Multi-Octave THz Wave Generation in PNPA Crystal at MHz Repetition Rates

L. A. Sterczewski,[1,+] J. Mnich,[1] and J. Sotor[1]
[1]*Faculty of Electronics, Photonics and Microsystems, Wroclaw University of Science and Technology, Wyb. Wyspianskiego 27, 50-370 Wroclaw, Poland*

Terahertz (THz) wave generation using organic nonlinear optical (NLO) crystals has received considerable attention in the past decades as a viable method to convert broadband near-infrared radiation to the far-infrared region. Nowadays, the rapid development of fiber laser oscillators at telecom wavelengths creates a demand for novel NLO crystals optimized for longer-wavelength excitation and lower pulse energy compared to conventional solutions optimized for millijoule-level pulses with kHz repetition rates at near-visible wavelengths. This requirement is partially addressed by well-established NLO materials like DAST, DSTMS, or OH-1. However, further improvements in terms of conversion efficiency are still desired. Very recently, PNPA ((E)-4-((4-nitrobenzylidene)amino)-*N*-phenylaniline) has been identified as a potential candidate to address this need. Spectra in the 0.2–5 THz range have been obtained using mJ-level 100 fs long pump pulses with kHz repetition rates, yet lower-energy MHz-rate excitation has not been tested to date. In this work, we demonstrate multi-octave THz generation from PNPA pumped by a simple all-fiber femtosecond laser providing 17 fs pulses at a 1550 nm wavelength with 50 MHz repetition rate (4 nJ pulse energy). Increasing the repetition rate 5×10^4 times accompanied by a 5-fold decrease in the pulse duration grants us access to the longwave infrared and THz region from 10 μm of wavelength (30 THz) to 300 μm (1 THz). Using the THz-induced lensing technique we optically sample the emitted field as shown in Fig. 1a. Without purging the measurement chamber, persistent oscillations of water molecules persist over tens of picoseconds, which appear as sharp absorption dips in the spectrum (Fig. 1b). We will discuss the time-frequency characteristics of the waveform and compare them with other well-established THz emitters like DSTMS.

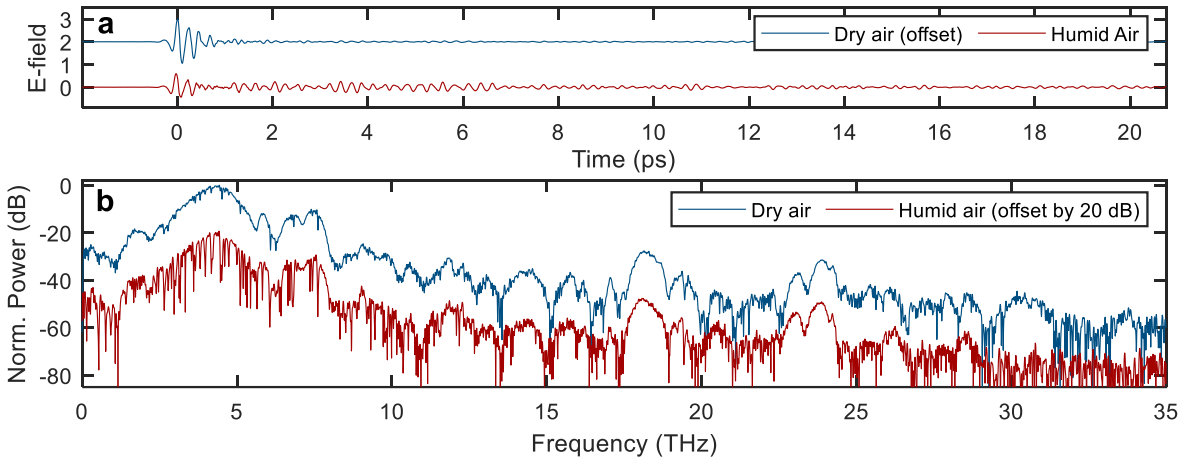

Fig. 1. Broadband THz generation from the PNPA crystal. (a) Sampled *E*-field. (b) Optical spectra obtained by Fourier-transforming the time-domain *E*-field waveforms.

[1] C. Rader et al., ACS Photonics **9**, 3720 (2022).

[+] Author for correspondence: lukasz.sterczewski@pwr.edu.pl; Funding from the European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No 101027721.

Supplementary Pages (Optional)

A time-frequency analysis (spectrogram) of the *E*-field emitted by the PNPA crystal is shown in Fig. S1. Interestingly, the emitted waveform is highly chirped – low frequency components are generated first. Next, the longwave infrared part (i.e. between 10–30 THz / 30–10 μm) appears in the emission spectrum, yet it persists for only ~2 picoseconds. This resembles earlier observations of organic nonlinear optical crystals, whose longwave infrared (LWIR) emission features correspond to a different generation mechanism than simple optical rectification [S1]. The high-frequency components are potentially attributable to molecular vibrations with picosecond-level dynamics that produce coherent infrared emission.

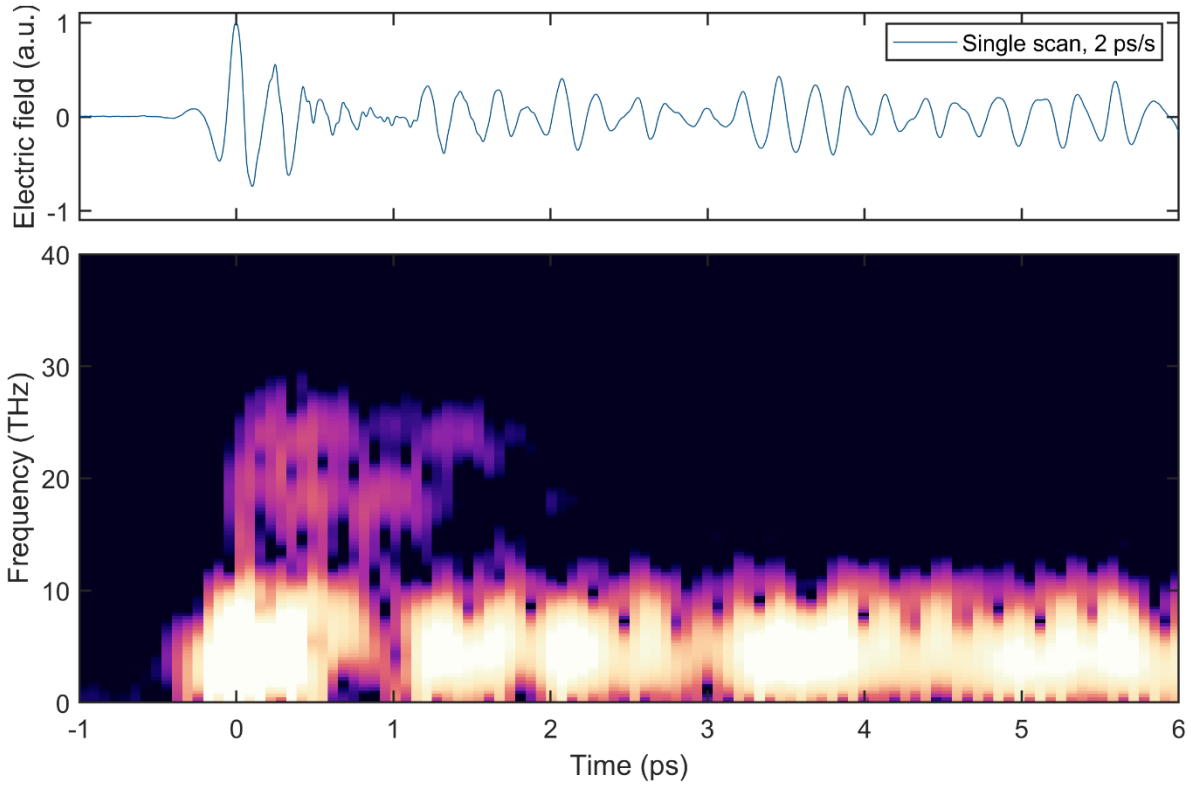

Fig. S1. Time-frequency analysis of the *E*-field waveform generated by the PNPA crystal pumped by a 17-fs 1550 nm laser.

Supplementary references

[S1] C. Somma, G. Folpini, J. Gupta, K. Reimann, M. Woerner, and T. Elsaesser, Ultra-Broadband Terahertz Pulses Generated in the Organic Crystal DSTMS, Opt. Lett. **40**, 3404 (2015).