UNIVERSITY OF PÉCS

Doctoral School of Physics

Pump pulse width and temperature effects in high energy lithium niobate THz sources

PhD Thesis

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1. PRELIMINARIES AND OBJECTS

Intense single-cycle terahertz source development in the last decade allowed the growth of new areas of research. Applications as THz-assisted attosecond pulse generation [1], THz pump - THz probe measurements [2], molecule alignment with an intense field [3] benefit highly from intense terahertz pulses. However, newly emerging areas as acceleration, longitudinal compression, and undulation of relativistic electron bunches [4-6], post-acceleration of laser-generated proton and ion beams with potential applications for hadron therapy [7], and multispectral single-shot imaging would benefit from field strengths higher than currently provided.

Today extremely high field strengths up to 100 MV/cm are available only in the higher frequency range [8]. Semiconductor [8] and organic crystal based terahertz sources [9-11] have good properties in the higher frequency range to achieve fields strengths due to tight focusing capabilities. Although low frequency range THz sources show an advantage for the mentioned applications – so that longer wavelength is preferably matching typical transversal sizes of particle beams –, pulse energies and peak electric fields necessary for these applications is presently not available. Zinc telluride is a good candidate, however high energy is only achievable above the three-photon absorption limit [12]. Tilted-pulse-front pumping [TPFP] [13] of inorganic lithium niobate ($LiNbO_3$, LN) shows good characteristics to achieve the highest possible pulse energies below 1 THz.

Therefore, my priority is to optimize THz sources generated by optical rectification of fs laser pulses with tilted-pulse-front pumping [13] in lithium niobate.

Theoretical studies [14-16] predict an increase in efficiency with optimization of pump pulse duration and cryogenic cooling of LN source crystal. In order to experimentally demonstrate these predictions I conducted experiments with terahertz sources in a tilted-pulse-front pumping scheme in various configurations using different pump pulse durations, source temperatures, and TPFP imaging configurations.

2. METHODS

I proved experimentally results of theoretical studies [14] that predict improvement of single-cycle terahertz source conversion efficiency:

(i) by studying a terahertz source where the lithium niobate crystal is pumped by laser source that emits transform-limited pulses between 0.38 ps and 0.65 ps with pulse energies up to 15 mJ at a central wavelength of 1030 nm, and giving an optimal pulse duration for efficient THz generation.

(ii) by studying a cryogenically-cooled lithium niobate terahertz source, and measuring the enhancement factor ${}^{\eta_{CT}}/_{\eta_{RT}}$ for moderate (below $45\,\mu J$) and high (50 to 180 μJ) terahertz pulse energies.

(iii) by waveform measurement of terahertz pulses with the possible highest pulse energies in the low frequency region using electro-optic sampling technique.

Also, I compared two, highly optimized terahertz sources to improve the knowledge about low-frequency terahertz sources using tilted-pulse-front pumping. A setup was built with two-lens imaging and another by a single achromat lens imaging. I realized spatial characterization and electro-optic measurement of the generated THz beam to receive information on parameters – beam divergence angle and ellipticity of beam cross section – that are important during focusing and beam transportation of such pulses. I conducted these measurements at various pump intensities for both setups.

3. NEW SCIENTIFIC ACHIEVEMENTS

I. With the aim to increase pump-to-THz generation efficiency I have investigated a THz source based on optical rectification of femtosecond laser pulses with tilted-pulse-front pumping in a lithium niobate crystal illuminated with variable-duration transform-limited pulses. I have shown that longer pump pulses are favorable for reaching higher conversion efficiency, in accordance with the general trend predicted by theoretical calculations [S1]. Moreover, I have shown that a broader THz spectrum can be achieved by using shorter pulses.

II. THz generation efficiency investigations were carried out by building diverse cryogenic-cooled THz sources based on tilted-pulse-front pumping in a lithium niobate crystal. I have shown up to 45μJ THz pulse energy that the conversion efficiency can be increased by a factor of 4 using cryogenic cooling of the source crystal. Above 50 μJ about up to 180 μJ an enhancement factor of 2.4 was demonstrated. [S1, S2]

III. By illuminating a room temperature lithium niobate crystal in a pulse front tilting setup with large pump energy and a large pump intensity in combination with optimal imaging and nearly optimal pump pulse duration I have demonstrated the highest pulse energies up to 0.4 mJ in the frequency range below 1 THz. I have demonstrated a generation efficiency of 0.77% which is the highest for large pulse energies. [S2]

IV. I have demonstrated for the first time generation of THz pulses with parameters that are applicable for charged-particle acceleration, for example for THz driven evanescent-wave proton post-accelerator. In this lithium niobate based cryogenic-cooled THz source I have measured in-focus waveforms that show a spectral peak at about 0.2 THz and a 0.65 MV/cm calculated peak electric field strength. [S2]

V. I have investigated experimentally near-field THz beam profiles. Measurements show that increasing the pump fluence leads to a beam diameter decrease in the horizontal plane while there is no change in the vertical plane. Moreover, performing the corresponding measurements for THz beam divergence I have found that the angle increases in the horizontal direction while no significant change can be observed in the vertical direction [S3]. These results indicate that nonlinear beam distortions have a high relevance at designing high energy THz sources.

4. ARTICLES RELATED TO THE TOPIC OF THIS THESIS

[S1] C Vicario, B Monoszlai, Cs Lombosi, A Mareczko, A Courjaud, JA Fülöp, CP Hauri, *"Pump pulse width and temperature effects in lithium niobate for efficient THz generation"* Optics Letters 38 (24), 5373-5376

[S2] JA Fülöp, Z Ollmann, Cs Lombosi, Ch Skrobol, S Klingebiel, L Pálfalvi, F Krausz, S K, J Hebling, "*Efficient generation of THz pulses with 0.4 mJ energy*", Optics Express 22 (17), 20155-20163

[S3] C Lombosi, G Polónyi, M Mechler, Z Ollmann, J Hebling, JA Fülöp, "*Nonlinear distortion of intense THz beams*", New Journal of Physics 17 (8), 083041

5. OTHER ARTICLES

[S4] W.Schneider, A Ryabov, Cs Lombosi, T Metzger, Zs Major, JA Fülöp, Peter Baum, *“800-fs, 330-μJ pulses from a 100-W regenerative Yb: YAG thin-disk amplifier at 300 kHz and THz generation in LiNbO 3”*, Optics Letters 39 (23), 6604-6607

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[15] J. A. Fülöp, L. Pálfalvi, M. C. Hoffmann, and J. Hebling, Opt. Express **19**, 15090 (2011).

[16] M. I. Bakunov, S. B. Bodrov, and E. A. Mashkovich, J. Opt. Soc. Am. B **28**, 1724 (2011).