# Efficient Broadband THz Generation from New Standards in Optical Rectification

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**Abstract:** We present the optical and THz properties of highly efficient organic nonlinear optical crystals PNPA and MNA. These crystals outperform industry standards in broadband THz generation through optical rectification of IR light pulses. 

## 1. Introduction

THz frequencies (0.1 to 10 THz) are used in a number of spectroscopic applications, most notably for direct excitation of phonon modes and other quasiparticles [1]. Optical rectification of infrared light pulses using nonlinear organic crystals is the most efficient means of generating string, single cycle THz pulses with broad bandwidths. Organic NLO crystals possess higher nonlinear optical coefficients ($\chi^2$) and exhibit stronger molecular hyperpolarizabilities and are therefore often preferred in comparison to their inorganic counterparts. Through data mining of crystallographic databases combined with 1st-principles calculations, our group has discovered and characterized a number of new organic THz generation crystals that exceed the performance of industry standards [2].

In this work, we report the optical and THz properties of NLO organic crystals PNPA and MNA and discuss how they pave the way for the future of THz spectroscopy applications [3-4].

## 2. Results

PNPA and MNA crystals were characterized for THz generation using the optical rectification and electro-optic sampling setup described in Ref. [5]. With this setup, we measured the field strengths in PNPA crystals of up to 2.3 MV/cm, which far outperforms measurements taken (using the exact same experimental setup) with commonly used crystals DAST and OH-1. **Figure 1** shows a comparison of the performance of these three crystals. The normalized spectra, plotted in **Figure 1b**, highlights the unique absorptive features of each crystal. From the frequency range of 0.5 to 3 THz, PNPA has few strong absorptions, in contrast with DAST and OH1, which have strong absorptions at 1 THz and 2 THz, respectively. This makes PNPA an excellent candidate for driving excitations at lower frequencies.

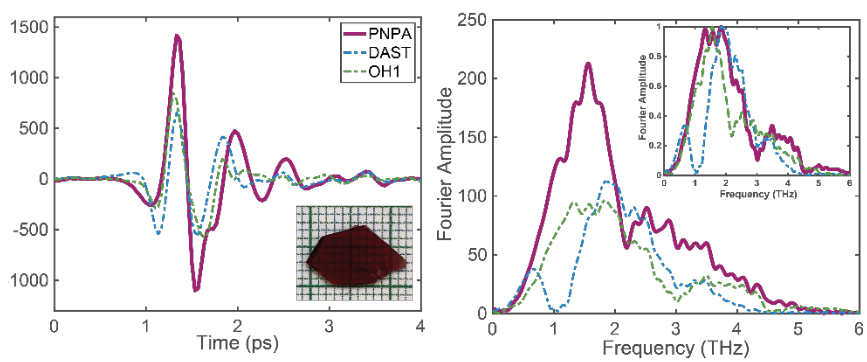


Fig. 1. (a) Electric field of PNPA, DAST, and OH1 measured at the same pump fluence and (b) the Fourier transform spectrum of each crystal. The inset of (a) is an image of PNPA. The inset of (b) shows the normalized spectra.

The pump wavelength of 800 nm is particularly interesting due to the commercial availability of Ti:sapphire lasers. Red NLO crystals such as DAST and OH-1 are not suitable for high-intensity THz generation at 800 nm due to poor phase matching, as well as lower damage thresholds at this pump wavelength due to significant two-photon absorption. Yellow crystals, however, have absorption cutoffs approaching 400 nm, which means there is less risk of two-photon absorption at the 800 nm pump wavelength. This raises the damage threshold of yellow crystals at this wavelength and enables research groups without optical parametric amplifiers (OPA) to perform THz spectroscopy. **Figure 2a** depicts the time traces of yellow crystals MNA, BNA, and inorganic GaP for reference pumped with the same fluence at 800 nm. The associated spectra are included in **figure 2c**. MNA meets the performance of its widely used derivative, BNA, at this technologically important wavelength. Additionally, MNA has a higher melting point range (128 – 129˚C versus 101˚C) and therefore a higher laser-induced damage threshold (LIDT) than BNA, allowing it to be pumped with higher laser fluences [5]. We propose that MNA would have excellent phase matching at a pump wavelength of 1030 nm, resulting in efficient THz generation at this wavelength as well.

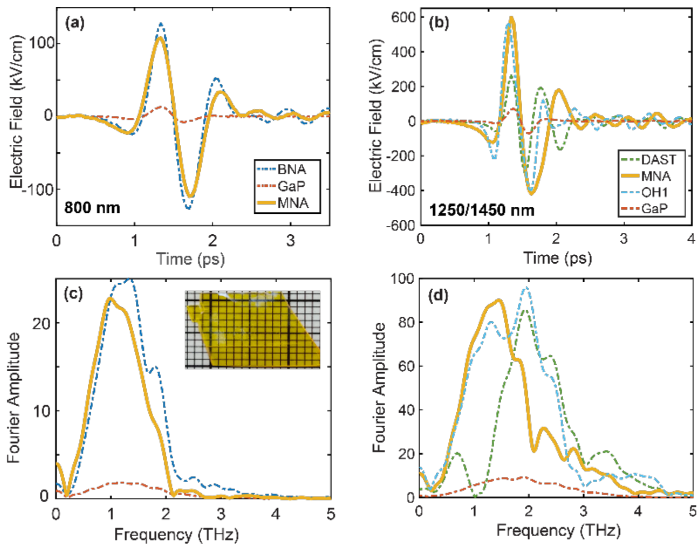


Fig. 2. (a) Time traces of MNA, BNA, and GaP with a pump wavelength of 800 nm. (c) Time traces of MNA at 1250 nm compared with DAST, GaP, and OH1 at 1450 nm. (b) and (d) show the spectrum of each of the traces in (a) and (c), respectively.

**Figure 2b** shows a comparison of the THz electric field produced by MNA with DAST and OH-1 at longer wavelengths obtained via the output of an OPA. MNA is the only known yellow crystal that rivals the performance of these organic THz generators at longer wavelengths. The spectrum of each is depicted in **figure 2d**.

## 3. Summary

We conclude that PNPA and MNA set new standards for THz generation through optical rectification with a large range of pump wavelengths. The THz field strengths produced by PNPA are higher than any other known NLO crystal, and MNA is well phase-matched at both 800-nm and longer pump wavelengths.

## 4. References

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