YUCOMAT 2014, Herceg Novi, Montenegro

TEMPERATURE DEPENDENCE OF GRAPHENE TRANSPORT PROPERTIES

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INTRODUCTION

The exceptional transport properties of graphene, coupled with its high thermal, mechanical and chemical stabilities, provide wide opportunities for practical application. Temperature dependencies of graphene thermal and electrical characteristics will be hereby analyzed in the wide range of 50–400 K, by solving semiclassical Boltzmann equations in the approximations of relaxation time. Basic relaxation mechanisms in graphene monolayer will be accounted, with corresponding relaxation times introduced phenomenologically. In the case of graphene thermal conductivity, the exact contribution of all phonon branches will be accounted. For calculation of graphene electrical conductivity, different mechanisms of scattering of charge carriers and their partial and overall contributions will be accounted. The theoretical results will be compared with the experimentally observed.

THERMAL CONDUCTIVITY OF GRAPHENE

Thermal conductivity of graphene is basically phonon-based, since its electronic-based thermal conductivity represents less than 1% of the total thermal conductivity at room temperature. Therefore, we consider the thermal conductivity of graphene as a consequence of phonon scatterings on boundaries, on defects, and on phonons.

Characteristics of phonon spectrum are determined by 2D structure of graphene. Graphene has a hexagonal structure with two carbon atoms in each cell. This determines the appearance of the six phonon branches in the dispersion spectrum: three acoustic (LA, TA, ZA) and three optical (LO, TO, ZO). The main contribution to thermal conductivity of graphene is provided by acoustic phonon branches, while contribution of optical phonon branches is neglected.

LA and TA acoustic modes correspond to longitudinal and transverse phonon oscillations in a graphene plane, and have linear dispersion laws, $\omega_1 = v_1 q$, $\omega_2 = v_2 q$, while ZA acoustic mode corresponds to oscillations of phonons in the direction normal to the direction of LA and TA oscillation modes, and has nonlinear dispersion law, $\omega_3 = v_3 q^{1.5}$.

The separate contribution of all three acoustic phonon branches, as well as their total contribution to thermal conductivity of graphene are analysed. To calculate thermal conductivity of graphene, the Boltzmann equation in the approximation of relaxation time is used:

$$\kappa = \frac{1}{4\pi\hbar k_B T^2} \sum_s \int_{\omega_{\min}}^{\omega_{\max}} (\hbar\omega_s)^2 e^{\frac{\hbar\omega_s}{k_B T}} \left(e^{\frac{\hbar\omega_s}{k_B T}} - 1 \right)^{-2} (\tau_B^{-1} + \tau_U^{-1} + \tau_{pd}^{-1})^{-1} \omega_s d\omega_s$$

with adopted relaxation times for phonon scatterings on boundaries, phonons and impurities:

$$\frac{1}{\tau_B} = \frac{1-p}{1+p}\frac{\upsilon}{L}, \qquad \frac{1}{\tau_u} = 2\gamma^2 \frac{k_B T}{M\upsilon^2}\frac{\omega^2}{\omega_D}, \qquad \frac{1}{\tau_{pd}} = \frac{\omega^3 \Gamma S}{2\upsilon^2} \qquad \Gamma = \sum_i c_i (1-c_i)\left(\frac{\Delta M_i}{M}\right),$$

Calculations are performed using the software package *Mathematica*. Index *s* has values *s* ϵ *(TA, LA, ZA)* while relevant parameters are given in table below:

h nm	L μm	v_{LA} km/s	v_{TA} km/s	v_{ZA} km/s	γ_{LA}	γ_{TA}	γ_{ZA}
0.35	100	18.4	16.5	9.2	1.8	1.6	1.2
M kg	Γ	S_0 m^2	p	$\omega_{D,LA}$ Hz	$\omega_{D,TA}$ Hz	$\omega_{D,ZA}$ Hz	$\omega_{\min}$ Hz
$2\cdot10^{-26}$	0.1	$4.61\cdot10^{-20}$	0.9	$2.66\cdot10^{14}$	$2.38\cdot10^{14}$	$1.32\cdot10^{14}$	$3.0\cdot10^{12}$

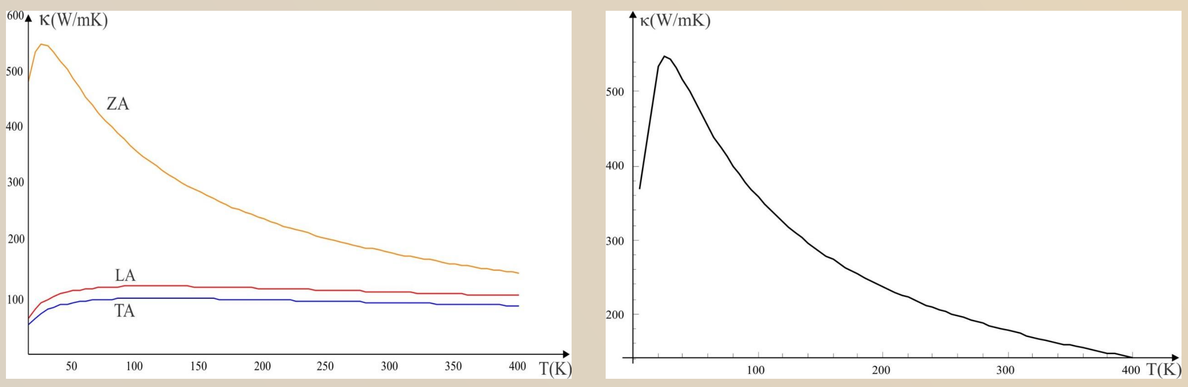

Fig 1. Temperature dependence of graphene thermal conductivity calculated separately for three acoustic phonon branches and for all three relaxation mechanisms accounted (left), and their total contribution to graphene thermal conductivity (right)

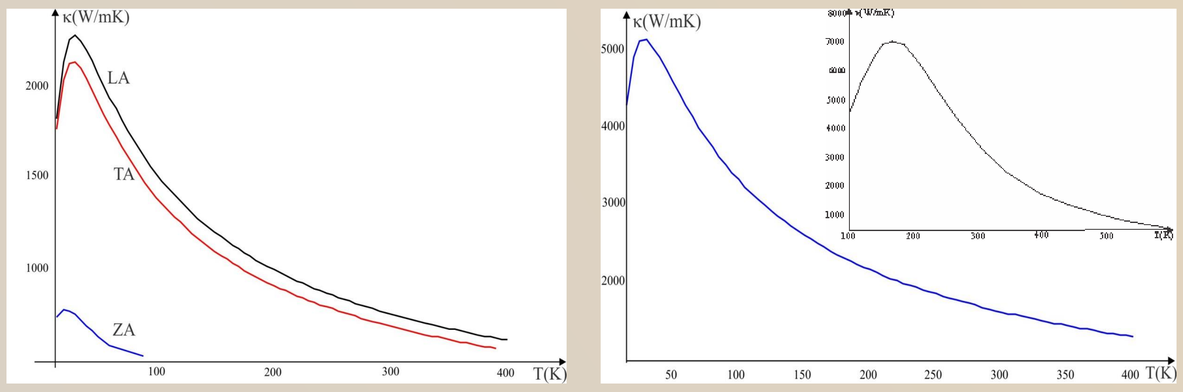

*Fig 2. Temperature dependence of graphene thermal conductivity calculated separately for three acoustic phonon branches , without boundaries and impurities (left), and their total contribution to graphene thermal conductivity (right) with upper insert presenting theoretical and experimental data from ref. A. Alofi, G.P. Srivastava, Phys. Rev B **87**, 115421, 2013.*

ELECTRICAL CONDUCTIVITY OF GRAPHENE

The graphene dispersion low, in case we take into account only area in the vicinity of Dirac points, can be significantly simplified: $E = \hbar v_F k$.

Further on, for finding graphene electrical conductivity we apply semiclassical Boltcmann transport equation in the approximation of relaxation time:

$$\sigma = \frac{e^2}{2}\int_0^\infty \frac{g_s g_v}{2\pi(\hbar v_F)^2} v^2 \tau(\varepsilon)\left(-\frac{\partial f_0}{\partial \varepsilon}\right) = \frac{e^2}{2}\int_0^\infty D(\varepsilon) v^2 \tau(\varepsilon)\left(-\frac{\partial f_0}{\partial \varepsilon}\right).$$

To account for total relaxation time we apply Matthiessen rule, with adopted relaxation times for electron scattering on impurities (charged, with long-range Coulomb interaction, and neutral, with short-range Coulomb interaction), and for electron scattering on phonons (in high temperature range, $T \geq T_{BG}$, and in low temperature range, $T << T_{BG}$; T_{BG} is Bloch-Grüneisen temperature):

$$\frac{1}{\tau_{im}} = \begin{cases} \dfrac{u_0^2}{\hbar\varepsilon_F}; \\ \dfrac{n_d V_0^2}{\hbar}\dfrac{\varepsilon_F}{4(\hbar v_F)^2}, \end{cases} \qquad \frac{1}{\tau_{ph}(\varepsilon)} = \begin{cases} \dfrac{1}{\hbar^3}\dfrac{\varepsilon}{4v_F^2}\dfrac{D^2}{\rho_m v_{ph}^2} k_B T; \\ \dfrac{1}{\pi}\dfrac{1}{\varepsilon_F}\dfrac{1}{k_F}\dfrac{D^2}{2\rho_m v_{ph}}\dfrac{4!\zeta(4)}{(\hbar v_{ph})^4}(k_B T)^4. \end{cases}$$

For numerical calculation of electrical conductivity, we adopt the following values of parameters:

$$D = 30{,}4\cdot10^{-19}\text{J}; \quad \rho_m = 7{,}6\cdot10^{-7}\frac{\text{kg}}{\text{m}^2}; \quad v_{ph} = 2\cdot10^4\frac{\text{m}}{\text{s}}; \quad \varepsilon_F = 4{,}168\cdot10^{-19}\text{J};$$

$$v_F = 10^6\frac{\text{m}}{\text{s}}; \quad k_F = 4{,}8\cdot10^8\,\text{m}^{-1}; \quad n_i^C = 4\cdot10^{15}\,\text{m}^{-2}; \quad Z = 1; \quad \tilde{\varepsilon}_r = 2{,}4; \quad \tilde{\gamma} = 4{,}2;$$

$$n_d = 0{,}4\cdot10^{14}\,\text{m}^{-2}; \quad V_0 = 16\cdot10^{-37}\,\text{Jm}^2; \quad T_{BG} = 54\text{K}.$$

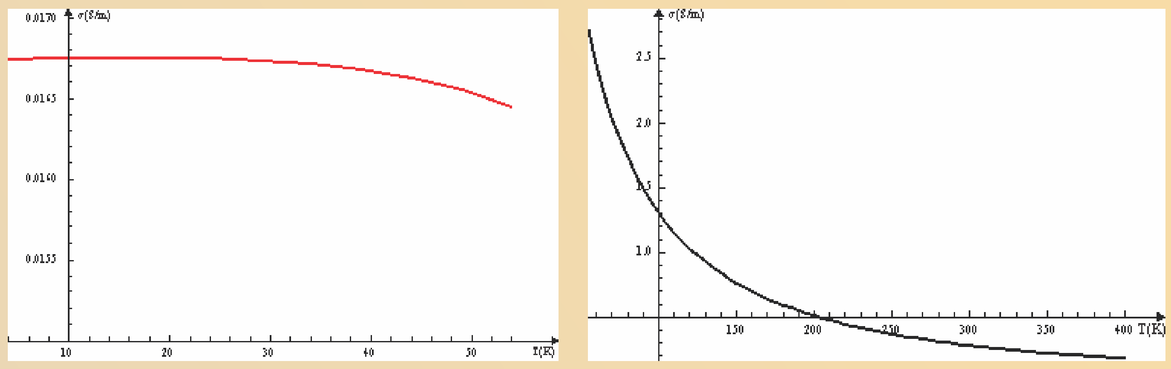

Fig 3. Temperature dependence of graphene electrical conductivity for relaxation on neutral impurities, in temperature ranges $T << T_{BG}$ (left) and $T \geq T_{BG}$ (right)

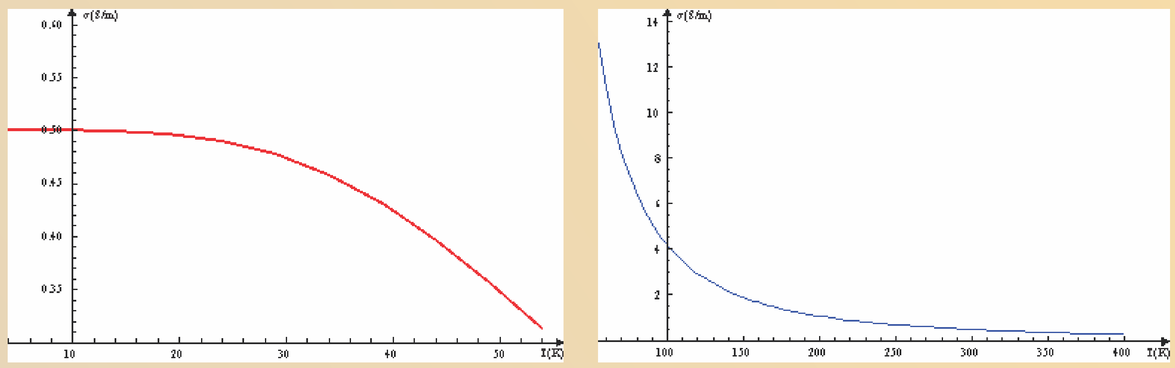

Fig 4. Temperature dependence of graphene electrical conductivity for relaxations on both charged impurities and phonons, in temperature ranges $T << T_{BG}$ (left) and $T \geq T_{BG}$ (right)

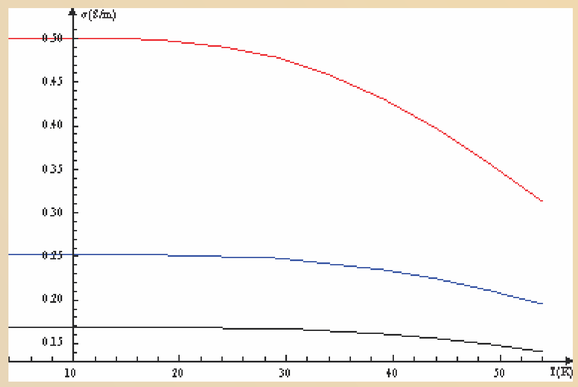

Fig 5. Low-temperature dependence of graphene electrical conductivity (for $T << T_{BG}$) for relaxations on both charged impurities and phonons, for several concentrations of impurities as parameters

CONCLUSION

Graphene thermal conductivity is phonon-based, and behaves at low-temperatures as $\kappa \approx T^2$ while at high-temperatures behaves as $\kappa \approx 1/T$. Among dominating three phonon acoustic branches (LA, TA, ZA), the numerical contribution of ZA acoustic mode in pure graphene samples is only up to 10% of its total thermal conductivity, but in graphene samples with boundaries and impurities the contribution of ZA acoustic mode to graphene thermal conductivity is dominating.

Graphene electrical conductivity decreases with temperature for all electron scattering mechanisms accounted (on charged and neutral impurities, and on phonons). The numerical calculations and graphs are hereby presented separately in low- and high-temperature regions, due to different temperature dependencies of chemical potentials and relaxation times in these regions.

Our theoretical results are in fairly good agreement with the available literature theoretical and experimental data.