Charge and spin transport in two-dimensional materials and their heterostructures

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1 Introduction

Abstract

In this chapter, the need for scaling of field-effect transistors (FETs) and the bottleneck towards further scaling are discussed. Later, two-dimensional (2D) materials such as graphene and germanane are introduced; these materials could pave the way for fabricating even smaller FETs than the current silicon-based FETs. Further, a brief introduction is given into the field of spin-electronics (spintronics), which involves using the spin of an electron to compute logic instead of the conventionally used electron charge-based devices. New device concepts based on the crossroad of 2D materials and spintronics, such as transition metal di-chalcogenides/graphene heterostructures, are introduced. Finally, an outline of the contents of this thesis is listed.

1.1 Scaling of field-effect transistors

In this hi-tech world, numerous electrical gadgets are around us, be it a compact mobile phone or a large display television set. The basic building block of all these electronic gadgets is a field-effect transistor (FET)[1]. FET is a three-terminal device consisting of a gate, a source and a drain electrode; a semiconductor is connected between the source and the drain electrode, and the semiconducting channel is capacitively coupled to the gate. The operation of FET relies on the control of the current flowing through the semiconducting channel (between source and drain electrodes) by the gate electrode. No flow of current between the source and the drain electrode is regarded as '0', while a constant current flow is regarded as '1'; forming the basis for defining binary bits used to store or compute information in a FET. The semiconducting channel in the FET is usually composed of three-dimensional (3D) silicon[2] or III-V semiconductors like GaN[3]and GaAs[4].

For the past 30 years, Moore's law has driven the semiconductor industry in scaling the FET, so that our electronic gadgets could become faster and smaller. According to Moore's law, the number of FETs in an integrated cicuit (IC) doubles every two years[5]. Scaling of FET facilitates higher density of transistors in an IC, faster FET performance, less power consumption and cheaper cum large-scale IC fabrication. However, in few years from now, we will hit a bottleneck in the scaling of FETs as we are nearing the physical scaling limit down to the size of individual atoms at which the semiconductor becomes unstable and the scaling cannot continue further. This bottleneck is also due to the charge current leakage between the source-drain electrodes of FET (also resulting in power dissipation) as a result of a shorter semiconducting channel. Further, thinning down of 3D semiconductor leads to[6]:

1. Dangling bonds at the surfaces, resulting in the scattering of charge carriers.
2. Mobility (μ) degradation, since mobility is directly proportional to the thickness (t) of the 3D semiconductor as, $\mu \alpha t^6$.
3. Increase in the Bandgap (E_g) of the material, since the bandgap is directly proportional to the thickness (t) of the 3D semiconductor as, $E_g \alpha t^2$.

In order to continue scaling, we need to explore new channel materials and devices based on alternative logic.

1.2 Two-dimensional materials

It is predicted by scaling theory that, FET with a thin oxide dielectric and a thin gate-controlled channel region would be robust against short-channel effects, down to very short gate lengths[7]. Hence, single layers of two-dimensional (2D) materials, which are only an atomic layer thick, seem to be very attractive in their use as channel and oxide materials for new generation of FETs. Contrary to limitations with thinning down of 3D semiconductor as explained in the previous section, 2D materials offers various advantages like:

1. There exists numerous 2D materials with different bandgap magnitudes and types, which could be metallic, or semi-metallic, or semiconducting, or insulating.
2. Naturally passivated surface exists in 2D materials, and hence, there are no dangling

bonds for a stable and perfect crystal of 2D material. Also, 2D materials are perfectly flat unlike 3D semiconductors that exhibit thickness variation or film discontinuity leading to scattering of the charge carriers.

3. Charge carrier mobility of 2D material like graphene[8] is around 10,000 $cm^2V^{-1}s^{-1}$, while that of 3D silicon[9] is only ~1,400 $cm^2V^{-1}s^{-1}$.

1.2.1 Graphene

In 2004, Andre K. Geim and K. S. Novoselov discovered graphene in search of new channel materials for FET[8]. Graphene is 2D in nature with naturally occurring atomically thin layers of ~0.35 nm thickness made up of honeycomb lattice of sp^2 carbon atoms. Further, the charge carriers in graphene can be uniformly controlled by applying a gate voltage. Graphene offers a high charge carrier mobility[8] of ~2,00,000 $cm^2V^{-1}s^{-1}$ (theoretically) enabling fabrication of high-performance devices alongside window to study new physics at quantum scale due to its two dimensionality[10]. However, graphene in its pristine form lacks bandgap, which is essential for switching action in logic circuits[11]. Efforts to introduce a bandgap in graphene by making nanoribbons of graphene[12] or chemically functionalising the graphene surface[13] has always resulted in complexity in device fabrication and in mobility degradation.

1.2.2 Germanane

As mentioned before, although graphene promises excellent charge carrier mobility, it still lacks a bandgap which is necessary to achieve turning ON and OFF of the FET. A search for an alternative has resulted in the discovery of other 2D materials like germanane, transition metal-dichalcogenides (TMD), hexagonal boron nitride (hBN), Bismuth Selenide (Bi_2Se_3), black phosphorus and many others[14]. Germanane (GeH) is a 2D material with a hexagonal lattice of germanium (Ge) atoms, wherein alternate Ge atoms are covalently bonded to hydrogen (H) atoms in up and down direction respectively. Germanane used in this thesis was synthesised for the first time by Bianco et al.[15] using topochemical deintercalation of $CaGe_2$. The electron mobility in GeH, which is limited by electron-phonon scattering, was calculated to be ~20,000 $cm^2V^{-1}s^{-1}$ at room temperature[15]. The calculated electron mobility in GeH is about five times that of germanium which is promising towards realising faster FETs.

The first-ever realisation of FET from GeH is reported in chapter 5. We found that the GeH transistor is ambipolar; hence, one can use GeH FET in the realisation of complementary metal-oxide semiconductor (CMOS) devices. Furthermore, GeH transistor showed electrical response upon shining light which is useful in building optoelectronic devices.

1.3 Spintronics

Conventional electronics utilises the charge property of electrons. However, electrons contain not only charge information but also angular momentum, which is called spin. Moreover, the spin of an electron can either point up or down corresponding to ‘1’ and ‘0’ states for logic applications and the electronics built using the spin aspect of an electron is called spin electronics, rather abbreviated as spintronics. Spintronics offers low power operation and faster switching times since the power required to switch the spin of an electron is low, and the switching speed is faster than the conventional charge-based electronics.

Spintronics has its roots in Mott's theoretical work in 1936, wherein he presented a simple two current model for spin-polarised transport[16,17]. In 1985, Johnson and Silsbee for the first time demonstrated the spin injection from a ferromagnetic metal into pure aluminium at 4K[18] temperature. In 1988, Albert Fert[19] and Peter Grünberg[20] discovered giant magneto-resistance (GMR) for which they won the Nobel Prize in Physics in 2007. To understand GMR, let us consider a pair of ferromagnets sandwiching a conductive non-magnet in between. This multilayer system offers a significant change in the electrical resistance depending on whether the adjacent ferromagnetic layers have their magnetisation in parallel or antiparallel configuration. In the case of parallel magnetisation, the overall resistance is low while it is high for anti-parallel configuration. By applying an external magnetic field, one can control the magnetisation direction of the ferromagnets. GMR has been used by IBM to develop read heads of computer disc in 1997 significantly increasing the storage capacity.

Furthermore, if a thin insulator separates the ferromagnets in GMR instead of a non-magnetic conductive layer, it leads to the observation of tunnel magnetoresistance (TMR). The basic working principle of TMR is: if the magnetisation of the ferromagnet are parallelly aligned, the probability of electrons tunnelling across the insulating barrier is higher than when they are aligned anti-parallelly[21]. TMR is the basis for the operation of magnetic random access memory (MRAM), which is a new type of non-volatile memory. It was also observed that, using the concept of spin-transfer torque, the magnetisation of an electrode could be switched by spin-polarized electrons [22]. The concept of spin-transfer torque is used in the development of spin-transfer torque MRAM (STT-MRAM), which has lower power consumption and better scalability over MRAM[23].

1.3.1 Graphene spintronics

Graphene has attracted huge interest as an excellent material for spin transport; partly because of the theoretically predicted long spin relaxation length and time[24,25]. This long spin relaxation length and time in graphene is mainly due to the presence of low spin-orbit coupling (SOC) and negligible hyperfine interaction. The first experimental demonstration of spin transport in graphene was shown by Tombros et al.,[26] who found a spin relaxation length of ~2 μm at room temperature which was weakly dependent on the charge carrier density in graphene. Since then, extensive research has been carried out[27] exploring different substrates, tunnel barriers and the quality of graphene to obtain longer spin relaxation length. The highest experimentally reported[28] spin relaxation length is ~30 μm at room temperature, wherein the graphene is covered with hBN and suspended over Co/MgO electrodes. Further, recent studies have probed spin transport in fully hBN encapsulated graphene[29], alongside using hBN as an efficient tunnel barrier[30].

Unlike single-layer graphene, bilayer graphene (BLG) exhibits an electronic bandgap in the presence of an electric field; this electric field could further tune the spin transport in BLG. Fully encapsulated BLG in hBN has been studied[31], showing gate tuneable spin relaxation length of ~24 μm. Further, it has also been observed[32] that the spin relaxation length can be increased to ~90 μm using carrier drift by applying direct current (DC). Anisotropy in spin relaxation times for out-of-plane spins to in-plane spins in hBN encapsulated BLG is around 8±2 near the charge neutrality point at 75 K[33], and up to ~12 at 100 K[34].

Seeing the potential in graphene as the future and emerging technology, the European Commission has invested 1 billion euros for graphene research under the umbrella of Graphene Flagship. "The Graphene Flagship is tasked with bringing together academic and industrial researchers to take graphene from the realm of academic laboratories into European society in the space of 10 years, thus generating economic growth, new jobs and new opportunities," reads an excerpt from www.graphene-flagship.eu. Further, a work package in the Graphene Flagship is exclusively set up on spintronics towards the engineering of efficient room-temperature spin injection and detection, long-distance spin transport, spin gating, and spin manipulation in graphene devices.

1.3.2 Spintronics with graphene-TMD heterostructures

Although graphene promises long-distance spin transport, it has weak intrinsic spin-orbit coupling (SOC), which is essential in the electrical control of spin transport in order to realise the spin polarized field-effect transistor (Spin-FET)[35]; Spin-FET promises logic operation based on the spins rather than the conventionally used charge of electrons. However, there exists other 2D materials like TMDs, having lower electron mobility than graphene but stronger spin-orbit coupling. These TMDs can be placed in the proximity of graphene to induce SOC in graphene[36,37], resulting from the hybridisation of the d-orbitals of TMD with the π-orbitals of graphene. Also, the spatial inversion symmetry is broken at the graphene/TMD interface, leading to Rashba type SOC in graphene; the SOC can further be tuned by applying an electric field, which allows the electrical control of spins.

For TMD-graphene heterostructures, reports suggest a reduced spin life time[38,39] and its further tuning by electrical gating[39] with a modulation of the spin current[39,40], enabling their use as 2D spin field-effect switch. However, these experiments do not explore the spin lifetime anisotropy present for the spin transport in graphene under the TMD proximity. Recently, there have been reports observing anisotropy in the spin relaxation time in graphene encapsulated by single[41] ($MoSe_2$) and multi[42] (WS_2) layer TMD. In chapter 6, we study the tungsten disulfide (WSe_2) on graphene heterostructures, and show that the spin lifetime anisotropy exists in these heterostructures. Further, we show that the spin lifetime anisotropy in graphene under the TMD proximity can also be studied by probing the non-local spin transport in the nearby graphene region. In addition, we also demonstrate the spin injection through WSe_2 into graphene in chapter 6.

For a monolayer TMD, the location of the conduction and valence band edges are at non-equivalent K points (K and K′) of the 2D hexagonal Brillouin zone known as the valleys. Further, the valence and the conduction bands at the K and K′ valleys are spin split due to a strong SOC, and this leads to a coupled spin and valley degree of freedom in the monolayer TMD. The charge (coupled to spins) transition between valence and conduction band in the K and K′ valleys can be achieved by using a right and left circularly polarised light respectively. Recently, valley polarised electron spins were generated using circularly polarised light in TMDs[43,44]; further, these generated spins were injected into graphene and were detected non-locally via ferromagnetic contacts.

Electronic band structure of a bi-layer graphene (BLG) on WSe_2 has been studied from first principles by Gmitra et al.,[45] and they found that an indirect bandgap of 12 meV opens up in BLG due to the built-in electric field across the BLG/WSe_2 heterostructure. Interestingly, the split in valence band due to induced SOC is huge (of the order 2 meV) and is two orders of magnitude higher in comparison to that of the conduction band. Further, the intrinsic bandgap of BLG on WSe_2 can be tuned or even reversed (the characters of the valence and conduction bands flip and we get a spin-orbit valve) by the application of an external electric field of the order 1 V/nm. Along this direction, we studied spin-transport in BLG on tungsten disulfide (WS_2) heterostructures, presented in chapter 7; we recorded huge spin lifetime anisotropy of ~40-70, which is the ratio between the out-of-plane ($\tau_\perp$) and the in-plane ($\tau_\parallel$) spin relaxation time. A new technique was used to calculate the spin lifetime anisotropy, which we developed in-house called the oblique spin-valve measurement.

Presented in chapter 8, we studied WSe_2 on BLG, wherein we clearly show that there is no spin transport of in-plane spins across the WSe_2 covered BLG, while the out-of-plane spin transport is unaffected. We also show that the spin lifetime anisotropy for in-plane and out-of-plane spins can be extracted by studying the non-local spin transport in BLG which is close to WSe_2 covered BLG. Furthermore, we have demonstrated the spin injection through WSe_2 into BLG.

1.4 Outline of the thesis

Here is a brief look at the outline of the contents of this thesis.

Chapter 1: In this chapter, the need for scaling and the bottleneck faced in the scaling of conventional silicon-based FETs are briefly discussed. Later, the new generation of materials which are fundamentally two-dimensional in structure like graphene, germanane, etc., are introduced; and their benefits to be used as a channel material for FETs in the future are noted. Further, an introduction and a brief history of the research undertaken in the field of spin-electronics (spintronics) in discussed. The section on spintronics, dwells more into the emerging area of study on spintronics in graphene and graphene-TMD heterostructures.

Chapter 2: Basic structural and electronic properties of two-dimensional materials used in this thesis are introduced in this chapter. The materials include germanane, graphene (single and bi-layer), transition metal di-chalcogenides (WS_2, WSe_2), and their heterostructures.

Chapter 3: In this chapter, the concepts necessary to understand the electronic and spin transport in two-dimensional materials are introduced; starting with a comparison of the electronic charge transport properties of bulk and isolated two-dimensional materials. Followed by a short introduction to the working principle of the field-effect transistor. Later, the concept of spin injection into non-magnetic material like graphene is discussed, and the two-channel model for spin transport in a typical spin-valve is briefly explained. The issues of conductivity mismatch and spin relaxation induced by contacts are also addressed in this chapter. Further, description on the spin transport in graphene is provided, including concepts on spin diffusion equation, non-local spin-valve (non-local SV) measurement, and Hanle spin precession measurement. Later in the chapter, various spin relaxation mechanisms for spin transport in graphene are discussed. Lastly, a short overview of the theoretical and experimental studies on spin transport in single and bi-layer graphene in proximity of TMD, are presented.

Chapter 4: In this chapter, the protocol for fabricating field-effect transistors and non-local SVs of 2D materials studied in this thesis, are presented. In the fabrication section, the mechanical exfoliation and the realisation of 2D materials heterostructures using pick-up technique are discussed. In the last section of this chapter, the electrical and magnetic experimental characterization setup and techniques, used to study the 2D flakes and their heterostructures, are briefly outlined.

Chapter 5: The fabrication and electrical characterisation of the first ever field-effect transistor made of multi-layer germanane are described in detail in this chapter. Here, it is shown that the germanane FET has charge transport in both electron and hole doped regimes with ON-OFF current ratio of up to 10^5 (10^4) and charge carrier mobilities of 150 $cm^2V^{-1}s^{-1}$ (70 $cm^2V^{-1}s^{-1}$) at 77 K (room temperature). A significant enhancement of the FET conductivity is illustrated under an illumination with 650 nm red laser. Both ambipolar charge transport and opto-electronic response observed in germanane FET has great potential for applications in CMOS and (opto)electronics.

Chapter 6: In this chapter, the fabrication and characterisation of non-local SV of graphene/WSe_2 heterostructures are explained. The study of the proximity induced SOC in graphene by WSe_2 via non-local SV and Hanle spin precession measurements are presented. The key observations discussed in this chapter are:

1. No spin transport was observed in graphene over WSe_2 covered region longer than 3 μm, which alludes to a large magnitude of the proximity induced SOC in graphene by WSe_2.
2. Observation of anisotropy in spin lifetime for in-plane and out-of-plane spin transport. Further, the proximity induced SOC in graphene by WSe_2 could be non-locally determined in the nearby graphene region not covered by WSe_2; which could be due to the diffusing nature of the itinerant spins which explore the WSe_2 covered graphene region.
3. Use of multi-layer WSe_2 as an intermediate layer for spin injection into graphene.
4. Modelling of Hanle curves using time-independent Bloch-diffusion equation for spin transport in graphene under, near and far from the WSe_2 covered graphene region. These modelled Hanle curves showed similar behaviour as our experimental observations.

Chapter 7: In this chapter, the fabrication and characterisation of non-local SV of bi-layer graphene/WS_2 heterostructures are presented. A new technique we developed called oblique spin-valve measurement is described; we used this technique to determine the spin lifetime anisotropy for in-plane and out-of-plane spins in bi-layer graphene, arising due to induced SOC by WS_2 substrate. With this measurement we were able to measure huge spin lifetime anisotropy of ~40-70, the ratio between the out-of-plane ($\tau_\perp$) and in-plane spin relaxation time ($\tau_\parallel$). Further in this chapter, the origin of such high spin lifetime anisotropy in BLG coupled with WS_2 is discussed.

Chapter 8: This chapter details the fabrication and characterisation of non-local SV of bi-layer graphene (BLG)/WSe_2 heterostructures. The spin transport studies carried out on this heterostructure is similar to the studies carried out on single layer graphene/WSe_2 heterostructure, discussed in chapter 6. The key observations discussed in this chapter are,

1. Using Hanle precession measurement at a high magnetic field we measured anisotropy in spin relaxation time for in-plane ($\tau_\parallel$) and out-of-plane ($\tau_\perp$) spins in BLG, which is covered by WSe_2, with $\tau_\perp/\tau_\parallel = 3.64$.
2. Anisotropy in the spin relaxation time present in BLG covered by WSe_2, could be non-locally determined in the nearby region of BLG not covered by WSe_2; this could be due to the diffusing nature of the itinerant spins which explore the neighbouring WSe_2 covered BLG region.
3. Due to the presence of anisotropy in $\tau_\parallel$ and $\tau_\perp$, any in-plane spin transport across the WSe_2 covered BLG was not observed, although an out-of-plane spin transport was detected.

The FETs of WSe_2/BLG in vertical geometry, which showed an n-type behaviour with a current ON-OFF ration $>10^3$, are also discussed in this chapter.

Chapter 9: In this chapter, a brief discussion on the conclusions of all the chapters discussed in this thesis, are presented.

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