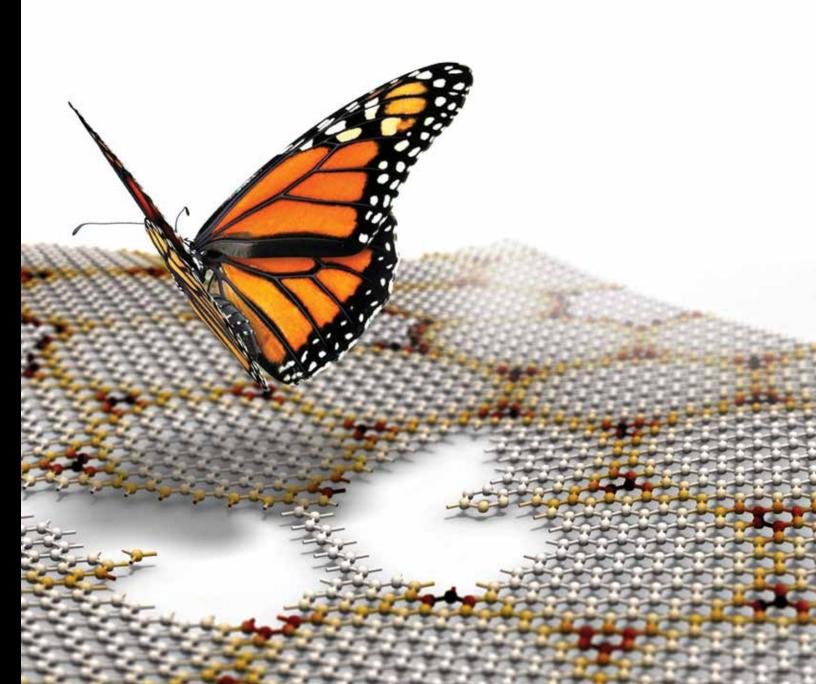
Physics in Low-Dimensional Materials



Edwin Abbot, in his 19th-century satirical novel, *Flatland: A Romance of Many Dimensions*, describes life in two dimensions. The narrator, named Square, is a knowledgeable scholar in his two-dimensional flatland.

by Prof. Philip Kim

Through the course of the novel, he realizes that many scientific mysteries in Flatland can be elegantly explained through the addition of an extra third dimension. Out of this elucidation, Square says, "Three Dimensions seems almost as visionary as Land of One or None."

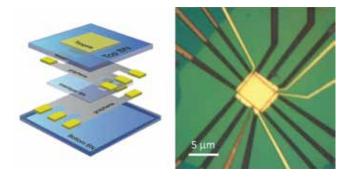
Similarly, physicists have been considering the concept of a multi-dimensional universe in order to explain the mysteries of our own three-dimensional world. The idea of a four-dimensional space-time provided the foundation for Einstein's theory of relativity. Today, in efforts to explain the underlying structure of the universe, physicists are considering as many as ten or more dimensions. However, through the study of these "extra" dimensions we might have missed the intricacy and beauty of the physics that happens at the other extreme: the reduced two- or one-dimensional worlds.

The notion of reduced dimensionality is often considered a purely mathematical idea and is seen as an abstract concept that cannot be realized in an actual physical or material world. Yet, quantum physics teaches us how to effectively describe a three-dimensional object within the confines of a reduced dimensional system. Imagine a particle confined to a box. If we start to flatten the box, quantum mechanics tells us that the energy of the spatially confined particle becomes discontinuous. The separation between energy levels increases as the box is flattened even further. Eventually, the particle's motion along the direction in which the box is flattened freezes as the discrete and quantized energy scale becomes larger than any

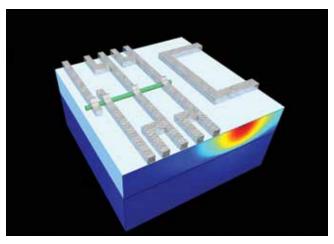
other characteristic energy in the system. Thus, in our flattened box, the particle is restricted to two dimensions—essentially an object in Abbot's Flatland, although it still exists in the three-dimensional world.

It turns out that these reduced dimensional systems are already crucial to modern electronic devices. Transistors, the core element of today's electronics, employ electrons in an effective, two-dimensional space formed at the interface between the silicon surface and its oxide layer. The reduced dimensionality of the space electrons inhabit often enables an unusual quantum mechanical effect. In particular, in a strong magnetic field and at ultra-low temperatures, the electron orbit can be completely quantized. In this case, the resistance along the direction of the current vanishes completely, indicating a nondissipating flow of electrons, just like in superconductors. Additionally, the Hall effect —characterized by the resistance measured perpendicular to the current (see details in the latter part of this article)—exhibits discrete steps with defined plateaus over large intervals of magnetic fields. This quantization is called the quantum Hall effect. Two Nobel prizes have been awarded in this subject, one for the discovery of the quantization at integer multiples of e2/h in 1985 (von Klitzing) and another for fractional multiples in 1998 (Stormer/Tsui/Laughlin). Over the years, Harvard's condensed-matter physics group has also made important contributions to the study of these beautiful quantum mechanical effects.

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Van der Waals heterostructure of graphene and hexa boron nitride to form a mesoscopic quantum device



Quantum engineered thermoelectric and thermal transport

In order to create low-dimensional semiconductor structures, physicists often borrow microfabrication technology developed over the past half century. Microfabrication, combined with nanotechnology, produces effectively 2-dimensional (2D) or even 1-dimensional (1D) semiconducting devices in the forms of the quantum wells, quantum wires, and quantum dots. Ironically, to create these tiny structures we often need to access huge facilities, properly equipped with sophisticated instruments for material manipulation, fabrication, and characterization. Harvard physicists enjoy world-class microfabrication facilities at the Center for Nanoscale Systems (CNS), which provides shared research space dedicated to the fabrication and characterization of nanometer-scaled devices. (See the sidebar, "Center for Nanoscale Systems.")

Historically, reduced dimensional semiconducting structures have been fabricated in a top-down method in which a larger system splits to create smaller ones. A completely different approach, the bottomup assembly method of nanostructures, was first used in nanoscience and technology starting in the 1990s. The bottom-up method, when applied to materials science and chemistry, assembles molecules to create larger material structures. Among these material structures, carbon-based nanostructures have attracted the most interest. The first kind of carbon nanostructure was the C_{60} "buckyball." This spherical molecule consists of 60 carbon atoms forming a cage connected via hexagonal and pentagonal rings of carbon. Richard Smalley and his colleagues, who discovered C_{60} in 1985, were awarded the Nobel Prize in Chemistry in 1996, celebrating the arrival of this new type of chemical nanostructure. Within a few

years, the buckyball was followed by carbon nanotubes, a 1D form of the tubular-shaped hexagonal carbon ring that was discovered in 1991 by Sumio Iijima. Unlike their predecessor, nanotubes immediately attracted the attention of many physicists as an idealization of quantum wires that can confine electrons in a 1D system. Nanotubes also carry the promise of many potential applications. Their excellent electronic, optical, thermal, chemical, and mechanical properties, resulting from reduced dimensionality, have inspired their use in the development of a wide variety of technologies. Interestingly, the 2D form of carbon nanostructure, graphene, was theoretically hypothesized by the Canadian physicist, Philip Wallace, more than 60 years ago. However, the actual experimental discovery was made only 10 years ago. In late 2004, Andre Geim and Konstantin Novoselov at Manchester University experimentally demonstrated the existence of 2D graphene, later winning the 2010 Nobel Prize in Physics.

Graphene is a two-dimensional, hexagonally-arranged layer of carbon that is only a single atom thick. In a way, graphite, which you can find in pencil lead, can be regarded as a three-dimensional (3D) stack of atomic graphene sheets that are held together by weak interactions, often referred to as van der Waals bonding. It is this weak van der Waals coupling between graphene layers that allows graphite to be cleaved easily. In fact, the Nobel duo at Manchester University initially used scotch tape to exfoliate a piece of graphite down to the thickness of a few atomic layers to demonstrate the existence of graphene—a procedure that fully exploited the weak interaction between graphene layers.

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It turns out that the electronic properties of graphene are exceptional, as well as unique. Although the electrons in graphene are the same electrons that we find in free space, their behavior in graphene is drastically different. The quantum mechanical description of the Bloch wave of electrons in graphene renders a new kind of quasiparticle—a concept used in condensed-matter physics to describe the alteration of the original particle's properties. Quasiparticles move like electrons that have completely lost their mass, following an analogous description of the Dirac equation. The idea of massless Dirac particles has been used to describe the relativistic quantum mechanical behaviors of spin-1/2 fermions traveling at the speed of light, independent of their energy or momentum. Similar to these relativistic particles, quasiparticles in graphene always move at a constant speed, about 1/300th the "real" speed of light. Except for the scaled down "speed of light," the quantum dynamics of graphene's quasiparticles are completely relativistic. This analogical mapping to the Dirac equation of electrons in graphene, however, can be rather intriguing since many "high energy" experiments can be realized in the setting of a condensed-matter physics laboratory. For example, the Klein tunneling of Dirac fermions, a relativistic quantum tunneling through a barrier, enables a perfect tunneling of Dirac fermions through any barrier due to the particle-antiparticle pair generated at the interface. Experimentally, Klein tunneling has already been observed in graphene devices fabricated using the microfabrication techniques discussed above.

Following the discovery of graphene, several other crystals, also one atomic layer thick, have been created in laboratories using similar experimental approaches. In fact, it turns out that nature provides us with many different "flatlands." In these layered systems, as we've seen in graphene, strong covalent chemical bonds exist within the single atomic layer, and weak van der Waals (vdW) forces hold the different layers together. The exciting news is that these vdW materials can exhibit very diverse electronic behaviors. Some of them are semiconductors with exceptional magnetic properties, some are superconductors at relatively high temperatures, and some are strongly correlated metals exhibiting exotic charge density waves. Building on graphene research, we are now exploring the new subfield of physics enabled by this emerging class of reduced dimensional material systems.

Furthermore, the recent advent of vdW material systems has also given rise to a new type of heterogeneous quantum material with atomically sharp interfaces. As discussed above, one unique feature of vdW materials is their rich functionality in 2D electronic systems.



Center for Nanoscale Systems at Harvard

Center for Nanoscale Systems

DIRECTOR: ROBERT WESTERVELT EXECUTIVE DIRECTOR: WILLIAM WILSON

The Center for Nanoscale Systems (CNS) at Harvard provides world-class facilities for users in academia and industry, supporting research that pushes the frontiers of electronics, photonics, materials science, and bioengineering.

IMAGING AND ANALYSIS

The center is equipped with sub-angstrom resolution Transmission Electron Microscope and Scanning Transmission Electron Microscopes, an automated Cryo-Bio TEM, an Atom Probe for 3D Tomography with atom identification, Scanning Electron Microscopes with Energy Dispersive Spectroscopy and Electron Backscatter Diffraction Analysis (EBSD), and sample preparation facilities.

NANOFABRICATION

A full range of nanofabrication techniques are available, including: Electron-beam Lithography; Optical Lithography; Reactive Ion Etching (RIE); Chemical Vapor Deposition (CVD); Atomic Layer Deposition (ALD); Physical Vapor Deposition (PVD); Metrology; and Wet-Processing Tools.

NANO/BIO/SOFT MATERIALS

The tools include Atomic Force Microscopy, X-ray Photoelectron Spectroscopy, X-ray Fluorescence, X-ray Computed Tomography, Fourier Transform Infrared Spectroscopy, Optical and Confocal Microscopy, and a soft lithography facility to construct Microfluidic systems.

STAFF AND RESOURCES

- CNS provides the staff and resources needed to train and assist students, postdocs, faculty, and industry investigators in the use of our advanced facilities.
- CNS offers internal and external users open access to our equipment.
- CNS provides lab courses and summer internships for undergraduates.
- CNS promotes collaborations between faculty groups and commercial firms.
- CNS hosts seminars and workshops on a broad range of topics.

The Hofstadter butterfly is a butterfly-shaped fractal energy spectrum. Fractals are infinitely repeating, self-recursive geometrical structures. They often appear in complex classical systems but rarely in the quantum mechanical world.

Capitalizing on the weak vdW interaction between two separate single-atom-thin vdW layers, we can simply stack them to form a heterogeneous blend of materials. This atomic stack can provide ample opportunities for the realization of novel collective interfacial quantum phenomena.

One interesting example of these new interfacial phenomena is our recent experimental realization of "Hofstadter's butterfly." Predicted by Douglas Hofstadter in 1976, the Hofstadter butterfly emerges when electrons are confined to a 2D sheet and subjected to both a periodic potential energy and a strong magnetic field. The Hofstadter butterfly is a butterfly-shaped fractal energy spectrum. Fractals are infinitely repeating, self-recursive geometrical structures. They often appear in complex classical systems but rarely in the quantum mechanical world. In fact, the Hofstadter butterfly was one of the first quantum fractals theoretically postulated in physics. In the past, experimental efforts to study the Hofstadter butterfly attempted to use artificially created structures to achieve the required periodic

potential energy. In our experiment, we used an effect called a moiré pattern that arises naturally when two similar atomic lattices overlap. We stacked a single atomic layer of graphene on top of a boron nitrate (BN) substrate, which has the same honeycomb atomic lattice structure as graphene, to create a vdW heterostructure with a periodic potential. (See figure page 10.) We mapped the graphene energy spectrum by measuring the electronic conductivity of the heterostructure at very low temperatures in extremely strong magnetic fields. For this experiment, we had to travel to the National High Magnetic Field Laboratory, where we could use a large magnet with immense magnetic fields—up to 35 Tesla, consuming 35 megawatts of power. Remarkably, our measurements showed the predicted fractal energy spectrum pattern, providing the strongest direct evidence to date of the Hofstadter butterfly.

From the early days of graphene and other 2D materials research, Harvard has been a powerhouse in exploring reduced-dimensional materials and their heterostructures. Recently our research has been

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further accelerated by the Center for Integrated Quantum Materials (CIQM), a science and technology facility funded by NSF. The mission of CIQM is to study extraordinary new quantum materials with striking nonconventional properties. (See the CIQM sidebar for further details.) My research group, located in the Laboratory of Integrated Science and Engineering (LISE), one of Harvard's most advanced laboratory buildings, has been an active participant in CIQM-sponsored programs. At CIQM, we are developing functional heterostructures of different 2D vdW materials to investigate the interaction between various correlated vdW layers.

In condensed-matter physics, fundamental discoveries can often be directly applied to engineering. Professor Edwin H. Hall, the discoverer of the Hall effect, joined the faculty in 1881 and served as a Professor of Physics at Harvard University from 1895 to 1921. His seminal work on measuring Hall resistance, the ratio between the transverse voltage to the longitudinal current, experimentally confirmed the existence of a magnetic force on a moving charge. It also gave rise to many practical applications. The Hall effect is one of the essential tools in characterizing semiconductors in the electronic industry. His work also provided the basis for the discovery of the integer and fractional quantum Hall effects a hundred years later, as mentioned before.

Interestingly, Professor Hall's experiment was made possible by using what was then considered a "thin" metal film to increase the current density to amplify weak signals. The thin specimen that brought him his initial success was gold leaf, which was about a micrometer thick and was probably the thinnest material that physicists could reliably access in his time. Today, we are investigating a new class of 2D materials—more than a thousand times thinner than the Hall's gold leaf—and possibly the thinnest material we can ever find. Just as the Hall's gold leaf was the lynchpin to his success, the heterogeneous vdW quantum structures have given us a glimpse of exciting new interfacial quantum effects and potential applications. These promising new findings have hinted at the prospect of "stacked Flatlands," which could yield both exciting new physics and many technological advances in the coming years.



Science & Technology Center for Integrated Quantum Materials

HARVARD UNIVERSITY

(DIRECTOR & PI: ROBERT WESTERVELT)
HOWARD UNIVERSITY (CO-PI: GARY HARRIS)
MASSACHUSETTS INSTITUTE OF TECHNOLOGY
(CO-PI: RAYMOND ASHOORI)
MUSEUM OF SCIENCE, BOSTON
(CO-PI: CAROL LYNN ALPERT)

The Science & Technology Center for Integrated Quantum Materials was created in October 2013 through a 5-year renewable grant from the National Science Foundation.

MISSION:

Transform electronics and photonics from 3D structures to 2D atomic layers, 2D electron surface states, and single-atom devices using Quantum Materials:

Atomic layer materials - Graphene, BN, Transition Metal Dichalcogenides

atomic-scale devices that are only a single atom or molecule thick

Topological Insulators

topologically protected data channels using surface edge states

Nitrogen Vacancy (NV) Centers in Diamond atomic memory sites and ultrasensitive magnetosensors

Broader Impacts:

Attract young students to careers in science and engineering. Engage public audiences in the quest for new frontiers. Commercialize science through new technologies and products.

College Network:

Share Center facilities and expertise with young students to encourage them to enter careers in science and engineering:

Bunker Hill Community College (co-PI JoDe LaVine)
Gallaudet University (co-PI Paul Sabila)
Mt. Holyoke College (co-PI Katherine Aidala)
Olin College (co-PI Rebecca Christianson)
Prince George's Community College (co-PI Scott Sinex)
Wellesley College (co-PI Robbie Berg)