3.1. Introduction

As discussed in the Chapter 2, thermopower $S$ is one of the important transport properties which finds application in power generation and small scale cooling. Besides, it provides information of electronic structure and scattering processes in the transport phenomena. The efficiency of a thermoelectric device is determined by its figure of merit $ZT (\sim S^2)$. In order to examine the applicability of graphene from the point of view of thermoelectric devices it is necessary to understand the behavior of its thermopower. During the last five years investigations of thermopower in graphene are carried out [3.1].

There exist theoretical [3.2-3.10] and experimental [3.11-3.14] studies of thermopower in monolayer graphene, which is briefly reviewed in the following. Peres and co-workers [3.2, 3.3], using the semi-classical Boltzmann equation, gave the phenomenological theory of transport in MLG. They studied carrier concentration dependence of $S^d$ considering the scattering due to impurities in the substrates and mid gap states, arising due to vacancies and other defects. It was pointed out that Boltzmann approach breaks down at charge neutrality point (CNP). Using the linear response theory and considering strong impurity scattering, Löfwander and Fogelström [3.4] have shown, at low $T$, $S^d$ to be linear in $T$ with slope proportional to the inverse of impurity density and their strength. Kubakaddi has developed [3.6, 3.7] theory of diffusion thermopower, using the Boltzmann transport equation approach, and obtained the equation for $S^d$ which is similar to the one in conventional 2DEG [3.15]. Considering the electron scattering due to ionized impurities, vacancies and acoustic phonons, it was studied as a function of $T$ and $n_s$ [3.6]. Kubakaddi [3.7] obtained Mott formula for $S^d$ and predicted the $n_s^{-1/2} (n_s$ the 2D electron concentration) and linear $T$ dependence. Besides, $S^d$ is shown to be dominant over the phonon-drag contribution for about $T < 2K$ and for $T > 10 K$.

The pioneering measurements of thermopower were made on monolayer graphene samples, mechanically exfoliated on SiO$_2$/Si substrates, in the temperature range $10 < T < 300$ K [3.11-3.14]. To measure thermopower, in a typical experiment, a controlled temperature difference $\Delta T$ is maintained along the plane of the sample and resulting thermoelectric voltage $\Delta V$ is measured to give $S = \Delta V/\Delta T$. Carrier concentration is controlled by gate voltage $V_g$. Measurements of Zuev et al [3.11] for $10 < T < 300$ K, for the samples with mobilities in the range $1-7 \times 10^3$ cm$^2$/Vs, show a
linear $T$ dependence for two $V_g$ far away from the charge neutrality point, indicating the diffusive thermopower. Change in sign of $S$ is observed across CNP as the majority carrier density is changed from electrons to holes. Experimental results of Wei et al [3.12] show $S \sim n_s^{-1/2}$, agreeing with the Mott formula prediction [3.7], which is a direct evidence of linear dispersion of Dirac Fermion in graphene, in contrast with $n_s^{-1}$ in conventional 2D systems [3.15]. $S$, for electrons, remains linear in low $T$ region and remains nonlinear in high $T$ region. The measured $S$ is found to be $\sim 100 \mu$V/K at room temperature. Checkelsky and Ong [3.13] find change in sign of $S$ with $V_g$ across CNP and nearly linear in $T$.

Hwang et al [3.8] developed the theory of $S^d$, using Mott formula, explained the experimental data taking the energy dependence of the relaxation times due to random charged impurities, short-range disorder and acoustic phonons. Mott formula is found to apply well for $T \leq 0.2T_F$ ($T_F$ is the Fermi temperature) and at higher $T$, $S$ is shown to be independent of $T$. The calculated $S^d$ behaves as $n_s^{-1}$, agreeing with the predictions of Kubakaddi [3.7] and experimental results [3.11-3.13]. Mott formula is shown to fail in low $n_s$ limit where formation of electron-hole puddles takes place. A quadratic $T$ dependence of $S^d$ is predicted due to screening of charged impurity scattering. Following the balance equation approach, $S^d$ is calculated [3.9] considering the electron scattering by impurities and phonons and analyze, taking account of phonon-drag contribution, the experimental results of Zuev et al [3.11]. Calculations of Bao et al [3.9] include both $S^e$ and $S^d$ and obtain good agreement with the experimental data. It is shown that $S^e$ is dominant for $T < 10$ K, agreeing with the predications of Kubakaddi [3.7] and $S^d$ dominating in the high $T$ region. Attempt is also made to fit the data of Zuev et al [3.11] by considering the electron scattering due to impurities, vacancies, surface roughness, acoustic phonons and optical phonons [3.10].

Later, different groups [3.16-3.18] have reported measurements of thermopower and its carrier mobility dependence. The data of Wu et al [3.14], in samples with relatively high mobility ($\sim 2x10^4$ cm$^2$/Vs) grown epitaxially on SiC substrates, away from CNP, is shown to fit to $AT+B{T^2}$, where $A$ and $B$ are $T$ independent constants. As predicted by Hwang et al [3.8], this quadratic $T$ dependence seems to arise due to temperature dependent screening effects.

In bilayer graphene, there are only few reports of study of thermopower [3.19-3.22]. Hao and Lee [3.19], using Kubo formula, have theoretically studied diffusion
thermopower in gapped and relatively clean BLG, treating impurity scattering in self consistent Born approximation. Enhancement of thermopower is predicted with the introduction of gap. Theoretically phonon-drag thermopower has been investigated (Chapter 4) [3.20]. Thermopower has been studied experimentally by Nam et al [3.21] and Wang et al [3.22] for temperatures $T=15-300$ K. Their observations largely show nearly linear $T$ dependence and inverse $n_s$ dependence for larger electron concentrations.

Although there exist a few theoretical studies of $S^d$ in BLG [3.20, 3.21], its sensitivity to the scattering mechanisms has not been addressed as being done in monolayer graphene [3.6-3.10]. In this study we report the calculations of $S^d$ due to different scattering mechanisms using Boltzmann transport theory for suspended and supported BLG and compare with the existing experimental observations. In the process, we address relaxation time and mobility also. The possible scattering mechanisms considered are due to acoustic phonons (AP) via deformation potential coupling [3.23, 3.24], surface polar phonons (SPP) [3.25] and impurity (charged impurity (CI) and short-range disorder (SD)) [3.26]. Since the energy of optical phonons is very high to cause any scattering, their role in governing the transport properties up to 300 K is ignored [3.27].

### 3.2. Theory of diffusion thermopower in bilayer graphene

BLG is a two weakly coupled layers of graphene separated by interlayer spacing $c = 0.34$ nm. As discussed in section 1.4.2, the weak interaction between the two layers leads to parabolic dispersion relation, in the low energy regime.

$$E_k = \frac{\hbar^2 k^2}{2m} [3.28, 3.29],$$

where $s = +1$ and $-1$ for conduction and valence band, respectively, $k$ is the 2D electron wave vector and $m$ is the effective mass of the electron with density of states $D(E_k)=\frac{2m}{\pi \hbar^2}$.

Following the theory given in the section 2.2.1, employing the Boltzmann transport formalism in the relaxation time approximation, $S^d$ in this system is shown to be

$$S^d = -L^d/\sigma = (1/|e|T) K_1 K_0^{-1}. \quad \text{(3.2.1)}$$

$K_0$ and $K_1$ appearing in this equation are given by Eqn. (2.2.3). These integrals are with respect to surface element $d^2k = (m/\hbar^2) dE_k d\psi$, for BLG with quadratic dispersion relation, where $\psi$ is the polar angle of $k$. Then expression for $K_n$ becomes...
\[ K_n = \frac{8 \tau s}{4 \pi^2} \frac{m}{\hbar^2} \int \tau(E_k) \nu_k \nu_k (E_k - E_f)^n \left( \frac{\partial f^0_k}{\partial E_k} \right) dE_k d\psi. \]  

(3.2.2)

Hence, \( K_0 \) and \( K_1 \) are shown to be

\[ K_0 = \frac{n_s}{m} \int \tau(E_k) E_k (E_k - E_f) \left( \frac{\partial f^0_k}{\partial E_k} \right) dE_k \]

\[ = \frac{n_s}{m} \langle \tau(E_k) \rangle - E_f \langle \tau(E_k) \rangle \]

and

\[ K_1 = \frac{n_s}{m} \int E_k \left( \frac{\partial f^0_k}{\partial E_k} \right) dE_k \]

\[ = \frac{n_s}{m} \tau(E_k) \]

(3.2.3)

where

\[ n_s = \frac{8 \tau s m}{2 \pi^2} \int f^0_k(E_k) dE_k. \]

(3.2.4)

(3.2.5)

\[ \langle \tau(E_k) \rangle = \frac{1}{\int E_k (E_k - E_f) \left( \frac{\partial f^0_k}{\partial E_k} \right) dE_k} \]

(3.2.6)

and

\[ E_f = k_B T \ln[\exp(\psi n_s / 2mk_B T) - 1] \]

(3.2.7)

is the Fermi energy.

Then, Eqn. (3.2.1) reduces to Eqn. (2.2.10) i.e.

\[ S^d = -(1/|e|T)\langle \{ \tau(E_k) E_k \} / \langle \tau(E_k) \rangle \rangle - E_f. \]

(3.2.8)

In degenerate case, i.e. for large \( n_s \) and low temperatures, expressing \( \tau(E_k) \sim E_k^p \), the above equation gives Mott formula in BLG [3.20]

\[ S^d = -[\pi^2 k_B^2 T (p + 1)]/3 |e| E_f. \]

(3.2.9)

Hence, we predict \( S^d \sim T \) and \( n_s^{-1} \), and it is consistent with the predictions in conventional 2DEG [3.15].

In the following we discuss the relaxation times due various scattering mechanisms that possibly limit the transport in a BLG.

### 3.2.1 Relaxation time due to acoustic phonon scattering

In the following we derive the electron relaxation time \( \tau(E_k) \) due to acoustic phonons, following the basic formalism given in Refs. [3.23, 3.30], which will be valid for a large range of temperature and carrier concentration. Assuming that the
Diffusion thermopower in a bilayer graphene

electrons interact with 2D in-plane longitudinal acoustic phonons of frequency \( \omega_q \) and wave vector \( \mathbf{q} \) via deformation potential coupling, the basic expression for \( \tau(E_k) \) is found to be

\[
1/\tau_{AP}(E_k) = \sum_{k'}(1-\cos\theta_{kk'})P(k,k')|1-f^0(E_{k'})/[1-f^0(E_k)],
\] (3.2.10)

where

\[
P(k,k')=(2\pi/h)|M(k,k')|^2 \Delta(E_k,E_{k'})
\] (3.2.11)
is the transition probability, with

\[
\Delta(E_k,E_{k'})=\{N_q^0\delta(E_k-E_{k'}+h\omega_q)+(N_q^0+1)\delta(E_k-E_{k'}-h\omega_q)\}.
\] (3.2.12)
The electron-acoustic phonon matrix element (Eqn. (1.5.2)) can be written as

\[
|M(k,k')|^2 = |C(q)|^2 g(\theta_{kk'}),
\] (3.2.13)

with

\[
|C(q)|^2 = (hD^2q/2\rho A\tau_s).
\] (3.2.14)

\( E_k \) and \( E_{k'} \) are, respectively, the energies of the electron in the initial (\( k \)) and final (\( k' \)) states, \( N_q^0=\exp(h\omega_q/k_BT)-1 \) is the acoustic phonon distribution function and \( g(\theta_{kk'}) \) is the form factor (Eqn. (1.5.5)) arising due to the chiral nature of carriers in BLG.

Converting summation over \( k' \) to integration

\[
\sum_{k'} \rightarrow \frac{A}{4\pi^2} \int d2k' = \frac{A}{4\pi^2} \int d\theta_{kk'} = \frac{A}{4\pi^2} \int dE_{k'} \frac{2\pi}{h^2} d\theta_{kk'},
\] (3.2.15)

Eqn. (3.2.10) becomes

\[
\frac{1}{\tau_{AP}(E_k)} = \frac{A}{4\pi^2} \frac{m}{h^2} \int dE_{k'} \frac{2\pi}{h^2} \int d\theta_{kk'} (1-\cos\theta_{kk'}) P(k,k') \frac{|1-f^0(E_{k'})|}{[1-f^0(E_k)]} \times |C(q)|^2 g(\theta_{kk'}) \Delta(E_k,E_{k'}).
\] (3.2.16)

Carrying out the integration over \( E_{k'} \) using the Dirac \( \delta \)-function, we get

\[
\frac{1}{\tau_{AP}(E_k)} = \frac{mD^2}{4\pi\hbar^2 v_s} \frac{\pi}{2} \int d\theta_{kk'} (1-\cos\theta_{kk'}) \frac{(1+\cos2\theta_{kk'})}{2} \frac{q}{[1-f^0(E_k)]} \times \{N_q^0[1-f^0(E_k+h\omega_q)]+(N_q^0+1)[1-f^0(E_k-h\omega_q)]\}.
\] (3.2.17)

For quasi-elastic scattering \( q=|\mathbf{k} - \mathbf{k'}|=2k\sin(\theta_{kk'}/2) \) and \( (1-\cos \theta_{kk'})=2(2/k)^2 \). Then \( g(\theta_{kk'}) \) can be expressed in terms of \( q \) and \( k \) and it is given by

\[
g(\theta_{kk'}) = g(q,k) = [1-2(2/k)^2]^2.
\] (3.2.18)

Using the above expression, Eqn. (3.2.17) becomes
\[
\frac{1}{\tau_{AP}(E_k)} = \frac{4mD^2}{\pi \hbar^3 v_s} \int_0^{2k} \frac{(q/2k)^3}{\sqrt{1-(q/2k)^2}} g(q,k) \times \{ N_q^0[1-f^0(E_k+h\omega_q)]+(N_q^0+1)[1-f^0(E_k-h\omega_q)] \} dq. \tag{3.2.19}
\]

### 3.2.2. Relaxation time due to surface polar phonon scattering

As discussed in section (1.5.3) the 2D electrons are assumed to interact with the 2D surface polar phonons of the substrate via Fröhlich interaction. The relaxation time due to surface polar phonons has been given by Li et al [3.25]

\[
\frac{1}{\tau_{SPP}(E_k)} = \frac{2\pi}{\hbar} \sum_q \frac{e^{2F^2q^2}}{|e(q)|^2} \left[ \frac{e^{-2qd}+e^{-2q(d+c)}}{2q} \right] \left\{ N_q^0 + \frac{1}{2} \pm \frac{1}{2} \right\} \\
\times g(\theta_{kk'}) \delta(E_{k'}-E_k \pm h\omega_q), \tag{3.2.20}
\]

where \( F^2 \) is the Fröhlich coupling constant as given in (Eqn. 1.5.14), \( d \) is the distance between the graphene and substrate, \( h\omega_s \) is the energy of SPPs and \( N_q^0=\left[\exp(h\omega_s/k_BT)-1\right]^{-1} \) is the SPPs distribution function. Converting summation over \( q \) to integration and carrying out integration with respect to \( q \), following Leburton [3.31], we obtain

\[
\frac{1}{\tau_{SPP}(E_k)} = \frac{2e^2F^2mA}{\pi \hbar^3} \left\{ \left( N_q^0 + 1 \right) \int_0^{\varphi_{\text{max}}} \frac{1}{|e(q^a_{\pm})|^2} \left[ \frac{e^{-2q^a_{\pm}d}+e^{-2q^a_{\pm}(d+c)}}{2} \right] g(q^a_{\pm}) dq^a_{\pm} \right\} \\
+ \left\{ N_q^0 \int_0^{\varphi_{\text{max}}} \frac{1}{|e(q^a_{\pm})|^2} \left[ \frac{e^{-2q^a_{\pm}d}+e^{-2q^a_{\pm}(d+c)}}{2} \right] g(q^a_{\pm}) dq^a_{\pm} \right\}, \tag{3.2.21}
\]

where

\[
\varphi_{\text{max}} = a \cos \sqrt{\hbar\omega_s/E_k}, \quad q^a_{\pm} = k \cos \varphi \pm \sqrt{k^2 \cos^2 \varphi - (2m\hbar\omega_s)/\hbar^2}, \quad q^a_{\pm} = -k \cos \varphi \pm \sqrt{k^2 \cos^2 \varphi + (2m\hbar\omega_s)/\hbar^2},
\]

\[
g(q^a_{\pm}) = \frac{\sqrt{E_k-E_{q^a_{\pm}}} \cos \varphi}{\sqrt{E_k-h\omega_s}}, \quad g(q^a_{\pm}) = \frac{\sqrt{E_k+E_{q^a_{\pm}}} \cos \varphi}{\sqrt{E_k+h\omega_s}} \quad \text{and} \quad E_{q^a_{\pm}} = \frac{\hbar^2}{2m} (q^a_{\pm})^2.
\]
3.2.3. Relaxation time due to impurity scattering

The relaxation time due to impurity is discussed by Das Sarma et al [3.26] and it is given by

$$\frac{1}{\tau_{\text{IMP}}(E_k)} = \frac{2m_0}{\hbar} \int \frac{d^2k'}{(2\pi)^2} |M(k,k')|^2 (1 - \cos \theta_{kk'}) g(\theta_{kk'}) \delta(E_k - E_{k'}).$$  \hspace{1cm} (3.2.22)

where $n_0$ is the impurity concentration. This is elastic scattering. Generally, impurities are taken to be ionized (charged) impurities and neutral impurities. The matrix element $|M(k,k')|^2$ due to charged impurity scattering is given by Eqn. (1.5.18). After carrying out integration with respect to $E_{k'}$ and expressing $\theta_{kk'}$ in terms of $q$, the explicit expression for $\tau_{\text{CI}}(E_k)$ due to charged impurity scattering is given by

$$\frac{1}{\tau_{\text{CI}}(E_k)} = \frac{2m_0}{\pi \hbar^3} \int_0^{2k} \frac{q}{2k} \frac{g(q,k)}{k[1-(q/2k)^2]^{1/2}(q+q_{TF})^2} dq,$$ \hspace{1cm} (3.2.23)

for charged impurity concentration $n_i$.

For neutral impurity scattering, which is a short-range disorder, the corresponding matrix element is given by Eqn. (1.5.22). The relaxation time $\tau_{\text{SD}}(E_k)$ due to short-range disorder, with concentration $n_d$, and potential $V_0$ is given by

$$\frac{1}{\tau_{\text{SD}}(E_k)} = \frac{n_d V_0^2 m}{\pi \hbar^3} \int_0^{2k} \frac{q}{2k} \frac{g(q,k)}{k[1-(q/2k)^2]^{1/2}} dq.$$ \hspace{1cm} (3.2.24)

For the later use, we denote $g(q,k)=g(E_k)$ appearing in all the above equations as it turns out to be function of $E_k$ after integrating over $q$.

Resultant relaxation time is obtained by Matthiessen’s rule

$$[1/\tau(E_k)] = [1/\tau(E_k)]_{\text{AP}} + [1/\tau(E_k)]_{\text{SPP}} + [1/\tau(E_k)]_{\text{CI}} + [1/\tau(E_k)]_{\text{SD}}.$$ \hspace{1cm} (3.2.25)

$S_d$ due to each of the scattering mechanisms can be studied using the respective relaxation times (Eqns. 3.2.19, 3.2.21, 3.2.23 and 3.2.24) in Eqn. (3.2.8) and the resultant $S_d$ is obtained using Eqn. (3.2.25).

3.3. Results and discussion

The material parameters used in the present calculations are listed in Table 3.1. We chose the value of $D = 20$ eV [3.7, 3.20, 3.23, 3.32, 3.33] for illustration, which is closure to the values largely used in the literature.
Table 3.1: Material parameters of BLG.

<table>
<thead>
<tr>
<th>Material parameters</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m/m_0$</td>
<td>0.033 [3.29]</td>
</tr>
<tr>
<td>$\rho$</td>
<td>$7.6 \times 10^{-8}$ g/cm$^2$ [3.29]</td>
</tr>
<tr>
<td>$v_f$</td>
<td>$1 \times 10^8$ cm/s [3.23]</td>
</tr>
<tr>
<td>$v_s$</td>
<td>$2 \times 10^6$ cm/s [3.23]</td>
</tr>
<tr>
<td>$d$</td>
<td>0.4 nm [3.25]</td>
</tr>
<tr>
<td>$c$</td>
<td>0.34 nm [3.25]</td>
</tr>
</tbody>
</table>

Table 3.2: Surface polar phonon energy and dielectric constant for different dielectric substrates[3.27].

<table>
<thead>
<tr>
<th></th>
<th>SiO$_2$</th>
<th>SiC</th>
<th>HfO$_2$</th>
<th>hBN</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_s$</td>
<td>3.9</td>
<td>9.7</td>
<td>22.0</td>
<td>5.09</td>
</tr>
<tr>
<td>$\varepsilon_{\infty}$</td>
<td>2.5</td>
<td>6.5</td>
<td>5.03</td>
<td>4.10</td>
</tr>
<tr>
<td>$\hbar \omega_{s01}$ (meV)</td>
<td>59.98</td>
<td>116.0</td>
<td>19.42</td>
<td>101.7</td>
</tr>
<tr>
<td>$\hbar \omega_{s02}$ (meV)</td>
<td>146.51</td>
<td>167.58</td>
<td>52.87</td>
<td>195.7</td>
</tr>
</tbody>
</table>

3.3.1. Supported bilayer graphene

First we illustrate relaxation times due to various scattering mechanisms and $S^d$ for a supported BLG on SiO$_2$ substrate. The parameters corresponding to SiO$_2$ substrate are listed in Table 3.2. There is a range of values being used in the literature for $n_i$ and $n_dV_0^2$ [3.26]. We chose reasonable values of $n_i=1 \times 10^{11}$ cm$^{-2}$ and $n_dV_0^2 = 2.0$ (eVÅ)$^2$ [3.26] with a typical carrier concentration of the order $10^{12}$ cm$^{-2}$, for illustration.

As mentioned in section 2.2.1, $S^d$ is sensitive to the energy dependence of the relaxation time. We first present $\tau(E_k)$ dependence on $E_k$ due to all the mechanisms considered above.
Figure 3.1(a). Inverse relaxation time $1/\tau_{AP}(E_k)$ due to acoustic phonons vs normalized electron energy $E_k/E_f$ at different temperatures. Inset is to show the effect of chiral property by taking the ratio of $\tau_{AP}(E_f)$ with that due to setting $g(E_k)=1$.

In Figure 3.1(a), $1/\tau_{AP}(E_k)$ is shown as a function of $E_k$ for $T = 4.2$, 10 and 77 K for $n_s=1x10^{12}$ cm$^2$. In the Bloch-Grüneisen (BG) regime ($q \ll 2k_f$, where $k_f = (\pi n_s)^{1/2}$ is the Fermi wave vector), $1/\tau_{AP}(E_k)$ increases with the increasing $E_k$ with a characteristic dip in a narrow region around $E_k=E_f$. The dip arises due to the statistical occupation factor, which may be defined as a composite Fermi-Boson distribution function, $\{N_q^0[1-f^0(E_k+h\omega_q)]+(N_q^0+1)[1-f^0(E_k-h\omega_q)]\}$ in Eqn. (3.2.19). The depth of the dip increases with the decreasing temperature. For $g(E_k)=1$, a case without chiral property of the electrons, the depth of the dip is smaller and inverse relaxation time is larger. This dip disappears at higher temperatures, that is in the equipartition regime ($h\omega_q<<k_B T$). This is similar to the behaviour observed in a conventional 2DEG in GaAs HJs [3.30]. $\tau_{AP}(E_k)$ is found to be almost independent of energy at higher $T$. In the inset of Figure 3.1(a) we have shown the ratio $\tau_{AP}(E_k)/[\tau_{AP}(E_f)]_{g(E_k)=1}$ at $E_k=E_f$ as a function of temperature to bring out the effect of chiral nature of electrons. The ratio shows a maximum at 10 and 20 K, respectively, for $n_s=1x10^{12}$ and $5x10^{12}$ cm$^2$, then decreases and becomes constant at higher temperatures. This indicates that the chiral nature of electrons reduces the scattering rate.
Figure 3.1(b). Inverse relaxation time $1/\tau_{SPP}(E_k)$ vs $E_k/E_f$ for scattering due to surface polar phonons of SiO$_2$ substrate with two energies 59.98 and 146.51 meV at $T=300$ K.

Figure 3.1(c). Inverse relaxation time $1/\tau(E_k)$ vs $E_k/E_f$ for scattering due to charged impurity and short-range disorder for SiO$_2$ substrate.

Behaviour of $1/\tau_{SPP}(E_k)$ is shown (Figure 3.1(b)) for SPPs of two energies at $T=300$ K. The sudden increase in $1/\tau_{SPP}(E_k)$ is attributed to the emission of SPP when the electron energy is equal to that of SPP. The contribution from the larger energy (146.51 meV) SPP is negligible and is ignored in our later calculations. In Figure 3.1(c) $1/\tau(E_k)$ vs $E_k$ is shown for scattering due to charged impurity and short-range
disorder. $1/\tau_{CI}(E_k)$ decreases with the increasing $E_k$ where as $1/\tau_{SDI}(E_k)$ is almost independent of $E_k$. It is to be noted that the impurity scattering rate is independent of temperature unless the temperature dependent screening is considered. From the Figures 3.1 (a-c) we observe that the short-range disorder is the dominant scattering mechanism for the chosen parameters. $1/\tau_{SPP}(E_k)$ illustrated in Figure 3.1(b) for 300 K is much smaller compared to other mechanisms. It will be almost negligible at lower $T$.

![Figure 3.1(d)](image)

**Figure 3.1(d).** Scattering parameter $p$ vs electron concentration $n_s$ for different scattering mechanisms at $T = 4.2$ K with SiO$_2$ substrate.

The scattering parameter $p = [(E_k/\tau(E_k)) \{d\tau(E_k)/dE_k\}]_{E_k=E_f}$ due to of each of these mechanisms is shown in Figure 3.1(d), for illustration, at $T = 4.2$ K. For short-range disorder scattering $p$ is almost zero for the range of $n_s$ considered. It is interesting to note that the $p$ due to resultant relaxation time is found to be closer to that of short-range disorder indicating the dominancy of this scattering. Low $T$ value of $p$ can be used to evaluate $S^d$ from the Mott formula (Eqn. 2.2.12) for each mechanism and all the mechanisms combined. It is to be noted that for AP scattering $p$ is almost zero in the higher $T$ region as can be seen in Figure 3.1(a).

In order to understand the relative importance of various scattering mechanisms, we plot mobility $\mu$ (Figure 3.2), which is obtained from $\mu = e <\tau(E_k)>/m$, as a function of $T$ ($1 < T < 300$ K) for $n_s = 1 \times 10^{12}$ cm$^{-2}$. At very low $T$ ($< 10$ K), the mobility $\mu_{AP}$ due to acoustic phonons decreases very rapidly, showing $T^{-4}$ dependence.
Figure 3.2. Mobility $\mu$ vs temperature $T$ for $n_s = 1 \times 10^{12}$ cm$^{-2}$. Inset shows $\mu_{\text{ap}}T^{-1}$ vs $T$ due to acoustic phonons in the BG regime.

It is a characteristic of 2D phonons agreeing with the theoretical predictions [3.23, 3.33] and experimental observation of resistivity [3.32] in MLG. However, we observe a bump in the curve around $T= 6$ K (inset of Figure 3.2), an additional interesting feature, that is attributed to the overlap integral due to chiral wave function. Such feature is not noticed in MLG [3.23, 3.33] and conventional 2DEG [3.30, 3.34]. Also, it is to be noted that $S^g$ vs $T$ has shown a kink around this region of temperature and hence validating Herring’s law $S^g_{\mu_{\text{AP}}^0} \sim T^{-1}$ over the entire BG regime [3.20]. At higher $T$, $\mu_{\text{AP}} \sim T^{-1}$ and this decrease of $\mu_{\text{AP}}$ with the increasing $T$ is also agreeing with the qualitative predictions made earlier in BLG [3.35]. $\mu_{\text{SD}}$ due to short-range disorder scattering is found to be almost independent of $T$ where as $\mu_{\text{CI}}$ increases with $T$ but very slowly. $\mu_{\text{CI}}$ and $\mu_{\text{SD}}$ contribute largely to the resultant $\mu$, with the dominance of $\mu_{\text{SD}}$, almost over the entire temperature region for the chosen parameters of $n_i$ and $n_dV_0^2$. However, significant and comparable contribution comes from acoustic phonon scattering in the higher $T$ region. The contribution due to scattering by surface polar phonons becomes slightly significant only around room temperatures. The resultant mobility is found to decrease very slowly with increase in $T$. 


Figure 3.3. Diffusion thermopower $S_d$ vs temperature $T$ for $n_s = 1 \times 10^{12}$ cm$^{-2}$ with SiO$_2$ substrate. Curve due to SD mechanism coincides with that of resultant $S_d$. Inset is $S_d$ vs $T$ due to acoustic phonons in BG regime for different $n_s$.

In Figure 3.3, we have shown the temperature dependence of $S_d$ for $n_s = 1 \times 10^{12}$ cm$^{-2}$. $S_d$ curves due to short-range disorder, acoustic phonon and the resultant are almost coinciding. Equal values of $S_d$ due to acoustic phonons and short-range disorder may be attributed to the almost same energy dependence of the respective relaxation times, except for low $T$ ($< 20$ K) in which energy dependence of acoustic phonons is different. Coincidence of $S_d$ due to short-range disorder with the resultant $S_d$ is due to the dominance of this mechanism. Although, individual value of $S_d$ due to CI is larger compared to that due to SD mechanism, its contribution in the resultant is relatively small. Similarly, contribution from AP scattering to the resultant $S_d$ is small. $S_d$ due to all the mechanisms, but SPP, shows almost linear temperature dependence below about 100 K. A similar observation is made in MLG [3.6-3.9] and conventional 2DEG [3.15] at low temperatures. In the higher $T$ region, $S_d$ shows sublinear behaviour, which is similar to the deviation from Mott formula observed in MLG [3.11, 3.13]. This may be attributed to partial non-degeneracy of electron gas as $T$ increases. The resultant $S_d$ also shows linear $T$ dependence at low $T$ and sublinear behaviour in the higher $T$ region. The $S_d$ due to acoustic phonons shows a non-monotonous behaviour around $T = 10$ K (inset of Figure 3.3). This may be attributed to the dip arising around $E_k = E_f$ in the energy dependence of the relaxation time.
The \( S^d \) vs \( T \) calculations are carried out for the sample of Nam et al [3.21], with \( n_s = 0.79, 1.51 \) and \( 3.67 \times 10^{12} \) cm\(^{-2}\), in the temperature range \( 30 < T < 250 \) K and presented in Figure 3.4. The linear \( T \) behaviour is found over the large temperature range for the samples of larger \( n_s \) showing the Mott formula being obeyed. We observe a qualitative agreement between the calculated and observed values for \( n_s = 0.79 \) and \( 1.51 \times 10^{12} \) cm\(^{-2}\). We point out that the similar \( T \) dependence of \( S^g \) is predicted in the range \( 30 < T < 70 \) K [3.20]. Due to lower dimensionality of the phonons the \( T \) dependence of \( S^g \), in this temperature regime, is almost closer to that of \( S^d \). The observed linear \( T \) dependence (\( 15 < T < 300 \) K) of \( S \) in the sample of Wang et al [3.22] for \( V_g = 80 \) V (corresponding to \( n_s = 12 \times 10^{12} \) cm\(^{-2}\)) is agreeing with our theoretical predictions [3.20]. However, the calculated values are 2-3 times smaller than the experimental values. In order to explain the experimental data more quantitatively, the sum of \( S^d \) and \( S^g \) need to be taken. At this juncture we feel, more experimental data of thermopower over the large range of temperature will be of great help to sort out the issue of relative contributions of \( S^d \) and \( S^g \).

In order to see the effect of substrates, \( S^d \) is calculated for a BLG on SiO\(_2\), SiC, hBN, HfO\(_2\) substrates using the substrate parameters given in Table 3.1 and is shown in Figure 3.5. \( S^d \) with substrates SiO\(_2\), SiC, hBN showing almost the same magnitude
Figure 3.5. Resultant diffusion thermopower (due to all the mechanisms) \( S^d \) vs temperature \( T \), for \( n_s = 1 \times 10^{12} \text{ cm}^{-2} \), for different substrates.

and \( T \) dependence where as it differs for HfO\(_2\) substrate, although increasing with \( T \).

We note that HfO\(_2\) has very high dielectric constant 22 and very low SPP energy 19.42 meV as compared to their values in other substrates [3.27].

Figure 3.6. Diffusion thermopower \( S^d \) vs electron concentration \( n_s \) at \( T = 300 \text{ K} \) with SiO\(_2\) substrate.
Figure 3.6 shows the calculations of $S^d$ as a function of $n_s = (0.5-10) \times 10^{12}$ cm$^{-2}$ at $T = 300$ K for all the four scattering mechanisms. These calculations are for SiO$_2$ substrate. All the mechanisms, but due to SPP ($S^d \sim n_s^{-0.75}$), show almost $S^d \sim n_s^{-1}$ behavior which is consistent with the case of degenerate conventional 2DEG [3.15] but differs from MLG ($S^d \sim n_s^{-1/2}$) [3.6, 3.7]. Again, as found in Figure 3.4, the $S^d$ values due to short-range disorder and acoustic phonon scattering are almost coinciding with the resultant one. $S^d$ due to short-range disorder is found to dominate over the entire $n_s$ range considered.

### 3.3.2 Suspended bilayer graphene

In the following we give similar illustrations for suspended BLG in which case $\varepsilon = 1$ and scattering due to SPP does not arise. Moreover, concentration of the charged impurity and short-range disorder will be much smaller than their values in supported BLG [3.33].

![Figure 3.7](image.png)

**Figure 3.7.** Inverse relaxation time $1/\tau(E_k)$ vs $E_k/E_f$ for scattering due to charged impurity and short-range disorder for suspended BLG.

The $\tau_{AP}(E_k)$ vs $E_k$ in suspended BLG will be same as in supported BLG (Figure 3.1 (a)) for unscreened deformation potential coupling. In Figure 3.7, $1/\tau(E_k)$
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$\text{vs } E_k$ is shown for scattering due to charged impurity and short-range disorder for reasonable parameters $n_i = 1 \times 10^{10} \text{ cm}^{-2}$, $n_d V_0^2 = 0.1 \text{ (eV } \text{Å})^2$ [3.25] and for $n_s = 1.0 \times 10^{12} \text{ cm}^{-2}$ [3.33]. $1/\tau_{CI}(E_k)$ decreases with the increasing $E_k$ whereas $1/\tau_{SD}(E_k)$ is almost independent of $E_k$. The magnitude of $1/\tau_{CI}(E_k)$ and $1/\tau_{SD}(E_k)$ in suspended BLG is greater than those in sample with substrate. This is attributed to the smaller value of strength of the short-range disorder potential and charged impurity concentration. Besides, the reduced dielectric constant in the screening function in the charged impurity scattering may also enhance these scattering rates.

The scattering parameter $p$ due to each of these mechanisms is shown in Figure 3.8, for illustration, at $T = 4.2 \text{ K}$ for suspended BLG. For short-range disorder scattering $p$ is almost zero for the range of $n_s$ considered. $p$ due to charged impurity scattering is weakly increasing with the increasing $n_s$. $p$ due to acoustic phonon scattering seems to be almost independent of $n_s$. Interesting to note that the $p$ due to resultant relaxation time is found to be closer to that of short-range disorder indicating the dominancy of this scattering.

![Figure 3.8](image)

**Figure 3.8.** Scattering parameter $p$ vs electron concentration $n_s$ for different scattering mechanisms at $T = 4.2 \text{ K}$ for suspended BLG.

Mobility vs temperature is presented in Figure 3.9. $\mu_{AP}$ vs $T$ is same as discussed in supported BLG. For the chosen parameters, with low impurity concentration, the resultant mobility is found to be dominated by impurity scattering below 50 K. However, we notice that $\mu_{AP}$ is found to dominate the impurity scattering
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for about $T > 50$ K. The resultant mobility is found to decrease with the increase of $T$, but more rapidly compared to the case of the sample with the substrate.

![Figure 3.9](image1)

**Figure 3.9.** Mobility $\mu$ vs temperature $T$ for $n_s = 1 \times 10^{12}$ cm$^{-2}$ for a suspended BLG. Curve due to SD mechanism coincides with that of resultant $\mu$.

![Figure 3.10](image2)

**Figure 3.10.** Diffusion thermopower $S^d$ vs temperature $T$ for $n_s = 1 \times 10^{12}$ cm$^{-2}$ for a suspended BLG. Curve due to SD mechanism coincides with that of resultant $S^d$.

$S^d$ calculations are carried out for suspended BLG taking scattering due to impurities and acoustic phonons. In Figure 3.10, $S^d$ calculations are presented...
choosing \( n_s = 1.0 \times 10^{12} \text{ cm}^{-2} \). It is found to increase linearly at low \( T \) and sublinearly in the high \( T \) region as found in supported BLG. The \( S^d \) due to short-range disorder is closer to the resultant \( S^d \), still showing its dominancy as found in case of sample with the substrate. There is an enhancement in the value of \( S^d \) to a small extent, in the larger temperature region, as compared to that of supported BLG. A relatively smaller value of \( S^d \) in supported BLG may be attributed to the significant screening of the charged impurity scattering.

![Figure 3.11](image-url)

**Figure 3.11.** Diffusion thermopower \( S^d \) vs electron concentration \( n_s \) at \( T = 300 \) K for a suspended BLG. Curve due to SD mechanism coincides with that of resultant \( S^d \).

\[
S^d \text{ vs } n_s = (0.5-10) \times 10^{12} \text{ cm}^{-2}, \text{ at } T=300 \text{ K, for suspended BLG, is found to show (Figure 3.11) similar behavior as that of supported BLG with } n_s^{-1}\text{dependence. Curves due to short-range disorder and acoustic phonon are found to coincide with the resultant } S^d. \text{ The values of } S^d \text{ due to each of the mechanisms are found to remain almost same which is attributed to the first term in Eqn. (3.2.8). This term contains the ratio } \langle \tau(E_k)E_k \rangle/\langle \tau(E_k) \rangle \text{ which is least affected by the energy independent parameters of the relaxation time.}

In suspended graphene, there exists calculations of electron scattering by flexural (out-of-plane) phonons and this is shown to dominate the intrinsic phonon contribution to the resistivity [3.36] and mobility [3.37, 3.38] in MLG and resistivity in BLG [3.39]. It is also shown that the effect of flexural phonons is strongly
suppressed by applying strain [3.37-3.39]. Consequently, our calculations presented here are valid for strained suspended BLG. For the unstrained BLG there is need for inclusion of scattering due to flexural phonons in the phonon contribution to $S^d$.

### 3.4 Conclusions

In summary, we have presented calculation of $S^d$ for both supported and suspended BLG along with the mobility $\mu$ calculations due to different scattering mechanisms. At very low $T$, acoustic phonon limited mobility $\mu_{AP} \sim T^{-4}$, with an additional interesting new feature of non-monotinous behavior due to chiral property of the electrons. At higher $T$, $\mu_{AP} \sim T^{-1}$, where as, mobilities due to short-range disorder $\mu_{SD}$ and ionized impurity $\mu_{CI}$ are nearly independent of temperature. Resultant mobility is limited, largely, by impurity scattering.

Low temperature $S^d$ shows linear $T$ dependence and inverse $n_s$ dependence. $S^d$ is found to increase almost linearly with $T$ nearly up to 100 K and becoming sublinear at higher $T$. A qualitative agreement is obtained with the experimental data. It is found that, largely, short-range disorder scattering limits $S^d$. $S^d$ found to get enhanced marginally in suspended BLG compared to the one grown on a substrate. $S^d$ found to vary as $n_s^{-1}$. The calculations presented here may help to provide a better understanding of relative importance of the various scattering mechanisms that contribute to $\mu$ and $S^d$ in BLG. More experimental results are required to confirm the predictions in the present calculations.
Reference


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