AC Conductivity Parameters of Graphene Films with THz Spectroscopy*

W-D. Zhang and E. R. Brown
Terahertz Sensor Lab, Depts. of Physics and Electrical Engineering, Wright State University, Dayton, OH, 45435
wzzhang@fastmail.fm

Phi H. O. Pham and P. Burke
Department of Electrical Engineering and Computer Science, University of California, Irvine, CA, 92697

Abstract—The complex conductivity of CVD-grown graphene films between 0.1 and 1.6 THz are obtained using a non-destructive THz etalon transmittance technique. Critical parameters such as ionized-impurity scattering width and chemical potential are derived. The technique can be extended to extract the complex ac conductivity parameters of other thin conducting films or 2DEG materials with high sheet conductance.

Keywords— Graphene; conductivity; THz; Transmittance;2D

I. Introduction

The complex conductivity of graphene or other two dimensional (2D) materials with zero or narrow band gaps involves two type transitions: intraband and interband. The intraband and interband conductivities can be calculated from the material's band Hamiltonian along with several key parameters such as the temperature *T*, the chemical potential μ, as well as the scattering parameter Γ. For example, the conductivity of graphene is written in the summation of [1]

$$\sigma = \sigma_{intra} + \sigma_{inter} \qquad (1)$$

The Drude-Boltzmann type intraband conductivity is

$$\sigma_{intra} = \frac{2ie^2}{\pi}\frac{k_B T}{(\omega + i\tau^{-1})}\ln[2\cosh(\frac{\mu}{2k_B T})] \qquad (2)$$

where k_B is the Boltzmann constant, e is the electron charge, is the Planck constant, and $\omega = 2\pi f$ is the angular frequency.

The optical transition conductivity is

$$\sigma_{inter} = \frac{ie^2\omega}{\pi}\int_0^\infty d\varepsilon \frac{F(-\varepsilon) - F(\varepsilon)}{(\omega + i\delta)^2 - 4\varepsilon^2} \qquad (3)$$

where $F()$ is the Fermi function and ε is the band energy. δ is a mathematical quantity $\delta \to 0$.

Hence, knowing the critical parameters μ, Γ, one should be able to calculate the conductivity, which is often necessary for design of high-speed electronic devices or long-wavelength optical devices. In this paper, we discuss how to derive the ac parameters with a THz Etalon Transmittance technique, and apply it to characterize CVD-grown graphene films.

II. THz Etalon Transmittance Measurments

First, 2D film is mounted onto a THz-transparent substrate with a few hundred micron thickness. For example, CVD - grown graphene film was transferred from copper coil to a high resistivity Si substrate through an poly-methyl methacrylate (PMMA) deposition-copper etching-acetone washing procedure, and followed by an annealing process. The high resistivity silicon was used as thin film "carrier" because it has little THz absorption, and its dielectric constant has almost none frequency dependence at the lower THz band.

Then, the Etalon- a substrate with the graphene film attached - was placed into the beam path of a frequency-domain Emcore PB7100 transceiver for transmittance measurements. (Fig. 1) The operation of this spectrometer is based upon two-laser photomixing. It produces THz frequency radiation continuously from 0.1 to 1.6 THz in steps of 500 MHz. Note the minimum resolution can even reach the level of 100 MHz if application requires. The dynamic range is 80 dB at 0.1 THz and 40 dB at 1.0 THz.

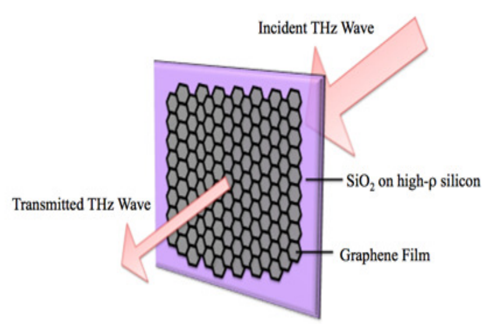

Figure 1 The THz transmittance measurements of 2D film mounted on the Si substare.

*This material is based upon work supported by, or in part by, the U. S. Army Research Laboratory and the U. S. Army Research Office under the contract number W911NF-11-1-0024.

Three spectrum were measured: background $P_B(f)$ (without the sample in the beam path), signal $P_S(f)$ (with the sample in the beam path) and noise $P_N(f)$ (with a metal blocking the beam path). Finally, the transmission was calculated from: $T_e=[P_S(f)-P_N(f)]/[P_B(f)-P_N(f)]$. All the measurements were performed at the room temperature, i.e T=300K.

III. MODELING

Transmission matrix method is applied for analysis of the transmittance data. [2] As the THz beam was incident to the etalon normally, the transmittance is calculated with

$$T_e = \left| \frac{t_1 t_2 \exp(-jk_s L)}{1 + r_1 r_2 \exp(-2jk_s L)} \right|^2 \tag{3}$$

where L is the thickness of the Si substrate, k_s is the wavevector inside the substrate. k_s is equal to $2\pi\sqrt{\varepsilon_s} f / c$, where ε_s is the dielectric constant of silicon and c is the light speed. r_1 is the reflection coefficient at the air/Si interface, and t_1 is the transmission coefficient at the air/Si interface. They are given by

$$r_1 = \frac{1-\sqrt{\varepsilon_s}}{1+\sqrt{\varepsilon_s}} \tag{4}$$

$$t_1 = \frac{2}{1+\sqrt{\varepsilon_s}} \tag{5}$$

r_2 and t_2 are the coefficients of reflection and transmission at the Si/graphene and graphene/air interfaces, respectively [1], and given by

$$r_2 = \frac{\sqrt{\varepsilon_s} - (\sqrt{\varepsilon_g}+1)}{\sqrt{\varepsilon_s} + (\sqrt{\varepsilon_g}+1)} \tag{6}$$

$$t_2 = \frac{2\sqrt{\varepsilon_s}}{\sqrt{\varepsilon_s} + (\sqrt{\varepsilon_g}+1)} \tag{7}$$

where $\varepsilon_g = \mathrm{Re}\{\varepsilon_g\} + j\,\mathrm{Im}\{\varepsilon_g\}$ is the complex dielectric constant of graphene. Equations (6) and (7) are obtained strictly from the Maxwell equations along with a set of boundary conditions at the interface between Si and graphene. [1] For some of 2D materials, boundary conditions can become complicated, Eqns (6) and (7) may be revised. The complex sheet conductance is computed as

$$\sigma_g = \frac{\sqrt{\varepsilon_g}}{377} \tag{8}$$

IV. RESULTS

A typical transmittance spectrum is plotted in the upper part of Fig. 2. It shows multiple interference peaks and valleys. The peak-valley ratio is pronounced due to the long wavelength of THz radiation. This can be examined from the frequency separation between two adjacent peaks, which is $\Delta f = c/2L\sqrt{\varepsilon_s}$. With the thickness of substrate being a few hundreds microns, the separation is estimated in the order of 100GHz.

In Equations (3)-(8), the thickness of silicon substrate was measured L=392 μm, and the dielectric constant of silicon is accurately known in the THz region ε_s = 11.65. [3] $\mathrm{Re}\{\varepsilon_g\}$ and $\mathrm{Im}\{\varepsilon_g\}$ are unknown, so they are the fitting parameters. The fitting regions are designated such that each contains a center frequency where a peak is located, and two end frequencies where the adjacent valleys are located (Fig. 2). By curve-fitting these regions, one can get spot conductivities. This technique is accurate because each fitting includes at least 100 data points thank to the 500-MHz frequency resolution of the THz spectrometer while there are only two fitting parameters.

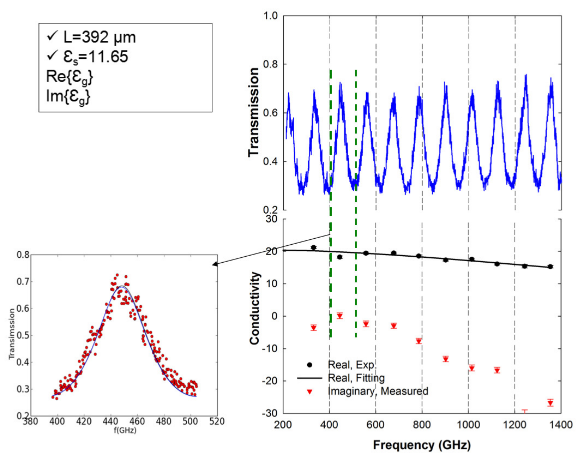

Figure 2 On the top of the right is the transmission spectrum; on the bottom is the extracted conductivity with unit of $e^2\pi/2h$. On the left is the example of fitting on one transmission peak.

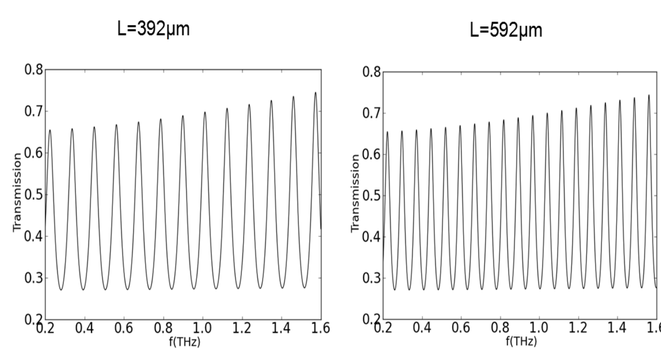

Figure 3 More spot-conductivities can be obtained by increasing the thickness of the substrate.

A Python numerical package was used for data processing. The extracted conductivity, both real and imaginary parts, is shown in the lower part of Fig. 2. The frequencies of the spot conductivities are where the peaks of transmittance are located. To get more spot-conductivities, one can properly increase the thickness of the substrate, as is illustrated in Fig. 3 with calculations based upon Eqns (2)-(8).

Furthermore, the extracted real conductivity is compared to the Drude type intraband Eq. (2). This step enables the derivation of the ac parameters $\Gamma = 4.59$ meV and $|\mu| = 0.15$ eV. The fitting curve is plotted in Fig. 2, showing an excellent consistency between the measurements and the Drude theory.

Because the THz photon energy is small and the Fermi level is large, the interband conductivity is less significant than the intraband one, and thus isn't included in the fitting above.

V. CONCLUSIONS

A simple THz technique to derive the ac conductivity and related parameters is demonstrated. The technique requires none-contact of the test samples, and thus is not destructive. This technique can be applied to study the dielectric properties of 2D films.

REFERENCES

[1] L. A. Falkovsky, "Optical properties of graphene and IV-VI semiconductors", Physics-Uspekhi, vol. 51, pp. 887–897, 2008.

[2] M. Born and E. Wolf, Principles of Optics: Electromagnetic Theory of Propagation, Interference and Diffraction of Light, Cambridge University Press, 1993.

[3] P. H. Bolivar, M. Brucherseifer, J. G. Rvivas, R. Gonzalo, I. Ederra, A. L. Reynolds, M. Holker and P. Mgt, "Measurement of the dielectric constant and loss tangent of high dielectric-constant materials at THz frequencies ", IEEE Transaction on Microwave Theory and Techniques, 51, pp. 1062–1066, 2003.