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Polarization state and Mueller matrix measurements in terahertz-time domain spectroscopy

Hui Dong [a,*], Yandong Gong [a], Varghese Paulose [a], Minghui Hong [b]

[a] *Institute for Infocomm Research, 1 Fusionopolis Way, #21-01 Connexis, 138632, Singapore*
[b] *Data Storage Institute, 5 Engineering Drive 1, 117608, Singapore*

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ABSTRACT

We derive the complete formulae governing the polarization state measurement in terahertz-time domain spectroscopy (THz-TDS) by using a rotatable THz polarizer. Four Stokes parameters can be uniquely obtained by spectrally-resolved measurement in THz-TDS. Further, we propose a new approach to measure the Mueller matrix of a pure birefringent material, using THz-TDS, by rotating the material under test. Based on the above techniques, we successfully measured the Mueller matrices of a quartz crystal in the frequency domain.

1. Introduction

Terahertz-time domain spectroscopy (THz-TDS) technology has been successfully used to measure refractive index and attenuation coefficient of a material in the THz range using single-cycle pulses [1,2]. The fingerprints obtained, based on the two parameters, have been used to distinguish and characterize different materials. In addition to refractive index and attenuation, some materials have polarization properties [3–5]. Therefore, THz-TDS technology has been extended to realize spectrally-resolved polarization measurement [6–12]. To detect the polarization properties of a material, the polarization state of THz wave should be measured in THz-TDS. So far, two kinds of polarization state measurement technologies have been proposed in THz-TDS. The first one uses a multi-contact (three or four) THz detector and two separate lock-in amplifiers to measure the orthogonal components of THz electric field simultaneously [6–10]. This approach can accomplish the measurement using only one time-scanning process. However, its performance is limited by inhomogeneity of probe beam intensity, inhomogeneity of the electrical property of the photoconductive substrate, imperfect shape of the contacts and gap, misalignment of the antenna orientation and inconsistent responses of two separate lock-in amplifiers [9]. The second approach uses the rotatable THz waveplates or THz polarizers in front of a two-contact THz detector. In optics, generally, a quarter waveplate and a linear polarizer are used for the polarization state measurement [13]. This configuration can certainly be adopted in the THz polarization state measurement. To achieve this goal, Masson and Gallot proposed the use of a combination of several quartz plates to flatten the phase retardance in a wider frequency range as the frequency range of THz wave in THz-TDS is up to several THz and the phase retardance of a quartz plate is very sensitive to frequency [14]. So far, the working frequency range of this THz quartz quarter waveplate is still less than that of a wire-grid THz polarizer. On the other hand, Masson and Gallot also presented the use of two polarizers to measure the polarization states because the THz wire-grid polarizers are almost frequency-insensitive [14,15]. However in Masson's method, only $\cos\delta$ (δ is the phase difference between two polarization components) is explicitly expressed. But δ cannot be uniquely determined. For example, if $\delta = \pm\pi/4$, we have the same $\cos\delta$, but two different $\sin\delta$, which denote two different polarization states. To measure the unique polarization state, $\sin\delta$ should also be determined independently. In this paper, we derive the complete formulae governing the polarization state measurement using a rotatable THz polarizer in THz-TDS. In this technique, we use only one rotatable THz polarizer, because a two-contact photoconductive THz detector can be considered as a fixed THz polarizer, followed by a polarization-insensitive detector. It responds only to the component of the electric field, which is parallel to the direction of the gap between two contacts. Therefore, only one rotatable THz polarizer is really required in this technique. The fixed THz polarizer, described in [15], is actually unnecessary.

With the successful measurement of the polarization states, THz-TDS can be applied to detect the polarization properties of a material. It is well-known that Mueller matrix governs all polarization properties of a material under test. Thus, the measurement of Mueller matrix using THz-TDS is of great importance. In optics, two

* Corresponding author.
E-mail address: hdong@i2r.a-star.edu.sg (H. Dong).

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input polarization states are needed, which should be orthogonal in Stokes space to statistically achieve the best measurement accuracy [16], for the measurement of Mueller matrix of a pure birefringent material. In THz-TDS, to generate two desired input polarization states, we have three options: (1) insert a rotatable THz waveplate between the THz emitter and the material under test; (2) change the output polarization state of the THz emitter; and (3) change the orientation of the material under test, which is equivalent to a relative change of the input polarization state. For option 1, the use of THz waveplate will induce excess loss. This will reduce the dynamic range of the THz-TDS system. For option 2, two techniques have been adopted. In the first one, the photoconductive emitter is mounted on a graduated rotation stage that allows the polarization state of the emitted THz wave to be rotated [6]. The second one uses a four-contact photoconductive THz emitter to generate two orthogonal output polarization states by rotating the bias voltages by 90° [8]. Apparently, the two techniques suffer from the problems of inhomogeneity of probe beam intensity, inhomogeneity of the electrical property of the photoconductive substrate and the imperfect shape of the contacts and gap. In this paper, the material under test is rotated. This solution is simple, does not introduce any excess loss and does not have problems that exist in the polarization modulation of the THz emitter. A theoretical analysis of this solution is presented in this paper and experimental results, on a quartz crystal, confirm the validity of the proposed technique.

2. Polarization state measurement in THz-TDS

In THz-TDS, the THz emitter launches sub-picosecond THz pulses that correspond to a frequency bandwidth of several terahertzes. In real space, two polarization components of a monochromatic electric field, with an angular frequency ω, is expressed in a given coordinated system as

$$\begin{cases} E_x = H\cos\omega t, \\ E_y = K\cos(\omega t+\delta), \end{cases} \tag{1}$$

where H and K are the magnitudes of the two components of electric field, respectively; t is the time and δ is the relative phase difference between two components. In Stokes space, the polarization state of an electromagnetic wave is expressed using four Stokes parameters given by [13]

$$\begin{cases} S_0 = H^2+K^2, \\ S_1 = H^2-K^2, \\ S_2 = 2HK\cos\delta, \\ S_3 = 2HK\sin\delta. \end{cases} \tag{2}$$

To measure the polarization state, a THz polarizer is placed in front of a two-contact photoconductive THz detector. We assume the direction of the gap between two contacts is along the x-axis. When the THz polarizer is rotated to an angle θ (θ is the angle between the x-axis and the polarizing axis of the THz polarizer), the resulting electric field E_θ at the THz detector is

$$E_\theta = A_\theta\cos\Theta_\theta. \tag{3}$$

where

$$\begin{cases} A_\theta = |\cos\theta|\sqrt{H^2\cos^2\theta+K^2\sin^2\theta+2HK\cos\theta\sin\theta\cos\delta}, \\ \Theta_\theta = \omega t+\varphi \end{cases} \tag{4}$$

and φ can be calculated using

$$\tan\varphi = -\frac{K\sin\theta\sin\delta}{H\cos\theta+K\sin\theta\cos\delta}. \tag{5}$$

A_θ and Θ_θ in Eq. (4) can be measured using THz-TDS. By rotating the THz polarizer to three specified angles: 0°, 45° and −45°, from Eqs. (3)–(5), we have

$$\begin{cases} H = A_{0^\circ}, \\ K = \sqrt{2\left[A^2_{45^\circ}+A^2_{-45^\circ}\right]-A^2_{0^\circ}}, \\ \cos\delta = \left(A^2_{45^\circ}-A^2_{-45^\circ}\right)\Big/\mathrm{HK}, \\ \sin\delta = (H^2-K^2)\tan(\Theta_{-45^\circ}-\Theta_{45^\circ})/2\mathrm{HK}. \end{cases} \tag{6}$$

The first three sub-equations in Eq. (6) were already presented in [15]. However, δ cannot be uniquely determined by the first three sub-equations. Now, since $\sin\delta$ can be calculated independently from the fourth sub-equation, δ can be uniquely determined. Substituting Eq. (6) into Eq. (2), we have the formulae for the Stokes parameter measurement as

$$\begin{cases} S_0 = 2\left(A^2_{45^\circ}+A^2_{-45^\circ}\right), \\ S_1 = 2\left(A^2_{0^\circ}-A^2_{45^\circ}-A^2_{-45^\circ}\right), \\ S_2 = 2\left(A^2_{45^\circ}-A^2_{-45^\circ}\right), \\ S_3 = 2\left(A^2_{0^\circ}-A^2_{45^\circ}-A^2_{-45^\circ}\right)\tan(\Theta_{-45^\circ}-\Theta_{45^\circ}). \end{cases} \tag{7}$$

If the THz wave is completely polarized, the normalized three-dimensional Stokes vector can be given as $\vec{S} = (s_1\quad s_2\quad s_3)^{\mathrm{T}} = (S_1\quad S_2\quad S_3)^{\mathrm{T}}/S_0$. Here "T" denotes the matrix transpose.

3. Mueller matrix measurement in THz-TDS

In this paper, we consider the Mueller matrix measurement only for a pure birefringent material. In this case, the Mueller matrix under test is a 3×3 orthogonal matrix and the polarization state is described by a 3×1 unitary Stokes vector as $\vec{S} = (s_1\quad s_2\quad s_3)^{\mathrm{T}}$ [13]. To obtain Mueller matrix, two input polarization states $\vec{S}_{\mathrm{in}}$ and $\vec{T}_{\mathrm{in}}$ are required to be launched into the material one by one. Moreover, $\vec{S}_{\mathrm{in}}$ and $\vec{T}_{\mathrm{in}}$ need to be orthogonal in Stokes space to achieve the statistically best measurement accuracy [16]. We will demonstrate, in this paper, that the desired $\vec{S}_{\mathrm{in}}$ and $\vec{T}_{\mathrm{in}}$ can be generated by rotating the material under test by an angle of 45°. The measurement procedure is in three steps:

(1) Measure the output polarization state of the THz wave emerging from the emitter when there is no material. This is just the input polarization state $\vec{S}_{\mathrm{in}}$ (if a THz polarizer is inserted between the THz emitter and the material under test, this step is unnecessary).
(2) Place the material between the THz emitter and the THz polarizer and measure the output polarization state $\vec{S}_{\mathrm{out}}$ emerging from the material.
(3) Rotate the material under test by an angle α and measure the corresponding output polarization state $\vec{T}_{\mathrm{out}}$.

To calculate the Mueller matrix $\mathbf{M}$ using the measured data, the following algorithm is adopted. First we have [13]

$$\begin{cases} \vec{S}_{\mathrm{out}} = \mathbf{M}\vec{S}_{\mathrm{in}}, \\ \vec{T}_{\mathrm{out}} = \mathbf{R}(-\alpha)\mathbf{M}\mathbf{R}(\alpha)\vec{S}_{\mathrm{in}}, \end{cases} \tag{8}$$

where $\mathbf{R}(\alpha) = \begin{pmatrix} \cos2\alpha & \sin2\alpha & 0 \\ -\sin2\alpha & \cos2\alpha & 0 \\ 0 & 0 & 1 \end{pmatrix}$ is a rotation matrix. And we have $\mathbf{R}^{-1}(\alpha) = \mathbf{R}(-\alpha)$, where $\mathbf{R}^{-1}$ denotes the inverse matrix of $\mathbf{R}$.

From the second sub-equation of Eq. (8), we have

$$\vec{\tilde{T}}_{\mathrm{out}} = \mathbf{M}\vec{T}_{\mathrm{in}}, \tag{9}$$

where $\tilde{\vec{T}}_{out} = \mathbf{R}(\alpha)\vec{T}_{out}$ and $\vec{T}_{in} = \mathbf{R}(\alpha)\vec{S}_{in}$. Eq. (9) shows that, if we consider the Mueller matrix **M** is unchanged by the rotation, the input polarization state should be changed to $\vec{T}_{in}$. The dot product of two input polarization states is

$$\vec{S}_{in} \cdot \vec{T}_{in} = \cos 2\alpha (s_{in1}^2 + s_{in2}^2) + s_{in3}^2, \tag{10}$$

where s_{in1}, s_{in2} and s_{in3} are three components of $\vec{S}_{in}$. We should choose an appropriate α to make the above dot product close to zero so that two input polarization states are nearly orthogonal in Stokes space. Since the THz emitter launches linearly polarized THz pulses ($s_{in3} \approx 0$), then $\alpha = 45°$ can lead to $\vec{S}_{in} \cdot \vec{T}_{in} \approx 0$. This will bring the statistically best measurement accuracy [16]. From Eqs. (8) and (9), we can calculate the Mueller matrix **M** using the following equation that [16]

$$\mathbf{M} = \mathbf{F}_{out}^{-1} \cdot \mathbf{F}_{in}, \tag{11}$$

where

$$\mathbf{F}_{out} = \begin{pmatrix} \vec{S}_{out}^{T} \\ \tilde{\vec{T}}_{out}^{T} \\ \left(\vec{S}_{out} \times \tilde{\vec{T}}_{out}\right)^{T} \end{pmatrix} \quad \text{and} \quad \mathbf{F}_{in} = \begin{pmatrix} \vec{S}_{in}^{T} \\ \vec{T}_{in}^{T} \\ \left(\vec{S}_{in} \times \vec{T}_{in}\right)^{T} \end{pmatrix}.$$

4. Experimental setup and results

An experiment is setup, as shown in Fig. 1, to verify the proposed theory and measurement method. In this setup, we use only one THz polarizer which is mounted on a rotatable holder. A Ti:sapphire femtosecond laser provides 10 fs optical pulses at 80 MHz repetition rate with a central wavelength of 800 nm. A two-contact photoconductive switch THz emitter is biased with a ±40 V square wave at a frequency of 65 kHz. The THz detector is also a two-contact photoconductive detector. A beam splitter splits the laser beam into the pump beam and the probe beam with an average power of 28 mW and 32 mW, respectively. The material under test and the THz polarizer are mounted on two separate rotatable holders. The optical time delay line and the lock-in amplifier are controlled by a computer. By moving the delay line and by recording the signal from the lock-in amplifier, the time-domain THz waveforms can be measured. The purging box is filled with nitrogen gas to avoid the absorption of the THz energy by water vapour.

Using this setup, three time-domain THz waveforms can be obtained one by one when the THz polarizer is rotated to the three angles: 0°, 45° and −45°. By doing Fourier transformation, and using the algorithm presented in Section 2, the spectrally-resolved Stokes vectors can be calculated. In the experiment, the polarization state of THz wave from the THz emitter is measured first. This is shown in Fig. 2 in the frequency range from 0.165 to 1.5 THz, which is $\vec{S}_{in} = (s_{in1} \quad s_{in2} \quad s_{in3})^T$ that was defined in Section 3. In Fig. 2, as well as the following figures, marks stand for the measured results and solid lines are the fourth-order polynomial fitting results based on the least-square method. Obviously when the frequency is larger than 1 THz, the circular polarization component s_3 increases [17].

When the material under test, a quartz crystal with a thickness of 5 mm, is inserted in the THz path, the output polarization state $\vec{S}_{out}$ is measured as shown in Fig. 3.

To achieve better measurement accuracy, the quartz crystal is then rotated by an angle of 45°. Based on Eqs. (8)–(10) and Fig. 2, although the real input polarization state $\vec{S}_{in}$ remains unchanged and the Mueller matrix has been rotated, we can assume that the Mueller matrix is unchanged and the input polarization state is changed to $\vec{T}_{in} = (s_{in2} \quad -s_{in1} \quad s_{in3})^T$. And obviously $\vec{T}_{in}$ is nearly orthogonal to $\vec{S}_{in}$ in Stokes space. Then the present output polarization state $\vec{T}_{out} = (t_{out1} \quad t_{out2} \quad t_{out3})^T$ emerging from the material is measured as shown in Fig. 4. Please note that we should use $\tilde{\vec{T}}_{out} = (t_{out2} \quad -t_{out1} \quad t_{out3})^T$, defined in Eq. (9), to calculate the Mueller matrix.

After $\vec{S}_{in}, \vec{T}_{in}, \vec{S}_{out}$ and $\tilde{\vec{T}}_{out}$ have been measured, we can calculate the Mueller matrices based on Eq. (11) in the frequency domain. For example, three calculated Mueller matrices at 0.5 THz, 1.0 THz and 1.5 THz are

$$\mathbf{M}(0.5\ \text{THz}) = \begin{pmatrix} -0.5762 & -0.0534 & 0.8216 \\ -0.0601 & 0.9797 & 0.1901 \\ -0.8334 & 0.1981 & -0.5312 \end{pmatrix}, \tag{12}$$

$$\mathbf{M}(1.0\ \text{THz}) = \begin{pmatrix} -0.2207 & 0.1382 & -0.9706 \\ -0.1183 & 0.9968 & 0.1516 \\ 0.9723 & 0.0212 & -0.2099 \end{pmatrix} \tag{13}$$

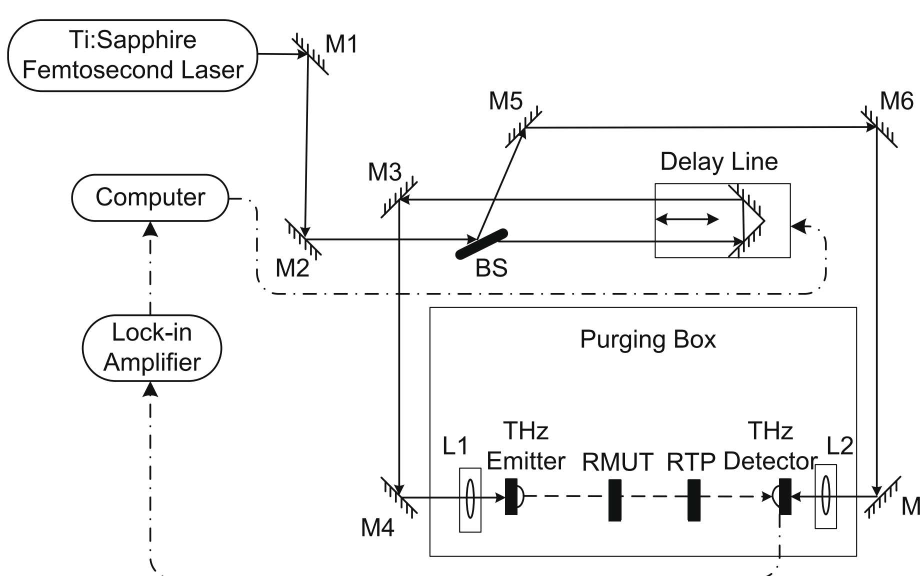

Fig. 1. Experimental configuration for THz polarization measurement in THz-TDS. M: mirror; L: lens; BS: beam splitter; RMUT: material in a rotatable holder; and RTP: THz polarizer in a manually-rotatable holder.

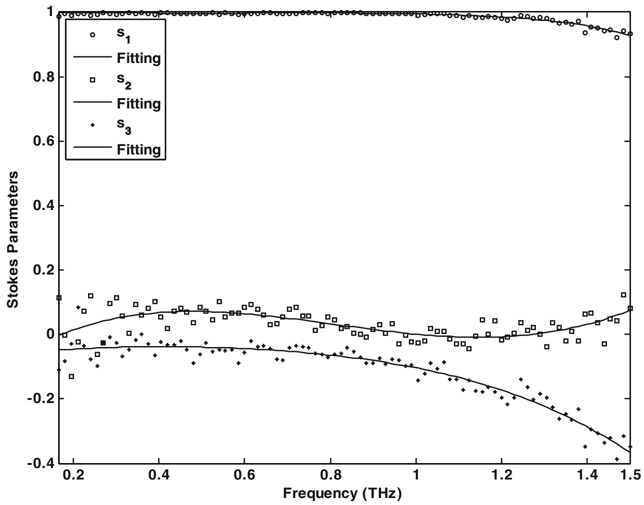

Fig. 2. Polarization state measurement results of Stokes parameters for the THz wave from the emitter.

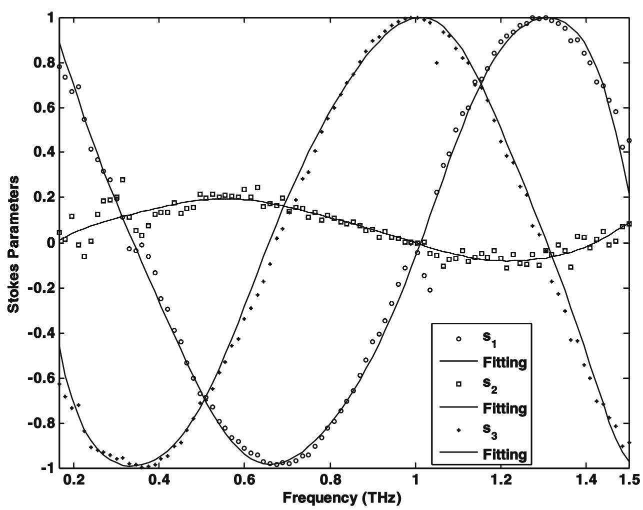

Fig. 3. Polarization state measurement results of Stokes parameters for the THz wave passing through a 5 mm thick quartz crystal.

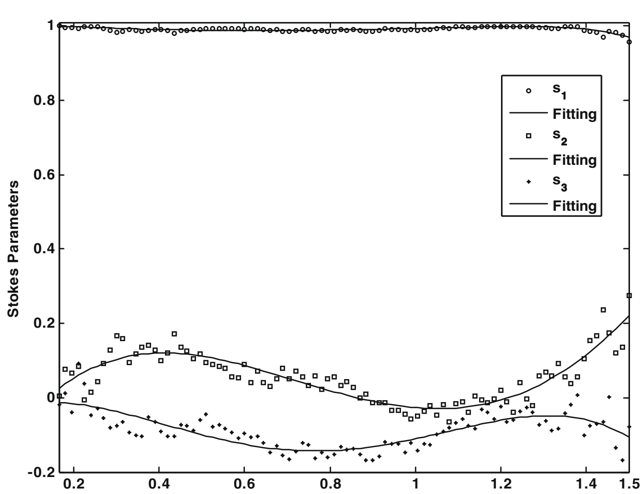

Fig. 4. Polarization state measurement results of Stokes parameters for the THz wave passing through the quartz crystal after a 45° rotation.

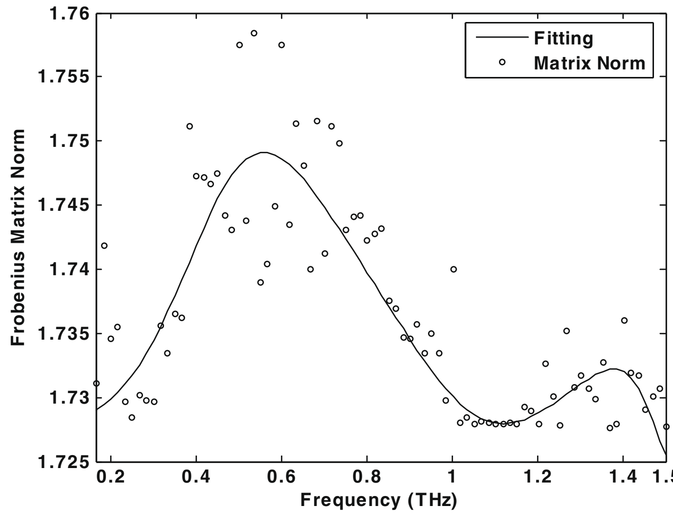

Fig. 5. The Frobenius matrix norms of the measured Mueller matrices.

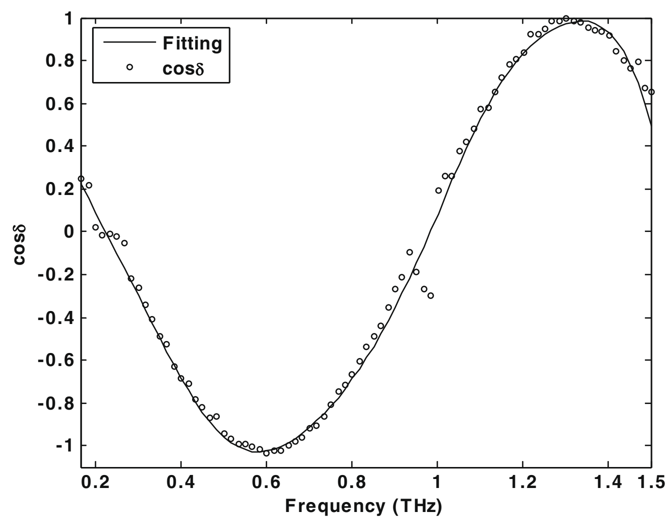

Fig. 6. Cosine of the phase retardance δ of the quartz crystal.

and

$$\mathbf{M}(1.5\,\mathrm{THz}) = \begin{pmatrix} 0.5476 & -0.4576 & 0.6777 \\ 0.1421 & 0.8710 & 0.4701 \\ -0.8077 & -0.1039 & 0.5598 \end{pmatrix}. \qquad (14)$$

Since the quartz crystal is a pure birefringent material, its Mueller matrix should be orthogonal. This means that the Frobenius matrix norms of the measured Mueller matrices should be $\sqrt{3} \approx 1.732$ [16]. This criterion can be used to verify the obtained Mueller matrices. Hence, we can calculate the Frobenius matrix norms and plot them in Fig. 5. Obviously the measured Mueller matrices meet this criterion with a relative error less than 1.5%, which also confirms the validity of our measurement technique.

With the obtained Mueller matrices, we can calculate the cosine of the phase retardance δ using $\cos\delta = [\mathrm{Tr}(\mathbf{M}) - 1]/2$ ("Tr" denotes the trace of the matrix $\mathbf{M}$) [18] and results are shown in Fig. 6.

5. Conclusion

We present the algorithm to uniquely determine the four Stokes parameters using one rotatable THz polarizer to measure the polarization state of THz wave in THz-TDS. Based on this technique, we can obtain the Mueller matrix of a material using the THz-TDS setup. To achieve a simple measurement approach, we propose to rotate the material under test. A 5 mm thick quartz crystal is used as a test sample for the proposed techniques. To the best of our knowledge, this is the first reported measurement result of Mueller matrix in THz-TDS. Of course, if the material under test is very large, or has to be stored in a cryostat, or is a living being, rotation of the same may be inconvenient. In these situations, we still need to find other techniques, as mentioned in Section 3, to control the input polarization states. However in most of the usual tests, rotating the material is simple and cost-effective.

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