

Mathematical modeling and computer simulation of interactions of charged particles with 2D materials

by

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Author's Declaration

I hereby declare that I am the sole author of this thesis. This is a true copy of the thesis, including any required final revisions, as accepted by my examiners.

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Abstract

Recent interest in the nanophotonic and nanoplasmonic devices has led researchers to a detailed study of different aspects of the interactions of the moving charged particles with two dimensional (2D) materials. When a charged particle moves above a 2D material, electromagnetic forces due to the polarization of the charges in the target material cause energy dissipation. Analyzing the energy spectra and momentum change of the reflected particles provides valuable information about the internal structure of the target material. The analyses of this kind are extensively performed in the electron energy loss spectroscopy (EELS) and the high resolution EELS (HREELS) experiments. To model such interactions, we used the classical dielectric response theory in the non-retarded approximation. We obtained closed form expressions for the forces acting on the charged particles moving parallel to doped phosphorene using analytical models for its dielectric function, which expose the strongly anisotropic character of the electronic structure and dynamic response of this 2D material. The parameters of these models, which we call the optical and the semiclassical models, are supplied by the *ab initio* calculations. It was found that the force on the incident charge has three components, showing strong dependence on the direction of motion of the charged particle. Furthermore, we performed an analysis of the electric potential in the plane of phosphorene, revealing a rich variety of the plasmonic wake patterns induced by the moving charged particle. Our computations showed surprising analogies of these wakes with Kelvin's ship wakes and atmospheric wakes regarding the asymmetry of the wake, slowing down of the plasmon dispersion, and the formation of Mach-like wake due to nonlocal effects in the semiclassical dielectric response function. In a related effort, we adopted the energy loss method (ELM) to compute the phosphorene carrier mobility tensor when the carrier scattering on charged impurities in the substrate is the main limiting factor of the mobility in the DC regime at low temperatures. The ELM provides a closed form expression for the phosphorene carrier's mobility tensor in terms of a double integral, which is superior to the traditional approach for mobility calculation based on the Boltzmann transport equation that requires numerical solution of an integral equation. Using the ELM, we examined the mobilities for different statistical distributions of the charged particles, revealing strong effects of the inter-particle correlation distance on the mobilities. Finally, we evaluated the energy loss function of doped graphene when there exist a random distribution of charged impurities in the substrate. We modeled the resulting random potential landscape in graphene via local Fermi energy, with its in-plane spatial correlation governed by a Gaussian distribution, giving rise to an expression for the loss function that includes a memory function, which is the solution of a nonlinear integral equation. The overall effect of this random potential landscape was found to be a broadening and shifting of the plasmon peak in the energy loss function of doped graphene.

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Dedication

To Maryam, Mohammad, and in loving memory of my father.

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Chapter 1

Introduction

1.1 Literature review and motivation

From a fully theoretical point of view, many years before the discovery of the first physical representative of a two dimensional electron gas (2DEG), or more generally, two dimensional (2D) crystals, they were of great importance to theoretical physicists due to their intrinsic extraordinary electrical, mechanical, and thermal behavior in comparison with three dimensional (3D) electron gas or crystals [4, 5]. One of the early representatives of a 2DEG in which the integer quantum Hall effect was observed as a consequence of two dimensional confinement, was the inversion layer of a Si-SiO₂ based system [6].

In 2004, Novoselov *et al.* discovered graphene, which is a single layer of graphite with a hexagonal lattice structure, making it the first isolated 2D material [7–10]. The first theoretical prediction of the electronic energy band structure of graphene was published much earlier by a Canadian physicist in Ref. [11]. Since this important discovery, graphene has become an important 2D material that has found many applications in different fields, such as photonics [12], nanoelectronics [13], and plasmonics [14, 15]. The discovery of graphene accelerated the development of fabrication techniques such as exfoliation, allowing researchers to isolate new 2D materials, such as phosphorene, with an orthorhombic crystal structure that is the single layer of black phosphorus. In 2014, the electrical properties of monolayer and few-layer black phosphorus were studied [16–19]. Based on these initial studies, this material possesses a direct bandgap from ~ 0.3 eV for bulk to ~ 2.0 eV for monolayer and highly mobile carriers with mobilities in the range 10,000 to 26,000 (in cm²V⁻¹s⁻¹). Unlike the transition metal dichalcogenide (TMDC) [20, 21] family, the direct topology of the bands in phosphorene remains unchanged as the number of layers

increases or decreases to a monolayer, making it a potential material for photonics and photoelectronic applications. Due to the intrinsic anisotropy combined with its puckered structure, phosphorene has highly anisotropic electrical, mechanical, and optical properties along its principal crystalline axes, labeled as the armchair (AC) and the zigzag (ZZ) directions [22–25]. Moreover, the bandgap value (0.3-2.0 eV) of phosphorene is typically smaller than the bandgap of TMDC compounds (1.1-2.5 eV), and larger than graphene, which is in fact a semimetal; this makes phosphorene a promising material for semiconductor devices, which comprises a suitable on/off ratio ($10^4 - 10^5$) [16, 18] with significantly large carrier mobility.

The intrinsic anisotropy of the in-plane conductivity tensor of the phosphorene makes it a promising candidate to be a natural representative of hyperbolic metasurfaces [26, 27], which have the unique property to give rise to the so called hyperbolic plasmon polariton modes identified by hyperbolic isofrequency dispersion curves. Hyperbolic plasmon modes are strongly direction dependent in terms of in-plane propagation, which allows for modes with relatively large wave numbers. Therefore, hyperbolic metasurfaces can serve in many applications, such as wave guiding, hyperlensing, and negative refraction [28]. Anisotropic 2D materials such as phosphorene, which can host hyperbolic plasmon modes, are also important from a technological point of view because of their ease of fabrication and the tunability of their optical response [29, 30].

The tunability of the optical response of black phosphorus (BP) has been deeply studied theoretically as layer thickness, mechanical strain, doping density, or the nearby dielectrics vary [31–34]. Recent experiments demonstrated the dynamic tunability of hyperbolic plasmon polariton modes in BP in the midinfrared (MIR) frequency range. In these experiments, the intraband electronic transitions were controlled via changing the doping density in phosphorene layers by applying an external gate potential [35, 36]. Additionally, theoretical studies revealed that in the terahertz (THz) and the MIR frequency ranges, phosphorene exhibits a topological transition between the elliptic and hyperbolic isofrequency plasmon polariton dispersion curves as the density of quasifree charge carriers varies [37–40]. Moreover, a theoretical model was developed to take the optical anisotropy of doped phosphorene into account by means of an in-plane conductivity tensor [32, 39]. This model was used to demonstrate a relation between the tunability and the directionality effects in launching the elliptic and hyperbolic plasmons, induced by a nearby stationary radiating dipole with adjustable polarization [38, 41–43]. Although analytical models are extensively used for the optical conductivity of doped phosphorene in the technologically important THz-MIR frequency range [32, 38, 41–44], there are several *ab initio* calculations which are based on the density functional theory (DFT), providing conductivity tensor of phosphorene over a much broader range of frequencies, from the THz to the vacuum

ultraviolet [45–47].

A moving charged particle, usually an electron, induces secondary electron emission (SEE), cathodoluminescence (CL) light emission, and many other excitations, such as plasmons and phonons, upon impinging on a microscope specimen. Analyzing the energy of this electron after reflection or passing through the sample provides information on the excitation spectrum of the sample. A practical setup to measure this energy loss is electron energy-loss spectroscopy (EELS) performed in scanning transmission electron microscope (STEM). The main advantage of electron microscopy in comparison with light based experiments is that EELS can provide practical nanometer spatial resolution [48], allowing the investigation of localized excitations that involve finite wave-vector components of the incident electromagnetic beam. As a result, electron beams were utilized in discovering and characterizing collective excitations of conduction electrons in metals, commonly known as plasmons because they are quasiparticles associated with quantized oscillations of the electron gas plasma [49–53]. Plasmons can be confined at the surface of metals or in the 2D materials to constitute the so called surface plasmons (SPs) [54–56] or sheet plasmons [57]. The first theoretical analysis of the interaction of a moving charged particle with 2DEG appeared in the paper by Fetter [58], who used the hydrodynamic model to describe SPs in the 2DEG.

In a more advanced setup, which is called the momentum-resolved EELS [59] the momentum change in the reflected or transmitted electron beam is measured as well as the energy change. This experiment in fact probes the *loss function* of a material, which is defined as the negative of the imaginary part of the inverse of the material dielectric function: $L(\mathbf{q}, \omega) = -\Im\{1/\varepsilon(\mathbf{q}, \omega)\}$. This spectroscopy has successfully been used to characterize the loss function of materials such as graphene [60–62], TMDCs [63, 64], and BP [65]. Moreover, if the low energy surface excitations are to be studied, several techniques, such as the vibrational and valence aloof beam EELS in a monochromated STEM [66–69], a high-resolution EELS (HREELS) that analyzes the low energy electrons reflected from the surface [70–75], and grazing scattering of medium-energy ions from surfaces [76–81] have proven to generate the most sensitive experiments. For instance, the momentum resolved HREELS experiments were utilized to reveal the tunability of the surface plasmons in doped graphene and their hybridization with optical phonon modes in an insulating polar substrate, happening in the THz-MIR range of frequency [57, 75, 82–84]. It was shown that graphene is most sensitive to the incident particle trajectories that are almost parallel to it (near-grazing scattering angles) [85]; in fact, this type of scattering is the most surface-sensitive regime in HREELS and ion-surface scattering experiments [71, 80]. Therefore, it is appropriate to ask whether a similar type of experiment could effectively probe the effect of directionality in exciting the elliptic and hyperbolic plasmon modes in doped phosphorene.

In order to provide a mathematical model for EELS experiments using a charged particle moving on an aloof trajectory or under grazing incidence upon the target surface, the particle could be assumed to move with constant velocity in a trajectory that is parallel to the surface of the 2D target material; in fact, its velocity component that is perpendicular to the target surface may be treated as adiabatically slow on a time scale of the target response [79, 86, 87]. Such a model proposes the existence of two different forces acting on the charged particle. The first force is a dissipative force, which is parallel to the particle's velocity vector in the case of isotropic target materials and acts to slow down the particle; hence, it is commonly called the stopping force. The second force is a conservative force perpendicular to the particle's trajectory and the target surface; this force is necessarily attractive, trying to bend the particle's trajectory toward the target; hence, commonly called the dynamic image force [71, 77, 79, 86]. If the target material is anisotropic, the stopping force is not necessarily parallel to the particle velocity vector and can have a component that is perpendicular to the particle velocity vector but parallel to the target surface. This component, which is a result of anisotropy in the dielectric response of the target [88, 89] will be called the *transverse* stopping force, while the component of the stopping force that is still parallel to the particle velocity is called the *longitudinal* stopping force. In this study, first a full description of these forces acting on a charged particle that is moving parallel to a general anisotropic 2D material is provided, then the effect of the existence of a supporting material is taken into account, and finally the results are specialized for the phosphorene supported by SiO₂ via appropriate models and relevant parameters. In most of the realizations of the EELS experiment, especially HREELS, the speed of the moving electrons is significantly less than the speed of light, and relativistic effects can be ignored. This assumption, which is commonly known as the non-retarded approximation, works well in many situations and provides useful approximation to the energy loss spectra of fast electrons [48]. On the other hand, we assume that the energy of the incident particle is large enough, so that the lost energy constitutes a small fraction of it, giving rise to the so-called no-recoil approximation to its trajectory.

Note that by the assumptions stated above, in fact a classical description of the particle and its trajectory in the non-retarded approximation, allow us to use the dielectric-response formalism within the Poisson equation to describe the Coulomb interactions throughout the structures studied in this thesis. This classical treatment of the problem is independent of the specific model that we use for the dielectric response of the target. In this study, we limit our focus to the THz-MIR frequency range and consider three different models with increasing complexity for the conductivity tensor of an anisotropic 2D material that might be supported by a substrate. These models, which we call the optical model, the semiclassical model, and the quantum mechanical model [87, 90, 91] can be adjusted to

simulate phosphorene when appropriate parameters, which will be discussed in detail, are set.

A standard output of the Poisson problem for a point charge moving uniformly above a surface, which supports a plasmon mode, is the electrostatic potential that exhibits a wake pattern akin to the Kelvin wake due to a ship moving on a deep calm water [92]. We computed the wake potential using the three dielectric models for phosphorene and found that due to the anisotropy in the phosphorene response functions, the geometry and topology of these plasmon wakes strongly vary in different directions, and the wake angle is a function of the movement direction. Our investigation of the wake potential using the method of stationary phase [92–94] revealed that a charged particle moving parallel to the anisotropic phosphorene can induce a rich variety of the plasmonic wake patterns [95], similar to those happening in other physical systems involving a fluid and a disturbing agent that are moving relatively, such as Kelvin’s ship wakes and atmospheric wakes [94, 96–99].

In practice, free standing 2D materials are rarely stable, and they are almost always mounted on a supporting material. On the other hand, no perfect supporting material is available, and there is always a distribution of charged impurities in the supporting material. The DC conductivity of a perfect phosphorene sample is mainly limited by some scattering processes, such as interactions between charge carriers and phonons, which are the quantized lattice vibrations in the phosphorene, and interactions between charge carriers and charged impurities (CIs) which are distributed inside the substrate material. At low temperatures, electron-phonon scattering does not play an important role, and the DC conductivity and mobility tensors are mainly limited by the Coulomb scattering with CIs. A common approach to compute the mobility tensor of carriers in this regime is solving the linearized Boltzmann transport equation (BTE) which ends up with an integral equation for the carriers momentum relaxation time (MRT) that must be solved numerically. Major limitations of the BTE formalism in this problem are the dependency of the relaxation times on the direction of the applied electric field [2, 91, 100–102], and the singular nature of the resulting integral equation that makes it unstable to be solved using ordinary numerical methods. Therefore, BTE is seriously limited when exploring various designs of nanoelectronic devices based on phosphorene [103–105], and is extremely expensive from a computational viewpoint when considering the effects of a spatial arrangement of CIs on the mobility of phosphorene, as a Monte Carlo simulation is required in that case.

The energy loss method (ELM) [106–108] is an alternative approach to compute the mobility tensor of phosphorene in DC regime. The main idea behind the ELM is that the energy dissipation rate could be computed from two different view points. Firstly, we can choose the frame of reference of the substrate in which charged impurities are fixed and carriers in phosphorene move at constant drift velocity. Considering the effective external

electric field in the presence of charged impurities and the current density due to the drift velocity, it is possible to evaluate the power dissipation of the system. Secondly, we can change the frame of reference to that in which phosphorene charge carriers are fixed but the charged impurities are rigidly moving with the reversed drift velocity. With the induced electric field due to the response of the phosphorene carriers to the moving impurities and the associated current density of those impurities, the energy loss rate of the system of impurities could be computed. Finally, these two power dissipations must be equivalent, which is the core assumption of the ELM [109–111]. The ELM has several advantages in comparison with BTE method. The ELM provides a closed form expression for the mobility tensor that is given as a definite double integral involving the dielectric function for the intraband electronic transitions in a 2D conducting material, which only needs to be available to the leading order in frequency [106, 107], and the geometric structure factor, which takes the effect of the distribution of the charged impurities into account. Moreover, using dielectric functions with different precision levels can reveal the role of different physical effects in the DC mobility, enabling the inclusion of various material properties into the mobility calculations, such as the band structure anisotropy [2, 91, 101, 102], finite temperature [100], or contribution of interband transitions to the static screening of the impurity potential [1, 112–115]. On the other hand, the geometric structure factor enables us to perform a statistical analysis of the effects of spatial correlation among the impurities. For instance, using the quantum mechanical model for the dielectric function of the phosphorene and the structure factor of the hard-disk model of a planar distribution of pointlike impurities, we analyzed the effects of the correlation distance on the mobility of carriers at different doping densities of phosphorene [116], a calculation that is extremely difficult, if not impossible, using the BTE approach.

It is also of great importance to study the effect of the random distribution of the charged impurities inside the substrate, on the plasmon dispersion in a supported 2D material. In this study, the effect of randomly distributed charged impurities is taken into account via a random spatial modulation of the energy level associated with the Dirac point in doped graphene with respect to the constant Fermi level. Note that the effect of the random surface roughness was studied before [117, 118]. A common feature of these problems is that at some point in the modeling process, an eigenvalue problem for the electrical potential in graphene is obtained in terms of frequency within the Drude model. Considering the randomness in the Dirac point position, we applied the so called smoothing method due to Maradudin [119] to obtain an integral equation in terms of the unknown function $m(\mathbf{q}, \omega)$, which we call the memory function. Finally, the loss function of the system that includes the effect of the randomness in the Dirac point position was expressed by $L(\mathbf{q}, \omega) = -\Im\{(\varepsilon(\mathbf{q}, \omega) - m(\mathbf{q}, \omega))^{-1}\}$. We used a Gaussian correlation

function to describe this randomness in which the correlation distance models the spacing of the randomly distributed charges. Our results reveal that the overall effect of this randomness is a broadening in the loss function peaks that gives rise to a decay of the sheet plasmon in doped graphene, which is consistent with physical interpretations for similar problems on rough metallic surfaces [118]. It should be mentioned that the integral equation, for which $m(\mathbf{q}, \omega)$ is a solution, is a nonlinear Fredholm integral equation with no known analytic solutions; hence, numerical methods are required. We were able to solve this integral equation by a modified iteration method, which we call the double iteration scheme (DIS).

1.2 Organization of the thesis

In Chapters 2 and 3, the main physical assumptions, mathematical tools, and models used are introduced. Chapters 4 and 5 present in full detail the results of our research on the forces and wakes due to a charged particle moving parallel to phosphorene, and the ELM-based approach to calculate the phosphorene carrier mobility tensor, which are published in Refs. [87, 95, 116]. In Chapter 6, we present the most recent results of our research on the energy loss function of a system consisting of graphene supported on a SiO_2 substrate in the presence of a random distribution of charged impurities inside the substrate. This thesis is organized so that the main results are presented in the main text, whereas the proofs of some relations and parts of the discussion pertaining to chapter 4 are moved to the appendices.

Chapter 2

Theoretical Background

2.1 Physical assumptions

In this section, a detailed description of the basic physical assumptions used throughout this thesis is provided. These assumptions mainly consist of:

- Maxwell's equations, which consist of four coupled vector equations, formulated in either the integral or the differential form. Solutions of this set of equations are the fundamental electric and magnetic fields that can provide a full description of a system from an electromagnetic point of view.
- Non-retarded approximation, which is equivalent to formally setting the speed of light c to be infinity. This assumption significantly reduces the mathematical complexity of the Maxwell's equations, while the results are still in excellent agreement with experimental data for problems that are justified to be modeled in this regime. It was shown in Refs. [58] and [120] that non-retarded approximation is applicable when the speed of the moving charged particle is such that $v \ll c$, in which case the effects of the magnetic field are negligible, whereas the electric field may be considered conservative.
- Translational invariance, which is a geometric constraint on the problem that allows the use of Fourier transformation to reduce the complexity of the governing differential equations. For instance, consider a flat 2D material supported by a semi-infinite substrate with a sharp interface. If both structures extend infinitely without any edges, the geometric completeness of this system implies symmetry in all physical

parameters in the planes parallel to the interface. Therefore, a 2D Fourier transformation in these planes converts differential and integral operators into simple algebraic operations, simplifying the problem.

2.1.1 Maxwell's equations

The most fundamental physical parameters in electromagnetic theory are the electric and magnetic fields, denoted by $\mathbf{e}(\mathbf{R}, t)$ and $\mathbf{b}(\mathbf{R}, t)$, respectively; and the electric charge density and electric current density, denoted by $\rho(\mathbf{R}, t)$ and $\mathbf{J}(\mathbf{R}, t)$, respectively. Maxwell's equations describe the complicated interdependencies of these functions in the form of a set of coupled differential equations. These equations in their microscopic form, and in the Gaussian system of units, which is often used units system in research fields such as quantum mechanics and optical properties of solids, are given by [121, 122]

$$\begin{cases} \nabla \times \mathbf{e}(\mathbf{R}, t) + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{b}(\mathbf{R}, t) = 0, \\ \nabla \cdot \mathbf{b}(\mathbf{R}, t) = 0, \\ \nabla \times \mathbf{b}(\mathbf{R}, t) - \frac{1}{c} \frac{\partial}{\partial t} \mathbf{e}(\mathbf{R}, t) = \frac{4\pi}{c} \mathbf{J}_{\text{micro}}(\mathbf{R}, t), \\ \nabla \cdot \mathbf{e}(\mathbf{R}, t) = 4\pi \rho_{\text{micro}}(\mathbf{R}, t), \end{cases} \quad (2.1)$$

where $\rho_{\text{micro}}(\mathbf{R}, t)$ and $\mathbf{J}_{\text{micro}}(\mathbf{R}, t)$ are the microscopic charge and current densities, respectively, $\mathbf{R} = (x, y, z)$ is the position vector, and t is time. These microscopic sources include the contribution of the electrons and atomic nuclei that are the basic charge elements in materials. For instance, if the electrons and atomic nuclei are modeled by point charges, a possible expression for the microscopic charge density is given by

$$\rho_{\text{micro}}(\mathbf{R}, t) = \sum_i q_i \delta(\mathbf{R} - \mathbf{r}_i),$$

where q_i is the charge of the i^{th} particle and $\delta(\cdot)$ stands for the Dirac delta function. In a more precise approximation, one can use the following approximation for the electronic charge contribution of the microscopic charge

$$\rho_{\text{microelectronic}}(\mathbf{R}, t) = -e \Psi(\mathbf{R}) \Psi^*(\mathbf{R}),$$

where e is the magnitude of the fundamental electric charge of a single electron, and $\Psi(\mathbf{R})$ is the electron wave function of the material. Note that, according to the Born-Oppenheimer approximation, atomic nuclei can still be accurately approximated by point charges.

Since the number of atoms in a volume of the size $\sim 1 \text{ cm}^3$ is around 10^{23} , microscopic Maxwell's equations are not practical to use in studying optical properties of solids. On the other hand, in the frequency range of the important optical properties, the wavelength of the incident electromagnetic wave is large enough to ensure that millions of atoms are contained in a volume with characteristic dimensions of the order of a wavelength. Therefore, it is possible to smooth out the unnecessary details through an averaging operation on the Maxwell's equations. Macroscopic electromagnetic functions are defined by

$$\mathbf{E}(\mathbf{R}, r) = \frac{1}{\Delta V} \int_{\Delta V} \mathbf{e}(\mathbf{R} + \boldsymbol{\xi}) d\boldsymbol{\xi}, \quad (2.2)$$

$$\mathbf{B}(\mathbf{R}, r) = \frac{1}{\Delta V} \int_{\Delta V} \mathbf{b}(\mathbf{R} + \boldsymbol{\xi}) d\boldsymbol{\xi}, \quad (2.3)$$

$$\rho_{\text{total}}(\mathbf{R}, r) = \frac{1}{\Delta V} \int_{\Delta V} \rho_{\text{micro}}(\mathbf{R} + \boldsymbol{\xi}) d\boldsymbol{\xi}, \quad (2.4)$$

$$\mathbf{J}_{\text{total}}(\mathbf{R}, r) = \frac{1}{\Delta V} \int_{\Delta V} \mathbf{J}_{\text{micro}}(\mathbf{R} + \boldsymbol{\xi}) d\boldsymbol{\xi}, \quad (2.5)$$

where all new functions defined by the smoothing integrals are macroscopic counterparts of the corresponding microscopic functions that is appeared in the integrand, and ΔV is a convex volume around the origin, large enough to contain many millions of atoms. Macroscopic Maxwell's equations are obtained by taking the average of the microscopic equations and using the definitions of the smoothed functions stated above. Macroscopic Maxwell's equations could be written as [121]

$$\begin{cases} \nabla \times \mathbf{E}(\mathbf{R}, t) + \frac{1}{c} \frac{\partial}{\partial t} \mathbf{B}(\mathbf{R}, t) = 0, \\ \nabla \cdot \mathbf{B}(\mathbf{R}, t) = 0, \\ \nabla \times \mathbf{B}(\mathbf{R}, t) - \frac{1}{c} \frac{\partial}{\partial t} \mathbf{E}(\mathbf{R}, t) = \frac{4\pi}{c} \mathbf{J}_{\text{total}}(\mathbf{R}, t), \\ \nabla \cdot \mathbf{E}(\mathbf{R}, t) = 4\pi \rho_{\text{total}}(\mathbf{R}, t). \end{cases} \quad (2.6)$$

Macroscopic Maxwell's equations are commonly employed to investigate the optical properties of solids. In this thesis, the term "Maxwell's equations" will refer specifically to their macroscopic form, given by Eqs. (2.6).

2.1.2 Non-retarded approximation

In many practical applications, such as EELS with low energy electrons or in the HREELS experiments the speed of the moving electrons is significantly smaller than the speed of

light c . In this regime, the results that were derived in Refs. [58] and [120] (Chapter 14) in the non-retarded limit may be achieved by formally setting the speed of light to be infinity in the above equations from the outset, which gives the so-called quasi-static approximation to the Maxwell's equations

$$\begin{cases} \nabla \times \mathbf{E}(\mathbf{R}, t) = 0, \\ \nabla \cdot \mathbf{B}(\mathbf{R}, t) = 0, \\ \nabla \times \mathbf{B}(\mathbf{R}, t) = 0, \\ \nabla \cdot \mathbf{E}(\mathbf{R}, t) = 4\pi\rho_{\text{total}}(\mathbf{R}, t). \end{cases} \quad (2.7)$$

In the non-retarded regime, both the curl and the divergence of the magnetic field $\mathbf{B}(\mathbf{R}, t)$ are zero, which means the magnetic field is a constant vector that without loss of the generality could be assumed to be zero, $\mathbf{B}(\mathbf{R}, t) = \mathbf{0}$. Moreover, the curl of the electric field $\mathbf{E}(\mathbf{R}, t)$ is zero, which by virtue of the Helmholtz theorem means that

$$\mathbf{E}(\mathbf{R}, t) = -\nabla\Phi(\mathbf{R}, t), \quad (2.8)$$

for some scalar function $\Phi(\mathbf{R}, t)$ that is called the electric potential.

The total charge density $\rho_{\text{total}}(\mathbf{R}, t)$ consists of two different parts

$$\rho_{\text{total}}(\mathbf{R}, t) = \rho_{\text{pol}}(\mathbf{R}, t) + \rho_{\text{ext}}(\mathbf{R}, t), \quad (2.9)$$

where $\rho_{\text{ext}}(\mathbf{R}, t)$ is the external charge density, and $\rho_{\text{pol}}(\mathbf{R}, t)$ is the polarization charge density. $\rho_{\text{ext}}(\mathbf{R}, t)$ is independent of the existing materials and is introduced by some external source such as the moving electrons in the EELS experiments. On the other hand, $\rho_{\text{pol}}(\mathbf{R}, t)$ is the induced charge (sometimes denoted by $\rho_{\text{ind}}(\mathbf{R}, t)$) in the existing materials describing the response of the electric charges of the materials to the external charge. Note that $\rho_{\text{ext}}(\mathbf{R}, t)$ is known and is given in the problem statement, while $\rho_{\text{pol}}(\mathbf{R}, t)$ is not known *a priori*. A basic assumption on the polarization charge density is that for some polarization density vector $\mathbf{P}(\mathbf{R}, t)$ that is defined in the materials we have

$$\rho_{\text{pol}}(\mathbf{R}, t) = -\nabla \cdot \mathbf{P}(\mathbf{R}, t). \quad (2.10)$$

Moreover, from a quite mathematical viewpoint, there always exists a tensor χ_e that relates the polarization and the electric field vectors through

$$\mathbf{P}(\mathbf{R}, t) = \chi_e \mathbf{E}(\mathbf{R}, t). \quad (2.11)$$

In general, such a tensor χ_e that is called the electric susceptibility, is a function of the electric field as well as its dependence on the position and time variables. In the linear

response theory, we assume that χ_e is independent of the electric field, but still a function of position and time. This final assumption is well-supported by physical principles and remains valid provided the electric field strength does not become excessively large. While Eq. (2.11) implies a local relation between the polarization and electric fields, it should be generalized into a convolution of these fields when non-local effects are present in the time and/or space domains. Combining the last Maxwell equation from Eqs. (2.7), and Eqs. (2.10, 2.11, 2.8, and 2.9) we find

$$\nabla \cdot [\epsilon \mathbf{E}(\mathbf{R}, t)] = 4\pi \rho_{\text{ext}}(\mathbf{R}, t), \quad (2.12)$$

or

$$\nabla \cdot [\epsilon \nabla \Phi(\mathbf{R}, t)] = -4\pi \rho_{\text{ext}}(\mathbf{R}, t), \quad (2.13)$$

where $\epsilon = \mathbf{1} + 4\pi\chi_e$ is called the dielectric constant of the material, in which $\mathbf{1}$ is the unit tensor. Equation (2.13) is the most fundamental equation that we solve subject to some appropriate boundary and matching conditions that are determined by the problem. Note that χ_e is generally an integral operator, so that in Eq. (2.12) we can define the electric displacement vector $\mathbf{D}(\mathbf{R}, t) = \epsilon \mathbf{E}(\mathbf{R}, t)$ that in a more general viewpoint may be expressed as

$$\mathbf{D}(\mathbf{R}, t) = \epsilon(\mathbf{R}, t) * \mathbf{E}(\mathbf{R}, t) = \int d\mathbf{R}' \int dt' \epsilon(\mathbf{R}, \mathbf{R}', t, t') \mathbf{E}(\mathbf{R}', t'), \quad (2.14)$$

where $*$ stands for the convolution operator, which defines the dielectric function $\epsilon(\mathbf{R}, \mathbf{R}', t, t')$ to be the response function connecting the electric field and the electric displacement. Note that in this general relation, the effect of $\mathbf{E}(\mathbf{R}, t)$ on $\mathbf{D}(\mathbf{R}, t)$ at all other positions and all earlier times is taken into account.

2.1.3 Translational invariance and Fourier transformation

Throughout this thesis, the underlying system consists of a semi-infinite supporting material with flat surface, a 2DEG that is located parallel to the surface of the supporting material at some distance h , and a single or a system of point charges. Moreover, we assume that both the 2DEG and the substrate are homogeneous. A perspective of this standard system is depicted in Fig. 2.1.

If we choose a Cartesian coordinate system in such a way that x-y plane is parallel to the plane of the 2DEG, then there is no preference to choose the origin. In other

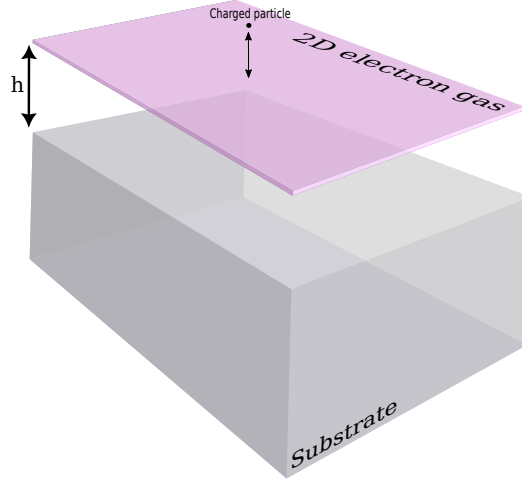


Figure 2.1: A perspective of the standard system.

words, all points with the same z coordinate are equally acceptable to be the origin. This full symmetry in the x - y plane is called the translational invariance, allowing to use the 2D Fourier transformation in the x and y directions and reducing the complexity of the governing differential equations. Throughout this study, the following convention for the Fourier transformation is used to transform $\mathbf{r} = (x, y) \rightarrow \mathbf{q} = (q_x, q_y)$, and $t \rightarrow \omega$

$$\mathcal{F}\{A(\mathbf{r}, z, t)\} = \tilde{A}(\mathbf{q}, z, \omega) = \iint d\mathbf{r} \int dt A(\mathbf{r}, z, t) e^{-i(\mathbf{q} \cdot \mathbf{r} - \omega t)}, \quad (2.15)$$

and

$$\mathcal{F}^{-1}\{\tilde{A}(\mathbf{q}, z, \omega)\} = A(\mathbf{r}, z, t) = \frac{1}{(2\pi)^3} \iint d\mathbf{q} \int d\omega \tilde{A}(\mathbf{q}, z, \omega) e^{i(\mathbf{q} \cdot \mathbf{r} - \omega t)}, \quad (2.16)$$

where A could be any square integrable vector or scalar valued function with $\tilde{A}$ as its Fourier transformation. Note that besides the purely mathematical advantages of utilizing the Fourier transforms, it has the important physical interpretation that in the reciprocal space, $\mathbf{q}$ and ω are the wave vector and the frequency of the particles or the collective, i.e., wave-like modes in the material.

The 2D Fourier transformation introduced by Eq. (2.15), transforms the differential operators such as the gradient and divergence to the algebraic ones, according to the

following rules

$$\mathcal{F}\{\nabla_{\parallel} A(\mathbf{r}, z, t)\} = i\mathbf{q}\tilde{A}(\mathbf{q}, z, \omega), \quad (2.17)$$

$$\mathcal{F}\{\nabla_{\parallel} \cdot \mathbf{A}(\mathbf{r}, z, t)\} = i\mathbf{q} \cdot \tilde{\mathbf{A}}(\mathbf{q}, z, \omega), \quad (2.18)$$

which gives rise to the following expression for the transformed Laplacian operator

$$\mathcal{F}\{\nabla_{\parallel}^2 A(\mathbf{r}, z, t)\} = \mathcal{F}\{\nabla_{\parallel} \cdot \nabla_{\parallel} A(\mathbf{r}, z, t)\} = -q^2 \tilde{A}(\mathbf{q}, z, \omega). \quad (2.19)$$

In these expressions $\nabla_{\parallel}$ stands for the in-plane differential operator $(\frac{\partial}{\partial x}, \frac{\partial}{\partial y})$. Another exceptionally important property of the Fourier transformation that makes it of great practical importance is its ability to transform convolutions into simple algebraic multiplications. For instance, taking the Fourier transformation of the constitutive relation in Eq. (2.14) gives

$$\tilde{\mathbf{D}}(\mathbf{q}, z, \omega) = \epsilon(\mathbf{q}, z, \omega) \tilde{\mathbf{E}}(\mathbf{q}, z, \omega), \quad (2.20)$$

in which all entities are the Fourier transformation of the corresponding functions of the Eq. (2.14), and it is assumed that

$$\epsilon(\mathbf{R}, \mathbf{R}', t, t') = \epsilon(\mathbf{r} - \mathbf{r}', z, t - t') \delta(z - z'), \quad (2.21)$$

which describes a homogeneous dependency in the $\mathbf{r}$ and t variables, and a local dependency in the z variable. Of course, the relation between the displacement and electric fields in Eq.(2.20) should be written more generally involving a convolution over the z variable, because the translational invariance in that direction is broken in heterostructures involving 2D materials. It is customary to omit the tilde sign above the Fourier transformed functions as was done in Eq. (2.20) for dielectric function $\epsilon(\mathbf{q}, z, \omega)$. However, their dependence on $\mathbf{q}$ and ω indicates that they are described in reciprocal space.

2.2 Response functions of 2D materials

In this section three important response functions of the 2D materials are introduced. These functions are the connecting bridges between the electromagnetic theory, and the internal structure and dynamics of the electric charges inside the 2DEG and the supporting material. The precise description of the internal charges of a material and their response to external fields demands a fully quantum mechanical treatment and there is no simple closed form expressions for these functions. Therefore, different models with different

precision levels are adopted to model the materials. In this section the definition of these characteristic functions and their relation to each other is provided. In the next chapter specific assumptions are made on the behavior of the internal charges of the materials, and the corresponding models for the characteristic functions are derived. Each of these models have their advantages and disadvantages, and each of them can provide a precise description of the material in some specific frequency and wave number range.

2.2.1 Polarization function

For the standard system shown in Fig. 2.1, the Poisson's equation for the electric potential Eq. (2.13) takes the following form in terms of the in-plane gradient and divergence operators

$$\nabla_{||} \cdot [\epsilon(\mathbf{r}, z, t) * \nabla_{||} \Phi(\mathbf{r}, z, t)] + \frac{\partial}{\partial z} \left[\epsilon(\mathbf{r}, z, t) * \frac{\partial}{\partial z} \Phi(\mathbf{r}, z, t) \right] = -4\pi [\rho_{\text{ext}}(\mathbf{r}, z, t) + \delta(z - z_0) \rho_{2D}(\mathbf{r}, t)], \quad (2.22)$$

where the z axis is chosen to be perpendicular to the flat surfaces of the substrate and the 2DEG. The surface charge density function $\rho_{2D}(\mathbf{r}, t)$ is the induced charge density about the equilibrium state of the 2D material at the position $z = z_0$. The function $\rho_{2D}(\mathbf{r}, t)$ imports the effect of the existing 2DEG into our model as a 2D charge density that appears in the Poisson's equation and the matching conditions of its solution. Then the Fourier transformed version of the Poisson's equation (2.22) is given by

$$q^2 \epsilon(\mathbf{q}, z, \omega) \tilde{\Phi}(\mathbf{q}, z, \omega) - \frac{\partial}{\partial z} \left[\epsilon(\mathbf{q}, z, \omega) \frac{\partial}{\partial z} \tilde{\Phi}(\mathbf{q}, z, \omega) \right] = 4\pi \tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega); \quad z \neq z_0, \quad (2.23)$$

where $q^2 = \mathbf{q} \cdot \mathbf{q}$. In Eq.(2.23), we assumed a local form of the dielectric response in the regions with the perpendicular coordinates $z \neq z_0$. This ordinary differential equation (ODE) in z can be then analytically solved in a piece-wise manner, because $\epsilon(\mathbf{q}, z, \omega)$ is a constant function of z in each region. The solution of the Eq. (2.23) is the fundamental electric potential, which is the most important quantity in our problems. In the standard system shown in Fig. 2.1 there are two interfaces (i.e., the substrate-vacuum and the vacuum-2DEG), and each interface is defined by its z coordinate. According to the principles of the electromagnetic theory, at each interface appropriate matching conditions must be satisfied. For instance, these matching conditions at z_0 , which is the z coordinate

of the 2DEG could be written as [123]

$$\begin{cases} \tilde{\Phi}(\mathbf{q}, z_0^+, \omega) = \tilde{\Phi}(\mathbf{q}, z_0^-, \omega), \\ \epsilon(\mathbf{q}, z_0^+, \omega) \frac{\partial}{\partial z} \tilde{\Phi}(\mathbf{q}, z_0^+, \omega) - \epsilon(\mathbf{q}, z_0^-, \omega) \frac{\partial}{\partial z} \tilde{\Phi}(\mathbf{q}, z_0^-, \omega) = -4\pi \tilde{\rho}_{2D}(\mathbf{q}, \omega), \end{cases} \quad (2.24)$$

where the first equation implies the continuity of the electric potential at the interface, and in the second equation $\tilde{\rho}_{2D}(\mathbf{q}, \omega) = \mathcal{F}\{\rho_{2D}(\mathbf{r}, t)\}$ is used to impose the electric displacement jump condition. Note that the solutions of the ODE in Eq. (2.23) for $\tilde{\Phi}(\mathbf{q}, z, \omega)$, on both sides of the 2DEG are connected by the surface charge density $\tilde{\rho}_{2D}(\mathbf{q}, \omega)$.

The polarization function is the response function that is postulated to make a connection between the induced charge density on a 2D material, and the electric potential on the 2D material in the Fourier domain. The polarization function $\chi(\mathbf{q}, \omega)$ is defined by

$$\tilde{\rho}_{2D}(\mathbf{q}, \omega) = -e^2 \chi(\mathbf{q}, \omega) \tilde{\Phi}(\mathbf{q}, z_0, \omega), \quad (2.25)$$

where e is the electron charge, $\tilde{\rho}_{2D}(\mathbf{q}, \omega)$ is the Fourier transformation of the induced charge density on the 2D material, and $\tilde{\Phi}(\mathbf{q}, z_0, \omega)$ is the Fourier transform of the total electric potential at the position of the 2D material that we assumed to be situated in the $z = z_0$ plane. Note that based on Eq. (2.25), the solution of the Poisson's problem Eq. (2.23) is used in the matching conditions of the same problem through Eq. (2.24), hence, our solution would be a self consistent solution of the Poisson's equation. Therefore, it is the polarization function $\chi(\mathbf{q}, \omega)$ that takes the behavior of the specific 2D material of the problem into account. Moreover, explicit mathematical models for $\chi(\mathbf{q}, \omega)$ are required for solving the Eq. (2.23). Specific models and their mathematical expressions for the $\chi(\mathbf{q}, \omega)$ are introduced in the next chapter.

It should be mentioned that for the standard system shown in Fig. 2.1

$$\epsilon(\mathbf{q}, z, \omega) = \begin{cases} 1 & \text{for } z > z_0, \\ 1 & \text{for } z > z_0 - h \text{ and } z < z_0, \\ \epsilon_s(\mathbf{q}, \omega) & \text{for } z < z_0 - h, \end{cases} \quad (2.26)$$

where $\epsilon_s(\mathbf{q}, \omega)$ is the dielectric function or permittivity of the substrate.

2.3 Dielectric function

If in the standard system, we remove the substrate and the 2D material, then Eq. (2.23) reduces to

$$q^2 \tilde{\Phi}_{\text{ext}}(\mathbf{q}, z, \omega) - \frac{\partial^2}{\partial z^2} \tilde{\Phi}_{\text{ext}}(\mathbf{q}, z, \omega) = 4\pi \tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega), \quad (2.27)$$

where the dielectric function $\epsilon(\mathbf{q}, z, \omega)$ is set to 1, that is, the dielectric constant of the free space, and $\tilde{\Phi}_{\text{ext}}(\mathbf{q}, z, \omega)$ is the external potential, due to the external charge density $\tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega)$. The boundary conditions of the Eq. (2.27) are the zero at infinity condition that implies $\lim_{z \rightarrow \pm\infty} \tilde{\Phi}_{\text{ext}}(\mathbf{q}, z, \omega) = 0$. If one solves the Eq. (2.23) along with the discussed matching conditions in a piece-wise manner, then the ratio of the solution of Eq. (2.27) to the solution of Eq. (2.23), when $z = z_0$, may be called the *effective 2D dielectric function* of the 2D material denoted by $\epsilon_{2D}(\mathbf{q}, \omega)$. In other words

$$\tilde{\Phi}(\mathbf{q}, z_0, \omega) = \frac{\tilde{\Phi}_{\text{ext}}(\mathbf{q}, z_0, \omega)}{\epsilon_{2D}(\mathbf{q}, \omega)}, \quad (2.28)$$

which expresses a constitutive relation between the external potential and the screened total potential in the plane of the 2DEG. The effective 2D dielectric function can be decomposed as

$$\epsilon_{2D}(\mathbf{q}, \omega) = \epsilon_{\text{bg}} + V_C(q)\chi(\mathbf{q}, \omega), \quad (2.29)$$

where ϵ_{bg} is the background dielectric function coming from the supporting material. We assume that the supporting material is isotropic so that ϵ_{bg} may be a function of $q = \sqrt{q_x^2 + q_y^2}$ and/or ω , and $V_C(q) = 2\pi e^2/q$ is the Fourier transform of the in-plane Coulomb interaction energy between two electrons of charge e .

2.4 Conductivity tensor

The in-plane Ohm's law in the 2DEG can be written as

$$\tilde{\mathbf{J}}_{2D}(\mathbf{q}, \omega) = \vec{\sigma}(\mathbf{q}, \omega) \cdot \tilde{\mathbf{E}}_{\parallel}(\mathbf{q}, z_0, \omega), \quad (2.30)$$

where $\tilde{\mathbf{J}}_{2D}(\mathbf{q}, \omega)$ is the 2D induced current density in the 2D material, and $\tilde{\mathbf{E}}_{\parallel}(\mathbf{q}, \omega)$ is the tangential component of the total electric field evaluated at the plane of 2D material. On the other hand, taking Fourier transform from both sides of the Eq. (2.8) gives

$$\tilde{\mathbf{E}}_{\parallel}(\mathbf{q}, z_0, \omega) = -i\mathbf{q}\tilde{\Phi}(\mathbf{q}, z_0, \omega). \quad (2.31)$$

Moreover, consistently with the Maxwell's equations, we invoke the fundamental law of the continuity of the electric charge that inside the plane of the 2D material takes the form

$$\nabla \cdot \mathbf{J}_{2D}(\mathbf{r}, t) + \frac{\partial}{\partial t} \rho_{2D}(\mathbf{r}, t) = 0. \quad (2.32)$$

The 2D charge continuity equation in the Fourier domain is expressed in the following form

$$\omega \tilde{\rho}_{2D}(\mathbf{q}, \omega) = \mathbf{q} \cdot \tilde{\mathbf{J}}_{2D}(\mathbf{q}, \omega). \quad (2.33)$$

After some simple algebraic manipulations on Eqs. (2.25), (2.30), (2.31), and (2.33) we find

$$\chi(\mathbf{q}, \omega) = \frac{i}{e^2 \omega} \mathbf{q} \cdot \vec{\sigma}(\mathbf{q}, \omega) \cdot \mathbf{q}, \quad (2.34)$$

which is the relation between the polarization function and the conductivity tensor of an anisotropic 2D material. This relation is of great theoretical importance, because in many situations, the conductivity is easier to model rather than the polarization.

It is worthwhile to mention that the in-plane conductivity tensor can generally be written in terms of dyadic products of the unit vectors $\hat{\mathbf{x}}$ and $\hat{\mathbf{y}}$ along the x and y axes as

$$\vec{\sigma}(\mathbf{q}, \omega) = \sigma_{xx} \hat{\mathbf{x}} \hat{\mathbf{x}} + \sigma_{xy} \hat{\mathbf{x}} \hat{\mathbf{y}} + \sigma_{yx} \hat{\mathbf{y}} \hat{\mathbf{x}} + \sigma_{yy} \hat{\mathbf{y}} \hat{\mathbf{y}}, \quad (2.35)$$

with its Cartesian components satisfying certain symmetry conditions. [124, 125]

2.5 Green's functions of two specific setups

In this section the Green's function $G(z, z')$ of two systems that are extensively used in this thesis are introduced. The main structure for both of these systems is quite similar to the standard system introduced in Fig. 2.1, and the difference is in the position of the source (z') and the probe (z). The detailed derivations for these results is provided in appendix A.

2.5.1 Source and probe above the 2DEG

Assume that a 2DEG is situated in the plane $z = 0$, and the region $z \leq -d$ is occupied by a semi-infinite substrate with dielectric permittivity $\epsilon_s(\omega)$. We are interested in the Green's function of the Poisson problem in the region $z \geq 0$, when the external electric charge distribution might be non-zero only for $z > 0$. This Green's function is the solution of the following problem

$$q^2 \epsilon(z) G(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G(z, z') \right] = 4\pi [\delta(z - z') + \delta(z) \rho_{2D}]; \quad z, z' \geq 0, \quad (2.36)$$

in which the $\mathbf{q}$ and ω dependence of the functions $\epsilon(z)$, ρ_{2D} , and $G(z, z')$ are omitted for simplicity. Eq. (2.36) is in fact Eq. (2.23) in which $\rho_{\text{ext}}(z)$ is replaced by $\delta(z - z')$, which represents a unit point source charge that is required in the definition of the Green's function. According to the linearity of the equation (2.36), its solution can be decomposed as

$$G(z, z') = G_0(z, z') + G_{\text{ind}}(z, z'), \quad (2.37)$$

where $G_0(z, z')$ is the external potential that is caused by the unit point charge $\delta(z - z')$ as if there were no substrate or 2DEG, and $G_{\text{ind}}(z, z')$ is the induced potential that is the response of the 2DEG and the substrate to the external charge. Therefore, $G_0(z, z')$ and $G_{\text{ind}}(z, z')$ are the solutions to the following problems

$$\begin{cases} q^2 G_0(z, z') - \frac{\partial}{\partial z} \left[\frac{\partial}{\partial z} G_0(z, z') \right] = 4\pi\delta(z - z'), \\ q^2 \epsilon(z) G_{\text{ind}}(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_{\text{ind}}(z, z') \right] = 4\pi\delta(z) \rho_{2D}. \end{cases} \quad (2.38)$$

Note that the matching conditions for the solutions of the Poisson equation must also be considered. The continuity condition can be considered separately for each equation, but the other condition, which is the jump in electric displacement must be considered for the total potential $G(z, z')$. In the appendix A.1 it is shown that for this problem these Green's functions are given by

$$G_0(\mathbf{q}, z, z') = \frac{2\pi}{q} e^{-q|z-z'|}, \quad (2.39)$$

and

$$\begin{aligned} G_{\text{ind}}(\mathbf{q}, z, z', \omega) &= \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - 1 \right] \frac{2\pi}{q} e^{-q(z+z')} \\ &= \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - 1 \right] G_0(\mathbf{q}, 0, z') e^{-qz}; \quad z, z' \geq 0, \end{aligned} \quad (2.40)$$

with $\epsilon_{2D}(\mathbf{q}, \omega)$ from Eq. (2.29), in which

$$\epsilon_{\text{bg}}(q, \omega) = \left[1 - \frac{\epsilon_s(\omega) - 1}{\epsilon_s(\omega) + 1} e^{-2qd} \right]^{-1}. \quad (2.41)$$

Although the total potential $\tilde{\Phi}(\mathbf{q}, z, \omega)$ can be obtained from the Green's function via

$$\tilde{\Phi}(\mathbf{q}, z, \omega) = \int dz' \tilde{\rho}_{\text{ext}}(\mathbf{q}, z', \omega) G(z, z'), \quad (2.42)$$

in many cases the induced potential

$$\tilde{\Phi}_{\text{ind}}(\mathbf{q}, z, \omega) = \int dz' \tilde{\rho}_{\text{ext}}(\mathbf{q}, z', \omega) G_{\text{ind}}(z, z') \quad (2.43)$$

plays the critical role. For instance, the external potential does not produce any force on the charged particles, and the induced potential is responsible for all forces acting on the charges.

2.5.2 Source and probe inside the substrate

Assume that the region $z \leq 0$ is occupied by a semi-infinite substrate with dielectric permittivity $\epsilon_s(\omega)$, and a 2DEG is placed in the plane $z = h$. We are interested in the Green's function of the Poisson problem Eq. (2.23) in the region $z \leq 0$, which is the region occupied by the substrate. This Green's function is the solution of the ODE

$$q^2 \epsilon(z) G(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G(z, z') \right] = 4\pi [\delta(z - z') + \delta(z - h) \rho_{2D}]; \quad z, z' \leq 0, \quad (2.44)$$

in which the $\mathbf{q}$ and ω dependence of the functions $\epsilon(z)$, $G(z, z')$, and ρ_{2D} are omitted for simplicity. Linearity allows to write

$$G(z, z') = G_0(z, z') + G_{\text{ind}}(z, z'), \quad (2.45)$$

with $G_0(z, z')$ and $G_{\text{ind}}(z, z')$ being the solutions of

$$\begin{cases} q^2 \epsilon(z) G_0(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_0(z, z') \right] = 4\pi \delta(z - z'), \\ q^2 \epsilon(z) G_{\text{ind}}(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_{\text{ind}}(z, z') \right] = 4\pi \delta(z - h) \rho_{2D}. \end{cases} \quad (2.46)$$

In this decomposition of the Green's function, $G_0(z, z')$ represents the potential that is merely due to the external unit point charge $\delta(z - z')$ and its interaction with the substrate. On the other hand, $G_{\text{ind}}(z, z')$ represents the potential that arises as the response of the 2DEG to the external charge and the substrate. This decomposition is important, because this Green's function will be used to compute the energy dissipation that is caused by the interactions between the external charges and the charge carriers in the 2DEG, without considering the dissipation that is merely related to the interaction of the external charges

and the substrate. In the appendix [A.2](#) it is shown that the explicit expression for $G_{\text{ind}}(z, z')$ happens to be

$$G_{\text{ind}}(\mathbf{q}, z, z', \omega) = \frac{2\pi}{q} \left[\frac{1}{\epsilon_{2\text{D}}(\mathbf{q}, \omega)} - \frac{1}{\epsilon_{\text{bg}}(q, \omega)} \right] \left[\frac{\epsilon_{\text{bg}}(q, \omega)}{\epsilon_{\text{bg}}^0} \right]^2 e^{q(z+z'-2h)}; \quad z, z' \leq 0, \quad (2.47)$$

where $\epsilon_{2\text{D}}(\mathbf{q}, \omega)$ and $\epsilon_{\text{bg}}(q, \omega)$ are given in Eqs. [\(2.29\)](#) and [\(2.41\)](#), respectively, and

$$\epsilon_{\text{bg}}^0 = \frac{\epsilon_s(\omega) + 1}{2}. \quad (2.48)$$

In this configuration, the frequency dependence of the substrate dielectric function $\epsilon_s(\omega)$ will be dropped and ϵ_s will be considered a real-valued constant describing lossless substrate.

2.6 Energy loss function and the energy loss rate

In this section, first the energy loss function is defined, then it is shown how this function naturally appears in the relations that describe the energy loss rate of the moving particles in the standard system shown in [Fig. 2.1](#).

2.6.1 Energy loss function

For the standard system, the energy loss function (ELF) of a 2D material is defined as

$$L(\mathbf{q}, \omega) = \Im \left\{ \frac{-1}{\epsilon_{2\text{D}}(\mathbf{q}, \omega)} \right\}, \quad (2.49)$$

where $\Im$ stands for the imaginary part.

2.6.2 Energy loss rate

Consider the standard system that consists of a semi-infinite substrate that occupies the region $z \leq -d$, and a 2DEG that is situated in the plane $z = 0$. Assume that a charged particle with electric charge Ze is moving with velocity $\mathbf{v}$ parallel to the 2DEG, at height z_0 above it. This particle can be characterized by

$$\rho_{\text{ext}}(\mathbf{r}, z, t) = Ze\delta(\mathbf{r} - \mathbf{v}t)\delta(z - z_0). \quad (2.50)$$

Moreover, the Fourier transform of this charge density is given by

$$\tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega) = 2\pi Ze\delta(\omega - \mathbf{q} \cdot \mathbf{v})\delta(z - z_0). \quad (2.51)$$

According to classical mechanics, a particle with the velocity $\mathbf{v}$ that is under the action of some force $\mathbf{F}$, loses its energy at the rate

$$\frac{dW}{dt} = \mathbf{F} \cdot \mathbf{v}. \quad (2.52)$$

Therefore, in order to compute the energy loss rate of the moving charge introduced in the Eq. (2.51), first we need to find the electric force $\mathbf{F}$ acting on it. On the other hand, the electric force acting on a point charge is the product of the charge and the induced electric field at its position, in other words

$$\mathbf{F} = Ze\mathbf{E}_{\text{ind}}(\mathbf{v}t, z_0, t) = -Ze\nabla\Phi_{\text{ind}}(\mathbf{v}t, z_0, t) = -Ze\left(\nabla_{\parallel} + \hat{\mathbf{z}}\frac{\partial}{\partial z}\right)\Phi_{\text{ind}}(\mathbf{v}t, z_0, t). \quad (2.53)$$

In the Fourier space Eq. (2.53) takes the form

$$\tilde{\mathbf{F}} = \left[-Ze\left(i\mathbf{q} + \hat{\mathbf{z}}\frac{\partial}{\partial z}\right)\tilde{\Phi}_{\text{ind}}(\mathbf{q}, z, \omega)\right]\Bigg|_{z=z_0}. \quad (2.54)$$

On the other hand

$$\tilde{\Phi}_{\text{ind}}(\mathbf{q}, z, \omega) = \int dz' \tilde{\rho}_{\text{ext}}(\mathbf{q}, z', \omega)G_{\text{ind}}(\mathbf{q}, z, z', \omega), \quad (2.55)$$

with $\tilde{\rho}_{\text{ext}}(\mathbf{q}, z', \omega)$ from Eq. (2.51), that gives

$$\tilde{\Phi}_{\text{ind}}(\mathbf{q}, z, \omega) = 2\pi Ze\delta(\omega - \mathbf{q} \cdot \mathbf{v})G_{\text{ind}}(\mathbf{q}, z, z_0, \omega). \quad (2.56)$$

Therefore,

$$\tilde{\mathbf{F}} = \left[-2\pi(Ze)^2\left(i\mathbf{q} + \hat{\mathbf{z}}\frac{\partial}{\partial z}\right)\delta(\omega - \mathbf{q} \cdot \mathbf{v})G_{\text{ind}}(\mathbf{q}, z, z_0, \omega)\right]\Bigg|_{z=z_0}, \quad (2.57)$$

which can be transformed to the real space using the inverse Fourier transformation in Eq. (2.16) as

$$\mathbf{F} = \frac{-(Ze)^2}{(2\pi)^2} \int d^2\mathbf{q} \left[\left(i\mathbf{q} + \hat{\mathbf{z}}\frac{\partial}{\partial z}\right)G_{\text{ind}}(\mathbf{q}, z, z_0, \mathbf{q} \cdot \mathbf{v})\right]\Bigg|_{z=z_0}, \quad (2.58)$$

in which the sifting property of the Dirac delta function is used. Note that this force consists of two components, the first component is in the $\mathbf{q}$ plane and is parallel to the 2DEG and is called the stopping force, while the second component involves the unit vector $\hat{\mathbf{z}}$ and is perpendicular to the 2DEG and is called the image force. For the purposes of this section, let us just consider the stopping force, which is denoted by $\mathbf{F}_{\parallel}$ while we drop the image force because it does no work. The explicit expression for this force is given by

$$\mathbf{F}_{\parallel} = \frac{-(Ze)^2}{(2\pi)^2} \int d^2\mathbf{q} \, i\mathbf{q} G_{\text{ind}}(\mathbf{q}, z_0, z_0, \mathbf{q} \cdot \mathbf{v}), \quad (2.59)$$

that can be decomposed based on the real and imaginary parts of the Green's function $G_{\text{ind}}(\mathbf{q}, z_0, z_0, \mathbf{q} \cdot \mathbf{v})$,

$$\mathbf{F}_{\parallel} = \frac{-(Ze)^2}{(2\pi)^2} \int d^2\mathbf{q} [i\mathbf{q} \Re\{G_{\text{ind}}(\mathbf{q}, z_0, z_0, \mathbf{q} \cdot \mathbf{v})\} - \mathbf{q} \Im\{G_{\text{ind}}(\mathbf{q}, z_0, z_0, \mathbf{q} \cdot \mathbf{v})\}]. \quad (2.60)$$

Since $G_{\text{ind}}(\mathbf{q}, z, z', \omega)$ is the Fourier transform of the electric potential that is a real valued function, it necessarily satisfies the Hermitian parity in $(\mathbf{q}, \omega)$ that means

$$G_{\text{ind}}(-\mathbf{q}, z, z, -\omega) = G_{\text{ind}}^*(\mathbf{q}, z, z, \omega), \quad (2.61)$$

where the superscript $*$ stands for the complex conjugate operation. Equation (2.61) means that the real and imaginary parts of the Green's functions are even and odd functions in $(\mathbf{q}, \omega)$, respectively. By virtue of this parity, the integral in the Eq. (2.60) that is taken over the real part vanishes and Eq. (2.60) reduces to

$$\mathbf{F}_{\parallel} = \frac{(Ze)^2}{(2\pi)^2} \int d^2\mathbf{q} \, \mathbf{q} \Im\{G_{\text{ind}}(\mathbf{q}, z_0, z_0, \mathbf{q} \cdot \mathbf{v})\}, \quad (2.62)$$

which could be written as

$$\mathbf{F}_{\parallel} = \frac{(Ze)^2}{2\pi} \int d^2\mathbf{q} \, e^{-2qz_0} \hat{\mathbf{q}} \Im\left\{\frac{1}{\epsilon_{2D}(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})}\right\}, \quad (2.63)$$

with $\hat{\mathbf{q}} = \frac{\mathbf{q}}{q}$, upon using the Eq. (2.40). The derivation above demonstrates how the energy loss function emerges in the expressions related to power dissipation. Referring to Eqs. (2.37), (2.40), (2.42) and (2.43), we note that our replacing G with G_{ind} is equivalent to the procedure in Chapter 14 of Ref. [120], where $1/\epsilon_{2D}$ is replaced by $(1/\epsilon_{2D} - 1)$.

2.7 Plasmon dispersion

When all the damping parameters in the models used for $\epsilon_{2D}(\mathbf{q}, \omega)$ are set to zero, the equation

$$\epsilon_{2D}(\mathbf{q}, \omega) = 0, \quad (2.64)$$

has real roots and defines a surface in the $(\mathbf{q}, \omega)$ space. This surface that is a relation between the wave vectors $\mathbf{q}$ and the frequencies ω , is called the plasmon dispersion relation. On the other hand, Eq. (2.28) can be written as

$$\epsilon_{2D}(\mathbf{q}, \omega) \tilde{\Phi}(\mathbf{q}, z_0, \omega) = \tilde{\Phi}_{ext}(\mathbf{q}, z_0, \omega), \quad (2.65)$$

that means for $(\mathbf{q}, \omega)$ satisfying the dispersion relation Eq. (2.64), the total potential $\tilde{\Phi}(\mathbf{q}, z_0, \omega)$ can be non-zero even if the external potential $\tilde{\Phi}_{ext}(\mathbf{q}, z_0, \omega)$ is zero. Therefore, plasmons can be interpreted as the self-sustaining oscillations of the electron gas. Moreover, when the damping parameters in the model are non-zero, the loss function is strongly peaked around the $(\mathbf{q}, \omega)$ values satisfying the dispersion relation of the damp-less system. This shows that the main energy loss channels are occurring around the dispersion curves.

Chapter 3

Theoretical models for phosphorene dielectric function

3.1 Introduction

In this chapter three different models for the dielectric function $\epsilon_{2D}(\mathbf{q}, \omega)$ of phosphorene are introduced. These models that we call optical, semiclassical, and quantum mechanical models are presented in the order of the increasing complexity. The third model is completely different in nature from the first two models, and will be used to find an approximation for the dielectric function of the phosphorene that is only expanded to the linear term in ω . On the other hand the first two models are closely related to each other and a precise comparison will be made between them and with the Drude model, which is an important component of those two models. This chapter is organized so that for clarity, the results are presented within the main text, while the proofs of the important relations are moved to appendix [B](#).

3.2 Band theory of the 2D crystals

Band theory of the 2D solids in the one-electron approximation relies on the solutions of the stationary Schrodinger equation [\[126\]](#)

$$-\frac{\hbar^2}{2m}\nabla_{\parallel}^2\psi(\mathbf{r}) + V(\mathbf{r})\psi(\mathbf{r}) = \varepsilon\psi(\mathbf{r}), \quad (3.1)$$

where $\psi(\mathbf{r})$ is the stationary wavefunction of the electron, and $V(\mathbf{r})$ is the potential energy due to the nuclei of the atoms in the crystal and the charge distribution of the other outer-shell electrons. In a perfect 2D crystal, the potential $V(\mathbf{r})$ is a periodic function of $\mathbf{r}$ with the same periods as the crystal. In this situation, the solutions of the Eq. (3.1) are of the form [5, 121, 127]

$$\psi(\mathbf{p}, \mathbf{r}) = u_{\mathbf{p}}(\mathbf{r}) e^{i \frac{\mathbf{p} \cdot \mathbf{r}}{\hbar}}, \quad (3.2)$$

where $\psi(\mathbf{p}, \mathbf{r})$ is called the Bloch wavefunction for the electron with state (wave vector) $\mathbf{p} = \hbar \mathbf{k}$, and $u_{\mathbf{p}}(\mathbf{r})$ is a periodic function with the same period as $V(\mathbf{r})$. In the Bloch function representation, each state $\mathbf{p}$ is associated with a single or a set of eigen energies denoted by $\varepsilon_{\mathbf{p}}$. The function $\varepsilon = \varepsilon_{\mathbf{p}}$ defines the electronic energy band structure and is sometimes called the electronic dispersion relation of the material.

3.2.1 Effective mass approximation

In the simplest approximation, one can assume that $V(\mathbf{r})$ is a constant function (i.e. the free space potential), which results in the plane wave solutions to the Schrodinger equation. In this case the electronic dispersion relation of the material reduces to

$$\varepsilon_{\mathbf{p}} = \frac{p^2}{2m} = \frac{1}{2m} (p_x^2 + p_y^2). \quad (3.3)$$

In the case of 2D crystals, the exact electronic band structure could have a quite complicated structure that must be computed by solving an eigen value problem upon substituting Eq. (3.2) into Eq. (3.1). For the semiconductors such as phosphorene, the electronic dispersion relation can still be well approximated by an elliptic paraboloid at the vicinity of the minimum of the lowest conduction band. Taking that minimum as the zero of energy located at $\mathbf{p} = \mathbf{0}$ allows us to approximate the conduction band with

$$\varepsilon_{\mathbf{p}} = \frac{p_x^2}{2m_x} + \frac{p_y^2}{2m_y}, \quad (3.4)$$

where m_x and m_y are called the effective masses of the electrons. Similar analysis applies in the vicinity of the maximum of the topmost valence band occupied by holes with the effective masses m_x and m_y . From a mathematical viewpoint, m_x and m_y are proportional to the curvature of the band structure at the local minima to ensure the perfect match between the derivatives of the approximating function and the exact band structure, up to second order derivative. On the other hand, from a physical point of view, the periodicity

of the crystal potential is modeled via these effective masses such that one can think of the 2DEG as a non-interacting free electron gas with the modified masses [126].

Fermi energy is a specific energy level with the unique property that at zero temperature and in the ground state all states with energies up to the Fermi level are filled with electrons, and all other states are empty. According to the common facts from the solid state physics [127], a completely empty or completely full band does not contribute in the electrical conduction. Therefore, in the semiconductors like phosphorene the closest bands to the Fermi level, specifically those that are crossed by the Fermi level are the most important bands to determine the conductivity of the material. Moreover, doping densities of charge carriers in many applications are small enough to ensure that the lowest conduction band (the first band above the Fermi level) or the topmost valence band (the first band below the Fermi level) are slightly crossed by the Fermi level so that an effective mass approximation can still provide a precise description of the material. Note that in the presence of weak fields or low energy photons, transitions mostly occur between the states with energies close to the Fermi level in a single band, while in the presence of the stronger fields or higher energy photons, transitions might occur between two different bands with finite energy gap between them. These transitions are called the *intraband* and *interband* electronic transitions, respectively.

3.3 Hydrodynamic model of isotropic 2DEG

In the case of an isotropic 2DEG, where the effective electron masses are equal, $m_x = m_y = m_*$, Fetter developed in Ref. [58] a hydrodynamic model, which yields the conductivity in the form

$$\sigma_{\text{hydro}}(q, \omega) = \frac{i}{\pi} \frac{\omega D}{\omega(\omega + i\gamma) - s^2 q^2},$$

where $D = \pi e^2 n / m_*$ is the so-called Drude weight with n being the equilibrium density of electrons, γ is a phenomenological damping constant, and $s = v_F / \sqrt{2}$ is the speed of sound in the (neutral, $e = 0$) 2DEG with $v_F = \hbar k_F / m_*$ being the Fermi speed and $k_F = \sqrt{2\pi n}$ the Fermi wavenumber.

The term $s^2 q^2$ in the denominator of the above expression, which results from the Fermi pressure in the 2DEG at zero temperature, can be perceived as a non-local, i.e., q -dependent, correction due to finite density n . In the limit of small n , the above expression may be approximated by its long-wavelength (or local, i.e., $q \rightarrow 0$) limit

$$\sigma_{\text{Drude}}(\omega) = \frac{i}{\pi} \frac{D}{\omega + i\gamma}.$$

This is the well-known Drude conductivity model, which expresses a universal form of the dynamic response of any 2DEG in the optical limit ($q \rightarrow 0$).

In the case of graphene, which exhibits a linear energy dispersion around the Dirac point of graphene's π bands, $\varepsilon_{\mathbf{p}} = v_F^g \|\mathbf{p}\|$, where $v_F^g \approx c/300$ is the (constant) Fermi speed, the effective mass in the hydrodynamic model should be replaced by the cyclotron mass [128] $m_* = \varepsilon_F^g / (v_F^g)^2$, where $\varepsilon_F^g = \hbar v_F^g k_F^g$ is the Fermi energy (chemical potential at zero temperature) in graphene doped by the equilibrium charge density n with $k_F^g = \sqrt{\pi n}$ being its Fermi wavenumber. Thus, the Drude weight in the Drude conductivity model for doped graphene may be expressed as $D = \frac{e^2}{\hbar^2} \varepsilon_F^g$.

However, for an anisotropic 2DEG, several modifications in the conductivity model(s) are required to generalize the Drude model and to take into account quantum-mechanical corrections for non-zero wavevectors that go beyond the hydrodynamic model of the isotropic 2DEG.

3.4 Optical model

From a technological point of view, the THz-MIR frequency range is of great importance and satisfactory empirical analytical models for the dielectric function of the materials in the range of low frequencies and long wavelength are required. The $\mathbf{q}$ dependence of the components of the conductivity tensor in Eq. (2.35) describes the non-local effects in the conductivity tensor that demonstrate the effect of the electric field at $\mathbf{r}'$ on the conductivity at $\mathbf{r}$. When the wavelength of the incident electromagnetic fields is long enough as in the THz-MIR frequency range, one can set $\mathbf{q} \rightarrow \mathbf{0}$. This limit in which the components of the conductivity $\vec{\sigma}(\omega)$ are just functions of the frequency ω is called the optical limit, and the models for the dielectric or polarization functions that rely on this assumption are called the optical models [129]. In the optical limit the conductivity tensor is diagonalizable [1, 32], so that the diagonal elements, $\sigma_{xx} = \sigma_x(\omega)$ and $\sigma_{yy} = \sigma_y(\omega)$ remain functions of frequency, whereas the off-diagonal elements vanish, $\sigma_{xy} = \sigma_{yx} = 0$. Therefore, in the optical limit the polarization function from Eq. (2.34) takes the form

$$\chi(\mathbf{q}, \omega) = \frac{i}{e^2 \omega} [q_x^2 \sigma_x(\omega) + q_y^2 \sigma_y(\omega)]. \quad (3.5)$$

In general, for an anisotropic 2D material such as phosphorene, the diagonal elements of the conductivity tensor are not identical, $\sigma_x(\omega) \neq \sigma_y(\omega)$, with the principal axes x and y that may be taken along its AC and ZZ directions, respectively [1, 130]. Moreover, the

diagonal elements of the conductivity tensor can be decomposed into two components due to the intraband or interband electron transitions [1, 129]

$$\sigma_j(\omega) = \sigma_j^{\text{intra}}(\omega) + \sigma_j^{\text{inter}}(\omega) \quad \text{for } j = x, y, \quad (3.6)$$

which results in the following decomposition of the polarization function into the intraband and interband components upon using Eqs. (2.34) and (3.6)

$$\chi(\mathbf{q}, \omega) = \chi_{\text{intra}}(\mathbf{q}, \omega) + \chi_{\text{inter}}(\mathbf{q}, \omega). \quad (3.7)$$

It turns out that simple phenomenological models can be developed for both the intraband and interband contributions for the optical conductivity of a 2D material at low frequencies, covering the technologically relevant THz-MIR range of frequencies.

3.4.1 Drude model for intraband conductivity

Drude model is a well established model for the in-plane conductivity of 2D materials due to the intraband transitions. The anisotropic Drude model can be used to describe the intraband conductivity tensor of the phosphorene $\sigma_x^{\text{intra}}(\omega)$ and $\sigma_y^{\text{intra}}(\omega)$, in the THz-MIR frequency range with an acceptable accuracy for many applications. The mathematical expressions for the diagonal elements of the phosphorene intraband conductivity tensor within the Drude model are given by [1, 32, 36]

$$\sigma_j^{\text{intra}}(\omega) = \frac{i}{\pi} \frac{D_j}{\omega + i\gamma_j}, \quad \text{for } j = x, y. \quad (3.8)$$

In this expression, D_j is the Drude weight in the direction $j = x, y$, which in the zero temperature can be described in terms of the equilibrium doping density n and an effective electron or hole mass m_j as

$$D_j = \frac{\pi e^2 |n|}{m_j}. \quad (3.9)$$

Moreover, $\gamma_j = 1/\tau_j$ is the damping rate in the direction $j = x, y$, which is an empirical parameter describing all the internal processes that scatter the electrons with mean free time τ_j . Note that using the definition of the DC conductivity tensor $\sigma_j^{\text{intra}}(0) = e|n|\mu_j$, the mean free time can be described as $\tau_j = \mu_j m_j / e$, for $j = x, y$. While the validity of the Drude model Eqs. (3.8) and (3.9) was demonstrated in the ab initio calculations [1], a heuristic derivation of this model is given in appendix B.1.

3.4.2 Linear approximation for interband conductivity

When considering the impact of the interband transitions on the conductivity tensor of semiconductors with band gap energy ε_g , the real part of this contribution $\Re\{\sigma_j^{\text{inter}}(\omega)\}$ can be neglected in the frequency range $0 \leq \omega \lesssim \omega_g$, with $\omega_g = \varepsilon_g/\hbar$. This characteristic of the $\Re\{\sigma_j^{\text{inter}}(\omega)\}$ that is confirmed by the *ab initio* calculations [1, 131], shows that the minimum energy required for an electromagnetic wave or a photon to induce an electron transition from the valence band to the conduction band is equal to the band gap of the semiconductor. Therefore, in the THz-MIR frequency range, the interband conductivity of phosphorene can be modeled only by its imaginary part $\Im\{\sigma_j^{\text{inter}}(\omega)\}$ in the limit $\omega \rightarrow 0$, because $\omega \ll \omega_g$ with $\omega_g \approx 2$ eV for phosphorene, and we can set $\Re\{\sigma_j^{\text{inter}}(\omega)\} = 0$. According to the *ab initio* calculations [1, 131], $\Im\{\sigma_j^{\text{inter}}(\omega)\}$ could be adequately approximated by a linear function with negative slope at the vicinity of $\omega = 0$. Hence, in the THz-MIR frequency range, the components of the phosphorene interband conductivity tensor can be represented by

$$\sigma_j^{\text{inter}}(\omega) = -i\alpha_j\omega \quad \text{for } j = x, y, \quad (3.10)$$

where $\alpha_j > 0$ may be regarded as the Cartesian components of its in-plane polarizability tensor in the *static limit*, which arise from high-energy interband electronic transitions. Moreover, based on the *ab initio* data, the numerical values of the constants α_j are almost independent of the doping density $|n|$ for the phosphorene when the doping density $|n|$ is sufficiently small.

Having the *ab initio* data of the interband conductivity tensor $\sigma_j^{\text{inter}}(\omega)$, one can obtain the parameters α_j defined in Eq. (3.10) via two different methods. The first method relies on the interpretation of $-\alpha_j$ in Eq. (3.10) as the slope in the limit as $\omega \rightarrow 0$ that gives

$$\alpha_j = \lim_{\omega \rightarrow 0} \frac{-\Im\{\sigma_j^{\text{inter}}(\omega)\}}{\omega}. \quad (3.11)$$

The second method uses the fact that $\sigma_j^{\text{inter}}(\omega)$ are complex valued analytic functions of ω , satisfying requirements of the Kramers-Kronig relations [121, 122, 132], hence, their imaginary part can be expressed as an integral in terms of the real part

$$\Im\{\sigma_j^{\text{inter}}(\omega)\} = -\frac{2}{\pi}\omega\mathcal{P}\int_0^\infty d\omega' \frac{\Re\{\sigma_j^{\text{inter}}(\omega')\}}{\omega'^2 - \omega^2}, \quad (3.12)$$

where $\mathcal{P}$ stands for the Cauchy's principal value of the integral over ω' . Therefore, we can

combine Eqs. (3.11) and (3.12) to write

$$\alpha_j = \frac{2}{\pi} \int_0^\infty d\omega \frac{\Re\{\sigma_j^{\text{inter}}(\omega)\}}{\omega^2}, \quad (3.13)$$

which allows to compute α_j from ab initio data for the real part of the interband optical conductivity as an integral in the form of a sum rule for each direction $j = x, y$. Note that the singularity of the integrand in Eq. (3.13) is removed as $\Re\{\sigma_j^{\text{inter}}(\omega)\}$ vanishes at $\omega = 0$.

In order to show the applicability of the linear approximation Eq. (3.10), both the real and imaginary parts of the interband optical conductivity of phosphorene that are obtained from the *ab initio* calculations are depicted in Fig. 3.1. These calculations are performed with different values of the doping density n , and the numerical values of α_j are obtained from Eq. (3.13). Figure 3.1 confirms that for $\hbar\omega \ll 2$ eV, the imaginary part of the interband conductivities can be precisely approximated by Eq. (3.10), while the real parts are negligibly small, as expected.

3.4.3 Phosphorene effective dielectric function within the optical limit

Given the expressions for the intraband and interband conductivities in Eqs. (3.8) and (3.10), one can obtain the intraband and interband contributions of the polarization function in Eq. (3.7) as

$$\chi_{\text{intra}}(\mathbf{q}, \omega) = \frac{-1}{\pi e^2 \omega} \left(\frac{D_x q_x^2}{\omega + i\gamma_x} + \frac{D_y q_y^2}{\omega + i\gamma_y} \right), \quad (3.14)$$

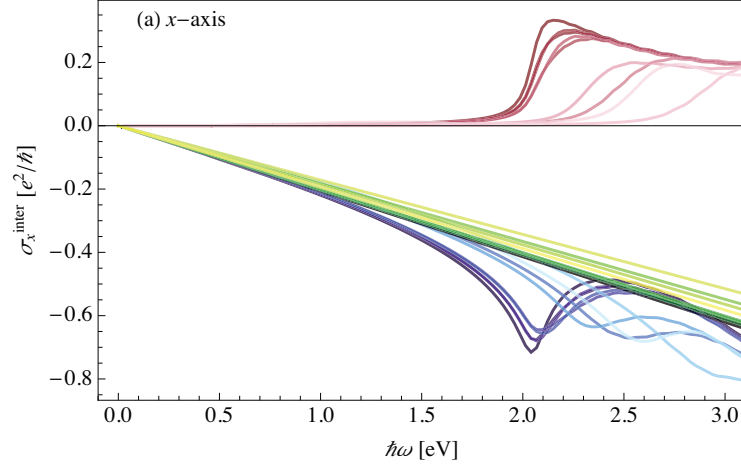
and

$$\chi_{\text{inter}}(\mathbf{q}, \omega) = \frac{1}{e^2} (\alpha_x q_x^2 + \alpha_y q_y^2). \quad (3.15)$$

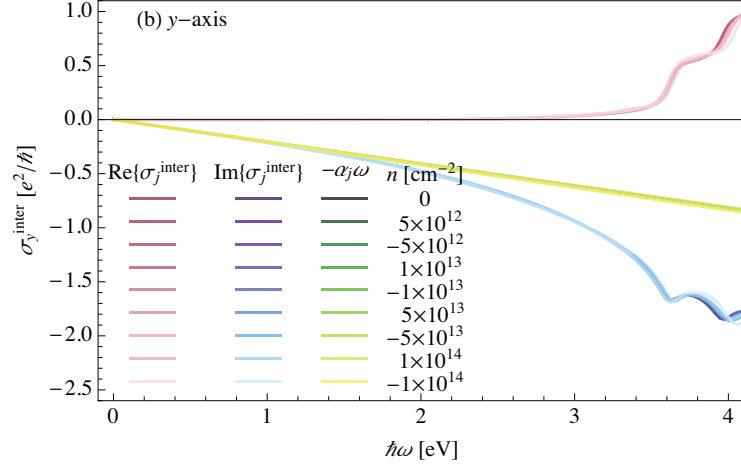
Therefore, the effective dielectric function of the phosphorene within the optical limit can be obtained from the Eqs. (2.29), (3.14), and (3.15) as

$$\epsilon_{2D}(\mathbf{q}, \omega) = \epsilon_{\text{bg}} + \frac{2\pi}{q} (\alpha_x q_x^2 + \alpha_y q_y^2) - \frac{2}{q\omega} \left(\frac{D_x q_x^2}{\omega + i\gamma_x} + \frac{D_y q_y^2}{\omega + i\gamma_y} \right). \quad (3.16)$$

This constitutes what we call the *optical model* of the effective dielectric function of phosphorene, which is expected to work well in the THz-MIR range of frequencies.



(a) Real (red curves) and imaginary (blue curves) parts of the interband optical conductivity for doped phosphorene, along the AC axis. Green curves are the linear approximation from Eq. (3.10).



(b) Real (red curves) and imaginary (blue curves) parts of the interband optical conductivity for doped phosphorene, along the ZZ axis. Green curves are the linear approximation from Eq. (3.10).

Figure 3.1: Numerical data presented in these figures, are obtained from *ab initio* calculations, performed with different doping densities (shown on panel b) [1].

3.5 Semiclassical model

In this section, we present a model for the effective dielectric function of the doped phosphorene that we call the *semiclassical model*. While the non-local effects of the intraband transitions are omitted by setting $\mathbf{q} \rightarrow \mathbf{0}$ in the optical model, the spatially dispersive nature of the intraband conductivity is taken into account in the semiclassical model through the semiclassical Boltzmann equation. This model can accurately describe the non-local effect up to wavenumbers $q = \sqrt{q_x^2 + q_y^2}$ that are smaller than about 0.5 nm^{-1} within the parameter values that is used in our study [87, 124]. Moreover, the semiclassical model is consistent with the anisotropic Drude model in the sense that its leading terms coincide with Eq. (3.8), when expanded in $\mathbf{q}$. On the other hand, within the semiclassical model the interband transitions are taken into account through the same linear approximation that is introduced in Eq. (3.10).

3.5.1 Semiclassical model for intraband polarization function

Considering the electron doped phosphorene with equilibrium doping density n , the electron dispersion relation of the conduction band in the effective mass approximation for a Bloch wave with the momentum vector $\mathbf{p} = p_x \hat{\mathbf{x}} + p_y \hat{\mathbf{y}}$ is given by

$$\varepsilon_{\mathbf{p}} = \frac{p_x^2}{2m_x} + \frac{p_y^2}{2m_y}. \quad (3.17)$$

It is shown that the polarization function $\chi(q_x, q_y, \omega)$ of an anisotropic 2DEG described by Eq. (3.17) might be expressed as [114, 133]

$$\chi(q_x, q_y, \omega) = \chi_{\text{iso}}(Q, \omega), \quad (3.18)$$

where $\chi_{\text{iso}}(Q, \omega)$ is the polarization function of an isotropic 2DEG that possess the same density, with the electron effective mass given by $m_d = \sqrt{m_x m_y}$ that is known as the *density of state effective mass*. Moreover, Q is the wavenumber of the equivalent isotropic 2DEG that is related to the wavenumber components of the anisotropic 2DEG through

$$Q(q_x, q_y) = \left[q_x^2 \sqrt{\frac{m_y}{m_x}} + q_y^2 \sqrt{\frac{m_x}{m_y}} \right]^{\frac{1}{2}}. \quad (3.19)$$

The polarization function $\chi_{\text{iso}}(Q, \omega)$ of an isotropic 2DEG, can be obtained from the Boltzmann equation within a well established approximation that is commonly known as

the relaxation time approximation as [124, 134]

$$\chi_{\text{iso}}(Q, \omega) = \chi_s^0 \int_0^\infty d\varepsilon f_0'(\varepsilon) \int_0^{2\pi} d\vartheta \frac{v_\varepsilon Q \cos \vartheta}{\omega + i\gamma - v_\varepsilon Q \cos \vartheta}. \quad (3.20)$$

where $\chi_s^0 = m_d/(\pi\hbar^2)$ is the static polarizability at zero temperature, $v_\varepsilon = \sqrt{2\varepsilon/m_d}$ is the speed of an electron with the energy ε relative to the band bottom, and $f_0(\varepsilon)$ is the Fermi-Dirac distribution

$$f_0(\varepsilon) = \frac{1}{1 + \exp\left(\frac{\varepsilon - \varepsilon_F^{\text{iso}}}{k_B T}\right)}, \quad (3.21)$$

in which $\varepsilon_F^{\text{iso}}$ is the Fermi energy of the isotropic 2DEG and k_B is the Boltzmann constant. Using the complex analysis integration techniques based on the Cauchy's residue theorem, the integration over ϑ in Eq. (3.20) can be computed analytically [134]. In particular, in the limit of zero temperature, the Fermi-Dirac distribution $f_0(\varepsilon)$ is given by

$$\lim_{T \rightarrow 0} f_0(\varepsilon) = H(\varepsilon_F^{\text{iso}} - \varepsilon), \quad (3.22)$$

where $H(\cdot)$ is the Heaviside unit step function, which means in this limit $f_0'(\varepsilon)$ is related to the Dirac delta function

$$f_0'(\varepsilon) = -\delta(\varepsilon_F^{\text{iso}} - \varepsilon). \quad (3.23)$$

Therefore, in the zero temperature limit, the integration over energy ε in Eq. (3.20) can also be performed analytically using the sifting property of the Dirac delta function that is equivalent to substitute the Fermi speed of the equivalent isotropic 2DEG, v_F^{iso} , into the analytic expression for the integral over ϑ . Note that $v_F^{\text{iso}} = \hbar k_F^{\text{iso}}/m_d$, with $k_F^{\text{iso}} = \sqrt{2\pi n}$ being its Fermi wavenumber. Moreover, the relaxation time approximation violates the crucial principle of the continuity of charges, hence, for more accurate results one can impose this condition during the model development, which may be implemented through the Mermin's procedure [135, 136]. The Mermin corrected expression for the polarization function is given by

$$\chi(Q, \omega) = \frac{\chi_{\text{iso}}(Q, \omega)}{1 - \frac{i\gamma}{\omega + i\gamma} \left(1 - \frac{\chi_{\text{iso}}(Q, \omega)}{\chi_s}\right)}, \quad (3.24)$$

where χ_s is the static polarization function that is the limiting case of the Eq. (3.20) when both ω and γ approach zero. Note that the semiclassical intraband polarization function Eq. (3.24) can be equivalently obtained via implementing the Bhatnagar-Gross-Krook [137] procedure in the process of solving the linearized Boltzmann equation for an anisotropic 2DEG in the RTA [124], which is given in full detail in the appendix B.2.

Finally, we find the following expressions for the real and imaginary parts of the semiclassical model for the intraband polarization function, that are obtained from Eqs. (3.18) and (3.20) [134]

$$\Re\{\chi_{\text{intra}}\} = \chi_s^0 \left[1 - |\omega| \frac{H\left(\omega^2 - (v_F^{\text{iso}} Q)^2\right)}{\sqrt{\omega^2 - (v_F^{\text{iso}} Q)^2}} \right], \quad (3.25)$$

$$\Im\{\chi_{\text{intra}}\} = \chi_s^0 \omega \frac{H\left((v_F^{\text{iso}} Q)^2 - \omega^2\right)}{\sqrt{(v_F^{\text{iso}} Q)^2 - \omega^2}}, \quad (3.26)$$

where we assumed that there is no damping ($\gamma = 0$), the temperature is set to zero, and Q is given by Eq. (3.19). This semiclassical model which is described in the above equations is supposed to work well for the small wavenumbers Q , which do not exceed k_F^{iso} [124]. Note that, according to the Eq. (2.63), the exponential factor e^{-2qz_0} in the integrand that naturally emerges in the expressions related to the dissipative forces, reduces the effective integration domain to the small wavenumbers $q \lesssim \frac{1}{2z_0}$. Hence, for an anisotropic 2DEG such as phosphorene with $m_y > m_x$ and doping density $n = 10^{13} \text{ cm}^{-2}$, we have $\max\{Q(q_x, q_y)\} = q \left(\frac{m_y}{m_x}\right)^{1/4}$ that means for distances $z_0 \gg \frac{1}{2k_F^{\text{iso}}} \left(\frac{m_y}{m_x}\right)^{1/4} \approx 1 \text{ nm}^{-1}$ the semiclassical model is expected to perform very well to provide the stopping force $F_{\parallel}$. Moreover, if we expand Eq. (3.24) in $v_F^{\text{iso}} Q/|\omega|$ to the leading order, the result is

$$\chi(Q, \omega) \sim -\frac{\chi_s^0}{2} \frac{(v_F^{\text{iso}} Q)^2}{\omega(\omega + i\gamma)}, \quad (3.27)$$

that is equivalent to the Drude contribution Eq. (3.8) in our optical model upon using Eq. (3.19) with the isotropic damping, $\gamma_x = \gamma_y = \gamma$. On the other hand, in the limit of vanishing frequency $\omega \rightarrow 0$ with finite damping, from Eq. (3.24) we find that

$$\lim_{\omega \rightarrow 0} \chi(Q, \omega) = \chi_s^0 = \frac{q_s}{2\pi e^2}, \quad (3.28)$$

that describes the static screening of a 2DEG, with $q_s = 2m_d e^2 / \hbar^2$ being the Thomas-Fermi inverse screening length at zero temperature.

3.5.2 Phosphorene effective dielectric function within the semiclassical model

Given the semiclassical representation of the real and imaginary parts of the phosphorene intraband polarization function in Eqs. (3.25) and (3.26), and using the linear approxi-

mation introduced in Eq. (3.15) for the interband contributions, we propose the following model for the effective dielectric function of doped phosphorene

$$\begin{aligned} \epsilon_{2D}(\mathbf{q}, \omega) = & \epsilon_{bg} + \frac{2\pi}{q} (\alpha_x q_x^2 + \alpha_y q_y^2) \\ & + \frac{2\pi e^2}{q} \chi_s^0 \left[1 - |\omega| \frac{H(\omega^2 - (v_F^{\text{iso}} Q)^2)}{\sqrt{\omega^2 - (v_F^{\text{iso}} Q)^2}} + i\omega \frac{H((v_F^{\text{iso}} Q)^2 - \omega^2)}{\sqrt{(v_F^{\text{iso}} Q)^2 - \omega^2}} \right], \end{aligned} \quad (3.29)$$

that we call the semiclassical model. This model is expected to provide a good account of the dynamic response of doped phosphorene in the THz-MIR frequency range that includes the spatially dispersive phenomena, which were omitted in the optical model.

3.6 Plasmon dispersion

In this section, we first briefly review the dispersion relations for an isotropic 2DEG based on the hydrodynamic model of Fetter [58], which is followed by a presentation of several models that are suitable for describing the plasmon dispersion in an anisotropic 2DEG in both local and non-local regimes.

3.6.1 Hydrodynamic model of isotropic 2DEG

From the hydrodynamic conductivity model of a previous section, we can write the corresponding effective 2D dielectric permittivity of a free-standing isotropic 2DEG in the limit of zero damping as

$$\epsilon_{\text{hydro}}(q, \omega) = 1 - \frac{2Dq}{\omega^2 - s^2 q^2}.$$

Solving the equation $\epsilon_{\text{hydro}}(q, \omega) = 0$ gives the plasmon frequency as

$$\omega_p^{\text{hydro}}(q) = \sqrt{2Dq + s^2 q^2},$$

which exhibits acoustic dispersion $\omega \approx sq$ for increasing wavenumbers, where $s = v_F/\sqrt{2}$.

On the other hand, taking the long-wavelength limit of the plasmon frequency $\omega_p^{\text{hydro}}(q)$ corresponds to the isotropic Drude model, which gives $\omega_p^{\text{Drude}}(q) = \sqrt{2Dq}$ when $q \rightarrow 0$. The resulting plasmon dispersion, $\omega = \omega_p^{\text{Drude}}(q)$, exhibits the same characteristic square-root dependence on the wavenumber as the waves on the surface of a deep calm water,

$\omega = \sqrt{gq}$, with the role of the acceleration due to gravity, g , taken by the Drude weight as $g = 2D$. This close analogy between the isotropic 2DEG and the water waves illustrates why Kelvin's ship wake analysis will be found relevant in a subsequent chapter devoted to the discussion of the wake effect in a 2DEG. In particular, it will be shown there that dramatic changes can arise in the classic Kelvin's wake pattern due to modifications in the plasmon frequency that are suitable for an anisotropic 2DEG.

3.6.2 Anisotropic 2DEG

In this subsection, a comparison is made between the models for the effective dielectric function of the doped phosphorene introduced in the previous sections. The non-local effects are discussed for plasmon dispersion of free-standing phosphorene ($\epsilon_{\text{bg}} = 1$) using three different models in the limit of zero damping and zero temperature.

1. Anisotropic *Drude model* for intraband electron transitions given in Eq. (3.8) that results in the effective dielectric function given by Eq. (3.16) with $\alpha_x = \alpha_y = 0$.
2. Full *optical model* that is the anisotropic Drude model for the intraband transitions, plus the linear approximation for the interband correction, given in Eq. (3.16).
3. The *semiclassical model* that consists of the semiclassical description of the intraband electron transitions and the linear approximation for the interband correction, given in Eq. (3.29).

Consider a sample of free-standing doped phosphorene with negligible damping. In this limit the effective dielectric functions within the anisotropic Drude model, optical model, and the semiclassical model can be written as

$$\epsilon_{2\text{D}}^{\text{Drude}}(\mathbf{q}, \omega) = 1 - \frac{2}{q\omega} \left(\frac{D_x q_x^2}{\omega} + \frac{D_y q_y^2}{\omega} \right), \quad (3.30)$$

$$\epsilon_{2\text{D}}^{\text{optical}}(\mathbf{q}, \omega) = 1 + \frac{A}{q} - \frac{2}{q\omega} \left(\frac{D_x q_x^2}{\omega} + \frac{D_y q_y^2}{\omega} \right), \quad (3.31)$$

and

$$\epsilon_{2\text{D}}^{\text{semi}}(q_x, q_y, \omega) = 1 + \frac{A}{q} + \frac{q_s}{q} \left[1 - \frac{|\omega|}{\sqrt{\omega^2 - (v_{\text{F}}^{\text{iso}} Q)^2}} \right], \quad (3.32)$$

respectively. Note that for the semiclassical model Eq. (3.32) it is assumed that $|\omega| > v_F^{\text{iso}}Q$, where Q is a function of q_x and q_y that is given in Eq. (3.19), q_s is equal to $2\pi e^2\chi_s^0$ based on Eq. (3.28), and A stands for the function

$$A(q_x, q_y) = 2\pi (\alpha_x q_x^2 + \alpha_y q_y^2), \quad (3.33)$$

which describes a contribution from the static polarizabilities due to the interband transitions.

Plasmon dispersion relations of these models are defined as the roots of the equation $\epsilon_{2D}(q_x, q_y, \omega) = 0$, when solved for each model to find ω as a function of (q_x, q_y) . Demanding Eqs. (3.30), (3.31), and (3.32) to be identically zero, yields

$$\omega_p^{\text{Drude}}(q_x, q_y) = \sqrt{\frac{2}{q} (D_x q_x^2 + D_y q_y^2)}, \quad (3.34)$$

$$\omega_p^{\text{optical}}(q_x, q_y) = \sqrt{2 \frac{D_x q_x^2 + D_y q_y^2}{q + a_x q_x^2 + a_y q_y^2}}, \quad (3.35)$$

and

$$\omega_p^{\text{semi}}(q_x, q_y) = \frac{v_F^{\text{iso}}Q(q_s + q + A)}{\sqrt{(q + A)(2q_s + q + A)}}, \quad (3.36)$$

for the plasmon dispersion relation of the anisotropic Drude, optical, and semiclassical models, respectively. An important prediction of the semiclassical model is that the non-local effects give rise to a region in the (q_x, q_y, ω) space characterized by a continuum of the intraband electron-hole (e-h) excitations that occur in the vanishing damping limit at frequencies $|\omega| < v_F^{\text{iso}}Q(q_x, q_y)$, see (3.26). This region can have significant effects on the plasmon dispersion curves in free-standing phosphorene, which can only occur at frequencies $\omega > v_F^{\text{iso}}Q(q_x, q_y)$, see (3.25), and are prevented from crossing the boundary $\omega = v_F^{\text{iso}}Q(q_x, q_y)$ as the wavenumbers increase. Notice that the semiclassical model Eq. (3.36) is generalization of the plasmon dispersion given in Eq. (3.35) that is based on the optical model.

Plasmon dispersion curves along the AC and the ZZ directions for these three models are shown in Fig. 3.2. In this figure we used an extended range of wavenumbers, which exceeds the domain of applicability of the semiclassical model. We see that the effect of interband transitions on the Drude model (green curves) is slowing down their increase (already for $q \gtrsim 0.1 \text{ nm}^{-1}$) and leveling them off, which is the full optical model (red curves). But the non-local effects from the semiclassical model, prevent the leveling off (already for $q \gtrsim 0.3 \text{ nm}^{-1}$) of the dispersion in the optical model and make them softly approach the edges of the e-h continuum (orange curves) at large wavenumbers.

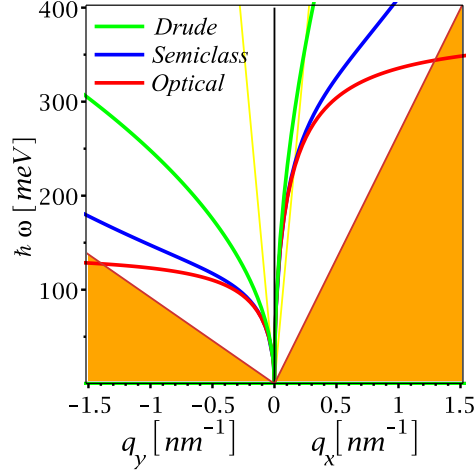


Figure 3.2: Cross sections of the dispersion surface $\omega = \omega_p(q_x, q_y)$ with the planes $q_x = 0$ (left side, $q_y < 0$) and $q_y = 0$ (right side, $q_x > 0$) for plasmon modes in free-standing phosphorene using: the Drude model (green curves), optical model (red curves), and the semiclassical model (blue curves). The two orange shaded regions indicate the continuum of e-h intraband excitations.

3.7 Quantum mechanical model

In this section we introduce a model for the polarization function of an isotropic 2DEG that stems from the fundamental assumptions of the quantum mechanics to describe the system [90, 138]. This model relies on assuming the Heisenberg equation as the main equation describing the dynamics of a non interacting 2DEG within the linear response theory. While the resulting expression for the polarization function is usually called the Lindhard function, it is equivalent to the so-called random phase approximation of the many-body perturbation theory, which is known to give an accurate description of the plasmon dispersion in the context of quantum mechanical treatment of the electron gas [138]. In the zero temperature limit, the following closed form expression for the Lindhard function $\chi = \chi_1 + i\chi_2$ could be obtained for an isotropic 2DEG with the effective electron mass m [90, 138]

$$\begin{aligned}\chi_1 &= \frac{m}{\pi\hbar^2} \left[1 + \frac{k_F}{q} \left(\text{sgn}(v_-)H(v_-^2 - 1)\sqrt{v_-^2 - 1} - \text{sgn}(v_+)H(v_+^2 - 1)\sqrt{v_+^2 - 1} \right) \right] \\ \chi_2 &= \frac{m}{\pi\hbar^2} \frac{k_F}{q} \left[H(1 - v_-^2)\sqrt{1 - v_-^2} - H(1 - v_+^2)\sqrt{1 - v_+^2} \right],\end{aligned}\tag{3.37}$$

where $v_{\pm} = \frac{\omega}{qv_F} \pm \frac{q}{2k_F}$, $\text{sgn}(\cdot)$ is the sign function, and $H(z)$ is the ordinary Heaviside step function. In the limit of vanishingly small frequency, where this model is used in this thesis, $|v_{\pm}| \rightarrow \frac{q}{2k_F}$, so that Eqs. (3.37) reduce to

$$\chi_1 = \frac{m}{\pi\hbar^2} \left[1 - \frac{2k_F}{q} \sqrt{\left(\frac{q}{2k_F}\right)^2 - 1} + \mathcal{O}(\omega^2) \right] H\left(\left(\frac{q}{2k_F}\right)^2 - 1\right) \quad (3.38)$$

$$\chi_2 = \frac{m}{\pi\hbar^2} \left[\frac{1}{\sqrt{1 - \left(\frac{q}{2k_F}\right)^2}} \left(\frac{\omega}{qv_F}\right) + \mathcal{O}(\omega^3) \right] H\left(1 - \left(\frac{q}{2k_F}\right)^2\right). \quad (3.39)$$

Note that χ_1 and χ_2 in Eqs. (3.38) and (3.39) are expanded as Taylor series to leading order in $\frac{\omega}{qv_F}$, which is sufficiently accurate for our purposes. Therefore, in the limit of low frequency, we find the following approximation for the quantum mechanical polarization function for an isotropic 2DEG, after some algebraic manipulations on Eqs. (3.38) and (3.39)

$$\chi_{\text{iso}}^{\text{quantum}}(q, \omega) = \frac{m}{\pi\hbar^2} \left[1 - \frac{1}{q} \sqrt{q^2 - (2k_F)^2} H(q - 2k_F) \right] + i\omega \frac{2m^2}{\pi\hbar^3 q} \frac{H(2k_F - q)}{\sqrt{(2k_F)^2 - q^2}}. \quad (3.40)$$

This polarization function can be used to describe the intraband electronic transitions in an anisotropic 2DEG through the transformation $q \rightarrow Q(q_x, q_y)$ that is introduced in Eq. (3.19) [91, 114, 133]. Ultimately, our quantum mechanical model for the polarization function consists of

$$\chi^{\text{quantum}}(q_x, q_y, \omega) = \chi_{\text{iso}}^{\text{quantum}}(Q(q_x, q_y), \omega), \quad (3.41)$$

where

$$Q(q_x, q_y) = \left[q_x^2 \sqrt{\frac{m_y}{m_x}} + q_y^2 \sqrt{\frac{m_x}{m_y}} \right]^{\frac{1}{2}}, \quad (3.42)$$

and $m = m_d$, with $m_d = \sqrt{m_x m_y}$ being the density of states effective mass. This polarization function will be amended by a contribution from the interband electronic transitions discussed in previous sections and used in chapter 5 to compute the DC mobility tensor of doped phosphorene, using the energy loss method (ELM).

3.8 Dielectric permittivity of the supporting material

In this thesis, we consider SiO₂ as the substrate of our standard system, which exhibits two prominent transverse optical (TO) phonon modes at frequencies ω_{TO1} and ω_{TO2} and its bulk dielectric permittivity in the THz-MIR range of frequencies can be modeled using the Lorentz model by [87, 122]

$$\epsilon_s(\omega) = \epsilon_\infty + (\epsilon_0 - \epsilon_i) \frac{\omega_{\text{TO1}}^2}{\omega_{\text{TO1}}^2 - \omega(\omega + i\gamma_{\text{TO1}})} + (\epsilon_i - \epsilon_\infty) \frac{\omega_{\text{TO2}}^2}{\omega_{\text{TO2}}^2 - \omega(\omega + i\gamma_{\text{TO2}})}, \quad (3.43)$$

where ϵ_0 , ϵ_i and ϵ_∞ are the static, intermediate and infinite-frequency dielectric constants, and γ_{TO1} and γ_{TO2} the corresponding damping rates.

Chapter 4

Forces and wakes induced by a point charge moving above phosphorene

4.1 Introduction

In this chapter, we consider a charged particle moving parallel to the surface of doped phosphorene, and analyze the induced electric forces acting on that particle. We use the standard dielectric response theory alongside the optical and semiclassical models introduced in the preceding chapter to model phosphorene and its remarkable anisotropic nature. Computations reveal how a topological transition between elliptic and hyperbolic isofrequency curves for plasmon modes in the THz-MIR frequency range affects the forces acting in the moving particle. Moreover, we consider the effects of plasmon hybridization with optical phonons in the SiO_2 substrate underneath phosphorene through a simple description of the SiO_2 dielectric function as introduced earlier.

It is shown that the force on the incident charge consists of three components that are quite sensitive to the velocity, both in magnitude and direction relative to the principal axes of the conductivity tensor. These force components are the longitudinal stopping force, the transverse stopping force, and the dynamic image force. While the existence of the transverse stopping force is a consequence of the anisotropic response of phosphorene, it is surprising that its magnitude is comparable to that of the longitudinal force. It acts parallel to the target surface and perpendicular to the trajectory of the particle, making it experimentally observable. Furthermore, our anisotropic model for the phosphorene conductivity predicts the existence of a repulsive dynamic image force when the particle

moves along the armchair direction of freestanding phosphorene at certain range of speeds. This unexpected repulsive image force disappears in the presence of the substrate.

Additionally, we study the total electric potential, a standard output of the dielectric response theory, in detail, revealing a rich variety of plasmonic wake patterns. Using the method of stationary phase, we discover surprising analogies with Kelvin's ship wakes in the presence of shear current and atmospheric wakes regarding the asymmetry of the wake and the onset of a Mach-like wake due to non-local effects in the dielectric response.

The materials presented in this chapter are a representation of our research papers, published in Refs. [87, 95].

4.2 Forces on the moving charge

The underlying system in this study is the standard system consisting of a semi-infinite substrate occupying the region $z \leq -d$ and an anisotropic 2DEG that represents the phosphorene, situated in the plane $z = 0$. A perspective of this system is shown in Fig. 2.1, and the Green's functions of its electric potential are given in Eqs. (2.40) and (2.39). Assuming that a charged particle is moving parallel to the surface of the 2DEG at the height $z = z_0$, the electric force acting on this particle could be obtained from the dielectric response theory, as performed in Sec. (2.6.2) and expressed in Eq. (2.58). Moreover, the total electric force acting on the charged particle could be decomposed as $\mathbf{F} = \mathbf{F}_{\parallel} + \hat{\mathbf{z}}F_i$, with components called the stopping force ($\mathbf{F}_{\parallel}$) and the dynamic image force (F_i), given by

$$\mathbf{F}_{\parallel} = \frac{(Ze)^2}{2\pi} \int d^2\mathbf{q} e^{-2qz_0} \hat{\mathbf{q}} \Im \left\{ \frac{1}{\epsilon_{2D}(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})} \right\}, \quad (4.1)$$

and

$$F_i = \frac{(Ze)^2}{2\pi} \int d^2\mathbf{q} e^{-2qz_0} \Re \left\{ \frac{1}{\epsilon_{2D}(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})} - 1 \right\}, \quad (4.2)$$

which were derived before. Note that Eq. (4.2) is equivalent with the second term in the right hand side of the Eq. (2.58), after accounting for the Hermitian parity of the Green's function for the electric potential.

Note that in Eqs. (4.1) and (4.2), the substitution $\omega = \mathbf{q} \cdot \mathbf{v}$ that is a manifestation of the Galilean kinematic resonance between the layer's and particle's frames of references, results in the directionality effects of the forces. On the other hand, the stopping force can have a component that is perpendicular to the particle's trajectory, motivating the decomposition $\mathbf{F}_{\parallel} = \hat{\mathbf{v}}F_l + (\hat{\mathbf{z}} \times \hat{\mathbf{v}})F_t$ of the stopping force to its longitudinal, F_l , and

transverse, F_t , components. Note that $\hat{\mathbf{v}} = \mathbf{v}/v$, and $\hat{\mathbf{z}}$ is the unit vector in the direction of the z axis. Moreover, switching to the polar coordinate system with $\hat{\mathbf{q}} = \hat{\mathbf{v}} \cos(\theta - \theta_0) + (\hat{\mathbf{z}} \times \hat{\mathbf{v}}) \sin(\theta - \theta_0)$, in which θ_0 is the angle between the velocity $\mathbf{v}$ and the phosphorene's AC principal axis, allows us to describe the force components in terms of the integrals in the polar representation of the wave vector $\mathbf{q} = (q, \theta)$ as

$$F_l = \frac{(Ze)^2}{2\pi} \int_0^\infty dq q e^{-2qz_0} \int_0^{2\pi} d\theta \cos(\theta - \theta_0) \Im \left[\frac{1}{\epsilon_{2D}(q, \theta, \mathbf{q} \cdot \mathbf{v})} \right], \quad (4.3)$$

$$F_t = \frac{(Ze)^2}{2\pi} \int_0^\infty dq q e^{-2qz_0} \int_0^{2\pi} d\theta \sin(\theta - \theta_0) \Im \left[\frac{1}{\epsilon_{2D}(q, \theta, \mathbf{q} \cdot \mathbf{v})} \right], \quad (4.4)$$

and

$$F_i = \frac{(Ze)^2}{2\pi} \int_0^\infty dq q e^{-2qz_0} \int_0^{2\pi} d\theta \Re \left[\frac{1}{\epsilon_{2D}(q, \theta, \mathbf{q} \cdot \mathbf{v})} - 1 \right]. \quad (4.5)$$

In the polar representations of the forces, $\epsilon_{2D}(q, \theta, \mathbf{q} \cdot \mathbf{v})$ is obtained by the substitutions $q_x = q \cos \theta$, $q_y = q \sin \theta$, and $\omega = \mathbf{q} \cdot \mathbf{v} = qv \cos(\theta - \theta_0)$ into $\epsilon_{2D}(\mathbf{q}, \omega)$. Moreover, the effective dielectric function $\epsilon_{2D}(\mathbf{q}, \omega)$ will be chosen from one of the anisotropic Drude model, optical model, or the semiclassical model given in Eqs. (3.30), (3.31), and (3.32), respectively. Note that the angle θ is also measured with respect to the AC direction of phosphorene, which is adopted for the x axis.

Additionally, Equations (4.3), (4.4), and (4.5) imply that the longitudinal stopping force and the image force are even functions of θ_0 with respect to the principal directions $\theta_0 = 0$ (AC) and $\theta_0 = \pi/2$ (ZZ), while the transverse stopping force is an odd function with respect to the principal directions. Moreover, these forces are all periodic functions of θ_0 with the period π which means that the force components do not change upon reversing the direction of motion.

In order to complete the dielectric models, we adopted the values of the parameters from Ref. [1], which are obtained from *ab initio* calculations. Accordingly, for an electron doped phosphorene with $n = 10^{13} \text{ cm}^{-2}$, the Fermi energy is 52.2 meV with respect to the bottom of the lowest-lying conduction band. At this doping density, the slopes α_j in Eq. (3.10) are given by $\alpha_x = 0.572 \text{ nm}$ and $\alpha_y = 0.488 \text{ nm}$, whereas the effective masses of the electrons in directions x and y are given by $m_x = 0.133m_0$ and $m_y = 1.119m_0$ with m_0 being the bare electron mass. On the other hand, according to Ref. [3], an approximation for the carrier mobilities due to electron scattering on charged impurities

is given by $\mu_x = 700 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ and $\mu_y = 113 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, which could be translated to $\hbar\gamma_x = 12.4 \text{ meV}$ and $\hbar\gamma_y = 9.2 \text{ meV}$. The numerical values for the phosphorene parameters are summarised in Table. 4.1. These numeric values show that the anisotropies in the static polarizabilities α_j and the damping rates γ_j are not significant, and our computations of the force components confirm that the anisotropy in the effective masses is the main source of the phosphorene anisotropic behavior.

Parameter	Value
E_F	52.2 meV
α_x	0.572 nm
α_y	0.488 nm
m_x	$0.133m_0$
m_y	$1.119m_0$
$\hbar\gamma_x$	12.4 meV
$\hbar\gamma_y$	9.2 meV

Table 4.1: Phosphorene parameters [1, 3].

In this research, SiO_2 with two prominent transverse optical (TO) phonon modes is served as the substrate. Silicon dioxide is a technology compatible and feasible supporting material that its dielectric permittivity could be adequately approximated by a Lorentz model, given in Eq. (3.43). In order to complete this dielectric permittivity model, parameters are taken from Ref. [139]. Accordingly, $\epsilon_0 = 3.9$, $\epsilon_i = 3.05$, and $\epsilon_\infty = 2.5$; $\hbar\omega_{\text{TO1}} = 55.6 \text{ meV}$ and $\hbar\omega_{\text{TO2}} = 138.1 \text{ meV}$; $\hbar\gamma_{\text{TO1}} = 5.37 \text{ meV}$ and $\hbar\gamma_{\text{TO2}} = 8.95 \text{ meV}$. These values for the parameters give rise to the two Reststrahlen bands, occupying the frequency intervals $\omega_{\text{TO1}} < \omega < \omega_{\text{SO1}}$ and $\omega_{\text{TO2}} < \omega < \omega_{\text{SO2}}$, with $\hbar\omega_{\text{SO1}} = 60.9 \text{ meV}$ and $\hbar\omega_{\text{SO2}} = 146.5 \text{ meV}$ corresponding to the frequencies of the related Fuchs-Kliwer, or surface optical phonons in the SiO_2 substrate. Reststrahlen bands are defined by a range of frequencies where the real part of the dielectric permittivity in Eq. (3.43) is negative. Finally, for the gap size between the substrate and the 2DEG, we take $d = 0.4 \text{ nm}$ as a representative value for a van der Waals distance.

In this study the speed of the incident charged particle is normalized by the Bohr velocity $v_B = e^2/\hbar \approx c/137$, with c being the speed of light in free space. Hence, the non-dimensionalized speed of the charged particle is given by $\bar{v} = v/v_B$.

4.3 Plasmon dispersion relation

In this section, plasmon dispersion relations, described by the equation $\epsilon_{2D}(q_x, q_y, \omega) = 0$ in the vanishing damping limit are discussed. These analyses are performed using the effective dielectric function models introduced in Eqs. (3.30), (3.31), and (3.32) that are called anisotropic Drude, optical, and semiclassical models, respectively. Moreover, these models are completed with the parameter values given in the previous section. In an effort to understand the importance of the intraband contribution in our conductivity models, we start with a free standing phosphorene ($d \rightarrow \infty$). In this case the optical model gives rise to the open isofrequency curves in the $\mathbf{q} = (q_x, q_y)$ plane in the frequency range $\omega_{h1} < \omega < \omega_{h2}$, with $\hbar\omega_{h1} = \hbar e \sqrt{\frac{n}{m_y \alpha_y}} \approx 142$ meV and $\hbar\omega_{h2} = \hbar e \sqrt{\frac{n}{m_x \alpha_x}} \approx 379$ meV. These open isofrequency curves which follow a hyperbolic geometrical pattern in the $\mathbf{q}$ plane are called the hyperbolic modes. On the other hand, the inclusion of the non-local effects via the semiclassical model, prevents the dispersion curves to cross the continuum of e-h excitations at increasing wave numbers, see Fig. 3.2. This results in the closed isofrequency curves in the same frequency range for which the optical model predicts the open hyperbolic isofrequency curves. Note that for the MIR frequency range and small wave numbers, the semiclassical model exhibits quasihyperbolic isofrequency curves, which are similar to those of the optical model. On the other hand, in the THz range, phosphorene operates in the elliptic regime [38, 39] with closed isofrequency dispersion curves that resemble an elongated numeral eight, making it more appropriate to call them the quasielliptic modes. In the quasielliptic regime, the wave numbers are typically $\ll 1 \text{ nm}^{-1}$ and the interband and spatially dispersive corrections are negligible. In the quasielliptic regime, the effective Dielectric function of the doped phosphorene could be adequately approximated by the anisotropic Drude model.

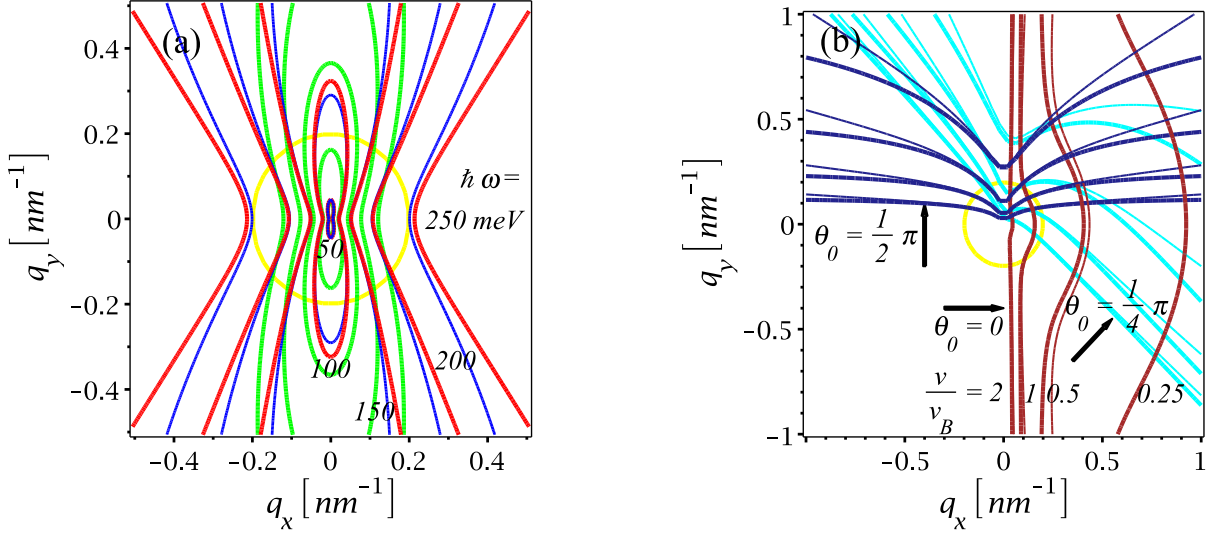
In order to exhibit a transition from the elliptic to hyperbolic regimes in freestanding phosphorene, the isofrequency curves from the three effective dielectric function models are depicted in Fig. 4.1a. These curves are obtained from the dispersion surface $\omega = \omega_p(q_x, q_y)$ for $\hbar\omega = 50, 100, 150, 200$, and 250 meV, based on Eqs. (3.34), (3.35), and (3.36) for the anisotropic Drude (green curves), optical (red curves), and semiclassical (blue curves) models, respectively. One can verify that the two innermost curves for 50 and 150 meV are quasielliptic for all three models. Moreover, while the isofrequency curves for the Drude and semiclassical models remain closed for 150, 200, and 250 meV, the isofrequency curves of the optical model become open and hyperbolic-shaped in this range. In Fig. 4.1a, the circle $q = \frac{1}{2z_0}$ with $z_0 = 2.5 \text{ nm}$ is also shown. Due to the exponential factor $\exp(-2qz_0)$ in the definition of the force components in Eqs. (4.3), (4.4), and (4.5), the main contribution to these forces is related to the region enclosed by this circle. One

notices that the dispersion curves from the optical and semiclassical models are close inside that circle for all frequencies, whereas the Drude model only agrees with them for the curve with $\hbar\omega = 50\text{meV}$.

The investigation into the directionality effects in freestanding phosphorene is driven by the definitions of the longitudinal and transverse stopping forces found in Eqs. (4.3) and (4.4). These forces are expressed as double integrals, which include a common factor, $\Im\{\frac{1}{\epsilon_{2D}(q_x, q_y, \omega)}\}$, that is related to the energy loss function of the system and becomes strongly peaked at $\omega = \omega_p(q_x, q_y)$ when losses are negligible. The value of $\omega_p(q_x, q_y)$ is determined by one of the equations (3.34), (3.35), or (3.36), depending on the model used for $\epsilon_{2D}(q_x, q_y, \omega)$. Additionally, in the integrand of these double integrals, $\omega = \mathbf{q} \cdot \mathbf{v}$ is implied by the kinematic resonance. Consequently, the curves defined by $\omega_p(q_x, q_y) = \mathbf{q} \cdot \mathbf{v} = q_x v \cos \theta_0 + q_y v \sin \theta_0$ and their neighborhoods correspond to the peaks in the integrands of Eqs. (4.3) and (4.4), making these regions in the $\mathbf{q}$ domain the most significant contributors to the stopping force components. Figure (4.1b) shows three sets of these curves for $\theta_0 = 0$ (brown curves), $\pi/4$ (cyan curves), and $\pi/2$ (navy curves), with each set containing four curves for reduced speeds $\bar{v} = 0.25, 0.5, 1$, and 2 . Within each set, the thick curves (same color) represent the optical model, while the thin curves represent the semiclassical model. In each set, the curve with the highest speed ($\bar{v} = 2$) is closest to the origin of the $\mathbf{q}$ plane, whereas the curve with $\bar{v} = 0.25$ for $\theta_0 = 0$ in the semiclassical model is out of the visible range of q_x and q_y in the figure. These curves exhibit a wide variety of shapes in different directions, bending away from the origin for the AC direction ($\theta_0 = 0$) and towards the origin for the ZZ direction ($\theta_0 = \pi/2$). This illustrates the complex interplay between the parameters θ_0 , v , and z_0 , which define the particle's trajectory, in relation to the sampling of different plasmon dispersion relations in phosphorene. The same circle $q = \frac{1}{2z_0}$ is also depicted in Fig. 4.1b, enclosing a region with small wave numbers relevant to the particle distances considered in this study.

On the other hand, the directionality effects in the dynamic image force are influenced by the integrand of Eq. (4.5), which incorporates the factor $\Re\{1/\epsilon_{2D}(q_x, q_y, \omega) - 1\}$. This factor is a Kramers-Kronig transformation of the system's energy loss function and can change its sign rapidly near the dispersion surfaces. Consequently, in systems with anisotropic materials, the dynamic image force may change its sign, in contrast to the stopping force components, which are strictly negative due to $\Im\{1/\epsilon_{2D}(q_x, q_y, \omega)\} < 0$.

In the presence of the substrate, the effective dielectric functions for both the optical and semiclassical models can be derived from Eqs. (3.31) and (3.32) by replacing the first term (i.e. 1) with $\epsilon_{bg}(q, \omega)$ from Eq. (2.41). This substitution results in the hybridization between the plasmons of phosphorene and the optical phonons of SiO_2 , leading to three distinct plasmon dispersion surfaces $\omega = \omega_{pj}(q_x, q_y)$ for $j = 1, 2$, and 3 , where $\omega_{p1} < \omega_{p2} <$



(a) The isofrequency curves $\hbar\omega_p(q_x, q_y) = 50, 100, 150, 200,$ and 250 meV for plasmon modes in freestanding phosphorene. Results from the anisotropic Drude, optical, and semiclassical models are shown in green, red, and blue, respectively. The circle $q = 1/(2z_0)$ is also shown with $z_0 = 2.5$ nm in yellow.

(b) Curves defined by $\omega_p(q_x, q_y) = q_x v \cos \theta_0 + q_y v \sin \theta_0$, determining the wave vectors with highest contribution in the stopping force components for a particle moving above freestanding phosphorene. Thick and thin curves represent results from the optical and semiclassical models, respectively. Results for $\theta_0 = 0, \pi/4,$ and $\pi/2$ are shown in brown, cyan, and navy, respectively. In each set, curves are plotted for the reduced speeds $\bar{v} = v/v_B = 0.25, 0.5, 1,$ and 2 (the curves with $\bar{v} = 2$ pass the closest to the coordinate origin for all three directions). The circle $q = 1/(2z_0)$ is also shown with $z_0 = 2.5$ nm in yellow.

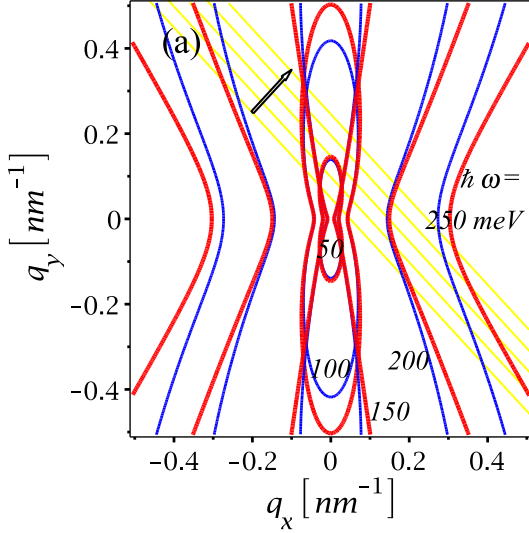
Figure 4.1: Isofrequency and kinetic resonance curves for freestanding phosphorene.

ω_{p3} for all wave vectors, see Fig. 4.3 below. To investigate the impact of plasmon-phonon hybridization, a more realistic value of the gap size of $d = 0.4$ nm is used. The isofrequency curves from the optical and semiclassical models are depicted in Fig. 4.2a for the same set of ω values as in Fig. 4.1a, represented by red and blue curves, respectively. These models show good agreement for wave numbers $q \lesssim 0.2$ nm⁻¹, as previously confirmed in Fig. 4.1a for free-standing phosphorene. In Fig. 4.2a, the yellow lines denote the isofrequency lines

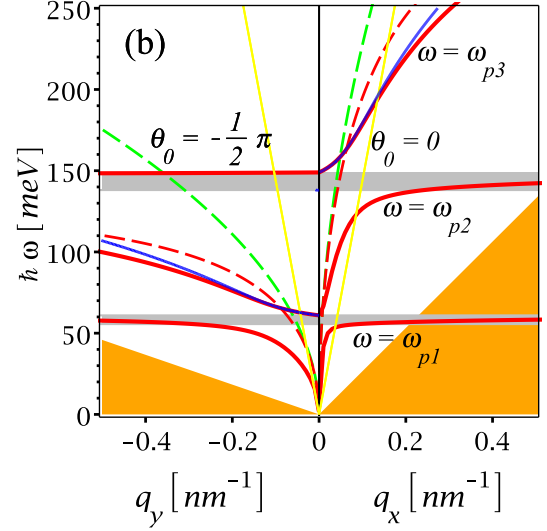
for the plane $\omega = q_x v \cos \theta_0 + q_y v \sin \theta_0$, with the black arrow indicating the direction of increasing frequency. These yellow lines, which represent the condition of kinematic resonance, are plotted for the speed $v = v_B$ and the direction angle $\theta_0 = \pi/4$. This confirms that for supported phosphorene in the THz-MIR frequency range, the optical and semiclassical models yield similar forces, suggesting that non-local effects can be ignored for large distances ($z_0 \gtrsim 1$ nm) and high particle speeds ($v \gtrsim v_B$).

Unlike the free standing phosphorene, the isofrequency curves for the supported phosphorene with $\hbar\omega = 50$ and 100 meV intersect the curve with $\hbar\omega = 150$ meV for both optical and semiclassical models. This is a result of having three different dispersion surfaces $\omega = \omega_{pj}(q_x, q_y)$ for $j = 1, 2$, and 3 in the case of supported phosphorene which exhibit avoided crossings over the entire (q_x, q_y) plane [139]. Using the optical model and assuming that $d = 0$, these three plasmon-phonon hybridized dispersion surfaces are shown in Fig. 4.3 with $\omega = \omega_{p1}(q_x, q_y)$ in cyan (lowest-lying), $\omega = \omega_{p2}(q_x, q_y)$ in brown (intermediate), and $\omega = \omega_{p3}(q_x, q_y)$ in navy (highest-lying). In Fig. 4.3, the plasmon dispersion of the freestanding phosphorene based on the optical model (gray surface), and the surface $\omega = q_x v \cos \theta_0 + q_y v \sin \theta_0$ with $v = 2v_B$ and $\theta_0 = \pi/3$ describing the condition of kinematic resonance, are also plotted. To elaborate on this, the cross sections of the three dispersion surfaces $\omega_{pj}(q_x, q_y)$ with the $q_x = 0$ and $q_y = 0$ planes are shown in Fig. 4.2b, in the left and right sides of the panel, respectively. Notice that the red and blue curves are obtained from the optical and semiclassical models, respectively. The two horizontal gray shaded areas represent the Reststrahlen bands of the SiO_2 substrate. One notices that, the ω_{p1} curves start from 0 and gradually enter the lower-lying Reststrahlen band as both $|q_x|$ and $|q_y|$ increase. The ω_{p2} curves start at ω_{SO1} and gradually enter the higher-lying Reststrahlen band as $|q_x|$ increases, and very slowly increases toward that band when $|q_y|$ increases. Finally, the ω_{p3} curves start at ω_{SO2} and increase as $|q_x|$ increases, but stays (almost) constant on the top of the higher-lying Reststrahlen band as $|q_y|$ increases. Moreover, the regions with a continuum of the intraband e-h excitations are shown in Fig. 4.2b with the orange shaded areas. One can see that except for the $\omega_{p2}(0, q_y)$ and $\omega_{p3}(q_x, 0)$ curves, where the phosphorene plasmons do not hybridize strongly with the substrate phonons, optical and semiclassical models are not distinguishable, demonstrating the fact that in the regions of the (q_x, q_y, ω) space with strong plasmon-phonon coupling, non-local effects are comparatively less significant in supported phosphorene.

Furthermore, in Fig. 4.2b, the dispersion curves for the freestanding phosphorene, obtained from the optical model (dashed red curves) and the anisotropic Drude model (dashed green curves) are plotted. Clearly, these dispersion curves are significantly different, demonstrating the important role of the interband transitions for the wave numbers exceeding $q \sim 0.1 \text{ nm}^{-1}$. On the other hand, in the regions with weak plasmon-phonon coupling, i.e.,



(a) The isofrequency curves $\hbar\omega_p(q_x, q_y) = 50, 100, 150, 200,$ and 250 meV for plasmon modes in phosphorene supported by SiO_2 substrate. Results from the optical and semiclassical models are shown red and blue, respectively. The yellow lines represent the isofrequency lines for the plane $\omega = q_x v_B \cos \theta_0 + q_y v_B \sin \theta_0$ with $\theta_0 = \pi/4$ at the order of increasing frequency, as the black arrow shows.



(b) Cross sections of the dispersion surfaces $\omega = \omega_{pj}(q_x, q_y)$ for $j = 1, 2,$ and 3 of the supported phosphorene with the planes $q_x = 0$ (left side ($q_y < 0$)) and $q_y = 0$ (right side ($q_x > 0$)). The red and blue curves are obtained from the optical and semiclassical models, respectively. The dashed green and red curves represent the same cross sections obtained from the dispersion relation $\omega = \omega_p(q_x, q_y)$ of the free phosphorene from the Drude and optical models, respectively. The two grey shaded regions are the Reststrahlen bands of the SiO_2 substrate, while the two orange shaded areas are the electron-hole intraband continuum in phosphorene. Yellow lines show the cross sections for the plane $\omega = q_x v_B \cos \theta_0 + q_y v_B \sin \theta_0$ with $\theta_0 = 0$ and $\theta_0 = -\pi/2$.

Figure 4.2: Isofrequency and dispersion curves for phosphorene supported by SiO_2 .

$\omega_{p2}(0, q_y)$ for increasing $|q_y|$ and $\omega_{p3}(q_x, 0)$ for increasing $|q_x|$, the freestanding dispersion curves within the optical model tend to those of the supported phosphorene. Moreover, the

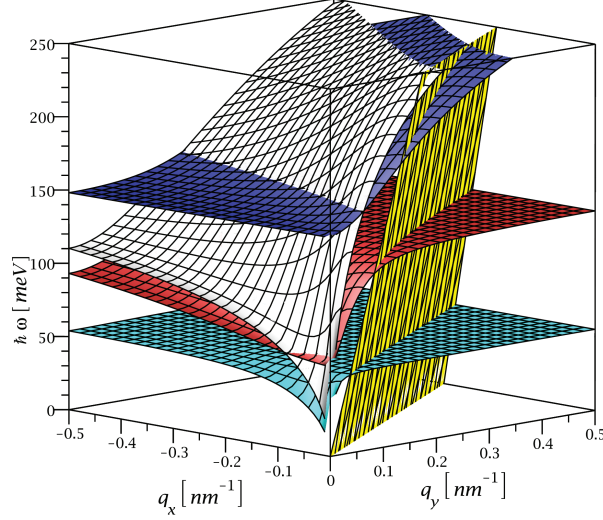


Figure 4.3: Plasmon-phonon hybrid dispersion surfaces $\omega = \omega_{pj}(q_x, q_y)$ for $j = 1$ (cyan surface), 2 (brown surface), and 3 (navy surface). The gray surface is the plasmon dispersion surface of the freestanding phosphorene based on the optical model. The yellow surface is the plane $\omega = q_x v \cos \theta_0 + q_y v \sin \theta_0$ with $v = 2v_B$ and $\theta_0 = \pi/3$, which describes the kinematic resonance between the particle and the system.

yellow lines in the Fig. 4.2b are the cross sections with the plane $\omega = q_x v \cos \theta_0 + q_y v \sin \theta_0$, enforcing the kinematic resonance condition at the speed $v = v_B$ for two directions of motion: $\theta_0 = 0$ (AC) and $\theta_0 = -\pi/2$ (ZZ). Note that the cross sections of these yellow lines with the dispersion curves in different modes correspond to the regions in the (q_x, q_y, ω) space with the dominant contribution to the energy loss of the charged particle, which amount to the longitudinal stopping force. A qualitative analysis of these lines can be used to surmise that the magnitude of the force F_l should be larger in the AC direction, because in this direction, cross sections are happening at the generally larger plasmon energies.

4.4 Calculation of the force components

In the previous section, we established that non-local effects are negligible for sufficiently small wave numbers, which can be ensured by choosing sufficiently large particle distance z_0 . Therefore, we now use our optical model for both free-standing and supported phosphorene to demonstrate the dependence of all three force components, given in Eqs. (4.3), (4.4), and (4.5), on the reduced speed $\bar{v} = v/v_B$ and the direction angle θ_0 for a charged particle moving at a fixed distance of $z_0 = 50$ nm above the phosphorene sheet. This choice of distance warrants the suitability of using our model via the inequality $q \lesssim 10^{-2} \text{ nm}^{-1}$. All forces are presented in a reduced form, normalized by $F_0 = (Ze)^2/(2z_0)^2$, which represents the image force on a static point charge Ze at a distance z_0 from an ideal flat conductor.

In Fig. 4.4, the longitudinal stopping force, F_ℓ , is plotted as a function of the reduced speed $\bar{v}$ and the movement angle θ_0 , showing a typical speed dependence similar to that in isotropic 2D materials like supported graphene [86, 136, 139] that starts at zero when the particle is not moving, reaches a dip at some intermediate speed, and then again its magnitude decreases with increasing velocity. For doped phosphorene, however, this force varies significantly with θ_0 , especially near $\bar{v} \sim 4$, where a prominent dip occurs. The dip is largest for motion along the AC direction ($\theta_0 = 0, \pi$) and smallest along the ZZ direction ($\theta_0 = \pi/2$). This can be explained by the same reasoning as in Refs. [86, 136, 139], which found that increasing the doping density n in the Drude model for graphene, generally increases the stopping force magnitude. In doped phosphorene, the effective electron masses are highly unequal, $m_x < m_y$, that can be translated by referring to the Drude weights in Eq. (3.9) to a larger number of charge carriers in the AC direction than in the ZZ direction, $n_x^e > n_y^e$ [1]. Thus, a charged particle moving in the AC direction above phosphorene interacts with more charge carriers than in the ZZ direction, leading to a larger F_ℓ in the AC direction. Based on our calculations, the ratio of these values is about 2.3 for freestanding phosphorene. With a substrate, the overall magnitude of F_ℓ is reduced compared to freestanding phosphorene, but the anisotropy remains significant near the main dip, with the AC to ZZ ratio reduced to about 1.7. Moreover, at higher speeds, $\bar{v} \gtrsim 10$, the anisotropy effects in F_ℓ are greatly diminished for both free-standing and supported phosphorene.

In Fig. 4.5, we present the transverse stopping force, F_t , for both freestanding and supported phosphorene. While this force is typically zero above isotropic surfaces, its non-zero values demonstrate the anisotropy effects for both free-standing and supported phosphorene. As mentioned before, F_t is an odd function of θ_0 , symmetric about $\theta_0 = 0, \pi$ (AC direction) and $\theta_0 = \pi/2$ (ZZ direction). Thus, F_t is zero when the particle moves

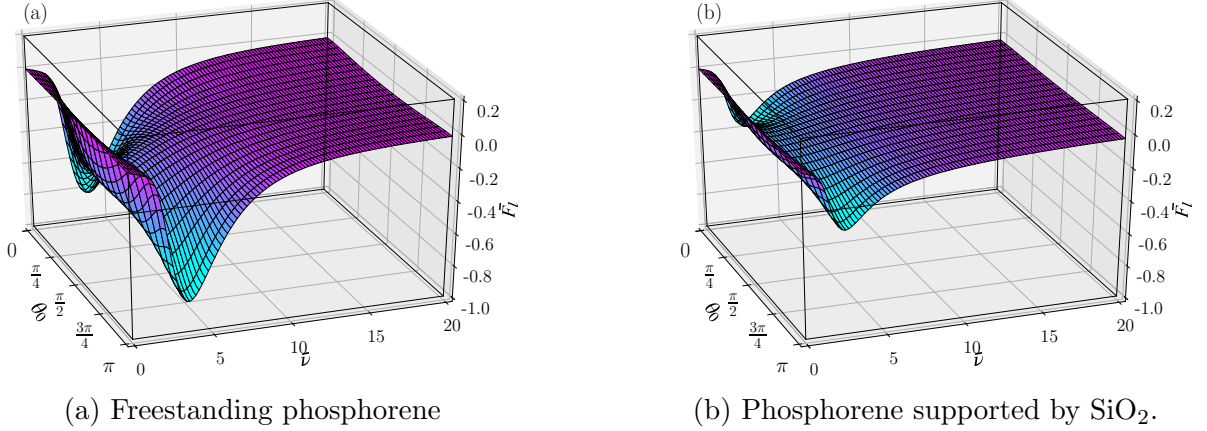


Figure 4.4: Normalized longitudinal stopping force $\bar{F}_l = \frac{F_l}{F_0}$ obtained from the optical model, as a function of the normalized speed $\bar{v} = \frac{v}{v_B}$ and direction angle θ_0 for a particle moving at height $z_0 = 50$ nm.

exactly along the AC and ZZ directions at any speed, but it reaches opposite minimum and maximum values when moving along intermediate directions. For intermediate directions, F_t shows two local peaks at speeds $\bar{v} \sim 2$ and $\bar{v} \sim 7$, forming a minimum/maximum pair for $0 < \theta_0 < \pi/2$ and a maximum/minimum pair for $\pi/2 < \theta_0 < \pi$. The sign of F_t in these intervals suggests that at $\bar{v} \sim 2$, the force directs the particle toward the AC direction, while at $\bar{v} \sim 7$, it directs it toward the ZZ direction. Moreover, the magnitude of F_t near these peaks is comparable to the longitudinal stopping force F_l , and should be observable in HREELS experiments when the target is rotated about the z axis. Similar to F_l , the overall magnitude of F_t decreases at the presence of the substrate, but unlike F_l , the anisotropy effects in F_t persist even at speeds above $\bar{v} \sim 10$. Particularly, Fig. 4.5 shows that F_t changes sign between its extremal points, depending on both θ_0 and $\bar{v}$.

In Fig. 4.6, we show the image force, F_i , for both free-standing and supported phosphorene. Its dependence on the speed $\bar{v}$, for most motion directions is similar to the dynamic image force above an isotropic 2D material [86, 136, 139]. Specifically, F_i starts from a finite value at $v = 0$ that is $F_i = -F_0$, which results from a prevalent role of the Drude model at vanishing speeds, reaches a significant dip at $\bar{v} \sim 2$, and gradually levels off at a constant value for $\bar{v} \gg 1$. However, F_i shows a significant anisotropy, especially near the dip, with the largest magnitude for motion along the AC direction ($\theta_0 = 0, \pi$) and the smallest magnitude along the ZZ direction ($\theta_0 = \pi/2$), with a ratio about 1.7. This anisotropy can be explained with the same reasoning as that of F_ℓ , with more effective

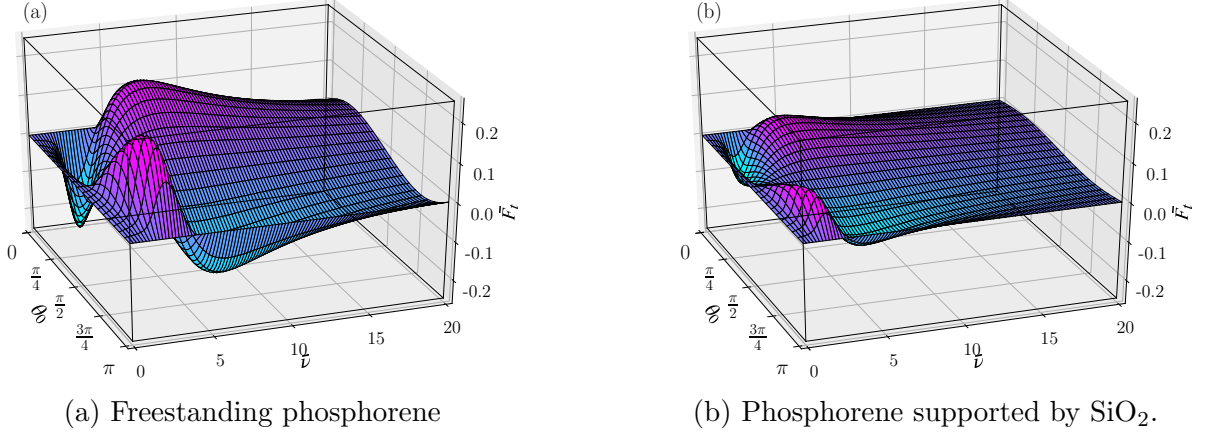


Figure 4.5: Normalized transverse stopping force $\bar{F}_t = \frac{F_t}{F_0}$ obtained from the optical model, as a function of the normalized speed $\bar{v} = \frac{v}{v_B}$ and direction angle θ_0 for a particle moving at height $z_0 = 50$ nm.

charge carriers in the AC direction than in the ZZ direction. Moreover, the presence of a substrate reduces the magnitude of the image force across a broad speed range near the dip and it diminishes the anisotropy effects by decreasing the ratio of magnitudes in the AC and ZZ directions near the dip to about 1.3.

Regarding the image force values when $\bar{v} \gg 1$, Fig. 4.6a shows that F_i approaches 0, while Fig. 4.6b shows that $F_i \approx -0.5 \times F_0$, still exhibiting residual angular dependence due to anisotropy. These limiting values can be assessed using the infinite-frequency limit of the effective dielectric permittivity in Eq. (4.3), which exposes the role of screening by the substrate at high speeds [86]. The most striking effect of our optical model is that the dynamic image force above free-standing phosphorene goes through a broad maximum, becoming positive and thus repulsive for a particle moving close to the AC direction at speeds between $5.7 \lesssim \bar{v} \lesssim 16.5$. This is not the case when the substrate enters the system, where the image force levels off at about $-0.5 \times F_0$ for speeds $\bar{v} \gtrsim 6$, while still showing some angular dependence. This repulsive dynamic image force for free-standing phosphorene cannot be easily explained, given that the image force above an isotropic surface is necessarily negative (attractive), which can be rigorously proved when the factor $\Re\{1/\epsilon_{2D}(\mathbf{q}, \omega) - 1\}$ in Eq. (4.5) is expressed in terms of the energy loss function $\Im\{-1/\epsilon_{2D}(\mathbf{q}, \omega)\} > 0$ using the Kramers-Kronig relations [86]. A further analysis of forces on moving charged particle, including a detailed discussion of the attractive image force, is given in appendix C.

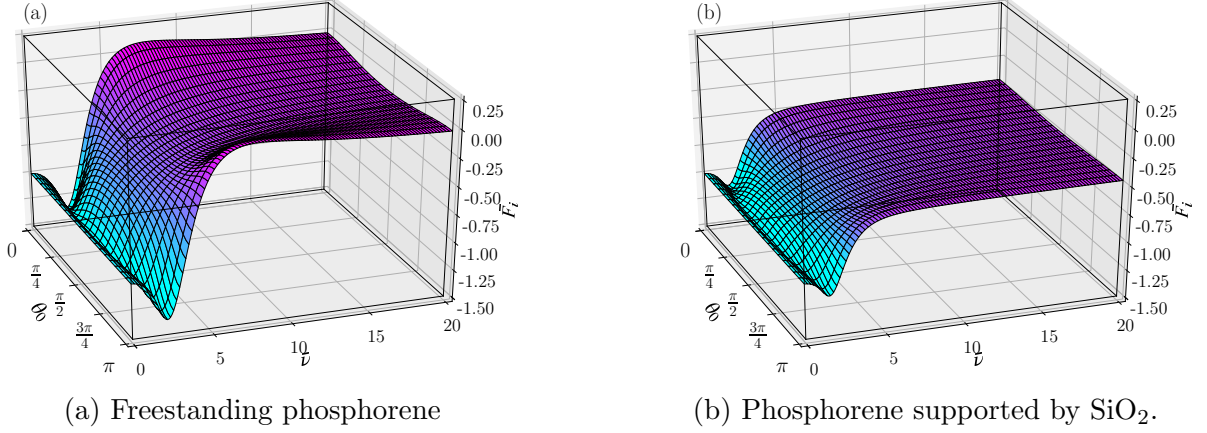


Figure 4.6: Normalized image force $\bar{F}_i = \frac{F_i}{F_0}$ obtained from the optical model, as a function of the normalized speed $\bar{v} = \frac{v}{v_B}$ and direction angle θ_0 for a particle moving at height $z_0 = 50$ nm.

4.5 Plasmon wake in phosphorene

In the preceding section, we examined the forces acting on a charged particle moving parallel to a phosphorene layer, which could be either free-standing or supported by SiO₂. This analysis only required the induced electric potential (specifically, its gradient) at the particle's position. In this section, we thoroughly investigate the total electric potential across the entire plane of the phosphorene, $\Phi(\mathbf{r}, z = 0, t)$, for the same system in which a charged particle moves parallel to a freestanding phosphorene layer (located in the $z = 0$ plane) with velocity $\mathbf{v}$, at a distance z_0 above it.

The total potential in the plane of phosphorene for such a system in the Fourier domain is given by

$$\tilde{\Phi}(\mathbf{q}, 0, \omega) = \int dz' \tilde{\rho}_{\text{ext}}(\mathbf{q}, z', \omega) G(\mathbf{q}, 0, z', \omega), \quad (4.6)$$

where $G(\mathbf{q}, 0, z', \omega) = G_{\text{ind}}(\mathbf{q}, 0, z', \omega) + G_0(q, 0, z')$ and $\tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega)$, $G_{\text{ind}}(\mathbf{q}, z, z', \omega)$, and $G_0(q, z, z')$ are given in Eqs. (2.51), (2.40), and (2.39), respectively. Substituting $\tilde{\rho}_{\text{ext}}(\mathbf{q}, z, \omega)$ from Eqs (2.51) into Eq. (4.6) gives

$$\tilde{\Phi}(\mathbf{q}, 0, \omega) = 2\pi Ze\delta(\omega - \mathbf{q} \cdot \mathbf{v})G(\mathbf{q}, 0, z_0, \omega), \quad (4.7)$$

with its inverse Fourier transformation given by

$$\Phi(\mathbf{r}, 0, t) = \frac{Ze}{(2\pi)^2} \int d^2\mathbf{q} \Re\{G(\mathbf{q}, 0, z_0, \mathbf{q} \cdot \mathbf{v}) e^{i\mathbf{q} \cdot (\mathbf{r} - \mathbf{v}t)}\}, \quad (4.8)$$

where, we used the Hermitian parity of the Green's function to eliminate the integral involving the imaginary part. Now we change the frame of reference based on $(\mathbf{r} - \mathbf{v}t) \rightarrow \mathbf{r}$, which represents the reference frame in which the charged particle is fixed and the phosphorene sheet is moving at $-\mathbf{v}$. Note that in this frame of reference, the electric potential becomes stationary, i.e., independent of time t . Therefore, we take the simplified notation $\Phi(\mathbf{r}, 0, t) \rightarrow \phi(\mathbf{r} - \mathbf{v}t)$ with the electric potential in the moving reference at the plane of phosphorene given by

$$\phi(\mathbf{r}) = \frac{Ze}{2\pi} \int d^2\mathbf{q} \frac{e^{-qz_0}}{q} \Re\left\{ \frac{e^{i\mathbf{q} \cdot \mathbf{r}}}{\epsilon_{2D}(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})} \right\}, \quad (4.9)$$

upon using Eqs.(2.40) and (2.39). We used this closed form representation of the electric potential (Eq. (4.9)) to numerically compute the potential profile. Moreover, for the effective dielectric function $\epsilon_{2D}(\mathbf{q}, \omega)$ appearing in Eq. (4.9), we employed our anisotropic Drude, optical, and semiclassical models, similar to their use in the force calculations. These models were explained in detail in the preceding chapter. Unless clearly stated otherwise, the numerical values used for the parameters in these models are also the same as those used in the force calculations. In this section, the x axis is again aligned with the phosphorene AC direction, and the direction angle of the particle velocity is measured relative to the AC direction. This allows us to express $\mathbf{v}$ in polar form as $\mathbf{v} = (v, \theta_0)$. Moreover, all distances in the plane of phosphorene are normalized according to $\bar{x} = x/l_c$ and $\bar{y} = y/l_c$, where $l_c = 2\pi m_0 e^2 / (\hbar^2 n) \approx 1.19 \mu\text{m}$.

Our setup is designed for electron energy loss spectroscopy (EELS) experiments using aloof electron beams [67, 140, 141]. We choose a distance of $z_0 = 50 \text{ nm}$, typical for these experiments, and set the particle speed to $v = 2v_B$, where $v_B = e^2/\hbar$ is the Bohr speed. This speed ensures an average Froude number [142–144], $Fr = v/\sqrt{z_0 g_d}$, of about 1, with $g_d = 2\pi e^2 n/m_d$ and $m_d = \sqrt{m_x m_y}$ representing the density of states (DOS) mass of phosphorene. Under these conditions, we can ignore relativistic effects [145, 146] and the quantum mechanical wavelength of the incident electron [141] in the wake potential.

A qualitative information about the wake patterns in the far-field region of the $\mathbf{r} = (x, y)$ plane is derived using the method of stationary phase [92–94]. Because of the analogy with Kelvin's wake, the wave pattern in the electric potential in the plane of the material that are obtained from the method of stationary phase may be called the Kelvin-Drude (KD) wake.

Roughly, stationary phase method considers the singularities of the integrand in Eq. (4.9) in the $\mathbf{q}$ plane as the dominant contributors in the integral. Constant phase curves are found from the critical points of $f = \mathbf{q} \cdot \mathbf{r}$ in Eq. (4.9) for a fixed $\mathbf{r}$, under the constraint $\epsilon(\mathbf{q}, \mathbf{q} \cdot \mathbf{v}) = 0$. This is equivalent to solving $\epsilon(\mathbf{q}, \omega) = 0$ without damping to obtain the plasmon frequency $\omega = \omega_p(\mathbf{q})$, while applying the radiation condition $\omega_p(\mathbf{q}) = \mathbf{q} \cdot \mathbf{v}$, which was called the kinematic resonance condition in the context of force calculations. The curves of constant phase f are then obtained as [92, 94]

$$\mathbf{r} = f \frac{\mathbf{v}_g(\mathbf{q}) - \mathbf{v}}{\mathbf{q} \cdot \mathbf{v}_g(\mathbf{q}) - \omega_p(\mathbf{q})}, \quad (4.10)$$

where $\mathbf{v}_g(\mathbf{q}) = \frac{\partial \omega_p(\mathbf{q})}{\partial \mathbf{q}}$ is the plasmon group velocity. It is more convenient to express this equation in the polar coordinates, $\mathbf{q} = (q, \theta)$, giving rise to the polar representation of the constant phase curves

$$x(\theta) = \frac{f}{\Delta} \left(\frac{\partial \omega_p}{\partial q} \cos \theta - \frac{\partial \omega_p}{\partial \theta} \frac{\sin \theta}{q} - v \cos \theta_0 \right), \quad (4.11)$$

$$y(\theta) = \frac{f}{\Delta} \left(\frac{\partial \omega_p}{\partial q} \sin \theta + \frac{\partial \omega_p}{\partial \theta} \frac{\cos \theta}{q} - v \sin \theta_0 \right), \quad (4.12)$$

where $\Delta \equiv q \frac{\partial \omega_p}{\partial q} - \omega_p$, and $\omega_p(q, \theta)$ could be obtained from one of the Eqs. (3.34), (3.35), or (3.36) after the change of the variables $q_x \rightarrow q \cos \theta$ and $q_y \rightarrow q \sin \theta$. Note that these parametric expressions for the constant phase curves must be applied with the radiation condition $\omega_p(\mathbf{q}) = \mathbf{q} \cdot \mathbf{v}$, which defines a curve in the $\mathbf{q}$ plane. This condition allows us to express q as a function of θ , $q = q_r(\theta)$. Moreover, the function $q_r(\theta)$ can be analytically obtained for all three models that we use to describe the effective dielectric function of the freestanding phosphorene. Therefore, Eqs.(4.11) and (4.12), after substituting $q \rightarrow q_r(\theta)$, yield the parametric representation of the constant phase curves (for different values of the phase f) with the angle $\theta \in [\theta_0 - \frac{\pi}{2}, \theta_0 + \frac{\pi}{2}]$ serving as the parameter [94]. Note that by examining the extremal points in the slope function $S(\theta) = -y/x$, the opening angles φ_w of the wedges, which enclose various types of waves in the wake, can be determined. These angles are measured with respect to the direction opposite to $\mathbf{v}$ [147].

4.6 Potential profile calculations

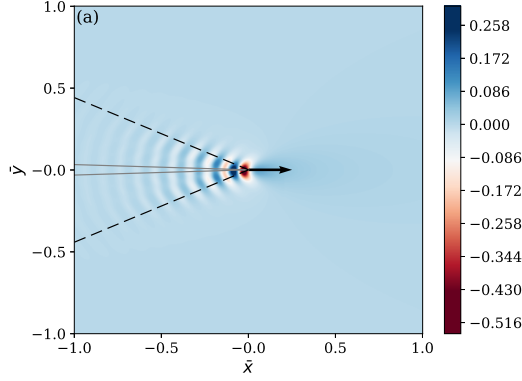
In Fig. 4.7 we present the density plots of the wake potential in doped phosphorene for a charged particle moving at $v = 2v_B$ in three different directions, obtained from Eq. (4.9).

In the AC direction, depicted in Fig. 4.7a, where the electron mass m_x is light, the wake is mainly composed of transverse waves that are concave relative to the direction of motion, differing from the typical convex shape of transverse waves in Kelvin’s wake [92, 94, 143, 144]. This is a surprising result, which resembles the wake patterns found in a system that exhibits the dispersion relation with negative group velocity [148]. In our system, this transition from the convex to the concave transverse waves in the AC direction is a consequence of increasing anisotropy between the effective electron masses in the 2DEG, as shown in Fig. 4.8a below. The half-opening angle of the wedge enclosing the wake is found to be $\varphi_{AC} \approx 23.8^\circ$ from the stationary phase analysis, which is slightly larger than $\varphi_K \approx 19.5^\circ$ of the classic Kelvin wake. In the ZZ direction, illustrated in Fig. 4.7c, where the electron mass m_y is heavier, the wake predominantly consists of diverging waves, forming a wedge with a significantly larger half-angle of $\varphi_{ZZ} \approx 57.2^\circ$. The stationary phase analysis shows that the wakes consist of transverse, diverging, and hybrid waves in the AC direction, though some of these waves are suppressed depending on the Froude number, $Fr = v/\sqrt{z_0 g}$. For the classic Kelvin’s wake, transverse waves dominate at smaller $Fr < 1$, while diverging waves are more prominent at larger $Fr > 1$ values [142–144]. Using our parameters to calculate the Froude number with $g_{x/y} = 2\pi e^2 |n|/m_{x/y}$, we find $Fr \approx 0.57$ in the AC direction and $Fr \approx 1.64$ in the ZZ direction, explaining the types of waves observed in Figs. (4.7a) and (4.7c).

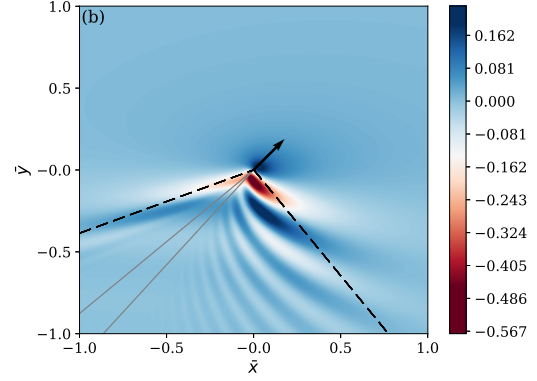
We do not pursue further discussion of the role of the distance z_0 between the moving particle and phosphorene, but we note that it effectively introduces an ultraviolet cut-off, $q_0 \propto 1/z_0$, in the integral in Eq. (4.9). There is a simple way to understand the effects of variable z_0 on the resulting wake patterns based on the observation that Kelvin’s construction is effectively stopped at q_0 [149].

The most notable wake pattern is shown in Fig. 4.7b for the direction at $\theta_0 = \pi/4$, midway between the AC and ZZ directions. This wake is mainly composed of diverging waves that display significant anisotropy with respect to the particle’s direction of motion. The wedge formed by these waves has opening angles of $\varphi_{\text{right}} \approx 82.7^\circ$ to the right (towards AC) and $\varphi_{\text{left}} \approx 23.9^\circ$ to the left (towards ZZ). This wake asymmetry resembles the asymmetry found in Kelvin’s ship wake when a sideways shear current is present in water underneath the ship [93, 147] (see, for example, Fig. (4) in Ref. [93]).

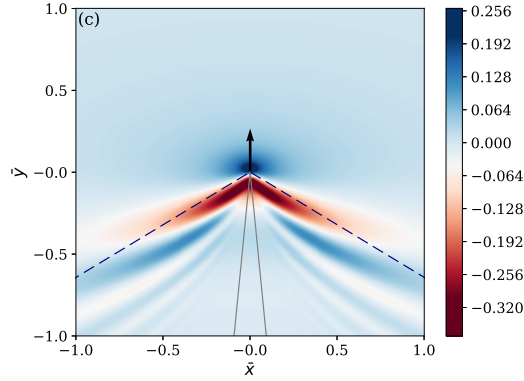
In the KD potential wake for phosphorene obtained from Eq. (4.9), the asymmetry seen in the far-field area is predominantly driven by the differences in effective electron masses. Thus, in Fig. 4.8, we analyze the isophase contours derived from an anisotropic Drude model, where the DOS mass $m_d = \sqrt{m_x m_y}$ is constrained to be equal to that from the *ab initio* data for phosphorene [1], while the anisotropy ratio $\mu = m_y/m_x$ is varied as a parameter. It is important to note that the isotropic Drude case with $\mu = 1$ yields



(a) $\theta_0 = 0$, AC direction.



(b) $\theta_0 = \pi/4$, midway direction.



(c) $\theta_0 = \pi/2$, ZZ direction.

Figure 4.7: The normalized wake potential, $\frac{\phi z_0}{(Ze)^2}$, in doped phosphorene, as a function of the normalized coordinates $\bar{x}$ (AC axis) and $\bar{y}$ (ZZ axis) from semiclassical model. The arrow shows the direction of motion for the particle that moves at distance $z_0 = 50$ nm with the speed $v = 2v_B$. The dashed black lines define the wedge enclosing the KD wake, while the solid grey lines are the asymptotes of the fan waves, defining the Mach-like wedge, see Fig. 4.9.

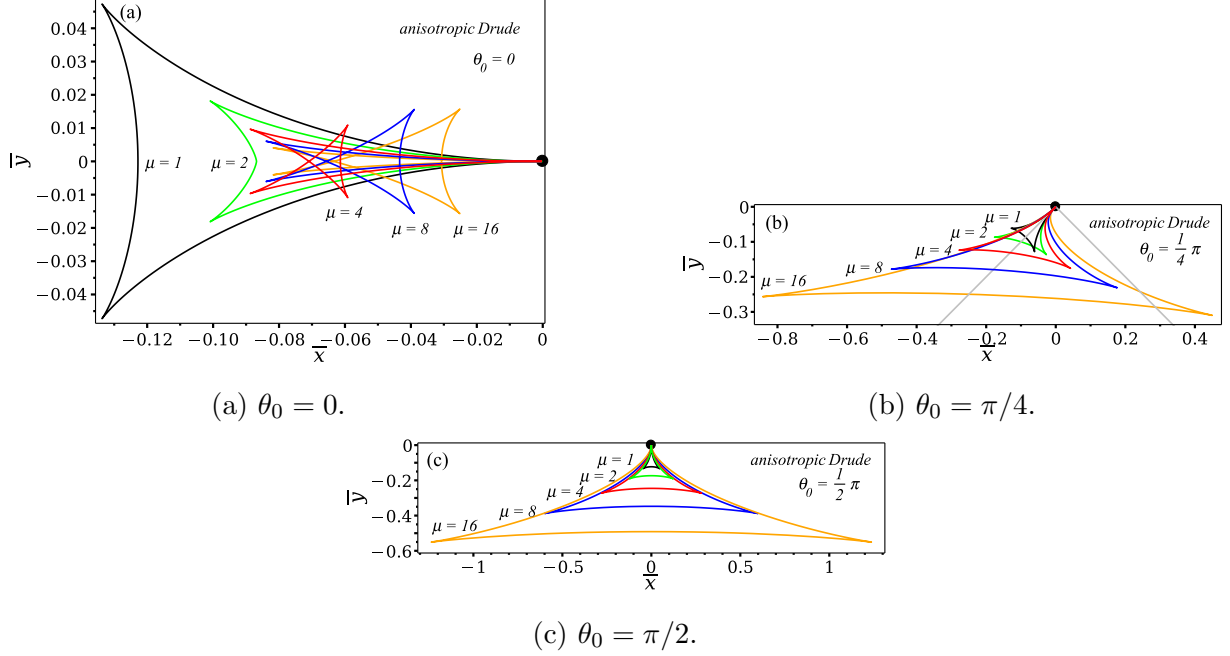


Figure 4.8: The isophase curves for $f = -\pi$, in the normalized coordinates $\bar{x}$ (AC axis) and $\bar{y}$ (ZZ axis), obtained from the anisotropic Drude model. The DOS mass $m_d = \sqrt{m_x m_y}$ is kept constant while varying the ratio $\mu = m_y/m_x$ to values of 1 (black), 2 (green), 4 (red), 8 (blue), and 16 (orange) for a particle speed of $v = 2v_B$. The gray lines in (b) represent the axes rotated by $\theta_0 = \pi/4$.

the solid black deltoid curve in Fig. 4.8, which corresponds to the classic Kelvin pattern featuring two diverging waves and one convex transverse wave.

As shown in Fig. 4.8a, when μ exceeds 2, the transverse and diverging waves no longer converge at the same pair of cusps. Instead, two new branches appear, which we call hybrid waves because they link the endpoints of the transverse wave to those of the diverging waves on the opposite sides of the wake. This transition leads to a topological change from a three-cusped Kelvin's deltoid to a five-cusped pentacle when μ reaches 2. In Figs. (4.8b) and (4.8c), the KD wake maintains a deltoid shape, which enlarges and broadens with an increase in μ , while becoming asymmetrically distorted when $\theta_0 = \pi/4$ in Fig. 4.8b. Interestingly, in Fig. 4.8b, the diverging wave in the wake with $\mu = 16$ appears ahead of the particle's position on the side toward the AC direction. Similarly, in Kelvin's ship wake, one of the wedge angles also exceeds $\pi/2$ in the presence of a strong sideways shear current [93]. Note that based on the values of our model parameters for phosphorene,

$\mu \approx 8.4$.

It is important to highlight that within the anisotropic Drude model, the wake patterns remain unchanged regardless of particle speed. Similar to the classic Kelvin wake, the isophase curves depicted in Fig. 4.8 can be scaled with $\lambda = v^2/g_d$, eliminating the speed dependency when coordinates are normalized as $\tilde{x} = x/\lambda$ and $\tilde{y} = y/\lambda$. Conversely, the optical and semiclassical models, while approximating the Drude model wake patterns at high speeds, reveal significant shape changes in the isophase curves at lower speeds, particularly when $v < v_B$. Investigating these changes necessitates more complex dielectric function models to account for non-local effects at low speeds, which are beyond the scope of the current study.

Significant differences among the three dielectric models for doped phosphorene are most evident when examining particle motion in the AC direction. In Fig. 4.9, three isophase curves are compared to illustrate these differences. The Drude model (green) serves as a baseline. The optical model (red) includes the interband contributions that shift the central cusp of the pentacle, where diverging waves meet, much further behind the particle. Despite this shift, two wedges enclosing the transverse (dashed) and diverging (dotted) waves remain. The semiclassical model (blue) largely follows the optical model's isophase curve, but with a notable distinction at the cusp region where diverging waves meet. Here, nonlocal effects give rise to a pair of "fan" waves [150], disrupting the cusp and extending asymptotically behind the particle, creating a Mach-like wedge (gray lines in Fig. 4.7). These fan branches meet the diverging branches at new cusps, forming an additional wedge (dash-dotted) in Fig. 4.9, similar to structures observed in recent capillary-gravity wake studies (see Fig. 7 in Ref. [144] or Fig. 8 in Ref. [150]).

In order to make a deep comparison between the three different conductivity models, in Fig. 4.10, we examine the isophase curves for these models at a speed of $v = 2v_B$ using the phosphorene parameters given before. In this analysis, we use the normalized coordinates $\tilde{x} = x/\lambda$ and $\tilde{y} = y/\lambda$, where $\lambda = v^2/g_d$ with $g_d = 2\pi e^2 n/m_d$, which make the isophase curves scale-invariant with respect to the speed v , within the anisotropic Drude model. Note that, Fig. 4.10a reproduces Fig. 4.9 with different normalized coordinates x and y . In both the Drude and optical models, the wake pattern exhibits two pairs of cusps in the pentacle where hybrid waves meet transverse and diverging waves. These cusps, along with the particle's position at the origin, define the arms of a wider wedge (dashed lines) enclosing a concave transverse wave which we label by W_t and a narrower wedge (dotted lines) enclosing diverging waves which we label by W_d . In the semiclassical model, the emergence of fan waves creates a third pair of cusps, disrupting the confluence of diverging waves and forming the narrowest wedge (dash-dotted lines). A detailed examination of isophase curves in the semiclassical model shows that fan waves extend asymptotically

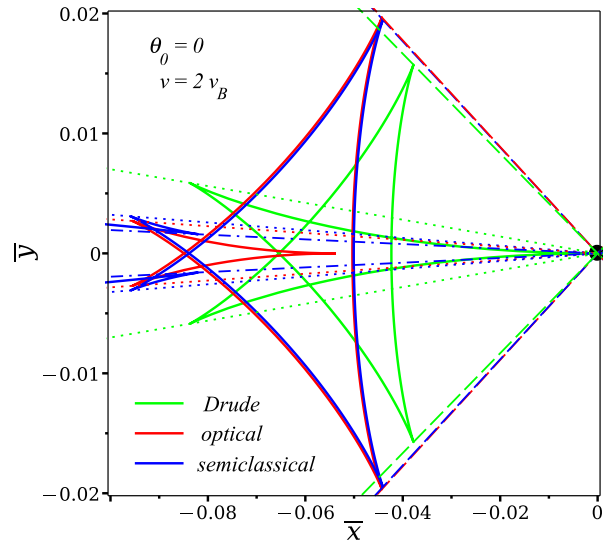


Figure 4.9: The isophase curves (solid lines) for $f = -\pi$ in normalized coordinates $\bar{x}$ (AC axis) and $\bar{y}$ (ZZ axis) are shown using three models for doped phosphorene: Drude (green), optical (red), and semiclassical (blue). These curves depict a particle moving in the AC direction ($\theta_0 = 0$) at a speed of $v = 2v_B$. The wedges enclosing the transverse and diverging waves are marked by dashed and dotted lines, respectively. The dash-dotted lines indicate the cusps where "fan" waves detach from the diverging waves in the semiclassical model.

behind the particle (indicated by gray lines in Fig. 4.7), converging towards the arms of the middle wedge (W_d) that enwraps the diverging waves (dotted lines). This behavior, driven by a minimum in the plasmon phase speed (ω/q) near the e-h continuum (see Fig. 3.2), is reminiscent of Mach-like features seen in Kelvin’s ship wake under capillary effects [94, 144, 150]. For directions $\theta_0 = \pi/4$ and $\theta_0 = \pi/2$ in Figs. (4.10b) and (4.10c), the isophase curves in the semiclassical and optical models align more closely with the Drude model. In these cases, hybrid waves are absent, leaving only two wedges in the semiclassical model: one defined by the confluence of diverging and transverse waves (dashed lines), showing a classic Kelvin’s convex shape, and a narrower wedge (dash-dotted lines) where fan waves disrupt the diverging waves.

Expecting that the Drude model would become increasingly accurate for the wake patterns at higher speeds, we present in Fig. 4.11 the isophase curves for all three models in the same directions of motion as in Fig. 4.10, but with the speed increased to $v = 7v_B$. The green curves representing the Drude model remain unchanged in both shape and size when compared to those in Fig. 4.10. However, the red curves (optical model) and blue curves (semiclassical model) show a closer alignment with the green curves in Fig. 4.11 than in Fig. 4.10, as anticipated. In panels (b) and (c) of Fig. 4.11, the blue and red curves nearly coincide with the green curves, except in narrow segments around the direction of motion. Here, the blue curves exhibit Mach-like wedges, resulting from the asymptotic behavior of the fan waves. This demonstrates the expected convergence of the optical and semiclassical models towards the Drude model predictions at higher particle speeds.

We finish this section by giving in Fig. 4.12, the density plots from Fig. 4.7 that are superimposed with isophase curves for $f = -\pi(2\ell - 1)$ for $\ell = 1, 2, 3, \dots$. These curves are derived as parametric plots from Eqs. (4.11) and (4.12) using the semiclassical model with phosphorene parameters. The isophase curves align remarkably well with the near-zero regions of the wake potential (pale blue areas in the density plots). The dashed lines indicate the wedge encompassing all waves in the wakes. Additionally, there is a significant clustering of the fan waves along the asymptotic lines, creating narrow Mach-like regions, as shown by the gray lines in Fig. 4.7. The angles of these asymptotes are found by analyzing the extremal points of the slope function $S(\theta)$, yielding the following values: $\varphi_M \approx \pm 1.8^\circ$ for $\theta_0 = 0$, $\varphi_M \approx 4.2^\circ$ and 3.8° for $\theta_0 = \pi/4$, and $\varphi_M \approx \pm 5.3^\circ$ for $\theta_0 = \pi/2$. The rich structures in the isophase curves of Fig. 4.12a due to diverging, hybrid, and fan waves are not visible in the corresponding density plot. This is likely because of the small Froude number in the AC direction and finite damping used in the computations. Similarly, the suppression of transverse and fan waves in Figs. (4.12b) and (4.12c) is probably due to the larger Froude numbers in those directions [142].

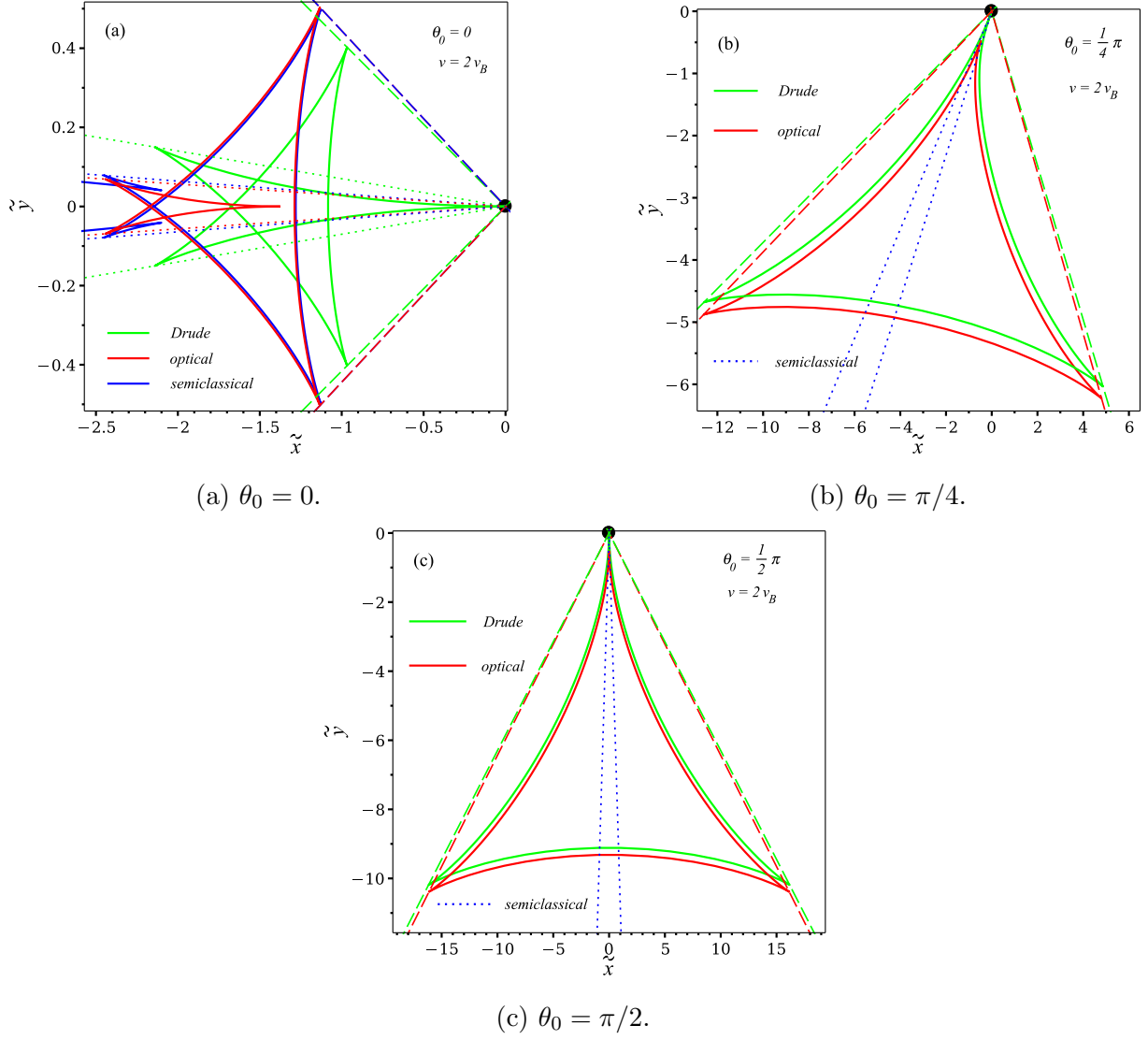


Figure 4.10: The isophase curves for $f = -\pi$, in the normalized coordinates $\tilde{x} = xg_d/v^2$ (AC axis) and $\tilde{y} = yg_d/v^2$ (ZZ axis), obtained from the anisotropic Drude model (green), optical model (red) and semiclassical models (blue) of doped phosphorene. The particle speed is $v = 2v_B$.

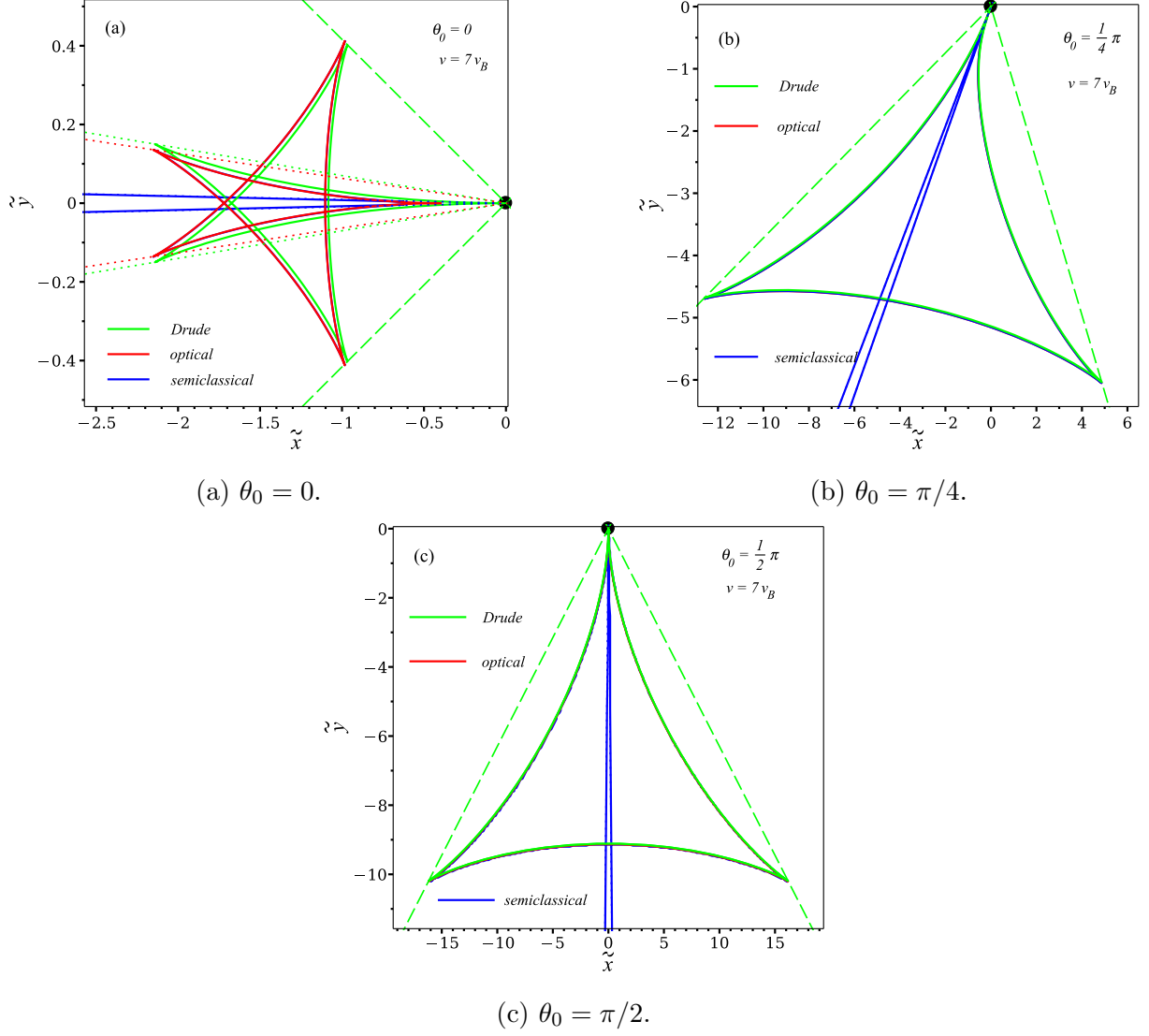


Figure 4.11: The isophase curves for $f = -\pi$, in the normalized coordinates $\tilde{x} = xg_d/v^2$ (AC axis) and $\tilde{y} = yg_d/v^2$ (ZZ axis), obtained from the anisotropic Drude model (green), optical model (red) and semiclassical models (blue) of doped phosphorene. The particle speed is $v = 7v_B$.

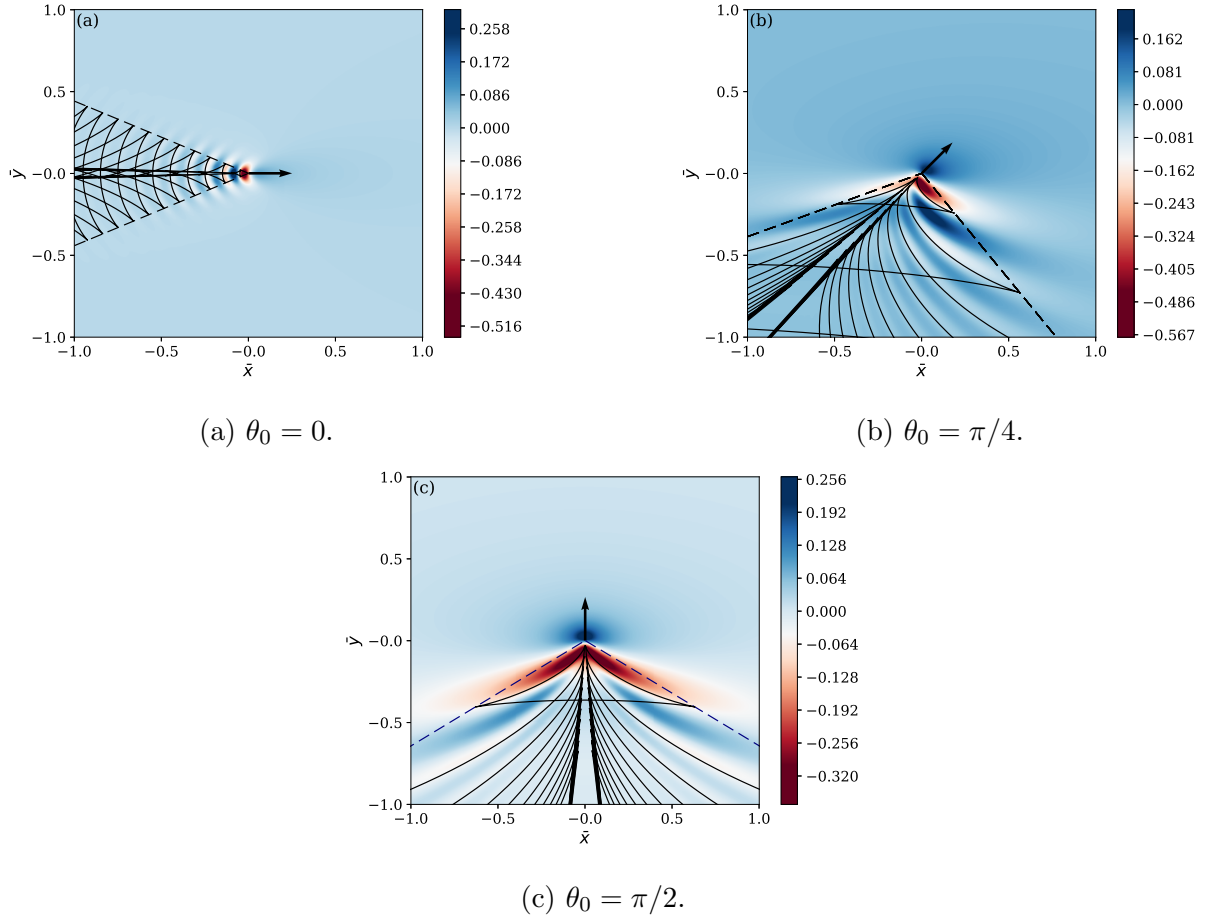


Figure 4.12: The normalized wake potential, $\frac{\phi z_0}{(Ze)^2}$, in doped phosphorene, as a function of the normalized coordinates $\bar{x}$ (AC axis) and $\bar{y}$ (ZZ axis) from semiclassical model. The arrow shows the direction of motion for the particle that moves at distance $z_0 = 50$ nm with the speed $v = 2v_B$. The solid black curves in represent the isophase curves derived from Eqs. (4.11) and (4.12) using the same semiclassical model with phosphorene parameters. The dashed black lines delineate the arms of the wedge that encompasses the wake. The solid grey lines indicate the asymptotes of the fan waves, which define the Mach-like wake region.

4.7 Numerical integration

To compute the forces in Eqs. (4.3), (4.4), and (4.5), or the wake potential in Eq. (4.9), numerical integration over the entire plane is required. Fortunately, the exponential factors e^{-2qz_0} or e^{-qz_0} in the integrands help reduce computational expenses by making the contributions from regions far from the origin negligible. However, the strongly oscillatory phase factor $e^{i\mathbf{q}\cdot\mathbf{r}}$ can complicate the computation of the potential $\phi(\mathbf{r})$.

Nevertheless, these numerical computations are performed using the built-in methods of `scipy`, a well-known Python library for scientific computing [151]. The methods in this library employ sophisticated mesh refinements and error predictions to ensure the accuracy [152]. In all calculations, the cut-off condition for the integration domain was set to $q < \frac{15}{z_0}$ for force calculations and $q < \frac{30}{z_0}$ for potential calculations, ensuring a prefactor of $e^{-30} \approx 9.3 \times 10^{-14}$ in the regions excluded from the integration process.

Chapter 5

Phosphorene carrier mobility tensor limited by charged impurity scattering: an energy loss method analysis

5.1 Introduction

In free space, applying a constant electric field to a point charge results in constant acceleration, leading to potentially unlimited speeds. In materials, however, due to scattering processes, the final speed of quasi-free charge carriers is proportional to the electric field for small fields and is bounded at high fields. The proportionality constant between drift velocity and electric field is known as the direct current (DC) mobility of charge carriers within the material. Generally, mobility is represented as a tensor $\vec{\mu}$, satisfying the relation $\mathbf{v} = \vec{\mu} \cdot \mathbf{E}$, where $\mathbf{v}$ is the drift velocity and $\mathbf{E}$ is the constant electric field in the material. At finite temperatures, the main scattering process affecting mobility is carrier-phonon scattering, which involves the impact of lattice vibrations on carrier mobility. In nano-electronic devices based on phosphorene, measurements of transport properties are still limited [103], and there is a significant gap between experimental results and theoretical models, especially for carrier-phonon scattering at room temperature [104, 105, 153, 154]. Synthesizing freestanding phosphorene is nearly impossible, so it is typically supported by other materials, which introduce a random distribution of charged impurities. At low temperatures, carrier-phonon scattering is negligible, and the primary limiting factor for

mobility is scattering from charged impurities (CIs) due to Coulomb interaction. Solving the linearized Boltzmann transport equation (BTE) is a well-established method for computing drift mobility tensors for anisotropic 2D materials due to carrier scattering on CIs at low temperatures [2, 91, 100–102]. In this method the carrier mobility tensor can be described in terms of the momentum relaxation times, which are the numerical solutions of an integral equation depending on the direction of the applied electric field. However, the BTE approach has significant limitations when exploring various designs of nanoelectronic devices based on phosphorene [103–105], as it is extremely computationally expensive, if not impractical, to study the effects of spatial arrangements of CIs on phosphorene mobility.

The Energy Loss Method (ELM) is an elegant but less commonly used technique compared to the Boltzmann transport equation for calculating the drift mobility of carriers [106–108]. The core idea of ELM for carrier scattering on charged impurities (CIs) involves a straightforward Galilean transformation between the reference frame where the impurities are stationary and the frame moving with the drift velocity of the charge carriers in the material [109, 110, 155, 156]. In the reference frame where the charged impurities are stationary and phosphorene charge carriers are moving with drift velocity $\mathbf{v}$, the energy dissipation rate consists of Joule heating from the electric field $\mathbf{E}$ and the current density from the drift of the phosphorene carriers. Conversely, in the frame where the charged impurities are moving at velocity $-\mathbf{v}$, there is no drift current in phosphorene. Instead, energy dissipation is due to the friction force experienced by the moving CIs in the electric field coming from dynamic polarization of the phosphorene charge carriers by the impurities themselves. The energy loss rate for moving impurities must be equal to the Joule heating occurring in the material [111]. Notice that unlike the BTE approach, which requires solving a complex integral equation for MRT, the ELM provides an explicit analytical expression for the drift mobility or the direct current (DC) electric conductivity, represented as an integral involving a suitably screened external potential for scattering of charge carriers.

In this chapter, we analyze the standard structure depicted in Fig. 5.1, which includes a semi-infinite substrate and an anisotropic 2DEG representing the phosphorene positioned at a distance h above the substrate. It is important to note that the supporting material contains a random distribution of charged impurities. We use silicon dioxide (SiO_2) as the substrate and consider several models for the distribution of the charged impurities: a uniform 3D distribution, a uniform planar distribution at a depth d within the substrate, and the hard disk (HD) model for the planar distribution [157]. In the case of the HD model, we conduct a thorough investigation into how the correlation distance affects the mobility of the carriers.

The materials presented in this chapter are a representation of our research paper,

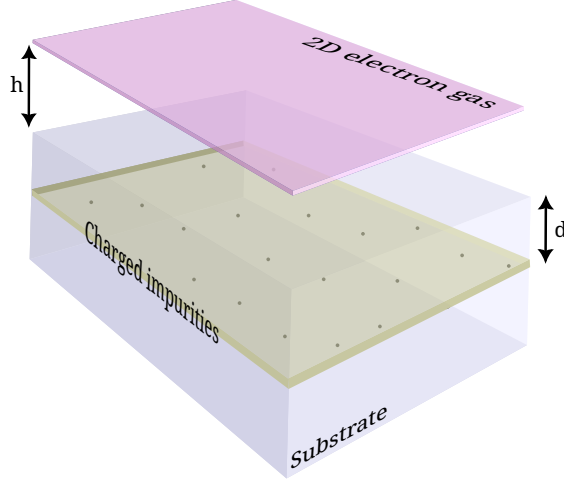


Figure 5.1: Perspective of the standard system. Black dots show the CIs inside the substrate. 2D electron gas represents the phosphorene sheet. h is the gap size between the substrate and phosphorene, whereas d is the depth of the CIs that are distributed in a plane parallel to the phosphorene.

published in Ref. [116].

5.2 Modeling and the energy loss method

In a Cartesian coordinate system with coordinates $\mathbf{R} \equiv \{\mathbf{r}, z\}$, where $\mathbf{r} = \{x, y\}$, we consider a large-area flat phosphorene sheet positioned in vacuum at a height $h \geq 0$ above the substrate with a relative dielectric constant ϵ_s . The substrate occupies the half-space $z \leq 0$ and contains a significant number N of point-like impurities, each with a charge of Ze (where e is the elementary charge).

The impurities are distributed statistically, with a charge density $\rho_0(\mathbf{r}, z)$ in the laboratory frame, which is translationally invariant in the $\mathbf{r}$ directions. The average density, obtained by ensemble averaging over the impurity arrangements, is denoted $\langle \rho_0(\mathbf{r}, z) \rangle \equiv \bar{\rho}_0(z)$ and depends only on the perpendicular distance z . This average density does not affect carrier scattering in phosphorene, but its integral, $\int dz \bar{\rho}_0(z) = Ze n_{\text{imp}}$, with $n_{\text{imp}} = \frac{N}{A}$ representing the areal density of impurities, determines the doping level of phosphorene with electrons or holes, resulting in an equilibrium areal density n . The scattering of

carriers is influenced by the fluctuation part of the impurity density, given by $\delta\rho_0(\mathbf{r}, z) = \rho_0(\mathbf{r}, z) - \bar{\rho}_0(z)$.

The ELM relies on a thought experiment related to conductivity measurement [107, 110, 155]. In this setup, a constant electric field $\mathbf{E}$ is applied tangentially to phosphorene, inducing an in-plane drift current $\mathbf{J}_d$. The total Joule power dissipated in phosphorene is given by $P_J = \iint d^2\mathbf{r} \mathbf{J}_d \cdot \mathbf{E} = A \mathbf{J}_d \cdot \mathbf{E}$. If the current is expressed as $\mathbf{J}_d = -en\mathbf{v}_d$, where $\mathbf{v}_d$ is the constant drift velocity of the charge carriers in electron-doped phosphorene, Ohm's law can be written as $\mathbf{J}_d = \overleftrightarrow{\sigma} \cdot \mathbf{E}$, where $\overleftrightarrow{\sigma}$ is the in-plane DC conductivity tensor of phosphorene. This tensor can be related to the mobility tensor by $\overleftrightarrow{\sigma} = -en\overleftrightarrow{\mu}$. Thus, we have $P_J = -Aen\mathbf{v}_d \cdot \mathbf{E}$ and the drift velocity $\mathbf{v}_d = \overleftrightarrow{\mu} \cdot \mathbf{E}$. By substituting this expression into the formula for Joule power, we can eliminate the electric field in terms of the drift velocity to obtain the final expression for the Joule power as

$$P_J = -Aen\mathbf{v}_d \cdot \overleftrightarrow{\mu}^{-1} \cdot \mathbf{v}_d, \quad (5.1)$$

where $\overleftrightarrow{\mu}^{-1}$ is the inverse of the mobility tensor of phosphorene.

The equivalence principle applied in the ELM by changing reference frames states that the Joule power observed in a conductivity measurement with drifting carriers in phosphorene is equivalent to the energy dissipation experienced by the system of charged impurities moving with an opposite velocity $\mathbf{v} = -\mathbf{v}_d$ in a frame where phosphorene's carriers are in a state with zero drift. In this stationary frame, the fluctuation part of the impurity density, given by $\delta\rho(\mathbf{R}, t) = \delta\rho_0(\mathbf{r} - \mathbf{v}t, z)$, creates an electrostatic potential, $\delta\Phi_{\text{pol}}(\mathbf{R}, t)$, due to the polarization of charge carriers in phosphorene from their equilibrium state. This potential can be expressed using the Green's function (GF) of the Poisson equation, which includes the effects of static screening by nearby dielectrics [158]

$$\delta\tilde{\Phi}_{\text{pol}}(\mathbf{q}, z, \omega) = \int_{-\infty}^{\infty} dz' G_{\text{ind}}(\mathbf{q}, z, z', \omega) \delta\tilde{\rho}(\mathbf{q}, z', \omega), \quad (5.2)$$

where

$$\delta\tilde{\rho}(\mathbf{q}, z, \omega) = \mathcal{F}\{\delta\rho_0(\mathbf{r} - \mathbf{v}t, z)\} = 2\pi \delta(\omega - \mathbf{q} \cdot \mathbf{v}) \delta\tilde{\rho}_0(\mathbf{q}, z) \quad (5.3)$$

and $G_{\text{ind}}(\mathbf{q}; z, z'; \omega)$ is given in Eq. (2.47). Notice that the factor $\delta(\omega - \mathbf{q} \cdot \mathbf{v})$ expresses the Galilean transformation between the moving frame and the rest frame of phosphorene. Moreover, the Fourier transform of the fluctuating part of the CI density with respect to the in-plane coordinates in the laboratory frame is given by

$$\delta\tilde{\rho}_0(\mathbf{q}, z) = \tilde{\rho}_0(\mathbf{q}, z) - (2\pi)^2 \delta(\mathbf{q}) \bar{\rho}_0(z). \quad (5.4)$$

The energy loss rate of the moving system of CIs can be obtained from

$$P_{\text{imp}} = \iiint d^3\mathbf{R} \mathbf{J}_{\text{imp}}(\mathbf{R}, t) \cdot \mathbf{E}_{\text{pol}}(\mathbf{R}, t), \quad (5.5)$$

where $\mathbf{J}_{\text{imp}}(\mathbf{R}, t) = \mathbf{v}\delta\rho_0(\mathbf{r} - \mathbf{v}t, z)$ is the current density due to the rigidly moving system of the CIs and $\mathbf{E}_{\text{pol}}(\mathbf{R}, t) = -\nabla_{\parallel}\delta\Phi_{\text{pol}}(\mathbf{r}, z, t)$ is the induced electric field parallel to phosphorene. Switching to the Fourier domain and using Eqs. (5.2) and (5.3) in Eq. (5.5) allows us to write an ensemble average of the impurity energy loss rate as

$$\langle P_{\text{imp}} \rangle = -i \iint \frac{d^2\mathbf{q}}{(2\pi)^2} (\mathbf{q} \cdot \mathbf{v}) \int dz \int dz' G_{\text{ind}}(\mathbf{q}, z, z', \mathbf{q} \cdot \mathbf{v}) \langle \delta\tilde{\rho}_0(-\mathbf{q}, z) \delta\tilde{\rho}_0(\mathbf{q}, z') \rangle. \quad (5.6)$$

Now we introduce the function defined by

$$\mathcal{S}(\mathbf{q}) = \frac{1}{Ne^2Z^2} \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \langle \delta\tilde{\rho}_0(-\mathbf{q}, z) \delta\tilde{\rho}_0(\mathbf{q}, z') \rangle, \quad (5.7)$$

that only depends on the distribution of the CIs, which is why it is referred to as the geometric structure factor. Note that the structure factor is a real-valued and even function, $\mathcal{S}(\mathbf{q}) = \mathcal{S}(-\mathbf{q})$. Moreover, the Green's function

$$G_{\text{ind}}(\mathbf{q}, z, z', \omega) = \frac{2\pi}{q} \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - \frac{1}{\epsilon_{\text{bg}}(q)} \right] \left[\frac{\epsilon_{\text{bg}}(q)}{\epsilon_{\text{bg}}^0} \right]^2 e^{q(z+z'-2h)}, \quad z, z' \leq 0, \quad (5.8)$$

satisfies the Hermitian parity and the reciprocity property, $G_{\text{ind}}(-\mathbf{q}, z, z', -\omega) = G_{\text{ind}}^*(\mathbf{q}, z', z, \omega)$, whereas ϵ_{bg} and ϵ_{bg}^0 were defined in Eqs. (2.41) and (2.48), but are applied here for the case of constant ϵ_s . Therefore, after taking care of the symmetries and using the definition of the structure factor, Eq. (5.6) can be written as

$$\langle P_{\text{imp}} \rangle = -NZ^2 \iint \frac{d^2\mathbf{q}}{(2\pi)^2} (\mathbf{q} \cdot \mathbf{v}) V_C(q) \left[\frac{\epsilon_{\text{bg}}(q)}{\epsilon_{\text{bg}}^0} \right]^2 e^{-2qh} \mathcal{S}(\mathbf{q}) \Im \left\{ \frac{-1}{\epsilon_{2D}(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})} \right\}, \quad (5.9)$$

where $V_C(q) = 2\pi e^2/q$ is the in-plane FT of the Coulomb potential with $q = \sqrt{q_x^2 + q_y^2}$.

5.2.1 Loss function

Note that the energy loss function $\Im\{-1/\epsilon_{2D}(\mathbf{q}, \omega)\}$ is emerged here again in an expression that describes the energy loss of the system. In order to construct the effective dielectric

function

$$\epsilon_{2D}(\mathbf{q}, \omega) = \epsilon_{bg}(q) + V_C(q) \chi(\mathbf{q}, \omega), \quad (5.10)$$

with $\chi(\mathbf{q}, \omega) = \chi_{\text{intra}}(\mathbf{q}, \omega) + \chi_{\text{inter}}(\mathbf{q}, \omega)$, we need to adopt the appropriate models for the intraband and interband components of the polarization function. In this chapter, for the intraband polarization function we set $\chi_{\text{intra}}(\mathbf{q}, \omega) = \chi^{\text{quantum}}(\mathbf{q}, \omega)$, with $\chi^{\text{quantum}}(\mathbf{q}, \omega)$ from Eq. (3.41). Note that this polarization function is actually the 2D Lindhard function, expanded to the leading term in ω . This approximation is adequate for our purposes in this chapter, where we assume that in the friction regime, the drift velocity is sufficiently small. On the other hand, for the interband electron transitions, we use the linear conductivity model discussed in section 3.4.2 that yields

$$\chi_{\text{inter}}(\mathbf{q}, \omega) = \frac{1}{e^2} (\alpha_x q_x^2 + \alpha_y q_y^2). \quad (5.11)$$

Upon substitution $\omega = \mathbf{q} \cdot \mathbf{v}$, these assumptions give an expression for the loss function to be used in Eq. (5.9) as

$$\Im \left\{ \frac{-1}{\epsilon(\mathbf{q}, \mathbf{q} \cdot \mathbf{v})} \right\} = \frac{\mathbf{q} \cdot \mathbf{v}}{[\epsilon_{\text{stat}}(\mathbf{q})]^2} \frac{q_s}{q} \frac{2m_d}{\hbar Q} \frac{H(2k_F - Q)}{\sqrt{(2k_F)^2 - Q^2}}, \quad (5.12)$$

where $q_s = 2m_d e^2 / \hbar^2$ is the Thomas-Fermi inverse screening length of the equivalent isotropic 2DEG and $\epsilon_{\text{stat}}(\mathbf{q})$ is a static dielectric function of phosphorene, defined in the presence of the interband contribution as [1]

$$\epsilon_{\text{stat}}(\mathbf{q}) = \epsilon_{bg}(q) + \frac{2\pi}{q} (\alpha_x q_x^2 + \alpha_y q_y^2) + \frac{q_s}{q} \quad (5.13)$$

for wavevectors constrained by the condition $Q(\mathbf{q}) < 2k_F$, which follows from Eq. (6.20) with $Q(\mathbf{q})$ given in Eq. (3.42).

5.2.2 Structure factor

The structure factor of the CIs given in Eq. (5.7) can be analytically computed for certain distributions of the CIs. Note that this structure factor is limited to the half-space $z \leq 0$, because we assume that CIs are only distributed inside the substrate and the Green's function in Eq. (5.8) is obtained under this constraint. Assuming the impurities are located

at positions $\mathbf{R}_j \equiv \{\mathbf{r}_j, z_j\}$ in the laboratory frame, with $z_j \leq 0$ for $j = 1, 2, \dots, N$, their charge density can be described by

$$\rho_0(\mathbf{r}, z) = eZ \sum_{j=1}^N \delta(\mathbf{r} - \mathbf{r}_j) \delta(z - z_j). \quad (5.14)$$

The statistical properties of the impurity position distribution can be modeled by assuming translational invariance in the $\mathbf{r}$ directions and utilizing the one- and two-particle distribution functions [159]

$$F_1(\mathbf{r}, z) = \frac{N}{A} f_1(z), \quad (5.15)$$

and

$$F_2(\mathbf{r}_1, z_1; \mathbf{r}_2, z_2) = \frac{N(N-1)}{A^2} f_1(z_1) f_1(z_2) g(\mathbf{r}_2 - \mathbf{r}_1; z_1, z_2). \quad (5.16)$$

Here, $f_1(z)$ represents the distribution of particle positions in the perpendicular direction, being non-zero for $z \leq 0$ and normalized to one. Meanwhile, $g(\mathbf{r}; z_1, z_2)$ is associated with the standard *radial distribution* function, which approaches the limit $g \rightarrow 1$ as $r \equiv \|\mathbf{r}\| \rightarrow \infty$ [159]. Given these definitions, and assuming that the impurities are distributed isotropically in the directions parallel to phosphorene, the structure factor is finally obtained as

$$\begin{aligned} \mathcal{S}(q) = & \int_{-\infty}^0 dz f_1(z) e^{2qz} + 2\pi n_{\text{imp}} \int_{-\infty}^0 dz_1 f_1(z_1) e^{qz_1} \\ & \times \int_{-\infty}^0 dz_2 f_1(z_2) e^{qz_2} \int_0^{\infty} dr r J_0(qr) [g(r; z_1, z_2) - 1], \end{aligned} \quad (5.17)$$

where $J_0(\cdot)$ is a Bessel function of the zeroth order. A proof of this result is given in appendix D.

Modeling a complete three-dimensional (3D) structure of CIs in the substrate that involves correlation is quite challenging and to the best of our knowledge there is no closed form expression for the structure factor of such distributions (see Appendix B of Ref. 158). Therefore, we will focus on two special cases. First, an ensemble of spatially uncorrelated impurities is defined by setting $g = 1$ in Eq. (5.17), which reduces the structure

factor to the first term in Eq. (5.17), describing the impurity distribution along the z axis [101, 158]. In this scenario, we consider a uniform 3D distribution over a layer of finite thickness L in the substrate, as in Ref. 2, with $f_1(z) = 1/L$ for $-L \leq z \leq 0$, resulting in $\mathcal{S}(q) = (1 - e^{-2qL})/(2qL)$ from Eq. (5.17). Second, a more common model in the literature assumes a planar distribution of impurities at a constant depth $d \geq 0$ from the substrate surface, without spatial correlation among the impurities. This is achieved by setting $f_1(z) = \delta(z + d)$ and $g = 1$ in Eq. (5.17), yielding $\mathcal{S}(q) = e^{-2qd}$ [2, 158, 159].

However, we will investigate the impact of correlation among impurities within a planar distribution by setting $f_1(z) = \delta(z + d)$ and assuming that the radial distribution in Eq. (5.17) only depends on r in the form that arises from the hard-disk (HD) model, resulting in $\mathcal{S}(q) = e^{-2qd} S_{\text{HD}}(q)$. For the HD structure factor $S_{\text{HD}}(q)$, we use Rosenfeld's parametrization [157], given by

$$S_{\text{HD}}(q) = \left[1 + 16a \left(\frac{J_1(qr_c/2)}{qr_c} \right)^2 + 8b \frac{J_0(qr_c/2)J_1(qr_c/2)}{qr_c} + \frac{8p}{1-p} \frac{J_1(qr_c)}{qr_c} \right]^{-1}, \quad (5.18)$$

$$\text{with} \quad \begin{cases} a = 1 + x(2p - 1) + \frac{2p}{1-p} \\ b = x(1 - p) - 1 - \frac{3p}{1-p} \\ x = \frac{1+p}{(1-p)^3} \end{cases}, \quad (5.19)$$

where r_c is the interparticle correlation distance and $J_0(\cdot)$ and $J_1(\cdot)$ are Bessel functions of the order 0 and 1, respectively. A crucial parameter in the HD model is the packing fraction $p = \pi n_{\text{imp}} r_c^2/4$. This parameter will reveal deviations from the simple inverse proportionality between mobility and the average impurity density n_{imp} as r_c increases. Rosenfeld's parametrization of $S_{\text{HD}}(q)$ accurately represents the HD model for packing fractions up to $p \approx 0.69$ [157]. Note that in the HD model, charged particles are treated as point charges located at the centers of imaginary disks with a radius of $r_c/2$, ensuring that the minimum distance between any two charged impurities is r_c . A sample distribution of charged impurities according to the HD model is illustrated in Fig. 5.2.

For small packing fractions, the HD structure factor shows weak oscillations around the value $S_{\text{HD}}(q) = 1$, which represents uncorrelated impurities ($r_c = 0$). The peaks are located near $q_\ell = 2\pi\ell/r_c$, where $\ell = 1, 2, \dots$, corresponding to the ℓ th coordination shell of the HD model [157, 159]. As the packing fraction p increases, $S_{\text{HD}}(q)$ is characterized by a strong peak at q_1 , related to the first coordination shell, followed by heavily damped oscillations at subsequent coordination shells, eventually approaching $S_{\text{HD}}(q) = 1$ as $q \rightarrow \infty$. On

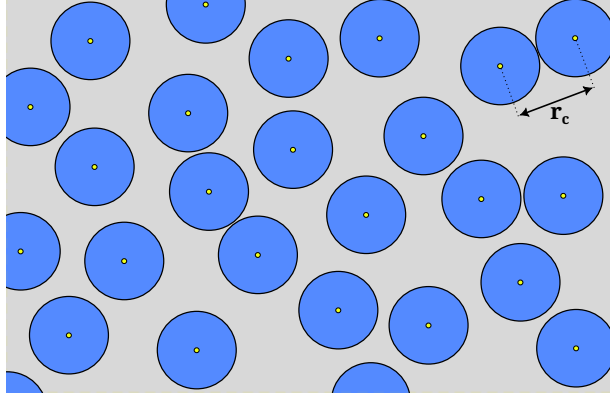


Figure 5.2: A sample distribution of the charged impurities based on the HD model. Blue disks represent the imaginary hard disks. Yellow dots at the center of the disks represent the point CIs.

the other hand, at long wavelengths, $q < q_1$, the magnitude of the HD structure factor significantly decreases with increasing packing fraction, reaching $S_{\text{HD}}(0) = (1-p)^3/(1+p)$ as $q \rightarrow 0$ [158]. In Fig. 5.3 the function $S_{\text{HD}}(q)$ is plotted for different packing fractions $p = 7.85 \times 10^{-5}$, 0.07, 0.5, and 0.63.

5.2.3 Mobility tensor

When Eq. (6.20) is substituted into Eq. (5.9) for $\langle P_{\text{imp}} \rangle$, the integrand includes a term $(\mathbf{q} \cdot \mathbf{v})^2$, which can be rewritten as $\mathbf{v} \cdot (\mathbf{q}\mathbf{q}) \cdot \mathbf{v}$. This reformulation allows the two velocity factors to be taken outside the integral, while the $(\mathbf{q}\mathbf{q})$ factor that is treated as the outer (or dyadic) product remains within the integral:

$$\iint d^2\mathbf{q} (\mathbf{q} \cdot \mathbf{v})^2 \Theta \equiv \mathbf{v} \cdot \left(\iint d^2\mathbf{q} (\mathbf{q}\mathbf{q}) \Theta \right) \cdot \mathbf{v}, \quad (5.20)$$

where Θ represents all other factors of the integrand except for $(\mathbf{q} \cdot \mathbf{v})^2$ and $\mathbf{v} \cdot (\mathbf{q}\mathbf{q}) \cdot \mathbf{v}$. Applying this re-arrangement in Eq. (5.9) gives an expression for $\langle P_{\text{imp}} \rangle$ that can be directly equated with the Joule power P_J in Eq. (5.1). Considering that the velocity $\mathbf{v} = -\mathbf{v}_d$ is *arbitrary*, one can deduce an expression for the inverse of the drift mobility tensor as follows

$$\overleftrightarrow{\mu}^{-1} = \frac{\hbar}{e} \frac{n_{\text{imp}}}{n} q_s^2 \iint \frac{d^2\mathbf{q}}{2\pi} (\hat{\mathbf{q}}\hat{\mathbf{q}}) \mathcal{S}(q) \left[\frac{\epsilon_{\text{bg}}(q)e^{-qh}}{\epsilon_{\text{bg}}^0 \epsilon_{\text{stat}}(\mathbf{q})} \right]^2 \frac{1}{Q} \frac{H(2k_F - Q)}{\sqrt{(2k_F)^2 - Q^2}}, \quad (5.21)$$

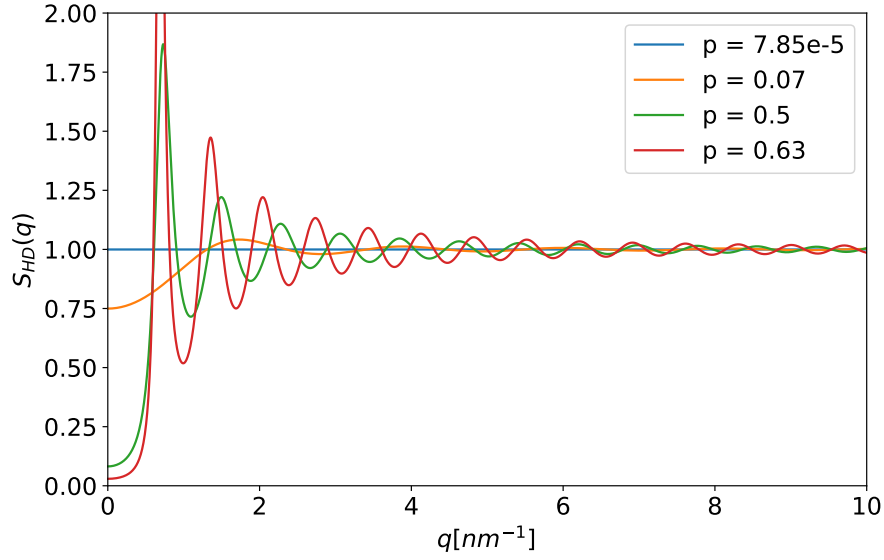


Figure 5.3: $S_{\text{HD}}(q)$ as a function of q for different values of the packing fraction. For small packing fractions this function is close to 1, whereas for large values of p it strongly oscillates. In this figure we set $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$.

where, $(\hat{\mathbf{q}}\hat{\mathbf{q}})$ is the outer product of two unit vectors $\hat{\mathbf{q}} = \mathbf{q}/q$, and Q is a function of the wavevector $\mathbf{q}$ as introduced in Eq. (3.42). This formula represents the core result of our ELM implementation, which serves as an alternative to the conventional BTE approach. Note that the BTE method typically involves numerically solving an integral equation for the direction-dependent MRT and then integrating this MRT to obtain numerical values for the mobility tensor components based on a given set of model parameters [2, 91, 101, 102].

The integration in Eq. (5.21) can be carried out using the polar coordinates, $\mathbf{q} = (q \cos \theta, q \sin \theta)$. By switching to a double angle, $\phi = 2\theta$, it becomes evident that the inverse mobility tensor is diagonal, with the components $1/\mu_{xx}$ and $1/\mu_{yy}$ representing the inverses of the mobilities along the AC and ZZ directions of phosphorene, respectively. Additionally, by transforming the wavenumber as $q = 2k_F \frac{\sin \gamma}{b(\phi)}$, where

$$b(\phi) = \sqrt{\frac{m_y + m_x}{2m_d} + \frac{m_y - m_x}{2m_d} \cos \phi}, \quad (5.22)$$

it is possible to express the inverse mobility components as double integrals over the angles γ and ϕ as

$$\frac{1}{\mu_{xx/yy}} = 4 \frac{\hbar}{e} n_{\text{imp}} \int_0^{\pi/2} d\gamma \int_0^\pi d\phi \mathcal{S}^* \left[\frac{\epsilon_{\text{bg}}^*}{\epsilon_0} e^{-2k_F h \frac{\sin \gamma}{b}} \right]^2 \frac{(1 \pm \cos \phi) \sin^2 \gamma}{(b^2 + \epsilon_{\text{bg}}^* b \nu \sin \gamma + a \nu^2 \sin^2 \gamma)^2} \quad (5.23)$$

where $\nu = 2k_F/q_s$. Note that $\mathcal{S}^*$ and ϵ_{bg}^* stand for functions of γ and ϕ that are obtained by substituting $q = 2k_F \frac{\sin \gamma}{b(\phi)}$ in the original functions $\mathcal{S}(q)$ and $\epsilon_{\text{bg}}(q)$, and a stands for the auxiliary function

$$a(\phi) = \pi q_s [\alpha_x + \alpha_y + (\alpha_x - \alpha_y) \cos \phi], \quad (5.24)$$

while the $\pm$ sign in Eq. (5.23) refers to the AC/ZZ direction.

5.3 Results

First, we will verify the validity of the formula in Eq. (5.21), which is equivalent with Eq. (5.23), by reproducing the results shown in Fig. 3 of Ref. 2 using the model parameters from that reference. Subsequently, we will use the parameters derived from the *ab initio* data of Ref. 1 for n-doped phosphorene with an electron density of $n = 10^{13} \text{ cm}^{-2}$ as given in table 4.1. While the electron effective masses vary slightly with doping density, they

remain nearly constant up to about $n = 5 \times 10^{13} \text{ cm}^{-2}$ [1], so we will treat m_x and m_y as constant over the range of n considered here. The hole effective masses show greater anisotropy and more significant variation with doping density within the same range [1], so we will focus on n-doped phosphorene to study the effects of variable doping density and different impurity distributions. Moreover, for the substrate, we use $\epsilon_s = 3.9$ to represent SiO_2 .

In Fig. 5.4, we applied Eq. (5.23) using the hole effective masses $m_x = 0.15 m_0$ and $m_y = m_0$ as used in Ref. 2, while setting $\alpha_x = \alpha_y = 0$ and $h = 0$ to be closest to their setup. We considered uncorrelated impurities ($r_c = 0$) that translates to the uniform distributions with an areal density of $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$, distributed either in a planar layer at three different depths or in a layer with a thickness of $L = 300 \text{ nm}$ within a SiO_2 substrate. Our mobility component results are shown as solid lines, while the curves from Fig. 3 of Ref. 2 are depicted as dashed lines. There is nearly complete agreement between the ELM and BTE methods, except for a slight discrepancy in the mobilities shown in panels (a) and (b) for the planar layer at finite depths of $d = 1$ and 2 nm . This discrepancy becomes more pronounced in the plots of the mobility ratio in panel (c) when the doping density exceeds $\sim 10^{13} \text{ cm}^{-2}$.

Note that the BTE approach involves an integral equation for the momentum relaxation times, which contains singularities in the integrand. Consequently, numerically solving this integral equation is challenging. In Ref. 2, the integral equation is converted into a system of linear equations by approximating the integrals with finite sums. This results in a strongly ill-conditioned system, which is solved using iterative methods. To address the source of the discrepancy between the BTE and ELM results, we solved the integral equation of the BTE method using the method of successive substitutions, which is a type of iteration. Although this method had a slow convergence rate, it yielded more accurate MRTs, resulting in mobilities that, while still differing from the ELM results, showed less deviation compared to the results from Ref. 2. This indicates that the ELM offers a promising and considerably simpler method for calculating the drift mobility in anisotropic 2D materials. In Fig. 5.5, we present the results from Ref. 2 (dashed lines) alongside our calculations using the BTE with more precise numerical methods (solid lines) and the ELM results (dotted lines).

It should be noted that, as carrier density rises, the mobility components increase as shown in Figs. 5.4a and 5.4b. However, the ratio of these mobilities decreases, as depicted in Fig. 5.4c. This reduction is due to enhanced screening of impurities from intraband electron transitions in phosphorene, which mitigates the asymmetry effects in the effective masses. For impurities at zero depth or distributed in a thick layer of the substrate, the ratio consistently decreases. In contrast, when impurities are in a planar layer at a finite

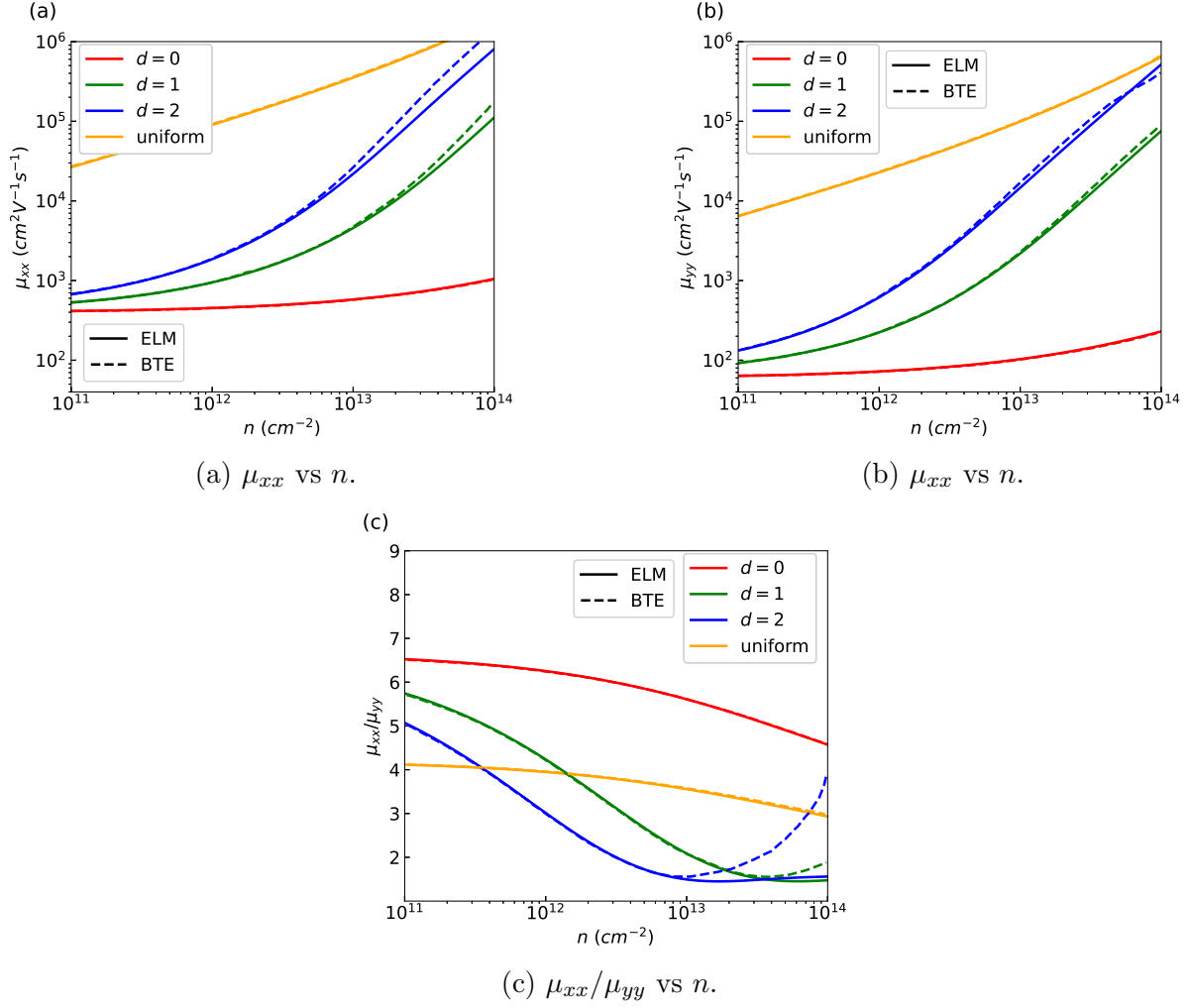
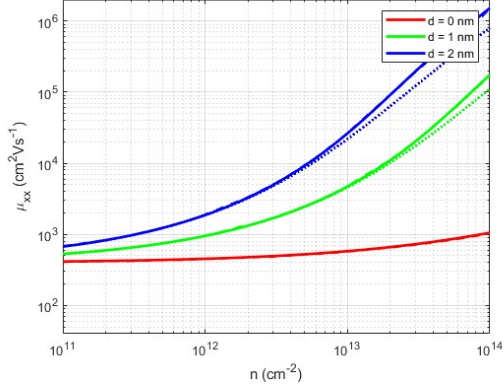
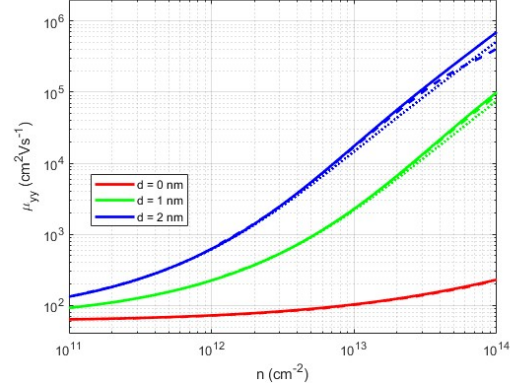


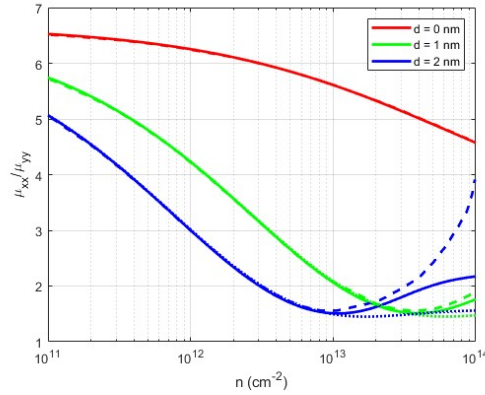
Figure 5.4: Mobilities along the (a) x -axis, (b) y -axis, and (c) their ratio versus carrier density n , calculated using the ELM (solid lines) and BTE (dashed lines) from Ref. 2. In all cases $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$ with zero correlation and a gap of $h = 0$. CIs are positioned in a plane at depths of $d = 0 \text{ nm}$ (red), $d = 1 \text{ nm}$ (green), and $d = 2 \text{ nm}$ (blue). The orange curves present the results for a uniform distribution of CIs across a layer with a thickness of $L = 300 \text{ nm}$ in an SiO_2 substrate. The model parameters used are consistent with those from Ref. 2.



(a) μ_{xx} vs n .



(b) μ_{yy} vs n .



(c) μ_{xx}/μ_{yy} vs n .

Figure 5.5: These curves are obtained with the same parameters as in Fig. 5.4. The dashed lines present the BTE results from Ref. 2, the solid lines present our results obtained from BTE approach with more precise numerical methods for solving the MRT integral equations, and the dotted lines present the ELM results.

depth, the ratio shows a minimum value, happening between $\sim 10^{13}$ and 10^{14} cm^{-2} for $d = 1$ and 2 nm.

In Fig. 5.6, we use effective electron masses from table 4.1 that are based on Ref. 1, and compare cases with $\alpha_x = \alpha_y = 0$ (solid lines) and $\alpha_x = 0.572$ nm, $\alpha_y = 0.488$ nm (dashed lines). We examine planar impurity distributions at three different depths with a gap $h = 0$, similar to Fig. 5.4. Comparing the solid lines from Figs. 5.4 and 5.6 highlights the impact of changing the effective masses from $m_x \approx 0.15 m_0$ and $m_y \approx m_0$ to $m_x \approx 0.133 m_0$ and $m_y \approx 1.119 m_0$. This effect is most apparent in the mobility ratios at $d = 0$ and the lowest density of $n = 10^{11}$ cm^{-2} .

Notably, Fig. 5.6 reveals that the influence of screening due to nonzero values of $\alpha_{x/y}$ from interband transitions grows with doping density. This effect is especially significant when the impurities are in the phosphorene plane (i.e., $d = 0$ and $h = 0$), causing the asymmetry ratio to decrease more rapidly with increasing carrier density. For instance, at $n = 10^{14}$ cm^{-2} , the ratio μ_{xx}/μ_{yy} drops to approximately 2.5, compared to about 5.5 when $\alpha_x = \alpha_y = 0$.

On the other hand, as the depth of the impurities increases, the effect of finite $\alpha_{x/y}$ becomes less significant. This can be understood by examining Eq. (5.21), where the structure factor $\mathcal{S}(q)$ includes a term $\exp(-2dq)$. This should be compared to the term involving $\alpha_{x/y}$ in the dielectric function from Eq. (5.10). The conclusion is that while both impurity depth screening and interband electron transition screening reduce the mobility asymmetry ratio as doping density increases, the former will dominate when the impurity depth d is roughly greater than or equal to $\pi \max(\alpha_x, \alpha_y) \approx 1.8$ nm due to the exponential factor $\exp(-2dq)$.

The relatively large thickness of the phosphorene layer can be modeled by considering a finite gap between the effective plane of charge density in the phosphorene sheet and the adjacent dielectric surface. Moreover, the van der Waals bond between the phosphorene and substrate has a typical length of ~ 0.3 to ~ 0.6 nm. Although modeling the transport of charge carriers in phosphorene is often assumed with zero gap between the phosphorene and the substrate [2,91], we investigate the impact of a finite gap h in Fig. 5.7 at a doping density of $n = 10^{13}$ cm^{-2} . The variation in mobilities is shown for different values of h , under the constraint $h + d = 1$ nm, thereby maintaining a constant 1 nm distance between charge carriers in phosphorene and uncorrelated impurities with a density of $n_{\text{imp}} = 10^{12}$ cm^{-2} in a planar layer. Although the dependence of the mobility ratio on h is not significant, it suggests that gap size should be considered in a detailed transport property modeling for phosphorene. Fig. 5.7 includes both cases with $\alpha_x = \alpha_y = 0$ (solid lines) and $\alpha_x = 0.572$ nm, $\alpha_y = 0.488$ nm (dashed lines), showing an offset between mobility curves comparable

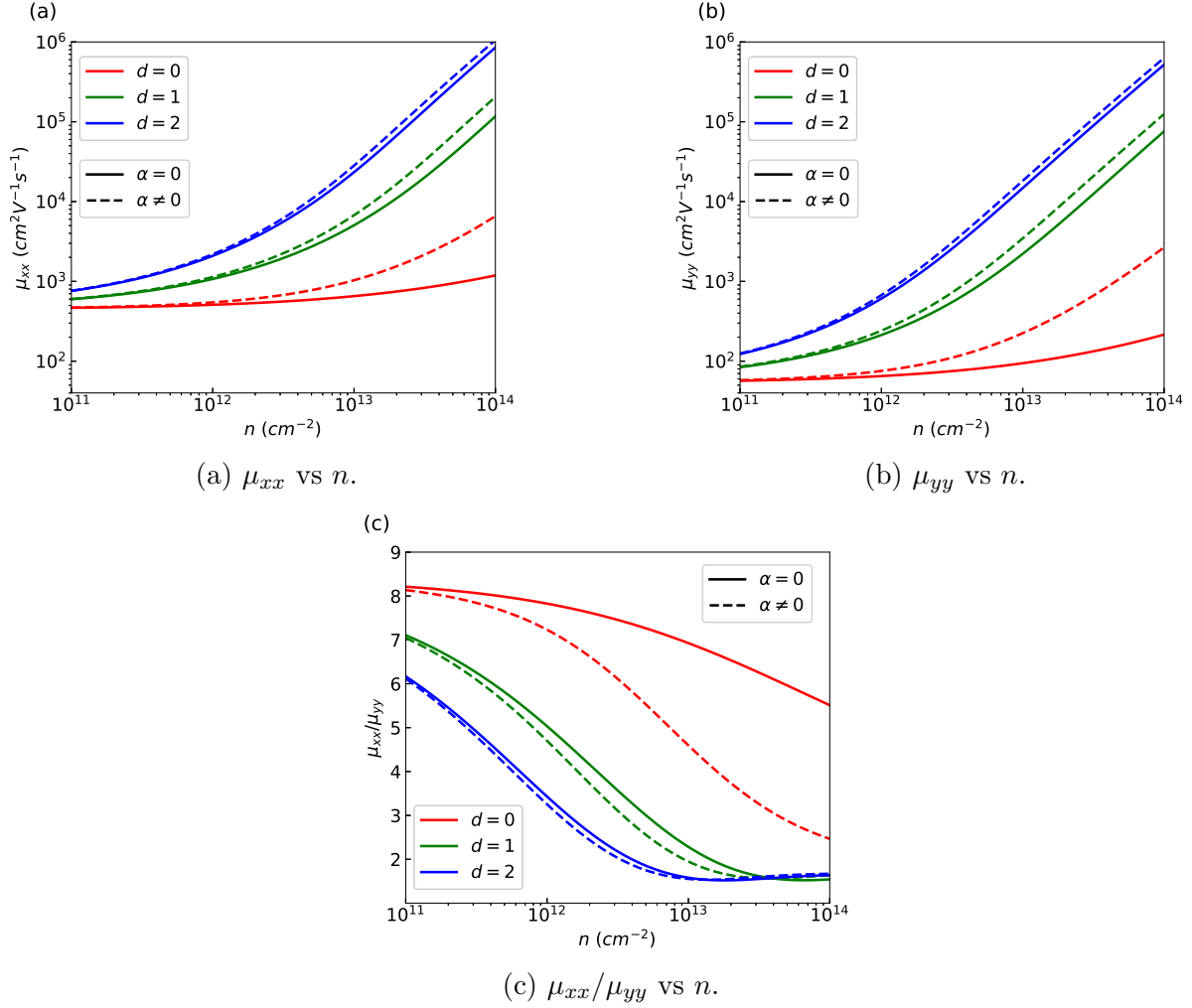
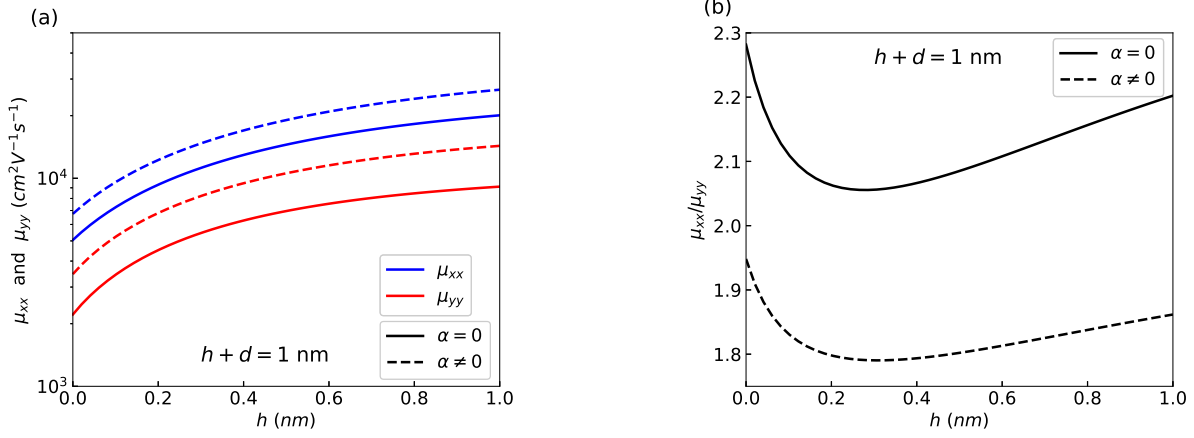


Figure 5.6: Mobilities along (a) the x -axis, (b) the y -axis, and (c) their ratio as a function of carrier density n . Results are shown for two cases: with $\alpha_x = \alpha_y = 0$ (solid lines) and with non-zero polarizabilities α_x and α_y (dashed lines). Impurities are distributed with a density of $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$ and no correlation ($r_c = 0$), at various depths: $d = 0$ (red), $d = 1$ nm (green), and $d = 2$ nm (blue). The calculations are conducted for a SiO_2 substrate with a gap $h = 0$. Parameters are based on Ref. 1 (see table 4.1).



(a) Mobilities along the x -axis (blue) and the y -axis (red) as a function of h .

(b) Mobility ratio $\frac{\mu_{xx}}{\mu_{yy}}$ as a function of h .

Figure 5.7: Solid curves correspond to $\alpha_x = \alpha_y = 0$, whereas dashed curves correspond to non-zero α_x and α_y from table 4.1. Phosphorene carrier density is fixed at $n = 10^{13} \text{ cm}^{-2}$ and the planar density of the non-correlated CIs is set to $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$. SiO_2 is served as the substrate, under the constraint $h + d = 1 \text{ nm}$.

to their variation with changing h .

In Fig. 5.8, we investigate the impact of impurity correlations at a density of $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$, with the impurities located in a plane at a depth of 1 nm in the SiO_2 substrate, with $\alpha_x = \alpha_y = 0$ and $h = 0$. The correlation distance r_c is varied in such a range that ensures the packing fraction remains below 0.69, hence the HD structure factor adopted in this study remains valid. Panels (a) and (b) display a significant spread in the mobility curves for different r_c values at doping densities roughly lower than n_{imp} . As the density increases beyond certain critical values that is distinct for the AC and ZZ directions the curves converge abruptly. This behavior can be explained by the constraint $Q(\mathbf{q}) \equiv qb(\phi) < 2k_F$ in Eq. (5.21) that is implied by the step function, and the structure factor $\mathcal{S}(q) = S_{\text{HD}}(q) \exp(-2qd)$, which involves the HD factor $S_{\text{HD}}(q)$. For small packing fractions, $S_{\text{HD}}(q)$ oscillates around 1, but is significantly reduced at wavenumbers $q < q_1 = 2\pi/r_c$ as p increases (see Fig. 5.3). Therefore, for doping densities such that $k_F r_c \lesssim \pi b(\phi)$, or $n \lesssim \frac{\pi}{2} [b(\phi)/r_c]^2 \equiv n_c(\phi)$, the mobility is expected to increase for larger r_c , as easily seen in the long-wavelength limit by $S_{\text{HD}}(0) = (1 - p)^3/(1 + p)$. On the other hand, once the doping density exceeds n_c , the $\mathbf{q}$ -integral in Eq. (5.21) is dominated by large q regions, where $S_{\text{HD}}(q)$ shows damped oscillations around 1. Consequently, the impact of finite

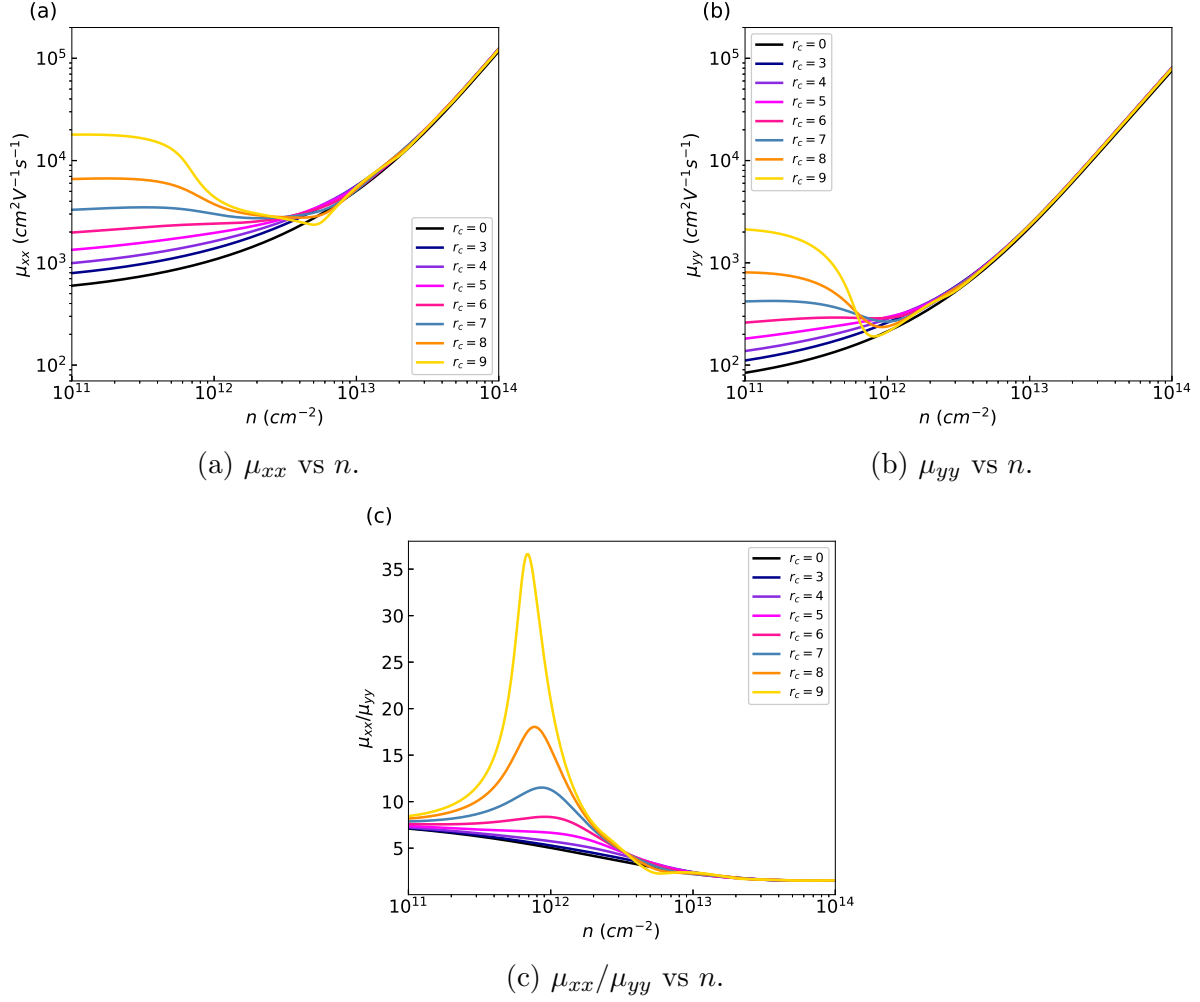


Figure 5.8: Mobilities along (a) the x -axis, (b) the y -axis, and (c) their ratio as a function of carrier density n for different correlation lengths $r_c = 0, 3, 4, 5, 6, 7, 8$, and 9 nm . In all cases $\alpha_x = \alpha_y = 0$ and impurities are distributed in the plane $z = -1 \text{ nm}$ with the density $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$. The calculations are conducted for a SiO_2 substrate with a gap $h = 0$. Parameters are based on Ref. 1 (see table 4.1).

correlation distances diminishes, and the mobility curves converge toward the uncorrelated impurity case as the density rises above n_c . The influence of the peak at q_1 is clearly seen at high packing fractions, as indicated by the weak local minima in the mobility curves for $r_c = 9$ nm in Figs. 5.8a and 5.8b. The location of these minima can be estimated by setting $\phi = 0$ (AC direction) and $\phi = \pi$ (ZZ direction) in $n_c(\phi)$, giving $n_c^{\text{AC}} \approx 5.6 \times 10^{12} \text{ cm}^{-2}$ and $n_c^{\text{ZZ}} \approx 6.7 \times 10^{11} \text{ cm}^{-2}$, closely matching the observed minima in the $r_c = 9$ nm curves in Figs. 5.8a and 5.8b.

In Fig. 5.8c, we observe that increasing values of r_c have a relatively minor impact on the mobility ratio, except around the doping densities n near the impurity density n_{imp} . Here, the ratio exhibits a broad peak whose height increases with r_c . This peak arises from different doping densities where the AC and ZZ mobility curves begin to converge. The weak dependence of the mobility ratio on r_c at low doping densities can be explained by the formula in Eq. (5.21). In this regime of the "complete screening", the integration in the $\mathbf{q}$ plane is limited to the small Q values by the step function $H(2k_F - Q)$, where the structure factor $\sim S_{\text{HD}}(0)$ is a common factor in both directions. Similarly, the decreasing influence of r_c at densities $n \gtrsim 10^{13} \text{ cm}^{-2}$ in Fig. 5.8c results from the convergence of the mobility curves towards those for uncorrelated impurities, as seen in Figs. 5.8a and 5.8b, for $n \gtrsim n_c^{\text{AC}}$ and $n \gtrsim n_c^{\text{ZZ}}$, respectively.

It is important to note that the results presented in Fig. 5.8, particularly the anisotropy ratio μ_{xx}/μ_{yy} that is predicted to exceed 35 at certain densities n (with $r_c = 9$), are potentially valuable. Achieving these results using the BTE approach typically requires Monte Carlo simulations with thousands of realizations of the CI configurations within the hard disk model, which is very expensive from a computational viewpoint.

In Fig. 5.9, we examine the impact of screening due to interband electron transitions, by comparing scenarios with $\alpha_x = \alpha_y = 0$ (solid curves) and $\alpha_x = 0.572 \text{ nm}$ and $\alpha_y = 0.488 \text{ nm}$ (dashed curves) for impurities with a density of $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$ and selected correlation distances r_c from Fig. 5.8. Panels (a) and (b) show that at low doping densities, the effects of impurity correlation dominate, whereas at high doping densities, the curves for different r_c converge into two groups, similar to Fig. 5.8. The group with finite $\alpha_{x/y}$ values exhibits higher mobility than the group with $\alpha_x = \alpha_y = 0$, consistent with Fig. 5.6. Additionally, finite $\alpha_{x/y}$ values lead to a slight reduction in the mobility ratio in panel (c) near the doping density corresponding to n_{imp} .

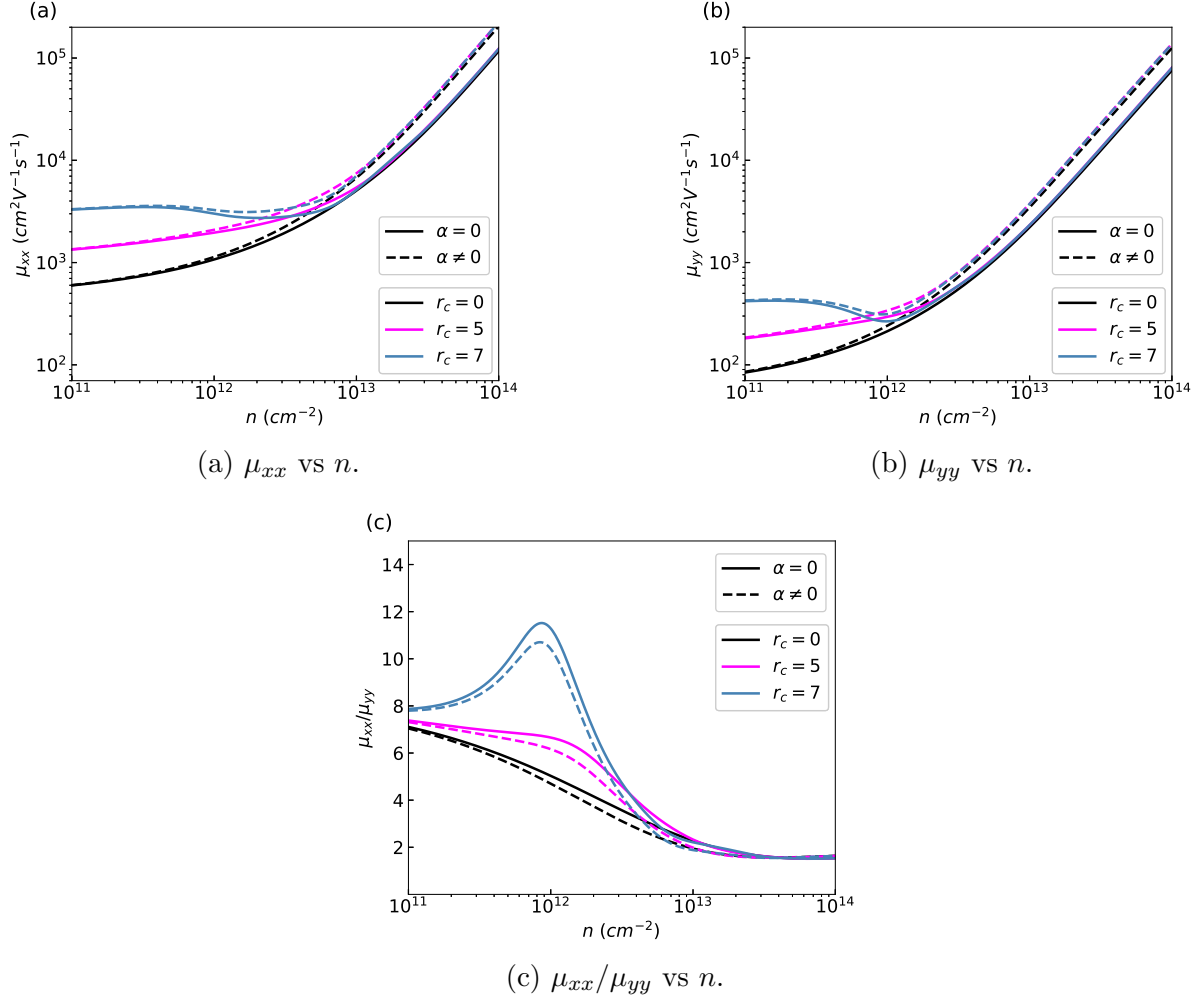


Figure 5.9: Mobilities along (a) the x -axis, (b) the y -axis, and (c) their ratio as a function of carrier density n for different correlation lengths $r_c = 0, 5$, and 7 nm. Solid curves correspond to $\alpha_x = \alpha_y = 0$ and dashed curves correspond to $\alpha_x = 0.572$ nm and $\alpha_y = 0.488$ nm. Impurities are distributed in the plane $z = -1$ nm with the density $n_{\text{imp}} = 10^{12} \text{ cm}^{-2}$. The calculations are conducted for a SiO_2 substrate with a gap $h = 0$. Parameters are based on Ref. 1 (see table 4.1).

Chapter 6

Sheet plasmon in graphene in the presence of random potential

6.1 Introduction

The existence of charged impurities is ubiquitous in the systems where graphene is supported by an insulating substrate (e.g., trapped charges in an oxide). Those impurities are randomly distributed in the substrate and may cause a nonuniform doping of graphene so that, close to the nominally neutral state of graphene, a system of electron-hole puddles may arise [160, 161], which are responsible for a finite minimum value of the DC conductivity in graphene [160, 162]. When a clean graphene sheet is doped with a relatively high concentration of electrons or holes by an external gate, a long-lived sheet plasmon mode arises with a characteristic square-root dispersion relation in the THz to MIR frequency range [160]. However, the presence of charged impurities in the oxide separating the gate from graphene still gives rise to a ground state with the dominant charge carriers exhibiting a randomly nonuniform density across its surface. That density gives rise to a random electrostatic potential landscape in the plane of graphene, which is controlled by the static screening due to the graphene itself and by the nearby dielectrics. At the same time, such a potential landscape provides strong disorder that can break up the coherence of the plasmon mode, thereby causing its decay, which is manifested in the broadening of its dispersion relation [163, 164].

In this chapter, we attempt to estimate the sheet plasmon broadening in graphene for a realistic set of parameters by using a model for its dynamical conductivity that is suitable in the THz-MIR frequency range and a stochastic description of the random electrostatic

potential landscape in the ground state without going into details of its microscopic origins [165,166]. Similar effects of disorder are observed in the broadening of surface plasmon modes on metallic surfaces that generally exhibit various degrees of roughness [118]. While a single sheet of graphene can exhibit a significant deviation from a perfect plane, especially when deposited on an oxide surface [117], we shall ignore the effects of roughness, but rather focus on the random electrostatic potential landscape in the plane of graphene by adapting the so-called “smoothing method” [119] that was used in Ref. [118]. This method is powerful enough to treat the effects of disorder nonperturbatively in an analytical manner, giving an eigenvalue problem for the sheet plasmon frequency in the presence of disorder in graphene. However, the analytical formulation of the problem involves a highly nonlinear integral equation that presents a substantial computational challenge, which we have overcome successfully.

6.2 Derivation of the eigenvalue problem

We consider a large sheet of doped graphene that occupies the $z = 0$ plane of a Cartesian coordinate system with an insulating substrate underneath graphene. The substrate contains an ensemble of stationary charged impurities that occupy randomly distributed positions, giving rise to a static potential $\phi_0(\mathbf{r})$ in the plane of graphene, which is a random function of the in-plane position $\mathbf{r} = (x, y)$. Statistical properties of that potential were studied theoretically in Ref. [166], yielding an autocovariance that was found to be in close agreement with the experimental observations of Ref. [165].

In the so-called rigid-band approximation, the potential landscape $\phi_0(\mathbf{r})$ causes a spatial fluctuation of the Dirac energy level in graphene’s π electron bands across its surface, so that a position-dependent Fermi energy $\varepsilon_F(\mathbf{r})$ may be defined to describe the local doping density of electrons (or holes) in graphene. This energy varies about some mean value, $\bar{\varepsilon}_F = \hbar v_F \bar{k}_F$, with $v_F \approx c/300$ being the Fermi speed of graphene’s π electron bands near the Dirac point and the Fermi wavenumber, $\bar{k}_F = \sqrt{\pi \bar{n}}$, encoding the mean (or nominal) doping density of graphene. The random function $\varepsilon_F(\mathbf{r})$ has the same autocovariance as $e\phi_0(\mathbf{r})$, which we model here by a Gaussian,

$$C_F(\mathbf{r} - \mathbf{r}') = \langle \Delta\varepsilon_F(\mathbf{r}) \Delta\varepsilon_F(\mathbf{r}') \rangle = \sigma_F^2 e^{-(\mathbf{r}-\mathbf{r}')^2/\ell_c^2}, \quad (6.1)$$

where $\Delta\varepsilon_F(\mathbf{r}) = \varepsilon_F(\mathbf{r}) - \bar{\varepsilon}_F$ is the fluctuating part of the Fermi energy, σ_F is its (local) standard deviation, and the parameter ℓ_c defines the correlation distance of the potential fluctuations across the surface of graphene. The parameters σ_F and ℓ_c in Eq. (6.1) can

be related to microscopic modeling of the impurities [166] or may be deduced from the experimental data [165]. We note that the choice of the Gaussian autocovariance is not essential for the development of the theory, but it helps in making analytical progress. Notice that the above model of impurities rests on the assumption of their translational invariance and isotropy in the directions of the in-plane positions $\mathbf{r}$. Those assumptions, along with our consideration of an isotropic 2D material, such as graphene, and the use of the Gaussian autocovariance, make it possible to derive a highly nonlinear integral equation in one variable for the plasmon self-energy in the presence of a random ground-state potential $\phi_0(\mathbf{r})$ caused by the impurities.

Some typical values of the relevant parameters are: $\bar{n} = 8 \times 10^{12} \text{ cm}^{-2}$ giving $\bar{k}_F \approx 0.5 \text{ nm}^{-1}$ and $\bar{\epsilon}_F \approx 0.33 \text{ eV}$ for graphene [164] and $\sigma_F \sim 0.05 \text{ eV}$ and $\ell_c \sim 20 \text{ nm}$ for impurities [165, 166].

To study the dynamic polarization of charge carriers in such an inhomogeneously doped graphene sheet, we assume that a weak, time-dependent electrostatic potential is applied externally, $\Phi_{\text{ext}}(\mathbf{r}, z, t)$, giving rise to a time-dependent perturbation of the carrier density, $\delta n(\mathbf{r}, t)$, which gives rise to a perturbation of the total in-plane electrostatic potential, $\delta\phi(\mathbf{r}, t)$, about the potential landscape $\phi_0(\mathbf{r})$. Performing a Fourier transform with respect to time,

$$\delta\phi(\mathbf{r}, \omega) = \int_{-\infty}^{\infty} dt e^{i\omega t} \delta\phi(\mathbf{r}, t),$$

the perturbation of the potential may be obtained from the integral equation [167–169]

$$\delta\phi(\mathbf{r}, \omega) = \Phi_{\text{ext}}(\mathbf{r}, 0, \omega) - e \iint d^2\mathbf{r}' G_0(\mathbf{r} - \mathbf{r}') \delta n(\mathbf{r}', \omega), \quad (6.2)$$

where $G_0(\mathbf{r} - \mathbf{r}')$ denotes the in-plane ($z = z' = 0$) value of the Green's function for the Poisson equation, which we shall approximate by only considering the dielectric background materials in the absence of graphene. A further progress can be made by invoking the in-plane continuity equation for charge carriers in graphene,

$$\delta n(\mathbf{r}, \omega) = \frac{i}{e\omega} \nabla_{\mathbf{r}} \cdot \delta \mathbf{j}(\mathbf{r}, \omega), \quad (6.3)$$

where the in-plane current fluctuation may be expressed via the Ohm's law in local form as

$$\delta \mathbf{j}(\mathbf{r}, \omega) = -\sigma(\mathbf{r}, \omega) \nabla_{\mathbf{r}} \delta\phi(\mathbf{r}, \omega). \quad (6.4)$$

Here, $\sigma(\mathbf{r}, \omega)$ is the conductivity due to intraband electronic transitions in doped graphene, which may be adequately modeled in the limit of high frequencies and long wavelengths

by the Drude model (see section 3.3)

$$\sigma(\mathbf{r}, \omega) = \frac{i}{\pi} \frac{e^2}{\hbar^2} \frac{\varepsilon_F(\mathbf{r})}{\omega + i\gamma}, \quad (6.5)$$

which is locally controlled by the position-dependent Fermi energy, whereas γ is a phenomenological damping constant. We note that a similar position-dependent local model of graphene AC conductivity was used to calculate localized plasmon modes on graphene flakes of finite area [167–169]. An analogous approach was also adopted for calculations of the stopping force on metallic surfaces [170] and in the bulk of a material [171], where the dynamic response of an inhomogeneous electron gas in those structures was treated in the so-called local plasma approximation.

Finally, by combining Eqs. (6.2), (6.3), (6.4) and (6.5), we obtain a self-consistent equation for the perturbation of the total potential,

$$\delta\phi(\mathbf{r}, \omega) = \Phi_{\text{ext}}(\mathbf{r}, 0, \omega) - \frac{e^2}{\pi\hbar^2} \frac{1}{\omega(\omega + i\gamma)} \iint d^2\mathbf{r}' G_0(\mathbf{r} - \mathbf{r}') \nabla_{\mathbf{r}'} \cdot [\varepsilon_F(\mathbf{r}') \nabla_{\mathbf{r}'} \delta\phi(\mathbf{r}', \omega)].$$

The right-hand-side of the above equation may be transformed using one of the Green's identities for the large-area graphene, giving

$$\delta\phi(\mathbf{r}, \omega) = \Phi_{\text{ext}}(\mathbf{r}, 0, \omega) + \frac{e^2}{\pi\hbar^2} \frac{1}{\omega(\omega + i\gamma)} \iint d^2\mathbf{r}' \varepsilon_F(\mathbf{r}') [\nabla_{\mathbf{r}'} \delta\phi(\mathbf{r}', \omega)] \cdot \nabla_{\mathbf{r}'} G_0(\mathbf{r} - \mathbf{r}'). \quad (6.6)$$

We may further perform a 2D Fourier transform with respect to the spatial coordinates, e.g.,

$$\delta\phi(\mathbf{q}, \omega) = \iint d^2\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \delta\phi(\mathbf{r}, \omega),$$

allowing us to convert Eq. (6.6) to the following form in the Fourier domain

$$\delta\phi(\mathbf{q}, \omega) = \Phi_{\text{ext}}(\mathbf{q}, 0, \omega) + \frac{e^2}{\pi\hbar^2} \frac{G_0(q)}{\omega(\omega + i\gamma)} \iint \frac{d^2\mathbf{q}'}{(2\pi)^2} \mathbf{q}' \cdot \mathbf{q} \varepsilon_F(\mathbf{q} - \mathbf{q}') \delta\phi(\mathbf{q}', \omega). \quad (6.7)$$

Expressing the Fermi energy in the Fourier domain as

$$\varepsilon_F(\mathbf{q}) = (2\pi)^2 \delta(\mathbf{q}) \bar{\varepsilon}_F + \Delta\varepsilon_F(\mathbf{q}), \quad (6.8)$$

allows us to rewrite Eq. (6.7) as

$$\epsilon_{2D}(q, \omega) \delta\phi(\mathbf{q}, \omega) = \Phi_{\text{ext}}(\mathbf{q}, 0, \omega) + \frac{e^2}{\pi\hbar^2} \frac{G_0(q)}{\omega(\omega + i\gamma)} \iint \frac{d^2\mathbf{q}'}{(2\pi)^2} \mathbf{q}' \cdot \mathbf{q} \Delta\varepsilon_F(\mathbf{q} - \mathbf{q}') \delta\phi(\mathbf{q}', \omega), \quad (6.9)$$

where

$$\epsilon_{2D}(q, \omega) = 1 - \frac{e^2}{\pi \hbar^2} \bar{\epsilon}_F \frac{q^2 G_0(q)}{\omega(\omega + i\gamma)} \quad (6.10)$$

is the effective 2D dielectric permittivity of a homogeneously doped graphene with the electron (or hole) density $\bar{n}$.

If we set the external potential to be zero, $\Phi_{\text{ext}}(\mathbf{q}, 0, \omega) = 0$, then Eq. (6.9) becomes an eigenvalue problem, written as a homogeneous integral equation for the potential $\delta\phi(\mathbf{q}, \omega)$,

$$\epsilon_{2D}(q, \omega) \delta\phi(\mathbf{q}, \omega) = \frac{e^2}{\pi \hbar^2} \frac{G_0(q)}{\omega(\omega + i\gamma)} \iint \frac{d^2 \mathbf{q}'}{(2\pi)^2} \mathbf{q}' \cdot \mathbf{q} \Delta\epsilon_F(\mathbf{q} - \mathbf{q}') \delta\phi(\mathbf{q}', \omega). \quad (6.11)$$

This equation has to be solved in the presence of the fluctuating part of the Fermi energy with zero mean, $\langle \Delta\epsilon_F(\mathbf{q}) \rangle = 0$, and the autocovariance given in the Fourier domain by

$$\langle \Delta\epsilon_F(\mathbf{q}) \Delta\epsilon_F(\mathbf{q}') \rangle = (2\pi)^2 \delta(\mathbf{q} + \mathbf{q}') C_F(q), \quad (6.12)$$

where $C_F(q)$ is the Fourier transform of the Gaussian correlation function in Eq. (6.1), given by

$$C_F(q) = \pi \sigma_F^2 \ell_c^2 e^{-\ell_c^2 q^2/4}. \quad (6.13)$$

When fluctuations are absent, $\Delta\epsilon_F(\mathbf{q}) = 0$, a nontrivial solution of Eq (6.11) requires that $\epsilon_{2D}(q, \omega) = 0$. Using an expression for the Green's function in the Fourier domain $G_0(q) = \frac{2\pi}{\epsilon_{\text{bg}} q}$, where $\epsilon_{\text{bg}} = (\epsilon_{\text{sub}} + 1)/2$ corresponds to graphene on a substrate with the dielectric permittivity ϵ_{sub} and zero gap between them, we obtain the familiar form of the 2D permittivity [168]

$$\epsilon_{2D}(q, \omega) = 1 - \frac{\alpha q}{\omega(\omega + i\gamma)}, \quad (6.14)$$

where $\alpha = \frac{2}{\epsilon_{\text{bg}}} v_B v_F \bar{k}_F$ with $v_B = e^2/\hbar \approx c/137$ being the Bohr speed. Thus, the condition $\epsilon_{2D}(q, \omega) = 0$ gives the familiar square-root dispersion relation of a sheet plasmon in a homogeneously doped graphene, given in the absence of damping by $\omega = \sqrt{\alpha q}$.

Our goal is to convert Eq. (6.11) into an effective eigenvalue problem for the expectation value of the potential, $\langle \delta\phi(\mathbf{q}, \omega) \rangle$, defined by taking an average over the ensemble of all realizations of the fluctuating part of the Fermi energy, $\Delta\epsilon_F(\mathbf{r})$. To that effect, we use the “smoothing method” due to Maradudin [119], which was successfully applied to calculate the broadening of the dispersion relation of the surface plasmon on a randomly rough metallic surface [118].

6.3 Smoothing method

In this section, we outline the main steps of Maradudin's smoothing method, noting that its details are presented in Ref. [119]. The integral equation in Eq. (6.11) for $\delta\phi(\mathbf{q}, \omega)$ may be written compactly in an operator form $(H - V)\delta\phi = 0$, or in the momentum (i.e., wavevector) representation as

$$\iint \frac{d^2\mathbf{p}}{(2\pi)^2} [H(\mathbf{q}, \mathbf{p}) - V(\mathbf{q}, \mathbf{p})] \delta\phi(\mathbf{p}, \omega) = 0, \quad (6.15)$$

where

$$H(\mathbf{q}, \mathbf{p}) = (2\pi)^2 \delta(\mathbf{p} - \mathbf{q}) \epsilon_{2D}(q, \omega) \quad (6.16)$$

describes a non-random or deterministic term, while

$$V(\mathbf{q}, \mathbf{p}) = \frac{1}{\omega(\omega + i\gamma)} \frac{\alpha}{q} \mathbf{q} \cdot \mathbf{p} \Delta\tilde{\epsilon}_F(\mathbf{q} - \mathbf{p}), \quad (6.17)$$

describes the random term of the stochastic eigenvalue equation for the potential $\delta\phi$, in which we have introduced the scaled fluctuation of the Fermi energy as $\Delta\tilde{\epsilon}_F = \Delta\epsilon_F/\bar{\epsilon}_F$. Note that, while one may expect $|\Delta\tilde{\epsilon}_F| \ll 1$ for heavily doped graphene, it is essential to treat the random term V in Eq. (6.15) nonperturbatively because V may not be small compared to the deterministic term H in the regions of frequencies and wavenumbers close to the plasmon condition, where one expects $\epsilon_{2D}(q, \omega) \approx 0$ and hence $H \approx 0$ [119].

As a result of the smoothing method, the stochastic eigenvalue problem in Eq. (6.15) is converted to a deterministic one for $\langle\delta\phi(\mathbf{p}, \omega)\rangle$

$$\iint \frac{d^2\mathbf{p}}{(2\pi)^2} [H(\mathbf{q}, \mathbf{p}) - \langle M(\mathbf{q}, \mathbf{p}) \rangle] \langle\delta\phi(\mathbf{p}, \omega)\rangle = 0, \quad (6.18)$$

with the random term $V(\mathbf{q}, \mathbf{p})$ being replaced by the expectation value, $\langle M(\mathbf{q}, \mathbf{p}) \rangle$, of the self-energy M , which satisfies the operator equation $M = V + M\langle\mathcal{G}\rangle(V - \langle M \rangle)$, where $\langle\mathcal{G}\rangle$ is the expectation value of the Green's operator $\mathcal{G} = (H - V)^{-1}$ for Eq. (6.15). The smoothing method shows that the average of the Green's function for Eq. (6.15) may be expressed as the Green's function of Eq. (6.18), given in an operator form by $\langle\mathcal{G}\rangle = (H - \langle M \rangle)^{-1}$ [119]. Writing in the momentum representation $\langle M(\mathbf{q}, \mathbf{p}) \rangle = (2\pi)^2 \delta(\mathbf{p} - \mathbf{q}) m(q, \omega)$, where $m(q, \omega)$ is a memory function that needs to be determined, one obtains

$$\langle\mathcal{G}\rangle = \frac{(2\pi)^2 \delta(\mathbf{p} - \mathbf{q})}{\epsilon_{2D}(q, \omega) - m(q, \omega)}, \quad (6.19)$$

so that the loss function of the 2D plasmonic system in the presence of disorder may be written as

$$L(q, \omega) = \Im \left\{ \frac{-1}{\epsilon_{2D}(q, \omega) - m(q, \omega)} \right\}. \quad (6.20)$$

This function will be used to illustrate that both shift and broadening may arise in the plasmon resonance in graphene in the presence of a disordered potential landscape. This broadening can be responsible for substantial damping, or attenuation of the sheet plasmon, even when the phenomenological damping parameter is absent, i.e., $\gamma = 0$. Whereas this parameter is often modeled by a constant, our results for the memory function $m(q, \omega)$ will show that its real part will be responsible for shifting, and the imaginary part for broadening of the plasmon resonance in a manner that is dependent on both the frequency and the wavenumber.

A progress towards determining the memory function is made by solving the above operator equation for M iteratively to the second-order in the fluctuating term, $\langle M \rangle \approx \langle V \langle \mathcal{G} \rangle V \rangle$. This approximation gives in the momentum representation, using Eqs. (6.17) and (6.19)

$$\begin{aligned} (2\pi)^2 \delta(\mathbf{p} - \mathbf{q}) m(q, \omega) &= \iint \frac{d^2 \mathbf{p}'}{(2\pi)^2} \iint \frac{d^2 \mathbf{q}'}{(2\pi)^2} \frac{\frac{\alpha}{q} \mathbf{q} \cdot \mathbf{p}'}{\omega(\omega + i\gamma)} \frac{\frac{\alpha}{q'} \mathbf{q}' \cdot \mathbf{p}}{\omega(\omega + i\gamma)} \\ &\times \frac{(2\pi)^2 \delta(\mathbf{p}' - \mathbf{q}')}{\epsilon_{2D}(q', \omega) - m(q', \omega)} \langle \Delta \tilde{\epsilon}_F(\mathbf{q} - \mathbf{p}') \Delta \tilde{\epsilon}_F(\mathbf{q}' - \mathbf{p}) \rangle. \end{aligned} \quad (6.21)$$

Upon using Eq. (6.12) and cancelling the factors $(2\pi)^2 \delta(\mathbf{p} - \mathbf{q})$ on both sides of the above equation, we finally obtain an integral equation for the memory function

$$m(q, \omega) = \frac{1}{[\omega(\omega + i\gamma)]^2} \frac{\alpha}{q} \iint \frac{d^2 \mathbf{p}}{(2\pi)^2} \frac{\alpha}{p} \frac{(\mathbf{p} \cdot \mathbf{q})^2}{\epsilon_{2D}(p, \omega) - m(p, \omega)} \tilde{C}_F(\mathbf{q} - \mathbf{p}), \quad (6.22)$$

where $\tilde{C}_F = C_F / \tilde{\epsilon}_F^2$ with C_F defined in Eq. (6.13). The above equation may be written more explicitly by defining the scaled memory function as $\mu(q, \omega) = \omega(\omega + i\gamma) m(q, \omega)$, which gives

$$\mu(q, \omega) = \frac{\alpha}{q} \iint \frac{d^2 \mathbf{p}}{(2\pi)^2} \frac{\alpha}{p} \frac{(\mathbf{p} \cdot \mathbf{q})^2}{\omega(\omega + i\gamma) - \alpha p - \mu(p, \omega)} \tilde{C}_F(\mathbf{q} - \mathbf{p}). \quad (6.23)$$

Performing the integration over $\mathbf{p}$ in polar coordinates, one can take advantage of the Gaussian form of C_F in Eq. (6.13) and perform the integration over the polar angle to

finally get the nonlinear integral equation in one variable,

$$\tilde{\mu}(\tilde{q}, \tilde{\omega}) = \frac{s}{8} \tilde{\ell}_c^2 \tilde{q} \int_0^\infty d\tilde{p} \frac{\tilde{p}^2 e^{-\frac{1}{4}\tilde{\ell}_c^2(\tilde{p}^2 + \tilde{q}^2)}}{\tilde{\omega}(\tilde{\omega} + i\tilde{\gamma}) - \tilde{\alpha}\tilde{p} - \tilde{\mu}(\tilde{p}, \tilde{\omega})} \left[I_0\left(\frac{1}{2}\tilde{\ell}_c^2\tilde{p}\tilde{q}\right) - \frac{2}{\tilde{\ell}_c^2\tilde{p}\tilde{q}} I_1\left(\frac{1}{2}\tilde{\ell}_c^2\tilde{p}\tilde{q}\right) \right], \quad (6.24)$$

where I_0 and I_1 are the modified Bessel functions of the first kind. In the above equation, we have introduced the parameter $s = (2\tilde{\alpha}\tilde{\sigma}_F)^2$, along with the scaled quantities $\tilde{\alpha} = \frac{2}{\epsilon_{bg}} \frac{v_B}{v_F}$, $\tilde{\sigma}_F = \sigma_F/\bar{\epsilon}_F$, $\tilde{\ell}_c = \ell_c \bar{k}_F$, $\tilde{q} = q/\bar{k}_F$, $\tilde{p} = p/\bar{k}_F$, $\tilde{\omega} = \hbar\omega/\bar{\epsilon}_F$, $\tilde{\gamma} = \hbar\gamma/\bar{\epsilon}_F$ and $\tilde{\mu} = \hbar^2\mu/\bar{\epsilon}_F^2$, which in terms of the original memory function reads $\tilde{\mu} = \frac{\hbar^2}{\bar{\epsilon}_F^2} \omega(\omega + i\gamma) m$.

6.4 Results

In this section we presents the numerical results for the energy loss function

$$L(\tilde{q}, \tilde{\omega}) = \Im \left\{ \frac{-\tilde{\omega}(\tilde{\omega} + i\tilde{\gamma})}{\tilde{\omega}(\tilde{\omega} + i\tilde{\gamma})\epsilon_{2D}(\tilde{q}, \tilde{\omega}) - \tilde{\mu}(\tilde{q}, \tilde{\omega})} \right\} = \Im \left\{ \frac{-1}{\epsilon_{2D}(\tilde{q}, \tilde{\omega}) - m(\tilde{q}, \tilde{\omega})} \right\}, \quad (6.25)$$

where

$$\epsilon_{2D}(\tilde{q}, \tilde{\omega}) = 1 - \frac{\tilde{\alpha}\tilde{q}}{\tilde{\omega}(\tilde{\omega} + i\tilde{\gamma})}, \quad (6.26)$$

and $\tilde{\mu}(\tilde{q}, \tilde{\omega})$ is the solution to the non-linear integral equation introduced in Eq. (6.24). Note that solving Eq. (6.24) is quite challenging due to the non-linear dependence of the integrand on the unknown function $\tilde{\mu}(\tilde{q}, \tilde{\omega})$. We developed a numerical method that was successful to solve this non-linear equation using a modified iteration technique that we call double iteration scheme (DIS). In this technique a sequence of functions $\tilde{\mu}_n(\tilde{q}, \tilde{\omega})$ is generated from the recursive equation

$$\tilde{\mu}_n(\tilde{q}, \tilde{\omega}) = \mathcal{T}[\tilde{\mu}_{n-1}(\tilde{q}, \tilde{\omega})], \quad (6.27)$$

where $\mathcal{T}[\cdot] : C(\mathbb{C}^2) \rightarrow C(\mathbb{C}^2)$ represents a transformation in the space of continuous functions of two complex variables, $C(\mathbb{C}^2)$. The precise definition of the transformation $\mathcal{T}$ and the details of the DIS is provided in appendix E. In our computation we start with the initial guess $\tilde{\mu}_0(\tilde{q}, \tilde{\omega}) = 1$ and use Eq. (6.27) to generate the sequence of functions. Although it is not easy to prove that $\mathcal{T}$ is a contraction on the underlying space, this iteration scheme converges to a function and it is easy to show that the limiting function satisfying

$\tilde{\mu}(\tilde{q}, \tilde{\omega}) = \mathcal{T}[\tilde{\mu}(\tilde{q}, \tilde{\omega})]$ is in fact the solution of the integral equation Eq. (6.24). Moreover, for the convergence criterion, we continue the iteration process until for some N

$$\left[\frac{1}{\tilde{p}_{\max}} \int_0^{\tilde{p}_{\max}} \left| \tilde{\mu}_N(\tilde{q}, \tilde{\omega}) - \tilde{\mu}_{N-1}(\tilde{q}, \tilde{\omega}) \right|^2 d\tilde{q} \right]^{\frac{1}{2}} \leq \varepsilon_{\text{cut}}, \quad (6.28)$$

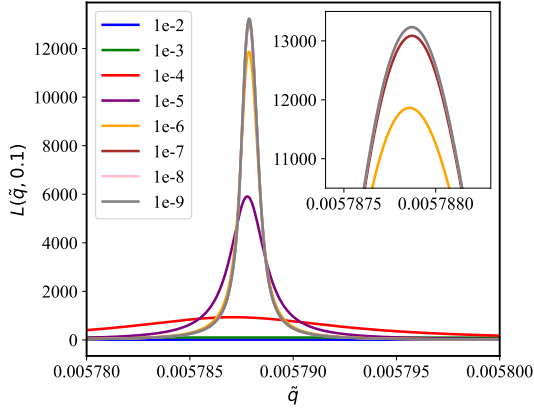
with $\varepsilon_{\text{cut}} = 10^{-8}$; then we set $\tilde{\mu}(\tilde{q}, \tilde{\omega}) = \tilde{\mu}_N(\tilde{q}, \tilde{\omega})$ as the solution of the Eq. (6.24). Note that in Eq. (6.24), $\tilde{\omega}$ is not affected by the integral, meaning that in each computation $\tilde{\omega}$ can be treated as a constant. Furthermore, in our computations the upper limit of the integral in Eq. (6.24), is approximated by $\tilde{p}_{\max} = \max(\frac{10}{\ell_c}, 3)$ that ensures a prefactor of at most $e^{-25} \approx 1.39 \times 10^{-11}$ in the integrand at the regions which are excluded from the integral.

First, we study the effect of the phenomenological damping parameter $\tilde{\gamma}$ on the energy loss function $L(\tilde{q}, \tilde{\omega})$. Note that, in the absence of the random potential ($\tilde{\sigma}_F = 0$), $\tilde{\mu}(\tilde{q}, \tilde{\omega}) = 0$ is the solution of Eq. (6.24) and the energy loss function reduces to

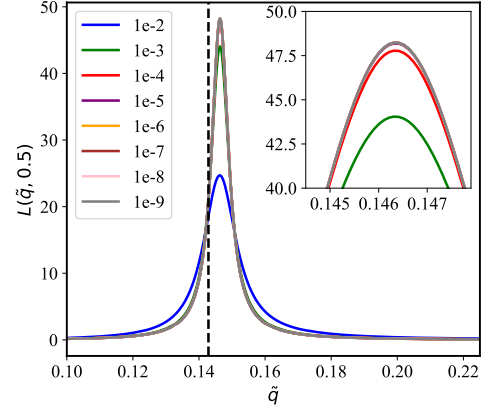
$$L(\tilde{q}, \tilde{\omega}) = \Im \left\{ \frac{-1}{\epsilon_{2D}(\tilde{q}, \tilde{\omega})} \right\} = \frac{\tilde{\alpha}\tilde{q}}{\tilde{\omega}} \frac{\tilde{\gamma}}{\left(1 - \frac{\tilde{\alpha}\tilde{q}}{\tilde{\omega}^2}\right)^2 + \tilde{\gamma}^2} \xrightarrow{\text{as } \tilde{\gamma} \rightarrow 0} \frac{\pi\tilde{\alpha}\tilde{q}}{\tilde{\omega}} \delta\left(1 - \frac{\tilde{\alpha}\tilde{q}}{\tilde{\omega}^2}\right), \quad (6.29)$$

converging to a sharp δ -like peak on the dispersion curve $\tilde{\omega} = \sqrt{\tilde{\alpha}\tilde{q}}$, and being zero elsewhere, as $\tilde{\gamma}$ approaches 0.

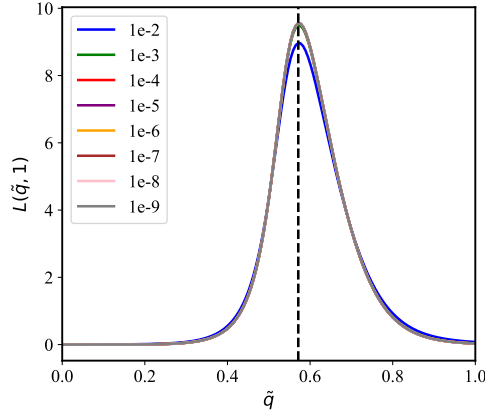
In Fig. 6.1 we show the energy loss function $L(\tilde{q}, \tilde{\omega})$ at fixed frequencies $\tilde{\omega} = 0.1, 0.5$, and 1.0 as a function of the wave number $\tilde{q}$, for different values of the damping parameter $\tilde{\gamma}$. In these curves we use the typical values for the parameters as introduced before, giving the values 0.28, 10, and 1.75 for the parameters s , $\tilde{\ell}_c$, and $\tilde{\alpha}$, respectively. Moreover, in each case the plasmon wavenumber of the pristine system with no random potential is shown with the vertical dashed line $\tilde{q} = \frac{\tilde{\omega}^2}{\tilde{\alpha}}$. Fig. 6.1a shows that for the small frequency $\tilde{\omega} = 0.1$, the peak in the energy loss function of the system occurs at wavenumbers that are considerably larger than the corresponding value of the pristine system, such that the vertical line representing the peak wavenumber of the pristine system is out of $\tilde{q}$ -range shown in Fig. 6.1a. The curves in Fig. 6.1a show a prominent peak at some finite wavenumber with some broadening around it, and vanishingly small values at wavenumbers far from the peak. On the other hand, for smaller values of the $\tilde{\gamma}$, the peak is larger, while its broadening is smaller. Although this is the typical behavior of the energy loss function of the pristine system Eq. (6.29), in this case the energy loss function does not converge to a



(a) $\tilde{\omega} = 0.1$.



(b) $\tilde{\omega} = 0.5$.

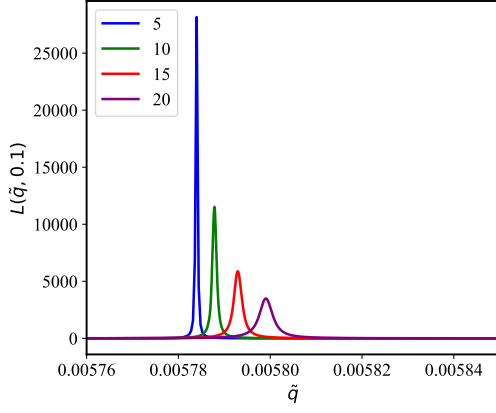


(c) $\tilde{\omega} = 1.0$.

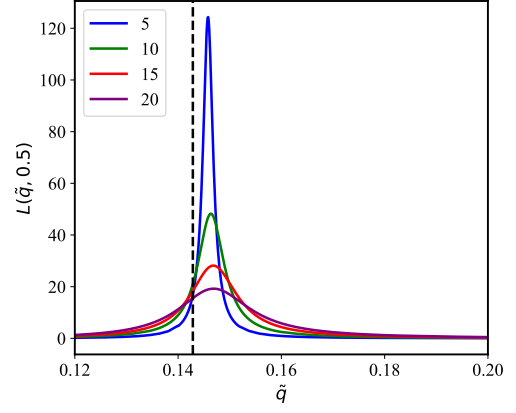
Figure 6.1: Energy loss function $L(\tilde{q}, \tilde{\omega})$, as a function of $\tilde{q}$ at fixed frequencies $\tilde{\omega} = 0.1, 0.5$, and 1.0 . The insets in panels (a) and (b) present the same curves of those panels in smaller ranges to magnify the details. Each color correspond to a specific value of $\tilde{\gamma}$ shown on each panel. The vertical dashed lines present the peak wavenumber of the pristine system at the corresponding frequency. s , $\tilde{\ell}_c$, and $\tilde{\alpha}$ are set to 0.28 , 10 , and 1.75 , respectively.

δ -like function but rather converges to a finite function with a non-zero broadening around the peak. Note that the curves for $\tilde{\gamma} = 10^{-8}$ and 10^{-9} in Fig. 6.1a are indistinguishable, even in the inset, which provides a more detailed view of the peak. This suggests that the random potential might be modeled by a finite effective damping parameter $\tilde{\gamma}_{\text{eff}}$ in the Drude model for small frequencies and small s values. In Fig. 6.1b, we present the same curves at $\tilde{\omega} = 0.5$. The peaks are clearly shifted to larger wavenumbers and the difference between the peak wavenumber of the pristine system (vertical line) and the system with random potential is relatively smaller in this case. Moreover, the convergence in $\tilde{\gamma}$ occurs at larger dampings such that for $\tilde{\gamma} \leq 10^{-5}$, curves are indistinguishable. Fig. 6.1c shows the same set of curves for $\tilde{\omega} = 1.0$. In this case, the shift in the peak frequency is negligible and the peaks of the loss function of the system with random potential have almost the same wavenumber as those of the pristine system (vertical line). Fig. 6.1c shows that for $\tilde{\omega} = 1.0$, the convergence in $\tilde{\gamma}$ is the fastest and for $\tilde{\gamma} \leq 10^{-3}$ the curves are indistinguishable. Unlike the cases in the panels (a) and (b), we see that the broadening in the panel (c) is asymmetrical about the peak position. Overall, Fig. 6.1 shows that the broadening around the peak is an increasing function of the frequency and the peak height decreases at higher frequencies. Note that the peak height is not a monotone function of $\tilde{\omega}$. According to Fig. 6.1, in the rest of this chapter we set $\tilde{\gamma} = 10^{-6}$ that is a quite small number for this purpose, ensuring that the resulting loss function is adequately close to the limiting loss function, while keeping the computations feasible.

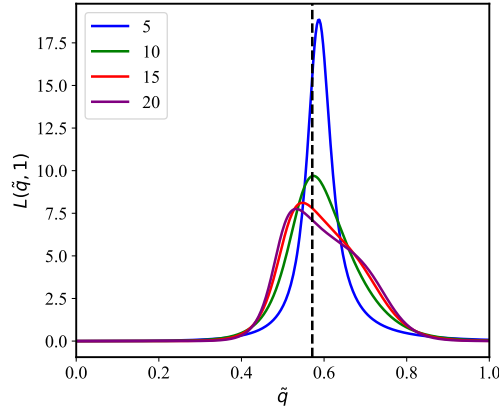
In Fig. 6.2, we study the effect of $\tilde{\ell}_c$ on $L(\tilde{q}, \tilde{\omega})$. Curves with different $\tilde{\ell}_c$ are shown in different colors as specified on each panel. The values of the other parameters are the same as those used in Fig. 6.1 with $\tilde{\gamma} = 10^{-6}$. Fig. 6.2a shows the loss function at $\tilde{\omega} = 0.1$ for four different values of $\tilde{\ell}_c$. One can see that when $\tilde{\ell}_c$ increases the peak wavenumber $\tilde{q}_{\text{peak}}$ for which $L(\tilde{q}, 0.1)$ reaches its maximum also increases almost linearly, deviating more from the pristine case. On the other hand, increasing $\tilde{\ell}_c$ decreases the magnitude of the peak, while rising the broadening of the peaks. Fig. 6.2b shows the same set of curves for $\tilde{\omega} = 0.5$. In this case the overall magnitude of the curves is reduced and peaks are shifted to right (larger wavenumbers). Moreover, the deviation of $\tilde{q}_{\text{peak}}$ with respect to the pristine peak frequency (vertical line) is less relative to the Fig. 6.2a. For $\tilde{\omega} = 0.5$, $\tilde{q}_{\text{peak}}$ is an increasing function of $\tilde{\ell}_c$, while its dependence on $\tilde{\ell}_c$ is relatively weak and peaks of the different curves overlap for the wavenumbers around $\tilde{q}_{\text{peak}}$. In Fig. 6.2c, we show the same curves for $\tilde{\omega} = 1.0$. In this case the peak wavenumber $\tilde{q}_{\text{peak}}$ is a decreasing function of the $\tilde{\ell}_c$ which opposes the cases with $\tilde{\omega} = 0.1$ and 0.5 . Here again increasing $\tilde{\ell}_c$ decreases the peak height, while broadening the peaks in a non-symmetric manner which makes it hard to model it by a finite effective damping rate $\tilde{\gamma}_{\text{eff}}$ in the Drude model. Nevertheless, for the rest of this chapter we set $\tilde{\ell}_c = 10$, which is the typical value of this parameter [165, 166].



(a) $\tilde{\omega} = 0.1$.

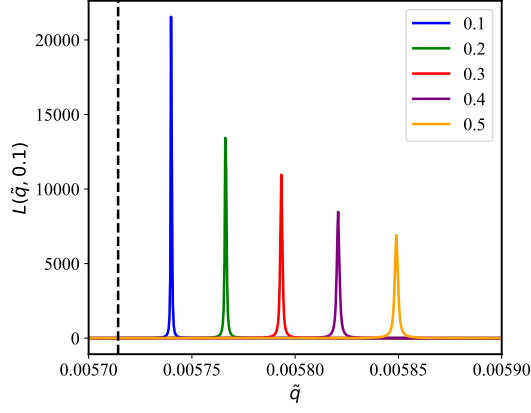


(b) $\tilde{\omega} = 0.5$.

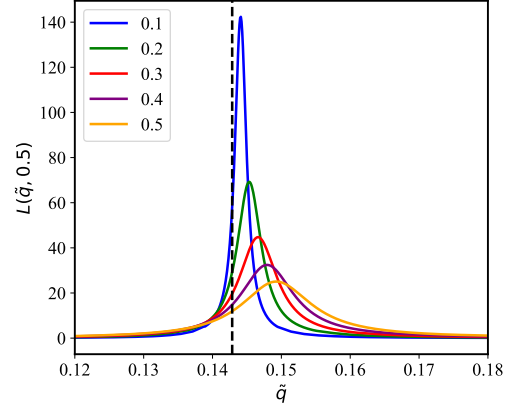


(c) $\tilde{\omega} = 1.0$.

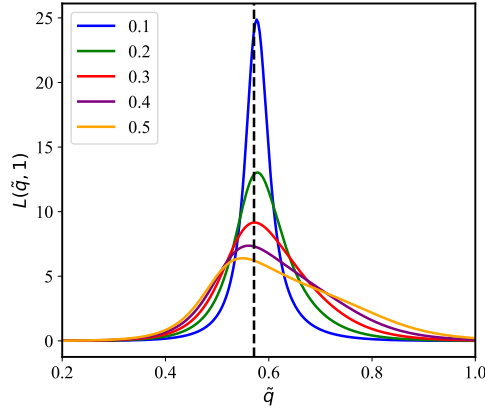
Figure 6.2: Energy loss function $L(\tilde{q}, \tilde{\omega})$, as a function of $\tilde{q}$ at fixed frequencies $\tilde{\omega} = 0.1, 0.5$, and 1.0 . Each color correspond to a specific value of $\tilde{\ell}_c$ shown on each panel. The vertical dashed lines present the peak wavenumber of the pristine system at the corresponding frequency. s , $\tilde{\gamma}$, and $\tilde{\alpha}$ are set to 0.28 , 10^{-6} , and 1.75 , respectively.



(a) $\tilde{\omega} = 0.1$.



(b) $\tilde{\omega} = 0.5$.



(c) $\tilde{\omega} = 1.0$.

Figure 6.3: Energy loss function $L(\tilde{q}, \tilde{\omega})$, as a function of $\tilde{q}$ at fixed frequencies $\tilde{\omega} = 0.1, 0.5$, and 1.0 . Each color correspond to a specific value of s shown on each panel. The vertical dashed lines present the peak wavenumber of the pristine system at the corresponding frequency. $\tilde{\gamma}$, $\tilde{\ell}_c$, and $\tilde{\alpha}$ are set to 10^{-6} , 10 , and 1.75 , respectively.

In Fig. 6.3, we analyze the effect of $s = (2\tilde{\alpha}\tilde{\sigma}_F)^2$ on the energy loss function, which is in fact analyzing the effect of the random potential scaled variance on the loss function. Note that the range of the s values in Fig. 6.3 corresponds to the reduced variance of $0 < \tilde{\sigma}_F^2 \lesssim 0.04$, which is quite small. One can see from Fig. 6.3a that for $\tilde{\omega} = 0.1$, the peak of the energy loss function occurs at $\tilde{q}_{\text{peak}}$ which is an increasing and seemingly linear function of s . Moreover, increasing s decreases the peak height, while increasing its broadening, as expected. The vertical dashed line in Fig. 6.3a represents the line $\tilde{q} = \frac{\tilde{\omega}^2}{\tilde{\alpha}}$ which is the peak wavenumber of the pristine system with no random potential, showing that, by increasing s the system deviates from its pristine state. Fig. 6.3b shows the same set of curves, at the intermediate frequency $\tilde{\omega} = 0.5$. In this case, $\tilde{q}_{\text{peak}}$ is again an increasing function of s , but not linear. Moreover, increasing s decreases the peak height and increases their broadening. Note that at higher frequencies, the relative deviation of the $\tilde{q}_{\text{peak}}$ from the peak wavenumber of the pristine system decreases. The same curves at the higher frequency $\tilde{\omega} = 1.0$ are shown in Fig. 6.3c. In this case, $\tilde{q}_{\text{peak}}$ is a decreasing function of s , whereas the height and broadening of the peaks are still decreasing and increasing functions of s , respectively. Note that for $s \gtrsim 0.2$, the curves broaden asymmetrically, showing the complexities coming from the non-linear nature of Eq. (6.24) and the fact that at high frequency regime and in the presence of the random potential, a simple Drude model can not adequately describe the material.

Fig. 6.4 shows a 2D density plot of $\sinh^{-1}(L(\tilde{q}, \tilde{\omega}))$, for the typical values of the parameters: $s = 0.28$, $\tilde{\ell}_c = 10$, $\tilde{\alpha} = 1.75$, and $\tilde{\gamma} = 10^{-6}$. Since the loss function $L(\tilde{q}, \tilde{\omega})$ in the range of $\tilde{q}$ and $\tilde{\omega}$ in Fig. 6.4 varies on an interval from 0 to 38000, we show the hyperbolic arcsine of L

$$\sinh^{-1}(L) = \ln \left(L + \sqrt{L^2 + 1} \right), \quad (6.30)$$

which varies on a much smaller interval, allowing us to follow the behavior of the function without losing data due to the change of the order of magnitudes. Fig. 6.4a shows that the loss function is vanishingly small over the unit square in the $(\tilde{q}, \tilde{\omega})$ plane, except at the vicinity of the curve $\tilde{\omega} = \sqrt{\tilde{\alpha}\tilde{q}}$, which is the dispersion curve of the pristine system. Moreover, Fig. 6.4a shows that for small wavenumbers $\tilde{q}$ the broadening of the peaks is small, while for large wavenumbers there exist a significant broadening. Note that since the loci of the peaks roughly satisfy the relation $\tilde{\omega}^2 = \tilde{\alpha}\tilde{q}$, the dependence of the properties on increasing $\tilde{q}$ is qualitatively the same as the dependence of those properties on increasing $\tilde{\omega}$. In Fig. 6.4b, we show the same density plot with different ranges for the $\tilde{q}$ and $\tilde{\omega}$. This figure show that there exists a small frequency window $0.03 \lesssim \tilde{\omega} \lesssim 0.08$, in which the loss functions takes the largest values, specifically $\tilde{L} \sim 37000$ for $\tilde{\omega} \sim 0.049$, with vanishingly small broadening. Finally, in Fig. 6.5, the energy loss function is shown, superimposed with

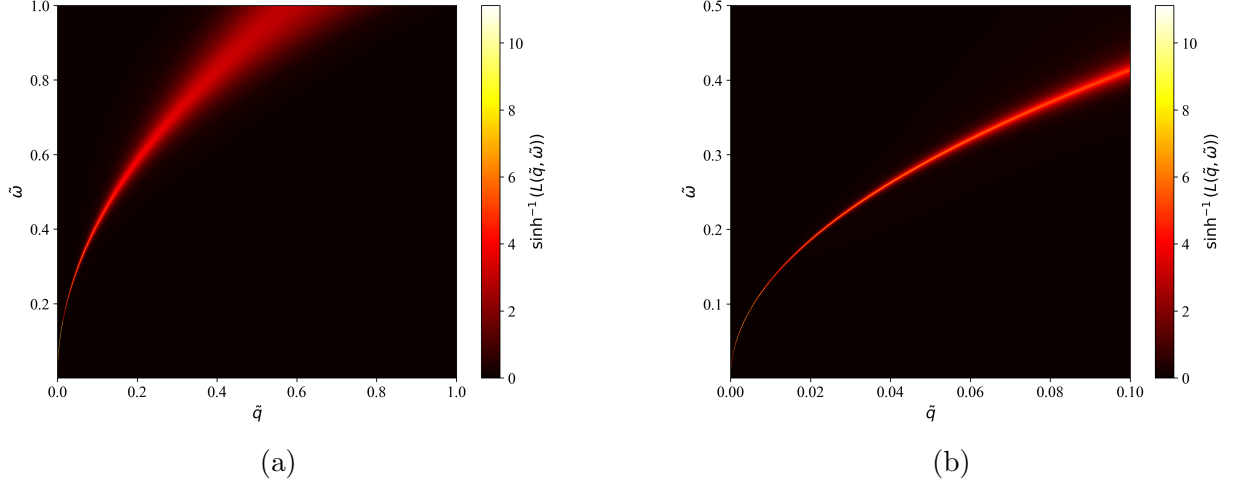


Figure 6.4: Hyperbolic arcsine of the energy loss function $\sinh^{-1}(L(\tilde{q}, \tilde{\omega}))$, as a function of $\tilde{q}$ and $\tilde{\omega}$. Parameters used in this figure are: $s = 0.28$, $\tilde{\ell}_c = 10$, $\tilde{\alpha} = 1.75$, and $\tilde{\gamma} = 10^{-6}$. Panels (a) and (b) present the same data in different ranges. One can see in panel (b) that $L(\tilde{q}, \tilde{\omega})$ takes largest values for very small wavenumbers and frequencies.

the dispersion curve of the pristine system in white, showing that the random potential makes the δ -like peaks broaden and finite.

We finish this chapter by showing in Fig. 6.6 a sample case of the function $\tilde{\mu}(\tilde{q}, \tilde{\omega})$. This function is obtained for the the same parameters used in Fig. 6.5, with $\tilde{\omega} = 1.0$. The vertical dashed line presents the wavenumber $\tilde{q} = \frac{\tilde{\omega}^2}{\tilde{\alpha}}$ for which $\epsilon_{2D}(\tilde{q}, \tilde{\omega}) = 0$. Although it is hard to track the role of each parameter in the shape of $\tilde{\mu}(\tilde{q}, \tilde{\omega})$ due to the non-linearity, Fig. 6.6 clearly shows that $\tilde{\mu}(\frac{\tilde{\omega}^2}{\tilde{\alpha}}, \tilde{\omega}) \neq 0$ at the wave numbers and frequencies satisfying $\epsilon_{2D}(\tilde{q}, \tilde{\omega}) = 0$, giving rise to the broadening and preventing δ -like peaks.

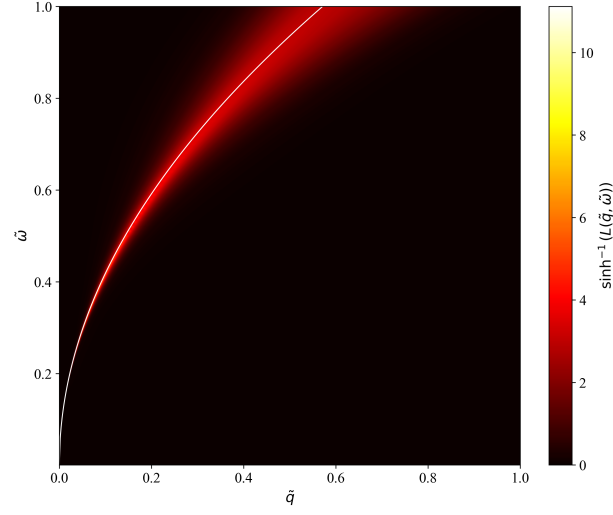


Figure 6.5: Hyperbolic arcsine of the energy loss function $\sinh^{-1}(\tilde{L}(\tilde{q}, \tilde{\omega}))$, as a function of $\tilde{q}$ and $\tilde{\omega}$. Parameters used in this figure are: $s = 0.28$, $\tilde{\ell}_c = 10$, $\tilde{\alpha} = 1.75$, and $\tilde{\gamma} = 10^{-6}$. The white curve represents the plasmon dispersion curve of the pristine system $\tilde{\omega} = \sqrt{\tilde{\alpha}\tilde{q}}$.

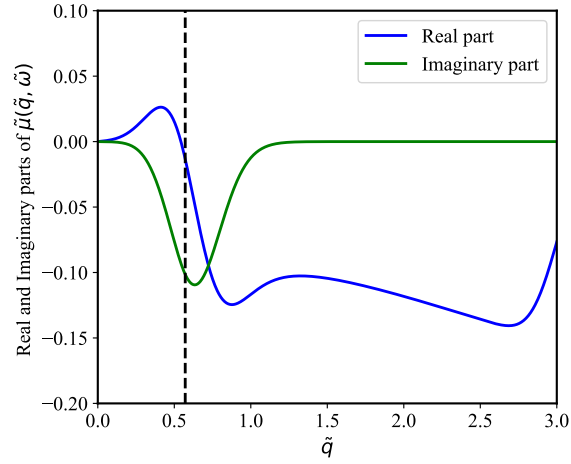


Figure 6.6: The real and the imaginary parts of $\tilde{\mu}(\tilde{q}, \tilde{\omega})$, as a function of $\tilde{q}$ for $\tilde{\omega} = 1.0$. The vertical dashed line presents the wavenumber $\tilde{q} = \frac{\tilde{\omega}^2}{\tilde{\alpha}}$ for which $\epsilon_{2D}(\tilde{q}, \tilde{\omega}) = 0$.

Chapter 7

Conclusions and future work

This thesis presents a comprehensive study of the interaction between charged particles with isotropic and anisotropic 2D materials in the context of EELS experiments with grazing scattering of incident electrons that move at the speeds on the order of the Bohr velocity. In Chapter 2, we introduced the essential concepts and assumptions, including Maxwell's equations in the non-retarded approximation, translational invariance in 2D materials of large area, and the 2D Fourier transform, which simplify the mathematical treatment while having clear physical interpretations in the reciprocal space. We also reviewed several basic analytical models for the electronic response functions, such as the effective dielectric function, the polarization function and the in-plane conductivity of 2D materials. Chapter 3 presents three models of increasing complexity for the polarization function of anisotropic 2D materials, focusing on the THz-MIR frequency range: the anisotropic Drude model, which accounts merely for the intraband electronic transitions, the optical model that adds a correction for interband transitions to the anisotropic Drude model, and the semiclassical model that includes non-local effects in the intraband transitions in addition to the same interband correction as in the optical model. The quantum mechanical 2D Lindhard function was also discussed separately, and was used only in the low-frequency limit in chapter 5 for mobility calculations.

In chapter 4, we studied the force components and the wake patterns using the anisotropic Drude, the optical, and the semiclassical models, which were compiled by using the parameters derived from *ab initio* calculations. A key aspect of the optical model is the inclusion of a contribution from high-energy interband electronic transitions in the static limit. This interband contribution, combined with the anisotropic Drude model for intraband transitions, give rise to a topological transition from quasi-elliptic (in the THz range) to hyperbolic (in the MIR range) iso-frequency dispersion curves for plasmon modes in doped

phosphorene. In addition to its application to free-standing phosphorene, this model was combined with a simple dielectric function to describe optical phonons in a SiO_2 substrate, allowing us to investigate plasmon-phonon hybridization in supported phosphorene. As anticipated, the plasmon dispersion surface splits into three branches corresponding to three plasmon-phonon hybrid modes, separated by two Reststrahlen bands of frequencies in the SiO_2 substrate. This hybridization maintains the elliptic and hyperbolic nature of iso-frequency curves within the frequency ranges defined by the phosphorene conductivity model, while exhibiting the avoided crossings between pairs of curves with different characteristics.

The developed dielectric models of the target system were further applied to calculate the forces acting on a classical point charge moving parallel to phosphorene at a distance of $z_0 = 50$ nm, with the speeds relevant to HREELS under oblique incidence or to the grazing ion scattering, which are both highly sensitive to surface anisotropy. Results show significant effects of phosphorene's anisotropy on the force components, which are strongly influenced by changes in the speed and direction of the charged particle's motion. Besides the longitudinal stopping force, which slows down the particle, there is also a transverse dissipative stopping force acting parallel to the surface but perpendicular to the particle's trajectory. This transverse force is quite strong, comparable to the longitudinal stopping force, and its character rapidly changes as the speed and direction of motion vary. For example, when the particle moves in a direction not aligned with phosphorene's principal axes, the transverse stopping force bends the particle's trajectory toward the AC direction at a normalized speed of $\bar{v} \sim 2$, whereas for $\bar{v} \sim 7$ it tries to bend the charged particle trajectory toward the ZZ direction.

Additionally, the dynamical image force, which typically is an attractive force pulling the particle toward the surface, also shows significant effects from phosphorene's anisotropy. Interestingly, this force can reverse its sign and become repulsive when the particle moves above free-standing phosphorene in a direction close to the AC direction at the speeds such that $6 \lesssim \bar{v} \lesssim 16$. This unexpected result may be due to the dominant role of the anisotropic Drude model at high frequencies, suggesting that further investigation is needed. We have shown that including a correction to the conductivity accounting for interband electron transitions in phosphorene can partially address this issue. This observation points to a required direction for future research, which will involve using *ab initio* data to explore the sign change in the dynamic image force across a wide frequency range in doped phosphorene [1]. Moreover, similar counterintuitive effects of a repulsive dynamic image force have been observed in other contexts using relativistic treatments of particle beam interactions with targets [172, 173]. It is possible that a relativistic approach could provide further insight into the behavior of the image force at high particle speeds

in this context, which will also be a focus of future work [145, 174, 175].

The primary effect of the SiO_2 substrate was observed to reduce the magnitude of all force components and to reduce the relative anisotropy in their dependence on the direction of motion. At speeds beyond the value $\bar{v} \approx 6$, the image force above supported phosphorene stabilizes at a finite value with minimal angular dependence, thereby removing the region where the image force appears repulsive in the case of free-standing phosphorene. Despite this, the transverse stopping force remains significant and continues to show strong anisotropy in the presence of a substrate, which could be detectable in well-designed HREELS experiments using reflection of electrons at grazing incidence with energies around ~ 100 eV. To accurately model such experiments, it is necessary to have expressions for the force components that are valid for all particle distances, including scenarios where the particle strongly interacts with the bulk modes of the substrate. It was shown that employing a bulk dielectric function for a polar substrate is sufficient to capture the essential features of its phonon spectra in HREELS under near-specular reflection at grazing incidence, particularly when the substrate thickness exceeds 2–3 nm [72, 73]. A more significant limitation for potential applications of this modeling is the restriction to large distances, which was applied in our work to simplify the derivation of a conductivity model for phosphorene by invoking its optical limit. In future research, we plan to fully account for non-local effects by utilizing momentum-dependent *ab initio* data for phosphorene’s conductivity [1] to numerically calculate the force components across a complete range of particle distances from phosphorene. This approach would enable the simulation of realistic trajectories of incident particles and help assess the role of anisotropy in the reflected electron spectra. One possible method to achieve this is to use a Monte Carlo simulation based on classical equations for the electron trajectory under grazing incidence on phosphorene [176], enhanced by a velocity- and distance-dependent anisotropic in-plane friction force, while treating motion in the perpendicular direction as adiabatic [79, 176].

Moreover, using the dielectric models introduced in this study, we demonstrated a variety of wake structures in the potential created by a charged particle moving parallel to an anisotropic 2D material. Due to the analytical simplicity of our dielectric response models, we conducted a detailed stationary phase analysis, allowing us to uncover surprising similarities in the wake patterns due to plasmon excitation in an anisotropic 2D electron gas with analogous effects seen in macroscopic ship and atmospheric wakes. These analogies include asymmetry caused by shear currents, flattening of dispersion seen in orographic lee waves, and Mach-like wakes resulting from capillarity, despite the fact that the microscopic mechanisms behind the wake in 2D materials differ significantly from those in macroscopic wakes.

While our modeling is well-suited for the scattering geometry in EELS using an aloof

electron beam with medium energy [67, 140], observing the displayed wake patterns in this setting might be challenging due to the rapid time scales and short length scales involved in the response of 2D target materials. An alternative approach could involve a static obstacle within an electron flow in the material, similar to a recently conducted experiment [177]. This experiment aligns with the hydrodynamic limit of electron motion in 2D [178], but it also highlights the need to extend the current study of the wake effects to lower relative speeds, where wake patterns are heavily influenced by plasmon dispersion at higher wavenumbers. In this context, it would be valuable to explore various analytical models of electron dynamics in both hydrodynamic and RPA regimes. This exploration could extend the stationary-phase analysis to the speeds near the threshold for wake formation, offering deeper insights into the directionality of plasmon excitation in anisotropic 2D materials and the impact of nonlocal effects. On the other hand, including the relativistic effects is still an open problem for our future studies.

Finally, phosphorene is known to be unstable in atmospheric conditions, but its applicability in nanoelectronic and plasmonic devices can be achieved by its encapsulation with hexagonal Boron Nitride (hBN) as a protective insulating layer [179]. Accordingly, our calculations of the forces and wakes should be repeated in future work by considering, e.g., a supported phosphorene layer that is covered by a single sheet of hBN with the external charge moving over that sheet.

In chapter 5, we adapted the ELM to compute the drift mobility tensor for charge carriers in an anisotropic 2D material, which are scattered by charged impurities inside the substrate, using model parameters specific to doped phosphorene on an oxide layer. By comparing our results with the available computational data in the literature, which used the Boltzmann transport equation (BTE) approach at zero temperature, we demonstrated that the ELM-derived formula is reliable and provides a more computationally efficient method for studying the DC transport properties of these materials. To highlight the ELM's efficiency, we investigated the impact of randomly distributed charged impurities using a structure factor, which naturally emerges in the mobility tensor formula derived from the ELM. Additionally, we modified the Lindhard's polarization function of quasi-free charge carriers in doped phosphorene by incorporating a static polarizability tensor with components α_x and α_y , which result from the interband electron transitions in phosphorene.

For a planar distribution of impurities, we observed that increasing their depth within the substrate leads to an increase in the mobility tensor components and a decrease in their asymmetry ratio, which results from a weakened interaction with charge carriers. These effects become more significant at higher doping densities, highlighting the growing importance of static screening due to intraband electron transitions in phosphorene. This impurity screening mechanism is further enhanced by including static polarizabilities $\alpha_{x/y}$

from interband electron transitions, which is especially noticeable for shallow impurities and higher doping levels. The presence of non-zero $\alpha_{x/y}$ in phosphorene also contributes to reducing the mobility asymmetry ratio as doping density increases, similar to the effect observed with greater impurity depth. Meanwhile, the presence of a finite correlation distance r_c among impurities in the hard disk model for a planar layer in the substrate leads to an increase in the mobility tensor components as well as an increase in the mobility asymmetry ratio, as r_c increases, but these effects are noticeable only at doping densities lower than the impurity density.

Investigating more complex models for the geometric arrangements of charged impurities near phosphorene is a logical next step for future research. Although our study focused on an isotropic distribution of impurities in the directions parallel to phosphorene, it would be valuable to explore how anisotropic models of the impurities' arrangement could impact the anisotropy in the mobility tensor of doped phosphorene.

In Chapter 6, we applied the smoothing method to examine how a random distribution of charged impurities affects the energy loss function of a system comprising a doped graphene supported by SiO_2 . The random distribution of impurities within the SiO_2 substrate is represented by a random potential that shifts the Dirac point of the graphene relative to the (fixed) Fermi energy, which can be alternatively considered as a random local Fermi energy relative to the Dirac (or neutrality) point of graphene. We modeled this random potential using a Gaussian distribution with a correlation distance l_c and a standard deviation σ_F . This approach led to a nonlinear integral equation for a memory function, whose solution is a critical component of the system's energy loss function. We developed a modified iteration scheme, referred to as DIS, that was successful to solve this nonlinear integral equation. Our computations revealed that the energy loss function of such a disordered structure remains finite and broadened around the peaks where the loss function of the pristine system with no random potential displays a sharp δ -like function along the dispersion curve corresponding to the sheet plasmon in doped graphene. The broadening caused by the random potential introduces an inherent energy loss that is independent of the phenomenological damping parameter γ in the Drude model. Additionally, our computations showed that changing the correlation distance l_c or the random potential variance σ_F^2 shifts the peak location and alters its broadening. Therefore, the random potential can slightly modify the plasmon signature of the material as well as introduce inherent losses.

A potential future direction for this research is to consider random potentials with different distributions. We initially chose the Gaussian distribution due to its favorable analytic properties, which allowed us to derive our formulation in a compact and closed form. Additionally, similar research could be conducted with anisotropic materials such as

phosphorene. However, for anisotropic materials, solving the nonlinear integral equation could be computationally expensive, as it may not be possible to perform integration over one variable analytically, leading to an integral equation that involves a double integral.

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APPENDICES

Appendix A

Green's functions of two specific problems

In this appendix detailed proofs for the Green's functions introduced in chapter (2) are provided. For the sake of completeness, the differential equation for each problem is stated again.

A.1 Source and probe above the 2DEG

Consider a system consisting of a semi-infinite substrate, occupying the region $z \leq -d$, and a 2DEG that is situated in the plane $z = 0$. We are looking for the electric potential in the region $z \geq 0$, when the source is also in the same region $z \geq 0$. We need to solve

$$q^2 \epsilon(z) G(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G(z, z') \right] = 4\pi [\delta(z - z') + \delta(z) \rho_{2D}]; \quad z, z' \geq 0, \quad (\text{A.1})$$

with the decomposition

$$G(z, z') = G_0(z, z') + G_{\text{ind}}(z, z'), \quad (\text{A.2})$$

where $G_0(z, z')$ and $G_{\text{ind}}(z, z')$ satisfy

$$q^2 G_0(z, z') - \frac{\partial}{\partial z} \left[\frac{\partial}{\partial z} G_0(z, z') \right] = 4\pi \delta(z - z'), \quad (\text{A.3})$$

and

$$q^2 \epsilon(z) G_{\text{ind}}(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_{\text{ind}}(z, z') \right] = 4\pi \delta(z) \rho_{2D}. \quad (\text{A.4})$$

Adopting the piece-wise manner to solve differential equations and using the fact that $\lim_{z \rightarrow \pm\infty} G_{\text{ext}}(z, z') = 0$, the following function for the solution of the equation (A.3) can be considered

$$G_0(z, z') = \begin{cases} ae^{-qz}, & \text{for } z \geq z', \\ be^{qz}, & \text{for } z \leq z'. \end{cases} \quad (\text{A.5})$$

In this expression a and b are allowed to be functions of $\mathbf{q}$ and ω , with their precise form being determined by applying the matching conditions. The continuity of the electric potential, and the jump in the electric displacement conditions at $z = z'$ imply that

$$\begin{cases} ae^{-qz'} = be^{qz'}, \\ -qae^{-qz'} - qbe^{qz'} = -4\pi. \end{cases} \quad (\text{A.6})$$

Note that the second equation in Eqs. (A.6) can be obtained by taking the definite integral from both sides of Eq. (A.3) on the interval $[z' - \varepsilon, z' + \varepsilon]$ as $\varepsilon \rightarrow 0^+$ and using the fact that $G_0(z, z')$ is finite. Solving the system of equations (A.6) reveals that

$$\begin{cases} a = \frac{2\pi}{q} e^{qz'}, \\ b = \frac{2\pi}{q} e^{-qz'}. \end{cases} \quad (\text{A.7})$$

Hence,

$$G_0(z, z') = \begin{cases} \frac{2\pi}{q} e^{q(z'-z)}, & \text{for } z \geq z', \\ \frac{2\pi}{q} e^{q(z-z')}, & \text{for } z \leq z'. \end{cases} \quad (\text{A.8})$$

In order to solve Eq. (A.4), we use the following expression for the induced potential that is based on the piece-wise manner and the fact that $\lim_{z \rightarrow \pm\infty} G_{\text{ind}}(z, z') = 0$

$$G_{\text{ind}}(z, z') = \begin{cases} Ae^{-qz}, & \text{for } z \geq 0, \\ Be^{qz} + Ce^{-qz}, & \text{for } 0 \geq z \geq -d, \\ De^{qz}, & \text{for } z \leq -d; \end{cases} \quad (\text{A.9})$$

in which A , B , C , and D are unknowns. The continuity of the electric potential implies that

$$\begin{cases} A = B + C, \\ Be^{-qd} + Ce^{qd} = De^{-qd}. \end{cases} \quad (\text{A.10})$$

On the other hand, the conditions for the jump in the electric displacement can be obtained by taking the definite integral from both sides of Eq. (A.4) over the intervals $[-\varepsilon, \varepsilon]$ and $[-d - \varepsilon, -d + \varepsilon]$ as $\varepsilon \rightarrow 0^+$, and noting that in these conditions the total Green's function $G(z, z')$ must be considered. Therefore,

$$\begin{cases} \left[-qA + \frac{\partial}{\partial z} G_0(0^+, z') \right] - \left[qB - qC + \frac{\partial}{\partial z} G_0(0^-, z') \right] = -4\pi\rho_{2D}, \\ \left[qBe^{-qd} - qCe^{qd} + \frac{\partial}{\partial z} G_0(-d^+, z') \right] - \epsilon_s \left[qDe^{-qd} + \frac{\partial}{\partial z} G_0(-d^-, z') \right] = 0. \end{cases} \quad (\text{A.11})$$

Using the fact that

$$\rho_{2D} = -e^2 \chi G(0, z') = -e^2 \chi [A + G_0(0, z')], \quad (\text{A.12})$$

and

$$\frac{1}{q} \frac{\partial}{\partial z} G_0(-d, z') e^{qd} = \frac{2\pi}{q} e^{-qz'} = G_0(0, z'), \quad (\text{A.13})$$

finally, the following system of equations for A , B , C , and D is obtained

$$\begin{cases} A - B - C = 0, \\ B + Ce^{2qd} - D = 0, \\ -(1 + 2V_C \chi) A - B + C = 2V_C \chi G_0(0, z'), \\ B - Ce^{2qd} - \epsilon_s D = (\epsilon_s - 1) G_0(0, z'); \end{cases} \quad (\text{A.14})$$

in which

$$V_C = \frac{2\pi e^2}{q}. \quad (\text{A.15})$$

Since we only need the Green's function for the induced potential in the region $z \geq 0$, the system of equations (A.14) must be solved only for A . Ultimately,

$$A = \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - 1 \right] G_0(0, z'), \quad (\text{A.16})$$

with

$$\epsilon_{2D}(\mathbf{q}, \omega) = \epsilon_{bg}(q, \omega) + V_C \chi(\mathbf{q}, \omega), \quad (\text{A.17})$$

and

$$\epsilon_{bg}(q, \omega) = \left[1 - \frac{\epsilon_s(\omega) - 1}{\epsilon_s(\omega) + 1} e^{-2qd} \right]^{-1}. \quad (\text{A.18})$$

Therefore,

$$G_{\text{ind}}(z, z') = \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - 1 \right] G_0(0, z') e^{-qz} = \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - 1 \right] \frac{2\pi}{q} e^{-q(z+z')}. \quad (\text{A.19})$$

A.2 Source and probe inside the substrate

Consider a system consisting of a semi-infinite substrate, occupying the region $z \leq 0$, and a 2DEG that is situated in the plane $z = h$. We are looking for the electric potential in the region $z \leq 0$, when the source is also in the same region $z \leq 0$. We need to solve

$$q^2 \epsilon(z) G(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G(z, z') \right] = 4\pi [\delta(z - z') + \delta(z - h) \rho_{2D}]; \quad z, z' \leq 0. \quad (\text{A.20})$$

with the decomposition

$$G(z, z') = G_0(z, z') + G_{\text{ind}}(z, z'), \quad (\text{A.21})$$

where $G_0(z, z')$ and $G_{\text{ind}}(z, z')$ satisfy

$$q^2 \epsilon(z) G_0(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_0(z, z') \right] = 4\pi \delta(z - z'), \quad (\text{A.22})$$

and

$$q^2 \epsilon(z) G_{\text{ind}}(z, z') - \frac{\partial}{\partial z} \left[\epsilon(z) \frac{\partial}{\partial z} G_{\text{ind}}(z, z') \right] = 4\pi \delta(z - h) \rho_{2D}. \quad (\text{A.23})$$

Note that the effect of the substrate is also taken into account in Eq. (A.22). Based on the piece-wise approach to solve Eq. (A.22), and the fact that $\lim_{z \rightarrow \pm\infty} G_0(z, z') = 0$, one can write

$$G_0(z, z') = \begin{cases} ae^{-qz}, & \text{for } z \geq 0, \\ be^{qz} + ce^{-qz}, & \text{for } 0 \geq z \geq z', \\ de^{qz}, & \text{for } z \leq z', \end{cases} \quad (\text{A.24})$$

where a , b , c , and d are unknowns that are allowed to be functions of $\mathbf{q}$ and ω . Imposing the continuity of the electric potential gives

$$\begin{cases} a = b + c, \\ be^{qz'} + ce^{-qz'} = de^{qz'}. \end{cases} \quad (\text{A.25})$$

On the other hand, we can take the definite integral from both sides of the Eq. (A.22) over the intervals $[-\varepsilon, \varepsilon]$ and $[z' - \varepsilon, z' + \varepsilon]$ as $\varepsilon \rightarrow 0^+$, to find

$$\begin{cases} [-qa] - \epsilon_s [qb - qc] = 0, \\ \epsilon_s [qbe^{qz'} - qce^{-qz'}] - \epsilon_s [qde^{qz'}] = -4\pi. \end{cases} \quad (\text{A.26})$$

Note that Eqs. (A.26) are in fact the condition of the jump in the electric displacement that can be imposed separately, without taking the definite integral from the main differential equation. After some algebraic manipulations the following system of equations arises

$$\begin{cases} a - b - c = 0, \\ b + ce^{-2qz'} - d = 0, \\ a + \epsilon_s b - \epsilon_s c = 0, \\ b - ce^{-2qz'} - d = \frac{-4\pi}{q\epsilon_s} e^{-qz'}. \end{cases} \quad (\text{A.27})$$

In the procedure of solving the Eq. (A.23), we only need $G_0(z, z')$ in the region $z \geq 0$, hence, we just need to find a from Eqs. (A.27) that is given by

$$a = \frac{4\pi}{q(1 + \epsilon_s)} e^{qz'}. \quad (\text{A.28})$$

Therefore,

$$G_0(z, z') = \frac{4\pi}{q(1 + \epsilon_s)} e^{q(z' - z)}, \quad z \geq 0 \geq z'. \quad (\text{A.29})$$

In order to solve Eq. (A.23), we use the following expression for the induced potential that is based on the piece-wise manner and the fact that $\lim_{z \rightarrow \pm\infty} G_{\text{ind}}(z, z') = 0$

$$G_{\text{ind}}(z, z') = \begin{cases} Ae^{-qz}, & \text{for } z \geq h, \\ Be^{qz} + Ce^{-qz}, & \text{for } h \geq z \geq 0, \\ De^{qz}, & \text{for } z \leq 0. \end{cases} \quad (\text{A.30})$$

In which A , B , C , and D are unknowns. Imposing the continuity condition gives

$$\begin{cases} Ae^{-qh} = Be^{qh} + Ce^{-qh}, \\ B + C = D. \end{cases} \quad (\text{A.31})$$

Now we impose the jump condition for the electric displacement on the total potential $G(z, z')$, that can be equally obtained by integrating the Eq. (A.23) over the intervals $[-\varepsilon, \varepsilon]$ and $[h - \varepsilon, h + \varepsilon]$ as $\varepsilon \rightarrow 0^+$ when the effect of the potential G_0 is also considered. The resulting equations are

$$\begin{cases} \left[-qAe^{-qh} + \frac{\partial}{\partial z} G_0(h^+, z') \right] - \left[qBe^{qh} - qCe^{-qh} + \frac{\partial}{\partial z} G_0(h^-, z') \right] = -4\pi\rho_{2D}, \\ \left[qB - qC + \frac{\partial}{\partial z} G_0(0^+, z') \right] - \epsilon_s \left[qD + \frac{\partial}{\partial z} G_0(0^-, z') \right] = 0. \end{cases} \quad (\text{A.32})$$

Using the facts that

$$\rho_{2D} = -e^2 \chi G(h, z') = -e^2 \chi [Ae^{-qh} + G_0(h, z')] , \quad (\text{A.33})$$

$$G_0(h, z')e^{qh} = G_0(0, z'), \quad (\text{A.34})$$

and

$$\frac{\partial}{\partial z} G_0(0^+, z') - \epsilon_s \frac{\partial}{\partial z} G_0(0^-, z') = 0, \quad (\text{A.35})$$

as it is the first equation in Eqs. (A.26), the following system of equations in A , B , C , and D can be obtained after some algebraic manipulations

$$\begin{cases} A - Be^{2qh} - C = 0, \\ B + C - D = 0, \\ -(1 + 2V_C \chi)A - Be^{2qh} + C = 2V_C \chi G_0(0, z'), \\ B - C - \epsilon_s D = 0; \end{cases} \quad (\text{A.36})$$

in which V_C is given by Eq. (A.15). Since we are only interested in $G_{\text{ind}}(z, z')$ in the region $z \leq 0$ and $z' \leq 0$, Eqs. (A.36) can be solved just for D . Hence,

$$G_{\text{ind}}(z, z') = De^{qz} = \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - \frac{1}{\epsilon_{\text{bg}}(q, \omega)} \right] \frac{\epsilon_{\text{bg}}^2(q, \omega)}{\epsilon_{\text{bg}}^0} e^{-2qh} G_0(0, z') e^{qz}, \quad (\text{A.37})$$

with $\epsilon_{\text{bg}}(q, \omega)$ from Eq. (A.18), and

$$\epsilon_{\text{bg}}^0 = \frac{\epsilon_s + 1}{2}. \quad (\text{A.38})$$

Ultimately, using Eq. (A.29) in Eq. (A.37) gives

$$G_{\text{ind}}(z, z') = \frac{2\pi}{q} \left[\frac{1}{\epsilon_{2D}(\mathbf{q}, \omega)} - \frac{1}{\epsilon_{\text{bg}}(q, \omega)} \right] \left[\frac{\epsilon_{\text{bg}}(q, \omega)}{\epsilon_{\text{bg}}^0} \right]^2 e^{q(z+z'-2h)}; \quad z, z' \leq 0. \quad (\text{A.39})$$

Note that setting $\chi = 0$ in Eq. (A.39) results in $G_{\text{ind}}(z, z') = 0$, which shows that this induced Green's function merely accounts for the interactions between the source and the 2DEG through the substrate, and the direct interactions between the source and the substrate are included in the $G_0(z, z')$.

Appendix B

Drude and BGK models for the phosphorene intraband conductivity tensor

B.1 Anisotropic Drude model

In this section a derivation for the anisotropic Drude model that is based on a classical treatment of the anisotropic 2DEG is presented. The equation of motion for a particle in an anisotropic 2D gas of particles (electrons or holes), by virtue of the Newton's second law can be written as

$$q\mathbf{E} - \overleftrightarrow{\lambda}\mathbf{r}' = \overleftrightarrow{m}\mathbf{r}'', \quad (\text{B.1})$$

where q is the particle charge ($-e$ for electrons and e for holes), and $\overleftrightarrow{\lambda}$ is the friction tensor that describes the effect of all other particles, atomic nuclei, and lossy processes on the particle that its position is $\mathbf{r}$. The friction tensor could be represented in the matrix notation as

$$\overleftrightarrow{\lambda} = \begin{bmatrix} \lambda_x & 0 \\ 0 & \lambda_y \end{bmatrix}, \quad (\text{B.2})$$

and $\overleftrightarrow{m}$, the effective mass tensor, could be represented by

$$\overleftrightarrow{m} = \begin{bmatrix} m_x & 0 \\ 0 & m_y \end{bmatrix}. \quad (\text{B.3})$$

Note that this tensor notation allows for having different friction constants and effective masses in different directions that is the main feature of the anisotropic materials. The Fourier transformation of the Eq. (B.1) is given by

$$q\tilde{\mathbf{E}} + \overleftrightarrow{\lambda}i\omega\tilde{\mathbf{r}} = -\overleftrightarrow{m}\omega^2\tilde{\mathbf{r}}, \quad (\text{B.4})$$

where each time derivative is transformed to a factor of $-i\omega$. Moreover, the drift current is defined by

$$\mathbf{J} = \rho\mathbf{r}', \quad (\text{B.5})$$

with $\rho = q|n|$, that in the Fourier domain reads as

$$\tilde{\mathbf{J}} = -q|n|i\omega\tilde{\mathbf{r}}. \quad (\text{B.6})$$

Combining Eqs. (B.4) and (B.6) gives

$$\left[\frac{1}{|n|}\overleftrightarrow{\lambda} - \frac{i\omega}{|n|}\overleftrightarrow{m} \right] \tilde{\mathbf{J}} = q^2\tilde{\mathbf{E}}, \quad (\text{B.7})$$

which can be solved for $\tilde{\mathbf{J}}$ as

$$\tilde{\mathbf{J}} = q^2 \left[\frac{1}{|n|}\overleftrightarrow{\lambda} - \frac{i\omega}{|n|}\overleftrightarrow{m} \right]^{-1} \tilde{\mathbf{E}}. \quad (\text{B.8})$$

On the other hand, because the tensor in the brackets is diagonal, a simple comparison between Eq. (B.8) and the Ohm's law $\tilde{\mathbf{J}} = \overleftrightarrow{\sigma}\tilde{\mathbf{E}}$ reveals that

$$\overleftrightarrow{\sigma} = q^2 \left[\frac{1}{|n|}\overleftrightarrow{\lambda} - \frac{i\omega}{|n|}\overleftrightarrow{m} \right]^{-1} = q^2 \left(\frac{1}{|n|} \begin{bmatrix} \lambda_x - i\omega m_x & 0 \\ 0 & \lambda_y - i\omega m_y \end{bmatrix} \right)^{-1}, \quad (\text{B.9})$$

which reduces to

$$\overleftrightarrow{\sigma} = \begin{bmatrix} \frac{q^2|n|}{\lambda_x - i\omega m_x} & 0 \\ 0 & \frac{q^2|n|}{\lambda_y - i\omega m_y} \end{bmatrix} = \begin{bmatrix} \frac{i}{\pi} \frac{\frac{\pi q^2|n|}{m_x}}{\omega + i\gamma_x} & 0 \\ 0 & \frac{i}{\pi} \frac{\frac{\pi q^2|n|}{m_y}}{\omega + i\gamma_y} \end{bmatrix}, \quad (\text{B.10})$$

with $\gamma_x = \lambda_x/m_x$ and $\gamma_y = \lambda_y/m_y$. Finally, for the electron doped phosphorene, $q = -e$ and the Drude intraband conductivity tensor components are given by

$$\sigma_j^{\text{intra}}(\omega) = \frac{i}{\pi} \frac{D_j}{\omega + i\gamma_j}, \quad \text{for } j = x, y, \quad (\text{B.11})$$

with

$$D_j = \frac{\pi e^2 |n|}{m_j}. \quad (\text{B.12})$$

Note that the Drude model can be obtained from a more rigorous method that relies on the Kubo formula [122], which is a quantum mechanical based approach. In this formalism

$$\vec{\sigma}(\mathbf{q}, \omega) = \frac{1}{\hbar \omega} \sum_s \int dt \langle s | \mathbf{J}(\mathbf{q}, 0) \mathbf{J}^*(\mathbf{q}, t) | s \rangle e^{-i\omega t}, \quad (\text{B.13})$$

with the assumption

$$\mathbf{J}(\mathbf{q}, t) = \begin{bmatrix} J_x(\mathbf{q}, 0) \exp\left(-\frac{t}{\tau_x}\right) \\ J_y(\mathbf{q}, 0) \exp\left(-\frac{t}{\tau_y}\right) \end{bmatrix}, \quad (\text{B.14})$$

which means a current impulse will decay exponentially with the current-current correlation time τ_j that is the same for all states in each direction j .

B.2 Semiclassical intraband conductivity for isotropic 2D materials

In this part we derive the semiclassical intraband polarization function that is introduced in chapter 3. Using the Boltzmann equation in the Bhatnagar-Gross-Krook (BGK) approximation [137], we can find an expression for the intraband conductivity tensor, which is called BGK conductivity tensor. This section repeats the derivation given in Ref. [124] for isotropic 2D material, which serves as a basis for its generalization to anisotropic 2D material, as outlined in section 3.5.1 of the main text.

Consider a 2D material that its lowest conduction band could be approximated by

$$\varepsilon_{\mathbf{p}} = \frac{p_x^2}{2m} + \frac{p_y^2}{2m}. \quad (\text{B.15})$$

The probability density function $f(\mathbf{p}, \mathbf{r}, t)$ for the electrons of this 2D layer is the solution of the Boltzmann equation

$$\frac{\partial f}{\partial t} + \mathbf{v}_{\mathbf{p}} \cdot \frac{\partial f}{\partial \mathbf{r}} - e \mathbf{E}_{\parallel}^0 \cdot \frac{\partial f}{\partial \mathbf{p}} = I_{\text{coll}}, \quad (\text{B.16})$$

where $\mathbf{r} = x\hat{\mathbf{x}} + y\hat{\mathbf{y}}$, and

$$\mathbf{v}_{\mathbf{p}} = \frac{\partial \varepsilon(\mathbf{p})}{\partial \mathbf{p}} = \frac{p_x}{m} \hat{\mathbf{x}} + \frac{p_y}{m} \hat{\mathbf{y}} \quad (\text{B.17})$$

is the electron group velocity. Moreover, e is the electron charge, $\mathbf{E}_{\parallel}^0$ is the in-plane electric field, and I_{coll} is the collision term. Using perturbation methods, for a weak field $\mathbf{E}_{\parallel}^0$, the solution of Eq. (B.16) can be expanded as

$$f(\mathbf{p}, \mathbf{r}, t) = f_0(\mathbf{p}) + f_1(\mathbf{p}, \mathbf{r}, t) + \cdots, \quad (\text{B.18})$$

where $f_0(\mathbf{p}) = F(\varepsilon_{\mathbf{p}} - \mu(n_0))$ is the solution of the Boltzmann equation in the equilibrium, with

$$F(\varepsilon - \mu) \equiv \left(1 + \exp \left[\frac{\varepsilon - \mu}{k_B T} \right] \right)^{-1}, \quad (\text{B.19})$$

being the Fermi-Dirac (FD) distribution at temperature T . The equilibrium doping density n_0 and the chemical potential μ are related to each other by

$$n_0 = g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} F(\varepsilon_{\mathbf{p}} - \mu), \quad (\text{B.20})$$

where $g_d = 2$ is the spin degeneracy factor. Moreover, we assume that the collision term in Eq. (B.16), may be modeled within the relaxation-time approximation by

$$I_{\text{coll}} = -\gamma [f(\mathbf{p}, \mathbf{r}, t) - f_{\text{loc}}(\mathbf{p}, \mathbf{r}, t)], \quad (\text{B.21})$$

where γ is the constant electron-electron collision rate, and

$$f_{\text{loc}}(\mathbf{p}, \mathbf{r}, t) = F(\varepsilon_{\mathbf{p}} - \mu(n(\mathbf{r}, t))), \quad (\text{B.22})$$

with the mean electron density defined by

$$n(\mathbf{r}, t) = g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} f(\mathbf{p}, \mathbf{r}, t). \quad (\text{B.23})$$

If we substitute Eq. (B.18) into the Eq. (B.23), it turns out that $n(\mathbf{r}, t) = n_0 + n_1(\mathbf{r}, t) + \cdots$ that is commensurate with Eq. (B.18). Therefore, we can expand the local equilibrium density as $f_{\text{loc}}(\mathbf{p}, \mathbf{r}, t) = f_0(\mathbf{p}) + h(\mathbf{r}, t) + \cdots$, with the first-order term written as

$$h(\mathbf{r}, t) = -F'_0(\varepsilon_{\mathbf{p}}) \alpha n_1(\mathbf{r}, t), \quad (\text{B.24})$$

where $F_0'(\varepsilon) \equiv \frac{\partial}{\partial \varepsilon} F(\varepsilon - \mu(n_0))$, the coefficient α is obtained from (B.20) as

$$\alpha = \left(\frac{dn_0}{d\mu} \right)^{-1}, \quad (\text{B.25})$$

and the induced electron density to the first order is given by

$$n_1(\mathbf{r}, t) = g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} f_1(\mathbf{p}, \mathbf{r}, t). \quad (\text{B.26})$$

The first-order terms satisfy the following Boltzmann equation

$$\frac{\partial f_1}{\partial t} + \mathbf{v}_{\mathbf{p}} \cdot \frac{\partial f_1}{\partial \mathbf{r}} - e \mathbf{E}_{\parallel}^0 \cdot \mathbf{v}_{\mathbf{p}} F_0'(\varepsilon_{\mathbf{p}}) = -\gamma(f_1 - h), \quad (\text{B.27})$$

which is based on the approximation (substitution)

$$\frac{\partial f}{\partial \mathbf{p}} = \frac{\partial f}{\partial \varepsilon} \cdot \frac{\partial \varepsilon}{\partial \mathbf{p}} \rightarrow F_0'(\varepsilon_{\mathbf{p}}) \cdot \mathbf{v}_{\mathbf{p}} \quad (\text{B.28})$$

that is extensively used to solve the linearized BTE. Using the Fourier transformation with respect to $\mathbf{r}$ and t , one can solve Eq. (B.27) to find $\tilde{f}_1(\mathbf{p}, \mathbf{q}, \omega)$ as

$$\tilde{f}_1 = i F_0'(\varepsilon_{\mathbf{p}}) \frac{-\alpha \gamma \tilde{n}_1 + e \mathbf{v}_{\mathbf{p}} \cdot \tilde{\mathbf{E}}_{\parallel}^0}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}. \quad (\text{B.29})$$

If we put this expression for $\tilde{f}_1$ into the definition of the first-order current density of electrons

$$\mathbf{j}_1(\mathbf{r}, t) = -e g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} \mathbf{v}_{\mathbf{p}} f_1(\mathbf{p}, \mathbf{r}, t), \quad (\text{B.30})$$

we find the following expansion of the first-order current density in the Fourier domain

$$\tilde{\mathbf{j}}_1 = e \omega \tilde{n}_1 \mathbf{d} + \vec{\sigma}_{\text{RTA}} \cdot \mathbf{E}_{\parallel}^0. \quad (\text{B.31})$$

Here the auxiliary vector field $\mathbf{d}(\mathbf{q}, \omega)$ is defined by

$$\mathbf{d} = i \frac{\gamma}{\omega} \alpha g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} F_0'(\varepsilon_{\mathbf{p}}) \frac{\mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}, \quad (\text{B.32})$$

and $\vec{\sigma}_{\text{RTA}}$ is the in-plane conductivity tensor in the relaxation time approximation (RTA), defined as

$$\vec{\sigma}_{\text{RTA}} = -ie^2 g_d \iint \frac{d^2 \mathbf{p}}{(2\pi\hbar)^2} F_0'(\varepsilon_{\mathbf{p}}) \frac{\mathbf{v}_{\mathbf{p}} \mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}, \quad (\text{B.33})$$

where $\mathbf{v}_p \mathbf{v}_p$ is the outer product of the velocity vectors of electrons. The first term in Eq. (B.31) is the diffusion current and the second term is the drift current. Invoking the in-plane continuity equation for electrons, we may write in the Fourier domain

$$\tilde{n}_1 = -\frac{\mathbf{q} \cdot \tilde{\mathbf{j}}_1}{e\omega}, \quad (\text{B.34})$$

enabling us to convert Eq. (B.31) to

$$\tilde{\mathbf{j}}_1 = -\mathbf{d}(\mathbf{q} \cdot \tilde{\mathbf{j}}_1) + \vec{\sigma}_{\text{RTA}} \cdot \tilde{\mathbf{E}}_{\parallel}^0. \quad (\text{B.35})$$

A solution of this linear equation may be expressed in the form $\tilde{\mathbf{j}}_1 = \vec{\sigma}_{\text{BGK}} \cdot \tilde{\mathbf{E}}_{\parallel}^0$, which defines an in-plane conductivity tensor in the BGK approximation, denoted by $\vec{\sigma}_{\text{BGK}}(\mathbf{q}, \omega)$. In order to find $\vec{\sigma}_{\text{BGK}}$, we write Eq. (B.35) in the matrix form

$$\begin{bmatrix} \tilde{j}_x \\ \tilde{j}_y \end{bmatrix} = \begin{bmatrix} -(q_x \tilde{j}_x + q_y \tilde{j}_y) d_x \\ -(q_x \tilde{j}_x + q_y \tilde{j}_y) d_y \end{bmatrix} + \begin{bmatrix} \sigma_{xx}^{\text{RTA}} & \sigma_{xy}^{\text{RTA}} \\ \sigma_{yx}^{\text{RTA}} & \sigma_{yy}^{\text{RTA}} \end{bmatrix} \begin{bmatrix} \tilde{E}_x \\ \tilde{E}_y \end{bmatrix}, \quad (\text{B.36})$$

that can be transformed to

$$\begin{bmatrix} 1 + q_x d_x & q_y d_x \\ q_x d_y & 1 + q_y d_y \end{bmatrix} \begin{bmatrix} \tilde{j}_x \\ \tilde{j}_y \end{bmatrix} = \begin{bmatrix} \sigma_{xx}^{\text{RTA}} & \sigma_{xy}^{\text{RTA}} \\ \sigma_{yx}^{\text{RTA}} & \sigma_{yy}^{\text{RTA}} \end{bmatrix} \begin{bmatrix} \tilde{E}_x \\ \tilde{E}_y \end{bmatrix}, \quad (\text{B.37})$$

after some algebraic manipulations. Therefore the BGK conductivity tensor is given by

$$\vec{\sigma}_{\text{BGK}} = \begin{bmatrix} \sigma_{xx}^{\text{BGK}} & \sigma_{xy}^{\text{BGK}} \\ \sigma_{yx}^{\text{BGK}} & \sigma_{yy}^{\text{BGK}} \end{bmatrix} = \begin{bmatrix} 1 + q_x d_x & q_y d_x \\ q_x d_y & 1 + q_y d_y \end{bmatrix}^{-1} \begin{bmatrix} \sigma_{xx}^{\text{RTA}} & \sigma_{xy}^{\text{RTA}} \\ \sigma_{yx}^{\text{RTA}} & \sigma_{yy}^{\text{RTA}} \end{bmatrix}, \quad (\text{B.38})$$

in which all components are known from Eqs. (B.32) and (B.33).

In order to find the semiclassical polarization function, we start with multiplying both sides of the Eq. (B.35) by $\mathbf{q}$ from left that yields

$$\mathbf{q} \cdot \tilde{\mathbf{j}}_1 = -(\mathbf{q} \cdot \mathbf{d})(\mathbf{q} \cdot \tilde{\mathbf{j}}_1) - i\mathbf{q} \cdot \vec{\sigma}_{\text{RTA}} \cdot \mathbf{q} \tilde{\Phi}_0, \quad (\text{B.39})$$

in which we made use of $\tilde{\mathbf{E}}_{\parallel}^0 = -i\mathbf{q} \tilde{\Phi}_0$. Combining Eqs. (B.39) and (B.34) gives

$$\tilde{n}_1 = \frac{i}{e\omega} \frac{\mathbf{q} \cdot \vec{\sigma}_{\text{RTA}} \cdot \mathbf{q}}{1 + \mathbf{q} \cdot \mathbf{d}} \tilde{\Phi}_0. \quad (\text{B.40})$$

From the definition of the polarization function, $\tilde{\rho}_1 = -e^2 \chi \tilde{\Phi}_0 = -e \tilde{n}_1$, and Eq. (B.40) we find

$$\chi = \frac{i}{e^2 \omega} \frac{\mathbf{q} \cdot \vec{\sigma}_{\text{RTA}} \cdot \mathbf{q}}{1 + \mathbf{q} \cdot \mathbf{d}}, \quad (\text{B.41})$$

which is the semiclassical polarization function. Now we show Eq. (B.41) is in fact equal to the expression

$$\chi(q, \omega) = \frac{\chi_{\text{iso}}(q, \omega)}{1 - \frac{i\gamma}{\omega + i\gamma} \left(1 - \frac{\chi_{\text{iso}}(q, \omega)}{\chi_s^0} \right)}, \quad (\text{B.42})$$

that was introduced in chapter (3), which can be used to describe anisotropic materials upon using an appropriate change of variable $q \rightarrow Q(q_x, q_y)$ that is discussed before. The numerator of Eq. (B.41) could be written as

$$i\mathbf{q} \cdot \vec{\sigma}_{\text{RTA}} \cdot \mathbf{q} = e^2 g_d \iint \frac{d^2 \mathbf{p}}{(2\pi \hbar)^2} F_0'(\varepsilon_{\mathbf{p}}) \frac{[\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}]^2}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}, \quad (\text{B.43})$$

upon using the definition of $\vec{\sigma}_{\text{RTA}}$ in Eq. (B.33). On the other hand,

$$\frac{[\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}]^2}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}} = -\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}} + \frac{(\omega + i\gamma) \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}} \quad (\text{B.44})$$

that results in

$$i\mathbf{q} \cdot \vec{\sigma}_{\text{RTA}} \cdot \mathbf{q} = \frac{e^2 g_d}{(2\pi \hbar)^2} (\omega + i\gamma) \iint d^2 \mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) \frac{\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}, \quad (\text{B.45})$$

by virtue of the identity

$$\iint d^2 \mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}} = \mathbf{q} \cdot \iint d^2 \mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) \mathbf{v}_{\mathbf{p}} = 0, \quad (\text{B.46})$$

since the integrand is a symmetric vector field around the origin, see Eqs. (B.15) and (B.17).

Now if we define

$$\chi_{\text{iso}} = \frac{g_d}{(2\pi \hbar)^2} \iint d^2 \mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) \frac{\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}, \quad (\text{B.47})$$

$$\chi_s = \frac{-g_d}{(2\pi \hbar)^2} \iint d^2 \mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) = \frac{1}{\alpha} = \frac{m}{\pi \hbar^2} \textcolor{blue}{1}, \quad (\text{B.48})$$

and use the Eq. (B.45), we conclude that the polarization function in Eq. (B.41) could be written as

$$\chi = \frac{i}{e^2\omega} \frac{e^2(\omega + i\gamma)\chi_{\text{iso}}}{1 + i\frac{\gamma}{\omega}\frac{1}{\chi_s}\chi_{\text{iso}}} = \frac{\chi_{\text{iso}}(q, \omega)}{1 - \frac{i\gamma}{\omega + i\gamma} \left(1 - \frac{\chi_{\text{iso}}(q, \omega)}{\chi_s}\right)}. \quad (\text{B.49})$$

This is the semiclassical intraband polarization function introduced before, upon using

$$\iint d^2\mathbf{p} F_0'(\varepsilon_{\mathbf{p}}) \frac{\mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}}{\omega + i\gamma - \mathbf{q} \cdot \mathbf{v}_{\mathbf{p}}} = m \int_0^\infty d\varepsilon F_0'(\varepsilon) \int_0^{2\pi} d\vartheta \frac{v_\varepsilon q \cos \vartheta}{\omega + i\gamma - v_\varepsilon q \cos \vartheta}, \quad (\text{B.50})$$

that is obtained via changing the variables from Cartesian to polar coordinate, based on $v_\varepsilon = \sqrt{2\varepsilon/m}$ and Eq. (B.15).

¹This last equality holds only at zero temperature.

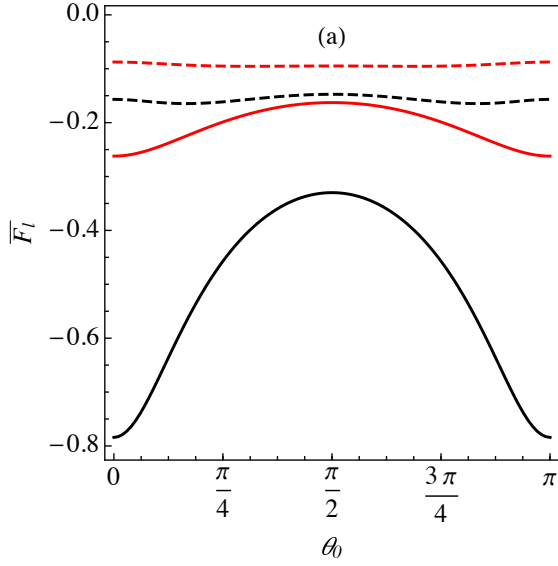
Appendix C

Additional analysis of forces

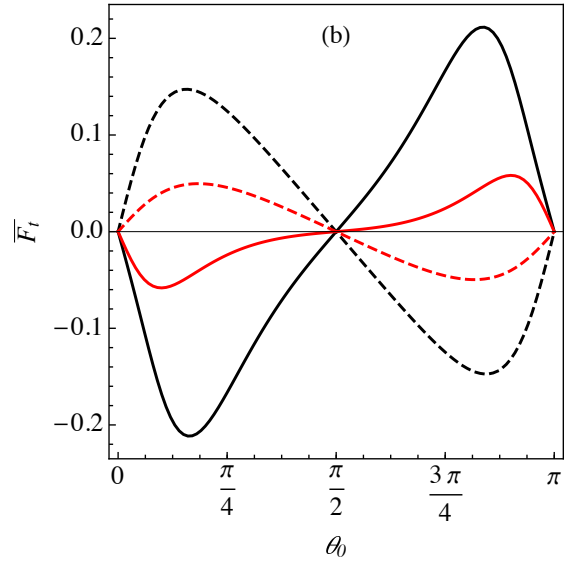
In this appendix, further analyses of the forces, and the role of the different dielectric models are provided. This appendix is a continuation of chapter 4. Recall that all forces are presented in a reduced form, normalized by $F_0 = (Ze)^2/(2z_0)^2$.

In Fig. C.1, we illustrate cross sections of the surfaces from Figs. 4.4, 4.5, and 4.6 at specific speeds, chosen to highlight the prominent angular dependencies of the three forces in free-standing phosphorene. For the longitudinal stopping force (Fig. C.1a), we select $\bar{v} = 4$ that corresponds to the dip region and $\bar{v} = 9$ that corresponds to the leveling off region. For the transverse stopping force (Fig. C.1b), we choose $\bar{v} = 2$ and $\bar{v} = 7$ to probe the local extremes. For the image force (Fig. C.1c), we pick $\bar{v} = 2$ that corresponds to its dip and $\bar{v} = 7$ that correspond to the region with a broad maximum and repulsive image force. Comparing the curves for free-standing phosphorene (black) with supported phosphorene (red) shows the substrate's impact in reducing force magnitudes and weakening anisotropic angular dependencies. In Fig. C.1c, at $\bar{v} = 2$, F_i peaks in the AC direction and dips in the ZZ direction for both free-standing and supported phosphorene. At $\bar{v} = 7$, this pattern reverses, with the image force becoming positive near $\theta_0 = 0$ for free-standing phosphorene but showing a slight oscillation around $-0.5 \times F_0$ for supported phosphorene. Finally, Fig. C.1b demonstrates that the transverse stopping force remains notably strong in the presence of the substrate and retains its antisymmetric angular dependence with respect to the principal directions, as seen in free-standing phosphorene.

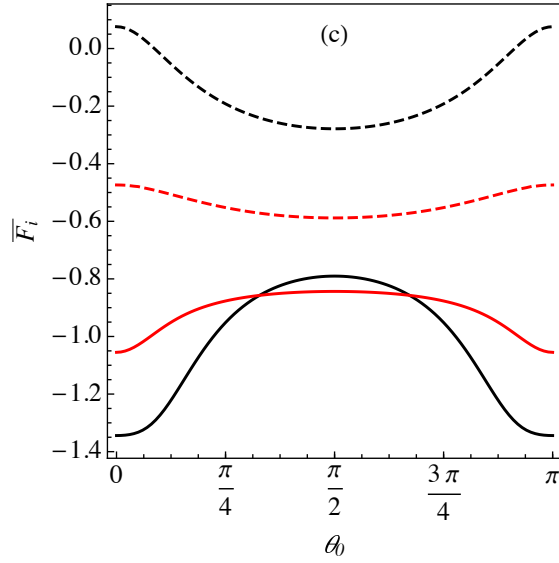
At this stage, it is advisable to give a detailed discussion on the unusual repulsive image force that occurs around the AC direction in the case of freestanding phosphorene. For this purpose, we introduce the isotropic Drude model, which is obtained from the anisotropic Drude model Eq. (3.30) with substitutions $m_x = m_y = m$ with $m = \frac{2m_x m_y}{m_x + m_y} \approx 0.238m_0$



(a) Longitudinal stopping force $\bar{F}_l$, at the normalized speeds: $\bar{v} = 4$ (solid lines) and 9 (dashed lines).



(b) Transverse stopping force $\bar{F}_t$, at the normalized speeds: $\bar{v} = 2$ (solid lines) and 7 (dashed lines).



(c) Dynamic image force $\bar{F}_i$, at the normalized speeds: $\bar{v} = 2$ (solid lines) and 7 (dashed lines).

Figure C.1: Normalized forces obtained from the optical model, as a function of the direction angle θ_0 with constant $\bar{v}$ for a particle moving at height $z_0 = 50$ nm, above freestanding (black curves) and supported (red curves) phosphorene.

and $\hbar\gamma_x = \hbar\gamma_y = \hbar\gamma \approx 12$ meV that is based on the assumption $\mu_x = \mu_y = \mu$ with $\mu = \frac{\mu_x + \mu_y}{2} \approx 406$ cm²V⁻¹s⁻¹. Moreover, we introduce the isotropic optical model that is obtained from Eq. (3.31) with $\alpha_x = \alpha_y = \alpha$, where $\alpha = \frac{\alpha_x + \alpha_y}{2} \approx 0.53$ nm, and the parameters m , μ , and γ are the same as for the isotropic Drude model. In order to examine the speed intervals where the image force becomes repulsive, this force is displayed in Fig. C.2, as a function of the distance z_0 , for the optical model (red curves), isotropic optical model (dotted green curves), and anisotropic Drude model (dotted blue curves). This figure shows that the anisotropic Drude model consistently results in an interval of distances, $0 \leq z_0 \leq z_c$, where the image force is repulsive. The upper limit, z_c , of this interval appears to increase proportionally with v^2 . Similarly, Fig. C.3 shows that the image force becomes positive in the anisotropic Drude model at speeds $v > v_c$, with a lower cut-off speed v_c that scales with $\sqrt{z_0}$. Note that including the interband contribution in the optical model significantly reduces the image force to negative values at small distances, making it similar to the isotropic model. The interband contribution also lowers the image force to negative values at speeds $v \gtrsim v_c$, but this effect diminishes with increasing distance. Consequently, the optical model exhibits a domain of large distances and high speeds where the image force remains positive, as illustrated in Fig. C.3b. Thus, it is confirmed that the anisotropic nature of the Drude contribution in the optical model is responsible for the positive image force, while the interband contribution tends to lower these values to negative. In contrast, the isotropic model never produces a repulsive image force (green curves in Figs. (C.2) and (C.3)).

Finally, we investigate the role of anisotropy in producing positive values of the dynamic image force above freestanding phosphorene for angles near the AC direction. As shown in Figs. (C.2) and (C.3), for $\theta_0 = 0$, the anisotropy in the Drude model causes the image force to become positive when the combination of speed and distance, given by v^2/z_0 , exceeds a certain threshold for the parameters used here. To explore this further, Fig. C.4 displays the reduced image force for $\theta_0 = 0$ from the Drude model. Here, we keep the product of the reduced effective electron masses fixed at the *ab initio* value, $\frac{m_x m_y}{m_0^2} = 0.149$, while varying their ratio $R = m_x/m_y$ from $R = 0.1$ (very anisotropic) to $R = 1$ (isotropic) in steps of 0.1. The results for the normalized speed fixed at $\bar{v} = 5$ are shown in Fig. C.4a, where the horizontal axis represents the distance z_0 . In Fig. C.4b, the normalized image force is plotted as a function of speed $\bar{v}$, for the distance fixed at $z_0 = 20$ nm. Even a slight deviation from isotropy causes the image force to become positive for distances $z_0 < z_c$ at fixed $\bar{v}$ and for speeds $\bar{v} > \bar{v}_c$ at fixed z_0 , with z_c and $\bar{v}_c$ depending on the mass ratio R in a non-monotonic manner.

However, as shown in Figs. (C.2) and (C.3), including the interband contribution in the optical model reduces the Drude model's effect, thereby lowering the image force values. At

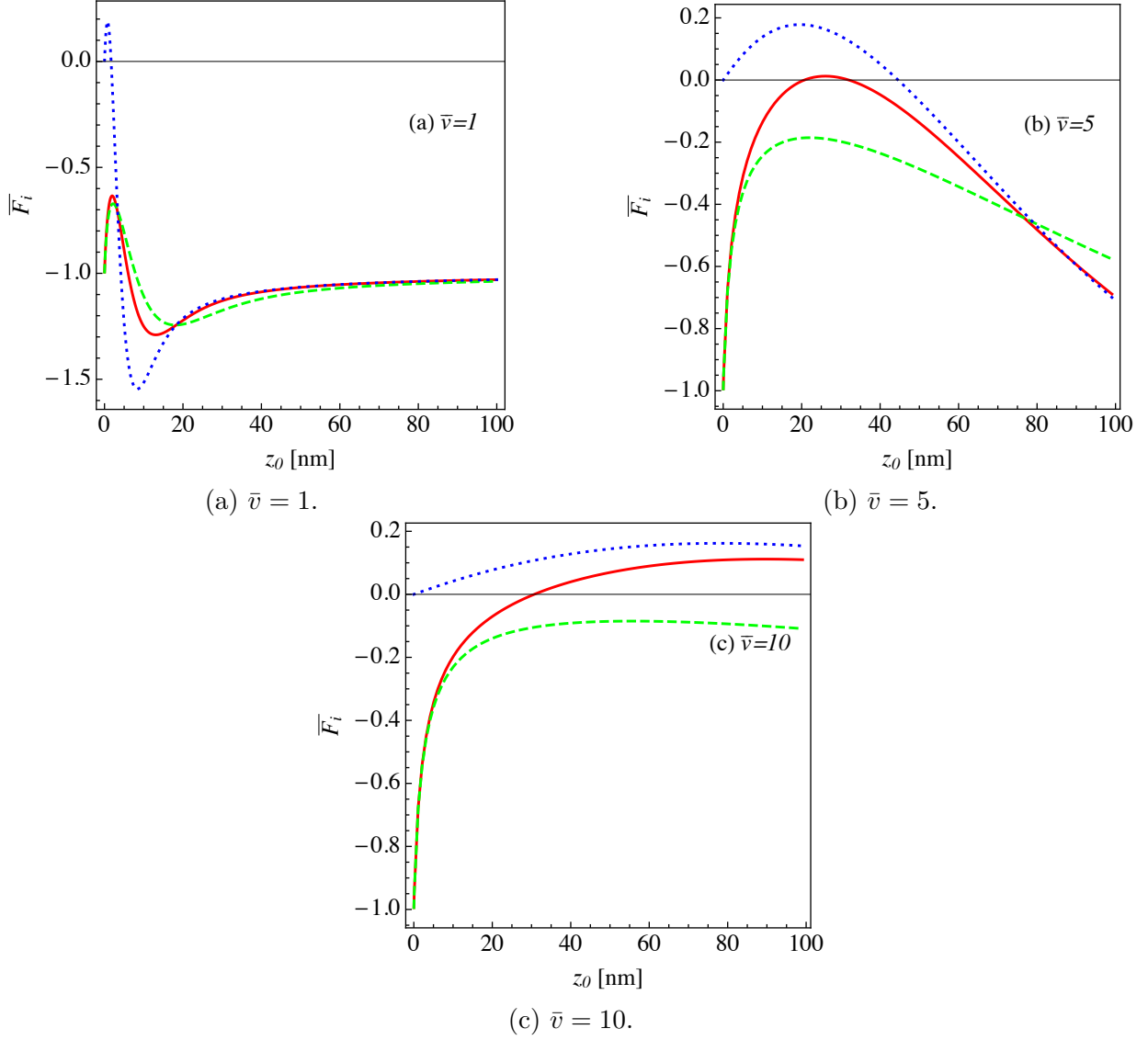


Figure C.2: Normalized image force $\bar{F}_i = \frac{F_i}{F_0}$ as a function of distance z_0 for three different reduced speeds $\bar{v}$, with $\theta_0 = 0$, for free phosphorene. Results from the anisotropic Drude, isotropic optical, and optical models are plotted in blue, green, and red, respectively.

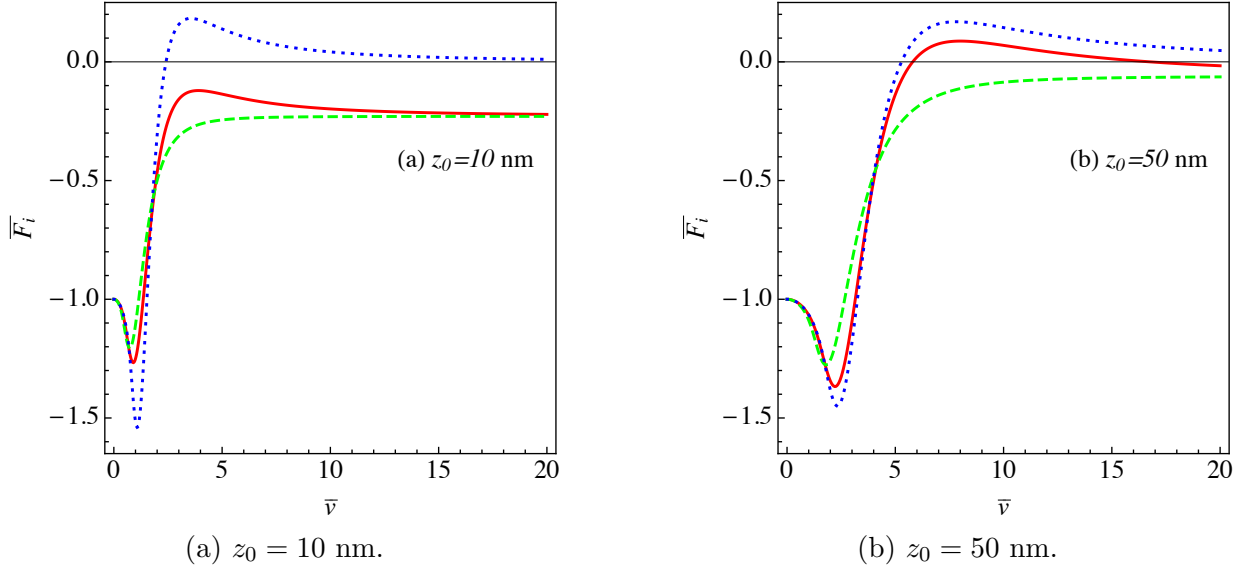


Figure C.3: Normalized image force $\bar{F}_i = \frac{F_i}{F_0}$ as a function of reduced speed $\bar{v}$ for $z_0 = 10$ nm and 50 nm, with $\theta_0 = 0$, for free phosphorene. Results from the anisotropic Drude, isotropic optical, and optical models are plotted in blue, green, and red, respectively.

lower speeds and shorter distances, both the optical and semiclassical models can produce negative image force values, while positive values are observed in other scenarios. Given that the integration domain for the image force in Eq. (4.5) involves high frequencies, a full description for interband electron transitions over a broad frequency range might take over the anisotropic Drude model, resulting in a negative image force for all values of θ_0 , v , and z_0 .

Next, we investigate the influence of non-local effects on free-standing phosphorene, focusing on the three force components experienced by a charged particle moving at a short distance of $z_0 = 5$ nm at low speeds $v < v_B$. In Fig. C.5, we present the results for the image force and the longitudinal stopping force for two angles of incidence: $\theta_0 = 0$ and $\theta_0 = \pi/2$. Obviously, the results from the optical and semiclassical models are adequately close to each other (even for such a small distance z_0), confirming the assertion that the optical model captures the main properties of doped phosphorene in the range of values for v and z_0 that are used in this research. At the same time, the Drude model gives results that are both quantitatively and qualitatively different from the other two models, showing that the interband correction plays an important role at low speeds and relatively short distances.

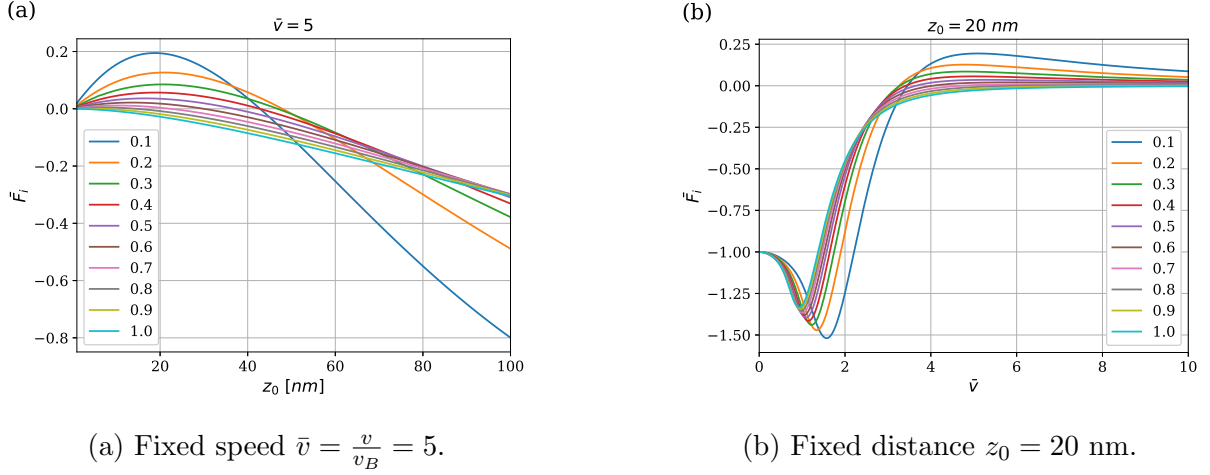
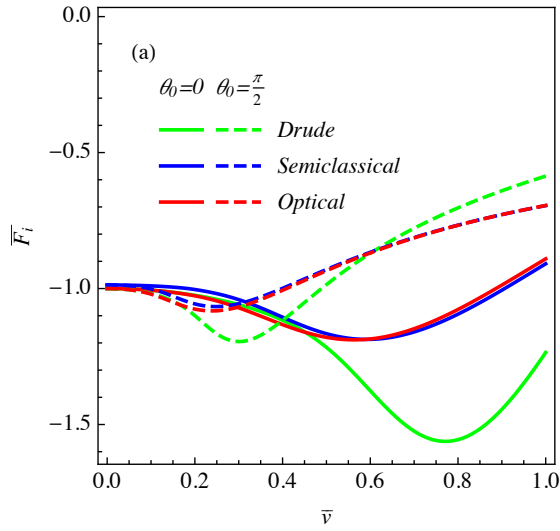


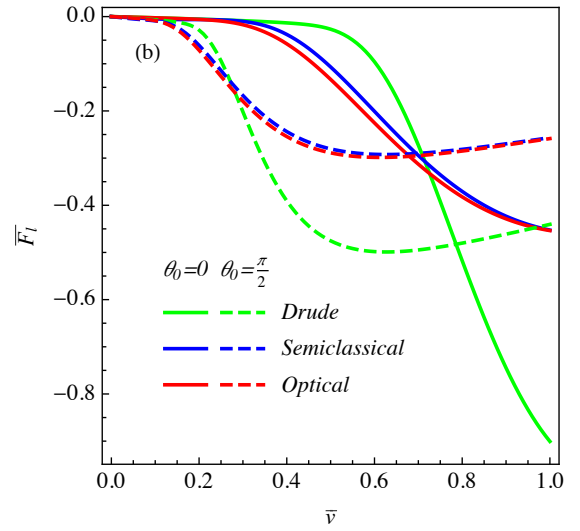
Figure C.4: Normalized image force $\bar{F}_i = \frac{F_i}{F_0}$, obtained from the anisotropic Drude model for doped phosphorene with $n = 10^{13} \text{ cm}^{-25}$. The effective mass ratio $R = \frac{m_x}{m_y}$ is varied in an interval from $R = 0.1$ to $R = 1$ in steps of 0.1, whereas the product of the effective masses is constant, $m_x m_y = 0.149 \times m_0^2$.

In Fig. C.6, we present the transverse component of the stopping force, as a function of the incidence angle θ_0 , for three different values of the reduced speed. The results from the optical and semiclassical models in this figure are quite close to each other, which confirms the adequacy of the Drude model for our choice of the particle related parameters. Note that the slight quantitative disagreement is acceptable, since Figs. (C.5) and (C.6) are obtained with $z_0 = 5$ nm and we know that increasing the distance z_0 , increases the accuracy of the Drude model in prediction of the forces.

To finish the discussion of the forces, in Figs. C.7, C.8, and C.9, we present the variations of the three force components obtained from the semiclassical model, using the same parameters as in computations based on the optical model in Figs. (4.4), (4.5), and (4.6), but with $\gamma_x = \gamma_y = 10$ meV. The surfaces shown in these figures appear indistinguishable from those obtained from the optical model. This similarity indicates that non-local effects have minimal impact on all three forces at a distance of $z_0 = 50$ nm within the speed range $0 < \bar{v} < 20$.



(a) Normalized image force $\bar{F}_i = \frac{F_i}{F_0}$.



(b) Normalized longitudinal stopping force $\bar{F}_l = \frac{F_l}{F_0}$.

Figure C.5: Normalized image and longitudinal forces, as a function of the normalized speed $\bar{v} = \frac{v}{v_B}$, with distance $z_0 = 5$ nm. Results from $\theta = 0$ and $\pi/2$ are shown with solid and dashed lines, respectively. Calculations are performed with the Drude model (green curves), optical model (red curves) and the semiclassical model (blue curves).

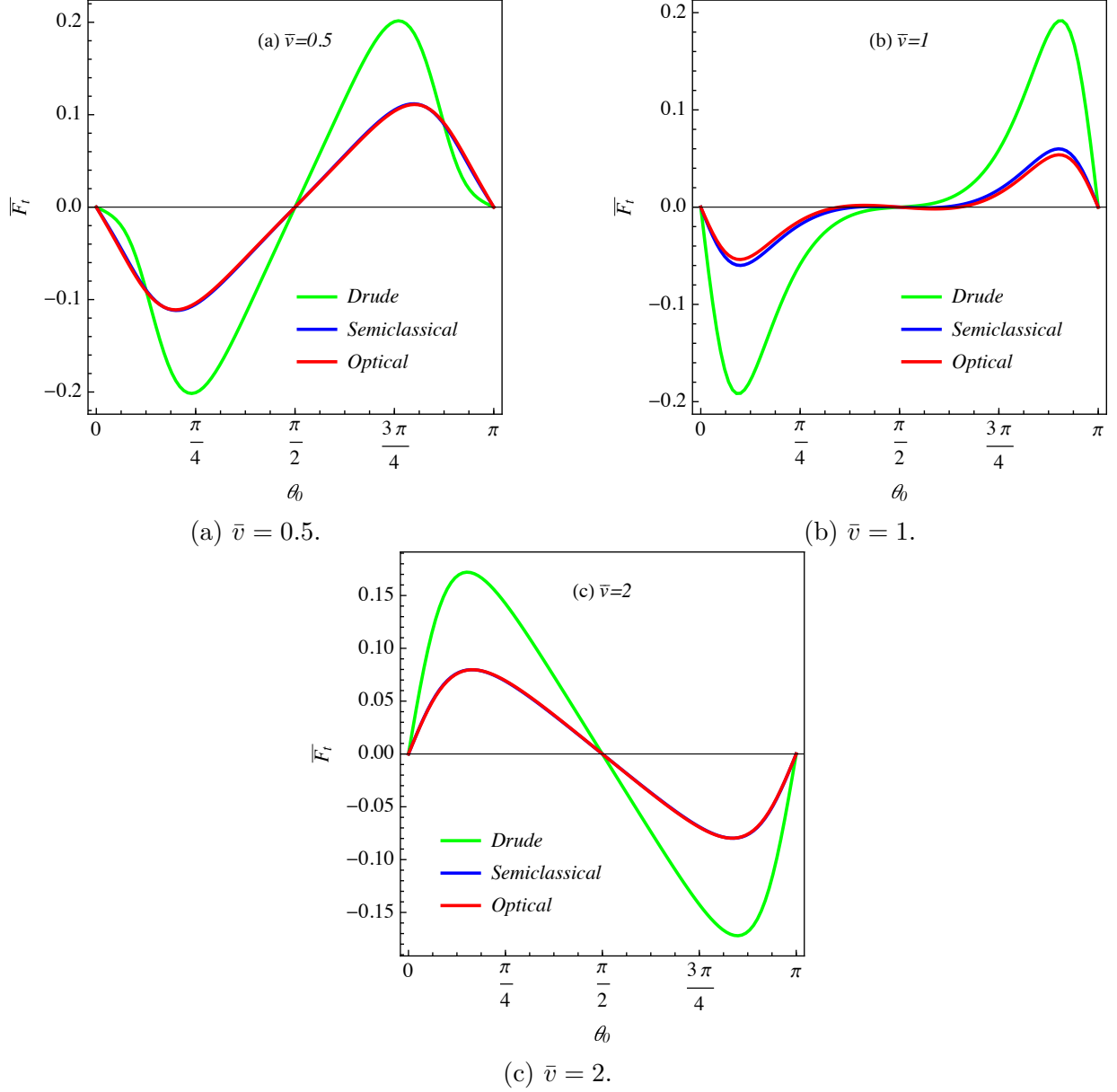


Figure C.6: Normalized transverse stopping force $\bar{F}_t = \frac{F_t}{F_0}$ as a function of incidence angle θ_0 for three different reduced speeds $\bar{v}$, with $z_0 = 5$, for free phosphorene. Results from the anisotropic Drude, optical, and semiclassical models are plotted in green, red, and blue, respectively.

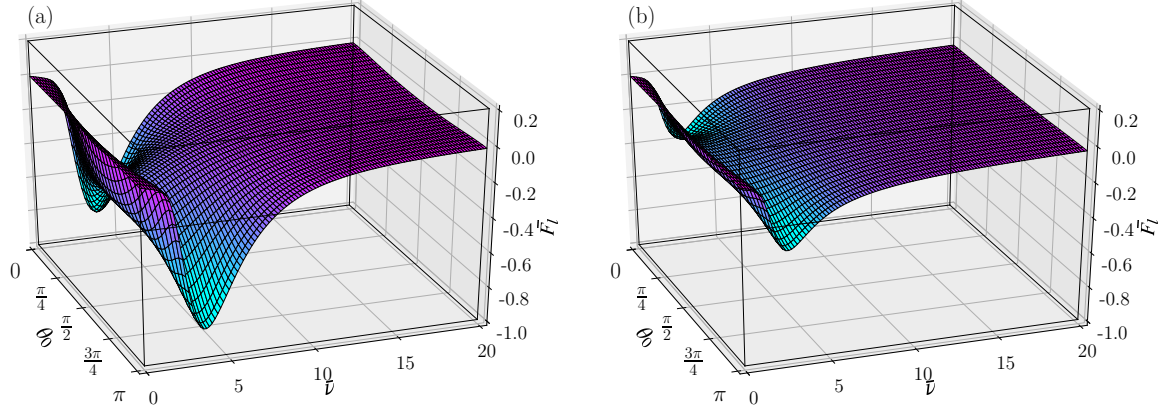


Figure C.7: Normalized longitudinal stopping force $\bar{F}_l = F_l/F_0$ as a function of normalized speed $\bar{v} = v/v_B$ and the direction angle θ_0 , obtained from the semiclassical model for a particle moving at height $z_0 = 50$ nm above: (a) free phosphorene and (b) phosphorene supported by SiO_2 .

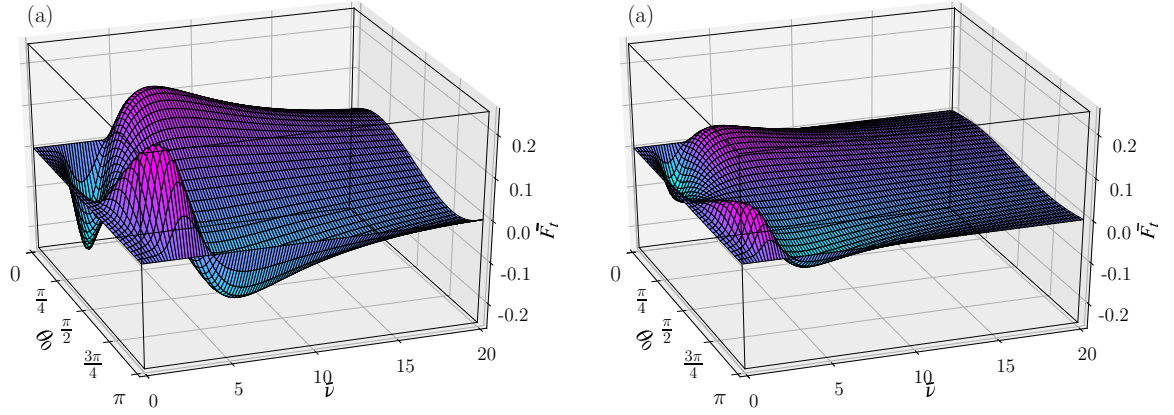


Figure C.8: Normalized transverse stopping force $\bar{F}_t = F_t/F_0$ as a function of normalized speed $\bar{v} = v/v_B$ and the direction angle θ_0 , obtained from the semiclassical model, for a particle moving at height $z_0 = 50$ nm above: (a) free phosphorene and (b) phosphorene supported by SiO_2 .

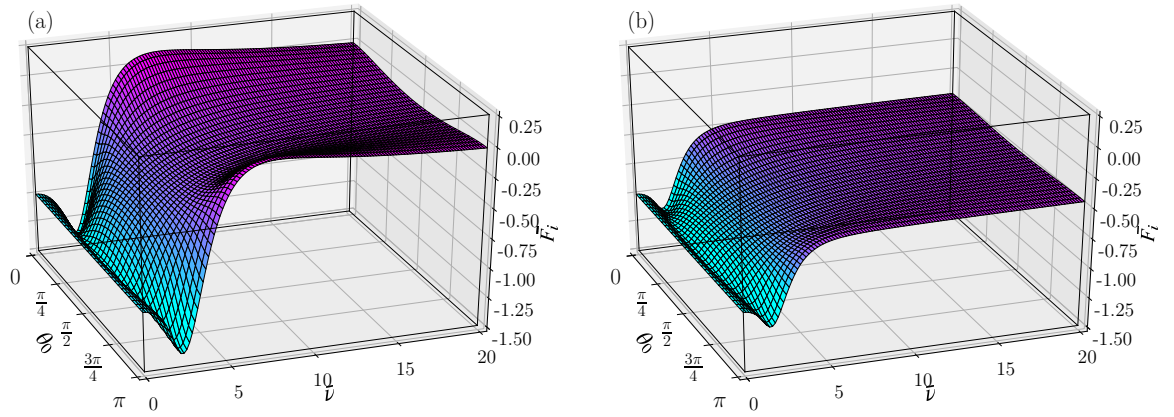


Figure C.9: Normalized image force $\bar{F}_i = F_i/F_0$ as a function of normalized speed $\bar{v} = v/v_B$ and the direction angle θ_0 , obtained from the semiclassical model, for a particle moving at height $z_0 = 50$ nm above: (a) free phosphorene and (b) phosphorene supported by a SiO_2 substrate.

Appendix D

Structure factor

In this appendix we derive a simplified expression for the structure factor

$$\mathcal{S}(\mathbf{q}) = \frac{1}{Ne^2Z^2} \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \langle \delta\tilde{\rho}_0(-\mathbf{q}, z) \delta\tilde{\rho}_0(\mathbf{q}, z') \rangle, \quad (\text{D.1})$$

where $\delta\tilde{\rho}_0(\mathbf{q}, z) = \tilde{\rho}_0(\mathbf{q}, z) - \langle \tilde{\rho}_0(\mathbf{q}, z) \rangle$ is the Fourier transform of $\delta\rho_0(\mathbf{r}, z) = \rho_0(\mathbf{r}, z) - \langle \rho_0(\mathbf{r}, z) \rangle$, with

$$\rho_0(\mathbf{r}, z) = eZ \sum_{j=1}^N \delta(\mathbf{r} - \mathbf{r}_j) \delta(z - z_j) \quad (\text{D.2})$$

being the random distribution of the CIs inside the substrate. Since

$$\langle \delta\tilde{\rho}_0(-\mathbf{q}, z) \delta\tilde{\rho}_0(\mathbf{q}, z') \rangle = \langle \tilde{\rho}_0(-\mathbf{q}, z) \tilde{\rho}_0(\mathbf{q}, z') \rangle - \langle \tilde{\rho}_0(-\mathbf{q}, z) \rangle \langle \tilde{\rho}_0(\mathbf{q}, z') \rangle, \quad (\text{D.3})$$

the function $\mathcal{S}(\mathbf{q})$ can be written as $\mathcal{S}(\mathbf{q}) = \frac{P - Q}{Ne^2Z^2}$, with

$$P = \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \langle \tilde{\rho}_0(-\mathbf{q}, z) \tilde{\rho}_0(\mathbf{q}, z') \rangle, \quad (\text{D.4})$$

and

$$Q = \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \langle \tilde{\rho}_0(-\mathbf{q}, z) \rangle \langle \tilde{\rho}_0(\mathbf{q}, z') \rangle. \quad (\text{D.5})$$

Moreover, the Fourier transform of the charge distribution $\rho_0(\mathbf{r}, z)$ is given by

$$\tilde{\rho}_0(\mathbf{q}, z) = \iint d^2\mathbf{r} e^{-i\mathbf{q}\cdot\mathbf{r}} \rho_0(\mathbf{r}, z) = eZ \sum_{j=1}^N e^{-i\mathbf{q}\cdot\mathbf{r}_j} \delta(z - z_j). \quad (\text{D.6})$$

Hence, P can be written as

$$\begin{aligned} P &= \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle eZ \sum_{j=1}^N e^{i\mathbf{q}\cdot\mathbf{r}_j} \delta(z - z_j) \times eZ \sum_{k=1}^N e^{-i\mathbf{q}\cdot\mathbf{r}_k} \delta(z' - z_k) \right\rangle \\ &= e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle \sum_{j=1}^N \sum_{k=1}^N e^{i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_k)} \delta(z - z_j) \delta(z' - z_k) \right\rangle \\ &= e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle \sum_{j=1}^N \delta(z - z_j) \delta(z' - z_j) + \sum_{j=1}^N \sum_{\substack{k=1 \\ k \neq j}}^N e^{i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_k)} \delta(z - z_j) \delta(z' - z_k) \right\rangle \\ &= P_1 + P_2, \end{aligned} \quad (\text{D.7})$$

with

$$P_1 = e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle \sum_{j=1}^N \delta(z - z_j) \delta(z' - z_j) \right\rangle \quad (\text{D.8})$$

and

$$P_2 = e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle \sum_{j=1}^N \sum_{\substack{k=1 \\ k \neq j}}^N e^{i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_k)} \delta(z - z_j) \delta(z' - z_k) \right\rangle. \quad (\text{D.9})$$

The averaging operations in Eqs. (D.8) and (D.9) can be analytically performed by means of classical 1- and 2-particle joint probability density functions $F_1(\mathbf{r}_1, z_1)$ and $F_2(\mathbf{r}_1, z_1; \mathbf{r}_2, z_2)$ for impurity positions, which gives

$$\begin{aligned} P_1 &= e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left[\iint d^2\mathbf{r}_1 \int_{-\infty}^0 dz_1 F_1(\mathbf{r}_1, z_1) \delta(z - z_1) \delta(z' - z_1) \right] \\ &= N e^2 Z^2 \int_{-\infty}^0 dz_1 f_1(z_1) e^{2qz_1}, \end{aligned} \quad (\text{D.10})$$

and

$$\begin{aligned}
P_2 &= e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \\
&\times \left[\iint d^2 \mathbf{r}_1 \int_{-\infty}^0 dz_1 \iint d^2 \mathbf{r}_2 \int_{-\infty}^0 dz_2 F_2(\mathbf{r}_1, z_1; \mathbf{r}_2, z_2) e^{i\mathbf{q} \cdot (\mathbf{r}_1 - \mathbf{r}_2)} \delta(z - z_1) \delta(z' - z_2) \right] \\
&= N e^2 Z^2 n_{\text{imp}} \iint d^2 \mathbf{r} \int_{-\infty}^0 dz_1 \int_{-\infty}^0 dz_2 e^{i\mathbf{q} \cdot \mathbf{r}} e^{q(z_1 + z_2)} f_1(z_1) f_1(z_2) g(\|\mathbf{r}\|; z_1, z_2). \quad (\text{D.11})
\end{aligned}$$

Notice that in the derivation of Eqs. (D.10) and (D.11), we made use of the sifting property of the Dirac delta function, the Fubini's theorem, Eqs. (5.15) and (5.16), and the following relations

$$\begin{cases} \iint d^2 \mathbf{r} = A, \\ \frac{N-1}{A} \approx \frac{N}{A} = n_{\text{imp}}. \end{cases} \quad (\text{D.12})$$

On the other hand, using the same assumptions and Eq. (D.6), Q can be written as

$$\begin{aligned}
Q &= \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \left\langle eZ \sum_{j=1}^N e^{i\mathbf{q}\cdot\mathbf{r}_j} \delta(z - z_j) \right\rangle \left\langle eZ \sum_{j=1}^N e^{-i\mathbf{q}\cdot\mathbf{r}_j} \delta(z' - z_j) \right\rangle \\
&= e^2 Z^2 \int_{-\infty}^0 dz \int_{-\infty}^0 dz' e^{q(z+z')} \\
&\quad \times \left[\iint d^2\mathbf{r}_1 \int_{-\infty}^0 dz_1 F_1(\mathbf{r}_1, z_1) e^{i\mathbf{q}\cdot\mathbf{r}_1} \delta(z - z_1) \right] \left[\iint d^2\mathbf{r}_2 \int_{-\infty}^0 dz_2 F_1(\mathbf{r}_2, z_2) e^{-i\mathbf{q}\cdot\mathbf{r}_2} \delta(z' - z_2) \right] \\
&= e^2 Z^2 \left[\iint d^2\mathbf{r}_1 \int_{-\infty}^0 dz_1 \frac{N}{A} f_1(z_1) e^{qz_1} e^{i\mathbf{q}\cdot\mathbf{r}_1} \right] \left[\iint d^2\mathbf{r}_2 \int_{-\infty}^0 dz_2 \frac{N}{A} f_1(z_2) e^{qz_2} e^{-i\mathbf{q}\cdot\mathbf{r}_2} \right] \\
&= Ne^2 Z^2 n_{\text{imp}} \iint d^2\mathbf{r}_1 \int_{-\infty}^0 dz_1 \iint d^2\mathbf{r}_2 \int_{-\infty}^0 dz_2 \frac{1}{A} e^{i\mathbf{q}\cdot(\mathbf{r}_1 - \mathbf{r}_2)} e^{q(z_1 + z_2)} f_1(z_1) f_1(z_2) \\
&= Ne^2 Z^2 n_{\text{imp}} \iint d^2\mathbf{r} \int_{-\infty}^0 dz_1 \int_{-\infty}^0 dz_2 e^{i\mathbf{q}\cdot\mathbf{r}} e^{q(z_1 + z_2)} f_1(z_1) f_1(z_2). \tag{D.13}
\end{aligned}$$

Note that Q has the same form as P_2 , except for the factor $g(\|\mathbf{r}\|; z_1, z_2)$. Therefore, the structure factor $\mathcal{S}(\mathbf{q}) = \frac{P - Q}{Ne^2 Z^2}$ can be written as

$$\mathcal{S}(\mathbf{q}) = \int_{-\infty}^0 dz f_1(z) e^{2qz} + n_{\text{imp}} \iint d^2\mathbf{r} \int_{-\infty}^0 dz_1 \int_{-\infty}^0 dz_2 e^{i\mathbf{q}\cdot\mathbf{r}} e^{q(z_1 + z_2)} f_1(z_1) f_1(z_2) [g(\|\mathbf{r}\|; z_1, z_2) - 1], \tag{D.14}$$

upon using the expressions that we found for P_1 , P_2 , and Q . This expression can be further simplified by converting the integration in $\mathbf{r}$ from Cartesian coordinates to polar coordinates

as

$$\begin{aligned}
& \iint d^2\mathbf{r} \int_{-\infty}^0 dz_1 \int_{-\infty}^0 dz_2 e^{i\mathbf{q}\cdot\mathbf{r}} e^{q(z_1+z_2)} f_1(z_1) f_1(z_2) [g(|\mathbf{r}|; z_1, z_2) - 1] \\
&= \int_0^\infty dr \int_0^{2\pi} d\theta \int_{-\infty}^0 dz_1 \int_{-\infty}^0 dz_2 r e^{iqr \cos \theta} e^{q(z_1+z_2)} f_1(z_1) f_1(z_2) [g(r; z_1, z_2) - 1].
\end{aligned} \tag{D.15}$$

This transformation also reveals that for the distribution of the CIs used in our study, the structure factor only depends on the magnitude of the wavenumber q . Therefore, the structure factor is given by

$$\begin{aligned}
\mathcal{S}(q) &= \int_{-\infty}^0 dz f_1(z) e^{2qz} \\
&+ 2\pi n_{\text{imp}} \int_{-\infty}^0 dz_1 f_1(z_1) e^{qz_1} \int_{-\infty}^0 dz_2 f_1(z_2) e^{qz_2} \int_0^\infty dr r J_0(qr) [g(r; z_1, z_2) - 1],
\end{aligned} \tag{D.16}$$

in which we made use of the identity [\[132\]](#)

$$J_0(x) = \frac{1}{2\pi} \int_0^{2\pi} d\theta e^{ix \cos \theta}. \tag{D.17}$$

Appendix E

Double iteration scheme; a modified iteration method to solve non-linear integral equations

In this appendix, an iteration scheme that can be used to solve linear and non-linear integral equations is introduced. Although no rigorous discussion is given on the convergence and its necessary conditions, this new scheme is able to solve a wide variety of linear and non-linear Fredholm's integral equations for whose solving the ordinary iteration techniques are not successful. The main idea of this modified iteration scheme for solving Fredholm's integral equations is breaking the integration domain at a variable x and taking one of the integrals into the non-homogeneous term followed by solving the resulting Volterra's equation with ordinary iteration schemes. Since during this procedure, the non-homogeneous term of the resulting Volterra equation depends on an unknown function, two loops of iteration are required. At each step we use the previous approximation of the solution to define the non-homogeneous term, and solve the resulting Volterra equation to find the new approximation. Therefore, we may call this method a double iteration scheme (DIS). Moreover, we can generalize this DIS to solve non-linear integral equations. For this purpose, a Volterra integral equation can be made from the given non-linear integral equation in such a way that the solution of this Volterra's equation is a solution of the initial non-linear integral equation and vice-versa.

E.1 Volterra and Fredholm integral equations

The equation

$$u(x) = f(x) + \lambda \int_a^x K(x, t)u(t)dt, \quad (\text{E.1})$$

where λ and a are constants and $f(x)$ and $K(x, t)$ are known functions is called a linear integral equation of the second kind or *Volterra's integral equation*. On the other hand the equation

$$u(x) = f(x) + \lambda \int_a^b K(x, t)u(t)dt, \quad (\text{E.2})$$

that differs from Eq. (E.1) in the upper limit of the integral (b is constant), is called *Fredholm's integral equation*.

It has been shown that the ordinary iteration method will always converge to the solution of the Volterra integral equation [180]. The procedure is as follows: I) Start with a continuous function $u_0(x)$ as the initial guess. II) Put this initial guess in the right-hand side of the Volterra integral equation Eq. (E.1) and name the result $u_1(x)$, which is the first approximation. III) Continue this procedure by repeating step II with using the last obtained approximation $u_{n-1}(x)$ instead of the initial guess. Continuing this procedure produces a sequence of functions $u_0(x)$, $u_1(x)$, $u_2(x)$, $\dots$, $u_n(x)$ satisfying

$$u_{n+1}(x) = f(x) + \lambda \int_a^x K(x, t)u_n(t)dt. \quad (\text{E.3})$$

Note that the sequence $u_n(x)$, defined by Eq. (E.3) converges to the solution of the Volterra's equation, regardless of the initial guess $u_0(x)$. Nevertheless, choosing a good initial guess will increase the convergence rate and reduce the required computational power. On the other hand, solving Fredholm integral equations using the iteration method is infeasible unless the kernel function $K(x, t)$ and the parameter λ meet certain restrictive conditions.

E.2 Double Iteration Scheme (DIS)

In this section we show that Fredholm integral equations could be transformed into a new integral equation requiring a double iteration scheme to be solved. For simplicity, these two iteration schemes are named *internal* and *external* iterations. The significance of this technique is that one of its iteration loops (the internal iteration) is just an iteration on a Volterra integral equation for which the convergence is ensured. On the other hand, the external loop has a more complicated mathematical structure, and despite the lack of a proof of convergence, numerical computations show that this external loop also converges in many practical cases. This allows the DIS method to solve Fredholm integral equations that the ordinary iteration scheme does not.

Consider the Fredholm integral equation

$$u(x) = f(x) + \lambda \int_a^b K(x, t)u(t)dt, \quad (\text{E.4})$$

which could be written as

$$u(x) = f(x) + \lambda \int_x^b K(x, t)u(t)dt + \lambda \int_a^x K(x, t)u(t)dt. \quad (\text{E.5})$$

Let us define

$$F(x, u) = f(x) + \lambda \int_x^b K(x, t)u(t)dt, \quad (\text{E.6})$$

hence Eq. (E.5) becomes

$$u(x) = F(x, u) + \lambda \int_a^x K(x, t)u(t)dt, \quad (\text{E.7})$$

which is a Volterra integral equation in which the non-homogeneous term is a function of x and a function of the unknown function $u(x)$. DIS uses the following scheme to solve Eq. (E.7):

DIS:

- 1) Choose a continuous initial guess for the solution and name it $u_0^0(x)$.
- 2) Construct the recursive relation

$$u_{n+1}^{m+1}(x) = F_n + \lambda \int_a^x K(x, t) u_{n+1}^m(t) dt, \quad (\text{E.8})$$

with $F_n = F(x, u_n(x))$ based on Eq. (E.6), where $u_n(x) = \lim_{m \rightarrow \infty} u_n^m(x)$ and $u_0(x) = u_0^0(x)$.

- 3) Solve the Volterra integral equation in step (2) by iteration on m to obtain $u_{n+1}(x) = \lim_{m \rightarrow \infty} u_{n+1}^m(x)$ from $u_n(x)$, starting with $u_{n+1}^0(x) = u_n(x)$.
- 4) Repeat step (3) until the sequence $\{u_n(x)\}$ converges.

Notes:

Step (3) is just solving a Volterra integral equation using the iteration method, this iteration is called the internal iteration loop. Step (4) is in fact an iteration loop to find the limit of the sequence $u_n(x)$, which is called the external iteration loop. While the convergence in step (3) is ensured, no rigorous proof is available for the convergence of the external iteration loop in step (4). Numerical computations generally show stable and fast convergence behavior, although there are some cases in which the external iteration diverges. Moreover, note that in the DIS, the solution of the integral equation is obtained as the limit of a double index sequence

$$u(x) = \lim_{n \rightarrow \infty} \left[\lim_{m \rightarrow \infty} u_n^m(x) \right], \quad (\text{E.9})$$

provided that this limit exists, under the constraint that $u_{n+1}^m(x)$ depends on $u_n^m(x)$, and $u_n^{m+1}(x)$ depends on $u_n^m(x)$, hence, the order of the limits in Eq. (E.9) can not be changed. Note that in this viewpoint, the inner limit presents the internal loop, while the outer limit presents the external loop.

E.3 Non-linear integral equations

A non-linear integral equation could have many different forms. In this study, by "*non-linear integral equation*" we refer to an equation having the form

$$u(x) = f(x) + \int_a^b H(x, t, u(t))dt, \quad (\text{E.10})$$

where $H(x, t, u)$ is some *well-behaved* function of three variables, $f(x)$ is the non-homogeneous term, and $u(x)$ is the unknown function. Note that with $H(x, t, u) = K(x, t)u$ this non-linear integral equation reduces to Eq. (E.2) which is the linear Fredholm's integral equation, hence, Eq. (E.10) may be called the non-linear Fredholm's integral equation.

It is possible to transform the non-linear Fredholm's integral equation to an integral equation which has the form of Eq. (E.7). Then the DIS technique might be applied to find $u(x)$, which is the solution of the non-linear integral equation. Eq. (E.10) could be written as

$$u(x) = f(x) + \int_x^b H(x, t, u(t))dt + \int_a^x H(x, t, u(t))dt, \quad (\text{E.11})$$

where the integration domain is divided into two parts. Moreover, Eq (E.11) could be rearranged to get

$$u(x) = f(x) + \int_x^b H(x, t, u(t))dt + \int_a^x \left[H(x, t, u(t)) - A(x, t)(u(t) - u_0) + A(x, t)(u(t) - u_0) \right] dt, \quad (\text{E.12})$$

or equivalently

$$u(x) = f(x) + \int_x^b H(x, t, u(t))dt + \int_a^x \left[H(x, t, u(t)) - A(x, t)(u(t) - u_0) \right] dt + \int_a^x A(x, t)(u(t) - u_0), \quad (\text{E.13})$$

where $A(x, t)$ is a *well-behaved* function of two variables and we may call it the *auxiliary function*, and u_0 is an arbitrary number that can facilitate the numerical computation. Therefore, if we define

$$F(x, u) = f(x) + \int_x^b H(x, t, u(t)) dt + \int_a^x \left[H(x, t, u(t)) - A(x, t)(u(t) - u_0) \right] dt, \quad (\text{E.14})$$

then Eq. (E.13) becomes

$$u(x) = F(x, u) + \int_a^x A(x, t)(u(t) - u_0). \quad (\text{E.15})$$

The most natural choice for $A(x, t)$ is the coefficient of the first-order term in the Taylor's expansion of $H(x, t, u)$ in $(u - u_0)$. In fact, we may write

$$\begin{aligned} H(x, t, u) = H(x, t, u_0) &+ \left. \frac{\partial}{\partial u} H(x, t, u) \right|_{u=u_0} \cdot (u - u_0) \\ &+ \frac{1}{2!} \left. \frac{\partial^2}{\partial u^2} H(x, t, u) \right|_{u=u_0} \cdot (u - u_0)^2 + \dots \end{aligned} \quad (\text{E.16})$$

that suggests to set

$$A(x, t) = \left. \frac{\partial}{\partial u} H(x, t, u) \right|_{u=u_0}, \quad (\text{E.17})$$

which is equivalent to a kind of linearization to solve the non-linear Eq. (E.10). Sometimes this linear coefficient $A(x, t)$ defined in Eq. (E.17) vanishes. In this situation, it is possible to find some well-chosen auxiliary function $A(x, t)$ which is non-zero and helps DIS to converge. Note that setting $A(x, t)$ to zero and using DIS is equivalent to solving Eq. (E.10) using an ordinary iteration scheme.

Therefore, if we replace the second step of the DIS algorithm given in Section E.2 with the following step

2) Construct the recursive relation

$$u_{n+1}^{m+1}(x) = F_n + \int_a^x A(x, t) (u_{n+1}^m(t) - u_0) dt, \quad (\text{E.18})$$

with $F_n = F(x, u_n(x))$ based on Eq. (E.14), where $u_n(x) = \lim_{m \rightarrow \infty} u_n^m(x)$ and $u_0(x) = u_0^0(x)$;

then the DIS could be used to solve non-linear integral equations. Note that Eq. (E.18) is a linear Volterra integral equation that has a solution $u_{n+1}(x)$ for every given $u_n(x)$. Hence, It can be seen as a transformation $\mathcal{T}[\cdot]$, satisfying $u_{n+1}(x) = \mathcal{T}[u_n(x)]$.

E.4 Numerical verification

In this section, some linear and nonlinear Fredholm's integral equations are solved using the DIS algorithm introduced in the previous section. In all cases, the numerical integrations are performed using the Trapezoidal rule on a uniform mesh grid. Moreover, for the convergence criterion, we continue the external iteration loop until for some n

$$E_n = \|u_n(x) - u_{n-1}(x)\| \leq \varepsilon, \quad (\text{E.19})$$

where

$$\|u(x)\| = \left[\frac{1}{b-a} \int_a^b |u(x)|^2 dx \right]^{1/2}, \quad (\text{E.20})$$

and ε the critical error that determines the acceptable accuracy.

E.4.1 Case 1: Linear Fredhoml's integral equation

In this section DIS is used to solve the following equation

$$u(x) = x + \lambda \int_0^1 u(t) dt, \quad (\text{E.21})$$

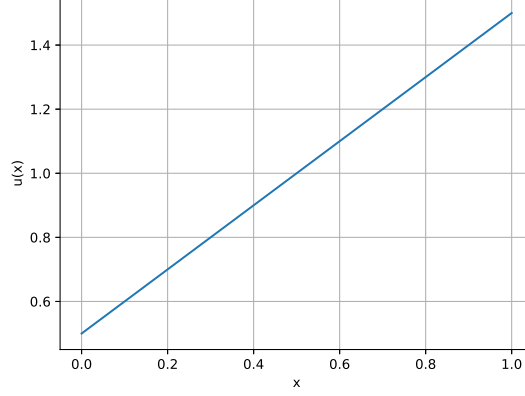


Figure E.1: Solution of Eq. (E.21) for $\lambda = 0.5$.

with the exact solution [180]

$$u(x) = x + \frac{\lambda}{2(1-\lambda)}, \quad \lambda \neq 1. \quad (\text{E.22})$$

All numerical integrations are performed on the following grid

$$x_i = a + i \left(\frac{b-a}{N-1} \right), \quad i \in \{0, 1, 2, \dots, N-1\}, \quad (\text{E.23})$$

so that the numerical approximations $\tilde{u}(x_i)$ are the output of computations. In this case, $a = 0$, $b = 1$, $N = 100$, and $\varepsilon = 10^{-5}$. Eq. (E.21) for two different values of λ . For $\lambda = \frac{1}{2}$, the exact solution is

$$u(x) = x + \frac{1}{2}, \quad (\text{E.24})$$

and the numerical solution is depicted in Fig. E.1. One can see that for $\lambda = \frac{1}{2}$, the numerical solution is equal to the exact solution. Moreover, the ordinary iteration method produces the same solution as DIS produced.

For $\lambda = -2$, the exact solution is

$$u(x) = x - \frac{1}{3}, \quad (\text{E.25})$$

and the numerical solution obtained from DIS is depicted in Fig. E.2. One can see that

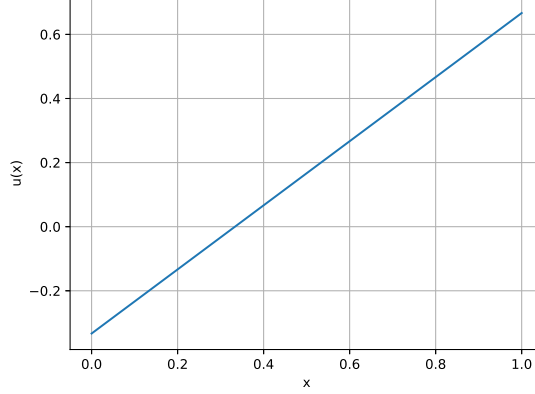


Figure E.2: Solution of Eq. (E.21) for $\lambda = -2$.

for $\lambda = -2$, the numerical solution is equal to the exact solution. Moreover, in this case, the ordinary iteration method diverges and the numerical result in Fig. E.2 represents the effectiveness of DIS algorithm in comparison with ordinary iteration. In fact, numerical computations show that the ordinary iteration only converges for $\lambda \in (-1, 1)$, while the convergence interval of DIS is $\lambda \in (-\infty, 1)$.

E.4.2 Case 2: Non-linear Fredholm's integral equation

In this section DIS is used to solve the following integral equation

$$S(\xi, \omega + \eta i) = \frac{1}{4} \left(\frac{\delta}{a} \right)^2 e^{-\xi^2/4} \int_0^\infty dx \frac{x^2 e^{-x^2/4}}{\gamma(\omega + \eta i) - xS(x, \omega + \eta i)} \left[3I_0\left(\frac{\xi x}{2}\right) - 4I_1\left(\frac{\xi x}{2}\right) + I_2\left(\frac{\xi x}{2}\right) \right], \quad (\text{E.26})$$

which is an important integral equation in physics and its solution can determine the normalized spectral density $\hat{\rho}(\xi, \omega)$ of surface plasmons on randomly rough surfaces through [118]

$$\hat{\rho}(\xi, \omega) = \frac{\xi}{\pi} \cdot \frac{\xi S_I(\xi, \omega) + \gamma_I(\omega)}{[\gamma_R(\omega) - \xi S_R(\xi, \omega)]^2 + [\gamma_I(\omega) + \xi S_I(\xi, \omega)]^2}, \quad (\text{E.27})$$

where the subscripts $_R$ and $_I$ stand for real and imaginary parts, respectively. Moreover, $\gamma(\omega)$ is given by

$$\gamma(\omega) = 1 - \left(\frac{\omega}{\omega_{sp}} \right)^2, \quad (\text{E.28})$$

$I_0(x)$, $I_1(x)$, and $I_2(x)$ are modified Bessel functions, and the constant ratio δ/a is called the corrugation strength [118]. Solving this equation is quite power-consuming from a computational point of view and the convergence rate of the iteration method is low. Therefore, the following integral equation is solved

$$u_{\omega+\eta i}(x) = \frac{1}{4} \left(\frac{\delta}{a} \right)^2 e^{-x^2/4} \int_0^6 dt \frac{t^2 e^{-t^2/4}}{\gamma(\omega + \eta i) - t u_{\omega+\eta i}(t)} \left[3I_0\left(\frac{xt}{2}\right) - 4I_1\left(\frac{xt}{2}\right) + I_2\left(\frac{xt}{2}\right) \right], \quad (\text{E.29})$$

in which the upper limit of the integral is restricted to 6, and the variables and the unknown function are renamed to match our terminology in previous sections, in fact

$$u_{\omega+\eta i}(x) = S(x, \omega + \eta i). \quad (\text{E.30})$$

The mesh grid is obtained from Eq. (E.23) with $a = 0$, $b = 6$, and $N = 31$; and $\varepsilon = 10^{-4}$ is used in the convergence criterion.

Although the ordinary iteration method was able to solve this equation as mentioned in Ref. [118], here we show the results obtained from DIS. In this case

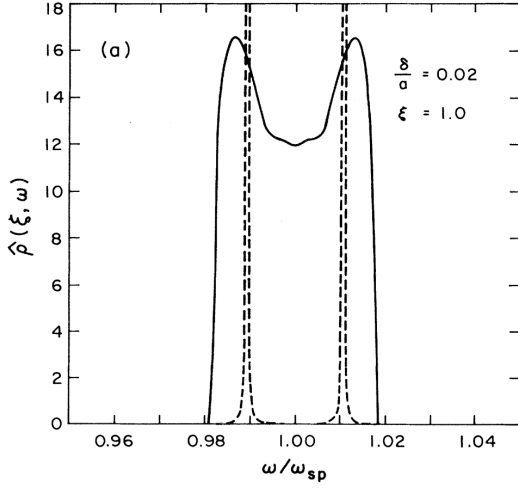
$$H(x, t, u) = \frac{1}{4} \left(\frac{\delta}{a} \right)^2 e^{-x^2/4} \frac{t^2 e^{-t^2/4}}{\gamma(\omega + \eta i) - tu} \left[3I_0\left(\frac{xt}{2}\right) - 4I_1\left(\frac{xt}{2}\right) + I_2\left(\frac{xt}{2}\right) \right], \quad (\text{E.31})$$

and the linear auxiliary function from Eq. (E.17) is given by

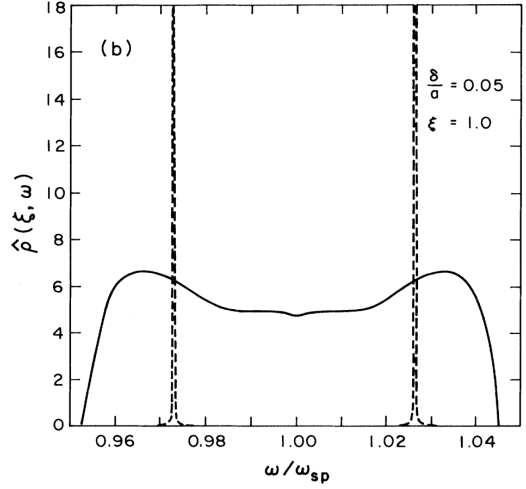
$$A(x, t) = \frac{1}{4} \left(\frac{\delta}{a} \right)^2 e^{-x^2/4} \frac{t^3 e^{-t^2/4}}{[\gamma(\omega + \eta i) - tu_0]^2} \left[3I_0\left(\frac{xt}{2}\right) - 4I_1\left(\frac{xt}{2}\right) + I_2\left(\frac{xt}{2}\right) \right], \quad (\text{E.32})$$

with $u_0 = 2$.

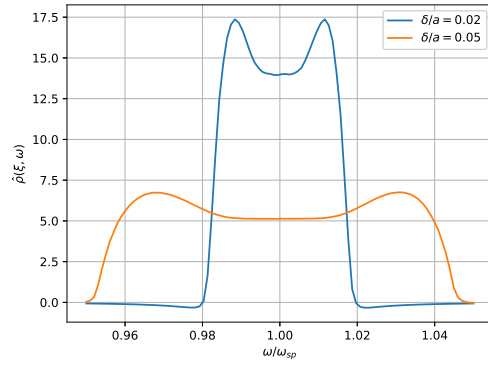
Note that the ordinary iteration method and DIS are able to deal with complex-valued functions, here, $\gamma(\omega + \eta i)$ with $\eta = 10^{-3}$ has complex values and the solution $u_{\omega+\eta i}(x)$ is allowed to be complex as well. To be able to compare the results with those of Ref. [118], the same set of parameters have been chosen and the results are shown in Fig. E.3. Figs. E.3a and E.3b show the results from Ref. [118], while Fig. E.3c shows the results obtained from DIS.



(a) $\delta/a = 0.02$.



(b) $\delta/a = 0.05$.



(c) The blue curve is a reproduction of the solid curve in panel (a), while the orange curve is a reproduction of the solid curve in panel (b).

Figure E.3: Cross-section of spectral density in Eq. (E.27) for $\delta/a = 0.02$, and $\delta/a = 0.05$, with $\xi = 1$.