Invited Paper

Accurate extraction of ultrabroadband terahertz conductivity of thin films

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Abstract: A thin film conductivity extraction algorithm has been developed for ultrabroadband terahertz time-domain spectroscopy (THz-TDS). To verify the accuracy of the extraction algorithm, the finite difference time-domain (FDTD) simulation has been performed. It turns out the complex conductivity extracted with the algorithm matches the predefined Drude response very well, while the conductivity calculated with the thin film equations differs dramatically. Finally, the extraction algorithm is applied to the transparent conducting oxides, and the significant divergence between the conductivities extracted with the two methods emphasizes that it is crucial to use the extraction algorithm to characterize the broadband terahertz conductivity of thin films.

Keywords: Thin films, Optical properties, Spectroscopy, Terahertz

Doi:

1. Introduction

Terahertz time-domain spectroscopy (THz-TDS) has been demonstrated as a powerful tool for characterizing material properties [1]. The terahertz spectra are usually Fourier transformed from their time traces. By comparing the terahertz spectra through sample to air, the optical properties of the material can be directly calculated. However, for certain samples, due to the low refractive index and thin sample thickness, the Fabry-Perot (FP) reflections inside the sample cannot be easily distinguished. Efforts have been done to solve the problem, even for samples with unknown thickness [2-4]. Yet, these works are mainly focused on single sample slab. Meanwhile, THz-TDS has been widely utilized to measure the conductivity of thin films [5-9]. It is usually straightforward to calculate the complex conductivity with thin film equations [10]. However, the condition for thin film equations does not necessarily hold for all the thin film samples [8]. It requires an accurate extraction method to be universally adequate for thin conducting films without limitations.

In this study, based on the same principle of extracting optical properties of slab samples, the thin film conductivity extraction algorithm has been developed. To verify the accuracy of the method, the finite difference time domain (FDTD) method has been performed to simulate the THz-TDS through a thin conducting layer on top of substrate with known Drude parameters [11]. The extracted complex conductivity matches very well with that of the predefined Drude response. In contrast, the complex conductivity calculated with the thin film equations deviates dramatically from the designed Drude conductivity, as the thin film criterion is not satisfied; the error for the real part is more than 30% and 40% for the imaginary part. Furthermore, the conductivity extraction algorithm has been applied to TCOs, and the significant divergence between the conductivities calculated with the two methods indicates it is crucial to use the extraction algorithm to derive the terahertz conductivity of thin films in broadband THz-TDS.

2. Conductivity extraction algorithm

To measure the frequency dependent conductivity of thin films in broadband terahertz time-domain spectroscopy, the thin film with a thickness of less than hundreds of nanometers, is usually deposited on top of a high resistivity silicon substrate, as shown in Fig. 1. It is convenient to treat the thin film as a conducting layer with no thickness as long as the thin film condition $n\omega d / c \ll 1$ is satisfied. That is the phase retardation caused by the thin film which can be ignored, then the thin film conductivity can be directly calculated through the Tinkham thin film equations [10]

$$\sigma_1(\omega) = \frac{n_{sub}+1}{Z_0 d}\left(\frac{\cos(\theta_{exp}(\omega))}{A_{exp}(\omega)} - 1\right) \tag{1}$$

$$\sigma_2(\omega) = -\frac{n_{sub}+1}{Z_0 d} \cdot \frac{\sin(\theta_{exp}(\omega))}{A_{exp}(\omega)}, \tag{2}$$

Where, $\sigma_1(\omega)$ and $\sigma_2(\omega)$ are the real and imaginary part of the complex conductivity, n_{sub} is the refractive index of the substrate, and $A_{exp}(\omega)$ and $\theta_{exp}(\omega)$ are amplitude and phase of the experimental transfer function E_s / E_{ref} respectively. This equation is mostly used to extract the complex conductivity of thin films at low terahertz frequencies. However, the criterion $n\omega d / c \ll 1$ is not necessarily satisfied for thin film samples, especially, for samples with high refractive index or at high terahertz frequencies.

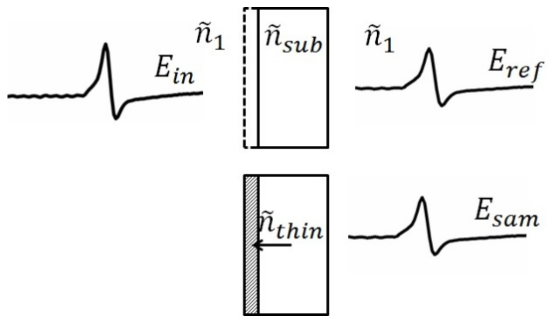

Fig. 1. Terahertz pulses transmitted through a substrate E_{ref} and a thin film on top of the substrate E_s .

Instead of treating the thin film as a conducting layer of no thickness, the complete transfer function should be considered without phase approximation. As shown in Fig.1, the reference terahertz pulse transmitted through the air-substrate-air interfaces is

$$E_{ref} = \exp(j\tilde{n}_1 \omega d_{thin} / c)\tilde{t}_{1s} \exp(j\tilde{n}_{sub}\omega d_{sub} / c)\tilde{t}_{s1} \cdot E_{in}, \tag{3}$$

Accordingly, the sample terahertz pulse transmitted through the air-thin film-substrate-air is

$$E_{sam} = \tilde{t}_{1thins} \exp(j\tilde{n}_{sub}\omega d_{sub} / c)\tilde{t}_{s1} \cdot E_{in}, \tag{4}$$

Where, $\tilde{t}_{1thins}$ is the amplitude transmission coefficient from air through the thin film to the substrate including the Fabry-Perot (FP) reflections inside the thin film [12], which is

$$\tilde{t}_{1thins} = \tilde{t}_{1thin}\tilde{t}_{thins} \exp(j\tilde{n}_{thin}\omega d_{thin} / c)\big/(1 + \tilde{r}_{1thin}\tilde{r}_{thins} \exp(2j\tilde{n}_{thin}\omega d_{thin} / c)), \tag{5}$$

Where, $\tilde{t}_{ij} = 2\tilde{n}_i / (\tilde{n}_i + \tilde{n}_j)$ and $\tilde{r}_{ij} = (\tilde{n}_i - \tilde{n}_j) / (\tilde{n}_i + \tilde{n}_j)$ are the complex amplitude transmission coefficient and reflection coefficient from material $\tilde{n}_i$ to material $\tilde{n}_j$. Thus, the entire theoretical transfer function is

$$E_{trans} = E_{sam}\big/E_{ref} = \tilde{t}_{1thins}\big/\tilde{t}_{1s} \exp(-j\tilde{n}_1\omega d_{thin}\big/c) = A_{theo}(\omega)\exp(j\theta_{theo}(\omega)). \tag{6}$$

Then, the complex conductivity of the thin film can be extracted to minimize the error function [2]

$$\Delta(\omega) = (A_{theo}(\omega) - A_{exp}(\omega))^2 + \left|\theta_{theo}(\omega) - \theta_{exp}(\omega)\right| \tag{7}$$

by simultaneously finding the optimal real and imaginary parts with the standard Nelder-Mead Simplex Method [13]. The initial guess for the complex conductivity can be calculated from the thin film equations Eq.(1) and Eq.(2) by[14]

$$n = (1/2(\varepsilon_1^2 + \varepsilon_2^2)^{1/2} + \varepsilon_1/2)^{1/2} \tag{8}$$

$$k = (1/2(\varepsilon_1^2 + \varepsilon_2^2)^{1/2} - \varepsilon_1/2)^{1/2} \tag{9}$$

Where, $\varepsilon_1 = \varepsilon_b - \sigma_2/(\varepsilon_0\omega)$ and $\varepsilon_2 = \sigma_1/(\varepsilon_0\omega)$ are the real and imaginary part of the dielectric function and ε_b is the background dielectric function. Finally, the complex conductivity of the thin film can be extracted from the complex refractive index through

$$\sigma_1 = 2nk\varepsilon_0\omega \tag{10}$$

$$\sigma_2 = (\varepsilon_b - n^2 + k^2)\varepsilon_0\omega. \tag{11}$$

3. Verification of the algorithm

With a thin film sample to be explored, estimating the accuracy of the extraction algorithm is challenging. Here, the finite difference time-domain (FDTD) method is used to simulate terahertz time-domain spectroscopy. For the simulation, a 200*nm* thick thin film with a specified Drude response is deposited on top of high resistivity silicon substrate with refractive index of 3.417 [15]. To be realistic, the Drude parameters are chosen comparably to that of the transparent conducting oxides [8], as represented in table 1. To be simplified, the silicon-air interface is ignored since it has no contribution to the whole transfer function.

Tab. 1 Parameters for the Drude response

DC conductivity σ_{DC} (10^3S/cm)	Carrier concentration N/cm^{-3}	Electron effective mass m^* (m_e)	Scattering time τ (ps)
2.4874	1.0×10^{19}	0.01	$1/(36\pi)$

The FDTD simulation process is the same as performing a normal THz-TDS experiment, the reference and sample terahertz pulses are recorded in Fig. 2. Both the reference and the sample pulses cover the spectra up to 20 *THz*.

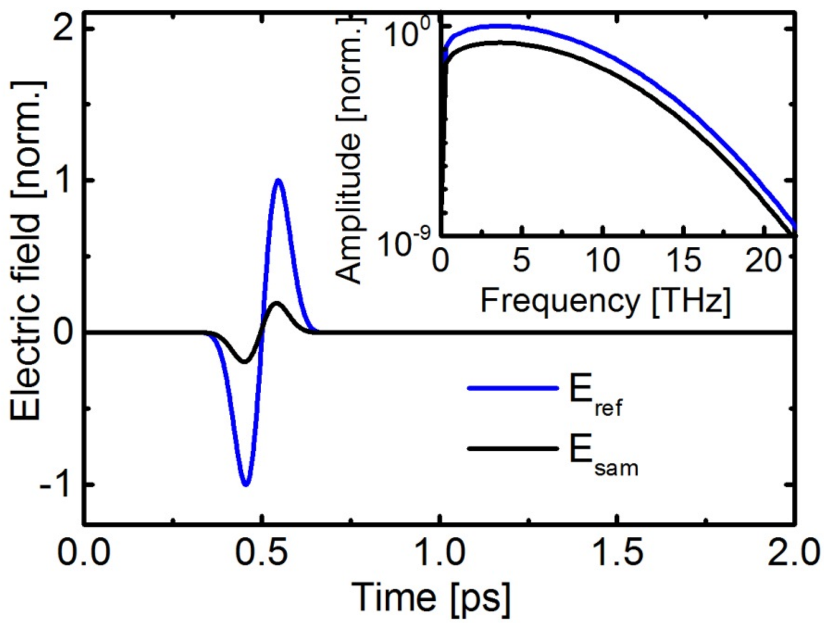

Fig. 2 Terahertz pulses transmitted through the substrate and the thin film sample. The inset shows the corresponding Fourier transformed spectrum.

The extracted conductivity from the algorithm well matches the input data, presented in Fig. 3(a); while the value from the thin film equations differs significantly in the whole frequency range, which results in considerable error for the scattering time, as shown in Fig. 3(b). The frequency dependent scattering time extracted with the algorithm is constant at 8.86 *fs*, while the value from the thin film equations is slightly frequency dependent with an average of 6.0 *fs*, corresponding to an error of 32%. At the low frequencies in Fig. 3(a), the real parts of the conductivity tend to overlap at σ_{DC}, while the imaginary parts are still different dramatically due to the small value, as indicated in Fig. 3(c). From 0.5 *THz* to 20 *THz*, the error induced by the thin film equations increases to 40% for the real part and reduces from more than -30% to -7% for the imaginary part. It is obvious that the criterion for thin film equations is not satisfied for the whole spectra region, as shown in Fig. 3(d). The maximum value of $n\omega d_{thin}/c$ is almost 0.42 at 10 *THz*, even in the low frequency region around 0.5 *THz*, and it is about 0.15. In short, the complex conductivity extracted with the algorithm matches very well with the predefined ac conductivity.

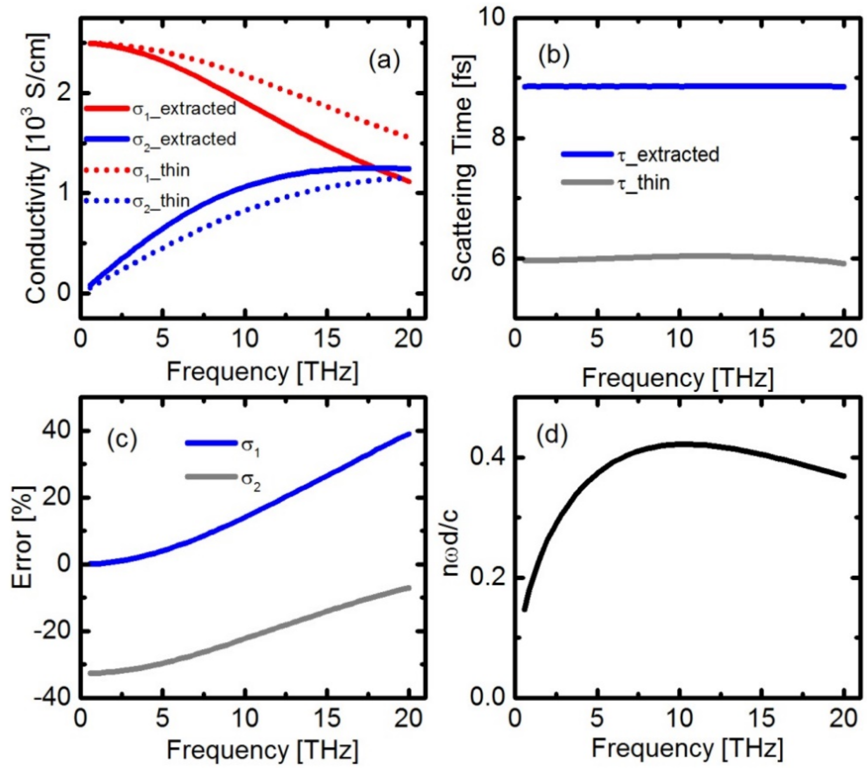

Fig. 3 The results extracted with the algorithm and thin film equations as function of terahertz frequency: (a) The complex conductivity, (b) the scattering times, (c) the error caused by the thin film equations and (d) the thin film criterion.

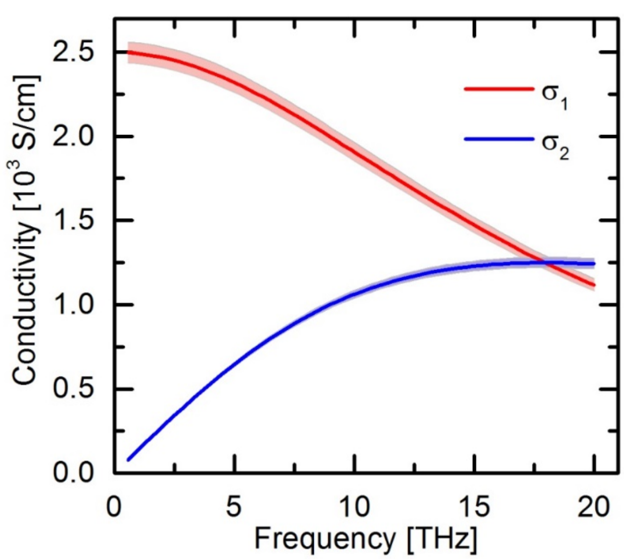

Fig. 4 Conductivities extracted for the thickness of 200 *nm* (line plot), 195 *nm* (upper limit of the filled area) and 205 *nm* (lower limit of the filled area).

The thickness error of the thin film caused by the uniformity of the silicon substrate wafer is also considered. Instead of using 200 *nm* for the extraction process, the thickness of 205 *nm* and 195 *nm* with 5 *nm* error are applied. The extracted values correspond to the lower and upper limit of the filled gray area in Fig. 4. The thickness error of 2.5% contributes to the conductivity error of less than 3%, which illustrates the thickness error will not induce the severe deviation to the extracted conductivity.

4. Experimental results

Transparent conducting oxides (TCOs) have many applications in optoelectronic devices due to their low loss and metallic properties [16-18]. The broadband terahertz conductivities of tin-doped indium oxide (ITO), aluminum or gallium-doped zinc oxide (AZO or GZO) have been discussed in the paper [8]. The extraction algorithm is essential for accurately extracting broadband terahertz conductivities of these TCOs. The terahertz time traces are recorded in the broadband two-color laser induced air plasma system with air biased coherent detection (ABCD) method [19, 20], which extends the useful terahertz frequencies for spectroscopy up to 18 *THz*. The details of the experimental set up have been introduced previously [21, 22]. The structure of the sample is illustrated in Fig. 1. The thin films are deposited on part of high resistivity silicon substrate with thickness of 525 μm for achieving both sample and reference terahertz pulses.

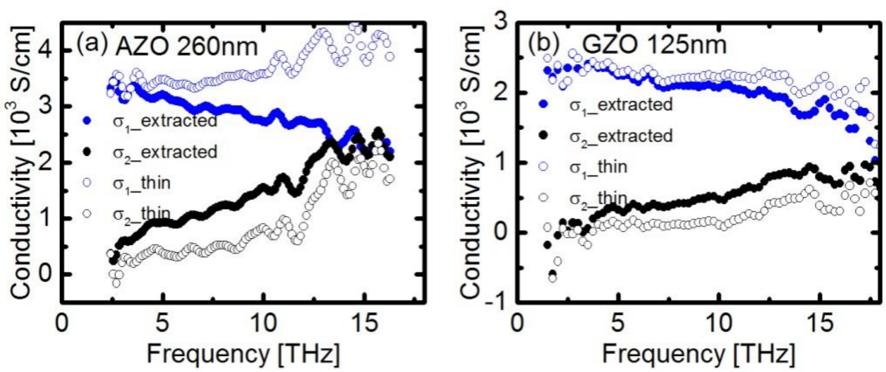

Fig. 5 Frequency dependent complex conductivities of (a) 260 *nm* AZO and (b) 125 *nm* GZO extracted with algorithm and thin film equations.

Figures 5(a) and (b) compare the conductivities of 260 *nm* thick AZO and 125 *nm* thick GZO extracted with the algorithm and thin film equations. Clearly, the results of these two methods differ significantly with each other. The distinctions increase dramatically with increasing frequency, considering the criteria for thin film equation become worse at high frequencies. Even for the 125 *nm* thick GZO, the extracted conductivities match insufficiently at low frequencies. Overall, the mismatch indicates the algorithm is vital for accurate conductivity extraction of thin films in broadband THz-TDS.

5. Conclusions

In conclusion, the conductivity extraction algorithm has been developed and applied for thin films in THz-TDS with no criterion. To verify the accuracy of the extraction method, the FDTD simulation has been performed. Comparing to the thin film equations, the conductivity extracted with the algorithm matches the predefined Drude parameters very well. Furthermore, the importance of using the algorithm for conductivity extraction of thin films has been demonstrated with the TCOs. The extraction method is not limited to thin conducting films deposited on a substrate. It works well for the semi-conducting materials in the broadband time-resolved terahertz spectroscopy (TRTS).

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