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Multiple phonon processes contributing to inelastic scattering during phonon transmission in thermal boundary conductance at solid interfaces

Patrick E. Hopkins

Engineering Sciences Center

Sandia National Laboratories

Albuquerque, NM, USA

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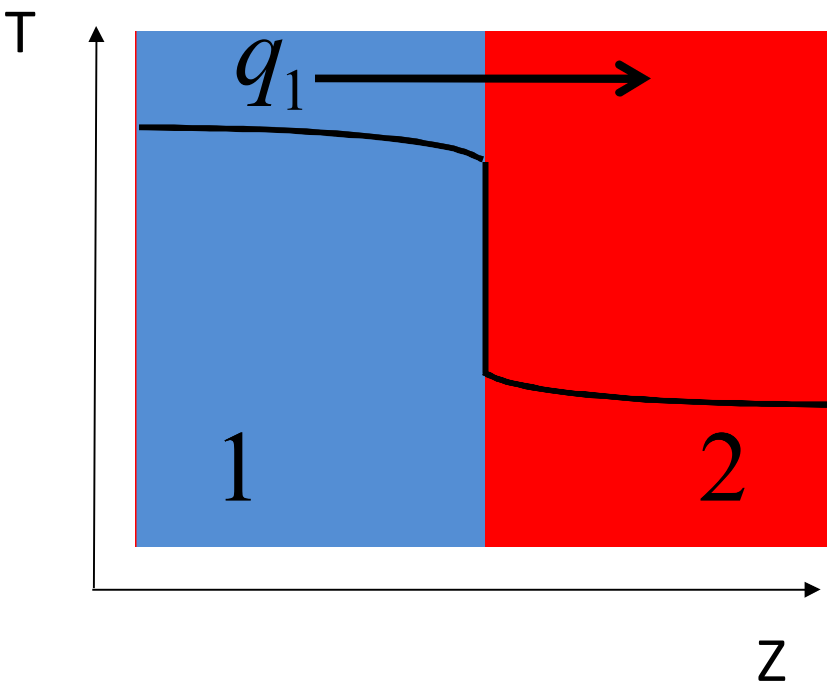
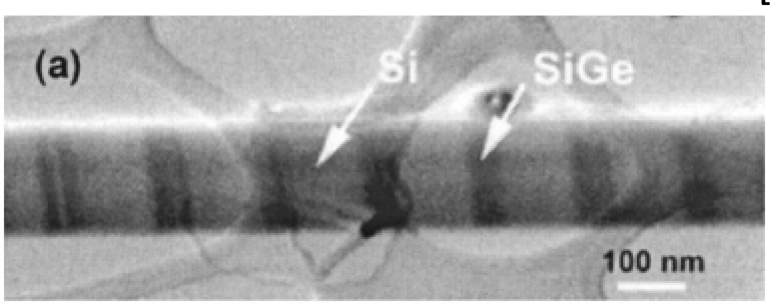
Thermal boundary conductance



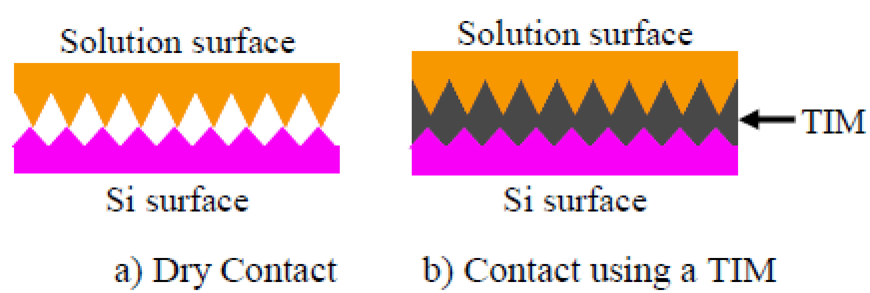
- G_K dominant thermal transport mechanism in nanostructures with thicknesses less than carrier mean free path
- Current nanoapplications rely on controlling phonon scattering in driving G_K

$$q_1 = G_K \Delta T$$

G_K= Thermal boundary conductance [$Wm^{-2}K^{-1}$]



Li *et al.*, *APL* **85**, 3186 (2003)



Samson *et al.*, *Intel technology Journal* **9**, 75 (2005)

Outline

Goal: Develop model that accounts for multiple phonon processes (inelastic scattering) in G_K

- Predicting G_K – the diffuse mismatch model (DMM)
- Assumptions and limitations of the DMM
- n-phonon processes in DMM transmission
- Inelastic DMM compared to data

Diffuse mismatch model (DMM)

$$q_1 = \frac{1}{2}\sum_j \int_0^{\pi/2} \int_0^{\omega_{c,1,j}} D_{1,j}(\omega) f(\omega,T) \hbar\omega v_{1,j} \alpha_{1,j}(\theta,\omega) \cos(\theta)\sin(\theta)\, d\omega\, d\theta = G_K \Delta T$$

Diffuse Mismatch Model (DMM)

E. T. Swartz and R. O. Pohl, *Reviews of Modern Physics*, **61**, 605 (1989)

diffuse scattering – phonon "looses memory" when scattered

$$G_K = \frac{1}{4}\sum_j \int_0^{\omega_{c,1,j}} D_{1,j}(\omega) \frac{f(\omega,T)}{\partial T} \hbar\omega v_{1,j} \alpha_1(\omega)\, d\omega$$

Isotropic Debye solid

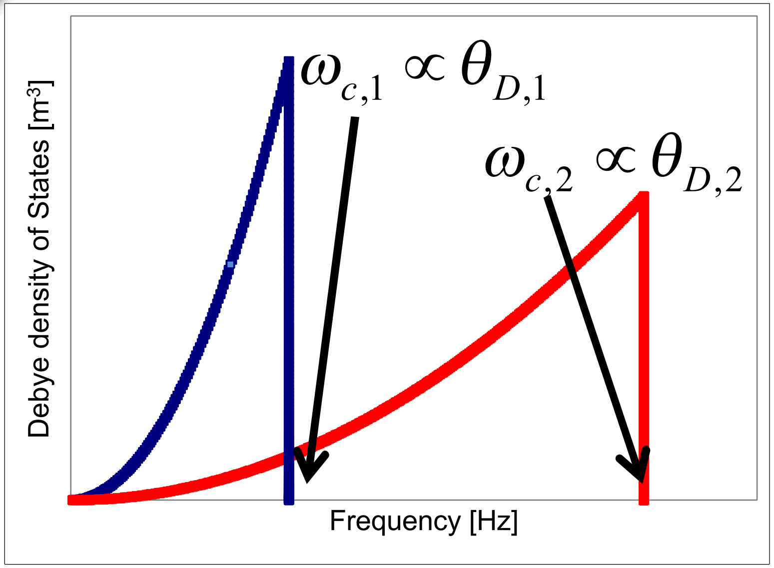
$$G_K = \frac{1}{6\pi^2}\sum_j v_{1,j}^{-2} \int_0^{\omega_{c,1,j}} \frac{f(\omega,T)}{\partial T} \hbar\omega^3 \alpha_1(\omega)\, d\omega$$

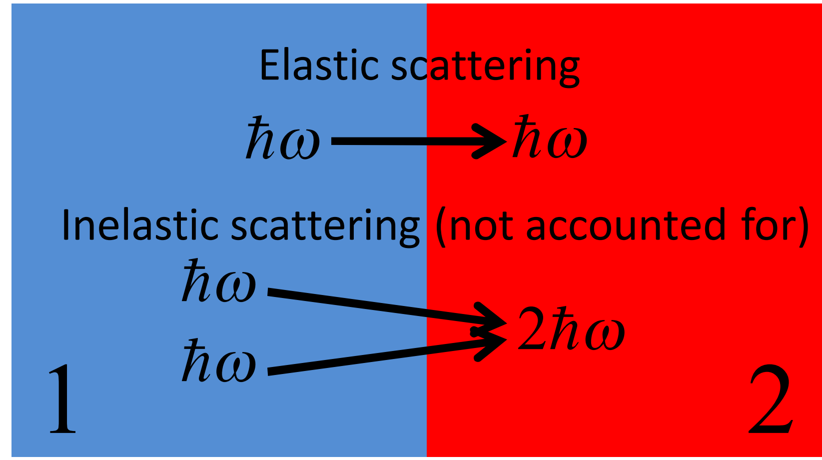
Elastic phonon scattering only in transmission

Loses frequency dependence

$$G_K = \frac{1}{6\pi^2}\sum_j v_{1,j}^{-2} \int_0^{\omega_{c,1,j}} \frac{f(\omega,T)}{\partial T} \hbar\omega^3 \alpha_1^{(2)}\, d\omega$$

Elastic phonon scattering





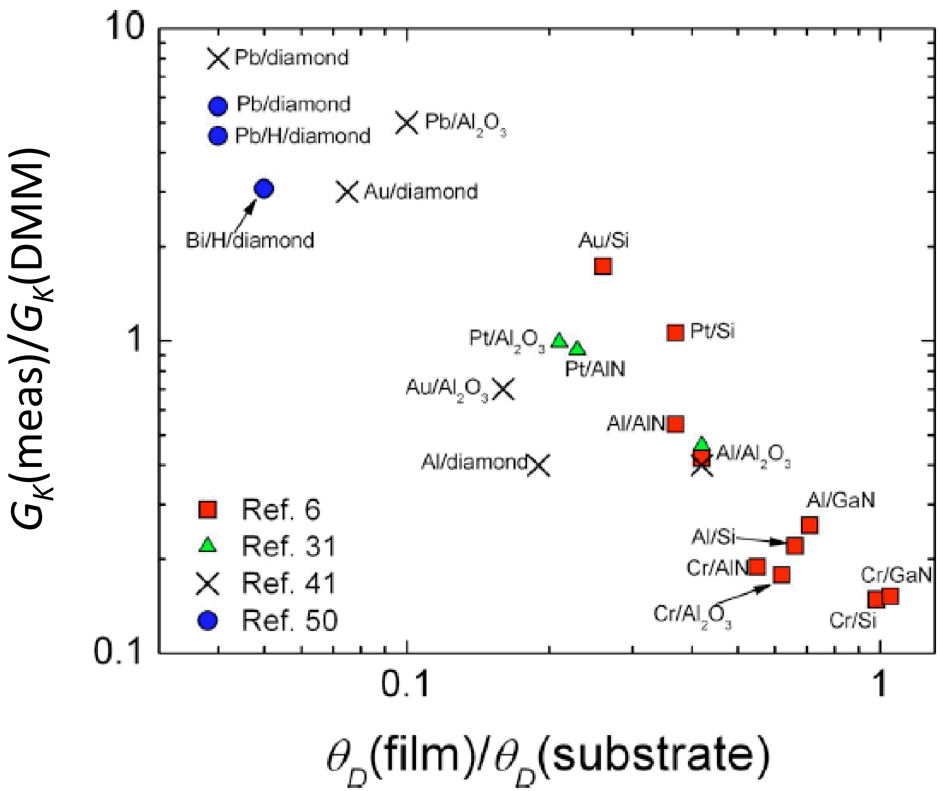
How do we calculate DMM elastic (2-phonon) transmission?

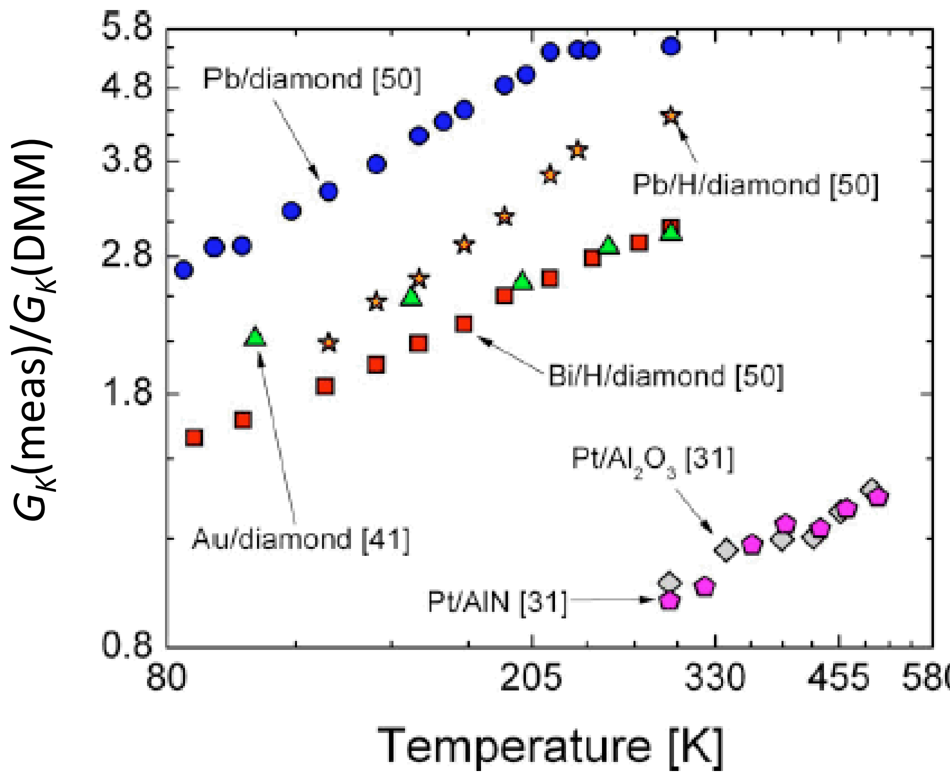
Principle of detailed balance

$$q_{1,\hbar\omega} = q_{2,\hbar\omega} \qquad \alpha_2 = r_1 = 1 - \alpha_1$$

$$\alpha_1^{(2)} = \frac{\sum_j v_{2,j}^{-2} \frac{f(\omega,T)}{\partial T}\omega^3}{\sum_j v_{1,j}^{-2} \frac{f(\omega,T)}{\partial T}\omega^3 + \sum_j v_{2,j}^{-2} \frac{f(\omega,T)}{\partial T}\omega^3} = \frac{\sum_j v_{2,j}^{-2}}{\sum_j v_{1,j}^{-2} + \sum_j v_{2,j}^{-2}}$$

Consequences





Norris and Hopkins, *JHT*, **131**, 043207 (2009)

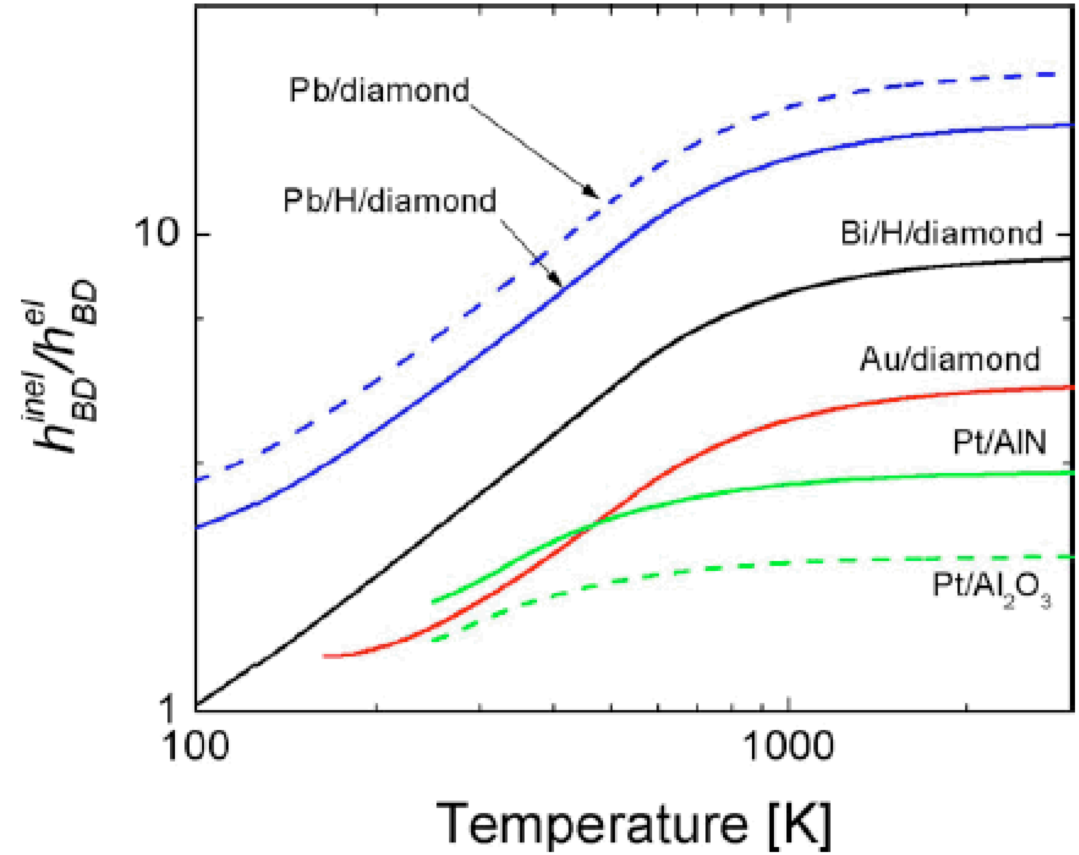
Ref. 6 – Stevens *et al.*, *JHT*, **127**, 315 (2005); Ref. 31 – Hopkins *et al.*, *JHT*, **130**, 022401 (2008)

Ref. 41 – Stoner and Maris, *PRB*, **48**, 16373 (1993);Ref. 50 – Lyeo and Cahill, *PRB*, **73**, 144301 (2006)

When $T>\theta_D$, Inelastic scattering causes linear trend in G_K

DMM predicts constant trend in G_K

Inelastic vs. elastic scattering



$\theta_{D,1}/\theta_{D,2} \propto 0.05$

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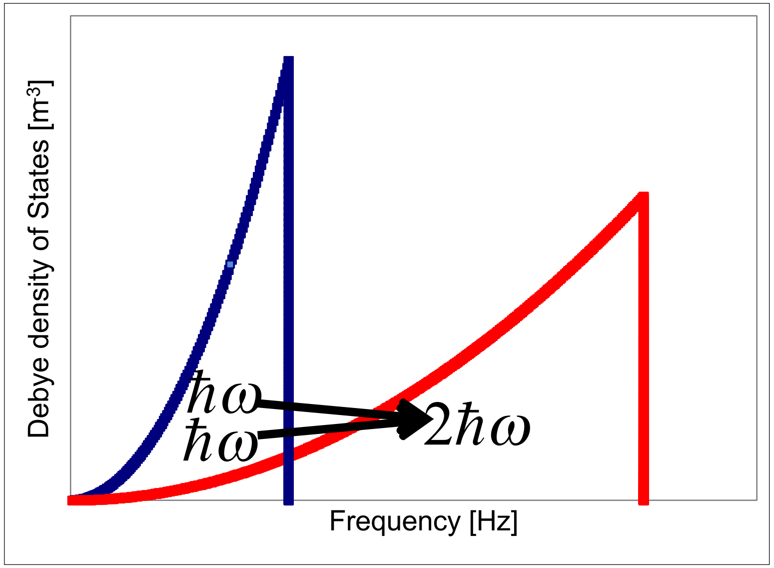
$\theta_{D,1}/\theta_{D,2} \propto 0.07$

$\theta_{D,1}/\theta_{D,2} \propto 0.2$

Hopkins and Norris, *JHT*, **141**, 022402 (2009)

- Inelastic (n-phonon) processes outweigh elastic (2 phonon) processes – more significant as $\theta_{D,1}/\theta_{D,2}$ decreases
- DMM accurately predicts elastic contribution when $\theta_{D,1}/\theta_{D,2} < 10\%$

3-phonon interface transmission



Principle of detailed balance

$$q_{1,2\hbar\omega,j} = q_{2,\hbar 2\omega,j}$$

Accounts for # phonons remaining after 2-phonon elastic processes

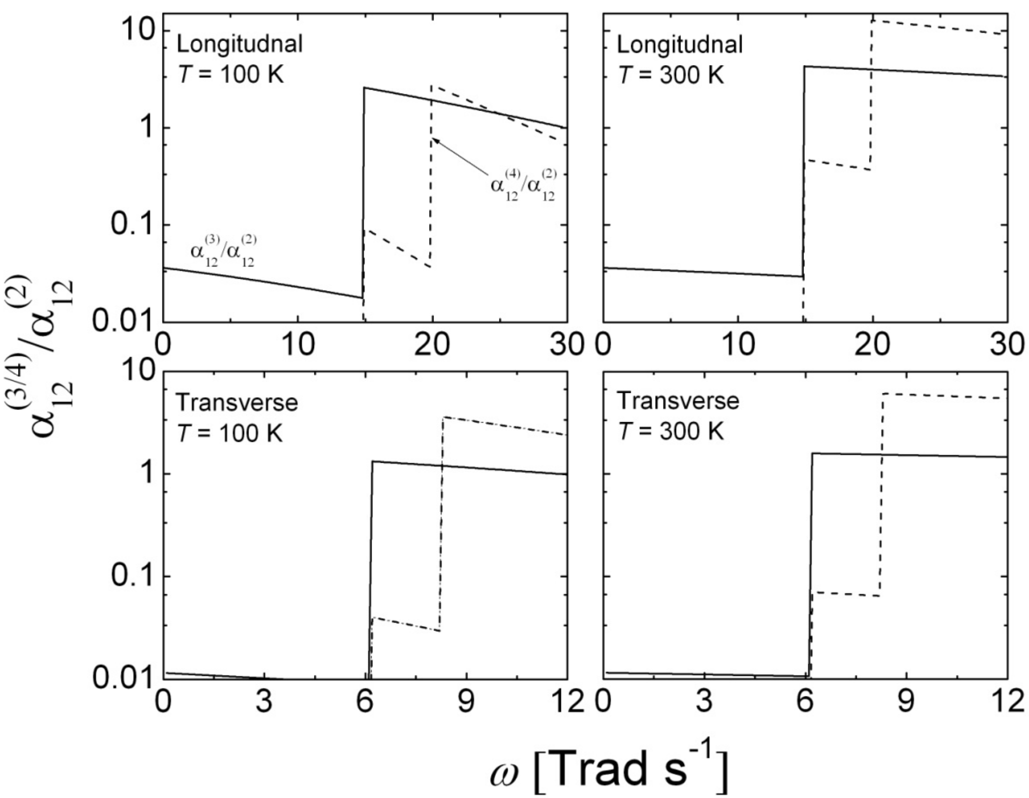
$$2(\hbar\omega)v_{1,j}\left(g_{1,j}(\omega)\right)f_0(\omega,T)\left[1-\alpha_{12}^{(2)}\right]\alpha_{12}^{(3)}(\omega,T)=\hbar(2\omega)v_{2,j}g_{2,j}(2\omega)f_0(2\omega,T)\alpha_{12}^{(2)}\left[1-\alpha_{12}^{(3)}(\omega,T)\right]$$

$$0<\omega\le\omega_{c,1,j}/2$$

$$2(\hbar\omega)v_{1,j}\left(g_{1,j}(\omega)\right)f_0(\omega,T)\left[1-\alpha_{12}^{(2)}\right]\alpha_{12,j}^{(3)}(\omega,T)=\hbar(2\omega)v_{2,j}g_{2,j}(2\omega)f_0(2\omega,T)\left[1-\alpha_{12,j}^{(3)}(\omega,T)\right]$$

$$\omega_{c,1,j}/2<\omega\le\omega_{c,1,j}$$

Higher order transmission: Pb/diamond



$$0 < \omega \le \omega_{c,1,j}/2$$

$$\alpha_{12,j}^{(3)}(\omega,T) = \frac{\dfrac{4\alpha_{12}^{(2)}}{v_{2,j}^2\left(\exp\left[\dfrac{2\hbar\omega}{k_B T}\right]-1\right)}}{\dfrac{\left[1-\alpha_{12}^{(2)}\right]}{v_{1,j}^2\left(\exp\left[\dfrac{\hbar\omega}{k_B T}\right]-1\right)}+\dfrac{4\alpha_{12}^{(2)}}{v_{2,j}^2\left(\exp\left[\dfrac{2\hbar\omega}{k_B T}\right]-1\right)}}$$

$$\omega_{c,1,j}/2 < \omega \le \omega_{c,1,j}$$

$$\alpha_{12,j}^{(3)}(\omega,T) = \frac{\dfrac{4}{v_{2,j}^2\left(\exp\left[\dfrac{2\hbar\omega}{k_B T}\right]-1\right)}}{\dfrac{\left[1-\alpha_{12}^{(2)}\right]}{v_{1,j}^2\left(\exp\left[\dfrac{\hbar\omega}{k_B T}\right]-1\right)}+\dfrac{4}{v_{2,j}^2\left(\exp\left[\dfrac{2\hbar\omega}{k_B T}\right]-1\right)}}$$

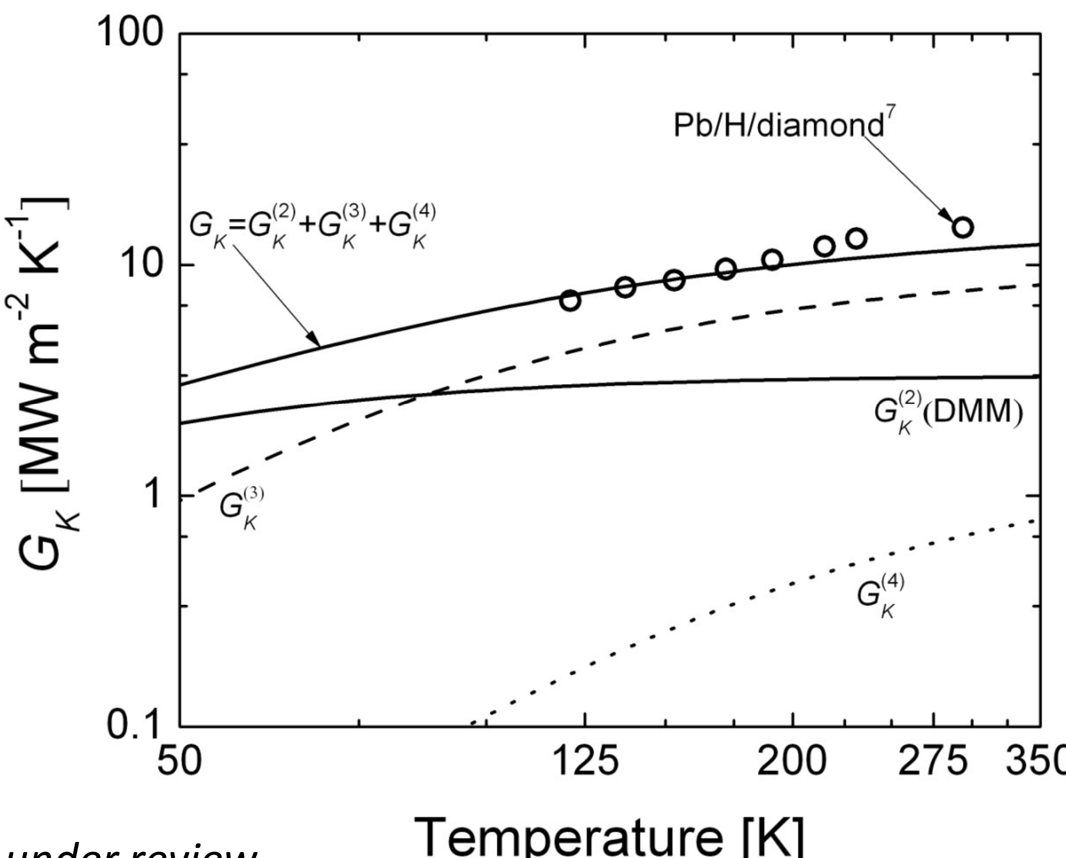
Inelastic G_K

$$G_K = \sum_n G_K^{(n+1)} = G_K^{(2)} + G_K^{(3)} + G_K^{(4)}$$

Elastic (2 phonon) DMM

$\propto \alpha_{12}^{(3)}$

$\propto \alpha_{12}^{(4)}$



Pb/H/diamond data from: Lyeo and Cahill, *PRB* **73**, 144301 (2006)

$\theta_{D,1} / \theta_{D,2} \propto 0.05$

Hopkins, *currently under review*

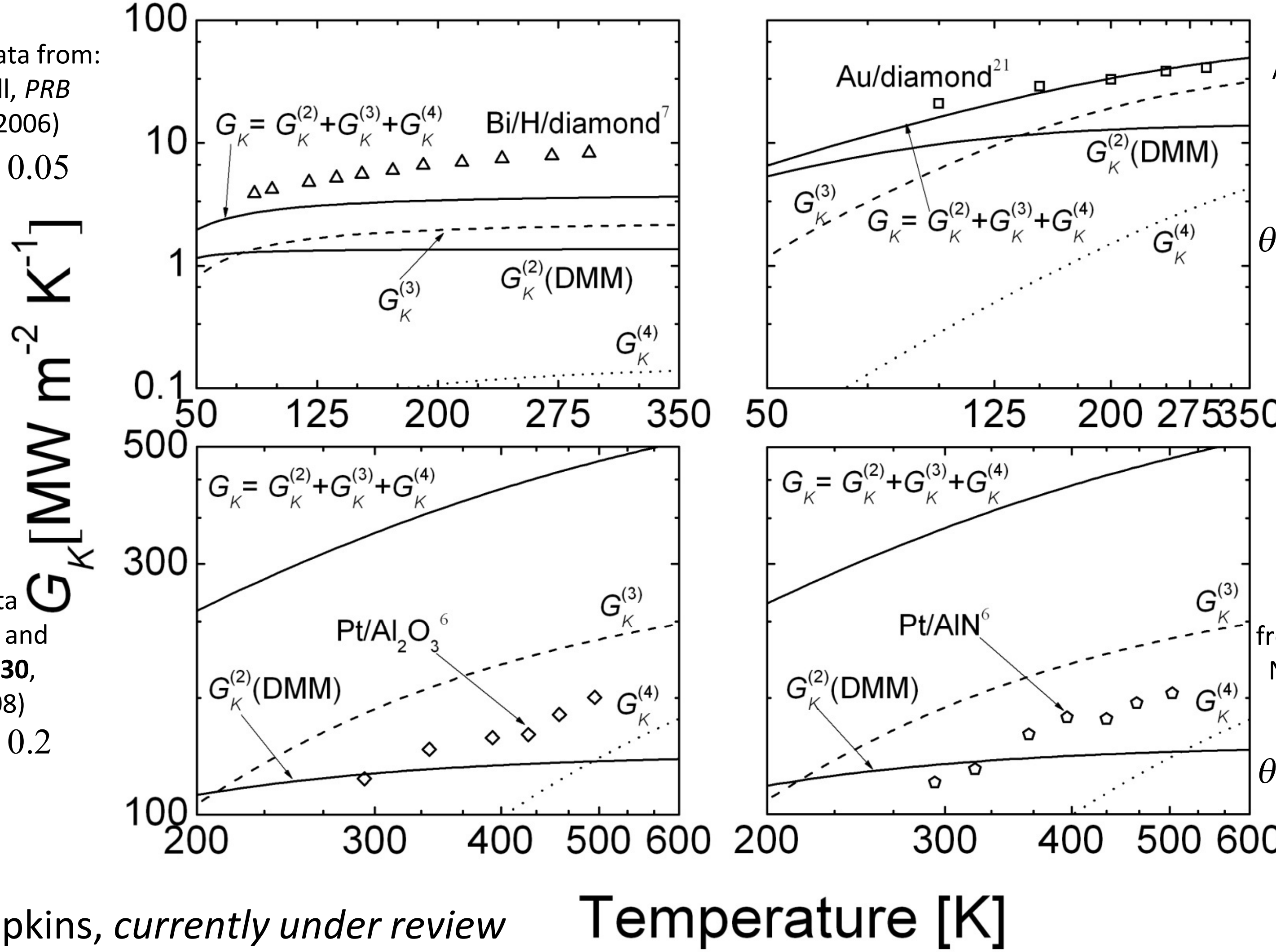
Inelastic G_K

Bi/H/diamond data from: Lyeo and Cahill, *PRB* **73**, 144301 (2006)

$\theta_{D,1} / \theta_{D,2} \propto 0.05$

Au/diamond data from: Stoner and Maris, *PRB* **48**, 16373 (1993)

$\theta_{D,1} / \theta_{D,2} \propto 0.07$



Pt/Al_2O_3 data from: Hopkins and Norris, *JHT*, **130**, 022401 (2008)

$\theta_{D,1} / \theta_{D,2} \propto 0.2$

Pt/AlN data from: Hopkins and Norris, *JHT*, **130**, 022401 (2008)

$\theta_{D,1} / \theta_{D,2} \propto 0.2$

Hopkins, *currently under review*

Conclusions

Goal: Develop model that accounts for multiple phonon processes (inelastic scattering) in G_K

- Only accounting for 2 phonon (elastic) processes in the DMM does not account for all phonon processes
- By conservation of energy and phonon number, new diffusive *n*-phonon transmission is developed
- Effects of *n*-phonon interfacial processes on G_K is calculated with new *n*-phonon DMM
- Improved agreement with data using *n*-phonon DMM

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LABORATORY DIRECTED RESEARCH & DEVELOPMENT

n-phonon trasmission

$$\alpha_{12,j}^{(n)}(\omega,T)=\frac{\dfrac{(n-1)^2\prod_{x=0}^{n-3}\alpha_{12}^{(x+2)}}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}}{\dfrac{\prod_{x=0}^{n-3}\left[1-\alpha_{12}^{(x+2)}\right]}{v_{1,j}^2\left(\exp\left[\dfrac{\hbar\omega}{k_BT}\right]-1\right)}+\dfrac{(n-1)^2\prod_{x=0}^{n-3}\alpha_{12}^{(x+2)}}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}},\ 0<\omega<\frac{\omega_{c,j}}{(n-1)},$$

$$\alpha_{12,j}^{(n)}(\omega,T)=\frac{\dfrac{(n-1)^2\prod_{x=1}^{n-3}\alpha_{12}^{(x+2)}}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}}{\dfrac{\prod_{x=0}^{n-3}\left[1-\alpha_{12}^{(x+2)}\right]}{v_{1,j}^2\left(\exp\left[\dfrac{\hbar\omega}{k_BT}\right]-1\right)}+\dfrac{(n-1)^2\prod_{x=1}^{n-3}\alpha_{12}^{(x+2)}}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}},\ \frac{\omega_{c,j}}{(n-1)}<\omega<\frac{2\omega_{c,j}}{(n-1)},$$

$$\vdots$$

$$\alpha_{12,j}^{n}(\omega,T)=\frac{\dfrac{(n-1)^2}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}}{\dfrac{\prod_{x=0}^{n-3}\left[1-\alpha_{12}^{(x+2)}\right]}{v_{1,j}^2\left(\exp\left[\dfrac{\hbar\omega}{k_BT}\right]-1\right)}+\dfrac{(n-1)^2}{v_{2,j}^2\left(\exp\left[\dfrac{(n-1)\hbar\omega}{k_BT}\right]-1\right)}},\ \frac{(n-2)\omega_{c,j}}{(n-1)}<\omega<\omega_{c,j}.$$