Temperature Dependence of Terahertz Wave Generation from DAST Crystal Illuminated by 1.56 μm Fiber Laser

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Abstract: 4-Dimethylamino-N-methyl-4-stilbazolium tosylate (DAST) crystal can generate broadband terahertz (THz) wave by optical rectification using femtosecond laser pulses. However, the frequency spectrum of generated THz waves has dips derive from the absorptions of the DAST crystal. We observed temperature dependence of THz wave generation from the DAST crystal. In terms of the frequency spectrum, broad dips at 1.1 and around 4.5 *THz* at room temperature shift to higher frequency with temperature decreasing, while a small dip at 3 *THz* shows no temperature dependence.

Keywords: Terahertz, DAST crystal, absorption, temperature dependence

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1. Introduction

Terahertz (THz) time-domain spectroscopy (TDS) and THz imaging have been utilized for applications in biological and fundamental materials sciences [1, 2]. Many applications require high power and broad-bandwidth THz wave generation. In general, semiconductor photoconductive antennas and surfaces of semiconductor have been used as typical THz wave sources using femtosecond pulsed laser [3-5]. Recently, a high power and broad THz wave generation from 4-Dimethylamino-N-methyl-4-stilbazolium tosylate (DAST) has been reported, which is a nonlinear organic crystal, using femtosecond laser pulses excitation with a wavelength of 1.56□ *μm* [6]. The DAST crystal has a great potential to be used as high power and broad bandwidth THz wave source for many applications, whereas the DAST crystal has several characteristic absorption peaks in THz region. The absorptions of the DAST crystal adversely affect the result of THz-TDS, because of a decrement of signal to noise ratios and an increment of instabilities at specific frequency. Optical properties of the DAST crystal were researched in THz region from 0 to 3 *THz* [7]. However, the origin of absorptions is not understood well. In this study, to investigate shift and peak amplitude change of the absorption of the DAST crystal, we measured THz wave generation from the DAST crystal at low temperatures and broad optical properties of the DAST crystal using a THz-TDS and cryo-cold head system.

2. Experimental Methods

In this study we measured the waveform of THz pulses generated from the DAST crystal, and

the waveform transmitted through the DAST crystal using a THz-TDS system equipped with the DAST crystal as a emitter, a dipole type low-temperature-grown GaAs (LT-GaAs) photoconductive switch as a detector, and Er doped fiber laser (λ = 1.56 μm; pulse width: 100 *fs*) as a femtosecond pulsed laser source. The femtosecond laser pulse is first separated into a "pump pulse" and a "probe pulse." The pump pulse is polarized in [100] *a* axis of the crystal for effective THz generation. The THz pulse is generated at the DAST crystal by the pump-pulse illumination and collimated by using an off-axis parabolic mirror and then collimated again onto the LT-GaAs detector by using another mirror. Fig. 1 shows a schematic diagram of the system. Since the system equips cryostat and vacuum vessel which encloses the detector, the emitter and all path of THz pulse wave, generated THz pulses are not affected by air and optic window. When we measure the absorption of the DAST crystal, the sample is mounted on a copper plate connected with a cold head of the cryostat. The temperature of the sample can be controlled from 15 *K* to room temperature, and is corrected by a silicon diode mounted on the copper plate near the sample. All DAST crystals in this study are grown by the slow-cooling technique coupled with a new creation method, which is described in detail in previous reports [8, 9]. The experimental results of THz-TDS are the average of five measurements.

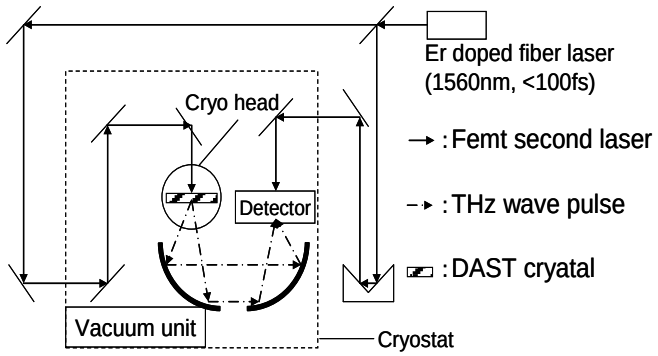

Fig. 1 Schematic diagram of the system for THz-wave generations from the DAST crystal.

3. Results

Fig. 2(a) shows the time domain spectra of generated THz wave from the DAST crystal at temperatures of 15, 46 and 295 *K*. The frequency spectra obtained from fast Fourier Transformation (FFT) of the time domain spectra in Fig. 2(a) are shown in Fig. 2(b). While some dips are observed in frequency spectra at 1.1, 3 and 4.7 *THz* at room temperature (Fig. 2(b)), the dips at 1.1 and 4.7 *THz* shift to higher frequency of 1.2 and 5.5 *THz* below 46 *K*. On the other hand, no shift is observed on the dip at 3 *THz* during temperature change. The shift of the dip from 1.1 to 1.2 *THz* with temperature decreasing is consistent with the previous study [10].

We measured the absorption of the DAST crystal at 2.0-4.5 *THz* using THz-TDS system equipped with the DAST crystal as an emitter. Here, the absorption spectra are calculated as $\alpha(\omega) = -\ln[E_{sam}(\omega)/E_{ref}(\omega)]/L$, where ω is the frequency, L is the thickness of the DAST crystal, $E_{sam}(\omega)$ is the FFT spectrum of the THz-pulse propagated through the crystal, and $E_{ref}(\omega)$ is the reference FFT spectrum. Fig. 3 shows the absorption spectra of the DAST crystal around 3.3 *THz* at 54 and 296 *K* when the incident direction of THz wave is parallel to *a* axis. Comparing the results of time domain spectra and absorption spectra, no characteristic change of the absorption with temperature change is observed (Fig. 3), which is consistent with the lack of a shift of the dip at 3

THz in Fig. 2(b). Hence, the absorption at 3 THz has no temperature dependence.

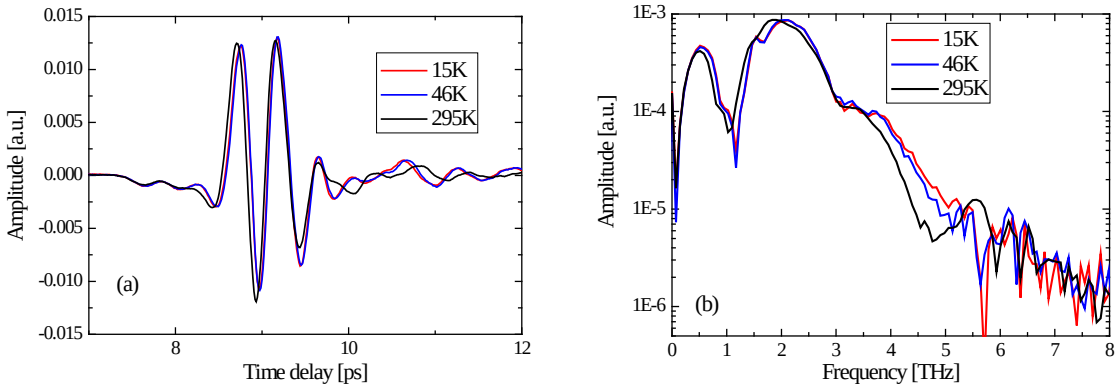

Fig. 2(a) THz generation from the DAST crystal in time domain and (b) frequency spectra obtained from the time-domain spectra.

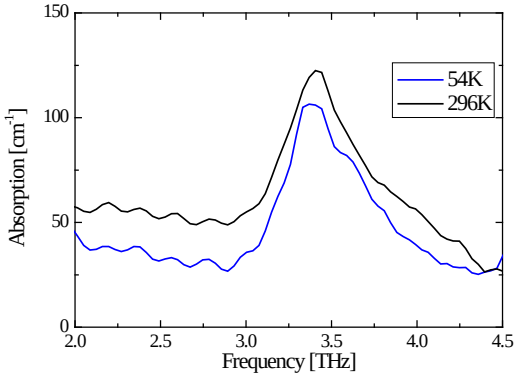

Fig. 3 An absorption of the DAST crystal around 3.3 *THz* measured by THz-TDS system using the DAST crystal as emitter.

The frequency region of the terahertz wave approximately corresponds to the low energy excitation, such as fundamental vibration modes and the thermal emission lines of molecules. Hence, the absorption around 3.3 *THz* in this study should be contributed by a molecular vibration mode. Here, it is reported that a vibration mode at 8.5 *THz* in the DAST crystal originates from the in-plain ring deformation vibration of the anion part [11]. The absorptions observed in this study should originate from vibration modes which have lower energy. Further, Walther et al has reported that the simple vibration of the anion–cation pair is the main cause of the absorption of the DAST crystal in the far infrared [10]. Hence, the shift of the absorption peaks observed at 1.1 and 4.5 *THz* with temperature decreasing might be caused by a change of a vibration and/or motion which contribute to the absorptions.

4. Conclusion

We have measured temperature dependence of THz generation and absorption of the DAST crystal using a THz-TDS system with cryostat. According to the results of time domain spectra,

THz generation from the DAST crystal has some dips at 1.1, 3 and 4.7 *THz* at room temperature. The dip at 3 *THz* is consistent with the absorption spectra of the DAST crystal, hence the dips are contributed from absorption of the DAST crystal. With a decreasing temeprature, the dips at 1.1 and 4.7 *THz* shift to 1.2 and 5.5 *THz* below 46 *K*, while the dip at 3 *THz* shows no temperature dependence.

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