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INVITED REVIEW

Flattening is flattering: The revolutionizing 2D electronic systems*

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Two-dimensional (2D) crystals are known to have no bulk but only surfaces and edges, thus leading to unprecedented properties thanks to the quantum confinements. For half a century, the compression of z-dimension has been attempted through ultra-thin films by such as molecular beam epitaxy. However, the revisiting of thin films becomes popular again, in another fashion of the isolation of freestanding 2D layers out of van der Waals (vdW) bulk compounds. To date, nearly two decades after the nativity of the great graphene venture, researchers are still fascinated about flattening, into the atomic limit, all kinds of crystals, whether or not they are vdW. In this introductive review, we will summarize some recent experimental progresses on 2D electronic systems, and briefly discuss their revolutionizing capabilities for the implementation of future nanostructures and nanoelectronics.

Keywords: 2D electronics, 2D superconductivity, Coulomb drag, twistronics

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1. Introduction

Flattening crystals into the atomic limit is a way to study model systems of such as superconducting quantum phase transitions in two dimensions (2D),[1,2] 2D quantum well and related physics,[3–6] the modification of critical behaviors in functional materials in 2D limit,[7–11] and the modern nanoelectronics based on multilayered thin films.[12–16] It is known that, to obtain those thin 2D films, techniques such as molecular beam epitaxy have been fostered in the community of surface sciences for decades,[17] with the grown atomically thin films often bound to the epitaxial substrates.

In early 2000s, researchers discovered that graphene, a free-standing single layer of graphite, can be obtained through a couple of rather simple methods including Scotch tape exfoliation[18] and chemical vapor deposition,[19] thus liberating the degrees of freedom of lattice-matching as well as the harsh growth conditions required in conventional epitaxial methods. After the big bang of mechanical exfoliation of van der Waals (vdW) crystals, single layer crystals with all kinds of compositions are routinely isolated from their bulk forms, thanks to the bountiful library of more than 5000 layered compounds on earth.[20]

When looking back to the above history, a question comes up naturally: what is the definition of a real 2D? To some extent, 2D physics thrives based on thin films (including vdW atomic layers), yet 2D systems are often disputable because of a lack of pure two dimensions that can be strictly achieved experimentally. It seems that a thin film will always have a certain thickness, even at the atomic level. Indeed, theories used to predict the non-existence of any ideal 2D crystals at finite temperatures.[21–23] However, in reality, nano-corrugations may take places that overcomes the thermodynamic limitations in theory.[24] Even though they are not ideally flat, laws of 2D physics prevail, as phenomena such as quantum Hall effect and Berezinski–Kosterlitz–Thouless phase transition can be observed in many x–y systems with a finite z-value.

A simplified view to describe the dimension crossover from 3D to 2D system is a slab model as given in Fig. 1. Carriers are free to move in the xy plane of the slab, but confined to a potential well along the z direction. The quasi-two-dimensional carriers in a well of size d will have discrete energy bands, for example, as shown in Fig. 1(b), at $k_x = k_y = 0$ the eigenvalues are given by $E_n = \frac{\hbar^2\pi^2 n^2}{2md^2}$, with n as the elevated quantum number and m as the effective mass. Using the conventional quadratic dispersion relation for the free in-plane motion of carriers, the total density of states (DOS) is simply the sum over all the constant values of DOS from each sub-band, namely, $g(E) = \sum_{n=1}^{n} \frac{m}{\pi\hbar^2}$, as shown in Fig. 1(c). It does not take a genius to see that, when the carrier is thermally excited to a high energy level n, $g(E) = \frac{m}{\pi\hbar^2} n = \frac{m}{\pi\hbar^2}\frac{d}{\pi}\sqrt{\frac{2mE}{\hbar^2}}$, carrier DOS takes a parabolic function, showing a behavior crossover from 2D to 3D. Therefore, to achieve two-dimensional behavior, it is reasonable to

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keep the energy separation ΔE_n $(= E_n - E_{n-1})$ between the neighboring discrete levels well above the thermal energy (i.e., $\Delta E_n > k_B T$), by controlling the well width d, temperature, or effective mass. The criterion is $d < \sqrt{\frac{\hbar^2\pi^2(2n-1)}{2mk_B T}}$, $n = 1, 2, \ldots$, for example, for carriers with effective mass m_e at room temperature behaving like 2D, the thickness d for the confinement has to be no more than 3.8 nm. We have to emphasize that the simple criteria formula for determining 2D has to be used by caution in terms of the followings: the formulas above are from solving the Schrödinger equation for non-relativistic particles with non-zero mass, and basically not suitable for the relativistic particles such as the Dirac fermions in graphene. For the latter case, which should be treated by solving relativistic Dirac equation in certain conditions, the criterion takes a form like $d < \sqrt{\frac{\hbar \pi c}{4k_B T}}$.[25] But we have to be cautious again since multilayered graphene may lose its characters as Dirac fermions due to interlayer interaction.

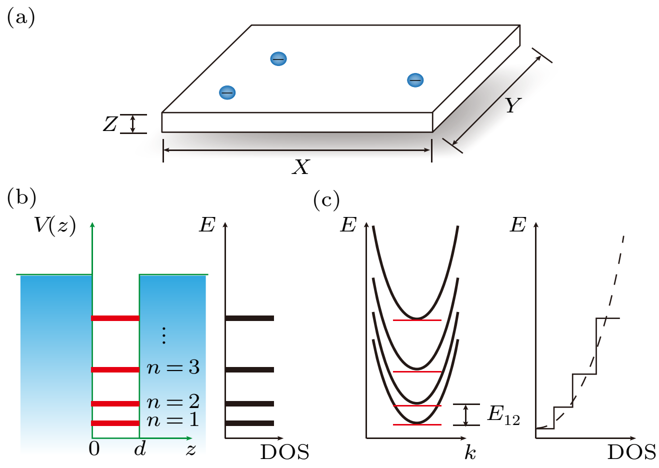

Fig. 1. (a) Schematic 2D structure with z direction confinement. (b) The z-direction confinement spectrum of one-dimensional quantum well model. (c) Band structure and density of states for quasi-two-dimensional electron gas.

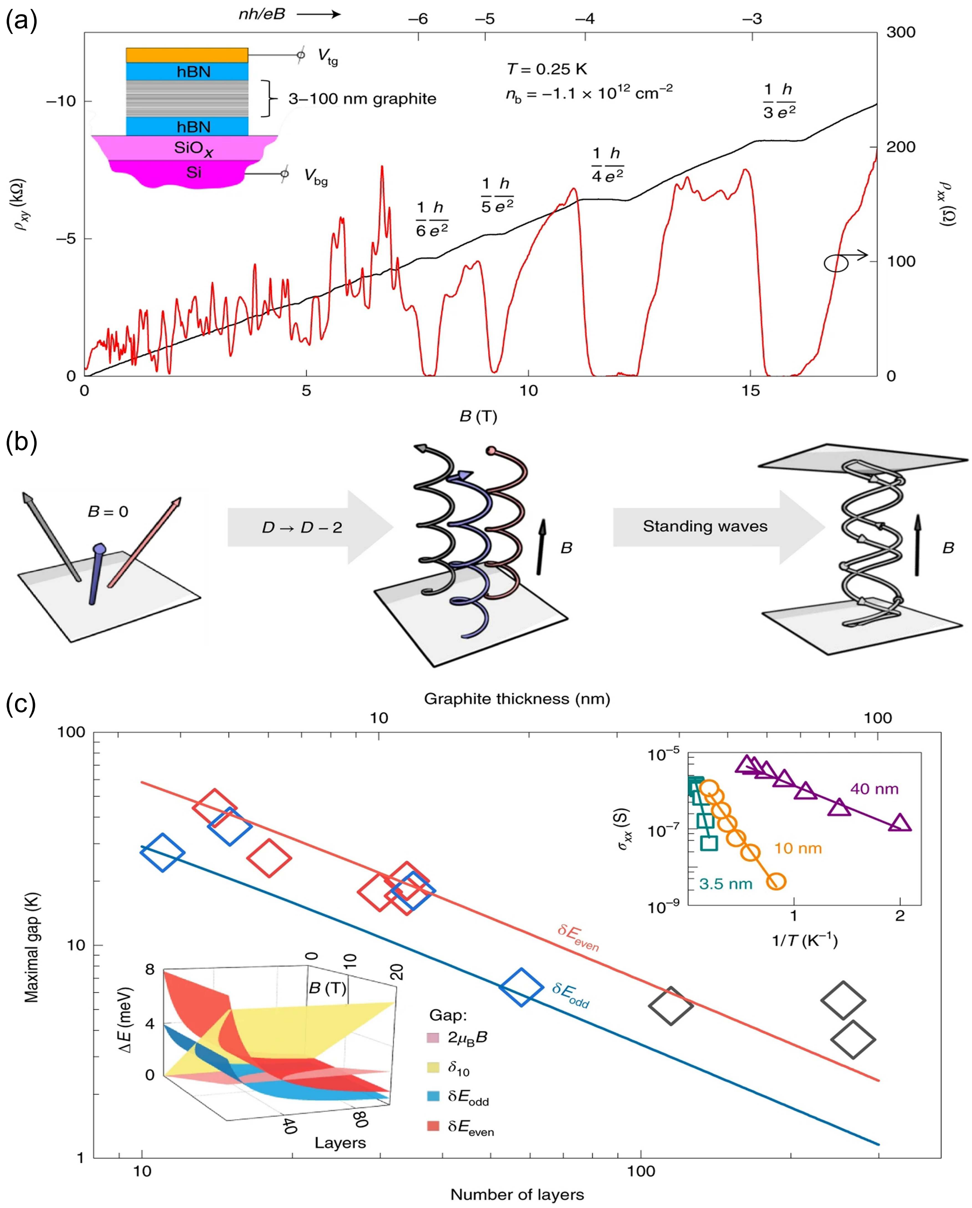

Fig. 2. (a) The QHE measured in a 6-nm-thick graphite flake at 0.25 K. (b) Schematic illustrations of electron trajectories under different conditions, (c) Energy gaps for the so-called 2.5D QHE as a function of thickness.[26] Reproduced with permission from Ref. [26].

Recent experiment showed that, in as thick as 100 nm graphite samples (usually considered as 3D bulk), clear signatures of quantum Hall (a scenario that is supposed to happen only in 2D systems) can still be observed,[26] as shown in Fig. 2(a). This striking observation can be attributed to the fact that, at the ultra-quantum regime (when only the lowest few Landau bands cross the Fermi level), quasi-one-dimensional electron motions are allowed only in the direction parallel to magnetic field B (Fig. 2(b)). As a result, the z-dimension is effectively reduced (defined as a 2.5D system, in between 2D and 3D) by introducing the standing wave within the top and bottom surfaces of the sample, where the parity of number of layers, Zeeman splitting, as well as the Landau cyclotron gap interplay and determine the observed 2.5D QHE behavior (Fig. 2(c)). It is noteworthy that although charge carriers induced by gate voltages are mainly distributed near the surface because of electrostatic screening, they mix with the bulk states since the 0 and 1 Landau bands of graphite cross the Fermi level, making the two graphite surfaces correlated (For sufficiently high doping, higher Landau bands can also become occupied at the surface, which results in charge localization near the surfaces, giving rise to surface states decay exponentially into the bulk).

The QHE behavior is also reported in other 3D systems such as bulk topological semimetal Cd_3As_2 where the conventional 1D chiral Landau cyclotron orbits at the QHE regime can be re-constructed via connected anti-symmetric Fermi-arcs on opposite surfaces of the sample,[27] and bulk (100-μm-thick) $ZeTe_5$ where the transversal conductivity is quantized with respect to a product of the conductance quantum and a fraction of the electron Fermi wavelength, defined as a new type of 3D QHE.[28] All these recent experimental progresses suggest that dimensionality is dominated by the corresponding law of physics rather than the actual thickness of a sample. Therefore, the mindset that only monolayer can be called 2D is incorrect for nanoelectronics. Nevertheless, it is noticed that electronic band structures can be sensitive to the number of layers, such as in the graphene case. Monolayer graphene has a linear dispersion, whilst bilayer graphene has a parabolic one.[18,29]

With a clear definition of 2D in mind, one can summarize the revolutionizing nature of a freestanding 2D crystal (or, more specifically, 2D vdW layers), as illustrated in Fig. 3. Taking the 2D electronic system as an example, free electrons (Fig. 3(a)) are confined in a flat playground, which behavior as 2D gases or liquid depending on the strength of e–e interaction.[30] The state-of-art manipulation techniques allow us to stack freely those vdW 2D layers, and to study physics of inter-layer interactions when properly spaced by insulating vdW layers (Fig. 3(b)).[3,10,31,32] Because many of the vdW crystals are isolated from correlated electronic bulks, and they retain fascinating physical properties such as 2D superconductivity[12,33–36] and 2D ferroic ordering,[10,37–42] as shown in Fig. 3(c). Furthermore, thanks to the open-surfaces of 2D vdW crystals, they can be piled up with a certain, controllable, rotation angle with respect to each other, illustrated in Fig. 3(d). This is technically direct engineering of novel crystals in real space, which could lead to a modification of electronic band structure in reciprocal space – a property that belongs only to two dimensions.[43] In this review article, we will mainly go through recent experimental progresses according to the above several categorizations. The summarized systems belong mainly to vdW crystals, but not limited within the library of vdW family. They can be expanded into a broader conception of flattened crystals, as long as the physics is at play.

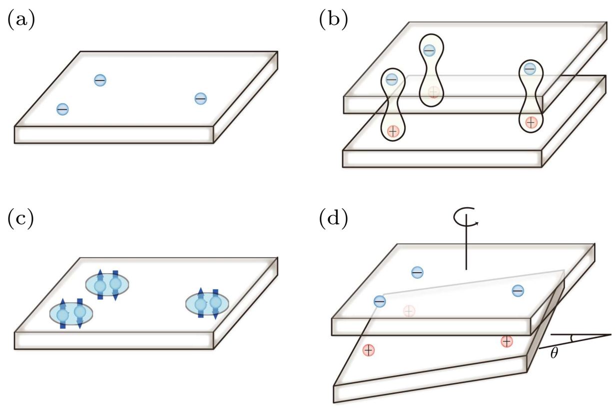

Fig. 3. Schematic illustrations of (a) free 2D electrons, (b) interlayer-interacted 2D electrons, (c) correlated 2D electrons, and (d) twisted 2D electronic systems.

2. New matters created when confined in 2D

Apparently, one of the noticeable achievements, in the great hunting of 2D crystals, is the discovery of a total new category of matters that exist in 2D, with distinct crystallographic arrangement and physical properties as compared to their forms in any other dimensionality. For example, the 2D form of group 14 elemental materials, silicene, germanene, and stanene, were proved to have distinguishing structural and electronic properties from their bulk ones.[44–51] Unlike their diamond-like semiconducting bulk structures, the metallic buckled honeycomb 2D forms were reported to be massless Dirac fermions holders.[47–49,51–53] Mechanically exfoliation method is not appliable to get their 2D forms, since their bulks structures are not van der Waals layered ones like graphite, while the specialized "bottom-up" (eg., MBE, CVD)[45–47,49,51–57] and "top-down" (eg., chemical exfoliation)[58,59] methods were broadly explored. Non planer but buckled 2D structures were found with sp^3+sp^2 mixed hybridizations forming the hexagonal symmetry lattice. And the massless Dirac fermions were predicted to be existed in them, while the linear energy–momentum dispersion coupled with a large graphene-liked Fermi velocity $\sim 10^6$ m/s[47] was revealed in the low-buckled epitaxial silicene sheet on the silver (111) substrate. Due to the stronger spin–orbit coupling effect compared with graphene, significant quantum spin hall effect was predicted to be existed in an accessible temperature regime.[49,60–63] Superconductivity, which is absent in the bulk α-tin, was also reported in the few layered stanene grown on the PbTe substrate.[64,65] Enhanced in-plane upper critical magnetic field (that can be diverged at low temperature) was found in few-layered stanene, which achieves a type-II Ising pairing superconductivity even without the participation of inversion symmetry breaking.[65]

As 2D crystals have two surfaces, their top and bottom surfaces can actually be of different chemical compositions. It is thus forming the famed 2D Janus material – a new matter that is believed to hold great promise for such as ferroelectricity thanks to the very low lattice symmetry.[66–72] So far, experimental progresses of 2D Janus materials are way behind the corresponding theoretical studies,[68,70,71] the physical properties of 2D Janus crystals remain largely unexplored.

3. Flattening functional materials into the 2D limit

Graphene and few-layered transitional metal dichalcogenides are two examples of the 2D crystals widely studied in the community since more than a decade ago.[18,31,34,73,74] Recently, 2D vdW functional materials, such as those with ferroic orderings, have attracted much attention because of emerging new physics and related potential applications.[8,10,37–41] For example, spin valves,[75] magnetic tunnel junctions,[10,11,76–84] intrinsic magnetic semiconducting transistors,[10,39,40] as well as other prototype spintronic devices have been demonstrated in the 2D limit using vdW materials as a platform.[85–89] On the other hand, ferroelectricity as a collective property of materials is at play in in many materials in the 2D limit. 2D vdW ferroelectric (FE) materials with dangling-bond-free surfaces and weak interlayer interactions have been reported in systems including MBE grown unit cell thin films,[7,90] insulating and semiconducting vdW 2D layers,[7,91–97] and even in those vdW 2D crystals such as $MoTe_2$ and WTe_2 whose bulk counterparts are not ferroelectric.[98,99] As vdW materials are often compatible with mass production processes such as CVD methods,[100–104] and can even be exfoliated into millimeter size.[105,106] The layer-stackable and -rotatable assemblies of those flexible vdW ferroic (or multi-ferroic) electronic devices can be revolutionizing in the very near future.[107,108]

Correlated two-dimensional electrons can also lead to quantum states such as 2D superconductors. As depicted by the 2D XY model, when cooled below the superconducting critical temperature, the 2D superconducting system will first go into a BKT dissipating phase in usually rather wide temperature range, before reaching a zero resistance ground state where vortices and anti-vortices are paired.[36,65,109] In early literatures, superconducting thin films were widely used to study the 2D superconducting to insulating (or to metal) quantum phase transitions at the vicinity of quantum critical point. For example, the quantum Griffiths singularity of superconducting-to-metal transition was reported in quite a few systems.[110] In the meantime, hybrid materials such as decorated graphene and related Josephson junctions brought the proximity effect into the 2D superconductivity, whilst keeping micron-meter-sized coherence length and gate tunable Fermi levels of the normal channel.[111–113]

Later on, thanks to the h-BN encapsulation in inert atmosphere, few layers with low air-stability isolated from intrinsic superconducting vdW materials were able to be assembled into nano-devices and go through the nano-fabrication process.[116,117] For example, $NbSe_2$ was found to be a Bose metal when thinned down to the 2D limit.[118] However, those bosonic metallic states are yet under debate because of the improper filtering may exist in many cryogenic setups.[119] It is noticed that when heavily doped, many of transition metal dichalcogenides (TMDCs) can turn into superconducting as well.[36] Some of the 2D crystals were reported to have spin–orbit locking, leading to the so-called Ising superconductivity.[34,65] 2D superconducting layers are also expected to exhibit topological behavior such as 1D Majorana edge mode when dressed with ferromagnetic 2D islands.[120] Recent growth method was reported to yield air-stable $NbSe_2$ monolayers in a mass production manner,[121] thus enabling possible batch fabrication of 2D superconducting devices from atomic vdW layers. Again, it can be compatible for foldable superconducting electronics because of the flexible nature of vdW 2D films.

Interestingly, the influence of underlying substrates can as well be a strong factor to affect the properties of 2D functional materials. For example, single layer FeSe film grown on $SrTiO_3$ (001) substrate exhibits a superconducting transition temperature of ~ 110 K (Figs. 4(a) and 4(b)), which is one order of magnitude higher than its bulk value.[114] FeSe has a very complicated phase diagram, the origin of enhanced superconducting T_c in monolayer tetragonal FeSe is attribute to the interfacial strain induced by the substrate underneath. Notice that as $SrTiO_3$ substrate undergoes a ferroelectric transition at low temperature, the resulted ultra-high dielectric constant can lead to a suppression of Coulomb scattering, thus giving rise to a robust quantum spin Hall state in graphene at modest magnetic field and relative rather high temperature.[122] By substrate engineering, p–n junctions,[123] mobility enhancement,[124] and non-volatile opto-electronics[125] can be achieved.

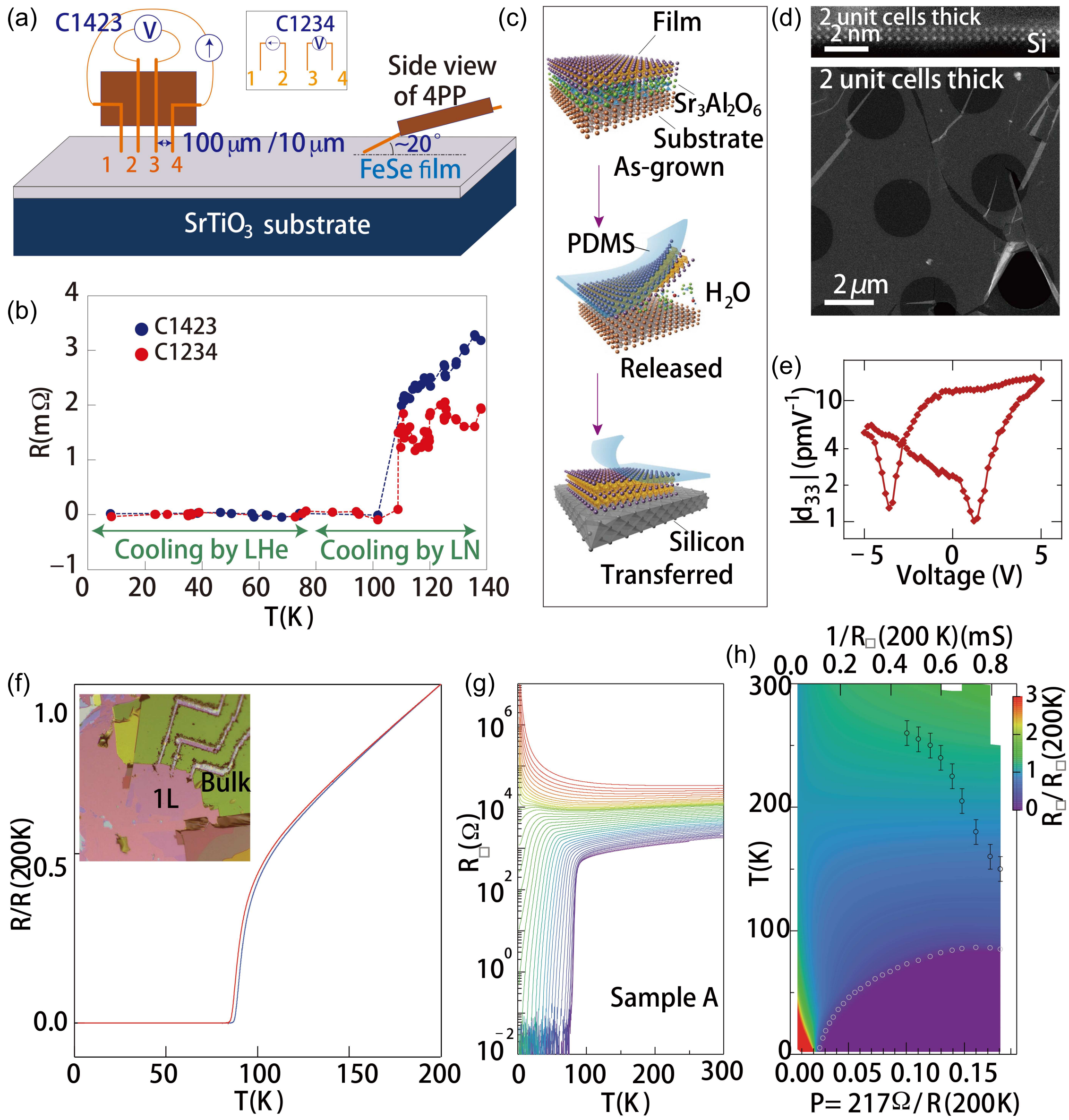

Fig. 4. (a), (b) FeSe monolayer epitaxially grown on $SrTiO_3(001)$ substrate, showing superconducting transition temperature above 100 K.[114] (c) Schematic picture of transfer process of isolated freestanding $SrTiO_3$ layers.[115] (d), (e) Characterizations of the $SrTiO_3$ few unit cells.[115] (f)–(h) Superconducting behavior of monolayered BSCCO.[9] Reproduced with permission from Refs. [9,114,115].

As discussed in the introduction part of this review, to be 2D or not, it does not really depend on whether the crystal is monolayer – it is rather a physical limit to pursue, even for non-vdW crystals. Indeed, to demonstrate the freestanding unit cell limit of the non-vdW perovskite crystal $SrTiO_3$ or $BiFeO_3$, a water soluble sacrificial buffer layer $Sr_3Al_2O_6$ was introduced to finish the removal of substrate via the assistant of a polymer stamp (Figs. 4(c) and 4(d)).[115] It was found that the ferroelectricity of $BiFeO_3$ sustains down to 4 unit cells, as shown in Fig. 4(e).

In many cases, flattening of a vdW crystal requires special effort as sometimes it is extremely difficult to tackle with because of both air-instability and quite strong inter-layer vdW bonding. For example, the cuperates high temperature superconductors, such as $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO), are known to be of vdW type. However, their thin layers degrade in air rapidly due to moisture and the loss of oxygen, and they are not easily-exfoliated. A dedicated Al_2O_3-assisted method was invented to overcome the later problem,[39] while the former can be mitigated by minimizing the exposure to air before electrical tests.[9] Strikingly, monolayered BSCCO shows almost the same superconducting transition temperature as that of its bulk, as shown in Fig. 4(f). The monolayer BSCCO is immune to Coulomb screening, and therefore allows one to study doping effect via ozone annealing, giving rise to a tunable critical temperature, as shown in Figs. 4(g) and 4(h).

Recently, $MnBi_2Te_4$ was found to be a co-host topological insulator and anti-ferromagnetic inter-layer coupling. When thinned down to the 2D limit with an odd number of layers, the system behaves as an Ising-type ferromagnetic topological insulator (MTI). The spontaneously broken time-reversal symmetry opens a gap at the gapless Dirac point, leading to the observation of a quantum anomalous Hall effect with chiral edge channels from both top and bottom surfaces at moderate magnetic fields close to the gap (Fig. 5(a)), as well as the observation of the conventional quantum Hall effects at higher magnetic fields and higher doping levels (Fig. 5(b)).[126] An MTI phase was also suggested in Sb-doped $Mn(Sb_xBi_{(1-x)})_2Te_4$ phase (Figs. 5(c)–5(f)).[127] More recently, high-Chern-number QHE, in which quantization is believed not of a Landau-level origin, was reported up to 60 K in seven-layered $MnBi_2Te_4$.[128]

Apparently, magnetic topological insulators or magnetic Weyl semimetals have become outstanding platforms to investigate exotic quantum states in the 2D limit.[129] Future experiments with broader range of carrier density via such as ionic gating or elemental doping, and at even higher magnetic fields, will be of interests to reveal more complicated transport features in such systems.

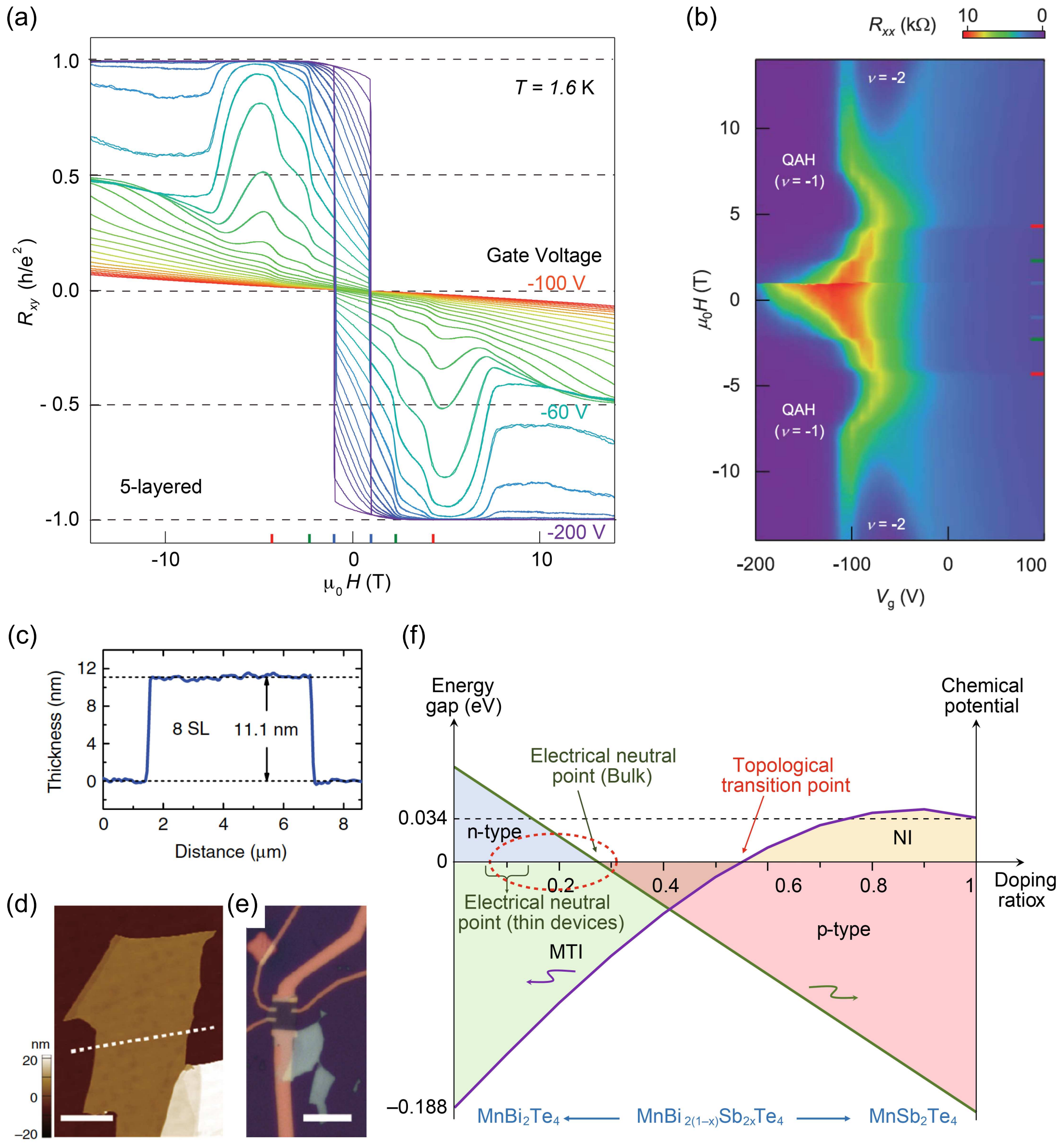

Fig. 5. Magnetic hysteresis loops measured in a 5-layered $MnBi_2Te_4$, with Hall resistance R_{xy} quantization indicated by dashed lines in (a) and color map of R_{xx} in (b).[126] (c)–(e) Morphology of a Sb-doped $MnBi_2Te_4$device with 8 monolayers.[127] (f) Phase diagram of Sb-doped $MnBi_2Te_4$ in the parameter space of gap energy, doping ratio, and chemical potential.[127] Reproduced with permission from Refs. [126,127].

4. Inter-layer interactions of 2D crystals

Inter-layer coupling in 2D vertical stacks is another widely studied phenomenon. For example, in TMDs, the optical response is dominated by intra-layer excitons, which are electron–hole paired quasi-particles due to the Coulomb interaction. When interfaced together, two pieces of TMDCs with different intrinsic doping levels give rise to a strong out-of-plane electric field, which results in a larger binding energy and longer lifetime for the interlayer excitons.[130,131] The interfacial coupling will take into account the lattice mismatch of each layer as well as their own physical properties, revealing unprecedented optical phenomenon, including moiré interlayer excitons[132–136] and exciton condensations probed by photoluminescence.[137,138]

Electronically speaking, separated double layer 2D electrons are also expected to yield exotic physics, due to long range Coulomb interactions when the spacing between them is small enough. In addition, the absence of interlayer tunneling significantly enhances the exciton lifetime. Typical experiment can be described in Fig. 6(a), in which two layers of 2D electrons are separated by a thin vacuum of spacing d, where current flow in one layer induces electrical signal in another, defined as a drag system. Here, the drag can happen between interlayer charges, spins, and other quasiparticles.[139–150] Before the flourish of graphene research, such kind of experiments can only be realized in the GaAs/AlGaAs double quantum wells,[151] which requires substantial efforts in sample preparations as illustrated in Fig. 6(b). The pitfalls of large interlayer space and non-tunable carrier types make the study of drag phenomena limited in these quantum wells.[152]

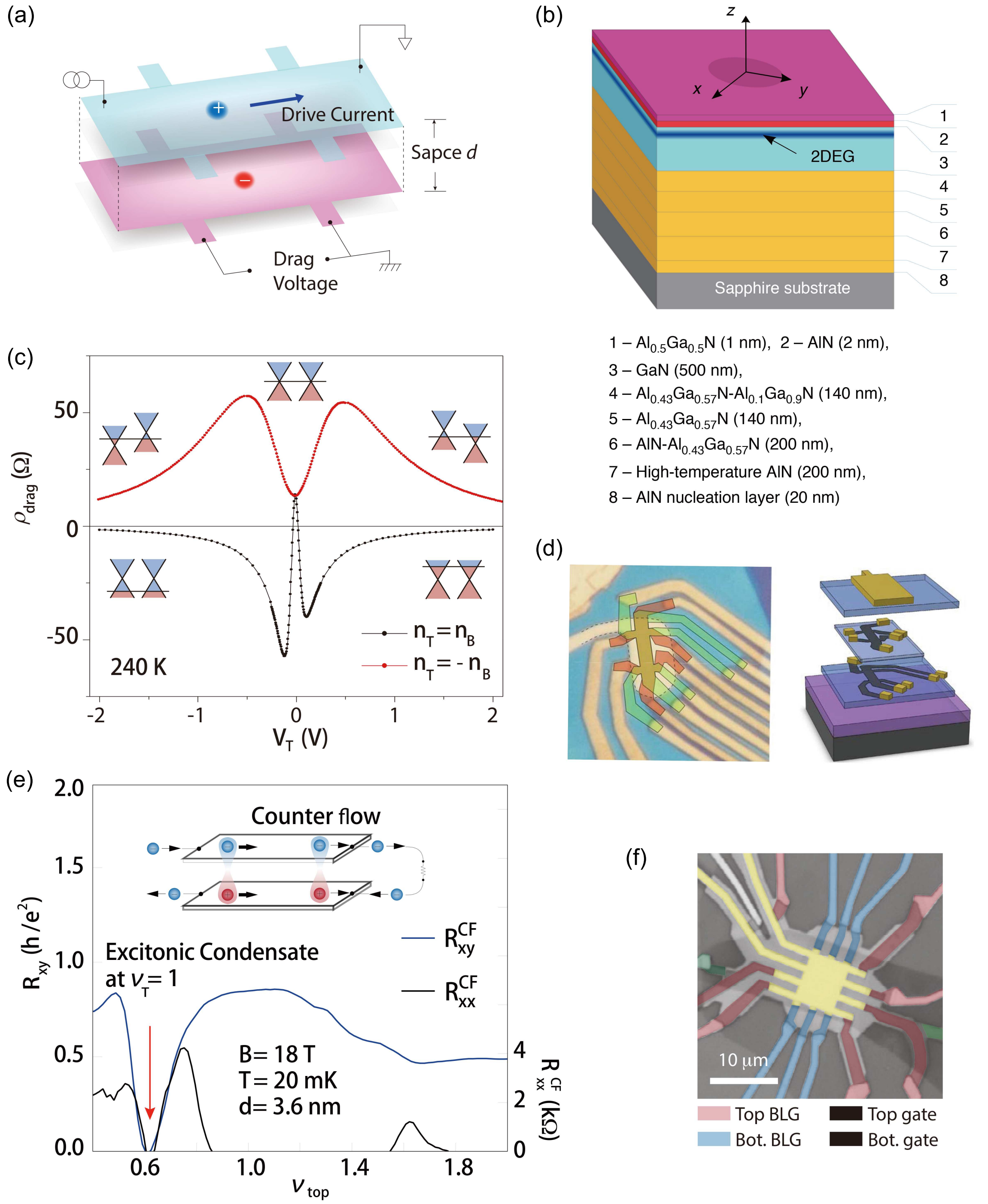

Fig. 6. (a) Schematic picture of a drag system with two separated 2D electrons. (b) Schematic picture of a GaAs/AlGaAs double quantum well sample.[152] (c), (d) Demonstration of Coulomb drag in graphene at zero magnetic field.[147] Width of the Hall bar is about 1.5 μm. (e), (f) Super fluid condensate realized in double bilayer graphene drag system.[142] The excitonic condensate, indicated by the red arrow, was examined in the top drag layer in a counter flow configuration at $\nu_T = 1$. (b)–(f) Reproduced with permission from Refs. [142,147,152].

The 2D insulating hexagonal boron nitride (h-BN) can act as an ideal spacer between the two drive-drag 2D electron layers. The thickness of h-BN can be tuned by simply choosing an appropriate number of layers, down to the single layer limit (0.34 nm). Furthermore, when encapsulated by h-BN, the carrier mobility of graphene can be much boosted as compared to that supported directly on SiO_2 wafers because of the clean and flat interface.[131,153] It therefore provides an outstanding platform to investigate Coulomb drag in double layer graphene with h-BN spacer using the transfer technique recently developed.[10,39]

Indeed, as shown in Figs. 6(c) and 6(d), graphene double-layer was found to show strong frictional drag at zero magnetic field, especially the drag signal became strongest at low density near the charge neutrality.[147] Recent efforts expanded this system into graphene double-layer structure in the quantum Hall regime in the presence of a large magnetic field.[142,154,155] As illustrated in Figs. 6(e) and 6(f), a signature of excitonic condensate was found at the filling fraction of one on the top drag layer in a counter-flow configuration.[142] In this scenario, due to the electron–hole paring, the bosonic excitons became immune to the magnetic field and the Hall voltage in the drag layer thus dropped to zero,[156] as indicate by the red arrow in Fig. 6(e).

5. Twistronics in rotatedly assembled 2D crystals

On the other hand, non-separated double layer systems can also yield bountiful exotic physics in electronic transport. When stacked together, two pieces of 2D crystal can form a moiré superlattice. For example, at the interface of graphene and h-BN, the moiré wavelength λ is defined as $\lambda=\frac{(1+\delta)a}{\sqrt{2(1+\delta)(1-\cos\phi)+\delta^2}}$,[157] where a is the graphene lattice constant, ϕ is the rotation angle between graphene and h-BN, and δ is the lattice mismatch (1.8% in the case of graphene/h-BN). Because of the moiré superlattice, a cloned miniband with Dirac dispersion at higher energy was formed in the band structure of graphene, giving rise to a pair of satellite resistance peaks versus gate voltage, and the so-called Hofstadter butterfly fractal Landau fan at high magnetic fields, as confirmed by transport,[158] capacitance,[159] and optical measurements.[160,161] Further experiments showed that this intriguing graphene–BN moiré superlattice can be constructed via chemical vapor deposition,[162] and even controlled with arbitrary ϕ in a rather high precision using mechanical manipulation of a scanning-probe tip.[163–165]

Graphene continues to amaze when considering the simplest case – by rotating-&-stacking two monolayered graphene with a certain angle, the consequential graphene–graphene (or twisted bilayer graphene TBLG) moiré superlattice will yield counter intuitive states including insulating,[13,14] superconducting,[12] and even magnetic behavior,[166] despite of the fact that both of the original layers are simple Dirac semi-metals (Fig. 7). The early pioneering theories predicted that, provided by a so-called magic angle of about 1.05° between two twisted layers of monolayer graphene, the entire lowest moiré band will be flattened (Figs. 7(a) and 7(b)).[167] Inside this flat band, at partial fillings with a much enhanced density of states, phenomena including superconductivity due to strong e–e interaction are supposed to happen.[168–170] These ground breaking results immediately triggered a worldwide gold-mining in twisting different 2D electronic layers, and gave birth to a new branch of condensed matter physics, called twistronics.[171]

TBLG with 1.8° rotation angle was first studied in 2016, and insulating states (corresponding to a single particle gap opening of 50–60 meV) were found at the Γ_s of the mini Brillouin zone induced by the moiré superlattice.[14] The smoking gun experiment of the superconductivity of BKT-type in the TBLG, with a magic rotation angle of $1.1^\circ \pm 0.5^\circ$, was reported very soon by the same group using the "tear-and-stack" method.[12,13] By defining carrier density n_s with 4 electrons per moiré unit cell (it is a band insulator at doping of $\pm n_s$ in magic angle TBLG, shown in Fig. 7(c)), a Mott-like insulating behavior was confirmed at the half-filling where $|n| = |n_s|/2 \sim 1.2\text{–}1.6 \times 10^{12}$ cm^{-2}, projecting owing to the many-body interaction picture. It was noticed that this kind of Mott-like behavior can be killed by bring the TBLG to the proximity of a monolayered WSe_2, possibly due to spin–orbit coupling reasons.[172]

Superconducting domes (with superconducting behavior possibly close to the crossover between BCS and BEC condensate) can be found slightly doped away from half filling. By examining the conductance evolution in parameter space of carrier density n and temperature T, it is seen that the magic angle TBLG system has a very similar phase diagram as that of high temperature superconductors.[12] Experiment confirmed that, by tuning interlayer spacing with hydrostatic pressure, TBLG with rotation angle larger than the magic angle can also exhibit a superconducting phase.[43]

Surprisingly, at 3/4 filling of the flat mini band, anomalous Hall signature was observed in the TBLG system at base temperature, with a hint of chiral edge state.[166] It is also noticed that this ferromagnetic hysteresis loop can be activated by exerting a current instead of sweeping the B field.[166] Higher quality magic-angled TBLG sample allows one to fully probe in details, at each fractional filling of the moiré flat band, the broken-symmetry states, interaction-driven insulators, orbital magnets, states with non-zero Chern numbers, and superconducting domes.[173] Further experiment demonstrated that the anomalous Hall signature in TBLG can actually be reaching the quantum anomalous Hall regime.[174]

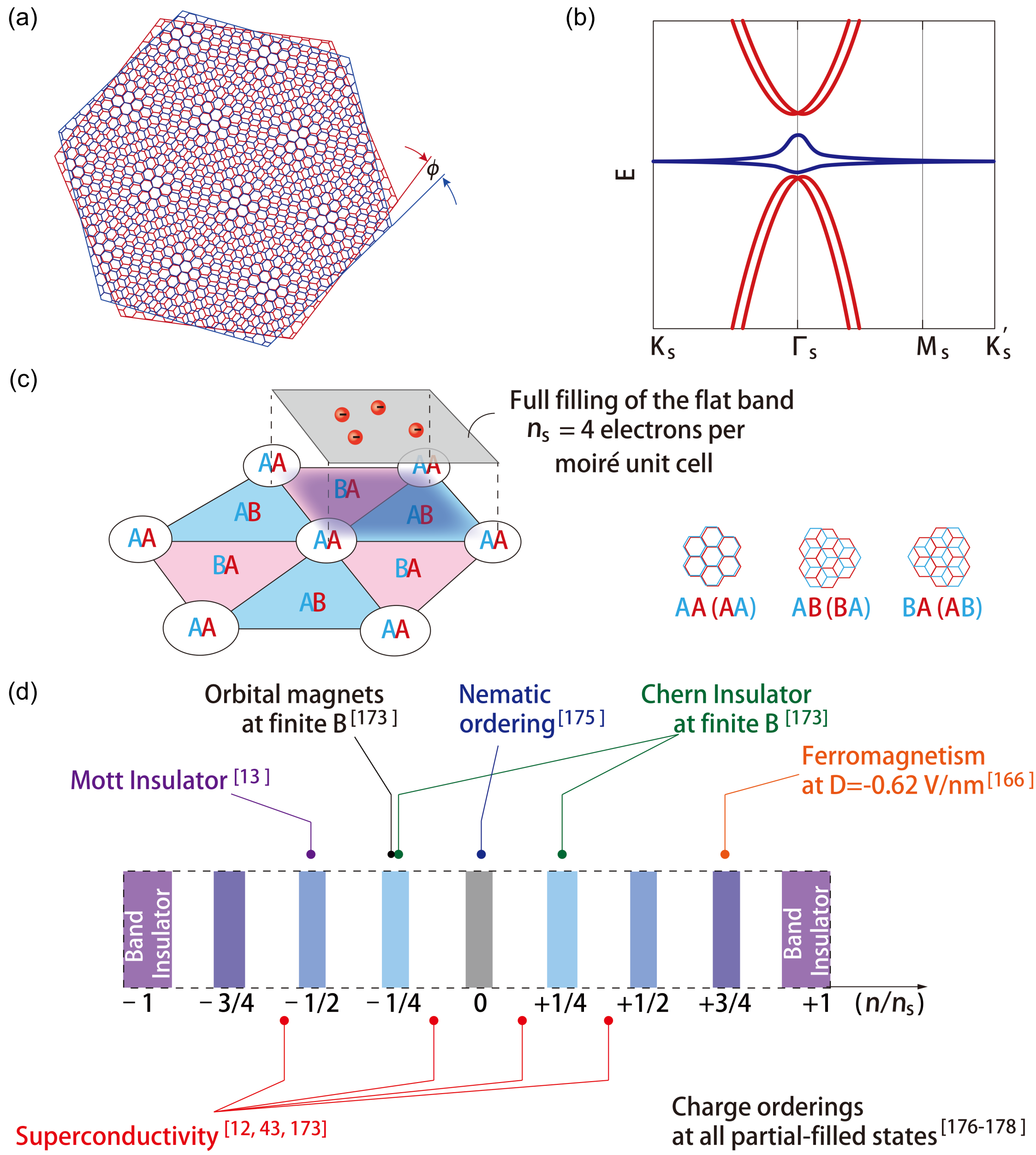

Fig. 7. (a) Schematic picture of a TBLG. (b) Illustration of the electronic band structure of a flat band (blue) induced by the magic angle moiré superlattice. (c) Schematic picture of the moiré superlattice with their stacking order marked with colors. A full filling of the mini flat band corresponds to 4 electrons in the moiré unit cell originated from the 4-fold degeneracy. (d) Summarization of state-of-the-art experimentally observed correlated quantum states in TBLG with the rotation close to the magic angle.

To unveil the underlying physics, visualization of the local density of states and charge distribution in magic angle TBLG was studied via scanning tunneling microscopy and spectroscopy (STM/STS).[175–179] Pseudogap phase with real-space global stripe charge ordering that breaks the rotational symmetry was observed at 4.6 K at partial filling of the flat mini band of the moiré superlattice, which is in close resemblance to high temperature superconductors.[176] Lower temperature (1.4–1.5 K) STS measurements show better developed $\mathrm{d}I/\mathrm{d}V$ spectra,[175,178] with those pseudogaps described using an extended Hubbard model cluster calculation,[175,178] and a possible nematic ordering at the charge neutrality point.[175] By checking TBLG samples with different rotation angles under STS at 5.7 K, it is found that the ratio of the Coulomb interaction to the bandwidth of each individual van Hove singularities (U/W) is maximized, which is the condition needed for Cooper pairing.[177] So far, dilution fridge temperature and high magnetic field studies are yet to be done using STS, which may release more fundamental physics such as to dig into the superconducting domes and the Chern insulator regimes.

The above emerging correlated quantum states (summarized in Fig. 7(d)), such as Chern insulator, superconducting condensate, and ferromagnetism, are also found in the flat band formed between a tri-layered ABC-stacking graphene and h-BN,[180–183] or in TBLG with rotation angles lower than the magic one,[184] and even systems of graphene/h-BN moiré lattice without flat band.[185] The experimental exploration of twistronics including twisted graphene is ongoing,[186–197] and has been expanded into twisted TMDCs and other systems.[164,198–204]

6. Conclusion and going beyond the flat crystals

As discussed above, flattened crystals have shown tremendous opportunities for studying fundamental physics. They also act as promising candidates for the implementation of future nanoelectronics. For example, by dual gating both top and bottom surfaces of a few-layered TMDC channel, photoswitching logic and memory can be integrated in a single small footprint device (Fig. 8(a)).[205] Ballistic avalanche phenomenon in a InSe/black phosphorous (BP) heterostructure was found to show sensitive 4 μm wavelength mid-infrared light detection with a sub 1 V avalanche threshold (Fig. 8(b)).[206] Local probe programmed ferroelectric domains were utilized to generate a TMDC homojunction, which gives rise to outstanding photo-detecting and rectifying behaviors (Fig. 8(c)).[207] Furthermore, structural engineering in a nano scale is also intriguing for developing the nanoelectronics based on 2D crystals. For instance, recent experiments have shown the feasibility to alter the conventional bulk semiconducting fin channel of a FinFET with a monolayered TMD, shrinking the fin-width into sub 1 nm limit (Fig. 8(d)).[208]

Noticeably, renewed efforts are being devoted to prepare 2D crystals recently, such as large scale production of high quality 2D single crystals, direct assembly of 2D heterostructures in a mass production manner, and new routes for engineering their electrical properties.[102,103,105,209–217] Moreover, the designs and fabrications of 2D layers with/from non-vdW materials are also thriving in the community.[218–220] Due to the limited length of this review, we regret that there are far more related works not included here. Nevertheless, it is believed that the plethora of 2D electronic systems will continue revolutionizing our research fields and future applications.

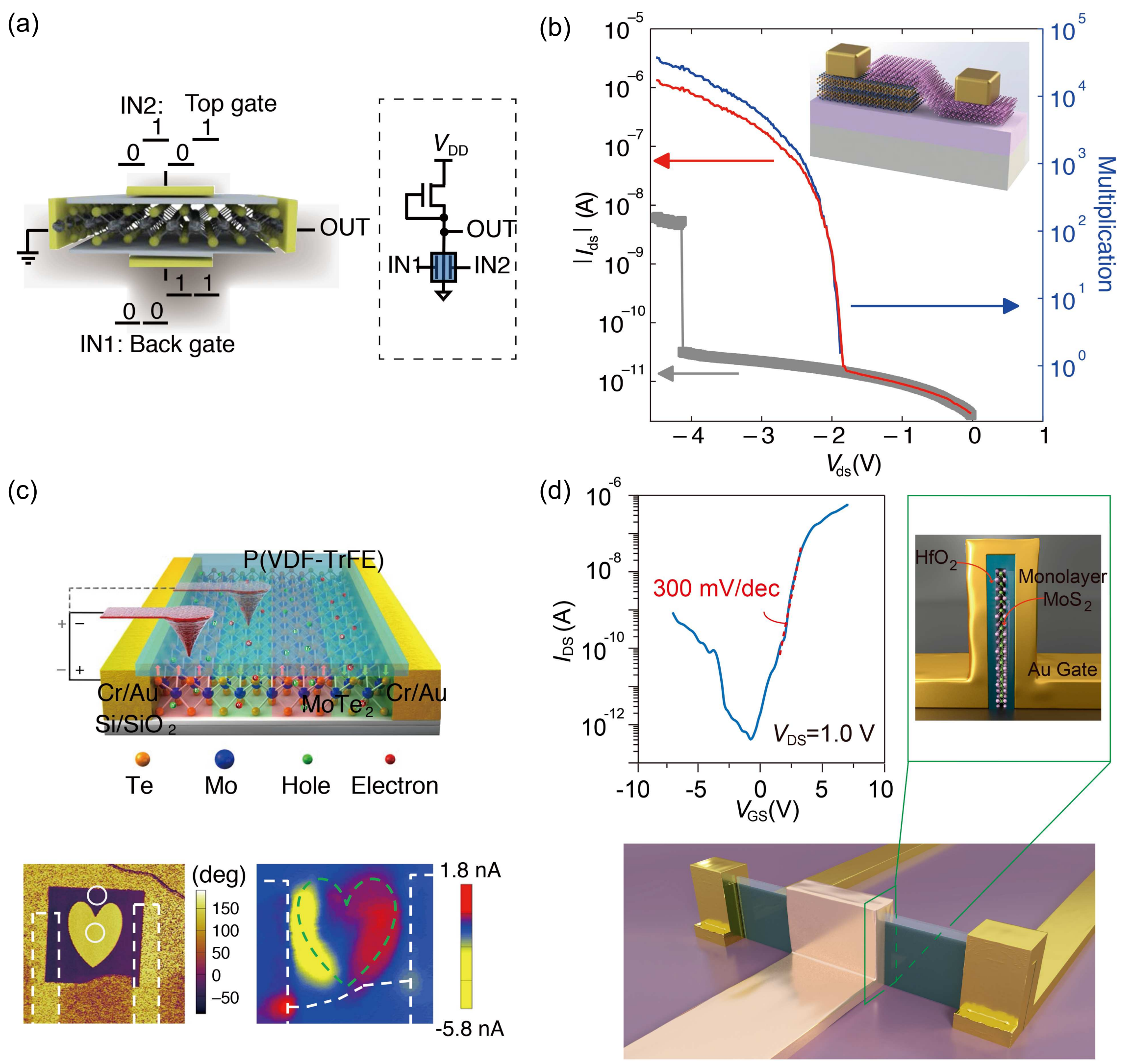

Fig. 8. (a) Schematic picture of a double gate programmable MoS_2 transistor, which is capable to integrate photoswitching logic and memory in a single cell.[205] (b) Ballistic avalanche transistor made of InSe/BP heterostructure.[206] (c) Ferroelectric domains realized by local patterning and the resulted photo detector with arbitrarily defined shapes.[207] (d) A FinFET using monolayered TMDC to replace the conventional Si fin channel.[208] Reproduced with permission from Refs. [205–208].

At this point, we would like to come to an end of this brief review article. As inspired by brain-experiments proposed by Feynman in early 1950s that there are plenty of rooms at the bottom,[221] it finally comes true that humankind can now stand at a technologically new era to play with isolated atomic sheets as demanded. Indeed, it has already been proven to be an exciting wealth to investigate the 2D electronic systems in all kinds of geometrical and electrical configurations. The resulted nanostructures have yielded insanely great amount of electronic states, sometimes of topological and/or quantum origins. An ending sentence is hereby drawn: when flattened, whether it is vdW or non-vdW, it is always flattering to explore the unexplored physics emerging in them or in their heterostructures, mechanically, optically, electronically, and more to be continued.

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