

# Broadband terahertz pulse generation driven by an ultrafast thin-disk laser oscillator

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**Abstract:** We demonstrate broadband THz generation driven by an ultrafast thin-disk laser (TDL) oscillator. By optical rectification of 50-fs pulses at 61 MHz repetition rate in a collinear geometry in crystalline GaP, THz radiation with a central frequency at around 3.4 THz and a spectrum extending from below 1 THz to nearly 7 THz are generated. We realized a spectroscopic characterization of a GaP crystal and a benchmark measurement of the water-vapor absorption spectrum in the THz range. Sub-50-GHz resolution is achieved within a 5 THz bandwidth. Our experiments show the potential of ultrafast TDL oscillators for driving MHz-repetition-rate broadband THz systems.

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## 1. Introduction

THz time-domain spectroscopy (THz-TDS) is a powerful tool to explore material properties and the dynamics of complex molecular systems through static and time-resolved investigations [1–5]. The employed THz source must fulfill a set of requirements including a spectral coverage in agreement with the studied system and a sufficient signal-to-noise ratio. Additionally, a compact system and a short acquisition time are often preferable. These criteria result in the demand for table-top high-power high-repetition-rate broadband THz sources.

Amongst other techniques, optical rectification of femtosecond pulses is a well-suited approach to produce high-power broadband THz radiation [6,7]. The development of THz sources based on optical rectification has been closely linked to the advances of sub-100-fs Ti:sapphire laser systems, which contributed to tremendous progress in this area. The efficiency of the optical rectification process is typically in the order of  $10^{-7}$  to  $10^{-2}$  [6,7]. Therefore, THz systems would benefit from power scalable laser technology. Recently developed diode-pumped ultrafast Yb-based lasers emitting at  $\sim 1 \mu\text{m}$  central wavelength demonstrated operation at average powers up to the kW level and high repetition rates with hundreds of femtosecond pulse duration [8–11]. In these lasers, detrimental thermal effects are significantly reduced due to alternative gain medium geometries (fibre, slab, thin disk) which allow for an efficient heat dissipation.

Most commonly, high-power ultrafast lasers rely on amplifier schemes based on complex multi-stage architecture (typically a seeding master oscillator, pulse stretching, multiple amplification stages and compression). In contrast, ultrafast thin-disk laser (TDL) oscillators offer a one-box solution for delivering nearly-ideal  $\text{sech}^2$ -shaped femtosecond pulses at MHz repetition rates, high average powers in diffraction-limited Gaussian beams. Nearly 300 W of average power have been achieved [12,13], but TDL oscillators typically operate at pulse durations longer than 100 fs [14]. However, sub-100-fs TDL oscillators based on broadband gain materials were recently demonstrated, albeit at moderate average power [15].

Despite appropriate laser parameters, Yb-based laser technology remains widely unexplored for THz generation [16]. Only few results have been reported attempting to use

Yb-based lasers to produce THz radiation. A high average power of 4 mW with a spectrum extending up to 1.2 THz has been demonstrated using tilted pulse front optical rectification in LiNbO<sub>3</sub> from 7 W average pump power and 1.3 ps pulse duration delivered by a 1 kHz repetition rate regenerative amplifier based on an Yb:YAG thin-disk crystal [17]. Compared to LiNbO<sub>3</sub>, broader spectra are achieved in semiconductors such as ZnTe and GaP due to a broad collinear phase matching in the near infrared [18–20]. GaP has a large rectification bandwidth up to 40 THz, although it possesses a transverse optical phonon resonance at 11 THz [19]. Despite the associated dispersion, collinear phase matching up to 8 THz is achievable using crystal thicknesses in the order of hundreds of microns [21]. In 2006, the first experiments of optical rectification in GaP conducted with an Yb-based ultrafast pump laser yielded 6.5  $\mu$ W THz average power from a 10-W 210-fs fiber amplifier operating at 120 MHz [22]. Later in 2013, 300  $\mu$ W THz average power has been obtained with frequency content up to 2 THz using an Yb-doped fibre amplifier delivering 21 W of average power in 52-fs pulses at a repetition rate of 42 MHz [23]. Broad spectra with frequency content up to 5 THz have been achieved both from a 3-W, 120-fs, 100-MHz Yb-doped fibre laser amplifier [24] and a 5-W, 20-fs, 78-MHz Yb-doped fibre laser amplifier [25].

Here, we report on MHz-repetition-rate broadband THz generation using the output of a mode-locked TDL. The 50-fs oscillator allows for a pulse-compression-free generation and detection of THz radiation up to 7 THz via optical rectification and electro-optic sampling (EOS) in crystalline GaP in a simple collinear geometry. Employing this THz source, we refine the Sellmeier coefficients of GaP in the THz range, enabling accurate phase matching calculations. Additionally, we benchmarked our system via THz spectroscopy of water vapor. Due to their power scalability and the high temporal resolution provided by sub-100-fs pump pulses, we believe that this result confirms the potential of ultrafast TDL oscillators to drive high-power broadband THz sources for static and time-resolved THz-TDS.

## 2. Broadband THz generation and detection

The experimental setup is depicted in Fig. 1. The driving laser source (described in [26]) is a Kerr lens mode-locked TDL oscillator based on the gain material Yb:Lu<sub>2</sub>O<sub>3</sub> [27]. It delivers a diffraction limited beam ( $M^2 = 1$ ) with 4 W of average power in 50-fs sech<sup>2</sup>-shaped pulses at 61 MHz repetition rate. This leads to >60 nJ pulse energy and >1 MW peak power available directly at the output of the laser oscillator. The optical spectrum is centered at around 1031 nm with a FWHM of 20 nm (corresponding to 5 THz). THz radiation is generated via optical rectification of the femtosecond near-infrared pulses in a <110>-cut GaP crystal. The pump pulses are pre-chirped by three dispersive mirrors accounting for  $-1500 \text{ fs}^2$  of group delay dispersion to compensate for the propagation through the focusing lens and crystal. The pump beam is focused into the crystal to a  $45 \mu\text{m } 1/e^2$  beam radius. Accounting for the 20% Fresnel reflection at the front interface of the uncoated GaP crystal, the peak intensity inside the crystal is estimated to be  $27 \text{ GW/cm}^2$  and the fluence  $1.7 \text{ mJ/cm}^2$ . These values remain below the damage threshold of the material which has been measured to be  $4.3 \text{ mJ/cm}^2$  (corresponding to  $60 \text{ GW/cm}^2$  peak power) at 1040 nm with 61 fs pulses [28]. We used a  $\sim 5$  mm diameter mirror to deflect the unconverted pump light. Due to the stronger divergence, only a small fraction of the THz radiation is reflected. An optical chopper running at 2.5 kHz modulates the pump beam for phase sensitive detection using a lock-in amplifier. The THz signal is then characterized via field-resolved detection using EOS in a second <110>-cut GaP [29,30]. The 50 fs gating probe pulses offer sufficient temporal resolution and do not require any additional temporal compression for a distortion-free broadband EOS measurement. The system is operated at room temperature ( $\sim 22 \text{ }^\circ\text{C}$ ) in a purged atmosphere with  $\sim 9\%$  relative humidity.

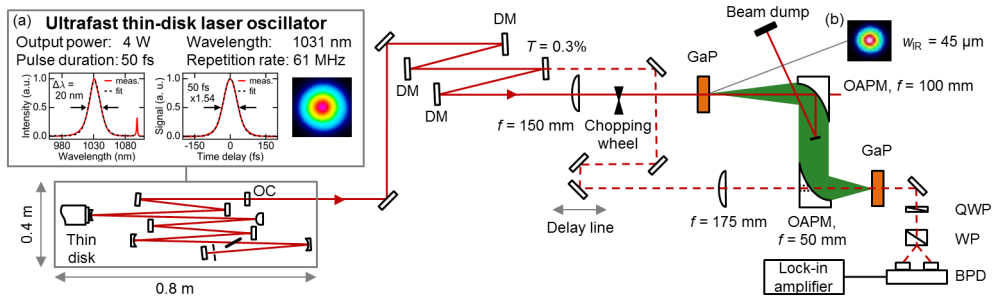


Fig. 1. Experimental setup for THz generation driven by the output of an ultrafast thin-disk laser oscillator. The inset (a) shows (from left to right) the optical spectrum, the autocorrelation trace and the output beam profile of the laser; the inset (b) shows the beam profile of the near-infrared laser at the focus. BPD: balanced photo-detector; DM: dispersive mirror; OAPM: off-axis parabolic mirror; OC: output coupler; QWP: quarter-wave plate; T: transmission; WP, Wollaston prism;  $w_{IR}$ : infrared beam radius.

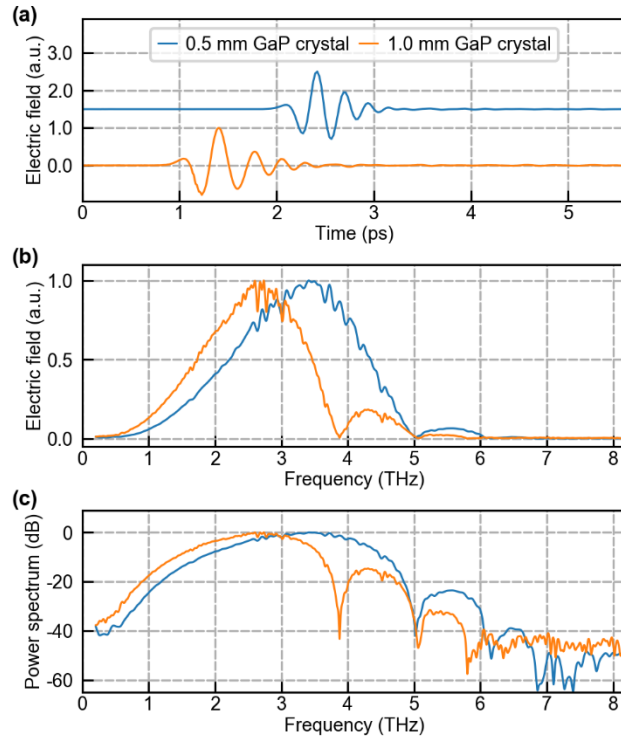


Fig. 2. (a) Time-resolved THz signals generated and detected in 0.5 mm (blue) and 1.0 mm (orange) thick GaP crystals (an offset in time and electric field has been added for better visibility) and (b) corresponding spectra of the electric field amplitude after Fourier transformation. (c) Corresponding power spectrum represented in logarithmic scale.

Quasi-single-cycle THz pulses were produced via optical rectification in 0.5 mm and 1.0 mm GaP crystals. The THz waveforms acquired in single scans with 30 ms and 10 ms integration constant, respectively, are shown in Fig. 2(a). They are detected via EOS using crystals with the same thicknesses as the rectification crystals. The corresponding THz spectra obtained by Fourier transformation of the waveforms have a central frequency at around 3.4 THz and 2.7 THz and extend up to nearly 6 THz and 7 THz, respectively [Fig. 2(b)]. The noise-like features in the spectrum are caused by residual water absorption. For both measurements, a dynamic range greater than 40 dB is achieved [Fig. 2(c)].

We evaluated the THz average power produced in the configuration with 1.0 mm GaP crystal using a calibrated pyroelectric photodetector (Ophir, RM9-THz) placed at the position of the detection crystal. The total path length from crystal to the detector is  $\sim 50$  cm. Two filters made of black paper and fabric are used to block residual pump light. At a relative humidity level of  $\sim 25\%$ , we measured an average power of  $0.2 \mu\text{W}$ . We calibrated the measurement by characterizing the filters spectral transmission using the EOS setup. They exhibit a 2% total transmission for our spectrum. Thus the estimated THz average power is in the order of  $10 \mu\text{W}$ , implying a conversion efficiency in the order of  $10^{-6}$ , which is in a reasonable agreement with results obtained with similar laser parameters [22,23,25].

### 3. Spectroscopic characterization of GaP

Prompted by the discrepancies among the published data [31,32], we performed an independent measurement of the refractive index of GaP in the THz region via THz-TDS [3,4]. For this, we inserted a 1.0 mm  $\langle 110 \rangle$ -cut GaP test-crystal into the collimated THz beam between two 0.5 mm GaP rectification and detection crystals. Comparing the spectral phase of this measurement to a reference measurement without the test-crystal allows to extract the refractive index of GaP [4]. Our data shown in Fig. 3 are consistent with [31] but disagree with more recent work [32]. Using our data in the range 1-6 THz and the data from [33] in the near infrared, the refractive index  $n$  is fitted to the Sellmeier equation given by

$$n^2 = 1 + \frac{B_1 \lambda^2}{\lambda^2 - \lambda_1^2} + \frac{B_2 \lambda^2}{\lambda^2 - \lambda_2^2}, \quad (1)$$

with  $B_1 = 2.064$ ,  $\lambda_1 = 27.284 \mu\text{m}$ ,  $B_2 = 8.089$  and  $\lambda_2 = 0.2707 \mu\text{m}$  and  $\lambda$  the wavelength in  $\mu\text{m}$ .

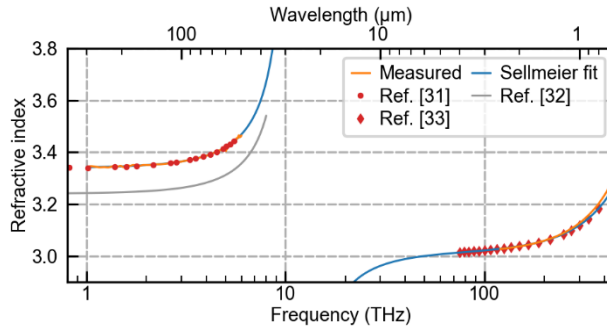


Fig. 3. Refractive index of GaP. In the THz region, it is retrieved from a THz time-domain spectroscopy measurement and compared to values taken from [31,32]. In the near-infrared range, it is measured by spectroscopic ellipsometry combined with transmission data and compared to [33].

The origin of the pronounced modulations in the THz spectra, e.g. at 5 THz, can be explained by phase matching. It accounts for the difference between the group velocity of GaP in the near-infrared and its phase velocity in the THz domain. The curves presented in Fig. 4 are calculated using the model introduced in [18] taking into consideration both generation and detection processes. In the calculation, we used a value of 3.31 for the optical group index of GaP at 1031 nm [33] and the refractive index in the THz range given by Eq. (1). Our calculated spectra are in a reasonable agreement with the measured ones in the range from 2 to 5 THz. In comparison, the refractive index given in [32] would yield a much broader phase matching for our system. The low-frequency behavior is not explained by this

simple calculation because it does not include the influence of the pump pulse shape and propagation effects. The discrepancies in the amplitude at frequencies above 5 THz are due to the linear absorption in GaP and vanishing nonlinear constant [32]. Based on phase matching considerations, we estimate an optimal crystal thickness to be 150  $\mu\text{m}$  for producing a gap-less spectrum spanning up to 7 THz using an Yb-based driving laser source.

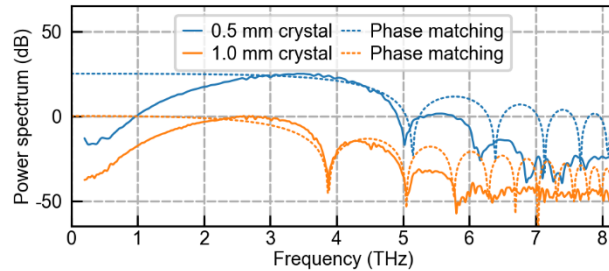


Fig. 4. THz spectra generated in 0.5 mm and 1.0 mm thick GaP crystals (solid lines). The corresponding phase matching curves (dashed lines) are calculated following the model presented in [18], using a value of 3.31 for the optical group index of GaP at 1031 nm and the refractive index in the THz domain given by (1).

#### 4. THz spectroscopy of water vapor

To confirm the suitability of the system for broadband THz-TDS, we performed a benchmarking spectroscopic measurement of water vapor absorption [2] [Fig. 5]. For this, we compared the THz spectra acquired at two humidity levels (23% and 9% relative humidity) in a setup using 1.0 mm GaP crystals for generation and detection. Each data set is acquired in a single scan with 10 ms integration constant. The water vapor absorption coefficient is given by the logarithm of the ratio of the two amplitude spectra as a function of the frequency. The resulting water vapor absorption spectrum is compared to the one reported in [34]. Water vapor lines are reliably detected with a sub-50-GHz resolution up to 5 THz. A better reliability in the 5-6 THz range could be obtained by averaging over multiple scans or by increasing the integration time constant. A finer spectral resolution is achievable by acquiring longer temporal scans including the echo pulses from the reflections inside the crystals, which would not affect the data analysis as they divide out in the frequency domain [2].

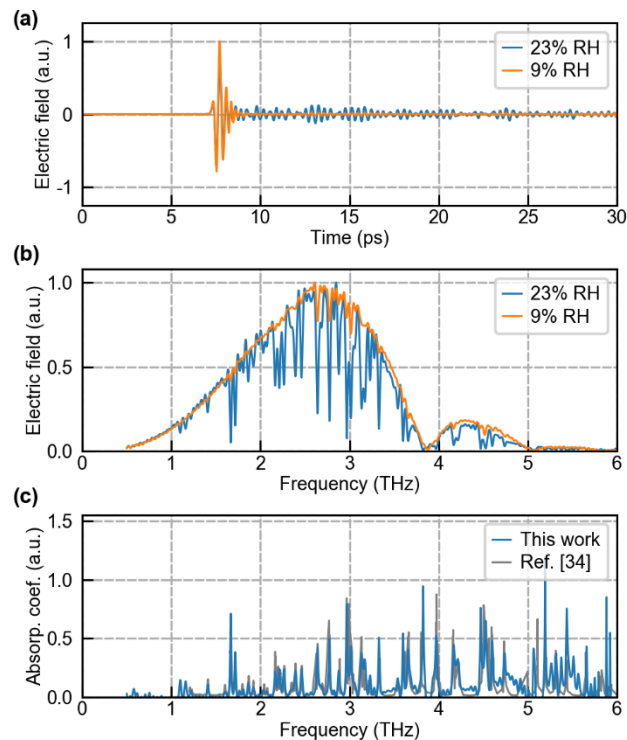


Fig. 5. (a) THz time-domain waveforms and (b) THz power spectra generated in a 1.0 mm GaP crystal at different relative humidity (RH) levels. (c) Frequency-dependent absorption coefficient of the water vapor, compared to the data taken from [34].

## 5. Conclusion and outlook

In conclusion, we have demonstrated broadband THz generation at MHz repetition rate using an ultrafast TDL oscillator as pump source. This simple single-stage laser source enabled the generation and detection of spectra with frequency content extending up to nearly 7 THz. We performed a spectroscopic measurement of GaP and refined its Sellmeier equation in the THz range. It is used for phase matching calculations, which are consistent with the acquired THz spectra. We conducted benchmarking linear THz-TDS experiment, measuring water vapor absorption spectrum with a sub-50-GHz resolution achieved in a frequency range between 0.5 and 5 THz.

We believe that higher THz frequencies are within reach by using thinner GaP crystals for improved phase matching or different types of emitters [35] such as ZnTe [20], nonlinear organic crystals [36], plasma [37] or metallic spintronic emitters [38]. On the other hand, higher THz average power should be achievable by using thicker crystals, albeit at the expense of a reduced bandwidth. A compromise fulfilling the demands of a particular experiment is certainly possible. Similar to the case of THz generation in LiNbO<sub>3</sub> [7], our preliminary calculations indicate that a pump pulse duration of 50 fs is not optimal for efficient THz generation with a targeted bandwidth of 7 THz. As a next step, we will investigate the influence of the pulse duration on the THz generation in GaP crystals. TDL oscillators delivering more than 10 W of average power with 90 fs pulses at 61 MHz repetition rate have been as well recently demonstrated [26]. We expect that such parameters would allow increasing the THz average power due to the availability of higher pump power and longer pulses offering an adequate spectral bandwidth for efficient conversion. Therefore, we believe that TDL oscillators are a promising technology for scaling up the average power

of broadband THz radiation. We expect that such compact sources of broadband THz pulses based on thin-disk laser oscillators will be beneficial for linear static THz-TDS and time-resolved THz spectroscopy.

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