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Quasiparticles in graphene and other 2D materials: modulation by a surface acoustic wave and contribution to Coulomb drag

TESIS DOCTORAL

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M.Tech

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The ring she wants

The rings I want

The ring I can afford

Doctoral Thesis: Quasiparticles in graphene and other 2D materials: modulation by a surface acoustic wave and contribution to Coulomb drag

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El tribunal nombrado por el Mgfco. y Exmco. Sr. Rector de la Universidad Politécnica de Madrid, el día de 2020, para juzgar la Tesis arriba indicada, compuesto por los siguientes doctores:

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Resumen

Los materiales bidimensionales (2D), como el grafeno, el h-BN, el fósforo negro (BP) o los dicalcogenuros de metales de transición (TMDCs), han mostrado un gran potencial en el área de la electrónica y la fotónica. Su ensamblado en heteroestructuras verticales de van der Waals es uno de los campos de investigación más candentes en la actualidad con un gran número de fenómenos recién observados. En esta tesis, hemos investigado varias cuasipartículas en grafeno y otros materiales 2D, con especial atención a su acoplo y a fenómenos relacionados, así como a su interacción con la luz, donde esta última se modula o asiste mediante una onda acústica superficial (SAW). En particular, hemos estudiado los plasmones en grafeno y sus heteroestructuras, su hibridación con fonones y su acoplo con la luz por medio de una SAW, el efecto de arrastre de Coulomb en grafeno, el acoplo de plasmones y excitones en TMDCs, BP y metal y el efecto de la deformación producida por una SAW en los fonones del grafeno.

Primero hemos estudiado teóricamente los plasmones en sistemas de grafeno monocapa (SLG) y de doble monocapa (DLG) y su acoplo con los fonones de superficie e hiperbólicos de una capa intermedia de h-BN y del sustrato, que dan lugar a polaritones de tipo plasmón-fonón superficial (SPPPs) y plasmón-fonón hiperbólico (HPPPs). Se ha demostrado que la SAW permite generar SPPPs propagantes en sistemas SLG y DLG con capa intermedia de h-BN en un sustrato piezoeléctrico como AlN y ZnO, donde la modulación de la superficie producida por la SAW actúa como una red de difracción proporcionando el momento necesario para generar los polaritones. La capa intermedia de h-BN entre el SLG y el sustrato o entre las capas de grafeno del sistema DLG induce cambios significativos en la dispersión de los SPPPs e incrementa su tiempo de vida.

También hemos estudiado teóricamente las interacciones electrón-electrón en el arrastre de Coulomb en el caso de un sistema DLG separado por una capa intermedia fina de h-BN. Hemos demostrado que estas interacciones pueden estar influenciadas por varias cuasipartículas, como los plasmones y fonones, así como por los efectos de intercambio y correlación (XC). Hemos evaluado numéricamente los efectos de estas cuasipartículas y los XC, y hemos mostrado que todas estas contribuciones en conjunto modifican la resistividad de arrastre permitiendo reproducir con gran precisión los resultados experimentales reportados en la literatura con el modelo teórico desarrollado.

Hemos continuado nuestros estudios teóricos de otros materiales 2D con TMDCs (MoS_2 y WS_2) y BP. Hemos propuesto el acoplo entre los excitones en estos materiales con los plasmones generados en una capa fina de plata mediante una SAW, de una manera similar al caso del grafeno, demostrando que se alcanza el régimen de acoplo fuerte.

Finalmente, hemos demostrado experimentalmente la modulación de los fonones ópticos del grafeno SLG activos en procesos Raman mediante la deformación dinámica creada por un resonador SAW en un sustrato de LiNbO_3 . Aquí hemos mostrado que el campo de deformación de la SAW induce una variación de la intensidad de la dispersión Raman de un 15 % y un desplazamiento en la frecuencia de los fonones de 10 cm^{-1} para una deformación máxima inducida del 0.24 % a una frecuencia SAW de $\sim 400 \text{ MHz}$. Así, se ha probado que las SAWs son herramientas muy potentes para modular las propiedades vibracionales del grafeno a través de las deformaciones locales de alta frecuencia creadas por los transductores acústicos, cuyo uso puede ser extendido a otros materiales 2D y heteroestructuras de van der Waals.

Abstract

Two-dimensional (2D) materials, such as graphene, h-BN, black phosphorus (BP) and transition metal dichalcogenides (TMDCs), have recently shown a lot of potential in the area of electronics and photonics. Assembling them into vertical van der Waals heterostructures is currently one of the hottest research fields with a wide range of newly observed phenomena. In this thesis, we have investigated various quasiparticles in graphene and other 2D materials, with special attention to their coupling and related phenomena as well as their interaction with light, the latter being modulated or mediated by a surface acoustic wave (SAW). In particular, we have addressed plasmons in graphene and its heterostructures, their hybridization with phonons, and their coupling with light by means of SAW, Coulomb drag effect in graphene, exciton-plasmon coupling by SAW in TMDCs, BP and metal and the effect of strain produced by a SAW on the phonons of graphene.

We have first theoretically studied the plasmons in single (SLG) and double layer graphene (DLG) systems and their coupling with surface and hyperbolic phonons of a h-BN interlayer and the substrate leading to hybridized surface plasmon-phonon polaritons (SPPPs) and hyperbolic plasmon-phonon polaritons (HPPPs). We have demonstrated that a SAW can be used to launch propagating SPPPs in SLG and DLG system with h-BN interlayers on a piezoelectric substrate like AlN and ZnO, where the SAW-induced surface modulation acts as a dynamic diffraction grating providing the extra necessary momentum for the generation of the polaritons. The h-BN interlayer between the graphene and the piezoelectric substrate and between the two graphene layers significantly changes the dispersion of the SPPPs and increases their lifetime.

We have also theoretically studied the electron-electron interactions in Coulomb drag in the case of a DLG system separated by a thin h-BN interlayer. We have demonstrated that these interactions can be influenced by various quasiparticles like plasmons and phonons, and also by exchange and correlation (XC) effects. We have numerically evaluated the effect of these quasiparticles and XC on the drag resistivity, and have shown how all these contributions altogether can modify the drag resistivity allowing us to replicate the experimental data reported in the literature by our theoretical model with high accuracy.

We have continued our theoretical studies of 2D materials with TMDCs (MoS₂ and WS₂) and BP. We have proposed the coupling between excitons in these 2D materials and plasmons in a thin silver film by means of a SAW, in a similar way as in the case of graphene, demonstrating that the strong coupling regime is achieved.

Finally, we have put forward the experimental study of the modulation of the Raman-active optical phonon modes of SLG by the dynamic strain created by a SAW resonator on a LiNbO₃ substrate. Here we have showed that the SAW strain field induces a variation in the Raman scattering intensity as large as 15 % and a shift in the phonon modes of up to 10 cm⁻¹ for a maximum strain of 0.24 % at a SAW frequency of ~400 MHz. Thus, SAWs are proven to be powerful tools to modulate the vibrational properties of graphene by means of the high-frequency localized deformations tailored by the acoustic transducers, which can also be extended to other 2D materials and van der Waals heterostructures.

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List of publications

Publications covered in this thesis:

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- **R Fandan**, J Pedrós, F Guinea, A Boscá, F Calle. “*Effect of quasiparticle excitations and exchange-correlation in Coulomb drag in graphene*”. Communications Physics 2 (1) (2019), 1-9.
- **R Fandan**, J Pedrós, J Schiefele, A Boscá, J Martínez, F Calle. “*Acoustically-driven surface and hyperbolic plasmon-phonon polaritons in graphene/h-BN heterostructures on piezoelectric substrates*”. Journal of Physics D: Applied Physics 51 (20) (2018), 204004, (10pp).
- **R Fandan**, J Pedrós, et al. “*Exciton-plasmon coupling in 2D semiconductors by surface acoustic waves*”. (under preparation)

Other publications by the author:

- S Fernández, A Boscá, J Pedrós, A Inés, M Fernández, I Arnedo, J González, M Cruz, D Sanz, A Molinero, **R Fandan**, M Pampillón, F Calle, J Gandía, J Cárabe, J Martínez. “*Advanced graphene-based transparent conductive electrodes for photovoltaic applications*”. Micromachines 10 (6) (2019), 402, (11pp).
- T Tomašević-Ilić, Đ Jovanović, I Popov, **R Fandan**, J Pedrós, M Spasenović, R Gajić. “*Reducing sheet resistance of self-assembled transparent graphene films by defect patching and doping with UV/ozone treatment*”. Applied Surface Science 458 (2018), 446-453.
- M Romero, A Boscá, J Pedrós, J Martínez, **R Fandan**, T Palacios, F Calle. “*Impact of 2D-Graphene on SiN Passivated AlGaIn/GaN MIS-HEMTs Under Mist Exposure*”. IEEE Electron Device Letters 38 (10) (2017), 1441-1444.

For a full list of publications see **Rajveer Fandan** or go to the urls:

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Oral contribution to conferences:

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List of abbreviations

BLG	Bilayer Graphene
BP	Black Phosphorous
BWF	Breit Wigner Fano
CVD	Chemical Vapour Deposition
DLG	Double Layer Graphene
FET	Field Effect Transistor
FTIR	Fourier Transform Infrared Spectroscopy
HPP	Hyperbolic Phonon Polariton
HPPP	Hyperbolic Plasmon Phonon Polariton
IDT	Interdigital Transducer
LFF	Local Field Factor
LP	Lower Polariton
MIR	Mid Infrared
PHC	Particle Hole Continuum
QED	Quantum Electrodynamics
RPA	Random Phase Approximation
SAW	Surface Acoustic Wave
SAWR	Surface Acoustic Wave Resonator
SLG	Single Layer Graphene
SO	Surface Optical
SPP	Surface Plasmon Polariton
SPPP	Surface Plasmon Phonon Polariton
TLM	Transmission Line Method
TMDC	Transition Metal Dichalcogenides
TMM	Transfer Matrix Method
UP	Upper Polariton
XC	Exchange Correlation

List of symbols

α	Fine structure constant
q	Wavevector
E_F	Fermi energy
T_F	Fermi temperature
v_F	Fermi velocity
k_F	Fermi wavevector
$F(E)$	Fermi-Dirac distribution
τ_e	Carrier relaxation time
τ	Carrier transport time
τ_{SO}	Surface optical phonon lifetime
τ_p	Plasmon lifetime
μ	Mobility
μ_c	Chemical potential
μ_D	Drude weight
P_{rf}	Radio frequency power
P_{IDT}	SAW power density
λ_{SAW}	SAW wavelength
q_{SAW}	SAW wavevector
δ	SAW amplitude
ε	Strain
r_s	Wigner-Seitz radius
V	Bare Coulomb interaction
U	Screened Coulomb interaction
$\chi^{(0)}$	Irreducible density-density correlation function
X	Reducible density-density correlation function
L	Loss function
ϵ	Dielectric function
O	Optical Mode
A	Acoustic Mode
G	Local field factor
σ	Conductivity
ρ_D	Drag resistivity
σ_D	Drag conductivity
I	Drag intensity
Γ	Rectification function or non-linear susceptibility
E_g	Band gap
$\hbar\omega_R$	Rabi frequency
g	Coupling strength
J_{DC}	DC current density
ϕ	Electric potential

*To my family
To those who inspired it
and will not read it*

Chapter 1

Introduction

1.1 Overview of the state of the art	1
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1.1 Overview of the state of the art

Since its discovery in 2004, graphene, being a one-atom-thick two-dimensional (2D) material, has opened a new paradigm in condensed matter physics and nanotechnology. Charge carriers (electrons and holes) in graphene behave as massless Dirac fermions due to its unique band structure. A direct consequence is that backscattering of electrons from impurities is forbidden, providing them a huge room temperature mobility of up to $10^5 \text{ cm}^2 \text{ V}^{-1} \text{ s}^{-1}$ [1]. Graphene also presents unique optical, mechanical, and thermal properties such as, a thermal conductivity of $3000 \text{ W m}^{-1} \text{ K}^{-1}$ [2], a Young's modulus of 1 TPa, an intrinsic strength of 130 GPa, and the capability of sustaining an elastic tensile strain of up to 25 % [3]. Among the optical properties, its capability to host surface plasmons in the mid infrared (MIR) and terahertz (THz) ranges has garnered a plethora of research both in the area of theory and experiment. Being carriers in graphene Dirac fermions, surface plasmons are much less lossy in the MIR than those hosted by metals in the visible range. In addition, the carrier density in graphene, or equivalently its Fermi level, can be modulated via electrostatic gating, thus making it an excellent material for tuning light-matter interactions in this wavelength range.

These plasmons once coupled with light, also referred as surface plasmon polaritons (SPPs) (half-matter half-light quasiparticles), can squeeze down the light (photon wavelength compression) beyond the diffraction limit, thus playing a leading role towards the miniaturization of photonic structures, along with their integration with current semiconductor technologies. Plasmons in graphene can confine the light to the order of $1/\alpha^3 (\sim 10^6)$ [4], where α is the fine structure constant. SPPs in metals have attracted a strong interest over the last few decades for manipulating light and light-matter interactions at scales well beyond the diffraction limit [5] and have laid the foundation of a whole new range of fields including SPP-based nanophotonic devices [6, 7], metamaterials [8], metasurfaces [9], and quantum plasmonics [10]. However, the lifetime of SPPs in metals is extremely short and that is where graphene plasmons come into picture with larger lifetime. The lifetime of the graphene plasmons can be further enhanced by isolating the graphene from the substrate underneath by inserting a thin interlayer of hexagonal BN (h-BN), a 2D insulator providing graphene with highly increased carrier mobilities. In graphene/h-BN

systems, the graphene plasmons couple with the phonons in h-BN leading to a larger number of quasiparticles, namely surface plasmon-phonon polaritons (SPPPs) [11] with a lifetime of ~ 1 ps. Graphene plasmons have shown their potential for several applications such as THz lasers [12], antennas [13, 14], plasmonic waveguides and one-atom thick Luneburg lenses [15], modulators [16], polarizers [17], plasmonic light harvesting [18, 19], surface-enhanced Raman scattering [20], plasmonic detectors and sensors [21–24].

Similarly to plasmons in metals, being on the right side of the light line in the dispersion curve (i.e., outside the light cone), free radiation cannot be coupled directly to graphene plasmons, i.e. a large momentum mismatch needs to be overcome, see for example figure 1.1.

FIGURE 1.1: Graphene plasmon dispersion for different E_F levels.

This can be achieved mainly by either using near-field techniques [25–27] or frequency wave mixing [28] or prism coupling [29–32] or tapered materials [33] or patterning the graphene film itself [34–36] or the substrate, where the graphene has been transferred to [37, 38], see for example figure 1.2. Surface acoustic waves (SAWs) have been proposed to overcome momentum mismatch, permitting to launch propagating SPPPs in unpatterned graphene using simple far-field excitation [39]. SAWs are low-frequency acoustic phonons confined to the surface of an unbounded solid [40]. They can be generated by an interdigital transducer (IDT) on a piezoelectric material. Having a large energy density extremely confined at the surface, their usage can be exploited with 2D materials when placed on top of piezoelectric material with maximum interaction efficiency. SAWs were already experimentally demonstrated to couple metal plasmon to light [41, 42]. An IDT on a piezoelectric film can launch a SAW across the graphene sheet creating a tunable optical grating without the need of any patterning in either the graphene layer or the substrate, thus eliminating edge scattering and fragility issues. Furthermore, the propagating plasmons can be switched electrically via the high-frequency signal at the IDT.

Apart from squeezing the light, graphene plasmons can also play a role in carrier transport, for example in Coulomb drag in double layer graphene systems separated by a thin h-BN interlayer, where transport is dominated by momentum and energy transfer between carriers in the two layers [44]. Coulomb drag is a phenomenon which occurs in two 2D electrical conductors placed very close to each other and electrically isolated by an insulating material in between, such that an electric current in one conductor, called active layer, drags carriers inducing a closed-circuit current (or an open-circuit voltage) in the other conductor, called passive layer due to the interlayer electron-electron interactions that can be mediated by phonons

FIGURE 1.2: Various experimental techniques to generate graphene plasmons (ref. [43])

and dipole [45–48]. Coulomb drag is identically zero unless these interactions are present, thus it is an ideal experimental probe to provides a detailed insight into the many-particle interactions in low-dimensional systems, which are at the center of a wide range of condensed matter phenomena such as the fractional quantum Hall effect, the high-temperature superconductivity, the Wigner crystallisation, the exciton condensates, and the Mott transition. The first experimental demonstration of Coulomb drag was carried out using the 2D electron gases (2DEGs) in GaAs double quantum well (QW) structures [49, 50] and, during the next two decades, they were the most used material systems for experiments until the arrival of graphene [51], along with h-BN as the insulating interlayer [52, 53], and the intense theoretical studies that has accompanied them [44, 54–64]. The thickness of the h-BN interlayer can be thinned down to 1 nm (a few h-BN monolayers) thus allowing the two graphene layers to be placed in close vicinity of each other as in contrast with a double quantum well hosting a 2DEG, which are typically 10 nm wide and separated by 15 nm or more. In addition, the graphene/h-BN/graphene (G/h-BN/G) system allows to tune in-situ the Fermi energy (E_F), i.e. the carrier density, by electrostatic gating. This permits to move from the Fermi-liquid regime to the Dirac point ($E_F = 0$). Furthermore, the carrier density (of either electrons (n) or holes (p)) can be easily tuned in the range of 10^{11} to 10^{13} cm^{-2} , thus providing another degree of freedom for tuning the drag resistivity.

Coulomb drag can be enhanced by not only plasmons but also by phonon exchange through the dielectric interlayer. The contribution of both plasmons and phonons was first addressed in doped polar semiconductor heterostructures. The enhancement of the drag by plasmons was shown to become significant for $T \geq 0.2T_F$ [65–67] (see figure 1.3), where T_F is the Fermi temperature. In the case of phonons, at very low temperatures, these materials only showed a contribution from the acoustic phonons [68–71]. However, at higher temperatures, the electron-energy scales (Fermi and plasma energies) become comparable to the LO-phonon energy [72]. This was shown to generate coupled plasmon-LO phonon modes further contributing to the Coulomb drag by virtual phonon exchange [73]. The role of graphene plasmons in Coulomb drag has been evaluated in double layer graphene systems [74] as well as in lateral graphene waveguides [75]. In both cases, it has been shown that the drag resistivity can be enhanced by the plasmon contribution for $T \geq 0.15T_F$. However, none of these studies considered the full frequency-dependent permittivity of the dielectric in between the graphene layers or waveguides but rather a static permittivity, thus lacking the contribution to the Coulomb drag from the phonon exchange mediated by the dielectric material. In G/h-BN/G

FIGURE 1.3: Temperature dependence of the Coulomb drag rate. The full bold (dotted) curve corresponds to calculations using the finite-T ($T = 0$) approximation. OP: optical plasmon; AP: acoustic plasmon (ref. [65]).

systems, the contribution to the Coulomb drag from the virtual phonon exchange through the h-BN interlayer has been studied [76], though only the contribution from the intraband region was taken into account, neglecting that from the interband region, where the plasmons lie. Therefore, there is a lack of a comprehensive study of the effect of plasmons and phonons altogether on the Coulomb drag in graphene. Also, there exists several theoretical models where attempts had been made to fit the experimental data with as high accuracy as possible. But due to complexity added to the calculations, a good agreement between theory and experiment had always been a compromise.

In addition to graphene and h-BN, 2D semiconductors such as, for example, transition metal dichalcogenides (TMDCs) (MoS_2 , WS_2 , etc) and phosphorene/black phosphorus (BP), have also attracted a lot of interest in photonics [77, 78], where excitons dominate the optical properties. Because of their reduced dimensionality, 2D semiconductors present strong light-matter interactions and reduced screening, making them very sensitive to the environment. They also present enhanced Coulomb interactions, which lead to various many-body phenomena, for example, electron-hole quasiparticles, such as excitons, trions, and biexcitons, even at room temperature and with high binding energies. Thus, this provides stronger confinement and more efficient light absorption as compared to quasiparticles in 3D semiconductors. These excitons, in the visible and near infrared ranges, can couple to surface plasmons in metals (see figure 1.4), leading to exciton-plasmon polaritons in the strong coupling regime where the rate of energy transfer exceeds the total damping rate of the system. For enabling this coupling, metal is typically nanostructured to host surface plasmons followed by the transfer of 2D semiconductor onto the nanostructures [79]. The interaction of SAWs with 2D semiconductors and the modulation of their optical and electrical properties has already been demonstrated experimentally [80–82]. Thus, exciton-plasmon coupling might also be facilitated by using SAWs in a similar manner as in graphene, as the SAW can lead to the generation of plasmons in metals [41, 42] which can then couple with optically generated excitons in the 2D semiconductors. A similar SAW-mediated approach has been proven in GaAs based QW heterostructures with metal stripes leading to Tamn plasmon-exciton polaritons [83].

FIGURE 1.4: Plasmon-exciton coupling in planar structures consisting of an atomically thin TMDC crystal deposited onto a metal substrate (ref. [84])

In addition to the undulating the graphene sheet forming a diffraction grating that overcomes the plasmon momentum mismatch, the strain field produced by the SAW deforms the graphene lattice itself, which in turn could be reflected in the modulation of its phonon spectrum. Strain in graphene has been shown to give rise to various extraordinary phenomena such as the shifting of the Dirac cones [85], the shifting and splitting of the graphene Raman modes [86, 87], the enhancement of the electron–phonon coupling and superconductivity [88], the generation of pseudomagnetic fields [89], and the zero-field quantum Hall effect [90]. Strain in graphene has also been reported to affect the interaction of the graphene surface with the environment, leading, for example, to the stabilization of the adsorption of metal atoms by preventing their clustering [91, 92], or to the increase in the coverage and self-assembly of hydrogen atoms [93, 94].

In practice so far, strain in graphene is generated by placing graphene on a stretchable and bendable substrate [86, 87, 95, 96] (see figure 1.5). However, ap-

FIGURE 1.5: (a) Two point and (b) four-point bending of graphene placed on top of a substrate coated with SU8 (ref. [95])

proaches capable of generating strain locally with fast actuation mechanisms are highly desirable for the development of integrated devices. Local strain techniques require transferring the graphene onto nano-patterned substrates [97, 98] or nano-indentation [99]. However, the complexity of most of these methods hinders the usage of graphene in practical device configurations, where the effects induced by the locally generated strain could be exploited. That is where the SAW technique can come handy to locally generate strain on supported graphene (or any other 2D material) which can be actively controlled electrically allowing to change the magnitude of the dynamic strain. Therefore, SAWs might open up new possibilities for strain engineering and straintronics in graphene and more generally in 2D materials [100].

1.2 Goal and objectives

The goal of this thesis is to study the quasiparticles (plasmons, phonons and excitons) arising in graphene and other 2D materials, how they interact with a SAW (being either mediated or modulated by the wave) and how they can contribute to carrier transport. We will theoretically study the plasmon dispersion in single and double layer graphene, understanding the effects of graphene carrier density, their hybridization with the phonons of the piezoelectric substrate and h-BN, and their coupling with light by the strain field of a SAW. We will extend the study of these plasmons and phonons to the Coulomb drag phenomenon in a pair of graphene sheets separated by a thin h-BN interlayer along with the effect of many body corrections by introducing exchange-correlations, also known as local field factors (LFFs), and comparison of these numerical results with experimental data reported in the literature for validation of the theoretical model used. Experimental study of visualization of the strain field of SAW by interferometry, dependence of vertical surface displacement on the applied rf power and the effect of this strain field on the graphene optical phonons will be addressed thoroughly. We will theoretically study exciton-plasmon coupling in 2D semiconductors (MoS_2 , WS_2 and black phosphorous (BP)) and metal (Ag) by means of the strain field of a SAW and will numerically evaluate their coupling efficiency.

1.3 Outline of the thesis

The outline of this thesis is as follows:

In **Chapter 2**, the various methods used in this thesis, including both theoretical models and experimental tools and techniques, are summarized.

In **Chapter 3**, the dispersion of plasmons in graphene and its various heterostructures with h-BN interlayers and AlN and ZnO substrates, their lifetime and their generation efficiency by a SAW are discussed. The case of plasmons in the double layer graphene system and its advantages over single layer graphene case is also discussed.

In **Chapter 4**, Coulomb drag in graphene/h-BN/graphene is discussed. The contribution to drag from quasiparticles and exchange-correlations are quantified and the obtained theoretical results are compared with experimental data from the literature.

In **Chapter 5**, the concept of coupling of excitons in 2D semiconductors and surface plasmons in a thin metal layer on a piezoelectric substrate by a SAW is developed. The generation efficiency is also calculated.

In **Chapter 6**, the experimental study of the strain field of a SAW in graphene by interferometry and the modulation of the phonons in graphene by this dynamic field is presented.

In **Chapter 7**, the important results of the thesis are summarized and the conclusions are presented, indicating also some possible future work to continue these research lines.

Chapter 2

Methods

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2.1 Theory

2.1.1 Random phase approximation (RPA)

The plasmon dispersion in graphene can be calculated within the random phase approximation (RPA). The RPA is a successor of Hartree–Fock approximation for an electron gas with a high density of particles. The RPA is self energy correction to the photon propagator in QED and represents a change in the potential felt by the electrons due to the effect of screening. The idea is that the interaction wiggle creates an electron–hole pair, which annihilates shortly afterwards. The RPA assumes that the electrons respond to the external field plus the time-dependent Hartree potential determined by the external charges and the polarization charges.

The Wigner-Seitz radius for graphene is given as $r_s = \alpha/\kappa$, where α is the fine structure constant in graphene and κ is the background dielectric constant. Thus, this dimensionless parameter is independent of carrier density in graphene unlike in the usual 2D and 3D electron liquids, where it increases with decreasing carrier density. This is a result of the relativistic Dirac-like nature of the free carriers in graphene. For the case of standard SiO₂ substrates ($\kappa = 3.9$), $r_s \sim 0.5$. This small value of r_s indicates that graphene is a weakly interacting system for all carrier densities, making the RPA an excellent approximation since the latter is asymptotically exact in the $r_s < 1$ limit. The two main ingredients for the RPA are the Coulomb interactions, $V(q, \omega)$, and the temperature-dependent irreducible density-density correlation function, $\chi^{(0)}(q, \omega, T)$ [101]. The total dielectric function, $\epsilon_{RPA}(q, \omega, T)$

within the RPA can be written as

$$\epsilon_{RPA}(q, \omega, T) = 1 - V(q, \omega) \chi^{(0)}(q, \omega, T) \quad (2.1)$$

where

$$V(q, \omega) = \frac{e^2}{2q\epsilon(\omega)} \quad (2.2)$$

with $\epsilon(\omega)$ as the background effective permittivity. $\chi^{(0)}(q, \omega, T)$ for graphene is given by the modified Lindhard function

$$\chi^{(0)}(q, \omega, T) = -\frac{1}{\pi^2} \int d^2k \sum_{ss'} f_{ss'}(k, q+k) \frac{F(E_s(q)) - F(E_{s'}(|q+k|))}{E_s(q) - E_{s'}(|q+k|) + \hbar\omega\tau_e^{-1}} \quad (2.3)$$

where $s, s' = \pm 1$ denote the band indices, $E_{\pm}(q) = \pm \hbar v_F q - \mu_c$, μ_c is the chemical potential, $F(E_s(q)) = [\exp(E_s(q)/k_B T) + 1]^{-1}$ is the Fermi-Dirac distribution and $f_{ss'}(k, q+k) = 1/2(1 + ss'k + q \cos \theta / |q+k|)$ is the band overlap wavefunction.

In the limit of small q and $\omega > v_F q$, $\chi^{(0)}$ can be approximated as

$$\chi^{(0)}(q, \omega) \approx \frac{q^2 E_F}{\pi \hbar^2 (\omega + i\tau_e^{-1})^2} \quad (2.4)$$

where E_F is graphene Fermi energy and τ_e is graphene carrier lifetime.

In the case of a double layer graphene system (DLG), where the two graphene layers are separated by a distance d with a dielectric in between, the $V(q, \omega)$ and $\chi^{(0)}(q, \omega, T)$ terms take the form of a matrix. For a DLG system submerged in a homogeneous anisotropic uniaxial dielectric material ($\epsilon_x = \epsilon_y \neq \epsilon_z$, see figure A.3), for example h-BN, the intra- ($i = j$) and interlayer ($i \neq j$) Coulomb interactions in the graphene layers ($i, j = 1, 2$) are then given by

$$V_{ij}(q, \omega) = \frac{e^2}{2q\epsilon_s(\omega)} e^{-qd(1-\delta_{ij})\epsilon_a(\omega)} \quad (2.5)$$

where $\epsilon_s(\omega) = \sqrt{\epsilon_z(\omega)\epsilon_x(\omega)}$, $\epsilon_a(\omega) = \sqrt{\epsilon_z(\omega)/\epsilon_x(\omega)}$ and δ_{ij} is the Kronecker delta function, see figure A.5 for the plots of $\epsilon_s(\omega)$ and $\epsilon_a(\omega)$. The total dielectric screening function is then given by

$$\epsilon_{RPA}(q, \omega, T) = [1 - V_{11}(q, \omega) \chi_1^{(0)}(q, \omega, T)][1 - V_{22}(q, \omega) \chi_2^{(0)}(q, \omega, T)] - V_{12}(q, \omega)^2 \chi_1^{(0)}(q, \omega, T) \chi_2^{(0)}(q, \omega, T) \quad (2.6)$$

The bare Coulomb interactions will then be screened by charge carriers in the two graphene layers. These screened interactions can be described by the coupled Dyson equations and written in matrix form as

$$U(q, \omega, T) = V(q, \omega) + V(q, \omega) \cdot \chi^{(0)}(q, \omega, T) \cdot U(q, \omega, T) \quad (2.7)$$

where $U(q, \omega, T)$ is the 2x2 matrix of the screened interaction. The interlayer dressed interaction is then given by

$$U_{12}(q, \omega, T) = \frac{V_{12}(q, \omega)}{\epsilon_{RPA}(q, \omega, T)} \quad (2.8)$$

Similarly, the reducible density-density correlation function $X(q, \omega, T)$ can also be defined in terms of $\chi^{(0)}(q, \omega, T)$ and written in matrix form as

$$X(q, \omega, T) = V(q, \omega) + V(q, \omega) \cdot \chi^{(0)}(q, \omega, T) \cdot X(q, \omega, T) \quad (2.9)$$

The loss function, $L(q, \omega, T)$, which characterizes the spectral density of collective charge excitations, is defined as follows,

$$L(q, \omega, T) = \begin{cases} -\text{Im}[\frac{1}{\epsilon_{RPA}(q, \omega, T)}], & \text{for single layer graphene (SLG)} \\ -\text{Im}[\text{Tr}[X(q, \omega, T)]]], & \text{for double layer graphene (DLG)} \end{cases} \quad (2.10)$$

Figures 2.1 (a) and 2.1 (b) show the loss function, $L(q, \omega, T)$, calculated at $T = 0$ within the RPA for SLG and DLG, respectively. For simplicity, in DLG, the two identical (same carrier density) graphene layers separated by 6 nm have been considered placed in an homogeneous medium of dielectric constant 4.74 (i.e. the $\epsilon_s(\omega = 0)$ of h-BN). The $L(q, \omega, T)$, characterizes the collective modes (plasmons) of the graphene layer(s) as shown in figure 2.1 (a) for SLG. In the case of DLG, there are two such modes, one where the electron densities in the two layers oscillate in phase, called as the optical mode (O, $\omega \sim \sqrt{q}$), and one where the oscillations are out of phase, called as the acoustic mode (A, $\omega \sim q$). Since both modes lie within the Pauli-blocked interband region ($v_F q < \omega < v_F(2k_F - q)$), i.e., outside of the particle-hole continuum (PHC), they cannot decay via intraband excitations. However, both modes get damped as soon as they enter into the region $\omega > (v_F q, v_F(2k_F - q))$, as they decay via interband PHC transitions (Landau damping) that are now allowed by the energy-momentum conservation. It has to be also noted that the acoustic plasmon becomes degenerate with the PHC as $d \rightarrow 0$.

FIGURE 2.1: Density plot of the loss function, $L(q, \omega, T = 0)$, for (a) SLG and (b) DLG for $E_F = E_{F1} = E_{F2} = 0.2$ eV and $d = 6$ nm

2.1.2 Exchange-correlation (XC)

One limitation of the RPA is that it neglects the short range effects due to the exchange and correlation, which can modify the Coulomb interactions [102]. The exchange and correlation (XC) effects can be taken into account by employing local field factors (LFFs) as a correction to the inter- and intralayer Coulomb interactions.

The LFFs describe the repulsion hole around an electron due to the exchange repulsion and the correlation effects. The LFFs become more important for large wavevectors and for small carrier densities, and in the case of 2D electron bilayers they have been shown to have a significant impact on the plasmon dispersion and the drag resistivity [103]. The simplest form of the LFF is given by the Hubbard approximation, but Hubbard form is only for the intralayer LFF, while it is zero for the interlayer. Other approaches for obtaining both inter- and intralayer LFFs are the self-consistent Singwi-Tosi-Land-Sjölander (STLS) [104] and the Fermi hypernetted-chain (FHNC) [105] methods. However, both STLS and FHNC methods are cumbersome. Therefore, in this work, quantum Monte Carlo (QMC) simulation data for LFFs [102, 106] have been used instead, which are applicable for drag in SLG, which has a small value of the interaction parameter (r_s). However, under different experimental setups such as drag in SLG in a large magnetic field or drag in BLG, r_s can take large values, and hence, lead to strong correlations. Thus, in such cases one might have to go beyond the QMC method and use other forms of the LFFs and/or express r_s in terms of $\epsilon_{RPA}(q, \omega, T)$ instead of $\epsilon_s(\omega)$ to fit the data. The Coulomb interactions get modified by the XC effects, thus leading to the effective Coulomb interactions given by

$$V_{ij}^{eff}(q, \omega) = V_{ij}(q, \omega)(1 - G_{ij}(q, \omega)) \quad (2.11)$$

where $G_{ij}(= G_{ji})$ are the intra- ($i = j$) and interlayer ($i \neq j$) LFFs [102]. The LFFs are typically functions of q and of the interaction parameter r_s . The frequency dependence in the LFFs has been introduced by making r_s frequency dependent. This can be done by making use of a frequency-dependent κ term by substituting κ with $\epsilon_s(\omega) = \sqrt{\epsilon_x(\omega)\epsilon_z(\omega)}$, as shown in figure 2.2 (a). Static LFFs are plotted in figure 2.2 (b). Frequency dependent LFFs are plotted in appendix C. The effect of XC on

FIGURE 2.2: (a) Absolute value of the interaction parameter, r_s , as a function of the frequency. The vertical dashed lines are the phonon frequencies of h-BN. (b) Static intralayer, $G_{11}(q)$, and interlayer, $G_{12}(q)$, LFFs for $r_s = 0.46$.

the plasmon dispersion and primarily on Coulomb drag resistivity will be discussed in more detail in chapter 4.

2.1.3 Transfer matrix method (TMM) and SAW coupling efficiency

The transmittance and reflectance of the any layered system can be calculated from the Fresnel reflection and transmission coefficients by means of the transfer matrix (TMM) method. For TM polarization, the transfer matrix is given by $M = B_{01}P_1B_{12}P_2B_{23}....$, where B_{ij} is the transmission matrix for the i and j layers (here the

0th layer is considered as semi infinite), and P_j is the propagation matrix that connects the fields propagating over a distance of j th layer thickness. Graphene enters in the transmission matrix as a surface charge plane via the frequency-dependent conductivity $\sigma(\omega)$ (see appendix A.1 for details). These matrices are given by the expressions

$$B_{ij} = \begin{bmatrix} 1 + \frac{\sigma(\omega)}{Z_j} + \frac{Z_i}{Z_j} & 1 - \frac{\sigma(\omega)}{Z_j} - \frac{Z_i}{Z_j} \\ 1 + \frac{\sigma(\omega)}{Z_j} - \frac{Z_i}{Z_j} & 1 - \frac{\sigma(\omega)}{Z_j} + \frac{Z_i}{Z_j} \end{bmatrix}; P_j = \begin{bmatrix} \exp\{-iq_j^z d\} & 0 \\ 0 & \exp\{iq_j^z d\} \end{bmatrix} \quad (2.12)$$

where $Z_j = \omega\epsilon_j/q_j^z$ and $q_j^z = \sqrt{\epsilon_j(\omega/c)^2 - q^2}$, q^z and q are perpendicular and parallel component, respectively, of the wavevector of the light. The Fresnel reflection (r) and transmission (t) coefficients are given in terms of elements of the transfer matrix M as $r = M_{21}/M_{11}$ and $t = 1/M_{11}$. The absorption of the light is given by $\text{Im}[r]$, which is similar to the loss function given by equation 2.10.

In the presence of a SAW of wavelength λ_{SAW} and an amplitude δ , the transmittance of light is reduced as the SAW-induced diffraction grating scatters the incident light with wave vector $(q_{\parallel 0}, q_{z0})$ into the various diffraction orders $(q_{\parallel m}, q_{zm})$ with

$$q_{\parallel m} = (\omega/c) \sin \theta + m2\pi/\lambda_{SAW} \quad (2.13)$$

$$q_{zm} = \sqrt{(\omega/c)^2 - q_{\parallel m}^2} \quad (2.14)$$

where θ is the angle of off-normal incidence and m is an integer (see figure 2.3 for illustration). The intensity of the diffracted light in m th order, normalized to the intensity of the incoming beam, is given by [107]

$$I^m = \left| r J_m(2q_{z0}\delta) \left[1 + \frac{m\pi}{\lambda_{SAW}q_{zm}} \frac{1+r}{r} \right] \right|^2 \quad (2.15)$$

where J_m is the Bessel function for the m th diffraction order.

FIGURE 2.3: Schematic of surface displacement and light diffraction by SAW produced by an IDT.

2.2 Experiment

2.2.1 Graphene transfer

The graphene used in this study was CVD grown on Cu foil and obtained from the Graphene Supermarket. To transfer the graphene from Cu foil to the desired

substrate, an automatic graphene system was used [108], which minimizes the issues like wrinkling and folding of graphene film which are common problems during scooping of graphene from water. Here, first the graphene/Cu/graphene (note that the Cu foil has graphene grown on both sides) was spin coated with PMMA (around 100 nm thick) on one of the side followed by backside graphene etching in oxygen plasma and then placed in the Cu etchant (0.3 M ammonium persulfate) for 12-15 hours to ensure complete etching of Cu before rinsing with clean DI water several times leaving behind PMMA/graphene stack floating on top of water. The water is then removed and PMMA/graphene stack transferred to the desired substrate and then left for drying in a glove box chamber by first 1 hour in dynamic vacuum and then in static vacuum overnight. Once dried, PMMA is then removed by acetone (heated at 50°C) leaving behind PMMA free clean graphene/substrate. This method is used for transferring h-BN also.

2.2.2 Raman spectroscopy and mapping

Raman spectroscopy is a non-destructive tool that allows us to better understand the behavior of electrons and phonons in graphene. The Raman spectrum of graphene is characterized by a set of common features in the region between 800 and 2000 cm^{-1} , in particular the so-called D and G bands, which lie at around ~ 1350 and 1580 cm^{-1} , respectively. The G peak corresponds to doubly degenerate E_{2g} mode at the Brillouin zone center while the D peak corresponds to modes associated with transverse optical (iTO) phonons around the edge of the Brillouin zone (K or Dirac point). In a perfect defect-free graphene, momentum conservation would be violated by the double resonance mechanism, and thus the D peak is absent. Its overtone (2D, $\sim 2660 - 2710 \text{ cm}^{-1}$), however, is always visible even when the D peak is absent as momentum conservation is always satisfied without the need for defect activation since the process involves two phonons with opposite momentum vectors.

After transferring the graphene onto the target substrate, we performed Raman spectroscopy. A WITec Alpha 300AR Raman confocal microscope has been used for the Raman spectroscopy mapping of the channel area of the graphene field effect transistors (GFETs). Raman spectra were obtained in the backscattering geometry using a 50x objective lens (numerical aperture NA = 0.8) in ambient conditions. A

FIGURE 2.4: Raman spectra of bare LiNbO₃ substrate and graphene/LiNbO₃.

FIGURE 2.5: Raman maps of the intensity, the full-width-at-half-maxima (FWHM) and the peak position of the G and 2D peaks of graphene/LiNbO₃.

532 nm wavelength laser set to a power of 3 mW was used as excitation source. Figure 2.4 shows an example of graphene/LiNbO₃ comparing the bare LiNbO₃ substrate before transfer and after transferring graphene followed by oxygen plasma etching to create graphene mesa structure (c.r. figure 2.10 (a)). Prominent G and 2D peaks from graphene can be seen in addition to the 2LO phonon from the LiNbO₃ substrate. The peak intensity ratio between the 2D and G peaks is greater than 2 confirming the monolayer character of graphene. In order to check the homogeneity of the transferred graphene, we also performed a Raman mapping over an area. The maps of the intensity, the full-width-at-half-maximum (FWHM) and the position of the G and 2D peaks are shown in figure 2.5 indicating a homogeneous graphene quality. Figure 2.6 shows the correlation between the frequencies of the G and 2D peaks in one of these Raman mappings. The diagonal evolution of the data distribution shows a slope value of $\partial\nu_{2D}/\partial\nu_G \approx 2.2$, which is consistent with graphene under biaxial strain and constant p-type doping, since $(\partial\nu_{2D}/\partial\nu_G)_{\epsilon_{biaxial}} = 2.2$ [109, 110]

and $(\partial\nu_{2D}/\partial\nu_G)_p = 0.7$ [111]. Using the reported sensitivity values of $\partial\nu_G/\partial\varepsilon_{\text{biaxial}} = -69.1 \text{ cm}^{-1}/\%$ and $\partial\nu_{2D}/\partial p = -1.04 \text{ cm}^{-1}/10^{12} \text{ cm}^{-2}$ [111], these frequencies are decomposed in a native strain and residual doping-related basis [111], as shown by the internal set of axes in figure 2.6.

FIGURE 2.6: Correlation of the G- and 2D-peak frequencies of the graphene transferred to LiNbO₃. The mean and standard deviation of the distribution are shown by the symbol and the error bars, respectively.

2.2.3 Electrical characterization

Graphene was transferred to standard SiO₂/Si substrates to characterize its electrical properties (mobility and Fermi energy) by the transmission line method (TLM) as described in [112] using electrical characterization and a model fitting method [113]. In these substrates, the SiO₂ (300 nm thick) served as gate dielectric and the doped

FIGURE 2.7: Mobility and E_F of graphene measured by the TLM method.

Si was used as back gate. After transfer, graphene was patterned into a mesa structure by optical lithography and O_2 plasma etching, various field effect transistors (FETs) and transmission line devices were defined by optical lithography followed by metallization (Ti/Au) and lift-off. The devices were measured at a probe station and the results of the mobility and E_F as a function of the gate voltage are shown in figure 2.7. Negative and positive value of E_F indicate p- and n-type doping region respectively. The dip in mobility near zero gate voltage is due to presence of residual charges, which arises due to residual PMMA during transfer process.

2.2.4 Fourier transform infrared spectroscopy (FTIR)

Fourier transform infrared spectroscopy (FTIR) provides us with information about the absorption of infrared (IR) light in graphene. It also gives information about the phonons of the polar dielectric materials used either as interlayer (h-BN) or substrate. A Magna-IR 760 FTIR spectrometer is used for the measurements [114]. The operating principle of a spectrometer is based on the 2-beam interference. IR light emitted from a resistively heated SiC rod is collimated and split into two paths with KBr beam splitter. One of the beams reflects at a fixed mirror and another one in a moving mirror. These two beams are then made to interfere and guided to the sample compartment and finally to the DGTS detector. The spectrometer has a N_2 purging system to evacuate atmospheric gases from the sample compartment to avoid their absorption. Transmittance spectra were then obtained by placing the sample vertical in between source and detector. For reflectance, a reflectance accessory guide was mounted in the sample compartment. The accessory guides the IR beam by a set of mirrors to the sample with an adjusted incidence angle, from sample surface IR beam is reflected and detected by the detector. In our case, we set a 45 degree incidence angle for reflectance.

FIGURE 2.8: FTIR spectra of (a) graphene and (b) h-BN transferred on AlN/Si substrate.

In figure 2.8 (a), measured FTIR transmittance spectra of G/AlN/Si are shown and compared with bare AlN/Si and Si substrates. The decrease in the transmittance between 0.08 eV and 0.12 eV is due to the reststrahlen band of the AlN ($\omega_{AlN}^{TO} = 0.083$ eV, $\omega_{AlN}^{LO} = 0.11$ eV [115]), while the overall transmittance is decreased to approximately 50% by the free carrier absorption in Si. Figure 2.8 (b) shows the FTIR reflectance spectra of h-BN/AlN/Si and AlN/Si. In the h-BN/AlN/Si spectrum, two dips can be seen, which corresponds to the TO phonons of the anisotropic h-BN

($\omega_{h-BN}^{TO,x} = 0.097$ eV, $\omega_{h-BN}^{TO,z} = 0.17$ eV [27]). Here the slight difference in $\omega_{h-BN}^{TO,x}$ between the measured value and the value from ref. [27] is attributed to the difference in the quality of h-BN.

2.2.5 Fabrication and characterization of SAW devices

SAWs are most commonly generated by electrical means using the inverse piezoelectric effect [116]. The electrical generation applies for piezoelectric materials and uses interdigital transducers (IDTs) deposited on the surface to excite the acoustic mode by applying radio frequency (rf) signal to IDTs. The spatial period of the IDTs defines the SAW wavelength λ_{SAW} , whereas the opposite polarity finger overlapping length determines the width of the acoustic beam W . IDTs allow for the

FIGURE 2.9: Fabrication of a 2-port SAW resonator (SAWR).

generation of high-frequency acoustic modes using processes compatible with the conventional planar technology for opto-electronic devices. The fabrication steps of a 2-port SAW resonator (SAWR), formed by IDTs and metal reflection gratings, patterned on 128° rotated Y-cut X-propagating LiNbO₃ by optical lithography followed by metallization (Cr/Au) and lift-off is shown schematically in figure 2.9. The period and aperture of the IDTs were chosen to be $\lambda_{SAW} = 10$ μm and $W = 100$ μm , respectively. For the study of the effect of dynamic strain on graphene produced by SAW, as described thoroughly in chapter 6, graphene was then transferred onto the substrate with IDTs and then etched selectively using an oxygen plasma, leaving a graphene strip in the region between IDTs. The final device design schematic is shown in 2.10 (a) along with the measured S-parameter of the SAWR 2.10 (b).

S-parameters were measured by a 4-port Agilent N5230A PNA-L network analyzer and coplanar probe station (Cascade Summit 9000-series) by using GSG (400 μm pitch) probes from Cascade Microtech. S-parameters relate the voltage waves traveling into each port to the voltage wave traveling away from each port. S_{ij} is determined by driving port j with an incident wave of voltage V_j^+ and measuring the voltage wave amplitude V_i^- coming out of port i . S_{ii} is therefore the reflection coefficient looking into port i and S_{ij} is the transmission coefficient from port j to i .

FIGURE 2.10: (a) SAWR layout schematic. (b) S-parameters of the SAW device measured as a delay line.

For 2-port network they can be written in matrix form,

$$\begin{bmatrix} V_1^- \\ V_2^- \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} \\ S_{21} & S_{22} \end{bmatrix} \begin{bmatrix} V_1^+ \\ V_2^+ \end{bmatrix} \quad (2.16)$$

where + and - sign are for incident and reflected wave respectively. The S-parameter are then defined as

$$S_{11} = \left. \frac{V_1^-}{V_1^+} \right|_{V_2^+ = 0} \quad (2.17)$$

$$S_{12} = \left. \frac{V_2^-}{V_1^+} \right|_{V_1^- = 0} \quad (2.18)$$

$$S_{21} = \left. \frac{V_1^-}{V_2^+} \right|_{V_1^+ = 0} \quad (2.19)$$

$$S_{22} = \left. \frac{V_2^-}{V_2^+} \right|_{V_1^+ = 0} \quad (2.20)$$

All these S-parameters show dip (in case of S_{11} and S_{22}) and peak (in case of S_{12} and S_{21}) around the same frequency value in figure 2.10 (b), which is the ratio of velocity of SAW and λ_{SAW} . The same device as schematically shown in figure 2.10 (a) is used for interferometry and Raman spectroscopy also.

2.2.6 Acoustic field mapping by SAW interferometry

To map the SAW i.e., to measure the surface displacement profile of the SAW, we used an interferometer setup located at the Paul-Drude institute (Berlin, Germany), developed by Dr. Paulo Santos which in principle is a Michelson interferometer [117]. A schematic of the experimental setup for the SAW interferometry is shown in figure 2.11. Here, a laser ($\lambda_{SAW} = 532$ nm) is passed through a chopper ($\omega_c = 317$ Hz) to a mirror, from where it is directed to a beam splitter. One beam goes to a

piezoelectric mirror (mirror displacement $\delta_m = 20$ nm) whereas the other one is focused on a small spot ($< 1\mu\text{ m}$) on the substrate by a microscope objective (50x).

FIGURE 2.11: Experimental setup for SAW Interferometry.

Both reflected laser beams are then made to interfere. This interference will not be static but dynamic due to the presence of SAW and will oscillate with a frequency equal to the SAW frequency (ω_{SAW}) and an amplitude proportional to the SAW amplitude (δ). The interference patterns consist of concentric circles as shown in figure 2.12. The output of the interferometer is detected by a fast photodiode. The amplitude of the diode signal, measured using a spectrum analyzer, is proportional to the amplitude of the z (out of plane) component of the particle displacement, u_z .

2.2.7 Acoustically-modulated Raman spectroscopy

For the study of the strain on graphene we made use of Raman spectroscopy which was performed in the backscattering configuration at room temperature using a confocal Horiba LabRam system located at the Paul-Drude institute (Berlin, Germany). The incident $\lambda_{SAW} = 473$ nm solid-state laser beam was focused onto a spot of approximately $1\mu\text{ m}$ diameter and 0.8 mW power on the sample surface using a long working distance microscope objective (50x) with a numerical aperture of 0.55. The backscattered light was collected by the same objective, spectrally analyzed by a monochromator with a 1800 grooves/mm dispersion grating and detected using a liquid-nitrogen-cooled charge coupled device (CCD).

In order to detect the changes in the Raman spectra induced by the SAW, the amplitude-modulation method described in ref. [118] was used. Here, both the rf signal generating the SAW at the IDTs and the excitation laser are chopped with the

FIGURE 2.12: SAW Interference fringes.

FIGURE 2.13: Schematic of the amplitude modulation of SAW

same modulation frequency (300 Hz) to be either in- or out-of-phase (see figure 2.13 for illustration).

Spectra were then recorded with the two excitations in phase (i.e. with the sample subjected to both the laser and acoustic excitations; called hereafter *SAW ON*) and out-of-phase (where the sample is exposed only to the laser; called hereafter *SAW OFF*) following the alternated sequence *SAW OFF* - *SAW ON* - *SAW ON* - *SAW OFF* (see figure 2.14).

FIGURE 2.14: Schematic of the laser and SAW in- and out-of-phase sequence

An acquisition time of 60 s per spectrum was used for each excitation condition and 20 sequences were measured (80 spectra in total). By averaging, respectively, over the 40 *SAW ON* and 40 *SAW OFF* cycles, the Raman spectra with (I_{on}) and without (I_{off}) the effects induced by the SAW were obtained. This method, besides increasing the signal-to-noise ratio due to the integration, minimizes the effects of the systematic fluctuation of the temperature and of the laser power on the Raman signal, allowing us to isolate the strain contribution [118].

The applied rf power from the signal generator (P_{SG}) was first amplitude modulated (AM) at a frequency of 300 Hz. Due to this modulation, the peak to peak instantaneous voltage, V_{pp} , becomes zero in one half of the AM cycle and gets doubled in the other half (see figure 2.14), leading to a 4-fold increase (+6 dB) in the instantaneous rf power (since instantaneous power, $P \propto (V_{pp}/\sqrt{2})^2$). Then the rf signal was then split into two equal (power) signals (-3 dB) and each of them was fed to an amplifier with +23 dB gain. Finally, the two equal signals from the output of each amplifier (P_{rf}) were applied simultaneously to the two IDTs forming the 2-port SAWR to create a standing wave (see figure 2.15 for illustration).

FIGURE 2.15: Schematic of the rf power scheme feeding the 2-port SAWR

Chapter 3

SAW-mediated plasmon-phonons in graphene/h-BN

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3.1 Graphene/h-BN/(AlN,ZnO)

Figures 3.1 plots the surface plasmon-phonon polariton (SPPP) dispersion calculated by the transfer matrix method (TMM) (see section 2.1.3) for the graphene (G) on AlN and ZnO compared with the case when a thin h-BN layer ($d = 1$ nm) is inserted between G and the AlN(ZnO). AlN and ZnO are piezoelectric materials [119, 120] supporting strong SAWs and are chosen in this study because they can be grown directly on substrates like Si and GaAs that can act as back gate, or allocate, alternatively, a buried metal gate, that can also be used for modulating the graphene Fermi energy. The graphene mobility has been considered to be enhanced by the h-BN film from $\mu = 5000$ to $10000 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$, whereas E_F has been taken to be 0.4 eV. The chosen mobility value for bare graphene is higher than the measured value shown in figure 2.7, as the graphene sample used there for measuring mobility was strongly polycrystalline with small grain size and unintentionally doped with PMMA residues during the wet transfer process. However, large mobility values as those selected

FIGURE 3.1: $SPPPi$ dispersion for (a) G/AlN and (e) G/ZnO compared with (c) G/h-BN/AlN and (g) G/h-BN/ZnO with a h-BN film thickness $d = 1$ nm. (b), (d), (f) and (h) Lifetime of the $SPPPi$. Dashed horizontal lines correspond to the phonon frequencies of AlN(ZnO) and h-BN, whereas dashed vertical lines indicates SAW wavelength of 0.25 and 1 μm .

FIGURE 3.2: Confinement ratio $\lambda_{\text{air}}/\lambda_{SPPPi}$ for the different $SPPPi$ supported by the G/h-BN ($d = 1$ nm)/AlN system.

here for the calculations can be expected from a high quality graphene sample with large grain size and dry transferred thus avoiding PMMA residues. The dispersion of the G/h-BN/AlN(ZnO) system presents a larger number of $SPPPi$ modes as compared to its G/AlN(ZnO) counterpart as a result of the interaction between the carriers in graphene and the phonons in h-BN. In particular, $SPPPi_1$ and $SPPPi_3$ arise from the interaction of the graphene surface plasmon with the surface optical (SO) phonon in AlN (ZnO), as shown in figures 3.1 (a), (c), (e) and (g), whereas the additional $SPPPi_2$ and $SPPPi_4$ in figures 3.1 (c) and (g), arise from the interaction with

the phonons of h-BN with two polarizations. Figures 3.1 (b), (d), (f) and (h) depict the lifetime of the corresponding $SPPP_i$ in figures 3.1 (a), (c), (e) and (g). Each $SPPP_i$ branch presents different spectral weight and lifetime, which depend on the oscillator strength and phonon lifetime of the underlying materials. The lifetime τ_p of the $SPPP_i$ spans between the limits imposed by the graphene carrier relaxation time, τ_e , and the phonon lifetime, τ_{SO} . Since the h-BN film enhances the graphene carrier mobility, and hence τ_e , the τ_p of $SPPP_1$ and $SPPP_3$ is increased. On the other hand, h-BN supports longer-lived phonons than AlN, providing $SPPP_2$ and $SPPP_4$ with the largest τ_p values (exceeding 1 ps for $SPPP_2$) (see appendix A for details of the material parameters). Thus, the h-BN interlayer allows both to increase the lifetime of the SPPs and to broaden the energy range that they cover. The case of $SPPP_2$ is especially interesting. Since the lower reststrahlen band (energy band bounded by TO and LO phonon of the respective material in which the material has 100 % reflection) of h-BN lies within the reststrahlen band of AlN, the $SPPP_2$ branch in this system is weak in intensity though its proximity to the SO phonon of the h-BN provides it with the largest lifetime. However, the intensity of $SPPP_2$ is enhanced in the case of ZnO as the reststrahlen band of ZnO does not overlap with h-BN. The confinement ratio, $\lambda_{air}/\lambda_{SPPP}$, corresponding to each $SPPP_i$ branch of figure 3.1 (c) is shown as a function of the incident light wavelength λ_{air} in figure 3.2.

3.2 Effect of h-BN thickness on plasmon dispersion

FIGURE 3.3: $SPPP_i$ and $HPPP_i^{U,L}$ dispersion for the G/h-BN/AlN system with a h-BN interlayer thickness d of (a) 5 nm, (b) 10 nm, (c) 50 nm, and (d) 70 nm. (e) and (f) are magnified views of the part of figures (c) and (d), respectively, around the AlN reststrahlen band. The superscripts in $HPPP_i^{U,L}$ denote the upper and lower reststrahlen bands.

The dispersion of the G/h-BN/AlN(ZnO) system strongly depends on the thickness, d , of the h-BN interlayer, as shown in figure 3.3. Here we consider AlN as substrate, but the results with ZnO will be qualitatively identical. As d increases, $SPPP_1$ and $SPPP_4$ shift towards higher energies, whereas $SPPP_3$ shifts towards lower energies and $SPPP_2$ does not vary substantially, as shown in figures 3.1 (c) and 3.3 (a) and 3.3 (b) for $d = 1, 5$, and 10 nm, respectively. For $d = 50$ and 70 nm, see figures 3.3 (c) and 3.3 (d) respectively, hyperbolic plasmon phonon polaritons (HPPPs) appear within the h-BN reststrahlen bands. These HPPPs appear also in the dispersion of the thinner h-BN films considered here but at higher q values than those depicted in figures 3.1 (b) and 3.3 (a) and 3.3 (b). In general, the thicker the h-BN interlayer, the stronger its influence on the dispersion curve of the G/h-BN/AlN system, with the $HPPP_i$ appearing at lower q values, and the weaker the influence of the AlN substrate. Also, increasing the thickness of the h-BN interlayer from 50 to 70 nm, a transition from $SPPP_3$ to $SPPP_2$ is observed, as shown in figures 3.3 (e) and 3.3 (f). This crossover is also attributed to the fact that increasing the h-BN thickness the influence of the AlN substrate weakens while that of the h-BN interlayer strengthens. In summary, by increasing the h-BN thickness the dispersion of the three-layer system (G/h-BN/AlN) seems to move gradually towards an effective two-layer system (G/h-BN).

3.3 Coupling of light and plasmons by SAW

FIGURE 3.4: ΔT^{TM} spectra of (a) $SPPP_1$, (b) $SPPP_3$, and (c) $SPPP_4$ for the G/AlN and G/h-BN($d = 1$ nm)/AlN systems for variable carrier mobility μ (in units of $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$).

A SAW acts as dynamic virtual grating for the incident light (or photons) that can provide the necessary extra momentum needed for the coupling of light and plasmons. This coupling can be probed by transmission or reflection spectroscopy, where for example, in case of transmission, the coupling of light and plasmons will result in a reduction of the transmission due to the enhanced absorption of light. Figure 3.4 plots the change in the transmittance, $\Delta T^{TM} = \Delta T^{TM}(0) - \Delta T^{TM}(\delta)$, in the G/AlN and G/h-BN($d = 1$ nm)/AlN systems for the first diffraction order ($m = 1$) induced by a SAW with an amplitude $\delta = 4$ nm and a wavelength $\lambda_{SAW} = 0.25 \mu\text{m}$ due to the generation of SPPPs (see section 2.1.3). This value of δ is unrealistically high for this small λ_{SAW} , and a lower value of δ will lead to a decrease in the coupling efficiency as the intensity of the first order diffracted light is proportional to δ^2 (see eq. 2.15). However, as can be seen from figure 3.3, larger λ_{SAW} can probe all $SPPP_i$ along with high values of δ . A more realistic choice of δ and λ_{SAW} will thus yield a coupling efficiency of 10^{-4} %, similar to that reported for SAW-driven plasmons in metal films [41]. As described in the previous section, the SPPP dispersion of the G/AlN system is modified by the introduction of a thin h-BN interlayer. This is also

reflected into the ΔT^{TM} spectra, as shown in figures 3.4 (a) and 3.4 (b) for the $SPPP_1$ and $SPPP_3$, respectively. In addition to a frequency shift of the peaks, the enhancement of the mobility induced by the h-BN leads to an increase in the peak intensity and to a reduced full-width-at-half-maximum of the peaks. This indicates that the h-BN strengthens $SPPP_1$ and $SPPP_3$ and allows them to live longer and propagate longer distances. Furthermore, new SPPs appear in the ΔT^{TM} spectra, as shown for example in figure 3.4 (c) for the $SPPP_4$. However, the increase in the mobility induced by the h-BN only enhances slightly the $SPPP_4$. This different behaviour, as compared to $SPPP_1$ and $SPPP_3$, can be explained considering that, as mentioned in the previous section, the lifetime τ_p of the $SPPP_i$ spans in between the limits imposed by τ_e and τ_{SO} . Thus, the closer the $SPPP_i$ is to a phonon in the dispersion curve, the stronger its phonon-like character and hence the stronger the influence of τ_{SO} rather than of τ_e . Hence, $SPPP_4$, that runs close to $\omega_{hBN}^{TO,z}$ within the studied q range, suffers a little influence of any enhancement in τ_e produced by an increment in the mobility.

FIGURE 3.5: ΔT^{TM} spectra of (a) $SPPP_1$, (b) $SPPP_3$ (and $SPPP_2$), and (c) $SPPP_4$ for the G/AlN and G/h-BN/AlN systems for various h-BN film thickness. For $d = 70$ nm, $SPPP_2$ mode appears instead of $SPPP_3$. The insets in (b) and (c) show magnified views of the energy range where $HPPP_1^L$ and $HPPP_1^U$ appear, respectively.

The ΔT^{TM} spectra are also strongly modulated by the thickness of the h-BN interlayer. Figure 3.5 shows the ΔT^{TM} spectra associated to the different SPPP and HPPP modes appearing in the dispersion curves of figure 3.2 for a q value corresponding to $\lambda_{SAW} = 0.25 \mu\text{m}$. It can be seen that, increasing the h-BN thickness from 1 to 70 nm, $SPPP_1$ and $SPPP_4$ present stronger peaks that blue shift as shown, respectively, in figure 3.5 (a) and 3.5 (c). The opposite trend is observed though for $SPPP_3$, figure 3.5 (b), where the peak intensity decreases and the peak position red shifts when increasing the h-BN thickness from 1 to 50 nm. This latter behaviour is due to the fact that $SPPP_3$ is strongly related to the SO phonon in AlN, and the electron-phonon coupling strength reduces as $\exp\{-2qd\}$. For a h-BN thickness of 70 nm, the $SPPP_2$ appears instead of $SPPP_3$, as shown in figures 3.3 (d) and 3.3 (e).

In addition to the tunability of the $SPPP_i$ and $HPPP_i$ dispersion in the G/h-BN/AlN system provided by the thickness of the h-BN interlayer, graphene presents an intrinsic degree of tunability by means of the modulation of its carrier density n , or $E_F = \hbar v_F \sqrt{\pi n}$, through electrostatic gating. All $SPPP_i$ and $HPPP_i$ blue shift as E_F increases, as expected from the hybridization with a higher energy graphene plasmon. However, the intensity of a particular $SPPP_i$ and $HPPP_i$ is especially enhanced at certain h-BN film thickness and E_F value combinations. Therefore, a detailed study of the dispersion of the G/h-BN/AlN system, both in terms of h-BN thickness as well as graphene doping, is required for the design of the most efficient SAW-mediated plasmonic devices.

Figure 3.6 shows the evolution of the maximum value of the peak in the ΔT^{TM} spectra of the different $SPPP_i$ and $HPPP_i$ induced by a SAW with $\lambda_{SAW} = 250$ nm. For thin h-BN films, the maximum of $SPPP_1$, figure 3.6 (a), occurs at energies below ω_{AIN}^{TO} as the $SPPP_1$ cannot propagate into the reststrahlen band of AlN. However, for thicker h-BN layers, this boundary condition relaxes and the maximum occurs at energies beyond ω_{AIN}^{TO} but below $\omega_{h-BN}^{TO,x}$, as now is the lower reststrahlen band of h-BN the one effectively limiting the propagation of $SPPP_1$. In all cases, the appearance of a maximum as E_F increases is related to the existence of two competing mechanisms, since a larger carrier density makes the plasmon stronger while its shift to higher energies is stopped by either one of the reststrahlen bands. The behaviour of $SPPP_2$ and $SPPP_3$, figure 3.6 (b), is more complex as it is affected by both the AlN reststrahlen band and the lower reststrahlen band of h-BN. As shown in figure 3.1 (c) for a 1-nm thick h-BN interlayer, $SPPP_2$ appears very close to $\omega_{h-BN}^{LO,x}$ and is very weak. In order to benefit from the large lifetime of this phonon, it is required to increase the h-BN interlayer thickness above 10 nm, as shown in figure 3.6 (b) and its magnified view in figure 3.6 (d), while keeping E_F low. In this regime, $SPPP_2$ evolves from $\omega_{h-BN}^{LO,x}$ onwards as E_F increases. However, for the q value imposed by the selected SAW wavelength, there is a transition from $SPPP_2$ to $SPPP_3$ at a certain threshold value of E_F . The threshold value that produces this crossover increases with the h-BN thickness, as depicted in figure 3.6 (d). The intensity of $SPPP_3$ increases monotonically in the E_F range studied as its upper limit, $\omega_{h-BN}^{TO,z}$ is far. However, increasing the h-BN thickness reduces the energy range covered by

FIGURE 3.6: Evolution of the maximum value ΔT^{TM} of (a) $SPPP_1$, (b) $SPPP_2$ and $SPPP_3$, and (c) $SPPP_4$ with E_F for graphene/h-BN/AlN systems with carrier mobility $\mu = 10000$ $\text{cm}^2\text{V}^{-1}\text{s}^{-1}$ and various h-BN film thickness d . (d) zoom-in view of the box in (b). The solid circles represent the $SPPP_3$ mode whereas hollow circles correspond to the $SPPP_2$ mode. Insets in (b) and (c) show magnified views of the modulation of the $HPPP_i^L$ and $HPPP_i^U$, respectively, that appear for the 50 and 70 nm thick h-BN interlayers. E_F varies in all cases from 0.1 to 0.6 eV in steps of 0.02 eV in the direction of the arrow.

$SPPP_3$ as the influence of AlN in the G/h-BN/AlN heterostructure reduces. The inset of figure 3.6 (b) shows the evolution of $HPPP_1^L$ and $HPPP_2^L$, which are only observed for the 50 and 70 nm-thick h-BN interlayers for the q value considered. These $HPPP_i^L$ modes are expected to be confined within the lower reststrahlen band of h-BN. However, it can be seen that $HPPP_1^L$ extends slightly below $\omega_{h-BN}^{TO,x}$. This seems to be related to the fact that the lower reststrahlen band of h-BN lies within the reststrahlen band of AlN, so that the lower limit imposed by $\omega_{h-BN}^{TO,x}$ relaxes. The exact mechanism behind this is still unclear. In case of $SPPP_4$, figure 3.6 (c), its strength increases monotonically with increasing E_F for all h-BN thickness values, as no boundary condition limits it. Conversely, $HPPP_1^U$ and $HPPP_2^U$, which are only observed for the 50 and 70 nm-thick h-BN interlayers for the q value considered (inset of figure 3.6 (c)), appear confined within the upper h-BN reststrahlen band. These $HPPP_i^U$ modes are thus expected to peak or saturate when evolving further towards $\omega_{h-BN}^{LO,z}$ if a broader parameter range is considered.

3.4 Graphene/h-BN/Graphene/ZnO

In this section we study the DLG system separated by h-BN on a ZnO substrate. This material choice avoids the reststrahlen band overlap between AlN and h-BN as described in section 3.1. However, apart from this detail, a DLG system on AlN will be qualitatively similar to that on ZnO. Figure 3.7 (a) and (b), plot the dispersion curve of G/h-BN/G/ZnO for a h-BN thickness of 1 and 50 nm, respectively. DLG leads to the emergence of an extra acoustic branch, $SPPP_{1,A}$, in addition to the optical branch, $SPPP_{1,O}$, and with ZnO as a substrate $SPPP_2$ and $SPPP_3$ branches can be distinctively observed for thick h-BN. Comparing with the SLG system with a 1 nm h-BN in figure 3.1 (g), it can be shown that $SPPP_i$ in DLG covers a wider energy range as shown in figure 3.7 (a).

FIGURE 3.7: $SPPP_i$ and $HPPP_i$ dispersion for the G/hBN/G/ZnO system for a hBN thickness of (a) 1 nm and (b) 50 nm.

Another advantage of using DLG over SLG is the enhancement in the intensity of the higher energy $SPPP_i$ branches, $SPPP_2$ and $SPPP_4$, especially $SPPP_4$ which has diminishing intensity in case of SLG. This is shown in figure 3.8 plotted for SLG

and DLG where all graphene layers were considered to be identical with same $E_F = 0.4$ eV and separated by 1 nm thick h-BN for $\lambda_{SAW} = 0.25 \mu\text{m}$.

Similarly to SLG, the $SPPP_2$ and $SPPP_4$ branches in DLG also can be tuned upon changing the values of E_F and the interlayer h-BN thickness. The evolution of the maximum value ΔT^{TM} of $SPPP_2$ and $SPPP_4$ for different h-BN thicknesses and varying E_F from 0.01 to 0.4 eV is plotted in figure 3.9 comparing both SLG and DLG for $\lambda_{SAW} = 250$ nm. For a fixed E_F , by increasing the h-BN thickness the maximum intensity of $SPPP_4$ increases and moves toward higher energy while the trend is opposite for $SPPP_2$ for both SLG and DLG. And for a fixed h-BN thickness, by varying E_F both $SPPP_2$ and $SPPP_4$ branches shift to higher energy with an increase in the maximum intensity value. However, DLG provides larger intensity and broader energy range coverage as compared to SLG for any value of h-BN thickness and E_F .

FIGURE 3.8: ΔT^{TM} spectra of $SPPP_2$ and $SPPP_4$ comparing both SLG and DLG systems on ZnO substrate with a h-BN interlayer thickness of 1 nm. Inset shows a magnified view of the ΔT^{TM} spectra of $SPPP_1$.

FIGURE 3.9: Evolution of the maximum value ΔT^{TM} of $SPPP_2$ and $SPPP_4$ with E_F comparing SLG and DLG system on ZnO with various h-BN film thickness values.

Chapter 4

Coulomb drag in graphene/h-BN/graphene

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4.1 Drag resistivity and intensity

Coulomb drag is characterized by the drag resistivity, ρ_D . The experimental setup consists of two charge carrying layers (e.g., graphene), placed closed to each other and electrically isolated by an insulator (e.g., h-BN) in between them (see the cover image of this chapter above). When a voltage (current) is applied to one of the layers, called as the active layer, the exchange of momentum between the two layers will induce a current (voltage) in the other layer, called as the passive layer. The momentum exchange between the layers is due to electron-electron interactions which can be mediated by phonons of the dielectric interlayer and plasmon modes of the conducting top and bottom layers depicted, respectively, as wiggly yellow lines and concentric red circles in the cover image. The fluctuations of the electron density of the active layer lead to a fluctuating electric potential which is sensed by the passive layer. Due to the applied voltage in the active layer, these fluctuations in the electric potential, $\phi(q, \omega)$, are not fully isotropic in the wavevector q and thus induce a DC current, J_{DC} , in the passive layer in a nonlinear response. For the double layer graphene system (DLG), the drag resistivity, ρ_D , is a 2x2 matrix given by

$$\rho_D = \frac{\sigma_{12}}{\sigma_{11}\sigma_{22} - \sigma_{12}\sigma_{21}} \simeq \frac{\sigma_{12}}{\sigma_{11}\sigma_{22}} \quad (4.1)$$

In this equation, $\sigma_{11}(\sigma_{22})$ is the temperature-dependent intralayer conductivity of the top (bottom) layer, given by the expression $e^2\mu_D\tau/(\pi\hbar^2)$, where τ is the graphene carrier transport time, which is assumed to be energy independent, and μ_D is the effective Drude weight, which depends on the carrier density and the temperature (see appendix A.1 for more details on the graphene conductivity, chemical potential, and Drude weight). And $\sigma_D \equiv \sigma_{12} = \sigma_{21}$ is the interlayer drag conductivity, which, up to second order perturbation in the interlayer interaction, is given by

$$\sigma_D = \frac{e^2}{64\pi^3k_BT} \int d^2q \int_0^\infty d\omega \frac{|(U_{12}(q, \omega, T)|^2 \Gamma_1(q, \omega, T) \Gamma_2(q, \omega, T))}{\sinh^2(\hbar\omega/2k_BT)} \quad (4.2)$$

where ω and q are the transferred energy and momentum from active to passive layer at temperature T with $U_{12}(q, \omega)$ being the dressed interlayer interaction given by eq. 2.8 and $\Gamma_l(q, \omega, T)$ being the rectification function or non-linear susceptibility of layer l [59]. $\Gamma_l(q, \omega, T)$ relates the external space and time dependent scalar potential, $\phi(q, \omega)$, in the active layer to the induced DC current, J_{DC} , in the passive layer in quadratic order (nonlinear response),

$$J_{DC} = \Gamma(q, \omega, T) |\phi(q, \omega)|^2. \quad (4.3)$$

$\Gamma_l(q, \omega, T)$ can be thought of as a measure of the particle-hole asymmetry. To induce a voltage (or generate a current) in the passive layer, one needs to move the charge carriers in the active layer. A current in the active layer leads to quasiparticles excitations (electrons and holes) around the Fermi energy of the electron gas, where they move in opposite directions. In the presence of particle-hole asymmetry such a state has nonzero total momentum which is transferred equally to electrons and holes in the passive layer, where the electrons and holes there are pushed in the same direction. This process can induce a voltage only due to particle-hole asymmetry. In conventional semiconductors the particle-hole asymmetry arises due to the curvature of the conduction band. In the Fermi-liquid theory the particle-hole asymmetry can be expressed as a derivative of the single-layer conductivity with respect to the chemical potential. Graphene exhibits exact particle-hole symmetry at the Dirac point at which the nonlinear susceptibility vanishes implying the absence of the drag effect. In contrast, experiment shows nonzero drag resistivity at Dirac point [52] which was attributed to charge density fluctuations near Dirac point [121], energy drag mechanism [122], and third-order drag effect [63]. The final expression for the drag resistivity can be written as

$$\rho_D = \int dq \int_0^\infty d\omega I(q, \omega, T), \quad (4.4)$$

where the integrand $I(q, \omega, T)$ is the drag intensity (also inferred as scattering rate) given by

$$I(q, \omega, T) = \frac{2\pi e^2}{64\pi^3k_BT} \frac{|(U_{12}(q, \omega, T)|^2 \Gamma_1(q, \omega, T) \Gamma_2(q, \omega, T))}{\sinh^2(\hbar\omega/2k_BT)} \left(\frac{\pi\hbar^2}{e^2\mu_D\tau} \right)^2 \quad (4.5)$$

4.2 Effect of exchange and correlation

Figures 4.1 (a) and 4.1 (b) show the loss function, $L(q, \omega, T)$ as defined in eq. 2.10, calculated for DLG at $T = 0$ within the RPA only (using the Coulomb interaction given

by eq. 2.5) and with the inclusion of the exchange and correlation (XC) effects (using the effective Coulomb interaction given by eq. 2.11), respectively. For simplicity, two identical ($E_{F1} = E_{F2} = 0.064$ eV) graphene layers separated by a distance $d = 6$ nm have been considered to be placed in a homogeneous medium of dielectric constant 4.74 (i.e. the $\epsilon_s(\omega = 0)$ of h-BN). The maximum value of $L(q, \omega, 0)$ in the XC case is almost double that in the RPA case as seen in colour scale bars of figures 4.1 (a) and 4.1 (b).

Also, after taking into account XC, the two plasmon branches shift to higher q values for a fixed ω and to lower ω values for a fixed q , as shown, respectively, in figures 4.1 (c) and 4.1 (d). In addition, the inclusion of the XC reduces the interlayer Coulomb interactions compared to those provided by the RPA alone, as shown in figures 4.1 (e) and 4.1 (f) for a fixed value of both ω and q , respectively. Thus, the XC increases the $\epsilon_{RPA}(q, \omega, T)$ and decreases the $V_{12}^{eff}(q, \omega)$, which in turn decreases the dressed interlayer Coulomb interaction, $U_{12}(q, \omega, T)$, see figure 4.2. As a result, the drag resistivity, ρ_D , which is directly proportional to $|U_{12}(q, \omega, T)|^2$ decreases with

FIGURE 4.1: Density plot of the loss function $L(q, \omega, 0)$ given by eq. 2.10 calculated for the case of (a) RPA and (d) XC for the DLG system with background $\epsilon = 4.74$. The solid lines separate the regions with Landau damping due to intraband ($\omega < v_F q$) and interband ($\omega > v_F q, v_F(2k_F - q)$) particle-hole continuum and the Pauli-blocked interband region ($v_F q < \omega < v_F(2k_F - q)$). Cross section of $L(q, \omega, 0)$ for (c) $\omega/E_F = 0.9$ and (d) $q/k_F = 0.65$, showing the shift of the plasmon branches with the inclusion of the XC effects. The latter also lead to a decrease in the magnitude of the effective interlayer Coulomb interaction $V_{12}^{eff}(q, \omega)$ as shown in (e) and (f) for $\omega/E_F = 0.9$ and $q/k_F = 0.65$, respectively.

FIGURE 4.2: $|U_{12}(\omega)|$ for (a) $q/k_F = 1$, (b) 2, (c) 3, and (d) 4 calculated both within RPA (dashed curves) and XC (solid curves). Parameters are: $T = 300$ K, $d = 6$ nm, and $E_F = 0.064$ eV.

FIGURE 4.3: Drag resistivity, ρ_D , as function of the temperature calculated for the RPA and XC cases.

the inclusion of XC as shown in figure 4.3.

FIGURE 4.4: Density plot of the $L(q, \omega, T)$, at (a) $T = 50$ K and (d) $T = 300$ K for the h-BN/G/h-BN/G/h-BN system. The superscript in HPP denotes the upper (U) and lower (L) reststrahlen bands. Density plot of the drag intensity, $I(q, \omega, T)$, at (b) $T = 50$ K without phonons, (c) $T = 50$ K with phonons, (e) $T = 300$ K without phonons, and (f) $T = 300$ K with phonons. The scale bars in (a) and (b) also apply, respectively, to (d) and (c), (e) and (f). The horizontal dashed lines are the phonon frequencies of h-BN. The hybridized surface and hyperbolic plasmon-phonon modes are labeled as SPP and HPP, respectively, where the superscript in HPP denotes the upper and lower reststrahlen bands.

4.3 Plasmons and phonons in Coulomb drag

Figure 4.4 (a) shows the loss function, $L(q, \omega, T)$, whereas figures 4.4 (b) and (c) show the Coulomb drag intensity, $I(q, \omega, T)$, without and with phonons, respectively, all calculated for the h-BN/G/h-BN/G/h-BN system for a temperature of 50 K. Figures 4.4 (d-e) plot the same magnitudes, respectively, as figures 4.4 (a-c) but for a temperature of 300 K. For simplicity, both graphene layers are considered to have identical carrier density ($E_{F1} = E_{F2} = 0.064$ eV) and the h-BN interlayer thickness is $d = 6$ nm. The hybridization of h-BN phonons with the plasmons in graphene can be eliminated by setting $\epsilon_s(\omega = 0)$ and $\epsilon_a(\omega = 0)$ in the expressions of $V_{11(22)}^{eff}(q, \omega)$ and $V_{12(21)}^{eff}(q, \omega)$ only, see figure 4.5. In the $L(q, \omega, T)$ plot, both plasmon modes, optical and acoustic, hybridize with the phonons of h-BN which start to get damped at higher temperature value (300 K, figure 4.4 (d)). The reason for this is the behaviour of the imaginary part of the graphene temperature-dependent polarizability $\text{Im}[\chi_l^{(0)}(q, \omega, T)]$, shown in figure 4.6, that represents the PHC. In this figure, the hybridized SPP modes are indicated by the superimposed black lines. At low temperature, the plasmon modes, which lie in the region $v_F q < \omega < v_F(2k_F - q)$, are undamped as they do not overlap with the PHC, as shown in figure 4.4 (a) for $T = 50$ K. Nevertheless, at high temperature, these plasmon modes overlap with the PHC, as shown in figure 4.4 (b) for $T = 300$ K, and now they can decay via particle hole excitations. The drag intensity, $I(q, \omega, T)$, which is directly proportional to the drag resistivity ρ_D , allows us to separate the contribution of the different quasiparticle excitations involved in the Coulomb drag. Figures 4.4 (b) and 4.4 (c) indicate that,

FIGURE 4.5: Density plot of the $L(q, \omega, T = 0)$, for $E_F = E_{F1} = E_{F2} = 0.2$ eV and $d = 6$ nm for (a) $\epsilon_a(\omega)$ and $\epsilon_s(\omega)$, (b) $\epsilon_a(\omega = 0)$ and $\epsilon_s(\omega)$, and (c) $\epsilon_a(\omega = 0)$ and $\epsilon_s(\omega = 0)$.

FIGURE 4.6: Density plot of the imaginary part of the graphene polarizability, $\text{Im}[\chi_l^{(0)}(q, \omega, T)]$, for a temperature (a) $T = 50$ K and (b) $T = 300$ K with $E_F = 0.064$ eV.

at low temperature (50 K), the major contribution to the drag resistivity comes from the intraband region, $\omega < v_F q$, and it is identical for both cases of with and without phonons because the temperature T in the $\sinh$ term in the denominator of eq. 4.2 sets an effective upper cut-off in ω . Thus, at low T , the HPP modes, appearing at higher frequencies (the two reststrahlen bands lie at around 100 and 180 meV), do not contribute to the Coulomb drag. On the other hand, the drag intensity is proportional to the non-linear susceptibility (see eq. 4.5), which vanishes at low temperature in the region where the SPP modes lie, $v_F q < \omega < v_F(2k_F - q)$, as shown in figure 4.7 (a). In this figure, the hybridized SPP modes are indicated by the superimposed black lines. Thus, at low temperature, these modes do not contribute either to the Coulomb drag.

$I(q, \omega, T)$ changes at high temperature, as shown, for example, in figures 4.4 (e) and 4.4 (f) for $T = 300$ K. In the case when phonons of h-BN are not considered, figure 4.2 (e), the drag intensity is enhanced due to the contribution from the optical and acoustic plasmon modes in the interband region. When the effect of the h-BN phonons is taken into account, as shown in figure 4.4 (f), the enhancement of the drag intensity is caused by the contribution of both hybridized SPP and HPP modes. These contributions mimic the dispersion shown by the $L(q, \omega, T)$ in figure 4.4 (d).

FIGURE 4.7: Density plot of the non-linear susceptibility of graphene, $\Gamma_l(q, \omega, T)$, for a temperature (a) $T = 50$ K and (b) $T = 300$ K with $E_F = 0.064$ eV.

Apart from the general higher effective upper cut-off in frequency, the specific reason for the enhancement of the Coulomb drag intensity at higher T is due to the fact that $U_{12}(q, \omega, T)$ is inversely proportional to $\epsilon_{RPA}(q, \omega, T)$. The latter becomes very small (vanishing at $T = 0$) for certain $\omega(q)$ corresponding to the plasmons of the system, here optical and acoustic modes, thus making $U_{12}(q, \omega, T)$ very large at the $\omega(q)$ of these plasmons. At low temperature, the contribution of the plasmons to the Coulomb drag is suppressed because the non-linear susceptibility, $\Gamma_l(q, \omega, T)$, vanishes in the region $v_F q < \omega < v_F(2k_F - q)$ where the plasmons lie, figure 4.7 (a). However, at high temperature, $\Gamma_l(q, \omega, T)$ acquires a non-vanishing finite value in this region that overlaps with the plasmon-phonon modes, as shown in figure 4.7 (b). In this figure, the hybridized SPP modes are indicated by the superimposed black lines. Therefore, the plasmon-phonon modes start to contribute to the Coulomb drag intensity, as pointed out in figures 4.4 (f). It should also be noted that the plasmon enhancement of the Coulomb drag is primarily due to acoustic mode, since it is lower in energy than the optical mode and runs closer to the PHC, thus facilitating its excitation at lower temperatures than the optical mode. Nonetheless, the optical mode also contributes to the drag, being its contribution larger the higher the temperature.

4.4 Theory vs experiment

The drag resistivity, ρ_D , has been calculated numerically as a function of the temperature, T , Fermi energy, E_F and the h-BN interlayer thickness, d . In order to isolate the effects of the different quasiparticle excitations, ρ_D has been calculated with and without phonons, for the whole $\omega - q$ space as well as for the inter- and intraband regions separately.

Figures 4.8 (a)-(c) present ρ_D as a function of T showing a good agreement with the experimental data of drag resistivity [52]. In all cases, ρ_D is initially enhanced with increasing temperature, peaking at around 300 K. Beyond that threshold, ρ_D starts to decrease. In the G/h-BN/G system considered here, this peak gets more intense when the plasmon-phonon hybridization is considered (solid lines), as compared to the case where only plasmons are taken into account (dashed lines). Thus,

the full consideration of the hybridized SPP and HPP modes leads to a 10, 16, and 25% percent higher values of ρ_D for $d = 4, 6$, and 8 nm, respectively. Although the overall magnitude of ρ_D decreases with increasing d , the larger difference between the with and without phonons cases for the thicker h-BN interlayer points out the strong contribution arising from the HPP modes confined in the h-BN reststrahlen bands, whose number increases with increasing d . The final reduction of ρ_D beyond the peak temperature is a consequence of the merging of the PHC with plasmon modes as explained earlier.

FIGURE 4.8: ρ_D as a function of the temperature for a h-BN thickness (a) $d = 4$ nm, (a) $d = 6$ nm and (b) $d = 9$ nm with $E_F = E_{F1} = E_{F2} = 0.064$ eV.

In figures 4.9 (a) and 4.9 (b), the ρ_D of figure 4.8 (b) is calculated separately for the inter- and intraband regions, respectively. In the interband region, figure 4.9 (a), ρ_D slightly decreases when considering the h-BN phonons because the two plasmon branches split into multiple hybridized SPP modes which have finite lifetime. This splitting occurs around the two reststrahlen bands of h-BN. Since only HPP modes are allowed to propagate in the reststrahlen bands and they primarily lie in the intraband region, the spectral weight of hybridized SPP modes in the interband region is smaller than that of their counterparts (plasmon modes) when h-BN phonons are not considered. Therefore, there is a net slight reduction in the interband contribution to ρ_D with inclusion of h-BN phonons. In the intraband region, figure 4.9 (b), the difference in the ρ_D when h-BN phonons are considered is the result of the enhancement of ρ_D by the HPP modes which become more prominent even at room temperature and beyond. Figure 4.9 (c) shows the qualitative evolution of ρ_D with respect to the temperature (normalized by the Fermi temperature T_F) due to the contributions of the SPP and HPP modes, which are shown separately as shaded colour regions. For $T \leq 0.01T_F$, $\rho_D \sim T^3$ as shown in the inset. Above that range and for $T \leq 0.07T_F$ (T_{PHC}), $\rho_D \sim T^{2.2}$ is dominated by the PHC. When the temperature is increased further, the SPP modes start to contribute with $\rho_D \sim T^{1.75}$ till $T_{SPP} (\sim 0.3T_F)$. In the absence of any other excitations, ρ_D is expected to decrease with increasing temperature. However, figure 4.9 (c) shows that ρ_D keeps increasing shifting the maximum value of ρ_D towards a higher temperature, $T_{HPP} (\sim 0.45T_F)$ and providing a $\rho_D \sim T^{0.35}$ dependence due to the contribution from the HPP modes. Beyond T_{HPP} , the system enters finally into the Boltzmann regime where ρ_D decreases as $\sim 1/T^2$.

The calculated dependence of ρ_D on E_F and d is shown in figures 4.10 (a)-(c) along with the experimental data from ref. [52]. The comparison shows an excellent agreement with the theory. The ρ_D values decrease with increasing E_F , as shown in figures 4.10 (a) and 4.10 (b) for $T = 240$ K and 300 K, respectively. The functional form of ρ_D varies as $\rho_D \sim 1/E_F^\beta$ (or $\sim 1/n^{\beta/2}$), with a β value of 2.3, 2.7, and 2.7 for $d = 4, 6$, and 9 nm respectively, which agrees well with the reported experimental data

FIGURE 4.9: ρ_D , same as in figure 4.8 (b) but calculated separately for the (a) interband and (b) intraband regions as a function of temperature with (solid line) and without phonons (dashed line). (c) Dependence of ρ_D in figure 6(b) on T/T_F with the contributions from the PHC and the different quasiparticles (SPPs and HPPs).

from ref. [52]. Similarly, ρ_D decreases with increasing d as the two graphene layers start to decouple, as shown in figure 4.10 (c) for both $T=240$ K and 300 K. In this case, the functional form of ρ_D has been assessed separately for the $k_F d < 1$ and $k_F d > 1$ regimes, with ρ_D varying as $1/d^\gamma$ with $\gamma = 1.5$ and 1.75, respectively. It should be noted that, close to Dirac point, the effects of the charge density fluctuations, known as electron-hole puddles, in the two graphene layers need to be taken into account [121]. In the calculations presented here, far from Dirac point, these effects have been neglected.

FIGURE 4.10: ρ_D as a function of the Fermi energy E_F ($E_F = E_{F1} = E_{F2}$) for (a) $T = 240$ K and (b) $T = 300$ K. (c) ρ_D as a function h-BN interlayer thickness d .

Chapter 5

SAW-mediated exciton-plasmons in 2D semiconductor/metal

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5.1 Exciton-plasmon coupling in MoS₂, WS₂

The strong coupling regime in light-matter interactions can be achieved by placing standard semiconductor nanostructures (quantum wells and quantum dots) in distributed Bragg reflectors (DBRs) and/or plasmonic microcavities [123, 124]. Recently, there has been a growing interest in coupling excitons in 2D semiconductor crystals to surface plasmon polaritons (SPPs, hereafter simply called plasmons) in metals [79]. The deep subwavelength confinement of the plasmons results in a strong light-matter interaction with ultra-small mode volume and hence enables the strong exciton-plasmon coupling. Excitons in 2D semiconductors have large binding energies, exceeding 100 meV, thus making their detection possible at room temperature [125, 126]. This contrasts with GaAs/AlGaAs quantum wells (QWs), where the exciton binding energy is approximately 5 meV causing the excitonic recombinations at low temperatures.

In contrast to excitons in semiconductors, plasmons in metals cannot be generated by directly shining light as their wave vectors lie on the right side of the light line in vacuum leading to a large momentum mismatch. To overcome this, metals are typically either fabricated into nanostructures (like nanoparticles, gratings etc.) or prism coupling techniques are used to excite plasmons. All these methods require extra fabrication steps and make the plasmon generation unscalable. Here we propose an alternative approach using a SAW to create a dynamic periodic grating using an IDT in a similar way as discussed in chapter 3 for the coupling of surface plasmon-phonon polaritons in graphene. However, in this case, a thin continuous (unpatterned) metal layer is placed between the piezoelectric substrate and the 2D semiconducting crystal.

In piezoelectric materials, a SAW is accompanied by both periodic strain and piezoelectric fields. The strain field induces a type-I modulation of the energy band gap E_g via the deformation potential interaction whereas the piezoelectric field creates a type-II modulation of E_g [116, 127, 128] (see the cover image for illustration of the modulation of E_g by a SAW, page 39). The latter has been shown to permit the long-range acoustic transport of electrons and holes by piezoelectric SAWs [129]. However, this modulation dissociates and ionizes the optically generated excitons into packets of electrons and holes confined in the moving lateral potential wells separated by one-half wavelength of the SAW [130]. Hence, we need type-I modulation only for sustaining excitons that can be produced by the strain field of the SAW, which coincidentally also leads to the generation of metal plasmons and hence the exciton-plasmon coupling. One option to create only SAW strain field is to use a non-piezoelectric propagation direction, for example the [100] direction in GaAs [129], but this configuration will not work in the case of 2D semiconductors as MoS₂ and WS₂ with an odd number of layers and black phosphorus (BP) with both odd and even number of layers, since they are intrinsically piezoelectric [131, 132]. Hence the strain field of the SAW propagating in [100] direction of GaAs can interact with piezoelectric 2D semiconductors and lead to quenching (or recombination) of the excitons [81] by establishing polarization due to the piezoelectric effect, i.e. a type-II modulation of E_g . Another possibility is to use a thin metal layer in between the piezoelectric substrate and the 2D semiconductor crystal, which can completely screen the piezoelectric fields of both the SAW and the 2D semiconductor crystal. The latter will be our proposed device configuration.

In this chapter, we theoretically demonstrate that the strong coupling regime can be achieved by coupling excitons in 2D crystals and metal plasmons by a SAW. This coupling can be inferred as a SAW-enhanced absorption in the reflection spectra with distinctively large Rabi splitting.

Here we consider a geometry of 2D crystal (MoS₂, WS₂, BP)/Ag/LiNbO₃ (see the cover image for the device schematic, page 39). Silver (Ag) is chosen as a plasmonic metal because its mass density is smaller than that of gold (Au), the other typically employed metal for plasmonics, as mass loading damps the SAW amplitude. The dielectric function of Ag can be given in Drude model form [133, 134] as

$$\epsilon_{Ag}(\omega) = \epsilon_{\infty} - \frac{\omega_p^2}{\omega(\omega + i\gamma_{Ag})} \quad (5.1)$$

where $\epsilon_{\infty} = 3$ is the high frequency dielectric constant, $\omega_p = 9.1$ eV is the plasma frequency and $\gamma_{Ag} = 20$ meV is the damping rate. LiNbO₃ is a strong piezoelectric material with a large electromechanical coupling coefficient and its dielectric constant is $\epsilon_{LiNbO_3} = 5$ [42]. The plasmon dispersion of Ag films of various thickness,

d_{Ag} , on a LiNbO₃ substrate is plotted in figure 5.1. We observe that for larger thicknesses the plasmonic resonance shifts toward shorter wavelengths. For example, for $d_{Ag} > 15$ nm, the Ag plasmon dispersion closely follows the light-line dispersion (accounting for the LiNbO₃ substrate refractive index, $n_{LiNbO_3} = \sqrt{\epsilon_{LiNbO_3}}$), whereas for $d_{Ag} = 5$ nm, there is a clear bending and a deviation from the light line. The 2D

FIGURE 5.1: Plasmon dispersion in Ag films of various thickness, d_{Ag} , on a LiNbO₃ substrate.

materials can be described by their 2D conductivity as

$$\sigma(\omega) = \frac{-2i\omega |\Psi|^2 \langle |e \cdot d|^2 \rangle}{E_{ex} - \omega - i\gamma_{ex}} \quad (5.2)$$

where Ψ is the exciton wave function, e is the photon polarization, d is the transition dipole moment matrix element, E_{ex} is the exciton energy and γ_{ex} is exciton linewidth taken as 1 ps^{-1} ($= 0.66 \text{ meV}$) [135, 136]. For the strong coupling, the Rabi splitting ($\hbar\omega_R$) must exceed the total damping rate, i.e. $\hbar\omega_R > \gamma_{ex} + \gamma_{Ag}$. The thickness of the metal should be chosen in such a way that it completely screen the piezoelectric field of both SAW and 2D crystal. For Ag, the minimum thickness required to completely screen the field was reported to be 5 nm [137, 138]. To calculate the polariton

dispersion we make use of the transfer matrix method (TMM, see section 2.1.3).

FIGURE 5.2: Exciton-Plasmon dispersion for (a) MoS₂ and (b) WS₂ on a 15 nm thick Ag layer on LiNbO₃ with their Hopfield coefficients shown beneath each graph. In the upper panel, the solid black line is the dispersion of the Ag plasmon and the dashed black line is the exciton energy.

In figure 5.2 we plot the exciton-plasmon polariton dispersion for MoS₂ and WS₂ on a Ag layer with a thickness of 15 nm. The exciton energy, E_{ex} , for MoS₂ and WS₂ is 1.9 and 2.0 eV respectively. The exciton-plasmon coupling results in two polaritonic branches, called upper polariton (UP) and lower polariton (LP). The SAW wavelength at which the Rabi splitting occurs is also indicated in the graphs. The exciton-plasmon energy dispersion can be described by a coupled oscillator model as:

$$\begin{bmatrix} E_{ex} + i\gamma_{ex} & g \\ g & E_{pl}(q) + i\gamma_{Ag} \end{bmatrix} \begin{bmatrix} X \\ C \end{bmatrix} = E \begin{bmatrix} X \\ C \end{bmatrix} \quad (5.3)$$

where E_{ex} is the exciton energy, $E_{pl}(q)$ is the q -dependent plasmon resonance energy, g is the exciton-plasmon coupling strength, E is the eigenvalue corresponding to the energies of the polariton modes, X and C are the eigenvectors referred to as the Hopfield coefficients such that $|X|^2 + |C|^2 = 1$. The analytical equation (the eigenvalue, E) of the two polariton branches, where the +, - sign corresponds to UP and LP respectively, can be given as

$$E_{\pm} = \frac{E_{ex} + E_{pl}(q)}{2} + \frac{i(\gamma_{ex} + \gamma_{Ag})}{2} \pm \frac{\sqrt{4g^2 + [E_{ex} - E_{pl}(q) + i(\gamma_{ex} - \gamma_{Ag})]^2}}{2} \quad (5.4)$$

where $g = \sqrt{(E_{ex} - E_-)(E_+ - E_{ex})}$ and $E_{pl}(q) - E_{ex} = \zeta$ is called as the detuning between the plasmon resonance and the exciton transition energy. The lifetime of

the two polaritons can be directly determined by the Hopfield coefficients and γ_{ex} , γ_{Ag} as:

$$\gamma_{LP} = |X|^2\gamma_{ex} + |C|^2\gamma_{pl} \quad (5.5)$$

$$\gamma_{UP} = |C|^2\gamma_{ex} + |X|^2\gamma_{pl} \quad (5.6)$$

A thinner metal will lead to a larger value of the Rabi splitting but at higher q ($= 2\pi/\lambda_{SAW}$) values. This is due to the fact that by decreasing the metal thickness, the plasmon dispersion, $E_{pl}(q)$, will move further away from the light line (light cone) towards a larger value of q , thus a smaller wavelength (see figure 5.1). And a smaller wavelength implies a larger electric field density and higher proportion of this electric field distribution lying in the vicinity of the 2D excitonic medium (i.e., the spatial overlap between the field of the plasmon and the exciton increases). Also, the Rabi splitting depends on the coupling strength, g , which can be enhanced by using emitters with a large dipole moment (i.e., a large oscillator strength) and/or by enhancing the electric field of the plasmonic cavity mode (plasmon with small wavelength). The latter is proportional to $1/\sqrt{V}$, where V is the mode volume, which for plasmonic nanocavities goes as $\sim \lambda^3$ [139, 140], λ being the plasmon wavelength. Therefore, confining the electromagnetic energy in a small volume V enhances the interaction and thus leads to a large Rabi splitting. One major challenge for the current SAW technology is the limitation in achieving smaller wavelengths. With e-beam and nano-imprint lithography one can typically reach down to 160 nm SAW wavelength [141–143] but beyond that is difficult. One way to overcome this challenge is to increase the metal thickness so that the metal plasmon dispersion will move towards the light line and hence the maxima of the exciton-plasmon splitting will occur at smaller q values (see figure 5.1). Although this is at the expense of a reduction in the Rabi splitting, this is still large enough to be in the strong coupling regime.

5.2 Exciton-plasmon coupling efficiency by SAW

FIGURE 5.3: Change in the reflectance spectra induced by a SAW with an amplitude of $\delta = 5$ Å in (a) MoS₂ at $\lambda_{SAW} = 200$ nm and (b) WS₂ at $\lambda_{SAW} = 185$ nm on a 15 nm thick Ag layer on LiNbO₃. The dashed curves are the bare reflectance curves of the Ag plasmon (i.e., without the exciton coupling) which once coupled to the exciton split into two peaks corresponding to upper (UP) and lower (LP) polariton branches.

Coupling of exciton and plasmon leads to enhanced absorption of light, which

can be interpreted as a change in reflectance that can be measured directly. As mentioned earlier, SAWs can be used to generate a plasmon and its coupling with the exciton. Figure 5.3 plots the differential reflectance or extinction spectra of MoS₂ and WS₂ on a 15 nm-thick Ag on a LiNbO₃ substrate and a SAW amplitude of 5 Å at a SAW wavevector corresponding to that at which the Rabi splitting ($\hbar\omega_R$) occurs as can be seen in figure 5.2. The solid peaks in the extinction spectra correspond to the coupling of exciton and plasmon and the separation between them is a direct measurement of the Rabi frequency, whereas the dashed peak corresponds to the generation of metal plasmon only. The obtained values of $\hbar\omega_R$ are comparable with the reported values in the literature [84].

5.3 Exciton-plasmon coupling in black phosphorus

Similarly to MoS₂ and WS₂, phosphorene or black phosphorus (BP) is a 2D semiconductor that has a layer-dependent direct band gap. MoS₂ and WS₂ are direct band gap only in the monolayer (ML) case otherwise being an indirect band gap material while mono- and few-layer BP is a direct band gap material. The exciton-plasmon dispersion for 1 ML BP on Ag layers with different Ag thickness is plotted in figure 5.4 (a)-(c). The exciton energy for monolayer BP is 1.1 eV [126], allowing

FIGURE 5.4: Exciton-plasmon dispersion for BP/Ag system on LiNbO₃ with Ag thickness of (a) 5 nm, (b) 10 nm and (c) 15 nm. The SAW wavelength, λ_{SAW} , at which Rabi splitting occurs is also indicated in the graphs. (d) Variation in the Rabi splitting, $\hbar\omega_R$, and the coupling strength, g , as a function of the Ag layer thickness.

the exciton-plasmon coupling to occur at low wavevector q values (i.e. large SAW wavelengths). Hence, in this case the Ag thickness can be reduced further down to 5 nm without compromising the upper limit of the SAW wavevector. This enhances the strong coupling as shown by the increase in the Rabi frequency with decreasing Ag thickness as depicted in figure 5.4 (d) along with the coupling strength, g , which is related to $\hbar\omega_R$ as

$$\hbar\omega_R = \sqrt{4g^2 - (\gamma_{ex} - \gamma_{Ag})^2} \quad (5.7)$$

5.4 Effect of layer number on exciton-plasmon in black phosphorus

Since BP remains direct band gap even with few layers, hence few-layer BP can also host excitons with a layer dependent exciton energy. In figure 5.5 (a)-(d) exciton-plasmon dispersion for 2 to 5 layer thick BP on top of a 5 nm thick Ag is plotted. The exciton energy decreases from 0.84 eV to 0.55 eV when going from 2 layer BP to 5 layer BP [126]. The increase in layer number makes the Rabi splitting to occur at small q values, which can ease the fabrication of the SAW devices while the system remains in the strong coupling regime as $\hbar\omega_R$ reduces only from 130 meV to 90 meV when going from 2 to 5 MLs.

FIGURE 5.5: Exciton-Plasmon dispersion for BP/Ag (5 nm) system on LiNbO₃ with varying monolayer (ML) number, (a)-(d) from 2 to 5 MLs. The SAW wavelength, λ_{SAW} , at which Rabi splitting occurs is also indicated in the graphs.

Chapter 6

Modulation of phonons in graphene by SAW

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6.1 Visualization of SAW by interferometry

The measurements of the SAW vertical component of the particle displacement, u_z , were performed using a Michelson interferometer as described in section 2.2.6. The spatial plot of the vertical particle displacement, u_z , is shown in figure 6.1 along with a reference signal (red) without SAW. Here only one IDT (IDT1) is excited by an rf signal while IDT2 remains idle thus creating a travelling wave. Even though IDT2

FIGURE 6.1: Particle displacement u_z measured by interferometry.

is not powered, but having several pairs of metal fingers, it acts as a reflector in addition with reflector 1 and 2 thus forming a kind of 1-port SAWR. The magnitude of u_z is large between the two IDTs indicating cavity effects formed by back and forth reflections from the two IDTs, i.e. between points c and d. The purpose of reflectors 1 and 2 here is to reflect back the SAW signal, as shown by the diminishing value of u_z within the two reflectors i.e., beyond points b and e. In a SAW transducer, a certain

FIGURE 6.2: Zoomed in portion of figure 6.1 showing the detail of part of region between IDT1 and IDT2

amount of detection, regeneration and reflection occurs at each finger, producing distortions in the time and frequency response of the SAW device. In transducers with large numbers of fingers such effects can be quite severe. IDT1 has 100 finger pairs, and because of this large number of finger pairs, part of IDT1 itself act as a reflector for the generated SAW. The large value of u_z at the edge of IDT1 confirms

FIGURE 6.3: Particle displacement u_z between IDT1 and IDT2 as a function of the applied rf power to IDT1

that the IDT1 itself is acting partially as a reflector for the SAW so that not the whole SAW is leaving the IDT1. Therefore an optimum design of the IDT should have a reduced number of finger pairs. Figure 6.2 plots a zoomed in portion of figure 6.1

between points c and d i.e., between the two IDTs. The period of oscillations of u_z here is $\sim 10 \mu\text{m}$, as expected for this device design ($\lambda_{\text{SAW}} = 10 \mu\text{m}$). In figure 6.3 u_z is plotted for an arbitrary region between IDT1 and IDT2 as a function of the rf power applied to IDT1 with IDT2 being idle (thus creating a travelling wave). The peak-peak amplitude of u_z increases as a square root of the applied rf power. A slight shift is also observed in u_z when changing the rf is due to the backlash of stage motor on which the sample is mounted over a long measurement time.

6.2 Standing-SAW-modulated Raman spectroscopy

We have investigated the effects of the SAW on the G and 2D Raman bands of the SLG. The 2-port SAWR produces two counter-propagating travelling SAWs forming a standing SAW pattern with a maximum amplitude at the anti-nodes that is twice as that of a single travelling SAW. Note that an IDT with a perfect reflector will have an amplitude twice that of an IDT without reflector, since the SAW generation at the IDT is bidirectional in nature. First, for the reference, we obtained the Raman spectra of the graphene G peak under very low SAW rf power excitation. When the rf power is very low, the SAW amplitude is also very small and thus the effect on the Raman intensity of any strain produced by the wave will be negligible as shown in figure 6.4. Here net differential response, $(I_{\text{off}} - I_{\text{on}})/I_{\text{off}}$, is plotted which can be obtained from equation 6.1 and 6.1.

FIGURE 6.4: Raman spectra of the G peak for the SAW OFF, I_{off} , and SAW ON conditions, I_{on} , for $f = 385.75 \text{ MHz}$.

The effects of the SAW have been first assessed by measuring the Raman spectra at different positions on the sample, marked as A, B, and C on the optical micrograph of the device shown in figure 6.5 (a). The corresponding Raman spectra of the G peak for the SAW ON and SAW OFF cases are shown in figure 6.5 (b). The intensity modulation and broadening of the peak are only observed within the path of

the SAW beam (position A), whereas the Raman spectrum remains unaltered at the edge of and outside that region (positions B and C, respectively). This proves that the modulation observed is solely due to the SAW strain field. In addition, the frequency of the rf signal applied to the IDTs has also been varied across the passband region of the SAWR, as shown in figure 6.5 (c). The dips in S_{11} (and peaks in S_{12}) are due to the different modes of the acoustic resonator. In this case, the maximum phonon modulation occurs at a frequency of 385.75 MHz, figure 6.5 (d), where the transmission losses (S_{12}) of the SAW devices are minimum. This modulation fades away rapidly out of the resonance frequency and is negligible for the main side lobes (at 378, 389, and 393 MHz). This also proves that the modulation observed is solely driven by the SAW strain field and rules out any thermal contribution by the laser illumination [144] and the rf dissipation. Furthermore, it demonstrates that the graphene phonons can be modulated dynamically at high frequencies by tuning the rf signal at the IDT. The slight variations in the center wavenumber of the G peak observed in figure 6.5 (b) at different laser positions on the sample are due to the variations in the native strain and residual doping of the graphene, whereas those observed in figure 6.5 (d) for the same laser position and different rf frequencies are produced by small mechanical and thermal drifts. However, these drifts do not af-

FIGURE 6.5: (a) Optical micrograph of the SAWR with the transferred graphene in between the IDTs. (b) Raman spectra of the G peak for the SAW ON and SAW OFF conditions ($f = 385.75$ MHz and $P_{rf} = 23$ dBm) at three different positions on the device marked as A, B, and C in (a). (c) S_{11} (reflection) and S_{12} (transmission) losses of the device. (d) Raman spectra of the G peak for the SAW ON and SAW OFF conditions ($P_{rf} = 23$ dBm) at the different frequencies indicated by dashed vertical lines in (c).

fect the differential analysis of the SAW OFF and SAW ON conditions measured at a fixed rf frequency and power, as the above mentioned modulation method cancels out the effects of any systematic fluctuations on the Raman signal.

Figure 6.6 displays the Raman spectra of the G and 2D peaks for various P_{rf} values for both SAW ON, I_{on} , and SAW OFF, I_{off} , cases. The net differential response $(I_{off} - I_{on})/I_{off}$ is also plotted. To fit the obtained Raman spectra of G and 2D peak of graphene in the SAW OFF case, we used Breit-Wigner-Fano (BWF) function [145], given as

$$I_{off}(\nu) = I_0 + b\nu + \frac{H \left(1 + \frac{\nu - \nu_c}{q_{BWF}\Gamma}\right)^2}{1 + \left(\frac{\nu - \nu_c}{\Gamma}\right)^2} \quad (6.1)$$

where I_0 is the base line, b is the slope of straight line used for background removal, H is the height of the peak, ν_c is the centre of the peak, Γ is the spectral width, and $1/q_{BWF}$ is the asymmetry factor. Setting $1/q_{BWF} = 0$ cancels out the contribution from the continuum of electronic excitations, giving a Lorentzian line shape, which represents a discrete phonon spectrum. For the SAW ON case, I_{on} , Raman spectra show an additional line broadening of the G peak (and 2D peak also) and a decrease in H for a certain range of applied rf power, P_{rf} values. This broadening in the G peak line shape is interpreted to be caused by the dynamic strain field of the SAW. Thus, the oscillating tensile and compressive cycles of the strain modulate periodically the phonon frequency around its equilibrium value, see figure 6.7. Since the acquisition time of the Raman spectra is much larger than the SAW period, the phonon oscillation manifests itself as a broadening of the peak line shape and a reduction of its maximum intensity, see figure 6.8. Both the broadening and the intensity reduction increase with increasing P_{rf} . In order to quantify the modulation of the graphene phonon by the strain field of the standing SAW, we have fitted the I_{on} spectra with the following integral equation:

$$I_{on}(\nu) = I_0 + b\nu + \frac{1}{2\pi} \int_0^{2\pi} \frac{H \left(1 + \frac{\nu - (\nu_c - M \cos \theta)}{q_{BWF}\Gamma}\right)^2}{1 + \left(\frac{\nu - (\nu_c - M \cos \theta)}{\Gamma}\right)^2} d\theta \quad (6.2)$$

where I_0 , b , H , ν_c , Γ , and $1/q_{BWF}$ are the fitting parameters extracted previously from the BWF fitting of $I_{off}(\nu)$ for each value of P_{rf} . The amplitude of the SAW-induced phonon frequency modulation is given by M , which is the only free parameter in the fitting of $I_{on}(\nu)$. M is the absolute value of the SAW-induced modulation, whereas its sign variation within the wave cycle is accounted for by the cosine term.

The effects of the local SAW strain observed in the graphene G and 2D Raman lines are found to be qualitatively similar to those described in the modulation of the LO phonon Raman line of bulk semiconductor crystals [118]. They are also similar to those observed in the photoluminescence of SAW-modulated quantum wells [146] and dots [147–149]. In Si, the differential Raman signal is slightly asymmetric [118]. The asymmetry was attributed to the scattering events under a SAW involving LO phonons with small wave vectors, which have energies slightly lower than the one for the zone-center mode. This type of asymmetry has not been observed for the graphene G Raman line, thus indicating that these scattering events are not important. Furthermore, the piezoelectric field accompanying the SAW can modulate the carrier concentration in semiconductors leading to broadenings and shifts of the Raman frequency [150]. In the case of SLG, however, the SAW-induced carrier density modulation has been estimated to be of the order of 10^{10} cm^{-2} , which is negligible as compared to the residual carrier density in the material. Nonetheless, the SAW strain modulation observed in graphene is quantitatively much stronger than in the semiconductors due to the combination of the larger Grüneisen parameters of

FIGURE 6.6: Raman spectra of the G (left) and 2D (right) peak for the SAW OFF, I_{off} , and SAW ON conditions, I_{on} , for $f = 385.75$ MHz and increasing values of the applied rf power P_{rf} . The experimental spectra (symbols) are fitted (lines) with Eq. 6.1 and 6.2, respectively. The net differential response $(I_{off} - I_{on})/I_{off}$ is also plotted (blue curve). Larger P_{rf} values were applied in case of 2D peak, as compared to the G peak, to achieve a noticeable modulation.

graphene and the more efficient generation of the strain in the 2D layer on a strong piezoelectric substrate, as we will discuss in the next section.

The amplitude of the SAW-induced phonon frequency modulation given by M (c.ref. equation 6.2) attains a non-zero value only within the SAW beam path, for the SAW resonance frequency, and for a large enough RF power. It must be noted that in the case of the 2D peak, $1/q_{BWF}$ is very small, as expected for a band hardly affected by the continuum of electronic excitations. A weaker acoustic modulation is observed for the 2D band as compared to the G band, so that much higher P_{rf} values have been required to obtain a comparable modulation to that of the G band. This is expected to be related to the larger width of the 2D peak that partially hinders the modulation making it less apparent.

6.3 Dynamic strain

Figures 6.9 (a) and 6.9 (b) display the evolution of the value of M for the G, $|M_G|$, and 2D, $|M_{2D}|$, peaks, respectively, as a function of the absolute value of the hydrostatic strain, ϵ_0 , generated by the standing SAW at the surface of the LiNbO₃ substrate where the SLG lies. Since the SAW is a Rayleigh mode [151], i.e. with sagittal polarization, it has only particle displacement component along the x and z directions (c.ref. figure 2.10 (a)). Therefore, the hydrostatic component of the strain field is then $\epsilon_0 = \epsilon_{xx} + \epsilon_{zz}$. The details of the calculation of ϵ_0 as a function of the P_{rf} applied to

FIGURE 6.7: Time evolution of the frequency of Raman G peak under a period of the SAW excitation.

FIGURE 6.8: (a) Instantaneous G peak position at different times within the SAW period. (b) G peak intensity reduction and broadening under SAW excitation.

the IDTs are described in Appendix B. Here it has been assumed that the graphene placed on top of LiNbO₃ is massless and has strong adhesion to the substrate, so that it mimics the exact strain profile at the surface of the LiNbO₃ substrate. Thus, the whole strain produced by the SAW is transferred to the graphene with 100 % efficiency, as it has been proven valid for small strain values [152]. Moreover, the applied strain by the SAW produced a reversible modulation in the Raman spectra, indicating that there was no slippage or permanent modification of the sample when stressed. Figures 6.9 (a) and 6.9 (b) display the data from two devices (A and B) with identical SAWR but with transferred graphene from two different batches. The linear fittings in figure 6.9 (a) provide a slope value or rate $\partial M_G / \partial \epsilon_0$ of 49 ± 2 and 46 ± 2 cm⁻¹/% for devices A and B, respectively. Similar fittings in figure 6.9 (b) provide a rate $\partial M_{2D} / \partial \epsilon_0$ of 39 ± 1 and 44 ± 1 cm⁻¹/%, respectively, for the same devices. The obtained values are then consistent for both graphene batches used. It has to be noticed that although the strain produced by the SAWR has a quasi-uniaxial character, since ϵ_{zz} is an order of magnitude smaller than ϵ_{xx} , the polycrystalline nature of

FIGURE 6.9: Absolute value of the modulation, $M_{G(2D)}$, of the (a) G- and (b) 2D-peak frequency as a function of the absolute value of the hydrostatic strain, ε_0 , produced by the SAW for devices A and B using graphene from two different batches. Experimental values (symbols) and their linear fittings (lines) are shown. The slight differences in the behaviour of the two devices are attributed to variations in the graphene quality and the IDT efficiency.

the CVD SLG used for this study averages out the contribution from several crystal domains, so that the well-known G- [86, 95, 96] and 2D-peak [87, 153, 154] splitting in (monocrystalline) exfoliated graphene flakes under uniaxial strain is not expected to occur here. In addition, these splittings typically require a uniaxial strain value beyond approximately 0.3 % [86, 87, 95, 96, 153, 154] to be observed clearly. Here, the maximum strain value produced by the SAWR, which depends on the P_{rf} applied to the IDTs, was limited by the maximum gain of the amplifier used. However, larger SAW-induced strain values can be achieved using a higher power amplifier along with an optimized SAWR design to minimize the losses. Therefore, these splittings are foreseen to be induced by the SAWR in SLG monocrystals (CVD or exfoliated).

From the $\partial M_{G(2D)} / \partial \varepsilon_0$ rates one can calculate the Grüneisen parameters, $\gamma_{G(2D)}$, which represent the rate of phonon mode softening (hardening) under tensile (compressive) strain given as

$$\gamma_{G(2D)} = \frac{1}{\nu_c^{G(2D)}} \frac{\partial M_{G(2D)}}{\partial \varepsilon_0}, \quad (6.3)$$

where $\nu_c^{G(2D)}$ is the centre of the position of the G (2D) peak for the unperturbed (SAW OFF) condition. Equation 6.3 does not include a negative sign as it has been particularized for the case under study where M and ε_0 are absolute values. The values of γ_G obtained for devices A and B are 3.1 ± 0.1 and 2.8 ± 0.1 , respectively, whereas the values of the corresponding γ_{2D} are 1.43 ± 0.04 and 1.63 ± 0.04 , respectively.

The Grüneisen parameters determine the thermomechanical properties of a material. In the case of graphene, there is still a large dispersion of values reported in the literature, with γ_G and γ_{2D} in the ranges of 0.69-2.4 and 2.98-3.8, respectively [86, 95, 155–157]. This variation in the γ can be attributed to the different type of strain applied, the quality of the graphene, the substrate used [158], and the effective adhesion of the graphene to the substrate [152]. The smaller value of γ_{2D} than of γ_G obtained in this study seems to be an artefact related to the less efficient SAW-induced modulation of the 2D band due to its broader nature, as shown in figure 6.6. On the other hand, the larger values of γ_G obtained in this study, as compared

to those reported in the literature, might arise from the large difference in the coefficient of thermal expansion, CTE , of the polymers used in the case of graphene on flexible substrates, such as SU8 in ref. [95] ($CTE = 5.2 \times 10^{-5} \text{ K}^{-1}$ [159]) or PDMS in ref. [86] ($CTE = 9\text{-}9.6 \times 10^{-4} \text{ K}^{-1}$ [160]), and of LiNbO_3 ($CTE = 7.5\text{-}15 \times 10^{-6} \text{ K}^{-1}$ [161]), as well as due to the dynamic nature of the strain produced by the SAW on the graphene/ LiNbO_3 system in contrast to the static strain in the case of the graphene/polymer systems on a flexible substrate.

Most bulk semiconductors have Grüneisen parameters for the zone-center TO and LO (or degenerated LTO) phonons γ_{LTO} with values around 1 [135] and phonon frequencies ν_c^{LTO} in the range of $250\text{-}550 \text{ cm}^{-1}$. Therefore, the Raman frequency sensitivity to strain of these materials is expected to be of approximately $5 \text{ cm}^{-1}/\%$, an order of magnitude smaller than the $\partial M_{G(2D)}/\partial \epsilon_0$ rates obtained here for graphene modulated by the SAW strain field. In addition, graphene can be easily transferred directly onto a strong piezoelectric substrate, for example LiNbO_3 used in this study, providing a large SAW strain amplitude. Conversely, semiconductors are typically weakly or non-piezoelectric, the latter requiring an additional piezoelectric layer on top of them to generate the SAW. Thus, in these cases, the SAW strain amplitude is either small or only a fraction of that produced at the piezoelectric layer reaches the active semiconductor region [162]. This makes SAWs specially powerful and well-suited tools for the strain modulation of graphene and the development of novel devices thereof.

Chapter 7

Conclusions and future work

In **chapter 3**, we have theoretically demonstrated that SAWs can be used to generate SPPs and HPPs in G/h-BN heterostructures on a piezoelectric substrate, such as AlN and ZnO, where the SAW creates a dynamic virtual diffraction grating overcoming the wave vector mismatch of SPPs and HPPs with the incident light. Graphene electrons couple to surface phonons of both the h-BN interlayer and the piezoelectric substrate providing a complex SPP dispersion relation that strongly depends on the thickness of the h-BN film and can be modulated by varying the Fermi energy of graphene. In addition, hyperbolic phonons appear in the case of multilayer h-BN films that also couple with the graphene carriers leading to tunable HPPs. The h-BN interlayer is demonstrated to not only significantly change the hybridized SPP dispersion but also to enhance the SPP lifetime, as compared to the simpler G/AlN(ZnO) system. The lifetime enhancement provided by the h-BN film is shown to be related to both the higher carrier mobility induced in graphene and the larger lifetime of the h-BN surface phonons. Therefore, the G/h-BN/AlN(ZnO) system provides an advantageous platform with greater plasmon robustness and tunability.

This study has also demonstrated that the choice of piezoelectric substrate is important, as the overlap of its reststrahlen band with that of h-BN hinders the full usage of the SPPs originating from the coupling of the graphene carriers and the h-BN phonons, which have the largest lifetime. This is the case of AlN, where the reststrahlen bands overlap, as compared to ZnO, where they do not. Moreover, the study has been extended to double layer graphene systems separated by h-BN, where the interaction between the graphene layers leads to a larger number of SPPs. In particular, the G/h-BN/G/ZnO system has been demonstrated to further broaden out the energy range covered by the SPPs as well as to increase their intensity, as compared to its G/h-BN/ZnO counterpart. All these results pave the way for engineering SAW-based G/h-BN plasmonic devices and metamaterials covering the mid-IR to THz range such as THz lasers [12], antennas [13, 14], waveguides [15], modulators [16], polarizers [17], plasmonic detectors and sensors [21–24].

In **chapter 4**, we have theoretically studied the Coulomb drag mechanism in a h-BN/G/h-BN/G/h-BN system and have evaluated the contribution of plasmons and phonons as well as their hybridization thereto. We have also included the effects of XC in the standard RPA, and found that these effects further modify the plasmon modes of the system and thus the drag resistivity. XC has allowed us to replicate the experimental data reported in the literature to a very good accuracy. In order to discriminate the different quasiparticles and their effects, the contributions from the inter- and intraband regions with and without h-BN phonons have been evaluated. We have showed that apart from the drag enhancement led by the graphene plasmons, their hybridization with the hyperbolic phonons of h-BN can further enhance

the drag resistivity even at room temperature and beyond. This contrasts with the surface-like hybridized plasmon-phonon modes in the interband region, whose contribution to the drag resistivity reduces, as compared to the unhybridized plasmon modes, due to their splitting imposed by the h-BN reststrahlen bands. In addition, the calculated maximum of the Coulomb drag resistivity takes place at room temperature for physically realizable experimental conditions. This result is relevant, as Coulomb drag experiments in G/h-BN/G systems have not been yet explored for temperatures larger than 240 K [52].

The described mechanisms of the enhancement of the Coulomb drag in the G/h-BN/G system, led by the graphene plasmons hybridized with the hyperbolic phonons of h-BN and XC, can also be used to describe and understand more complex heterostructures of double BLG systems such as BLG/WSe₂/BLG [163] and BLG/h-BN/BLG [164], where Coulomb drag has already been measured at room temperature [165, 166]. Furthermore, the same theoretical approach can be applied to understand the Coulomb drag mechanism in other 2D material systems with a h-BN interlayer, such as phosphorene/h-BN/phosphorene [167], or more generally to any future double 2D layer systems including a naturally hyperbolic 2D interlayer material, such as, for example, black phosphorous [168], transition metal dichalcogenides [169], or graphene superposed on black phosphorous. The latter has been shown to lead to strained superlattices, which in turn create large pseudomagnetic fields [170] that are shown to have substantial effects on the Coulomb drag [52].

In **chapter 5**, we have theoretically proven that SAWs can also be used to couple the optically generated excitons in 2D semiconductors (MoS₂, WS₂ and BP) and plasmons in a metal (Ag). The thickness of Ag was chosen in such a way that it completely screens the piezoelectric field of both SAW and 2D semiconductors which are also inherently piezoelectric (MoS₂ and WS₂ with an odd number of layer and BP with both odd and even number of layer), which can ionize the excitons, leaving behind only the SAW strain field. This coupling will lead to two polaritonic branches with large enough Rabi frequency leading to the strong coupling regime. This Rabi frequency can be increased (decreased) by increasing (decreasing) the thickness of Ag and will occur at large (small) SAW wavevector, which is a SAW device design parameter. In the case of BP, which remains as a direct band gap semiconductor up to few layers in contrast with MoS₂ and WS₂ which are direct band gap only in single layer, this Rabi frequency decreases with increasing the layer number.

The proposed exciton-plasmon coupling mechanism can be extended to any other direct band gap 2D semiconductor, which can be easily transferred onto a piezoelectric substrate that can generate a SAW. The obtained results can be extended to more complicated studies like trapping of exciton-plasmons in the moving potential created by the band gap modulation produced by the SAW. In addition, the band gap modulation by the SAW strain field can also induces a longitudinal force on the carriers which is proportional to dE_g/dx , where x is the SAW propagation direction. This longitudinal force is much smaller than that due to the piezoelectric field for frequencies smaller than 1 GHz. However, in our study, since the operating SAW frequencies are several GHz, this force could also allow us to transport exciton-plasmons over several hundreds of microns with reduced shape distortion.

In **chapter 6**, we have experimentally investigated the SAW-induced modulation of the phonons of graphene by Raman spectroscopy using the SAW resonators on LiNbO₃, where a standing wave is produced on the transferred graphene sheet. We have observed strong effects in both first-order and second-order (or two-phonon)

Raman modes, achieving intensity variations of 15 % and phonon frequency shifts of around 10 cm^{-1} when the SAW is applied. The strain produced by the SAW can be varied by changing the applied rf power and frequency allowing us to dynamically tune both the Raman intensity and the phonon frequency. SAWs might also permit to generate strain along multiple axes and tailor the strain fields. For example, a bi-axial strain pattern can be generated by either using orthogonal SAWRs or selecting crystal cuts and propagation directions providing comparable ϵ_{xx} and ϵ_{yy} components [171]. An adequate design of the in- and out-of-plane strain components of the SAW might also be useful for tailoring gauge fields in graphene [90, 172]. In addition, the amount of strain generated in the graphene could be enhanced by using focusing IDTs [173]. On the other hand, in this study, the piezoelectric field accompanying the SAW in LiNbO_3 leads to a variation of the graphene carrier density of the order of 10^{10} cm^{-2} . Although this value is 2 orders of magnitude smaller than the residual density of the PMMA assisted wet-transferred CVD graphene, it could be used for locally modulating highly-clean graphene encapsulated by h-BN (typically produced by dry transfer techniques) where the background/residual doping (fixed charges) is of the same order of magnitude. This carrier density modulation has been shown to modify the BWF line shape [145]. Thus, the h-BN would allow us to gate the graphene near the Dirac point where the carrier density is minimum and the SAW-induced carrier density modulation becomes significant.

The modulation mechanism reported here is not limited to single-layer graphene but can be extended to any other single- or few-layer 2D material, and where engineering opens up a whole range of new possibilities [174, 175]. In addition, many 2D materials are themselves piezoelectric [176], enhancing the capabilities of strain modulation. In the case of graphene, for example, the E_{2g} mode has been shown to modify the plasmonic properties of bilayer graphene [177], so that a SAW could be used to tune hybridized plamon-phonon modes for surface-enhanced Raman spectroscopy [178].

In summary, in this thesis, we have investigated the various quasiparticles (plasmons, phonons, and excitons) in graphene and other 2D materials, including h-BN, TMDCs, and BP. We have paid special attention to their coupling (plasmon-phonons and plasmon-excitons) and their influence on other phenomena such as Coulomb drag. We have also addressed in detail their interaction with light, the latter being modulated or mediated by a SAW. Here, the strain field of a SAW has been shown to modulate the phonons of graphene or to generate a virtual diffraction grating capable of coupling the light into plasmons in graphene or thin metals. This strain is tailored by means of the high-frequency localized deformations produced by the acoustic transducers fabricated on a piezoelectric substrate, where the graphene or any other 2D material is transferred to. Therefore, SAWs are powerful tools for the generation and manipulation of these quasiparticles, paving the way for new devices and technologies based on 2D materials and van der Waals heterostructures.

Appendix A

Materials

A.1 Graphene conductivity	61
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A.1 Graphene conductivity

The frequency-dependent conductivity of graphene in the local limit ($q \rightarrow 0$) and for $\omega \gg \tau_e^{-1}$ is given by the Kubo formula as

$$\sigma(\omega) = \frac{\omega + i\tau_e^{-1}}{i\pi\hbar} \left(\frac{1}{(\omega + i\tau_e^{-1})^2} \int_0^\infty dE E \left(\frac{\partial F(E)}{\partial E} - \frac{\partial F(-E)}{\partial E} \right) + \int_0^\infty dE \frac{F(E) - F(-E)}{(\omega + i\tau_e^{-1})^2 - 4(E/\hbar)^2} \right) \quad (\text{A.1})$$

where $F(E) = [\exp(E/k_B T) + 1]^{-1}$ is the Fermi-Dirac distribution, μ_c is the chemical potential and τ_e is the carrier relaxation time. The first and second terms in the above equation represent the contributions of the intraband ($\hbar\omega < 2E_F$) and interband ($\hbar\omega > 2E_F$) transitions, respectively as shown in figure A.1. In the static

FIGURE A.1: Frequency dependent conductivity of graphene.

FIGURE A.2: (a) Density plot of $\sigma_{ll}(T, E_F)/\tau$. The horizontal black and vertical red lines are the cross section of graphene conductivity for a fixed Fermi energy ($E_F = 0.064$ eV (carrier density, $n=3 \times 10^{11} \text{ cm}^{-2}$) and fixed temperature, $T=300$ K, and are plotted in (c) and (d) respectively. (b) Graphene Drude weight μ_D (black curve) and its chemical potential μ_c (red curve) as a function of temperature for $E_F = 0.064$ eV.

limit, the intralayer conductivity of graphene has the following form

$$\sigma_{ll} = e^2 \mu_D \tau / (\pi \hbar^2), \quad (\text{A.2})$$

where τ is the carrier transport time in graphene, which is assumed to be energy independent, and μ_D is the effective Drude weight, which depends on the carrier density and the temperature T , and is given by

$$\mu_D = \mu_c + 2k_B T \ln[1 + \exp(-\mu_c/k_B T)], \quad (\text{A.3})$$

The latter can be calculated solving the following relation for the Fermi energy:

$$E_F = k_B T \sqrt{2 \int_0^\infty (f_- - f_+) x dx}, \quad (\text{A.4})$$

where

$$f_\pm = \frac{1}{1 + \exp(x \pm \mu_c/k_B T)}. \quad (\text{A.5})$$

Equation A.4 can be simplified to

$$E_F = k_B T \sqrt{2 [Li_2[-\exp(-\mu_c/k_B T)] - Li_2[-\exp(\mu_c/k_B T)]]} \quad (\text{A.6})$$

where

$$Li_2(x) = \int_x^0 \frac{\ln(1-t)}{t} dt \quad (\text{A.7})$$

is Spence's function [179]. The density plot of the graphene conductivity normalized by the carrier transport time, $\sigma_{II}(T, E_F)/\tau$, is shown in figure A.2 (a). The chemical potential and the Drude weight as a function of the temperature for a fixed Fermi energy of $E_F = 0.064$ eV are plotted in figure A.2 (b). Figures A.2 (c) and A.2 (d) present, respectively, the temperature dependence of this function for $E_F = 0.064$ eV and the Fermi energy dependence of this function for $T = 300$ K, corresponding to the horizontal and vertical cross section lines indicated in figure A.2 (a).

A.2 h-BN permittivity

h-BN is a polar dielectric material with an extremely large crystalline anisotropy resulting from strong, in-plane covalent bonding of boron and nitrogen atoms and weak, out-of-plane, interlayer van der Waals bonding. The strong anisotropy results in a large birefringence, where within the reststrahlen bands, the dielectric permittivities along orthogonal crystal axes are not only different, but opposite in sign. hence referred as hyperbolic materials. The frequency-dependent dielectric function or relative permittivity of h-BN is given by [27]

$$\epsilon_m(\omega) = \epsilon_{\infty,m} \left(1 + \frac{(\omega_{LO}^m)^2 - (\omega_{TO}^m)^2}{(\omega_{TO}^m)^2 - i\gamma_m\omega - \omega^2} \right) \quad (\text{A.8})$$

where $m = x, z$ are the in-plane and out-of-plane components and $\epsilon_{\infty,x} = 2.95$ and $\epsilon_{\infty,z} = 4.87$ are the high-frequency dielectric constants, $\omega_{TO}^x = 0.096$ eV, $\omega_{LO}^x = 0.102$ eV, $\omega_{TO}^z = 0.169$ eV, and $\omega_{LO}^z = 0.199$ eV are the phonon frequencies, and $\gamma_x = 0.49$ meV and $\gamma_z = 0.62$ meV are the damping frequencies. The real and imaginary parts of $\epsilon_m(\omega)$ are plotted in figures A.3 (a) and A.3 (b), respectively. The

FIGURE A.3: (a) Real and (b) imaginary part of the h-BN dielectric function, where $\epsilon_{x(z)}(\omega)$ is the in- (out-of-) plane relative permittivity. The vertical dashed lines are the phonon frequencies of h-BN.

real part of $\epsilon_m(\omega)$ becomes negative in the reststrahlen bands ($\omega_{TO}^m < \omega < \omega_{LO}^m$). The opposite sign in the in-plane and out-of-plane components within these bands makes h-BN a hyperbolic material. The lower band presents type-I (two-sheeted) hyperbolicity ($Re[\epsilon_x] < 0, Re[\epsilon_z] > 0$), whereas the upper band has type-II (single-sheeted) hyperbolicity ($Re[\epsilon_x] > 0, Re[\epsilon_z] < 0$), as shown in figure A.3 (a). The isofrequency curves of h-BN, plotted in figure A.4, are described by the equation of a hyperboloid:

$$Re[\epsilon_z(\omega)]^{-1}k_z^2 + Re[\epsilon_x(\omega)]^{-1}(k_x^2 + k_y^2) = (\omega/c)^2 \quad (\text{A.9})$$

FIGURE A.4: Isofrequency surfaces of h-BN.

FIGURE A.5: Plot of (a) $\epsilon_s(\omega)$ and (b) $\epsilon_a(\omega)$. Both $\epsilon_s(\omega)$ and $\epsilon_a(\omega)$ vary rapidly around the two reststrahlen bands. The y axis is the absolute magnitude of $\epsilon_s(\omega)$ and $\epsilon_a(\omega)$ both of which are complex functions of ω and the filling colour is the argument.

The expressions $\epsilon_s(\omega) = \sqrt{\epsilon_z(\omega)\epsilon_x(\omega)}$ and $\epsilon_a(\omega) = \sqrt{\epsilon_z(\omega)/\epsilon_x(\omega)}$ are plotted in figure A.5. The phase remains the same for $\epsilon_s(\omega)$ in the two reststrahlen bands but it is of opposite sign for $\epsilon_a(\omega)$, hence leading to a variation in the total phase of the exponential term of eq. 2.5 within these two reststrahlen bands which gives rise to the hyperbolic modes confined within these bands [180].

A.3 AlN and ZnO permittivity

The frequency-dependent dielectric function of the piezoelectric materials, AlN and ZnO, is given by [115]

$$\epsilon(\omega) = \epsilon_\infty + (\epsilon_0 - \epsilon_\infty) \left(\frac{\omega_{TO}^2}{\omega_{TO}^2 - i\gamma\omega - \omega^2} \right) \quad (\text{A.10})$$

where $\epsilon_0 = 7.37$ (7.46) and $\epsilon_\infty = 3.93$ (3.61) are the static and high frequency dielectric constants, respectively. $\omega_{TO} = 0.083$ eV (0.05 eV) are the phonon frequencies and γ is the phonon damping rate for AlN (ZnO). We assumed $\gamma=1$ ps for both AlN

and ZnO. Dielectric function of AlN and ZnO are plotted in figure A.6 (a) and A.6 (b), respectively.

FIGURE A.6: Real and imaginary parts of $\epsilon(\omega)$ for (a) AlN and (b) ZnO. The vertical blue lines are the phonon frequencies of AlN and ZnO.

Appendix B

SAW-induced strain vs rf power density

The output signal from an rf generator was split and amplified, in order to drive each IDT of the SAWR with an rf power P_{rf} (dBm). The SAW power density (power per unit of SAW beam width) generated by each IDT was then calculated using the following relation [181]

$$P_{IDT}(W/m) = \frac{10^{\left[\frac{(S_{12}/2)(dB)}{10}\right]} 10^{\left[\frac{(P_{rf}(dBm) - 30)}{10}\right]}}{W_{IDT}(m)} \quad (B.1)$$

The half of the transmission parameter $S_{12}(dB)$ of the SAWR takes into account the electromechanical efficiency of a single IDT and reflection grating.

Rayleigh SAWs have a mixed compressional and shear character, leading to a displacement field vector

$$u_R = (u_x, 0, u_z), \quad (B.2)$$

where u_x and u_z are the longitudinal and shear vertical components, respectively, and the shear horizontal component u_y is typically zero. In the particular case of 128° rotated Y-cut X-propagating LiNbO₃, u_y is not zero but approximately 20 times smaller than the other two components, so it has been neglected. Thus, using the Voigt notation, the strain field is written as

$$\varepsilon_R = (\varepsilon_{xx}, 0, \varepsilon_{zz}, 0, 2\varepsilon_{xz}, 0), \quad (B.3)$$

where the strain tensor components are given by

$$\varepsilon_{kl} = \frac{1}{2} \left(\frac{\partial u_k}{\partial x_l} + \frac{\partial u_l}{\partial x_k} \right) \quad (B.4)$$

and the zero components in equation B.3 appear because $u_y = \partial u_x / \partial y = \partial u_z / \partial y = 0$. The particle displacements have been calculated by solving numerically the coupled elastic and electromagnetic equations for the LiNbO₃ substrate. This allow us to calculate the strain components and the hydrostatic strain $\varepsilon_0 = \varepsilon_{xx} + \varepsilon_{zz}$ produced by the SAWR in the LiNbO₃ substrate, and transmitted to the graphene layer on top. Figure B.1 shows the amplitude of ε_0 as a function of P_{IDT} (W/m), where a square root dependence is observed. The maximum strain is achieved at the anti-nodes of the standing wave produced by the SAWR (where both IDTs are excited), which is 2 times larger than that generated by a travelling wave excited by a single IDT. The sign of the strain depends in both cases on the part of the cycle of the wave.

FIGURE B.1: Amplitude of the hydrostatic strain ε_0 produced by the SAWR at the anti-nodes of the standing wave pattern (both IDTs excited, solid line) as function of P_{IDT} (W/m). For comparison, the strain associated to a travelling wave generated by a single IDT (dashed line) is also depicted.

Appendix C

Local field factors

The intra- and interlayer frequency-dependent LFFs, $|G_{11}(q, \omega)|$ and $|G_{12}(q, \omega)|$, are plotted in figures C.1, for various q values. It can be observed that $|G_{ij}(q, \omega)|$, similarly to $|r_s(\omega)|$, attains a maximum value around the two restrahlen bands of h-BN. The intensity of these maxima increases with increasing q/k_F , as shown in the density plots of $|G_{11}(q, \omega)|$ and $|G_{12}(q, \omega)|$ depicted in figure C.2.

FIGURE C.1: Absolute value of the (a) $|G_{11}(q, \omega)|$ and (b) $|G_{12}(q, \omega)|$ LFFs as a function of ω for $E_F = 0.064$ eV. The vertical dashed lines are the phonon frequencies of h-BN.

FIGURE C.2: Density plot of the absolute value of the (a) $|G_{11}(q, \omega)|$ and (b) $|G_{12}(q, \omega)|$ LFFs for $E_F = 0.064$ eV. The horizontal dashed lines are the phonon frequencies of h-BN.

Bibliography

- [1] Alexander S. Mayorov et al. "Micrometer-Scale Ballistic Transport in Encapsulated Graphene at Room Temperature". *Nano Letters* 11.6 (June 2011), pp. 2396–2399 (cit. on p. 1).
- [2] Alexander A. Balandin. "Thermal properties of graphene and nanostructured carbon materials". *Nature Materials* 10.8 (July 2011), pp. 569–581 (cit. on p. 1).
- [3] C. Lee et al. "Measurement of the Elastic Properties and Intrinsic Strength of Monolayer Graphene". *Science* 321.5887 (July 2008), pp. 385–388 (cit. on p. 1).
- [4] Frank HL Koppens, Darrick E Chang, and F Javier Garcia de Abajo. "Graphene plasmonics: a platform for strong light–matter interactions". *Nano letters* 11.8 (2011), pp. 3370–3377 (cit. on p. 1).
- [5] Antonio I Fernández-Domínguez, Francisco J Garcia-Vidal, and Luis Martín-Moreno. "Unrelenting plasmons". *Nature Photonics* 11.1 (2017), p. 8 (cit. on p. 1).
- [6] Yurui Fang and Mengtao Sun. "Nanoplasmonic waveguides: towards applications in integrated nanophotonic circuits". *Light: Science & Applications* 4.6 (2015), e294 (cit. on p. 1).
- [7] Mark L Brongersma and Pieter G Kik. *Surface plasmon nanophotonics*. Vol. 131. Springer, 2007 (cit. on p. 1).
- [8] Vladimir M Shalaev. "Optical negative-index metamaterials". *Nature photonics* 1.1 (2007), p. 41 (cit. on p. 1).
- [9] Nanfang Yu and Federico Capasso. "Flat optics with designer metasurfaces". *Nature materials* 13.2 (2014), pp. 139–150 (cit. on p. 1).
- [10] Mark S Tame et al. "Quantum plasmonics". *Nature Physics* 9.6 (2013), pp. 329–340 (cit. on p. 1).
- [11] Achim Woessner et al. "Highly confined low-loss plasmons in graphene–boron nitride heterostructures". *Nature materials* 14.4 (2015), p. 421 (cit. on p. 2).
- [12] Alexander A Dubinov et al. "Terahertz surface plasmons in optically pumped graphene structures". *Journal of Physics: Condensed Matter* 23.14 (2011), p. 145302 (cit. on pp. 2, 57).
- [13] Long Ju et al. "Graphene plasmonics for tunable terahertz metamaterials". *Nature nanotechnology* 6.10 (2011), p. 630 (cit. on pp. 2, 57).
- [14] Nikitas Papasimakis et al. "Graphene in a photonic metamaterial". *Optics express* 18.8 (2010), pp. 8353–8359 (cit. on pp. 2, 57).
- [15] Ashkan Vakil and Nader Engheta. "Transformation optics using graphene". *Science* 332.6035 (2011), pp. 1291–1294 (cit. on pp. 2, 57).
- [16] F Javier Garcia de Abajo. "Graphene nanophotonics". *Science* 339.6122 (2013), pp. 917–918 (cit. on pp. 2, 57).

- [17] Yu V Bludov, Mikhail I Vasilevskiy, and Nuno MR Peres. "Tunable graphene-based polarizer". *Journal of Applied Physics* 112.8 (2012), p. 084320 (cit. on pp. 2, 57).
- [18] Sukosin Thongrattanasiri, Frank HL Koppens, and F Javier Garcia De Abajo. "Complete optical absorption in periodically patterned graphene". *Physical review letters* 108.4 (2012), p. 047401 (cit. on p. 2).
- [19] A Yu Nikitin, F Guinea, and Luis Martin-Moreno. "Resonant plasmonic effects in periodic graphene antidot arrays". *Applied Physics Letters* 101.15 (2012), p. 151119 (cit. on p. 2).
- [20] VG Kravets et al. "Surface hydrogenation and optics of a graphene sheet transferred onto a plasmonic nanoarray". *The Journal of Physical Chemistry C* 116.6 (2012), pp. 3882–3887 (cit. on p. 2).
- [21] Zheyu Fang et al. "Graphene-antenna sandwich photodetector". *Nano letters* 12.7 (2012), pp. 3808–3813 (cit. on pp. 2, 57).
- [22] Pradeep Kumar Maharana and Rajan Jha. "Chalcogenide prism and graphene multilayer based surface plasmon resonance affinity biosensor for high performance". *Sensors and Actuators B: Chemical* 169 (2012), pp. 161–166 (cit. on pp. 2, 57).
- [23] Yuan Liu et al. "Plasmon resonance enhanced multicolour photodetection by graphene". *Nature communications* 2 (2011), p. 579 (cit. on pp. 2, 57).
- [24] Hai Hu et al. "Gas identification with graphene plasmons". *Nature communications* 10.1 (2019), p. 1131 (cit. on pp. 2, 57).
- [25] Jianing Chen et al. "Optical nano-imaging of gate-tunable graphene plasmons". *Nature* 487.7405 (2012), p. 77 (cit. on p. 2).
- [26] Zhe Fei et al. "Gate-tuning of graphene plasmons revealed by infrared nano-imaging". *Nature* 487.7405 (2012), p. 82 (cit. on p. 2).
- [27] S Dai et al. "Graphene on hexagonal boron nitride as a tunable hyperbolic metamaterial". *Nature nanotechnology* 10.8 (2015), p. 682 (cit. on pp. 2, 16, 63).
- [28] Thomas J Constant et al. "All-optical generation of surface plasmons in graphene". *Nature Physics* 12.2 (2016), pp. 124–127 (cit. on p. 2).
- [29] Choon How Gan. "Analysis of surface plasmon excitation at terahertz frequencies with highly doped graphene sheets via attenuated total reflection". *Applied Physics Letters* 101.11 (2012), p. 111609 (cit. on p. 2).
- [30] Yu V Bludov, MI Vasilevskiy, and NMR Peres. "Mechanism for graphene-based optoelectronic switches by tuning surface plasmon-polaritons in monolayer graphene". *EPL (Europhysics Letters)* 92.6 (2011), p. 68001 (cit. on p. 2).
- [31] Leyong Jiang et al. "Perfect terahertz absorption with graphene surface plasmons in the modified Otto configuration". *Plasmonics* 12.6 (2017), pp. 1825–1831 (cit. on p. 2).
- [32] Jin Yao et al. "Multiple resonant excitations of surface plasmons in a graphene stratified slab by Otto configuration and their independent tuning". *Photonics Research* 5.4 (2017), pp. 377–384 (cit. on p. 2).
- [33] A Yu Nikitin, P Alonso-González, and R Hillenbrand. "Efficient coupling of light to graphene plasmons by compressing surface polaritons with tapered bulk materials". *Nano letters* 14.5 (2014), pp. 2896–2901 (cit. on p. 2).

- [34] Huguen Yan et al. "Damping pathways of mid-infrared plasmons in graphene nanostructures". *Nature Photonics* 7.5 (2013), p. 394 (cit. on p. 2).
- [35] Isaac John Luxmoore et al. "Strong coupling in the far-infrared between graphene plasmons and the surface optical phonons of silicon dioxide". *ACS photonics* 1.11 (2014), pp. 1151–1155 (cit. on p. 2).
- [36] Huguen Yan et al. "Tunable infrared plasmonic devices using graphene/insulator stacks". *Nature nanotechnology* 7.5 (2012), p. 330 (cit. on p. 2).
- [37] Weilu Gao et al. "Excitation of plasmonic waves in graphene by guided-mode resonances". *ACS nano* 6.9 (2012), pp. 7806–7813 (cit. on p. 2).
- [38] Weilu Gao et al. "Excitation and active control of propagating surface plasmon polaritons in graphene". *Nano letters* 13.8 (2013), pp. 3698–3702 (cit. on p. 2).
- [39] Jurgen Schiefele et al. "Coupling Light into Graphene Plasmons through Surface Acoustic Waves". *Physical Review Letters* 111.23 (Dec. 2013), p. 237405 (cit. on p. 2).
- [40] Bertram Alexander Auld. *Acoustic fields and waves in solids.*, 1973 (cit. on p. 2).
- [41] Claudia Ruppert et al. "Surface acoustic wave mediated coupling of free-space radiation into surface plasmon polaritons on plain metal films". *Physical Review B* 82.8 (2010), p. 081416 (cit. on pp. 2, 4, 24).
- [42] X Sun, S Shiokawa, and Y Matsui. "Interactions of surface plasmons with surface acoustic waves and the study of the properties of Ag films". *Journal of applied physics* 69.1 (1991), pp. 362–366 (cit. on pp. 2, 4, 40).
- [43] DN Basov, MM Fogler, and FJ Garcia De Abajo. "Polaritons in van der Waals materials". *Science* 354.6309 (2016), aag1992 (cit. on p. 3).
- [44] BN Narozhny and A Levchenko. "Coulomb drag". *Reviews of Modern Physics* 88.2 (2016), p. 025003 (cit. on pp. 2, 3).
- [45] Antti-Pekka Jauho and Henrik Smith. "Coulomb drag between parallel two-dimensional electron systems". *Physical Review B* 47.8 (1993), p. 4420 (cit. on p. 3).
- [46] Lian Zheng and AH MacDonald. "Coulomb drag between disordered two-dimensional electron-gas layers". *Physical Review B* 48.11 (1993), p. 8203 (cit. on p. 3).
- [47] Alex Kamenev and Yuval Oreg. "Coulomb drag in normal metals and superconductors: Diagrammatic approach". *Physical Review B* 52.10 (1995), p. 7516 (cit. on p. 3).
- [48] Karsten Flensberg et al. "Linear-response theory of Coulomb drag in coupled electron systems". *Physical Review B* 52.20 (1995), p. 14761 (cit. on p. 3).
- [49] TJ Gramila et al. "Mutual friction between parallel two-dimensional electron systems". *Physical Review Letters* 66.9 (1991), p. 1216 (cit. on p. 3).
- [50] U Sivan, PM Solomon, and H Shtrikman. "Coupled electron-hole transport". *Physical Review Letters* 68.8 (1992), p. 1196 (cit. on p. 3).
- [51] Seyoung Kim et al. "Coulomb drag of massless fermions in graphene". *Physical Review B* 83.16 (2011), p. 161401 (cit. on p. 3).

- [52] RV Gorbachev et al. "Strong Coulomb drag and broken symmetry in double-layer graphene". *Nature Physics* 8.12 (2012), p. 896 (cit. on pp. 3, 30, 35, 36, 37, 58).
- [53] M Titov et al. "Giant magnetodrag in graphene at charge neutrality". *Physical Review Letters* 111.16 (2013), p. 166601 (cit. on p. 3).
- [54] Wang-Kong Tse, Ben Yu-Kuang Hu, and S Das Sarma. "Theory of Coulomb drag in graphene". *Physical Review B* 76.8 (2007), p. 081401 (cit. on p. 3).
- [55] NMR Peres, JMB Lopes Dos Santos, and AH Castro Neto. "Coulomb drag and high-resistivity behavior in double-layer graphene". *EPL (Europhysics Letters)* 95.1 (2011), p. 18001 (cit. on p. 3).
- [56] MI Katsnelson. "Coulomb drag in graphene single layers separated by a thin spacer". *Physical Review B* 84.4 (2011), p. 041407 (cit. on p. 3).
- [57] EH Hwang, Rajdeep Sensarma, and S Das Sarma. "Coulomb drag in monolayer and bilayer graphene". *Physical Review B* 84.24 (2011), p. 245441 (cit. on p. 3).
- [58] M Carrega et al. "Theory of Coulomb drag for massless Dirac fermions". *New Journal of Physics* 14.6 (2012), p. 063033 (cit. on p. 3).
- [59] BN Narozhny et al. "Coulomb drag in graphene: perturbation theory". *Physical Review B* 85.19 (2012), p. 195421 (cit. on pp. 3, 30).
- [60] Bruno Amorim and NMR Peres. "On Coulomb drag in double layer systems". *Journal of Physics: Condensed Matter* 24.33 (2012), p. 335602 (cit. on p. 3).
- [61] Jonathan Lux and Lars Fritz. "Kinetic theory of Coulomb drag in two monolayers of graphene: From the Dirac point to the Fermi liquid regime". *Physical Review B* 86.16 (2012), p. 165446 (cit. on p. 3).
- [62] Chuanyi Zhang and Guojun Jin. "Electron-hole pair condensation and Coulomb drag effect in a graphene double layer". *Journal of Physics: Condensed Matter* 25.42 (2013), p. 425604 (cit. on p. 3).
- [63] M Schütt et al. "Coulomb drag in graphene near the Dirac point". *Physical Review Letters* 110.2 (2013), p. 026601 (cit. on pp. 3, 30).
- [64] Justin CW Song, Dmitry A Abanin, and Leonid S Levitov. "Coulomb drag mechanisms in graphene". *Nano Letters* 13.8 (2013), pp. 3631–3637 (cit. on p. 3).
- [65] Karsten Flensberg and Ben Yu-Kuang Hu. "Coulomb drag as a probe of coupled plasmon modes in parallel quantum wells". *Physical Review Letters* 73.26 (1994), p. 3572 (cit. on pp. 3, 4).
- [66] Karsten Flensberg and Ben Yu-Kuang Hu. "Plasmon enhancement of Coulomb drag in double-quantum-well systems". *Physical Review B* 52.20 (1995), p. 14796 (cit. on p. 3).
- [67] N. P. R. Hill et al. "Correlation Effects on the Coupled Plasmon Modes of a Double Quantum Well". *Physical Review Letters* 78 (11 1997), pp. 2204–2207 (cit. on p. 3).
- [68] H. C. Tso, P. Vasilopoulos, and F. M. Peeters. "Direct Coulomb and phonon-mediated coupling between spatially separated electron gases". *Physical Review Letters* 68 (1992), pp. 2516–2519 (cit. on p. 3).

- [69] Chao Zhang and Y. Takahashi. “Dynamical conductivity of a two-layered structure with electron-acoustic phonon coupling”. *Journal of Physics: Condensed Matter* 5.28 (1993), p. 5009 (cit. on p. 3).
- [70] T. J. Gramila et al. “Evidence for virtual-phonon exchange in semiconductor heterostructures”. *Physical Review B* 47 (1993), pp. 12957–12960 (cit. on p. 3).
- [71] Martin Chr. Bønsager et al. “Frictional drag between quantum wells mediated by phonon exchange”. *Physical Review B* 57 (1998), pp. 7085–7102 (cit. on p. 3).
- [72] R. Jalabert and S. Das Sarma. “Quasiparticle properties of a coupled two-dimensional electron-phonon system”. *Physical Review B* 40 (1989), pp. 9723–9737 (cit. on p. 3).
- [73] K. Güven and B. Tanatar. “Coupled plasmon-phonon mode effects on the Coulomb drag in double-quantum-well systems”. *Physical Review B* 56 (1997), pp. 7535–7540 (cit. on p. 3).
- [74] Samvel M Badalyan and Francois M Peeters. “Enhancement of Coulomb drag in double-layer graphene structures by plasmons and dielectric background inhomogeneity”. *Physical Review B* 86.12 (2012), p. 121405 (cit. on p. 3).
- [75] Artsem A Shylau and Antti-Pekka Jauho. “Plasmon-mediated Coulomb drag between graphene waveguides”. *Physical Review B* 89.16 (2014), p. 165421 (cit. on p. 3).
- [76] Bruno Amorim et al. “Coulomb drag in graphene–boron nitride heterostructures: Effect of virtual phonon exchange”. *Physical Review B* 86.12 (2012), p. 125448 (cit. on p. 4).
- [77] Kin Fai Mak and Jie Shan. “Photonics and optoelectronics of 2D semiconductor transition metal dichalcogenides”. *Nature Photonics* 10.4 (2016), p. 216 (cit. on p. 4).
- [78] Xi Ling et al. “The renaissance of black phosphorus”. *Proceedings of the National Academy of Sciences* 112.15 (2015), pp. 4523–4530 (cit. on p. 4).
- [79] Wenjing Liu et al. “Strong exciton–plasmon coupling in MoS₂ coupled with plasmonic lattice”. *Nano letters* 16.2 (2016), pp. 1262–1269 (cit. on pp. 4, 39).
- [80] Amgad R. Rezk et al. “Acoustic–Excitonic Coupling for Dynamic Photoluminescence Manipulation of Quasi-2D MoS₂ Nanoflakes”. *Advanced Optical Materials* 3.7 (2015), pp. 888–894 (cit. on p. 4).
- [81] Amgad R. Rezk et al. “Acoustically-Driven Trion and Exciton Modulation in Piezoelectric Two-Dimensional MoS₂”. *Nano Letters* 16.2 (2016), pp. 849–855 (cit. on pp. 4, 40).
- [82] Edwin Preciado et al. “Scalable fabrication of a hybrid field-effect and acousto-electric device by direct growth of monolayer MoS₂/LiNbO₃”. *Nature Communications* 6 (2015), p. 8593 (cit. on p. 4).
- [83] JVT Buller et al. “Dynamical and Tuneable Modulation of the Tamm Plasmon/Exciton-Polariton Hybrid States Using Surface Acoustic Waves.” *Acta Physica Polonica, A*. 129 (2016) (cit. on p. 4).
- [84] PAD Gonçalves et al. “Plasmon-exciton polaritons in two-dimensional semiconductor/metal interfaces”. *Physical Review B* 97.4 (2018), p. 041402 (cit. on pp. 5, 44).

- [85] Vitor M. Pereira, A. H. Castro Neto, and N. M. R. Peres. "Tight-binding approach to uniaxial strain in graphene". *Physical Review B* 80.4 (July 2009) (cit. on p. 5).
- [86] M. Huang et al. "Phonon softening and crystallographic orientation of strained graphene studied by Raman spectroscopy". *Proceedings of the National Academy of Sciences* 106.18 (Apr. 2009), pp. 7304–7308 (cit. on pp. 5, 55, 56).
- [87] Duhee Yoon, Young-Woo Son, and Hyeonsik Cheong. "Strain-Dependent Splitting of the Double-Resonance Raman Scattering Band in Graphene". *Physical Review Letters* 106.15 (Apr. 2011) (cit. on pp. 5, 55).
- [88] Chen Si et al. "First-Principles Calculations on the Effect of Doping and Biaxial Tensile Strain on Electron-Phonon Coupling in Graphene". *Physical Review Letters* 111.19 (Nov. 2013), p. 196802 (cit. on p. 5).
- [89] N. Levy et al. "Strain-Induced Pseudo-Magnetic Fields Greater Than 300 Tesla in Graphene Nanobubbles". *Science* 329.5991 (July 2010), pp. 544–547 (cit. on p. 5).
- [90] F. Guinea, M. I. Katsnelson, and A. K. Geim. "Energy gaps and a zero-field quantum Hall effect in graphene by strain engineering". *Nature Physics* 6.1 (Sept. 2009), pp. 30–33 (cit. on pp. 5, 59).
- [91] Miao Zhou et al. "Strain effects on hydrogen storage capability of metal-decorated graphene: A first-principles study". *Applied Physics Letters* 97.10 (Sept. 2010), p. 103109 (cit. on p. 5).
- [92] Ovidiu Cretu et al. "Migration and Localization of Metal Atoms on Strained Graphene". *Physical Review Letters* 105.19 (Nov. 2010) (cit. on p. 5).
- [93] M. L. Ng et al. "Controlling Hydrogenation of Graphene on Transition Metals". *The Journal of Physical Chemistry C* 114.43 (Oct. 2010), pp. 18559–18565 (cit. on p. 5).
- [94] Z. F. Wang, Yu Zhang, and Feng Liu. "Formation of hydrogenated graphene nanoripples by strain engineering and directed surface self-assembly". *Physical Review B* 83.4 (Jan. 2011) (cit. on p. 5).
- [95] T. M. G. Mohiuddin et al. "Uniaxial strain in graphene by Raman spectroscopy: Gpeak splitting, Gruneisen parameters, and sample orientation". *Physical Review B* 79.20 (May 2009) (cit. on pp. 5, 55, 56).
- [96] Otakar Frank et al. "Compression Behavior of Single-Layer Graphenes". *ACS Nano* 4.6 (May 2010), pp. 3131–3138 (cit. on pp. 5, 55).
- [97] Hongyi Mi et al. "Creating periodic local strain in monolayer graphene with nanopillars patterned by self-assembled block copolymer". *Applied Physics Letters* 107.14 (2015), p. 143107 (cit. on p. 5).
- [98] Yuhang Jiang et al. "Visualizing Strain-Induced Pseudomagnetic Fields in Graphene through an hBN Magnifying Glass". *Nano Letters* 17.5 (2017), pp. 2839–2843 (cit. on p. 5).
- [99] Péter Nemes-Incze et al. "Preparing local strain patterns in graphene by atomic force microscope based indentation". *Scientific Reports* 7 (2017), p. 3035 (cit. on p. 5).
- [100] Per Delsing et al. "The 2019 surface acoustic waves roadmap". *Journal of Physics D: Applied Physics* 52.35 (July 2019), p. 353001 (cit. on p. 5).

- [101] MR Ramezanali et al. "Finite-temperature screening and the specific heat of doped graphene sheets". *Journal of Physics A: Mathematical and Theoretical* 42.21 (2009), p. 214015 (cit. on p. 7).
- [102] Giovanni Vignale Gabriele Giuliani. *Quantum Theory of the Electron Liquid*. Cambridge University Press, Mar. 1, 2008. ISBN: 0521527961 (cit. on pp. 9, 10).
- [103] S. M. Badalyan et al. "Exchange and correlation effects on plasmon dispersions and Coulomb drag in low-density electron bilayers". *Physical Review B* 75.12 (Mar. 2007) (cit. on p. 10).
- [104] K. S. Singwi et al. "Electron Correlations at Metallic Densities". *Physical Review* 176.2 (Dec. 1968), pp. 589–599 (cit. on p. 10).
- [105] Lauri J. Lantto. "Variational theory of multicomponent quantum fluids: An application to positron-electron plasmas at $T=0$ ". *Physical Review B* 36.10 (Oct. 1987), pp. 5160–5170 (cit. on p. 10).
- [106] Claudio Attacalite et al. "Correlation Energy and Spin Polarization in the 2D Electron Gas". *Physical Review Letters* 88.25 (June 2002) (cit. on p. 10).
- [107] Eric G. Lean. "III Interaction of Light and Acoustic Surface Waves". *Progress in Optics*. Elsevier, 1973, pp. 123–166 (cit. on p. 11).
- [108] Alberto Boscá et al. "Automatic graphene transfer system for improved material quality and efficiency". *Scientific Reports* 6.1 (Feb. 2016), p. 21676 (cit. on p. 12).
- [109] Dominik Metten et al. "All-optical blister test of suspended graphene using micro-Raman spectroscopy". *Physical Review Applied* 2.5 (2014), p. 054008 (cit. on p. 13).
- [110] Yuyoung Shin et al. "Raman spectroscopy of highly pressurized graphene membranes". *Applied Physics Letters* 108.22 (2016), p. 221907 (cit. on p. 13).
- [111] Ji Eun Lee et al. "Optical separation of mechanical strain from charge doping in graphene". *Nature Communications* 3 (2012), p. 1024 (cit. on p. 14).
- [112] Hua Zhong et al. "Comparison of mobility extraction methods based on field-effect measurements for graphene". *AIP Advances* 5.5 (2015), p. 057136 (cit. on p. 14).
- [113] Alberto Boscá et al. "Method for extracting relevant electrical parameters from graphene field-effect transistors using a physical model". *Journal of Applied Physics* 117.4 (2015), p. 044504 (cit. on p. 14).
- [114] Julen Tamayo-Arriola et al. "Controllable and Highly Propagative Hybrid Surface Plasmon-Phonon Polariton in a CdZnO-Based Two-Interface System". *ACS Photonics* 6.11 (2019), pp. 2816–2822 (cit. on p. 15).
- [115] R Fandan et al. "Acoustically-driven surface and hyperbolic plasmon-phonon polaritons in graphene/h-BN heterostructures on piezoelectric substrates". *Journal of Physics D: Applied Physics* 51.20 (Apr. 2018), p. 204004 (cit. on pp. 15, 64).
- [116] Mauricio M de Lima Jr and Paulo V Santos. "Modulation of photonic structures by surface acoustic waves". *Reports on progress in physics* 68.7 (2005), p. 1639 (cit. on pp. 16, 40).
- [117] Paulo V Santos. "Acoustic field mapping on GaAs using microscopic reflectance and reflectance anisotropy". *Applied physics letters* 74.26 (1999), pp. 4002–4004 (cit. on p. 17).

- [118] F. Iikawa et al. "Optical phonon modulation in semiconductors by surface acoustic waves". *Physical Review B* 93.19 (May 2016), p. 195212 (cit. on pp. 18, 20, 52).
- [119] J Pedrós et al. "Low attenuation of GHz Rayleigh-like surface acoustic waves in ZnO/GaAs systems immersed in liquid helium". *Applied Physics Letters* 102.4 (2013), p. 043507 (cit. on p. 21).
- [120] JG Rodriguez-Madrid et al. "Super-high-frequency SAW resonators on AlN/diamond". *IEEE Electron Device Letters* 33.4 (2012), pp. 495–497 (cit. on p. 21).
- [121] Derek YH Ho et al. "Theory of Coulomb Drag in Spatially Inhomogeneous Materials". *Communications Physics* 1.41 (2018), pp. 1–9 (cit. on pp. 30, 37).
- [122] Justin CW Song and Leonid S Levitov. "Energy-driven drag at charge neutrality in graphene". *Physical review letters* 109.23 (2012), p. 236602 (cit. on p. 30).
- [123] HM Gibbs, Galina Khitrova, and Stephan W Koch. "Exciton–polariton light–semiconductor coupling effects". *Nature Photonics* 5.5 (2011), p. 273 (cit. on p. 39).
- [124] Ralf Ameling and Harald Giessen. "Microcavity plasmonics: strong coupling of photonic cavities and plasmons". *Laser & photonics reviews* 7.2 (2013), pp. 141–169 (cit. on p. 39).
- [125] Ilkka Kylänpää and Hannu-Pekka Komsa. "Binding energies of exciton complexes in transition metal dichalcogenide monolayers and effect of dielectric environment". *Physical Review B* 92.20 (2015), p. 205418 (cit. on p. 39).
- [126] Guowei Zhang et al. "Determination of layer-dependent exciton binding energies in few-layer black phosphorus". *Science advances* 4.3 (2018), eaap9977 (cit. on pp. 39, 44, 45).
- [127] Adriano Violante et al. "Dynamics of indirect exciton transport by moving acoustic fields". *New Journal of Physics* 16.3 (2014), p. 033035 (cit. on p. 40).
- [128] PV Santos et al. "Band mixing and ambipolar transport by surface acoustic waves in GaAs quantum wells". *Physical Review B* 69.15 (2004), p. 155318 (cit. on p. 40).
- [129] J Rudolph, R Hey, and PV Santos. "Long-range exciton transport by dynamic strain fields in a GaAs quantum well". *Physical Review Letters* 99.4 (2007), p. 047602 (cit. on p. 40).
- [130] C Rocke et al. "Acoustically driven storage of light in a quantum well". *Physical review letters* 78.21 (1997), p. 4099 (cit. on p. 40).
- [131] Karel-Alexander N Duerloo, Mitchell T Ong, and Evan J Reed. "Intrinsic piezoelectricity in two-dimensional materials". *The Journal of Physical Chemistry Letters* 3.19 (2012), pp. 2871–2876 (cit. on p. 40).
- [132] Zuocheng Zhang et al. "Strain-modulated bandgap and piezo-resistive effect in black phosphorus field-effect transistors". *Nano letters* 17.10 (2017), pp. 6097–6103 (cit. on p. 40).
- [133] Honghua U Yang et al. "Optical dielectric function of silver". *Physical Review B* 91.23 (2015), p. 235137 (cit. on p. 40).
- [134] Zakaria M Abd El-Fattah et al. "Plasmonics in atomically thin crystalline silver films". *ACS nano* 13.7 (2019), pp. 7771–7779 (cit. on p. 40).

- [135] P. Yu and M. Cardona. *Fundamentals of Semiconductors: Physics and Materials Properties*. Graduate Texts in Physics. Springer Berlin Heidelberg, 2010. ISBN: 9783642007101 (cit. on pp. 41, 56).
- [136] C Ciuti et al. "Role of the exchange of carriers in elastic exciton-exciton scattering in quantum wells". *Physical Review B* 58.12 (1998), p. 7926 (cit. on p. 41).
- [137] S Fahy, Charles Kittel, and Steven G Louie. "Electromagnetic screening by metals". *American Journal of Physics* 56.11 (1988), pp. 989–992 (cit. on p. 41).
- [138] R Schad et al. "Metallic and nonmetallic conductivity of thin epitaxial silver films". *Physical Review B* 45.19 (1992), p. 11430 (cit. on p. 41).
- [139] Francois Marquier, Christophe Sauvan, and Jean-Jacques Greffet. "Revisiting quantum optics with surface plasmons and plasmonic resonators". *ACS photonics* 4.9 (2017), pp. 2091–2101 (cit. on p. 43).
- [140] Gleb M Akselrod et al. "Leveraging nanocavity harmonics for control of optical processes in 2D semiconductors". *Nano Letters* 15.5 (2015), pp. 3578–3584 (cit. on p. 43).
- [141] IV Kukushkin et al. "Ultrahigh-frequency surface acoustic waves for finite wave-vector spectroscopy of two-dimensional electrons". *Applied physics letters* 85.19 (2004), pp. 4526–4528 (cit. on p. 43).
- [142] S Büyükköse et al. "Ultrahigh-frequency surface acoustic wave transducers on ZnO/SiO₂/Si using nanoimprint lithography". *Nanotechnology* 23.31 (2012), p. 315303 (cit. on p. 43).
- [143] S Büyükköse et al. "Ultrahigh-frequency surface acoustic wave generation for acoustic charge transport in silicon". *Applied physics letters* 102.1 (2013), p. 013112 (cit. on p. 43).
- [144] N. Papasimakis et al. "Strain engineering in graphene by laser irradiation". *Applied Physics Letters* 106.6 (Feb. 2015), p. 061904 (cit. on p. 51).
- [145] Eddwi H. Hasdeo et al. "Breit-Wigner-Fano line shapes in Raman spectra of graphene". *Physical Review B* 90.24 (Dec. 2014), p. 245140 (cit. on pp. 52, 59).
- [146] T. Sogawa et al. "Dynamic band-structure modulation of quantum wells by surface acoustic waves". *Phys. Rev. B* 63 (12 Mar. 2001), p. 121307 (cit. on p. 52).
- [147] J. R. Gell et al. "Modulation of single quantum dot energy levels by a surface-acoustic-wave". *Applied Physics Letters* 93.8 (2008), p. 081115 (cit. on p. 52).
- [148] Jens Pustiowski et al. "Independent dynamic acousto-mechanical and electrostatic control of individual quantum dots in a LiNbO₃-GaAs hybrid". *Applied Physics Letters* 106.1 (2015), p. 013107 (cit. on p. 52).
- [149] B Villa et al. "Surface acoustic wave modulation of a coherently driven quantum dot in a pillar microcavity". *Applied Physics Letters* 111.1 (2017), p. 011103 (cit. on p. 52).
- [150] Fernando Cerdeira and Manuel Cardona. "Effect of Carrier Concentration on the Raman Frequencies of Si and Ge". *Phys. Rev. B* 5 (4 Feb. 1972), pp. 1440–1454 (cit. on p. 52).
- [151] Lord Rayleigh. "On Waves Propagated along the Plane Surface of an Elastic Solid". *Proceedings of the London Mathematical Society* s1-17.1 (Nov. 1885), pp. 4–11 (cit. on p. 53).

- [152] C. Bousige et al. "Biaxial Strain Transfer in Supported Graphene". *Nano Letters* 17.1 (2017), pp. 21–27 (cit. on pp. 54, 55).
- [153] Mingyuan Huang et al. "Probing Strain-Induced Electronic Structure Change in Graphene by Raman Spectroscopy". *Nano Letters* 10.10 (2010), pp. 4074–4079 (cit. on p. 55).
- [154] Otakar Frank et al. "Raman 2D-Band Splitting in Graphene: Theory and Experiment". *ACS Nano* 5.3 (2011), pp. 2231–2239 (cit. on p. 55).
- [155] Fei Ding et al. "Stretchable Graphene: A Close Look at Fundamental Parameters through Biaxial Straining". *Nano Letters* 10.9 (2010), pp. 3453–3458 (cit. on p. 55).
- [156] Constanze Metzger et al. "Biaxial Strain in Graphene Adhered to Shallow Depressions". *Nano Letters* 10.1 (2010), pp. 6–10 (cit. on p. 55).
- [157] Y. C. Cheng et al. "Gruneisen parameter of the Gmode of strained monolayer graphene". *Physical Review B* 83.11 (Mar. 2011), p. 115449 (cit. on p. 55).
- [158] Tao Jiang et al. "Equi-biaxial compressive strain in graphene: Gruneisen parameter and buckling ridges". *2D Materials* 6.1 (Dec. 2018), p. 015026 (cit. on p. 55).
- [159] Harald Steiner et al. "The non-linear thermal behavior of SU-8". *2012 35th International Spring Seminar on Electronics Technology*. IEEE, May 2012, pp. 450–454 (cit. on p. 56).
- [160] J.E. Mark. *Polymer Data Handbook*. Oxford University Press, 2009. ISBN: 9780195181012 (cit. on p. 56).
- [161] Y. S. Kim and R. T. Smith. "Thermal Expansion of Lithium Tantalate and Lithium Niobate Single Crystals". *Journal of Applied Physics* 40.11 (Oct. 1969), pp. 4637–4641 (cit. on p. 56).
- [162] J. Pedrós et al. "Guided propagation of surface acoustic waves and piezoelectric field enhancement in ZnO/GaAs systems". *Journal of Applied Physics* 110.10 (2011), p. 103501 (cit. on p. 56).
- [163] G. William Burg et al. "Strongly Enhanced Tunneling at Total Charge Neutrality in Double-Bilayer Graphene- WSe₂ Heterostructures". *Physical Review Letters* 120.17 (Apr. 2018) (cit. on p. 58).
- [164] M. Zarenia et al. "Multiband Mechanism for the Sign Reversal of Coulomb Drag Observed in Double Bilayer Graphene Heterostructures". *Physical Review Letters* 121.3 (July 2018) (cit. on p. 58).
- [165] J. I. A. Li et al. "Negative Coulomb Drag in Double Bilayer Graphene". *Physical Review Letters* 117 (4 2016), p. 046802 (cit. on p. 58).
- [166] J. Hu et al. "Coulomb drag and counterflow Seebeck coefficient in bilayer-graphene double layers". *Nano Energy* 40 (2017), pp. 42–48 (cit. on p. 58).
- [167] S. Saberi-Pouya et al. "Coulomb drag in anisotropic systems: a theoretical study on a double-layer phosphorene". *Journal of Physics: Condensed Matter* 28 (2016), p. 285301 (cit. on p. 58).
- [168] Edo van Veen et al. "Tuning Two-Dimensional Hyperbolic Plasmons in Black Phosphorus". *Phys. Rev. Applied* 12 (1 July 2019), p. 014011 (cit. on p. 58).
- [169] M. N. Gjerding et al. "Layered van der Waals crystals with hyperbolic light dispersion". *Nature Communications* 8 (2017), p. 320 (cit. on p. 58).

- [170] Yanpeng Liu et al. "Tailoring sample-wide pseudo-magnetic fields on a graphene-black phosphorus heterostructure". *Nature Nanotechnology* 13 (2018), pp. 828–834 (cit. on p. 58).
- [171] J. J. Campbell and W. R. Jones. "A method for estimating optimal crystal cuts and propagation directions for excitation of piezoelectric surface waves". *IEEE Transactions on Sonics and Ultrasonics* 15.4 (1968), pp. 209–217 (cit. on p. 59).
- [172] M.A.H. Vozmediano, M.I. Katsnelson, and F. Guinea. "Gauge fields in graphene". *Physics Reports* 496.4 (2010), pp. 109–148 (cit. on p. 59).
- [173] MM de Lima Jr et al. "Focusing of surface-acoustic-wave fields on (100) GaAs surfaces". *Journal of applied physics* 94.12 (2003), pp. 7848–7855 (cit. on p. 59).
- [174] B. Amorim et al. "Novel effects of strains in graphene and other two dimensional materials". *Physics Reports* 617 (2016), pp. 1–54 (cit. on p. 59).
- [175] Gerardo G Naumis et al. "Electronic and optical properties of strained graphene and other strained 2D materials: a review". *Reports on Progress in Physics* 80.9 (Aug. 2017), p. 096501 (cit. on p. 59).
- [176] Chaojie Cui et al. "Two-dimensional materials with piezoelectric and ferroelectric functionalities". *npj 2D Materials and Applications* 2 (2018), p. 18 (cit. on p. 59).
- [177] Tony Low et al. "Novel Midinfrared Plasmonic Properties of Bilayer Graphene". *Physical Review Letters* 112 (11 2014), p. 116801 (cit. on p. 59).
- [178] Brandon C. Marin et al. "SERS-enhanced piezoplasmonic graphene composite for biological and structural strain mapping". *Nanoscale* 9 (3 2017), pp. 1292–1298 (cit. on p. 59).
- [179] Eric W. Weisstein. *Spence's Function*. URL: <https://mathworld.wolfram.com/SpencesFunction.html> (cit. on p. 63).
- [180] Rajveer Fandan et al. "Effect of quasiparticle excitations and exchange-correlation in Coulomb drag in graphene". *Communications Physics* 2.1 (2019), pp. 1–9 (cit. on p. 64).
- [181] Rajveer Fandan et al. "Dynamic local strain in graphene generated by surface acoustic waves". *Nano Letters* 20.1 (2020), pp. 402–409 (cit. on p. 67).