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Proximity-induced spin-orbit and exchange coupling in graphene-based heterostructures

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Summary

The information age began with the invention of transistors in 1947. Since then, countless technological breakthroughs have enabled the realization of smaller and faster electronic devices, mainly based on the principles of field effect transistors (FETs) to perform logic operations. This process led to the progressive miniaturization of electronic systems that well followed the down-scaling trend predicted by Moore in 1965. However, considering the fundamental limits at the nanometer-scale that are enforced by e.g. the excessive energy dissipation, we would need to explore other ways to transport and store information in order to maintain the progress in the computing speed and power efficiency.

Spintronics or spin-based electronics offers alternative logic operations. The working principle of spintronic devices is based on the quantum mechanical property of an electron, spin, which is a measure of its intrinsic angular momentum. The main objectives in spintronics are to be able to maintain and transport the spin information over micrometer-length scales, and also to manipulate and store the spin signal for the integration of memory elements with logic circuits. The physics of spintronics was applied in the computer industry after the discovery of giant magnetoresistance (GMR) effect in the 1980s. Due to the GMR effect the resistance of a circuit containing two ferromagnets that are separated by an ultra-thin non-magnet depends on the relative orientation of the magnetization of the ferromagnets. The difference in resistance levels associated with the parallel and anti-parallel magnetization states, conceived by the computer as bits 0 and 1, became the working principle of today's nanoscale spin-valve devices. On the other hand, the interest in reaching ever-smaller devices boomed due to the practical realization of how to extract graphene and other two-dimensional (2D) materials from their bulk layered crystals in 2004 by mechanical cleavage.

The research presented in this thesis is the combined follow-up of the two above-mentioned Nobel prize winning discoveries. Here we cleave and separate atomically thin layers of 2D materials, bring them in close proximity of each other in the form of van der Waals heterostructures and fabricate graphene-based spintronic devices

with ferromagnetic electrodes. Graphene is chosen as an excellent spin transporter because of its small spin-orbit coupling and high charge carrier mobility. We inject and detect a non-equilibrium spin accumulation in graphene that diffuses in the channel from spin-polarized Co injector to detector electrodes and leads to the required different resistance levels based on the relative magnetization orientation of the Co electrodes.

For the design of our graphene-based spintronic devices we optimize the spin injection by introducing an insulating layer at the Co-graphene interface which acts as a tunnel barrier. Here we take advantage of atomically flat bilayer hBN that is an ideal pin-hole free tunnel barrier, providing a large spin-polarization for the Co electrodes by avoiding spin back-flow into the contact. In chapter 5, we also use bilayer hBN as an insertion layer between a semiconducting transition metal dichalcogenide (TMD) and metal contacts in TMD-based FETs. The presence of bilayer hBN at the TMD-metal interface eliminates chemical interactions, prevents Fermi-level pinning and reduces the Schottky barrier height. Thus, it leads to the observed efficient gate-tunability of contact resistances even at low biases. These observations introduce bilayer hBN as a suitable choice for high quality tunneling contacts that allows for low energy charge and spin injection in semiconducting layered materials.

In practical spin-logic circuitries one requires electric-field control and manipulation of the spin information. This is possible in materials with large spin-orbit or exchange coupling and both are absent in graphene. Thus, here we explore the possibility of tuning the spin-related properties of graphene by bringing it in the proximity of other 2D materials. We induce significant spin-orbit coupling in graphene by bringing it in contact with TMDs. As a consequence, the spin transport measurements of chapter 6 in graphene-TMD van der Waals heterostructures show strongly anisotropic spin dynamics with the lifetime of the out of plane spins to be one order of magnitude longer than that of the in-plane spins. This experimental observation confirms the theoretical predictions of the possibility to imprint the spin texture of the neighboring TMD layer in graphene, where the broken in-plane symmetry gives rise to the out-of-plane valley-Zeeman spin-orbit fields that rule the spin-dynamics. Beyond that, graphene loses its out-of-plane symmetry when brought in contact with other materials in van der Waals hybrids. The broken symmetry due to the out-of-plane electric field at the graphene-TMD interface leads to strong in-plane Rashba spin-orbit fields that affect the lifetime of the out-of-plane spins in the proximitized graphene.

Furthermore, we observe that both induced valley-Zeeman and Rashba spin-orbit fields allow for the emergence of charge-to-spin conversion mechanisms in graphene (chapter 7). The valley-Zeeman spin-orbit fields with opposite sign in the two valleys in graphene cause a spin-dependent modification of the electron trajectories. We measure this in a graphene Hall-bar in proximity of a monolayer TMD, where the applied charge current generates an out-of-plane polarized pure spin current in the transverse direction via the spin Hall effect. On the other hand, the Rashba spin-orbit fields result in a winding in-plane spin texture with spins perpendicular to the momentum. In this condition, an in-plane electric field can create a finite accumulation of the in-plane spins, perpendicular to the direction of the charge current via Rashba-Edelstein effect (REE). In chapter 7, we detect the co-existence of both effects that can be separated by the symmetry-resolved spin precession measurements under oblique magnetic fields. The REE signal, in particular, is shown to be gate-dependent and persistent up to room temperature.

In chapter 8, we go one step further by bringing graphene in the proximity of 2D magnetic materials which allows for inducing both exchange and spin-orbit coupling in graphene. The large spin-splitting in magnetized graphene (in proximity of CrSBr) results in an imbalance in carrier density for spin-up and spin-down electrons and thus a strong spin polarization of conductivity (up to 14%) is directly measured. The different conductivity for spin-up and down electrons also leads to the emergence of the spin-dependent Seebeck effect by which a thermal gradient in the magnetic graphene creates spin accumulation. Moreover, we detect non-linearity in the Hall voltage as a signature of anomalous Hall effect in graphene-CrSBr heterostructure which implies the co-presence of magnetism and spin-orbit coupling in this system.

The studied proof of principle devices that directly address spin lifetime anisotropy, spin-to-charge conversions, and spin-dependent conductivity in graphene evidence strong proximity-induced spin-orbit and exchange couplings in graphene while its superior charge and spin transport properties are preserved. The distinct spin transport properties that graphene acquires in the proximity of TMDs or 2D magnets and in particular, the observed efficient spin generation and manipulation by means of electric fields only, open the route towards 2D all-electrical spintronic and spin-caloritroic devices.