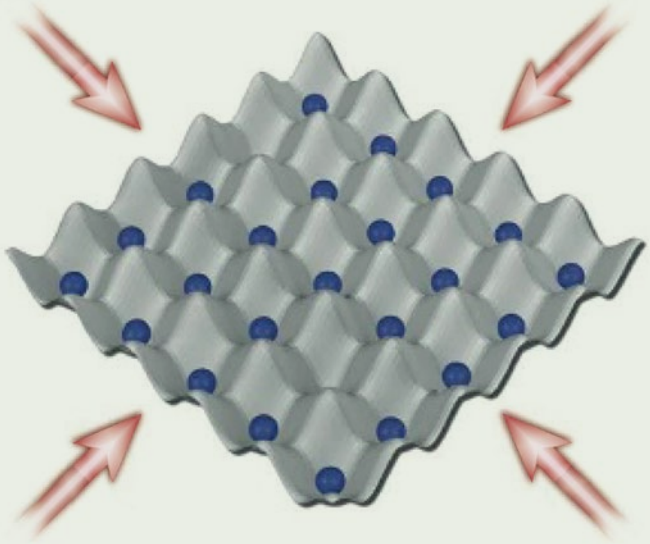
Magic Angle
A New Twist on

by Pablo Jarillo-Herrero
and Senthil Todadri

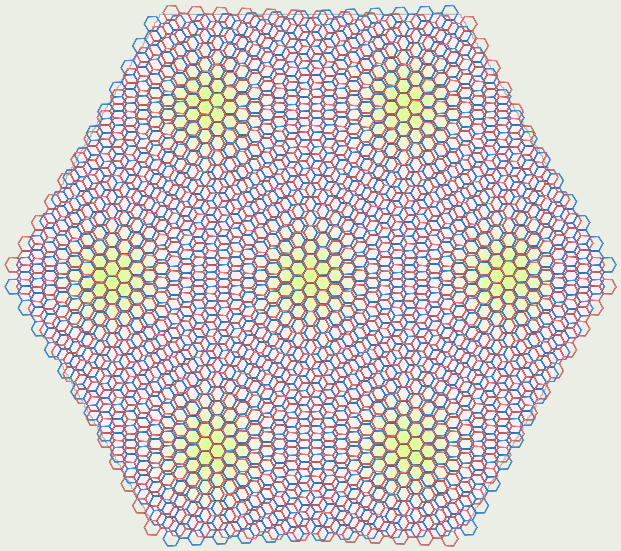
Graphene
Quantum Materials

The development of quantum mechanics was undoubtedly one of the largest scientific triumphs of the 20th century. Between the 1920s and 1940s, a precise mathematical formalism was developed that enabled extremely accurate descriptions of the quantum mechanical behavior of a number of diverse physical systems, ranging from the spectrum of the hydrogen atom to the basic electronic properties of semiconductors. This fundamental science success resulted in what one might call the first quantum engineering revolution—lasers, silicon transistors, magnetic resonance imaging and solar cells, to name a few.

COLD ATOMS OPTICAL LATTICES
Length scale ~ 1 micron
Temperature scale ~ 0.1-1 nanoKelvin



MAGIC-ANGLE GRAPHENE
Superlattice length ~ 10nm
Temperature scale ~ 1-10 Kelvin

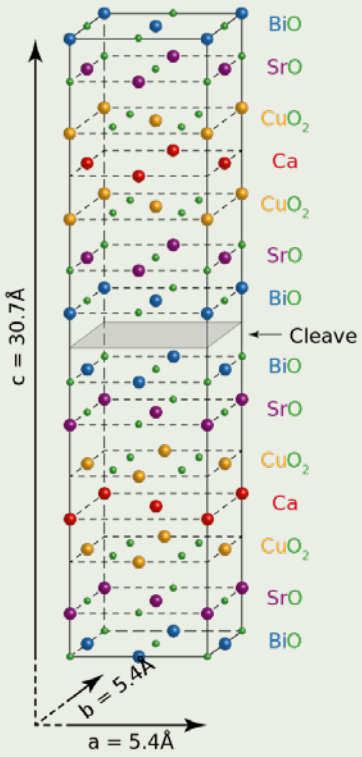


GIVEN THIS ENORMOUS SUCCESS, it may come as a surprise that significant challenges remain in dealing with the quantum mechanics of systems with a large number of particles. If these particles interact only *weakly* with each other, the physics can be described simply and often accurately. The difficulties arise in situations where a system consists of many particles that interact *strongly* with each other. In those cases, the standard mathematical methods used in quantum mechanics (developed for weak interactions) fail miserably. How important is this? It turns out that among the most fascinating states of matter are those where the interactions between the basic constituent particles are strong. Examples of such strongly interacting quantum systems abound in many areas of physics: in nuclear and particle physics (in describing properties of atomic nuclei and their fundamental structure in terms of quarks and gluons or in describing the quark-gluon plasma thought to be present in the early universe); in astrophysics (in understanding the interior of neutron stars); and in condensed matter physics (in understanding exotic phenomena such as high temperature superconductivity).

Strong correlations in quantum materials

Quantum mechanics plays an essential role in understanding the properties of macroscopic solids; thus, they are referred to as QUANTUM MATERIALS. For the electronic

QUANTUM MATERIALS
Lattice scale ~ few Å
Temperature scale ~ 100-1,000 Kelvin



$Bi_2Sr_2Ca_nCu_{n+1}O_{2n+6-d}$

FIGURE 1

Experimental platforms *to investigate strongly correlated quantum materials. [Credit: left: I. Bloch,* Nature *453, 1016 (2008).]*

properties of solids, the most important players are the electrons. In many simple quantum materials the electrons may be viewed as a gas of weakly interacting particles, and their behavior can then be easily described. However, in the last few decades a large class of quantum materials has been discovered wherein the motions of different electrons are tightly correlated with each other. The physics of such materials involves a delicate interplay between the kinetic energy of the electrons, their Coulomb repulsion [1], the crystalline lattice on which the electrons move, and their Fermi-Dirac statistics [2].

Strongly correlated quantum materials often show dramatic phenomena. A striking example is high-temperature superconductivity, first discovered more than 30 years ago. Apart from the high value (~100 Kelvin) of the superconducting transition temperature, various phenomena in these materials shocked physicists when first discovered. The superconductivity occurred in a then unprecedented place: by adding or removing charge carriers to a magnetic insulator. Such insulators were well known to be driven by strong inter-electron Coulomb repulsion acting in concert with the crystalline lattice. Adding charge carriers to an insulator typically leads to a poor conductor, yet these materials transformed into a superconductor! The theoretical description of high-temperature superconductors constitutes one of the grand challenges in contemporary physics. Currently, there is no known method to reliably solve even the simplest model believed to contain the essential physics responsible for their extraordinary behavior. This challenge, deeply connected to the more general problem of strongly correlated states of matter, is an active area of research at MIT and elsewhere.

The difficulties posed by strongly correlated quantum materials are not limited to their theoretical description. As most of such materials are compounds with complicated heavy elements containing impurities, their experimental study also poses a significant challenge. This led physicists to develop alternative platforms to try to simulate their behavior (*Figure 1*). An interesting, modern approach consists

of using lasers to create perfect artificial periodic lattices, where one can trap atoms. Atomic physicists can control the interactions between the atoms very precisely, and create "tunable artificial quantum materials" that they can directly study—a "quantum simulator." This field has made remarkable progress over the past 20 years, and recently a few groups, including our MIT and Harvard colleagues at the Center for Ultracold Atoms [3], have succeeded in getting a glimpse of some of the exotic behavior exhibited by some correlated materials. However, to reach the most sought-after states of correlated matter, such as unconventional superfluidity, atomic physicists need to cool down their lattices to a small fraction of a nanoKelvin(!)—an order of magnitude below the current state of the art—and the difficulties to reach them, while not fundamental, are technically complex.

2-D materials LEGOLAND® and the rise of twistronics

In the midst of all these decades-long difficulties, a totally unexpected discovery happened last year as a result of a new materials paradigm, and it is rapidly becoming a game-changer in our understanding of correlated quantum materials. The protagonist could not be anything else but the wonder material GRAPHENE, albeit with a new "twist."

Graphene is a one-atom thick sheet of graphite (the material in pencil lead), where the carbon atoms form a honeycomb-shaped lattice. Over the past 15 years, physicists have been studying the electronic properties of graphene with tremendous fascination because electrons propagate through graphene like ultra-relativistic massless particles, making graphene the world's best electrical conductor. Graphene was the first example of a monolayer two-dimensional crystal, *i.e.,* materials that can be obtained from mechanical exfoliation of a three-dimensional bulk layered crystal. You can think of a 3-D graphite sample as a stack of 2-D LEGO® pieces, with graphene being a single 2-D LEGO® piece that we are able to extract out of the stack. Different 3-D layered crystals are like different colored LEGO® stacks, and scientists have learned to make completely new material heterostructures by stacking different 2-D monolayer crystals on top of each other (following the analogy, by stacking LEGO® pieces of different colors). These new heterostructures, which can have properties quite different from those of the constituent monolayers, have been the subject of intense research during the past decade—a field sometimes referred to as 2-D Materials LEGOLAND®.

While this LEGOLAND® aspect of 2-D materials is fascinating, their most unique aspect is one that goes far beyond any analogy with LEGO® pieces, and that has no precedent in the history of materials science: two 2-D monolayer crystals can be stacked on top of each other with an arbitrary angle of rotation, or *twist* angle, between the crystalline lattices. This cannot be done with LEGO® pieces, as they have to be perfectly aligned to stack. This also cannot be done when growing bulk crystals, as equilibrium thermodynamics dictates the preferred atomic arrangement in a crystalline lattice, leading to a precise stacking. Even often-used non-equilibrium growth methods do not allow arbitrary angles of rotation between

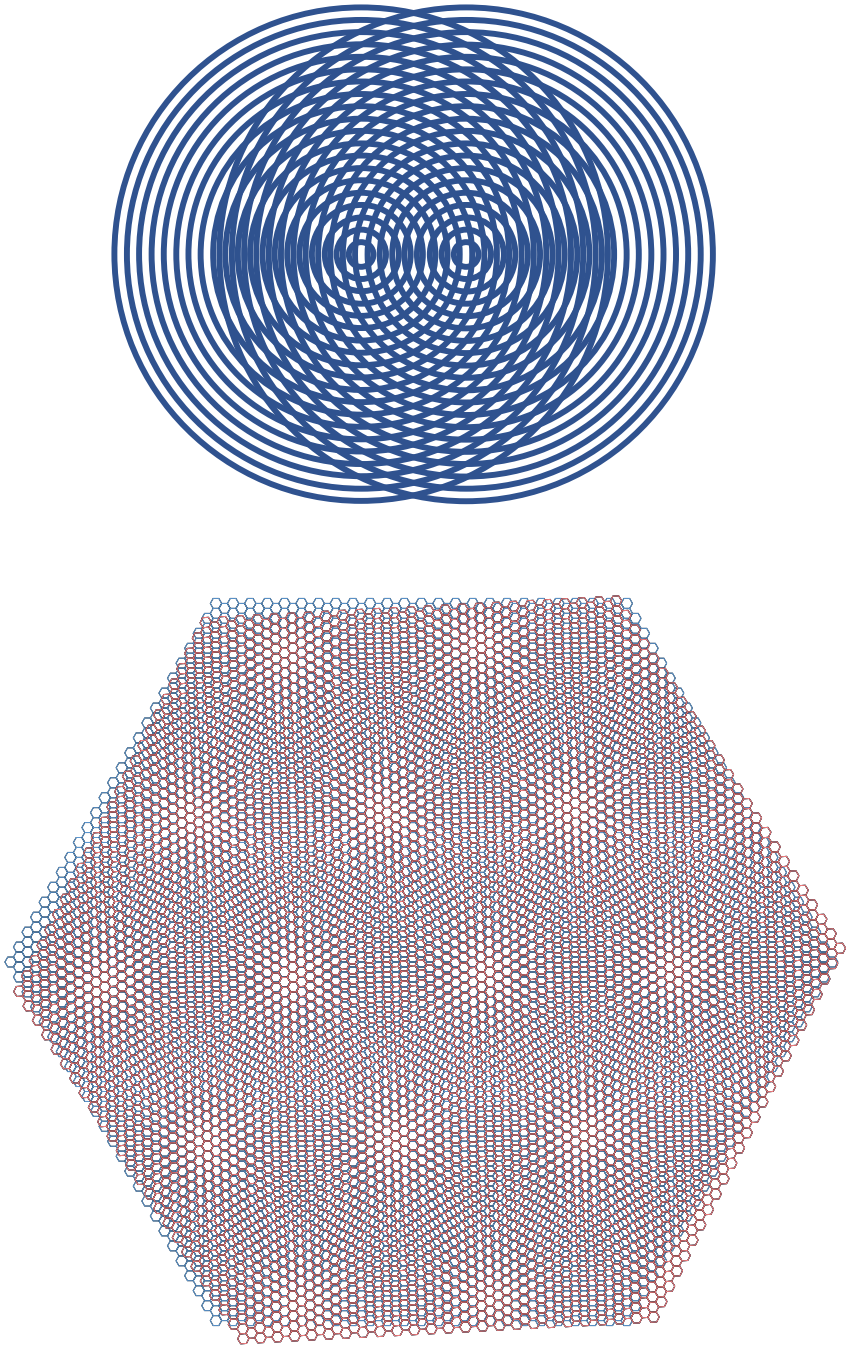


FIGURE 2

Moiré patterns *created by superimposing two sets of concentric rings (top) or by twisting two graphene sheets to create twisted bilayer graphene (bottom).*

different layers in the heterostructure. It is only with the advent of 2-D materials that scientists have been able to make such twisted heterostructures. Amazingly, the introduction of such a twist can change the electronic and optical properties in a dramatic way. This new field of physics and materials science is sometimes called TWISTRONICS.

The first question one may ask is: What happens when you place two periodic structures, such as two crystalline monolayers, on top of each other with a relative twist angle between them? The answer is that a moiré pattern forms (*Figure 2*).

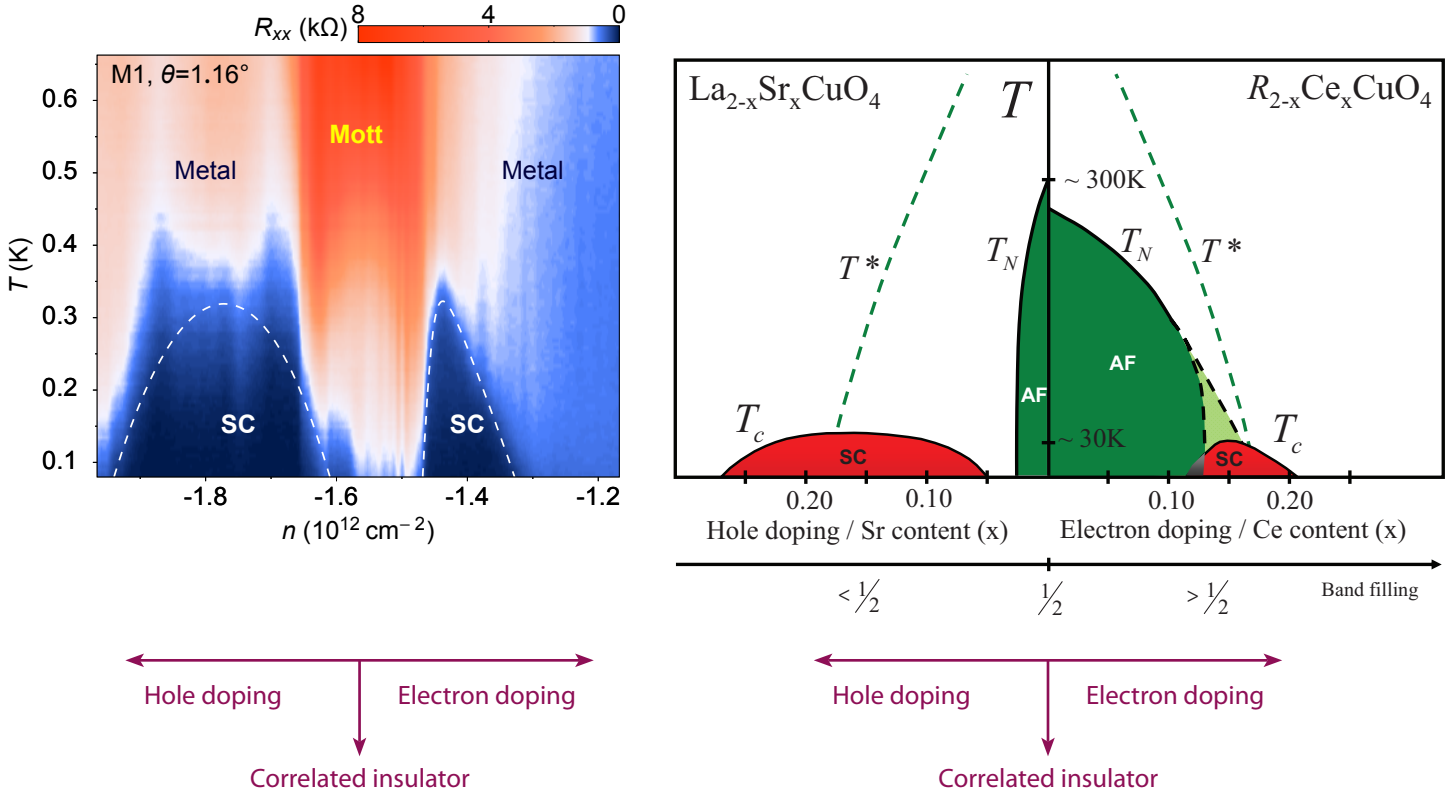


FIGURE 3

Phase diagram *of magic angle graphene and high temperature superconductors. In both figure panels, the horizontal axis refers to charge density,* n *(left) or doping (right). The vertical axis is temperature. SC refers to superconductivity (dark blue region in left, red regions in right). Mott (left, center) refers to Mott insulator, a type of correlated insulator. AF (right, center) refers to antiferromagnetic behavior, happening in the correlated insulator region. [Credit: Left, ref [7]; right, N.P. Armitage et al. Rev. Mod. Phys. 82, 2421 (2010).]*

Moiré patterns were originally conceptualized in silk textiles and appear in many areas of mathematics and physics, such as when two waves of slightly different frequency interfere and make a beating pattern. In 2-D, a moiré pattern forms even if two identical lattices are superimposed but rotated relative to each other. Physicists have been investigating rotated graphene lattices for the past ten years, but it was only last year that the full potential of twisted bilayer graphene was realized, and it required some "magic."

Magic angle graphene: the twist and shout of quantum materials

In 2010-2011, two theory groups [4, 5], predicted that if two graphene layers were stacked on top of each other, with a relative twist angle of about 1°, the velocity of the electrons—known as the Fermi velocity—would decrease to almost zero. They named this angle the "magic angle." Near-zero Fermi velocity implies that the kinetic energy of the electrons is very small. The theorists themselves realized back then that, under these circumstances, the Coulomb interaction among the electrons would likely acquire a disproportionate role in the electronic behavior of MAGIC ANGLE GRAPHENE, but they did not dare to predict what exactly would happen. This theoretical prediction, although quite unusual, did not gain much

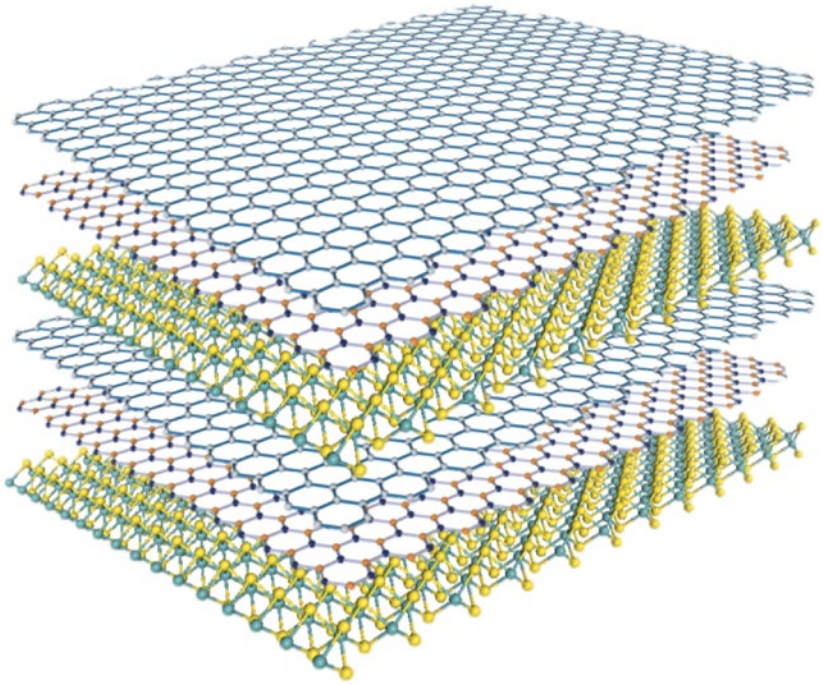
experimental attention and only a handful of experiments were performed in this area for several years. One of the groups that did perform experiments in twisted bilayer graphene was at MIT Physics, led by Pablo Jarillo-Herrero. Starting in 2010, the Jarillo-Herrero group investigated the electronic properties of twisted bilayer graphene, albeit first in the large twist angle regime, where another whole set of predictions and experiments (related to the field of topological condensed matter physics) were taking place. Then, in 2014 a new MIT graduate student, Yuan Cao, joined the group and together they decided to pursue the physics of small twist angle graphene (*Figure 2*). Initial experiments in 2016 in devices at twist angles of about 2° already showed promise, as the Fermi velocity seemed to be decreasing. The following year the group decided to try to fabricate devices with an angle near the magic angle, and explore what type of physics would occur.

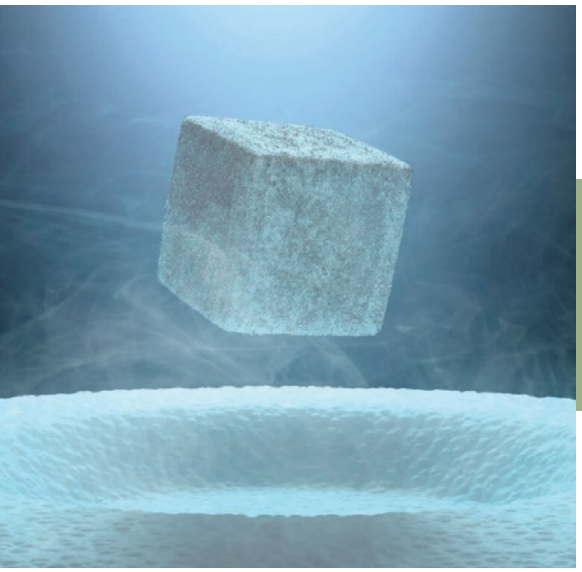
By spring 2017, the Jarillo-Herrero group finally managed to make some devices with an angle very close to the magic angle, and immediately surprises started to appear. The first was that these magic angle graphene devices exhibited an unusual type of insulating behavior. This was strange, as graphene is the world's best conductor. How can it be that you place the world's best conductor on top of itself and get an insulator? This did not make any sense! However, at the magic angle the electrons slow down a great deal and thus can interact very strongly with each other—hence these interactions, perhaps, were leading to a correlated type of insulator. This could be confirmed by changing the density of charge carriers in graphene, which was easily done through a voltage applied to a nearby metallic electrode. By doing this, the group realized the insulator appears only when the density of electrons happens to be a very particular value: when the electronic bands are partially filled, but such that there is an integer charge per moiré unit cell. At that point, the group was still struggling to fully understand the consequences of all this, when they made an even more surprising discovery—you may even say that then the "real magic" happened.

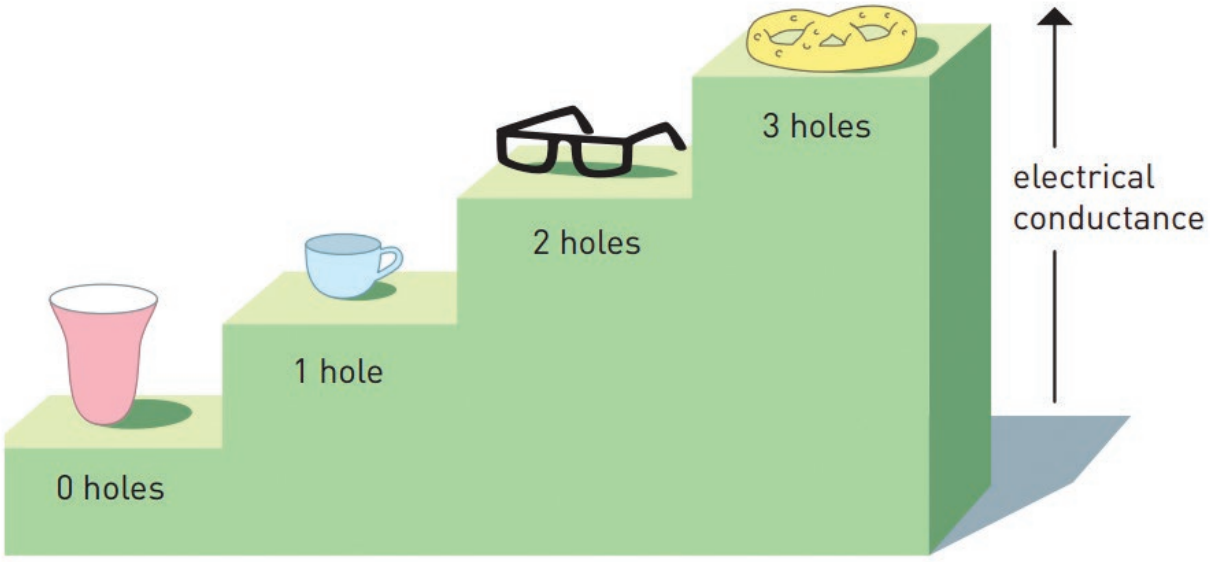
It turns out that adding a few charge carriers to the correlated insulator state led to superconductivity! In fact, they could control the superconductivity electrically, as it occurred only in a narrow range of density, or doping, and at temperatures below a "critical" value that exhibited a "dome-like" shape behavior. These types of superconducting domes, occurring near a correlated insulator, are typical of materials such as the high-temperature superconductors (*Figure 3*). Now, the big difference is that here they could control it electrically, whereas in most quantum materials changing the density of carriers implies growing a different crystal with different chemical impurities, different disorder, etc. Further, the group quickly realized that magic angle graphene was one of the strongest coupling superconductors known—the maximum critical temperature, of about one to a few Kelvin, was extremely high in comparison to the natural electronic temperature scale (known as the Fermi degeneracy scale), T_{Fermi}. All of these discoveries point in the direction of a very exotic superconductor.

FIGURE 4

The juncture of modern condensed matter fields into one system. *[Credits: below left, ktsdesign/Shutterstock.com; bottom, Johan Jarnestadt, The Royal Swedish Academy of Sciences.]*







Moiré is different, and the future of twistronics

These discoveries—correlated insulator behavior and superconductivity—were published in two back-to-back papers in the same issue of the journal *Nature* in the Spring of 2018 [6,7], generating great interest in the physics community. By now, several experimental groups around the world have reproduced these results and extended them in interesting directions. At the same time, new moiré heterostructures have been studied, exhibiting interesting correlated behavior and superconductivity with different characteristics. The burgeoning field of TWISTRONICS is quickly becoming a new paradigm in condensed matter physics.

In fact, apart from the ability to study correlation-driven phenomena in a highly tunable platform, moiré materials pose fascinating and fundamental challenges to theoretical physics. Remarkably, it brings together two of the grand themes of modern quantum matter physics (*Figure 4*). The first is the concept of strong correlations between the different electrons; the second is the concept of topology applied to electronic bands. Topology is the field of mathematics that deals with how shapes can be deformed into one another smoothly. In the context of electronic bands, topology deals with the global properties of the quantum mechanical wave function of the electrons, and how resilient they are to smooth deformations. We know now that there is a large class of materials which have bands that are topologically distinct from other, trivial ones. An insulator with a filled topological band has a number of characteristic signatures in experiments, notably in the presence of boundaries to the outside world, that have been thoroughly explored. [8]

Much of the advances in our understanding of topological materials has relied on the approximation of ignoring the interactions between the electrons, which is adequate for many systems. Nevertheless, theorists have speculated for a few years about the fate of strongly correlated electrons in a topological band. Remarkably, as shown by us and our collaborators [9], several of the moiré materials (including the twisted bilayer graphene system) possess topological bands. Thus, there is now a clear experimental context where the problem of strong correlations in a topological band needs to be confronted. Clearly, standard methods of modeling strongly correlated electron systems will fail in such a situation. The interplay of band topology and electron interactions requires a new theoretical framework that is currently unavailable. It remains to be seen what phenomena will be discovered in this fundamentally new arena.

As is often the case with fundamental physics discoveries, for now the physics community is mostly excited about the new knowledge and quantum phenomena arising in these fascinating moiré systems. However, it is tempting to speculate about what new quantum technologies, in the context of the current "second quantum revolution" (which involves, for example, quantum sensing, quantum cryptography and quantum computing), these twistronics systems will enable. At a basic level, magic angle graphene can be seen as an electrically tunable superconductor or a superconducting field effect transistor. These may be attractive as potential elements in cryogenic superconducting classical computing, or even as tunable superconducting qubits in quantum computation. Currently, the world's

best single-photon detectors are made with ultrathin superconductors and there is no thinner superconductor than magic angle graphene, so this may enable a new generation of ultrasensitive quantum photodetectors for communications, optical and space applications. These are just a couple of obvious examples, but it is exciting to see what the imagination of physicists and engineers will develop based on this new twistronics paradigm in materials science and condensed matter physics.

NOTES & REFERENCES

[1] *Coulomb repulsion* refers to the electrostatic repulsion between same charge sign particles, in this case electrons.

[2] *Fermi-Dirac statistics* refers to the quantum mechanical statistics of fermions, *i.e.*, particles with half-integer spin, which differ from those of bosons, *i.e.*, particles with integer spin, known as *Bose-Einstein statistics*.

[3] Professors Martin Zwierlein (MIT) and Markus Greiner (Harvard) groups.

[4] E. Suárez Morell, J. D. Correa, P. Vargas, M. Pacheco, and Z. Barticevic, *Phys. Rev.* B **82**, 121407 (2010).

[5] R. Bistritzer and A. H. MacDonald, *Proceedings of the National Academy of Sciences* **108**, 12233 (2011).

[6] Y. Cao, V. Fatemi, A. Demir, S. Fang, S. L. Tomarken, J. Y. Luo, J. D. Sanchez-Yamagishi, K. Watanabe, T. Taniguchi, E. Kaxiras, R. C. Ashoori, and P. Jarillo-Herrero, *Nature* **556**, 80 (2018).

[7] Y. Cao, V. Fatemi, S. Fang, K. Watanabe, T. Taniguchi, E. Kaxiras, and P. Jarillo-Herrero, *Nature* **556**, 43 (2018).

[8] An example is the famous quantum Hall effect, where charge carriers are transported only clockwise or counterclockwise by conducting channels at the boundary of the sample.

[9] Y.-H. Zhang, D. Mao, Y. Cao, P. Jarillo-Herrero, and T. Senthil, *Phys. Rev. B* **99**, 075127 (2019).

PABLO JARILLO-HERRERO *is the Cecil and Ida Green Professor of Physics at MIT. His research interests lie in the area of experimental condensed matter physics, particularly quantum electronic transport and optoelectronics in novel two-dimensional materials, with special emphasis on investigating their superconducting, magnetic and topological properties.*

A native of Valencia, Spain, Jarillo-Herrero joined MIT as an assistant professor of physics in January 2008 after postdoctoral appointments at Delft University of Technology (The Netherlands) and Columbia University. His many awards include an Alfred P. Sloan Fellowship; a David and Lucile Packard Fellowship; a DOE Early Career Award; a Presidential Early Career Award for Scientists and Engineers; an ONR Young Investigator Award; and a Moore Foundation Experimental Physics in Quantum Systems Investigator Award. In 2018, Jarillo-Herrero was elected a Fellow of the American Physical Society, and promoted to Full Professor of Physics.

SENTHIL TODADRI *is a Professor of Physics at MIT and senior member of the Department's renowned condensed matter theory group. His research is focused upon the theory of quantum many-particle systems, where he combines phenomenological modeling of experiments with abstract theoretical ideas and methods.*

A native of Chennai, India, Senthil Todadri joined MIT in January 2001 as an assistant professor of physics, receiving tenure in early 2007 and promotion to Full Professor in 2011. Prior to MIT, he was a postdoctoral fellow at the Kavli Institute for Theoretical Physics, University of California, Santa Barbara. Todadri's many awards and honors include an Alfred P. Sloan Research Fellowship; Research Innovation Award, The Research Corporation; Outstanding Investigator Award, Department of Atomic Energy, India; Outstanding Young Physicist Award, Indian Physics Association; and a Kavli Frontiers Fellowship. In 2013, he was elected a Fellow of the American Physical Society, and is currently Distinguished Visiting Research Chair (2011-2020) at the Perimeter Institute for Theoretical Physics, and a Simons Investigator (2013-2023) of the Simons Foundation.