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

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Summary

In the second half of the last century, we witnessed a revolution in microelectronics technology towards digital electronics, from the invention of transistor to the powerful microprocessor chips in our computers and smart phones. The electronic circuits in these devices express the data as binary digits or bits, 0 and 1. This technology created a billion-dollar industry over the years and now the electronic devices are getting miniaturized to fit in our pockets as a result of packing more logic devices into every chip. Currently, as of 2017, we have nearly reached the end of the Moore's law as the size of an individual bit approaches the dimension of atoms, 1 nanometer. In such compact devices, power dissipation is itself a challenge.

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, purely a quantum mechanical property. Spin of an electron is roughly akin to the spinning of a child's top or to the directional behavior of a compass needle. The top or compass needle can rotate clockwise or anticlockwise, equivalent to the electron's spin which has up-spin and down-spin directions in relation to the magnetic field. One advantage of spin over charge of electron is that the spin of an electron can carry information which can be manipulated by the external magnetic field without consuming or dissipating power. The very first devices of this relatively new field of research, called Spintronics, have already surfaced and have been commercialized, for example, the properties of electron spins are already in use in magnetic sensors and magnetic random access memories in the hard disk drives of our day to day electronic devices.

Moreover, in order to overcome the challenges with downscaling or miniaturizing the next generation spintronic devices, researchers are also exploring new type of materials that could host the movement of spins for a large distances and long durations. Graphene, a one-atom thick two-dimensional layer of carbon atoms, discovered in the beginning of this century, holds the promise for spintronics applications due to the predictions of a large spin transport length and a long spin relaxation time in this material. However, in practice, the transport of spins in graphene is difficult due to randomization of spins after a short while, and the present research community is focused on finding the problems and overcoming the challenges posed for such low performance of graphene based spintronics devices.

It has been nearly a decade since the first demonstration of the spin transport which showed relatively small spin relaxation time and spin diffusion lengths. The subsequent efforts in this field suggest that the underlying substrate, impurities on graphene's surface and the quality of the contacts play a crucial role in influencing its transport properties. The research work presented in this thesis contributes towards addressing these issues altogether using a new type of device geometry.

This thesis presents for the first time a new device structure for graphene spintronics where graphene is fully encapsulated between two hexagonal boron nitride (hBN) layers to address the challenges due to the substrate, the effect of contacts, and the inhomogeneity in the charge and the spin transport properties.

Several experiments suggest that one important factor limiting the spin transport in graphene is the quality of ferromagnetic tunnel contacts which consist of ferromagnet(FM)/tunnel-barrier/graphene junctions. In Chapter 5, we tried to address this issue using a single-layer hBN tunnel barrier in place of the conventional oxide barriers. Additionally, the same tunnel barrier layer protects the graphene underneath from coming in contact with the lithographic impurities due to the lithographic processes.

The presented charge transport characteristics in this device show that the extracted charge mobility values lie close to each other for different regions of the encapsulated graphene. The spin transport measurements resulted in consistent spin relaxation parameters which do not differ much for the different regions of the encapsulated graphene. Besides, we also observed spin transport across a 12.5 μm graphene channel encapsulated under a monolayer-hBN-tunnel barrier even in the presence of conductivity mismatched electrodes, indicating the pin-hole free nature of hBN and the clean interface of hBN with graphene.

These results indicate the potential for using hBN as a substrate and as a tunnel barrier for investigating the intrinsic spin transport properties of graphene in a clean environment. However, due to the relatively low interface resistance-area R_cA product, with monolayer-hBN barriers, there is a need to use a higher number of hBN layers for non-invasive spin injection and detection. The value of R_cA product quantifies the backflow of spins to FM. Moreover, large spin-injection polarizations have been predicted in FM/hBN/graphene systems as a function of bias with increasing number of hBN layers.

Biasing ferromagnet/thin-hBN/graphene systems has not been explored in the literature, whereas it is predicted to show rich physics in terms of creating electric-fields, electrostatic gating of the graphene, and potentially inducing magnetic proximity exchange splitting in graphene. In order to explore such phenomena, in Chapter 6, we study how the differential spin-injection and detection polarizations depend on the electric field applied across the cobalt/bilayer-hBN/graphene tunneling contacts. Our spin-transport measurements at room temperature reveal surprisingly large bias induced differential spin-injection and detection polarizations up to ±100%, and a unique sign inversion of polarizations near zero bias.

Using the large values of spin-injection and detection polarizations, we demonstrate a two-terminal spin-valve with a record magnitude of the differential inverted spin signals up to 800 Ω and magnetoresistance ratio of 2.7%. Moreover, we also observe unambiguous evidence of spin transport in the two-terminal measurement geometry via Hanle spin precession measurements using the bilayer-hBN tunnel barrier contacts, given in Chapter 8. This is the first demonstration of a two-terminal Hanle signal and these results are technologically desirable for practical graphene spintronic applications.

We also provide suggestions to improve the two-terminal magnetoresistance signals even further and enhance spin-accumulation in graphene beyond room-temperature thermal energy, k_BT, which would open up a completely new regime to study the spin transport in graphene. These results indicate the uniqueness of bilayer-hBN tunnel barriers for achieving unprecedented large spin-injection and detection polarizations in graphene in a fully hBN encapsulated environment. Furthermore, a two-terminal spin-valve effect with such controlled values of the large spin-accumulation and high magnetoresistance ratio is a step closer towards realizing practical graphene based spin field-effect transistors.

So far, we used exfoliated-hBN as a barrier for electrical spin injection and detection. This material is of higher quality and can be obtained easily by repeated peeling of the layers of hBN crystals down to the mono, bi or tri layers. However, for the future industry-scale applications, it is important to produce large scale chemical vapour deposition (CVD) grown materials. In this respect, in Chapter 7 we study the electrical spin injection in graphene using a layer-by-layer-stacked two-layers of CVD grown hBN tunnel barrier.

Ferromagnetic contacts with large-area CVD grown hBN tunnel barriers were reported to give high spin injection polarizations in graphene. It has also been reported that the high-mobility graphene on hBN substrate shows long distance spin transport. In this chapter, we address the possibility of combining high-mobility graphene on a hBN substrate with a large-area two-layer-CVD-hBN tunnel barrier for efficient spin transport.

We observe low-mobility and small spin relaxation time in our devices, due to the degraded quality of graphene by the traditional wet transfer of CVD-hBN. We also find both positive and negative differential spin polarizations for two-layer-CVD-hBN barrier contacts. Moreover, unlike exfoliated-hBN barrier contacts reported in Chapter 6, the spin polarization does not change its sign within the range of ±0.3 V bias, and its magnitude increases only at large negative bias values. These features mark a distinction between the two-layer-CVD-hBN, and the bilayer-exfoliated-hBN tunnel barriers reported in Chapter 6.

A large magnitude of spin polarization up to 15% at -0.2 V bias for two-layer-CVD-hBN barriers contacts indicates the potential of using CVD-hBN tunnel barrier for large-area spintronic applications. Moreover, considering the obtained results, we emphasize that, along with the thickness of the tunnel barrier, other parameters play a significant role in determining the spin tunneling characteristics such as, the quality and the relative alignment of two monolayers of hBN. From these results, we address the fundamental nature of the spin injection via a CVD-hBN tunnel barrier which is important for the spintronics community and also provides a deeper insight about the possibility of using large-area CVD-hBN for future commercial applications.

In conclusion, the results presented in this thesis represent important developments towards understanding the nature of spin transport in graphene and spin injection via hBN barriers. This understanding will certainly be helpful in overcoming the challenges in realizing practical spintronic devices based on graphene-hBN van der Waals heterostructures.

In the light of the growing interest in graphene spintronics, in Chapter 8 we review the progress that has been made to reach the current status of graphene spintronics research with respect to finding a suitable substrate for effective spin transport and a tunnel barrier for efficient spin injection and detection. We also discuss the recent state-of-the-art findings on spin transport in graphene-hexagonal boron nitride heterostructures and identify current challenges for realizing true potential of spin transport in graphene. We give our perspectives on what hBN entails for making heterostructures with graphene for exploring interesting physics and realizing spin based device applications.