



Numerical investigation of mid-infrared graphene plasmonics for tunable nanophotonic components

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Thesis presented in fulfilment of the requirements for the degree of Doctor of Sciences

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> Academic year 2015-2016

ISBN 978-2-87325-095-9

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This thesis was carried out in the context of a doctoral fellowship from the Fonds National de Recherche Scientifique (FRS-FNRS) - Wallonia (Belgium)

Acknowledgements

I would like to thank all the people who contributed, from a distance or close up, to the work described in this thesis.

My first thanks goes to my supervisor Prof. Bjorn Maes for giving me the opportunity to pursue a PhD thesis in his group, but also for his patience, availability and guidance. He scheduled regular meetings, gave accurate advices based on his knowledge or on an amazingly fast research in the literature. He was also a good mentor to inform me and to help me with the paper work to find a post-doctoral position.

I also thank Prof. Michel Voué and Dr. David Beljonne, members of my Support Committee for their interest in my work and for the constructive discussions we had.

My sincere thanks also goes to Prof. Philippe Tassin for his thoughtful welcome at Chalmers University of Technology in Gothenburg. He provided me the opportunity to join his team and gave me access to his research facilities. His support was precious to apprehend new simulation techniques and to widen my research from various perspectives.

I would like to thank Dr. Branko Kolaric for his trust and for giving me the opportunity to work with him.

I dedicate special thanks to Dr. Aimi Abass and Dr. Honghui Shen for their help on COMSOL modelling. Aimi for his assistance on COMSOL, and for providing me for useful advices and modelling tips at the beginning of my thesis, and Honghui for the model templates he sent me.

Of course I will not forget to thank my friends and colleagues, or if I may say officemates, Corentin Guyot, Mathieu Stock, Nicolas Rivolta, Fabio Vaianella and Galaad Altares. I appreciated the belote tournaments during lunch and after-works, challenges with balls, photo montages, special business cards, music tunes stuck in the head, artistic advice... But also the stimulating discussions and the mutual ii

assistance that prevails in the office.

But I do not restrain my acknowledgements to the office and I would like to emphasis on the pleasant atmosphere that reigns in the hall gathering together the Elementary Particles Physics Group, Materials Physics and Optics Group, and the Micro- and Nanophotonic Materials Group. Special thanks is devoted to Joseph Hanton for his natural help on computer issues, but also for his contribution to the great ambiance in the hall with the help of Francis Lequeux and Georges Kohnen.

Last but not least, I also thank my family for their general support in major decisions I have made and for their encouragements, and my girlfriend for supporting me throughout the writing process.

And thank you for reading those lines!

Summary

Graphene, a two-dimensional sheet of carbon atoms in a hexagonal lattice, has spread tremendous interest since its discovery. At first, graphene was highly attractive for its strong conductivity and high carrier mobility, but it also proved interesting with respect to the optical properties. Indeed, graphene exhibits a strong light-matter interaction and provides highly confined plasmons in the mid-infrared range, where it behaves as a metal. Furthermore, by adjusting the Fermi level by chemical doping or by a gate voltage one can significantly shift the properties of these plasmons. Our goal is to explore graphene plasmons and investigate their tunability in various configurations, which may lead to efficient and compact photodetectors, sensors and integrated optoelectronic components.

In this thesis we study the reflection and transmission of plasmons along a doping change with various profiles, and we derive simple laws to describe these effects. Doing so, we demonstrate that a graphene plasmon can be assimilated to twodimensional plane wave propagation, where a wide range of refractive indices (the effective indices of the plasmons) are available, paving the way to numerous applications currently explored in the photonics discipline. Furthermore, we investigate plasmons encountering graphene edge structuration, such as designed defects on the sheet. Next, plasmons coupling with graphene ribbon cavities show total absorption with a high tunability thanks to graphene doping. Finally, we consider beams coupling with graphene nano-disk dimers of asymmetric doping and we demonstrate strong resonances and tunability.

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1

Introduction

1.1 Introduction

In a world of constant change, innovation has never been so relevant. In the last decades optical fibers have replaced the old copper cables for communication, enabling the development of a giant and fast data network. From there, we have witnessed an impressive deployment of new technologies: smartphones have become the norm and their development still requests a growing need for more connected, faster and smaller integrated devices.

Although long-distance data transfers are currently handled by photonics, data processing is still mostly achieved by silicon micro-electronics. This sixties based technology was expected to grow exponentially according to Moore's law but it is actually reaching its physical limits. Therefore, although silicon micro-electronics still reigns supreme, it is clear that new technological developments are strongly desired and that the next technological generation will be optoelectronic, combining electrons and photons.

Over the past decades, with the increased control of materials down to the nanoscale, photonics has been widely investigated, tailoring the flow of light over an increased path length for diverse functionalities. Apart from telecommunications and information processing, photonics applications are numerous: lighting, displays, solar cells, detectors, microscopy, lasers and spectroscopy. A major problem is however due to the diffraction limit, restricting the compactness of photonic devices [1].

In this context, plasmonics has emerged [2]. It is a subfield of photonics dealing with the excitation, manipulation and utilization of the coupling between light and collective electron oscillations. These so-called plasmons demonstrate a high confinement and a strong field enhancement. Plasmonics research has mainly focused on noble metals such as gold and silver. It showed its benefit in nanophotonic integrated systems for its ability to control light at scales considerably smaller than the wavelength [1, 3, 4] and in metamaterials, where metals are a crucial ingredient for their implementation [5, 6, 7]. This led to unusual electromagnetic phenomena including negative refraction [8], superlensing [9], and cloaking [10]. However, these great progresses are substantially held up by the losses encountered in noble metals. Recently, a new, exciting and promising material arrived, offering less-damped light propagation and tunability of its optical properties: graphene.

Graphene is a two-dimensional (2D) one-atom-thick carbon sheet, thought for a long time to be unstable at finite temperature. In 1930 it was shown that thermal fluctuations would destroy and melt the 2D lattice at finite temperature [11, 12]. Therefore, Geim and Novoselov created a surprise in 2004 with their discovery of the first 2D crystal, with an astonishingly simple fabrication technique [13]. Graphene was purely and solely extracted by scotch tape exfoliation of graphite, which is a stack of weakly bound graphene planes. Nowadays, a myriad of fabrication techniques has been developed and are further discussed in Chapter 2. Graphene is flexible, strong and ecological (carbon is the basic ingredient of life and all organic chemistry). Furthermore, its expected low-cost production comes together with its alluring electronic and optical properties, which are still a subject of intense research and will lead to plenty of practical applications [14, 15, 16].

In the visible wavelength range, graphene is considered transparent since it absorbs only 2.3% of the normal incident light, but at the same time it demonstrates a strong light-matter interaction for a simple carbon sheet [17]. Moreover, its strong carrier mobility (theoretically it can reach $\mu > 10^7 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$) makes it suitable as a transparent electrode in solar cells [18], in OLED or in LEDs, the latter reaching the market this year.

Nowadays the mid-infrared range is under strong investigation with the development of novel sources [19]. In this range, going from 2 μ m to 15 μ m, graphene supports low-loss and confined plasmons. Graphene plasmons are very suitable for practical applications for three reasons: they are less damped, more confined and tunable in comparison to noble metal plasmons. These three properties are extensively studied and described in this thesis: Chapter 3 compares the damping and the confinement of graphene plasmons to noble metal plasmons, and the graphene tunability is explored in the following chapters. A complete understanding of graphene plasmons will play an important role in a wide variety of applications in information and communication technologies, medical sciences, chemical and biological sensing, and spectroscopy, among many others [20].

One of the most exciting applications stands in "flatland optics". This concept was introduced by Vakil and Engheta in [21] and exploits the tunability of the optical properties of graphene. They start from the fact that graphene can be locally doped, so that a single sheet of graphene will offer different patches of conductivity, and therefore various propagation properties of plasmons. In this way graphene is transformed in an optical platform for integrated circuitry or transformation optics. For example, this allows for the design of a completely planar beam splitter and a Luneberg lens.

This concept of flatland optics remains timely. After the discovery of the semimetal graphene, new two-dimensional materials have emerged with different properties, such as transition-metal dichalcogenides (mostly acting as semiconductors) and hexagonal boron nitride (acting as dielectric in visible or polaritonic in mid-infrared). The combination of these two-dimensional materials are required to realize flat allgated optoelectronic devices [22].

This thesis draws its inspiration from the work of Vakil and Engheta in order to study the electromagnetic behavior in pure graphene structures and to take advantage of the tunability of graphene. The problem is approached by rigorous finiteelement method calculations and is further described by more intuitive semi-analytic approaches. In the following section we describe the different subjects addressed in this work.

1.2 Outline

This thesis is structured as follows. Chapter 2 is devoted to the basic description of graphene. We first explain the hexagonal carbon lattice and we derive its electronic properties leading to the famous Dirac cones. Subsequently, the optical parameters are presented and rigorously investigated, since it is the basis of this work. Graphene is characterized by an optical conductivity, which is a sum of two contributions. The interband optical conductivity is responsible for the constant 2.3% absorption of the visible light, while the intraband part has a Drude form similar to noble metals. The latter is characterized by a small real part responsible for the small damping of graphene plasmons. The analytical expression of the conductivity depends on the scattering lifetime of electrons, the frequency, the doping (leading to graphene tunability) and the temperature. Finally, we discuss the production techniques of graphene sheets, depending on their size.

Chapter 3 addresses the background theoretical concepts relevant for the thesis. Starting from Maxwell's equations, we derive the Helmholtz equation useful to extract the modes of light, we define the Poynting vector, we present the boundary conditions and we determine the Fresnel coefficients. Then, an important part is devoted to graphene plasmons: they are characterized and compared to standard surface plasmons polaritons of noble metals, demonstrating their superiority in terms of propagation length and confinement. Finally, cavities and their resonances are considered together with the mathematical tools to describe them: localized surface plasmon resonances, Fabry-Pérot cavities and coupled mode theory.

Chapter 4 is devoted to nanoribbon resonances and exposes the work we have published in [23]. In recent works plasmons in graphene nanoribbons were studied fundamentally in single elements [24] or in gratings for a tunable optical response [20, 25, 26]. Other potential nanophotonic components were discussed such as bends and splitters [27], and directional couplers [28, 29]. It was therefore legitimate to investigate the coupling of graphene sheet plasmons with nanoribbons to provide the building blocks of plasmonic circuitry. It is known that the graphene plasmon reflectance at the edge is total [30]. In Chapter 4 we show how the reflectance can be modulated by a graphene nanoribbon. First, it is placed at the end of the sheet, leading to reflectance dips in the spectrum strongly depending on graphene doping, the distance to the sheet, and the scattering lifetime of electrons. The results are further described by coupled mode theory and an electrostatic scaling law is derived, allowing for a straightforward prediction of the resonant wavelength. Second, the reflectance is modulated by a nanoribbon on top of the graphene sheet and the results are supported by a scattering matrix formalism. A complex reflectance pattern depending on the position of the ribbon emerges, with particular zones of zero reflectance.

Chapter 5 provides a more detailed study of the nonuniform doping feature proposed in [21], examining the reflectance and transmittance of graphene plasmons impinging upon doping interfaces of different profiles. That work was published in [31]. This chapter also describes the behavior of graphene plasmons encountering local doping changes i.e. graphene defects that are common in manufactured sheets and can arise from local distortions in the lattice. Defects were addressed in [32, 33] where they demonstrated experimentally and theoretically the plasmon scattering caused by cracks in graphene, while doping inhomogeneities have been approached numerically in [34] neglecting losses. In Chapter 5, we provide a complete study, including losses, of abrupt or smooth doping variations along the propagation direction in order to create plasmonic tapers or couplers. We show that the transmittance and reflectance of the plasmon can be described by a Fresnel approach, including the effective index of the plasmon in the Fresnel coefficients. Finally, local inhomogeneities are designed, leading to small cavities that can reach total absorption under certain conditions.

After local doping defects, Chapter 6 considers structural defects at the end of a graphene sheet and extends results we have published in [35]. If graphene plasmons encountering the straight edge of a graphene sheet are totally reflected, in this chapter we reveal that the reflection is strongly determined by the edge structure. The examined ribbon grating case offers longitudinal and lateral edge mode Fabry-Pérot resonances that drastically decrease the reflection for particular ribbon length and width combinations, where it would have been nearly 100% for a non-structured edge. The edge mode coupling is associated with phase changes, influencing the positions of the longitudinal cavity modes. Furthermore, the rapidly changing transmittance beyond such a resonance leads to particular points of critical and near-zero reflection.

Finally, Chapter 7 investigates the behavior of graphene nanodisk dimers excited via an incident beam, following the results available in [36]. This chapter, therefore, introduces another excitation type of plasmonics: the localized surface plasmon resonance, already known for a long time in the visible range. At present, metallic nanoparticles with various shapes from spheres to triangles and nanorods are widely investigated [37, 38, 39, 40]. They demonstrate a strong interaction with incident light, with for example hot spots on tips or on adjacent edges [41], enabling

them for a wide range of applications such as biosensing [42], nonlinear optics [43], nanocircuits [3], optoelectronics [44] and metamaterials [45]. The combination of more metallic particles can also reveal higher order modes that are not excitable at normal incidence for a single particle [46]. However, in metallic dimers the only effective way to access these so-called dark modes is to change the physical configuration (size [40], metal [47],...). With graphene those modes are simply accessible with doping. Incidentally, in Chapter 7 we consider two graphene disks under normal incident plane wave and discuss their interaction when the disks are differently doped. The plasmonic modes hybridize and new resonances appear, converting dark higher order modes into visible, relatively narrow resonances. Two polarizations are considered and their slightly different response is analyzed by a Hamiltonian model. A final section discusses graphene dimers connected by a graphene ribbon. The tunable, conductive junction allows charge to oscillate in between the two disks, creating a new narrower and enhanced resonance, the charge transfer plasmon. The former non-bridged resonance is blue-shifted in the presence of a conductive bridge and weaker because it is screened by the charge transfer (screen bonding dipolar plasmon).

1.3 Publications

1.3.1 Publications in international journals

Here follow the contributions published in international journals during the PhD work:

- G. Rosolen and B. Maes, Graphene ribbons for tunable coupling with plasmonic subwavelength cavities, *Journal of Optical Society of America B*, vol. 31, pp. 1096-1102, 2014
- G. Rosolen and B. Maes, Nonuniform doping of graphene for plasmonic tapers, *Journal of Optics*, vol. 17, p. 015002, 2015
- F. Vaianella, G. Rosolen and B. Maes, Graphene as a transparent electrode for amorphous silicon-based solar cells, *Journal of Applied Physics*, vol. 117, p. 243102, 2015
- M.A. van der Veen, G. Rosolen, T. Verbiest, M.K. Vanbel, B. Maes and B. Kolaric, Nonlinear optical enhancement caused by a higher order multipole mode of metallic triangles, *Journal of Materials Chemistry C*, vol. 3, pp. 1576-1581, 2015
- G. Rosolen and B. Maes, Patterned graphene edges for tailored reflection of plasmonic modes, *Optics Letters*, vol. 40, pp. 2727-2730, 2015
- G. Rosolen and B. Maes, Asymmetric and connected graphene dimers for a tunable plasmonic response, *Physical Review B*, vol. 92, p. 205405, 2015

1.3.2 Contributions in international conferences

A list of our conference talks / proceedings contributions:

- G. Rosolen and B. Maes, *Graphene Nanophotonics: controlling ultra-confined light*, 7th Optoelectronics and Photonics Winter School, Trento, Italy, 2013
- G. Rosolen and B. Maes, *Graphene plasmonics: tunable coupling with nanocavities*, Annual symposium of the IEEE Photonics Society Benelux Chapter, Eindhoven, Netherlands, 2013
- G. Rosolen and B. Maes, *Graphene plasmonics: tunable coupling with nanocavities*, Nanolight 2014, Benasque, Spain, 2014
- G. Rosolen and B. Maes, *Graphene doping inhomogeneities: plasmonic taper applications*, Annual symposium of the IEEE Photonics Society Benelux Chapter, Twente, Netherlands, 2014
- G. Rosolen and B. Maes, *Graphene Plasmonics: Propagation control for optoelectronic applications*, Frontiers in Nanophotonics, Monte Verita, Ascona, Suisse, 2015
- G. Rosolen and B. Maes, *Patterned graphene edges for tailored reflection of plasmonic modes*, Metamaterials, Oxford, UK, 2015
- G. Rosolen and B. Maes, *Absorption tunability of asymmetric ad bridged graphene dimers*, Annual symposium of the IEEE Photonics Society Benelux Chapter, Brussels, 2016

2

Graphene

What was old is new again! This adage perfectly fits carbon, known since antiquity as graphite or diamond. It was recently brought up to date with the discovery of graphene in 2004 by Konstantin Novoselov and Andre Geim. The outstanding properties of graphene led them to the 2010 Nobel Prize: it is about 207 times stronger than steel by weight, is flexible, conducts heat and electricity efficiently, is nearly transparent and supports relatively low-loss propagative plasmons.

This chapter demonstrates the electronic dispersion of a graphene sheet and describes the Dirac cones in Sec. 2.1. Afterwards, the optical conductivity and permittivity of graphene are investigated in Sec. 2.2 under judicious conditions. Finally, a brief review on graphene production and doping is proposed in Sec. 2.3.

2.1 Dirac cones

This section is devoted to the derivation of the band diagram of graphene. First, Sec. 2.1.1 describes the honeycomb lattice. Then, the electronic dispersion of graphene is derived in Sec. 2.1.2, leading to the Dirac cones in Sec. 2.1.3.



Figure 2.1: (a) Hexagonal lattice of the graphene sheet. The unit cell constitutes of 2 atoms A and B (red and blue, respectively). $\mathbf{a_i}$ are the primitive unit vectors and $\mathbf{R_j}$ are the Bravais lattice vectors. The unit cell is the dashed parallelogram. (b) Reciprocal lattice and first Brillouin zone in k-space.

2.1.1 Graphene lattice

In its elemental form, the carbon atom has two core electrons (1s orbital) and four valence electrons (in 2s and 2p orbitals) in its ground state [48]. When carbon atoms form graphene, the 2s orbitals interact with the $2p_x$ and $2p_y$ orbitals to form three sp^2 hybrid orbitals. The sp^2 hybridization then creates three in-plane bonds that are called σ -bonds and are the strongest type of covalent bond. The $2p_z$ orbitals overlap and creates π -bonds that are normal to the plane of the σ bonds and weakly bound to the nuclei. These electrons are relatively delocalized and thus enable electron transport.

Such hybridization leads to a hexagonal lattice of carbon atoms. The primitive unit cell of the lattice is constituted of two atoms and can be considered an equilateral parallelogram with side $a = \sqrt{3}a_{C-C} = 2.46$ Å, where $a_{C-C} \approx 1.42$ Å is the carbon-carbon atom distance. Each carbon atom is bonded to its three nearest neighbours and the vectors describing the separation between type A atom and the nearest neighbour type B atom are subscripted \mathbf{R}_i on Fig. 2.1a. The primitive unit vectors are

$$\mathbf{a_1} = \frac{a}{2} \left(\sqrt{3}, 1\right), \qquad \mathbf{a_2} = \frac{a}{2} \left(\sqrt{3}, -1\right) \tag{2.1}$$

The reciprocal lattice vectors are given by

$$\mathbf{b_1} = \frac{2\pi}{a} \left(\frac{\sqrt{3}}{3}, 1\right), \qquad \mathbf{b_2} = \frac{2\pi}{a} \left(\frac{\sqrt{3}}{3}, -1\right) \tag{2.2}$$

The first Brillouin zone (BZ) is represented in Fig. 2.1b, with the particular \mathbf{K} points at the corners of the BZ, where the Dirac cones will appear. The six corners form two groups of \mathbf{k} points, labelled \mathbf{K} and \mathbf{K}' .

2.1.2 Electronic dispersion

In order to derive the analytical electronic band structure of graphene $E(\mathbf{k})$, we need to solve the Schrödinger equation $H\Psi(\mathbf{k}, \mathbf{r}) = E(\mathbf{k})\Psi(\mathbf{k}, \mathbf{r})$. A general solution satisfying the Bloch theorem adapted to a periodic basis of two atoms (A and B, Fig. 2.1a) gives

$$\Psi(\mathbf{k}, \mathbf{r}) = C_A \Phi_A(\mathbf{k}, \mathbf{r}) + C_B \Phi_B(\mathbf{k}, \mathbf{r})$$
(2.3)

where the Φ_m expresses the Bloch function as the linear combination of the atomic orbitals ϕ in a periodical environment (known as Wannier functions)

$$\Phi_m(\mathbf{k}, \mathbf{r}) = \frac{1}{\sqrt{N}} \sum_{j=1}^{N} e^{i\mathbf{k}\cdot\mathbf{R}_{\mathbf{m}_j}} \phi(\mathbf{r} - \mathbf{R}_{\mathbf{m}_j})$$
(2.4)

where N is the number of unit cells in the lattice and \mathbf{R}_{m_j} are the Bravais lattice vectors identifying the location of all type m atoms in the unit cell j of the graphene lattice. Inserting solution 2.4 in the Schrödinger equation, and integrating over the lattice (called Ω in Eq. 2.7), one finds

$$C_A(H_{AA} - ES_{AA}) = C_B(ES_{AB} - H_{AB})$$
(2.5)

$$C_A(H_{AB}^* - ES_{AB}^*) = C_B(ES_{AA} - H_{AA})$$
(2.6)

with the definitions of H_{mn} the Hamiltonian or transfer integral of the electrons and S_{mn} the overlap matrix elements between Bloch functions

$$H_{mn} = \int_{\Omega} \Phi_m^* H \Phi_n d\mathbf{r}, \quad S_{mn} = \int_{\Omega} \Phi_m^* \Phi_n d\mathbf{r}$$
(2.7)

Within the tight-binding approximation that the wavefunction of an electron in any primitive unit cell only overlaps with the wavefunctions of its nearest-neighbours (3 of them), one finds that $H_{AA}(\mathbf{k}) = E_{2p}$, close to the energy of the 2p orbital in isolated carbon (close because the Hamiltonian has a periodic potential) and $S_{AA} = 1$ because the Wannier functions are normalized. A step further assumes an electron-hole symmetry and leads to $S_{AB}(\mathbf{k}) = 0$. Finally, we get the energy dispersion proposed by Wallace in 1947 [49]

$$E(\mathbf{k})^{\pm} = E_{2p} \pm \sqrt{H_{AB}(\mathbf{k})H_{AB}^{*}(\mathbf{k})}$$
(2.8)

with H_{AB} the Hamiltonian of the interaction of one atom with its three neighbors

$$H_{AB}(\mathbf{k}) = \gamma \left(e^{-i\mathbf{k}\cdot\mathbf{R_1}} + e^{-i\mathbf{k}\cdot\mathbf{R_2}} + e^{-i\mathbf{k}\cdot\mathbf{R_3}} \right)$$
(2.9)

where $\gamma \approx 2.8$ eV is the nearest-neighbor hopping energy [50]. Hence the analytical π bands of graphene are given by

$$E(\mathbf{k})^{\pm} = \pm \gamma \sqrt{1 + 4\cos\frac{\sqrt{3}a}{2}k_x \cos\frac{a}{2}k_y + 4\cos^2\frac{a}{2}k_y}$$
(2.10)

where we set $E_{2p} = 0$ eV, as the energy is defined up to an arbitrary reference potential.

The next paragraph provides an interpretation of the band diagram derived in Eq. 2.10.



Figure 2.2: (a) Dispersion of the electronic π -bands of graphene following analytical Eq. 2.10. (b) Zoom on a Dirac cone, for pristine graphene, n-doped graphene and p-doped graphene (from left to right).

2.1.3 Band diagram

In Eq. 2.10, the upper band (+) is called the π^* or conduction band, while the lower (-) one is called the π or valence band. The two bands meet in the six corners of the BZ in the **K** and **K**'-points. This particular point is the Fermi energy level of charge neutral graphene. Indeed, since there are two atoms per unit cell, there are two electrons per unit cell (the electrons from $2p_z$ orbitals, see Sec. 2.1.1). Owing to Pauli's exclusion principle, the valence band is then filled with these two electrons, leaving the conduction band empty.

Fig. 2.2a represents the band diagram of Eq. 2.10 with the valence band in red and the conduction band in cyan. The Fermi energy (E_F) , defined as the highest occupied k-state when the solid is in its ground state, is then at the 6 K and K' points, called the Dirac points. Owing to the absence of a band gap and to the position of E_F , graphene is called a semi-metal. A zoom around these peculiar points is depicted in Fig. 2.2b, showing the Dirac cones.

In fact, in the vicinity of the Dirac points, the energy dispersion is linear. A Taylor expansion of Eq. 2.10 near \mathbf{K} yields

$$E(\mathbf{k})^{\pm} = \pm \frac{\sqrt{3}}{2} \gamma a |\mathbf{k}| = \pm v_F \hbar |\mathbf{k}|$$
(2.11)

where $v_F = \sqrt{3\gamma a/2\hbar} \approx 10^6 \text{ ms}^{-1}$ is the Fermi velocity. This linear dispersion is different from the usual parabolic dispersions and confers outstanding transport properties to graphene. The linearity implies a constant velocity v_F , independent on the energy or the momentum. It is similar to massless particles, which can be described by Einstein's special relativity in the form of Dirac's relativistic quantum mechanical wave equation, but this parallel is beyond the scope of this thesis.

2.2 Optical parameters

Within the energy bands derived from the tight-binding approximation (Eq. 2.11), we can obtain the optical conductivity of graphene. Indeed, the linear response of a collection of electrons to an external applied electromagnetic field can be described by the Kubo-Greenwood formula [51]. In this formulation, the Hamiltonian is modified through the application of a potential vector. Then, the random-phaseapproximation (RPA) is used to take into account the electron-electron interactions [52]. RPA is an extension of the tight-binding approximation from the previous section to include the dynamical response of the system. Neglecting spatial dispersion, the developments can be found in [53, 54] and give (with convention $e^{j\omega t}$)

$$\sigma(\omega, E_F) = \sigma_{\text{intra}}(\omega, E_F) + \sigma_{\text{inter}}(\omega, E_F)$$
(2.12)

with

$$\sigma_{\rm intra}(\omega, E_F) = \frac{-2je^2k_BT}{\hbar^2\pi(\omega - j\tau^{-1})}\ln\left[2\cosh\left(\frac{E_F}{2k_BT}\right)\right]$$
(2.13)

and

$$\sigma_{\text{inter}} = \frac{e^2}{4\hbar} \left[\frac{1}{2} + \frac{1}{\pi} \arctan\left(\frac{\hbar\omega - 2E_F}{2k_BT}\right) \right] + \frac{e^2}{4\hbar} \left[\frac{j}{2\pi} \ln\frac{(\hbar\omega + 2E_F)^2}{(\hbar\omega - 2E_F)^2 + (2k_BT)^2} \right]$$
(2.14)

with k_B the Boltzmann constant, T the temperature, e the elementary charge and τ the scattering lifetime of electrons.

The σ_{intra} term corresponds to the intraband electron-photon scattering process (see Fig.2.3b), with the τ^{-1} term in order to take the electron-disorder scattering processes into account. It corresponds to the Drude model of classical metals, except for the E_F factor conferring to graphene its tunability. Indeed, assuming $E_F \gg k_B T \approx 0.026$ eV at room temperature gives

$$\sigma_{\text{intra}} = \frac{-je^2|E_F|}{\hbar^2 \pi (\omega - j\tau^{-1})} \tag{2.15}$$

The σ_{inter} term originates from the direct interband electron transitions: charge carriers are excited from the valence band to the conduction band (see Fig.2.3a). They only occur for $\hbar\omega \geq 2E_F$ owing to Pauli's exclusion principle. That threshold distinctly appears at T = 0 K with the step function θ :

$$\sigma_{\text{inter}}(\omega, E_F) = \frac{e^2}{4\hbar} \left[\theta(\hbar\omega - 2E_F) + \frac{j}{2\pi} \ln \frac{(\hbar\omega + 2E_F)^2}{(\hbar\omega - 2E_F)^2} \right]$$
(2.16)

In the next paragraph, we provide a deeper explanation for the different parameters in these equations.



Figure 2.3: (a) Interband transitions occur when $\hbar \omega > 2E_F$ owing to Pauli's exclusion principle. (b) Intraband transitions occur at smaller energies.

2.2.1 Parameter discussion

Three essential parameters play a role in the optical conductivity of graphene: the scattering lifetime of electrons τ , the doping level of graphene E_F and the temperature T.

Scattering lifetime of electrons

The scattering lifetime of electrons τ is a fitting parameter depending on the quality of the graphene sheet. From Boltzmann transport theory [55], it is related to the electron mobility μ following

$$\tau = \frac{\mu E_F}{e v_F^2} \tag{2.17}$$

with v_F the Fermi velocity.

The mobility is a difficult parameter to evaluate. It depends on the carrier density (and thus doping E_F), the graphene sheet quality and the temperature. Theoretically, high electron mobilities are expected from simulations ($\mu > 10^7 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$), these theoretical results are 2 orders of magnitude higher in comparison to measurements in suspended graphene [56]. High graphene sheet quality is obtained on hexagonal boron nitride [57] thanks to their good lattice matching, demonstrating a mobility of $\mu = 0.5 \times 10^6 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$. In contrast, graphene on silicon carbide [57] gives rise to a smaller mobility $\mu = 4.5 \times 10^4 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$.

For our model of suspended graphene, we choose the very conservative value of $\mu = 10^4 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$ measured at room temperature and induced by a gate voltage [13]. This leads to a scattering lifetime of $\tau = 10^{-13}$ s for a doping of $E_F = 0.1 \text{ eV}$.

Furthermore, the choice of a conservative value is justified since losses could appear from other channels such as many-body interactions [58] or deviations from perfect Dirac-cone band structures [59]. Another known loss channel is the electronoptical phonon interaction [60], appearing when $\hbar \omega > 0.2$ eV or $\lambda < 6 \ \mu$ m. These losses are not accounted for in our model.

Doping

One of the greatest interests in graphene is that adjusting its Fermi level E_F changes its optical conductivity. There are two main ways to achieve doping in graphene: chemical modification and electric field gating. The literature mainly reports on adjusting the charge density n. It is related to the doping level E_F by [48]

$$E_F = \hbar v_F \sqrt{\pi n} \tag{2.18}$$

Electrostatic doping is achieved with a bias voltage. This technique reaches high carrier densities: $n > 10^{14}$ cm⁻² i.e. $E_F > 1.2$ eV with ionic glass mobility [61], or with ion gel [62, 63, 64]. Tuning the bias voltage results in a tuning of the doping level of graphene (more details on experimental setups in Sec. 2.3.3).

Chemical doping [65] is distinguished in two categories: surface transfer doping and substitutional doping. Surface charge transfer is achieved by electron transfer with a doped semiconductor or gas dopants that adsorb on the surface. This technique does not disrupt the structure of graphene and leads to a reversible doping. To the best of our knowledge, the maximal doping achieved yet is $E_F = 0.45$ eV [66]. Substitutional doping however is based on the replacement of a carbon atom by a dopant atom such as nitrogen or boron for example. That technique can lead to high doping levels close to $E_F = 1$ eV but may disrupt the band structure of graphene and create a bandgap [67, 68].

Hence, we can suppose a conservative tunable value of the doping level between 0 and 1 eV.

Temperature

Temperature is also an important parameter for graphene, since it plays a role on the real part of the conductivity: the thermal agitation reduces the electron mobility. For realistic purpose, we only consider room temperature i.e. T = 300 K.

2.2.2 Optical conductivity

The real and imaginary parts of the conductivity are represented in Fig. 2.4 for different doping levels, where we defined $\sigma_0 = e^2/4\hbar$. For small frequencies ($\hbar\omega \ll E_F$), the intraband term dominates and the Drude model appears, following the σ_{intra} term (compare Eqs. 2.15 and 2.14). Indeed, with the assumption $E_F \gg k_B T$ and $\hbar\omega \ll 2E_F$, σ_{inter} goes to 0 (see Eq. 2.14). That is called the **Drude approximation** throughout this thesis.

On the other hand, for higher frequencies, the real part of the conductivity shows a step from around $0.1\sigma_0$ to σ_0 , and the imaginary part demonstrates a local maximum. It occurs at the frequency $\hbar\omega = 2E_F$ where interband transitions appear. In visible frequencies the real part reaches a maximum. It is the well-known value of $\sigma_0 = e^2/4\hbar$ that leads to a constant absorption of 2.3 % for normal incident visible light on a graphene sheet [17]. The convergence can be checked in Eq. 2.14, the intraband transitions σ_{intra} in Eq. 2.15 being negligible at high frequencies.



Figure 2.4: Graphene optical conductivity normalized by $\sigma_0 = e^2/4\hbar$ for three doping levels : $E_F = 0.2$, 0.6 and 1 eV. The solid lines represent the real part of the conductivity and the dashed lines show the imaginary part. Increasing graphene doping drives the conductivity to higher frequencies.

The amazing properties of graphene doping are well illustrated in Fig. 2.4. Increasing doping shifts the threshold frequency of interband transitions, conferring metallic properties to graphene at higher frequencies.

This thesis focuses on the region in between, where the real part of the conductivity reaches a local minimum. In this regime, graphene supports low-losses surface plasmons as discussed in Sec. 3.2.2.

2.2.3 Permittivity

In simulations the conductivity can often be modelled as a boundary condition. However, some mode solvers require bulk materials, described by a permittivity. Therefore, an effective thickness d must be considered, resulting in an effective graphene volume. The latter can be described by an effective volume conductivity: $\sigma_{\text{eff}} = \sigma/d$.

Supposing a harmonic dependence of the fields, the permittivity of this bulk

graphene is found as follows [2]

$$\varepsilon_0 \varepsilon_r \mathbf{E} = \mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \tag{2.19}$$

$$=\varepsilon_0 \mathbf{E} + \frac{\mathbf{J}}{j\omega} \tag{2.20}$$

$$=\varepsilon_0 \mathbf{E} + \frac{\sigma_{\text{eff}} \mathbf{E}}{j\omega} \tag{2.21}$$

$$=\varepsilon_0 \mathbf{E} + \frac{\sigma \mathbf{E}}{j\omega d} \tag{2.22}$$

where ε_0 is the vacuum permittivity and **P** is the polarization and is related to the internal charge density (together with the charge conservation one gets $\mathbf{J} = \partial \mathbf{P} / \partial t$, see Eq. 3.8). The relative permittivity of a graphene sheet is then

$$\varepsilon_r = 1 - \frac{j\sigma}{\varepsilon_0 \omega d} \tag{2.23}$$

An equivalent plot of the conductivity in Fig. 2.4 is represented in Fig. 2.5 for 5 nm thick graphene permittivity. The three regimes explained in Sec. 2.2 become clearer. For low frequencies ($\hbar\omega \ll 2E_F$), the Drude model applies. For higher frequencies ($\omega \approx 0.1$ to 3×10^{15} rad/s for a 1 eV doped graphene sheet for example), the imaginary part of the permittivity reaches a steady value close to zero. The restricted regime $\omega \approx 0.1$ to 1.5×10^{15} rad/s (1 eV doped) is perfect for plasmons because graphene is metallic ($\Re{\{\varepsilon\}} < 0$) and losses are really small ($\Im{\{\varepsilon\}}$ close to zero). For even higher frequencies, the real part of the permittivity tends to 1, explaining the non-reflectivity ($R < 1.5 \times 10^{-4}$) of graphene in the visible range [17].

Note that graphene's optical parameters are defined as a surface conductivity that we recast here as a bulk permittivity. Supposing an anisotropic component along the effective thickness or perpendicular direction of the graphene sheet should be legitimate. The transverse component of the permittivity was measured with ellipsometry in [69]: $\varepsilon_{\perp} = 1.98 - 0.18j$. However, considering anisotropic or isotropic permittivity gives similar results in the visible range [70]. Up to now, no careful analysis has been made in the infrared range, but we can suppose a negligible effect of this component in our models regarding experimental observations of graphene plasmons [71].

2.2.4 Limitations

The optical conductivity of graphene defined in Eqs. 2.15 and 2.14 is established in the approximation of high frequencies ($\omega \gg \tau^{-1}$). With τ varying between 10^{-13} to 10^{-12} s for doping between 0.1 and 1 eV (see Eq. 2.17), the conductivity is valid for $\omega \gg 10^{13}$ rad/s or $\lambda \ll 190 \ \mu$ m. In this thesis, we will limit ourselves the mid-infrared range $1 < \lambda < 15 \ \mu$ m where graphene plasmons appear.

We also assume $E_F \gg k_B T \approx 0.026$ eV at room temperature. Hence, working with doping in between 0.1 and 1 eV, we are in the validity domain of the analytic conductivity established previously.

Finally, it is worth discussing the finite-size quantum effects. For small ribbons or nanodisks, edges constitute a new damping pathway. There are two types of



Figure 2.5: Graphene permittivity of 5 nm effective thickness for three doping levels: $E_F = 0.2$, 0.6 and 1 eV. The solid lines represent the real part of the permittivity and the dashed lines show the imaginary part. Increasing graphene doping drives the conductivity to higher frequencies.

graphene edges: armchair (AC) or zigzag (ZZ) depending on the shape of the edge. The small width of graphene flakes can lead to quantum confinement of electrons, which restricts their motion along the edge. These are called edge states. It was demonstrated [72] that for $\hbar \omega > E_F$, new transitions from ZZ edge to bulk appear, increasing the losses in graphene. For 0.4 eV doped graphene flakes smaller than 20 nm, ZZ edge damping broadens the plasmon resonances enormously [73]. The edge damping for both ZZ and AC edges is particularly active when the plasmon energy $E_p = \hbar \beta c$ is above E_F , allowing the decay through excitation of those states [74] (with β the propagation constant of plasmons, see Sec. 3.2.2).

2.3 Production

Graphite is a stack of two-dimensional sheets of a hexagonal carbon atom lattice. These sheets are known as graphene and were first extracted quite simply by exfoliation by Geim and Novoselov [13]. To do so, they used adhesive tape on graphite, cleaving the stack of sheets. They repeated this step, producing a slice with fewer layers, until only one remained. Finally the graphene flakes larger than 1 mm and visible to the naked eye were deposited on a silicon wafer. Since then, progress to achieve mass production of graphene has been realized.

Graphene production is out of the scope of this thesis, but we provide a concise summary of the fabrication techniques. The methods of fabrication depend on the size of the graphene sheets needed for the application [75] and are revised in Sec. 2.3.1 for small sheets and in Sec. 2.3.2 for large sheets. Finally, we briefly review doping possibilities in Sec. 2.3.3.

2.3.1 Small graphene sheets

Small graphene sheets are used in functional coatings, conductive inks, batteries and supercapacitors. Their fabrication rests on exfoliation of bulk graphite (top-down method). It is mainly done in three ways.

- In liquid by sonication [76], shearing [77] or ball milling [78], with or without the use of a surfactant. Although it is easy to realize, the technique shows a low yield, a non-uniformity of the sheet sizes (0.5 to 2 μ m) and thicknesses (down to 5-7 layers minimum) and it suffers from surfactant or organic solvent impurities [75].
- By solid exfoliation, where graphene is produced by a simple ball milling of graphite in the presence of dry ice (solid phase of carbon dioxide) [79]. This functionalizes the edges that tend to repel each other to effectively exfoliate graphene sheets. The edges are defunctionalized by thermal annealing to end up with nearly pristine graphene sheets. These small graphene sheets are easy to realize and the technique demonstrates a high yield. However the sheet sizes and thicknesses are not uniform (one to five layers) and they agglomerate.
- By chemical species in between graphene layers in graphite to weaken the van der Waals interactions. This can be realized by oxidation-exfoliation-reduction [80] or by intercalation-exfoliation [81]. The first oxidises graphene: oxygen covalently bounds to graphene and this weakens the interlayer interaction. This technique, however, leads to severe damage of the graphene sheets due to the distribution of graphite oxide. Stirred in water, oxidized graphite is soluble and then useful for many applications. The second technique avoids the oxidation to produce high-quality pristine graphene: it intercalates chemical species that will expand graphite with microwave heating or chemical reaction. The final pristine graphene sheets are obtained by sonication. This process is easier to scale up, is more efficient and causes less pollution than the oxidation route. However, the thickness varies from one to ten layers.

This shows that mass producing one-layer graphene sheets with high yield and purity is a challenge for all the techniques.

2.3.2 Large graphene sheets

Large graphene sheets are more expensive to manufacture but are required in upcoming applications such as transparent electrodes in touch panels, displays and photovoltaic devices and for next-generation electronics and optoelectronics for flexible and wearable devices. They are based on a bottom-up approach:

• Chemical assembly [82], which is simply assembling small graphene sheets on various substrates, like a puzzle, but the film quality is really poor.

• Chemical vapour deposition (CVD) [83] : high temperatures decompose the hydrocarbons onto metal surfaces. The films are then transferred to transparent substrates either by etching away the metal, or by non-destructive electrochemical bubbling. However, CVD-graphene is more expensive than indium-tin oxide (ITO) films and the graphene sheets are less efficient (higher absorption and smaller conductivity). The poor quality comes from the inefficient transfer of graphene from metallic to other substrates, which remains the biggest challenge for CVD.

These techniques are much more expensive than the previous ones and a long way remains in order to produce large, high-performance and low-cost graphene sheets.

2.3.3 Doping graphene

As briefly described in Sec. 2.2.1, there are two main ways to achieve graphene doping: by electrostatic field gating or by chemical doping.

Electrostatic field gating

This doping technique consists in applying a gate voltage between a graphene sheet and a metal plate separated by a material. This material will bring electrons/ions to the graphene sheet when a bias voltage is applied (see Fig. 2.6a). In [62], a polymer matrix (poly(ethylene)oxide, PEO) containing mobile ions (Li⁺ and ClO₄⁻) is used. Those materials are simply called ion gel. Ion gels are widely used in optical components to achieve a transparent device. Another device from [64] is represented in Fig. 2.6b. Graphene is sandwiched between gold and an ITO gate, and rests upon glass.

Finally, one can realize an uneven doping on a graphene sheet following the proposal in [21]. Biasing the graphene sheet with an uneven metallic ground plane, separated by a dielectric spacer will result in a nonuniform static biasing electric field along the graphene (see Fig. 2.6c). This results in a nonuniform distribution of local carrier densities i.e. doping. In the same vein, biasing the graphene sheet with an even metallic ground plane, but separated by a non-uniform dielectric spacer will also induce an nonuniform doping level of the graphene sheet (see Fig. 2.6d).

Chemical doping

There are mainly two chemical doping techniques [65]: surface transfer doping and substitutional doping.

Surface transfer doping occurs through charge transfer from the adsorbed dopant to graphene. The dopant can be a substrate semiconductor such as SiO_2 , silicon carbide (SiC) or hexagonal boron nitride (hBN) [84]. Note that the doping effect may be dependent on the interaction with the substrate [65]. Furthermore, the dopant may also be a gas such as NO₂ [66] and it reveals graphene as a very sensitive gas sensor [85].

On the other hand, substitutional doping disrupts the graphene structure, replacing carbon atoms by dopant atoms. An illustration from [67] shows the process



Figure 2.6: Electrostatic field gating structure. (a) Graphene and the gate electrode are separated by a polymer matrix containing ions (POE), commonly named ion gel. When a bias voltage is applied, the ions reach their corresponding electrodes and dope graphene. (b) Transparent device achieved by ion gel and ITO. (c-d) Proposal of Vakil and Engheta in [21] to achieve nonuniform doping on a graphene sheet with (c) an uneven ground plane or (d) a nonuniform dielectric spacer.



Figure 2.7: The atomic configuration and corresponding Dirac cone of the graphene sheet. (a) The atomic and band structures of the pristine graphene. (b) The representative vacancy defects of the irradiated graphene. (c) The atomic and band structures of the graphene annealed in NH_3 after irradiation. The doped graphene is formed by substituting some C atoms with N atoms.

in Fig. 2.3.3. The pristine graphene has a honeycomb lattice and is described by two Dirac cones shown in Fig. 2.7a. Then, N^+ irradiation induces defects in the graphene sheets (Fig. 2.7b). Finally, the damaged graphene sheet is annealed in NH₃ and the carbon vacancies are replaced by N atoms, leaving a doped graphene sheet represented in Fig. 2.7c.

3

Optical concepts

This chapter introduces the basic theoretical concepts we will employ throughout this thesis. Sec. 3.1 prepares the field with Maxwell's equations leading to the Helmoltz equation and related concepts such as the Poynting vector, boundary conditions, and Fresnel coefficients. Afterwards, graphene plasmons are characterized and compared to standard surface plasmon polaritons in Sec. 3.2. Finally, cavities and their resonances are described in Sec. 3.3 together with the mathematical tools to describe them.

3.1 Photonics

Controlling the flow of light is an important goal of photonic applications. A significant group of photons carrying the information has to be modulated and travel from point A to point B without significant losses. The latter can be achieved with a typical dielectric optical fiber using photonic modes, but also with a metal using plasmonic modes. The classical behavior of light is described by the Maxwell equations (Sec. 3.1.1). These equations together with Ohm's law lead to an equivalence between the permittivity and the conductivity of a metal derived in Sec. 3.1.2. Sec. 3.1.3 describes the Helmoltz equation and defines the Poynting vector, which will be useful to derive the Fresnel coefficients in Sec. 3.1.5. Note that transverse magnetic and transverse electric polarizations are defined in Sec. 3.1.4.

3.1.1 Maxwell equations

The macroscopic interaction of light with material is described by the Maxwell equations. Assuming harmonically varying fields with $e^{j\omega t}$ [2] the equations are

$$\nabla \cdot \mathbf{D} = \rho_{\text{ext}} \tag{3.1}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{3.2}$$

$$\nabla \times \mathbf{E} = -j\omega \mathbf{B} \tag{3.3}$$

$$\nabla \times \mathbf{H} = \mathbf{J}_{\text{ext}} + j\omega \mathbf{D} \tag{3.4}$$

These equations link the four macroscopic fields **D** (the dielectric displacement), **E** (the electric field), **H** (the magnetic field), and **B** (the magnetic induction) with the external charge and current densities ρ_{ext} and \mathbf{J}_{ext} . Here, we distinguish between the external ($\rho_{\text{ext}}, \mathbf{J}_{\text{ext}}$) and internal (ρ, \mathbf{J}) charge and current densities, so that in total $\rho_{\text{tot}} = \rho_{\text{ext}} + \rho$ and $\mathbf{J}_{\text{tot}} = \mathbf{J}_{\text{ext}} + \mathbf{J}$.

Since we only consider a linear optical response of the material, we can define the relations

$$\mathbf{D} = \varepsilon_0 \varepsilon \mathbf{E} \tag{3.5}$$

$$\mathbf{B} = \mu_0 \mu \mathbf{H} \tag{3.6}$$

where ε_0 and μ_0 are the electric permittivity and magnetic permeability of vacuum, respectively, and are related to the speed of light c with the relation $c = \frac{1}{\sqrt{\varepsilon_0\mu_0}}$. The dielectric permittivity ε characterizes the electric field response in a bulk material and it is intrinsically related to the conductivity of the material. Similarly, the permeability μ is related to its magnetic field response. All throughout this work, we consider non-magnetic ($\mu = 1$) and isotropic media (ε and μ are scalar).

3.1.2 Conductivity-permittivity discussion

The electric and displacement fields can be linked via the polarization \mathbf{P} as follows

$$\mathbf{D} = \varepsilon_0 \mathbf{E} + \mathbf{P} \tag{3.7}$$

P describes the orientation of the dipole with the electric field in the material and is linked to the internal charge density via $\nabla \cdot \mathbf{P} = -\rho$. With the charge conservation $\nabla \cdot \mathbf{J} = -j\omega\rho$ we get

$$\mathbf{J} = j\omega\mathbf{P} \tag{3.8}$$

Combining the latter two equations with Ohm's law, which supposes that current is a linear function of the field ($\mathbf{J} = \sigma \mathbf{E}$), lead in Sec. 2.2.3 to Eq. 2.23. In the latter, we dropped the (\mathbf{r}, t) dependence for the sake of simplicity, but the rigorous development should be carried on in the Fourier domain, leading to [2]

$$\varepsilon(\mathbf{k},\omega) = 1 - \frac{j\sigma(\mathbf{k},\omega)}{\omega\varepsilon_0}$$
(3.9)

where we can simplify this general equation of the dielectric response $\varepsilon(\mathbf{k}, \omega)$ to the limit of a spatially local response ($\mathbf{k} = 0$) as $\varepsilon(\mathbf{k}, \omega) = \varepsilon(\omega)$. That simplification is valid when the wavelength in the material is larger than the size of the unit cell and the mean free path of the electrons.

Eq. 3.9 is extremely useful. Indeed, it is usual in theory to calculate currents and therefore σ , since it is easier to study the response of the charges to electrical fields, as for graphene in Chapter 2. However, measuring the motion of electrons at optical frequencies is difficult for experimentalists: the optical permittivity ε is easier to access via ellipsometry. Eq. 3.9 is thus interesting to validate theory with experiments.

For graphene simulations Eq. 3.9 is used to model graphene as a bulk permittivity. In general in COMSOL Multiphysics, graphene can be modeled as a surface current, with the conductivity of graphene. This is the case for the *Mode Analy*sis module to compute the dispersion, and for the scattering field formulation for incident plane wave on a structured graphene sheet. However the *Boundary Mode Analysis* module, which considers propagating modes does not handle surface currents and requires the effective bulk permittivity.

3.1.3 Helmholtz equation

In the absence of external stimuli ($\mathbf{J}_{\text{ext}} = 0$ and $\rho_{\text{ext}} = 0$) and using the bulk permittivity of materials, Maxwell's equations lead to a propagating wave solution. One can find the wave equation

$$\nabla \times \nabla \times \mathbf{E} = \mu_0 \omega^2 \mathbf{D} \tag{3.10}$$

equivalent to

$$\nabla \left(\nabla \cdot \mathbf{E}\right) - \nabla^2 \mathbf{E} = \varepsilon \varepsilon_0 \mu_0 \omega^2 \mathbf{E} = k_0^2 \varepsilon \mathbf{E}$$
(3.11)

where we used $\omega/c = k_0$. In media where ε is piecewise constant, Eq. 3.1 reduces to $\nabla \cdot \mathbf{E} = 0$, and the previous equation becomes the Helmholtz equation, identical for the **H** field under these approximations:

$$\nabla^2 \mathbf{E} + k_0^2 \varepsilon \mathbf{E} = 0 \tag{3.12}$$

$$\nabla^2 \mathbf{H} + k_0^2 \varepsilon \mathbf{H} = 0 \tag{3.13}$$

where the electric (magnetic) field profile solution depends on the boundary conditions at the considered interfaces (see Sec. 3.1.5).

In homogeneous media, the solutions (or modes) are plane waves:

$$\mathbf{E} = \mathbf{E}_{\mathbf{0}} e^{-j\mathbf{k}\cdot\mathbf{r}} \tag{3.14}$$

with the condition $\mathbf{E}_{0} \cdot \mathbf{k} = 0$ (from $\nabla \cdot \mathbf{E} = 0$) meaning that the field is transverse. **k** is called the wavevector and indicates the direction of propagation. Its amplitude is defined by

$$k = \frac{\omega}{c} \sqrt{\varepsilon_{\text{eff}}} = \frac{\omega}{c} n_{\text{eff}}$$
(3.15)

where n_{eff} is the effective refractive index and is the connection between ω and k, generally called the dispersion relation. In homogeneous media the effective refractive index is equal to the refractive index of the material $n_{\text{eff}} = n = \sqrt{\varepsilon}$.

The energy flow of a plane wave is represented by the Poynting vector. Since we work with harmonic fields, the time average of the Poynting vector is given by [86]

$$\langle \mathbf{S} \rangle = \frac{1}{2} \Re \left\{ \mathbf{E} \times \mathbf{H}^* \right\}$$
(3.16)

with * denoting complex conjugate and \Re the real part. It can be shown that the energy flow of a plane wave (Eq. 3.14) is given by

$$|\langle \mathbf{S} \rangle| = \frac{\Re \{n\}}{2\mu_0 c} |\mathbf{E}|^2 \tag{3.17}$$

Note finally that the work rate done by the external electromagnetic fields **B** and **E** on a charge q is $q\mathbf{v} \cdot \mathbf{E}$, where \mathbf{v} is the charge velocity. The magnetic field does not do work, since its force is perpendicular to the velocity. Therefore, for a continuous distribution of charge in a surface S, the total work is [86]

$$W = \int_{S} \mathbf{J} \cdot \mathbf{E} \ d^{2}x. \tag{3.18}$$

This power represents a conversion of electromagnetic energy into mechanical or thermal energy. The time average of this quantity is

$$\langle W \rangle = \frac{1}{2} \int_{S} \Re \left\{ \mathbf{J}^* \cdot \mathbf{E} \right\} d^2 x.$$
 (3.19)

Throughout this thesis this formulation of the absorption of the electromagnetic field is used to compute the absorption of graphene in COMSOL, when it is represented by a surface current. Note that with the ε formulation, there is a COMSOL variable to extract directly the dissipative losses.

3.1.4 Polarization

In Sec. 3.2 we consider the dispersion relation of guided modes. Such a guidance is realized with a particular symmetry of the materials and the fields. In this section we suppose a dielectric profile invariant in y and z directions i.e. $n(\mathbf{r}) = n(x)$ (see Fig. 3.1) and we choose the propagation along z. The field should then have the following form

$$\mathbf{E}(x, y, z) = \mathbf{E}(x)e^{-j\beta z}$$
(3.20)

$$\mathbf{H}(x, y, z) = \mathbf{H}(x)e^{-j\beta z}$$
(3.21)

where $\beta = k_0 n_{\text{eff}}$ is the propagation constant.

It turns out that in these conditions the Maxwell equations accept two sets of solutions: the transverse electric (TE) polarization contains only E_y together with


Figure 3.1: Schematic of the slab structure, with the orange arrow depicting the propagation direction. The non-zero components of the field are represented on the right for TE and TM polarizations.

 H_x and H_z and the transverse magnetic (TM) polarization contains only H_y together with E_x and E_z (see Fig. 3.1). The Helmholtz equations therefore become

$$\frac{d^2 E_y(x)}{dx^2} + k_0^2 \varepsilon E_y(x) = \beta^2 E_y(x) \quad \text{for TE}$$
(3.22)

$$\frac{d^2H_y(x)}{dx^2} + k_0^2 \varepsilon H_y(x) = \beta^2 H_y(x) \quad \text{for TM}$$
(3.23)

It can be shown that plasmon propagation is only possible for TM polarization [2]. Therefore, Sec. 3.2 will use Eq. 3.23 to derive the dispersion of a plasmonic mode along a metallic interface.

3.1.5 Fresnel coefficients

Before describing plasmonic modes we briefly define the Fresnel coefficients for an incident plane wave (Eq. 3.14), which are useful for Chapter 5. They are established considering the boundary conditions of the electric and magnetic fields at the straight interface of two media labelled 1 and 2:

$$\widehat{\mathbf{n}} \cdot \{\mathbf{D}_1 - \mathbf{D}_2\} = 0 \qquad \widehat{\mathbf{n}} \cdot \{\mathbf{B}_1 - \mathbf{B}_2\} = 0 \tag{3.24}$$

$$\widehat{\mathbf{n}} \times \{ \mathbf{E}_1 - \mathbf{E}_2 \} = 0 \qquad \widehat{\mathbf{n}} \times \{ \mathbf{H}_1 - \mathbf{H}_2 \} = 0$$

$$(3.25)$$

where $\widehat{\mathbf{n}}$ is a unit vector normal to the interface. Note that those boundary conditions are valid without external current and charge ($\mathbf{J}_{\text{ext}} = 0$ and $\rho_{\text{ext}} = 0$).

Suppose a plane wave propagating in medium 1 and impinging the interface with medium 2. Using the superposition principle the transmitted electric field in medium 2 (E_T) should match with the superposition of the incident (E_I) and the reflected (E_R) electric fields in medium 1. At normal incidence the transmission tand reflection r coefficients are identical for both polarizations (TE and TM) and are [87]

$$r \equiv \frac{E_R}{E_I} = \frac{n_1 - n_2}{n_1 + n_2}, \qquad t \equiv \frac{E_T}{E_I} = \frac{2n_1}{n_1 + n_2}$$
 (3.26)

with n_1 (n_2) the complex refractive index of medium 1 (2).

The reflectance is defined as the reflected energy $|\langle \mathbf{S}_R \rangle|$ with respect to the incident energy $|\langle \mathbf{S}_I \rangle|$:

$$\frac{|\langle \mathbf{S}_R \rangle|}{|\langle \mathbf{S}_I \rangle|} = \left| \frac{E_R}{E_I} \right|^2 = |r|^2 \tag{3.27}$$

where we used Eq. 3.17 to find the reflection coefficient in Eq. 3.26. Similarly, the transmittance is defined as the transmitted energy $|\langle \mathbf{S}_T \rangle|$ divided by the incident energy:

$$\frac{|\langle \mathbf{S}_T \rangle|}{|\langle \mathbf{S}_I \rangle|} = \frac{\Re \{n_2\}}{\Re \{n_1\}} \left| \frac{E_T}{E_I} \right|^2 = \frac{\Re \{n_2\}}{\Re \{n_1\}} |t|^2$$
(3.28)

The transmittance is therefore normalized by the permittivity of the two media. These two Eqs. 3.27 and 3.28 will be useful to describe the reflectance and transmittance of graphene plasmons in Chapter 5.

3.2 Plasmonics

Plasmonics takes advantage of the coupling of light with electrons in metals, and allows for localization of light into subwavelength dimensions enabling strong field enhancements. A metallic permittivity is often described by the Drude model, which considers a harmonic oscillation response of the free electrons of the metal with an electric field excitation. The permittivity of the metal is then [2]

$$\varepsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega \left(\omega - j\tau^{-1}\right)} \tag{3.29}$$

with ω_p the plasma frequency, which depends on the metal considered and defines the range of frequencies where the material behaves as a metal ($\omega < \omega_p$) or a dielectric ($\omega > \omega_p$). The losses are caused by the limited scattering lifetime of electrons τ .

The Drude model is useful in the next sections in order to compare the dispersion of plasmons in classical noble metals and graphene.

3.2.1 Surface plasmon polaritons

An interface between a dielectric and a metal can support surface plasmon polaritons (SPPs): the electric field of the light excitation can couple with the longitudinal oscillations of the electrons at the interface. Metals are characterized by a negative permittivity ($\varepsilon_m < 0$), while dielectrics have a positive permittivity ($\varepsilon_d > 0$).

Suppose the metal-dielectric interface represented in Fig. 3.2a with ε_m for x < 0and ε_d for x > 0. Solving for TM polarization (Eq. 3.23) leads to the following profile of the magnetic field

$$H_{y}(x,z) = Ae^{-j\beta z}e^{-\delta x} \qquad \text{for } x > 0 \tag{3.30}$$

$$H_y(x,z) = Ae^{-j\beta z}e^{+\gamma x} \qquad \text{for } x < 0 \tag{3.31}$$



Figure 3.2: (a) Simple metal (ε_m) - dielectric (ε_d) interface configuration. (b) Insulator-metal-insulator configuration with a metal layer of thickness d in the center (zone 1).

with

$$\delta = \sqrt{\beta^2 - \varepsilon_d k_0^2} \tag{3.32}$$

$$\gamma = \sqrt{\beta^2 - \varepsilon_m k_0^2} \tag{3.33}$$

where A is a constant.

Eq. 3.30 shows that we have a mode when δ is real i.e. $\beta^2 > \varepsilon_d k_0^2$. In this condition, the field is exponentially confined in the *x* direction (see Eqs. 3.30 and 3.31), with an even stronger confinement in the metal since $\gamma > \delta$. The other components E_x and E_z also decay exponentially and their analytical profile can be determined with Eqs. 3.3 and Eqs. 3.4.

Examining the E_x component of the field and requiring continuity, one can show

$$\frac{\delta}{\gamma} = -\frac{\varepsilon_d}{\varepsilon_m}.\tag{3.34}$$

Since δ and γ should be positive for exponential confinement, this relation implies that ε_d and ε_m should have opposite sign. In other words, these confined modes only exist at metal/dielectric interfaces. Finally, one can show that these SPP modes are only supported for TM polarization. Mathematically, the continuity conditions do not allow exponential confinement of the electromagnetic wave for TE at the metal/dielectric interface. Physically, the longitudinal oscillation of the electrons cannot couple with a TE electromagnetic wave since there is no electric field in the propagation direction for that polarization (see Sec. 3.1.4).

Combining Eqs. 3.32, 3.33 and 3.34, we derive the dispersion relation of SPPs:

$$\beta = k_0 \sqrt{\frac{\varepsilon_d \varepsilon_m}{\varepsilon_d + \varepsilon_m}}.$$
(3.35)

This dispersion describes the SPP at a simple metal/dielectric interface. However, for a sheet of graphene with effective permittivity ε_{eff} and thickness d, the dispersion is different. In that case, we consider the insulator-metal-insulator (IMI) system represented in Fig. 3.2b. Writing the fields with an exponential profile similar to Eq. 3.30 and introducing the boundary conditions, one can show for the dispersion [88]

$$\frac{\left(\varepsilon_{2}k_{1}+\varepsilon_{1}k_{2}\right)\left(\varepsilon_{3}k_{1}+\varepsilon_{1}k_{3}\right)}{\left(\varepsilon_{2}k_{1}-\varepsilon_{1}k_{2}\right)\left(\varepsilon_{3}k_{1}-\varepsilon_{1}k_{3}\right)}=e^{-2k_{1}d}$$
(3.36)

with

$$k_i = \sqrt{\beta^2 - \varepsilon_i k_0^2} \tag{3.37}$$

where i depicts the number of the medium.



Figure 3.3: (a) Dispersion of the SPP at a simple metallic interface (green solid line) and the asymmetric (solid blue) and symmetric (solid red) SPPs of the IMI in vacuum ($\varepsilon_d = 1$). The black dashed line represents the light line in the dielectric ε_d . The metal of the IMI is a 70 nm thick silver layer, modelled by the Drude model ($\omega_p = 7.2 \times 10^{15}$ rad/s). Corresponding H_y field at $\omega = 0.65\omega_p$ of the (b) symmetric mode, (c) simple interface mode and (d) asymmetric mode. z is the propagation direction.

The dispersion of a simple metal-vacuum interface (Eq. 3.35) and the dispersion of a 70 nm thick silver IMI (Eq. 3.36) in vacuum are represented in Fig 3.3a, considering a non-lossy Drude model of silver ($\omega_p = 7.2 \times 10^{15} \text{ rad/s}, \tau^{-1} = 0$). The dashed straight line represents the light line in vacuum. All the modes are represented on the right of that light line, indicating that they are guided modes (modes on the left of the light line would be radiative modes). There is only one mode for a simple interface and two modes for the IMI, one symmetric and one antisymmetric, defined following the profile of the transversal H_y field.

The corresponding magnetic fields are represented in Fig. 3.3b for the symmetric mode of the IMI, in Fig. 3.3c for the simple interface mode and in Fig. 3.3d for the asymmetric mode of the IMI. From the H_y field plots one can observe that the asymmetric mode is more strongly confined than the symmetric mode. That corroborates with the dispersions in Fig 3.3a: a stronger β implies that the exponential decays more rapidly (for IMI and simple interface, see Eqs. 3.32 and 3.37). Note that in the limit of $d \rightarrow 0$, only the asymmetric mode survives, as we will see in the next section with graphene.

3.2.2 Graphene plasmons

In this subsection, we derive the graphene plasmon (GP) dispersion when graphene is modeled as a line current boundary condition. Afterwards, we analyze the field profile and the GP dispersion. Finally, we conclude with a comparison between gold SPPs and GPs.

Derivation

The graphene plasmon dispersion can be derived considering the current $\mathbf{J} = \sigma \mathbf{E}$, with σ the surface conductivity discussed in the previous chapter. The derivation developed here closely follows the treatment in [89]. The field profile of a graphene plasmon in TM polarization should have the form

$$\mathbf{E}_{\mathbf{i}} = (E_{i,x}; 0; E_{i,z}) e^{-j\beta z} e^{-\delta_i |x|}$$
(3.38)

$$\mathbf{H}_{\mathbf{i}} = (0; H_{i,y}; 0) e^{-j\beta z} e^{-\delta_i |x|}$$
(3.39)

where *i* depicts the media number as showed in Fig. 3.4a and $\delta_i = \sqrt{\beta^2 - \varepsilon_i k_0^2}$.

That set of solutions can be injected in the Maxwell equations 3.3 and 3.4 to find six equations of the fields for the two dielectrics surrounding graphene. The subsequent system of equation can be reduced with the boundary conditions, which need to be adjusted to take into account the presence of graphene at x = 0 [90]:

$$\widehat{\mathbf{n}} \cdot \{\mathbf{D}_1 - \mathbf{D}_2\} = \rho \tag{3.40}$$

$$\widehat{\mathbf{n}} \cdot \{\mathbf{B}_1 - \mathbf{B}_2\} = 0 \tag{3.41}$$

$$\widehat{\mathbf{n}} \times \{ \mathbf{E}_1 - \mathbf{E}_2 \} = 0 \tag{3.42}$$

$$\widehat{\mathbf{n}} \times \{\mathbf{H}_1 - \mathbf{H}_2\} = \mathbf{J} \tag{3.43}$$

where $\widehat{\mathbf{n}}$ is a unit vector normal to the graphene sheet.



Figure 3.4: (a) Cross-section of the considered graphene sheet modeled as a boundary current line $\mathbf{J} = \sigma \mathbf{E}$ and surrounded by two dielectrics indicated with 1 and 2. Corresponding field components of a GP for a 0.6 eV doped free-standing graphene sheet ($\varepsilon_1 = \varepsilon_2 = 1$) and operating wavelength of 5 μ m : (a) E_x , (b) E_y and (c) H_z in arbitrary units.

Therefore, Eq. 3.42 leads to the continuity of E_z $(E_{1,z} = E_{2,z})$ and Eq. 3.43 introduces the graphene conductivity with the H field $(H_{1,y} - H_{2,y} = J_z = \sigma E_z)$. Finally, the dispersion of the graphene plasmon is the transcendental equation

$$\frac{j\sigma}{\omega\varepsilon_0} = \frac{\varepsilon_1}{\delta_1} + \frac{\varepsilon_2}{\delta_2} \tag{3.44}$$

$$=\frac{\varepsilon_1}{\sqrt{\beta^2 - \varepsilon_1 k_0^2}} + \frac{\varepsilon_2}{\sqrt{\beta^2 - \varepsilon_2 k_0^2}}$$
(3.45)

Throughout this thesis, we consider the nonretarded regime since we work in the near infrared range where plasmons are strongly confined ($\beta \gg k_0$). That leads to [60]

$$\beta = -\frac{\varepsilon_1 + \varepsilon_2}{2} \frac{2j\omega\varepsilon_0}{\sigma\left(\omega, E_F\right)} \tag{3.46}$$

Eq. 3.46 can be further simplified in the Drude approximation $(E_F \gg \hbar\omega, \text{ see Sec. 2.2})$ of the graphene conductivity

$$\beta = -\frac{\varepsilon_1 + \varepsilon_2}{2} \frac{2j\omega\varepsilon_0}{\sigma_{\text{intra}}\left(\omega, E_F\right)}$$
(3.47)

$$=\frac{\varepsilon_1+\varepsilon_2}{2}\frac{\pi\hbar^2\varepsilon_0}{e^2E_F}\omega\left(\omega-j\tau^{-1}\right)$$
(3.48)



Figure 3.5: (a) Dispersion of a GP at room temperature propagating along 0.2 eV, 0.4 eV and 0.6 eV doped free-standing graphene sheet. Scattering lifetime τ from Eq. 2.17. Dashed blue line for the Drude model approximation of the graphene conductivity. The light line (solid green line) covers the ordinate indicating that GPs are strongly confined. (b) Normalized dispersion ($k_F = E_F/\hbar v_F$) [50].

Here, a small wavelength (large β) leads to a high x confinement of the GP mode (see Eq. 3.39), accompanied by a collective surface charge oscillation, similar to SPPs. However the differences in electronic dispersion (linear Dirac cones instead of parabolic bands) lead to qualitatively different dispersions of SPPs and GPs. Therefore, throughout this thesis, we will distinguish GPs from SPPs.

Dispersion

The three field component profiles of the GP are represented in Figs. 3.4b, 3.4c and 3.4d for a current line at x = 0. Graphene is 0.6 eV doped and the operating wavelength is $\lambda = 5 \ \mu$ m. The continuity of the E_z component is verified, as for the discontinuity of the H_y field and the E_x field (see Eq. 3.40) at the surface current. The H_y field component is asymmetric, and similar to the IMI (see Fig. 3.3d). Indeed, taking carefully the limit $d \to 0$ in Eq. 3.36 when replacing the permittivity by the graphene conductivity (see Eq. 3.9) leads to the same dispersion obtained with the surface conductivity (Eq. 3.46). The mathematical details are provided in Appendix A. Therefore, modelling graphene with the effective permittivity or the line current is equivalent if the effective thickness is sufficiently small.

The GP dispersion is represented in Fig. 3.5a. We consider a free-standing graphene sheet at room temperature (T = 300 K) and with a scattering lifetime of electrons depending on the doping (see Eq. 2.17). The light line (green solid line) covers the ordinate indicating that the propagation constant of GPs is very large $(\beta \gg k_0)$. Incidentally the lateral confinement is stronger and the effective wavelength of GP is smaller than for the conventional SPPs (compare with Fig. 3.3a).

The frequency shift induced by graphene doping appears when examining the solid lines representing GPs for different doping (Fig. 3.5a). Indeed, a larger doping implies that GPs exist at higher frequencies (the region where graphene permittivity



Figure 3.6: Comparison of the plasmonic properties of graphene (modelised with the full dispersion relation 3.46) and gold plotting the lateral decay length (C) as a function of the normalized propagation length (L/λ_{eff}). The best properties are reached in the bottom right of the figure, for an important propagation length and a small decay length. The results are plotted for a 10 nm thick gold film and a 0.5 and 1 eV doped free-standing graphene sheet at room temperature. The color represents the wavelength (μ m). Gold refractive index values are from Johnson and Christy [91].

is negative is shifted via doping, see Fig. 2.5).

The blue dashed line in Fig. 3.5a represents the dispersion of a GP on a 0.4 eV doped sheet following the Drude approximation and behaves as a square root (see Eq. 3.48). The validity of the Drude approximation is verified with the normalized dispersion in Fig. 3.5b. The Drude approximated dispersion is close to the GP dispersion (solid lines) for energies $\hbar\omega \ll E_F$. Note that the dispersion in the Drude approximation (dashed line in Fig. 3.5b) stands for all doping.

Losses

The comparison between the dispersion of GPs (Fig. 3.5a) and SPPs (Fig. 3.3a) gives a hint on the possible superiority of GPs for potential plasmonics circuitry applications. Therefore, we propose a comparison with two parameters: the confinement (in order to judge the compactness of the integrated circuits) and the propagation length (comparing the damping strength). Note that it is important to avoid the comparison in the same frequency range since GPs operate in the mid-infrared range while SPPs operate in the visible range.

The losses are determined by the imaginary part of the propagation constant

 $(\Im \{\beta\})$. The propagation length L is therefore defined as [2]

$$L = \frac{1}{2\Im\left\{\beta\right\}} \tag{3.49}$$

Normalizing L by the effective wavelength of the SPP (or GP) [92] leads to the unitless value

$$\frac{L}{\lambda_{\text{eff}}} = \frac{\frac{1}{2\Im\{\beta\}}}{\frac{2\pi}{\Re\{\beta\}}} = \frac{\Re\{\beta\}}{4\pi\Im\{\beta\}}$$
(3.50)

In order to judge the lateral confinement of the mode, we introduce the lateral decay length (from the field profile in Eq. 3.39)

$$C = \frac{1}{2\sqrt{\beta^2 - \varepsilon_d k_0^2}} \tag{3.51}$$

where ε_d is the dielectric surrounding the metal. The lateral decay length (C) as a function of the normalized propagation length (L/λ_{eff}) is represented in Fig. 3.6. The ideal plasmon should be strongly confined (small lateral decay length) and should have a large propagation length.

The 10 nm thick gold SPP is weakly confined: the best lateral decay length is 20 nm for a poor propagation length $(0.3\lambda_{\text{eff}})$. On the other hand, the GP propagation length is two orders of magnitude better than gold for the same lateral decay length: $6\lambda_{\text{eff}}$ for 0.5 eV and $20\lambda_{\text{eff}}$ for 1 eV doped graphene sheet. Gold SPPs can achieve those values of high propagation length, but that is at the price of a poor confinement. Note that the operating wavelength of GPs is around 8 μ m, while gold SPPs operate mainly in the visible range ($\lambda_0 < 1 \mu$ m, dark blue in Fig. 3.6).

Therefore, there is a trade-off between high confinement and small propagation length, which needs to be chosen judiciously for the concerned application. However, we can conclude that graphene plasmons are better than gold plasmons for the high confinement and the large propagation length they offer.

3.3 Circuits and cavities

In this section, we further introduce the concept of flatland photonics, which was the starting point of this PhD thesis. Next, we review the methods developed to describe the cavity resonances and the coupling with those resonances: Sec. 3.3.2 discusses localized surface plasmon resonances, Sec. 3.3.3 Fabry-Pérot cavities, and Sec. 3.3.4 coupled mode theory.

3.3.1 Flatland photonics

SPPs enjoyed a huge development since their discovery in the early 1960s. The phenomenon has mainly been applied in sensors, detector and Raman spectroscopy, but also in printed metal circuits [93]. With the high confinement of GPs, it was suggested to replace integrated photonics circuitry [1]. However, even if the losses



Figure 3.7: (a) *E*-field for a GP wave at f = 30 THz along the ribbon like section of graphene with different doping, which splits into two paths ($L_1 = 1077$ nm, $L_2 = 560$ nm, $w = w_1 + w_2 + w_3 = 600 + 200 + 600$ nm) [21]. (b) Luneburg lens: phase of the E_y field of the GP at f = 30 THz (D=1.5 μ m, w = 75 nm, $L = 1.6 \ \mu$ m) [21].

in graphene are far smaller than in noble metals as discussed in Sec. 3.2.2, GPs still suffer from larger damping than their photonics counterparts [94] and its application in circuits has been restricted.

Nonetheless, graphene's optical properties are strongly tunable with doping, which is easily achieved by electrostatic field gating or chemical doping (Sec. 2.3.3). That amazing property implies that we can implement different conductivity zones on a single flake of graphene, enabling graphene to become a flatland platform for metamaterials and transformation optical devices [21].

In [21] Vakil and Engheta propose various applications where graphene plays the role of an optical platform: an infrared splitter or a Luneburg lens (Figs. 3.7a and 3.7b, respectively). This thesis is mainly based on that idea of flatland optics, where the GP propagation along a two-dimensional graphene sheet with various doping is considered.

Flatland photonics is not restricted to graphene and suggests a bright future for optoelectronics. Indeed, since the discovery of graphene, other interesting 2D materials have been uncovered: transition-metal dichacolgenides (TMDs), often playing the role of semiconductor, and hexagonal boron nitride (h-BN), often in the role of insulator [22]. These three materials thus offer the triad required to realize all 2D gated optoelectronic devices.

3.3.2 Localized surface plasmon resonances

In the previous sections we only considered propagating plasmons. However, metallic nanoparticles and graphene flakes can also sustain localized surface plasmon resonances (LSPR) [2]: an electromagnetic wave induces an oscillation of the electrons of the particle, which will be optimal at a certain frequency depending on the material and the geometry of the particle. These LSPRs are characterized by a high near field intensity.

LSPR modes can be excited by direct light illumination. Suppose that a particle is placed in a beam of electromagnetic radiation. The presence of the particle therefore results in an *extinction* of the incident beam, which is a sum of the *absorption* and the *scattering* by the particle [95, 96]. It is convenient to define the efficiencies of the extinction Q_{ext} , absorption Q_{abs} and scattering Q_{sca} :

$$Q_{\rm ext} = Q_{\rm abs} + Q_{\rm sca} \tag{3.52}$$

defined as the extinct, absorbed or scattered energy normalized by the incident energy E_{inc} (Eq. 3.17 for a plane wave) and G the particle cross-sectional area projected onto a plane perpendicular to the incident beam (e.g. $G = \pi r^2$ for a normal incident wave impinging a graphene disk of radius r):

$$Q_{\text{ext}} = \frac{E_{\text{ext}}}{E_{\text{inc}}G}, \qquad Q_{\text{abs}} = \frac{E_{\text{abs}}}{E_{\text{inc}}G}, \qquad Q_{\text{sca}} = \frac{E_{\text{sca}}}{E_{\text{inc}}G}.$$
 (3.53)

The scattered energy $E_{\rm sca}$ is obtained integrating the norm of the scattered field Poynting vector (see Sec. 3.1.3), and the absorbed energy $E_{\rm abs}$ is the energy absorbed by the particle, established in Eq. 3.19. Note that in the case of a small particle $(d \ll \lambda)$, the quasi-static approximation applies, and the scattering efficiency is negligible $(Q_{\rm sca} \approx 0)$ [2].

The LSPR resonance frequency depends on the shape of the particle and its constitutive metal. Considering a graphene flake, a given geometrical shape will result in plenty of modes numbered by l, each one characterized by a constant value η_l with respect to graphene conductivity $\sigma(\omega, E_F)$, frequency ω and the diameter of the flake D

$$\eta_l = \frac{-j\sigma\left(\omega, E_F\right)}{4\pi\varepsilon_0\omega D} \tag{3.54}$$

A more detailed mathematical description is provided in Appendix B, and graphene nanodisk modes are described in Chapter 7.

3.3.3 Fabry-Pérot cavity

In the case of a 1D graphene cavity (e.g. a graphene ribbon) the characterization of the resonances can be even more straightforward. In conventional photonics a basic Fabry-Pérot (FP) cavity is defined by two parallel partially transmitting mirrors [97]. When they are illuminated by a monochromatic wave, resonances appear inside the cavity and the transmittance spectra is characterized by local maxima. That interference phenomenon was discovered by Charles Fabry and Alfred Pérot in 1897: they examined the transmittance of a thin film, and they realized that this interference-induced contrast could be exploited in interferometry [87].

A FP resonance is achieved with constructive interference of the wave after a round-trip. If β is the wave vector of the light in the cavity medium, and l the length of the cavity, then the phase condition for a constructive interference is

$$e^{-j\beta l}e^{-j\varphi_{r1}}e^{-j\beta l}e^{-j\varphi_{r2}} = e^{-j2m\pi}$$
(3.55)

Or equivalently

$$2\beta l + \varphi_{r1} + \varphi_{r2} = 2m\pi \tag{3.56}$$

where m is an integer defining the order of the resonance, and φ_{r1} and φ_{r2} the phases induced by the reflection of the mirrors. As $\beta = \omega n/c$ with n the refractive index in the cavity, the frequency separation $\Delta \omega$ of the peaks is given by

$$|\Delta\omega| = \frac{c}{2nl} \tag{3.57}$$

This model agrees well with graphene ribbon resonances. A GP with propagation constant β resonates along the ribbon and is totally reflected at the edge of the graphene sheet, with a reflection phase of $\varphi_r = 0.85$ rad, independent of the frequency and the doping of the graphene ribbon [30]. This formalism is applied in Chapters 4, 5 and 6.

Note that the reflectance and transmittance of a wave incident to a FP cavity can be computed assuming the partial reflections and transmissions at each interface. This is implemented in Chapter 5 for a non-uniformly doped graphene sheet.

3.3.4 Coupled Mode Theory

In this section we introduce Coupled Mode Theory (CMT), which will be useful for the description of the sub-wavelength cavity resonances in Chapter 4. CMT is strictly valid when the width of the resonance is far smaller than the resonance frequency [98]. Therefore, it is well-suited for various problems, and amongst them for the description of plasmonic [99] and photonic Fano resonances [64] or for aperture antennas [100].

The theory is based on the coupling of modes in a time-dependent formalism of optical resonators [97]. We suppose a system (see Fig. 3.8) composed of a single-mode optical resonator coupled with m ports, which are single-mode waveguides in our case. The temporal equation of CMT is [98]

$$\frac{da}{dt} = \left(j\omega_0 - \frac{1}{\tau}\right)a + \left(\langle\kappa|^*\right)|s_+\rangle \tag{3.58}$$

$$|s_{-}\rangle = C|s_{+}\rangle + a|d\rangle \tag{3.59}$$

where a is the complex amplitude of the mode in the cavity with ω_0 its resonance frequency and τ the lifetime of the resonance. s_+ and s_- correspond to the input and output waves in the ports, respectively. The different couplings are embodied by $\langle \kappa |$ for the resonator input and by $|d\rangle$ for the resonator output. In addition, the incoming and outgoing waves in the ports can also couple through a direct pathway, accounted for with a scattering matrix C.

Here we use the bracket Dirac's notation: the ket $|d\rangle$ is a column vector

$$|d\rangle = \begin{bmatrix} d_1 \\ \vdots \\ d_n \end{bmatrix}$$
(3.60)

and the bra $\langle d |$ a row vector

$$\langle d| = \begin{bmatrix} d_1^* & \cdots & d_n^* \end{bmatrix}$$
(3.61)



Figure 3.8: Schematic of an optical resonator system coupled with multiple ports. The arrows indicate the incoming and outgoing waves. The dashed lines stand for the reference planes of the ports.

with * indicating complex conjugate.

If τ expresses the lifetime of the cavity resonance, τ^{-1} represents the total decay rate. In the system we consider in Chapter 4 this rate is decomposed in a sum of different decay channels, such as the coupling τ_c^{-1} , the absorption τ_a^{-1} or the radiative τ_r^{-1} decay rates. The total decay rate is then given by

$$\tau^{-1} = \tau_c^{-1} + \tau_a^{-1} + \tau_r^{-1} \tag{3.62}$$

Note that the amplitude a is normalized such that $|a|^2$ corresponds to the energy in the resonator. Since $|s_{i+}|^2$ ($|s_{i-}|^2$) corresponds to the input (output) power propagating in port i, the conservation energy in the lossless case ($\tau_a^{-1} = \tau_r^{-1} = 0$) means

$$\frac{da}{dt} = \langle s_+ | s_+ \rangle - \langle s_- | s_- \rangle \tag{3.63}$$

The other quantities (τ and κ) are supposed to be independent of the frequency: that assumption is valid as far as the waveguide dispersion around the resonant frequency is weak (i.e. the width of the resonance is smaller than the resonance frequency). Going to the frequency domain, which means that quantities oscillates with $e^{j\omega t}$, we can rewrite Eq. 3.58

$$a = \frac{\left(\langle \kappa |^* \right) | s_+ \rangle}{j \left(\omega - \omega_0\right) + 1/\tau} \tag{3.64}$$

The parameters $\langle d |$ and $\langle \kappa |$ can be determined using time-reversal transformation and energy conservation as described in [97] and one can find

$$\langle \kappa | = \langle d | \tag{3.65}$$

$$\langle d|d\rangle = 2/\tau_c \tag{3.66}$$

$$C|d\rangle^* = -|d\rangle \tag{3.67}$$

Eq. 3.65 shows a symmetry between input $(\langle \kappa |)$ and output $(\langle d |)$ coupling. Eq. 3.66 shows that output coupling is related to the coupling lifetime (in the lossless case, it is linked to the resonance lifetime since $\tau_c = \tau$). The last equation shows that the coupling constants in general cannot be arbitrary but are instead related to the direct scattering matrix C.

These conditions lead to the scattering matrix S

$$|s_{-}\rangle = S|s_{+}\rangle = C\left[1 - \frac{|d\rangle^{\ast} \langle d|^{\ast}}{j(\omega - \omega_{0}) + 1/\tau}\right]$$
(3.68)

using Eqs. 3.65 and 3.67 in Eqs. 3.58 and 3.59. In the special case of only one input waveguide, Eq. 3.68 can be rewritten

$$|s_{-}\rangle = S|s_{+}\rangle = C\left[1 + \frac{-2/\tau_{c}}{j(\omega - \omega_{0}) + 1/\tau}\right]$$
 (3.69)

using Eq. 3.66. This equation will be useful to describe the resonance in a graphene ribbon, as we will see in Chapter 4.

From the general Eq. 3.64 the resonance of our cavity is characterized by a Lorentzian shape and the quality or narrowness of the resonance is quantified by the quality factor Q

$$Q = \frac{\omega_0}{\Delta\omega_{\rm FWHM}} = \frac{\omega_0 \tau}{2} \tag{3.70}$$

with $\omega_{\rm FWHM} = 2/\tau$ the full width at half maximum of the Lorentzian peak.

Finally, the decay rate of the cavity resonance may be easily evaluated for a given Fabry-Pérot resonator. One can find that the coupling quality factor Q_c and absorption quality factor Q_a are [97]

$$\frac{1}{Q_c} = \frac{2}{\omega_0 \tau_c} = \frac{T v_g}{2\omega_0 l} \tag{3.71}$$

$$\frac{1}{Q_a} = \frac{2}{\omega_0 \tau_a} = \frac{2\alpha v_g}{\omega_0} = \frac{2\Im\left\{\beta\right\} v_g}{\omega_0} \tag{3.72}$$

where T is the transmittance from the port to the cavity, v_g the group velocity of the mode, l the length of the cavity, and where we replace the spatial decay rate of the field α with the imaginary part of the propagation constant of the mode $\Im \{\beta\}$.

3.4 Optical modeling with COMSOL

Along this thesis we propose the study of complex structures that are difficult or impossible to solve with the previously mentioned analytical models. In order to solve the optical response of such structures we rely on a computational method called the Finite Element Method (FEM) [101, 102, 103], implemented in the commercial package COMSOL [104]. The Radio Frequency (RF) module in COMSOL provides a wide range of modeling tools to solve time-harmonic electromagnetic field distributions. The *Eigenfrequency* node is used to find eigen-frequencies and their associated



Figure 3.9: (a) Triangular mesh (in white) of a structure composed of two materials represented in blue and red. (b) Effective refractive index of the graphene plasmon $(E_F = 0.4 \text{ eV} \text{ and } \lambda = 5 \ \mu\text{m})$ as a function of the effective thickness of the graphene sheet. n_{eff} converges for small graphene thickness to the accurate value computed using the graphene conductivity (blue dashed line).

eigen-modes in resonant cavities and the *Scattered Field Formulation* is employed to compute the optical response of various devices (both employed in Chapter 7). The *Mode Analysis* node is applicable for 2D and 2D cross-section of waveguides where it is used to find the mode dispersion, while the *Boundary Mode Analysis* is used to excite the propagating modes from a port boundary (both mainly applied in Chapters 4, 5 and 6).

Solving the differential Maxwell equations directly dealing with continuous variation of the electric field is impossible for digital computers because they handle discrete numbers. A way to get around that problem is the FEM, which relies on dividing the space into a large number of volume elements. In that limit the field relationship transforms to a large set of linear equations solvable by matrix inversion techniques. In FEM, triangular or tetrahedral meshes are mainly used: they can better approximate curved boundaries compared to cubic or squared meshes required by other techniques. A representation of a white meshed curved geometry is represented in Fig. 3.9a.

Defining the mesh correctly is primordial since an extremely thin mesh will unnecessarily lead to an interminable computational time, while a coarser mesh may not converge to a physical result. In general, fixing the maximum element size in a medium to $\lambda/6$ (where λ is the wavelength of the wave in the medium) provides accurate solutions. However SPs and GPs are characterized by a strong field decay and they need thinner meshes. Therefore its size should be adapted to the confinement of the plasmon along the interface. To determine a proper mesh, a convergence investigation can be performed.

Note that modeling graphene with an effective permittivity implies choosing an effective thickness. Fig. 3.9b shows the dependence of the effective refractive index on the thickness of a 0.4 eV doped graphene sheet at $\lambda = 5 \ \mu$ m. The solid red line converges for small thickness to the accurate value computed through the optical conductivity of graphene. In this case, we can assume that 0.5 nm is a sufficiently

converged value.

The structure modeled should have the smallest size to reduce the number of finite elements and therefore the computation time. The RF module of COMSOL provides a set of boundary conditions to achieve this goal. First, perfect mirrors can be implemented by the *Perfect Electric Conductor* and the *Perfect Magnetic Conductor* boundaries, which force the perpendicular electric field or magnetic field to zero, respectively. Second, it is possible to set up a periodicity between selected boundaries with *Floquet periodicity* for oblique incidence. Finally, the *Perfectly Matched Layers* apply a complex coordinate scaling to a layer surrounding the physical region of interest, which will absorb all the outgoing wave energy.

In our thesis we will use two kinds of light source in order to excite the modes in our designed structures. The most used is the port boundary, which can launch and absorb specific modes. The port boundary together with the *Boundary Mode Analysis* allows for the excitation of GPs when graphene is modeled by an effective permittivity, as used in Chapters 4, 5 and 6. The second technique is the *Scattered Field Formulation*: a background field (usually a plane wave) is set up by the user and COMSOL computes the scattered field. That formulation reduces the computation time since it does not require a solution for the incident field, as it is the case when the incident plane wave is excited by a port boundary. Moreover if the incident field is much larger in magnitude than the scattered field, the accuracy of the simulation improves. The *Scattered Field Formulation* is implemented in Chapter 7.

4

Ribbon cavities

4.1 Introduction

Graphene is a promising material for optoelectronic applications, with functional graphene circuits using plasmonic modes as an interesting developing field, as we discussed in Sec. 3.3.1. In this chapter we describe the coupling of graphene plasmons between a semi-infinite sheet and a nanoribbon, which gives deep reflectance dips at certain resonant wavelengths. In these circuits the useful phenomenon of critical coupling leading to zero reflectance can be engineered. Furthermore, the resonant wavelength is adjustable by tuning the gate voltage on graphene. The width of the ribbon (only 75 nm or smaller) and the confinement of the graphene plasmon mode (< 50 nm, depending on the operating wavelength and doping) lead to ultra-small, tunable optoelectronic circuits.

Indeed, the promise of photonic integrated circuits is jeopardized by limiting problems with downscaling and integration [1, 105]. Consequently, graphene circuits create great interest for their tiny size (one atom thick material and very small plasmon wavelength, see Sec. 3.2) and their tunability via electrostatic gating (see Sec. 2.3.3).

In recent works plasmons in graphene nanoribbons were studied fundamentally in single elements [24] or in gratings for a tunable optical response [25, 20, 26]. Bends and splitters were discussed in [27], and directional couplers were demonstrated in [28], with a switching behaviour in [29]. Additionally, it was shown that curved graphene sheets support plasmons with low radiation losses leading to flexible graphene circuits [106]. Lately, carbon nanotubes coupled to graphene nanodisks show an efficient way of launching and phase controlling the propagation of graphene plasmons [4]. These and our results provide the building blocks to develop graphene plasmon circuits for future compact plasmon devices with potential application to optical signal processing and infrared sensing.

In Sec. 4.2, we consider a graphene ribbon close to the end of a semi-infinite graphene sheet. Without ribbons, the graphene plasmon reflectance at the edge is total. That phenomenon is discussed in Sec. 3.3.3, and is exploited in Chapters 5 and 6. Here however, the evanescent exponential field can couple with the ribbon cavity. This leads to reflectance dips in the spectra, depending on the distance to the cavity and the graphene doping. A theoretical model is introduced via the coupled mode theory defined in Sec. 3.3.4, and its constitutive parameters are analysed and compared to the physical parameters. Finally, an electrostatic scaling law is derived, allowing for a straightforward prediction of the resonant wavelength.

Sec. 4.3 describes the plasmon reflectance for a ribbon placed on the top of a semi-inifinite sheet. Here, the theoretical development is based on parallel waveguide coupling, adapted to our structure through the scattering matrix formalism. The theoretical results fit well with simulations, and we observe a rich behaviour of the graphene plasmon reflectance as a function of the position of the cavity and the wavelength. The results are shown for two different lengths of the ribbon cavity and the reflectance dip behavior is explained via the anti-symmetric and symmetric supermodes of the waveguide coupler. Finally, the tunability of the graphene ribbon cavity is considered, and is qualitatively explained with a Fabry-Pérot resonance model.

Throughout this chapter, graphene is modelled as a thin layer of 0.5 nm thickness and its optical parameters are defined via the relative permittivity (see Sec. 2.2). The scattering lifetime of electrons in graphene is fixed to $\tau_g = 0.16$ ps. We operate in the range $\omega = 2$ to 3×10^{14} rad/s and $E_F = 0.2$ to 1 eV. We perform two dimensional simulations, and graphene plasmons are propagating in the horizontal direction. The excitation is performed by a port handled by *Boundary Mode Analysis* in COMSOL, and the other boundaries are modelled by *Perfectly Matched Layers* in order to account for the radiation losses (which are very weak in these devices).

Most of the work in this chapter was presented in [23].

4.2 Ribbon at the end of a sheet

In this section we study a two-dimensional system composed of a semi-infinite sheet of graphene (the access waveguide) and a small ribbon (serving as cavity) of width L at distance d from the sheet (Fig. 4.1). The background medium is air ($\varepsilon_{air} = 1$).

Without the cavity the injected graphene plasmon propagates along the graphene sheet and is nearly totally reflected at the edge: the plasmon is so confined that very little light is radiated [30]. When a cavity ribbon is placed near the end of the sheet, the evanescent field can couple into and light can resonate in the cavity. An example



Figure 4.1: Side view of the ribbon cavity with a semi-infinite sheet of graphene (the access waveguide) and a small ribbon (serving as cavity) of width L at distance d from the sheet. The graphene plasmon (GP) is excited by a port along positive x direction (s_+) , and the reflection is measured (s_-) .

of the magnetic field profile of a resonance (0.3 eV doped graphene at $\lambda = 7.95 \ \mu m$) is represented in Fig. 4.2a. For a specific wavelength we observe a reflection minimum (Fig. 4.2b). Note that the presented reflections are 'normalized': the losses in the access waveguide are neglected, but the losses in the cavity are accounted for.

4.2.1 Coupled Mode Theory

The properties of the cavity modes are modeled very adequately by the Coupled Mode Theory (CMT). CMT describes the coupling of ports with resonators as discussed in Sec. 3.3.4. In our case a single mode port couples with a cavity, with coupling strength characterized by τ_c^{-1} . The dissipative and radiative losses occurring in the resonator are represented by τ_a^{-1} and τ_r^{-1} , respectively. From Sec. 3.3.4, the equations of temporal CMT are

$$\frac{\mathrm{d}a}{\mathrm{d}t} = \left(j\omega_0 - \frac{1}{\tau}\right)a + \sqrt{\frac{2}{\tau_c}}s_+ \tag{4.1}$$

$$s_{-} = r_0 s_{+} + \sqrt{\frac{2}{\tau_c}} a \tag{4.2}$$

with

$$\tau^{-1} = \tau_c^{-1} + \tau_a^{-1} + \tau_r^{-1} \tag{4.3}$$

and where s_+ and s_- are the input and output amplitudes of the (plasmon) mode transmitted through the port (Fig. 4.1), a is the amplitude of the cavity mode, r_0 is the reflection at the edge of the graphene sheet without the resonator, and ω_0 is the resonant frequency.

Going to the frequency domain, which means that quantities oscillate with $e^{j\omega t}$,

and isolating s_{-}/s_{+} , we find

$$\frac{s_{-}}{s_{+}} = r_0 \left(1 - \frac{\frac{2}{\tau_c}}{j(\omega - \omega_0) + \frac{1}{\tau}} \right)$$
(4.4)

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leading finally to the reflectance

$$R = \left|\frac{s_{-}}{s_{+}}\right|^{2} = \left|r_{0}\right|^{2} \frac{(\omega - \omega_{0})^{2} + \left(-\frac{1}{\tau_{c}} + \frac{1}{\tau_{a}} + \frac{1}{\tau_{r}}\right)^{2}}{(\omega - \omega_{0})^{2} + \left(\frac{1}{\tau_{c}} + \frac{1}{\tau_{a}} - \frac{1}{\tau_{r}}\right)^{2}}$$
(4.5)

As previously mentioned, because of the remarkable impedance mismatch between the plasmon and free space, radiative losses at the edge of a sheet can be neglected, so $|r_0|^2 \approx 1$ and $\tau_r^{-1} \approx 0$. That last assumption is verified in simulations, since we simulate a system delimited with Perfect Matched Layers in order to account for possible radiative losses.

The Lorentzian shaped resonance of Eq. 4.5 has its minimum at $\omega = \omega_0$. In addition, one obtains a useful critical coupling when $\tau_a = \tau_c$: all the energy coupled to the cavity is dissipated, and the reflection goes to zero.

As a reminder from Sec. 3.3.4, the absorption lifetime τ_a can be modeled as follows

$$\tau_a = \frac{1}{v_g \Im(\beta)} \tag{4.6}$$

where v_g is the group velocity of the considered mode and $\Im(\beta)$ is the imaginary part of its propagation constant.

4.2.2 Results and discussion

We consider a cavity of width L = 75 nm and the doping of graphene (access waveguide and cavity) is shifted from $E_F = 0.2$ eV to $E_F = 0.5$ eV. The results are represented in Fig. 4.2b and we observe a shift of the fundamental resonance wavelength from $\lambda = 10$ to 6 μ m. Such doping tunability can be achieved by introducing a gate voltage on graphene, as discussed in Sec. 2.3.3. The realization is feasible since both the sheet and the ribbon are doped in the simulations: a gate voltage tunable graphene disk grating has already been realized in [64]. Although the graphene plasmon is not influenced by the ground plane because of its high confinement [21], the ion gel needed for doping (see Sec. 2.3.3) has a permittivity of $\varepsilon = 1.82$ and can slightly shift our results. Table 4.2c shows different fitted parameters from the first order minima of these spectra using Eq. 4.5 and the corresponding fit is shown in Fig. 4.2b for $E_F = 0.3$ eV (dots).

First of all one observes that the theoretical absorption lifetime τ_a (Eq. 4.6) matches the fitted value, and it increases with the graphene doping (compare third and sixth column of Table 4.2c). This is understood from the interband transitions in graphene, occurring above a threshold related to the Fermi energy ($\hbar \omega > 2E_F$), which can be shifted to higher frequencies by larger doping (see Sec. 2.2 for more details). Subsequently, increasing doping decreases interband transitions, and thus



Figure 4.2: (a) Normalized $|H_z|$ field for $E_F = 0.3$ eV at $\lambda = 7.95 \,\mu$ m. The size of the cavity is L = 75 nm and the distance from the sheet is d = 10 nm. (b) Simulated reflection spectrum for different doping (E_F) . Theoretical points are shown for $E_F = 0.3$ eV. Three orders of resonances are shown for each E_F . (c) Table with fitted lifetimes and resonant frequency ω_0 , and the calculated absorption lifetime. The quality factors are computed from the fitted parameters as $Q = \omega_0 / \Delta \omega_{FWHM}$.

decreases losses. This impacts the imaginary part of the graphene plasmon propagation constant: doping graphene decreases its value and from Eq. 4.6 one finds an increase of τ_a .

Note that the absorption lifetime is correlated to the scattering lifetime τ_g of electrons in the graphene sheet. As discussed in Sec. 2.2, the latter is linearly dependent to the graphene doping E_F (see Eq. 2.17). For the sake of clarity we arbitrarily fix $\tau_g = 0.16$ ps in the permittivity model but the minima are also observed for lower values of this parameter (which can happen for low quality graphene sheets). This is illustrated in Fig. 4.3a for $E_F = 0.4$ eV (theoretically $\tau_g \approx 0.4$ ps). Varying τ_g keeps the resonant frequency ($\omega_0 = 2.8 \times 10^{14}$ rad/s) and the coupling lifetime ($\tau_c = 1.7 \times 10^{-13}$ s) constant while τ_a changes. A limiting problem is that the Lorentzian becomes wider with the losses.

Next, from Table 4.2c we note that τ_c decreases when increasing the doping level. This is explained by the confinement of the plasmon. As we will demonstrate in Sec. 4.2.3, the resonances always occur for the same value of the real part of the propagation constant $\Re(\beta) = n_{\text{eff}}\omega/c$ (where c is the speed of light, and \Re depicts the real part). This implies that n_{eff} decreases when ω increases, leading to a less confined plasmon for larger angular frequencies. Since τ_c^{-1} theoretically stems from an overlap integral of the evanescent fields, this gives stronger coupling (τ_c smaller) when ω increases as shown in Table 4.2c. Because of the integration model of τ_c^{-1} , the coupling lifetime is also tunable via the distance d, increasing with this parameter. This is shown in Fig. 4.3b for $E_F = 0.3$ eV, with all other parameters constant ($\omega_0 = 2.4 \times 10^{14}$ rad/s, $\tau_a = 2.0 \times 10^{-13}$ s).

Because of these evolutions of τ_a and τ_c , for the configuration of Fig. 4.2b, critical coupling (R = 0) is reached when $\tau_a = \tau_c = 2.0 \times 10^{-13}$ s and $\omega = \omega_0 = 2.4 \times 10^{14}$ rad/s (see Eq. 4.5). Note that for a fixed doping of the access waveguide, only doping the cavity will give similar results. The absorption lifetime will be equal to the results of Table 4.2c since τ_a is only related to the absorption in the cavity. However, the coupling lifetime will be different than the former results, but will follow the same trends (smaller doping, longer τ_c). The equality $\tau_a = \tau_c$ will then be reached for another doping configuration. For instance, when the access waveguide is $E_F = 0.4$ eV doped and the cavity $E_F = 0.25$ eV, a critical coupling R = 0 is reached when $\omega_0 = 2.1 \times 10^{14}$ rad/s (or $\lambda = 9 \ \mu m$).

When examining the second order minima (Fig. 4.3a) we do not reach critical coupling. Indeed, losses are too important at these wavelengths (τ_a too small in comparison to τ_c to reach $\tau_a = \tau_c$). Consequently, the minimum of reflection is only about 0.5. Note that for 0.4 eV doping, the second order dip has a quality factor Q = 15. This value is higher than the one obtained for the first order dip (Q = 13), but this is mainly due to the larger resonance frequency ω_0 .

4.2.3 Dispersion

The particular resonance frequency is strongly determined by the plasmon properties. Indeed, in order to get a resonance in the cavity one needs constructive interference during a round-trip (see Sec. 3.3.3):

$$2\Re\left\{\beta(\omega)\right\}L + 2\varphi_r = 2m\pi \tag{4.7}$$



Figure 4.3: Simulated reflection in function of the wavelength. (a) The resonances are modified with the scattering lifetime of the electrons in graphene. For the first order minimum, critical coupling is obtained for $\tau_g = 0.12$ ps. $E_F = 0.4$ eV, L = 75 nm and d = 10 nm. (b) The resonances depend on the cavity-waveguide distance d. A critical coupling (R = 0) is observed for d = 10 nm. $E_F = 0.3$ eV and L = 75 nm.

Figure 4.4: Dispersion of the plasmon propagating along a graphene sheet for different doping levels. The black vertical solid line represents the required real part of β for a resonance in a 75 nm long cavity. The horizontal lines indicate the resonance frequencies. The vertical grey solid line stands for the expected resonances in a 30 nm cavity.

where β is the propagation constant of the plasmon mode, φ_r is the phase induced by the reflection at the end of the ribbon and m is an integer.

It turns out that φ_r is relatively constant (≈ 0.85 rad, see Sec. 3.3.3) in the typical mid-infrared range of frequencies, so for a particular L the resonance always occurs at the same value of the propagation constant $\Re\{\beta\} \approx 30 \text{ rad}/\mu\text{m}$. In Fig. 4.4 the graphene dispersion is plotted for different doping levels. The crossing between this value of $\Re\{\beta\}$ (vertical solid black line) with the different dispersion curves corresponds with the resonance frequencies fitted in the Table 4.2c from our simulations. This is a consequence of the specific properties of graphene: applying a gate voltage shifts the optical properties of the plasmon and thus the resonance frequency (see Sec. 2.2).

In fact, that property is a consequence of the scaling law of the modes of the graphene flakes, extensively discussed in Chapter 7 for nanodisks. We can show that the resonance in a graphene ribbon can be characterized by a scalar η , characterizing the considered resonance. A more detailed mathematical demonstration is available in Appendix B. Once determined for a given geometry, η is fixed and is equal to a function of the frequency ω , doping level E_F and the length L of the ribbon via the transcendental equation

$$\eta = \frac{-j\sigma\left(\omega, E_F\right)}{4\pi\varepsilon_0\omega L} \tag{4.8}$$

where $\sigma(\omega, E_F)$ is the optical conductivity of graphene. Using the non-retarded regime ($\Re\{\beta\} \gg k_0$, see Sec. 3.2.2) of the graphene plasmon dispersion, the propa-

gation constant of the graphene plasmon is (see Eq. 3.46)

$$\beta = -\frac{2j\omega\varepsilon_0}{\sigma\left(\omega, E_F\right)}\tag{4.9}$$

Combining Eqs. 4.8 and 4.9, we conclude

$$\eta = -\frac{1}{2\pi\beta L} = -\frac{\lambda_p}{4\pi^2 L} \tag{4.10}$$

This means that the product βL is a constant for the considered cavity mode, corroborating with the phase φ_r being constant in Eq. 4.7 as a function of doping and wavelength.

Thanks to these properties, one can easily predict the resonance frequency for a given cavity length. In the previous section, the resonance of a cavity of L = 75nm occurs with a propagation constant $\Re \{\beta\} = 30 \text{ rad}/\mu\text{m}$. In order to keep the βL product constant, a L = 30 nm length cavity requires $\Re \{\beta\} = 75 \text{ rad}/\mu\text{m}$. This value is the grey straight vertical line in Fig. 4.4. Therefore, the first-order resonance frequency for 1 eV doping will occur at $\omega_0 = 6.8 \times 10^{14} \text{ rad/s}$.

In order to improve the quality factor, one would need smaller cavities, i.e. larger resonance frequencies. However, going in this direction increases the losses (interband transitions occur when $\hbar \omega > 2E_F$). To counter that effect, a high doping of graphene is required, so when $E_F = 1$ eV a quality factor of Q = 42 is reached with a cavity of L = 30 nm, at $\lambda_0 = 2.8 \ \mu$ m. This is of the same order of magnitude as the quality factor of localized surface plasmon resonances of metals like silver $(Q \approx 30)$ or gold $(Q \approx 10)$ [107].

4.3 Directional coupler cavity

In this section we study a graphene ribbon (cavity) on top of a free-standing semiinfinite graphene sheet (Fig. 4.5b). Without the cavity, the plasmon propagates along the semi-infinite graphene sheet, is reflected at the edge, and creates a stationary wave. If we put a ribbon on top of the sheet the plasmon can couple with this cavity, leading to more complex interference effects and resonances.

We introduce a simple model in the following subsection, we discuss the behavior and we compare with simulations in the next subsection.

4.3.1 Theory

The coupling between two parallel graphene sheets separated by a distance d can be described by directional coupler theory. We suppose the two plasmonic waveguides are identically doped, so they support a plasmon with the same propagation constant β . The coupling between the plasmon modes is characterized by κ . The solution for the lossless case is in [108]. One can take the phase and the losses into account via the exponential factor $e^{-j\beta x}$:

$$s_1(x) = A(x)s_1(0) + B(x)s_2(0) = \left[\cos(\kappa x)s_1(0) - j\sin(\kappa x)s_2(0)\right]e^{-j\beta x}$$
(4.11)

$$s_2(x) = B(x)s_1(0) + A(x)s_2(0) = \left[-j\sin(\kappa x)s_1(0) + \cos(\kappa x)s_2(0)\right]e^{-j\beta x} \quad (4.12)$$

Figure 4.5: Transversal view of the considered structures. (a) In a first step, we consider a simple directional coupler between two graphene sheets, in a 4-port system. (b) The final structure we study. The graphene ribbon (cavity) of length L is placed at a distance d from the semi-infinite graphene sheet, and at a distance x_2 from the end of the sheet.

where $s_i(x)$ are the amplitudes of the plasmonic mode in the i^{th} waveguide and x is the propagation direction.

As a first step, we write the general Eqs. 4.11 and 4.12 as a 4-port system, see Fig. 4.5a. The input mode amplitude s_i for the i^{th} port is subscripted + and the output is subscripted -. That leads to the 4 × 4 scattering matrix

$$\begin{pmatrix} s_{1-} \\ s_{2-} \\ s_{3-} \\ s_{4-} \end{pmatrix} = \begin{pmatrix} 0 & 0 & B(x) & A(x) \\ 0 & 0 & A(x) & B(x) \\ B(x) & A(x) & 0 & 0 \\ A(x) & B(x) & 0 & 0 \end{pmatrix} \begin{pmatrix} s_{1+} \\ s_{2+} \\ s_{3+} \\ s_{4+} \end{pmatrix}$$
(4.13)

The zero elements in the scattering matrix express the absence of reflection in the simple case of Fig. 4.5a, while its symmetry comes from the reciprocity of the two equivalently doped graphene sheets.

From the scattering matrix of Eq. 4.13, the reflection of the considered system of Fig. 4.5b can be more easily derived. Firstly, the waveguide 1 is reduced to a ribbon of length L. That means that the ports 2 and 3 are replaced by a reflection r_0 : $s_{2+} = r_0 s_{2-}$ and $s_{3+} = r_0 s_{3-}$. In the same vein, all the matrix coefficients drops their x dependence for an L dependence, because the waveguide coupler only operates on the length of the ribbon. Secondly, the 4th port is also replaced by the reflection, but is defined at a distance x_2 from the graphene ribbon. Thus, accounting for the propagation, we get $s_{4+} = r_0 s_{4-} e^{-2j\beta x_2}$. Finally, the 1st port is at a distance x_1 from the ribbon, so the resulting input amplitude becomes $s_{1+} = s_{1-}e^{-2j\beta x_1}$. Therefore, the Eq. 4.13 adapts for the problem of Fig. 4.5b in

$$\begin{pmatrix} s_{1-} \\ s_{2-} \\ s_{3-} \\ s_{4-} \end{pmatrix} = \begin{pmatrix} 0 & 0 & B(L) & A(L) \\ 0 & 0 & A(L) & B(L) \\ B(L) & A(L) & 0 & 0 \\ A(L) & B(L) & 0 & 0 \end{pmatrix} \begin{pmatrix} s_{1-}e^{-2j\beta x_1} \\ r_0s_{2-} \\ r_0s_{3-} \\ r_0s_{4-}e^{-2j\beta x_2} \end{pmatrix}$$
(4.14)

Since we are interested in the port 1 reflection, we need to solve the problem for s_{1-}/s_{1+} . The scattering matrix problem of Eq. 4.14 can be rewritten as follows

$$\begin{pmatrix} 0\\0\\Be^{-2j\beta x_1}\\Ae^{-2j\beta x_1} \end{pmatrix} = \begin{pmatrix} 1 & 0 & -Br_0 & -Ar_0e^{-2j\beta x_2}\\0 & 1 & -Ar_0 & -Br_0e^{-2j\beta x_2}\\0 & A & 1 & 0\\0 & -B & 0 & 1 \end{pmatrix} \begin{pmatrix} s_{1-}/s_{1+}\\s_{2-}/s_{2+}\\s_{3-}/s_{3+}\\s_{4-}/s_{4+} \end{pmatrix}$$
(4.15)

Eq. 4.15 is a linear system of four equations in four unknowns. The solution is

$$R = \left| \frac{s_{1-}}{s_{1+}} \right|^2 = \left| \frac{\left(A^2 r_0 - r_0^3 e^{-4j\beta L} \right) e^{-2j\beta(x_1+x_2)} + B^2 r_0 e^{-2j\beta x_1}}{1 - A^2 r_0^2 - B^2 r_0^2 e^{-2j\beta x_2}} \right|^2$$
(4.16)

The reflectance is thus determined by the parameters L and x_2 , which we discuss in the next section, but also by β , κ and r_0 . The latter one (r_0) was mentioned in the previous section, and was relatively constant as a function of the frequency and the doping. The coupling constant κ is fitted from simulations of a directional coupler composed of two sheets of graphene separated by a distance d using Eqs. 4.11 and 4.12 (Fig. 4.5a). Finally, the plasmon dispersion is known from simulations. We already discussed the real part of the propagation constant $\Re(\beta)$ in Sec. 4.2.3.

4.3.2 Results and discussion

First we examine the effect of x_2 , the distance between the side ribbon and the end of the semi-infinite sheet (see Fig. 4.5b). The total size of the system $x_1 + L + x_2 =$ 200 nm is kept constant. By fitting simulations of a basic 1 eV doped graphene waveguide coupler separated by a distance d = 30 nm (Fig. 4.5a), we obtain that κ monotonously increases with the wavelength from about 5 to 11 rad/ μ m. Note that varying the distance d changes κ (for example κ varies from about 14 to 18 rad/m for d = 20 nm) and one can tune resonance frequencies with this parameter (not shown here, see Eq. 4.16). Note also that we fairly arbitrarily chose to work with high doping ($E_F = 1$ eV), but the same effect could be shown for smaller doping levels at higher wavelengths.

Fig. 4.6a and 4.6b show, respectively, the simulated and theoretical reflectance in color as a function of the wavelength λ and the position of the cavity x_2 for a cavity length L = 30 nm. First, one observes a good agreement of simulations with theory. Second, we observe a general augmentation of the reflectance with increasing wavelength. Indeed, because of the interband transition threshold, losses are less important at larger wavelengths: the propagation length of the plasmon is greater (see Sec. 3.2.2) and the reflected plasmon is, therefore, less damped.

Figure 4.6: Map of the simulated (a) and theoretical (b) reflectance of the system with a L = 30 nm length cavity varying the wavelength and its position (x_2) . The blue zones represent the reflectance minima of the first order cavity mode. (c) Theoretical map for a L = 60 nm length cavity varying the wavelength and its position (x_2) . Rhe second order minima appears for small wavelengths and the symmetric and antisymmetric type resonances appear at higher wavelengths.

Figure 4.7: Normalized $|H_z|$ field of the cavity L = 60 nm, for two different resonances: the symmetric (left: $x_2 = 57$ nm and $\lambda = 4.05 \ \mu$ m) and antisymmetric (right: $x_2 = 92$ nm and $\lambda = 3.53 \ \mu$ m) mode, respectively.

Let us focus on the reflectance minima of the map (blue zones in Fig. 4.6a). As a first approximation, one can predict the resonant wavelengths as explained for the isolated ribbon in the previous section via Eq. 4.7. In this way, the expected wavelength would be $\lambda = 2.8 \ \mu m$, which is close to what is simulated. However, with the directional coupling and the semi-infinite sheet reflection, there is an interference via two other parameters: the coupling constant κ and x_2 .

First of all, for some values of x_2 ($x_2 = 20$ and 61 nm e.g.), there are no resonances. This appears every half plasmon wavelength $\lambda_p/2 = 41$ nm. Indeed, when there is no cavity, there is a stationary wave on the ribbon with incoming and reflected waves. Subsequently, if the cavity is centred on top of a node, the resultant field in the cavity will be very small: the forward incoming wave and the backward reflected wave create destructive interference in the cavity.

When the cavity is not centred on a node, different possible interferences appear depending on the cavity position and the coupling strength κ . An interpretation is provided by the supermodes of the directional coupler. Depending on the position of the cavity, one will excite the symmetric (large field between the graphene sections, Fig. 4.7 (left)) or antisymmetric (node between the graphene sections, Fig. 4.7 (right)) supermode. The two supermodes have a different dispersion leading to two corresponding wavelengths. This explains the slanted shape of the blue zone, with two reflection dips e.g. at $\lambda = 2.75 \ \mu m \ (x_2 = 10 \ nm)$ and at $\lambda = 2.78 \ \mu m \ (x_2 = 28 \ nm)$.

That slanted shape of the blue zone is profoundly modified with the dispersion of the two supermodes. That assumption is further verified in Fig. 4.6c, which shows the reflectance as a function of λ and x_2 for a larger cavity length L = 60 nm. As the supermodes differ more when κ is large, the wavelength gap between the reflectance dips becomes broader (see Fig. 4.6c between $\lambda = 3.5$ and 4 μ m). Indeed, a larger cavity size implies a larger plasmon wavelength (see Eq. 4.8) and so a larger coupling κ (the plasmon is less confined transversally and it interacts more with the other ribbon). In this case, one witnesses a larger wavelength gap between two minima ($\lambda = 3.7 \ \mu$ m, $x_2 = 27 \ nm$ and $\lambda = 3.95 \ \mu$ m, $x_2 = 53 \ nm$). Note that the minima appear at higher wavelength than in Fig. 4.6b because larger plasmon wavelengths are acquired with larger free space wavelengths (see Sec. 3.2.2). The other blue zones around $\lambda = 2.5$ and 2.6 μ m are second order resonances.

Figure 4.8: Reflectance as a function of the wavelength for different doping levels of the graphene ribbon cavity. One observes the tunability of the reflectance dip with E_F . In this case, L = 30 nm, d = 30 nm and $x_2 = 40$ nm.

Finally, we investigate the reflectance tunability of such a structure. The reflectance as a function of wavelength is plotted for different doping levels of the cavity ribbon in Fig. 4.8. The doping level of the access waveguide is fixed to 1 eV and the one of the cavity ribbon is modified from 0.7 to 1 eV. The graph shows a baseline of increasing reflectance for larger wavelengths with localized dips with the resonance depending on the doping of the cavity, and shifting from $\lambda = 3.3$ to 2.75 μ m. That increasing reflectance is a consequence of the longer propagation length of graphene plasmons for larger wavelengths. In our structure (L = 30 nm, d = 30 nm and $x_2 = 40$ nm), a minimum of reflectance (R = 2.5%) can be reached for $E_F = 0.93$ eV when $\lambda = 2.84 \ \mu$ m.

The tuning behavior is explained in a first approximation by the phase resonance condition of Eq. 4.7 or the scaling law (Eq. 4.8) as previously discussed (Sec. 4.2.2): with a constant cavity length L, the resonance is achieved with the same β and, as depicted in Fig. 4.4, smaller doping implies smaller resonant frequencies. A more accurate model can be developed similar to Sec. 4.3.1 for two dissimilar waveguides.

4.4 Summary

This chapter demonstrates how the graphene plasmon reflectance at the end of a graphene sheet can be tuned by a graphene nanoribbon. When it is placed at a small distance from the graphene sheet (about 10 nm), the evanescent field can couple and one can get a total absorption in the cavity, and therefore zero reflectance. The quality factor of these nanoribbon cavities can reach 42, which is slightly larger than the quality factor of regular plasmonic cavities. These results are interesting for the development of graphene bio-sensors, where the graphene ribbon parameters can be

tuned with a detectable molecule.

The simulated results are rigorously explained by coupled mode theory, and its constitutive parameters (coupling and absorption lifetimes) as a function of the distance or the doping are qualitatively or quantitatively explained. A critical coupling is achieved with a graphene doping of 0.3 eV and a free space wavelength of 8 μ m. We showed that critical coupling with another doping can be achieved by adjusting the distance from the sheet and the size of the cavity. The position of the resonance is further explained with two models: first with the well-known Fabry-Pérot model, where we verified that the reflection phase of a graphene plasmon at the end of a graphene sheet is fairly constant with doping and wavelength and second, with a scaling law, showing that the same resonance always occurs for a particular value of βL .

Second, when the cavity is on top of a semi-infinite sheet of graphene, the reflectance resonances depend on the vertical distance and the horizontal position of the cavity, the cavity size, and the graphene doping. The theory of directional couplers is in good agreement with the simulated results. We show that the horizontal position of the cavity plays an important role creating or annihilating the resonance. The coupling constant is also essential, giving two resonant wavelengths for two different positions because of the symmetric and antisymmetric supermodes. Finally, we illustrate the strong tunability of the reflection thanks to the doping level of graphene.

5

Nonuniform doping

5.1 Introduction

The previous chapter was devoted to evanescent coupling with graphene cavities, or in other terms, holes in graphene sheets. Here, we consider another interesting feature of graphene: its doping tunability. Instead of structuring graphene sheets, we can modulate the graphene plasmon (GP) response with various spatial doping patterns. That is the key point of Vakil and Engheta's paper [21]. They demonstrate that, instead of shaping graphene sheets into ribbons for optical circuitry, one can locally dope graphene to achieve the same goal. This chapter aims to study this feature in more details, examining the reflectance and transmittance of GPs impinging on such interfaces.

This chapter also describes the behavior of GPs encountering graphene defects that are common in manufactured graphene sheets. Indeed, as described in Sec. 2.3, many flaws can appear on a graphene sheet, even with the most careful fabrication techniques. For example, with the chemical vapor deposition technique, the transfer from the copper substrate to another substrate can create carbon vacancies in the graphene sheets (see Sec. 2.3.2). These vacancies in the graphene lattice favors molecular adsorption, locally increasing the carrier density [109]. These local adsorptions are known as grain boundaries and locally distort the graphene doping [32, 110].

Figure 5.1: Doping configurations: (a) abrupt interface (Section 5.2), (b) tapered interface (Section 5.3), (c) cavity with abrupt interfaces (Section 5.4), (d) cavity with tapered interfaces (Section 5.6).

A numerical investigation of the phenomenon of nonuniform doping in back-gated voltage graphene ribbons was reported [111]. In addition, plasmon scattering caused by cracks in graphene have been demonstrated experimentally and theoretically [32, 33], while doping inhomogeneities have been approached numerically in [34] neglecting losses. Moreover, a clever way to redirect the GP and then avoid the defect was designed, based on the invisibility cloaking technique [112]. However, a complete study, including losses, of abrupt or smooth doping variations along the propagation direction in order to create plasmonic tapers or couplers [113] has not been reported to our knowledge.

We perform two dimensional simulations, and graphene plasmons are propagating in the horizontal direction. The excitation is performed by a port handled by *Boundary Mode Analysis* in COMSOL, and the other boundaries are modelled by *Perfectly Matched Layers* in order to account for the radiation losses (which are very weak in these devices).

In this chapter, we perform two dimensional simulations, and graphene plasmons are propagating in the horizontal direction. The excitation and the measurements of the reflection and transmission are performed by a port handled by *Boundary Mode Analysis* in COMSOL: the GP is excited by a port and propagates in the horizontal direction to the other port. The boundaries in the transversal direction use *Perfectly Matched Layers* in order to account for the radiation losses (expected to be very weak).

In Sec. 5.2 we consider abrupt doping changes along the GP propagation direction. The considered doping profile is represented in Fig. 5.1a. This profile demonstrates a simple behavior: stronger doping contrast leads to stronger reflection. We show that these results are explained by Fresnel-type coefficients of reflection, owing to the small radiation losses. A quadratic law of the reflectance is derived, only depending on the doping levels considered.

Afterwards, Sec. 5.3 addresses a smooth interface as depicted in Fig. 5.1b. These tapered interfaces drastically decrease the reflectance when increasing their length, but also decrease the transmittance because of the losses encountered in the transition zone. Therefore, there is a trade-off between interface contrast and transition length.

Then, we are prepared to consider local doping inhomogeneities, similar to the

defects discussed previously. Sec. 5.4 considers a cavity delimited by two abrupt doping changes, see Fig. 5.1c. We show that small homogeneities of 50 nm can achieve 70% of absorption. The absorption pattern is described with a Fabry-Pérot model, and the corresponding modes are plotted.

In Sec. 5.5 an even smaller cavity is designed to achieve total absorption. For constant doping, a GP encountering the edge of a graphene sheet is nearly totally reflected. Here, we show that locally doping the end of the graphene sheet on a 6 nm length distance is sufficient to achieve total absorption. This critical absorption is explained with an analytical multi-layer approach based on Fresnel coefficients.

Finally, Sec. 5.6 is devoted to smooth cavities as shown in Fig. 5.1d. Although small doping changes or longer cavities lead to near-zero reflectance, nearly 80% of reflectance is achieved for a particular point. These results are explained employing the simple interface case from previous sections.

Most of the work in this chapter was presented in [31].

5.2 Abrupt doping change

In this section we assume a sheet of graphene with two differently doped, abruptly delimited zones ($E_{F,\text{in}}$ and $E_{F,\text{out}}$ in Fig. 5.1a). The GP is excited along the sheet (in the half-plane with $E_{F,\text{in}}$) towards this abrupt transition, and is reflected when it encounters a variation in the sheet refractive index caused by the doping change. Note that we compute the reflectance at the interface i.e. the power returned into the counter-propagating plasmon mode (excluding the losses in the incident section). The simulated reflectance of the GP mode under various doping variations is represented by points in Fig. 5.2 at $\lambda = 6 \ \mu \text{m}$. GPs are excited from $E_{F,\text{in}}$ doped graphene (legend) and are reflected on a $E_{F,\text{out}}$ doped sheet (abscissa).

As expected, uniform sheets $(E_{F,\text{in}} = E_{F,\text{out}})$ lead to zero reflection. Furthermore, exchanging $E_{F,\text{in}}$ with $E_{F,\text{out}}$ gives nearly the same reflectance: there are negligible radiation losses, so the reflectance is directly related to the transmittance (R = 1 - T). Transmittance being reciprocal, the incidence direction does not have consequences on the reflectance. Finally, stronger reflectance is found for bigger graphene doping (and thus refractive index) differences.

The latter observation can be explained by a simple plane wave model. Indeed, the reflectance can be modeled by the Fresnel coefficient (solid lines in Figure 5.2) if we assume an effective medium characterized by the complex effective index \tilde{n}_{eff} of the propagating GP mode. It is given by $\tilde{n}_{\text{eff}} = \tilde{\beta}/k_0$ where $\tilde{\beta}$ is the complex propagation constant and k_0 the wave vector in vacuum. From Sec. 3.2.2, we know that for free-standing graphene in the non-retarded regime

$$\widetilde{\beta} = -\frac{2j\omega\varepsilon_0}{\widetilde{\sigma}\left(\omega, E_F\right)} \tag{5.1}$$

where $\tilde{\sigma}$ is the conductivity of graphene described in Sec. 2.2. Transmittance and reflectance are now completely described by the Fresnel coefficients established in

Figure 5.2: Reflectance of GPs at an abrupt interface of two different dopings. GPs propagate from $E_{F,in}$ to $E_{F,out}$. Points are simulated results and lines are computed with the Fresnel coefficients in Eq. 5.3. The grey dashed line stands for the approximate relation (Eq. 5.5).

Sec. 3.1.5

$$T_{12} = \left| \frac{2\widetilde{n}_1}{\widetilde{n}_1 + \widetilde{n}_2} \right|^2 \frac{n_2}{n_1} \tag{5.2}$$

$$R_{12} = \left| \frac{\widetilde{n}_1 - \widetilde{n}_2}{\widetilde{n}_1 + \widetilde{n}_2} \right|^2 \tag{5.3}$$

where \tilde{n}_1 and \tilde{n}_2 are the complex effective refractive indices of the incoming GP and the transmitted one, respectively and n_1 (n_2) is the real part of \tilde{n}_1 (\tilde{n}_2). The theoretical solid lines in Fig. 5.2 (corresponding to R_{12}) match the simulated points, except for large doping changes. In fact, the Fresnel coefficients in Eqs. 5.2 and 5.3 are no longer valid for heavily lossy media [114]. For 0.2 eV doped graphene at $\lambda = 6 \ \mu m$, the effective refractive index of the GP is $n_{\text{eff}} = 97 - 12j$.

The reflectance can be written in a simple quadratic function of the doping variation. When doping and wavelength are large, i.e. for $\hbar\omega \ll E_F$ (at $\lambda = 6\mu$ m for example, $E_F \gg 0.2$ eV), interband losses disappear (σ_{inter} vanishes), and the conductivity of graphene reduces to a Drude-like expression (see Eq. 2.15)

$$\widetilde{\sigma}(\omega, E_F) \approx \widetilde{\sigma}_{\text{intra}}(\omega, E_F) \approx \frac{e^2 E_F}{\pi \hbar^2} \frac{-j}{\omega - j\tau_g^{-1}}$$
(5.4)

Combining Eqs. 5.1 and 5.3, one obtains the simple formula

$$R \approx \left(\frac{E_{F,\text{out}} - E_{F,\text{in}}}{E_{F,\text{out}} + E_{F,\text{in}}}\right)^2 \tag{5.5}$$
which does not depend on the wavelength in this approximation. An example for $E_{F,\text{in}} = 0.4 \text{ eV}$ is shown in Fig. 5.2 (grey dashed line) justifying the accuracy of the approximation. Note that deviations from the simulations are larger for smaller doping ($E_{F,\text{out}} < 0.4 \text{ eV}$), as expected from the approximation validities.

5.3 Tapered interface

In the previous section, the sudden interface led to strong reflections for large doping differences. In order to enhance the transmission, we examine here smooth doping transitions, depicted in Fig. 5.1b. We assume a cosine-like shape of the doping profile $E_F(x)$ with a transition zone of length d

$$E_F(x) = \frac{E_{F,\text{in}} + E_{F,\text{out}}}{2} \left[1 + \frac{E_{F,\text{in}} - E_{F,\text{out}}}{E_{F,\text{in}} + E_{F,\text{out}}} \cos\left(\frac{\pi x}{d}\right) \right]$$
(5.6)



Figure 5.3: Reflectance (solid lines) and transmittance (dashed lines) of GPs with a smooth cosine-shaped transition of doping. GP propagates from 1 eV to a range of smaller doping going from 0.2 eV to 0.8 eV. Points are simulated reflectance and lines are from theoretical considerations (multi-layer approach).

The reflectance (solid lines) and transmittance (dashed lines) as a function of the transition zone length d are represented in Fig. 5.3. The GP is always incident from doping $E_{F,in} = 1$ eV, and gradually reaches the doping level indicated in the legend. As a check, when d approaches zero, the reflection tends to the same reflection obtained for an abrupt interface (Fig. 5.2). Indeed, the reflection increases drastically when increasing the doping difference, corroborating Eq. 5.5. Longer taper lengths decrease the reflectance drastically, down to nearly zero for d > 100 nm. However, the transmittance does not always increase as a function of d, there is even an optimum point for smaller $E_{F,out}$. Indeed, absorption during propagation is important for the GP, especially at lower doping. Therefore, there is a trade-off between interface contrast and propagation length, leading to a maximum of transmission at a certain taper length. For example, the variation from 1 eV to 0.2 eV doping is optimal for a d = 100 nm transition.

The solid and dashed lines in Fig. 5.3 are computed via a one-dimensional multilayer Fresnel approach implemented with a software module (CAMFR): the smooth doping of the profile is discretized in 100 layers. Each layer thus corresponds to a particular graphene doping and the effective refractive index of the layer is determined with Eq. 5.1. The model fits very well with the rigorous simulation results, except for large doping changes, as also observed in the previous section.

Finally, we checked that the reflectance does not vary much upon change of incidence direction in this system (maximum 2%, not shown here). This equivalence is not straightforward. Indeed, although radiation losses are negligible, significant absorption occurs in the tapered section. Therefore, the relation T = 1 - R does no longer hold, and the transmittance reciprocity does not imply reflectance reciprocity.

5.4 Cavity with abrupt interfaces

In this section, we assume a localized inhomogeneity in the doping: the sheet has doping $E_{F,\text{in}}$ except in a particular zone of width d where the doping abruptly changes to $E_{F,\min}$ (Fig. 5.1c). The simulation results are represented in Fig. 5.4a and show that strong absorption can be achieved in this small zone depending on the width d and doping level ($E_{F,\min}$). The operating wavelength remains $\lambda = 6 \ \mu \text{m}$ and the sheet is $E_{F,\text{in}} = 1 \text{ eV}$ doped.

For small doping of the cavity ($E_{F,\min} < 0.2 \text{eV}$), graphene does not support GPs anymore at the considered wavelength and we observe a fairly constant absorption as a function of the size. Non-absorbed light is simply reflected, nothing is transmitted, except for cavities smaller than 20 nm: reflectance goes to zero in these conditions (not shown here, light tunnels to the exit waveguide, similar to the evanescent field coupling with the ribbon cavity described in Chapter 4).

For doping between 0.2 eV and 0.6 eV, bands of absorption show up. This is a Fabry-Pérot (FP) cavity: in order to get constructive interference during a round-trip, one needs (see Sec. 3.3.3)

$$2\Re\left\{\widetilde{\beta}\right\}d + 2\varphi_r = 2m\pi \tag{5.7}$$

where *m* is an integer for the mode order and φ_r is the phase induced by the reflection (found via simulations and varying monotonously from 0.6 to 0 rad for $E_{F,\min} = 0.2$ to 1 eV). The solutions of this equation for the five first modes are depicted as black lines in Fig. 5.4a and follow the simulated absorption maxima closely.

The norm of the electric field in the vertical direction for the two first modes is represented in Figure 5.4b, clearly indicating the FP-type profile. Note that strongly localized electric fields appear on the doping interfaces: this can be connected to



Figure 5.4: (a) Simulations of the absorption in the cavity (color plot) as a function of the size of the cavity and its doping level $E_{F,\min}$ (from 0 to 1 eV). Outside the cavity $E_{F,in} = 1$ eV. The black lines represent the five first theoretical modes of the FP cavity. (b) The norm of the electric field component in the vertical direction for the two points indicated in (a).

nanoribbon modes reported in [115] and [24]. It was also observed for back-gated graphene circuits in [21].

For higher doping of the cavity ($E_{F,min} > 0.6 \text{ eV}$), reflectance is too weak to create a strong resonance. That corroborates with the single interfaces we examined, see e.g. Eq. 5.5 or Fig. 5.5 in Sec. 5.2. For example, a doping difference of 0.4 eV only provides maximum 10% of reflection.

It is worth noticing that a small cavity of only 50 nm can absorb nearly 70% of the incoming light (point 1 in Fig. 5.4a). This absorption may be further improved by the use of an asymmetric FP cavity, i.e. when the outgoing graphene part has a different doping level than the incoming one. As an even tighter alternative, however, we examine a design with a semi-infinite sheet of graphene in the next section.



Figure 5.5: (a) A doping defect at the edge of a semi-infinite graphene sheet. The doping profile is represented underneath. (b) The norm of the total electric field for the resonance at $\lambda = 4 \ \mu m$ and $E_{F,cav} = 0.25$ eV for a d = 6 nm cavity.

5.5 Asymmetric cavity

In this section we propose an ultra-thin perfect absorber. In the previous FP symmetric cavity, the GP was trapped for many round trips and was gradually absorbed. However, we only reached about 70% absorption. To achieve critical coupling the easiest way is to limit the number of output ports. Thus, we design a semi-infinite graphene sheet of $E_{F,in} = 1$ eV doping (Fig. 5.5a), with the end zone (of length d) doped with $E_{F,cav} = 0.25$ eV. The latter value is chosen so that GPs are very lossy between $\lambda = 3$ and 5 μ m. Varying the size d of the low-doped zone, one obtains the reflectance represented in Fig. 5.6.

For d = 6 nm, one gets zero reflectance, meaning 100% absorption since there are almost no radiation losses (because of the remarkable impedance or density of states mismatch between the GP and free space). Incidentally, we observe a highly confined field in this zone for the mode without reflection (Figure 5.5b).

This resonant wavelength can be explained again via a Fabry-Pérot-type model. Indeed, one fits a quarter of the effective wavelength in the cavity: $d = \lambda_p/4 \approx 6$ nm, with $\lambda_p \approx 26$ nm being the GP wavelength at $\lambda = 4 \ \mu m$ and $E_F = 0.25$ eV.

Even if a 6 nm cavity is really small, one could envisage smaller cavities. Indeed, since the media involved are very lossy, from [116] one might get a resonance for cavities of ≈ 1 nm, but this is beyond the application regime of pure electromagnetic simulations, and ab initio computations would be more appropriate.

The calculations in Fig. 5.6 can be approximated by a multi-layer approach. The reflection in a 3-layer model becomes

$$R = \left| r_{12} + t_{12} t_{21} r_{23} e^{-2j\widetilde{\beta}d} + t_{12} t_{21} r_{23}^2 r_{21} e^{-4j\widetilde{\beta}d} + \dots \right|^2$$
(5.8)

$$= \left| r_{12} + t_{12} t_{21} r_{23} e^{-2j\widetilde{\beta}d} \{ 1 + r_{23} r_{21} e^{-2j\widetilde{\beta}d} + \dots \} \right|^2$$
(5.9)

$$= \left| r_{12} + t_{12} t_{21} r_{23} \sum_{n=0}^{\infty} \left(r_{23} r_{21} e^{-2j\tilde{\beta}d} \right)^n \right|^2$$
(5.10)

$$= \left| r_{12} + \frac{r_{23}t_{12}t_{21}e^{-2j\beta d}}{1 - r_{21}r_{23}e^{-2j\tilde{\beta}d}} \right|^2$$
(5.11)



Figure 5.6: Plot of the reflectance of GPs with a change of doping from $E_{F,\text{in}} = 1$ eV to $E_{F,\text{cav}} = 0.25$ eV at the end of a semi-infinite graphene sheet with length d indicated in the legend. Critical coupling is reached when the size of the smaller doping zone is 6 nm. The grey dashed line stands for analytical Fresnel reflectance.

with 1 indicating the semi-infinite graphene part, 2 the edge defect and 3 the air half-space, and where we used $|r_{23}r_{21}| < 1$ for the infinite series convergence. It is known that when the GP reaches an air edge, it is totally reflected [34, 71, 32], the phase shift induced by reflection being $\approx 0.27\pi$ [30, 23], so $r_{23} = e^{-0.27j\pi}$. The other contributions are computed via Fresnel coefficients as done in Sec. 5.2. The solution for d = 6 nm is plotted in Fig. 5.6 (grey dashed line). The match is not perfect because of the evanescent fields involved due to the small size of the cavity, which are not taken into account in Eq. 5.11.

5.6 Cavity with tapered interfaces

In this section we consider that the inhomogeneity is not abruptly shaped (as in Sec. 5.4) but smooth (Fig. 5.1d). A theoretical study on this geometry was performed for larger wavelengths neglecting losses [34], but here we focus on $\lambda = 6 \ \mu m$ with losses included. The reflectance and the transmittance are represented in Fig. 5.7a and 5.7b depending on the size of the cavity d and its minimum doping $E_{F,\min}$. The transition from the maximum $E_{F,\min} = 1$ eV doping to the varying minimum doping $(E_{F,\min})$ is the cosine shape of Eq. 5.6.

As a check, when the minimum doping is close to 1 eV, the GP is totally transmitted, since it is the doping of the main sheet. Moreover, for cavities longer than 200 nm, the reflectance is zero. That corroborates with Fig. 5.3 where the reflectance is close to zero for tapers longer than 100 nm independently of doping changes (the



Figure 5.7: Simulated color plot of (a) reflectance and (b) transmittance of a GP through a smooth cavity as a function of its size d and the minimum doping of the modulation $E_{F,\min}$. The GP is excited from an $E_{F,\min} = 1$ eV doped graphene sheet at $\lambda = 6 \ \mu$ m.

cavities here are composed of two tapers, compare Figs. 5.1b and 5.1d). On the other hand, when the minimum doping is lower than 0.15 eV, the reflection is small, and decreases even more for larger defect sizes. This leads to a large absorption reaching 100% for a cavity bigger than 250 nm (Figs. 5.7a and 5.7b, reflectance and transmittance are nearly zero): the doping change is smooth enough to avoid reflection (see Sec. 5.3) and the GP is gradually absorbed, with increasing losses when the doping decreases.



Figure 5.8: Plot of the normalized H field for the maximum reflection condition: $E_{F,\min} = 0.2$ eV and d = 75 nm. GP excited from the left. Strong reflection is observed.

An interesting feature is the maximum reflectance (around 80%) for a small defect of 70 nm and 0.2 eV minimum doping. The norm of the electric field is plotted in Fig. 5.8. A very small field amplitude is transmitted, a large standing-wave in reflection is observed. This is due to constructive interference in the reflection, which is only possible with the limited defect size here.

Finally we point out that in Fig. 5.7a, other orders of reflection are barely visible for larger cavity sizes (from 200 to 300 nm). Their intensity is very weak since the GP are more damped with longer transition zones. This is consistent with [34] where higher orders are clearly observed because the operating wavelength is larger, and losses are missing.

5.7 Summary

This chapter discusses the reflectance and transmittance of GPs through various doping change geometries. For all doping configurations the GP behavior is analogous to a simple plane wave Fresnel-type model, considering an effective medium index. The plane wave analogy works well since the GP is the guided mode and there are no radiation losses. However, the model deviates for smaller doping i.e. lossier media since Fresnel coefficients need small corrections in this range.

In the case of an abrupt doping change, an analytic approximation of the reflectance is derived and only depends on the considered doping level. This formula shows why the reflectance is stronger with higher doping contrast and why reflectance is independent from the incidence direction. However, small deviations for weak graphene doping appear because the approximation becomes less valid. In order to decrease the reflectance, we then consider a smooth doping transition. We calculate an increased transmission and find an optimal size of 100 nm for the 1 to 0.2 eV taper, which is a trade-off between high reflection for abrupt changes and high absorption for long transition zones. Indeed, reflectance diminishes rapidly when increasing the transition length, and is already negligible for a 150 nm length transition.

By combining these types of interfaces we investigate various cavity situations. We first describe a localized abrupt doping change leading to about 70% absorption for a 50 nm length cavity, which is described by a simple Fabry-Pérot model. Afterwards, the cavity is optimized to reach total absorption. The GP is impinging the graphene edge, and is absorbed in the cavity formed by a local doping of only 6 nm length. Finally, cavities composed of two tapered interfaces are considered, demonstrating 80% reflectance for a particular configuration.

We envisage this work to be useful for graphene circuitry. On the one hand, these theoretical considerations help to understand the behavior of GPs encountering grain boundaries, constituting a strong damping source. On the other hand, mastering the doping profiles is important to control GPs. We demonstrate the possibilities of trapping or transmitting the plasmon modes. Finally, the asymmetric cavity could lead to nano-sensor applications if an adsorbed molecule locally changes the electronic density.

6

Patterned graphene edges

6.1 Introduction

In Chapter 5, we found that graphene plasmons encountering the straight edge of a graphene sheet are totally reflected, with a special phase. The anomalous reflection phase was described later by Nikitin *et al.* in [30]. However, that particular total reflection and phase is valid for a straight edge only; what happens for a patterned edge? The previous two chapters were devoted to two-dimensional structures, where we experimented with doping and gaps between graphene sheets and ribbons. Here, we consider a more complex three-dimensional structure.

In this chapter we show that the reflection is strongly determined by the edge structure. The examined ribbon grating case offers longitudinal and lateral edge mode Fabry-Pérot resonances that drastically decrease the reflection for particular ribbon length and width combinations, whereas it would have been nearly 100% for a non-structured edge. The edge mode coupling is associated with phase changes, influencing the positions of the longitudinal cavity modes. Furthermore, the rapidly changing transmittance beyond such a resonance leads to particular points of critical and near-zero reflection.

In Sec. 6.2, we describe the considered structure. The semi-infinite graphene sheet is connected to a finite graphene ribbon grating. We characterize the grating

by length and width and we fix the period in order to avoid diffraction. As a first step, we assume a simple model with a fixed wavelength and doping.

Sec. 6.3 analyzes the reflection of a graphene plasmon impinging the patterned edge, which constitutes our main result in this chapter. We describe the complex reflection pattern observed by a simple Fabry-Pérot analytical model. Then, a deeper investigation follows on the constitutive variables of the Fabry-Pérot model. We analyse the case of a semi-infinite graphene ribbon connected to a semi-infinite graphene sheet. The mode dispersion, the reflection and transmission phase together with the reflectance and the transmittance are taken into account. This deeper analysis concludes that lateral edge modes play an important role for specific ribbon widths.

The final Sec. 6.4 discusses the broad validity of the results. The fixed doping condition is released and we show that the doping does not influence the results if they are judiciously normalized.

Most of the work in this chapter was presented in [35]

6.2 Graphene edge structure

The particular pattern we study is represented in Fig. 6.1. It is a grating at the end of a semi-infinite free-standing sheet in air, with an edge described as a ribbon array with elements of length l, width w and period p. This can be experimentally realized for a sheet of graphene obtained with chemical vapour deposition in order to have a large sheet of high quality, as discussed in Sec. 2.3.2. The micro-ribbon array constituting the edge can be shaped using standard optical lithography followed by oxygen plasma etching [117]. The grating is excited via an in-plane graphene plasmon (GP), perpendicular to the edge (incidence along y axis, Fig. 6.1).



Figure 6.1: The patterned edge on a graphene sheet, with period p, length l and width w. The norm of the incident graphene plasmon magnetic H-field is depicted in the background.

For this problem, full-wave three-dimensional calculations are needed. A single period is simulated, delimited by *Perfect Magnetic Conductor* boundaries in the x direction (since the GP propagates in parallel to the boundary), with *Perfectly Matched Layers* on the y and z boundaries in order to account for radiation losses (see Fig. 6.1 for axes). Graphene's optical properties are defined by the graphene permittivity established in Sec. 2.2. We fix the free-space wavelength to $\lambda_0 = 10 \ \mu \text{m}$ and the doping level to $E_F = 1 \text{ eV}$, but this is fairly arbitrary and does not influence our conclusions, as we will see in Sec. 6.4. The scattering lifetime of electrons is $\tau_g = 0.16 \text{ ps}$. Thus, the graphene permittivity for an effective thickness of t = 2 nmis $\varepsilon = -185 - 6.4j$, and the corresponding plasmon effective wavelength is $\lambda_p = 1.16$ μm . Note that the effective permittivity technique is used since the Boundary Mode Analysis node of COMSOL does not compute modes along a conductive boundary, see discussion in Sec. 3.1.2.

In order to make the analysis easier, we deliberately consider a grating period to avoid diffraction. That condition is achieved for a period $2\pi/p > 2\pi/\lambda_p$. Thus we choose $p = 0.95\lambda_p = 1.1 \ \mu \text{m}$.

6.3 Results and discussion

The reflectance of an in-plane plasmon mode perpendicularly incident on the grating constitutes our main result (Fig. 6.2), as we observe a rich characteristic as a function of the ribbon length l (normalized to λ_p) and the width w (normalized to the period p). Here, the reflectance is the power returned into the counter-propagating plasmon mode (excluding the losses in the incident section), as we fixed $p = 0.95\lambda_p$ there is no diffraction towards other directions. If the graphene edge was unstructured, it would give total reflectance, whereas now zones with near-zero reflectance appear (blue zones), which means near-complete absorption as the radiation losses remain very small. In the following we explain these features through models that are as compact as possible.

Fabry-Pérot model

The considered structure (Fig. 6.1) can be viewed as a graphene sheet connected with a finite graphene ribbon grating (GRG) of length l. A schematic of this problem is represented in Fig. 6.3. As one can see, even this simple picture implies many parameters to consider. However a simple model can be considered.

The reflectance dips appear when the plasmon is trapped in the grating: a GRG mode is reflected back and forth with internally constructive interference. This Fabry-Pérot (FP) resonance condition is

$$2\Re\{\beta\}l + \varphi_{r_{21}} + \varphi_{r_{23}} = 2m\pi \tag{6.1}$$

where $\Re{\{\beta\}}$ is the real part of the propagation constant of the (fundamental and only) GRG plasmonic mode, $\varphi_{r_{21}}$ and $\varphi_{r_{23}}$ are the reflection phases for the fundamental mode at the two different interfaces of the GRG cavity, and *m* is an integer. Upon calculating $\Re{\{\beta\}}$, $\varphi_{r_{21}}$ and $\varphi_{r_{23}}$ (through separate simulations), we can represent the first five orders with white lines in Fig. 6.2.



Figure 6.2: Reflectance of a GP impinging the patterned edge, as a function of normalized ribbon width w/p and length l/λ_p . Theoretical Fabry-Pérot modes depicted with white lines.

The single-mode description agrees well with the full-structure simulation results, with deviations mainly for widths w > 0.7p that increase for smaller lengths l. Indeed, at the interfaces evanescent modes are generated (similar to metallic edges [118]) and they are expected to have a larger influence for short cavities (small l) and larger w (better match with edge mode, discussed below). Note that the complex modal reflection (or transmission) is written following the convention $r_{ij} = \sqrt{R_{ij}}e^{-j\varphi_{r_{ij}}}$.

Parameter analysis

We are now in a position to analyze the behavior of the resonances in more detail, using as few parameters as possible, amongst others via the parameters of Eq. 6.1. Fig. 6.4 represents the real part of the effective refractive index ($\Re\{n_{\text{eff}}\}$) of the fundamental GRG mode (blue line) as a function of the ribbon width w (where $\beta = 2\pi n_{\text{eff}}/\lambda_0$). This mode (which builds up the resonance in the GRG) is highly confined (large n_{eff}) for narrow ribbons, while for wider ribbons the mode ultimately reaches the index of a pristine graphene sheet plasmon (red dashed line). If not embedded in a grating (single ribbon, black dashed line), the dispersion of the fundamental single ribbon mode approaches the edge mode dispersion (green dashed line). Note that they dissociate at $w \approx 0.7p$ when the ribbon modes in the GRG start to interact with their neighbours via the edge modes (edge mode and GRG dispersion crossing). These edge modes will play an important role in our description.

The following parameters we analyse are simulated with a simpler system than



Figure 6.3: Top view with the relevant reflection and transmission parameters at the two interfaces. Lateral edge modes c_e are sketched.



Figure 6.4: Dispersion of the relevant modes as a function of normalized width (w/p). The fundamental single ribbon mode is identical to the ribbon grating mode, except when w/p approaches 1. In one hand, the single ribbon mode tends to the edge mode effective refractive index as the field is more distributed on the edges. On the other hand the ribbon grating mode tends to the effective refractive index of the sheet mode since w/p = 1, physically corresponding to a graphene sheet. On the right, the norm of the electric field is plotted for the different mode at w/p = 0.75. From top to bottom: single ribbon, ribbon grating, edge and sheet modes.

the one depicted in Fig. 6.3. In order to extract the reflection and transmission from a GRG to a sheet, we simulate a semi-infinite graphene sheet, connected to a semi-infinite GRG. Then, we measure the reflection and transmission when the mode is incident from the GRG or the graphene sheet. The phases in Eq. 6.1 have a direct influence on the modelled resonances (white lines in Fig. 6.2). Fig. 6.5a represents the reflection and transmission phases of the GRG plasmon at the interfaces of the FP cavity, see Fig. 6.3 for their definitions. Fig. 6.5a shows two regions where the phases rapidly change (w/p < 0.2 and w/p > 0.7). These fast changes are responsible for the slope variations of the FP resonances (blue regions, Fig. 6.2), also explicit in the single-mode model (white lines, Fig. 6.2). They are both connected to a strong coupling with lateral (x direction) FP resonances via edge modes (discussed below). The variation in the reflection phase [119] of the GRG at w = 0.75p ($\varphi_{r_{23}}$, black dashed line) characterizes another lateral FP cavity resonance at the end of the GRG (into air, see below). Note that $\varphi_{r_{23}}$ tends to the value $0.85 = 0.27\pi$ rad for a straight edge (w/p = 1), as already mentioned in Sec. 3.3.3.

Finally, Fig. 6.5b shows the transmittance and reflectance from GRG to a graphene sheet $(T_{21} \text{ and } R_{21})$ and the reflectance at the end of the grating (R_{23}) , see Fig. 6.3. By reciprocity $T_{12} = T_{21}$, so T_{21} (red line) also applies for transmittance from graphene sheet to GRG. Similarly to the phase plot, there are two regions where the reflectance and transmittance rapidly change (w/p < 0.2 and w/p > 0.7), making those points particular.

We also calculated the average of the magnetic field z component at the lateral edges of the sheet-grating interface (Fig. 6.5b, green-dot line, right axis). At this interface only part of the energy stays in the y directed longitudinally propagating modes $(R_{21} + T_{21} < 1)$. With negligible radiation losses and without other longitudinally propagating modes, the remaining energy flows towards evanescent modes, determined as $c_e = 1 - R_{21} - T_{21}$ (green line, Fig. 6.5b). The evanescent contribution c_e mainly corresponds to the edge mode, propagating in the lateral x direction [115] (sketched in Fig. 6.3).

This is correlated with the averaged edge magnetic field (green line and dots in Fig. 6.5b): two peaks are observed at w = 0.15p and 0.7p. The corresponding electric fields are represented in Fig. 6.6a and Fig. 6.6b and they indicate a third order and a first order lateral FP resonance along the edge, respectively. Accordingly, the Poynting vectors (green arrows, Fig. 6.6a and 6.6b) exhibit an enhanced energy flow along the x direction edge. Note that the second order resonance, characterized by a node at the boundaries of the period, is not allowed by the symmetry of the excitation. At the other end of the grating (to air), a similar edge mode lateral FP resonance at the end of the ribbons is responsible for the R_{23} reflectance dip around w = 0.75p (black dashed line, Fig. 6.5b).

Synthesis

Now we have the elements to understand the complete structure. The first order (lateral) resonance (Fig. 6.6b) is responsible for the disappearance of the (longitudinal) FP modes around w = 0.7p in Fig. 6.2 (a band of higher reflectance appears, in between low reflectance zones). Indeed, in Fig. 6.5b the transmittance T_{21} falls to nearly zero, while edge mode coupling reaches 40%, which weakens the GRG cavity mode. Note that this particular width w = 0.7p corresponds with the crossing point of the edge mode and the grating ribbon dispersion (Fig. 6.4). The coupling with edge modes is also present at the other interface (towards air) of the GRG: the



Figure 6.5: (a) Reflection and transmission phase shift at the interfaces. (b) Reflectance and transmittance at the two interfaces. c_e represents coupling with evanescent modes. The right axis shows the z-component magnetic field average at sheet-grating interface edge (dots).



Figure 6.6: Top view of the graphene sheet with the real part of the y-component of the electric field for (a) w = 0.15p and (b) w = 0.7p, corresponding to lateral (x-direction) FP resonances of the edge mode. The excitation comes from the top via a semi-infinite GRG and is transmitted to a semi-infinite graphene sheet at the bottom. Poynting vectors are sketched in green arrows, with the arrow length depicting the vector norm in logarithmic scale.

reflection R_{23} is nearly 100% except around 0.75*p*, where the energy is absorbed at the edge (dashed black line in Fig. 6.5b).

The other lateral edge mode resonance (peak around w = 0.15p in Fig. 6.5b) however, does not spoil the reflectance dip in Fig. 6.2 at w = 0.15p. This is understood considering that the transmittance $T_{12} = T_{21}$ is not affected in this case (red line in Fig. 6.5b). The altered parameters are the reflectance of the GRG plasmon R_{21} where a dip appears (blue line in Fig. 6.5b), and the slope of the phase $\varphi_{r_{21}}$ (blue line in Fig. 6.5a). The latter influences the white line slopes in Fig. 6.2 for w < 0.15p.

Finally, we focus on the brutal changes of transmittance T_{21} and reflectance R_{21} when w/p > 0.8 (Fig. 6.5b). At the final value (w/p = 1) the grating becomes a graphene sheet, leading to the absence of an interface, thus total transmittance and zero reflectance. Therefore, in the range w/p > 0.8, the losses remain fairly constant, while reflectance and transmittance change very rapidly. This leads to a particular value of w/p that achieves critical coupling (losses equal coupling), whereas the other values are under- or overcoupled. This explains the localized minima (tight blue spots in Fig. 6.2) in this regime, in contrast to the more extended FP resonances for w/p < 0.8 (elongated blue minima zones).

6.4 Broad validity range

The results in the previous section were computed with fairly arbitrary wavelength and doping ($\lambda = 10 \ \mu m$ and $E_F = 1 \text{ eV}$). In this section, we show that the phenomena are very similar for different dopings and wavelengths, if the structural parameters are scaled with the plasmon wavelength of the graphene sheet, so the conclusions are more general.

6.4.1 Doping

To asses the broad validity range, we plot the graphene plasmon reflectance as a function of the normalized width in Fig. 6.7 for different doping. The length of the GRG is fixed to $l/\lambda_p = 0.93$ (ordinate of Fig. 6.2). Incidentally, the 1 eV curve (magenta in Fig. 6.7) is a horizontal cross-section of Fig. 6.2. All results in Fig. 6.7 are normalized by the graphene sheet plasmon wavelength, which depends on the doping. For example, 0.2 eV doping leads to a plasmon wavelength of $\lambda_p = 212$ nm, thus the width w is normalized by the period $p = 0.95\lambda_p = 201$ nm and the length $l = \lambda_p = 0.93\lambda_p = 212$ nm.



Figure 6.7: Graphene plasmon reflectance as a function of the normalized width for different doping with a fixed length of the GRG $(l = 0.93\lambda_p)$. The operating wavelength is 10 μ m. The normalization is thus different for each doping: for 0.2 eV, $\lambda_p = 0.21 \ \mu$ m, for 0.4 eV, $\lambda_p = 0.46 \ \mu$ m, for 0.6 eV, $\lambda_p = 0.70 \ \mu$ m, for 0.8 eV, $\lambda_p = 0.93 \ \mu$ m and for 1 eV, $\lambda_p = 1.16 \ \mu$ m.

The reflectance of the graphene plasmon (Fig. 6.7) behaves similarly for varying doping. The three dips represent three different orders of the FP cavity discussed in Sec. 6.3. We observe similar curves for all doping values, except for 0.2 eV, where the dips are less pronounced. In the following, we explain the invariant position of the peaks and the damping.

First, we show that the doping has no influence on the FP resonance positions. That assumption is correct if the parameters in Eq. 6.1 remain largely invariant with doping variation. Examining $\Re\{\beta\}$, we plot the propagation constant of the GRG (normalized by the propagation constant of the graphene sheet) as a function of the normalized ribbon width in Fig. 6.8a. The results of doping varying from 0.2 to 1 eV overlap each other. Therefore, no deviation is expected if the length of the GRG is normalized by the sheet plasmon wavelength $(2\pi/\beta_{\text{sheet}})$.

Note that Fig. 6.8a shows that the dispersion of a GRG (β_{GRG}) only depends



Figure 6.8: (a) Propagation constant of the GRG mode normalized by the propagation constant of the graphene sheet mode as a function of the width of the ribbon. The curves and points overlap. (b) Reflectance and (c) transmittance from the GRG to the graphene sheet. The doping influence is stronger with small doping.

on the graphene plasmon propagation constant and the width of the ribbon. That observation corroborates with the scaling law for single ribbons discussed in [24]. Finally, the remaining terms in Eq. 6.1 that we need to discuss are the reflection phases. The plots are not shown here, but the curves are close to each other.

Second, we focus on the reflectance and the transmittance at the interface between the graphene sheet and the GRG. They are represented in Figs. 6.8b and 6.8c for different doping levels. The curves are similar, with a deviation for smaller doping. That behavior can be explained by the losses. Indeed, with smaller doping and constant wavelength, damping is stronger: the condition $\hbar\omega \ll 2E_F$ is no longer fulfilled and interband transitions start to occur and, as already demonstrated in various works [114, 120] and discussed in Chapter 5, the increase of the imaginary part of the propagation constant has an influence on the reflection coefficients. If the phase of the reflection plays a role for the position of the dip, the amplitude has an influence on their depth (see also Eq. 5.11 from the previous chapter). That explains why the dips in Fig. 6.7 are less pronounced for 0.2 eV doping (reflectance is about 20% stronger), but occur at the same w/p.



Figure 6.9: Graphene plasmon reflectance as a function of the normalized width for different wavelength with a fixed length of the GRG $(l = 0.93\lambda_p)$. Graphene is 1 eV doped. The normalization is thus different for each wavelength: for $\lambda = 4 \ \mu m$, $\lambda_p = 0.18 \ \mu m$, for $\lambda = 8 \ \mu m$, $\lambda_p = 0.74 \ \mu m$, for $\lambda = 10 \ \mu m$, $\lambda_p = 1.16 \ \mu m$ and for $\lambda = 12 \ \mu m$, $\lambda_p = 1.67 \ \mu m$.

6.4.2 Wavelength

Following the same normalization procedure of the previous section for w and l, we perform simulations for different wavelengths, with doping fixed to 1 eV. The reflectance as a function of the normalized width is represented in Fig. 6.9. As we can see, the FP condition remains invariant, since the dip minima positions match for the different wavelengths. From the previous discussions it indicates that the reflection phase and the propagation constant are invariable with wavelength, within the considered normalization.

Similarly to the doping influence, the depth of the reflectance dips in Fig. 6.9 are adjusted by wavelength changes (about 20% under the considered wavelength range). That could be explained with an investigation of the amplitude of the reflection and transmission, as previously discussed (but not shown here).

Summarizing, owing to the similarities between various dopings (Fig. 6.7) and wavelengths (Fig. 6.9), with judicious normalization, we can conclude that the reported graphene grating behaviour is fairly general.

6.5 Summary

Plasmon reflectance at the edge of a graphene sheet is profoundly influenced by its shape. Indeed, the examined ribbon grating demonstrates a nearly zero reflection for particular size configurations, where it would have been nearly 100% for a non-structured edge.

The reflection is strongly influenced by the length and the filling factor of the ribbon grating. That is explained with simple Fabry-Pérot resonances, examining the propagation constant and the phase of reflection and transmission. However, the reflectance and transmittance are not trivial. We show that they are strongly influenced by lateral edge modes. This assumption is further supported by integrating the field along the edge and by analyzing the Poynting vector.

Although the edge mode lateral resonances hamper the longitudinal Fabry-Pérot resonances, they are also responsible for particular points of critical and near-zero reflection caused by the rapidly changing transmittance beyond such a longitudinal resonance.

Finally, we examine the influence of the doping on the reflectance, transmittance and effective index. We show that the effective index of the grating guided mode is independent of the doping, when normalized by the effective index of the graphene sheet plasmon. On the other hand, the doping influences slightly the reflectance and the transmittance, caused by the lossy interface arising from lower doping. We also show that the same conclusions are valid for a range of wavelengths, so our main result is general for various dopings and wavelengths.

These results are of importance for designs of graphene sheets and flatland circuits, since we show that geometric deviations from straight edges induce drastic changes on the reflection properties. The conclusions thus apply to the design of nano-optical circuitry, sensors and antennas that employ graphene plasmons.

7

Coupled graphene disks

7.1 Introduction

In this chapter, we introduce another excitation type of plasmonics: the localized surface plasmon resonance (LSPR). These LSPRs are non-propagating, which is different from the other chapters. The effects of this phenomenon has been known for a long time, especially by artists. They used them to generate brilliant colors in glass artifacts and artwork, including gold nanoparticles of different size into the glass to create a multitude of colors. For example, the Lycurgus cup from Roman empire has a green color with reflected light and shines red with transmitted light.

The theoretical description of LSPRs on spherical nanoparticles was first described by Gustav Mie in 1908. Since then, with the improvement of nano-fabrication techniques, or modern characterization techniques, applications of these LSPRs have been flourishing. At present, metallic nanoparticles with various shapes from spheres to triangles and nanorods are widely investigated [37, 38, 39, 40]. They demonstrate a strong interaction with incident light, with for example hot spots on tips, enabling them for a wide range of applications such as biosensing [42], nonlinear optics [43], nanocircuits [3], and optoelectronics [44]. Even more recently, their scattering properties were mastered to design a transparent screen display [121] and an invisibility cloak [45]. Strong field enhancements also appear on the edge of two adjacent nanoparticles [41], so that two dipolar modes hybridize and localize the charge density on the junction edges [122]. This hybridization can reveal higher order modes that are not excitable at normal incidence for a single particle [46] (higher order resonance can appear for a single particle under non-normal incidence [39]). These so-called dark modes become bright thanks to the near-field excitation of the neighbouring resonances, as experimentally observed [123, 124]. Recently, studies of clusters consisting of even more particles (dimer, trimers, pentamers etc.) have flourished, in search of hot spots or tunable asymmetric resonances known as Fano resonances [99].

Another interesting aspect of nanoparticle dimers concerns the regime where the two nanoparticles are connected by a conductive junction, allowing the charges to oscillate in between the two metallic particles [47]. Several experiments and simulations reveal that this connection drastically alter the optical spectra, because the charge transfer tends to neutralize the two individual plasmonic charges and it creates a new low energy resonance called a charge transfer plasmon (CTP) [47, 125]. The latter depends on the conductance of the junction, and is governed by quantum [126] or classical properties [127] depending on the width of the junction. A particular interest of this junction characterization is to achieve molecular junction conduction measurements, since standard electrical transport measurement cannot be performed in gigahertz and higher frequencies regime due to the strong capacitive coupling between electrodes.

However, in metallic clusters, the only effective way to vary the optical response of the system is to change the physical configuration. For example, one needs to vary the size [40], change the metal [47], shift the configuration [128] or create a disk-ring nanostructure [129]. With bridged dimers, the variation of the optical response is achieved through the conductance of the junction: in [127], they vary the size of the junction and in [47, 125] they choose another metal. As discussed throughout this thesis, graphene is highly tunable via Fermi level doping. This chapter demonstrates how the parameters flexibility can modulate the optical response of localized resonances.

Experimental realization of graphene nanodisks have already been achieved [64]. Generally, a graphene sheet is grown using the chemical vapour deposition method, as explained in Sec. 2.3. A monolayer of polystyrene spheres is prepared on the graphene surface. Then, oxygen plasma is applied to transform the closely packed nanosphere monolayer into arrays of separate nanospheres. Areas of graphene that are not shadowed by the nanospheres are etched away. Finally, the spheres are removed by sonication in ethanol. Local doping can be reached with N-doping of graphene: local N⁺ ion irradiation creates vacancy defects in graphene that are filled by N atoms after annealing in NH₃ [67]. More details are provided in Sec. 2.3.3.

In this chapter, we consider 60 nm wide nanodisks of graphene and 25 nm wide graphene junctions. As already explained in Sec. 2.2.4, the finite-size quantum effects are not taken into account in our simulations. Related to nanodisks, experimental measurements of the extinction cross-section of a 50 nm diameter graphene nanodisk closely correspond with classical simulations [64]. One can observe a small red-shift and broadening of the resonances, known as edge-state damping in zig-zag edge



Figure 7.1: Illustration of the dimer considered, with the hybridization of the modes.

configurations [72], but the main conclusions of classical theory remain. Note finally that the 5 nm gap between the two nanodisks does not have a strong influence on the extinction cross-section for both zig-zag and armchair configurations. The main contribution to the induced dipole originates from induced charges distributed over regions away from the gap [130]. In this case, however, the local electric field enhancement is hardly influenced by the edge conformation, so we will not analyse the field enhancement in this chapter. Concerning the quantum effect in the ribbon graphene junction, classical simulations are valid for ribbons wider than 20 nm [24]. In [24], this statement is for doping higher than 0.4 eV, so the doping of our 25 nm graphene junction only varies between 0.4 and 1 eV.

Sec. 7.2 expands the work of single graphene disk modes. Indeed, graphene disks have already been studied theoretically and experimentally in gratings [64, 74]. However, these studies only consider the first dipolar mode of single disks. We show that there exist higher order modes that are dark (or subradiant) under normal incidence. The study of those single disk modes is of prior importance regarding asymmetric graphene dimers.

In Sec. 7.3, we consider two graphene disks under normal incident plane wave. Symmetric graphene dimers have been reported before [64, 130, 131, 132], but here we discuss the interaction of two graphene disks with different doping levels. This asymmetric dimer structure creates a rather complicated infrared response, as the plasmonic modes hybridize and new resonances appear, as illustrated in Fig. 7.1. Indeed, mode coupling converts dark higher order modes into visible, relatively narrow resonances. On the other hand, the coupling of two dipolar, bright resonances can create a subradiant coupled mode, which thus disappears as the doping is tuned. The section is subdivided in two parts, considering parallel and perpendicular polarizations respectively. The slightly different response under the two polarizations is analysed, resting upon the single disk modes described before and a classical model of two coupled oscillators.

The last section discusses graphene dimers connected by a graphene ribbon. The tunable, conductive junction allows charges to oscillate in between the two disks, creating a new narrower and enhanced resonance, the charge transfer plasmon. The dipolar-dipolar resonance is blue-shifted and weaker because it is screened by the charge transfer (screen bonding dipolar plasmon).

In this chapter, we perform full-wave three-dimensional simulations, where the



Figure 7.2: Scheme of the graphene nanodisk structure, with tunable doping. The incident plane wave is z directed.

graphene disks are modeled with the optical conductivity in the x - y plane. A sphere includes the disks with a background field defined as $\mathbf{E} = (E_0 e^{jk_0 z}, 0, 0)$ for an x-polarized incident plane wave where $E_0 = 1$ V/m and k_0 is the wavevector in the air. A *Perfectly Matched Layer* is designed all around the sphere to absorb the outgoing wave energy.

Most of the work in this chapter was presented in [36].

7.2 Single disk

We investigate the absorption efficiency of a free-standing single graphene circular disk of diameter D = 60 nm under normal irradiation (z directed; see Fig. 7.2). Usually, the extinction (sum of absorption and scattering) of the incident electromagnetic field is considered, however, in our case scattering is negligible (by three orders of magnitude), since the nanodisk is small in comparison to the wavelength ($D << \lambda_0$) [74]. Therefore, we only consider the absorption efficiency (Q_{abs}) where the absorption (A) is normalized with the irradiance (I) and the surface area ($G = \pi (D/2)^2$) as demonstrated in Sec. 3.3.2

$$Q_{\rm abs} = \frac{A}{IG} = \frac{8Ac\mu_0}{|E_0|^2 \pi D^2}$$
(7.1)

where we used the energy of the plane wave (Eq. 3.17) for the irradiance, with c the speed of light in vacuum, μ_0 the vacuum permeability and E_0 the incident electric field [95].

The absorption efficiency of a single disk is plotted in Fig. 7.3 (logarithmic scale), as a function of doping and the incident wavelength. As demonstrated in previous studies [64, 74, 131], the dipolar mode (solid red band) dominates and reaches an absorption efficiency of 25, which is 10 times higher than for conventional metal nanodisks [39]. Another mode appears weakly for smaller wavelength and doping, with an absorption efficiency of $Q_{abs} \approx 10^{-2}$ (light blue-green band below 0.5 eV). Note that the curve slopes downward for the two modes: indeed, analogous to graphene plasmons described in Sec. 3.2.2, a smaller doping implies a smaller imaginary part of the optical conductivity of graphene i.e. a higher effective index or a higher confinement of the field [31, 60]. Finally, a fairly constant absorption is observed for doping levels beneath the continuous white line. The latter represents the limit $\hbar\omega = 2E_F$: for smaller E_F interband transitions occur and graphene becomes dielectric, with the well known value of 2.3% absorption of a graphene sheet [17] as already described in Sec. 2.2.



Figure 7.3: Logarithm of the absorption efficiency (Q_{abs}) for a single nanodisk as a function of disk doping E_F and incident wavelength. The dipolar mode dominates. A fairly constant absorption appears when graphene is dielectric (below the solid white line). The theoretical dashed black lines stand for the first six disk modes.

This optical response can be modeled using the Coulomb potential produced by the induced charge density [64, 74]. Searching for modes leads to an eigenvalue problem corresponding with a real, unitless value (noted as η_j) for each mode j, which depends on the geometrical shape of the graphene flake. A more detailed mathematical demonstration is available in Appendix B. The resulting transcendental equation is

$$\eta_j = \frac{-j\sigma(\omega, E_F)}{4\pi\varepsilon_0\omega D} \tag{7.2}$$

with ε_0 the vacuum permittivity and $\sigma(\omega, E_F)$ the optical conductivity of graphene from Sec. 2.2. Once the geometry of the graphene flake is fixed, the value of η_j is fixed for all doping E_F , frequency ω and disk diameter D.

In Fig. 7.4 we show the various mode profiles with the corresponding η_j value, with j numbering the mode. The mode profiles are the total charge density computed using the time-harmonic current continuity from Sec. 3.1.2 ($\rho = -\nabla \cdot \mathbf{J}/(i\omega)$), with \mathbf{J} the current density computed in COMSOL. We mainly observe a linear charge on the edge for the dipolar mode (mode 0), with more charge appearing on the surface for higher order modes. The sixth mode (mode 5) is a higher order dipolar mode and shows a radial node. Note that the first mode value ($\eta_0 = 0.07$) matches

the theoretical value of a perfect conductor disk [96] $(\eta_0 = (3\pi^2/2)^{-1} = 0.07)$ and a previous study of graphene disk resonances [64] $(\eta_0 = 0.08)$.



Figure 7.4: Six first mode profiles (charge distribution) of the graphene nanodisks with their corresponding η_j eigenvalue.

This fundamental parameter η_j is determined after computing the resonant frequency once for a fixed (but fairly arbitrary) $E_F = 0.4$ eV with the COMSOL eigenfrequency solver. With the η_j value we easily determine the resonant frequency for the particular mode for all doping levels, solving the transcendental Eq. 7.2. In this way the mode dispersions of the six first modes are plotted in black dashed lines on Fig. 7.3, in good agreement with the full simulations. One observes that only the dipolar modes (modes 0 and 5) couple with the incident electric field; all the other modes are dark due to the incompatibility of their symmetry with the excitation. In order to excite these dark modes, we need to employ asymmetric dimers, as in the following section.

7.3 Graphene dimers

We demonstrate that a free-standing graphene dimer gives rise to a more complicated pattern of absorption when the doping is tuned judiciously. Indeed, the modes of the two disks hybridize and give rise to various anti-crossing effects. We examine a 60 nm diameter disk with 0.4 eV doping, placed at 5 nm distance from a second disk with doping varying from 0 eV to 1 eV (see Fig. 7.5). We investigate two directions of polarization of the incident electric field: parallel (x directed, Sec. 7.3.1) and perpendicular (y directed, Sec. 7.3.2). The incidence direction remains perpendicular to the disk planes (along z axis).

7.3.1 Parallel polarization

The absorption efficiency as a function of wavelength and doping is plotted on Fig. 7.6 for x directed electric field excitation. The first observation is the increase of



Figure 7.5: Graphene dimer with one disk of 0.4 eV doping and the other of varying doping. Normal incidence along z direction.

the complexity of the absorption pattern in comparison with the single disk absorption efficiency (Fig. 7.3). The continuous black lines represent the first six modes of a single disk (with varying E_F values) as explained in Sec. 7.2, and the vertical dashed lines are for a fixed 0.4 eV doped disk. We observe a superposition and interaction of these modes. The fundamental dipolar modes (mode 0 in Fig. 7.4) of the two disks dominate the spectra, but complexity appears where modes cross, e.g. around points 1 and 2 and around points 3 and 4, which gives rise to anti-crossings of a different nature, as we will discuss.



Figure 7.6: Logarithm of the absorption efficiency of a graphene dimer with a fixed 0.4 eV disk and a varyingly doped disk (from 0 to 1 eV) as a function of the wavelength for x polarization. Solid black lines represent the first six modes with varying doping, the vertical dashed lines for 0.4 eV. The horizontal white dashed line indicates equally doped disks. Blue lines are from perturbation theory.

The particular shape of the anti-crossing bands can be described with a classical model of two coupled oscillators [26]. Following the energy level analogy, we convert

the resonant wavelength λ_r into energy level E, which is simply $E = 2\pi c\hbar/\lambda_r$. With E_i (E_v) the energy level of a single disk mode with invariant $E_F = 0.4$ eV (varying E_F), corresponding to the black dashed (solid black) lines in Fig. 7.6, the perturbed Hamiltonian of the model is

$$\widehat{H} = \widehat{H}_0 + \widehat{W} = \begin{pmatrix} E_i + W_{11} & W_{12} \\ W_{21} & E_v + W_{22} \end{pmatrix}$$
(7.3)

with \widehat{W} the perturbation induced by the disks coupling. For simplicity, we suppose a reciprocal coupling $(W_{12} = W_{21} = \Delta)$ and a non-diagonal perturbation matrix $(W_{11} = W_{22} = 0)$. This last assumption is not totally correct in parallel polarization, as we will explain later. However, it does not change our final result. Indeed, if W_{11} and W_{22} are non-zero, this can be induced via $\tilde{E}_i = E_i + W_{11}$ and $\tilde{E}_v = E_v + W_{22}$. All the results then remain true after replacing E_i and E_v by \tilde{E}_i and \tilde{E}_v . The eigenvalues of the coupling problem $\widehat{H}\Psi = E_{\pm}\Psi$ are [133]

$$E_{\pm} = \frac{1}{2}(E_i + E_v) \pm \frac{1}{2}\sqrt{(E_i - E_v)^2 + 4\Delta^2}$$
(7.4)

The only undetermined parameter Δ can be fitted to follow the particular anticrossing pattern. The results of this perturbation theory are plotted in blue lines for two anti-crossings on Fig. 7.6, as discussed below.

First, we describe the interaction between the two dipolar modes of similar doping $E_F = 0.4$ eV (around $\lambda_0 = 6.5 \ \mu$ m). This is the particular case of the bonding and anti-bonding dipolar dimer plasmon [127]; we observe an anti-crossing between two dipolar single-disk modes. The coupling Δ between these two modes lifts the degeneracy and we obtain two solutions (blue curves in Fig. 7.6) E_+ (on the left, smaller wavelengths) and E_- (on the right). Here, we fit $\Delta = 20$ meV, and take the single disk resonances $E_i = 0.20$ eV and E_v varying from 0.17 to 0.22 eV. The blue curves correctly follow the absorption pattern but are slightly blueshifted. Incidentally, the vertical dashed line of the fixed 0.4 eV doped graphene disk is also shifted from the absorption results in Fig. 7.6. This is due to the approximation $W_{11} = W_{22} = 0$ as discussed above and could be adjusted by adding non-zero W_{11} and W_{22} .

The resulting modes are a linear combination of the two (nearly) identical dipolar modes (mode 0 in Fig. 7.4). The two possible combinations are illustrated in Fig. 7.7 for parallel polarization (horizontal arrows in the table). The lower energy mode combination corresponds to E_{-} and to a strong resonance since the dipolar moments are parallel and add up (top-left of Fig. 7.7) [122]. On the other hand, the other combination requires higher energy (E_{+}) as the dipolar moments are opposed, but resulting in a zero total dipolar moment (bottom-right of Fig. 7.7). Due to the symmetry of the excitation, the latter mode is dark in our spectrum, which explains the lack of resonance at point 1 of Fig. 7.6, and the strong resonance at point 2. This is further supported by the plot of the corresponding field enhancement on Fig. 7.8 (1) and (2): they are both identically scaled in order to distinguish the dark mode (1) from the bright mode (2).

Secondly, we describe the anti-crossing of two qualitatively different modes: the dipolar mode of the 0.65 eV doped disk and the quadrupolar mode of the 0.4 eV



Figure 7.7: Possible linear combinations of the dipolar modes for parallel (horizontal) and perpendicular (vertical) polarizations. The arrows indicate the dipolar moment directions of the involved single disk resonances.

doped disk (point 3-4 around 5 μ m in Fig. 7.6). The blue curves for perturbation theory are plotted in this area and follow the absorption pattern. In this case $\Delta = 8$ meV, indicating a smaller coupling with higher order modes (compared to 10 meV of dipolar-dipolar interaction): the charge density on the edge near the other disk is smaller for the quadrupolar mode, as charges are more distributed all along the edge (see mode 1 in Fig. 7.4, 2 positive and 2 negative poles). For this coupling, the normally dark quadrupolar mode (no coupling in Fig. 7.3) becomes bright over a large E_F range thanks to the evanescent field of the neighbouring dipolar mode. At the crossing point, we then observe the quadrupolar resonance (point 3 on Fig 7.6) as it can be seen with the enhancement of the electric field on Fig. 7.8 (3) and the dipolar mode (point 4 on Fig 7.6) represented on Fig. 7.8 (4). Note that we still observe the redshift of the modes as explained previously.

Furthermore, the sixth mode for each disk is (weakly) visible in Fig. 7.6: vertical dashed line around $\lambda_0 = 3.3 \ \mu m$ and lowest black curve. At their crossing (around point 5) the modes are not perturbed (Δ negligible), and do not interact as suggested by the electric field enhancement (Fig. 7.8 (5)). Indeed, the evanescent field at the edges demonstrates a faster decay than the lower order modes reducing the coupling strength.

Finally, the charge density profile of point 6 in Fig. 7.6 is depicted in Fig. 7.8 (6). At this wavelength ($\lambda_0 = 6.35 \ \mu m$), the 0.1 eV doped graphene disk (at right) is dielectric, and the incident electric field only couples with the dipolar mode of the fixed 0.4 eV doped graphene disk. Note that the redshift of the mode compared to the theoretical single disk vertical dashed line does no longer appear: there is no free charges on the dielectric disk at this frequency to perturb the mode.

7.3.2 Perpendicular polarization

In this section we briefly discuss the perpendicular y directed electric field polarization (Fig. 7.5), with absorption efficiency in Fig. 7.9. Again, the first six modes of single disks are represented with a solid (varying E_F values) or dashed black line



Figure 7.8: Enhancement of the electric field (total electric field over incident electric field: $|E| / |E_0|$) for the particular points of Fig. 7.6. The dashed white disks represent the edge of the graphene disk when there is no resonance. The plot numbers indicate the points in Fig. 7.6.

 $(E_F = 0.4 \text{ eV}).$

Generally, one observes a smaller influence of the second disk than in the previous section. First, there is no redshift of the modes in this case. Second, the anticrossings are less pronounced. For example, the dipolar-dipolar anti-crossing around 0.4 eV and $\lambda_0 = 6.3 \ \mu\text{m}$ shows a coupling of $\Delta = 4.5 \text{ meV}$, twice smaller than $\Delta = 10 \text{ meV}$ for parallel polarization (Sec. 7.3.1). The quadrupolar-dipolar anticrossing (around 0.65 eV and 4.8 μm) is also characterized by a weak coupling of $\Delta = 4 \text{ meV}$. Indeed, with perpendicular polarization the charge densities are not as concentrated at the edge of the small gap between the two disks (Fig. 7.7 top-right and bottom-left), reducing the influence of the neighbouring disk, and explaining the better fit between single disk resonances and the simulated dimer absorption.

We focus now on an anti-crossing of dipolar-dipolar modes (between $\lambda_0 = 6$ to 7 μ m and around $E_F = 0.4$ eV). An important difference appears in comparison to the parallel polarization case in Fig. 7.6. In perpendicular polarization the bright resonance occurs at high energy (short wavelength), while the dipolar moments cancel at low energy. In this case, the resonant energy is smaller when the charge oscillates out of phase (see Fig. 7.7) and the dark mode therefore appears on the right (higher wavelength) side of the anti-crossing.

7.4 Tunable charge transfer dimer

An alternative way to induce a tunable response is to introduce a charge transfer component. This consists e.g. in a graphene bridge connecting the two disks, which



Figure 7.9: Logarithm of the absorption efficiency of a graphene dimer with a fixed 0.4 eV disk and a varyingly doped disk (from 0 to 1 eV) as a function of the wavelength for y polarization. Solid black lines represent the first six modes with varying doping, the vertical dashed lines for 0.4 eV. The horizontal white dashed line indicates equally doped disks. Blue lines are from perturbation theory.

allows electric charges to oscillate from one disk to the other: the charge transfer plasmon (CTP). It was demonstrated that the CTP properties for metals strongly depend on the junction conductance [127]. Graphene is therefore a very suitable CTP material due to its tunable conductivity.

Here, we examine two graphene disks (diameter D = 60 nm) separated by a distance of 15 nm and with the same doping (0.4 eV). We add a charge transfer junction of 25 nm wide, see Fig. 7.10a (classical electromagnetic simulations are accurate for ribbons wider than about 25 nm [24]).

In this system we vary the junction conductance via the graphene conductivity, from 0.4 eV to 1 eV. The absorption is plotted in Fig. 7.11 for the wavelength range 4-18 μ m. The hybridized non-bridged dipolar-dipolar resonance around $\lambda_0 = 6.5$ μ m is also shown for comparison (dashed line). Note that the maximal absorption efficiency is smaller than previously, due to the longer distance d = 15 nm between the two nanodisks (d = 5 nm in Sec. 7.3).

When the dimer is bridged, two groups of resonances are distinguished: one is redshifted, while the other group appears at smaller wavelengths. The blueshifted family of resonances is called the screened bonding dipolar plasmon (SBDP) [125]. The non-bridged dipolar-dipolar resonance, which is a capacitive coupling over the dimer gap, is screened by the charge transfer, inducing a blueshift and a weaker absorption efficiency. The charge densities are plotted in Fig. 7.10c for 0.4 eV doping throughout. The dipolar modes of the two nanodisks are still visible, as charge also appears on the edges of the junction. In the case of small doping, the resistance in



Figure 7.10: (a) Scheme of the charge transfer structure. Charge density of the 0.4 eV doped junction for (b) CTP at $\lambda_0 = 14.3 \ \mu m$ and (c) screened bonding dipolar plasmon (SBDP) at $\lambda_0 = 5.7 \ \mu m$.

the junction increases, reducing the screening and therefore the blueshift.

The redshifted resonances are the charge transfer plasmons, the particular oscillation of the electrons between the disks is presented on Fig. 7.10b: one disk is a positive pole and the other a negative one. This is completely different from Sec. 7.3 where the charges oscillate separately on each disk (Fig. 7.7). With the junction the dimer acts as a continuous particle of larger length, which consequently induces a redshift.

The magnitude of the redshift depends on the time needed for the electrons to cross the junction, which is inversely proportional to the conductance [127]. In our case the only varying factor is the conductivity as the junction size is invariant. This explains why 1 eV doped graphene (higher conductivity) resonates at smaller wavelength than lightly doped graphene junctions.

Note finally that the peak is narrower for higher doping i.e. better conductivity. This last assumption can be understood with a simple resistor model (Fig. 7.12) where the disk and the junction are replaced by serial resistors r_d and r_j [127]. The power dissipated W_j in the junction when a potential V is applied is

$$W_j = \frac{V^2 r_j}{(2r_d + r_j)^2}$$
(7.5)

The conductivity and the resistance are inversely proportional, with a constant depending on the geometry of the conductor (Pouillet's law). Therefore

$$W_j = V^2 \frac{\frac{c_j}{\sigma_j}}{\left(2\frac{c_d}{\sigma_d} + \frac{c_j}{\sigma_j}\right)^2} \tag{7.6}$$

$$=V^2 \frac{c_j \sigma_j}{\left(2\frac{c_d \sigma_j}{\sigma_d} + c_j\right)^2}$$
(7.7)

where c_j and c_d are the geometric constants of the junction and the disks, respectively. At those wavelengths ($\approx 12 \ \mu m$), we have $\hbar \omega \approx 0.1 \text{ eV} \ll E_F$, and we can



Figure 7.11: Absorption efficiency of a graphene charge transfer dimer with varying bridge doping. The non-bridged dipolar-dipolar resonance is plotted in black dashed line for comparison.

use the Drude approximation for the graphene conductivity (see Eq. 2.15). We then have $\sigma \propto E_F$ and that leads to

$$W_{j} = C \frac{E_{Fj}}{\left(2\frac{c_{d}E_{Fj}}{c_{j}E_{Fd}} + 1\right)^{2}}$$
(7.8)

where $C = \frac{-2jV^2e^2}{c_j\hbar^2\pi(\omega-j\tau^{-1})}$. In our case, the doping of the disks E_{Fd} is constant. E_{Fj} dominates in the dominator (squared) and therefore, the increase of the junction doping E_{Fj} decreases the energy dissipated in the junction. As discussed in Sec. 3.3.4, decreasing damping narrows the resonance, and we observe a sharper peak for high doping.

7.5 Summary

This chapter discusses the infrared response of graphene nanodisks under various geometries: single disks, asymmetrically doped dimers and charge transfer dimers. The description of single disks under normal incidence expands the work of mode characterization to the first six modes. The previous studies mainly consider the first dipolar mode of single disks. We show that there exist higher order modes that are dark (or subradiant) under normal incidence. This study also validates the theoretical model description over a wide range of doping and wavelengths.

We demonstrate that asymmetric graphene dimers under normal incidence, induced by different doping levels, lead to an intricate and tunable absorption spectrum. From the theoretical solutions of the single modes, we explain the anti-crossing



Figure 7.12: Resistor model describing the influence of the junction doping on the width of the CTP resonances. The disks (junction) are (is) represented by the resistance r_d (r_j).

of the absorption bands, with the help of a perturbation Hamiltonian. We show that the degeneracy of the dipolar modes prompted from the two equally doped disks leads to a bright and a dark mode. That phenomenon is explained by adding the dipolar moments and depends strongly on the incident polarization. Finally, the evanescent field of the dipolar mode changes higher order dark modes into bright ones.

Finally, we demonstrate strong and tunable absorption efficiencies with charge transfer plasmon resonances, when the graphene disks are coupled via a bridge of varying doping. These numerical considerations of junctions show their interest for the ease of tunability: with conventional metals the optical properties are only adjusted when the metal of the junction is exchanged for another metal. Further investigations can consider e.g. particular non-uniform graphene doping profiles of the junction. Our findings could lead to molecular junction conduction measurements, since standard electrical transport measurements cannot be performed in gi-gahertz and higher frequency regimes due to the strong capacitive coupling between electrodes [127]. Moreover bio-sensors could be designed by providing a molecule functionalization of the graphene junction.

Further work can also examine more complicated cluster structures, such as pentamers, where tunable responses should also appear. Other studies suggest the potential for Fano resonances [99, 128] when the symmetry is broken by the size or the position of the center particle. Graphene nanodisk pentamers could deliver a similar response by adjusting the doping level of the central disk.

8

Conclusion and outlook

Throughout this work we studied and exploited the low-damped, highly confined and tunable graphene plasmons in various fundamental structures and configurations. Low-loss and strong confinement are crucial for the design of compact infrared components and the tunability allows for a myriad of functionalities, strengthening graphene as a promising material for optoelectronic applications. In the following, we propose a concise summary of this work and discuss further opportunities.

First, we considered the strong resonance of a graphene nanoribbon, and its influence on the well-known total reflectance of a propagative plasmon encountering a graphene edge. Incidentally, a ribbon separated by 10 nm from a graphene edge creates a plasmonic cavity that can achieve total absorption. Its quality factor can reach 42 for 1 eV doped graphene, which is slightly larger than the quality factor of regular plasmonic cavities. The resonance frequency is tailored easily with the graphene doping and the size of the cavity, with a simple conservative law allowing for a direct prediction of the resonant frequency. Furthermore, we inquired the influence of the distance of the ribbon to the edge and the scattering lifetime of electrons on the critical coupling, which is convenient for applications since it strongly depends on the quality of the manufactured graphene. Finally, we investigated a ribbon placed at the top of a semi-infinite graphene sheet. In that case the resonance depends on the position of the cavity and its doping. We extended the directional coupler theory to agree with the simulated results and explained qualitatively the complex resonance dependence with symmetric and asymmetric supermodes. We concluded by emphasizing the interest of these resonances for graphene bio-sensors, where the graphene ribbon parameters could be tuned with a detectable molecule.

Afterwards, we investigated the influence of doping changes on graphene plasmon propagation. We showed that for all doping configurations reflectance and transmittance follow a simple plane wave Fresnel-type model. Indeed, graphene plasmon is single mode and there are no radiation losses, fulfilling the same conditions as a plane wave encountering an interface at normal incidence. However, a small deviation is noticed for smaller doping since Fresnel coefficients need a correction for lossier media. We first considered one interface, with abrupt and smooth doping changes and we revealed reasonably that smoother and smaller doping changes induce a weaker reflectance. However, it is worth noting that smooth doping changes induce losses. Therefore, there is a trade-off between high reflectance for abrupt changes and high absorption for long transition zones. Second, we examined double interfaces that can be seen as local inhomogeneities: they occur in manufactured graphene and originate from a vacancy in the lattice filled by a grain boundary. We proposed different designs of the cavity combining abrupt and smooth doping profiles and reached total absorption for a 6 nm length cavity at the end of a semiinfinite graphene sheet. We concluded with the benefit of these results for graphene circuitry: they help to understand the behavior of plasmons encountering grain boundaries, which constitutes a strong damping source and they demonstrate the possibilities of trapping or transmitting the plasmon modes.

Patterning the edge is another way to tailor the reflection of graphene plasmons. If the straight edge totally reflects plasmons, our ribbon grating pattern offers a more complex reflectance response depending on the length and the width of the ribbons. The explanation is based on a simple Fabry-Pérot model, but originates from two contributions. First, there are *longitudinal* resonances, properly described examining the propagation constant and reflection phase of a ribbon grating. But the second contribution is not trivial: the incident plasmon couples with *lateral* edge modes, which strongly influence the reflectance. Their presence is proved by inspecting the fields and the Poynting vector. Afterwards, the influence of the doping on the reflectance was explored and it was shown that the results are general providing a careful normalization of the pattern dimensions. Summing up, geometric deviations from straight edges induce drastic changes on the reflection properties and should be considered for designs of graphene sheets and flatland circuits.

Finally, we examined the infrared response of graphene nanodisks, broadening the quickly growing field of metallic resonant particles. Unlike noble metals, graphene is tunable and allows facile adaptability of the resonance with the doping. We expanded the description of recent works on single disk modes under normal incidence to the first six modes. In this way, we revealed higher order modes that are dark (or subradiant) under normal incidence and validate the theoretical model description over a wide range of doping and wavelengths. Then, we simulated an intricate and tunable absorption spectrum of asymmetric graphene dimers under normal incidence, induced by different doping levels. The plasmonic modes hybridize and new resonances appear. On the one hand, mode coupling converts dark higher order modes into visible and relatively narrow resonances. On the other
hand, two equally doped disks create a degeneracy of the dipolar modes, leading to a bright and a dark mode. Finally, we considered bridged dimers linked by a tunable graphene ribbon junction and we observed a stronger and narrower resonance than non-bridged dimers. The advantage of graphene compared to noble metals resides in the ease of tunability: simply varying the doping of the junction shifts the resonance, where conventional metal would have required a different junction size or constitutive metal.

Further work can also examine more complicated cluster structures, such as pentamers, where tunable responses should also appear. Other studies suggest the potential for Fano resonances when the symmetry is broken by the size or the position of the center particle. Graphene nanodisk pentamers could deliver a similar response by adjusting the doping level of the central disk.

However, before going further into pure graphene device developments, another issue needs to be addressed: the losses. Indeed, even if graphene plasmons are less damped than conventional surface plasmons, the damping is stronger than in their photonic counterpart. Fortunately, a novel idea to answer this problem is emerging. Recently, with the rise of graphene and the development of mid-infrared sources, the field of polaritonic materials was revitalised. These materials support surface phonon polaritons, a result of light interacting with optical phonons, creating a surface excitation mediated by the atomic vibrations. They offer an opportunity to simultaneously achieve sub-diffraction confinement, low optical losses (in comparison to their plasmonic counterparts) and operation in the mid-infrared range. By placing a polaritonic substrate next to graphene, plasmons and phonons can couple, leading to a new mixed state recently defined as a plasmon-phonon-polariton, with a two times longer propagation length than surface phonon polaritons. Therefore, exploring the coupling between plasmons and phonons will enable light detection, modulation functionality and plasmon-phonon circuitry, combining the tunability of graphene plasmons with the low-losses of surface phonon polaritons.

Furthermore, after the rise of graphene, new two-dimensional materials have been burgeoning. They all arise from a bulk material composed of a stack of weakly attached sheets by van der Waals forces (e.g. graphite for graphene). First principle calculations of 2D hexagonal boron nitride optical parameters have been computed and experimentally measured. An entire class of 2D semiconductors can be implemented via transition metal dichalcogenides, making up for the lack of a bandgap in pristine graphene. Together, they pave the way for the realization of all 2D gated optoelectronic devices.

To conclude, graphene may not be the complete solution to design cheap, fast and compact photonic devices because the damping is still stronger than in silicon photonics. However, throughout this thesis, we demonstrated its amazing tunability properties, designing fundamental devices and describing doping influence on graphene plasmon propagation. That facile tunability makes graphene essential in the development of future heterostructures, with a wise combination of the properties of polaritonic or other two-dimensional materials. Appendices

A

Graphene plasmon dispersion

In this appendix we demonstrate that the graphene dispersion obtained with the permittivity and the conductivity of graphene are equivalent. We start from the result of [88] considering the dispersion of a classical insulator-metal-insulator (IMI) structure represented in Fig. A.1. The metal is denoted 1 with a permittivity of ε_1 and the dielectrics are denoted 2 and 3, with respective permittivity ε_2 and ε_3 .



Figure A.1: Geometry of a planar layer of thickness *a* surrounded by two half-spaces. The absolute value of the transversal magnetic field component is drawn in grey solid lines (in the case of $\varepsilon_1 = \varepsilon_3$). Propagation direction is along *x* axis. Figure from [88].

The dispersion of such an IMI is given by

$$\frac{\left(\varepsilon_{2}k_{1}+\varepsilon_{1}k_{2}\right)\left(\varepsilon_{3}k_{1}+\varepsilon_{1}k_{3}\right)}{\left(\varepsilon_{2}k_{1}-\varepsilon_{1}k_{2}\right)\left(\varepsilon_{3}k_{1}-\varepsilon_{1}k_{3}\right)} = e^{-2k_{1}a}$$
(A.1)

$$= 1 - 2k_1a + \vartheta(a^2) \tag{A.2}$$

with

$$k_i = \sqrt{\beta^2 - \varepsilon_i k_0^2} \tag{A.3}$$

with *i* depicting the number of the medium and developing the Taylor series of the exponential around 0 $(a \rightarrow 0)$ in Eq. A.2.

Distributing and simplifying leads to

$$2\varepsilon_1\varepsilon_2k_3 + 2\varepsilon_1\varepsilon_3k_2 = -2a\left(\varepsilon_2\varepsilon_3k_1^2 - \varepsilon_1\varepsilon_2k_1k_3 - \varepsilon_1\varepsilon_3k_2k_1 + \varepsilon_1^2k_2k_3\right)$$
(A.4)

Multiplying each side of the equality by a, and expressing the permittivity of the metal in term of conductivity σ (Eq. 3.9) gives

$$2a\left(1-\frac{j\sigma}{\omega\epsilon_{0}a}\right)\varepsilon_{2}k_{3}+2a\left(1-\frac{j\sigma}{\omega\epsilon_{0}a}\right)\varepsilon_{3}k_{2} = -2a^{2}\left[\varepsilon_{2}\varepsilon_{3}k_{1}^{2}\left(1-\frac{j\sigma}{\omega\epsilon_{0}a}\right)\varepsilon_{2}k_{1}k_{3} - \left(1-\frac{j\sigma}{\omega\epsilon_{0}a}\right)\varepsilon_{3}k_{2}k_{1} + \left(1-\frac{j\sigma}{\omega\epsilon_{0}a}\right)^{2}k_{2}k_{3}\right]$$
(A.5)

And taking the limit $a \to 0$ the equation simplifies to

$$\frac{j\sigma}{\omega\epsilon_0}\varepsilon_2k_3 + \frac{j\sigma}{\omega\epsilon_0}\varepsilon_3k_2 = \left(-\frac{j\sigma}{\omega\epsilon_0}\right)^2k_2k_3 \tag{A.6}$$

or simply

$$\frac{\varepsilon_2}{k_2} + \frac{\varepsilon_3}{k_3} = -\frac{j\sigma}{\omega\epsilon_0} \tag{A.7}$$

and with Eq. A.3 we reach the same dispersion as the one demonstrated with a line of current

$$\frac{\varepsilon_2}{\sqrt{\beta^2 - \varepsilon_2 k_0^2}} + \frac{\varepsilon_3}{\sqrt{\beta^2 - \varepsilon_3 k_0^2}} = -\frac{j\sigma}{\omega\epsilon_0}$$
(A.8)

showing that computing the graphene plasmons with the effective permittivity or with the conductivity is similar in the limit of thin graphene $(a \rightarrow 0)$.

B

Nanodisk modes

In this section we describe the optical response of a graphene flake in term of the electrostatic potential ϕ [74, 64]. We consider a graphene flake of size D in vacuum, homogeneously doped. It is characterized by a charge density $\rho(\mathbf{r}')$. The coulomb potential in the plane of the graphene flake is then [86] :

$$\phi(\mathbf{r}) = \frac{1}{4\pi\varepsilon_0} \int \frac{\rho(\mathbf{r}')}{|\mathbf{r} - \mathbf{r}'|} d^2 r'$$
(B.1)

where $d^2r' = dx'dy'$ is a two-dimensional plane element.

The continuity equation in the harmonic fields approximation without charge creation is $(e^{j\omega t} \text{ convention})$

$$\rho(\mathbf{r}') = \frac{j}{\omega} \nabla \cdot \vec{J}(\mathbf{r}') = \frac{-j}{\omega} \nabla \cdot [\sigma(\mathbf{r}', \omega) \nabla \phi(\mathbf{r}')]$$
(B.2)

using Ohm's law $\mathbf{J} = \sigma \mathbf{E}$ and the definition of the scalar potential $\mathbf{E} = -\nabla \phi$.

We can remove the position in $\sigma(\mathbf{r}', \omega)$ introducing a function $f(\mathbf{r}')$ that takes value 1 on graphene and 0 elsewhere. Inserting Eq. B.2 in Eq. B.1, we find

$$\phi(\mathbf{r}) = \frac{-j\sigma(\omega)}{4\pi\varepsilon_0\omega} \int \frac{\nabla \cdot [f(\mathbf{r}')\nabla\phi(\mathbf{r}')]}{|\mathbf{r} - \mathbf{r}'|} d^2r'$$
(B.3)

Using dimensionless coordinates $\pmb{\theta} = (x/D, y/D)$ we can write the eigenvalue problem

$$\frac{1}{\eta}\phi(\boldsymbol{\theta}) = \int \frac{\nabla \cdot [f(\boldsymbol{\theta'})\nabla\phi(\boldsymbol{\theta'})]}{|\boldsymbol{\theta} - \boldsymbol{\theta'}|} d^2\theta'$$
(B.4)

with a graphene flake geometrical dependant operator

$$\mathcal{W}[\phi(\boldsymbol{\theta})] = \int \frac{\nabla \cdot [f(\boldsymbol{\theta}') \nabla \phi(\boldsymbol{\theta}')]}{|\boldsymbol{\theta} - \boldsymbol{\theta}'|} d^2 \theta', \qquad (B.5)$$

the eigenvectors ϕ and the eigenvalues or modes $1/\eta$ of the problem:

$$\eta = \frac{-j\sigma(\omega)}{4\pi\varepsilon_0\omega D} \tag{B.6}$$

Note that the eigenvalues are established for any geometrical form of the graphene flake. This means that once η is determined for one mode of a fixed geometry, then it will not vary as a function of the frequency ω , the conductivity of the graphene sheet $\sigma(\omega)$, or the size D of the graphene flake.

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