SYNTHESIS AND CHARACTERIZATION OF GRAPHENE: ITS OPTICAL PROPERTIES FOR APPLICATION IN SOLAR CELL

Nwankwo, Ndidiamaka Vivian^I, A.D. A. Buba^{II} Department of Physics, University of Abuja, Nigeria

Abstract

Graphene shows remarkable optical behavior, where above an incident light intensity threshold, transmission is attenuated. There is significant interest in synthesizing large-area graphene films at low temperatures by chemical vapor deposition (CVD) for optical device applications. The results obtained shows that therefractive index of graphene is $n_g = 1.86 - 1.56i$ while that of graphene conductance is $\sigma_r = 1.016$ and $\sigma_i \sim 0$ and graphene density 0.77 mg. The modified CVD method is expected to allow the direct growth of graphene in device manufacturing processes for practical applications while keeping underlying devices intact.

Keywords:Graphene, Optical, Incident, Synthesizing and chemical vapor deposition (CVD)

1. Introduction

Graphene is a single atom, thick two-dimensional (2D) material, thus, exhibiting $\sim 97.7\%$ transmittance throughout the entire visible light spectrum. Additionally, graphene has a flat transmittance spectrum from the ultraviolet (UV) region to the long wave length infrared (IR) region, thus exhibiting a wide window that allows a comprehensive range of photon wave length passed through it. Unconditionally, except graphene, these combinations of remarkable optical properties are yet to be observed in any types of materials till date. Similarly, graphene has unusual electronic transport properties, which follows the characteristics of 2D Dirac fermions, quantum hall effects, Landau level quantization, and so on. Consequently, graphene's free charges are immobile in one spatial dimension but mobile in other two dimensions, and thus, charge carrier mobility is $\sim 10^6 cm^2 / v/s$ in free-standing graphene (Yoshikawa K 2017). Similarly, also exhibits excellent and thermal graphene mechanical properties $(k \sim 300 - 500 W/mk)^1$ and chemical inertness. Hence, all these properties coupled with the optical properties put together graphene a stronger candidate for applications in transparent conducting electrodes (TCEs) flexible optoelectronics, energy harvesting devices, photo detectors, and many other optical devices (Yoshikawa 2017).

Furthermore, graphene is anticipated as an emerging alternate for conventional transparent conductive metal oxides, specifically, indium tin oxide (ITO), which contains indium as a toxic, costly, and scarce element. In particular, the graphene-based TCE for application in solar cells with enhanced efficiency is of utmost interest. To date, graphene electrodes have been applied for different types of solar cells, namely, solid-state solar cells, electrochemical solar cells, quantum dot solar cells (QDSCs), and polymer solar cells (Zang Y 2011).

2. Material and Method

The experiment setup and data measurement was done at the department of mineralogy Kaduna state Polytechnic. This research will involve Graphene films deposited on a glass substrate using spin coating. It employed a reduction protocol where a 4 Watt, 365 nm UV lamp was used for exposure of the synthesized Graphene films for different periods of time. The current-voltage (I-V) characteristics for the Graphene films were measured as a function of UV exposure time. For this, a four-probe configuration was used and current across the two inner terminals was taken as a function of the source voltage applied across the two outer terminals.

The evolution of the I-V characteristics with the UV exposure time (up to 2 hours) was analyzed. Raman and X-ray photoemission (XPS) spectra was obtained both prior to UV exposure as well as post 2 hours exposure for samples belonging to the same batch of synthesis. A comparison of the Raman spectra of the grown sample and a corresponding sample was synthesized after 2 hours exposure to UV light. The characteristic D, G and 2D peaks was labeled. A comparison of reduced GO Raman spectra with pristine graphene (CVD grown), graphene single layer ($I_{2D}/I_G = 2.7$), bilayer ($I_{2D}/I_G = 1.4$) and multilayer Raman spectra were compared to provide a recipe for estimating extent of disorder in Graphene.

A conservative estimate of the extent of disorder based on the observed I_D/I_G ratio of the 2 hour reduced Graphene sample was made and were estimated.

Distilled water of $11000cm^3$ was measured and mixed with the graphite sample, poured into the flotation cell and agitated for 3 minutes. During the agitation process, three drops of regulator (sodium hydroxide) was added to adjust the pulp pH to 8.5ml and

conditioned for seven minutes. Thereafter, 3ml of kerosene and pine oil added, immediately after the conditioning/agitation, air was allowed to pass through the pulp at a reasonable rate and froth emerged and collected till barren froth surfaced.

Table 1.0: Shows fixed carbon content of raw graphite (as received	d)
--	----

Where:

	Samples Content %									
He			V		AS		FC			
adSample	С	Μ								
			19.		39.		26.2			
	4.51	76		48		5				

MC = Moisture Content, VM = Volatile Matter, AS = Ash Content, FC = Fixed Carbon

Table 2.0: Shows fixed carbon content of the concentrate

	Samples Content %								
С			V		AS		FC		
oncentrate	С	Μ							
			5.4		10.		81.2		
	.97	5		30		8			

3. Result and Discussion

Optical properties of graphene

The electromagnetic properties of graphene are based on its unique atomic structure and the electronic band-gap between graphene sheet layers. As it is a two-dimensional material, the massless Dirac fermions in graphene have an amazingproperty: the optical response is universal and expressed only regarding to the fine-structure constant. To derive this response, we need determine the electric field of the incident light via avector potential:

$$\vec{A}(t) = \vec{A} \exp\left(-i\omega t\right) \tag{1.0}$$

$$\vec{E}(t) = -\frac{1}{c}\frac{\partial \vec{A}}{\partial t} = \frac{i\omega}{c}\vec{A}$$
(1.1)

Thus, the incident working on the Hamiltonian of Dirac electron in the presence of an electric field is:

$$\vec{H} = v\vec{\sigma}\left(\vec{P} - \frac{e}{c}\vec{A}\right) = \widehat{H_0} + \widehat{H_{int}}$$
(1.2)

Where:

$$H_{int} = \frac{ve}{2c}\vec{\sigma}\vec{A} = i\frac{ev}{2\omega}\vec{\sigma}\vec{E}$$
(1.3)

Is the Hamiltonian of the electron photon interaction, the factor $\frac{1}{2}$ in equation (4.3) is necessary since the standard expression for the complex field is:

$$\vec{E}(t) = Re\left[\vec{E}\exp(-i\omega t)\right] = \frac{1}{2}\left[\vec{E}\exp(-i\omega t) + \vec{E}\exp(i\omega t)\right]$$
(1.4)

And only the first term need to take into account. This interaction induces transitions from the occupied whole states $\varphi_h(\vec{k})$ to empty election state $\varphi_e(\vec{k})$ with the same wave vector \vec{K} , the intraband transitions being forbidden by momentum conservation. The matrix element of the Hamiltonian equation is given by:

$$\left\langle \varphi_{h} \middle| \widehat{H_{int}} \middle| \varphi_{e} \right\rangle = \frac{ev}{2\omega} \left(E_{y} Cos\varphi \mp E_{x} Sin\varphi \right)$$
(1.5)

Where the negative and positive signs correspond to k and k' Valleys, the sign depends only on the polar angle \emptyset of the \vec{K} vector, not on its length. After determination the average value, the square matrix element $|M|^2$ over \emptyset is found by:

$$|M|^{2} = \overline{\langle \varphi_{h} | \widehat{H_{int}} | \varphi_{\theta} \rangle} = \frac{e^{2} v^{2}}{8\omega^{2}} \left| \vec{E} \right|^{2}$$
(1.6)

Here, assuming the phone propagates perpendicular to the graphene plane, thus the electric field $\vec{E} = (E_x, E_y, 0)$ lies within the plane. The absorption probability per unit time, to the lowest order of perturbation theory is:

$$P = \frac{2\pi}{\hbar} |M|^2 N(\varepsilon = \frac{\hbar\omega}{2})$$
(1.7)

Where, $N(\varepsilon) = \frac{2|E|}{\pi \hbar^2 v^2}$ is the density of states of single layer graphene material, the spin and valley degeneracy has been considered and the energy of the final state is $\frac{\hbar \omega}{2}$ substituting the equation (1.6) result in:

$$P = \frac{e^2}{4\hbar^2 \omega} \left| \vec{E} \right|^2 \tag{1.8}$$

Thus the optical absorption energy per unit time is:

$$W_{\alpha} = p\hbar\omega = \frac{c}{4\hbar} \left|\vec{E}\right|^2$$
, therefore the absorption coefficient is:
 $A_0 \frac{W_{\alpha}}{W_i} = \frac{\pi e^2}{\hbar c}$

$$\frac{1}{137.04} \approx 2.3\%$$

Optical refractive index of graphene

From Maxwell equations and Snell's law, displacement \vec{D} magnetic field \vec{H} and surface current \vec{J} can be expressed as:

$$\vec{D} = \varepsilon_0 \vec{E} + \vec{P} = \varepsilon_0 \varepsilon_r \vec{E}$$
(1.9)

$$\vec{J} = -\frac{\sigma}{d}\vec{E}$$
(1.10)

$$\nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} = \vec{J} + \varepsilon_0 \frac{\partial \vec{E}}{\partial t}$$
(1.11)

Where \vec{P} is the dielectric polarization, σ is the conductance, d is the material thickness. ε_0 is the free space permittivity (8.854 × 10⁻¹² F/m), and r is the relative material permittivity, μ_0 is the open space magnetic permeability $4\pi \times 10^{-7} H/m$, μ_r is relative permeability of the material.

The optical conductance of graphene defined from its linear dispersion relation as:

$$\sigma = \sigma_r + \sigma_i \tag{1.12}$$

Where σ_r and σ_i are the real and imaginary parts respectively, which can be rewritten as;

$$\sigma_{r} = \sigma_{0} \left[\frac{1}{2} + \frac{1}{72} \frac{\hbar\omega^{2}}{\xi^{2}} \times \left(tan \frac{\hbar\omega + 2\mu_{k}}{4K_{B}T} + tan \frac{\hbar\omega - 2\mu_{k}}{4K_{B}T} \right) \right]$$
(1.13)
$$\sigma_{i} = \sigma_{0} \left[\frac{\mu_{c}}{\hbar\omega} \frac{4}{\pi} \left(1 - \frac{2\mu_{c}^{2}}{9\xi^{2}} \right) - \log \frac{|\hbar\omega + 2\mu_{c}|}{|\hbar\omega - 2\mu_{c}|} \left(\frac{1}{n} + \frac{1}{36\pi} \left(\frac{\hbar\omega}{\xi} \right)^{2} \right) \right]$$
(1.14)

Where $\xi \sim 3ev$ is the energy hopping between nearest atoms in the chemical potential near Dirac point, $K_B T$ is the incident photon energy and $\sigma_0 = \frac{\pi}{2} * \frac{e^2}{\hbar} = 6.08 \times 10^{-5} \Omega^{-1}$ is the universal graphene conductance. One theoretical work shows when the temperature T = 300k under visible incidents real part and imaginary part of graphene conductance are; $\sigma_r = 1.016$ and $\sigma_i \sim 0$ respectively.

Substitute equations (1.13) and (1.14) into (1.12), we can rewrite the equation as:

$$\frac{\sigma}{d}\vec{E} + i\omega\varepsilon_0\vec{E} = i\omega\varepsilon_0\varepsilon_r\vec{E}$$
(1.15)

We can extract the relative permittivity \mathcal{E}_r as:

$$\varepsilon_r = 1 + \frac{\sigma}{j\omega\varepsilon_0} \tag{1.16}$$

By definition of the refractive index of the medium is $n = \sqrt{\varepsilon_r \mu_r}$, which indicates the transmission efficiency in the medium. Regarding the linear susceptibility, the refractive

index is $n_0 = \sqrt{\varepsilon_r}, \mu_r = 1$ when the material is graphene. Therefore, we can obtain the graphene refractive index as:

$$n_g = \sqrt{\varepsilon_r} = \sqrt{1 + \frac{\sigma}{j\omega\varepsilon_0}}$$

Where the refractive index is expressed in terms of fundamental constants and the ratio of optical wavelength to graphene thickness. We note that a universal conductance implies a refractive index with strong dispersion. For $\lambda = 550nm$, $\sigma = 0.3354nm$ Therefore we find $n_g = 1.86 - 1.56i$



Figure 1.0: Refractive indexes of (a) monolayer graphene and (b) bilayer graphene

4. Density of graphene

The unit hexagonal cell of graphene contains two carbon atoms and has an area of $0.053nm^2$ and its density was found to be $0.77mg/m^2$. A hypothetical hammock measuring 1m2 made from graphene would thus weigh 0.77 mg

5. Electrical conductivity of graphene

The sheet conductivity of a 2D material is given by. The mobility is theoretically limited to $\mu = 200,000 cm^2$ by acoustic phonons at a carrier density of $n = 1014 cm^2$. The 2D sheet resistivity, also called the resistance per Square, is then 28Ω . Our fictional hammock measuring $1m^2$ would thus have a resistance of 28Ω . $\sigma = en\mu$ Using the layer thickness we get a bulk conductivity of $0.86 \times 10^2 \Omega^{-1} cm^{-1}$ for graphene. This is somewhat higher than the conductivity of copper which is $0.54 \times 10^2 \Omega^{-1} cm^{-1}$.

Conclusion

In general, solar cells are not that efficient. However, recent advances in graphenebased solar cells have seen the reflectance of solar rays reduced by 20%, which provides a potential efficiency increase up to 20%, the thesis explains and demonstrates the use of Graphene for making solar panels for deriving extreme power supply using the unlimited solar energy. The results obtained shows that the refractive index of graphene is $n_g = 1.86 - 1.56i$ while that of graphene conductance is $\sigma_r = 1.016$ and $\sigma_i \sim 0$ and graphene density 0.77 mg. The modified CVD method is expected to allow the direct growth of graphene in device manufacturing processes for practical applications while keeping underlying devices intact.

Acknowledgements

I am deeply grateful to my supervisor, Prof. Abdul. A. Buba, for his kind guidance and excellent supervision. His knowledge and expertise in the field of device technology and especially graphene devices added considerably to my graduate experience. I would really like to appreciate his invaluable help. I am forever grateful. I also wish to appreciate all my colleagues.

Bibliography

Angermann, H. H. and G. Hörz (1993). Influence of sulfur on surface carbon monolayer formation and graphite growth on nickel. Applied Surface Science,70-71,Part 1,(June 02, 1993) (163-168),0169-4332

Barone, V., O. Hod, et al. (2006). Electronic Structure and Stability of Semiconducting Graphene Nanoribbons. Nano Letters,6,12,(November 24, 2006) (2748-2754),1530-6984

Yong-Tae Kim, Department of Chemistry, BK-21 School of Chemical Materials Science SKKU Advanced Institute of Nanotechnology, Sung kyunkwon University, Swon, Korea

Byung Hee Hong, Department of Chemistry, Seoul National University, Seoul, Korea

Daeheon Choi, Department of Chemistry & Institute for Advanced Research, Nagoya University, Nagoya 464- 8620 Japa

Yasuo Azuma, Materials and Structures Laboratory, Tokyo Institute of Technology, Yokohama 226-8503, Japan

Ting YU,Division of Physics and Applied, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore

Riichiro Saito, Department of Physics, Tohoku University, Sendai, Miyagi 9808578, Japan

Book:Alternate energy:Assesment and Implementation,James J Winebrake

R. Olac-vaw, H. C. Kang, H. Karasawa, Y. Miyamoto, H. Handa, H. Fukidome, T. Suemitsu, M. Suemitsu, and T. Otsuji, Ambipolar behavior in epitaxial graphene based field-effect transistors on Si substrate. Jpn. J. Applied Physics. 49: 06GG01. (2010).

H. C. Kang, H. Karasawa, Y. Miyamoto, H. Handa, T. Suemitsu, M. Suemitsu, T. Otsuji, Epitaxial graphene field effect transistors on silicon substrates. Solid State Electron. 54:1010-1014. (2010).