

## Introducing New Optoelectronic Properties of Graphene-Multilayer

H.A.Elmeleegi<sup>\*</sup>, Z.S.Elmandouh, A.Abdel Moez

Physical Research Division  
National Research Centre (NRC), Dokki, Cairo, Egypt  
eml3121@gmail.com

F.Taher

Faculty of Science,  
AlAzhar University (Girls), Cairo, Egypt

---

**Abstract:** *This paper presents new properties for graphene-multilayer. In the preparation step, we use Hammer method and Pulsed Laser Deposition to create graphene-multilayer. We analyze the features of the prepared graphene-multilayered. Experimental results reveal the existence of a negative resistance upon applying low AC voltage and illumination. This negative resistance was attributed to the occupation of traps and, the change of polarization due to the change of their dipole moments. Then, we calculated various optical parameters for graphene-multilayered samples, such as optical energy gap, refractive index, extinction coefficient, dielectric loss, and dielectric energy loss. To determine both of dispersion energy and oscillating energy, we apply single oscillator theory equation as well.*

**Keywords:** *A. Chalcogenides B. Negative Resistance, C amorphous materials, D. photoelectron spectroscopy, E. transport properties.*

---

### 1. INTRODUCTION

Since single layer of graphene transmits 98% of the total incident light, graphene was found to be an alternative to ITO in organic photovoltaic cells. Graphene also shows remarkable optical properties [1]. Despite being few atoms thick, it can be optically visualized [2, 3]. Previous work show that Raman characterization for graphene is quite sensitive and diagnostic for crystallization quality, the number of layers, and electrical doping[4,5]. Graphene is zero-gapped semi-metal, whose electronic band structure shows linear dispersion near the charge neutral Dirac point. Therefore, it exhibits novel electronic properties involving ballistic transport [6]. Chemical and physical properties make it an ideal photonic and optoelectronic material [7-10]. Optical properties and optical conductivity of graphene are independent of the frequency of the transmitted light [11, 15]. The main contribution of this paper is discovering new phenomenon of graphene-multilayer and discussing the root of this phenomenon. Our experimental work uses unique preparation methods and presents three-dimensional graphs to visualize the results.

### 2. EXPERIMENTAL WORK

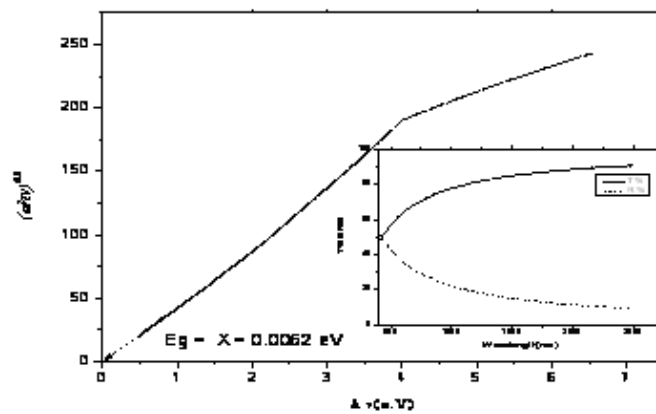
Graphene Oxide (GO) was synthesized from natural white graphite (sigma-Aldrich) by a modified Hummers' method [16, 17]. The GO paste collected from the filter paper is dispersed in deionized water (600 ml) in ultrasonic 70 % amplitude for 1 hour. The suspension is washed with deionized water for 5~7 times for five days by decantation and finally filtered, well-washed GO paste collected on the filter paper is re-dispersed into water by ultra-sonication. The obtained brown dispersion is then subjected to 30 mins of centrifugation at 5000 rpm to remove any un-exfoliated GO. The graphene oxide was obtained by dehydration at 60 °C. Afterward, graphene film was grown on Glass substrates by the pulsed laser deposition technique. 1 gram of the dark brown graphene oxide sheets was loaded for the PLD experiment. The energy of laser destroys the O bonds in Graphene Oxide and then Graphene was precipitated on glass slides [18]. A Titanium Sapphire LASER "femto-second laser" at wavelength of 800 nm, pulse duration of 40 fs, average pulse energy of 220mW and maximum repetition rate of 1000Hz was used. The Glass substrates

were separated from the target by a distance of approximately 9 cm with rotation speed of 8 rpm to guarantee a homogeneous consumption for typical irradiation time 20 mins. The deposition chamber was evacuated to a base pressure of approximately  $8.4 \times 10^{-4}$  torr. The maximum temperature of the glass substrates was 200 C°. UV/VIS absorption and transmission (normal incidence spectrum) were measured using T80+ UV/VIS spectrometer PG Instruments Ltd... Current-Voltage properties during illumination with uv-lamp and room light were taken from Virtins Instruments digital- computerized oscilloscope.

### 3. RESULTS AND DISCUSSION

#### 3.1. Optical Results

Graphene has a very high optical transmission with only 2.3% absorption of white light, so; optical properties of graphene were investigated by measuring optical transmission and reflection data of these samples as shown in figure 1. The absorption coefficient ( $\alpha$ ) of graphene films were calculated from both the transmission and reflection spectra according to the following equation [19]



**Figure1a.** Relation between optical transmission and wave length for graphene thin films

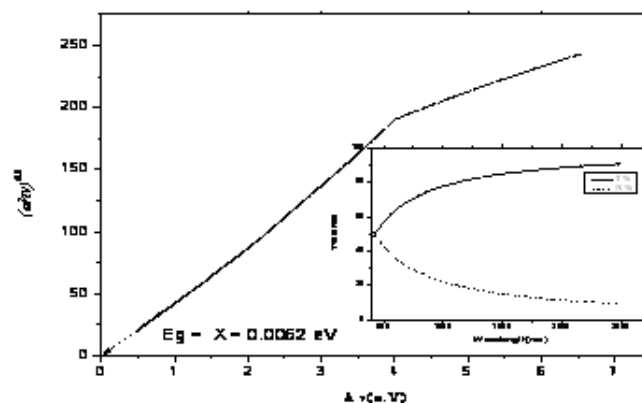
**Figure1b.** Relation between optical reflection and wave length for graphene thin films.

$$\alpha = \left(\frac{1}{d}\right) \ln\left(\frac{(1 - R)^2}{T}\right) \tag{1}$$

where d is the film thickness, R, T were the reflectance and transmittance of the films. The optical energy gap was determined from the absorption curve using the empirical equation (2):

$$\alpha = A(h\nu - E_g)^p \tag{2}$$

where A is a constant,  $E_g$  is the energy band gap,  $\nu$  is the frequency of the incident light waves and h is Planck's constant. The constant P takes different values depending on the kind of optical transition, P=0.5 for direct allowed transition (direct energy gap) and for indirect allowed transition the value of P equals 2.



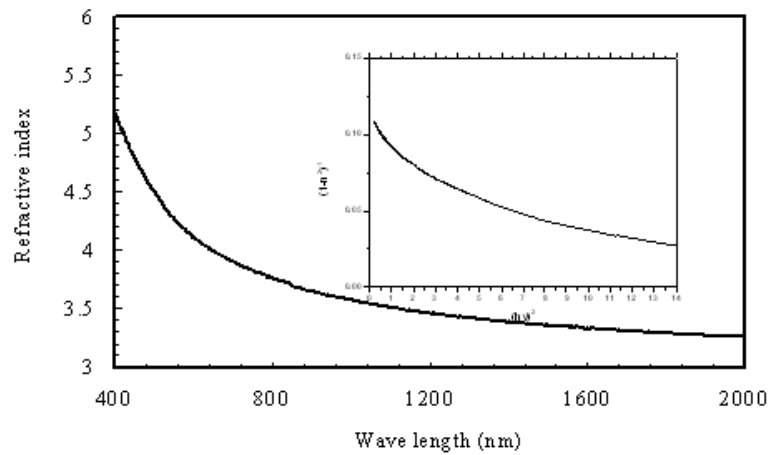
**Figure2.** Relation between  $(ah\nu)^{0.5}$  and photon energy  $(h\nu)$  for graphene thin films.

Figure 2 shows the relation between  $(\alpha hv)^{0.5}$  and photon energy  $(hv)$ , the indirect allowed optical energy gap for these films was determined from the extrapolation of the linear portion of the curves to x-axis as shown in Figure 2. From this figure it was found that, the graphene film had a indirect energy gap of 0.0062 eV.

The refractive index  $(n)$  for these films was calculated using the following equation [21]:

$$n = \frac{(1 + R) + \sqrt{4R - (1 - R)^2 K^2}}{(1 - R)} \tag{3}$$

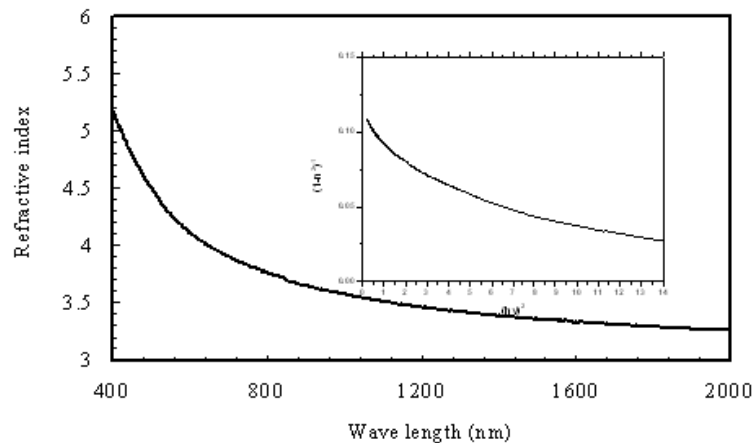
Figure 3 represents the refractive index dependence on the wave length for graphene thin films, from this figure; the refractive index decreases sharply at small wave length, while it has a constant value at high wave lengths. The obtained values of refractive index help us to calculate the single oscillator for the graphene layers. The single oscillator can be expressed by the Wemple–DiDomenico relationship [22]



**Figure3.** The refractive index dependence on wave length for graphene thin films

$$n^2(E) - 1 = \frac{E_o E_d}{E_o^2 - E^2} \tag{4}$$

where  $E$  is the photon energy,  $E_o$  is the single oscillator energy and  $E_d$  is the dispersion energy. The parameter  $E$  is a measure of the intensity of the inter-band optical transition. Plotting  $(n^2-1)^{-1}$  against  $(hv)^2$  as in figure 4 allows one to determine the oscillator parameters.  $E_o$  and  $E_d$  which can be directly determined from the slope which equals to  $(E_o E_d)^{-1}$ , and the intercept which equals to  $(E_o / E_d)$ . The calculated values of  $E_d$  was 1.46 eV while the value of  $E_o$  was 0.17 eV (as calculated from the above driven equations). The dielectric parameters for graphene were calculated from the values of the refractive index and the extinction coefficient using the following equations [23]:

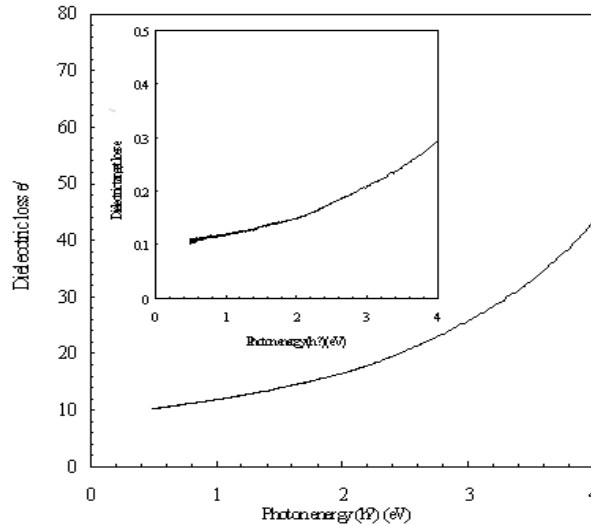


**Figure4.** Relation between  $(n^2-1)^{-1}$  and  $(\text{photon energy})^2$  for graphene thin films

$$\epsilon' = (n^2 + k^2) \tag{5}$$

$$\epsilon'' = [(n^2 + k^2)^2 - (n^2 - k^2)^{0.5}] \tag{6}$$

The relation between photon energy and both of the dielectric loss  $\epsilon'$  and dielectric tangent loss  $\epsilon''$  are shown in figures 5, and 6. From this figures it is clear that both  $\epsilon'$  and  $\epsilon''$  increase with increasing photon energy, while; optical conductivity (real part ( $\sigma_1$ ) and imaginary part ( $\sigma_2$ )) for these samples was calculated from equations 7 and 8 [24].



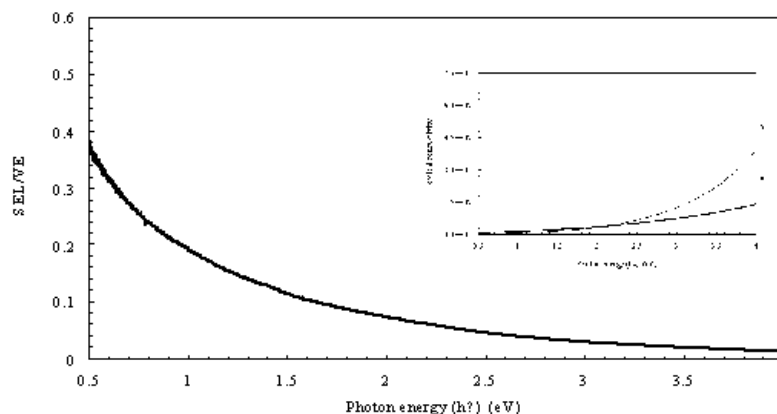
**Figure5** .The dielectric loss  $\epsilon'$  dependence on photon energy graphene thin films.

**Figure6**. The dielectric tangent loss  $\epsilon''$  dependence on photon energy graphene thin films.

$$\sigma_1 = \frac{(\epsilon'' \cdot c)}{(2 \cdot \lambda)} \tag{7}$$

$$\sigma_2 = \frac{(1 - \epsilon') \cdot c}{4 \lambda} \tag{8}$$

From figure 7; the optical conductivity increases with increasing the photon energy, this behavior could be attributed to high photon energy leads to increase of the electron mobility consequently to an increase in the optical conductivity of graphene layers. The optical transitions could be described by the Surface Energy Loss (SEL), which describes the electron transition in thin film and, the second parameter was Volume Energy Loss (VEL), which describe the electron transitions in the bulk materials [25], the value of SEL/VEL represent the electrons transitions in this material at low and high energy [25]. The values of both SEL and VEL can be calculated using the following equations [26]:



**Figure7**. The optical conductivity a) real part and b) imaginary part dependence on photon energy graphene thin layers.

$$VEL = \frac{\epsilon''}{\epsilon'^2 + \epsilon''^2} \tag{9-a}$$

$$SEL = \frac{\epsilon''}{(\epsilon' + 1)^2 + \epsilon''^2} \tag{9-b}$$

Where  $\epsilon'$  is the dielectric loss and  $\epsilon''$  is the dielectric tangent loss of the studied samples. The relation between SEL/VEL for these films and photon energy is presented in Figure 8. From this Figure it is clear that the SEL/VEL decrease with increasing the photon energy, this is because of the increase of the optical conductivity of this film with increasing photon energy according to the equation (7) and the relation [27]:

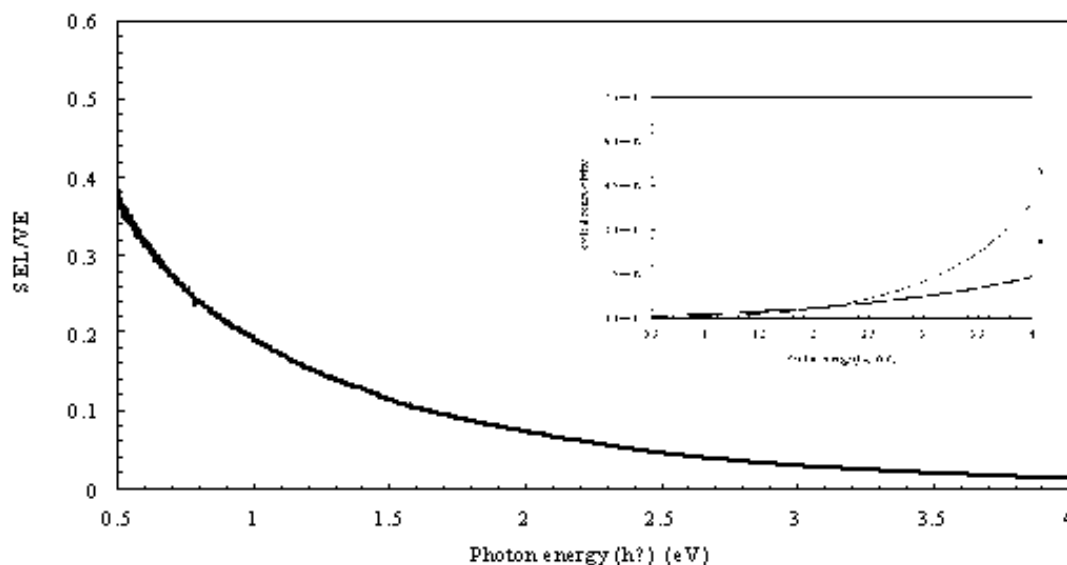


Figure8. The SEL/VEL dependence on photon energy graphene thin layers.

$$\sigma_T = \frac{c \alpha \sqrt{\epsilon'}}{4 \pi} \tag{10}$$

Where:  $\sigma_T$  is the total optical conductivity in  $\text{Ohm}^{-1} \cdot \text{cm}^{-1}$ ,  $\alpha$  is the absorption coefficient in  $\text{cm}^{-1}$  and is related to photon energy according to equation (2). The ratio of photoconductivity to the incident light intensity is termed photosensitivity  $S$  which can be calculated from the following equation [28]:

$$S = \frac{\sigma_T}{J} \tag{11}$$

Where  $\sigma_T$  is the total photoconductivity and  $J$  is the incident light intensity. Photosensitivity against wavelength of a halogen lamp is shown in figure 9 which indicates a decay of  $S$  with increasing wavelength  $\lambda$ . This phenomenon may be attributed to an increase in the number of free charged carriers'  $n$  by excitation of incident light till  $\lambda = 1500\text{nm}$  after that  $S$  tends to saturation. Figure 10-a, b, c shows the relation between optical resistivity against voltage and time at wavelengths 365nm, 254nm, and room light respectively. The figure shows a decrease in the resistivity with increasing both voltage and time, till a critical voltage equals 78m. Volts, after this voltage the optical resistivity increases. This phenomenon illustrated as the total free electron densities gradually increases with increasing applied voltage, and they become saturated above a critical voltage "78m.Volts" because all traps in the graphene layer at the high voltage are fully occupied by free electrons. The electron trapping rate corresponding to the increase of the total trapped electron density gradually increases with increasing applied voltage, as shown in figure 10, where the resistivity is inversely proportional to the current and directly proportional to the applied voltage, the free electrons injected from the electrode increase with increasing applied voltage, as shown in figure 11-a and could be deduced from the following relations [29]:

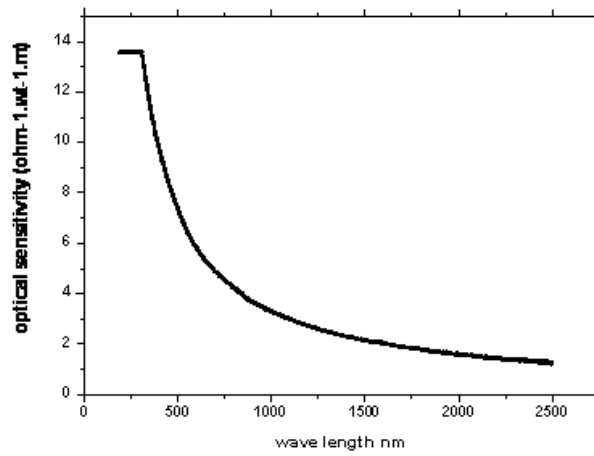
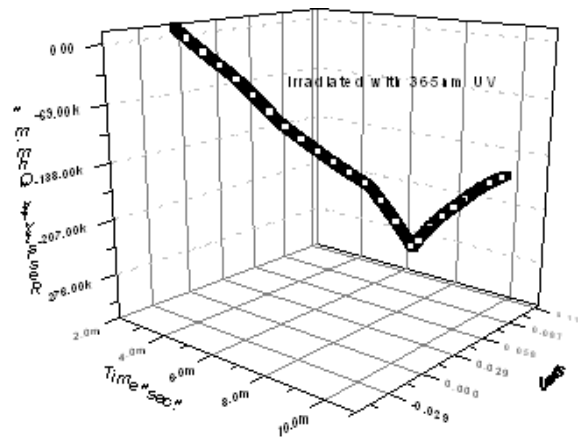
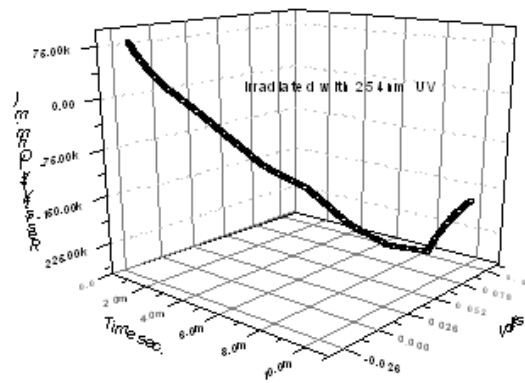


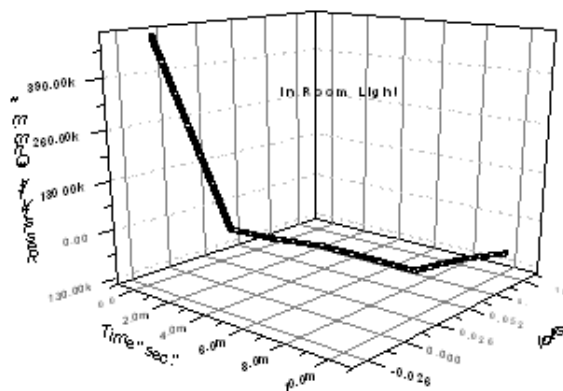
Figure9. The dependence of optical sensitivity on wavelength.



10a



10b



10c

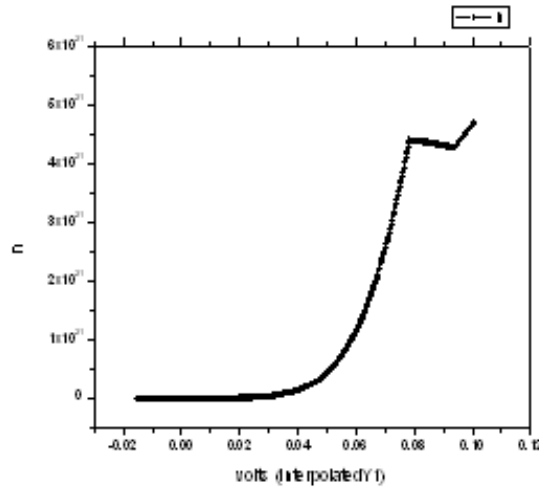


Figure11a. Density of free charged carriers.

$$n = N_c \left[ \frac{E_t}{KT} \right] \tag{12}$$

$$\theta = \frac{N_c}{N_t} \exp\left( -\frac{E_t}{KT} \right) \tag{13}$$

$$J = \frac{9 \epsilon_o \epsilon_r \mu \theta E^2}{8d} \tag{14}$$

Where  $\theta$  is the ratio between free carriers density and occupied traps density  $N_t$ ,  $N_c$  is the effective density of states,  $E_t$  is the energy level of carrier trap sites, where the free carriers is  $n$  and  $J$  is the current density,  $\mu$  is the charged carrier mobility,  $\epsilon_o$  is dielectric constant, and  $\epsilon_r$  is the dielectric permittivity. The number of traps occupied by the free electrons decreases results in the increase of the unoccupied traps as shown in figure 11-b. Even though the free electron density increases with increasing the applied voltage, the unoccupied trap density decreases at high voltages. The critical value of electron trapping rate and the applied voltage are proportional to the trap density  $N_t$  [30] according to equation (13) and as shown in figure 11-c. So the photocurrent rise is fully controlled by the carrier trapping. At longer times, rise of photocurrent depends on the excitation density [31] as represented in equation (14). It is shown that the polarization of volume traps alone is unable to describe the observed values and the behavior of the negative transient current. This fact shows the significant role of surface traps in capturing electrons which is the mechanism of negative current generation. This behavior is confirmed by figure 8 as surface energy loss is lower than volume energy loss and their ratio is less than one.

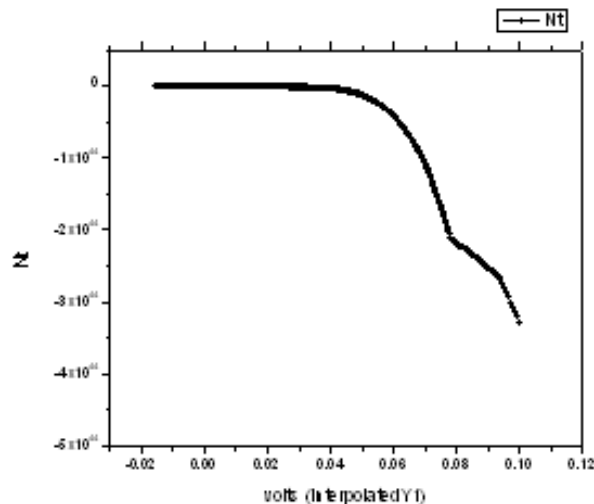
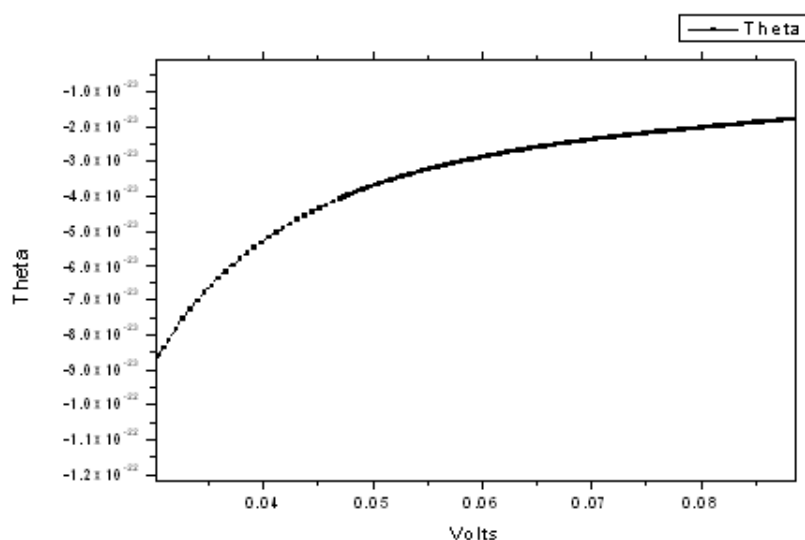


Figure11b. Density of occupied traps.



**Figure 11c.** Ratio between free electrons to occupied traps.

#### 4. CONCLUSION

The optical results showed that these samples had an allowed indirect energy gap of value 0.0062 eV. The refractive index was determined for these samples in order to determine both of the dielectric loss and dielectric tangent loss. The optical conductivity increases with increasing photon energy. The ratio of SEL/VEL dependence on photon energy was studied and showed that this ratio decreased with photon energy. Photosensitivity as a function of wavelength shows a decrease with increasing wavelength. The relative permittivity as a function of time and voltage shows an increase with time and voltage. Transient optical resistivity behavior was studied and the results confirmed the dependence of photo current on the density of traps.

#### REFERENCES

- [1] R.,Nair,etal. Science, **320**(5881), 1308, (2008).
- [2] C., Casiraghi etal Nano. Lett: **7**, 2711 (2007).
- [3] P.Blake etal, Appl.Phys.Lett 91, 063124(2007).
- [4] J.Yan, Y.Zhang, P. Kim, A. Pinczuk, Phys. Rev. Lett. **98** 166802(2007).
- [5] A.C. Ferrary etal., Phys. Rev. Lett.**97**187401/1-187401/4(2006).
- [6] K.S.Novoselov, etal, Nature **438**, 197-200(2005).
- [7] T.Gokus, etal, ACS. Nano **3**, 3963(2009).
- [8] G.Eda, etal Adv.Mater. **22**, 505 (2009).
- [9] X.Sun, etal, Nano. Res.**1**, 203(2008).
- [10] Z.Luo, P.M.Vora, E.J.Mele, A.T.C.Johnson, J.M.Kikkawa Appl. Phys. Lett. **94**, 111909(2009).
- [11] K.Ziegler, Phys. Rev. Lett. **97**, 266802(2006).
- [12] K. Ziegler, Phys. Rev. B **75**, 233407(2007).
- [13] Lei Liao, X.Duan, Materials Science and Engineering: R: Reports, **70**, 354-370(2010).
- [14] S.Sonde, etal, Physica E, **44**, 989-992(2012).
- [15] S.Ansari, E.P. Giannelis J Polym Sci Part B **47**, 888–97(2009).
- [16] N.Liu, F.Luo, H.Wu, Y.Liu, C.Zhang, Adv Funct Mater **16**, 1518–1525(2008).
- [17] W.S. Hummers, R.E. Offeman. J.Am.Chem.Soc. **80**(6), 1339(1958).
- [18] H.A. Becerril, etal . Nano **2**(3), 463–470(2008).
- [19] P.H.Berming, G. Hass (Ed.), Physics of Thin Films, Vol. **1**, Academic, New York, p. 69(1967).
- [20] M.Fadel, S.A.Fayek, M.O.Abou-Helal, M.M.Ibrahium, A.M.Shakra, J. Alloys and Compounds **48**, 5604-609(2009).



- [21] J.Aranda, J.L.Morenza, J.Esteve, J. M.Codina, Thin Solid Films. **120** 23-30(1984).
- [22] P.Nozieres, D.Pines, , Phys. Rev. **113**, 1254(1959).
- [23] J. Torris, J.I. Cisneros, G. Gordillo, F. Alvarez, Thin Solid Films. **289**, 238–41(1996).
- [24] B. Aleksandra, E. Djurić, Li. Herbert, Optics Communications **157** 72–76(1998).
- [25] A.H. Ammar, A.M. Frid and M.A.M.Sayam, Vacuum, 66, 27-38(2002).
- [26] G.L.Tan, L.K.DeNoyer, R.H.French, M.J.Guittet, M.Gautier-Soyer, Materials Science and Engineering **294–296** 867–870(2000).
- [27] A.H.Ammar, B.A. El-Sayed and E.A.El-Sayad, J. Material Science, **37** 3255-3260(2002).
- [28] P.S.Kireev, "Semiconductor Physics", Mir Publishers, Mosco, P522(1975).
- [29] J.H.Jung, T.W.Kim, Solid State Communications **149** (2009)1025-1028.
- [30] R. Fairman and, B.Ushkov, Semiconducting Chalcogenides II (2004) chapt 4, Elsevier Academic Press New York.
- [31] Kawae, etal., Appl.Phys.Lett.,94, (2009) 112904.