van der Waals heterostructures

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Asymmetry in the magnetic neighbourhood

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Transforming atomically thin materials by their magnetic neighbours reveals a surprising asymmetry that allows a versatile control of the valley degrees of freedom and band topology in van der Waals heterostructures.

Materials design often resembles the saying 'the grass is greener on the other side'. Even silicon, the key to electronics, is only of limited use in its pristine form. Silicon's missing desirable properties are typically acquired by adding impurities, a process of doping ubiquitous to many other classes of materials. Now, writing in *Nature Materials*, Choi and colleagues[1] report a clever, very different path to design materials (Fig. 1a), where desirable magnetic properties in an atomically thin semiconductor, $MoSe_2$, leak from its ferromagnetic neighbour, $CrBr_3$.

In a broader context, Choi and colleagues demonstrate an example of proximitized materials[2]. A given material can be transformed through proximity effects, whereby it acquires properties of its neighbours, for example, becoming magnetic, superconducting, topologically non-trivial, or with an enhanced spin–orbit coupling. Such proximity effects not only complement the conventional methods of designing materials by doping or functionalization, but can also overcome their various limitations. For example, direct doping of the common semiconductor GaAs by magnetic atoms (Mn) reveals the cost of adding magnetism: the high mobility of GaAs is lowered by several orders of magnitude, and its excellent optical emission properties are drastically degraded[2]. By contrast, through proximity effects in van der Waals heterostructures, desirable properties can be introduced more gently with a better-preserved identity of the host material.

In magnetic proximity interactions (MPIs), the spin-dependent wavefunction from a ferromagnet penetrates into a non-magnetic material and directly polarizes its electronic structure, which then acquires some of the magnetic properties of its neighbour. While this phenomenon has been studied for decades[2–4], the common understanding of MPIs was derived in analogy with spin effects in a uniform applied magnetic field, **B**. The electron (hole) levels in the conduction (valence) band are shifted based on the direction of their spin relative to **B**. As a result, with the broken time-reversal symmetry, an initially non-magnetic material acquires a net energy difference between electrons of different spins (say, 'up' versus 'down'). Surprisingly, Choi and colleagues show that there is more to MPIs than meets the eye. A hallmark of $MoSe_2$ is the presence of local extrema, known as valleys, marked by K and K′ in Fig. 1b. Unlike the expected equal **B**-induced shift

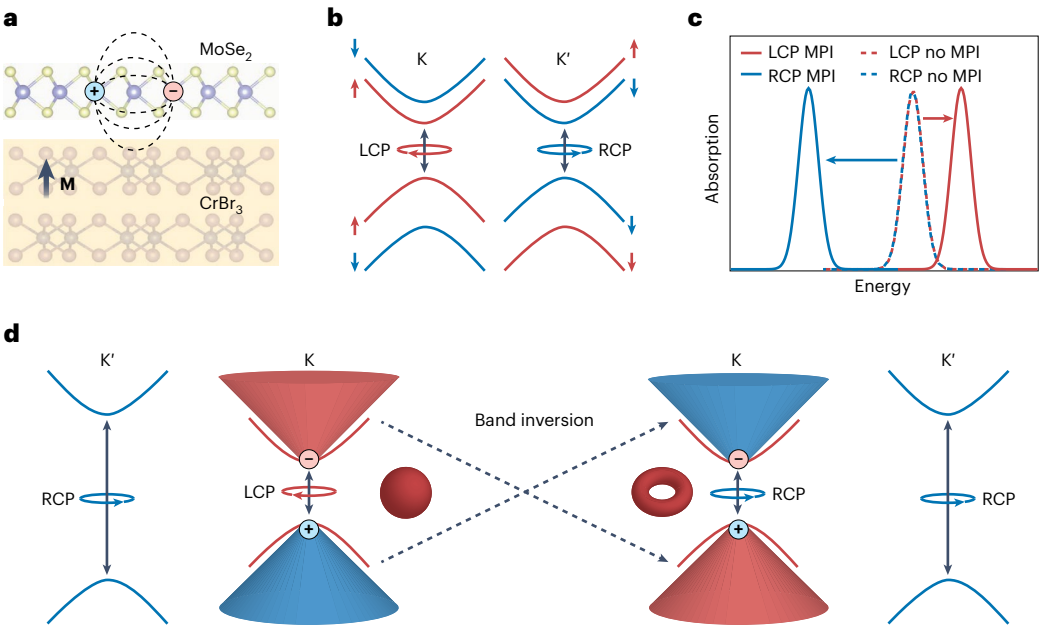

Fig. 1 | Optically probing asymmetric magnetic proximity interactions and band topology. a, Schematic of the $MoSe_2/CrBr_3$ heterostructure. An exciton is a bound state of the positively (negatively) charged hole (electron) with marked electric field lines. $CrBr_3$ has an out-of-plane magnetization, **M**. **b**, Schematic of the low-energy conduction (upper) and valence (lower) bands, which are spin resolved, as denoted by arrows. The resulting low-energy exciton at the K (K′) valley couples to a LCP (RCP) light. **c**, Valley-dependent exciton absorption spectrum shift induced by the MPIs. The solid (dashed) lines denote the magnetic (non-magnetic) $CrBr_3$. **d**, Schematic of the exciton helicity reversal when the trivial topology (ball) at the K valley is transformed into a non-trivial topology (doughnut) through the band inversion. Panel **a** adapted with permission from ref. 1, Springer Nature Ltd.

of energy levels at K and K′, there is a striking asymmetry: one valley is transformed more by the ferromagnetic $CrBr_3$ neighbour than the other. Monolayer $MoSe_2$ has different on-site energies for Mo and Se atoms in its honeycomb lattice and broken inversion symmetry, analogous to the chessboard with two types of square, but the K and K′ behaviour is equivalent, constrained by the time-reversal symmetry. However, the MPI breaks the time-reversal symmetry such that the previous real-space chessboard gets its partner in the momentum space where K and K′ are now inequivalent[4], just as different colours of its squares. The resulting valley-specific rules provide a versatile two-state system, similar to spin.

To elucidate the asymmetry of the MPI, Choi and colleagues used an enlightened approach. With small screening, two-dimensional $MoSe_2$ has tightly bound excitons (a bound state of electron–hole pairs), which dominate its optical properties. Given the out-of-plane magnetization, **M**, in $CrBr_3$, optical transitions between conduction and valence bands satisfy dipole selection rules with the conservation of the total angular momentum between the carrier (electron, hole) spin and light. Without MPIs, each valley supports a specific (left, right) circular polarization of light (LCP, RCP) and the accompanying angular moment resulting from the corresponding exciton (Fig. 1b). However, since the excitons at K and K′ have the same energy, without MPIs there is no shift in their two spectral lines (Fig. 1c), as experimentally verified at high temperature when $CrBr_3$ loses its magnetic order. By contrast, for a lower temperature, with a ferromagnetic $CrBr_3$ and MPIs, the excitonic energies become valley dependent. As depicted in Fig. 1c, a striking asymmetric shift of the spectral lines of the reflected LCP and RCP light is the fingerprint of the MPI asymmetry.

A tantalizing prospect of this discovery is that the MPI asymmetry could be ubiquitous to many other heterostructures. If, instead of $MoSe_2$ with its wide energy bandgap of more than 1 eV, we chose a narrow-bandgap hexagonal layer, the MPI asymmetry could create even more dramatic results (Fig. 1d). With inequivalent K and K′ valleys, the bandgap could be closed and reopened at only one of them (shown at K). While the MPI asymmetry alone is moderate and expected to yield energy shifts <10 meV (refs. 1, 2), together with a real-space asymmetry of the lattice sites[5], enhanced by an applied electric field[6], they can conspire to provide a much stronger total asymmetry. In the chessboard analogy, the colour contrast of the squares was only modest in momentum and real-space. However, by combining the two chessboards, the colour contrast becomes stark and tunable to even overturn the band order in a given valley. This is an example of a topological transition[7] where the band order between the conduction and valence bands is reversed, akin to tying a knot, which is accompanied by the change of the selection rules[8]. The resulting reversal of the circularly polarized light, from LCP to RCP[6], could be detected by the methods of Choi and colleagues.

At present, the reported results are limited to low temperature and modest MPI asymmetry[1]. However, with the development of two-dimensional ferromagnets[9], there is no fundamental obstacle to substantially enhance the magnitude of the energy shifts and the temperature range over which they are observed. One can envision scaled-down applications for tunable interconversion between the spin of electrons and polarization of light, which could even lead to a novel class of spin lasers[10].

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Competing interests

The authors declare no competing interests.