

Overlapping two sheets of graphene shows a characteristic pattern.

SUPERCONDUCTIVITY WITH A TWIST

RESEARCHERS ARE SCRAMBLING TO UNDERSTAND CURIOUS BEHAVIOUR IN MISALIGNED STACKS OF GRAPHENE.

BY ELIZABETH GIBNEY

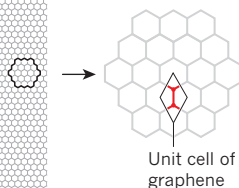
It was the closest that physicist Pablo Jarillo-Herrero had ever come to being a rock star. When he stood up in March to give a talk in Los Angeles, California, he saw scientists packed into every nook of the meeting room. The organizers of the American Physical Society conference had to stream the session to a huge adjacent space, where a standing-room-only crowd had gathered. “I knew we had something very important,” he says, “but that was pretty crazy.”

The throngs of physicists had come to hear how Jarillo-Herrero’s team at the Massachusetts Institute of Technology (MIT) in Cambridge had unearthed exotic behaviour in single-atom-thick layers of carbon, known as graphene. Researchers already knew that this wonder material can conduct electricity at ultra-high speed. But the MIT team had taken a giant leap by turning graphene into a superconductor: a material that allows electricity to flow without resistance. They achieved that feat by placing one sheet of graphene over another, rotating the other sheet to a special orientation, or ‘magic angle’, and cooling the ensemble to a fraction of a degree above absolute zero. That twist radically changed the bilayer’s properties — turning

JULIETTE HALSEY FOR NATURE

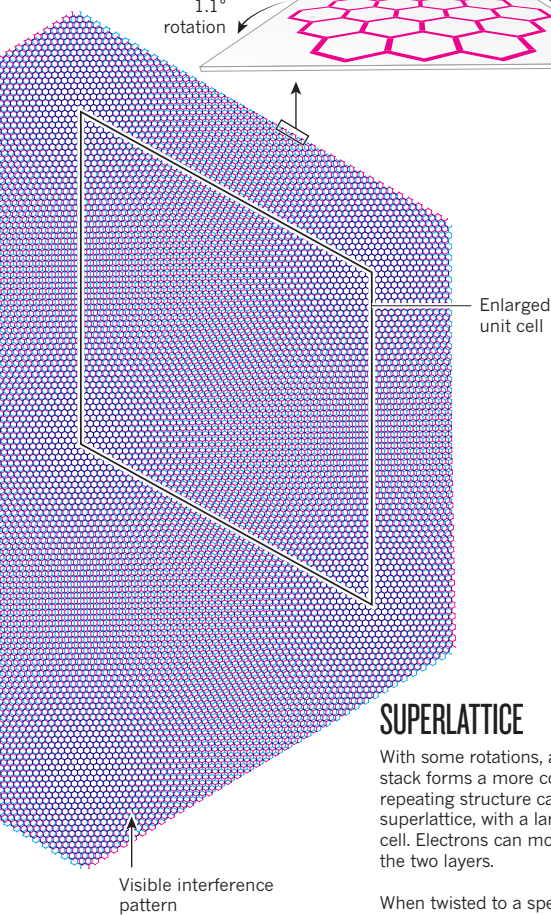
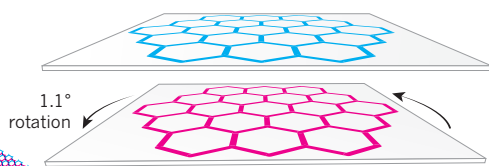
MAGIC ANGLE

Stacking one sheet of graphene on top of another can have a range of effects. If the sheets are rotated with respect to one another at just the right angle, the interaction of electrons in the two layers can give rise to new electronic properties.



SIMPLE STRUCTURE

The crystal structure of a single layer of graphene can be described as a simple repetition of two atoms — its 'unit cell'.



SUPERLATTICE

With some rotations, a two-layer stack forms a more complex repeating structure called a superlattice, with a larger unit cell. Electrons can move between the two layers.

When twisted to a specific 'magic angle', the stack seems to exhibit behaviour not seen in ordinary graphene, such as superconductivity.

it first into an insulator and then, with the application of a stronger electric field, into a superconductor.

Graphene had previously been cajoled into this behaviour by combining it with materials that were already known to be superconductors, or by chemically splicing it with other elements. This new-found ability to induce the same properties at the flick of a switch turned heads. "Now you put two, non-superconducting atomic layers together in a certain way and superconductivity pops up? I think that took everyone by surprise," says ChunNing Jeanie Lau, a physicist at the Ohio State University in Columbus.

Physicists at the meeting were even more excited because of the way in which a graphene bilayer seems to become a superconductor. There were hints that its remarkable properties arose from strong interactions or 'correlations' between electrons — behaviour that is thought to underlie bizarre states of matter in more-complex materials. Some of those materials, namely ones that superconduct at relatively high temperatures (although still well below 0°C), have baffled physicists for more than 30 years. If superconductivity in simple graphene is caused by the same mechanism, the material could be the Rosetta stone for understanding the phenomenon. That, in turn, could help researchers to engineer materials that superconduct close to room temperature, which would revolutionize many areas of modern technology, including transportation and computing.

"Immediately I could see pretty much everyone I know become really excited," says Lau. But while she listened in amazement to the talk, others couldn't wait. Andrea Young, a condensed-matter physicist at the University of California, Santa Barbara, had left the meeting to rush back to his laboratory. His team was one of a handful around the world already exploring twisted graphene, looking for hints of recently predicted strange behaviour. Young scanned the *Nature* papers^{1,2} from the MIT group, which were published two days ahead of the talk, and found what he needed to know to replicate the experiment. That turned out to be harder than anticipated. But by August, having joined forces with a group at Columbia University in New York City led by physicist and friend Cory Dean, he and his team succeeded³. "We had reproduced it many times ourselves," says Jarillo-Herrero. But having the confirmation of a second group, he says, "was tremendously reassuring".

Although the Young and Dean collaboration was the first to publicize its replication results, activity behind the

scenes is frenetic, says Lau. "I haven't seen this much excitement in the graphene field since its initial discovery," she says. Three other teams told *Nature* that they have replicated some or all of the MIT findings, although some are keeping their cards close to their chests while they experiment with other 2D materials and tweak layers in new ways, looking for other displays of strong electron interactions. "Everyone is taking their favourite thing and twisting it with their other favourite thing," says Young. Meanwhile, theorists trying to explain the behaviour have posted more than 100 papers on the topic to the arXiv preprint server. But sorting out whether the same mechanism that underlies superconductivity in high-temperature superconductors is at play in twisted graphene will take much more information, says Lau. "So far, apart from the fact that this is a really interesting system," she says, "I don't think the theorists agree on anything."

FINDING THE MAGIC

The audience at Jarillo-Herrero's talk in Los Angeles was excited but also sceptical. Conference delegates teased him that the last time someone had presented something so cool, it was Jan Hendrik Schön, whose string of dazzling results on superconductivity and other phenomena turned out to be fraudulent. "They were joking," Jarillo-Herrero says, "but they said they'd need to see this reproduced before they would believe it."

Although twisted graphene's superconducting behaviour came as a surprise, the idea that something intriguing could happen was not. Overlaid at angles of more than a few degrees, two graphene sheets usually behave independently. But at smaller angles, the misalignment of the two lattices can create a 'superlattice' in which electrons can move between layers. Theorists had predicted^{4,5} that at specific small twists — magic angles — the underlying structure of the superlattice would drastically change the behaviour of electrons, slowing them down and enabling them to interact in ways that change the material's electronic properties (see 'Magic angle'). In theory, all kinds of layered 2D material, when twisted to the proper angle, can form such superlattices. But no one knew how a material's properties might change, or at what angle such a change might occur.

Back in 2010, Eva Andrei, a physicist at Rutgers University — New Brunswick in New Jersey, and her colleagues saw hints of strange behaviour in graphene⁶ around the same magic angle later observed by Jarillo-Herrero and his team, but many doubted whether the theory worked at all. "I didn't believe it, says Philip Kim, an experimental



physicist at Harvard University in Cambridge, Massachusetts. “But I admit I was completely wrong,” he says.

When Young arrived back at his lab in March, he thought that reproducing the MIT group’s results seemed trivial, he says. Young’s team could achieve the very low temperatures needed, and the researchers were already experts in preparing very clean samples. But coaxing graphene sheets to align at just the right angle — a twist of around 1.1° — turned out to be a struggle.

Hitting the angle is difficult, not least because it subtly changes from sample to sample, depending on how each one is made. “You have to do some searching,” says Andrei. Moreover, because twisted graphene’s structure is so close to that of graphite, in which successive layers are all oriented in the same direction, the slightest heat or strain can cause the layers to fall into alignment. “It doesn’t want to stay where you put it,” says Young.

Dean’s lab, which was also working on the problem, hit on a solution: when the team overshot the twist in a number of devices, at least some samples would settle at the magic angle as they rotated back towards alignment. But getting those samples to superconduct required equipment that could reach a fraction of a degree above absolute zero, which his lab lacked. Working with Young’s team, the researchers soon measured several devices in which resistance shot up — characteristic of an insulator — but dropped to zero, as in superconductors, when they fed in more electrons by applying an electric field.

It is the only other team apart from Jarillo-Herrero’s to publish its findings so far, but that will not be the case for long, says Andrei. “Everyone I know is working on this,” she says.

SOMETHING UNCONVENTIONAL

One reason for the intense interest in twisted graphene is the stark similarities between its behaviour and that of unconventional superconductors. In many of these, electric current runs without resistance at temperatures well above what the conventional theory of superconductivity generally allows. But quite how that happens remains a mystery: one that, when solved, could allow physicists to engineer materials that conduct electricity with zero resistance near room temperature. Achieving that could enable radically more-efficient transmission of electricity, and, by slashing energy costs, allow superconductors to find uses in a host of new technologies.

All forms of superconductivity rely on electrons pairing up in ways that allow them to travel without resistance. In conventional superconductors — the kind that power the magnets in magnetic resonance imaging (MRI) machines — electrons pair up only indirectly, as a by-product of the interplay between the particles and vibrations in their

atomic lattice. Electrons ignore their fellows, but end up thrown together in a way that helps them to navigate without resistance at temperatures a few degrees above absolute zero. But in unconventional superconductors — many of which carry current with zero resistance at closer to 140 kelvin — electrons seem to pair up through a direct and much stronger interaction.

The MIT experiments showed hints of this unconventional superconductivity. Although twisted bilayer graphene became superconducting only at extremely low temperatures, it did so with very few freely moving electrons. That suggests that, unlike in a conventional superconductor, whatever force drew the electrons together must be relatively strong. The proximity of the superconducting state to an insulating one also mirrors what is seen in a group of high-temperature superconductors made from ceramics, called cuprates. In those systems, the zero-resistance state often borders a ‘Mott’ insulator — in which no current flows, despite the presence of free electrons, because mutual repulsion between the particles pins them in place.

If the same mechanisms are at play in twisted bilayer graphene, it could be boon to theorists. One problem with cuprates, such as yttrium barium copper oxide, is that they are a jumble of elements that proves difficult to model. “The hope is of finding the same phenomenology but in a much simpler system, one which theorists can stick their teeth into and make some progress,” says Andrei.

Graphene is also an experimentalist’s dream. Studying the switch to superconductivity means measuring what happens as more electrons are added to the material. In cuprates, this is done by inserting atoms of a different element into the material — a process known as doping — which means making an entirely new sample for each point on a chart. In twisted graphene, however, researchers can make the switch simply by turning a knob on a voltage source, says Andrei. “This is a huge benefit.”

No one knows yet whether twisted graphene is really acting like an unconventional superconductor, or even whether the behaviour arises exactly because of the conditions described by the magic-angle theory. The flood of theory papers posted since March covers every possibility. Because correlated systems such as those seen in twisted graphene are too complex to calculate in their entirety, theorists use approximations that differ from model to model. That makes theories flexible enough for physicists to sometimes tweak them to fit new data, says Young. Few theories explain the findings in full, and many do not include predictions that would allow experimentalists to tease apart different scenarios, adds Jarillo-Herrero. For “an experimentalist like me they all seem similarly sensible,” he says. “I’m a bit disoriented in theory land.”

Physicist Pablo Jarillo-Herrero (far left) with three graduate students in his lab at the Massachusetts Institute of Technology.

So far, there is evidence for both unconventional and conventional superconductivity in graphene. As-yet-unpublished data from the MIT group suggest that other phenomena seen in unconventional superconductors are also present in the material, says Jarillo-Herrero. For one thing, his team has observed that the strength of the magnetic field necessary to strip superconductivity from a sample, through a process known as the Meissner effect, varies with direction (it should be uniform in conventional superconductors).

CAUTIOUS APPROACH

But results from Young and Dean's groups suggest more caution is needed. Their samples are more uniform than those of the MIT group, says Young, and show some contrasting results. In particular, superconductivity appears when the number of electrons is turned down but not when it is turned up, an asymmetry that is arguably more consistent with a conventional superconductor. And, in contrast to cuprates, which can be insulating at higher temperatures than those at which they superconduct, in twisted graphene the two states seem to be present in a similar temperature range, he adds. Further tests, such as seeing whether the superconducting state still occurs when experimentalists restrict vibrations in the sample but still allow electron interactions, could help to clarify the situation, says Young. Andrei's group is also working on imaging the material at the atomic level, to reveal effects that could be washed out when studying the sample as a whole. Andrei says her team's preliminary data have revealed new phenomena that could help to make sense of the underlying physics, although she is so far unwilling to give any more away.

Understanding the outcomes of experiments — along with devising set-ups that work well on 2D materials — can be a challenge. In this delicate system, Young says that even the material used to make the electrodes can interfere with results. “You have to be careful about interpreting what you see, because you don't know what's an intrinsic property of the system and what's an effect of the set-up,” Young says the mechanism behind the superconductivity could well turn out to be conventional, but that it is exciting even if it doesn't help to explain high-temperature superconductivity. “This is already one of the coolest results to come out of this field in the past ten years,” he says.

Regardless of whether it resembles exotic forms of superconductivity, researchers say the system is fascinating because it is a rare example of dramatic change coming from a small physical tweak. “That fact alone is pretty amazing and remarkable,” says Dean. “What is it about this system that gives rise to superconductivity that is absent away from this precise twist angle?”

Whatever is going on in the superconducting state, physicists agree that the accompanying insulating state is almost impossible to explain without some kind of interaction between electrons. Like a metal, graphene is ordinarily conductive, with free electrons that interact only with the atomic lattice and not with one another. Somehow, despite the presence of these free electrons, which are absent in conventional insulators, bilayer graphene can block the flow of electricity, suggesting that interactions are at play.

This is exciting because electron interactions underlie many of the weird and wonderful states of matter that have been uncovered over the past few decades. These include quantum spin liquids — strange disordered states in which electrons' magnetic fields never align — and fractional quantum Hall states, phases of matter defined by topology, a previously unknown kind of unifying property that might be harnessed to build extremely robust quantum computers. “Understanding strongly correlated systems is where a lot of the big questions, and also perhaps big opportunities, are in condensed-matter physics right now,” says Young. Many of these states emerge under conditions that, at least to electrons, look similar to those that arise in graphene at the magic angle. This raises the possibility that other intriguing states could emerge from twisted bilayers, says Rebeca Ribeiro-Palau, a physicist at the Centre for

Nanoscience and Nanotechnology in Palaiseau, France, and formerly a postdoc in Dean's lab. “For me, the presence of a superconducting state is a symptom of something more interesting,” she says.

Crucially, graphene and other 2D systems allow for much greater experimental control than do other strongly correlated materials, she says. Researchers can smoothly tune not only the electric field to alter behaviour, but also the twist angle — while at Columbia, Ribeiro-Palau and her colleagues used the tip of an atomic force microscope to smoothly spin one layer with respect to the other⁷. As has been demonstrated by the Young and Dean collaboration, experimentalists can also fine-tune the distance between layers by applying pressure. Squeezing the layers closer together increases the strength of the interaction between electrons in the sheets, a boost that means magic-angle conditions can happen at much bigger — and more stable — rotations.

DOING THE TWIST

Kim and his colleagues have already replicated the graphene finding, he says. Now they are looking to see whether they can also generate superconductivity or perhaps magnetism in twisted layers of more-complex 2D semiconductors, called transition-metal dichalcogenides. Before the MIT result, Kim's was one of a few teams that was already probing the effects of rotating one 2D layer on top of another, a nascent area of research sometimes known as twistronics. With the possibilities demonstrated in graphene, the idea is now taking off. “In principle, you can apply the concept to all the 2D materials and twist to see what happens,” says Kim. “There is the possibility that you find something completely unexpected.”

Meanwhile, Feng Wang at the University of California, Berkeley, says he and his colleagues have seen signs of superconductivity in triple-stacked layers of graphene even without a twist. Layering three sheets in a particular orientation⁸ achieves a superlattice geometry similar to that in magic-angle twisted bilayers, and results in similarly strongly correlated physics, he says.

Physicists are optimistic that the crossover between two previously separate fields — 2D materials and strongly correlated systems — will lead to exciting results. “It's giving us an opportunity to talk to a whole community of people we haven't had the chance to talk to in the past,” says Dean. And applied physicists are thinking about how the unusual properties of twisted 2D stacks might be harnessed to store and process information in super-efficient ways. Rotating or squeezing materials could also become a new way to switch an electronic device's behaviour.

But for now, many researchers are focused on sorting out the fundamentals. This month, experimentalists and theorists will gather at the Kavli Institute for Theoretical Physics in Santa Barbara for a workshop that will thrash out key questions in the burgeoning field. Jarillo-Herrero hopes the meeting will help bring theorists into alignment. “At the moment, they can't even agree on the basics.” By then, more experimentalists might be willing to show their hand and publicly reveal their data, he adds.

Even though physicists don't know how significant the discovery will ultimately be, Young says there's a takeaway message from the dozens of theory papers that have appeared since the MIT publications: “Anything could come out of this, and something certainly will.” ■

Elizabeth Gibney is a senior reporter for Nature in London.

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