Spin and charge transport in graphene devices in the classical and quantum regimes
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Document Version
Publisher's PDF, also known as Version of record

Publication date:
2015

Link to publication in University of Groningen/UMCG research database

Citation for published version (APA):

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Download date: 03-03-2019
Chapter 3
Spin transport and relaxation in graphene

Abstract
Graphene is a very promising material for spin transport and spin based logic circuits. Due to its low spin orbit coupling and low hyperfine interactions, initial theoretical predictions pointed out that graphene has the potential for transferring spin information over very long distances even at room temperature. The first experimental works fell short of these initial predictions, which incited both theoreticians and experimentalists to explore the reasons for these results and find the experimental limits for graphene spintronics. The field of graphene spintronics is growing rapidly in both theoretical and experimental fronts, not only to understand the limitations for spins in graphene, but also to combine the electronic properties of graphene with spin transport.

In this chapter I will review the main concepts of spin transport such as spin injection via a ferromagnetic electrode, the conductivity mismatch problem, the two channel model, the nonlocal spin valve and the Hanle effect. I will discuss how to measure the spin properties of graphene and how to model a non-homogeneous graphene device. At the end of the chapter I will give a short review on the spin relaxation mechanisms for graphene.
3. Spin transport and relaxation in graphene

3.1 Spintronics

The use of the electronic spin to carry information, known nowadays as spintronics [1], is receiving increasing attention since the discovery of the giant magnetoresistance (GMR) effect [2, 3]. Spintronics is strongly application driven since its start, and is the basis of revolutionary inventions such as the hard disk drive and more recently magnetic random access memories (M-RAM) and spin transfer torque random access memories (STT-RAM). With the demand for increasingly smaller and faster devices, the understanding of the limits of spintronic effects in nanoscale devices is of high importance.

Graphene is one of the most promising materials for spintronic applications due to theoretical predictions of long spin relaxation times and lengths [4, 5]. But before I get into a detailed explanation of graphene’s spintronic properties we have to understand a few basic models and phenomena in spintronics.

I will start by explaining how to inject a spin polarized current in a non-magnetic material and the difficulties that might arise due to the difference in conductivity of the contacts and the non-magnetic material. Later I treat the specific case of spin transport in graphene and the most common methods to measure the spin transport parameters. Finally I finish the chapter by discussing the spin relaxation mechanisms in graphene.

3.2 Spin injection in non-magnetic materials

The most common way to produce a spin polarized current in a non-magnetic material (NM) is by the use of ferromagnetic (FM) electrodes. The density of states at the Fermi energy in a ferromagnetic material is different for the two spin species, which in here will be referred as “spin up” and “spin down” for simplicity. This difference in the density of states causes a difference in the conductivity for each spin direction. For a diffusive system we can calculate its conductivity using the Einstein’s relation:

\[ \sigma_{\uparrow(\downarrow)} = D_{\uparrow(\downarrow)} e^2 \nu(E)_{\uparrow(\downarrow)}, \]  

(3.1)

where \( e \) the electron charge, \( D_{\uparrow(\downarrow)} \) is the diffusion constant, and \( \nu(E)_{\uparrow(\downarrow)} \) the density of states at the energy \( E \) for spin up (down). The average diffusion constant can be written as \( \bar{D} = D_\uparrow D_\downarrow (\nu_\uparrow + \nu_\downarrow) / (D_\uparrow \nu_\uparrow + D_\downarrow \nu_\downarrow) \) [6, 7]. For the case of a non-magnetic conductor we have that \( \bar{D} = D_c \), where \( D_c \) is the charge diffusion constant.

When a current is driven through the FM/NM interface a spin imbalance is created in the NM which decays away from the interface in order to restore the equilibrium condition (see Fig. 3.1). This spin imbalance is called spin accumulation and it is measured by the difference in the chemical potentials for spin up and down: \( \mu_s = (\mu_\uparrow - \mu_\downarrow)/2 \). The characteristic distance in which \( \mu_s \) decays is called the spin
relaxation length, $\lambda_s$. The spin relaxation length, like the phase relaxation length discussed in the previous chapter, is related to a spin relaxation time $\tau_s$ and a spin diffusion constant $D_s$ by: $\lambda_s = \sqrt{D_s\tau_s}$. For the case where electron-electron interactions is not the dominant scattering mechanism, which is the case for graphene: $D_s \approx D_c$.¹

![Diagram of a FM/NM interface](image)

**Figure 3.1:** Scheme of a FM/NM interface with an applied current from the FM to the NM. Since in the FM the density of states (DOS) at the Fermi energy for one spin species is higher than the other current in the FM is spin polarized. When the current flows to the NM it creates a spin imbalance (spin accumulation) at the interface that decays towards the equilibrium condition with the characteristic length $\lambda_s$. Note that the magnetization of the FM points to the opposite direction of the spin accumulation. This is the case of cobalt, where the majority spins (down, red) have a lower DOS than minority spins (up, blue). The dark region at the interface represents a tunnel barrier, which is used to increase the spin injection efficiency as described in section 3.2.2.

### 3.2.1 Two channel model

An intuitive approach developed by Valet and Fert [9] to describe spin injection and transport in materials reduces the studied system into an equivalent resistor circuit. This model is commonly referred as the two channel model since it assumes the two spin species to flow in different branches of the circuit. A FM-NM-FM stack can be

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¹In the case of high electron-electron scattering $D_s < D_c$, due to a phenomenon called spin Coulomb drag [8].
described by assuming different resistances for spin up and down in the ferromagnets and equal resistances in the NM, as shown in Fig. 3.2.

![Diagram of spin transport in graphene](image)

**Figure 3.2:** Two channel model for a FM/NM/FM stack in a parallel (left) and antiparallel (right) state. The resistances (over the spin relaxation length) $R = R(1 + \gamma)$ and $r = R(1 - \gamma)$ represent the resistances for each spin channel depending on the magnetization orientation of the FM. The resistances $R_{rel}$ represent the spin relaxation resistances, connecting the spin up and down channels.

The magnetization of the ferromagnets is described by the resistances for the flow of spin up or down. We can quantify the polarization of the current through the FM by writing the equivalent resistance for spin up in the FM as $R_{\uparrow} = R(1 - \gamma)$ and for spin down as $R_{\downarrow} = R(1 + \gamma)$, where $\gamma$ is the polarization of the electrodes and $R$ the average resistance of the FM. The spin relaxation in the NM can be accounted for by including a spin relaxation resistance ($R_{rel}$) as shown in Fig. 3.2. It is easy to show that for a FM/NM/FM stack in the absence of spin relaxation ($R_{rel} \gg 1$), a spin polarized current flows when the FM are oriented parallel to each other (Fig. 3.2 left) and a spin accumulation (voltage between the spin up and down branches) develops for an antiparallel orientation (Fig. 3.2 right). This simple model can be used to describe even complicated systems with several contacts [10].

### 3.2.2 Conductivity mismatch and contact induced spin relaxation

One of the major issues that has to be circumvented in semiconductor spintronics is known as the conductivity mismatch problem [7, 11]. This issue arises from the fact that when spins are forced into a non-magnetic material by a ferromagnetic electrode they tend to go back to the ferromagnet where they loose their information due to the shorter lifetime of spins in the ferromagnetic material. To quantify the conductivity

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**References:**

[7, 11]
mismatch problem we introduce the spin resistance for a certain material as \( R_\lambda = \rho \lambda_s / A \), where \( \rho \) is the resistivity and \( A \) the channel cross-sectional area. \( R_\lambda \) can be written for the special case of a 2D material as graphene as:

\[
R_\lambda = \frac{R_{sq} \lambda_s}{W},
\]

(3.2)

where \( R_{sq} \) is the sheet resistance and \( W \) the channel width.

When the ratio between the spin resistances of the ferromagnet and the non-magnetic channel is much smaller than 1 the spins tend to go back and relax in the ferromagnets, therefore no spin accumulation is generated in the channel. This is usually the case since both the spin relaxation length and the resistivity of most ferromagnetic metals are much smaller than those of graphene or semiconductors.

The conductivity mismatch can be circumvented by including a highly resistive interface barrier between the ferromagnetic electrode and the non-magnetic channel \cite{12}. This barrier, usually made of metal oxides such as \( Al_2O_3, TiO_2 \) and \( MgO \), can have a resistance \( R_b \) much higher than the spin resistance of the channel which can be controlled by the oxide layer thickness.

In chapter 4 I will discuss the effect of conductivity mismatch for the specific case of the contacts used in the experimental research presented in this thesis.

## 3.3 Spin transport in graphene

In this section I discuss the specific case of graphene spin transport and the most used methods to extract its spin transport properties. I will also explain the model used to describe the diffusive transport of spins in graphene and then, at the end of the section, extend this model to more realistic devices.

### 3.3.1 Spin diffusion equation

To describe the motion of spins in graphene we use the spin diffusion equation in a steady state condition \cite{7}:

\[
\frac{d\bar{\mu}_s}{dt} = 0 = D_s \nabla^2 \bar{\mu}_s - \frac{\bar{\mu}_s}{\tau_s},
\]

(3.3)

where \( \bar{\mu}_s = (\mu_x, \mu_y, \mu_z) \) represents the spin accumulation in 3 dimensions. The first term on the right represents the diffusion of the spins due to a difference in the chemical potential with the diffusion constant \( D_s \) and the last term represents the spin relaxation with the relaxation time \( \tau_s \).

Equation 3.3 can be solved for 1D transport (e.g. in the \( x \) direction) resulting in \( \mu_s = \mu^0_s (A e^{-\frac{x}{\lambda_s}} + B e^{+\frac{x}{\lambda_s}}) \), where \( A \) and \( B \) are constants to be defined by the boundary conditions. In an infinite 1D channel, we have the boundary condition...
\( \mu_s(x = \pm \infty) = 0 \). This leads to the general solution: \( \mu_s = \mu_0 e^{-\frac{|x|}{\lambda_s}} \), i.e. the spin accumulation decays exponentially towards the equilibrium condition (\( \mu_s = 0 \)) with a characteristic length scale \( \lambda_s \). Therefore, it is possible to experimentally obtain \( \lambda_s \) by measuring the spin signal as a function of the distance between the spin injector and detector electrodes [13]. However, as it will be explained below, this method is usually prone to errors due to the randomly different polarization of the electrodes.

### 3.3.2 Nonlocal spin valve

In the previous section we discussed the properties of a FM/NM/FM stack. When a current is driven through these stacks we can observe a spin polarized current or a spin accumulation in the NM depending on the relative orientation of the FM electrodes. However, the spin polarization of the current is usually in the order of 10\%. This means that the difference in resistance between parallel and antiparallel orientation of the FM is only a small fraction of the total resistance, which makes it difficult to distinguish the spin related signal from the background signal. Furthermore, charge related phenomena such as the magneto Coulomb effect [14] and anomalous Hall effects [15] can mimic spin related signals. An elegant way to avoid this problem is to take advantage of the diffusive behaviour of the spins in the NM and isolate the current path from the voltage detection as illustrated in Fig. 3.3(a).

This configuration, called nonlocal spin valve, uses the fact that in diffusive transport all the information carried by the phase or momentum of the electrons gets randomized (typically) over a much shorter length than the spin relaxation length (\( \lambda_\phi, l_{mfp} \ll \lambda_s \)). As we will see later in chapter 7 this condition is not obeyed in phase coherent devices, which leads to interesting observations.

A typical nonlocal spin valve consists of 4 ferromagnetic electrodes with the 2 central electrodes separated by a distance \( L \), as shown in Fig. 3.3(a). Driving a current between contacts 1 and 2 generates a spin accumulation underneath these contacts. Since the current flows from contact 2 to contact 1, when both magnetizations are aligned in parallel, contact 2 injects spin up while contact 1 extracts spin up. The spin accumulation generated by contact 2 is: \( \mu_{s2}^0 = e P_2 IR \lambda_s \), where \( P_2 \) is the spin polarization of contact 2 and \( I \) the current used. The spin accumulation given by contact 1 is: \( \mu_{s1}^0 = -e P_1 IR \lambda_s \), since it extracts spins with an efficiency \( P_1 \). The total spin accumulation is a sum of the spin accumulation given by both contacts: \( \mu_s(x) = \mu_{s1}^0 e^{-|x-x_1|/\lambda_s} + \mu_{s2}^0 e^{-|x-x_2|/\lambda_s} \), where \( x_1(2) \) is the position of contact 1 (2).

Since contacts 3 and 4 are ferromagnetic, they probe preferentially the chemical potential of one spin species, Fig. 3.3(b). In the case where the ferromagnetic contacts have parallel magnetization for example, spins up are preferentially detected by the contacts. The voltage measured between the two contacts is: \( V_{nl} = P_3 \frac{\mu_s(x_3)}{e} - P_4 \frac{\mu_s(x_4)}{e} \), where \( x_3 \) and \( x_4 \) are the locations of contacts 3 and 4 respectively. Therefore, it can be shown that the nonlocal voltage including the effect of all
3.3. Spin transport in graphene

![Figure 3.3: (a) Nonlocal spin valve schematic view showing the 4 FM electrodes and the connection circuit. (b) Spin accumulation as a function of the distance for the case where all the electrodes pairs are parallel (top) and antiparallel (bottom). The dashed lines show the spin accumulation for spin up due to each injector. The total spin accumulation for spin up (down) is shown in blue (red).](image)

In order to change the magnetization direction relative to another electrode one by one, their coercive fields are made different. Therefore, by applying a magnetic field, the magnetization of each electrode can be switched individually.

The factor $1/2$ appears due to the fact that the injected spins diffuse in both positive and negative $x$-direction.

\begin{equation}
V_{nl} = \frac{IR_{sq}\lambda_s}{2W} \left[ P_3 \left( P_2 e^{-|x_3-x_2|/\lambda_s} - P_1 e^{-|x_3-x_1|/\lambda_s} \right) - P_1 \left( P_2 e^{-|x_4-x_2|/\lambda_s} - P_1 e^{-|x_4-x_1|/\lambda_s} \right) \right].
\end{equation}

\[ (3.4) \]
field parallel to the contacts, their magnetization switches at different values of magnetic field. We ensure a different coercive field for each electrode by changing their width between 100 - 800 nm. It is important to point out that in order to have sharp switches as shown in Fig. 3.5(a) the electrodes have to be composed of a single magnetic domain and the easy axis for the magnetization should be well defined and parallel to the field. For that all our electrodes have an aspect ratio length/width > 6, with lengths below 15 µm and the electrodes extend away from the sample keeping right angles to avoid domain wall propagation (Fig. 3.4). Furthermore, in order to avoid that adjacent contacts couple magnetically with one another via stray fields, we keep their “loose ends” on opposite sides.

Figure 3.4: Scanning electron micrograph of a suspended graphene sample as used in Chapter 8 showing the Co electrodes used to contact the graphene.

When contacts 1 and 4 are far away from the two inner contacts we have that $|x_3 - x_1|, |x_4 - x_2|, |x_4 - x_1| \gg \lambda_s$ and Eq. 3.4 simplifies to (using $|x_3 - x_2| = L$):

$$V_{nl} = \frac{IR_{sq}\lambda_s}{2W} P_2 P_3 e^{-L/\lambda_s}. \quad (3.5)$$

This means that if the first injector is far away, the generated spin accumulation decays to nearly zero before reaching the detection circuit. The outermost detector being far away means that $\mu_s$ decays to nearly zero before reaching it. Therefore, contact 4 effectively probes the equilibrium chemical potential: $\mu_s \approx 0$ eV.

Considering just the inner electrodes, when they are aligned in a parallel configuration, contact 2 injects spin up and contact 3 detects spin up, see Fig. 3.3(b). This results in a positive nonlocal voltage. If the inner electrodes are aligned in an antiparallel configuration the measured nonlocal voltage is negative since contact 2 injects spin up and contact 3 detects preferentially the chemical potential for spin down.

If we normalize $V_{nl}$ by $I$ we obtain a resistance called nonlocal resistance (using $P_2 = P_1$ and $P_3 = P_d$):
3.3. Spin transport in graphene

\[
R_{nl} = \pm P_l P_d R_{sq} \lambda_s \frac{\lambda_s}{2W} e^{-\frac{L}{\lambda_s}}. \tag{3.6}
\]

The \( \pm \) sign indicates the relative orientation of the FM contacts, + when they are parallel and − for antiparallel.

Equation 3.6 shows that by changing the relative orientation of the FM electrodes, we should observe a sudden jump on the measured nonlocal resistance from positive to negative and symmetric around \( R_{nl} = 0 \text{Ω} \). This measurement is often referred as a nonlocal spin valve measurement. Sometimes a small background, due to heating or regular Ohmic contributions for example, can be observed, Fig. 3.5(a).

\( R_{nl} \) decays exponentially with the distance \( L \) between electrodes with the characteristic decay length \( \lambda_s \) as expected for the spin signal, Fig. 3.5(b). By taking the natural logarithm of \( R_{nl} \) we expect a linear behaviour with a slope \(-1/\lambda_s\). However, we can now see that in order to accurately measure \( \lambda_s \) by obtaining \( R_{nl} \) for different \( L \), we cannot have a large spread in the polarization of the used contacts. If \( P \) varies strongly, the decaying trend of \( R_{nl} \) gets less clear, leading to large errors in \( \lambda_s \). This variation of the contact polarization is the main responsible for the deviation of the data in Fig. 3.5(b).

Figure 3.5: (a) Nonlocal spin valve for a device based on bilayer graphene on lift-off resist (LOR) for \( L = 1.8 \text{µm} \) and \( L = 2.8 \text{µm} \). (b) Natural logarithm of the nonlocal spin signal as a function of the distance \( L \). Using a linear fit we extract \( \lambda_s = (0.89 \pm 0.05) \text{µm} \). Data from: M.H.D. Guimarães, et al., unpublished.
3.3.3 Hanle effect

A more accurate way to obtain the spin transport parameters in the graphene channel is by the use of the Hanle effect \[7\]. When a perpendicular magnetic field \( \vec{B} = B_0 \hat{z} \) is applied to the device, the injected spins precess around the field while diffusing. To describe the spin transport in the system we can use a diffusive picture \[16\] in which spins injected from a contact have a probability distribution as a function of time, \( \mathcal{P}_D(t) \), to reach a detector electrode at a distance \( L \) given by:

\[
\mathcal{P}_D(t) = \frac{1}{\sqrt{4\pi D_s t}} e^{-\frac{L^2}{4D_s t}}. \tag{3.7}
\]

The spin relaxation can be included by multiplying Eq. 3.7 by \( \mathcal{P}_\tau = e^{-t/\tau_s} \). The spin precession due to the applied magnetic field is taken into account by including the term \( \cos(\omega t) \), where \( \omega = \frac{g\mu_B B_0}{\hbar} \) with \( g \) the Landé g-factor, \( \mu_B \) the Bohr magneton and \( \hbar \) the reduced Planck constant. Therefore, the contribution to the signal at a time \( t \) after the injection, including the effects of diffusion, precession and relaxation is given by:

\[
\mathcal{P}(t) = \frac{1}{\sqrt{4\pi D_s t}} e^{-\frac{L^2}{4D_s t}} e^{-t/\tau_s} \cos(\omega t). \tag{3.8}
\]

To find the spin accumulation (in the \( \hat{y} \) direction) we integrate Eq. 3.8 in time:

\[
\mu_s(L, B) = 2\sqrt{\frac{D_s}{\tau_s}} \mu_s^0 \int_0^\infty \mathcal{P}(t) dt, \tag{3.9}
\]

where the pre-factor \( 2\sqrt{\frac{D_s}{\tau_s}} \) comes from the condition \( \mu_s(L = 0, B = 0) = \mu_s^0 \).

The integral in Eq. 3.9 can be solved using a mathematical package software such as Wolfram Mathematica and, using \( R_{nl}(L, B) = P_d/(eI)\mu_s(L, B) \), an analytical expression for the measured nonlocal resistance can be obtained:

\[
R_{nl}(L, B) = \pm \frac{P_d P_d R_{sq} D_s}{W} \operatorname{Re} \left\{ \frac{1}{2\sqrt{D_s}} \frac{e^{-L\sqrt{\lambda_s^2 - i\frac{\omega}{\tau_s}}}}{\sqrt{\tau_s^{-1} - i\omega}} \right\}. \tag{3.10}
\]

Eq. 3.10 describes how the nonlocal resistance depends on an applied perpendicular magnetic field and the distance between electrodes. Since this equation depends on \( D_s \) and \( \tau_s \), this expression can be used to extract the spin transport parameters independently \[17\][18]. Measuring the nonlocal signal in a spin valve as a function of a perpendicular magnetic field, the experimental data can be fitted by equation 3.10 assuming \( g=2 \) and keeping \( D_s, \tau_s \) and the amplitude of the signal as fitting parameters. An example of a Hanle measurement and its fit is shown in Fig. 3.6.

\[3\] Here we are considering the projection of the spins in the \( \hat{y} \) direction, which is the easy axis of magnetization of the electrodes.
3.3. Spin transport in graphene

As it can be seen in Fig. 3.6, with an increase of the magnetic field, the non-local signal decreases until it crosses zero and reaches a maximum negative value. This happens since the spins precess around $B$ with the point where $R_{nl} = 0\ \Omega$ corresponding to an average 90$^\circ$ rotation and the maximum negative value corresponding to an average 180$^\circ$ rotation. With further increase of $B$ the spins precess to higher angles before reaching the detector electrode and $R_{nl}$ decays to zero due to incoherent precession and relaxation.

In order to exclude the background signal from other sources in the Hanle precession measurements we use the fact that the spin signal for parallel and antiparallel of the contacts is mirrored in respect to zero spin signal (inset of Fig. 3.6). By subtracting the nonlocal signal for the antiparallel from the parallel magnetization and dividing by 2 we have an average signal $R_s = (R_P - R_{AP})/2$. The resulting signal $R_s$ is in principle purely due to spin contributions.

\[ L = 2.8\ \mu m \]
\[ D_s = 0.004 \ m^2/s \]
\[ \tau_s = 508 \ \text{ps} \]
\[ \lambda_s = 1.42 \ \mu m \]

\[ B (T) \]
\[ R_{nl} (\Omega) \]

---

\footnote{Although rare, effects of temperature drift during the measurements due to, for example sample heating, might induce a background which is usually linear in time and can be accounted by adding a linear slope. This effect is usually suppressed at low temperatures.}
3.3.4 Spin diffusion in non-homogeneous systems

For some cases, the samples are non-homogeneous, consisting of regions with different properties, e.g. different diffusion constants, square resistances and spin relaxation times. One simple case is when the central region between the spin injector and detector has distinct properties from the ones of the identical outer regions (Fig. 3.7).

For this case the approach we used before to obtain an analytical expression for $R_{nl}$ is not as easy. Therefore, we solve the Bloch equations as proposed by Johnson and Silsbee [19] and worked out in detail by F. Jedema [6] and M. Popinciuc et al. [20], taking special attention to the boundary conditions, which are different from the cases treated before.

We start by modifying Eq. 3.3 to include a precession term (for transport in the $\hat{x}$ direction):

$$D_s \frac{d^2 \vec{\mu}_s}{dx^2} - \frac{\vec{\mu}_s}{\tau_s} + \vec{\omega} \times \vec{\mu}_s = 0,$$

This equation is commonly referred as Bloch equation.

The solution to the equation can be obtained assuming that the spin accumulation decays to zero at infinity and is continuous everywhere and the spin current (proportional to the derivative of $\mu_s$ as a function of $x$) is continuous everywhere except at the spin injection point. Therefore we assume the following boundary conditions:

1. The spin accumulation decays to zero at infinity: $\vec{\mu}_s(\pm \infty) = 0$,

2. Continuity of the spin accumulation: $\vec{\mu}_s(x)$ is continuous everywhere, and
3. Continuity of the spin current: \( 1/(eR_{sq})d\vec{\mu}/dx \) is continuous except at the injection point where the spin accumulation in the \( y \) direction is discontinuous by \( P_i I/2 \).

This results in a set of coupled differential equations for the \( x \) and \( y \) components of the spin accumulation \( \mu_x \) and \( \mu_y \):

\[
D_s \frac{d^2 \mu_x}{dx^2} - \frac{\mu_x}{\tau_s} + \frac{g\mu_B}{\hbar} B_0 \mu_y = 0 \quad (3.12a)
\]

\[
D_s \frac{d^2 \mu_y}{dx^2} - \frac{\mu_y}{\tau_s} - \frac{g\mu_B}{\hbar} B_0 \mu_x = 0 \quad (3.12b)
\]

where \( B_0 \) is the magnetic field strength in the \( z \) direction and \( D_s \) and \( \tau_s \) can have different values for the different regions. These equations have solutions of the type:

\[
\mu_j = c_1^j e^{+\frac{x}{\lambda_s} \sqrt{1+i(\gamma B_0 \tau_s)}} + c_2^j e^{+\frac{x}{\lambda_s} \sqrt{1-i(\gamma B_0 \tau_s)}} + c_3^j e^{-\frac{x}{\lambda_s} \sqrt{1+i(\gamma B_0 \tau_s)}} + c_4^j e^{-\frac{x}{\lambda_s} \sqrt{1-i(\gamma B_0 \tau_s)}},
\]

where \( c_k^j \) \( (k = 1, 2, 3, 4) \) are constants and \( j = x, y \). Considering the three regions (two outer and one inner), leads to 12 independent constants to be determined, which can be reduced to 8 after applying the first boundary condition. The remaining constants can then be obtained using a mathematical program, such as MATLAB, to solve the coupled equations in a matrix form and obtain the nonlocal resistance as a function of the different parameters involved.

This model can explain well different experiments shown in chapters 8 and 9. For comparison with the experiments, we can solve the equation numerically as a function of magnetic field to generate Hanle precession curves with known values for the diffusion coefficient and relaxation times for all the regions. These simulated Hanle curves are then fitted with the solution homogeneous systems as done in the experiments. From this fitting procedure we extract \( \tau_s \) and \( D_s \) that can be compared to the experiment and the values used for the simulation \(^{21}\).

It is worth noting that this procedure is required due to the diffusive nature of the spins. This means that the spins can probe a significant amount of the three regions before being detected, and therefore the Hanle precession curve contains information of the three regions and not only the central one. However, if the central region is long enough \((L \gg \lambda_s)\), the spins that reach the detection circuit spend a very long time in the central region and therefore the solution for the homogeneous system can be used with reasonable accuracy to extract the spin transport parameters of the central region. This can be also understood if we take, for example, that the

\(^{5}\)It is important to note that the square resistance \( R_{sq} \) is different for each region: \( R_{sq} = R_o \) for the outer regions and \( R_{sq} = R_i \) for the inner region.
Spin relaxation time for the outer regions is much smaller than the central region. If the time the spins take to diffuse from the injector to the detector $\tau_d = L^2 / 2D_s$ is higher than the spin relaxation time for all regions, only spins that go in a more straightforward path toward the detector can be measured before relaxing.

Therefore, the spin relaxation obtained by fitting the data with the solution of the Bloch equation for a homogeneous system will be a combination of the spin relaxation times in each region: $\tau_s^{-1} = \alpha/\tau_i + (1 - \alpha)/\tau_o$, where $\alpha$ is a constant that quantifies the contribution of each region on $\tau_s$. The value for $\alpha$ will depend on the average time the spin spends in the central region as compared to the relaxation time in that region.

### 3.4 Spin relaxation in graphene

In this section I am going to discuss the status of the understanding of the spin relaxation mechanisms in graphene. One of the reasons why graphene has attracted a lot of attention in the area of spintronics is due to initial theoretical predictions of very long spin relaxation times ($\tau_s \approx 1\ \mu s$) and lengths ($\lambda_s \approx 100\ \mu m$) [22, 23]. However, the first experimental results fell a few orders of magnitude short than the initial predictions [18]. Further theoretical [4, 5, 24–29] and experimental [20, 21, 30–40] efforts pointed out impurities and adatoms as possible sources of enhanced spin-orbit coupling (SOC) and therefore spin relaxation.

Two main mechanisms are often used to describe spin relaxation in metals and semiconductors. They are the D’Yakonov-Perel (DP) [41] and the Elliott-Yafet (EY) [42, 43] mechanisms. Both these mechanisms use the fact that SOC couples the electron momentum to the spin [7].

Specifically for graphene, the intrinsic SOC strength is very small [22, 23, 44] ($\Delta_{int} < 1\ \mu eV$) and often ignored when compared to SOC arising from other sources, as for example adatoms [25] ($\Delta_{SO} \approx 1\ \text{meV}$), ripples [4] ($\Delta_{SO} \approx 10\ \mu eV$) and electric fields [5, 22, 23, 26, 44, 46] ($\Delta_{SO} \approx 0 - 10\ \mu eV$). These extra SOC effects often break the inversion symmetry of the graphene flake and give rise to an extra Rashba-like term in the Hamiltonian [44]:

$$H_{SO} = \frac{\Delta_{SO}}{2} (\sigma \times s)_z,$$

where $\sigma$ and $s$ are the pseudospin and real spin Pauli matrices respectively. We ignore the Rashba term $(k_F \times s)_z$ since this is much smaller when compared to the term that takes the pseudospin [44]. It is important to point out that the electronic momentum and spin are still coupled since the pseudospin and momentum direction are intimately related.

The spin orbit term in Eq. 3.14 leads to a small separation in energy for bands with different spin directions (Fig. 3.8). In the picture of the EY and DP mecha-
nisms, this gives rise to two distinct mechanisms for spin scattering. In a EY-like mechanism, the spin-orbit interaction couples electronic states with opposite spin projections in different bands making it difficult to define Bloch states with a well defined spin polarization \cite{28,42}. Each momentum scattering event leads to a random change in momentum direction which causes changes to the spin direction. For a DP-like mechanism the SOC term gives rise to an effective momentum-dependent effective magnetic field, which causes the spins to precess with a direction and intensity related to their momentum.

**Figure 3.8:** (a) A simplified scheme of the graphene band structure in the presence of Rashba spin orbit coupling ($\Delta_{SO}$). The different colours for the branches indicate different spin species. (b) Fermi surface of the band structure in the left showing the effect of spin orbit coupling in the spin direction as a function of the momentum as in Ref. \cite{44}.

The resulting spin relaxation times for graphene due to the DP and EY mechanisms are, respectively \cite{28}:

\[
\tau_{s,DP} \approx \frac{\hbar^2}{\Delta_{SO}^2} \frac{1}{\tau_p}, \quad (3.15a)
\]

\[
\tau_{s,EY} \approx \frac{E_F^2}{\Delta_{SO}^2} \tau_p, \quad (3.15b)
\]

where $E_F$ is the Fermi energy and $\tau_p$ the momentum relaxation time. The result for the EY mechanism was calculated for the specific case of graphene with weak scatterers by Huertas-Hernando et al. \cite{4} and later shown to be general for several types of scatterers by Ochoa et al. \cite{28}.

The biggest noticeable difference between the two mechanisms is their scaling of $\tau_s$ as a function of $\tau_p$. Assuming the DP mechanism means that samples with low electronic mobility (low $\tau_p$) have higher spin relaxation times when compared to high mobility samples. On the other hand, the EY mechanism means that low electronic
mobility samples have lower spin relaxation times when compared to high mobility samples.

Following the reasoning of Ref. [4], if we take the ratio of the two contributions to the spin relaxation we find:

\[
\frac{\tau_s^{EY}}{\tau_s^{DP}} \approx \left( \frac{E_F \tau_p}{\hbar} \right)^2.
\] (3.16)

Since the total spin relaxation time is obtained by: \(1/\tau_s = (1/\tau_s^{EY}) + (1/\tau_s^{DP})\), if the ratio given by Eq. 3.16 is higher than 1, DP is the most relevant mechanism for spin relaxation in graphene. By assuming typical experimental values: \(E_F \approx 10\) - 100 meV (corresponding to a charge carrier density of \(n \approx 10^{11} - 10^{13} \text{ cm}^{-2}\)) and \(\tau_p \approx 10^{-14} - 10^{-13} \text{ s}\), we find that \(\tau_s^{EY}/\tau_s^{DP} \approx 0.1 - 10^3\). This means that the DP mechanism dominates the spin relaxation in graphene most of the time, with the EY mechanism being relevant only for very low quality samples (low \(\tau_p\)) at very low energies \((E_F \approx 10 \text{ meV})\).

Although early studies showed a linear relation between \(\tau_p\) and \(\tau_s\) in graphene, which suggested a EY mechanism for spin relaxation [47] when the Fermi energy of the carriers is taken into account, EY alone cannot be the responsible for the measured results [28]. Later experimental efforts have shown that when the electronic mobility (and therefore \(\tau_p\)) is modulated using an electrolyte [39] or by adatoms [40] while keeping \(E_F\) fixed, \(\tau_s\) does not show a clear behaviour as a function of \(\tau_p\). Experimental results can be fitted by a combination of the DP and EY mechanism assuming different strengths for the mechanisms, which is taken into account by assuming two different \(\Delta\text{SO}\) terms [31].

Later theoretical efforts have focused on explaining the experimental results by using adatoms [24, 25], the substrate [26] and magnetic impurities [27] as the main sources for spin relaxation. Hydrogen adatoms for example, cause the graphene atoms to locally change from a sp\(^2\) to a sp\(^3\) hybridization [25]. These sp\(^3\) bonds distort the lattice and cause a local enhancement of the SOC leading to spin scattering. The spin relaxation including this local (and random) enhancement of the SOC can be treated using random Rashba fields, which shows reasonable agreement with different experimental data [24]. In addition to locally enhancing the SOC, the distortion of the graphene lattice created by the covalent bonding with an adatom can also give rise to local magnetic moments. This effect was elegantly demonstrated by McCreary et al. [48] using hydrogen adatoms and pure spin currents. It was later theoretically estimated that even a very small amount of such magnetic moments can decrease the spin relaxation from the intrinsic values of \(\approx 100 \text{ ns}\) to the experimentally obtained values of \(\approx 100 \text{ ps}\) [27]. By studying the spin relaxation in few-layer graphene, it was experimentally shown that \(\tau_s\) increases by increasing the number of layers [38]. This was attributed to an impurity screening effect of the outer graphene layers which is in agreement with theoretical suggestions that
adatoms and impurities coming from the sample preparation procedures and the substrate have a demeaning effect on the spin transport.

In this thesis, in addition to exploring the possibility of using quantum coherence and confinement in graphene spin transport (chapters 6 and 7), I also focused on the study of spin transport in high quality graphene devices. In the quest for the highest electronic mobility we performed spin transport in suspended graphene devices, but found that the non-suspended low quality regions dominate the measurement results for the spin relaxation times. However, due to the high diffusion coefficient, the spin relaxation length in these devices was improved by more than a factor of 2 when compared to SiO₂ based devices. Using another approach and encapsulating graphene with a less invasive substrate (hBN) we show that \( \tau_s \) can be increased by at least one order of magnitude when compared to SiO₂ based devices (in the order of ns), which when combined with the large diffusion coefficients for these devices leads to a \( \lambda_s > 10 \, \mu \text{m} \). These encapsulated devices also provide a new platform for spin manipulation in graphene since a dual gated structure is possible. As it will be explained in chapter 9 the use of this double gated structure allows us to control separately the charge carrier density and a transverse electric field. I will briefly describe the effect of a transverse electric field on spin transport in graphene below.

Another relevant parameter is the direction of the SOC and the effect it has on the spin relaxation time in graphene. For most cases, e.g. impurities, substrate related effects and adatoms, the Rashba-type spin orbit fields are predicted to point parallel to the graphene plane [4, 24–26], while for curvature effects, e.g. ripples, the effective magnetic fields due to SOC are predicted to point out of the graphene plane [4]. Therefore, by studying the spin relaxation times for spins injected in-plane and out-of-plane we can establish the direction of the spin orbit fields since, if these point in-plane, the spin relaxation out-of-plane is approximately half the in-plane value: \( \tau_\perp = 0.5 \tau_\parallel \). This can be understood if one considers that the component of the spin accumulation in the direction of the spin orbit field does not dephase while the perpendicular components do. Following the same reasoning, in the case that the spin orbit fields point preferentially out-of-plane, we have: \( \tau_\perp \gg \tau_\parallel \).

**Electric field induced spin orbit coupling**

The graphene lattice is in principle inversion symmetric. This means that if you apply the inversion operation in a vector \( \vec{v}_1 = (x, y, z) \) obtaining \( \vec{v}_2 = (-x, -y, -z) \), both the initial and final vectors are indistinguishable. By applying a transverse electric field (\( \vec{E} = (0, 0, E_0) \)), this symmetry (along with several others) is broken [49]. This broken inversion symmetry allows the presence of a Rashba-like SOC term (eq. 3.14). The strength of the spin orbit coupling constant \( \Delta_{SO} \) can be controlled by the intensity of the electric field, leading to the electric control of spin precession in graphene. The possibility of electric spin manipulation is one of the most important
paradigms of spintronics and is a necessary characteristic for the development of the Datta-Das spin transistor [50].

Using the tight-binding approximation or density functional theory (DFT), the dependence of $\Delta_{SO}$ as a function of a perpendicular electric field $\vec{E}$ can be calculated. Theoretical works find that $\Delta_{SO} = \zeta \vec{E}$ with $\zeta$ in the range of $0.1$ to $66 \mu eV/Vnm^{-1}$ [5, 22, 23, 26, 44, 46]. As it will be shown in chapter 9, the first experimental estimations give $\zeta \approx 40 \mu eV/Vnm^{-1}$ and show that the spin orbit fields point preferentially in the graphene plane.
3.5 Supplementary literature

Spin injection and spin transport


Graphene spintronics


Spin relaxation mechanisms in semiconductors and graphene


References


3. Spin transport and relaxation in graphene


