Electron spin transport in graphene and carbon nanotubes

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Introduction

1.1 Spintronics: from metals, semiconductors to organics

Electronics and spintronics are two fields of technology which are very strongly coupled. This is due to the fact that both electronic and spintronic devices use the same elementary particles, electrons, for their operation. However, each field uses a different fundamental property of the particle, in electronics it is the charge and in spintronics the angular momentum, also better known as spin. For the latter, placing the particle in a magnetic field results in a coupling of its magnetic moment (generated by its spin) to the magnetic field. If now the spin is measured we obtain only two possible states, the spin-up state and the spin-down state. This last spintronic property can be used to perform Boolean logic operations, in a similar way as is already done in nowadays computer chips. For this type of logic operations two states are needed, a zero and a one state. This can be easily found in a spintronic device just by assigning the spin-down state to the zero and the spin-up state to the one state. However, to be able to create a computer chip containing only spintronic devices it is necessary to build fundamental spintronic devices in which the spin state can be manipulated.

An extremely useful spintronic device is used in the read head of hard disks. It contains a giant magneto-resistance sensor which reads the data written on the disc, on which information is registered magnetically, and converts this to electric current. It was in 1988 when Albert Fert and Peter Grünberg each independently discovered the Giant Magnetoresistance effect (GMR) [1, 2]. Here, the electrical resistance of an Fe/Cr multilayer structure was found to depend on the relative orientation of the magnetization of the magnetic layers. In 2007, Albert Fert and Peter Grünberg were awarded the Nobel prize of Physics for this breakthrough. The GMR device not only is one of the most important spintronic devices found in the past fifty years but it is also considered to be one of the first applications of nanotechnology. This is due to the fact that nanotechnology techniques, developed in the 1970s, enabled the production of very thin layers of materials that are necessary for the GMR to work.

The first steps in spintronics were done thirty years before the discovery of GMR, when spintronic effects were measured in metallic systems. The first was the discovery of the anomalous magneto-resistance (AMR) effect [3], being a pure bulk effect, which is the dependence of the resistance of a ferromagnetic strip on the relative orientation of the magnetization and current. In 1973 the spin polarization of ferromagnets was measured by using a system containing layers in the sequence ferromagnet/Al_2O_3/Al in which the Al is in the superconducting state. After the discovery of GMR a system was investigated in which the two ferromagnetic layers are separated by a thin tunnel barrier. This resulted in the discovery of tunneling magneto-resistance (TMR) [4]. Replacing the tunnel barrier by other anorganic non-magnetic materials, for example aluminum, copper or even by organic materials gives the possibility to investigate the spin transport mechanisms in non-magnetic materials. First experiments in this direction were already done in 1985 by Johnson and Silsbee in which they investigated spin transport in an Al crystal using a permalloy/ aluminum crystal/ permalloy spin valve device [5–7]. Experiments performed in these so-called spin valves can result in spurious signals which look very similar to spin valve signals. Examples of spurious effects are Hall and AMR effects. In order to eliminate any spurious signals from the spin signals Jedema *et al.* used the non-local detection techniques to investigate spin transport in Al and in Cu [8–10]. The same non-local technique has also been used to investigate spin transport in organic systems, for example in a carbon nanotube [11] and in a graphene layer [12].

Using a semiconductor instead of a metal is expected to result in useful spintronic devices. One of the most useful is perhaps the spin field effect transistor. This device was first proposed by Datta in 1990 [13] and its operation is based on the Rashba spin-orbit coupling found in semiconductors. Here, a gate voltage changes the spin-orbit coupling and this in turn induces spin precession. High on-off rations could be obtained by using ferromagnetic electrodes with unity polarization. Semiconductors can also be used to produce light-driven spintronic devices. Here the band-gap found in semiconductors allows for optical spin injection and detection. Optical injection and detection enabled the first experimental investigation of spin transport in semiconductors. For years scientists tried to measure spin signals in all electrical semiconductor spin valve devices without any success. The difficulty was found in the so-called conductivity mismatch [14]. Since the resistivity of the semiconductor is much higher than that of the ferromagnetic injector and detector, it dominates the total resistance of the device and reduces the spin injection and detection efficiencies. This problem can be solved by modifying the interface properties between the two systems in such a way that the spin dependent resistance at the interface matches the spin independent resistance. The first all electrical spin injection and detection was accomplished in 2007 by Xiou *et al.* in GaAs [15].

The interest in organic conductors for spintronic applications has similar sources as that for the use of organic conductors in electronics. For example, it is expected that fabrication costs will be low as the organic material can be spin coated on the substrate. However, the main reason of interest in organics is that very large spin lifetimes are expected since the spin-orbit interaction and the hyperfine interaction are expected to be small. This is due to the fact that organic systems contain mainly light carbon atoms having an atomic number Z = 6 and therefore the main source of spin relaxation time, which is the spin orbit interaction, is small since it is proportional to Z^4 [16]. Also the hyperfine interaction of the spins with the carbon nuclei is small, only 1% of carbon nuclei (with N = 13) have a magnetic moment. Organic spin valve devices based on carbon nanotubes [11,17–20], on molecules 8-hydroxy-quinoline aluminium (Alq3) [21] and on sexithienyl (T6) [22] have been reported. However, the fact that most experiments have been performed using the conventional two probe measurements which can result in spurious signals raises the question if the measured spin lifetimes are correct. Like in metallic systems, this spin lifetime can be correctly determined using the non-local technique.

Very recently a lot of attention is given to using pentacene and graphene in spintronic devices. Although electronic devices containing high mobility pentacene crystals can be made, no working pentacene based spintronic device has been reported yet. On the other hand, graphene, being a single plane of carbon atoms as strong as diamond, inert to most gases and fluids, stable to temperatures up to 500 ^{o}C in air and having excellent electronic properties, has proven to be an excellent material for spintronic devices. Non-local spin transport experiments performed in our group show that the spin relaxation length in graphene, which is the distance from the spin injector at which the spin signal decreases by a factor e, can be as large as 2 μm at room temperature [12]. Since the mobility μ in graphene is high, (up to 2 10^5 cm^2 V^{-1} s^{-1}) [23] an application of an in-plane electric field E results in a finite drift velocity of the charge carriers (=μE) with a magnitude comparable to the Fermi velocity $v_F = 10^6$ m/s. This can be used to manipulate the spin relaxation length in a graphene-based spin valve device. Here, switching the electric field direction (along/against the spin transport direction) or changing the type of charge carriers (electrons/holes) using a gate voltage allows us to control the spin relaxation length considerably. In one case the spins move "upstream", since the spins need to travel against the action of the carrier drift to reach the spin detector, resulting to a reduction of the spin valve signal and of the spin relaxation length. In the other case the spins move "downstream", resulting to an enhancement of the spin valve signal. Experiments performed in our group show that we can enhance/reduce the spin relaxation length by a factor of 2 by applying a $\pm$700 Vcm^{-1} electric field on a 2.5 10^3 cm^2 V^{-1} s^{-1} high mobility sample [24]. Higher enhancement/reduction factors can be obtained by increasing the drift velocity, for example by using higher mobility graphene or stronger electric fields. Graphene seems to be a very promising material for spintronic and electronic devices. However, one of the main problems in using this remarkable system in large-scale production is that very high temperatures (>1000^{o}C) are needed for graphene growth. Here, chemistry could offer a solution to this problem as polycyclic aromatic hydrocarbons, which can be regarded as two-dimensional graphite segments, can be produced at much lower temperatures [25].

1.2 Motivation and Outline

In this thesis I present experimental work on spin transport in the 2-dimensional electron/hole system graphene and in a carbon nanotube which is a 1-dimensional conductor. I also present measurements in superconducting ultrathin tin nanowires encapsulated in multi-walled carbon nanotubes. The chapters in this thesis are:

Chapter 2:

Some basic theoretical concepts of spintronics are introduced. I discuss the conventional spin valve device and introduce the non-local spin transport technique. I develop a model for spin transport and precession in a nonmagnetic material including the role of the contacts. A number of electronic and spintronic properties of carbon nanotubes and graphene layers are presented.

Chapter 3:

Here I describe the fabrication techniques used for the production of graphene and nanotube spin valve devices. Also the fabrication steps needed for the fabrication of a tin-based superconducting device are presented. A large part of the chapter is used to discuss a number of experimental techniques which I used for the characterization of single graphene layers.

Chapter 4:

I present our first spin transport measurements in an organic system. Nonlocal and local spin transport measurements in a single-walled carbon nanotube are presented. We show that the magnetoresistance changes measured in the conventional two-terminal geometry are dominated by effects not related to spin accumulation. Combining our results with a theoretical model which can be found in Appendix A, we deduce a spin polarization at the contacts, α_F, of approximately 25 %.

Chapter 5:

Here I discuss the influence of the magneto-coulomb effect (MCE) on the magnetoconductance of spin valve devices. When two ferromagnets are weakly connected to a Coulomb island then an application of an external magnetic field can result to a change in resistance of the spin valve device. Here the magnetic field changes the work function of the ferromagnetic contact, which in turn induces an additional charge to the Coulomb island. We show that MCE can induce magnetoconductances of several percents or more, dependent on the strength of the Coulomb blockade.

Chapter 6:

I present our first non-local spin transport measurements in a single graphene layer. Measurements at room temperature show that spins can travel over 1.5 μm distances in graphene before relaxing. Graphene can therefore be used as a basic building block for spintronic devices.

Chapter 7:

Here I investigate anisotropic spin relaxation in graphene. Our experiments indicate anisotropic spin relaxation in this two dimensional system. For spins injected perpendicular to the graphene layer we found a relaxation time which is about 20% smaller compared to spins injected parallel to the graphene layer.

Chapter 8:

In this chapter I present experiments on drift of electron spins under an applied DC electric field in single layer graphene spin valves. The spin valve signals are increased/decreased, depending on the direction of the DC field and the carrier type, by as much as ±50%. We observe a sign reversal of the drift effect when switching from hole to electron conduction. A drift-diffusion model of spin transport is used to fit the experimental results.

Chapter 9:

I present electronic measurements on superconducting tin nanowires encapsulated in multi-walled nanotubes. The multi-walled carbon nanotube protects the monocrystalline tin nanowire from oxidation and shape fragmentation and therefore allows the investigation of the electronic properties of stable wires with diameters as small as 25 nm.

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