Engineering Plasmonic Waves in Two-Dimensional Electron Systems

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Engineering Plasmonic Waves in Two-Dimensional Electron Systems

A dissertation presented

by

Yan Mui Kitty Yeung

to

The School of Engineering and Applied Sciences

in partial fulfillment of the requirements

for the degree of

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Engineering Plasmonic Waves in Two-Dimensional Electron Systems

Abstract

Plasmonic waves are waves of mobile charge carriers caused by their collective oscillations. They can be excited in solid-state conducting materials and behave distinctively in different numbers of dimensions. With fabrication technologies available for solid-state materials, one can functionalize the dimensional properties by engineering the boundaries and interfaces of the plasmonic wave medium. For instance, plasmonic waves in two-dimensional (2D) conductors, such as semiconductor heterojunction and graphene, exhibit strong subwavelength confinement – with a wavelength about a factor of 100 below the electromagnetic wavelength at the same frequency. Hence, 2D plasmonic devices can be constructed below the diffraction limit of light. To utilize this ultra-subwavelength confinement is the main motivation of this thesis.

This thesis establishes the machinery behind the unique behaviors of 2D plasmons, and compares them to plasmons in higher dimensions, namely plasma oscillations in bulk materials and surface plasmons on conducting-insulating interfaces. The Coulomb restoring force and mobile charge carrier inertia causing the collective oscillations are formulated into a transmission-line model. This formulation is used to engineer ultra-subwavelength plasmonic circuits in gigahertz integrated electronics and terahertz metamaterials.
As one of the demonstration platforms, we use GaAs/AlGaAs 2D electron gas. Amongst a variety of devices, the thesis focuses on an on-chip solid-state 2D plasmonic Mach-Zehnder interferometer operating at microwave frequencies. The gated 2D plasmonic waves achieve a velocity of \(~c/300\) (\(c\): free-space speed of light). Due to this ultra-subwavelength confinement, the resolution of the 2D plasmonic interferometer is two orders of magnitude higher than that of its electromagnetic counterpart at a given frequency.

Another material we use, which hosts mobile charge carriers in 2D, is graphene. We fabricate metamaterials in the form of graphene plasmonic crystals in a continuous graphene sheet with periodic structural perturbations. Plasmonic bands in the far-infrared are formed and excited via symmetry-based selection rules, in a manner akin to photonic crystals. The plasmonic bands can be engineered by manipulating the charge carrier concentration, the dimensions of the periodic lattice, the shape of the perturbation and the lattice symmetry. These demonstrations may generate new avenues for a wealth of subwavelength graphene plasmonic devices, such as band gap filters, modulators and switches.
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Citations to Previously Published Work

Large portions of this thesis have appeared in the following papers in publication:


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Dedicated to my parents and grandparents.
Chapter 1

Introduction

Plasmas appear in various forms in nature, with the collective electron density waves, or plasmonic waves, serving as a salient dynamic feature. Solid-state plasmas consisting of mobile electrons in conducting materials are especially interesting, as the fabrication technologies available for solid-state materials allow us to design the boundaries and interfaces of the plasma media in order to engineer the plasmonic waves. In particular, surface plasmons on three-dimensional (3D) bulk metals, as an active subject of photonics research, can achieve a velocity as low as \( \sim c/10 \) (\( c \): free-space speed of light), with a proportionally reduced wavelength [1]. Therefore, one can make the surface plasmons to react to light in structures smaller than the diffraction limit of light. While surface plasmons on 3D bulk metals typically appear in the optics regime, plasmonic waves in 2D conductors, such as GaAs/AlGaAs 2D electron gas (2DEG) and graphene, can appear at gigahertz (GHz) to terahertz (THz) frequencies [2-8]. These 2D plasmons exhibit a greater subwavelength confinement, with velocities far below \( \sim c/100 \) [6].
In this thesis, we harness this 2D plasmonic wave ultra-subwavelength confinement in 2DEG and graphene to make various devices from microwave to infrared frequencies.

Prior to the work presented in this thesis, our research group has demonstrated a variety of ultra-subwavelength devices\(^1\) based on geometry-shaping of GaAs/AlGaAs 2DEG. This set of circuits includes a 2DEG plasmonic cavity, in which standing plasmonic waves are trapped at frequencies defined by the cavity size; plasmonic crystals that can function as bandgap filters, in which a number of cavities are connected in series, with a mode-area reduction of up to 440,000-fold relative to their electromagnetic counterparts; interferometers that integrate and allow interference between electromagnetic and plasmonic waves; and negative refractive-index metamaterials that produce negative refractive indices as large as -700.

This thesis starts with the formalism used to develop these plasmonic circuits in general and proceeds to focus on a Mach-Zehnder interferometer operating on the interference between 2D plasmonic waves. Such an interferometer is motivated by efforts to develop surface plasmonic interferometers [9-12]. A prominent advantage of surface plasmonic interferometers is their high resolution due to subwavelength confinement, which makes surface plasmonic interferometers more sensitive to the path length difference and the surrounding dielectric media, leading to higher resolution than their electromagnetic counterparts in measurements of physical quantities such as length and dielectric constant. With greater subwavelength confinement than surface plasmons, the 2D plasmonic interferometer has even higher resolution and sensitivity. We develop at microwave frequencies in the electronics regime, drawing a parallel line of development to surface plasmonic interferometers in photonics.

\(^1\)These examples are published in [6] by W. F. Andress, et al., in which the author participated in the design and analysis of the devices, and in [7] by H. Yoon, et al., in which the author participated in the data analysis.
The next demonstrational material is graphene. In addition to the subwavelength confinement enabled by graphene’s low dimensionality [8, 13-20], graphene plasmons also feature technologically significant attributes such as voltage-controlled tunability [19, 20] and their peculiar dispersion relation, arising from graphene’s unique electronic band structure [21, 22]. The first observation of plasmonic resonance in graphene with far-infrared excitation [8] has spurred a surge of efforts to engineer graphene plasmons [13-18]. These efforts have largely focused on localized plasmonic resonance within a specifically shaped graphene “island” such as a ribbon [8, 13-15], a ring [16, 17], or a disk [16-18], where the boundary condition set by the island geometry determines the resonant frequency. While these foundational works arrange the graphene islands in periodic arrays, the array has served only as a structural repetition to enhance gross plasmonic absorption by summing up each independent localized effect.

As an exception, it has been shown with an array of disk islands [16] that electrostatic coupling between proximate islands offers an additional mechanism to control the localized plasmonic resonance frequency in each island. This result suggests one way to engineer the global dynamics across an array of graphene islands by exploiting its periodicity, despite the plasmon localization. In fact, engineering wave dynamics by medium periodicity is one hallmark paradigm to create wave-based devices. In contrast to reference [16], this principle can be applied to delocalized plasmons in a continuous graphene medium with a periodic structural perturbation, so as to pursue plasmonic band engineering in a manner akin to photonic crystals [23].

We create graphene plasmonic crystals by introducing a lattice of apertures in a graphene sheet. Delocalized plasmons interact with the medium periodicity, forming a plasmonic band structure. This is demonstrated by resonantly coupling far-infrared light to particular plasmonic
modes belonging to a unique set of plasmonic bands, where the light selects these specific modes because the spatial symmetry of the radiation field matches that of the plasmons within those modes. The band formation and excitation are controlled by the carrier concentration of graphene, the shape and dimensions of the periodic apertures and the lattice symmetry. Our work demonstrates a step toward graphene plasmon band engineering, paving the way for novel graphene plasmonic devices.

Before going into details, the remaining sections of this chapter will discuss the features of 2D plasmonic waves that differentiate themselves from plasmonic waves in higher dimensions. We will establish a self-consistent transmission-line model that captures the key characteristics in the 2D plasmonic systems in Chapter 2. This model is used to design plasmonic structures in the subsequent chapters. The model is also applied to surface plasmons in order to elucidate how they differ from 2D plasmons. We will introduce and analyze the 2DEG plasmonic interferometer in Chapter 3 and graphene plasmonic crystals in Chapter 4. The methods we use to engineer graphene plasmonic crystal band structures are demonstrated in Chapter 5. The experimental setups and measurement procedures specific for 2DEG and graphene plasmonic waves are recorded in Chapter 6.

1.1 Plasmons in 3D

Plasmons are excited when mobile charge carriers of one type (either positive or negative) collectively oscillate. In solid-state conductors, mobile charge carriers can be confined within the materials in different dimensions. The dimensionality of the confinement determines the behavior of the plasmonic waves.
In a 3D bulk metal, plasmonic oscillation is a well-known phenomenon and can be explained in a classical framework. Take electrons as an example of the mobile charge carriers, as illustrated in Fig. 1.1a. When they are displaced from their original positions by \( \vec{x} \), a net positive charge is left over in the volume vacated by the electrons. This separation of charges induces an electric field \( \vec{E} = (en_{3D}/\kappa\varepsilon_0)\vec{x} \), and thus a Coulomb restoring force \( -e\vec{E} \), which pulls the electrons back to their original positions (\( e \): elementary charge; \( n_{3D} \): number of electrons per unit volume; \( \kappa \): relative dielectric constant of the metal; \( \varepsilon_0 \): vacuum permittivity). However, since the electrons have inertia, they overshoot to the other side of their original position. The same procedure then repeats in the opposite direction. In this way, the electrons slosh back and forth collectively. The electron displacement obeys the harmonic oscillator equation \( \ddot{x} = -\omega_p^2x \), where 

\[
\omega_p = \sqrt{\left(n_{3D}e^2\right)/\left(m^*\kappa\varepsilon_0\right)}
\]

is the natural angular frequency for the collective oscillations known as the plasma frequency (\( m^* \): effective electron mass in metal). In typical metals, \( \omega_p \) lies in the optical regime.

Another kind of 3D plasmonic mode can exist along a conductor-insulator interface (Fig. 1.1a). Instead of a bulk oscillation, a propagating plasmonic wave can be excited. Take the surface of a metal in air as an example. The metal has a relative dielectric constant 

\[
\kappa_m(\omega) = \kappa_\infty - \frac{\omega_p^2}{\omega^2},
\]

assuming the metal is lossless (\( \kappa_\infty \): background dielectric constant). The air has a relative dielectric constant \( \kappa_d \). When a transverse-magnetic electromagnetic (EM) wave is incident on the metal surface, it decays exponentially from the surface into the metal. The thickness underneath the metal surface for the EM wave amplitude to decay by a factor of an Euler’s number is called the penetration depth, \( \delta \). The mobile electrons that can respond to the
electric field are thus concentrated within a distance $\delta$ of the surface. When the incident EM wave frequency is close to the surface plasmon resonant frequency $\omega_{sp} = \omega_p / \sqrt{\kappa_p + \kappa_d}$ [24], the mobile electrons move along the field direction in the form of a plasmonic wave as a result of the Coulomb restoring force and the electron inertia. The value of $\delta$ reduces with increasing frequency. The number of mobile electrons decreases. To maintain the same current, fewer electrons need to move faster. Therefore, the plasmonic wave gains more kinetic energy as the frequency approaches $\omega_{sp}$. At $\omega_{sp}$, $\kappa_d$ and $\kappa_m$ have equal magnitude and opposite signs. The dielectric constant near the interface, $(\kappa_m + \kappa_d)/2$, experienced by a propagating EM wave is effectively zero. When the frequency is too low, the electrons cannot be collectively driven. There is not enough kinetic energy to excite plasmonic waves. In this case, only the EM wave propagates in the dielectric along the interface. At intermediate frequencies, the coupling between photons and plasmons produces a hybrid mode called the surface plasmon polariton.

These behaviors of the surface plasmons are described by the dispersion relation (Fig. 1.1b) with the wavenumber $k_{sp}(\omega) = \frac{\omega}{c} \sqrt{\frac{\kappa_m(\omega)\kappa_d}{\kappa_m(\omega) + \kappa_d}}$. The slope of the dispersion relation indicates the propagation speed of the surface waves. At low frequencies, when surface plasmons are not excited (as explained above), the dispersion relation follows the light-line. At higher frequencies, as surface plasmons are excited, the dispersion deviates from the light-line, and the propagation speed slows down. As the frequency approaches $\omega_{sp}$, the propagation speed theoretically tends to zero. In reality, losses in metal only allow the dispersion to approach $\omega_{sp}$ asymptotically up to a finite wavenumber and bend the dispersion back to the light-line. The maximum reduction of speed from the speed of light is a factor of $\sim 10$ [1]. This factor of 10 has been attractive in the field of photonics. It implies that at the same frequency, the propagation wavelength of the
surface plasmon is ~10 times smaller than that of the EM wave. One can then make subwavelength structures to interact with light.

Figure 1.1 (a) Illustrations of charge density distributions in 3D plasmonic oscillation in a bulk metal, surface plasmonic wave on the surface of a 3D metal and 2D plasmonic waves in a 2D conductor. (b) Long-wavelength dispersion relations of the latter two cases compared to light-line (the free-space dispersion relation of light). The 3D plasmonic oscillation occurs at a constant optical frequency outside the wavenumber regime this thesis concerns, and is thus not drawn here.
1.2 Plasmonic Waves in 2D

In materials where mobile charges are strictly confined to two dimensions (Fig. 1.1a), there is no frequency-dependent penetration depth. There are fewer charges and the Coulomb interaction range is shorter than in 3D. The charges therefore have a lot of kinetic energy and more readily experience collective oscillations. The plasmonic wave dispersion relation, $\omega \propto \sqrt{k_p}$ ($k_p$: plasmonic wavenumber), which will be derived in the following chapter, deviates from the light-line at all frequencies (Fig. 1.1b). This feature enables the development of subwavelength structures at frequencies other than in the optical regime.

Figure 1.2 gives the plasmonic dispersions of AlGaAs/GaAs 2DEG and chemical vapor deposition (CVD) graphene on SiO$_2$/Si substrate using typical material properties. Already at gigahertz (GHz) frequencies, the propagation speed can be as low as $\sim c/100$ [6]. This much greater speed reduction translates to subwavelength confinement to a greater degree than surface plasmons, motivating the engineering of 2D plasmonics as the theme of this thesis.
Figure 1.2 Plasmonic dispersion relations of (a) AlGaAs/GaAs 2DEG and (b) CVD graphene on SiO$_2$/Si substrate, demonstrating clear subwavelength confinement. In this thesis, 2D plasmonic devices are developed for 2DEG in GHz range and for graphene in THz range, as suggested by these graphs. Carrier concentration in 2D, $n \sim 10^{12}$ cm$^{-2}$, is used as a typical value in both materials.
Chapter 2

Transmission-Line Model for 2D Plasmonics

As mentioned in the previous chapter, the charge carrier inertia and the Coulomb restoring force are the fundamental cause of plasmonic waves. The energy in a plasmonic system is converted back and forth between a kinetic form and an electric form, if there is no loss. This is similar to an EM wave, whose propagating energy cycles between a magnetic form and an electric form. Just as an EM wave can be modeled by a transmission line, with distributed magnetic inductors in series and distributed capacitors in parallel, so too can a plasmonic wave be modeled by a transmission line but with distributed kinetic, instead of magnetic, inductors (Fig. 2.1, $dz$: an infinitesimal segment of the line) [5, 25, 26]. The kinetic inductors store kinetic energy associated with the collective motion of charge carriers, and the capacitors store electric potential energy associated with the Coulomb restoring force. This chapter will first establish a lossless plasmonic transmission-line model consisting of kinetic inductors and capacitors for a 2D plasmonic system, with the 2D conductor either ungated or gated. The model is also applied to the surface plasmonic counterpart, giving consistent comparisons as in the previous chapter.
The effects of losses due to mobile charge scatterings are then added into the circuit model as a resistive component.

![Figure 2.1](image)

**Figure 2.1** The lossless transmission-line model of the 2D plasmonic waves.

## 2.1 Kinetic Inductance for 2D Plasmonics

While magnetic inductance opposes the changes in magnetic flux, kinetic inductance opposes changes in the collective motion of carriers. The kinetic inductance per unit length of a strip of 2D plasmonic wave medium with width $W$ is given by [7, 25, 27]

$$L_k = \frac{m^*}{ne^2} W = \frac{4\pi\hbar^2}{g_s g_v E_F e^2} \frac{1}{W}$$

(2.1)

($n$: number of mobile charge carrier per unit area; $\hbar$: reduced Planck constant; $E_F$: Fermi level of the 2D plasmonic wave medium; $g_s, g_v$: spin/valley degeneracy). The first expression is only true for a plasmonic material with non-zero charge carrier effective mass $m^*$. The second expression is applicable to a general 2D conductor, including graphene with $m^* = 0$. 

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2.2 Ungated 2D Plasmonics

The capacitance can take different forms based on the system setup. The following considers the cases when the 2D plasmonic wave medium is either ungated or gated. Both situations are used in the devices developed in this thesis.

When the 2D plasmonic wave medium is embedded in a dielectric material, with no conductors nearby, the ground of the transmission line is considered to be infinitely far away from the 2D system. The capacitance per unit length in this geometry is $C = 2\kappa \varepsilon_0 W$ [26]. Thus, the dispersion of this transmission line is simply

$$\omega_{\text{ungated}}(k_p) = \frac{k_p}{\sqrt{L_k C}} = \sqrt{\frac{ne^2 k_p}{2\kappa \varepsilon_0 m^*}} = \sqrt{\frac{E_k e^2 k_p}{2\pi \kappa \varepsilon_0 \hbar^2}}.$$ (2.2)

The relation between frequency and the square-root of wavenumber is typical for 2D plasmonic systems. We can also obtain the transmission-line characteristic impedance from

$$Z = \sqrt{\frac{L_k}{C}},$$ (2.3)

as well as the plasmonic wave group velocity (“p” stands for “plasmonic”),

$$v_p = \frac{d\omega}{dk_p} = \frac{1}{\sqrt{L_k C}}.$$ (2.4)

The above discussion does not consider the effect of electron degeneracy pressure, which also generates a restoring mechanism that acts against perturbations of the equilibrium charge carrier distribution. It is caused by Pauli’s exclusion principle when additional charges are placed
above the Fermi level. This quantum effect can also be modeled as a capacitance per unit length 
\[ C_q = e^2 g(E_F) W g(E_F) \] (density of states at the Fermi energy) at 0 K and can be included in the 
transmission line as a distributed quantum capacitor in series with the geometrical capacitor [28]. 
However, \( C_q \) is negligible compared to the ungated geometrical capacitance produced by the 
Coulomb restoring force in the long-wavelength regime \( (k_p << k_F, \text{ where } k_F \text{ is the Fermi} \) 
momentum) [27], which this thesis focuses on.

2.3 Gated 2D Plasmonics

In the case of a metal gate placed in proximity to a 2D conductor, the gate acts as the ground in the transmission-line picture (Fig. 2.1). If the gate is parallel to the 2D conductor at a separation \( d \) much smaller than the plasmonic wavelength \( (k_p d << 1) \), the geometrical capacitor is modified to have a parallel plate capacitance per unit length \( C = \kappa \varepsilon_0 \frac{W}{d} \). The dispersion relation becomes linear, as

\[
\omega_{\text{gated}}(k_p) = \frac{k_p}{\sqrt{L_k C}} = k_p \sqrt{\frac{n e^2 d}{\kappa \varepsilon_0 m^*}} = k_p \sqrt{\frac{E_F e^2 d}{\pi \kappa \varepsilon_0 \hbar^2}}. \tag{2.5}
\]

The dispersions of the ungated and gated cases are plotted together in Fig. 2.2. The much lower slope of the gated case with the same wavenumber shows that the gated 2D plasmonic wave propagates even slower. Physically, these are caused by the gate screening the Coulomb restoring force and reducing the Coulomb interaction range. The collective kinetic interaction between the charge carriers is therefore more pronounced in the gated case.
Figure 2.2 Plasmonic dispersion relations of AlGaAs/GaAs 2DEG when 2DEG has or not a gate in its proximity. \( n \sim 10^{12} \text{ cm}^{-2} \) and \( d = 80 \text{ nm} \) are used.

In the gated case, since the Coulomb restoring force is screened, the capacitance in the transmission line may no longer be dominated by the geometrical capacitance, and the quantum capacitance may become important. This happens when \( d \) is comparable to the Fermi wavelength \((k_Fd \leq 1)\). For a typical value of \( n \sim 10^{12} \text{ cm}^{-2} \) in 2DEG or graphene, this condition requires \( d < 5 \text{ nm} \). Since the gated circuit structures developed in this thesis used 2DEG with \( d = 80 \text{ nm} \), the quantum effect can still be safely ignored.
2.4 Comparison to Surface Plasmonics

With the transmission-line picture established, let us revisit the surface plasmonic-wave case and compare it to the 2D case. As the plasmonic oscillation is a result of kinetic inertia and Coulomb restoring force regardless of dimensionality, the surface plasmonic wave system can also be described by the kinetic inductance and capacitance [26]. Figure 1.1b already shows that the propagating wave in a conductor-insulator interface changes from EM to plasmonic as frequency increases. To describe this transition, the energetic component $\frac{1}{2}LI^2$ must have inductance $L$ dominated by magnetic inductance at low frequency and by kinetic inductance at high frequency ($I$: current). In a transmission-line model, both inductances can be included in series (Fig. 2.3). The magnetic inductance in an ungated transmission line can be modeled as a self-inductance per unit length $L_m = \mu_0/(2kW)$ ($\mu_0$: vacuum permittivity). (The light-line dispersion then has the correct form: $\omega = k/\sqrt{L_mC} = k/\sqrt{\kappa e_0\mu_0}$ with $C = 2\kappa e_0 kW$ as in Section 2.2 but with a general wavenumber $k$.) The surface plasmonic kinetic inductance per unit length is [26, 27]

$$L_{k,\text{surface}} = \frac{m^*}{n_{3D}e^2 W \delta} = \frac{3\pi^2 \hbar^2}{g_s g_v E_v e^2 W k_{F,3D} \delta^3}$$

(2.6)

where $k_{F,3D} = \frac{6\pi^2 n_{3D}}{g_s g_v}$ is the electronic wavenumber at the Fermi surface.

The penetration depth $\delta$ decreases with frequency [26], so the number of mobile charges $n_{3D}W \delta$ contained in the cross-section decreases. These fewer mobile charges need to accelerate to higher maximum velocity to maintain the same amount of current $I$. Their kinetic energy
therefore increases. Fig. 2.4 indicates that $L_{k,\text{surface}}$ increases and $L_m$ decreases as frequency increases. When $L_{k,\text{surface}}$ grows large enough to dominate over the magnetic inductance $L_m$, plasmonic waves are excited, and the dispersion curve deviates from the light-line, as seen in Chapter 1. This occurs at optical frequencies in typical surface plasmonic materials.

![Figure 2.3](image) The lossless transmission-line model of the surface plasmonic waves.

In contrast, as a 2D conductor has no penetration depth, its kinetic inductance has no frequency dependence (Eq. 2.1). Using typical AlGaAs/GaAs 2DEG parameters, its kinetic inductance per unit length is $L_k = (3.3916 \times 10^{-9}/W)$ H/m, which is orders of magnitude higher than the magnetic inductance at all frequencies in the long-wavelength regime (Fig. 2.4a).

Figure 2.4b, plotting of the surface $L_{k,\text{surface}}$, also illustrates clearly that $L_{k,\text{surface}} \ll L_k$, using typical parameters of gold [29] in Eq. 2.6. In addition, comparing Eq. 2.1 to 2.6, the kinetic inductance of the surface plasmon has an extra factor of $k_{F,3D}\delta$. Although as frequency approaches $\omega_p$, $\delta$ goes to zero, and $L_{k,\text{surface}}$ theoretically grows infinitely large, the loss in real metals prevents this from happening.
In summary, in the frequency range of interest, the value of $L_k$ in 2D is much greater than $L_{k,\text{surface}}$ and $L_m$ in 3D bulk materials so that the slowest plasmonic waves occur in 2D.

![Graphs showing frequency-dependent values of $L_m$ and $L_{k,\text{surface}}$](image)

**Figure 2.4** Values of the frequency-dependent (a) $L_m$ and (b) $L_{k,\text{surface}}$ (calculated using gold) in the frequency range of interest. Both are orders of magnitude smaller than the 2D $L_k = (3.3916 \times 10^{-9}/W) \text{ H/m}$.

### 2.5 Loss Consideration

We have so far ignored the effect of loss in the transmission-line model. In real materials, there are impurities, defects, phonons and other scattering centers of mobile charges that dissipate energy from the plasmonic wave. To take these damping mechanisms into account, distributed resistors are added in series with the inductors (Fig. 2.5). The value of the resistance per unit length is determined by the carrier momentum relaxation time $\tau$ via

$$R = \frac{L_k}{\tau}. \quad (2.7)$$
The total distributed series impedance per unit length is then

\[ R + i\omega L_k = \frac{1}{\sigma(\omega) W}, \tag{2.8} \]

where \( \sigma(\omega) \) is the mobile charge conductivity. The value of \( \tau \) includes all kinds of damping mechanisms. Factors such as acoustic and optical phonons increase the number of charge carrier scattering events and reduce \( \tau \) as temperature increases. When designing plasmonic structures, the effects of temperature must be taken into account. In order to observe plasmonic waves, the number of scatterings needs to be reduced, or the experiments need to be conducted at higher frequencies than the scattering frequency.

Figure 2.5 The transmission-line model of the 2D plasmonic waves with losses taken into account by connecting distributed resistors with kinetic inductors.

Quantitatively, this design criterion is described by the new characteristic impedance of the transmission line

\[ Z = \sqrt{(R + i\omega L_k) / (i\omega C)} = \sqrt{L_k / C} \sqrt{1 - i/Q}, \]

where \( Q \) is the quality factor of the plasmonic oscillation and represents the number of plasmonic oscillation cycles between scattering events. The extra factor \( \sqrt{1 - i/Q} \) indicates that the imaginary part of \( Z \) reduces to zero as \( Q \) grows infinitely large. \( Z \) then approaches the characteristic impedance of a
transmission line with no resistance (Eq. 2.3). In fact, \( Q = \omega L_k/R = \omega \tau \). In order for plasmonic waves not to be overwhelmed by scattering, \( L_k/R = \tau \) or \( \omega \) has to be large.

In this thesis, the AlGaAs/GaAs sample has values of \( \tau \sim 1.4 \times 10^{-13} \) s at room temperature and \( \sim 1.5 \times 10^{-10} \) s at 4.2 K. These values imply that in order to achieve \( Q = \omega \tau > 1 \), the frequencies need to be >1 THz at room temperature and >1 GHz at 4.2 K. Therefore, for proof-of-concept GHz demonstrations, we use cryogenic temperatures for the AlGaAs/GaAs sample and study the device behaviors as the temperature rises (Chapter 3). The CVD graphene sample used has \( \tau \sim 5 \times 10^{-14} \) s at room temperature, and is used for experiments at frequencies >3 THz (Chapters 4 and 5).
Chapter 3

Ultra-Subwavelength 2D Plasmonic Interferometer

The methodology behind engineering 2D plasmonic circuits is based on designing and fabricating the shapes of the 2D conductors. The shaped 2D conductors act as channels which restrict the plasmonic wave to within their boundaries and guide the plasmonic waves across the conducting plane. The plasmonic waves can reflect, refract, diffract or interfere as experienced by any classical waves. By shaping the channel geometry to promote these wave interactions, we can engineer useful functionalities.

This chapter focuses on an interferometer,\(^\text{ii}\) in which plasmonic waves propagate along two paths of different lengths and interfere to introduce a phase difference. Due to 2D plasmonic ultra-subwavelength confinement, the resolution derived from the phase difference is two orders

\(^{\text{ii}}\) Large portions of this chapter are derived from a paper in publication by the author [30].
of magnitude higher than that of its electromagnetic counterpart at a given frequency. We explain the procedures to design, fabricate, measure and analyze the performance and applications of the plasmonic interferometer.

3.1 Design of the 2D Plasmonic Interferometer

As a proof of concept, we use AlGaAs/GaAs 2DEG as a 2D plasmonic medium and construct a Mach-Zehnder interferometer, whose two paths are defined by mesa-etching the 2DEG (Fig. 3.1). The curved 2DEG path has a length of \( l_1 \sim 191 \mu m \) and the linear path has a length of \( l_2 \sim 120 \mu m \) while both have the same width of \( W \sim 8 \mu m \). The left metallic coplanar waveguide (CPW) consisting of a signal line (‘S’) and two ground lines (‘G’) guides an electromagnetic wave onto the left end of the interferometer; the CPW’s signal line makes an ohmic contact to the 2DEG. The excited plasmonic wave splits into two plasmonic waves along the two 2DEG paths, which superpose at the right end of the interferometer. By this junction, the two waves develop a frequency-dependent phase difference due to the path length difference, thus, their superposition exhibits an interference pattern with frequency. This superposed wave excites an electromagnetic wave onto the right CPW, which we measure to study the interference. The characteristic impedance, \( Z_0 \), of both CPWs is designed to be 50 \( \Omega \).

A metallic top gate placed 80 nm above both 2DEG paths serves as an ac plasmonic ground; each 2DEG signal path with the gate as the ac ground then forms a plasmonic transmission line. The plasmonic ground is merged with the CPWs’ ground lines. This ground sharing ensures seamless continuation of the purely plasmonic transmission lines from the purely electromagnetic CPWs \([6, 31]\); with no such ground sharing, the gate itself would act as an
electromagnetic path, instead of a plasmonic ground. As explained in Chapter 2, the gate also serves to further slow the 2D plasmonic velocity (which is already small due to the reduced dimensionality) by shortening the Coulomb interaction range within 2DEG [6, 32]. This further enhances subwavelength confinement and interferometer resolution. The gated 2D plasmonic velocity, \( v_p = \frac{\sqrt{(ne^2d)} / (m^* \kappa e_0)}{c/300} \), is \( \sim c/300 \) in our case as seen shortly in Section 3.2 (parameters defined in Chapters 1 and 2). Yet another role of the gate is to provide a \( dc \) bias to tune the electron density, hence \( v_p \). In actuality, we apply to the CPWs’ signal lines a \( dc \) bias with reference to the ground lines, so that the gate is effectively biased in the opposite polarity relative to the 2DEG.

Figure 3.1 Diagram of the 2D plasmonic Mach-Zehnder interferometer (not drawn to scale). The inset shows an optical microscope image of an actual implementation. 2DEG forms between AlGaAs and GaAs layers.
We use the Sonnet frequency-domain electromagnetic field solver to design the 50-Ω CPWs [6, 27, Appendix B]. The signal lines have a width of 24 μm and a length of 218 μm and are separated by 15 μm from the 123 μm-wide ground lines on each side. Each ohmic contact occupies an area of 6×24 μm². The separations between the top gate and the CPW signal lines on the left and right sides of the interferometer are 7 μm (Fig. 3.1). These ungated 2DEG paths are far shorter than the gated 2DEG paths. This is to minimize the nonlinear dispersion effect of the ungated 2DEG regions (Chapter 2).

3.1.1 Materials and Fabrication

Before device fabrication, the GaAs/AlGaAs 2DEG at 4.2 K has a carrier density, \( n \), of \( 1.54 \times 10^{11} \) cm\(^{-2} \) and a mobility, \( \mu \), of \( 2.5 \times 10^6 \) cm\(^2\)/Vs in the dark. Under illumination, \( n = 2.8 \times 10^{11} \) cm\(^{-2} \) and \( \mu = 3.9 \times 10^6 \) cm\(^2\)/Vs. At room temperature, \( n = 3.76 \times 10^{11} \) cm\(^{-2} \) and \( \mu = 3.66 \times 10^3 \) cm\(^2\)/Vs in the dark. We etch the AlGaAs/GaAs sample by > 80 nm from the top surface to define the two 2DEG mesa paths of the interferometer (above the 2DEG, there is a 48 nm undoped Al\(_{0.3}\)Ga\(_{0.7}\)As layer, a 26 nm Si-doped Al\(_{0.3}\)Ga\(_{0.7}\)As layer, and a 6 nm GaAs cap). The ohmic contacts are created by thermally evaporating layers of Ni (5 nm), Au (20 nm), Ge (25 nm), Au (10 nm), Ni (5 nm), and Au (40 nm), followed by annealing at 420 °C for 50 seconds. The CPWs and the top gate are deposited by thermal evaporation of Cr (8 nm) and Au (500 nm).

3.2 Experiments on the 2D Plasmonic Interferometer

We perform microwave scattering experiments in the dark up to 50 GHz with a vector network analyzer to obtain frequency-domain interference patterns. The measurements are done inside a LakeShore cryogenic probe station. Temperature is at 4.2 K so that plasmonic dynamics
is not masked by electron scatterings. The Agilent E8364A vector network analyzer generates an ac signal up to 50 GHz with -45 dBm power reaching the device via ground-signal-ground microwave probes (100-μm pitch) and coaxial cables. The network analyzer, cables, and probes all have a characteristic impedance of 50 Ω. The network analyzer measures s-parameters. The delay and loss from the network analyzer up to the probe tips are calibrated out by using the NIST-style multi-line TRL calibration method [33]; this procedure involves measuring a set of s-parameters for CPWs of varying lengths fabricated on a separate GaAs substrate with no 2DEG present [6, 7]. This calibration leads to the raw s-parameters, which include the effects of the interferometer, the phase delays through the on-chip CPWs, and the direct parasitic coupling between the two on-chip CPWs, which bypass the interferometer. The phase delays of the electromagnetic waves traveling through the CPWs are far smaller than the phase delays of the much slower plasmonic waves traveling through the interferometer, so we safely ignore the CPW phase delays. The effect of the parasitic coupling is separately measured by applying a gate bias of -0.4 V, thus depleting the 2DEG to imitate an open circuit (Fig. 3.2). These open-circuit-device s-parameters are then de-embedded from the interferometer’s raw s-parameters [34, Appendix C]. Figure 3.2 juxtaposes the raw and the de-embedded s-parameters at an example bias at 0.55 V. The s-parameters discussed from here on are de-embedded ones, unless otherwise stated.
Figure 3.2 (a) Magnitudes of raw $s_{21}$ (red, dashed), de-embedded $s_{21}$ (black, solid), and open-device $s_{21}$ (blue, semi-dashed) at 4.2 K with a gate bias of 0.55 V. (b) Values of the phases of the three $s_{21}$ parameters. In (b), y-axis is frequency and x-axis is $s_{21}$ phase, following the convention of showing dispersion.

3.2.1 Derivation of $S_{21}$ Parameter of the Plasmonic Interferometer

Theoretically, the transmission $s_{21}$ of the interferometer can be approximated. Figure 3.3a illustrates the interferometer consisting of two plasmonic transmission lines ‘1’ and ‘2’ along with the two on-chip CPWs ‘A’ and ‘B’. When an electromagnetic wave is launched onto CPW A, multiple transmissions and reflections will occur at the two junctions in the figure, and multiple waves will appear at CPW B through many different signal pathways. Superposition of these multiple waves represents the total transmitted wave. There are two 1st-order signal pathways from CPW A to CPW B that exhibit the lowest degree of loss: A→1→B and A→2→B. There are eight 2nd-order signal pathways from CPW A to CPW B that exhibit the second lowest degree of loss (in what follows, a path number with no prime signifies left-to-right propagation
on that path, and a path number with prime signifies right-to-left propagation): A→1→1’→1→B, A→1→1’→2→B, A→1→2’→1→B, A→1→2’→2→B, A→2→1’→1→B, A→2→1’→2→B, A→2→2’→1→B, and A→2→2’→2→B. Similarly we can identify higher-order signal pathways.

Figure 3.3 (a) Schematic showing the interferometer’s two plasmonic paths (1 and 2) along with the two on-chip CPWs (A and B). (b) Illustration of local transmissions and reflections for the incoming power wave from CPW A.

We first calculate the contribution of the 1st-order signal pathways (A→1→B and A→2→B) to $s_{21}$. Since s-parameters are defined in terms of power waves [35], we start by calculating local transmission coefficients for A→1, 1→B, A→2, and 2→B for power waves. Figure 3.3b shows the left junction. The incoming power wave $a_d^+$ on CPW A produces, at the junction, transmitted power wave $a_1^+$ and $a_2^+$ on path 1 and 2 (as well as the reflected power wave $a_d^-$, which is not involved in the $s_{21}$ calculation with the 1st-order signal pathways), where [35]:

$$a_d^+ = \frac{V_1^+ + Z_0(I_1^+ + I_2^+)}{2\sqrt{Z_0}}; \quad a_1^+ = \frac{V_1^+ + ZI_1^+}{2\sqrt{\text{Re}\{Z\}}}; \quad a_2^+ = \frac{V_2^+ + ZI_2^+}{2\sqrt{\text{Re}\{Z\}}}.$$  \hspace{1cm} (3.1)
Here $V_1^+$ and $V_2^+ (= V_1^+$ due to the shunt connections of path 1 and path 2 at the junction) are the voltages of the plasmonic waves transmitted onto path 1 and path 2, read at the junction; $I_1^+$ and $I_2^+ (= I_1^+$ because path 1 and path 2 have the same characteristic impedance, $Z$) are the currents of the plasmonic waves transmitted onto path 1 and path 2, read at the junction. Then the local transmission coefficients [35] $t_{A\rightarrow 1} = a_1^+ / a_A$ and $t_{A\rightarrow 2} = a_2^+ / a_A$ are given by

$$t_{A\rightarrow 1} = t_{A\rightarrow 2} = \sqrt{\frac{Z_0}{\text{Re}\{Z\}}} \frac{2Z}{Z + 2Z_0}.$$  

(3.2)

Similarly, we can calculate the local transmission coefficients $t_{1\rightarrow B}$ and $t_{2\rightarrow B}$ as:

$$t_{1\rightarrow B} = t_{2\rightarrow B} = \sqrt{\frac{\text{Re}\{Z\}}{Z_0}} \frac{2Z_0}{Z + 2Z_0}.$$  

(3.3)

Then the overall A-to-B transmission, $s_{21}$, through the 1st-order signal pathways is given by $t_{A\rightarrow 1} t_{1\rightarrow B} e^{-\alpha l_1} e^{-ij\beta_1} + t_{A\rightarrow 2} t_{2\rightarrow B} e^{-\alpha l_2} e^{-ij\beta_2}$, that is:

$$s_{21} = \frac{4ZZ_0}{(Z + 2Z_0)^2} \left[ e^{-\alpha l_1} e^{-ij\beta_1} + e^{-\alpha l_2} e^{-ij\beta_2} \right]$$

[1st order]  

(3.4)

($\beta \equiv \alpha \nu_p$ and $\alpha$ are the wavenumber and attenuation constant of the plasmonic lines, respectively.

The latter due to electron scatterings in 2DEG).

We now consider the contribution of the 2nd-order signal pathways to $s_{21}$. Given that $\alpha \sim 8000$ m$^{-1}$ (Subsection 3.2.2), traversing path 1 ($l_1 \sim 191$ µm) once will reduce the amplitude by a factor of $\exp(-\alpha l_1) \sim 0.22$ and traversing path 2 ($l_2 \sim 120$ µm) once will reduce the amplitude by a factor of $\exp(-\alpha l_2) \sim 0.38$. Since each of the eight 2nd-order signal pathways involves traversing path 1 and/or path 2 a total of three times, each pathway will suffer significant attenuation. In
addition, each 2\textsuperscript{nd}-order pathway involves two additional local reflections and/or transmissions, which further attenuates the signal. The eight substantially attenuated signals superpose in CPW B, but they have generally different phases, thus, their superposition does not help much in countering the attenuation. All in all, the contribution from the 2\textsuperscript{nd}-order pathways to \( s_{21} \) is negligibly small, which is confirmed by the actual calculation of the 2\textsuperscript{nd}-order contribution (Fig. 3.4). In sum, it is sufficient to calculate \( s_{21} \) up to the 1\textsuperscript{st} order and for simplicity neglect the ohmic contacts and the short ungated 2DEG regions (Fig. 3.1):

\[
\begin{align*}
\begin{equation}
\begin{aligned}
s_{21} &\approx \frac{4Z_0Z}{(Z+2Z_0)^2} \left[ e^{-\alpha l_1} e^{-i\beta l_1} + e^{-\alpha l_2} e^{-i\beta l_2} \right] = \frac{4Z_0Z}{(Z+2Z_0)^2} A_1 e^{-i\beta l_1} \left[ 1 + \frac{A_2}{A_1} e^{-i\Delta \phi} \right] \\
( A_1 &\equiv e^{-\alpha l_1} \text{ and } A_2 \equiv e^{-\alpha l_2} \text{; and } \Delta \phi \equiv \beta(l_1-l_2) = \omega(l_1-l_2)/v_p \text{ is the phase difference between the two plasmonic waves at the right junction of the interferometer}).
\end{aligned}
\end{equation}
\end{align*}
\]

Figure 3.4 Contributions of the 1\textsuperscript{st} (black, solid) and 2\textsuperscript{nd} (blue, dashed) order signal pathways to the \( s_{21} \) magnitude and their sum (red, semi-dashed). Physical parameters used (\( L, R, \alpha \) and \( \beta \)) for this plot are those (median values) extracted at 0.55 V (Subsection 3.2.3).
The $s_{21}$ is proportional to the superposition of the two plasmonic waves and will exhibit an interference pattern with frequency. The factor $4Z_0Z/(Z+2Z_0)^2 \equiv F$ is complex in general, as $Z$ is generally complex due to loss caused by electron scatterings; however, $F$ exhibits a practically constant magnitude and a negligible phase over the measurement frequency range as will be seen in Subsection 3.2.3.

### 3.2.2 Performance of the Plasmonic Interferometer

Figure 3.5 shows the magnitude of the measured $s_{21}$ vs. frequency at various effective gate biases. At each bias the $s_{21}$ magnitude shows the anticipated interference pattern with destructive dips marked as 1, 3 and 5, and constructive peaks as 2, 4 and 6. The reduction of the peak and dip magnitudes with frequency, which is not predicted by Eq. 3.5, stems from the frequency dependent behaviors of the ohmic contacts and the ungated 2DEG regions. According to Eq. 3.5, the destructive dips (constructive peaks) with amplitudes proportional to $1 - (+) A_2/A_1$ occur at frequencies that make $\Delta \phi \equiv \omega(l_1-l_2)/v_p$ odd (even) integer multiples of $\pi$. The increasing gate bias increases $v_p$, leading to the shift of the interference pattern to the right (Fig. 3.5a); for a larger $v_p$, higher frequencies are required to produce the same $\Delta \phi$. From these considerations in connection with Eq. 3.5, we can extract, at a given bias, $v_p$ at six different frequencies corresponding to the three dips and three peaks of the $s_{21}$ magnitude. Figure 3.6 shows the range (red bar) and median (red square) of such six extracted $v_p$ values at each bias, where the median is the average of the two middle $v_p$ values. While $v_p$ must be constant at a given bias, the extracted $v_p$ shows the frequency variation (bar). This is due to the frequency-dependent behaviors of the ohmic contacts and ungated 2DEG regions, which can shift the dip and peak positions from the ideal positions predicted by Eq. 3.5. While the actual $v_p$ should monotonically increase with gate bias,
the extracted $v_p$ shows deviation from this trend at biases below 0.4 V (Fig. 3.6). We suspect that this abnormality is also caused by the ohmic contacts, this time through their substantial bias dependence below 0.4 V, which considerably affects the magnitude of $s_{21}$.

Figure 3.5 (a) $s_{21}$ magnitude and (b) absolute value of $s_{21}$ phase at effective gate bias 0.45 V (red, semi-dashed), 0.55 V (green, dotted), 0.65 V (blue, dashed), and 0.75 V (black, solid). Temperature: 4.2 K.

We alternatively extract $v_p$ from the phase, $\theta$, of the measured $s_{21}$, whose absolute value vs. frequency is shown in Fig. 3.5b at various biases. Equation 3.5 yields the following expression for $\theta$ (as will be shown in Subsection 3.2.3, $4Z_0Z/(Z+2Z_0)^2$ of Eq. 3.5 has a negligible phase):

$$
\theta = -\arctan \left[ \frac{\sin(l_1\omega/v_p) + (A_2/A_1)\sin(l_2\omega/v_p)}{\cos(l_1\omega/v_p) + (A_2/A_1)\cos(l_2\omega/v_p)} \right],
$$

which reduces to $\theta = \omega(l_1+l_2)/(2v_p)$ for $A_1 = A_2$, as expected. Equation 3.6 predicts that with a larger bias, thus with a larger $v_p$, $\theta$ increases more slowly with frequency, as evident in Fig. 3.5b.

We extract $v_p$ by fitting Eq. 3.6 to Fig. 3.5b at each gate bias. The $A_2/A_1$ ratios required for this fitting are extracted from the amplitudes of the dips and peaks of the $s_{21}$ magnitude in connection
with Eq. 3.5; specifically, at a given bias, a total of five \( A_2/A_1 \) ratios are obtained over five frequency ranges, with each range spanning from a dip to its adjacent peak; while the actual \( A_2/A_1 \) ratio must be constant at a given bias, the variations of the extracted \( A_2/A_1 \) values with frequencies at a given bias are caused by the frequency-dependent effects of the ohmic contacts and ungated 2DEG regions. Subsequently, fitting of Eq. 3.6 to Fig.3.5b is performed in each of these five frequency ranges, yielding five values of \( v_p \) at each bias. The ranges (blue bars) and medians (blue dots) of \( v_p \) so extracted from the measured \( s_{21} \) phase are shown in Fig. 3.6. As compared to \( v_p \) extracted from the \( s_{21} \) magnitude, this \( s_{21} \)-phase based extraction shows a greater immunity to the ohmic contact and ungated 2DEG effects: first, the frequency-dependent variation (sizes of the bars) is smaller; second, \( v_p \) shows the expected monotonic increase with an increasing bias even below 0.4 V. At biases in excess of 0.4 V, the median values of \( v_p \) extracted from the \( s_{21} \) magnitude and those extracted from the \( s_{21} \) phase show a match within 5%. The high-fidelity \( v_p \) extracted from \( s_{21} \) phase ranges from \( \sim c/330 \) (0.2 V) to \( \sim c/250 \) (0.85 V), attesting to ultra-subwavelength confinement of the gated 2D plasmons.

In addition, since \( A_2/A_1 = e^{\alpha_l(l_2-l_1)} \), we can then extract the attenuation constant, \( \alpha \) (Fig. 3.7). The median values of \( \alpha \) are around 8000 m\(^{-1}\) at gate biases in excess of 0.4 V, where ohmic contact effects are less pronounced in the \( s_{21} \) magnitudes.
**Figure 3.6** The plasmonic velocities extracted from the measured $s_{21}$ magnitude (red squares and bars) and those extracted from the measured $s_{21}$ phase (blue dots and bars). The squares/dots and bars, respectively, represent the median values and frequency-dependent variations at given biases.

**Figure 3.7** (a) $A_2/A_1$ extracted from the $s_{21}$ magnitude. (b) $\alpha$ extracted from $A_2/A_1$ of part (a).
3.2.3 Extraction of Circuit Parameters

Both gated 2D plasmonic transmission lines of the interferometer have an identical 2DEG width, \( W \), thus the same characteristic impedance, \( Z \). The gated 2D plasmonic line can be modeled as a distributed ladder network of kinetic inductors and capacitors (Chapter 2); \( L_k = m^*/ne^2W \) is the kinetic inductance per unit length, \( C = \kappa\varepsilon_0 W/d \) is the capacitance per unit length. The ohmic resistance per unit length, \( R = 1/(ne\mu W) \), in series with \( L_k \) stems from the electron scatterings. To evaluate \( Q = \omega L_k/R > \omega \tau \) is the quality factor, we should first know the values for \( L_k \) and \( R \) (on the other hand, \( C = \kappa\varepsilon_0 W/d \sim 1.14 \times 10^{-8} \text{ F/m} \) can be readily evaluated using the known geometric parameters). By using \( v_p \) values extracted from the \( s_{21} \) phase (Fig. 3.6), in \( v_p = 1/\sqrt{L_kC} \) we extract \( L_k \) values (Fig. 3.8a). By noting that \( Q = \omega L_k/R \) on the one hand and \( Q \sim \beta/2\alpha = \omega/(2v_p\alpha) \) on the other hand [36], we can write \( R = 2\alpha v_p L_k \); then by using the extracted values of \( \alpha \) (Fig. 3.7b), \( v_p \) (Fig. 3.6), and \( L_k \) (Fig. 3.8a) in this formula, we can extract \( R \) (Fig. 3.8b). With the extracted \( R \) and \( L_k \) values, we can evaluate \( Z = \sqrt{(R+i\omega L_k)/(i\omega C)} = \sqrt{L_k/C/\sqrt{1-i/\omega}} \). As \( Q = \omega L_k/R \sim 1 \) occurs below 5 GHz, the imaginary part of \( Z \) becomes increasingly small as frequency rises. Moreover, by substituting this \( Z \) into \( F \equiv 4Z_0Z/(Z+2Z_0)^2 \) of Eq. 3.5, we find that \( F \) itself has a negligible imaginary part as compared to its almost constant real part over nearly all measurement frequency range (Fig. 3.9a). That is, \( F \) has a negligible phase (Fig. 3.9b) and a constant magnitude. This is self-consistent with our \( v_p \) extraction from \( s_{21} \) phase, where we ignored the phase of \( F \) (subsections 3.2.1 and 3.2.2).

The extracted \( L_k \) and \( R \) values also allow us to extract the electron density, \( n_e \), and the mobility, \( \mu \) (Fig. 3.8c) and Fig. 3.8d). The median values of the extracted \( \mu \) lie between \( 1 \times 10^6 \)

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\[ ~1.5 \times 10^6 \text{ cm}^2/\text{Vs} \], which are comparable to, but justifiably smaller due to fabrication steps than the mobility of the pristine sample (\(2.5 \times 10^6 \text{ cm}^2/\text{Vs}\)).

Figure 3.8 (a) Extracted \(L_k\). (b) Extracted \(R\). (c) Extracted \(n\). (d) Extracted \(\mu\).
3.3 Temperature Effects on the 2D Plasmonic Interferometer

We further confirm the plasmonic interferences by temperature-dependent measurements. As temperature rises, the electron scatterings are promoted to increase the ohmic resistance [37], degrading the 2DEG as a plasmonic medium. Quantitatively, while the plasmonic quality factor, \( Q=\omega \tau \), is greater than unity at 4.2 K at GHz frequencies, \( \tau \) and thus \( Q \) decreases with temperature, making the observation of plasmonic waves and their interferences increasingly difficult. This is attested by temperature-dependent measurements of Fig. 3.10, where we show, for a reason that will be clear shortly, raw \( s_{21} \)-parameters without de-embedding the direct coupling between the two CPWs. As temperature rises towards 40 K, the \( s_{21} \) magnitude gradually decreases, blurring the interference pattern (Fig. 3.10a). At room temperature, the ohmic resistance becomes so high that the interferometer becomes effectively open-circuited; the non-zero raw \( s_{21} \) magnitude at room temperature in Fig. 3.10a is due to the direct coupling between the CPWs. This is confirmed by the close similarity between this raw \( s_{21} \) magnitude at room
temperature and the $s_{21}$ magnitude of the actual open device at 4.2 K, where this open device is attained by biasing the gate at -0.4 V and depleting the 2DEG. It is for this comparison that Fig. 3.10 presents raw $s$-parameters. The raw $s_{21}$ phase in Fig. 3.10b tells the same physics: up to 40 K, $s_{21}$ phase does not change appreciably, as the plasmonic wave, while increasingly masked by electron scatterings, observably maintains the same velocity. At room temperature, the plasmonic dynamics is completely masked, and the interferometer behaves as an open circuit.

![Figure 3.10](image)

**Figure 3.10** (a) Magnitude and (b) phase of the interferometer’s raw $s_{21}$ parameters at 4.2 K (black, solid), 20 K (blue, dashed), 40 K (red, semi-dashed), and room temperature (green, dotted), and of the actual open circuits’ $s_{21}$ parameters at 4.2 K (magenta, thin).

### 3.4 Applications of the 2D Plasmonic Interferometer

Due to the ultra-subwavelength confinement of 2D plasmons, the interferometer achieves a high resolution. Since $\Delta \phi \equiv \omega (l_1-l_2)/v_p$ and $v_p \sim c/300$, a given path length difference, $l_1-l_2$, will lead to a $\sim 300/\sqrt{k_{EM}}$ times larger phase difference in the plasmonic interferometer than in an
electromagnetic wave interferometer whose wave speed is $c/\sqrt{\kappa_{\text{EM}}}$ ($\kappa_{\text{EM}}$ is the effective dielectric constant of the medium in the electromagnetic interferometer). So for the given $\Delta \phi$ resolution of the vector network analyzer, the plasmonic interferometer achieves a $\sim 300/\sqrt{\kappa_{\text{EM}}}$ times higher resolution than the electromagnetic interferometer at the same frequency. The plasmonic interferometer is also capable of a higher resolution detection of changes in the physical parameters on which $v_p$ depends, where this benefit is again attributed to the ultra-subwavelength confinement. For example, a change in the refractive index $\sqrt{\kappa}$ of the medium separating the gate and the 2DEG incurs a change in $\Delta \phi$ as 

$$d(\Delta \phi)/d\sqrt{\kappa} = \left[\omega(l_1 - l_2)/c\right] \times \left[c/(v_p\sqrt{\kappa})\right],$$

while this derivative for an electromagnetic interferometer with a wave speed of $c/\sqrt{\kappa_{\text{EM}}}$ is 

$$d(\Delta \phi)/d\sqrt{\kappa_{\text{EM}}} = \omega(l_1 - l_2)/c.$$ 

Thus the plasmonic interferometer is $\sim 300/\sqrt{\kappa}$ times more sensitive to the refractive index change, achieving a higher resolution by the same factor. The plasmonic interferometer can also detect the changes in the 2DEG electron density, $n$, and the effective electron mass, $m^*$, with high resolution, as $v_p$ depends on these quantities. They are quantities irrelevant to electromagnetic interferometers, thus, their detection is a bonus feature of our interferometer.

### 3.4.1 Frequency Scaling

Despite the high-resolution advantage, the GHz proof-of-concept prototype presented is limited to cryogenic operation. Specifically, $Q/2\pi$ signifies the number of local oscillation cycles of electrons in a given plasmonic wave during a mean momentum relaxation time $\tau$. A larger $Q$ facilitates plasmonic wave observation; if $Q$ becomes smaller than unity, scattering occurs before electrons undergo a fraction of one oscillation cycle, thus, the plasmonic dynamics is
increasingly masked by the ohmic resistance (scattering). To ensure $Q = \omega \tau$ in excess of 1 at the GHz frequencies, we increased $\tau$ by operating the plasmonic interferometer at 4.2 K thus by suppressing electron scatterings by phonons. With a higher frequency, $\tau$ can be made shorter (i.e., operation temperature can be higher) while maintaining large enough $Q = \omega \tau$. At higher THz and infrared frequencies, 2D plasmons in AlGaAs/GaAs 2DEG and graphene can be observed at room temperature [8, 18, 39], while maintaining the nature of subwavelength confinement. Design into these higher frequencies can be done by scaling down the device dimensions. For instance, to attain the first destructive dip [$\Delta \phi = \omega (l_1 - l_2)/v_p = \pi$] at 3 THz with $v_p \sim c/300$, the path length difference $l_1 - l_2$ can be set to 170 nm. Scaling our design into this higher frequency regime may enable high-resolution THz and infrared interferometric applications, such as biochemical detection, molecular spectroscopy, and precision modulation, at room temperature.
Chapter 4

Graphene Plasmonics

As we consider THz plasmonic materials for room temperature devices, graphene stands out as a natural 2D material with subwavelength confinement and tunability [19]. Again, our methodology of engineering graphene plasmonic wave dynamics relies on patterning the shape of graphene. While previous graphene plasmonic work [8, 13-18] has focused on isolated graphene ‘islands’ that support excitation of localized plasmons, here we generate delocalized plasmons in a continuous graphene medium with a periodic structural perturbation, so as to create plasmonic bands in a manner akin to photonic crystals. The fabricated periodic structures are thus called graphene plasmonic crystals. The plasmonic band formation and excitation are demonstrated with both simulation and Fourier transform infrared spectroscopy. This chapter presents the first demonstration of graphene plasmonic crystals and explains their design, fabrication, simulation and measurement procedures. Group theory is used to interpret the
selection of a specific set of plasmonic bands excited by phase matching and symmetry-based coupling between plasmons and photons.iii

It has been noted that this work differentiate itself from another report [40] which showed mid-infrared plasmon excitation in a similar aperture lattice. While sharing similar physics, [40] focuses on coupling graphene plasmons with mid-infrared substrate phonons. In contrast, we purposefully avoid such coupling by working in the far-infrared regime to focus on the medium-periodicity-based plasmon band engineering and symmetry-based plasmon band selection rule.

4.1 First Demonstration of Graphene Plasmonic Crystals with Hexagonal Lattices

One can imagine a variety of ways to introduce structural periodicity in a continuous graphene medium. The hexagonal lattice of apertures in our work is one proof-of-concept realization of medium periodicity. We fabricate [Appendix A] four graphene plasmonic crystals, which we call GPC1 to GPC4 (Fig. 4.1), by etching out hexagonal lattices of circular (GPC1) or hexagonal shape apertures (GPC2 to GPC4) via photolithography in four separate regions of the same 1.5×1.5 cm² graphene sheet. This graphene sheet, which is grown by CVD and is transferred onto a 289 nm SiO₂ / 381 μm Si substrate, exhibits the typical Raman spectrum (Fig. 4.2a) of monolayer graphene [41, 42]. The hexagonal lattice geometry of each plasmonic crystal, occupying an area of 2×2 mm², is characterized by the lattice constant \( a \), the aperture shape that is circular or hexagonal, and the aperture size (diameter \( D \) in case of circular apertures, edge-to-opposite-edge distance \( D' \) in case of hexagonal apertures). The geometric parameters are in the micrometer range; for instance, \( a \) ranges from 3 μm to 6 μm. An optical micrograph and a

iii Large portions of this chapter are derived from a paper in publication by the author [21, 56].
scanning electron microscopy (SEM) image of GPC1 \((a \sim 3 \mu m; D \sim 2 \mu m)\) are shown in Figs. 4.1b, c. A map of the integrated Raman 2D peak intensity [42, 43] from 2630 cm\(^{-1}\) to 2730 cm\(^{-1}\) (Fig. 4.2b) also confirms the hexagonal lattice in the graphene.

We also leave a certain region unpatterned (area: \(\sim 1.0 \times 0.5 \text{ cm}^2\)) in the same graphene sheet, as its interaction with far-infrared light provides a comparison to the interaction of plasmonic crystals with far-infrared light. On this unpatterned graphene, we also perform a Hall transport measurement via the four-probe van der Pauw method, from which we determine the charge carrier type (holes), its concentration \(n (\sim 1.05 \times 10^{13} \text{ cm}^{-2})\), and mobility \(\mu (\sim 1360 \text{ cm}^2/\text{Vs})\). These values correspond to a Fermi level \(E_F = -\hbar v_F \sqrt{(n \pi)} \sim -0.38 \text{ eV}\) and carrier momentum relaxation time \(\tau = \hbar^2 \pi n \mu/(eE_F) \sim 5 \times 10^{-14} \text{ s}\) \((v_F: \text{Fermi velocity})\) [22]. The sign of \(E_F\) is chosen to be negative for holes, with the Dirac point set as the reference 0 eV. \(E_F\) and \(\tau\) are important characteristics that influence the detailed behavior of graphene plasmons. While \(E_F\) and \(\tau\) spatially vary in large-area graphene [44], and may also assume degraded values in the patterned graphene plasmonic crystals due to the edge disorder introduced at the boundaries of the apertures [45], their grossly measured values in the unpatterned region give a rough feel for their values in the crystal regions, and signify certain characteristics of the graphene sample. For instance, our graphene sample is strongly doped (with holes), and the plasmonic quality factor \(Q = \omega \tau\) is \(\sim 1.9\) at 6 THz.

The length scales of the hexagonal lattice parameters \(a, D,\) and \(D'\) are comparable to the graphene plasmonic wavelengths in the far infrared region, where graphene plasmons emerge conspicuously [19]. Hence, plasmons are scattered by the lattice, and their dispersion relation is transformed from the continuous dispersion curve of unpatterned graphene into a plasmonic band structure, as seen theoretically [46, 47].
Figure 4.1 (a) Illustration (not drawn to scale) of the graphene sample on SiO\textsubscript{2}/Si substrate, containing four graphene plasmonic crystals: GPC1 ($a \sim 3$ $\mu$m, $D \sim 2$ $\mu$m), GPC2 ($a \sim 4$ $\mu$m, $D' \sim 3$ $\mu$m), GPC3 ($a \sim 5$ $\mu$m, $D' \sim 4$ $\mu$m), GPC4 ($a \sim 6$ $\mu$m, $D' \sim 5$ $\mu$m), an unpatterned graphene region, and bare SiO\textsubscript{2}/Si region uncovered by graphene. $T$ [$T_0$] is the light intensity through a crystal [bare SiO\textsubscript{2}/Si] in FTIR. (b) Optical microscope image and (c) SEM image of GPC1.

Figure 4.2 (a) Typical Raman spectrum of the graphene sample in the unetched area. (b) Integrated graphene Raman 2D peak intensity map from 2630 to 2730 cm\textsuperscript{-1} of GPC1, with dark areas indicating apertures.
4.1.1 Band Structure Simulation

We first show the plasmonic band structure of the hexagonal lattice by simulation, by solving Maxwell’s equations via the finite element method with appropriate boundary conditions using COMSOL Multiphysics [Appendix D]. Here, graphene is modeled as a 0.5-nm-thick conducting boundary layer with a conductivity corresponding to the intraband transitions at room temperature [48]:

\[
\sigma(\omega) = -j \frac{e^2 E_F}{\pi \hbar^2} \frac{1}{\omega - j/\tau}.
\]  

(4.1)

Bloch boundary conditions were used to represent the periodic structure. The material properties of the substrate are obtained from tabulated data [49]. The simulated band structure for GPC1 is displayed in Fig. 4.3 (horizontal axis: plasmonic wavenumber, \(k_p\); vertical axis: frequency, \(f\)), where the eleven lowest-lying bands are shown along the high-symmetry points in reciprocal space. For this particular simulation, we use \(E_F = -0.38\) eV, obtained in the unpatterned region, as the exact value of \(E_F\) of the crystal is unknown. Simulations with differing \(E_F\) values reveal that the band diagram scales vertically in proportion to \(\sim \sqrt{|E_F|}\), which is a key signature of graphene plasmons [8, 19]. For example, Fig. 4.4 shows this \(E_F\) dependence in the degenerate mode frequency of plasmonic bands 5 and 6 at the \(\Gamma\)-point.

To demonstrate the plasmonic band formation in the graphene plasmonic crystal, we perform Fourier transform infrared spectroscopy (FTIR) at room temperature by normally irradiating an unpolarized far-infrared plane wave along the \(z\)-axis onto the device, which lies in the \(x-y\) plane. The wave vector \(k\) of the normally incident light has no component in the plane of the graphene, yet the corresponding \(k_x = k_y = 0\) line can still excite plasmonic modes at the \(\Gamma\) point \((k_p = 0)\) on the bands; such phase-matching and resultant plasmonic excitation would not be possible in
unpatterned graphene, which exhibits a continuous plasmonic dispersion relation with no plasmonic band formation.

Among all available Γ-point plasmonic modes, only two pairs of degenerate Γ-point modes, belonging to bands (5, 6) and bands (10, 11), can be excited, because the spatial symmetry of these specific plasmonic modes matches the spatial symmetry of the fields of the normally incident plane waves [50-52]. All other Γ-point modes behave differently than the radiation field under symmetry operations (such as reflections with respect to planes parallel to the z-axis or rotations about the z-axis), and thus cannot be excited despite their phase matching to the normally incident wave. To help appreciate this, we illustrate the symmetries of all Γ-point plasmonic modes of the eleven lowest-lying bands by displaying the simulated spatial profiles of their electric field in the z-direction, \( E_{pz} \), just above the graphene (Fig. 4.5). The Γ-point plasmonic mode belonging to band 7, for example, has a \( 120^\circ \) rotational symmetry about the z-axis, a symmetry that radiation fields do not possess, and thus cannot be excited.

This symmetry-based selection rule can be formally proved. The hexagonal lattice possesses the \( C_{6v} \) point group symmetry, and thus, each Γ-point mode hosted by the lattice exhibits definite symmetry transformation properties under any symmetry operation belonging to the \( C_{6v} \) group. However, one can show that the symmetry transformation properties of only the degenerate Γ-point modes on bands (5, 6) and those on bands (10, 11) match the symmetry transformation properties of normally incident plane waves, being described by the same irreducible representation of the \( C_{6v} \) group [51, 53]. The following section explains in detail plasmonic band excitation using group theory.
Figure 4.3 Simulated plasmonic band structure of GPC1 ($E_F = -0.38$ eV) along high symmetry points of a hexagonal reciprocal lattice (inset).
Figure 4.4 Simulated frequencies of degenerate Γ-point plasmonic modes on bands 5 and 6 with varying $E_F$ and a least-square fit to $f \propto \sqrt{|E_F|}$.

Figure 4.5 Simulated $E_{p,z}$ just above graphene for each Γ-point mode (corresponding to Fig. 4.3). Color bar shows normalized filed strength.
4.1.2 Symmetry-Based Plasmonic Band Selection Rule

Our graphene plasmonic crystals are realized by creating hexagonal lattices of apertures in a continuous graphene medium. The hexagonal lattice—whether its apertures are circularly or hexagonally shaped—possesses the $C_{6v}$ point group symmetry with a 6-fold rotation axis and six reflection planes (see Fig. 4.6 in case of the hexagonal lattice of circular apertures). Following the crystallographic convention, we denote the 12 symmetry operations (elements) constituting the group as $E, 2 \times C_6 (C_6, C_6^{-1}), 2 \times C_3 (C_3, C_3^{-1}), C_2, 3 \times \sigma_d (\sigma_d, \sigma_d', \sigma_d''),$ and $3 \times \sigma_v (\sigma_v, \sigma_v', \sigma_v'').$ The six irreducible representations of the $C_{6v}$ group are denoted as $A_1, A_2, B_1, B_2, E_1, E_2,$ also following the convention. The character table of the $C_{6v}$ group is in Table 4.1.

The structural symmetry of the hexagonal lattice leads to the physical symmetry of the plasmonic waves hosted by the structure [51]. The plasmonic dynamics in the lattice are governed by the electromagnetic wave equation. Its stationary solutions represent plasmonic eigenmodes, which, for $k_p = 0$, become the $\Gamma$-point plasmonic modes on different plasmonic bands, as discussed before. The fields of these $\Gamma$-point plasmonic eigenmodes manifest definite spatial symmetry properties under any operation belonging to the $C_{6v}$ group; this is evident from, for example, the $E_{p,z}$ spatial profiles of the $\Gamma$-point modes right above GPC1, shown in Fig. 4.5. Formally, the symmetric transformation properties of any given non-degenerate $\Gamma$-point plasmonic eigenmode or any given set of degenerate $\Gamma$-point plasmonic eigenmodes are described uniquely by one of the six irreducible representations of the $C_{6v}$ group. For GPC1, for example, the eleven lowest-lying $\Gamma$-point modes can be classified as in Table 4.2, based on the irreducible representations that describe their symmetry transformation properties.
Figure 4.6 Point group symmetry of the hexagonal lattice.

Table 4.1 Character table of the $C_{6v}$ point symmetry group.

<table>
<thead>
<tr>
<th>$C_{6v}$</th>
<th>$E$</th>
<th>$2C_6$</th>
<th>$3C_3$</th>
<th>$C_2$</th>
<th>$3\sigma_v$</th>
<th>$3\sigma_d$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_1$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>$A_2$</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
</tr>
<tr>
<td>$B_1$</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
</tr>
<tr>
<td>$B_2$</td>
<td>1</td>
<td>-1</td>
<td>1</td>
<td>-1</td>
<td>-1</td>
<td>1</td>
</tr>
<tr>
<td>$E_1$</td>
<td>2</td>
<td>1</td>
<td>-1</td>
<td>-2</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>$E_2$</td>
<td>2</td>
<td>-1</td>
<td>-1</td>
<td>2</td>
<td>0</td>
<td>0</td>
</tr>
</tbody>
</table>

Table 4.2 Classification of the $\Gamma$-point plasmonic eigenmodes of GPC1.

<table>
<thead>
<tr>
<th>Band indices of $\Gamma$-point plasmonic eigenmodes</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5, 6 (degenerate)</th>
<th>7</th>
<th>8</th>
<th>9</th>
<th>10, 11 (degenerate)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Irreducible representation</td>
<td>$E_2$</td>
<td>$E_2$</td>
<td>$B_1$</td>
<td>$E_1$</td>
<td>$B_2$</td>
<td>$E_2$</td>
<td>$E_2$</td>
<td>$E_1$</td>
</tr>
</tbody>
</table>

48
This assignment of a plasmonic eigenmode to a unique irreducible representation in Table 4.2 is done as follows. Consider, as an example, the non-degenerate Γ-point plasmonic mode on band 4 for GPC1 (f = 5.7 THz; Figs. 4.3 and 4.5). By inspection of the spatial profile of \( E_{p,z} \) of this mode (Fig. 4.5), we see that a 60° rotation of \( E_{p,z} \) about the z-axis flips its sign, i.e.,

\[
C_6(E_{p,z}) = -E_{p,z}, \quad (4.2)
\]

and a reflection of \( E_{p,z} \) along the y-axis maintains the field component, i.e.,

\[
\sigma_v(E_{p,z}) = E_{p,z}, \quad (4.3)
\]

These relations identify the characters of \( C_6 \) and \( \sigma_v \) operations on the band-4 Γ-point mode as -1 and 1, respectively. By comparing these results with Table 4.1, we see that the symmetry properties of this Γ-point mode are uniquely described by irreducible representation \( B_1 \).

As another example, for doubly-degenerate Γ-point modes on bands 5 and 6 for GPC1 (f = 6.8 THz; Figs. 4.3 and 4.5), each operation of the \( C_{6v} \) group may be expressed as a 2×2 matrix. For instance, a 180° rotation of the \( E_{p,z} \) fields of the degenerate modes about the z-axis flips their signs (Fig. 4.5), which can be expressed as

\[
C_2 \begin{pmatrix} E_{5,p,z} \\ E_{6,p,z} \end{pmatrix} = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} E_{5,p,z} \\ E_{6,p,z} \end{pmatrix}. \quad (4.4)
\]

Thus the character of \( C_2 \)—the trace of the 2×2 matrix on the right hand side—for these doubly-degenerate modes is -2. By comparing Table 4.1, we then see that the symmetry properties of these degenerate Γ-point modes on bands 5 and 6 can be described only by irreducible representation \( E_1 \). In this way, we complete Table 4.2.
Next, we consider the physical symmetry of the normally incident excitation plane waves. The symmetry properties of these free-space light waves can also be uniquely identified with one of the irreducible representations of the $C_{6v}$ group in exactly the same manner as above. Firstly, they are doubly degenerate due to the two orthogonal polarizations, and secondly, $180^\circ$ rotations about the $z$-axis flip the signs of their fields, i.e.:

$$
C_2 \begin{pmatrix} E_{\text{light},x} \\ E_{\text{light},y} \end{pmatrix} = \begin{pmatrix} -1 & 0 \\ 0 & -1 \end{pmatrix} \begin{pmatrix} E_{\text{light},x} \\ E_{\text{light},y} \end{pmatrix}. \quad (4.5)
$$

Therefore, the character of $C_2$ for the doubly-degenerate free-space waves is $-2$, just like the doubly-degenerate Γ-point plasmonic modes on bands 5 and 6, and thus, the symmetry properties of the free-space radiation are described uniquely by the $E_1$ irreducible representation.

Finally, as we look at Table 4.2, we see that among all the eleven lowest-lying Γ-point plasmonic modes of GPC1, only the symmetry properties of those on bands (5, 6) and (10, 11) are described by the $E_1$ irreducible representation, just like the symmetry properties of the normally-incident excitation plane waves. In other words, only these particular plasmonic modes respond in the same way as the normally-incident plane waves under the symmetry operations of the $C_{6v}$ group. This is the reason why only these specific Γ-point plasmonic modes, whose overlap integral with light fields does not vanish, are coupled to (and excited by) the free-space radiation, even though all Γ-point modes satisfy the phase matching condition.
4.1.3 Simulated Plasmonic Band Excitation

We now solve Maxwell’s equations with a plane-wave excitation to obtain the extinction, $1 - \frac{T}{T_0}$ ($T$ and $T_0$ are the light intensity transmitted through the on-substrate graphene device of concern and through the substrate only, respectively; Fig. 4.1a), which indicates the degree of absorption or reflection by the device. Figure 4.7a displays simulated extinction spectra of GPC1 with $E_F = -0.38$ eV for differing values of $\tau$. Simulation with either $x$- or $y$-polarized excitation light gives the same result. For $\tau = 5 \times 10^{-12}$ s (this relatively long momentum relaxation time is feasible with high-mobility exfoliated graphene [54, 55]) the extinction spectrum exhibits two peaks, confirming the selective excitation; the tall 6.8-THz peak [small 8.6-THz peak] is due to the excitation of the degenerate $\Gamma$–point plasmonic modes at exactly the same frequency on bands 5 and 6 [bands 10 and 11] in Fig. 4.3. These peaks, which can also be designed to occur in the mid-infrared [40], are due to Fano resonances between the plasmon modes and direct transmission through the graphene, similar to the Fano resonance in photonic crystal slabs [23]. With decreasing $\tau$, i.e. lowering the plasmon quality factor (Chapter 3), each peak broadens and decreases in amplitude in simulation. For $\tau = 5 \times 10^{-14}$ s, that is, commensurate with the mobility of our CVD-grown graphene, the peak due to the degenerate $\Gamma$ point on bands 5 and 6 remains observable at a slightly lowered frequency (6.7 THz), while the peak due to the degenerate $\Gamma$ point on bands 10 and 11 is unresolvable. Simulated extinction spectra with different geometric parameters show the same behavior (Fig. 4.7); in the low scattering regime, the extinction shows multiple peaks corresponding to a subset of $\Gamma$-point plasmonic modes allowed by the symmetry selection rule; in the high scattering regime (as in CVD graphene), a single broad peak appears, typically around the originally dominant peak.
Figure 4.7 Simulated extinction spectra of GPC1-4 for various values of $\tau$. $E_F = -0.38$ eV is used.
4.1.4 Experimental Excitation of the Plasmonic Bands

Our work employs lower-mobility CVD graphene with $\tau \sim 5 \times 10^{-14}$ s, as its large area is amenable to maximal coupling with the far-infrared beam from an Ever-Glo IR source (beam diameter: $\sim$8.75 mm). To ensure the measurement of only the one particular plasmonic crystal under examination, a mask with a pinhole (diameter $\sim$ 2 mm) is aligned directly behind the particular crystal to permit only its signal to be transmitted. Thus, we expect from the simulation that the extinction spectrum will exhibit a single broad peak. In fact, the measured spectrum (Fig. 4.8a, blue) of a GPC1 in the frequency range of 3–14 THz (above the lower cutoff frequency of the Thermo Fisher FTIR6700 system used, and below the absorption bands of SiO$_2$) exhibits a single broad peak near 6 THz with an overall decreasing background, in agreement with the shape of the extinction spectrum simulated with $\tau = 5 \times 10^{-14}$ s (the measurement is done in a N$_2$ atmosphere with a polyethylene windowed deuterated triglycine sulfate (DTGS/PE) far-IR detector; the transmission spectrum of N$_2$ is separately measured and this background spectrum is subtracted from every device spectrum). This peak is due to the excitation of the degenerate $\Gamma$-point plasmonic modes on bands 5 and 6. The emergence of the peak demonstrates the band structure formation by the periodic structuring. This is because in unpatterned graphene, the plasmonic dispersion curve does not form bands and thus cannot meet with the $k_x = k_y = 0$ line representing the normally incident light. This lack of coupling between the light and plasmons in unpatterned graphene is clearly seen in the measured extinction of the unpatterned graphene region (Fig. 4.8a, red). The monotonic spectrum is due to the background interaction between the light and graphene free carriers [8] (as described below); no peak is observed due to the lack of light-plasmon coupling.
The extinction spectrum of the unpatterned graphene sheet on the SiO2/Si substrate in the N2 atmosphere is also calculated using the scattering matrix method through an N2/graphene/SiO2/Si/N2 stack (Fig. 4.8). Scattering matrices are constructed by calculating the transmission and reflection coefficients at the material interfaces and propagation factors accounting for phase delay and attenuation through different material layers [57]. The material properties needed in these calculations are obtained from tabulated data [49]; the free charge carrier conductivity, σ, of graphene is also needed for these calculations (Eq. 4.1).

The transmission coefficient through an interface, $t$, the reflection coefficient at an interface, $r$, and the attenuation coefficient through a medium, $\alpha$, are calculated as below ($n$: refractive index of the medium; $d$: thickness of the medium; $Z_f$: free-space impedance).

![Figure 4.8 Schematics showing the coefficients of the scattering matrix through the materials.](image-url)
The scattering matrices through the interfaces are

\[
S_{ags} = \begin{pmatrix} r_{ags} & t_{ags} \\ t_{ags} & r_{sga} \end{pmatrix}, \quad S_{ss} = \begin{pmatrix} t_{ss} & t_{ss} \\ r_{ss} & r_{ss} \end{pmatrix}, \quad S_{sa} = \begin{pmatrix} r_{sa} & t_{sa} \\ t_{sa} & r_{sa} \end{pmatrix}, \quad S_{as} = \begin{pmatrix} t_{as} & t_{as} \\ r_{as} & r_{as} \end{pmatrix}
\]

The scattering matrices through the media are

\[
S_{SiO_2} = \begin{pmatrix} 0 & \alpha_{SiO_2}^{-1} \\ \alpha_{SiO_2} & 0 \end{pmatrix}, \quad S_{Si} = \begin{pmatrix} 0 & \alpha_{Si}^{-1} \\ \alpha_{Si} & 0 \end{pmatrix}
\]

To obtain the effect of all the elements above, any scattering matrix, \( S = \begin{pmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{pmatrix} \), is converted into its transmission counterpart, \( T' \), as

\[
T' = \frac{1}{S_{11} \det S} \begin{pmatrix} 1 & -S_{22} \\ S_{21} & S_{11} \end{pmatrix}
\]

So the total transmission matrix through the graphene/SiO\(_2\)/Si stack is

\[
T'^{total} = T_{ags}^{'} \cdot T_{SiO_2}^{'} \cdot T_{ss}^{'} \cdot T_{Si}^{'} \cdot T_{sa}^{'}
\]

and through the bare SiO\(_2\)/Si substrate is
\[ T_0^{\text{total}} = T_{sa}^n T_{xs}^n T_{Si}^n T_{sa}^n. \]

From these, the total scattering matrix is obtained via

\[ S^{\text{total}} = \frac{1}{T_{11}^{\text{total}}} \begin{pmatrix} T_{21}^{\text{total}} & \det T_{11}^{\text{total}} \\ 1 & -T_{12}^{\text{total}} \end{pmatrix}, \]

which gives the transmission intensity, \( T = \left| \frac{1}{T_{11}^{\text{total}}} \right|^2 \) and \( T_0 = \left| \frac{1}{T_{0,11}^{\text{total}}} \right|^2 \), enabling the calculation of the extinction spectrum and comparison with experiment (Fig. 4.9).

![Figure 4.9](image)

**Figure 4.9 (a)** Extinction spectra of unpatterned graphene (red) and GPC1 (blue), measured by FTIR spectroscopy. **(b)** Measured (red) and calculated (black) extinction spectra of an unpatterned graphene sheet. \( E_F = -0.38 \) eV and \( \tau \sim 5 \times 10^{-14} \) s, measured in the unpatterned region, are used in the calculation.
The monotonic extinction spectrum calculated in this way compares well with the measured extinction spectrum of unpatterned graphene (Fig. 4.9b), which confirms that the monotonic spectrum is due to the background interaction between the light and free charge carriers in graphene.

For the graphene plasmonic crystals, we can obtain the extinction peak frequencies by fitting the experimental spectra to a single-peak Fano resonance lineshape [58],

\[
1 - \frac{T}{T_0} = A T_B(\omega) \frac{q_f + \frac{2(\omega - \omega_0)}{\Gamma_p}}{1 + \frac{2(\omega - \omega_0)}{\Gamma_p}}^2 + b,
\]

where the fitting parameters are: \(A\) (amplitude), \(q_f\) (Fano parameter), \(\Gamma_p\) (plasmon damping rate), \(\omega_0\) (natural frequency), and \(b\) (screening parameter). Here \(T_B\) is the background extinction spectrum obtained experimentally from transmission through unpatterned graphene (Fig. 4.9a, red). This single Fano resonance fit well approximates the single broad extinction peak in each graphene plasmonic crystal in the heavy carrier scattering regime, and the \(R^2\) statistics of the fits are in the range of 0.88 to 0.95. The peak frequency obtained this way, 6.3 THz, is close to the simulated peak frequency at 6.7 THz. The \(\sim 5\%\) difference from the simulation is largely due to spatial variations in \(E_F\); that is, the value of \(E_F = -0.38\) eV used in the simulation is from the unpatterned graphene region, while the crystal region under examination in general assumes a different \(E_F\) value.

The observation of a single plasmon peak is robust across the remaining three plasmonic crystals with differing hexagonal lattice geometries (Fig. 4.10), further affirming the formation of the plasmonic band structure by the periodic patterning. As expected, the peak frequency varies from device to device, because the band structure is altered by the lattice geometry. In our
setup, the dependence of the peak frequency solely on geometric parameters cannot be closely examined, due in part to the photolithographic inaccuracy in controlling the geometric parameters to submicron precision, and more fundamentally because $E_F$ varies from device to device and from region to region, even within a single device (spatial variations of $E_F$ on CVD graphene can be on the order of 0.1 eV [44, 59]). In fact, we estimate the gross effective $E_F$ of each plasmonic crystal by matching the peak frequency between the measured and simulated spectrum, where the simulation uses $E_F$ as a fitting variable, and the size and shape of the apertures are estimated from the SEM as fixed parameters ($\tau$ is kept at $\sim 5\times10^{-14}$ s in this simulation, because extinction peak frequencies are not sensitive to $\tau$, as far as $\tau$ varies within the range expected for CVD graphene); the device-to-device variation of $E_F$ so estimated is up to $\sim 0.1$ eV (Fig. 4.8).

In the next section, a new set of graphene plasmonic crystals are fabricated with an improved fabrication procedure. The value of $E_F$ of each plasmonic crystal is explicitly measured.
Figure 4.10 Extinction spectra (blue) of GPC1-4 and least-square fits (black) to an expression based on a Fano resonance. The grossly estimated value of $E_F$ by simulation peak fitting is indicated for each crystal. The insets are SEM images of the crystals.
4.2 Graphene Plasmonic Crystals with Square Lattices

We now demonstrate that the symmetry-based selective light-plasmon coupling is a general feature not limited to the hexagonal lattices with $C_{6v}$ point group symmetry. We study graphene plasmonic crystals with square lattices of circular apertures that possess $C_{4v}$ point group symmetry, and experimentally confirm the symmetry-based selective light-plasmon coupling via FTIR.

We fabricate four graphene plasmonic crystals (Fig. 4.11a), each occupying an area of $6 \times 6$ mm$^2$, on the same single sheet of graphene. The graphene sheet is grown on copper foil by CVD and transferred onto a 500-µm thick single-crystal quartz substrate. The diameter of the circular aperture, $D$, is fixed at 3 µm across all crystals, but the lattice constant, $a$, varies from crystal to crystal: 3.65 µm, 4 µm, 4.5 µm, and 5 µm. Figure 4.11a shows one of the four crystals ($a = 4$ µm). Fabrication of these structures entails photolithography and O$_2$ plasma etching. We use an i-line stepper for photolithography with ~ 0.65 µm resolution, which gives robust control of the lattice geometry and aperture shape.

We first measure $\tau$ and $E_F$, which influence the plasmonic excitation linewidth and frequency, respectively. As in Section 4.1, to measure $\tau$, we leave an area of $\sim 6 \times 12$ mm$^2$ on the same graphene sheet unpatterned, and perform a Hall measurement via the 4-probe van der Pauw method. This yields $\tau \sim 6.5 \times 10^{-14}$ s, corresponding to a plasmonic $Q = \omega \tau$ of 2 at 5 THz. While $\tau$ may vary somewhat from one crystal to another, as $Q$ is low and plasmonic excitation linewidths are broad, we do not need to measure $\tau$ for each crystal, but use $6.5 \times 10^{-14}$ s, measured above, as an estimate of $\tau$ for each crystal.
Figure 4.11 (a) Micrograph of a graphene plasmonic crystal with a square lattice of circular apertures ($D = 3 \, \mu m; \ a = 4 \, \mu m$). (b) Near-IR extinction spectrum (blue, solid) of the crystal in part (a), with a Gaussian-broadened-step-function fit (red, dashed).

On the other hand, we measure $E_F$ for each plasmonic crystal, as the plasmonic excitation frequency depends sensitively on $E_F$ (the unpatterned area gives $E_F \sim 0.597 \, eV$ from the Hall measurement). To this end, we measure interband transition via near-IR spectroscopy by using the same Thermo Fisher FTIR6700 system with N$_2$ atmosphere. This is separate from the far-IR spectroscopy, the main experiment to be performed to interrogate plasmon-light coupling, which relates to intraband transition. We measure transmission spectrum, $T$, of near-IR light through
each plasmonic crystal on the quartz substrate. The reference spectrum, \( T_0 \), through the quartz substrate area not covered by graphene is also measured (this quartz substrate is transparent in far-IR frequencies below \(~8\) THz and also from 100 to 1875 THz, to which our near-IR range belongs). These lead to the near-IR extinction spectrum \( 1-T/T_0 \). We use a quartz-halogen white light lamp as the near-IR source (through an aperture with \(~4\) mm diameter), a mercury cadmium telluride (MCT-A) detector, and a CaF\(_2\) beam splitter. A mask with a pinhole (diameter \(~5\) mm) is placed behind any one plasmonic crystal under examination, to align the crystal to the source and detector. Near-IR light with photon energy \( \hbar \omega \) in excess of \( 2E_F \) can induce interband transition [22]. The resulting absorption of the light creates the extinction spectrum in the form of a Gaussian-broadened step function, as in Fig. 4.11b, with the Gaussian peak frequency centered at \( \hbar \omega = 2E_F \) [44]. From this, we estimate \( E_F \) to have magnitudes of \(~0.400\) eV, \(~0.417\) eV, \(~0.423\) eV and \(~0.457\) eV for the crystals with \( a = 3.65 \) μm, \( 4 \) μm, \( 4.5 \) μm and \( 5 \) μm, respectively. Since our graphene is hole doped, as confirmed by the Hall measurement, the actual \( E_F \) values are negative (with the Dirac point set as the reference \(~0\) eV).

### 4.2.1 Plasmonic Band Structures and Excitations for Square Lattices

With the measured \( E_F \), we simulate the plasmonic band structure of the square lattice with \( a = 3.65 \) μm (subsection 4.1.1). Nine plasmonic bands emerge in the frequency range of 3.5-8 THz with \( k_p \) (Bloch wavenumber for plasmons) varying along the Γ-M direction in the square reciprocal lattice (Fig. 4.12). Normally incident far-IR light with no wavevector component in the graphene plane can couple only to plasmonic modes lying at the Γ-point \( (k_p = 0) \) of the bands. Moreover, among these Γ-point modes, only a select subset can be excited by the light (subsection 4.1.2); *i.e.*, symmetry between light fields and plasmonic modes must match for excitation. The plasmonic modes assume the point group symmetry of the lattice. For light and
plasmonic modes to have matching symmetry, their irreducible representations under the symmetry group must be the same.

To identify the excitable plasmonic modes according to this principle, we first consider the structural symmetry of the square lattice of circular apertures. It possesses $C_{4v}$ point group symmetry with the 4-fold rotation axis—z-axis orthogonal to graphene plane—and four reflection planes (Fig. 4.12b). Following convention, we denote the eight symmetry operations as $E$, $2 \times C_4$ ($C_4$, $C_4^{-1}$), $C_2$, $2 \times \sigma_v$ ($\sigma_v'$, $\sigma_v''$), and $2 \times \sigma_d$ ($\sigma_d'$, $\sigma_d''$), and the five irreducible representations as $A_1$, $A_2$, $B_1$, $B_2$, and $E$, with the character table in Fig. 4.12c [51]. Since the $\Gamma$-point plasmonic modes possess the same $C_{4v}$ point group symmetry, each $\Gamma$-point mode can be assigned to one of the five irreducible representations. This assignment is done by examining the symmetry of the spatial profiles of $E_{p,z}$, which is obtained from the band simulation (Fig. 4.12d). At the same time, the normally incident plane-wave light with any linear polarization possesses symmetry with irreducible representation $E$ ($180^\circ$ rotation about the z-axis flips the signs of the light fields) under the $C_{4v}$ point symmetry group. Only the degenerate $\Gamma$-point modes on bands 2 and 3, and those on bands 6 and 7, are described by irreducible representation $E$ (Fig. 4.12d), matching the symmetry of light. Hence only these two pairs of doubly degenerate $\Gamma$-point modes are excitable, independent of light polarization.
Figure 4.12 Simulations/theory of a square lattice of circular apertures ($C_{4v}$ point symmetry group; $D = 3 \, \mu m, a = 3.65 \, \mu m, E_F = -0.4 \, eV, \tau = 5 \times 10^{-12} \, s$). (a) Simulated band structure along the $\Gamma$-$M$ direction in the square reciprocal lattice. (b, c) Elements and character table of the $C_{4v}$ point symmetry group. (d) $E_{p,z}$ profile of each $\Gamma$-point plasmonic mode, which is labeled by the index of the band it belongs to and with the irreducible representation under the $C_{4v}$ point symmetry group. The color bar shows normalized field strength. (e) Simulated extinction spectrum. The spectrum is independent of the polarization of the excitation plane wave light.
Figure 4.13 Simulated extinction spectrum of the square-lattice graphene plasmonic crystal with circular apertures ($D = 3 \, \mu m$, $a = 3.65 \, \mu m$, $E_F = -0.4 \, eV$ and $\tau = 5 \times 10^{-12} \, s$), which is identical to Fig. 4.12e of the main text, but now shown with the $E_z$ profiles at the extinction peak frequencies. The color bar shows normalized field strength.

The light’s selective coupling to these particular modes is confirmed by a separate COMSOL simulation that calculates the extinction spectrum with plane-wave light excitation (Fig. 4.12e; the spectrum is independent of the polarization of the light). The two extinction peaks obtained here occur at exactly the same frequencies of the two pairs of degenerate $\Gamma$-point modes obtained from the band simulation. Moreover, the extinction spectrum simulation with graphene illuminated by the light also gives a profile of the electric field on the graphene plane at every frequency. This field profile corresponds to the response of graphene under the driving field of the incident plane wave. We call the $z$-component of this response electric field $E_{z}$, in order to compare it to the $E_{p,z}$ profile of the plasmonic mode from the band simulations. The profiles of $E_z$ that can be driven by the incident light at the extinction peaks should show the same feature as the $E_{p,z}$ profiles of the corresponding $\Gamma$-point modes with the matching symmetry to light. In fact, the doubly degenerate $E_z$ profiles at the extinction peaks (Fig. 4.13) agree with
the $E_{p,z}$ profiles of the two pairs of the degenerate $\Gamma$-point plasmonic modes (Fig. 4.12d), showing the consistency between the band and extinction spectrum simulations.

This symmetry-based light-plasmon coupling is generally applicable. For example, while the square lattice of circular apertures with $a = 5 \ \mu$m has a differing plasmonic band structure (Fig. 4.14a), again only those $\Gamma$-point modes assigned to irreducible representation $E$ can be excited by the plane-wave light. The $E_{p,z}$ profiles of the $\Gamma$-point modes obtained from the band simulation (Fig. 4.14b) show that four pairs of doubly degenerate $\Gamma$-point modes—in bands (2, 3), (5, 6), (8, 9), and (10, 11)—are assigned to the irreducible representation $E$. The simulated extinction peaks (Fig. 4.14c; independent of the polarization of light) indeed occur at exactly the same frequencies as those of the four pairs of degenerate modes, confirming the symmetry-based coupling. Moreover, the four pairs of doubly degenerate $E_z$ profiles from the extinction spectrum simulation at these four peaks (Fig. 4.15) agree with the $E_{p,z}$ profiles for the four pairs of doubly degenerate $\Gamma$-point plasmonic modes from the band simulation (Fig. 4.14b). One apparent exception is at the lowest-frequency peak at ~5 THz; the two degenerate $E_z$ profiles at this peak (Fig. 4.15) seemingly differ from the two degenerate $E_{p,z}$ profiles of the degenerate $\Gamma$-point modes in bands (2, 3) at the same frequency (Fig. 4.14b). But these two pairs of degenerate field profiles are actually equivalent, as one pair is obtained by linear superposition (addition and subtraction in this case) of the other; either pair forms a basis for the field solutions at the degenerate frequency.
Figure 4.14 Simulations of a square lattice of circular apertures ($C_4v$ point symmetry group; $D = 3 \ \mu m$, $a = 5 \ \mu m$, $E_F = -0.457 \ \text{eV}$, $\tau = 5 \times 10^{-12} \ \text{s}$). (a) Simulated band structure along the $\Gamma$-M direction. (b) $E_{p,z}$ profile of each $\Gamma$-point plasmonic mode, labeled by band index and irreducible representation. (c) Simulated extinction spectrum (the inset is a zoom-in of the peaks in the 7-8 THz range). The spectrum is independent of the polarization of the excitation plane wave light.
Figure 4.15 Simulated extinction spectrum of the square-lattice graphene plasmonic crystal with circular apertures ($D = 3 \, \mu m$, $a = 5 \, \mu m$, $E_F = -0.457 \, eV$ and $\tau = 5 \times 10^{-12} \, s$), which is identical to Fig. 4.14c of the main text, but now shown with the $E_z$ profiles at the extinction peak frequencies.

4.2.2 Experimental Results and Effects of Changing $a$ with a Fixed $D$

Figure 4.16a compares simulated extinction spectra for four square lattices of circular apertures (top and bottom panels are identical to Figs. 4.14c and 4.12e), where peaks correspond to the plasmon-light coupling with matching symmetry. As $a$ decreases, the spacing between the first and second peaks increases, because as the first Brillouin zone widens with decreasing $a$, fewer plasmonic bands, and thus fewer $\Gamma$-point modes, occupy a given frequency range.

These simulated extinction spectra assumed $\tau = 5 \times 10^{-12} \, s$ (corresponding to a plasmonic $Q$ of $\sim150$ at 5 THz). While such high-quality large-area graphene is not yet available, the hypothetically sharp extinction lines were used to show the light-plasmon coupling at more than one frequency, without being masked by broad lines. With the actual $\tau$ of $6.5 \times 10^{-14} \, s$ shown in Chapter 4, simulated extinction spectra are greatly broadened (Fig. 4.16b). For a larger $a$, with which the lowest- and second-lowest extinction peak frequencies are closer (Fig. 4.16a), the
second peak is blurred away by the now broadened first peak (Fig. 4.16b). For a smaller $a$, which separates the two peak frequencies farther (Fig. 4.16a), both peaks become resolvable (Fig. 4.16b).

Figure 4.16 Measurements and simulations of four square lattices with circular apertures ($C_4v$ point symmetry group; $D$ is fixed at 3 μm across the four lattices; $a$ varies from 5 μm to 3.65 μm). (a) Simulated extinction spectra of all four lattices ($\tau = 5 \times 10^{-12}$ s). Top and bottom panels are identical to Figs. 4.14c and 4.12e, respectively. The frequency-dependent dielectric constant of quartz used in the simulations is from the literature [49]. (b) The same simulations as (a), but with the measured $\tau$ of 6.5 $\times 10^{-14}$ s. (c) Measured Far-IR extinction spectra from FTIR experiments.
Figure 4.16c shows the measured extinction spectra for all four plasmonic crystals, where these far-IR spectroscopy measurements are done again with the Thermo Fisher FTIR6700 system, as described in Sections 4.2 and 4.3. The measured extinction spectra of Fig. 4.16c confirm the essence of the foregoing theory. First, when $a$ is larger (5 µm and 4.5 µm), only one peak appears, but when $a$ is reduced (4 µm and 3.65 µm), the spectrum shows slower roll-off because light coupling to the next excitable mode is less blurred away with decreasing $a$, as explained with the simulation (Fig. 4.16b). Second, the first plasmon-light coupling peaks indeed occur around at 5 THz, as predicted by simulation. Third, the frequency of the first peak increases as $a$ is reduced from 5 µm to 4.5 µm to 4 µm, but does not increase any more as $a$ is further reduced to 3.65 µm, which is largely consistent with simulation (Fig. 4.16b); in simulation, the frequency increases as $a$ is reduced from 5 µm to 4.5 µm to 4 µm, just like in the measurements, and then decreases as $a$ is further reduced to 3.65 µm.

While the symmetry-based selective excitation played a key role in demonstrating the formation of the plasmonic band structure in the present work, other coupling schemes [60, 61] may enable the plasmonic bands beyond the Γ points to be excited and probed.
Chapter 5

Graphene Plasmonic Band Engineering

With the graphene plasmonic band structures established in the previous chapter, we now discuss various methods to manipulate the band structures. The plasmonic band structures depend on a number of parameters: carrier concentration, lattice dimension, aperture shape and lattice symmetry. This chapter discusses each of their effects on the band structures manifested in the extinction spectra, either in experiments or in simulations\textsuperscript{iv}.

5.1 Carrier Concentration

We upshift the overall $|E_F|$ distribution across the entire sample in section 4.1 containing the four crystals via global chemical doping of holes, and verify if the peak frequency of each crystal increases. This method is beneficial in the face of device-to-device variation of $E_F$, since it does

\textsuperscript{iv} Large portions of this chapter are derived from a paper in publication by the author [21, 56].
not require the exact knowledge of the spatial distribution of $E_F$. After chemically hole-doping the sample by exposure to 70% HNO$_3$ vapor for 1 min, which shifts the measured $E_F$ from -0.38 eV to -0.55 eV in the unpatterned region, the measured peak frequency in every crystal shifts upward consistently (Fig. 5.1). Incidentally, the frequency upshift factor indicated in Fig. 5.1 is not constant amongst the devices, because the pre-doping $E_F$ spatial profile is non-uniform, doping itself may not be perfectly uniform, and the *ex situ* doping procedure may cause a slightly different crystal position to be probed by the FTIR before and after doping.

![Figure 5.1](image.png)

*Figure 5.1* Extinction spectra (thin lines) of the four graphene plasmonic crystals before (black) and after (red) hole doping. The factor by which the peak frequency increases is shown for each crystal. Bold lines are least-squares fits to an expression based on the Fano resonance (Chapter 4). A vertical cumulative offset of 10% is added between the spectra from different crystals for clarity.
5.2 Lattice Dimension

In Chapter 4, the effects of varying $a$ with a fixed $D$ are already shown using the graphene plasmonic crystals with square lattices. This section presents additional analysis of the lattice-dimension dependence of the band structure inferred by the extinction spectra.

5.2.1 Change $a$ with a Fixed $D/a$

![Graph showing simulated extinction spectra of circular-aperture hexagonal lattices with varying $a$.](image)

**Figure 5.2** Simulated extinction spectra of circular-aperture hexagonal lattices with varying $a$. (a) $\tau = 5 \times 10^{-12}$ s; (b) $\tau = 5 \times 10^{-14}$ s as in CVD graphene. $E_F = -0.38$ eV is used for all simulations.

As an example, let us consider GPC1-type plasmonic crystals—hexagonal lattice of circular apertures—as demonstrational vehicles (Fig. 4.1). Simulations show that for an increasing $a$ with a fixed $D/a$—this represents a simple scaling of the crystal, i.e., the hexagonal lattice and its inner structural details are stretched by the same factor in all directions—both the multiple
plasmonic peak frequencies in the weak scattering regime (Fig. 5.2a) and the single plasmon peak frequency in the heavy scattering regime decrease (Figs. 5.2b). This is explained as follows. As the plasmonic wavenumbers $k_p$’s at Brillouin zone boundaries are inversely proportional to the lattice constant $a$, the Brillouin zones become smaller with the increasing $a$, while the approximate relation $f \propto \sqrt{k_p}$ is maintained within Brillouin zones, regardless of the increasing $a$. Therefore, for a larger value of $a$, the plasmonic bands cross both the Brillouin zone boundaries and the $\Gamma$ points at lower frequencies. Therefore, the extinction peaks appear at lower frequencies with the increasing $a$.

### 5.2.2 Change $D$ with a Fixed $a$

![Figure 5.3](image.png)

Figure 5.3 Simulated extinction spectra of circular-aperture hexagonal lattices with varying $D$. (a) $\tau = 5 \times 10^{-12}$ s; (b) $\tau = 5 \times 10^{-14}$ s as in CVD graphene. $E_F = -0.38$ eV is used for all simulations.
Again for an example hexagonal lattice of circular apertures, when $D$ is increased for a fixed $a$ (we choose $a = 3 \ \mu m$ here)—this corresponds to the change in the inner structure of the lattice, although the aperture shape remains to be circular— the extinction spectrum changes in a more complicated manner. In the low scattering regime ($\tau = 5 \times 10^{-12} \ \text{s}$; Fig. 5.3a), for $D/a = 0.6$, a single peak appears, but as $D/a$ is increased, second and even third peaks emerge in the extinction spectra, and the frequency of the original single peak first increases and then decreases back. These richer behaviors reflect that as the inner structure of the lattice is changed, the band structure—and thus the subset of $\Gamma$-point plasmonic modes that satisfy the symmetry-based selection rule—is altered in a complex manner, as expected. In the heavy scattering regime ($\tau = 5 \times 10^{-14} \ \text{s}$; Fig. 5.3b), while the second and third peaks are largely blurred out, the behavior of the original single peak (that is now broadened) remains the same with the increasing $D$ for a fixed $a$; its frequency first increases, and then decreases back.

### 5.3 Aperture Shape

From the study of the foregoing sections, we can expect that the change of the aperture shape—another way of modifying the inner structure of the lattice—will also alter the band structure (and thus the extinction spectra) in a complex manner. To demonstrate, we compare a hexagonal lattice of circular apertures ($a = 6 \ \mu m$, $D = 5 \ \mu m$) and a hexagonal lattice of hexagonal apertures ($a = 6 \ \mu m$, $D' = 5 \ \mu m$; GPC4). In the weak scattering regime (Fig. 5.4a), the positions and resonance strengths of multiple plasmonic peaks differ in nontrivial ways between the extinction spectra of the two lattices. It follows that in the heavy scattering regime (Fig. 5.4b), the single plasmonic peaks occur at two noticeably differing frequencies.
Figure 5.4 Simulated extinctions for a hexagonal lattice of circular apertures \((a = 6 \mu m, D = 5 \mu m)\) and a hexagonal lattice of hexagonal apertures \((a = 6 \mu m, D^* = 5 \mu m; GPC4)\). (a) \(\tau = 5 \times 10^{-12} s\); (b) \(\tau = 5 \times 10^{-14} s\). \(E_F = -0.38\ eV\).

5.4 Lattice Symmetry Breaking

The above section leads us to investigate how the light-plasmon coupling behaviors are altered when the aperture shape is modified to break the lattice symmetry.

5.4.1 Degeneracy Lifting

As a first demonstrational example, we change the square lattice of circular apertures with a diameter of 3 \(\mu m\) (as used in the experiment in Section 4.3) into a square lattice of elliptical apertures, where the ellipse is elongated in the \(y\)-direction with semi-major and -minor axes of 1.7 \(\mu m\) and 1.5 \(\mu m\), respectively (Fig. 5.5a). The \(C_{4v}\) point group symmetry (Fig. 4.11b) is reduced to the \(C_{2v}\) point group symmetry, whose character table is in Fig. 5.5b. Under the \(C_{2v}\) point symmetry group, the normally-incident \(x\)- and \(y\)-polarized plane-wave lights are described
by distinct irreducible representations, $B_2$ and $B_1$, respectively. Thus only $\Gamma$-point plasmonic modes whose symmetry properties are represented by $B_2$ or $B_1$ will be excited, with the former [latter] excited by $x$-polarized [$y$-polarized] light.

We verify this for a square lattice of elliptical apertures with $a = 3.65 \, \mu m$ via extinction spectrum simulation with $x$- and $y$-polarized light. As seen in Fig. 5.5c, $E_z$ profiles at the two extinction peaks excited by $x$-polarized light have the symmetry described by $B_2$ irreducible representation, and $E_z$ profiles at the other two extinction peaks excited by $y$-polarized light have the symmetry described by $B_1$ irreducible representation, as predicted. Importantly, the $E_z$ profiles of the two polarization-dependent peaks around 5 THz [7 THz] (Fig. 5.5c) have the same spatial features as the $E_{p,z}$ profiles of the $\Gamma$-point degenerate plasmonic modes in bands 2 and 3 [bands 6 and 7] (Fig. 4.11d). This shows that as we alter the aperture from circle to ellipse in the square lattice, each pair of the doubly-degenerate $\Gamma$-point plasmonic modes that were polarization-independently excited at a single frequency now split into the two non-degenerate $\Gamma$-point plasmonic modes, which are excited at two different frequencies and polarizations.

A similar change in the light-plasmon coupling behavior occurs as the aperture is once again altered from circle to ellipse for a square lattice with $a = 5 \, \mu m$ (Fig. 5.6d vs. Fig. 4.13). Each of the four peaks of Fig. 4.13c—excitation of four pairs of doubly degenerate $\Gamma$-point plasmonic modes in bands (2, 3), (5, 6), (8, 9), and (10, 11) in the square lattice of circular apertures—now splits into two peaks (Fig. 5.5d), one described by irreducible representation $B_2$ and thus excitable by $x$-polarized light, and the other by irreducible representation $B_1$ and excitable by $y$-polarized light. $E_z$ profiles at the extinction peaks in this elliptical aperture case (Fig. 5.5d)
exhibit the same spatial features as $E_{p,z}$ profiles in Fig. 4.13b in the circular aperture case; this shows once again degeneracy lifting.

**Figure 5.5** Simulations/theory of two square lattices of elliptical apertures ($C_{2v}$ point symmetry group). (a, b) Elements and character table of the $C_{2v}$ group. (c) Simulated extinction spectra of a square lattice ($a = 3.65 \mu m$) of elliptical apertures (semi-minor axis = 1.5 \mu m, semi-major axis = 1.7 \mu m) with $x$-polarized (black, dashed) and $y$-polarized (red, solid) excitation light. $E_F = -0.4$ eV, $\tau = 5 \times 10^{-12}$. (d) The same simulation, but with $a = 5 \mu m$ and $E_F = -0.457$ eV. The bottom panel is a zoom-in of the 7-8 THz range. Both parts (c) and (d) show $E_z$ profiles at the peaks; their labels are the indices of the particular bands in Figs. 4.11 and 4.13, to which the original degenerate $\Gamma$-point plasmonic modes (before the degeneracy removal) belong.
5.4.2 Excitation of Inactive Modes

Besides the degeneracy removal of already excitable modes, the aperture shape alteration can also make originally inactive plasmonic modes excitable [50, 51]. To demonstrate, we now take a hexagonal lattice [21] and modify the aperture from circle ($D = 3 \, \mu m$, $a = 4 \, \mu m$; Fig. 5.6) to ellipse (semi minor axis = 1.5 \, \mu m, semi-major axis = 1.7 \, \mu m along the y-direction; Fig. 5.7), reducing the point group symmetry from $C_{6v}$ to $C_{2v}$. Figure 5.6 shows the simulated band structure of the hexagonal lattice of circular apertures with each $\Gamma$-point mode $E_{p,z}$ profile assigned to a $C_{6v}$ irreducible representation. Its doubly degenerate $\Gamma$-point plasmonic modes (4, 5), (8, 9) or (13, 14) have symmetry described by $E_1$ irreducible representation under the $C_{6v}$ point group, which is also the irreducible representation of both $x$- and $y$-polarized light under the same group [21], and thus only these modes can couple to light. Indeed, in the simulated extinction spectrum (Fig. 5.6e), three peaks appear at exactly the same frequencies of the three pairs of degenerate modes (see Fig. 5.7 for their $E_z$ profiles).

When aperture changes from circle to ellipse in the hexagonal lattice, symmetry is reduced to the $C_{2v}$ point group (Figs. 5.8a, b), and most of the extinction peaks (Figs. 5.8c, d) show degeneracy removal (subsection 5.4.1)—peak splitting with polarization dependence—as compared to Fig. 5.6e. These split peaks of Figs. 5.7c, d are labeled with the indices of the bands in Fig. 5.6a—(4, 5), (8, 9), and (13, 14)—that host the original degenerate modes, where we identify the original modes by comparing the $E_{p,z}$ profiles in Fig. 5.6 and the $E_z$ profiles at the peaks in Fig. 5.8.
Figure 5.6 Simulations/theory of a hexagonal lattice of circular apertures ($C_{6v}$ point symmetry group; $D = 3 \, \mu \text{m}, \, a = 4 \, \mu \text{m}, \, E_F = -0.38 \, \text{eV}, \, \tau = 5 \times 10^{-12} \, \text{s}, \, \text{SiO}_2/\text{Si substrate}$). (a) Simulated band structure along the $\Gamma$-M direction in the hexagonal reciprocal lattice. (b) $E_{p,z}$ profile of each $\Gamma$-point plasmonic mode. (c, d) Elements and character table of the $C_{6v}$ point symmetry group. (e) Simulated extinction spectrum (inset is a zoom-in of the 8-8.8 THz range). The spectrum is independent of polarization of excitation plane wave light.
Figure 5.7 Simulated extinction spectrum of the hexagonal-lattice graphene plasmonic crystal with circular aperture ($D = 3$ μm, $a = 4$ μm, $E_F = -0.38$ eV and $\tau = 5 \times 10^{-12}$ s), which is identical to Fig. 5.6(e) of the main text, but now shown with the $E_z$ profiles at the extinction peak frequencies.

The extinction spectra of Figs. 5.8c, d also show a new aspect of broken-symmetry: two additional peaks, labeled 1 and 11, are excited by light with y- and x-polarizations, respectively. These two peaks are labeled 1 and 11, as their $E_z$ profiles (Figs. 5.8c and d) show the same spatial features as the $E_{p,z}$ profiles of $\Gamma$-point modes in bands 1 and 11 for the hexagonal lattice with the original circular apertures (Fig. 5.6b). These two modes, described by $B_1$ and $B_2$ irreducible representations under the $C_{6v}$ group (under which the symmetry of light of any polarization is described by $E_1$ irreducible representation), were thus inactive in the hexagonal lattice of circular apertures and did not show up in Fig. 5.6e. By contrast, in the hexagonal lattice with elliptical aperture, they are described by $B_1$ and $B_2$ irreducible representations under the $C_{2v}$ group, just as the y- and x-polarized lights are, and therefore are excited. A gradual change from circular to elliptical aperture is presented in Fig. 5.9, showing again the emergence of the active modes from the originally inactive modes.
Figure 5.8 Simulations/theory of a hexagonal lattice of elliptical apertures (C\textsubscript{2v} point symmetry group; \(a = 4 \, \mu\text{m}, \) elliptical aperture semi minor axis = 1.5 \(\mu\text{m}, \) semi-major axis = 1.7 \(\mu\text{m}, \) \(E_F = -0.38 \, \text{eV}, \) \(\tau = 5 \times 10^{-12} \, \text{s}, \) SiO\textsubscript{2}/Si substrate). (a, b) Elements and character table of the C\textsubscript{2v} point symmetry group. (c, d) Simulated extinction spectra with x- and y-polarized light. The inset in part (c) shows a zoomed-in view of the 8-9 THz range. Both parts (c) and (d) show \(E_z\) profiles at the peaks, and their labels are indices of the particular bands from Fig. 5.6a, to which the original degenerate \(\Gamma\)-point plasmonic modes (before the degeneracy removal) belong.
Figure 5.9 Simulated extinction spectra of the hexagonal-lattice graphene plasmonic crystals with the aperture shape changing from circle to ellipse with the semi-major axis gradually increasing from 1.5 μm to 1.7 μm. Part (a) is with x-polarized light, with Part (b) zooming in the 8-9 THz range of Part (a). Part (c) is with y-polarized light, with Parts (d) and (e) zooming in the 3.9-4.4 THz and 8-9 THz ranges of Part (c). These plots subsume Fig. 5.6e for the case of 1.5 μm (circular aperture) and Figs. 5.8c, d for the case of 1.7 μm (most elongated ellipse) of the main text. The $E_z$ profiles shown here for extinction peaks are for the 1.7 μm case. Degeneracy removal (frequency splitting and polarization dependence) for certain modes and activation of originally inactive modes are both clearly seen.
The studies in this chapter show that the plasmonic band structure can be richly manipulated by means of the crystal geometry, symmetry and carrier tuning. This is a powerful feature for plasmonic band engineering, and will become even more useful, as the control of the spatial variation of the Fermi level of large-area graphene and its quality further improve. The degeneracy removal, polarization dependence control, and activation switch via alteration of the lattice symmetry may lead to useful far-IR and terahertz applications, such as polarization-dependent frequency filters with subwavelength confinement.
Chapter 6

Experimental Setups

As previous chapters described briefly, the 2DEG interferometer is measured in a cryogenic probe station using a vector network analyzer, and the graphene plasmonic crystals are characterized with FTIR. The general operation instructions of these systems can be found in the equipment manuals. However, for future reference, this chapter serves as a record of the specific setups and procedures required by the plasmonic structures studied in this thesis.

6.1 Setup and Procedures for 2DEG Interferometer Measurements

Measurements of the 2DEG interferometer are conducted in a cryogenic probe station as shown in Fig. 6.1. It has the capability to connect to two radio-frequency (RF) probes, two DC probes and a back-gate DC supply probe. The experiments in this thesis only require RF probes. They are connected to the vector network analyzer, which supplies AC signals up to 50 GHz.
The sample chips are mounted onto a copper sample holder using silver paint. The sample holder has an electrical connection to the back-gate inside the inner chamber. A microscope camera is attached above the chamber optical windows. This enables us to observe while the RF probes are manually touched down onto the on-chip CPWs.

The chambers are pumped down to about $10^{-7}$ Torr before the measurements. Helium gas is transferred into the outer chamber, which gradually cools down the samples to 4.2 K. There are three temperature controllers with heaters that monitor and separately adjust the temperatures of the sample holder, the probes and the radiation shield on the inner chamber.

Prior to taking measurements of the interferometer, a calibration is performed using the multi-line TRL method [33] to remove loss and phase delay in the microwave cables and probes. A set of thru/open/short structures and CPWs of different lengths are fabricated on an undoped GaAs substrate [27], using the same fabrication procedure (Appendix A) with the same signal-line and ground-line geometries as those of the interferometer CPWs. Cascade Microtech WinCal XE 4.2 software is used to calibrate and analyze the signals recorded by the network analyzer. (The detailed WinCal XE settings can be found in Ref. [27].) After the calibration, the reference planes are set to the tips of the RF probes. This calibration step is done each time the system is set to a new temperature.
Figure 6.1 Photographs of the measurement systems and setup for the 2DEG interferometer. (a) LakeShore cryogenic probe station with capability to connect to two RF probes, two DC probes, a backgate DC power supply, a vacuum pump, a helium dewar and a microscope camera. (b) Agilent Technologies E8364A PNA Series Network Analyzer 45 MHz – 50 GHz. (c) Two LakeShore 332 Temperature Controllers. The left one has two monitors. (The top two power supplies are not used). (d) The inner and outer chambers of the probe station. (e) Inside the inner chamber: the GaAs substrate and 2DEG interferometer chips are mounted onto the sample holder. The two RF probes are positioned above a chip before touch-down.
6.2 FTIR for Graphene Plasmonic Crystals

Both the far- and near-IR spectroscopy measurements of graphene plasmonic crystals are carried out using the Thermo Fisher Nicolet FTIR6700 spectrometer. Figure 6.2 shows the system setup. The sample chamber is purged with N₂ gas in order to reduce moisture in the air before any experiment.

For near-IR measurements, a quartz-halogen white-light source, a CaF₂ beam splitter, and an MCT-A detector are used. The MCT-A detector is cooled using liquid nitrogen. The iris diameter for the light source is set to ~4 mm. Larger iris diameter would let too much light through and saturate the detector. For far-IR, an Ever-Glo light source, a solid substrate and a DTGS/PE detector are used. The iris diameter is set to ~8.75 mm.

The measurements are controlled and recorded using the OMNIC software. After selecting the correct source, beam splitter and detector in the software, the height of the sample holder aperture, without the sample on, is adjusted manually until the peak-to-peak magnitude of the interferogram is maximized. An automated alignment procedure is then taken to align the source, beam splitter and detector.

In the software experiment setup, the “final format” is set up “%Transmission”. A background spectrum of the chamber atmosphere is collected and is removed from further sample measurements.

A large number of scans are conducted for every sample, typically lasting about 10 minutes per sample, in order to average out ambient noise as much as possible. The resultant data is the
transmission $T$ of the graphene on the substrate. A separate transmission $T_0$ is taken for the bare substrate so that the extinction $1 - T/T_0$ can be calculated post-experiment.

**Figure 6.2** Photographs of the FTIR system for the graphene plasmonic crystals. (a) The Thermo Fisher Nicolet FTIR6700 spectrometer. (b) The sample chamber with a sample holder inserted. (c) DTGS and MCT-A detectors are both placed in their holders. The OMNIC software can select which detector to use. (d) Three beam splitters, for far-, mid- and near-IR, are available. The user can choose and place one manually.
Appendix A

Fabrication Recipes

The detailed fabrication steps used in this thesis are listed below. Some portions have been published in Refs. [6, 7, 30]. The author acknowledges William Andress, Hosang Yoon, Charles Marcus group (formerly at Harvard University) and Jing Kong Group (MIT) for contributing to some parts of the recipes.

AlGaAs/GaAs 2DEG Fabrication

Photolithography

1. Successively clean the AlGa/GaAs chip in trichloroethylene (TCE), acetone and isopropyl alcohol (IPA) for 5 min each in ultrasound.
2. Blow dry with N₂ gas. Singe on hotplate at 160 °C for 5 min.
3. Spin Shipley 1805 photoresist (1818 for thick gates):
   500 rpm, 100 rpm/sec ramp, 5 sec.
   4000 rpm, 1000 rpm/sec ramp, 30 sec.
4. Bake at 115 °C for 3 min.
5. Suss MJB4 Mask Aligner: for lamp powers 31.8 mW/cm² & 60.7 mW/cm², expose 1 sec (2.2 sec for Shipley 1818) with constant power (CP) mode and hard contact. The exposure time needs to be adjusted according to the power of the machine. Use a sacrificial chip to test.
7. Wash in DI water and blow dry.

**Mesa etching**

1. Photolithography as outlined in photolithography recipe.
2. Measure initial chip profile.
3. Etch in $\text{H}_2\text{O}:\text{H}_2\text{O}_2:\text{H}_2\text{SO}_4 = 240:8:1$.
4. Immediately after etch, soak in DI water for 30 min and blow dry.
5. Measure chip profile again to obtain etching rate. Repeat steps 2 and 4 until the donor layer is etched away (>80 nm below the chip surface).

**Ohmic contact**

1. Photolithography with thin gate requirements.
2. Plasma clean, 30 W for 30 sec with 30 sccm $\text{O}_2$.
3. Dip in $\text{H}_2\text{O}:\text{NH}_3\text{OH}=3:1$ for 5 sec. Rinse with DI water for 15 sec.
4. Transport immediately to thermal evaporator. Store in $\text{N}_2$ environment to prevent oxidation if cannot transport immediately.
5. Evaporate 50 Å Ni, 200 Å Au, 250 Å Ge, 100 Å Au, 50 Å Ni and 400 Å Au.
6. Lift off in acetone with ultrasound and squirt from acetone bottle if necessary. For delicate structures, leave in acetone at least an hour before resorting to ultrasound or leave in acetone overnight.
7. Rinse in methanol and blow dry.
8. Anneal at 420 °C for 50 sec.
Gates

1. Photolithography with thick gate requirements.
2. Dip in $\text{H}_2\text{O}:\text{NH}_4\text{OH}=3:1$ for 5 sec. Rinse with DI water for 15 sec.
3. Evaporate 80 Å Cr (or Ti) and 5000 Å Au.
4. Lift off and clean as in the ohmic contact steps.

Graphene Fabrication

Graphene Transfer

1. Graphene is grown via CVD on Cu foil in Jing Kong group (MIT). Graphene is grown on both side of the Cu foil. A protection layer of PMMA is spin coated on graphene on one side. Graphene on the other side is removed with $\text{O}_2$ plasma. Cut the PMMA/graphene/Cu into the desired shape and size.
2. Remove the Cu by floating the PMMA/graphene/Cu piece on Cu etchant surface with the Cu side down. Cu disappears after about 20 min.
3. Fish out the PMMA/graphene layer with a glass slide and transfer into 3 big beakers of DI water one after another to wash away the Cu etchant.
4. Transfer into $\text{HCl}:\text{DI water} = 1:10$ for 20 min to remove remaining Cu etchant.
5. Transfer into 3 big beakers of DI water one after another to wash away the HCl.
6. Fish out the PMMA/graphene layer finally using the SiO$_2$/Si or quartz substrate. Position the piece onto the center of the substrate.
7. Blow N\textsubscript{2} directly on top of the PMMA/graphene to squeeze out the water between graphene and the substrate, reducing wrinkles and keep graphene flat.
8. Dry in vacuum overnight.
9. Remove PMMA in acetone for 10-15 min or longer. Be gentle when putting the chip into the liquid as the PMMA/graphene layer may be torn apart.

**Graphene Patterning**

1. Photolithography as in the 2DEG case with Shipley 1805.
2. Etch areas uncovered by photoresist with 40 sccm O\textsubscript{2} plasma at 100 W for 60 sec. Repeat (usually three times) until one can visually observe the color change of photoresist as it gets thinner on SiO\textsubscript{2}/Si to ensure graphene is definitely removed.
3. Remove photoresist by leaving chip in acetone overnight. Avoid shaking.

An alternative photolithography step is to use the AS200 i-line Stepper instead of the Suss MJB4 Mask Aligner. The stepper gives a better resolution (~0.65 μm) and a precision with a 5X reduction exposure tool. The corresponding recipes are as follows:

1. Spin HMDS 4000 rmp, 1000 rmp/s for 45 s, no baking.
2. Spin resist SOR-700-0.1, 4000 rmp, 1000 rmp/s for 45 s. Back for 65 s at 115 °C.
3. Expose with AS200 i-line Stepper after confirming the exposure dose on a sacrificial substrate.
4. Post exposure bake for 65 s at 115 °C.
5. Develop with CD-26 for 1 min. Wash in DI water and blow dry.
Appendix B

Microwave Frequency 2DEG Simulations

The 2DEG interferometer circuit is simulated using the Sonnet frequency-domain field solver. The structures are simulated in a box with $x = 500 \, \mu m$, $y = 850 \, \mu m$ and $z = 1000 \, \mu m$. As indicated in Fig. B.1, the design schematics reflect the interferometer structure shown in Chapter 3.

![Figure B.1 Schematics](image_url)

**Figure B.1** Schematics of the 2DEG interferometer in Sonnet frequency-domain field solver. (a) A 3D view of the structure in the simulation box, consisting of two layers and three material blocks: vacuum, AlGaAs and GaAs. (b) Top view of layer 1 where the 2DEG, ohmic contacts and signal lines are situated.
The 2DEG and signal lines are drawn on layer 1 and the gate and ground lines are drawn above on layer 0. The two layers are separated by 80 nm with AlGaAs in between. There is vacuum above layer 0 and GaAs below layer 1. The materials are defined by their dielectric constants in the Dielectric Layers section. The signal lines are connected to the 2DEG regions by two ohmic contacts. The signal lines, ground lines of the CPW and the gate are all defined to be lossless, as a good approximation in the 1-50 GHz frequency range. The ohmic contacts are defined to have a resistance of 400 Ω/sq. (This is an estimation obtained from previous measurements in [6, 7] using the same materials. Although the exact value varies from device to device, the estimation is a well-educated one for design purposes.) The 2DEG is defined to have an inductance estimated using Eq. (2.1) and a resistance using Eq. (2.7). These values are defined in Metal Types.

A Linear Frequency Sweep simulation of the CPW characteristic impedance is carried out prior to any circuit design. A thru CPW is drawn in the same dielectric layers. The two signal ports both have a resistance of 50 Ω. The spacing between the signal and the ground lines is varied until the characteristic impedance shows 50 Ω, which is the characteristic impedance of the network analyzer, cables and probes. This spacing is then used in the interferometer structure shown in Fig. B.1.

The widths of the 2DEG region are chosen to match the 50-Ω characteristic impedance of the CPWs, and therefore the rest of the measurement system. A Liner Frequency Sweep is carried out from 1 to 51 GHz with a 2-GHz step. The response of the circuit is analyzed with the $s_{21}$ magnitude shown in Fig. B.2. The 2DEG parameters were adjusted until the $s$-parameters give satisfactory characteristics, such as those shown in Fig. B.2, with well-defined interference patterns.
The simulations also provide current distribution visualizations. Figure B.3 gives an example of the current distribution in the 2DEG regions.

**Figure B.2** Simulated $s_{21}$ parameter magnitude by Sonnet.

**Figure B.3** Current distribution at an arbitrary frequency.
Appendix C

Parasitic Signal De-embedding

The microwave scattering measurements are conducted in an Agilent E8364A two-port vector network analyzer with Cascade Microtech WinCal XE 4.2 software. As mentioned in Section 3.2, the delay and loss from the network analyzer up to the probe tips are calibrated out by using the NIST-style multi-line TRL calibration method [33]. Therefore, the s-parameters read by the software contain only signals from both the interferometer and the parasitic coupling between the CPWs bypassing the interferometer.

![Figure C.1 Schematics explaining the de-embedding method.](image)

To de-embed the parasitic signal, we measure first the raw s-parameter of the interferometer with the parasitic signal and the s-parameters of an open circuit. The open circuit is obtained by applying a negative effective gate bias which completely depletes the 2DEG. We convert the measured s-parameters to admittance matrix parameters (the Y-parameters). With the circuit models shown in Fig. C.1, the Y-parameters of the interferometer alone are calculated by directly subtracting the total Y-parameters with interferometer and the parasitic signals by the open circuit Y-parameter, i.e.

\[ Y(\text{Interferometer}) = Y(\text{Interferometer + parasitic}) - Y(\text{Open}). \]
The interferometer Y-parameter is then converted back to an s-parameter [36].

The conversions between the s- and Y-parameters are [36]:

\[
Y_{11} = Y_0 \frac{(1 - S_{11})(1 + S_{22}) + S_{12}S_{21}}{(1 + S_{11})(1 + S_{22}) - S_{12}S_{21}},
\]

\[
Y_{12} = Y_0 \frac{-2S_{12}}{(1 + S_{11})(1 + S_{22}) - S_{12}S_{21}},
\]

\[
Y_{21} = Y_0 \frac{-2S_{21}}{(1 + S_{11})(1 + S_{22}) - S_{12}S_{21}},
\]

\[
Y_{22} = Y_0 \frac{(1 + S_{11})(1 - S_{22}) + S_{12}S_{21}}{(1 + S_{11})(1 + S_{22}) - S_{12}S_{21}},
\]

where \( Y_0 \equiv 1/Z_0 \), with \( Z_0 = 50 \Omega \) being the characteristic impedance of the measurement environment.

And then

\[
S_{11} = \frac{(Y_0 - Y_{11})(Y_0 + Y_{22}) + Y_{12}Y_{21}}{(Y_0 + Y_{11})(Y_0 + Y_{22}) - Y_{12}Y_{21}},
\]

\[
S_{12} = \frac{-2Y_0Y_{12}}{(Y_0 + Y_{11})(Y_0 + Y_{22}) - Y_{12}Y_{21}},
\]

\[
S_{21} = \frac{-2Y_0Y_{21}}{(Y_0 + Y_{11})(Y_0 + Y_{22}) - Y_{12}Y_{21}},
\]

\[
S_{22} = \frac{(Y_0 + Y_{11})(Y_0 - Y_{22}) + Y_{12}Y_{21}}{(Y_0 + Y_{11})(Y_0 + Y_{22}) - Y_{12}Y_{21}}.
\]
Appendix D

Far-infrared Graphene Plasmonic Simulations

We use the COMSOL Multiphysics 3D Wave Optics Module to simulate graphene plasmonic responses in the far-infrared regime. For future reference, this section describes briefly the method and setup used.

Figure D.1 shows the graphic design of a unit cell of the system, using the hexagonal lattice of circular apertures on SiO₂/Si as an example. It consists of a stack of four blocks, each labeled with the corresponding materials and thicknesses. The bottom 4 μm Si block is set to be a Perfectly Matched Layer. The side walls are defined to have a Periodic Condition with k-vector for Floquet periodicity. For transmission simulations, the topmost boundary is defined as a Port inputting an electric field. Graphene is embedded at the interface of the vacuum and SiO₂ blocks, i.e. the x-y plane at z = 0 and outside the central circle. Graphene is not drawn as a physical layer. Because graphene is so much thinner than the other materials, the mesh would have to be extremely fine around the graphene region, significantly consuming computational power and slowing down the simulation. Rather, graphene is defined as a Transition Boundary Condition with conductivity given by Eq. 4.1 and a thickness of 0.5 nm.

To obtain the transmission through the graphene plasmonic crystal, a Frequency Domain study is conducted with a sweep of frequency over the range of interest. A Surface Integration at the Perfectly Matched Layer boundary in Si, i.e. the x-y plane at z = -4 μm, is use to give the transmitted power through the stack, T. Transition Boundary Condition is turned off (so there is
no graphene) to simulate background transmission through the substrate alone, $T_0$. Thus, the extinction spectra $1 - T/T_0$ are calculated post-simulation. A Parametric Sweep over a range of $\tau$ values can be added.

For band structure simulations, the Perfectly Matched Layer and the Port are not needed. In this case, an Eigenfrequency study is run to search for eigenfrequencies around each Brillouin Zone k-vector. The band structures are all simulated with a high $\tau = 5 \times 10^{-12}$ s.

**Figure D.1** A unit cell of the graphene plasmonic crystal setup in the COMSOL Multiphysics finite element simulation.
Bibliography


