Gate-Controlling Emergent Physics in Layered Quantum Materials

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Probing and controlling electronic states in solids is a key goal in contemporary condensed matter physics. Layered quantum materials provide a versatile platform for tailoring quantum states, thereby enabling the emergence of many novel physical phenomena. We specialize in fabricating field-effect nanodevices made of layered quantum materials with sample dimensions comparable to, or smaller than the characteristic length scales of the states under study. This allows us to probe the emergent physics in the mesoscopic and quantum regimes and to control them utilizing the ionic gating technique. Here we describe our latest research progress on gate-controlling emergent physics in layered quantum materials. Recent highlights include gate-controlling interface Rashba spin-orbit coupling, gate-tuning anomalous Hall effect in a layered polar semiconductor, the observation of the concurrence of directional Kondo behavior and incommensurate magnetic ordering, and the discovery of two-dimensional superconductivity in a thick exfoliated kagome film.

Layered quantum materials are solids with highly anisotropic electronic properties, consisting of strongly bonded sheets that are weakly bonded to adjacent layers. These materials exhibit novel electronic or magnetic properties arising from their quantum mechanical nature, reduced dimensionality and layer-by-layer stacking. Therefore, layered quantum materials have emerged as a versatile playground for exploring fundamental physical principles and for the development of next-generation electronic devices. The key to fully realizing their potential is to gain deterministic manipulation of their correlated electronic states in a controllable manner. Our research sits at this frontier, with the aim to achieve new levels of control and thus gain new insights into the interacting states in layered quantum materials.

Many of the layered quantum materials are known to be sensitive to changes in carrier density, which allows for a powerful route for controlling their electronic states. Here, we mainly employ field-effect devices made of layered quantum materials to probe and control their physical properties. The solid-gated field effect transistor (Fig. 1a) has been widely applied in narrow-band-gap semiconductors for nearly 80 years. The recently developed innovative approach – the ionic-gated transistor (Fig. 1b) – generates a high electric field at the interface between the channel material and the ionic liquid, extending the research to wide-band-gap semiconductors and semimetals.

We have developed a good knowledge in the field of gate-controlling emergent physics [1, 2], and have achieved advancements in various layered quantum materials [3, 4, 5, 6, 7, 8, 9]. Our interest spans across various kinds of layered quantum materials, ranging from semiconductors like elemental material tellurium (Te) to semimetals like transition metal compounds. Single crystals of the layered materials were provided by the internal collaborators from the Chemical Metals Science department of our institute or external collaborators. We utilize state-of-the-art nanofabrication techniques, including e-beam lithography and sputtering, which are set up in-house, to pattern the layered materials into nanodevices (Fig. 1c), and explore their quantum transport behavior at low temperatures, in high magnetic fields, and, more interestingly, at different doping levels controlled in-situ by the gating technique.

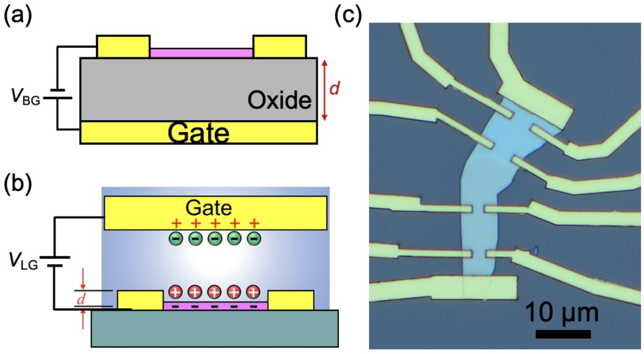

Fig. 1: Schematic representation of (a) a solid gated field effect device and (b) an ionic gated device. (c) An optical image of an example device made of layered quantum materials, fabricated through nanofabrication using ebeam lithography.

Controlling Rashba spin-orbit coupling in layered tellurium

We first worked with Te, a single elemental semiconductor, to control its interface Rashba spin-orbit coupling (SOC). Te has been attracting significant scientific interest recently due to its chiral structure (Fig. 2a, b) and unusual topological nature. Efficient tuning of SOC in this material is, therefore, highly desirable, as it may lead to nontrivial topological quantum properties [10].

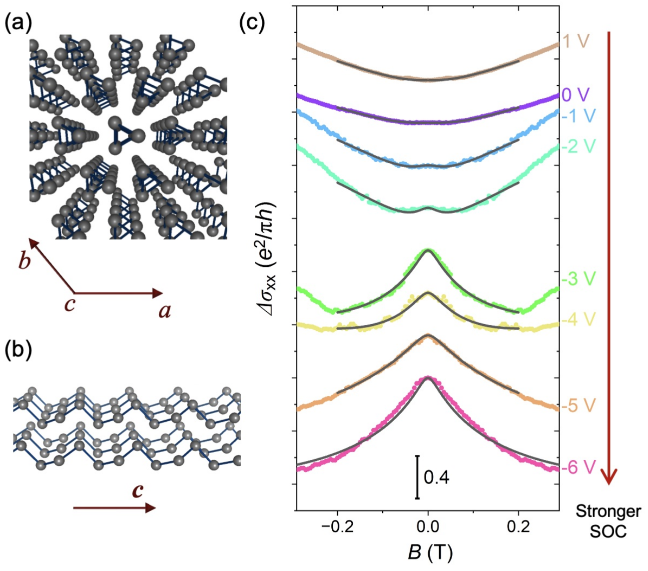

Fig. 2: (a) Crystal structure of trigonal Te. (b) Schematic side view of the exfoliated Te films. (c) Magnetoconductivity of an exfoliated film modulated by ionic gating. The data were taken at a temperature of 2 K with the magnetic field applied along the out-of-plane direction. The solid lines are the fitting data. (Figure taken from [5].)

We have made progress in this area by investigating electronic transport in ionic-gated Te devices [5]. To be specific, we have carried out a systematic study on the electric field control at the interfaces of ionic liquid/Te thin flakes. The large interfacial electric field enables drastic tuning of the in-plane conductivity, allowing for a transition from insulating states to metallic states by simply changing the polarity of the applied gate voltage. As shown in Fig. 2 c, measuring the dependence of the magnetoconductance, $\Delta\sigma_{xx}(B) = \sigma_{xx}(B) - \sigma_{xx}(0)$, on the gate voltage reveals a crossover from weak localization to weak antilocalization. This demonstrates that a stronger SOC is induced by gate voltage (or electric field) control of the system. Quantitative analysis of the magnetoconductance at different gate voltages and temperatures allows us to determine the spin- and phase-relaxation mechanisms in our sample, which demonstrates that the Rashba parameter is enhanced by a factor of 4 through ionic gating. Our results not only clarify how the SOC can be tuned in Te, but also demonstrate the wide range of SOC tuning, highlighting its potential impact on spintronics devices.

Controlling magnetism in a layered polar semiconductor

The work described above focused on inducing spin-polarized electrons in non-magnetic materials. In parallel with this, we also exploited the possibility of controlling spin-polarization in layered magnetic materials, as it enables fundamental studies of magnetic properties and practical applications based on the magnetic layered materials.

Recently, we have presented a series of studies of the layered magnet $AgCrSe_2$, where $CrSe_2$ transition metal dichalcogenide layers alternate with Ag layers and form a polar structure [3, 6, 7, 8] (see also reports: **PQM_01_Baenitz** and **PQM_09_Rosner**). By conducting electrical transport measurements on $AgCrSe_2$ crystals, we have observed a spontaneous anomalous Hall effect (AHE) [7]. Specifically, we find that its Hall resistivity exhibits anomalous jumps as a function of the applied magnetic field at low temperatures. Quantitative analysis of the AHE reveals that the anomalous Hall conductivity does not depend on the scattering rate, suggesting that it is an intrinsic AHE but distinct in origin from one based on conventional ferromagnetism.

This observation has motivated a set of research efforts in exploring the origin of this observed effect. Firstly, we performed ionic gating experiments and we found that the anomalous Hall conductivity magnitude can be enhanced by tuning the carrier density (Fig. 3).

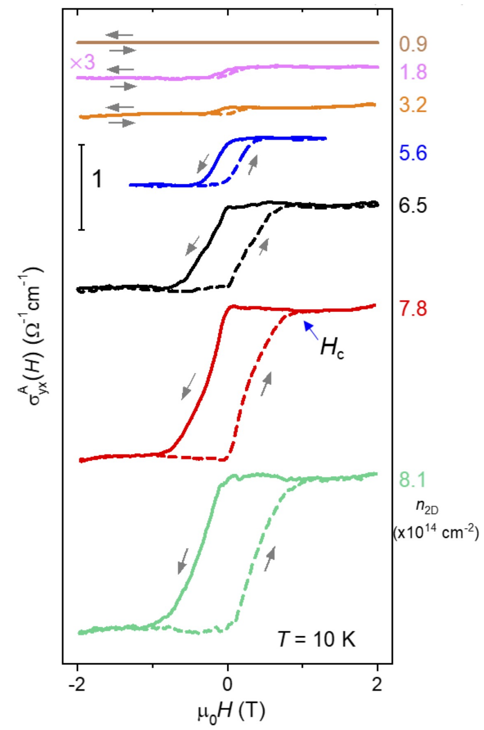

Fig. 3: The anomalous Hall conductivity $\sigma^A_{yx}(H)$ of $AgCrSe_2$ modulated by ionic gating. The data were obtained at T = 10 K with the magnetic field applied along the c-axis. The carrier density n_{2D} extracted based on the single band model from the field linear part of the Hall effect at each V_G are labeled beside the curves. (Figure taken from [7].)

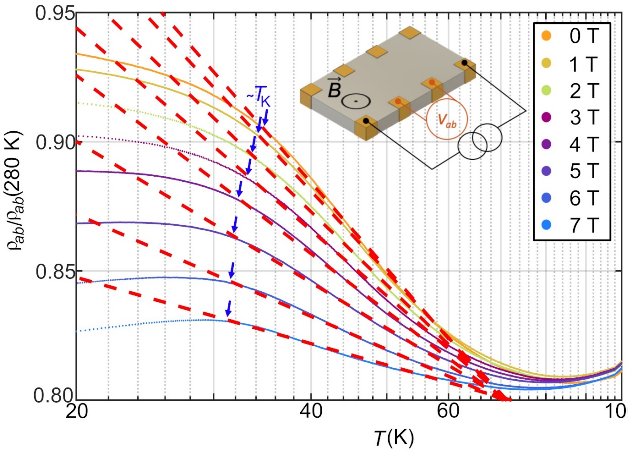

Fig. 4: Variation of the Kondo temperature estimated from the resistivity measurements as a function of the out-of-plane magnetic field. (Figure taken from [8].)

Secondly, theoretical calculations, which were achieved in collaboration with Libor Šmejkal (Johannes Gutenberg Universität Mainz, now a joint group leader between MPI-CPfS and MPI-PKS), demonstrate that the observed gate-tunable AHE can be reasonably accounted for by Berry curvature that correlates closely to the previously suggested magnetic ordering in $AgCrSe_2$.

Our findings open up new avenues for exploring the physical properties that emerge from the interplay between magnetic and polar structures. We believe these results will prove to have broad significance, because they reveal a single material platform that allows for the coexistence of polar and time-reversal symmetry broken effects, which has long been sought after in the fields of semiconducting spintronics and magnetoelectrics. The gate-controllable AHE in this material provides a new possibility for the local manipulation of spin states, which can facilitate the realization of stable and compact spintronic and magnetoelectric devices.

There are other puzzles in the transport properties of $AgCrSe_2$. We have observed an upturn of the in-plane resistivity at low temperatures, indicating Kondo-like transport whose characteristic temperature coincides with the magnetic ordering temperature. Interestingly, this Kondo-like transport feature occurs only in slightly off-stoichiometric samples and disappears in the gated devices. At a first glance, the concurrent emergence of Kondo-like transport and magnetic ordering seems contradictory, as magnetic order typically competes with Kondo impurity scattering and eventually suppresses the latter in the Doniach phase diagram. Through a combined study involving transport measurements (Fig. 4), susceptibility measurements and high resolution wavelength X-ray diffraction spectroscopy, we are able to find a consistent physical picture within which magnetic order is indeed required for the appearance of the Kondo behaviour [8]. The ability to enable Kondo effect, coupled with its association with the antiferromagnetic order, provides a novel route to tune the competition between magnetic correlations and Kondo screening.

Controlling competing orders in a layered kagome system

We have started a new project aimed at probing and controlling competing orders in a layered kagome superconductor. The recently discovered kagome system CsV_3Sb_5 has proven to host intertwined superconducting (SC) states and charge density wave states, which is putatively related to the nested Van Hove singularities (VHSs) near the Fermi level. The intertwined states present both an exceptional opportunity and a challenge: the opportunity lies in the fact that the intertwined orders lead to exotic physical phenomena, while the challenge is that it complicates the interpretation of the emergent physics and the disentanglement of the various orders. In the following, we first describe some recently achieved results on the direct probe of the SC pairing symmetry in CsV_3Sb_5 thin films without applying external perturbations [9], and then discuss future experimental endeavours.

We focus on the exfoliated thin flakes of CsV_3Sb_5 with the optimal thickness ranging from 40 to 60 nm with maximal T_c. By measuring the temperature dependent superfluid stiffness, we have seen a clear Berezinskii-Kosterlitz-Thouless (BKT) transition (Fig. 5), which is a hallmark of 2D superconductivity. Yet, the observation of 2D superconductivity in samples with thickness

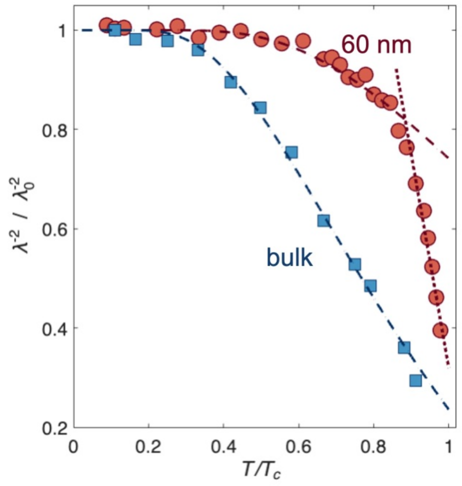

Fig. 5: The temperature dependent superfluid stiffness. The dashed curves are fits based on the hypothesis of an s-wave order parameter. The experimental data of the 60-nm-thick film exhibits a BKT transition near T_c as indicated by the dotted line. (Figure taken from [9].)

around 40-60 nm is striking, as the thickness is approximately 70 times larger than the interlayer lattice spacing. Our finding sheds new light on the competing correlations in the kagome superconductors, and provides fresh insights on realizing 2D superconductivity in films far thicker than the atomic limit.

This achievement motivates us to carry out further experimental and theoretical investigations on the CsV_3Sb_5 thin films. One major task is to investigate the origin of the 2D superconductivity, particularly regarding the underlying correlations at play within the optimal thickness. Another focus is to investigate the crossover from 3D to 2D behavior with thickness reduction. Last but not least, tuning the doping level towards the VHSs nested near the Fermi level, to further enhance electronic correlations, remains a natural and long-sought goal.

External Cooperation Partners

P.D.C. King (University of St Andrews, UK), S.D. Wilson (University of California Santa Barbara, USA), H. Zhou and J. Liu (University of Tennessee, USA), C. Berthod and A.F. Morpurgo (University of Geneva, Switzerland), J.J. Lin (National Yang Ming Chiao Tung University, Taiwan), C. Hooley (MPI-PKS, Germany), L. Šmejkal (Johannes Gutenberg Universität Mainz, Germany).

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