q_z} {}_{v_{\mathrm{g}z}>0}\int_0^{q_{r\max}} \frac{C_{\mathbf{q}p} q_r^2 \Lambda_{\mathrm{C}r}}{\omega\tau_\mathrm{N}} \left\{2 + \frac{s-1}{1-s\exp\left(-1/Kn_{\mathrm{C}z}\right)}\left[\exp\left(-\frac{\bar{z}}{Kn_{\mathrm{C}z}}\right) + \exp\left(\frac{\bar{z}-1}{Kn_{\mathrm{C}z}}\right)\right]\right\} dq_r$$

$$K\left(\bar{z},\bar{z}'\right) = \exp\left(-\frac{\left|\bar{z}'-\bar{z}\right|}{Kn_{\mathrm{C}z}}\right) + \frac{s^2}{1-s^2\exp\left(-2/Kn_{\mathrm{C}z}\right)}\left[\exp\left(\frac{\bar{z}'-\bar{z}-2}{Kn_{\mathrm{C}z}}\right) + \exp\left(\frac{\bar{z}-\bar{z}'-2}{Kn_{\mathrm{C}z}}\right)\right]$$
$$+ \frac{s}{1-s^2\exp\left(-2/Kn_{\mathrm{C}z}\right)}\left[\exp\left(-\frac{\bar{z}'+\bar{z}}{Kn_{\mathrm{C}z}}\right) + \exp\left(\frac{\bar{z}'+\bar{z}-2}{Kn_{\mathrm{C}z}}\right)\right]$$

Methodology

- Heat flux calculation

$$J_x(z) = \sum_p \int v_{gx} \hbar \omega g \frac{d\mathbf{q}}{(2\pi)^3}$$

$$J_x(z) = \underbrace{J_{x1}(z)}_{\text{SMRT}} + \underbrace{J_{x2}(z)}_{\text{Drift correction}}$$

SMRT Drift correction

Fuchs-Sondheimer (F-S) model

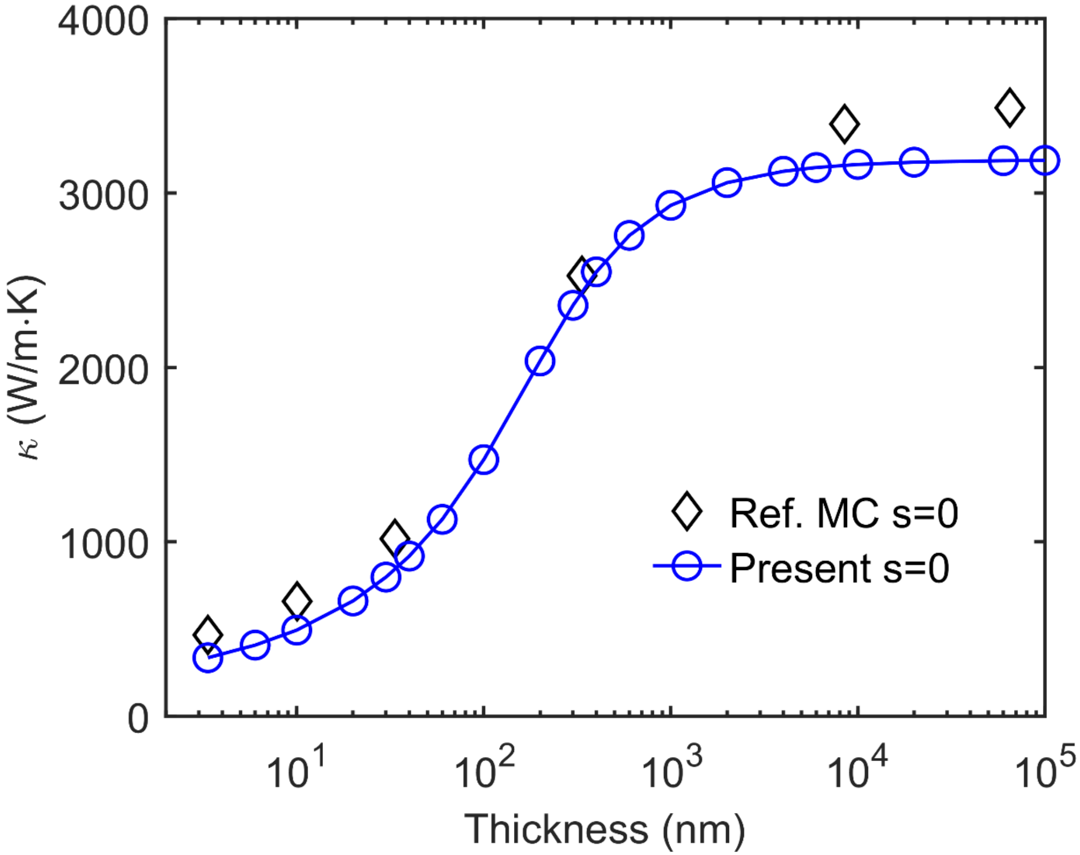
$$J_{x1}(\bar{z}) = -C_4 \frac{\partial T}{\partial x} - \frac{\partial T}{\partial x} \frac{\Delta q_z}{8\pi^2} \sum_p \sum_{q_z} {}_{v_{gz}>0} \int_0^{q_{r\max}} C_{\mathbf{q}p} v_{gr} \Lambda_{Cr} q_r \left\{ 2 + \frac{s-1}{1 - s\exp\left(-1/Kn_{Cz}\right)} \left[\exp\left(-\frac{\bar{z}}{Kn_{Cz}} \right) + \exp\left(\frac{\bar{z}-1}{Kn_{Cz}} \right) \right] \right\} dq_r$$

$$J_{x2}(\bar{z}) = C_1 T u_x(\bar{z}) + \frac{\Delta q_z}{8\pi^2} \sum_p \sum_{q_z} {}_{v_{gz}>0} \int_0^{q_{r\max}} \frac{C_{\mathbf{q}p} v_{gr} q_r^2 d}{\omega \Lambda_{Nz}} \int_0^1 T u_x(\bar{z}') K(\bar{z}, \bar{z}') d\bar{z}' dq_r$$

Results and Discussions

- Model validation

T = 200 K;
Isotopically pure;
Fully diffuse surface

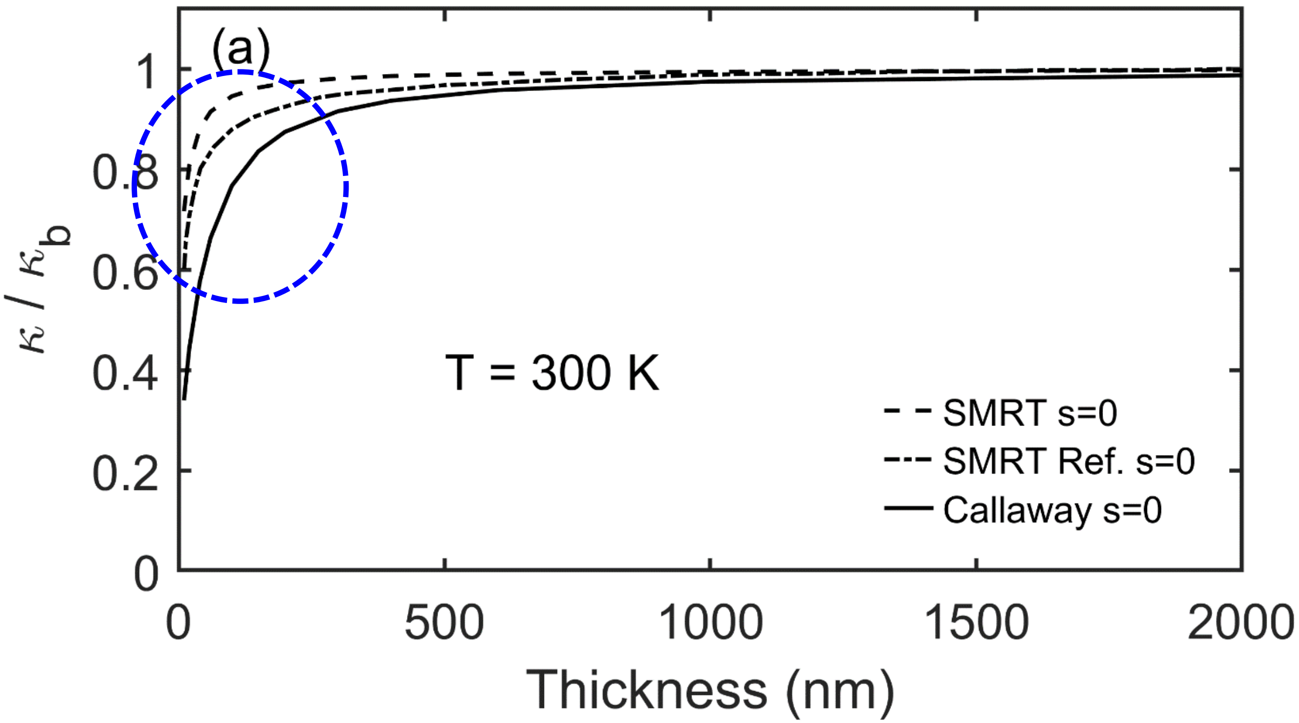


Ref. MC: Li *et al.* JAP, 2022 (MC solution of BTE with full scattering term)

Results and Discussions

- Thickness effect: SMRT vs. Callaway model

Nature abundance;
Fully diffuse surface



The SMRT approximation underestimates the size effect from finite thickness.

SMRT Ref.: Minnich APL 2015

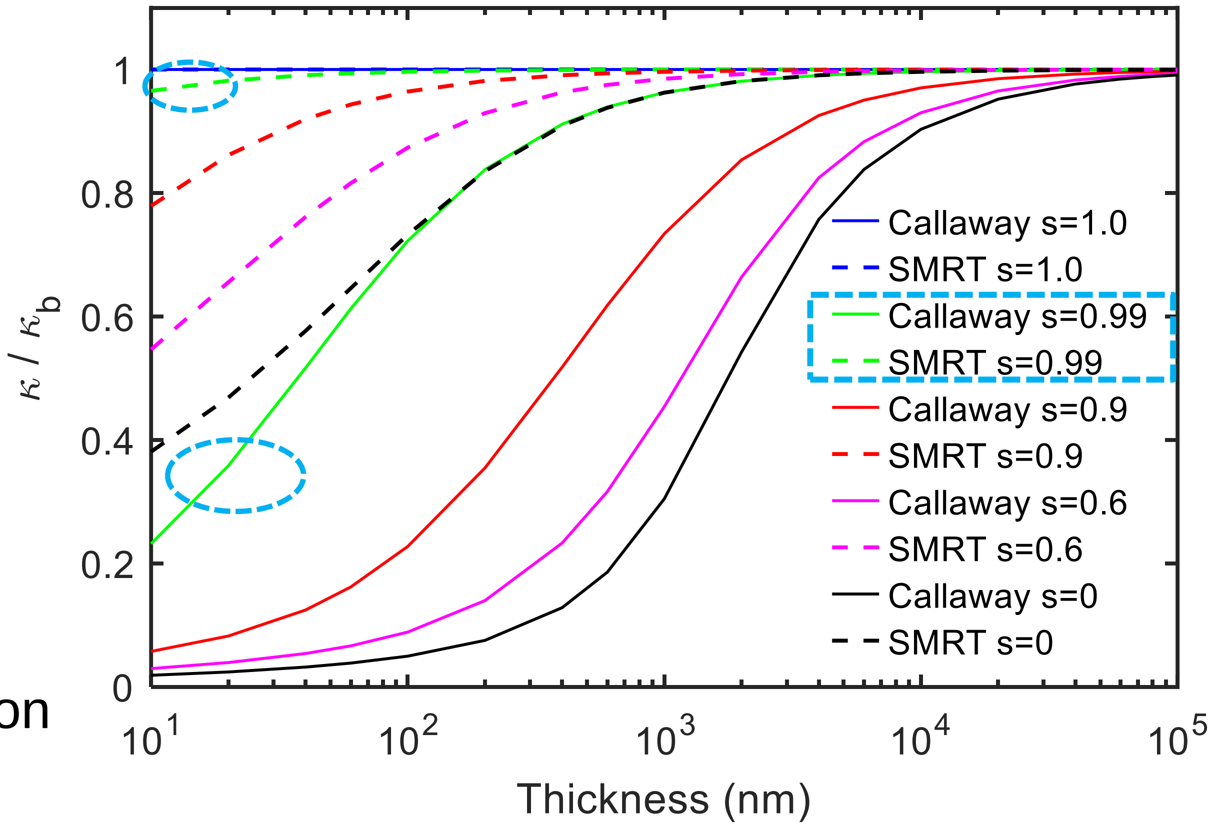
Results and Discussions

- Surface roughness effect: SMRT vs. Callaway's model

T = 100 K;
Isotopically pure

1) The SMRT model much underestimates the effect of surface roughness !

2) Small surface roughness would induce strong reduction of thermal conductivity !

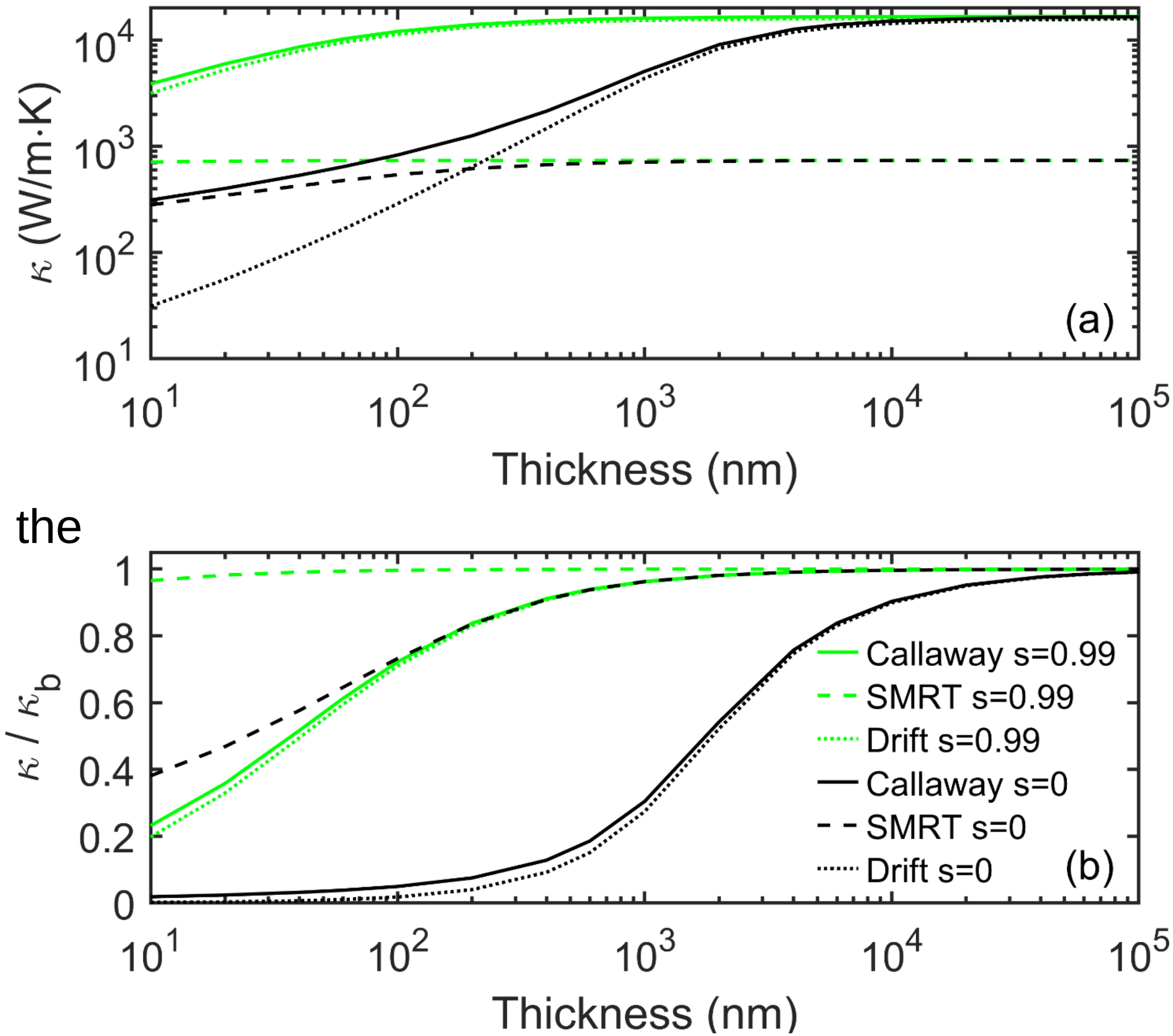


Results and Discussions

- Underlying mechanism

T = 100 K;
Isotopically pure

Strong size effect on the drift correction part !



Conclusions

- ✓ A semi-analytical BTE model for basal-plane heat transport along graphite thin film with finite thickness

- ✓ Significant thickness and surface roughness effects, which are much underestimated by the SMRT model

- ✓ The underlying mechanism comes from the strong size effect on the phonon drift motion.

Thank you for your attention ^_^

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- CREST JST (Grant Number JPMJCR19Q3)
- ANR project NearHeat (LCF Orsay and iLM Villeurbanne)