Perspectives on the 2010 Nobel Prize in Physics for Graphene

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ABSTRACT The 2010 Nobel Prize in physics was awarded to Andre Geim and Konstantin Novoselov for their groundbreaking experiments regarding the two-dimensional material graphene. Some personal perspectives about this award are presented.

Graphene provides the missing piece in the carbon nanostructures puzzle. Carbon nanostructures have been at the center stage of science for at least 25 years, since the discovery of fullerenes in 1985,[1,2] which was the topic of the 1996 Nobel Prize in chemistry. Following this surge in public interest in carbon nanostructures, this research community increased dramatically after the publications by the Iijima[3] and Bethune[4] groups on single-wall carbon nanotubes in 1993. The numerous researchers working on carbon nanostructures were ideally positioned to appreciate the importance of the 2004 publication by Geim and Novoselov[5] announcing a remarkably simple method for preparing monolayer graphene and presenting groundbreaking physical measurements on graphene that the research community could easily reproduce, expand upon, and fit into the fabric of the carbon nanostructures puzzle and into the broader framework of frontier science.

> The 2004 paper by Geim and Novoselov described some highly innovative experiments that attracted a great deal of attention from the physics community.

The concept of graphene has been around for a long time, at least going back to the pioneering and highly cited 1947 publication of Wallace,[6] who developed the linear $E(k)$ dispersion for the electronic structure of graphene around the K point of the Brillouin zone. However, the interest in carbon nanostructures in these early days was minuscule, as can be seen in Figure 1, showing an ISI listing of nanostructured carbon publications per annum going back to 1960 when M.S.D. entered the field. Figure 1 shows that the entry of each new carbon nanostructure has impacted the carbon nanostructure literature, after introducing an appropriate time delay for new entrants to the field to do significant work and to get their work published. This early 1947 work by Wallace was long appreciated by entrants to the field as a construct and vision for graphene as researchers slowly entered this newly emerging research field that would perhaps someday become important.

Monolayer and few-layer graphene was, in fact, quietly synthesized in 1962 by Boehm,[7] who became better known starting in the 1970s for his work on graphite intercalation compounds. Boehm spoke about his graphene research at carbon conferences, but M.S.D., for one, was not present at his talks on graphene and did not actively follow his work on this topic. The method for graphene synthesis used by Boehm in his early work is based on the reduction of graphene oxide. Variants of this method have since been used by many investigators, such as Ruoff's group[8−10] and Kaner's group.[11−13] In the very early graphene characterization studies by Boehm, X-ray diffraction measurements of the structure of these films were made, along with efforts to measure the thickness of the layers and the specific surface areas of these films. Although characterization tools in 1962 were primitive compared to those available today, the early work by

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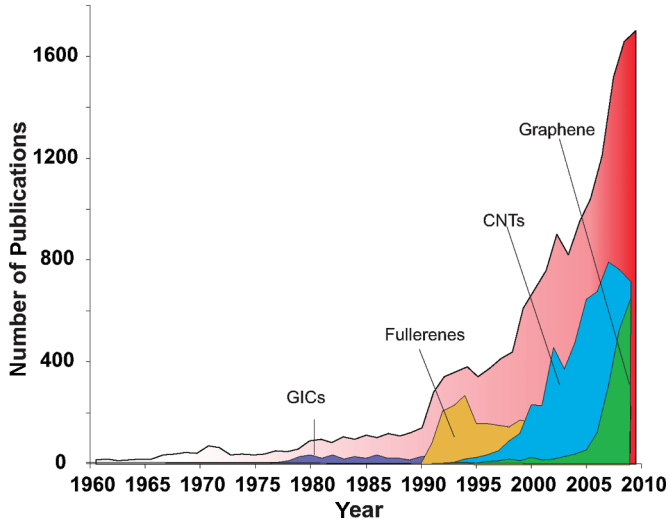

Figure 1. History of scientific publications on carbon-based materials showing the timeline for each research topic and the number of researchers in the field.

Boehm is notable for its clarity and insight.

The field of graphite intercalation compounds (GICs) became active in the 1970s and gained sufficient importance by 1977 to merit its own dedicated conference. The first conference at La Napoule in France in 1977 and subsequent GIC conferences were instrumental in building up the ranks of the nanostructured carbon community of which co-author M.S.D. was part. In graphite intercalation compounds,[14] n layers of graphene ($n = 1, 2, ...$) are sandwiched between an intercalant layer of guest atoms or molecules in a periodic two-dimensional lattice. Boehm was one of the key speakers at the first graphite intercalation compound conference and at subsequent conferences in this conference series, but M.S.D.'s recollection of his talks focused on the intercalation of graphite and not on few-layer or monolayer graphene, as discussed in his 1962 paper.[7]

Other methods for preparing monolayer and few-layer graphene were subsequently developed. Some methods are based on the mechanical exfoliation of few-layer flakes from much thicker flakes or bulk-like materials. Natural graphite flakes from Ceylon (Sri Lanka) and Ticonderoga, New York, were well-known to the carbon research community in the 1950s and two synthetic forms of graphitic material (pyrolytic graphite[15] and kish graphite) appeared on the scene at the time M.S.D. started working in carbon science. Highly oriented pyrolytic graphite (HOPG) synthesized in 1960 in the Ubbelohde Laboratory at Imperial College, London, soon became commercialized by the General Electric Company, in Schenectady, New York, and was used in commercial neutron monochromators and elsewhere, while kish graphite produced as a precipitate in high-quality steels also became available to the research community at about the same time, mostly from Japanese sources.

With HOPG and kish graphite being available to many people both for research (Figure 1) and for product development, there were numerous people in the research community using adhesive (scotch) tape to peel a few graphene layers off their HOPG and kish graphite materials. In so doing, beautiful mirror-like surfaces were produced on samples being prepared for use in connection with their optical and transport experiments, while throwing away, without further examination, the few-layer graphene materials remaining on the adhesive tape. It was, in fact, this residual material on the adhesive tape that was involved in the research for which the 2010 Nobel Prize in Physics was awarded.

Starting in 2001, other methods for preparing graphene were developed. The group of Walt de Heer,[16] coming from a surface science background, started reporting graphene-related results at carbon conferences on both the fabrication of a planar graphene layer by heating SiC to 1300 °C and on the physical properties of these layers. He showed year-by-year improvements in the quality of his material, which was prepared with the goal of one day creating carbon-based planar electronics. In 2004, the de Heer group published a detailed paper on their ultrathin epitaxial graphene,[16] and in the same paper, they highlighted the two-dimensional (2D) electron gas properties displayed by the graphene charge carriers in an electric field.

In the same 2001 time frame, Enoki and co-workers in Japan were preparing monolayer and few-layer graphene ribbons to examine their structural and magnetic properties.[17] This interest was strengthened by predictions of a high density of electronic states for zigzag graphene edges at the Fermi level,[18] in contrast to armchair graphene edges, which have a much lower density of electronic states. Interest in making appropriate samples for these experiments led to the development of a procedure to make monolayer and few-layer graphene ribbon samples from nanodiamond precursor materials.[19] These nanographite samples, as they were called in their publications, were studied for both their structural and their spectroscopic properties, and Raman spectroscopy[20,21] provided detailed polarization information that could distinguish between armchair and zigzag edges of graphene, as could also be done with high-resolution transmission electron microscopy. This work did not focus on the number of layers in their few-layer graphene ribbons but rather focused on the edge properties, which were studied by many techniques including scanning probe methods.

Shortly thereafter, Philip Kim and collaborators at Columbia University started talking about their interest in few-layer graphene at APS meetings and other conferences.[22,23] This group developed their own version of a mechanical exfoliation process for producing few-layer graphene samples. At about the same time, Geim and Novoselov used a simple mechanical exfoliation method for preparing monolayer and few-layer graphene based on

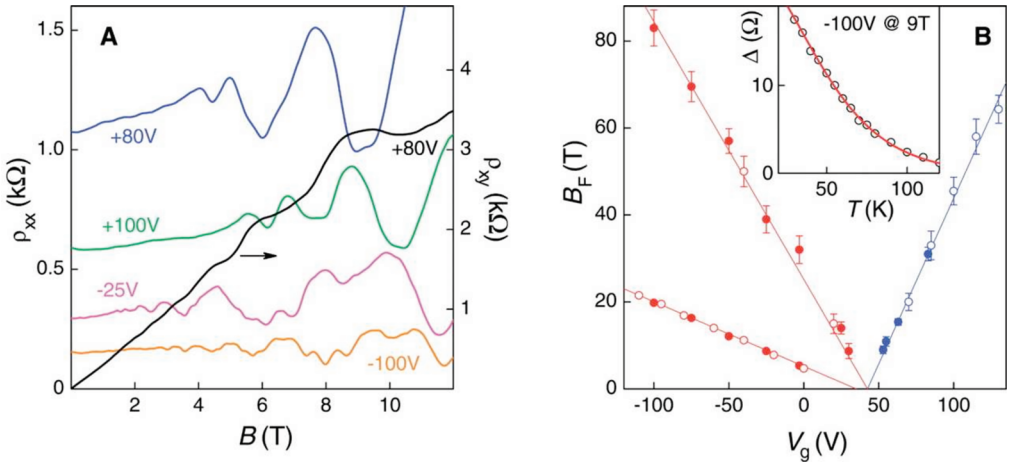

Figure 2. Dirac fermions in graphene.[5] (A) Examples of Shubnikov−de Haas oscillations observed in the resistivity ρ_{xx} and the Hall conductivity σ_{xy} of few-layer graphene at various gate voltages. (B) Dependence of the frequency of these oscillations on gate voltage for samples with band gaps of 6 meV (solid circles) and 20 meV (open circles) showing the double valley degeneracy for both electrons and holes. The inset shows data for the temperature-dependent amplitude for the Shubnikov−de Haas oscillations used to determine the cyclotron frequency $\omega_c = eB/m^*$ and the effective mass m^*.[5] Reproduced with permission from ref 5. Copyright 2004 American Association for the Advancement of Science.

adhesive tape and repeatedly transferring their graphene samples to the desired surface (often SiO_2) in order to obtain graphene of the desired graphene thickness for their monolayer and few-layer graphene samples.[5] What was special about their work was the many breakthrough physics discoveries they made in a short period of time[24] with the materials they produced and continued to perfect.

Although the mechanical exfoliation method used by Geim and Novoselov yields high-quality graphene material exhibiting very high values of the carrier mobility (*e.g.*, $\mu \sim 200\,000$ cm^2/V · s when suspended across a trench), a long-range synthesis goal has been to prepare square centimeter size monolayer films with controlled shapes, which would be especially useful for applications. Large-area monolayer graphene films can now be prepared by controlled chemical vapor deposition (CVD) techniques, as has been demonstrated in growth from a carbon-saturated copper foil.[25] By annealing this material at 1050 °C for 1 h, large-area CVD graphene growth without grain boundaries can now be achieved.[26] Although great strides have already been made in controlled graphene synthesis, there appears to be much further opportunity remaining for advances to be made in this high-profile research area.

The Breakthrough of Geim and Novoselov. From Figure 1, it can be seen that although the nanocarbon community had grown rapidly from the entry first of fullerene-based researchers starting in 1985, and later by nanotube-based researchers starting in 1992, graphene *per se* did not attract much of a following until the 2004 publication of Geim and Novoselov,[5] whereupon several important advances were made in rapid succession. One advance relates to sample preparation of interest to synthetic chemists whereby these authors developed (as described above) a simple technique that could be easily replicated by others in the preparation of high-quality monolayer and few-layer graphene flakes. This 2004 paper also described some highly innovative experiments that attracted a great deal of attention from the physics community.[5] This publication first showed a clear linear $E(k)$ dispersion relation for electrons and holes, which the authors related to massless Dirac fermions (see Figure 2), thereby attracting not only the interest of chemists, condensed matter physicists, and materials scientists but also the interest of a much broader range of scientists, including particle physicists.[5] This paper goes on to describe the temperature-dependent resistivity and the dependence of the Hall coefficient on gate voltage, emphasizing the linear $E(k)$ relation and the symmetry between electrons and holes. In this initial work, a small band overlap of 4 meV was reported, which, as the authors noted in their paper, is much smaller than the 40 meV band overlap in semimetallic bulk graphite.[27]

The paper also reports Shubnikov−de Haas oscillations in both the longitudinal and transverse resistivity of graphene in a magnetic field, thereby providing accurate values for the cyclotron frequency from which they determined the effective mass for electrons and holes as a function of energy from the Dirac point. Soon thereafter, a more detailed paper on the quantum Hall effect appeared,[24] emphasizing the special properties of the half-integral character of the quantum Hall effect in monolayer graphene and the integer quantum Hall effect in bilayer graphene,[28] which were here revealed for the first time, as shown in Figure 3. These early discoveries triggered many invited talks by Geim with standing-room-only attendance. From this great interest came a huge rush of graphene publications by the already large nanocarbon research community, as documented in Figure 1. Once launched, the Geim−Novoselov team moved forward quickly in graphene research, bringing with them a large cadre of well-informed researchers, many of them coming with a strong nanocarbon background based largely on their involvement in carbon nanotube research. (See Figure 1 where a small downturn in

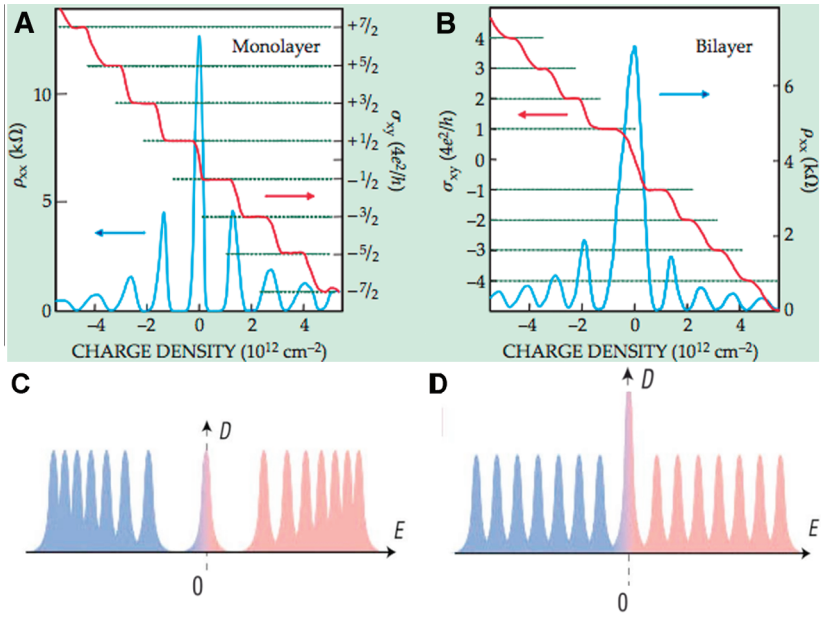

Figure 3. Quantum Hall effect for monolayer (A) and bilayer (B) graphene, showing both σ_{xy} and ρ_{xx} as a function of charge density for both electrons and holes.[29] Reproduced with permission from ref 29. Copyright 2007 American Institute of Physics. Also shown in (C) and (D) are the corresponding density of states for electrons and holes in the various Landau levels.[30] Reproduced with permission from ref 30. Copyright 2007 Nature Publishing Group.

carbon nanotube papers accompanies a rapid rise in graphene publications.) However, the graphene field also attracted many people who were new to carbon nanoscience.

In principle, it was thought in the 2004 time frame that 2D single-layer materials would not be stable below a certain thickness. In fact, before 2004, no paper had emphasized the actual observation of single layers of a 2D material, although, as summarized above, evidence had appeared for the existence of such single layers without discussion of this contradiction between observations and conversations at conferences.

In 2005, Novoselov and Geim performed comparative studies in other atomically layered 2D systems like boron nitride (BN), MoS_2, $NbSe_2$, and $Bi_2Sr_2CaCu_2O_x$.[31] All of the materials were exfoliated in the same way as had been done for graphene and were shown to be morphologically stable, although with electrical and mechanical properties much inferior to those of 2D graphene. Later, the production of monolayer graphene suspended over microfabricated trenches exhibited striking stability and opened a new channel for technological devices.[32,33]

The authors of the various early graphene papers suggested that graphene could then become the basis for the next generation of technologically important materials. Because of their special electronic and vibrational structures, single and bilayer graphene have been envisioned as a prototype material for many previously unobserved quantum mechanical phenomena, such as unconventional quantum Hall effects,[24,28,30] direct observation of Berry's phase, which had been predicted by Ando[35] in 1998, and the Klein paradox.[34] The Klein paradox says that relativistic particles can move unimpeded through high and wide potential barriers (this is one of the most exotic consequences of quantum electrodynamics). Actually, because of the chiral nature of graphene-related quasi-particles, quantum tunneling in monolayer graphene and bilayer graphene is quite anisotropic, and this anisotropic behavior allowed the Klein paradox tunneling phenomena to be tested quantitatively.[34] In the case of the quantum Hall effect, it is known that there are two distinct types of integer quantum Hall effects: (1) a more conventional effect exhibited in 2D semiconductor systems and (2) its relativistic counterpart, which takes place in graphene, where the electrons and holes mimic Dirac fermions, which carry a Berry phase of π.[24,28,30] In bilayer graphene, the Berry phase is found to equal 2π due to the parabolic dispersion of bilayer graphene, generating anomalous electrical conductivity properties in high magnetic fields.[28]

Graphene is an atomically layered material with a very low optical absorption (2.3% absorption/layer) in the visible spectrum.[5] Such flakes are thus nearly invisible under visible illumination, but they can be probed optically by placing them on a silicon substrate covered by a SiO_2 surface layer. In addition, the electronic and vibrational properties of graphene change as the number of graphene layers changes. In this context, Raman spectroscopy provides a rapid and noninvasive technique to characterize monolayer and few-layer graphene, where effects like double resonance features in the Raman spectra reveal a lot of information about the electronic and vibrational structures of these films.[36] In 2006, Novoselov, Geim, Ferrari, and co-workers[36] showed that the G′-band[37] provides an excellent spectroscopic signature to reveal the number of graphene layers contained in their few-layer exfoliated graphene flakes.[36] The G′-band feature changes its shape, width, integrated area, and frequency as the number of layers increases, reflecting changes in the graphene electronic structure. The Raman signature of monolayer graphene, having a very strong G′-band signal relative to the G-band signal, can be used to identify monolayer graphene relative to bilayer and trilayer graphene with AB interlayer stacking.[37]

Graphene has also been shown to provide excellent materials for sensor applications.[33] The best sensitivity a detection system can achieve is the detection of a single atom or molecule. What limits the sensitivity of chemical sensors is the thermal motion of charges and impurities (which act as defects). Novoselov and Geim showed that micrometer-sized monolayer graphene chemical sensors are able to detect one single molecule in the case of several different gases like NH_3, CO, NO_2, and

At the many conferences where graphene work was reported, the Geim–Novoselov team maintained their leadership position in this fast moving field, by continually generating new, exciting results. It was the high quality and impact of these publications and public presentations that made an indelible mark on the graphene field and on science more generally.

H_2O. This sensitivity is mostly due to the exceptionally low electronic noise of graphene, making graphene a powerful material for potential electronics applications such as, for example, in field effect transistors.[38,39]

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Some Perspectives. The 2010 Physics Nobel Prize in graphene will almost certainly stimulate interest and activity in graphene for the next few years, as reflected by an acceleration of the increase in the upward slope of the graphene projection of Figure 1. New ideas are now entering the field. The complexities of chirality and metallicity in single-wall nanotubes will sustain interest in the carbon nanostructure field for a while, especially if commercial applications become important. Likewise, we can expect to see increased attention given to developing graphene applications that will first come in areas related to complementary nanotube applications, such as for transparent electrical conducting films, sensors, and biomedical applications. The very high carrier mobilities that can be achieved and the complementary behaviors of electron and hole carriers are particularly attractive aspects of graphene for applications, and important and unique applications in this area are expected in the future. One area of high promise is high-frequency terahertz applications going far beyond 100 GHz, which was reported in February 2010.[40] These applications are expected to generate a new wave of applications explaining the complementary science areas. We can thus expect a high level of activity in graphene for some years into the future.[40–43]

Because the science of graphene is still under rapid development and large scale demonstrated applications are still in the future, many scientists were surprised by the awarding of the graphene Nobel Prize in Physics only six years after the groundbreaking seminal work was published in 2004. However, there are other examples in recent history when the Nobel prize committee decided to award the Nobel prize in Physics relatively soon after the groundbreaking work was published and widely disseminated.

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