

# Generation of 0.9-mJ THz pulses in DSTMS pumped by a Cr:Mg<sub>2</sub>SiO<sub>4</sub> laser

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We report on high-field terahertz transients with 0.9-mJ pulse energy produced in a 400 mm<sup>2</sup> partitioned organic crystal by optical rectification of a 30-mJ laser pulse centered at 1.25  $\mu$ m wavelength. The phase-locked single-cycle terahertz pulses cover the hard-to-access low-frequency range between 0.1 and 5 THz and carry peak fields of more than 42 MV/cm and 14 Tesla with the potential to reach over 80 MV/cm by choosing appropriate focusing optics. The scheme based on a Cr:Mg<sub>2</sub>SiO<sub>4</sub> laser offers a high conversion efficiency of 3% using uncooled organic crystal. The collimated pump laser configuration provides excellent terahertz focusing conditions. © 2014 Optical Society of America

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High fields up to 100 MV/cm, or 1 V/Å, in the low-frequency part of the THz spectrum (0.1–10 THz, LF-THz) are beneficial for many new applications, such as THz-enhanced attosecond pulse generation [1], the manipulation of electron beams [2], switching and controlling of magnetic domains [3,4], the initiation of surface catalytic reactions [5], free electron laser and high-harmonic generation-based pulse characterization [6], and the post-acceleration of laser-driven protons and ions. Assuming a diffraction limit focus and a single-cycle transient for constant pulse energy, the THz field strength  $E$  scales as  $E^2 = 1/f^3$ . Higher frequencies give rise to higher field because they are focused more tightly and turn in shorter single cycle pulse. Due to this physical law the production of high field strength in the low-frequency part of the THz spectrum requires pulse millijoule energies in a diffraction-limited spot size, while only a few  $\mu$ J are needed to realize equivalent field amplitude in the mid-infrared part (20–100 THz) of the spectrum [7]. Up to now, the production of low-frequency THz pulses at mJ pulse energy for laser-based sources were hampered due to hurdles in pump laser technology, the production of large size nonlinear crystals and the realization of velocity matching between the THz beam and the pump pulses.

In the LF-THz range, only a few techniques have shown potentials for high-energy THz generation. Among them, optical rectification (OR) of femtosecond laser pulses with a tilted pulse front has shown to be efficient for high-energy pulse generation both with uncooled [8,9] and cryocooled LiNbO<sub>3</sub> (LN) [10,11]. Terahertz pulse energies up to 175  $\mu$ J could be generated [12]. However, the radiation spectrum of LN is mostly confined to frequencies lower than 1.5 THz, and the realization of high field strength is challenging because of the unfavorable THz beam properties (elliptical beam profile, unequal divergence in  $x$  and  $y$ ) [13]. Laser-driven plasma sources, on the other hand, can deliver ultrabroadband THz pulses with up to a few  $\mu$ J pulse energy in the range of 1–100 THz, but

upsampling toward higher energies seems difficult due to plasma instabilities [14–16]. Finally, electron accelerator-based THz sources deliver pulse energies of more than 100  $\mu$ J and field strength of up to 20 MV/cm for high-frequency THz part (>10 THz) [17]. Unfortunately, the benefits of this source for the large user community are limited due to the restricted accessibility of the large-scale facilities. The low repetition rate and synchronization issues of the electron accelerator with respect to an external optical laser poses some challenges on time-resolved experiments.

In this report, we present a table-top and compact terahertz source-emitting phase-stable single-cycle transients between 0.1–5 THz with maximum fields larger than 42 MV/cm, and pulse energies as large as 900  $\mu$ J. The scheme is based on optical rectification in recently developed large-size partitioned crystal surface (PCS) DSTMS [18], which is combined with a novel type of drive laser. Up to 30 mJ energy pump beam is delivered by a multistage chirped-pulse amplification Cr:Mg<sub>2</sub>SiO<sub>4</sub> (Cr:forsterite) laser system. This source offers several benefits over the conventionally used Ti:sapphire-driven optical parametric amplifier (OPA) approach. The direct pumping reduces the overall complexity and provides one order of magnitude larger pulse energy as well as enhanced stability, reliability, and beam quality. Indeed, the transverse intensity profile from a high-energy OPA often suffers from hot-spots and inhomogeneities that limit the maximum fluence applicable to OR on organic crystals as surface damage could occur.

High-field THz sources based on organic crystals offer a simple, compact, and powerful scheme for time-resolved experiments [19–21]. A variety of small-size organic crystals have been investigated in the past in view of bandwidth and field strength, with the most prominent ones being DSTMS (0.15–5.5 THz, 1.5 MV/cm) [19], OH1 (0.1–3 THz, 0.5 MV/cm) [20], DAST (0.5–5 THz, 1 MV/cm) [21], and TMS (1–10 THz) [22,23].

In the past, there have been two hurdles to upscale the THz pulse energy/field strength of organic-based sources pumped with a SWIR laser system. The first difficulty was the growth and polishing of several  $\text{cm}^2$  size organic crystals. The second challenge was the availability of a pump source with sufficient energy to efficiently pump those large crystals at wavelength between 1.2 and 1.5  $\mu\text{m}$ .

To overcome the restriction in crystal size, we have recently proposed and realized 4  $\text{cm}^2$ -area organic crystal assembly by arranging a number of small-size crystals on a transparent host substrate. With this mosaic-like pattern, we demonstrated THz pulse energy and field reached up to 40  $\mu\text{J}$  and 5 MV/cm, respectively [18]. The generation of higher THz pulse energy was still hampered by the pump technology consisting of a Ti:sapphire-driven OPA operated at  $\lambda = 1.5 \mu\text{m}$  [21,24] and delivering  $\approx 2$  mJ. The upscaling of the OPA output is difficult and is associated with severe beam distortions. Therefore, efforts have been undertaken to explore conventional laser sources (e.g., Ti:sapphire emitting at a wavelength of 0.8  $\mu\text{m}$ ) to drive OR in organic crystals. However, the low nonlinearity at this pump wavelength limits the optical to THz energy conversion efficiency to  $6 \times 10^{-5}$  [25].

The Cr:forsterite CPA amplifier system investigated here overcomes above mentioned shortcomings since it can provide more than 30 mJ pulse, almost an order of magnitude higher energy with respect to present OPA systems. The main advantage of Cr:forsterite laser is that it delivers femtosecond pulses at 1250 nm. This wavelength is suited for optical rectification in the organic crystal DSTMS as it offers good phase-matching across a large THz frequency range paired to low absorption. Furthermore, with respect to the OPA, the laser offers a more homogeneous intensity beam profile that is beneficial for efficient pumping the organic crystals.

The simple experimental setup is shown in Fig. 1. The Cr:Mg<sub>2</sub>SiO<sub>4</sub> laser system consists of an oscillator running at 90 MHz which, after stretching, is injected into a regenerative ring amplifier. Several consecutive multipass amplifier stages running at 10 Hz and pumped with a total energy of 650 mJ from Nd:YAG lasers (1064 nm) boost the pulse energy up to 55 mJ with an energy stability of 2% rms [26]. After compression, the resulting 95-fs, 30-mJ pulses are collimated and directed to the organic mosaic crystal DSTMS (4-N,N-dimethylamino-4'-N'-methyl-stilbazolium 2,4,6-trimethylbenzenesulfonate). The laser pulse spectrum is centered at 1.25  $\mu\text{m}$  with a bandwidth of 20 nm FWHM. After the organic crystal, a low pass filter with cut-off frequency at 10 THz is used to reject the residual nIR pump. Calibrated THz attenuators are used to reduce the intensity for the beam characterization.

Shown in Fig. 2 are the spectrum and the interferometric autocorrelation of the terahertz pulse produced in DSTMS at a pump fluence of 10 mJ/cm<sup>2</sup>. The single-cycle pulse carries a spectrum that covers the full range between 0.1 and 7 THz and is peaked at around 2.8 THz. The measurements have been performed in ambient air. This causes some spectral modulation due to absorption in water vapor (humidity 40%), which results in a slight reduction of the integrated terahertz pulse energy at the position of the energy detector. The dip at 1 THz is due to a phonon absorption in DSTMS.

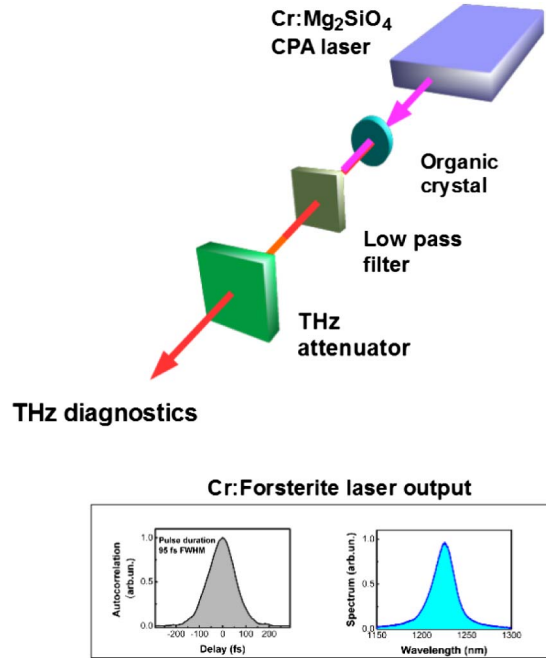


Fig. 1. Experimental setup: Cr:Mg<sub>2</sub>SiO<sub>4</sub> laser pumps a large-size partitioned DSTMS with 30 mJ, 95 fs FWHM pulse at 1250 nm central wavelength. The residual beam is rejected by low pass filters, and the THz intensity is reduced by THz attenuators.

DSTMS provides very large nonlinear optical susceptibility for optical rectification (212 pm/V) paired with low absorption for both the THz and the pump laser ( $\alpha < 1 \text{ cm}^{-1}$  between 900 and 1550 nm) [27]. The large PCS DSTMS used in our experiment can be pumped by the maximum pulse energy (30 mJ) available from the Cr:Mg<sub>2</sub>SiO<sub>4</sub> system. As described in Ref. [18], the PCS DSTMS consists of multiple parts of uncoated small DSTMS crystals of 0.5-mm thickness equally oriented and fixed on a host substrate transparent to the pump wavelength.

The advantage of organic crystals is their large NIR-to-THz conversion efficiency of a few percent. Shown in Fig. 3 are the measured THz pulse energy and conversion efficiency in dependence of the pump pulse energy. For these measurements, the pump diameter (full width at  $1/e^2$ ) has been fixed to 20 mm in order to illuminate

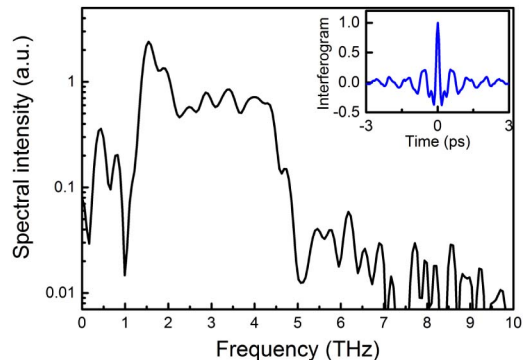


Fig. 2. (Black curve) Emitted terahertz spectrum retrieved by the interferometric autocorrelation (blue curve in the inset) using a THz Michelson interferometer.

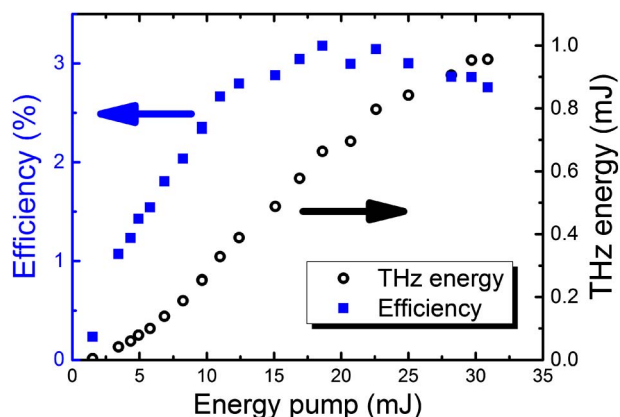


Fig. 3. THz energy (black circles) and conversion efficiency (blue squares) as function of the pump energy, for laser size covering the full organic crystal (2 cm diameter).

the entire PCS. An almost-linear increase in THz output energy is observed for an increasing pump energy, which reaches 900  $\mu\text{J}$  for 33-mJ nIR pulses (laser fluence of 10  $\text{mJ}/\text{cm}^2$ ). This corresponds to an emitted THz energy density of about 300  $\mu\text{J}/\text{cm}^2$ . For the energy measurements, a pyroelectric sensor (SPI-D, Spectrum Detector Inc.) and a calibrated Golay cell (Tydex Inc.) have been used. Both detectors provide similar results, within an error of less than 10%. As reported previously [18], the PCS DSTMS structure does not substantially worsen the laser to THz conversion efficiency. The maximum conversion efficiency of about 3% is measured at intermediate pump energy (20 mJ) before it drops to approximately 2.7% for higher pump intensities. The large conversion efficiency is reached without cooling the crystal and is due to cascaded optical rectification processes. The conversion efficiency is larger than previous results where the organic crystal was pumped by an OPA [18]. The  $\text{Cr:Mg}_2\text{SiO}_4$  pump provides a smoother beam profile that is beneficial for higher OR conversion efficiency. Moreover the longer  $\text{Cr:Mg}_2\text{SiO}_4$  pulse duration matches better than the OPA the OR spectral bandwidth of the PCS used here, which is limited to frequency below 7 THz.

Pumped by a collimated beam OR in the PCS, organic crystal yields THz radiation in a symmetric spatial intensity profile with excellent wavefront properties that result in high fields at focus. To measure the THz beam profile in the focus, an uncooled microbolometer THz imager (NEC Corporation, IRV-T0830, 23.5  $\mu\text{m}$  pixel pitch) has been used. The profile in the focus is symmetric and Gaussian-like. The THz field strength is determined from the measured THz pulse energy, the intensity spot size, and the pulse duration. As mentioned before, the higher frequencies are focused more tightly. Our diagnostics provides an average beam size integrated over all the spectral components. Due to the large- $f$  optics ( $f = 2.5$ ) focusing system available during the measurements, the THz focus was 260  $\mu\text{m}$  at FWHM. Being thus far from the diffraction-limited spot size, the pulse still shows very large field strength of 42 MV/cm and 14 Tesla, respectively, which is the largest ever reported terahertz field in the 0.1–5 THz frequency range. As recently shown, the collinear THz generation scheme

presented here provides radiation that can be focused to its diffraction-limited spot size using small  $f$ -number focusing optics and wavefront optimization [28]. Under these conditions, field strength of 80 MV/cm and 27 Tesla, respectively, is feasible, which surpasses any laser-based and accelerator based THz sources by about an order of magnitude in the frequency range of 0.1–5 THz. This opens new opportunities in nonlinear terahertz science.

In conclusion a record high-THz pulse energy of 0.9 mJ is presented that is the highest reported value for optical rectification in nonlinear crystals for 0.1–5 THz frequency range today. The single-cycle, phase-stable THz pulse shows excellent focusability properties that results in extremely large field strength of 42 MV/cm (experimentally measured) and 80 MV/cm (theoretical limit). The novel pump source presented here is based on a powerful  $\text{Cr:Mg}_2\text{SiO}_4$  laser that provides high pulse energy and an excellent beam profile for efficient terahertz generation in the partitioned large-size organic crystal DSTMS.

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