The evolution of graphene-based electronic devices

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Successful isolation of single-layer graphene, the two-dimensional allotrope of carbon from graphite, has fuelled a lot of interest in exploring the feasibility of using it for fabrication of various electronic devices, particularly because of its exceptional electronic properties. Graphene is poised to save Moore’s law by acting as a successor of silicon-based electronics. This article reviews the success story of this allotrope with a focus on the structure, properties and preparation of graphene as well as its various device applications.

Keywords: electronic device; FET; field effect; graphene; nanoelectronics

1. Introduction

Graphene, a single-atom-thick sheet of graphite that combines aspects of semiconductors and metals, is a promising replacement for conventional semiconductor materials such as silicon. It consists of $sp^2$-hybridized carbon atoms arranged in a two-dimensional (2D) honeycomb crystal structure, which is also the basic constituent of other important allotropes of carbon (Figure 1), i.e. it can be stacked to form the three-dimensional (3D) allotrope graphite, rolled into one-dimensional (1D) carbon nanotubes or wrapped up into 0D fullerenes, and hence is described as the mother of all graphitic forms [1,2].

Some of the promising features of graphene [1,3–5] include:

- the very high mobility of electrons in graphene ($\sim 15,000$ cm$^2$ V$^{-1}$s$^{-1}$) compared to that in silicon ($\sim 1400$ cm$^2$ V$^{-1}$s$^{-1}$) at room temperature;
- very low resistivity ($\sim 1.0$ $\mu\Omega$ cm), which is about 35% less than the resistivity of copper;
- very large breaking strength ($\sim 40$ N m$^{-1}$), and hence stronger than diamond;
- the offer of ballistic transport.

Unlike all other known materials, graphene remains highly stable and conductive even when it is cut into ribbons a nanometer wide. The advantage of graphene over carbon nanotubes in electronics is its planar 2D structure that enables circuit design with standard lithography techniques. Hence, it can be used to make transistors at the true-nanometer scale.

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Although graphene has been a topic of study for about the last 60 years, its existence in the free state remained elusive. But, ultimately in 2004, a group of researchers led by Dr Andre Geim from the University of Manchester isolated graphene from graphite for the first time [3]. Since then, graphene has spurred great excitement in the chip research community because of its promising electrical properties and bare-minimum atomic size.

Scientists have reported using graphene to create a variety of simple electronic devices. Its importance has been duly recognized in the 2009 edition of the ITRS [6]. As exciting as the findings are, many challenges must be overcome before graphene semiconductors can be manufactured in bulk for use in consumer products.

2. History of graphene

Well before the breakthrough in 2004, there indeed were a number of earlier efforts to make very thin films of graphite. Right from the 1970s, surface scientists were growing single layers of graphite epitaxially on top of other materials like Ni, SiC, etc. [7–9]. Also, scientists prepared intercalated graphite compounds by the insertion of additional chemical species between the individual layers of graphite [10]. Micromechanically thinned graphite, first reported in 1990, could only produce films several layers thick [11,12]. There were also observations of thin graphitic films on top of graphite, e.g. an atomic force microscope tip was used for manipulating small pillars patterned into highly oriented pyrolytic graphite (HOPG) [13,14]. However, in all such cases, the layers remained in bulk or bound to the top of some substrate. The long and tenacious journey of finding a method for ‘isolating’ single-layered graphene sheets ultimately reached its goal in 2004 with the reporting of a simple yet ingenious method by Novoselov et al. along with the observation
of its various unusual properties, such as electric field effect, ballistic transport of carriers, quantum oscillations, etc. [3]. This also marked the onset of a new era of experimental physics on graphene and a rush for unearthing its various potential uses. The evolution of this wonder material from that point onwards has been tracked in the rest of this text.

3. Preparation of graphene

3.1. Mechanical exfoliation

This is the foremost method devised by the Manchester group in 2004 by which single and few layer graphene sheets are separated out of bulk graphite onto a support substrate using a micromechanical cleavage technique [3,15]. Their method involves repeated peeling of flakes out of graphite-mesas attached to a photoresist by using simple scotch tapes. The interlayer van der Waals interaction energy in graphite is about 2 eV/nm² and the required normal force to cleave off a 1 μm² graphite sheet from the top of the crystallite is ∼300 nN [16]. Such a tiny force is indeed what is achieved using the adhesive tape. Ultimately, thin flakes (of thickness < 10 nm) are left on the photoresist, which are released using acetone solution. Then, an SiO₂-covered Si wafer is put in this solution, which causes some flakes to become attached to the SiO₂ surface due to van der Waals forces.

Thus, single and few-layer graphene samples were obtained on the SiO₂ surface. It may be called a coincidence that the same SiO₂ layer helps in identification of the thin flakes atop it amid thicker flakes and other impurities. Although graphitic films thinner than 5 nm are transparent to visible light, when present on an SiO₂ layer of particular thickness above a Si wafer, they become easily observable in an optical microscope (Figure 2a) [17]. This is because the graphene layers lead to an added optical path to the reflected light, which causes some change in the interference color and gives an indication of the thickness of the layer above the oxide.

This seemingly simple technique is actually time-consuming and demands a high degree of patience and precision. Nonetheless, the technique results in crystallites that can be more than 100 μm² in size and superior in both electrical as well as structural aspects, which is mainly due to the high quality of the starting single crystalline graphite source. Further characterization of the graphene samples can be done by atomic force microscopy (Figure 2b), scanning electron microscopy (Figure 2c), Raman spectroscopy, etc. [17–20].

3.2. Alternative procedures

Mechanical exfoliation is a low-throughput procedure. Thus, the search is on for alternative methods that can produce high-quality, single-layer graphene films and also satisfy the industrial bulk processing requirements. To this end, the previously futile attempts of producing single sheets of graphene are now being revisited successfully. Even several newer methods have been reported. A few of such innovative procedures are discussed here in brief.

3.2.1. Chemically derived graphene

A solution-based process for the large-scale production of single-layer graphene was reported in 2006 [21,22]. In this technique, oxidation of graphite is first carried out, which yields graphite oxide (GO). This can be easily exfoliated mechanically to produce stable aqueous dispersions of individual sheets. Then hydrazine is added to this suspension, which removes the oxygen functionalities in GO (introduced during oxidation) producing planar single sheets of graphene.
Figure 2. Image of a big mechanically exfoliated graphite flake containing regions of different thicknesses taken using an optical microscope (a), an atomic force microscope (b), and a scanning electron microscope (c). An ultra thin graphite region (thickness below 2 nm) is highlighted by a dashed rectangle. (d) The flake is deposited on top of an SiO$_2$/Si substrate with an oxide thickness $\Delta$SiO$_2$ = 500 nm [17]. Reprinted with permission from Nano Letters, S. Roddaro, P. Pingue, V. Piazza, V. Pellegrini, and F. Beltram, The optical visibility of graphene: Interference colors of ultrathin graphite on SiO, Vol. 7, Issue 9, pp. 2707–2710. Copyright 2007 American Chemical Society.

The major advantages of producing chemically converted graphene are its low cost and large scalability. Also the sheets produced have large area (up to 20 $\mu$m $\times$ 40 $\mu$m), making them easier to process and characterize. However, the comparison of electrical properties of such sheets with those prepared by mechanical exfoliation revealed that the resistivity of the former is some two to three orders of magnitude higher than that of the latter [22]. This is due to the presence of residual point defects within the 2D structure that are introduced during oxidation.

3.2.2. Epitaxial graphene (EG)

Graphene can be grown over specially chosen support substrates. The advantage of such substrate-based synthesis techniques is their compatibility with the present CMOS technology and their scalability, which can help in realizing the dream of incorporating graphene in the mainstream electronics industry. One such method uses ultrahigh vacuum (UHV) annealing of single-crystal SiC causing its thermal reduction and eventually producing graphene as an epitaxial layer [23–25]. Silicon desorbs from SiC at about 1000°C and the carbon-enriched surface undergoes reorganization, leaving behind small islands of graphitized carbon. The process is called vacuum graphitization. The size of the film produced depends on the size of the substrate used. The face of the SiC substrate used for growing graphene, i.e. either the silicon-terminated face or carbon-terminated face has a role in determining the properties of the produced film. Recently, Emstev et al. [26] have reported that films of much better quality can be synthesized by the ex situ graphitization of Si-terminated SiC(0001) in 900 mbar argon atmosphere and a temperature of about 1500°C followed by annealing at about 1650°C [26]. The resulting wafer-scale monolayer films are found to have electronic mobility of $\sim$2000 cm$^2$/V$^{-1}$s$^{-1}$ at $T = 27$ K. Once grown,
the EG layers can be easily patterned using standard nanolithography methods. Techniques for transferring the EG films from SiC to arbitrary substrates like SiO$_2$, GaN and Al$_2$O$_3$ have also been reported [27]. It has been shown that the EG grown on SiC has electronic properties similar to those of isolated graphene [24].

The potential of SiC epitaxial graphene for large-scale integrated electronics was first proposed by a team of researchers under Dr Walt A. de Heer in 2004 [28], only a couple of months after the reporting of isolated graphene by Geim’s group.

3.2.3. Chemical vapor deposition of graphene

Low-cost synthesis of large-scale graphene films using chemical vapor deposition (CVD) on thin (less than 300 nm) metal layers (e.g. Ni, Co, Pt, Ru, etc.) is another alternative technique [29,30]. The metal film (e.g. Ni film on SiO$_2$/Si substrate) is exposed to a flow of a hydrocarbon gas (e.g. methane) at high temperatures (900–1000°C) causing carbon saturation of the metal. This is followed by rapid cooling of the sample, leading to a decrease in the solid solubility of carbon in Ni and, thus, precipitation of carbon in the form of ultrathin graphitic films (1–10 layers) over the metal surface occurs as a consequence. The deposited films can be transferred to arbitrary substrates by etching the underlying metal film, and can also be patterned using standard lithographic processes. The quality of graphene obtained by CVD is almost as high as mechanically cleaved graphene. Another option is to prepattern the metal surface producing graphene patterns of desired geometries at precise locations and hence favoring device fabrication (Figure 3).

Recently, Dr. Rodney Ruoff’s team has demonstrated the growth of graphene on copper foils (of thickness $\sim 25\mu$m) via the CVD method using a mixture of methane and hydrogen, yielding films that can be arbitrarily large in size and of high quality [31]. Primarily, because of the low solubility of carbon in copper, the growth is self-limited and automatically stops after a single graphene layer. They have also devised means of transferring the produced graphene films to arbitrary substrates using polymethyl methacrylate (PMMA). Ci et al. have demonstrated hybrid 2D structures made by merging graphene and hexagonal boron nitride (h-BN, having the same lattice structure as graphene) through CVD that has made band-gap engineering possible [32]. This is due to the fact that graphene is a conductor and h-BN is an insulator and the proportion of one to the other determines the tunable band gap.
4. Structure of graphene

Graphene is conceptually a single (one-atom thick) layer of graphite, which consists of carbon atoms arranged in a honeycomb crystal lattice. The in-plane unit cell vectors are $|a_G| = |b_G| = 2.4589$ Å (Figure 4a). Each unit cell contains two carbon atoms at locations $(0, 0)$ and $(a_G/3, 2b_G/3)$ [24]. The carbon–carbon bond length in graphene is about 0.142 nm. The carbon atoms are hybridized into an $sp^2$ configuration. Thus, there are three in-plane sigma ($\sigma$) bonds per C atom, which are extremely strong and form the basis of the extreme tensile strength of the material. Also, the partially filled $pz$ or pi ($\pi$) orbitals are perpendicular to the plane of the lattice (Figure 4b) and are responsible for providing the electrons for current conduction. Also, due to these out-of-plane $\pi$ orbitals, interactions occur between the graphene layer and the supporting substrate or between different layers of graphene, hence influencing its electronic properties.

5. Electronic properties of graphene

The tremendous potential of graphene as a material of interest in the electronic devices scenario is principally due to its unique electronic properties, which are also the most researched. Some of these are discussed here.

5.1. Large mobility, less scattering

High-quality graphene crystals have a very low density of defects, which serve as scattering centers inhibiting charge transport in most conductors. It is inferred that interactions with the underlying substrate play a major role in limiting the mobility in graphene (i.e. always below the theoretically predicted values), although effects like surface charge traps, interfacial phonons, fabrication residues, etc. may all lead to the degradation. Thus, we need to improve the substrate quality or eliminate the substrate from beneath the graphene layer to minimize substrate-induced scattering. Using the latter technique, Kim's group at Columbia measured carrier mobilities as high as $200,000$ cm$^2$V$^{-1}$s$^{-1}$ at electron densities of $\sim 2 \times 10^{11}$ cm$^{-2}$ for a single-layer mechanically exfoliated and annealed graphene sheet suspended over a trench between metallic contacts forming a device (Figure 5) suitable for performing Hall measurements [33]. Thus, electron transport in such graphene films is ballistic at sub-micron distances. The carrier mean-free path, which is highly limited by impurity scattering, has been estimated in epitaxial graphene grown on SiC to be about...
600 nm [23]. In suspended graphene, this is about 1 μm [33]. Schedin et al. have shown that the mobility remains high even at the highest carrier concentration levels induced by applied electric fields and is not much affected due to chemical doping by gaseous species [34]. They also reported that the initial undoped state of the graphene structure can be recovered easily by a gentle heating of it in vacuum. As defect scattering is the dominant scattering mechanism, mobility remains almost unchanged with temperatures in the range of 10 K to 100 K [4].

5.2. Band structure and ambipolar field effect

The charge carriers in graphene behave like mass-less Dirac fermions, relativistic particles with zero rest mass, having an effective ‘speed of light’ $v_F$ and are most effectively described by the Dirac relativistic equation rather than the usual Schrödinger equation [35].
This is verified by the observation of a relativistic analogue of the quantum Hall effect, described later. The quasiparticles in graphene exhibit a linear dispersion relation \( E(k) = \hbar k v_F \), \( v_F \) being the Fermi velocity \( \approx c/300 \), hence, behaving differently from those in metals and semiconductors where the energy spectrum can be approximated by a parabolic dispersion relation [24,36,37]. The \( k = 0 \) point is called the Dirac or charge-neutrality point (CNP). The density of states at the Fermi level is zero; hence, single-layer graphene is a semi-metal or zero-band gap semiconductor (Figure 6) with its valence and conduction bands touching at discrete points in the Brillouin zone, and the \( E-k \) dispersion relation becomes linear in the vicinity of these points.

By the electric field effect in graphene we mean the ability to modulate the current flowing through it by variation of its carrier concentration, hence, changing conductivity, \( \sigma \) (Figure 7a), due to the application of an external electric field [1,3,4,36,38]. An added advantage is the ambipolar behavior. Thus, by changing the polarity of the applied (back) gate voltage, we can change the graphene into either a completely electron (Fermi level rises above the Dirac point), or, completely hole conductor (Fermi level drops below the Dirac point) through an intrinsic state (around \( V_g = 0 \) V) when both electrons and holes are present [3], as illustrated in Figure 7b. The induced carrier concentration of electrons (or holes) is given by \( n = (\epsilon_o V_g/\varepsilon_{ox}) \propto V_g \), where \( \epsilon_{ox} \) is the permittivity of the dielectric (SiO\(_2\) in the figure), \( e \) is the electronic charge and \( t_{ox} \) the thickness of dielectric between the graphene and the Si back-gate. Corresponding variations of resistivity (\( \rho \)) and Hall coefficient (\( R_H \)) are illustrated in Figure 7b.

Graphene can also be doped due to impurities adsorbed on its surface, causing a shift of the charge-neutrality point from \( V_g = 0 \) V [39,40]. Thus, exposure to \( \text{H}_2\text{O}, \text{NO}_2 \) (electron-withdrawing species) and \( \text{NH}_3 \) (electron donor) causes p-type and n-type doping, respectively [3,34]. The zero band gap leads to a low \( I_{on}/I_{off} \) ratio and, hence, leakage currents, i.e. graphene has a minimum conductivity even when the Fermi level is aligned at the Dirac point [4,42].

A band gap of up to 400 meV can be imparted to the band structure by patterning graphene sheets into narrow ribbons or strips called graphene nanoribbons (GNR), with the energy gap scaling inversely with the ribbon width [43–45]. According to the measurements performed on such GNRs by Chen et al., as the 2D-confined electrons of graphene become further confined due to the patterning into quasi-1D ribbons, a split of the original 2D energy dispersion of graphene into a number of one-dimensional modes occurs and a finite energy gap is formed [45]. Band gaps can also appear if an electric field is applied perpendicular to bilayer graphene flakes, but the advantage of smaller leakage current seems to disappear with the increase of temperature and drain bias [46].

5.3. Quantum Hall effect (QHE)

Single-layer graphene exhibits an unconventional form of the quantized Hall conductivity, \( \sigma_{xy} = \pm 4e^2/h(N + 1/2) \), \( n = 0, 1, 2, \ldots ; N \) is the Landau level index, i.e. it forms an uninterrupted ladder of equidistant steps persisting through the Dirac point (Figure 8). This is different from both the integer QHE and fractional QHE and is named as half-integer QHE [1,3,4,47,48]. Also, in graphene the longitudinal resistance exhibits maxima rather than minima for integral values of the Landau filling factor in the Shubnikov–de Haas oscillations (SdHO) measurements. This reveals a phase shift of 180°, popularly recognized as Berry’s phase, which arises due to the zero effective mass of carriers near the Dirac points [4,49]. That the carriers have a non-zero cyclotron mass is revealed by studying the temperature dependence of the SdHO in graphene [4].
Figure 7. Ambipolar electric field effect in graphene. (a) Variation of conductivity (\(\sigma\)) with applied back-gate voltage (\(V_g\)). The inset shows the Hall-bar device with patterned graphene between metallic contacts and Si substrate (acting as back-gate) separated by an oxide-layer of thickness 300 nm. [Reprinted with permission from [38], Physics Review B, S.V. Morozov, K.S. Novoselov, F. Schedin, D. Jiang, A.A. Firsov, and A.K. Geim, *Two-dimensional electron and hole gases at the surface of graphite*, Vol. 72, p. 201401(R), 2005. Copyright 2005 The American Physical Society.] (b) Change of resistivity (\(\rho\)) with applied back-gate voltage. The low-energy spectrum, \(E(k)\), indicating the positions of the Fermi energy (\(E_F\)) for different values of \(V_g\) are shown in the three insets. [Adapted by permission from [1], Macmillan Publishers Ltd: Nature Materials, A.K. Geim and K.S. Novoselov, *The rise of graphene*, Vol. 6, pp. 183–191. Copyright 2007 Macmillan Publishers Ltd.] The inset in top-left shows the corresponding variation of Hall coefficient (\(R_H\)) and its change of sign. [Adapted by permission from [4], Macmillan Publishers Ltd: Nature, K.S. Novoselov; A.K. Geim, S.V. Morozov, D. Jiang, M.I. Katsnelson et al., *Two-dimensional gas of massless Dirac fermions in graphene*, Vol. 438, pp. 197–200. Copyright 2005 Macmillan Publishers Ltd.]

6. Devices and applications in electronics

In addition to the scalability to nanometer sizes, graphene seems to offer ballistic transport, linear current–voltage characteristics, high transconductance and huge sustainable
currents (>10^8 A/cm^2), which are all ideal for analogue/RF applications [42,50]. Although graphene transistors show a rather modest on/off resistance ratio (less than ~100 at 300 K) due to thermally excited carriers, nonetheless, such on/off ratios are considered sufficient for logic circuits [3,51]. These unusual properties make it useful for myriad applications [52].

6.1. Field-effect devices

A graphene layer can be utilized as the channel between the source and drain of a field-effect device, hence producing a graphene field-effect transistor (GFET). Configurations proposed include the back-gated pseudo-MOS structures (with the surface of graphene remaining exposed) [3,49], top-gated graphene MOS transistor (with the silicon substrate acting as a global back-gate) [42,50,53,54], dual-gated structures [55], etc. A comparison of some properties of graphene with those of a few other semiconductors commonly used in fabricating field-effect devices is provided in Table 1.

The schematic cross-section of a top-gated GFET is shown in Figure 9a. The local top-gate and global back-gate are independently used to control the carrier concentration and polarity in the graphene channel. The $I_D$–$V_{DS}$ characteristic obtained is almost linear (Figure 10a). The transconductance changes sign with increasing gate voltages due to ambipolar transport (Figure 10b). Compared to uncovered graphene, due to interaction of the top $\pi$-orbitals by van der Waals bonds to the top dielectric, some reduction in mobility and, hence, the drain current is observed [53,54]. Nonetheless, the values are far better than conventional silicon-based devices. Also, graphene devices operating in the microwave frequency range, with intrinsic current-gain decreasing with increasing frequency following the ideal $1/f$ dependence and a cutoff frequency ($f_T$) of 26 GHz have been reported [56]. Recently, the problem of low on/off current ratio has been alleviated in GFETs made with bilayer graphene [51]. Here, bilayer graphene biased with a perpendicular E-field has
Table 1. Comparison of the properties of graphene with those of some common semiconductors.

| Property                                      | Graphene | Si  | Ge  | GaAs | InAs | InP |
|-----------------------------------------------|----------|-----|-----|------|------|-----|
| Electron mobility (cm$^2$ V$^{-1}$ s$^{-1}$)@ 300 K | 200,000  | 1400| 3900| 4600 | 16,000| 2800 |
| Band gap energy, $E_g$ (eV)                   | 0        | 1.12| 0.66| 1.42 | 0.36 | 1.35 |
| Electron saturation velocity $V_{sat}$ (10$^7$ cm/s) | >5       | 1   | 0.6 | 2.2  | 4.0  | 2.2  |
| Density-of-states electron effective mass ($m^*/m_o$) | 0        | 1.08| 0.56| 0.067| 0.023| 0.077|
| Relative dielectric constant, $\varepsilon_r$ | 2.4      | 11.9| 16.0| 13.1 | 14.6 | 12.4 |
| Thermal conductivity (W m$^{-1}$ K$^{-1}$)    | 5000     | 150 | 60.2| 46   | 27   | 68   |
| Lattice constant (Å)                          | 2.46     | 5.43| 5.65| 5.65 | 6.06 | 5.87 |

Figure 9. A top-gated graphene field effect transistor on a SiO$_2$/Si substrate: (a) schematic diagram; (b) SEM image [37]. Reprinted with permission from Proceedings of the IEEE/ACM International Conference on Computer-Aided Design, K.L. Shepard, I. Meric, and P. Kim, Characterization and modeling of graphene field-effect devices, pp. 406–411, November 2008. Copyright 2008 IEEE.

demonstrated a large electrical band gap (>130 meV) and an on/off current ratio of around 100 and 2000 at room temperature and 20K respectively.

Besides mechanically exfoliated graphene, research on devices using epitaxial and CVD graphene is gradually gaining emphasis as these are scalable techniques and provide the means to reach the graphene technology goal. Multilayered epitaxial graphene on insulating SiC substrate has been used for fabricating hundreds of transistors on a single chip [57]. The world’s first RF graphene field-effect transistor has been accomplished using 1–2 layered EG on SiC [58]. Recently, IBM has reported the creation of top-gated transistors using graphene grown on the silicon face of a 2 inch thick SiC wafer that can operate at speeds of 100 GHz with an electron carrier density of about $3 \times 10^{12}$ cm$^{-2}$ and peak mobility of 1500 cm$^2$ V$^{-1}$ s$^{-1}$ at room temperature (Figure 11) [59]. This far surpasses the performance of the fastest GaAs transistors. The major innovation lies in the usage of a 10 nm thick “interfacial polymer layer” of poly-hydroxystyrene separating the graphene layer from its metal gate dielectric. The stress now is on improving the purity of graphene films, scaling down of such transistors, optimizing the device structure and finding ways of imparting a sizeable band gap for enabling its use in digital applications. Using CVD graphene on copper foils, Li, et al. have realized dual-gated FETs with Al$_2$O$_3$ as the gate...
dielectric. A mobility of about 4050 cm² V⁻¹ s⁻¹ at room temperature has been measured in such devices [31].

The use of graphene nanoribbons (GNR) in making transistors is perhaps another alternative that promises higher on/off current ratios [45]. New experiments have revealed that...
a band gap can be opened up regardless of the chiral orientation of the GNRs, i.e. there is no crystallographic directional dependence, but a strong dependence on the boundary conditions at the edges has been observed [44]. FETs produced using such ribbons have exhibited well behaved transistor action at room temperature with $I_{on}/I_{off}$ ratios ($\propto \exp[E_g/k_BT]$) greater than $10^6$. The extracted band gap values show a variation of $E_g(eV) = [0.8/\text{width(nm)}]$ [60,61]. But, due to the limitations of the present lithographic fabrication technology, the edges of the GNR produced are always somewhat rough leading to significant alteration of their electronic properties, e.g. due to increased scattering at the edges.

FET photodetectors have also been innovated utilizing the strong photoresponse near metal/graphene interfaces. When light is incident on graphene, electron–hole pairs are generated, which then get separated due to the internal electric field formed near the metal electrode/graphene interfaces (thus, no externally applied field is needed), producing a photocurrent [62]. Mueller et al. have achieved a photoresponsivity of 6.1 mA W$^{-1}$ (at 1.55 $\mu$m wavelength) and an internal quantum efficiency of 15–30% in such detectors with an achievable operational wavelength range of 300 nm to 6 $\mu$m [41,62]. These photodetectors have been successfully employed for data communication at a speed of 10 Gbit s$^{-1}$ on an optical link.

The choice of the metal and the effect of the gap between the metallic electrodes (source and drain) of a GFET have been examined by Nouchi et al. [63]. They have tested short channel GFETs with cobalt contacts and have obtained a somewhat more distorted transfer characteristic than the same transistor with conventional Cr/Au electrodes which is attributed to band alteration of the graphene channel due to the particular metal used. Superconducting transistors have been made utilizing the Josephson effect in mesoscopic junctions consisting of a graphene layer contacted by two closely spaced superconducting electrodes [64]. Thus, the supercurrent can be controlled by the gate voltage and is carried by either electrons or by holes. As reported by Liang and Dong, such superconductor–GNR–superconductor junctions (SGS) can also be used as superconducting switches [65].

6.2. $p$–$n$ Junctions

The ambipolar electric field doping in graphene has once again been utilized to make $p$–$n$ junctions. Williams et al. have used a local top-gate in combination with a global back-gate to control the carrier type and concentration in adjacent regions of the same graphene sheet [66]. A key feature of this innovation is the usage of a non-covalent functionalization layer between the graphene film and the high-$k$ dielectric (Al$_2$O$_3$) layer, which not only stops any interaction between Al$_2$O$_3$ and graphene, but also acts as a catalyst in the formation of the oxide layer above it using atomic layer deposition (ALD) technique. In addition, graphene $p$–$n$ junctions made by chemical doping have also been reported [67,68]. The added advantage here is that unlike top-gating, chemical doping does not reduce the mobility of carriers in the graphene film. Demonstrations of Klein tunneling [69], Vaselago lensing [70], etc., in graphene $p$–$n$ junctions have also been reported. Klein tunneling describes the unity probability of relativistic electrons in graphene to tunnel through any potential barrier (from the conduction band to the valence band) when it has a normal incidence on the barrier. This occurs due to the absence of backscattering coupled with a conservation of momentum in a direction parallel to the straight $p$–$n$ junction interface [71]. A graphene Vaselago lens is based on the fact that we can have a photon-like behavior of charge carriers in a graphene $p$–$n$ junction arising from the linear electron dispersion. Hence, focusing, defocusing and
total internal reflection of carriers can be achieved in such graphene-based optical analogue devices, with the Fermi energy level playing the role of the refractive index [70,72]. Huard et al. have studied the effect of metallic contacts on transport properties of graphene and have proved that charge transfer from a metal contact to graphene may lead to a p–p, n–n or p–n junction depending on the polarity of carriers in the bulk of the graphene sheet [73]. Thus, a clear difference can be observed between the conductances and, hence, the drain currents of GFETs at exactly opposite charge densities (i.e. at the same carrier densities but with opposite charge polarities).

6.3. Transparent electrodes and interconnects

Graphene is highly conductive and, being a 2D sheet of single-atom thickness, is virtually transparent (with a transparency of 97–98%), and, hence, is suitable for making flexible transparent electrodes and conductors for optoelectronic devices including touch screens, LCDs, LEDs and solar cells. It also has a higher mechanical strength than conventional transparent electrodes like the metal-alloy indium tin oxide (ITO), which is brittle, expensive (the supply of indium being scarce), has a low-throughput manufacturing process, may diffuse into other materials and is very difficult to recycle [30]. Graphene films deposited over large areas using chemical vapor deposition [74,75] or from solutions [76,77] have been used for such purposes, e.g. as an anode for application in photovoltaic devices, as window electrodes in solar cells, etc.

Multilayer graphene flakes obtained from reduced graphene oxide have been used as transparent electrodes in solar cells, but they exhibit a rather restrained performance like lower transparency (~80%), higher sheet resistance (> 5 kΩ/□), moderate power conversion efficiency (~0.4%) and a leakage current under dark conditions [78,79]. Such drawbacks can be explained by the various disadvantages associated with this graphene preparation method, as discussed earlier. On the contrary, CVD has emerged as a better method. Li et al. have shown in a recent report that graphene grown on copper foils by CVD can be transferred to desirable substrates yielding high performance large area electrodes [80]. The superior properties, such as high optical transmittance (>90%) and low sheet resistance, of such films are illustrated in Figure 12.

![Figure 12.](image-url)
Graphene films synthesized using nickel films [81] by a scalable CVD method have also yielded high performance large area cost-effective electrodes with minimal surface roughness (∼0.9 nm). They exhibited comparable power conversion efficiency as with ITO, but an outstanding capability to operate under bending conditions up to 138° in contrast to ITO-based devices which had cracks and irreversible failure under a bending of only 60°. Stanford University scientists have reported light-weight, flexible, large-area and environment-friendly organic LEDs (OLED) on flexible plastic substrates using graphene as a transparent conductor [82]. Thus, optical and electrical performance at par with ITO anodes is obtained minus its shortcomings. Recently, Matyba et al. have innovated an all-plastic (made entirely of organic materials, i.e. without any metallic electrode) solution-processed organic light-emitting electrochemical cell (OLEC) based on graphene, which promises to be an alternative to OLEDs [83].

Graphene nanoribbons (GNRs) are able to sustain current levels as high as $10^8$ A/cm$^2$ and hence, can outperform metallic conductors like copper conventionally used for on-chip interconnects [84–86]. This current-carrying capacity is about two orders of magnitude higher than copper at the same size scales and also, the problem of electromigration is absent and there is better reliability. However, low-temperature synthesis of graphene is a crucial requirement for the practical success of such interconnects. As the thermal conductivity of GNRs is also quite high ($>1$ kW m$^{-1}$ K$^{-1}$) for structures with less than 20 nm width, they can be utilized as efficient heat spreaders in integrated circuits.

### 6.4. Ultracapacitance devices

Ultracapacitors are an alternative to rechargeable batteries for the capture and storage of electrical energy. Advantages of these include larger power storage capacity, wider thermal operating range, lighter in weight, longer life, higher reliability, etc. Also, they can deliver the stored charge at much higher power ratings. The exceptionally high surface area to mass ratio of graphene (2630 m$^2$/g) makes it an ideal candidate for the conductive plates of ultracapacitor cells with a large energy-storage density. Such ultracapacitors are based on electrochemical double layer capacitance (EDLC) concept and their specific capacitance has been measured to be 135 and 99 F/g in aqueous and organic electrolytes, respectively [87]. The values are comparable to those of existing ultracapacitors and the chance of graphene beating them is quite bright. Also, hybrid films of graphene and carbon nanotubes have been used for realizing supercapacitors [88]. This concept can ultimately lead to the realization of high performance, electrical energy storage devices, which are in great demand in present times.

### 6.5. Chemical and biological sensors

Graphene is an ideal material for making sensors. It has been shown by Schedin et al. that even a single molecule adsorbed on its surface can cause a local change in the carrier density and hence, the resistance, thus imparting a high sensitivity for such sensors. This is accentuated by features like large surface area, atomic-thickness, high electrical conductivity, low electronic noise and a structure which is easily accessible for adsorption of various molecules for their detection [34,89]. Researchers have demonstrated good sensitivity for chemicals like NH$_3$, NO$_2$, dinitrotoluene, etc. [90] as well as biocellular and biomolecular sensors [91] using chemically modified graphene (CMG) nanostructures.
Zhang et al. have demonstrated that the sensitivity of these sensors can be immensely increased by using graphene doped with particular species like phosphorus, boron, nitrogen etc. or due to the presence of defects [92]. Thus, for example N-doped graphene shows weak interactions with CO, NO and NH₃, but strong binding with NO₂, hence, promising a good selectivity of NO₂ for such a sensor. Similarly, Ao et al. have theoretically shown that aluminum-doped graphene strongly chemisorbs CO molecules due to the formation of Al–CO bonds, thus Al-doped graphene is a potential candidate for the detection of CO [93]. Recently, scientists have reported sensors using suspended graphene (in an aqueous solution), which provide reduced noise levels due to the absence of interactions with the substrate [94]. A good review of graphene-based sensors can be found in the article by Shao et al. [95].

6.6. Nanoelectromechanical systems (NEMS)

The potential for graphene being used as the base material for nanoelectromechanical systems such as resonators, pressure sensors, mass sensors, etc., is also considerable. This is due to its exceptional mechanical properties, i.e. very light but extremely stiff at the same time. Mechanically cleaved graphene sheets placed over trenches in a SiO₂ substrate and provided with electrical contacts were used as nanoscale resonators by Bunch et al. The actuation is by electrical or optical means and the measured room-temperature charge sensitivity is \(\sim 8 \times 10^{-4}\) electrons per root Hz [96]. Such graphene-based nanomechanical resonators offer incredible features like low inertial mass, ultra-high operating frequencies (up to \(10^9\) Hz), excellent quality factors (up to 4000) and extreme sensitivities (Figure 13) [96,97]. Recently, it has been theoretically proved that armchair-edge GNRs are more suitable for MEMS/NEMS than the zigzag-edged ones as the former suffer changes in the electronic character owing to applied strain deformations [98]. Researchers have also revealed graphene-based electromechanical switches, which can be used at quite low bias voltages (<5 V) and have better stiction qualities, hence promising a low power consumption and better reliability [99].

6.7. Non-volatile memories

Field-induced reversible chemical modification of the crystalline structure of graphene led to the demonstration of a new switching effect in graphene field-effect devices by

![Figure 13. Nanomechanical drum-resonators using few-monolayer graphene oxide films [97]. Reprinted with permission from Nano Letters, J.T. Robinson, M. Zalalutdinov, J.W. Baldwin, E.S. Snow, Z. Wei, P. Sheehan, and B.H. Houston, Wafer-scale reduced graphene oxide films for nanomechanical devices, Vol. 8, Issue 10, pp. 3441–3445. Copyright 2008 American Chemical Society.](image-url)
Echtermeyer et al. The modification of the channel resistivity occurs due to the forma-
tion of insulating graphene derivatives like graphane (having $H^+$ ions attached), graphene
oxide (having $OH^-$ ions attached), etc., depending on the presence of other molecules like
water in the ambient. The initial structure can be restored by applying an electric field of
opposite polarity. On/off ratios greater than six orders of magnitude have been measured
in such devices [100]. These reversible switches can be applied for design of non-volatile
memories.

Li et al. have reported a solid-state memory using nanocables with a silicon dioxide
core and a shell of stacked sheets of graphene which possesses stable, rewritable,
non-volatile and non-destructive properties with on/off ratios of up to $10^7$ and switching
times of up to 1 $\mu$s [101]. It has the potential of increasing the amount of storage in
a 2D array by a factor of five, as individual graphitic thin film sheets could be made
much thinner compared to the circuitry in today’s flash memory chips. Ferroelectric FET
(FeFET) memory devices fabricated using graphene (or FLG) and ferroelectric thin film
have demonstrated reversible non-volatile switching between a high resistance state and
a low resistance state realized by controlling the electrical doping level in the graphene
film hysteretically, which is caused by a hysteretic switching of the polarization in the
ferroelectric film using gate voltage sweep [102]. Nonetheless, the bistability and retention
properties are highly sensitive to the FLG thickness, which is attributed to a charge
screening effect in the FLG films [103].

6.8. Frequency multipliers
By intelligent use of the ambipolar V-shaped current–voltage ($I_{DS}$ vs. $V_{GS}$) characteristic
of a GFET (Figure 14b), which is approximately identical to an ideal full-wave rectifier
characteristic (Figure 14a), design of signal-rectifiers and frequency multipliers have been
proposed for use in radio-frequency (RF) and mixed-signal applications by researchers at
MIT [104]. The signal to be full-wave rectified is required to be applied to the gate of
the GFET in common-source configuration with the gate being biased at the minimum
conduction point of the GFET $I_{DS}$–$V_{GS}$ plot, i.e. at $I_{DS\text{min}}, V_{GS\text{min}}$. The rectified output is
obtained at the drain terminal. This is due to the fact that electrons and holes conduct in
alternate half-cycles producing an output signal at the drain, whose fundamental frequency
is twice that of the input. Using this setup they were able to convert a 10 kHz input signal
to a 20 kHz output with high spectrum purity ($\sim94\%$) and low noise in the absence of
any filtering element. Although such graphene devices are predicted to open up a range
of new applications, such as in communication, radio-astronomy, THz-sensing, etc., their
practical use can be limited by a negligible voltage-gain, with the amplitude of the output

![Figure 14](image_url)

Figure 14. (a) Ideal full-wave rectifier characteristic and (b) piecewise linear approximation of a
GFET $I_{DS}$ vs. $V_{GS}$ characteristic.
signal being about 40 times less than that of the input signal. Besides, the operational frequencies demonstrated have not yet been more than a few MHz. Recently, a top-gated GFET based frequency doubler has been reported where the gain has been increased by about 10 times compared to that of the previous back-gated GFET based devices [105].

6.9. Digital integrated circuits

The realization of four basic logic-gates, NAND, XOR, OR and NOT, out of just a single graphene transistor has been reported by Sordan et al. [106]. This was done by carefully using the region around the maxima of the transfer characteristic \( R \) versus \( V_G \) of a GFET (similar to Figure 7b). The output resistance \( R \) was set at particular values by proper adjustment of the gate-voltage for corresponding logic-gate realizations. Complementary inverters have been obtained using p-type and n-type GFETs realized on a single graphene layer [107].

Recently, Harada et al. have realized a digital phase modulator using a single GFET inverter with top and back-gates where the polarity of the transistor can be switched by switching the voltage applied to the back-gate [108]. Logic gates based on graphene p–n junctions whose logic functionality can be dynamically reconfigured by adjusting the control gate voltages have been designed by Tanachutiwat et al. [109]. This utilizes the fact that a graphene p–n interface may be made to reflect away most of the incident carriers by a proper selection of gate geometry and bias voltages, whereas current flows when we have a p–p or n–n interface. All these seem to have initiated a new era of graphene-based digital ICs.

7. Future challenges

Graphene is justifiably called ‘a material for all seasons’. With its outstanding physical, chemical, electrical, optical and structural properties, graphene is being studied worldwide for electronics, sensors, displays, solar cells, electromechanical systems and energy storage devices. It has the ability to facilitate terahertz computing at processor speeds that can be up to 1000 times faster than that with silicon. It can also serve as a potential candidate for the fabrication of ‘beyond-CMOS’ devices [6]. Graphene-related research has now assumed an overwhelming pace since it was first isolated in 2004. However, some challenges still need to be addressed before graphene-based electronic devices can really taste commercial success:

1. fast and reliable methods for producing high-quality, large-sized graphene for device applications;
2. methods for precise patterning of nanostructures to achieve desirable electronic characteristics;
3. the need for other device materials, e.g. suitable dielectrics;
4. device fabrication and architecture optimization;
5. application of computer aided design (CAD).

It can be concluded that the roadblocks currently faced are bound to be circumvented in the near future with an active collaboration among the various scientific communities researching in graphene.
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