HIGH-FIELD LASER-BASED TERAHERTZ SOURCE FOR SWISSFEL

C. Vicario, B. Monoszlai and C. Ruchert, Paul Scherrer Institute, 5232 Villigen PSI, Switzerland
C. P. Hauri, Ecole Polytechnique Federale de Lausanne, 1015 Lausanne, Switzerland and Paul Scherrer Institute, 5232 Villigen PSI, Switzerland

Abstract

We present efficient laser-driven THz generation by optical rectification in various organic materials yielding transient fields up to 150 MV/m, 0.5 Tesla and energy per pulse up to 45 μJ. The generated spectra extend over the entire THz gap (0.1-10 THz). Manipulation of the absolute phase by dispersion control is demonstrated for 5-octave spanning, single-cycle pulses. The presented source will be applied to the future SwissFEL as Xray photon temporal diagnostics and for pump-and-probe experiments.

INTRODUCTION

THz radiation located between the optical and the microwave frequency region known as terahertz gap (0.1-10 THz) is well suited to explore fundamental physical phenomena and to drive applications in condensed matter, chemistry, medicine and biology [1]. Few MV/cm THz electric transients and tesla magnetic fields open new opportunities to study ultrafast magnetization dynamics, collective effects and charged particle manipulations [2]. High-peak THz sources are required at X-ray free electron laser facilities, such as SwissFEL, for novel pump and probe experiments as well as for the temporal characterization of the Xray pulse on the femtosecond time scale [3]. The generation of few-cycle fields exceeding 1 MV/cm in the THz gap has remained challenging. To access high field in the full THz gap we recently developed a compact and powerful laser-driven THz source based on nonlinear organic crystals. The radiation is generated by optical rectification in organic salt material such as DAST, OH1 and DSTMS. The optical rectification in these nonlinear materials permits the realization of extremely intense and phase-stable THz transient [4-6]. These organic crystals provide in fact low THz absorption and the highest susceptibility for optical rectification. Moreover, velocity matching between laser pump and THz radiation is achieved in a collinear geometry for pump wavelengths between 1.35 and 1.5 μm. THz radiation is emitted collinearly to the pump with excellent focusing characteristics which is key feature to achieve the highest peak field. Spectra covering the full THz gap become accessible for femtosecond laser pump and sufficiently thin crystal supporting this bandwidth. The crystals can be anti-reflection coated for maximum optical rectification efficiency, and, as we show in the experiment, their damage threshold for femtosecond pulses is in excess of 160 GW/cm^2

THZ GENERATION AND CHARACTERIZATION

In the experimental setup a TW-class Ti:Sa at 100 Hz, producing 60 fs FWHM pulses is used to drive a white-light continuum optical parametric amplifier (OPA). The multi-mJ OPA delivers 70 fs transform-limited infrared pulses to pump the organic crystal for THz generation.

In order to prevent organic crystal damage while using the maximum available pump flux, large crystals with up to 10 mm aperture at a nominal thickness of 0.5 mm are utilized. The THz beam is emitted collinearly to the pump and is tightly focused for the realization of the highest field. The THz temporal shape is measured by electro-optical sampling (EOS). The EOS gives direct access to the THz the absolute electric field and the spectral content. The electro-optical spectral sensitivity for our electro-optical setup decreases above 5 THz. Higher frequencies are measured by means of Michelson interferometer equipped with THz sensitive detectors.

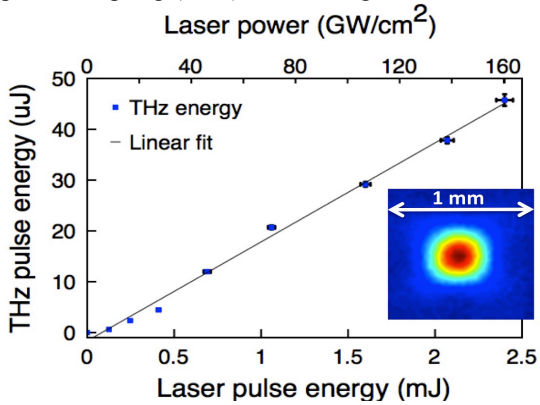
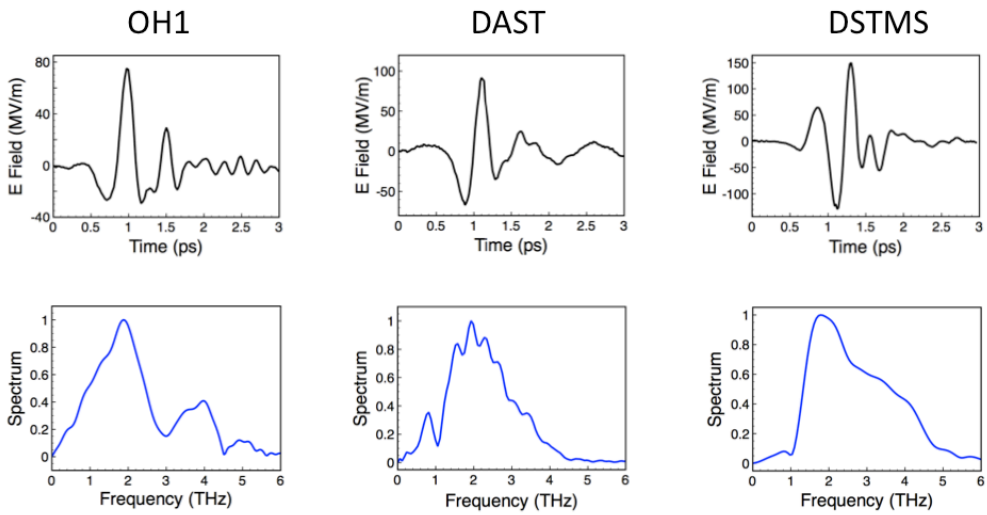


Figure 1: THz pulse energy as function of the pump fluence. Inset, 360 μm FWHM terahertz spot achieved at the waist.

Energy Conversion and Focal Spot

Absolute energy measurements are carried out by means of a calibrated Golay cell. The transverse beam profile is recorded with a bolometer un-cooled camera having a pixel size of 23.5 microns. In Figure 1 the THz pulse energy generated in a 0.5 mm thick DAST crystal with an aperture of 8 mm is shown as function of the infrared laser energy and power density. Maximum THz pulse energy of 45 μJ is reached when the crystal is pumped OPA pulse energy of 2.4 mJ (at 160 GW/cm^2). The highest pump-to-THz energy conversion is about 2% corresponding to photon conversion efficiency larger than 220%. Furthermore remarkable is the shot-to-shot THz

ISBN 978-3-95450-126-7



2: (Upper plots) Electric field shape and (lower curves) corresponding THz spectra generated in OH1, DAST and DSTMS.

energy variation, which is comparable to the OPA energy stability (1% rms). The quasi-linear dependence of the generated energy indicates that the upscale of the THz output by increasing the source energy is further feasible. It is worth noting that the maximum power density of 160 GW/cm^2 is not inducing damages in the organic crystal at 100 Hz. For other organic materials, the THz energy yield recorded in the same experimental conditions is approximately 1%.

In our setup, close to diffraction limit focusing is produced by parabolic mirror. The focus shape is circular and not affected by astigmatism or other visible aberrations, inset of1. The intensity is well fitted by a Gaussian with full width half maximum of 360 μm close to diffraction limit value (300 μm).

Time and Spectral Domain Reconstruction

Single-cycle intense THz field are generated in the organic crystals. The temporal shape of the THz pulse is retrieved through electro-optical sampling in 100 μm thick gallium-phosphide crystal.

The THz electric field produced in OH1, DAST and DSTMS are shown in Figure 2 (upper black curves) [7]. The spectral intensity, lower graphs in Figure 2, is calculated by Fourier transformation of the corresponding temporal evolution. The THz temporal transients approximate a single-cycle oscillation with different grade of asymmetry. High energy per pulse concentrated in one or two optical cycles and very tight focusing, reported before, allow for high THz field. The maximum field recorded for the DSTMS displays peak field of 150 MV/m and therefore magnetic field of 0.5 Tesla. For OH1 and DAST organic crystal the peak field is slightly less than 100 MV/m when pumped by the same pump fluence. The higher frequencies for the DSTMS result in a tighter focus, shorter THz pulse and thus in higher peak field. The calculated spectra reveal the absorption properties and phonon resonances of the each organic material in good agreement with data reported in literature. More of 5-octave spectra are generated for all the crystal. As shown in the following with thinner organic crystals and interferometric measurement is possible to measure higher frequency.

Control of the THz Phase

The THz pulse generated by optical rectification is characterized by stable absolute phase. This is an important feature for multishot field-sensitive experiments. Equally important, for exploring nonlinear dynamics, is the generation of the highest field at the focus by proper control of the absolute phase of the THz oscillation. In fact, for the highest asymmetry and zero absolute phase offset, the THz pulse could potentially drive nonlinear phenomena as a quasi-unipolar stimulus.

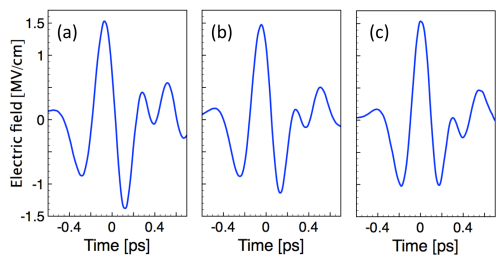


Figure 3: Direct control of the THz field absolute phase for the generation of quasi half-cycle transient.

We demonstrated recently an efficient method to directly control the absolute phase of the THz pulse by combining dispersion properties of different transparent plastics. Teflon and other polymer sheets with proper thickness were used in order to vary the THz absolute phase and forming a fully asymmetric pulse as shown in Figure 3. It is worth noting that the phase manipulation introduces minor energy losses and negligible decrease in the peak field. Moreover, in our setup, the THz field

ISBN 978-3-95450-126-7

polarity can be easily inverted by 180 deg rotation of the organic crystal.

Ultra-broadband THz Generation

Both the thickness of the organic emitter and the pump pulse duration determine the maximum accessible THz bandwidth. The highest THz frequency is related to the spectral components of the pump pulse. The phase velocity bandwidth of the optical rectification is inversely proportional to the thickness of the organic crystal. For the generation of higher frequencies thin crystals are then necessary.

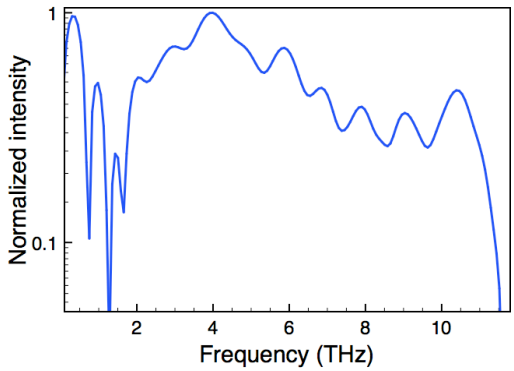


Figure 4: Broadband spectrum generated by optical rectification in180 μm thick DAST crystal.

In Figure 4 the broadband spectrum generated in 180 μm thick DAST is shown. To avoid the decrease of sensitivity caused by the electro-optic detection (at frequencies higher than 5 THz), the spectra are reconstructed by Fourier interferometry. The measured spectrum is impressively large and extends far beyond 10 THz. The measurement indicates that with organic crystals it is possible to produce high fields in the whole THz gap (0.1-10 THz) and furthermore, it seems feasible to extend the spectral region of the emitted radiation up to frequencies only accessible by optical difference frequency generation (>20THz). The absorption line corresponds to the phonon resonances of the DAST. This is an ideal source for time resolved spectroscopy in conjunction with the Xray FEL probe.

Ultrafast Magnetization driven by Intense THz Field

The intense terahertz source presented here is an ideal tool to explore nonlinear phenomena and to perform time resolved spectroscopy. THz magnetic field approaching 1 tesla, opens moreover, new opportunities for coherent control of magnetization on femtosecond timescale. We demonstrated experimentally that the magnetization in a cobalt thin film could be directly steered by the intense magnetic Terahertz laser field in non-resonant conditions [8].

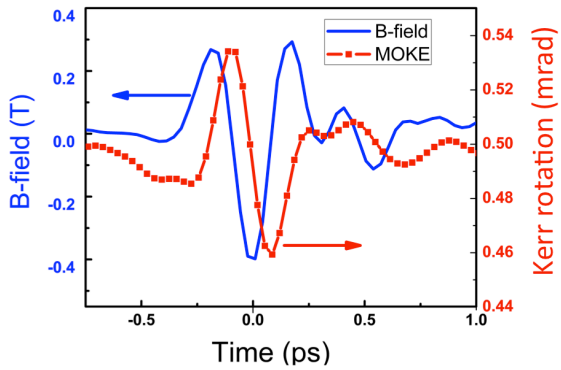


Figure 5: Ultrafast magnetization represented by the MOKE curve drive by intense THz magnetic field.

As shown in Figure 5, ultrafast magnetization (red curve) in Co film is initiated by THz magnetic field (blue plot). The magnetic vector rotation is visualized by time-resolved magneto-optic Kerr effect (MOKE). The strong (0.4 T) single-cycle Terahertz pulse acts as a cold stimulus and permits the exact manipulation of the magnetization. Dissimilarly from incoherent magnetization dynamics initiated by conventional lasers, in the presented results, the phase and the amplitude information of the THz field are directly imprinted in the magnetization response. The resulted precession of the magnetic vector occurs on the same time scale of the THz stimulus. This precise control of magnetization on an ultrafast timescale is a novel tool and is expected to pave the way to purely optical ultrafast data storage.

CONCLUSIONS

In this paper we report terahertz source based on OH1, DAST, DSTMS organic crystals in view of their potential of high-field generation in the THz gap (0.1-10 THz) for FEL. All the above organic crystals turned out to be highly efficient (up to 2% energy conversion yield) and broadband THz emitters when pumped by mJ femtosecond infrared pulses. The generated THz radiation offers multi-octave spanning, single-cycle and phase-stable pulses with up to 1.5 MV/cm electric and 0.5 Tesla magnetic field strength. The generation scheme based on a collimated pumping geometry provides excellent THz focusing characteristics. We present a new method to efficiently control the absolute phase and the polarity of the THz field. Ultra-broadband THz spectra covering the frequency range of 1-10 THz could be realized by optical rectification in a thin organic crystal.

High peak magnetic field produced in organic crystal has been used to initiate ultrafast magnetization in Co film. Due to the absence of heating deposition associated to the THz stimulus the phase and amplitude of the magnetic field are directly and coherently imprinted in the magnetic response. The results represent an important milestone for demonstrating exact control of the magnetization by laser magnetic field.

ISBN 978-3-95450-126-7

The source presented here paired with hard Xray FEL will be a power tool to investigate field-sensitive and extreme nonlinear phenomena. The excellent stability and reproducibility make the THz generation in organic crystal well suited for temporal photon diagnostic in Xray FEL.

ACKNOWLEDGMENT

We acknowledge support from SwissFEL, the Swiss National Science Foundation (grant no. PP00P2128493 and 20002-1122111) and SCIEX under grant 12.159.

REFERENCES

[1] M. Tonouchi, Nature Photon. 1, 97 (2007).
[2] M. C. Hoffmann and J. J. Turner, Synchr. Radiat. News, 25-2, 17 (2012).
[3] U. Fruehling et al Nature Photon. 3, 523 (2009).
[4] C. P. Hauri, C. Ruchert, F. Ardana and C. Vicario, Appl. Phys. Lett. 99, 161116 (2011).
[5] C. Ruchert, C. Vicario and C. P. Hauri, Opt. Lett. 37, 899 (2012).
[6] C. Ruchert, C. Vicario and C. P. Hauri, Phys. Rev. Lett., 110, 123902 (2013).
[7] C. Vicario, C. Ruchert and C. P. Hauri, J. Mod. Opt. 2013 DOI:10/1080/0500340.2013.800242 http://dx.doi.org/10.1080/09500340.2013.800242
[8] C. Vicario, C. Ruchert, F. Ardana-Lamas, P.M. Derlet, B. Tudu, J. Luning and C.P. Hauri, Nature Photon. (2013) DOI: 10.1038/nphoton.2013.209

ISBN 978-3-95450-126-7