Verification of unevaluated nonlinear optical process of DAST crystal using the prism coupled Cherenkov phase matching method

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***Abstract*—The 4-N,N-dimethylamino-4'-N'-methyl stilbazolium tosylate (DAST) crystal has a high figure of merit among nonlinear optical crystals [1]. In second-order nonlinear optical (NLO) processes using a DAST crystal, the d_{111} process generally used is the process of generating *a*-axis polarized light from incident *a*-axis polarized light. However, there has been very little investigation of other effective nonlinear processes utilizing other second-order NLO processes. In this study, we investigated generation of terahertz (Thz) waves via the d_{133} process of the DAST crystal using the prism-coupled Cerenkov phase matching (PCC-PM) method [2,3]. THz waves were generated efficiently using the d_{133} process, and the magnitude of the nonlinear optical coefficient d_{133} estimated from the THz intensity ratio was 34.1 pm/V. These results indicate that PCC-PM can be used to examine unevaluated *d*-values of NLO crystals and estimate their values.**

I. INTRODUCTION

ONE way to generate THz waves is the optical rectification process using nonlinear optical crystals. The second-order nonlinear optical coefficients related to NLO crystal orientation determine the efficiency of the NLO process and serve as an indicator of its performance. Studies on second-order NLO processes of the DAST crystal have been widely conducted because they have larger nonlinear optical coefficients than other NLO crystals. For example, d_{111} (DAST) = 1,010 pm/V at $\lambda = 1{,}318$ nm, whereas d_{333} (inorganic lithium niobite) = 17 pm/V at $\lambda = 458$ nm [4].

For THz wave generation under collinear phase-matching conditions using the DAST crystal, a refractive index of 2.1 for near-infrared (NIR) light along the *a*-axis, and of 2.3 for THz wavelengths, provides a relatively long coherence length. However, the DAST crystal has an anisotropic refractive index, with a refractive index of 1.6 for NIR light along the *c*-axis, which results in a large refractive index difference in the THz range and an extremely short coherence length. The anisotropy of the DAST crystal is an important parameter in THz wave generation.

DAST crystals grown using a solution method are flat and long along the *a*-axis and *b*-axis, and short along the *c*-axis [5]. Because the DAST crystal is an organic material, processing is difficult and the crystal orientation for efficient THz wave generation is greatly affected by crystal habit [6]. Therefore, THz wave generation involving the *ab*-plane is widely used in practical applications. For the above reasons, few experimental reports have compared the various NLO constants for the DAST crystal.

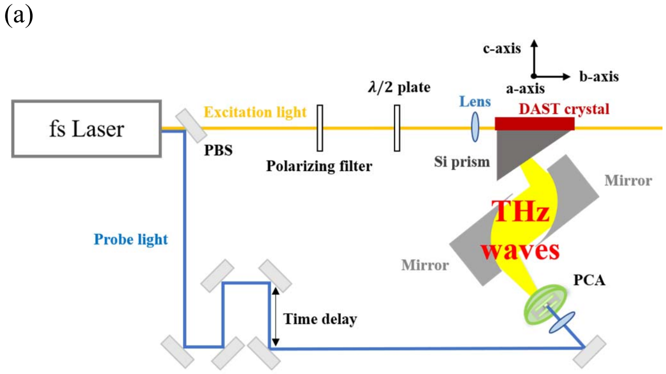
In this study, we validated a second-order NLO process along the *c*-axis of the DAST crystal using prism-coupled Cerenkov phase matching. In the PCC-PM method, THz waves are generated in the direction in which spherical waves overlap. In other words, since the travel direction of the excitation light and the travel direction of the THz wave are different, there is no restriction on the coherence length. Further, because the THz wave is extracted from the side surface of the crystal, the influence of absorption by the crystal can be reduced. Therefore, by using PCC-PM, the influence of phase mismatch and crystal absorption can be diminished, and THz waves can be generated by excitation light parallel to the *c*-axis.

DAST is a monoclinic crystal that belongs to the space group *Cc* and point group *m* [5,7]; thus, the theoretical quadratic NLO coefficients include d_{111}, d_{122}, d_{133}, d_{113}, d_{223} and d_{333}. In this study, we focused on d_{133}, which can be evaluated experimentally by simply rotating the polarization.

II. RESULTS

Figure 1a shows a schematic diagram of the setup of the THz-TDS system using PCC-PM. A femtosecond fiber laser (Femtolite HFX 400; IMRA America Inc., Ann Arbor, MI, USA) was used as the excitation light source (wavelength: 1,560 nm; maximal power: 242 mW; repetition frequency: 70 MHz; pulse width: 48 fs). In addition, a dipole-type PC antenna (Hamamatsu Photonics, Hamamatsu, Japan) was used to detect the THz wave. Prism-coupled Cherenkov phase-matching (PCC-PM) was used to extract THz waves generated the Cherenkov angle from the sides of the DAST crystal, via a silicon lens attached to the *ab*-plane of the crystal (Fig. 1b). The polarization of the excitation light was rotated in the plane formed by the *a*-axis and *c*-axis of the DAST crystal via a half-wavelength plate. The excitation light was linearly polarized, as confirmed by inserting a polarization filter (SPFN-30C-26; SIGMAKOKI, Tokyo, Japan) with a four-digit extinction ratio in front of the half-wavelength plate. Figure 1b shows a detailed view of the polarization direction of the excitation light and the polarization direction of the generated THz waves.

(a)



(b)

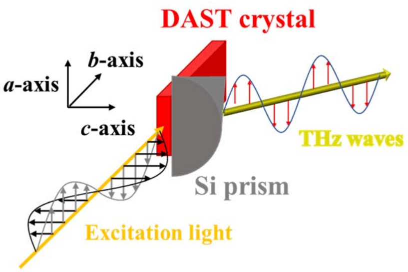
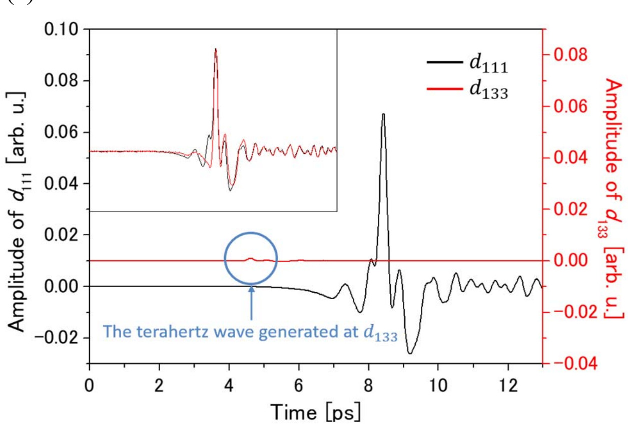


Fig. 1.(a) Schematic diagram of the experimental system for THz wave generation using prism-coupled Cherenkov phase matching. The polarization filter is used to align the excitation light into perfectly linearly polarized light. In addition, the polarization of the excitation light was adjusted by rotating the half-wave plate. (b) Detailed view of the polarization direction of the excitation light and the polarization direction of the generated THz wave.

Figure 2 shows the time waveform and frequency spectrum of the THz wave generated by the excitation light parallel to the a-axis and c-axis. In both cases, generation of THz waves was confirmed. To check the polarization direction of the generated THz wave, a wire grid was inserted into the propagation path of the THz wave. The results confirmed that the polarization direction of the THz wave generated by both processes was parallel to the a-axis of the DAST crystal. Therefore, it was confirmed that the THz wave was generated by the d_{111} process in the case of the a-axis polarization excitation and by the d_{133} process in the case of the c-axis polarization excitation. Figure 2a shows that the amplitude of the THz wave generated by the d_{111} process was 67.3 mV, whereas that of the d_{133} process was 0.93 mV. From these results, the amplitude of the THz wave generated in the d_{133} process was about 1/72.4 times that generated in the d_{111} process [0.93 = (1/72.4) 67.3]. Thus, the NLO coefficient d_{133} was estimated to be 34.1 pm/V based on the already known NLO coefficient d_{111} and the results of this experiment. In the frequency spectrum (Fig. 2b), there was no significant difference. The absorption line of THz waves at 1.1 THz is consistently observed, which is the intrinsic absorption of the DAST crystal.

(a)



(b)

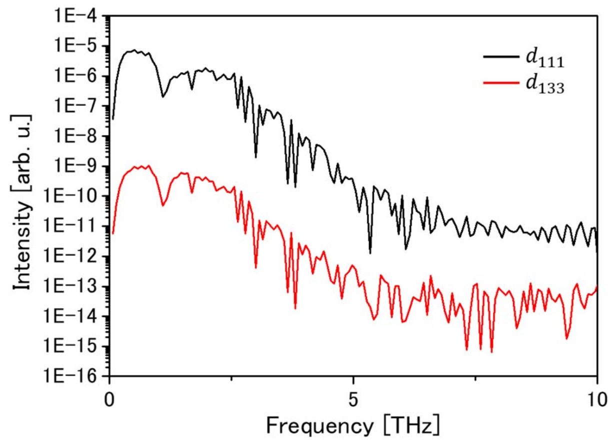


Fig. 2.(a) The THz timedomain waveform observed by the THz-TDS method; the black line corresponds to d_{111} , and the red line corresponds to d_{133}. A scaled up figure is also included for comparison. (b) Each frequency spectrum.

III. Summary

This experiment demonstrated PCC-PM-enabled THz wave generation using NLO processes that had not been attempted until now. The amplitude of the THz wave generated in the d_{133} process was about 1/72.4 times that generated in the d_{111} process. The NLO coefficients of d_{133} were estimated to be 34.1 pm/V, based on the relative NLO coefficients calculated from the value of d_{111}. In addition, PCC-PM provides a new method for measuring and examining unevaluated NLO coefficients.

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