Laser driven generation of intense single-cycle THz field

Carlo Vicario^{1 a}, Clemens Ruchert^a, Fernando L. Ardana^{a,b}, Christoph P. Hauri^{* a, b}

^a Paul Scherrer Institute, 5232 Villigen PSI, Switzerland;

^b Ecole Polytechnique Federale de Lausanne, 1015 Lausanne, Switzerland.

ABSTRACT

We report on laser-based, high power single-cycle THz source. The THz radiation is generated by four-wave mixing in plasma and by optical rectification in organic salt crystal pumped by powerful optical parametric amplifier. The first approach permits the generation of electric field of hundreds of kV/cm at central frequency of 0.7 THz. The second technique allows the synthesis of an electric field exceeding 1 MV/cm paired with an unprecedented conversion efficiency of more than 2%, at frequency of 2 THz. The presented sources can be focused to a diffraction-limited spot and are suitable-versatile tool for time resolved THz experiment.

Keywords: Terahertz frequency high electric fields, picosecond THz pulse, organic crystals, plasma source.

1. INTRODUCTION

THz radiation located between the optical and the microwave frequency region (0.1-10 THz) is well suited to explore fundamental physical phenomena and to drive potential applications in biology, homeland security and medicine. Electric and magnetic field strengths exceeding on the order of a few MV/cm and Tesla respectively, are suited for exploring nonlinear phenomena [1, 2], to study ultrafast magnetization dynamics and to realize a novel type of THz-based streak camera [3] for characterizing sub-50 femtosecond x-ray pulses generated in free electron laser (FEL) [4]. Laser based sources of short THz pulse are also ideal for time resolved spectroscopy and pump and probe experiments. While laser-based THz radiation at frequencies 20-100 THz has been produced by difference frequency mixing in nonlinear inorganic crystals [5] the generation of high-power single-cycle field above 1 MV/cm in the so-called THz gap (0.1-10 THz), is still challenging. Only recently single-cycle pulses at 0.8 THz central frequency of 1 MV/cm field have been demonstrated by optical rectification in LiNbO₃ [6]. Nevertheless in this crystal a complicated non-collinear setup is required to overcome the phase velocity mismatch between the pump and the THz signals.

In this paper, we present a compact powerful single-cycle THz sources capable of delivering high-strength electromagnetic fields. We investigated two different techniques for the generation of such intense sub-mm wavelength pulses. The THz formation is realized in laser driven gas plasma or in nonlinear organic salt crystal. The two setups provide complementary characteristics and cover the frequency range between 0.1 and 5 THz. In the plasma source, the fundamental wavelength and the second harmonic of a femtosecond Ti:Sa laser are focused in gas [7]. At the focal location, the intense field initiates air plasma, which emits THz radiation. The second source is based on optical rectification (OR) in organic salt crystal (4-N,N-dimethylamino-4'-N'-methyl stilbazolium tosylate) known as DAST [8]. The crystal is pumped by a powerful optical parametric amplifier (OPA) driven by the same Ti:Sa laser running. The OPA allows infrared tunability in the range between 1.2 and 1.5 μm , which are the required wavelengths for performing efficient OR in DAST.

Intense single cycle THz radiation is generated by both the sources [9,10]. The output beam can be focus into a sub-mm spot size. The THz pulses have stabilized carrier envelope phase and it is possible to control their electric field polarity. The optical rectification in DAST permits higher energy per pulse and more intense field. The plasma source is suited to generate frequencies below 1 THz while DAST emission extends above 1 THz up to 5 THz.

*carlo.vicario@psi.ch ; phone +41 (0)56 310 3162 ; http://www.psi.ch/index_e.shtml

*christoph.hauri@psi.ch ; phone +41 (0)56 310 4197 ; http://www.psi.ch/index_e.shtml

In our setup, the temporal and spectral characteristics of the THz pulse are reconstructed by applying the well-known electro-optical sampling (EOS) technique in $\approx 100 \mu\text{m}$ GaP crystal. A weak 50 fs pulse acts as probe ($\lambda=810 \text{ nm}$) and originates from the Ti:Sapphire amplifier used to drive the THz sources. A calibrated Golay cell is used for absolute energy measurement and paired with a knife blade scanner to reconstruct the THz transverse distribution.

Pulses with spectral components above 1 THz suffer from a significant distortion and broadening when propagating in ambient air. For typical environment conditions in our laboratory (temperature=20 °C and relativity humidity of 40%), the water vapour absorption lines lead to a drop of the peak electric field by 30% over half meter propagation distance. Therefore transporting high-field THz pulses from the source to the target required beam lines which could be flooded with dry air or nitrogen to avoid re-absorption. For significantly reducing the distortions caused by THz absorption in water vapor, the EOS measurements were performed in dry air conditions (relative humidity <5%).

2. LASER PLASMA BASED THZ SOURCE

The formation of THz radiation in a plasma can be explained in the framework of the four-wave mixing process $(\omega_{2L} + \omega_{\text{THz}}) - \omega_L - \omega_L = \omega_{\text{THz}}$, with ω_L , ω_{2L} and ω_{THz} being the laser fundamental, its second harmonic and the resulting THz frequency, respectively. This third-order interaction is initiated by focusing an intense femtosecond laser pulse and its second-harmonic in a gas. A schematic of our setup is shown in Fig. 1a). As a laser system, we use a Ti:sapphire amplifier system delivering up to 20 mJ, 20 fs pulses at a repetition rate of 100 Hz. The pulses are focused in argon by a 90 degrees off-axis parabola ($f=100 \text{ mm}$). Before reaching the focus the signal is frequency-doubled in a beta-barium borate (BBO, 0.1 mm) nonlinear crystal. A 3-mm thick polytetrafluoroethylene filter is located after the crystal and acts as separator of the transmitted THz and the residual infrared radiation being scattered. The radiation is then focused in the GaP crystal for EOS reconstruction of the electric field. Typically, 7 mJ pulse energy is used to drive the plasma-based THz scheme. The reconstructed electric field and spectrum are shown in figure 1b) and 1c). The generated spectrum extends between 0.3 and 3 THz and it is peaked at 0.7 THz. We achieved absolute field strengths of up to 430 kV/cm at a central frequency of 0.7 THz.

Figure 1, THz generation in plasma. a) experimental setup used for the THz generation and detection in GaP crystal: off axis parabola (L), quarter wave plate (QWP), polarizer beam splitter (PBS) and balance photo-detector (BPD). b) temporal evolution of the THz electric field, and c) corresponding spectral intensity [9].

For several applications it is desirable to provide the appropriate THz field polarity at the experiment location. The control of polarity (figure 2) can be achieved in our setup by changing the time delay between the fundamental and second harmonic using a phase plate, for example. The polarity reverses for a delay corresponding to a half cycle of the second harmonic wave ($\approx 0.7 \text{ fs}$ for 400 nm) with respect to the fundamental wave.

Figure 2 Control of field polarization by introducing a delay between fundamental and second harmonic.

The THz radiation generated in our setup is free from high order aberrations and therefore can be easily focused into a diffraction limited spot size (figure 3). The transverse intensity, at the focus of 5 cm off-axis parabola, is reconstructed using a blade knife scanning technique and the Golay cell (upper plot). The horizontal intensity profile is well approximated by Gaussian distribution with sigma of 56 μm and FWHM of 131 μm .

Figure 3 Energy versus knife-blade position and reconstructed beam profile at the focus of an off-axis parabola ($f=5\text{cm}$).

The plasma-based approach has limited potentials for reaching higher field strength ($>>0.4\text{ MV/cm}$). Strong defocusing provoked by the free electrons in the plasma prevents the nonlinear mixing process from working at higher intensities in

the interaction zone. We observed saturation in THz yield for fundamental wavelength laser energies exceeding 7 mJ. Up-scaling the electric field strengths toward MV/cm and tuning the generated spectrum toward higher frequencies seems challenging with the presented scheme.

3. THZ GENERATED VIA OPTICAL RECTIFICATION IN DAST

To increase the peak electric field beyond the intensity provided by the laser plasma setup, we developed a powerful THz source based on optical rectification in DAST organic salt crystal. This technique permits single-cycle sub-ps THz pulses with electric fields strength above 1 MV/cm. The corresponding magnetic transient is larger than 0.3 Tesla. In our setup high pump-to-THz energy conversion efficiency is achieved. The source provides aberration-free THz beam, which allows for tight focusing.

Figure 4 THz generation in DAST organic crystal: (a) source and electro-optic detection; picture of the DAST (inset). QWP: quarter wave plate, WP: Wollaston prism, BPD: balance photodiode detector. (b) Pump laser autocorrelation trace, (c) 50 fs probe pulse centered at 800 nm used for the EOS[10].

In our experiment (figure 4), a 0.2 mm thick 5 mm diameter uncoated DAST is pumped by a 850 uJ pulse from an optical parametric amplifier. The pump pulse (figure 4(b)) has duration of 68 fs (FWHM) and spectral width larger than 100 nm. For THz generation the collimated infrared pump beam is propagated through the DAST crystal at normal incidence. For EOS a weak 50 fs pulse at 800 nm central wavelength acts as probe, figure 4(c).

Optical rectification in DAST is enabled by the broad spectrum of the infrared pump delivered by the OPA. Due to the OR process the generated THz electric field is intrinsically carrier-envelope phase (CEP) stabilized. In fact the phase fluctuation of the THz radiation are canceled since OR is equal to the frequency difference between two intra-pulse spectral components with the same phase offset. Compared to other crystals, the DAST provides very large nonlinear optical susceptibility for OR ($\chi_{111}^{(2)} = 480 \text{ pm/V}$ at $\lambda_p = 1.5 \mu\text{m}$). Low absorption and velocity matching between the THz and the pump laser is also relevant to achieve the highest THz field. For the DAST, the pump wavelengths for velocity matching are located between 1.2 and 1.5 μm .

Figure 5 THz intensity profile recorded at the focus of the recombining off axis parabola ($f=10$ cm).

THz radiation in DAST is emitted in the forward direction collinear with the pump and has the same linear polarization as the infrared pulse. The polarity of the output THz can therefore be controlled by simultaneous rotation of the input polarization and the DAST crystal. Our simple generation scheme allows for the formation of well-collimated THz radiation, which can be tightly focused. In figure 5 the THz intensity profile at the focus of the parabolic mirror with hole is shown. The THz distribution measured with a piroelectric camera (Spiricon PirocamIII) indicates a spot size of 0.6 mm FWHM. The asymmetric shape is attributed to the diffraction from the 5 mm aperture in the focusing optics.

Figure 6 (a) THz electric field and (b) spectral intensity generated when the DAST is driven by 1.5 μm wavelength and (c) (d) same plots for 1.2 μm .

Using the EOS technique, we measured the temporal evolution and spectral intensity of the THz radiation for different pump laser wavelengths. The results shown in figure 6 indicate that the temporal and spectral THz pulse characteristics (figure 6 (a), (b)) using a 1.5 μm pump source are almost identical to the results obtained with $\lambda_p=1.2 \mu\text{m}$ (figure 6 (c), (d)). For both pump wavelengths, the generated spectra cover the frequency range between 0.1 and 5 THz. The absorption lines at 1.1 and 3.1 THz are due to the two well-known phonon resonances in DAST. Similar THz characteristics in the temporal and spectral domain are also measured for other pump wavelengths (Fig. 7). Only small variations in the peak THz electric field (figure 7(a)) and output bandwidth (figure 7(b)) are observed across the full spectral range of the pump laser.

Figure 7 (a) Peak THz electric field and (b) spectral intensity generated for different pump wavelengths.

Pulse energies of up to 20 μJ have been measured with calibrated Golay cells. The value of the peak electric field is derived from the following formula:

$$E_{THz} = \sqrt{0.5 \varepsilon_{THz} / (c \varepsilon_0 \tau_{THz} \pi r_{THz}^2)} \quad (1)$$

with E_{THz} peak electric field of THz, c speed of light, ε_{THz} THz pulse energy (Golay cell measurement), r_{THz} spot size, τ_{THz} pulse duration provided by EOS, radius, ε_0 electric permittivity of free space. The maximum field strength of our THz source is calculated as 1.35 MV/cm at focus for central frequency of 2.1 THz and a bandwidth $\Delta\nu$ of 2 THz (FWHM). On the other hand, the field strength reconstructed by measuring the maximum modulation signal of the balanced photo-detector $(I_1 - I_2)/(I_1 + I_2) = 0.34 (\pm 3\%)$ on with signals I_1 and I_2 measured at the peak THz field indicates a somewhat lower field (936 kV/cm $\pm 10\%$). The field reconstruction is performed according to the formula:

$$E_{THz} = \frac{\lambda_0 \sin(\Delta I / I)}{2 \pi n_0^3 r_{41} t L} \quad (2)$$

with electro optical coefficient $r_{41} = 0.88 \text{ pm/V}$ at the probe wavelength $\lambda_0 = 800 \text{ nm}$ and crystal length $L \approx 100 \mu\text{m}$. The coefficient t takes into account the overall THz intensity transmission losses introduced by the mirrors, the low pass filter and the mirror with aperture in the beam transport. This discrepancy between the peak field calculated with the two formulas is attributed to the measured CEP offset $\neq 0$ (see Fig. 5), to pulse distortions and accumulated small uncertainties of filter transfer functions and of EOS crystal thickness.

A remarkable pump-to-THz energy conversion efficiency of 2.2% is achieved. Considering the central frequency of 2.1 THz and the pump wavelength of 1.5 μm the corresponding photon conversion efficiency is 235%. Higher fields could be realized by use of antireflection-coated DAST crystals, up-scaling the pump pulse energy and by implementing a full control on the CEP offset. These implementations are currently under way. Laser-induced damage on the DAST surface is observed for intensities beyond 220 GW/cm^2 .

4. CONCLUSION

Two powerful single-cycle phase-stable THz sources are presented. The plasma-based source provides high strength fields at frequencies between 0.3 and 3 THz. The radiation can be focused down to a diffraction limit spot size and the field sign can be easily reversed for polarity sensitive experiments. Unfortunately this approach has limited potential for reaching higher field strengths ($>>0.4$ MV/cm). On the other hand, optical rectification in DAST permits the realization of compact and more powerful sources with emitted fields exceeding 1 MV/cm and 0.3 T magnetic field strengths at 2.1 THz. The pulse energy of ≈ 20 uJ is ideally suited to drive extreme nonlinear phenomena in gases and solids. The results are the most intense single cycle THz generated by a laser-based table-top THz source. The THz generation in DAST has the potential to realize transients with even higher field strengths by proper CEP offset control and by scaling up both the pump energy and the crystal size. The plasma and DAST emitters are well suited for studying ultrafast dynamics and performing time resolved spectroscopy experiments in the frequency range between 0.3-5 THz. The results demonstrated the THz specifications needed for novel streak camera for characterizing sub-50 femtosecond x-ray pulses generated FEL.

This work has been supported by the Swiss National Foundation (PP00P2_128493/1 and 200021_122111/1).

REFERENCES

- [1] M.C. Hoffmann, J. Hebling, H.Y. Hwang, K.L. Yeh, K.A. Nelson, "Impact ionization in InSb probed by terahertz pump terahertz probe spectroscopy". *Phys. Rev. B* 79, 161201 (2009).
- [2] M. Tonouchi, "Cutting-edge terahertz technology" *Nature Photonics* 1, 97 (2007).
- [3] U. Fruehling, M. Wieland, M. Gensch, T. Gebert, B. Schuette, M. Krikunova, R. Kalms, F. Budzyn, O. Grimm, J. Rossbach, E. Ploenjes, M. Drescher, "Single-shot terahertz-field-driven X-ray streak camera", *Nature Photonics* 3, 523 (2009).
- [4] BD Patterson, R. Abela, H.H. Braun, U. Flehsig, R. Ganter, Y. Kim, E. Kirk, A. Oppelt, M. Pedrozzi, S. Reiche, L. Rivkin, Th. Schmidt, B. Schmitt, V.N. Strocov, S. Tsujino, A.F. Wrulich, "Coherent science at the SwissFEL x-ray laser", *New J. Phys.* 12, 035012 (2010).
- [5] F. Jungiger, A. Sell, O. Schubert, B. Mayer, D. Brida, M. Marangoni, G. Cerullo, A. Leitenstorfer, R. Huber, "Single-cycle multiterahertz transients with peak fields above 10 MV/cm", *Opt. Lett.* 35, 2645 (2010).
- [6] H. Hirori, A. Doi, F. Blanchard, K. Tanaka, "Single-cycle terahertz pulses with amplitudes exceeding 1 MV/cm generated by optical rectification in LiNbO_3 ", *Appl. Phys. Lett.* 98, 091106 (2011).
- [7] T. Bartel, P. Gall, K. Reimann, M. Woerner, T. Elsaesser, "Generation of single-cycle THz transients with high electric-field amplitudes", *Opt. Lett.* 30, 2805 (2005).
- [8] A. Schneider, M. Neis, M. Stillhart, B. Ruiz, R. Kahn, P. Gunter, "Generation of terahertz pulses through optical rectification in organic DAST crystals: theory and experiment", *JOSA B* 23, 1822 (2006).
- [9] C. Ruchert, F. Ardana, A. Trisorio, C. Vicario, and C. P. Hauri "Toward High-power Single-cycle THz Laser for Initiating High-field-sensitive Phenomena", *Chimia* 65, 320 (2011).
- [10] C. P. Hauri, C. Ruchert, F. Ardana and C. Vicario "Strong-field single-cycle THz pulse generated in organic crystal", *Appl. Phys. Lett.* 99, 161116 (2011).