University of Pécs

Doctoral School of Physics
Laser physics, nonlinear optics and spectroscopy program

Scalable terahertz pulse sources based on lithium-niobate

PhD thesis

Krizsán Gergő

Supervisor:

Dr. Fülöp József András

Pécs, 2022.

1. Introduction and aims

Thanks to the widespread use of femtosecond lasers in the last three decades, the whole of the terahertz (THz) range (~0.1-10 THz), which was previously unavailable or difficult to reach, became available, as well as the possibility of direct measurement of electric field strength. The ranges of ~0.1–2 THz, ~1–4 THz, and ~0.1–10 THz became available by optically rectifying ultrashort pulses (~100 fs) in different nonlinear optical materials. THz pulses in these frequency ranges typically occur in lithium niobate (LiNbO3, LN) [1], semiconductors (e.g. zinc telluride (ZnTe) [2], gallium phosphide (GaP) [3]) or organic crystals (e.g. : DAST [4], OH1 [5], DSTMS [6,7]), respectively.

Applications of THz pulses include safety technology, time-domain linear and nonlinear spectroscopy, imaging and non-destructive material testing, as well as the application with perhaps the greatest interest today: charged particles (electrons [8,9] or even ions [10]) manipulation and acceleration.

In the past two-decade terahertz (THz) pulse sources went through a rapid development thanks to the increasing pump energies and the tilted pulse front excitation scheme [11]. LN based sources provided the highest pulse energies and electric field strength [12] in the low frequency part of the THz range (0.1-2 THz), which is especially advantageous for particle acceleration. Increasing the THz generation efficiency and scaling the size of the conventional THz pulse sources, which contains an optical grating, imaging optical element and a prism shape lithium niobate (LN) crystal, have strong limitations.

These are the following: (i) Limited interaction length due to angular dispersion [13]. (ii) Imaging errors, which results curved pulse fronts and pump pulse lengthening at the sides of large pump beams [13,14]. (iii) Prism shape of the LN crystal with a large ($\gamma \approx 63°$) wedge angle, resulting in different temporal waveform along the THz beam [15]. Energy scalability is limited by (i)-(iii), while the availability of uniform THz beam is limited mainly by (ii) and (iii). The good beam quality is required by many high-field application e.g., particle acceleration. In the past few years extensive effort has been made to reduce or even eliminate these limitations.

Our group proposed several new setups for THz generation. All of them requires the microstructuring of the LN surface. In some cases a contact grating should be created in the front side of the LN crystal [16,17], in others microstructuring a stair-step (echelon) structure is needed [15,18,19]. Depending on the pre-tilt angle introduced by the optical grating (and on the use of any imaging element) a nonlinear echelon slab (NLES) or a NLES with small wedge angle can be used. Meanwhile all of these proposed setups are reducing the limitations, they are not scalable without any principal limitations. During my work one of my goals was to design a setup, which is scalable without any limitations and can produce THz pulses with relatively high efficiency. Another advantage would be if the nonlinear material surface microstructuring would not be required.

2. Methodes

Most of my work was to design new, scalable THz pulse sources. This included the further development of previously proposed sources, in which I proposed the use of new optical elements. Such an optical element was the volume phase holographic grating in combination with the nonlinear echelon slab. In this setup the imaging element can be omitted. The other optical element, which I proposed to use was the external structured reflector. The micromachining of this element is much easier than that of LN.

I built, demonstrated, and characterized the setup containing a nonlinear echelon slab, which was pumped through a volume phase holographic grating. The pumping wavelength was 1030 nm and the pulse length was 200 fs.

Using the Lorentz-Lorenz equation, I designed a nanocomposite liquid with a high refractive index, which can be ideal as a refractive index matching liquid for LN (or lithium tantalate), and can enable efficient coupling between the reflective nonlinear slab and the external structured reflector.

3. New scientific results

[1] I suggested to use volume phase holographic grating in combination with the nonlinear echelon slab. In the resulted setup any imaging element can be omitted and the whole setup can become plan-parallel and scalable without any principal limitations. With numerical simulations volume phase holographic grating have been designed for the setup. Furthermore, we showed that applying a refractive index matching liquid between the grating and the slab reflection losses can be decreased, and THz generation efficiency can be increased. [S2]

[2] At low pump intensity proof-of principle experiment had been demonstrated with a prototype nonlinear echelon slab in combination with a volume phase holographic grating. The THz generation efficiency was nearly the same as with the setup containing imaging element. The maximum conversion efficiency was 0.03% and the generated maximum THz pulse energy was 0.2 μJ. [S2]

[3] I suggested to use an external structure reflector in combination with a plane-parallel nonlinear slab. Microstructuring a metal plate is much easier and can be

made with a better surface quality. For efficient incoupling between the external structured reflector and the nonlinear material slab a high refractive index liquid has to be used. The setup is especially advantageous for nonlinear materials requiring pulse-front-tilt angle bigger than 60°. With numerical simulations we showed that for lithium niobate and lithium tantalate materials at 800 and 1030 nm pump wavelengths at appropriately high diffraction order the diffraction efficiency can be exceptionally high (>85%). [S1]

[4] I suggested to use nanocomposite liquid as high refractive index liquid in the reflective nonlinear echelon slab – external structured reflector setup. I have selected materials for the nanocomposite liquid and gave their refractive indices and the nanopowder concentration values for lithium niobate and lithium tantalate nonlinear materials at 800 and 1030 nm pump wavelengths. [S1]

4. Related publications

[S1] **G. Krizsán**, Z. Tibai, J. Hebling, L. Pálfalvi, G. Almási, and G. Tóth, *„Lithium niobate and lithium tantalate based scalable terahertz pulse sources in reflection geometry”*, Opt. Express **28**, 34320 (2020).

[S2] **G. Krizsán**, Z. Tibai, G. Tóth, P. S. Nugraha, G. Almási, J. Hebling, and J. A. Fülöp, *„Uniformly scalable lithium niobate THz pulse source in transmission geometry”,* Opt. Express **30**, 4434 (2022).

5. Other publications

[S3] P. S. Nugraha, **G. Krizsán**, Gy. Polónyi, M. Mechler, J. Hebling, Gy. Tóth, and J. Fülöp, *„Efficient semiconductor multicycle terahertz pulse source”*, Journal of Physics B: Atomic, Molecular and Optical Physics **51**, 094007 (2018).

[S4] P. S. Nugraha, **G. Krizsán**, Cs. Lombosi, L. Pálfalvi, Gy. Tóth, G. Almási, J. A. Fülöp, and J. Hebling, *„Demonstration of a tilted-pulse-front pumped plane-parallel slab terahertz source”*, Opt. Lett. **44**, 1023 (2019)

[S5] Gy. Tóth, L. Pálfalvi, J. A. Fülöp, **G. Krizsán**, N. H. Matlis, G. Almási, and J. Hebling, *„Numerical* *investigation of imaging-free terahertz generation setup using segmented tilted-pulse-front excitation"*, Opt. Express **27**, 7762 (2019).

[S6] V. Stummer, T. Flöry, **G. Krizsán**, Gy. Polónyi, E. Kaksis, A. Pugžlys, J. Hebling, J. A. Fülöp, and A. Baltuška, *„Programmable generation of terahertz bursts in chirped-pulse laser amplification"*, Optica **7**, 1758 (2020)

[S7] Sz. Turnár, **G. Krizsán**, J. Hebling and Z.Tibai „*Waveguide structure based electron acceleration using THz pulses"*, Optics Express (2022, Elfogadva)

[S8] N. M. Mbithi, Gy. Tóth, Z. Tibai, I. Benabdelghani, L. Nasi, **G. Krizsán**, J. Hebling and Gy. Polónyi „*Investigation of terahertz pulse generation in semiconductors pumped at long infrared wavelengths"*, Optics Express (2022, Beadva)

[S9] Z.Tibai, **G. Krizsán**, Gy. Tóth, G. Almási, G. Illés, L. Pálfalvi and J. Hebling „*Scalable Microstructured Semiconductor THz Pulse Sources"*, Optics Express (2022, Beadva)

6. References

[1] J. A. Fülöp, Z. Ollmann, C. Lombosi, C. Skrobol, S. Klingebiel, L. Pálfalvi, F. Krausz, S. Karsch, and J. Hebling, Opt. Express **22**, 20155 (2014).

[2] G. Polónyi *et al.,* Optics express **24**, 23872 (2016).

[3] G. Polónyi, M. I. Mechler, J. Hebling, and J. A. Fülöp, IEEE J. Sel. Top. Quantum Electron. **23**, 1 (2017).

[4] C. P. Hauri, C. Ruchert, C. Vicario, and F. Ardana, Appl. Phys. Lett. **99**, 161116 (2011).

[5] C. Ruchert, C. Vicario, and C. P. Hauri, Opt. Lett. **37**, 899 (2012).

[6] M. Shalaby and C. P. Hauri, Nature communications **6**, 1 (2015).

[7] C. Vicario, A. V. Ovchinnikov, S. I. Ashitkov, M. B. Agranat, V. E. Fortov, and C. P. Hauri, Opt. Lett. **39**, 6632 (2014).

[8] D. Zhang *et al.,* Nat. Photonics **12**, 336 (2018).

[9] S. Turnár, J. Hebling, J. Fülöp, G. Tóth, G. Almási, and Z. Tibai, Appl. Phys. B **127**, 1 (2021).

[10] L. Pálfalvi, J. Fülöp, G. Tóth, and J. Hebling, Physical Review Special Topics-Accelerators and Beams **17**, 031301 (2014).

[11] J. Hebling, G. Almasi, I. Z. Kozma, and J. Kuhl, Opt. Express **10**, 1161 (2002).

[12] B. Zhang *et al.,* Laser & Photonics Reviews **15** (2021).

[13] J. Fülöp, L. Pálfalvi, G. Almási, and J. Hebling, Optics express **18**, 12311 (2010).

[14] L. Tokodi, J. Hebling, and L. Pálfalvi, Journal of Infrared, Millimeter, and Terahertz Waves **38**, 22 (2017).

[15] G. Tóth, L. Pálfalvi, J. A. Fülöp, G. Krizsán, N. H. Matlis, G. Almási, and J. Hebling, Optics Express **27**, 7762 (2019).
[16] Z. Ollmann, J. Hebling, and G. Almási, Appl. Phys. B **108**, 821 (2012).
[17] L. Pálfalvi, Z. Ollmann, L. Tokodi, and J. Hebling, Optics Express **24**, 8156 (2016).
[18] L. Pálfalvi, G. Tóth, L. Tokodi, Z. Márton, J. A. Fülöp, G. Almási, and J. Hebling, Optics express **25,** 29560 (2017).
[19] P. S. Nugraha, G. Krizsán, C. Lombosi, L. Pálfalvi, G. Tóth, G. Almási, J. A. Fülöp, and J. Hebling, Opt. Lett. **44**, 1023 (2019).