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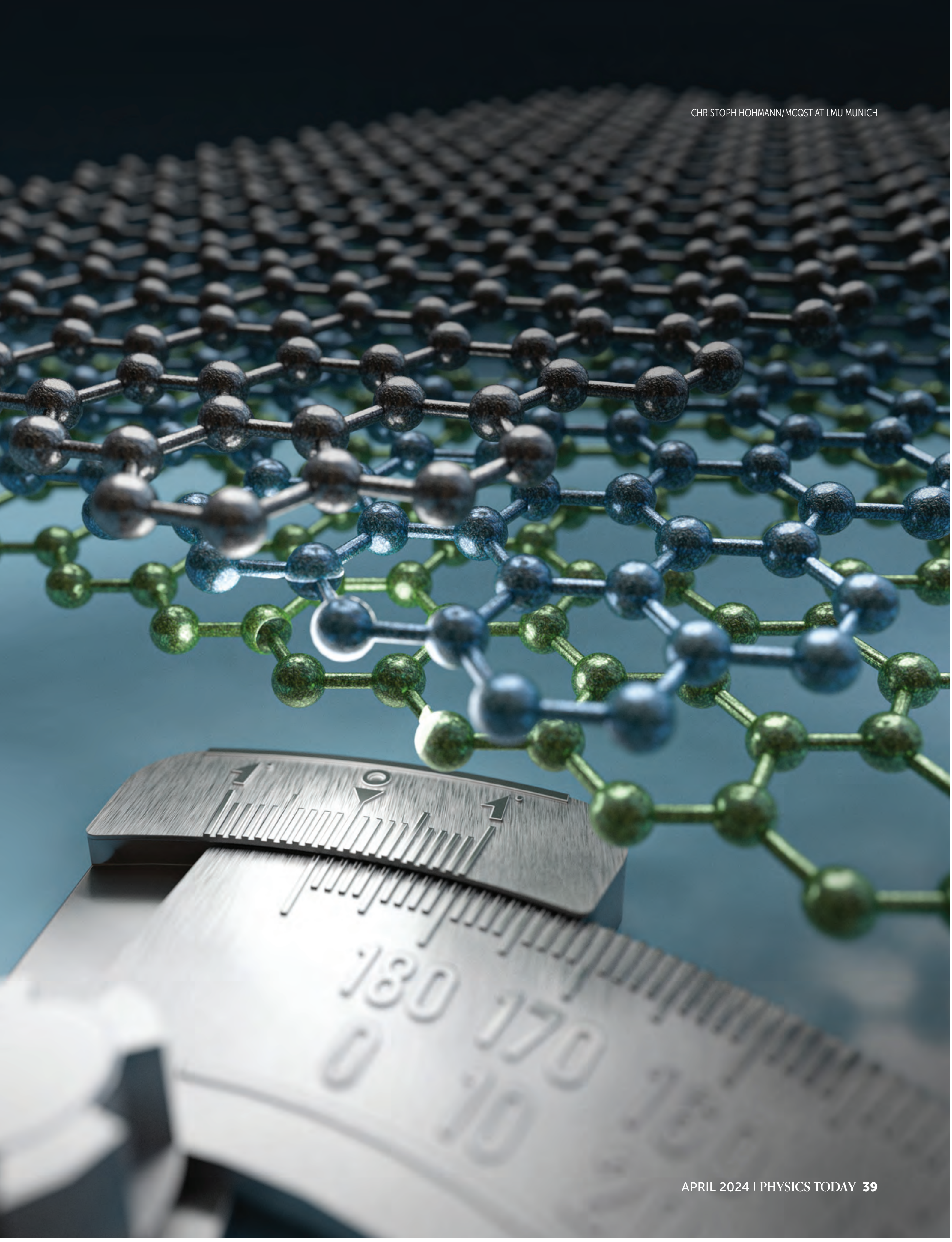
Twisted bilayer graphene's gallery of phases

B. Andrei Bernevig and Dmitri K. Efetov

The simultaneous occurrence of exotic phases, and the ability to easily tune them, has positioned magic-angle twisted bilayer graphene as one of the richest materials platforms in condensed-matter physics.

A simple twist between two sheets of carbon crystals has taken the condensed-matter-physics community by storm. The discovery of superconductivity and other phenomena in that system, announced in 2018, has revealed an array of new options to also realize interacting topology, magnetism, and other many-body states of matter, in an entirely novel and simple way. Now, six years after that initial revelation, researchers are still trying to grasp the full details of its complex phase diagram. Since the universal principles that give rise to those phases can be transferred to other 2D materials, big discoveries continue to happen almost every month.



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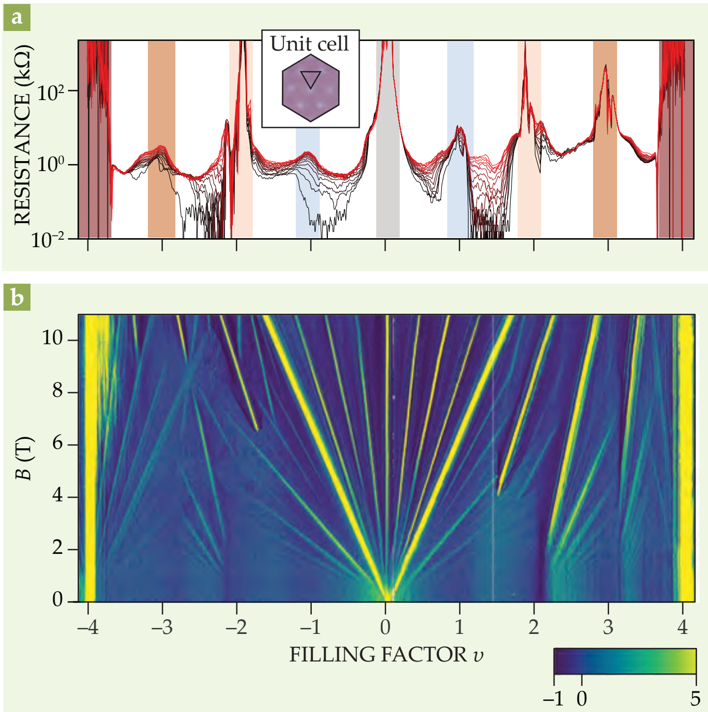


FIGURE 1. EMERGENT STATES of magic-angle twisted bilayer graphene. **(a)** Correlated insulator states, characterized by peaks in the resistance, are found at almost all integer values of the filling factor *v*. Integer filling factor *v* is the number of excess electrons (positive) or holes (negative) in each unit cell of a moiré lattice. Lines ranging from black to red correspond to increasing values of the applied magnetic field. (Adapted from ref. 4.) **(b)** The yellow streaks in this plot of inverse electron compressibility map the rich sequence of topological Chern-insulator states with quantized Hall conductance that appear when a perpendicular magnetic field *B* is applied. (Adapted from ref. 8.)

Traditionally, the electronic properties of a material are the result of its atomic composition and the arrangement of its atoms. So-called strongly correlated materials are one of the richest and yet least understood material classes. Their electrons do not behave as individual particles but are highly entangled with one another. Such compounds include, for example, heavy-fermion systems and cuprate high-temperature superconductors that exhibit a rich phase diagram governed by a mysterious interplay between magnetic and superconducting phases, in which current flows without resistance. Typically, strongly correlated materials are complex, multicomponent systems that often contain large atoms with *f*- and *d*-shell electrons. (See the article by Antoine Georges and Gabriel Kotliar on page 46.) In search of novel physical effects, scientists have been striving to synthesize crystals with evermore complex structures and compositions.

The observation of strongly correlated electrons and multiple many-body ground states in magic-angle twisted bilayer graphene (MATBG) devices shocked and excited the physics community.[1–3] The surprise lies in the fact that it was achieved by an entirely novel and previously unthinkable approach and in the unlikeliest of materials. In stark contrast to traditional strongly correlated systems, MATBG consists entirely of light and simple carbon atoms, and its building blocks, the single-layer graphene sheets, show no signs of strongly correlated electron effects. Those properties can, however, be turned on by a conceptually simple trick—stacking two graphene sheets, one on top of another, and twisting them. Just like when a combination lock is opened by turning to the right sequence of symbols, strong electronic correlations are unlocked when the twist angle between the layers is set to a well-defined value, 1.1°, the so-called magic angle. (See box 1.)

2D materials and moiré superlattice

The fabrication of MATBG was enabled by a breakthrough in the engineering of structures composed of vertically assembled 2D van der Waals materials, a large class of materials that consists of weakly coupled sheets. For example, the van der Waals material graphite, called graphene when separated out as single layer, consists of carbon atoms arranged on an atomically thin hexagonal lattice. Because of the weak coupling between the layers, it's possible not only to extract 2D layers from bulk van der Waals crystals but also to assemble them one on top of another using a simple colamination technique, thus creating vertical van der Waals heterostructures.

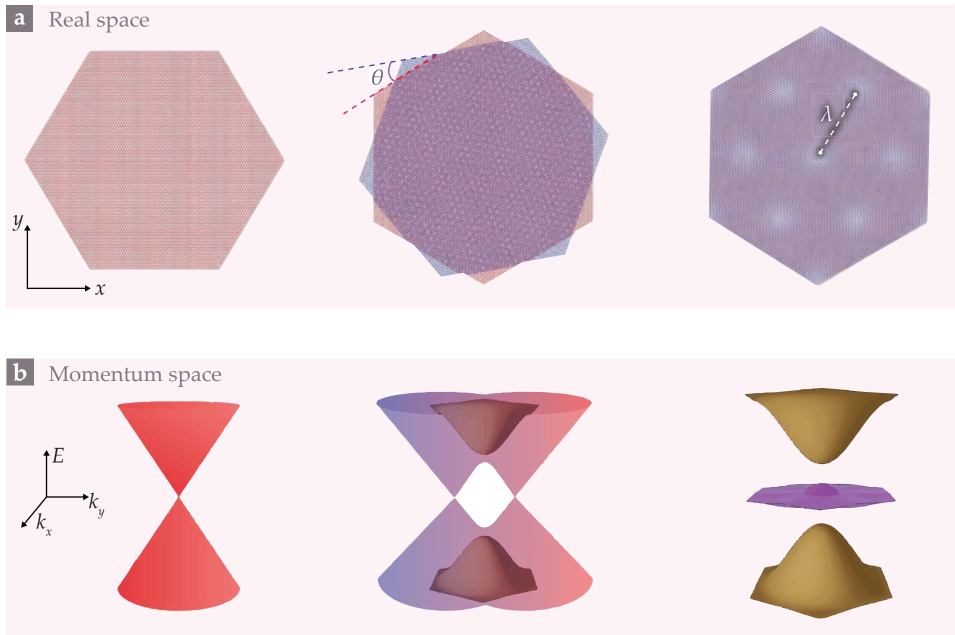
In more-traditional techniques, crystals are typically grown by epitaxy of individual atoms, and the grown layers automatically lock into the crystallographic orientation of the underlying substrate. But since van der Waals heterostructures are assembled from preformed 2D crystals, it has become possible to freely choose the crystallographic orientation between the layers during the assembly process and to set a twist angle between the crystals with a precision of a tenth of a degree. When two misaligned 2D crystals are stacked one on top of another, the individual crystal lattices produce between them a type of geometric interference known as a moiré pattern.

The moiré pattern forms a periodic, triangular superlattice on top of the original graphene lattice, which, for small twist angles, forms a unit cell with a lattice constant λ of about 10 nm, which is orders of magnitude larger than that of single-layer graphene. The enlarged unit cell in real space gives rise to a reduced unit cell in momentum space. In addition, the moiré pattern can naturally break some of the internal symmetries of the underlying 2D crystal, such as its inversion symmetry. In high-quality devices, where the electron mean free path is bigger than the lattice constant λ, the moiré superlattice can strongly affect the electronic wavefunctions and lead to a strongly altered electronic band structure.

When scientists started to theoretically investigate the effect of the moiré pattern of two twisted graphene layers, they found that for a set of well-defined twist angles—the magic angles—the effect of the electronic coupling between the layers becomes extremely strong.[1] In particular, for the first magic angle of 1.1°, the electrons slow down dramatically and condense into extremely flat energy bands, which have a bandwidth of only about 10 meV. Further, scientists have realized that those renormalizations possess all the necessary ingredients to turn on topology in the MATBG bands. In such topological bands, while the interior of the device behaves as an insulator, the edges of the device behave as a metal, forming

Box 1. What makes the magic angle magical

Graphene is a single 2D layer of hexagonally arranged carbon atoms, as shown in real space in panel a. Much of the fascination with the material arises from the fact that in special places in momentum space, the valence and conduction bands meet at one point, and the energy varies linearly with momentum. In the immediate vicinity of those points—the so-called valleys—the band structure thus has the shape of two cones, called Dirac cones, seen in panel b.



When one graphene sheet is stacked on top of another at a twist angle θ, a geometric interference pattern between the individual lattices emerges. Known as the moiré pattern, it takes the form of a triangular superlattice.

For large θ, the separation of the Dirac cones of the individual lattices in momentum space is large, and the two graphene layers are almost decoupled and only weakly affected by the moiré pattern.

When θ is small, however, the Dirac cones in the individual layers strongly overlap and begin to hybridize. For a twist angle close to the magic angle of 1.1°, the Dirac cones flatten and converge to zero energy. That results in the occurrence of ultraflat bands with a bandwidth of only about 10 meV. Because all the electrons in those bands have almost the same energy, the density of states is extremely large and peaks in the centers of the moiré lattice sites. That produces the topological flat bands, induced by the moiré superpotential, observed in magic-angle twisted bilayer graphene.

states similar to the quantum Hall effect, in which the electrons can only move along the device edges.

Tunable topological flat bands

The resulting topological flat bands are the key starting point to understand the rich phenomenology of MATBG. The electrons' lack of kinetic energy makes the electron–electron Coulomb interactions the dominant contribution to the system energy. Since all electrons in the flat bands have almost the same energy, their density of states is also extremely large, which in turn further increases the electronic interactions and favors the formation of strongly correlated phases. The combination of topology and strong electron interactions represents a long-sought blend of properties that enables the formation of entirely new electronic phases.

As a result of the strong electronic interactions, MATBG exhibits many complex quantum phases that are not present in a single layer of graphene, including correlated insulators,[2,4] superconductors,[3–5] and a so-called strange-metal phase.[6] Additionally, as described below, the inherent topological property of the flat bands gives rise to orbital magnets[4,7] and Chern-insulator states.[8,9] Incredibly, all those phases can coexist in a single device, and they can be tuned into one another by a set of external experimental knobs, a capability that is not possible in other strongly correlated systems. One such tuning knob is the voltage applied to an electrostatic-gate electrode that is placed underneath each device. It acts as a capacitor plate that can charge the MATBG sheet and allows for direct, clean, and reproducible control of the electron density in the device. (See Physics Today, January 2020, page 18.)

One emergent property of MATBG that can be directly observed through the manipulation with the electrostatic gate is a strong electron-density dependence of the resistance, as shown in figure 1a. When the moiré lattice sites are fully occupied by an integer number of electrons or holes, known as integer filling factors, and no free lattice positions remain, resistance can jump by orders of magnitude, while remaining low at electron densities that fall between those values. In the absence of an applied magnetic field, the resistance entirely vanishes in some regions, indicative of superconducting behavior.

Highly resistive regions at integer filling factors are interpreted as correlated insulators. Since the cost in Coulomb energy that each electron has to pay by tunneling to an already-occupied lattice site is much higher than its kinetic energy, the electrons localize on the moiré lattice sites and induce interaction-driven energy gaps that give rise to insulating states. Because of the Pauli exclusion principle, each energy state can be occupied by two electrons with spin-up and spin-down. In addition to the spin quantum number, electrons in MATBG have quantum numbers defined by the sublattice (×2) and valley (×2), totaling

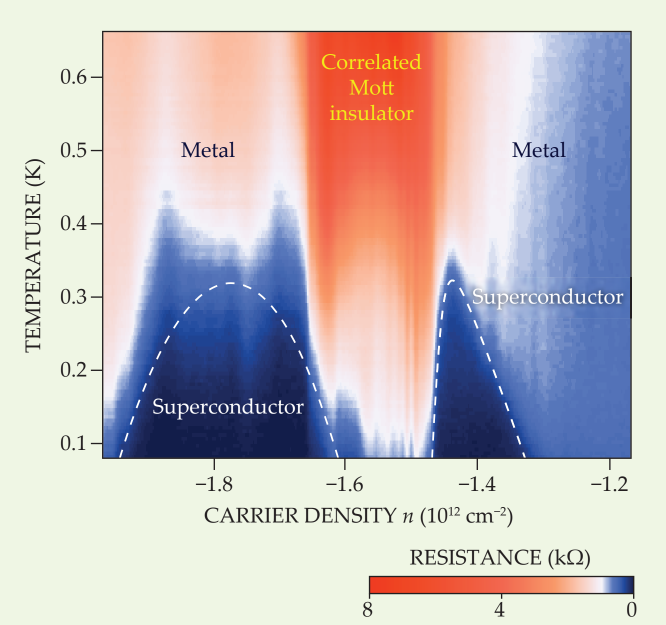


FIGURE 2. A PHASE DIAGRAM of magic-angle twisted bilayer graphene. Dome-shaped superconducting regions appear in close proximity to correlated Mott insulator states. (Adapted from ref. 3.)

an overall eight electrons per energy state. The interaction-induced energy gaps naturally polarize the energy bands with respect to those quantum numbers and produce a cascade of phase transitions, forming complex many-body ground states with broken spin, sublattice, and valley symmetries.

To understand in detail the possible low-energy many-body states in MATBG, one must also consider the inherent nontrivial topological properties of the energy bands. The abundance of low-lying topological states becomes visible when a perpendicular magnetic field B is applied. As shown in figure 1b, MATBG exhibits a robust sequence of Chern-insulator states of quantized Hall conductance with Chern numbers C of ±1, ±2, ±3, and ±4 (proportional to the slopes of the strongest yellow lines) nucleating from filling factors ν of ±3, ±2, ±1, and 0, respectively.[8] The Chern numbers indicate the number of topological edge states, each of which contributes to the conductance e^2/h, where e is the electric charge and h is Planck's constant; the sign of the Chern numbers reflects its propagation direction.

More-complex Chern insulators with fractional Chern numbers have also been discovered.[9] And at odd-integer filling factors, specifically when ν is 1 or 3, MATBG has exhibited the quantum anomalous Hall effect—quantized conductivity in the absence of a magnetic field.[7] Those zero-field Chern-insulator states have insulating magnetic bulk and metallic topological edge states that arise from circular currents rather than from spin symmetry breaking, hence they form an orbital magnet.

Superconductivity and quantum geometry

As shown in figure 2, upon slight doping away from (most of) the correlated insulators at integer filling factors in MATBG, the resistance drops to zero, and extended dome-shaped superconducting regions appear with a critical temperature T_c of roughly 0.1–0.3 K (and up to 5 K in other experiments)—and of yet-unknown mechanism.[3] Such behavior initially sparked direct comparisons to unconventional superconductors, like cuprates and heavy-fermion materials, in which superconductivity also appears upon doping localized, correlated insulating states. Such superconductors are widely believed to have a purely electronic coupling mechanism. That picture contrasts strongly with that of conventional superconductors, which are well-described by the Bardeen-Cooper-Schrieffer (BCS) theory, which states that electrons form bound pairs because of an attractive electron–phonon interaction.

One of the key attributes of the superconducting phase in MATBG is its unprecedentedly low electron density, which is only about 10^{11} cm^{-2}. Another is the very high ratio—roughly 0.1—of its critical temperature T_c to its Fermi temperature T_F, which is the Fermi energy divided by Boltzmann's constant. That ratio is comparable to only that of unconventional superconductors. The unconventional nature of the superconducting phase in MATBG is further supported by the fact that the size of the superconducting electron pairs is similar to the spacing between moiré lattice sites, which suggests that the system lies close to the crossover between a BCS-like state and a Bose–Einstein condensate.[3]

It still cannot be ruled out that MATBG superconductivity could also arise from a simple BCS mechanism, where the electron–phonon coupling could be strongly enhanced by the immense density of states in the flat bands. Initial experimental studies based on scanning tunneling microscopy[10] and thermal-conductivity measurements have spurred researchers to propose a nodal superconducting pairing symmetry: The superconducting energy gap vanishes in certain crystallographic directions. That behavior contrasts with the uniform pairing usually found in a conventional BCS-like superconductor, but could still arise from electron-phonon pairing or bands with topological properties.

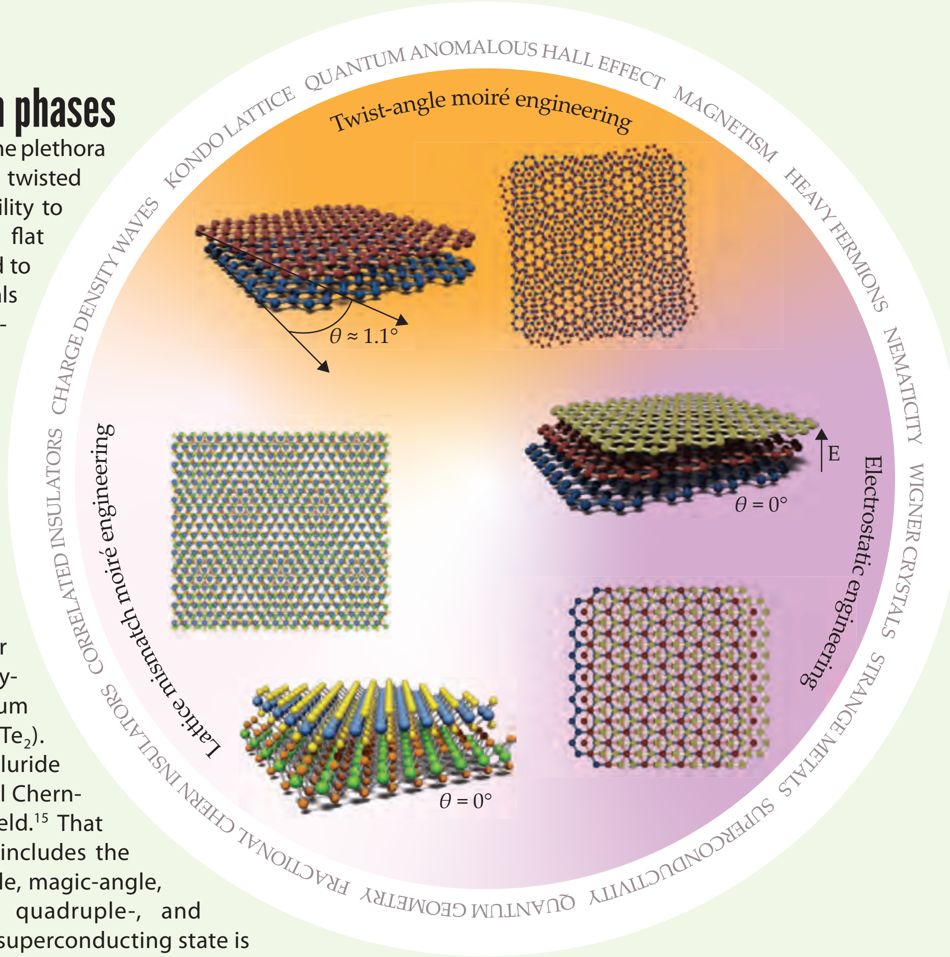
Meanwhile researchers have also recognized an entirely novel aspect of superconductivity in MATBG. Previous research has shown that superconductivity is hindered in a conventional localized flat band, but it can be made possible through the so-called quantum geometry and the topology of the band structure.[11] It arises from the overlap of the orbital wavefunctions, which facilitates movement of interacting particles even when noninteracting particles would usually be localized. The flat-band dispersion and quantum geometry together hence guarantee a lower bound for superfluid weight in MATBG even if the bands were exactly flat.

Strange metals

Another exotic phase in MATBG is the metallic phase that exists between the integer filling factors. It is markedly different than the metallic phases observed in standard metals, which are typically well described by a Fermi-liquid theory in which the electrons behave as noninteracting quasiparticles. In contrast, the metallic phase in MATBG shows features similar to the ones observed in other strongly correlated materials, such as cuprates and heavy fermions, where a strange-metal phase is observed.

The key attribute of such a phase is that its low-temperature resistivity scales linearly with temperature, which is in stark contrast to the quadratic dependence observed in normal met-

Box 2. Emergent quantum phases



The main principle that gives rise to the plethora of quantum phases in magic-angle twisted bilayer graphene (MATBG) is the ability to engineer topologically nontrivial flat bands. That ability can be transferred to a much larger set of van der Waals materials, which has led to the discovery of an even bigger multitude of exotic ground states. Graphene-based systems, however, remain the only ones to show a robust superconducting phase.

Similar to MATBG, twist-angle engineering can be used to create moiré-induced flat bands in various twisted layers of sheet-like materials, such as monolayer-on-bilayer graphene, bilayer-on-bilayer graphene, tungsten diselenide bilayers (t-WSe_2/WSe_2), and molybdenum ditelluride bilayers (t-$MoTe_2/MoTe_2$). Twisted bilayer molybdenum ditelluride has been shown to have a fractional Chern-insulator state in zero magnetic field.[15] That class of twist-angle materials also includes the alternating multilayers—for example, magic-angle, mirror-symmetrical twisted tri-, quadruple-, and quintuple-layer graphene—whose superconducting state is believed to have a spin-triplet pairing.

It is also possible to create moiré flat bands without any twisting of the layers. One such way is by stacking and aligning layers of different materials, which produces a moiré lattice because of the mismatch between the different crystal species. Those stacked systems include graphene sheets on hexagonal boron nitride substrates, bilayers of WSe_2/WSe_2, and $MoTe_2/WSe_2$, in which a Kondo lattice state was observed that shows highly localized and magnetically ordered heavy fermions on the moiré lattice sites,[16] similar to MATBG.

Van der Waals engineering further allows alteration of materials properties by, for example, inducing strong spin–orbit coupling through the introduction of heavy element layers, such as WSe_2.[17] It is also possible to introduce close-by metallic layers, which can be used as electrostatic gates and as screening layers,[5] which reduce the Coulomb interaction. Additionally, it is possible to tune the band structure by applying strong out-of-plane electric fields. In some moiré-free stacked bi- and trilayer graphene sheets, that dramatically flattens the bands and induces strong interactions and superconductivity.[18] The absence of the moiré superpotential and critical temperatures that are two orders of magnitude lower than MATBG, however, could mean that the nature of those states is distinct from its moiré counterparts.

als. Electrons in the strange-metal phase also have an extremely high electron-scattering rate, with an upper limit dictated by the Heisenberg uncertainty principle. Such a strange-metal phase is often found close to a quantum critical point in the vicinity of magnetically driven zero-temperature phase transitions (see the article by Subir Sachdev and Bernhard Keimer, Physics Today, February 2011, page 29).

In MATBG, the characteristic Planckian-limited linear-in-temperature resistivity was found down to the extremely low temperature of 40 mK and over a range of filling factors.[3,6,12] At elevated temperatures, a linear-in-temperature resistivity would not be too surprising, as one is also found in noncorrelated single-layer graphene devices above 10 K, where it is a result of conventional electron–phonon interactions.

The persistence of the linear scaling down to millikelvin temperatures in MATBG, however, makes a similar scenario quite unlikely, as phonons at such low temperatures are effectively frozen out. That behavior points rather to the existence of a quantum critical point and the prevalence of strong electron–electron interactions. An understanding of how electrons interact with each other in the strange-metal phase can likely shed light on the origin of all the other low-temperature phases, because the strange-metal phase acts as their "parent phase." In particular, the superconducting phase directly nucleates from the strange-metal phase.

A heavy-fermion picture

The striking similarities between the phenomenology of MATBG and other strongly correlated systems have found a solid theoretical foundation, and the physics of the MATBG

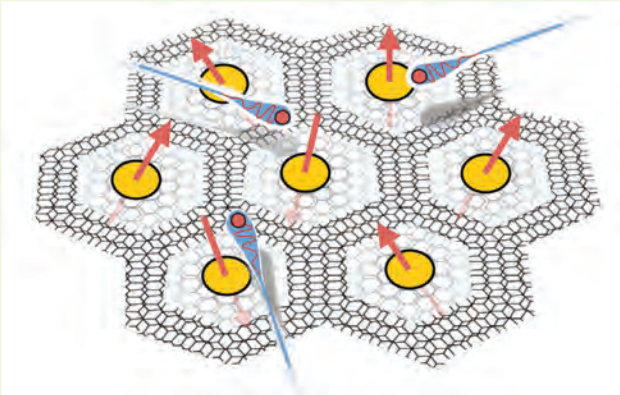


FIGURE 3. IN A TOPOLOGICAL HEAVY-FERMION MODEL of magic-angle twisted bilayer graphene, electrons can have light or heavy effective masses. Heavy fermions (yellow dots) remain localized in the moiré lattice, while light fermions (red dots with blue tails) move freely though the crystal lattice. (From ref. 14.)

low-energy manifold of states can be understood through several approaches. Two initial complementary strategies have been proposed to provide a theoretical basis for understanding the strongly correlated and topological MATBG bands. One is to construct extended Hubbard models in which the symmetries are represented nonlocally in real space. The other is to adopt a full momentum-space formalism, which hides the local nature of the interactions. Neither of the approaches, however, could easily explain the presence of two carrier types with local and itinerant moments.

Recently it has become possible to write down a fully symmetric model with a simple real-space picture that remarkably and elegantly explains the ground states of MATBG and their topologies.[13] The interacting MATBG can be reformulated as an effective topological heavy-fermion system consisting of local f orbitals and delocalized topological conduction bands (c). As illustrated in figure 3, the f electrons are so localized that they have a kinetic energy of only about 0.1 meV, but they have a strong on-site Coulomb repulsion, computed to be about 60 meV. The c electrons, however, carry the symmetry anomaly and have unbounded kinetic energies. The actual flat bands emerge from a hybridization between the f and c bands, and several types of interactions also couple the f and c electrons. With that understanding, scientists can explain the coexistence of quantum-dot-like behavior and superconductivity: They come from two different types of carriers (f and c).

Challenges and opportunities

The simultaneous occurrence of all those phases and the ability to modulate between them by simply applying a voltage have positioned MATBG as one of the richest and most tunable materials platforms in condensed-matter physics. MATBG also established the novel concept of twisting of materials as a simple but extremely powerful technique to dramatically alter materials properties and to induce strong electron correlations and topology in a large variety of systems. (For more on the engineering of other van der Waals materials, see box 2.) Those innovations have been achieved in a highly controllable, albeit detail-sensitive, system environment, and currently the community is struggling to grasp the full details of the MATBG's colorful phase diagram.

The complex quantum phases of MATBG create both an unprecedented opportunity to crack the mysteries of strong-coupling superconductivity and unprecedented challenges in understanding the myriad phases of the system. A plethora of different possible ground states arise from the combination of broken-symmetry phases and crystallographic symmetry operations, intertwined with nuances in twist angle, dielectric environment, strain, disorder, and other material properties. The details of the experimental findings finely depend on the competition between all the different orders that are closely adjacent to each other, which results in different phase diagrams for different samples. The ultimate challenge is to find a unified formalism that both explains all the possible phases and points out universal features of the problem.

Meanwhile, each year researchers discover more and more novel 2D flat-band platforms with seemingly similar many-body phases. Among those discoveries have been the transition-metal-dichalcogenide moirés and the crystalline nonmoiré graphene multilayers. The many material platforms enabled by van der Waals materials and their degree of controllability allow for significant progress in the coming years.

The intriguing link between MATBG and heavy-fermion physics offers a bridge between two formerly disjointed communities. The machinery of heavy fermions—including dynamical mean-field theory—can now be applied to the physics of MATBG. Van der Waals moiré materials also provide an avenue for controllable tests of heavy-fermion physics and possibly their quantum critical points, in ways that the doping of 3D crystals cannot. The interplay between topology, interaction, heavy fermions, and superconductivity is a fundamental strength of MATBG research, which promises to provide an exceptional range of discoveries in the future.

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