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# Single trace terahertz spectroscopic ellipsometry

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**Abstract:** A new technique for terahertz time-domain ellipsometry is presented. Information of reflection coefficients of the sample in two orthogonal polarizations is encoded on the same terahertz trace by using a birefringent medium. This allows for single measurement refractive index extraction without the need for a moving analyzer. A comparison of the complex refractive index measurements of optical grade fused silica and non birefringent sapphire are carried out both in reflection ellipsometry and with a standard terahertz transmission spectrometer showing good agreement.



## 1. Introduction

Terahertz-time domain spectroscopy (THz-TDS) is becoming a widespread technique to measure optical properties in many kinds of scenarios ranging from research laboratories to industrial environments [1–3], being the transmission configuration the most common due to its high accuracy and reliability. However, in certain applications a reflection setup is more convenient due to either elevated absorption of the material or difficulty in accessing the back of the sample. In these cases, typical sample-reference measurements suffer from phase errors due to misalignments between sample and reference mirror which are difficult to overcome [4,5], this is why self-referencing methods such as [6] or ellipsometry, are best suited.

In THz reflection ellipsometry, information from the sample is obtained by measuring the reflection coefficient in two polarization directions. Conventional ellipsometry is performed in a two-step configuration, in which the angle of one of the polarizers is mechanically changed to select the desired polarization (parallel or perpendicular to the incidence plane) [7–9]. One method to speed up the measurement is fast mechanical polarization modulation [10,11]. Also, instruments with simultaneous polarization measurement have been proposed based on balanced electrooptical detection of orthogonal polarizations [12,13]. Here, a new method that eliminates the need to turn the polarizers based on a birefringent medium is proposed. Mechanical elements are avoided just by adding a single passive component. The method simplifies measurements since, after initial system calibration, all later measurements are self-referenced and all the information needed for the extraction of the optical properties is recorded in one single terahertz trace.

## 2. Method

The technique described in this work employs the scheme outlined in Fig. 1. The polarization of the emitter and the first polarizer (P1) are set to the same angle $\alpha$. This is needed to ensure that no cross-polarization coming from the antenna goes though the setup.

The signal after P1 passes through element B which is a flat window of birefringent material with its fast axis matching the direction parallel to the plane of incidence. After B, each polarization will acquire a different time delay due to the refractive index of each axis, as well as a different attenuation. By introducing this element, there is no need for an analyzer polarizer since

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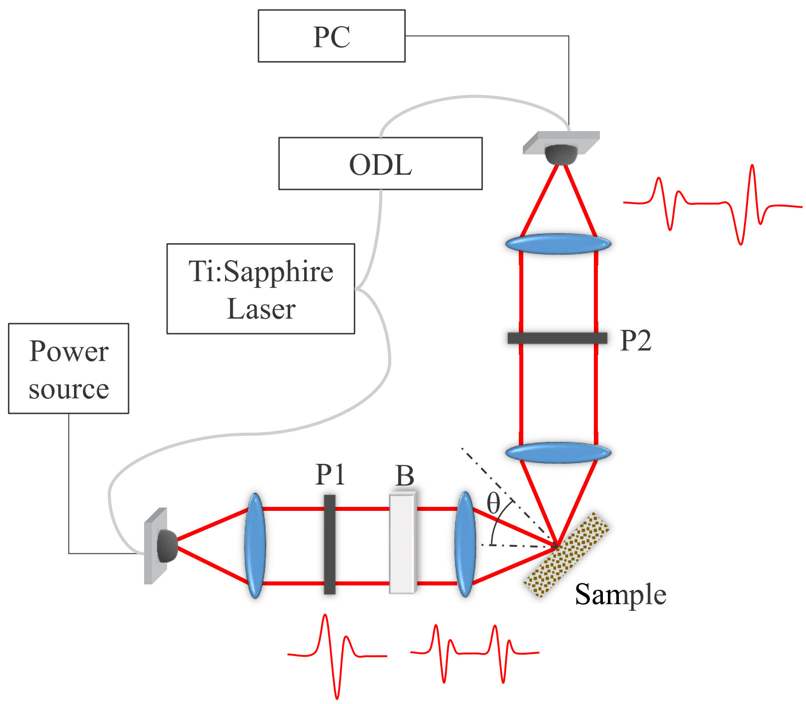


**Fig. 1.** Schematics of the THz ellipsometry setup. P1 and P2 are THz wire grid polarizers. B is a birefringent c-cut sapphire plate. The red pulse shapes show the evolution of the temporal waveform of the THz signal along the path of the ellipsometer.

*p*- and *s*- polarizations are already separated in time. Due to the pulsed nature of time-domain terahertz systems, information is stored in a very short span of time and interference between polarizations is easily avoidable, which allows for typical ellipsometric data extraction [7] after processing of the temporal trace.

Radiation reflected on the sample will then change its amplitude and phase according to Fresnel reflection coefficients. The Jones vector of this THz pulse takes the following shape:

$$|P_{sample}\rangle = \frac{r_p}{\sqrt{2}} t_{pb} \cos\alpha |p\rangle + \frac{r_s}{\sqrt{2}} t_{sb} \sin\alpha exp(i\delta)|s\rangle, \tag{1}$$

where $\delta = f(n_0 - n_e)d/c$ is the phase difference introduced by the birefringent material that depends on the complex ordinary and extraordinary indexes ($n_o$, $n_e$), the thickness $d$ of the material, the frequency $f$ of the incoming wave and $c$, the speed of light in vacuum. The transmission coefficients of the birefringent window are denoted by $t_{pb}$ and $t_{sb}$; $r_p$ and $r_s$ are the reflection coefficients of the sample.

Finally, the THz signal arrives at another polarizer oriented at 45° and the time trace is recorded by a receiver whose polarization axis is also oriented at the same angle. This setup ensures a balanced measurement of both polarizations and the detected signal is proportional to the projection of the amplitude of the incoming wave in the axis of the polarizer:

$$\langle 45^\circ | P_{sample}\rangle = \frac{r_p}{2} t_{pb} \cos\alpha + \frac{r_s}{2} t_{sb} \sin\alpha exp(i\delta). \tag{2}$$

Each term in Eq. (2) corresponds to the complex amplitude of the pulses in Fig. 2. If the time delay between the two contributions $\Delta t = (n_o - n_e)d/c$ is greater than the pulse duration, it will be possible to separate the pulses corresponding to the two polarizations by applying a window function to each one.

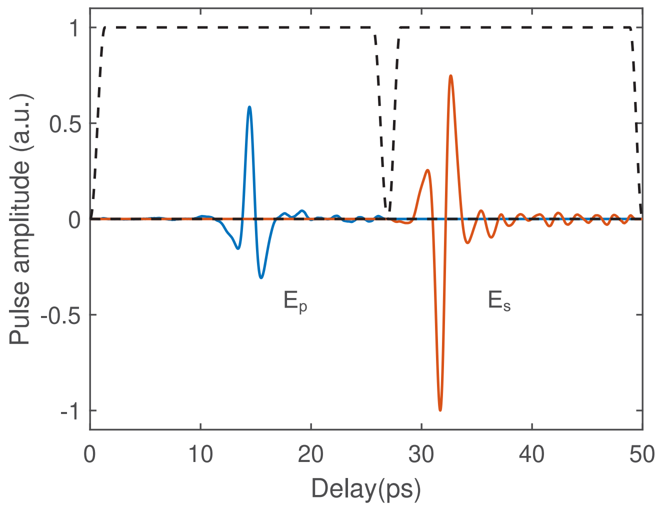


**Fig. 2.** In solid line, raw recording of the pulses after an ellipsometric measurement. In dashed line, windows used to separate orthogonal polarizations.

Separation of the pulses is best achieved by using a window function that distorts the pulses as little as possible. This is achieved with two cosine-tapered rectangular windows that meet at a certain delay position in between the pulses. This middle position is calculated for each trace following the criteria that the extracted optical parameters using a given pair of windows show the least amount of ripple. For this, a minimization of the Total Variation of the refractive index is performed in the frequency range with maximal signal to noise ratio. [14].

After windowing, the ellipsometric quantity $\rho_{meas}$ is computed by dividing the Fourier transform of each pulse. This quotient is represented by the ratio of the terms in Eq. (2):

$$\rho_{meas} = \frac{E_p}{E_s} = \frac{t_{pb}}{t_{sb}} \frac{r_p}{r_s} \frac{\cos\alpha}{\sin\alpha} e^{i\delta}, \tag{3}$$

In order to obtain the ellipsometric parameter of the sample $\rho = r_p/r_s$, two additional measurements, are needed to substract the effect of the birefringent window. This one-time calibration is performed by measuring a mirror instead of the sample material. In each of the measurements the birefringent window is turned to align its fast and slow axis respectively with the polarization angle of P1. This results in the acquisition of two traces that only show one pulse and carry the information of the transmission coefficients and relative time delay of the birefringent window axis:

$$\begin{aligned} P_{cals} &= \frac{t_s}{\sqrt{2}}(\cos\alpha|p\rangle + \sin\alpha|s\rangle)e^{i\delta} \\ P_{calp} &= \frac{t_p}{\sqrt{2}}(\sin\alpha|p\rangle + \sin\alpha|s\rangle) \end{aligned} \tag{4}$$

Then, the calibration ratio is obtained as follows:

$$\rho_{cal} = \frac{P_{calp}}{P_{cals}} = \frac{t_p}{t_s} e^{-i\delta}. \tag{5}$$

Using Eqs. (4) and (5) the ellipsometric parameter can be found:

$$\rho = \frac{r_p}{r_s} = \frac{\rho_{meas}}{\rho_{cal}} \tan\alpha \tag{6}$$

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To obtain the complex refractive index $N = n - i\kappa$ of the sample, the exact analytical solution of the dielectric permittivity $\varepsilon$ in terms of $\rho$ was used [15] and then, its square root was taken.

$$N = \sqrt{\varepsilon} = \sin\theta\sqrt{1 + \left(\frac{1-\rho}{1+\rho}\right)^2 \tan^2\theta}. \quad (7)$$

In ellipsometric measurements made in the visible spectrum, the effect of the uneven surface of the sample that may contain thin films of other materials has to be taken into account. Drude proposed a correction for the phase of the $\rho$ parameter [15], however, this correction depends inversely on the wavelength of the incident light that is, typically, around 1 $\mu$m. Since THz ellipsometry employs light whose wavelength is 300 times greater than that of optical ellipsometry, it can be assumed that Eq. (7) provides a good approximation.

## 3. Results and discussion

Measurements have been carried out using the setup shown in Fig. 1. It is based on an all-fiber implementation using pigtailed photoconductive antennas and a femtosecond pulse laser at 1550 nm with a repetition rate of 100 MHz. A set of lenses made of TPX polymer, with 35.5 mm clear aperture and a focal length of 50 mm are used for collimation and focusing of the radiation. This system provides an effective bandwidth of 1.5 THz and a maximum dynamic range of 60 dB after averaging 200 traces. The polarizers used in this setup have been characterized by measuring the ratio of the amplitudes transmitted at the passing and blocking positions. Result in Fig. 3 shows a polarization extinction ratio better than 25 dB from 0.2 to 1 THz.

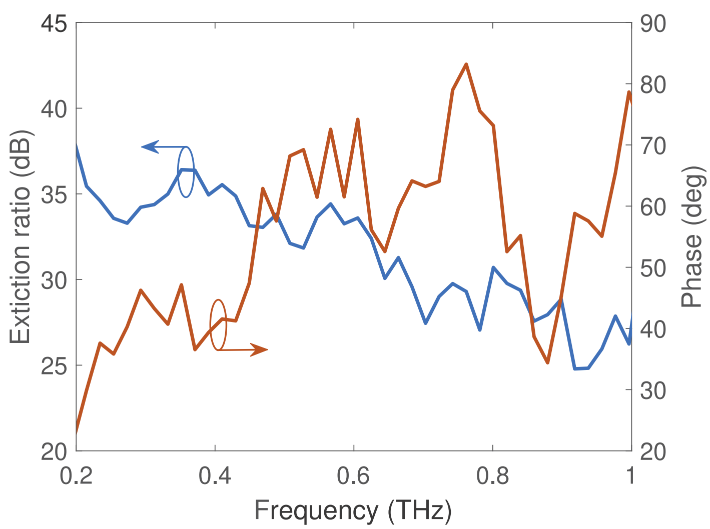


**Fig. 3.** Frequency characteristics of the employed wire grid polarizers.

Recording of the THz signal is achieved by sampling the electric current generated in the receiver when a delayed copy of the laser pulse illuminating the emitter arrives at the antenna. A voice-coil driven optical delay line (ODL) controls the delay of the sampling pulse and allows mapping the full shape of the signal in the time domain at a speed of 10 traces per second.

The birefringent material used was a 15 mm thick sapphire plate cut with the c-axis parallel to its faces. Sapphire is a convenient material since shows a birefringency of around 0.32 and an absorption of 1 $cm^{-1}$ at 1THz [16,17]. Measurements have shown that the Sapphire plate introduces a delay of 17.2 ps. The thicker the window, the better to separate polarization-dependent traces.

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To prove the feasibility of this technique, measurements where carried out using samples of two materials: 5mm thick UV grade fused silica and 5 mm thick optical grade sapphire cut along the ordinary axis, so there is no birefringence in the propagation direction. Both materials where also measured in transmission configuration to obtain a reference value of their refractive index.

Figure 2 shows the shape of the temporal trace for the silica measurement. After windowing and performing the Fourier transform of the pulses, the $\rho$ parameter is found using Eq. (6). Then, Eq. (7) is used to obtain the refractive index of the sample.

For the results shown in Fig. 4, 5 sets of 200 traces per material were recorded with an angle of incidence $\theta$ close to the Brewster angle. This is done so that the relative amplitude of *p*- and *s*-components is most different and thus, sensitivity in the value of the ellipsometric parameter is improved. The angular position of the first polarizer P1 and the emitter antenna is set to $\alpha = 18°$ following [18] in order to minimize the error sensitivity on the angular position of B an P1.

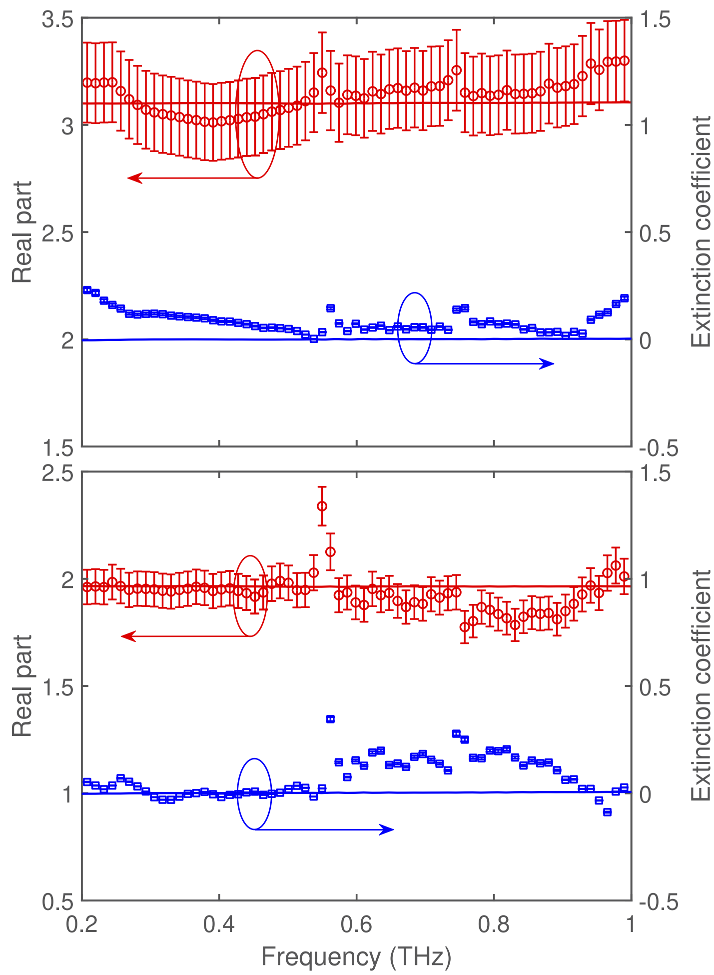


**Fig. 4.** Real (red) and imaginary (blue) parts of the refractive index of fused silica measured at an incident angle of 50° (a) and ordinary axis sapphire measured at an incidence angle of 70° (b) obtained with the proposed ellipsometric technique. Continuous lines correspond to transmission measurements. Smoothing is carried out by zero padding the temporal trace.

Refractive index calculations were performed for each set and then their average value was taken. The results are shown in Fig. 4. The error bars come from the error propagation of the

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variables with the most uncertainty, which are the incidence angle and the angular orientation of the birefringent plate. Refractive indexes obtained in transmission mode and using time delayed ellipsometry agree with the typical margin in conventional THz ellipsometry [18,19].

The delay introduced by the birefringent medium limits the trace length for each pulse, which bounds the frequency resolution of this approach. For that reason, this technique might be better suited for measuring materials with flat or slowly changing refractive index. The maximum trace length is in the order of the delay introduced by the sapphire window which, for the setup used in the experiments, allows for a frequency resolution of approximately 60 GHz.

In addition, some degree of interference between the two pulses in the trace can appear due to the presence of water vapor in the optical path of the ellipsometer. Water vapor effect in THz pulses is seen as an oscillating tail that accounts for the absorption of the rotational transitions of the $H_2O$ molecule [20]. Due to the proximity of the two pulses, the tail of the first one can affect the recorded amplitude of the second one changing this way the obtained refractive index. In the frequency domain, water vapour creates absorption peaks that distort the shape of the spectra and are prone to cause artifacts in the optical parameters like the ones observed in Fig. 4 at around 0.55 THz. In order to avoid this pulse interference, care has been taken to align the birefringent window axis in such a way that the smaller *p*- component traverses the fast axis, gets recorded first in time and therefore, a lower amount of tail distortion is applied to the slower *s*- pulse.

Another factor to be taken into account in the system is the correct alignment of the optical axis of the birefringent plate with the axis of the polarizers. This was done by placing and then rotating the sapphire window between two polarizers set to 0° and 90° until the THz signal at the receiver was minimal. At any other angle not matching the optical axis, the incoming polarization would split in two mutually delayed pulses with polarizations not crossed to the second polarizer and thus, some signal would be detected. Mechanical components in this setup allowed an accuracy in the optical axis of 1° following this method. In order to measure the effect of a possible misalignment, ellipsometric measurements of a HRFZ silicon sample where conducted with the birefringent window in different angular positions deviating from the correct one. Then, an average refractive index in the range of 0.2 to 1 THz was obtained and its value for

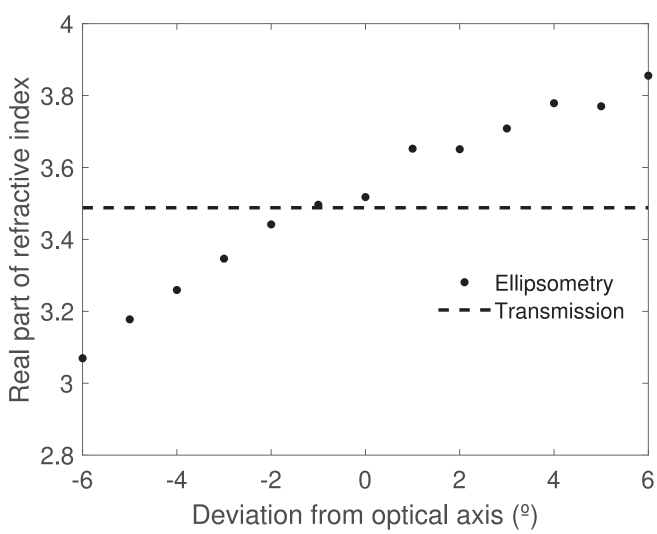


**Fig. 5.** Real part of the refractive index of HRFZ silicon calculated for different angular positions of the birefringent plate averaged from 0.2 to 1 THz. Dashed line corresponds to the refractive index measured in transmission.

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different angles can be seen in Fig. 5. There, it is shown that deviation due to a 1° misalignment of the window produces an error in the refractive index of 2%.

## 4. Conclusion

It has been shown that moving elements in terahertz ellipsometry can be easily removed only by adding a passive element, i.e. a birefringent crystal. Experimental results on fused silica and sapphire show good agreement with transmission spectroscopy. Applications such as industrial quality control and in-line monitoring, where simple instruments are a need, will benefit from this approach.

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## References

1. Y. S. Lee, *Principles of Terahertz Science and Technology* (Springer US, 2009).
2. S. Dexheimer, ed., *Terahertz Spectroscopy: Principles and Applications* (CRC Press, 2008).
3. A. Bockelt, J. Palací López, and B. Vidal, "All-fiber centralized architecture for parallel terahertz sensors," IEEE Trans. Terahertz Sci. Technol. **5**(1), 137–144 (2015).
4. M. Khazan, R. Meissner, and I. Wilke, "Convertible transmission-reflection time-domain terahertz spectrometer," Rev. Sci. Instrum. **72**(8), 3427–3430 (2001).
5. H. B. Liu, Y. Chen, G. J. Bastiaans, and X. Zhang, "Detection and identification of explosive rdx by thz diffuse reflection spectroscopy," Opt. Express **14**(1), 415–423 (2006).
6. F. Sanjuan, A. Bockelt, and B. Vidal, "Birefringence measurement in the terahertz range based on double fourier analysis," Opt. Lett. **39**(4), 809–812 (2014).
7. T. Nagashima and M. Hangyo, "Measurement of complex optical constants of a highly doped si wafer using terahertz ellipsometry," Appl. Phys. Lett. **79**(24), 3917–3919 (2001).
8. N. Matsumoto, T. Hosokura, T. Nagashima, and M. Hangyo, "Measurement of the dielectric constant of thin films by terahertz time-domain spectroscopic ellipsometry," Opt. Lett. **36**(2), 265–267 (2011).
9. A. A. Galuza, V. K. Kiseliov, I. V. Kolenov, A. I. Belyaeva, and Y. M. Kuleshov, "Developments in THz-range ellipsometry: Quasi-optical ellipsometer," IEEE Trans. Terahertz Sci. Technol. **6**(2), 183–190 (2016).
10. C. M. Morris, R. V. Aguilar, A. V. Stier, and N. P. Armitage, "Polarization modulation time-domain terahertz polarimetry," Opt. Express **20**(11), 12303–12317 (2012).
11. T. Iwata, H. Uemura, Y. Mizutani, and T. Yasui, "Double-modulation reflection-type terahertz ellipsometer for measuring the thickness of a thin paint coating," Opt. Express **22**(17), 20595–20606 (2014).
12. M. B. Byrne, M. U. Shaukat, J. E. Cunningham, E. H. Linfield, and A. G. Davies, "Simultaneous measurement of orthogonal components of polarization in a free-space propagating terahertz signal using electro-optic detection," Appl. Phys. Lett. **98**(15), 151104 (2011).
13. Q. Guo, Y. Zhang, Z. Lyu, D. Zhang, Y. Huang, C. Meng, Z. Zhao, and J. Yuan, "Thz time-domain spectroscopic ellipsometry with simultaneous measurements of orthogonal polarizations," IEEE Trans. Terahertz Sci. Technol. **9**(4), 422–429 (2019).
14. I. Pupeza, R. Wilk, and M. Koch, "Highly accurate optical material parameter determination with thz time-domain spectroscopy," Opt. Express **15**(7), 4335–4350 (2007).
15. H. G. Tompkins and E. A. Irene, eds., *Handbook of Ellipsometry* (William Andrew, 2005).
16. D. Grischkowsky, S. Keiding, M. van Exter, and C. Fattinger, "Far-infrared time-domain spectroscopy with terahertz beams of dielectrics and semiconductors," J. Opt. Soc. Am. B **7**(10), 2006–2015 (1990).
17. Y. Kim, M. Yi, B. G. Kim, and J. Ahn, "Investigation of thz birefringence measurement and calculation in Al2O3 and LiNbO3," Appl. Opt. **50**(18), 2906–2910 (2011).
18. X. Chen, E. P. J. Parrott, Z. Huang, H.-P. Chan, and E. Pickwell-MacPherson, "Robust and accurate terahertz time-domain spectroscopic ellipsometry," Photonics Res. **6**(8), 768–775 (2018).
19. M. Neshat and N. P. Armitage, "Terahertz time-domain spectroscopic ellipsometry: instrumentation and calibration," Opt. Express **20**(27), 29063–29075 (2012).
20. M. van Exter, C. Fattinger, and D. Grischkowsky, "Terahertz time-domain spectroscopy of water vapor," Opt. Lett. **14**(20), 1128–1130 (1989).