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Exotic Physical Properties of 2D Materials Modulated by Moiré Superlattices

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Abstract

Van der Waals heterostructures of two-dimensional materials are naturally endowed with the nanoscale moiré pattern, which has become a versatile platform for studying novel quantum phenomena during past decades. Here, we discussed the moiré superlattice combining graphene and hexagonal boron nitride (h-BN) can lead to the observation of electronic minibands, superlattice Dirac points and anomalous physical phenomena such as the quantum Hall effect and the Hofstadter butterfly pattern. The twist bilayer graphene (TBG) system has witnessed the appearance of angle-dependent van Hove singularities. Moreover, in the magic-angle twisted bilayer graphene (MATBG), there is a superconductor-insulator transition due to the enhancement of van der Waals forces by interlayer resonant conditions. Besides, owing to the moiré superlattice constant comparable to the Bohr radius of excitons, exciton physics dominated by moiré potential for the heterojunction based on transition metal dichalcogenides (TMDs) monolayers have attracted extensive attention. This review mainly covers the exotic physical properties, ranging from electronic structure to excitonic property of graphene-based and TMDs-based van der Waals-coupled twistronic materials, and discusses the remaining unsolved strongly correlated questions and possible expected semiconductor device applications in the quantum matter world.

Keywords:
Two-dimensional materials, Graphene, Transition metal dichalcogenides (TMDs), Moiré Superlattices, Quantum phenomena, Twistronics

1. Introduction

Two-dimensional (2D) materials have become the most popular material system since 2004 when graphene is mechanically exfoliated from bulk graphite. Galvanized by this breakthrough, researchers have by now isolated and studied dozens of 2D crystals and predicted the existence of many more. These atomically layered materials that exhibit intriguing properties distinct from those of their bulk counterparts can be fabricated as build blocks for advanced material properties. The so-called van der Waals (vdW) material is characterized by covalent bonding within the layer and weak vdW forces between the 2D plane. This has led many researchers to study the flatland regime of 2D materials ranging from fundamental physics2–3 to device applications such as high-performance photonic and optoelectronic devices.5–8 When two single layers are stacked with a rotation, they produce a moiré pattern of much larger length scale than the periodicity of each layer. The pattern is more often than not an incommensurate structure and exhibits twist-dependent emergent electronic behavior.9–12 Whereas an overarching periodicity emerges in the local atomic registry of the constituent crystal structures, which is known as a moiré superlattice.13–18

The moiré superlattice induced by the lattice mismatch or crystal orientation misalignment can
significantly modify the electronic and optical properties of heterostructures. Graphene/hexagonal boron nitride (h-BN), a typical vdW heterostructure, exhibits greatly improved properties for device applications by stacking graphene on top of h-BN. In the Graphene/h-BN heterostructure, the h-BN can reduce ripples and suppress charge inhomogeneities to improve the carrier mobility of graphene, as well as provides unique opportunities to open the band gap of graphene at the Dirac point by the periodic superlattice potential, which leads to various anomalous quantum phenomena including the quantum Hall effect and the Hofstadter butterfly pattern. Twisted bilayer graphene (TBG) is simply fabricated by changing the relative orientation between the layers, which induces low-energy Van Hove singularities (VHSs). This may intrigue prospects for VHS engineering of electronic phases. In some special case, the two graphene layers are twisted relative to each other at a small angle which called the magic angle, and there is a correlated insulator and unconventional superconductor transition due to the localization of electron within the moiré superlattice.

Though the distinctive linear dispersion of graphene yields unique electronic and optical properties, the zero-band gap greatly hinders its applicability in semiconductor based elements and devices. The TMDs monolayer exfoliated from bulk crystal possess direct energy gaps in the range of 1~2 eV, opened up a new window for optoelectronics. The reduced dimensionality and the associated reduced dielectric screening result in the greatly enhanced excitons binding energy in monolayer TMDs. The strong excitonic effects allow for room temperature excitonic devices that have promising prospects for overcoming the dilemma of response time and integration in electron or/and photon based devices. Heterobilayer of TMDs is particularly attractive for low-dimensional semiconductor optics because they host interlayer excitons—with electrons and holes localized in different layers and thereby possess various novel and appealing properties, especially in a typical type-II band alignment, where electrons and holes find their energy minima in different layers. Besides, owing to the high sensitivity of TMDs to crystal structure symmetry and interlayer coupling, the bandgap of TMDs heterobilayers composed of various 2D materials is of big difference. Furthermore, the moiré potential determined by interlayer coupling in TMDs heterobilayers can localize interlayer excitons and modulate the multiple interlayer exciton resonances, suggesting the feasibility of engineering artificial excitonic crystals can provid a unique platform for studying novel quantum phases of matter.

In this review, we mainly aim to describe exotic electronic and excitonic properties modulated by nanoscale moiré pattern in graphene-based and TMDs-based heterostructures. Firstly, we overview how the electronic structure of graphene is modified by moiré superlattice and the anomalous quantum phenomena of graphene/h-BN. Then we discuss the transport properties of twisted bilayer graphene. Moreover, the interplay of correlated insulating states and superconductivity of twisted bilayer graphene is also described. Finally, we review excitonic phenomena determined by moiré potential in twisted TMDs heterobilayers and came up with serval possible perspectives to inspire the research enthusiasm for semiconductor quantum light-emitting devices based on moiré superlattices.

2. Quantum Phenomena in Graphene Moiré Superlattice
Graphene has attracted great attention owing to its remarkable properties as the first 2D vdW
material. The recent development of experimental techniques for combining graphene with other atomically thin vdW crystals has enabled the exploration of the properties of these so-called vdW heterostructures, which provides an exciting approach to design quantum materials that can harness and extend the already fascinating properties of the constituents. In this section, we first discuss the exotic experiment phenomena in the emergent graphene/h-BN moiré superlattice, such as superlattice Dirac points and Hofstadter butterfly spectrum. Then we illustrate the intriguing properties in the twisted bilayer graphene (TBG) due to stronger interlayer coupling than that in graphene/h-BN moiré superlattice. Finally, we describe two correlated quantum states of correlated insulating states and unconventional superconductivity caused by strong electron-electron Coulomb interaction in the magic-angle twisted bilayer moiré superlattice of graphene.

2.1 Graphene/h-BN Moiré Superlattice

Heterostructures of 2D vdW materials offer a platform for engineering novel optoelectronic properties. H-BN is another well-known vdW material that is commonly used. It can provide high and homogeneous dielectric circumstances for graphene, enabling the observation of the quantum Hall effect (QHE) and fractional quantum Hall effect (FQHE) in graphene/hBN heterostructures. Additionally, h-BN can modify the optoelectronic properties of graphene when aligned with graphene under certain circumstances, for example, the arising of superlattice Dirac points, accompanied by the appearance of the Hofstadter butterfly spectrum. The graphene/h-BN heterostructure provides an opportunity for engineering the properties that can be achieved by tuning each of these degrees of freedom, thereby facilitates the design and realization of graphene-based devices.

2.1.1 Quantum Hall Effect in Graphene

Quantum-mechanically enhanced transport phenomena such as the quantum Hall effect can be observed when electrons are confined in two-dimensional materials. Fig. 1a shows a schematic of a Hall bar device made of graphene and h-BN used to measure the quantum Hall effect. Highly degenerate Landau-levels (LLs) can be formed when graphene is applied with a perpendicular magnetic field. The energy dispersion of the LLs is given by

\[ E_n = \text{sgn}(n)\sqrt{2\hbar v_F^2|nB|}, \]

where \( e \) is the electron charge, \( \hbar \) is Planck’s reduced constant, the integer \( n \) represents an electron-like (\( n > 0 \)) or a hole-like (\( n < 0 \)) LL index, \( v_F \) is Fermi velocity and \( B \) is the magnitude of the applied magnetic field. Unlike the traditional two-dimensional electron gas, the LLs formed in the graphene under magnetic field are not equally spaced in energy, as shown in Fig. 1b. When the Fermi level \((E_F)\) of graphene crosses a LL by adjusting the magnetic field, the Hall conductance \( \sigma_{xy} \) appears quantized plateau

\[ \sigma_{xy} = \nu \frac{e^2}{h} = g(n + \frac{1}{2}) \frac{e^2}{h}, \]

where \( \frac{1}{2} \) comes from the Berry phase, \( \nu \) is the quantized filling factor of LLs and \( E_F \) jumps by an amount of \( g e^2/h \). The longitudinal resistance \( R_{xx} \) is zero while the Hall resistance \( R_{xy} \) exhibits quantum Hall plateau, marking the system enters the quantum Hall effect (Fig. 1c). Note that \( \sigma_{xy} = -R_{xy}^{-1} \) and \( \sigma_{xx} = -R_{xx}^{-1} \). This Landau quantization is a result of the linear band
dispersion of graphene. Additionally, the valley degree of freedom in graphene adds an extra twofold degeneracy, but in most materials, the degeneracy of each LL is doubled owing to the spin degree of freedom. Considering the valley and spin degree of freedom of graphene, there are half-integer multiples quantum Hall plateaus, that is, the fractional quantum Hall effect. Fig. 1d shows an expanded view in the $N = 0$ LL between $\nu = 0$ and $\nu = 1$. Two-flux composite fermion states (centered around $\nu = 1/2$) and four-flux composite fermion states (centered around $\nu = 1/4$) are observed.\textsuperscript{53}
Fig. 1 (a) Schematic of a Hall bar device made of graphene and h-BN used to measure the quantum Hall effect.51 Copyright 2011, Springer Nature Limited. (b) A schematic diagram of LLs DOS formed by quantization of Dirac fermions under external perpendicular magnetic field.50 Copyright 2005, Springer Nature Limited. (c) Hall resistance (black) and magnetoresistance (orange) as a function of gate voltage under fixed magnetic field and low temperature. The upper
inset is a diagram of high-filling-factor plateau measured at different temperature.\textsuperscript{50} Copyright 2005, Springer Nature Limited. (d) Device conductance ($\sigma_{xx}$) at B = 15 T between $\nu = 0$ and $\nu = 1$. The red labels indicate 2-flux fractional quantum Hall effect and blue labels indicate 4-flux sequence.\textsuperscript{54} Copyright 2019, Springer Nature Limited.

### 2.1.2 Quantum Phenomena in Graphene/h-BN Moiré Superlattice

In the graphene/h-BN heterostructure that rotational mismatch is small, a long-period geometric interference pattern can be produced. Owing to the electronic and optical properties of heterostructure can be significantly modified by moiré pattern, the unique low-temperature transport phenomena of graphene/h-BN moiré superlattice have been studied extensively. Fig. 2a shows the STM image of the graphene/h-BN moiré pattern. The period of this moiré pattern is very sensitive to lattice mismatch $\delta$ between graphene and h-BN and their relative rotational orientation. This relationship can be described as\textsuperscript{16}

\[ \lambda = \frac{(1+\delta)a}{\sqrt{2(1+\delta)(1-\cos \theta)+\delta^2}} \]  

(3)

where $\lambda$ is the moiré periodicity, $\delta$ is the lattice mismatch between h-BN and graphene which is less than 2%, $a$ is the lattice constant of graphene, and $\theta$ is the relative rotation angle between the two lattices. Fig. 2b inset shows that the moiré periodicity increases when the relative rotation angle decreases. STM measurements reveal that the moiré superlattice has a fluctuation of 20 pm in space through the epitaxial growth of graphene/h-BN samples. The strength of van der Waals interaction between carbon atoms and the h-BN substrate is inversely proportional to their distance. Therefore, the moiré pattern effectively acts as a periodic superlattice electrical potential for graphene.\textsuperscript{17} The resulting periodic potential enables scattering processes along the directions of its reciprocal lattice vectors which are otherwise forbidden in graphene. The so-called superlattice Dirac points (SDPs) are created in the band structure where the periodic potential connects the $k$ and $-k$ bands. The appearance of the SDPs originates from the moiré superlattice attached to graphene and is located at the Brillouin zone boundaries of the superlattice in the reciprocal space. Indeed, the researchers have systematically studied the SDP. The scanning tunneling spectroscopy (STS) discovered the DOS of these SDPs symmetric with respect to the primary Dirac point (PDP) of graphene/h-BN moiré superlattice.\textsuperscript{16} The energy dispersion of these SDPs is extracted as a function of the moiré periodicity (Fig. 2b). It is clear that the energy of the SDP decreases as the moiré periodicity increases. The observation of SDPs confirms the modulation of the band structure of graphene by moiré periodic potential.\textsuperscript{13, 16, 17} Moreover, various experimental measurements demonstrate the sizeable bandgaps at the PDP and SDPs in the valence band.\textsuperscript{13, 25, 55-60} A schematic band structure of graphene/h-BN heterostructure with a small twist angle is shown in Fig. 2c. These bandgaps are understood as a result of the sublattice symmetry breaking, owing to the imbalance of carbon-on-boron and carbon-on-nitrogen stacking within the graphene. The measured bandgaps at the primary and superlattice Dirac points are extracted as a function of twist angle (Fig. 2d). It is clear that the gap at the SDP is typically smaller than that at the PDP and vanishes at $> 1^\circ$ misalignment.\textsuperscript{18, 58} Note that the bandgap can be further broadened by many-body interactions.\textsuperscript{61, 62}
Fig. 2 (a) STM image of moiré pattern observed in graphene/h-BN heterostructure owing to the lattice mismatch between them. (b) The energy of the superlattice Dirac points as a function of moiré periodicity. The inset displays the wavelength of the moiré superlattice as a function of the twist angle (θ) between graphene and the h-BN substrate. (c) Schematic band structure for graphene/h-BN heterostructure with a small twist angle. E, energy; k, momentum. (d) Measured bandgaps at the primary and superlattice Dirac points as a function of twist angle within a single graphene/h-BN device. Figure adapted with permission from: (a) ref.25 Copyright 2014, Springer Nature Limited. (b) ref.54 Copyright 2019, Springer Nature Limited. (c), (d) ref.58 Copyright 2018, AAAS.

The long-period moiré pattern in graphene/h-BN heterostructure provide a unique system to explore the interplay between electrical potentials and magnetic fields over similar length scales. Transport properties of graphene/h-BN devices indicate that the SDPs manifest themselves as additional peaks in the resistance symmetrically flanking the primary Dirac point.13-15 The charge carrier density n-dependent conductance measurements show that the longitudinal resistivity $\rho_{xx}$ is a maximum at both sides of PDP and SDP (Fig. 3a), whereas the Hall resistivity $\rho_{xy}$ drops suddenly at one side of the PDP and SDP and increases at the other side (Fig. 3b), reversing in sign. To present the detailed electronic structure of PDP and SDPs, the $R_{xx}$ and $R_{xy}$ are plotted as a function of gate voltage and magnetic field, which is called Landau fan. The Landau fan diagram shows that the degeneracy of Landau level formed by two-dimensional electron gas increases and enters the quantum Hall state gradually as the magnetic field increasing. $R_{xx}$
reaches a minimum when Fermi level crosses a LL and $R_{xy}$ exhibits quantum Hall plateau at the corresponding position. Similarly to the Dirac point, these SDPs break up into sequences of LLs when applied magnetic field. Transport measurements show that these LLs emerge from each of these points, including PDP and SDPs, at low magnetic fields and intersect as the magnetic field increasing, resulting in series of energy levels, which is known as a Hofstadter butterfly spectrum (Fig. 3c, d).

The Hofstadter butterfly was predicted in 1976, it pointed out that the 2D electron gas would form a fractal energy spectrum if it is simultaneously under periodic electric field and magnetic field. By tuning the magnetic field, a fractal of the energy band is completed when a unit cell is filled with one magnetic flux quantum $\hbar/e$. However, the small period (~1nm) of electrical potentials in ordinary crystal lattice leads that one magnetic flux quantum filling needs magnetic field of more than 1000 T, which is difficult to achieve in experiment. Intriguingly, Graphene/h-BN moiré superlattice with a moiré period about 10nm can reduce this magnetic field to approximately 30 T, making Hofstadter butterfly spectrum visible in laboratory-scale magnetic fields. Moreover, in graphene/h-BN moiré superlattice system, these LLs intersect when

$$\frac{\Phi_0}{\phi} = q \quad (4)$$

where $\Phi_0$ is the magnetic flux quantum through superlattice unit cell, $\phi$ is the magnetic flux and $q$ is a positive integer. Fig. 3e displays a Wannier diagram showing the quantum Hall effect states as a function of charge carrier density, highlighting the Hofstadter states experimentally observed in graphene/h-BN. Particularly, black, blue and red lines indicate fractal integer quantum Hall, conventional fractional quantum Hall and anomalous quantum Hall states, respectively. Though there are many researches about Hofstadter spectrum, further studies are needed to understand the rich physics in Hofstadter butterfly.
2.2 Strongly Correlated Quantum States in Twisted Bilayer Graphene

Among existing 2D materials, graphene is the most stable material with the most mature and successful preparation technology as well as the most extensive and in-depth research in its physical properties. In previous reports, the properties of graphene can be understood very well based on single electron approximation, without considering the electron-electron interaction. However, there is a wealth of strongly correlated quantum states in magic-angle twisted bilayer graphene. Jarillo-Herrero’s group observed two correlated quantum states of correlated insulating states and unconventional superconductivity caused by strong electron-electron interaction.

Note that there are three methods that can experimentally induce electron-electron interaction in graphene. The most commonly used one is to generate Landau levels in graphene by applying a
perpendicular external magnetic field. The fractional quantum Hall effect is a kind of strongly correlated quantum state of matter induced by a strong magnetic field. The second method is to modulate the Fermi level of monolayer graphene located on the Van Hove peak, which may make graphene exhibit anomalous magnetic or superconducting properties.\textsuperscript{64-69} However, the gap between Van Hove peaks of monolayer graphene and the neutral point, which is the Dirac point, is about \(3\) eV, the current experimental technology is unable to realize such modulation. Therefore, inducing magnetism and superconductivity into monolayer graphene through electron-electron interaction has always been studied theoretically. The third method proposes that a strongly correlated quantum state can be induced in the twisted bilayer graphene. The intuitive physical scenario is as follows: there are twist-angle-dependent low-energy Van Hove singularities (VHSs) in the TBG and the energy gap between VHSs and Dirac points of the system is generally less than a few hundred millielectron volts, which make it possible to tune Fermi level to VHS accurately in the experiment.\textsuperscript{70-72} The twisted bilayer graphene system provides an opportunity to experimentally realize the strongly correlated electronic state.

2.2.1 Van Hove Singularities in TBG

Prior to the experimental discovery of graphene/h-BN superlattices, the related experimental research on twisted bilayer graphene has attracted intense interests since 2009. The most important difference between the two kinds of moiré superlattice is that the band gap of h-BN is about \(5.9\) eV, which makes the low energy band of graphene unable to hybridize with h-BN. While in twisted bilayer graphene moiré superlattice, the upper and lower graphene energy bands are coupled with each other. In graphene/h-BN structures, the presence of a moiré superlattice can lead to the observation of electronic minibands, whereas in twisted graphene bilayers its effects are enhanced by interlayer resonant conditions. The interlayer coupling strength of electrons is periodically distributed in the TBG moiré superlattice system, which may strongly modify the electronic property of the individual graphene.

The TBG moiré superlattice is a homostructure, thus the lattice mismatch \(\delta\) is zero. Therefore, the formula (3) can be simplified as

\[
\lambda = \frac{a}{\sqrt{2(1-\cos \theta)}} \quad (5)
\]

In the energy band structure of TBG, Dirac points corresponding to the upper and lower layers are separated under the rotation angle, forming \(K\) and \(K_0\) points (Fig. 4a). Along the direction of separation of the Dirac points, a saddle point is formed in the middle of the two Dirac points, which is called Van Hove singularity.\textsuperscript{73} corresponding to the two maximum of the density of states. The twisted bilayer graphene with different rotation angles was studied by using STM and the scanning tunneling spectroscopy.\textsuperscript{29, 74, 75} Because the STS spectra reflect the local electronic density of states of the sample, there are two characteristic peaks corresponding to Van Hove peaks, to the two saddle points in the band structure of TBG (Fig. 4b). The left panel is the spectra of two VHSs and the right panel shows two saddle points form at \(k = 0\) between the two Dirac cones, \(K\) and \(K_0\), with a separation of \(\Delta K = 2K \sin(\theta/2)\). Many intriguing phenomena caused by the VHSs in the TBG, including enhanced optical absorption, Raman G-band resonance and enhanced chemical reactivity, are studied in the last decades.\textsuperscript{76-84} Moreover, for TBG moiré superlattice, it has been demonstrated that the energy interval of Van Hove peaks, \(\Delta E_{\text{VHS}}\), is twist
angle-dependent, and the relationship approximately satisfies

$$\Delta E_{VHS} = \hbar v_F \Delta K - 2t_\theta$$  \hspace{1cm} (6)$$

where $v_F$ is the renormalized Fermi velocity and $t_\theta$ is the interlayer coupling coefficient. Assuming the coupling coefficient between layers is constant, $\sin \theta \approx \theta$ for small rotation angle ($\theta < 6^\circ$), the gap between the Van Hove peaks is proportional to the twist angle. $\Delta E_{VHS}$ increases as the twist angle increasing, which is consistent with the experimental results (Fig. 4c). Moreover, the photocurrent can be significantly enhanced when the incident photon energy matches the energy interval between VHSs of the conduction and valance band in the TBG photodetection device, which suggests the existence of strong light–matter interaction. Therefore, the emergence of twist angle-dependent VHSs in TBG can selectively enhance photocurrent generation, providing valuable insight for designing graphene-based photodetectors with variable wavelength.

Fig. 4 (a) Schematic Dirac cones, K and $K_\theta$ of two misoriented graphene layers in the reciprocal space. (b) Left: Tunneling spectra recorded at different positions of a moiré pattern. Right:
Electronic band structure of the TBG with a finite interlayer coupling. (c) Angle-dependent VHSs in twisted graphene bilayers. Solid blue circles: average experimental data measured in different twisted graphene bilayers. The gray dotted line: linear fit of the experimental data. (d) Renormalized Fermi velocity as a function of twist angle obtained from Landau-level spectroscopy (symbols) compared to a theoretical simulation (solid line). (e) Renormalized Fermi velocity, \( v^* = \frac{v_F(1 - 3\alpha^2)}{(1 + 6\alpha^2)} \), as a function of twist angle in the range \( 0.18^\circ < \theta < 1.2^\circ \). shows a sequence of magic twist angles at which the Fermi velocity vanishes. (f) The band structure of MATBG shows the formation of flat bands (blue lines) when approaching the magic angle. Figure adapted with permission from: (a), (c) ref. 85 Copyright 2014, American Physical Society; (b) ref. 74 Copyright 2012, American Physical Society. (d) ref. 88 Copyright 2011, American Physical Society. (e) ref. 72 Copyright 2011, National Academy of Sciences. (f) ref. 30 Copyright 2018, Springer Nature Limited.

Additionally, the experiments also proved that the low-energy region of the TBG has an angle-dependent Fermi velocity.\(^{88, 89}\). When the twist angle is not very small (\( \theta > 3^\circ \)), the Fermi velocity can be renormalized, and the relationship is as following\(^{70-72, 90}\)

\[
\frac{v^*}{v_F} = 1 - 9\left(\frac{t_0}{\hbar v_F \Delta K}\right)^2
\]

Obviously, the Fermi velocity in the low-energy region of the TBG continues to decrease as the twist angle becomes smaller and drops to zero at the first magic angle (Fig. 4d, e), leading to the appearance of two flat bands in the low-energy band structure of the system (Fig. 4f).\(^{72}\) Fig. 4e shows the twist angle expressed in terms of the dimensionless parameter \( \alpha = \frac{w}{\hbar v_F \Delta K} \), where \( w \) is the interlayer tunnelling strength and \( \Delta K \approx 2K \sin(\theta/2) \) as defined in the above text. The calculation results show that the bandwidth of flat bands in magic-angle TBG is approximately 12 meV (\( E > 0 \)) or 2 meV (\( E < 0 \)). The flat band of the MATBG was measured experimentally for the first time in 2015, and the corresponding magic angle was about 1.11\(^{\circ}\).\(^{91}\)

2.2.2 Strongly Correlated Quantum States in MATBG

Due to the lack of mature and accurate twisted graphene preparation technology, it was until 2018 when Cao et al. successfully measured two correlated quantum states of insulating and unconventional superconductivity caused by strong electron-electron interaction in the magic-angle twisted bilayer moiré superlattice of graphene under extremely low-temperature.\(^{30, 31}\)

In Hubbard model, the correlation effect of electrons is determined by \( U/W \), in which \( U \) and \( W \) represent mutual Coulomb repulsion between electrons and kinetic energy of electrons, respectively. The correlation effect of electrons is significant when the mutual Coulomb repulsion between electrons dominates over their kinetic energy, \( U/W \gg 1 \).\(^{92-96}\) In the TBG superlattice, the Coulomb interaction \( U \) satisfies

\[
U = \frac{e^2}{4\pi \varepsilon_0 \varepsilon_r d}
\]

where \( d \) is the distance between electrons, \( \varepsilon_0 \) and \( \varepsilon_r \) are the vacuum permittivity and relative permittivity, respectively. Let’s take \( d = \lambda/2 = 6.5nm \) (\( \theta = 1.1^\circ, \lambda = 13nm \)) and \( \varepsilon_r \) is set to 4, then \( U \approx 55.4 meV \). Here, 1.1\(^{\circ}\) is the so-called first magic angle. As mentioned above, the Fermi velocity could drop to zero at the magic angle in TBG, leading to the emergence of flat bands, which presents weak dispersion in momentum space, with a bandwidth of about 10meV. The upper limit
of electrons kinetic energy $W$ is the flat band bandwidth when the electron concentration is tuned to limited fill the flat band near zero energy. The upper limit of electron kinetic energy $W$ is the flat band bandwidth when the electron concentration is only tuned to fill near the zero energy of flat bands (Fig. 5a). Therefore, strongly correlated electronic phases are expected to be generated in MATBG because the condition $U/W >> 1$ is satisfied. The flat-band DOS peak in MATBG revealed by STM/STS measurements illustrates that the electron is highly localized in space with the inversion symmetry breaking of the electronic state distribution when the Fermi level lies within the flat bands in the low-energy region, which was attributed to the strong electron-electron interaction in the system (Fig. 5b).
distribution diagram of electrons local density of states (LDOS) calculated for the flat bands with $E > 0$ in the MATBG. The electron density is strongly concentrated at the regions with AA stacking order, but it is mostly depleted at AB- and BA-stacked regions. (c) Carrier dependence of conductance in MATBG. (d) Phase diagram showing the carrier density-dependent superconducting critical temperature in the partially filled flat band. There are two dome-shaped superconducting regions (blue) flank an insulator (red). (e) Density dependence of $T_c/T_F$ as a function of doping for MATBG (red filled circles) compared to other strongly correlated systems. $T_c$: superconducting temperature, $T_F$: Fermi energy. Figure adapted with permission from: (a)-(c), ref.\textsuperscript{30} Springer Nature Ltd; (d), (e), ref.\textsuperscript{31} Springer Nature Ltd.

The strong correlation effect of electrons often accompany by the Mott-like insulator states transition.\textsuperscript{97} The sample is expected to behave like metal by single-particle band energy theory when the band is not full filled with electrons. However, it actually behaves like an insulator owing to the strong correlation effect of electrons when the energy band is half-filled, which is usually observed in cuprate materials.\textsuperscript{96-101} Similarly, low-temperature (~1K) electrical transport measurements on MATBG revealed interaction-induced correlated insulating states when the flat band is half-filled with electrons, thus the state near $\pm n_s/2$ is called half-filling insulating states (Fig. 5c). The MATBG system exhibits two pronounced superconducting domes on each side of the half-filling correlated insulating state when using gate voltage to continuously tune the carrier concentration of the system, with the currently obtained highest superconducting transition temperature of 1.7 K (Fig. 5d). Whereas the MATBG system exhibits a quantum anomalous Hall effect (QAHE) at 3/4 filling of the conduction miniband. The QAHE combines topology and magnetism to produce precisely quantized Hall resistance at zero magnetic field. The effect is driven by the spontaneous valley polarization in valley resolved moiré bands with opposite Chern number.\textsuperscript{102-105} In the recent papers, the researchers observed emergent ferromagnetic hysteresis, reflecting a topological state with Chern number $C = 1$ that is an orbital ferromagnet. Moreover, the magnetic order of chiral edge states of opposite polarization can be reversed by applying a small direct current, which may have potential applications such as an electrically rewritable magnetic memory.\textsuperscript{106, 107}

In MATBG, the ratio of the superconducting transition temperatures to Fermi temperatures, $T_c/T_F \approx 0.1$, exceeds the range at which the weak coupling theory of superconductivity can be used, and is similar to $T_c/T_F$ in other materials (Fig. 5e) that exhibit superconductivity close to metal–insulator phase transitions. Very recently, the nematicity has been observed across the entire superconducting dome of TBG, which indicates that nematic fluctuations might play an important role in the low-temperature phases of MATBG.\textsuperscript{108} The phenomenology of MATBG is very similar to the phase-diagram of cuprate and pnictide\textsuperscript{109, 110} superconductors in which superconducting domes flank Mott-like insulating phases. Moreover, it is much easier to characterize and control the MATBG than high-temperature cuprates superconducting system. Therefore, in-depth research on MATBG is expected to help solve the high-temperature superconducting mechanism that troubles physicists for more than thirty years.

However, there are still a lot of efforts required to confirm the resemblance between the phenomenology of MATBG and high-temperature superconductors. For example, the parent
material of the high-temperature cuprates superconducting system is an anti-ferromagnetic Mott insulator. Current experiments have confirmed that the MATBG is an insulator when the flat band is half-filled, but there is no definite evidence of whether it is a Mott insulator. Moreover, it is needed to be further studied whether the MATBG has anti-ferromagnetism if Mott-like insulator states are confirmed in it. Besides, we still don’t know the superconducting pairing mechanism in the MATBG. Apart from this, it is unclear whether there is a pseudogap. On the other hand, there are many problems in the MATBG superconductivity needed to be settled. For example, it is still not clear how does the superconductivity in MATBG change along with angles near the magic angle under the same filling condition. Whether there is a smaller magic-angle in which can lead to similar superconductivity phenomena except for the first magic angle is also indefinite. In each moiré pattern of the TBG, the stacking order undergoes a change from AB stacking to BA stacking and there are topologically protected boundary states on the AB-BA stacking domain boundary, which can intrigue an interesting question that how do the superconducting and topological boundary state networks in the MATBG interact and affect each other. Nevertheless, we hold optimistic opinions about more “magic” properties in the MATBG in the foreseeable future.

3. Moiré Exciton in TMDs heterobilayer

Transition metal dichalcogenides are another kind of materials that attract people’s attention apart from graphene, because it is a natural semiconductor material with a considerable band gap. Similar to graphene, strong intralayer covalent bonding and weak interlayer vdW interactions make it possible to exfoliate the thin 2D material from its counterpart bulks. Moreover, TMDs convey unique semiconducting properties which renders them optically active. The anomalous phenomena in graphene-based moiré superlattices arouse the interest of scientists to investigate twisted bilayer TMDs superlattices. A TMDs moiré superlattice is produced when two layers with lattice mismatch are grown on top of one another, naturally endowing with nanoscale moiré pattern, which can significantly modify the electronic and optical properties of TMDs heterobilayers. In this section, we mainly present the contemporary experimental and theoretical understanding of the unique effects of a moiré superlattice on the interlayer exciton potential landscape and optical properties.

3.1 Theory of Interlayer Excitons Moiré Potential Energy

Various TMDs heterobilayers have a type II band alignment, where electrons (holes) have lower energy in different layers. The excitons composed of electrons and holes located at different layers through strong Coulomb interaction are referred to as interlayer excitons, shown as violet circle in Fig. 6a, whereas intralayer excitons are blue and red circles, in which formed by electrons and holes coming from individual layers. Fig. 6b shows excitation, charge separation and emission processes of interlayer and intralayer excitons. The photoluminescence spectra expected from the heterostructure are shown as Fig. 6c. The blue (X) and red (Y) photoluminescence peaks correspond to pristine intralayer excitons, whereas the violet (I) peaks are from interlayer excitons.
The moiré pattern in TMDs superlattices induced by the lattice mismatch or crystal orientation misalignment can cause modulations in the electronic band structure and optical property, such as a spatially dependent band gap, or twist-controlled interlayer excitons. Considering a moiré superlattices of the common chalcogen TMD bilayer WX$_2$/MoX$_2$ ($X = \text{Se, S}$) with a small twist angle $\theta$ and an in-plane displacement $d$, which has two distinct stacking orders of AA stacking (Fig. 7a) and AB stacking (Fig. 7d). This heterojunction has a small lattice mismatch (~0.1%) and the metal heteroatoms of adjacent layers overlap each other only for periodic moiré periodicities, thus the wavelength of moiré superlattice $a_M$ can be simplified as $a_M = a_0/\theta$, in which $a_0$ is the lattice constant of a monolayer TMD. $a_M$ (~18.3nm) is much larger than exciton Bohr radius (~1.3nm) when the twist angle is smaller than 1°. Therefore, the exciton energy can be approximated by following the local band gap.

In this system, the moiré potential modulated interlayer excitons energy is $E_g - E_b$, where $E_g$ is the band gap between the conduction band minimum (CBM) of MoX$_2$ and the valence band maximum (VBM) of WX$_2$ at the $\textbf{K}$ point, and $E_b$ is the exciton binding energy. The variation of $E_b$ in the moiré pattern is usually neglected, since it is typically smaller than the variation of $E_g$. According to the Ab initio continuum model, the electronic properties calculated from the untwisted reference stackings can be leveraged to approximate the twisted bilayer with a smoothly varying field over the moiré pattern, which can accurately capture atomic relaxations and electronic phenomena. The calculation procedures of excitons moiré potential energy in TMDs heterobilayers by using the Ab initio continuum model are as follows:

Firstly, calculating the Density Functional Theory (DFT) band structure of untwisted reference cells. Obtain $E_g$ as a periodic function of relative displacement $d$ at the zero-twist angle from the DFT band structure (Fig. 7b, e). The bandgap $E_g$ is defined as the energy difference between the conduction band minimum (CBM) of MoX$_2$ and the valence band maximum (VBM) of WX$_2$ at the $\textbf{K}$ point.

Secondly, knowing the band structure at different local stackings. Since the potential is a smooth periodic function of $d$ in both AA and AB stacking, it can be approximated as $\Delta(d) \equiv E_g(d) - \langle E_g \rangle \approx \sum_{j=1}^{6} V_j \exp(i \textbf{G}_j \cdot \textbf{d})$, in which $\langle E_g \rangle$ is the average of $E_g$ over $d$, $\textbf{G}_j$ is one of the
first-shell reciprocal lattice vectors (Fig. 7i), and \( V_j \) is the moiré potential parameter, denoting the coupling strength between MoX\(_2\) and WX\(_2\) in TMDs heterobilayers. Moreover, the existence of threefold rotational symmetry (\( \hat{C}_3 \)) in each TMD layer leads to \( V_1 = V_3 = V_5 \), \( V_2 = V_4 = V_6 \) (Fig. 7h). Therefore, \( \Delta(\mathbf{d}) \) is always real.

Finally, approximating the interlayer Bloch-wave coupling over a single moiré supercell. Assume that in a moiré supercell, the variation of \( E_g \) locally follows \( \Delta(\mathbf{r}) \approx \Delta[\mathbf{d}(\mathbf{r})] \approx \Sigma_{j=1}^6 V_j \exp[i\mathbf{G}_j \cdot \mathbf{d}(\mathbf{r})] \approx \Sigma_{j=1}^6 V_j \exp[i\mathbf{b}_j \cdot \mathbf{r}] \) (Fig. 7c, f), where the local configuration, \( \mathbf{d} \), is a function of space, \( \mathbf{d}(\mathbf{r}) \). \( \mathbf{b}_j = \theta \mathbf{G}_j \times \hat{z} \) is the reciprocal lattice vector of the moiré pattern (Fig. 7g).

Therefore, the moiré pattern formed in the twisted TMD heterobilayer can lead to a
position-dependent potential for excitons across a single moiré supercell, with spatial modulation up to ~150 meV for interlayer excitons in MoS$_2$/WSe$_2$ heterobilayer with rotational alignment and lattice mismatch, which has been experimentally identified by STM/STS (Fig. 8a, b).\textsuperscript{118} Additionally, TMDs are highly sensitive to crystal structure symmetry and interlayer coupling, which makes them highly susceptible to stacking orders. Atoms stacked in different configurations are known as atomic registries, which can cause valence and conduction bands to possess different energies, giving additional control over the electronic and optical properties in these systems.\textsuperscript{124-128}

![Fig. 8](image_url)

**Fig. 8** (a) STM measurement of the topographic height across a single moiré supercell of the MoS$_2$/WSe$_2$ moiré pattern. (b) A height profile along the diagonal line from AA to AA in a moiré supercell (arrow in a). (c) Illustration of the 2D K valley moiré potentials in both 3D graphs and 2D projections that can trap long-lived interlayer excitons (red and black spheres) in the local minima as they move from one location to another. 1, 2 and 3 indicate high symmetry points in a TMD heterobilayers of WS$_2$/WSe$_2$ with a twist angle of 0° (top) and 60° (bottom). Figure adapted with permission from: (a), (b) ref.\textsuperscript{118} Copyright 2017, AAAS. (c) ref.\textsuperscript{129} Copyright 2020, Springer Nature Limited.

For interlayer excitons in a moiré pattern, the theory predicts that the moiré potential generally results in localized exciton states near the potential minimum positions. **Fig. 8c** depicts that the depth of moiré potential for 0° is much larger than that for 60°. It implies that the moiré pattern leads to twist-angle-dependent periodic potential landscapes that govern the transport and relaxation of interlayer excitons, which has been experimentally confirmed.\textsuperscript{129} The existence of moiré potential in twist TMDs heterobilayers suggests a vdW heterostructure based strategy for realizing two-dimensional nanoscale arrays of uniform quantum dots, which has a great prospect on quantum photonics devices. Yu et al.\textsuperscript{40} studied interlayer excitons in MoX$_2$/WX$_2$ heterobilayer and found that they have a unique spin-dependent hopping, which can be switched off when applied a perpendicular electric field according to DFT calculation, giving the possibility to turn the superlattices into identical arrays of nanodots that act as uniform quantum emitters. Besides, the spin optical selection rules can be flipped by switch the positions of emitters in the moiré due to the electric field. These properties of moiré excitons in TMDs heterobilayers suggest the
feasibility of engineering artificial excitonic crystals for nanophotonics and quantum information applications. 130

3.2 Twist-dependent Moiré Excitons in TMDs Heterobilayers

Although the theory of exciton potential energy in twist TMDs heterobilayers has predicted notable effects of moiré potential on interlayer excitons years before, 38, 40, 119 the impacts of moiré potentials on light emission and absorption in heterobilayers of TMDs have not been detected experimentally until 2019. It was found that interlayer excitons have a valley degree of freedom, different interlayer excitons at a given valley can absorb light with different circular polarization, besides, the excitons interact with light of a specific polarization that depends on the local symmetry of the atoms arranged periodically in the heterobilayer. 41-44

Recently, the WSe2/WS2 moiré superlattice system with different twist angles was studied experimentally. Jin et al. 42 measured PL of WSe2/WS2 heterobilayer and found that there is no monolayer PL in the heterostructure, which indicates the existence of strong interlayer coupling. There emerge three peaks around the original A exciton resonance of WSe2 in the reflection spectrum in the small twisted WSe2/WS2 moiré superlattice, corresponding to distinct moiré exciton states. Whereas in superlattice with a large twist angle, it shows only a single resonance in the energy near the A exciton of WSe2 (Fig. 9a). These phenomena can be explained with a theoretical model in which the moiré potential is much stronger than the kinetic energy of exciton, generating multiple flat exciton minibands.

Seyler et al. studied individual interlayer excitons trapped in the moiré potential in MoSe2/WSe2 moiré superlattice with a small twist angle. 43 They found that there are several narrow peaks near the free interlayer exciton energy under extremely low temperatures when using low excitation power. The linewidth of peaks is about 110 µeV, making it comparable with quantum emitters. However, the excitons can delocalize with broadened linewidth emissions under high power excitation. Besides, the trapped interlayer excitons exhibit strong circular polarization of the same helicity for a given twist angle (Fig. 9b, c). The samples are excited with σ+-polarized light at 1.72 eV. The σ+ and σ− components of the photoluminescence are shown in red and blue, respectively. The PL from the heterobilayer with a twist angle of 57° is co-circularly polarized, but from the heterobilayer with a twist angle of 2° is cross-circularly polarized. This phenomenon suggests that the moiré-defined trapping sites preserve Ĉ3 symmetry. Therefore, the observation of narrow interlayer PL lines verified the existence of moiré potential with inherited valley-contrasting physics, trapping the interlayer excitons.

The experimental result of Tran et al. is the observation of multiple interlayer exciton resonances with circularly polarized emission in the same moiré superlattice system, MoSe2/WSe2 heterobilayer with a small twist angle. 41 They can locate the emission peaks of interlayer excitons and determine the extent to the energy separation between peaks dependent on twist angle in the moiré superlattice. This result provides an exciting opportunity for high-performance semiconductor lasers, quantum emitter arrays and a platform for studying exotic quantum phases.

Additionally, the research also reveals that intralayer excitons can hybridize with interlayer
excitons in MoSe₂/WS₂, owing to the conduction band delocalized over both layers, which leads to a resonant enhancement of moiré superlattice effects. The essence of hybridization between intralayer and interlayer excitons is that the conduction band edges of MoSe₂ and WS₂ are near-degeneracy (Fig. 9d), which can amplify the impact of the moiré potential on optical features of these heterobilayers. The energy of hybridized excitons relies on the interlayer twist angle because hybridized excitons are formed by holes that reside in MoSe₂ binding to a twist-dependent superposition of electron states in the adjacent monolayers, which gives an additional degree of freedom to modulate the optical properties of TMDs heterojunctions.

Fig. 9 (a) Reflection contrast spectrum of small twisted WSe₂/WS₂ heterostructure (light blue, top) compared to that of a WSe₂/WS₂ heterostructure with a large twist angle (black, bottom). Helicity-resolved PL spectra of trapped interlayer excitons of MoSe₂/WSe₂ heterobilayers with twist angles of 57° (b) and 2° (c). The insets illustrate the twist angles of the two samples. (d) The band diagram showing intralayer excitons can be hybridized with interlayer excitons in heterobilayers of MoSe₂/WS₂ (indicated in purple). Figure adapted with permission from: (a) ref. 42 Copyright 2019, Springer Nature Limited. (b), (c) ref. 43 Copyright 2019, Springer Nature Limited. (d) ref. 131 Copyright 2019, Springer Nature Limited.

The above four researches all studied the impact of the moiré superlattice on optical properties of moiré excitons in TMDs heterobilayers with different twist angles experimentally and found many novel phenomena. The origin of the observed effects is interlayer excitons trapped in a smooth moiré potential with inherited valley-contrast physics. However, these studies are only the first footstep toward complete understanding and control over individual moiré potentials. In-depth knowledge of the scale and depth of potentials is expected to control over single exciton traps,
which provides opportunities for obtaining quantum emitter arrays of versatile tunability in twisted TMDs heterostructures.

4. Conclusion and perspective

In this paper, we have mainly reviewed quantum phenomena in two-dimensional heterostructures. The electronic and optical properties of graphene-based and TMDs-based heterojunctions have been summarized, and the physical mechanism of quantum phenomena and the moiré potential in moiré superlattices have been elaborated. The moiré superlattice modulated properties in 2D materials have been studied at a remarkable level for a wide variety of materials, among graphene/h-BN, (magic-angle) twisted graphene bilayer and TMDs heterobilayers. In graphene/h-BN heterostructures, the moiré superlattice can strongly modify the Dirac spectrum and assist optical spectroscopy in the fractional quantum Hall regime, which results in superlattice minibands and leads to Hofstadter-butterfly spectra in a strong magnetic field, whereas in twisted graphene bilayers its effects are enhanced by interlayer resonant conditions, resulting in a superconductor–insulator transition at magic twist angles. Moreover, in TMDs heterobilayers, the appearance of moiré superlattice accompanied by the twist angle-dependent periodic potential landscape that can trap interlayer excitons near the potential minima, resulting in anomalous optical phenomena. Although lots of intriguing physical phenomena have been observed experimentally, exploiting the distinct advantages of angle-dependent moiré superlattice in 2D materials and their superior devices is still full of challenges. In perspective, we propose several research directions for 2D materials-based vdW-coupled twistronic materials of great importance, which are worth being discussed thoroughly in the future.

1) Precise control of twist angle. 2D materials vdW moiré heterostructures can be fabricated by using a variety of methods, including CVD epitaxial growth, monolayer folding and tear-and-stack methods. Each method has its advantages and disadvantages, but the issue of common concern is how to precisely control the twist angle, which is also the most important obstacle to immediate progress in understanding the properties of 2D twistronic materials. Uncontrolled strains related to details of how mechanical contact is established between the twisted layers can strongly influence physical properties, such as basic features of the phase diagram that expresses gapped states, the appearance or absence of superconductivity and orbital magnetism in the MATBG.

2) Physical mechanism of superconductivity in the MATBG. Two strongly correlated quantum states have been observed in the MATBG due to the enhanced interlayer coupling compared with graphene/h-BN heterostructures. However, there are some important problems remained to be solved. For example, there is no definite evidence whether the MATBG is a Mott insulator when the flat band is half-filled, moreover, it is needed to be further studied whether the MATBG has anti-ferromagnetism if Mott-like insulator state is confirmed in it. Besides, we are not yet clear of the superconducting pairing mechanism in the MATBG. The resemblance between the phenomenology of high-temperature superconductors and MATBG is striking, but whether these similarities reflect a deep connection remains to be seen.

3) Development of multi-functional twistronic devices. TMDs twistronic materials are a big family with rapid development and innovation. In twisted TMDs heterobilayers, moiré potential
generally results in localized exciton states near the potential minimum position, which suggests a vdW heterostructure based strategy for realizing 2D periodic arrays that act as tunable uniform quantum light emitters. Furthermore, the number of trapped excitons could go down to one by carefully tuning the size and depth of the moiré potentials, offering a source of single-photon emitter arrays. In addition, interlayer excitons at a given valley can absorb light with different circular polarization, which provides the possibility to fabricate high-performance semiconductor lasers and entangled photon sources as well as artificial excitonic crystals.

In conclusion, the moiré superlattice in 2D materials provides a platform of versatile tunability for studying exotic quantum phases of matter with imaging possibilities. Progress is being made in this direction, and we can expect that studies of moiré superlattices can advance twistronics physics and help us understand other condensed matter systems.

Conflict of Interest
The authors declare no conflict of interest.

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