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## Charge and spin transport across graphene and multifunctional oxide interfaces

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### 1.1 Magnetism and spintronics

Magnetism has played a crucial role throughout human history. People invented the first compass as early as the ancient Han dynasty (ca. 206 BC) in China, based on magnetic principles. Due to this invention, people could explore and connect the world. However, understanding of how the compass works by the general public was absent until 1600 AD, when William Gilbert published the first scientific book on magnetism. He investigated the magnetic field of the earth and gave insight into the working principle of the compass. Another milestone in the human's exploitation of magnetism is the discovery of the relationship between electricity and magnetism. This led to the booming development in both fundamental understanding and applications of magnetism.

With the development of quantum mechanics, it is now understood that the atomic origin of magnetism lies in electron spins. Meanwhile, when investigating thin films of magnetic materials, researchers discovered a new field of spin transport, which later on developed into a separate subject: spintronics, short for spin electronics. Spintronics uses spin as the carrier of information and thus brings a new degree of freedom compared to conventional electronics, which only depends on electron charge as the information carrier. A pioneering work by Sir Nevill Mott on the resistivity of transition metals and their alloys kicked off the era of spintronics in 1936. To explain the abnormal resistivities observed, he introduced a two-current model, in which the contributions of the down-spin electrons and up-spin electrons to the conductivity are treated independently.[1] Since then, the researchers started to realize the influence of the spins on the electronic transport properties.

Spintronics involves multidisciplinary research, such as magnetism, semiconductor physics, mesoscopic physics. The main goal of spintronics is to control the electronic properties using the spin degree of freedom, and vice versa. The beginning of the spintronics era is marked by the discovery of giant magneto-resistance (GMR) by Albert Fert and Peter Grunberg[2, 3], who were awarded the 2007 Nobel Prize in Physics. GMR was observed in the magnetic heterostructures consisting of two ferromagnetic (FM) layers and a non-magnetic layer in between. This device showed

different resistance depending on the relative magnetization alignment of two FM layers as a result of spin-dependent scattering. This fundamental scientific work of GMR was rapidly applied to the data storage technologies as a method of data reading, which accelerated the rate of areal information density growth.

Spintronics plays a role in magnetic random access memory (MRAM, primary data storage) and the computer's hard drive disk (HDD, secondary data storage). Both are facing fierce competition from their electronic counterpart. The main bottleneck for the spintronic storage is the downscaling. A revolutionary concept to overcome this bottleneck was to use spin-transfer torque (STT) in the MRAM.[4] Instead of switching the spins by a magnetic field, STT-MRAM switches the spins by a polarized spin current due to spin-transfer torque, which results in higher storage densities, lower energy consumption and reduced cost. What is more, the main advantage of STT-MRAM over the electronic-based RAM is its non-volatility and near-zero leakage power consumption. Due to all these superiorities, STT-MRAM is the new rising star in the memory industry.

Besides the application of spintronics in data storage, the spin-based logic device has been proposed. In 1990, Datta and Das came up with the proposal of spin field-effect transistor (spinFET)[5], where the electric field can control the spins in the semiconductor channel via spin-orbit coupling and thus the device can be switched on or off. Since then, different variations of the Datta-Das transistor have been proposed.[6–8] Although the spinFET has not been realized yet, it allows for a possibility of all-spin computing with faster processing speed, lower power consumption, larger storage density and nonvolatility, compared to conventional electronic-based computing.

Nowadays, the field of spintronics is flourishing. New concepts, such as (inverse) spin Hall effect[9–11], spin pumping[12], spin Nernst effect[13, 14], magnon transport[15], and magnetic skyrmions[16, 17], invigorate the field. New materials, such as two-dimensional materials, topological insulator, are utilized in the field of spintronics, offering new platforms. Eventually, all the efforts lead to new applications, such as quantum computing using qubits based on the spin state of a quantum box[18] and TMR sensors[19].

## 1.2 Graphene spintronics

Graphene is an ideal candidate to be used in spintronic devices. Its superior intrinsic mobility, low spin-orbit coupling, and weak hyperfine interaction allow for long spin relaxation time and length.[20–22] Since the first demonstration of spin transport in graphene, it has been intensively investigated in terms of spin transport. Besides graphene alone, we can stack different two-dimensional materials with graphene

to form two-dimensional heterostructures. Recently, considerable new phenomena are emerging in graphene and its van der Waals heterostructure: proximity effect, electrical tunability, magnetism, and spin-light coupling.

There are two main challenges in this field. One challenge is the control of spins in graphene. This is due to the low spin-orbit coupling. To solve this problem, researchers made use of the proximity effect, where graphene can inherit the properties from the neighbor material. Thus, the neighboring transition metal dichalcogenides or topological insulators can introduce spin-orbit coupling in graphene. The other challenge is that the discrepancy between the theoretical prediction and the experimental observation of the spin relaxation time has driven the researchers to scrutinize the dominant spin relaxation mechanism.

### 1.3 Functionalize graphene with the complex oxides

An innovative approach to manipulating the charge and spin transport in graphene is to functionalize its interface with multifunctional oxides. Multifunctional oxides possess diverse functional properties, ranging from ferroelectricity, multiferroicity, tunability of resistance with thickness, high spin-polarization in half-metal oxides, large temperature-dependent dielectric constant, etc. Therefore, at the interface of graphene with the oxides, we can expect different functionalities from both materials. For example, the spin-orbit coupling might be modified. The spin-efficiency of transport can be maximized by interface engineering. Spin and charge transport can be manipulated by switching the electric polarization and magnetization of ferroic materials. In short, combining graphene and strongly correlated transition metal perovskites empowers the interface with novel functionalities, which can be tailored to open up new possibilities for new information technology beyond CMOS.

### 1.4 Thesis outline

This thesis explores the spin and charge transport in graphene and complex oxides. There are two parts in this thesis. In the first part, from chapter 2 to chapter 4, the essential material knowledge, theoretical concepts, and fabrication techniques are discussed. In the second part, from chapter 5 to chapter 8, the scientific findings of this thesis are discussed. A brief overview is given below:

- *Chapter 5* reports the first investigation of spin transport in graphene on SrTiO<sub>3</sub> and discusses its temperature and electric field dependence. Peculiar non-monotonous temperature dependence of the spin transport parameters in

graphene is present. We show that spin transport in graphene on SrTiO<sub>3</sub> is influenced by the surface electric dipoles, intrinsic spin-orbit coupling in SrTiO<sub>3</sub>, and temperature-induced rippling of the graphene due to the phase transition of SrTiO<sub>3</sub>. Furthermore, we find that an electric field can control the spin transport parameters, which is attributed to the modulation of the strength of surface dipoles in SrTiO<sub>3</sub>.

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- *Chapter 6* investigates the temperature and gate bias dependence of charge transport in graphene on SrTiO<sub>3</sub> with and w/o intermediate layer hexagonal boron nitride (hBN). We find that the anti-hysteresis in the gate dependence of the graphene resistance is related to the movement of oxygen vacancies, and domain walls imprint their potential landscape in graphene.

Besides SrTiO<sub>3</sub>, another attractive complex oxide is BiFeO<sub>3</sub>, whose ferroelectricity and antiferromagnetism are coupled. However, the surface magnetic properties of BiFeO<sub>3</sub> are not well understood.

- *Chapter 7* deviates from graphene study and examines the surface magnetic properties of BiFeO<sub>3</sub> of different strains using spin Hall magnetoresistance and spin Seebeck effect. We demonstrate different magnetic orders in BiFeO<sub>3</sub> of different strains, which possess different antiferrodistortive and ferroelectric distortions.
- *Chapter 8* investigates the influence of the BiFeO<sub>3</sub> on the graphene transport properties by studying the graphene magneto-transport. We report linearity in graphene magnetoresistance, which indicates inhomogeneity in the carrier densities and mobilities of graphene caused by the local gating effect from BiFeO<sub>3</sub> domains. Furthermore, from the weak localization analysis, we have extracted the spin relaxation time to be 1.2 ps. Such low spin relaxation time is attributed to the strong influence of local magnetic moments in BiFeO<sub>3</sub>.

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