Decay and Frequency Shift of Inter and Intravalley Phonons in
Graphene –Dirac Cone Migration–

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Abstract

By considering analytical expressions for the self-energies of intervalley and intravalley phonons in graphene, we describe the behavior of D, 2D, and D’ Raman bands with changes in doping ($\mu$) and light excitation energy ($E_L$). Comparing the self-energy with the observed $\mu$ dependence of the 2D bandwidth, we estimate the wavevector $q$ of the constituent intervalley phonon at $\hbar v q \simeq E_L/1.6$ ($v$ is electron’s Fermi velocity) and conclude that the self-energy makes a major contribution (60%) to the dispersive behavior of the D and 2D bands. The estimation of $q$ is based on a concept of shifted Dirac cones in which the resonance decay of a phonon satisfying $q > \omega/v$ ($\omega$ is the phonon frequency) into an electron-hole pair is suppressed when $\mu < (\hbar v q - \hbar \omega)/2$. We highlight the fact that the decay of an intervalley (and intravalley longitudinal optical) phonon with $q = \omega/v$ is strongly suppressed by electron-phonon coupling at an arbitrary $\mu$. This feature is in contrast to the divergent behavior of an intravalley transverse optical phonon, which bears a close similarity to the polarization function relevant to plasmons.
The Raman spectrum of graphene has two prominent peaks called the G and 2D (or G') bands that are very informative characterization tools. The 2D band at $\sim 2600 \text{ cm}^{-1}$ has been used to distinguish a single layer from graphene layers. The G band at $\sim 1580 \text{ cm}^{-1}$ can be used to determine whether or not the position of the Fermi energy $\mu$ is close to the Dirac point, since the width broadens when $\mu \simeq 0$. By contrast, the 2D bandwidth sharpens when $\mu \simeq 0.4$. What is the origin of the difference between the $\mu$ dependencies of the G and 2D bands?

As illustrated in Fig. 1(a), the presence (absence) of a resonant process by which the phonon decays into a real electron-hole pair, enhances (suppresses) the spectral broadening. Because the G band consists of $\Gamma$ point phonons, a direct transition is a unique decay channel that conserves momentum. Thus, the $\mu$ dependence of the G bandwidth is readily understood in terms of the Pauli exclusion principle. Meanwhile, the 2D band involves two near K point (intervalley) phonons, and the spectral broadening is induced by an indirect transition that crosses two valleys, as shown in Fig. 1(b). The presence or absence of a resonance decay channel for a phonon with a nonzero wavevector is the key to answering the question posed above. In this paper, we provide the answer in a unified manner by translating the Dirac cone.

Figure 1(b) shows that an intervalley phonon (zigzag line) can change into an electron-hole pair (loop) as a result of an electron-phonon interaction. The wavevector of an intervalley phonon is written as $k_F + q$, where $k_F$ is a wavevector pointing from the K point to the K' point and $q (= |q|)$ is much smaller than $|k_F|$. Suppose that a hole is located at $k$ measured from the K point, then the wavevector of the electron is given by $k + (k_F + q)$ because of momentum conservation. As a result, the wavevector of the electron measured from the K' point is $k + q$, and the energies of the hole and electron are given by $-\hbar v k$ ($= -\hbar v |k|$) and $\hbar v |k + q|$, respectively, where $v (~10^6 \text{ m/s})$ denotes the Fermi velocity. Hereafter, we use units in which $\hbar = 1$.

An indirect transition between two valleys can be regarded as a “direct” transition by translating the Dirac cone at the K' point to $-(k_F + q)$ as shown in Fig. 1(b). With the shifted Dirac cones, it is easy to capture the essential feature of the broadening of a $q \neq 0$ phonon. When $\mu = 0$, we see in Fig. 1(c) that there is an energy gap, $v q$, between the conduction and valence bands. This energy gap precludes a phonon with frequency $\omega (< v q)$ from decaying into a real electron-hole pair. On the other hand, when sufficient doping is
FIG. 1: (a) The $\mu$ dependence of G band broadening (frequency $\omega_G$). (b) An electron-hole pair between two valleys. The Dirac cone at the K (K') point is indicated in black (red). With the migration of the Dirac cone, the electron-hole pair creation process is viewed as a direct transition. (c) The $\mu$ dependence of the broadening of an intervalley phonon is different from that of the G band. Note that spectral broadening of an intravalley phonon can be discussed with the replacement $K' \rightarrow K$ (or $K \rightarrow K'$).

achieved as shown in Fig. 1(c), the phonon can decay into an intraband electron-hole pair. This intraband decay channel results in spectral broadening. When $q = 0$, the two Dirac cones are merged into one [inset in Fig. 1(b)] and the energy gap vanishes. Then, it is clear that the broadening of the $q = 0$ phonon bears similarities to that of the G band.\textsuperscript{2-6} The $\mu$ dependence of the broadening of an intervalley phonon with $vq > \omega$ differs greatly from that of the G band, and the concept of the shifted Dirac cones is useful for understanding the $\mu$ and $q$ dependencies of the broadening in a unified manner.

More detailed information about the broadening can be obtained by calculating the self-
energy. The self-energy of the intervalley phonon with \( q \) and \( \omega (> 0) \) is defined by

\[
\Pi_\mu(q, \omega) \equiv g_{ep} \frac{(2\pi)^2}{V} \sum_{s,s'} \sum_k \frac{f_{k,\mu}^s - f_{k+q,\mu}^{s'}}{\omega + \epsilon v q - s' v|k + q| + i\epsilon} \left( 1 - s s' \frac{k + q \cos \varphi}{|k + q|} \right) \quad (1)
\]

In Eq. (1), \( s (= \pm 1) \) and \( s' (= \pm 1) \) are band indices, \( f_{k,\mu}^s = \lim_{\beta \to \infty} (1 + e^{\beta (s\epsilon v|k| - \mu)})^{-1} \) is the Fermi distribution function defined at zero temperature and with a finite doping \( \mu \), and \( \epsilon \) is a positive infinitesimal. We can assume \( \mu > 0 \) without losing generality because of particle-hole symmetry. The factor \( g_{ep} \) denotes the electron-phonon coupling strength, \( \varphi \) denotes the polar angle between \( k \) and \( q \), and the term, \( g_{ep} \times (1 - s s' \frac{k + q \cos \varphi}{|k + q|}) \), is the square of the electron-phonon matrix element for the intervalley phonon, which will be discussed later. The broadening and modified frequency are given by \(-\text{Im}\Pi_\mu(q, \omega)\) and \( \omega + \text{Re}\Pi_\mu(q, \omega) \), respectively.

In the continuum limit of \( k \), the broadening normalized by \( g_{ep} \) leads to \([22]\)

\[
-\text{Im}\Pi_\mu(q, \omega) = \pi \sqrt{\omega^2 - vq^2} \theta_{\omega - vq} \left[ \theta_{\frac{\omega - \mu}{vq} - \frac{\pi}{2}} + \theta_{\frac{\omega + vq}{vq} - \frac{\pi}{2}} \left\{ \frac{\pi}{2} - \sin^{-1} \left( \frac{2\mu - \omega}{vq} \right) \right\} \right] \\
+ \pi \sqrt{vq^2 - \omega^2} \theta_{vq - \omega} \left[ \theta_{\frac{2\mu + \omega}{vq} - \frac{\pi}{2}} - \theta_{\frac{\omega - vq}{vq} \frac{\pi}{2}} \left\{ \frac{2\mu - \omega}{vq} \right\} \right], \quad (2)
\]

where \( \theta_x \) denotes the step function satisfying \( \theta_{x \geq 0} = 1 \) and \( \theta_{x < 0} = 0 \), and \( g(x) \equiv \log(x + \sqrt{x^2 - 1}) \). Figure 2(a) shows a 3-dimensional (3d) plot of \(-\text{Im}\Pi_\mu(q, \omega)\) as a function of \( vq \) and \( \mu \) when \( \omega = 0.2 \) (eV), which corresponds to the Debye frequency of carbon (\( \omega_D \)). As indicated in Fig. 2(a), a line node appears for \( vq = \omega_D \). In Eq. (2), this line node is a critical line separating the two terms, which are proportional to \( \theta_{\omega - vq} \) and \( \theta_{vq - \omega} \), and it can be shown that the first (second) term originates from the contributions of interband (intraband) electron-hole pair creation. For example, the \( q = 0 \) phonon satisfies \( \theta_{vq - \omega} = 0 \), and only the interband electron-hole pairs cause spectral broadening. The first term in Eq. (2) leads to \(-\text{Im}\Pi_{\mu < \omega/2}(0, \omega) = \pi^2 \omega \) and \( \text{Im}\Pi_{\mu > \omega/2}(0, \omega) = 0 \), which are consistent with the behavior of the G band\([2-4]\). Contrastingly, for the phonon satisfying \( vq > \omega \), we can confirm that from the second term of Eq. (2) spectral broadening is possible only when there is sufficient doping, namely when \( \mu > \frac{\omega^2 - \omega vq - vq^2}{2} \) is satisfied. A sharp step appears at \( vq = 2\mu + \omega_D \), as indicated in Fig. 2(a). In Fig. 2(b), we plot \(-\text{Im}\Pi_\mu (q, \omega_D)\) as a function of \( \mu \) to show more clearly the \( q \) dependence of the broadening. It is seen that for \( vq > 0.2 \) [red curves], \(-\text{Im}\Pi_\mu(q, \omega_D)\) is suppressed when the Fermi energy is close to the Dirac point \( (\mu < \frac{vq - 0.2}{2}) \), and broadening appears when \( \mu > \frac{vq - 0.2}{2} \).
FIG. 2: (a) 3d plot of $-\text{Im}\Pi_{\mu}(q, \omega_D)$. The variables $vq$ and $\mu$ are given in units of eV. The cross section of (a) for different $vq$ values (b), and for different $\mu$ values (c). In (c), $vq$ is proportional to the light excitation energy $E_L$.

Because the Raman 2D band consists of two intervalley phonons satisfying $vq > \omega_D$ the suppressed broadening when $\mu \simeq 0$ also holds for the 2D band. Das et al. have shown that the 2D bandwidth sharpens when $\mu \simeq 0$. A suppressed broadening of the 2D band ($2\omega_q \simeq 0.32$ eV) has also been observed when $\mu \leq 0.4$ eV in a recent experiment reported by Chen et al., from which we estimate the $q$ value to be $vq \simeq 0.96$ eV using $0.4 \simeq \frac{vq-0.16}{2}$. 
The validity of this estimation ($vq \simeq 0.96$ eV) can be further investigated by changing $q$. In Fig. 2(c), we show the plot of $-\text{Im}\Pi_\mu(q, \omega_D)$ as a function of $q$ for different $\mu$ values. For $vq \simeq 0.96$, increasing $q$ would cause the broadening to decrease (increase) when $\mu = 0.4$ (0.8) eV. Because $vq$ is related to light excitation energy $E_L$ through momentum conservation, the broadening can also depend on $E_L$. If we assume $E_L = \alpha vq$, $\alpha \simeq 1.6$ is obtained as a fitting parameter because $E_L = 1.58$ eV is used in the experiment. A similar $\alpha$ parameter value ($\alpha \simeq 1.3$) can be obtained by calculation.

A 3d plot of the real part of the self-energy, $\text{Re}\Pi_\mu(q, \omega_D)$, is shown in Fig. 3(a). The plot is based on the analytical expression of $\text{Re}\Pi_\mu(q, \omega)$ given by

$$\begin{align*}
\text{Re}\Pi_\mu(q, \omega) &= 4\pi\mu \\
&+ \pi \sqrt{\omega^2 - v^2 q^2} \theta_{\omega-vq} \left[ -g \left( \frac{\omega + 2\mu}{vq} \right) + \theta_{\omega-vq} - \mu g \left( \frac{\omega - 2\mu}{vq} \right) \right] \\
&+ \pi \sqrt{v^2 q^2 - \omega^2} \theta_{\omega-vq} \left[ \theta_{\omega-vq} - \mu \left( \frac{\pi}{2} - \sin^{-1} \left( \frac{\omega + 2\mu}{vq} \right) \right) \right] + \theta_{\omega+vq} - \mu \left( \frac{\pi}{2} - \sin^{-1} \left( \frac{2\mu - \omega}{vq} \right) \right) \right].
\end{align*}$$

(3)

For the $q = 0$ phonon, Fig. 3(a) shows that the softening is maximum at $\mu = 0.1$ (eV). Equation 3 is simplified in the limit of $q \to 0$, as

$$\text{Re}\Pi_\mu(0, \omega) \simeq 4\pi\mu + \pi\omega \log \left| \frac{\omega - 2\mu}{\omega + 2\mu} \right|,$$

(4)

and the large softening is caused by the logarithmic singularity at $\mu = \omega_D/2$. This feature is exactly the same as the Kohn anomaly of the G band. It also shows that the logarithmic singularity is removed gradually as we increase $q$ from zero. Figure 3(a) shows that when $\mu$ is sufficiently large the real part increases linearly with $\mu$ as $\text{Re}\Pi_\mu(q, \omega_D) \simeq 4\pi\mu$ for an arbitrary $q$ value. Interestingly, $\text{Re}\Pi_\mu(q, \omega_D)$ increases as we increase $q$ (or $E_L$), even for a fixed $\mu$ value. This feature is more clearly seen in Fig. 3(c), and suggests that the self-energy contributes to the dispersive behavior of the 2D (or D) band; the 2D band frequency increases linearly with $E_L$ ($\partial \omega_{2D}/\partial E_L \simeq 100$ cm$^{-1}$/eV). If we use $g_{ep} = 5$ cm$^{-1}$, which is obtained from the broadening data published by Chen et al., the self-energy can account for $\sim 60\%$ of the dispersion because $2 \times \text{Re}\Pi_{\mu=0}(q, \omega_q) \simeq 2g_{ep} \pi^2 vq = 2g_{ep} \pi^2 E_L/\alpha$ and $2g_{ep} \pi^2 /\alpha = 61.6$ cm$^{-1}$.

In Fig. 3(a), for a fixed $vq$ that is larger than $\omega_D$, $\text{Re}\Pi_\mu(q, \omega_D)$ undergoes two discontinuities at $\mu = \frac{\omega_D - \omega q}{2}$ and $\frac{\omega_D + \omega q}{2}$. A modest softening appears as $\mu$ approaches $\frac{\omega_D - \omega q}{2}$ from
FIG. 3: (a) A 3d plot of $\text{Re}\Pi(q, \omega_D)$. The variables $vq$ and $\mu$ are given in eV. (b) The cross section of (a) for different $vq$ values, and (c) for different $\mu$ values. Note that $\text{Re}\Pi(q, \omega)$ does not include the $q$ dependence of the bare frequency.

zero. This is consistent with the observations by Das et al., Chen et al., and Araujo et al. showing that the 2D band frequency remains almost constant (disregarding a small modulation of about 8 cm$^{-1}$) when the Fermi energy is near the Dirac point. On the other hand, the 2D band frequency exhibits a slight hardening of $\sim 2$ cm$^{-1}$ in the observation reported by Yan et al. We consider that the data actually show that the 2D band frequency does not depend on doping because the observed small amount of hardening is within the spectral resolution (2 cm$^{-1}$).

As we increase $\mu$ further, $\text{Re}\Pi(q, \omega_D)$ undergoes slight hardening and subsequent soft-
ening until $\omega > \mu$. These features can also be seen in Fig. 3(b). The discontinuities of $\text{Re}\Pi_\mu(q, \omega)$ can be explained by the perturbation theory: the energy correction by a virtual state with energy $\varepsilon$ is proportional to

$$\frac{1}{\omega - \varepsilon}. \quad (5)$$

Because the sign of $(\omega - \varepsilon)^{-1}$ is positive (negative) when $\varepsilon < \omega$ ($\varepsilon > \omega$), the lower (higher) energy electron-hole pair makes a positive (negative) contribution to $\text{Re}\Pi_\mu(q, \omega)$. Therefore, when $\nu \nu > \omega$, softening is induced by the doped carriers since the energy of a virtual state is approximately given by $\nu \nu$, which is larger than $\omega$, and thus $1/(\omega - \nu \nu) < 0$ is satisfied [see Fig. 4(a)]. In fact, the energy $\varepsilon$ corresponds to $v|k + \nu q| - v k$ in Eq. (1) and $\varepsilon \simeq \nu \nu$ when $k \simeq 0$. The softening magnitude is tiny as shown in Fig. 3(a) and (b) because the electron density vanishes at the Dirac point. When $\mu = \frac{\nu \nu - \omega}{2}$, an intraband electron-hole pair with $\varepsilon \leq \omega$ can start to be excited [see Fig. 3(b)], and this electron-hole pair causes hardening. Note that some of the doped carriers satisfying $\frac{\nu \nu - \omega}{2} < \mu < \frac{\nu \nu + \omega}{2}$ contribute to the softening, and the hardening is partly cancelled by the softening. The details of the cancellation are determined by the $\varphi$ dependence of the electron-phonon coupling term, $1 - ss' \frac{k + \nu q \cos \varphi}{|k + \nu q|}$, in Eq. (1). Because the intraband transition satisfies $ss' = 1$, the matrix element vanishes when $\varphi = 0$ and thus hardening dominates softening (unless $\omega$ is negligible compared with $\nu \nu$). When $\mu$ approaches $\frac{\nu \nu + \omega}{2}$, the Pauli exclusion principle forbids the occurrence of some of the intraband transitions that contribute to the hardening, which accounts for the appearance of the softening. For $\mu \geq \frac{\nu \nu + \omega}{2}$, the frequency exhibits hardening due to the suppression of the softening induced by interband ($ss' = -1$) virtual electron-hole pairs.

As we have seen, a phonon’s self-energy can be very sensitive to the electron-phonon matrix element. In particular, the sign of the coefficient of $ss'$ in

$$1 - ss' \frac{k + q \cos \varphi}{|k + q|}, \quad (6)$$

is critical in determining the behavior of the self-energy. In fact, if the minus sign is replaced with a plus sign as follows

$$1 + ss' \frac{k + q \cos \varphi}{|k + q|}, \quad (7)$$

the corresponding self-energy exhibits a singularity at $\nu \nu = \omega$. In addition, the real part exhibits softening as $\sim -4\pi \mu$ when $\nu \nu > \omega$ because the matrix element is maximum when
FIG. 4: The projection of the Dirac cones. (a) Slight doping (red) causes a softening, while (b) a heavy doping (blue) causes hardening as well as softening.

ϕ = 0 and thus softening dominates hardening. These features are in marked contrast to the fact that the imaginary part of the self-energy given by Eq. (6) exhibits a nodal structure at \( vq = \omega \) and that the real part exhibits hardening as \( \sim 4\pi\mu \). The self-energy with Eq. (7) corresponds to the self-energy of the Coulomb potential known as the polarization function, and the singularity at \( vq = \omega \) is important for plasmons in graphene.18,19

Intravalley longitudinal optical (LO) and transverse optical (TO) phonons are related to the intervalley phonon and plasmon. The corresponding elements of the electron-phonon interactions are given by

\[
1 - ss \frac{k + q \cos \varphi}{|k + q|} + ss \frac{2k \sin^2 \varphi}{|k + q|}, \quad \text{(LO),} \tag{8}
\]
\[
1 + ss \frac{k + q \cos \varphi}{|k + q|} - ss \frac{2k \sin^2 \varphi}{|k + q|}, \quad \text{(TO).} \tag{9}
\]

The first two terms for the LO [TO] phonon are the same as Eq. (6) [Eq. (7)]. By constructing an analytical expression for the self-energy of the LO and TO phonons27 we visualize the self-energies in Fig. 5. In Fig. 5(a), we show the imaginary part of the LO phonon. A notable feature of Fig. 5(a) is that for \( vq > \omega_D \), the broadening increases as \( \mu \) is increased. This is a sharp contrast to the broadening of the intervalley phonon, which is suppressed for heavy doping (see Fig. 2(a)). For the real part shown in Fig. 5(b), a discontinuous feature caused by the last term in Eq. (8) is clearly seen at \( vq = \omega_D \). Interestingly, the TO phonon has some similarities to the polarization function: the existence of a singularity and the
frequency softening for $vq > \omega_D$, as shown in Fig. 5(c) and (d). It is instructive to compare the self-energy of an intravalley LO phonon with that of a TO phonon. The self-energy of the LO phonon is the same as that of the TO phonon for the $\Gamma$ point $q = 0$, and their difference is highlighted for nonzero $q$ values ($q > \omega/v$). The difference between the LO and TO phonons will be useful in allowing us to determine the optical phonon (LO or TO) composing the $D'$ band.

![Graphs showing LO and TO decay and shift](image_url)

**FIG. 5:** 3d plot of $\Pi_\mu(q, \omega_D)$ for an intravalley LO phonon (a,b) and TO phonon (c,d). (a,c) the imaginary part, and (b,d) the real part. The variables $vq$ and $\mu$ are given in eV.

Coulomb interactions among electrons which we did not consider in this paper might make an effect on the self-energies of phonons.\textsuperscript{20} Attaccalite et al. point out the importance of vertex-corrections to electron-phonon coupling by electron-electron interaction.\textsuperscript{21} However, their results concern the phonon at the exact K point, which corresponds to the $q = 0$ phonon, and the $q = 0$ phonon has nothing to do with the experimentally observed 2D band phonon. Our main results remain unchanged even if we include such a correction, because our approach - shifting the Dirac cones - is based only on momentum conservation and does not depend on any dynamical details. Moreover, since we have determined the electron-phonon coupling using experimental data, such effects, if any, are all included.
In conclusion, employing a concept of shifted Dirac cones, we clarified that a phonon satisfying $vq > \omega$ does not decay into an electron-hole pair when $\mu < (vq - \omega)/2$. This is a general consequence that is independent of the details of electron-phonon coupling and can be applied to both inter and intravalley phonons. Based on the self-energy, which includes the effect of electron-phonon coupling, we estimated the $q$ value of the 2D band at $vq \simeq 1$ (eV) by referring to recent experimental data on the $\mu$ dependence of broadening. This value $vq \simeq 1$ (eV) also suggests that about 60% of the dispersive behavior can be attributed to the self-energy. Since $vq$ is proportional to $E_L$, the $q$ dependence of the self-energy may be explored by using a tunable laser, without changing $\mu$ by controlling the gate voltage. For example, the Fermi energy position of graphene can be determined from the $E_L$ dependence of the broadening. Several anomalous features have been pointed out in the self-energies for intravalley LO and TO phonons. The differences between the LO and TO phonons will be useful for specifying the mode and $q$ value of the D’ band.

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22 See Supplemental Material at [URL] for derivation.
23 See Supplemental Material at [URL] for details.
24 See Supplemental Material at [URL] for derivation.
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27 See Supplemental Material at [URL] for derivation.