Classification of terahertz spectrometer for transmittance measurements of refractive materials

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Abstract: A comparison between commercially available transmission terahertz spectroscopy systems using photoconductive antennas owned by three participating institutions was performed to verify the effects of the specifications of each system on the transmittance measurement uncertainty. Three types of sample with different compositions and shapes were circulated around the participating institutions, and the data obtained from their systems were compared. We verified that sample thickness and its non-parallelism affected measurement data by carrying out measurements and calculations. On the basis of the components of variations in the measurement data, we tried to classify the five measurement data sets. The results indicated that the system users can easily compare their measurement conditions with those of other systems using statistical analysis.

Keywords: transmittance, photoconductive antenna, terahertz spectroscopy, statistical analysis

Classification: Optical systems

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1 Introduction

Applications in the terahertz (THz) range whose frequency is approximately from 0.1 to 3 THz have become increasingly common [1, 2] since the development of THz time-domain spectroscopy (TDS) [3] in the 1980s. As a result, quantitative TDS measurements are required more than ever before. Furthermore, wireless communication at 120 GHz [4] was launched in 2014, and a 300 GHz system is also under development [5]. Accurate dielectric constants of various substances in the terahertz band will be needed, and thus we consider wave properties such as propagation and absorption in various materials.

On the other hand, a commercial THz-TDS system that can measure relative complex permittivity has no common measurement protocol, and the measured data may vary depending on the system and sample properties [6]. In particular, a sample configuration that changes the terahertz beam propagation direction is a dominant factor for variations owing to the small size of the photoconductive antenna for detecting terahertz beams. To obtain valid measurement results, it is necessary to understand optical specifications such as the beam radius and mirror position. However, many system users hardly check the optical specifications inside a system box in detail. Therefore, an easy statistical method is needed to examine measurement conditions from a comparison with several different systems.

In this paper, we have therefore performed the comparison test among three institutes to verify the effects of the specifications of each system on the transmittance measurement uncertainty, which affects the calculation of the relative complex permittivity. On the basis of the components of variations in the study data, we examined whether optical conditions can be classified.

2 Samples and systems

2.1 Samples and measurement procedures

Three types of sample with different compositions and shapes were circulated around the participating institutions. The first set of samples were thin-film attenuators (four samples), whose sample holder diameter and thickness were φ22 and 5 mm, respectively. The amounts of attenuation were designed to be approximately 3 and 12 dB for a film thickness of 25 µm, and 6 and 13 dB for a thickness of 6 µm. The thin-film attenuators were developed by AIST [7]. To fabricate these attenuators, a metal (Inconel®) layer was deposited onto a polyethylene terephthalate (PET) film by evaporation to form a transmittance-controlled double-layer thin film, which was fixed using a metallic holder. Samples fabricated by AIST were circulated around the participating institutions. The transmittances of the samples...
obtained using their systems were compared. The agreement among the measurement results was examined.

The second set of samples were azurite pellets (one reference pellet and five samples), whose diameter and thickness were φ13 and approximately 1.6 mm, respectively. The mixing ratio (weight ratio) of azurite/polyethylene (PE) was 0.02. Here, the grain diameter was less than 20 µm, and the applied pressure was 15 kN for 10 min. The azurite pellets were fabricated by mixing azurite \([\text{Cu} (\text{CO}_3)_2 (\text{OH})_2]\) crystal powder with high-density polyethylene (HDPE) powder and compressing the mixture into pellet form. Azurite exhibits absorption peaks in the frequency range of 0.1–3 THz, which can be covered by general TDS systems. The samples fabricated by NICT were circulated around the participating institutions. The effect of thickness nonuniformity, which is a problem arising in the fabrication of pellets, was examined.

The third set of samples were high-resistivity silicon plates (four samples), whose diameter and thicknesses were φ15 and 0.5, 1.0, 2.0, and 3.0 ± 0.1 mm, respectively. The resistivity was over 10 kΩ·cm and the floating zone (FZ) method was employed for single-crystal growth. The high-resistivity silicon plates are industrial products with high permittivity, low loss, and uniform thickness, and the changes in their physical constants attributed to environmental factors are small. Four samples with different thicknesses (specifications other than the thickness were the same) were circulated around the participating institutions to compare the transmittances obtained using their systems. The effect of thickness was examined.

Photographs of the samples are shown in Fig. 1. In these measurements, the samples were set perpendicular to the direction of terahertz wave propagation. In the case of film samples, the angle between a film and its holder was approximately 2° to minimize the interference effects caused by reflected waves. Other sample data have information of only a direct wave by eliminating the reflection waves with time domain gating to simply analyze the data.

### 2.2 Systems

Five types of TDS systems with different optical systems (named A, B, C, D, and E), as shown in Table I, were used in the participating institutions. System A was a typical TDS system with a femtosecond (fs) laser and two photoconductive antennas comprising of a small dipole antenna for THz generation and detection. The optical system of System A was in the focusing mode, and the sample chamber...
could be purged with dry air under atmospheric pressure (humidity less than 2%). System B has a generator comprising a trans-4’-(dimethylamino)-N-methyl-4-stilbazolium tosylate (DAST) crystal [8], and its other components are similar to those of System A. System C was also a typical TDS system similar to System A. System C could evacuate air including vapor from a sample chamber using a vacuum pump. In the case of System D, the optical system was in the parallel mode, and measurements were carried out at atmospheric pressure without purging with dry air. The measurement conditions of System E were also no purging, but the optical system was a typical TDS system with the focusing mode similar to those of Systems A and C, even though the optical configurations differed. The ambient temperature and humidity around the samples were different depending on the measurement system or date. The type of THz source and detector, optical mode and configuration, and humidity could affect the measurement results. The number of spectra to average was selected as the number of measurements required to achieve a high accuracy for each system. The reference spectra for the systems in this study are shown in Fig. 2. Here, the spectra were normalized by the maximum intensity to enable their comparison. We consider that the study data can be compared in the frequency range from approximately 0.5 to 3 THz even though the dynamic ranges were different.

3 Results and discussions

3.1 Measurement of attenuators

The thin-film attenuators were circulated around the participating institutions to compare the transmittances (amplitude change) measured using their respective
systems. In this measurement, the position of the samples was fixed, and the mean and standard deviation ($\sigma$) of the measured values obtained from five successive measurements were plotted. As shown in Fig. 3, the deviations in the measured values among Systems A–E were small. Note that large errors were observed at approximately 1 THz for System B. This was due to the low THz signal absorbed by the organic nonlinear optical crystal (DAST) used to generate terahertz waves. System B can obtain comparable data at frequencies over 1.5 THz, even though the generator of System B differs from those of other systems. The relative deviations were smallest at 2 THz, particularly in the case of the 6 dB attenuator, and the values were in agreement within $\pm 2\%$ (Fig. 4). Larger attenuations (12 and 13 dB) caused a low signal to noise ratio, whereas lower attenuations (3 dB) clearly reflected the THz source noise. Therefore, we considered using a 6 dB thin-film attenuator as a fundamental sample in statistical analysis. If the spectrum of the 6 dB attenuator was different from those of other systems, we could predict measurement errors.

### 3.2 Measurement of mixed pellets

The transmittance of each azurite pellet (Samples a–e) was measured once using each system and the mean of each set of five values and the standard deviation ($\sigma$)
are indicated in Fig. 5. The positions of the peaks at approximately 1.8 and 2.2 THz were almost in agreement among the five systems. Specifically, Table II shows the absorption peak frequencies of Sample a observed near 2.2 THz. In the measurement, the frequency resolution of each system was set to the maximum value. The results showed that the measured values were in agreement among the five systems to one decimal place.

On the other hand, the variations in the transmittance among the samples became large at high frequencies. The magnitude of the standard deviation was different between the focusing-mode systems (Systems A, B, C, and E) and the parallel-mode system (System D). To determine the cause of the difference, the measurement using System C and D was performed by rotating a sample (θ = 0, 90, 180, and 270°) with respect to an arbitrary position on the sample, as shown in Fig. 6. The maximum variations in thickness for the reference and sample pellets were approximately 70 and 65 µm, respectively. Marked differences in transmittance were only observed for System D. The erroneous value of the transmittance of over 100% was due to the intensity of the terahertz waves passing through the reference pellet being lower than that of the terahertz waves passing through the sample. We simulated the changes in current magnitude at the center of a small dipole antenna (50 µm-length and 10 µm-width) on GaAs substrate, using electromagnetic field simulation software (FEKO suite 7.0.2 version, Altair Engineering,

![Transmittance spectra of azurite pellets (Samples a–e).](image)

**Table II.** Absorption peak frequencies of pellet sample a

| System | Resolution [THz] | Absorption peak frequency [THz] | Mean value [THz] | 1σ [THz] |
|--------|------------------|---------------------------------|-----------------|---------|
| A      | 0.0075           | 2.2350 2.2350 2.2425 2.2500 2.2575 2.2440 | 2.2385           | 0.0098  |
| B      | 0.0075           | 2.2350 2.2350 2.2350 2.2350 2.2425 2.2365 | 2.2335           | 0.0034  |
| C      | 0.0061           | 2.2323 2.2323 2.2323 2.2384 2.2323 2.2323 | 2.2323           | 0.0027  |
| D      | 0.0061           | 2.2323 2.2323 2.2323 2.2323 2.2323 2.2323 | 2.2323           | 0.0000  |
| E      | 0.0244           | 2.2461 2.2461 2.2461 2.2461 2.2461 2.2461 | 2.2461           | 0.0000  |
| Mean   |                  |                                 | 2.2385           |         |
Inc.). Here, we constructed two simple small-sized parallel-mode models, one with a parallel and one with a non-parallel PE plate, whose diameter and average thickness were $\phi 6\text{mm}$ and 0.8 mm, respectively. The minimum and maximum thicknesses of the non-parallel PE plate were 0.835 and 0.765 mm, respectively. The slope direction of PE thickness was vertical to the THz electric field direction, and the interval between the substrate and the PE plate was 50 mm. Plane waves were irradiated to the PE plate, and then only the direct waves through the plate were focused on the antenna, using a silicon semispherical lens. Fig. 7 shows the ratio of the current magnitude $I$ depended on for the conditions of the PE plate ($I_{\text{parallel}}/I_{\text{nonparallel}}$). The current ratio increased with the increase of frequency. Thus, the receiving level of the antenna lowered, in particular at high frequencies when the non-parallel PE plate was placed in the parallel-mode model. Therefore, the erroneous values shown in Fig. 6 were considered, because of the large variation of the propagation direction of the terahertz waves when the non-parallel pellet was placed. In the case of the focusing mode, the effect was not significant compared with the parallel mode. These results indicate that the non-parallel pellet can be used to classify the optical systems.

### 3.3 Measurement of high-resistance silicon

The transmittance of a 0.5-mm-thick high-resistivity silicon plate was measured five times successively without changing the position of the sample so that the

![Fig. 6. Changes in transmittance by rotating a pellet sample in the measurement using Systems C and D.](image-url)
conditions of the sample and optical system were kept constant. Fig. 8 shows the mean and standard deviation (1σ) of the transmittance. At frequencies over 1.5 THz, the spectra obtained using Systems D and E were similar and we found differences between the results obtained using these two systems and the other systems.

To determine the reason for the difference, the absorption coefficients of four samples with different thicknesses were calculated (from the amplitude and phase) and compared. In Fig. 9, the squares, circles, and triangles respectively show the results of measuring the absorption coefficients at 1.2, 1.5, and 2.1 THz using Systems C and D. The absorption coefficients tended to increase with the thickness and frequency when incident terahertz waves were focused. When parallel terahertz waves were incident, the dependence of the absorption coefficient on the thickness of the samples was not significant, and the variability of the frequency characteristics was also small. As shown in Ref. 9 the absorption coefficients of a high-resistivity silicon plate at 1.2, 1.5, and 2.1 THz were found to be approximately 0.70, 0.63, and 0.48 cm$^{-1}$, respectively. We considered that the differences between

Fig. 7. Changes in the ratio of the current depended on the condition of the PE plate ($I_{\text{parallel}}/I_{\text{nonparallel}}$).

Fig. 8. Transmittance spectra of 0.5-mm-thick silicon plate.
the reference values and the thick sample data measured using the parallel mode were caused by the slight thickness distribution and slope of the sample against a holder. The values obtained using the focusing-mode system were greater than the reference values. Because the silicon materials used for the plate samples were single-crystalline and homogeneous, regardless of their thickness, the absorption coefficients and the shape of the absorption spectra should be constant. However, the absorption coefficient was affected by the sample thickness. In addition, in the case of an electromagnetic simulation example using the focusing-mode model with short focus parabolic mirrors, a significant decrease in the receiving level of the antenna was observed, even at 500 GHz, when a silicon plate was placed. Here, note that it was difficult to conduct the simulation at the actual size of the optical system, due to the huge memory needed to carry out calculations.

To examine the factors contributing to the variations in the data obtained using focusing-mode systems, we used ray tracing. As shown in Fig. 10, the following two changes occur in focusing-mode system when a high-resistivity silicon plate sample is placed. (i) The focal point of a terahertz wave at the sample position moves toward the light-receiving antenna, and the diameter of the terahertz beam incident to a parabolic mirror decreases, resulting in the increased radius of the focused terahertz beam. The beam radius \( r_0 \) at the beam waist can be derived from Eq. (1) [10]. Here, \( \lambda \), b, and D are the wavelength, the distance between the parabolic mirror and beam waist position, and the diameter of the terahertz beam on

Fig. 9. Effect of silicon thickness on absorption coefficient at 1.2, 1.5, and 2.1 THz. Dotted lines are calculation results for System C and reference values for System D.
the mirror, respectively. (ii) The focal point of the terahertz wave focused via the parabolic mirror and silicon hemispherical lens moves in the z-direction in Fig. 10, resulting in the increased radius of the terahertz beam on the light-receiving antenna. The beam radius \( \omega \) on the antenna can be derived from Eq. (2). Here, \( z \) is the distance from a focal point. \( \alpha \) in Eqs. (1) and (2) is an empirical compensation coefficient related to diffraction, corresponding to \( M^2 \), in this paper.

\[
\omega_0 \sim 2\alpha\lambda b/\pi D \\
\omega^2(z) = \omega_0^2[1 + (\alpha\lambda z/(\pi\omega_0^2))^2]
\]

As a result of the increased radius of the terahertz beam incident to the light-receiving antenna, the power \( P \) detected by the antenna, as expressed by the following Eq. (3), decreases and leads to an increase in loss. Here, \( A \), \( W \), and \( I \) are the effective area of the antenna, the power density, and the terahertz beam intensity, respectively.

\[
P = AW = AI/\pi\omega^2
\]

When the effective area of the small dipole antenna used in the spectroscopy system and the intensity of the generated terahertz beam are assumed to be constant, the power detected by the antenna decreases as the radius of the terahertz beam increases. The approximate losses caused by the changes in the beam radius were calculated from Eq. (4) using ray tracing, as shown in Fig. 11. We then calculated the absorption coefficient from each loss and the reference value. To obtain the above \( \omega_0 \) and \( z \), we used values of 0.89, 0.88, and 0.85 mm for the 1/e² intensity beam radius (FWHM values of approximately 1.02, 1.01, and 0.99 mm) measured at the sample position in the focusing mode at 1.2, 1.5, and 2.1 THz, respectively. It was assumed that the distances \( a \) and \( b \) defined in Fig. 11 were equal (\( a = b = 2\Omega = 150 \text{ mm} \)) and that the divergence angle \( \theta_b \) (\( \sim \alpha\lambda/\pi\omega_0 \)) was approximately equal to \( \tan \theta_b \). From these values, the diameter of the terahertz beam on the mirror \( D \) was calculated using Eq. (5). The radius and position of the silicon hemispherical lens were also assumed, however the different assumptions did not significantly affect the loss.

![Fig. 10. (Left) Schematic of System C and (right) variation in THz beam radius on the receiving antenna.](image-url)
\[
\text{Loss} = -\log\left(\frac{P_{\text{sample}}}{P_{\text{ref}}}\right) = \log\left(\frac{o_{\text{sample}}}{o_{\text{ref}}}\right)
\]

(4)

\[
D \sim 2a \cdot \theta_d - 2d\left(\tan\theta_d - \tan[\sin^{-1} \left(\frac{\sin \theta_d}{n}\right)]\right)
\]

(5)

Fig. 9 show that the tendencies of the results calculated using Eqs. (1)–(5) for the focusing mode and the reference values for the parallel mode (dashed lines) at 1.2, 1.5, and 2.1 THz were in agreement with the measured absorption coefficients (squares, circles, and triangles). When the empirical compensation coefficients were assumed to be 1.1, 1.5, and 1.9 at 1.2, 1.5, and 2.1 THz, respectively, the calculated results were close to the measured values. This indicates that the beam quality is high at 1.2 THz, near the center frequency of the photoconductive antenna. The radius of the terahertz beam incident to the light-receiving antenna was significantly related to the cause of the variations in measurement data; the beam radius at the sample position significantly affected the measured value. The results suggest that the measurement data of the silicon plate is suitable as second data for the system classification.

### 3.4 Comparison of systems

On the basis of the above-mentioned results, we used the transmittance data of a 6 dB thin-film attenuator, a non-parallel azurite pellet, and a 0.5-mm-thick high-resistivity silicon plate to examine whether measurement conditions can be classified. In the case of the pellet, standard deviations were used. Data under 1.5 THz and over 2.5 THz were eliminated because the deviation of System B at 1 THz and vapor significantly changed a classification result. Here, we assumed that the criterion data were the mean of five attenuator data, standard deviation of 0 at all frequencies, and the interpolation data of the reference values of silicon [9]. The criterion data were subtracted from the measurement data, and then integrated variances were indicated in a three-dimensional graph. In Fig. 12, we verified that we could classify five systems into two groups, two systems (D and E), and the others as discussed in a previous section. In the case of System E with the focusing mode, we inferred that the beam radius at the sample position was larger than that in the case of other focusing-mode systems, indicating that the optical condition was close to that of System D.
From the graph and compared spectra, the systems included in the X group are typically of focusing mode, which cannot precisely measure a thick sample. In the Y group, system users should consider that their system may have properties similar to those of the parallel mode even if called focusing mode. The best system is closest one to the coordinate point $(0,0,0)$ shown with red circle in Fig. 12. These results suggest that THz-TDS system users and developers can easily compare their measurement conditions with those of other systems using the three type samples and statistical analysis database including data in this study.

4 Conclusions

Comparisons of transmittance measurements using thin-film attenuators, non-parallel azurite pellets, and silicon plates with different thicknesses were performed. We discussed the variations in transmission measurement related to the changes in beam pass and radius against a small photoconductive antenna. On the basis of the components of the variations, we verified that we could classify the five measurement conditions. The results indicate that the spectral database will provide system users with the statistical relationship between their measurement condition and deviation.

In future works, we intend to expand targets to electro-optic detectors, and also cooperate with the manufacturers of spectroscopy systems and overseas standardization institutions.

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