

TEMPERATURE DEPENDENCE OF THERMAL CONDUCTIVITY OF POLYCRYSTALLINE GRAPHENE

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INTRODUCTION

The temperature dependence of thermal conductivity of polycrystalline graphene is analyzed, as polycrystalline graphene is more often experimentally realized than monocrystalline one. Phonons make the greatest contribution to the thermal conductivity of graphene (while its electronic component is negligibly small). The contribution of all three branches of acoustic phonons to thermal conductivity is calculated taking into account phonon scattering at the sample boundaries, point defects, grain boundaries, and phonon-phonon scattering. The temperature dependence of thermal conductivity is calculated numerically in approximation of the relaxation time. The obtained results for the thermal conductivity of polycrystalline graphene are compared with the results for monocrystalline graphene and experimental results.

THERMAL CONDUCTIVITY OF GRAPHENE

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 direction of LA and TA oscillation modes, and has nonlinear dispersion law, $\omega_3 = v_3 q^{1.5}$.

Coefficient of phonon conductivity is determined by relation:

$$\kappa = \frac{1}{4\pi L_z k_B T^2} \sum_s \int_{\omega_{\min}}^{\omega_{\max}} (\hbar \omega_s)^2 \frac{e^{\frac{\hbar \omega_s}{k_B T}}}{\left(e^{\frac{\hbar \omega_s}{k_B T}} - 1 \right)^2} \tau(\omega_s) \frac{v}{u} \omega_s d\omega_s. \quad \frac{1}{\tau} = \sum_i \frac{1}{\tau_i}$$

where L_z is the sample thickness, v is the phonon group velocity, u is the phonon phase velocity, and τ is the phonon relaxation time.

PHONON RELAXATION TIMES

At low temperatures in sufficiently pure sample the most important mechanism is scattering at boundaries of the sample. Relaxation time for this case is defined as:

$$\frac{1}{\tau_B} = \frac{1-p}{1+p} \frac{v}{L_x},$$

where p is a measure of surface roughness, and L_x is smallest dimension of sheet. In the case of elastic scattering on isotopes, the relaxation time is defined by:

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

For U-processes the relaxation time is defined as:

$$\frac{1}{\tau_u} = 2\gamma_s^2 \frac{k_B T}{Mv^2} \frac{\omega^2}{\omega_D}$$

where γ is Grüneisen constant.

In the literature it is proposed that polycrystalline boundaries can be modelled by disclinations (rotationary dislocations). The boundary grains are represented by static two-axes dipoles of the length L . The character of phonon relaxations on these dipoles depends on the ratio of the phonon wavelength λ and length L . The relaxation time on the grain boundaries is represented by:

$$\frac{1}{\tau_{GB}} = 2D^2 v^2 L^2 n_i \omega G(qL)$$

which is obtained in the approximation of the deformation potential, with

$$D = \pi\gamma(1-2\sigma)/(1-\sigma)$$

where γ is Grüneisen constant, σ is Poisson constant, v is Frank index, and n_i is the surface density of dipoles. The function $G(qL)$ is given as

$$G(qL) = J_0^2(qL) + J_1^2(qL) - J_0(qL)J_1(qL)/qL$$

where $J_0(qL)$ and $J_1(qL)$ are Bessel functions.

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

Table 1. Relevant parameters adopted in thermal calculations above.

L_z [μm]	L_x [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]	A_d	p	$\omega_{D,LA}$ [Hz]	$\omega_{D,TA}$ [Hz]	$\omega_{D,ZA}$ [Hz]	$\omega_{\min} = f(\omega, T)$	
$2 \cdot 10^{-26}$	$4.5 \cdot 10^{-4}$	0.9	$2.66 \cdot 10^{14}$	$2.38 \cdot 10^{14}$	$1.32 \cdot 10^{14}$	-	
σ	L [m]	v	n_i [m ⁻²]	D	-	-	-
0.16	$2.46 \cdot 10^{-9}$	0.2	$1.8 \cdot 10^{11}$	2.6	-	-	-

RESULTS OF NUMERICAL CALCULATIONS OF GRAPHENE THERMAL CONDUCTIVITIES

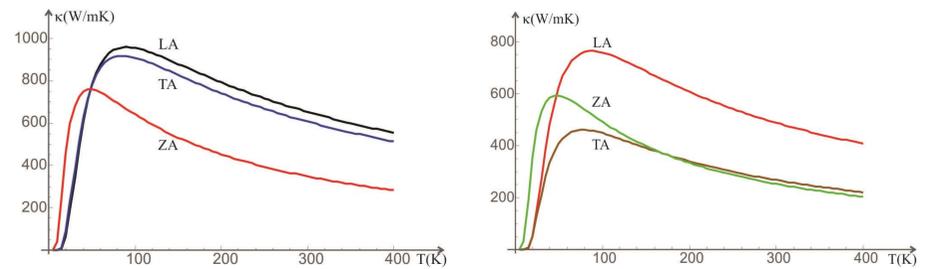


Figure 1. Contributions of particular phonon branches to thermal conductivity of monocrystalline (left) and polycrystalline graphene (right).

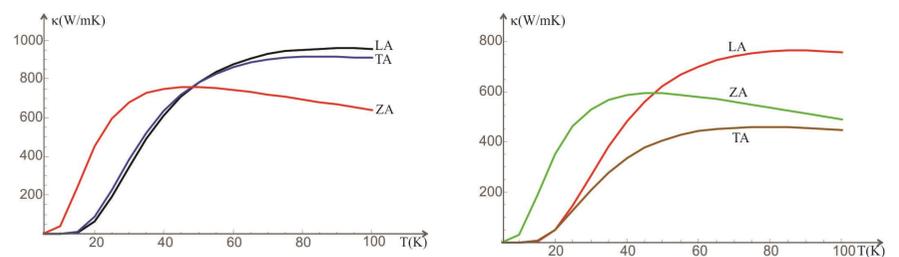


Figure 2. Contributions of particular phonon branches to thermal conductivity of monocrystalline (left) and polycrystalline graphene (right) at low-temperatures.

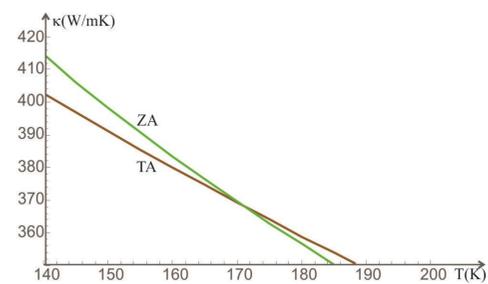


Figure 3. Thermal conductivities originating from TA and ZA phonon branches in polycrystalline graphene.

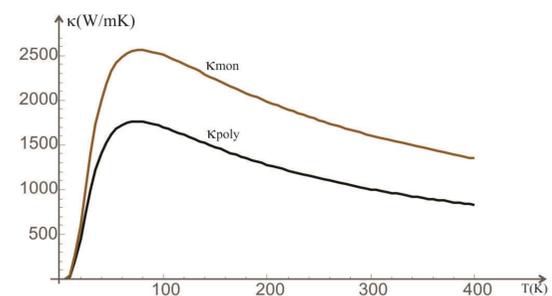


Figure 4. Total thermal conductivities in monocrystalline and polycrystalline graphenes.

CONCLUSION

Larger samples of graphene are, as a rule, of polycrystalline structure. Heat transfer in graphene, at room temperatures, is of interest due to its possible application in electronics and photonics. It has been experimentally proven that the basic contribution to the thermal conductivity of graphene is the phonon thermal conductivity, while the electronic thermal conductivity can be neglected. The coefficient of thermal conductivity in polycrystalline graphene was found by solving the Boltzmann transport equation in the relaxation time approximation. For different phonon scattering mechanisms, relaxation times as a function of temperature have been adopted from the literature, as well as the values of the corresponding parameters. Based on that, the contribution to the thermal conductivity of each individual phonon branch of acoustic phonons was calculated numerically. The results obtained in polycrystalline graphene were compared with the results obtained in monocrystalline one. It is noticeable (Fig. 1) that the contribution of the ZA phonon branch in polycrystalline graphene is higher than in monocrystalline one, especially at lower temperatures. After 50 K, the contribution of the ZA phonon branch is less than the contribution of the LA phonon branch, but greater than the contribution of the TA phonon branch. Only after 170 K, the contribution of the TA phonon branch becomes greater than the contribution of the ZA phonon branch. Also, in monocrystalline graphene, the contributions of LA and TA phonon branches are functionally the same, except that the contribution of TA phonon branch is somewhat smaller. In polycrystalline graphene, the contributions of LA and TA phonon branches differ significantly although they are functionally similar (Figs. 2 and 3). Finally, the total thermal conductivity of polycrystalline graphene is about 50% lower than the thermal conductivity of monocrystalline graphene, in accordance with the experimental data (Fig. 4).