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RESEARCH ARTICLE

Investigation of aqueous acetone solution with THz pulse spectroscopy

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Abstract The absorption spectra from 0.2 to 1.7 THz of five water-acetone mixtures, whose concentrations are 0%, 25%, 50%, 75%, and 100%, has been investigated by THz time-domain spectroscopy (THz-TDS). The result indicates that of acetone solutions with different concentrations have different absorption for THz radiation, which is analyzed and attributed to the variation of the hydrogen bond. Furthermore, we also find the absorption of the water-acetone mixture almost linearly increases with the increase of water concentration in the THz range.

Keywords THz time-domain spectroscopy (THz-TDS), relative absorption coefficient, aqueous acetone solution

1 Introduction

THz radiation, generally in the frequency range of 0.1–10.0 THz, belongs to the far infrared electromagnetic radiation range. Since there had been no effective methods to generate and detect THz wave for a long time, the THz range was initially called “THz gap”. In the beginning, optical rectification was used to generate THz radiation [1], through which the generated THz signal had not been widely used until the existing of high-power mode-lock lasers in the late of 1980s. Nowadays, THz time-domain spectroscopy (THz-TDS) has been utilized in a wide range of research fields, such as biological molecules identification, materials investigation, explosives inspection and medicine analysis [2–6]. Compared to traditional detection methods, such as infrared radiation (IR), X-ray, etc, THz-TDS has many advantages, i.e., it is insensitive to the thermal background and has a high signal to noise ratio (SNR). At the same time, THz-TDS can penetrate many non-polar materials. Unlike X-ray, as a kind of non-ionizing radiations, THz wave can be used as a non-destructive detection technique that can provide information about physicochemical properties, such as composition, structure, physical state and rheology, which means THz spectroscopy is becoming a competitive method that can identify different materials. However, up to now, there are few papers focusing on investigating the property of aqueous solution, in that water shows strong absorption at THz frequencies ($\alpha = 200\ cm^{-1}$ at 1.0 THz) [7,8] in normal case.

In this paper, water-acetone mixtures with different water content have been investigated by using THz-TDS and the different responses of these solutions in the THz region are obtained. Acetone solution with high content water has a large number of hydration bonds, which shows a strong adsorption capability in the THz range. Furthermore, we find the absorptivity of the 100%, 75%, 50%, 25%, and 0% acetone solutions almost linearly increases when the concentration of water content increased, which indicates that the hydration bonds in the water play an important role for the THz wave absorption. This work contributes to the build of water content detection of mixed liquid solution.

2 Experiment

The THz-TDS experimental setup is illustrated in Fig. 1. Coherent Ti: sapphire laser (central wavelength at 800 nm, pulse duration around 80 fs, repeat frequency at 76 MHz, and output power at 1.1 W) was applied to pump and detect THz wave. The laser was split into pump beam and probe beam, and the former was focused on ZnTe crystal. The THz setup had four parabolic mirrors arranged in a confocal geometry [9]. TheTHz wave emitted from GaAs intrinsic Schottky diode was gathered by parabolic M1. In order to compress the THz beam to a diameter comparable to the size of small samples, an additional pair of parabolic

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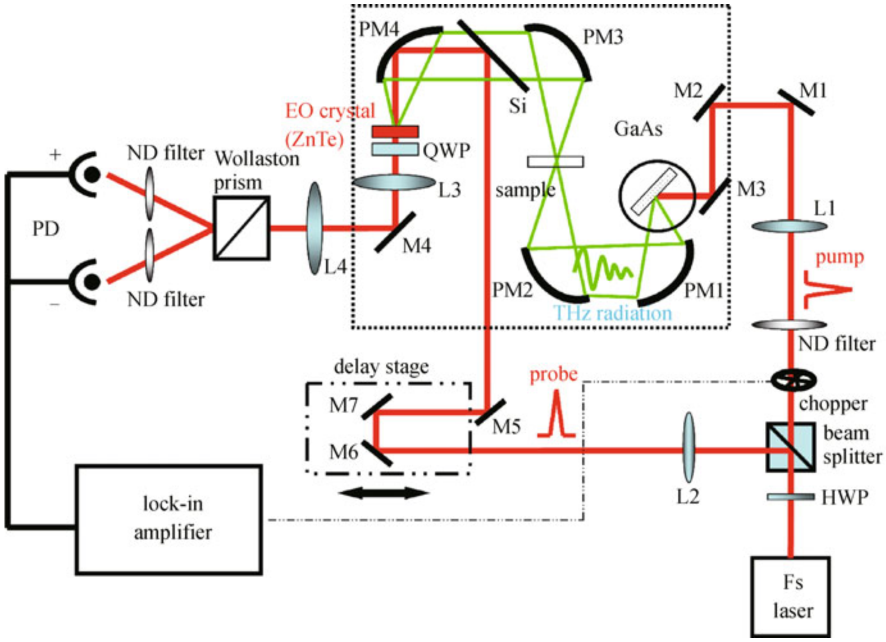

Fig. 1 Schematic diagram of THz-TDS system. M1-M7: reflecting mirrors, L1-L4: lenses, PM1-PM4: off-axis parabolic mirrors, Si: high resistant silicon wafer, EO: electro-optic crystal, HWP: half-wave plate, QWP: quarter-wave plate

mirrors M2 and M3 was employed. The THz waves with the information of samples, together with the probe beam, gathered by the parabolic M4 were focused onto ZnTe electro-optical (EO) crystal. Then, the probe beam was split into two beams with orthogonal polarizations by Wollaston prism, which was demodulated by balanced photodiode. To avoid the strong absorption of vapor, nitrogen gas was infused into a covered box which is indicated as dashed line in Fig. 1. The humidity in the covered box is kept less than 8% and the temperature was maintained at 23.5°C to 24.5°C.

Figure 2 shows the schematic diagram of the sample box made by high-density polyethylene (HDPE) for THz transmission experiment. THz signal from the sample box without and with sample information, $E_{ref}(t)$ and $E_{sam}(t)$, respectively, called reference and sample signals. Furthermore, the THz spectra of the reference and the samples, $E_{ref}(\omega)$ and $E_{sam}(\omega)$, can be obtained from the Fourier transform of the transmitted THz pulses. In this experiment, six groups of samples were used. Group 1 just included a piece of filter paper in the sample box as reference. Group 2–6 included a piece of filter paper dipped with 0%, 25%, 50%, 75% and 100% acetone solution, respectively. In order to accurately obtain the thickness of samples together with their power absorption coefficient, the samples were gently placed between HDPE slabs, which have excellent optical transmission properties in THz wave range [10]. The thicknesses of the samples were measured by micrometer.

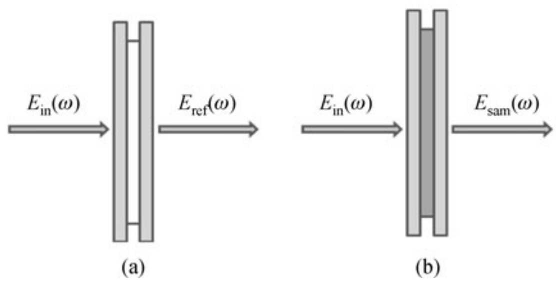

Fig. 2 Schematic diagram of the structure of sample boxes. (a) Reference cell with the structure of HDPE-filter paper-HDPE; (b) sample cell with the same structure filled with the solution of acetone

3 Results and discussion

THz waveforms of the reference and five samples in time domain and their corresponding spectra from Fourier transform are shown in Figs. 3(a) and 3(b), respectively. It is obvious that the THz traces of five samples have different delay time comparing with the reference signal, which is caused by the difference absorption coefficient of the solutions. From Fig. 3(b), it can be found that the amplitudes of transmitted spectra of the samples are much small comparing with the situation without acetone solutions, which indicates that THz waves are strongly absorbed by the acetone solutions in the THz range.

It is well known that the intensity I of electromagnetic

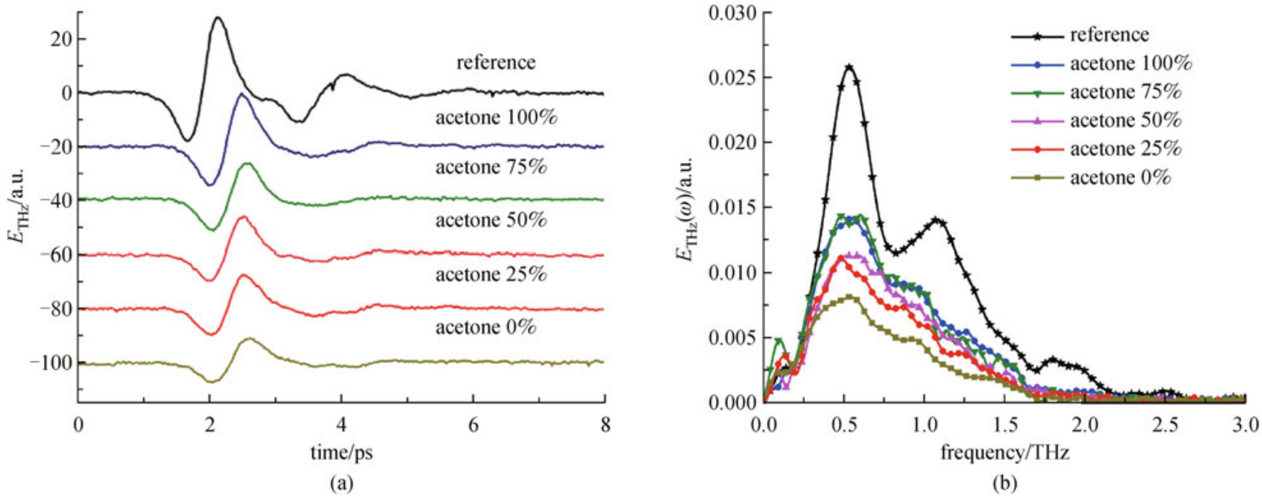

Fig. 3 (a) Transmitted THz pulses of the vacant sample box and 0%, 25%, 50%, 75% and 100% acetone solution; (b) the corresponding THz spectra from the Fourier transform of time domain waves

radiation should decrease exponentially with the permeation distance d:

$$I = I_0 \exp(-\alpha d), \quad (1)$$

where α is absolute absorption coefficient. Meanwhile, from the reference and sample signals, we can obtain,

$$I_{\text{reference}} = I_{\text{in}} \exp(-\alpha_{\text{ref}} d), \quad (2)$$

and

$$I_{\text{sample}} = I_{\text{in}} \exp(-\alpha_{\text{sam}} d), \quad (3)$$

where α_{ref} and α_{sam} are the absolute absorption coefficients of the vacant sample box and samples, d is the thickness of the vacant sample box and the samples. Then we can gain the relative absorption coefficients of these samples by Bouguer-Lambert-Beer law,

$$\alpha = -\ln\left(\frac{I_{\text{sample}}}{I_{\text{reference}}}\right). \quad (4)$$

The relative absorption coefficients of these acetone solutions, as shown in Fig. 4, can be obtained easily by using Eq. (4), which shows that the five samples have different absorption spectra within the range from 0.2 to 1.7 THz. Among these samples with different water content, 100% aqueous solution has the highest absorptivity. The 100% acetone solution shows relatively low absorptivity in the THz frequency range due to the lack of hydrogen bond. We can clearly see that when the concentration of water increases, the absorption coefficient becomes larger.

Furthermore, the relative absorption coefficient at 1 THz is selected to analyze the relation between the THz absorption coefficients and the concentrations of acetone solutions. The result as shown in Fig. 5 indicates that there is almost linear relation between them. This can be explained by molecular vibration theory: water has very high absorption coefficient due to the vibration of hydrogen bond [11], whose frequency locates in the THz range. Water molecule provides two hydrogen bonds, whereas these bonds do not appear in pure acetone solution. When the acetone is mixed by the water, the concentration of hydrogen bonds increases, which causes the relative large absorptivity of the mixed solution. The relative absorption coefficient provides a quantitative response of the water solutions to THz radiation, which is a new method to identify various types of water solutions by using THz spectroscopy technique.

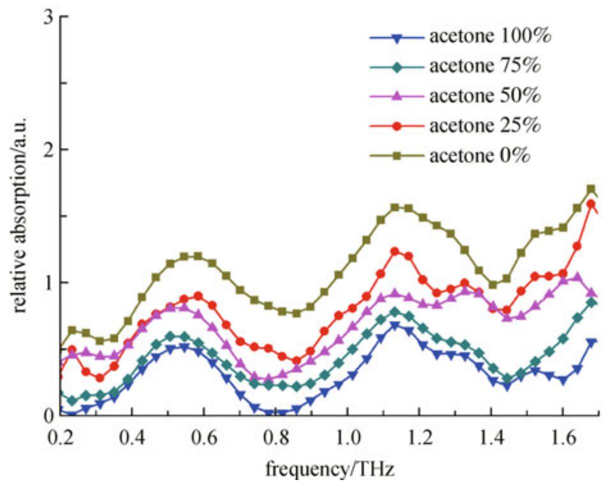

Fig. 4 Relative absorption of 0%, 25%, 50%, 75% and 100% acetone-water mixture

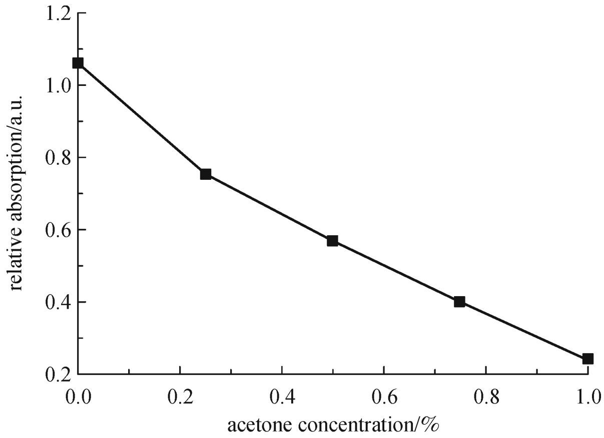

Fig. 5 Relation between relative absorption and concentration of acetone in acetone-water mixture at 1 THz

4 Conclusions

We used THz-TDS system to measure five groups of acetone solutions with different content of water and obtained their relative absorption coefficients in the THz frequency range. We find that when the water content increases, the absorption coefficient becomes larger. Molecular vibration theory is used to explain this phenomenon, and it can be concluded that the hydration bonds in the water play an important role for the absorption of these solutions in the THz frequency range. This work contributes to the build of water content detection of mixed liquid solution.

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