Plasmonic Properties of Nanopatterned Graphene

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PLASMONIC PROPERTIES OF NANOPATTERNED GRAPHENE

by

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M.S. University of Central Florida, 2019

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ABSTRACT

Graphene is an atomically thin two-dimensional material with carbon atoms arranged in a honeycomb lattice. It has been successfully mechanically exfoliated from graphite and characterized in 2004 by Andre Geim and Konstantin Novoselov at the University of Manchester, UK. While suspended graphene holds the world record in mobility exceeding 100,000 cm²/V s, which means that graphene could increase the operating frequency of electronic devices up to the THz regime. For example, imaging computer chips running at a few THz frequency, which would be about 1000 times faster than current chips. However, there are two main obstacles for using graphene in electronics and optoelectronics. Firstly, graphene has no band gap, which means that the current through graphene cannot be turned off by a gate voltage, hampering the creation of a graphene transistor. Secondly, graphene’s absorbance is only about \( \pi \alpha = 2.3\% \), where \( \alpha = e^2 / hc = 1/137 \) is the fine structure constant. In my thesis I focus on a unique method to increase the absorbance of single-layer graphene (SLG) and multi-layer graphene (MLG) to nearly 100%. This method relies on the creation of localized surface plasmons (LSPs) around holes patterned in SLG and MLG. These LSPs are able to absorb almost 100% of incident infrared (IR) light over a wide range of wavelengths from \( \lambda = 1.3 \, \mu m \) down to \( \lambda = 12 \, \mu m \) and beyond, covering the near-infrared (NIR), short-wavelength infrared (SWIR), mid-wavelength infrared (MWIR), and long-wavelength infrared (LWIR) regimes. Taking advantage of the high absorbance in nanopatterned graphene we developed the proof-of-concepts of IR photodetectors based on the photothermoelectric effect in nanopatterned graphene (NPG) and nanopatterned multilayer graphene intercalated with \( FeCl_3 \) (NPMLG-\( FeCl_3 \)), an IR photodetector based on the combined effect of phase transition from insulator to metal in \( VO_2 \) and NPG in a heterostructure consisting of NPG on top of \( VO_2 \), and a thermal IR emission source based on NPG. During my thesis I have studied several physical phenomena for developing these devices. Due to the 2D nature of graphene and its Dirac fermions, it is possible
to tune the LSP absorption and emission resonance wavelength by means of a gate voltage. This means all the proposed IR photodetectors and thermal IR sources can be tuned electrostatically. In the case of the IR photodetectors based on NPG and NPMLG-\(FeCl_3\) the detection of incident IR light relies on the plasmonically enhanced photothermoelectric effect due to asymmetric nanopatterning of NPG or NPMLG. For the thermal IR emission from NPG we generalized Planck’s theory to any grey body and derived the completely general nonlocal fluctuation-dissipation theorem with nonlocal response of surface plasmons in the random phase approximation (RPA). The nonlocality of the fluctuation-dissipation theorem allows for the description of the coherence in the excited LSPs and the thermally emitted photons. For the description of the devices we developed a multiscale modeling platform using various software packages for performing finite-difference time domain (FDTD) simulations, COMSOL solvers for the drift-diffusion and heat equations coupled by thermoelectric effects, density functional theory (DFT) calculations, and analytical formulas.
I dedicate this work to my beloved wife, Khadija, my parents and siblings for their unconditional love, support and encouragement.
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CHAPTER 1: INTRODUCTION

Light-matter interaction play a fundamental role in quantum processing, sensing, detection, and lasers. The impact of incident light on different materials varies based on size, shape, and intrinsic properties of the materials, like, bandgap, Fermi energy, carrier concentration and dimensionality etc. [34, 27, 47]. Modern engineering techniques have made it possible to control optical and electronic properties of materials by using different kinds of patterning at nano-scale [57]. Absorption in pristine graphene is limited to 2.3% only [81], but doped graphene with periodic nanostructure has shown potential for electrically tunable high absorption [105, 81], low losses, and significant wave localization. Excitation of plasmons generate hot carriers via Landau damping [45], which have wide potential in energy harvesting and photodetection devices.

Graphene plasmons

Plasmons are collective excitations of conduction electrons at the interface between metal and dielectric materials. With the advancement of experimental and computational techniques, the work of Ritchie [74], and Mie [60] have attracted researchers from all scientific fields to explore plasmons based phenomena and its wide range applications. Surface plasmons in Dirac materials show distinct properties due to the presence of massless Dirac fermions with linear dispersion relation around Dirac cone [71]. Graphene is a single layer sheet of carbon atoms arranged in a honeycomb lattice, as shown in Fig. 1.1. Because of its unique band structure, graphene has a very high carrier mobility and fast relaxation time [31, 30, 91], which makes it a promising material for ultrafast devices [28, 12]. Exciting plasmons leads to enhancement of absorption in graphene [82, 81, 29]. The surface plasmons couples the electromagnetic wave to the conducting medium, which results in a direct absorption in mono layer graphene, and provides an opportunity to tune
the resonance peak[86, 88, 82].

Figure 1.1: a) Honeycomb lattice structure of single layer graphene sheet, \(a_1\) and \(a_2\) are lattice vectors. (b) first Brillouin zone of graphene sheet with reciprocal lattice vectors \(b_1\) and \(b_2\) and symmetry points \(\Gamma\), \(M\) and \(K\). (c) Dirac cone at K point.

Using the linear dispersion relation, the intraband optical conductivity for graphene is[82, 68]

\[
\sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{2k_B T}{\tau - 1 - i\omega} \ln \left[ 2 \cosh \left( \frac{\varepsilon_F}{2k_B T} \right) \right],
\]

which in the case of \(\varepsilon_F \gg k_B T\) is reduced to

\[
\sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{\varepsilon_F}{\tau - 1 - i\omega} = \frac{2\varepsilon_m \omega_p^2}{\pi \hbar^2 (\tau - 1 - i\omega)},
\]

where \(\tau\) is determined by impurity scattering and electron-phonon interaction \(1/\tau = 1/\tau_{\text{imp}} + 1/\tau_{e-\text{ph}}\). Using the mobility \(\mu\) of the NPG sheet, it can be presented in the form \(1/\tau = e \nu_F^2 / (\mu E_F)\), where \(\nu_F = 10^6\) m/s is the Fermi velocity in graphene. \(\omega_p = \sqrt{e^2 \varepsilon_F / 2\varepsilon_m}\) is the bulk graphene plasma frequency.

Due to the dominance of Coulomb interaction collision rate, hydrodynamic effects play an important role in graphene, i.e. \(1/\tau_{ee} \gg 1/\tau_{\text{imp}}\) and \(1/\tau_{ee} \gg 1/\tau_{e-\text{ph}}\), which corresponds to the hydrodynamic regime. \(1/\tau_{\text{imp}}\) and \(1/\tau_{e-\text{ph}}\) are the electron-impurity and electron-phonon collision rates. To achieve high absorbance and emittance, we choose a large Fermi energy, which takes us
in the Fermi liquid regime of the graphene sheet. Taking the hydrodynamic correction into account, the hydrodynamically adjusted intraband optical conductivity is,[6, 21]

$$\sigma_{\text{HD, intra}}(\omega) = \frac{\sigma_{\text{intra}}(\omega)}{1 - \eta^2 k^2/\omega^2},$$  \hspace{1cm} (1.3)

where $\eta^2 = \beta^2 + D^2\omega(\gamma + i\omega)$, $\beta^2 \approx \frac{3}{4}v_F^2$ is the intraband pressure velocity, $D \approx 0.4 \mu m$ is the diffusion length in graphene, and $\gamma = 1/\tau$ is the relaxation rate. The dielectric function for graphene is given by[82, 68]

$$\varepsilon_{\|}(\omega) = \varepsilon_g + \frac{i\sigma_{2D}(\omega)}{\varepsilon_0\omega d},$$  \hspace{1cm} (1.4)

where $\varepsilon_g = 2.5$ is the dielectric constant of graphite and $d$ is the thickness of graphene. For the case of graphene surrounded by dielectric mediums with permittivities $\varepsilon_1, \varepsilon_2$ dispersion relation for TM mode is,

$$\sqrt{\frac{\varepsilon_1}{q^2 - \frac{\varepsilon_1\omega^2}{c^2}}} + \sqrt{\frac{\varepsilon_2}{q^2 - \frac{\varepsilon_2\omega^2}{c^2}}} = -\frac{\sigma_{2D}(\omega, q) i}{\omega \varepsilon_0}$$

which can be presented, for $q = \omega/c$ case, in this form,

$$q \approx \frac{\varepsilon_1 + \varepsilon_2}{2} \frac{2i\omega}{\sigma(\omega, q)}$$

Charge carriers in graphene are bound in a normal direction, which leads to more confined electromagnetic field. Furthermore, localized surface plasmons decay through Landau damping into electron-hole pairs, which thermalize and thus transfer energy through electron-phonon interaction to the lattice as heat. This heat process occurs at a time scale of $\sim 100$ fs, while cooling happens much slower on a time scale of $\sim$10 ps [92, 13].
Aim of this work

In this work, we study, theoretically, the properties of plasmonics in nanopatterned graphene sheet, and their applications as light absorbers, photodetectors and thermal emitters in mid wavelength infrared (MWIR) and long wavelength infrared (LWIR) regimes. In Chapter 2, we demonstrate an efficient way to convert light signal in MWIR region into electrical signal with a very high responsivity. By patterning a graphene sheet with nano sized holes we enhance the absorption in single layer of graphene sheet from 2.3% to 60%. Nano sized holes introduce extra edges in graphene sheet, which play an important role in enhancement of light absorption. Recent studies have shown multiple ways to enhance absorption, but conversion of light signal into significant electrical signal has suffered due to absence of the bandgap in graphene sheet.

We demonstrate a new phenomenon for room-temperature infrared (IR) photodetection by generating Dirac-plasmons assisted hot carriers in graphene sheet. We introduce patterning in half area of graphene sheet to create asymmetric electronic environment. After the mid-IR photons get absorbed around the edges of the holes at a wavelength that matches the localized surface plasmons resonance, hot carriers are generated, which travel to the colder end of the sheet. Source drain voltage provides extra driving force to these hot carriers by creating differential Joule heating. Patterning leads to difference in intrinsic properties of graphene sheet, such as mobility, electrical conductivity, thermal conductivity and seebeck coefficient are lower for patterned graphene. All these factors play a role to create an effective temperature gradient between source and drain. Additionally, our design ensures gate voltage tunability upto $2\mu m$. Our device exhibits a very high responsivity of $\sim 3000 \, V/W$, detectivity $D^* \sim 10^9 \, Jones$ with a time response of $\sim 100 \, ns$ in the wavelength regime 8 - 12 $\mu m$.

In Chapter 3, We present a proof of concept for a spectrally selective thermal mid-IR source based on nanopatterned graphene (NPG) with a typical mobility of CVD-grown graphene (up to 3000
cm²V⁻¹s⁻¹), ensuring scalability to large areas. For that, we solve the electrostatic problem of a conducting hyperboloid with an elliptical wormhole in the presence of an *in-plane* electric field. The localized surface plasmons (LSPs) on the NPG sheet allow for the control and tuning of the thermal emission spectrum in the wavelength regime from λ = 3 µm to 12 µm by adjusting the size of and distance between the circular holes in a hexagonal or square lattice structure. Most importantly, the LSPs along with an optical cavity increase the emittance of graphene from about 2.3% for pristine graphene to 80% for NPG, thereby outperforming state-of-the-art pristine graphene light sources operating in the near-infrared (NIR) by at least a factor of 100. According to our COMSOL calculations, a maximum emission power per area of 11 × 10³ W/m² at T = 2000 K for a bias voltage of V = 23 V is achieved by controlling the temperature of the hot electrons through the Joule heating.

By generalizing Planck’s theory to any grey body and deriving the completely general nonlocal fluctuation-dissipation theorem with nonlocal response of surface plasmons in the random phase approximation (RPA), we show that the coherence length of the graphene plasmons and the thermally emitted photons can be as large as 13 µm and 150 µm, respectively, providing the opportunity to create phased arrays made of nanoantennas represented by the holes in NPG. The spatial phase variation of the coherence allows for beamsteering of the thermal emission in the range between 12° and 80° by tuning the Fermi energy between ε₉ = 1.0 eV and ε₉ = 0.25 eV through the gate voltage. Our analysis of the nonlocal hydrodynamic response leads to the conjecture that the diffusion length and viscosity in graphene are frequency-dependent. Using finite-difference time domain (FDTD) calculations, coupled mode theory, and RPA, we develop the model of a mid-IR light source based on NPG.

In Chapter 4, we present the model of an ultrasensitive mid-infrared (mid-IR) photodetector operating in the mid-wavelength infrared (MWIR) and long-wavelength infrared (LWIR) domains consisting of a hybrid heterostructure made of nanopatterned graphene (NPG) and vanadium diox-
ide (VO$_2$) which exhibits a large responsivity of $R \sim 10^4$ V/W, a detectivity exceeding $D^* \sim 10^{10}$ Jones, and a sensitivity in terms of noise-equivalent power $\text{NEP} \sim 100$ fW/√Hz close to room temperature by taking advantage of the phase change of a thin VO$_2$ film. Our proposed photodetector can reach an absorption of nearly 100% in monolayer graphene due to localized surface plasmons (LSPs) around the patterned circular holes. The geometry of the nanopattern and an electrostatic gate potential can be used to tune the absorption peak in the mid-IR regime between 3 and 12 µm. After the photon absorption by the NPG sheet and the resulting phase change of VO$_2$ from insulating to metallic phase the applied bias voltage $V_b$ triggers a current through the VO$_2$ sheet, which can be detected electronically in about 1 ms, shorter than the detection times of current VO$_2$ bolometers. Our envisioned mid-IR photodetector reaches detectivities of cryogenically cooled HgCdTe photodetectors and sensitivities larger than VO$_2$ microbolometers while operating close to room temperature.

In Chapter 5, We present a proof of concept for a spectrally selective near-IR photodetector based on nanopatterned multilayer graphene (NPMLG) intercalated with FeCl$_3$, enabling large modulation p-doping of NPMLG. The localized surface plasmons (LSPs) on the NPMLG sheets allow for electrostatic tuning of the photodetection in the near-infrared (NIR) and short-wavelength infrared (SWIR) regimes from $\lambda = 1.3$ µm to 3 µm, which is out of range for nanopatterned single layer graphene (NPG). Most importantly, the LSPs along with an optical cavity increase the absorbance of MLG from about 2% for pristine graphene to nearly 100% for NPMLG. Our NIR and SWIR detection scheme relies on the photo-thermoelectric effect induced by asymmetric patterning of the MLG sheets. The LSPs on the nanopatterned side create hot carriers that give rise to Seebeck photodetection at room temperature achieving a responsivity of $= 6300$ V/W, a detectivity of $D^* = 2.3 \times 10^9$ Jones, and an ultrafast response time of the order of 100 ns. Our theoretical results pave the way to graphene-based optical IR communication, IR color displays, and IR spectroscopy. We conclude the dissertation in Chapter 6.
CHAPTER 2: PHOTODETECTION BASED ON DIRAC PLASOMONS ASSISTED THERMAL CARRIERS IN GRAPHENE

Due to low photon energy, detection of infrared photons at room temperature is very challenging. Thermoelectric effect provides an alternate method overcoming the restriction of bandgap in materials. In this chapter, we present a scheme for photodetection using plasmons assisted thermal carriers at room temperature in an asymmetric electronic environment with an extraordinary high responsivity, detectivity and ultrafast time response.

Introduction

Two-dimensional materials have shown a great potential for the detection of photons in infrared regime, especially graphene [106]. An ultrafast detection of infrared photons is based on exitation of thermal carriers in absence of electron-phonon scattering [94, 39, 92] and examining the change in electron temperature due to thermoelectric effect [14, 108, 109]. Plasmonic excitations compress the energy of electromagnetic waves into sub-wavelength volumes, i.e. they are responsible for the high confinement of light, which increases strongly the light absorption in photodetectors [4]. Plasmons decay through Landau damping into electron-hole pairs, which thermalize and thus transfer energy through electron-phonon interaction to the lattice as heat, thereby generating a thermoelectric voltage $\Delta V$. Thermoelectric devices contain materials which convert a temperature difference to a potential difference, or vice versa, via the Seebeck effect or the Peltier effect, respectively, i.e.

$$\Delta V = S\Delta T \quad and \quad \Pi = ST$$

The thermoelectric voltage $\Delta V$ depends on the Seebeck coefficient $S$ and the temperature difference $\Delta T$. Seebeck coefficient is a material related property, and can be altered to some degree [99]. $\Delta T$ can be enhanced by multiple factors, e.g., high absorption of light, low energy loss in hot region, and low emissivity in hot region and high emissivity in cold region. Low thermal conductivity also leads to higher temperature gradient [50, 49, 33]. Since seebeck coefficient plays an important role for thermoelectric voltage, we will first discuss seebeck coefficient for graphene.

Seebeck coefficient for graphene

From linear response theory and using the Boltzmann transport equation, thermal and electrical current densities in terms of electric field and temperature gradient can be written as [44]

$$
\begin{pmatrix}
\vec{j}_q \\
\vec{j}
\end{pmatrix} = L
\begin{pmatrix}
\vec{E} \\
-\vec{\nabla}T
\end{pmatrix}
$$

(2.2)

where

$$
L =
\begin{pmatrix}
L_{11} & L_{12} \\
L_{11} & L_{12}
\end{pmatrix}
= \begin{pmatrix}
K^{(0)} & \frac{1}{eT} K^{(1)} \\
\frac{1}{e} K^{(1)} & \frac{1}{e^2 T} K^{(2)}
\end{pmatrix}
$$

and

$$
K^{(\alpha)} = \int_{-\infty}^{+\infty} (\varepsilon - \mu)^{\alpha} \sigma(\varepsilon) \left( -\frac{\partial f^{(0)}}{\partial \varepsilon} \right) d\varepsilon
$$

(2.3)

Here, $\mu$ is chemical potential, $\varepsilon = \hbar v_F k$, and $f^{(0)}$ is the Fermi Dirac distribution function at equilibrium. Seebeck coefficient, Peltier coefficient, electrical and thermal conductivities are defined from matrix elements as [44]:

$$
S = \frac{L_{12}}{L_{11}} \quad , \quad \Pi = \frac{L_{21}}{L_{11}} \quad , \quad \sigma = L_{11} \quad , \quad \kappa = L_{22}
$$
Thus, using equation (2.3) we can write the Seebeck coefficient as

$$ S = -\frac{1}{eT} \int_{-\infty}^{+\infty} (\varepsilon - \mu) \sigma(\varepsilon) \left( -\frac{\partial f(0)}{\partial \varepsilon} \right) d\varepsilon $$

The Seebeck coefficient for graphene depends on the energy dependent differential conductivity, which is proportional to the density of states $D(\varepsilon)$ and the relaxation time $\tau(\varepsilon)$, which can be calculated as

$$ \frac{1}{\tau(\varepsilon_k)} = \frac{\pi n_i}{\hbar} \left| \int k | u_i(q) \varepsilon(q, T) |^2 (1 + \cos \theta)(1 - \cos \theta) \delta(\varepsilon_k - \varepsilon_{k'}) \right|, $$

where $\int k = \int \frac{d^2k}{(2\pi)^2}$.

The differential conductivity is $\sigma(\varepsilon) = e^2 v_F^2 D(\varepsilon) \frac{\tau(\varepsilon)}{2}$, and the RPA screening function $\varepsilon(q, T) = 1 + \bar{u} D_0 \Gamma(q, T)$, where $D_0$ is density of states at zero Fermi energy $\epsilon_F = 0$. $\Gamma(q, T)$ is the irreducible polarization function. In degenerately doped systems, equation (2.4) can be modified to

$$ S = \frac{\pi^2 k_B^2 T}{3e} \left( \frac{d}{d\varepsilon} \ln (\sigma(\varepsilon)) \right)_{\varepsilon = \varepsilon_F} $$

From equation (2.7), we can see that the factors enhancing the energy dependent differential of conductivity can lead to enhancement in the Seebeck coefficient. One example is the energy dependent density $n(\varepsilon)$, which is directly proportional to the density of states. 2D materials have a higher density of states as compared to 3D materials, which affects the Seebeck coefficient. For graphene on a substrate, screening lengths and ratio of densities are the fitting parameter. Fig. 2.1 shows the plot of the Seebeck coefficient as a function of Fermi energy at room temperature. Across the charge neutral point (CNP), the sign of the Seebeck coefficient is opposite due to the opposite charges of electrons and holes. It has two peaks close to CNP, but decreases by increasing the absolute value of the gate voltage as $1/\sqrt{\epsilon_F}$ [98]. At the CNP the electron and hole Seebeck effects cancel each other. Furthermore, the Seebeck coefficient is dependent on the temperature,
and it increases with temperature. Duan et al. [24] have reported experimental results for temperature dependent $S(T)$ for graphene/hBN and graphene/SiO$_2$, which shows an increase in $S(T)$ with temperature, but it does not follow the linear trend expected by Mott’s equation [35]

Photo-Thermoelectric Effect in graphene

Monolayer pristine graphene has a very low light absorption ($< 3\%$) in the visible and the near-IR spectrum, but there are several approaches to enhance light absorption in graphene. Alireza et al., [81] have reported a 45% absorption in a patterned graphene coupled to an optical cavity in the $8 - 12 \mu m$ infrared band using holes arranged in square lattice. In this work, we have enhanced absorption from 45% to 60% by arranging holes in hexagonal lattice. Since the packing fraction of
hexagonal lattice in 2D is higher than square lattice, the total length of the edges in graphene sheet is increased for the case of a hexagonal arrangement of holes, thus enhancing plasmons excitations. In next chapters, we have even achieved perfect absorption in a graphene sheet. The main reason of low absorption here is that silicon is used for doping and it blocks almost 30% incoming light. Before modeling a full detector geometry, we studied behavior of photothermal electrical voltage in patterned graphene sheet only without optical cavity. Fig. 2.2 show two graphene sheet with square and hexagonal patterning, and respective photothermoelectric voltage $V_{PTE}$ is shown in Fig. 2.3.

Half area of the graphene sheet is patterned with a square or a hexagonal lattice of etched circles. The radius of the etched circles is 200 nm and the period of both hexagonal and square lattices is 600 nm. The incident power of the light beam, used in simulations, is 155 nW in form of a rectangular pulse, which has a period of 4 ms. Absorption of light around the etched holes excites localized edge plasmons, which in turn generate hot carriers in the graphene sheet. The hot carriers travel form the high temperature region to low temperature regions, which generates the Seebeck
voltage $\Delta V$ in the graphene sheet. As we can see from Fig. 2.3 (a) the patterned side of the sheet is at high temperature and unpatterned side is at low temperature. Figure 2.3 (b) and (c) shows the voltage produced in the graphene sheet due to the photo-thermoelectric effect: the left end of sheet is fixed at 0 V (ground), and the Seebeck voltage $\Delta V$ is measured at right end. The maximum voltage measured for hexagonal patterning is 2.9 $\mu$V and 1.6 $\mu$ V for square patterning. Higher absorption in hexagonal patterning leads to higher thermoelectric voltage.

We also checked the dependence of $\Delta V$ on the patterning area, which is plotted in Fig. 2.4. The photo-thermoelectric voltage strongly depends on the ratio of patterned and unpatterned graphene area. The maximum $\Delta V$ is produced when this ratio is approximately between 50% and 60%. When the patterned area is small, absorption of light is also low, which means less hot carries are generated, and they have to travel long distance to reach the collector. In result, we get a significantly lower value of the thermoelectric voltage. But, for higher ratio, more thermal carriers are produced, which travel quickly to cooler regions, thereby decreasing the temperature gradient, which also decreases the Seebeck voltage $\Delta V$. Our results show that maximum seebeck voltage is
generated only for a balanced combination of patterned and unpatterned area of graphene sheet.

Schematic design of the detector

The geometry of the photodetector is shown in Fig. 2.4. The length and width of the detector are $20 \mu m$ and $200 \mu m$, respectively. Graphene is on top of aluminum oxide and gated silicon, $SU-8$ of thickness $\lambda_{res}/4n$ is used for a spacer, where $n$ is refractive index of $SU-8$ and a gold mirror of thickness $200 \text{ nm}$. $5 \mu m$ wide and $50 \text{ nm}$ thick Gold is used as metallic contacts for source and drain voltages. Thus, the channel of the detector is $10 \mu m$ wide and $200\mu m$ long. The half width of the channel, i.e. $5 \mu m$, is patterned graphene. By doing so, an asymmetry is introduced in the system, which assists infrared detection. Another source of asymmetry is gold contacts used for applied bias. The detector takes advantage of the phenomena discussed above for 2D graphene, i.e. patterning the graphene enhances absorption due to excitation of plasmons associated with intraband transitions [83, 80]. As a result, hot carriers are generated, which travel towards colder end following the gradient introduced in carried density between source and drain contacts. The length of channel, between gold contacts, is $10 \mu m$, which is comparable to diffusion length of charge carriers [82, 83, 86].

Finite Element Method simulations

In this section, we discuss the results of the simulations of an ultrafast, uncooled long wavelength infrared photodetector based on monolayer p-doped graphene. We use COMSOL Multiphysics 5.3a software to simulate detector. The overall goal of simulations is to find the time dependent solution for bias dependent photothermoelectric current, which is further used to calculate the photothermoelectric voltage $V_{PTE}$ and the responsivity $V_{PTE}/P_{inc}$, where $P_{inc}$ is the power of the
Figure 2.4: Schematic showing our proposed tunable ultrafast mid-IR photodetector.

Incident electromagnetic field. We use the built-in modules “Electric Currents” and “Heat Transfer in Solids” coupled with the multiphysics module “Thermoelectric Effect.” The equations solved in these modules are:

\[
\mathbf{E} = -\nabla V
\]
\[
\nabla \cdot \mathbf{J} = 0
\]
\[
\mathbf{J} = \sigma \mathbf{E} + \frac{\partial \mathbf{D}}{\partial t} + \mathbf{J}_e
\]
\[
\rho c_p \frac{\partial T}{\partial t} + \rho c_p \mathbf{u} \cdot \nabla T - \nabla \cdot \mathbf{q} = Q
\]  
\text{(2.7)}
\]
\[
\Pi = ST
\]
\[
\mathbf{q} = \Pi \mathbf{J}
\]
\[
\mathbf{J}_e = -\sigma S \nabla T
\]
where \( J \) is the current density, \( E \) is the electric field, \( V \) is the electric potential, \( D \) is the displacement field, \( T \) is the temperature, \( S \) is the Seebeck coefficient, and \( \Pi \) is the Peltier coefficient. \( J_e \) is an external current density, contributed by the generated hot electrons. The other constants represent the electrical conductivity, the mass density, the specific heat capacity at constant pressure, and the thermal conductivity, respectively. Gold, Silicon, and Aluminum oxide materials are directly imported from COMSOL material library, while we use the experimentally measured parameters for graphene. The Seebeck coefficient is calculated using Mott’s approximation, given in eq. 2.7. The electrical conductivity and Seebeck coefficient are gate dependent for graphene, and low electrical conductivity and Seebeck coefficient are measured experimentally for patterned graphene as compared to unpatterned graphene. For example, at Fermi energy \( E_F = -1.0 \ eV \), \( \sigma = 1.58 \times 10^4 \ S/m \) and \( S = 90 \ \mu V/K \) for unpatterned graphene, and \( \sigma = 0.46 \times 10^4 \ S/m \) and \( S = 69 \ \mu V/K \) for patterned graphene. The heat capacity of graphene at room temperature is set to \( c_p = 700 \ J/(kg - K) \). We use the temperature independent electrical conductivity for all materials because we do not consider bolometric effects.

The bias voltage is applied across the gold terminals; one side is set to ground, and the other at high potential. Except gold terminals, everything is electrically insulated, the current conservation boundary condition is applied for the whole geometry, and the initial values are set to \( V = 0 \). Electrical contacts are introduced between gold and graphene in order to add contact resistances. In the heat transfer module, the unpatterned graphene side and the gold electrodes are thermally insulated. The heat flux is applied in form of rectangular pulse of period 4 \( ms \), which means, for the first two milliseconds there is zero heat flux, corresponding to the dark state. For the next two milliseconds nonzero heat flux is applied on the patterned side of graphene using laser heating. We use the Gaussian beam with spot size, \( R_{\text{spot}} = 2 \ mm \), and incident power \( P_{\text{inc}} = 155 \ nW \).
Figure 2.5: Photothermoelectric $V_{PTE}$ as a function of Fermi energy for different bias voltages $V_{SD}$.

The heat flux is

$$q_0 = \frac{2P_{inc}}{\pi R_{spot}^2} \exp\left(-\frac{2R_{focus}^2}{R_{spot}^2}\right)$$  \hspace{1cm} (2.8)$$

The absorbed heat flux depends on the absorption at different Fermi levels. The gate dependence of the light absorption is calculated using Lumerical FDTD software, which ranges from $A = 34\%$ at $EF = -0.55 \ eV$ to $A = 60\%$ at $EF = -1.0 \ eV$ for patterned graphene. The absorbed heat flux is $q = Aq_0$. The bottom side of the detector is kept at fixed temperature using the boundary condition “temperature” in the software. The initial value of the temperature is set to
\[ T_0 = 293.15 \ K \], and the boundary condition “open boundary” was used across all the sides of the detector, except top and bottom surfaces, which means heat can flow inside or outside across the cross-sectional boundary depending on the ambient temperature. Thermal contacts are used between graphene, aluminum oxide, and silicon to control heat transfer in the vertical direction. We use a user-controlled mesh, namely the free tetrahedral mesh for gold, the free triangular mesh at the graphene surface, which were swept in vertical direction for the remaining geometry. The time dependent solver with very low relative tolerance of \(10^{-5}\) is used to measure the time dependent thermoelectric current passing through the terminal for different fermi levels. The dark and light thermoelectric current \(I_{TE,D}\) and \(I_{TE,L}\) were measured in absence and presence of the incident heat flux, respectively. The photothermoelectric \(I_{PTE}\) current is then calculated by subtracting the dark from the light current. The bias voltage also generates thermoelectric voltage due to the Peltier effect. To differentiate between the plasmonically driven and the bias driven thermoelectric voltage, we have modeled the current flowing through the graphene channel for opposite bias voltages. The total current flowing through the channel for opposite polarities of \(V_{SD}\) is the sum of the bias current \(I_{SD}\) and thermoelectric current, i.e.

\[
I_1 = I_{SD} + I_{TE}\quad \text{and}\quad I_2 = -I_{SD} + I_{TE}
\]

(2.9)

The direction of the thermoelectric current is independent of the sign of \(V_{SD}\), as it flows only from the hot towards the cold region. Thus \(I_{TE} = \frac{I_1 + I_2}{2}\), and the difference of \(I_{TE}\) between the illuminated and non-illuminated graphene sheet is the photothermoelectric current: \(I_{PTE} = I_{TE,L} - I_{TE,D}\).
Photoresponse of the detector

The photothermoelectric voltage is calculated by multiplying $I_{PTE}$ with the combined resistance of patterned and unpatterned graphene sheets: $V_{PTE} = I_{PTE}R_G$, where $R_G$ is resistance of graphene. Fig. 2.5 shows the photothermoelectric voltage for different bias voltages. Our results show that higher bias leads to higher photothermoelectric voltage. We can infer that the bias voltage increases the drift of the hot carriers in the patterned region, giving rise to high responsivity. The maximum bias voltage is limited by the breakdown carrier density of graphene $\approx 12$ A/cm$^2$, therefore 0.9V was selected as an upper limit. In Fig. 2.6, we show comparison of experimentally measured and simulated $V_{PTE}$, which shows a nice agreement. The temperature and potential profiles across channel length are plotted in Fig. 2.7, highest value of $V_{PTE}$ measured was 0.44 mV for bias voltage of $V_{SD} = 0.9V$, which corresponds to temperature gradient $\Delta T = 4.7$ K.
Figure 2.7: Temperature and potential profile on graphene surface. a) and b) show temperature distribution across the channel length between gold contacts, while c) and d) show potential distribution.

The responsivity is calculated by dividing a photothermoelectric voltage with incident power. $R = \frac{V_{PTE}}{P_{inc}}$. Fig.2.7 a) shows plot of responsivity for a constant applied bias, and it goes to maximum value at Fermi level $0.8\ eV$ and then start decreasing. We have achieved a very high responsivity of $\approx 3000\ V/W$, which is much higher than so far reported values for graphene [14]. To study the effect of ambient temperature on responsivity, we simulate the device from $70 - 320\ K$, as shown in Fig.2.7 b). As temperature is decreased, responsivity vanishes, which is opposite to the case of bolometers, where it is enhanced upon cooling [113, 96, 25]. Thermal carrier generation at low temperatures is unaffected, as absorption remains same for this temperature range, but Seebeck coefficient is temperature dependent, which affects the responsivity at low
temperatures. This gives another proof that our detection process is purely based on thermoelectric effect.

Noise equivalent power and detectivity

The sensitivity of the photodetector is determined by the noise-equivalent power NEP, being an important figure of merit for the performance of a photodetector. The NEP of a photodetector provides a measure for the minimum detectable power per 1 Hz of bandwidth. The formula for NEP reads [69]

\[
\text{NEP} = \frac{\nu_n}{R},
\]

where \(\nu_n = \sqrt{\nu_t^2 + \nu_b^2 + \nu_f^2}\) is the root-mean-square of the spectral noise density, which consists of the sum over all possible noise voltages, such as the thermal Johnson-Nyquist noise \(\nu_t\), due to thermal motion of the charge carriers and independent of the bias voltage \(V_b\), the shot noise \(\nu_b\), due to the discrete nature of uncorrelated charge carriers, and the 1/f noise \(\nu_f\), also called flicker noise, due to random resistance fluctuations. Based on experimentally measured spectral noise density, \(\sim 12nV/\sqrt{Hz}\) at room temperature, we calculate \(\text{NEP} \sim 7pW/\sqrt{Hz}\) for applied bias of 0.5V. Detectivity, \(D^*\) is calculated by means of formula, \(D^* = \sqrt{A/\text{NEP}}\), where \(A\) is active area of detector. The highest value of detectivity is found to be \(1.1 \times 10^9 \text{ Jones}\) at Fermi energy 0.8 eV and \(V_{SD} = 0.9 \text{ V}\). Time response is measured by A.C. response of the detector, and it is found to be 100 ns, which validates our claim for an ultrafast detection process.

Hydrodynamic equations for graphene

Hydrodynamics explain a great number of phenomena, for example, flow of fluids, aircraft motion, atmospheric phenomena, blood flow in human beings and animals. It has been recognized
Figure 2.8: Responsivity of the detector, a) as a function of Fermi energy for a bias voltage $V_{SD} = 0.5 \ V$, b) for different substrate temperature at Fermi energy $-0.8 \ eV$ and $V_{SD} = 0.5 \ V$, c) as a function of bias voltage $V_{SD}$ at Fermi energy $-0.8 \ eV$.

that, under certain conditions, electrons in conductors also follow the equations of hydrodynamics. The hydrodynamic theory relies on the condition that electron-electron scattering is very high as compared to electron-phonon and electron-impurity scattering rates. Graphene is considered an excellent material for hydrodynamic flow of electrons [5, 65, 62], because electron-phonon scatterings on suitable substrates are weak, which leads to very high carrier mobilities at moderately high temperatures. Furthermore, the high quality of graphene fabrication minimizes impurity
scattering.

Starting from semi-classical Boltzmann equation [52, 9], for 2D massless Dirac Fermions, MDFs.

\[
\frac{\partial}{\partial t} + \vec{v}_k \cdot \vec{\nabla}_r \pm \frac{1}{\hbar} \vec{\nabla}_r U_{\text{eff}}(\vec{r}, t) \cdot \vec{\nabla}_k f = \mathcal{I}_{\text{coll}}[f]
\]

(2.11)

where negative sign is for electrons, and positive sign for holes. \( \mathcal{I}_{\text{coll}} \) is collision integral, and function \( f = f(\vec{k}, \vec{r}, t) \) is the distribution function for finding probability of an electron or hole with momentum \( \hbar \vec{k} \) at position \( \vec{r} \) and time \( t \). \( U_{\text{eff}}(\vec{r}, t) \) is an effective potential energy of an electron. All external potentials, and electron distribution’s self consistent potential should be included in \( U_{\text{eff}} \). The velocity \( \vec{v}_k \) and wave vector \( \vec{k} \) are related as;

\[
\vec{v}_k = \vec{v}_F \frac{k}{\vec{k}}
\]

(2.12)

In the ordinary Schroedinger Equation \( \vec{v}_k \) is linear, \( \vec{v}_k = \frac{\hbar \vec{k}}{m} \), but, for MDFs case it is strongly nonlinear[61]. As Discussed earlier, theory of hydrodynamics depends on assumption that system is in local equilibrium w.r.t. e-e scatterings, which means we can choose distribution function in such a way that \( \mathcal{I}_{\text{coll}}[f] = 0 \) at any point in space and time. In view of conservation of particles, momentum, and energy, we may write distribution function [95] as

\[
f \left( \vec{k}, \vec{r}, t \right) = \left( 1 + e^{\beta(\vec{r}, t) [h v_F \vec{k} - \hbar \vec{u} \cdot \vec{k} - \mu(\vec{r}, t)]} \right)^{-1}
\]

(2.13)

where \( u(\vec{r}, t) \) is average velocity, and \( \mu \) is chemical potential. Local Macroscopic averages are defined as [52]

\[
\langle A(\vec{r}, t) \rangle = \frac{\sum_{\ell \in \{e,h\}} \int d\vec{k} \ A_k f_\ell \left( \vec{k}, \vec{r}, t \right)}{\sum_{\ell \in \{e,h\}} \int d\vec{k} \ f_\ell \left( \vec{k}, \vec{r}, t \right)}
\]

(2.14)
Taking average of Boltzmann equation 2.11

\[ \int d\vec{k} A_k \left[ \partial_t + v_k \cdot \hat{\nabla}_r = \frac{1}{\hbar} \left[ \hat{\nabla}_r U_{\text{eff}} (\vec{r}, t) \cdot \hat{\nabla}_k \right] \right] f (\vec{k}, \vec{r}, t) = 0 \]  (2.15)

\[ \partial_t \langle nA \rangle + \vec{\nabla}_r \cdot \langle n\vec{v}_k A \rangle - n \left( \vec{v}_k \cdot \left( \hat{\nabla}_r \langle A \rangle \right) \right) \pm n \left( \hat{\nabla}_r U_{\text{eff}} (r, t) \cdot \left( \hat{\nabla}_k \langle A \rangle \right) \right) = 0 \]  (2.16)

A set of hydrodynamic equations can be derived by using different values for \( A \); \( A = 1 \) for continuity equation, \( A = \hbar \vec{k} \) for Euler equation, and \( A = \hbar v_F \vec{k} \) for energy balance equation.

The particle density, macroscopic current, and energy density can be calculated from distribution function following standard procedure [52]

\[ n = N \int \frac{d^2 k}{(2\pi)^2} f (\vec{r}, \vec{k}, t) \]

\[ j = N \int \frac{d^2 k}{(2\pi)^2} \vec{v}_k f (\vec{r}, \vec{k}, t) \]

\[ \varepsilon = N \int \frac{d^2 k}{(2\pi)^2} \varepsilon_k f (\vec{r}, \vec{k}, t) \]

Where \( N \) is degeneracy and is equal to 4 for the case of graphene. The conduction and valence band densities give total charge and imbalance charge.

\[ n = n_c - n_v \quad \text{and} \quad n_I = n_c + n_v \]

Similarly current densities can be combined to get total and imbalance current.

\[ j = j_c - j_v \quad \text{and} \quad j_I = j_c + j_v \]
Following all above steps, we derive continuity equation for hydrodynamic densities.

\[
\frac{\partial}{\partial t} n + \frac{\partial}{\partial x_i} (n u_i) = \partial_t n + \vec{\nabla} \cdot \vec{j} = 0
\]  

(2.17)

and for imbalance charge

\[
\partial_t n_I + \vec{\nabla} \cdot \vec{j}_I = 0
\]

Continuity equation is similar to ordinary parabolic band for two dimensional electron gas 2DEG. But, Euler and energy balance equations are totally different than 2DEG for graphene MDFs.

Euler equation

\[
\frac{\partial}{\partial t} (n u_\alpha) + \frac{\partial}{\partial x_\beta} \Pi_{\alpha\beta} \pm n \frac{\partial}{\partial x_\alpha} U_{\text{eff}} = 0
\]  

(2.18)

where \(\Pi_{\alpha\beta}\) is stress tensor, simplifying equation (2.18)

\[
\frac{\partial}{\partial t} \vec{k}(\vec{r}, t) + \frac{1}{\hbar} \frac{1}{n} \vec{\nabla}_r P + [\vec{u} \cdot \vec{\nabla}_r] \vec{k}(\vec{r}, t) + \frac{1}{\hbar} n e - n_h \vec{\nabla}_r U_{\text{eff}} = 0
\]  

(2.19)

As discussed earlier that, for 2D MDFs case, velocity and wave vector does not obey linearity like Schrödinger equation, where \([\hbar \vec{k}(\vec{r}, t) = m \vec{v}(\vec{r}, t)]\), instead follows [78]

\[
\hbar \vec{k}(\vec{r}, t) = \frac{3P}{n} \frac{1}{1 - \frac{v^2}{v_F^2}} \vec{u}
\]  

(2.20)

where \(P = P(\vec{r}, t)\) is pressure and can be calculated from equation of state for 2D MDFs [78]. For limit \(\vec{v}(\vec{r}, t) << v_F\), we can simplify Euler equation in following form;

\[
\frac{\partial}{\partial t} \vec{u} + [\vec{u} \cdot \vec{\nabla}_r] \vec{u} + \frac{v_F^2}{3P} \vec{\nabla}_r P + \frac{\vec{u}}{P} \partial_t P + \frac{v_F^2}{3P} [n_e - n_h] \vec{\nabla}_r U_{\text{eff}} (r, t) = 0
\]  

(2.21)

Equation (2.21) is Navier Stokes equation for graphene derived under the condition that local aver-
age velocity is much smaller than Fermi velocity. Comparing with non relativistic hydrodynamics, we see there is an extra term proportional to $\partial_t P$, which has very small effect on low frequency.

Now, following similar procedure for energy balance equation, we can write as;

$$\frac{\partial}{\partial t}(n\varepsilon) + \hbar v_F^2 \vec{\nabla}_r.(n\vec{k}) - v_F \vec{\nabla}_r U_{eff} \int \frac{d^2k}{(2\pi)^2} \vec{\nabla}_k f = 0$$

using equation (2.20), and continuity equation, we simplify above equation for graphene case,

$$\frac{\partial}{\partial t}(n\varepsilon) + 3P\vec{\nabla}.\vec{u} + 3P\vec{u} \cdot \vec{\nabla}P + 2n\vec{u} \cdot \vec{\nabla} U_{eff} = 0 \quad (2.22)$$

Now, we include the effect of dissipation, then distribution fiction is

$$f = f^0 + \delta f$$

where $f^0$ is the distribution function at equilibrium, and $\delta f$ is non-equilibrium correction. and Boltzmann equation is

$$[\frac{\partial}{\partial t} + \vec{v}_k \cdot \vec{\nabla}_r + \frac{1}{\hbar} \vec{\nabla}_r U_{eff}(\vec{r}, t) \cdot \vec{\nabla}_k]f = \mathcal{I}_{coll}[f] + \frac{\delta f}{\tau}$$

Dissipative correction in currents, $\vec{j}$ and $\vec{j}_{I}$ leads to an additional term in continuity equation, because they are not conserved due to non zero contribution from collision integral. The resulting continuity equations are;

$$\frac{\partial}{\partial t} n + \vec{\nabla}.\vec{j} = \vec{\nabla}.\delta \vec{j} \quad \text{and} \quad \frac{\partial}{\partial t} n_{I} + \vec{\nabla}.\vec{j}_{I} = \vec{\nabla}.\delta \vec{j}_{I} \quad (2.23)$$

25
Non equilibrium correction to stress tensor $\delta \Pi_{\alpha\beta}$ adds viscous term in Navier Stokes equation.

$$\frac{3P}{v_F^2} \left( \frac{\partial}{\partial t} \vec{u} + \left[ \vec{u}, \nabla_r \right] \vec{u} \right) + \nabla_r P + \frac{3\vec{u}}{v_F^2} \partial_t P + v_F^2 \left[ n_e - n_h \right] \nabla_r U_{eff}(r,t) + \eta \nabla^2 \vec{u} = 0$$  \hspace{1cm} (2.24)

Equation (2.24) is generalized form of Navier Stokes equation for graphene. Equations (2.17, 2.19, 2.22, 2.23, 2.24) represent complete system of hydrodynamic equations for two dimensional graphene. Signatures of hydrodynamics behavior in graphene are, longitudinal conductivity in mono layer, viscosity and nonlocal transport, linear magnetoresistance, and thermal transport properties etc. Nonlinear hydrodynamic phenomena and viscous effects are hallmark of hydrodynamic flow in graphene.

Theory of hydrodynamics has been very successful in explaining behavior of classical gases and liquids, and it depends on conservation of mass, momentum, and energy. Some recent articles report that, hydrodynamics is applicable to strongly interacting quantum critical systems, including ultra cold Fermi gases, and ultra hot nuclear matter\cite{Jacak}, \cite{26}. Hydrodynamics can also be employed to explain many electrons phenomena in condensed matter. Hydrodynamics is applicable when electron-electron scattering length is the shortest in spatial scale of space.

$$a_{ee} \ll D, a$$

where $a_{ee}$ is scattering length of electrons, and $D$ is width of sample being studied, $a = v_F \tau$ is related to electron-phonon and electron-impurity scatterings. For small scattering lengths, motion of electrons can be studied by hydrodynamic equations.

$$\nabla \cdot \vec{j} = 0 \quad \text{and} \quad \vec{j} - D_\eta \nabla^2 \vec{j} + \sigma_0 \nabla \phi(r) = 0$$  \hspace{1cm} (2.25)

where $\vec{j} = j(\vec{r})$, and $\phi$ is electric potential in 2D plane. $D_\eta$ is a spacial scale dependent on shear
Figure 2.9: Plot of surface current density. (a) Hydrodynamic ohm’s law, for $D_\eta = 0.4 \mu m$, (b) Ohm’s law $D_\eta = 0$.

viscosity and $\tau$, $D_\eta = \sqrt{\eta \tau}$. For, $D_\eta = 0$, second equation in eq(2.7) becomes traditional Ohm’s law $\vec{j} = \sigma_0 \vec{E}$ with Drude like conductivity. The packages we are using for simulations doesn’t have any built-in module to observe effects of hydrodynamic ohm’s law on graphene samples. Comsol Mulitiphysics [64] allows to built new modules in package, using mathematical equations. We have simluted thermoelectric graphene sheet, discussed in previous section, including hydrodynamics effect on electron’s motion. Viscosity of electrons in graphene is reported with very high values of $0.1 \frac{m^2}{s} [5]$, even higher than honey. We have simulated graphene surface and observed sub micrometer whirlpools, which depends on spacial scale $D_\eta$. Figure ?? shows result of simulations with and without hydrodynamic effects, and found that electron’s motion in highly viscous graphene produces whirlpools. Bandurin et. al. [5] have recently reported experimental evidence for whirlpools for special geometry. We have found strong dependence of whirlpools on $D_\eta$, and disappear as we increase $D_\eta$ or decrease sample size. The effect of hydrodynamics term in graphene absorption is discussed in Chapter 3, and plotted in Figure 3.3.
Conclusion

In conclusion, this chapter shows schematic for infrared photodetection using nanopatterned single layer of graphene sheet. The detection process involves enhancement of tunable absorption in graphene sheet coupled to a cavity, engineering of carrier mobility, and an asymmetric electrical environment. The asymmetric environment created in channel enhanced temperature gradient to 4.7K, which leads to high photothermoelectric voltage. For a low incident power of 155 nW the merit of figures for our proposed detector outperform graphene based detectors in MWIR with a responsivity of 3000 V/W and detectivity of $10^9$ Jones, and ultrafast time response of 100 nanosecond. In last part, we have derived hydrodynamic equations for graphene, and observed the whirlpools in current density due to high viscosity of electrons. Our finite element simulations measure the limit of $D_\eta = 0.4\mu m$ for whirlpools.
CHAPTER 3: TUNABLE SPECTRALLY AND DIRECTIONAL SELECTIVE THERMAL EMISSION FROM NANOPATTERNED GRAPHENE

In this chapter, we present a proof of concept for a spectrally selective thermal mid-IR light source based on nanopatterned graphene (NPG)\(^1\).

Introduction

An object that is kept in equilibrium at a given temperature \( T > 0 \) K emits electromagnetic (EM) radiation because the charge carriers on the atomic and molecular scale oscillate due to their heat energy.[7] Planck’s law describes quantitatively the energy density \( u(\omega) \) of the EM radiation per unit frequency \( \omega \) for black-body radiation, which is \( u_{BB}(\omega)d\omega = \frac{\omega^2}{\pi^4 c^3} \Theta(\omega)d\omega \), where \( c \) is the speed of light in vacuum, \( \hbar \) is the Planck constant, and \( k_B \) is the Boltzmann constant. \( \Theta(\omega, T) = \frac{\hbar \omega}{\exp(\hbar \omega/k_B T) - 1} \) is the thermal energy of a photon mode. Consequently, the energy emitted per unit surface area and per unit frequency, also called spectral radiance, of a black body into three-dimensional (3D) space is given by

\[
I_{BB}(\omega)d\omega = \frac{1}{4\pi} cu(\omega) = \frac{\omega^2}{4\pi^3 c^2} \Theta(\omega)d\omega.
\]  

The total energy density \( u \) can then be obtained by integrating over all frequencies and angles over the half-sphere, leading to the Stefan-Boltzmann law for the energy density of black-body

\[^1\text{Plasmonically enhanced mid-IR light source based on tunable spectrally and directionally selective thermal emission from nanopatterned graphene, Muhammad Waqas Shabbir & Michael N. Leuenberger, Sci Rep 10, 17540 (2020). https://doi.org/10.1038/s41598-020-73582-3}\]
radiation,

\[ u_{BB} = \left( \frac{8\pi^5 k_B^4}{15c^3\hbar^3} \right) T^4 = a_{BB} T^4, \]  

\[ (3.2) \]

with \( a_{BB} = 7.566 \times 10^{-16} \text{ Jm}^{-3} \text{K}^{-4} \). The total power emitted per unit surface area \( P/A \) of a black-body is

\[
I_{BB} = \frac{P}{A} = \int_0^\infty I_{BB}(\omega) d\omega \int_0^{2\pi} d\varphi \int_0^{\pi/2} \cos \theta \sin \theta d\theta 
= \pi \int_0^\infty I_{BB}(\omega) d\omega = \frac{1}{4\pi} uc 
= \frac{a_{BB} c}{4\pi} T^4 = b_{BB} T^4 = \left( \frac{\pi^2 k_B^4}{60c^2\hbar^3} \right) T^4,
\]

\[ (3.3) \]

where \( b_{BB} = 5.67 \times 10^{-8} \text{ Wm}^{-2} \text{K}^{-4} \) is the Stefan-Boltzmann constant. The factor \( \cos \theta \) is due to the fact that black bodies are Lambertian radiators.

In recent years, several methods have been implemented for achieving a spectrally selective emittance, in particular narrowband emittance, which increases the coherence of the emitted photons. One possibility is to use a material that exhibits optical resonances due to the band structure or due to confinement of the charge carriers.[7] Another method is to use structural optical resonances to enhance and/or suppress the emittance. Recently, photonic crystal structures have been used to implement passive pass band filters that reflect the thermal emission at wavelengths that match the photonic bandgap.[22, 56] Alternatively, a truncated photonic crystal can be used to enhance the emittance at resonant frequencies.[18, 111]

Recent experiments have shown that it is possible to generate infrared (IR) emission by means of Joule heating created by means of a bias voltage applied to graphene on a SiO\(_2\)/Si substrate.[32, 59] In order to avoid the breakdown of the graphene sheet at around \( T = 700 \) K, the graphene sheet can be encapsulated between hexagonal boron nitride (h-BN) layers, which remove efficiently the
heat from graphene. The top layer protects it from oxidation.[48, 58] In this way, the graphene sheet can be heated up to $T = 1600 \, \text{K}$,[58] or even above $T = 2000 \, \text{K}$.[48, 87] Kim et al. and Luo et al. demonstrated broadband visible emission peaked around a wavelength of $\lambda = 725 \, \text{nm}$.[48, 58] By using a photonic crystal substrate made of Si, Shiue et al. demonstrated narrowband near-IR emission peaked at around $\lambda = 1600 \, \text{nm}$ with an emittance of around $\epsilon = 0.07$.[87] To the best of our knowledge, there are neither theoretical nor experimental studies on spectrally selective thermal emission from graphene in the mid-IR range.

Here, we present the proof of concept of a method to tune the spectrally selective thermal emission from nanopatterned graphene (NPG) by means of a gate voltage that varies the resonance wavelength of localized surface plasmons (LSPs) around the circular holes that are arranged in a hexagonal or square lattice pattern in a single graphene sheet in the wavelength regime between 3 $\mu \text{m}$ and 12 $\mu \text{m}$. By generalizing Planck’s radiation theory to grey-body emission, we show that the thermal emission spectrum can be tuned in or out of the two main atmospheric transparency windows of 3 to 5 $\mu \text{m}$ and 8 to 12 $\mu \text{m}$ in the mid-IR regime, and also in or out of the opaque mid-IR regime between 5 and 8 $\mu \text{m}$. In addition, the gate voltage can be used to tune the direction of the thermal emission due to the coherence between the localized surface plasmons (LSPs) around the holes due to the nonlocal response function in graphene, which we show by means of a nonlocal fluctuation-dissipation theorem. The main element of the nanostructure is a circular hole of diameter $a$ in a graphene sheet. Therefore let us focus first on the optoelectronic properties of a single hole.
The frequency-dependent dipole moment of the hole is

\[ p(r, \omega) = -\varepsilon_0 \varepsilon_{\parallel}(r, \omega)|E_0| = -\alpha_{1,2}(r, \omega)|E_0|, \]

(3.4)

where the polarizabilities \( \alpha_{1,2} \) are given along the main axes \( x \) and \( y \) of the elliptic hole, and \( r = r_0 \) is the position of the dipole moment, i.e. the hole. Graphene’s dielectric function is isotropic in the xy-plane, i.e. \( \varepsilon'' = \varepsilon''_{xx} = \varepsilon''_{yy} \). \( V_0 \) is the volume of the graphene sheet. In the Appendix A we derive the general polarizabilities of an uncharged single-sheet hyperboloid with dielectric function \( \varepsilon(\omega) \) inside a medium with dielectric constant \( \varepsilon_m \) [see Eq. (A.106)]. The polarizabilities of an elliptical
wormhole in $x$- and $y$-direction read

\[
\alpha_1(\omega) = \frac{2abd\pi(\pi/2 - 1)}{3} \frac{\varepsilon_{||}(\omega) - \varepsilon_m}{\varepsilon_m + L_1[\varepsilon_{||}(\omega) - \varepsilon_m]},
\]

\[
\alpha_2(\omega) = \frac{2abd\pi(\pi/2 - 1)}{3} \frac{\varepsilon_{||}(\omega) - \varepsilon_m}{\varepsilon_m + L_2[\varepsilon_{||}(\omega) - \varepsilon_m]},
\]

\[(3.5)\]

\[(3.6)\]

respectively, for which the in-plane polarizabilities lies in the plane of the graphene sheet that is parallel to the $xy$-plane. $a$ and $b$ are the length and the width of the elliptical wormhole, as shown in Fig. A.1 in the Appendix A. $\varepsilon_{||}(\omega)$ is the dielectric function of graphene. We assumed that the thickness $d$ of the graphene sheet is much smaller than the size of the elliptic hole. The geometrical factors in this limit are

\[
L_1 \approx abd \int_{-\infty}^{\infty} \frac{d\eta'}{(\eta' + a^2)R_{\eta'}},
\]

\[(3.7)\]

\[
L_2 \approx abd \int_{-\infty}^{\infty} \frac{d\eta'}{(\eta' + b^2)R_{\eta'}},
\]

\[(3.8)\]

In the case of a circular hole of diameter $a$ the polarizability simplifies to

\[
\alpha_{||}(\omega) = \frac{2a^2d\pi(\pi/2 - 1)}{3} \frac{\varepsilon_{||}(\omega) - \varepsilon_m}{\varepsilon_m + L_{||}[\varepsilon_{||}(\omega) - \varepsilon_m]},
\]

\[(3.9)\]

The localized surface plasmon resonance (LSP) frequency of the hole can be determined from the equation

\[
\varepsilon_m + L_{||}[\varepsilon_{||}(\omega) - \varepsilon_m] = 0,
\]

\[(3.10)\]

the condition for which the denominator of $\alpha_{||}$ vanishes.
Using the linear dispersion relation, the intraband optical conductivity is \[ \sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{2 k_B T}{\tau - 1 - i \omega} \ln \left[ 2 \cosh \left( \frac{\epsilon_F}{2 k_B T} \right) \right], \] (3.11)
which in the case of \( \epsilon_F \gg k_B T \) is reduced to

\[ \sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{\epsilon_F}{\tau - 1 - i \omega} = \frac{2 \epsilon m \omega_p^2}{\pi \hbar^2 (\tau - 1 - i \omega)}, \] (3.12)

where \( \tau \) is determined by impurity scattering and electron-phonon interaction \( \tau^{-1} = \tau_{\text{imp}}^{-1} + \tau_{\text{e-ph}}^{-1} \). Using the mobility \( \mu \) of the NPG sheet, it can be presented in the form \( \tau^{-1} = e v_F^2 / (\mu E_F) \), where \( v_F = 10^6 \) m/s is the Fermi velocity in graphene. \( \omega_p = \sqrt{e^2 \epsilon_F / 2 \epsilon_m} \) is the bulk graphene plasma frequency.

It is well-known by now that hydrodynamic effects play an important role in graphene because
the Coulomb interaction collision rate is dominant, i.e. $\tau_{ee}^{-1} \gg \tau_{imp}^{-1}$ and $\tau_{ee}^{-1} \gg \tau_{e-ph}^{-1}$, which corresponds to the hydrodynamic regime. $\tau_{imp}^{-1}$ and $\tau_{e-ph}^{-1}$ are the electron-impurity and electron-phonon collision rates. Since for large absorbance and emittance, we choose a large Fermi energy, we are in the Fermi liquid regime of the graphene sheet. Taking the hydrodynamic correction into account, we also consider the hydrodynamically adjusted intraband optical conductivity,[6, 21]

$$\sigma_{\text{intra}}^{\text{HD}}(\omega) = \frac{\sigma_{\text{intra}}(\omega)}{1 - \eta^2 k_{||}^2/\omega^2},$$

(3.13)

where $\eta^2 = \beta^2 + D^2 \omega(\gamma + i\omega)$, $\beta^2 \approx \frac{3}{4} v_F^2$ is the intraband pressure velocity, $D \approx 0.4 \mu m$ is the diffusion length in graphene, and $\gamma = \tau^{-1}$ is the relaxation rate. Interestingly, the optical conductivity becomes $k$-dependent and nonlocal. Also, below we will conjecture that the diffusion length $D$ must be frequency-dependent. The effect of the hydrodynamic correction on the LSP resonances at around $\lambda = 4 \mu m, 7 \mu m, \text{and} 10 \mu m$ is shown in Figs. 3.3(a), (b), and (c), respectively.

Note that since $\varepsilon = 1 + \chi$, where $\chi$ is the susceptibility, it is possible to replace $\varepsilon'' = \chi''$. Alternatively, using the formula of the polarizability $\alpha = \varepsilon_0 \chi$ we can write $\varepsilon'' = \alpha''/\varepsilon_0$. The dielectric function for graphene is given by[82, 68]

$$\varepsilon_{||}(\omega) = \varepsilon_g + \frac{i\sigma_{2D}(\omega)}{\varepsilon_0 \omega d},$$

(3.14)

where $\varepsilon_g = 2.5$ is the dielectric constant of graphite and $d$ is the thickness of graphene. Inserting this formula into Eq. (3.10) gives

$$\varepsilon_m + L_{||}[\varepsilon_g - i \frac{e^2}{\pi \hbar^2} \varepsilon_F \frac{\varepsilon_F}{\varepsilon_0 \omega d (\tau^{-1} - i\omega)}] - \varepsilon_m] = 0,$$

(3.15)
Solving for the frequency and using the real part we obtain the LSP frequency,

\[
\text{Re} \omega_{\text{LSP}} = \frac{2L^2|\varepsilon_m\omega_p^2}{\pi \hbar^2 \left( L^2 + d^2 \varepsilon_0^2 \left[ L_{\parallel} (\varepsilon_g - \varepsilon_m) + \varepsilon_m \right]^2 \right)},
\]

(3.16)

which is linear in the Fermi energy \( \epsilon_F \).

The materials in Fig. 3.2 from top to bottom are: one single layer of hexagonal boron nitride (h-BN), for preventing oxidation of graphene at higher temperatures, one single layer of patterned graphene, 50 nm of Si\(_3\)N\(_4\), for large n-doping and gating, 50 nm of ITO, metallic contact for gating, which is also transparent in mid-IR, \( \lambda/4n_{\text{SU-8}} \) of SU-8,[82] which is transparent in mid-IR, and Au back mirror. \( n_{\text{SU-8}} = 1.56 \) is the refractive index of SU-8.

2D array of holes in graphene

Let us now consider the 2D array of circular holes in a graphene sheet. Since the dipole moments \( p_j = \delta p(R_j, \omega) \) interact with each other by inducing dipole moments, we need to consider the dressed dipole moment at each site \( R_j \) as source of the electric field, which is

\[
\tilde{p}_j = p_j + \alpha \sum_{j' \neq j} G_{jj'} \tilde{p}_{j'},
\]

(3.17)

where \( G_{jj'} \) is the dipole-dipole interaction tensor. Using Bloch’s theorem \( p_j = p_0 \exp(i k_{\parallel} \cdot R_{\parallel}) \),

the effective dipole moment becomes

\[
\tilde{p}_0 = p_0 + \tilde{p}_0 \alpha \sum_{j' \neq j} G_{jj'} e^{i k_{\parallel} \cdot (R_j - R_{j'})}.
\]

(3.18)
Figure 3.3: Emittance $\epsilon(\lambda)$ [equal to absorbance $A(\lambda)$] of the structure shown in Figs. 3.1 and 3.2 with Fermi energy $E_F = 1.0$ eV, mobility $\mu = 3000$ V/cm$^2$s, hole diameter of (a) $a = 30$ nm, (b) $a = 90$ nm, (c) $a = 300$ nm, and period (a) $P = 45$ nm, (b) $P = 150$ nm, (c) $P = 450$ nm at $T = 300$ K.

for each site $j$, and thus

$$\tilde{p}_0 = \frac{p_0}{1 - \alpha \tilde{G}}.$$  (3.19)
The lattice some over the dipole-dipole interaction tensor $G = \sum_{j' \neq j} G_{jj'} e^{i|k|(|R_j - R_{j'}|)}$ can be found in Ref. [97], i.e.

$$\text{Re}G \approx g/P^3, \quad (3.20)$$

$$\text{Im}G = S - 2k^3/3, \quad (3.21)$$

where $P$ is the lattice period,

$$S = \frac{2\pi k}{\Omega_0} \times \left\{ \begin{array}{ll}
\arccos \theta & \text{for s polarization,} \\
\cos \theta & \text{for p polarization.}
\end{array} \right\}, \quad (3.22)$$

$\Omega_0$ is the unit-cell area, and the real part is valid for periods much smaller than the wavelength. The factor $g = 5.52$ ($g = 4.52$) for hexagonal (square) lattice. The electric field created by the effective dipole moment is determined by

$$\tilde{p}_0 = \tilde{\alpha}E_0, \quad (3.23)$$

from which we obtain the effective polarizability of a hole in the coupled dipole approximation (CDA),

$$\tilde{\alpha} = \frac{\alpha}{1 - \alpha G}, \quad (3.24)$$

This formula is the same as in Refs. [116, 97], where the absorption of electromagnetic waves by arrays of dipole moments and graphene disks were considered, respectively. Thus, our result corroborates Kirchhoff’s law (see below). Consequently, we obtain the same reflection and transmission amplitudes as in Ref. [97], i.e.

$$r = \frac{\pm iS}{\alpha^{-1} - G}, \quad t = 1 + r, \quad (3.25)$$
where the upper (lower) sign and $S = 2\pi\omega/e\Omega_0 \cos \theta$ ($S = 2\pi\omega \cos \theta/e\Omega_0$) apply to s (p) polarization. Thus, the emittance and absorbance of the bare NPG sheet are given by\[87, 115, 55\]

$$\epsilon_g = A_g = 1 - |r|^2 - |t|^2. \quad (3.26)$$

The coupling to the interface of the substrate with reflection and transmission amplitudes $r_0$ and $t_0$, respectively, which is located basically at the same position as the NPG sheet, yields the combined reflection and transmission amplitudes\[97\]

$$R = r + \frac{tt'r_0}{1 - r_0t'}; \quad T = \frac{tt_0}{1 - r_0t'}, \quad (3.27)$$

where $r' = r$ and $t' = 1 - r$ are the reflection and transmission amplitudes in backwards direction, respectively. The results for the LSP resonances at around $\lambda = 4 \mu m, 7 \mu m,$ and $10 \mu m$ are shown in Figs. 3.3 (a), (b), and (c), respectively.

If we include also the whole substrate including cavity and Au mirror, we need to sum over all possible optical paths in the Fabry-Perot cavity, yielding

$$R_{FP} = R + TT'r_Aue^{i\delta} \sum_{m=0}^{\infty} r_m, \quad (3.28)$$

with

$$r_m = r_AuR'e^{i\delta}, \quad (3.29)$$

where $r_Au$ is the complex reflection amplitude of the Au mirror in the IR regime. $\delta = 2kL \cos \theta$ is the phase accumulated by one back-and-forth scattering inside the Fabry-Perot cavity of length $L$. $k \approx n_{SU-8}k_0$ is the wavenumber inside the cavity for an external EM wave with wavenumber

39
$k_0 = 2\pi/\lambda$. Since the sum is taken over a geometric series, we obtain

$$R_{FP} = R + \frac{TT' r_{Au} e^{i\delta}}{1 - r_{Au} R' e^{i\delta}}. \quad (3.30)$$

Since the transmission coefficient through the Au mirror can be neglected, we obtain the emittance $\epsilon$ and absorbance $A$ including cavity, i.e.

$$\epsilon_{FP} = A_{FP} = 1 - |R_{FP}|^2. \quad (3.31)$$

The results for the LSP resonances at around $\lambda = 4 \mu m$, $7 \mu m$, and $10 \mu m$ are shown in Figs. 3.3 (a), (b), and (c), respectively. The solid (black) curve represents the result of FDTD calculation. The dashed (blue) curve and the solid (black) curve are the emittances $\epsilon_g$ and $\epsilon_{FP}$ calculated by means of Eq. (3.26) and Eq. (3.31) for the bare NPG sheet without cavity and the NPG including cavity, respectively. The dotted (green) line exhibits a blue-shift due to the hydrodynamic correction shown in Eq (3.13) with $D(\nu = 30 \text{ THz}) \approx 0$. The blue-shifted dashed (magenta) curve and the blue-shifted dot-dashed (cyan) curve are the RPA-corrected LSP peaks due to the Coulomb interaction and the Coulomb interaction including electron-phonon interaction with the optical phonons of graphene, boron nitride, and Si$_3$N$_4$. This NPG sheet emits (a) into the atmospheric transparency window between 3 and 5 $\mu m$, (b) into the atmospheric opacity window between 5 and 8 $\mu m$, and (c) into the atmospheric transparency window between 8 and 12 $\mu m$.

Spectral radiance of incoherent photons

Using these results, let us consider the excitation of the graphene sheet near the hole by means of thermal fluctuations, which give rise to a fluctuating EM field of a localized surface plasmon (LSP). This can be best understood by means of the fluctuation-dissipation theorem, which provides a re-
lation between the rate of energy dissipation in a non-equilibrium system and the quantum and thermal fluctuations occurring spontaneously at different times in an equilibrium system.[66] The standard (local) fluctuation-dissipation theorem for fluctuating currents \( \delta \hat{J}_\nu(r, \omega) \) in three dimensions reads

\[
\left\langle \delta \hat{J}_\mu(r, \omega) \delta \hat{J}_\nu(r', \omega') \right\rangle = \omega \varepsilon_0 \varepsilon''_{\mu\nu}(r, \omega) \Theta(\omega) \\
\times \delta(\omega - \omega') \delta(r - r'),
\tag{3.32}
\]

where the relative permittivity \( \varepsilon(r, \omega) = \varepsilon'(r, \omega) + i\varepsilon''(r, \omega) = f(r)\varepsilon(\omega) \) and \( \mu, \nu = x, y, z \) are the coordinates. Note that since \( \varepsilon = 1 + \chi \), where \( \chi \) is the susceptibility, it is possible to replace \( \varepsilon'' = \chi'' \). Alternatively, using the formula of the polarizability \( \alpha = \varepsilon_0 \chi \) we can write \( \varepsilon'' = \alpha''/\varepsilon_0 \).

\( f(r) = 1 \) on the graphene sheet and 0 otherwise. Since the fluctuating currents are contained inside the two-dimensional graphene sheet, we write the local fluctuation-dissipation theorem in its two-dimensional form, i.e.

\[
\left\langle \delta \hat{J}_\mu(r_{||}, \omega) \delta \hat{J}_\nu(r'_{||}, \omega') \right\rangle = \sigma^{2D}_{\mu\nu}(r_{||}, \omega) \Theta(\omega) \\
\times \delta(\omega - \omega') \delta(r_{||} - r'_{||}),
\tag{3.33}
\]

where the fluctuating current densities have units of A/m\(^2\) and the coordinates are in-plane of the graphene sheet.

Using the method of dyadic Green’s functions, it is possible to express the fluctuating electric field generated by the fluctuating current density by

\[
\delta \hat{E}(r, \omega) = i\omega \mu_0 \int_{\Omega} G(r, r_0_{||}; \omega) \delta \hat{J}(r_0_{||}, \omega) d^2r_0_{||},
\tag{3.34}
\]

where \( \Omega \) is the surface of the graphene sheet. The LSP excitation around a hole can be well
approximated by a dipole field such that

\[
\delta \mathbf{J}(r_{0||}, \omega) = -i \omega \sum_j \delta \tilde{p}(\mathbf{R}_j, \omega)
\]

\[
= -i \omega \delta \tilde{p}_0(\omega) \sum_j \delta(\mathbf{r}_{0||} - \mathbf{R}_j),
\]

(3.35)

where \( \mathbf{R}_j = (x_j, y_j) \) are the positions of the holes in the graphene sheet.

Consequently, we have

\[
\delta \mathbf{E}(\mathbf{r}, \omega) = \omega^2 \mu_0 \delta \tilde{p}_0(\omega) \sum_j \mathbf{G}(\mathbf{r}, \mathbf{R}_j; \omega).
\]

(3.36)

The dyadic Green function is defined as

\[
\mathbf{G}(\mathbf{r}, \mathbf{r}'; \omega) = \left[ \mathbf{1} + \frac{1}{k(\omega)^2} \nabla \nabla \right] G(\mathbf{r}, \mathbf{r}'; \omega)
\]

(3.37)

with the scalar Green function given by

\[
G(\mathbf{r}, \mathbf{r}'; \omega) = \frac{e^{-i k(\omega) |\mathbf{r} - \mathbf{r}'|}}{4\pi |\mathbf{r} - \mathbf{r}'|},
\]

(3.38)

and \( k(\omega)^2 = (\omega^2/c^2)[\varepsilon_{xx}(\omega), \varepsilon_{yy}(\omega), \varepsilon_{zz}(\omega)] \).

Then, the fluctuation-dissipation theorem can be recast into the forms

\[
\langle \delta \tilde{p}_\mu(r_{0||}, \omega) \delta \tilde{p}_\nu^*(r'_{0||}, \omega') \rangle = \frac{\sigma^{2D}_{\mu\nu}(\mathbf{R}_i, \omega)}{\omega^2} \Theta(\omega) \delta(\omega - \omega')
\]

\[
	imes \delta(\mathbf{r}_{0||} - \mathbf{r}'_{0||}),
\]

(3.39)
Figure 3.4: Spectral radiance of NPG including cavity, as shown in Figs. 3.1 and 3.2, as a function of wavelength $\lambda$ with Fermi energy $E_F = 1.0$ eV, mobility $\mu = 3000$ V/cm$^2$s, hole diameter (a) $a = 30$ nm, (b) $a = 90$ nm, (c) $a = 300$ nm, and period (a) $P = 45$ nm, (b) $P = 150$ nm, (c) $P = 450$ nm at 1300 K, 1700 K, and 2000 K.

and thus we obtain

$$
\langle \delta \tilde{E}_\mu(r, \omega) \delta \tilde{E}^*_\nu(r', \omega') \rangle = \omega^4 \mu_0^2 \sum_{m, m'} \int_{\Omega} d^2 r_0 |G_{\mu m}(r, r_0; \omega)\rangle \times \int_{\Omega'} d^2 r_0' |G_{m' \nu}^*(r', r_0'; \omega')\rangle \langle \delta \tilde{p}_m(r_0, \omega) \delta \tilde{p}^{*}_{m'}(r_0', \omega)\rangle
$$

(3.40)
\[
\begin{align*}
I(r, \omega) &= \frac{\omega^2}{4\pi\varepsilon_0 c^3} \frac{1}{N} \sum_{\mu; m=x, y, z} |G_{\mu m}(r, R_j; \omega)|^2 \nonumber \times \Theta(\omega) \sigma_{\mu m}^{2D}(R_j, \omega) \\ &= \frac{\omega^2}{4\pi\varepsilon_0 c^3} \frac{1}{N} \sum_{\mu, m} |G_{\mu m}(r, R_0; \omega)|^2 \Theta(\omega) \sigma_{||}^{2D}(\omega) \sum_{\mu, m} |G_{\mu m}(r, R_0; \omega)|^2,
\end{align*}
\]

assuming that the dipole current of the LSP is in the plane of the graphene sheet, i.e. the xy-plane, and the polarizability is isotropic, i.e. \(\sigma_{||}^{2D} = \sigma_{xx}^{2D} = \sigma_{yy}^{2D}\), and the same for all holes. \(N\) is the number of holes. In order to obtain the spectral radiance in the far field, we need to integrate over the spherical angle. Using the results from the Appendix A, we obtain

\[
I_\infty(\omega) = \frac{\omega^2 \Theta(\omega)}{3\pi^2 \varepsilon_0 c^3} \sigma_{||}^{2D}(\omega) = \frac{\omega^2 \Theta(\omega)}{3c^2 \pi^2} A_{||}^{2D}(\omega),
\]
where we used the definition of the absorbance of a 2D material, i.e.

$$A_{2D}(\omega) = \frac{1}{\varepsilon_0} \text{Re} \sigma_{2D}(\omega) = \frac{1}{\varepsilon_0 c} \sigma'_{2D}(\omega),$$

(3.45)

with 2D complex conductivity $\sigma_{2D}(\omega)$. According to Kirchhoff’s law, emittance $\epsilon(\omega)$, absorbance $A(\omega)$, reflectance $R(\omega)$, and transmittance $T(\omega)$ are related by[54]

$$\epsilon(\omega) = A(\omega) = 1 - R(\omega) - T(\omega),$$

(3.46)

from which we obtain the grey-body thermal emission formula

$$I_\infty(\omega) = \frac{\omega^2 \Theta(\omega)}{3\pi^2 c^2} \epsilon_{2D}^p(\omega),$$

(3.47)

whose prefactor bears strong similarity to Planck’s black body formula in Eq. (3.1).

Using FDTD to calculate the emittance $\epsilon_{2D}^p(\omega)$, we evaluated the grey-body thermal emission according to Eq. (3.47) for the thermal emitter structure based on NPG shown in Figs. 3.1 and 3.2. Using COMSOL, we calculated the temperature distribution inside the NPG sheet, as shown in Fig. 3.5, when a bias voltage $V_{SD}$ is applied, which gives rise to Joule heating. The geometry of the simulated device is shown in Figs. 3.1 and 3.2. The area of the graphene channel is 10 $\mu$m x 10 $\mu$m. The thickness of the graphene sheet is 0.5 nm. The size of the gold contacts is 5 $\mu$m x 10 $\mu$m, with a thickness of 50 nm. Our results are shown in Figs. 3.4 (a), (b), and (c) for the temperatures 1300 K, 1700 K, and 2000 K of NPG. After integrating over the wavelength under the curves, we obtain the following thermal emission power per area:

Let us consider the dependence of the thermal emission of NPG on the angle $\theta$. Integrating over $r^2 \varphi$ we obtain

$$I(\theta, \omega) = \frac{\omega^2}{4\pi c^2} \Theta(\omega) \frac{11 + \cos(2\theta)}{16\pi} \epsilon_{2D}^p(\omega),$$

(3.48)
Figure 3.5: Temperature distribution inside the NPG sheet for various values of the bias voltage $V_{SD}$, calculated by means of COMSOL. As the bias voltage is increased, the maximum of temperature shifts away from the center of the NPG sheet due to the Peltier effect.

which is a clear deviation from a Lambert radiator. The pattern of the thermal radiation can be determined by

$$\dot{I}(\theta) = \frac{\int_0^{2\pi} \int_0^\pi I(r, \omega) r^2 d\phi}{\int_0^{2\pi} \int_0^\pi I(r, \omega) r^2 \sin \theta d\theta d\phi} = \frac{3}{64} [11 + \cos(2\theta)],$$

(3.49)

which is shown in Fig. 3.6. Interestingly, since we assumed that thermal emission is completely
Table 3.1: Power per unit area for different resonant peaks

<table>
<thead>
<tr>
<th>Resonance wavelength</th>
<th>Power per area</th>
</tr>
</thead>
<tbody>
<tr>
<td>4 µm</td>
<td>11,221 W/m²</td>
</tr>
<tr>
<td>7 µm</td>
<td>9820 W/m²</td>
</tr>
<tr>
<td>10 µm</td>
<td>6356 W/m²</td>
</tr>
</tbody>
</table>

incoherent [see Eq. (3.43)] the thermal emission from NPG is only weakly dependent on the emission angle \( \theta \), which can be clearly seen in Fig. 3.6.

Partial coherence of plasmons in graphene and the grey-body radiation

However, the assumption that thermal emission of radiation is incoherent is not always true. Since Kirchhoff’s law is valid, thermal sources can be coherent.[37] After theoretical calculations predicted that long-range coherence may exist for thermal emission in the case of resonant surface waves, either plasmonic or phononic in nature,[16, 43] experiments showed that a periodic microstructure in the polar material SiC exhibits coherence over many wavelengths and radiates in well-defined and controlled directions.[38] Here we show that the coherence length of a graphene sheet patterned with circular holes can be as large as 150 µm due to the plasmonic wave in the graphene sheet, thereby paving the way for the creation of phased arrays made of nanoantennas represented by the holes in NPG.

The coherence of thermal emission can be best understood by means of a nonlocal response function.[42] First, we choose the nonlocal hydrodynamic response function in Eq. (3.13). Using the 2D version of the fluctuation-dissipation theorem in Eq. (3.33), we obtain the nonlocal fluctuation-
dissipation theorem in the hydrodynamic approximation,

\[
\langle \delta \hat{J}_\mu(r_\parallel, \omega) \delta \hat{J}_\nu(r'_\parallel, \omega') \rangle = \sigma_{\mu\nu}^{\text{HD}}(\Delta r_\parallel, \omega) \Theta(\omega) \delta(\omega - \omega')
\]

\[
= \frac{1}{D} \int_0^\infty dk_\parallel \sigma_{\text{intra}}(\omega) e^{-ik_\parallel \Delta r_\parallel} \frac{1}{1 - \eta^2 k_\parallel^2 / \omega^2} \Theta(\omega) \delta(\omega - \omega')
\]

\[
= \sigma_{\text{intra}}(\omega) \frac{\omega \sqrt{\pi/2}}{D \eta} \sin \left( \frac{\omega \Delta r_\parallel}{\eta} \right) \Theta(\omega) \delta(\omega - \omega'),
\]

where \( \Delta r_\parallel = r_\parallel - r'_\parallel \) and \( \eta^2 = \beta^2 + D^2 \omega (\gamma + i\omega) \). This result suggests that the coherence length is given approximately by \( D \), which according to Ref. [6] would be \( D \approx 0.4 \, \mu\text{m} \). However, the resulting broadening of the LSP resonance peaks would be very large and therefore in complete contradiction to the experimental measurements of the LSP resonance peaks in Refs. [82, 80, 79]. Thus, we conclude that the hydrodynamic diffusion length must be frequency-dependent with \( D(\nu = 0) = 0.4 \, \mu\text{m} \). Using the Fermi velocity of \( v_F = 10^6 \, \text{m/s} \) and a frequency of \( \nu = 30 \, \text{THz} \), the average oscillation distance is about \( L = v_F \nu^{-1} = 0.033 \, \mu\text{m} \), which is much smaller than \( D(\nu = 0) \) in graphene. Thus we can approximate \( D(\nu = 30 \, \text{THz}) = 0 \). We conjecture that there is a crossover for \( D \) into the hydrodynamic regime when the frequency is reduced below around \( \nu_0 = 1 \) to 3 THz, below which the hydrodynamic effect leads to a strong broadening of the LSP peaks for NPG. Consequently, the viscosity of graphene should also be frequency-dependent and a crossover for the viscosity should happen at about the same frequency \( \nu_0 \). We plan to elaborate this conjecture in future work. Future experiments could corroborate our conjecture by measuring the absorbance or emittance as a function of wavelength for varying scale of patterning of the graphene sheet.

Next, let us consider the coherence of thermal emission by means of the nonlocal optical conduc-
Figure 3.6: Spherical density plot of the normalized angular intensity distribution \( \hat{I}(\theta) \) of the thermal emission from NPG in the case of incoherent photons.

Activity in the RPA approximation. Using the general formula

\[
\sigma(q, \omega) = \frac{ie^2\omega}{q^2} \chi^0(q, \omega),
\]

(3.51)

with

\[
\chi^0(q, \omega) \approx \frac{\epsilon_F q^2}{\pi \hbar^2 \omega (\omega + \frac{i}{\tau - 1})}
\]

(3.52)

in the low-temperature and low-frequency approximation, one obtains Eq. (5.2). Now, let us use the full polarization in RPA approximation including only the Coulomb interaction,

\[
\chi^{\text{RPA}}(q, \omega) = \frac{\chi^0(q, \omega)}{1 - v_c(q)\chi^0(q, \omega)},
\]

(3.53)
from which we obtain

\[ \sigma_{RPA}(q, \omega) = \frac{i e^2 \omega}{q^2} \chi(q, \omega) \]

\[ = \frac{i e^2 \omega \epsilon_F}{\pi \hbar^2 \omega (\omega + i \tau^{-1}) - \frac{e^2 \epsilon_F^2}{2 \epsilon_0}} q, \]

which introduces the nonlocal response via the Coulomb interaction in the denominator. The effect of the RPA correction on the LSP resonances at around \( \lambda = 4 \mu m, 7 \mu m, \) and \( 10 \mu m \) is shown in Figs. 3.3 (a), (b), and (c), respectively. After taking the Fourier transform, we obtain the nonlocal fluctuation-dissipation theorem in RPA approximation,

\[ \langle \delta \hat{J}_\mu(r||, \omega) \delta \hat{J}_\nu(r'||, \omega') \rangle = \sigma^RPA_{\mu\nu}(\Delta r||, \omega) \Theta(\omega) \delta(\omega - \omega') \]

\[ = \frac{\sqrt{2\pi \epsilon_0 \omega}}{C_{RPA}} e^{i K_{RPA} \Delta r||} e^{-\frac{\Delta r||}{C_{RPA}}} \Theta(\omega) \delta(\omega - \omega'), \]

where the coherence length in RPA approximation is

\[ C_{RPA} = \frac{e^2 |\epsilon_F|}{2\pi \hbar^2 \epsilon_0 \gamma \omega}, \]

and the coherence wavenumber is given by

\[ K_{RPA} = \frac{2\pi \hbar^2 \epsilon_0 \omega^2}{e^2 |\epsilon_F|}. \]

For simplicity, we switch now to a square lattice of holes. In the case of the LSP resonance for a square lattice of holes at \( \lambda = 10 \mu m, \) corresponding to \( \nu = 30 \) THz, \( \epsilon_F = 1.0 \) eV, \( \omega = 2\pi \nu, \) and \( \gamma = ev_F^2/(\mu E_F) = 0.3 \) THz for \( \mu = 3000 \) cm\(^2\)V\(^{-1}\)s\(^{-1}\), which results in a coherence length of \( C_{RPA} = 3 \mu m. \) This result is in reasonable agreement with the full width at half maximum (FWHM) values of the widths of the LSP resonance peaks in Refs. [82, 80, 79]. This coher-
ence length would allow to preserve coherence for a linear array of period $\mathcal{P} = 300$ nm and $C_{RPA}/\mathcal{P} = 10$ holes. In order to show the coherence length that can be achieved with graphene, we can consider a suspended graphene sheet with a mobility of $\mu = 15000$ cm$^2$V$^{-1}$s$^{-1}$. Then the coherence length increases to a value of $C_{RPA} = 13$ $\mu$m, which would allow for coherence over a linear array with $C_{RPA}/\mathcal{P} = 43$ holes.

![Figure 3.7](image.png)

Figure 3.7: Coherence length $C_{\text{FDTD}}$ and coherence time $\tau_{\text{FDTD}}$ of emitted photons, extracted from the full-width half-maximum (FWHM) of the spectral radiances shown in Figs. 3.4 (a), (b), and (c).

In the case of the LSP resonance for a square lattice of holes at $\lambda = 5$ $\mu$m, corresponding to $\nu = 60$ THz, $\epsilon_F = 1.0$ eV, $\omega = 2\pi\nu$, and $\gamma = ev_F^2/(\mu E_F) = 0.3$ THz for $\mu = 3000$ cm$^2$V$^{-1}$s$^{-1}$, which results in a coherence length of $C_{RPA} = 1.5$ $\mu$m. Considering again a suspended graphene sheet, the coherence length can be increased to $C_{RPA} = 6.7$ $\mu$m. Since the period in this case is $\mathcal{P} = 45$ nm, the coherence for $\mu = 3000$ cm$^2$V$^{-1}$s$^{-1}$ and $\mu = 15000$ cm$^2$V$^{-1}$s$^{-1}$ can be preserved for a linear array of $C_{RPA}/\mathcal{P} = 33$ and 148 holes, respectively.
The coherence length and time of thermally emitted photons is larger because the photons travel mostly in vacuum. Taking advantage of the Wiener-Kinchine theorem,[37] we can extract the coherence length $C_{\text{FDTD}}$ and coherence time $\tau_{\text{FDTD}}$ of thermally emitted photons by means of the full-width half-maximum (FWHM) of the spectral radiances shown in Figs. 3.4 (a), (b), and (c). Our results are shown in Fig. 3.7. The coherence length of the thermally emitted photons can reach up to $C_{\text{FDTD}} = 150 \, \mu\text{m}$ at a resonance wavelength of $\lambda = 4 \, \mu\text{m}$. This means that the coherence length of the thermally emitted photons is about 37 times larger than the wavelength.

Phased array of LSPs in graphene

Thus, the latter large coherence length allows for the coherent control of a 150x150 square array of holes with period $\mathcal{P} = 45 \, \text{nm}$, individually acting as nanoantennas, that can be used to create a phased array of nanoantennas. One of the intriguing properties of a phased array is that it allows to control the directivity of the emission of photons, which is currently being implemented for large 5G antennas in the 3 to 30 GHz range. The beamsteering capability of our NPG sheet is shown in Fig. 3.8. In contrast, our proposed phased array based on NPG can operate in the 10 to 100 THz range.

The temporal control of the individual phases of the holes requires an extraordinary fast switching time of around 1 ps, which is not feasible with current electronics. However, the nonlocal response function reveals a spatial phase shift determined by the coherence wavenumber $K_{\text{RPA}}$, which is independent of the mobility of graphene. In the case of the LSP resonance at $\lambda = 4 \, \mu\text{m}$, we obtain $\lambda_{\text{RPA}} = 2\pi/K_{\text{RPA}} = 6 \, \mu\text{m}$, resulting in a minimum phase shift of $2\pi\mathcal{P}/\lambda_{\text{RPA}} = 0.042 = 2.4^\circ$ between neighboring holes, which can be increased to a phase shift of $9.7^\circ$ by decreasing the Fermi energy to $E_F = 0.25 \, \text{eV}$. Thus, the phase shift between neighboring holes can be tuned arbitrarily between $2.4^\circ$ and $9.7^\circ$ by varying the Fermi energy between $\epsilon_F = 1.0 \, \text{eV}$ and $\epsilon_F = 0.25 \, \text{eV}$.
Fig. 3.8 shows the capability of beamsteering for our proposed structure by means of directional thermal emission, which is tunable by means of the gate voltage applied to the NPG sheet.

Due to the full control of directivity with angle of emission between $\theta = 12^\circ$ and $\theta = 80^\circ$ by tuning the Fermi energy in the range between $\epsilon_F = 1.0$ eV and $\epsilon_F = 0.25$ eV, thereby achieving beamsteering by means of the gate voltage, our proposed mid-IR light source based on NPG can be used not only in a vertical setup for surface emission, but also in a horizontal setup for edge emission, which is essential for nanophotonics applications.

Figure 3.8: Directivity of the thermal emission from NPG where the holes act as nanoantennas in a phased array. This emission pattern for $\epsilon_F = 1.0$ eV can be used for surface-emitting mid-IR sources.

In the case of a 150x150, 75x75, 56x56, 37x37 square lattice of holes (size of lattice matches coherence length) with period $P = 45$ nm and hole diameter of 30 nm, introducing a relative phase of 2.43°, 4.86°, 7.28°, 9.71° between the nanoantennas allows for beamsteering in the range between $\theta = 12^\circ$ and $\theta = 80^\circ$ by tuning the Fermi energy in the range between $\epsilon_F = 1.0$ eV and $\epsilon_F = 0.25$ eV. In the case of a 150x150, 75x75, 56x56, 37x37 square lattice of holes (size of lattice matches coherence length) with period $P = 45$ nm and hole diameter of 30 nm, introducing a relative phase of 2.43°, 4.86°, 7.28°, 9.71° between the nanoantennas allows for beamsteering in
the range between $\theta = 12^\circ$ and $\theta = 80^\circ$ by tuning the Fermi energy in the range between $\epsilon_F = 1.0$ eV and $\epsilon_F = 0.25$ eV.

Conclusion

In conclusion, we have demonstrated in our theoretical study that NPG can be used to develop a plasmonically enhanced mid-IR light source with spectrally tunable selective thermal emission. Most importantly, the LSPs along with an optical cavity increase substantially the emittance of graphene from about 2% for pristine graphene to 80% for NPG, thereby outperforming state-of-the-art graphene light sources working in the visible and NIR by at least a factor of 100. Combining our proposed mid-IR light source based on patterned graphene with our demonstrated mid-IR detector based on NPG[79], we are going to develop a mid-IR spectroscopy and detection platform based on patterned graphene that will be able to detect a variety of molecules that have mid-IR vibrational resonances, such as CO, CO$_2$, NO, NO$_2$, CH$_4$, TNT, H$_2$O$_2$, acetone, TATP, Sarin, VX, etc. In particular, a recent study showed that it is possible to detect the hepatitis B and C viruses label-free at a wavelength of around 6 $\mu$m.[77] Therefore, we will make great effort to demonstrate that our platform will be able to detect with high sensitivity and selectivity the COVID-19 virus and other viruses that pose a threat to humanity.
In this chapter, we study the model of an ultrasensitive mid-infrared (mid-IR) photodetector operating in the mid-wavelength infrared (MWIR) and long-wavelength infrared (LWIR) domains consisting of a hybrid heterostructure made of nanopatterned graphene (NPG) and phase changing material vanadium dioxide (VO$_2$). 

Introduction

Due to the low photon energy of IR radiation cryogenic cooling is required for highly sensitive photodetection based on low band gap materials like mercury-cadmium-telluride (HgCdTe). Various kinds of microbolometers primarily based on vanadium oxide (VO$_x$) offer uncooled detection of IR radiation. However, microbolometers suffer from low sensitivity, slow response and tedious multi-step complex lithographic processes in comparison to cooled photodetectors [75]. Photodetection based on the bolometric effect takes advantage of the dependence of the resistivity on the temperature to detect incident light, typically in the infrared regime. Vanadium oxide (VO$_2$) is considered as one of the standard materials for building microbolometers with broadband mid-IR photodetection because it features a reversible insulator-to-metal phase transition (IMT) when heated above the phase transition temperature $T_c$, which is slightly above and close to room temperature [76]. Bulk VO$_2$ undergoes a phase transition from an insulating state with monoclinic crystal structure below 68°C to a metallic state with rutile crystal structure above $T_c = 68$°C (=341 K) [36, 63, 19]. This phase transition is fully reversible with a hysteresis loop, occurs on a sub-

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1Spectrally selective mid-IR light detection based on hybrid nanopatterned graphene and vanadium oxide heterostructure operating close to room temperature, Muhammad Waqas Shabbir, Sayan Chandra, Michael N Leuenberger, 2021. Manuscript submitted for publication.
picosecond timescale [3, 23], and can be initiated either thermally, electrically [93], or optically [17]. While for bulk VO$_2$ optically induced IMT can only be achieved by pumping above the band gap of $E_g = 670$ meV, it is possible to induce IMT in thin films at energies of 200 meV and above (corresponding to wavelengths of $\lambda = 6.2 \, \mu m$ and below) due to electronic defects inside the band gap [73]. This is the reason why VO$_2$ thin films containing defects can be used for IR detection in a wide IR range, in particular in the 3-5 $\mu m$ range. Interestingly, when VO$_2$ is in the form of a thin film, its transition temperature $T_c$ depends strongly on the thickness $d$ of the film, i.e. $T_c$ decreases from 65°C (=338 K) for $d = 25$ nm down to 52°C (=325 K) for $d = 3$ nm [107]. While IR radiation with wavelengths above about $\lambda = 1 \, \mu m$ cannot detect the change in thickness of around 20 nm, it certainly distinguishes between the insulating and the metallic phase of VO$_2$. All these properties make VO$_2$ the ideal material for developing mid-IR photodetectors based on the IMT effect.

However, photodetection of mid-IR light with wavelength above about 6 $\mu m$ is inefficient with VO$_2$ bulk or thin films because of the relatively weak interaction between the incident photons and the optical phonons in VO$_2$. This interaction is so weak that the IMT cannot be achieved. That is why in the wavelength regime of 8 to 12 $\mu m$ the photodetection is based purely on the bolometric effect in the semiconducting phase of VO$_2$ [8, 20]. Here, by adding a single layer of nanopatterned graphene (NPG) on top of a layer of VO$_2$, we present the model of a photodetector that not only greatly enhances the absorption of mid-IR light energy in the longer wavelength regime from $\lambda = 6 \, \mu m$ and exceeding 12 $\mu m$ but also narrows the absorption bandwidth to 0.1 $\mu m$ within the mid-IR range of 3 to 12 $\mu m$, thereby enabling plasmonically enhanced spectrally selective absorption of mid-IR light for the IMT effect in a heterostructure made of NPG and VO$_2$. 

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Figure 4.1: Schematic showing our proposed IMT-based mid-IR light photodetector consisting of hybrid NPG-VO$_2$.

Design of the hetero-structure device

Building upon the knowledge we acquired for developing spectrally selective photodetectors made of NPG that detect mid-IR light by means of the photothermoelectric effect [82, 80, 79] and the knowledge we acquired for developing thermal emitters based on NPG [86], we develop here the model of a mid-IR microbolometer that consists of an hexagonal boron nitride (h-BN) coated NPG, silicon nitride (Si$_3$N$_4$), indium tin oxide (ITO), VO$_2$, polymer, and gold (Au) mirror, as shown in Fig. 4. The materials from top to bottom are: 1 single layer of hexagonal boron nitride (h-BN), for preventing oxidation of graphene at higher temperatures (*), 1 single layer of patterned graphene (*), 65 nm of Si$_3$N$_4$ (*), for large n-doping and gating, 65 nm of ITO (*), metallic contact for gating, which is also transparent in mid-IR, 20 nm of h-BN (*), used for efficient heat transfer, 3 nm of
VO$_2$ (*), contacted with source and drain Au leads, $\lambda/4n_{\text{SU8}}$ of SU8,[82] which is transparent in mid-IR, and Au back mirror. $n_{\text{SU8}} = 1.56$ is the refractive index of SU8. All the layers marked with an asterix (*) are patterned with the same hexagonal lattice of holes.

The main working principle of our envisioned mid-IR photodetector can be summarized as follows. After mid-IR photons get absorbed by the NPG sheet at a wavelength that matches the localized surface plasmon (LSP) resonance, the NPG’s temperature increases and transfers the heat through the Si$_3$N$_4$ and ITO layers to the VO$_2$ layer. By patterning not only the graphene sheet but also the Si$_3$N$_4$ and ITO layers we maximize the heat transfer to the VO$_2$ layer. Once the VO$_2$ layer’s temperature increases above the phase transition temperature $T_c$, the VO$_2$ layer undergoes a transition from insulating to metallic phase. The patterning of the VO$_2$ layer decreases its volume, thereby decreasing the heat required to drive the VO$_2$ layer over $T_c$, which in turn increases the sensitivity (NEP) of our proposed NPG-VO$_2$ photodetector. During this whole time a bias voltage $V_b$ is applied to the VO$_2$ layer, which upon phase transition triggers a current through the VO$_2$ layer, which can be detected electronically in about 1 ms, shorter than the detection time of typical VO$_2$ bolometers [20].

Theoretical model

For the electronic response of the graphene sheet and the VO$_2$ layer to the incident mid-IR photons, we need to consider the intraband conductivity of graphene and the dielectric function of VO$_2$ in the insulating and metallic regimes. Using the linear dispersion relation, the intraband optical conductivity of graphene is [82, 68]

$$\sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{2k_B T}{\tau - i\omega} \ln \left[ 2 \cosh \left( \frac{E_F}{2k_B T} \right) \right], \quad (4.1)$$
Figure 4.2: Absorbance of the NGP-VO$_2$ photodetector shown in Fig. 4 (a) when VO$_2$ is in the insulating phase. (c) when VO$_2$ is in the metallic phase. (b) and (d) show absorbance as a function of wavelength and Fermi energy, showing the overall tunability of the LSP resonance peaks, when VO$_2$ is in the semiconducting and metallic phase, respectively.

which in the case of $E_F \gg k_B T$ is reduced to

$$\sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{E_F}{\tau^{-1} - i\omega} = \frac{2\varepsilon_m \omega_p^2}{\pi \hbar^2 (\tau^{-1} - i\omega)}; \quad (4.2)$$

where $\tau$ is determined by impurity scattering and electron-phonon interaction $\tau^{-1} = \tau_{\text{imp}}^{-1} + \tau_{\text{e-ph}}^{-1}$. Using the mobility $\mu$ of the NPG sheet, it can be presented in the form $\tau^{-1} = ev_F^2/\mu E_F$, where $v_F = 10^6$ m/s is the Fermi velocity in graphene. $\omega_p = \sqrt{e^2 E_F/2\varepsilon_m}$ is the bulk graphene plasma
Following Refs. Wang2014 and Barker1966, the dielectric function of VO$_2$ can be modeled by means of a Drude-Lorentz model for the insulating phase,

$$
\epsilon_i(\omega) = \epsilon_\infty + \sum_{j=1}^{N} \frac{S_j \omega_j^2}{\omega_j^2 - i\gamma_j \omega - \omega^2},
\tag{4.3}
$$

where $j = 1, \ldots, N$ denotes the $N$ phonon modes, $\omega_j$ are the phonon vibration frequencies, $\gamma_j$ are the scattering rates, and $S_j$ are the oscillation strengths. The value of these parameters can be found in Ref. Barker1966. The metallic phase of VO$_2$ can be modeled by means of the Drude formula,

$$
\epsilon_m(\omega) = -\epsilon_\infty \frac{\Omega_p^2}{\omega^2 + i\omega \Gamma},
\tag{4.4}
$$

where $\Omega_p = \sqrt{N_f e^2/\epsilon_0 m^*} = 8000 \text{ cm}^{-1}$ is the plasma frequency, $\Gamma = e/m^* \mu_{VO_2} = 10000 \text{ cm}^{-1}$ is the collision frequency, with $\mu_{VO_2} = 2 \text{ cm}^2/\text{Vs}$ being the mobility, $N_f = 1.3 \times 10^{22} \text{ cm}^{-3}$ the free-carrier concentration, and $m^* = 2m_e$ the effective mass of the charge carriers. $m_e$ is the free electron mass.

Numerical simulations of absorbance

We used the finite-difference time domain method (FDTD) to calculate the absorbance of the hybrid NPG-VO$_2$ photodetector as shown in Fig. 4.2 with hexagonal nanopattern made of a hole diameter of 300 nm and a period of 450 nm. (a) Absorbance when VO$_2$ is in the insulating phase. The resonance peaks of the LSPs are clearly visible and tunable by means of a gate voltage that shifts the Fermi energy inside the NPG sheet. Absorbance of mid-IR light in this wavelength regime in insulating VO$_2$ is very low. (b) Absorbance as a function of wavelength and Fermi energy, showing the overall tunability of the LSP resonance peaks, when VO$_2$ is in the insulating phase.
phase. (c) Absorbance of the NGP-VO$_2$ photodetector when VO$_2$ is in the metallic phase. The resonance peaks of the LSPs are still clearly visible. Compared with the absorbance for insulating VO$_2$ shown in (a), the metallic VO$_2$ layer absorbs mid-IR light over a large wavelength regime and exhibits a maximum of about 30% at a wavelength of about $\lambda = 9 \mu$m. (d) Absorbance as a function of wavelength and Fermi energy, showing the overall tunability of the LSP resonance peaks, when VO$_2$ is in the metallic phase.

The resonance peaks due to the absorption of mid-IR light by localized surface plasmons (LSPs) in NPG are clearly visible and similar to previous reports [82, 80, 79, 86]. The main difference is that the VO$_2$ layer exhibits very low absorbance (around 6%) of mid-IR light in the insulating phase but a larger broadband absorption in the metallic phase. The thicker the metallic VO$_2$ layer is, the stronger is the absorption of mid-IR light. We chose a VO$_2$ layer thickness of 3 nm for two important reasons: Firstly, a thinner VO$_2$ layer results in weaker absorption and thus makes it easier for the photodetector to cool down after the incident mid-IR radiation is turned off. Secondly, the thinner VO$_2$ layer has a lower phase transition temperature of $T_c = 52^\circ$C ($=325$ K) and thus requires less external heating for keeping the photodetector at an optimum operating temperature (see below).

The absorbance resonance peak as a function of wavelength can be tuned by means of the Fermi energy of NPG, as shown in Fig. 4.2 (b) when VO$_2$ is in the insulating phase and in Fig. 4.2 (d) when VO$_2$ is in the metallic phase. Thus, the gate voltage allows the operating wavelength of the VO$_2$-NPG photodetector to be tuned.
Figure 4.3: Temperature and photocurrent as functions of temperature and time of the NGP-VO$_2$ photodetector shown in Fig. 4 with nanopattern made of a hole diameter of 300 nm and a period of 450 nm. (a) Temperature $T$ of the VO$_2$ layer as a function of time, showing the photodetection process while the incident mid-IR light is turned on periodically for 1.3 ms, and the applied bias voltages of $V_b = 1.7$ V are turned on periodically for 1.3 ms, as shown in (b). (c) Photocurrent $I_{ph}$ through the VO$_2$ layer as a function of temperature $T$ for a constant applied bias voltage $V_b = 1.7$ V during the heating (red curve) and cooling (blue curve) process. The base temperature of the substrate is kept at $T_0 = 319$ K. The temperature difference of the hysteresis is $\Delta T = 5$ K. (d) Temperature profile at times $t = 1.2$ ms when VO$_2$ is in the metallic phase. The maximum temperature is $T_{\text{max}} = 337$ K. The gap between the NPG and the source and drain contacts ensures that there is no electrical current flowing through the NPG. (e) Temperature profile at times $t = 2.9$ ms when VO$_2$ is in the insulating phase. The maximum temperature is $T_{\text{max}} = 319.2$ K.

Operation of the NPG-VO$_2$ detector

For modeling the operation of the proposed NPG-VO$_2$ mid-IR photodetector, we used COMSOL and the following theory for the thermoelectric properties of VO$_2$ close to the phase transition temperature $T_c$. The VO$_2$ layer is operated around the IMT phase transition temperature $T_c$. The
performance of the bolometric detection can be analyzed by means of the heat equation and a hysteresis model [2]. The heat equation reads

$$C \frac{dT}{dt} = \alpha P + I^2 R(T) - G(T - T_h),$$

where $C$ is the heat capacity, $\alpha$ is the absorbance, $P$ is the power of the incident radiation, $I$ is the time-independent bias current, $R(T)$ is the temperature-dependent resistance, $G$ is the thermal conductivity of the heat sink, and $T_h$ is the time-independent temperature of the heat sink. The hysteretic behavior of $R(T)$ for VO$_2$ layer can be calculated by

$$R(T) = 17 \exp \left( \frac{2553}{T + 273} \right) g(T) + 140,$$

where the semiconductor volume fraction is given by

$$g(T) = \frac{1}{2} + \frac{1}{2} \tanh \beta \left[ \frac{\delta w}{2} + T_c - \left( T + T_{pr} P \left( \frac{T - T_r}{T_{pr}} \right) \right) \right],$$

where $w$ is the width of the hysteresis, $\beta$ is a function of $dg/dT$ at $T_c$, $P(x)$ is an arbitrary monotonically decreasing function, and $\delta = \text{sign}(dT/dt)$. The proximity temperature is given by

$$T_{pr} = \frac{\delta w}{2} + T_c - \frac{1}{\beta} \arctan \left( 2g_r - 1 \right) - T_r.$$

Eqs. (4.5)-(4.8) describe the hysteretic behavior of the plasmonically driven bolometric photodetector. The incident power $P_{inc}$ is then given by the energy pumped into the plasmonic nanostructure.

Using this thermoelectric theory and combining it with our FDTD results, we developed a photothermoelectric theory of the NPG-VO$_2$ heterostructure. We optimized the geometry of the pho-
detector in terms of performance. We choose a channel width of 5 µm (distance between the source and drain contacts) and a channel length of 5 µm. The larger the channel length, the smaller is the resistance of the VO$_2$ channel, resulting in reduced Joule heating. This is the method we choose to reduce the resistance instead of increasing the thickness of the VO$_2$ layer, which would increase the absorption of the VO$_2$ layer in the metallic phase. For the initial study we consider a thickness of 3 nm. Note that the current flows only through VO$_2$, not through NPG. NPG is used only as a photothromoelectric heating element. When the incident mid-IR light is off, a very weak dark current $I$ on the scale of µA is flowing when a bias voltage $V_b$ is applied in the range from 0.1 to 1.7 V. In stark contrast, when the incident mid-IR light is on, a much larger light current $I$ on the scale of mA is flowing with the same applied bias voltage $V_b$. This effect is due to the phase transition of VO$_2$ between insulating and metallic phases. We take advantage of this effect to develop the model of an ultrasensitive photodetector based on the NPG-VO$_2$ heterostructure.

After modeling the heating and cooling of the NPG-VO$_2$ heterostructure as a function of time, we identified the optimum photodetection process. Fig. 4.3 (a) shows the temperature $T$ of the VO$_2$ layer as a function of time $t$ resulting from the incident light and voltage pulse trains depicted in Fig. 4.3 (b). The voltage pulse train is required to let the VO$_2$ layer cool down after each signal detection, i.e. it ensures the cooling of the photodetector below the phase transition temperature $T_c$, as shown in Fig. 4.3 (d) and (e). The amplitude of the voltage pulse train is set at a relatively high value of $V_b = 1.7$ V in order to maximize the photoresponsivity $R$, detectivity $D^*$, and sensitivity NEP. The inset of Fig. 4.3 (a) shows the magnification between times $t = 0$ ms and $t = 3$ ms in order to demonstrate that the temperature increase of VO$_2$ with pristine graphene or without graphene is not sufficient to trigger an IMT in VO$_2$. This means that NPG is absolutely necessary for triggering the IMT in VO$_2$. The photocurrent $I_{ph}$ through the VO$_2$ layer as a function of temperature $T$ for a constant applied bias voltage $V_b = 1.7$ V exhibits the expected hysteresis loop as shown in Fig. 4.3 (c). We used the theoretical models and experimental values of conductivity,
Figure 4.4: Illustration of NPG-VO$_2$ photodetector with a VO$_2$ layer whose thickness has a gradient varying from 3 nm to 12 nm. Note that the gate voltage is applied by means of a gold contact (not shown) above h-BN.

thermal conductivity, and heat capacity of VO$_2$ given in Refs. [70, 117, 84, 67]. Our results reveal that the NPG-VO$_2$ photodetector has a response time on a time scale of the order of 1 ms, shorter than current microbolometers based on VO$_2$ alone [20]. The temperature profiles shown in Fig. 4.3 (d) and (e) reveal the stark contrast between the heating in the metallic phase and the insulating phase of VO$_2$, respectively.

Since we want to realize a linear dependence of the photocurrent $I_{ph}$ as a function of input power $P_{inc}$ of the mid-IR light, we add a gradient in the thickness of the VO$_2$ layer as shown in Fig. 4.4, i.e. the VO$_2$ layer thickness is varied from 3 nm to 12 nm. Fig. 4.5 shows four temperature profiles of the cross section of the VO$_2$ layer showing nine different thicknesses. The main idea is based on the
Figure 4.5: Temperature and photocurrent as functions of temperature and time of the NGP-VO$_2$ photodetector shown in Fig. 4 with nanopattern made of a hole diameter of 300 nm and a period of 450 nm. (a) Temperature $T$ of the VO$_2$ layer as a function of time (b) voltage and light pulse. (c) Photocurrent $I_{ph}$ through the VO$_2$ layer. (d) Four temperature profiles of the cross section of the VO$_2$ layer showing nine different thicknesses. (e) Time dependent temperature curve for a light and voltage pulse pulse train shown in (f).

The fact that the larger the power of the incident light $P_{inc}$, the more steps undergo the phase transition from insulating to metallic phase, which results in a larger photocurrent $I_{ph}$. After modeling the heating and cooling of the NPG-VO$_2$ gradient heterostructure as a function of time, we identified the optimum photodetection process. The widths and thicknesses of the steps are chosen such that the total resistance of step remains the same, i.e. the resistance is $R = \rho w/A$, where $\rho$ is the resistivity of VO$_2$ and $w$ is the channel width. The cross-sectional area $A = ld$, where $l$ is the length of a step and $d$ is its thickness. This geometry gives rise to a photocurrent $I_{ph}$ that is linear as a function of $P_{inc}$. In particular, we find that the voltage pulse train must consist of pulses with two applied
bias voltages $V_{b1}$ and $V_{b2}$, where the former is responsible for the triggering of the IMT in VO$_2$ and the latter is substantially lower than the former in order to reduce the Joule heating and to ensure the linear dependence of $I_{ph}$ as a function of input power $P_{inc}$. Fig. 4.5 shows temperature and photocurrent as functions of temperature and time of the NGP-VO$_2$ photodetector shown in Fig. 4 (a) Temperature $T$ of the VO$_2$ layer as a function of time, showing the photodetection process while the incident mid-IR light is turned on periodically for 1.3 ms, and the applied bias voltages of $V_b = 1.7$ V are turned on periodically for 1.3 ms, as shown in (b). (c) shows linear dependence of current on incident power.(d) Four temperature profiles of the cross section of the VO$_2$ layer showing nine different thicknesses. The larger the power of the incident light $P_{inc}$, the more steps undergo the phase transition from insulating to metallic phase. This gradient of thickness gives rise to the linear photocurrent $I_{ph}$ as a function of $P_{inc}$, shown in (c). As before, the voltage pulse train is required to let the VO$_2$ layer cool down after each signal detection, i.e. it ensures the cooling of the photodetector below the phase transition temperature $T_h$. The amplitude of the first part of the voltage pulse is set at a relatively high value of $V_{b1} = 1.7$ V in order to maximize the photoresponsivity $R$, detectivity $D^*$, and sensitivity NEP, and the second part of the voltage pulse is set to the substantially lower value of $V_{b2} = 0.2$ V in order to reduce Joule heating. Therefore it is possible to operate the NPG-VO$_2$ photodetector in pulse train mode, as shown in Fig. 4.5 (e). The dependence of the temperature $T$ of the VO$_2$ as a function of incident light power is depicted by means of several graphs for $P_{inc} = 50$ nW, 67 nW, and 95 nW. Owing to the thickness gradient of the VO$_2$ layer, we obtain the linear photocurrent $I_{ph}$ as a function of input power $P_{inc}$, as shown in Fig. 4.5 (c). This provides an optimized mapping of $P_{inc}$ onto $I_{ph}$ for maximum dynamic range.
Responsivity and detectivity of the photodetector

The responsivity $\mathcal{R}$ of the NPG-VO$_2$ photodetector can be calculated by means of the formula [79]

$$\mathcal{R} = \frac{(I_{\text{light}} - I_{\text{dark}})R}{P_{\text{inc}}}$$  \hspace{1cm} (4.9)

where $R$ is the resistance of VO$_2$ in the metallic phase, $I_{\text{light}}$ is the light current when the incident light is on, $I_{\text{dark}}$ is the dark current when the incident light is off, and $P_{\text{inc}}$ is the power of the incident light. The responsivity as a function of Fermi energy $E_F$ of NPG is shown in Fig. 5.5.

The sensitivity of the NPG-VO$_2$ photodetector is determined by the noise-equivalent power NEP, being an important figure of merit for the performance of a photodetector. The NEP of a photodetector provides a measure for the minimum detectable power per 1 Hz of bandwidth. The formula for NEP reads [79]

$$\text{NEP} = \frac{\nu_n}{\mathcal{R}},$$  \hspace{1cm} (4.10)

where

$$\nu_n = \sqrt{\nu_t^2 + \nu_b^2 + \nu_f^2}$$  \hspace{1cm} (4.11)

is the root-mean-square of the total noise voltage, which consists of the sum over all possible noise voltages, such as the thermal Johnson-Nyquist noise $\nu_t$, due to thermal motion of the charge carriers and independent of the bias voltage $V_b$, the shot noise $\nu_b$, due to the discrete nature of uncorrelated charge carriers, and the 1/f noise $\nu_f$, also called flicker noise, due to random resistance fluctuations. The Johnson noise is given by [40]

$$\nu_t = \sqrt{4k_BTR},$$  \hspace{1cm} (4.12)

where $k_B$ is the Botzmann constant, $T$ is the temperature, and $R$ is the resistance. The shot noise
is given by \[ (4.13) \]

\[ \nu_b = \sqrt{2eI_dR^2}, \]

where \( e \) is the elementary charge and \( I_d \) is the dark current. Since the dark current is very low and the NPG-VO\(_2\) photodetector operates close to room temperature, the shot noise is much smaller than the Johnson noise. Therefore, we can safely neglect the shot noise. At a modulation frequency of \( V_b \) of around 1 kHz we can also neglect the \( 1/f \) noise. Using the NEP, we can calculate the detectivity of the NPG-VO\(_2\) photodetector by means of the formula \[ (4.14) \]

\[ D^* = \frac{\sqrt{A}}{\text{NEP}}, \]

where \( A \) is the area of the photodetector. The results of these figures of merit are shown in Table 4.1.

Table 4.1: Figures of Merit of the NPG-VO\(_2\) gradient photodetector at a Fermi energy of \( E_F = 1.0 \) eV.

<table>
<thead>
<tr>
<th>( \Delta T ) [K]</th>
<th>( P ) [nW]</th>
<th>( T_0 ) [K]</th>
<th>( R ) [V/W]</th>
<th>( \text{NEP} ) [fW/( \sqrt{Hz} )]</th>
<th>( D^* ) [Jones]</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.0</td>
<td>50</td>
<td>319</td>
<td>( 4.4 \times 10^5 )</td>
<td>21</td>
<td>( 2.4 \times 10^{10} )</td>
</tr>
<tr>
<td>5.0</td>
<td>67</td>
<td>319</td>
<td>( 3.3 \times 10^5 )</td>
<td>27</td>
<td>( 1.8 \times 10^{10} )</td>
</tr>
<tr>
<td>5.0</td>
<td>95</td>
<td>319</td>
<td>( 2.3 \times 10^5 )</td>
<td>39</td>
<td>( 1.3 \times 10^{10} )</td>
</tr>
</tbody>
</table>

Table 4.1 shows that the detectivity \( D^* \) of the NPG-VO\(_2\) photodetector operating close to room temperature is close to \( D^* \) of cryogenically cooled HgCdTe photodetectors. The NPG-VO\(_2\) photodetector reaches a sensitivity close to VO\(_2\) microbolometers while exhibiting a shorter detection time of around 1 ms and being able to detect photons with spectral selectivity in the MWIR and LWIR regimes, which is impossible for VO\(_2\) microbolometers.
Conclusion

In conclusion, we present the model of an ultrasensitive mid-infrared (mid-IR) photodetector based on a heterostucture made of NPG and VO\textsubscript{2} with spectral selectivity in the MWIR and LWIR regimes. This hybrid NPG-VO\textsubscript{2} photodetector has a narrowband absorption in the mid-IR regime (3-12 \( \mu \)m) that can be tuned by means of a gate voltage. Our results show that the NPG-VO\textsubscript{2} photodetector can reach a large responsivity \( R \sim 10^5 \) V/W, a detectivity \( D^* \sim 10^{10} \) Jones, and a sensitivity in terms of NEP \( \text{NEP} \sim 10 \) fW/\( \sqrt{\text{Hz}} \) close to room temperature by taking advantage of the phase change of a thin VO\textsubscript{2} layer. The NPG sheet achieves an absorption of nearly 100\% due to localized surface plasmons (LSPs) around the patterned circular holes in a hexagonal 

Figure 4.6: Responsivity of the NPG-VO\textsubscript{2} gradient photodetector as a function of Fermi energy \( E_F \) of the NPG.
lattice symmetry. The electrostatic gate potential can be used to tune the wavelength peak in the MWIR and LWIR regimes between 3 and 12 $\mu$m. Our COMSOL simulations show that the NPG-VO$_2$ photodetector is able to operate on a time scale of 1 ms. Our proposed mid-IR photodetector reaches detectivities of cryogenically cooled HgCdTe photodetectors and sensitivities close to VO$_2$ microbolometers while operating close to room temperature.
In this chapter, we study a spectrally selective near-IR photodetection based on nanopatterned multilayer graphene (NPMLG) intercalated with FeCl$_3$, enabling large modulation p-doping of NPMLG.\(^1\)

Introduction

Because of the weak absorbance of pristine graphene of around 2%, we created nanopatterned CVD-grown single-layer graphene (NPG) that exhibits absorbance exceeding 60% in the long-wavelength infrared (LWIR) regime between $\lambda = 8 \ \mu\text{m}$ to $12 \ \mu\text{m}$.\([82, 80]\) Recently, we showed that NPG with smaller sizes of hexagonally arranged holes and smaller lattice constants exhibits absorbance of 80% in the mid-wavelength (MWIR) regime between $\lambda = 3 \ \mu\text{m}$ and $8 \ \mu\text{m}$. Due to the resolution limit of e-beam lithographic systems it is currently impossible to create smaller nanopatterns for increasing the absorbance of graphene using nanopatterning at shorter wavelengths.

Here, we show that LSP resonances can be realized in the technologically relevant NIR and SWIR regimes between $\lambda = 1.3 \ \mu\text{m}$ and $3 \ \mu\text{m}$ by means of nanopatterned multilayer graphene (NPMLG) intercalated with FeCl$_3$. Multilayer graphene intercalated with ferric chloride FeCl$_3$, dubbed graphexeter, was created for realizing an all-graphene photodetector which operates at around $6 \ \mu\text{m}$.\([103]\) The intriguing proximity effect of single layers of FeCl$_3$ between the graphene sheets is

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\(^1\)Plasmonically enhanced tunable spectrally selective NIR and SWIR photodetector based on intercalation doped nanopatterned multilayer graphene, Muhammad Waqas Shabbir & Michael N. Leuenberger, 2021. Manuscript submitted for publication.
to p-dope the graphene sheets to a Fermi energy of $E_F = -0.6$ eV due to the large work function of FeCl$_3$ of $W_{FeCl_3} = 5.1$ eV and the resulting charge transfer between FeCl$_3$ and graphene, which has a work function of around $W_g = 4.6$ eV.[11, 46] Remarkably, the bandstructures of graphene and FeCl$_3$ remain completely decoupled due to the incommensurate lattice structures, i.e. their lattice constants are 2.46 Å and 6.06 Å, respectively.[112] According to Ref. Zhan2010, MLG-FeCl$_3$ has a gap of 1.2 eV, to which the Fermi energy can be tuned by n-doping, corresponding to a wavelength of 1.03 µm, and is therefore transparent in the NIR and SWIR regimes between $\lambda = 1.3$ µm and 3 µm. The refractive index of FeCl$_3$ is $n = 1.365$, which gives a dielectric constant of $\varepsilon = n^2 = 1.86$. By creating a hexagonal nanopattern of holes inside the multilayer graphene/FeCl$_3$-intercalated heterostructure (NPMLG-FeCl$_3$), we show that it is possible to achieve LSP resonances in the NIR and SWIR regimes between $\lambda = 1.3$ µm and 3 µm with absorbances of nearly 100%. The method to tune the spectrally selective absorbance in NPMLG by means of a gate voltage $V_g$ is based on the fact that $V_g$ varies the Fermi energy $E_F$ inside NPMLG, thereby varying the charge density and therefore resonance wavelength of the LSPs around the circular holes in the wavelength regime between 1.3 µm and 3 µm.

Taking advantage of the special properties of FeCl$_3$, we present the proof of concept for a NIR photodetector based on NPMLG intercalated with FeCl$_3$.

Schematic design

Fig. 5.1 shows the schematic of our envisioned NPMLG-FeCl$_3$ photodetector. When the incident NIR or SWIR light field is maximized at the position of the NPMLG-FeCl$_3$ heterostructure by means of the Au mirror and also in resonance with the LSPs around the holes of the NPMLG, the NIR or SWIR light gets strongly absorbed with absorbance exceeding 95%, as shown in Fig. 5.2. Subsequently, the LSPs decay by creating hot carriers inside the graphene sheets due to boundary-
assisted intraband Landau damping.[110] While it is possible to detect the NIR or SWIR light by means of the bolometric effect relying on the change in conductance through NPG, this method is relatively slow because the lattice of NPG needs to be heated by means of the electron-phonon interaction before a signal can be detected, which is typically of the order of 1 ms.[85] In order to reduce the response time to about 100 ns, we choose to nanopattern only about half of the area of each graphene sheet, as shown in Fig. 5.1. Due to the asymmetric heating inside each partially nanopatterned graphene, a temperature gradient is created, which leads to the diffusion of the hot carriers from the nanopatterned side to the pristine side of the graphene sheet. This charge motion yields a Seebeck voltage across the Au source-drain contacts, giving rise to the plasmonically enhanced photothermoelectric effect inside each partially nanopatterned graphene sheet. Due to the record-high temperature difference $\Delta T = 5$ K, our envisioned NPMLG-FeCl$_3$ NIR and SWIR photodetectors exhibit extraordinarily large responsivity of $R = 6300$ V/W and detectivity of
$D^* = 2.33 \times 10^9 \text{ Jones.}$

The role of intercalated layers

As mentioned above, due to limited resolution of e-beam lithography it is currently impossible to realize LSP resonances in NPG in the NIR or SWIR. A natural way to increase the plasmon frequency is to increase the charge density of the material. In graphene this can be achieved by stacking several graphene sheets on top of each other. While for small twist angles between $\theta = 0^\circ$ up to about $15^\circ$, twisted bilayer, trilayer, and multilayer graphene exhibit interlayer coupling, resulting in e.g. electrostatically tunable band gaps in bilayer graphene\cite{114} at $\theta = 0$ and exotic many-body correlations, such as superconductivity, in twisted bilayer graphene at magic angle $\theta = 1.1^\circ$,\cite{15} twist angles around $\theta = 30^\circ$ in twisted bilayer graphene suppress completely interlayer coupling due to the mismatch of the k-space locations of the Dirac cones of the two layers.\cite{100} At first, such incommensurate stackings of graphene layers might look like being ideal candidates for increasing the plasmon frequency of the LSPs. However, the electrostatic doping of all the incommensurate graphene layers is impossible due to screening and also due to electronic decoupling of the layers.\cite{41}

In the seminal paper by Withers et al.\cite{103} the authors studied the optoelectronic properties of graphene/FeCl$_3$-intercalated few layer graphene, which they called graphexeter. They demonstrated a photodetector based on the photothermoelectric effect with a responsivity of $\mathcal{R} = 1.\infty \text{ V/W}$ at a wavelength of around $\lambda = 6 \mu\text{m}$. Interest in such intercalation doped multilayer graphene heterostructures has been recently rekindled by the realization of modulation doping of multilayer graphene by means of $\alpha$-RuCl$_3$.\cite{101} Due to the large work function of $\alpha$-RuCl$_3$, $W_{\alpha-\text{RuCl}_3} = 6.1 \text{ eV}$, substantial p-doping of all graphene layers of around $E_F = -0.8 \text{ eV}$ can be achieved without electrostatic gating. There are conflicting reports on the band gap of RuCl$_3$. In Ref. Reschke2017
Figure 5.2: Absorbance $A(\lambda)$ of the NPMLG-FeCl$_3$ heterostructure shown in Figs. 5.1 with Fermi energy $E_F = -1.0$ eV, mobility $\mu = 1500$ V/cm$^2$s, hole diameter of $a = 40$ nm and period $P = 60$ nm at $T = 300$ K. The results are obtained by FDTD calculations. (a) For a single NPG sheet the LSP resonance is at around 4.5 $\mu$m. At $N = 5$, the LSP resonance is at 1.55 $\mu$m. At $n = 8$, the LSP resonance is at 1.30 $\mu$m. (b) The LSP resonance gets shorter and shorter with larger number $N$ of NPG layers. (c) The full width at half maximum (FWHM) of the LSP resonance gets smaller and smaller for larger $N$.

IR reflectivity and transmission measurements reveal a band gap of 200 meV, which agrees with the energies of spin-orbit excitons measured in Ref. Warzanowski2020. However, photoemission and inverse photoemission spectroscopies find a much larger band gap of 1.9 eV.[89]

Since RuCl$_3$ has an optical band gap of the order of 200 meV,[72] here we focus on NPMLG-FeCl$_3$ for a NIR and SWIR photodetector based on the plasmonically enhanced photothermoelectric effect, which is shown in Fig. 5.1. The MLG channel length, which is the distance between the Au
source and drain contacts, is chosen to be \( L = 10 \ \mu m \), of the same order as the diffusion length of charge carriers in graphene.[90] The MLG channel width is chosen to be \( W = 10 \ \mu m \) as well because carrier collection does not need to be enhanced. The optimized hexagonal nanopattern on about half of the MLG-FeCl\(_3\) heterostructure (see Fig. 5.1) has a period of 60 nm and a hole diameter of 40 nm.

**Numerical simulations**

In order to perform the finite-difference time domain (FDTD) calculations, we use the linear dispersion relation of graphene, which gives rise to the intraband optical conductivity[82, 68]

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

which in the case of \( \epsilon_F \gg k_B T \) is reduced to

\[
\sigma_{\text{intra}}(\omega) = \frac{e^2}{\pi \hbar^2} \frac{E_F}{\tau - i\omega} = \frac{2\epsilon_m \omega_p^2}{\pi \hbar^2 (\tau - i\omega)},
\]

where \( \tau \) is determined by impurity scattering and electron-phonon interaction \( \tau^{-1} = \tau_{\text{imp}}^{-1} + \tau_{\text{e-ph}}^{-1} \). Using the mobility \( \mu \) of the NPG sheet, it can be presented in the form \( \tau^{-1} = \frac{e v_F^2}{\mu E_F} \), where \( v_F = 10^6 \) m/s is the Fermi velocity in graphene. \( \omega_p = \sqrt{e^2 E_F/2\epsilon_m} \) is the bulk graphene plasma frequency. Since the graphene sheets are electronically decoupled from each other by the insulating FeCl\(_3\) layers, the optical conductivity of MLG-FeCl\(_3\) is given by

\[
\sigma_{\text{intra}}^{\text{MLG-FeCl}_3}(\omega) = N \frac{e^2}{\pi \hbar^2} \frac{E_F}{\tau - i\omega},
\]
where $N$ is the number of graphene layers. This formula is only valid for excitation energies below the band gap $E_g = 1$ eV of FeCl$_3$. Since the LSP resonances occur at energies between 250 meV and 950 meV, we can safely neglect the optical phonons in graphene at 200 meV and the optical phonons in FeCl$_3$ at 2.7 meV, 7 meV.

Figure 5.3: Absorbance as a function of Fermi energy $E_F$ for NPMLG-FeCl$_3$ (a) containing 5 layers of graphene, the LSP resonance occurs at $\lambda = 1.55 \mu$m. (b) The tunable LSP resonance for Fermi energy between $E_F = -0.5$ eV and -1.0 eV. for 5-layers (c) containing 8 layers of graphene, the LSP resonance occurs at $\lambda = 1.30 \mu$m. (d) The tunable LSP resonance of the 8-graphene layer structure for Fermi energy between $E_F = -0.5$ eV and -1.0 eV.
We use FDTD to calculate the absorbance as a function of Fermi energy $E_F$ for NPMLG-FeCl$_3$ containing 5 and 8 layers of graphene, as shown in Fig. 5.3. The LSP resonances for the 5-graphene layer and 8-graphene layer structures exhibit wide tunability as a function of the Fermi energy $E_F$. Compared to our previous results on NPG, the absorbance remains above 80% even for lower Fermi energies of $E_F = -0.5$ eV. This large absorbance gives rise to strong heating of the nanopatterned side of the NPMLG structure.

Seebeck voltage

Using COMSOL, we obtain the temperature distribution inside the NPMLG-FeCl$_3$ heterostructure, as shown in Fig. 5.4. The photothermoelectric effect in the NPMLG-FeCl$_3$ heterostructure relies on large difference in absorbance between the patterned and the unpatterned side, i.e. the patterned side exhibits a plasmonically enhanced absorbance of nearly 100% while the unpatterned side absorbs only about 2% of the incident IR light. This results in a temperature gradient across the channel length $\Delta X = X_L - X_R$, where $X_L$ and $X_R$ are the edge positions of the left and right Au contacts, respectively. The hot carriers created on the patterned side due to Landau damping diffuse to the unpatterned side, resulting in a charge separation and Seebeck voltage

$$V_S = \int_{X_L}^{X_R} S(x) \frac{\partial T_e(x)}{\partial x} dx,$$

(5.4)

where $S(x)$ is the Seebeck coefficient, which has two values $S_{\text{pat}}$ and $S_{\text{unpat}}$ on the patterned and unpatterned side, respectively. $T_e(x)$ is the temperature profile of the charge carriers across the channel length, as shown in Fig. 5.4 (b). The Seebeck coefficient is approximated well by Mott’s formula

$$S = \frac{\pi^2 k_B^2 T}{3e} \frac{\partial \ln \sigma}{\partial E_F},$$

(5.5)
where $\sigma$, $k_B$, and $e$ are the electrical conductivity, Boltzmann constant, and elementary charge, respectively. $\sigma$ and $S$ depend on the $E_F$.

Figure 5.4: COMSOL simulated temperature gradient and Seebeck voltage generated by the plasmonically enhanced photothermoelectric effect in NPMLG-FeCl$_3$ heterostructure containing $N = 5$ graphene layers for the LSP resonance at $\lambda = 1.55$ $\mu$m. The channel length and width are both 10 $\mu$m. For an IR light incident power of $P_{\text{inc}} = 120$ nW a temperature difference of $\Delta T = 5.38$ K (a,b) and a Seebeck voltage of $\Delta V = 0.78$ mV (c,d) are achieved.

Starting from room temperature at $T = 300$ K and an incident power of the IR light of $P_{\text{inc}} = 120$ nW we obtain a temperature difference of $\Delta T = 5.38$ K and a Seebeck voltage of $V_S = 0.78$ mV. The Fermi energy of the graphene layers is kept at $E_F = -1.0$ eV, close to the intrinsic p-doping level due to the intercalation with FeCl$_3$. A bias voltage of $V_b = \pm 0.6$ V is applied for measuring the photocurrents in both directions. Owing to the bias voltage, both plasmonically enhanced photothermoelectric and bolometric effects contribute. In order to remove the bolometric
effect for the measurements, the dark and light Seebeck voltages $V_{S,d}$ and $V_{S,l}$ are calculated in the absence and presence of the incident IR light, respectively. The Seebeck voltage is then calculated by $V_S = V_{S,d} - V_{S,l}$. For a constant Fermi energy $E_F$, a DC bias voltages $+V_b$ and $-V_b$ are applied for two separate simulations across the channel width, which yields the currents $I_> = I + I_S$ and $I_< = -I + I_S$, respectively. $I$ is the current driven by the bias voltage and $I_S = (I_> + I_<)/2$ is the Seebeck current, which is captured in the absence ($I_{S,d}$) and presence ($I_{S,l}$) of the incident IR light. Since the holes are circularly symmetric, $I_S$ is independent of the polarization of the IR light. The Seebeck current and voltage are then calculated by $I_S = I_{S,l} - I_{S,d}$ and $V_S = R I_S$, respectively, where $R$ is the resistance of the NPMLG-FeCl$_3$ heterostructure. For the incident IR light, we assume a Gaussian beam with spot size radius of $R_{\text{spot}} = 2$ mm, a focus size radius of $R_f = \sqrt{(x-x_f)^2 + (y-y_f)^2}$, and an incident power of $P_{\text{inc}} = 120$ nW. The heat flux through the edges of the holes in the nanopattern is then

$$q_0 = \frac{2P_{\text{inc}}}{\pi R_{\text{spot}}^2} e^{-2R_f^2/R_{\text{spot}}^2}.$$  \hfill (5.6)

For an absorbance $A$ obtained from FDTD, the absorbed heat flux is determined by $q_A = Aq_0$.

Responsivity and detectivity

The responsivity is obtained by

$$\mathcal{R} = \frac{V_S}{P_{\text{inc}}}.$$  \hfill (5.7)

The responsivity as a function of Fermi energy $E_F$ is shown in Fig. 5.5. $\mathcal{R}$ ranges from a minimum of about $2.6 \times 10^3$ V/W for $E_F = -0.5$ eV to a maximum of about $6.3 \times 10^3$ V/W for $E_F = -1.0$ eV. Assuming a typical noise equivalent power of $\text{NEP} = 7$ pW/Hz$^{1/2}$ for CVD graphene,[79] we obtain a maximum detectivity of $D^* = \sqrt{LW/\text{NEP}} = 0.74 \times 10^9$ Jones. If we scale the channel
width to $W = 200 \, \mu m$, it is possible to achieve $D^* = 2.33 \times 10^9$ Jones.

Figure 5.5: COMSOL simulated temperature gradient and Seebeck voltage generated by the plasmonically enhanced photothermoelectric effect in NPMLG-FeCl$_3$ heterostructure containing $N = 5$ graphene layers for the LSP resonance at $\lambda = 1.55 \, \mu m$. The channel length and width are both $10 \, \mu m$. For an IR light incident power of $P_{inc} = 120 \, nW$ a temperature difference of $\Delta T = 5.38 \, K$ (a,b) and a Seebeck voltage of $\Delta V = 0.78 \, mV$ (c,d) are achieved.

Conclusion

In conclusion, we have demonstrated in our theoretical study that NPMLG-FeCl$_3$ can be used to develop a plasmonically enhanced NIR and SWIR photodetector with spectrally tunable selective light absorption. Most importantly, the LSPs along with an optical cavity increase substantially the absorbance of graphene from about 2% for pristine graphene to nearly 100% for NPMLG, thereby
outperforming state-of-the-art graphene-based photodetector. Remarkably, by taking advantage of NPMLG-FeCl$_3$ with an arbitrary number of graphene layers $N$ it is possible to develop photodetectors that operate over a wide wavelength range from $\lambda = 1.3$ $\mu$m down to $\lambda = 12$ $\mu$m and beyond, covering the NIR, SWIR, MWIR, and LWIR regimes. In the future it would be interesting to study LSPs in NPG also in the THz regime. Our proposed IR photodetector can be used to develop an IR spectroscopy and detection platform based on NPMLG that will be able to detect a variety of molecules that have IR vibrational resonances, such as CO, CO$_2$, NO, NO$_2$, CH$_4$, TNT, H$_2$O$_2$, acetone, TATP, Sarin, VX, viruses, etc.
CHAPTER 6: CONCLUSION AND FUTURE WORK

In this dissertation, following the techniques of enhancing light absorption in graphene sheet, we have presented different models involving graphene sheet for detection and emission of low energy photons in mid-IR regime. In Chapter 2 we have demonstrated an outstanding photodetection process which depends on multiple phenomena, enhancement of tunable absorption in single layer graphene sheet, effect of mobility, thermal carriers generated by means of plasmons, and photothermoelectric effect. We have observed temperature difference of $4.7K$ between hot and cold edges of graphene sheet under the effect of incident power of $155nW$, which in turn produces a signal of $0.44 \text{ mV}$. The responsivity and detectivity of detector are $3000 \text{V/W}^{-1}$ and $10^9 \text{ Jones}$, respectively with an ultrafast time response of $100 \text{ ns}$.

In Chapter 3, we have demonstrated theoretically that NPG can be used to develop a plasmonically enhanced mid-IR light source with spectrally tunable selective thermal emission. Most importantly, the LSPs along with an optical cavity increase substantially the emittance of graphene from about 2% for pristine graphene to 80% for NPG, thereby outperforming state-of-the-art graphene light sources working in the visible and NIR by at least a factor of 100.

In Chapter 4 we have presented the model of an ultrasensitive mid-infrared (mid-IR) photodetector based on a heterostructure made of NPG and VO$_2$, thereby extending the responsivity of a VO$_2$ microbolometer to the LWIR domain. Moreover, this hybrid NPG-VO$_2$ photodetector has a narrowband absorption in the MWIR and LWIR that can be tuned by means of a gate voltage. Our results show that the NPG-VO$_2$ photodetector can reach a large responsivity $R \sim 10^4 \text{ V/W}$, a detectivity $D^* \sim 10^{10} \text{ Jones}$, and a sensitivity in terms of NEP $\text{NEP} \sim 100 \text{ fW}/\sqrt{\text{Hz}}$ close to room temperature by taking advantage of the phase change of a thin VO$_2$ layer. The NPG sheet achieves an absorption of nearly 100% due to localized surface plasmons (LSPs) around the pat-
terned circular holes in a hexagonal lattice symmetry. The electrostatic gate potential can be used to tune the wavelength peak in the MWIR and LWIR regimes between 3 and 12 microns, thereby overcoming the intrinsic upper limit of 6 microns for microbolometers based on VO\(_2\). Our COMSOL simulations show that the NPG-VO\(_2\) photodetector is able to operate on a time scale of 1 ms, much shorter than the response times of current microbolometers based on VO\(_2\) alone. Our proposed mid-IR photodetector reaches detectivities of cryogenically cooled HgCdTe photodetectors and sensitivities close to and field of view similar to VO\(_2\) microbolometers while operating close to room temperature.

In Chapter 4, we have demonstrated in our theoretical study that NPMLG-FeCl\(_3\) can be used to develop a plasmonically enhanced NIR and SWIR photodetector with spectrally tunable selective light absorption. Most importantly, the LSPs along with an optical cavity increase substantially the absorbance of graphene from about 2% for pristine graphene to nearly 100% for NPMLG, thereby outperforming state-of-the-art graphene-based photodetector. Remarkably, by taking advantage of NPMLG-FeCl\(_3\) with an arbitrary number of graphene layers \(N\) it is possible to develop photodetectors that operate over a wide wavelength range from \(\lambda = 1.3\ \mu m\) down to \(\lambda = 12\ \mu m\) and beyond, covering the NIR, SWIR, MWIR, and LWIR regimes.

In future, Combining our proposed mid-IR light source based on patterned graphene[86] with our demonstrated mid-IR detectors based on NPG[79] and NPMLG, we are going to develop a mid-IR spectroscopy and detection platform based on patterned graphene that will be able to detect a variety of molecules that have mid-IR vibrational resonances, such as CO, CO\(_2\), NO, NO\(_2\), CH\(_4\), TNT, H\(_2\)O\(_2\), acetone, TATP, Sarin, VX, etc. In particular, a recent study showed that it is possible to detect the hepatitis B and C viruses label-free at a wavelength of around 6 \(\mu m\).[77] Therefore, we will make great effort to demonstrate that our platform will be able to detect with high sensitivity and selectivity the viruses that pose a threat to humanity.
Ellipsoidal coordinates

For determining the EM properties of an infinitesimally thin conducting elliptical disk of radius $R$ or an infinitesimally thin conducting plane with a elliptical hole, including coated structures, it is most convenient to perform the analytical calculations in the ellipsoidal coordinate system $(\xi, \eta, \zeta)$,[1, 53, 51, 10] which is related to the Cartesian coordinate system through the implicit equation

$$\frac{x^2}{a^2 + u} + \frac{y^2}{b^2 + u} + \frac{z^2}{c^2 + u} = 1 \quad \text{(A.1)}$$

for $a > b > c$. The cubic roots $\xi$, $\eta$, and $\zeta$ are all real in the ranges

$$-a^2 \leq \zeta \leq -b^2, \quad -b^2 \leq \eta \leq -c^2, \quad -c^2 \leq \xi < \infty,$$  \quad \text{(A.2)}

which are the ellipsoidal coordinates of a point $(x, y, z)$. The surfaces of constant $\xi$, $\eta$, and $\zeta$ are ellipsoids, hyperboloids of one sheet, and hyperboloids of two sheets, respectively, all confocal with the ellipsoid defined by

$$\frac{x^2}{a^2} + \frac{y^2}{b^2} + \frac{z^2}{c^2} = 1. \quad \text{(A.3)}$$

Each point $(x, y, z)$ in space is determined by the intersection of three surfaces, one from each of the three families, and the three surfaces are orthogonal to each other. The transformation between the two coordinate systems is given by the solutions of Eq. (A.1), i.e.

$$x = \pm \sqrt{\frac{(\xi + a^2)(\eta + a^2)(\zeta + a^2)}{(b^2 - a^2)(c^2 - a^2)}}, \quad \text{(A.4)}$$

$$y = \pm \sqrt{\frac{(\xi + b^2)(\eta + b^2)(\zeta + b^2)}{(c^2 - b^2)(a^2 - b^2)}}, \quad \text{(A.5)}$$

$$z = \pm \sqrt{\frac{(\xi + c^2)(\eta + c^2)(\zeta + c^2)}{(a^2 - c^2)(b^2 - c^2)}}, \quad \text{(A.6)}$$
defining 8 equivalent octants. The length elements in ellipsoidal coordinates read

\[ dl^2 = h_1^2 d\xi^2 + h_2^2 d\eta^2 + h_3^2 d\zeta^2, \tag{A.7} \]

\[ h_1 = \sqrt{\frac{(\xi - \eta)(\xi - \zeta)}{2R_\xi}}, \tag{A.8} \]
\[ h_2 = \sqrt{\frac{(\eta - \zeta)(\xi - \zeta)}{2R_\eta}}, \tag{A.9} \]
\[ h_3 = \sqrt{\frac{(\zeta - \xi)(\zeta - \eta)}{2R_\zeta}}, \tag{A.10} \]

\[ R_u^2 = (u + a^2)(u + b^2)(u + c^2), \; u = \xi, \eta, \zeta. \tag{A.11} \]

For the transformation from cartesian to ellipsoidal coordinates, one can use the following system of equations:

\[ \dot{\xi} = \frac{\frac{\partial x}{\partial \xi} \dot{x} + \frac{\partial y}{\partial \xi} \dot{y} + \frac{\partial z}{\partial \xi} \dot{z}}{\sqrt{\left(\frac{\partial x}{\partial \xi}\right)^2 + \left(\frac{\partial y}{\partial \xi}\right)^2 + \left(\frac{\partial z}{\partial \xi}\right)^2}}, \tag{A.12} \]

\[ \dot{\eta} = \frac{\frac{\partial x}{\partial \eta} \dot{x} + \frac{\partial y}{\partial \eta} \dot{y} + \frac{\partial z}{\partial \eta} \dot{z}}{\sqrt{\left(\frac{\partial x}{\partial \eta}\right)^2 + \left(\frac{\partial y}{\partial \eta}\right)^2 + \left(\frac{\partial z}{\partial \eta}\right)^2}}, \tag{A.13} \]

\[ \dot{\zeta} = \frac{\frac{\partial x}{\partial \zeta} \dot{x} + \frac{\partial y}{\partial \zeta} \dot{y} + \frac{\partial z}{\partial \zeta} \dot{z}}{\sqrt{\left(\frac{\partial x}{\partial \zeta}\right)^2 + \left(\frac{\partial y}{\partial \zeta}\right)^2 + \left(\frac{\partial z}{\partial \zeta}\right)^2}}, \tag{A.14} \]
whose elements $J_{ij}$ define the Jacobian matrix. The derivatives are explicitly:

\[
\frac{\partial x}{\partial \xi} = \frac{1}{2} \sqrt{\frac{(a^2 + \eta)(a^2 + \zeta)}{(a^2 + \xi)(a^2 - b^2)(a^2 - c^2)}},
\]
\[
\frac{\partial x}{\partial \eta} = \frac{1}{2} \sqrt{\frac{(a^2 + \xi)(a^2 + \zeta)}{(a^2 + \eta)(a^2 - b^2)(a^2 - c^2)}},
\]
\[
\frac{\partial x}{\partial \zeta} = \frac{1}{2} \sqrt{\frac{(a^2 + \xi)(a^2 + \eta)}{(a^2 + \zeta)(a^2 - b^2)(a^2 - c^2)}},
\]
\[
\frac{\partial y}{\partial \xi} = \frac{1}{2} \sqrt{\frac{(b^2 + \eta)(b^2 + \zeta)}{(b^2 + \xi)(b^2 - a^2)(b^2 - c^2)}},
\]
\[
\frac{\partial y}{\partial \eta} = \frac{1}{2} \sqrt{\frac{(b^2 + \xi)(b^2 + \zeta)}{(b^2 + \eta)(b^2 - a^2)(b^2 - c^2)}},
\]
\[
\frac{\partial y}{\partial \zeta} = \frac{1}{2} \sqrt{\frac{(b^2 + \xi)(b^2 + \eta)}{(b^2 + \zeta)(b^2 - a^2)(b^2 - c^2)}},
\]
\[
\frac{\partial z}{\partial \xi} = \frac{1}{2} \sqrt{\frac{(c^2 + \eta)(c^2 + \zeta)}{(c^2 + \xi)(c^2 - a^2)(c^2 - b^2)}},
\]
\[
\frac{\partial z}{\partial \eta} = \frac{1}{2} \sqrt{\frac{(c^2 + \xi)(c^2 + \zeta)}{(c^2 + \eta)(c^2 - a^2)(c^2 - b^2)}},
\]
\[
\frac{\partial z}{\partial \zeta} = \frac{1}{2} \sqrt{\frac{(c^2 + \xi)(c^2 + \eta)}{(c^2 + \zeta)(c^2 - a^2)(c^2 - b^2)}}.
\]

The coordinate $\eta$ is constant on the surfaces of oblate spheroids defined by

\[
\frac{x^2 + y^2}{(R \cosh \eta)^2} + \frac{z^2}{(R \sinh \eta)^2} = 1
\]

The surface associated with the limit $\eta \to 0$ is an infinitesimally thin circular disk of radius $R$. In contrast, the surface in the limit $\eta \gg 1$ is a sphere of radius $r = R \cosh \eta \approx R \sinh \eta$. Thus, the
Laplace equation in ellipsoidal coordinates reads

\[ \Delta \Phi = \frac{4}{(\xi - \eta)(\xi - \zeta)(\zeta - \eta)} \left[ (\eta - \zeta) R_\xi \frac{\partial}{\partial \xi} \left( R_\xi \frac{\partial \Phi}{\partial \xi} \right) + (\zeta - \eta) R_\eta \frac{\partial}{\partial \eta} \left( R_\eta \frac{\partial \Phi}{\partial \eta} \right) + (\xi - \eta) R_\zeta \frac{\partial}{\partial \zeta} \left( R_\zeta \frac{\partial \Phi}{\partial \zeta} \right) \right] = 0. \]  
(A.25)

Charged conducting ellipsoid

The surface of the conducting ellipsoid is defined by \( \xi = 0 \). Thus, the electric field potential \( \Phi(\xi) \) is a function of \( \xi \) only, thereby defining the equipotential surfaces by confocal ellipsoids. Laplace’s equation is then simplified to

\[ \frac{d}{d\xi} \left( R_\xi \frac{d\Phi}{d\xi} \right) = 0. \]  
(A.26)

The solution outside the ellipsoid is

\[ \Phi_{\text{out}}(\xi) = A \int_\xi^\infty \frac{d\xi'}{R_{\xi'}}. \]  
(A.27)

From the asymptotic approximation \( \xi \approx r^2 \) for large distances \( r \rightarrow \infty \), i.e. \( \xi \rightarrow \infty \), we identify \( R_\xi \approx \xi^{3/2} \) and thus

\[ \Phi_{\text{out}}(\xi \rightarrow \infty) \approx \frac{2A}{\sqrt{\xi}} = \frac{2A}{r}. \]  
(A.28)

using the boundary condition \( \lim_{\xi \rightarrow \infty} \Phi(\xi) = 0 \). Since the Coulomb field should be \( \Phi(\xi \rightarrow \infty) \approx e/r \) at large distances from the ellipsoid, \( 2A = e \) and

\[ \Phi_{\text{out}}(\xi) = \frac{e}{2} \int_\xi^\infty \frac{d\xi'}{R_{\xi'}.} \]  
(A.29)

is obtained, corresponding to the far-field of a monopole charge.
The solution inside the ellipsoid is

\[
\Phi_{\text{in}}(\xi) = B \int_{-c}^{\xi} \frac{d\xi'}{R_{\xi'}}. \tag{A.30}
\]

Using the asymptotic approximation \( R_{\xi \to -c^2} \propto \sqrt{\xi + c^2} \) we obtain

\[
\Phi_{\text{in}}(\xi \to -c^2) \approx B \sqrt{\xi + c^2}. \tag{A.31}
\]

This solution satisfies the boundary condition \( \lim_{\xi \to -c^2} \Phi(\xi) = 0 \). The constant \( B \) can be found from the boundary condition \( \Phi(\xi = 0) = V \), where \( V \) is the potential on the surface of the charged ellipsoid. Thus, \( B = V/c \) and

\[
\Phi_{\text{in}}(\xi) = \frac{V}{c} \sqrt{\xi + c^2}. \tag{A.32}
\]

**Dipole moment of conducting ellipsoid induced by an external electric field in z-direction**

Following Ref. [10], let us consider the case when the external electric field is parallel to one of the major axes of the ellipsoid. For the external potential let us choose

\[
\Phi_0 = -E_0 z = -E_0 \sqrt{\frac{(\xi + c^2)(\eta + c^2)(\zeta + c^2)}{(a^2 - c^2)(b^2 - c^2)}}. \tag{A.33}
\]

Let \( \Phi_p \) be the potential caused by the ellipsoid, with the boundary condition \( \Phi_p(\xi \to \infty) = 0 \). Requiring continuous boundary condition on the surface of the ellipsoid, we have

\[
\Phi_{\text{in}}(0, \eta, \zeta) = \Phi_0(0, \eta, \zeta) + \Phi_p(0, \eta, \zeta). \tag{A.34}
\]
We make the ansatz
\[ \Phi_p(\xi, \eta, \zeta) = F_p(\xi)\sqrt{(\eta + c^2)(\zeta + c^2)}, \] (A.35)

which after insertion into the Laplace equation yields
\[ R_\xi \frac{d}{d\xi} \left[ R_\xi \frac{dF_p}{d\xi} \right] - \left( \frac{a^2 + b^2}{4} + \frac{\xi}{2} \right) F_p(\xi) = 0. \] (A.36)

Thus, one obtains for the field caused by the ellipsoid
\[ \Phi_p(\xi, \eta, \zeta) = C_p F_p(\xi)\sqrt{(\eta + c^2)(\zeta + c^2)}, \] (A.37)

with
\[ F_p(\xi) = F_{in}(\xi) \int_\xi^\infty \frac{d\xi'}{F_{in}^2(\xi')R_{\xi'}}, \] (A.38)

where
\[ F_{in}(\xi) = \sqrt{\xi + c^2}, \] (A.39)

the function we used in the case of the charged ellipsoid (see above). Thus, the field inside the ellipsoid is given by
\[ \Phi_{in} = C_{in} F_{in}(\xi)\sqrt{(\eta + c^2)(\zeta + c^2)}. \] (A.40)

Using the boundary condition shown in Eq. (A.34), one obtains the first equation
\[ C_p \int_0^\infty \frac{d\xi'}{(c^2 + \xi')R_{\xi'}} - C_{in} = \frac{E_0}{\sqrt{(a^2 - c^2)(b^2 - c^2)}}, \] (A.41)

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The boundary condition of the normal component of $D$ at $\xi = 0$, equivalent to

$$
\varepsilon_m \frac{\partial \Phi_{\text{in}}}{\partial \xi} = \varepsilon_m \frac{\partial \Phi_0}{\partial \xi} + \varepsilon_m \frac{\partial \Phi_p}{\partial \xi},
$$

yields the second equation

$$
\varepsilon_m C_p \left[ \int_0^\infty \frac{d\xi'}{(c^2 + \xi') R_{\xi'}} - \frac{2}{abc} \right] - \varepsilon_{\text{in}} C_{\text{in}} = \frac{\varepsilon_m E_0}{\sqrt{(a^2 - c^2)(b^2 - c^2)}},
$$

Consequently, the potentials are

$$
\Phi_{\text{in}} = \frac{\Phi_0}{1 + \frac{L_3(\varepsilon_{\text{in}} - \varepsilon_m)}{\varepsilon_m}},
$$

$$
\Phi_p = \frac{\Phi_0}{1 + \frac{L_3(\varepsilon_{\text{in}} - \varepsilon_m)}{\varepsilon_m}} \left[ \frac{abc \varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_m} \right] \frac{\int_0^\infty \frac{d\xi'}{(c^2 + \xi') R_{\xi'}}}{\xi},
$$

where

$$
L_3 = \frac{abc}{2} \int_0^\infty \frac{d\xi'}{(c^2 + \xi') R_{\xi'}}.
$$

Far away from the ellipsoid for $\xi \approx r^2 \to \infty$, one can use the approximation

$$
\int_\xi^\infty \frac{d\xi'}{(c^2 + \xi') R_{\xi'}} \approx \int_\xi^\infty \frac{d\xi'}{\xi^{5/2}} = \frac{2}{3} \xi^{-3/2},
$$

yielding the potential caused by the ellipsoid, i.e.

$$
\Phi_p \approx \frac{E_0 \cos \theta}{r^2} \frac{abc \varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_m} \frac{3}{1 + \frac{L_3(\varepsilon_{\text{in}} - \varepsilon_m)}{\varepsilon_m}},
$$

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from which we identify the dipole moment

\[ p = p \hat{z} = 4\pi \varepsilon_m abc \frac{\varepsilon_{in} - \varepsilon_m}{3\varepsilon_m + 3L_3(\varepsilon_{in} - \varepsilon_m)} E_0 \hat{z}. \]  

(A.49)

This result determines the polarizability of the charged ellipsoid, i.e.

\[ \alpha_3 = 4\pi \varepsilon_m abc \frac{\varepsilon_{in} - \varepsilon_m}{3\varepsilon_m + 3L_3(\varepsilon_{in} - \varepsilon_m)} \]  

(A.50)

If the external electric field is applied along the other major axes of the ellipsoid, \( x \) or \( y \), the polarizabilities are

\[ \alpha_1 = 4\pi \varepsilon_m abc \frac{\varepsilon_{in} - \varepsilon_m}{3\varepsilon_m + 3L_1(\varepsilon_{in} - \varepsilon_m)}, \]  

(A.51)

\[ \alpha_2 = 4\pi \varepsilon_m abc \frac{\varepsilon_{in} - \varepsilon_m}{3\varepsilon_m + 3L_2(\varepsilon_{in} - \varepsilon_m)}, \]  

(A.52)

respectively, where

\[ L_1 = \frac{abc}{2} \int_0^\infty \frac{d\xi'}{(a^2 + \xi')R_{\xi'}}, \]  

(A.53)

\[ L_2 = \frac{abc}{2} \int_0^\infty \frac{d\xi'}{(b^2 + \xi')R_{\xi'}}, \]  

(A.54)

For oblate spheroids \( a = b \), \( L_1 = L_2 \),

\[ L_1 = \frac{g(e_o)}{2e_o^2} \left[ \frac{\pi}{2} - \arctan g(e_o) \right] - \frac{g^2(e_o)}{2}, \]

\[ g(e_o) = \sqrt{1 - \frac{e_o^2}{e_o^2}}, \quad e_o^2 = 1 - \frac{c^2}{a^2}, \]  

(A.55)
where $e_o$ is the eccentricity of the oblate spheroid. The limiting cases of an infinitesimally thin disk and a sphere are obtained for $e_o = 1$ and $e_o = 0$, respectively.

The geometrical factors $L_i$ are related to the depolarization factors $\hat{L}_i$ by

\[
E_{\text{in}x} = E_{0x} - \hat{L}_1 P_{\text{in}x}, \quad (A.56)
\]
\[
E_{\text{in}y} = E_{0y} - \hat{L}_2 P_{\text{in}y}, \quad (A.57)
\]
\[
E_{\text{in}z} = E_{0z} - \hat{L}_3 P_{\text{in}z}, \quad (A.58)
\]

with

\[
\hat{L}_i = \frac{\varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_{\text{in}} - \varepsilon_0 \varepsilon_m} L_i. \quad (A.59)
\]

Dipole moment of conducting ellipsoid induced by an external electric field in $x$-direction

In analogy to Ref. [10], let us consider the case when the external electric field is parallel to one of the major axes of the ellipsoid, in this case along the $x$-axis. For the external potential let us choose

\[
\Phi_0 = -E_{0x} = -E_0 \sqrt{\frac{(\xi + a^2)(\eta + a^2)(\zeta + a^2)}{(b^2 - a^2)(c^2 - a^2)}}. \quad (A.60)
\]

Let $\Phi_p$ be the potential caused by the ellipsoid, with the boundary condition $\Phi_p(\xi \to \infty) = 0$. Requiring continuous boundary condition on the surface of the ellipsoid, we have

\[
\Phi_{\text{in}}(0, \eta, \zeta) = \Phi_0(0, \eta, \zeta) + \Phi_p(0, \eta, \zeta). \quad (A.61)
\]

Thus, one obtains for the field caused by the ellipsoid

\[
\Phi_p(\xi, \eta, \zeta) = C_p F_p(\xi) \sqrt{(\eta + a^2)(\zeta + a^2)} \quad (A.62)
\]
with

\[ F_p(\xi) = F_{\text{in}}(\xi) \int_{\xi}^{\infty} \frac{d\xi'}{F_{\text{in}}^2(\xi') R_{\xi'}}, \]  

(A.63)

where

\[ F_{\text{in}}(\xi) = \sqrt{\xi + a^2}, \]  

(A.64)

the function we used in the case of the charged ellipsoid (see above). Thus, the field inside the ellipsoid is given by

\[ \Phi_{\text{in}} = C_{\text{in}} F_{\text{in}}(\xi) \sqrt{(\eta + a^2)(\zeta + a^2)}. \]  

(A.65)

Using the boundary condition shown in Eq. (A.61), one obtains the first equation

\[ C_p \int_{0}^{\infty} \frac{d\xi'}{(a^2 + \xi') R_{\xi'}} - C_{\text{in}} = \frac{E_0}{\sqrt{(b^2 - a^2)(c^2 - a^2)}}, \]  

(A.66)

The boundary condition of the normal component of \( \mathbf{D} \) at \( \xi = 0 \), equivalent to

\[ \varepsilon_{\text{in}} \frac{\partial \Phi_{\text{in}}}{\partial \xi} = \varepsilon_m \frac{\partial \Phi_0}{\partial \xi} + \varepsilon_m \frac{\partial \Phi_p}{\partial \xi}, \]  

(A.67)

yields the second equation

\[ \varepsilon_m C_p \left[ \int_{0}^{\infty} \frac{d\xi'}{(a^2 + \xi') R_{\xi'}} - \frac{2}{abc} \right] - \varepsilon_{\text{in}} \varepsilon_m \frac{E_0}{\sqrt{(b^2 - a^2)(c^2 - a^2)}} \]  

\[ = \frac{\varepsilon_m E_0}{\sqrt{(b^2 - a^2)(c^2 - a^2)}}, \]  

(A.68)

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Consequently, the potentials are

\[
\Phi_{in} = \Phi_0 \left(1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}\right),
\]

(A.69)

\[
\Phi_p = \Phi_0 \frac{abc \varepsilon_{m} - \varepsilon_{in}}{2 \varepsilon_m} \int_0^\infty \frac{d\xi'}{(a^2 + \xi')(R_{\xi'})} \left(1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}\right),
\]

(A.70)

where

\[
L_1 = \frac{abc}{2} \int_0^\infty \frac{d\xi'}{(a^2 + \xi')(R_{\xi'})}.
\]

(A.71)

Far away from the ellipsoid for \(\xi \approx r^2 \to \infty\), one can use the approximation

\[
\int_\xi^\infty \frac{d\xi'}{(a^2 + \xi')(R_{\xi'})} \approx \int_\xi^\infty \frac{d\xi'}{\xi'^{5/2}} = \frac{2}{3} \xi^{-3/2},
\]

(A.72)

yielding the potential caused by the ellipsoid, i.e.

\[
\Phi_p \approx \frac{E_0 \cos \theta}{r^2} \frac{abc \varepsilon_{m} - \varepsilon_{in}}{3 \varepsilon_m} \frac{3}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}},
\]

(A.73)

from which we identify the dipole moment

\[
p = p\dot{x} = 4\pi \varepsilon_m abc \varepsilon_{in} - \varepsilon_m \left(3 \varepsilon_m + 3L_1(\varepsilon_{in} - \varepsilon_m)\right)E_0 \cdot \dot{x}.
\]

(A.74)

This result determines the polarizability of the charged ellipsoid, i.e.

\[
\alpha_1 = 4\pi \varepsilon_m abc \frac{\varepsilon_{in} - \varepsilon_m}{3 \varepsilon_m + 3L_1(\varepsilon_{in} - \varepsilon_m)}
\]

(A.75)
Dipole moment of conducting single-sheet hyperboloid with a small elliptical wormhole induced by an external electric field

Let us consider a conducting single-sheet hyperboloid with a small elliptical wormhole, as shown in Fig. A.1. Contrary to the case of an uncharged ellipsoid, where the solutions when applying the external electric field in $x$, $y$, or $z$ direction are similar, the solutions in the case of an uncharged hyperboloid depend strongly on the axis in which the external field $E$ points. While the solutions for $E = E_0 \hat{x}$ and $E = E_0 \hat{y}$ are similar, the solution for $E = E_0 \hat{z}$ is completely different. The reason for this fundamental difference is that the ellipsoid resembles a sphere from far away. However, a single-sheet hyperboloid has elliptical cylindrical symmetry.

Here, let us first calculate the electrostatic potential $\Phi(\xi, \eta, \zeta)$ of a conducting single-sheet hyperboloid with an elliptical hole, which can be represented by a limiting hyperboloid from a family of
hyperboloids described by the implicit equation

$$\frac{x^2}{a^2 + u} + \frac{y^2}{b^2 + u} + \frac{z^2}{c^2 + u} = 1$$  \hspace{1cm} (A.76)

for \( a > b > c \). The cubic roots \( \xi, \eta, \) and \( \zeta \) are all real in the ranges

$$-a^2 \leq \zeta \leq -b^2, \quad -b^2 \leq \eta \leq -c^2, \quad -c^2 \leq \xi < \infty,$$  \hspace{1cm} (A.77)

which are the ellipsoidal coordinates of a point \((x, y, z)\). The limiting hyperboloid is a single planar sheet with an elliptical hole, i.e. it belongs to the family of solutions \( \eta \) in the limit \( \eta \to -c^2 \). Therefore, let us choose this limiting case as our origin in ellipsoidal coordinates with \( c = 0 \). Then Eq. (A.76) becomes

$$\frac{x^2}{a^2 + u} + \frac{y^2}{b^2 + u} + \frac{z^2}{u} = 1$$  \hspace{1cm} (A.78)

for \( a > b > c = 0 \). The cubic roots \( \xi, \eta, \) and \( \zeta \) are all real in the ranges

$$-a^2 \leq \zeta \leq -b^2, \quad -b^2 \leq \eta \leq 0, \quad 0 \leq \xi < \infty,$$  \hspace{1cm} (A.79)

The surface of the conducting hyperboloid is defined by \(-b^2 \leq \eta = \eta_1 < 0\).

Let us consider the case \( E_0 = E_0 \hat{x} \), which in the limit when the hyperboloid becomes a flat plane is the most relevant one. Therefore

$$\Psi_0 = -E_0 x = \pm E_0 \sqrt{\frac{(\xi + a^2)(\eta + a^2)(\zeta + a^2)}{(b^2 - a^2)(-a^2)}}$$  \hspace{1cm} (A.80)

in the lower-half plane, where the negative sign corresponds to positive \( x \) values and the positive sign to negative \( x \) values. Since the equipotential surfaces are determined by \( \eta \), let \( \Psi_p \) be the potential caused by the hyperboloid, with the boundary condition \( \Psi_{in}(\eta = 0) = 0 \). Requiring
continuous boundary condition on the surface of the hyperboloid, we have

\[ \Psi_{\text{in}}(\xi, \eta_1, \zeta) = \Psi_0(\xi, \eta_1, \zeta) + \Psi_p(\xi, \eta_1, \zeta), \quad (A.81) \]

\[ \varepsilon_{\text{in}} \frac{\partial \Psi_{\text{in}}}{\partial \eta} \bigg|_{\eta_1} = \varepsilon_m \frac{\partial \Psi_0}{\partial \eta} \bigg|_{\eta_1} + \varepsilon_m \frac{\partial \Psi_p}{\partial \eta} \bigg|_{\eta_1}, \quad (A.82) \]

where in the second equation the normal component of \( D \) at \( \eta = \eta_1 \) must be continuous. Then we make the ansatz for the electrostatic potential inside the hyperboloid,

\[ \Psi_{\text{in}}(\xi, \eta, \zeta) = -C_{\text{in}} E_0 x, \quad (A.83) \]

where \( C_{\text{in}} \) is a constant. This ansatz satisfies the boundary condition \( \Psi_{\text{in}}(\xi = 0, \eta \to 0, \zeta) = 0 \).

For the outside polarization field we choose

\[ \Psi_p(\xi, \eta, \zeta) = -C_p E_0 x F_1(\xi) K_1(\eta), \quad (A.84) \]

where \( C_p \) is a constant, and we defined

\[ F_1(\xi) = \int_\xi^\infty \frac{ad\xi'}{2\xi'^{1/2}(\eta + a^2)} - \int_\xi^\infty \frac{ad\xi'}{2(\xi' + a^2)^{3/2}}, \]

\[ = \arctan \left( \frac{a}{\sqrt{\xi}} \right) - \frac{a}{\sqrt{\xi} + a^2}. \quad (A.85) \]

Note that \( \lim_{\xi \to 0^+} \arctan \left( \frac{a}{\sqrt{\xi}} \right) = \pi/2 \), whereas \( \lim_{\xi \to 0^-} \arctan \left( \frac{a}{\sqrt{\xi}} \right) = -\pi/2 \). Therefore, in order to avoid discontinuity at \( \xi = 0 \), we must have \( \arctan \left( \frac{a}{\sqrt{\xi}} \right) = \pi - \arctan \left( \frac{a}{\sqrt{\xi}} \right) \).

\[ K_1(\eta) = \int_\eta^\infty \frac{d\eta'}{(\eta' + a^2)R_{\eta'}}, \quad (A.86) \]
where \( R_\eta = \sqrt{(\eta + a^2)(\eta + b^2)(-\eta)} \). The boundary conditions at \( z \to \pm\infty \) are satisfied:

\[
F_1(\xi) = \begin{cases} 
0 & \text{for } z \to +\infty \\
\pi & \text{for } z \to -\infty
\end{cases}
\]  

(A.87)

At large distances \( r = \sqrt{x^2 + y^2 + z^2} \) from the wormhole we have \( \xi \approx r^2 \). Then the far-field potential in the upper half-space, which is given by the pure polarization field, is

\[
\Psi_p(\xi, \eta, \zeta) \approx -C'_pE_0\frac{a^3}{3}\frac{x}{r^3}.
\]  

(A.88)

The polarization far-field has the form of a dipole field at large distances \( r \) from the wormhole.

In order to determine the polarizability of the wormhole, let us find the solution at \( \xi = 0 \), corresponding to the plane that passes through the center of the wormhole. For \( \xi = 0 \), the unit vectors \( \hat{x} \) and \( \hat{\eta} \) are parallel. In this near-field limit, the polarization potential has the form

\[
\Psi_p(\xi, \eta, \zeta) = -C'_{p_0}E_0xK_1(\eta),
\]  

(A.89)

where \( C'_{p_0} = C_p(\pi/2 - 1) \).

Using the boundary conditions shown in Eq. (A.82), we obtain the first equation

\[
\tilde{C}_pK_1(\eta_1) - C_{\text{in}} = 1,
\]  

(A.90)
and the second equation

$$
\varepsilon_m \tilde{C}_p \left[ K_1(\eta_1) \frac{\partial x}{\partial \eta} \bigg|_{\eta_1} + K'_1(\eta_1) x \bigg|_{m} \right] \\
- \varepsilon_{in} C_{in} \frac{\partial x}{\partial \eta} \bigg|_{\eta_1} = \varepsilon_m \frac{\partial x}{\partial \eta} \bigg|_{\eta_1}.
$$

(A.91)

Using the derivatives

$$
\frac{\partial x}{\partial \eta} \bigg|_{\xi=0,m} = \frac{a}{2} \sqrt{\frac{(\zeta + a^2)}{(\eta_1 + a^2)(a^2 - b^2)(a^2 - c^2)}},
$$

(A.92)

$$
K'_1(\eta_1) = \frac{1}{(\eta_1 + a^2)R_{\eta_1}}.
$$

(A.93)

we can rewrite the second equation as

$$
\varepsilon_m \tilde{C}_p \left[ \frac{K_1(\eta_1)}{\eta_1 + a^2} + K'_1(\eta_1) \right] \\
- \varepsilon_{in} C_{in} \frac{1}{\eta_1 + a^2} = \varepsilon_m \frac{1}{\eta_1 + a^2}.
$$

(A.94)

which is equivalent to

$$
\varepsilon_m \tilde{C}_p \left[ K_1(\eta_1) + \frac{1}{R_{\eta_1}} \right] \\
- \varepsilon_{in} C_{in} = \varepsilon_m.
$$

(A.95)

Thus, the potentials are

$$
\Psi_{in} = \frac{\Psi_0}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}},
$$

(A.96)

$$
\Psi_p = \Psi_0 \frac{R_{\eta_1} \varepsilon_{m} - \varepsilon_{in}}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}} \frac{F_1(\xi) K_1(\eta) (\pi/2 - 1)}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}}.
$$

(A.97)
Then the far-field potential in the upper half-space, which is given by the pure polarization field, is

$$\Psi_p \approx -E_0 \frac{R_{\eta_1} \frac{\varepsilon_m - \varepsilon_{in}}{\varepsilon_m} K_1(\eta \approx -b^2)(\pi/2 - 1) a^3 x}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}} \frac{3}{r^3}$$

$$\approx -E_0 \frac{ab\sqrt{-\eta_1 \frac{\varepsilon_m - \varepsilon_{in}}{\varepsilon_m}} 2\pi (\pi/2 - 1) a^3 x}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}} \frac{3}{r^3}$$

$$= -E_0 \frac{ab\sqrt{-\eta_1 \frac{\varepsilon_m - \varepsilon_{in}}{\varepsilon_m}} \pi (\pi/2 - 1) 2x}{1 + \frac{L_1(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}} \frac{3}{r^3},$$  \hspace{1cm} (A.98)

where we assumed that $a \approx b$. The polarization far-field has the form of a dipole field at large distances $r$ from the wormhole. If the external electric field is applied in $y$-direction, we obtain the potentials

$$\Psi_{in} = \Psi_0 \frac{1}{1 + \frac{L_2(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}},$$  \hspace{1cm} (A.99)

$$\Psi_p = \Psi_0 \frac{R_{\eta_1} \frac{\varepsilon_m - \varepsilon_{in}}{\varepsilon_m} F_2(\xi) K_2(\eta)(\pi - 1)}{1 + \frac{L_2(\varepsilon_{in} - \varepsilon_m)}{\varepsilon_m}},$$  \hspace{1cm} (A.100)

with

$$F_2(\xi) = \int_{\xi}^{\infty} \frac{bd\xi'}{2\xi'^{3/2}(\xi' + b^2)} - \int_{\xi}^{\infty} \frac{bd\xi'}{2(\xi' + b^2)^{3/2}},$$  \hspace{1cm} (A.101)

$$K_2(\eta) = \int_{\eta}^{\infty} \frac{d\eta'}{(\eta' + b^2)R_{\eta'}.}$$  \hspace{1cm} (A.102)

We defined the geometrical factors

$$L_1 = R_{\eta_1} K_1(\eta_1) \approx ab\sqrt{-\eta_1} \int_{\eta_1}^{\infty} \frac{d\eta'}{(\eta' + a^2)R_{\eta'}},$$  \hspace{1cm} (A.103)

$$L_2 = R_{\eta_1} K_2(\eta_1) \approx ab\sqrt{-\eta_1} \int_{\eta_1}^{\infty} \frac{d\eta'}{(\eta' + b^2)R_{\eta'}},$$  \hspace{1cm} (A.104)
which are related to the depolarization factors by

\[ L_i = \frac{\varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_{\text{in}} - \varepsilon_0} L_i \]  \hspace{1cm} (A.105)

This result determines the polarizability of the uncharged hyperboloid observable in the far-field, i.e.

\[ \alpha_1 = \frac{2ab\sqrt{-\eta_1 \pi (\pi/2 - 1)}}{3} \frac{\varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_m + L_1(\varepsilon_{\text{in}} - \varepsilon_m)}. \]  \hspace{1cm} (A.106)

Similarly, we obtain the polarizability in y-direction, i.e.

\[ \alpha_2 = \frac{2ab\sqrt{-\eta_1 \pi (\pi/2 - 1)}}{3} \frac{\varepsilon_{\text{in}} - \varepsilon_m}{\varepsilon_m + L_2(\varepsilon_{\text{in}} - \varepsilon_m)}. \]  \hspace{1cm} (A.107)

Comparing to the polarizabilities of ellipsoids,[10] the polarizabilities of hyperboloids are proportional to \( ab\sqrt{-\eta_1} \), which corresponds to the volume of the ellipsoid \( abc \).

In the case of circular wormholes, we have \( a = b \), and therefore \( \alpha_1 = \alpha_2 = \alpha_{||} \), with \( L_1 = L_2 = L_{||} \).

Dispersion relations

In our proposed mid-IR light source the effective combination of silicon nitride (Si\(_3\)N\(_4\)) and hexagonal boron nitride (h-BN) behaves as an environment with polar phonons. Both materials are polar with ions of different valence, which leads to the Frohlich interaction between electrons and optical phonons.[110] Fig. A.2 shows that the interaction between the electrons in graphene and the polar substrate/graphene phonons modifies substantially the dispersion relations for the surface plasmon
polaritons in graphene. The RPA dielectric function of graphene is given by \[82, 68\]

\[
\varepsilon^{RPA}(\mathbf{q}, \omega) = \varepsilon_m - v_c(\mathbf{q}) \chi^0(\mathbf{q}, \omega) \\
- \varepsilon_m \sum_l v_{sph,l}(\mathbf{q}, \omega) \chi^0(\mathbf{q}, \omega) \\
- \varepsilon_m v_{oph}(\mathbf{q}, \omega) \chi_{j,j}^0(\mathbf{q}, \omega).
\]

(A.108)

The second term is due to the effective Coulomb interaction, and \(v_c(\mathbf{q}) = e^2/2q\varepsilon_0\) is the 2D Coulomb interaction. The effective electron-electron interaction mediated by the substrate optical phonons,

\[
v_{sph,l}(\mathbf{q}, \omega) = |M_{sph}|^2 G^0_l(\omega),
\]

(A.109)
gives rise to the third term, where \(|M_{sph}|^2\) is the scattering and \(G^0_l(\omega)\) is the free phonon Green function. The effective electron-electron interaction due to the optical phonons in graphene,

\[
v_{oph}(\mathbf{q}, \omega) = |M_{oph}|^2 G^0(\omega),
\]

(A.110)
gives rise to the last term of Eq. (A.108). \(|M_{oph}|^2\) is the scattering matrix element, and \(G^0(\omega)\) is the free phonon Green function. \(\chi_{j,j}^0(\mathbf{q}, \omega)\) is the current-current correlation function in Eq. (A.108).

The relaxation time \(\tau\) of the momentum consists of the electron-impurity, electron-phonon, and the electron-edge scattering, \(\tau^{-1} = \tau_{DC}^{-1} + \tau_{edge}^{-1} + \tau_{e-ph}^{-1}\), which determines the plasmon lifetime and the absorption spectrum bandwidth. It can be evaluated via the measured DC mobility \(\mu\) of the graphene sample through \(\tau_{DC} = \mu \hbar \sqrt{\pi \rho} / ev_F\), where \(v_F = 10^6\) m/s is the Fermi velocity, and the charge carrier density is given by \(\tau_{DC} = \mu \hbar \sqrt{\pi \rho} / ev_F\). The edge scattering time is \(\tau_{edge} \approx (1 \times 10^6(\text{m/s})/w - w_0)^{-1}\), where \(w\) is the edge-to-edge distance between the holes, and \(w_0 = 7\) nm is the adjustment parameter. The electron-phonon scattering time is \(\tau_{e-ph} = \hbar / 2\Im(\sum_{e-ph})\).
Figure A.2: The energy loss function for graphene with $\epsilon_F = 1.0$ eV. $k_{LSP4}$, $k_{LSP7}$, and $k_{LSP10}$ are the plasmon wavenumbers associated with the nanopatterning of the graphene sheet shown in Figs. 3.4 (a), (b), and (c), respectively. $\omega_{LSP4}$, $\omega_{LSP7}$, and $\omega_{LSP10}$ represent the LSP resonances shown in Figs. 3.4 (a), (b), and (c), respectively. The polar phonon resonance of h-BN and the surface polar phonon resonance of Si$_3$N$_4$ are denoted by $\omega_{BN}$, and $\omega_{SN}$, respectively. The Landau damping region is marked by the shaded area.

The imaginary part of the electron-phonon self-energy reads

$$\text{Im} \left( \sum_{e-ph} \right) = \gamma |\hbar \omega - \text{sgn} (\hbar \omega - E_F) \hbar \omega_{oph}|,$$

where $\gamma = 18.3 \times 10^{-3}$ is the electron-phonon coupling coefficient. The optical phonon energy of graphene is given by $\hbar \omega_{oph} \approx 0.2$ eV.

The loss function $Z$ describes the interaction of the SPPs and the substrate/graphene phonons. In RPA we have

$$Z \propto -\text{Im} \left( \frac{1}{\varepsilon_{RPA}} \right).$$

Fig. A.2 (a) shows the loss function for graphene with carrier mobility $\mu = 3000$ cm$^2$/V·s and a Fermi energy of $\epsilon_F = 1.0$ eV. In order to take advantage of the enhancement of the electromagnetic field at the position of the graphene sheet, the thickness of the optical cavity must be $\lambda/4n$, where
\( n \) is the refractive index of the cavity material.[82] The LSP resonance frequencies \( \omega_{\text{LSP}4}, \omega_{\text{LSP}7}, \) and \( \omega_{\text{LSP}10} \) mark the frequencies around the resonance wavelengths of 4 \( \mu m \), 7 \( \mu m \). and 10 \( \mu m \).

The resonance frequencies of the polar phonons are denoted by \( \omega_{\text{BN}} \) for h-BN and by \( \omega_{\text{SN}} \) for Si\(_3\)N\(_4\).

**Integral of dyadic Green function elements over spherical angle**

For the calculation of the spectral radiance we need to integrate the elements of the dyadic Green function over the spherical angle. We can split the total dyadic Green function into a free space term \( \vec{\mathcal{G}}_0(\mathbf{r}, \mathbf{r}'; \omega) \) and a term \( \vec{\mathcal{G}}_{\text{SPP}}(\mathbf{r}, \mathbf{r}'; \omega) \) that creates surface plasmon polaritons inside graphene.

Since the absorbance of the pristine graphene sheet is only 2.3\%, we can safely neglect \( \vec{\mathcal{G}}_{\text{SPP}}(\mathbf{r}, \mathbf{r}'; \omega) \).

Our goal is to calculate the gray-body emission of the EM radiation from the LSP around the holes in graphene into free space. Therefore, we need to evaluate

\[
I_{\text{GB}}(\omega) = \lim_{r \to \infty} \int r^2 \sin \theta d\theta d\varphi I_{\text{GB}}(r, \omega),
\]

where can use the approximation

\[
I_{\text{GB}}(r, \omega) = I_0(r, \omega) - I_{\text{SPP}}(r, \omega) \approx I_0(r, \omega).
\]

In Cartesian coordinates, we can write down the dyadic Green function as[66]

\[
\vec{\mathcal{G}}_0(\mathbf{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ \frac{1}{kr} - \frac{1}{k^2 r^2} \right] \mathbf{1} + \left( \frac{3}{k^2 r^2} - \frac{3i}{kr} - 1 \right) \hat{\mathbf{r}} \hat{\mathbf{r}}.
\]
Since we are interested only in the far field, we consider only the far-field component of the dyadic Green function, which is

\[ \vec{G}_{FF}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ \hat{\mathbf{r}} - \hat{\mathbf{r}} \right], \quad (A.116) \]

which possesses only angular (transverse) components but no radial (longitudinal) components. Then the necessary components are

\[ G_{xx}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin^2 \theta \cos^2 \varphi \right], \]
\[ G_{yx}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin^2 \theta \cos \varphi \sin \varphi \right], \]
\[ G_{zx}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin \theta \cos \theta \cos \varphi \right], \]
\[ G_{xy}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin^2 \theta \cos \varphi \sin \varphi \right], \]
\[ G_{yy}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin^2 \theta \sin^2 \varphi \right], \]
\[ G_{zy}(\vec{r}; \omega) = \frac{e^{ikr}}{4\pi r} \left[ 1 - \sin \theta \cos \theta \sin \varphi \right], \quad (A.117) \]

The corresponding integrals are

\[ \int r^2 \sin \theta d\theta d\varphi |G_{xx}(\vec{r}; \omega)|^2 = \frac{2}{15\pi}, \]
\[ \int r^2 \sin \theta d\theta d\varphi |G_{yx}(\vec{r}; \omega)|^2 = \frac{4}{15\pi}, \]
\[ \int r^2 \sin \theta d\theta d\varphi |G_{zx}(\vec{r}; \omega)|^2 = \frac{4}{15\pi}, \]
\[ \int r^2 \sin \theta d\theta d\varphi |G_{xy}(\vec{r}; \omega)|^2 = \frac{4}{15\pi}, \]
\[ \int r^2 \sin \theta d\theta d\varphi |G_{yy}(\vec{r}; \omega)|^2 = \frac{2}{15\pi}, \]
\[ \int r^2 \sin \theta d\theta d\varphi |G_{zy}(\vec{r}; \omega)|^2 = \frac{4}{15\pi}. \quad (A.118) \]
Doping of graphene due to Si$_3$N$_4$

The Silicon nitride, Si$_3$N$_4$, dielectric layer causes an effective n-type doping in graphene sheet.[102, 104] The shift in Fermi energy is given by

$$E_F = \hbar v_F \sqrt{\pi n}, \quad (A.119)$$

where $v_F$ is the Fermi velocity ($v_F \approx 10^6 m/s$ for graphene), $\hbar$ is Planck’s constant, and $n$ is the carrier density. The carrier density $n$ depends on the gate voltage and capacitance, i.e.

$$n = C \Delta V/e, \quad (A.120)$$

where $\Delta V = (V_G - V_{CNP})$ is the gate voltage relative to charge neutrality point, $e$ is electric charge, and $C$ is the capacitance of dielectric layer, given by $C' = \frac{\varepsilon_r \varepsilon_0}{d}$, $\varepsilon_r$ is relative permittivity, $\varepsilon_0$ is permittivity of free space, and $d$ is the thickness of dielectric layer.

The gate capacitance for a 50 nm thick Si$_3$N$_4$ layer in the infrared region is $C_G = 4.5 \times 10^{-8} F/cm^2$. From Eq. (A.119) we conclude that the Fermi energy $E_F = 1 \text{ eV}$ corresponds to a gate voltage relative to the CNP of $\Delta V = (V_G - V_{CNP}) = 6.9 \text{ V}$.

Wang et al.[102] observed that a Si$_3$N$_4$ film with a thickness of 50 nm shifts the CNP in a graphene sheet to -20 V, which shows that graphene is n-doped at zero gate voltage and the Fermi energy is $E_F = 1.74 \text{ eV}$. The Fermi energy can be tuned by applying a gate voltage to a desired value. In our work, we have used a Fermi energy of $E_F = 1 \text{ eV}$, which corresponds to $\Delta V = 6.59 \text{ V}$, i.e. for the CNP at -20 V, $V_G = -13.41 \text{ V}$ results in a Fermi energy of $E_F = 1 \text{ eV}$. From Eqs. A.119 and eq: density, the carrier density required to achieve a Fermi energy of $E_F = 1 \text{ eV}$ is $n = 1.94 \times 10^{12} \text{ cm}^{-2}$, which corresponds to an electric field of $E_{1.0eV} = \frac{en}{\varepsilon_r \varepsilon_0} = 1.38 \times 10^6 \text{ Vcm}^{-1}$, which is in the
safe zone compared to the reported breakdown field of the order of $10^7$ Vcm$^{-1}$.[118]
LIST OF REFERENCES


