Spin transport in graphene - hexagonal boron nitride van der Waals heterostructures

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Chapter 1

Introduction and outline

Abstract

In the second half of the last century, we witnessed a revolution in microelectronic technology from the invention of transistor to powerful microprocessor chips in electronic devices. In order to overcome the current challenges of microelectronic devices such as the power dissipation and downscaling, researchers have been exploring an additional intrinsic property of electron, called spin. The field of spin electronics or spintronics has explored new spin related physics, including giant magnetoresistance, tunneling magnetoresistance, and spin transfer torque, which led to spin based device applications, for example, magnetic sensors in hard disk drive read heads, and magnetic random access memory data storage devices. Moreover, spintronics explores new type of materials that could host the transport of spins for long distances and durations. An atomically thin layer of graphene, discovered in the beginning of this century, holds the promise for spintronics applications due to the predictions of a large spin transport length and long spin relaxation time in this material. However, earlier experiments showed a lower magnitude of graphene's spin transport properties, and further research focused on finding the problems and overcoming the challenges that posed for such low performance of graphene based spin transport devices. The main challenges include finding a tunnel barrier for obtaining a consistent, and efficient spin injection and detection in graphene, and protecting the spin transport channel from the unwanted influence of the underlying substrate and the lithographic polymers. This chapter provides a brief history of the progress in spintronics with a focus on the current challenges in graphene spintronics, and a brief outline of the research work presented in this thesis.

1.1 Spintronics

Spintronics or spin electronics is a field of study that exploits the intrinsic spin angular momentum of an electron. Conventional electronics utilizes the charge degree of freedom of electron and focuses on improving the mobility or conductivity of the charge carriers. Whereas, spintronics utilizes the spin degree of freedom of an electron in addition to its charge state, and focuses on generation or manipulation of a spin polarized population of electrons, aiming at using the electron spins for efficient data storage and communication methods.

The origin of spintronics goes back to the first understanding of the electrical conduction in transition metals by Mott [1, 2] in 1936, who described the conduction of electrons in ferromagnetic (F) materials as a combination of two individual current channels, one channel consisting of electrons with spins parallel to the magnetization axis of F and the other with electron spins oriented in the opposite direction (anti-parallel). Due to an exchange splitting between the two spin subbands in the ferromagnet, the corresponding electrons at the Fermi level (E_f), which contribute to the electric current, have different densities of states (DoS) and conductivities. As a result, the current in F is spin polarized. The idea of the two channel model in F is further validated by Fert and Campbell [3–8] who studied the electronic transport of the doped ferromagnets.

The first proof for the existence of a non-equilibrium spin polarization in a material other than F is provided by Meservey *et al.* [9] in 1970, who showed that an application of a magnetic field in the plane of a superconductor (SC) results in a Zeeman splitting of its quasiparticle DoS. The spin-split DoS of the SC was utilized by Tedrow and Meservey [10–15] to determine the spin polarization of different Fs and their alloys by employing a spin polarized tunneling current technique to F/insulator(I)/SC tunnel junctions.

Instead of using a superconductor as a spin analyzer, Julliere [16] used a F to study the spin polarized conductance across F/I/F junctions, also known as magnetic tunnel junctions (MTJs), and reported that the tunneling of the spin polarized electrons across a F/I interface leads to a large change in the junction resistance when the magnetization orientation of the two Fs is changed from the parallel to the anti-parallel configuration. According to the model developed by the author, a relative conductance variation or a tunneling magnetoresistance (TMR) is defined based on the conductance of an MTJ in parallel (G_p) and anti-parallel (G_{ap}) orientations, given by, TMR = $\frac{G_p - G_{ap}}{G_{ap}} = \frac{2P_1P_2}{1-P_1P_2}$ where P_1 and P_2 are the spin polarization of two Fs.

Following the experiments of Meservey and Tedrow [10, 11] on the spin polarized tunneling in F/I/SC heterostructures, theoretical works of Aronov and Pikus [17], in 1976, suggested the possiblity of producing spin polarization in semiconductors(S) by flowing an electrical current from F into S, and Aronov [18] suggested an injection of non-equilibrium spins into metals, however with a small spin polarization. When a charge current is flowed across the interface of F with a nonmagnet (N) (either semiconductor or metal), a non-zero spin population will be created in N, near the interface, which leads to different electrochemical potentials for up-spin and down-spin electrons near the Fermi level, and their difference is called the spin accumulation. The non-equilibrium spin accumulation decays away from the interface into bulk. The characteristic distance over which the spin accumulation survives is called the spin diffusion length $\lambda_s = \sqrt{D_s\tau_s}$ where D_s is the spin diffusion coefficient and τ_s is the spin relaxation time of N.

According to a nonlocal device geometry proposed by Silsbee [19] in 1980, one can use a second F to detect the spin accumulation in N, before the complete decay of the spin accumulation, at a farther distance from the F/N injector interface. Indeed, it was later demonstrated experimentally by Johnson and Silsbee [20, 21] in 1985, that the non-equilibrium decaying spin accumulation can be detected as an electric voltage due to spin-charge coupling, and the spin relaxation time can be determined by the Hanle spin precession measurements. These measurements were realized at cryogenic temperatures in a single crystalline aluminium channel with Permalloy ferromagnetic injector and detector contacts in the nonlocal geometry. The authors also developed a theoretical framework [22, 23] to explain these results. In another independent attempt, van Son *et al.* [24, 25] theoretically gave a similar explanation on the charge-spin conversion at a F/N interface. 1

In a similar MTJ device geometry of Julliere [16] experiments, by replacing the insulator with a non-magnetic metal or semiconductor, the groups of Fert [26] and Grünberg [27] independently discovered the giant magnetoresistance (GMR) phenomenon in 1988. A typical GMR device consists of a thin non-magnetic material sandwiched between two F layers. The resistance of the device changes depending on the relative orientation of magnetization of the Fs which allows a spin dependent transmission of the conduction electrons between the F layers through the N layer. Early measurements on GMR devices were conducted by passing a current in the plane (CIP) of the layers. Later the GMR measurements were extended by Pratt *et al.* [28, 29] to the same device geometry where the current is now injected perpendicular to the plane (CPP) of the layers which showed similar GMR behaviour but with a higher magnitude. A theoretical model was proposed by Johnson [30] to explain the GMR behaviour in the CPP geometry on the basis of spin-coupled interface resistance formalism, which was previously developed by Johnson *et al.* [22, 23] and van Son *et al.* [24, 25] to understand the experiments involving the conduction of the spin polarized electrons from F into N [11, 31]. Another theoretical framework was developed on the basis of two-channel conductance for each spin direction to explain the GMR results of the CPP geometry [32, 33].

The multilayer structure of the GMR devices can be used as a magnetic sensor and has found its applications for reading the data in hard disk drives, storing bits of information in magnetoresistive random-access memory (MRAM) devices, and other devices [34]. The discovery of the GMR led to the 2007 Nobel prize in physics, awarded to Fert and Grünberg [35, 36].

Compared to the GMR device geometry, the nonlocal measurement geometry [19] has several advantages for studying spin transport in nonmagnetic materials. Detection of the spin accumulation in the nonlocal geometry avoids the spurious magnetoresistive signals that may arise due to the charge transport such as the anisotropic magnetoresistance or the Hall effect which may mask small spin signals due to spin injection in the GMR multilayer geometry. Moreover, a lateral spin transport geometry allows for the integration of multiple components, for example, backgate, topgate, and for applications in multiterminal devices, for example, three-terminal spin-flip transistor [37], spin-torque transistor [38, 39], spin field-effect transistors [40–43], and logic devices [44–46].

Despite the superiority of the nonlocal geometry, it was only Jedema *et al.* [47] in 2001 who demonstrated the electrical spin injection and detection at room temperature. The authors measured spin transport in all-metallic mesoscopic spin valves including a variety of materials [48] and demonstrated a controlled spin precession wherein the spin injection was achieved through an oxide tunnel barrier [49]. Thereafter a significant progress in understanding the spin transport in different non-magnetic systems was achieved including the zero-dimensional mesoscopic islands [50, 51], one-dimensional metallic channels [52–64], two-terminal metallic pillars [65, 66], semiconductors [67–69], superconductors [70–72], magnetic insulators [73, 74], toplogical insulators [75–78], carbon nanotubes [79, 80], graphene [81, 82], and transition metal dichalcogenides [83].

1.2 Spintronics materials

The spins undergo certain scattering processes during transport, causing the spin relaxation. The characteristic distance over which spins are transported is called the spin diffusion length (λ_s) and the characteristic time over which the spins do not dephase is called spin relaxation time (τ_s). The performance of a spintronics device is characterized based on these figures of merit. Therefore, it is required to find a material in which the electron spins can travel long distances without any scattering, i.e, a material retaining the spin polarization for a long duration.

When an unpolarized charge current is passed through a F conductor, the current becomes spin polarized due to unequal DoS for up-spin and down-spin electrons. A current carrying F can be used as a spin polarized current source or a spin injector for N. Moreover, F can also be used as a spin detector for sensing the spin polarized current in N due to spin-charge coupling [20]. However, naturally available ferromagnets viz., Fe, Ni, Co, and Gd, are not suitable for electrical transport of non-equilibrium spins due to the presence of a large spin orbit coupling (SOC) in such materials which leads to a very fast relaxation of spins, making these materials not suitable for data processing and data communication technologies.

On the other hand, non-magnetic materials, metals or semiconductors, have no spin polarization at equilibrium due to equal number of both spin states at E_f. However, an electrical current passing through a F/N interface can inject spin polarized carriers into N, near the interface. The injected non-equilibrium spin accumulation in N diffuses away from the interface towards the equilibrium region in the bulk of N with no spin accumulation. In metals, even though the spin backflow can be circumvented by inserting a tunnel barrier between the F and metal, the electrical spin transport in metals faces problem from a relatively short spin relaxation times, typically $\tau_s < 1$ ns. Moreover, large carrier density in metals makes it difficult to manipulate the carrier spins via a dielectric gate in a lateral spin valve device geometry.

Semiconductor(S) spintronics device physics has also been explored in parallel. Even though the presence of Schottky barrier at the F/S interface complicates the spin injection process, the N semiconductors exhibit a large room-temperature spin lifetimes above 1 ns and long spin relaxation lengths, and electric-field controllable carrier density which makes N semiconductors viable for the spintronics applications with a possibility of realizing logic, communication, and storage technologies within the same material [84].

Another class of materials, organics, are based on carbon element, and display a small spin-orbit coupling and negligible hyperfine interactions, the two phenomena deemed to cause relaxation of spins in a semicoductor material. These properties make organic materials promising for spintronics with a potential for large spin relaxation times. Newly introduced organic materials such as carbon nanotubes and graphene are non-magnetic, and are a main subject of interest for carbon-based spintronics. In fact, since the first demonstration of unambiguous spin transport in graphene by Tombros *et al.* [81], graphene has attracted much attention in the spintronics research community due to its theoretically predicted long spin relaxation time and large spin relaxation length at room temperature. Moreover, the magnitude of spin signal in graphene [85] is much higher than in metals [47] and semiconductors [67].

1.3 Motivation: Graphene spintronics

Two-dimensional graphene is attractive for spintronics due to a number of reasons. First and foremost is the theoretically predicted long spin relaxation time and large spin diffusion length which primarily result from the low SOC and negligible hyperfine interactions of carbon lattice [86, 87]. Besides, its large carrier mobility and electric field tunable carrier density make a compelling case for studying the spin dynamics in graphene.

The first demonstration of spin transport in graphene [81] showed small magnitudes of spin transport parameters, and the subsequent efforts [82] improved these parameters to the significant values by overcoming the challenges due to the underlying substrate, tunnel barriers, and the quality of graphene itself via various device geometries. However, there are few challenges still remain to bring graphene close to the spintronics applications.

First is the quality of graphene channel for spin transport. Even though graphene on hexagonal boron nitride (hBN) substrate was found to give a high electron mobility, its spin transport properties are limited by the lithography impurities on the top surface coming from the device fabrication process [88].

Second is the quality of ferromagnetic tunnel contacts for electrical spin injection into graphene. In the past decade, a big volume of spintronics research focused on finding a better way to effectively inject a spin polarized current into graphene via tunneling contacts having oxide tunnel barriers. The problem with conventional oxide tunnel barriers is two fold. One is the conductivity mismatch problem due to the inhomogeneous growth of tunnel barriers resulting in a frequent low-resistance contacts. This results in a low spin injection efficiency and small spin lifetimes in graphene. Even with the high resistive contacts, the spin lifetime reported to be very small. Therefore, besides the contact resistance, morphology of the tunnel barrier-graphene interface seems to play an important role in determining the spin lifetime in graphene [88]. This is possibly due to a direct growth of oxide barriers on graphene which might create roughness and dangling bonds at the interface.

Third is the efficiency of electrical spin injection and detection in graphene. Spin polarization is akin to fuel for spin transport. Most of the graphene spintronics studies were focused on aforementioned two challenges in order to achieve the large spin relaxation lengths and long spin relaxation times while using only a small spin injection polarization ≈1-10% in graphene which is just enough to study the basic spin transport physics in graphene. Even with high quality oxide tunnel barriers reported in the literature, the maximum value of spin injection polarization achieved was only up to 36% [89]. However, such high polarization values are rarely reported thereafter, probably due the irreproducibility of high quality tunnel barriers in different laboratories. In order to incorporate graphene spintronics to new avenues such as quantum dots and GMR-based technology, we need to achieve large spin polarizations by introducing new materials or new methods.

This thesis addresses these three problems by proposing a new device geometry for graphene spin valves with hBN tunnel barriers and substrate, and by introducing a method of applying a bias across the ferromagnetic tunnel contacts.

1.4 Thesis outline

A short summary of the chapters presented in this thesis is as follows,

- **Chapter 1** gives a brief introduction to the field of spintronics from both historical and technological perspectives, and the scope of the thesis in this field.

- **Chapter 2** describes the concepts of spin transport in general and derivation of spin signals in spin valve and Hanle measurements for two, three, and four-terminal measurement geometries. An account on various definitions of spin polarization and their relevance to the nonlocal spin transport is also given.

- **Chapter 3** gives a brief overview of the structural and electronic properties of graphene and hexagonal boron nitride materials.
- **Chapter 4** describes the experimental techniques used in this thesis work including the device preparation methods and measurement schemes.
- **Chapter 5** presents the first demonstration of spin transport in fully hBN encapsulated graphene where a monolayer-hBN is used as a tunnel barrier. Using this new spin valve device geometry we address the problems with the lithography impurities and the conventional oxide tunnel barriers [90].
- **Chapter 6** focuses on a bilayer-hBN tunnel barrier for electrical spin injection and detection in fully hBN encapsulated graphene. Here we achieve spin-injection and detection polarizations reaching up to 100% in ferromagnet/bilayer-hBN/graphene/hBN heterostructures using an electrical bias. Besides, we demonstrate two-terminal spin valve signals with a large magnetoresistance ratio which is attractive for practical graphene based spintronic devices [85].
- **Chapter 7** presents the spin injection and transport in graphene using two-layers of large-area chemical vapour deposition (CVD) grown hexagonal boron nitride tunnel barriers. Our results emphasize the importance of the quality of the CVD material growth and its transfer process for an efficient spin injection and transport in graphene spin valves. Moreover, the importance of the crystallographic orientation of the two layers of hBN is also discussed [91].
- **Chapter 8** gives a critical review of the experimental progress on spin transport in graphene, up until the work done in this thesis, emphasizing on the graphene/hBN heterostructures. A few current challenges and future perspectives are also given [92].

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