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Doctoral Dissertation

Monte Carlo Analysis of Carrier Transport in
Turbostratic Multilayer Graphene Systems

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Chapter 1

Introduction

1.1 Research Background

Graphene is flat monolayer allotrope of carbon atoms densely packed in a hexagonal structure as shown in Fig. 1.1. Although its electronic properties were theoretically studied much earlier [1], graphene was long regarded as difficult to realize experimentally. This was mainly due to the common expectation that atomically thin crystals would be unstable and could not exist freely [2]. However, the experimental success of obtaining few-layer graphitic sheets using micromechanical exfoliation [3] has initiated cascades of theoretical and experimental research of graphene [4–11]. This breakthrough made it possible to examine graphene’s intrinsic electronic, mechanical, and optical properties directly, leading to rapid progress in understanding its fundamental physics and accelerating research on graphene [12–19].

A remarkable electronic property of graphene is that its charge carriers behave as massless Dirac fermions with a Fermi velocity of $v_f \approx 10^8$ cm/s near the Dirac point

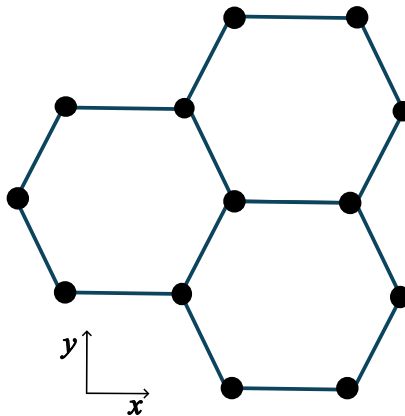


Figure 1.1: Carbon atoms arranged in a flat monolayer hexagonal lattice of graphene.

[4, 5]. Due to the linear energy-momentum relationship, charge carriers maintain very high velocities even at low energies, contributing to the exceptional mobility observed in pristine samples [20–22]. These unique transport characteristics make graphene a strong candidate for high-speed electronic applications and for exploring transport phenomena in two-dimensional systems [23–27].

Graphene has been experimentally obtained through various approaches, including epitaxial growth on SiC and chemical vapor deposition synthesis [28–33]. However, the mobility in fabricated graphene samples is often degraded primarily by the presence of charged impurities, alongside other structural imperfections [34, 35]. Consequently, to overcome these limitations, it is essential to investigate the intrinsic and extrinsic obstacles that hinder graphene’s high carrier mobility.

One of the intrinsic scattering mechanisms that affects the transport properties of graphene is phonon scattering [34, 36]. Although phonon scattering can be suppressed at low temperatures [37, 38], it remains an important factor in the application of room-temperature devices. Besides phonons, charged impurities also play an important role in determining the transport properties of graphene as an external scattering source [34, 39, 40]. These impurities appear between the graphene sheet and its substrate and induce potential fluctuations. As a result, the perturbations caused by charged impurities change the transport properties of carriers, resulting in reduced carrier mobility [35, 41]. Figure 1.2 illustrates the schematics of the potential landscape in free-standing graphene and in graphene placed on a substrate with impurities.

A few methods have been suggested to lessen the effects of impurities on the carriers. One of these methods is the use of colloidal quantum dots to cancel the fields

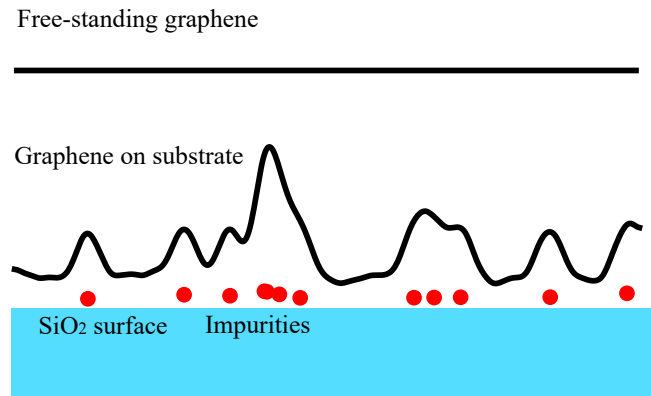


Figure 1.2: Schematic of free-standing graphene and impurity-induced potential fluctuations in substrate-supported graphene.

caused by the impurities [42, 43]. In addition, the choice of substrate for graphene also influences its mobility. Hence, changing the substrate from the commonly used SiO_2 to other materials such as h-BN leads to improved mobility [34, 44, 45].

Beside these approaches, which deal with the improvement of the mobility in the monolayer graphene, another option is to switch to multilayer graphene. This simple, yet structural change, has a unique merit. One major advantage is that the lower graphene layers screen the induced potential, effectively shielding the upper layers from these effects, thus improving their mobility. Specifically, the graphene system with turbostratic structure is attractive [46–49]. In turbostratic structure, each layer is positioned on top of other layer with a random degree of rotation, enabling each layer to preserve the electronic structure of the pristine graphene [46, 50, 51]. Thus, each graphene layer exhibits the transport properties of the pristine graphene.

1.2 Research Objectives and Thesis Organization

Based on the aforementioned background, this research aims to study the transport properties of carriers in different systems of turbostratic multilayer graphene. Particularly, this study examines the different phenomena affecting the transport properties, how the multilayer structure mitigates impurity scattering, and the factors limiting carrier transport, with an investigation of possible structural configurations to improve these properties.

The thesis is structured as follows. Chapter 2 introduces the overall structure of the multilayer graphene examined in this study. This chapter provides the details of the simulation framework, ranging from the methods used for calculating the electrostatic potential to the Monte Carlo simulation calculations. These calculation methods are common across the different systems examined in this study, and the distinctive features of each system are explained in their respective chapters. Chapter 3 presents the simulation results for turbostratic multilayer graphene, and subsequently analyzes the transport behavior in this turbostratic structure. Systems with different numbers of layers, along with varying impurity densities, are studied, and an analytical model explaining the impurity scattering effects on carrier mobility is introduced. Chapter 4 focuses on edge scattering in the multilayer graphene structure, and examines carrier transport under different scales of edge scattering. The calculation model is first explained, followed by the simulation results. This chapter concludes with an empirical model that reproduces the simulation results accurately. Chapter 5 introduces a new multilayer graphene structure, where twisted

bilayer graphene is placed between the turbostratic multilayer graphene and the substrate. The suppression of the Fermi velocity in the bilayer structure, which depends on the twist angle, enables the formation of a new class of multilayer graphene. In Chapter 5, the transport properties of these structures with different suppression ratios are explained. Chapter 6 provides a conclusion to the discussions and results presented in this thesis.

Chapter 2

Simulation Framework

2.1 Introduction

This chapter explains the overall structure of the simulation methods used in this research. The ultimate objective of this thesis is to study the transport properties of various multilayer graphene systems, which differ in aspects such as the number of layers and substrate impurity concentration. As a result, the focal points of subsequent chapters vary, covering factors that affect transport such as impurity density and edge scattering, as well as structural configurations designed to improve it, such as the inclusion of twisted bilayer graphene. Despite these different aspects, the underlying simulation method employed throughout all calculations is consistent, with specific modifications tailored according to each chapter's goals. Here, the basic structure of the multilayer system along with the foundational simulation method is explained.

The simulation is performed in two main parts. The first part involves calculating the impurity-induced potential profile for the multilayer system. This is done by self-consistently solving the Poisson equation using the Thomas-Fermi approximation. From the resulting potential profile for each layer, the fluctuating electric fields induced by the impurities are obtained, thereby allowing the extrinsic factors that influence carrier mobility to be determined. The second part of the simulation is the calculation of the transport properties by solving the Boltzmann transport equation using the Monte Carlo method. In this stage, phonon scattering mechanisms, which represent the intrinsic scattering sources, are also implemented alongside the impurity-induced fields. This makes it possible to study the trajectories of carriers as they move through the graphene sheet and are scattered by these mechanisms. The procedures for these calculations are described in the following sections.

2.2 System Structure

First, the structure of the multilayer graphene system studied in this research is presented. The structure described here has the foundational features common to all the subsequent chapters, and specifically, this structure is studied in Chapter 3. The following Chapters 4 and 5 have largely similar structures to that described here, with specific modifications in each chapter, tailored to the objectives of each one. Hence, the respective structural changes are described in those chapters.

A schematic diagram of the model used in this research is presented in Fig. 2.1. The system consists of a square-shaped SiO_2 substrate of size $L \times L$, with donor impurities on its surface. Except for the substrate surface, the rest of the substrate contains no impurities. The total number of impurities is denoted by N_{imp} , and the location of each impurity is determined stochastically. From these, the area density of impurities is given by $N_{\text{D}} = N_{\text{imp}}/(L \times L)$. On top of the substrate, M layers of graphene are placed. The uppermost part of the system is an insulating layer devoid of any charge, located above the last graphene layer. The size of each section of the system is as follows: the heights of the substrate and the insulating layer are h_{b} , and h_{t} respectively. The first graphene layer (the closest graphene layer to the substrate) is referred to as the first layer, and the farthest layer is called the last layer throughout this thesis) is at a distance of d_{sg} from the substrate. The interlayer distance between graphene layers is d_{gg} . The actual values of the parameters characterizing the system are listed in Table 2.1.

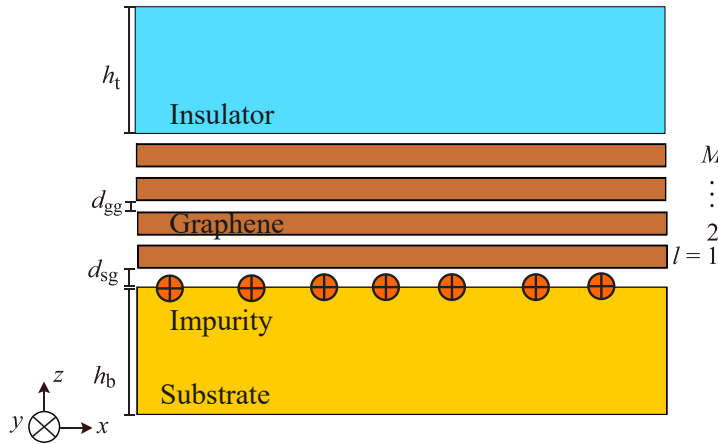


Figure 2.1: Schematic representation of the simulated system, showing the substrate with impurities, the M stacked graphene layers, and the insulating region above them.

Table 2.1: Structural parameters of the multilayer graphene system. All values are presented in nm.

Height of the substrate h_b	136
Height of the insulator h_t	136
Distance of the 1st graphene layer from the substrate d_{sg}	0.68
Distance between each graphene layer d_{gg}	0.34
Sheet thickness of graphene layer d_p	0.34
Length of the square-shaped graphene sheet L	100

2.3 Self-Consistent Calculation of the Electrostatic Potential

In order to obtain the potential profile of the multilayer graphene system, a few steps need to be carried out. First, the parameters defining the structure of the system, such as the number of graphene layers M and the size of the graphene sheets are determined. Then, the positions of the impurities are also determined. Next, the positions of the charged impurities are assigned to the mesh grid for the calculation of the grid-based charge density. Finally, the self-consistent calculation of the Poisson equation within the Thomas–Fermi approximation is performed. Here, the detailed procedure of each step is explained.

2.3.1 Impurity Position

The positions of donor impurities on the substrate surface are determined stochastically using uniformly distributed random numbers, ensuring an equal probability of locating an impurity anywhere on the substrate. These impurities remain fixed at their assigned positions throughout the calculations. However, a technical issue arises due to the use of periodic boundary conditions in the subsequent Monte Carlo transport simulation, which necessitate determining the impurity positions in a manner compatible with these conditions. If the impurity positions were generated completely randomly within the primary $L \times L$ domain, the potential near opposite boundaries would differ. Since the periodic boundaries in the transport simulation connect these edges, such discontinuities in the potential profile would lead to unwanted forces, especially significant at low external electric fields where impurity-induced fields dominate and therefore influence carrier transport.

To avoid this inconsistency, the impurity distribution is constructed to be periodic. Thus, the following mirroring procedure is used:

1. Impurity positions are first generated randomly within the primary $L \times L$ region ($0 < x, y < L$).
2. The impurity set is mirrored across the line $x = L$ to produce the adjacent domain to the right.
3. The original set is mirrored across $y = L$ to create the domain above.
4. Mirroring across both $x = L$ and $y = L$ completes the top-right region.

Through this process, a periodic $2L \times 2L$ impurity pattern is constructed, ensuring that the potential is continuous across the boundaries of the primary simulation domain. A schematic representation of this mirroring procedure is shown in Fig. 2.2. In this figure, the impurities in the lower-left region correspond to the initially generated random positions, while the remaining three regions illustrate the horizontally, vertically, and diagonally mirrored distributions. The arrows indicate how the impurity positions in the primary region are mapped to their corresponding mirrored locations. By constructing the impurity pattern in this manner, the potential landscape becomes fully consistent with the periodic boundary conditions imposed during the transport simulation, preventing artificial discontinuities at the boundaries.

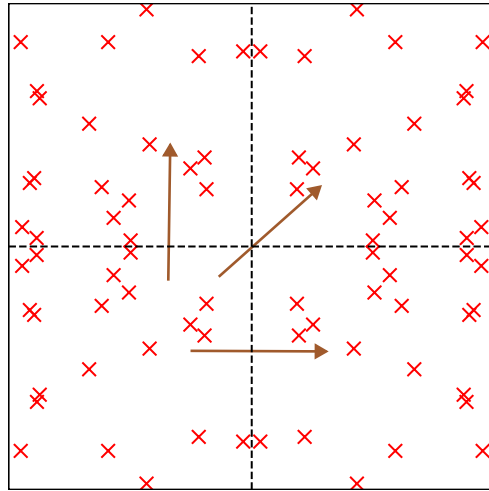


Figure 2.2: Schematic illustration of the impurity-mirroring procedure used to generate a periodic impurity pattern. Impurities are first placed randomly in the lower-left region and then mirrored horizontally, vertically, and diagonally to form the corresponding regions. The arrows indicate how impurity positions in the primary region map to their mirrored counterparts in the adjacent areas.

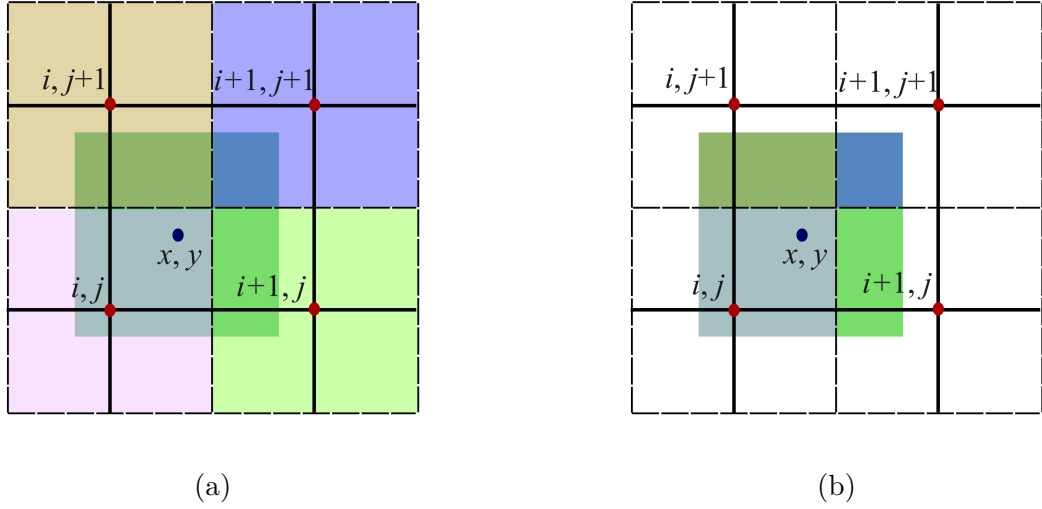


Figure 2.3: (a) Illustration of the cloud-in-cell (CIC) representation of an impurity. The impurity located at (x, y) is treated as the center of a finite-sized charged cloud, shown relative to the four surrounding grid cells indexed by (i, j) , $(i+1, j)$, $(i, j+1)$, and $(i+1, j+1)$. (b) The corresponding overlapping areas between the impurity cloud and each of the four neighboring grid cells, which determine the fractional charge assigned to each grid point in the CIC method.

2.3.2 Cloud-in-cell Method

For the numerical calculations, the system is discretized with a mesh spacing of $\delta = 1$ nm. The impurity charges are then assigned to the grid points using the cloud-in-cell (CIC) method [52], which distributes the charge of each impurity among its neighboring grid points. In this method, the coordinate of each impurity is considered to be the center of a finite-sized charged cloud. The four grid points surrounding the impurity are treated as the centers of the four surrounding cells. The charges are assigned to the grid points as follows: first, the overlapping area between the impurity cloud and each of the four surrounding grid cells is calculated. The charge assigned to a grid point is then determined by the ratio of this overlapping area to the area of the corresponding cell. Consequently, the charge is distributed proportionally, with grid points closer to the impurity receiving a larger share. Figure 2.3 shows the schematic illustrations of this procedure.

2.3.3 Outline of Self-consistent Calculation

Here, the outline of the self-consistent calculation is explained. Having an initial charge distribution of the system and using Poisson equation, the electrostatic po-

tential induced by the charges in the system can be obtained. Then, by using the Thomas-Fermi approximation, the new charge distribution of the system is calculated. In the self-consistent Poisson and Thomas-Fermi cycle, the calculations of the charge density and the electrostatic potential are repeatedly alternated until the change converges to a certain error range. Here, the self-consistent calculation is performed with charge neutrality valid in the system and Poisson equation is solved with appropriate boundary conditions applied to the system. The following relations simplify the aforementioned process.

Denoting F as the function which, given the input of electrostatic potential, yields n , the carrier density, and G as the function which yields electrostatic potential V from n , the following sequence holds:

$$\begin{aligned} n &= F[V] \quad (\text{Thomas-Fermi gives } n \text{ from } V), \\ V &= G[n] \quad (\text{Poisson equation gives } V \text{ from } n). \end{aligned} \tag{2.1}$$

Hence, the composite update would be

$$f[V] \equiv G(F[V]), \tag{2.2}$$

which yields

$$V = f[V]. \tag{2.3}$$

Therefore, iterations over the functions would be

$$V^{(0)} \xrightarrow{F} n^{(0)} \xrightarrow{G} V^{(1)} \xrightarrow{F} n^{(1)} \xrightarrow{G} V^{(2)} \dots \text{ until } \|V^{(k+1)} - V^{(k)}\| < \epsilon, \tag{2.4}$$

where ϵ is the predetermined error range used for the calculation. Hence, the aforementioned algorithm could be summarized as

1. Guess an initial potential $V^{(0)}$.
2. For $k = 0, 1, 2, \dots$:
 - (a) $n^{(k)} \leftarrow F[V^{(k)}]$ (compute density via Thomas-Fermi)
 - (b) $V^{(k+1)} \leftarrow G[n^{(k)}]$ (solve Poisson equation)
 - (c) If $\|V^{(k+1)} - V^{(k)}\| < \epsilon$, stop; else continue.
3. Final density: compute
$$n^{(k+1)} = F[V^{(k+1)}].$$
4. Return $V^{(k+1)}$ and $n^{(k+1)}$.

2.3.4 Poisson Equation

For a given charge density $\rho(x, y, z)$, Poisson equation

$$\nabla^2 \phi = -\frac{\rho}{\varepsilon}, \quad (2.5)$$

is solved where ε is the permittivity and $\phi(x, y, z)$ is the electrostatic potential. The permittivity of SiO_2 is assigned across the system. Both the charge density $\rho(x, y, z)$ and the potential $\phi(x, y, z)$ are discretized on the xy-plane with a grid size of 1 nm. Here, the charge density is formed by two sections. The first is the density of the charged donor impurities, $en_{\text{D}}(x, y)$, whose positions are randomly chosen on fixed locations on the substrate. Here, $n_{\text{D}}(x, y)$ denotes the impurity density at each point on (x, y) on the substrate. The total number of impurities N_{imp} is obtained by integrating the impurity density over the surface of the substrate. The second part of the charge density is the induced electron and hole densities in each layer of graphene. $n_l(x, y)$ and $p_l(x, y)$ denote the electron and hole densities for the l -th layer, where $l = 1, 2, \dots, M$. Contrary to the fixed impurity density, electron and hole densities adjust to the potential profile of the system in each iteration of the self-consistent calculation. It is noteworthy that the charge densities in this section are area densities, being in accordance with the stacked two-dimensional layers of graphene in the system. However, in the simulation, a parameter $d_{\text{p}} = 0.34 \text{ nm}$ is utilized to represent the thickness of each two-dimensional layer. Thus, the three-dimensional Poisson equation is solved, and the charge density is treated as a volume density. As for the boundary condition of the system, the potential is fixed to zero at the top and bottom of the system, thus applying a zero Dirichlet boundary condition, while on each of the four lateral faces, zero Neumann boundary conditions are applied where the derivative of the potential is set to vanish.

2.3.5 Carrier Density by Thomas-Fermi Approximation

Once the potential profiles for each layer of graphene are obtained in each self-consistent iteration of the calculation, the induced electron and hole densities are calculated for the given profile. The following linear dispersion relation near the Dirac points is assumed for each layer of the stacked graphene structure:

$$E = \hbar v_{\text{f}} |\mathbf{k}|, \quad (2.6)$$

where the magnitude of the 2D wave vector $\mathbf{k} = (k_x, k_y)$ is denoted by $|\mathbf{k}|$, and the Fermi velocity of $v_f = 10^8$ cm/s is used. Therefore, the density of states in each graphene layer is given by

$$g(E) = \frac{2|E|}{\pi\hbar^2 v_f^2}. \quad (2.7)$$

The Fermi-Dirac distribution function, $f(E) = [1 + \exp((E - \mu)/k_B T)]^{-1}$, with the Fermi level μ , temperature of T , and the Boltzmann constant k_B , is used to determine the carrier densities. Defining the Dirac point as the origin of the energy level and in accordance with the Thomas-Fermi approximation, it is considered that the electrostatic potential $\phi(\mathbf{r})$ is a very slowly varying function of $\mathbf{r} = (x, y, z)$, and hence, due to slowly varying potential, the local Dirac point is shifted by $\varphi(\mathbf{r}) = -e\phi(\mathbf{r})$ [53]. By utilizing this assumption, the local potential profile determines the carrier densities. Thus, the density of states of graphene at the local point S can be written as

$$g(E - \varphi_S) = g_c(E - \varphi_S) + g_v(E - \varphi_S), \quad (2.8)$$

where g_c denotes the density of states of the conduction band for energies higher than the local potential ($E > \varphi_S$), defined as

$$g_c(E - \varphi_S) = 2(E - \varphi_S)/(\pi\hbar^2 v_f^2). \quad (2.9)$$

Similarly, g_v represents the density of states of the valence band for the energies lower than the local potential ($E < \varphi_S$), defined as

$$g_v(E - \varphi_S) = 2(\varphi_S - E)/(\pi\hbar^2 v_f^2). \quad (2.10)$$

Thus, using each density of states, the induced electron density at the given point S in the graphene sheet, n_S , can be calculated by

$$n_S = \int_{\varphi_S}^{E_{\max}} g_c(E - \varphi_S) f(E) dE, \quad (2.11)$$

where $E_{\max}$ is the upper energy bound for the integration. Equation 2.11 yields the electron density corresponding to the occupied states above the Dirac point in the conduction band.

Similarly, the following is used to calculate the hole density p_S at the given point S :

$$p_S = \int_{E_{\min}}^{\varphi_S} g_v(E - \varphi_S) (1 - f(E)) dE, \quad (2.12)$$

where $E_{\min}$ is the lower energy bound for the integration. Here, the hole density in the valence band below the Dirac point is calculated.

The upper and lower bounds of the energy are set to be $E_{\max} = 5 \text{ eV}$ and $E_{\min} = -5 \text{ eV}$. Therefore, for a point S , the net carrier density is determined by

$$\begin{aligned}
 n_S - p_S &= \int_{\varphi_S}^{E_{\max}} g_c(E - \varphi_S) f(E) dE - \int_{E_{\min}}^{\varphi_S} g_v(E - \varphi_S) (1 - f(E)) dE \\
 &= \int_{\varphi_S}^{E_{\max}} g_c(E - \varphi_S) f(E) dE + \int_{E_{\min}}^{\varphi_S} g_v(E - \varphi_S) f(E) dE \\
 &\quad - \int_{E_{\min}}^{\varphi_S} g_v(E - \varphi_S) dE \\
 &= \int_{E_{\min}}^{E_{\max}} g(E - \varphi_S) f(E) dE - \int_{E_{\min}}^{\varphi_S} g_v(E - \varphi_S) dE.
 \end{aligned} \tag{2.13}$$

Taking into account the charge neutrality of the system, the number of the induced electrons in the system should be equal to the number of the induced holes plus the donor impurities. Eq. 2.13 gives the net carrier density at a point in the system, thus by integrating it for all the M layers of the graphene, the following equation is obtained:

$$\sum_{l=1}^M \int_0^L \int_0^L (n_S - p_S) dx dy = N_{\text{imp}}. \tag{2.14}$$

Eq. 2.14 denotes the charge neutrality of the system. Therefore, to enforce the charge neutrality in the system, an appropriate Fermi level for the whole system is determined. To be more specific, as mentioned earlier, the Dirac points are shifted by $-e\phi(\mathbf{r})$, thus the local potential perturbations are reflected in the calculation of the induced carriers. Simultaneously, a common Fermi level is adjusted for the entire system, in such a way that the Eq. 2.14 remains valid. Hence it can be interpreted that Eq. 2.13 is a function of the Fermi level, and the search for an appropriate Fermi level to fulfill Eq. 2.14 is the main numerical problem that needs to be solved. Here, the bisection method is used to determine it.

2.4 Monte Carlo Simulation Method

The transport properties of carriers in devices can be studied theoretically by solving the Boltzmann transport equations and Poisson equation. Solving the Boltzmann

transport equation directly is quite challenging, and thus using alternative methods to simulate the trajectories of carriers is more practical and desirable. One of the widely used methods for this purpose is the Monte Carlo simulation method. In the Monte Carlo method, the transport history of each individual carrier is updated and tracked throughout the simulation. The duration of the free-flight and the scattering mechanism that terminates it are determined stochastically. Additionally, since the band structure of the material under study along with its scattering behavior is incorporated in the Monte Carlo method, the transport properties of the material can be studied. With a large enough number of simulated trajectories, the final results provide a reliable approximation of the average behaviors of the carriers in real devices [54–56]. Hence, this approach captures the statistical behavior of carriers, leading to results that represent the ensemble-averaged transport properties of the system. Here, the implementation of the Monte Carlo simulation for the multilayer graphene system is explained.

2.4.1 Initial States

First, the initial states of the electrons are determined (the explanation here focuses on electrons, and only the differences for holes are mentioned separately). These initial states include the electron positions in the graphene sheet, which are generated using uniformly distributed random numbers across the simulation area, as described earlier. The electron energies are sampled so that they follow the equilibrium energy distribution, which is given by the product of the graphene density of states and the Fermi-Dirac distribution. To achieve this in the simulation, the rejection sampling method is used so that only the energy values matching the desired distribution are kept. The initial direction of motion of each carrier is also determined stochastically.

2.4.2 Free-flight

Here, the free-flight of the carriers is described. The free-flight refers to the movement of the carriers between the scattering events. In this study, the carriers are subjected to an in-plane external electric field in the x direction, and there is no additional field applied to the carriers along the y direction. The external field is denoted by

$$\mathbf{F}_{\text{ext}} = (F_{\text{ext},x}, 0). \quad (2.15)$$

This field is applied to drive the electrons from the left side of the graphene sheet to the right side, hence ($F_{\text{ext},x} < 0$). Additionally, the electrons are subjected to the local electric fields originating from the charged impurities on the substrate. These local fields, denoted by

$$\mathbf{F}_{\text{imp}} = (F_{\text{imp},x}, F_{\text{imp},y}), \quad (2.16)$$

are obtained from the self-consistently calculated potential profile ($\mathbf{F}_{\text{imp}} = -\nabla\phi$). Thus, the total electric field applied to the electrons during the free-flight is

$$\mathbf{F}_{\text{tot}} = \mathbf{F}_{\text{ext}} + \mathbf{F}_{\text{imp}}. \quad (2.17)$$

In graphene, the electrons move with the Fermi velocity during the free-flight. Here, the simulation is done for an ensemble of 10^4 electrons. The total duration of the simulation is defined as t_{fin} . The evolution of electrons' trajectories over t_{fin} is divided into smaller intervals. The duration of each interval is denoted by the time-step parameter t_s . The positions along with the components of the wave vector and the energy are updated in each time interval. The time intervals may contain no scattering events or one or more scattering events (in rarer cases). The beginning and end of the intervals are denoted by t_b and t_e , respectively. Therefore, at the beginning of the simulation for the first interval, $t_b = 0$ and $t_e = t_s$. Similarly, for the n -th interval, $t_b = (n-1)t_s$ and $t_e = nt_s$.

The duration of the free-flight time is related to the scattering rate, in which higher scattering rate leads to the shorter free-flight. Considering a system with m scattering mechanisms, the total scattering rate, Γ , is obtained by

$$\Gamma(E) = \sum_{k=1}^m \frac{1}{\tau_k(E)}, \quad (2.18)$$

where τ_k is the relaxation time for the k -th scattering mechanism. Since the scattering rate Γ varies with energy, the well-known method of introducing fictitious self-scattering into scattering mechanisms [57], which does not alter the carriers' momentum, but makes the scattering rate constant, is used. Hence the constant scattering rate Γ_0 is fixed and the duration of the free-flight, t_f , is then calculated by

$$t_f = -\Gamma_0^{-1} \ln r, \quad (2.19)$$

where r is a uniformly distributed random number between 0 and 1. Here, t_f is calculated for each electron, and denotes the duration during which no scattering events occur, and carriers are only influenced by $\mathbf{F}_{\text{tot}}$. Subsequently, a scattering

event occurs after the free-flight duration t_f . After the scattering event, another t_f is calculated using Eq. 2.19 to determine the duration of next free-flight. Consequently, the cumulative free-flight time $t_{f,\text{cum}}$ is defined as the aggregate of the free-flight durations from the first through the n -th scattering event, and expressed as

$$t_{f,\text{cum}}^{(n)} = \sum_{i=1}^n t_f^{(i)}. \quad (2.20)$$

Here, the cumulative free-flight time for each electron represents the occurrence time of the n -th scattering event. As the transport properties of the carriers are calculated by moving through the t_s , it is checked whether $t_{f,\text{cum}}$ falls inside the current time-step. If it does, it indicates that a scattering event is going to happen in the current time-step.

During the time-step, the x and y coordinates of the electrons at the beginning of the free-flight, x_b and y_b , are advanced to the new coordinates, x_e and y_e , at the end of the free-flight. This update is done in two scenarios: one in the absence of any scattering event within the time-step, where the new coordinates are obtained according to the following expressions:

$$x_e = x_b + t_s v_f \cos \alpha, \quad (2.21a)$$

$$y_e = y_b + t_s v_f \sin \alpha. \quad (2.21b)$$

Here, the electrons move with the Fermi velocity during the free-flight, and their direction of movement is denoted by the angle α , which is obtained using the components of the wave vectors at the beginning of the time-step, $k_{x,b}$ and $k_{y,b}$ by the following equation:

$$\alpha = \tan^{-1}(k_{y,b}/k_{x,b}). \quad (2.22)$$

Accordingly, the components of the wave vector are also updated until the end of the time-step by

$$k_{x,e} = k_{x,b} \mp \frac{et_s}{\hbar} (F_{\text{ext},x} + F_{\text{imp},x}), \quad (2.23a)$$

$$k_{y,e} = k_{y,b} \mp \frac{et_s}{\hbar} F_{\text{imp},y}. \quad (2.23b)$$

In Eqs. 2.23a and 2.23b, the negative signs correspond to the expressions for updating electron wave vectors, whereas the positive signs denote the analogous updates for hole wave vectors. Obtaining $k_{x,e}$ and $k_{y,e}$ furthermore yields the magnitude of

the wave vector,

$$|\mathbf{k}| = (k_{x,e}^2 + k_{y,e}^2)^{1/2} \quad (2.24)$$

as well as the new angle ($\alpha = \tan^{-1}(k_{y,e}/k_{x,e})$). Subsequently, the energy for each electron at the end of the time-step is calculated via Eq. 2.6.

Now when there is a scattering event in the time-step, the movement of the electrons is divided into two parts: one before the scattering, and one after. From the beginning of the time-step to $t_{f,cum}$, where the scattering happens, the electron's position is updated by

$$x_f = x_b + (t_{f,cum} - t_b)v_f \cos \alpha, \quad (2.25a)$$

$$y_f = y_b + (t_{f,cum} - t_b)v_f \sin \alpha. \quad (2.25b)$$

Here, x_f and y_f correspond to the electron's x and y coordinates when scattering happens. Once x_f and y_f are obtained, the wave-vector components are updated over the interval $(t_{f,cum} - t_b)$ using Eqs. 2.23a and 2.23b. Subsequently, the electron energy upon completion of free flight is calculated. In order to update the transport properties of the electrons after $t_{f,cum}$, first the type of the scattering event needs to be determined. Since the scattering rate varies with the energy, the probability of a certain scattering happening also varies with energy. Taking these into account and using the electrons' energy before scattering, the appropriate scattering mechanism is chosen. Specifically, a random number r is generated, and the specific mechanism l is selected by identifying the interval in the cumulative scattering where the following condition is met:

$$\frac{\sum_{i=1}^{l-1} \frac{1}{\tau_i(E)}}{\Gamma_0} \leq r < \frac{\sum_{i=1}^l \frac{1}{\tau_i(E)}}{\Gamma_0}, \quad (2.26)$$

Here, l ranges from 1 to $k + 1$ scattering mechanisms including self-scattering [54].

Depending on the type of the scattering mechanism, the electron might gain, lose, or retain its energy. Except in the case of self-scattering, the electron's trajectory is altered. Thus, a random number is used to randomize the direction of movement isotropically. The details of the scattering mechanisms considered here are described in Section 2.4.3. After determining the scattering mechanism and the electrons' new transport property attributes, the movement of the electrons is resumed until the end of the time step, using:

$$x_e = x_f + (t_e - t_{f,cum})v_f \cos \alpha', \quad (2.27a)$$

$$y_e = y_f + (t_e - t_{f,cum})v_f \sin \alpha', \quad (2.27b)$$

where α' denotes the direction of the electron movement after the scattering. Similarly, the components of the wave vector are updated by

$$k_{x,e} = k_{x,b} \mp \frac{e(t_e - t_{f,cum})}{\hbar} (F_{ext,x} + F_{imp,x}), \quad (2.28a)$$

$$k_{y,e} = k_{y,b} \mp \frac{e(t_e - t_{f,cum})}{\hbar} F_{imp,y}, \quad (2.28b)$$

and the energy of the electrons at the end of the time-step is also calculated.

2.4.3 Scattering Mechanism

The carriers in the graphene are scattered by phonons, besides the impurity scattering. Here the phonon scattering implementation is explained. Acoustic phonon (AP) and optical phonon (OP) scattering mechanisms are considered. The elastic AP scattering rate for electrons in the conduction band is expressed as [36, 58]

$$S_{ap}^{elas} = \frac{4E_D^2 k_B T}{\hbar^3 v_f^2 v_s^2 \rho_s} E, \quad (2.29)$$

where E_D denotes the deformation potential, ρ_s represents the density of graphene, and v_s is the sound velocity.

The inelastic scattering rates of electrons due to OP and AP interactions are given by [45, 58]

$$S_{op(ap)}^{inelas} = \frac{D_{f,op(ac)}^2}{\hbar^2 \omega_{op(ac)} \rho_s v_f^2} \left[(E + \hbar \omega_{op(ac)}) N_{op(ac)} + (E - \hbar \omega_{op(ac)}) (N_{op(ac)} + 1) \Theta(E - \hbar \omega_{op(ac)}) \right], \quad (2.30)$$

where $D_{f,op(ac)}$ represents the deformation field, $\omega_{op(ac)}$ is the optical (acoustic) phonon frequency and $N_{op(ac)}$ is the phonon occupation number. The Heaviside function, $\Theta(E)$, ensures that the electron energy is sufficient for phonon emission.

Table 2.2: Parameters used for phonon scattering rates.

Deformation potential (acoustic) E_D (eV)	4.6
Deformation field (optical) D_{op} (eV/cm)	1.0×10^9
Deformation field (acoustic) D_{ac} (eV/cm)	3.5×10^8
Density of graphene ρ (g/cm ²)	7.6×10^{-8}
Sound velocity v_s (cm/s)	2×10^6
Optical phonon energy $\hbar \omega_{op}$ (meV)	164
Intervalley acoustic phonon energy $\hbar \omega_{ac}$ (meV)	124

Table 2.2 lists the parameters utilized for calculating the scattering rates in Eqs. 2.29 and 2.30.

2.4.4 Ensemble Drift Velocity

The Ensemble Monte Carlo simulation is performed on N carriers using the previously described approach, incorporating periodic boundary conditions (except at the edges in systems where edge states are considered). To calculate the drift velocity of carriers in the x -direction (the direction in which the external field is applied) at the l -th graphene layer, an ensemble average of the x -components of the individual carrier velocities is taken. Each carrier moves with the Fermi velocity v_f , and α_j denotes the angle of its velocity direction measured from the x -axis. Thus, $v_f \cos \alpha_j$ is the x -component of the j -th carrier's velocity, and the drift velocity for electrons in layer l is

$$v_{x,l,e} = \frac{1}{N} \sum_{j=1}^N v_f \cos \alpha_j. \quad (2.31)$$

The same ensemble averaging procedure is applied to holes to obtain $v_{x,l,h}$, using the corresponding angles of hole velocity directions. The net drift velocity in the x -direction for the l -th layer, $v_{x,l}$, is then the weighted average of the electron and hole contributions. Here, n_l and p_l are the sheet densities of electrons and holes in layer l , respectively. The total drift velocity for the l -th layer is then obtained by

$$v_{x,l} = \frac{n_l v_{x,l,e} - p_l v_{x,l,h}}{n_l + p_l}. \quad (2.32)$$

2.5 Summary

This chapter outlined the simulation framework used to study carrier transport in multilayer graphene systems. The computational approach was divided into two main parts: the calculation of the electrostatic potential and the simulation of carrier transport.

In Sec. 2.2, the structural model used in this study is introduced. It consists of a SiO₂ substrate containing randomly distributed donor impurities, stacked graphene layers placed on top of it, and an insulating layer covering the graphene. Section 2.3 describes the procedure for obtaining the electrostatic potential profile, which involves solving Poisson equation self-consistently in conjunction with the Thomas-Fermi approximation. This iterative process updates both the potential and the induced carrier densities until the system converges. A specific impurity-mirroring

technique is employed to generate a periodic impurity pattern, ensuring consistency with the periodic boundary conditions applied in the transport simulation. The Cloud-in-Cell method is used to assign the impurity charges to the computational grid.

The second phase of the simulation is described in Sec. 2.4, which explains the transport calculation using the Ensemble Monte Carlo method. This approach simulates the trajectories of a large ensemble of carriers moving under the influence of both the impurity-induced electric fields and an externally applied field. Carrier dynamics are modeled as a sequence of free flights interrupted by scattering events. The framework explicitly accounts for intrinsic scattering mechanisms, including acoustic phonon scattering and optical phonon scattering. Finally, the transport properties of the system are obtained by computing the ensemble average of the individual carrier velocities, yielding the drift velocity for each graphene layer.

Overall, this chapter establishes the computational foundations used throughout this thesis and provides the methodological basis for the analyses presented in the subsequent chapters.

Chapter 3

Carrier Transport Simulation in Turbostratic Graphene

3.1 Introduction

As it has been briefly explained in Chapter 1, the transport properties of monolayer graphene are highly sensitive to external perturbations arising from its surrounding environment [23, 24]. On substrates such as SiO_2 , randomly distributed charged impurities generate electrostatic potential fluctuations. These fluctuations locally shift the charge neutrality point in graphene, resulting in spatially varying electron-rich and hole-rich regions known as electron puddles and hole puddles [39, 59, 60]. Accordingly, this perturbative potential landscape is recognized as a major factor for the reduction of mobility in graphene well below its intrinsic limit [22, 34, 40, 61–63].

A promising route to alleviating this impurity-induced problem and achieving mobility enhancement is the use of multilayer graphene structures with a turbostratic configuration. Figure 3.1 illustrates the schematic configuration of this structure. The attractiveness of the turbostratic arrangement lies in the fact that adjacent graphene layers are rotated by a relatively large angle, typically around 30° , resulting in a decoupling of graphene layers. Hence, due to this rotational misalignment, each layer preserves the linear Dirac dispersion characteristic of pristine monolayer graphene, which essentially enables each layer to behave as monolayer graphene when the carrier transport is considered [51, 64–66]. Thus, carriers maintain a high Fermi velocity and excellent intrinsic transport characteristics. Beyond preserving the desirable monolayer-like electronic properties, the multilayer structure offers an additional advantage, enhanced screening of impurity-induced potential. Lower graphene layers in the stack shield the upper layers from the fluctuating

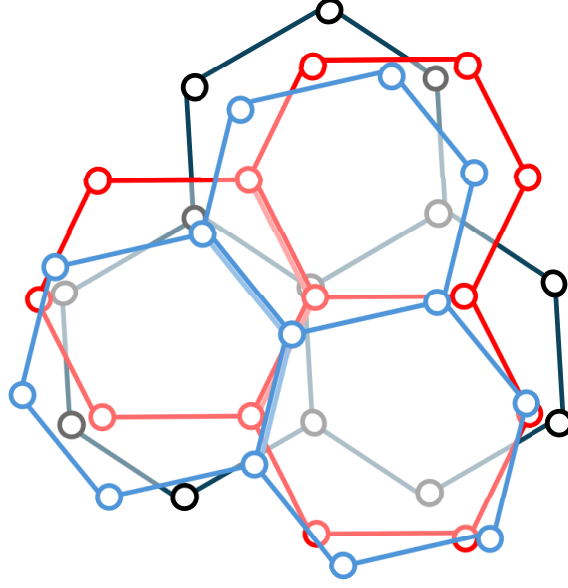


Figure 3.1: Schematic illustration of a turbostratic multilayer graphene structure, in which each graphene sheet is stacked with random rotational angles with respect to one another.

electrostatic potential generated by charged impurities on the substrate surface. As a result, it is expected that the potential inhomogeneity experienced by carriers diminishes rapidly as the distance from the substrate increases. Previous studies have shown that graphene layers can change the potential landscape and cause a redistribution of charges among the layers, which leads to a smoother landscape [48, 67–70]. Thus, turbostratic multilayer graphene provides a structural pathway to suppress extrinsic disorder, and retain the intrinsic transport characteristics.

The objective of this chapter is to investigate the effectiveness of this multilayer approach by examining how turbostratic graphene layers influence potential fluctuations in each graphene layer, and ultimately, affect the carrier mobility. The details of the model and methodology were explained in the preceding chapter, and the following section presents the numerical results and their physical interpretation.

3.2 Results and Discussion

3.2.1 Potential Profile

The general configurations of the multilayer structures have been explained in Section 2.2 of Chapter 2. The specific configurations considered in this section are as follows. The calculations are performed for systems with different numbers of layers

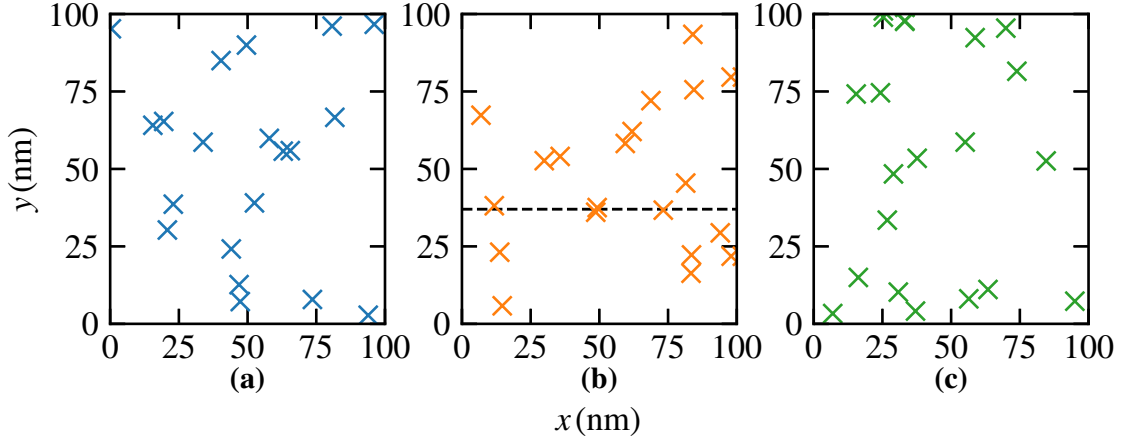


Figure 3.2: Three configurations of randomly distributed impurities on the substrate, comprising $N_{\text{imp}} = 20$ impurities (corresponding to a density of $N_D = 2 \times 10^{11} \text{ cm}^{-2}$), are considered. The cross-sectional potential along the dashed line in pattern (b) is shown in Fig. 3.3.

to investigate the changes in the potential profile with the addition of further layers in the system. Multilayer graphene systems with $M = 2, 4, 8, 12$, and 16 are analyzed. The electrostatic potential is calculated using a mesh size of $\delta = 1 \text{ nm}$ in the xy plane and 0.34 nm in the z direction, corresponding to the interlayer spacing d_{gg} . The potential profiles for three different impurity densities of $N_D = 2 \times 10^{11} \text{ cm}^{-2}$, $5 \times 10^{11} \text{ cm}^{-2}$, and $1 \times 10^{12} \text{ cm}^{-2}$ (corresponding to $N_{\text{imp}} = 20, 50$, and 100) are determined for systems with $M = 16$. In order to minimize the impact of a specific impurity distribution on the transport properties, the potential profiles for ten different impurity patterns are calculated. Figure 3.2 illustrates three samples of impurity patterns, corresponding to a density of $N_D = 2 \times 10^{11} \text{ cm}^{-2}$ used in this study. The cross marks indicate the position of the impurities on the substrate. There are no correlations between the positions of the impurities, and each position is determined independently and randomly on the substrate.

Figure 3.3 illustrates a cross-section of the potential profile at $y = 37 \text{ nm}$, indicated by the dashed line in pattern (b) of Fig. 3.2. The figure shows that the peaks of potential fluctuations coincide with the impurity positions; however, their magnitude varies significantly across the graphene layers. In the 1st layer (black line), pronounced peaks appear. For example, at $x = 49 \text{ nm}$, a sharp potential maximum of approximately 0.28 V is observed. In contrast, the 4th layer potential (blue dashed line) decreases to about 0.07 V , while the 6th and 8th layers show further reductions, with approximate potential values of 0.03 V and 0.01 V , respectively. Finally, the 16th layer becomes nearly flat, exhibiting minimal fluctuation. This

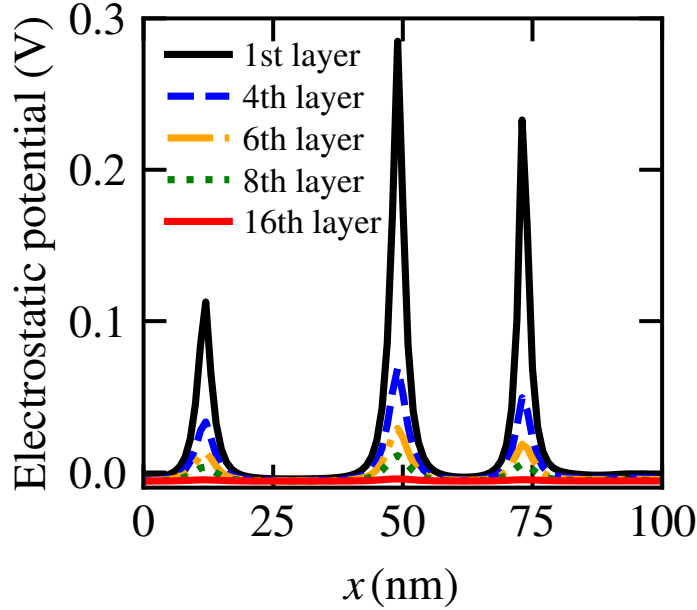


Figure 3.3: Cross-sectional electrostatic potential along the dashed line in impurity pattern (b) of Fig. 3.2, calculated for a 16-layer graphene structure with an impurity density of $N_D = 2 \times 10^{11} \text{ cm}^{-2}$. The trend here indicates that the potential profile becomes noticeably smoother in the upper layers.

demonstrates that the amplitude of the potential curves diminishes substantially as the layer index increases.

Figure 3.4 presents a color map of the potential profiles of the 1st, 4th, 8th and 16th graphene layers in the multilayer graphene system corresponding to the impurity pattern (b) shown in Fig. 3.2. The bright orange regions indicate areas of high potential. By comparing the 1st layer profile with the impurity positions in Fig. 3.2 (b), it is observed that locations near impurities correspond to prominent potential peaks, whereas the rest of the graphene sheet exhibits only minor fluctuations. As the layer index increases from the 1st to the 16th layer, the orange spots gradually fade, indicating a progressive reduction of potential variations. In the uppermost layer, there is no significant difference between the regions near the impurities and the remainder of the graphene sheet, indicating an overall flattening of the potential profile.

Figure 3.5 presents how the standard deviation σ_l of the electrostatic potential evolves across the graphene layers for systems with $M = 16$ and different impurity densities. The quantity σ_l characterizes the spatial fluctuation of the potential

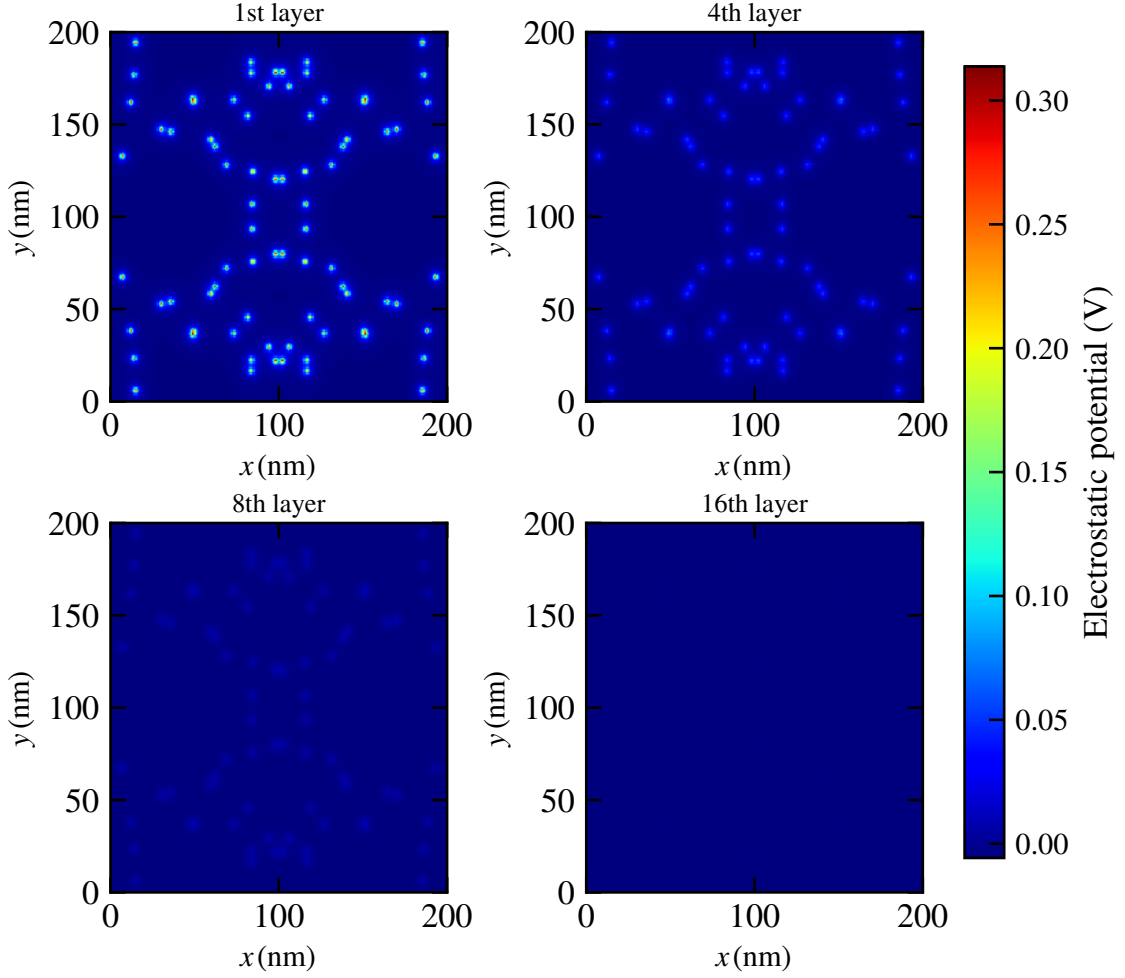


Figure 3.4: Electrostatic potential profile for the 1st, 4th, 8th, and 16th layers for a system with an impurity density of $N_D = 2 \times 10^{11} \text{ cm}^{-2}$ and impurity pattern (b) in Fig. 3.2. Bright spots indicate regions with higher potential peaks. It is seen that the magnitude of the potential peaks decreases with increasing layer index.

within each graphene sheet and is defined as

$$\sigma_l = \left[\frac{1}{N_m} \sum_{j=1}^{N_m} (\phi_{jl} - \bar{\phi}_l)^2 \right]^{1/2}, \quad (3.1)$$

where ϕ_{jl} denotes the potential at grid point j in the l -th graphene layer, $\bar{\phi}_l$ is the average value of the potential in the layer, and N_m is the number of grid points in the layer.

Large σ_l values in the lower layers indicate strong spatial inhomogeneity of the potential, where sharp local peaks appear near charged impurities and dominate the local electrostatic landscape. As the distance from the substrate increases, these

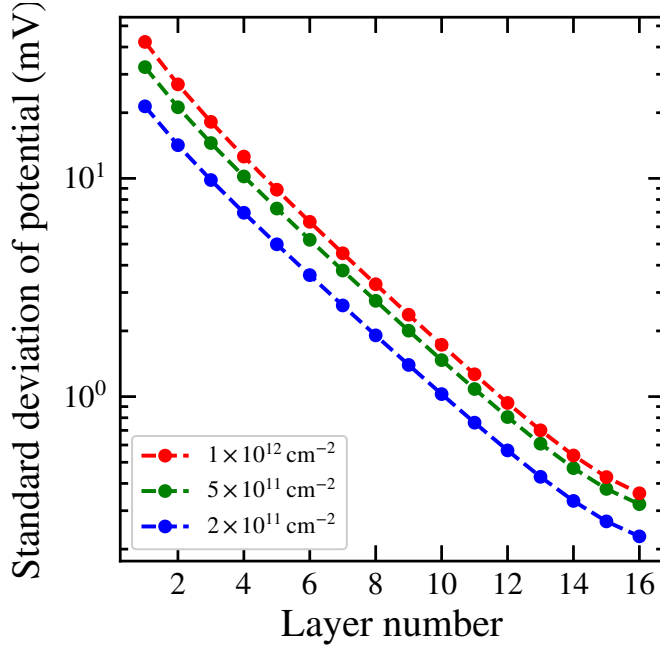


Figure 3.5: Standard deviation of the electrostatic potential for a 16-layer graphene system at different impurity densities. Here, the amplitude of potential fluctuations within each graphene sheet is shown. The lower layers exhibit large fluctuation magnitudes due to strong impurity-induced variations, whereas the upper layers show nearly vanishing fluctuations. A rapid decrease in fluctuation amplitude with increasing layer index is observed for all impurity densities.

variations diminish rapidly. For example, in the system with an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$, the standard deviation decreases by approximately 67% in the 4th layer and by more than 90% in the 8th layer compared to the 1st layer. By the 16th layer, the fluctuation amplitude is reduced by about 99%, suggesting that the potential profile in the uppermost layer has become almost uniform.

This exponential decay of σ_l is consistently observed even when the impurity density increases. Systems containing a larger number of impurities exhibit stronger potential fluctuations within each individual layer. However, the rate of reduction in standard deviation with layer number remains nearly unchanged, and the potential variation in the upper layers becomes negligible regardless of impurity concentration. This rapid reduction can be attributed to the large accumulation of electrons near the charged impurity regions. These electron concentrations effectively screen the electrostatic potential induced by the impurities, which weakens exponentially with distance, resulting in an increasingly smoother potential profile in the upper graphene layers.

3.2.2 Transport Properties

The Monte Carlo simulation is performed for 10^4 carriers, with $t_s = 1$ fs and $t_{\text{fin}} = 1$ ns. The averaged drift velocity of each graphene layer for a system with $M = 16$ and impurity density of $2 \times 10^{11} \text{ cm}^{-2}$ under different external fields is depicted in Fig. 3.6. The results are averaged for 10 impurity patterns with Eq. 2.32. Here it is seen that the lower layers closer to the substrate have the lower drift velocity, and as the layers get farther from the substrate, the velocity improves significantly, eventually approaching the velocity of a system with no impurity (shown by the solid line). Additionally, for each layer, as the magnitude of the external field increases, the velocity also increases, but at higher fields the rate of increase diminishes due to phonon emission. This increase in the velocity is more prominent for the layers farther from the substrate, where the impact of the impurity scattering is reduced due to screening. These velocity results are compatible with the potential results, shown in the last section, where the upper layers had smaller potential fluctuations compared to the lower layers, and hence the carriers in the upper layers face fewer obstacles from the impurity potential while moving through the graphene. Here, the velocity of carriers in the 16th graphene layer becomes almost identical to the phonon-limited drift velocity of electrons in an impurity-free system, indicating that the impact of impurity scattering becomes so inconsequential that electrons feel no hindrance from the impurities.

Figure 3.7 (a) presents the carrier mobility of each layer for multilayer graphene systems with 2, 4, 8, 12, and 16 layers under an electric field of 10^2 V/cm and in the presence of an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$. The first observation is that the lower layers exhibit nearly identical mobilities across all systems, implying that the impurity influence dominates similarly across the systems in that region. However, clear differences in mobility appear in the upper layers. For instance, the 8th layer in the $M = 8$ system shows a lower mobility value than the same layer in the $M = 12$ system, and a similar relationship holds between the 12th layers of the $M = 12$ and $M = 16$ systems. This is because the screening of the potential does not occur only in the lower layers, but the upper layers also contribute to the screening to some extent. Thus, the additional upper layers further enhance the screening, and reduce the impact of impurities. The mobility of the 12th layer in a system with $M = 16$ improves simply due to the presence of four more layers above it. However, beyond approximately 16 layers, this improvement saturates, and the upper-layer mobility approaches the phonon-limited value, meaning that the impurity potential is almost entirely screened out by the lower layers. This trend is also inferred from

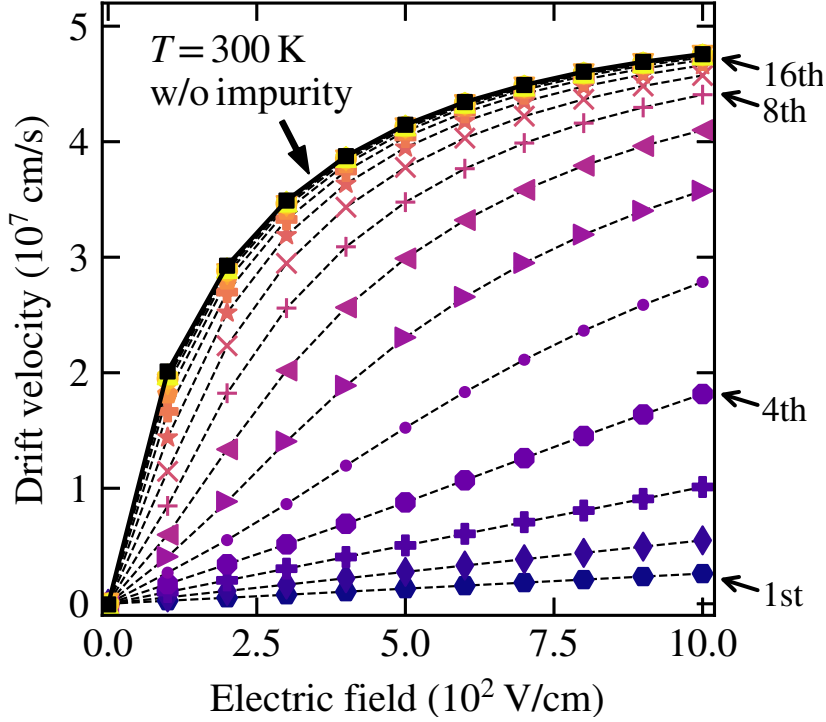


Figure 3.6: Averaged drift velocities of each graphene layer in a 16-layer system with an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$ under various electric fields. Lower layers exhibit reduced velocities due to strong impurity scattering, while upper layers show progressively higher velocities as the impurity potential becomes more effectively screened. As a result, the 16th layer approaches the phonon-limited drift velocity of an impurity-free system. The increase in velocity with electric field is most pronounced in the upper layers, where phonon emission eventually limits further increase.

the curvature of the mobility profile of the system with $M = 16$, where a rapid increase in mobility is followed by saturation. More specifically, it is observed that around the 7th to 9th layers in the systems, a pronounced boost in mobility is achieved with the addition of just one more layer. However, in the upper layers, this rapid growth slows down.

Figure 3.7 (b) shows the drift velocity profiles of carriers under an applied field of 10^3 V/cm and an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$ for systems with 2, 4, 8, 12, and 16 layers. Here, the overall trend resembles that observed for the mobility in Fig. 3.7 (a). The screening effect originating from the lower layers weakens the influence of impurities in the upper layers, resulting in an improved drift velocity in the upper layers. However, at this higher applied field, the increase in drift velocity becomes more pronounced in the layers near the substrate. This behavior arises because carrier transport is governed by two fields: the externally applied electric field and

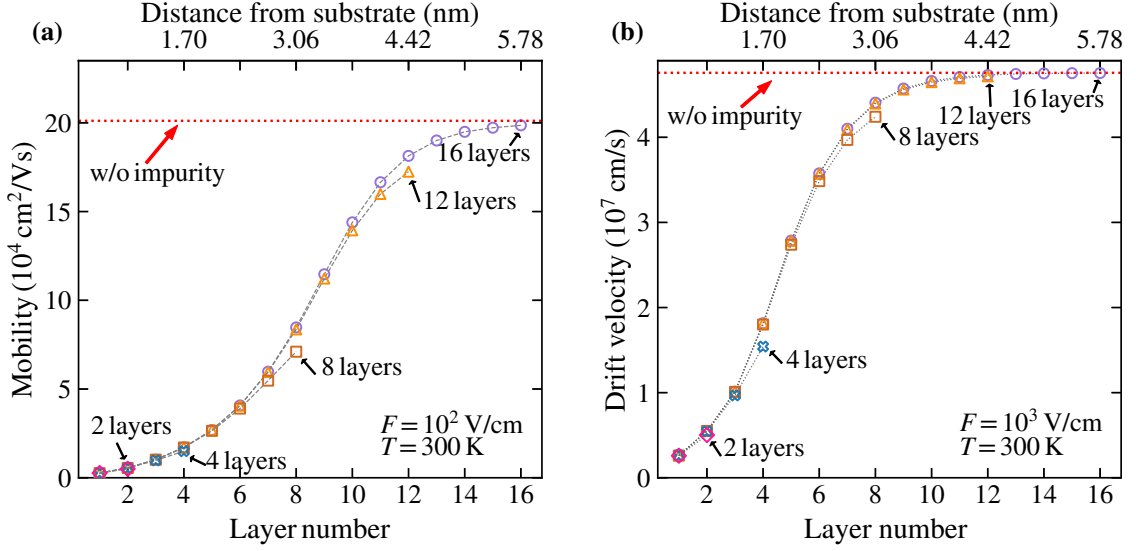


Figure 3.7: (a) Carrier mobility for each layer in multilayer graphene systems with $M = 2, 4, 8, 12$, and 16 under an electric field of 10^2 V/cm . As the number of layers increases, the mobility in the upper part of the stack improves due to a smoother potential landscape, and this enhancement gradually saturates as it approaches the phonon-limited value. (b) Drift velocity for each layer under an applied field of 10^3 V/cm for the same systems. A similar trend of improvement followed by saturation is observed, with the higher field causing a steeper rise in the velocity before it reaches its saturated value.

the local field induced by the charged impurities. When the applied field is large, the external field rapidly outweighs the impurity-induced field, thereby reducing the relative impact of impurities more effectively than in the lower-field case shown in Fig. 3.7 (a). Consequently, as illustrated in Fig. 3.7 (b), the drift velocity exhibits a steep rise followed by saturation, ultimately approaching the velocity observed in the impurity-free system.

Figure 3.8 (a) shows the carrier mobility of each layer in a 16-layer multilayer graphene system under an electric field of 10^2 V/cm for impurity densities of $2 \times 10^{11} \text{ cm}^{-2}$, $5 \times 10^{11} \text{ cm}^{-2}$, and $1 \times 10^{12} \text{ cm}^{-2}$. Here, it is observed that for each layer, the system with the higher impurity density has the lower carrier mobility. Nevertheless, there is a mobility enhancement for all systems, irrespective of their impurity density, in the higher layers. Additionally, the mobility differences between the different impurity densities are quite small up to the third graphene layer, and the difference increases in the middle layers of the systems in the range of the 7th to 9th layers, and eventually the mobilities converge to the phonon-limited mobilities in the upper layers. In other words, for the system with a lower impurity density, the rise in the mobility of graphene layers occurs in the lower layers and the eventual

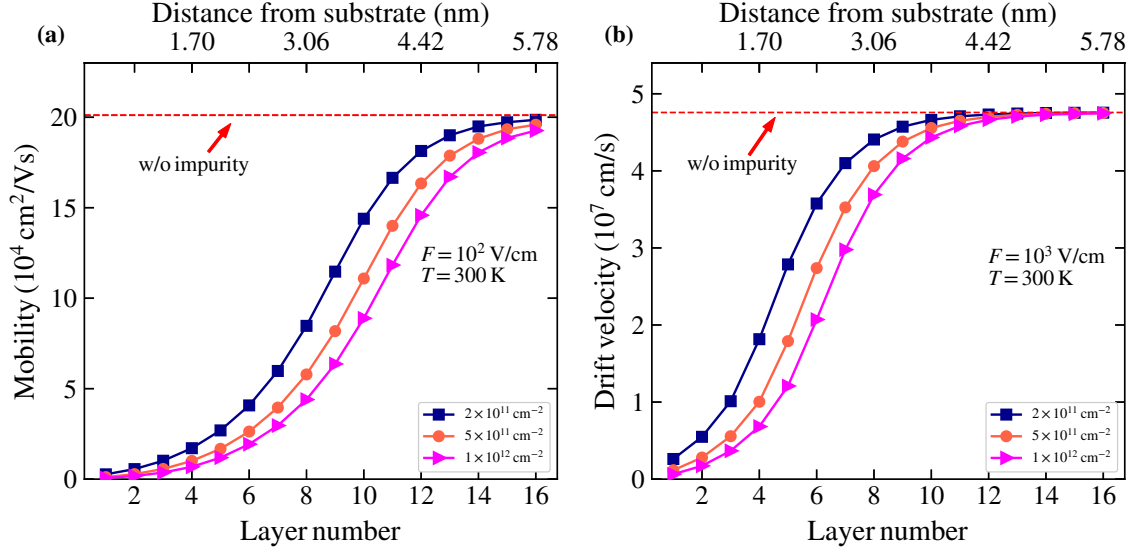


Figure 3.8: (a) Carrier mobility for each layer in a 16-layer graphene system under an electric field of 10^2 V/cm for impurity densities of 2×10^{11} , 5×10^{11} , and $1 \times 10^{12} \text{ cm}^{-2}$. Higher impurity densities yield lower mobilities, yet all profiles exhibit mobility enhancement in the upper layers and eventual convergence toward the phonon-limited value. (b) Drift velocity for each layer under an applied field of 10^3 V/cm for the same impurity densities. The overall behavior mirrors the mobility trends, though the stronger field induces a sharper rise in velocity and an earlier convergence to the phonon-limited regime.

convergence also happens earlier, and this trend becomes delayed (the rise and convergence occur in higher layers) for the systems with higher impurity densities.

Figure 3.8 (b) illustrates the drift velocity trends for the same systems as Figure 3.8 (a). Here, the general trends of the velocity curves are the same as the mobility curves. However, due to the application of a higher external field, the rise in the velocities is more pronounced, and the convergence to the phonon-limited velocity also happens in the lower layers compared to the mobility case.

Figure 3.9 (a) illustrates the total mobilities of systems with 2, 4, 8, 12, and 16 layers under an electric field of 10^2 V/cm and an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$. These mobilities are derived using a method similar to Eq. 2.32, where the average is taken over all layers comprising the multilayer graphene system and weighted by their carrier densities across the entire system. As observed, the addition of more layers to the system contributes positively to the total mobility of the system. It is important to note that the carriers are disproportionately induced in the graphene layers; consequently, the average total mobility of the system becomes lower than the highest mobility observed in the upper layers, as seen in the case of 16-layer graphene. Figure 3.9 (b) shows the total velocity for the same systems as Fig. 3.9

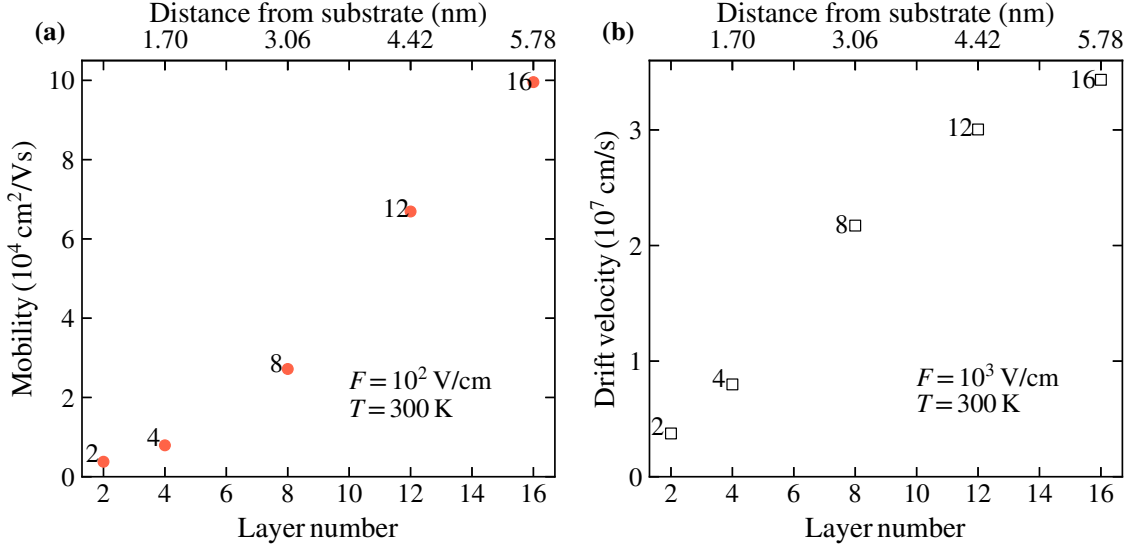


Figure 3.9: (a) Total mobility of multilayer graphene systems with 2, 4, 8, 12, and 16 layers under an electric field of 10^2 V/cm and an impurity density of $2 \times 10^{11} \text{ cm}^{-2}$. The total mobility is obtained by averaging over all layers comprising the multilayer graphene system and weighting them by their carrier densities across the entire system. (b) Total drift velocity for the same systems under an electric field of 10^3 V/cm . The velocity trends mirror those in part (a), showing an enhancement as more layers are added, although a clear disparity remains between the averaged velocity and the highest velocities occurring in the upper layers.

(a). The velocity trends here are the same as those of the mobilities, with an enhancement of the total velocity as more layers are added, although a noticeable gap remains between the averaged velocity and the highest layer-specific velocity in each multilayer scheme.

3.3 Analytical Model of Mobility for Turbostratic Graphene

3.3.1 Potential Fluctuation

Here, an analytical model for the mobility of a multilayer graphene system is considered. N_{imp} impurities are assumed to be randomly distributed over a plane of area S . If the in-plane position of the i -th impurity is $\mathbf{r}_i = (x_i, y_i)$, the areal density of impurities is written as [71]

$$\rho(\mathbf{r}) = \sum_{i=1}^{N_{\text{imp}}} \delta(\mathbf{r} - \mathbf{r}_i). \quad (3.2)$$

Taking the Fourier transform gives

$$\rho(\mathbf{q}) = \int \rho(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} = \int \sum_{i=1}^{N_{\text{imp}}} \delta(\mathbf{r} - \mathbf{r}_i) e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} = \sum_{i=1}^{N_{\text{imp}}} e^{-i\mathbf{q}\cdot\mathbf{r}_i}. \quad (3.3)$$

The electrostatic potential induced by these impurities at a plane with a distance of d from the impurity plane is considered. The potential is obtained by

$$V(\mathbf{r}) = \int \frac{e^2}{4\pi\epsilon\sqrt{|\mathbf{r} - \mathbf{r}'|^2 + d^2}} \rho(\mathbf{r}') d\mathbf{r}', \quad (3.4)$$

whose Fourier transform reads

$$\begin{aligned} V(\mathbf{q}) &= \int V(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} = \iint \frac{e^2}{4\pi\epsilon\sqrt{|\mathbf{r} - \mathbf{r}'|^2 + d^2}} e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} \rho(\mathbf{r}') d\mathbf{r}' \\ &= \int \frac{e^2}{4\pi\epsilon\sqrt{|\mathbf{r}''|^2 + d^2}} e^{-i\mathbf{q}\cdot\mathbf{r}''} d\mathbf{r}'' \int \rho(\mathbf{r}') e^{-i\mathbf{q}\cdot\mathbf{r}'} d\mathbf{r}' = v(q) \rho(\mathbf{q}). \end{aligned} \quad (3.5)$$

Here, $v(q)$ is the Fourier transform of the potential due to a single impurity, given by

$$v(q) = \int v(\mathbf{r}) e^{-i\mathbf{q}\cdot\mathbf{r}} d\mathbf{r} = \int v(r) e^{-iqr \cos \phi} r dr d\phi = \int 2\pi v(r) J_0(qr) r dr. \quad (3.6)$$

$J_0(x)$ denotes the zeroth-order Bessel function of the first kind

$$J_0(x) = \frac{1}{2\pi} \int_0^{2\pi} e^{ix \cos \phi} d\phi, \quad J_0(-x) = J_0(x), \quad (3.7)$$

which gives

$$2\pi J_0(qr) = \int_0^{2\pi} e^{-iqr \cos \phi} d\phi. \quad (3.8)$$

Hence, Eq. 3.6 becomes

$$\begin{aligned} v(q) &= 2\pi \int \frac{e^2}{4\pi\epsilon\sqrt{r^2 + d^2}} J_0(qr) r dr \\ &= \frac{e^2}{2\epsilon} \int \frac{J_0(qr) r}{\sqrt{r^2 + d^2}} dr \\ &= \frac{e^2}{2\epsilon q} e^{-qd}. \end{aligned} \quad (3.9)$$

To account for screening effects, the Fourier transform of the potential is modified

by the screening constant q_s as

$$v(q) = \frac{e^2}{2\epsilon(q + q_s)} e^{-qd}. \quad (3.10)$$

Here, q_s is given by [72]

$$\begin{aligned} q_s &= \frac{e^2}{2\epsilon} \int \left(-\frac{df(E)}{dE} \right) g(E) dE \\ &= \frac{e^2}{2\epsilon} \frac{n_e}{k_B T}, \end{aligned} \quad (3.11)$$

where n_e is the electron density.

The mean square of the potential fluctuations is given by [34]

$$\begin{aligned} \langle \delta V_{\text{imp}}^2 \rangle &= \langle [V(\mathbf{r}) - \langle V(\mathbf{r}) \rangle]^2 \rangle = \langle V(\mathbf{r})^2 \rangle - \langle V(\mathbf{r}) \rangle^2 \\ &= n_{\text{imp}} \int \frac{d\mathbf{q}}{(2\pi)^2} v(q)^2, \end{aligned} \quad (3.12)$$

where $n_{\text{imp}} = N_{\text{imp}}/S$ is the impurity density. This integral can be evaluated analytically to yield

$$\begin{aligned} \langle \delta V_{\text{imp}}^2 \rangle &= n_{\text{imp}} \int \frac{d\mathbf{q}}{(2\pi)^2} \left[\frac{e^2}{2\epsilon(q + q_s)} e^{-qd} \right]^2 = n_{\text{imp}} \left(\frac{e^2}{2\epsilon} \right)^2 \int \frac{q dq}{2\pi} \frac{e^{-2qd}}{(q + q_s)^2} \\ &= \frac{1}{2\pi} n_{\text{imp}} \left(\frac{e^2}{2\epsilon} \right)^2 C(q_s d), \end{aligned} \quad (3.13)$$

where the function C is defined by

$$C(x) = (1 + 2x) e^{2x} E_1(2x) - 1, \quad (3.14)$$

and E_1 denotes the exponential integral

$$E_1(z) = \int_z^\infty \frac{e^{-t}}{t} dt. \quad (3.15)$$

3.3.2 Mobility of Monolayer Graphene

The conductivity of electrons, in terms of the momentum relaxation time, is written as [72, 73]

$$\sigma = \frac{e^2}{\pi \hbar^2} \langle \tau \rangle \int f(E) dE, \quad (3.16)$$

where

$$\langle \tau \rangle = \frac{\int E \tau(E) \left(-\frac{df}{dE} \right) dE}{\int E \left(-\frac{df}{dE} \right) dE}. \quad (3.17)$$

Here, $f(E)$ represents the electron distribution function. Consequently, the density of states is given by

$$g(E) = \frac{g_s g_v E}{2\pi \hbar^2 v_f^2} = \frac{g_s g_v E}{2\pi \gamma^2}, \quad (3.18)$$

where $g_s = 2$ is the spin degeneracy, $g_v = 2$ is the valley degeneracy and $\gamma = \hbar v_f$. The electron density is then given by

$$n = \int g(E) f(E) dE = \frac{g_s g_v}{2\pi \gamma^2} \int E f(E) dE. \quad (3.19)$$

Hence, the mobility can be expressed as

$$\mu = \frac{\sigma}{ne} = \frac{ev_f^2}{2} \frac{\int E \tau(E) \left(-\frac{df}{dE} \right) dE}{\int E \left(-\frac{df}{dE} \right) dE} \frac{\int f(E) dE}{\int E f(E) dE}. \quad (3.20)$$

Approximating $f(E)$ with the Boltzmann distribution, and $\beta = 1/k_B T$, the following expression is obtained

$$\begin{aligned} \mu = \frac{\sigma}{ne} &= \frac{ev_f^2}{2} \frac{\int E \tau(E) e^{-\beta E} dE}{\int E e^{-\beta E} dE} \frac{\int e^{-\beta E} dE}{\int E e^{-\beta E} dE} \\ &= \frac{ev_f^2}{2k_B T} \int \beta E \tau(E) e^{-\beta E} d\beta E = \frac{ev_f^2}{2k_B T} \int \varepsilon \tau(k_B T \varepsilon) e^{-\varepsilon} d\varepsilon, \end{aligned} \quad (3.21)$$

where $\varepsilon = \beta E$.

The momentum relaxation time $\tau(E)$ due to impurity scattering of electrons is given by [72]

$$\frac{\hbar}{\tau(E)} = 2\pi n_{\text{imp}} \frac{E}{2\pi \gamma^2} \int_0^\pi \frac{d\theta}{\pi} \frac{1}{2} (1 - \cos^2 \theta) v(q)^2, \quad (3.22)$$

where

$$q = \frac{2E}{\gamma} \sin(\theta/2) = 2k \sin(\theta/2), \quad (3.23)$$

and $E = \gamma k$.

Here, the potential obtained in Eq. 3.10 is used in Eq. 3.22, hence, the momentum

relaxation time is expressed as:

$$\begin{aligned}
 \frac{\hbar}{\tau(E)} &= 2\pi n_{\text{imp}} \frac{E}{2\pi\gamma^2} \int_0^\pi \frac{d\theta}{\pi} \frac{1}{2} (1 - \cos^2 \theta) \left(\frac{e^2}{2\epsilon(q + q_s)} \right)^2 e^{-2qd} \\
 &= n_{\text{imp}} \left(\frac{e^2}{2\epsilon} \right)^2 \frac{1}{E} \int_0^\pi \frac{d\theta}{2\pi} (1 - \cos^2 \theta) \frac{e^{-4kd \sin(\theta/2)}}{(2 \sin(\theta/2) + q_s/k)^2} \\
 &= \frac{1}{2} n_{\text{imp}} \left(\frac{e^2}{2\epsilon} \right)^2 \frac{1}{EG(q_s d, kd)},
 \end{aligned} \tag{3.24}$$

where $G(x, y)$ is defined by

$$\frac{1}{G(x, y)} = \int_0^\pi \frac{d\theta}{\pi} (1 - \cos^2 \theta) \frac{e^{-4y \sin(\theta/2)}}{(2 \sin(\theta/2) + (x/y))^2}. \tag{3.25}$$

Therefore, using Eq. 3.21 and Eq. 3.24, the mobility is given by

$$\mu = \left(\frac{2\epsilon}{e^2} \right)^2 \frac{\hbar e v_f^2}{n_{\text{imp}}} \int \varepsilon^2 G(q_s d, kd) e^{-\varepsilon} d\varepsilon. \tag{3.26}$$

Since $k = \varepsilon k_B T / \gamma$, the following expression is obtained

$$\begin{aligned}
 \mu &= \left(\frac{2\epsilon}{e^2} \right)^2 \frac{\hbar e v_f^2}{n_{\text{imp}}} \int \varepsilon^2 G(q_s d, \varepsilon k_B T d / \gamma) e^{-\varepsilon} d\varepsilon \\
 &= \left(\frac{2\epsilon}{e^2} \right)^2 \frac{\hbar e v_f^2}{n_{\text{imp}}} H(q_s d, k_B T d / \gamma),
 \end{aligned} \tag{3.27}$$

where $H(x, y)$ is defined by

$$H(x, y) = \int \varepsilon^2 G(x, y\varepsilon) e^{-\varepsilon} d\varepsilon. \tag{3.28}$$

Figure 3.10 illustrates the mobility in terms of the screening constant q_s , calculated using Eq. 3.27, for three different impurity densities of $n_{\text{imp}} = 2.0 \times 10^{11}$, 5.0×10^{11} , and $1.0 \times 10^{12} \text{ cm}^{-2}$. The dashed lines show the MC results for the mobility of the first layer of a 16-layer graphene system, corresponding to each of these impurity densities. It is seen that the MC simulation values are consistent for $q_s \lesssim 10^6 \text{ m}^{-1}$. It is noteworthy that this result does not imply that the screening is weak, but rather demonstrates the semiclassical limit of MC simulation. Therefore, the classical limit is considered as $k \gg q_s$, where the electron wavelength is much shorter than the length scale in which the potential changes. In this case, $G(q_s d, kd)$ can be approximated by $G(0, kd)$, and $G(0, y)$ can be analytically expressed using

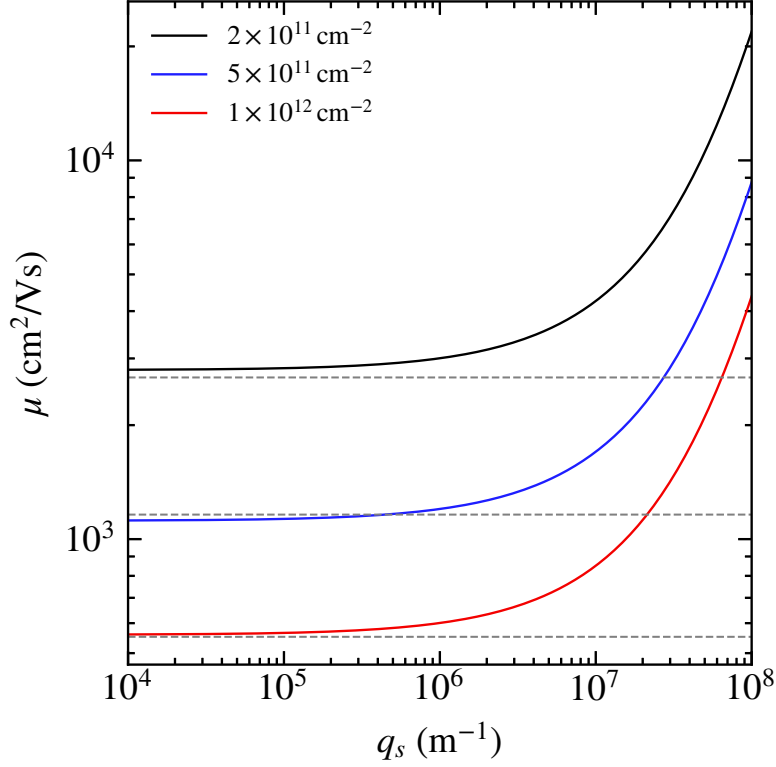


Figure 3.10: Mobility as a function of the screening constant q_s for three impurity densities ($n_{\text{imp}} = 2.0 \times 10^{11}$, 5.0×10^{11} , and $1.0 \times 10^{12} \text{ cm}^{-2}$), calculated using Eq. 3.27. The dashed lines indicate the MC results for the first layer of a 16-layer graphene system.

the modified Bessel function of the first kind $I_n(z)$, and the modified Struve function $L_n(z)$ as

$$\frac{1}{G(0, y)} = \frac{I_1(4y) - L_1(4y)}{4y}. \quad (3.29)$$

Since $k_B T d / \gamma \ll 1$, $G(0, y)$ in Eq. 3.28 is given by

$$G(0, y) = 2 + \frac{32}{3\pi} y + \dots \quad (3.30)$$

and hence, the approximate expression for the mobility is obtained as

$$\mu = \frac{\hbar e v_f^2}{n_{\text{imp}}} \left(\frac{2\epsilon}{e^2} \right)^2 \left(4 + \frac{64}{\pi} \frac{k_B T d}{\gamma} + \dots \right). \quad (3.31)$$

3.3.3 Mobility of Multilayer Graphene

Here, it is assumed that there are $\ell - 1$ screening layers between the electron transport layer and the impurity plane. Due to the screening, the potential at the transport plane is reduced by a factor η_ℓ , such that

$$v(q) = \eta_\ell \frac{e^2}{2\epsilon(q + q_s)} e^{-qd}. \quad (3.32)$$

Using this screened potential and proceeding as in the aforementioned derivation, the lowest-order mobility from Eq. 3.31 becomes

$$\mu = 4\eta_\ell^{-2} \frac{\hbar e v_f^2}{n_{\text{imp}}} \left(\frac{2\epsilon}{e^2} \right)^2. \quad (3.33)$$

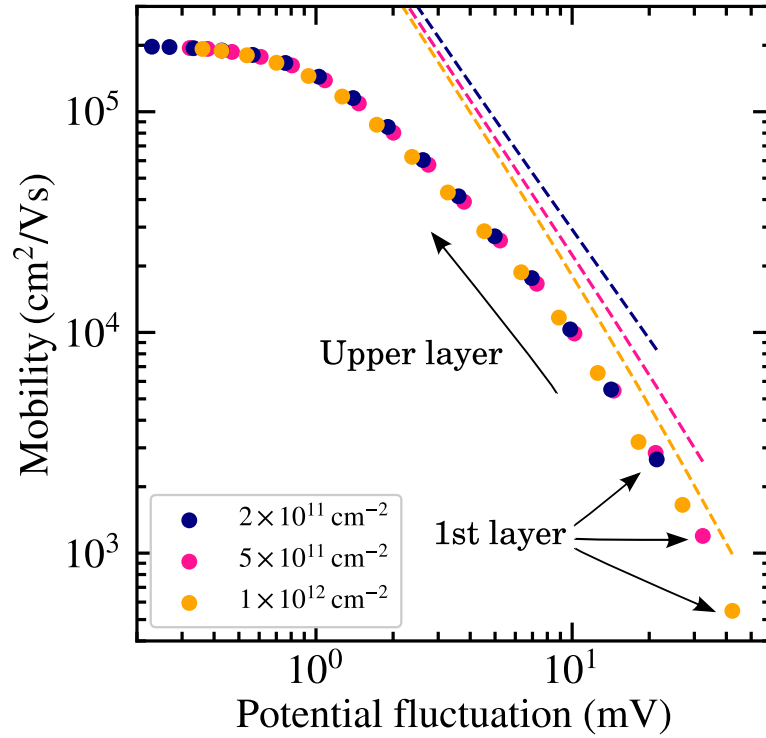


Figure 3.11: Carrier mobility versus potential fluctuations for each graphene layer in the $M = 16$ systems with impurity densities of $2 \times 10^{11} \text{ cm}^{-2}$, $5 \times 10^{11} \text{ cm}^{-2}$, and $1 \times 10^{12} \text{ cm}^{-2}$. Mobility improves with decreasing potential fluctuation. Large fluctuations near the substrate result in strong mobility degradation, while fluctuations below 1 mV result in the mobility approaching the phonon-limited transport value. The dashed lines show the analytical mobility of Eq. 3.35, in agreement with the Monte Carlo results.

The variance of the potential fluctuation is

$$\langle \delta V_{\text{imp}}^2 \rangle = \frac{1}{2\pi} \eta_{\ell}^2 n_{\text{imp}} \left(\frac{e^2}{2\epsilon} \right)^2 C(q_s d), \quad (3.34)$$

where $C(q_s d)$ is the same function defined earlier. Hence, expressing the mobility in terms of the potential fluctuation gives

$$\mu = \frac{2}{\pi} \frac{\hbar e v_{\text{f}}^2}{\langle \delta V_{\text{imp}}^2 \rangle} C(q_s d), \quad (3.35)$$

indicating that μ is inversely proportional to $\langle \delta V_{\text{imp}}^2 \rangle$.

Figure 3.11 shows the dependence of the carrier mobility on the standard deviation of the impurity-induced potential profile for each graphene layer in the $M = 16$ systems with impurity densities of $2 \times 10^{11} \text{ cm}^{-2}$, $5 \times 10^{11} \text{ cm}^{-2}$, and $1 \times 10^{12} \text{ cm}^{-2}$. A layer-by-layer correlation between the magnitude of the potential fluctuations and the resulting mobility is observed. In the layers close to the substrate, large potential variations are accompanied by significantly reduced mobility, indicating that the transport is strongly limited by impurity scattering. As the layer index increases, the potential fluctuations diminish, and the corresponding mobility increases. When the fluctuation amplitude falls below approximately 1 mV, as occurs around the 10th layer in the $2 \times 10^{11} \text{ cm}^{-2}$ system, the mobility approaches the phonon-limited value. Thus, suppressing the potential variation to approximately 1 mV level effectively removes most impurity-induced mobility degradation. The dashed lines represent the analytical model of Eq. 3.35, which captures the trend of the Monte Carlo results.

3.4 Summary

This chapter examined how charged impurities on the substrate influence the electrostatic potential and carrier transport in multilayer graphene systems, and how increasing the number of graphene layers suppresses impurity-induced scattering. The numerical results from self-consistent electrostatic calculations and ensemble Monte Carlo simulations are presented, followed by an analytical model that connects mobility to potential fluctuations.

The potential analysis revealed that impurities generate strong, localized potential peaks in the graphene layer closest to the substrate. These fluctuations decay rapidly with distance from the impurity plane due to screening by carriers in the lower graphene layers. In systems containing up to sixteen layers, the potential

variation diminishes, and the potential profile becomes almost flat in the uppermost layer. The standard deviation of the potential in each graphene sheet, used to quantify the fluctuation variation, exhibits an approximately exponential decay with layer index, and this trend holds even when impurity density is increased. Thus, the multilayer structure effectively screens the impurity potential, with most of the screening achieved within the first several layers.

Transport calculations further showed that the suppression of potential fluctuations translates directly into enhanced carrier mobility and drift velocity in upper layers. Layers near the substrate experience strong impurity scattering and exhibit significantly reduced transport properties. In contrast, layers farther from the substrate exhibit improved mobility and, in the upper layers, the transport becomes nearly identical to the phonon-limited behavior of an impurity-free graphene sheet. At high electric fields, the applied field more strongly dominates the impurity-induced field, leading to a steeper velocity increase and earlier velocity saturation.

Comparison across different impurity densities demonstrated that higher impurity concentrations degrade mobility in all layers, but the qualitative trend of rapid recovery with layer index remains unchanged. Nevertheless, the convergence to phonon-limited mobility shifts to higher layers when impurity density is larger, reflecting the stronger potential variations created by the higher density impurities.

An analytical model was then introduced to relate mobility to the impurity-induced potential fluctuations. The model established that mobility is inversely proportional to the potential fluctuation. This framework reproduces the layer-by-layer trend observed in the MC results and captures how mobility improves as the potential landscape becomes smoother.

Overall, this chapter shows that multilayer graphene provides an effective pathway to mitigate impurity scattering originating from the substrate. Accordingly, the upper graphene layers achieve higher mobility than those closer to the substrate.

Chapter 4

Edge Scattering in Turbostratic Graphene

4.1 Introduction

As discussed in Chapter 3, the multilayer graphene systems with turbostratic stacking have promising characteristics for utilizing the high mobility of graphene. Thus far, the impact of impurity scattering on the transport properties has been discussed. Here, the impact of the edge scattering, as another key limiting factor of the mobility, is also discussed. The size of the system and the edge geometry itself has long been known to affect graphene's electronic structure [74–76]. Consequently, the edge scattering plays an important role in determining the transport properties, as it alters the transport behavior of the carriers [77–82]. In the systems with smaller scales such as nanoribbon graphene, the influence of the boundary becomes increasingly important because the lateral dimensions are comparable to the mean free path [76, 83–85]. This has the implication that even when impurity scattering is minimized, edge scattering can still impose a fundamental limitation on carrier mobility. Previous studies have shown that rough or disordered graphene edges induce diffusive scattering, in which the collisions randomize the direction of carrier motion [86, 87]. Since diffusive scattering is commonly observed in graphene, and strongly affects transport behavior, it is important to incorporate edge-scattering effects into the simulation model.

In this chapter, the effects of boundary collisions on the transport properties are studied. While in the model studied here, carriers are not restricted as strongly as in graphene nanoribbons, the finite system width defined by the edges still imposes a limiting factor on carrier transport and consequently alters the transport behavior.

In multilayer graphene systems, the interplay between interlayer screening and edge collisions introduces additional complexity. The lower layers can effectively screen the impurity potential, while the outermost layers remain susceptible to boundary-induced scattering. Therefore, even when impurity scattering is reduced, the edge-scattering effects can still significantly affect the mobility of the system. Understanding this interplay is essential for accurately evaluating the transport behavior of multilayer systems, where both impurity and edge effects coexist.

In this chapter, a probabilistic model for boundary collisions is introduced, followed by Monte Carlo transport simulations incorporating this model. In Sect. 4.2, the schematic configuration of the multilayer system studied in this chapter is described. In Sect. 4.3, the boundary treatment and the implementation of edge-scattering events in the Monte Carlo framework are explained. Section 4.4 presents the simulation results and discusses how edge scattering modifies transport behavior across different layers and conditions. Finally, in Sect. 4.5, an analytical formulation is developed to describe the edge-induced carrier dynamics and to validate the numerical trends.

4.2 System Structure

The multilayer graphene system studied in this chapter is shown in Fig. 4.1. Here, similar to the previous chapter, the graphene layers are placed on top of a SiO_2 substrate ($\kappa_{\text{SiO}_2} = 3.9$), with impurities residing below the graphene, on the surface of the substrate. Both the substrate and the graphene sheet have the same length, L . The width of the graphene is set to w , whereas the width of the substrate is equal to its length. Each graphene layer is indexed by l , which ranges from 1 to

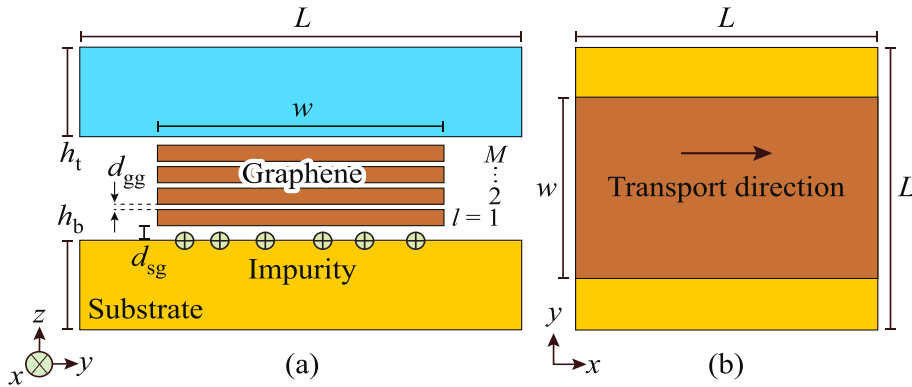


Figure 4.1: A schematic figure of the multilayer graphene system for studying the transport properties with the inclusion of edge scattering. The carriers are confined in the graphene sheet with a width of w .

M . The layer closest to the substrate is represented by $l = 1$. An insulating layer is also placed on top of the graphene layers. The heights of the substrate and the insulating layer are h_b and h_t , respectively. The top insulator has the same relative permittivity as the substrate. Except for the surface of the substrate, both the substrate and the insulator are devoid of charge. The distance d_{sg} represents the gap between the closest graphene sheet to the substrate, and d_{gg} represents the gap between the graphene sheets. Donor impurities are placed at randomly chosen positions in the area below the graphene sheet. Therefore, N_{imp} impurities yield the impurity areal density of $n_D = N_{\text{imp}}/(w \times L)$.

4.3 Monte Carlo Simulation

Here, the self-consistent calculation of Poisson equation and Thomas-Fermi approximation is performed as explained in Chapter 2. However, it is important to mention that in the present model, the impurities are located directly below the graphene sheet on the substrate, while the induced carriers reside within the graphene layers. The other regions of the structure are devoid of impurities and carriers. Thus, no charge exists in the areas outside the graphene sheet ($y < (L - w)/2$ and $y > (L + w)/2$).

After obtaining the potential profile for each layer, the Monte Carlo simulation is performed to calculate the transport properties. The details of the MC method are explained in Section 2.4. However, in this chapter, a probabilistic model is incorporated into the MC simulation, to model the transport behaviors due to edge scattering. In the structures studied in Chapter 3, periodic boundary conditions were applied to all sides of the graphene sheet. However, here, the assumption is that the carriers in the graphene move along the x -axis, confined between edges perpendicular to the y -axis. Thus, the periodic boundary condition applies only along the transport direction. When the carriers arrive at the top edge in the positive side of the y -axis, or on the contrary at the bottom edge in the negative side of the y -axis, the edge scattering occurs and they bounce back.

The post-collision direction of the carriers is determined stochastically by the edge-scattering parameter r . The parameter r characterizes the extent of randomness in edge scattering. $r = 0$ denotes specular reflection at the edges, where carriers are reflected with the same angle as the incident angle. On the other hand, $r = 1$ corresponds to fully diffusive scattering, where scattering angles are determined randomly. The schematic illustration of the edge interaction employed in this study is

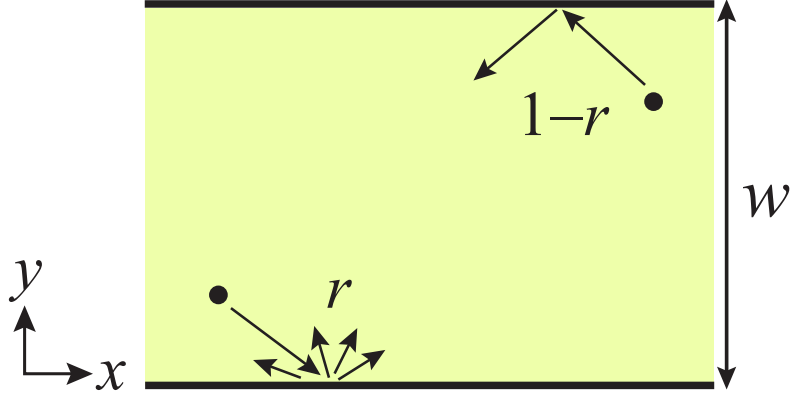


Figure 4.2: A schematic illustration of the probabilistic edge interaction model used in this chapter. The parameter r indicates the scale of diffusive scattering occurring at the edges, while $1 - r$ represents the scale of specular reflection.

shown in Figure 4.2.

The edge scattering is incorporated into the MC calculations as described below. As discussed in the section 2.4, at the end of the free-flight, the updated x and y positions of the carriers along with their wave vectors, are determined using Eqs. 2.21a, 2.21b, 2.23a and 2.23b. Here, after updating the positions, a check is performed to identify any possible boundary encounters. Since the edges are located at fixed positions perpendicular to the y -axis, a carrier is considered to have reached an edge if its y -position goes beyond these limits during free-flight. Hence, the edge scattering mechanism is taken into account when the carrier contacts a boundary. First, a random number t is drawn from a uniform distribution, ranging from 0 to 1, and its value is compared with parameter r . If $t > r$, the carrier undergoes specular reflection at the edge, while $t < r$ indicates diffusive scattering. Once the type of edge interaction is determined, the positions and the wave vector are updated accordingly. For specular reflection events, the x component of the wave vector is updated according to

$$k_{x,\text{new}} = k_x \mp \frac{et_{\text{edge}}}{\hbar} (F_{\text{ext},x} + F_{\text{imp},x}), \quad (4.1)$$

where $t_{\text{edge}} = t_s$, the time-step parameter. In the case that phonon scattering takes place during the time step, $t_{\text{edge}} = t'_f$, where t'_f corresponds to the free-flight time within the current time step. Unlike the x component, updating the y component occurs in two steps. This is because, irrespective of whether electrons collide with the top edge or bottom edge, their direction of movement along the y -axis reverses after the collision, resulting in a change in the sign of the y component of the wave vector. Accordingly, t_{edge} is split into two segments: t_1 , corresponding to the free-flight time

before arriving at the edge, and t_2 , representing the time after the collision. Thus, k_y is computed through a three-step process:

$$k_{y,1} = k_y \mp \frac{et_1}{\hbar} F_{\text{imp},y}, \quad (4.2)$$

$$k_{y,2} = -k_{y,1}, \quad (4.3)$$

$$k_{y,\text{new}} = k_{y,2} \mp \frac{et_2}{\hbar} F_{\text{imp},y}. \quad (4.4)$$

Here, $k_{y,1}$ denotes the y component of the wave vector for the free-flight segment up to the edge before collision, $k_{y,2}$ is the y component with a reversed sign, signifying the change in direction along the y -axis after reflecting from the edge, $k_{y,\text{new}}$ is the final y component of the wave vector including the free-flight after edge reflection. Following the calculation of the new wave vector, the corresponding direction of carrier movement is calculated as

$$\alpha = \arctan \left(\frac{k_{y,\text{new}}}{k_{x,\text{new}}} \right). \quad (4.5)$$

Accordingly, the positions of the carriers are also calculated.

For diffusive scattering ($t < r$), the free-flight time t_{edge} is also split into two intervals, t_1 and t_2 , corresponding to the duration of free-flight before and after the scattering event. For the free-flight prior to the scattering event, the following equations are used to describe the carrier movement:

$$k_{x,1} = k_x \mp \frac{et_1}{\hbar} (F_{\text{ext},x} + F_{\text{imp},x}), \quad (4.6)$$

$$k_{y,1} = k_y \mp \frac{et_1}{\hbar} F_{\text{imp},y}. \quad (4.7)$$

After completing the free-flight with duration of t_1 , the following series of equations are used to describe the diffusive scattering:

$$|\mathbf{k}_{\text{mag},1}| = \sqrt{k_{y,1}^2 + k_{x,1}^2} \quad (4.8)$$

$$k_{x,2} = |\mathbf{k}_{\text{mag},1}| \cos \theta, \quad (4.9)$$

$$k_{y,2} = |\mathbf{k}_{\text{mag},1}| \sin \theta. \quad (4.10)$$

Using Eq. 4.8, the magnitude of the wave vector is determined, and with Eqs. 4.9 and 4.10, new components of the wave vector after the scattering event are calculated. Angle θ defines the new direction of the carriers and is computed using a random number. Finally, the remaining portion of free-flight after the scattering is described

by

$$k_{x,\text{new}} = k_{x,2} \mp \frac{et_2}{\hbar} (F_{\text{ext},x} + F_{\text{imp},x}), \quad (4.11)$$

$$k_{y,\text{new}} = k_{y,2} \mp \frac{et_2}{\hbar} F_{\text{imp},y}. \quad (4.12)$$

Edge scattering is modeled as elastic scattering in this study; therefore, the magnitude of the wave vector $|\mathbf{k}_{\text{mag},1}|$ does not change due to the scattering event. Additionally, in the present model, localized electronic edge states of graphene are not included. Therefore, neither carrier exchange with such states nor Coulomb interactions with charges localized at the edges are considered.

4.4 Results and Discussion

4.4.1 Potential Profile

Figure 4.3 shows the positions of $N_{\text{imp}} = 100$ donor impurities placed on the substrate below the graphene sheet of width $w = 100$ nm and length $L = 200$ nm, with an impurity density of $n_D = 5 \times 10^{11} \text{ cm}^{-2}$. Here, the remaining structural parameters of the systems are adopted from Table 2.1. The bottom edge is located at $y = 0$ and the top edge is located at $y = 100$ nm. The initial impurity locations are

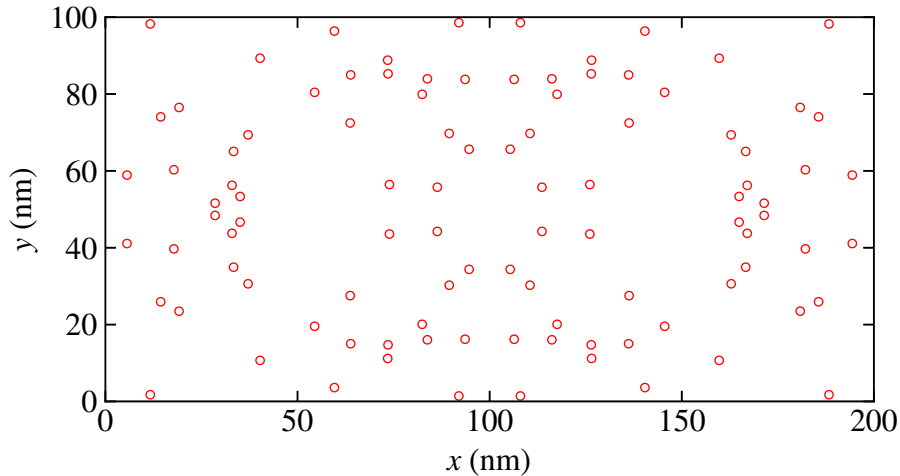


Figure 4.3: Spatial distribution of $N_{\text{imp}} = 100$ donor impurities on a substrate, with impurities distributed over a $100 \text{ nm} \times 200 \text{ nm}$ area corresponding to an impurity density of $n_D = 5 \times 10^{11} \text{ cm}^{-2}$. Impurity positions are randomly allocated from 0 to 100 nm in the x -axis and from 0 to 50 nm in the y -axis and subsequently mirrored across both the $x = 100 \text{ nm}$ and $y = 50 \text{ nm}$ lines. The symmetrical arrangement near the edges preserves consistent scattering environments.

randomly chosen between 0 and 100 nm along the x -axis and between 0 and 50 nm along the y -axis.

To maintain a balanced environment, the impurity distribution is mirrored in two directions. First, it is mirrored across $x = 100$ nm, which cancels out small differences in the potential profile caused by periodic boundary conditions. Second, the distribution is mirrored across $y = 50$ nm, so that both edges of the graphene experience a similar impurity environment. Although edge scattering is stochastic and varies at each event, this structure maintains similar potential conditions near the top and bottom edges of graphene. This ensures that impurity effects are not unevenly introduced between the two boundaries.

Figure 4.4 presents a three-dimensional representation of the induced potential calculated by the self-consistent simulation. An offset of 0.37 V is applied between each potential profile of the layers to enhance visibility. The figure shows that the lower layers exhibit sharper potential peaks, whereas the upper layers display more

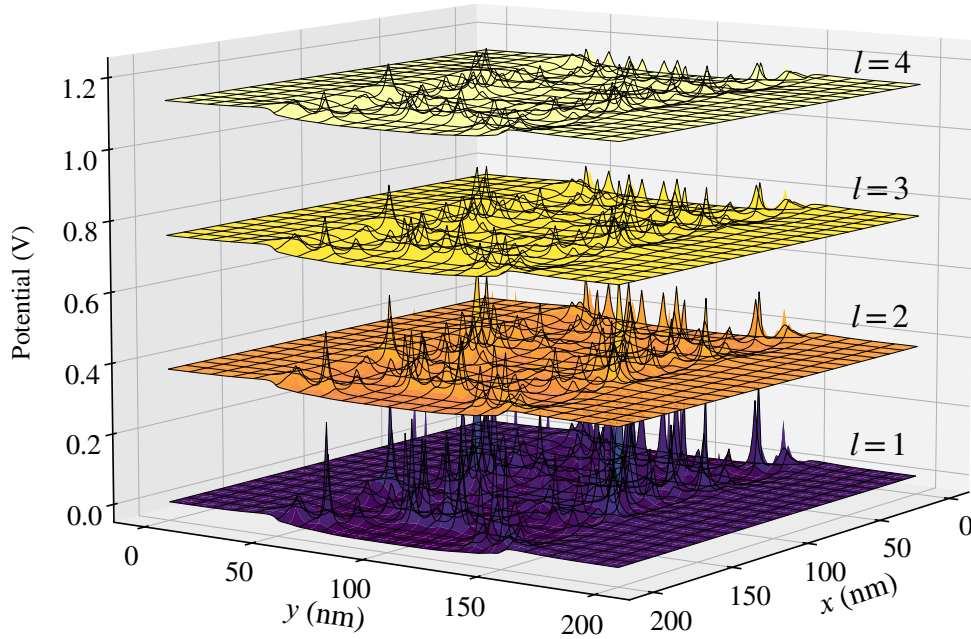


Figure 4.4: A three-dimensional figure of the induced potential, obtained from self-consistent simulations of a graphene system with $M = 4$ layers. To improve visual clarity, an offset of 0.37 V has been introduced between layers. The potential profiles exhibit sharp peaks in the lower layers, which become less pronounced in the upper layers, indicating the screening effect of the lower layers on the overall potential distribution.

subdued peaks, reflecting the potential flattening effect caused by screening from the lower layers. To be more specific, the standard deviation of the potential fluctuations is reduced from 23.9 mV in the first layer to 12.2 mV in the fourth layer.

4.4.2 Transport Properties

Upon obtaining the potential profile of each layer, the transport properties are evaluated using the MC simulation. Prior to incorporating the impurity effects, the transport properties are examined for systems with different scattering parameters in the absence of impurities. Hence, the MC simulation is performed for a graphene system with $M = 4$ and values of $r = 0$ to $r = 1.0$ with an increment of 0.1.

Figure 4.5 depicts the drift velocity of the graphene layers for various values of r , plotted against the applied electric field in the impurity-free system. For the case of $r = 0$, in which electrons undergo specular reflection at the boundaries, the drift velocity is shown by the solid line. This curve matches the impurity-free drift velocity of a graphene sheet calculated in Chapter 3, where periodic boundary

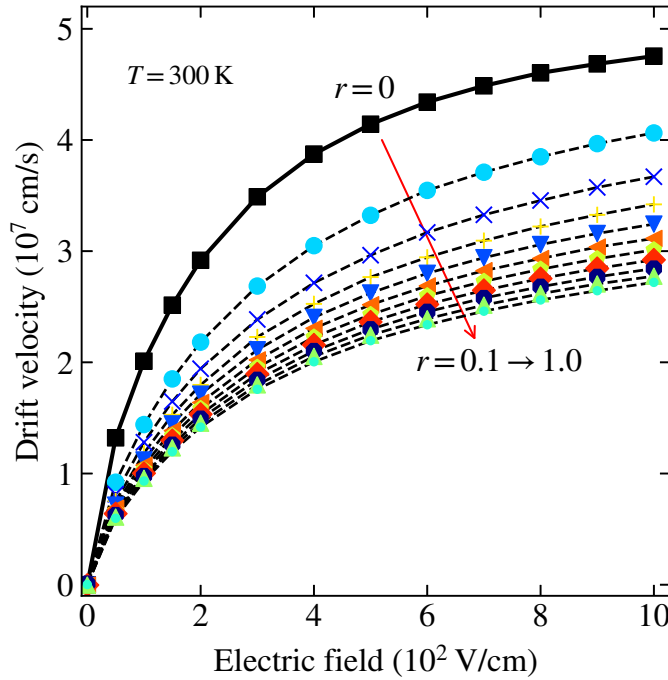


Figure 4.5: Drift velocity versus applied electric field for graphene layers with different edge scattering parameters r in the absence of impurities. The solid line ($r = 0$) denotes specular edge reflection. Curves below the solid line represent increasing r values (from 0.1 to 1.0), in which stronger random edge scattering reduces the drift velocity. The figure illustrates the achievable drift velocity range when only phonon and edge-scattering mechanisms are present.

conditions were applied along all edges. As r increases, the corresponding drift velocity curves appear below the $r = 0$ line, with the lowest curve corresponding to $r = 1$.

Larger values of r correspond to stronger random edge scattering, meaning that the electron motion becomes more randomized at the edges. As a result, the drift velocity decreases systematically as r increases. Particularly, the velocity reduction becomes more pronounced in the high-field region, where electrons with higher energy interact with the boundaries, and consequently their motion is altered more frequently. Thus, Fig. 4.5 demonstrates the range of drift velocities achievable in the considered graphene layers under phonon and edge scattering alone, excluding impurity effects. From this, it can be deduced that even in systems without impurity scattering, edge scattering alone can noticeably reduce the drift velocity of the electrons.

Fig. 4.6 presents the average drift velocity of carriers in a four-layer graphene structure ($M = 4$) under four different impurity configurations and varying values of parameter r . When $r = 0$, the fourth (uppermost) layer exhibits a considerably higher drift velocity compared to the lower layers. As edge scattering becomes more pronounced, the drift velocities of the upper layers (the third and fourth layers) decrease, whereas the velocities of the lower layers show only a slight decrease. For example, at $r = 1$, the velocity contrast between the fourth and lower layers becomes much smaller compared to configurations with weaker edge scattering. As established in Chapter 3, multilayer graphene with a turbostratic structure tends to exhibit enhanced transport properties in the upper layers, mainly due to the screening effect from the lower layers. However, Fig. 4.6 demonstrates that this advantage is largely undermined when edge scattering is frequent. Even though the upper layers experience smoother potential landscapes, stronger boundary scattering suppresses their drift velocity and limits the overall transport properties of the upper layers.

The drift velocities of carriers in a four-layer graphene system ($M = 4$), subjected to an external electric field of $F = 10^3$ V/cm, are illustrated in Fig. 4.7. The results are averaged for four distinct impurity distributions across different values of r . Here, two primary effects are observed. First, it can be seen that the drift velocity is almost unchanged for the first layer and decreases slightly in the second layer. This is because in the lower layers, impurity scattering is the most prominent scattering mechanism; therefore, the edge scattering has little impact on carrier velocity. However, in layers farther from the substrate (third and fourth layers), the drift velocity decreases with increasing r . This occurs due to the reduced impact of

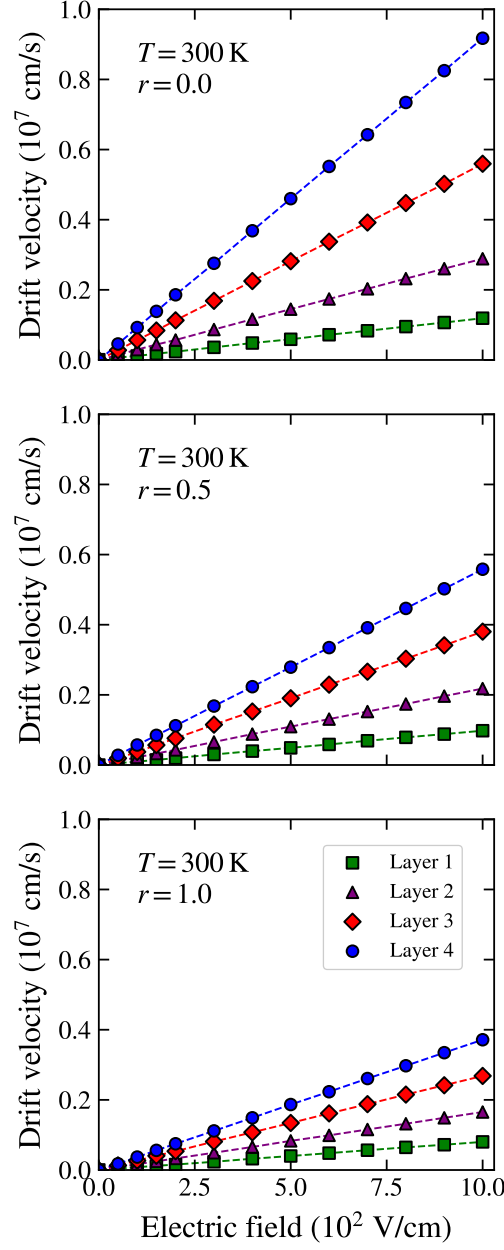


Figure 4.6: Drift velocity of carriers across four impurity distribution patterns in a four-layer graphene system ($M = 4$) under varying edge-scattering parameters r . For $r = 0$, the system exhibits the highest drift velocity for every layer index l , while increasing r (approaching 1) reduces the velocity gap between the upper and lower layers. This indicates that although the upper layers in turbostratic graphene generally exhibit better transport due to screening by lower layers, this benefit is reduced with more random edge scattering.

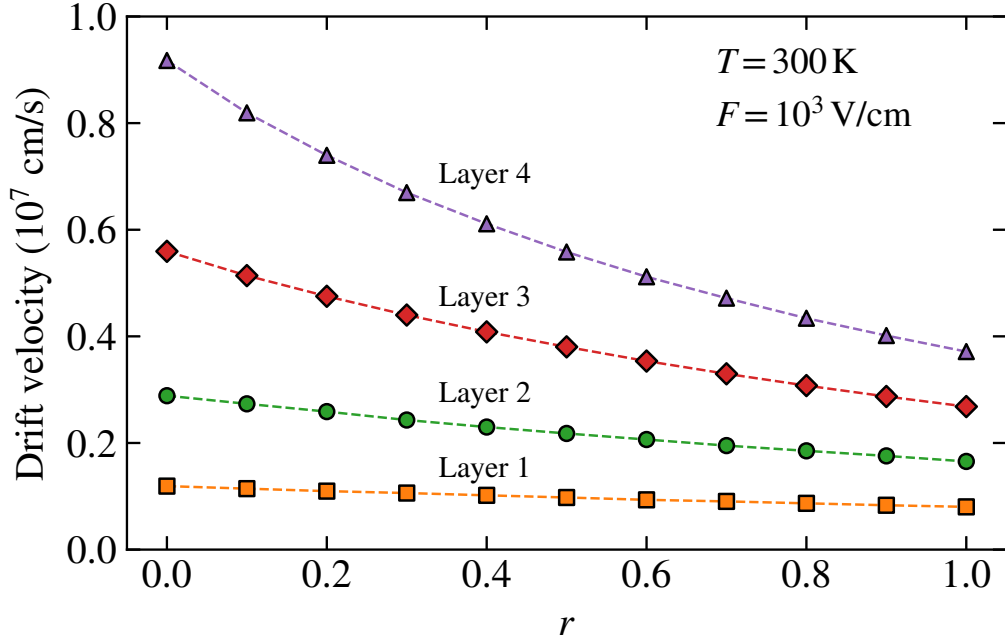


Figure 4.7: Drift velocities of carriers in a four-layer graphene system under an applied electric field of $F = 10^3$ V/cm for different values of the edge scattering parameter r . In the lower layers, impurity scattering dominates, and edge effects are negligible, while in the upper layers, increasing r significantly reduces the drift velocity. The velocity gap between layers also decreases with increasing r , as enhanced edge scattering affects the upper layers more strongly. Results are averaged over four impurity distributions.

impurity scattering in the upper layers, where edge effects become more influential, resulting in a decrease in drift velocity with increasing r . For instance, in the fourth layer, the velocity declines from roughly 9.2×10^6 cm/s at $r = 0$ to about 3.7×10^6 cm/s at $r = 1$. The second observation is that the velocity differences among layers decrease as r increases. For example, the gap between the first and fourth layers narrows from 8.0×10^6 cm/s at $r = 0$ to 2.9×10^6 cm/s at $r = 1$. This reduction is caused by enhanced random edge scattering at higher r values, which affects the upper layers to a greater extent, where impurity scattering is weaker, thus leading to a reduction in the velocity differences among the layers.

Figure 4.8 shows the drift velocity normalized by its value at $r = 0$ for each graphene layer. It is observed that the normalized drift velocity decreases monotonically with increasing r for all layers. For systems with relatively large edge-scattering parameters ($r \gtrsim 0.6$), the rate of decrease is nearly identical across all layers. In contrast, for smaller values of r , the reduction in the normalized drift velocity is more pronounced in the upper layers.

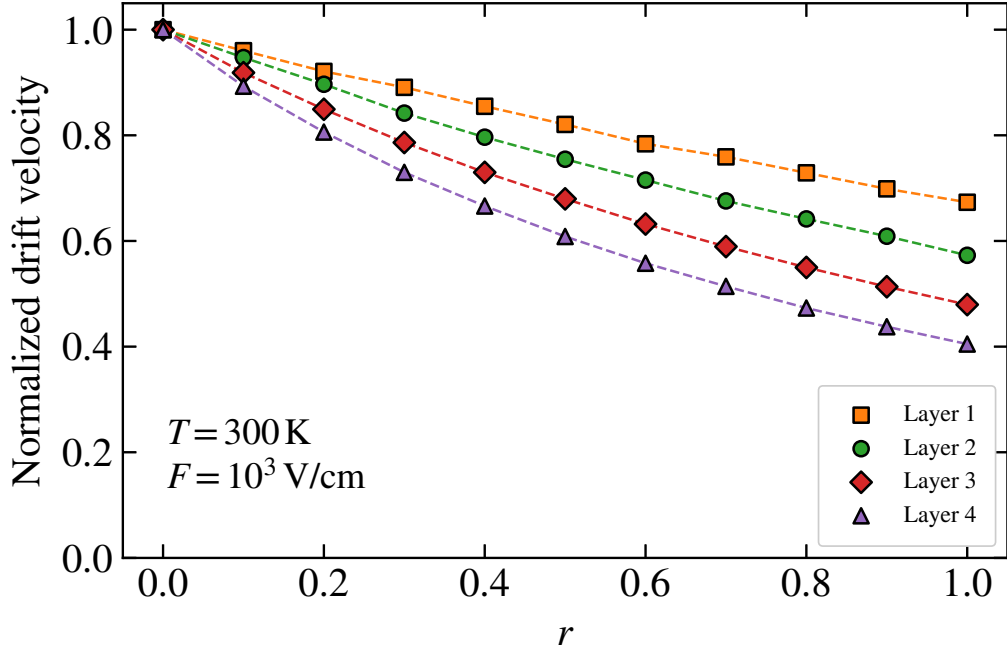


Figure 4.8: Normalized drift velocity as a function of the edge-scattering parameter r for each layer in a four-layer graphene system at $T = 300$ K and $F = 10^3$ V/cm. The drift velocity in each layer is normalized by its corresponding value at $r = 0$. For larger values of the edge-scattering parameter ($r \gtrsim 0.6$), the rate of decrease in the normalized drift velocity is nearly the same for all layers, whereas for smaller r values, the reduction is more pronounced in the upper layers.

This behavior arises because the absolute magnitude of the potential fluctuations is smaller in the upper layers due to screening, such that even a small increase in edge scattering leads to a relatively sharp reduction in the drift velocity. In the lower layers, impurity scattering acts as a strong transport limiting mechanism, and therefore a modest increase in edge scattering results in a comparatively smaller decrease in velocity. As r increases further, edge scattering becomes strong enough that its relative influence is nearly the same for all layers. Nevertheless, the normalized drift velocity is lower in the higher layers for all values of r .

4.5 Analytical Model

An analytical model is developed here to characterize the transport properties of graphene with edge scattering taken into account. In Chapter 3, an analytical model of the transport properties with respect to impurity scattering was presented. Based on that model, the electron conductivity in graphene in terms of the transport

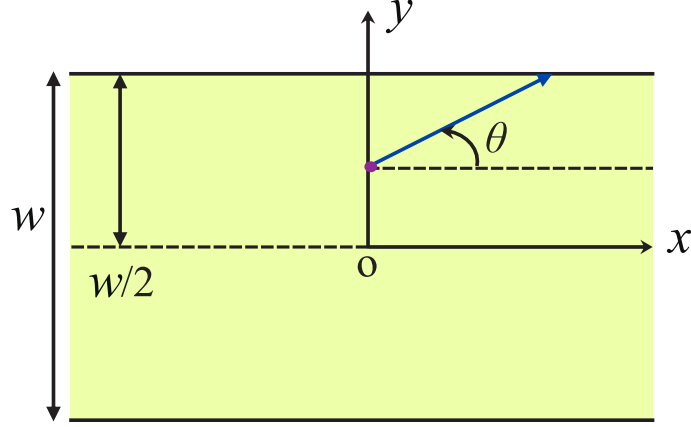


Figure 4.9: Schematic of a graphene sheet of width w . The x -axis is along the transport direction, y -axis across the width, and origin o at the center. At $t = 0$, an electron moves at angle θ to the x -axis toward the nearest edge.

relaxation time τ_{tr} , can be written as [72, 73]

$$\sigma = \frac{e^2}{\pi \hbar^2} \tau_{\text{tr}} \int f(E) dE. \quad (4.13)$$

Here, τ_{tr} is expressed in terms of the energy-dependent relaxation time $\tau(E)$ as

$$\tau_{\text{tr}} = \frac{\int E \tau(E) \left(-\frac{\partial f}{\partial E}\right) dE}{\int E \left(-\frac{\partial f}{\partial E}\right) dE}. \quad (4.14)$$

In the case of non-degenerate carriers, the mobility μ can be written as

$$\mu = \frac{ev_{\text{f}}^2}{2k_{\text{B}}T} \tau_{\text{ave}}, \quad (4.15)$$

where τ_{ave} is the average relaxation time, described by

$$\tau_{\text{ave}} = \frac{1}{(k_{\text{B}}T)^2} \int E \tau(E) e^{-E/k_{\text{B}}T} dE. \quad (4.16)$$

Based on these formulations, an analytical model that includes edge scattering is constructed. Here, the coordinate system is defined such that the x -axis aligns with the transport direction, and the y -axis extends across the width of the graphene sheet. The graphene sheet has a width of w , and the origin of the coordinate system is set at its center, as depicted in Fig. 4.9. It is considered that an electron moves from the point $(0, y)$ towards the graphene edge with an angle θ with respect to the x -axis. More specifically, the initial location of the electron is set at a position $0 \leq y \leq \frac{1}{2}w$, at an angle $0 \leq \theta \leq \frac{\pi}{2}$ with respect to the x -axis. Knowing the initial

y coordinate of the position of the electron and its angle relative to the edge, the time τ_r at which an electron reaches the edge can be estimated as

$$\tau_r = \frac{\frac{1}{2}w - y}{v_f \sin \theta}, \quad (4.17)$$

where v_f denotes the Fermi velocity. Assuming that τ_0 is the relaxation time due to phonon and impurity scattering, and using Matthiessen's rule, the total relaxation time τ is obtained by

$$\tau = \left(\frac{r}{\tau_r} + \frac{1}{\tau_0} \right)^{-1} = \left(\frac{rv_f \sin \theta}{\ell} + \frac{1}{\tau_0} \right)^{-1} = \frac{\ell}{rv_f} (\sin \theta + a)^{-1}. \quad (4.18)$$

Here, r denotes the edge-scattering parameter ranging from 0 to 1, $\ell = \frac{1}{2}w - y$, and a is a dimensionless parameter calculated by $a = \ell/(rv_f\tau_0)$. Averaging τ over the angles yields

$$\langle \tau \rangle = \frac{2}{\pi} \int_0^{\pi/2} \tau d\theta = \frac{2}{\pi} \frac{\ell}{rv_f} \int_0^{\pi/2} (\sin \theta + a)^{-1} d\theta = \tau_0 F(a). \quad (4.19)$$

Here, $F(a)$ is defined as

$$F(a) = \begin{cases} \frac{2a}{\pi} \frac{\arctan(\sqrt{a^2 - 1})}{\sqrt{a^2 - 1}}, & (a > 1), \\ \frac{2a}{\pi} \frac{\operatorname{arctanh}(\sqrt{1 - a^2})}{\sqrt{1 - a^2}}, & (a < 1). \end{cases} \quad (4.20)$$

Furthermore, averaging $\langle \tau \rangle$ over y yields

$$\langle \langle \tau \rangle \rangle = \frac{2}{w} \int_0^{w/2} \langle \tau \rangle dy = \tau_0 G(b), \quad (4.21)$$

where $G(b)$ is described by

$$\begin{aligned} G(b) &= \frac{1}{b} \int_0^b F(a) da \\ &= \begin{cases} \frac{2}{\pi b} \left[\beta_+ \arctan \beta_+ - \log \left(\frac{1}{2}b \right) \right], & (b > 1), \\ -\frac{2}{\pi b} \left[\beta_- \operatorname{arctanh} \beta_- + \log \left(\frac{1}{2}b \right) \right], & (b < 1). \end{cases} \end{aligned} \quad (4.22)$$

Here, $b = w/(2rv_f\tau_0)$ and $\beta_{\pm} = [\pm(b^2 - 1)]^{1/2}$.

Using Eq. 4.15 and the averaged relaxation time $\langle \langle \tau \rangle \rangle$ calculated from Eq. 4.21,

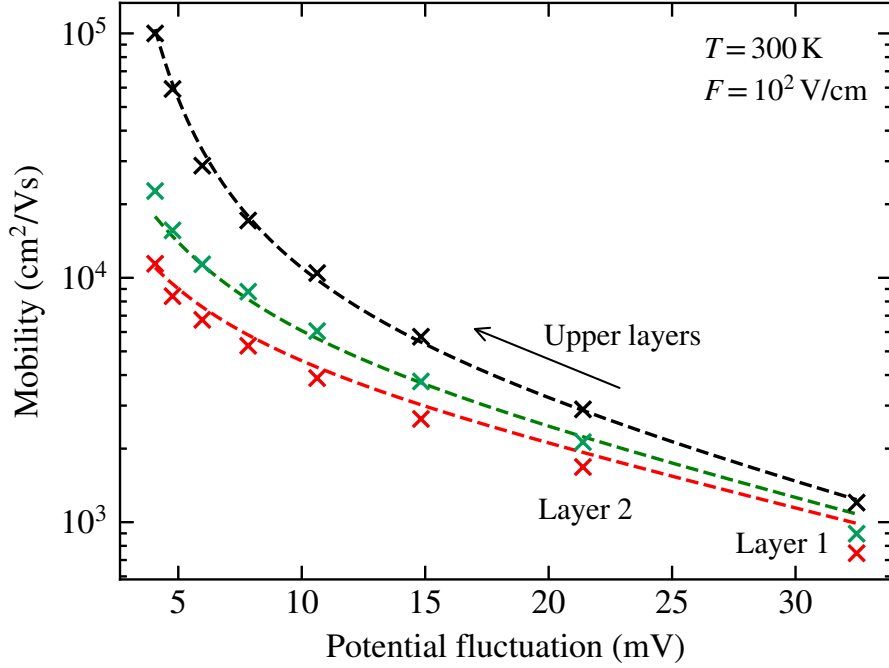


Figure 4.10: Comparison of Monte Carlo simulation results (symbols) and the analytical model (dashed lines) for electron mobility in an $M = 8$ layer system. The impurity density is fixed at $n_D = 5 \times 10^{11} \text{ cm}^{-2}$, with data averaged over four impurity distributions. The black dashed line represents the fit for the specular case ($r = 0$), which serves as the baseline for determining the relaxation time τ_0 . The green and red curves depict the theoretical predictions for edge scattering parameters $r = 0.5$ and $r = 1.0$, respectively.

the mobility μ is then obtained by

$$\mu = \frac{ev_f^2}{2k_B T} \langle \langle \tau \rangle \rangle = \mu_0 G(b). \quad (4.23)$$

Here, μ_0 denotes the mobility of a graphene sheet in the absence of edge scattering, defined as $\mu_0 = ev_f^2 \tau_0 / (2k_B T)$. Accordingly, the dimensionless parameter b is given in terms of μ_0 by $b = ev_f w / (4r\mu_0 k_B T)$. From Eq. 4.23, it can be inferred that edge scattering reduces the mobility by a factor of $G(b)$. Here, $G(b)$ is a monotonically increasing function, with $G(0) = 0$ and $G(\infty) = 1$. Thus, $G(b)$ serves as a measure of how strongly edge scattering suppresses transport, starting from high suppression at narrow widths to negligible influence at large widths. In particular, edge scattering can be neglected when b is sufficiently above unity. This condition implies that the width w must be much larger than $2rv_f \tau_0$. This criterion aligns with the basic expectation that edge scattering becomes less significant for system widths larger than the mean free path ($v_f \tau_0$). It should be noted that the probabilistic model

explained in this section is based on the assumption that electrons move toward the nearest edge. This assumption excludes cases where electrons reach the farther edge. However, this simplification is useful, as it allows this empirical model to reproduce the Monte Carlo results with good accuracy.

Figure 4.10 presents the influence of potential fluctuations on electron mobility in an 8-layer system with an impurity density of $n_D = 5 \times 10^{11} \text{ cm}^{-2}$ for three different values of r . The symbols indicate the MC results averaged over four impurity configurations. The black dashed line corresponds to a fit of the $r = 0$ data and is taken as a reference for extracting μ_0 , which includes only phonon and impurity scattering. From this baseline, mobilities for $r = 0.5$ and $r = 1.0$ are evaluated using Eq. 4.23. The corresponding analytical predictions are plotted as dashed lines: a green line for $r = 0.5$, and a red line for $r = 1.0$. Despite being empirical, the aforementioned model reproduces the overall edge-scattering effects on mobility across different impurity fluctuations. However, deviations are observed in the lower layers under strong-scattering conditions, most notably for $r = 0.5$ and $r = 1.0$, where Matthiessen's rule may not hold. The discrepancy becomes most significant at $r = 1.0$.

The trends in Fig. 4.10 corroborate experimental findings for turbostratic multilayer graphene [49], showing that the turbostratic configuration enhances transport properties. Even though the experimental mobilities exhibit lower values than the simulation results, mainly due to narrower widths and higher impurity densities, the qualitative behavior is consistent. This validates the effectiveness of turbostratic stacking in preserving graphene's high mobility

4.6 Summary

In this chapter, the transport properties of turbostratic multilayer graphene were investigated with a focus on the role of edge scattering. Edge scattering was integrated into the calculation using probabilistic modeling. A parameter r was introduced to describe boundary scattering, where $r = 0$ corresponds to specular reflection and $r = 1$ to fully diffusive collisions. The results are consistent with those of Chapter 3 indicating that carrier transport is governed substantially by impurity scattering, and the screening of the fluctuating potential has a positive effect on the transport properties of the upper layers. However, the analysis in this chapter shows that, with the introduction of edge scattering, an additional limitation on carrier mobility emerges that affects the graphene layers. Since edge scattering originates from

the intrinsic structure of graphene layers, it affects every layer in the stack, even when the impurity potential is screened. Furthermore, the simulation results indicate that the edge-scattering effect becomes more pronounced in the upper layers, where the impurity potential is weaker due to screening. In these layers, the relative importance of boundary collisions increases, leading to a reduction in the drift velocity. Finally, an empirical model was introduced in this chapter, which agrees well with the Monte Carlo results. Through the calculations and results of this chapter, it has been shown that integrating edge scattering is vital for constructing an accurate model for the transport behavior of the carriers. Overall, this chapter demonstrates that a comprehensive transport model must simultaneously account for impurity scattering, boundary scattering, and interlayer screening, in order to correctly predict the transport properties of multilayer graphene devices.

Chapter 5

Carrier Transport Simulation in Twisted Bilayer and Multilayer Graphene Structures

5.1 Introduction

In Chapters 3 and 4, it was shown that turbostratic stacking serves as an effective mechanism for screening impurity-induced potential fluctuations and thereby enhances the transport properties of multilayer graphene systems. Building on these results, this chapter investigates the application of twisted bilayer graphene (tBLG) as an interfacial screening layer within turbostratic multilayer structures.

In a tBLG system, two graphene layers are stacked with a relative rotational misalignment characterized by the twist angle θ , generating a spatially periodic Moiré pattern. The twist angle acts as a key parameter that governs the degree of electronic coupling between the layers [66, 88–91]. Figure 5.1 presents the schematics for systems with twist angles of $\theta = 3^\circ$ and $\theta = 6^\circ$ as examples. At most twist angles, the bilayer retains a band structure similar to that of monolayer graphene [92, 93].

However, tBLG has attracted considerable interest because it provides a controllable way to tune interlayer interactions without altering the atomic structure of each layer [94, 95]. As θ decreases, the interaction between the layers becomes stronger, and the carrier motion gradually deviates from the ideal linear dispersion of monolayer graphene. In particular, near the so-called “magic angle”, the band structure becomes extremely flat [89, 96–100], leading to a substantial suppression of the Fermi velocity.

In contrast, the turbostratic structures examined in the previous chapters cor-

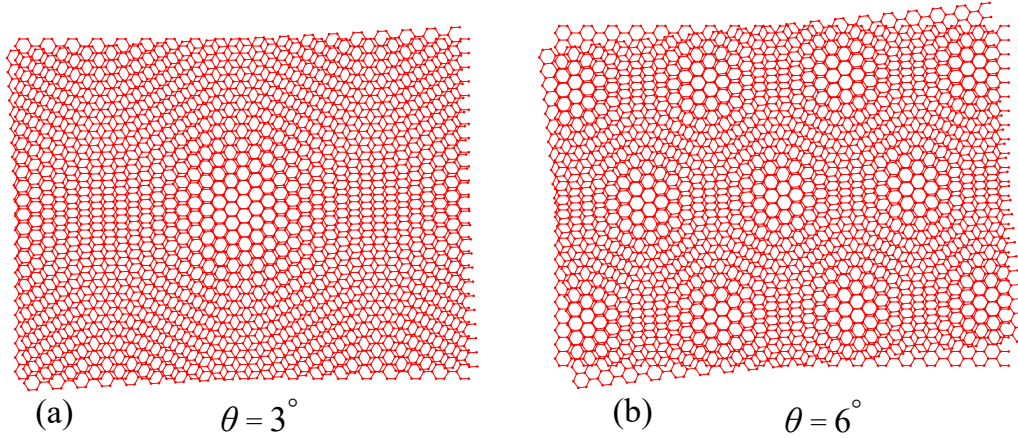


Figure 5.1: Schematics of twisted bilayer graphene for (a) $\theta = 3^\circ$ and (b) $\theta = 6^\circ$.

respond to larger twist angles, typically around 30° , where the graphene layers are effectively misaligned enough to behave as monolayer graphene. In this large-angle regime, the interlayer coupling is very weak, and each sheet retains its own electronic character similar to that of monolayer graphene [51, 101]. When the twist angle becomes smaller, however, the interlayer interaction can no longer be neglected. Theoretical studies have reported that as the twist angle decreases to values on the order of several degrees, the Fermi velocity drops noticeably below that of monolayer graphene, and at sufficiently small angles it becomes even less than one-tenth of the monolayer value [65, 101, 102]. Therefore, the flattening of the band structure and the strong suppression of carrier velocity lead to an increase in the density of states within tBLG layers. Consequently, it is expected that carriers more readily accumulate near charged impurities, thereby weakening and smoothing out the surrounding potential fluctuations.

This behavior suggests that twisted bilayer regions can serve as natural “buffer layers” that help reduce potential variations between adjacent graphene sheets. In particular, a tBLG layer placed near the substrate can act as an effective screening interface that compensates for impurity-induced disorder, complementing the role of turbostratic stacking in multilayer systems [69]. Thus, incorporating tBLG layers within a turbostratic multilayer structure could provide a new pathway toward enhanced carrier transport through reinforced interlayer screening.

The objective of this chapter is to examine multilayer graphene systems that include twisted bilayer configurations and to evaluate their capacity as an effective means for achieving high intrinsic mobility in graphene-based structures. To this end, this chapter analyzes how the presence of tBLG modifies the electrostatic potential profile, alters carrier dynamics, and contributes to the overall mobility

enhancement across the multilayer stack.

5.2 Calculation Model

The calculations presented in this chapter follow the simulation framework described in Chapter 2, with several modifications introduced to incorporate a multilayer graphene system containing twisted bilayer graphene. A brief description of the procedure and the modifications is summarized below. As described in the introduction, the degree of band flattening in tBLG depends on the twist angle. In order to estimate the effects of integrating the tBLG into the turbostratic multilayer graphene system, the exact twist angles of the tBLG are not taken into account, and only the extent of flattening of the bands near the Dirac points is considered. Therefore, in constructing this model, a parameter α is introduced to describe the ratio of the Fermi velocity in tBLG to that in monolayer graphene. Here, α represents the degree of Fermi velocity suppression in tBLG defined by

$$\alpha = \frac{v_{f,\text{tBLG}}}{v_{f,\text{monolayer}}}. \quad (5.1)$$

A smaller α corresponds to heavier carriers, whereas a larger α corresponds to faster carriers. Given these points, the entire graphene system is characterized by two parameters: α , which represents the degree of Fermi velocity suppression in the tBLG layers, and M , which represents the total number of layers, including both the tBLG and turbostratic graphene layers.

The objective of the calculations here is to clarify how inserting a tBLG between the turbostratic graphene layers and the substrate influences impurity-induced potential fluctuations and the eventual transport properties of the graphene layers. The simulation procedure consists of two stages. First, the electrostatic potential is obtained through a self-consistent solution of Poisson equation under the Thomas–Fermi approximation. Second, carrier transport is computed by solving the Boltzmann transport equation using MC simulations. In addition, hypothetical cases with $\alpha > 1$ are considered, to examine the trade-off between tBLGs with faster carriers with weaker screening and slower carriers with stronger screening capability.

Figure 5.2 shows the schematic structure of the systems studied in this chapter. Similar to the systems studied in Chapter 3, the graphene layers are located between the substrate and an insulator. The specific values of parameters denoting the system size are the same as those of the model used in Chapter 3.

The same assumption used in the previous chapters applies here as well. Near the

Dirac points of the turbostratic layers, a linear dispersion $E = \hbar v_f |\mathbf{k}|$ is used. Here, v_f is the Fermi velocity of graphene, and $|\mathbf{k}|$ denotes the magnitude of the carrier wave vector $\mathbf{k} = (k_x, k_y)$. For tBLG layers, a reduction of the Fermi velocity must be considered. This is introduced by the parameter α , resulting in the dispersion relation

$$E = \hbar \alpha v_f |\mathbf{k}|, \quad (5.2)$$

where $\alpha (\leq 1)$ encapsulates the extent of this suppression of the Fermi velocity. Subsequently, the density of states in tBLG is obtained as

$$g(E) = \frac{2|E|}{\pi \hbar^2 (\alpha v_f)^2}. \quad (5.3)$$

It is noteworthy that in both Eqs. 5.2 and 5.3, inserting $\alpha = 1$ would yield the characteristics of monolayer graphene, which is the same as the turbostratic layers in this study.

The details of the MC simulation are the same as those of the systems in Chapter 3. However, as for the phonon scattering, the equations are modified to incorporate the parameter α in the model. Here, acoustic phonon (AP) scattering and optical phonon (OP) scattering are also considered. The elastic AP scattering rates for electrons in the conduction band are obtained by

$$S_{\text{ap}}^{\text{elas}} = \frac{4E_D^2 k_B T}{\hbar^3 (\alpha v_f)^2 v_s^2 \rho_s} E. \quad (5.4)$$

Here, the variables are defined as follows: ρ_s is the density of graphene, v_s is the sound velocity, and E_D denotes the deformation potential. The inelastic scattering

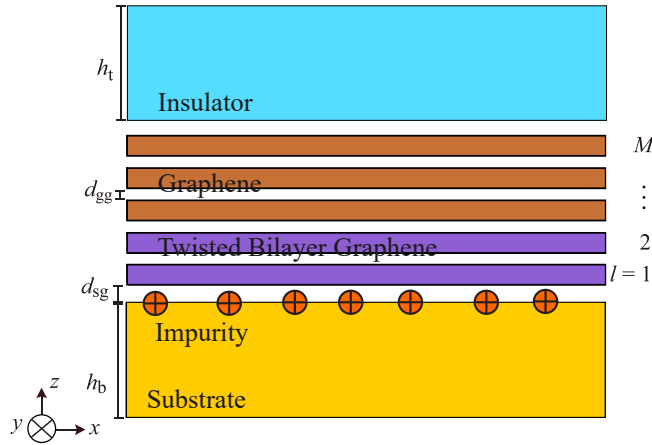


Figure 5.2: Schematic of the multilayer graphene system containing a tBLG layer.

Table 5.1: Parameters for the phonon scattering rates

Density of graphene ρ (g/cm ²)	7.6×10^{-8}
Sound velocity v_s (cm/s)	2×10^6
Optical phonon energy $\hbar\omega_{\text{op}}$ (meV)	164
Intervalley acoustic phonon energy $\hbar\omega_{\text{ac}}$ (meV)	124
Deformation potential of acoustic phonon scattering E_D (eV)	4.6
Deformation field of optical phonon scattering $D_{\text{f,op}}$ (eV/cm)	1.0×10^9
Deformation field of acoustic phonon scattering $D_{\text{f,ac}}$ (eV/cm)	3.5×10^8

rates for electrons are governed by the following expression

$$S_{\text{op(ap)}}^{\text{inelas}} = \frac{D_{\text{f,op(ac)}}^2}{\hbar^2 \omega_{\text{op(ac)}} \rho_s (\alpha v_f)^2} \left[(E + \hbar\omega_{\text{op(ac)}}) N_{\text{op(ac)}} + (E - \hbar\omega_{\text{op(ac)}}) (N_{\text{op(ac)}} + 1) \Theta(E - \hbar\omega_{\text{op(ac}})) \right]. \quad (5.5)$$

The parameters in the equation are defined as follows: $\omega_{\text{op(ac)}}$ is the phonon frequency (optical or acoustic). $D_{\text{f,op(ac)}}$ is the deformation field corresponding to the scattering process. $\Theta(x)$ represents the Heaviside function. This function acts as a threshold, to ensure that the electron's energy is sufficient for a phonon emission process to occur.

After conducting the MC simulation and obtaining the drift velocity of each layer, the transport properties of the entire system are obtained by averaging the carrier velocities over all layers, weighted by the corresponding carrier densities. In each layer l , the velocities of the electrons and holes are denoted by $v_{x,l,e}$ and $v_{x,l,p}$, and the respective carrier densities are n_l and p_l , yielding

$$v_{x,\text{avg}} = \frac{\sum_{l=1}^M (n_l v_{x,l,e} - p_l v_{x,l,p})}{\sum_{l=1}^M (n_l + p_l)}, \quad (5.6)$$

as the drift velocity and

$$\mu_{\text{avg}} = \frac{v_{x,\text{avg}}}{|F_x|}, \quad (5.7)$$

as the mobility of the entire system under an electric field of magnitude F_x . Thus, the dynamics of transport properties of the entire system can be evaluated and studied. The parameters used for the phonon scattering rate calculations are listed in Table 5.1.

5.3 Results and Discussion

5.3.1 Potential Profile

The simulations were carried out for multilayer graphene systems with four different impurity patterns. In all cases, the impurity density was set to $N_D = 2.0 \times 10^{11} \text{ cm}^{-2}$, and the parameters M and α were varied. A smaller value of α results in tBLG with flatter bands, which can improve the screening of potential fluctuations caused by impurities. By changing the total number of layers M , it becomes possible to examine how the influence of impurities extends to the upper layers and affects their transport properties.

Figure 5.3 shows the electron and hole densities of each layer in the system with $M = 12$ and $\alpha = 0.1, 0.2, 0.5$, and 0.8 . A significantly higher carrier concentration appears in the first two layers, which correspond to the twisted bilayer graphene. The remaining turbostratic layers exhibit a sharp decrease in both electron and hole densities from the third layer onward. Since a lower Fermi velocity increases the density of states near the Dirac point, these layers induce a higher carrier concentration

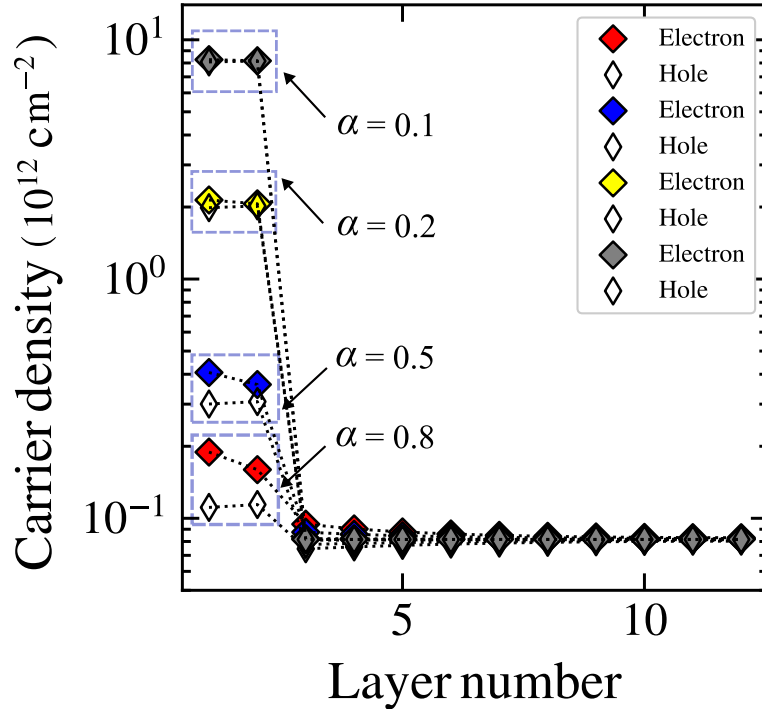


Figure 5.3: Carrier densities across layers for $M = 12$ and $\alpha = 0.1, 0.2, 0.5$, and 0.8 . The first two layers (tBLG) contain most of the carriers, while densities drop sharply from the third layer onward. Smaller α values yield higher carrier densities and stronger screening compared to larger α .

compared to the upper turbostratic layers. As a result, a larger portion of the total carriers becomes concentrated in the first two layers, which enhances the screening of the impurity-induced potential. In the upper turbostratic layers, where the density of states is smaller, fewer carriers are induced, leading to the rapid decrease in carrier density observed from the third layer onward. The effect of α is also evident. Larger values of α correspond to a higher Fermi velocity, which reduces the density of states in the twisted bilayer graphene. Cases with small α (0.1 or 0.2) induce more carriers in the first two layers than cases with larger α (0.5 or 0.8). Therefore, small α leads to higher carrier densities in the first two layers and stronger screening of the impurity-induced potential, while larger α weakens this screening effect.

Figure 5.4 illustrates the spatial distribution of the electrostatic potential in the first graphene layer for systems with $M = 12$ and parameter α of 0.1, 0.5, 0.8, and 1.0. Here, a clear trend is observed: systems with smaller α exhibit noticeably

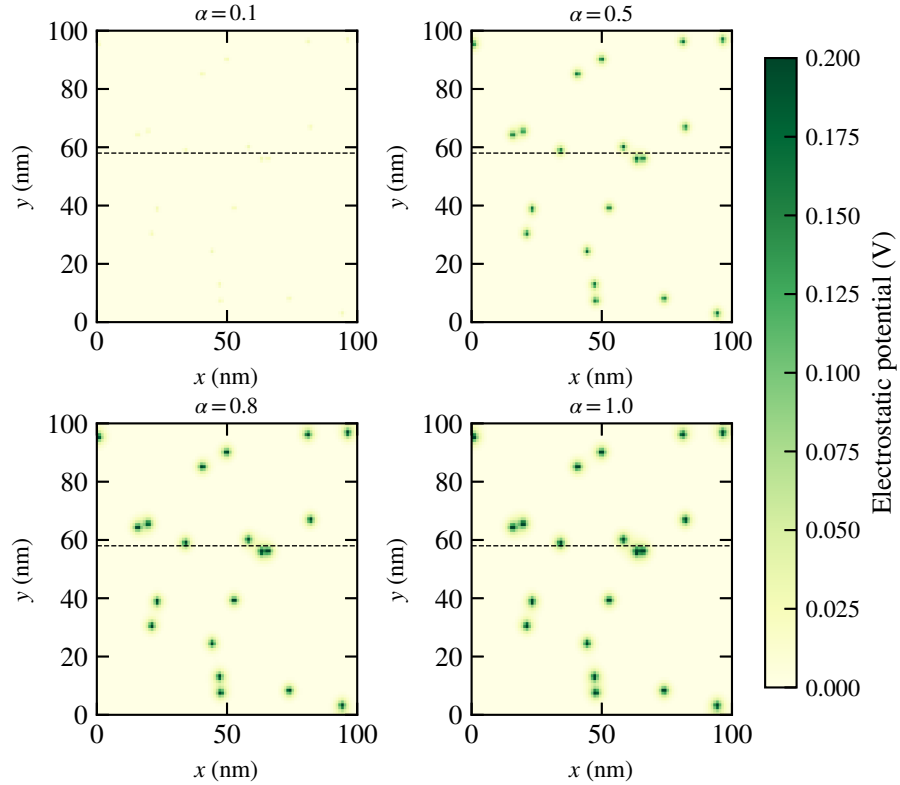


Figure 5.4: Spatial distribution of the electrostatic potential in the first graphene layer for systems with $M = 12$ and $\alpha = 0.1, 0.5, 0.8$, and 1.0 . Smaller α values lead to stronger carrier accumulation near the interface, resulting in enhanced screening of impurity-induced fluctuations and a smoother potential landscape. In contrast, systems with larger α exhibit more pronounced potential peaks, visible as bright regions in the plot, indicating weaker screening capability.

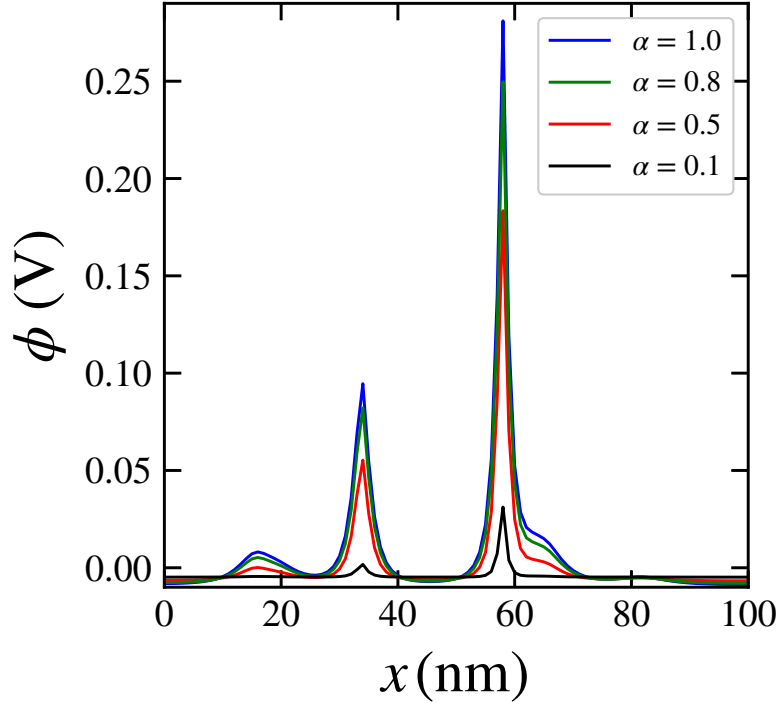


Figure 5.5: Cross-sectional potential profiles extracted along the dashed lines in Fig. 5.4, taken at a fixed position of $y = 58$ nm. The $\alpha = 0.1$ system shows the smallest fluctuation amplitude, while the $\alpha = 1.0$ system exhibits strong peaks. This comparison reflects the dependence of screening effectiveness on the parameter α .

reduced potential fluctuations across the surface, whereas systems with larger α show more pronounced potential peaks. As mentioned earlier, a smaller α corresponds to higher carrier density in the tBLG region. Thus, the accumulation of carriers in these bottom layers of systems with smaller α alters the overall potential landscape of the layer to a smoother profile compared to the systems with larger α .

For a more quantitative evaluation of these fluctuations, Fig. 5.5 presents the one-dimensional potential profiles extracted along the dashed lines shown in Fig. 5.4. The slice is taken at a fixed y coordinate of 58 nm for all four α values for direct visual comparison. The contrast in the magnitude of fluctuations becomes more apparent here, where the system with $\alpha = 0.1$ demonstrates the smallest potential variations, whereas the $\alpha = 1.0$ case exhibits pronounced peaks and dips, reflecting a much weaker screening effect. These results indicate that the electronic properties introduced by the tBLG layers directly affect the strength of screening and the resulting potential profile.

In Fig. 5.5, it is shown that for $\alpha = 1$, the peak electrostatic potential reaches

approximately 0.28 V, whereas for $\alpha = 0.1$ it decreases drastically to 0.031 V. Thus, the peak amplitude for $\alpha = 1$ is nearly nine times larger than that for $\alpha = 0.1$. This reduction clearly demonstrates that using a tBLG with a smaller α significantly mitigates impurity-induced potential fluctuations by providing stronger electrostatic screening.

5.3.2 Transport Properties

Figure 5.6 shows the carrier mobility versus the velocity ratio under an electric field of 10^2 V/cm for a system with $M = 12$ layers, where each mobility value is averaged over four different impurity configurations. The first two layers correspond to the tBLG segment, whereas the remaining ten layers correspond to turbostratic graphene.

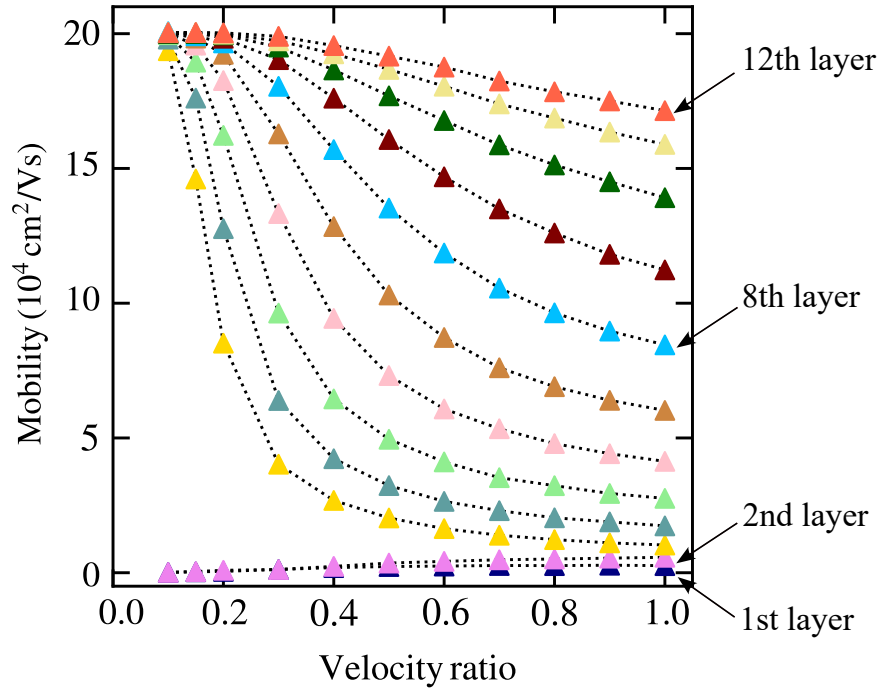


Figure 5.6: Carrier mobility of a 12-layer multilayer graphene system versus the velocity ratio α . In the tBLG segment, mobility increases with α . In contrast, mobility in the turbostratic layers decreases as α increases, showing that weaker screening at larger α allows impurity-induced potential fluctuations to influence the turbostratic layers more and consequently, reduce the mobility of the upper turbostratic layers. For the systems with small values of α , it is observed that the turbostratic layer closer to the substrate reaches the phonon-limited mobility, showcasing the effectiveness of screening of potential fluctuations by tBLG layers.

For the tBLG layers (layers 1 and 2), the mobility increases monotonically with α , reflecting the direct dependence of the mobility on the Fermi velocity. For example, in the first layer of a system with $\alpha = 0.1$, $\mu_1 = 1.26 \times 10^2 \text{ cm}^2/\text{Vs}$, whereas for a system with $\alpha = 1.0$, the mobility increases to $\mu_1 = 2.63 \times 10^3 \text{ cm}^2/\text{Vs}$. On the other hand, the mobility of carriers in the turbostratic layers decreases with increasing α . The most drastic decrease occurs in the third layer, where for $\alpha = 0.1$, $\mu_3 = 1.94 \times 10^5 \text{ cm}^2/\text{Vs}$, which is nearly equal to the phonon-limited mobility, whereas for $\alpha = 1.0$, the mobility decreases to $\mu_3 = 1.03 \times 10^4 \text{ cm}^2/\text{Vs}$. Even in the twelfth layer, from $\alpha = 0.1$ to $\alpha = 1.0$, there is a decrease of mobility from $\mu_{12} = 2.00 \times 10^5 \text{ cm}^2/\text{Vs}$ to $\mu_{12} = 1.71 \times 10^5 \text{ cm}^2/\text{Vs}$. Nevertheless, systems with smaller α reach mobility values closer to the phonon-limited value at much lower layers within the turbostratic segment. Overall, this demonstrates that for sufficiently small α , the tBLG efficiently screens the impurity-induced potential fluctuations, which are one of the dominant scattering sources in graphene transport, and thus allows the upper turbostratic layers to approach the phonon-limited mobility of pristine graphene at a layer much closer to the substrate.

While using tBLG with a smaller Fermi velocity ratio α effectively enhances the screening of impurity-induced potential fluctuations, it also reduces the carrier velocity within the tBLG layers, thereby reducing their transport properties. Thus, the overall behavior of the multilayer system is governed by two opposing effects: (i) slower carrier motion within the tBLG layers, and (ii) enhanced potential screening that benefits the upper turbostratic layers.

Figure 5.7 (a) presents the averaged mobility, μ_{ave} , of the entire multilayer system as a function of the velocity ratio α for systems with $M = 4, 6, 8$, and 12. Each mobility value is averaged over four different impurity configurations. For small α , μ_{ave} increases with increasing α and reaches a maximum around $\alpha \approx 0.7$, after which it begins to decrease. When α is small, carriers in the tBLG layers have significantly reduced Fermi velocities and exhibit higher carrier densities compared to the upper turbostratic layers. Even though the tBLG layers in systems with small α efficiently screen impurity-induced potential fluctuations and therefore support improved mobility in the upper layers, the slow carrier motion within the tBLG dominates the total transport properties, leading to a lower overall mobility of the system. As α increases, the situation reverses: the Fermi velocity in the tBLG layers becomes larger, and the carrier density within the tBLG decreases. For instance, in the $M = 12$ system, approximately 95% of the carriers reside in the tBLG layers at $\alpha = 0.1$, whereas this fraction decreases to 45% at $\alpha = 0.5$ and 18% at $\alpha = 1.0$. This redistribution of carriers reduces the dominance of the tBLG contribution to

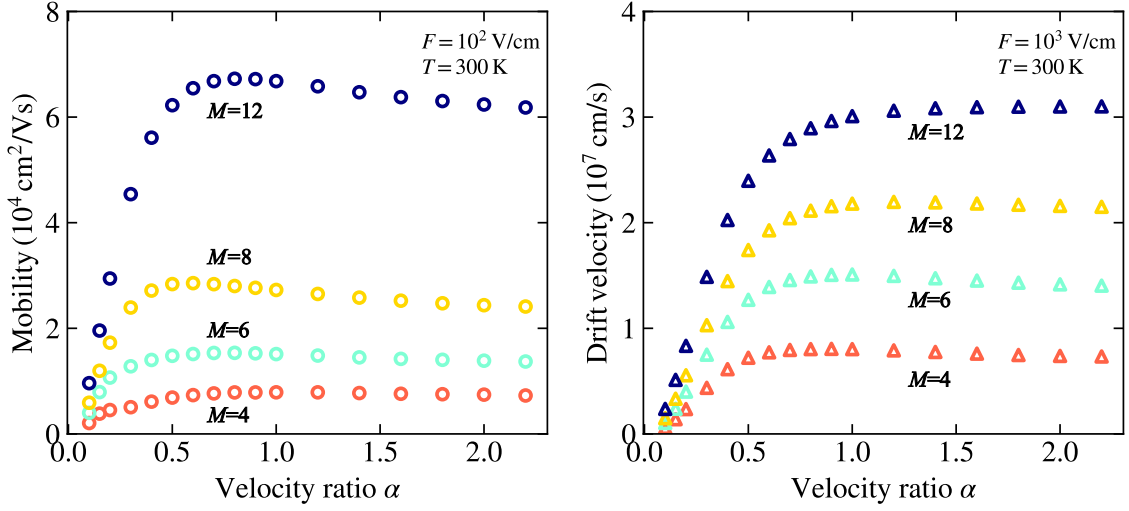


Figure 5.7: (a) Averaged mobility μ_{ave} of the entire system versus the velocity ratio α for $M = 4, 6, 8$, and 12 , averaged over four impurity configurations. For small α , μ_{ave} increases as the carriers in the tBLG layers gain higher Fermi velocities. The mobility reaches its maximum around $\alpha \approx 0.7$, after which it decreases because the reduced screening from the tBLG layers becomes insufficient to suppress impurity-induced potential fluctuations in the upper layers. (b) Averaged drift velocity $v_{x,\text{ave}}$ of the entire system under an electric field of 10^3 V/cm . For $\alpha < 1$, the strong electric field drives carrier transport and the influence of weaker screening is minor, resulting in an increase in $v_{x,\text{ave}}$ with α . When $\alpha > 1$, carriers in the first two layers experience stronger impurity scattering and provide little screening to the upper layers, leading to a reduction in the overall drift velocity of the system.

the total mobility, allowing the upper layers to play a more significant role in the transport.

A reduction in screening by the first two layers would intuitively suggest a decrease in the overall mobility. However, the screening is shared among all layers of the system, so the reduction in screening by the tBLG layers is partially compensated by the turbostratic layers above. Consequently, as α increases, the benefit of faster carrier motion in the tBLG outweighs the moderate reduction in screening, leading to an overall increase in μ_{ave} with α for $\alpha \lesssim 0.7$.

Figure 5.7 (b) shows the dependence of the overall drift velocity, $v_{x,\text{ave}}$, on the velocity-ratio parameter α under an applied electric field of 10^3 V/cm . For $\alpha < 1.0$, the average drift velocity increases as α becomes larger. In this regime, a larger α causes a greater portion of carriers to reside in the graphene layers located above the tBLG region. Although these carriers are subject to stronger potential fluctuations, the relatively strong electric field dominates their motion, and the reduced screening plays a secondary role. As a result, the drift velocity of the entire system increases.

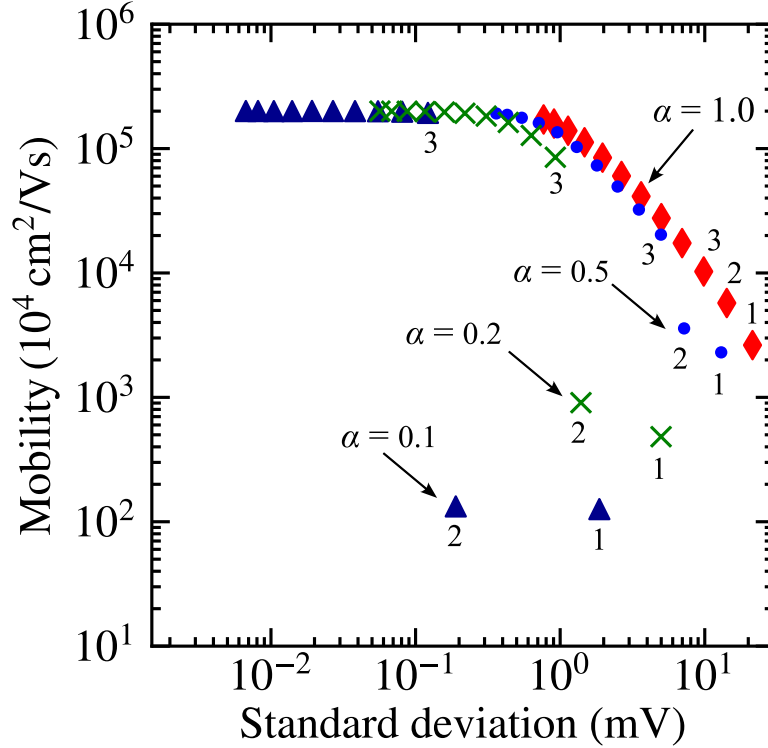


Figure 5.8: Relationship between the standard deviation of the electrostatic potential and the mobility of each graphene layer for $M = 12$ and $\alpha = 0.1, 0.2, 0.5, 1.0$. Layers closer to the substrate experience stronger impurity-induced fluctuations and exhibit lower mobility, while screening from the lower layers reduces the potential variation and mobility approaches the phonon-limited value. A sharp mobility increase between the 2nd and 3rd layers is observed when α is small, marking the transition from the tBLG segment to the turbostratic layers due to stronger screening.

When $\alpha > 1.0$, however, the trend reverses, and $v_{x,\text{ave}}$ gradually decreases. In this regime, the carriers in the first two layers have higher Fermi velocities than those in the upper layers, yet they are more strongly affected by impurity-induced scattering, which suppresses their drift velocities. For example, in the 12-layer system, the drift velocities of the first layer for $\alpha = 1.0, 1.6$, and 2.2 are 2.65×10^6 cm/s, 2.13×10^6 cm/s, and 1.73×10^6 cm/s, respectively, and the drift velocities of the second layer for the same cases are 5.56×10^6 cm/s, 5.47×10^6 cm/s, and 4.70×10^6 cm/s, respectively. Even though α increases, the drift velocity of these layers decreases due to the enhanced scattering from impurities. Because the carriers in the first two layers also provide limited screening to the upper layers, the overall drift velocity of the multilayer graphene system becomes smaller.

Figure 5.8 presents the relationship between the standard deviation of the elec-

trostatic potential and the carrier mobility for each graphene layer in systems with $M = 12$ and velocity ratios $\alpha = 0.1, 0.2, 0.5$, and 1.0 . In each system, the rightmost data point corresponds to the layer nearest to the substrate, where impurity-induced potential fluctuations are the strongest and mobility is the lowest. As the layer index increases, screening from the lower graphene layers reduces the standard deviation, which allows the mobility to increase. When the standard deviation decreases to approximately 1 mV, mobility becomes unaffected by the remaining potential fluctuations and converges to the phonon-limited value, μ_{phonon} . Under these conditions, transport is governed predominantly by intrinsic phonon scattering rather than by impurity-induced potential variations. Since the upper turbostratic graphene layers have the same Fermi velocity for all values of α , the convergence value of mobility is identical among all four systems. In contrast, mobility behavior changes at the interface between the tBLG segment and the upper turbostratic layers, where the Fermi velocity shifts from its reduced value in the tBLG to the higher value in the turbostratic graphene. A sharp increase in mobility between the 2nd and 3rd layers marks the transition from the tBLG segment to the upper turbostratic graphene. As α decreases, the tBLG layers provide stronger screening, resulting in smaller standard deviations in the first turbostratic layer. For $\alpha = 0.1$, this screening is sufficiently strong that the mobility of the 3rd layer already reaches μ_{phonon} , even though it is only a few layers above the substrate.

These observations suggest that the choice of α plays an important role in determining the transport characteristics of the multilayer system, and different values of α can shift the balance between screening efficiency and carrier mobility. Therefore, the influence of α should be considered carefully when evaluating or designing multilayer graphene systems incorporating tBLG.

Based on the results shown in Figs. 5.6 and 5.8, a device concept emerges in which the multilayer graphene stack is functionally divided into two distinct roles: the tBLG layers act as a screening buffer that suppresses impurity-induced potential fluctuations, while the upper turbostratic layers serve as the primary transport channel carrying most of the electrical current. In this configuration, selecting a small velocity ratio ($\alpha \ll 1$) for the tBLG layers is advantageous because it increases the density of states and strengthens electrostatic screening. Consequently, the turbostratic layers above can approach phonon-limited mobility within only a few layers, as illustrated for $\alpha = 0.1$, where the third layer already exhibits nearly intrinsic transport characteristics.

These findings imply that, in principle, even a minimal stack composed of a tBLG and a monolayer graphene ($M = 3$) could operate as a high-mobility structure.

Achieving such small values of α , however, requires rotational alignment extremely close to the magic angle, where α becomes highly sensitive to the twist angle, making fabrication challenging. If the overall layer count is not a strict constraint, a more practical approach is to employ tBLG with a moderately larger velocity ratio and compensate by adding several turbostratic layers. The transport calculations indicate that, for example, a configuration with four turbostratic layers atop a tBLG buffer with $\alpha \approx 0.2$ can attain comparable mobility performance while easing the requirements on twist-angle precision. In this sense, the velocity ratio α acts as a design parameter that balances screening effectiveness and device thickness, which needs to be considered in device fabrication.

5.4 Summary

In this chapter, multilayer graphene systems incorporating twisted bilayer graphene (tBLG) were investigated. In tBLG, the Fermi velocity is suppressed depending on the twist angle, and in this study, the extent of the suppression is represented by the parameter α , defined as the ratio of the Fermi velocity in tBLG to that of pristine graphene. A smaller value of α indicates stronger suppression and lower carrier velocity in the tBLG region. Based on this characteristic, the possibility of using tBLG as a means to improve mobility in turbostratic multilayer graphene systems were investigated. Here, electrostatic potential profiles of the multilayer structures were calculated and Monte Carlo transport simulations were performed to evaluate the transport behavior of these multilayer systems. Since velocity reduction increases the density of states, the tBLG layers attract more carriers and become effective in screening substrate-induced impurity potentials. As a result, the electrostatic potential experienced by the upper graphene layers becomes significantly smoother, allowing carriers there to move under reduced impurity scattering conditions. The mobility behavior is determined by the competition between two effects: (i) enhanced screening due to increased carrier density in tBLG, and (ii) reduced carrier velocity within the tBLG region. For small α , the upper turbostratic layers experience greatly weakened impurity scattering, and their mobility rapidly approaches the phonon-limited value μ_{phonon} . In systems with $M = 12$, for example, the 3rd layer already reaches μ_{phonon} when $\alpha = 0.1$, only a few layers above the substrate. In contrast, at $\alpha = 1.0$, screening is weaker, the potential fluctuations remain large, and impurity scattering continues to limit mobility over more layers. When the mobility is evaluated for the entire multilayer stack, this trade-off pro-

duces a non-monotonic dependence on α , with the highest overall mobility occurring near $\alpha \approx 0.7$, where screening and carrier velocity are optimally balanced. These results demonstrate that the reduced Fermi velocity inherent to tBLG can serve as a functional design element: tBLG can be used as an intentional screening layer to mitigate impurity-induced potential fluctuations and enable the upper layers to attain mobility values close to the intrinsic phonon-limited regime.

Chapter 6

Conclusion

This thesis has presented a theoretical and numerical investigation into the carrier transport properties of multilayer graphene systems. By utilizing a self-consistent electrostatic calculation combined with an ensemble Monte Carlo simulation, this work has shown the distinct roles and combined effects of substrate-induced impurity scattering, edge scattering, and the structural modifications introduced by twisted bilayer graphene (tBLG). The findings provide a framework for understanding how electrostatic screening and structural design can be leveraged to optimize the performance of graphene-based electronic devices.

The first part of this research focused on the limitations imposed by charged impurities on the substrate. The results demonstrated that the multilayer structure serves as an effective electrostatic shield. The potential analysis revealed that impurities generate strong, localized potential peaks in the graphene layers closest to the substrate, which decay exponentially with the layer index. This behavior confirmed that the lower graphene layers effectively screen the impurity potential, creating a significantly smoother potential landscape for the upper layers. Consequently, transport calculations indicated a recovery of mobility and drift velocity in the upper layers, where transport characteristics converge toward the intrinsic phonon-limited regime of impurity-free graphene. The development of an analytical model establishing that mobility is inversely proportional to the potential fluctuations provided a theoretical explanation for this phenomenon, describing the general layer-by-layer mobility trends observed in the Monte Carlo simulations.

While the screening of impurity potentials significantly improves transport in upper layers, this study also took into account the impact of geometrical limitations. In particular, edge scattering was integrated into the analysis and modeled using the parameter r , which quantifies the degree of scattering randomness. Since edge

scattering is an intrinsic factor in graphene layers, it influences carrier transport throughout the stack. Crucially, as the impurity potential is screened in the upper layers, the relative dominance of edge scattering increases. This leads to a scenario where, even in the absence of significant impurity scattering, the carrier mobility is restricted by diffusive collisions at the boundaries. The empirical model derived in this study successfully captures this behavior, demonstrating a clear correlation between the randomness parameter and the resulting mobility. Notably, while boundary collisions impose a constraint on transport properties, the multilayer stacking remains highly advantageous because it effectively screens the substrate impurities, enabling the upper layers to reach higher mobilities where carriers move with fewer impediments.

Finally, the research explored the functional engineering of multilayer systems by incorporating twisted bilayer graphene (tBLG). This study specifically examined the impact of the Fermi velocity suppression, characterized by the parameter α . The reduction of Fermi velocity in tBLG layers results in an increased density of states, which enhances the screening capacity of the tBLG layers. The simulations demonstrated that these layers effectively suppress substrate-induced potential fluctuations, allowing the upper turbostratic layers to achieve phonon-limited mobility with fewer stacked layers. At the same time, while strong velocity suppression (small α) maximizes screening, it directly limits the carrier velocity within the tBLG layers themselves. The evaluation of the total mobility of the stack revealed a non-monotonic dependence on parameter α , with an optimal balance found near $\alpha \approx 0.7$. This finding suggests that tBLG can be utilized not merely as a simple screening layer, but as an intentional design element to mitigate impurity scattering while maintaining high overall transport properties.

Prospective extensions of this work could involve introducing a device structure with defined source and drain regions to quantitatively assess performance. Additionally, the simulation framework could be further extended by expanding on disorder configurations, such as introducing clustered impurity distributions and defects.

In summary, this thesis establishes that while substrate impurities are a major obstacle for carrier transport in graphene, their effects can be effectively mitigated by using turbostratic structures. Additionally, intrinsic factors such as edge scattering are also important to consider. Finally, using tBLG systems is effective in improving the carrier mobility of the system. Collectively, these insights provide essential guidelines for the design of high-mobility multilayer graphene devices, showing that high transport properties are achievable through the careful balancing of layer count,

boundary quality, twist-angle engineering, and stacking configuration.

Research Achievements

Journal Papers

1. R. Negishi, K. Yamamoto, H. Tanaka, Seyed Ali Mojtahedzadeh, N. Mori, Y. Kobayashi, “Crossover point of the field effect transistor and interconnect applications in turbostratic multilayer graphene nanoribbon channel,” *Scientific Reports* **11**, 10206 (1-11), 2021.
2. Seyed Ali Mojtahedzadeh, H. Tanaka, and N. Mori, “Monte Carlo simulation of mobility enhancement in multilayer graphene with turbostratic structure,” *Japanese Journal of Applied Physics* **63**, 031004 (1-10), 2024.
3. Seyed Ali Mojtahedzadeh, H. Tanaka, and N. Mori, “Carrier transport simulations in twisted bilayer and turbostratic multilayer graphene systems,” *Japanese Journal of Applied Physics* **63**, 05SP09 (1-7), 2024.
4. Seyed Ali Mojtahedzadeh, H. Tanaka, and N. Mori, “Monte Carlo simulation of edge scattering effects on carrier mobility in turbostratic multilayer graphene,” *Japanese Journal of Applied Physics* **64**, 121001 (1-9), 2025.

International Conferences

1. Seyed Ali Mojtahedzadeh and N. Mori, “Monte Carlo simulation of electron transport in multilayer graphene,” International Conference on Electron Dynamics in Semiconductors, Optoelectronics and Nanostructures, July 15, 2019, Nara, Japan.
2. Seyed Ali Mojtahedzadeh and N. Mori, “Effects of impurity concentration on electron transport in multilayer graphene,” International Symposium on Hybrid Quantum Systems 2019, December 2, 2019, Matsue, Japan.
3. Seyed Ali Mojtahedzadeh, H. Tanaka, and N. Mori, “Carrier transport simulations in twisted bilayer and turbostratic multilayer graphene systems,” The 36th International Microprocesses and Nanotechnology Conference, November 14-17, 2023, Hokkaido, Japan.

Domestic Conferences

1. Seyed Ali Mojtahedzadeh, 田中 一, 森 伸也, “モンテカルロ法を用いた多層

グラフェンの電子輸送解析に関する研究”，新学術領域「ハイブリッド量子科学」第9回領域会議, 2019年8月8日, 北陸先端科学技術大学院大学, 石川県.

2. 森 伸也, 橋本 風渡, Seyed Ali Mojtahedzadeh, 田中 一, “ハイブリッド量子デバイスのシミュレーション”, 新学術領域「ハイブリッド量子科学」第10回領域会議, 2020年2月26日, 理化学研究所, 埼玉県.
3. 根岸 良太, 周 詠凱, 魏 超鵬, 小川 友以, 谷保 芳孝, 赤堀 誠志, Seyed Ali Mojtahedzadeh, 森 伸也, 増田 健作, 橋本 克之, 平山 祥郎, 小林 慶裕, “乱層積層した多層グラフェンにおけるキャリア散乱長の向上”, 第67回応用物理学会春季学術講演会, 2020年3月12日, 上智大学, 東京都.

References

- [1] P. R. Wallace. The band theory of graphite. *Physical Review*, 71(9):622–634, 1947.
- [2] A. K. Geim and K. S. Novoselov. The rise of graphene. *Nature Materials*, 6:183–191, 2007.
- [3] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, and A. A. Firsov. Electric field effect in atomically thin carbon films. *Science*, 306:666–669, 2004.
- [4] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, M. I. Katsnelson, I. V. Grigorieva, S. V. Dubonos, and A. A. Firsov. Two-dimensional gas of massless Dirac fermions in graphene. *Nature*, 438:197–200, 2005.
- [5] Y. Zhang, Y.-W. Tan, H. L. Stormer, and P. Kim. Experimental observation of the quantum Hall effect and Berry’s phase in graphene. *Nature*, 438:201–204, 2005.
- [6] K. S. Novoselov, D. Jiang, F. Schedin, T. J. Booth, V. V. Khotkevich, S. V. Morozov, and A. K. Geim. Two-dimensional atomic crystals. *Proceedings of the National Academy of Sciences*, 102(30):10451–10453, 2005.
- [7] M. I. Katsnelson. Nonlinear screening of charge impurities in graphene. *Physical Review B*, 74:201401(R), 2006.
- [8] M. I. Katsnelson, K. S. Novoselov, and A. K. Geim. Chiral tunnelling and the Klein paradox in graphene. *Nature Physics*, 2:620–625, 2006.
- [9] A. C. Ferrari, J. C. Meyer, V. Scardaci, C. Casiraghi, M. Lazzeri, F. Mauri, S. Piscanec, D. Jiang, K. S. Novoselov, S. Roth, and A. K. Geim. Raman spectrum of graphene and graphene layers. *Physical Review Letters*, 97:187401, 2006.

- [10] E. H. Hwang and S. Das Sarma. Dielectric function, screening, and plasmons in two-dimensional graphene. *Physical Review B*, 75:205418, 2007.
- [11] K. F. Mak, M. Y. Sfeir, Y. Wu, C. H. Lui, J. A. Misewich, and T. F. Heinz. Measurement of the optical conductivity of graphene. *Physical Review Letters*, 101:196405, 2008.
- [12] J. S. Bunch, A. M. van der Zande, S. S. Verbridge, I. W. Frank, D. M. Tanenbaum, J. M. Parpia, H. G. Craighead, and P. L. McEuen. Electromechanical resonators from graphene sheets. *Science*, 315:490–493, 2007.
- [13] C. Lee, X. Wei, J. W. Kysar, and J. Hone. Measurement of the elastic properties and intrinsic strength of monolayer graphene. *Science*, 321:385–388, 2008.
- [14] R. R. Nair, P. Blake, A. N. Grigorenko, K. S. Novoselov, T. J. Booth, T. Stauber, N. M. R. Peres, and A. K. Geim. Fine structure constant defines visual transparency of graphene. *Science*, 320:1308, 2008.
- [15] X. Du, I. Skachko, A. Barker, and E. Y. Andrei. Approaching ballistic transport in suspended graphene. *Nature Nanotechnology*, 3:491–495, 2008.
- [16] A. A. Balandin, S. Ghosh, W. Bao, I. Calizo, D. Teweldebrhan, F. Miao, and C. N. Lau. Superior thermal conductivity of single-layer graphene. *Nano Letters*, 8:902–907, 2008.
- [17] L. Liao, Y.-C. Lin, M. Bao, R. Cheng, J. Bai, Y. Liu, Y. Qu, K. L. Wang, Y. Huang, and X. Duan. High-speed graphene transistors with a self-aligned nanowire gate. *Nature*, 467:305–308, 2010.
- [18] E. Hendry, P. J. Hale, J. Moger, A. K. Savchenko, and S. A. Mikhailov. Coherent nonlinear optical response of graphene. *Physical Review Letters*, 105:097401, 2010.
- [19] N. Levy, S. A. Burke, K. L. Meaker, M. Panlasigui, A. Zettl, F. Guinea, A. H. Castro Neto, and M. F. Crommie. Strain-induced pseudo-magnetic fields greater than 300 tesla in graphene nanorubbles. *Science*, 329:544–547, 2010.
- [20] S. V. Morozov, K. S. Novoselov, M. I. Katsnelson, F. Schedin, D. C. Elias, J. A. Jaszczak, and A. K. Geim. Giant intrinsic carrier mobilities in graphene and its bilayer. *Physical Review Letters*, 100:016602, 2008.

- [21] K. I. Bolotin, K. J. Sikes, Z. Jiang, M. Klima, G. Fudenberg, J. Hone, P. Kim, and H. L. Stormer. Ultrahigh electron mobility in suspended graphene. *Solid State Communications*, 146:351–355, 2008.
- [22] J.-H. Chen, C. Jang, S. Xiao, M. Ishigami, and M. S. Fuhrer. Intrinsic and extrinsic performance limits of graphene devices on SiO₂. *Nature Nanotechnology*, 3:206–209, 2008.
- [23] A. H. Castro Neto, F. Guinea, N. M. R. Peres, K. S. Novoselov, and A. K. Geim. The electronic properties of graphene. *Reviews of Modern Physics*, 81:109–162, 2009.
- [24] S. Das Sarma, S. Adam, E. H. Hwang, and E. Rossi. Electronic transport in two-dimensional graphene. *Reviews of Modern Physics*, 83:407–470, 2011.
- [25] Y.-M. Lin, C. Dimitrakopoulos, K. A. Jenkins, D. B. Farmer, H.-Y. Chiu, A. Grill, and P. Avouris. 100-GHz transistors from wafer-scale epitaxial graphene. *Science*, 327:662, 2010.
- [26] P. Avouris. Graphene: Electronic and photonic properties and devices. *Nano Letters*, 10(11):4285–4294, 2010.
- [27] Y. Singh and H. Subbaraman. Recent advances in graphene-enabled silicon-based high-speed optoelectronic devices—a review. *Photonics*, 10(12):1292, 2023.
- [28] C. Berger, Z. Song, T. Li, X. Li, A. Y. Ogbazghi, R. Feng, Z. Dai, A. N. Marchenkov, E. H. Conrad, P. N. First, and W. A. de Heer. Ultrathin epitaxial graphite: 2D electron gas properties and a route toward graphene-based nanoelectronics. *The Journal of Physical Chemistry B*, 108:19912–19916, 2004.
- [29] C. Virojanadara, M. Syväjarvi, R. Yakimova, and L. I. Johansson. Homogeneous large-area graphene layer growth on 6H-SiC(0001). *Physical Review B*, 78:245403, 2008.
- [30] A. L. Vázquez de Parga, F. Calleja, B. Borca, M. C. G. Passeggi Jr., J. J. Hinarejos, F. Guinea, and R. Miranda. Periodically rippled graphene: Growth and spatially resolved electronic structure. *Physical Review Letters*, 100:056807, 2008.

- [31] A. Reina, X. Jia, J. Ho, D. Nezich, H. Son, V. Bulovic, M. S. Dresselhaus, and J. Kong. Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition. *Nano Letters*, 9:30–35, 2009.
- [32] K. S. Kim, Y. Zhao, H. Jang, S. Y. Lee, J. M. Kim, K. S. Kim, J.-H. Ahn, P. Kim, J.-Y. Choi, and B. H. Hong. Large-scale pattern growth of graphene films for stretchable transparent electrodes. *Nature*, 457:706–710, 2009.
- [33] H. Ago, Y. Ito, N. Mizuta, K. Yoshida, B. Hu, C. M. Orofeo, M. Tsuji, K. Ikeda, and S. Mizuno. Epitaxial chemical vapor deposition growth of single-layer graphene over cobalt film crystallized on sapphire. *ACS Nano*, 4(12):7407–7414, 2010.
- [34] S. Adam, E. H. Hwang, V. M. Galitski, and S. Das Sarma. A self-consistent theory for graphene transport. *Proceedings of the National Academy of Sciences*, 104:18392–18397, 2007.
- [35] J.-H. Chen, C. Jang, S. Adam, M. S. Fuhrer, E. D. Williams, and M. Ishigami. Charged-impurity scattering in graphene. *Nature Physics*, 4:377–381, 2008.
- [36] E. H. Hwang and S. D. Sarma. Acoustic phonon scattering limited carrier mobility in two-dimensional extrinsic graphene. *Physical Review B*, 77:115449, 2008.
- [37] D. K. Efetov and P. Kim. Controlling electron–phonon interactions in graphene at ultrahigh mobility. *Physical Review Letters*, 105:256805, 2010.
- [38] K. I. Bolotin, K. J. Sikes, J. Hone, H. L. Stormer, and P. Kim. Temperature-dependent transport in suspended graphene. *Physical Review Letters*, 101:096802, 2008.
- [39] Y. Zhang, V. W. Brar, C. Girit, A. Zettl, and M. F. Crommie. Origin of spatial charge inhomogeneity in graphene. *Nature Physics*, 5:722–726, 2009.
- [40] E. H. Hwang, S. Adam, and S. Das Sarma. Carrier transport in two-dimensional graphene layers. *Physical Review Letters*, 98:186806, 2007.
- [41] J. H. Gosling, O. Makarovskiy, F. Wang, N. D. Cottam, M. T. Greenaway, A. Patané, R. D. Wildman, C. J. Tuck, L. Turyanska, and T. M. Fromhold. Universal mobility characteristics of graphene originating from charge scattering by ionised impurities. *Communications Physics*, 4:30, 2021.

- [42] L. Turyanska, O. Makarovsky, L. Eaves, A. Patané, and N. Mori. Mobility enhancement of cvd graphene by spatially correlated charges. *2D Materials*, 4:025026, 2017.
- [43] O. Makarovsky, L. Turyanska, N. Mori, M. Greenaway, L. Eaves, A. Patané, M. Fromhold, S. Lara-Avila, S. Kubatkin, and R Yakimova. Enhancing optoelectronic properties of SiC-grown graphene by a surface layer of colloidal quantum dots. *2D Materials*, 4:031001, 2017.
- [44] W. Gannett, W. Regan, K. Watanabe, T. Taniguchi, M. F. Crommie, and A. Zettl. Boron nitride substrates for high mobility chemical vapor deposited graphene. *Applied Physics Letters*, 98:242105, 2011.
- [45] H. Hirai, H. Tsuchiya, Y. Kamakura, N. Mori, and M. Ogawa. Electron mobility calculation for graphene on substrates. *Journal of Applied Physics*, 116:083703, 2014.
- [46] R. Negishi, K. Yamamoto, H. Kitakawa, M. Fukumori, H. Tanaka, T. Ogawa, and Y. Kobayashi. Synthesis of very narrow multilayer graphene nanoribbon with turbostratic stacking. *Applied Physics Letters*, 110:201901, 2017.
- [47] C. Wei, R. Negishi, Y. Ogawa, M. Akabori, Y. Taniyasu, and Y. Kobayashi. Turbostratic multilayer graphene synthesis on CVD graphene template toward improving electrical performance. *Japanese Journal of Applied Physics*, 58:SIIB04, 2019.
- [48] R. Negishi, C. Wei, Y. Yao, Y. Ogawa, M. Akabori, Y. Kanai, K. Matsumoto, Y. Taniyasu, and Y. Kobayashi. Turbostratic stacking effect in multilayer graphene on the electrical transport properties. *Physica Status Solidi (b)*, 257:1900437, 2019.
- [49] R. Negishi, K. Yamamoto, H. Tanaka, S. A. Mojtahedzadeh, N. Mori, and Y. Kobayashi. Crossover point of the field effect transistor and interconnect applications in turbostratic multilayer graphene nanoribbon channel. *Scientific Reports*, 11:10206, 2021.
- [50] J. Hass, F. Varchon, J. E. Millán-Otoya, M. Sprinkle, N. Sharma, W. A. de Heer, C. Berger, P. N. First, L. Magaud, and E. H. Conrad. Why multilayer graphene on 4H-SiC(000 $\bar{1}$) behaves like a single sheet of graphene. *Physical Review Letters*, 100(12):125504, 2008.

- [51] J. Hass, W. A. de Heer, and E. H. Conrad. The growth and morphology of epitaxial multilayer graphene. *Journal of Physics: Condensed Matter*, 20:323202, 2008.
- [52] C. K. Birdsall and D. Fuss. Clouds-in-clouds, clouds-in-cells physics for many-body plasma simulation. *Journal of Computational Physics*, 3:494–511, 1969.
- [53] N. W. Ashcroft and N. D. Mermin. *Solid State Physics*. Cengage Learning, 1976.
- [54] M. Lundstrom. *Fundamentals of Carrier Transport*. Cambridge University Press, 2000.
- [55] C. Moglestue. *Monte Carlo Simulation of Semiconductor Devices*. Chapman & Hall, 1993.
- [56] C. Jacoboni and L. Reggiani. The Monte Carlo method for the solution of charge transport in semiconductors with applications to covalent materials. *Reviews of Modern Physics*, 55:645, 1983.
- [57] H.D. Rees. Calculation of distribution functions by exploiting the stability of the steady state. *Journal of Physics and Chemistry of Solids*, 30:643, 1969.
- [58] R. S. Shishir and D. K. Ferry. Velocity saturation in intrinsic graphene. *Journal of Physics: Condensed Matter*, 21(34):344201, 2009.
- [59] J. Martin, N. Akerman, G. Ulbricht, T. Lohmann, J. H. Smet, K. von Klitzing, and A. Yacoby. Observation of electron–hole puddles in graphene using a scanning single-electron transistor. *Nature Physics*, 4:144–148, 2008.
- [60] E. Rossi and S. Das Sarma. Ground-state of graphene in the presence of random charged impurities. *Physical Review Letters*, 101:166803, 2008.
- [61] T. Ando. Screening effect and impurity scattering in monolayer graphene. *Journal of the Physical Society of Japan*, 75(7):074716, 2006.
- [62] S. Adam, E. H. Hwang, E. Rossi, and S. Das Sarma. Theory of charged impurity scattering in two-dimensional graphene. *Solid State Communications*, 149(27–28):1072–1079, 2009.
- [63] N. Sule, S. C. Hagness, and I. Knezevic. Clustered impurities and carrier transport in supported graphene. *Physical Review B*, 89(16):165402, 2014.

- [64] W. A. de Heer, C. Berger, X. Wu, M. Sprinkle, Y. Hu, M. Ruan, J. A. Stroscio, P. N. First, R. Haddon, B. A. Piot, C. Faugeras, M. Potemski, and J.-S. Moon. Epitaxial graphene electronic structure and transport. *Journal of Physics D: Applied Physics*, 43(37):374007, 2010.
- [65] S. Shallcross, S. Sharma, E. Kandelaki, and O. A. Pankratov. Electronic structure of turbostratic graphene. *Physical Review B*, 81:165105, 2010.
- [66] G. Trambly de Laissardière, D. Mayou, and L. Magaud. Localization of Dirac electrons in rotated graphene bilayers. *Nano Letters*, 10:804–808, 2010.
- [67] F. Guinea. Charge distribution and screening in layered graphene systems. *Physical Review B*, 75:235433, 2007.
- [68] S. S. Datta, D. R. Strachan, E. J. Mele, and A. T. Charlie Johnson. Surface potentials and layer charge distributions in few-layer graphene films. *Nano Letters*, 9(1):7–11, 2009.
- [69] C.-P. Lu, M. Rodriguez-Vega, G. Li, H.-Z. Tsai, J.-H. Chen, A. H. MacDonald, and E. Y. Andrei. Local, global, and nonlinear screening in twisted double-layer graphene. *Proceedings of the National Academy of Sciences*, 113(24):6623–6628, 2016.
- [70] H. Rokni and W. Lu. Layer-by-layer insight into electrostatic charge distribution of few-layer graphene. *Scientific Reports*, 7(1):42821, 2017.
- [71] C. Hamaguchi. *Basic Semiconductor Physics*. Springer, 2001.
- [72] T. Enoki and T. Ando. *Physics and Chemistry of Graphene*. Stanford Publishing, Singapore, 2019.
- [73] J. H. Gosling. *Electronic Transport in Graphene: 2D Materials, Composites, and 3D Printed Structures*. PhD thesis, The University of Nottingham, Nottingham, United Kingdom, 2022.
- [74] K. Nakada, M. Fujita, G. Dresselhaus, and M. S. Dresselhaus. Edge state in graphene ribbons: Nanometer size effect and edge shape dependence. *Physical Review B*, 54:17954, 1996.
- [75] L. Brey and H. A. Fertig. Electronic states of graphene nanoribbons studied with the Dirac equation. *Physical Review B*, 73:235411, 2006.

- [76] Y.-W. Son, M. L. Cohen, and S. G. Louie. Energy gaps in graphene nanoribbons. *Physical Review Letters*, 97:216803, 2006.
- [77] D. A. Areshkin, D. Gunlycke, and C. T. White. Ballistic transport in graphene nanostrips in the presence of disorder: Importance of edge effects. *Nano Letters*, 7(1):204–210, 2007.
- [78] Youngki Yoon and Jing Guo. Effect of edge roughness in graphene nanoribbon transistors. *Applied Physics Letters*, 91(7):073103, 2007.
- [79] D. Basu, M. J. Gilbert, L. F. Register, S. K. Banerjee, and A. H. MacDonald. Effect of edge roughness on electronic transport in graphene nanoribbon channel metal-oxide-semiconductor field-effect transistors. *Applied Physics Letters*, 92(4):042114, 2008.
- [80] T. Fang, A. Konar, H. Xing, and D. Jena. Mobility in semiconducting graphene nanoribbons: Phonon, impurity, and edge roughness scattering. *Physical Review B*, 78:205403, 2008.
- [81] A. Yazdanpanah, M. Pourfath, M. Fathipour, H. Kosina, and S. Selberherr. A numerical study of line-edge roughness scattering in graphene nanoribbons. *IEEE Transactions on Electron Devices*, 59(2):433–440, 2012.
- [82] K. Moors, A. Contino, M. L. Van de Put, W. G. Vandenberghe, M. V. Fischetti, W. Magnus, and B. Sorée. Theoretical study of scattering in graphene ribbons in the presence of structural and atomistic edge roughness. *Physical Review Materials*, 3:024001, 2019.
- [83] E. R. Mucciolo, A. H. Castro Neto, and C. H. Lewenkopf. Conductance quantization and transport gaps in disordered graphene nanoribbons. *Physical Review B*, 79:075407, 2009.
- [84] Y. M. Banadaki and A. Srivastava. Effect of edge roughness on static characteristics of graphene nanoribbon field effect transistor. *Electronics*, 5(1):11, 2016.
- [85] Z. Xiao and C. Durkan. Size effects in the resistivity of graphene nanoribbons. *Nanotechnology*, 30(44):445203, 2019.
- [86] S. Masubuchi, K. Iguchi, T. Yamaguchi, M. Onuki, M. Arai, K. Watanabe, T. Taniguchi, and T. Machida. Boundary scattering in ballistic graphene. *Physical Review Letters*, 109:036601, 2012.

- [87] E. Walter, T. Ö. Rosdahl, A. R. Akhmerov, and F. Hassler. Breakdown of the law of reflection at a disordered graphene edge. *Physical Review Letters*, 121:136803, 2018.
- [88] E. J. Mele. Commensuration and interlayer coherence in twisted bilayer graphene. *Physical Review B*, 81:161405, 2010.
- [89] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero. Unconventional superconductivity in magic-angle graphene superlattices. *Nature*, 556:43–50, 2018.
- [90] G. Tarnopolsky, A. J. Kruchkov, and A. Vishwanath. Origin of magic angles in twisted bilayer graphene. *Physical Review Letters*, 122:106405, 2019.
- [91] E. Y. Andrei and A. H. MacDonald. Graphene bilayers with a twist. *Nature Materials*, 19:1265–1275, 2020.
- [92] S. Shallcross, S. Sharma, and O. A. Pankratov. Quantum interference at the twist boundary in graphene. *Physical Review Letters*, 101:056803, 2008.
- [93] J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto. Continuum model of the twisted graphene bilayer. *Physical Review B*, 86:155449, 2012.
- [94] S. Carr, S. Fang, and E. Kaxiras. Electronic-structure methods for twisted moiré layers. *Nature Reviews Materials*, 5(10):748–763, 2020.
- [95] Mikito Koshino. Band structure and topological properties of twisted double bilayer graphene. *Physical Review B*, 99:235406, 2019.
- [96] R. Bistritzer and A. H. MacDonald. Moiré bands in twisted double-layer graphene. *Proceedings of the National Academy of Sciences*, 108(30):12233–12237, 2011.
- [97] H. Polshyn, M. Yankowitz, S. Chen, Y. Zhang, K. Watanabe, T. Taniguchi, C. R. Dean, and A. F. Young. Large linear-in-temperature resistivity in twisted bilayer graphene. *Nature Physics*, 15:1011–1016, 2019.
- [98] M. Yankowitz, S. Chen, H. Polshyn, Y. Zhang, K. Watanabe, T. Taniguchi, D. Graf, and C. R. Dean. Tuning superconductivity in twisted bilayer graphene. *Science*, 363(6431):1059–1064, 2019.

- [99] A. Kerelsky, L. J. McGilly, D. M. Kennes, L. Xian, M. Yankowitz, S. Chen, K. Watanabe, T. Taniguchi, J. Hone, C. Dean, A. Rubio, and A. N. Pasupathy. Maximized electron interactions at the magic angle in twisted bilayer graphene. *Nature*, 572:95–100, 2019.
- [100] L. Balents, C. R. Dean, D. K. Efetov, and A. F. Young. Superconductivity and strong correlations in moiré flat bands. *Nature Physics*, 16:725–733, 2020.
- [101] J. M. B. Lopes dos Santos, N. M. R. Peres, and A. H. Castro Neto. Graphene bilayer with a twist: Electronic structure. *Physical Review Letters*, 99:256802, 2007.
- [102] E. Suárez Morell, J. D. Correa, P. Vargas, M. Pacheco, and A. H. C. Neto. Flat bands in slightly twisted bilayer graphene: Tight-binding calculations. *Physical Review B*, 82:121407, 2010.