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Spin and charge transport in graphene devices in the classical and quantum regimes

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Chapter 10

Conclusions

Abstract

This chapter presents an overall conclusion of the works presented in this thesis. Furthermore, I give a comparison between the results obtained during my research period with recent results from different groups. Next I give a short outlook on graphene spintronics and future directions that could be taken to answer the main questions still present in the field and open the door for new spin-based applications for graphene.

10.1 Conclusions

The work presented in this thesis is mainly focused on the spin transport in graphene. During the period of research in which the works presented here were performed (2010 - 2014) several breakthroughs were made in graphene research. New methods of fabrication were developed which increased the electronic mobility of graphene devices either by removing the substrate, therefore suspending the graphene flake, or using less invasive substrates, such as hexagonal boron nitride (hBN) [1–4]. Electronic mobilities above 10^5 cm^2/Vs were demonstrated in these high quality devices. Further improvement of the technique used to suspend graphene flakes allowed the fabrication of suspended graphene devices by a much safer acid-free method [5].

While studying these suspended graphene devices we found that ballistic nanoconstrictions can be formed after the cleaning procedure of current annealing [6]. Some of the measurements of this work were used in chapter 2 to illustrate conductance quantization due to size confinement and the evolution to the quantum Hall state.

In order to understand how this evolution from quantum confinement to the quantum Hall state happens in ballistic graphene nanostructures we simulated various shapes of graphene nanoconstrictions and compared our simulation results (chapter 5) to the experiments. Our simulations showed that in order to obtain results comparable to the experiments, the graphene nanoconstrictions have to be short, with its length comparable to its width. We also showed that the width of the graphene nanoconstrictions estimated by analysing the transition between quantum confinement and the quantum Hall state with a semiclassical approach is in good agreement with the real values used in our simulations.

The remaining of the thesis is focused on spin transport in graphene devices. Trying to understand if the edges of the flake introduce enhanced spin-flip processes, and also to study the effect of confinement on spins in graphene, in chapter 6 we studied the spin dynamics in graphene nanostructures. There we showed that contact induced spin relaxation can have a stronger influence in devices where the dimensions are smaller than the spin relaxation length when compared to larger devices. We also showed that enhanced spin-flip processes due to the edges is not the main source of spin relaxation for graphene on SiO_2 substrates.

When the dimensions of the device are comparable to the phase coherence length, quantum interference effects can influence the spin transport in graphene. In chapter 7 we demonstrate this fact showing that the nonlocal spin signal in quantum coherent graphene spin valves can be strongly modulated by the use of gate voltages. The strong modulation on the spin signal comes from quantum interference effects that give origin to the universal conductance fluctuations. Since the spin up and down channels have slightly different Fermi energies, the transmission for each spin species is slightly different and can be modulated using a gate voltage in a similar way as done in carbon nanotube quantum dots [7, 8].

It has been suggested by several theoretical works that the main source of spin relaxation in graphene is due to adatoms or (magnetic) impurities [9–18]. To verify this fact we fabricated high-quality suspended graphene spin valves using the same sample fabrication technique used for the study of ballistic suspended graphene nanoconstrictions. We showed in chapter 8 that, due to the enhanced electronic mobilities, the spin diffusion constant is increased by a factor higher than 5 when compared to the standard SiO_2 based graphene spin valves, reaching values around 0.1 m^2/s. However, we did not observe an improvement on the spin relaxation time for these devices, finding $\tau_s \approx$ 150 ps. These results lead to spin relaxation lengths in the order of $\lambda_s \approx$ By using a model that takes into account the clean central suspended region and the outer non-suspended regions, we show that the measured spin relaxation time is limited by the dirtier supported regions. Due to the diffusive nature of the spin transport, the spins probe both the suspended and supported regions. Therefore the system as a whole, not only the central region, contributes to the spin relaxation.

The use of graphene in devices in which spins are used to perform logical operations depend not only on a long spin relaxation time, but also include the ability to easily manipulate spins in graphene. In chapter 9 we showed that hBN encapsulated graphene spin valves not only show very long spin relaxation times, but also provide the control of spin relaxation using an electric field. There we used the fact that a dual gated geometry (with a top and bottom gate) allows for the independent control of the carrier density and the electric field in the graphene flake. The higher electronic mobility in these devices leads to a high spin diffusion constant in the order of 0.05 m^2/s and, in combination with the long spin relaxation times around 2 ns, leads to a spin relaxation length above 12 μm. By comparing the spin relaxation time for spins injected in-plane ($\tau_{||}$) with the one for spins out-of-plane ($\tau_\perp$) as a function of a transverse electric field ($\bar{E}$) we observed that an increase in the electric field reduces the ratio $\tau_{||}/\tau_\perp$ from 0.75 at $\bar{E}$=0 V/nm to $\tau_{||}/\tau_\perp \approx$ 0.65 at $\bar{E} \approx$ -0.6 V/nm. Our results are in agreement with an electric field induced Rashba-like spin orbit field pointing in the graphene plane as theoretically predicted [13, 15, 17]. This study is the first demonstration of electric field control of spin relaxation in graphene and will guide future researchers in more detailed studies and the application of graphene in spin logic devices.

10.2 Outlook

Since the development of the ”scotch-tape method” in 2004 [19] graphene research has been growing at increasing rate, with more than 32 thousand papers published in 2013 alone, and about 700 of those contain the words *graphene* and *spin*. In the past few years, crucial development for graphene spintronics has taken place. Graphene spintronics grew from a new field to a well established one, with very relevant results not only for the general graphene community but also for the spintronics community. Considerable knowledge on the effects of contact induced spin relaxation was obtained using graphene as an example material in theory works [20, 21] and as the spin transport channel in experiments [21, 22]. Several mechanisms for spin relaxation were proposed [9–18] and tested [20, 23–31] in order to understand the discrepancy between the initial theoretical predictions and experiments.

Although graphene spintronics grew considerably in the past year, with several groups around the world joining the field, there are still several questions to be answered and potential applications and phenomena to be explored. Perhaps the main intriguing question in the field nowadays is the discrepancy between theoretical predictions and experimental data for the spin relaxation in graphene [32]. Several works indicated that the main mechanism for spin relaxation in graphene can be due to adatoms [9–17], magnetic impurities [18, 33] or due to contact induced spin relaxation [22, 34]. However, experiments shown that there is no apparent correlation between graphene's momentum and spin relaxation times [28, 31, 35], or the range of contact resistances in the experiments and the obtained spin relaxation times [20, 21, 27] [1]. The influence of the substrate on the spin relaxation is also not completely clear [31]. The full suspension of the graphene flake shows similar spin transport parameters as SiO_2 based graphene spin valves [29], which agrees with theoretical predictions that substrate effects are not the main responsible for spin relaxation in graphene [12]. However, as shown in chapter 9, encapsulating the graphene flake in hBN results in a much higher spin relaxation time, even when the momentum relaxation time is comparable to graphene on SiO_2 or non-encapsulated hBN devices. Furthermore, the effect of defects and localized states in the substrate was shown to have a major effect on the obtained Hanle precession curves [36], but no detailed study has been done in these systems yet.

Spin transport in graphene in devices where quantum effects play a role is still an unexplored field. Initial experiments showed that Fabry-Perot interference can modulate the spin signals in graphene spin valves [37]. Still, these experiments were performed with the cavity defined by the two inner contacts, therefore they are not very well-defined and relatively big Fabry-Perot cavities. Chapter 7 showed that another quantum interference effect, known as universal conductance fluctuations, can also result in a modulation of the nonlocal spin signal by orders of magnitude. The recent advances in the fabrication of quantum coherent graphene devices [38, 39] can lead to very interesting studied when combined with spin transport in graphene.

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[1] Here I refer to the spin relaxation times obtained using nonlocal Hanle precession measurements. Although a much higher spin relaxation time was estimated using a local method, it is known that several effects could mimic spin signals, leading to an erroneous estimation of the spin relaxation time.

Well defined and small Fabry-Perot cavities can give an easy electrical control on the amplitude and signal of the spin signals. In a similar way as done in carbon nanotubes [7, 8], the electrical control of the spin signal could also be obtained in high quality graphene-based coulomb blockade devices [40].

As discussed in the introduction, logical operations in spintronic devices require spin injection, manipulation and detection combined in the same device. The first demonstration of electrical spin manipulation in graphene was shown in chapter 9. However, plenty of work have to be done before logical operations can be performed in graphene spintronic devices. Due to the low spin-orbit interaction in graphene, high electric fields are necessary to induce a Rashba-type spin-orbit coupling in the devices. For that, gating could be done using ionic liquids [41], or high-κ dielectrics [42] could be used as gate insulators. After a better electric control of the spin-orbit coupling is achieved, electric control over spin precession, and therefore controllable spin precession, can be shown.

New materials, such as layered transition metal dichalcogenides (TMDs) [43] and topological insulators (TIs) [44], can also open new roads for electronics and spintronics in low dimensional devices. These materials can make use of similar synthesis and fabrication procedures as the ones developed for graphene, and not only have properties complementary to graphene, but also show completely different physical phenomena, as spin-momentum locking in TIs and the valley Hall effect in TMDs. Since the knowledge acquired for graphene devices can be partially used to fabricate and understand devices based on these materials, several research groups mainly focused on graphene are now broadening their research to incorporate also TMDs and TIs. The main advantage of layered materials is that they can be easily stacked to create heterostructures which combine their different properties [45], leading to new possibilities for electronic, optical and spintronic devices.

In conclusion, graphene offers many possibilities for spintronics, not only as a channel with low spin relaxation but also easy spin manipulation. Although spin transport and relaxation in graphene has been widely studied, a lot remains to be done. One of the most puzzling questions, ”what limits the spin relaxation in graphene”, still remains open. The relationship between the momentum and spin relaxation times is also not understood. Moreover, the combination of quantum effects, such quantum coherence, with spin transport is just starting to be explored. The understanding of effects, like spin relaxation, spin dependent quantum coherence effects and electric spin manipulation in graphene, will be used to fabricate improved graphene-based spintronic devices which can lead to promising new applications. Furthermore, new layered materials and their combination with graphene opens new paths for spintronic devices based in low dimensional materials.

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