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Optoelectronic Properties

and Plasmonic Devices of Graphene

A dissertation submitted in partial satisfaction of the
requirements for the degree Doctor of Philosophy
in Electrical Engineering

by

I-Tan Lin

2016
ABSTRACT OF THE DISSERTATION

Optoelectronic Properties and Plasmonic Devices of Graphene

by

I-Tan Lin

Doctor of Philosophy in Electrical Engineering

University of California, Los Angeles, 2016

Professor Jia-Ming Liu, Chair

Graphene is a one-atom-thick 2D system that has a unique hexagonal crystal structure of two carbon atoms per unit cell. Unlike any other 2D semiconductor material known today, intrinsic graphene has a zero bandgap with its charged carriers behaving like Dirac fermions with a zero mass, resulting in many extraordinary properties that are very different from other materials. Such properties can be controllably modified by proper impurity doping or by electrical or optical modulation, making graphene extremely attractive for novel device applications. The salient electronic, optical, and optoelectronic properties of graphene, together with its unique nanostructure, offer innovative opportunities to many potentially revolutionary applications for high-speed/high-frequency electronic and optoelectronic devices, terahertz (THz) oscillators and sensors, and ultrafast nonlinear optical elements. Realization of these exciting graphene-based devices, as well as the possibility for further innovation, relies on a good understanding of graphene’s electronic, optical and optoelectronic properties in the broad
spectral range from THz to the visible. In this thesis, the electronic property of graphene is investigated first, followed by the discussion of THz property of graphene. Based on the derived models of the electronic and THz properties of graphene, we can describe the plasmonic behavior of graphene in various configurations, such as graphene-based THz waveguide. A concept THz device is demonstrated both theoretically and experimentally in the last.
The dissertation of I-Tan Lin is approved.

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University of California, Los Angeles
2016
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Last but not least, I would like to thank my family for their encouragement and support through my graduate study. It is my parents who encouraged me to enroll in the Ph.D. program, and I am glad that I did. Therefore, I dedicate my achievement to my family.
Biography

I-Tan Lin received the B.S. degree in electrical engineering from National Chiao Tung University, Taiwan, in 2009, and M.S degree in electrical engineering in 2012 from the University of California, Los Angeles, where he conducted the research of optoelectronic properties and plasmonic devices of graphene in Dr. Jia-Ming Liu’s group. He was awarded Dissertation Year Fellowship in 2015. He has 9 first-author and 2 co-author papers published in peer-review journals. His current research interests include graphene-based optoelectronics, terahertz frequency devices, and plasmonics.
Chapter 1. Introduction

1.1. Motivation and outline

With the advancement of the terahertz (THz) generation technique and the development of THz time-domain spectroscopy (THz-TDS), THz research has been experiencing explosive interests in the last decade. THz technology has found many applications in various sectors, such as communication, security screening, medical imaging, biology sciences, and non-destructive evaluation. Much interest has been focused on the THz communication and information applications. In the past two decades, the bandwidth demand for wireless communication has increased rapidly. The demand for bandwidth has doubled every 18 months for the last 25 years, following the Edholm’s law of bandwidth. If this trend continues, the current microwave communications systems operating at carrier frequencies within gigahertz (GHz) spectral region will be replaced or compensated by THz communication system in the near future.\(^1\) THz wireless communication also enjoys many advantages over microwave and infrared wireless links. For example, higher bandwidth compared to microwave links and lower attenuation under certain weather conditions compared to IR links. Due to the high data rates, THz communication links also serves as a suitable solution for the “last mile” or “first mile” problem. Therefore, due to these advantages and recent breakthrough in high-power THz generation and the development of THz-TDS, there has been an increasing interest in THz communication technology in the past decade.\(^2\)-\(^6\).

To realize the THz communication systems, it is necessary to develop low-loss THz waveguides and highly functional THz modulators and detectors. One approach to modulate THz waves is based on the free-carrier absorption, achieved by gated two-dimensional electron gas
structure, such as gated GaAs/AlAs multiple-quantum-well. Other way is to induce the geometrically induced plasmons, also known as the spoof plasmons, to manipulate the THz wave. Some novel THz waveguides have also been proposed in the literature, such as metal wires, hollow-glass metallic waveguides, and air-core microstructure fiber. However, they all suffer from high loss and large footprint. The typical lengths of the relevant structural parameters are in the range of 50µm to few millimeters for the resonance frequency in the sub-THz region. Furthermore, the resonance frequency is usually fixed and cannot be tuned. Instead of using metal or semiconductor as the material in building the THz communication devices, in this thesis we investigate the possibility of using graphene to manipulate the THz wave for graphene being naturally compact and its strong response to the THz wave.

The successful isolation of graphene in 2004 has triggered great interest in the graphene research in the past decade. As graphene is the “true” 2D material, it is naturally compact, and can be easily integrated into currently existent devices by putting or wrapping thin films on the devices. The carrier response of graphene to the THz electromagnetic waves can be described by the Drude model; in the right condition, graphene plasmons can be excited in the THz spectral region, whereas the plasmon frequency of metal is within or above the optical spectral region, and thus can be hardly used as the material in the plasmonic THz devices. The plasmon dispersion of graphene can be found by calculating the dynamic polarization function within random phase approximation (RPA). Because of the vanishing density of states and bandgap around the Dirac point, the Fermi energy changes dramatically with low carrier density, rendering a remarkable tunability of graphene plasmon frequency by electrical gating or doping. The plasmonic properties of graphene can also be tuned by numerous extrinsic and intrinsic factors; these factors include the number of graphene layers, the stacking orders and rotational
angles among layers, the substrate, light frequency, and temperature. Plasmons under different geometries are also considered in the literature, such as double-layer graphene with a varying distance between them, graphene nanostructures (antidots, ribbons, nanodisks, and rings), and graphene deposited on grating structures or stacked with other materials. The physics becomes even more interesting when graphene plasmons are coupled with other plasmons of different materials, with surface phonons of substrate, or with photons to form plasmon-polaritons. Therefore, because of these aforementioned advantages, such as small footprint, great tunability, strong THz response and strong interaction with surrounding materials, graphene is regarded as a promising building block for the THz devices.

Despite the rapidly increasing number of theoretical and experimental studies of the optical properties of graphene, there are still many unresolved open questions, especially in the realm of optoelectronic devices in the THz spectral region. Therefore, another focus of my thesis is to address these questions along the line of developing the graphene-based THz devices. For example, the frequency dependence of the scattering rate in the THz spectral region, various modes in the graphene waveguide and their propagation distances, and the excitation efficiency of graphene plasmons on a grating, to name a few. In the following of this chapter, we first discuss the general physics of graphene and multilayer graphene including the band structures, density of states, and temperature dependence of the chemical potential. Following Chapter 1, the electronic properties, the terahertz optical properties, and the plasmonic properties of graphene are investigated in Chapter 2, Chapter 3, and Chapter 4, respectively. Various scattering mechanisms are discussed in Chapter 2. The surface polar optical phonon scattering of carriers in graphene on various substrates is thoroughly studied using Rode’s iteration method. The dependence of the scattering rate on carrier energy, temperature, and Fermi energy is
investigated. In Chapter 3, we present an experimental study where the optical conductivity of monolayer and multilayer graphene in the terahertz spectral region is measured using terahertz time-domain spectroscopy. Physical parameters, including carrier density, dc conductivity, and carrier mobility, of each sample can also be deduced from the theoretical fitting of experimental data. The plasmonic properties of graphene and graphene-based structures are discussed in Chapter 4, and their applications are presented in Chapter 5, where concept devices such as waveguides and terahertz devices are thoroughly studied both theoretically and experimentally. The unique combination of graphene and other materials in various miniature structures is promising for many graphene-based plasmonic devices, such as THz filters, modulators, detectors, and highly integrated THz devices. The conclusion is drawn in Chapter 6.

1.2. Graphene band structure

The electronic band structure of graphene can be obtained by using the tight-binding model. An analytical expression for the band structure can be found by assuming all long-range interactions much weaker than the self-interaction of each carbon atom at a lattice point and the interactions of the atom with its three nearest neighbors. Then, the band structure is found as

\[ E(k) = \varepsilon \pm \gamma_0 \sqrt{3 + 2 \cos(\sqrt{3} k_y a) + 4 \cos \left( \frac{\sqrt{3}}{2} k_y a \right) \cos \left( \frac{3}{2} k_x a \right)} , \]

where \( \varepsilon \) is the self-interaction energy and \( \gamma_0 \) is the nearest-neighbor hopping energy in graphene. The band structure is plotted in Figure 1.1(a) using (1.1).
As can be seen in Figure 1.1(a), the conduction band touches the valence band at $E = \varepsilon$. The self-interaction energy $\varepsilon$ can be set to be zero for simplicity so that an electron in a state on the conduction band has a positive energy and that in a state on the valence band has a negative energy. The points at $E(k) = 0$ are called the Dirac points; the vicinities of these points are referred to as the valleys, where the graphene band structure can be described by the relativistic Dirac equation. As in the case of the real space where there are two sublattices A and B, in the $k$ space there are also two distinct groups of Dirac points K and K'. Inside the Brillouin zone, two Dirac points can be found at the $k$-space locations represented by the reciprocal space vectors:

$$K = \frac{4\pi}{3\sqrt{3}a} \left( \frac{\sqrt{3}}{2} \hat{x} + \frac{1}{2} \hat{y} \right), \quad K' = \frac{4\pi}{3\sqrt{3}a} \left( \frac{\sqrt{3}}{2} \hat{x} - \frac{1}{2} \hat{y} \right).$$

(1.2)

Figure 1.1. (a) Band structure of graphene plotted using (1.1) while setting the self-interaction energy to be $\varepsilon = 0$. The top surface of a higher energy is the conduction band ($\pi^*$ band), and the bottom surface of a lower energy is the valence band ($\pi$ band). The conduction band is projected onto a two-dimensional plane on the top. The circled region near the Dirac point $K'$ is enlarged.
For perfectly intrinsic graphene, the chemical potential, i.e., the Fermi level, is located at the energy level of the Dirac points, \( E = 0 \), so that the valence band is completely filled and the conduction completely empty. As most physics and carrier transitions happen at energy levels around the chemical potential, we shall rewrite (1.1) by shifting the origin of the \( k \) space to one of the Dirac points and assume a small \( k \) to simplify (1.1). Substituting \( k \) in (1.1) with \( K + k \) or \( K' + k \), we obtain, for \( |k| a \ll 1 \) in the vicinity of a Dirac point,

\[
E = \pm \gamma_0 \sqrt{\frac{9}{4} k_y^2 a^2 + \frac{9}{4} k_x^2 a^2} = \pm \frac{3}{2} a \gamma_0 |k| = \pm \hbar v_F k, \tag{1.3}
\]

where \( a = 0.142 \text{ nm} \) is the distance between two neighboring carbon atoms, \( \hbar \) is the reduced Planck’s constant, and

\[
\nu_F = \frac{3a \gamma_0}{2\hbar} \approx 1.0 \times 10^6 \text{ m s}^{-1} \tag{1.4}
\]

is the Fermi velocity of graphene. Equation (1.3) indicates that there is a region in the \( k \) space around each Dirac point where the carrier energy is linearly proportional to the wave number that is measured with respect to the given Dirac point. This region is called the Dirac cone, as shown in the blow-up in Figure 1.1(b). Because (1.3) can also be derived from the massless Dirac equation, electrons on the Dirac cone are also called Dirac electrons.

1.3. Density of states and carrier concentration
With the band structure near the Dirac point described by (1.3), the density of electron states of graphene in the energy range between $E$ and $E + dE$ for $E > 0$ near the conduction-band edge is

$$D_c(E) dE = \frac{1}{A} \sum_{k, E_k > 0} \delta(E_k - E) dE = \frac{g}{(2\pi)^2} \int_0^\infty \delta(\hbar \nu, k - E) 2\pi k \, dk dE = \frac{2E}{\pi (\hbar \nu)^2} dE, \quad (1.5)$$

and that for $E < 0$ near the valence-band edge is

$$D_v(E) dE = \frac{1}{A} \sum_{k, E_k < 0} \delta(E_k - E) dE = \frac{g}{(2\pi)^2} \int_0^\infty \delta(-\hbar \nu, k - E) 2\pi k \, dk dE = -\frac{2E}{\pi (\hbar \nu)^2} dE, \quad (1.6)$$

where $A$ is the area of the graphene sheet and $g = 4$ is the total degeneracy due to the spin degeneracy and the valley degeneracy (two valleys $K$ and $K'$ in one Brillouin zone). Because of the symmetry between the conduction band and the valence band, $D_c(E) dE = D_v(E) dE$ for the same absolute value of energy $|E|$.

From (1.6), the concentrations of electrons and holes are, respectively,

$$n_0 = \int_0^\infty D_c(E) f_0(E) dE, \quad (1.7)$$

$$p_0 = \int_{-\infty}^0 D_v(E) [1 - f_0(E)] dE,$$

where

$$f_0(E) = \frac{1}{e^{(E - \mu)/k_B T} + 1}, \quad (1.8)$$

is the equilibrium Fermi-Dirac distribution function with a chemical potential $\mu$ at a temperature $T$, and $k_B$ is the Boltzmann constant. The probability of finding an electron in a state at the
energy level $E$ is $f_0(E)$, and the probability of finding a hole in a state at the energy level $E$ is $1 - f_0(E)$. When the condition $|\mu| \gg k_B T$ is satisfied, $f_0(E)$ is approximately a Heaviside function $\theta(\mu - E)$, which has a value of 1 for $E < \mu$, 0.5 for $E = \mu$, and 0 for $E > \mu$. Therefore, for $|\mu| \gg k_B T$, we obtain

$$n_0 + p_0 = \int_0^\infty D_c(E) \theta(\mu - E) dE + \int_{-\infty}^0 D_v(E) \left[1 - \theta(\mu - E)\right] dE = \frac{\mu^2}{\pi (\hbar v_F)^2}.$$  \hspace{1cm} (1.9)

1.4. Temperature dependence of the chemical potential

The temperature dependence of $\mu(T)$ of graphene can be found by recognizing the fact that each additional thermally excited electron always comes with the creation of a hole to result in an electron-hole pair. Therefore, for a given piece of graphene, the difference between the electron concentration and the hole concentration, $n_0 - p_0$, is a conserved quantity that is independent of temperature. At zero temperature when $\mu(0) = E_F > 0$ so that it lies within the conduction band, we have $p_0 = 0$ and $n_0 - p_0 = n_0 + p_0 = \frac{E_F^2}{\pi \hbar^2 v_F^2}$ from (1.9). Therefore we can relate $\mu$ and $E_F$ by combining (1.7) and (1.9) to write

$$\frac{E_F^2}{\pi (\hbar v_F)^2} = \int_0^\infty D_c(E) f_0(E) dE - \int_{-\infty}^0 D_v(E) \left[1 - f_0(E)\right] dE,$$  \hspace{1cm} (1.10)

which can be written in a compact form
\[
\frac{E_k^2}{2} = \int_0^\infty \frac{E \sinh(\mu/k_B T)}{\cosh(E/k_B T) + \cosh(\mu/k_B T)} dE,
\]  
(1.11)

from which \( \mu \) can be obtained numerically. When \( \mu \gg k_B T \), the integral approaches \( \mu^2/2 \), and we have \( \mu = E_k \) from (1.11).

1.5. Band structure of multilayer graphene

When multiple layers of graphene are brought together to form a sheet of multilayer graphene, the interlayer interaction can fundamentally change the band structure. In this section, the band structures of AA and AB (Bernal) stacking orders are discussed. The misoriented stacking arrangement is discussed at the end.

Figure 1.2. (a) Intralayer (\( \gamma_0 \)) and interlayer (\( \gamma_1 \)) interactions of AB-stacked multilayer graphene showing one set of \( \alpha \) and \( \beta \) layers. (b) Top view of (a), showing the overlap between \( \alpha \) (bottom) and \( \beta \) (top) layers. (c) Side view of (a). A chain of alternating A and B carbon atoms connected by \( \gamma_1 \) along the \( z \) direction is shown.

**AB stacking order**
In AB-stacked multilayer graphene, the arrangement of layers follows the $\alpha\beta\alpha\beta \ldots$ order. Figure 1.2 illustrates the AB stacking by showing one set of $\alpha$ and $\beta$ layers. Nearest-neighbor intralayer ($\gamma_0$) and interlayer ($\gamma_1$) interactions are considered, as shown in Figure 1.2(a); high-order interactions beyond nearest neighbors are neglected for simplicity. The top layer ($\beta$ layer) is shifted relative to the bottom layer ($\alpha$ layer) by one C–C distance, as shown in Figure 1.2(b).

The band structure of $N$-layer AB-stacked multilayer graphene can be obtained in a manner similar to the derivation of the band structure of monolayer graphene. The eigenenergy $E$ is found as$^{15}$

$$E = \frac{1}{2} \left( E_0 \pm \sqrt{4 \left( \hbar \nu_t k \right)^2 + E_0^2} \right), \quad (1.12)$$

where $k_\parallel = \left( k_x^2 + k_y^2 \right)^{1/2}$ is the in-plane wave number, in contrast to the out-of-plane wave number $k_z$, and $E_0$ is given by

$$E_0 = 2\gamma_1 \cos \left( r \frac{\pi}{N+1} \right), \quad (1.13)$$

where $r$ is an integer. In the limit of $|E_0| \gg \hbar \nu_t k$, (1.12) has two low-$k_\parallel$ solutions:

$$E = E_0 + \frac{\left( \hbar \nu_t k \right)^2}{E_0} \quad (1.14)$$

and

$$E = -\frac{\left( \hbar \nu_t k \right)^2}{E_0} \quad (1.15)$$

Note that in both (1.14) and (1.15), $E$ varies quadratically, not linearly, with $k$.

By defining the
effective mass as \( m^* = |E_0|/2\nu_f^2 \), the first solution (1.14) has the form of \( E = E_0 \pm \left( \hbar k \right)^2 / 2m^* \), and the second solution (1.15) has the form of \( E = \mp \left( \hbar k \right)^2 / 2m^* \), where the upper signs are taken when \( E_0 > 0 \) and the lower signs are taken when \( E_0 < 0 \). Accordingly, the energy bands given by (1.14) are called massive bands, in contrast to the massless Dirac bands of monolayer graphene.

For even \( N \), there are \( N \) different values of \( E_0 \) given by (1.13). Therefore, (1.14) and (1.15) respectively gives \( N \) energy bands and thus a total of \( 2N \) massive energy bands. Because half of \( E_0 \) values are positive and the other half are negative, there are \( N \) conduction bands and \( N \) valence bands. As an example, the band structure of AB-stacked bilayer graphene \((N = 2)\) is shown in Figure 1.3(a), where the solid curves are obtained from (1.12) and the dashed curves are obtained from (1.14) and (1.15) under the low-\( k \) approximation, using \( \gamma_0 = 3.16 \text{ eV} \) and \( \gamma_1 = 0.37 \text{ eV} \) obtained from experiments.\(^{16} \)
Figure 1.3. Band structures of AB-stacked (a) bilayer and (b) trilayer graphene. The solid curves in (a) for both \( r = 1 \) and 2 and those in (b) for \( r = 1 \) and 3 are obtained from (1.12), and the dashed curves are the low-\( k_p \) approximation obtained from (1.14). The linear bands for \( r = 2 \) in (b) are the Dirac cone. \( a_0 = \sqrt{3}a \) is the lattice constant.

For odd \( N \), we can always find a value of \( r \) such that \( E_0 = 0 \) given by (1.13), for which (1.12) gives the linear bands \( E = \pm \hbar v_F k_\parallel \). Therefore, there are \( 2(N-1) \) massive conduction and valence bands described by (1.14) and (1.15), and two linear bands defining one massless Dirac cone given by \( E = \pm \hbar v_F k_\parallel \). As an example, the band structure of AB-stacked trilayer of \( N = 3 \) is shown in Figure 1.3(b), where six bands can be seen; four are massive bands and two are linear Dirac bands.

**AA stacking order**

![Figure 1.4. Atomic structure of AA-stacked bilayer graphene.](image)

In the AA stacking order, the sublattice A of the upper layer is directly above the sublattice A of the lower layer, and the sublattice B is also directly above the sublattice B, as shown in Figure 1.4. Nearest-neighbor intralayer (\( \gamma_0^{AA} \)) and interlayer (\( \gamma_1^{AA} \)) interactions are considered. These interaction energies \( \gamma_0^{AA} \) and \( \gamma_1^{AA} \) of AA-stacked multilayer graphene are not
necessarily the same as $\gamma_0$ and $\gamma_1$ of AB-stacked multilayer graphene. It is found that the eigenenergies of the AA-stacked multilayer graphene is given by\textsuperscript{15}

\[ E = \pm \hbar v_F A A |k_\parallel| + 2\gamma_1^{AA} \cos k_z d^{AA}, \quad (1.16) \]

where the identity $\gamma_+ \sqrt{\gamma_- / \gamma_+} = \hbar v_F A A |k_\parallel|$ is used, and $k_z$ is given by

\[ k_z = \pm r \frac{\pi}{(N+1)d} \quad (1.17) \]

with $d = 0.36 \text{nm}$ being the distance between two AA-stacked graphene layers.\textsuperscript{17,18} Different from the bands of AB-stacked multilayer graphene, the energy bands of AA-stacked graphene given in (1.16) are all massless linear bands. These linear bands form Dirac cones of shifted energies.

As examples, the band structures of AA-stacked bilayer graphene ($N = 2$) and trilayer graphene ($N = 3$) are shown in Figure 1.5(a) and Figure 1.5(b), respectively, using $\gamma_0^{AA} = 2.59 \text{eV}$ and $\gamma_1^{AA} = 0.217 \text{eV}$.\textsuperscript{17,19} As can be seen, there are two bands for each value of $r$ (marked next to the curves) and a total of $2N$ bands for the system. The shift of the Dirac point is determined by the second term in (1.16). Note that each Dirac cone of a given $r$ value has its unique $k_z$ value. Therefore, unless an out-of-plane momentum is provided, carrier transitions between two different Dirac cones are forbidden.
Misorientation and other stacking orders

For chemically deposited multilayer graphene, misorientation might happen among graphene layers. When the relative rotation is small, large areas of locally AA-stacked and AB-stacked regions are formed, as shown in Figure 1.6 for misoriented bilayer graphene. In such a case, each of these regions can be locally treated as AA or AB stacking if the mean free path of the carriers is much smaller than the physical size of the region. Otherwise, one must identify the unit cell of the misoriented multilayer system and numerically calculate the band structure. It is
found that while the energy band is still linear around the K point, the Fermi velocity is greatly reduced and therefore the localization of electrons is possible in misoriented multilayer graphene.\textsuperscript{21}

Figure 1.6. Misoriented bilayer graphene with locally AA-stacked and AB-stacked regions.
Chapter 2. Electronic Properties of Monolayer and Multilayer Graphene

2.1. Elastic scattering mechanisms

Many scattering mechanisms have been suggested for the explanation of the observation in the graphene carrier-transport experiments.\textsuperscript{22-28} Among all possible scattering channels, phonon scattering is the intrinsic scattering mechanism that serves as the lower bound of the scattering rate that fundamentally limits the mobility of carriers in graphene. Two types of phonon scattering mechanisms are considered in this chapter for carrier transport in graphene: elastic acoustic phonon scattering and inelastic optical phonon scattering; the former is discussed in this section and the latter is discussed in the next section. The scattering rate arising from the longitudinal acoustic phonon scattering is given by\textsuperscript{29}

\[
\tau_{\text{ac}}^{-1}(E) = \frac{1}{\hbar^3} \frac{k_B T}{4v_F^2} \frac{D_A^2}{\rho v_{\text{ph}}^2} E, \tag{2.1}
\]

where \( E \) is the energy of charged carriers, \( \hbar \) is the reduced Planck constant, \( k_B \) is the Boltzmann constant, \( T \) is the temperature, \( v_F \) (\( \approx 10^6 \) m/s) is the Fermi velocity of carriers in graphene, \( D_A \) is the acoustic deformation potential, \( \rho = 7.6 \times 10^{-7} \) kg/m\(^2\) is the density of graphene per layer, and \( v_{\text{ph}} = 2 \times 10^4 \) m/s is the phonon velocity of the longitudinal acoustic mode.\textsuperscript{30} In this thesis, \( D_A = 18 \) eV is assumed, as measured in the experiments.\textsuperscript{24, 28, 29, 31}

The impurity scattering due to the charged impurity is also considered in the literature for carrier transport in graphene.\textsuperscript{22, 23, 25} For a monolayer graphene sandwiched between two media
of permittivities $\varepsilon_1$ and $\varepsilon_2$, the average dielectric permittivity of such a system is $\varepsilon_{\text{avg}} = (\varepsilon_1 + \varepsilon_2) / 2$. The elastic scattering rate arising from the charged impurity scattering can then be written as $^{32}$

$$
\tau^{-1}_{\text{imp}}(E) = \frac{n_i}{4\pi h} \frac{E}{(\hbar v_F)^2} \int_0^{2\pi} d\theta \frac{\tilde{V}^2}{(1 + q_s / q)^2} (1 - \cos^2 \theta),
$$

(2.2)

where $n_i$ is the impurity density, $q$ is the scattering wave number, $\tilde{V} = e^2 / 2\varepsilon_{\text{avg}}q$ is the Fourier transform of the 2D potential energy, and $q_s$ is the screening wave number. With $q = 2k \sin (\theta / 2)$, where $k$ is the wave number of carriers and $\theta$ is the scattering angle, we obtain the analytical expression for the impurity scattering rate in the limit of $k \gg q_s$:

$$
\tau^{-1}_{\text{imp}}(E) = \frac{n_i}{4\pi h} \frac{1}{E} \left( \frac{e^2}{2\varepsilon_{\text{avg}}} \right)^2.
$$

(2.3)

In fact, $q_s$ is a function of both temperature $T$ and wave number $q$. In this chapter, we consider the case of a Fermi energy $E_F \gg k_B T$ and a wave number $q \leq 2k_F$. Within this limit, $q_s$ is approximately the Thomas-Fermi screening wave number given by $e^2D(E_F) / 2\varepsilon_{\text{avg}}$, where $D(E_F) = 2E_F / \pi (\hbar v_F)^2$ given by (1.5) is the density of states at the Fermi energy. $^{32}$

Another elastic scattering mechanism considered in this chapter is the scattering from weak scatterers, whose physical origin is still debatable. Possible scattering sources are graphene ripples$^{33}$ and point defects (local substitution or dislocations in the carbon lattice).$^{34, 35}$ The resistivity $\rho_s$ contributed by weak scatterers is fairly constant, independent of the carrier density and the temperature. The scattering rate is given by
\[
\tau_s^{-1}(E) = \frac{e^2}{\pi \hbar^2 \rho_s E},
\]

(2.4)

where \( \rho_s \) is in the range from 40 \( \Omega \) to 100 \( \Omega \) obtained from experiments.\(^{26,31,36,37}\) It is also shown that the value of \( \rho_s \) is insensitive to substrates where graphene sample is deposited,\(^{31}\) suggesting that scattering off ripples might not be important. Therefore, in this chapter, \( \rho_s \) is set to be 100 \( \Omega \) as a reasonable parameter obtained from the experiments, and the same value of \( \rho_s \) is used for the calculation of carrier transport of graphene on different substrates.

2.2. Inelastic scattering mechanisms

Recent experimental studies show that the measured resistivity of graphene increases rapidly with temperature.\(^{24,26,28}\) This temperature-dependent behavior is explained by Chen \textit{et al.} considering the surface polar optical phonon modes of graphene on a SiO\(_2\) substrate.\(^{24}\) The excellent fit of their experimental data suggests that surface optical phonon scattering plays an important role in the carrier transport of graphene above 200 K. It has also been shown experimentally that scattering by surface optical phonons limits the saturation velocity in a graphene field-effect transistor.\(^{38}\) In this section, we theoretically study the carrier scattering rate arising from surface optical phonons using Rode’s iteration method. The carrier energy dependence of the scattering rate is clearly illustrated.
Table 2-1. Physical parameters of selected materials.a

<table>
<thead>
<tr>
<th>Quantity (units)</th>
<th>SiO$_2^{b,c}$</th>
<th>Al$_2$O$_3^{c}$</th>
<th>ZrO$_2^{c}$</th>
<th>HfO$_2^{c}$</th>
<th>SiC $^{c,d}$</th>
<th>h-BN$^{b,c}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\varepsilon_{\text{low}} (\varepsilon_0)$</td>
<td>3.90</td>
<td>12.53</td>
<td>24.0</td>
<td>22.0</td>
<td>9.7</td>
<td>5.09</td>
</tr>
<tr>
<td>$\varepsilon_{\text{int}} (\varepsilon_0)$</td>
<td>3.36</td>
<td>7.27</td>
<td>7.75</td>
<td>6.58</td>
<td>-</td>
<td>4.58</td>
</tr>
<tr>
<td>$\varepsilon_{\text{high}} (\varepsilon_0)$</td>
<td>2.40</td>
<td>3.20</td>
<td>4.00</td>
<td>5.03</td>
<td>6.5</td>
<td>4.10</td>
</tr>
<tr>
<td>$\hbar\omega_{\text{TO1}} (\text{meV})$</td>
<td>55.6</td>
<td>48.18</td>
<td>16.67</td>
<td>12.40</td>
<td>98.82</td>
<td>97.08</td>
</tr>
<tr>
<td>$\hbar\omega_{\text{TO2}} (\text{meV})$</td>
<td>138.10</td>
<td>71.41</td>
<td>57.70</td>
<td>48.35</td>
<td>-</td>
<td>187.22</td>
</tr>
<tr>
<td>$\hbar\omega_1 (\text{meV})$</td>
<td>58.94</td>
<td>61.63</td>
<td>28.18</td>
<td>21.60</td>
<td>118.03</td>
<td>101.42</td>
</tr>
<tr>
<td>$\hbar\omega_2 (\text{meV})$</td>
<td>156.39</td>
<td>100.20</td>
<td>76.33</td>
<td>54.21</td>
<td>-</td>
<td>195.83</td>
</tr>
<tr>
<td>$F_1^2 (\text{meV/} \varepsilon_0)$</td>
<td>0.74</td>
<td>1.45</td>
<td>1.05</td>
<td>0.96</td>
<td>2.35</td>
<td>0.76</td>
</tr>
<tr>
<td>$F_2^2 (\text{meV/} \varepsilon_0)$</td>
<td>5.06</td>
<td>5.87</td>
<td>3.27</td>
<td>0.92</td>
<td>-</td>
<td>1.65</td>
</tr>
</tbody>
</table>

*a $\varepsilon_{\text{low}}$, $\varepsilon_{\text{int}}$, and $\varepsilon_{\text{high}}$ are the low-frequency, intermediate, and high-frequency dielectric permittivities, respectively. $\hbar\omega_{\text{TO}}$ is the transverse optical phonon energy, and $\hbar\omega_i$ is the surface optical phonon energy of mode $v$. For a material with only one surface optical phonon mode, $\hbar\omega_1 = \hbar\omega_{\text{TO1}} \left[ \left( \varepsilon_{\text{low}} + \varepsilon_0 \right) / \left( \varepsilon_{\text{high}} + \varepsilon_0 \right) \right]^{0.5}$. For a material with two surface optical phonon modes, $\varepsilon_{\text{high}}$ is replaced with $\varepsilon_{\text{int}}$ in the above equation for $\hbar\omega_1$ ($\hbar\omega_2$), respectively. $F_v^2$ is the coupling parameter for the surface optical phonon mode $v$.\n
b Reference $^{40}$
c Reference $^{39}$
d Reference $^{41}$
e Reference $^{42}$

Consider a graphene monolayer placed at a distance $d$ above a substrate. The graphene monolayer is sandwiched between the substrate and the air, which has the permittivity of free space $\varepsilon_0$. For such a system, there exist surface optical phonon modes which can individually correspond to a certain bulk transverse optical (TO) phonon mode of the substrate, assuming that the coupling between different TO modes is small. Various substrate materials are studied, and their physical parameters are listed in Table 2-1. For scattering involving surface optical phonon modes of phonon energies $\hbar\omega_i$, where $v$ stands for different phonon modes, the carrier transition rate $S(k,k')$ from state $k$ to state $k'$ can be written as$^{43}$
\[
S(k,k') = \frac{2\pi}{\hbar^2} \sum_{\nu,\alpha} F_v^2 \left\{ \frac{N^\alpha e^2}{A} \left( \frac{e^{2q_d}}{q} \right) \frac{1 + s \cdot \cos \theta}{2} \delta(E - E' \mp \hbar \omega_v) \right\},
\]
(2.5)

where for the \( \pm \) and \( \mp \) signs, the positive sign has to be chosen for phonon emission and the minus sign is for phonon absorption of the scattering process; \( A \) is the area of graphene under consideration; \( \theta \) is the angle between carrier wave vectors \( k' \) and \( k \); \( E \) and \( E' \) are respectively the carrier energy before and after the collision; \( N^\alpha \) is the equilibrium phonon occupation number, where \( k_{B} \) is the Boltzmann constant and \( T \) is the temperature; \( s = 1 \) for intraband scattering and \( s = -1 \) for interband scattering; \( q \) is the scattering phonon wave number; \( q_s \) is the Thomas-Fermi screening wave number given by \( q_s = aE_F / \hbar \nu_F \), where \( E_F \) is the Fermi energy and \( a \) is a dimensionless constant given by \( a = e^2 / \epsilon_{\text{avg}} \pi \hbar \nu_F \); \( \epsilon_{\text{avg}} = (\epsilon_0 + \epsilon_{\text{low}}) / 2 \) is the average dielectric permittivity of the air/graphene/substrate system, and \( \epsilon_{\text{low}} \) is the low-frequency dielectric permittivity of the substrate. The factor \( F_v \) is the electron-phonon coupling parameter, which has the following relation:\(^{43,44}\)

\[
F_v^2 = \frac{\hbar \omega_v}{2 \left( \frac{1}{\epsilon_{\text{high}} + \epsilon_0} - \frac{1}{\epsilon_{\text{low}} + \epsilon_0} \right)},
\]
(2.6)

where \( \epsilon_{\text{high}} \) is the high-frequency dielectric permittivity of the substrate. In the case of two dominant surface optical phonon modes, as with all substrate materials except SiC considered in this chapter, it is useful to introduce an intermediate permittivity \( \epsilon_{\text{int}} \) that describes the dielectric permittivity for some intermediate frequency between the two transverse optical phonon frequencies \( \omega_{\text{TO1}} < \omega_{\text{int}} < \omega_{\text{TO2}} \).\(^{39}\) Thus, from (2.6), \( F_1^2 \) is obtained by replacing \( \epsilon_{\text{high}} \) with \( \epsilon_{\text{int}} \),
and $F_2^2$ is obtained by replacing $\epsilon_{\text{low}}$ with $\epsilon_{\text{int}}$. With $S(k,k')$ given in (2.5), it can be shown that the carrier scattering rate is\textsuperscript{45}

$$
\tau^{-1}(E) = \frac{S_0(E)}{1 + \sum_{\nu} S_{a,\nu}(E) \tau(E - \hbar \omega_{\nu}) + \sum_{\nu} S_{e,\nu}(E) \tau(E + \hbar \omega_{\nu})},
$$

where the functions $S_{a,\nu}(E)$ and $S_{e,\nu}(E)$ effectively describe the in-scattering contributions to the state $E$ from states $E - \hbar \omega_{\nu}$ by phonon absorption and from states $E + \hbar \omega_{\nu}$ by phonon emission, respectively, and $S_0(E)$ describes the out-scattering process from the state $E$. To solve (2.7), we adopt the iteration method first formulated by Rode.\textsuperscript{46} We first set $S_{a,\nu}(E)$ and $S_{e,\nu}(E)$ to zero so that in-scattering processes are ignored; then, the zeroth-order solution $\tau^{(0)} = S_0^{-1}$ can be obtained. The $\ell$th-order solution $\tau^{(\ell)}$ can be obtained by iteratively replacing $\tau$ in (2.7) with the $(\ell - 1)$th-order solution $\tau^{(\ell-1)}$ so that

$$
\tau^{(\ell)}(E) = \frac{1 + \sum_{\nu} S_{a,\nu}(E) \tau^{(\ell-1)}(E - \hbar \omega_{\nu}) + \sum_{\nu} S_{e,\nu}(E) \tau^{(\ell-1)}(E + \hbar \omega_{\nu})}{S_0(E)}.
$$

The iteration process continues until the difference between the last iterative solution and the current solution is below a certain threshold. Elastic scattering mechanisms can also be included in the iteration process by replacing $S_0(E)$ with $S_0(E) + \tau^{-1}_{\text{el}}(E)$ in (2.7), where $\tau^{-1}_{\text{el}}(E)$ is the scattering rate contributed by the elastic scattering.\textsuperscript{45} In this chapter, acoustic phonon scattering, charged impurity scattering, and scattering from short-range scatterers are considered.
Figure 2.1. Surface optical phonon scattering rates of graphene on (a) SiO$_2$, and (b) HfO$_2$ at the temperatures of 100 K (black), 200 K (blue), and 300 K (red) (from bottom to top). The dashed curves are the elastic-limit solutions of (2.9), and the solid curves are the iterative results of (2.7). The energy locations of $\hbar \omega_1$, $\hbar \omega_2$, $\mu$, $\mu + \hbar \omega_1$, and $\mu + \hbar \omega_2$ are also indicated. The Fermi energy is set at 100 meV in our numerical calculation.

The scattering rate arising from the surface optical phonon scattering for graphene on top of SiO$_2$ and HfO$_2$ substrates are plotted in Figure 2.1 as a function of the carrier energy. The physical parameters used are listed in Table 2-1. Here we choose $d = 3.4$ Å as the separation between graphene and the substrate, and the Fermi energy $E_F$ to be 100 meV, which is in the neighborhood of the experimentally observed $E_F$ values of graphene samples synthesized by chemical vapor deposition.$^{47-49}$ The corresponding chemical potential $\mu$ is a function of temperature, which is determined by the conservation of the total electron density.$^{32}$ Two sets of
data are drawn in Figure 2.1: the iterative solution (exact solution) of (2.7) (solid curves), and the elastic-limit approximation of the scattering rate (dashed curves) given by

\[ \tau^{-1}(E) = \frac{A}{4\pi^2} \int d^2k' (1-s \cdot \cos \theta) S(k, k') \],

which is a popular way to approximate the solution of (2.7) in the literature.\(^ {43, 51}\) For the elastic-limit scattering rates, two physical features can be observed: (1) The scattering rate initially drops as the carrier energy \( |E| \) increases away from the Dirac point because the contribution from the interband phonon emission decreases with decreasing density of states at \( E' \). (2) Kinks at phonon energies \( \hbar \omega_1 \) and \( \hbar \omega_2 \) can be seen in the figure, especially at low temperatures. These kinks indicate the onset of the intraband phonon emission processes, which increase the overall scattering rate. However, as can be seen in Figure 2.1, the elastic-limit approximation works better for substrates that have small surface optical phonon energies, such as HfO\(_2\) shown in Figure 2.1(b), than substrates that have large surface optical phonon energies, such as SiO\(_2\) shown in Figure 2.1(a). For the exact scattering rates (solid curves) shown in Figure 2.1, due to limited available states below the chemical potential \( \mu \) for electrons to be scattered into, the scattering rate is strongly suppressed for carrier energies around \( \mu \). This dip of the scattering rate at \( \mu \) translates into multiple dips at the energy locations \( \mu \pm c_1 \hbar \omega_1 \pm c_2 \hbar \omega_2 \), where \( c_1 \) and \( c_2 \) are integers, through the coupling among scattering rates at these energy locations via (2.7). Three dips at \( \mu, \mu + \hbar \omega_1 \), and \( \mu + \hbar \omega_2 \) are indicated in Figure 2.1(a) for graphene on the SiO\(_2\) substrate at 100 K. Also note in Figure 2.1(a) that the location of the dip for the minimum scattering rate shifts with \( \mu \), which approaches the Dirac point with increasing temperature. At low temperatures, \( \mu \) approximately equals \( E_F \), which is set at 100 meV for our numerical calculation.
Figure 2.2. Resistivity of graphene on different substrates due to the surface optical phonon scattering (a), (b) as a function of temperature with $E_F = 100$ meV, and (c), (d) as a function of Fermi energy at 300 K. All solid curves are obtained using iteration method. The dashed curves are for SiO$_2$ assuming the coupling strength $F^2$ is 7 times higher than the theoretical value in Table 2-1, and the dotted curves are for SiO$_2$ using the elastic-limit approximation of (2.11).

Once the scattering rate is obtained, the 2D conductivity can be calculated semiclassically from the relation$^{34}$

$$\sigma = -\frac{e^2}{\pi\hbar^2} \int dE \frac{E}{\tau(E)} \frac{df(E)}{dE}.$$  \hspace{1cm} (2.10)

The resistivity $\rho = \sigma^{-1}$ arising from surface optical phonon scattering for graphene on different substrates is shown in Figure 2.2(a) and (b) as a function of temperature for a fixed Fermi energy of 100 meV and in Figure 2.2(c) and (d) as a function of Fermi energy at a fixed temperature of
300 K. The resistivity of the graphene/SiO$_2$ system calculated using the elastic-limit approximation is also shown in Fig. 2. However, instead of (2.9), which is a bad approximation for SiO$_2$ as can be seen in Figure 1.1(a), we keep the Fermi function term in the approximation so that\(^{50}\)

\[
\tau(E)^{-1} = \frac{A}{4\pi^2} \int d^2 k' \frac{1 - f_0(E')}{1 - f_0(E)} (1 - s \cdot \cos \theta) S(k, k').
\] (2.11)

The resultant resistivity (dotted curves in Figure 2.2) is slightly underestimated; the maximum error, about 20\%, happens at the highest temperature, as seen in Figure 2.2(b), and around the Fermi energy, as seen in Figure 2.2(d).
Figure 2.3. Scattering rates of graphene monolayer on Al$_2$O$_3$ (left panel (a) and (c)) and SiO$_2$ (right panel (b) and (d)) at the temperatures of 100 K (top panel (a) and (b)) and 300 K (bottom panel (c) and (d)). For each figure, the solid curve is the total scattering rate calculated by the iteration method, the dotted curve is the total scattering rate using Matthiessen's rule, and the dashed curve is the elastic scattering rate contributed by the impurity scattering and the acoustic phonon scattering. The charged impurity density is chosen to be $n_i = 2 \times 10^{11}$ cm$^{-2}$, and other parameters in the calculations are the same as those of Figure 2.1.

2.3. Comparison between inelastic and elastic scattering rates

To compare the surface optical phonon scattering rate with the elastic scattering rate, we plot scattering rates for graphene monolayer on SiO$_2$ and Al$_2$O$_3$ at the temperatures of 100 K and 300 K, as shown in Figure 2.3. In these calculations, we assume a low-impurity “clean” graphene/substrate with $n_i = 2 \times 10^{11}$ cm$^{-2}$. Other parameters are the same as those used for Figure 2.1. Three curves are drawn for each figure in Figure 2.3. Both the solid curve and the
dotted curve represent the total scattering rate contributed by both the elastic scattering and the inelastic surface optical phonon scattering. The difference is that the solid curve is obtained from the exact iterative (IR) result of (2.7) by replacing $S_o(E)$ with $S_o(E) + \tau^{-1}_o(E)$, while the dotted curve is obtained using Matthiessen's rule (MR), representing the total scattering rate $\tau^{-1}_s(E) + \tau^{-1}_e(E)$ where $\tau^{-1}_s(E)$ is the iterative result of (2.7) for the surface optical phonon scattering. The dashed curve is the elastic contribution from the impurity scattering and acoustic phonon scattering. As can be observed from the figure, while the IR and MR results are similar for carrier energies near the chemical potential $\mu$ (approximately at $E_F = 100$ meV), the discrepancy becomes large when the carrier energy is away from $\mu$ or when the temperature is low. Also, both IR and MR results change dramatically from 100 K to 300 K due to the strong temperature dependence of the surface optical phonon scattering. This temperature-dependent effect is especially strong for the case of Al$_2$O$_3$ substrate shown in Figure 2.3(a) and (c).

The relative importance of the surface optical phonon scattering can also be seen in Figure 2.4, which shows the mobility of monolayer graphene on different substrates. As can be seen in Figure 2.4(a), the mobility decreases as the temperature increases due to the increasing surface optical phonon scattering rate. Although the surface optical phonon scattering is weaker at a higher Fermi energy, as shown in Figure 2.2(c) and Figure 2.2(d), the carrier density increases rapidly with increasing Fermi energy as $n = \left(\frac{E_F}{\hbar v_F}\right)^2 / \pi$, which leads to a decreasing mobility ($\propto \sigma / ne$) with increasing Fermi energy, as shown in Figure 2.4(b).

Because the value of the mobility depends on many physical parameters such as the density of impurities and defects, the Fermi energy of graphene, and the distance between
graphene and the substrate, etc., the experimentally measured values may differ from those shown in Figure 2.4. For example, it is measured to be around 25,000 cm$^2$V$^{-1}$s$^{-1}$ for a graphene sheet on a $h$-BN substrate. However, for a clean graphene sheet with negligible impurity density, the mobility is limited only by the acoustic phonon scattering; it is found that the mobility can be as high as 100,000 cm$^2$V$^{-1}$s$^{-1}$.23

![Figure 2.4](image.png)

**Figure 2.4.** Mobility of monolayer graphene on different substrates at (a) different temperatures with $E_F = 100$ meV, and (b) at 300 K with different $E_F$. Parameters of the calculations are the same as those of Figure 2.3.
2.4. Electronic Properties of AA-stacked Multilayer Graphene

Figure 2.5. From left to right: energy band diagrams of monolayer graphene, AA-stacked bilayer and AA-stacked trilayer graphene. The shaded area is the carrier filling of the lowest band for the same $\mu$. The effective chemical potentials $\mu_i = \mu - \Delta_i$ are measured from the Dirac point for each energy band. Arrows pointing up means positive values, whereas arrows pointing down means negative values. The location of the Dirac point for band $l$ is marked by $\Delta_l$.

For $N$-layer AA-stacked graphene, there are $N$ Dirac bands that are shifted in energy, as shown in Figure 2.5. The amount of the energy shift $\Delta_l$ for the $l$-th Dirac band ($l = 1, 2, 3, ..., N$) is given by $\Delta_l = 2\alpha_1 \cos(l\pi/(N+1))$,\textsuperscript{15} where $\alpha_1 = 217$ meV is the interaction energy of vertically neighboring atoms.\textsuperscript{17, 54} For an electric field that is parallel to the graphene surface, scatterings among different Dirac bands are forbidden due to the momentum conservation of carriers. Therefore, the optical conductivity of multilayer AA-stacked graphene can be calculated by the summation of the contribution from different Dirac bands. The optical conductivity per layer is given by
\[ \sigma_{\text{AA}}(\omega) = \frac{1}{N} \sum_{l=1,2,\ldots,N} \sigma_{l,\text{AA}}(\omega) = -\frac{e^2}{\pi \hbar^2 N} \sum_{l=1,2,\ldots,N} \int_{-\infty}^{\infty} dE \frac{|E|}{\tau^{-1}_l(E)} \frac{df_0(E,\mu_l)}{dE}, \]  

(2.12)

where \( \tau^{-1}_l \) is the scattering rate as a function of the carrier energy on the Dirac band \( l \), and the effective chemical potential \( \mu_l \) is given by \( \mu - \Delta_l \), as shown in Figure 2.5.

However, it is difficult to fit the experimental data with models (2.12) because the energy dependence of \( \tau^{-1}_l \) might be complicated. Therefore, we introduce the concept of the effective scattering rate \( \tau^{-1}_{\text{eff}} \); for monolayer graphene, we replace the term \( \tau(E) \) in (2.10) with \( \tau^{-1}_{\text{eff}} \) and removed it from the integral. The remaining integral is easy to evaluate, and (2.10) becomes

\[
\sigma(\omega) = \frac{e^2}{\pi \hbar^2} \frac{2k_BT}{\tau_{\text{eff}}} \ln \left[ 2 \cosh \left( \frac{\mu}{2k_BT} \right) \right].
\]  

(2.13)

By comparing (2.10) and (2.13), we can see that \( \tau^{-1}_{\text{eff}} = \tau^{-1}(\mu) \) at zero temperature because the conductivity is only contributed by carriers at the \( \mu \) level. At a nonzero temperature, \( \tau^{-1}_{\text{eff}} \) gives the effective scattering rate contributed by carriers near the \( \mu \) level. The theoretical \( \tau^{-1}_{\text{eff}} \) can be obtained by equating (2.10) and (2.13).

For AA-stacked multilayer graphene, a similar form can also be derived by replacing the term \( \tau_l(E) \) in (2.12) with \( \tau_{l,\text{eff}} \):

\[
\sigma_{\text{AA}}(\omega) = \frac{e^2}{\pi \hbar^2 N} \sum_{l=1,2,\ldots,N} \ln \left[ 2 \cosh \left( \frac{\mu_l}{2k_BT} \right) \right].
\]  

(2.14)

It can be shown that \( \tau^{-1}_{\text{eff}} = \sum_{l,\text{eff}} \sigma_{l,\text{AA}} / N \sigma_{\text{AA}} \) from (2.12) and (2.14). Therefore, \( \tau^{-1}_{\text{eff}} \) is a
weighted effective scattering rate contributed by carriers on different Dirac bands. Note that these models are based on the semiclassical Boltzmann equation. For a low Fermi energy $E_F$ (related with $\mu$ by the conservation of the total electron density\textsuperscript{32}, $E_F$ might be comparable to $\hbar \tau^{-1}_{\text{eff}}$, i.e., the Fermi wave vector is comparable to the carrier mean free path. Modification of the models might be needed to account for the enhanced quantum effects.

For AA-stacked multilayer graphene, the theoretical DC conductivity is obtained from (2.12) with $\tau^{-1}_i(E) = \tau^{-1}_{\text{ac}}(E) + \tau^{-1}_{\text{imp}}(E) + \tau^{-1}_s(E) + \tau^{-1}_{\text{app}}(E)$, where $\tau^{-1}_{\text{app}}(E)$ is the approximated solution of (2.11) for the surface optical phonon scattering rate given by\textsuperscript{56}

$$\tau^{-1}_{\text{app}}(E) = \frac{e^2}{4\hbar^2 v_F^2} \sum_{\nu, s} \frac{1}{1 - f_0(E, \mu_i)} \frac{\omega_{\nu} N v^2 e^{-2\nu q}}{q(1 + q_s / q)^2} \left(1 - f_0(E \mp \hbar \omega_{\nu} \mu)\right) \frac{E}{\hbar \omega_{\nu} + 1}. \tag{2.15}$$

This approximation is very close to the exact iteration result for most materials when the Fermi energy is above 100 meV.\textsuperscript{56} The elastic scattering rates $\tau^{-1}_{\text{ac}}(E)$, $\tau^{-1}_{\text{imp}}(E)$, and $\tau^{-1}_s(E)$ are given by (2.1), (2.2), and (2.4), respectively. These equations are still applicable to AA-stacked multilayer graphene because the band structure of AA-stacked graphene also consists of Dirac cones. However, the density of states (DOS) of $N$-layer AA-stacked multilayer graphene is different from that of monolayer graphene. For $N$-layer AA-stacked multilayer graphene, the DOS is given by

$$D(E_F) = \sum_{i=1,2,N} \frac{2 \left| E_i - \Delta \right|}{\pi \left( \hbar v_F \right)^2}, \tag{2.16}$$

as shown in Figure 2.6(a). Therefore, in (2.2) the screening wave number $q_s$ is also different for $N$-layer AA-stacked multilayer graphene.
Figure 2.6. (a) Density of states at the Fermi energy for monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene. (b) Mobilities of monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene on Al₂O₃ with \( n_i = 2.5 \times 10^{12} \text{ cm}^{-2} \) and \( T = 300 \text{ K} \). The corresponding real part of the effective scattering rate \( \tau_{\text{eff}}^{-1} \) for monolayer graphene and AA-stacked bilayer graphene is shown in (b) and (c), respectively. The areas shaded in different gray scales from bottom to top are the contributions from impurity scattering (imp), short-range weak scatterers (ws), acoustic phonon scattering (ac), and surface optical phonon scattering (sp), respectively.

The DC mobilities of monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene on Al₂O₃ at 300 K are shown in Figure 2.6(b). Samples are assumed to be dirty with \( n_i = 2.5 \times 10^{12} \text{ cm}^{-2} \). As can be seen, there is a kink at \( E_F = \Delta_1 \) for the mobility of the AA-stacked bilayer graphene. This kink can be explained by \( D(E_F) \) calculated from (2.16), as shown in Figure 2.6(a). As \( E_F \) crosses the Dirac point located at \( \Delta_1 \) (Fig. 1), \( D(E_F) \) starts to increase, resulting in a stronger screening effect \( q_s \) and weaker impurity scattering, which lead to an increased mobility. Also can be seen in Figure 2.6(b) is that the mobility of AA-stacked graphene is generally higher than that of monolayer graphene, and the highest one among the three is that of AA-stacked trilayer graphene. This is due to the strong screening of impurities in
AA-stacked multilayer graphene because of the higher density of states at the Fermi energy. By comparing Figure 2.6(c) and Figure 2.6(d), it is clear that the impurity scattering (marked “imp” in the figure) of bilayer graphene is much weaker than that of monolayer graphene. Note that in Figure 2.6, and in the figures below in this section, we only show the real part of $\tau_{\text{eff}}^{-1}$. The imaginary part of $\tau_{\text{eff}}^{-1}$ is negligibly small compared to the real part and thus is ignored in the plotting.

![Graphene Mobility and Effective Scattering Rate](image)

Figure 2.7. (a) Mobilities of monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene on Al$_2$O$_3$ with $n_i = 4 \times 10^{11}$ cm$^{-2}$ and $T = 300$ K. The corresponding real part of effective scattering rate for monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene is shown in (b), (c), and (d), respectively. The areas shaded in different gray scales from bottom to top are the contributions from impurity scattering (imp), short-range weak scatterers (ws), acoustic phonon scattering (ac), and surface phonon scattering (sp), respectively.

However, for clean samples with $n_i = 4 \times 10^{11}$ cm$^{-2}$, the situation is reversed, i.e., the mobility is lower for AA-stacked multilayer graphene, as shown in Figure 2.7(a). This can be explained by the strong scattering from weak scatterers and the high carrier density of AA-
stacked graphene. As shown in Figure 2.7, although the impurity scattering is inefficient for AA-stacked graphene, the scattering rate from weak scatterers (marked “ws” in the figure) is enhanced. Also, despite a smaller $\tau_{\text{eff}}^{-1}$ of AA-stacked multilayer graphene, the carrier density of AA-stacked graphene is much higher than that of monolayer graphene for the same $E_F$; therefore, the mobilities of AA-stacked multilayer graphene are lower than that of monolayer graphene in case of clean samples. In fact, for a higher density of weak scatters, $\tau_{\text{eff}}^{-1}$ of AA-stacked multilayer graphene can be potentially higher than that of monolayer graphene, as have been experimentally observed in our previous study.\textsuperscript{47}
Chapter 3. Terahertz Optical Properties of Monolayer and Multilayer Graphene

3.1. Drude model

In the THz spectral region, the optical conductivity of graphene is dominantly determined by the intraband carrier scattering.\(^5\) Semiclassically, the intraband conductivity of monolayer graphene can be expressed as

\[
\sigma_{\text{mono}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{2k_B T}{\tau - i\omega} \ln \left( 2\cosh \left( \frac{2}{k_B T} \right) \right),
\]

which is similar to (2.13) with \(\tau_{\text{eff}}^{-1}\) replaced by \(\tau^{-1} - i\omega\); the subscript \(\text{eff}\) is dropped for simplicity. Equation (1) can also describe the optical conductivity of misoriented (turbostratic) multilayer graphene, which is frequently observed in chemical vapor deposition (CVD) synthesis.\(^5, 5\) The energy dispersion of misoriented multilayer graphene is linear near the Dirac point, like that of monolayer graphene, but with a reduced Fermi velocity.\(^2\) Because (3.1) is not a function of the Fermi velocity, it can also be used to calculate the optical conductivity per layer of misoriented multilayer graphene.

Similarly, the optical conductivity per layer of AA-stacked multilayer graphene can be expressed as the average of the contribution from \(N\) different Dirac bands:

\[
\sigma_{\text{AA}}(\omega) = \frac{e^2}{\pi \hbar^2 N} \frac{2k_B T}{\tau - i\omega} \sum_{r=1,2,N} \ln \left( 2\cosh \left( \frac{\mu + 2\alpha \cos \left( \frac{r\pi}{N+1} \right)}{2k_B T} \right) \right),
\]

which is similar to (2.14) with \(\tau_{\text{eff}}^{-1}\) replaced by \(\tau^{-1} - i\omega\); again, the subscript \(\text{eff}\) is dropped for simplicity.
simplicity. It is also possible that a multilayer graphene consists of misoriented layers as well as layers that are AA or AB stacked. For example, it has been suggested that for ABA’ graphene (an AB-stacked bilayer plus a misoriented monolayer), the Raman spectrum is the combination of the spectra from an AB bilayer and a misoriented monolayer,\(^59\) and its band structure is a superposition of that of the AB-stacked bilayer and a linear dispersion band.\(^60\) The superposition of band structures provides us with the convenience in calculating the optical conductivity of such a mixed system of different stacking schemes. In this case, the optical conductivity of ABA’ graphene is the sum of the optical conductivity of an AB-stacked bilayer and that of a misoriented monolayer. Here we generalize this conclusion to multilayer graphene comprising AA-stacked and misoriented layers as supported by the fitting results from our experimentally measured data described below. For example, from Eqs. (3.1) and (3.2), the optical conductivity of an AAA’ graphene trilayer, which comprises one AA-stacked bilayer and one misoriented monolayer, is \(\sigma_{\text{AAA'}} = 2 \times \sigma_{\text{AA}}|_{N=2} + \sigma_{\text{mono}}\). Hereafter, for the ease of reference, we shall call (3.1) the graphene or misoriented model, (3.2) the AA model, and any combination of them the mixed model.

3.2. Experimental studies

Sample preparation

Our graphene samples were synthesized by CVD on a copper substrate. By carefully controlling the airflow of a mixture of methane and hydrogen in a heated furnace, monolayer graphene was uniformly grown on a copper substrate. The samples were then spin coated with poly(methyl methacrylate) (PMMA). The PMMA/graphene can be detached from the copper
substrate by etching the copper with an aqueous nitric acid solution. The detached PMMA/graphene was then transferred and deposited on the sapphire substrate by direct contact. The PMMA was dissolved subsequently, leaving monolayer graphene on our target substrate. PMMA residues on the sample were then eliminated by the annealing process. $N$-layer graphene samples were obtained by repeating the aforementioned process $N$ times on the same sapphire substrate. For this work, we used five graphene samples on sapphire substrate with $N = 1, 2, 3, 5$ and 7, respectively. The number of layers in each sample was confirmed by the quantized absorption level of each sample seen in the optical transmission measurement using broadband visible light, as shown in Figure 3.1. The quantized absorption level can be tuned by varying the synthesis condition in CVD process; for this reason, different values are observed by different groups.\textsuperscript{61,62}

![Figure 3.1. Transmittance (%) (measured at the wavelength of 600 nm) of samples with different numbers of graphene layers.](image)

**Raman spectroscopy**

The Raman measurements were performed at room temperature under ambient conditions with a laser of 488 nm excitation wavelength and 5 mW power. As shown in Figure 3.2, every one of the Raman G’-band spectra of our multilayer samples can be described by a single Lorentzian shape, which excludes the possibility of any of our samples being AB-stacked.\textsuperscript{63}
Furthermore, except for the bilayer sample, the other multilayer samples \((N = 3, 5, 7)\) all show a \(G'\) blueshift with respect to that of the monolayer graphene (Table 3-1). This indicates that the Fermi velocity is renormalized and some layers, if not all, are misoriented.\(^{59, 64}\) The \(G'\)-band intensity, shown in Figure 3.2 inset, is greatly influenced by the number of layers and the rotational angle of misorientation. It has been shown theoretically and experimentally that for misoriented bilayer graphene, the integrated intensity of \(G'\)-band increases with increasing rotational angle.\(^{65, 66}\) The integrated intensity can even be higher than twice that of monolayer graphene.\(^{65}\) If this trend also applies to multilayer graphene with \(N > 2\), the average rotational angle among layers in our trilayer graphene sample should be very small because of the ultralow \(G'\)-peak intensity and the large full width at half-maximum (FWHM) of the \(G'\)-band spectrum observed for this sample (Table 3-1), which are both signatures of a small rotational angle.\(^{65}\) For our 5- and 7-layer graphene samples, the peak intensity of the 7-layer sample is around 7 times that of monolayer graphene while that of the 5-layer sample is only about 3 times that of monolayer graphene. Therefore, we conclude that the average rotational angle among layers in the 7-layer graphene sample is larger than that in the 5-layer graphene sample. For our bilayer graphene sample, we believe that it has AA stacking because its Raman spectrum has no \(G'\) blueshift and its \(G'\)-peak intensity is similar to that of the monolayer sample (Figure 3.2 inset). The small redshift might be caused by the finite amount of charged impurity.\(^{67}\)

<table>
<thead>
<tr>
<th>Number of layers</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>5</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>(G') shift (cm(^{-1}))</td>
<td>-3</td>
<td>6</td>
<td>15</td>
<td>15</td>
<td>6</td>
</tr>
<tr>
<td>(G') FWHM (cm(^{-1}))</td>
<td>40</td>
<td>37</td>
<td>48</td>
<td>45</td>
<td>43</td>
</tr>
</tbody>
</table>
Figure 3.2. Raman $G'$-band spectra of monolayer ($N = 1$) and multilayer graphene with different numbers of layers ($N = 2, 3, 5, 7$). From left to right, as the arrow indicates, are curves for 2-, 1-, 7-, 3- and 5-layer graphene samples. The intensity is scaled to the same level to show the shifts of curves. In the inset are the absolute Raman scattering intensities of the same data. Curves associated with different numbers of layers are indicated by the arrow. Corresponding curves are shown in the same color.

**Terahertz time-domain spectroscopy**

To gain more physical details about our samples, THz-TDS is used to measure the THz optical conductivity. The THz-TDS experiments are briefly described in the following. The generation and detection of THz pulsed radiation using the free-space electro-optic sampling technique was set up on a mode-locked Ti:Sapphire laser operating at 800 nm wavelength with 30-fs pulses at an 80 MHz repetition rate. The femtosecond-pulse laser beam was split into a pump beam and a probe beam. The THz pulses were generated by using the pump laser pulses to trigger a semi-insulating-InP photoconductive switch that consists of two parallel electrodes spaced 0.5 mm apart and biased with a DC voltage of 100 V. The generated THz-pulse beam was focused by a pair of off-axis parabolic mirrors onto a graphene sample at normal incidence with the incident THz electric field polarized parallel to the sample surface. The temporal electric-field-amplitude profile of an incident or transmitted THz pulse was sampled by scanning
the delay between the pump and probe optical pulses. For this sampling measurement, the incident or transmitted THz pulse being measured was focused by another pair of off-axis parabolic mirrors on a 1-mm-thick (110)-ZnTe sensor crystal. The polarization of the optical probe beam changes when the ZnTe crystal was irradiated by the THz electric field. The THz-induced phase retardation of the optical probe beam was converted into an intensity modulation and detected by using balanced photodiodes. In order to avoid any undesirable effects from the humidity in the laboratory air, the THz-TDS system was placed in a Plexiglas box and was purged with dry nitrogen flow. The measured time-domain profile of the incident THz electric field amplitude is shown by the gray curve (labeled $E_i$) in Figure 3.3(a).

All samples were measured at the room temperature of 294 K. For each sample before graphene was placed on the substrate, the time-domain profile of the transmitted electric field amplitude of the THz pulse through the bare sapphire substrate was first measured as the background reference. After each sample with graphene on the sapphire substrate was made, the time-domain profile of the transmitted electric field amplitude of the THz pulse through the sample was measured. These time-domain data are shown in Figure 3.3(a). As can be seen, the transmitted signal is reduced as the number of layers increases. The spectra of the transmission coefficients in the frequency domain were obtained by taking the Fourier transform on the data in Figure 3.3(a). The derived data are complex numbers, carrying both the phase and amplitude information. Using Maxwell’s equations with appropriate boundary conditions, the ratio of the transmission coefficients $t_{sam}$ (graphene sample on the sapphire substrate) to $t_{sub}$ (bare sapphire substrate) in the frequency domain can be related to the optical conductivity of the graphene sample without the substrate as
\[ \frac{t_{\text{sam}}(\omega)}{t_{\text{sub}}(\omega)} = \frac{1+n}{1+n+N\sigma(\omega)/c\varepsilon_0}, \]

where \( n = 3.07 \) is the refractive index of the sapphire substrate, which was measured in our experiments and found to be consistent with literature; \( c \) is the speed of light; \( \varepsilon_0 \) is the permittivity of free space; and \( \sigma(\omega) \) is the measured optical conductivity per layer of the sample.

Figure 3.3. (a) Time-domain profiles of the incident electric field amplitude \( (E_i) \) and the transmitted electric field amplitudes \( (E_t) \) of THz pulses for the sapphire substrate and the graphene samples with different number of layers. \( E_i \) is obtained by measuring the transmitted signal through the air without the substrate in the optical path. The framed region of the transmitted profiles for \( E_t \) is enlarged and shown in the inset. Indicated by the arrow from top to bottom are the data for bare sapphire substrate \( (N=0) \) and for graphene samples with \( N = 1, 2, 3, 5, 7 \) number of layers. (b, c) Measured optical conductivity spectra (solid curves) and theoretical optical conductivity spectra (dashed curves) for (b) monolayer \( (N=1) \) and bilayer graphene \( (N=2) \), and (c) multilayer graphene \( (N=3, 5, 7) \). Different models are used for fitting the experimental data of different samples (see text). The fitting values for the parameter \( \tau^{-1} \) are presented in Table 3-2A.
Table 3-2. Fitting results of scattering rate $\tau^{-1}$ for graphene samples of different number of layers.

<table>
<thead>
<tr>
<th></th>
<th>Number of layers</th>
<th></th>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>1</td>
<td>2</td>
<td>3</td>
<td>5</td>
</tr>
<tr>
<td>A</td>
<td>$\mu$ (meV)</td>
<td>64</td>
<td>64</td>
<td>64</td>
<td>63</td>
</tr>
<tr>
<td></td>
<td>$\tau^{-1}$ ($10^{12}$ s$^{-1}$)</td>
<td>9.9</td>
<td>29.2</td>
<td>40.1</td>
<td>18</td>
</tr>
<tr>
<td>B</td>
<td>$\mu$ (meV)</td>
<td>91.4</td>
<td>190.9</td>
<td>248.1</td>
<td>70.3</td>
</tr>
<tr>
<td></td>
<td>$\tau^{-1}$ ($10^{12}$ s$^{-1}$)</td>
<td>14.7</td>
<td>25.4</td>
<td>44.5</td>
<td>20.3</td>
</tr>
</tbody>
</table>

A: Fitting results for $\tau^{-1}$ based on the assumption of possible AA stacking with experimentally measured values of chemical potential $\mu$ obtained by the pump-probe experiment.

B: Fitting results for $\mu$ and $\tau^{-1}$ based on the assumption that all samples are decoupled graphene layers.

**Ultrafast optical pump mid-infrared probe experiment**

We also experimentally obtained the chemical potential $\mu$ for each sample by using the ultrafast optical pump mid-infrared probe technique.$^{69, 70}$ Due to the redistribution around $\mu$ of the carriers excited by the optical pump pulse, the reflectivity of the sample increases ($\Delta R > 0$) for a probe photon energy higher than $\mu$ and decreases ($\Delta R < 0$) for a probe photon energy lower than $\mu$. By finding the sign change of $\Delta R$ using different wavelengths of the mid-infrared probe beam, the location of $\mu$ can be found. For all samples, $\mu$ was found to be 63 or 64 meV (Table 3-2A). The details of the measurement are beyond the scope of this chapter.
Figure 3.4. (a) Misoriented bilayer graphene with locally AA-stacked region illuminated by the incident THz pulse beam. (b) Schematically drawing of the transmission measurement without ($t_{\text{sub}}$) and with ($t_{\text{sam}}$) a graphene sample on a sapphire substrate. (c) (From left to right) Filling of the lowest band by carriers in monolayer, AA-stacked bilayer, and AA-stacked trilayer graphene with the same chemical potential $\mu$.

3.3. Analysis and discussions

With both $\mu$ and $\sigma(\omega)$ experimentally determined for each sample, we can now determine the scattering rate $\tau^{-1}$ and the stacking arrangement of each sample by fitting the real part of $\sigma(\omega)$ to the real part of our theoretical conductivity models using $\tau^{-1}$ as the fitting parameter. The fitting curves are shown in Figure 3.3b and Figure 3.3c. The measured values of $\mu$ and the fitting values of $\tau^{-1}$ for these fitting curves are listed in Table 3-2A. We find that the monolayer and 5-layer graphene samples fit well with the graphene/misoriented model. We have also tried fitting the data with the mixed model to see if there are AA-stacked layers in the 5-layer sample. It is found that the theoretical optical conductivity is too high for the experimental data to be fitted with this model. For the bilayer and trilayer samples, the experimental data fit well with the AA model with an interaction energy of $\alpha_1 = 217$ meV, which is consistent with the theoretical value of $\alpha_1$ reported in the literature.\(^{17, 54}\) Note that a different theoretical value of $\alpha_1$ (361 meV) is
also suggested in the literature,\textsuperscript{71} which does not fit with our experimental data. This fitting result suggests that there are locally AA-stacked regions in our misoriented trilayer sample. The local existence of AA stacking is possible when layers are misoriented, as illustrated in Figure 3.4(a). As suggested by its Raman spectrum in Figure 3.2, our trilayer sample is misoriented with small rotational angles. Because the size of the local AA region in a unit cell increases rapidly with a rotational angle smaller than $5^\circ$,\textsuperscript{21,72} we can potentially have an AA region that has a dimension comparable to the mean free path of carriers in the resultant supercell if the angle is sufficiently small. The Fermi velocity of AA-stacked trilayer graphene is about $8.37 \times 10^5$ m/s.\textsuperscript{17} With a carrier lifetime of $\tau = 25$ fs that is obtained by fitting with the AA model, the mean free path is 20 nm, which is around one hundred lattice constants. Because the number of carbon atoms in a supercell can easily reach thousands,\textsuperscript{72} it is possible that our conductivity data are the direct measurement of locally AA-stacked regions in the sample. For the 7-layer sample, $\sigma(\omega)$ fits well with a mixed model of 5 misoriented layers plus an AA-stacked bilayer. By comparison, the existence of AA-stacking coupling is difficult to tell from its Raman spectrum because it is overwhelmed by the Raman intensity contributed by the misoriented layers.

To further show the existence of AA stacking in our samples, we also fit the experimental data using (3.1) by assuming that all the samples have decoupled graphene layers, with both $\tau^{-1}$ and $\mu$ as fitting parameters. The fitting results are listed in Table 3-2B, which show a strong disagreement with the values of $\mu$ that were experimentally obtained by the pump-probe experiment. The large discrepancies in these fitting values of $\mu$ with its experimentally measured values cannot be explained by the inhomogeneity in the graphene layers or in the impurity density. Clearly, the assumption of decoupled graphene layers is not valid for all of our multilayer samples.
Table 3-3. Derived physical parameters of graphene samples based on the parameters listed in Table 3-2A.

<table>
<thead>
<tr>
<th>Number of layers</th>
<th>Relaxation time $\tau$ (fs)</th>
<th>Number of layers</th>
<th>Carrier density ($10^{12}$ cm$^{-2}$/layer)</th>
<th>DC conductivity ($10^{3}$ Ω$^{-1}$/layer)</th>
<th>Mobility (cm$^2$ V$^{-1}$ s$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>101</td>
<td>2</td>
<td>34.2</td>
<td>0.44</td>
<td>8.04</td>
</tr>
<tr>
<td></td>
<td></td>
<td>3</td>
<td>24.9</td>
<td>5.58</td>
<td>8.82</td>
</tr>
<tr>
<td></td>
<td></td>
<td>5</td>
<td>55.6</td>
<td>7.22</td>
<td>6.33</td>
</tr>
<tr>
<td></td>
<td></td>
<td>7</td>
<td>41.5</td>
<td>0.48</td>
<td>4.25</td>
</tr>
</tbody>
</table>

Other physical parameters can also be derived using the values of $\tau^{-1}$ and $\mu$ listed in Table 3-2A; the results are listed in Table 3-3. With the Fermi velocity of monolayer $v_{F_{\text{mono}}} \equiv 10^6$ m/s,$^{16}$ the mobility obtained for the monolayer graphene sample is very high, around 11420 cm$^2$V$^{-1}$s$^{-1}$. The mobility in a practical monolayer device might be much lower than this value as we have to consider the contact potential or ineffective gate coupling.$^{34,58}$ For AA-stacked bilayer and trilayer graphene samples, the carrier density can be calculated with known band overlapping and $v_f \equiv 8.37 \times 10^5$ m/s.$^{17}$ The resultant mobilities for our bilayer and trilayer graphene samples as listed in Table 3-3 are below 1000 cm$^2$V$^{-1}$s$^{-1}$, which are much lower than that of the monolayer sample. Also note that, as seen in Figure 3.3(b) and Figure 3.3(c), their optical conductivities are relatively high among the samples due to the semimetal nature of AA stacking. For the 5-layer sample, the average rotational angle is about 10° judging from its large blueshift of the $G'$ peak.$^{65}$ The corresponding Fermi velocity is $0.95v_{F_{\text{mono}}}$,$^{21}$ and the resultant carrier density and mobility are obtained and listed in Table 3-3. For the 7-layer sample, its rotational angle is difficult to tell from its $G'$ -peak shift because the observed shift corresponds to many possible angles.$^{65}$ However, because the peak intensity is almost 7 times that of monolayer graphene, we can confidently assume that the rotational angle is sufficiently large that the Fermi velocity is the same as $v_{F_{\text{mono}}}$.$^{21}$ The obtained mobility is lower than that of the 5-layer.
sample due to the existence of AA coupling in two of the layers in this 7-layer sample. From Table 3-3 we also notice that the scattering rate is relatively high for the bilayer and trilayer samples, which can be explained by the dependence of the scattering rate on the carrier density. For the same chemical potential, the carrier density is different in different subbands, as shown in Figure 3.4(c) for the lowest conduction band. As can be seen, the carrier density is especially high in AA-stacked samples due to the large overlapping of bands. Because the scattering rate of short-range scatters is proportional to the square root of the carrier density \( \tau^{-1} \propto \sqrt{n} \), our data indicate that short-range scatters are the dominant source of scattering in our bilayer and trilayer samples. Due to the AA-stacking coupling of two layers as suggested by the fitting result, our 7-layer sample also has a relatively high scattering rate. The sources of short-range scatters can be many, which cannot be identified from our experimental data.

To further confirm our fitting results, we calculated the refractive index

\[
n(\omega) = \left[ \left( \epsilon_\infty + i \sigma(\omega)/\omega d \right)/\epsilon_0 \right]^{1/2},
\]

where \( d \) is the thickness per layer, \( \sigma(\omega) \) is obtained from the experiment as in (3.3), and \( \epsilon_\infty \) is contributed by the interband transitions, which is negligibly small compared to the other term for the THz frequency region.\(^{73}\) We choose \( d = 3.6 \) Å for the bilayer and trilayer graphene samples (AA stacking),\(^{18}\) and \( 3.4 \) Å for the 5- and 7-layer graphene samples (misoriented).\(^{16}\) The choice of 3.4 Å is an approximation as we are not able to accurately determine the rotational angles among the layers in the 5-layer sample and in the 7-layer sample. Figure 5 shows, for each sample, the resultant refractive index that is deduced from the experimental data, plotted along with the theoretically calculated refractive index that is obtained using the appropriate model determined above for each sample. The values of the fitting
parameter $\tau^{-1}$ listed in Table 3-2A are used for these theoretical calculations. As can be seen in Figure 3.5, the experimental results can be fitted quite well by the theoretical models.

![Graph showing measured and theoretically calculated refractive index](image)

Figure 3.5. Measured refractive index (solid curves) and theoretically calculated refractive index (dashed curves) using a proper model for each sample (see text). $n'$ and $n''$ are the real and imaginary parts of the refractive index, respectively. From (a) to (e) are the data for samples of 1, 2, 3, 5 and 7 layers, respectively. Values of the fitting parameter $\tau^{-1}$ are listed in Table 3-2A.
Chapter 4. Plasmonic Properties of Graphene

4.1. Numerical model

Plasmons are the quantized collective oscillation of electrons that can exist on the surface or in the bulk of a three-dimensional material. Because graphene is a two-dimensional material, there is only a surface plasmon mode of graphene, in contrast to the metallic materials where both surface and bulk plasmon modes exist. To solve the characteristic dispersion curve of the surface plasmon mode, we consider a sheet of graphene located at a distance \( d_2 \) above a substrate that has a thickness of \( d_1 \), as shown in Figure 4.1. Permittivities in different regions are marked by \( \varepsilon_i \) \((i = 1, 2, 3)\). For our model to be applicable to different situations, the substrate can be either metallic with finite plasma frequency \( \omega_p \), or a dielectric of \( \omega_p = 0 \). Thus, the permittivity of the substrate can be modeled as \( \varepsilon_p = \varepsilon_\infty(1 - \omega_p^2 / \omega^2) \), where \( \varepsilon_\infty \) is the permittivity of the metallic substrate in the limit of the angular frequency \( \omega \to \infty \), or the constant permittivity of the dielectric substrate of \( \omega_p = 0 \). For simplicity, we set \( \varepsilon_\infty \) to be the permittivity of free space, i.e., \( \varepsilon_\infty = \varepsilon_0 \). By solving the Maxwell equations with appropriate boundary conditions, the characteristic equation to be solved for the dispersion relations of the coupled surface plasmons can be found:

\[
\frac{\gamma_2 \gamma_p \varepsilon_1 + \gamma_2^2 \varepsilon_p \tanh \gamma_p d_1}{\varepsilon_2 \gamma_2^2 \Gamma + \gamma_2^2 \varepsilon_2 \tanh \gamma_2 d_2} + \frac{\gamma_1 \gamma_p \varepsilon_1 + \gamma_1^2 \varepsilon_p \tanh \gamma_p d_1}{\gamma_3 \varepsilon_p \Gamma + \gamma_2 \varepsilon_p \tanh \gamma_2 d_2} = 0 \tag{4.1}
\]

where \( \Gamma = \varepsilon_3 / \varepsilon_2 + i \sigma q / \omega \varepsilon_2 \), \( q^2 - \gamma_i^2 = \varepsilon_i \omega^2 / \varepsilon_0 c^2 \) \((i = 1, 2, 3, p)\), \( c \) is the speed of light, and \( \sigma \) is the optical conductivity of the graphene sheet.
\[ \sigma(q, \omega) = -i \omega \frac{\varepsilon^2}{q^2} \Pi(q, \omega), \]  

where the polarizability \( \Pi(q, \omega) \) can be obtained analytically within random phase approximation (RPA) at zero temperature.\(^{13, 75}\) In this chapter, we assume that \( \text{Re} \, \Pi \) is much larger than \( \text{Im} \, \Pi \) so that the imaginary part of \( q \) can be ignored.\(^{76}\) Therefore, the surface plasmon dispersion characteristics inside the interband or intraband damping regions as shown in the figures of this chapter can only be regarded as qualitative results that ignore the damping effect.

![Figure 4.1. General schematic of the structures considered in this chapter.](image)

The solutions given by (4.1) represent surface plasmon polariton (SPP) modes that have fields confined on the graphene surface or on the surfaces of the substrate if the substrate is metallic. In contrast to the SPP mode, a surface plasmon (SP) mode of graphene is a solution of (4.1) under the quasistatic approximation, namely a solution only valid in the non-retarded region. The quasistatic approximation is suitable when the propagation of field is instantaneous in the physical region under consideration.\(^{77}\) This means that the speed of light \( c \) is much larger...
than the phase velocity \( \omega/q \) of the electromagnetic field so that the effect of time retardation can be ignored. As we shall see, in most cases considered in the literature, the phase velocity \( \omega/q \) of the SPP mode is in the order of the Fermi velocity \( v_F \) of graphene. Then, because \( c \gg v_F \), the SPP mode is essentially decoupled from the photon, and thus is better referred to as the SP mode; the term “polariton” is dropped from SPP because the coupling with photon is essentially non-existent. In our current case, the SP modes can be found from (4.1) by assuming \( c \gg \omega/q \). The retardation effect and its influence on our analytical results are later discussed. In this approximation, Eq. (4.1) can be simplified:

\[
\frac{\epsilon_1 + \epsilon_\text{p} \tanh q d_1}{\epsilon_\text{p} + \epsilon_1 \tanh q d_1} + e \frac{\epsilon_0 + \epsilon_\text{p} \tanh q d_2}{1 + \epsilon_\text{p} \tanh q d_2} = 0. \tag{4.3}
\]

As an example, the numerical solutions of Eq. (4.3) are plotted in Figure 4.2(a) for the coupled plasmon modes of graphene that is 30 nm away from a doped semiconductor thin film with other parameters specified in the caption. Three coupled plasmon modes \( \omega_{\text{sp}1}, \omega_{\text{sp}2} \) and \( \omega_g \) are found with \( \omega_{\text{sp}1} > \omega_{\text{sp}2} > \omega_g \) (thick solid curves). The decoupled modes are also plotted as dashed curves by setting large distances between different layers of the structure \( (d_1 = d_2 = 5 \mu\text{m}) \) in (4.3); these modes are decoupled as long as \( q^{-1} \ll d_{1,2} \). Two dashed lines, approximately \( \omega_\text{p}\sqrt{\epsilon_\text{p}/(\epsilon_0 + \epsilon_2)} \) and \( \omega_\text{p}\sqrt{\epsilon_\text{p}/(\epsilon_0 + \epsilon_1)} \), are the surface plasmon modes associated with the top and bottom surfaces of the semiconductor substrate. Another rising dashed curve is the graphene plasmon mode, which in the long-wavelength limit \( (q \to 0) \) is given by

\[
\omega_{\text{mono}} = \sqrt{e^2 E_F q / \pi (\epsilon_2 + \epsilon_3) \hbar^2}, \]

where \( E_F \) is the Fermi energy of graphene. When \( d_{1,2} \gg q^{-1} \), surface plasmon fields on the graphene sheet and on the semiconductor surfaces can hardly “see”
each other; in this situation, the plasmon modes are weakly coupled, following the dashed curves in the high-\( q \) region. By contrast, the plasmon modes are strongly coupled in the limit \( q \to 0 \) when retardation is neglected, resulting in the solid dispersion curves that significantly deviate from the dashed curves. This situation is similar to the case of planar metallic waveguides where the surface plasmon mode on each surface are strongly coupled when \( q \to 0 \).\cite{78, 79}
Figure 4.2. (a) Coupled plasmon modes ($\omega_{sp1}$, $\omega_{sp2}$ and $\omega_g$, solid curves) of graphene 30 nm away from a semiconductor thin film of 50 nm thickness. The parameters used are $\epsilon_1 = 3.9\epsilon_0$, $\epsilon_2 = 2\epsilon_0$, $\epsilon_3 = \epsilon_0$, $E_F = 100$ meV for graphene, and $\hbar\omega_p = 200$ meV for the doped semiconductor thin film. Decoupled modes, calculated with $d_1 = d_2 = 5\mu$m, are also plotted as dashed curves. Thin curves in the low-$q$ region are the low-$q$ approximation for each mode given by (4.7-9) and (4.11). The dotted curve represents the plasmon mode $\omega_g$ when the substrate is changed to a metal of $\hbar\omega_p = 9$ eV with other parameters unchanged. The red dots mark the modes at $q = 0.36k_F$ and $0.6k_F$ near the gap opening. The crossing of the $\omega_{sp2}$ and $\omega_g$ modes around $q = 0.36k_F$ is enlarged in the inset. Colored areas are interband and intraband damping regions.

(b) Coupled plasmon modes ($\omega_{sp1}$ and $\omega_g$, solid curves) of graphene 30 nm away from a semi-infinite semiconductor substrate ($d_1 \rightarrow \infty$). Other parameters are the same as those used in (a). Decoupled modes, calculated with $d_2 = 5\mu$m, are also plotted as dashed curves. Thin curves in the low-$q$ region are the low-$q$ approximation for each mode given by (4.9), (4.12) and (4.13). Inset: $\omega_g (q \rightarrow 0) = qrv_F$ (black circles) and Eq. (29) of Ref. $^{80}$ (red circles) are plotted along with the numerical result (solid curve) with $d_2 = 4$ nm while other parameters remain unchanged.
4.2. Analytical models

The coupled plasmon modes $\omega_{sp1}$, $\omega_{sp2}$ and $\omega_g$ can be analytically obtained in the long-wavelength limit. By plugging (4.2) in (4.3) and keeping only the constant $q^0$ term and the first-order $q^1$ term, we obtain

$$
\epsilon_p \epsilon_2 (\epsilon_1 + \epsilon_3) + (\epsilon_1 \epsilon_3 + \epsilon_2^2) \epsilon_p q d_2 + (\epsilon_1 \epsilon_3 + \epsilon_2^2) \epsilon_2 q d_1 + G(q \rightarrow 0, \omega) = 0,
$$

(4.4)

where $G$ is the contribution from the graphene sheet:

$$
G(q \rightarrow 0, \omega) = \left[ \epsilon_2 \epsilon_p + \epsilon_1 \epsilon_3 (\epsilon_1 d_1 + \epsilon_p d_2) \right] \frac{\epsilon^2}{q} \text{Re} \Pi(q \rightarrow 0, \omega),
$$

(4.5)

and

$$
\frac{\epsilon^2}{q} \text{Re} \Pi(q \rightarrow 0, \omega) = -\frac{\omega_{mono}^2}{\omega^2} \left( \epsilon_2 + \epsilon_3 \right) \left( \frac{\hbar \omega}{4 E_F} \ln \left[ \frac{2 E_F - \hbar \omega}{2 E_F + \hbar \omega} \right] + 1 \right).
$$

(4.6)

Note that the singularity at $\hbar \omega = 2|E_F|$ in Eq. (4.6) is removed for finite temperature. As shown in Figure 4.2(a), $\omega_{sp1}(q \rightarrow 0) \equiv \omega_p \gg \omega_{mono}$ and $G(q \rightarrow 0, \omega_{sp1})$ is much smaller than the other terms in Eq. (4.4); therefore, $\omega_{sp1}(q \rightarrow 0)$ manifests the strongly coupled mode between the two surfaces of the plasma substrate. Dropping the $G$ term in (4.4), we obtain

$$
\omega_{sp1}(q \rightarrow 0) = \omega_p \left( 1 - \frac{\epsilon_2 q d_1}{2 \epsilon_0 (\epsilon_1 + \epsilon_3)} \right).
$$

(4.7)

Interesting enough, $\omega_{sp1}(q \rightarrow 0)$ is independent of $\epsilon_2$, due to the fact that the plasmonic field penetrates deeply into the regions of $\epsilon_1$ and $\epsilon_3$ dielectrics when $q d_2 = 1$ (recall that $d_2$ is the thickness of $\epsilon_2$ dielectric). By contrast, $\omega_{sp2}$ approaches zero as $q \rightarrow 0$, as shown in Figure
4.2(a); therefore, the $G$ term in (4.4) cannot be ignored in this case. However, because 
\[
\hbar \omega_{q2} (q \to 0) \ll 2 |E_F|,
\]
(4.6) can be simplified by taking \( \ln \left| (2E_F - \hbar \omega) / (2E_F + \hbar \omega) \right| = 0 \).

Plugging (4.5) and the simplified (4.6) in (4.4), we obtain
\[
\omega_{q2}^2 (q \to 0) = \frac{\epsilon_2 - \epsilon_1}{\epsilon_3 + \epsilon_1} \omega_{p}^2 + \frac{(\epsilon_2 + \epsilon_3)}{(\epsilon_3 + \epsilon_1)} \omega_{\text{mono}}^2.
\]

For the \( \omega_g (q \to 0) \) mode, the analytical solution cannot be obtained by taking \( q \to 0 \) while assuming a constant \( \omega \) in \( \Pi (q \to 0, \omega) \) for (4.6) because the group velocity \( d\omega_g / dq \) will diverge in this approach while \( d\omega_g / dq \) should be a constant as shown in Figure 4.2(a) for \( \omega_g (q \to 0) \). This difficulty can be resolved by assuming
\[
\omega_g (q \to 0) = q r \nu_F,
\]
and solving for \( r \). Instead of (4.6), the polarizability can be found as
\[
\lim_{q \to 0} \Pi (q, \omega) = \lim_{q \to 0} \Pi (q, q r \nu_F) = \frac{2E_F}{\pi (\hbar \nu_F)^2} \left( 1 - \frac{r}{\sqrt{r^2 - 1}} \right).
\]

Plug (4.5) and (4.10) in (4.4), and find the coefficient for the lowest order of \( q \); this coefficient should vanish as \( q \to 0 \). Hence, we obtain the equation
\[
1 + \frac{2 e^2 E_F}{\pi \hbar^2} \left( 1 - \frac{r}{\sqrt{r^2 - 1}} \right) \left[ \frac{d_2}{\nu_F^2 \epsilon_2} - \frac{r^2}{\epsilon_0 \omega_p^2 d_1} \right] = 0,
\]
from which \( r \) can be obtained numerically. From (4.9) and (4.11), we see that \( \omega_g (q \to 0) \) is independent of \( \epsilon_1 \) and \( \epsilon_3 \), due to the fact that \( \omega_g (q \to 0) \) manifests the strong coupling between graphene and the substrate with a plasmonic field that is highly confined within the \( \epsilon_2 \) region. In
the case of a metal substrate where $h\omega_p$ are generally much larger than $E_F$, (4.11) can be simplified by dropping the $r^2$ term; then, the root can be found analytically:

$$r = \frac{1 + \Lambda}{\sqrt{1 + 2\Lambda}}$$

with

$$\Lambda = \frac{2e^2 E_F d_2}{\pi \hbar^2 v_F \epsilon_2^2}.$$  \hspace{1cm} (4.12)

The analytical results of (4.7), (4.8), and (4.9) for the $\omega_{sp1}, \omega_{sp2}$ and $\omega_g$ modes, respectively, in the limit of $q \to 0$ are plotted in Figure 4.2(a) as thin solid curves. The $\omega_g$ mode for the case of a metal substrate ($h\omega_p = 9$ eV) with other structural parameters unchanged is also plotted as dotted curve; in this case, the $\omega_{sp1}$ and $\omega_{sp2}$ modes are too energetic to be seen in the figure.

In fact, in the limit $d_1 \to \infty$ (semi-infinite substrate), the solution of $\omega_g (q \to 0) = q r v_F$ is the same as that of the case in the limit $\omega_p \to \infty$ (metal substrate) because both $\omega_p$ and $d_1$ are in the denominator of (4.11); a plasma substrate of infinite thickness or infinite plasma frequency is physically identical and impenetrable for a plasmonic field of $q \to 0$. However, in the case of a semi-infinite substrate, our result given in (4.9) and (4.12) is different from the relation of $\omega_g (q \to 0) = \sqrt{e^2 E_F d_2 / \epsilon_2 \pi \hbar^2 q}$ found in Eq. (29) of Ref. 80. We believe our result is correct because, as mentioned above, it is improper to take $q \to 0$ while assuming constant $\omega$ in $\Pi (q \to 0, \omega)$, as is done in Ref. 80.

In the case of a semi-infinite plasma substrate ($d_1 \to \infty$), besides the $\omega_g$ mode there is also a high-energy mode $\omega_{sp}$ that can be obtained in the same way as the $\omega_{sp1}$ mode is derived in the case of finite $d_1$. The result is
\[
\omega_{sp}(q \to 0) = \frac{\omega_p}{\sqrt{1 + \frac{\epsilon_2 (\epsilon_3 + \epsilon_2 q d_2)}{\epsilon_0 (\epsilon_2 + \epsilon_2 q d_2)}}} = \sqrt{\frac{\epsilon_0}{\epsilon_0 + \epsilon_3}} \omega_p \left[ 1 + \frac{q d_2}{2} \left( \frac{\epsilon_3^2 - \epsilon_2^2}{\epsilon_2 (\epsilon_3 + \epsilon_0)} \right) \right].
\] (4.13)

In Figure 4.2(b), both the \(\omega_{sp}\) and \(\omega_g\) modes are numerically solved and plotted (thick solid curves) for a graphene sheet 30 nm above a semi-infinite plasma substrate; other parameters used for Figure 4.2(b) are the same as those used for Figure 4.2(a). The analytical results for \(\omega_g(q \to 0)\), given by (4.9) and (4.12), and for \(\omega_{sp}(q \to 0)\), given by (4.13), are plotted as thin solid curves. The decoupled \(\omega_{sp}\) and \(\omega_g\) modes are also plotted as dashed curves by setting \(d_2 = 5 \mu m\). As can be seen, because \(\epsilon_2\) is larger than \(\epsilon_3\) in our case, \(\omega_{sp}\) decreases from \(\omega_p \sqrt{\epsilon_0 / (\epsilon_0 + \epsilon_3)}\) with increasing \(q\) (see (4.13)). To compare our analytical result of \(\omega_g(q \to 0)\) with Eq. (29) of Ref. 80, we decrease \(d_2\) to 4 nm with other parameters unchanged so that the difference is more apparent in the plotting. We plot both \(\omega_g(q \to 0) = q r \nu_F\) (black circles) and Eq. (29) of Ref. 80 (red circles) in the inset of Figure 4.2(b) along with the numerical result (solid curve). As can be seen, the black circles are a better approximation of the accurate values than the red circles.
Figure 4.3. Comparison between the retarded SPP and nonretarded SP modes. The SP modes are plotted as dashed and solid curves, as also shown in Figure 4.2(b) for the curves in the same style. All the blue curves and the black curves are numerically calculated using $d_2 = 30 \text{ nm}$ and $d_2 = 5 \mu \text{m}$, respectively. The retarded $\omega_g$ mode is indistinguishable from the nonretarded $\omega_g$ mode. A waveguide mode appears between light lines $qc/n_3$ and $qc/n_2$ when $d_2 = 5 \mu \text{m}$.

4.3. Retardation effect

The retardation effect due to the finite speed of light $c$ is considered in Figure 4.3. The two straight thin lines are $\omega = qc/n_3$ and $\omega = qc/n_2$, respectively, where $n_i$ is the refractive index of medium $i$ given by $\sqrt{\epsilon_i/\epsilon_0}$. The nonretarded plasmon dispersion characteristics given by the thick solid curves ($d_2 = 30 \text{ nm}$) and the dashed curves ($d_2 = 5 \mu \text{m}$) in Figure 4.2(b) are plotted again in Figure 4.3 to be compared with the retarded plasmon dispersion characteristics given by the dotted curves that take into account the retardation effect. The retarded $\omega_{\text{sp}}$ approaches $qc/n_3$ asymptotically, resulting in a large deviation from the nonretarded $\omega_g$ as $q \to 0$. However, $\omega_g$ is barely affected by the retardation effect; the retarded curve for $\omega_g$ is indistinguishable from the nonretarded curve in Figure 4.3. When $d_2$ is sufficiently large (e.g., $d_2 = 5 \mu \text{m}$), a waveguide mode guided by the $\epsilon_2$ dielectric layer appears between $\omega = qc/n_3$ and
\[ \omega = q c / n_2 \] at an energy comparable to those of the plasmon modes; other high-order waveguide modes for this structure exist deeply inside the interband damping region with energies above \( 2E_F \) and are not shown in Figure 4.3.

![Figure 4.4](image-url)

Figure 4.4. (a) The graphene sheet is sandwiched between free space of permittivity \( \varepsilon_0 \) above and a dielectric of permittivity \( \varepsilon_d \) below. The thickness of the dielectric layer is \( d_2 \), and below it is a periodic structure characterized by a period of \( \Lambda \), a gap width of \( w \), a permittivity of \( \varepsilon_p \), and a thickness of \( d_1 \). Inside the gap and below the grating is also filled with the dielectric of permittivity \( \varepsilon_d \). (b) Photonic crystal (PC) model of the structure shown in (a) for one period. The effective indices for the first and second cavities are \( n_1 \) and \( n_2 \), respectively.

4.4. Excitation of graphene plasmons

To experimentally observe the SPP mode, a grating structure is usually needed to provide the extra momentum for the excitation of the surface plasmon modes\(^{83-85} \). According to the waveguide theory, it is generally assumed that the wave number of the excited first-order waveguide mode is given by \( 2\pi/\Lambda \), where \( \Lambda \) is the period of the grating; the frequency of the waveguide mode and that of the incident light is then obtained accordingly by plugging the known wavenumber into the eigenvalue equations of the waveguide structure without the grating.\(^{86} \) This is also true for the excitation of graphene SPPs if the gaps of the grating are small.
compared to the period. In this section, we obtain the dispersion characteristics of the graphene SPP modes on a grating structure both qualitatively and quantitatively. We show that the dispersion of the excited SPP modes highly depends on the size of the gap of the grating.

Consider the structure shown in Figure 4.4(a). The graphene sheet is sandwiched between the free space of permittivity $\varepsilon_0$ and a dielectric of permittivity $\varepsilon_d$. The thickness of $\varepsilon_d$ is $d_2$, and below which is a periodic structure characterized by a period of $\Lambda$, a gap width of $w$, a permittivity of $\varepsilon_p$, and a thickness of $d_1$. Inside the gap and below the grating is also filled with the dielectric of permittivity $\varepsilon_d$. With an optical wave of frequency $\omega$ that is normally incident from the above of the structure, graphene surface SPPs are excited with the field confined on the graphene surface. The SPP field is characterized by a longitudinal electric field component $E_z$, a transverse electric field component $E_x$, and a transverse magnetic field component $H_y$. According to the fundamental grating-coupling theory, the propagating mode that is phase matched to the normally incident wave is characterized by the wave number $q = 2n\pi/\Lambda$, where $n$ is a positive integer. The momentum $hq$ of the propagating mode is provided by the grating structure, and the plasmon energy $\hbar\omega$ is supplied by the photon energy. The dispersion relation between $q$ and $\omega$ is easy to solve when the grating is absent, namely, either $w \to 0$ or $w \to \Lambda$. In the case of $w \to \Lambda$, the graphene is simply sandwiched between dielectrics of permittivities $\varepsilon_0$ and $\varepsilon_d$. By using the Drude model, the dispersion in the low-temperature limit is given by

$$q = \frac{\pi\hbar^2\omega^2}{e^2E_F}(\varepsilon_0 + \varepsilon_d), \quad (4.14)$$

where $E_F$ is the Fermi energy of graphene. In (4.14) and equations below, the scattering rate is
ignored because to the first order the dispersion relation is not affected by the scattering rate when the scattering rate is low.\textsuperscript{76} We have also assumed that the phase velocity of the SPP mode $\omega/q$ is much smaller than the speed of light $c$. It is generally true considering the fact that the Fermi velocity $v_F$ is much smaller than $c$.\textsuperscript{74} As discussed in the previous sections, the SPP mode in this region of $\omega/q \ll c$ is essentially the SP mode in analogy to the definition of the SP mode of metals.\textsuperscript{87, 88}

In the limit of $w \to 0$, the configuration shown in Figure 4.4(a) becomes a graphene sheet located at a distance of $d_2$ above a uniform substrate of permittivity $\epsilon_p$. Therefore, the characteristic dispersion relation is given by (4.3) with $\epsilon_1 = \epsilon_2 = \epsilon_d$:

\[
\frac{\epsilon_p}{\Gamma + \tanh qd_2} \frac{\epsilon_p + \epsilon_d \tanh qd_1}{1 + \Gamma \tanh qd_2} = 0,
\]

where we have used the Drude model, and

\[
\Gamma = \frac{\epsilon_0}{\epsilon_d} - \frac{q}{\omega \epsilon_d} \frac{e^2 E_F}{\pi \hbar^2 \omega}.
\]

Equation (4.15) can be much simplified if the substrate is a metal. For most metals, which have plasmon energies on the order of electron volts, $\epsilon_p$ is comparatively much larger than the permittivity $\epsilon_d$ of most dielectrics in the terahertz frequency region where graphene plasmons oscillate. Therefore, we can write (4.15) as

\[
1 + \Gamma \tanh qd_2 = 0,
\]

which has to be solved numerically. For a finite $w$ smaller than the period $\Lambda$, equations (4.14) and (4.17) can only be treated at most as an approximation for the cases of small $w$ or $w$ close to
Λ. To obtain the dispersion of graphene SPP modes on a grating with an arbitrary gap size \( w \), another technique is needed as discussed in the following.

**Fabry-Pérot approximation**

Certainly, one can always resort to a full simulation to find the plasmon dispersion on the grating structure. However, this approach is not only time-consuming for the design process of the grating, but it also provides little physical insight. In fact, a simple analysis without much calculation can yield qualitative results for the graphene plasmon dispersion on a grating.

Essentially, one can regard the structure shown in Figure 4.4(a) as two Fabry-Pérot cavities. The edge of the grating slabs serves as boundaries that scatter the propagating plasmons in same manner as the boundaries of a Fabry-Pérot cavity that scatter the propagating plane waves. By taking one side of the gap to be at \( z = 0 \), the first cavity is characterized by a width of \( w \) corresponds to the region \( 0 < z < w \) in Figure 4.4(a); then, the second cavity is characterized by a width of \( \Lambda - w \) corresponds to the region \( w < z < \Lambda \). The presence of the boundaries results in a series of discrete resonant modes; the wavelength of the modes is given by \( \lambda = 2w/m \) and \( \lambda = 2(\Lambda - w)/n \) for the first and second cavities, respectively, where \( m \) and \( n \) are positive integers; hereafter, we call these modes FP (Fabry-Pérot) modes. The corresponding wave numbers are given by

\[
q = \frac{m \pi}{w}
\]  

(4.18)

for the FP modes of the first cavity, and
for the FP modes of the second cavity. The resonant frequencies are found by plugging (4.18) into (4.14) for the FP modes of the first cavity, and by plugging (4.19) into (4.17) for the FP modes of the second cavity.

\[ q = n \frac{\pi}{\Lambda - w} \]  

(4.19)

Figure 4.5. (a) FP modes and transmittance of the grating structure shown in Figure 4.4(a) for normally incident light. The structure is characterized by \( \Lambda = 10 \mu m \), \( d_1 = d_2 = 50 \text{nm} \), \( \epsilon_d = 3.9 \epsilon_0 \), and \( E_F = 100 \text{meV} \). The dispersion curves of the first five FP modes of each cavity are plotted as solid curves using (4.14), (4.17), (4.18), and (4.19). The transmittance is obtained using the simulation based on the classical modal method.\(^89\)

The resonant frequency as a function of \( w \) is plotted in Figure 4.5 for the first five FP modes of each cavity. The curves are calculated using the period \( \Lambda = 10 \mu m \), \( d_1 = d_2 = 50 \text{nm} \), \( \epsilon_d = 3.9 \epsilon_0 \), and \( E_F = 100 \text{meV} \). From (4.18) and (4.14), we find that as \( w \) increases, \( q \) decreases and thus the frequency decreases for the FP modes in the first cavity, as also shown in Figure 4.5. The trend is opposite for the modes in the second cavity: from (4.19) and (4.17), we can find that as \( w \) increases, \( q \) increases and thus the frequency increases. These cavity modes are compared with the transmittance of the structure shown in Figure 4.4(a) as a transmittance map in the
background. The transmittance is obtained using the simulation based on the classical modal method. For a fixed $w$, the excitation of plasmons can be seen by locating the transmittance minimum in the map. When $w$ is extremely small, the plasmon modes of the grating are simply given by (4.19) with $n$ being an even number, as shown in Figure 4.5 for the dashed curves at $w \approx 0$. This is because of the periodic structure of the grating; the phase shift $q \Lambda$ of the SPP mode has to be multiple of $2\pi$, namely $\cos q \Lambda = \cos n\pi = 1$, for one period in the case of normally incident light.

As $w$ increases, the phase shift across the gap can no longer be ignored, and the plasmon modes start to deviate from the even $n$ modes. Nevertheless, the requirement of the phase shift of the plasmon mode being multiple of $2\pi$ in one period still holds. Therefore, instead of $n$ being an even number, we find that the plasmon modes can be found approximately at the locations where $n + m$ is an even number; these locations are shown as dotted symbols at the intersections of dispersion curves; the excitation of plasmons is suppressed at the locations where $n + m$ is an odd number. The fact that for a large $w$ the frequency of the SPP modes can be qualitatively located by regarding the grating structure as two Fabry-Pérot cavities suggests strong reflection of propagating plasmons at the gap edges; the plasmon modes become highly localized inside the gap or within the metallic slabs in contrast to the propagating SPP when $w$ is small.

**Photonic crystal approximation**

To quantitatively find the SPP modes on the grating, instead of viewing one period of the grating as two Fabry-Pérot cavities, we shall resort to the theory of photonic crystal (PC). We regard the grating structure as alternating layers of dielectrics of the refractive index $n_1$ and $n_2$, 

as shown in Figure 4.4(b) for one pair of layers. The effective refractive indices \( n_1 \) and \( n_2 \) are not given by \( \sqrt{\epsilon_d/\epsilon_0} \) and \( \sqrt{\epsilon_p/\epsilon_0} \), because along the \( x \)-axis the permittivities are not homogeneously \( \epsilon_d \) and \( \epsilon_p \) in the gap region and substrate region, as shown in Figure 4.4(a). Instead, the effective indices have to be obtained from \( n_1 = c\beta/\omega \) where \( \beta \) is given by (4.14), and \( n_2 = c\beta/\omega \) where \( \beta \) and \( \omega \) satisfy (4.17). If the scattering rate of the Drude model is small, the PC represents a lossless and reciprocal system; the PC band structure is thus described by\(^{90}\)

\[
\cos(q\Lambda) = \text{Re} \frac{1}{t},
\]

(4.20)

where \( t \) is the transmission coefficient for one period of the PC:

\[
t = \frac{4n_1n_2e^{i\omega n_1(\Lambda-w)/c}}{(n_1 + n_2)^2 - (n_2 - n_1)^2 e^{2i\omega n_1(\Lambda-w)/c}}.
\]

(4.21)

Because of the cosine function in (4.20), the PC band structure has a period of \( \Lambda \) as expected. It can be numerically shown that numerous PC bands can be found and plotted on the \( q-\omega \) map using (4.20). The field inside the PC can be characterized by an \( E_x \) electric field shown in Figure 4.4(b). A few examples of the \( E_x \) distribution as a function of \( z \) are shown in Figure 4.6(c), (e), (f), and (h) for \( w=0.7\Lambda \) and \( \cos(q\Lambda)=1 \). Each continuous curve represents one snapshot at a certain time. If we look at one snapshot for the field distribution of \( E_x \), we find that PC modes are either characterized by a symmetric or antisymmetric profile to the middle dashed lines in two layers. If \( \cos(q\Lambda) \neq 1 \), i.e., the phase shift of the PC mode over one period is not a multiple of \( 2\pi \), the field distribution is neither symmetric nor anti-symmetric.
Figure 4.6. (a) Transmittance of normally incident light through the grating structure shown in Figure 4.4(a), calculated using the simulation based on the classical modal method. The parameters used for the calculation are the same as those for Figure 4.5. The dispersion of the plasmon modes are plotted as solid and dashed curves obtained using (4.20) with \( \cos q\Lambda = 1 \) considering the PC model shown in Figure 4.4(b). The distributions of \( E_x \) at the selected points for \( w = 0.7\Lambda \) are plotted in (b-h). Field distributions are antisymmetric and symmetric for the solid and dashed curves, respectively. The exact field distributions in (b), (d), and (g) are calculated on the graphene sheet using the mode-matching method. The field distributions in (c), (e), (f), and (h) are calculated using the PC model.

The PC modes and the plasmon modes are highly related. In fact, each PC mode corresponds to one plasmon mode. For normally incident light on the grating, \( \cos q\Lambda = 1 \) is satisfied; thus, the plasmon modes can be found by finding the solution of (4.20) with \( \cos q\Lambda = 1 \). For each fixed structure of a certain \( w \), a set of solution can be found from (4.20), and we can plot the plasmon modes as a function of \( w \), as shown in Figure 4.6(a) for the solid curves. Plasmon modes that are characterized by a symmetric field distribution of \( E_x \) are not shown except for the lowest order mode plotted as a dashed curve. For a normally incident plane wave with symmetric \( E_z \) and \( H_y \), the excited field is also characterized by symmetric \( E_z \) and \( H_y \),
and the excited $E_x$ is antisymmetric because $E_x \propto \partial H_y / \partial z$. Thus, the plasmon modes characterized by a symmetric $E_x$ cannot be excited in our configuration.

The plasmon modes found from (4.20) is compared with the transmittance map as shown in Figure 4.6(a) using the simulation based on the classical modal method.\(^{89}\) The exact solution of the plasmon dispersion is found by locating the transmittance minimum. The exact field distribution on the graphene sheet is shown in Figure 4.6(b), (d), and (g) for the selected points at the transmittance minimum for $w = 0.7 \Lambda$. The field distribution on the graphene sheet is calculated using the mode-matching method.\(^{89}\) Rayleigh modes up to 31 orders are considered, and 25 waveguide modes in the gap are accounted. Higher order modes can also be included, but the correction of the fields shown in Figure 4.6(b), (d), and (g) can be hardly seen. As can be seen, all the fields shown are characterized by antisymmetric $E_x$ distributions, which resemble the field distributions derived using the PC model. Some discrepancies in the field distributions and in the dispersion curves are expected because the PC model is simplified structure of the original grating structure; any effect arising from the edges of the grating cannot be taken into account in the PC model. As can be seen in Figure 4.6, the dispersion of the plasmon modes found from equation (4.20) is in quantitative agreement with the exact solution. Therefore, we find that the dispersion characteristics of such a complex system can be well approximated by using the photonic crystal model, and therefore a full simulation is not needed.
Chapter 5. Plasmonic graphene applications

5.1. Plasmonic waveguide based on monolayer graphene

Theoretical models

Figure 5.1. (a) Planar waveguide, and (b) nonplanar rectangular waveguide considered in this chapter. In (a) from bottom to top are a metal slab of permittivity \( \varepsilon_1 \), a dielectric layer of permittivity \( \varepsilon_2 \) and thickness \( d_2 \), a sheet of monolayer graphene, a dielectric layer of permittivity \( \varepsilon_3 \) and thickness \( d_3 \), and a metal slab of permittivity \( \varepsilon_4 \). In (b), the core has a thickness \( d_2 + d_3 \), a width \( w \), and a permittivity \( \varepsilon_{\text{core}} \) both above and below the graphene sheet; the rest of the space between the metal slabs is filled with a dielectric of permittivity \( \varepsilon_{\text{clad}} \) as cladding regions.

The structure shown in Figure 4.1 is essentially a plasmonic waveguide. To increase the confinement of the field in the waveguide, we modify the structure in Figure 4.1 to the ones shown in Figure 5.1 where two types of waveguides, namely planar and nonplanar waveguides, are schematically drawn. As shown in Figure 5.1, the graphene sheet is sandwiched between dielectrics of permittivity \( \varepsilon_2 \) and \( \varepsilon_3 \). Below the dielectric \( \varepsilon_2 \) and above the dielectric \( \varepsilon_3 \) are metal slabs of infinite thickness. The metal slabs are characterized by permittivities

\[
e_i = \varepsilon_0 \left[1 - \omega_i^2/(\omega^2 + i\omega\gamma_i)\right],
\]

where \( i = 1 \) for the bottom metal and \( i = 4 \) for the top metal, and \( \omega \) and \( \gamma \) are respectively the plasma frequency and the carrier scattering rate of metal \( i \). Because of
the presence of the metal slabs, the plasmonic field arising from the collective electronic oscillation on the graphene sheet is confined within the dielectric layers. The resultant dispersion relations of the confined surface plasmon modes can be found by solving Maxwell’s equations with appropriate boundary conditions. The characteristic equation to be solved for the plasmon dispersion is similar to the one given by (4.3) as

\[
\frac{\varepsilon_2}{\varepsilon_3 + \varepsilon_4 \tanh q d_3} + \frac{\varepsilon_4}{\varepsilon_2 + \varepsilon_3 \tanh q d_2} = -\frac{i}{\omega} \sigma(q,\omega),
\]

(5.1)

where \(\sigma(q,\omega)\) is the optical conductivity of graphene given by (4.2), and \(d_2\) and \(d_3\) are the thicknesses of the dielectric layers \(\varepsilon_2\) and \(\varepsilon_3\), respectively. In (5.1), the retardation effect is ignored so that (5.1) describes a SP mode of the structure. In Figure 4.1(a), metals of infinite thickness are assumed. For metal slabs of finite thicknesses, (5.1) serves as a very good approximation for the plasmonic dispersion if their thicknesses are much larger than their skin depths; for gold, the skin depth is about \(c/\omega_p \approx 22\) nm for the bulk plasma energy of \(\hbar\omega_p = 8.89\) eV.\(^{91}\)

Unlike the previous sections where we have ignored the loss, for the study of a waveguide it is crucial to include the scattering loss in the model. The optical conductivity can be obtained within the relaxation-time approximation\(^{75, 92, 93}\) combined with the random-phase approximation (RPA) in the low-temperature limit:\(^{13}\)

\[
\sigma(q,\omega) = -i\omega \chi(q,\omega)
\]

(5.2)

where the 2D susceptibility is given as
\[ \chi(q, \omega) = \frac{e^2}{q^2} \frac{\omega + i\gamma}{\omega \Pi(q, \omega + i\gamma)^{-1} + i\gamma \Pi(q, 0)^{-1}} \]  

(5.3)

where \( \gamma \) is the carrier scattering rate of the graphene sheet and the polarizability function \( \Pi(q, \omega + i\gamma) \) has the same form as that in Ref. 76. For finite carrier scattering rates \( \gamma_i \) of the metal slabs and a finite carrier scattering rate \( \gamma \) of the graphene sheet, (5.1) can be satisfied only when the plasmon wave number is a complex number: \( q = q_1 + iq_2 \). Assuming low scattering rates such that \( q_1 \gg q_2 \), we can expand (5.1) in terms of \( q_2/q_1 \) and ignore the high-order terms. The real part of (5.1) is given as

\[ \frac{\text{Re}\left(\epsilon_4\right)}{\epsilon_3 + \text{Re}\left(\epsilon_4\right)\tanh q_1d_3} + \frac{\text{Re}\left(\epsilon_1\right)}{\epsilon_2 + \text{Re}\left(\epsilon_1\right)\tanh q_1d_2} = -e^2 \frac{\text{Re}\left(\Pi_i, \omega\right)}{q_1} \]  

(5.4)

As can be seen, \( q_2 \) does not enter (5.4); therefore \( q_1 \) can be numerically solved from (5.1). In the THz spectral region, it is usually true for metals that \( \text{Re}\left(\epsilon_1\right) \) and \( \text{Re}\left(\epsilon_4\right) \) are the dominant terms in the denominators and numerators of (5.4). In such a case, an analytical solution can be found in the low-\( q_1 \) region where \( q_1d_2 \ll 1 \) and \( q_1d_3 \ll 1 \):

\[ q_1 = \omega/\nu_F, \]  

(5.5)

where

\[ r = \frac{1 + \Lambda}{\sqrt{1 + 2\Lambda}}, \text{ with } \Lambda = \frac{2e^2k_F}{\pi hv_F} \left( \frac{\epsilon_3}{d_3} + \frac{\epsilon_2}{d_2} \right), \]  

(5.6)

which is similar to the solution given by (4.12) for a waveguide with only one side being metallic.

The imaginary part of (5.1) gives the solution of \( q_2 \) in the limit of \( q_1 \gg q_2 \):
\[
q_2 = \frac{\text{Im} \Pi(q_1, \omega) + \left( \frac{\gamma}{\partial \omega} + \frac{\gamma}{\omega} \right) \text{Re} \Pi(q_1, \omega) - \frac{\gamma}{\omega} \left[ \text{Re} \Pi(q_1, \omega) \right]^2}{R + \left( \frac{1}{q_i} - \frac{\partial}{\partial q_i} \right) \text{Re} \Pi(q_1, \omega)},
\]

(5.7)

where

\[
R = \left( \frac{\varepsilon_2 d_2}{\sinh^2 q_id_2} + \frac{\varepsilon_3 d_3}{\sinh^2 q_id_3} \right) q_i \varepsilon^2
\]

(5.8)

is the “reduction factor” contributed by the metals. A large \( R \) leads to a small \( q_2 \) and hence a long propagation distance of the plasmon mode. When the metal slabs are removed by setting \( d_2 \to \infty \) and \( d_3 \to \infty \), the reduction factor \( R \) becomes zero; then, (5.7) reduces to the expression for \( q_2 \) in the case of a graphene sheet sandwiched between dielectrics \( \varepsilon_2 \) and \( \varepsilon_3 \) in the absence of the metal slabs. \(^{76}\) Note that in the derivation of (5.7), it is assumed that \( \text{Re} \varepsilon_1 \gg \varepsilon_2 \) and \( \text{Re} \varepsilon_4 \gg \varepsilon_3 \), which is generally true in the terahertz spectral region for metals compared to common dielectrics. In the limit of \( q_id_2 \ll 1 \) and \( q_id_3 \ll 1 \), (5.7) can be further reduced to

\[
q_2 = \gamma / 2rv_e,
\]

(5.9)

where \( r \) is given by (5.6).
Figure 5.2. (a) Dispersion characteristics and (b) the attenuation length of surface plasmons as a function of surface plasmon energy for the structure shown in Figure 5.1(a) with $\varepsilon_2 = \varepsilon_3 = \varepsilon_0$, and (c) the current density on the graphene surface divided by that in the absence of the metal slabs. The metal is assumed to be gold with $\hbar \omega_1 = \hbar \omega_4 = 8.89$eV and $\gamma_1 = \gamma_4 = 17$ ps$^{-1}$. The scattering rate $\gamma = 4$ ps$^{-1}$ and the Fermi energy $E_F = 100$meV are taken for the graphene sheet. The curves are calculated using different values of $d_2 = d_3 = d$ marked next to each curve. In (a) and (b), the solid curves are numerically calculated using $(5.4)$ and $(5.7)$. The dashed curves are analytically obtained using $(5.5)$ and $(5.9)$. The dots are located at $q_i = 1.2/d$ for different values of $d$. In (c), the scattering rates of $\gamma_1 = \gamma_4 = \gamma = 0$ are assumed.
Simulation results

The dispersion characteristics of surface plasmons for the structure shown in Figure 5.1(a) are plotted in Figure 5.2(a). The solid curves are numerically obtained from (5.4), and the dashed curves are analytically calculated using (5.5). For all curves, $\epsilon_2 = \epsilon_3 = \epsilon_0$ is assumed for the dielectric layers, the scattering rate $\gamma = 4 \text{ ps}^{-1}$ and the Fermi energy $E_F = 100 \text{ meV}$ are taken for the graphene sheet, and the parameters of gold are used for $\epsilon_1 = \epsilon_4$ of the metal slabs. The curves are calculated using different values of $d_2 = d_3 = d$ marked next to each curve. As can be seen, the approximate analytical results agree very well with the exact numerical results in the low-frequency region as long as the condition $q_1d \ll 1$ is satisfied. In this region, the plasmon dispersion is a strong function of $d$; at a fix frequency, $q_1$ increases with decreasing $d$, signifying that the confinement of the plasmonic field can be consistently enhanced with decreasing graphene–metal distance. However, in the high-frequency region, all numerical curves merge into the blue curve because of the strong confinement of the plasmonic field on the graphene sheet: the presence of metal slabs becomes insignificant as the plasmonic wavelength becomes much smaller than the physical distances between the graphene sheet and the metal surfaces.

The attenuation length $q_2^{-1}$ is plotted in Figure 5.2(b) with the graphene–metal distance $d$ marked next to each curve. The solid curves are obtained from (5.7) using the values of $q_i$ given in Figure 5.2(a), and the dashed curves are analytically calculated using (5.9). As can be seen, the approximate analytical results agree very well with the exact numerical results in the low-frequency region. Note that below $\hbar \omega = 2.5 \text{ meV}$ where $\omega$ is of the same order of magnitude as
the curves in Figure 5.2(a) and Figure 5.2(b) can only be regarded as an approximation because the condition \( q_1 \gg q_2 \) is no longer valid. In the high-frequency region, all curves merge into the blue curve and start to increase rapidly near the Landau damping region approximately located at \( \omega E_F = 1.49 \) , which is obtained by finding the intersection of the blue curve and the boundary of the Landau damping region. The equation to be solved is approximately given by

\[
0.23 \varepsilon_{\text{ave}} \left( \frac{\hbar \omega}{E_F} \right)^2 + \frac{\hbar \omega}{E_F} - 2 = 0,
\]

where \( \varepsilon_{\text{ave}} = \left( \varepsilon_2 + \varepsilon_3 \right) / 2 \varepsilon_0 = 1 \) for the case considered in Figure 5.1. In the intermediate-frequency region where \( q_1^{-1} \) is comparable to \( d \), the black curves are below the blue curve, signifying reduced loss in the presence of the metal slabs. Therefore, as seen in Figure 5.2(a) and Figure 5.2(b), we find that an enhanced confinement and an increased propagation distance can be simultaneously achieved by the presence of the metal slabs within a certain spectral region.

The phenomena of increased propagation in the presence of the metal slabs can be explained as follows. The highly conductive graphene sheet and a metal surface effectively form a Fabry–Pérot cavity that tends to push the Poynting flux from the graphene surface toward the center of the graphene–metal cavity. As a result, when the plasmonic wavelength is comparable to the graphene–metal distance, the Poynting flux and the current density on the graphene sheet is reduced so that the Joule loss due to the carrier scattering rate \( \gamma \) of graphene decreases. To show the shift of the Poynting flux, we simplify the problem by ignoring the losses in the graphene sheet and the metal slabs so that \( q_2 = 0 \). By normalizing the electromagnetic power over the transverse cross section between the two metal surfaces, the magnitude of the current density on the graphene sheet can be shown to be proportional to \( \sqrt{\omega} \) when the metal slabs are
absent, and it is proportional to $\sqrt{\omega/\left(\tanh dq_1 + dq_1 \sech^2 dq_1\right)}$ for finite distances of $d_2 = d_3 = d$ between graphene and the metal surfaces. The ratio of the current density on the graphene sheet for different values of $d$ to that for the case of free-standing graphene in the absence of the metal slabs are shown in Figure 5.2(c). As can be seen from the relative current density shown in Fig. 5.2(c), for $q_1 d = 1$, the relative current density has a value greater than unity, indicating a higher loss in the presence of the metal slabs than that in the case of free-standing graphene, as shown in Figure 5.2(b). For $q_1 d \gg 1$, the presence of the metal slabs becomes insignificant as the plasmonic wavelength is much smaller than the physical distances between graphene and the metal surfaces. As a result, the relative current density has the value of unity in the high-frequency region. In the intermediate-frequency region where $q_1^{-1}$ is comparable to $d$, strong interaction between the graphene sheet and the metal surfaces pushes the Poynting flux away from the graphene surface, resulting in a reduced loss in the presence of the metal slabs, as seen in Figure 5.2(b). The minimum of the relative current density in Figure 5.2(c) does not pinpoint the peak of $q_1/q_2$ near the interband scattering region, such as the case of $d = 10\text{ nm}$, because we have assumed lossless graphene for the calculation to obtain the data in Figure 5.2(c).

The locations of the troughs for the $q_2$ curves with respect to the blue curve in Figure 5.2(b) can be obtained by finding the minimum of the relative current density given by

$$1/\sqrt{\tanh dq_1 + dq_1 \sech^2 dq_1}$$

in Figure 5.2(c) for each value of $d$; the solution is $dq_1 \tanh dq_1 = 1$ or $dq_1 \approx 1.2$. The locations of $dq_1 = 1.2$ are dotted for all curves in each figure of Figure 5.2, except for the case of $d = 10\text{ nm}$, for which the value of $q_1 = 1.2/d$ is inside the Landau damping region. As can be seen, these dots quantitatively mark the peaks in Figure 5.2(b), and in Figure
5.2(a) they mark the points where the dispersion curves transition from linear relations to quadratic relations. For the case of $d = 50 \text{ nm}$, we found a maximum of 20% increase in the propagation distance from 1.04 $\mu\text{m}$ to 1.25 $\mu\text{m}$ and a 42% increase in $q_1/q_2$ at $\hbar \omega = 71.9 \text{ meV}$ where the dot is located. By further reducing the surface plasmon energy $\hbar \omega$ to $9 \text{ meV}$, a maximum of 97% increase in $q_1/q_2$ can be obtained.

Nonplanar waveguide

The structure in Figure 5.1(a) can only confine the plasmonic field in the direction perpendicular to the surface. Two-dimensional confinement can be realized using the nonplanar structure in Figure 5.1(b) by introducing dielectric boundaries in the lateral direction. For such a structure, the plasmonic waveguide modes can be found by using the effective-index method. In this method, the surface plasmon modes of the cladding and core regions are solved individually as a planar waveguide shown in Figure 5.1(a). The obtained wave number $q$ for each region gives the effective permittivity of the corresponding region by $\epsilon_{\text{eff}} = \epsilon_0 c^2 q^2/\omega^2$. With $\epsilon_{\text{eff}}$ known for both core and cladding regions, the plasmonic waveguide modes are solved numerically in the same manner as the modes of a dielectric slab waveguide with a core width of $w$. The characteristics of the plasmonic waveguide modes for the structure shown in Figure 5.1(b) with the parameters $\epsilon_{\text{core}} = 4 \epsilon_0$, $\epsilon_{\text{clad}} = \epsilon_0$, $d_2 = d_3 = 10 \text{ nm}$, and $w = 100 \text{ nm}$ are plotted in Figure 5.3. The black curve is identical to the curve associated with $d = 10 \text{ nm}$ in Figure 5.2(a) and Figure 5.2(b) with $\epsilon_2 = \epsilon_3 = \epsilon_{\text{clad}}$, and the red curve is obtained in the same manner as that for the black curve using (5.4) and (5.7) with $\epsilon_2 = \epsilon_3 = \epsilon_{\text{core}}$ for the waveguide core. As can be seen, the
plasmonic waveguide modes are mostly confined between red and black curves. Except for the fundamental mode, which does not have a cutoff frequency, the cutoff frequency $f_c$ of a high-order mode $v$ is located near the intersection of the blue curve $v$ and the black curve. At $f_c$, the plasmonic field is not confined within the core but spreads through the cladding regions. Due to the finite imaginary parts of the effective permittivities, $f_c$ is not located exactly on the black curve but slightly below it, as shown in the blowup in Figure 5.3(a). It can be shown that for an even larger scattering rate $\gamma$, the cutoff frequency $f_c$ further decreases, moving away from the black curve because the black curve is not a function of $\gamma$ determined by (5.4). As an example, $f_c$ of mode $v = 4$ as a function of $\gamma$ is plotted in the inset of Figure 5.3(a), which clearly shows the described trend.

The values of $q_1/q_2$ for different modes are plotted as blue curves in Figure 5.3(b). The kinks of the blue curves around $h\omega = 110 \text{meV}$ are due to the kink of the red curve at the boundary of the Landau damping region shown in Figure 5.3(a). The kink is also observed for the case of free-standing graphene.\textsuperscript{76} As the frequency approaches $f_c$, the value of $q_1/q_2$ increases sharply at the expanse of poor confinement of the plasmonic field. Away from $f_c$, the $q_1/q_2$ curves for different modes are all below the black and red curves; the attenuation length $q_2^{-1}$ is sacrificed for the lateral confinement, which is visualized in Figure 5.3(b) as line segments for the comparison among modes $v = 1$ and $v = 2$ of nonplanar waveguides and modes of slab waveguides at $h\omega = 70 \text{meV}$. In general, the values of $q_1/q_2$ and $q_2^{-1}$ for the fundamental mode $v = 1$ are the highest among all modes of the nonplanar plasmonic waveguide,
but they are lower than those of slab waveguides shown in Figure 5.1(a) using the same
dielectrics.

![Graph](image_url)

Figure 5.3. (a) Dispersion characteristics and (b) $q_1/q_2$ as a function of surface plasmon energy for different plasmonic waveguide modes. The nonplanar waveguide structure is schematically drawn in Figure 5.1(b) with parameters $d_2 = d_3 = 10$ nm, $w = 100$ nm, $\epsilon_{core} = 4\epsilon_0$, and $\epsilon_{clad} = \epsilon_0$. Other parameters are the same as those used for Figure 5.2. The four thin blue curves are associated with four different waveguide modes with the mode number $\nu$ marked next to each curve. The black curve is identical to the curve associated with $d = 10$ nm in Figure 5.2(a) and Figure 5.2(b) with $\epsilon_2 = \epsilon_3 = \epsilon_{Core}$, and the red curve is obtained in the same manner as the black curve except for $\epsilon_2 = \epsilon_3 = \epsilon_{core}$. In (a), the colored areas are the Landau damping regions. For mode $\nu = 4$, the dispersion curve near the cutoff frequency $f_c$ is blown up in the bottom inset to show that the cutoff appear below the black curve, and its cutoff frequency $f_c$ as a function of $\gamma$ is plotted in the top inset. In (b), values of $q_2^{-1}$ at $\hbar\omega = 70$ meV are visualized as line segments, which from bottom to top as the arrow indicates are associated with $\nu = 2$, $\nu = 1$, red and black curves, respectively.
5.2. Plasmonic waveguide based on multilayer graphene

There are several ways to reduce the loss of the surface plasmon of single-layer graphene (SLG) besides improving the quality of graphene. For example, in the previous section we find that by placing SLG in close proximity to a high-index dielectric or a plasma layer such as metal, the attenuation of surface plasmons can be effectively reduced by 16% to 20%. In this section, we show that the propagation distances of surface plasmons can also be increased by placing two sheets of SLG in close proximity to form a double-layer graphene (DLG) structure. Coupled-plasmon modes of graphene sheets and graphene nanoribbons are widely studied in the literature. In contrast to previous treatments, we focus on the attenuation loss of these coupled-plasmon modes by considering both extrinsic scattering and intrinsic Landau damping. To systematically analyze the source of the loss, different models reported in the literature are considered to obtain a consistent result.

Theoretical models

Consider a DLG structure consisting of a dielectric of permittivity \( \varepsilon_2 \) filling the space between two graphene sheets of spacing \( d \) and a dielectric of permittivity \( \varepsilon_1 \) filling the rest of the space. Both graphene sheets have the same properties characterized by a scattering rate \( \gamma \) and a Fermi energy \( E_F \). The characteristic equation to be solved for the plasmon dispersion of DLG is
\[
\frac{i\sigma q + \epsilon_1 \omega}{\epsilon_2 \omega} + \coth^{-1}\left(\frac{qd}{2}\right) = 0, \tag{5.10}
\]
where the plus and minus signs correspond to the acoustic and optical modes, respectively. In (5.10). In the presence of extrinsic scattering or intrinsic electron–hole excitation (Landau damping), the optical conductivity \( \sigma \) is a complex-valued function of \( q \) and \( \omega \). Then, (5.10) can be satisfied only when the wave number is a complex number: \( q = q_1 + iq_2 \), where \( q_2 \) quantifies the attenuation of the plasmon mode.

The simplest model for the optical conductivity of graphene is the Drude model, which ignores Landau damping and assumes an energy-independent scattering rate \( \gamma \) within the relaxation-time approximation. The Drude conductivity is given by

\[
\sigma(\omega) = \frac{2e^2 k_B T}{\pi} \ln\left(\frac{\cosh(\mu / 2k_B T)}{\pi \hbar^2 (\gamma - i\omega)}\right), \tag{5.5, 5.6}
\]
where \( k_B \) is the Boltzmann constant and \( \mu \) is the chemical potential. In the limit of \( E_F \gg k_B T \), \( \mu \) can be approximated by

\[
\mu = E_F \left[1 - \pi^2 k_B T^2 / 6E_F^2 \right]. \tag{5.32}
\]
By plugging the Drude conductivity in (5.10), we obtain the expression for the plasmon dispersion assuming \( q_1 \gg q_2 \):

\[
\hbar \omega = \sqrt{\frac{2q_1 e^2 k_B T}{\pi} \ln\left(\frac{\cosh(\mu / 2k_B T)}{\pi \hbar^2 \left(\epsilon_1 + \epsilon_2 \coth^{-1}\left(\frac{qd}{2}\right)\right)}\right), \tag{5.11}
\]
and the attenuation parameter \( q_2 \) is given by

\[
q_2 = \frac{\gamma q_1}{\omega} \left\{1 \pm \frac{q_d \epsilon_2}{\epsilon_1 \cosh q_1 d + \epsilon_2 \sinh q_1 d + \epsilon_1}\right\}^{-1}. \tag{5.12}
\]
As can be seen in (5.12), the assumption \( q_1 \gg q_2 \) implies \( \omega \gg \gamma \), but the reverse is not true because \( q_2 \) is also contributed by intrinsic Landau damping, as we will discuss shortly. In the
long-wavelength limit such that $q_i \to 0$, (5.11) reduces to the result obtained in Ref. 100 for the optical mode.

To account for both Landau damping and extrinsic scattering, the optical conductivity is derived within the random-phase approximation (RPA)\textsuperscript{13, 75} together with the relaxation-time approximation (RT).\textsuperscript{76, 92, 93} The RPA–RT conductivity is expressed as (5.2):

$$\sigma(q, \omega) = -i \omega \chi(q, \omega).$$

where the 2D susceptibility is given by (5.3):

$$\chi(q, \omega) = \frac{e^2}{q^2} \frac{\omega + i \gamma}{\omega \Pi(q, \omega + i \gamma)^{-1} + i \gamma \Pi(q, 0)^{-1}},$$

and the polarizability function $\Pi(q, \omega)$ has the same form as given in Ref. 76. In the case of $\gamma = 0$, (5.14) reduces to the usual RPA susceptibility $\chi(q, \omega) = e^2 \Pi(q, \omega)/q^2$. To obtain the RPA–RT results, $\Pi(q, \omega + i \gamma)$ is expanded into Taylor series:

$$\Pi(q, \omega + i \gamma) = i q_2 \partial \Pi(q, \omega)/\partial q_1 + i \gamma \partial \Pi(q_1, \omega)/\partial q + ...$$

where an analytical form of $\Pi(q, \omega)$ in the limit of temperature $T \to 0$\textsuperscript{13, 75, 93} and a semi-analytical form of $\Pi(q_1, \omega)$ at a finite temperature\textsuperscript{108} have previously been derived. By plugging the RPA–RT conductivity of (5.13) in (5.10), the characteristic equation can be separated into real and imaginary parts in the limit of $q_i \gg q_2$. The real part of (5.10) in the limit of $q_i \gg q_2$ is given by

$$\frac{1}{\epsilon_2} \left[ \epsilon_1 + e^2 \frac{\text{Re} \Pi(q_1, \omega)}{q_1} \right] + \coth^{-1} \left( \frac{q_2 d}{2} \right) = 0.$$  

With proper manipulation of $\text{Re} \Pi(q_1, \omega)$ in (5.15),\textsuperscript{74} it can be shown that in the long-
wavelength limit (5.15) reduces to the result obtained in Ref. 103 for the acoustic mode. The imaginary part of (5.10) in the limit of \( q_i \gg q_2 \) is given by

\[
q_2 = \frac{\text{Im} \Pi(q_i, \omega) + \left( \frac{\gamma}{\omega} + \frac{\gamma}{\omega} \right) \text{Re} \Pi(q_i, \omega) - \frac{\gamma \left[ \text{Re} \Pi(q_i, \omega) \right]^2}{\omega \text{Re} \Pi(q_i, 0)}}{R + \left( \frac{1}{q_i} - \frac{\partial}{\partial q_i} \right) \text{Re} \Pi(q_i, \omega)},
\]

(5.16)

where we have ignored the imaginary part of \( \Pi(q, 0) \) by writing \( \Pi(q, 0) = \text{Re} \Pi(q_i, 0) \) because \( \Pi(q, 0) \) is approximately a constant \( 2k_i / \pi \nu_i \hbar \) for \( q_i \leq 2k_i \) in the temperature range we consider in this section. The \( R \) parameter in (5.16) is a function of \( q_i \):

\[
R = \frac{\varepsilon_i q_i d}{2e^2} \left[ \coth^2 \left( \frac{q_i d}{2} \right) - 1 \right],
\]

(5.17)

which has a role of reducing \( q_2 \) if \( R > 0 \). In the limit of \( d \to \infty \), the reduction factor \( R \) becomes zero as the two graphene layers are decoupled. As a result, the expression of \( q_2 \) given in (5.16) reduces to that for SLG sandwiched between dielectrics \( \epsilon_i \) and \( \epsilon_2 \).76

Simulation results

The complex \( q \) of the plasmon modes for DLG as a function of plasmon energy in the limit of \( T \to 0 \) is shown in Figure 5.4, where the black and red curves represent acoustic and optical modes, respectively. The structural parameters used for this figure are \( d = 50 \text{nm} \) and \( \epsilon_1 = \epsilon_2 = 4 \epsilon_0 \), where \( \epsilon_0 \) is the permittivity of free space. For both graphene sheets, the Fermi energy is taken to be \( E_F = 100 \text{meV} \), and a scattering rate of \( \gamma = 4 \text{ps}^{-1} \) is chosen as a reasonable
value, which is experimentally observed in chemically deposited graphene.\textsuperscript{19, 48, 109} Both the solid and dashed curves are obtained from (5.15) and (5.16). The difference is that the solid curves represent the RPA–RT results, and the dashed curves represent the RPA results obtained by setting $\gamma = 0$ so that extrinsic scattering is ignored. By contrast, Landau damping is ignored for the dotted curves, which are numerically calculated from (5.10) using the Drude conductivity. As can be seen in Figure 5.4(a) and Figure 5.4(b), in the low-frequency region where $q_1d = 1$, the RPA–RT curves converge to the Drude curves as Landau damping is not significant. By comparing the Drude curves with the RPA–RT curves, we find that the RPA–RT curves are reasonably accurate down to $\hbar \omega \approx 0.05 E_F$ even though the assumption $q_1 \gg q_2$ is not strictly valid. In the high-frequency region where $q_1d \gg 1$, the two graphene sheets are decoupled, and both plasmon modes converge to the SLG plasmon mode represented by the grey curve. As the plasmon modes enter into the Landau damping region, $q_2$ increases rapidly for both the RPA–RT and RPA curves; this behavior cannot be captured by the Drude model.

The intersection of $q_2$ curves for the two plasmon modes in Figure 5.4(b) can be solved by equating $q_2$ of the acoustic mode to that of the optical mode, both obtained from (5.12). Considering a homogeneous dielectric background of $\epsilon = \epsilon_1 = \epsilon_2$, the identity

$$q_1^*d = 2\left[1 + \tanh^{-1}\left(q_1^*d/2\right)\right]^{-1}$$

is valid at the intersection, where $q_1^+$ and $q_1^-$ are the real parts of the wave numbers of acoustic and optical modes, respectively. Subsequently, the plasmon energy $\hbar \omega$ can be numerically solved from (5.11) using the above identity:
\[ h\omega = \frac{3}{4} \sqrt{\frac{2e^2 k_b T}{\pi \epsilon d}} \ln \left[ 2 \cosh \left( \frac{\mu}{2k_b T} \right) \right], \tag{5.18} \]

Equation (5.18) gives the plasmon energy below which the optical mode is less lossy than the acoustic mode, and above which the acoustic mode is less lossy than the optical mode. Plugging the physical parameters used for Figure 5.4 into (5.18), we obtain \( h\omega = 0.4E_F \), which accurately pinpoints the intersection in Figure 5.4(b).

Figure 5.4. (a) Dispersion characteristics, (b) \( q_2/k_F \), and (c) \( q_1/q_2 \) of the acoustic mode (black curves) and the optical mode (red curves) of DLG at temperature \( T \to 0 \). Other parameters used are \( \epsilon_1 = \epsilon_2 = 4\epsilon_0 \), \( d = 50 \text{ nm} \), \( \gamma = 4 \text{ ps}^{-1} \), and \( E_F = 100 \text{ meV} \). The dotted curves are numerically calculated by using the Drude conductivity in (5.10). The dashed and solid curves are obtained from (5.15) and (5.16), representing the RPA and RPA–RT results, respectively. The extrinsic scattering is ignored by setting \( \gamma = 0 \) for the RPA curves. The RPA–RT results for SLG plasmon mode are plotted as the grey curve for comparison. In (a), the colored areas are the Landau damping regions. The RPA and RPA–RT curves are overlapped. In (b), the location of the arrow is obtained from (5.18). In (c), the distributions of the longitudinal electric field component \( E_L \) are plotted for the acoustic mode (upper inset) and the optical mode (lower inset) at the plasmon energy \( h\omega = 0.5E_F \). The location of the dot is obtained by solving (5.11) with \( q_1d = 2 \).

To investigate the aforementioned intersection of \( q_2 \) curves, it is intuitive to divide \( q_1 \) by \( q_2 \) as both \( q_1 \) and \( q_2 \) increase with frequency. The value of \( q_1/q_2 \), as shown in Figure 5.4(c),
can be regarded as the Q-factor of a plasmonic spectrum\textsuperscript{107} or a figure of merit for the plasmon propagation distance relative to the plasmon wavelength. As can be seen, the acoustic mode has a larger value of \( q_1/q_2 \) than that of the SLG plasmon, and the optical mode has a lowest value of \( q_1/q_2 \) than that for the acoustic mode and that for the SLG plasmon. The difference in \( q_1/q_2 \) can be explained by the characteristic field distributions of the two modes shown in the insets of Figure 5.4(c), where the distribution of the longitudinal electric field component \( E_L \) for \( \hbar \omega = 0.5 E_{\downarrow} \) is plotted. The \( E_L \) field of the acoustic mode is suppressed between the two graphene sheets, as shown in the top inset. As a result, the Joule loss arising from the current density due to the longitudinal \( E_L \) field is less than that for the SLG plasmon. This phenomenon is manifested by the larger value of \( q_1/q_2 \) for the acoustic mode than that for the SLG plasmon, as shown in Figure 5.4(c). By contrast, the \( E_L \) field of the optical mode is enhanced between the two graphene sheets. As a result, the Joule loss of the optical mode is high, and the optical mode has the lowest \( q_1/q_2 \) compared to both the acoustic mode and the SLG plasmon.

Because the current density is related to the value of \( q_1/q_2 \), we can calculate the current density for the DLG structure to locate the peak of \( q_1/q_2 \) for the acoustic mode with respect to the \( q_1/q_2 \) curve for the SLG plasmon in Figure 5.4(c). We first simplify the problem by ignoring the scattering rate \( \gamma \), which only scales \( q_2 \) and thus \( q_1/q_2 \), as seen from (5.12). By normalizing the electromagnetic power of the acoustic mode, it can be shown that the ratio of the current density on the graphene sheets of DLG to that on a SLG sheet is proportional to

\[
\left[ \frac{\epsilon_1}{\epsilon_2} + \coth \left( \frac{q_1d}{2} \right) \right] / \sqrt{\frac{\epsilon_1}{\epsilon_2} + q_1d \text{csch}^2 \left( \frac{q_1d}{2} \right)} + \coth \left( \frac{q_1d}{2} \right),
\]

which has the minimum value
at \( q_d d = 2 \) in the case of \( \epsilon_1 = \epsilon_2 \). Plugging the physical parameters used for Figure 5.4 and \( q_d d = 2 \) in (5.11), we obtain \( \hbar \omega \approx 0.5 E_F \), which approximately locates the peak of \( q_1/q_2 \) for the acoustic mode with respect to the \( q_1/q_2 \) curve for the SLG plasmon, as shown in Figure 5.4(c).

In the case when \( \epsilon_1 \neq \epsilon_2 \), the value of \( q_d d \) for the minimum value of the current density ratio has to be numerically solved from the relation \( (2\epsilon_1 - \epsilon_2 q_d) \tanh(q_d d / 2) = \epsilon_1 q_d d - 2\epsilon_2 \).

Figure 5.5. (a) Attenuation (\( q_2/k_F \)) and (b) \( q_1/q_2 \) of the acoustic mode (black curves) and the optical mode (red curves) of DLG at \( T = 300 K \). Unless otherwise specified, the parameters used for the calculation are the same as those for Figure 5.4. The line styles of the curves have the same meaning as those for Figure 5.4. In (a), the locations of the intersections for the solid and dotted curves are indicated. In (b), \( \gamma = 10 \text{ps}^{-1} \) is assumed for the bottom two solid curves. The location of the two dots are obtained by solving (5.11) with \( q_d d = 2 \).

The features shown in Figure 5.4(b) and Figure 5.4(c) are also observable at room temperature, as shown in Figure 5.5 for \( T = 300 K \). Unless otherwise specified, other parameters used for Figure 5.5 are the same as those used for Figure 5.4. As can be seen in Figure 5.5(a), the RPA curves start rising at photon energies as low as \( \hbar \omega = 0.2 E_F \) because the Landau damping region is extended into the low-frequency region. For this reason, the Drude model is reasonably
accurate only below $\hbar \omega = 0.2 E_F$. The intersection of the Drude curves for the two plasmon modes can be found from (5.18), which gives $\hbar \omega = 0.38 E_F$. This value serves as an approximation for the intersection of the RPA–RT curves at $\hbar \omega = 0.35 E_F$. The extension of the Landau damping region can also be observed in Figure 5.5(b). In the low-frequency region below $\hbar \omega = 0.2 E_F$, the value of $q_1/q_2$ is primarily determined by the extrinsic scattering rate $\gamma$. By contrast, above $\hbar \omega = 0.2 E_F$ the effect of Landau damping becomes increasingly important to suppress the growth of $q_1/q_2$ value. The peak location of $q_1/q_2$ for the acoustic mode with respect to the $q_1/q_2$ curve for the SLG plasmon can be found by plugging $q_d = 2$ into (5.11), which gives $\hbar \omega = 0.51 E_F$ marked by the dot in Figure 5.5(b). As can be seen, for $\gamma = 4 \text{ps}^{-1}$ the dot does not pinpoint the peak because the condition $q_d = 2$ is derived classically and the effect of Landau damping cannot be accounted for. The dot can pinpoint the peak only when Landau damping is not significant compared to extrinsic scattering. For example, by increasing the scattering rate from $\gamma = 4 \text{ps}^{-1}$ to $\gamma = 10 \text{ps}^{-1}$, the dot approximates the location of the peak more accurately, as shown in Figure 5.5(b).
Figure 5.6. Loss of the acoustic mode for DLG as a function of interlayer spacing \( d \). The attenuation \( q_2 \) is normalized to the Fermi wave number \( k_F = 100 \text{meV}/\hbar v_F \). Unless otherwise specified, the parameters used for the calculation are the same as those for Figure 5.4. All the curves are the RPA–RT results at a fixed plasmon energy indicated next to each curve. The dashed curve is obtained at \( T = 100 \text{K} \) and the dotted curve is obtained by assuming \( E_F = 104 \text{meV} \).

For a fixed frequency, the attenuation parameter \( q_2 \) of the acoustic mode is minimized at an optimum spacing \( d_{\text{opt}} \) between the two graphene sheets. By plugging \( q_1d_{\text{opt}} = 2 \) into (5.11), the optimum spacing \( d_{\text{opt}} \) can be found as

\[
d_{\text{opt}} = \frac{4e^2k_B^2T}{\pi\epsilon\hbar^2\omega^2} \frac{\ln\left[2\cosh\left(\mu/2k_BT\right)\right]}{1+\coth 1},
\]

where \( \epsilon = \epsilon_1 = \epsilon_2 \) is assumed. The attenuation parameter \( q_2 \) of the acoustic mode obtained using (5.15) and (5.16) within the framework of RPA–RT is shown in Figure 5.6 as a function of \( d \). The solid curves are obtained using the same parameters as used for Figure 5.4 except that \( d \) is now the variable. The optimum spacing obtained from (5.19) is marked in Figure 5.6 for each curve. As can be seen, the dots accurately mark the locations of minimum attenuation loss for all curves. For comparison, the dashed curve is plotted for \( T = 100 \text{K} \) and the dotted curve is
obtained by setting $E_F = 104\text{meV}$, with other parameters being the same as those used for the top solid curve. As can be seen, the loss increases with temperature but reduces with Fermi energy at fixed $d$ and fixed plasmon energy.

Figure 5.7. (a) Attenuation $q_2/k_F$ as a function of surface plasmon energy, and (b) as a function of the interlayer distance $d$ for 3-layer (solid curves) and 4-layer (dashed curves) Coulomb-coupled graphene sheets. The graphene sheets are equispaced at a distance of $d = 50\text{nm}$. Other parameters are the same as those for Figure 5.4. All the curves are the RPA–RT results, and curves in different colors represent different modes. In the inset of (a), the corresponding $q_1$ from $\hbar \omega = 0$ to $0.5E_F$ is shown. The colored area is the intraband Landau damping region. In (b), selected modes in (a) are plotted in the corresponding styles. Curves obtained for the same plasmon energy are grouped by an ellipse with the plasmon energies indicated.

The characteristic behaviors of $q_2$ for the plasmons of DLG structures shown in Figure 5.4(b) and Figure 5.6 are also present in 3-layer and 4-layer Coulomb-coupled graphene. As can be seen in Figure 5.7(a), the intersection of $q_2$ curves for different plasmon modes also exists for these multilayer graphene systems. Except for the optical mode, there is an optimum interlayer spacing $d_{\text{opt}}$ for minimum $q_2$. Selected modes in Figure 5.7(a) are plotted in Figure 5.7(b) in corresponding styles as a function of interlayer spacing $d$ at three fixed plasmon energies of
\( \hbar \omega = 40 \text{meV}, 50 \text{meV}, \text{and} 60 \text{meV}. \) As can be seen, for most of the modes the optimum interlayer spacing exists, which varies for different modes and different plasmon energies.

### 5.3. Graphene-based THz devices

The majority of the proposed graphene-based THz devices consist of a metamaterial that can optically interact with graphene.\(^9, 10, 83, 84, 110\) This coupled graphene-metamaterial system gives rise to a family of resonant modes such as the SPP modes of graphene,\(^74, 83, 84, 95, 111\) the geometrically induced SPPs, also known as the spoof SPP modes,\(^9, 10, 110, 112-115\) and the Fabry-Perot (FP) modes.\(^116-119\) Even in a simple metamaterial structure such as a one-dimensional (1D) metallic slit grating, these modes all exist and can potentially interact with each other. As an example, a graphene-metamaterial hybrid system is shown in Figure 5.8(a), where \( \epsilon_d, \epsilon_m, \epsilon_1, \epsilon_2, \) and \( \epsilon_3 \) are the permittivities of the dielectric gap filling, the medium that is used to construct the metamaterial, the medium below the metamaterial, the medium below graphene, and the medium above graphene, respectively; \( d_1 \) is the thickness of the metamaterial; \( d_2 \) is the distance between the graphene sheet and the metamaterial; other structural parameters charactering the miniature patterns of the metamaterial are also specified, such as the width \( w \) and the height \( h \) of the engraved grooves. If the structure is periodic, the periods in two directions and the unit cells can be defined. Some of the patterns studied in the literature are shown in the insets of Figure 5.8(a); from left to right are 1D grating,\(^83, 84\) two-dimensional (2D) holes,\(^120, 121\) rings,\(^9\) and split ring resonators (SRR).\(^10, 114, 115\) Upon the incidence of an optical field, the induced locally oscillating fields on each unit cell emulate the atomic resonance. The corresponding bound modes also exist for a metamaterial made of a perfect conductor because the electric field can still penetrate inside
the material through holes or grooves in the structure, resembling the decaying field of the true metal SPPs. Therefore, these geometrically induced modes are also known as spoof SPP modes. The resonance frequency $v$ of a spoof SPP mode is determined by the design parameters of the structure. For normally incident radiation of a wavelength $\lambda$ that is much larger than the structural dimensions of the metamaterial patterns, the resonance frequencies of 1D grooves with $h < d_1$, 1D slits with $h = d_1$, 2D holes, and 2D rings, as shown in the insets of Figure 5.8(a), are given approximately by

\[ v \approx \frac{c}{4n_{\text{eff}}d_1}, \quad v \approx \frac{c}{2n_{\text{eff}}d_1}, \quad v \approx \frac{c}{2n_{\text{eff}}w}, \quad v \approx \frac{c}{2\pi r n_{\text{eff}}}, \]

respectively, where $n_{\text{eff}}$ is the effective index of the mode and $r$ is the radius of the ring. If the ring is cut so that the symmetry is broken, the resulting SRR structure can be regarded as an LC circuit with a new resonance frequency of $v \approx \frac{c}{2\pi \sqrt{LC}}$. Besides these spoof SPP modes, the periodic structure of the metamaterial also provides the necessary phase-matching momentum for the excitation of metal SPP modes and graphene SPP modes. To reduce the complexity without loss of generality, in the following we consider a periodic slit grating as the metamaterial structure for the discussion of the coupling effect and the dispersion characteristics of all possible modes of a graphene-metamaterial structure.
Figure 5.8. (a) Sketch of a graphene-metamaterial structure. The miniature structure of the metamaterial can take the form of periodic slits, holes, rings, or SRR, as shown in the insets. By coupling graphene with a metamaterial surface, (b) a graphene SPP mode, (c) a spoof SPP mode, or (d) a FP mode can be excited by normally incident light. For all figures, \( \varepsilon_d, \varepsilon_m, \varepsilon_1, \varepsilon_2, \) and \( \varepsilon_3 \) are the permittivities of the gap filling, the metamaterial, the medium below the metamaterial, the medium below graphene, and the medium above graphene, respectively; \( d_1 \) is the thickness of the metamaterial; \( d_2 \) is the distance between the graphene sheet and the metamaterial. In the case of a 1D grating structure being the metamaterial structure as shown in (b)-(d), \( \Lambda \) is the period, and the gap is characterized by a width of \( w \) and a height of \( h \). The representative transmittance curves as a result of the excitation of graphene SPP, spoof SPP, and FP modes are shown in the insets in (b)-(d), respectively, calculated using the classical modal method.\(^{89}\) The parameters are \( h = d_1, \ w = 0.1 \ \mu m, \ \Lambda = 3 \ \mu m, \) chemical potential \( \mu_1 = 100 \ \text{meV}, \) polarization angle \( \phi = 0^\circ, \) scattering rate \( \gamma = 1 \ \text{ps}^{-1} \) for the graphene sheet,\(^{26}\) \( \varepsilon_3 = \varepsilon_0, \) and \( \varepsilon_d = \varepsilon_1 = \varepsilon_2 = 3.9 \varepsilon_0, \) where \( \varepsilon_0 \) is the permittivity of free space. The Drude model is used for the graphene sheet, and the metal is assumed to be a perfect conductor. The increased chemical potentials used for the insets in (b)-(d) are \( \mu_2 = 260 \ \text{meV}, \ 200 \ \text{meV}, \) and \( 400 \ \text{meV}, \) respectively. In (b), \( d_1 = d_2 = 10 \ \text{nm}. \) In (c), \( d_1 = 25 \ \mu m, \ d_2 = 10 \ \text{nm}. \) In (d), \( d_1 = 10 \ \text{nm}, \ d_2 = 25 \ \mu m. \)

For a graphene-metamaterial structure that consists of a metallic slit grating, there are graphene SPP modes, spoof SPP modes of the metallic grating, and FP modes supported by the graphene and the metallic surfaces, as shown in Figure 5.8(b-d), respectively, for the case of a 1D grating. For normal incidence from above with an optical field that is linearly polarized at an azimuth angle of \( \phi = 0^\circ, \) the corresponding characteristic transmittance curves are shown in the
insets next to each figure for two different graphene chemical potentials of $\mu_1 = 100\text{meV}$ and $\mu_2 > \mu_1$. By increasing the chemical potential, the average kinetic energy of electrons in graphene increases, and the resonance frequency of a graphene SPP mode, corresponding to the transmittance trough, is blue-shifted, as shown in the inset of Figure 5.8(b). A graphene SPP mode can also couple with a spoof SPP mode of the metallic grating when the spoof SPP mode is supported by the grating structure. As a result, the spoof SPP mode is split into two coupled modes, as can be seen in the inset of Figure 5.8(c) for two transmittance peaks. When the graphene sheet is placed sufficiently far away from the metallic grating, FP modes are supported between the graphene sheet and the metallic grating. By moving the Fermi level away from the Dirac point, the graphene sheet changes from weakly conducting to highly conducting. As a result, the boundary condition on the graphene side of the FP cavity is modified, and the corresponding resonance frequency of the FP mode changes accordingly. Thus, from the insets of Figure 5.8(b-d), we find that by adjusting the chemical potential of graphene, the resonance frequency of the relevant surface modes can be tuned and thus the transmitted signal can be switched on and off.

**Numerical simulation**

The dispersion curves of graphene SPP, spoof SPP, and FP modes can be found by locating the resonance frequency in the transmittance spectrum. For normally incident light, the dispersion map calculated using the modal method\textsuperscript{89} are plotted in Figure 5.9 as a function of $d_1$ (right panel) and $d_2$ (left panel). The parameters are the same as those used for the insets in
Figure 5.8(b-d). Three regions are clearly seen where graphene SPP, spoof SPP, and FP modes are respectively dominant. In the case of \( w = \Lambda \), the resonance frequency \( \omega \) of the \( m \)th-order SPP mode is given by\(^7\)

\[
\epsilon_2 + \left( \epsilon_3 - \frac{e^2|\mu|}{\pi \hbar^2 \omega^2} \right) \tanh mKd_2 = 0,
\]

(5.20)

where \( K = 2\pi/\Lambda \) is the wave number of the periodic metallic grating that has a period of \( \Lambda \). In (5.20), the Drude model for graphene is used, and the losses assumed to be negligible. The metal is assumed a perfect conductor, which is a good approximation in the THz spectral region. The dispersion curves of graphene SPP modes solved by using (5.20) are plotted in Figure 5.9(a); the dotted curves labeled (b) and (c) are for the first- and second-order graphene SPP modes. These curves follow the transmittance troughs that correspond to the location of the SPP excitation. The fields of these graphene SPP modes are confined on the graphene sheet, as shown in Figure 5.9(b) and Figure 5.9(c) for the first and second orders, respectively. Above 10 THz in Figure 5.9(a), the dispersion curves are no longer observable because the Drude conductivity of graphene that supports these SPP modes drops significantly in the high frequency region.

As the optical frequency increases, the excitation of the metal SPP mode becomes possible. In the case of \( \epsilon_m \to -\infty \) for metal in the THz spectral region, the metal SPP mode satisfies the Rayleigh condition \( \lambda = \sqrt{\epsilon_1 \Lambda} \),\(^1\) which gives a transmittance trough at \( \nu = c/\sqrt{\epsilon_1 \Lambda} = 50.64 \) THz in Figure 5.9(a). For a finite but large \( \epsilon_m \), for example \( \epsilon_m = -260\epsilon_0 \), the coupling of the two surfaces of metallic grating becomes possible for a small thickness \( d_1 \); then, the dispersion curve is altered as shown in Figure 5.9(d) for the location marked by a white box labeled (d) in Figure 5.9(a). A sharp resonance characterized by a transmittance maximum
followed by a transmission minimum as shown in Figure 5.9(d) is a characteristic feature of a Fano resonance supported by a metallic grating. A representative electric field distribution of the metal SPP mode is plotted in Figure 5.9(e).

Figure 5.9. (a) Transmittance as a function of $d_1$ and $d_2$ calculated using the classical modal method. Other physical parameters used for the simulation are the same as those used for the insets in Figure 5.8(b-d). Areas of the dominant modes are separated approximately by the white dashed curves. The dotted curves for the first two orders of graphene SPP modes and the spoof SPP modes are obtained from (5.20) and (5.21), respectively. The dotted curves for the FP modes are given by $\nu = mc/2n_d^2$, where $m$ is a positive integer. The dispersion curves of the high-order FP and spoof SPP modes are not plotted as the linewidth become exponentially thin below the resolution of the figure. (b-c) The electric field $E_x$ of the graphene SPP modes for $d_1 = d_2 = 10$ nm and $\nu = 0.6$ THz and 1.2 THz labeled (b) and (c) in (a), respectively. (d) The metal SPP dispersion curve for the region marked by the white box labeled (d) in (a) assuming $\epsilon_m = -260 \epsilon_0$. (e) Field distribution of $E_x$ for the parameters of $d_1 = 12.6$ nm and $\nu = 47$ THz marked by (e) in (d). (f) Field distribution of $E_x$ for the parameters of $d_1 = 25$ µm and $\nu = 2.9$ THz labeled (f) in (a). The structure shown is not to scale. (g) Field distribution of $E_x$ for the parameters of $d_2 = 25$ µm and $\nu = 2.4$ THz labeled (g) in (a). The metallic grating is not discernible in (e) and (g) because of its small thickness $d_1$ compared to the dimensions of the graphs.
As the thickness $d_1$ of the grating increases, the excitation of spoof SPP modes in THz spectral region becomes possible. We consider the first-order grating because the effect of the high-order gratings is hardly observable in the transmission spectrum.\textsuperscript{120, 121} For a first-order grating, the dispersion of the spoof SPP modes in the long-wavelength limit of $\lambda \gg \Lambda$ and $\lambda \gg w$ is given by\textsuperscript{121}

$$\frac{\sqrt{K^2 - k_d^2}}{k_d} = \pm \frac{w}{\Lambda} \left[ \tan \left( k_d d_1 / 2 \right) \right]^{\pm 1}, \quad (5.21)$$

where $k_d = n_d \omega / c$ and $n_d = \sqrt{\varepsilon_d / \varepsilon_0}$. Because of the periodicity of the tangent functions, (5.21) gives two sets of solutions when plus and minus signs are taken respectively. When $K \gg k_d$, the left-hand side of (5.21) approaches infinity, and thus the solution is found by requiring the argument $k_d d_1 / 2$ on the right-hand side of (5.21) to be multiples of $\pi / 2$, which gives $\nu = mc / 2 n_d d_1$ for the resonance frequency of the $m$th-order mode. The first two modes are plotted as dotted curves labeled (f) as a group, which agree well with the simulation results in the right panel of Figure 5.9(a). In the low-frequency region where the coupling with the metal SPP mode is negligible, the field is confined inside the gap, as shown in Figure 5.9(f). In this case, a spoof SPP mode is also called a cavity mode, which plays an important role in extraordinary optical transmission (EOT).\textsuperscript{113, 122} Spoof SPP modes can also couple with graphene SPP modes to open up gaps in the dispersion curves, as indicated by the white arrows in Figure 5.9(a). The location of the gaps can be tuned by adjusting the chemical potential of graphene; thus, the transmittance of the graphene-metamaterial structure can be tuned accordingly, as shown in Figure 5.8(c).
When the wavelength of light is comparable to the distance $d_2$, a FP cavity is formed between the graphene and the metallic grating with the boundary conditions determined by the chemical potential of graphene. The $m$th-order resonance frequency of a FP cavity characterized by a dielectric constant $n_2$ and a thickness $d_2$ is given by $v = mc^2/2n_2d_2$, where $m$ is a positive integer. The first two modes are plotted as dotted curves labeled (g) as a group in the left panel of Figure 5.9(a). The curves approximately agree with the simulation results. The discrepancy is due to the finite conductance of graphene and the unbalanced dielectric indices $n_1$ and $n_3$ that make the FP cavity imperfect. The transmittance is tuned by adjusting the chemical potential of graphene to achieve the functionality of the device shown in Figure 5.8(d).

**Experimental demonstration**

To demonstrate the concept device based on (5.20) as well as the simulation results shown in Figure 5.9, we fabricate a structure shown in Figure 5.8(b). 1D and 2D grating structures of different periods $\Lambda$ are fabricated with other structural parameters kept unchanged for all structures, as shown in Figure 5.10(a). Terahertz time-domain spectroscopy (THz-TDS)\(^{19}\) is used to measure the transmitted electric field through the grating, $E_g = E_g(\cos\phi \hat{x} + \sin\phi \hat{y})$, and through the free space without the grating, $E_{air}$, at room temperature, where $\phi$ is the azimuth angle of the incident light. The values of the ratio $\text{Re} E_g / \text{Re} E_{air}$ are plotted as dotted curves in Figure 5.10(b) for two different 1D gratings that have periods of $\Lambda = 3.9$ µm and $\Lambda = 4.9$ µm, respectively. For the 2D grating, the values of the transmitted electric field ratio $\text{Re} E_g / \text{Re} E_{air}$
for the incident THz fields polarized at the angle of $\phi = 0^\circ$ along the long period of $\Lambda = 4.9 \ \mu\text{m}$ and at the angle of $\phi = 90^\circ$ along the short period of $\Lambda = 1.5 \ \mu\text{m}$, respectively, are plotted as solid curves in Figure 5.10(b). The agreement between the experimental data and the simulation results shows the good quality of our grating structures.

Monolayer graphene grown by chemical vapor deposition (CVD) is then transferred onto the grating structure so that we have a system shown in Figure 5.10(b). The transmitted electric field of the graphene-grating system $\text{Re} \ E_{gg}$ is divided by $\text{Re} \ E_g$ so that the resonance locations of the graphene SPP mode can be observed. The experimental and simulation results are plotted in Figure 5.10(c) and Figure 5.10(d), respectively. The simulation is carried out using the fitting parameters of $\mu = 60 \ \text{meV}$ and $\gamma = 16 \ \text{ps}^{-1}$ except for the period $\Lambda = 4.9 \ \mu\text{m}$ of the 2D grating, which is fitted using a scattering rate of $\gamma = 29 \ \text{ps}^{-1}$. The high scattering rate might be due to the ripples caused by the uneven surface of the 2D grating structure, or due to angle-sensitive scattering centers such as grain boundaries that prefer to grow in a certain direction resulting in a high scattering rate at specific polarization angles. By rotating the angle from $0^\circ$ to $90^\circ$ for the 2D sample, as plotted in Figure 5.10(e), a smooth transition is shown for the curves of $\text{Re} \ E_{gg}/\text{Re} \ E_g$ at different angles. This further indicates the angle-dependent scattering rate and the polarization-sensitive nature of our 2D structure. It can also be seen that the resonance frequency of the SPP mode shifts to a higher frequency for a shorter grating period. In our case where $Kd_2 \ll 1$, the resonance frequency varies as $\omega \propto \Lambda^{-1}$ according to (5.20). Therefore, the resonance frequency increases with decreasing grating period, which is consistent with the
experimental results. The exact location of resonance can be found by modifying (5.20) to account for a finite scattering rate.

The effect of Drude conductivity is observed using Fourier-transform infrared spectroscopy (FTIR). The transmittance of 1D gratings with periods of $\Lambda = 1.5 \, \mu m$ and $\Lambda = 3 \, \mu m$ are measured with and without the graphene layer. As can be seen in Figure 5.10(f), the differences in the transmittance, which are shown as the green areas, indicate the frequency region where the Drude conductivity is significant. Because the graphene SPP mode is supported by the Drude conductivity, these differences shrink quickly as the frequency increases, indicating that the excitation of the graphene SPP mode at high frequencies gradually becomes difficult, which is consistent with the simulation results shown in Figure 5.9(a). Therefore, the operational range of the graphene SPP-based THz device shown in Figure 5.8(b) is limited not only by intrinsic Landau damping, but also by the frequency-dependent nature of the Drude conductivity of graphene.
Figure 5.10. (a) Images of scanning electron microscope for 1D and 2D gratings. The 1D gratings shown in Figure 5.8(b) are characterized by the parameters: $w = 0.4 \mu m$, $d_1 = 50 \text{nm}$, $d_2 = 30 \text{nm}$, $\epsilon_3 = \epsilon_0$, $\epsilon_2 = 3.9 \epsilon_0$ for silicon dioxide, $\epsilon_1 = \epsilon_d = 11.68 \epsilon_0$ for silicon, $\Lambda = 3.9 \mu m$ and 4.9 $\mu m$, respectively, for two samples. The 2D structure is the same as the 1D structures except that the 2D structure is also periodic in the $y$ direction. The long and short periods are $\Lambda = 4.9 \mu m$ and 1.5 $\mu m$, respectively. These metallic gratings are made of titanium nitride (TiN), represented by the yellow regions in the insets. (b) Ratio of the real part of the transmitted field through gratings, $E_g$, to that of the free space without the grating, $E_{air}$. For the 2D grating (solid curves), both $\phi = 0^\circ$ and 90$^\circ$ for the polarization along the periods of $\Lambda = 4.9 \mu m$ and 1.5 $\mu m$, respectively, are measured. For the 1D gratings (dashed curves), only $\phi = 0^\circ$ is measured. Theoretical curves, plotted as grey curves, are calculated using the classical modal method$^{89}$ for the 1D structures, and using COMSOL software for the 2D structure. A conductivity of $\sigma = 6000 \Omega^{-1}\text{cm}^{-1}$ is taken for TiN.$^{123}$ (c) Ratio of the measured real part of the transmitted field through graphene-grating structure, $E_{gg}$, to the real part of $E_g$. (d) Simulation results for (c). The fitting parameters are $\mu = 60 \text{ meV}$ and $\gamma = 16 \text{ ps}^{-1}$ except for the period $\Lambda = 4.9 \mu m$ of the 2D
grating, which is fitted using the scattering rate $\gamma = 29$ ps$^{-1}$. (e) $\text{Re} E_{gg}/\text{Re} E_g$ for different values of the polarization angle $\phi = 0^\circ, 35^\circ, 45^\circ, 65^\circ, \text{and} 90^\circ$. The arrow indicates the direction of increasing angles. (f) Transmittance measurement of FTIR for 1D gratings with (thick curves) and without (thin curves) graphene on top; the green areas show the transmittance difference. Two different samples of periods $\Lambda = 3 \, \mu\text{m}$ and $1.5 \, \mu\text{m}$ are measured.
Chapter 6. Conclusion

In this thesis, we have discussed numerous physical properties of graphene. In Chapter 1, some basics of graphene and multilayer graphene are discussed, such as the band structures, the density of states, and the carrier concentration. These physical properties are repeatedly used in the later chapters. In Chapter 2, various scattering mechanisms are discussed. The dependence of the scattering rates on carrier energy, temperature, Fermi energy, and the surrounding permittivities is investigated. It is found that the relative importance of the scattering mechanisms highly depends on the substrate beneath the graphene sheet. It is also found that the mobility of AA-stacked multilayer graphene can be potentially higher than that of monolayer graphene because of the high carrier density and the high screening effect of the AA-stacked multilayer graphene. The scattering rate derived in Chapter 2 is then used as a parameter in the Drude model, which is introduced in Chapter 3. The optical conductivity of monolayer and multilayer graphene in the terahertz spectral region is experimentally measured using terahertz time-domain spectroscopy. The physical parameters are extracted by fitting the measured data with the Drude model. It is found that the THz optoelectronic properties of multilayer graphene can be tuned by purposefully misorienting layers or employing different stacking schemes. To describe the plasmonic property of graphene and the characteristics of the SPP modes, we modify the Drude model to account for the spatial nonlocality in Chapter 4. The great tunability of SPP characteristics is shown by changing the surrounding dielectric constant, the coupling distance, and the plasma frequency of the substrate. We also show that the dispersion of the SPP modes for graphene on a grating can be well approximated by using the photonic crystal model, and therefore a full simulation is not needed. Finally, by understanding how graphene interacts
with the electromagnetic wave and its immediate surroundings as discussed in Chapter 2-4, graphene-based THz devices can be modeled and the performance can be simulated, as presented in Chapter 5. It is shown that by placing the graphene sheet near a plasma substrate such as metal or another graphene sheet, the confinement is enhanced and the attenuation length is increased. The propagation length divided by the plasmonic wavelength is estimated by the figure of merit $q_1/q_2$. For the double-layer graphene waveguide and the graphene-metal hybrid waveguide of a scattering rate $\gamma = 4\text{ps}^{-1}$, it is shown that in the THz spectral region $q_1/q_2$ is around 20 to 30, which is comparable to that of a metal waveguide in the visible and ultraviolet spectral regions. If the graphene sheet is perfectly clean without impurity and defects, the scattering rate can be as low as $1\text{ps}^{-1}$, limited by the intrinsic acoustic phonon scattering. This ultra-low scattering rate results in a giant electron mobility in the order of $10^5 \text{cm}^2/\text{V s}$ as confirmed by the experiment. As the technology for graphene synthesis becomes mature with large-size and high-quality graphene available in the future, a graphene-based waveguide with $q_1/q_2$ above 100, or the propagation length of few micrometers for $E_f < 100\text{meV}$, is foreseeable.

From the numerical analysis in Chapter 5, we also find that the RPA model is more accurate than the Drude model, especially when the wave number is comparable to the Fermi wave number near or inside the interband and intraband scattering regions (Landau damping regions), where the scattering events are not accounted for by the Drude model. If the excited SPP has a wave number much smaller than the Fermi wave number, then the Drude model is sufficiently accurate. In such a case, the consideration of the spatial nonlocality is not necessary, and the Drude model can be used directly in the standard EM solvers, i.e., FDTD or FEM models. In Chapter 5, a graphene SPP-based THz device is also fabricated and measured. The
experimental results show a consistent trend between the SPP resonance frequency and the grating period, as predicted by our theoretical analysis. We also find that the excitation of the graphene SPP mode is most efficient in the terahertz spectral region due to the Drude conductivity of graphene in this spectral region.

Overall, in this thesis we have acquired a fundamental understanding of the optoelectronic properties of graphene, and obtained a better understanding of coupling of graphene plasmons with extrinsic quasiparticles. Equipped with these knowledge and various physics models that have been successfully applied in the experiments, we are one step closer toward the realization of more sophisticated graphene-based THz devices of better performance. For example, by using the conductivity derived in this thesis with appropriate boundary conditions similar to those used for the grating structure in Chapter 5, the resonance frequency of graphene nano-antennas can be approximated,\textsuperscript{125} although not discussed in the thesis, graphene nano-antenna is also a major class of proposed THz applications of graphene.\textsuperscript{126-128} Research of other devices can also benefit from our research, such as low-loss THz waveguides and highly functional THz modulators and detectors, which are essential and necessary components to realize the wireless communication system in the THz spectral region.
References


