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DISCOVERY, PHYSICAL PROPERTIES, SYNTHESIS AND APPLICATION OF GRAPHENE

BY

ABREHAM TESHOME

**A GRADUATE PROJECT SUBMITTED TO THE SCHOOL OF
GRATUATE STUDIES
IN PARTIAL FULFILMENT OF THE REQUIREMENTS FOR THE
DEGREE OF MASTERS OF SCIENCE IN PHYSICS
AT
ADDIS ABABA UNIVERSITY
ADDIS ABABA, ETHIOPIA
MARCH, 2015**

ADVISOR: PROF. VADIM N. MAL'NEV

CO-ADVISOR: Dr. TESHOME SENBETA

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ADDIS ABABA UNIVERSITY

COLLEGE OF NATURAL SCIENCES

DEPARTMENT OF PHYSICS

The undersigned hereby certify that they have read and recommend to the School of Graduate Studies for acceptance a graduate project entitled "**DISCOVERY, PHYSICAL PROPERTIES, SYNTHESIS AND APPLICATION OF GRAPHENE"** by **ABREHAM TESHOME** in partial fulfillment of the requirements for the degree of Master of Science in Physics.

Dated:______________________

Supervisor: Name ______________________ Signature ____________

Examiners: Name ______________________ Signature ____________

Name ______________________ Signature ____________

ABSTRACT

Among systems with only carbon atoms, like diamond and graphite, graphene is the world's first two dimensional allotrope of carbon. It is a thin layer of pure carbon; it is tightly packed layer of carbon atoms that are bonded together in a hexagonal honeycomb lattice. In more complex terms, it is an allotrope of carbon in the structure of a plane of sp^2 *bonded atoms with a molecular bond length of 0.142 nanometres.* Layers of graphene stacked on top of each other form graphite*, with an interplanar spacing of 0.335 nanometers. Charge carriers in graphene are discribed by the Dirac-like equation and have zero effective mass. It is the thinnest compound known to man at one atom thick, the lightest material known, the strongest compound discovered, the best conductor of heat at room temperature and also the best conductor of electricity known.* Other notable properties of graphene *are its unique levels of light absorption at 2.3% of white light. There are probably a dozen methods being used and developed to prepare graphene of various dimensions, shapes and quality. The mobility of graphene is very high which makes the material very interesting for electronic high frequency application. In the near future graphene used on commercial scale on optoelectronics; specifially touch screens, liquid crystal displays (LCD) and organic light emitting diodes (OLEDs) for smart phones, tablet and desktop computers and televisions.*

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First I would like to thank the Almighty God for His endless grace and blessing on me during all these months here at AAU and all my life.

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Abreham Teshome

March, 2015

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INTRODUCTION

Carbon is one of the most versatile chemical elements. Because it can form single, double and triple bonds, it forms thousands of chemical compounds, and has numerous elemental structures, or allotropes. The most common allotropes of carbon are diamond and graphite. Diamond consists of carbon atoms single-bonded to four other carbon atoms producing a tetrahedral crystal lattice. Its structure leads to its extreme hardness and thermal conductivity, but diamond is a very poor electrical conductor. In contrast, graphite consists of stacked layers of carbon sheets within an individual carbon sheet, known as graphene 1 .

"Graphene" is a combination of graphite and the suffix -ene, named by Hanns-Peter Boehm, who described single-layer carbon foils in 1962 1 . The term graphene first appeared in 1987 1 to describe single sheets of graphite as a constituent of graphite intercalation compounds (GICs); conceptually a GIC is a crystalline salt of the intercalant and graphene.

Graphene is pure carbon in the form of a very thin, nearly transparent sheet, one atom thick single layer of carbon packed in a hexagonal (honeycomb) lattice, with a carbon-carbon distance of 0.142nm. It is remarkably strong for its very low weight (100 times stronger than steel) and it conducts heat and electricity with great efficiency. Because it is virtually two-dimensional, it interacts oddly with light and with other materials 2 .

Technically, graphene is a crystalline allotrope of carbon with 2-dimensional properties. In graphene, carbon atoms are densely packed in a regular sp^2-bonded atomic-scale chicken wire (hexagonal) pattern. Graphene can be described as a one-atom thick layer of graphite. It is the basic structural element of other allotropes, including graphite, charcoal, carbon nanotubes and fullerenes. It can also be considered as an indefinitely large aromatic molecule, the limiting case of the family of flat polycyclic aromatic hydrocarbons 1 .

Graphene, the world's first 2D material, has many extraordinary properties and characteristics which are either unique or surpass those of other materials. It possesses superior mechanical stiffness, strength and elasticity, electrical and thermal conductivity. It is optically active, chemically inert and impermeable to gases. The possession of all of these properties in a single material makes graphene a potentially disruptive technology in sectors like optoelectronics, flexible electronics, bioelectric devices, energy storage and ultra filtration 2 .

CHAPTER ONE

DISCOVERY OF GRAPHENE

More than 70 years ago, Landau and Peierls argued that strictly two-dimensional (2D) crystals were thermodynamically unstable and could not exist 2 . Their theory pointed out that a divergent contribution of thermal fluctuations in low-dimensional crystal lattices should lead to such displacements of atoms that they become comparable to inter atomic distances at any finite temperature 2 . The argument was later extended by Mermin 3 and is strongly supported by a whole omnibus of experimental observations. Indeed, the melting temperature of thin films rapidly decreases with decreasing thickness, and they become unstable at a thickness of dozens of atomic layers 4,5 . For this reason, atomic monolayers have so far been known only as an integral part of larger 3D structures, usually grown epitaxially on top of monocrystals with matching crystal lattices 4,5 . Without such a 3D base, 2D materials were presumed not to exist until 2004, when the common wisdom was flaunted by the experimental discovery of graphene and other free-standing 2D atomic crystals 6 .

Graphene, though only recently confirmed experimentally, has been discussed in conjunction with graphite for many years. Many of its properties had long been studied in conjunction with the properties of graphite, including its band structure. Graphene as a free substance was largely ignored as a purely academic substance because it was accepted that thermodynamic stresses prevented the existence of any free one- or two-dimensional crystals 4 . Additionally, there had been previous attempts to achieve two-dimensional crystals, but in all cases, it was confirmed that reducing the thickness made the crystals melt at increasingly low temperatures and it was agreed that two dimensional crystals were too unstable to exist in a free state. A possible explanation for the disparity between theory predicting the non-existence of two-dimensional crystals and their experimental confirmation may be that the graphene monolayers are only approximately two-dimensional and owe some of their stability to rippling perpendicular to the plane 6 .

Graphene had already been studied theoretically in 1947 by P.R. Wallace 7 as a text book example for calculations in solid state physics. He predicted the electronic structure and noted the linear dispersion relation. The wave equation for excitations was written down by J.W. McClure 8 already in 1956, and the similarity to the Dirac equation was discussed by G.W. Semenoff in 1984 9 .

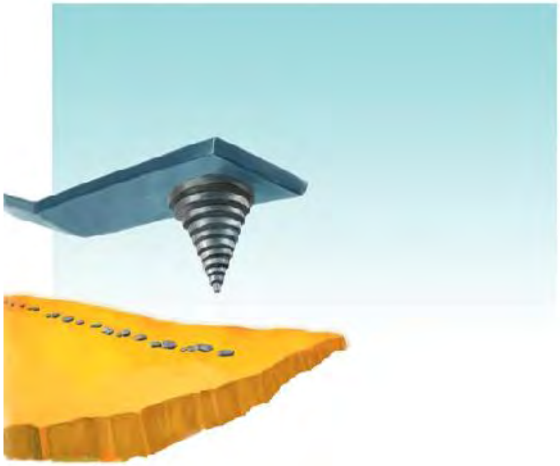
Although graphene is the building block of graphite, a material studied for many years because of its interesting mechanical properties, it is only recently [10] that a free-standing stable atomic monolayer of carbon was observed experimentally, proving wrong the theories of Landau and Mermin [11,12]. Despite all the skepticism numerous attempts to make graphene took place over the years.

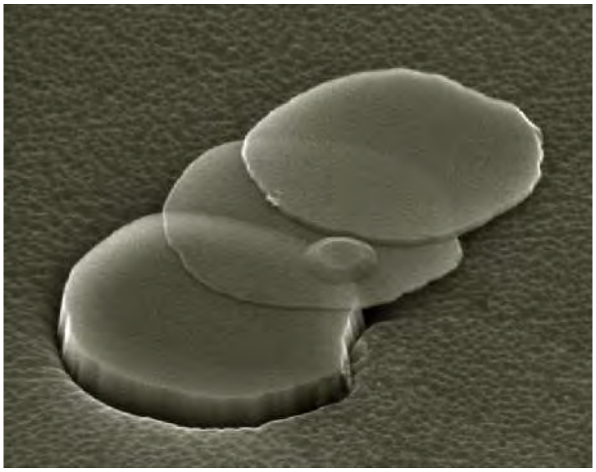
Graphene comes from graphite the “lead’’ a pencil: a kind of pure carbon formed from flat, stacked layers of atom. The tiered structure of graphite was discerned centuries ago, and so it was natural for physicists and material scientists to try splitting the mineral into its constituent sheets. Graphene is the name given to one such sheet. It is made up entirely of carbon atoms bound together in a network of repeating hexagons within a single plane just one atom thick 13 .

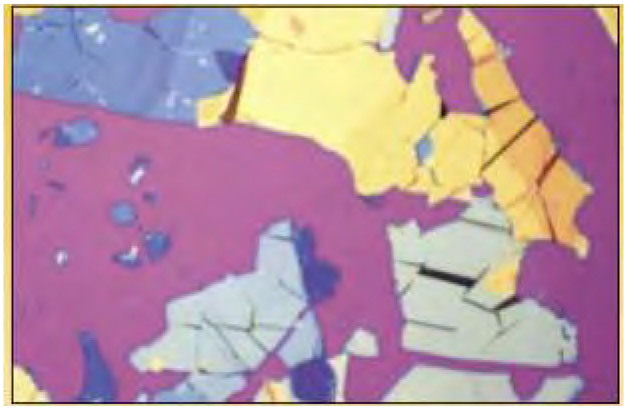
For years, however, all attempts to make graphene ended in failure. The most popular early approach was to insert various molecules between the atomic planes of graphite to wedge the planes apart. Although graphene layers almost certainly detached from the graphite at some transient stage of the process, they were never identified as such. Instead the final product usually emerged as slurry of graphitic particles. The early interest in chemical exfoliation faded away 13 .

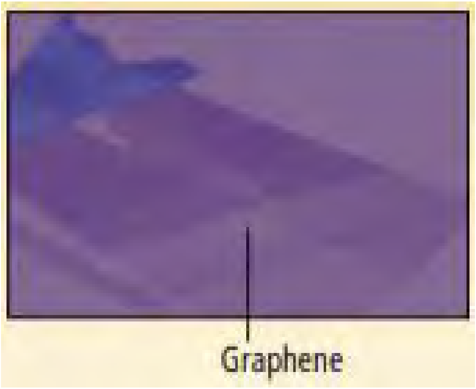
Soon thereafter experimenters attempted a more direct approach. They split graphite crystals into progressively thinner wafers by scraping or rubbing them against another surface. In spite of its crudeness, the technique, known as micromechanical cleavage, worked surprisingly well. Investigators managed to peel off graphite films made up fewer than 100 atomic planes. German physicists at Rheinisch-Westfälische Technische Hochschule (RWTH) Aachen University had isolated graphite films thin enough to be optically transparent by 1990 13 .

A decade later Kim, working with Yuanbo Zhang, then a graduate student at Columbia University, refined the micromechanical cleavage method to create a high-tech version of the pencil—a “nanopencil,” of course. “Writing” with the








Graphene

CHAPTER TWO

GRAPHENE

Carbon, the elementary constituent of graphene and graphite, is the 6th element of the periodic table. Its atom is, therefore, built from 6 protons, a neutrons, and 6 electrons, where A = 6 and 7 yield the stable isotopes ^{12}C and ^{13}C, respectively, and A = 8 characterizes the radioactive isotope ^{14}C. The isotope ^{12}C, with a nuclear spin I = 0, is the most common one in nature with 99% of all carbon atoms, whereas only 1% are ^{13}C with nuclear spin $I = \frac{1}{2}$. Carbon is the elementary building block of all organic molecules and, therefore, responsible for life on Earth 14 .

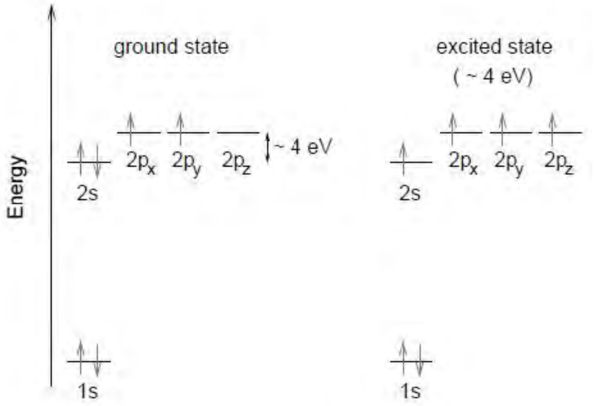
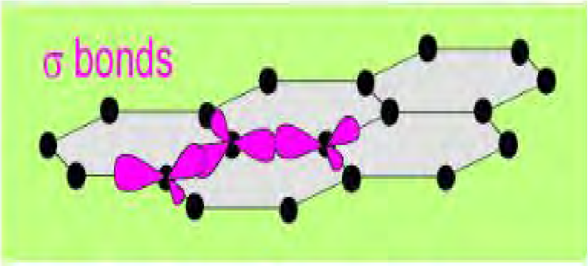
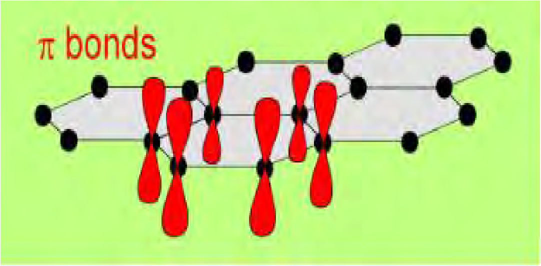


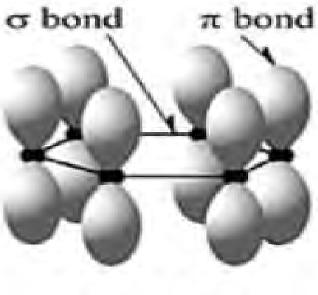
Figure 2.1 Electronic configurations for carbon in the ground state (left) and in the excited state (right) [54].

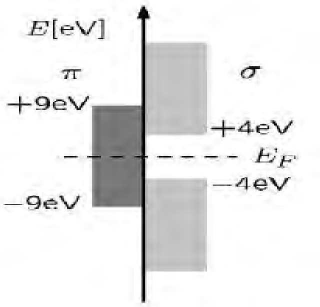
In the atomic ground state, the 6 electrons are in the configuration $1s^2 2s^2 2p^2$, i.e. 2 electrons fill the inner shell 1s, which is close to the nucleus and which is irrelevant for chemical reactions, whereas 4 electrons occupy the outer shell of 2s and 2p orbitals. Because the 2p orbitals ($2p_x$, $2p_y$, and $2p_z$) are roughly 4eV

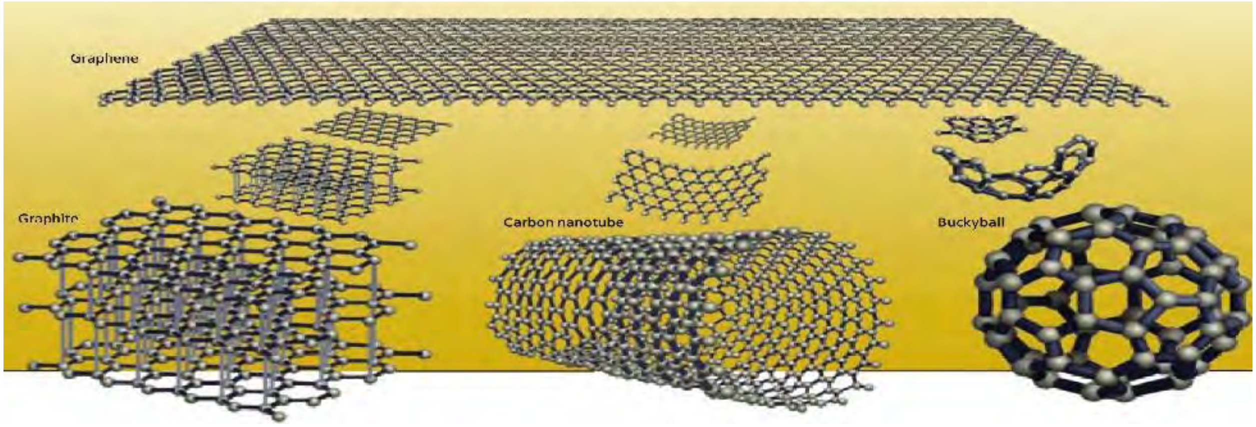




p_z






Graphene
Graphite
Carbon nanotube
Buckyball

When atoms form molecules or solids the total energy decreases due to overlap of the electron wave functions at various sites and formation of molecular orbitals (in molecules), or energy bands (in solids); for a compact introduction to chemical bonding in solids. This energy gain can be sufficient to provide the energy 16 which is necessary to promote a 2s electron into a 2p state in the carbon atom. In order to maximize the energy gained during the formation of a covalent bond, the overlap of the wave functions with those at neighboring atoms should also be maximal. This is possible if the neighboring atoms are situated in such directions from the central atoms that the atomic wave functions take on maximum values. The larger these values are the stronger the bond is 2 .

Graphene has a honeycomb crystal lattice as shown in Fig.2.5 (a). The Bravais lattice is triangular, with the lattice vectors

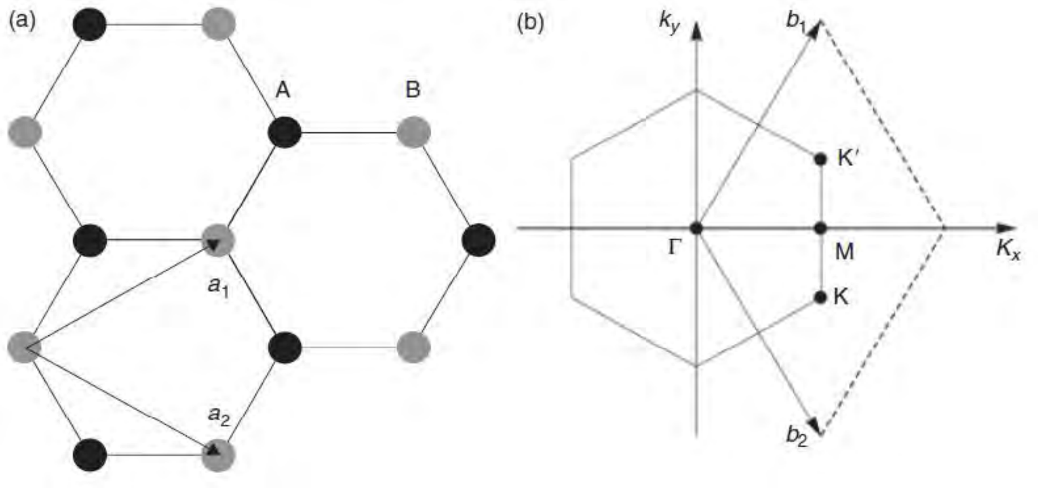


Figure2.5 A honeycomb lattice, subblattices A and B are shown as black and grey. (b) Reciprocal lattice vectors and some special points in the Brillouin zone 14

$$\vec{a}_1 = \frac{a}{2}\ 3, \sqrt{3} \qquad \vec{a}_2 = \frac{a}{2}\ 3, -\sqrt{3} \tag{2.1}$$

where a≈1.42A^0 is the nearest-neighbour distance. It corresponds to the so-called conjugated carbon carbon bond (like in benzene) intermediate between a single bond and a double bond, with lengths r_1≈1.54A^0 and r_2≈1.31A^0 , respectively 17 . The honeycomb lattice contains two atoms per elementary cell. They belong to two sublattices, A and B, each atom from sublattice A being surrounded by three atoms from sublattice B, and vice versa (a bipartite lattice) 17 . The nearest neighbour vectors are

$$\bar{\delta}_1 = \frac{a}{2}\ 3,\sqrt{3} \quad , \quad \bar{\delta}_2 = \frac{a}{2}\ 3,-\sqrt{3} \quad , \quad \bar{\delta}_3 = a\ -1,0 \qquad (2.2)$$

The reciprocal lattice is also triangular, with the lattice vectors

$$\vec{b}_1 = \frac{2\pi}{3a}\ 1,\sqrt{3} \qquad , \quad \vec{b}_2 = \frac{2\pi}{3a}\ 1,-\sqrt{3} \qquad (2.3)$$

The Brillouin zone is presented in Fig.2.5 (b) Special high-symmetry points K, K' and M are shown there, with the wave vectors 14

$$\vec{K}' = \frac{2\pi}{3a},\frac{2\pi}{3\sqrt{3}a} \qquad \vec{K} = \frac{2\pi}{3a},-\frac{2\pi}{3\sqrt{3}a} \qquad \vec{M} = \frac{2\pi}{3a},0 \qquad (2.4)$$

The sp^2 hybridized states (σ states) form occupied and empty bands with a huge gap, whereas π states form a single band, with a conical self-crossing point in K (the same point, by symmetry, exists also in K'). This conical point is a characteristic of the peculiar electronic structure of graphene and the origin of its unique electronic properties 17,18,19 .

Graphene is a single layer of carbon atoms densely packed in a honeycomb lattice, or it can be viewed as an individual atomic plane pulled out of bulk graphite. From the point of view of its electronic properties, graphene is a two-dimensional zero-gap semiconductor with the energy spectrum and its low-energy quasiparticles are formally described by the Dirac-like Hamiltonian

$$H_0 = V_F \vec{\sigma}.\vec{P} \qquad \text{but} \vec{P} = -i\hbar\nabla \tag{2.5}$$

$$H_0 = -i\hbar V_F \sigma \nabla \tag{2.6}$$

where V_F is the Fermi velocity and $\sigma = (\sigma_x, \sigma_y)$ are the Pauli matrices given by

$$\sigma_x = \begin{matrix} 0 & 1 \\ 1 & 0 \end{matrix}, \qquad \sigma_y = \begin{matrix} 0 & -i \\ i & 0 \end{matrix} \tag{2.7}$$

$$\widehat{H}_0 \psi = E\psi \tag{2.8}$$

$$\upsilon_F \;\; \sigma_x \widehat{P}_x + \sigma_y \widehat{P}_y \quad \begin{matrix} \psi_1 \\ \psi_2 \end{matrix} = E \;\; \begin{matrix} \psi_1 \\ \psi_2 \end{matrix}$$

$$\begin{matrix} 0 & \widehat{P}_x - i\widehat{P}_y \\ \widehat{P}_x + i\widehat{P}_y & 0 \end{matrix} \quad \begin{matrix} \psi_1 \\ \psi_2 \end{matrix} = \frac{\mathrm{E}}{\mathrm{v_F}} \;\; \begin{matrix} \psi_1 \\ \psi_2 \end{matrix}$$

$$\widehat{P}_x - i\widehat{P}_y \;\; \psi_2 = \;\; \frac{\upsilon_F}{E} \;\; \psi_1 \tag{2.9}$$

$$\widehat{P}_x + i\widehat{P}_y \;\; \psi_1 \;\; = \;\; \frac{E}{\nu_F} \;\; \psi_2 \tag{2.10}$$

Solving (2.10) for $\psi_2$and substituting in (2.9), we get

$$\psi_2 = \frac{\upsilon_F}{E} \;\; \widehat{P}_x + i\widehat{P}_y \;\; \psi_1 \tag{2.11}$$

$$\widehat{P}_x - i\widehat{P}_y \quad \widehat{P}_x + i\widehat{P}_y \;\; \psi_1 \;\; = \;\; \frac{E}{\nu_F}^{\;\;2} \psi_1$$

$$\widehat{P}_x^{\;2} + \widehat{P}_y^{\;2} \;\; \psi_1 = \;\; \frac{E}{\nu_F}^{\;\;2} \psi_1 \tag{2.12}$$

Let us seek a solution of equation (2.12) in the form $\psi = Ae^{i \;\; k_x.x_{+k_y.y}}$

$$\hbar^2\ k_x{}^2 + k_y{}^2\ =\ \frac{E}{\nu_F}\ ^2 \qquad (2.13)$$

$$E = \sqrt{\hbar^2 k^2} \upsilon_F \qquad (2.14)$$

$$E = \pm \hbar k \nu_F \qquad (2.15)$$

$$E = \pm P \nu_F \qquad (2.16)$$

where $P = \hbar k$ is momentum and ν_F is the velocity of electrons in graphene, known as Fermi velocity.

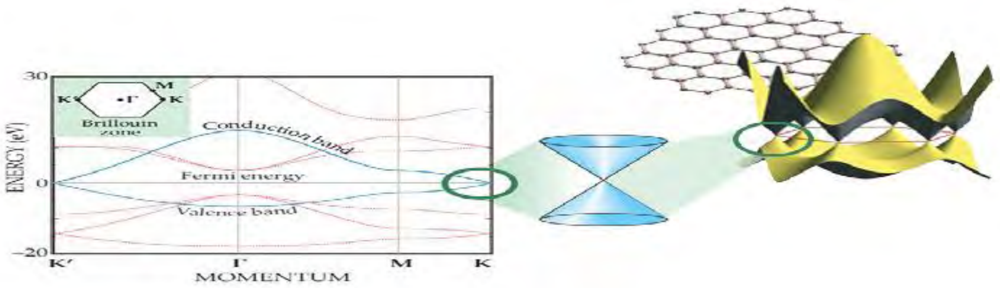
Using (2.15) we can find the velocity $\nu\ k$

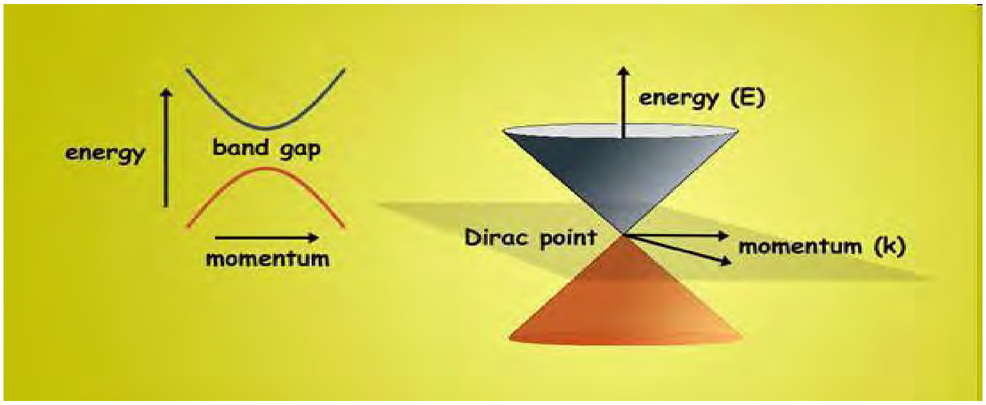
$$\nu\ k\ = \frac{1}{\hbar}\frac{\partial E}{\partial k} = \nu_F \qquad (2.14)$$

$$\nu\ k\ \ = \nu_F \approx 1\text{x}10^6 \frac{m}{s}$$

As we see from (2.16), $E \propto P = \sqrt{P^2 + 0}$ implies the effective mass is zero, since the energy of a particle having effective mass m^* and moving with a velocity υ in a medium is E = $\overline{P^2 \upsilon^2 + m^{*2} c^4}$, where c is the speed of light. Thus, the charge carriers in graphene have zero effective mass and move at a constant velocity. Electrons in graphene are not actually massless. The effective mass is a parameter that describes how an electron at particular wave vectors responds to applied force. Since the velocity of electrons confined on graphene remain constant that indicates that the parameter (effective mass) vanishes 20 and E Vs P is linear.

Neglecting many-body effects, this description is accurate theoretically and has also been proved experimentally by measuring the energy-dependent


30
0
−20
ENERGY (eV)
K′
Γ
M
K
Brillouin zone
Conduction band
Fermi energy
Valence band
K′
Γ
M
K
MOMENTUM



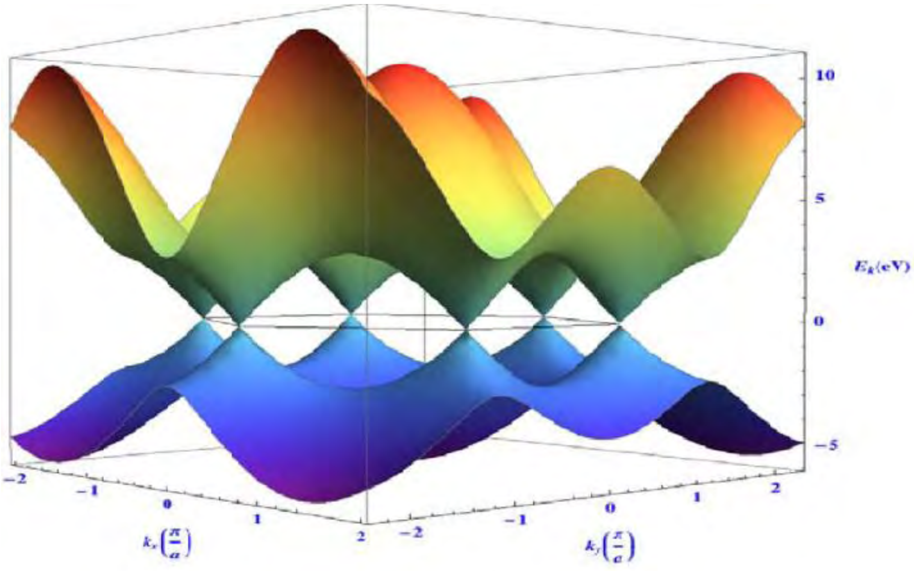
$$E = \frac{P^2}{2m} = \frac{\hbar^2 k^2}{2m}$$

$$k = \frac{2\pi}{\lambda}$$

$\sim$

The electronic structure of graphene is rather different from usual three-dimensional materials. Its Fermi surface is characterized by six double cones, as shown in Fig.2.8. In intrinsic (undoped) graphene the Fermi level is situated at the connection points of these cones. Since the density of states of the material is zero at that point, the electrical conductivity of intrinsic graphene is quite low and is of the order of the conductance quantum $\sigma \sim \frac{e^2}{h}$; the exact prefactor is still debated. The Fermi level can however be changed by an electric field so that the material becomes either n-doped (with electrons) or p-doped (with holes) depending on the polarity of the applied field. The electrical conductivity for doped graphene is potentially quite high, at room temperature it may even be higher than that of copper 21 .

Close to the Fermi level the dispersion relation for electrons and holes is linear. Since the effective masses are given by the curvature of the energy bands, this corresponds to zero effective mass. The equation describing the excitations in graphene is formally identical to the Dirac equation for massless fermions which travel at a constant speed. The connection points of the cones are therefore called Dirac points 21 .


10
5
0
−5
E_k(eV)
−2
−1
0
1
2
$k_x\left(\frac{\pi}{a}\right)$
−2
−1
0
1
2
$k_y\left(\frac{\pi}{a}\right)$

distribution of charge carriers on the conductor's surface. This separation of charges establishes an electric field that opposes further charge build-up. As long as charges flow, a steady electric potential exists called the Hall voltage and the resistivity of the conductor depends linearly on the magnetic field strength. This is known as the classical Hall Effect [22], and is the basis of many practical applications and devices such as magnetic field measurements, and position and motion detectors. With the measurements he made, Hall was able to determine for the first time the sign of charge carriers in a conductor. Even today, Hall effect measurements continue to be a useful technique for characterizing the electrical transport properties of metals and semiconductors. Indeed, the failure of the simple model of metallic conductivity to account for many experimental measurements of the Hall Effect has been one of the principal motivators leading to a better understanding of electronic properties of materials [23].

Electric field E_x applied in x direction produces following electric current j_x in x direction. Magnetic field B in z direction exert Lorentz force $\vec{j} \times \vec{B}$ on the moving charges carrying the current $\vec{j}$. For $\vec{j}$ in the x direction and $\vec{B}$ in z direction, the Lorentz force $\overline{F_L}$ is in the –y direction. The Lorentz force is given by

$$\vec{F}_L = \frac{d\vec{P}}{d\tau} = -e \quad \vec{E} + \frac{\vec{P}\times\vec{B}}{m_b} \quad - \frac{\vec{P}}{\tau}$$

$$\vec{F}_L = -e\vec{E} - \frac{e\vec{P}\times\vec{B}}{m_b} - \frac{\vec{P}}{\tau} \qquad (2.1.1)$$

where $\vec{F}_L$Lorentz force, E is the electric field, $\vec{P}$ is momentum, $\vec{B}$ is magnetic field, τ is relaxation time and m_b is band mass. The last term takes into account relaxation processes due to the diffusion of electrons by generic impurities.

The components of the Lorentz forces given by

$$F_x = -eE_x - \frac{eB}{m_b}P_y - \frac{P_x}{\tau} \qquad (2.1.2)$$

$$F_y = -eE_y + \frac{eB}{m_b}P_x - \frac{P_y}{\tau} \qquad (2.1.3)$$

In the above expression, one can notice the appearance of a characteristics frequency (cyclotron frepuency)

$$\omega_c = \frac{eB}{m_b} \qquad (2.1.4)$$

Let electrons move in the xy-plane with a magnetic field in the z direction and an electric field also in the xy-plane. In MKS units and standard notation (e > 0)====

$$F_x = -eE_x - ev_yB - m_b\frac{v_x}{\tau} \qquad (2.1.5)$$

$$F_y = -eE_y + ev_xB - m_b\frac{v_y}{\tau} \qquad (2.1.6)$$

The current density is given by

$$j_x = -nev_x \qquad (2.1.7)$$

$$j_y = -nev_y \qquad (2.1.8)$$

where n is the number of electrons per unit volume. Letting the dc conductivity be

$$\sigma_0 = \frac{ne^2}{m_b}\tau \qquad (2.1.9)$$

we can write (in the steady state when $F_x, F_y = 0$) from (2.1.5) and (2.1.6)

$$\mathrm{e}E_x = -\mathrm{e}\nu_y \mathrm{B} - \mathrm{m_b}\frac{\nu_x}{\tau} \qquad (2.1.10)$$

$$\mathrm{e}E_y = +\mathrm{e}\nu_x \mathrm{B} - \mathrm{m_b}\frac{\nu_y}{\tau} \qquad (2.1.11)$$

From (2.1.10) and (2.1.11), we get

$$\begin{matrix} E_x \\ E_y \end{matrix} = \begin{matrix} \frac{-m_b}{e\tau} & -B \\ B & \frac{-m_b}{e\tau} \end{matrix} \begin{matrix} \upsilon_x \\ \upsilon_y \end{matrix} \qquad (2.1.12)$$

$$= -\frac{m_b}{e\tau} \begin{matrix} 1 & \frac{Be\tau}{m_b} \\ \frac{-Be\tau}{m_b} & 1 \end{matrix} \begin{matrix} \upsilon_x \\ \upsilon_y \end{matrix}$$

$$\begin{matrix} E_x \\ E_y \end{matrix} = -\frac{m_b}{e\tau} - \begin{matrix} 1 & \omega_c\tau \\ \omega_c\tau & 1 \end{matrix} \begin{matrix} \upsilon_x \\ \upsilon_y \end{matrix} \qquad (2.1.13)$$

Inserting the value of υ_x and υ_y from (2.1.7) and (2.1.8) in equation (2.1.13) we obtain

$$\begin{matrix} E_x \\ E_y \end{matrix} = \frac{m_b}{ne^2\tau} \begin{matrix} 1 & \omega_c\tau \\ -\omega_c\tau & 1 \end{matrix} \begin{matrix} j_x \\ j_y \end{matrix} \qquad (2.1.14)$$

But $\frac{1}{\sigma_0} = -\frac{m_b}{ne^2\tau}$

$$\begin{matrix} E_x \\ E_y \end{matrix} = \frac{1}{\sigma_0} \begin{matrix} 1 & \omega_c\tau \\ -\omega_c\tau & 1 \end{matrix} \begin{matrix} j_x \\ j_y \end{matrix} \qquad (2.1.15)$$

Since $\vec{E} = \sigma^{-1}\vec{j}$ we obtain the current density tensor

$$\rho = \sigma^{-1} = \frac{1}{\sigma_0} \begin{matrix} 1 & \omega_c\tau \\ -\omega_c\tau & 1 \end{matrix} \qquad (2.1.16)$$

where ω_c is the cyclotron frequency and we can show (by (2.1.9))

$$\frac{B}{ne} = \frac{\omega_c \tau}{\sigma_o} \qquad \text{E2.1.17F}$$

The inverse to (3.1.15) can be written using law of inverse of matrix

$$A^{-1} = \frac{1}{det|A|} cof \;\; A_{ji}$$

$$\begin{matrix} j_x \\ j_y \end{matrix} = \frac{\sigma_o}{1+ \; \omega_c \tau^{\;2}} \begin{matrix} 1 & -\omega_c \tau \\ \omega_c \tau & 1 \end{matrix} \; \begin{matrix} E_x \\ E_y \end{matrix} \qquad (2.1.18)$$

We will use the geometry as shown in Fig. 2.1.1 We rederive the Hall coeffcient.

Setting j_y= 0, then

$$E_y = |\nu x B| = -\frac{\omega_c \tau}{\sigma_o} j_x = -\frac{B}{ne} j_x \qquad (2.1.19)$$

where $\upsilon = \upsilon_x = \frac{j_x}{ne}$ from (2.1.7). The Hall coefficient is defined as

$$R_H = \frac{E_y}{j_x B_z} = -\frac{1}{ne} \qquad (2.1.20)$$

as usual. The Hall voltage over the length w would then be

$$V_H = -E_y w = B \frac{j_x w}{ne} \qquad \text{EOKNKONF}$$

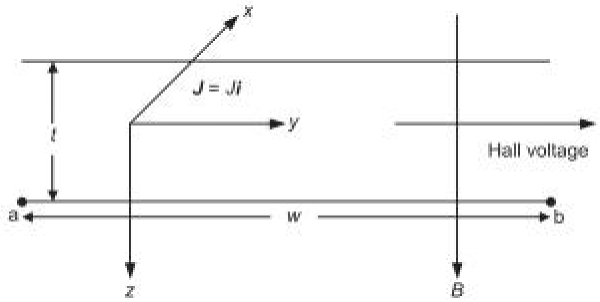
The current through the segment of area tw is

$$I_x = j_x tw \qquad \text{EOKNKOOF}$$

where t is the thickness of the slab.

So

$$V_H = \frac{B I_x}{nte} \qquad \text{EOKNKOPF}$$



n_a

$$V_H = \frac{I_x B}{n_a w e}$$

$$\frac{1}{R_{xy}}$$

$$\frac{1}{R_{xy}} = \frac{I_x}{V_H} = \frac{n_a w e}{B}$$

$$V_L = E_x L = \frac{j_x L}{\sigma_o}$$

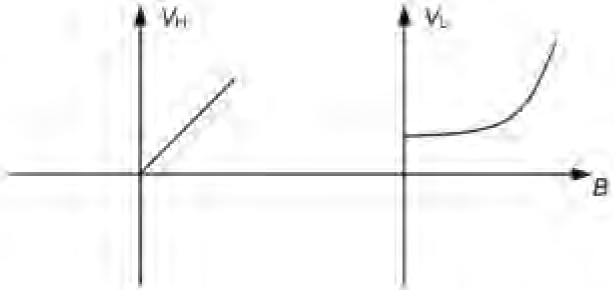
$$j_x = \frac{I_x}{tw}$$

$$V_L = \frac{I_x L}{tw\sigma_0}$$

$$\sigma = \frac{\sigma_o}{1+ \sigma_o R_H{}^2 \frac{jxB^{2}}{j^{2}}}$$

$$j \times B =$$

$$I_x =$$



conductivity and explain why charge carriers in graphene tend not to localize 20 .

Graphene shows very interesting behavior in the presence of a strong perpendicular magnetic field at low temperatures. In the presence of strong magnetic field B, electrons (holes) confined in two dimensions are considered to move in close cyclotron orbits that in quantum mechanics are quantized. The quantization of cyclotron orbits is reflected in the quantization of energy levels: at infinite B, the B = 0 dispersion is replaced by a discrete set of energy levels, known as Landau levels (LLs). In other words, we can say that electrons occupy discrete Landau energy levels as a result of their quantized orbits. That quantum behavior shows up as plateau in the conductance measured transverse to the current flow 20 .

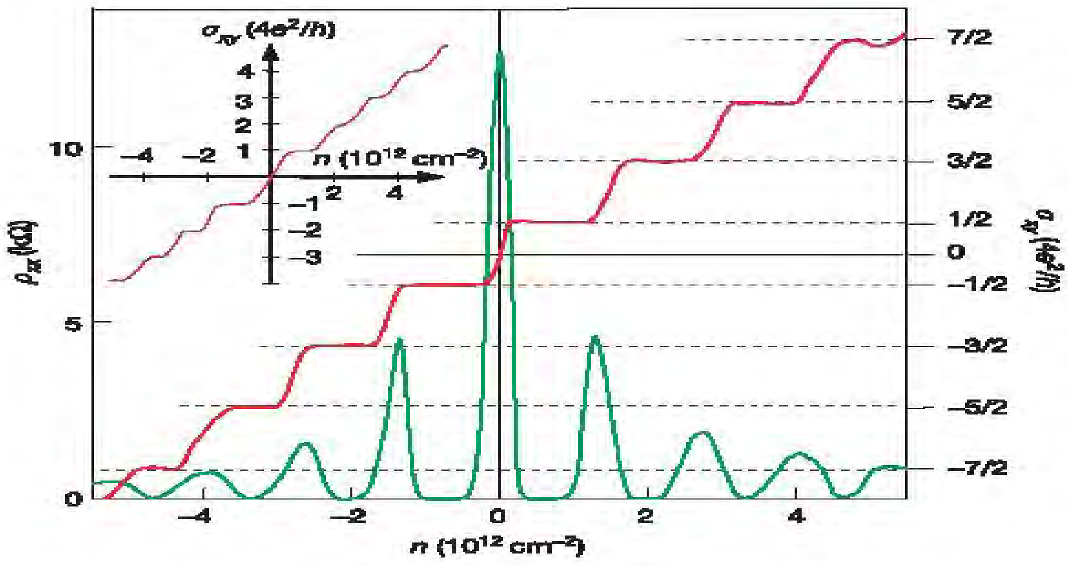
2.1.1 ANOMALOUS QUANTUM HALL EFFECT IN GRAPHENE

The quantum Hall effect is only observed in two-dimensional systems. Thus the quantum Hall effect must be explained using the properties of two-dimensional systems. Thus we can say quantum Hall effect is a quantum-mechanical version of the Hall effect, observed in two-dimensional electron systems subjected to low temperatures and strong magnetic fields, in which the Hall conductance σ undergoes certain quantum Hall transitions to take on the quantized values

$$\sigma = \frac{I_{\mathrm{channel}}}{V_{\mathrm{Hall}}} = \nu \frac{e^2}{h} \qquad (2.1.1.1)$$

where $I_{channel}$ is the channel current, V_{Hall} is the Hall voltage, e is the elementary charge and h is Planck's constant. The prefactor ν is known as the "filling factor", and can take on either integer or fractional values. The quantum Hall effect is referred to as the integer or fractional quantum Hall effect depending on whether ν is an integer or fraction, respectively. The integer quantum Hall effect is very well understood, and can be simply explained in terms of single-particle orbitals of an electron in a magnetic field. The fractional quantum Hall effect is more complicated, as its existence relies fundamentally on electron–electron interactions. Although the microscopic origins of the fractional quantum Hall effect are unknown, there are several phenomenological approaches that provide accurate approximations. For example the effect can be thought of as an integer quantum Hall effect, not of electrons but of charge-flux composites known as composite fermions. In 1988, it was proposed by an American scientist F. D. M. Haldane that there was quantum Hall effect without Landau levels. This quantum Hall effect is referred to as the anomalous quantum Hall (QAH) effect 39 .

Graphene exhibits an anomalous quantum hall effect. In classical electromagnetism, the Hall effect arises when a magnetic field is applied perpendicular to the surface of a solid carrying a current parallel to the surface. The Lorentz force causes positive and negative charges to build up on opposite sides of the solid, parallel to the current, producing a potential difference known as the Hall voltage. The direction the voltage points determines the charge of the charge carriers in the material. The quantum



σ_{xy} ($4e^2/h$)
4
3
2
1
−1
−2
−3
−4
−2
2
4
n (10^{12} cm^{-2})
ρ_{xx} (kΩ)
10
5
0
−4
−2
0
2
4
n (10^{12} cm^{-2})
7/2
5/2
3/2
1/2
0
−1/2
−3/2
−5/2
−7/2
σ_{xy} ($4e^2/h$)

Graphene is an ideal system for examining the quantum Hall effect for a number of reasons. First, graphene samples are available in such purity that the charge carrier concentration can be tuned continuously from high concentrations of electrons to high concentrations of holes simply by changing the gate voltage. Second, the purity of the graphene samples is so high that the QHE can be observed even at room temperature, whereas most materials only exhibit the QHE at much lower temperatures. Finally, graphene's anomalous quantum Hall effect, by being shifted by half compared to most systems, exhibits non-zero conductivity even as the charge carriers change from electrons to holes (the neutrality point or the Dirac point). For most materials, as the charge carrier concentration tends towards zero, so does the conductivity, so that there is a metal to insulator transition at no temperature difference. But graphene has shown no signs of a metal-insulator transition even down to liquid helium temperatures of −269°C 21 .

CHAPTER THREE

PROPERTIES OF GRAPHENE

3.1 DENSITY OF GRAPHENE

The unit hexagonal cell of graphene contains two carbon atoms and has an area of 0.052nm^2. We can thus calculate its density as being 0.77mg/m^2. A hypothetical hammock measuring 1m^2 made from graphene would thus weigh 0.77mg 21 .

3.2 OPTICAL TRANSPARENCY OF GRAPHENE

Graphene's unique optical properties produce an unexpectedly high opacity for an atomic monolayer in vacuum, absorbing $\pi a \approx 2.3\%$ of red light, where a is the fine-structure constant which is equal to 1/137. This is a consequence of unusual low-energy electronic structure of monolayer graphene that features electron and hole conical bands meeting each other at the Dirac point 50 .

3.3 STRENGTH OF GRAPHENE

Graphene has a breaking strength of 42N/m^2. Steel has a breaking strength in the range of 250-1200 MPa = $0.25\text{-}1.2\text{x}10^9\text{N/m}^2$. For a hypothetical steel film of the same thickness as graphene (which can be taken to be 3.35Å = $3.35\text{x}10^{-10}\text{m}$, *i.e.* the layer thickness in graphite), this would give a 2D breaking strength of $0.084\text{-}0.40\text{N/m}^2$. Thus graphene is more than 100 times stronger than the strongest steel and more than forty times stronger than diamond.

In our 1m^2 hammock tied between two trees you could place a weight of approximately 4kg before it would break [50]. It should thus be possible to make an almost invisible hammock out of graphene that could hold a cat without breaking. The hammock would weigh less than one mg, corresponding to the weight of one of the cat's whiskers [21].

3.4 ELECTRICAL CONDUCTIVITY OF GRAPHENE

The sheet conductivity of a 2D material is given by $\sigma = en\mu$. The mobility is theoretically limited to μ=200,000 cm^{2}V^{-1}s^{-1} by acoustic phonons at a carrier density of $n = 10^{12}$cm^{-2}. The 2D sheet resistivity, also called the resistance per square, is then 31Ω. Our fictional hammock measuring 1m^2 would thus have a resistance of 31Ω [21].

Using the layer thickness we get a bulk conductivity of 0.96x10^{6}Ω^{-1}cm^{-1} for graphene. This is somewhat higher than the conductivity of copper which is 0.60x10^{6}Ω^{-1}cm^{-1} [21], which means it can carry more electricity more efficiency, faster and with more precision than any other material.

3.5 THERMAL CONDUCTIVITY

Early measurements of the thermal conductivity of suspended graphene reported an exceptionally large thermal conductivity of approximately 5,300 W·m^{-1}·K^{-1}, compared with the thermal conductivity of pyrolytic graphite of approximately 2,000 W·m^{-1}·K^{-1} at room temperature [50]. However, later studies have questioned whether this ultrahigh value had been overestimated, and have instead measured a wide range of thermal conductivities between 1,500 - 2,500 $W \cdot m^{-1} \cdot K^{-1}$ for suspended single layer graphene. The large range in the reported thermal conductivity can be caused by large measurement uncertainties as well as variations in the graphene quality and processing conditions 50 .

3.6 IMPERMEABLITY

Graphene is also the most impermeable material ever discovered. Even helium atoms cannot squeeze through. This makes it a great material for building highly sensitive gas detectors, since even the smallest quantity of a gas will get caught in its lattice 21

CHAPTER FOUR

PRODUCTION TECHNIQUES

The market of graphene applications is essentially driven by progress in the production of graphene with properties appropriate for the specific application, and this situation is likely to continue for the next decade or at least until each of graphene's many potential applications meets its own requirements. Currently, there are probably a dozen methods being used and developed to prepare graphene of various dimensions, shapes and quality. Here we concentrate only on those that are scalable 26 .

4.1 LIQUID PHASE AND THERMAL EXFOLIATION

Liquid-phase exfoliation of graphite 26, 27 is based on exposing the materials to a solvent with a surface tension that favors an increase in the total area of graphite crystallites. The solvent is typically non-aqueous, but aqueous solutions with surfactant can also be used. With the aid of sonication, graphite splits into individual platelets, and prolonged treatment yields a significant fraction of monolayer flakes in the suspension, which can be further enriched by centrifugation. Related method is the graphite oxide route in which graphite pellets are first oxidized and then ultrasonically exfoliated in an aqueous solution 28 . After exfoliation of graphite oxide the suspension may be further processed by centrifugation, and can then be deposited as a thin film on almost any surface and reduced in situ back to the parent graphene state.

An industrially important variation of the fully aqueous-based graphite-oxide route makes use of a thermal-shock procedure to achieve exfoliation and reduction simultaneously 29 . Even though the resulting material may contain graphene components with several layers, it still preserves many of the appealing properties of single-layer graphene. Similarly to oxidation, the parent graphite stacking can be disturbed via intercalation of small molecules. Such graphite intercalation compounds may then be used in a similar way as precursors and can subsequently be subjected to thermal or plasma processes to achieve their delamination into single sheets.

Also, there are several methods of producing suspensions of graphene nanoribbons—via unzipping of single-wall carbon nanotubes 30,31 . Although they are more expensive than chemical exfoliation of graphite or graphite oxide, these methods allow one to achieve suspensions with well-defined distributions (potentially very narrow) of graphene platelets. Similarly, nanotube unzipping allows better control over the chemical functionalization and quality of the edges.

Such bulk grades of graphene are already available on the tonne scale and are currently being evaluated in numerous fields of application 32 . Thus, graphene-based paints and inks will find their way into printed electronics, electromagnetic shielding, barrier coatings, heat dissipation, supercapacitors, smart windows 33 , and so on. A number of flake-based products can be expected in the marketplace within a few years, and prototype applications for conductive inks have already been demonstrated on the commercial level.

4.2 CHEMICAL VAPOUR DEPOSITION

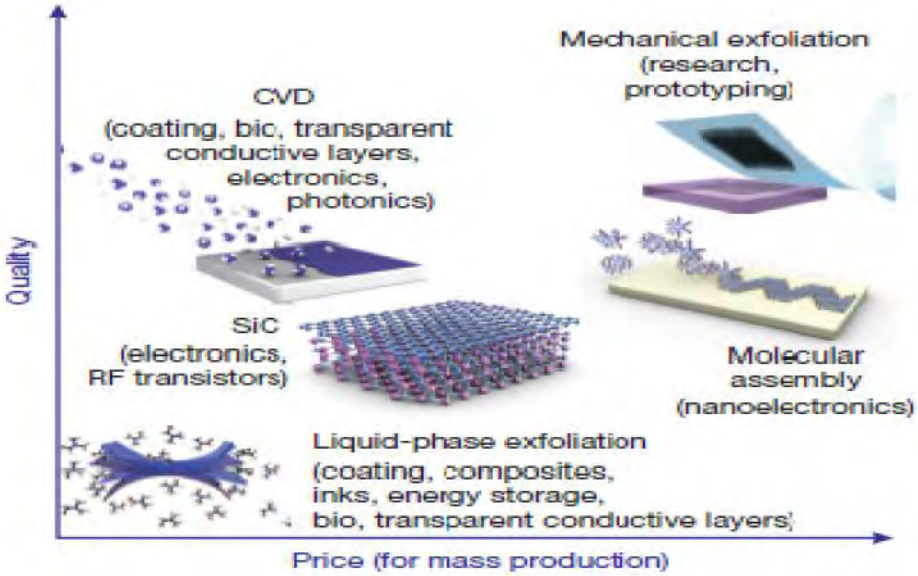
Large-area uniform polycrystalline graphene films are now being grown by chemical vapour deposition (CVD) on copper foils and films, and show promise for many applications 34 . Despite the fact that the complete process typically requires transfer from the copper support to a dielectric surface or other substrate of interest 2 , the production of square metres of graphene has already been achieved 35 . These films have also been transferred onto 200 mm Si wafers on which state-of-the-art devices have been demonstrated. On a smaller scale, these films show transport properties equivalent to those of exfoliated graphene on both SiO_2 and hexagonal boron nitride substrates. Despite the presence of defects, grain boundaries, inclusions of thicker layers, and so on, such films are ready for use in transparent conductive coating applications (such as touch screens).

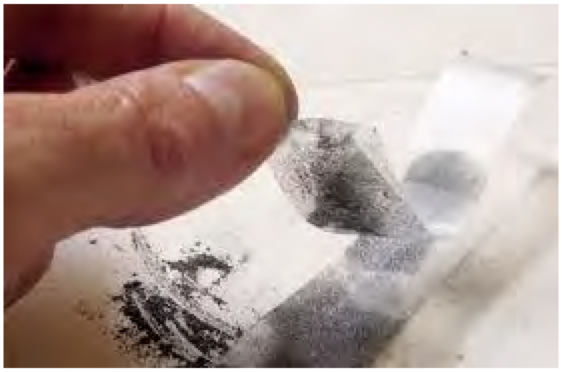
At present, the process is expensive, as shown in Fig. 4.3.1, owing to large energy consumption and because the underlying metal layer has to be removed. However, once the transfer process is optimized this method may indeed be disruptive and cost-effective. A number of issues need to be resolved before graphene CVD technology can become widely used. Graphene growth on thin (tens of nanometres) films of metals needs to be achieved, simultaneously gaining control of the domain (grain) size, ripples, doping level and the number of layers. Control of the number and relative crystallographic orientation of the graphene layers is critical because it will enable a number of applications which would require double, triple and even thicker layers of graphene. Simultaneously, the transfer process should be improved and optimized with the objectives of minimizing the damage to graphene and of recovering the sacrificial metal.

4.3 SYNTHESIS ON SiC

Silicon carbide is a common material used for high-power electronics. It has been demonstrated that graphitic layers can be grown either on the silicon or carbon faces of a SiC wafer by sublimating Si atoms, thus leaving a graphitized surface 36 . Initially, the C-terminated face of SiC was used to grow a turbostratic stack of many randomly oriented polycrystalline layers 37 , but now the number of graphene layers grown 38 can be controlled. The quality of such graphene can be very high, with crystallites approaching hundreds of micrometres in size 39 .

The two major drawbacks of this method are the high cost of the SiC wafers and the high temperatures (above 1,000^{0}C) used, which are not directly compatible with silicon electronics technology. There are potentially several ways to take advantage of the growth of graphene on SiC, including the growth of thin SiC on Si, although this approach requires further development. As a result of the high-temperature growth, high substrate cost, and small-diameter wafers, the use of graphene on SiC will probably be limited to niche applications. High-frequency transistors based on SiC-grown graphene 40 may well find applications within a decade when the existing technology, based


Mechanical exfoliation
(research,
prototyping)
CVD
(coating, bio, transparent
conductive layers,
electronics,
photonics)
Quality
SiC
(electronics,
RF transistors)
Molecular
assembly
(nanoelectronics)
Liquid-phase exfoliation
(coating, composites,
inks, energy storage,
bio, transparent conductive layers)
Price (for mass production)

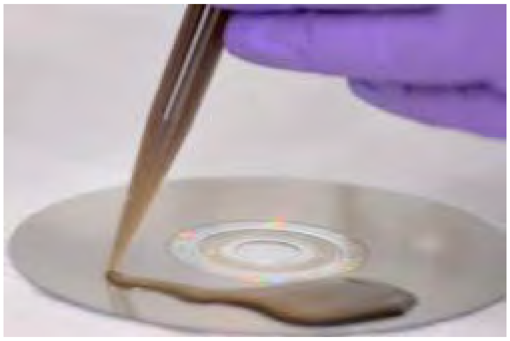


4.6 REDUCTION OF GRAPHITE OXIDE

Graphite oxide reduction was probably the first method of graphene synthesis. P. Boehm reported producing monolayer flakes of reduced graphene oxide in 1962. Geim acknowledged Boehm's contribution. Rapid heating of graphite oxide and exfoliation yields highly dispersed carbon powder with a few percent of graphene flakes. Reduction of graphite oxide monolayer films, e.g. by hydrazine with annealing in argon/hydrogen, was reported to yield graphene films. However, the quality is lower compared to scotch-tape graphene, due to incomplete removal of functional groups. Furthermore, the oxidation protocol introduces permanent defects due to over-oxidation. The oxidation protocol was enhanced to yield graphene oxide with an almost intact carbon framework that allows efficient removal of functional groups. The measured charge carrier mobility exceeded 1,000centimetres (393.70 in)/Vs. Spectroscopic analysis of reduced graphene oxide has been conducted 50 .

4.7 SHEARING

In 2014 defect-free, unoxidized graphene-containing liquids were made from graphite using mixers that produce local shear rates greater than 10. Commercial graphene products using the output were announced. The method was claimed to be applicable to boron nitride, Molybdenum disulfide and other layered crystals 50 .



CHAPTER FIVE

APPLICATIONS

Graphene has a number of properties which makes it interesting for several different applications. It is an ultimately thin, mechanically very strong, transparent and flexible conductor. Its conductivity can be modified over a large range either by chemical doping or by an electric field. The mobility of graphene is very high 42 which makes the material very interesting for electronic high frequency applications 40 . Recently it has become possible to fabricate large sheets of graphene. Using near-industrial methods, sheets with a width of 70cm have been produced 34, 43 . Since graphene is a transparent conductor it can be used in applications such as touch screens, light panels and solar cells, where it can replace the rather fragile and expensive Indium-Tin-Oxide (ITO). Flexible electronics and gas sensors 44, 45 are other potential applications. The quantum Hall effect in graphene could also possibly contribute to an even more accurate resistance standard in metrology 46 . New types of composite materials based on graphene with great strength and low weight could also become interesting for use in satellites and aircraft 47, 48 .

Graphene is a material that can be utilized in numerous disciplines including, but not limited to: bioengineering, composite materials, energy technology and nanotechnology.

5.1. BIOLOGICAL ENGINEERING

Bioengineering will certainly be a field in which graphene will become a vital part of in the future; though some obstacles need to be overcome before it can be used. Current estimations suggest that it will not be until 2030 when we will begin to see graphene widely used in biological applications as we still need to understand its biocompatibility (and it must undergo numerous safety, clinical and regulatory trials which, simply put, will take a very long time). However, the properties that it displays suggest that it could revolutionize this area in a number of ways. With graphene offering a large surface area, high electrical conductivity, thinness and strength, it would make a good candidate for the development of fast and efficient bioelectric sensory devices, with the ability to monitor such things as glucose levels, hemoglobin levels, cholesterol and even DNA sequencing. Eventually we may even see engineered 'toxic' graphene that is able to be used as an antibiotic or even anticancer treatment. Also, due to its molecular make-up and potential biocompatibility, it could be utilized in the process of tissue regeneration 51 .

5.2. OPTICAL ELECTRONICS

Graphene strongly interacts with photons, with the potential for direct band-gap creation. This is promising for optoelectronic and nanophotonic devices. Light interaction arises due to the Van Hove singularity 52 . Graphene displays different time scales in response to photon interaction, ranging from femtoseconds (ultra-fast) to picoseconds. Potential uses include transparent films, touch screens and light emitters or as a plasmonic device that confines light and alters wavelengths 52 is a potential application of graphene in optoelectronics.

5.3. ULTRAFILTRATION

Another standout property of graphene is that while it allows water to pass through it, it is almost completely impervious to liquids and gases (even relatively small helium molecules). This means that graphene could be used as an ultrafiltration medium to act as a barrier between two substances. The benefit of using graphene is that it is only 1 single atom thick and can also be developed as a barrier that electronically measures strain and pressures between the 2 substances (amongst many other variables). A team of researchers at Columbia University have managed to create monolayer graphene filters with pore sizes as small as 5nm (currently, advanced nanoporous membranes have pore sizes of 30-40nm). While these pore sizes are extremely small, as graphene is so thin, pressure during ultrafiltration is reduced. Co-currently, graphene is much stronger and less brittle than aluminium oxide (currently used in sub-100nm filtration applications). This mean that graphene is developed to be used in water filtration systems, desalination systems and efficient and economically more viable biofuel creation 51 .

5.4. COMPOSITE MATERIALS

Graphene's strength, stiffness and lightness suggested it for use with carbon fiber 52 . Currently, aerospace engineers are incorporating carbon fibre into the production of aircraft as it is also very strong and light. However, graphene is much stronger whilst being also much lighter. Ultimately it is expected that graphene is utilized (probably integrated into plastics such as epoxy) to create a material that can replace steel in the structure of aircraft, improving fuel efficiency, range and reducing weight. Due to its electrical conductivity, it could even be used to coat aircraft surface material to prevent electrical damage resulting from lightning strikes. In this example, the same graphene coating could also be used to measure strain rate, notifying the pilot of any changes in the stress levels that the aircraft wings are under. These characteristics can also help in the development of high strength requirement applications such as body armour for military personnel and vehicles 51 .

Graphene has been used as a reinforcing agent to improve the mechanical properties of biodegradable polymeric nanocomposites for engineering bone tissue 52 .

5.5. PHOTOVOLTAIC CELLS

Offering very low levels of light absorption (at around 2.7% of white light) whilst also offering high electron mobility means that graphene can be used as an alternative to silicon or ITO in the manufacture of photovoltaic cells. Silicon is currently widely used in the production of photovoltaic cells, but while silicon cells are very expensive to produce, graphene based cells are potentially much less so. When materials such as silicon turn light into electricity it produces a photon for every electron produced, meaning that a lot of potential energy is lost as heat. Recently published research has proved that when graphene absorbs a photon, it actually generates multiple electrons. Also, while silicon is able to generate electricity from certain wavelength bands of light, graphene is able to work on all wavelengths, meaning that graphene has the potential to be as efficient as, if not more efficient than silicon, ITO or (also widely used) gallium arsenide. Being flexible and thin means that graphene based photovoltaic cells could be used in clothing; to help recharge your mobile phone, or even used as retro-fitted photovoltaic window screens or curtains to help power your home 51 .

5.6. ENERGY STORAGE

Due to graphene's high surface area to mass ratio, one potential application is in the conductive plates of supercapacitors. In February 2013 researchers announced a novel technique to produce graphene supercapacitors based on the DVD burner reduction approach. In 2014 a supercapacitor was announced that was claimed to achieve energy density comparable to current lithium-ion batteries 52 .

In 2015 the technique was adapted to produce stacked, 3-D supercapacitors. Laser-induced graphene was produced on both sides of a polymer sheet. The sections were then stacked, separated by solid electrolytes, making multiple microsupercapacitors. The stacked configuration substantially increased the energy density of the result. In testing, the researchers charged and discharged the devices for thousands of cycles with almost no loss of capacitance. The resulting devices were mechanically flexible, surviving 8,000 bending cycles. This makes them potentially suitable for rolling in a cylindrical configuration. Solid-state polymeric electrolyte-based devices exhibit areal capacitance of >9 mF/cm^2 at a current density of 0.02 mA/cm^2, over twice that of conventional aqueous electrolytes 52 .

Also in 2015 another project announced a microsupercapacitor that is small enough to fit in wearable or implantable devices. Just one-fifth the thickness of a sheet of paper, it is capable of holding more than twice as much charge as a comparable thin-film lithium battery. The design employed laser-scribed graphene, or LSG with manganese dioxide. They can be fabricated without extreme temperatures or expensive “dry rooms”. Their capacity is six times that of commercially available supercapacitors. The device reached volumetric capacitance of over 1,100 F/cm3. This corresponds to a specific capacitance of the constituent MnO_2 of 1,145 F/g, close to the theoretical maximum of 1,380 F/g. Energy density varies between 22 and 42 Wh/l depending on device configuration 52 .

CHAPTER SIX

CONCLUSION

Before monolayer graphene was isolated in 2004, it was theoretically believed that two dimensional compounds could not exist due to thermal instability when separated. However, once graphene was isolated, it was clear that it was actually possible, and it took scientists some time to find out exactly how. After suspended graphene sheets were studied by transmission electron microscopy, scientists believed that they found the reason to be due to slight rippling in the graphene, modifying the structure of the material. However, later research suggests that it is actually due to the fact that the carbon to carbon bonds in graphene are so small and strong that they prevent thermal fluctuations from destabilizing it 51 . Graphene is one of the hottest new materials in physics today. The development of this new material opens new exiting possibilities. It is the first crystalline 2D-material and it has unique properties, which makes it interesting both for fundamental science and for future applications It is undoubtedly emerging as one of the most promising nanomaterials because of its unique combination of superb properties, which opens a way for its exploitation in a wide spectrum of applications ranging from electronics to optics, sensors, and biodevices.

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DECLARATION

This graduate project paper is my original work, has not been presented for a degree in any other University and that all the sources of material used for the thesis have been dully acknowledged.

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This graduate project paper has been submitted for examination with my approval as University co-advisor.

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