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# TEMPERATURE DEPENDENCE OF THERMAL CONDUCTIVITY OF GRAPHENE MONOLAYER IN THE FRAMEWORK OF DEBYE AND CALLAWAY MODELS

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## INTRODUCTION

The temperature dependence of thermal conductivity of graphene monolayer is analyzed, based on semi-classical Boltzmann transport equation in the approximation of relaxation time in the framework of Debye and Callaway models. In both cases the phonon relaxation mechanisms on monolayer boundaries, impurities, and phonon-phonon interactions are considered. Electron-phonon and electron-electron interactions are neglected as they provide small contributions to thermal conductivity. The three-phonon scattering *N*-processes in graphene monolayer thermal conductivity are accounted within Callaway model, while neglected within Debye model. It is shown that Callaway model is more succesful than Debye model in explaining experimental measurements of thermal conductivity in graphene.

## SIGNIFICANCE OF THERMAL CONDUCTIVITY

Thermal conductivity is one of the most important indicators for the reliable and efficient use of graphite-based nanoelectronic devices. At nano level, one of the important problems that arises is the local heating of individual parts of the device. This localized heat, if not distributed, causes the formation of hot spots limiting the maximum current density, which can cause material damage of the device.

## COEFFICIENT OF THERMAL CONDUCTIVITY

The main contribution to thermal conductivity of graphene is given by phonons. The coefficient of thermal conductivity is found by solving the Boltzmann transport equation in the approximation of the relaxation time. In this paper, two theoretical approaches are used to find coefficient of thermal conductivity: Debye and Callaway.

Debye model is based on the following assumptions:

1. Solid state is considered an isotropic elastic continuum;
2. Thermal energy of the system is conducted only by acoustic phonon branches;
3. Dispersion law for 3 acoustic branches is: $\omega_{\mathrm{LA}} = v_{\mathrm{LA}} q$ , $\omega_{\mathrm{TA}} = v_{\mathrm{LA}} q$ , $\omega_{\mathrm{ZA}} = v_{\mathrm{LA}} q^{1,64}$ .
4. Phonon frequences are in the range: $0 \le \omega_s \le \omega_D$ , where $s \in (\mathrm{LA,TA,ZA})$ and $\omega_D$ is Debay frequency, maximum frequency in the spectrum.
5. Phonon density of states is given by: $D(\omega_s) = A\omega_s / 2\pi v_s^2$ .
6. In phonon spectrum there is no difference between phase and group velocities.

Coefficient of thermal conductivity in Debye model is:

$$\kappa_D = \frac{1}{4\pi L_z k_B T^2} \sum_s \int_0^{\omega_D} (\hbar\omega_s)^2 \bar{n}_s \left(\bar{n}_s + 1\right) \tau(\omega_s) \omega_s \mathrm{d}\omega_s . \qquad \bar{n}_s = 1/\left(\mathrm{e}^{\hbar\omega_s / k_B T} - 1\right)$$

## MECHANISMS OF PHONON SCATTERING

The phonon relaxation mechanisms on monolayer boundaries, impurities, and phonon-phonon interactions are considered. In Debye approach only so called *U* processes of phonon-phonon scattering are accounted. Although Peierls showed that only *U* phonon-phonon scattering contributes to final values of the coefficient of thermal conductivity, it has been shown later that this conclusion does not hold for 2D structures, and that so called *N*-processes of phonon-phonon scattering must be accounted as well.

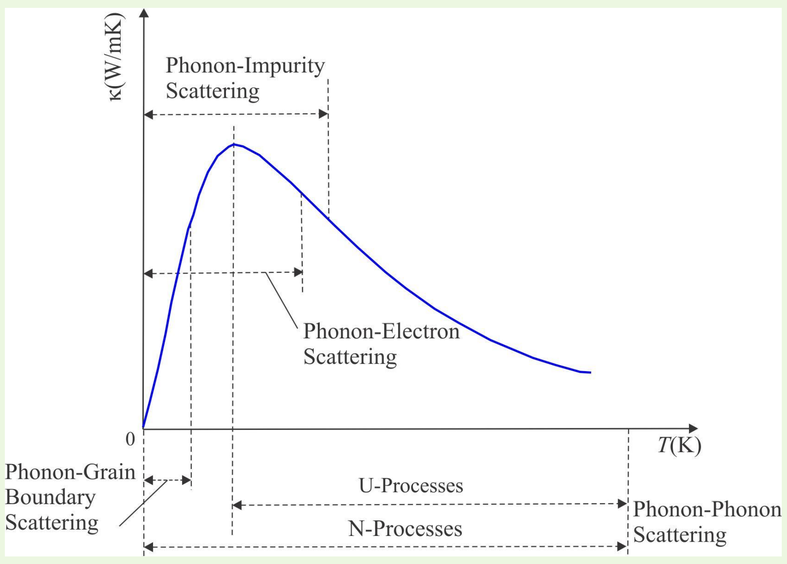


*Figure 1. Mechanisms of phohon scattering with thermal ranges of their dominance.*

## CALLAWAY MODEL

In Callaway approach the non-trivial role of *N*-processes of phonon-phonon scattering is accounted. Also, difference between phase and group velocities of phonons is made.

$$\kappa_{\mathrm{C}} = \kappa_{\mathrm{D}} + \kappa_{\mathrm{N\text{-}drift}} \qquad \kappa_{\mathrm{D}} = \frac{1}{4\pi L_z k_B T^2} \sum_s \int_0^{\omega_{\mathrm{D}}} (\hbar\omega_s)^2 \bar{n}_s \left(\bar{n}_s + 1\right) \tau(\omega_s) \frac{\upsilon_s}{u_s} \omega_s \mathrm{d}\omega_s .$$

$$\kappa_{\mathrm{N\text{-}drift}} = \frac{1}{4\pi L_z k_B T^2} \sum_s \frac{\left\{ \int_0^{\omega_{\mathrm{D}}} \left(\hbar\omega_s\right)^2 \bar{n}_s \left(\bar{n}_s + 1\right) \tau(\omega_s) \tau_{\mathrm{N}}^{-1}(\omega_s) \frac{\upsilon_s}{u_s} \omega_s \mathrm{d}\omega_s \right\}^2}{\int_0^{\omega_{\mathrm{D}}} \left(\hbar\omega_s\right)^2 \bar{n}_s \left(\bar{n}_s + 1\right) \tau_{\mathrm{N}}^{-1}(\omega_s) \left(1 - \tau(\omega_s)\tau_{\mathrm{N}}^{-1}(\omega_s)\right) \frac{\upsilon_s}{u_s} \omega_s \mathrm{d}\omega_s} .$$

where $u_s$ is phase velocity $v_s$ is group velocity of phonons.

## PHONON RELAXATION TIMES

Total phonon relaxation time is given by Matthiessen rule: $\tau^{-1} = \sum_i \tau_i^{-1}$

Phonon relaxation time on boundaries is:

$$\frac{1}{\tau_B} = \frac{1 - p}{1 + p} \frac{v_s}{L_x},$$ ($p$ is coefficient of reflextion, $L_x = 2{,}9\,\mu\mathrm{m}$ is adopted linear dimension of graphen sample)

Phonon relaxation time on impurities is:

$$\frac{1}{\tau_{pd}} = A_d \frac{2\pi\omega^3}{\omega_D^2},$$

Phonon relaxation time for *U* phonon-phonon scattering is:

$$\tau_U^{-1} = B_U \mathrm{e}^{-\frac{T_D}{\alpha T}} \omega^2 T^3$$ (with adopted coefficient $\alpha$ = 3)

Phonon relaxation time for *N* phonon-phonon scattering is:

$$\tau_N^{-1} = B_N \omega^2 T^3$$

## RESULTS OF NUMERICAL ALANALISYS

*Table 1. Hereby adopted relevant parameters for calculations of graphene monolayer coefficients of thermal conductivities.*

| $L_z$ [nm] | $v_{LA}$ [km/s] | $v_{TA}$ [km/s] | $v_{ZA}$ [km/s] | $B_N$[sK$^{-3}$] | $B_U$[sK$^{-3}$] | $A_d$ |
|---|---|---|---|---|---|---|
| 0.35 | 18.4 | 16.5 | 9.2 | $3{,}85\cdot10^{-25}$ | $7{,}7\cdot10^{-25}$ | $4.5\cdot10^{-4}$ |
| $p$ | $\omega_{D,LA}$ [Hz] | $\omega_{D,TA}$ [Hz] | $\omega_{D,ZA}$ [Hz] | $T_{D,LA}$[K] | $T_{D,TA}$[K] | $T_{D,ZA}$[K] |
| 0.9 | $2.66\cdot10^{14}$ | $2.38\cdot10^{14}$ | $1.32\cdot10^{14}$ | 2032 | 1818 | 1008 |

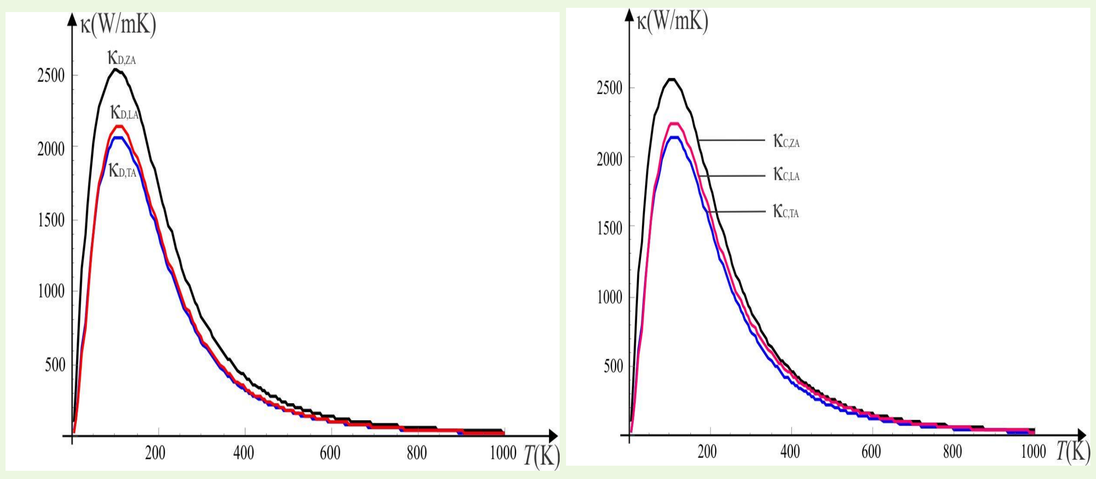


*Figure 2. Coefficients of graphene monolayer thermal conductivity for all three phonon polarizations (LA, TA, ZA) in Debye and Callaway models.*

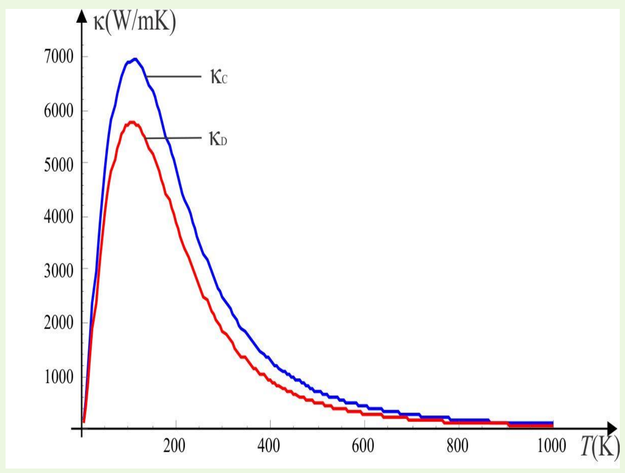


*Figure 3. Coefficients of graphene monolayer total thermal conductivity in Debye and Callaway models.*

## CONCLUSION

The obtained values of coefficients of graphene monolayer thermal conductivity in both Debye and Callaway models vary for differerent phonon polarizations. In both models the highest coefficients of graphene monolayer thermal conductivity are achieved for ZA, then LA, and the least for TA phonon polarizations.

By analysis of the obtained coefficients of total termal conductivity of graphene monolayer in the temperature range 0 – 1000 K, we see that Debye model predicts maximum value of ~ 5800 W/mK at ≤ 200 K, while Callaway model predicts higher maximum value of ~ 7000 W/mK at higher temperature, in better agreement with experimentally observed values up to 1000 K. This means that *N*-processes of phonon-phonon scattering must be accounted, especially having in mind the nonlinear dispersion law for ZA phonon branch.

Results of theoretical approaches of other authors predict graphene monolayer thermal conductivity from several hundreds W/mK up to 6600 W/mK in Debye model, and from several tens W/mK up to 9500 W/mK in Callaway model. Results of different experimental measurements of graphene monolayer thermal conductivity also vary significantly: from 2000 up to 10000 W/mK. These discrepances are considered as a result of non-ideal flatness of different graphen monolayer samples. The reason could also be in the uncertain concept of "thickness" in 2D structures, which might change our concepts of diffusion, electrical and thermal conductivities, and their quantitative values.