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Correlation, superconductivity and topology in graphene moiré superlattice

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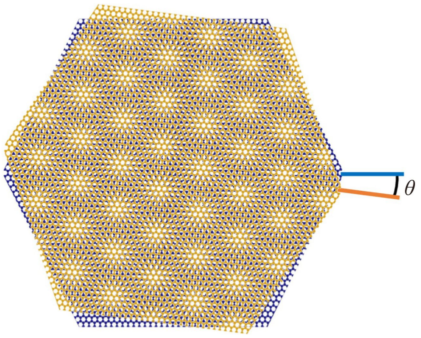
ABSTRACT

Moiré superlattices created by stacking different pieces of two-dimensional layered materials with a slight lattice mismatch have recently emerged as an exceptional platform for exploring emergent quantum phenomena. In stark contrast to the “parent” materials, the electronic band structures are significantly modified from moiré engineering due to the large-scale periodic moiré potential and interlayer hybridization. In this paper, we mainly focus on the recent progresses achieved in graphene-based moiré systems which have been a condensed-matter playground showing unprecedented abundance of quantum states such as strongly correlated states, superconductivity and novel band topologies.



Keywords twisted graphene, moiré superlattices, correlation, superconductivity, topology

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1 Introduction

Graphene-based moiré superlattice has been a rich platform to study emergent quantum phenomena due to the high tunability of its electronic structure. Band engineering arising from moiré superlattices and interface reconstructions makes it possible to create the unique flat bands which is crucial to study correlated physics. On one hand, the moiré superstructures formed from lattice mismatch can effectively fold the initial bands into a set of moiré minibands. More importantly, due to the formation of reduced mini-Brillouin zone, the first conduction/valance band is significantly lowered down in terms of energy and can be easily traversed with electrostatic gating. On the other hand, at some magic twist angles, the interlayer tunnelling between different graphene layers further flattens out the energy dispersion, resulting in flat bands which host a large density of states and amplify the interactions [1]. Twisted bilayer graphene (TBG) is the first studied flat-band graphene system both theoretically and experimentally. The TBG device can be obtained by stacking one sheet of monolayer graphene on the top of another with a rotated angle θ [Fig. 1(a)]. Periodical moiré patterns with θ dependent wavelength λ and modified unit cells [Fig. 1(c)] appear. As a result, the electronic structures of graphene can be effectively modified. Based on a continuum model, a

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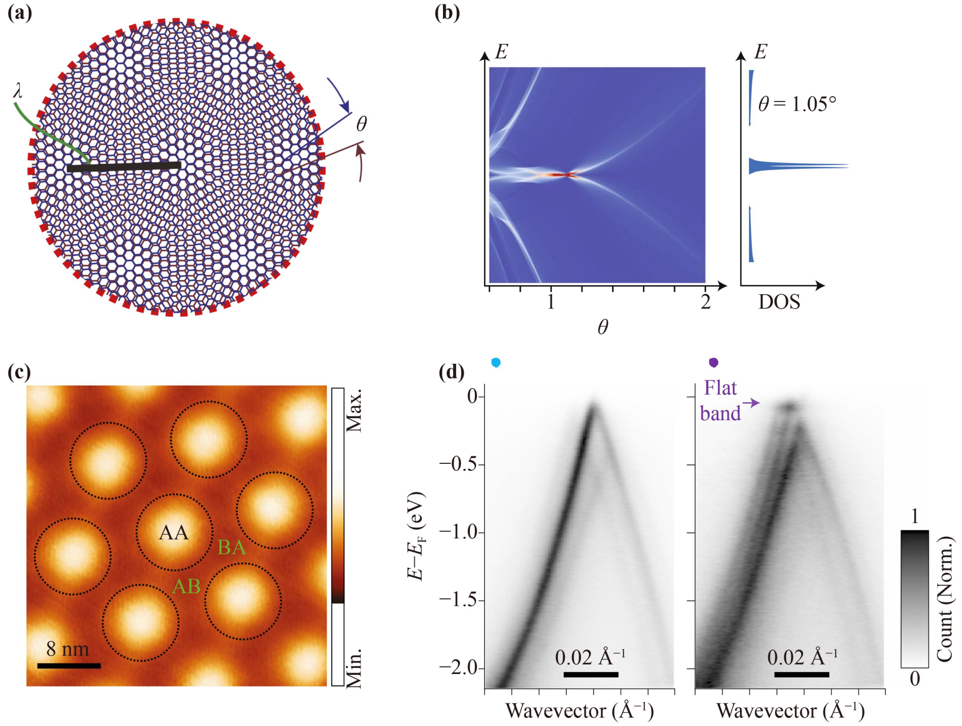

Fig. 1 **(a)** Schematic of moiré pattern formed by stacking two layers of graphene with twisted angle θ [5]. **(b)** Color plot of schematic density of states (red color denotes high value and blue color denotes low value) in TBG as a function of energy E and twist angle θ (left). Density of states as a function of energy E with $\theta = 1.05^{\circ}$ (right) [2]. **(c)** Moiré pattern of magic-angle TBG measured by scanning tunnelling microscopy (STM) image [6]. **(d)** Band structures resolved by ARPES for monolayer graphene and TBG. The monolayer graphene displays linear band dispersion with Dirac cone structure (left). By contrast, flat electronic band is formed in TBG (right) [4].

series of magic twist angles for generating low-energy flat bands were firstly predicted [1]. As shown in Fig. 1(b), pronounced flat bands are formed at $\theta \approx 1.1^{\circ}$ (the first "magic-angle") [2]. Experimentally, the existence of the flat bands in TBG has also been directly observed through angle-resolved photoemission spectroscopy (ARPES) [Fig. 1(d)] [3, 4].

2 Correlation in twisted graphene

Since Cao *et al.* [5] first reported their discovery of strong correlation marked by the half-filling (also indicated by $\nu = \pm 2$, each moiré unit cell filled with two electrons/holes) insulating states [Fig. 2(a)], strongly correlated physics has been studied extensively in twisted graphene systems. The half-filling insulating states have been attributed to a Mott-like insulator induced by the strong electron-electron interactions in the flat bands [5]. STM further demonstrated the presence of electron correlations in magic-angle TBG [6–9]. Besides the insulating states at $\nu = \pm 2$, a full sequence of integer-filling correlated states have been further observed in TBG devices with more homogeneous twist angles [10]. Temperature-dependent measurements show the $\nu = 0, \pm 2, +3$ states are thermally activated with gap values ~1 meV, while the $\nu = \pm 1, -3$ states are semi-metallic [10]. Subsequent scanning measurements of local electronic compressibility studies further revealed the symmetry breaking mechanism of the correlated phases in this system [11]. Under pressure or strain effect, correlated insulating states can also appear at a twist angle away from the magic angle [Fig. 2(b)] [12, 13], indicating high tunability with interlayer spacing and coupling strength.

In addition to magic-angle TBG, a few more twisted graphene systems showing similar correlated physics have been discovered. In ABC-stacked trilayer graphene/hexagonal boron nitride (h-BN) heterostructure, electrically tunable correlated insulating states have been found at one quarter and half fillings [Fig. 2(c)] [17]. In twisted double bilayer graphene, correlated insulating states appear at half filling $(\nu = 2)$ [Fig. 2(d)] [15, 18–22]. Interestingly, an in-plane magnetic field significantly enhanced half filling $(\nu = 2)$ state and gave birth to other integer-filling states including $\nu = 1, 3$, implying a spin-polarized ground state [15, 19, 21]. For twisted monolayer-bilayer graphene [Fig. 2(f)], the correlated states are related to the direction of electric displacement field due to the breaking of mirror symmetry [16, 23]. More theorical and experimental studies further reveal that the correlated states can also emerge at

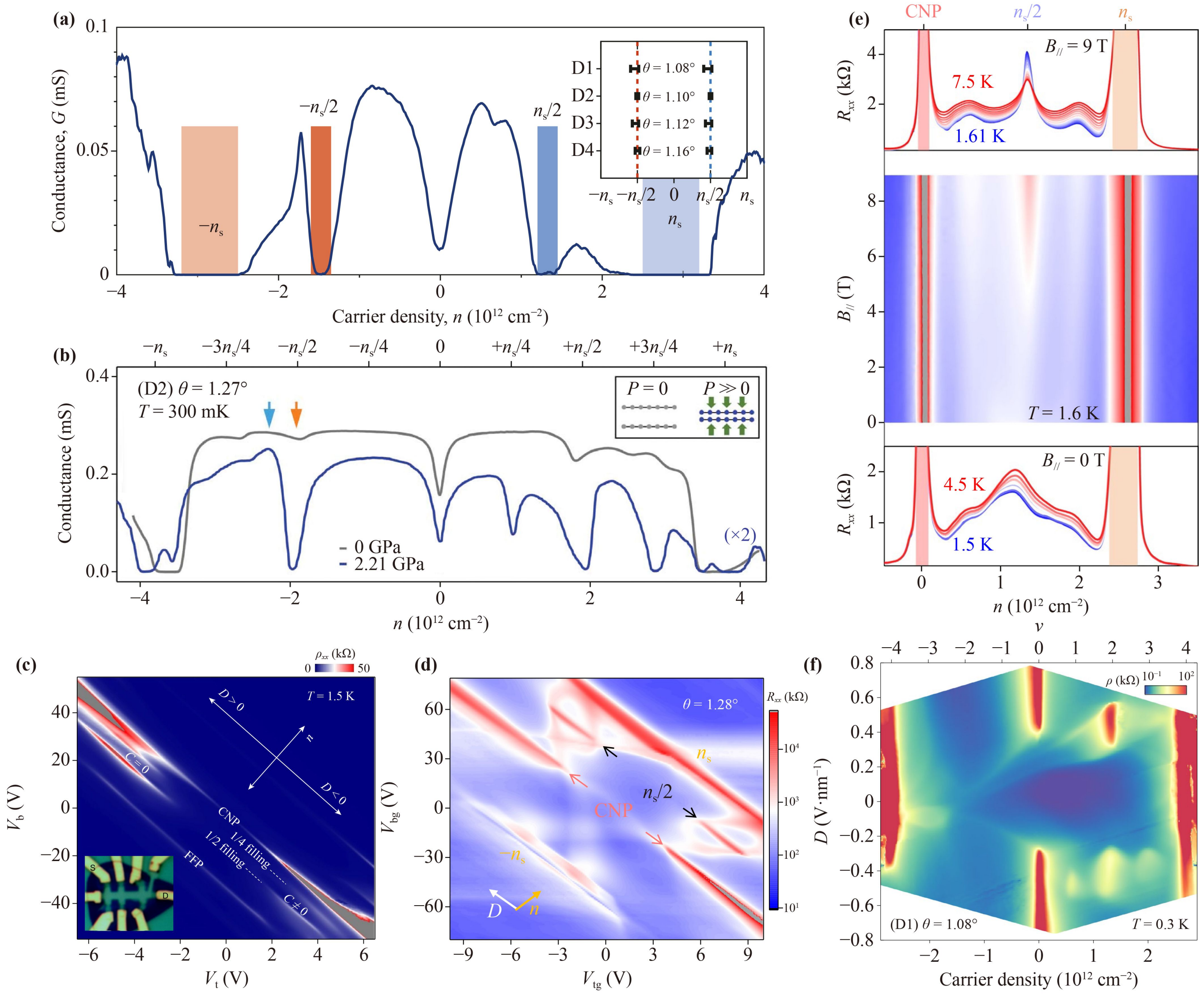

Fig. 2 **(a)** Conductance versus carrier density in magic-angle TBG at 0.3 K [5]. **(b)** Conductance versus carrier density in magic-angle under different pressure values [12]. **(c)** Color plot of the longitudinal resistivity as a function of dual gates in ABC-stacked trilayer graphene/h-BN heterostructure [14]. **(d)** Color plot of the longitudinal resistance as a function of dual gates at 4 K in twisted double bilayer graphene with $\theta = 1.28°$ [15]. **(e)** Evolution of correlated insulator under in-plane magnetic field with $D/\varepsilon_0 = -0.306\ \mathrm{V\cdot nm^{-1}}$ in twisted double bilayer graphene [15]. **(f)** Color plot of the longitudinal resistivity as a function of carrier density and displacement field in twisted monolayer-bilayer graphene with $\theta = 1.08°$ [16].

twisted multilayer graphene systems [24–27]. Moreover, some of the correlated states are rendered with a non-trivial topology which will be further discussed in the following section.

3 Superconductivity in twisted graphene

Superconductivity resided in flat bands is another inspiring discovery in twisted graphene superlattices. Cao *et al.* [28] first discovered superconductivity in magic-angle TBG, directly positioning the system as a radically new platform to study superconductivity. Specifically, a robust superconducting dome is observed slightly beyond the region of half-filling valance band ($\nu = -2$), showing a similar phase diagram with cuprate superconductors [Fig. 3(a)]. Meanwhile, the large ratio of superconducting critical temperature T_c over Fermi temperature T_F (T_c/T_F) or Bose–Einstein condensation temperature T_{BEC} (T_c/T_{BEC}), further indicates the unconventional texture of superconductivity. A few more superconducting domes all flanking symmetry-breaking correlated states have been further observed [Fig. 3(b)] [10]. Such phenomena have raised a new question about the pairing mechanism of electrons in magic-angle TBG. Additionally, the superconducting state is robust against the

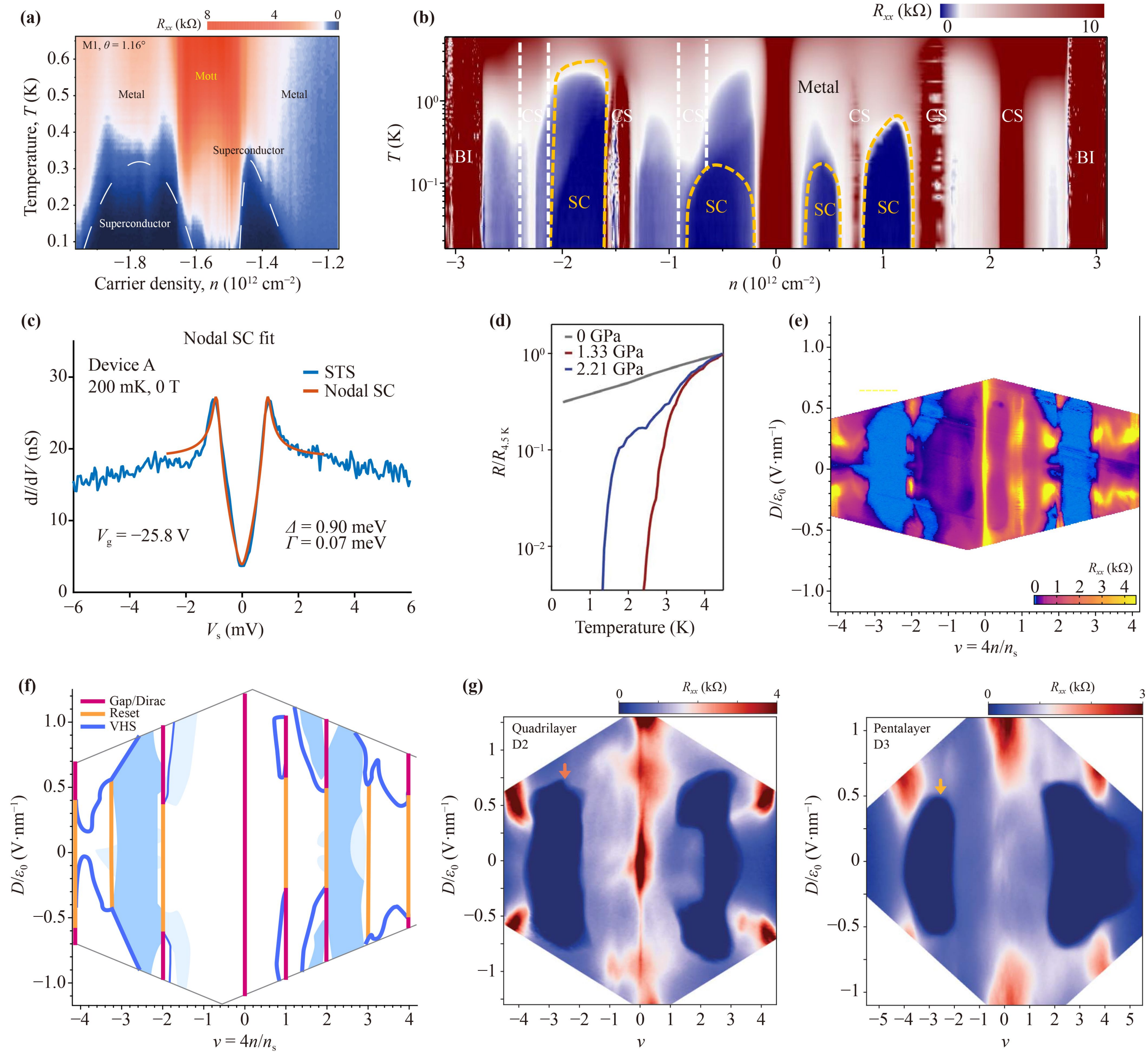

Fig. 3 **(a, b)** Superconducting phase diagram in magic-angle TBG, showing correlated state (CS) and superconducting state (SC) [10, 28]. **(c)** V-shaped tunnelling spectrum is well with the nodal fitting in TBG with $\theta = 1.13°$ [32]. **(d)** Pressure dependent superconducting state in TBG with $\theta = 1.27°$ [12]. **(e)** Color plot of longitudinal resistance against electric displacement field and filling factor in twisted trilayer graphene. Superconducting behavior observed at $2 < |\nu| < 3$ [36]. **(f)** Illustration of phase diagram in twisted trilayer graphene. VHS: Van Hove singularity [36]. **(g)** Measured superconductivity in twisted quadri-, and pentalayer graphene placed on monolayer $\mathrm{WSe_2}$ [27].

screening environment while correlated states are quenched in TBG [29–31]. The later spectroscopy study demonstrated that as observed superconducting states exhibited signatures of a nodal superconductor which is in stark contrast to conventional s-wave superconductors. [Fig. 3(c)] [32]. Analogous to correlated insulating states, the superconducting transition can be tuned under hydrostatic pressure [Fig. 3(d)] [12].

Compared to TBG, the coexistence of dispersive bands and flat bands in twisted trilayer graphene systems offer an unusual platform for studying the electric field tunable superconductivity [33–36]. At $2 < |\nu| < 3$, symmetry-broken superconducting transition was observed in twisted trilayer graphene [Fig. 3(e)]. Under high displacement fields, the superconductivity at $2 < |\nu| < 3$ is suppressed and bounded by the van Hove singularity as shown in the schematic diagram [Fig. 3(f)]. This behavior is somewhat inconsistent with conventional Bardeen–Cooper–Schrieffer theory. Strong coupling and unconventional feature of the superconductivity at

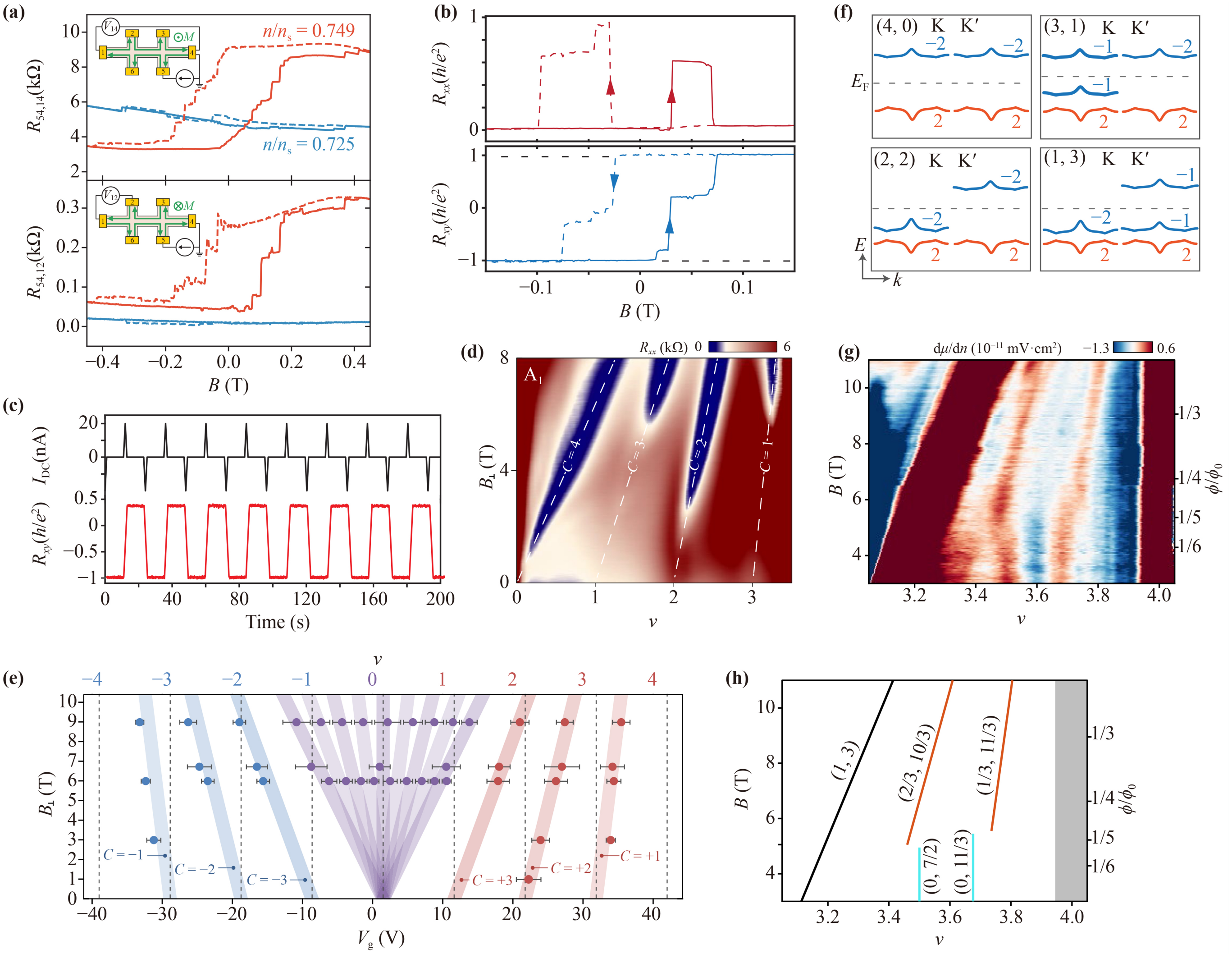

Fig. 4 **(a)** Nonlocal resistance measurements to probe the edge state, a characteristic of topological phase [39]. **(b)** Magnetic field dependent R_{xx} and R_{xy} observed in magic angle TBG aligned with h-BN at 3/4 filling [41]. Quantized R_{xy} with R_{xx} approaching to zero provides an unambiguous evidence of quantized anomalous Hall effect. **(c)** Sign reversals of quantized anomalous Hall effect under small direct currents [41]. **(d)** Correlated Chern insulators at different filling factors with non-zero magnetic field [48]. **(e)** Schematic of Chern insulators at different fillings of TBG under magnetic field measured by STM [47]. **(f)** Schematic band structures showing proposed filling arrangements of different Chern insulators. [48]. **(g)** Color plot of inverse compressibility versus magnetic field and filling factor. The corresponded Wannier diagram is present in **(h)** [51].

$2 < |\nu| < 3$ in twisted trilayer graphene was suggested [33, 36] and further verified by high-resolution scanning tunnelling microscopy and spectroscopy measurements [37]. Recently, superconducting states in twisted quadrilayer and pentalayer graphene have been experimentally observed, referring to twisted few-layer graphene systems as a magic superconducting family. [Fig. 3(g)] [27, 38].

4 Topological properties in twisted graphene

In addition to correlated and superconducting states, novel band topology in twisted graphene systems is also a rich and highly tunable source of quantum phenomena. The first experimental indication of topological feature is the presence of conducting chiral edge states in TBG. In 2019, Sharpe *et al.* [39] revealed that the correlated insulator state at three-quarter filling ($\nu = 3$) of magic angle TBG aligned with h-BN substrate presents orbital ferromagnetism with giant anomalous Hall effect. More importantly, a large and hysteretic resistance can be detected by nonlocal measurements in this ferromagnetic insulator state [Fig. 4(a)], demonstrating the existence of chiral edge states, the nature of topological phase [39], which implies an incipient Chern insulator. Similar features are also observed at $\nu = 1$ state in later work [40]. Later, the well-quantized Hall resistance $R_{xy} = h/e^2$ with the longitudinal resistance R_{xx} less than 1 kilohm near filling factor $\nu = 3$ was successfully observed at zero magnetic field [Fig. 4(b)], indicting the emergence of a Chern insulator [41]. The underlying mechanism for the Chern insulator can be understood in terms of the C_{2z} symmetry broken by aligning h-BN, leading to a nontrivial band topology with a well-defined Chern number $C = 1$ or -1 for each moiré band that breaks spin and valley degrees of freedom [42–44]. Interestingly, an extremely small direct current can switch the sign of the quantized Hall resistance. That is to say, electrical current can switch the magnetic order [Fig. 4(c)] [41], indicating giant orbital magnetoelectric effect [45]. Furthermore, the quantum anomalous Hall insulators with Chern number $C = 2$ has been observed in magic angle TBG, twisted monolayer-bilayer graphene and ABC-trilayer graphene/h-BN moiré superlattices [14, 23, 31].

Under moderate magnetic field, strongly correlated Chern insulators can emerge. Those with different Chern numbers are ascribed to interaction-driven symmetry breaking. Particularly, in TBG systems, topologically distinct states were mapped through STM [Fig. 4(e)] [46, 47]. Subsequently, these Chern insulating states were corroborated by transport measurements [48, 49]. As shown in Fig. 4(d), the Chern insulators emerge at all integer fillings ($\nu = 1$, 2, 3), and the corresponding Chern number is $\nu = 3$, 2, 1, respectively [48], which can be succinctly explained by the topological flat bands and pseudo-Landau-level picture [Fig. 4(f)] [48–50]. Besides the correlated Chern insulators, a clue for fractional Chern insulator at a filling of 3.5 was suggested by observing a van Hove singularity with magnetic field $B > 5\ \mathrm{T}$ [51]. Additionally, fractional Chern insulators have been substantiated experimentally in Bernal-stacked bilayer graphene–h-BN heterostructure and TBG [51, 52]. As opposed to the fact that fractional Chern insulator was only reported to appear at strong magnetic fields in bilayer-graphene-h-BN heterostructure [52], in TBG, such evidence showed up at weaker magnetic fields, described by magnetic-field-and-carrier-density-dependent inverse compressibility with fractional quantum numbers as shown in Figs. 4(g) and (h) [51]. The magnetic field required here has been speculated to stabilize the fractional Chern insulating states by distributing the Berry curvature uniformly across the whole Brillouin zone.

5 Conclusion and outlook

Technically, great progress has been made in recent years in fabricating quality devices with well-controlled twist angles. Different methods such as chemical vapor deposition [53], tear-and-stack [54] and cut-and-stack [55] techniques have been developed to fabricate twistronic devices. However, how to achieve more precise control over twist-angle disorder remains to be an obstacle. To reveal new exotic states and richer phase diagrams of twisted moiré devices, the control over twist angles needs to be further protected. In addition, more tunable parameters can be assessed in determining the electronic properties of twisted graphene layers. For example, the effects of carbon isotope may contribute to a different phonon-mediated interaction. An alternative substrate for twisted graphene devices to h-BN could provide a new tunable knob by changing the dielectric environment.

Although the mechanisms for the correlated insulating and superconducting states are not yet completely understood, a number of novel phenomena discovered in twisted graphene moiré systems pointed out that the moiré superlattice provides an opportunity to engineer and study the unique quantum states of condensed matter. Besides correlated insulating, superconducting and topological states themselves, recent advances have revealed many exotic behaviors related to these quantum states, such as the tunable superconducting diode effect [56, 57], nematic phase [58], bistable superconducting field-effect transistor [59]. We could expect more exciting findings such as intrinsic fractional Chern insulator to be made in the future.

Conflicts of interest There are no conflicts to declare.

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