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Investigation of two-dimensional lattice thermal transport in bilayer graphene using phonon scattering mechanism

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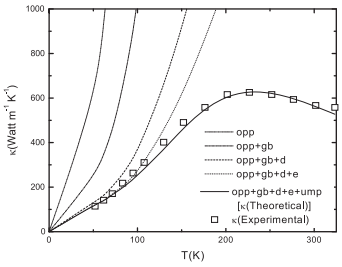
HIGHLIGHTS

- Two-dimensional lattice thermal conductivity in bilayer graphene.
- Phonon scattering mechanism incorporating the phonon-defect, -electron, -grain boundaries, -phonon umklapp and out-of-plane phonon scattering process.
- Temperature and frequency dependence of relaxation rates and their effect on thermal conductivity.

GRAPHICAL ABSTRACT

Two-dimensional lattice thermal conductivity of bilayer grapheme.



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ABSTRACT

Two-dimensional lattice thermal transport in bilayer graphene is investigated using phonon scattering mechanism. In the plane layer of carbon atoms the thermal conductivity (κ) is demonstrated by incorporating the phonon–defect, phonon–electron, phonon–grain boundaries, phonon–phonon umklapp scatterings and out-of-plane phonon scattering process in the model Hamiltonian. A typical $T^{1.5}$ dependence of thermal conductivity observed at low temperatures (lower than 150 K) is the resultant of various operating phonon scattering mechanisms. Above room temperatures, the thermal conductivity decreases and follows almost T^{-2} dependence which is an artifact of the dominant Umklapp phonon scattering at higher temperatures. The phonon peak appear at around 225 K is due to the competition between the increase in the phonon population and decrease in phonon mean free path due to umklapp phonon scattering with increasing temperature. The results obtained from present model are in good agreement with the available experimental data and reflect the two-dimensional nature of phonon transport in graphene which is dominated by phonon scatterings.

1. Introduction

Graphene that consists of only one plain layer of carbon atoms arranged in a honeycomb lattice [1] has attracted tremendous interest for both fundamental studies and applications in high-speed electronic devices because of its extremely high carrier mobility [2–4], outstanding thermal transport property [5–7] and Seebeck coefficient [8]. The extremity high observed thermal conductivity (κ) [5–7] of graphene establishes it as an excellent material for thermal management. With the continuously decreasing size of electronic devices and increasing dissipation power density in downscaled circuits, one observes a tremendous growth of materials that can conduct heat efficiently. Besides carbon nanotubes (CNTs) those are known to have very high thermal conductivity κ with the experimentally determined room temperature value of thermal conductivity ranging from $\approx$1500 to 6000 $Wm^{-1}K^{-1}$ for different structures of CNTs [9–11], the single plane layer of carbon atoms (*i.e.* graphene) are another structures who demonstrates an extremely high thermal conductivity. Despite of high observed thermal conductivity and potential

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applications, no clear theoretical explanation is yet available which motivates us for theoretical investigations on thermal conductivity of graphene. In present work we have demonstrated a theoretical model for investigation of Two-dimensional lattice thermal transport in graphene using phonon scattering mechanism.

The thermal conductivity (κ) of two bilayer graphene samples each suspended between two microresistance thermometers was measured to be 620 ± 80 and $560 \pm 70\ \mathrm{Wm^{-1}\,K^{-1}}$ at room temperature by Michael et al. [5]. A typical $T^{1.5}$ dependence of the temperature dependant thermal conductivity is observed at temperatures lower than 125 K. Alexander et al. have reported the experimental investigation of thermal conduction in a suspended single-layer graphene performed with the help of confocal micro-Raman spectroscopy. The room temperature values of the thermal conductivity of up to $5300\ \mathrm{W\,m^{-1}\,K^{-1}}$ were extracted for a single-layer graphene from the dependence of the Raman G peak frequency on the excitation laser power [6]. A Raman measurement of a large graphene flake suspended over a 44 μm diameter hole was reported by Cai et al. [12]. By measuring the laser heating and monitoring the Raman G peak, the room-temperature thermal conductivity of $(370+650/-320)\ \mathrm{W\,m^{-1}\,K^{-1}}$ is obtained for the supported graphene. However, the thermal conductivity of the suspended graphene exceeds $(2500+1100/-1050)\ \mathrm{W\,m^{-1}\,K^{-1}}$ near room temperature 350 K, decreases on increase in temperature, and becomes $(1400+500/-480)\ \mathrm{W\,m^{-1}\,K^{-1}}$ at about 500 K [12].

Several investigations [7,13,14] indicate that the phonons are the sole carrier of heat, leading to such a high thermal conductivity observed in graphene. The main contribution to the thermal conductivity of graphene comes from the in-plane longitudinal acoustic (LA) and transverse acoustic (TA) phonons, which are characterized by the high phonon group velocities and small Gruneisen parameters leading to the large phonon mean free path [7]. Whereas the contribution to the thermal conductivity from the out-of-plane phonon modes (generally called as flexural phonon) is small because of the large Gruneisen parameter and small group velocity [13]. The role of flexural phonon on diffusive thermal conductivity of suspended rectangular single layer graphene sheet is analyzed by Verma et al. [14]. A quadratic dependence of the out-of-plane phonon frequency (flexural phonons), on the phonon wave vector has been taken into account to analyze the behavior of thermal conductivity at lower temperatures. It is clearly demonstrated that thermal conductivity follows a $T^{1.5}$ at lower temperatures and T^{-2} at higher temperatures due to the edge roughness scattering and the umklapp scattering respectively [14].

The lattice thermal conductivity of freestanding graphene has been predicted to be dominated by contributions from the out-of-plane acoustic (ZA) phonons, that is, the flexural modes [15]. The damping of the ZA phonons by a silicon dioxide support limits the thermal conductivity of supported single-layer graphene which has been reported to $\sim 600\ \mathrm{W\,m^{-1}\,K^{-1}}$ [16] considerably lower than the basal-plane values of highly ordered pyrolytic graphite [17,18]. The low-temperature thermal conductivity values of the bilayer samples are reported [5] slightly higher than those reported for the single-layer graphene samples supported on SiO_2 [16], and reported a few times higher than the few-layer graphene samples encased in SiO_2 [19]. Furthermore, it is reported that the thermal conductivity values of the few-layer graphene samples increases with the layer thickness which is attributed as weaker effect of interlayer coupling than graphene-medium interaction on phonon transport [19]. This difference is attributed as a weaker effect of the interlayer coupling than that of interaction with the polymer residue that is present only on the top surface of the bilayer graphene, or to weaker interaction between the polymer residue and the bilayer sample than that between the single-layer samples and the SiO_2 support.

The theoretical calculations [15,16] revealed that, in bilayer graphene the interlayer coupling break the reflection symmetry, reducing the ZA contribution and resulting in a suppressed thermal conductivity. However, this effect is not strong enough to suppress the thermal conductivity to a value much lower than that of the basal plane of graphite [20]. Ghosh et al. [20] has presented phonon dispersion curves for single-layer and bilayer graphene which reflects that the longitudinal acoustic (LA_2) and transverse acoustic (TA_2) phonon branches in bilayer graphene have very small slope than in single layer graphene, which translates to low phonon group velocity, affecting the thermal conductivity. It is of considerable interest to study the thermal conductivity of bilayer graphene in order to analyze the effect of interlayer interaction on phonon scattering machenism and thermal conductivity.

Looking at above experimental observations and theoretical interpretations, it seems that the propagation of phonons in an ordered single plane layer of carbon atoms, play an important role in the heat transmission in the graphene and responsible for high observed thermal conductivity. Despite of several investigations the exact role of phonons, the phonon scattering mechanism in graphene, the phonon relaxation rates, dependence of phonon relaxation rates on temperatures and frequency and their effect on thermal conductivity is not clearly understood. Several fundamental questions on phonon scattering mechanism remains unanswered, which motivates us to carry out a systematic theoretical investigation of thermal conductivity of graphene using phonon scattering mechanism. In present paper we have incorporated the scattering of phonons with defects, grain boundaries, electrons, out-of-plane phonon and umklapp phonon scatterings in the model Hamiltonian within the relaxation time approximation to demonstrate the anomalies observed in thermal conductivity of graphene. We have also quantitatively estimated the strength of various phonon scattering rates, based on transport parameters which appear in the theoretical model. We find that the thermal conductivity can be well explained using the Debye like model based on phonon scattering mechanism. The detailed model formulism has been presented in the next section followed by results and discussion and conclusions.

2. The model

Looking to the fact that the phonons play an important role in describing the thermal conductivity of graphene [7,13,14], we follow a simple model where the phonons are described in the Debye model. The use of the Debye model is reasonable since the temperature region of interest lies around the Debye temperature. As the simplest approximation to the problem at hand, the isotropic Debye model approach can be used to derive qualitative results, as demonstrated later.

We begin with a model Hamiltonian that follows [21,22]

$$
\begin{aligned}
H = {} & \sum_k \varepsilon_k a_k^+ a_k + \sum_q \omega_q b_q^+ b_q + \sum_{i=1,2} \varepsilon_i c_i^+ c_i \\
& + \sum_{k_1,k_2} \varphi(k_1,k_2) a_{k_1}^+ a_{k_2} + D_p \sum_{k,q} q \left[\frac{\hbar}{2\rho\omega_q}\right]^{1/2} a_{k+q}^+ a_k (b_k + b_{-k}^+) \\
& + \frac{R}{2N} \sum_{q_1,q_2} e^{i(q_1+q_2)r_i} \left[\frac{\hbar\omega_{q_1}\hbar\omega_{q_2}}{4}\right]^{1/2} (b_{q_1} - b_{-q_1}^+)(b_{q_2} - b_{-q_2}^+) \\
& + \sum_{q;i,j=1,2} \gamma_{ij} q \left[\frac{\hbar}{2\rho\omega_q}\right]^{1/2} c_i^+ c_j (b_q + b_{-q}^+) + H_{ph-ph}. \qquad (1)
\end{aligned}
$$

here, initial three terms represent electrons, in plane phonons, and out-of-plane phonon excitation. The fourth and fifth terms represent the carrier–defect interactions and carrier–phonon interactions, respectively. The sixth and seventh terms denote the phonon–defect interactions and out-of-plane phonon