Nano-Graphene and Single-walled Carbon Nanotube Electrical and Optical Conductivity Properties by Terahertz Time-Domain Spectroscopy

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Abstract

This paper studies the graphene single-layer and single-walled carbon nanotube thin-films electrical and optical conductivity properties using terahertz time-domain spectroscopy. We analytically compare the graphene and single-walled carbon nanotube thin-films total transmission deposition on high resistivity silicon wafers as sheet conductivity function in same range terahertz spectroscopy of 0-3THz.

1. Introduction

Carbon nanostructures have been used in bio-systems because of great mobility to further electron transfer and reaction. Previously, far infrared spectroscopy used in order to obtain the nano-carbon crystal properties [1]. The main restriction was that it derived a complicated numerical transformation process of the Kramers-Kronig relationship. In an opposite position, terahertz (THz) time-domain spectroscopy (TDS) can enable the precise, simple and exact complex permittivity with high signal-tonoise ratio about 10000 compared to the far infrared and microwave spectroscopy. Recently, electromagnetic THz wave spectroscopy based on the pumped optical source has been most widely studied for nanostructure carbon [2-3]. The graphene optical characteristics in the terahertz frequency range is related to the intraband conductivity which relays on the total carrier concentration as well as the energy of carrier distribution [4]. The quick fast relaxation time of less than 1ps and slow recombination time greater than 1ps of graphene carriers as well as gapless energy spectrum in THz time-domain emission mixture pumped optical-probe energy of $\hbar\Omega$ can be archived for studying intraband and interband process[5]. The ac conductivity real part is commensurate to the absorption coefficient of energy photons with frequency ω and it is negative in the range of terahertz frequency[6]. The conductivity of graphene by THz frequency is relayed on two carrier specifications; concentration and distribution energy. Due to perfect characteristics of graphene such as high carrier density and high optical transparency through air-graphene-wafer components [7], the graphene conductivity can measure without fabrication and pattering of electrical contact by THz spectroscopy. In this work, we will study about the optical and electrical conductivity of single-layer graphene and single-walled carbon nanotube (SWCNT) using THz (0 to 3 THz) time-domain spectroscopy. Furthermore, a total transmission comparison of SWCNT and grapheme through the obtained sheet conductivity will be presented.

2. THz-TDS graphene configuration and methodology

This report analytically investigates to find the total transmission based on the relative power transmitted through graphene/Si sample and then compares the obtained results of total transmission with single-walled carbon nanotube as function of sheet conductivity. The graphene and SWCNT samples have a similar condition with multi-layer structure consisting of graphene [8] or SWCNT [9], silicon (Si) and air layers with various refractive indices of each layer. The THz reference pulses were generated by the femtosecond Ti:sapphire laser. The refractive index of high-resistivity Si resistivity and air is known by $n_S=3.42$ and $n_{air}=1$, respectively. The both devices (graphene and SWCNT) analysed by thin-film Fresnel coefficients and the Drude model.

$$TH_{Z}(\omega)_{output} = TH_{Z}(\omega)_{input} e^{\left(-\frac{d \alpha(\omega)}{2}\right)} \times e^{\left(i\frac{2\pi}{\lambda}n_{real}(\omega)d\right)}$$
(1)

Here, d is the sample thickness; λ is the free-space wavelength, $\alpha(\omega)$ is the power absorption coefficient and $n_{real}(\omega)$ is the real part of sample refractive index. The absorption coefficient can be calculated from the proportional in spectral phase magnitude between the input and transmitted signal through sample($\frac{THz(\omega)_{output}}{THz(\omega)_{input}}$). The real part of index of refraction can be obtained by phase delay $(\varphi_{output} - \varphi_{input})$ that introduced by

$$n_{real}(\omega) = 1 + \left[\frac{\varphi_{output} - \varphi_{input}}{d\omega c^{-1}}\right]$$
(2)

$$\alpha(\omega) = \frac{1}{d} \ln \frac{TH_{Z}(\omega)_{output}}{TH_{Z}(\omega)_{input}} = \frac{4\pi n_{img}}{\lambda}$$
(3)

Where φ_{input} is the input signal phase, φ_{output} is the output signal phase, c is the speed of light in vacuum and n_{img} is imaginary part of the index of refraction of the supposed sample ($n = n_{real} +$ in_{ima}). Following that, the real conductivity σ_{real} conductivity can be obtained

$$\sigma_{real}(\omega) = 2\omega\varepsilon_0 n_{real}(\omega) n_{img}(\omega) = c\alpha(\omega) n_{real}(\omega)\varepsilon_0$$
(4)

The graphene layer was treated as a zero-thickness conductive film and SWCNT thickness of 7um whereas the Si substrate is considered an optically thick dielectric medium. The transmission through the first interface (air-graphene/SWCNT-Si-air) and internal reflection from the graphene or SWCNT interface are

$$t(\sigma_s) = \frac{2}{n_s + 1 + Z\sigma_s} \tag{5}$$

$$r(\sigma_s) = \frac{\frac{n_s - 1 - Z\sigma_s}{n_s - 1 + Z\sigma_s}}{\frac{n_s - 1 - Z\sigma_s}{n_s - 1 + Z\sigma_s}}$$
(6)

Z is the vacuum impedance of 376.73 Ω . The ratio of the total transmission or relative transmission of the graphene/Si or SWCNT/Si sample is given by

$$T_{relative}(\sigma_s) = \left(\frac{t(\sigma_s)}{t_{13}}\right)^2 \frac{1 - (r_{34}r_{31})^2}{1 - (r_{34}r(\sigma_s))^2}$$
(7)

In this case, the incident and reflection and transmission angles (β) are equal to zero ($cos\beta=I$). The Freshel's coefficients of the electrical field that is parallel to the incident sample are $t_{ij} = \frac{2n_i}{(n_i + n_j)}$ and $r_{ij} = \frac{(n_i - n_j)}{(n_i + n_j)}$ and refractive indices of $n_1 = n_4 = n_{air}$ and $n_3 = n_5$. t_{13} is the transmission through second

interface from air to silicon wafer, r_{34} and r_{31} are internal reflections inside the silicon wafer.

3. Results and discussion

The amplitude and phase of THz electromagnetic wave will be changed (THz transmitted signal) as results of pump photon energy into grapheme and SWCNT samples (equation (1)). In case of grapheme single-layer as an experimental setup, the optical contrast and conductivity of graphene sample will change as well. The power transmission measured in range of 0.36 to 0.41 corresponding to heterogeneous graphene sample [8]. Also, the power average transmission Si as reference power evaluated to be 0.57. From equation (5), we have obtained the amount of sheet conductivity as function of transmission through the graphene sample as demonstrated in Fig.1 (left). Hereby, the range conductivity of 1.2×10^{-3} and $3 \times 10^{-3} \Omega^{-1}$ is obtained. After calculating the internal reflection of graphene interface and according to equation (7), we have simulated the relative transmission according to the Frensel coefficients $(T_{relative}(\sigma_s) = \left(\frac{t(\sigma_s)}{t_{13}}\right)^2 \frac{1 - (r_{34}r_{31})^2}{1 - (r_{34}r(\sigma_s))^2}$) versus obtained results in Fig. 1 (left) as shown in Fig. 1 (center). However, by dividing the graphene sample power transmission over Si substrate reference power $(T_{relative} = \frac{T_{graphene-Silicon}}{T_{silicon}})$, the local conductivity is between 1.7×10^{-3} and $2.4 \times 10^{-3} \Omega^{-1}$. The both experimental and analytical results relative transmission results are same in average conductivity of $2.04 \times 10^{-3} \Omega^{-1}$. In other words, we can assume the average sheet conductivity as reference point in order to obtain the average total transmission through the graphene sample even if using theoretical solution. In order to obtain the sheet conductivity of SWCNT with sample thickness d as same way of the graphene sample, we derive the real sheet conductivity σ_s from conductivity σ ($\rho_s =$ $d/\sigma = 1/\sigma_s$). Fig. 1 (right) shows a comparison between total or relative transmission of both samples in Ref. 8 (garphene mounted on Si) and Ref. 9 (SWCNT mounted on Si) as sheet conductivity function in the same THz frequency of 0 to 3 THz. The results represent that the total transmission of SWCNT sample a little more than total transmission of graphene sample because of the number of surface defects exists in graphene nanostructure. Jung et al. [1] studied in terms of a comparison between different carbon nanostructures for the real conductive. Their results show that the real conductivity is strongly depended on the crystallinity of the graphitic layers and the number of surface defects in assumed carbon nanostructure. Due to this reason, the conductivity is declined by ordering single-walled>double-walled>multi-walled \approx graphene nanotubes.

4. Conclusion and future works

We have analytically studied the THz time-domain spectroscopy single-layer graphene sheet conductivity mounted on high-resistivity and high refractive index silicon wafer using the Fresnel coefficients and the Drude mode. We have also found that the total transmission of SWCNT sample is higher than graphene sample in the same sheet conductivity due to the surface defects in graphene nanostructure. The future works can be obtaining the total transmission of graphene and nano-carbon-structure as function of frequency. Beside that, an investigation through doping level of uniform graphene conductivity and spatial variation in conductivity by utilizing THz-TDS would be meaningful in future.



Fig. 1. (left) Sheet conductivity versus graphene sample transmission (center) relative transmission versus sheet conductivity and (right) comparing the total transmission between the SWNCT and graphene samples versus sheet conductivity.

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