ENGINEERING ELECTRON SPIN IN VAN DER WAALS HETEROSTRUCTURES

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Two-dimensional materials revolutionize spintronics, enabling unprecedented manipulation of electron spin for next-generation information technologies. Particularly exciting is the possibility of spin engineering van der Waals heterostructures, expected to lead to breakthroughs in many realms of quantum physics. This feature article explores tailoring spin interactions in van der Waals heterostructures via stacking and twisting.

What is spintronics

Spintronics covers a vast research landscape of solid-state physics in which the central role is played by electron spin [1]. While spin is essential for the Fermi-Dirac statistics and thus the overall electronic structure of solids, spintronics focuses on actively manipulating spin by controlling spin interactions such as spin-orbit coupling (SOC) or exchange coupling. SOC preserves time-reversal symmetry and does not lead to magnetism, whereas exchange coupling breaks time-reversal symmetry and results in magnetic materials. However, both spin interactions influence spin dynamics and spin relaxation and can be used for spin manipulation. Such manipulation is particularly important for memory and magneto-logic applications, given that the electron spin is used to store information.

In magnetic solids, electron spins have a preferential orientation dictated by the magnetization. Spin manipulation occurs through the rotation of this magnetization. For example, in spin-transfer-torque magnetic random access memory, spin-polarized electrons from a reference ferromagnet are injected into a magnetic layer, aligning its magnetization parallel to that of the reference layer. A more recent version of this technology is based on spin-orbit torque (SOT), which requires only a single magnetic layer. In this case, electrons become spin-polarized through SOC rather than exchange interactions.

Nonmagnetic solids are generally spin-unpolarized. However, they can maintain nonequilibrium electron spin populations created by electrical or optical spin injection. In a typical spin transport experiment, spin-polarized electrons are injected from a ferromagnetic electrode. The nonequilibrium spin, also known as spin accumulation, precesses around an applied magnetic field and is detected by another magnetic electrode as a voltage signal, or more precisely, as electromotive force. This signal is then analyzed as a function of the magnetic field, providing relevant information such as spin relaxation time and spin diffusivity.

2D spintronics

The era of 2D spintronics started with the first successful spin injection and spin detection in graphene achieved by the group of Bart van Wees [2]. Early experiments found that the spin relaxation time in graphene is hundreds of picoseconds, which was significantly below theoretically predicted microseconds. Later experiments employed cleaner samples—graphene encapsulated between hexagonal BN—extending the measured spin lifetime up to 10 ns. Despite numerous proposed mechanisms for spin relaxation in graphene, a definitive experimental determination of the origin behind ultrafast spin-flip processes remains elusive. Quantitatively closest agreement with the experiment provides the mechanism based on resonant magnetic scatterers [3].

Graphene exhibits exceptional electron transport properties, characterized by high group velocities of about 10^6 m/s. This translates to remarkably long spin diffusion lengths in the micrometer range, making it a promising candidate for spintronic applications [4] [5]. Combining exchange and SOC, see Box 2, has great potential for breakthroughs in both fundamental science and applications. Adding more complexity by stacking two-dimensional (2D) spin-orbit materials, magnets, ferroelectrics, and superconductors should further enhance our ability to influence electron spin, potentially leading to fascinating new phenomena. Finally, merging spintronics and correlated physics will open new venues for designing quantum materials with controllable properties.

However, a significant limitation arises from the intrinsically weak SOC in graphene, in the range of 20-30 micro eV. This falls short of the 1-100 meV regime necessary for efficient electrical manipulation of spin and for realizing devices based on SOC-driven topological edge states.

Fortunately, the inherent versatility of 2D materials offers a compelling solution. By employing a bottom-up approach, individual 2D monolayers can be stacked vertically to form van der Waals heterostructures. These engineered structures are bound together by weak interlayer van der Waals forces, enabling the deliberate design of spin interactions by the proximity effects [6].

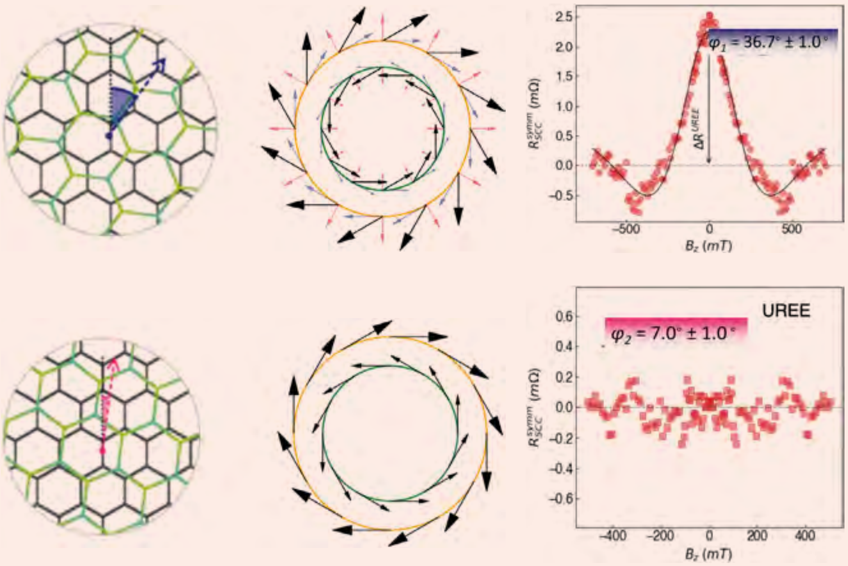
Proximity effects

The math of "1 + 1 = 2" would not capture the essence of van der Waals engineering. Indeed, stacking individual 2D monolayers can lead to the emergence of entirely new materials. The properties of these materials transcend the simple sum of their individual constituents. However, if two materials interact weakly, their electrons are not strongly perturbed and we can still describe the monolayers individually. This has profound effects on spintronics. The prototypical example to design spin interactions is graphene, which has weak SOC and is nonmagnetic. For example, placing graphene on a material with strong SOC, such as semiconducting WSe_2, can enhance SOC in graphene by two orders of magnitude. Similarly, using a semiconducting magnetic material like $Cr_2Ge_2Te_6$ as a substrate can induce weak magnetism in graphene. This would manifest as the graphene electrons experiencing an exchange field on the meV scale, effectively splitting their spin-up and spin-down energy levels. Due to the weak interlayer coupling, such proximity effects are short-ranged: they primarily affect the neighboring layers.

Why are 2D semiconductors preferred for proximity effects, in contrast to metals and insulators? Semiconductors appear to provide the optimal balance between interaction and isolation, allowing proximity effects to influence graphene's properties while still maintaining its essential characteristics and enabling independent control over its behavior.

Furthermore, 2D semiconductors exhibit unique spintronic properties. Electrons and holes in semiconducting transition metal dichalcogenides, such as WSe_2, have their spin "locked" with the momentum. As a result, employing circularly polarized light, researchers can achieve optical orientation, generating spin-polarized electrons and holes. In a heterostructure these spin-polarized carriers can tunnel to neighboring layers, offering exciting possibilities for opto-spintronics, such as the development of efficient spin LEDs where the light emission could be controlled by spin.

BOX 1: SPIN-ORBIT FIELDS ENGINEERING IN 2D MATERIALS



There are two basic types of spin-orbit fields in 2D materials: in-plane Rashba field and out-of-plane valley Zeeman (Ising) spin-orbit field. The Rashba field is usually tangential to the electron's velocity. However, changing the crystal symmetry by twisting, it was recently discovered that Rashba fields can also have a radial component, leading to a tilting of the Rashba field away from the tangential direction. The Rashba field can also be purely radial, yielding what is also called a chiral spin structure, since the scalar product of spin and momentum, which appears in the radial Rashba Hamiltonian, is not preserved by reflection through a vertical mirror plane along the velocity vector: velocity is polar, spin is axial.

Remarkably, tilting of the Rashba field was recently seen in experiment. Felix Casanova's group studied twisted heterostructures of graphene and WSe2, observing conventional tangential Rashba for twist angles of 7°, and unconventional tilted Rashba for a twist of 36,7°. The presence of an unconventional Rashba field (plotted next to the atomic structure) was detected by the presence of spin-charge conversion resistance (Rscc) peak as a function of the applied magnetic field. Figure courtesy of F. Casanova [11].

Spin-orbit fields

Materials lacking a center of inversion symmetry are special. Their SOC manifests as spin-split electron energy levels, opposite for opposite electron momenta (to maintain overall spin polarization). Furthermore, each electron feels an effective magnetic field, called a spin-orbit field, which provides an efficient control for the spin manipulation. Unlike a conventional magnetic field that acts in real space, the spin-orbit field influences the electron's momentum.

There are several types of spin-orbit fields found in 2D materials. One of the most common is the Rashba field, an in-plane vector field typically oriented tangentially to the electron's velocity. Another significant type is the valley-Zeeman, or Ising, spin-orbit field, which points out of the 2D plane. The valley-Zeeman field plays a crucial role in various phenomena: it causes spin-valley locking in WSe_2 and related semiconductors (hexagonal transition metal dichalcogenides), contributes to Ising superconductivity in $NbSe_2$, and results in the giant spin relaxation anisotropy observed in graphene/WSe_2 heterostructures.

This last example is particularly fascinating. Pristine graphene, which possesses a center of inversion, inherently lacks a spin-orbit field. However, when graphene is in contact to WSe_2, the proximity effect induces both Rashba and valley-Zeeman spin-orbit field in graphene. This fundamentally alters graphene's spin properties, demonstrating the profound impact of SOC in 2D materials. A recent example is the nearly 10-fold increase in the critical temperature of superconducting (untwisted) bilayer graphene due to the proximity-induced valley-Zeeman spin-orbit coupling [7].

Twist and spin

Accessing the spin-orbit fields is a key goal in 2D spintronics, as it offers the potential to control spin dynamics and enable spin-controlled functionalities such as spin-charge conversion and spin-orbit torque. While traditional strategies to tailor spin-orbit fields include straining and gating, a novel approach involving twisting has recently emerged as a promising method.

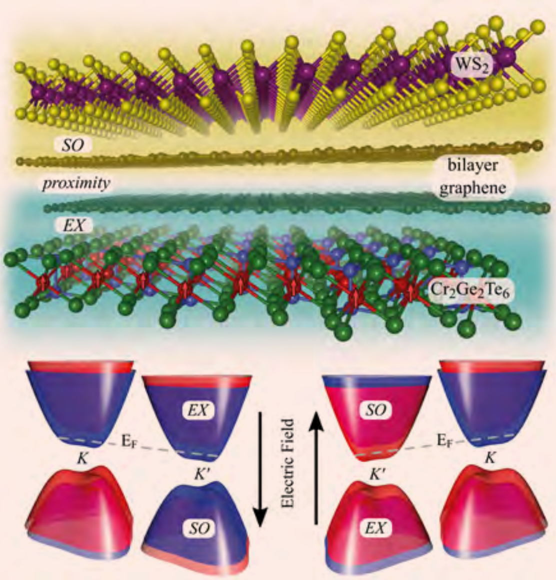
At first glance, twisting appears a terrible choice. Twisting two monolayers, even if they have the same lattice geometry (hexagonal, rectangular), creates a seemingly messy atomic structure. Commensurate unit cells—supercells—if they exist, comprise tens to thousands of atoms, each seeing a different local environment of the neighboring layer. Can one hope for a simple description of the spin properties of the proximitized monolayers? Theoretically, the challenge is to describe large supercells, requiring significant computational resources. Experimentally, one is typically restricted to materials stable in air. Prominent examples are semimetallic graphene, semiconducting transition metal dichalcogenides, or magnetic CrSBr.

One of the most extensively studied heterostructures is that of graphene and WSe_2, which fortunately also happens to be highly intriguing for two reasons. First, upon twisting the out-of-plane valley-Zeeman spin-orbit coupling is expected to diminish monotonically to zero as the twist angle approaches 30°. This is important since all the effects claimed to originate from this spin interaction should vanish at this twist angle. Second, the Rashba spin-orbit field, which is usually perpendicular to the electron's velocity, can become tilted, acquiring a radial (parallel to the velocity) component. As a result,

BOX 2: EX-SO-TIC HETEROSTRUCTURES

The interplay of exchange and spin-orbit coupling is a highly active research area in condensed matter physics. This interest is driven by discoveries of topological magnetic materials, predictions for exotic quasiparticles like Majorana fermions, or potential applications in spin-orbit torque devices.

Van der Waals heterostructures emerge as a perfect platform for this research. Ex-so-tic heterostructures, designed to comprise exchange (ex) and spin-orbit (so) interactions, offer a unique ability to dynamically control these two spin interactions, even swapping them, by electrical signals. The figure shows an ex-so-tic structure based on bilayer graphene (BLG) sandwiched between a magnetic monolayer $Cr_2Ge_2Te_6$ and a strong spin-orbit monolayer transition metal dichalcogenide. Applying an electric field perpendicular to the 2D sheets, the conduction electrons at the Fermi level (dashed line) in bilayer graphene can experience either spin-orbit (seen in the band structure on the right: top bands at momenta K and K′ = - K have opposite spin polarization indicated by red and blue) or exchange coupling (left band structure indicating the same spin polarization at K and K′), or their combination. Figure from Ref. [12]



unconventional spin-charge conversion can be observed, with the accumulated spin also pointing along the direction of velocity, see Box. 1. Ramifications of the Rashba spin-orbit coupling are yet to be explored.

Outlook

Combining exchange and spin-orbit coupling, see Box 2, holds great potential for breakthroughs in both fundamental science and applications. This includes advancements in spin-orbit torques [8], spin-logic circuits [9], or 2D versions of MESO memories [10]. Furthermore, adding more complexity by stacking two-dimensional spin-orbit materials, magnets, ferroelectrics, and superconductors should further enhance our ability to influence electron spin. Finally, establishing a synergy between spintronics and correlated physics would open new avenues for designing quantum materials with controllable properties. ■

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