High-Power, Ultra-Broadband THz Generation in Organic Crystal MNA

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***Abstract*—We present a THz-TDS operating with a high source average power of 4.4 mW at 400 kHz repetition rate, with a broad bandwidth of more than 10 THz, detected using the organic crystal 2-amino-5-nitrotoluene MNA as generation and detection crystals. The pump laser is an industrial Yb-based laser system, temporally compressed to a pulse duration of 35 fs. An optical-to-THz conversion efficiency of 0.08% is achieved.**

I. Introduction

Optical rectification (OR) in organic crystals is a well-established path to obtain intense and broadband THz radiation with high optical to THz conversion efficiency. Due to the intrinsically lower dispersion of the refractive index in organic nonlinear crystals, they are collinearly phase-matched in a much broader THz bandwidth compared to inorganic crystals such as gallium phosphide or zinc telluride [1]. 2-amino-5-nitrotoluene (MNA) is one among many organic crystals identified about 40 years ago as a powerful material for nonlinear applications due to its high molecular hyperpolarizability, favorable noncentrosymmetric crystal packing, and relatively large molecular number density. However, it has not been widely used to generate THz radiation due to the difficulties in synthesizing large enough crystal sizes. Very recently, B. Palmer et al. in [2] were able to grow large single crystals which are suitable for THz generation based on OR. Their studies confirmed that MNA is a perfect candidate for intense and broadband THz generation, as it does not require the additional synthetic steps required for synthesizing BNA as it was shown in [3]. In these results, a conversion efficiency of 3% was achieved using ~1.3 mJ/cm² pump laser at a central wavelength of 1250 nm and low repetition rate of 1 kHz.

One area of great current interest for organic crystals is to increase their average power using Yb-based high-power lasers, with several recent demonstrations showing great potential: for example in [4], BNA was pumped with a 540 kHz pump pulse which resulted in 5.6 mW of THz average power with a bandwidth of 7.5 THz. In [5], HMQ-TMS was pumped with a 10 MHz-repetition rate laser and the emitted THz radiation had an average power of 1.38 mW.

Here, we demonstrate a high average power and ultra-high-bandwidth THz source based on OR in MNA. It is driven by a commercial, Yb-based laser capable of delivering 18.5 W average power and externally compressed pulse duration of 35 fs, operating at 400 kHz repetition rate. Our THz source reaches 4.4 mW of THz average power, and our TDS achieves ultra-broad bandwidth extending more than 10 THz, with a conversion efficiency of 0.08%. To the best of our knowledge this is the first demonstration of average power scaling of an MNA crystal. The demonstrated system reaches impressive performance combining high average power, a bandwidth spanning >10 THz and high repetition rates.

II. Experimental setup and results

The full experimental setup is shown in Figure 1. The laser system is an industrial material processing ultrafast laser (TruMicro 2000, TRUMPF) providing 46 µJ pulse energy at the maximum repetition rate of 400 kHz. A home-built, Herriott-type multi-pass cell (MPC) compressor is used to reduce the pulse duration down to 35 fs with the power transmission efficiency of 94%. After the MPC, the laser beam splits in two parts: 99% of the total beam is used to generate THz radiation based on OR in MNA directly fused on a sapphire substrate. The crystal is mounted in such a way that the pump passes through the sapphire before reaching the MNA crystal. The reflection of the pump beam from the sapphire substrate is considered in the calculation of the THz conversion efficiency (loss of about 7.5%). The $1/e^2$ diameter of the laser beam on the position of MNA is 1.9 mm generated by a focusing lens with a focal length of 200 mm placed before the crystal. The generated THz radiation is collimated and refocused using two identical off-axis parabolic mirrors (OAP) with a diameter of 76.2 mm and a focal length of 152.4 mm. In order to reduce the thermal load on MNA, an optical chopper is placed before the crystal. The remaining 1% of the total laser beam is used to probe the THz radiation in an electro optic sampling (EOS) setup. A shaker with sinusoidal movement is used as a delay line to provide the delay between the THz and the probe pulses.

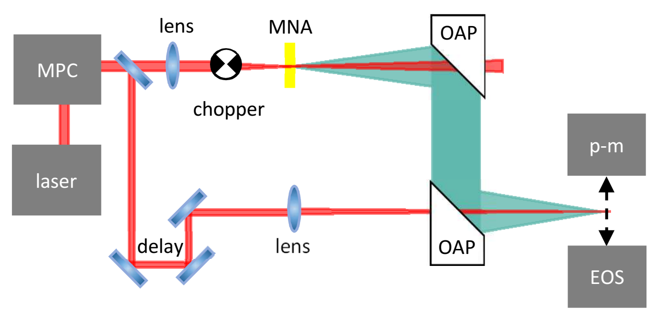

Figure 1. Experimental setup including pump laser, multi-pass cell (MPC) and THz-TDS. OAP: off-axis parabolic mirrors, p-m: power meter, EOS: electro optic sampling.

To suppress the residual laser radiation and parasitic second harmonic generation, two high density polyethylene (HDPE) disks with total thickness of 2.8 cm and THz transmission of 54% and polytetrafluorethylene (PTFE) tapes with THz transmission of 79% are used. The generated THz radiation is characterized using either a calibrated pyroelectric power meter (THz 20, SLT GmbH) or an EOS setup. A 0.1 mm gallium phosphide (GaP) or an MNA crystal without substrate are used to sample the generated THz trace using ~100 mW of the laser power in the EOS setup. The data is acquired using a lock-in amplifier (Zurich instruments, MFLI), which records the signal from the balanced photodetector in the EOS setup and the digitized position of the shaker. The modulation frequency of the pump beam, which is generated by the previously mentioned chopper, is used as a reference for the lock-in amplifier, and it is set to 2.7 kHz. A bandwidth of 250 Hz is chosen for the low-pass filter of the lock-in amplifier, and the frequency of the shaker is set to 0.5 Hz to sample the THz trace.

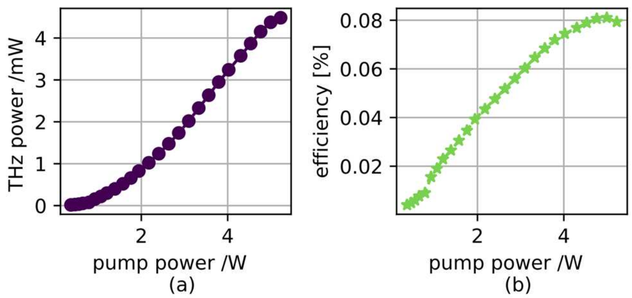

Figure 2. a) THz average power vs. pump power b) conversion efficiency vs. pump power. The loss from the sapphire substrate is considered in the efficiency calculation.

Figure 2.a) shows the THz power versus pump power on the crystal is measured by the THz power meter placed in the focus of the second OAP. The power meter is calibrated at a modulation frequency of 18 Hz at the German metrology institute (Physikalisch-Technische Bundesanstalt, PTB). Therefore, the frequency of the chopper before MNA is set to 18 Hz as well for the THz power measurement. The result shows a typical expected behavior for a second order OR process like in inorganic crystals such as GaP and lithium niobate [6]. At low pumping levels a quadratic dependence on the pump power is observed. At intermediate pump levels, the dependency becomes linear and at high pump levels the crystal shows a slight saturation. The saturation is most likely due to multi-photon absorption of the MNA at 1030 nm of pump wavelength, but the exact mechanism remains to be explored in more detail. The crystal is pumped up to 5.3 W, without any irreversible damage, which results in a maximum THz average power of 4.4 mW. The corresponding conversion efficiency is shown in Figure 2.b) which has the maximum value of about 0.08%.

In order to detect the THz electric field using the EOS setup, the THz power meter is replaced first with a thin GaP crystal. The purple curve in Figure 3.a) indicates the THz trace in time domain averaged over 150 traces and recorded in 150 s. The corresponding power spectrum on the logarithmic scale is obtained by Fourier transformation from the measured THz trace and is shown purple in Figure 3.b). The spectrum has a wide bandwidth which spans up to 7.6 THz with a dynamic range of about 60 dB, limited by the detection crystal response. To increase the detection bandwidth, the 0.1 mm GaP is substituted by a second MNA crystal without substrate. The THz trace is recorded with the previous parameters mentioned for GaP. The turquoise curve in Figure 3.a) shows the time trace. The corresponding spectrum is shown in turquoise in Figure 3.b). The detected bandwidth using MNA spans to more than 10 THz. The strong dip at ~6 THz comes in both cases from the transmission frequency response of the PTFE at this frequency [7].

III. Summary

In conclusion, we show a high average power, ultrabroadband, and high dynamic range THz-TDS. It is pumped with a 1030 nm laser at a 400 kHz repetition rate with a pulse duration of 35 fs. Using a driving power of 5.3 W, the maximum measured THz power is 4.4 mW, which is the highest THz power obtained using MNA to the best of our knowledge and is comparable to the state-of-the-art THz sources. Due to the good crystal quality and low roughness, we use MNA as detection crystal. The detected spectrum bandwidth using MNA is more than 10 THz. This source represents a unique tool for a variety of spectroscopy experiments, currently limited by dynamic range and bandwidth, and potentially for nonlinear THz spectroscopy. By optimizing the pump spot, reducing the repetition rate of the laser and operating in purged condition, we believe that this source has a high power-scaling potential.

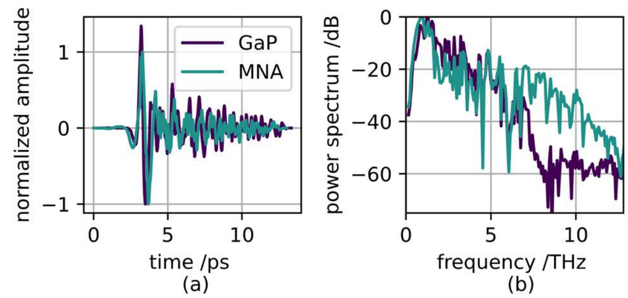

Figure 3. EOS trace: THz generation crystal is MNA. The detection crystal is GaP (purple) and MNA (turquoise). a) time domain. b) frequency domain.

References

[1] M. Jazbinsek, U. Puc, A. Abina, and A. Zidansek, "Organic Crystals for THz Photonics," *Appl. Sci.*, vol. 9, no. 5, Art. no. 5, Jan. 2019, doi: 10.3390/app9050882.

[2] B. W. H. Palmer *et al.*, "Large Crystal Growth and THz Generation Properties of 2-Amino-5-Nitrotoluene (MNA)," *ACS Appl. Electron. Mater.*, vol. 4, no. 9, pp. 4316–4321, Sep. 2022, doi: 10.1021/acsaelm.2c00592.

[3] I. C. Tangen *et al.*, "Comprehensive characterization of terahertz generation with the organic crystal BNA," *JOSA B*, vol. 38, no. 9, pp. 2780–2785, Sep. 2021, doi: 10.1364/JOSAB.420597.

[4] S. Mansourzadeh, T. Vogel, A. Omar, M. Shalaby, M. Cinchetti, and C. J. Saraceno, "Broadband, high power THz source at 540 kHz using organic crystal BNA," *APL Photonics*, vol. 8, no. 1, p. 011301, Jan. 2023, doi: 10.1063/5.0126367.

[5] T. O. Buchmann *et al.*, "High-power few-cycle THz generation at MHz repetition rates in an organic crystal," *APL Photonics*, vol. 5, no. 10, p. 106103, Oct. 2020, doi: 10.1063/5.0022762.

[6] M. C. Hoffmann, K.-L. Yeh, J. Hebling, and K. A. Nelson, "Efficient terahertz generation by optical rectification at 1035 nm," *Opt. Express*, vol. 15, no. 18, pp. 11706–11713, Sep. 2007, doi: 10.1364/OE.15.011706.

[7] F. D'Angelo, Z. Mics, M. Bonn, and D. Turchinovich, "Ultra-broadband THz time-domain spectroscopy of common polymers using THz air photonics," *Opt. Express*, vol. 22, no. 10, Art. no. 10, May 2014, doi: 10.1364/OE.22.012475.