The marvels of moiré materials

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Moiré systems formed by 2D atomic layers have widely tunable electrical and optical properties and host exotic, strongly correlated and topological phenomena, including superconductivity, correlated insulator states and orbital magnetism. In this Viewpoint, researchers studying different aspects of moiré materials discuss the most exciting directions in this rapidly expanding field.

Microscopy of moiré materials

Eva Y. Andrei. 2D atomic layers have changed the way we think about materials. Because 2D layers have all their atoms at the surface, it is possible to transform their electronic properties without changing their chemical composition. One of the simplest methods to do so, superposing two or more 2D crystals with a relative twist between the layers, has opened a new research frontier, that of moiré physics.

Serendipity played a key role in the field of 2D materials, from its inception by scotch-tape exfoliation to the observation of exotic physics in a moiré system. Although moiré patterns have been known to exist on the surface of graphite since the invention of the scanning tunnelling microscope (STM), the discovery that they can alter the electronic properties of a material was serendipitous. In 2009, while studying a graphene sample grown by chemical vapour deposition (CVD), we realized that, instead of the expected structure of single-layer graphene, the STM image revealed large-period moiré patterns typical of twisted bilayers and the presence of a wide range of twist angles. Despite our plan to study a single-layer sample, we decided to check how the moiré pattern affected the electronic properties of the material. To our astonishment, the scanning tunnelling spectra featured two Van Hove singularity peaks - indicative of two saddle points in the electronic band structure - whose separation increased linearly with the twist angle. This observation was intriguing because new phases of matter can arise near Van Hove singularities, thus, the possibility to tune such singularities by changing the

twist angle can be used to engineer material properties. In particular, at a twist angle of $\sim 1.1^\circ$, later termed the magic angle, we observed that the two peaks merged to form a flat energy band. Flat bands are interesting because they can host various correlated electron phases, including superconductivity and magnetism.

Many theoretical proposals were put forward for the realization of novel correlated electron phases in moiré systems, but their implementation had to wait for the invention of the tear-and-stack fabrication technique. This technique enables precise control of the twist angle and led to the discovery of superconductivity in magic-angle twisted bilayer graphene (MATBG). MATBG, intriguingly, has a phase diagram similar to that of hightemperature superconductors. Because a STM probes the sample locally, it provides insight into the microscopic origin of this phase diagram. So far, STM measurements have highlighted the importance of broken symmetries underlying the electronic correlations in MATBG. They also revealed that twist-angle inhomogeneity is inherent to the tear-and-stack technique. For the field to progress, this inhomogeneity must be overcome. This might be achieved by learning how to control the twist angle in CVD-grown materials. Another option is to introduce strain or buckling in a monolayer system, for example, through substrate-induced periodic potentials. As we have shown for graphene, this generates a local pseudo-magnetic field that can result in the formation of flat bands.

More generally, there are many opportunities in moiré physics beyond MATBG. Both smaller and larger twist angles are expected to produce new correlated states and topological phases whose exploration has barely begun. Finally, combining the vast array of 2D van der Waals crystals with the flexibility of moiré physics opens a huge swath of possibilities for engineering heterostructures with novel electronic, magnetic and topological properties.

Understanding superconductivity in MATBG

Dmitri K. Efetov. Understanding the microscopic mechanisms underlying the superconducting phase in MATBG is one of the major frontiers in moiré materials research. Superconductivity in MATBG exhibits several unusual attributes. It arises in a 2D system and originates from an electron ensemble with a density several orders of magnitude lower than that of any other known superconductor. Nonetheless, its superconducting coupling strength is extremely high, with a critical temperature that is exceedingly large compared with its ultra-low Fermi temperature, which is only comparable to high-temperature superconductors. Most strikingly, superconductivity in MATBG exists in the presence of strong electronic correlations, which initially motivated comparisons with other strongly correlated superconductors, such as the cuprates, pnictides and heavy fermion compounds. This similarity led to the conjecture that superconductivity in MATBG is assisted by electron correlations; however, recent work could not exclude a more conventional electronphonon-mediated mechanism. Owing to the recent discovery of topologically nontrivial bands in MATBGs, the possibility that the superconductivity is also topological gained formidable attention. Experimental evidence is still scarce, with only a handful of experiments performed on superconducting MATBG; thus, the exact origin of superconductivity remains elusive.

The study of superconductivity in MATBG is facilitated by the fact that this system offers several novel control knobs, allowing us to tune its electronic band structure, interaction energies and symmetries. MATBG can be integrated with other 2D materials into heterostructures

with interlayer alignments as small as a fraction of a degree. The kinetic energy of the electrons in MATBG can be tuned by slightly changing its twist angle and their interaction energy by controlling its dielectric environment, in particular, using metallic screening layers. Owing to the ultra-low density of its electron system, the flat bands of MATBG can be completely filled and emptied by electrostatic gating, and junctions can be effortlessly created. Exploiting these different control knobs will be a major avenue for future research, and work in this direction is slowly beginning.

A challenge in the study of superconductivity in MATBG is that many experimental techniques used for bulk systems are not readily applicable to this 2D system with ultra-low density. For example, due to the low electron density, the Meissner effect may be too weak to sense with state-of-the-art magnetic sensors; the study of the magnetic penetration depth is hindered by the fact that MATBG is thinner than its expected penetration depth; specific heat and thermal transport techniques cannot be applied, owing to the lack of proper nanoscale thermometers; inelastic neutron scattering is challenging, owing to the tiny cross section of the 2D samples; and the resolution of angle-resolved photoemission may be too coarse to resolve superconducting gaps smaller than a few meV.

• Pushing the moiré quantum matter frontier

Pablo Jarillo-Herrero. The past three years have seen an explosion of theoretical and experimental research on correlated moiré systems. Triggered by the discovery of

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Dmitri K. Efetov joined Institut de Ciencies Fotoniques in Barcelona as a Professor and Group Leader in 2017. His group investigates moiré materials with electronic transport and optoelectronic techniques, and is credited for the development of the highest-quality magic-angle twisted bilayer graphene devices demonstrated so far.

Pablo Jarillo-Herrero is currently Cecil and Ida Green Professor of Physics at Massachusetts Institute of Technology and is the recipient of the American Physical Society 2020 Oliver E. Buckley Condensed Matter Physics Prize and the 2020 Wolf Prize in Physics for the discovery of correlations and superconductivity in magic-angle graphene. His research interests lie in the area of experimental condensed matter physics, in particular, quantum electronic transport and optoelectronics in novel 2D materials, with special emphasis on investigating their superconducting, magnetic and topological properties.

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Kin Fai Mak is an Associate Professor of Physics and of Applied and Engineering Physics at Cornell University. His research group uses optical and electrical probes to explore condensed matter phenomena in atomically thin materials and their heterostructures.

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Ali Yazdani is the Class of 1909 Professor of Physics and the Director of the Princeton Center for Complex Materials. He specializes in the development and the application of high-resolution microscopy and spectroscopy to quantum materials. His group has uncovered a wide range of novel correlated and topological phenomena, including in moiré systems.

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correlated insulator states and superconductivity in MATBG, subsequent experiments quickly uncovered a plethora of novel phenomena, such as orbital magnetism, quantum anomalous Hall states, cascades of phase transitions, Chern insulators, unconventional ferroelectricity and even entropy-driven electronic liquid-to-solid transitions. It is reasonable to expect that several other typical condensed matter phases, from multiferroics to quantum spin liquids, might, one day, be realized in moiré materials. Moreover, several new moiré systems have appeared, including double twisted bilayer graphene, twisted monolayer-bilayer graphene and twisted transition metal dichalcogenides (TMDs), leading to the emergence of the field of moiré quantum matter (BOX 1).

A key characteristic of moiré quantum materials is the presence of tunable flat bands. Indeed, the high degree of tunability in moiré systems is one of the aspects that make them so exciting. Perhaps the most obvious tuning knob available is the twist angle, which changes the ratio between interaction strength and electronic bandwidth (that is, kinetic energy). Unfortunately, right now, the twist angle is set during nanofabrication, with no possibility to tune it in situ during lowtemperature experiments. Being able to tune the twist angle in situ would allow the direct exploration of correlated physics, as the ratio of bandwidth over interaction energy is continuously changed, possibly enabling the exploration, in a single experiment, of the weak, intermediate and strong coupling regimes.

Another, largely unexplored, knob in moiré heterostructures is strain. Strain can play a similar role to the twist angle and is easier to tune in situ. Moreover, in principle, twisted 2D materials allow the application of tunable heterostrain, that is, of a different strain (both in magnitude and in direction) to each of the two sheets forming the heterostructure. Heterostrain is very hard to obtain in bulk materials or heterostructures fabricated by molecular beam epitaxy. This knob would allow us to tune the bandwidth, the bands degeneracy and their topological character in twisted moiré systems, and, hence, the relative importance of correlations and the character of the associated topological phases.

Another promising direction is to push beyond pair combinations of layers. Interesting theoretical work suggests that going towards three or four layers and beyond would give rise

Box 1 | Moiré materials and phenomena

Moiré materials can be made of two layers of a same material stacked with a twist angle (homobilayers), of two layers of different materials with a lattice mismatch and, potentially, a twist angle (heterobilayers) or of multiple layers of twisted 2D materials (multilayers). The constituent 2D materials employed so far include graphene, hexagonal boron nitride (hBN), transition metal dichalcogenides (TMDs) and various 2D magnets and superconductors. Many exotic phenomena have been observed or are predicted to emerge in these moiré materials. Superconductivity, with its phase diagram reminiscent of that of high-temperature superconductors, was the focus of most initial studies, but many other interesting states of matter arising in moiré materials are starting to attract researchers' attention.

- Wigner crystal states: electronic crystals driven by strong Coulomb repulsions.
- Chern insulator states: states of matter that exhibit the quantum Hall effect at zero magnetic field.
- Fractional Chern insulator states: states of matter that exhibit the fractional quantum Hall effect at zero magnetic field and support excitations that obey fractional statistics.
- Mott insulator states: states of matter that are expected to be metallic according to band theory but behave as insulators, owing to strong electron correlations.
- Stripe phases: electronic crystals and electronic liquid crystals that spontaneously break rotational symmetry.
- Chiral spin liquids: spin liquids that break time-reversal symmetry and may exhibit chiral edge states.

Materials



Plus Wigner crystal states, Chern insulator states, simulation of Hubbard physics and other models, moiré excitons...

to novel tunable electronic structures, possibly leading to higher-temperature superconductivity, paving the way for a new generation of moiré quantum matter. The very recent experimental realization of ultra-strong coupling and highly tunable superconductivity in magic-angle twisted trilayer graphene has ushered the race for moiré magic 3.0 and beyond. • For their own sake and for the greater good

Allan H. MacDonald. When 2D crystals with a small difference in lattice constant or orientation are overlaid, they form a moiré pattern. When the starting crystals are semiconductors or semimetals, the combined system is a moiré material, that is, a material in which low-energy electronic

states, as well as excitons and other collective bosonic excitations, are described by a Hamiltonian with the periodicity of the moiré pattern. The vision that moiré materials can provide a platform to study fundamental condensed matter physics has been realized over the past few years with a delightful series of discoveries, including the identification of superconductors, orbital ferromagnets, Mott insulators, Chern insulators, Wigner crystals and charge density wave states. The dizzying pace of recent progress suggests that there is much more to come.

The physics of moiré materials is the same as that of real atomic-scale crystals, in that they share discrete lattice translational symmetry, which implies that single-particle Hamiltonian eigenstates carry current. Sometimes, the similarity is deeper. For example, moiré p-type TMD heterobilayers map to one-band Hubbard models. However, moiré materials and atomic crystal Hubbard model systems differ in that the attractive potentials at lattice sites in moiré materials have finite depth, not the unbounded depth of Coulomb attraction to ion cores. This difference is significant and is being actively explored. Nevertheless, as material quality improves, it will be possible to map out the phase diagrams of moiré Hubbard materials and gain new understanding. For example, I expect an experimental answer to a deep question that occupies much research on high-temperature superconductivity: are doped Mott insulators superconductors? We should know soon whether the answer is always, never or sometimes and — in the case of the more nuanced answer perhaps get a hint of the secret sauce of cuprate superconductivity.

In addition to simulating atomic crystals, moiré materials raise new fundamental questions. For example, MATBG is captivating because it combines non-trivial momentum space topology with strong interactions in a way that has yet to be realized in atomic-scale crystals. The topology of MATBG derives from the Dirac cones of isolated graphene layers and their honeycomb lattices. Some TMD homobilayer moiré materials map to honeycomb lattices and open up unexplored possibilities to study the strong correlation physics of topologically non-trivial crystals.

The energy scale of moiré materials varies from sub-meV to about 100 meV, depending on the phenomena of interest, the system and the moiré twist angle. Larger moiré energy scales tend to be

associated with weaker correlations but still allow for collective electronic states. My great hope is that high-energy moiré material physics will, in some cases, lead to robust room-temperature interaction phenomena — for example, itinerant electron ferromagnetism or strong electro-optical responses — that can be engineered by adjusting the twist angle. We would then have a new paradigm for band structure engineering with happy implications for both physics and technology.

IMD moiré superlattices

Kin Fai Mak. Moiré superlattices in TMD bilayers exhibit unique features complementary to those of graphene moiré systems. First, depending on the TMDs involved, both non-topological and topological moiré flat bands can be obtained. Systems with non-topological bands, in particular, allow us to explore the strong correlation physics of lattice models with unprecedented control. Second, the effects of Coulomb interactions in TMD bilayers are generally stronger than in graphene moiré systems, as exemplified by the emergence of Wigner crystal states at fractional fillings. Third, unlike in magic-angle graphene, in TMD bilayers, moiré flat bands that support strong correlation physics can form for a wide range of twist angles, making these systems rather insensitive to twist angles and twist-angle disorder (especially for TMD heterobilayers). Finally, the strong lightmatter interaction and the valley-dependent optical selection rule in TMDs allow optical probing of their electronic and magnetic properties.

Although studies on TMD moiré systems are in their infancy, recent experiments have revealed novel phenomena driven by strong Coulomb interactions. These include the appearance of the Mott insulating state, Wigner crystal states, stripe phases, antiferromagnetism and pseudospin ferromagnetism. Many exotic states of matter remain to be uncovered: for example, TMDs could host fractional Chern insulating states. In addition, TMDs provide a tunable platform to simulate Hubbard model physics on a triangular or a honeycomb lattice and may answer the fundamental question of whether unconventional superconductivity can emerge in a Hubbard model system. TMD homobilayer systems that support multi-orbital Hubbard physics remain largely unexplored and are, thus, another promising research direction. Furthermore, achieving continuous tuning of the

interaction strength will allow us to study the Mott transition with unprecedented control and to realize chiral spin liquids.

Last but not least, TMD moiré systems are promising quantum simulators for other important models in condensed matter physics. For example, by trapping an interlayer exciton gas, the Bose–Hubbard model and, possibly, supersolid phases can be realized. Coupling two moiré bands with different bandwidths could enable the realization of Kondo lattice physics.

With opportunities come challenges. The formation of ohmic contacts and reliable four-point resistance measurements are difficult but crucial for the success of these proposals. We also need new experimental probes that can access the charge, spin, valley and collective excitations of moiré systems to understand the underlying physics and to uncover new phases of matter.

Strong correlations meet band topology

T. Senthil. What I find most fascinating about moiré graphene systems is that they provide a tunable experimental platform on which both strong electronic correlations and band topology are often present. Moiré graphene forces theorists to confront the question of the description of strong correlations in a system with partially filled topological bands. The band topology severely complicates the use of the standard theoretical framework of lattice models of interacting electrons typically employed to describe correlated materials. Some guidance can be found in quantum Hall phenomena in a Landau level (which can be regarded as a special kind of topological Chern band with uniform Berry curvature), but moiré systems differ from traditional quantum Hall systems in that, first, they are time-reversal invariant at the microscopic level; second, their bandwidth is non-zero and, in some cases, tunable; and, third, the band topology can be more complex than that of a Landau level. Moreover, when the bands have a non-zero Chern number, the Berry curvature is not uniform.

So, these systems pose a new kind of theoretical problem for which we have no pre-existing theoretical framework, forcing us to think afresh. A key feature of strong correlation physics arises in the insulating states at total integer filling. Unlike conventional solids (but similarly to a Landau level), a correlated insulator in a topological band is likely to have ferromagnetic ordering of its flavour (spin or valley degrees of freedom for moiré graphene). The experimental observation of ferromagnetism in some moiré materials, and its possible theoretical explanation in terms of orbital ferromagnetism in bands with a non-zero valley Chern number is, thus, encouraging. It gives us confidence in proposing flavour-ordered ferromagnetic states as candidates for the correlated insulating states in other moiré materials with topological bands.

So far, I believe we have only seen the tip of the iceberg, and there will be many more surprising discoveries in moiré graphene. I am excited by the opportunity to study questions such as how a quantum anomalous Hall orbital ferromagnet evolves into a paramagnetic Fermi liquid metal as the bandwidth is tuned; whether there is a fractional quantum anomalous Hall effect at a neighbouring filling; and what we will learn from realizing such a state in moiré graphene. What are the implications, if any, of the topology of the band structure on the superconductivity that occurs in some moiré materials? Finally, moiré materials that do not have topological bands provide the opportunity to study transport near a correlation-driven metal-insulator transition more closely than has ever been possible.

I Beyond the single moiré

Emanuel Tutuc. MATBG has been the playground for a remarkable set of discoveries, with the observation of superconducting, correlated insulator and ferromagnetic states, to name a few. With research on MATBG entering a phase in which its properties are probed simultaneously by different techniques, such as transport, thermodynamic and scanning probe measurements, what other vistas can be opened in the moiré world? Mindful that the answer is, inevitably, subjective, I describe two particularly promising avenues.

The first avenue is the investigation of systems beyond the single moiré. Most studies have focused on single moiré patterns, initially in graphene on hexagonal boron nitride and subsequently in twisted bilayer graphene. Here, the moiré patterns are realized by either controlling the relative twist between two adjacent layers (in homobilayers) or by lattice mismatch (in heterobilayers). The next frontier in the moiré landscape involves heterostructures in which more than one angle between layers is controlled. An example is twisted trilayer graphene,

which has aligned top and bottom layers and the middle layer twisted by a small angle. The twisted trilayer graphene band structure has a larger magic angle ($\sim 1.5^{\circ}$) than MATBG (~1.1°) at which flat bands coexist with Dirac cones, which is expected to increase the ordering temperature for both superconducting and magnetic phases. Indeed, two very recent studies show robust superconductivity in this system. A second type of interesting heterostructure beyond the single moiré is the double moiré pattern, consisting of two moiré patterns in close proximity and possibly separated by a tunnel barrier. By combining two layers of MATBG, one can expect the stabilization of paired states at fractional and complementary moiré band filling factors, for example, $\frac{1}{2} + \frac{1}{2}$ or $\frac{1}{2} + \frac{3}{4}$. Such heterostructures also provide a pathway to stabilize indirect equilibrium exciton condensates between particles and vacancies in the two moiré patterns.

The second avenue is using 2D materials other than graphene. Moiré patterns are beginning to be explored in TMD bilayers with small twist angles, and correlated insulator phases were recently observed in twisted WSe₂ bilayers. Although TMDs have a lower carrier mobility than graphene in the starting crystal, and lattice relaxation is expected to play a role at large twist angles, flat bands are present in twisted TMDs across a range of twist angles, often with topologically non-trivial, gate-tunable gaps.

As with any endeavour, progress will depend on advances in sample fabrication techniques and high-quality crystals. Protocols will have to be devised to control multiple angles in one heterostructure and to avoid individual lattice relaxation in increasingly complex fabrication process flows.

Visualizing correlations and topology

Ali Yazdani. The discovery of moiré materials, like the discovery of hightemperature superconducting cuprates three decades ago, has shifted the landscape of condensed matter physics. The advent of cuprates laid bare our inadequate theoretical understanding of many-body quantum physics, while it experimentally challenged us to tease out the signatures of electronic correlations in very complex and often disordered materials.

To understand the cuprates, powerful new ideas on electronic correlations and a new generation of experimental techniques were developed. Techniques such as high-resolution angle-resolved photoemission spectroscopy and scanning tunnelling microscopy and spectroscopy became so advanced that, when topological insulators were discovered a decade ago, their application readily revealed their beautiful single-particle physics. The discovery of moiré materials is now driving quantum condensed matter physics in a similar fashion. In moiré materials, theorists are confronted with a new platform on which to understand strong correlations and challenged to determine the interplay between correlations and topology. From the experimental point of view, with all the physics happening not in bulk samples but in atomically thin films that can be doped through gating — basically, transistor-like devices — there are new opportunities for probing, but also for controlling, correlations and topology.

The large periodicity of moiré superlattices allows us to fully control the electronic occupation of their correlated flat bands and to tune their properties in situ with a gate. Integrating this tunability with the power of high-resolution scanning tunnelling microscopy and spectroscopy enables the study of strong correlations in ways that are impossible for other materials. Already, such experiments on twisted MATBG have revealed that, when the gate is far away from the MATBG, the strength of electron-electron interactions exceeds the bandwidth of the system. We now know that, in MATBG, strong correlations are not only at work in creating the insulating states but are also present in the normal state before superconductivity arises, as is the case for other correlated superconductors. An unexpected recent discovery is that these interactions can drive the formation of new Chern insulators, providing a rare example of correlated topological quantum states. In situ spectroscopy of MATBG moiré devices was critical to establish the role of interactions in the formation of such novel states and to distinguish them from non-interacting topological phases.

Moiré systems are a game changer. They not only host many novel quantum states but provide the opportunity to study these phases with high energy and spatial resolution in a pristine chemical setting, in which we can tune the interactions among the electrons. Undoubtedly, we still need to improve sample fabrication techniques. However, it is clear that each moiré system has its own advantages for studying a particular flavour of electronic phenomena, and we are just starting down the path of exploring them with increasing sophistication as we develop our ability to look inside these moiré devices.

The (orbital) magnetic pull of moiré materials

Andrea F. Young. When the strength of interparticle interactions exceeds the width of an isolated energy band, the electronic ground states are determined primarily by such interactions. In a 2D system subjected to a magnetic field, for example, the bandwidth of the Landau levels vanishes (that is, the bands become flat) in the absence of disorder, and interactions dominate the behaviour of the system, leading to a panoply of correlated states at partial band filling. The non-trivial topology of the Landau bands further enriches the phase diagram, so that, in addition to symmetry-breaking ferromagnets, Wigner crystals and charge density waves, interactions favour topologically ordered ground states characterized by fractional charge and statistics. The low electron density makes the entire density-dependent phase diagram experimentally accessible via electrostatic gating, enabling precise comparisons of experiment and theory.

Moiré patterns, which arise from the interference between misaligned or mismatched crystal lattices, provide an alternate route to flat-band engineering. One example is the Hofstadter butterfly, an intricate fractal structure of topological bands that arises from the interplay between the periodic electrostatic potential of a moiré superlattice and a magnetic field. When a Hofstadter band is sufficiently flat, new ground states arise from the interplay of interactions, topology and the symmetry of the moiré superlattice. These include Chern insulators that spontaneously break superlattice symmetries, as well as fractional Chern insulators that bind quantized fractional charges to the superlattice sites.

The realization of flat bands at zero magnetic field in twisted bilayer graphene and rhombohedral trilayer graphene has opened the possibility of realizing ground states with unusual magnetic properties. Crucially, the absence of a magnetic field that violates time-reversal symmetry allows for ferromagnetic states in which this symmetry breaks spontaneously. It is remarkable that moiré ferromagnetism is stabilized not by spin-orbit interactions but, instead, by the anisotropy of the 2D electron orbitals. This fact contrasts with 2,500 years of magnetic materials. Already, orbital ferromagnetism combined with the topology of the moiré bands has revealed quantized anomalous

Hall effects at record high temperatures, and the ingredients are available for the potential realization of other exotic Chern insulators. Orbital magnets allow for new ways of controlling the magnetization, including magnetic switching actuated by minuscule currents and by electrostatic gating. These novel effects ultimately arise from the fact that the microscopic magnetization does not arise from the electron spin but from the band wavefunctions, which can be directly designed and controlled. Exploration of orbital magnetism is still in its infancy, but this paradigm seems poised to impact magnetic devices for quantum and classical information science in moiré systems and beyond.

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