Accurate and rapid extraction of optical parameters for thin plates with terahertz time-domain spectroscopy technology

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Abstract: Due to the low THz radiation energy and narrow pulse width (picosecond range), as a new spectral analysis method, terahertz time-domain spectroscopy(THz-TDS) technology is nondestructive and high-temporal-resolution when being used to extract optical parameters of samples. Fabry-Pérot interference is a crucial obstacle in extracting optical parameters of thin wafers in the THz-TDS through-transmission mode. A reliable algorithm was proposed and tested to simultaneously filter Fabry-Pérot interference and obtain precise optical characterization of thin wafers. The algorithm employs a band-stop filter immediately and exclusively designed for every single sample to the initial refractive index and absorption coefficient. Experimental results of doped silicon wafers and amino acid tablets confirming the utility of the algorithm were also presented.

Key words: terahertz; optical parameters; Fabry-Pérot; silicon; amino acid

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太赫兹时域光谱技术用于准确快速地提取薄片的光学参数

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摘 要: 由于太赫兹辐射能量低并且脉冲宽度窄(皮秒量级),太赫兹时域光谱技术作为一种提取材料光学参数的新兴光谱分析手段具有无损伤和高时间分辨率的特点。法布里-珀罗震荡是透射模式太赫兹时域光谱提取薄片的光学参数的主要障碍。为了即时地滤除法布里-珀罗震荡以获得薄片准确的光学参数提出了一种可靠的方法。该方法为每一片测量样品实时地设计其独有的带阻滤波器以滤除叠加在折射率和吸收系数的初始值中的法布里-珀罗震荡。同时不同掺杂的硅片和氨基酸薄片的实验结果证实了该方法的可行性。

关键词: 太赫兹; 光学参数; 法布里-珀罗震荡; 硅; 氨基酸

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0 Introduction

The terahertz(THz) region of the electromagnetic spectrum ranges from tens of gigahertz (GHz) to several THz[1-2]. THz wave technology, especially terahertz time-domain spectroscopy (THz-TDS) has broad application prospects in communication, sensing, and imaging, particularly in security inspection, nondestructive testing, and bio-medical detection, because it is broadband, prompt, spatially and temporally coherent [3-4]. THz-TDS systems can be divided into through-transmission mode, which is used in experiments in this paper, and reflection mode according to whether the incident THz wave travels through the sample or is reflected from the sample surface. THz-TDS systems provide measurements of the full THz electric field over a wide frequency range, rather than just intensity or phase, thus THz-TDS contains abundant spectral information. The vibrational and rotational energy levels of large molecules mostly lie in the THz wave band, while large molecules especially biological and chemical macromolecules whose spectroscopic signatures corresponding to conformational changes and other large scale deformations mostly fall into the THz region[5-6] making THz-TDS a suitable and efficient approach to extract optical parameters of materials to identify their structures and physical properties.

In the past two decades, numerous researchers have realized and used THz-TDS to determine optical characterization for various materials, such as dielectrics[7], semiconductors[8], superconductors[9-10], and biological materials [11]. Duvillaret et al. presented a numerical inversion algorithm which predicated on knowledge of accurate sample thickness to arrive at optical parameters[12]. This method has its limitations with thin samples because sample thickness measurement is not completely accurate and a thin sample acts as a Fabry-Pérot etalon, which highly affects the exactitude of extracted optical parameters such as refractive index and absorption coefficient. To obtain the correct sample thickness together with optical parameters for paint films, Dorney, et al. proposed a method called Total Variation (TV) which performed search for the minimum of the total variation between the measured optical transfer function and a preselected model over a series of thickness[13]. The TV approach can measure the optical parameters of high scattering materials and inhibits noises well. But its calculation speed is quite slow and convergence is often affected by the presence of local minima, nor is it applicable to materials that strongly absorb THz radiation. Pupeza, et al. introduced an update-iterative algorithm that changed the iterative initial value of extinction coefficient in the TV approach of Dorney, et al. This algorithm not only improved accuracy but also reliably measured a sample of 100 μm thick with ultralow refractive index[14]. But the update method solves neither the low computing speed nor the local convergence. Scheller, et al. provided a simple approach in a quasi space which performed another Fourier transform over computed material parameters to filter out periodic Fabry-Pérot oscillations[15]. This method is especially suitable for measurements in which the Fabry-Pérot effect is significant, but is susceptible to isolated absorption features in the optical parameters of materials and noises of the system, which has negative impact on the accuracy of the sample thickness and optical parameters.

The present work concerns itself with a practical method, with consideration for filtering out Fabry-Pérot interference, to accurately and simultaneously determinate optical characterization of materials. As a demonstration, we considered a full family of silicon wafers and amino acid tablets obtaining their full suites of optical parameters in the THz region. The silicon wafers have systematic variation of wafer thickness and doping densities of both n-type and p-type dopants, ranging from lightly doped to heavily doped samples. The amino acid samples include three kinds of amino acids, D-alanine(D-Ala), DL-alanine

(DL-Ala) and L-tyrosine (L-Tyr). Beyond the mere goal of rapid and accurate extraction of optical parameters of thin samples, the veracity and rapidity of the proposed approach make it highly suitable for spectral imaging of materials of technological importance.

1 Algorithm

When extracting optical parameters of thin samples in the through-transmission mode, the crucial issue we are confronted with is how to remove the omnipresent Fabry-Pérot interference. The potential of a temporal window to remove the Fabry-Pérot effect emerges in cases in which multiple echoes are far separated from the first directly transmitted terahertz pulse. But the use of a temporal window leads to questionable validity of the measured signal in time-domain at the edges of the window[14]. Moreover, in some cases the Fabry-Pérot oscillatory interference that is superimposed on valuable information we need overlaps with each other or even is not evident in time domain so cannot be filtered by a temporal window. Thus, instead of the window-based method, we chose to employ a novel but easy-to-implement method to remove the Fabry-Pérot interference in the present work. Such a procedure contains the following steps: At the outset, with the THz radiation perpendicularly incident, we calculate the initial values of the refractive index and the absorption coefficient, using the following equations (derived from the Fresnel′s Law of refraction/reflection, together with the Beer-Lambert Law of attenuation. Details of the derivation can be found in Ref[12].

$$n_s(\omega)=\frac{\varphi(\omega)c}{\omega d}+1 \tag{1}$$

$$\alpha(\omega)=\frac{2\kappa(\omega)\omega}{c}=\frac{2}{d}\ln\frac{4n_s(\omega)}{A(\omega)(n_s(\omega)+1)^2} \tag{2}$$

Here n_s is the real part of the refractive index, φ is the phase difference between the Fourier transform of sample signal recorded with the measured sample and reference signal recorded in air, c is the THz wave propagation speed in vacuum, ω is the angular frequency, d is the sample thickness, α is the absorption coefficient proportional to the extinction coefficient κ, A is the ratio of amplitudes of the sample signal and the reference in the frequency domain.

In the second step, an exclusive and instantaneous band-stop filter $H(f)$ is developed for each measured sample. As can be observed in Fig.1(dotted line), the

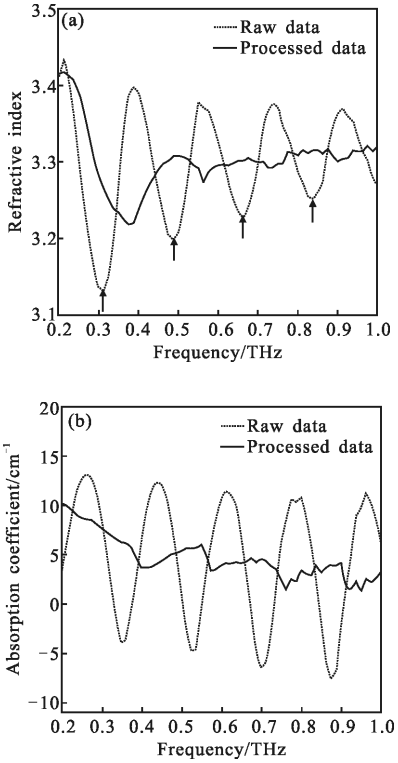

Fig.1 Calculated optical parameters using Eqs.(1)-(2): (a) refractive index and (b) absorption coefficient

periodic Fabry-Pérot oscillation, which we need to remove, is superimposed on the calculated refractive index and absorption coefficient using Eqs. (1)-(2), from which the Fabry-Pérot resonance frequency can be obtained. The procedure of constructing the filter $H(f)$ is composed of the following three steps:

(1) Find all the trough positions indicated by the arrows in the refractive index in Fig.1(a) $f=\{f_1, f_2, \cdots, f_n\}$ (the corresponding crests have the same effect). The frequency difference between every two adjacent troughs is the Fabry-Pérot resonance frequency, namely,

$$f_{\mathrm{fp}\_n}=\Delta f=f_n-f_{n-1} \tag{3}$$

The troughs and crests in the absorption coefficient in Fig.1 (b) can play the same roles as those in the refractive index.

(2) The actual Fabry-Pérot oscillation period in the frequency domain is taken as the average of all the Fabry-Pérot resonance frequencies calculated in step (1) by Eq.(3) to minimize the error of individual Fabry-Pérot resonance frequency obtained from trough positions that have uncertainties of the order of the spectral resolution of the THz-TDS system employed (12.5 GHz in the present case).

$$f_{\mathrm{fp}}=\frac{1}{n}\sum_{k=1}^{n}f_{\mathrm{fp}\_k} \tag{4}$$

The reciprocal of Fabry-Pérot oscillation period in the frequency domain mentioned above is the Fabry-Pérot oscillation frequency in a quasi space domain, which will be the center frequency of the designed band-stop filter.

$$f_{\mathrm{qs}}=\frac{1}{f_{\mathrm{fp}}} \tag{5}$$

(3) The designed band-stop filter can be described as follows:

$$H(f)=\begin{cases}0 & \text{if } f_{\mathrm{qs}}-BW/2<f<f_{\mathrm{qs}}+BW/2\\ 1 & \text{otherwise}\end{cases} \tag{6}$$

Where BW is the rejection bandwidth of the filter; the most effective value of BW is as follows:

$$BW=\frac{2f_0^2}{f_{\mathrm{fp}}(f_{\mathrm{fp}}-f_0)(f_{\mathrm{fp}}+f_0)} \tag{7}$$

Here f_0 is the spectral resolution of THz-TDS system and at same time is the determined resolution of f_{fp}. The rejection bandwidth of the filter defined above contains the inaccuracy range to avoid the misdiagnosis of f_{fp}.

Lastly, we apply the designed filter to the initial values of the refractive index and the absorption coefficient abtained in step (1). To eliminate the influence of the approximation in the derivation of Eqs.(1)-(2), the errors of the ratio of amplitudes and phase difference between the sample signal and the reference are employed to modify the initial value of the refractive index and absorption coefficient.

$$\delta_A=\ln(A(\omega))-\ln(A_m(\omega)) \tag{8}$$

$$\delta_\varphi=\varphi(\omega)-\varphi_m(\omega) \tag{9}$$

Where δ_A and δ_φ are respectively the errors of the amplitudes ratio and phase difference between the sample signal and the reference, $A(\omega)$ and $A_m(\omega)$ are the theoretical and measured value of the amplitude ratio, and $\varphi(\omega)$ and $\varphi_m(\omega)$ are the theoretical and measured value of the phase difference. The error transfer functions are included to calculate the uncertainty caused by such errors. The modification procedure is to go on iteratively until δ_A and δ_φ are less than some predefined fraction 1% of the refractive index and absorption coefficient ratios.

$$\Delta\delta_A=\frac{\Delta A_m}{A_m}+|\kappa-\kappa_a|\frac{\omega}{c}\Delta d \tag{10}$$

$$\Delta\delta_\varphi=\Delta\varphi_m+|n-n_a|\frac{\omega}{c}\Delta d \tag{11}$$

Here ΔA_m, $\Delta\varphi_m$ and Δd are respectively the resolution of measured amplitude, phase and thickness, and κ_a and n_a are the imaginary and real part of the complex refractive index. Figure 1 shows the raw data and the processed data of Sample 1, the characterization information of which can be found in Tab.1, after being filtered by the aforementioned filter: (a) is the

Tab.1 Characterization information of silicon wafers

Label	Resistivity /Ω·cm	Doping type	Dopant	Doping concentration (10^{11} cm^{-3})*	Thickness /μm
Sample 1	1-10	n-type	Phosphorus	13 400-146 000	247.4
Sample 2	1-5	n-type	Phosphorus	9 470-146 000	445.8
Sample 3	25-35	n-type	Phosphorus	569-718	446.6
Sample 4	>20 000	n-type	Phosphorus	<6.6	516.1
Sample 5	0.5-1	p-type	Boron	146 000-321 000	521.9
Sample 6	10-20	p-type	Boron	6 670-13 310	516.2
Sample 7	>2 500	p-type	Boron	<53.2	314.9

* Doping concentration was looked up from Ref [17], based on the manufacture supplied resistivity data

refractive index and (b) is the absorption coefficient. From Fig.1, we can see that the Fabry-Pérot oscillation has been neatly eliminated from the optical parameters.

2 THz-TDS system and samples

The robust THz-TDS system deployed in our research is the T-Gauge 5000 unit from Advanced Photonix, Inc. [16]. The femtosecond laser pulse produced from the Sapphire oscillator has a duration of 80 fs, a central wavelength of 1 064 nm, a repetition rate of 100 MHz, and an average power of probe beam 20 mW. The system can work in both the reflection and the through-transmission modes, with the working mode being determined by the fashion in which the transmitter and receiver are installed on a travelling rail. Regardless of the modality of operation, the system provides a spectral bandwidth ranging from about a few tens of GHz to 3.5 THz, a signal to noise ratio better than 70 dB at the low-frequency end (down to a few tens of GHz), and not less than 35 dB at the high-frequency end. The sampling interval of the TDS is 0.1 ps, and the spectral resolution is 12.5 GHz.

The doped silicon samples under investigation in our experiment were purchased from Harbin Turbo Technology Co. Ltd. They included two types: n-Si (phosphorus-doped) and p-Si (boron-doped). The thinnest Si wafer was 247.4 μm phosphorus-doped, labeled as Sample 1, with nominal resistivity of 1-10 Ω·cm. The thickest sample was 521.9 μm, boron-doped, labeled as sample 5, whose characterization information together with that of the other doped silicon wafers is shown in Tab.1. The designated thickness of the samples represents the average of 10 measurements, by a micrometer with a 1μm resolution.

During the preparation of the tablets of D-Ala, DL-Ala and L-Tyr, 20 mg of the three kinds of amino acids are respectively mixed thoroughly with 20 mg polyethylene(PE) powder and then pressed (at 4 tons cm^{-2} for 1 minute) into tablets labeled as Sample 8, Sample 9, Sample 10. The diameter of pressed pellets was 15 mm which matches the size of the terahertz light spot and the thicknesses are all 300 μm.

3 Experimental results

Figure 2 shows the refractive index (a) and the absorption coefficient (b), processed by the band-stop filter described in Section 3 for three phosphorus-doped Si samples: Sample 2, Sample 3 and Sample 4. Solid lines are processed parameters and dotted lines are data obtained directly from Eqs.(1)-(2). Obviously

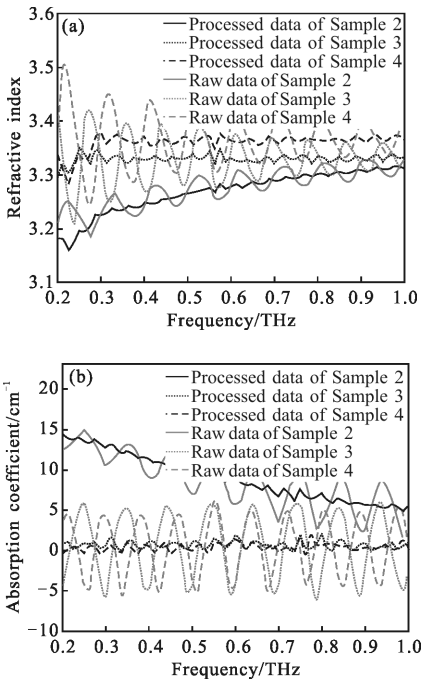

Fig.2 Processed data and raw data: (a) refractive index and (b) absorption coefficient of Sample 2, Sample 3, and Sample 4

the Fabry-Pérot interference superposed on optical parameters is neatly removed and the processing time is several minutes even tens of seconds. In the term of calculating speed our method is prior to TV approach. Over the frequency range of 0.2-1 THz the refractive index and the absorption coefficient curves of these three samples are rather featureless. At each frequency, the value of refractive index of these three samples follows the order of Sample 4> Sample 3> Sample 2, while the absorption coefficient is in the reverse order. Because silicon wafers with higher doping concentrations have lower resistivity, resulting

in stronger absorption and a lower index of refraction. The refractive index and the absorption coefficient obtained here are in agreement with those given in the literature[18].

These observations are also true for boron-doped p −type silicon samples. Processed data for the refractive index(a) and the absorption coefficient (b) of Sample 5, Sample 6 and Sample 7 are shown in Fig.3.

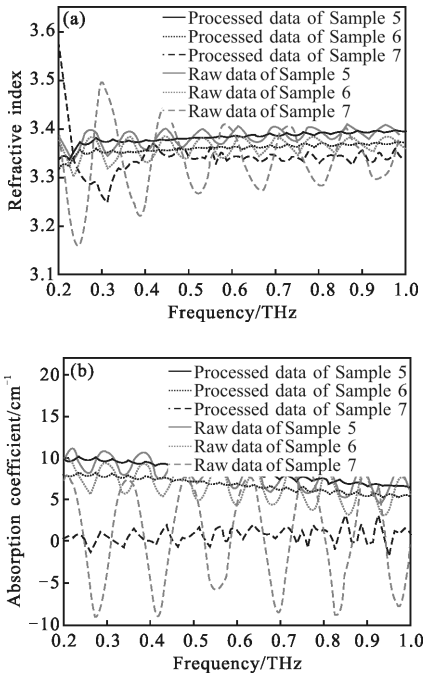

Fig.3 Processed data and raw data: (a) the refractive index and (b) the absorption coefficient of Sample 5, Sample 6 and Sample 7

The raw and processed refractive index and absorption coefficient of Sample 8, Sample 9 and Sample 10 are respectively shown in Fig.4 (a) and Fig. 4 (b). The dotted lines are the raw parameters obtained directly from Eqs.(1)−(2) and the solid lines are the processed parameters handled by the band-stop filter described in Section 3 for every single sample. In the curves of raw data of both the refractive index and the absorption coefficient the Fabry-Pérot interference superposed on optical parameters among the frequency band lower than 0.78 THz. Additionally an absorption peak is observed at 1.24 THz in the curves of raw data of the absorption coefficient for DL−Ala and another absorption peak is observed at 0.962 5 THz in the curves of raw data of the absorption coefficient for L −Tyr. At the same frequencies with those of the two absorption peaks which are in accordance with literature data[19−20] in the absorption coefficient locate, the curves of the refractive index show the corresponding peaks. Through the filtering by the instantly designed band-stop filter the Fabry-Pérot oscillation at the low frequency band is thoroughly filtered meanwhile the peaks mentioned above are perfectly preserved. So our method is more effective in dealing with Fabry-Pérot oscillation interfering optical parameters with absorption peaks comparing with QS method.

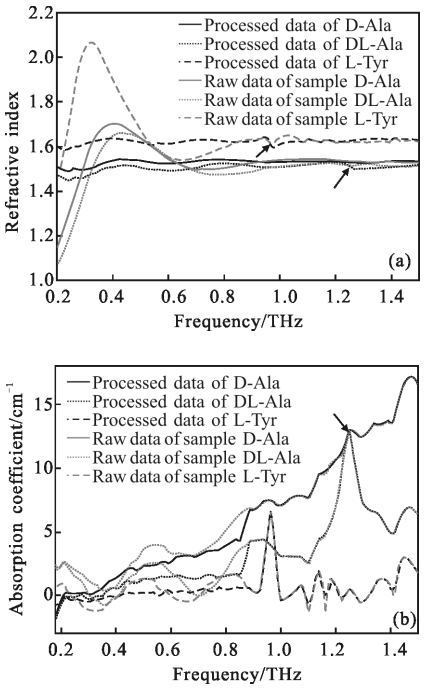

Fig.4 Raw and processed data: (a) the refractive index and (b) the absorption coefficient of D−Ala, DL−Ala and L−Tyr

4 Conclusions

A simple and reliable method is developed for ridding of the Fabry-Pérot interference to precisely and simultaneously determinate material optical characterizations for homogeneous thin, planar solid samples. For each measured sample, an exclusive band-pass filter is promptly and exclusively designed to remove the Fabry-Pérot multi-reflections

superimposed on the optical parameters. Because of its rapidity and precision the proposed approach suits well for spectral imaging applications. The systematic experiments on a full suite of silicon wafers and three kinds of amino acids proved the feasibility of our method. Under the premise of thoroughly filtering the Fabry-Pérot and ensuring accuracy, comparing with QS method our method is more adaptive to remove Fabry-Pérot oscillation superposed on optical parameters with absorption peaks and considering the calculating speed our method is prior to TV approach.

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