

Extracting Complex Optical Properties of Ultra-thin Conductors Using Time-domain THz Spectroscopy

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Abstract—We use terahertz time-domain spectroscopy (THz-TDS) in reflection geometry to investigate the complex optical properties of ultra-thin (i.e., $\lesssim 10$ nm) conductors from 0.3 to 3 THz. We discuss the advantages of this approach, and report complex optical properties of graphene and a 9 nm Ti thin-film.

I. INTRODUCTION

EXTRÊMELY thin materials can have properties quite unlike—and often far superior to—their bulk counterparts. For this reason, increased attention is being paid to two-dimensional materials in the search for novel devices, including those that are functional at THz frequencies. Graphene is currently the most actively studied of these materials, as it shows potential for plasmonic, optoelectronic, and metamaterials applications in the THz frequency range [1]. To realize many of these applications, the complete optical properties of graphene in this spectral range must be well understood. Here we extract and compare the complex THz optical properties of a single graphene layer and a Ti thin-film using THz-TDS in a self-referencing reflection geometry.

II. ANALYSIS AND RESULTS

As shown in Fig. 1, a p -polarized THz pulse is incident from below on an undoped Si substrate at an angle $\theta_i = 10^\circ$. We refer to the first interface encountered by the THz pulse as the “front”, and the opposite side as the “back”. A portion of

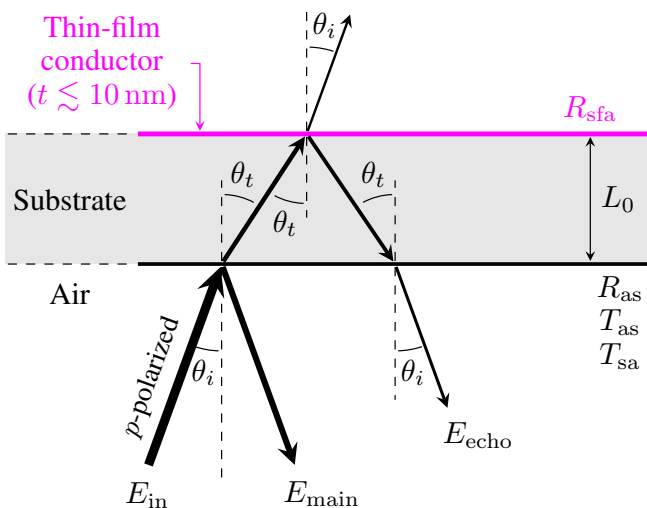


Figure 1. Measurement geometry with thin-film sample present. Optical paths of incident THz pulse, main reflection, and echo signals are shown.

the incident pulse will be reflected from the front (main), and the rest of the signal will propagate into the substrate with a refraction angle θ_t . Some of this transmitted signal will then reflect from the back interface (echo) and exit out the front of the sample. Subscripts a, s, and f denote air, substrate, and thin-film, respectively.

To analyze the signals, we calculate a transfer function \tilde{H} using the main reflection as the input and the echo as the output.

$$\tilde{H} = \frac{|E_{\text{echo}}|}{|E_{\text{main}}|} \exp(-j(\phi_{\text{echo}} - \phi_{\text{main}})) \quad (1)$$

We calculate the refractive index of the substrate n_s from the argument of \tilde{H} , and the extinction coefficient of the substrate κ_s from the magnitude of \tilde{H} . We note that since the main and echo signals are obtained simultaneously—without disturbing the substrate—misalignment and positioning errors are effectively eliminated. The delay of the echo signal is determined by referencing its earlier self (the main reflection), and since the substrate thickness is known, the optical properties of the substrate can be accurately determined. We then repeat the measurement with a sample atop the substrate. The presence of the sample modifies the boundary conditions at the substrate-sample interface, indicated by R_{sfa} in Fig. 1. From these two measurements, and an approach similar to that described in [2], we calculate the Fresnel reflection coefficient using the equation

$$R_{\text{sfa}} = \tilde{H}_f \frac{R_{\text{as}}}{T_{\text{as}} T_{\text{sa}}} \exp\left[j\tilde{n}_s \frac{\omega L_{\text{eff}}}{c}\right], \quad (2)$$

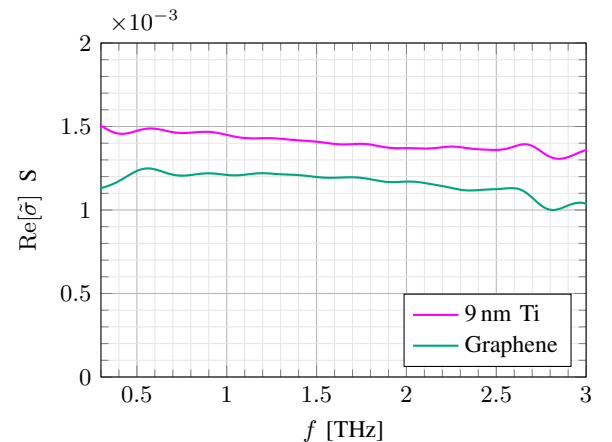


Figure 2. Real part of surface conductivity for a Ti thin-film and single-layer graphene on undoped Si with a 300 nm oxide layer.

in which $L_{\text{eff}} = L_0 / \cos \theta_t$ is the effective path length through the substrate. We then calculate the complex conductivity $\tilde{\sigma}$ of the sample from R_{sfa} using the following equation [3].

$$R_{\text{sfa}} = \frac{\tilde{n}_b \cos \theta_i - n_a \cos \theta_t - \cos \theta_i \cos \theta_t \tilde{\sigma} Z_0}{\tilde{n}_b \cos \theta_i + n_a \cos \theta_t + \cos \theta_i \cos \theta_t \tilde{\sigma} Z_0} \quad (3)$$

The real parts of $\tilde{\sigma}$ for single-layer graphene and for the Ti thin-film obtained by this analysis are plotted in Fig. 2. We note that the values obtained for the Ti thin-film are in agreement with those reported in [4] using transmission geometry.

Of the many optical properties related to the complex conductivity, the two values of most interest are the relative permittivity and the absorption coefficient, since they give insight into the behaviour of thin conducting films in the presence of THz radiation. The absorption coefficient α is directly proportional to the imaginary part of the complex refractive index of the thin film (κ_f). It is given by

$$\alpha = \frac{2\omega}{c} \kappa_f. \quad (4)$$

Relative permittivity ϵ_r is the real part of the dielectric function, which is related to the complex conductivity as follows [5]:

$$\epsilon_r = \text{Re}[\tilde{\epsilon}] = -\frac{\text{Im}[\tilde{\sigma}]}{\omega \epsilon_0 t_f}. \quad (5)$$

Extracted values for these two properties are plotted in Fig. 3.

Given that the surface conductivity of the Ti thin-film is higher than that of single-layer graphene (Fig. 2), this suggests that Ti absorbs more of the THz radiation than graphene. While this may be true of the film as a whole, we find that the absorption coefficient of graphene is considerably larger than that of Ti in the THz range (Fig. 3a). Another important observation is that the relative permittivity of graphene is negative, whereas that of thin-film Ti is positive (Fig. 3b). This explains why single-layer graphene supports surface plasmons in the THz range [6] but a conventional metal thin-film, such as Ti, does not.

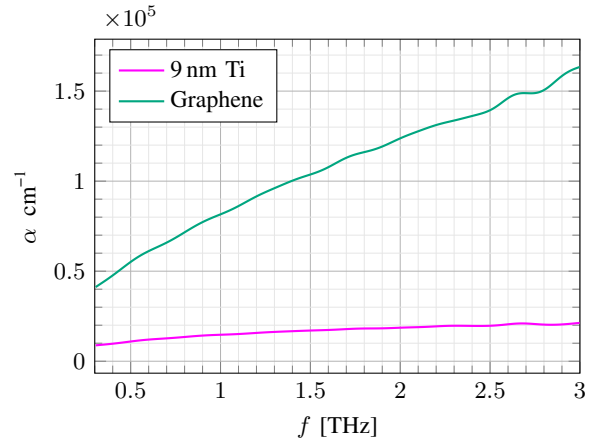
Since graphene behaves as a metal, the complex conductivity should be described by the Drude equation:

$$\tilde{\sigma} t_f = \frac{\sigma_{\text{dc}}}{1 - j\omega\tau}. \quad (6)$$

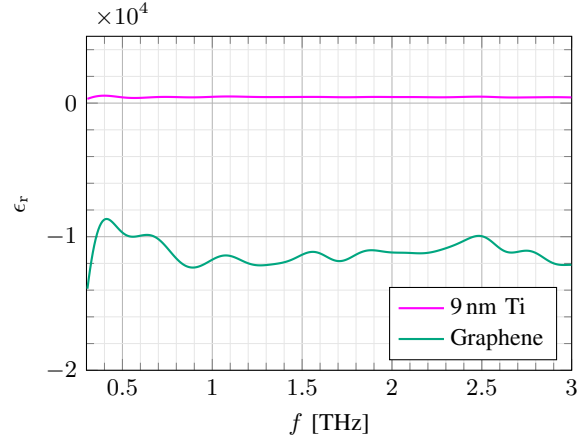
From this equation, we find that for our CVD-grown monolayer graphene transferred onto an undoped Si wafer with a 300 nm oxide layer, the mean scattering time $\tau \approx 29$ fs, the dc conductivity $\sigma_{\text{dc}} \approx 4 \times 10^4 \text{ S cm}^{-1}$, and the Fermi energy $E_F \approx 0.38 \text{ eV}$. These values are similar to ones that have been previously obtained by transmission and reflection measurements of graphene on a quartz substrate [7].

III. CONCLUSION

Here we report a robust method to determine complex optical parameters for ultra-thin conductors—including one-atom-thick graphene—using a self-referencing THz-TDS measurement. Using the Drude model, we also determine the electrical characteristics of graphene, namely the dc conductivity and Fermi level, for 0.3 to 3.0 THz. Our approach ensures reliable values even for ultra-thin conductors, which is critical for the understanding of materials at the forefront of new device research, in particular graphene-based THz plasmonics.



(a) Absorption coefficient for 9 nm Ti and single-layer graphene.



(b) Relative permittivity for 9 nm Ti and single-layer graphene.

Figure 3. Optical parameters of single-layer graphene and a Ti thin-film.

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