

# Graphene and Topological Insulators

## Meet the Novel Materials Pushing the Physics Frontier

**Q**uantum mechanics is the fundamental theory that describes most physical phenomena that occur in nature. Its basic equation, the Schrödinger equation, accurately describes the electronic properties of solids—of great importance in modern technology. To describe the behavior of particles that travel near the speed of light, such as neutrinos, the theory must be modified to take Einstein’s theory of relativity into account. This was accomplished by physicist Paul Dirac in the 1930s. However, particles behaving according to the Dirac equation are usually thought to lie within the realm of high energy physics.



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Imagine then that such fast-traveling particles were electrically charged. How would they conduct electricity, and what sort of new quantum phenomena would this give rise to? Remarkably, low-dimensional realizations of this hypothetical scenario have recently become accessible to physicists with the discovery of materials whose electronic properties are governed by the Dirac equation rather than the non-relativistic Schrödinger equation. This novel behavior is shaking the foundations of modern condensed matter physics and suggests radically new electronic properties with far-reaching consequences beyond the academic domain, including potential applications ranging from terahertz (THz) transistors to fault-tolerant quantum computation. All this excitement has resulted in a worldwide effort to explore novel quantum phenomena in new regimes made possible by the low-dimensionality and unique relativistic-like electronic structure of two such classes of materials: GRAPHENE and TOPOLOGICAL INSULATORS.

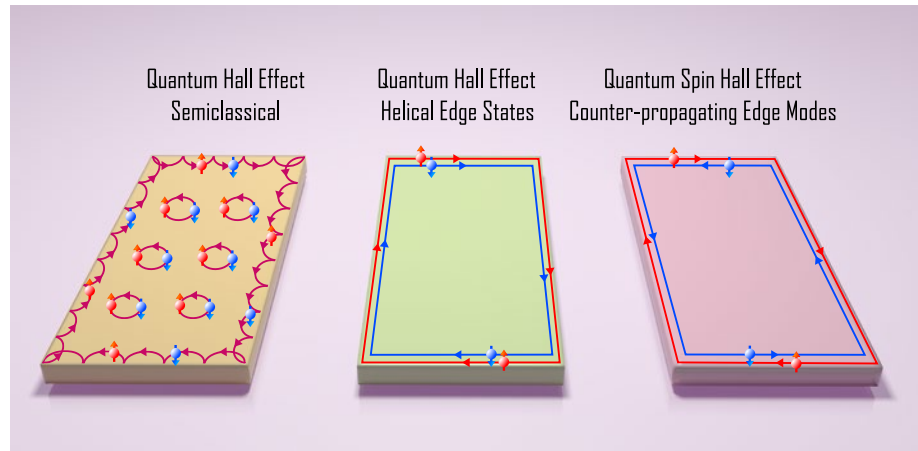
A single layer of graphite, or graphene, consists of a single-atom thick sheet of carbon atoms arranged in a honeycomb lattice. In graphene, charge carriers move through the layer with a velocity of  $10^6$  m/s—1/300th the speed of light. Yet, this velocity is independent of their energy, a property usually possessed only by massless particles moving at the speed of light, such as photons or nearly massless neutrinos. The earliest experiments—which demonstrated that the behavior of electrons in graphene is well described by the two-dimensional (2D) Dirac equation for massless particles—were related to a fundamental phenomenon in condensed matter physics called the QUANTUM HALL EFFECT (QHE).

FIGURE 1

**(left) Semiclassical picture of orbits of electrons in a two-dimensional electronic system with an applied perpendicular magnetic field.** Notice the “skipping orbits at the edge.”

**(center) Quantum mechanical picture of edge states in the quantum Hall effect:** edge states of opposite spin travel in the same direction.

**(right) Quantum spin Hall effect:** edge states of opposite spin travel in opposite directions.



## The quantum Hall effect

Twenty-five years before the discovery of graphene, there was a remarkable observation (which subsequently won a Nobel prize) in two-dimensional semiconductor systems, known as the quantum Hall effect. If you take a long piece of semiconductor and apply a voltage to the two ends, in the presence of a perpendicular magnetic field, a voltage appears in the transverse direction (known as Hall voltage). Moreover, the Hall resistance (the Hall voltage divided by the current flowing through the semiconductor) is, to within a few parts in a billion, quantized and given by  $h/\nu e^2$  where  $\nu$  is an integer number called the “filling factor” whose value depends on the 2D electron density and magnetic field strength. ( $\nu$  is precisely equal to the number of electrons in the sample divided by the magnetic flux threading sample, as measured in units of the magnetic flux quantum  $h/e$ .)

One can get an intuitive view of the QHE by using a semiclassical picture. In the presence of an applied magnetic field, electrons in the interior (or bulk) of the 2D system execute circular motion (*Fig. 1, left*). At the edges of the 2D system, however, the electrons cannot complete the circular orbits, and instead bounce off the edges, following skipping orbits—always moving clockwise or counter-clockwise around the sample, depending on the direction of the magnetic field. In a quantum mechanical picture, electrons in the 2D system occupy discrete energy levels called Landau levels. In the quantum Hall (QH) regime, in the bulk of the 2D system, the Landau levels are fully occupied, and there is no room for electrons to move around. However, near the edge of the system, the Landau levels are only partly occupied, allowing for “one-dimensional highways” for electrons. These are called QH edge modes, and they have the property that electrons only move in one-direction, either clockwise or anti-clockwise. As a result, electrons moving in one edge mode cannot turn back on that side of the sample—they cannot “backscatter.” To backscatter, they would have to jump all the way to the other side of the system, where electrons are moving in the opposite direction. But this jumping (or tunneling) is very strongly suppressed because of the large distance between the two edges.

The edge modes thus carry current in one direction and are perfectly transmitting. In 1957, Rolf Landauer at IBM had predicted that one-dimensional electronic modes that transmit perfectly, if they existed, would each have a conductance (current divided by voltage) exactly equal to  $e^2/h$ . This value is a universal contact resistance of  $25,812.8076 \Omega$  for injecting currents into these perfectly conducting channels, and it leads to the observed quantized Hall conductance. The ultra-precise quantization of the Hall conductance is now used as a metrological resistance standard, precise to within nine decimal places!

## Topology in condensed matter physics

The fact that the Hall conductance is quantized to a ratio of fundamental constants multiplied by an integer  $\nu$ , made some physicists realize that there was a deeper connection between the QHE and a branch of mathematics called TOPOLOGY. Topology deals with the properties of space that are preserved under continuous transformations. Speaking metaphorically, topology allows you to distinguish at a fundamental level an orange from a doughnut. The distinction lies in the number of handles that the object has. For an orange, it's zero, and for a doughnut it's one. An orange and a cucumber are topologically equivalent, because both have zero holes and one can smoothly deform (at least mathematically!) an orange into a cucumber. A doughnut and a coffee mug with a handle are also topologically equivalent, because both are threaded by one hole. Now, handles come in precise integers: one cannot have an object threaded by half a hole, or by 999 thousandths of a hole, so there is a topological quantity (or, genus) that tells you how many handles an object has.

What is the connection between topology and QHE? In the case of the QHE, the filling factor is also mathematically a topological quantity, called the Chern number, which is a precise integer and tells you the number of edge modes that are carrying current. So, from a modern physics perspective insulators can be divided into two types: *topologically trivial*, such as glass, which are insulators in the bulk of the material and do not conduct electricity; and *topologically non-trivial* insulators, such as a QH insulators. In non-trivial insulators, while the bulk of the material is an insulator, from the electronic transport point of view they are conductors, where the current gets carried by topological objects, that is, the number of edge (or boundary) modes.

## Graphene and the quantum Hall spin effect

How does this all link to graphene? In graphene, because of the massless Dirac equation governing the behavior of electrons, the QHE is very different from that occurring in standard semiconductors, such as silicon. Moreover, different types of topological QH states can be obtained, as recently demonstrated in a collaboration between the Ashoori and Jarillo-Herrero groups.[1]

Just as in the Dirac equation for massless neutrinos, particles can exist with less-than-zero energy, called antiparticles. In graphene they are called simply “holes” (a hole is the absence of an electron). Holes are positively charged particles, so in a magnetic field they move in opposite direction to electrons. Graphene is an electrically tunable system, where the charge density can be changed such that you have zero electrons or holes. If this is the case, we say that the Fermi energy (the energy of the last occupied electronic state) is at the Dirac point (*Fig. 4a, page 52*).

What happens when a magnetic field is applied perpendicular to the plane of a graphene system with the Fermi energy set to the Dirac point? Theory predicts that a zero energy Landau level will form. This zero-energy Landau level is actually composed of mixed electron and hole states. But which way will the edge modes move? MIT’s Dimitri Abanin, Leonid Levitov, and Patrick Lee addressed this question in 2006.[2] They predicted something unusual: that particles with spin-up would move in one direction (say, clockwise) around the edges of the sample, whereas particles with spin-down would move in the opposite direction. So you have counter-propagating, spin-polarized edge modes. If the particle spins could flip, then particles would constantly reverse direction, and one would find zero conductivity through the sample. If the spins did not flip, one would instead measure a conductivity of  $2e^2/h$ .

Experimenters attempted to see this effect, but it didn’t work. It turned out that many-body physics of the 2D layer instead created a new state within the layer, and this system had zero conductivity. This many-body state behaves somewhat like an antiferromagnet, with particles localized within the 2D system and with equal numbers of spin-up and spin-down particles.

Pappalardo Fellow Andrea Young, together with postdoc Ben Hunt, graduate student Javier Sanchez-Yamagishi, and undergraduate Sang-Hyun Choi, worked on this problem in a collaboration with Professors Pablo Jarillo-Herrero and Ray Ashoori. They decided to apply a large magnetic field in the plane of the graphene to try to destroy the antiferromagnetic state by forcing the spins to align with the applied field, thereby creating the system predicted by Abanin, Lee, and Levitov.

After characterizing samples and seeing indications of the new state at MIT, Young, Hunt, and Sanchez-Yamagishi took a trip to the National High Magnetic Field Lab in Tallahassee, Florida, where they applied fields of up to 35 Tesla to a graphene sample. They saw a remarkable change in the sample conductance upon adding a parallel magnetic field. *Figure 2* displays the sample conductance while holding constant the perpendicular applied magnetic field to a field of 1.4 Tesla and varying the total magnetic field strength so as to change the component of the magnetic field applied parallel to the plane of the graphene. The black curve labeled as  $B_T=1.4T$  shows the results for a solely perpendicular applied magnetic field. The curves correspond each to successively increasing the field in the graphene plane. The horizontal axis on the graph marks the changing gate voltage, and the gate voltage controls the density of electrons (or holes) in the sample. The Dirac point (where, in zero magnetic field, there are no electrons and no holes in the sample) is at about 35 mV of gate voltage.

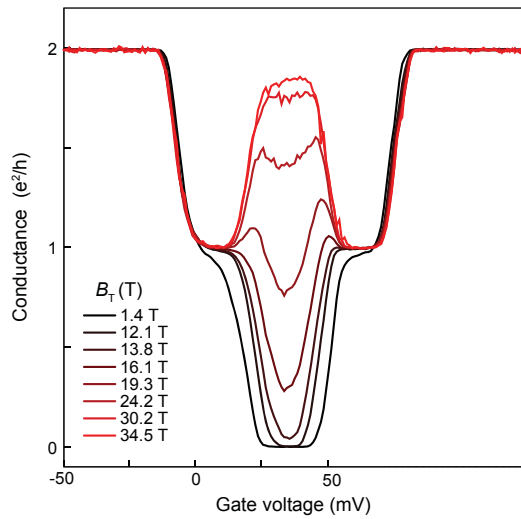


FIGURE 2

**Conductance of a graphene sample** as a function of a gate voltage that controls the electron density. The electron density is zero (the Dirac point) when the gate voltage is set to about 35 mV. In all of the data, there is an applied magnetic field of 1.4 Tesla perpendicular to the graphene plane. The different curves are for different values of in-plane field (to give a total field of  $B_T$ ).

For the  $B_T=1.4$  T curve, the minimum showing zero conductance between about 20 mV and 40 mV arises from the collective antiferromagnetic state described above. As the parallel field is increased, the conductance in this range increases until it reaches nearly  $2e^2/h$ , as Abanin, Lee, and Levitov had predicted. The system has counter-propagating edge states of opposite spins. The fact the conductance doesn't quite reach  $2e^2/h$  indicates that there is some backscattering of these

edge channels—some spins must be flipping. In fact, the state created is a novel topological state, different from the quantum Hall state, called the quantum spin Hall (QSH) state. However, it is a very different realization of the QSH state than originally predicted.

### Adding metal to a quantum conductor can increase its resistance!

An even more remarkable result happens when small, disconnected, electrically “floating” gold contacts are added to the edges of the sample. *Figure 3a* shows schematic pictures of a piece of graphene with four gold contacts on it. Consider case “A” with two contacts on either side of the sample shunted together. On both the top and bottom of the sample, there will be two counter-propagating edge states with opposite spin. As only one of the two edge states on the top moves from left to right, a current flowing from left to right will be handled entirely by this state. That state will have the potential of the left contact since charges in the right flowing state stay in equilibrium with the reservoir (the left contact) from which they emanated. The left flowing state is instead in equilibrium with the right reservoir. The two counter-propagating edge states on the top are thus out of equilibrium. However, they

do not equilibrate because this would require flipping spins, and there is no mechanism for doing this. As there is one edge state on the top that moves from left to right and another (with opposite spin) on the bottom, each giving  $e^2/h$  conductance, the measured conductance is  $2e^2/h$ . This is just what is seen for the red curve in *Figure 3b*.

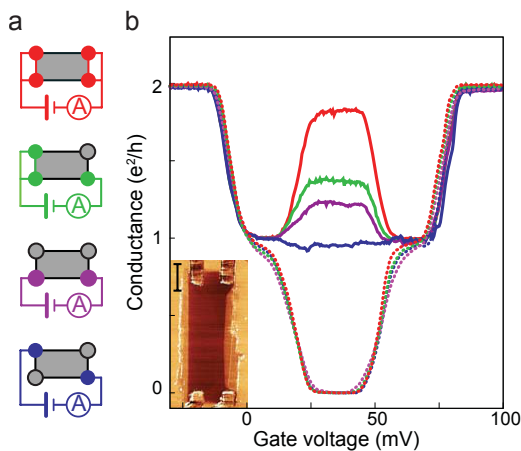


FIGURE 3

**Conductance of the same graphene sample** described in *Figure 2* under different contact arrangements. Inset photo: graphene sample with four contacts (scale bar = 1 micron).

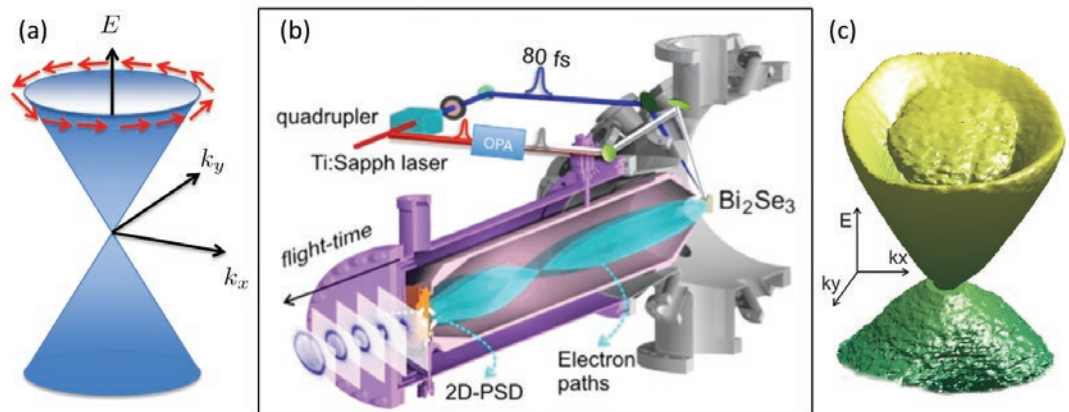


FIGURE 4

**(a) Theoretical dispersion relation of a topological insulator.** The tip of the cone where the bands cross is known as the Dirac point. Red arrows show the spin of the electrons, which are locked perpendicularly to their momentum.

**(b) A sketch of the novel tr-ARPES setup.** Fourth harmonic of Ti-Sapphire laser is used to photo-eject electrons, which are detected with angle, resolved time of flight detector. This enables simultaneous detection of 3D band structure ( $E$ ,  $k_x$  and  $k_y$ ) as shown in (c).

**(c) For the case of  $\text{Bi}_2\text{Se}_3$ .** The intensity inside the cone is due to parabolic bulk band. To study time resolved changes, part of the laser pulse is split and used as the excitation beam. An optical parametric amplifier (OPA) allows tuning the pump photon energy.

Now consider the green schematic in *Figure 3a*. The path from the top left contact to the bottom right contact now passes through an electrically disconnected “floating” contact on the upper right. Ordinarily, one might expect that adding a dab of gold to a sample would do nothing, or perhaps enhance the conductance. Instead, the conductance shown in *Figure 3b* drops. This happens because the floating contact equilibrates the oppositely traveling edge states by providing them with a volume in which they can flip their spins. Spins can flip inside the gold contact either by spin-orbit effects from the heavy gold atoms or through magnetic impurities in the metal. The floating contact, by symmetry, will have electric potential exactly halfway between the connected contacts. Considering each leg in the top path as having a conductance of  $e^2/h$ , the current through the top path will be divided by two. So, the total conductance through the structure will be given by  $e^2/h$  for the bottom path plus  $e^2/2h$  for the top path, giving a total of  $3e^2/2h$ .

Each leg along the edges of the sample can be seen as having a resistance of  $h/e^2$ . Placing a floating gold contact on one side effectively adds another leg. Amazingly, the resistance could be made to increase indefinitely just by adding more floating gold contacts. Adding gold increases the resistance!

## Topological insulators in two- and three-dimensions

To create the weird and novel state described above, the team from the Ashoori and Jarillo-Herrero groups had to apply high magnetic fields and make measurements at low temperatures. Theorists have thought of ways of realizing similar phenomena at room temperature and at zero magnetic field. In 2005 and 2006, theorists at the University of Pennsylvania and Stanford proposed a clever idea: the external magnetic field needed in the QHE can be replaced by the effective magnetic field due to spin-orbit coupling. In spin-orbit coupling, in the rest frame of the moving electron, the electric field of the nucleus is felt as an effective magnetic field. The effect of this effective magnetic field on the energy of electrons depends on the sign of their spins— increasing energies for one sign of spin and decreasing them for the other. Therefore, electrons with opposite spins are pushed in opposite directions by the spin-orbit interaction. Finally, the magnitude of this effective

field is larger for elements near the bottom of the periodic table since they have a bigger atomic number.

If one pictures the QHE in a two-dimensional sample with this effective magnetic field, similar edge states should form. But, in this case, since the direction of the field switches for different spin species, two copies of edge states should form. If, for example, electrons with spin up move clockwise, the ones with spin down should move counterclockwise. The nice thing is that, even though there will now be two copies of edge states traveling in opposite directions, non-magnetic impurities will still not be able to scatter electrons backwards, since the spins do not match. The bulk of the material will be insulating just as in the quantum Hall effect. This phenomenon is called quantum spin Hall effect (QSHE). As you can see, the phenomenology is identical to the graphene case discussed above, but this time it occurs at zero applied magnetic field.

The quantum spin Hall effect refers to one-dimensional edge states of a two-dimensional sample, but one can generalize this effect into three dimensions. In this case, instead of 1D spin-polarized edge states of a 2D material, one pictures 2D surface states of a 3D material. The bulk of this material is an insulator, but a 2D conducting state exists at the surface. Similar to the QSHE, electrons travelling in opposite directions have opposite spins. These materials are called 3D topological insulators. Typical examples are  $\text{Bi}_2\text{Se}_3$ ,  $\text{Bi}_2\text{Te}_3$ , as well as several other related materials.

The surface electrons in topological insulators have a number of unusual characteristics. First, similar to electrons in graphene, they behave like ultrarelativistic particles. The energy of the topological electrons plotted against their momentum forms a cone known as a Dirac cone (*Fig. 4a*). Secondly, much like the graphene edge-states described above, the spin of the surface electrons in the topological insulators is locked perpendicular to their momentum (*Fig. 4a*). Finally, in the absence of magnetic impurities, the spins on the surface don't flip and this surface state is robust against moderate disorder. The crossing of the bands at the tip of the Dirac cone (the Dirac point), is protected by time reversal symmetry (TRS).

## Seeing electrons with light: ARPES

The exotic surface state of topological insulator was first experimentally discovered by angle-resolved photoemission spectroscopy (ARPES), the standard method to measure the electronic band structure. In this technique, a high-energy photon is used to photo-emit electrons from the solid. Measuring the energy and momentum of these photoelectrons and using energy momentum conservation yields their energies and momenta inside the solid. This experiment is usually performed in synchrotrons where high-intensity soft X-ray light is available to do photoemission experiments. Conventional ARPES detectors measure energy and *one* component of momentum of the electrons ( $E$  vs  $k_x$ ). To measure the other component of the momentum ( $k_y$ ), one needs to rotate the sample and measure the ARPES spectra at different angles. Patching together these spectra, 3D disper-



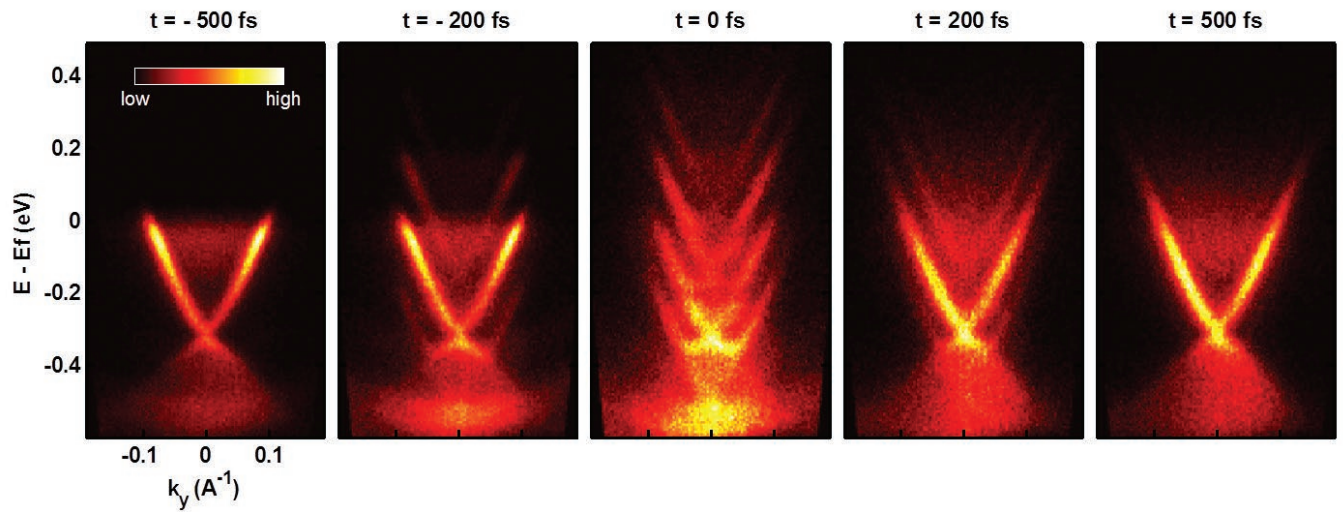


FIGURE 5

**Angle-resolved photoemission spectra (ARPES)** of  $\text{Bi}_2\text{Se}_3$  at several pump-probe time delay  $t$  under strong mid-infrared excitation. At  $t = -500$  fs time delay, the probe pulse arrives earlier than the pump, seeing no change to the band structure of  $\text{Bi}_2\text{Se}_3$ . When the two pulses start to overlap in  $t$ , the spectrum shows duplicates of the original Dirac cone at higher and lower energies. Five hundred fs after the excitation, the side-bands disappear and there are occupation in both bulk and surface states above the Fermi level.

sion relation ( $E$  vs  $k_x$  and  $k_y$ ) can be obtained. Graduate student Yihua Wang in the Gedik group developed a novel ARPES system at MIT that can measure the 3D dispersion without needing to rotate the sample (Fig 4b, page 52). This system uses the fourth harmonic of Ti-sapphire laser at 6 eV as the light source, as well as a new type of detector based on time of flight. The energy of the electrons is obtained from the travel time and the  $x$  and  $y$  positions of the location they hit at the detector yield, both  $k_x$  and  $k_y$ . Figure 4c on page 52 shows the experimentally measured Dirac cone of  $\text{Bi}_2\text{Se}_3$  using this technique.

When the surface state of TI is effectively coupled to certain orders, *broken symmetry states* are predicted. For example, the presence of magnetic order on the surface breaks the time reversal symmetry and opens a gap at the Dirac point. This should lead to a quantum Hall state where electrostatics similar to that of an axion are predicted to exist. Experiments have only recently demonstrated the existence of an anomalous quantum Hall state by doping a TI with magnetic impurities.

## Time-periodic perturbations and Floquet-Bloch physics

Breaking time-reversal symmetry using electromagnetic fields represents a completely different method to engineer novel states of matter on the surface of a TI. The Gedik group has recently used such a scheme to achieve breaking time reversal symmetry on the TI surface with light. Light, being an oscillating electromagnetic field, gives rise to a periodic perturbation in Hamiltonian. Floquet's theorem states that a Hamiltonian periodic in time has quasistatic eigenstates that are evenly spaced in energy by the drive photon energy. These so-called "Floquet states" can be regarded as a time analog of Bloch states, which are the eigenstates of a Hamiltonian periodic in space. In the case of the Bloch states, spatial periodicity of the Hamiltonian results in electron wavefunctions that are periodic in momentum. In the case of the Floquet states, temporal periodicity of the Hamiltonian leads to wavefunctions that are periodic in energy. Combining the two situations, a periodic excitation on a crystalline lattice induces Floquet-Bloch bands that repeat in both momentum and energy. Just as different Bloch bands hybridize and develop band

gaps at the crossing points, the crossing points between different orders of the Floquet-Bloch bands should open dynamic gaps. Being able to open up gaps in the excitation spectrum with light can potentially be used to modify the electronic and optical properties of the materials.

Prior to this work, Floquet-Bloch states had never been observed experimentally. Incoherent effects such as scattering of excited electrons as a result of direct absorption of light via inter-band electronic transitions can mask the Floquet-Bloch states. The Gedik group suppressed the incoherent effects by exciting the system with below band-gap light. They then measured the band structure of  $\text{Bi}_2\text{Se}_3$  at different times both before and after the arrival of this mid-infrared excitation pulse.

Graduate students Yihua Wang and Fahad Mahmood in the Gedik group managed to capture these time-resolved ARPES spectra at high resolution. *Figure 5* shows vertical cuts from 3D energy-momentum spectra of  $\text{Bi}_2\text{Se}_3$  obtained at several time delays  $t$  after the intense mid-infrared radiation excitation. At  $t = -500$  fs, the probe pulse is ahead of the pump pulse, and the band structure is similar to that of an unperturbed system (*Fig. 5, far left*). When the two pulses start to overlap in time ( $t = -200$  fs), duplicates of the surface Dirac cone appear above and below the original band. The energy difference between the bands equals exactly the pump photon energy. The intensity of such side bands becomes stronger while the original band gets weaker when the pump and probe pulses are completely overlapped at  $t = 0$ . The fact that the sidebands are only present during the time duration of the pump pulse suggests that it is a consequence of the coherent interaction between the mid-infrared photons and the electron system.

The band structure of these Floquet-Bloch states is composed of a manifold of Dirac cones stacked in energy. When these Dirac cones cross, depending on the momentum direction, they either cross or anti-cross (*Fig. 5, left and center*) and open up band gaps at the crossing points. When the mid-infrared pulse is circularly polarized, an additional band gap opens up at the Dirac point (*Fig. 6*), a signature of broken time-reversal symmetry on the surface. This represents a completely novel way of breaking TRS using circularly polarized light instead of magnetic field. The photo-induced gap at the Dirac point can be as big as 50 meV. To open up a similar gap using a magnetic field, 500 T would be necessary, larger than has ever been achieved on Earth! Furthermore, this gapped photo-excited state is not trivial; it is predicted to show many exotic features such as QHE.

Floquet states have previously only been observed in atomic systems. The realization of coupled photon-Dirac fermion states in three-dimensional topological insulators represents the first such observation in a solid. This has far-reaching

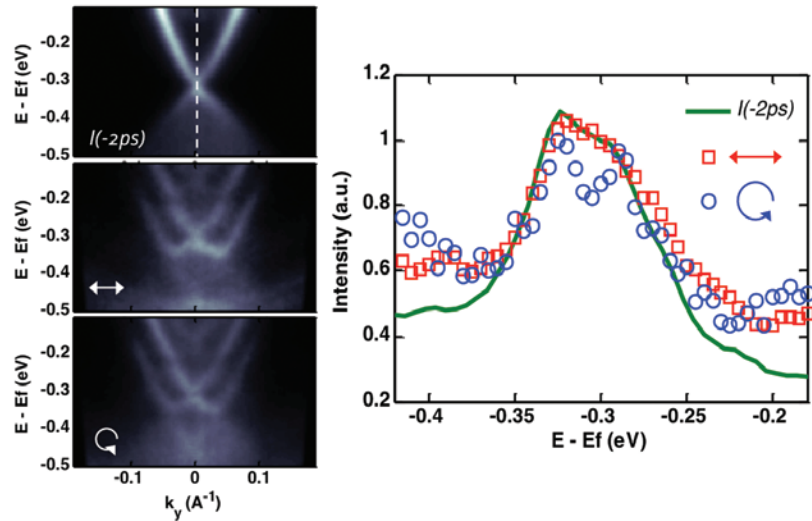


FIGURE 6

**(a) The ARPES spectra** obtained at  $t = -2$  ps (top panel), at time zero under linearly (center panel) and under circularly (bottom panel) polarized mid-infrared light excitation.

**(b) The photoemission intensity cuts along the dashed vertical line through the Dirac point.**

Around the Dirac point ( $E = -0.3$  eV), intensity profile before the excitation (green curve) agrees well with the one under linearly polarized excitation (red squares). Under circularly polarized light (blue circles), a band gap opens at the Dirac point.

implications both in terms of fundamental science and device applications. The observed topological surface states with broken time-reversal symmetry indicate the existence of the long sought-after quantum Hall phase without Landau levels induced by the light. Also, these findings open up an avenue for optical controlling and switching topological orders and provide a platform for realizing many other light-induced phases in these materials. It can eventually pave the way for using lasers to engineer materials with different properties by modifying their band structure. This has the potential of fundamentally changing how we think about making materials with novel properties. As opposed to the conventional methods (such as changing the material chemistry and applying pressure), shining light could be enough to realize a completely different quantum phase of a material. Light excitation could be quickly switched on and off in ultrafast timescales and could be spatially patterned, allowing many intriguing possibilities for designing devices.

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RAYMOND ASHOORI *The principle focus of research of the Ashoori group lies in the study of interacting electronic systems in low-dimensional structures in semiconductors, graphene, and other materials. His group uses extremely sensitive charge sensing methods to probe these systems in novel ways that give basic information on the state of the electrons. They have also used charge sensing methods to develop a pulsed electron tunneling technique that yields precise high resolution tunneling spectra for a variety of materials, including insulators.*

*A graduate of the University of California at San Diego (BA 1984) and Cornell University (PhD 1991), Ray Ashoori joined the MIT faculty in 1993, after serving as a postdoctoral member of the technical staff at AT&T Bell Laboratories. He was promoted to Associate Professor in 1998 and full Professor in July 2004.*

NUH GEDIK *Professor Gedik's research centers on developing and using advanced optical techniques for investigating ultrafast processes in solids, nanostructures and interfacial molecular assemblies. The Gedik group uses these techniques to search for answers to important problems in condensed matter physics. One primary focus is to understand strongly correlated electron systems. In these materials, the interplay between spin, charge and lattice excitations leads to fascinating properties such as high temperature superconductivity and colossal magneto-resistance. Using the experimental techniques they developed, the Gedik group studies the spatiotemporal dynamics of these excitations, with the goal of identifying the mechanisms behind the striking macroscopic behavior of these materials.*

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**PABLO JARILLO-HERRERO** *Professor Jarillo-Herrero's research interests lie in the area of experimental condensed matter physics, in particular quantum electronic transport and optoelectronics in novel low dimensional materials, such as graphene and topological insulators (TIs).*

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