UNIVERSITY OF PÉCS

DOCTORAL SCHOOL OF PHYSICS

Laser Physics, Nonlinear Optics and Spectroscopy Program

Analysis of tilted-pulse-front excitation terahertz sources

Summary of the PhD Thesis

Levente Tokodi

Supervisor:
László Pálfalvi, DSc
full professor

PÉCS, 2020

1. Overview

High-intensity terahertz (THz) source development is a high-priority research area in the last decade. Due to the widespread availability of compact, or table-top laser sources, THz generation via utilize nonlinear optics – such as difference frequency generation (DFG) and especially optical rectification (OR) – became the prominent method. Nowadays highly interested areas of applications required the growth of higher and higher THz pulse energies and peak electric fields. For example, linear- and nonlinear THz spectroscopy requires few fJ pulse-energies with few-hundreds V/cm electric fields and 1-10 μJ pulse-energies with peak electric fields on the order of 100 kV/cm, respectively. Extremely high THz pulse energies (>1mJ) with 10-100 MV/cm peak-electric fields are necessary for such exotic areas as THz driven electron accelerators [1], intense THz-laser driven proton acceleration in plasmas [2] and single-cycle attosecond pulse generation with THz pulses [3, 4].

Optical rectification of femtosecond (fs) laser pulses in nonlinear materials – as was mentioned earlier – is a promising and dominant way for generation of intense THz pulses. It is essential for the generation to fulfill the requirement of velocity-matching, or in other words the group velocity of the pumping pulse needs to be equivalent with the phase velocity of the generated THz pulse in the medium. Inorganic materials with extremely high nonlinearity (e.g. LiNbO3 (LN) and LiTaO3 (LT)) are promising but having extremely large gap between the index of refraction on the optical and on the THz-spectral range inflict the impossibility of collinear velocity matching. To overcome this, the so-called tilted-pulse-front method was born in the early 2000's by Hebling et al. [5] where one can achieve the velocity matching condition – in materials like the mentioned ones – by tilting the intensity-front of the pumping pulses. This simple and elegant method proved so effective, that many research-groups use it since its presentation, and nothing shows the method's performance better than in 2014, Fülöp et al. successfully reached nearly 0.5 mJ THz pulse energy in LN [6].

With the increasing need for higher THz pulse energies, and better THz sources, numerical methods and simulation techniques are evolved together to assist the source-development. Quantitative and qualitative information from ray-tracing simulations and/or outputs of mathematical codes are often have cardinally importance on the point of view of the designing. Simulations on the process of THz generation are evolved well from the beginning [7] complemented by such effects as the distortion in the pump pulse lengths [8], the occurrence of multi-photon absorption [9, 10] to nowadays where the most complete models are taking into the account all the effects mentioned before and the so-called cascade-effect as well [11, 12].

2. Objectives and methods

My first goal was to develop a ray-tracing simulation based numerical method, which is suitable to analyze the realizable THz excitation geometries by its imaging system's optical aberrations effects on the pump pulses. This is an important step before the realizations of such experimental systems because basing on the results one can implement further optimizations, thus can make source developments more time and cost effective at the institute. To realize this my tasks was numerical investigations by ray-tracing analyses of systems with well-described geometries, to these I was need to search a capable ray-tracing software and develop a mathematical code which is able to give quantitative and qualitative results from the distortion of the pump pulse in time- (pulse length broadening) and space (deviation of the shape of the pulse front from plane) domain based on the outputs of the ray-tracing simulation software.

Lithium-tantalate is a promising candidate for THz generation as well as LN due to its high nonlinear coefficient. Furthermore, the possibility of applying the widely available Ti:Sapphire pumping laser source (at 800 nm central wavelength) without the occurrence of the three-photon absorption was an alluring property too. To characterize and examine the crystal and its possibility to design a system based on it was the following of my goals.

Through all my examinations and optimizations, the technical feasibility was a main criterion. Based on this, although the dissertation contains theoretically analyzes, simulations and numerical calculations, the descriptions and results can be considered as a "recipe" to realize or build a given THz source.

3. New scientific results

1. I have developed a ray tracing simulation based numerical method for analyzing the distortions of the pump pulses in tilted-pulse-front excitation terahertz sources. With this method I analyzed the telescopic THz source and compared the sources containing one BK7 lens and one achromatic lens in conventional setup, and two BK7 lenses- and two achromatic lenses in telescopic setup. I showed that the using of an achromatic lens in conventional setup cause significantly reduced pulse distortions comparing to the case of using a BK7 lens. Furthermore, I stated that instead of using the BK7 telescope which is slightly better than the case of using an achromatic lens we can achieve even better results if we use a telescopic setup containing two achromatic lenses. [S1]

2. With adapting the method which was mentioned in the first thesis point, I made ray-tracing simulations and numerical analysis on the hybrid contact grating terahertz source. Examined the distortions of the

pump beam for a few case (geometrical and grating parameters) I showed that the hybrid system with one BK7 lens performs excellently: Assuming a 16 mm pump beam diameter I have calculated about one third time-domain distortion at the edges of the pump beam compared to the one BK7 lens containing conventional setup. Furthermore, in the case of the hybrid source, the wedge angle of the LiNbO3 crystal needs to be just 25-30° instead of the high (63°) wedge angle in the conventional setup and this may result in a higher quality THz beam profile in the case of the hybrid source. [S2]

3. I have optimized the diffraction elements of the hybrid source containing lithium tantalate for maximizing its efficiency. With systematically changing the geometrical parameters (filling factor, groove depths and grating constant) of the binary grating structure on the input surface of the crystal I showed those conditions, with which the grating's diffraction efficiency will be maximum. To increase the THz generation efficiency, I proposed the

changing of the initial pulse-front-tilting grating – which was formerly a reflection one – to a transmission one. I have shown with optimization simulations with the usage of the proposed grating in near Littrow configuration leads to that the pump beam can be coupled into the terahertz generator crystal with optimized contact grating with greater efficiency. [S3]

4. I have made the raytracing analysis on the hybrid THz source containing a plane-parallel nonlinear crystal with a stair-step echelon input surface. Since the limits of the ray-tracing software I searched and proposed a solution to make the analyses which was suitable to examine the effects of the system's optical aberrations on the pump pulses. With the modified solution I stated that as the results of the reduced imaging errors the distortions of the pump pulse will significantly reduce in the hybrid stair-step echelon system compared to the conventional one. [S4]

4. List of publications related to the thesis

S1. **L. Tokodi**, J. Hebling, and L. Pálfalvi, "*Optimization of the Tilted-Pulse-Front Terahertz Excitation Setup Containing Telescope*," Journal of Infrared, Millimeter, and Terahertz Waves **38**, 22-32 (2017).

S2. L. Pálfalvi, Z. Ollmann, **L. Tokodi**, and J. Hebling, "*Hybrid tilted-pulse-front excitation scheme for efficient generation of high-energy terahertz pulses*," Opt. Express **24**, 8156-8169 (2016).

S3. **L. Tokodi**, A. Buzády, J. Hebling, and L. Pálfalvi, "*Possibility of high-energy THz generation in* $LiTaO_3$," Applied Physics B **122**, 1-7 (2016).

S4. L. Pálfalvi, G. Tóth, **L. Tokodi**, Z. Márton, J. A. Fülöp, G. Almási, and J. Hebling, "*Numerical investigation of a scalable setup for efficient terahertz generation using a segmented tilted-pulse-front excitation*," Opt. Express **25**, 29560-29573 (2017).

5. References

1. A. Sharma, Z. Tibai, J. Hebling, and J. A. Fülöp, "*Terahertz-driven wakefield electron acceleration*," Journal of Physics B: Atomic, Molecular and Optical Physics **51**, 204001 (2018).
2. A. Sharma, Z. Tibai, and J. Hebling, "*Intense tera-hertz laser driven proton acceleration in plasmas*," Physics of Plasmas **23**, 063111 (2016).
3. E. Balogh, K. Kovacs, P. Dombi, J. A. Fulop, G. Farkas, J. Hebling, V. Tosa, and K. Varju, "*Single attosecond pulse from terahertz-assisted high-order harmonic generation*," Physical Review A **84**, 023806 (2011).
4. G. Tóth, Z. Tibai, A. Sharma, J. A. Fülöp, and J. Hebling, "*Single-cycle attosecond pulses by Thomson backscattering of terahertz pulses*," J. Opt. Soc. Am. B **35**, A103-A109 (2018).
5. J. Hebling, G. Almási, I. Z. Kozma, and J. Kuhl, "*Velocity matching by pulse front tilting for large-area THz-pulse generation*," Opt. Express **10**, 1161-1166 (2002).

6. J. A. Fülöp, Z. Ollmann, C. Lombosi, C. Skrobol, S. Klingebiel, L. Pálfalvi, F. Krausz, S. Karsch, and J. Hebling, "*Efficient generation of THz pulses with 0.4 mJ energy*," Opt. Express **22**, 20155-20163 (2014).
7. K. L. Vodopyanov, "*Optical generation of narrow-band terahertz packets in periodically-inverted electro-optic crystals: conversion efficiency and optimal laser pulse format*," Opt. Express **14**, 2263-2276 (2006).
8. J. A. Fülöp, L. Pálfalvi, G. Almási, and J. Hebling, "*Design of high-energy terahertz sources based on optical rectification*," Opt. Express **18**, 12311-12327 (2010).
9. X. Wu, S. Carbajo, K. Ravi, F. Ahr, G. Cirmi, Y. Zhou, O. D. Mücke, and F. X. Kärtner, "*Terahertz generation in lithium niobate driven by Ti:sapphire laser pulses and its limitations*," Opt. Lett. **39**, 5403-5406 (2014).
10. S.-C. Zhong, Z.-H. Zhai, J. Li, L.-G. Zhu, J. Li, K. Meng, Q. Liu, L.-H. Du, J.-H. Zhao, and Z.-R. Li, "*Optimization of terahertz generation from*

$LiNbO_3$ under intense laser excitation with the effect of three-photon absorption," Opt. Express **23**, 31313-31323 (2015).

11. K. Ravi, W. R. Huang, S. Carbajo, X. J. Wu, and F. Kartner, "*Limitations to THz generation by optical rectification using tilted pulse fronts*," Opt. Express **22**, 20239-20251 (2014).
12. K. Ravi, W. R. Huang, S. Carbajo, E. A. Nanni, D. N. Schimpf, E. P. Ippen, and F. X. Kärtner, "*Theory of terahertz generation by optical rectification using tilted-pulse-fronts*," Opt. Express **23**, 5253-5276 (2015).