J Infrared Milli Terahz Waves (2018) 39:509–513
https://doi.org/10.1007/s10762-018-0481-7

Single-Cycle Terahertz Pulse Generation from OH1 Crystal via Cherenkov Phase Matching

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Received: 19 January 2018 / Accepted: 6 March 2018 /
Published online: 16 March 2018

Abstract OH1 crystal is an organic nonlinear optical crystal with a large nonlinear optical constant. However, it has dispersion of refractive indices in the terahertz (THz) frequency. This limits the frequencies that satisfy the phase matching conditions for THz wave generation. In this study, we addressed the phase matching conditions for THz wave generation by combining an OH1 crystal with prism-coupled Cherenkov phase matching. We observed the generation of single-cycle THz pulses with a spectrum covering a frequency range of 3 THz. These results prove that combining prism-coupled Cherenkov phase matching with nonlinear optical crystals yields a THz wave generation method that is insusceptible to crystal dispersion.

Keywords Nonlinear optical crystal · OH1 crystal · Terahertz source · Cherenkov phase matching

1 Background

Terahertz (THz) waves are electromagnetic waves corresponding to frequencies from 100 GHz to 10 THz between radio waves and light waves. Several unique applications of these waves are currently under development [1, 2]. In particular, there are many applications based on THz time-domain spectroscopy (THz-TDS), such as high-resolution tomography [3], measurements of tablet-coating thicknesses [4], determination of chloride ion concentrations in concrete [5], and communications [6]. We must improve the performance of THz sources before we can use them to make advanced measurements.

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THz-TDS uses a femtosecond laser to generate and detect THz wave pulses [7]. The main mechanisms are either exciting carriers in photoconductive antennas (PCAs) grown on gallium arsenide (GaAs) substrates, or generating THz waves by optical rectification using nonlinear optical crystals. The latter method is based on the excitation of crystals with large nonlinear optical constants by high-output lasers. This enables us to efficiently generate THz waves with relative ease. In fact, there are many studies on THz sources with various nonlinear optical crystals [8–19]. The references [16–18] describe THz wave generation using the tilted pulse front method which is an alternative method of the Cherenkov phase matching. While the tilted pulse front method demands a crystal which is able to cut obliquely, the prism-coupled Cherenkov phase matching (PCC-PM) can select many crystals. Further, reference [19] describes a possibility of ring-shaped wave generation with suitable crystal and clad material. Hence, if appropriate, in a crystal and clad material, one can expect to generate further well THz waves from nonlinear optical crystals.

During THz wave generation using nonlinear optical crystals, complex-shaped THz waveforms arise due to refractive index dispersion and absorption in the crystal's THz frequency range. This range is unique to each crystal. Furthermore, in the frequency spectrum, if the refractive indices of the visible and THz frequencies differ significantly, or the crystal has high absorption in the THz frequency range, we will obtain a THz wave with a complex spectrum. These issues can be addressed by applying a THz wave generation method based on PCC-PM [13, 14]. This method enables us to determine the THz wave generation conditions (i.e. Cherenkov angle) by controlling the difference between the refractive indices of the nonlinear optical crystal and a silicon (Si) prism. In addition, the absorption of the waves by the crystal is regulated because the THz waves are generated close to the crystal surface. These advantages have enabled researchers to obtain single-cycle THz frequency pulses from 4-N,N-dimethylamino-4′N′-methyl-stilbazolium (DAST) crystal [15]. Furthermore, prism-coupled methods allow us to ignore the refractive index of the crystal in the THz frequency band. Thus, various nonlinear optical crystals can be used for THz wave generation. In this study, we used an OH1 crystal, which is a molecular organic crystal with a relatively large nonlinear optical constant (120 pm/V at 1.9 μm) [20, 21], and verified the feasibility of single-cycle THz pulse generation by PCC-PM. Normally, the refractive index of OH1 crystals is 2.13 in the visible light frequency range and about 2.30 in the THz frequency range [21]; thus, the Cherenkov angle, θ, becomes extremely shallow and is not suitable for Cherenkov phase matching. However, if the prism-coupled method is applied, the generation angle θ is determined by [11]:

$$\theta = \arccos\left(\frac{n_g}{n_{\text{clad}}}\right), \tag{1}$$

where n_{g} is the group refractive index of the pumping laser in the crystal, and n_{clad} is the refractive indices of the THz waves in the prism. Therefore, the generation angle is determined by the THz frequency refractive index of the prism material and the refractive index of the crystal at the applied excitation light wavelengths. Thus, the refractive index of the crystal in the THz frequency range can be ignored during wave generation. On the other hand, PCC-PM has problems to be achieved, such as collection of excitation light, surface roughness of the crystal, and design of waveguide structure from bulk crystal. In this paper, we used Si as the prism material, and the Cherenkov angle θ was estimated to be 49.8°.

2 Experiment

The OH1 crystal used in this study was grown by spontaneous nucleation. OH1 was dissolved in methanol, and then slowly cooled [22]. Figure 1 shows the microscope image of the OH1 crystal grown by the solution method and a structural formula of OH1 molecule.

The THz wave generation and detection discussed in this study are essentially based on the principle of THz-TDS. We used a femtosecond fibre laser (femtolite HFX400; IMRA Inc.) with a wavelength of 1.560 nm, maximum output of 242 mW, pulse rate of 70 MHz, and pulse width of 48 fs as the excitation light source. We used PCA (bow-tie type) to detect the THz waves, and a femtosecond pulse with a wavelength of 780 nm as the probe beam (see references [12, 14, 15] for details of the generation and detection of THz waves and data acquisition). Here, the OH1 crystal was not damaged under this excitation condition.

Figure 2a shows a cross-section of the OH1 crystal used for the wave generation along the plane of the incident excitation beam. The size of the OH1 crystal used in the presented experiment was $4.48 \times 2.41 \times 0.63$ mm^3 (thickness in the *a*-axis direction). We attached a Si prism to its *bc*-plane to extract the THz radiation from the crystal and filtered out the excitation beam by placing a polyethylene terephthalate (PET) film between the OH1 crystal and the Si prism. Figure 2b shows the setup of the OH1 crystal mounted in the optical system. We optimised the Si prism for THz radiation with the Cherenkov angle. According to Eq. (1), the THz wave generation angle was 49.8°. The crystal was assumed to refract the excitation beam with refractive index n_{opt} 2.20 [23]. Similarly, we assumed the refractive index of the high-resistance Si in the THz frequency n_{clad} to be 3.41. The excitation beam was focused on the *ac*-plane of the OH1 crystal using an objective lens. Prior to focusing, the diameter of the incident beam was 2.5 mm, and its Rayleigh length was assumed to be 13 mm. The excitation beam was linearly polarised, with its polarisation direction parallel to the *c*-axis. The radius of the Si prism base was 10 mm, and its length was 15 mm. The focal length of the objective lens was 4.5 mm. The arrows in the figure show the optical axes of the OH1 crystal.

3 Results

Figure 3a, b shows the time-domain waveform and frequency spectrum of the obtained THz wave. The THz wave generator used a prism-coupled OH1 crystal, and the THz wave detector used a bow-tie PCA.

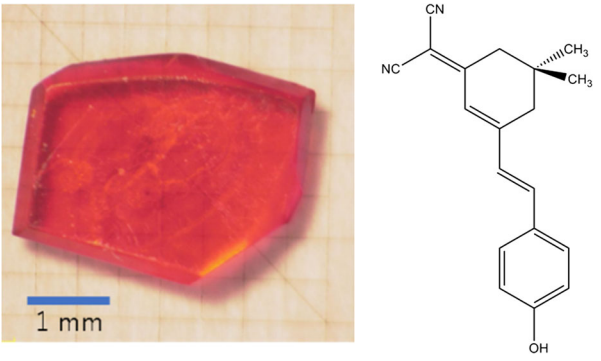

Fig. 1 The bulk OH1 single crystal we used (left). The structural formula of OH1 molecule (right)

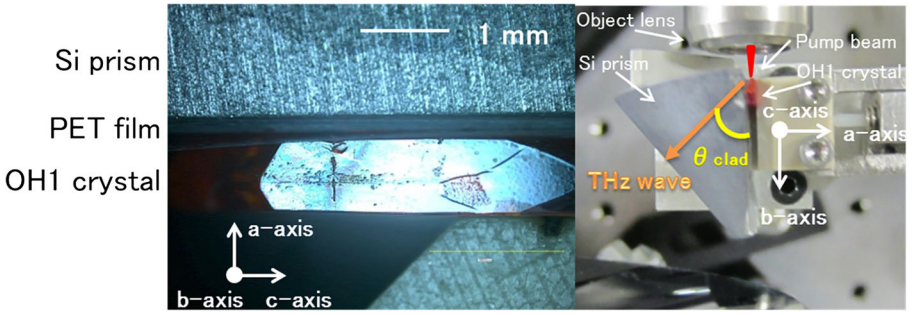

Fig. 2 **(Left)** Cross-section of the OH1 crystal along the plane of the incident excitation beam. **(Right)** Arrangement of the OH1 crystal, object lens, and silicon (Si) prism relative to the excitation beam

As shown in Fig. 3a, we obtained a single-cycle THz pulse wave with a pulse width of 560 fs. Generally, when THz wave is generated from a crystal in a collinear configuration, it often has a complex time-domain waveform due to the absorption caused by the crystal and the effect of multiple reflections inside the crystal. However, in this experiment, the observed time-domain waveform was comparable to a single-cycle waveform. These results are similar to previously reported results from studies that used PCC-PM with DAST [15], where the use of PCC-PM with an OH1 crystal allowed the generation of THz wave pulses that were close to single-cycle pulses. Figure 3b shows the frequency spectrum of the generated THz pulse. We confirmed that the generated pulse consisted of frequencies up to 3 THz. We also observed attenuation of the THz wave due to absorption at 1.47 THz along the *c*-axis of the OH1 crystal [24]. This indicates that as the OH1 crystal used in this study is a bulk crystal with an *a*-axis thickness of 0.63 mm, its focal point in the *ac*-plane (the distance between the focal point and the Si prism) and the beam diameter of the excitation light caused crystal absorption and thus attenuated the generated THz wave.

OH1 crystals have complex optical properties in the THz frequency range. For example, the refractive index is large at 0.4, 0.6, and 1.5 THz, and the absorption coefficient reaches its peak values at the same frequencies. There is large absorption when the frequency is beyond 2 THz [21]. This large absorption means that OH1 crystals are not suited to broadband generation. However, the method presented herein achieved a relatively broad THz wave over a 3-THz frequency range. This shows the advantage of using the Cherenkov phase matching method.

We were able to verify that THz-TDS with PCC-PM using the organic nonlinear optical crystal OH1 can generate short THz pulses that are useful for applications based on time-

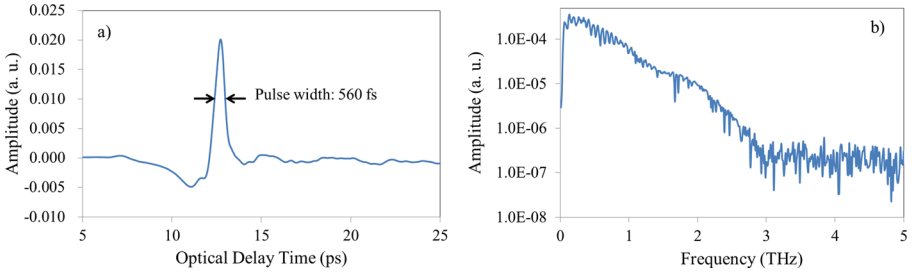

Fig. 3 **a** Time-domain waveform observed during terahertz wave generation obtained using the prism-coupled Cherenkov phase matching (PCC-PM) method using an OH1 crystal. **b** Frequency spectrum obtained by taking the Fourier transform of the time-domain waveform

domain waveforms (e.g. tomography). This result constitutes an experimental verification that the PCC-PM is a well THz wave generation method with various organic nonlinear optical crystals. In future work, we will prepare waveguides with restricted crystal thicknesses to cancel out the phase mismatches due to the finite diameter of the excitation beam, and optimise the excitation conditions (excitation wavelength, pulse width, power, etc.). This should enable us to create extremely short THz frequency pulsed light sources that are suitable for industrial applications.

4 Conclusions

In this study, we built a THz-TDS system based on the PCC-PM method that uses an OH1 crystal to generate a THz wave. We successfully generated short single-cycle THz pulses with a pulse width of 560 fs, and a frequency spectrum covering a range of 3 THz. This result confirmed that the PCC-PM with organic nonlinear optical crystals is a well THz wave generation method.

Acknowledgements We acknowledge the contributions of T. Minami, J. Seto, and H. Okano of Nagoya University for their experimental support and helpful discussion.

Funding Information This work was supported by the Japan Science and Technology Agency (JST) and JSPS KAKENHI (Grant No. 25220606).

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