Use of the organic crystal DAST for terahertz beam applications

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Received January 3, 2000

We report the use of the organic crystal DAST as a freely propagating electromagnetic-wave sensor at terahertz (THz) frequency. We also report the result of using a DAST crystal as a mid-IR THz emitter. Compared with the optical rectification from our best ZnTe emitter, that obtained by use of DAST demonstrates a sixfold enhancement of radiation at 7–20 THz. © 2000 Optical Society of America

OCIS codes: 160.4890, 190.4710, 230.2090, 300.6270, 320.7150, 260.1440.

The organic ionic salt crystal 4-dimethylamino-*N*-methylstilbazolium tosylate (DAST) is a promising material with very high nonlinear optical and electro-optical (EO) coefficients. The second-order nonlinear coefficient of DAST is 840 pm/V at 1542 nm. Its Pockels coefficient of 75 pm/V at 820 nm[1] is almost two orders higher than that of ZnTe, an EO crystal widely used for terahertz (THz) spectroscopy sampling. DAST has been used to generate broadband THz radiation by optical rectification[2] and narrow-band THz radiation by difference-frequency mixing with a dual-wavelength laser.[3] Moreover, the low dielectric constants of DAST allow high-speed EO modulation and detection. Intensity modulation of as much as 18 GHz was observed for a DAST waveguide.[4] DAST was also used to monitor a millimeter wave field by the interferometric method.[5] With the recent development of a free-space THz EO sampling technique,[6–8] it is of great interest to utilize the high Pockels coefficient of DAST as a far-IR sensor, even as a mid-IR sensor or emitter.[9]

In this Letter we report the implementation of DAST as an ultrafast THz wave sensor for what is believed to be the first time, employing a new setup that completely compensates for the intrinsic birefringence of DAST. Furthermore, DAST is demonstrated as a mid-IR THz emitter, showing a sixfold increase at 7–20 THz compared with our best ZnTe emitter.

Phase matching between optical and THz pulses is a major factor determining the efficiency and the bandwidth of a particular EO crystal and hence the optimum thickness of the crystal for specific applications. The coherent length,[8] which is decided by the phase-matching condition, provides a very important reference for selecting EO crystals and their thicknesses. Calculation of coherent length requires a complete knowledge of dielectric constants at THz frequency. The dielectric constant of DAST has been characterized by THz transmission spectroscopy.[2,10] However, owing to the strong absorption above 1 THz, it is difficult to obtain accurate data on that region. Hence, one can employ THz reflection spectroscopy[11–14] to obtain the complex dielectric constant of DAST. One and two resonant structures (phonons) are observed at the a and b axes, respectively. Fitting the data with an oscillator model yields a complete characterization of dielectric constants.[15] Based on the knowledge of the constants of DAST in both the optical and the THz regions, we calculate the coherence length, assuming an optical wavelength of 800 nm and the same polarization of both the THz and the optical beams in either the a or the b axis. The result is shown in Fig. 1, indicating that a DAST crystal with a thickness of a few hundred micrometers is suitable for EO sampling up to a few THz.

DAST has very strong birefringence, with group refractive indices of $n_{g,a} = 2.70$ and $n_{g,b} = 1.89$ at a wavelength of 800 nm.[1] After transmission of an optical pulse through a c-cut DAST crystal with a thickness of 0.7 mm, the field components of the optical pulse at the a and b axes of the DAST crystal will have a phase difference of $713 \times 2\pi$, equivalent to 1.9 ps. Considering the pulse duration of 100 fs or less that was used in the experiment, optical pulses along the a or the b axis will lose coherence after transmission through a 0.7-mm DAST crystal. The birefringence cannot be compensated for by a compensator, which usually has

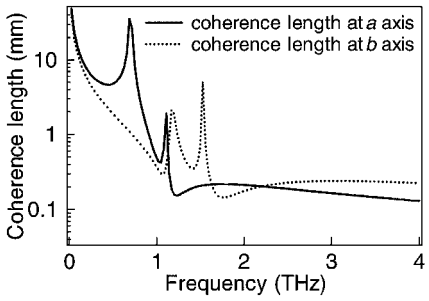

Fig. 1. Coherence length of the DAST crystal when both a THz pulse and an optical pulse have the same polarization in the a and b axes. The optical wavelength is assumed to be 800 nm.

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a retardation range from 0 to π. Therefore, this situation is very different from isotropic crystal (such as ZnTe) or weakly anisotropic crystal (such as $LiTaO_3$), of which the phase delay of the components along the different optical axes is either negligible or can be compensated for by a compensator. However, to monitor the small phase shift between the different optical axes of DAST induced by an applied THz pulse, it is necessary to compensate for the time shift between pulses at the a and b axes.

Figure 2 shows the experimental setup used to compensate for the birefringence. The optical probe beam goes through a quarter-wave plate after transmission through a c-cut 0.7-mm-thick DAST crystal. The beam is then reflected back by a mirror and goes through the same quarter-wave plate and the DAST crystal again. The quarter-wave plate is oriented so that s polarization (forward beam) becomes p polarization (backward beam) and vice versa. This polarization change can be verified by monitoring of the polarization of the probe beam after it transmits the beam splitter through a polarizer. Because each component goes through the same DAST twice with two orthogonal polarization states, each component has the same phase delay, and birefringence is exactly canceled. The temporal window between the forward and the backward optical probe beams is longer than the THz pulse duration, so the returned optical probe pulse in the DAST crystal will not be further modulated by the THz pulse. A Ti:sapphire laser oscillator with a 100-fs pulse width and 1.5-W average power is used in this experiment. A THz pulse generated by optical rectification in a 1-mm ZnTe is applied along the a axis of the DAST crystal and synchronized with the optical probe pulse when it passes through the DAST crystal. The probe beam is polarized at 45° from the a axis. After the intrinsic birefringence of the DAST is compensated for, it is biased by another quarter-wave plane, which yields the linear response. Assuming good phase matching, the balanced current is given by

$$\Delta I/I_0 = \Delta\Gamma = 0.5\pi E_{\mathrm{THz}}({n_a}^3 r_{11} - {n_b}^3 r_{21})L/\lambda\,, \quad (1)$$

where r_{ij} are the corresponding components of the EO tensor, L is the effective crystal thickness, and λ is the optical wavelength.

Figure 3 shows the THz waveforms and (inset) spectra measured with the DAST sensor and with a 0.5-mm ZnTe sensor. The amplitudes, waveforms, and spectra are all comparable. Since ZnTe is known to be an ideal THz sensor at less than 4 THz, we conclude that DAST is indeed a good EO sensor up to the frequency detected. The difference between the measured THz waveforms and spectra comes from the different spectral response functions of two sensors.[16] From the known EO coefficients, the THz amplitude from DAST should be several times larger than that of ZnTe. We attribute the deviation from theory to imperfections in the optical quality of DAST, such as nonflatness, inhomogeneity, misorientation, and imperfect surface conditions. These imperfections make it difficult to cancel the birefringence exactly. Hence the bias point is not optimized. Moreover, strong absorption above 1 THz reduces the signal significantly. An intense laser beam can cause a photorefractive effect, or even melting of DAST. We also notice an apparent difference from sample to sample.

Nevertheless, this technique has some very attractive advantages. In addition to the total cancelation of intrinsic birefringence, local effects, such as thermal-induced birefringence in the crystal, can also be compensated for. Furthermore, the technique can be applied to any other EO crystal, such as $LiNbO_3$ and $LiTaO_3$, with intrinsic birefringence. Thus the technique provides a simple method for the THz application of EO materials with high Pockels coefficients and intrinsic birefringence.

In the development of THz spectroscopy, it has often been desirable to perform it at higher frequency. For example, phonon dynamics is an interesting phenomenon and occurs at ~10 THz for some polar semiconductors.[14] DAST has been demonstrated to be an excellent emitter at less than 3 THz.[2] Here a Ti:sapphire oscillator with a 15-fs pulse rate is used to generate high-frequency mid-IR radiation. The setup is similar to that reported in Ref. 9. Figure 4 illustrates a mid-IR pulse generated by optical rectification in a 100-μm DAST crystal, with an observable bandwidth up to 20 THz, showing a dramatic sixfold increase compared with that of a 30-μm ZnTe sample in the same conditions. Since the coherence length of ZnTe is less than 30 μm for the frequency region 7–20 THz,[9] a thicker ZnTe crystal will not increase the amplitude of the spectrum. Thus thickness is not the

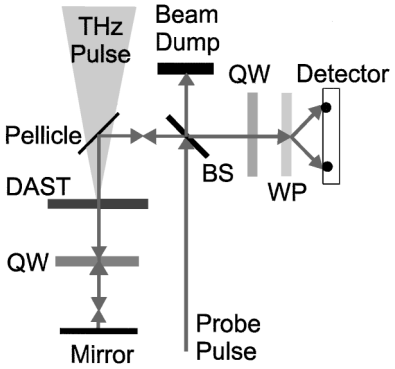

Fig. 2. Schematic of the experimental setup: QW, quarter-wave plate; WP, Wollaston prism; BS, 50–50 optical beam splitter.

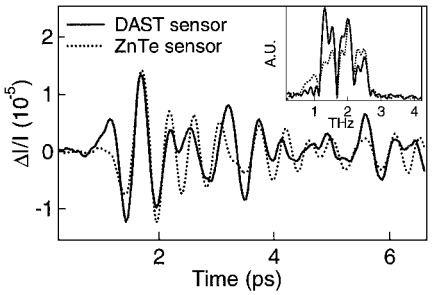

Fig. 3. THz waveforms obtained with DAST and ZnTe as electro-optic sensors. Inset, the corresponding spectra.

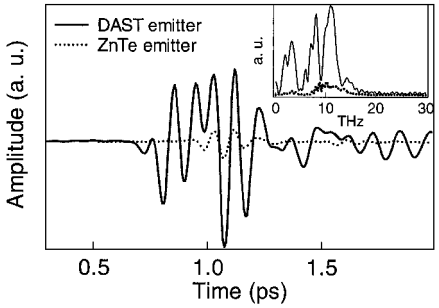

Fig. 4. Experimental mid-IR pulse from a 100-μm DAST emitter, showing a sixfold increase compared with a 30-μm ZnTe sample. Inset, pulse spectra.

cause of the enhancement after thickness reaches the coherence length.

In conclusion, the use of DAST as an ultrafast EO sensor at THz frequency has been demonstrated by means of a novel scheme. This technique extends the frequency response of DAST from the gigahertz to the THz range and opens the door for application of DAST in EO waveguide modulators, EO sensors, and other ultrafast EO devices in the THz-frequency region. Furthermore, the technique can be applied to other EO crystals with intrinsic birefringence. Finally, DAST showed dramatic enhancement of mid-IR THz emission at 7–20 THz.

This work was supported by the U.S. Army Research Office and the National Science Foundation. X.-C. Zhang's e-mail address is zhangxc@rpi.edu.

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