## Ph.D. THESIS

# Graphene-based heterostructures under pressure

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### List of abbreviations

**2D** two-dimensional **2DEG** two-dimensional electron gas **AFM** atomic force microscopy **BLG** bilayer graphene **BM** Bistritzer-MacDonald **BZ** Brillouin zone **CNP** charge neutrality point **CVD** chemical vapor deposition **DOS** density of states **EBL** Electron beam lithography **hBN** hexagonal boron nitride LL Landau level mBZ moiré-Brillouin zone **NNH** nearest neighbor hopping PC polycarbonate **PDMS** polydimethylsiloxane **RIP** remote interfacial phonon SdH Shubnikov-de Haas SLG single-layer graphene **SOC** spin-orbit coupling **TBG** twisted bilayer graphene **TDBG** twisted double bilayer graphene **TMD** transition metal dichalcogenide **TMF** transverse magnetic focusing **TO** transverse optical **TRS** time-reversal symmetry  $\mathbf{vdW}$  van der Waals **VRH** variable-range hopping WAL weak antilocalization WL weak localization

### 1. Chapter

### Introduction

The continuous development of our technology always needs novel materials with the desired properties. With the gate all around field effect transistors, the conventional semiconducting industry in a few years is reaching its limit by reaching the  $0.5 \,\mathrm{nm}$  node[1]. To go beyond it, a fundamental change is necessary to be able to engineer the material on the level of atomic precision in the next generation of microelectronic devices [2]. An alternative approach became available with the discovery of two-dimensional (2D) materials [3] after the first isolation of graphene [4]. Among the 2D materials, besides graphene, which is a semimetal, there are insulators like hexagonal boron nitride (hBN), semiconductors like transition metal dichalcogenides (TMDs), topological insulators and so on[5, 6]. The field of 2D materials since then became one of the most studied field in condensed matter physics. There have been various promising attempts to integrate the 2D crystals into the silicon-based technology making this field very attractive to industry besides fundamental research [7]. Besides potentially taking the place of silicon in conventional devices, with 2D materials, completely new electrical elements could be realized, like reconfigurable logic and logic-in-memory, and nanosensors [8, 9].

What makes 2D crystals unique compared to conventional materials is that they can be placed on top of each other layer by layer with atomic precision in the vertical direction, realizing van der Waals (vdW) heterostructures[5, 10]. Their name comes from the fact that the bonding force between the layers are vdW forces and they are weakly bonded compared to covalently bonded crystals. The properties of these heterostructures are determined by the nature of the constituting materials and the interlayer interactions[11, 12]. For example, hBN can be used as protective layers by encapsulating the graphene within hBN layers to protect it from the environment and improve the charge carrier mobility in it[13, 14]. In hBN/graphene heterostructures, the lattice mismatch may lead to the formation of a moiré pattern, which depends on the relative orientation of the crystals, the twist angle. A superlattice forms, where the moiré pattern acts as a periodic potential[15]. At small twist angles, as the lattice mismatch is small between the hBN and graphene, the electronic properties at low energies are modified, leading to interesting phenomena such as Hofstadter's butterfly[16, 17]. The twisting is a unique degree of freedom of vdW heterostructures, unlike conventional heterostructures. Twisting can fundamentally change the properties of a heterostructure, leading to new properties such as correlated phases and superconductivity in twisted bilayer graphene[18, 19]. The field of twistronics, which focuses on the effect of twisting is dynamically growing by including newer and newer systems such as twisted TMD heterostructures[20].

Besides twistronics, vdW materials are also exciting for the field of spintronics. Spintronics focuses on using the spin degree of freedom of the charge carriers to realize electronic devices, which are controlled with the manipulation of electron spins instead of the charge[21]. The manipulation is usually done by a magnetic field or with the help of spin-orbit coupling (SOC). Most of the spintronic devices are based on spin valves, which are heterostructures made of magnetic layers with a nonmagnetic layer between them. For instance, an insulator in a tunneling magnetoresistance device which is a ferromagnet/insulator/ferromagnet heterostructure can be used as a memory element in hard drives. Moreover, a non-magnetic metal is used in spin-transfer torque magnetic random-access memories, where the spin-polarized current is used to control the memory state [22]. These heterostructures can also be realized with 2D materials, where the graphene usually serves as a transport channel due to its excellent properties like negligible SOC and the near-absence of nuclear spins [23, 24]. Moreover, combining graphene with TMDs induces SOC in graphene with proximity effects [25], and the spin current in these heterostructures could be controlled with electrical gating [26-29] and with spin-to-charge conversion [30–33] bringing the 2D materials closer for industrial applications.

In vdW heterostructures, as the interlayer interactions play a crucial role, their properties could be altered by tuning the strength of these interactions[12]. Varying the interlayer distance could significantly change the strength of interlayer interactions, which could be done in practice by pressing the heterostructure[34], which can lead to structural and electronic transitions, such as driving a material from a metallic to a superconducting state[35–38]. Realizing novel phases with pressure could lead to new applications and are very important in fundamental research. Therefore, the aim of this thesis is to investigate the effect of pressure on vdW heterostructures using electrical transport techniques.

In this thesis, I focus on the effect of pressure on graphene-based heterostructures. The outline of this thesis is the following. At the beginning of the thesis, in **Chapter 2**, I introduce graphene and describe its most important properties for the experiments presented in the main part. I give a brief introduction to the band structure of twisted bilayer graphene and the role of SOC in graphene-based heterostructures. Apart from these, a basic introduction to the theory of transport and magnetotransport is also given, which are used to study the devices in the main part of this thesis. In Chapter **3**, the most important experimental methods for the main part are presented focusing on the sample fabrication and the low-temperature transport measurements along with the method of applying the pressure on nanodevices. The main part of this thesis consists of 4 parts. In **Chapter 4**, the modeling of TMD and bilayer graphene (BLG) based heterostructures are presented including the calculation of Landau levels in them along with the calculation of the Fermi surfaces. In the second part of the Chapter, the calculation of the pressure dependence of the band structure of twisted double bilayer graphene is presented. In Chapter 5, the electronic properties of twisted double bilayer graphene around the magic angle are investigated under pressure. The transport measurements focus on determining how the pressure affects the single-particle moiré gaps, the correlated phases and the twist angle. The findings are compared with the model described in Chapter 4. In Chapter 6, the change of the proximity-induced spin-orbit coupling by applying hydrostatic pressure is investigated in WSe<sub>2</sub> and BLGbased heterostructures with transport measurements, focusing on the change of the SOC strength with pressure, which are determined using the calculations presented in Chapter 4. In Chapter 7, a transport study of high-mobility devices made of singlelayer graphene (SLG), which is encapsulated within hBN crystals is presented, focusing on the evolution of transport properties with pressure. In **Chapter 8**, a brief summary is given, which is followed by the thesis points, the list of publications and the appendix. The appendix includes fabrication recipes, details of the derivation of the Landau levels in bilayer graphene, calculation of surface phonon modes, and further measurements of BLG/WSe<sub>2</sub> heterostructures and high-mobility SLG devices. At the end of this thesis, the list of references from the literature is given.

### 2. Chapter

### Theoretical background

In this chapter, the theoretical background of the studied materials and effects are summarized. Firstly, single-layer graphene (SLG) and bilayer graphene (BLG) are described, focusing on their electrical structure. Secondly, the role of the twist angle between graphene layers is discussed and the Bistritzer – MacDonald model is derived for twisted double bilayer graphene. Thirdly, the role of the spin-orbit coupling (SOC) is presented. Afterward, the transport properties of graphene are described at finite temperatures using the Boltzmann transport equation. Finally, the magneto-transport in graphene is discussed.

# 2.1 Electronic properties of graphene and bilayer graphene



Figure 2.1: (a) Crystal structure of single-layer graphene. A and B atoms indicate the two sublattices and are shown with blue and red circles.  $\mathbf{a_1}$  and  $\mathbf{a_2}$  are the lattice vectors and a is the lattice constant. (b) Reciprocal space of SLG and BLG in  $\mathbf{b_1}$  and  $\mathbf{b_2}$  reciprocal basis vectors. The hexagon is the first Brillouin zone (BZ) of graphene with  $\Gamma$  at the center and K and K' are two non-equivalent corners.

Graphene is a single atomic sheet of graphite. It was studied experimentally for the first time in 2004[4]. Since then, it has become one of the most studied materials in condensed matter physics, and other 2D crystals have also been discovered. In graphene,

the Carbon atoms sit on a triangular lattice with two-atom unit cell with a lattice constant of  $a = 2.46 \text{\AA}[39]$  as illustrated in Fig. 2.1a. The basis vectors are given by

$$\mathbf{a_1} = \begin{pmatrix} \frac{a}{2} \\ \frac{\sqrt{3a}}{2} \end{pmatrix}, \qquad \mathbf{a_2} = \begin{pmatrix} \frac{a}{2} \\ -\frac{\sqrt{3a}}{2} \end{pmatrix}.$$
(2.1)

The Brillouin zone is a hexagon and the reciprocal basis vectors are given by

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

The A and B sublattice positions are given by  $\tau_{\rm A} = 0$  and  $\tau_{\rm B} = (\mathbf{a_1} - \mathbf{a_2})/3$ . In graphite, the perpendicular lattice constant is d = 3.35Å[40].

#### 2.1.1 Single-layer graphene



Figure 2.2: (a) Dispersion relation of SLG. The valence and conduction bands touch each other at the Dirac points. (b) Zoom in near the K-point. The dispersion is linear in the low-energy regime.

The band structure of graphene is symmetric around the  $\Gamma$  point. The valence and conduction bands are touching at the K and K' points, which are called Dirac points as shown in Fig. 2.2a[41]. Around them, the bands have linear dispersion as depicted in Fig. 2.2b. The Hamiltonian describing the bands around the K points are given by

$$H_{\rm SLG} = \hbar v_{\rm F} \left( \xi \sigma_x k_x + \sigma_y k_y \right), \tag{2.3}$$

where  $\hbar$  is the reduced Planck's constant,  $\mathbf{k} = (k_x, k_y)$  is measured from the K(K')point,  $v_{\rm F}$  is the Fermi velocity of graphene and it is related to the  $\gamma_0$  hopping<sup>1</sup> by

 $<sup>^{1}\</sup>gamma_{0} \approx 3 \,\mathrm{eV}$  is the nearest neighbor hopping integral between sublattice A and B[39].

 $v_{\rm F} = \sqrt{3}a\gamma_0/2\hbar$ . Here,  $\xi$  is the valley index  $\xi = 1(-1)$  for K(K'),  $\sigma_i$  are the pseudospin Pauli matrices acting on sublattices (A and B atoms in Fig. 2.1) of graphene. The electronic states are described by

$$H_{\rm SLG}\Psi(\mathbf{r}) = E\Psi(\mathbf{r}),\tag{2.4}$$

where the wave function is written as

$$\Psi_{s\mathbf{k}}(\mathbf{r}) = \frac{1}{\sqrt{A}} F_s(\mathbf{k}) e^{i\mathbf{k}\mathbf{r}},\tag{2.5}$$

where A is the area of the system [42–45]. Introducing  $k = |\mathbf{k}|$  and the angle  $\theta_{\mathbf{k}}$  in the momentum space as  $k_x = k \cos(\theta_{\mathbf{k}})$  and  $k_y = k \sin(\theta_{\mathbf{k}})$ , the eigen energies of Eq.(2.4) are given by  $E(k) = s\hbar v_{\mathrm{F}}|\mathbf{k}|$  around the K and K' points in the Brillouin zone with s = 1 for the conduction and s = -1 for the valence bands, whereas  $F_s(\mathbf{k})$  is given by

$$F_s(\mathbf{k}) = \frac{1}{\sqrt{2}} \begin{pmatrix} e^{-i\theta_{\mathbf{k}}} \\ s \end{pmatrix}.$$
 (2.6)

The density of states (DOS) g(E) for graphene is given by

$$g(E) = \frac{g}{4\pi^2} 2\pi k \left| \frac{dE(k)}{dk} \right|^{-1} = \frac{2E}{\hbar^2 v_{\rm F}^2 \pi},$$
(2.7)

where g is the degeneracy factor, g = 4 for graphene (spin and valley degeneracy). The charge carrier density n relative to the charge neutral case, i.e. a filled valence band, in the zero temperature limit is defined by

$$n = \int_{0}^{E_{\rm F}} g(E) dE = \frac{E_{\rm F}^2}{\hbar^2 v_{\rm F}^2 \pi} = \frac{k_F^2}{\pi},$$
(2.8)

where  $E_{\rm F}$  is the Fermi energy relative to the Dirac points and  $k_F$  is the Fermi wave vector. The effective mass  $m^*$  of SLG due to its linear dispersion depend on  $k_F[46-48]$ as

$$m^*(k_F) = \hbar k_F / v_F. \tag{2.9}$$

#### 2.1.2 Bilayer graphene

BLG consists of two layers of graphene with a distance of d between them. In the common, Bernal-stacked configuration, the B atom in the bottom layer (B1) is below the A atom in the top layer (A2) as shown in Fig. 2.3. Unlike SLG, BLG has parabolic band dispersion with a slight modification by the remote hopping terms ( $\gamma_3$  and  $\gamma_4$  in



Figure 2.3: Crystal structure of bilayer graphene. On the bottom layers A1 and B1 atoms are shown with blue and red circles and the A2 and B2 atoms on the top layer are shown with blue and grey circles. The  $\gamma_i$  hopping parameters are also shown with lines between the atoms.

Fig. 2.3) at the low-energy regime and touching around the Dirac points as shown in Fig. 2.4b. The effective low-energy Hamiltonian of pristine BLG near Dirac points can be written in the  $|A1, B1, A2, B2\rangle$  basis[45, 49] as

$$H_{\rm BLG} = \begin{pmatrix} \frac{u}{2} & v_0 \pi^{\dagger} & -v_4 \pi^{\dagger} & v_3 \pi \\ v_0 \pi & \frac{u}{2} + \Delta' & \gamma_1 & -v_4 \pi^{\dagger} \\ -v_4 \pi & \gamma_1 & -\frac{u}{2} + \Delta' & v_0 \pi^{\dagger} \\ v_3 \pi^{\dagger} & -v_4 \pi & v_0 \pi & -\frac{u}{2} \end{pmatrix},$$
(2.10)

where  $\pi = \hbar(\xi k_x + ik_y)$  and  $v_i = \frac{\sqrt{3}a}{2\hbar}\gamma_i$ .  $\gamma_i$  are the hopping parameters[45]:  $\gamma_0 = 2.61 \text{ eV}$ is the intralayer nearest neighbor hopping,  $\gamma_1 = 0.361 \text{ eV}$  is the interlayer coupling between orbitals on the dimer sites B1 and A2, and  $\gamma_4 = 0.138 \text{ eV}$  is the interlayer coupling between dimer and non-dimer orbitals A1 and A2 or B1 and B2. The parameter  $\Delta' = 0.015 \text{ eV}$  describes the on-site energy difference between dimer and non-dimer sites and  $\xi$  is the valley index and u is the interlayer potential difference.

The interlayer potential difference can be related to an external displacement field D by

$$u = \frac{ed}{\epsilon_0 \epsilon_{\rm BLG}^{\perp}} D, \qquad (2.11)$$

where e is the elementary charge,  $\epsilon_0$  is the vacuum permittivity, d = 0.33 nm is the



Figure 2.4: Band structure of bilayer graphene using the hopping parameters given in the main text (a) at u = -10 meV, (b) at u = 0 and (c) at u = 10 meV near the *K*-point. The color of the bands shows the layer-polarization with the color bar plotted in panel (b).

BLG interlayer separation,  $\epsilon_{\text{BLG}}^{\perp} = 4.3$  is the out-of-plane dielectric constant of BLG [50].  $\gamma_3 = 0.1$  eV is the interlayer coupling of A1 and B2, which is responsible for the trigonal warping. The effect of u is illustrated in Fig. 2.4a. It opens a gap at the charge neutrality point (CNP) and layer polarizes the bands. With positive (negative) u the valence band electrons are localized on the bottom (top) layer and the conduction band electrons are localized on the top (bottom) layer[51].

#### 2.2 Twisted graphene structures

Unlike conventional semiconductor heterostructures, van der Waals (vdW) heterostructures are made from 2D materials and the bonding force between the layers are vdW forces[5]. A specialty of these heterostructures is that their properties depend also on the relative orientation of the 2D constituent crystals[52–58]. The angle between their relative crystal orientation is called twist angle  $\vartheta$ . Experiments showed many interesting phenomena such as Hofstadter butterfly[16, 17, 59] and fractional Bloch band quantum Hall effect[14, 60] in aligned hBN/graphene heterostructures[61]. A huge impact on the field of twistronics was the discovery of superconductivity[19, 62] and correlated phases[18, 63–66] in twisted bilayer graphene (TBG) near a so-called magic angle. At the magic angle, the bands closest to the Fermi energy become completely flat[53], and this phenomenon can be described with a continuum model called the Bistritzer-MacDonald (BM) model. This model is discussed later in Section 2.2.2.

#### 2.2.1 Twisted graphene lattice

Stacking two-dimensional crystals with the same lattice structure with small relative orientation angle and similar lattice constants leads to the formation of Moiré pattern. The rotation of the lattice can be simply done by modifying the basis vectors with the rotation matrix  $\mathbf{R}(\vartheta)$  defined as

$$\mathbf{R}(\vartheta) = \begin{pmatrix} \cos(\vartheta) & -\sin(\vartheta) \\ \sin(\vartheta) & \cos(\vartheta) \end{pmatrix}, \qquad (2.12)$$

where  $\vartheta$  is the angle of rotation. In the following, the case of two honeycomb lattices is discussed. Then Eq.(2.1) is modified to

$$\mathbf{a_1} = \mathbf{R}(\vartheta) \begin{pmatrix} \frac{a}{2} \\ \frac{\sqrt{3a}}{2} \end{pmatrix}, \quad \mathbf{a_2} = \mathbf{R}(\vartheta) \begin{pmatrix} \frac{a}{2} \\ -\frac{\sqrt{3a}}{2} \end{pmatrix}.$$
 (2.13)

The corresponding primitive reciprocal lattice vectors in Eq.(2.2) are modified to

$$\mathbf{b_1} = \left(\frac{2\pi}{a}, \frac{2\pi}{\sqrt{3}a}\right) \mathbf{R}^T(\vartheta) = \frac{2\pi}{a} \left(\cos(\vartheta) - \frac{\sin(\vartheta)}{\sqrt{3}}, \sin(\vartheta) + \frac{\cos(\vartheta)}{\sqrt{3}}\right), \quad (2.14)$$
$$\mathbf{b_2} = \left(\frac{2\pi}{a}, -\frac{2\pi}{\sqrt{3}a}\right) \mathbf{R}^T(\vartheta) = \frac{2\pi}{a} \left(\cos(\vartheta) + \frac{\sin(\vartheta)}{\sqrt{3}}, \sin(\vartheta) - \frac{\cos(\vartheta)}{\sqrt{3}}\right).$$

The coordinate system is chosen such that the basis vectors of the two honeycomb lattices are described with Eq.(2.13) with the rotation of  $\vartheta/2$  for the top and  $-\vartheta/2$  for the bottom lattice:

$$\mathbf{a_1^t} = \mathbf{R}(\vartheta/2) \begin{pmatrix} \frac{a_t}{2} \\ \frac{\sqrt{3}a_t}{2} \end{pmatrix}, \qquad \mathbf{a_2^t} = \mathbf{R}(\vartheta/2) \begin{pmatrix} \frac{a_t}{2} \\ -\frac{\sqrt{3}a_t}{2} \end{pmatrix},$$
$$\mathbf{a_1^b} = \mathbf{R}(-\vartheta/2) \begin{pmatrix} \frac{a_b}{2} \\ \frac{\sqrt{3}a_b}{2} \end{pmatrix}, \qquad \mathbf{a_2^b} = \mathbf{R}(-\vartheta/2) \begin{pmatrix} \frac{a_b}{2} \\ -\frac{\sqrt{3}a_b}{2} \end{pmatrix},$$

where  $\mathbf{a}_{1,2}^{t}$  are the basis vectors of the top layer with  $a_t$  lattice constant and  $\mathbf{a}_{1,2}^{b}$  are the basis vectors of the bottom layer with  $a_b$  lattice constant. The lattice constant of the bottom layer can always be written as  $a_b = a_t(1 + \delta)$ , where  $\delta$  describes the lattice mismatch.

The Moiré pattern, which is illustrated in Fig. 2.5, can be regarded as a beating of the two lattices [67, 68] similarly to the case of sound waves. Reciprocal lattice vectors can be constructed to this superlattice as the difference of the constituting reciprocal

lattice vectors. Here, they are defined as

$$\mathbf{b_1^{(s)}} = \mathbf{b_1^b} - \mathbf{b_1^t},$$

$$\mathbf{b_2^{(s)}} = \mathbf{b_2^t} - \mathbf{b_2^b},$$
(2.15)

where  $\mathbf{b_i^{t/b}}$  are the original reciprocal lattice vectors of the top and bottom layers which are given by Eq.(2.14) as depicted in Fig. 2.5a. These vectors form a triangular lattice in the reciprocal space and its Wigner-Seitz cell is called the moiré-Brillouin zone (mBZ). If the lattice constants of the two layers are the same (like TBG), then Eq.(2.15) can be written as

$$\mathbf{b_1^{(s)}} = \frac{2\pi}{a} \cdot 2\sin(\vartheta/2) \cdot \left(\frac{1}{\sqrt{3}}, -1\right), \qquad (2.16)$$
$$\mathbf{b_2^{(s)}} = \frac{2\pi}{a} \cdot 2\sin(\vartheta/2) \cdot \left(\frac{1}{\sqrt{3}}, 1\right).$$

From Eq.(2.16) the basis vectors can be calculated with the relation of  $\mathbf{a_i b_j} = 2\pi \delta_{ij}$ . The length of the superlattice basis vectors defines the Moiré-wavelength as  $a^{(s)} = \lambda = \frac{a}{2|\sin(\vartheta/2)|}$  for  $\delta = 0$ . The area of the Moiré-unit cell is given by

$$A_{\rm m} = \frac{\sqrt{3}}{2} \left(\frac{a}{2\sin(\vartheta/2)}\right)^2. \tag{2.17}$$

The maximum electron density per Bloch band is given by  $1/A_{\rm m}$ . In the case of graphene, these bands are 4-fold degenerate due to the spin and valley degeneracy, thus a quantity of  $n_{\rm s} = 4/A_{\rm m}$  can be introduced, which is the charge carrier density of these degenerate bands.

#### 2.2.2 Bistritzer-MacDonald model

The moiré pattern leads to a rearrangement of the band structure in the mBZ and moiré Bloch bands form in the electronic structure of twisted materials. The Bistritzer-MacDonald (BM) model describes these electronic states with a low-energy continuum Hamiltonian, which consists of the Hamiltonians of the isolated layers and tunneling terms, which describes the hopping between the layers[69]. The model Hamiltonian and its derivation are shown in Section 2.2.3.

The result of the Bistritzer-MacDonald model for twisted bilayer graphene, which consists of two layers of SLG, is plotted for a few angles in Fig. 2.6 in the mBZ. At large angles, the layers are decoupled, and the spectrum resembles the band structure of SLG as shown in Fig. 2.6a at  $\vartheta = 5^{\circ}$ . The bands of the SLG layers are folded into the mBZ, and the valance and conduction bands are touching at the  $K_M$  and  $K'_M$  points of the



Figure 2.5: (a) Brillouin-zone (BZ) of the top (red) and bottom (blue) graphene with their basis vectors. The black honeycomb is the Moiré-BZ with its corresponding reciprocal basis vectors of  $b_i^{(s)}$ . (b) Moiré-pattern formed from two honeycomb lattices.  $\mathbf{a}_1^{(s)}$  and  $\mathbf{a}_2^{(s)}$  are the lattice vectors of the formed superlattice.



Figure 2.6: Calculated band structure of twisted bilayer graphene using the BM model plotted in the mBZ using the hopping parameters given in the main text with  $\gamma = 3.1 \text{ eV}$  and w = 110.7 meV at (a)  $\vartheta = 5^{\circ}$ , (b)  $\vartheta = 1.05^{\circ}$  and (c)  $\vartheta = 0.5^{\circ}$ .

mBZ. As the twist angle is decreased, the interlayer interaction strength is increasing leading to hybridization of the layers. At the magic angle  $\vartheta_m \approx 1.05^\circ$  (Fig. 2.6b), the interlayer coupling leads to completely flat bands near the Fermi level. At even smaller angles, the layers are strongly coupled leading to narrow, but not completely flat bands near and away from the Fermi level as shown in Fig. 2.6c.

#### 2.2.3 Derivation of the BM model

The effect of twisting on the band structure can be described with a continuum model[69] described in the following. Assume that, the crystal states  $|\Psi_{n,\mathbf{k}}\rangle$  of twisted structures can be expanded as a linear combination of the Bloch-states  $|\Phi_{s\mathbf{k}}^{(l)}\rangle$  of the layers as

$$|\Psi_{n,\mathbf{k}}\rangle = \sum_{l} \sum_{s} c_{s\mathbf{k}}^{(l)} |\Phi_{s\mathbf{k}}^{(l)}\rangle, \qquad (2.18)$$

where l indexes the layers and s is the site index<sup>2</sup> in layer l and  $\mathbf{k}$  is the wave vector in the reciprocal space. The Bloch states are defined as

$$|ls\mathbf{k}\rangle = |\Phi_{s\mathbf{k}}^{(l)}\rangle = \frac{1}{\sqrt{N}} \sum_{\mathbf{R}_{n}^{(l)}} e^{i\mathbf{k}(\mathbf{R}_{n}^{(l)} + \tau_{s})} |\mathbf{R}_{n}^{(l)} - \tau_{s}\rangle, \qquad (2.19)$$

where  $|\mathbf{R}_{n}^{(l)} - \tau_{s}\rangle = |ls\mathbf{R}_{n}\rangle$  are the Wannier-states<sup>3</sup> of atom s with atomic positions of  $\tau_{s}$  in layer l on site  $\mathbf{R}_{n}$  and N is the number of unit cells in layer l. In general, the system can be described with a Hamiltonian (H) consisting of a kinetic and a potential term, which includes lattice periodic potentials. H acts on the crystal states of  $|\Psi_{n,\mathbf{k}}\rangle$  as

$$\sum_{\mathbf{k}'} H_{\mathbf{k}\mathbf{k}'} |\Psi_{n,\mathbf{k}'}\rangle = E_n(\mathbf{k}) |\Psi_{n,\mathbf{k}}\rangle, \qquad (2.20)$$

where  $E_n(\mathbf{k})$  are the eigenenergies of the system of the *n*th band. The Hamiltonian is non-diagonal in  $\mathbf{k}$  as shown later unlike the untwisted case like Eq.(2.10) for Bernalstacked BLG.

To calculate the band structure of twisted systems, Eq.(2.20) needs to be solved. For this purpose,  $H_{\mathbf{k}\mathbf{k}'}$  must be diagonalized in  $\mathbf{k}$  space. To do this, the lattice vectors of the bottom layer are chosen as

$$\mathbf{R}_n = n_1 \mathbf{a}_1 + n_2 \mathbf{a}_2. \tag{2.21}$$

The top layer is rotated with  $\vartheta$  counter-clockwise and its Bravais lattice is described with

$$\mathbf{R}'_m = R(\vartheta)(n_1\mathbf{a}_1 + n_2\mathbf{a}_2) + \mathbf{d}, \qquad (2.22)$$

where **d** is the shift between the two lattices. To solve (2.20), the matrix element between the Bloch-states defined in Eq.(2.19) can be constructed as

$$\langle ls\mathbf{k}|H|l's'\mathbf{k}'\rangle = \frac{1}{\sqrt{NN'}} \sum_{\mathbf{R}_n,\mathbf{R}'_m} \langle ls\mathbf{R}_n|H|l's'\mathbf{R}'_m\rangle e^{-i\mathbf{k}(\mathbf{R}_n+\tau_s)} e^{i\mathbf{k}'(\mathbf{R}'_m+\tau_{s'})}, \qquad (2.23)$$

<sup>&</sup>lt;sup>2</sup>In case of SLG s = 1 for sublattice A and s = 2 for sublattice B atoms.

<sup>&</sup>lt;sup>3</sup>Atomic orbitals of atom s in layer l on site  $\mathbf{R}_n$  in linear combination of atomic orbitals (LCAO) method.

where  $\langle ls \mathbf{R}_n | H | l's' \mathbf{R}'_m \rangle$  is the hopping amplitude that depends on the local coordination of the layers l and l' and the sites within the 2D crystal unit cell labeled with s.

The l = l' terms are the intralayer matrix elements. In this case,  $\mathbf{R}_n$  and  $\mathbf{R}_m$  are on the same layer. In two-center approximation,  $\langle ls \mathbf{R}_n | H | ls' \mathbf{R}'_m \rangle$  depend only on the difference between atomic positions<sup>4</sup> and by introducing  $\mathbf{R} = \mathbf{R}_n - \mathbf{R}'_m$ , Eq.(2.23) can be rewritten as

$$\langle ls\mathbf{k}|H|ls'\mathbf{k}'\rangle = \frac{1}{N}\sum_{\mathbf{R}_n} e^{-i(\mathbf{k}-\mathbf{k}')\mathbf{R}_n} \sum_{\mathbf{R}} e^{-i\mathbf{k}'\mathbf{R}} \langle ls\mathbf{R}|H|ls'\mathbf{0}\rangle e^{-i\mathbf{k}\tau_s + i\mathbf{k}'\tau_{s'}}.$$
 (2.24)

The summation of  $\mathbf{R}_n$  can be carried using the orthogonality relations of

$$\sum_{\mathbf{R}_n} e^{i(\mathbf{k}-\mathbf{k}')\mathbf{R}_n} = N \sum_{\mathbf{G}_p} \delta_{\mathbf{k}-\mathbf{k}',\mathbf{G}_p}, \qquad (2.25)$$

where  $\mathbf{G}_p = p_1 \mathbf{b}_1 + p_2 \mathbf{b}_2$  are reciprocal lattice vectors, where  $p_i$  are integers and  $\delta_{i,j}$  is the Kronecker delta function. Therefore,

$$\langle ls\mathbf{k}|H|ls'\mathbf{k}'\rangle = \sum_{\mathbf{G}_p} \delta_{\mathbf{k}-\mathbf{k}',\mathbf{G}_p} \sum_{\mathbf{R}} e^{-i\mathbf{k}'\mathbf{R}} \langle ls\mathbf{R}|H|ls'\mathbf{0}\rangle e^{-i\mathbf{k}'(\tau_s-\tau_{s'})-i\mathbf{G}_{\mathbf{p}}\tau_s} = (2.26)$$

$$= \sum_{\mathbf{G}_p} \delta_{\mathbf{k}-\mathbf{k}',\mathbf{G}_p} H_{ls,ls'}(\mathbf{k}+\mathbf{G}_p), \qquad (2.27)$$

where, as we are only interested in the low-energy regime, we can restrict  $\mathbf{k}$  and  $\mathbf{k}'$  to be in the first BZ, i.e.  $\mathbf{G}_p=0$ . The intralayer hopping terms results

$$\langle ls\mathbf{k}|H|ls'\mathbf{k}'\rangle = \delta_{\mathbf{k},\mathbf{k}'}H_{ls,ls'}(\mathbf{k}), \qquad (2.28)$$

where  $H_{ls,ls'}(\mathbf{k})$  is the Hamiltonian of a separate layer. For example, for SLG in nearestneighbor approximation,  $H_{ls,ls'}(\mathbf{k})$  is given by Eq.(2.3) around the **K** and **K'** point in the low-energy regime.

In two-center approximation as  $\langle ls \mathbf{R}_n | H | l's' \mathbf{R}'_m \rangle$  depends only on the difference of the atomic positions as  $(\mathbf{R}_n + \tau_s - \mathbf{R}'_m - \tau_{s'})$ , its Fourier transform can be written as

$$\langle ls\mathbf{R}_n|H|l's'\mathbf{R}_m'\rangle = \frac{1}{N}\sum_{\mathbf{q}} e^{i\mathbf{q}(\mathbf{R}_n + \tau_{\mathbf{s}})} H_{ls,l's'}(\mathbf{q}) e^{-i\mathbf{q}(\mathbf{R}_m' + \tau_{\mathbf{s}}')}, \qquad (2.29)$$

where  $H_{ls,l's'}(\mathbf{q})$  is the Wannier representation Bloch-band Hamiltonian and it is a nontrivial function of the displacement of the top layer with respect to the bottom layer. Inserting Eq.(2.29) in Eq.(2.23) and using the the orthogonality relations of Eq.(2.25),

<sup>&</sup>lt;sup>4</sup>In LCAO method, the atomic potentials  $U(\mathbf{r})$  are lattice periodic and this approximation is exact.

 $\langle ls {\bf k} | H | l's' {\bf k}' \rangle$  can be written as

$$\langle ls\mathbf{k}|H|l's'\mathbf{k}'\rangle = \sum_{\mathbf{G}_p,\mathbf{G}_q'} e^{i\mathbf{G}_p\tau_s} e^{-i\mathbf{G}_q'\tau_{s'}} H_{ls,l's'}(\mathbf{k}+\mathbf{G}_p)\delta_{\mathbf{k}+\mathbf{G}_p,\mathbf{k}'+\mathbf{G}_q'}.$$
 (2.30)

In this expression, the Moiré pattern is hidden in the Kronecker delta. To make it more apparent,  $\delta_{\mathbf{k}+\mathbf{G}_p,\mathbf{k}'+\mathbf{G}'_q}$  can be rewritten as  $\delta_{\mathbf{k}-\mathbf{k}',\mathbf{G}'_q-\mathbf{G}_p}$ , where  $\mathbf{G}'_q - \mathbf{G}_p = \mathbf{G}^{(s)}$  are the reciprocal lattice vectors of the super lattice if  $l \neq l'$ . This means that, the only non-zero matrix elements of  $\langle ls\mathbf{k}|H|l's'\mathbf{k}'\rangle$  for  $l \neq l'$  are those where  $\mathbf{k} - \mathbf{k}' = \mathbf{G}^{(s)}$ .

In transport studies, the low-energy regime is interesting. In the case of graphene, these states are close to the Dirac points  $\mathbf{K}$  and  $\mathbf{K}'$ , which is indexed in this section as  $\mathbf{K}_{\xi}$ , where  $\xi = \pm 1$  stand for the  $\mathbf{K}$  and  $\mathbf{K}'$  points. Around these points, the intralayer terms are described in Section 2.1.1 for SLG and in Section 2.1.2 for BLG. It is convenient to introduce  $\mathbf{q} = \mathbf{k} - \mathbf{K}_{\xi}$  and  $\mathbf{q}' = \mathbf{k}' - \mathbf{K}'_{\xi}$ , which are measured from the Dirac points<sup>5</sup>. For small twist angles, it's enough to consider only these states. To separate the intra and interlayer terms, Eq.(2.30) can be rewritten after introducing  $\mathbf{q}$  as

$$\langle ls\mathbf{k}|H|l's'\mathbf{k}'\rangle = \delta_{l,l'}H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k},\mathbf{k}'} + (1 - \delta_{l,l'})T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k}+\mathbf{G},\mathbf{k}'+\mathbf{G}'}, \quad (2.31)$$

where the first term is diagonal in  ${\bf k}$  and it is the Hamiltonian of an isolated layer. The second term with

$$T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q}) = \sum_{\mathbf{G},\mathbf{G}'} e^{i\mathbf{G}\tau_s} e^{-i\mathbf{G}'\tau_{s'}} H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q} + \mathbf{G})$$
(2.32)

describes the interlayer tunneling processes.

The tunneling amplitude  $\langle ls \mathbf{R}_n | H | l's' \mathbf{R}'_m \rangle$  between the layers depends only on the distance between the sites, which can be written as  $\sqrt{r^2 + d^2}$ , where r is the inplane distance of sites and d is the distance between the layers. d is larger than the distance between the atoms within the layers  $(a/\sqrt{3})$ . The hopping is decaying with the distance between the sites, and it is the strongest when the atoms are on top of each other. The tunneling amplitude for farther atoms, where the in-plane distance is comparable to or larger than the distance between the layers, is exponentially small. Thus, the tunneling amplitude is only large in a certain point of the space, where the atoms are on top of each other. As the other terms are exponentially small, their Fourier transform of  $H_{ls,l's'}(\mathbf{k})$  is sharp and goes to zero rapidly with  $\mathbf{k}$  when  $|\mathbf{k}| \cdot d > 1$  [54, 69]. In Eq.(2.31), the tunneling amplitude  $H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q} + \mathbf{G})$  is the largest at  $\mathbf{G} = \mathbf{0}$ , and due to the  $C_3$  symmetry (around the  $\Gamma$ -point) of graphene, there are

<sup>&</sup>lt;sup>5</sup>Here,  $\mathbf{K}_{\xi}$  are the K-points of layer l and  $\mathbf{K}'_{\xi}$  are the K-points of layer l'.

3 equivalent points<sup>6</sup> with  $\mathbf{G}_i$  as  $\mathbf{G}_1 = \mathbf{0}$ ,  $\mathbf{G}_2 = -\xi \mathbf{b}_1$ , and  $\mathbf{G}_3 = -\xi \mathbf{b}_2$ . In short,  $\sum_{\mathbf{G}} H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q} + \mathbf{G}) = \sum_{i=1}^{3} H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q} + \mathbf{G}_i)$  and all the other terms can be neglected. Ab initio supercell density-functional theory results in the same nonnegligible terms [70].

As  $\mathbf{q} \ll \mathbf{K}_{\xi} + \mathbf{G}_i$ , the  $\mathbf{q}$  dependence of  $H_{ls,l's'}(\mathbf{q} + \mathbf{K}_{\xi} + \mathbf{G}_i)$  is assumed to be negligible for  $l \neq l'$ :  $H_{ls,l's'}(\mathbf{q} + \mathbf{K}_{\xi} + \mathbf{G}_i) \approx H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{G}_i)$ . The  $C_3$  symmetry of  $H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{G}_i)$  produces the same matrix elements for every  $\mathbf{G}_i$  as

$$H_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{G}_i) = H_{ls,l's'}(\mathbf{K}_{\xi}).$$
(2.33)

This is shown perturbatively in Ref[71]. Substituting Eq.(2.33) in the interlayer term of Eq.(2.31) and writing out the summation of  $\mathbf{G}_i$  leads to

$$T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k}+\mathbf{G},\mathbf{k}'+\mathbf{G}'} = H_{ls,l's'}(\mathbf{K}_{\xi})\sum_{\mathbf{G}'} \left[ e^{-i\mathbf{G}'\tau_{s'}}\delta_{\mathbf{K}_{\xi}+\mathbf{q},\mathbf{K}'_{\xi}+\mathbf{q}'+\mathbf{G}'} + e^{-i(\xi\mathbf{b}_{1}\tau_{s}+\mathbf{G}'\tau_{s'})}\delta_{\mathbf{K}_{\xi}+\mathbf{q}-\xi\mathbf{b}_{2},\mathbf{K}'_{\xi}+\mathbf{q}'+\mathbf{G}'} \right].$$

$$(2.34)$$

At small twist angles, as  $|\mathbf{q}| \ll |\mathbf{K}_{\xi}|$  and  $|\mathbf{q}'| \ll |\mathbf{K}'_{\xi}|$ , there is only one  $\mathbf{G}'$  reciprocal lattice vector for each term in Eq.(2.34), that satisfies the Kronecker delta of the term: in the first term  $\delta_{\mathbf{K}_{\xi}+\mathbf{q},\mathbf{K}'_{\xi}+\mathbf{q}'+\mathbf{G}'} \neq 0$  if  $\mathbf{G}' = 0$ , in the second term  $\delta_{\mathbf{K}_{\xi}+\mathbf{q}-\xi\mathbf{b}_1,\mathbf{K}'_{\xi}+\mathbf{q}'+\mathbf{G}'} \neq 0$ o if  $\mathbf{G}' = -\xi \mathbf{b}'_1$  and in the last term  $\delta_{\mathbf{K}_{\xi}+\mathbf{q}-\xi\mathbf{b}_2,\mathbf{K}'_{\xi}+\mathbf{q}'+\mathbf{G}'} \neq 0$  if  $\mathbf{G}' = -\xi \mathbf{b}'_2$ . Writing these back to Eq.(2.34) and using the original notation of  $\mathbf{k}$  and  $\mathbf{k}'$ , the simplified expression is given for the interlayer tunneling matrix element as

$$T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k}+\mathbf{G},\mathbf{k}'+\mathbf{G}'} = H_{ls,l's'}(\mathbf{K}_{\xi})\left[\delta_{\mathbf{k},\mathbf{k}'} + e^{-i(\xi\mathbf{b}_{1}\tau_{s}-\xi\mathbf{b}'_{1}\tau_{s'})}\delta_{\mathbf{k}-\xi\mathbf{b}_{1},\mathbf{k}'-\xi\mathbf{b}'_{1}} + (2.35)\right]$$
$$e^{-i(\xi\mathbf{b}_{2}\tau_{s}-\xi\mathbf{b}'_{2}\tau_{s'})}\delta_{\mathbf{k}-\xi\mathbf{b}_{2},\mathbf{k}'-\xi\mathbf{b}'_{2}}\left].$$

With the derived expressions above, the matrix elements of the Bistritzer-MacDonald Hamiltonian are given by

$$\langle ls\mathbf{k}|H|l's'\mathbf{k}'\rangle = \delta_{l,l'}H_{ls,l's'}(\mathbf{k})\delta_{\mathbf{k},\mathbf{k}'} + (1-\delta_{l,l'})T_{s,s'}(\mathbf{K})\left[\delta_{\mathbf{k},\mathbf{k}'} + e^{-i(\xi\mathbf{b}_{1}\tau_{s}-\xi\mathbf{b}_{1}'\tau_{s'})}\delta_{\mathbf{k}-\xi\mathbf{b}_{1},\mathbf{k}'-\xi\mathbf{b}_{1}'} + e^{-i(\xi\mathbf{b}_{2}\tau_{s}-\xi\mathbf{b}_{2}'\tau_{s'})}\delta_{\mathbf{k}-\xi\mathbf{b}_{2},\mathbf{k}'-\xi\mathbf{b}_{2}'}\right],$$

$$(2.36)$$

where l is the layer index, s is the sub lattice index and  $\xi = \pm 1$  for the K and K' points, respectively. **k** is a **k**-space vector and  $\delta_{l,l'}$  is the Kronecker delta function, which is 1 for l = l' and zero otherwise. The first term describes the intralayer tunnelling with the **k**-space Hamiltonian of each individual layer of  $H_{ls,l's'}(\mathbf{k}) = H_{s,s'}^{(l)}(\mathbf{k})$ . The second term describes the interlayer tunneling, where  $T_{s,s'}(\mathbf{K})$  is the tunneling matrix amplitude

<sup>&</sup>lt;sup>6</sup>The graphene has 3 equivalent  $K_{\xi}$ -points.



Figure 2.7: Illustration of the coupled **k**-points in the **k**-space of the BM model, where  $a^{(s)}$  is the length of the superlattice unit vector. The positions of the red dots correspond to the matrix elements with **k'** of the top layer. These points are coupled to the blue dots which positions corresponds to the matrix elements from the bottom layer. The coupling between them is illustrated with black, red, and blue for the tunneling amplitudes of  $T_0$ ,  $T_+$ , and  $T_-$ , which are defined in Eq.(2.38), respectively. The **q**<sub>i</sub> vectors, shown in the figure, connect the two layers and they are defined in Eq.(2.39).

between the layers. In the BM model, the tunneling matrix amplitudes are constants  $T_{s,s'}(\mathbf{K}) = w[69].$ 

Rewriting Eq.(2.35) in matrix notation (for the sublattice index s) for graphene, the final expression of the interlayer tunneling is given by

$$T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k}+\mathbf{G},\mathbf{k}'+\mathbf{G}'} = T_{\mathbf{q}_0}\delta_{\mathbf{q}'-\mathbf{q},\mathbf{q}_0} + T_{\mathbf{q}_-}\delta_{\mathbf{q}'-\mathbf{q},\mathbf{q}_-} + T_{\mathbf{q}_+}\delta_{\mathbf{q}'-\mathbf{q},\mathbf{q}_+}, \qquad (2.37)$$

where

$$T_{\mathbf{q}_{0}} = \begin{pmatrix} H_{A,A'}(\mathbf{K}) & H_{A,B'}(\mathbf{K}) \\ H_{B,A'}(\mathbf{K}) & H_{B,B'}(\mathbf{K}) \end{pmatrix}, \qquad (2.38)$$

$$T_{\mathbf{q}_{-}} = e^{i\mathbf{b}_{1}'\mathbf{d}} \begin{pmatrix} H_{A,A'}(\mathbf{K}) & H_{A,B'}(\mathbf{K})e^{-\xi i 2\pi/3} \\ H_{B,A'}(\mathbf{K})e^{\xi i 2\pi/3} & H_{B,B'}(\mathbf{K}) \end{pmatrix},$$

$$T_{\mathbf{q}_{+}} = e^{i\mathbf{b}_{2}'\mathbf{d}} \begin{pmatrix} H_{A,A'}(\mathbf{K}) & H_{A,B'}(\mathbf{K})e^{\xi i 2\pi/3} \\ H_{B,A'}(\mathbf{K})e^{-\xi i 2\pi/3} & H_{B,B'}(\mathbf{K}) \end{pmatrix}$$

are tunnelling matrices connecting  $\mathbf{q}$  with  $\mathbf{q}' = \mathbf{q} + \mathbf{q}_i$ , where

$$\mathbf{q}_{0} = \mathbf{K}_{\xi} - \mathbf{K}_{\xi}', \ \mathbf{q}_{-} = \mathbf{K}_{\xi} - \xi \mathbf{b}_{1} - (\mathbf{K}_{\xi}' - \xi \mathbf{b}_{1}'), \ \text{and} \ \mathbf{q}_{+} = \mathbf{K}_{\xi} - \xi \mathbf{b}_{2} - (\mathbf{K}_{\xi}' - \xi \mathbf{b}_{2}') \ (2.39)$$

wave vectors connect the layers in **k**-space. The tunneling amplitudes  $H_{s,s'}(\mathbf{K})$  can be obtained from ab initio supercell density-functional theory[70, 72, 73]. The full matrix in the **k**-space is illustrated in Fig. 2.7.

The Hamiltonian connects an arbitrary **k**-point from the top layer with a diagonal term of  $H_{1s,1s'}(\mathbf{k})$ , to three other  $\mathbf{k} + \mathbf{q_i}$  points on the bottom layer with diagonal matrix elements of  $H_{2s,2s'}(\mathbf{k} + \mathbf{q_i})$ . The off-diagonal terms which connect these points are  $T_{ls,l's'}(\mathbf{K}_{\xi} + \mathbf{q})\delta_{\mathbf{k}',\mathbf{k}+\mathbf{q_i}}$ . The  $\mathbf{k} + \mathbf{q_i}$  points of the bottom layer are also connected to three other points from the top layer, which are also connected to 3 other points from the bottom layer, and so on. These couplings are shown with colored lines in the figure. The constructed matrix is infinitely large. It is solved by introducing a truncation vector such that the change of the low-energy levels is negligible by further increasing the size.

#### 2.3 Role of spin-orbit coupling in graphene

The spin-orbit coupling (SOC) is a relativistic effect and in the representation of 2s2p states of carbon, it can be represented as  $H_{SOC} = \lambda \mathbf{L} \times \mathbf{s}$ , where  $\lambda$  is the coupling strength,  $\mathbf{L}$  is the orbital moment operator and  $\mathbf{s}$  is the spin operator[74, 75]. From this, in graphene, the symmetry only allows one term in the second order, which can be written as

$$H_{\rm int} = \lambda_{\rm int} \xi \sigma_z s_z, \qquad (2.40)$$

where  $\lambda_{\text{int}}$  is the intrinsic spin orbit strength[76]. From first-principle calculations,  $\lambda_{\text{int}} \approx 12 \,\mu\text{eV}$  [77]. This opens a 24  $\mu\text{eV}$  gap, which can be neglected in transport. A finite perpendicular electric field may also induce spin-splitting as it breaks the inversion symmetry, allowing an additional SOC term, the Rashba-type SOC. Its strength depends on the electric field and approximately  $\lambda_{\text{R}} = 10 \,\mu\text{eV} \,\text{nm} \,\text{V}^{-1} \cdot E_z$ , where  $E_z$ is the perpendicular electric field[77, 78]. This is of no importance with the reachable electric fields with gate electrodes in experiments.

This negligible SOC and hyperfine coupling make graphene an ideal material for spintronics[24, 79]. The long spin relaxation times combined with the large mobilities of the charge carriers lead to an exceptionally long spin relaxation length of several tens of micrometers[24, 79–81]. These properties also make graphene an ideal platform for spin qubits defined by quantum dots[82–86]. In bilayer graphene (BLG), where the

electrons can be confined using electric fields, more than  $100\,\mu s$  to even half second of spin relaxation time have been demonstrated [87–90].

#### 2.3.1 Spin-orbit coupling in graphene/TMD heterostructures

The proximity of different materials to the graphene can substantially change its properties[5]. There are a lot of fascinating 2D materials that can be combined with graphene. Among them, the transition metal dichalcogenides are very interesting to the field of spintronics as they exhibit large intrinsic SOC [91, 92]. TMDs have a similar lattice structure to graphene and by placing graphene on a TMD, their bands can hybridize which increases the SOC in the graphene[25]. The enhancement of SOC in these heterostructures was demonstrated with weak localization measurements[27, 93–98], Shubnikov-de Haas (SdH) oscillations[93, 94, 99–101], spin valves[26, 28, 102–105] and quasiparticle interference imaging[106]. Similar measurements are shown in Chapter 6.

The proximity of the TMD breaks the inversion symmetry, which allows new SOC terms to add to the Hamiltonian of the graphene[107, 108]. From these terms, the most important is the Ising-type

$$H_{\rm I} = \xi \frac{\lambda_{\rm I}}{2} \sigma_z \tag{2.41}$$

also called valley-Zeeman SOC and the Rashba-type SOC

$$H_{\rm R} = \frac{\lambda_{\rm R}}{2} (\xi \sigma_x s_y - \sigma_y s_x). \tag{2.42}$$

Here,  $s_i$  and  $\sigma_i$  are Pauli-matrices acting on the spin and sublattice degree of freedom, respectively.  $\lambda_{\rm R}$  and  $\lambda_{\rm I}$  are the SOC strengths of the Rashba-type and Ising-type SOC, respectively. Their theoretical values are in the meV range and depend on the TMD[107] and also on the twist angle between the TMD and the graphene[109–111]. The Isingtype SOC leads to an effective valley-dependent magnetic field, that splits the spin degeneracy.

To use graphene in spintronic devices, electrical control of the spin information is necessary[25, 112], for which the induced SOC in graphene offers a way. The control of the spin currents in graphene/TMD heterostructures was successfully performed with electrical gating[26–29] and with spin-to-charge conversion, where a charge current induces a transverse spin current by SOC[30–33]. Moreover, the low-energy behavior is greatly influenced by the SOC not only in Bernal-stacked[112, 113] but also in twisted structures[114–117].

#### 2.3.2 SOC in BLG/TMD heterostructures



Figure 2.8: Effect of SOC on the band structure of BLG with only a bottom TMD layer  $(\lambda_{\rm I}^{\rm t} = \lambda_{\rm R}^{\rm t} = 0)$  using (a,b)  $\lambda_{\rm I}^{\rm b} = 2.5 \,\text{meV}$ ,  $\lambda_{\rm R}^{\rm b} = 0$ , in the K and K' valley, respectively, and (c)  $\lambda_{\rm I}^{\rm b} = 0$  and  $\lambda_{\rm R}^{\rm b} = 10 \,\text{meV}$  and (d)  $\lambda_{\rm I}^{\rm b} = 2.5 \,\text{meV}$  and  $\lambda_{\rm R}^{\rm b} = 10 \,\text{meV}$ . The color of the bands shows the spin polarization with the color bar plotted in panel (a).

The proximity effect is short-ranged in van der Waals heterostructures. As a result, in BLG/TMD heterostructures, the TMD affects only the closest layer of BLG[45, 118]. In general, BLG can be encapsulated between two TMDs, which induces SOC in both layers, but their strength can be different depending on the TMD and the twist angle between the BLG and TMD. This can be modeled with the addition of SOC terms in both layers in Eq.(2.10). The Hamiltonian of the heterostructure can be written in the  $(|A1\rangle, |B1\rangle, |A2\rangle, |B2\rangle) \otimes (|\uparrow\rangle, |\downarrow\rangle)$  basis as

$$H = H_{\rm BLG} + H_{\rm SOC}, \tag{2.43}$$

where  $H_{\text{SOC}}$  is the spin-orbit coupling term, defined as

$$H_{\rm SOC} = \begin{pmatrix} \xi \lambda_{\rm I}^b s_z/2 & i\lambda_{\rm R}^b s_-^{\xi} & 0 & 0\\ -i\lambda_{\rm R}^b s_+^{\xi} & \xi \lambda_{\rm I}^b s_z/2 & 0 & 0\\ 0 & 0 & \xi \lambda_{\rm I}^t s_z/2 & i\lambda_{\rm R}^t s_-^{\xi} \\ 0 & 0 & -i\lambda_{\rm R}^t s_+^{\xi} & \xi \lambda_{\rm I}^t s_z/2 \end{pmatrix}.$$
 (2.44)

 $H_{\text{SOC}}$  describes the proximity-induced Ising-type SOC with the parameters of  $\lambda_{\text{I}}^{i}$  and Rashba-type SOC parametrized with  $\lambda_{\text{R}}^{i}[113, 119]$ . Here,  $s_{\pm}^{\xi} = \frac{1}{2}(s_{x} + i\xi s_{y})$  and t and b indexes the top and bottom layers, respectively. In a BLG/TMD heterostructure, there is SOC at only one layer of the BLG (e.g.  $\lambda_{\rm I}^{\rm t} = \lambda_{\rm R}^{\rm t} = 0$ ). The band structure of the heterostructure is shown in Fig. 2.8. In panels (a) and (b), the effect of Ising-type SOC is shown at the K and K' point: it splits the spin degeneracy of the bands and makes them spin-polarized in the out-of-plane direction oppositely in the two valleys. The Rashba-type SOC splits the spin degeneracy at larger k points, as illustrated in Fig. 2.8c, and gives a complex in-plane spin texture. The combined effect of SOC terms makes the band spin-polarized in the out-of-plane direction at low energies, whereas for higher energies a more complicated, canted spin structure arises[112].



Figure 2.9: Effect of SOC and u on BLG encapsulated within two TMDs using  $\lambda_{\rm R} = 0$ . (a-c) With the same SOC strength in both layers of  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t} = 2.5$  meV. If  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t}$ , without u in (a) there is no gap. (b,c) Above  $|u| \approx |\lambda_{\rm I}^{\rm b} - \lambda_{\rm I}^{\rm t}|/2$  a gap begins to open similar to pristine BLG. (d-f) With opposite sign of the Ising-type SOC strengths in the layers as  $\lambda_{\rm I}^{\rm b} = 2.5$  meV and  $\lambda_{\rm I}^{\rm t} = -2.5$  meV. (d) At u = 0 a non-trivial gap opens. (e) At  $u \approx \pm |\lambda_{\rm I}^{\rm b} - \lambda_{\rm I}^{\rm t}|/2$ , the non-trivial gap closes. (f) Above  $|u| \approx |\lambda_{\rm I}^{\rm b} - \lambda_{\rm I}^{\rm t}|/2$  a trivial gap opens as in (panel c). The color of the bands shows the spin polarization with the color bar plotted in panel (a).

In a TMD/BLG/TMD heterostructure, SOC is induced in both graphene layers. Depending on the twist angle of the TMDs, the induced Ising-type SOC can be the opposite in the two layers[120]. If the TMDs are aligned, then  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t}$ . In this case, the Ising-type SOC behaves as an effective magnetic field and splits the bands as shown in Fig. 2.9a. Applying an electric field has a similar effect as in pristine BLG, it opens a band gap, but in this case, a band only opens if  $|u| > |\lambda_{\rm I}^{\rm b}|$  as illustrated in Fig. 2.9b and Fig. 2.9c.

If the relative twist angle between the TMDs is  $60^{\circ}\pm 120^{\circ}$ , then  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t}[113]$ . This is similar to Kane–Mele SOC[76]. In this case, Ising-type SOC opens a band gap, but the bands remain spin degenerate as shown in Fig. 2.9d. This gap is topologically different from the electric field opened gap. However, it is not a strong topological insulator, due to the  $2\pi$  Berry phase in BLG[50]. This gap can be closed by electric field by applying  $u \approx \pm |\lambda_{\rm I}^{\rm b} - \lambda_{\rm I}^{\rm t}|/2$  as shown in Fig. 2.9e. Applying a larger electric field opens a topologically trivial gap similarly to the case of  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t}$ .

#### 2.4 Transport in graphene

Graphene has remarkable transport properties. Its electrical mobility ( $\mu$ ), which describes how fast a charge carrier can move through the material with an electric field (**E**), can exceed 10<sup>5</sup> cm<sup>2</sup>/Vs[121–136]. In experiments, the mobility is usually determined using the Drude formula

$$\sigma_{xx} = ne\mu, \tag{2.45}$$

where  $\sigma_{xx}$  is the longitudinal conductivity. The conductivity is theoretically calculated either with the semiclassical Boltzmann transport theory or using the Kubo formalism[137]. The former fails to describe quantum phenomena like weak localization (described in Section 2.5.1), which is a quantum correction derived from the Kubo formalism.

In this section, the transport properties of graphene are described using the Boltzmann equation. The conductivity tensor in relaxation time approximation is given by

$$\sigma = \frac{e^2}{2} \int \mathrm{d}\epsilon g(\epsilon) \mathbf{v_k}^2 \tau_{\mathrm{m}}(\epsilon) \left(-\frac{\partial f}{\partial \epsilon}\right), \qquad (2.46)$$

where  $\mathbf{v}_{\mathbf{k}} = \hbar^{-1} \frac{dE(\mathbf{k})}{d\mathbf{k}}$  is the group velocity,  $\tau_{\mathrm{m}}(\epsilon)$  is the momentum relaxation time also called transport scattering time,  $f(E(\mathbf{k})) = \{1 + \exp[(E(\mathbf{k}) - \mu)/k_{\mathrm{B}}T]\}^{-1}$  is the Fermi-Dirac distribution where  $k_{\mathrm{B}}$  is the Boltzmann constant, T is the temperature and  $\mu(T)$  is the chemical potential[138]. At T = 0,  $E_{\mathrm{F}} = \mu(T = 0)$  and the longitudinal component of the conductivity tensor is given by

$$\sigma_{xx} = \frac{e^2 v_{\rm F}^2}{2} g(E_{\rm F}) \tau_{\rm m}(E_{\rm F}).$$
(2.47)

The transport scattering time  $\tau_{\rm m}$  arises from impurity scattering such as charged im-

purities, adsorbates, substrate corrugations, ripples, and also from phonon scattering. The mobility is related to  $\tau_{\rm m}$  by

$$\tau_{\rm m} = m^* \mu/e. \tag{2.48}$$

There is another important characteristic time in transport, the single-particle relaxation time also called quantum scattering time ( $\tau_q$ ), which determines the quantum level broadening  $\Gamma = \hbar/2\tau_q$  of the momentum eigenstates[139]. The difference between  $\tau$  and  $\tau_q$  from the following relations

$$\tau_{\rm m}^{-1} = \int_{0}^{\pi} Q(\theta)(1+\cos\theta)(1-\cos\theta)\mathrm{d}\theta \qquad (2.49)$$

$$\tau_{\mathbf{q}}^{-1} = \int_{0}^{\pi} Q(\theta) (1 + \cos \theta) \mathrm{d}\theta \qquad (2.50)$$

is the  $(1 - \cos \theta)$  weighting factor[140, 141]. Here,  $Q(\theta)$  is a function defined by the scattering mechanisms, and  $\theta$  is the angle of scattering. The  $(1 - \cos \theta)$  term weights the backward scattering of charge carriers by impurities, and makes the forward scattering relatively unimportant in  $\tau_{\rm m}$ , i.e. in the resistivity. The  $(1 + \cos \theta)$  is absent in normal 2D systems and it comes from the overlap of the wave function of graphene<sup>7</sup> and it is associated with Klein tunneling. This factor suppresses the large-angle scattering. Thus  $\tau_{\rm q}$  is sensitive only to small angle scattering events, while  $\tau$  is insensitive to both small and large angle scatterings and the dominant contribution comes from the  $\theta = \pi/2$  "right angle" scattering.

In the following, the T = 0 limit is investigated in the ballistic and diffusive regime. In the second part of this section, the *T*-dependence of  $\sigma_{xx}$  is described.

#### 2.4.1 Ballistic transport in graphene

The elastic mean free path  $(l_m)$  gives the average length a charge carrier can travel without significant back scattering. It is related to the transport scattering time as

$$l_{\rm m} = v_{\rm F} \tau_{\rm m}.\tag{2.51}$$

The transport is considered ballistic when  $l_{\rm m}$  is longer than the length of the device (L) between the source and drain  $(l_{\rm m} \gg L)$ . In this regime, the conductivity can be

<sup>&</sup>lt;sup>7</sup>From Eq.(2.6),  $F_s^{\dagger}(\mathbf{k})F_{s'}(\mathbf{k}') = [\exp(i\theta_{\mathbf{k}'} - i\theta_{\mathbf{k}}) + ss']/2$ . In the calculation of the scattering times, a term of  $|F_s^{\dagger}(\mathbf{k})F_{s'}(\mathbf{k}')|^2$  appears. Considering only elastic scatterings (s = s')[44],  $|F_s^{\dagger}(\mathbf{k})F_s(\mathbf{k}')|^2 = (1 + \cos(\theta_{\mathbf{k}'} - \theta_{\mathbf{k}})/2$ , where the scattering angle is  $\theta_{\mathbf{k}'} - \theta_{\mathbf{k}} = \theta$ .

calculated using the Landauer formalism as

$$\sigma_{xx} = \frac{L}{W} \frac{4e^2}{h} M(E_{\rm F}), \qquad (2.52)$$

where  $M(E_{\rm F})$  is the number of transport channels assumed to be  $M(E_{\rm F}) \gg 1$  and Wis the width of the device[142]. Using the periodic boundary condition,  $M(E_{\rm F})$  can be approximated as  $M(E_{\rm F}) = Wk_F/\pi = W\sqrt{n/\pi}$  by assuming perfect transmission. Here, Eq.(2.8) is used to convert  $k_F$ . In this approximation,  $\sigma_{xx} \propto \sqrt{n}$  as shown in Fig 2.10 with a red line, which is observed in high-mobility devices[143]. Another signature of ballistic transport is the visibility of transverse magnetic focusing, which is described in Section 2.5.4.

#### 2.4.2 Scattering mechanisms in graphene



Figure 2.10: The conductivity of SLG. In the ballistic limit(red curve),  $\sigma_{xx} \propto \sqrt{n}$  plotted with Eq.(2.52) with parameters of  $L = W = 1 \,\mu\text{m}$ . In the diffusive limit (blue curve), the combined conductivity of the short-ranged and long-ranged scatterers is plotted using Eq.(2.62) with the parameters of  $\kappa = 4.3$ ,  $n_i = 1 \cdot 10^{15} \,\text{cm}^{-2}$ ,  $n_d = 1 \cdot 10^{14} \,\text{cm}^{-2}$  and  $V_0 = 100 \,\text{eV} \cdot A_c$ .

In real graphene samples, like in metals, the mobility is hindered by the disorder. Depending on the type of the disorder, there are different scattering mechanisms present, which are hidden in  $\tau_{\rm m}(\epsilon)$  in Eq.(2.46). The most important scatterers at low temperatures are the charged impurities, short-range impurities and ripples[138, 144].

#### Scattering on charged impurities

One of the main limiting factors of mobility is the presence of charged impurities, which can be inside the substrate or near the graphene/substrate interface. Assuming random charge distribution, the relaxation time can be calculated as

$$\tau_{\rm m}^{\rm (Coulomb)} = \frac{\sqrt{n}}{F_1(2r_s)v_{\rm F}\sqrt{\pi}n_i},\tag{2.53}$$

where  $n_i$  is the number of impurities per unit area,

$$\frac{F_1(x)}{x^2} = \frac{\pi}{4} + 3x - \frac{3\pi x^2}{2} + x(3x^2 - 2)f(x)$$
(2.54)

with

$$f(x) = \begin{cases} \frac{\operatorname{arcosh}(1/x)}{\sqrt{1-x^2}} & \text{for } x < 1\\ \frac{\operatorname{arccos}(1/x)}{\sqrt{x^2-1}} & \text{for } x > 1 \end{cases}$$
(2.55)

 $r_s = e^2/\kappa \hbar v_{\rm F} 4\pi \epsilon_0$  is the effective fine structure constant for SLG,  $\kappa$  is the substrate dielectric constant<sup>8</sup>[44, 138, 144, 145] and  $\epsilon_0$  is the vacuum permittivity.  $r_s$  is in range of  $0 \leq r_s \leq 2.2^9$  and  $r_s \approx 0.51$  for a hBN encapsulated graphene ( $\kappa = 4.3$ ). As  $r_s$  is independent of n in SLG, from Eq.(2.53)  $\tau_{\rm m} \propto \sqrt{n}$  and the conductivity has a linear dependence on n as  $g(\epsilon) \propto \sqrt{n}$  in Eq.(2.47).

The quantum scattering time can be calculated within the random phase approximation (RPA)[140] as

$$\tau_{\rm q}^{\rm (Coulomb)} = \frac{2\sqrt{n}}{F_2(2r_s)v_{\rm F}\sqrt{\pi}n_i},\tag{2.56}$$

where

$$\frac{F_2(x)}{x^2} = \frac{1}{x} + xf(x) - \frac{\pi}{2}.$$
(2.57)

The ratio of  $\tau_{\rm m}^{\rm (Coulomb)}/\tau_{\rm q}^{\rm (Coulomb)}$  depends on  $r_s$  and it is always larger than 2. In the weak screening limit, when  $r_s \to 0$ ,  $\tau_{\rm m}^{\rm (Coulomb)}/\tau_{\rm q}^{\rm (Coulomb)}$  diverges.

#### Scattering on short-ranged impurities

Atoms (eg. hydrogen) or molecules (eg. hydrocarbon) can be absorbed on the surface of graphene[146] and they can act as short-ranged scatterers[147]. The short-ranged scatterers can be weak or strong. In the weak limit, the scattering time is given by

$$\tau_{\rm m}^{\rm (short)} = \frac{4\hbar^2 v_{\rm F}}{n_d V_0^2 \sqrt{\pi n}},\tag{2.58}$$

 $<sup>{}^8\</sup>kappa$  is the mean value of the dielectric constant of the medium above and below the graphene. <sup>9</sup>In vacuum ( $\kappa = 1$ )  $r_s \approx 2.2$ .

where  $n_d$  is the 2D impurity density and  $V_0$  is the strength of the short-ranged disorder potential and has units of  $\text{Jm}^2[148]$ . It can be calculated as  $V_0 \approx A_c V_{eff}$ , where  $A_c = \sqrt{3}a^2/2$  is the area of the unit cell of graphene and  $V_{eff} \sim 100 \text{ eV}$  is the effective impurity potential[149]. This gives a charge density independent conductivity as the DOS is  $g(\epsilon) \propto \sqrt{n}$  in Eq.(2.47).

Adsorbed hydrocarbons or vacancies give strong local potentials and the formula above gives an inappropriate result for them[149]. When  $V_0$  is strong, eg. a hydrogen atom binds covalently to a carbon atom, it acts as a resonant impurity at that site of the graphene[150, 151] meaning that it increases the likelihood of a charge carrier to be trapped in the vicinity of the adatom for a longer time, i.e. generating a resonant state[147]. Cracks and vacancies have similar effects, they all give rise to midgap states (zero-energy states)[147, 152]. The disorder potential can be modeled with a circularly symmetric potential with  $V(r < R) = \infty$  and V(r > R) = 0, which corresponds to a circular void of radius R, where the electron wave function is zero. Scattering at this potential induces a phase shift of the wave function of the scattering electron[153] and the phase shift ( $\delta_k$ ) at small k-vectors are given by

$$\delta_k \approx -\frac{\pi}{2} \frac{1}{\ln(kR)}.\tag{2.59}$$

In the strong scattering limit, the scattering rate of s-wave elastic scattering is given by

$$\frac{1}{\tau_{\rm m}} = \frac{8n_d}{\pi\hbar g(E)} \sin^2(\delta_k). \tag{2.60}$$

From Eq.(2.59) and Eq.(2.60) the scattering time for midgap states is

$$\tau_{\rm m} = \frac{k \ln^2(kR)}{\pi^2 v_{\rm F} n_{\rm D}},\tag{2.61}$$

which gives a sublinear n dependency of the conductivity mimicking the effect of charged impurities.

The combined effect of different scattering mechanisms on the conductivity can be taken into account (see the blue curve in Fig 2.10) using Matthiessen's rule ( $\tau_{\rm m}^{-1} = \tau_{\rm m}^{({\rm Coulomb})^{-1}} + \tau_{\rm m}^{({\rm short})^{-1}}$ ) and the resistivity is given by

$$\rho_{xx} = \frac{1}{\sigma_{\text{Coulomb}}} + \frac{1}{\sigma_{\text{short}}},\tag{2.62}$$

where the first term is computed from Eq.(2.53) and the second term is computed in case of weak short-range scattering from Eq.(2.58).

The quantum transport time can also be calculated for short-ranged scattering (e.g. see Eq.(17b) in Ref.[140]). The ratio of  $\tau_{\rm m}/\tau_{\rm q}$  depends on  $r_s$  and its value is always

between 1 and 2 for SLG. As for short-ranged scattering the ratio of  $\tau_{\rm m}/\tau_{\rm q} \leq 2$  and for long-range scattering  $\tau_{\rm m}/\tau_{\rm q} \geq 2$ , the ratio can help in determining the relevant scattering mechanism is disordered graphene. In experiments,  $\tau_{\rm m}/\tau_{\rm q}$  were found in the range of 1.5-5 of graphene devices on SiO<sub>2</sub> indicating that the long-ranged scattering dominates over short-ranged scattering in high-mobility devices[141, 154, 155].

#### Surface corrugations

Graphene is not atomically flat, it exhibits surface corrugations such as wrinkles, ripples, and crumples. They are formed around defects, edges, or impurities and also due to the substrate and thermal fluctuations [156, 157]. For instance, out-of-plane corrugations can originate from the substrate roughness or the strain during the fabrication. The deformation of graphene changes its properties [158], including its electrical structure. It suppresses weak localization (discussed in Section 2.5.1), opens a band gap [159], and also generates random pseudomagnetic fields [160], which leads to scattering.

Generally, ripples can be described with the height-height correlation function, which grows with the distance r as  $\langle (h(r) - h(0))^2 \rangle \sim r^{2H}$ , where H characterizes the fractal dimension of ripples[161, 162]. The scattering rate  $(\tau^{-1})$  is proportional to the correlation function of the scalar and vector potential induced by the corrugations, which have similar dependency as the height-height correlation function[160].

An exponent of 2H < 1 leads to density-independent conductivity, while an exponent of 2H = 1 indicates short-range correlations and  $\sigma_{xx} \propto 1/\ln^2(k_F a)$ . For 2H > 1,  $\sigma_{xx} \propto n^{2H-1}$  [162] and the electron scattering off ripples would mimic short-range disorder.

An exponent of 2H = 2 indicates a thermally excitable membrane, which is only loosely bound to the substrate. In this limit, scattering mimics the long-range Coulomb scattering as  $\sigma_{xx} \propto n[162, 163]$ . It is debated whether the main scattering mechanism in mediocre quality devices or negligible[144, 163]. The temperature induces such corrugations, and this conductivity contribution is proportional to  $\sigma_{xx} \propto n/T^2$ . During the fabrication of samples, when the graphene touches the substrate, the vdW forces pin the rippled configuration. Therefore, due to the substrate, the corrugations induced by the temperature become *T*-independent[162]. Besides ripples, a finite amount of wrinkles may also lead to linear *n*-dependence of the conductivity of SLG[164].

#### Remarks

According to experiments, in devices with mobility less than tens of thousands in  $cm^2/Vs$ , the conductivity is limited by screened charged impurities[123, 143, 165, 166].

The effect of short-ranged impurities is the deviation of the conductivity from linear behavior, such as saturation at high densities[167], as shown in Fig 2.10 with the blue curve at large n, which implies that the long-range Coulomb scattering becomes irrelevant in the high-density limit. Higher mobility devices ( $\mu > 10\,000\,\mathrm{cm^2/Vs}$ ) show sublinear conductivity[165, 168], in accordance with the long-range scattering being suppressed at large densities while other scattering mechanisms such as short-range scattering dominate, which most likely originate from the edge of the sample or corrugations varying on the atomic scale.

In the low-density regime  $(n \to 0)$ , the conductivity saturates due to density inhomogeneity (puddles), which is due to charged impurities and ripples[138, 147]. Moreover, even in the intrinsic limit, when there is no disorder, the conductivity at  $n \to 0$ remains finite and has a universal value of  $\sigma_{\min} = \sigma_{xx}(n \to 0) = 4e^2/\pi h$  due to the relativistic nature of electrons in graphene[142].

In BLG, similar scattering mechanisms determine the conductivity. However, the calculated conductivity is qualitatively different. For example, the short-ranged scattering gives  $\sigma \propto n$  and the screened Coulomb disorder gives  $\sigma \propto n^{\alpha}$ , where  $\alpha$  is n dependent and changes from 1 at low density to 2 at high density[138].

#### 2.4.3 Temperature-dependent transport

Besides the scattering mechanisms presented in the previous section, the electronphonon interaction also plays an important role at finite temperatures. For example, in normal metals (eg. Cu) the electron-phonon scattering dominates their resistance such that by increasing temperature, their resistivity increases. On the other hand, in an insulator, the conductivity increases by increasing the temperature due to thermal activation or variable-range hopping transport (see Eq.3.3). Due to the combination of different scattering mechanisms, both behaviors can be observed in graphene: it shows a metallic behavior at high densities and it can also show insulating behavior at low densities[169–171].

#### Temperature-dependence of the Coulomb scattering

In the previous section,  $\tau_{\rm m}^{\rm (Coulomb)}$  is discussed in the zero temperature limit. At finite temperatures, the Coulomb scattering has two independent sources of temperature dependency: from the definition<sup>10</sup> of  $\sigma_{xx}$  in Eq.(2.46) and from the temperature dependence of the dielectric function through screening<sup>11</sup>[173]. Their combined effect leads to

<sup>&</sup>lt;sup>10</sup>The explicit temperature dependence is in the Fermi-Dirac distribution.

<sup>&</sup>lt;sup>11</sup>The dielectric function contains the polarizability function, and its T dependence come from the Fermi-function in it (for the explicit formula, see for example Eq.(3) in Ref.[172]).

a non-monotonic dependence: at low temperatures  $(T \ll T_{\rm F})$  the conductivity decreases quadratically with the temperature as  $\sigma_{\rm Coulomb}(T)/\sigma_{\rm Coulomb}(T=0) \approx 1 - C_1(T/T_{\rm F})^2$ , where  $T_{\rm F} = E_{\rm F}/k_{\rm B}$  is the Fermi temperature and  $C_1$  is a positive constant that depends on  $r_s$ . At high temperatures  $(T \gg T_{\rm F})$  conductivity increases with T as  $\sigma_{\rm Coulomb}(T)/\sigma_{\rm Coulomb}(0) \approx C_2(T/T_{\rm F})^2$  with a positive constant of  $C_2[173]$ . As the Fermi temperature is very high  $(T_{\rm F} \gtrsim 1000 \,\mathrm{K}$  for  $n \gtrsim 10^{11} \,\mathrm{cm}^{-2})$ , the large temperature limit usually only applies near the Dirac point in experiments.

Moreover, there is another effect of the impurity-scattering, the collisional level broadening[174], which suppresses the temperature-dependent screening. For  $T \leq T_{\rm D}$ , where  $T_{\rm D} = \Gamma/(\pi k_{\rm B})$  is the Dingle temperature with the impurity-scattering induced collisional level broadening of  $\Gamma = \hbar/2\tau_{\rm q}$ , the temperature-dependence of screening is suppressed by scattering effects<sup>12</sup>[174–176]. This suppression leads to effectively linear instead of quadratic T dependence of the resistivity at low temperatures  $(T < T_{\rm D})$ [173], which is consistent with the experimental findings[143].

#### Temperature-dependence of the short-ranged scattering

The temperature-dependence of the conductivity from the short-range disorder can be calculated from Eq.(2.46) as

$$\sigma_{\rm short}(T) = \frac{\sigma_{\rm short}(T=0)}{1+e^{-\beta\mu}},\tag{2.63}$$

where  $\beta = 1/k_{\rm B}T$ .  $\mu$  is the chemical potential and  $\sigma_{\rm short}(T = 0) = e^2 v_{\rm F}^2 g(E_{\rm F}) \tau_{\rm m}^{\rm (short)}/2[173]$ . This temperature dependence is suppressed at low-temperature, but in the large T limit it approaches  $\sigma(0)_{\rm short}/2$ .

#### **Electron-phonon scattering**

At low temperatures (typically  $T \leq 20 \text{ K}[177]$  depending on the sample quality and the charge density<sup>13</sup>) the *T* dependence of  $\rho_{xx}(T)$  is completely dominated by disorder. At higher temperatures, the electron-phonon coupling starts to dominate  $\rho_{xx}(T)$ . In graphene, there are three different types of phonon scattering: intravalley acoustic and optical phonon scattering and intervalley phonon scattering[138]. In graphene, for the acoustic modes, the Debye temperature ( $\Theta_{\rm D}$ ), which is the temperature scale of the highest phonon mode if all modes are occupied, is quite high  $\Theta_{\rm D} = 2300 \text{ K}[179]$ . At  $T < \Theta_{\rm D}$ , mainly phonons with  $k_{ph} = k_{\rm B}T/\hbar v_{ph} < k_{\rm D}$  are populated, where  $k_{\rm D} = \omega_{\rm D}/v_{ph}$ 

<sup>&</sup>lt;sup>12</sup>In an illustrative way, as long as the collision level broadening is larger than the thermal broadening  $(k_{\rm B}T)$ , the polarizability function has a negligible T dependence[174].

<sup>&</sup>lt;sup>13</sup>At higher n as the screening is enhanced, the effect of charge impurities is less pronounced [178].

is the Debye wave vector<sup>14</sup> and  $v_{ph}$  is the sound velocity of a given phonon mode. In the low-density regime, which is important in transport in graphene,  $k_F \ll k_D$ , electrons can scatter only on a small fraction of the acoustic phonons, those with  $k_{ph} < 2k_F^{15}$ . This defines the Bloch-Grüneisen (BG) temperature ( $T_{BG} = 2\hbar v_{ph}k_F/k_B$ ). Below  $T < T_{BG}$ , the largest momentum of most of the occupied acoustic phonon modes is smaller than  $\hbar k_F$ , which allows only small-angle scattering processes. Above  $T > T_{BG}$ , the acoustic phonon modes are well populated above  $\hbar k_F$  in the momentum space allowing also large-angle scatterings.

In Fig. 2.11a, the acoustic phonon contribution to the resistance is shown at two different densities. There are two regions depending on  $T_{\rm BG}$ . If  $T \ll T_{\rm BG}$ ,  $\rho_{ac}(T) \approx \rho_0 + \alpha T^4$  with  $\rho_0$  the residual resistance from the disorder. If  $T \gtrsim T_{\rm BG}$ ,  $\rho_{ac}(T) \approx \rho_0 + \gamma T$  with the proportionality factors of

$$\gamma = \frac{\pi D_{\rm A}^2 k_{\rm B}}{4e^2 \hbar \rho_m v_{\rm F}^2 v_{ph}^2} \tag{2.64}$$

and

$$\alpha = \frac{12\zeta(4)\pi D_{\rm A}^2 k_{\rm B}^4}{e^2 \hbar^4 \rho_m v_{\rm F}^2 v_{ph}^5} \left(\pi n\right)^{-3/2},\tag{2.65}$$

where  $D_{\rm A} \approx 10 - 30 \,\mathrm{eV}[180\text{--}183]$  is an effective acoustic deformation potential<sup>16</sup>,  $\rho_m = 7.6 \cdot 10^{-7} \,\mathrm{kg/m^{-2}}$  is the graphene mass density,  $v_{ph} = 2 \cdot 10^4 \,\mathrm{m/s}[185]$  is an effective acoustic phonon velocity and  $\zeta$  is the Riemann-Zeta function[177, 180, 184, 186-188]. The charge density dependence of  $T_{\rm BG}$  is illustrated with red and black curves in Fig. 2.11a, which are plotted at different charge densities as a function of T. As n is increased, the transition temperature, when the behavior of  $\rho_{xx}$  changes from linear T to  $T^4$  dependence and characterized with  $T_{\rm BG}$ , increases.

At temperatures above  $T \gtrsim 270$  K, the optical phonon modes, especially the optical  $A'_1$  intervalley phonon mode[189], gives a non-negligible contribution to the temperature dependence of the resistance which can be calculated numerically from Eq.(2.46)[190, 191].

#### Remote phonon scattering

Besides ripples and remote charge impurities, the substrate has another important scattering mechanism that limits the mobility at higher temperatures ( $T \gtrsim 200 \text{ K}[178]$ ): scattering by surface optical phonons at the substrate which is called remote interfacial phonon (RIP) scattering[178, 191–193]. It originates from the transverse optical (TO)

<sup>&</sup>lt;sup>14</sup>With  $\hbar\omega_{\rm D} = k_{\rm B}\Theta_{\rm D}$ .

<sup>&</sup>lt;sup>15</sup>The factor of 2 comes from the full backscattering.

<sup>&</sup>lt;sup>16</sup>Here, the effect of acoustic phonon modes is described by scattering on a single effective phonon mode[184].



Figure 2.11: Temperature dependence of the resistivity of SLG. (a) The resistivity contribution from the acoustic phonons of graphene. The full formula[177] is used here instead of the asymptotic ones in Eq.(2.64) and Eq.(2.65) defined as  $\rho_{ac} = 8D_{\rm A}^2 k_F f_s(T_{\rm BG}/T)/e^2 \rho_m v_{ph} v_{\rm F}^2$ , where  $f_s(z) = \int_0^1 \frac{zx^4 \sqrt{1-x^2}e^{zx}}{(e^{zx}-1)^2} dx$  and  $D_{\rm A} = 19 \,\mathrm{eV}$  is used. At  $T \leq 0.1T_{\rm BG}$ ,  $\rho_{xx}(T) \propto T^4$  and at  $T \gtrsim 0.2T_{\rm BG}$ ,  $\rho_{xx}(T) \propto T$ . (b) The combined resistivity ( $\rho_{tot}$ ) using Eq.(2.67) which consists of  $\rho_{ac}$  (blue dashed lines) and  $\rho_{\rm RIP}$  (red dotted lines). The parameters used here are  $D_{\rm A} = 19 \,\mathrm{eV}$ ,  $n = 2 \cdot 10^{12} \,\mathrm{cm}^{-2}$ ,  $\alpha = 1$ ,  $B = 5 \cdot 10^{19} \,\mathrm{cm}^2 \mathrm{Ohm}$ ,  $\hbar \omega_1 = 102.12 \,\mathrm{meV}$ ,  $\hbar \omega_2 = 198.3 \,\mathrm{meV}$  and S = 2.05.

phonon modes of the substrate. The contribution of RIP scattering to the resistivity of graphene is given as

$$\rho_{\rm RIP}(T,n) = \frac{B}{n^{\alpha}} \left( \frac{1}{e^{\hbar\omega_1/k_{\rm B}T} - 1} + \frac{S}{e^{\hbar\omega_2/k_{\rm B}T} - 1} \right), \tag{2.66}$$

where the resistivity coefficient  $Bn^{-\alpha}$  is *n* dependent with phenomenological constants of  $\alpha$  and *B*. Here, only two modes exhibiting the largest oscillator strength with frequencies of  $\omega_1$  and  $\omega_2$  are considered with the ratio between their coupling strength of *S*.

By combining the scattering mechanism described in this section, for a good quality device at higher densities, where the temperature-dependence of the resistance is due to the electron-phonon scattering, the resistivity can be well described with

$$\rho_{xx}(T,n) = \rho_0(n) + \rho_A(T) + \rho_{\rm RIP}(n,T), \qquad (2.67)$$

where  $\rho_0(n)$  is the residual resistivity at low temperature,  $\rho_A(n,T) = \frac{\pi D^2 k_B T}{4e^2 \hbar \rho_m v_F^2 v_{ph}^2}$  is the contribution from the acoustic phonons (see Eq.(2.64)) and  $\rho_{\text{RIP}}(n,T)$  as defined in

Eq.(2.66). Eq.(2.67) is plotted at  $n = 2 \cdot 10^{12} \text{ cm}^{-2}$  in Fig 2.11b. As described,  $\rho_{xx}(T)$  is dominated by RIP scatterings above  $T \gtrsim 200 \text{ K}$  and below  $\rho_{xx}(T) \propto T$  due to the acoustic phonons in graphene.

#### Temperature dependence of the low-density regime

At low charge densities ( $n < n_r$ , where  $n_r$  is the magnitude of disorder-induced carrier density fluctuation also called residual doping),  $\sigma_{\min}$  is expected to vary with temperature only for  $k_{\rm B}T > E_{\rm puddle}$ , where  $E_{\rm puddle} = \hbar v_{\rm F} \sqrt{\pi n_r}$  for SLG[13, 143, 194]. The combined effect of the temperature dependence of the short-range and long-range disorder and the phonon contribution lead to the temperature dependence of  $\sigma_{\min}$ [195] as

$$\sigma_{\min}(T) = \left[ \left( \sigma_{\text{short}}(T) + A_c T^2 \right)^{-1} + B_p T + \rho_{\text{RIP}} \right]^{-1}, \qquad (2.68)$$

where  $\sigma_{\text{short}}(T)$  is defined in Eq.(2.63),  $A_c$  is a coefficient related to the Coulomb disorder<sup>17</sup> and  $B_p$  is related to acoustic phonon scattering ( $\gamma$  defined in Eq.2.64).

#### 2.5 Magneto-transport in graphene

When a material is placed in an external magnetic field, the charge carriers are affected by the magnetic field during transport. The magnetic field dependence of the resistance is called magnetoresistance, and it is a versatile tool to study the properties of the materials. In magneto-transport, a variety of interesting phenomena occurs, like weak localization, Shubnikov-de Haas oscillations, quantum Hall effect, large magnetoresistance, or magnetic focusing. In this section, a short description of these effects is given below.

#### 2.5.1 Weak localization

Due to inelastic scatterings, the charge carriers' energy changes which leads to a loss of coherence. The time scale that characterizes this is called phase coherence time  $\tau_{\phi}$ . The semiclassical Boltzmann equation which is discussed in Section 2.4, doesn't include coherence effects. However, the coherence effects lead to modification of the conductivity, and in shorter length scales than the phase coherence length ( $L_{\phi} = \sqrt{\tau_{\phi}D}$ , where D is the diffusion coefficient) the transport is coherent. The coherence length decreases with the temperature as T leads to thermal broadening of the energy of the particles (dephasing), and also at higher temperatures, the number of inelastic electronphonon scattering events is increased. Let's consider the probability of a charge carrier

 $<sup>^{17}</sup>A_c$  is related to  $C_2$  in the  $(T >> T_{\rm F})$  limit, which is relevant near the CNP.
going from  $\mathbf{r}_1$  to  $\mathbf{r}_2$  with the probability amplitudes of the path  $A_i$  as

$$P(\mathbf{r}_1, \mathbf{r}_2) = \left| \sum_i A_i \right|^2 = \sum_i |A_i|^2 + \sum_{i \neq j} A_i A_j^*.$$
(2.69)

The first term describes the classical probability of propagation, and the second term comes from the interference. In a diffusive system, the phase of  $A_i$  is random, and they average to zero in the second term except when  $\mathbf{r}_1 = \mathbf{r}_2$  (backscattering). In the case of backscattering, if the time-reversal symmetry (TRS) is conserved, the phase of a backscattered trajectory is the same as the phase of its time-reversed pair. These pairs interfere constructively and the probability of backscattering is twice as much as in the classical case when the second term in Eq.(2.69) is absent. This coherent backscattering is called weak localization (WL)[196].



Figure 2.12: An example of a closed loop that contributes to the interference. Multiple scattering events lead to backscattering. If TRS is present, the backscattered path and its reversed path interfere constructively (destructively in the presence of SOC). The enclosed surface is shown with a gray area, which  $\phi_{AB}$  depends on.

If SOC is present, then  $A_i$  become  $2 \times 2$  matrices in the spin space and can be written as  $A_i = A_i^0 \mathbb{1}_{2\times 2} + \mathbf{A} \cdot \mathbf{s}$ , where  $A_i^0$  is the spin-independent part of  $A_i$ ,  $\mathbf{A}$  is the spindependent probability amplitude and  $\mathbf{s} = (s_x, s_y, s_z)$  is the vector of Pauli matrices. The time reversed pair of  $A_i$  is  $\mathcal{T}A_i = A_i^0 \mathbb{1} - \mathbf{A} \cdot \mathbf{s}$ . The different sign of  $A_i$  and  $\mathcal{T}A_i$ results in a probability of backscattering which is half of the classical value, which results in destructive interference, which is called weak antilocalization (WAL)[197]. In a magnetic field, the particles pick up another phase called the Aharonov-Bohm phase  $(\phi_{AB} = 2e\pi BS/h)$ . As  $\phi_{AB}$  depends on the enclosed surface S perpendicular to  $\mathbf{B}$ , as  $\mathbf{B}$  is increased, first the contribution from the largest circles is suppressed by averaging over the trajectories and the contribution from the smallest ones remain unchanged. Thus, as the magnetic field is increased, fewer and fewer closed paths contribute to the interference and the resistance drops to its classical value.

In graphene, elastic scattering can happen between the valleys, which is characterized by the intervalley scattering time  $(\tau_{iv})$ , or within a valley, which is characterized by the intravalley scattering time  $\tau_z$ . The intravalley scattering is caused by long-ranged scattering, whereas the intervalley scattering is due to atomically sharp scatterers. The trigonal warping in graphene breaks the  $\mathbf{p} \to -\mathbf{p}$  symmetry within a valley as  $\epsilon(K_{\pm}, \mathbf{p}) \neq \epsilon(K_{\pm}, -\mathbf{p})$ , but due to the TRS, it has an opposite sign in the two valleys leading to  $\epsilon(K_{\pm}, \mathbf{p}) = \epsilon(K_{\mp}, -\mathbf{p})$ . As a result, the trigonal warping suppresses the interference effects from intravalley scattering while the correction to the resistance from the intervalley scatterings is unchanged. A time scale can also be attributed to the trigonal warping  $\tau_w$ . These scattering mechanisms contribute the following correction to the magnetoconductivity[198]

$$\Delta\sigma(B) = \sigma(B) - \sigma(0) = \frac{e^2}{\pi h} \left[ F\left(\frac{B}{B_{\phi}}\right) - F\left(\frac{B}{B_{\phi} + 2B_{\rm iv}}\right) - 2F\left(\frac{B}{B_{\phi} + B_{\rm iv} + B_{\ast}}\right) \right],\tag{2.70}$$

where  $F(x) = \ln(x) + \Psi(1/2 + 1/x)$  with the digamma function  $\Psi(x)$ ,  $B_j = \hbar/(4eD\tau_j)$ and  $\tau_* = (1/\tau_w + 1/\tau_z)^{-1}$ . For simplicity, in the rest of this thesis,  $\tau_*$  is referred as the intravalley scattering time. Eq.(2.70) is plotted in Fig. 2.13a for the relevant scenarios. The colors in the figure show curves with different phase coherence times, which changes the curvature of the curves around B = 0. In the limit of  $\tau_{\phi} > \tau_{iv}$ , the magnetoresistance has a WL behavior meaning that  $\Delta \sigma$  increases by increasing the magnetic field, which is usually observed in graphene samples[163, 199–202]. The other limit is when  $\tau_{iv} >$  $\tau_* \geq \tau_{\phi}$ , when the magnetoresistance shows WAL-like behaviour meaning that  $\Delta \sigma$ decreases by increasing the magnetic field[200].

The presence of SOC in graphene, like in graphene/TMD heterostructures, introduces new scattering mechanisms, which are characterized by  $\tau_{\rm R}$  for the Rashba-type SOC and  $\tau_{VZ}$  for the Ising-type SOC. The scattering mechanisms can be combined into  $z \rightarrow -z$  symmetric part  $\tau_{\rm sym}$ , which depend on  $\tau_{VZ}$  and an antisymmetric part  $\tau_{\rm asy}$ , which depend on  $\tau_R$ . The magnetoconductivity correction in the limit when the intervalley scattering dominates is described by[203]

$$\Delta\sigma(B) = -\frac{e^2}{2\pi h} \left[ F\left(\frac{B}{B_{\phi}}\right) - F\left(\frac{B}{B_{\phi} + 2B_{\text{asy}}}\right) - 2F\left(\frac{B}{B_{\phi} + B_{\text{asy}} + B_{\text{sym}}}\right) \right]. \quad (2.71)$$

If  $\tau_{iv}$  and  $\tau_*$  are comparable with the other scattering times, more correction terms have to be included, and it can be described with Eq.(28) in Ref.[204]. The relevant scenarios of Eq.(2.71) are shown in Fig. 2.13b. In the case of small  $\tau_{asy}$  and  $\tau_{sym}$  compared to  $\tau_{\phi}$ , it shows WAL behavior, and the background is mostly affected by  $\tau_{sym}$ . If  $\tau_{asy} \gg \tau_{\phi}$ 



Figure 2.13: Magnetoresistance corrections for different scattering times. (a) Eq.(2.70) with  $\tau_{iv} = 1$  ps and  $\tau_* = 0.1$  ps at different phase coherence times. If  $\tau_{\phi} > \tau_{iv} > \tau_*$ , the curve has WL behaviour (orange curve). If  $\tau_{iv} > \tau_* \ge \tau_{\phi}$ , the (blue) curve has WAL-like behaviour, i.e. negative magnetoconductance. If  $\tau_{iv} \gg \tau_* \gg \tau_{\phi}$ , there is no effect (black). (b) Eq.(2.71) at different scenarios. If  $\tau_{\phi}$  is similar to  $\tau_{asy}$  and  $\tau_{sym}$  there is WL and if  $\tau_{\phi}$  is larger than  $\tau_{asy}$  and  $\tau_{sym}$ , there is WAL.

the curve shows WL behavior due to the cancellation of the first two terms.

In an ideal two-dimensional system, the in-plane magnetic field  $\mathbf{B}_{\parallel}$  spin polarizes the graphene carriers through the Zeeman effect leading to positive  $B_{\parallel}^2$  dependent magnetoconductance[205], while having no effect on the interference corrections[206]. However, real 2D systems are rippled and they have out-of-plane corrugations originating from the substrate roughness or the strain during the fabrication. The height of the corrugations usually follows a Gaussian distribution with root-mean-square (RMS) height fluctuations of Z. The in-plane expansion of corrugations is characterized by the lateral correlation length of the corrugations R. When the 2D crystal is put in  $\mathbf{B}_{\parallel}$ , it has random out-of-plane magnetic field components originating from corrugations. These out-of-plane components lead to dephasing and changes the dephasing rate by  $\tau_{\phi}^{-1} \rightarrow \tau_{\phi}^{-1} + \tau_{\parallel}^{-1}$ [201, 202, 207, 208], where

$$\tau_{\parallel}^{-1} = \sqrt{\pi} \frac{e^2}{\hbar^2} v_{\rm F} Z^2 R B_{\parallel}^2.$$
 (2.72)

The dephasing rate in Eq.(2.72) holds for Gaussian correlated corrugations with an in-plane correlation length of R and with the RMS height fluctuations of Z.

### 2.5.2 Shubnikov-de Haas oscillations

The Shubnikov-de Haas (SdH) effect is the appearance of oscillations in the magnetoresistivity as a function of magnetic field, B at low temperatures when  $k_{\rm B}T \ll \hbar\omega_c$ , where  $k_{\rm B}$  is the Boltzmann constant, T is the temperature and  $\omega_c = eB/m^*$  is the cyclotron frequency [209, 210]. The oscillation is periodic over 1/B and the periodicity is given by

$$\Delta\left(\frac{1}{B}\right) = \frac{2\pi e}{\hbar S_{\rm F}},\tag{2.73}$$

where  $S_{\rm F}$  is greatest cross-section of the Fermi surface perpendicular to the magnetic field[210-212]. The origin of the oscillations is the formation of Landau-levels in a magnetic field, which leads to a magnetic field-dependent DOS. The scattering rate is proportional to the DOS in Born approximation and the longitudinal resistivity  $\rho_{xx}$ is proportional to the scattering rate [213]. Therefore, the oscillations in the magnetoresistivity and in the DOS at the Fermi level are due to the Landau level quantization. Generally, the longitudinal resistance can be written in the region of the SdH oscillations[48, 214-217] as

$$\rho_{xx} = \rho_0 \left[ 1 + c \sum_{s=1}^{\infty} D(sx) e^{-\frac{s\pi}{\omega_c \tau_q}} \cos\left(s \frac{\hbar S_F}{eB} - s\pi + s\phi_0\right) \right], \qquad (2.74)$$

where c is a constant prefactor,  $\tau_{q}$  is the lifetime of the charge carrier, which is defined with Eq.(2.50), s is the harmonic order,  $\phi_{0}$  is the Berry phase, which is zero in conventional 2D systems,  $\pi$  in SLG[47, 218] and  $2\pi$  in BLG[219, 220]. D(sx) is the temperature damping factor given as

$$D(sx) = \frac{s2\pi^2 k_{\rm B}T/\hbar\omega_c}{\sinh(s2\pi^2 k_{\rm B}T/\hbar\omega_c)}.$$
(2.75)

At a low magnetic field, it is enough to sum only at the first few harmonics of s[215]and in experimental studies, usually only the s = 1 term is considered[47, 221–223].

In experiments,  $S_{\rm F}$  can be obtained from the oscillation frequency using Eq.(2.73) and if there is more than one Fermi surface, all of them can be determined from the magnetoresistivity. From the exponential part of Eq.(2.74), which is called the Dingle factor,  $\tau_{\rm q}$  can be obtained. Lastly, from the *T* dependence, the effective mass can also be obtained.

#### 2.5.3 Quantum Hall measurements

In magnetic field  $(\mathbf{B})$ , the charge carriers, with  $\mathbf{v}$  velocity, experience the Lorentz force

$$\mathbf{F} = -e\left(\mathbf{E} + \mathbf{v} \times \mathbf{B}\right),\tag{2.76}$$

which leads to finite off-diagonal matrix elements of the conductivity tensor as

$$\sigma_{xy} = ne/B, \tag{2.77}$$

which is called classical Hall conductivity. The Hall resistance can be calculated as

$$\rho_{xy} = \frac{\sigma_{xy}}{\sigma_{xx}^2 + \sigma_{xy}^2},\tag{2.78}$$

which increases linearly with B as shown at low magnetic fields in Fig. 2.14c. At higher magnetic fields,  $\rho_{xy}$  starts to behave non-linearly and has plateaus of  $h/e^2\nu$ , where  $\nu$  is the Landau level filling factor. This quantization of the Hall resistance was first discovered in two-dimensional electron gas (2DEG)[224].

The quantized Hall resistance (quantum Hall effect) can be understood as the following. In a magnetic field Landau levels (LL) form. Their energy can be calculated by replacing the canonical momentum in the Hamiltonian with the kinetic impulse, and then the wave vector  $\mathbf{k}$  is replaced with  $\mathbf{q} = \mathbf{k} - \frac{e}{\hbar}\mathbf{A}$ , where  $\mathbf{A}$  is the vector potential. For 2DEG, the solution is given by

$$E_N = \left(N + \frac{1}{2}\right)\hbar\omega_c,\tag{2.79}$$

where  $\omega_c = eB/m^*$  is the cyclotron frequency and N quantum number indexes the LLs and it is a non-negative integer number[210]. The degeneracy of these levels is given by

$$N_p = g \frac{\Phi}{\Phi_0},\tag{2.80}$$

where g is the degeneracy factor (g = 2 for spin degeneracy),  $\Phi_0 = h/e$  is the magnetic flux quantum and  $\Phi = BA$  is the magnetic flux. The charge density is defined as the number of occupied states divided by the volume or in 2D the area, which is for the Nth occupied LL given as

$$n = \frac{NN_p}{A} = \frac{gNBe}{h},\tag{2.81}$$

which can be rewritten for the filling factor, which gives the amount of filled LLs  $(\nu = gN)$  as

$$\nu = \frac{nh}{eB}.\tag{2.82}$$



Figure 2.14: Quantum Hall effect in graphene. (a) Schematic illustration of the LL structure of SLG as a function of y or k. (b) Illustration of the edge states of  $\nu = 6$  in SLG with red and magenta arrows in a Hall-bar geometry in perpendicular B field. The chemical potentials of the contacts are labeled with  $\mu_i$ . (c-d) Hall resistance  $\rho_{xy}$  and magnetoresistance  $\rho_{xx}$  as a function of out-of-plane magnetic field (c) in SLG and (d) in BLG.  $\rho_{xx}$  is calculated with Eq.(2.74) with  $\phi_0 = \pi$  for SLG and  $\phi_0 = 2\pi$  for BLG.  $\rho_{xy}$  is calculated with Eq.(2.78), where  $\nu$  is calculated with Lorentzian Landau-level broadening[225] as  $\nu = g \int \sum_{n=0}^{\infty} g_{LL}(E) f(E) dE$ , where g is the LL degeneracy (g = 4 for graphene), f(E) is the Fermi-Dirac distribution and  $g_{LL} = \frac{1}{\pi\Gamma[1+(E-E_N)^2/\Gamma^2]}$ , where  $\Gamma$  is the quantum level broadening and  $E_N$  is calculated with Eq.(2.85) for SLG and Eq.(2.87) for BLG. (c) QHE in SLG calculated with the parameters  $n = 10^{12} \text{ cm}^{-2}$ , T = 5 K and  $\Gamma = 0.5 \text{ meV}$ . (d) QHE in BLG calculated with  $E_{\rm F} = 56.6 \text{ meV}$ , from which  $n \approx 1.5 \cdot 10^{12} \text{ cm}^{-2}$ , T = 5 K and  $\Gamma = 0.5 \text{ meV}$ .

Real samples have finite size and the physical boundaries give a confining potential, which bends the LLs. The Landau states can be written in the form of  $\Psi(x,y) = e^{ikx}\phi(y)$  with a wave number k in the x direction and  $\phi(y)$  which is the harmonic oscillator solution of Hamiltonian of the system with Hermite-polynomials and centered to  $y_k = \hbar k/eB$ . In Fig. 2.14a the bending is illustrated as a function k or y-coordinate. The LLs in the middle of the sample look like the LLs in an unconfined system. However, near the edge of the sample, there are allowed states. Consequently, when the Fermi energy lies between two LLs, only states near the edge of the sample are allowed. These are called edge states and have a velocity parallel to the edge[226]. These propagating states, which carry charge current in one direction, are spatially separated from those which carry the charge in the opposite direction. This separation leads to an exponentially small overlap of the wave functions of the states carrying current in the opposite direction, which leads to the suppression of backscattering. As a result, in Fig. 2.14b the edge states carry current from left to right and from right to left on the opposite side of the sample. The edge states moving to the right (left) are in equilibrium with the contact on the left (right) with a chemical potential of  $\mu_{\rm L}(\mu_{\rm R})$ . As a result, the chemical potential of the floating contact is  $\mu_2 = \mu_3 = \mu_{\rm L}$  and  $\mu_1 = \mu_{\rm R}[227]$ . The longitudinal voltage is zero, as  $V_{xx} = (\mu_2 - \mu_3)/e = 0$ . Whereas the longitudinal voltage is finite  $V_{xy} = (\mu_1 - \mu_2)/e = (\mu_{\rm L} - \mu_{\rm R})/e$ . In the Landauer formalism, the current is given by

$$I = \frac{e}{h}M(\mu_{\rm L} - \mu_{\rm R}),$$
 (2.83)

where M is the number of transport channels, which is in this case the number of edge states  $M = \nu$ . Thus, the longitudinal resistance  $(R_{xx} = V_{xx}/I)$  is zero and the Hall resistance is

$$R_H = \frac{V_{xy}}{I} = \frac{h}{\nu e^2}.$$
 (2.84)

These statements above are only valid if the Fermi level is between the LLs, If the LLs are partially filled, there are states in the bulk allowing backscattering, and the Hall conductance is no longer quantized. In real systems there is disorder. The disorder leads to the broadening of the LLs, characterized by the level broadening  $\Gamma$ . The states induced by the disorder are localized, thus the bulk is insulating and the current is carried by the propagating edge states[209].

For single-layer graphene, the LL energies are given by[75]

$$E_N = \operatorname{sgn}(N) \sqrt{2ev_{\rm F}^2 \hbar |NB|}, \qquad (2.85)$$

where N is an integer. This is non-equidistant and the N = 0 LL has zero energy unlike Eq.(2.79). In graphene, there is usually spin and valley degeneracy, therefore  $g = g_s g_v = 4$  and as the N = 0 state is equally shared by electrons and holes, it contains half as many states as all the other states when counting only to electrons or holes, thus the LL filling factor is given as  $\nu = g\left(N + \frac{1}{2}\right)$ , which takes the values of  $\nu = \pm 2, \pm 6, \pm 10 \dots$  The Hall resistance for SLG is given by

$$\rho_{xy} = \frac{h}{g_s g_v \left(N + \frac{1}{2}\right) e^2},\tag{2.86}$$

and it is shown in Fig. 2.14c with black curves as a function of the magnetic field. At large magnetic fields, only the zeroth LL is occupied, and as B is decreased other LLs are filled which are 4-fold degenerate.

For bilayer graphene, if the remote hoppings are neglected, the LL energies are given by

$$E_N^{(\pm)} = \pm \hbar \omega_c \sqrt{N(N-1)},$$
 (2.87)

where  $m^*$  in  $\omega_c$  is given by  $m^* = \gamma_1/2v_F^2[45]$ . The degeneracy of the LLs for  $N \ge 2$  is the same as in SLG, but the zeroth and first levels are degenerate. The Hall resistance is the same as for a 2DEG, but there is no plateau at n = 0:

$$\rho_{xy} = \frac{h}{g_s g_v N e^2},\tag{2.88}$$

N is a non-zero integer<sup>18</sup>.  $\rho_{xy}$  for BLG is shown in Fig. 2.14d with black curves as a function of B. Plateaus of  $\rho_{xy}$  occur at  $h/N4e^2$ . The effect of remote hoppings, the electric field, and SOC on the LL energies is discussed in Section 4.1.1.

The longitudinal resistance  $\rho_{xx}$  is shown with blue curves in Fig. 2.14c for SLG and Fig. 2.14d for BLG as a function of B at fixed n.  $\rho_{xx}$  has zero value when the LLs are filled and the Fermi level lies between two LLs. When  $E_{\rm F}$  is on a LL, it has a non-zero value (see Eq.(2.74)).

### 2.5.4 Magnetic focusing

The transport is ballistic when the mean free path is larger than the dimension of the sample, i.e. the electron can travel long distances without scattering. In this case, an out-of-plane magnetic field may focus electrons in a certain device geometry, which phenomenon is called transverse magnetic focusing (TMF). Generally, the states that are detectable in transport are only those within a  $k_{\rm B}T$  window near the Fermi level and are moving with Fermi velocity[228]. The electron trajectories can be obtained from the semiclassical equation of motion

$$\hbar \frac{\mathrm{d}\mathbf{k}}{\mathrm{d}t} = -e\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{r} \times \mathbf{B},\tag{2.89}$$

which gives that, the real space trajectory of the electrons can be obtained by rotating the Fermi surface by 90° and scaling it by  $\hbar/eB$ . If the Fermi surface is a circle, then the cyclotron radius is

$$r_c = \frac{\hbar k_F}{eB}.\tag{2.90}$$

In TMF experiments, the charge carriers are injected from narrow contacts isotropically as illustrated in Fig. 2.15a. Near the boundary, the charge carriers bounce back leading to skipping orbits[229] which bunch into caustics[230, 231]. The resulting focal points are at integer multiples of  $2r_c$  as illustrated for one bouncing of the edge in Fig. 2.15b. By placing a detector contact at a distance of L from the injector contact,

<sup>&</sup>lt;sup>18</sup>For BLG,  $\nu = g_s g_v N = \pm 4, \pm 8, \pm 12...$ 



Figure 2.15: (a-b) Classical trajectories of electrons injected from a point contact isotropically when the Fermi surface is a circle. Electrons are focused at  $2r_cp$ , where p is an integer. Trajectories (a) without reflections of the y = 0 lower boundary and (b) including one bounce. (c) Schematic set-up of the TMF measurements: the injector (I) contact is used to inject the charge carriers of the material by injecting I current and the collector (c) contact is used to detect the signal by measuring  $V_c$ . (d) TMF peaks from Eq.(2.92 as a function of n and B. Inset in (c): schematic illustration of  $R_{nl} = V_{nl}/I$  at  $n = 10^{12}$  cm<sup>-2</sup> as a function of B. The area of the first peak is marked with grey color.

as illustrated in Fig. 2.15c, the magnetic field required to focus the electrons is

$$B_j = \frac{2\hbar k_{\rm B} p}{eL},\tag{2.91}$$

where j = 1, 2, ... and j - 1 is the number of reflections from the boundary. In SLG  $k_{\rm F}$  can be calculated from Eq.(2.8) leading to

$$B_j = \frac{2\hbar\sqrt{\pi n}p}{eL},\tag{2.92}$$

which is plotted in Fig. 2.15d. An equidistant series of peaks appear at a fixed n and the deflection of the charge carrier depends on the sign of the charge of the carriers[232–234].

The temperature affects the TMF as the energy window of the excited carriers is  $\sim k_{\rm B}T$  near the Fermi level. At low temperatures, the electrons are phase coherent,

and the TMF signal exhibits a fine structure [235-237]. When the temperature-induced broadening of the Fermi wave vector  $(k_{\rm F})$  is comparable with 1/L, the electrons are no longer coherent, and the quantum interference is lost [238] which affects the fine structure of the TMF signal. On the other hand, T introduces new scattering channels at high temperatures, which leads to the loss of ballistic behavior of charge carriers, thus the TMF signal. In metals, mainly the electron-phonon scattering is responsible for the loss of TMF signal as the temperature is increased [239]. The suppression of the TMF signal upon heating is caused by electron-electron scattering in graphene superlattices [233, 234] and also in 2DEGs [240].

In experiments, the area under the first focusing peak  $A_1$ , which is shown with grey are in the inset of Fig. 2.15d, can be used to study the effect of temperature[233].  $A_1$ is proportional to the fraction of charge carriers, that propagate from the injector to the collector ballistically along the semicircle. The electrons need  $t = \pi L/2v_F$  time to propagate along the semicircle, where L is the distance between the injector and collector.  $A_1(T)$  can be expressed as

$$A_1(T) = A_1(T_{\text{base}}) e^{-\frac{\pi L}{2v_{\text{F}}\tau_{\text{eff}}}},$$
(2.93)

with an effective scattering time  $\tau_{\rm eff}$ . In experiments,  $\tau_{\rm eff}$  can be extracted as

$$\tau_{\rm eff}(T) = -\frac{\pi L}{2v_{\rm F}} / \ln\left(\frac{A_1(T)}{A_1(T_{\rm base})}\right).$$
(2.94)

If the temperature-dependent scattering mechanism is mainly scattering on acoustic phonons,  $\tau_{\text{eff}}^{-1}(T)$  depends linearly on T[180, 232]. If the electron-electron scattering is the main source of the loss of TMF signal, quadratic T dependence is expected[233, 234].

# 3. Chapter

# Experimental methods

In this chapter, experimental methods that are important to this thesis are presented. The first part is devoted to the sample fabrication including the fabrication of van der Waals (vdW) heterostructures and electrical contacts to them. The second part focuses on electrical transport measurements including the measurement set-up, the method of applying pressure, and the background of some important measurement techniques which is used in the following chapters.

## 3.1 Sample fabrication

Since many of the investigated phenomena are only observable on samples with large mobilities, it is very important to fabricate samples with high quality. In this section, I introduce the fabrication method I have used to make devices including the exfoliation of crystals which are used to assemble VdW heterostructures.

## 3.1.1 Building a vdW heterostructure



Figure 3.1: Optical microscope image of exfoliated (a) graphene, (b) hBN, and (c)  $WSe_2$  crystals on  $Si/SiO_2$  wafers. (a) The SLG, BLG, and thicker graphite flakes are well distinguishable from each other.

A vdW heterostructure is made of different layered crystals and held together by

the van der Waals interaction. The initial step to build them is the preparation of starting materials. There are various methods to synthesize 2D crystals such as chemical vapor deposition (CVD), and mechanical and liquid phase exfoliation[5]. Among them, mechanical exfoliation provides one of the highest quality flakes. However, the yield is far worse than CVD[241]. During the exfoliation, a low adhesive tape<sup>1</sup> is used as a transfer medium: the top layers of the bulk are peeled off with it and transferred to a Si/SiO<sub>2</sub> substrate. The exfoliated flakes are inspected on the wafer with an optical microscope. In Fig. 3.1 examples of exfoliated graphene, hBN, and WSe<sub>2</sub> are shown. The visibility of the flakes depends on their thickness and the substrate[242]. From the color of the flakes, their thickness can be estimated. To distinguish SLG and BLG from each other and thicker graphite flakes, their contrast can be used[243–245].

#### vdW stacking



Figure 3.2: Schematic representation of the process of dry stacking of vdW heterostructures. (a) A PC/PDMS stamp is prepared on a glass slide and used to pick up the first layer (hBN) by placing the stamp on the wafer which is heated to 60-100 °C. (b) Pickup of the second layer (graphene) with the top layer. (c) Stamping the finished stack onto an empty wafer and melting the PC onto the substrate.

In general, any kind of layered materials can be placed on top of each other (which is called stacking for simplicity) under a transfer stage[121, 246]. The process is illustrated in Fig 3.2. It involves using transparent polymers on a glass slide: a polydimethylsiloxane (PDMS) piece is put on a glass slide and a polycarbonate (PC) film is placed on top of it. This stamp is mounted on an XYZ-stage under the transfer microscope (in the figure, it is illustrated with the PC and PDMS on a glass slide). Then the wafer (SiO<sub>2</sub> in the figure) with the flake intended as the top layer is placed on the xy-stage of the microscope. A clean region of the PC layer is pushed to the surface of the wafer at the position of the chosen flake. Then the stage is heated up

<sup>&</sup>lt;sup>1</sup>Eg. Nitto ELP-BT-150P-LC

to 100 °C, which increases the adhesion between the polymer and the flake. After the wafer is heated up, the stamp is slowly retracted while the flake is peeled off from the substrate. Then the wafer with the flake intended as the second layer is placed on the stage and the flake is picked up by the layer on the stamp. This process continues until all the desired crystals are picked up and then the whole heterostructure is stamped on a wafer. It is heated to 180 °C to melt the PC on the wafer. The stamp is slowly retracted and the PC with the heterostructure remains on the substrate. After this, the heterostructure is annealed at 180 °C for 5 minutes to increase the adhesion between the substrate and the heterostructure. Heating and stacking at large temperatures also help to reduce the amount of contamination in the samples[130, 135]. Afterwards, the PC is dissolved in chloroform and the heterostructure is ready to be made into a device for transport measurements.

#### Characterization of the heterostructures

To have a high-quality sample, it is essential to perform the following studies before further fabrication steps on the heterostructure. The most widely used noninvasive methods are Raman spectroscopy and atomic force microscopy (AFM). With Raman spectroscopy for example nanometre-scale strain variations can be spatially resolved[247], which limits the mobility in high-quality graphene samples[163].

The AFM is a type of scanning probe microscope. The tip is moved by piezoelectric transducers and it is mounted on a cantilever which gets deflected when it interacts with the sample. The deflection is measured optically: a laser beam gets reflected from the cantilever to a four-segment photodetector. From the deflection, feedback control is used to keep the force or the distance between the tip and the surface[248]. The AFM can be used to determine the accurate thickness of the layers and to map the surface topology of the heterostructure. In Fig. 3.3 an optical microscope image and an AFM image are shown on the same heterostructure. The darker spots in the optical image are contamination between the layers, probably organic residuals. These along with a lot of smaller ones are more pronounced in Fig. 3.3b. Two bubble-free regions are circled in the figure, which can be used later in the fabrication. The reason behind avoiding the bubbles is that they induce local strain fluctuations[249, 250] which lead to lower mobility.

### 3.1.2 Making an electronic device

Electronic devices made from heterostructures are usually made by the following steps: the sample geometry is chosen to be suitable for the application. Then electrical contacts are made by lithography and the sample is shaped in the designed geometry.



Figure 3.3: Comparison of an optical microscope and an AFM image of a heterostructure. (a) Optical microscope image of a heterostructure. (b) AFM image of the same heterostructure. The white circles are bubbles of contamination between the layers. The black-circled regions are contamination-free and could be used to fabricate electrical devices.

#### Designing the sample

The samples made for transport measurements are most commonly made into Hall bar or Corbino geometry as well as two-terminal geometries[251]. The Corbino geometry is a cylindrical symmetric configuration where the sample is the annulus between the inner and outer contacts as shown in Fig. 3.4a. In this configuration  $\sigma_{xx}$  is measured as the radial part of the electric field is responsible for the measured current. As there are no sample edges between the contacts, the bulk response dominates and good electrical contact is not as important as in Hall bar geometries[252] to observe eg. fractional QHE[253]. On the other hand, the Hall bar geometry allows the measurements of  $\rho_{xx}^2$  and  $\rho_{xy}^3$  simultaneously by measuring the longitudinal and transverse Hall voltage on the terminals shown in Fig. 3.4b. This is the most widely used geometry, and the measurements presented in this thesis are also performed in this configuration.

After the heterostructure is finished, a clean region and the geometry is chosen. A design for the next steps is made by a vector graphics software<sup>4</sup>. A design of a device is shown in Fig. 3.5a. In the figure, the differently colored polygons belong to different lithography steps including contact evaporation and etching of the sample, which are described in the next paragraphs.

<sup>&</sup>lt;sup>2</sup>From the longitudinal voltage  $(V_{xx})$ , the measured current (I) and the geometrical parameters, the width W and the length L of the sample  $\rho_{xx} = W/L \cdot V_{xx}/I$ .

<sup>&</sup>lt;sup>3</sup>From the transverse voltage  $(V_{xy})$  and the measured current  $(I) \rho_{xy} = V_{xy}/I$ .

<sup>&</sup>lt;sup>4</sup>Eg. Adobe Illustrator, Inkscape, or Autodesk AutoCAD



Figure 3.4: Schematic device geometries. (a) Corbino geometry. The grey region is the sample and the conductance can be measured between the two yellow contacts. (b) Hall bar geometry. The sample is formed into the gray area and longitudinal voltage  $V_{xx}$  and transverse voltage  $V_{xy}$  can be measured at once. The sample width is W and the length between the side arms of the Hall bar is L.



Figure 3.5: Designing a sample. (a) The yellow polygons outlined in black are the areas where electrical contacts are to be fabricated, the red ones are the areas where the crystals are planned to be etched away, and the darker yellow areas are where the top gate is to be placed. (b) Almost completed device: the contacts are finished, and the device is shaped, but the top gate is yet to be fabricated.

#### Electron beam lithography

Electron beam lithography (EBL) offers a very high resolution (up to 10 nm) nanofabrication method and it is very flexible, as the electrons are deflected to expose a mask and create any custom design[254]. Thanks to these properties, this is one of the most commonly used patterning methods in the field of 2D materials. Before the lithography, the wafer is coated with a resist. During the lithography, the resist gets irradiated at a designed pattern. After this, the resist is developed, which means that

the regions that were exposed to the electron beam are removed for positive resist and the opposite happens for negative resist.

After the pattern is made in the resist, it can be used in various ways: the sample can be shaped using reactive ion etching, or contacts for electrical measurements can be made for electrical measurements. An example is given in Fig. 3.5: in panel (a) the colored polygons are the design of different lithography steps: the outer yellow ones are directly evaporated by Cr/Au in Fig. 3.5b, the inner yellow ones were first etched (to access the graphene) then evaporated to make edge contacts[121]. The brown one in the bottom left is etched till the bottom graphite is reached to contact it as a bottom gate. The red ones are the regions that were etched completely to achieve a Hall bar shape. The fabrication recipes I used in the samples presented in this thesis are given in Appendix A.1.

## 3.2 Transport measurement

Certain phenomena, such as weak localization and SdH oscillations presented in the previous chapter, are only observable at low temperatures. In this thesis, the presented experiments were carried out in different cryogenic measurement set-ups in which the base temperature is 35 mK, 1.5 K or 4.2 K. In Helium-4 cryostats, the dewar<sup>5</sup> is filled with liquid <sup>4</sup>He, for which boiling point is at 4.2 K at atmospheric pressure, and cryogenic liquid is directly used to cool down the sample. If a variable temperature insert (VTI) is placed in the dewar, then by letting in liquid He and pumping the volume of the VTI, the latent heat is removed by evaporation, and temperatures as low as 1.5 K can be reached thanks to the small pressure and the large vapor pressure<sup>6</sup> of <sup>4</sup>He. In a VTI a heater is also present allowing to control the temperature between 1.5 K to 300 K. To achieve 35 mK, a <sup>3</sup>He/<sup>4</sup>He dilution refrigerator is used, where the cooling power comes from dilution of <sup>3</sup>He from the concentrated phase to the diluted phase[255]. The 4.2 K cryostats is equipped with a 15 T out-of-plane magnet, one of the Helium-4 cryostat has a 8 T magnet, and the other one has a 9/3 T vector magnet.

Electronic measurements were carried out using standard lock-in techniques to measure the differential conductance and resistance. Low-noise and low-drift current and voltage amplifiers were used to amplify the signal. DC voltage sources were used on the gates. All measurements were controlled with our measurement codes based on QCoDeS[256], which is a Python-based data acquisition framework.

<sup>&</sup>lt;sup>5</sup>A double-walled vacuum isolation vessel.

 $<sup>^{6}</sup>$ At 10 mbar the boiling point of liquid  $^{4}$ He is around 1.67 K.

In the rest of this section, the effect of gating on heterostructures is introduced, and a useful method of determining the band gap of the sample.

### 3.2.1 Electrical gating

The electrochemical potential and thus the charge carrier density n in a field effect transistor can be tuned by applying voltage on a capacitively coupled gate electrode[210]. Similarly, in graphene, the gate voltage also tunes n[4]. In BLG, as a result of gate voltage, a potential difference can be formed between the layers, and a band gap opens[219, 257, 258] as it was shown in Section 2.1.2. Experimentally, this can be well controlled with a dual-gated geometry as in this case the charge density (n) and electric displacement field (D) are related to the top and bottom gate voltage by

$$n = \alpha_{\rm TG} V_{\rm TG} + \alpha_{\rm BG} V_{\rm BG} + n_0$$

$$\frac{D}{\epsilon_0} = \frac{e}{2\epsilon_0} \left( \alpha_{\rm TG} V_{\rm TG} - \alpha_{\rm BG} V_{\rm BG} \right) + \frac{D_0}{\epsilon_0},$$
(3.1)

where  $\alpha_{\rm TG}$  and  $\alpha_{\rm BG}$  are the lever arms<sup>7</sup> for the top and bottom gate respectively,  $\epsilon_0$  is the vacuum permittivity,  $V_{\rm TG}$  and  $V_{\rm BG}$  are the top and bottom gate voltage, respectively[259, 260].  $n_0 = -\alpha_{\rm TG}V_{\rm TG0} - \alpha_{\rm BG}V_{\rm BG0}$  is the carrier density when the gate voltages are set to zero, whereas  $D_0 = -\frac{e}{2} (\alpha_{\rm TG}V_{\rm TG0} - \alpha_{\rm BG}V_{\rm BG0})$  is a built-in offset electric field.  $V_{\rm TG0}$  and  $V_{\rm BG0}$  are the values of the top and bottom gate at the zero density and zero displacement field point.

The procedure to obtain the lever arms can be done by measuring the resistivity  $\rho_{xx}$  as a function of the gate voltages and magnetic field. For example, the ratio of the lever arms  $s = \alpha_{\text{TG}}/\alpha_{\text{BG}}$  in dual-gated samples can be obtained from the slope of the charge neutrality line (CNL) at n = 0. A typical  $\rho_{xx}$  measurement as a function of gate voltages is plotted in Fig 3.6a. Here, the high resistance region corresponds to the CNL, and the slope of the dashed line which shows the direction of D is the ratio between the lever arms. After obtaining the ratios, for BLG,  $n_0$  and  $D_0$  can be obtained from the solution of the CNL and from the minimum resistance along the CNL. After these actions, the charge carrier density can be tuned with the gates. Using the quantum Hall effect, which is described in more detail in Section 2.5.3, n can be easily calculated as at certain magnetic fields B, the resistance  $\rho_{xx}$  shows minima and the charge carrier

<sup>&</sup>lt;sup>7</sup>In a parallel-plate capacitor model,  $\alpha = \epsilon/ed$ , where  $\epsilon$  is the dielectric constant of the oxide between the graphene and the gate electrode and d is the thickness of the oxide.

density can be calculated from the LL filling as

$$n = \frac{eB\nu}{h},\tag{3.2}$$

where  $\nu$  is the LL filling factor. The absolute value of the lever arms can be obtained as shown in Fig. 6.2 or Fig. 5.5. In these figures, the quantum Hall features at integer LL filling (constant integer  $\nu$ ) can be used to scale the *x*-axis according to Eq.(3.2).



Figure 3.6: Conversion of the gate voltages to n and D. (a)  $\rho_{xx}$  as a function of the top and bottom gate voltages of a BLG device. The high resistance region is the CNL. The dashed white lines show the direction of increasing n and D. (b) The same as (a) but plotted as a function of n and D.

#### **3.2.2** Thermal activation measurements

At high temperatures, semiconductors, insulators and also BLG possess a conductivity even when the chemical potential is in the gap due to the thermal activation of charge carriers across the gap ( $\Delta$ ) between the valence and conduction bands[261]. At small temperatures, where the intrinsic charge carrier concentration is smaller than the carrier concentration induced by impurities, the conduction is determined by the impurities. The conduction at small temperatures occurs by the hopping of electrons through localized states of impurities, which is called nearest neighbor hopping (NNH). At even smaller temperatures, the conduction happens via electron-assisted hopping through localized states far from each other, which is called variable-range hopping (VRH)[261]. These processes act as parallel conduction channels, and the temperature dependence of the conductivity can usually be well described with

$$\sigma_{xx} = \rho_1^{-1} e^{-\frac{\Delta}{2k_{\rm B}T}} + \rho_2^{-1} e^{\left(-\frac{T_1}{T}\right)} + \rho_3^{-1} e^{\left(-\frac{T_{\alpha}}{T}\right)^{\alpha}},\tag{3.3}$$

where  $\rho_i$  are constant prefactors and  $T_i$  are phenomenological constants. The resistance is plotted in Fig. 3.7: at large temperatures, the first term of Eq.(3.3) dominates, which is the Arrhenius equation that corresponds to contributions from thermal activation of the band gap  $(\Delta)^8$ . At intermediate temperatures, the second term of Eq.(3.3) dominates, when conduction happens through the NNH of carriers within the defect band. The third term of Eq.(3.3) comes from VRH[262] and dominates at low temperatures.  $\alpha$  in VRH is less than 1 and it is expected to be  $\alpha = 1/3$  in two dimensions like in BLG near the CNP[260, 263, 264].

At high temperatures, thermal activation dominates. As the thermal activation is the main contribution to the conductance at high temperatures, the gap energy can be determined from the T dependence of the resistivity at high temperatures by a linear fit of 1/T as

Δ

$$\ln(\rho_{xx}) \propto \frac{1}{2k_{\rm B}T}.$$
(3.4)



Figure 3.7: Simulated thermal activation of a gap. (a) Resistance as a function of the temperature using the parameters of  $\rho_1 = 0.01 \Omega$ ,  $\rho_2 = 10 \Omega$  and  $\rho_3 = 100 \Omega$  with  $\Delta = 50 \text{ meV}$ ,  $T_1 = 29 \text{ K}$  and  $T_2 = 3 \text{ K}$  for the blue and  $\Delta = 100 \text{ meV}$ ,  $T_1 = 58 \text{ K}$  and  $T_2 = 8 \text{ K}$  for the red curves, respectively. (b) Arrhenius plot of resistance as a function of 1/T.

(0, 1)

<sup>&</sup>lt;sup>8</sup>The activation energy is usually larger than the real gap due to the disorder-induced localization[227].

## 3.3 Pressure-dependent transport measurement

In layered materials, like a vdW heterostructure, the interlayer interactions play an essential role in determining their properties. The strength of these interactions depends vastly on the interlayer distance, thus the properties of layered materials can be tuned by varying the interlayer distance[12]. Application of hydrostatic pressure on a solid leads to compression, i.e. interlayer distance can be decreased by pressing a layered material. As the constituting particles get closer to each other, the overlap of the electron wavefunction increases, which leads to a change in the band structure. Besides the decrease of the lattice constants, the pressure can induce a structural phase transition. For example, with pressure, the NaCl crystal with face-centered cubic structure can change to a primitive cubic lattice like that of CsCl[265]. In the field of vdW materials, pressure is a very important experimental tuning knob for tuning the strength of the proximity effect between the layers.



Figure 3.8: (a) Schematic view of the pressure cell. 1. CuBe upper screw. 2. WC push rod. 3. WC piston backup. 4. NiCrAl/CuBe double-layered cell wall. 5. WC piston. 6. Teflon cup. 7. CuBe plug. (b) Optical image of the cell. (c) Optical image of the sample holder plug.

Pressure is usually applied with commercial pressure cells, however, to apply it on a nanodevice and perform electrical measurements a specially designed cell is necessary. The design of our pressure cell is described in Ref. [266] and its simplified schematic is shown in Fig. 3.8. In short, it is a piston-cylinder pressure cell with a sample holder plug with a PCB that allows us to bond the sample to connect it to the measurement

instruments. Its small size, with its diameter of 25 mm (Fig. 3.8b), allows us to put it in all of our cryostats to make low-temperature measurements.

The process of applying pressure is the following: after the device is fixed to the PCB on the plug (7 in Fig. 3.8a), which is shown in Fig. 3.8c, the sample is bonded. Then a Teflon cup (6 in Fig. 3.8a) is filled with a pressure mediating special inert hydraulic oil<sup>9</sup> and it is placed on the plug. This structure is placed in the pressure cell as shown schematically in Fig. 3.8 and the cell is assembled. The cell is placed into a hydraulic press and the piston is pressed with the help of a pushing rod (2 in Fig. 3.8a). When the desired pressure is reached, the screw on the top is tightened which holds the applied pressure in the cell. This procedure is done at room temperature. To change the pressure applied to the sample, it has to be warmed up to room temperature.

<sup>&</sup>lt;sup>9</sup>We use Daphne 7373 kerosene compound.

# 4. Chapter

# Modeling graphene heterostructures

In this chapter, modeling of different heterostructures is presented including the effect of pressure. Firstly, the effect of a transition metal dichalcogenide (TMD) on BLG is modeled. Secondly, the calculation of the band structure of twisted double bilayer graphene (TBDG) is discussed.

## 4.1 SOC in $BLG/WSe_2$ heterostructures

Among the TMDs, the SOC in the valence band of  $WSe_2$  is one of the strongest[267], and the induced SOC strength that is inherited in graphene is also one of the largest[107]. The effect of  $WSe_2$  on the band structure of graphene is described in Section 2.3. Here, the influence of the SOC on the electric field-induced gap and the Landau levels is described.



Figure 4.1: Band gap at the CNP in bilayer graphene with  $\lambda_{\rm R} = 0$  and different Isingtype SOC strengths. The black line corresponds to the pristine BLG. The blue line corresponds to a BLG/WSe<sub>2</sub> heterostructure. And the red and green lines correspond to a WSe<sub>2</sub>/BLG/WSe<sub>2</sub> device.

The electric field opens a gap in BLG, which can be modified with SOC as illustrated in Fig. 4.1. Without SOC in a pristine BLG, a gap opens and increases linearly with |u| (black line). In a BLG/WSe<sub>2</sub> heterostructure, where  $\lambda_{\rm I}^{\rm b} \neq 0$ , but  $\lambda_{\rm I}^{\rm t} = 0$ , the gap opens if  $|u| > \lambda_{\rm I}^{\rm b}/2$  (blue line). Otherwise, if |u| is smaller a correlated insulating phase can form[268, 269]. In a WSe<sub>2</sub>/BLG/WSe<sub>2</sub> device, depending on the twist angle, the relative sign of the SOC strength can be the same or the opposite. If both are positive (or negative), then a gap opens if  $|u| > |\lambda_{\rm I}^{\rm b} + \lambda_{\rm I}^{\rm t}|/2$  (red line). On the other hand, if their sign is the opposite, there is already a gap present at u = 0, which closes at  $|u| = (|\lambda_{\rm I}^{\rm b}| - |\lambda_{\rm I}^{\rm t}|)/2$  (green line). At larger |u|, a new gap opens similarly to the other cases. This is experimentally studied in Chapter 6.

### 4.1.1 Landau levels in BLG

The SOC also modifies the Landau level (LL) spectrum in BLG. To calculate the LL spectrum in a large out-of-plane magnetic field, I follow the footsteps of Ref. [270]. The  $\gamma_3$  hopping is neglected, which would introduce a small mixing of the LLs at small magnetic fields[219]. The ladder operators are introduced as  $\hat{a} = \frac{l_B}{2}(q_x + iq_y)$ ,  $\hat{a}^{\dagger} = \frac{l_B}{2}(q_x - iq_y)$ , which can be written back to the original Hamiltonian in Eq.(2.10). In the ladder operators,  $l_B = \sqrt{\hbar/e|B|}$  is the magnetic length and  $q_i = k_i - e/\hbar A_i$  is the canonical impulse with the vector potential  $A_i$ . In valley K,  $\pi = \sqrt{2eB\hbar}\hat{a}^{\dagger}$ , which can be substituted in the Hamiltonian as

$$H_{\rm BLG}^{K} = \begin{pmatrix} u & v_0 \sqrt{2eB\hbar}\hat{a}^{\dagger} & -v_4 \sqrt{2eB\hbar}\hat{a}^{\dagger} & 0\\ v_0 \sqrt{2eB\hbar}\hat{a} & u + \Delta' & \gamma_1 & -v_4 \sqrt{2eB\hbar}\hat{a}^{\dagger}\\ -v_4 \sqrt{2eB\hbar}\hat{a} & \gamma_1 & -u + \Delta' & v_0 \sqrt{2eB\hbar}\hat{a}^{\dagger}\\ 0 & -v_4 \sqrt{2eB\hbar}\hat{a} & v_0 \sqrt{2eB\hbar}\hat{a} & -u \end{pmatrix} \otimes s_0. \quad (4.1)$$

In the K' valley,  $\pi = -\sqrt{2eB\hbar}\hat{a}^{\dagger}$  and  $\pi^{\dagger} = -\sqrt{2eB\hbar}\hat{a}$  and the Hamiltonian can be written as

$$H_{\rm BLG}^{K'} = \begin{pmatrix} u & -v_0\sqrt{2eB\hbar}\hat{a} & v_4\sqrt{2eB\hbar}\hat{a} & 0\\ -v_0\sqrt{2eB\hbar}\hat{a}^{\dagger} & u + \Delta' & \gamma_1 & v_4\sqrt{2eB\hbar}\hat{a}\\ v_4\sqrt{2eB\hbar}\hat{a}^{\dagger} & \gamma_1 & -u + \Delta' & -v_0\sqrt{2eB\hbar}\hat{a}\\ 0 & v_4\sqrt{2eB\hbar}\hat{a}^{\dagger} & -v_0\sqrt{2eB\hbar}\hat{a}^{\dagger} & -u \end{pmatrix} \otimes s_0. \quad (4.2)$$

The magnetic field also leads to spin splitting, which introduces an additional term, the Zeeman term  $H_Z = E_Z s_z$  in Eq.(2.43), where  $E_Z = -\mu_B B$  is the Zeeman energy with the Bohr magneton  $\mu_B$ . The eigenenergies  $E_{\xi,n,s_z}$  of  $H = H_{\text{BLG}} + H_{\text{SOC}} + H_Z$  are defined as

$$H|\xi, n, s_z\rangle = E_{\xi, n, s_z}|\xi, n, s_z\rangle, \qquad (4.3)$$

with the eigenstates of  $|\xi, n, s_z\rangle$ , where  $n = 0, 1, \ldots$  is the orbital number. The single-

particle LL energies can be obtained from the matrices derived from Eq.(4.3) in Appendix B.

#### $BLG/WSe_2$ heterostructures



Figure 4.2: Landau levels in BLG. (a) Schematic representation of the real-space localization of the LL orbits. They can be represented with 3 quantum numbers: the valley (+/-), the orbital number (n = 0, 1...) and the spin  $(\uparrow / \downarrow)$ . The + orbits are localized at the bottom and the – are localized at the top layer of the BLG. As the WSe<sub>2</sub> is at the bottom, the SOC only modifies the + orbit energies. The n = 1 LLs with the same valley index are localized on the same layer as the n = 0 LLs (solid lines in b and c). (b) and (c) Calculated LL energies as a function of the interlayer potential difference (u) with (b)  $\lambda_{\rm I} = 0$  and (c)  $\lambda_{\rm I} = 1.6$  meV at B = 8 T. The values of the hopping parameters are given in Section 2.1.2. The  $\nu = \pm 3$  crossing points are marked with a red and a blue dot and are found at  $u = u_{\pm 3}^*$ . The arrows with the letter p-s next to them show how the LL energies change by increasing pressure i.e. increasing the  $\lambda_{\rm I}$  in (c). The LL filling factors are shown with black numbers between LLs.

If all the parameters except  $\gamma_0$  and  $\gamma_1$  are set to zero, the first 8 LLs (zeroth LLs) are degenerate (spin, valley and n = 0, 1 orbital degeneracy) with zero energy (see Eq.(2.87)). The other parameters lift this degeneracy. Since for the low-energy bands, only A1 and B2 atoms (non-dimer sites) play a role, the layer and the pseudo-spin (sublattice) degree of freedom become identical. Moreover, the lowest LLs from the K valley (+) are localized on the bottom layer, while the lowest LLs with K' index (-) are localized on the top layer, which is illustrated in Fig. 4.2a. As a result, the LL energies of different valleys depend oppositely on the application of the electric field. In Fig. 4.2b the LLs are shown if  $\lambda_{\rm I} = 0$  at B = 8 T. In this case, the LLs are spin split due to the Zeeman term and also split in the orbital index due to  $\gamma_4$  and  $\Delta'$ . A finite u further splits these LLs and their energy is linear in u. In the figure, the LL filling factors  $\nu$  with numbers between the LLs are also shown. There are several crossings observable between the LLs, from which the LL crossing points at  $\nu = \pm 3$  are marked with red and blue dots. The position of these crossings without SOC remains at u = 0 independently of the applied magnetic field. These crossings are interesting since their position is unaffected by the electronic interactions[51] and their position is strongly affected by the VZ interaction. This is illustrated in Fig. 4.2c by including  $\lambda_{\rm I}^{\rm b} = 1.6 \,\mathrm{meV}$  in a geometry shown in Fig. 4.2a. In this geometry, as the SOC has an effect only at the bottom layer, it affects only the LLs with + index. Since the Ising-type SOC behaves as an effective *B*-field, as we increase  $\lambda_{\rm I}$ , LL energies of the bottom layer (indicated by the solid lines in Fig. 4.2b) shift according to their spin polarization, as indicated by the arrows with letter p-s next to them in the figure. The effect of  $\lambda_{\rm R}$  is negligible on these crossings, and only the Ising-type SOC has a measurable effect on the lowest LLs[270].

As the SOC also modifies the LL spectrum, it allows the extraction of the Ising term[50, 271, 272] in BLG/TMD heterostructures. For this, the magnetic field dependence of the  $\nu = \pm 3$  crossings can be calculated as shown in Fig. 4.3a, which I also measured on my devices and it is discussed in Chapter 6. At small magnetic fields, the Ising-type SOC is stronger than the Zeeman splitting, and  $u_3^*$  is negative. The LLs with spin  $\uparrow$  in K valley have larger energy than the LLs indexed with spin  $\downarrow$  as opposed to the LLs in the K' valley (Fig. 4.3b). The external magnetic field, as increased, compensates the SOC at  $2g\mu_B B \approx \lambda_1^{\rm b}$ ,  $u_3^* = u_{-3}^* \approx 0$  as shown in Fig. 4.3c. At larger B fields,  $u_3^* > 0$  as shown in Fig. 4.2c. By further increasing B, near  $4g\mu_B B \approx \lambda_1^{\rm b}$ the slope changes in Fig. 4.3a and at this magnetic field, the LLs with spin  $\uparrow$  in K valley have the same energy as the LLs indexed with spin  $\downarrow$ . At larger magnetic fields, these LLs change position and the LLs that cross at  $\nu = \pm 3$  have the same spin and orbital number but different valleys as opposed to smaller magnetic fields. As shown in Chapter 6, I have used these crossings to extract the pressure tuning of the Ising-type SOC coupling strength[273].

#### $WSe_2/BLG/WSe_2$ heterostructures

In a TMD/BLG/TMD heterostructure, the LLs indexed with K are influenced by the induced SOC from the bottom TMD, and the LLs indexed with K' depend on the SOC induced from the top TMD. If the induced SOC is the opposite in the layers  $(\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t})$ , the spectrum is very similar to the spectrum of a pristine BLG without SOC. The  $\nu = \pm 3$  crossing points are at  $u_{\pm 3}^* = 0$  and they are independent of B. On the other hand, the positions of the other crossing points are different compared to both the case of without SOC and in the case of induced SOC only at one layer.

When  $\lambda_{\rm I}^{\rm t}$  and  $\lambda_{\rm I}^{\rm b}$  have an opposite sign but their magnitudes are different, the u, -u symmetry is lost, leading to non-zero  $\pm u_3^*$  crossings as shown in Fig. 4.4b with  $\lambda_{\rm I}^{\rm b} = 1.2 \,\text{meV}$  and  $\lambda_{\rm I}^{\rm t} = -0.8 \,\text{meV}$ . In this case,  $u_{\pm 3}^*$  depend on B as shown with solid



Figure 4.3: (a)  $u_{\pm 3}^*$  magnetic field dependence with  $\lambda_{\rm I}^{\rm b} = 1 \,\text{meV}$  (dashdotted lines) and  $\lambda_{\rm I}^{\rm b} = 1.5 \,\text{meV}$  (solid lines). The  $u_{\pm 3}^*$  lines cross at  $2g\mu_B B_{comp} \approx \lambda_{\rm I}$ . (b) and (c) Calculated LL energies as a function of the interlayer potential difference (u) with  $\lambda_{\rm I}^{\rm b} = 1.5 \,\text{meV}$  at (b)  $B = 4 \,\text{T}$  and (c) at  $B = B_{comp} = 6.188 \,\text{T}$ . The  $\nu = \pm 3$  crossing points are marked with a red and a blue dot.



Figure 4.4: LLs in a TMD/BLG/TMD heterostructure. (a) and (b) Calculated LL energies as a function of the interlayer potential difference (u) at B = 5 T with (a)  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} = 1 \text{ meV}$  and (b)  $\lambda_{\rm I}^{\rm b} = 1.2 \text{ meV}$  and  $\lambda_{\rm I}^{\rm t} = -0.8 \text{ meV}$ . The  $\nu = \pm 3$  crossing points are marked with a red and a blue dot and are found at  $u = u_{\pm 3}^*$ . (c) Magnetic field dependence of  $u_{\pm 3}^*$  with  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t} = 1 \text{ meV}$  (dashdotted lines) and  $\lambda_{\rm I}^{\rm b} = 1.2 \text{ meV}$  and  $\lambda_{\rm I}^{\rm t} = -0.8 \text{ meV}$ .

lines in Fig. 4.4c. Comparing this scenario with the *B* dependence of the crossings in a BLG/TMD heterostructure, which is shown 4.3a, the tendency differs. In this case,  $u_{\pm 3}^*$  are more or less constant with *B* with  $|u_3^* - u_{-3}^*| \sim |\lambda_I^b - \lambda_I^t|$  and near  $4g\mu_B B \approx |\lambda_I^b - \lambda_I^t|/2$  they change sign.  $u_{\pm 3}^*$  in the symmetric case of  $\lambda_I^b = \lambda_I^t$ , which is shown with dashdotted lines in Fig. 4.4c, shows similar tendency as in a BLG/TMD heterostructure, but there's no crossing at  $2g\mu_B B \approx |\lambda_I^b + \lambda_I^t|/2$ .

The magnetic field dependence of the other crossings can also be calculated. In Fig. 4.5 and Fig. 4.6, the Landau level crossings are plotted, with SOC and without



Figure 4.5: Landau level crossings at  $\nu = 0$  as a function of B with (a)  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} = -5.6 \,\mathrm{meV}$ , (b)  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} = -2 \,\mathrm{meV}$  and (c)  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t} = 0$ .

SOC, for  $\nu = 0$  and  $\nu = \pm 1$ , respectively. Without SOC the crossings go to zero as  $B \to 0$  as opposed to the case of  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} \neq 0$ . Comparing these figures with the measurements discussed in Section 6.2, the experiments show similarities to the model: the  $\nu = 1$  crossings show a similar tendency, so do the higher  $u_0^*$  branches in the  $\nu = 0$  crossings. The discrepancy between the model, which is calculated in a single-particle picture, and the experiment comes from the fact that in the model electron-electron interactions are neglected [51].



Figure 4.6: Landau level crossings at  $\nu = \pm 1$  as a function of B with (a)  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} = -5.6 \,\mathrm{meV}$ , (b)  $\lambda_{\rm I}^{\rm b} = -\lambda_{\rm I}^{\rm t} = -2 \,\mathrm{meV}$  and with (c)  $\lambda_{\rm I}^{\rm b} = \lambda_{\rm I}^{\rm t} = 0$ .

### 4.1.2 Fermi surface of BLG/TMD heterostructures

In the previous section, I showed that the LL spectroscopy can be used to extract  $\lambda_{\rm I}^{\rm b}$  from the LL crossings. Here, I show that from SdH oscillations, which is presented in Section 2.5.2,  $\lambda_{\rm R}^{\rm b}$  can also be obtained from the difference of the Fermi surfaces  $(dS_{\rm F})$ . From the period of the SdH oscillations, the spin-split Fermi surface  $S_{\rm F}^{(\sigma)}$  can be extracted.

To calculate  $S_{\rm F}^{(\sigma)}$ , where  $\sigma = \uparrow, \downarrow$  indexes the spin split bands, I start from the Hamiltonian of the BLG/TMD in Eq.(2.44). I calculate the dispersion relation  $E_{\sigma}(k)$  and numerically integrate the Fermi function near the K valley to obtain  $S_{\rm F}^{(\sigma)}$  as

$$S_{\rm F}^{(\sigma)} = \iint f(E_{\sigma}(k) - \mu) \mathrm{d}k^2, \qquad (4.4)$$

where  $\mu$  is the chemical potential and  $f(E_i - \mu)$  is the Fermi-Dirac distribution defined as

$$f(E - \mu) = \frac{1}{1 + e^{(E - \mu)/k_{\rm B}T}}.$$
(4.5)

The charge density n can be calculated similarly as

$$n = \frac{1}{2\pi^2} \sum_{\sigma} \iint f(E_{\sigma}(k) - \mu) dk^2 = \frac{1}{2\pi^2} \sum_{\sigma} S_{\rm F}^{(\sigma)}, \qquad (4.6)$$

where the valley degeneracy is included with a factor 2.



Figure 4.7: (a) Calculated Fermi surfaces of BLG/TMD heterostructure with  $\lambda_{\rm R}^{\rm b} = 10 \text{ meV}$ . The black dashed line denotes the degenerated case when  $S_{\rm F} = \pi^2 n$ . (b) Difference of the two spin split Fermi surface  $dS_{\rm F}$  as a function of n using  $\lambda_{\rm R}^{\rm b} = 10 \text{ meV}$  and  $\lambda_{\rm R}^{\rm b} = 20 \text{ meV}$ .

In the calculations,  $S_{\rm F}^{(\sigma)}$  and *n* are calculated as a function of  $\mu$ . Then  $\mu$  is converted to *n*. The calculated Fermi surface as a function of the charge density is shown in

Fig. 4.7a using  $\lambda_{\rm R}^{\rm b} = 10 \,{\rm meV}$ . The average of the two spin split  $S_{\rm F}$  is equal to the degenerate case, when  $\lambda_{\rm R}^{\rm b} = 0$ , which is shown in black dashed lines and equals  $S_{\rm F} = \pi^2 n$ . To visualize the splitting, the difference of the spin split Fermi surfaces  $dS_{\rm F} = |S_{\rm F}^{(1)} - S_{\rm F}^{(2)}|$  is shown in Fig. 4.7b for  $\lambda_{\rm R}^{\rm b} = 10 \,{\rm meV}$  and  $\lambda_{\rm R}^{\rm b} = 20 \,{\rm meV}$ : the larger  $\lambda_{\rm R}^{\rm b}$ , the larger the splitting. This is fitted on my experiments on BLG/WSe<sub>2</sub> heterostructures in Section 6.1.5.



## 4.2 Twisted double bilayer graphene

Figure 4.8: Illustration of the band structure of TDBG in a corner of the Brillouin zone: (a) No coupling between the top and bottom BLGs which are depicted with red and blue colors respectively, (b) a small coupling is introduced between the two BLGs which hybridizes the bands and leads to avoided crossings, (c) an external electric field can open a band gap at the Dirac points.

The twisted double bilayer graphene (TDBG) consists of two BLGs with a rotation angle of  $\vartheta$  between them. The rotation between the top and bottom BLG lattices, illustrated in Fig. 5.1b, leads also to a rotation between their Brillouin-zones (BZ) as it was shown in Section 2.2.1. These, along with the simplified BLG spectrum of each lattice, are presented in Fig. 4.8a with red and blue for the top and bottom bilayer, respectively. For small rotation angles, the spectrum of the bottom and top bilayers overlap. The coupling  $V_{int}$  between the closest monolayers of the BLGs hybridizes the bands as shown in Fig. 4.8b. Moreover, at magic twist angles the low-energy moiré bands become flat [69, 274–276] due to the strong interlayer coupling driven avoided crossings. Unlike TBG, in TDBG the electric field opens a bandgap as illustrated in Fig. 4.8c.

To model the TDBG, the BM model is applied, which is derived in Section 2.2.3. It is assumed that only the nearest graphene layers have non-negligible tunneling matrix elements between them. This is the case in the lowest order when only nearest neighbor hoppings are taken into account between the layers. This can be used to construct a continuum Hamiltonian from  $2 \times 2$  blocks encoding the BLG structure as these blocks are  $4 \times 4$  matrices due to the two sublattices. In Eq.(2.36) the diagonal terms are given by the Hamiltonian of BLG defined in Eq.(2.10). The coordinate system is chosen such that, the top layer is rotated with  $\vartheta/2$  counter-clockwise and the bottom layer is rotated with  $-\vartheta/2$ , i.e.  $\pi \to \pi e^{\pm i\vartheta/2}$ . In this representation Eq.(2.10) for the top layer is modified to

$$H^{t}(\mathbf{q},\vartheta) = \begin{pmatrix} \frac{u}{2} & v_{0}\pi^{\dagger}e^{-i\vartheta/2} & -v_{4}\pi^{\dagger}e^{-i\vartheta/2} & v_{3}\pi e^{i\vartheta/2} \\ v_{0}\pi e^{i\vartheta/2} & \frac{u}{2} + \Delta' & \gamma_{1} & -v_{4}\pi^{\dagger}e^{-i\vartheta/2} \\ -v_{4}\pi e^{i\vartheta/2} & \gamma_{1} & -\frac{u}{2} + \Delta' & v_{0}\pi^{\dagger}e^{-i\vartheta/2} \\ v_{3}\pi^{\dagger}e^{-i\vartheta/2} & -v_{4}\pi e^{i\vartheta/2} & v_{0}\pi e^{i\vartheta/2} & -\frac{u}{2} \end{pmatrix}$$
(4.7)

and the Hamiltonian for the bottom layer is given as  $H^{\rm b}(\mathbf{k},\vartheta) = H^{\rm t}(\mathbf{q},-\vartheta)$ , where  $\mathbf{q}$  is measured from the Dirac point of the top layer.

The BM model, with the Hamiltonian given in Eq.(2.36), is a tripod model in a sense that it connects a **q**-point on the top layer, which is measured from the Dirac points, with three other  $\mathbf{q} + \mathbf{q}_i$ -points of the bottom layer in the **k**-space, where  $\mathbf{q}_i$  are defined in Eq.(2.39), i.e. the Hamiltonian of the BM model is constructed in basis of  $\{|\mathbf{q}, t\rangle, |\mathbf{q} + \mathbf{q}_0, b\rangle, |\mathbf{q} + \mathbf{q}_+, b\rangle |\mathbf{q} + \mathbf{q}_-, b\rangle\}$ , where t and b stand for top and bottom layer and  $\mathbf{q}_i$ s are defined in Eq.(2.39). In BM model, there is tunneling only between the neighboring layers and in a matrix notation in the basis defined here the Hamiltonian can be written as

$$H(\mathbf{q}) = \begin{pmatrix} H^{t}(\mathbf{q},\vartheta) & T_{0} & T_{+} & T_{-} \\ T_{0}^{\dagger} & H^{b}(\mathbf{q} + \mathbf{q}_{0},\vartheta) & 0 & 0 \\ T_{+}^{\dagger} & 0 & H^{b}(\mathbf{q} + \mathbf{q}_{+},\vartheta) & 0 \\ T_{-}^{\dagger} & 0 & 0 & H^{b}(\mathbf{q} + \mathbf{q}_{-},\vartheta) \end{pmatrix}, \quad (4.8)$$

where the tunneling matrices between the top and bottom bilayer graphene are defined as

$$T_i = \begin{pmatrix} 0 & 0 \\ T_{\mathbf{q}_i} & 0 \end{pmatrix}, \tag{4.9}$$

where  $T_i$  is acting only between the upper layer of the BLG and the bottom layer of the top BLG and  $T_{\mathbf{q}_i}$  is defined in Eq.(2.38).

To calculate the band structure more conveniently, the coordinate system is shifted

to the center of the mBZ ( $\Gamma_M$ -point) by introducing

$$\mathbf{q}_{t} = \frac{\mathbf{b}_{1}^{(s)} + 2\mathbf{b}_{2}^{(s)}}{3} = \frac{8\pi}{3a}\sin(\vartheta/2)\left(\sqrt{3}/2, 1/2\right), \qquad (4.10)$$
$$\mathbf{q}_{b} = \frac{2\mathbf{b}_{1}^{(s)} + \mathbf{b}_{2}^{(s)}}{3} = \frac{8\pi}{3a}\sin(\vartheta/2)\left(\sqrt{3}/2, -1/2\right)$$

vectors in the **k**-space, where the reciprocal lattice vectors of the moiré lattice  $(\mathbf{b}_i^{(s)})$  are defined in Eq.(2.16) and the shifting is  $\mathbf{q} = \mathbf{q}' + \mathbf{q}_t$ , where  $\mathbf{q}'$  is measured from the Dirac points of the top layer. The direction of  $\mathbf{q}_{t,b}$  is shown in Fig. 4.9a. These vectors point to the  $K_M$  and  $K'_M$  points in the mBZ. In this notation, Eq.(4.8) can be rewritten as

$$H(\mathbf{q}) = \begin{pmatrix} H^{t}(\mathbf{q} - \mathbf{q}_{t}, \vartheta) & T_{0} & T_{+} & T_{-} \\ T_{0}^{\dagger} & H^{b}(\mathbf{q} - \mathbf{q}_{b}, \vartheta) & 0 & 0 \\ T_{+}^{\dagger} & 0 & H^{b}\left(\mathbf{q} - (\mathbf{q}_{b} + \mathbf{b}_{2}^{(s)}), \vartheta\right) & 0 \\ T_{-}^{\dagger} & 0 & 0 & H^{b}\left(\mathbf{q} - (\mathbf{q}_{b} - \mathbf{b}_{1}^{(s)}), \vartheta\right) \end{pmatrix}$$
(4.11)

Eq.(4.11) describes the matrix elements that couple to the top layer at  $\mathbf{q} - \mathbf{q}_t$ . However, the other points in the bottom layer couple other **k**-points of the top layer resulting in an infinitely large Hamiltonian, as illustrated graphically in Fig. 4.9a with red and blue points of the top and bottom layer, which are connected with the red, blue and black lines

blue and black lines. To visualize the construction of the full Hamiltonian, I introduce the basis of  $|(h,k),t/b\rangle = |\mathbf{q} + h\mathbf{b}_1^{(s)} + k\mathbf{b}_2^{(s)}, \text{top/bottom}\rangle$ , which goes through all the  $\mathbf{k} = \mathbf{q} + h\mathbf{b}_1^{(s)} + k\mathbf{b}_2^{(s)}$ -points which are coupled to  $\mathbf{q}$  with integers of h and k. In this basis, the BM model Hamiltonian in the second nearest neighbors approximation<sup>1</sup> in the  $\mathbf{k}$ -space takes the form of

(0,0),t angle	(0,-1),t angle	$ (1,0),t\rangle$	(1,1),t angle	(0,1),t angle	$ (-1,0),t\rangle$	(-1,-1),t angle	(0,0),b angle	(0,1),b angle	$ (-1,0),b\rangle$	
$H_t$	0	0	0	0	0	0	$T_0$	$T_+$	$T_{-}$	$\langle (0,0),t $
0	$H_t$	0	0	0	0	0	$T_{+}$	0	0	$\langle (0, -1), t  $
0	0	$H_t$	0	0	0	0	$T_{-}$	0	0	$\langle (1,0),t $
0	0	0	$H_t$	0	0	0	0	$T_{-}$	0	$\langle (1,1),t $
0	0	0	0	$H_t$	0	0	0	$T_0$	0	$\langle (0,1),t $
0	0	0	0	0	$H_t$	0	0	0	$T_0$	$\langle (-1, 0), t  $
0	0	0	0	0	0	$H_t$	0	0	$T_+$	$\langle (-1, -1), t  $
$T_0^{\dagger}$	$T^{\dagger}_{\pm}$	$T_{-}^{\dagger}$	0	0	0	0	$H_b$	0	0	$\langle (0,0),b $
$T_{+}^{\dagger}$	0	0	$T_{-}^{\dagger}$	$T_0^{\dagger}$	0	0	0	$H_b$	0	$\langle (0,1),b $
$T_{-}^{\dagger}$	0	0	0	0	$T_0^{\dagger}$	$T^{\dagger}_{+}$	0	0	$H_b$	$\langle (-1,0),b $

where the argument of the diagonal terms is  $\mathbf{q} - \mathbf{q}_{b/t} + h\mathbf{b}_1^{(s)} + k\mathbf{b}_2^{(s)}$ . The full Hamiltonian is constructed by considering more and more points that are connected via the Kronecker deltas in Eq.(2.37) as visualized in Fig. 4.9a. As the full Hamiltonian is infinitely large, during the calculation a cut-off momenta is defined such that the change of the low-energy levels is negligible by further increasing the size.

<sup>&</sup>lt;sup>1</sup>In a sense, that  $|(0,0),t\rangle$  is coupled to three  $|(h,k),b\rangle$ , which are coupled to two more  $|(h,k),t\rangle$  beside  $|(0,0),t\rangle$ .



Figure 4.9: (a) Illustration of the BM model showing the moiré reciprocal basis vectors and  $\mathbf{q}_{t,b}$ . The mBZ is shown with black dashed lines. The red and blue dots and the colored lines are the same as in Fig. 2.7. (b) mBZ showing the high-symmetry points. The blue dashed line shows the path in reciprocal space that is used in the figures shown later.

I implemented this model using Eq.(4.11) to construct the Hamiltonian and used it to calculate the band structure of TDBG. In the calculations I performed, the chosen momentum cut-off was the radius of  $4|\mathbf{b}_1^{(s)}|$  to obtain converged results. This implied Hamiltonian matrices with the size of  $648 \times 648$ .

During the calculation, the used parameter set for the interlayer matrix elements was calculated in Ref. [73] with ab initio EXX+RPA calculations with equilibrium distances, and the average of the sum over the three symmetric stacking configurations was taken to obtain

$$\omega = \omega_{AB'} = \omega_{BA'} \simeq \frac{H_{B,A'}(\mathbf{K})}{3}, \qquad (4.12)$$
$$\omega' = \omega_{AA'} = \omega_{BB'} \simeq \frac{H_{A,A'}(\mathbf{K})}{3}.$$

Using these values, the tunneling matrix elements can be written as

$$T_{\mathbf{q}_0} = \begin{pmatrix} \omega' & \omega \\ \omega & \omega' \end{pmatrix}, \qquad T_{\mathbf{q}_{\pm}} = e^{i\mathbf{b}_1'\tau} \begin{pmatrix} \omega' & \omega e^{\pm i2\pi/3} \\ \omega e^{\pm i2\pi/3} & \omega' \end{pmatrix}.$$
(4.13)

The  $\gamma_1$  hopping element is related to  $\omega$  by  $\gamma_1 = 3\omega$ . They also calculated the pressure

dependence of  $\omega$  and  $\omega'$  as

$$\omega(P) = 0.0546 + \sqrt{0.0044 + 0.0031 \cdot P}, \qquad (4.14)$$
  
$$\omega'(P) = 0.0561 + \sqrt{0.0018 + 0.0018 \cdot P},$$

where  $\omega$  is in eV and P is in GPa.

The calculated band structure along the blue line in Fig. 4.9b for the full model is shown in Fig. 4.10 at different angles at p = 0 GPa with red dotted lines and a minimal model without including the remote hopping terms ( $\gamma_3$  and  $\gamma_4$ ) are plotted with black lines. Without the remote hopping terms the bandwidth of the bands is smaller, electron-hole symmetric and the effect of trigonal warping is not present. At small angles, the two layers are strongly coupled, similar to TBG, and the bands are strongly hybridized. At the magic angle  $\vartheta_m \approx 1.05^\circ$  (Fig. 4.10b) the bandwidth of the bands near the Fermi level is the smallest as a function of  $\vartheta$  and almost completely flat in the minimal model. These bands are separated from the other dispersive bands with a gap  $\Delta_{\pm n_s}$ . At large angles (Fig. 4.10c), the layers are decoupled and the original parabolic dispersion of BLG is recovered near the  $K_M$  points, which is folded into the mBZ.



Figure 4.10: Band structure of twisted double bilayer graphene at p = 0 GPa at (a)  $\vartheta = 0.8^{\circ}$ , (b)  $\vartheta = 1.05^{\circ}$  and (c)  $\vartheta = 3^{\circ}$ . The band structure of not including the remote hopping terms ( $\gamma_3$  and  $\gamma_4$ ) is in black, and the full model including the remote hopping terms is plotted in red dotted lines.

In Ref.[277] we investigated a TDBG with a twist angle of  $\vartheta = 1.067^{\circ}$ . Applying the model at this twist angle, the calculated band structure along with the DOS is shown in Fig. 4.11. This  $\vartheta$  is close to the magic angle and the bands near the Fermi level are very narrow with a large DOS as shown in Fig. 4.11a at p = 0 GPa. These narrow bands are separated from the dispersive bands (colored black) with a gap of  $\Delta_{\pm n_s}$ . In the transport measurements shown in Section 5.2, large resistance is expected when the Fermi energy is in the gapped regions of the band structure. The application of electric field is illustrated in Fig. 4.11b by setting the interlayer potential difference u = 12 meV. The electric field opens a band gap  $\Delta_{\rm CNP}$  at the CNP similarly as in BLG but at the same time, it decreases and eventually closes the gaps  $\Delta_{\pm n_s}$ . Overall, the band structure calculations are in agreement with the measurements presented in Fig. 5.3a.



Figure 4.11: Band structure of twisted double bilayer graphene at  $\vartheta = 1.067^{\circ}$  at (a) p = 0 GPa and u = 0, (b) p = 0 GPa and u = 12 meV, (c) p = 2 GPa and u = 0 and (d) p = 2 GPa and u = 12 meV. The gaps separating the flat bands are indicated with  $\Delta_{\pm n_s}$  and the electric field opened band gap is shown with  $\Delta_{\text{CNP}}$ .

The pressure tuning of the band structure is done by varying  $\omega$  and  $\omega'$  according to Eq.(4.14). These parameters increased by  $\approx 30\%$  at p = 2 GPa from their value at p = 0 GPa while the interlayer distance decreased by  $\approx 5\%$ . The pressure has a drastic effect on the band structure as shown in Fig. 4.11c. The moiré gaps  $\Delta_{\pm n_s}$  are completely closed at p = 2 GPa, and the narrow bands are no longer separated from the other bands and slightly narrow down. The application of electric field is similar to p = 0 GPa at p = 2 GPa as it opens a band gap at the CNP.

From the band structure calculations, I also calculated the pressure and electric field dependence of the gaps in Ref.[277], which are shown in chapter 5 along with the measured values.

# 5. Chapter

# Tailoring the band structure of twisted double bilayer graphene with pressure

Twisted two-dimensional structures open new possibilities in band structure engineering. At magic twist angles, flat bands emerge, as discussed in Section 2.2. Twisted double bilayer graphene (TDBG), which consists of two Bernal stacked bilayer graphene (BLG) crystals with a rotation angle of  $\vartheta$  between them (depicted in Fig. 5.1b.), offer a versatile platform where an external electric field can be used to control the band structure via tuning the interlayer potential. Recent experimental [278–286] and theoretical [73, 287–292] studies showed the presence of correlated insulator states and topologically non-trivial phases in TDBG. The detailed band structure calculation of TDBG with a continuum model is given in Section 4.2.

Since the reconstruction of the band structure and the appearance of the correlated phases depend on the interactions between the layers, these are extremely sensitive to the interlayer distance. Therefore, tuning the interlayer distance in these structures is of central interest. This can be achieved by applying external pressure (p) as discussed in Section 3.3. In order to understand the influence of pressure on correlated phases in TDBG, the first step is to study the pressure dependence of the main parameters of the band structure in the single-particle picture. In this chapter, tuning of the band structure of TDBG is investigated experimentally close to the magic angle  $(1.05^{\circ})$  by applying hydrostatic pressure. Using bias spectroscopy and thermal activation measurements a strong modulation of the single-particle band gaps of the system is demonstrated. By applying p = 2 GPa, the moiré gaps can be fully closed. These findings agree well with the band-structure calculations presented in Section 4.2. Moreover, the measurements presented in this chapter indicate that pressure can lead to a topologically non-trivial gap at finite magnetic fields at the charge neutrality point and weaken the correlations.



Figure 5.1: Properties of the twisted double bilayer graphene device. (a) Schematic of the TDBG (black lattice) with a bottom graphite gate (orange) isolated by a hBN layer (light blue) and a top metallic gate (gray) isolated by a hBN layer and an AlO<sub>x</sub> layer (blue). The sample is contacted by edge contacts (yellow). The red arrows represent the pressure p, which modifies the distance between the layers. (b) Illustration of the twisted double bilayer structure. The purple arrow shows the direction of the transverse displacement field (D),  $\vartheta$  is the twist angle and d is the distance between the graphene layers which is tuned with the pressure. (c) and (d) Schematic pictures of the DOS in the vicinity of the flat band in magic-angle TDBG without and with an electric field, respectively. The external electric field splits the degenerate flat bands and opens a gap  $\Delta_{\rm CNP}$  at the charge neutrality point.  $\Delta_{\rm n_s}$  and  $\Delta_{\rm -n_s}$  are the band gaps separating the flat bands from the conduction and valence bands. These gaps are tunable with external pressure p.

## 5.1 TDBG heterostructure

The rotation between the top and bottom BLG lattices, illustrated in Fig. 5.1b, leads also to a rotation between their Brillouin zones (BZ). Near  $\vartheta_m$  the bands are very narrow near the CNP and these bands are separated from the dispersive bands as illustrated with the DOS schematically in Fig. 5.1c. The flat bands (red and blue) have a small bandwidth (BW) and are separated from the dispersive conduction bands by a gap  $\Delta_{n_s}$  and from the valence bands by  $\Delta_{-n_s}$ . Here  $n_s$  is the carrier density required to fill a single moiré band, either red or blue, with 4-fold degeneracy corresponding to four holes or electrons per superlattice unit cell in real space due to the spin and valley degeneracy. The indices of the gaps  $\Delta_{\pm n_s}$  signify that in order to fill either flat band and move the Fermi level into either gap, the carrier density must be  $\pm n_s$ . In TDBG the external perpendicular displacement field opens a band gap  $\Delta_{\rm CNP}$  and splits the
flat bands into two fourfold degenerate bands and also the gaps separating them from the dispersive bands  $\Delta_{n_s}$  and  $\Delta_{-n_s}$  are decreased as illustrated in Fig. 5.1d.

#### 5.1.1 Sample fabrication

van der Waals heterostructure was fabricated using the dry-transfer The technique [293] in ETH Zürich by Peter Rickhaus and Fokko de Vries. Half of a single BLG is picked up with the top hBN layer ( $\sim 32 \,\mathrm{nm}$ ), whereafter the remaining BLG is rotated by 1° and subsequently picked up. Then the bottom hBN ( $\sim 54$  nm) and graphite layer, which serves as a global bottom gate, are added. The device is fabricated in a cleanroom facility, using standard electron beam lithography techniques to define the different components and shapes. Side contacts to the twisted double bilayer graphene (TDBG) are created by reactive ion etching and evaporation of 10 nm of chromium (Cr) and 50 nm of gold (Au). After this the first top gate layer (blue in Fig. 5.2b) is added, consisting of 10/70 nm of Cr/Au, and the device boundaries are defined by reactive ion etching (red in Fig. 5.2b). Finally, atomic layer deposition is used to create a 30 nm aluminum oxide dielectric layer, which isolates the second top gate layer of  $10/110 \,\mathrm{nm}$  Cr/Au (grey transparent in Fig. 5.2b) from the device and first top gate layer. The two top gate layers are used as one in the measurements by adjusting their potential such that the density in the TDBG is constant throughout the device.



Figure 5.2: (a) Optical microscope image of the measured device. The scale bar is 2 µm (b) Schematic device geometry showing the four-terminal measurement setup.

#### 5.1.2 The pressure-dependence of the lever arms

The characterization of the sample involves the determination of the lever arms, which was done as described in Section 3.2.1 from the gate dependence of the resistance (Fig. 5.3) and from the quantum oscillations in magnetoconductance measurements (Fig. 5.5). The magnetoresistance of TDBG has a richer texture compared to SLG as shown in Fig. 5.5 as a function of n and B. Beside the LLs that starts from n = 0, LLs emerge also from  $n_s$  and  $\pm 2n_s$  following a Diophantine equation[16, 17, 59] of

$$n = \frac{\nu eB}{h} + sn_{\rm s},\tag{5.1}$$

where  $\nu$  is the LL filling factor and  $s = 0, \pm 1, \pm 2$  is an integer that corresponds to the Bloch band filling index[294]. These features are similar at 1 GPa and 2 GPa, and the secondary fans originate from the same density of  $n_{\rm s}$ .

The lever arms along with the residual charge density are shown in Table 5.1. The pressure dependence of the lever arms originates from the compression of the dielectrics and pressure-dependent dielectric constant of the hBN as already reported in Refs. [295, 296]. The extracted lever arms within the margin of error were the same at the same pressures at different cooldowns.

P (GPa)	0	1	2
$\alpha_{\mathrm{TG}} \left( \frac{10^{15}}{\mathrm{Vm}^2} \right)$	3.38(5)	3.75(5)	3.84(5)
$\alpha_{\rm BG} \left( \frac{10^{15}}{\rm Vm^2} \right)$	4.54(6)	4.94(7)	4.91(8)
$n_0 \ (10^{15} / \mathrm{m})$	2.3(2)	2.5(2)	2.4(2)
$D_0/\epsilon_0 ~({ m V/nm})$	0.016(5)	0.018(5)	0.017(5)

Table 5.1: The extracted lever arms and residual charge density and offset displacement field at different pressures.

## 5.2 Gate and pressure dependence of the resistance

Transport measurements were carried out in a four-terminal and two-terminal geometry with typical AC voltage excitation of 0.1 mV using a standard lock-in technique at 177.13 Hz. The measurement geometry is depicted in Fig. 5.2b. Four-probe resistance  $(R_{xx})$  measurement as a function of top and bottom gate voltages is plotted as a function of electron density n and displacement field D at temperature T = 1.5 K in Fig. 5.3. Lighter-colored regions of higher resistance correspond to conditions when the Fermi energy is in a gap. If the flat bands are completely filled with electrons or holes at  $n = \pm n_s$  the device shows single-particle gaps which are the most prominent at D = 0and start to fade away for larger displacement fields. Moreover, at the charge neutrality point (CNP, n = 0), a gap opens by increasing |D| as demonstrated by the increase of the resistance with the increase of |D| in Fig. 5.3a. This behavior is explained by the band structure calculations in Section 4.2.



Figure 5.3: Four-probe resistance of the TDBG as a function of the charge density (n) and electric displacement field (D) measured in (a) at ambient pressure p, in (b) at p = 1 GPa and in (b) at p = 2 GPa. Besides the charge neutrality point, there are two other high-resistance regions at  $\pm n_s$ , when the flat bands are completely filled. These regions fade away as the pressure is increased. The white arrows show the position of the emerging correlated phases at half-filling in panel (a).



Figure 5.4: Band structure of twisted double bilayer graphene at  $\vartheta = 1.067^{\circ}$  at u = 0(a) p = 0 GPa and (b) p = 2 GPa. The gaps separating the flat bands are indicated with  $\Delta_{\pm n_s}$ . The flat bands are highlighted with red and blue curves. Near the band structure, the calculated density of states is also shown.

After the measurements at ambient pressure, we applied hydrostatic pressure, which is detailed in Section 3.3, and  $R_{xx}$  was remeasured as a function of the gate voltages. We applied 1 GPa and 2 GPa pressure, and the results are is shown in Fig. 5.3b at p = 1 GPa and in Fig. 5.3c at p = 2 GPa. The features are similar to p = 0 GPa, except that the resistance values at  $n = \pm n_s$  are significantly smaller than in Fig. 5.3a. For example, along  $n = -n_s$  the resistance decreased by 70% at p = 2 GPa. To understand the origin of this change, the evolution of the band structure using the pressure dependence of the interlayer coupling parameters was calculated, which is detailed in Section 4.2. At p = 0 GPa, the moiré band gaps  $\Delta_{\pm n_s}$  are finite as shown in Fig. 5.4a. As the applied pressure is increased in numerical calculations, the moiré gaps decrease and then fully close with pressure, as shown at p = 2 GPa in Fig. 5.4b, in agreement with the observed decrease of the resistance. Moreover, in Fig. 5.3a there's a sign of local resistance peaks at half filling (marked with white arrows) which are attributed to emerging correlated phases, which disappears at p = 2 GPa (see Fig. 5.3b and Section 5.5.2 for more details), similarly to what was observed in magic-angle TBG[37, 72, 297].



Figure 5.5: Magnetic oscillation measurements of the four-terminal conductance G. (a), (b), (c) show dG/dn versus n and B at p = 0 GPa, 1 GPa and 2 GPa respectively. The white dotted lines are guides to the eye and follow the filled LLs with the filling factor of  $\nu = \pm 4, \pm 8...$  Beside the LLs that starts from n = 0, there are secondary fans, that start from  $n = \pm n_s$  following Eq.(5.1).

# 5.3 Acquiring the twist angle and its pressure dependence

There is a density-independent oscillation in a magnetic field which also depends on the superlattice size: at certain magnetic fields, the magnetic length is commensurable with the lattice periodicity which results in the recovery of the translation symmetry thus the electron feels effectively zero magnetic field. This results in an oscillation in the resistance called Brown-Zak oscillation[298, 299]. A common and reliable method to determine the twist angle  $(\vartheta)$  in superlattice structures such as the TDBG via transport measurements is to calculate it from the Brown-Zak oscillations. It can be calculated using that the Brown-Zak oscillations have maxima in the conductance at  $\phi = BA_{\rm m} = \frac{\phi_0}{q}$ , where  $\phi_0 = h/e$  is the flux quantum, q is an integer number and  $A_{\rm m}$ is the area of the superlattice unit cell which is given by Eq.(2.17).



Figure 5.6: Brown-Zak oscillations. (a) 2D color map of dG/dB versus  $n/n_s$  and  $\Phi_0/\Phi$  at D = 0 and at ambient pressure. (b) 2D color map of dG/dB versus  $n/n_s$  and  $\Phi_0/\Phi$  at D = 0 under 2 GPa pressure. On the figures, the oscillating pattern is well observable and their periodicity is the same. The dashed lines are a guide to the eye for the oscillations.

Brown-Zak oscillation measurements are shown in Fig. 5.6 at p = 0 GPa and at p = 2 GPa, where the oscillation is periodic in  $\Phi_0/\Phi$ . I determined the twist angle  $(\vartheta = 1.067^{\circ} \pm 0.003^{\circ})$  by fitting  $A_{\rm m}$  in the Brown-Zak oscillations as a function of  $\Phi_0/\Phi$ . At 2 GPa I found that the determined twist angle is identical to  $\vartheta$  at p = 0 GPa within the uncertainty of the measurements, which indicate that the pressure doesn't change the twist angle.

For consistency check, the twist angle is also determined from the position of the secondary peaks of the resistance at  $\pm n_{\rm s}$ . As  $n_{\rm s}$  is  $n_{\rm s} = 4/A_m$ ,  $\vartheta$  can be extracted directly from  $n_{\rm s}$ . Beside the value of  $\vartheta$ , its inhomogeneity is also estimated from the position and width of the resistance peaks at  $n = \pm n_{\rm s}$  in Fig. 5.3 at different pressures.

These varied in a similar range: at p = 0 GPa  $\vartheta$  is between 1.06° and 1.09°, at p = 1 GPa  $\vartheta$  is between 1.06° and 1.1° and at p = 2 GPa  $\vartheta$  is between 1.05° and 1.08°. These values coincide with the values obtained from the BZ oscillations. In regions containing substantial twist angle inhomogeneity, this inhomogeneity could result in smaller measured gap values, which are discussed in the next section, but it doesn't change the results qualitatively.

## 5.4 Band gap engineering of TDBG with pressure

To study quantitatively the effect of pressure on the band structure and to understand the decrease of the resistance at  $\pm n_s$  in Section 5.2, I made thermal activation measurements and bias spectroscopy. From these measurements, I determined the moiré band gaps at different displacement fields and pressures. I also compared them to my numerical calculations described in Section 4.2.

#### 5.4.1 Thermal activation measurements

When the Fermi level is in the gap, the resistance temperature (T) dependence is given by the Arrhenius equation

$$R_{xx} \propto e^{\frac{\Delta}{2k_{\rm B}T}},\tag{5.2}$$

where  $k_{\rm B}$  is the Boltzmann constant and  $\Delta$  is the gap energy[260, 262, 263]. To make thermal activation measurements, we measured the four-terminal resistance at a fixed D in a small range of n near the gapped regions as a function of the temperature (T).

A typical activation measurement is shown in Fig. 5.8a, i.e. a resistance map as a function of n and T. The resistance decreases with increasing temperature due to thermal activation over the gap. To determine the gap size, I extract the resistance maximum for each temperature value,  $R_{xx}(T)$ . Then the gap value was extracted from the Arrhenius plot, where the logarithm of  $R_{xx}(T)$  was plotted as a function of  $T^{-1}$  (see Fig. 5.8b). The linear region at high temperature (low  $T^{-1}$ ) is used to extract the gap energies by Eq.(5.2). The slope of the linear fit (black line) provided the gap values.

#### 5.4.2 Bias spectroscopy

The gap energies at the CNP are also estimated with bias spectroscopy measurements. For this, two-terminal resistance of the device with lock-in technique at low frequency was measured while a DC voltage bias ( $V_{\rm B}$ ) was applied to the sample. If a voltage higher or equal to the gap is applied, the gap as a barrier to transport is



Figure 5.7: Bias measurements. (a) The resistance versus n and the bias voltage ( $V_{\rm B}$ ) at  $D = -0.3 \,\mathrm{V/nm}$ . (b) Line-cut from (a) at n = 0 and  $n = -3 \cdot 10^{11} \,\mathrm{cm}^{-2}$ .

cancelled and conduction increases drastically. I performed bias measurements over a small range of n at fixed D (see Fig. 5.7a). First the maximum resistance  $(R_{\text{max}})$  was determined (occurring in Fig. 5.7a at n = 0 and  $V_{\text{B}} = 0$ ). I defined the gap value as the difference of the points  $V_{B\pm}$  where the resistance drops to 15% of the maximum resistance value, also taking into account a background value. The background  $R_{\text{gapless}}$  is determined at a different density value, where the resistance R is virtually independent of  $V_{\text{B}}$ . In other words, I approximated the gap energy with  $e|V_{\text{B}}|$  where  $V_{\text{B}} = (V_{\text{B}+} + V_{\text{B}-})/2$  with  $R(\pm V_{\text{B}\pm}) = R_{\text{gapless}} + 0.15(R_{\text{max}} - R_{\text{gapless}})$  as it is shown in Fig. 5.7b.

#### 5.4.3 Comparison between theory and experiments

The gap energies were extracted with the methods described in the previous sections, at several D points along the three dashed lines shown in Fig. 5.8c, which are denoted as  $\pm n_{\rm s}$  and CNP. These data are shown in Fig. 5.8d at p = 0 GPa, p = 1 GPa and p = 2 GPa with black, red and blue colors, respectively. The middle panel presents the evolution of the gap with D at the CNP ( $\Delta_{\rm CNP}$ ). The system is not gapped at D = 0 and a gap opens at  $D/\epsilon_0 \approx 0.2$  V/nm where  $\epsilon_0$  is the vacuum permittivity. The top panel shows the D dependence of the moiré gap at the electron side. A non-zero gap is present at D = 0, which decreases with |D| until it closes at  $|D|/\epsilon_0 \approx 0.4$  V/nm. The bottom panel summarizes the behaviour of the gap at  $-n_{\rm s}$  ( $\Delta_{-n_{\rm s}}$ ). It behaves similarly to  $\Delta_{\rm n_s}$  as it is finite at zero displacement field and decreases with |D|, but it closes at a smaller displacement field ( $|D|/\epsilon_0 \approx 0.3$  V/nm). The results at ambient pressure are in accordance with the previous findings[73, 278, 280, 284, 288]. The measurement results obtained at a pressure of p = 1 GPa and p = 2 GPa are shown in the



Figure 5.8: Thermal activation measurements of band gaps and comparison to the theoretical results. (a) A typical resistance map measured at  $D/\epsilon_0 = 0.375 \text{ V/nm}$  and p = 0 GPa near n = 0 as a function of the temperature T and charge density n. At every measured temperature the highest resistance value was taken. (b) R peaks extracted from panel (a) as a function of 1/T. The black line is the fit from which the gap values were obtained according to the Arrhenius equation. (c) The same n-D map of the resistance as Fig. 5.3a showing the lines along which gap values were estimated for several D values in panel (d) using similar thermal activation measurements as presented in panel (b). (d) The measured gaps with respect to D. The different colors show the gaps at different pressures. The gaps obtained from different methods are shown with different markers: the squares and diamonds show gaps obtained from four-probe and two-probe thermal activation measurements, respectively, and the circles are gaps obtained from bias measurements.

same plots with red and blue color, respectively. At finite pressure,  $\Delta_{\text{CNP}}$  behaves similarly to p = 0 GPa: the gap opens around the same displacement field (middle panel). However, for  $\Delta_{\pm n_s}$  the displacement field dependence strongly deviates from results at ambient pressure. At 1 GPa (red symbol)  $\Delta_{n_s}$  is smaller than at p = 0 GPa by a factor of 4. On the other hand, at 2 GPa (blue symbol)  $\Delta_{n_s}$  is closed at D = 0 and opens at finite D. The  $\Delta_{-n_s}$  data at p = 1 GPa and p = 2 GPa are similar to each other: they exhibit a peak at finite |D| before decaying but they are strongly reduced compared to the p = 0 GPa values.

I note that the gap energies remained approximately the same at the same hydrostatic pressures during different cooldowns and pressurization cycles. Part of the thermal activation measurements was confirmed at 2 GPa and 1 GPa using bias spectroscopy shown by circle symbols (for details see the previous subsection). These results are generally consistent with the thermal activation data.



Figure 5.9: Comparison of the band gap of TDBG between the measurement and the model. The displacement field was calculated with  $\epsilon_{BLG} = 5$ . The markers are the same gap values plotted in Fig 5.8, extracted from the thermal activation measurements. Whereas, the solid lines are theoretically calculated single-particle gaps.

A good qualitative understanding of the pressure dependence of the measured gaps can be obtained by comparing them to the theoretical calculations shown in Fig. 5.9 with solid lines. In these calculations, as it was described in Section 4.2, the electronelectron interactions and the quantum capacitance were neglected. The band gaps as a function of the interlayer potential difference (u), which is proportional to the displacement field, are plotted in Fig. 4.2e. The gaps in the measurements show qualitatively the same dependence on the displacement field as our calculation and the measured gap values are also comparable to the calculated ones. In the calculations, for  $\Delta_{n_s}$ , the gap values at ambient pressure and at p = 1 GPa decrease as a function of u, the latter being smaller than the former. Furthermore, the p = 1 GPa gap closes for smaller u. At p = 2 GPa the gap opens with u and closes at a higher interlayer potential difference. This tendency is similar to the experimental results. However, the theory suggests a faster opening of the gap at 2 GPa compared to the experiment. For the hole sides, at ambient pressure, a finite gap is present in the calculations, which closes by increasing u. In contrast, the gap is absent both for p = 1 GPa and p = 2GPa for all interlayer potential differences. These tendencies are qualitatively the same as observed in the experiments where only small gaps are seen at large pressure. Altogether, the measured spectrum is in good agreement with the theoretical expectations considering the simplicity of the model.

In order to compare the interlayer potential to the displacement fields applied in the experiment, I used Eq.(2.11) with the relative dielectric constant of bilayer graphene of  $\epsilon_{BLG} = 6 \pm 2[300, 301]$ . Using  $\epsilon_{BLG} = 5$  gives a relatively good agreement for the gap at the CNP, and less good for the moiré gaps (see Fig. 5.9). I emphasize that as a simplification I used equal potential drop between all four layers in the calculations. However, the potential drop between the layers can be different originating from crystal fields[302] and the different layer distances and dielectric constants. Moreover, I neglected all quantum capacitance corrections, and most importantly all correlation effects in the calculations (see section 4.2).

## 5.5 Effect of magnetic field

In a weak out-of-plane magnetic field, the band energy can be expressed as

$$\epsilon_{N,\xi,s_z}(\mathbf{k},B) = \epsilon_{N,\xi}(\mathbf{k}) + \mu_B g_s s_z B + m_{N,\xi}(\mathbf{k})B, \qquad (5.3)$$

where the second term is the Zeeman energy and the third term is the orbital Zeeman energy. Here, N is the band index,  $g_s$  is the spin g-factor and  $m_{N,\xi}(\mathbf{k})$  is the orbital magnetization[285, 303, 304]. If an insulating state is spin or valley polarized, and the magnetic field enhances the Zeeman splittings, the magnetic field increases the band gap. The last two terms can be combined as  $g\mu_B B$ , where g is the effective g-factor. In the case of in-plane magnetic field  $B_{\parallel}$ , the third term is absent and  $B_{\parallel}$  has an effect only on the spin-polarized states (then  $g = g_s/2$ ).

The out-of-plane magnetic field dependence of moiré band gaps is shown in Fig 5.10.  $\Delta_{-n_s}$  increases with B at D = 0 for both p = 0 GPa and p = 2 GPa (shown in panel a), and from the increase, the effective g-factor is larger than 4. The same tendency is visible for  $\Delta_{n_s}$  at 2 GPa and at D = 0, but at p = 0 GPa  $\Delta_{n_s}$  seems independent of Bin a limited magnetic field range as depicted in panel b. Both  $\Delta_{\pm n_s}$  are valley polarized



Figure 5.10: Out-of-plane magnetic field dependence of the band gaps. (a)  $\Delta_{-n_s}$  versus B at D = 0 at p = 0 GPa and at p = 2 GPa. From the fit of the blue dashed line,  $g(p = 2 \text{ GPa}) = 6 \pm 2$ . (b)  $\Delta_{n_s}$  versus B at D = 0 at p = 0 GPa and at p = 2 GPa. From the fit of the blue dashed line,  $g(p = 2 \text{ GPa}) = 30 \pm 2$ . (c)  $\Delta_{\text{CNP}}$  at  $D/\epsilon_0=0.3$  V/nm and -0.3 V/nm shown with squares and circles respectively.

and g is large at p = 0 GPa according to Ref.[285, 288]. At p = 2 GPa, I observe that, the effective g-factor remains large, larger than 4 for both  $\Delta_{\pm n_s}$ .  $\Delta_{CNP}$  at finite D is independent of B at small magnetic fields as it is shown in Fig. 5.10c.

#### 5.5.1 The closing of the band gap at the CNP



Figure 5.11: The charge neutrality gap (n = 0) at D = 0 as a function of the magnetic flux in the superlattice unit cell ( $\Phi = BA_{\rm m}$ ). The corresponding magnetic field B is given on the top axis. The gap starts to close above ~ 4 T and closes completely approximately at  $\Phi/\Phi_0 = 1/2$  for 1 GPa and  $\Phi/\Phi_0 = 1/3$  for 2 GPa. The blue and red lines are extrapolations for the gap at p = 2 GPa and p = 1 GPa, respectively. The shaded areas are the 95% confidence interval of the extrapolation.

At the CNP (D = 0) I observed signatures of interesting topological effects in an out-of-plane magnetic field. By increasing the magnetic field B, a gap opens and surprisingly above  $\sim 4 \,\mathrm{T}$  this gap starts to close. This is shown in Fig. 5.11 for 1 and 2 GPa respectively. This finding is similar to the results in Ref. [282], where the authors argued that if the gap is non-trivial with a non-zero Chern-number (C) then the gap should close at  $\Phi/\Phi_0 = 1/|C|$ . Here  $\Phi = BA_{\rm m}$  is the magnetic flux penetrating the superlattice unit cell. The measurements of Ref. [282] were performed at ambient pressure using a sample of  $\vartheta = 1.01^{\circ}$  twist angle. Their results suggested the presence of a non-trivial gap with C = 2 at finite D, which agrees with the theory [288–290, 305], and similar gap opening and closing was observed for D = 0. Our device shows a similar behavior, however, for D = 0 and finite pressure as shown in Fig. 5.11. At 2 GPa the gap closes near  $\Phi/\Phi_0 = 1/3$ , which could suggest a band gap with a Chern-number of C = 3. Surprisingly at 1 GPa (red symbols and red line), the gap starts to close at the same magnetic field as for 2 GPa, but the extrapolation suggests that it would go to zero at a much higher magnetic field than accessible in our setup (8T), probably near  $\Phi/\Phi_0 = 1/2$ . This suggests that the Chern-number may depend on the pressure. Another more likely possibility stems from the decrease of correlation effects which is well visible in Fig. 5.13, by the disappearance of correlated features at half-filling at finite pressures. According to the calculations of Ref[282], this leads to a smaller value of  $\Phi/\Phi_0$  where the gap closes. For a better understanding, further studies are required both theoretically and experimentally.



#### 5.5.2 Signature of spin-polarized correlated states

Figure 5.12: Four-probe resistance of the TDBG as a function of the charge density (n) and electric displacement field (D) measured at ambient pressure (a) at B = 0, (b) at B = 1 T and (c) at B = 2 T.



Figure 5.13: Four-probe resistance of the TDBG as a function of the charge density (n) and electric displacement field (D) measured at B = 2 T (a) at p = 0 GPa, (b) at p = 1 GPa and (c) at p = 2 GPa.

When the bandwidth of a narrow band is comparable with the characteristic Coulomb energy scale of electron-electron interaction, correlated states can emerge. For example, at half filling, TBG can exhibit correlated insulating states that are spin unpolarized[18, 19, 37]. Similarly, in TDBG at half fillings, correlated phases can emerge at finite D, but only at the electron side due to the larger bandwidth at the hole side (see Fig. 4.11d)[278, 279, 283, 284]. The resistance of these states is enhanced with the magnetic field, which is a sign of a spin-polarized correlated insulating state. On the sample, I observed similar behavior near  $n = n_s/2$  and  $D/\epsilon_0 = \pm 0.3$  V/nm as shown in Fig. 5.12: by increasing the out-of-plane magnetic field, the resistance increases.

Similar magnetic field dependence were also measured at p = 1 GPa and at p = 2 GPa, which is shown in Fig. 5.13. Under pressure, the correlated gap disappears for all applied magnetic fields which is an indicator of the decrease of correlations by increasing the pressure.

The effect of the in-plane magnetic field on the sample was also investigated at p = 2 GPa, as shown in Fig. 5.14. Comparing  $B_x = 0$  (panel a) and  $B_x = 3$  T (panel b) it is visible that the effect of the in-plane magnetic field is negligible. At a finite perpendicular magnetic field, the effect of applying an in-plane field is also negligible (panels c,d). In thermal activation measurements,  $B_x$  also had a negligible effect on the extracted gaps.

### 5.6 Conclusion

In conclusion, in this chapter, I presented our results of Ref.[277], where we investigated a TDBG with  $\vartheta = 1.067^{\circ}$  under pressure at different temperatures and magnetic fields. I found that the band structure significantly changes with pressure:



Figure 5.14: Four-probe resistance of the TDBG as a function of n and D measured at 2 GPa in different in- and out-plane magnetic fields. (a) presents the data for zero magnetic field, (b) for  $B_x = 3 \text{ T}$  in-plane magnetic field, (c) for  $B_z = 2 \text{ T}$  vertical magnetic field whereas (d) shows the measurement in  $B_z = 2 \text{ T}$  and  $B_x = 2 \text{ T}$ .

the single-particle moiré gaps present at zero displacement fields can be fully closed. I compared our findings with a single-particle continuum model and found a reasonable agreement with the experimental data. The changes achievable with the pressure are large and on the order of the bandwidth of the central flat band. The large tunability and the theoretical predictability suggest that the band structure of twisted structures can be precisely designed by taking into account the modified layer separation. The pressure combined with the electric field tunability allows extensive control of the band structure in situ. The exploration of the single-particle band structure is an important milestone toward tuning and understanding the emerging topological and correlated states of these systems. In addition, I showed in Section 5.5.1 that in an out-of-plane magnetic field, the closing of the gap at the charge neutrality point strongly depends on pressure, suggesting pressure dependence of the Chern-number.

# 6. Chapter

# Tuning the proximity-induced SOC in BLG/WSe<sub>2</sub> heterostructures with pressure

Combining graphene with TMDs induces SOC in graphene as shown in Section 2.3. The induced SOC has a substantial effect on the low-energy part of the band structure leading to or stabilizing novel phases such as topological phases[50, 306] or superconductivity[272]. In this chapter, the pressure-dependence of the SOC strength is investigated in BLG/WSe<sub>2</sub> and WSe<sub>2</sub>/BLG/WSe<sub>2</sub> heterostructures. Magnetoconductance measurements are shown here, such as weak localization, quantum Hall, and Shubnikov-de Haas oscillation measurements to extract the different SOC terms that determine the low-energy band structure of BLG. The WSe<sub>2</sub>/BLG/WSe<sub>2</sub> device has been fabricated by the group of S. Goswami in Delft and has been measured by M. Kedves, whereas I have fabricated and measured the BLG/WSe<sub>2</sub> samples. I have performed the modeling for both kinds of devices.

# 6.1 Varying the SOC in BLG/WSe<sub>2</sub> heterostructures with pressure

Applying hydrostatic pressure is a reliable way to increase the proximity-induced SOC in these heterostructures as it was shown in our pioneering work with single-layer graphene[307]. In BLG/WSe<sub>2</sub> heterostructures the presence of SOC in the conduction or valence band is expected to be controllable using gate electrodes[112, 118]. In the following, the presence of the SOC is shown by performing weak localization measurements. From a detailed study of SdH oscillations and quantum Hall effect the Rashba-type and Ising-type SOC strengths are extracted. By applying pressure as discussed in Section 3.3, a sizable increase of the SOC is detected than expected from theoretical predictions.



Figure 6.1: Properties of the BLG/WSe<sub>2</sub> heterostructure. (a) Schematic picture of the heterostructure. The BLG and the WSe<sub>2</sub> flakes are sandwiched between two hBN crystals. The heterostructure is contacted with Cr/Au edge contacts and a 30 nm thick ALD AlO<sub>x</sub> layer is deposited on the heterostructure to separate the gate from the device. A bottom graphite gate and a Cr/Au top gate are used to tune the charge density n and displacement field D. (b) Optical microscope image of the measured BLG/WSe<sub>2</sub> device A. The device is shaped into a Hall bar and it is covered with a global top gate. The scale bar is 5 µm. (c) Schematic structure of the BLG/WSe<sub>2</sub> heterostructure. The direction of the applied electric displacement is also shown between the graphene layers. (d) Low-energy band structure of the bilayer graphene including the spin-orbit coupling (SOC) with  $\lambda_{\rm R} = 15$  meV and  $\lambda_{\rm I} = 2.5$  meV. (e) An u = 10 meV interlayer potential difference opens a bandgap and helps spin-polarize the conduction bands. The bands in panel (d) and (e) are colored by their spin polarization shown by the color bar on panel (e).

#### 6.1.1 $BLG/WSe_2$ heterostructures

I fabricated the studied BLG/WSe<sub>2</sub> heterostructures using the dry-transfer technique, which is detailed in Section 3.1. The schematic of our device is shown in Fig. 6.1a, whereas an optical micrograph of device A can be found in Fig. 6.1b. It consists of a bilayer graphene (BLG) flake placed on top of a WSe<sub>2</sub> flake and encapsulated between two hBN layers. The graphene is contacted with etched side contacts. The graphite bottom gate and the metallic top gate allow to separately tune the charge carrier density, n, and the displacement field, D. During the fabrication, I used a WSe<sub>2</sub> layer with a thickness of 7 nm, the thickness of the top hBN is 22 nm, the bottom hBN is 58 nm thick and the aluminum oxide dielectric layer is 30 nm. Details of the other devices are given in Appendix C.

The schematic illustration of our BLG/WSe<sub>2</sub> heterostructure is shown in Fig. 6.1c. The low-energy band structure of pristine BLG near the K and K' points is described by Eq.(2.10), which exhibits parabolic bands touching near the K points with small modifications from the remote hopping terms ( $\gamma_3$  and  $\gamma_4$ ). The presence of the WSe<sub>2</sub> flake leads to the modification of the band structure of BLG, which is reflected by the appearance of additional SOC terms: a Rashba-type  $H_{\rm R} = \frac{\lambda_{\rm R}}{2} (\xi \sigma_x s_y - \sigma_y s_x)$  and an Ising-type  $H_{\rm I} = \xi \frac{\lambda_{\rm I}}{2} \sigma_z$  SOC term, also referred to as valley-Zeeman (VZ) SOC

as discussed in Section 2.3.2. The SOC terms are assumed to be present only in the bottom carbon layer, which is in proximity with  $WSe_2[45, 118]$ . The band structure is calculated using Eq.(2.43) around the K point with  $\lambda_{\rm R} = 15 \,{\rm meV}$  and  $\lambda_{\rm I} = 2.5 \,{\rm meV}$ (Fig. 6.1d). These parameters are plausible values based on the measurements shown in this chapter. The effect of  $\lambda_{\rm R}$  is depicted with black arrows, which splits the bands at higher k values and the splitting becomes larger at higher energies. The effect of  $\lambda_{\rm I}$  on the low-energy spectrum is shown with red and blue arrows. At low energies, it splits the spin degeneracy of the bands and makes them spin-polarized in the out-ofplane directions, whereas for higher energies a more complicated, canted spin structure arises as the combination of the two types of spin-orbit interactions [112]. The effect of the electric field is illustrated in Fig. 6.1e with u = 10 meV. The electric field leads to layer-polarization of electrons in the conduction and valence bands. For positive u, the conduction electrons are localized on the bottom layer neighboring the WSe<sub>2</sub> crystal. Hence the spin splitting caused by the proximity-induced SOC appears in this band (see panel e). For negative u, the layer-polarization is reversed and the spin splitting appears in the valence band.

#### Lever arms and their pressure dependence

The characterization of the sample involves the determination of the lever arms, which was done as described in Section 3.2.1 from the gate dependence of the resistance (Fig. 3.6) and from the quantum oscillations in magnetoconductance measurements (Fig. 6.2) using the full filling of the Landau levels (LL) being at  $n = \nu e|B|/h$ , where  $\nu = 0, \pm 4, \pm 8...$  The lever arms along with the residual charge density are shown in Table 6.1. The pressure dependence of the lever arms originates from the compression of the dielectrics<sup>1</sup> and pressure-dependent dielectric constant of the hBN as already reported in Refs. [295, 296] and was observed also in Section 5.1.2. Measurements under pressure are done using a pressure cell which is detailed in Section 3.3.

p (GPa)	0	2
$\alpha_{\rm TG} \ (10^{15} {\rm V}^{-1} {\rm m}^{-2})$	4.09(4)	4.45(4)
$\alpha_{\rm TG} \ (10^{15} {\rm V}^{-1} {\rm m}^{-2})$	2.47(3)	2.80(3)
$n_0 (10^{15} \mathrm{m}^{-2})$	4.20(5)	4.79(5)
$D_0/\epsilon_0 ~(V/nm)$	0.047(2)	0.069(2)

Table 6.1: The extracted parameters of device A for the calculation of n and D at p = 0 GPa and p = 2 GPa with the uncertainty of the last digit in the bracket originating from the measurement resolution, the readout precision and from the fitting.

<sup>&</sup>lt;sup>1</sup>Here the dielectrics are hBN and  $hBN/WSe_2$  on the other side.



Figure 6.2: Magnetic oscillation measurements of  $\rho_{xx}$  of device A (a) at p = 0 GPa and (b) at p = 2 GPa respectively. The white dashed lines show the Landau levels at different fillings ( $\nu = 0, \pm 4, \pm 8...$ ).



6.1.2 Gate and pressure dependence of the resistance

Figure 6.3: Longitudinal resistivity of the BLG/WSe<sub>2</sub> device A as a function of the charge density (n) and electric displacement field (D) measured at B = 0 T at (a) ambient pressure, T = 35 mK and (b) at p = 2 GPa, T = 50 mK. Near the charge neutrality line (n = 0), the resistance is high and increases with |D| as it opens a gap in the BLG. (c) Longitudinal resistivity as a function of D for n = 0 at 0 and 2 GPa pressures.

Fig. 6.3a shows longitudinal resistivity ( $\rho_{xx}$ ) as a function of top and bottom gate voltages, plotted as a function of n and D at temperature  $T = 35 \,\mathrm{mK}$  at ambient pressure. Lighter-colored regions of higher resistance correspond to the displacementfield-opened gap along the charge neutrality line. This gap is increasing by increasing |D| as expected in bilayer graphene. In Fig. 6.3b,  $\rho_{xx}$  is plotted at  $p = 2 \,\mathrm{GPa}$ . The features are similar compared to the  $p = 0 \,\mathrm{GPa}$  case. However, the resistance is increased with pressure. A line-cut along n = 0 is shown in Fig. 6.3c. At  $p = 0 \,\mathrm{GPa}$ ,  $\rho_{xx}$ depend non-monotonously on D. There is a small local maximum at D = 0 and minima at  $D/\epsilon_0 \sim \pm 10 \,\mathrm{mV/nm}$ . These features are already observed at low-temperatures in BLG/MoS<sub>2</sub> heterostructures[268] and BLG/WSe<sub>2</sub> heterostructures, and attributed this as an emergence of a correlated insulating state due to the SOC[269]. This feature is absent from the data at p = 2 GPa. The explanation may be that when the sample is pressed, the mobility decreases significantly<sup>2</sup>, and the correlated insulating states are not resolved due to the increased disorder.

#### 6.1.3 Weak localization in BLG/WSe<sub>2</sub> heterostructures



Figure 6.4: Magnetoconductivity of device A at D = 0 and  $n = \pm 1, \pm 0.5 \cdot 10^{12} \text{ cm}^{-2}$  (a) at p = 0 GPa and (b) at p = 2 GPa. Here, I averaged over 41 curves in a  $0.2 \cdot 10^{12} \text{ cm}^{-2}$  range around each density value and a fixed D to average out the universal conductance fluctuations. A clear antilocalization signal is visible in all density ranges.

Weak localization as discussed in Section 2.5.1 is a useful tool to determine the SOC in graphene. Thereby, to confirm the presence of induced SOC in the heterostructure, I performed magnetoconductance measurements at a low magnetic field B. In pristine BLG weak localization is expected[308]. However, the presence of SOC leads to weak antilocalization (WAL)[27, 93, 94, 96–99, 309]. I performed magnetic-field-dependent measurements at T = 1.5 K at zero, and at p = 2 GPa pressures. I averaged over 41 curves in a  $2 \cdot 10^{11}$  cm<sup>-2</sup> density range around each density value and a fixed displacement fields to average out the universal conductance fluctuations. The magnetoconductivity  $\delta \sigma = \sigma(B) - \sigma(B = 0)$  is shown in Fig. 6.4. In the measurements, weak antilocalization (WAL) is observed at both ambient pressure (Fig. 6.4a) and at p = 2 GPa (Fig. 6.4b), which is clear evidence for the presence of SOC in BLG.

The magnetoconductivity at different displacement fields and pressures at  $n = \pm 1 \cdot 10^{12} \text{ cm}^{-2}$  is shown in Fig. 6.5. Most curves show weak antilocalization (WAL) and the signal depends on D. The dependence is similar to that in Ref.[98] at ambient pressure.

<sup>&</sup>lt;sup>2</sup>At ambient pressure, the field effect mobility ( $\mu = \sigma_{xx}/ne$ ) in the electron regime is  $\mu \sim 80\,000\,\mathrm{cm}^2/\mathrm{Vs}$  and in the hole regime is  $\mu \sim 40\,000\,\mathrm{cm}^2/\mathrm{Vs}$ . At  $p = 2\,\mathrm{GPa}$ , in the electron regime is  $\mu \sim 10\,000\,\mathrm{cm}^2/\mathrm{Vs}$  and in the hole regime is  $\mu \sim 5000\,\mathrm{cm}^2/\mathrm{Vs}$ .



Figure 6.5: Weak localization measurements of device A at  $n = \pm 1 \cdot 10^{12} \text{ cm}^{-2}$ . (a-b) measurements at p=0 GPa and (c-d) measurements at p=2 GPa. Measurements made at different displacement fields at T = 1.5 K.

At p = 0 GPa, at the hole side (Fig. 6.5a), the WAL signal disappears by changing D from positive to negative. However, at the electron side (Fig. 6.5b), the signal decreases by increasing |D| in both directions. At p = 2 GPa, at the hole side (Fig. 6.5c), the signal weakly decreases at large displacement fields both in positive and negative directions. At the electron side under pressure (Fig. 6.5d), the WAL signal decreases by increasing D from negative to positive. Though this resembles the layer-tunable SOC effect, the tendency on a larger dataset is not that clear. At a large displacement field, the layer-polarization decreases and the layer specificity of the SOC is expected to disappear. Moreover, the WAL signal is a complex combination of different time scales. Therefore, the amplitude of the WAL signal alone do not necessarily reflect the SOC strength[98].

#### 6.1.4 Obtaining the Ising-type SOC strength

The LL crossings can be reliably used to extract the Ising-type SOC strength as shown in Section 4.1.1. I extract the position of the  $\nu = \pm 3$  crossing points in D by two methods. On the one hand, I measure  $\rho_{xx}$  maps as a function of n and D at a fixed out-of-plane magnetic field. In Fig. 6.6a,b,  $\rho_{xx}$  as a function of n and D at B = 8 T



Figure 6.6: Four-probe resistance of device A at B = 8 T as a function of n and D at (a) p = 0 GPa and (b) at p = 2 GPa. On the top, the corresponding Landau level (LL) filling factors ( $\nu = nh/eB$ ) are shown. LLs correspond to extended areas of large resistance and crossings correspond to sharp features of large resistance. We use the  $\nu = \pm 3$  crossing points to extract  $\lambda_{\rm I}$ . These are shown with red and blue arrows. (c) and (d) Line-cuts from the 2D maps along D at  $\nu = \pm 3$  (a) at B = 6.5 T and (b) at B = 8 T.

is shown. On the top, the corresponding Landau level (LL) filling factors  $\nu = \frac{nh}{eB}$  are also shown. The dark (bright) regions correspond to situations where the Fermi level is situated between (on) LLs, with (half-) integer  $\nu$ . Landau level crossings as a function of D appear as bright spots along certain  $\nu$ s at particular D values. The  $\nu = \pm 3$ crossing points are highlighted with red and blue arrows in panel (a) and these are also highlighted in the LL spectrum in Fig. 4.2. Here, I take a line-cut at  $n = \nu e|B|/h$ and from the resistance peak, the position of the crossing point is obtained. Line-cuts are shown in Fig. 6.6c,d at  $\nu = \pm 3$  at p = 0 GPa and p = 2 GPa pressures. As the measurement of these resistance maps takes a long time, instead of taking a map at a different magnetic field, I measure the  $\nu = \pm 3$  crossing points as a function of the magnetic field by measuring a D trace at a fixed filling factor at different magnetic fields (therefore also changing the density as  $n = \nu e|B|/h$  at every magnetic field to keep the filling factor constant). These measurements are shown in Fig. 6.7 for zero and 2 GPa pressures.



Figure 6.7: Measurements of the  $\nu = \pm 3$  crossing points of device A.  $\rho_{xx}$  as a function of B and D while setting  $n = \nu e|B|/h$  as  $\nu$  fixed at p = 0 GPa (a-b) and at p = 2 GPa (c-d). The evolution of  $\nu = -3$  is shown in (a) and (c) and the evolution of  $\nu = 3$  is shown in (b) and (d). The white arrows are guides to the eye to show the peak of the crossings.

The extracted position of the crossings as a function of B is shown with red and blue symbols for  $\nu = \pm 3$  in Fig. 6.8. The main contribution of the error comes from the read-off error of the peaks and from the uncertainty of the lever arms, it is less than  $\Delta D/\epsilon_0 = 0.0015 \text{ V/nm}$  ( $\Delta u = 0.12 \text{ meV}$ ). To determine the Ising-type SOC strength, the LL spectrum is calculated as described in Section 4.1.1 for different magnetic fields and  $\lambda_{\rm I}$  values. Then the magnetic field dependence of crossings is fitted having only  $\lambda_{\rm I}^{\rm b}$  as the only fitting parameter. The best fit is shown with dashed lines in the figure. From the fits we obtain  $\lambda_{\rm I}^{\rm b}(p = 0) = 1.6 \pm 0.2 \text{ meV}$ , which is in agreement with the experiments [50, 97, 106, 271] and the theoretical values in the literature [109–111].

To investigate the changes in the SOC coupling strength as the layer distance is



Figure 6.8: The position of the  $\nu = \pm 3$  crossing points extracted from the measurements of device A. The dashed lines represent the fits of the model on the measurement data using  $\lambda_{\rm I}^{\rm b}(p=0) = 1.6 \,\mathrm{meV}$  and  $\lambda_{\rm I}^{\rm b}(p=2 \,\mathrm{GPa}) = 2.5 \,\mathrm{meV}$ .

decreased, the same procedure is performed also at p = 2 GPa. The extracted crossing point values are shown in Fig. 6.8 with dark red and dark blue markers for  $\nu = \pm 3$ , respectively. Fitting similarly, the obtained  $\lambda_{\rm I}^{\rm b}(p = 2$  GPa) =  $2.5 \pm 0.2$  meV, which is a significant enhancement. We expect that, the extracted  $\lambda_{\rm I}$  and its enhancement with pressure leads to such a large spin splitting, that could potentially modify and tune the correlated states in twisted graphene/TMD heterostructures[116, 310].

#### 6.1.5 Obtaining the Rashba-type SOC strength

In the previous section, the Ising-type SOC is determined from LL related magnetoresistance measurements. Similarly, the Rashba-type SOC can also be determined from magneto-oscillations. For this, SdH oscillation measurements are used in this section. As visible in Fig. 6.1d, the Fermi surface is split due to the SOC coupling, and the splitting is dominated at high doping by the Rashba term (black arrows). As a result, there are two Fermi surfaces for the split bands with slightly different area  $S_{\rm F}^1$  and  $S_{\rm F}^2$ in momentum space. Therefore, the SdH oscillations give us the opportunity to extract the splitting of the Fermi surface as discussed in Section 2.5.2.

Shubnikov-de Haas oscillations are performed at 1.5 K at different charge densities. A quadratic background from the longitudinal resistance signal is subtracted and the result at a selected density is depicted in Fig. 6.9a. A fast Fourier transformation (FFT) on the signal is performed (Fig. 6.9c) and the obtained frequency  $f_B$  is converted into the area of the Fermi surface using Eq.(2.73) as  $S_{\rm F} = \frac{2\pi e}{\hbar} f_B$ . The same procedure is performed over several densities and a few of them are shown in Fig. C.2. The peaks



Figure 6.9: Shubnikov-de Haas oscillations. (a)  $\rho_{xx}$  after subtracting a quadratic background at  $n = -5 \cdot 10^{12} \,\mathrm{cm}^{-2}$  at  $p = 0 \,\mathrm{GPa}$  of device A. (b) Fermi surface areas extracted from the Shubnikov-de Haas oscillations as a function of n. The black markers are measurements made at zero pressure and the red ones are made at  $p = 2 \,\mathrm{GPa}$ . Their uncertainty comes from read-off error of the frequencies. The yellow line would be the Fermi surface  $S_{\rm F} = \pi^2 n$  if the spectrum was spin degenerate. (c) Fourier-transform of (a) as a function of the Fermi surface  $(S_{\rm F})$  converted from the frequency  $f_B$  as  $S_{\rm F} = \frac{2\pi e}{\hbar} f_B$ . The arrows show the peaks  $(S_{\rm F}^{1,2})$  and their difference  $(dS_{\rm F})$  which is used to get  $\lambda_{\rm R}^{\rm b}$ .

shown in Fig. 6.9c with arrows are extracted at different charge densities at D = 0 which is shown in Fig. 6.9b. The two peaks are the result of the slightly different Fermi surfaces produced by the Rashba-type SOC. At higher densities as the splitting is larger from the SOC, the peaks are more visible and separated. For better visibility, the difference of the obtained Fermi surfaces  $dS_{\rm F} = |S_{\rm F}^1 - S_{\rm F}^2|$  is shown in Fig. 6.10 by blue symbols for a large range of charge density. To extract the Rashba SOC strength, the Fermi surfaces are calculated using Eq.(4.4) at a fixed *n* using the previously obtained  $\lambda_{\rm I}^{\rm b}$ .  $dS_{\rm F}$  from the model is fitted to the measurement data using  $\lambda_{\rm R}^{\rm b}$  as the only fitting parameter<sup>3</sup>. From the fitting,  $\lambda_{\rm R}^{\rm b}(p = 0) = 11 \pm 2 \,\mathrm{meV}$ , which is shown in Fig. 6.10, where the dashed line and the shaded area shows the calculated Fermi surface splitting

 $<sup>^{3}\</sup>lambda_{\rm I}^{\rm b}$  is mostly relevant in the low-density regime. An uncertainty of  $\Delta\lambda_{\rm I}^{\rm b} = 1.5$  meV would bring an uncertainty of  $\Delta\lambda_{\rm R}^{\rm b} = 0.5$  meV, which is much less than the uncertainty of the fitting.



Figure 6.10: The difference of the Fermi surfaces as a function of n of device A. FFT is performed on the SdH data, where two peaks are observed. From the Fourier analysis, the frequency difference of the two peaks is extracted. The frequencies  $(f_B)$  are converted to Fermi surfaces  $(S_F = 2\pi e f_B/\hbar)$ . The blue error bars are measured at p = 0 GPa, and the red error bars are measured at p = 2 GPa. Their uncertainty comes from read-off error of the frequencies. The dashed lines are the fitting of the model with the corresponding  $\lambda_R^b$  values. The colored ranges show the uncertainty interval of the fits.

and the error, respectively. This is similar to the experimentally found values in the literature with similar techniques [94, 106, 271], but the theoretically predicted values are an order of magnitude smaller [107, 109, 110, 311]. The reason for this discrepancy is not clear.

The same analysis is performed at 2 GPa. The extracted  $dS_{\rm F}$  are shown with red markers in Fig. 6.10. The model is fitted similarly, and from it,  $\lambda_{\rm R}^{\rm b}(p = 2 \,{\rm GPa}) =$  $18 \pm 3 \,{\rm meV}$ , which is shown with a red dashed line and the shaded area shows the error in the Figure. The relative increase is similar to the increase of  $\lambda_{\rm I}^{\rm b}$ .

As it was already stated in Section 2.3.1, the strength of SOC strongly depends on the rotation angle between WSe<sub>2</sub> and graphene. Though during the fabrication the rotation angle between the flakes is not controlled, the large VZ splitting suggests rotation angles close to  $\vartheta \sim 11^{\circ}$  or  $\vartheta \sim 22^{\circ}$  according to Ref. [110] and according to Ref. [111] it is close to 0°. From the optical image of the flakes shown in Fig C.1, the twist angle is likely around  $\vartheta \sim 16^{\circ}$ . In Appendix C, measurements of the other devices are shown. Similar enhancement of the SOC strengths is found with pressure on these devices as well. On one of the samples, I find negative SOC at a similar value  $(\lambda_{\rm I} = -1.7 \pm 1 \text{ meV})$  from Quantum Hall studies, which implies that  $\vartheta$  is negative[111].

#### 6.1.6 Conclusion

In conclusion, the presence of proximity SOC in graphene is observed from WAL measurements. To determine the strength of the Ising-type SOC term in my BLG/WSe<sub>2</sub> heterostructures, I performed quantum Hall measurements as a function of the displacement field. From studying the LL crossings at  $\nu = \pm 3$ , I estimated  $\lambda_{\rm I}^{\rm b}$ . I performed a magnetoconductance study and from the FFT analysis of Shubnikov-de Haas oscillation, I extracted  $\lambda_{\rm R}^{\rm b}$ . I found that both are enhanced by more than 50% by applying hydrostatic pressure of 2 GPa on the device. These results can be important in realizing novel phases in graphene/TMD heterostructures for which large SOC is required.

# 6.2 Stabilizing the inverted phase in a $WSe_2/BLG/WSe_2$ heterostructure via hydrostatic pressure

In this section, our results of a BLG flake encapsulated between two layers of WSe<sub>2</sub> are shown, where the application of pressure led to the stabilization of a peculiar, band-inverted phase[306]. The measurements were performed by Máté Kedves and these are detailed in his PhD thesis. The samples have been fabricated by the group of Srijit Goswami in Delft. I supported the work with theoretical modeling. Details of the work can be found in Ref.[306]. Here I summarize the main results.

The schematic of the device is similar to the  $BLG/WSe_2$  devices as shown in Fig. 6.1c, but it has a second  $WSe_2$  layer on the top of the BLG under the top hBN. The gate-dependence of the sample is characterized similarly to the  $BLG/WSe_2$  devices, and the increase of the lever arms with pressure is also comparable as described in the Supporting Information of Ref.[306].

A summary on the effect of SOC is shown in Fig. 6.11. In panel (a) and panel (b) the resistance of the device is shown as a function of n and D at p = 0 GPa and at p = 1.65 GPa, respectively. Both at p = 0 GPa and at p = 1.65 GPa, there is a high resistance region at n = 0, which starts to decrease with |D|, then has a local minimum and starts to increase. It is qualitatively different compared to Fig. 6.3c. However, the resistance at D = 0 is similar in Fig. 6.11c and Fig. 6.3c. This behavior can be explained by the oppositely induced SOC in the two graphene layers from the WSe<sub>2</sub>, which is expected due to the inversion symmetry if they are aligned. However, the relative signs of the induced SOC is determined by the twist angle between the two WSe<sub>2</sub> crystals. As it is described in Section 4.1, if the sign of  $\lambda_{\rm I}^{\rm b}$  is different from the sign of  $\lambda_{\rm I}^{\rm t}$ , a gap is opened. This gap closes at  $u = \pm |\lambda_{\rm I}^{\rm b} - \lambda_{\rm I}^{\rm t}|/2$ . By further increasing the electric field, a



Figure 6.11: Measurements on the WSe<sub>2</sub>/BLG/WSe<sub>2</sub> device. (a-b) Resistance of the WSe<sub>2</sub>/BLG/WSe<sub>2</sub> device as a function of n and D at T = 1.4 K, B = 0 T at (a) ambient pressure, and (b) at p = 1.65 GPa. Near the charge neutrality line (n = 0), the resistance is high, it first decreases then after a local minimum it increases with |D| as it first closes and then opens a gap in the device. (c) Resistance as a function of D at n = 0 at 0 and 1.65 GPa pressures. (d) Band gap from thermal activation measurements at n = 0 as a function of D. (e)  $\nu = 0$  LL crossings as function of B. (f)  $\nu = 1$  LL crossings as function of B.

new gap opens. Here, the behavior of the gap was also verified with thermal activation measurements shown in Fig. 6.3d, where the trend of the gap  $\Delta$  is similar to the trend of the resistance in Fig. 6.3c. The local minimum with D is used to extract the Ising-type SOC and the extracted SOC is  $\lambda_{\rm I}^{\rm b}(p=0) = -\lambda_{\rm I}^{\rm t}(p=0) = 2.2 \pm 0.4$  meV. After pressing the heterostructure, the same measurements were performed and at p = 1.65 GPa, we obtained  $\lambda_{\rm I}^{\rm b}(p=1.65$  GPa) =  $-\lambda_{\rm I}^{\rm t}(p=1.65$  GPa) =  $5.6 \pm 0.6$  meV.

The presence of the SOC is also verified with quantum Hall measurements, where the  $\nu = 0, 1$  crossings along D were measured as a function of B similarly to Fig. 6.7, which is shown in Fig. 6.11e,f. The crossings when B goes to zero remain finite as opposed to the case when there is no SOC induced in the BLG as shown in Fig. 4.5c and Fig. 4.6c. The increase in the position of the crossings with pressure can also be explained by the increase of the SOC. Comparing the measured LL crossings with the calculated crossings in a single-particle picture in Section 4.1.1, the increase of the LL crossings by increasing the Ising-type SOC is consistent. However, to quantitatively compare the experiment with the model, the electron-electron interaction<sup>4</sup> has to be considered which is beyond our model.

To conclude, in a  $WSe_2/BLG/WSe_2$  device if the  $WSe_2$  crystals are oriented such that the induced SOC is the opposite in the two layers of the BLG, a band-inverted phase appears which can be closed with electric field and the closing point can be used to extract the induced SOC in the graphene. By applying hydrostatic pressure, the gap increases, and the closing point of the gap as a function of D is also enhanced which is a signature of the increased SOC.

 $<sup>^{4}</sup>$ As the width of LLs are defined by the disorder, in clean systems the electronic interactions become effectively strong[51].

# 7. Chapter

# Effect of pressure on the transport properties of SLG

A high-quality device with high mobility is essential to observe novel phenomena in graphene. When graphene was discovered, only rather low-quality devices, with mobility in the order of  $10^3 \,\mathrm{cm}^2/\mathrm{Vs}$  were available [4, 47] due to the charge traps in the SiO<sub>2</sub> substrate[178] and contamination originating mostly from the fabrication[161, 312]. To get rid of these obstacles, a separation of the graphene from the  $SiO_2$  is required, which was first achieved with current-annealed suspended graphene [123, 143] by realizing free-standing graphene and removing the fabrication residues with an applied large current. Later, a more reliable and versatile method was realized, by using hBN as a substrate<sup>[13]</sup> and fully encapsulating the graphene between hBN crystals<sup>[121, 313]</sup>. Both methods resulted in devices with mobilities over  $10^5 \,\mathrm{cm}^2/\mathrm{Vs}$ . These technical developments of realizing high-quality devices made it possible to observe new physics in graphene such as electron optics[314], Moiré physics[53], fractional quantum Hall effect[315] or highly viscous electron fluids[316, 317]. Also for the industrial application of graphene, reliably high-quality sample fabrication is essential. To achieve this, it is essential to know the main source of disorder and the limiting factors of the mobility to be able to eliminate them during fabrication. In the first devices, when the graphene was placed on a  $SiO_2$  substrate uncovered, the contamination on its surface and the substrate lead to short-ranged scatterings and also long-ranged scattering from the charge traps in  $SiO_2$ , which resulted their poor quality as discussed in Section 2.4.2. Since then, the encapsulation of graphene between hBN layers lead to atomically smooth interfaces with the substrate [318] and reduced scattering from short-ranged scatterers. There have been many experimental studies on the resistivity of graphene, which showed the importance of long-ranged scatterers [123, 143, 165, 166] and the proper choice of substrate to reduce the effect of the substrate-induced RIP scattering [178, 191–193]. If the scattering comes from interfacial effects like in the case of RIP scattering, the change of layer distance could help to understand better these scattering mechanisms, which could be achieved with pressure.

In this chapter, the basic transport properties of graphene are investigated as a

function of pressure, to reveal the scattering mechanisms, that limit the charge carrier mobility in the samples. The main limiting factors of resistance are described in Section 2.4. The devices investigated here were made in the clean room facility of RWTH Aachen University by T. Ouaj in the group of Prof. C. Stampfer using the fabrication method described in Section 3.1.

## 7.1 Low-temperature transport properties

The device geometry is the following: a SLG is encapsulated in hBN flakes and placed on a  $Si/SiO_2$  wafer, where the Si is used as a gate to tune *n* in graphene. We studied four devices, named A-D, for which optical images are shown in Fig. 7.1a-d, respectively. The devices are shaped into a Hall bar geometry. Here, mostly the results of device A are presented and the measurements on the other devices are given in Appendix E.



Figure 7.1: SLG devices. (a-d) Optical micrographs of SLG samples A, B, C and D, respectively. The devices are hBN encapsulated SLG etched into a Hall bar and they are on a Si/SiO<sub>2</sub> wafer, where the Si is used as a back gate. The scale bar is 5 µm. (e-h) Four-terminal conductivity of devices A-D, respectively as a function of n. The conductivity at p = 0 GPa is shown by black lines, whereas conductivity at 1 and 2 GPa is shown by teal and red lines, respectively.

The four-terminal conductivity of the investigated devices is shown in Fig. 7.1e-h at T = 1.5 K at ambient pressure and under hydrostatic pressure as a function of n. The measurements at 0 GPa are shown by black lines, whereas measurements at 1 and 2 GPa are shown by teal and red lines, respectively. The conductivity increases sublinearly with n, following Eq.(2.62). The measurements show that the increase of

the conductivity with n is always the largest at p = 0 GPa in every device. During pressurization of device B, a small region of the top hBN tore away from the graphene (based on optical microscopy performed later), and there the graphene got in touch with kerosene leading to spatially inhomogeneous doping, which resulted a second Dirac point as visible in Fig. 7.1f around  $n = 1.5 \cdot 10^{11}$  cm<sup>-2</sup> at 2 GPa.

The charge density for SLG is calculated as [319]

$$n = \sqrt{n_*^2 + (\alpha_{\rm BG} |V_g - V_{\rm D}|)^2} \cdot \text{sgn}(V_g - V_{\rm D}),$$
(7.1)

where  $\alpha_{BG}$  is the lever arm of the bottom Si gate,  $V_D$  is the gate voltage at the Dirac point, where  $\sigma_{xx}(n)$  has a minimum and  $n_*$  is the residual charge density at the Dirac point due to electron-hole puddles arising from inhomogeneity of the impurity-induced potential[145, 320].  $n_*$  is extracted from the log-log  $\sigma_{xx}(n)$  plots, which is detailed in Appendix E.1 and the extracted values are shown in Table 7.1 at different pressures. The lever arm  $\alpha_{BG}$  is extracted from quantum oscillations in magnetoconductance measurements as shown in Fig. 7.2. For device A,  $\alpha_{BG}$  and  $V_D$  is shown in Table E.1 at different pressures.



Figure 7.2: Magnetic oscillation measurements of  $\rho_{xx}$  of device A at p = 0 GPa. White dashed lines show the Landau levels at different fillings ( $\nu = 2, \pm 6, \pm 10...$ ).

To quantify the increase of  $\sigma_{xx}$  with n, Eq.(2.62) is fitted separately for electron and hole regions as

$$\sigma_{xx} = \left(\frac{1}{en\mu^{e/h}} + \rho_0^{e/h}\right)^{-1},$$
(7.2)

where e and h indexes the electron and hole regions, respectively. Here, I supposed that the mobility is constant, which is expected for long-ranged scatterers such as charged impurities or strain fluctuations [138, 163]. The fit for device A is shown in Fig. 7.3a with blue dashed curves, and the fitting parameters are given in Table 7.1. From the fitting, the charge carriers' mobility in the device is larger than  $\mu^{e/h} = 200\,000 \text{ cm}^2/\text{Vs}$  indicating high sample quality. As the pressure is increased, the mobility decreases significantly. The origin of its decrease will be investigated in this chapter.

The other fitting parameter  $\rho_0^{e/h}$ , which describes the density-independent resistivity from short-ranged scatterers, is negligible at ambient pressure in device A. At higher pressures,  $\rho_0^{e/h}$  becomes relevant, resulting in the saturation of  $\sigma_{xx}$  at larger pressures in Fig. 7.3a. For the other devices, the fitting parameters are given in Appendix E.2. Their pressure dependence is similar to the obtained values of device A in this section.

p (GPa)	0	1	2
$n_* (10^{10} \mathrm{cm}^{-2})$	1.6(5)	3.2(7)	2(7)
$\mu^h \left(\frac{m^2}{Vs}\right)$	22.12(6)	17.13(4)	16.27(5)
$\rho_0^h(\Omega)$	< 0.1	3.9(1)	4.8(1)
$\mu^e \left(\frac{m^2}{Vs}\right)$	26.47(6)	16.47(5)	15.05(4)
$\rho_0^e (\dot{\Omega})$	< 0.1	14.8(1)	26.6(2)

Table 7.1: Extracted parameters of device A of the fit of Eq.(7.2) on the conductivity and  $n_*$  at p = 0 GPa, p = 1 and p = 2 GPa with the uncertainty of the last digit in the bracket originating from the fitting.

From Eq.(2.45) combined with Eq.(2.48) and Eq.(2.9), the elastic mean free path can be calculated from  $\sigma_{xx}$  as

$$l_{\rm m} = \frac{\hbar}{e^2} \sqrt{\frac{\pi}{n}} \sigma_{xx}.$$
(7.3)

In Fig. 7.3b,  $l_{\rm m}$  is calculated as a function of n. At every density,  $l_{\rm m}$  is smaller than the width of the sample and the length between the voltage probes, which are shown with dashed lines in the figure, thus the transport is diffusive. At high densities,  $l_{\rm m}$ is comparable with the distance of the contacts, which allows to perform magnetic focusing experiments, which are shown later in this chapter. By increasing the pressure,  $l_{\rm m}$  decreases, which is related to the decrease in mobility.

The scatterings that contribute to the reduced  $l_{\rm m}$  can be divided into short- and long-ranged scatterers. The transport scattering time from short-ranged scattering  $\tau_{\rm m}^{\rm short}$  can be calculated from  $\rho_0^{e/h}$  using Eq.(2.45) combined with Eq.(2.48) and Eq.(2.9) as

$$\tau_{\rm m}^{\rm short} = \frac{\hbar}{e^2 v_{\rm F} \rho_0^{e/h}} \sqrt{\frac{\pi}{n}}.$$
(7.4)

At ambient pressure, as  $\rho_0^{e/h}$  is negligible, thus the calculated  $\tau_m^{\text{short}}$  is very large compared to  $\tau_m^{\text{long}}$ . At finite pressures,  $\tau_m^{\text{short}}$  gets smaller as shown in Fig. 7.3c, where at



Figure 7.3: Zero field transport properties of device A at T = 1.5 K. The measurements at 0 GPa are shown by black lines, whereas measurements at 1 and 2 GPa are shown by teal and red lines, respectively. (a) Conductivity of device A at different pressures as a function of n. The blue dashed lines are fits of Eq.(7.2). (b) The elastic mean free path ( $l_m$ ) calculated from  $\sigma_{xx}$  with Eq.(7.3). For comparison, the width of the sample and the length between the two inner contacts in the four-terminal measurement setup are also shown with dashed lines. (c) Transport scattering time from short-ranged scattering as a function of n at different pressures calculated from  $\rho_0$ . (d) Transport scattering time from long-ranged scattering as a function of n at different pressures calculated with Eq.(7.5).

higher densities,  $\tau_{\rm m}^{\rm short}$  starts to dominate the momentum relaxation rate, based on the saturation of  $\sigma_{xx}$ .

The measured transport scattering time from long-ranged scattering can be calculated from  $\tau_{\rm m} = l_{\rm m}/v_{\rm F}$  using the Matthiessen's rule as

$$\tau_{\rm m}^{\rm long} = \left(\frac{1}{\tau_{\rm m}} - \frac{1}{\tau_{\rm m}^{\rm short}}\right)^{-1}.$$
(7.5)

For device A,  $\tau_{\rm m}^{\rm long}$  is plotted in Fig. 7.3d as a function of *n*. It increases with *n* and is proportional to  $\sqrt{n}$  at every measured pressure, as expected from Eq.(2.56) for

both strain fluctuations and charged impurities [138, 163], where the mobility is doping independent. By increasing the pressure,  $\tau_{\rm m}^{\rm long}$  decreases, i.e. the long-range scattering rate increases, which result in a decrease in mobility.

To sum up, at ambient pressures, the main contribution to the conductivity comes from long-ranged scatterers as expected for a high-mobility device. The pressure decreases the mobility of SLG by increasing the scattering rates of both short- and longranged scatterers. For small charge densities, scattering from long-ranged scatterers remains the main source of scattering, and at high densities short-ranged scatterers start to dominate the transport.

## 7.2 Magnetotransport studies

To get a more in-depth view of the scattering mechanisms and how the pressure modifies the transport properties of SLG, magnetotransport measurements are also performed. Whereas field effect measurements can be used to extract transport lifetime, the so-called quantum lifetime, which is defined in Eq.(2.50), can be obtained from SdH oscillations. Secondly, WL measurements can be used to study the phase-coherent transport in the device and give information on dephasing due to inelastic scatterings. Finally, TMF measurements can be used to to probe modification of the band structure and the Fermi surface and can give information on scattering mechanisms that lead to the loss of ballistic behaviour of charge carriers.

#### 7.2.1 Shubnikov-de Haas oscillations

The longitudinal resistivity is shown in Fig. 7.4a as a function of  $B^{-1}$  after removing a quadratic background at a fixed charge density of  $n = 0.7 \cdot 10^{12} \text{ cm}^{-2}$ , at ambient pressure and different temperatures. An oscillation pattern is clearly visible, which vanishes by increasing  $B^{-1}$  or by increasing the temperature. These oscillations correspond to SdH oscillations and  $\rho_{xx}$  obeys to Eq.(2.74), which can be used to obtain the density, the effective mass, and the quantum lifetime. To fit  $\rho_{xx}$ , I used Eq.(2.74) with a small modification of limiting the harmonic order at 5 as

$$\rho_{xx} = \rho_0 \left[ 1 + c \sum_{s=1}^5 D(sx) \exp\left(-\frac{sm_*\pi}{eB\tau_q}\right) \cos\left(\frac{\hbar\pi^2 ns}{eB} - s\pi + s\phi_0\right) \right], \quad (7.6)$$

where  $D(sx) = \frac{X}{\sinh(X)}$  with  $X = s2\pi^2 k_{\rm B}Tm_*/eB\hbar$ . During the fitting procedure, a global fit is performed such that, at fixed charge densities  $\rho_{xx}(B,T)$  is fitted as a function of B at different temperatures simultaneously, where  $\tau_{\rm q}$  and  $m_*$  are assumed to be independent of T. The free parameters of the fitting are  $\rho_0$ , c,  $m_*$ , n,  $\phi_0$  and  $\tau_{\rm q}$ 



Figure 7.4: SdH oscillations measurements on device A. (a)  $\rho_{xx}$  after subtracting a quadratic background at  $n = 0.7 \cdot 10^{12} \,\mathrm{cm}^{-2}$  at  $p = 0 \,\mathrm{GPa}$  at different temperatures. The fit of Eq(7.6) on the data is shown with dashed lines and the most important fitting parameters are also shown in the figure, where the uncertainty of the last digit is in the bracket, which originates from the fitting. (b) Extracted quantum scattering time as a function of pressure at  $\pm 10 \,\mathrm{V}$  gate voltages. (c)  $\tau_{\rm m}/\tau_{\rm q}$  ratio as a function of pressure. 10 V correspond to  $n(p = 0 \,\mathrm{GPa}) = 7.2 \cdot 10^{11} \,\mathrm{cm}^{-2}$ ,  $n(p = 1 \,\mathrm{GPa}) = 7.3 \cdot 10^{11} \,\mathrm{cm}^{-2}$  and  $n(p = 0 \,\mathrm{GPa}) = -6.6 \cdot 10^{11} \,\mathrm{cm}^{-2}$ ,  $n(p = 1 \,\mathrm{GPa}) = -7.3 \cdot 10^{11} \,\mathrm{cm}^{-2}$  and  $n(p = 0 \,\mathrm{GPa}) = -8.2 \cdot 10^{11} \,\mathrm{cm}^{-2}$ .

from which the most important parameters  $(m_*, n, \tau_q)$  are shown in the figure along with the fitted curve with dashed lines. n is related to the oscillation period, and the fit results the same as extracted from quantum Hall measurements. The cyclotron mass  $m^*$ , which is related to the damping of the oscillations by the temperature, is close to the expected  $m^* = \hbar \sqrt{\pi n}/v_F = 0.017m_e$  value, where  $m_e$  is the electron mass. Moreover,  $\phi_0 = 3.2 \pm 0.1$  is close to  $\pi$ , as expected for SLG. Lastly,  $\tau_q$  is also obtained, which is responsible for the decay of  $\rho_{xx}$  with  $B^{-1}$ .

This analysis is performed at two fixed gate voltages  $V = \pm 10$  V at different pressures. The results from the global fits on the data for  $\tau_{\rm q}$  are shown in Fig. 7.4b as a function of pressure. As the pressure is increased,  $\tau_{\rm q}$  is decreased similarly to  $\tau_{\rm m}$ , which suggest that they have the same microscopic origin. Their ratio  $\tau_{\rm m}/\tau_{\rm q}$  describes how anisotropic the scattering is. As it is discussed in Section 2.4.2, for short-ranged scattering  $\tau_{\rm m}/\tau_{\rm q}$  is more or less isotropic and the ratio is  $\tau_{\rm m}/\tau_{\rm q} < 2$ . For long-ranged scattering such as scattering on charged impurities, the ratio is  $\tau_{\rm m}/\tau_{\rm q} > 2$ . The calculated ratio of  $\tau_{\rm m}/\tau_{\rm q}$ , which is shown in Fig. 7.4c is always larger than 2, indicating that the long-ranged scattering is the dominant scattering mechanism at every pressure. One possible explanation of large  $\tau_{\rm m}/\tau_{\rm q}$  is the presence of charged impurities<sup>1</sup>, which

<sup>&</sup>lt;sup>1</sup>For example charge traps in  $SiO_2$  under the hBN.

leads to a very strong enhancement in  $\tau_{\rm m}/\tau_{\rm q}$  similarly as in 2DEGs[139, 140]. For strain fluctuations, this has not yet been investigated. No clear pressure dependence can be observed for the  $\tau_{\rm m}/\tau_{\rm q}$  in device A. This analysis is performed for another device, device D, shown in the appendix E.5. In device D, an increasing tendency can be observed with the pressure. Nevertheless, further measurements are needed to make statements on the pressure dependency of  $\tau_{\rm m}/\tau_{\rm q}$ .

From SdH measurements, I found that the dominant scattering mechanism is the long-ranged scattering, which is consistent with the zero magnetic field transport studies in the previous section. One possible explanation can be charged impurities far from the graphene plane. However, corrugations cannot be ruled out as the origin of scattering in the device. By applying hydrostatic pressure,  $\tau_{q}$  is found to decrease similarly to  $\tau_{m}$ . By increasing the pressure, if remote charge impurities are present, their distance to graphene is slightly reduced, which leads to a decrease of  $\tau_{m}/\tau_{q}$ . However, the dielectric constant of hBN increases with pressure, which can lead to larger effective distances for the remote charge carriers[296]. Their combined effect determines the pressure dependence of  $\tau_{m}/\tau_{q}$ , which is beyond the scope of this thesis thus the presence of remote charged impurities cannot be ruled out.

#### 7.2.2 Magnetic focusing experiments

To address the scattering mechanisms, I turned to transverse magnetic focusing (TMF), where the amount of ballistic electrons carries information on the scattering processes within the sample. For that, I used the measurement arrangement shown in Fig. 7.5. Contact b is the injector and contact c is the collector contact. The non-local voltage  $V_{\rm nl}$  is measured between contact c and e. The reference contact e is much further than  $l_{\rm m}$  so no TMF signal is expected, which would distort the measurements. Contact a is grounded and a fixed voltage is applied between contacts a and b and the current I is measured as well.

The non-local resistance  $R_{\rm nl} = V_{\rm nl}/I$  is shown in Fig. 7.6a at T = 1.5 K and ambient pressure as a function of n and B. Above  $|B| \ge 0.3$  T, SdH oscillations are observed, which is expected from longitudinal resistance measurements. For  $|B| \le 0.2$  T, various peaks can be observed, which do not resemble SdH oscillations. In a magnetic field the electrons follow circular trajectories, and the magnetic field required for an injected electron to reach the collector contact<sup>2</sup> as described in Section 2.5.4, is given by

$$B_j = \frac{2\hbar\sqrt{\pi n}j}{eL},\tag{7.7}$$

 $<sup>^{2}</sup>$ When the distance between the contacts matches twice the cyclotron radius or its multiples.


Figure 7.5: Schematic measurement arrangement for TMF on device D. The current flows between contacts a and b. The current is injected at contact b and contact a is grounded. Contact c is used as a detector contact. The voltage is measured between contacts c and e, where the contact e serves as a reference. The black scale bar is  $5 \,\mu$ m.

where j = 1, 2, ... and L is the distance between the injector and collector contacts. Eq.(7.7) is plotted with black curves in the figure. The location of the peaks in the range of  $|B| \leq 0.2$  T follow these lines, indicating that they correspond to TMF. The first peak comes from the charge carriers which propagate directly from the injector to the collector, whereas the other peaks correspond to charge carriers which are reflected from the edge before they reach the collector. For n > 0, positive B focuses the electrons to the collector, while negative B directs them away from the collector, which leads to the absence of TMF peaks. For holes, the effect of the magnetic field is reversed, i.e. negative B results TMF peaks.

The temperature dependence of  $R_{\rm nl}$  is shown in Fig. 7.6b as a function of B at p = 0 GPa and at a fixed density of  $n = 10^{12}$  cm<sup>-2</sup>. Two clear peaks are visible, which correspond to TMF. Higher order peaks are not observed either because the elastic mean free path is not long enough or the edge roughness, due to the etching of the shape of the sample, is too large, which leads to random scattering[321]. The amplitude of these peaks decays by increasing the temperature and completely vanishes at large temperatures. To quantify this, the area under the first focusing peak  $A_1(T)$  is calculated after removing a linear background, which is fitted on  $R_{\rm nl}$  in the -B range. Using Eq.(2.94),  $\tau_{\text{eff}}$  is calculated, where for  $A_1(T_{\text{base}}) = A_1(T = 1.5 \text{ K})$  is used.  $\tau_{\text{eff}}$  is extracted at  $n = \pm 10^{12} \,\mathrm{cm}^{-2}$ , which is shown in Fig. 7.6c and in Fig. 7.6d on a log-log scale as a function of T. By increasing the temperature,  $\tau_{\text{eff}}$  decreases linearly in the log-log scale, as expected. The slope of the linear T dependence of  $\tau_{\rm eff}$  on a log-log scale gives information about the dephasing of the TMF. If only the electron-phonon scattering is responsible for the decay of  $\tau_{\rm eff}$  with T, a linear dependency  $(\tau_{\rm eff}^{-1} \propto T)$  is expected [180, 232]. However, if the main source of decay is originated from electronelectron scattering, a quadratic dependence  $(\tau_{\text{eff}}^{-1} \propto T^2)$  is expected[233, 234]. Within the Thomas-Fermi approximation, from the electron-electron interaction, for  $T \lesssim T_*$ 



Figure 7.6: Magnetic focusing on device A. (a)  $R_{\rm nl}$  as a function of n and B at T = 1.5 K and p = 0 GPa. The solid lines are the TMF peaks from Eq.(2.92) with L = 3.5 µm. (b)  $R_{\rm nl}$  after subtracting a linear background at  $n = 1 \cdot 10^{12}$  cm<sup>-2</sup> at p = 0 GPa at different temperatures. (c-d) Extracted effective scattering time as a function of temperature near  $n = \pm 1 \cdot 10^{12}$  cm<sup>-2</sup> at p = 0 GPa, p = 1 GPa and at p = 2 GPa with black, teal and red dots, respectively. The solid lines are linear fits. On panel (c) the slope of the fitted lines are  $-2.3 \pm 0.3$ ,  $-1.9 \pm 0.1$  and  $-1.8 \pm 0.1$  for p = 0, 1, 2 GPa, respectively. On panel (d) the slope of the fitted lines are  $-1.6 \pm 0.2$ ,  $-1.6 \pm 0.1$  and  $-1.7 \pm 0.1$  for p = 0, 1, 2 GPa, respectively. The blue dashed line in panels (c) and (d) is the calculated scattering time from e - e interaction using Eq.(7.8).

with  $k_{\rm B}T_* = 2v_{\rm F}\sqrt{k_F/L\pi}$ , the scattering time can be described by

$$\tau_{e-e}^{-1} \approx \frac{(k_{\rm B}T)^2}{2\hbar v_{\rm F} k_F} \log\left(\frac{3.6L}{w}\right),\tag{7.8}$$

where w is the width of the emitting and collecting contacts<sup>3</sup>[233]. In the figures, the solid lines are linear fits, their slope  $\alpha$  gives the temperature dependence of  $\tau_{\text{eff}} \propto T^{\alpha}$ .

<sup>&</sup>lt;sup>3</sup>For device A,  $w = 1.3 \,\mu\text{m}$ .

For  $n = -10^{12} \text{ cm}^{-2}$ ,  $\alpha = -2.3 \pm 0.3$ , which is close to -2, which is expected if the scattering mechanism is electron-electron scattering. For  $n = 10^{12} \text{ cm}^{-2}$ ,  $\alpha = -1.6 \pm 0.1$ , which is between -1 and -2 suggesting that both electron-electron and electron-phonon scatterings contribute to the reduction of the TMF signal. In the figures, the decay from the electron-electron interaction is also shown with a blue dashed line, which is calculated with Eq.(7.8). The extracted  $\tau_{\text{eff}}$  is close to  $\tau_{e-e}$  and the deviation is presumably caused by that Eq.(7.8) is an approximation which overestimates the the scattering rate[233].

The same measurements and extraction of  $\tau_{\text{eff}}$  is also performed at p = 1 GPaand p = 2 GPa, which are shown in Fig. 7.6c and in Fig. 7.6d with teal and red dots, respectively. The slopes of the curves are very similar, and from the log-log fits, the extracted  $\alpha$ s coincide with the  $\alpha$ s at ambient pressure, which means that the pressure doesn't change the scattering mechanism.

#### 7.2.3 Weak localization measurements on device A

In disordered samples with sufficiently long phase coherence length  $l_{\phi}$ , time-reversed trajectories can interfere leading to weak localization (WL) as discussed in Section 2.5.1. WL can be used to detect the presence of SOC by observing WAL as discussed in Section 6.1.3 for BLG/WSe<sub>2</sub> devices. Here, as the studied devices are SLG encapsulated in hBN, the SOC is negligible and WL signal is expected.

On device A, two terminal magnetoconductance measurements are performed at low out-of-plane magnetic fields between the two farthest contacts (contacts a and e in Fig. 7.5), which are  $L_{2pt} = 18.7 \,\mu\text{m}$  away from each other, much longer than  $l_{\phi}$  at every n. The phase coherence time, due to electron-phonon scattering [180, 186] and electron-electron scattering in the diffusive regime [322], depends on  $T^{-1}$ . In the ballistic regime, due to electron-electron scattering [200, 323],  $\tau_{\phi}$  depends on  $T^{-2}$ . In both cases, as  $\tau_{\phi}$  decays with T, the WL correction is suppressed. At T = 20 K and above, no WL signal is observed in this device. Measurements shown here are performed at  $T = 1.5 \,\mathrm{K}$ and a high-temperature background is also measured at T = 20 K, which is subtracted from the magnetoconductivity. The low-temperature data, before background subtraction and the high-temperature background is shown separately in Appendix E.3. The magnetoconductivity  $\delta\sigma(T) = \sigma(B,T) - \sigma(B = 0,T)$ , after subtracting the hightemperature background  $\delta\sigma = \delta\sigma(T = 1.5 \,\mathrm{K}) - \delta\sigma(T = 20 \,\mathrm{K})$  and averaging 37 curves over a density window of  $n = 5 \cdot 10^{11} \,\mathrm{cm}^{-2}$ , is shown in Fig. 7.7a at ambient pressure near the CNP displaying a typical WL signal. To quantify the scattering times, Eq.(2.70) is used to fit the data. In the fitting procedure, the diffusion coefficient is calculated from  $l_{\rm m}$ , which is obtained from the conductance measurements, as D is



Figure 7.7: Weak localization measurements on device A at T = 1.5 K. (a) and (c) Magnetoconductance  $\delta\sigma(B) - \delta\sigma(B = 0)$  after removing a high temperature background, measured at T = 20 K, near the CNP at p = 0 GPa in panel (a) and at p = 2 GPa in panel (c). The dashed lines are the result of the fit on the data with different boundaries for  $\tau_*$  using  $D = 1.27 \,\mathrm{m}^2/\mathrm{s}$ . (b) The extracted scattering times as a function of n at p = 0 GPa and at p = 2 GPa. (d) Scattering lengths as a function of n, calculated from the scattering times with  $L_i = \sqrt{\tau_i D}$ .

defined as  $D = v_{\rm F} l_{\rm m}/2[138]$  and D is averaged over the density window, which is used to obtain  $\delta\sigma$ . During the fitting, only the scattering times were the free parameters and a lower boundary is set to  $\tau_*$ : 1 fs, 100 fs or 10 ps. The fitting results are also shown in the figure with dashed curves, labeled with the fitted scattering times. Limiting  $\tau_*$  at both 1 fs and 100 fs gives a good fit, where  $\tau_*$  always converges to the lower boundary. However, setting  $\tau_*$  larger than 1 ps results in a bad fit, which suggests that  $\tau_*$  is in the 1 - 100 fs range. From the fits, the intervalley scattering time is  $\tau_{\rm iv} = 11 \pm 3$  ps. The phase coherence time is  $\tau_{\phi} = 3.7 \pm 0.6$  ps, which is smaller than  $\tau_{\rm iv}$  in device A, which is common for SLG samples[201, 202].

Similar measurements and fitting to n = 0 is performed at different charge densities and the result is shown in Fig. 7.7b. The fitting parameters that correspond to the lowest bound of  $\tau_* = 1$  fs are shown here.  $\tau_{iv}$  is shown with red error bars and the lines between them are guides to the eye. A small dependence of the intervalley scattering time on the density can be observed, which is similar to the *n* dependence of  $\tau_{\rm m}$ . I also calculated the intervalley scattering length  $L_{\rm iv} = \sqrt{D\tau_{\rm iv}}$ , which is shown in Fig. 7.7d.  $L_{\rm iv}$  is found to be in the same order as the width of the sample. The intervalley scattering time is related to short-ranged scatterers such as atomically sharp impurities or the edge of the sample and for high-quality devices usually the edges are the dominant source of intervalley scattering[138, 324]. From the extracted  $L_{\rm iv}$  values in device A, which width is 3.9 µm, the intervalley scattering is caused mainly by the sample edges. The phase coherence time, which is plotted with black error bars, is related to inelastic electron-electron scattering and electron-phonon scattering. By increasing n,  $\tau_{\phi}$  is also increasing which is consistent with Refs.[200, 322, 323, 325].

Similar measurements and analysis are performed at p = 2 GPa, but no high temperature background is subtracted. At n = 0,  $\delta\sigma$  is shown in Fig. 7.7c as a function of *B*. Similarly to p = 0 GPa, WL is observed. It is fitted in similar  $\tau_*$  ranges: for  $\tau_*$ in the range of 1 - 100 fs, Eq.(2.70) fits the data very well and the large  $\tau_*$  gives a bad result. I found that both  $\tau_{\phi}$  and  $\tau_{iv}$  decreased under pressure.  $\delta\sigma$  is measured also at different densities at p = 2 GPa, which are processed similarly to p = 0 GPa. The results are plotted in Fig. 7.7b. The intervalley scattering time is shown with blue error bars with a guiding dashed curve. Altogether, the charge density dependence of  $\tau_{iv}$  at 2 GPa is similar at p = 0 GPa, but a small decrease of  $\tau_{iv}$  can be observed. In Fig. 7.7d,  $L_{iv}$  is shown with blue error bars at p = 2 GPa. It is still comparable to the sample width, but it gets smaller by applying hydrostatic pressure, which is a signature of the increased effect of atomically sharp defects. The coherence time, which is shown with magenta error bars in Fig. 7.7b, is also reduced by increasing the pressure. However, a similar *n* dependency is observed at p = 0 GPa, as excepted.

All in all, from the WL measurements, I found that the intervalley scattering time is long and remains long under pressure, and that its origin is most likely the edge of the sample. I also found that all scattering time decreased under pressure following the momentum scattering time.

### 7.2.4 Suppression of WL in an in-plane magnetic field on device C

At the end of section 2.5.1, the effect of in-plane magnetic field on the phase coherence time is discussed. Due to corrugations and ripples, an applied in-plane magnetic field has also out-of-plane magnetic field components, which can be interpreted as random vector potentials that lead to dephasing[201, 202]. As  $B_{\parallel}$  increases the dephasing, measurement of  $\tau_{\phi}$  as a function of  $B_{\parallel}$  gives details about out-of-plane corrugations of the system.



Figure 7.8: WL in-plane magnetic field dependence of device C. (a-b)  $\delta\sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity as a function of B at various in-plane magnetic fields  $(B_{\parallel})$  at T = 1.5 K,  $n = 1.25 \cdot 10^{12} \text{ cm}^{-2}$ , (a) at p = 0 GPa and (b) at p = 2 GPa. The black dashed lines correspond to global fits on the data, which results  $\tau_* = 1$  fs and  $\tau_{iv}(p = 0) = 6.7 \pm 0.4$  ps with  $D(p = 0) = 1.27 \text{ m}^2/\text{s}$  for p = 0 GPa in panel (a) and  $\tau_* = 1$  fs and  $\tau_{iv}(p = 2 \text{ GPa}) = 3.6 \pm 0.8$  ps with  $D(p = 2 \text{ GPa}) = 0.76 \text{ m}^2/\text{s}$  for p = 2 GPa in panel (b). (c) Extracted  $\tau_{\phi}^{-1}$  as a function of  $B_{\parallel}^2$  at 0 and 2 GPa pressures. The dephasing rate depends linearly on  $B_{\parallel}^2$  according to Eq.(2.72). The solid lines are line fits which gives  $Z^2R = 25 \pm 3 \text{ nm}^3$  for p = 0 GPa and  $Z^2R = 47 \pm 15 \text{ nm}^3$  for p = 2 GPa.

Magnetotransport measurements are performed on device C. From the device, an optical micrograph is shown in Fig. 7.1c. It is a similar hBN/SLG/hBN device etched into a Hall bar like device A. The magnetoconductivity  $\delta \sigma = \sigma(B) - \sigma(B = 0)$  is shown in Fig. 7.8a at  $n = 1.25 \cdot 10^{12} \text{ cm}^{-2}$ , at T = 1.5 K and at ambient pressure as a function of B at different in-plane magnetic fields  $(B_{\parallel})$ . The curves show WL behavior. By increasing  $B_{\parallel}$  from 0 to 1 T, shown by yellow to purple curves, the magnetoconductivity drops at larger Bs. To quantify the decrease, a global fit is performed on the data using

Eq.(2.70) to extract  $\tau_{\phi}(B_{\parallel})$  such that, at a fixed charge density,  $\delta\sigma$  is fitted as a function of *B* at different in-plane fields simultaneously, where  $\tau_*$  and  $\tau_{iv}$  are assumed to be independent of  $B_{\parallel}$  and only  $\tau_{\phi}$  is assumed to depend on  $B_{\parallel}$ . The diffusion coefficient for the fitting is calculated from  $l_{\rm m}$ . The results of the fitting are shown with black dashed lines in the figure. Similarly to the WL measurements on device A, the best fits on the data is acquired with  $\tau_* = 1$  fs. The fitted  $\tau_{iv}(p = 0) = 6.7 \pm 0.4$  ps is similar to the fitted  $\tau_{iv}$  on device A. From  $\tau_{iv}$ ,  $L_{iv}(p = 0) = 2.9 \pm 0.3$  µm, which is comparable to the sample width (W = 3.4 µm) like in device A, thus the intervalley scattering is most likely caused mainly by the sample edges.

Real systems exhibit corrugations such as ripples and wrinkles. These corrugations are usually Gaussian correlated and can be parametrized with the RMS height fluctuation (Z) and the in-plane correlation length (R). Due to these corrugations, an applied in-plane magnetic field has an out-of-plane component, which leads to dephasing as discussed at the end of section 2.5.1. From Eq.(2.72), the total dephasing rate  $\tau_{\parallel}^{-1}$  is proportional to  $B_{\parallel}^2$ . The dephasing rate  $\tau_{\phi}^{-1}$  from the fitting is shown in Fig. 7.8c as a function of  $B^2$  with black error bars. A linear tendency is observed, as expected from Eq.(2.72). A linear fit of  $\tau_{\phi}^{-1}$  on  $B_{\parallel}^2$  is shown with a black line in the figure. From the slope, the corrugation volume  $Z^2R$  is extracted using Eq.(2.72). The extracted volume  $Z^2R = 25 \pm 3 \text{ nm}^3$  fits in range of previously reported values between 1.6 nm and 156 nm[201, 202].

Similar magnetotransport measurements are performed under 2 GPa hydrostatic pressure. The magnetoconductivity  $\delta\sigma$  is shown in Fig. 7.8b at  $n = 1.25 \cdot 10^{12} \,\mathrm{cm}^{-2}$ , at  $T = 1.5 \,\mathrm{K}$  and at  $p = 2 \,\mathrm{GPa}$  as a function of B at different in-plane magnetic fields  $(B_{\parallel})$ . A similar tendency is observed at ambient pressure:  $\delta\sigma$  is decreasing by increasing  $B_{\parallel}$ . A similar analysis is performed, and the best result is also obtained with  $\tau_* = 1 \,\mathrm{fs}$ . From the fit, the extracted  $\tau_{\mathrm{iv}}(p = 2 \,\mathrm{GPa}) = 3.6 \pm 0.8 \,\mathrm{ps}$  decreases with pressure, and  $L_{\mathrm{iv}}(p = 0) = 1.7 \pm 0.2 \,\mathrm{\mu m}$  is considerably smaller than the sample width, which implies that the effect of atomically sharp impurities is enhanced with pressure.

The dephasing rate is shown in Fig. 7.8c as a function of  $B^2$  with red error bars for p = 2 GPa. Overall, the dephasing rate is increased with pressure similar to device A. A linear fit is shown with a red line in the figure, which gives  $Z^2R = 47 \pm 15 \text{ nm}^3$ for p = 2 GPa. The increase of the corrugation volume with pressure implies that the ripples or wrinkles in the device are increased. This is a counter-intuitive finding, as the hydrostatic pressure compresses the volume of a solid and if it is isotropic, the shape remains unchanged. A possible explanation is that, as the the sample is pressed and the hBN layer is pushed closer to the graphene, the corrugations are wrinkled, leading to an increased number of wrinkles and crumples, which increases the average corrugation volume. As the wrinkle size is increased, the scattering region is increased, which results in more scatterers that reduce the phase coherence time and the mobility of the device consistently with the zero magnetic field measurements, which are discussed in Appendix E.2 for this device. Similar measurements are performed on device D, which are discussed in Appendix E.4. From those measurements, an increase in the corrugation volume is also observed consistently with the findings in device C.

## 7.3 Temperature-dependent transport and the role of RIP scattering

At large temperatures (typically above  $T \gtrsim 20 \text{ K}$ ), as it is discussed in Section 2.4.3, the electron-phonon scattering mechanisms start to dominate the resistance[177, 178, 180]. In graphene devices, the main phonon scattering mechanisms are the acoustic[180, 184] and optical[190] phonon scattering and also the remote interfacial phonon (RIP) scattering[326–329], which originates from the polar optical modes of the substrate. Below the Bloch-Grüneisen temperature ( $T_{BG}$ ), the contribution to the resistivity of acoustic phonon-electron interaction is linear in temperature ( $\rho_A = \gamma T$ ) and independent of charge density. The scattering due to optical phonons of graphene becomes important above  $T \gtrsim 270 \text{ K}[190]$ , which is around the top of the measurement range presented here, thus it is neglected in the following. The third phonon scattering mechanism, the RIP scattering originates from the hBN layers, which encapsulate the graphene in the devices studied here. The scattering due to RIP of hBN can be well described with Eq.(2.66). The combined effect of the scattering mechanisms on the resistivity in our measurement range can be described with

$$\rho(T,n) = \rho_0(n) + \gamma T + \frac{B}{n^{\alpha}} \left( \frac{1}{e^{\hbar\omega_1/k_{\rm B}T} - 1} + \frac{S}{e^{\hbar\omega_2/k_{\rm B}T} - 1} \right),\tag{7.9}$$

where the first term, the residual resistance of  $\rho_0$ , describes the low-temperature resistance of the device, the second term describes the charge carrier scattering on acoustic phonons and the third term is the resistance contribution from RIP scattering, where  $\omega_{1,2}$  are the strongest surface optical phonon modes, and S is the ratio of their coupling strength. In Eq.(7.9), the T dependence of  $\rho_0$  is neglected, which is valid only away from the CNP. For example, if  $n \geq 10^{11} \text{ cm}^{-2}$ , the Fermi temperate is  $T_{\rm F} \geq 1000 \text{ K}$ , and in our temperature range this results a negligible T dependency of the short-range and long-range scattering as discussed in Section 2.4.3.

The resistivity as a function of the temperature at a series of charge densities at ambient pressure of device A is shown with dots in Fig 7.9a.  $\rho_{xx}$  follows a superlinear



Figure 7.9: Temperature dependence of the resistivity of device A. (a) The resistivity at some densities as a function of the temperature at ambient pressure plotted with dots. The red and blue dashed lines are the result of the global fit with fixed polar optical surface frequencies for the electron and hole regimes, respectively. (b)  $\Delta \rho_{xx} = \rho_{xx} - \rho_0$  as a function of temperature at  $n = -0.8 \cdot 10^{12} \text{ cm}^{-2}$  and at p = 0 GPa, p = 1 GPa and p = 2 GPa plotted with black, teal and red dots, respectively. The dashed lines are global fits at different pressures.

tendency: below  $T \leq 140$  K,  $\rho_{xx}$  has a linear T dependence, which is attributed to acoustic phonon scattering. At higher temperatures, the resistance becomes non-linear due to the RIP scattering. To quantitatively analyse the data,  $\rho_0$  is subtracted from the resistivity, and their difference  $\Delta \rho_{xx} = \rho_{xx} - \rho_0$ , which is shown with black dots as a function of the temperature at  $n = -0.8 \cdot 10^{12} \text{ cm}^{-2}$  in Fig 7.9b, is fitted. During the fitting, Eq.(7.9) is used to fit  $\Delta \rho_{xx}$  on the charge density and temperature dependence simultaneously. The fitting involved the following steps:

- 1.  $\rho_0$  is determined by fitting a line on the linear regime (20-115 K) and the constant part is taken as  $\rho_0$ .
- 2.  $\Delta \rho_{xx}$  is fitted with Eq.(7.9) on the charge density and temperature dependence simultaneously in the temperature range of 20 - 285 K and in the charge density range of  $[-11, -7] \cdot 10^{11}$  cm<sup>-2</sup> and  $[7, 11] \cdot 10^{11}$  cm<sup>-2</sup>. The fitting parameters are *B* and  $\gamma$ .  $\alpha$  is chosen arbitrarily ( $\alpha = 1$ ). *S*,  $\omega_1$  and  $\omega_2$  is fixed, and their value is calculated below.
- 3. From the previous step  $\gamma$  is used to calculate  $\rho_{\text{RIP}} = \rho_{xx} \rho_0 \gamma T$ . At a fixed temperature, a linear curve is fitted on  $\ln(\rho_{\text{RIP}})$  as a function of  $\ln(|n|)$  in the density range used in the previous step. The slope of the fitted line determines  $\alpha$ .
- 4.  $\Delta \rho_{xx}$  is fitted with Eq.(7.9) on the charge density range of  $[-11, -7] \cdot 10^{11} \,\mathrm{cm}^{-2}$

and  $[7, 11] \cdot 10^{11} \text{ cm}^{-2}$  and the temperature range of 20 - 285 K. The fitting parameters are B and  $\gamma$ .  $\alpha$  used here is obtained in the previous step.

 $\rho_{\text{RIP}}$  is recalculated from the parameters of step 4, and its logarithm is shown as a function of  $\ln(|n|)$  in Fig. 7.10 at T = 240 K. A linear tendency is expected with the slope of  $\alpha$  in the range where Eq.(2.66) is valid. The tendency is linear at larger densities, where the fitting is performed. However, at lower densities, the resistivity deviates. This deviation from linear tendency at small densities is also present in theory[326, 330]. For consistency check, a linear fit is performed, which result in the same value for  $\alpha$  as obtained in step 3. By performing the same analysis at different temperatures, I observe that,  $\alpha$  varies within a small range, which determines the uncertainty of  $\alpha$ . At the hole regime (Fig. 7.10a),  $\alpha(p = 0) = 1 \pm 0.1$ , which is in range of the experimentally observed values[178, 191, 192] and in the theoretically expected range of [0.5, 1][326, 330]. At the electron regime, which is shown Fig. 7.10b,  $\ln(\rho_{\text{RIP}})$  is slightly non-linear in our measurement range<sup>4</sup>, and the fitting gives an unreliable result. For the sake of completeness, near the largest measured densities, I obtain  $\alpha(p = 0) = 0.9 \pm 0.2$ .



Figure 7.10: Logarithm of the longitudinal resistivity after subtracting the residual resistivity and the resistivity contribution of the acoustic phonons as a function of the logarithm of the charge density at T = 240 K of device A is shown with black, teal and red lines at 0, 1 GPa and 2 GPa, respectively. The blue dashed lines are linear fits on the linear regime of the curves. (a)  $\ln(\rho_{\rm RIP})$  at n < 0. From the slope of the fitted lines  $\alpha(p = 0) = 1 \pm 0.1$ ,  $\alpha(p = 1 \text{ GPa}) = 1.1 \pm 0.1$  and  $\alpha(p = 2 \text{ GPa}) = 1.1 \pm 0.1$ . (b)  $\ln(\rho_{\rm RIP})$  at n > 0. From the slope of the fitted lines  $\alpha(p = 0) = 0.9 \pm 0.2$ ,  $\alpha(p = 1 \text{ GPa}) = 0.8 \pm 0.2$  and  $\alpha(p = 2 \text{ GPa}) = 1 \pm 0.2$ .

The lower limit in step 2 and step 4 is chosen at  $n = \pm 7 \cdot 10^{11} \text{ cm}^{-2}$ , as at smaller densities, the RIP contribution to the resistivity starts to deviate from Eq.(7.9)[193], which can also be seen in Fig. 7.10.

<sup>&</sup>lt;sup>4</sup>At different temperatures it is more non-linear, which is not shown.

During the fitting (in step 2 and step 4),  $\omega_{1,2}$  and S is calculated using Eq.(D.4) where the dielectric response function of the top and bottom dielectric is the same (both are hBN) and equal to the dielectric function of the hBN  $\epsilon_{top}(\omega) = \epsilon_{bottom}(\omega) = \epsilon_{hBN}(\omega)$ , which results the equation of

$$2\epsilon_{\rm hBN}(\omega) = 0. \tag{7.10}$$

Substituting Eq.(D.1) in Eq.(7.10) with the parameters of hBN, which are  $\epsilon_{\text{hBN}}^0 = 5.09$ ,  $\epsilon_{\text{hBN}}^\infty = 4.1$ ,  $\epsilon_{\text{hBN}}^i = 4.58$ ,  $\hbar\omega_{TO1} = 97.08 \text{ meV}$  and  $\hbar\omega_{TO2} = 187.22 \text{ meV}[328, 331]$  leads to

$$0 = \epsilon_{\rm hBN}^{\infty} + \left(\epsilon_{\rm hBN}^{0} - \epsilon_{\rm hBN}^{i}\right) \frac{\omega_{TO1}^{2}}{\omega_{TO1}^{2} - \omega^{2}} + \left(\epsilon_{\rm hBN}^{i} - \epsilon_{\rm hBN}^{\infty}\right) \frac{\omega_{TO2}^{2}}{\omega_{TO2}^{2} - \omega^{2}},\tag{7.11}$$

which has two solution for  $\omega$  that gives the surface optical phonon modes. The solutions are  $\hbar\omega_1 = 102.13 \text{ meV}$  and  $\hbar\omega_2 = 198.29 \text{ meV}$ . The ratio of the coupling strengths of the surface optical modes is calculated from Eq.(D.5) as

$$S = \frac{\omega_2}{\omega_1} \left( \frac{1}{\epsilon_{\rm hBN}^{\infty} + \epsilon_{\rm hBN}^i} - \frac{1}{2\epsilon_{\rm hBN}^i} \right) / \left( \frac{1}{2\epsilon_{\rm hBN}^i} - \frac{1}{\epsilon_{\rm hBN}^0 + \epsilon_{\rm hBN}^i} \right), \tag{7.12}$$

which gives S = 2.04.



Figure 7.11: The extracted fitting coefficients of the global fitting of  $\Delta \rho_{xx}(T, n)$  with Eq.(7.9) as a function of pressure on device A in the hole regime. Coefficients of  $\gamma$  and B are shown in panel (a) and (b), respectively.

The result of the global fitting with  $\alpha = 1$ , which is obtained from the third step of the fitting, is shown in Fig. 7.9a with red and blue dashed lines for the electron and hole regime, respectively and is also shown in Fig. 7.9b with a black dashed line at  $n = -0.8 \cdot 10^{12} \text{ cm}^{-2}$ . The fitted curves follow the measured data except below  $T \leq 10 \text{ K}$ , which outside of the fitting regime. In that region,  $T < T_{\text{BG}}$ , and the linear T dependency of the acoustic phonon-electron coupling is no longer valid (see Section 2.4.3). A small deviation is also expected from other temperature dependent scattering mechanisms such as short and long-ranged scattering, which is discussed in Section 2.4.3. The fitted parameters for the hole side are shown in Fig. 7.11 with blue markers. The uncertainty of the fitting parameters comes from the uncertainty of the fitting and their deviation by varying the fitting range in n and T. From  $\gamma$ , which is shown in panel (a), the acoustic deformation potential  $D_A$  can be extracted using Eq.(2.64). By substituting  $\rho_m = 7.6 \cdot 10^{-7} \text{ kg/m}^{-2}$  and  $v_{ph} = 2 \cdot 10^4 \text{ m/s}[185]$ ,  $D_A = 15.7 \pm 0.1 \text{eV}$ . The obtained acoustic deformation potential is in range of the previously obtained values in the literature [143, 177, 178, 180–183]. The parameter B is shown in Fig. 7.11b. In case of  $\alpha = 1$ , according to Eq.(S12) in Ref.[193], B can be estimated with

$$B = \frac{g_1}{4\hbar v_{\rm F}^2 d(\sqrt{2} + a^2)^2},\tag{7.13}$$

where d is the interlayer distance of hBN and graphene,  $a = e^2/\epsilon_{avg}\pi\hbar v_{\rm F}$  is a dimensionless constant with  $\epsilon_{avg} = \epsilon_{\rm hBN}^0$  and  $g_1$ , which is the electron-phonon coupling strength, which is twice of Eq.(D.5), because of the two hBN layers, given as

$$g_1 = \frac{\hbar\omega_1}{\epsilon_0} \left( \frac{1}{2\epsilon_{\rm hBN}^i} - \frac{1}{\epsilon_{\rm hBN}^0 + \epsilon_{\rm hBN}^i} \right).$$
(7.14)

As the only unknown parameter in Eq.(7.13) is the interlayer distance, d can be estimated from B. From the fitting,  $d = 3.1 \pm 0.2$ Å, which is in the range of the theoretically expected value of 3.24Å[332, 333] between the hBN and the graphene.

To study the pressure dependence of the electron-phonon interaction, similar measurements are made at p = 1 GPa and at p = 2 GPa.  $\Delta \rho_{xx}$  is shown in Fig 7.9b as a function of the temperature at  $n = -0.8 \cdot 10^{12} \text{ cm}^{-2}$  at p = 1 GPa and at p = 2 GPa with teal and red dots, respectively. At intermediate temperatures, where the acoustic phonons dominate the transport, the curves coincide, from which I expect only a very small pressure dependence of the acoustic phonon-electron coupling. An increase of the resistance with pressure can be observed at larger temperatures, where the RIP scattering starts to dominate the transport, from which it is expected that, the pressure increases the effect of the RIP scattering.

To perform similar fitting as at ambient pressure, with the only free parameters of B and  $\gamma$ , the pressure dependence of the polar surface phonon frequencies and the ratio between them is calculated from the pressure dependence of the dielectric function and the TO modes of the hBN. The pressure dependence of the dielectric constant is  $\epsilon_{\rm hBN}(p) \approx \epsilon_{\rm hBN}(p = 0) + 0.047p$ , where p is in GPa[334]. The pressure dependence of the TO phonon modes is calculated with  $\omega_{\rm TOi}(p) = \omega_{\rm TOi}(p = 0) + c_i p$ ,

p (GPa)	0	1	2
$\hbar\omega_1 \;(\mathrm{meV})$	102.13	101	99.98
$\hbar\omega_2 \;(\mathrm{meV})$	198.29	198.68	199.07
S	2.04	2.06	2.09

Table 7.2: Calculated surface optical phonon frequencies of a hBN/SLG/hBN heterostructure with Eq.(7.11) and ratio of their coupling strengths using Eq.(7.12) at different pressures. The pressure dependency of the parameters of the hBN is taken from Ref.[334].

where  $c_1 = -1.03 \text{ meV/GPa}^5$  and  $c_2 = 0.5 \text{ meV/GPa}^6$ , which are determined from pressure dependent Raman measurements in Ref.[334]. From these, the calculated surface polar modes and the ratio between them are collected in Table 7.2. With the calculated parameters, the fitting procedure is performed at each pressure, where I fixed  $\alpha = 1$  as within the range of uncertainty, step 3 gives  $\alpha = 1$  also at finite pressures. The result of the fitting is shown with dashed lines in Fig 7.9b.

The fitted parameters at p = 1 GPa and p = 2 GPa are shown in Fig. 7.11 for the hole regime.  $\gamma$ , which is shown in panel (a), shows a decreasing tendency, however, within the uncertainty of the fits it is insensitive to pressure. As  $\gamma$  is inversely proportional to the mass density of graphene, a small decrease of  $\gamma$  is expected during compression. However, since the acoustic deformation potential is inversely proportional to the lattice constant of graphene[180, 187], a small increase of  $\gamma$  is also expected.

An increasing tendency of B can be observed with pressure as shown in Fig. 7.11b. The increase of B is expected from Eq.(7.13). The estimated interlayer distance from B in the hole regime results  $d(p = 1 \text{ GPa}) = 2.7 \pm 0.2 \text{ Å}$  and  $d(p = 2 \text{ GPa}) = 2.5 \pm 0.2 \text{ Å}$ . The decreasing tendency of the interlayer distance with pressure is consistent with the theory[335]. However, in the measurements, the decrease is larger than an averaged interlayer distance decrease with pressure in DFT calculations in Ref.[335].

Similar measurements are also performed in device D, which is discussed in Appendix E.7. The results are similar to in device A in this section: the main finding is that the pressure enhances the RIP coupling (B), and the acoustic phonon-electron coupling  $(\gamma)$  is independent of the pressure within the uncertainty of our measurements. I note that, in other devices and in the electron regime, no clear linear behavior is observed thus the fitting procedure presented in this section could not be done reliably.

<sup>&</sup>lt;sup>5</sup>The phonon mode of  $A_{2u}(TO)$ .

<sup>&</sup>lt;sup>6</sup>The phonon mode of  $E_{1u}(TO)$ .

### 7.4 Conclusion

In conclusion, in high-mobility hBN/SLG/hBN devices, the long-range scattering is the main contribution to the resistivity at small temperatures, which can come from charge impurities far from the graphene or from corrugations. At finite pressures, I found that by increasing the pressure, the mobility decreases due to the increase of the scattering rates of both short- and long-ranged scatterers. From in-plane magnetic field-dependent WL measurement I found that the volume of ripples increased with pressure, which is an indication of the enhanced scattering of electrons on ripples with pressure. At finite temperatures, from TMF experiments, I found that the electron-electron scattering and the acoustic phonon-electron scattering are insensitive to pressure. The latter is also verified with zero-magnetic field transport measurements. Finally, it is found that the RIP scattering is enhanced with pressure, which further decreases the mobility at large temperatures. These results can be important in understanding the main source of the pressure-dependent conductivity for novel devices such as graphene-based pressure sensors.

## 8. Chapter

## Summary

Pressure offers a versatile tool to investigate and tune the properties of van der Waals (vdW) heterostructures as in these structures the interlayer vdW forces are much weaker than the covalent bonds, which allows an efficient modulation of the interlayer spacing[38]. As the properties of the vdW heterostructures are highly affected by the interlayer interactions, with pressure their properties can be tuned. This thesis mainly focuses on the effect of hydrostatic pressure applied on graphene-based van der Waals materials. The vdW materials are investigated with electrical transport measurements. I focus on the electronic and transport properties such as the evolution of the band gaps, the change of the spin-orbit coupling strength and the change of the scattering mechanisms by applying hydrostatic pressure.

In the first part of the thesis, I give a brief introduction to graphene and the band structure of twisted bilayer graphene along with a short description of the most relevant transport properties of graphene and the phenomena behind the transport measurement methods used in this thesis. In Chapter 3, I give a brief summary of experimental methods, such as sample fabrication and low-temperature measurements.

The electronic properties of twisted double bilayer graphene around the magic angle are investigated under pressure in Chapter 5. From both thermal activation and bias voltage-dependent measurements, I extracted the single-particle moiré gaps, which were found to decrease and fully close by increasing the pressure. To verify this theoretically, I calculated the pressure dependence of the moiré band gaps with the Bistritzer-MacDonald model in Chapter 4, which qualitatively agreed with the experiments. From magnetotransport measurements, I verified the insensitivity of the twist angle to pressure by analyzing the Brown-Zak oscillations. Moreover, I also observed the signature of the decrease of the correlations, as at the half filling the gaps due to correlations close with pressure and I also observed an unusual magnetic field dependence of the gap at the charge neutrality.

The proximity-induced spin-orbit coupling in  $WSe_2$  and BLG-based heterostructures is investigated under hydrostatic pressure in Chapter 6. The heterostructures are studied with low-temperature magnetotransport measurements. From Shubnikovde Haas oscillations, I observed two Fermi surfaces of the spin split bands due to the SOC. I calculated the low-energy band structure of the heterostructure in Chapter 4, which I used to calculate the Fermi surfaces at finite SOC. I found that at large charge densities, the difference of the bands is mostly determined by the Rashba-type SOC. To obtain the Rashba-type SOC strength, I fitted the model on the experiments where I found a large increase of the coupling strength with increasing pressure. I also obtained the Ising-type coupling strength from quantum Hall measurements. For this, I measured the positions of the Landau level crossings at different magnetic fields. I calculated the Landau level energies numerically and fitted the measured crossing points at  $\nu = \pm 3$  filling factor to obtain the Ising-type SOC, which increased a large amount with applying pressure.

The transport properties of high-mobility devices, made of single-layer graphene, which is encapsulated within hBN crystals, are investigated under pressure in Chapter 7. From field effect measurements, I observed that the main scattering mechanism that is responsible for the resistance of graphene at low temperatures is the long-ranged scattering and it remains dominant under pressure. I also verified this with Shubnikov-de Haas oscillation measurements. I observed an increase in both the short- and long-range scattering by increasing the pressure, which led to a decrease in the mobility of the charge carriers. I observed with weak localization measurements the increase of the volume of ripples by increasing the pressure, which is consistent with the increased scattering rates with pressure. I also observed in the weak localization measurements that the short-range scattering is mainly due to the sample edges and their pressure dependence is negligible. I observed in magnetic focusing experiments, that the pressure doesn't change the scattering mechanism and has a negligible effect on the combined contribution of the electron-electron interactions and the acoustic phonon-electron coupling to the dephasing of the magnetic focusing signal. With temperature-dependent field effect measurements, I observed that the acoustic phonon-electron coupling has negligible pressure dependence and I showed that the remote interfacial phonon-electron coupling increases with pressure.

The experimental findings in this thesis give an insight into how important the interlayer interactions are in vdW heterostructures and their tunability with pressure. In the near future, I believe a vast amount of similar studies on various heterostructures will be executed especially focusing on twisted structures, where most of their properties are the result of the interlayer coupling. I believe that these results will be beneficial in the field of graphene, especially in graphene spintronics and twistronics.

## Thesis points

- 1. I showed for the first time the extensive tunability of the moiré gaps of twisted double bilayer graphene with pressure in agreement with the theory. I performed temperature and bias voltage-dependent transport experiments. From the thermal activation and bias voltage-dependent measurements, I measured the moiré gaps of a TDBG near the magic angle and showed that the moiré gaps in a TDBG can be decreased and fully closed by applying hydrostatic pressure. Furthermore, I showed by measuring Brown-Zak oscillations that, the twist angle doesn't change with pressure. Finally, I also observed a decrease in the correlation effects by increasing the pressure. [T1]
- 2. I showed for the first time in WSe<sub>2</sub>/BLG heterostructures the enhancement of the proximity-induced Rashba-type and Ising-type SOC with hydrostatic pressure in agreement with the theory. I made low-temperature magnetic field-dependent experiments. In the experiments, I used the Shubnikov-de Haas oscillations to obtain the Rashba-type SOC strength, and I used the quantum Hall effect to obtain the Ising-type SOC strength by measuring the positions of the Landau level crossings. I showed that with pressure, the positions of the crossing points change and also I showed that the pressure increases the splitting of the Fermi surface, which is due to the lifted spin-degeneracy of the SOC. From these, I found that the spin-orbit coupling strength increased by more than 50% under pressure. [T2]
- 3. I showed the decrease and closing of the moiré gaps in twisted double bilayer graphene with pressure with simulations, which showed a good agreement with the experiments. I simulated the change of the Fermi surfaces of WSe<sub>2</sub>/BLG heterostructures by varying the Rashbatype SOC and I calculated the change of the Landau level crossings by varying the Ising-type SOC, which I used to obtain the SOC strengths by fitting the experimental data with my simulations. I successfully applied the Bistritzer-MacDonald model on twisted double bilayer graphene, where I used the pressure dependence of the interlayer tunneling from the literature to calculate the band structure at different pressures. I found that, by increasing the pressure, the moiré gaps decrease and fully close, which is in good agreement

with my experiments. In the case of WSe<sub>2</sub>/BLG heterostructures, I calculated band structure from a low-energy model. From the band structure, I calculated the Fermi surfaces at different Rashba-type coupling strengths. Then I fitted the experimentally obtained Fermi surfaces with the model and obtained the Rashba-type SOC strength. From the low-energy model of the heterostructure, I calculated numerically the Landau level energies as a function of an applied external electric field. I calculated the change of the positions of the Landau level crossing by varying the Ising-type SOC strength. I fitted the model on the experimentally determined Landau level positions and obtained the Ising-type SOC strength. Furthermore, I also modeled a WSe<sub>2</sub>/BLG/WSe<sub>2</sub> heterostructure, where the Ising-type SOC is the opposite of the two graphene layers. I found that, by increasing the electric field, the SOC-induced gap closes and reopens, and the closing point could be used to obtain the Ising-type SOC strength. [T1, T2, T3]

4. I showed that the mobility of charge carriers in high-mobility singlelayer graphene decreases with pressure due to the increased shortrange and long-range scattering and also the increased effect of remote interfacial phonon coupling with hydrostatic pressure. From field effect and Shubnikov-de Haas oscillations measurements, I showed that in highmobility devices, the long-range scattering is the main source of the resistance and it remains dominant under pressure. From weak localization measurements, I found that in high-mobility devices, the short-range scattering is mainly caused by the edges of the sample which is insensitive to the pressure. I showed with in-plane magnetic field-dependent weak localization measurements, that the volume of the corrugations increased by increasing the pressure. From field effect measurements, I showed the increase of short-range and long-range scattering with pressure. From temperature-dependent magnetic focusing experiments, I found that the pressure has a negligible effect on the combined effect of the electron-electron interactions and the acoustic phonon-electron coupling. From temperature-dependent field-effect transport measurements, I showed that the remote interfacial phonon-electron coupling increases with pressure. [T4]

### Publications related to thesis points

T1 Bálint Szentpéteri, Peter Rickhaus, Folkert K. de Vries, Albin Márffy, Bálint Fülöp, Endre Tóvári, Kenji Watanabe, Takashi Taniguchi, Andor Kormányos, Szabolcs Csonka and Péter Makk, Tailoring the band structure of twisted double bilayer graphene with pressure. Nano Letters 21(20), 8777–8784 (2021)

- T2 Bálint Szentpéteri, Albin Márffy, Máté Kedves, Endre Tóvári, Bálint Fülöp, István Kükemezey, András Magyarkuti, Kenji Watanabe, Takashi Taniguchi, Szabolcs Csonka and Péter Makk, Increasing the proximity-induced spin-orbit coupling in bilayer graphene/WSe<sub>2</sub> heterostructures with pressure. Submitted to Physical Review B, arXiv:2409.20062
- T3 Máté Kedves, Bálint Szentpéteri, Albin Márffy, Endre Tóvári, Nikos Papadopoulos, Prasanna K. Rout, Kenji Watanabe, Takashi Taniguchi, Srijit Goswami, Szabolcs Csonka and Péter Makk, Stabilizing the Inverted Phase of a WSe<sub>2</sub>/BLG/WSe<sub>2</sub> Heterostructure via Hydrostatic Pressure. *Nano Letters* 23(20), 9508–9514 (2023)
- T4 Bálint Szentpéteri *et al.* Origin of the reduced charge carrier mobility in graphene under pressure. Manuscript in preparation.

## Additional unrelated publications

- T5 Zoltán Kovács-Krausz, Anamul Md Hoque, Péter Makk, Bálint Szentpéteri, Mátyás Kocsis, Bálint Fülöp, Michael Vasilievich Yakushev, Tatyana Vladimirovna Kuznetsova, Oleg Evgenevich Tereshchenko, Konstantin Aleksandrovich Kokh, István Endre Lukács, Takashi Taniguchi, Kenji Watanabe, Saroj Prasad Dash, and Szabolcs Csonka, Electrically Controlled Spin Injection from Giant Rashba Spin–Orbit Conductor BiTeBr. Nano Letters 20(7), 4782–4791 (2020)
- T6 Bálint Fülöp, Albin Márffy, Simon Zihlmann, Martin Gmitra, Endre Tóvári, Bálint Szentpéteri, Máté Kedves, Kenji Watanabe, Takashi Taniguchi, Jaroslav Fabian, Christian Schönenberger, Péter Makk, and Szabolcs Csonka, Boosting proximity spin–orbit coupling in graphene/WSe<sub>2</sub> heterostructures via hydrostatic pressure. npj 2D Materials and Applications 5, 82 (2021)

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# A. Appendix

## Fabrication processes

### A.1 Assembly of van der Waals heterostructures

- 1. A PC film is prepared on a glass slide: a drop of PC solution is dripped on the glass slide then with a second glass slide the solution is dispersed homogeneously on both slides by pressing them together. The PC is dissolved in chloroform, and the solid content is  $\sim 3\%$ .
- 2. The thin PC layer is transferred on a PDMS cube mounted on a glass slide with scotch tape. The PDMS is made from two parts: Sylgard 184 base and EPS cure with a rate of 10:1.
- 3. Exfoliate the crystals on a  $Si/SiO_2$  substrate
- 4. Pick up the layers with the PC stamp at 60 100 °C
- 5. Stamp the completed stack on a  $Si/SiO_2$  substrate by heating it to  $180 \,^{\circ}C$
- 6. Anneal the stack at  $180\,^{\circ}\mathrm{C}$  for  $5\,\mathrm{min}$
- 7. Remove the PC with chloroform  $\sim 10 \text{ min}$

### A.2 EBL and development

Most commonly polymers are used as resist like poly(methyl-methacrylate) (PMMA). The PMMA is dissolved in ethyl-lactate. Here I gave a recipe for a 300 nm thick PMMA coating. PMMA 600K is used with a solid content of 4.7%. The receipt for EBL is:

- 1. Clean the wafer and preheat it for about 1 min at 180 °C. Let it cool down.
- 2. Spin-coat PMMA (for 300 nm 4000 rpm for 40 sec.) Bake at 180 °C for 3 min.
- 3. Expose the sample with electron beam (U=20 keV, Dose $\approx$ 560 µC cm<sup>-2</sup> for cold development and Dose $\approx$ 360 µC cm<sup>-2</sup> for MIBK:IPA
- 4. Cold-development in IPA:H<sub>2</sub>O (7:3 ratio) at  $0^{\circ}$ C for 60 sec then blow-dry

The regular developer is MIBK:IPA (1:3) for 60 sec, and then the stopper is IPA for 30 sec. However, the PMMA can be cracked on the heterostructures. To avoid it, a low-stress developer like the IPA:H<sub>2</sub>O is advised[336].

### A.3 Evaporation of contacts and top gates

To contact the graphene, Cr is used. For the top gate, Ti is used as an adhesive layer.

- 1. EBL and development
- 2. Evaporate 5+10 nm Cr (rate: 1 Å/s). The first 5 nm with a closed sample shutter to clean the surface of the metal.
- 3. Evaporate 80-110 nm Au (rate: 4-5 Å/s)

For the Ti adhesive layer, the receipt is the same as the receipt for Cr (5+10 nm with 1 Å/s).

### A.4 Reactive ion etching

#### A.4.1 $CHF_3/O_2$ plasma

Parameters are CHF<sub>3</sub>/O<sub>2</sub> (40/4 sccm), P = 60 W,  $p_{\text{work}} = 60$  mTorr,  $p_{\text{base}} = 5 \cdot 10^{-5}$  mbar and T = 20 °C. This is used to define the edge contacts. The etching rates are

SiO<sub>2</sub>: ~ 10 nm/min PMMA: ~ 50 nm/min graphite: ~ 7 nm/min hBN: ~ 25 nm/min WSe<sub>2</sub>: ~ 24 nm/min

#### A.4.2 $SF_6/Ar/O_2$ plasma

Parameters are SF<sub>6</sub>/Ar/O<sub>2</sub> (50/5/5 sccm), P = 60 W,  $p_{\text{work}} = 25$  mTorr,  $p_{\text{base}} = 5 \cdot 10^{-5}$  mbar, T = 20 °C and t = 30 sec. This is used to shape the device. The etching rates are

SiO<sub>2</sub>:  $\sim 14 \text{ nm/min}$ PMMA:  $\sim 170 \text{ nm/min}$ graphite:  $\sim 12 \text{ nm/min}$  hBN: > 200 nm/min

 $WSe_2: > 200 \text{ nm/min}$ 

#### A.4.3 $Ar/O_2$ plasma

Parameters are Ar/O<sub>2</sub> (22/8 sccm), P = 30 W,  $p_{\text{work}} = 25$  mTorr,  $p_{\text{base}} = 5 \cdot 10^{-5}$  mbar and T = 20 °C. The etching rates are

 ${\rm SiO}_2: < 2\,{\rm nm}/{\rm min}$ 

PMMA:  $\sim 70 \,\mathrm{nm/min}$ 

graphite:  $\sim 10\,\mathrm{nm}/\mathrm{min}$ 

hBN:  $< 2 \,\mathrm{nm}/\mathrm{min}$ 

 $WSe_2: < 2 \, nm/min$ 

#### A.4.4 SF<sub>6</sub> plasma

Parameters are SF<sub>6</sub> (40 sccm), P = 30 W,  $p_{\text{work}} = 30$  mTorr,  $p_{\text{base}} = 5 \cdot 10^{-5}$  mbar and T = 20 °C. The etching rates are

PMMA:  $\sim 80 \text{ nm/min}$ graphite: < 1 nm/minhBN: > 200 nm/minWSe<sub>2</sub>: > 200 nm/min

#### A.4.5 AlO<sub>x</sub> deposition

The AlO<sub>x</sub> layer is deposited with atomic layer deposition (ALD). The recipe for a 30 nm thick layer is 300 cycles, T = 225 °C, two components: water and TMA. Before the deposition, a 2 min O<sub>2</sub> plasma cleaning at P = 60 W is done.

## **B.** Appendix

## Derivation of LL energies in BLG

The ladder operators act on the Landau level wavefunctions as  $\hat{a} |n\rangle = \sqrt{n} |n-1\rangle$ and  $\hat{a}^{\dagger} |n+1\rangle = \sqrt{n+1} |n+1\rangle$ . To solve Eq.(4.3), the same ansatz can be used as in Ref. [270]:

$$|+, n, s_{z}\rangle = (|A1, \uparrow\rangle |n\rangle, |A1, \downarrow\rangle |n-1\rangle, |B1, \uparrow\rangle |n-1\rangle, |B1, \downarrow\rangle |n-2\rangle, \quad (B.1)$$

$$|A2, \uparrow\rangle |n-1\rangle, |A2, \downarrow\rangle |n-2\rangle, |B2, \uparrow\rangle |n-2\rangle, |B2, \downarrow\rangle |n-3\rangle),$$

$$|-, n, s_{z}\rangle = (|A1, \uparrow\rangle |n-2\rangle, |A1, \downarrow\rangle |n-3\rangle, |B1, \uparrow\rangle |n-1\rangle, |B1, \downarrow\rangle |n-2\rangle, (B.2)$$

$$|A2, \uparrow\rangle |n-1\rangle, |A2, \downarrow\rangle |n-2\rangle, |B2, \uparrow\rangle |n\rangle, |B2, \downarrow\rangle |n-1\rangle).$$

Using these, the LL energies for  $n\geq 3$  are given by the eigenvalues of  $H_{n\geq 3}^{\xi}+H_Z$  with the matrices of



where  $\hbar\omega_i = v_i \sqrt{2eB\hbar}$  is introduced for simplicity and  $H_Z = E_Z s_z$ .

Using the ansatz also allows to obtain the zeroth LLs from the following Hamiltonians. For n = 0, the solution is given by  $H_0^K = (u + \lambda_{\rm I}^b)/2 + E_Z$  and  $H_0^{K'} = -(u + \lambda_{\rm I}^t)/2 + E_Z$ . For n = 1, the solution is given by

$$H_{1}^{K} = \begin{pmatrix} \frac{1}{2}(u + \lambda_{1}^{b}) + E_{Z} & 0 & \hbar\omega_{0} & -\hbar\omega_{4} \\ 0 & \frac{1}{2}(u - \lambda_{1}^{b}) - E_{Z} & i\lambda_{R}^{b} & 0 \\ \hbar\omega_{0} & -i\lambda_{R}^{b} & \frac{1}{2}(u + \lambda_{1}^{b}) + \Delta' + E_{Z} & \gamma_{1} \\ -\hbar\omega_{4} & 0 & \gamma_{1} & \frac{1}{2}(-u + \lambda_{1}^{t}) + \Delta' + E_{Z} \end{pmatrix},$$
(B.3)  
$$H_{1}^{K'} = \begin{pmatrix} \frac{1}{2}(u - \lambda_{1}^{b}) + \Delta' + E_{Z} & \gamma_{1} & \hbar\omega_{4} & 0 \\ \gamma_{1} & \frac{1}{2}(-u - \lambda_{1}^{t}) + \Delta' + E_{Z} & -\hbar\omega_{0} & i\lambda_{R}^{t} \\ \hbar\omega_{4} & -\hbar\omega_{0} & \frac{1}{2}(-u - \lambda_{1}^{t}) + E_{Z} & 0 \\ 0 & -i\lambda_{R}^{t} & 0 & \frac{1}{2}(-u + \lambda_{1}^{t}) - E_{Z} \end{pmatrix}.$$
(B.4)

For n = 2, the matrices can be written as

$$H_2^K = \begin{pmatrix} \frac{1}{2}(u+\lambda_1^b) & 0 & \hbar\omega_0\sqrt{2} & 0 & -\hbar\omega_4\sqrt{2} & 0 & 0 \\ 0 & \frac{1}{2}(u-\lambda_1^b) & i\lambda_R^b & \hbar\omega_0 & 0 & -\hbar\omega_4 & 0 \\ \hbar\omega_0\sqrt{2} & -i\lambda_R^b & \frac{1}{2}(u+\lambda_1^b) + \Delta' & 0 & \gamma_1 & 0 & -\hbar\omega_4 \\ 0 & \hbar\omega_0 & 0 & \frac{1}{2}(u-\lambda_1^b) + \Delta' & 0 & \gamma_1 & 0 \\ -\hbar\omega_4\sqrt{2} & 0 & \gamma_1 & 0 & \frac{1}{2}(-u+\lambda_1^t) + \Delta' & 0 & \hbar\omega_0 \\ 0 & -\hbar\omega_4 & 0 & \gamma_1 & 0 & \frac{1}{2}(-u-\lambda_R^t) + \Delta' & i\lambda_R^t \\ 0 & 0 & -\hbar\omega_4 & 0 & \gamma_1 & 0 & \hbar\omega_0 \\ -\hbar\omega_0 & \frac{1}{2}(u-\lambda_1^b) + \Delta' & 0 & \gamma_1 & 0 & \hbar\omega_4 \\ 0 & 0 & -\hbar\omega_4 & 0 & \gamma_1 & 0 & \hbar\omega_4\sqrt{2} & 0 \\ -i\lambda_R^b & 0 & \frac{1}{2}(u+\lambda_1^b) + \Delta' & 0 & \gamma_1 & 0 & \hbar\omega_4 \\ \hbar\omega_4 & \gamma_1 & 0 & \frac{1}{2}(-u-\lambda_1^t) + \Delta' & 0 & -\hbar\omega_0\sqrt{2} & i\lambda_R^t \\ 0 & 0 & \gamma_1 & 0 & \frac{1}{2}(-u-\lambda_1^t) + \Delta' & 0 & -\hbar\omega_0 \\ 0 & \hbar\omega_4\sqrt{2} & 0 & -\hbar\omega_0\sqrt{2} & 0 & \frac{1}{2}(-u-\lambda_1^t) & 0 \\ 0 & 0 & \hbar\omega_4 & -i\lambda_R^t & -\hbar\omega_0 & 0 & \frac{1}{2}(-u+\lambda_1^t) \end{pmatrix} \end{pmatrix} + H_{2Z}^{K'},$$
 where

$$H_{2Z}^{K} = \begin{pmatrix} E_{Z} & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & -E_{Z} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & E_{Z} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -E_{Z} & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & E_{Z} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -E_{Z} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & -E_{Z} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & E_{Z} \end{pmatrix} \quad \text{and} \quad H_{2Z}^{K} = \begin{pmatrix} E_{Z} & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & E_{Z} & 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & -E_{Z} & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & -E_{Z} & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & E_{Z} & 0 \\ 0 & 0 & 0 & 0 & 0 & 0 & 0 & -E_{Z} \end{pmatrix}.$$

$$(B.7)$$

# C. Appendix

# Further details about $BLG/WSe_2$ heterostructures

Further details and measurements on  $BLG/WSe_2$  heterostructures are presented in this appendix.

## C.1 Further details of device A

The optical image of the flakes is shown in Fig C.1. From the angle of the triangles which is used to estimate the orientation of the crystals, the twist angle is likely around  $\vartheta \sim 16^{\circ}$ .



Figure C.1: (a) Optical microscope image of the measured stack. The scale bar is  $20 \,\mu\text{m}$ . The black and magenta lines show the perimeter of the graphene and the WSe<sub>2</sub>, respectively. The red triangle is an estimate of the crystal orientation of the graphene from its edges. The yellow triangle is an estimate of the crystal orientation of the WSe<sub>2</sub>. (b) Optical microscope image of the used WSe<sub>2</sub> showing the same yellow triangle as in (a). The scale bar is  $10 \,\mu\text{m}$ . (b) Optical microscope image of the used BLG showing the same red triangle as in (a). The scale bar is  $20 \,\mu\text{m}$ .

#### C.1.1 More FFT curves

Some FFT curves that were used to extract  $S_{\rm F}$  in Fig. 6.9b are shown in Fig. C.2. The two main peaks are more visible at large densities. Their splitting is decreasing at lower densities. The quality of the data measured at p = 2 GPa is worse likely due to the increased disorder with pressure.



Figure C.2: FFT of the measured SdH oscillations at different densities at D = 0 (a) at ambient pressure and (b) at p = 2 GPa.

### C.2 Device B

An optical image of device B is shown in the inset of Fig. C.3c. Its structure is similar to device A: a graphite bottom gate, a bottom hBN with the thickness of 17 nm, a WSe<sub>2</sub> with the thickness of 4 nm, a BLG, a top hBN with the thickness of 6 nm, a 30 nm thick  $AlO_x$  layer and a metallic top gate. From the optical micrograph, the twist angle of device B is  $\vartheta \sim 0^{\circ}$ .

Fig. C.3 shows longitudinal resistivity ( $\rho_{xx}$ ) of device B as a function of top and bottom gate voltages, plotted as a function of n and D at temperature T = 4.2 K at ambient pressure. Similarly to device A, lighter-colored regions of higher resistance correspond to the displacement-field-opened gap along the charge neutrality line. This



Figure C.3:  $\rho_{xx}$  of device B as a function of n and D at T = 4.2 K (a) at p = 0 GPa and (b) at p = 1.95 GPa. (c) Longitudinal resistivity as a function of D at n = 0 at 0 and 1.95 GPa pressures. Inset is an optical microscope image of device B with a scale bar of 5 µm.

p (GPa)	0	1.95
$\alpha_{\mathrm{TG}} \left( \frac{10^{15}}{\mathrm{Vm}^2} \right)$	6.50(6)	8.09(12)
$\alpha_{\rm BG} \left( \frac{10^{15}}{{ m Vm}^2} \right)$	7.23(1)	9.71(6)
$n_0 \ (10^{15}/{\rm m}^2)$	5.23(5)	6.32(5)
$D_0/\epsilon_0 ~({\rm V/nm})$	0.080(2)	0.063(3)

Table C.1: The extracted parameters of device B for the calculation of n and D at p = 0 GPa and p = 1.95 GPa with the uncertainty of the last digit in the bracket originating from the measurement resolution, the readout precision and from the fitting.

gap is increasing by increasing |D|. At p = 1.95 GPa the resistance is increased with pressure as shown in Fig. C.3b. A line-cut along n = 0 is shown in Fig. C.3c, where  $\rho_{xx}$ depend non-monotonously on D. There is a local maximum at D = 0 and minima at  $D/\epsilon_0 \sim 11 \text{ mV/nm}$  which shift to  $D/\epsilon_0 \sim 18 \text{ mV/nm}$  at p = 1.95 GPa. Here as opposed to device A, this feature is observable at p = 1.95 GPa. The lever arms were determined similarly as in device A and the increase of the lever arms with pressure is also similar as shown in Table C.1.

In this device, only the Ising-type SOC is determined from  $\nu = \pm 3$  LL crossings. In Fig. C.4a  $\rho_{xx}$  as a function of n and D is shown at B = 14 T at p = 0 GPa. The LL crossings are observed similarly as in device A. Remeasuring the same map at p = 1.95 GPa (Fig. C.4b), the LL crossings are more blurred than at p = 0 GPa similarly as before. The extracted position of the crossings is shown in Fig. C.4c. Here, the Ising-type SOC is fitted, where a sublattice asymmetry term  $H_{asy} = u_{asy}\sigma_z$  is also included, which shifts all the LLs with  $u_{asy}$  without any mixing. This term takes into account the on-site potential difference of the BLG. From the fitting on device B (see Fig. C.4c),  $\lambda_{\rm I}^{\rm b}(p = 0) = 1.25(10)$  meV with  $u_{asy}(p = 0) = -0.22$  meV and  $\lambda_{\rm I}^{\rm b}(p = 1.95$  GPa) = 1.7(1) meV with  $u_{asy}(p = 1.95$  GPa) = -0.42 meV.



Figure C.4: (a-b)  $\rho_{xx}$  of device B as a function of n and D at T = 4.2 K at B = 14 T (a) at p = 0 GPa and (b) at p = 1.95 GPa. (c) Extracted crossing points from the quantum Hall measurements at zero and at 1.95 GPa. The dashed lines show the corresponding fits with  $\lambda_{I}(p = 0) = 1.25$  meV and  $\lambda_{I}(p = 1.95$  GPa) = 1.7 meV respectively.

### C.3 Device C

An optical image of device C is shown in the inset of Fig. C.5c. Its structure is similar to device A: a graphite bottom gate, a bottom hBN with the thickness of 33 nm, a WSe<sub>2</sub> with the thickness of 4 nm, a BLG, a top hBN with the thickness of 30 nm, a 30 nm thick AlO<sub>x</sub> layer, and a metallic top gate. From the optical micrograph, the twist angle of device C is  $\vartheta \sim 6^{\circ}$ .



Figure C.5:  $\rho_{xx}$  of device C as a function of n and D at T = 4.2 K (a) at p = 0 GPa and (b) at p = 1.95 GPa. (c) Longitudinal resistivity as a function of D at n = 0 at 0 and 1.8 GPa pressures. The red curve is measured at 1.8 K Inset is an optical microscope image of device C with a scale bar of 5 µm.

Fig. C.5a shows longitudinal resistivity of device C as a function of n and D at temperature T = 4.2 K at ambient pressure. Similarly to devices A and B, lightercolored regions of higher resistance correspond to the displacement-field-opened gap along the charge neutrality line. This gap is increasing by increasing |D|. At p = 1.8 GPa

p (GPa)	0	1.8
$\alpha_{\rm TG} \left( \frac{10^{15}}{{ m Vm}^2} \right)$	3.34(4)	3.54(4)
$\alpha_{\rm BG} \left( \frac{10^{15}}{\rm Vm^2} \right)$	3.70(4)	4.09(5)
$n_0 \ (10^{15}/\mathrm{m}^2)$	2.79(4)	2.75(4)
$D_0/\epsilon_0 ~({\rm V/nm})$	0.034(2)	0.058(2)

Table C.2: The extracted parameters of device C for the calculation of n and D at p = 0 GPa and p = 1.8 GPa with the uncertainty of the last digit in the bracket originating from the measurement resolution, the readout precision and from the fitting.

the resistance is increased with pressure as shown in Fig. C.5b. A line-cut along n = 0 is shown in Fig. C.5c, where  $\rho_{xx}$  depend non-monotonously on D. There is a local maximum at D = 0 and minima at  $D/\epsilon_0 \sim 12 \,\mathrm{mV/nm}$ . Under  $p = 1.8 \,\mathrm{GPa}$ , these features are not observable similarly to device A. The lever arms were determined similarly as in devices A and B and the increase of the lever arms with pressure is also similar as shown in Table C.2.



Figure C.6: (a-b)  $\rho_{xx}$  of device C as a function of n and D at T = 4.2 K at B = 14 T (a) at p = 0 GPa and (b) at p = 1.95 GPa. (c) Extracted crossing points from the quantum Hall measurements at zero and at 1.95 GPa. The dashed lines show the corresponding fits with  $\lambda_{\rm I}(p = 0) = 1.25$  meV and  $\lambda_{\rm I}(p = 1.8$  GPa) = 1.7 meV respectively.

In Fig. C.6a  $\rho_{xx}$  as a function of n and D is shown at B = 14 T at p = 0 GPa. The LL crossings are observed similarly as in devices A and B. Remeasuring the same map at p = 1.8 GPa (Fig. C.6b), the LL crossings are more blurred than at p = 0 GPa similarly as before. The extracted position of the crossings is shown in Fig. C.6c. Here, the Ising-type SOC is fitted, where a sublattice asymmetry term  $H_{asy} = u_{asy}\sigma_z$  is also included, which shifts all the LLs with  $u_{asy}$  without any mixing. This term takes into account the on-site potential difference of the BLG. From the fitting on device B (see Fig. C.4c),  $\lambda_{\rm I}^{\rm b}(p = 0) = -1.7(1)$  meV with  $u_{asy}(p = 0) = -0.03$  meV and  $\lambda_{\rm I}^{\rm b}(p =$ 1.8 GPa) = -2.35(10) meV with  $u_{asy}(p = 1.8$  GPa) = -0.31 meV. The increase of the SOC is similar and consistent with the obtained values of devices A and B. The opposite sign is possible as it depends on the twist angle  $\vartheta$  as  $\lambda_{I}(\vartheta) = -\lambda_{I}(-\vartheta)[111]$  and it was seen in previous measurements[271].



Figure C.7: The difference of the Fermi surfaces as a function of n based on Shubnikovde Haas oscillations on device C. The blue points are at p = 0 GPa and the red points are at p = 1.8 GPa. The dashed lines are the fitting of the model with the corresponding  $\lambda_{\rm R}$ s. The colored ranges show the confidence interval of the fits.

On device C, Shubnikov-de Haas oscillation measurements are also performed and  $\lambda_{\rm R}$  is extracted the same way as on device A: I subtracted a quadratic background from the magnetoresistance. I Fourier-transformed it as a function of  $B^{-1}$ . I converted the frequency peaks to Fermi surface using  $S_{\rm F} = 2\pi e f_B/\hbar$ . I extracted the peaks that correspond to split Fermi surfaces of the sample at a given n. I took their difference, which is plotted in Fig. C.7. I calculated the Fermi surfaces with respect to n and used  $\lambda_{\rm R}$  as the only fitting parameter to fit the data. For this device  $\lambda_{\rm R}(p=0) = 12\pm 4$  meV, which increased with pressure to  $\lambda_{\rm R}(p=1.8 \,{\rm GPa}) = 20 \pm 4 \,{\rm meV}$ , though the data quality is lower than for device A presented in the main text.

# D. Appendix

# Calculation of the surface optical phonon modes

In an ionic dielectric, by considering only two modes exhibiting the strongest oscillator strength, the dielectric response function can be written as

$$\epsilon_{\rm ox}(\omega) = \epsilon_{\rm ox}^{\infty} + \left(\epsilon_{\rm ox}^0 - \epsilon_{\rm ox}^i\right) \frac{\omega_{TO1}^2}{\omega_{TO1}^2 - \omega^2} + \left(\epsilon_{\rm ox}^i - \epsilon_{\rm ox}^{\infty}\right) \frac{\omega_{TO2}^2}{\omega_{TO2}^2 - \omega^2},\tag{D.1}$$

where  $\omega_{TO1} \leq \omega_{TO2}$ ,  $\epsilon_{ox}^0 = \epsilon_{ox}(\omega = 0)$  is the static permittivity of the insulator,  $\epsilon_{ox}^{\infty} = \epsilon_{ox}(\omega \to \infty)$  is the optical permittivity of the ionic crystal,  $\epsilon_{ox}^i$  is an intermediate permittivity of the insulator and  $\omega_{TO1}$  and  $\omega_{TO2}$  are transverse optical phonon modes in the long-wavelength limit[337]. The value of  $\epsilon_{ox}^i = \epsilon_{ox}(\omega_{int})$  is related to an intermediate frequency  $\omega_{int}$  between the two TO modes and can be calculated using the Lyddane–Sachs–Teller relation, which determines the relation between the longitudinal and transverse optical phonon modes, which can be written for 2 longitudinal and 2 transverse optical phonon modes as [338]

$$\epsilon_{\rm ox}(\omega) = \epsilon_{\rm ox}^{\infty} \frac{(\omega_{\rm LO2}^2 - \omega^2)(\omega_{\rm LO1}^2 - \omega^2)}{(\omega_{\rm TO2}^2 - \omega^2)(\omega_{\rm TO1}^2 - \omega^2)},\tag{D.2}$$

where  $\omega_{\text{LO1}}$  and  $\omega_{\text{LO2}}$  are the longitudinal optical phonon modes in the long-wavelength limit.

By writing up the electrostatic boundary conditions (e.g. the continuity of the electric displacement field) at the interface of the dielectric medium and graphene, the surface optical phonon frequencies can be obtained[192, 193, 326, 327, 337, 339]. The surface optical phonon modes interact with the graphene plasmons, which modifies the surface optical phonon frequencies and leads to interfacial plasmon-phonon modes besides the surface optical phonon modes[329, 340]. However, the phonon-plasmon coupling is usually neglected in the literature[326–328]. By neglecting the plasmonic excitations of the graphene[329, 339, 340], if there is only one oxide, a secular equation

from the boundary conditions for the dielectric response function can be written as

$$\epsilon_{\rm ox}(\omega) + \epsilon_0 = 0, \tag{D.3}$$

where  $\epsilon_0$  is the vacuum permittivity. If there is also a second oxide layer on the top of the graphene, the secular equation is given by

$$\epsilon_{\rm bot}(\omega) + \epsilon_{\rm top}(\omega) = 0,$$
 (D.4)

where  $\epsilon_{top}(\omega)$  and  $\epsilon_{bot}(\omega)$  is the dielectric response function of the dielectric medium above and below the graphene defined with Eq.(D.1)[192, 339]. The solution of Eq.(D.3) or Eq.(D.4) gives the surface optical phonon modes of  $\omega_1$  and  $\omega_2$ .

The electron-phonon coupling strength  $(g_i)$  is proportional to

$$g_i^l = \frac{\hbar\omega_i}{2\epsilon_0} \left( \frac{1}{\epsilon_l^{hi} + \epsilon_{l'}^i} - \frac{1}{\epsilon_l^{lo} + \epsilon_{l'}^i} \right) \tag{D.5}$$

on each dielectric medium, where l = top/bottom indexes the dielectric, l' the dielectric on the other side of the graphene<sup>1</sup>. For i = 1,  $\epsilon_l^{hi} = \epsilon_l^i$  and  $\epsilon_l^{lo} = \epsilon_l^0$ , whereas for i = 2,  $\epsilon_l^{hi} = \epsilon_l^{\infty}$  and  $\epsilon_l^{lo} = \epsilon_l^i [329, 339]$ . If the dielectrics are different, the equations above result in 4 different surface modes with  $S^l = g_2^l/g_1^l$ . If the dielectrics are made of the same material, there are only two surface optical modes with twice the coupling strength and  $S = g_2/g_1$ . For example, using hBN as the bottom dielectric without dielectric above the graphene, from Eq.(D.3) the surface phonon frequencies are calculated as  $\hbar\omega_1 = 101 \text{ meV}$  and  $\hbar\omega_1 = 196 \text{ meV}$  and the ratio of their coupling strength from Eq.(D.5) results S = 2.17[328, 331].

<sup>&</sup>lt;sup>1</sup>If there is only one dielectric layer  $\epsilon_{l'}^i = \epsilon_0$ .

## E. Appendix

# Further measurements on high-mobility SLG devices

In this section, more measurement data is shown, which are performed on highmobility SLG devices. Optical micrographs of the devices are shown in Fig. 7.1a-d, and the details of them are given in Section 7.

### E.1 Calculating the charge density in SLG

The charge density is calculated with Eq.(7.1) in SLG devices. For  $n, n_*$  is calculated by plotting  $\ln(\sigma_{xx})(n)$  on a logarithmic scale and fitting a line on the linearly increasing part of the conductivity and an another line at the constant part of  $\sigma_{xx}$  at low densities. The intersection of the two lines defines  $n_*$  as shown in Fig. E.1a.

The lever arms are obtained from quantum oscillations in magnetoconductance as described in Section 3.2.1. For device A, the longitudinal resistance is shown in Fig. E.1b as a function of n at ambient pressure at 1.5 K. For SLG, from Eq.(2.86), the full filling of the LLs are at  $\nu = \pm 2, \pm 6, \pm 10...$  The position of the LLs as a function of n and B is shown with white lines in the figure, which is used to obtain  $\alpha_{BG}$ . The obtained lever arms are shown in Table E.1 for device A. For device B-D, the lever arms are shown in Table E.3 and Table E.4, respectively. As the dielectric here is SiO<sub>2</sub>/hBN, the change of lever arm is expected to be small with pressure[341] as opposed to having only hBN as a dielectric like in Chapter 5 and chapter 6.

p (GPa)	0	1	2
$\alpha_{\rm BG} \left(\frac{10^{14}}{\rm Vm^2}\right)$	6.92(7)	7.31(7)	7.74(8)
$V_{\rm D}$ (V)	-0.538(2)	-0.384(4)	0.474(5)

Table E.1: Extracted lever arms and  $V_{\rm D}$  of device A for the calculation of n at p = 0 GPa, p = 1 GPa and p = 2 GPa with the uncertainty of the last digit in the bracket originating from the measurement resolution, the readout precision and from the fitting.



Figure E.1: Extraction of  $n_*$  and  $\alpha_{BG}$  of device A at T = 1.5 K and at ambient pressure. (a) Extracting  $n_*$  from  $\sigma_{xx}$ . The natural logarithm of conductivity as a function of the natural logarithm of the charge density is shown, where the blue (red) dots correspond to the electron (hole) doped region. The black dashed lines are linear fits on the data, which are used to obtain  $n_*$  from their intersection. (b) Magnetic oscillation measurements of  $\rho_{xx}$  of device A as a function of n and B. The white dashed lines show the Landau levels at different fillings ( $\nu = \pm 2, \pm 6, \pm 10...$ ).

### E.2 Zero field transport properties

The main findings of the pressure dependence of low-temperature electronic transport of SLG graphene are presented in Section 7.1, which are made on device A. Here, measurements on devices B, C and D are discussed. The conductivity is fitted using Eq.(7.2) for every device, which are shown in Fig. E.2a, Fig. E.3a and Fig. E.4a with blue dashed lines for device B, C and D, respectively. The fitted values are summarized in Table E.2, Table E.3 and Table E.4. The fitted parameters show universal pressure dependence: on every device by applying hydrostatic pressure, the mobility decreases and  $\rho_0^e$  increases.

In Fig. E.2b, Fig. E.3b and Fig. E.4b,  $l_{\rm m}$  is shown for the devices as a function of n at T = 1.5 K. In every device,  $l_{\rm m}$  is shorter than the sample width and length, so every sample is considered diffusive. The decrease of  $l_{\rm m}$  with pressure is universal and consistent with the decrease of mobility.

Similarly to device A, the scattering time is divided into short- and long-ranged scattering times, which are shown in Fig. E.2c-d, Fig. E.3c-d and Fig. E.4c-d as a function of n at T = 1.5 K for device B, C, and D, respectively.  $\tau_{\rm m}^{\rm short}$  decreases with pressure and starts to dominate  $\tau_{\rm m}$  at larger densities, similarly to device A.  $\tau_{\rm m}^{\rm long}$  is



Figure E.2: Zero field transport properties of device B at T = 1.5 K. (a) Conductivity of device B at different pressures as a function of n. The black dashed lines are fits of Eq.(7.2). (b) The elastic mean free path  $(l_m)$  calculated from  $\sigma$  with Eq.(7.3). For comparison, the width of the sample and the length between the two inner contacts in the four-terminal measurement set-up are also shown with dashed lines. (c) Transport scattering time from short-ranged scattering as a function of n at different pressures calculated from  $\rho_0$ . (d) Transport scattering time from long-ranged scattering as a function of n at different pressures calculated with Eq.(7.5).

also decreasing with pressure, and at low densities, it dominates  $\tau_{\rm m}$ , as expected.
p (GPa)	0	2
$n_* (10^{10} \mathrm{cm}^{-2})$	3.1(7)	2.1(1.5)
$\mu^h \left( \frac{m^2}{Vs} \right)$	29.27(5)	21.47(5)
$ ho_0^h(\Omega)$	9.64(3)	16.4(1)
$\mu^e \left(\frac{m^2}{Vs}\right)$	34.51(5)	9.62(7)
$ ho_0^e (\overline{\Omega})$	14.0(1)	43.1(5)
$\alpha_{\rm BG} \left( \frac{10^{14}}{{ m Vm}^2} \right)$	6.81(7)	7.25(7)
$V_{\rm D}$ (V)	-0.800(2)	-0.298(1)

Table E.2: Extracted parameters of device B for the fit of Eq.(7.2) on the conductivity,  $n_*$ ,  $V_{\rm D}$  and  $\alpha_{\rm BG}$  at p = 0 GPa and p = 2 GPa with the uncertainty of the last digit in the bracket originating from the fitting.

p (GPa)	0	2
$n_* (10^{10} \mathrm{cm}^{-2})$	5(2)	5(1)
$\mu^h \left( \frac{m^2}{Vs} \right)$	23.62(3)	20.47(3)
$ ho_0^h(\overline{\Omega})$	8.16(2)	11.25(3)
$\mu^e \left(\frac{m^2}{Vs}\right)$	32.08(8)	18.47(2)
$ ho_0^e ( \widetilde{\Omega} )$	10.01(1)	15.30(3)
$\alpha_{\rm BG} \left( \frac{10^{14}}{{ m Vm}^2} \right)$	7.38(7)	7.04(7)
$V_{\rm D}$ (V)	-4.08(2)	4.004(2)

Table E.3: Extracted parameters of device C for the fit of Eq.(7.2) on the conductivity,  $n_*$ ,  $V_{\rm D}$  and  $\alpha_{\rm BG}$  at p = 0 GPa and p = 2 GPa with the uncertainty of the last digit in the bracket originating from the fitting.

p (GPa)	0	1.5
$n_* (10^{10} \mathrm{cm}^{-2})$	3.1(5)	4(1)
$\mu^h \left( \frac{m^2}{Vs} \right)$	14.33(7)	15.16(5)
$ ho_0^h(\overline{\Omega})$	< 0.1	4.6(1)
$\mu^e \left(\frac{m^2}{Vs}\right)$	21.5(1)	9.80(3)
$ ho_0^e ( \widetilde{\Omega} )$	2.45(7)	6.1(2)
$\alpha_{\rm BG} \left( \frac{10^{14}}{{ m Vm}^2} \right)$	7.00(7)	7.40(7)
$V_{\rm D}$ (V)	0.877(2)	2.155(2)

Table E.4: Extracted parameters of device D for the fit of Eq.(7.2) on the conductivity,  $n_*$ ,  $V_{\rm D}$  and  $\alpha_{\rm BG}$  at p = 0 GPa and p = 1.5 GPa with the uncertainty of the last digit in the bracket originating from the fitting.

### E.3 Raw data of WL measurements of device A

 $\delta\sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity is shown in Fig. E.5a as a function of B at different charge densities at T = 1.5 K. A WL signal is clearly visible in the raw data. The high temperature background at T = 20 K, which is subtracted from  $\delta\sigma$  at T = 1.5 K, is shown in Fig. E.5b. The background is nearly constant.



Figure E.3: Zero field transport properties of device C at T = 1.5 K. (a) Conductivity of device B at different pressures as a function of n. The black dashed lines are fits of Eq.(7.2). (b) The elastic mean free path  $(l_m)$  calculated from  $\sigma$  with Eq.(7.3). For comparison, the width of the sample and the length between the two inner contacts in the four-terminal measurement set-up are also shown with dashed lines. (c) Transport scattering time from short-ranged scattering as a function of n at different pressures calculated from  $\rho_0$ . (d) Transport scattering time from long-ranged scattering as a function of n at different pressures calculated with Eq.(7.5).



Figure E.4: Zero field transport properties of device D at T = 1.5 K. (a) Conductivity of device B at different pressures as a function of n. The black dashed lines are fits of Eq.(7.2). (b) The elastic mean free path  $(l_m)$  calculated from  $\sigma$  with Eq.(7.3). For comparison, the width of the sample and the length between the two inner contacts in the four-terminal measurement set-up are also shown with dashed lines. (c) Transport scattering time from short-ranged scattering as a function of n at different pressures calculated from  $\rho_0$ . (d) Transport scattering time from long-ranged scattering as a function of n at different pressures calculated with Eq.(7.5).



Figure E.5: Raw data of WL measurements on device A at p = 0 GPa. (a)  $\delta \sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity as a function of B at different charge densities at T = 1.5 K. (b)  $\delta \sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity as a function of B at different charge densities at T = 20 K. This is used as a background, which is subtracted from  $\delta \sigma$  at T = 1.5 K in Fig. 7.7.

## E.4 Additional data on the in-plane magnetic field dependence of WL



Figure E.6: WL in-plane magnetic field dependence of device C. (a-b)  $\delta\sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity as a function of B at various in-plane magnetic fields  $(B_{\parallel})$  at T = 1.5 K,  $n = -10^{12}$  cm<sup>-2</sup>, (a) at p = 0 GPa and (b) at p = 2 GPa. The black dashed lines correspond to global fits on the data, which results  $\tau_* = 1$  fs and  $\tau_{iv}(p = 0) = 7.5 \pm 1$  ps with  $D(p = 0) = 1.11 \text{ m}^2/\text{s}$  for p = 0 GPa in panel (a) and  $\tau_* = 1$  fs and  $\tau_{iv}(p = 2$  GPa) =  $1.5 \pm 0.4$  ps with D(p = 2 GPa) =  $0.93 \text{ m}^2/\text{s}$  for p = 2 GPa in panel (b). (c) Extracted  $\tau_{\phi}^{-1}$  as a function of  $B_{\parallel}^2$  at 0 and 2 GPa pressures. The dephasing rate depends linearly on  $B_{\parallel}^2$  according to Eq.(2.72). The solid lines are line fits which gives  $Z^2R = 49 \pm 13 \text{ nm}^3$  for p = 0 GPa and  $Z^2R = 69 \pm 22 \text{ nm}^3$  for p = 2 GPa.

In addition to magnetotransport measurement on device C at  $n = 1.25 \cdot 10^{12} \,\mathrm{cm}^{-2}$ , which is discussed in Section 7.2.4, measurements at  $n = -10^{12} \,\mathrm{cm}^{-2}$  is also performed on device C and D at  $T = 1.5 \,\mathrm{K}$ .  $\delta\sigma$  is shown in Fig. E.6a at ambient pressure for device C as a function of B at different in-plane magnetic fields. A global fit is also performed on  $\delta\sigma$  as a function of B, which resulted  $\tau_* = 1$  fs and  $\tau_{iv}(p=0) = 7.5 \pm 1$  ps, which is, within the range of the fitting error, the same as at  $n = -10^{12}$  cm<sup>-2</sup>. From the fits, the dephasing rate is plotted in Fig. E.6c as a function of  $B_{\parallel}^2$  with black error bars. A linear fit on  $\tau_{\phi}^{-1}$  is shown with a black line in the figure, which slope gives  $Z^2R = 49 \pm 13$  nm<sup>3</sup>. The deviation from the corrugation volume at  $n = 1.25 \cdot 10^{12}$  cm<sup>-2</sup> is unexpected, as ripple size is expected to be independent of n. Further measurement of the n dependence of  $Z^2R$  is needed to explain this discrepancy, however, it is beyond the scope of this thesis.

After applying 2 GPa hydrostatic pressure on the sample the same analysis is performed, which is shown in Fig. E.6d. From the fit,  $\tau_{iv}(p = 2 \text{ GPa}) = 1.5 \pm 0.4 \text{ ps}$  decreases with the applied pressure similarly as at  $n = 1.25 \cdot 10^{12} \text{ cm}^{-2}$ . From the inplane magnetic field dependence of  $\tau_{\phi}^{-1}$ , which is shown with red markers in Fig. E.6c,  $Z^2R = 69 \pm 22 \text{ nm}^3$ , which increases with pressure similarly as at  $n = 1.25 \cdot 10^{12} \text{ cm}^{-2}$ .

Similar measurements are performed at  $n = -10^{12} \text{ cm}^{-2}$  on device D, which are shown in Fig. E.7a at p = 0 GPa and in Fig. E.7b at p = 1.5 GPa, respectively. From the fittings,  $\tau_{\phi}^{-1}$  is shown in Fig. E.7c as a function of  $B_{\parallel}^2$  at 0 and 2 GPa with black and red markers, respectively. The slope of linear fits on the dephasing rates, which is shown with solid lines in the figure, results  $Z^2R = 32 \pm 3 \text{ nm}^3$  for p = 0 GPa and  $Z^2R = 78 \pm 23 \text{ nm}^3$  for p = 1.5 GPa, which is similar to the volumes obtained on device C. From the measurements of the suppression of WL with in-plane magnetic fields, I conclude that the increase of the corrugation volume is universal by increasing the applied hydrostatic pressure in hBN-encapsulated graphene devices.

#### E.5 SdH oscillations measurements on device D

Similar magnetotransport measurements are also carried out on device D.  $\rho_{xx}$  is plotted as a function of  $B^{-1}$  at  $n = 2.4 \cdot 10^{12} \,\mathrm{cm}^{-2}$  at ambient pressure at different temperatures in Fig. E.8a. A global fit is also performed on the data similarly as it is discussed in Section 7.2.1 for device A. The result of the fit is also shown in the figure with dashed curves along with the fitting parameters.  $m^*$  is similar to its expected value from the theory.  $\tau_q$  is extracted at different charge densities, which is shown in Fig. E.8b with black markers.  $\tau_q$  is increasing with n, which is expected from Eq.(2.56) if the dominant scattering mechanism is long-ranged scattering. The ratio of  $\tau_m/\tau_q$  is also calculated as a function of n, which is shown in Fig E.8c with black markers. An increase of  $\tau_m/\tau_q$  with n is observed, which is expected if the charged impurities are away from the graphene plane[140].



Figure E.7: WL in-plane magnetic field dependence of device D. (a-b)  $\delta\sigma = \sigma(B) - \sigma(B = 0)$  magnetoconductivity as a function of B at various in-plane magnetic fields  $(B_{\parallel})$  at T = 1.5 K,  $n = -10^{12}$  cm<sup>-2</sup>, (a) at p = 0 GPa and (b) at p = 1.5 GPa. The black dashed lines correspond to global fits on the data, which results  $\tau_* = 1$  fs and  $\tau_{iv}(p = 0) = 8 \pm 1.5$  ps with  $D(p = 0) = 0.58 \text{ m}^2/\text{s}$  for p = 0 GPa in panel (a) and  $\tau_* = 1$  fs and  $\tau_{iv}(p = 2 \text{ GPa}) = 8 \pm 2$  ps with  $D(p = 2 \text{ GPa}) = 0.66 \text{ m}^2/\text{s}$  for p = 1.5 GPa in panel (b). (c) Extracted  $\tau_{\phi}^{-1}$  as a function of  $B_{\parallel}^2$  at 0 and 2 GPa pressures. The dephasing rate depends linearly on  $B_{\parallel}^2$  according to Eq.(2.72). The solid lines are line fits which gives  $Z^2R = 32\pm 3 \text{ nm}^3$  for p = 0 GPa and  $Z^2R = 78\pm 23 \text{ nm}^3$  for p = 1.5 GPa.

From similar SdH oscillations measurements under p = 1.5 GPa hydrostatic pressure,  $\tau_{\rm q}$  is extracted, which is shown in Fig. E.8b with blue markers as a function of n. A similar decrease of  $\tau_{\rm q}$  is observed as in device A. The ratio of  $\tau_{\rm m}/\tau_{\rm q}$  at p = 1.5 GPa is shown with blue markers in Fig. E.8c. The ratio is increasing with pressure in this device. An increasing trend with n can be observed similarly as at p = 0 GPa. However, the lack of measurement data prevents us to make quantitative statements.



Figure E.8: SdH oscillations measurements on device D. (a)  $\rho_{xx}$  after subtracting a quadratic background at  $n = 2.4 \cdot 10^{12} \text{ cm}^{-2}$  at p = 0 GPa at different temperatures. (b) Extracted quantum scattering time as a function of n at p = 0 GPa and at p = 1.5 GPa pressures. (c)  $\tau_{\text{m}}/\tau_{\text{q}}$  ratio as a function of n at p = 0 GPa and at p = 1.5 GPa pressures.

#### E.6 Magnetic focusing measurements on device D

Magnetic focusing experiments are also performed on device D under zero and 2 GPa hydrostatic pressures. The measurement configuration, which is used in the measurements, is illustrated in Fig. E.9. Contact d is the injector and contact p is grounded. Contact e is the collector and contact i, which is a lot farther (16.3 µm) from the collector than  $l_{\rm m}$ , is used as a reference in the non-local measurements.



Figure E.9: Schematic measurement arrangement for TMF on device D. The current flows between contacts d and p. The current is injected at contact d and contact p is grounded. Contact e is used as a detector contact. The voltage is measured between contacts e and i, where contact i serves as a reference. The black scale bar is 5 µm.

 $R_{\rm nl}$  as a function of *n* and *B* is shown in Fig. E.10a at T = 1.5 K at p = 0 GPa. Unlike sample A, here no SdH oscillations are observed in the range of  $|B| \le 0.4$  T. For  $|B| \le 0.2$  T, TMF peaks are observed, which follow the black curves, that correspond





Figure E.10: Magnetic focusing on device D. (a)  $R_{\rm nl}$  as a function of n and B. The solid lines are the TMF peaks from Eq.(2.92) with  $L = 3.5 \,\mu{\rm m}$ . (b)  $R_{\rm nl}$  after subtracting a linear background at  $n = 1.7 \cdot 10^{12} \,{\rm cm}^{-2}$  at p = 0 GPa at different temperatures. (c-d) Extracted effective scattering time as a function of temperature near p = 0 GPa and p = 1.5 GPa. The solid lines are linear fits. On the panel (c) the slope of the fitted lines are  $-1.6 \pm 0.3$  and  $-1.6 \pm 0.1$  for p = 0, 2 GPa, respectively. On the panel (d) the slope of the fitted lines are  $-1.7 \pm 0.1$  and  $-1.8 \pm 0.2$  for p = 0, 2 GPa, respectively.

Along  $n = 1.7 \cdot 10^{12} \,\mathrm{cm}^{-2}$ , the temperature dependence of  $R_{\rm nl}$  is shown in Fig. E.9b as a function of B at p = 0 GPa. A peak around  $B \sim 0.1 \,\mathrm{T}$  is clearly visible, which corresponds to the first peak of TMF. The amplitude of the peak decays by increasing the temperature and completely vanishes at large temperatures. Similarly to device A, the area under the peak is numerically integrated after removing a linear background, which is fitted in the regions (-B) where no TMF peaks are expected.  $\tau_{\rm eff}$  is calculated with Eq.(2.94), where  $A_1(T_{\rm base}) = A_1(T = 1.5 \,\mathrm{K})$  is used. The results are shown in

Fig. E.9d with black markers in a logarithmic scale as a function of T.  $\tau_{\text{eff}}$  decreases with T as expected. The exponent  $\alpha = -1.7 \pm 0.1$  of  $\tau_{\text{eff}} \propto T^{\alpha}$  is extracted by a linear fit in the log-log plot, which is shown with a black line in the figure. Similarly from the TMF experiment on device A,  $\alpha$  is between -1 and -2 meaning that both electron-electron and electron-phonon scatterings contribute to the reduction of the TMF signal.

Similar experiments are performed at  $n = -1.96 \cdot 10^{12} \text{ cm}^{-2}$ . The extracted  $\tau_{\text{eff}}$  is shown in Fig. E.9c with black markers. From a similar line fit,  $\alpha = -1.6 \pm 0.3$  gives the same results as at  $n = 1.7 \cdot 10^{12} \text{ cm}^{-2}$ .

Under p = 2 GPa hydrostatic pressure, similar TMF experiments are also performed, which results are shown with red markers in Fig. E.9d at  $n = 1.3 \cdot 10^{12} \text{ cm}^{-2}$ and Fig. E.9c at  $n = -1.8 \cdot 10^{12} \text{ cm}^{-2}$ . From similar line fits, the extracted exponents  $(\alpha(n = -1.8 \cdot 10^{12} \text{ cm}^{-2}) = -1.6 \pm 0.1$  and  $\alpha(n = 1.3 \cdot 10^{12} \text{ cm}^{-2}) = -1.8 \pm 0.2)$  are the same as the ones obtained at ambient pressure within the uncertainty of the fittings, similarly to the results on device A. These measurements further enforce that, the pressure doesn't qualitatively change the transport properties of hBN-encapsulated SLG.

## E.7 Temperature-dependent measurements on device D



Figure E.11: Fitting the temperature dependence of device D with Eq.(7.9). (a) The resistivity at some densities as a function of the temperature at ambient pressure is plotted with dots. (b)  $\Delta \rho_{xx}$  as a function of temperature at  $n = -0.8 \cdot 10^{12} \,\mathrm{cm}^{-2}$  and at  $p = 0 \,\mathrm{GPa}$  and  $p = 1.5 \,\mathrm{GPa}$  plotted with black and red dots, respectively. The dashed lines are global fits at different pressures. The extracted fitting coefficients of  $\gamma$  and B are shown in Table E.5.

The main findings of the role of electron-phonon coupling and its pressure dependence in device A are presented in Section 7.3. Here, measurements of device D are presented. At fixed temperatures, the longitudinal resistance is measured as a function of charge density, and a similar fitting procedure with Eq.(7.9) on the resistivity as a function of n and T is performed. Here  $\Delta \rho_{xx}$  is fitted in the charge density range of  $[-16, -8] \cdot 10^{11} \text{ cm}^{-2}$  and in the temperature range of 20 - 285 K.

 $\rho_{xx}$  of device D at some densities as a function of T is shown in Fig. E.11a. Similarly to device A, an increasing tendency is observed. Similar measurements are performed at 1.5 GPa, which is shown with red markers in Fig. E.11b. At large temperatures, similarly to the observation in device A, the resistance increases with pressure due to the increased effect of the RIP, whereas at intermediate temperatures, where the acoustic electron-phonon scattering dominates, the change of resistance is negligible with pressure.



Figure E.12: Logarithm of the longitudinal resistivity after subtracting the residual resistivity and the resistivity contribution of the acoustic phonons as a function the logarithm of the charge density at T = 230 K of device D is shown with black and red lines at 0 and 2 GPa, respectively. The blue dashed lines are linear fits on the linear regime of the curves. (a)  $\ln(\rho_{\rm RIP})$  at n < 0. From the slope of the fitted lines  $\alpha(p=0) = 1 \pm 0.1$  and  $\alpha(p=2$  GPa) =  $1 \pm 0.1$ . (b)  $\ln(\rho_{\rm RIP})$  at n > 0. From the slope of the fitted lines  $\alpha(p=0) = 1 \pm 0.2$  and  $\alpha(p=2$  GPa) =  $0.8 \pm 0.2$ .

 $\ln(\rho_{\rm RIP})$  is shown in Fig. E.12. In the hole regime in panel (a), at large densities, a linear regime is observed, where the blue dashed line is fitted both at zero and 1.5 GPa. From the fit  $\alpha = 1$ , similarly from the fits in device A. In the hole regime in panel (b), a non-linear behavior is observed, i.e. Eq.(7.9) gives an unreliable result if fitted in this regime.

The fitted curves are shown in Fig. E.11a and Fig. E.11b with dashed lines. At the hole regime, the fits follow the data badly due to the non-linear behavior at large

p (GPa)	0	1.5
$\gamma (\omega/K)$	$0.095 \pm 0.004$	$0.087 \pm 0.004$
B $(10^{19}\Omega m^{2\alpha})$	$1.92\pm0.08$	$2.17\pm0.08$

Table E.5: Results of the global fit of Eq.(7.9) on  $\Delta \rho_{xx}$  of device D at zero and 1.5 GPa.

densities, thus only the fitting results at the hole regime is discussed here. The results of the fitting is summarized in Table E.5. The fitted parameters show a similar tendency as the obtained values in device A:  $\gamma$  has a decreasing tendency. However, it is independent of p within the margin of its uncertainty in the hole regime. The parameter B increases with p, as expected.

# Bibliography

- Valasa, S., Kotha, V. R. & Vadthiya, N. Beyond moore's law a critical review of advancements in negative capacitance field effect transistors: A revolution in next-generation electronics. *Materials Science in Semiconductor Processing* 173, 108116 (2024).
- [2] Yoo, J., Nam, C.-Y. & Bussmann, E. Atomic precision processing of twodimensional materials for next-generation microelectronics. ACS Nano 18, 21614–21622 (2024).
- [3] Novoselov, K. S. et al. Two-dimensional atomic crystals. Proceedings of the National Academy of Sciences 102, 10451–10453 (2005).
- [4] Novoselov, K. S. *et al.* Electric field effect in atomically thin carbon films. *Science* 306, 666–669 (2004).
- [5] Geim, A. K. & Grigorieva, I. V. Van der waals heterostructures. Nature 499, 419–425 (2013).
- [6] Qian, X., Liu, J., Fu, L. & Li, J. Quantum spin hall effect in two-dimensional transition metal dichalcogenides. *Science* 346, 1344–1347 (2014).
- [7] Akinwande, D. et al. Graphene and two-dimensional materials for silicon technology. Nature 573, 507–518 (2019).
- [8] Huang, X., Liu, C. & Zhou, P. 2d semiconductors for specific electronic applications: from device to system. npj 2D Materials and Applications 6 (2022).
- [9] Liu, A. et al. The roadmap of 2d materials and devices toward chips. Nano-Micro Letters 16 (2024).
- [10] Ajayan, P., Kim, P. & Banerjee, K. Two-dimensional van der waals materials. *Physics Today* 69, 38–44 (2016).
- [11] Ferrari, A. C. et al. Science and technology roadmap for graphene, related twodimensional crystals, and hybrid systems. Nanoscale 7, 4598–4810 (2015).

- [12] Bian, Z., Miao, J., Zhao, Y. & Chai, Y. Strong interlayer interaction for engineering two-dimensional materials. Accounts of Materials Research 3, 1220–1231 (2022).
- [13] Dean, C. R. et al. Boron nitride substrates for high-quality graphene electronics. Nature Nanotechnology 5, 722–726 (2010).
- [14] Yankowitz, M., Ma, Q., Jarillo-Herrero, P. & LeRoy, B. J. van der waals heterostructures combining graphene and hexagonal boron nitride. *Nature Reviews Physics* 1, 112–125 (2019).
- [15] Hennighausen, Z. & Kar, S. Twistronics: a turning point in 2d quantum materials. Electronic Structure 3, 014004 (2021).
- [16] Ponomarenko, L. A. et al. Cloning of dirac fermions in graphene superlattices. Nature 497, 594–597 (2013).
- [17] Dean, C. R. et al. Hofstadter's butterfly and the fractal quantum hall effect in moiré superlattices. Nature 497, 598–602 (2013).
- [18] Cao, Y. et al. Correlated insulator behaviour at half-filling in magic-angle graphene superlattices. Nature 556, 80–84 (2018).
- [19] Cao, Y. et al. Unconventional superconductivity in magic-angle graphene superlattices. Nature 556, 43–50 (2018).
- [20] Shah, S. J. et al. Progress and prospects of moiré superlattices in twisted tmd heterostructures. Nano Research 17, 10134–10161 (2024).
- [21] Fabian, J., Matos-Abiague, A., Ertler, C., Stano, P. & Zutic, I. Semiconductor spintronics. Acta Physica Slovaca 57, 565–907 (2007).
- [22] Ralph, D. & Stiles, M. Spin transfer torques. Journal of Magnetism and Magnetic Materials 320, 1190–1216 (2008).
- [23] Seneor, P. et al. Spintronics with graphene. MRS Bulletin 37, 1245–1254 (2012).
- [24] Han, W., Kawakami, R. K., Gmitra, M. & Fabian, J. Graphene spintronics. *Nature Nanotechnology* 9, 794–807 (2014).
- [25] Gmitra, M. & Fabian, J. Graphene on transition-metal dichalcogenides: A platform for proximity spin-orbit physics and optospintronics. *Physical Review B* 92, 155403 (2015).

- [26] Yan, W. et al. A two-dimensional spin field-effect switch. Nature Communications 7, 13372 (2016).
- [27] Yang, B. et al. Tunable spin-orbit coupling and symmetry-protected edge states in graphene/WS2. 2D Materials 3, 031012 (2016).
- [28] Dankert, A. & Dash, S. P. Electrical gate control of spin current in van der waals heterostructures at room temperature. *Nature Communications* 8, 16093 (2017).
- [29] Omar, S. & van Wees, B. J. Spin transport in high-mobility graphene on WS2 substrate with electric-field tunable proximity spin-orbit interaction. *Physical Review B* 97, 045414 (2018).
- [30] Benitez, L. A. *et al.* Tunable room-temperature spin galvanic and spin hall effects in van der waals heterostructures. *Nature Materials* 19, 170–175 (2020).
- [31] Herling, F. et al. Gate tunability of highly efficient spin-to-charge conversion by spin hall effect in graphene proximitized with wse2. APL Materials 8, 071103 (2020).
- [32] Safeer, C. K. et al. Room-temperature spin hall effect in graphene/MoS2 van der waals heterostructures. Nano Letters 19, 1074–1082 (2019).
- [33] Yang, H. et al. Twist-angle-tunable spin texture in wse2/graphene van der waals heterostructures. Nature Materials (2024).
- [34] Pimenta Martins, L. G. et al. High-pressure studies of atomically thin van der waals materials. Applied Physics Reviews 10 (2023).
- [35] Shirotani, I. et al. Phase transitions and superconductivity of black phosphorus and phosphorus-arsenic alloys at low temperatures and high pressures. *Physical Review B* 50, 16274–16278 (1994).
- [36] Chi, Z. et al. Superconductivity in pristine 2ha-mos2 at ultrahigh pressure. Physical Review Letters 120, 037002 (2018).
- [37] Yankowitz, M. et al. Tuning superconductivity in twisted bilayer graphene. Science 363, 1059–1064 (2019).
- [38] Pei, S., Wang, Z. & Xia, J. High pressure studies of 2d materials and heterostructures: A review. *Materials & Design* 213, 110363 (2022).

- [39] Neto, A. H. C., Guinea, F., Peres, N. M. R., Novoselov, K. S. & Geim, A. K. The electronic properties of graphene. *Reviews of Modern Physics* 81, 109–162 (2009).
- [40] Baskin, Y. & Meyer, L. Lattice constants of graphite at low temperatures. *Phys-ical Review* 100, 544–544 (1955).
- [41] Wallace, P. R. The band theory of graphite. *Physical Review* **71**, 622–634 (1947).
- [42] Slonczewski, J. C. & Weiss, P. R. Band structure of graphite. *Physical Review* 109, 272–279 (1958).
- [43] Ando, T. Theory of electronic states and transport in carbon nanotubes. Journal of the Physical Society of Japan 74, 777–817 (2005).
- [44] Ando, T. Screening effect and impurity scattering in monolayer graphene. Journal of the Physical Society of Japan 75, 074716 (2006).
- [45] McCann, E. & Koshino, M. The electronic properties of bilayer graphene. *Reports on Progress in Physics* 76, 056503 (2013).
- [46] Novoselov, K. S. et al. Two-dimensional gas of massless dirac fermions in graphene. Nature 438, 197–200 (2005).
- [47] Zhang, Y., Tan, Y.-W., Stormer, H. L. & Kim, P. Experimental observation of the quantum hall effect and berry's phase in graphene. *Nature* 438, 201–204 (2005).
- [48] Tan, Z. et al. Shubnikov-de haas oscillations of a single layer graphene under dc current bias. Physical Review B 84, 115429 (2011).
- [49] Jung, J. & MacDonald, A. H. Accurate tight-binding models for theπbands of bilayer graphene. *Physical Review B* 89, 035405 (2014).
- [50] Island, J. O. *et al.* Spin–orbit-driven band inversion in bilayer graphene by the van der waals proximity effect. *Nature* 571, 85–89 (2019).
- [51] Hunt, B. M. et al. Direct measurement of discrete valley and orbital quantum numbers in bilayer graphene. Nature Communications 8, 948 (2017).
- [52] Morell, E. S., Correa, J. D., Vargas, P., Pacheco, M. & Barticevic, Z. Flat bands in slightly twisted bilayer graphene: Tight-binding calculations. *Physical Review* B 82 (2010).

- [53] Andrei, E. Y. & MacDonald, A. H. Graphene bilayers with a twist. Nature Materials 19, 1265–1275 (2020).
- [54] Bistritzer, R. & MacDonald, A. H. Transport between twisted graphene layers. *Physical Review B* 81 (2010).
- [55] Song, J. C. W., Shytov, A. V. & Levitov, L. S. Electron interactions and gap opening in graphene superlattices. *Physical Review Letters* **111**, 266801 (2013).
- [56] Diez, M., Dahlhaus, J., Wimmer, M. & Beenakker, C. Emergence of massless dirac fermions in graphene's hofstadter butterfly at switches of the quantum hall phase connectivity. *Physical Review Letters* **112**, 196602 (2014).
- [57] Slotman, G. et al. Effect of structural relaxation on the electronic structure of graphene on hexagonal boron nitride. *Physical Review Letters* **115**, 186801 (2015).
- [58] Jung, J., DaSilva, A. M., MacDonald, A. H. & Adam, S. Origin of band gaps in graphene on hexagonal boron nitride. *Nature Communications* 6 (2015).
- [59] Hunt, B. et al. Massive dirac fermions and hofstadter butterfly in a van der waals heterostructure. Science 340, 1427–1430 (2013).
- [60] Wang, L. et al. Evidence for a fractional fractal quantum hall effect in graphene superlattices. Science 350, 1231–1234 (2015).
- [61] Du, Y., Xu, N., Lin, X. & Jauho, A.-P. Moiré effects in graphene-hBN heterostructures. *Physical Review Research* 2, 043427 (2020).
- [62] Lu, X. et al. Superconductors, orbital magnets and correlated states in magicangle bilayer graphene. Nature 574, 653–657 (2019).
- [63] Kerelsky, A. et al. Maximized electron interactions at the magic angle in twisted bilayer graphene. Nature 572, 95–100 (2019).
- [64] Jiang, Y. et al. Charge order and broken rotational symmetry in magic-angle twisted bilayer graphene. Nature 573, 91–95 (2019).
- [65] Xie, Y. et al. Spectroscopic signatures of many-body correlations in magic-angle twisted bilayer graphene. Nature 572, 101–105 (2019).
- [66] Choi, Y. et al. Electronic correlations in twisted bilayer graphene near the magic angle. Nature Physics 15, 1174–1180 (2019).

- [67] N'Diaye, A. T., Coraux, J., Plasa, T. N., Busse, C. & Michely, T. Structure of epitaxial graphene on ir(111). New Journal of Physics 10, 043033 (2008).
- [68] Artaud, A. *et al.* Universal classification of twisted, strained and sheared graphene moiré superlattices. *Scientific Reports* **6** (2016).
- [69] Bistritzer, R. & MacDonald, A. H. Moire bands in twisted double-layer graphene. Proceedings of the National Academy of Sciences 108, 12233–12237 (2011).
- [70] Jung, J., Raoux, A., Qiao, Z. & MacDonald, A. H. Ab initiotheory of moiré superlattice bands in layered two-dimensional materials. *Physical Review B* 89, 205414 (2014).
- [71] Bernevig, B. A., Song, Z.-D., Regnault, N. & Lian, B. Twisted bilayer graphene.
  i. matrix elements, approximations, perturbation theory, and a k·p two-band model. *Phys. Rev. B* 103, 205411 (2021).
- [72] Chittari, B. L., Leconte, N., Javvaji, S. & Jung, J. Pressure induced compression of flatbands in twisted bilayer graphene. *Electronic Structure* 1, 015001 (2018).
- [73] Chebrolu, N. R., Chittari, B. L. & Jung, J. Flat bands in twisted double bilayer graphene. *Physical Review B* 99, 235417 (2019).
- [74] Bransden, B. H. & Joachain, C. J. Physics of atoms and molecules (Prentice Hall, Harlow, 2008), 2. ed., [nachdr.] edn. Includes bibliographical references and index.
- [75] Katsnelson, M. I. The Physics of Graphene (Cambridge University Press, Cambridge, United Kingdom, 2020), second edition edn. Includes bibliographical references and index.
- [76] Kane, C. L. & Mele, E. J. Quantum spin hall effect in graphene. *Physical Review Letters* 95 (2005).
- [77] Gmitra, M., Konschuh, S., Ertler, C., Ambrosch-Draxl, C. & Fabian, J. Bandstructure topologies of graphene: Spin-orbit coupling effects from first principles. *Physical Review B* 80, 235431 (2009).
- [78] Min, H. et al. Intrinsic and rashba spin-orbit interactions in graphene sheets. Physical Review B 74, 165310 (2006).
- [79] Roche, S. et al. Graphene spintronics: the european flagship perspective. 2D Materials 2, 030202 (2015).

- [80] Drögeler, M. et al. Spin lifetimes exceeding 12 ns in graphene nonlocal spin valve devices. Nano Letters 16, 3533–3539 (2016).
- [81] Bisswanger, T. et al. Cvd bilayer graphene spin valves with 26 um spin diffusion length at room temperature. Nano Letters 22, 4949–4955 (2022).
- [82] Eich, M. et al. Spin and valley states in gate-defined bilayer graphene quantum dots. Physical Review X 8, 031023 (2018).
- [83] Banszerus, L. et al. Gate-defined electron-hole double dots in bilayer graphene. Nano Letters 18, 4785–4790 (2018).
- [84] Eich, M. et al. Coupled quantum dots in bilayer graphene. Nano Letters 18, 5042–5048 (2018).
- [85] Banszerus, L. et al. Single-electron double quantum dots in bilayer graphene. Nano Letters 20, 2005–2011 (2020).
- [86] Eich, M. et al. Coulomb dominated cavities in bilayer graphene. Physical Review Research 2, 022038 (2020).
- [87] Gächter, L. M. et al. Single-shot spin readout in graphene quantum dots. PRX Quantum 3, 020343 (2022).
- [88] Banszerus, L. et al. Spin relaxation in a single-electron graphene quantum dot. Nature Communications 13, 3637 (2022).
- [89] Garreis, R. et al. Long-lived valley states in bilayer graphene quantum dots. Nature Physics 20, 428–434 (2024).
- [90] Denisov, A. O. *et al.* Ultra-long relaxation of a kramers qubit formed in a bilayer graphene quantum dot (2024).
- [91] Cheng, Y. C., Zhu, Z. Y., Tahir, M. & Schwingenschlögl, U. Spin-orbit-induced spin splittings in polar transition metal dichalcogenide monolayers. *EPL (Euro-physics Letters)* **102**, 57001 (2013).
- [92] Liu, G.-B., Shan, W.-Y., Yao, Y., Yao, W. & Xiao, D. Three-band tight-binding model for monolayers of group-vib transition metal dichalcogenides. *Physical Review B* 88, 085433 (2013).
- [93] Wang, Z. et al. Strong interface-induced spin-orbit interaction in graphene on WS2. Nature Communications 6, 8339 (2015).

- [94] Wang, Z. et al. Origin and magnitude of 'designer' spin-orbit interaction in graphene on semiconducting transition metal dichalcogenides. *Physical Review* X 6, 041020 (2016).
- [95] Yang, B. et al. Strong electron-hole symmetric rashba spin-orbit coupling in graphene/monolayer transition metal dichalcogenide heterostructures. *Physical Review B* 96, 041409 (2017).
- [96] Wakamura, T. et al. Strong anisotropic spin-orbit interaction induced in graphene by monolayer ws2. Physical Review Letters 120, 106802 (2018).
- [97] Zihlmann, S. et al. Large spin relaxation anisotropy and valley-zeeman spin-orbit coupling in wse2/graphene/h-bn heterostructures. Physical Review B 97, 075434 (2018).
- [98] Amann, J. et al. Counterintuitive gate dependence of weak antilocalization in bilayer graphene/wse2 heterostructures. Physical Review B 105, 115425 (2022).
- [99] Afzal, A. M. et al. Gate modulation of the spin-orbit interaction in bilayer graphene encapsulated by ws2 films. Scientific Reports 8, 3412 (2018).
- [100] Tiwari, P. et al. Experimental observation of spin-split energy dispersion in high-mobility single-layer graphene/wse2 heterostructures. npj 2D Materials and Applications 6, 68 (2022).
- [101] Rao, Q. et al. Ballistic transport spectroscopy of spin-orbit-coupled bands in monolayer graphene on wse2. Nature Communications 14, 6124 (2023).
- [102] Omar, S. & van Wees, B. J. Graphene-ws2 heterostructures for tunable spin injection and spin transport. *Physical Review B* 95, 081404 (2017).
- [103] Avsar, A. et al. Spin-orbit proximity effect in graphene. Nature Communications 5, 4875 (2014).
- [104] Benítez, L. A. et al. Strongly anisotropic spin relaxation in graphene-transition metal dichalcogenide heterostructures at room temperature. Nature Physics 14, 303–308 (2017).
- [105] Ghiasi, T. S., Ingla-Aynés, J., Kaverzin, A. A. & van Wees, B. J. Large proximityinduced spin lifetime anisotropy in transition-metal dichalcogenide/graphene heterostructures. *Nano Letters* 17, 7528–7532 (2017).
- [106] Sun, L. et al. Determining spin-orbit coupling in graphene by quasiparticle interference imaging. Nature Communications 14, 3771 (2023).

- [107] Gmitra, M., Kochan, D., Högl, P. & Fabian, J. Trivial and inverted dirac bands and the emergence of quantum spin hall states in graphene on transition-metal dichalcogenides. *Physical Review B* 93, 155104 (2016).
- [108] Kochan, D., Irmer, S. & Fabian, J. Model spin-orbit coupling hamiltonians for graphene systems. *Physical Review B* 95 (2017).
- [109] David, A., Rakyta, P., Kormányos, A. & Burkard, G. Induced spin-orbit coupling in twisted graphene-transition metal dichalcogenide heterobilayers: Twistronics meets spintronics. *Physical Review B* 100, 085412 (2019).
- [110] Li, Y. & Koshino, M. Twist-angle dependence of the proximity spin-orbit coupling in graphene on transition-metal dichalcogenides. *Physical Review B* 99, 075438 (2019).
- [111] Naimer, T., Zollner, K., Gmitra, M. & Fabian, J. Twist-angle dependent proximity induced spin-orbit coupling in graphene/transition metal dichalcogenide heterostructures. *Physical Review B* 104, 195156 (2021).
- [112] Gmitra, M. & Fabian, J. Proximity effects in bilayer graphene on monolayer WSe2
  : Field-effect spin valley locking, spin-orbit valve, and spin transistor. *Physical Review Letters* 119, 146401 (2017).
- [113] Zollner, K. & Fabian, J. Bilayer graphene encapsulated within monolayers of WS2 or cr2ge2te6 : Tunable proximity spin-orbit or exchange coupling. *Physical Review B* 104, 075126 (2021).
- [114] Lin, J.-X. et al. Spin-orbit-driven ferromagnetism at half moiré filling in magicangle twisted bilayer graphene. Science 375, 437-441 (2022).
- [115] Bhowmik, S. et al. Spin-orbit coupling-enhanced valley ordering of malleable bands in twisted bilayer graphene on wse2. Nature Communications 14, 4055 (2023).
- [116] Chou, Y.-Z., Tan, Y., Wu, F. & Das Sarma, S. Topological flat bands, valley polarization, and interband superconductivity in magic-angle twisted bilayer graphene with proximitized spin-orbit couplings. *Physical Review B* **110**, 1041108 (2024).
- [117] Zhang, Y. et al. Twist-programmable superconductivity in spin-orbit coupled bilayer graphene (2024). 2408.10335.

- [118] Khoo, J. Y., Morpurgo, A. F. & Levitov, L. On-demand spin-orbit interaction from which-layer tunability in bilayer graphene. *Nano Letters* 17, 7003–7008 (2017).
- [119] Konschuh, S., Gmitra, M., Kochan, D. & Fabian, J. Theory of spin-orbit coupling in bilayer graphene. *Physical Review B* 85, 115423 (2012).
- [120] Péterfalvi, C. G., David, A., Rakyta, P., Burkard, G. & Kormányos, A. Quantum interference tuning of spin-orbit coupling in twisted van der waals trilayers. *Physical Review Research* 4, 1022049 (2022).
- [121] Wang, L. et al. One-dimensional electrical contact to a two-dimensional material. Science 342, 614–617 (2013).
- [122] Bolotin, K. et al. Ultrahigh electron mobility in suspended graphene. Solid State Communications 146, 351–355 (2008).
- [123] Du, X., Skachko, I., Barker, A. & Andrei, E. Y. Approaching ballistic transport in suspended graphene. *Nature Nanotechnology* 3, 491–495 (2008).
- [124] Mayorov, A. S. et al. Micrometer-scale ballistic transport in encapsulated graphene at room temperature. Nano Letters 11, 2396–2399 (2011).
- [125] Kretinin, A. V. et al. Electronic properties of graphene encapsulated with different two-dimensional atomic crystals. Nano Letters 14, 3270–3276 (2014).
- [126] Amet, F., Williams, J., Watanabe, K., Taniguchi, T. & Goldhaber-Gordon, D. Selective equilibration of spin-polarized quantum hall edge states in graphene. *Physical Review Letters* **112**, 196601 (2014).
- [127] Banszerus, L. et al. Ultrahigh-mobility graphene devices from chemical vapor deposition on reusable copper. Science Advances 1 (2015).
- [128] Banszerus, L. *et al.* Ballistic transport exceeding 28  $\mu$ m in cvd grown graphene. Nano Letters **16**, 1387–1391 (2016).
- [129] Terrés, B. et al. Size quantization of dirac fermions in graphene constrictions. Nature Communications 7 (2016).
- [130] Pizzocchero, F. *et al.* The hot pick-up technique for batch assembly of van der waals heterostructures. *Nature Communications* **7** (2016).

- [131] Zimmermann, K. et al. Tunable transmission of quantum hall edge channels with full degeneracy lifting in split-gated graphene devices. Nature Communications 8 (2017).
- [132] Yamoah, M. A., Yang, W., Pop, E. & Goldhaber-Gordon, D. High-velocity saturation in graphene encapsulated by hexagonal boron nitride. ACS Nano 11, 9914–9919 (2017).
- [133] De Fazio, D. et al. High-mobility, wet-transferred graphene grown by chemical vapor deposition. ACS Nano 13, 8926–8935 (2019).
- [134] Kim, M. et al. Control of electron-electron interaction in graphene by proximity screening. Nature Communications 11 (2020).
- [135] Purdie, D. G. et al. Cleaning interfaces in layered materials heterostructures. Nature Communications 9 (2018).
- [136] Wang, W. et al. Clean assembly of van der waals heterostructures using silicon nitride membranes. Nature Electronics 6, 981–990 (2023).
- [137] Liu, Z., Jiang, L. & Zheng, Y. Conductivity tensor of graphene dominated by spin-orbit coupling scatterers: A comparison between the results from kubo and boltzmann transport theories. *Scientific Reports* 6 (2016).
- [138] Das Sarma, S., Adam, S., Hwang, E. H. & Rossi, E. Electronic transport in two-dimensional graphene. *Reviews of Modern Physics* 83, 407–470 (2011).
- [139] Das Sarma, S. & Stern, F. Single-particle relaxation time versus scattering time in an impure electron gas. *Physical Review B* 32, 8442–8444 (1985).
- [140] Hwang, E. H. & Das Sarma, S. Single-particle relaxation time versus transport scattering time in a two-dimensional graphene layer. *Physical Review B* 77, 195412 (2008).
- [141] Hong, X., Zou, K. & Zhu, J. Quantum scattering time and its implications on scattering sources in graphene. *Physical Review B* 80, 241415 (2009).
- [142] Tworzydło, J., Trauzettel, B., Titov, M., Rycerz, A. & Beenakker, C. W. J. Subpoissonian shot noise in graphene. *Physical Review Letters* 96, 246802 (2006).
- [143] Bolotin, K. I., Sikes, K. J., Hone, J., Stormer, H. L. & Kim, P. Temperaturedependent transport in suspended graphene. *Physical Review Letters* 101, 096802 (2008).

- [144] Yudhistira, I. & Adam, S. Theory for electron transport in graphene. Synthetic Metals 210, 2–8 (2015).
- [145] Adam, S., Hwang, E. H., Galitski, V. M. & Das Sarma, S. A self-consistent theory for graphene transport. *Proceedings of the National Academy of Sciences* 104, 18392–18397 (2007).
- [146] Pálinkás, A. et al. The composition and structure of the ubiquitous hydrocarbon contamination on van der waals materials. Nature Communications 13 (2022).
- [147] Peres, N. M. R. Colloquium: The transport properties of graphene: An introduction. Reviews of Modern Physics 82, 2673–2700 (2010).
- [148] Shon, N. & Ando, T. Quantum transport in two-dimensional graphite system. Journal of the Physical Society of Japan 67, 2421–2429 (1998).
- [149] Ferreira, A. et al. Unified description of the dc conductivity of monolayer and bilayer graphene at finite densities based on resonant scatterers. *Physical Review* B 83, 165402 (2011).
- [150] Robinson, J. P., Schomerus, H., Oroszlány, L. & Fal'ko, V. I. Adsorbate-limited conductivity of graphene. *Physical Review Letters* **101**, 196803 (2008).
- [151] Castro Neto, A. H. & Guinea, F. Impurity-induced spin-orbit coupling in graphene. *Physical Review Letters* 103, 026804 (2009).
- [152] Stauber, T., Peres, N. M. R. & Castro Neto, A. H. Conductivity of suspended and non-suspended graphene at finite gate voltage. *Physical Review B* 78, 085418 (2008).
- [153] Hentschel, M. & Guinea, F. Orthogonality catastrophe and kondo effect in graphene. *Physical Review B* 76, 115407 (2007).
- [154] Monteverde, M. et al. Transport and elastic scattering times as probes of the nature of impurity scattering in single-layer and bilayer graphene. Physical Review Letters 104, 126801 (2010).
- [155] Kotsakidis, J. C. et al. Charged impurity scattering and electron-electron interactions in large-area hydrogen intercalated bilayer graphene. ACS Applied Materials & Interfaces 16, 61194–61203 (2024).
- [156] Fasolino, A., Los, J. H. & Katsnelson, M. I. Intrinsic ripples in graphene. Nature Materials 6, 858–861 (2007).

- [157] Deng, S. & Berry, V. Wrinkled, rippled and crumpled graphene: an overview of formation mechanism, electronic properties, and applications. *Materials Today* 19, 197–212 (2016).
- [158] Yildiz, G., Bolton-Warberg, M. & Awaja, F. Graphene and graphene oxide for bio-sensing: General properties and the effects of graphene ripples. Acta Biomaterialia 131, 62–79 (2021).
- [159] Gui, G., Zhong, J. & Ma, Z. Electronic properties of rippled graphene. Journal of Physics: Conference Series 402, 012004 (2012).
- [160] Vozmediano, M., Katsnelson, M. & Guinea, F. Gauge fields in graphene. *Physics Reports* 496, 109–148 (2010).
- [161] Ishigami, M., Chen, J. H., Cullen, W. G., Fuhrer, M. S. & Williams, E. D. Atomic structure of graphene on sio2. *Nano Letters* 7, 1643–1648 (2007).
- [162] Katsnelson, M. & Geim, A. Electron scattering on microscopic corrugations in graphene. Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences 366, 195–204 (2007).
- [163] Couto, N. J. et al. Random strain fluctuations as dominant disorder source for high-quality on-substrate graphene devices. *Physical Review X* 4, 041019 (2014).
- [164] Pereira, V. M., Castro Neto, A. H., Liang, H. Y. & Mahadevan, L. Geometry, mechanics, and electronics of singular structures and wrinkles in graphene. *Physical Review Letters* 105, 156603 (2010).
- [165] Chen, J.-H. et al. Charged-impurity scattering in graphene. Nature Physics 4, 377–381 (2008).
- [166] Jang, C. et al. Tuning the effective fine structure constant in graphene: Opposing effects of dielectric screening on short- and long-range potential scattering. *Physical Review Letters* 101, 146805 (2008).
- [167] Hwang, E. H., Adam, S. & Sarma, S. D. Carrier transport in two-dimensional graphene layers. *Physical Review Letters* 98, 186806 (2007).
- [168] Tan, Y.-W. et al. Measurement of scattering rate and minimum conductivity in graphene. Physical Review Letters 99, 246803 (2007).
- [169] Tan, Y.-W., Zhang, Y., Stormer, H. L. & Kim, P. Temperature dependent electron transport in graphene. *The European Physical Journal Special Topics* 148, 15–18 (2007).

- [170] Heo, J. et al. Nonmonotonic temperature dependent transport in graphene grown by chemical vapor deposition. *Physical Review B* 84, 035421 (2011).
- [171] Li, Q., Hwang, E. H. & Das Sarma, S. Disorder-induced temperature-dependent transport in graphene: Puddles, impurities, activation, and diffusion. *Physical Review B* 84, 115442 (2011).
- [172] Hwang, E. H. & Das Sarma, S. Dielectric function, screening, and plasmons in two-dimensional graphene. *Physical Review B* 75, 205418 (2007).
- [173] Hwang, E. H. & Das Sarma, S. Screening-induced temperature-dependent transport in two-dimensional graphene. *Physical Review B* 79, 165404 (2009).
- [174] Das Sarma, S. Theory of finite-temperature screening in a disordered twodimensional electron gas. *Physical Review B* 33, 5401–5405 (1986).
- [175] Ando, T., Fowler, A. B. & Stern, F. Electronic properties of two-dimensional systems. *Reviews of Modern Physics* 54, 437–672 (1982).
- [176] Lyo, S. K. The effect of level broadening on the screening in a degenerate twodimensional electron gas. *Journal of Physics C: Solid State Physics* 21, 3577–3583 (1988).
- [177] Efetov, D. K. & Kim, P. Controlling electron-phonon interactions in graphene at ultrahigh carrier densities. *Physical Review Letters* 105, 256805 (2010).
- [178] Chen, J.-H., Jang, C., Xiao, S., Ishigami, M. & Fuhrer, M. S. Intrinsic and extrinsic performance limits of graphene devices on SiO2. *Nature Nanotechnology* 3, 206–209 (2008).
- [179] Tewary, V. K. & Yang, B. Singular behavior of the debye-waller factor of graphene. *Physical Review B* 79, 125416 (2009).
- [180] Hwang, E. H. & Sarma, S. D. Acoustic phonon scattering limited carrier mobility in two-dimensional extrinsic graphene. *Physical Review B* 77, 115449 (2008).
- [181] Woods, L. M. & Mahan, G. D. Electron-phonon effects in graphene and armchair (10,10) single-wall carbon nanotubes. *Physical Review B* 61, 10651–10663 (2000).
- [182] Ono, S. & Sugihara, K. Theory of the transport properties in graphite. Journal of the Physical Society of Japan 21, 861–868 (1966).
- [183] Vasko, F. T. & Ryzhii, V. Voltage and temperature dependencies of conductivity in gated graphene. *Physical Review B* 76, 233404 (2007).

- [184] Kaasbjerg, K., Thygesen, K. S. & Jacobsen, K. W. Unraveling the acoustic electron-phonon interaction in graphene. *Physical Review B* 85, 165440 (2012).
- [185] Cong, X. et al. Probing the acoustic phonon dispersion and sound velocity of graphene by raman spectroscopy. Carbon 149, 19–24 (2019).
- [186] Stauber, T., Peres, N. M. R. & Guinea, F. Electronic transport in graphene: A semiclassical approach including midgap states. *Physical Review B* 76, 205423 (2007).
- [187] Castro, E. V. et al. Limits on charge carrier mobility in suspended graphene due to flexural phonons. *Physical Review Letters* 105, 266601 (2010).
- [188] Ochoa, H., Castro, E. V., Katsnelson, M. I. & Guinea, F. Temperature-dependent resistivity in bilayer graphene due to flexural phonons. *Physical Review B* 83, 235416 (2011).
- [189] Piscanec, S., Lazzeri, M., Mauri, F., Ferrari, A. C. & Robertson, J. Kohn anomalies and electron-phonon interactions in graphite. *Physical Review Letters* 93, 185503 (2004).
- [190] Sohier, T. et al. Phonon-limited resistivity of graphene by first-principles calculations: Electron-phonon interactions, strain-induced gauge field, and boltzmann equation. Physical Review B 90, 125414 (2014).
- [191] You, Y. G. et al. Role of remote interfacial phonons in the resistivity of graphene. Applied Physics Letters 115, 043104 (2019).
- [192] Zou, K., Hong, X., Keefer, D. & Zhu, J. Deposition of high-quality hfo<sub>2</sub> on graphene and the effect of remote oxide phonon scattering. *Physical Review Letters* 105, 126601 (2010).
- [193] Lin, I.-T. & Liu, J.-M. Surface polar optical phonon scattering of carriers in graphene on various substrates. *Applied Physics Letters* 103 (2013).
- [194] Feldman, B. E., Martin, J. & Yacoby, A. Broken-symmetry states and divergent resistance in suspended bilayer graphene. *Nature Physics* 5, 889–893 (2009).
- [195] Das Sarma, S. & Hwang, E. H. Density-dependent electrical conductivity in suspended graphene: Approaching the dirac point in transport. *Physical Review* B 87, 035415 (2013).

- [196] Heinzel, T. Mesoscopic electronics in solid state nanostructures (Wiley-VCH Verl., Weinheim, 2010), 3., completely rev. and enlarged ed. edn. Literaturangaben.
- [197] Nazarov, J. V. & Blanter, Y. M. Quantum Transport (Cambridge University Press, Cambridge, 2009).
- [198] McCann, E. et al. Weak-localization magnetoresistance and valley symmetry in graphene. Physical Review Letters 97 (2006).
- [199] Tikhonenko, F. V., Horsell, D. W., Gorbachev, R. V. & Savchenko, A. K. Weak localization in graphene flakes. *Physical Review Letters* 100, 056802 (2008).
- [200] Tikhonenko, F. V., Kozikov, A. A., Savchenko, A. K. & Gorbachev, R. V. Transition between electron localization and antilocalization in graphene. *Physical Review Letters* 103 (2009).
- [201] Lundeberg, M. B. & Folk, J. A. Rippled graphene in an in-plane magnetic field: Effects of a random vector potential. *Physical Review Letters* 105, 146804 (2010).
- [202] Zihlmann, S. et al. Out-of-plane corrugations in graphene based van der waals heterostructures. Physical Review B 102, 195404 (2020).
- [203] McCann, E. & Fal'ko, V. I.  $z \rightarrow zSymmetry$  of spin-orbit coupling and weak localization in graphene. *Physical Review Letters* **108** (2012).
- [204] Ilić, S., Meyer, J. S. & Houzet, M. Weak localization in transition metal dichalcogenide monolayers and their heterostructures with graphene. *Physical Review B* 99 (2019).
- [205] Hwang, E. H. & Das Sarma, S. Graphene magnetoresistance in a parallel magnetic field: Spin polarization effect. *Physical Review B* 80, 075417 (2009).
- [206] Meyer, J. S., Altland, A. & Altshuler, B. L. Quantum transport in parallel magnetic fields: A realization of the berry-robnik symmetry phenomenon. *Physical Review Letters* 89, 206601 (2002).
- [207] Mathur, H. & Baranger, H. U. Random berry phase magnetoresistance as a probe of interface roughness in si mosfet's. *Physical Review B* 64, 235325 (2001).
- [208] Minkov, G. M. et al. Transverse negative magnetoresistance of two-dimensional structures in the presence of a strong in-plane magnetic field: Weak localization as a probe of interface roughness. *Physical Review B* 70, 035304 (2004).

- [209] Beenakker, C. W. J. & van Houten, H. Quantum transport in semiconductor nanostructures. In Semiconductor Heterostructures and Nanostructures, 1–228 (Elsevier, 1991).
- [210] Sólyom, J. (ed.) Fundamentals of the Physics of Solids. SpringerLink (Springer Berlin Heidelberg, Berlin, Heidelberg, 2009).
- [211] McClure, J. W. Band structure of graphite and de haas-van alphen effect. *Phys-ical Review* 108, 612–618 (1957).
- [212] Soule, D. E., McClure, J. W. & Smith, L. B. Study of the shubnikov-de haas effect. determination of the fermi surfaces in graphite. *Physical Review* 134, A453–A470 (1964).
- [213] Abrikosov, A. A. Fundamentals of the theory of metals (North Holland, 1988).
- [214] Adams, E. & Blount, E. Energy bands in the presence of an external force field—ii. Journal of Physics and Chemistry of Solids 10, 286–303 (1959).
- [215] Isihara, A. & Smrcka, L. Density and magnetic field dependences of the conductivity of two-dimensional electron systems. *Journal of Physics C: Solid State Physics* 19, 6777–6789 (1986).
- [216] Sharapov, S. G., Gusynin, V. P. & Beck, H. Magnetic oscillations in planar systems with the dirac-like spectrum of quasiparticle excitations. *Physical Review* B 69, 075104 (2004).
- [217] Gusynin, V. P. & Sharapov, S. G. Magnetic oscillations in planar systems with the dirac-like spectrum of quasiparticle excitations. ii. transport properties. *Physical Review B* 71, 125124 (2005).
- [218] Peres, N. M. R., Guinea, F. & Castro Neto, A. H. Electronic properties of disordered two-dimensional carbon. *Physical Review B* 73, 125411 (2006).
- [219] McCann, E. & Fal'ko, V. I. Landau-level degeneracy and quantum hall effect in a graphite bilayer. *Physical Review Letters* 96, 086805 (2006).
- [220] Novoselov, K. S. *et al.* Unconventional quantum hall effect and berry's phase of  $2\pi$  in bilayer graphene. *Nature Physics* **2**, 177–180 (2006).
- [221] Wu, H.-C. et al. Enhanced shubnikov-de haas oscillation in nitrogen-doped graphene. ACS Nano 9, 7207–7214 (2015).

- [222] He, L. et al. Quantum transport evidence for the three-dimensional dirac semimetal phase incd3as2. Physical Review Letters 113, 246402 (2014).
- [223] Matsumoto, N. et al. Shubnikov-de haas measurements on a high mobility monolayer graphene flake sandwiched between boron nitride sheets. Journal of Physics: Condensed Matter 29, 225301 (2017).
- [224] Klitzing, K. v., Dorda, G. & Pepper, M. New method for high-accuracy determination of the fine-structure constant based on quantized hall resistance. *Physical Review Letters* 45, 494–497 (1980).
- [225] Poux, A. et al. Microscopic model for the magnetic-field-driven breakdown of the dissipationless state in the integer and fractional quantum hall effect. *Physical Review B* 94, 075411 (2016).
- [226] Datta, S. Electronic Transport in Mesoscopic Systems (Cambridge University Press, 1995).
- [227] Ferry, D. K. Transport in nanostructures (Cambridge University Press, Cambridge, UK, 2009), second edition edn. Includes bibliographical references and index.
- [228] Tsoi, V. S., Bass, J. & Wyder, P. Studying conduction-electron/interface interactions using transverse electron focusing. *Reviews of Modern Physics* 71, 1641–1693 (1999).
- [229] Bohr, N. Niels Bohr Collected Works, vol. Vol. 3 (North-Holland, Amsterdam [u.a.], 1976).
- [230] Davies, N. et al. Skipping and snake orbits of electrons: Singularities and catastrophes. Physical Review B 85, 155433 (2012).
- [231] Patel, A. A., Davies, N., Cheianov, V. & Fal'ko, V. I. Classical and quantum magneto-oscillations of current flow near ap-njunction in graphene. *Physical Review B* 86, 081413 (2012).
- [232] Taychatanapat, T., Watanabe, K., Taniguchi, T. & Jarillo-Herrero, P. Electrically tunable transverse magnetic focusing in graphene. *Nature Physics* 9, 225–229 (2013).
- [233] Lee, M. et al. Ballistic miniband conduction in a graphene superlattice. Science 353, 1526–1529 (2016).

- [234] Berdyugin, A. I. et al. Minibands in twisted bilayer graphene probed by magnetic focusing. Science Advances 6 (2020).
- [235] Beenakker, C. W. J., Houten, H. v. & Wees, B. J. v. Mode interference effect in coherent electron focusing. *Europhysics Letters (EPL)* 7, 359–364 (1988).
- [236] Aidala, K. E. et al. Imaging magnetic focusing of coherent electron waves. Nature Physics 3, 464–468 (2007).
- [237] Rakyta, P., Kormányos, A., Cserti, J. & Koskinen, P. Exploring the graphene edges with coherent electron focusing. *Physical Review B* 81, 115411 (2010).
- [238] Cheianov, V. V., Fal'ko, V. & Altshuler, B. L. The focusing of electron flow and a veselago lens in graphene p-n junctions. *Science* **315**, 1252–1255 (2007).
- [239] van Son, P. C., van Kempen, H. & Wyder, P. New method to study the electronphonon interaction in metals. *Physical Review Letters* 58, 1567–1570 (1987).
- [240] Gupta, A. et al. Precision measurement of electron-electron scattering in GaAs/AlGaAs using transverse magnetic focusing. Nature Communications 12 (2021).
- [241] Huang, Y. et al. Reliable exfoliation of large-area high-quality flakes of graphene and other two-dimensional materials. ACS Nano 9, 10612–10620 (2015).
- [242] Blake, P. et al. Making graphene visible. Applied Physics Letters 91, 063124 (2007).
- [243] Ni, Z. H. et al. Graphene thickness determination using reflection and contrast spectroscopy. Nano Letters 7, 2758–2763 (2007).
- [244] Wang, X., Zhao, M. & Nolte, D. D. Optical contrast and clarity of graphene on an arbitrary substrate. Applied Physics Letters 95 (2009).
- [245] Lee, H. et al. Nondestructive thickness determination of polymers based on optical contrast of graphene. Applied Nanoscience 13, 4427–4435 (2022).
- [246] Zomer, P. J., Guimarães, M. H. D., Brant, J. C., Tombros, N. & van Wees, B. J. Fast pick up technique for high quality heterostructures of bilayer graphene and hexagonal boron nitride. *Applied Physics Letters* 105, 013101 (2014).
- [247] Neumann, C. *et al.* Raman spectroscopy as probe of nanometre-scale strain variations in graphene. *Nature Communications* **6** (2015).

- [248] Eaton, P. & West, P. Atomic Force Microscopy (Oxford University Press, 2010).
- [249] Levy, N. et al. Strain-induced pseudo-magnetic fields greater than 300 tesla in graphene nanobubbles. Science 329, 544–547 (2010).
- [250] Qi, Z. et al. Pseudomagnetic fields in graphene nanobubbles of constrained geometry: A molecular dynamics study. Physical Review B 90, 125419 (2014).
- [251] Ihn, T. Semiconductor Nanostructures: Quantum states and electronic transport (Oxford University Press, Oxford, 2010).
- [252] Weis, J. & von Klitzing, K. Metrology and microscopic picture of the integer quantum hall effect. *Philosophical Transactions of the Royal Society A: Mathematical, Physical and Engineering Sciences* **369**, 3954–3974 (2011).
- [253] Zeng, Y. et al. High-quality magnetotransport in graphene using the edge-free corbino geometry. Physical Review Letters 122, 137701 (2019).
- [254] Rai-Choudhury, P. Handbook of Microlithography, Micromachining, and Microfabrication. Volume 1: Microlithography (SPIE PRESS, 1997).
- [255] Pobell, F. Matter and Methods at Low Temperatures (Springer, Berlin, 1996), second edition edn.
- [256] Nielsen, J. H. et al. Qcodes. https://github.com/microsoft/Qcodes (2025).
- [257] Castro, E. V. et al. Biased bilayer graphene: Semiconductor with a gap tunable by the electric field effect. Physical Review Letters 99, 216802 (2007).
- [258] Castro, E. V. et al. Electronic properties of a biased graphene bilayer. Journal of Physics: Condensed Matter 22, 175503 (2010).
- [259] Zhang, Y. et al. Direct observation of a widely tunable bandgap in bilayer graphene. Nature 459, 820–823 (2009).
- [260] Taychatanapat, T. & Jarillo-Herrero, P. Electronic transport in dual-gated bilayer graphene at large displacement fields. *Physical Review Letters* 105, 166601 (2010).
- [261] Shklovskii, B. I. & Efros, A. L. Electronic Properties of Doped Semiconductors (Springer Berlin Heidelberg, 1984).
- [262] Sui, M. et al. Gate-tunable topological valley transport in bilayer graphene. Nature Physics 11, 1027–1031 (2015).

- [263] Zou, K. & Zhu, J. Transport in gapped bilayer graphene: The role of potential fluctuations. *Physical Review B* 82, 081407 (2010).
- [264] Lee, J. *et al.* Transitions of electrical conduction mechanism in graphene flake van der waals thin film. *Materials Today Communications* **39**, 108859 (2024).
- [265] Bhardwaj, P. & Singh, S. Pressure induced structural phase transitions—a review. Open Chemistry 10, 1391–1422 (2012).
- [266] Fülöp, B. et al. New method of transport measurements on van der waals heterostructures under pressure. Journal of Applied Physics 130, 064303 (2021).
- [267] Kormányos, A. et al. k · p theory for two-dimensional transition metal dichalcogenide semiconductors. 2D Materials 2, 022001 (2015).
- [268] Masseroni, M. et al. Spin-orbit proximity in mos2/bilayer graphene heterostructures. Nature Communications 15 (2024).
- [269] Seiler, A. M. *et al.* Layer-selective spin-orbit coupling and strong correlation in bilayer graphene (2024). 2403.17140.
- [270] Khoo, J. Y. & Levitov, L. Tunable quantum hall edge conduction in bilayer graphene through spin-orbit interaction. *Physical Review B* 98, 115307 (2018).
- [271] Wang, D. et al. Quantum hall effect measurement of spin-orbit coupling strengths in ultraclean bilayer graphene/WSe2 heterostructures. Nano Letters 19, 7028– 7034 (2019).
- [272] Zhang, Y. et al. Enhanced superconductivity in spin–orbit proximitized bilayer graphene. Nature 613, 268–273 (2023).
- [273] Szentpéteri, B. et al. Tuning the proximity induced spin-orbit coupling in bilayer graphene/wse<sub>2</sub> heterostructures with pressure (2024). 2409.20062.
- [274] dos Santos, J. M. B. L., Peres, N. M. R. & Neto, A. H. C. Continuum model of the twisted graphene bilayer. *Physical Review B* 86, 155449 (2012).
- [275] de Laissardière, G. T., Mayou, D. & Magaud, L. Numerical studies of confined states in rotated bilayers of graphene. *Physical Review B* 86, 125413 (2012).
- [276] Fang, S. & Kaxiras, E. Electronic structure theory of weakly interacting bilayers. *Physical Review B* 93, 235153 (2016).
- [277] Szentpéteri, B. et al. Tailoring the band structure of twisted double bilayer graphene with pressure. Nano Letters 21, 8777–8784 (2021).

- [278] Burg, G. W. et al. Correlated insulating states in twisted double bilayer graphene. Physical Review Letters 123, 197702 (2019).
- [279] Cao, Y. et al. Tunable correlated states and spin-polarized phases in twisted bilayer-bilayer graphene. Nature 583, 215–220 (2020).
- [280] He, M. et al. Symmetry breaking in twisted double bilayer graphene. Nature Physics 17, 26–30 (2020).
- [281] Rickhaus, P. et al. Correlated electron-hole state in twisted double-bilayer graphene. Science 373, 1257–1260 (2021).
- [282] Wang, Y. et al. Emergent symmetry and valley chern insulator in twisted doublebilayer graphene. Physical Review Letters 133, 246401 (2024).
- [283] Liu, X. et al. Tunable spin-polarized correlated states in twisted double bilayer graphene. Nature 583, 221–225 (2020).
- [284] Shen, C. et al. Correlated states in twisted double bilayer graphene. Nature Physics 16, 520–525 (2020).
- [285] Liu, L. et al. Isospin competitions and valley polarized correlated insulators in twisted double bilayer graphene. Nature Communications 13 (2022).
- [286] Kuiri, M. et al. Spontaneous time-reversal symmetry breaking in twisted double bilayer graphene. Nature Communications 13 (2022).
- [287] Koshino, M. Band structure and topological properties of twisted double bilayer graphene. *Physical Review B* 99, 235406 (2019).
- [288] Lee, J. Y. et al. Theory of correlated insulating behaviour and spin-triplet superconductivity in twisted double bilayer graphene. Nature Communications 10, 5333 (2019).
- [289] Crosse, J. A., Nakatsuji, N., Koshino, M. & Moon, P. Hofstadter butterfly and the quantum hall effect in twisted double bilayer graphene. *Physical Review B* 102, 035421 (2020).
- [290] Wang, Y.-X., Li, F. & Zhang, Z.-Y. Phase diagram and orbital chern insulator in twisted double bilayer graphene. *Physical Review B* 103, 115201 (2021).
- [291] Lin, X., Zhu, H. & Ni, J. Pressure-induced gap modulation and topological transitions in twisted bilayer and twisted double bilayer graphene. *Physical Review* B 101, 155405 (2020).

- [292] Haddadi, F., Wu, Q., Kruchkov, A. J. & Yazyev, O. V. Moiré flat bands in twisted double bilayer graphene. *Nano Letters* 20, 2410–2415 (2020).
- [293] Kim, K. et al. van der waals heterostructures with high accuracy rotational alignment. Nano Letters 16, 1989–1995 (2016).
- [294] MacDonald, A. H. Landau-level subband structure of electrons on a square lattice. *Physical Review B* 28, 6713–6717 (1983).
- [295] Solozhenko, V., Will, G. & Elf, F. Isothermal compression of hexagonal graphitelike boron nitride up to 12 gpa. Solid State Communications 96, 1–3 (1995).
- [296] Yankowitz, M. et al. Dynamic band-structure tuning of graphene moiré superlattices with pressure. Nature 557, 404–408 (2018).
- [297] Carr, S., Fang, S., Jarillo-Herrero, P. & Kaxiras, E. Pressure dependence of the magic twist angle in graphene superlattices. *Physical Review B* 98, 085144 (2018).
- [298] Brown, E. Bloch electrons in a uniform magnetic field. *Physical Review* 133, A1038–A1044 (1964).
- [299] Zak, J. Magnetic translation group. *Physical Review* **134**, A1602–A1606 (1964).
- [300] Hwang, C. et al. Fermi velocity engineering in graphene by substrate modification. Scientific Reports 2, 590 (2012).
- [301] Bessler, R., Duerig, U. & Koren, E. The dielectric constant of a bilayer graphene interface. Nanoscale Advances 1, 1702–1706 (2019).
- [302] Rickhaus, P. et al. Gap opening in twisted double bilayer graphene by crystal fields. Nano Letters 19, 8821–8828 (2019).
- [303] Wu, Q., Liu, J., Guan, Y. & Yazyev, O. V. Landau levels as a probe for band topology in graphene moiré superlattices. *Physical Review Letters* **126**, 056401 (2021).
- [304] Zhang, S., Dai, X. & Liu, J. Spin-polarized nematic order, quantum valley hall states, and field-tunable topological transitions in twisted multilayer graphene systems. *Physical Review Letters* **128**, 026403 (2022).
- [305] Zhang, F., Jung, J., Fiete, G. A., Niu, Q. & MacDonald, A. H. Spontaneous quantum hall states in chirally stacked few-layer graphene systems. *Physical Review Letters* 106, 156801 (2011).

- [306] Kedves, M. et al. Stabilizing the inverted phase of a wse2/blg/wse2 heterostructure via hydrostatic pressure. Nano Letters 23, 9508–9514 (2023).
- [307] Fülöp, B. et al. Boosting proximity spin-orbit coupling in graphene/WSe2 heterostructures via hydrostatic pressure. npj 2D Materials and Applications 5, 82 (2021).
- [308] Gorbachev, R. V., Tikhonenko, F. V., Mayorov, A. S., Horsell, D. W. & Savchenko, A. K. Weak localization in bilayer graphene. *Physical Review Letters* 98, 176805 (2007).
- [309] Völkl, T. *et al.* Magnetotransport in heterostructures of transition metal dichalcogenides and graphene. *Physical Review B* **96**, 125405 (2017).
- [310] Arora, H. S. et al. Superconductivity in metallic twisted bilayer graphene stabilized by wse2. Nature 583, 379–384 (2020).
- [311] Alsharari, A. M., Asmar, M. M. & Ulloa, S. E. Topological phases and twisting of graphene on a dichalcogenide monolayer. *Physical Review B* 98, 195129 (2018).
- [312] Dan, Y., Lu, Y., Kybert, N. J., Luo, Z. & Johnson, A. T. C. Intrinsic response of graphene vapor sensors. *Nano Letters* 9, 1472–1475 (2009).
- [313] Wang, L. et al. Negligible environmental sensitivity of graphene in a hexagonal boron nitride/graphene/h-bn sandwich structure. ACS Nano 6, 9314–9319 (2012).
- [314] Chakraborti, H. *et al.* Electron wave and quantum optics in graphene. *Journal of Physics: Condensed Matter* **36**, 393001 (2024).
- [315] Bolotin, K. I., Ghahari, F., Shulman, M. D., Stormer, H. L. & Kim, P. Observation of the fractional quantum hall effect in graphene. *Nature* 462, 196–199 (2009).
- [316] Crossno, J. et al. Observation of the dirac fluid and the breakdown of the wiedemann-franz law in graphene. Science 351, 1058–1061 (2016).
- [317] Bandurin, D. A. et al. Negative local resistance caused by viscous electron backflow in graphene. Science 351, 1055–1058 (2016).
- [318] Haigh, S. J. et al. Cross-sectional imaging of individual layers and buried interfaces of graphene-based heterostructures and superlattices. Nature Materials 11, 764–767 (2012).
- [319] Zhong, H., Zhang, Z., Xu, H., Qiu, C. & Peng, L.-M. Comparison of mobility extraction methods based on field-effect measurements for graphene. *AIP Advances* 5, 057136 (2015).
- [320] Gosling, J. H. *et al.* Universal mobility characteristics of graphene originating from charge scattering by ionised impurities. *Communications Physics* 4 (2021).
- [321] Jo, M. et al. Quantum hall valley splitters and a tunable mach-zehnder interferometer in graphene. Physical Review Letters 126, 146803 (2021).
- [322] Altshuler, B. L., Aronov, A. G. & Khmelnitsky, D. E. Effects of electron-electron collisions with small energy transfers on quantum localisation. *Journal of Physics C: Solid State Physics* 15, 7367–7386 (1982).
- [323] Narozhny, B. N., Zala, G. & Aleiner, I. L. Interaction corrections at intermediate temperatures: Dephasing time. *Physical Review B* 65, 180202 (2002).
- [324] Horsell, D. et al. Mesoscopic conductance fluctuations in graphene. Solid State Communications 149, 1041–1045 (2009).
- [325] Ki, D.-K., Jeong, D., Choi, J.-H., Lee, H.-J. & Park, K.-S. Inelastic scattering in a monolayer graphene sheet: A weak-localization study. *Physical Review B* 78, 125409 (2008).
- [326] Fratini, S. & Guinea, F. Substrate-limited electron dynamics in graphene. *Phys-ical Review B* 77, 195415 (2008).
- [327] Konar, A., Fang, T. & Jena, D. Effect of high-κ gate dielectrics on charge transport in graphene-based field effect transistors. *Physical Review B* 82, 115452 (2010).
- [328] Perebeinos, V. & Avouris, P. Inelastic scattering and current saturation in graphene. *Physical Review B* 81, 195442 (2010).
- [329] Ong, Z.-Y. & Fischetti, M. V. Theory of interfacial plasmon-phonon scattering in supported graphene. *Physical Review B* 86, 165422 (2012).
- [330] Schiefele, J., Sols, F. & Guinea, F. Temperature dependence of the conductivity of graphene on boron nitride. *Physical Review B* **85**, 195420 (2012).
- [331] Geick, R., Perry, C. H. & Rupprecht, G. Normal modes in hexagonal boron nitride. *Physical Review* 146, 543–547 (1966).

- [332] Giovannetti, G., Khomyakov, P. A., Brocks, G., Kelly, P. J. & van den Brink, J. Substrate-induced band gap in graphene on hexagonal boron nitride:ab initiodensity functional calculations. *Physical Review B* 76, 073103 (2007).
- [333] Slotman, G. J., de Wijs, G. A., Fasolino, A. & Katsnelson, M. I. Phonons and electron-phonon coupling in graphene-h-bn heterostructures. *Annalen der Physik* 526, 381–386 (2014).
- [334] Segura, A. et al. High-pressure softening of the out-of-plane a2u(transverse-optic) mode of hexagonal boron nitride induced by dynamical buckling. The Journal of Physical Chemistry C 123, 17491–17497 (2019).
- [335] Ouyang, W. et al. Mechanical and tribological properties of layered materials under high pressure: Assessing the importance of many-body dispersion effects. Journal of Chemical Theory and Computation 16, 666–676 (2019).
- [336] Rooks, M. J. et al. Low stress development of poly(methylmethacrylate) for high aspect ratio structures. Journal of Vacuum Science and Technology B: Microelectronics and Nanometer Structures Processing, Measurement, and Phenomena 20, 2937–2941 (2002).
- [337] Fischetti, M. V., Neumayer, D. A. & Cartier, E. A. Effective electron mobility in si inversion layers in metal-oxide-semiconductor systems with a high-κ insulator: The role of remote phonon scattering. *Journal of Applied Physics* **90**, 4587–4608 (2001).
- [338] Chaves, A. & Porto, S. Generalized lyddane-sachs-teller relation. Solid State Communications 13, 865–868 (1973).
- [339] Ong, Z.-Y. & Fischetti, M. V. Theory of remote phonon scattering in top-gated single-layer graphene. *Physical Review B* 88, 045405 (2013).
- [340] Ong, Z.-Y. & Fischetti, M. V. Erratum: Theory of interfacial plasmon-phonon scattering in supported graphene [phys. rev. b86, 165422 (2012)]. *Physical Review* B 86, 199904 (2012).
- [341] Liu, L. *et al.* First-principles simulation of raman spectra and structural properties of quartz up to 5 gpa. *Chinese Physics B* **24**, 127401 (2015).