UNIVERSITY OF PÉCS

Physics Doctoral School

Laser physics, nonlinear optics and spectroscopy program

Investigation of scalable concepts for intense terahertz pulse generation

PhD Thesis

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1. BACKGROUND AND OBJECTIVES

The study and control of materials with extremely strong THz fields [1, 2], or the acceleration and manipulation of electrons [3] and protons [4] are emerging applications which require THz sources with unprecedented parameters. Besides the pulse energy, an excellent focusability is also essential to achieve the highest possible field strengths.

Optical rectification of ultrashort laser pulses with tilted pulse front in lithium niobate (LN) [5] has become a standard technique for efficient THz generation. Conventionally, a prism-shaped LN crystal is used with a large wedge angle equal to the pulse-front tilt (63°). Such a source geometry results in a nonuniform pump propagation length across the beam, which can lead to a spatially varying interaction length for THz generation [6].

This negatively affects the THz beam quality and, consequently, the focusability, thereby limiting the achievable field strength. Lateral beam (and eventually waveform) nonuniformity is especially problematic in high-energy THz sources [7], where a large-diameter pump beam is needed.

Different approaches have been proposed to mitigate limitations of tilted-pulse-front pumped THz sources. Recently, a modified hybrid approach to provide uniform interaction length across large pump and THz beams have been proposed [8]. The setup uses a plane-parallel LN slab as the nonlinear medium, which is equipped with an echelon structure on its input surface. Inside the LN slab, a segmented tilted pulse front is formed with an average tilt angle as required by phase matching.

Intense nearly single-cycle terahertz (THz) pulses can be used in materials science [2] and for the acceleration of electrons [3] and protons [4, 9]. The waveguide and resonator structures proposed for electron acceleration can be more efficiently driven by multicycle THz pulses. Multicycle THz pulses also could be used as drivers of coherent X-ray generation [10] and electron beam diagnostics. In THz spectroscopy experiments, multicycle narrowband THz pulses with high spectral brightness can selectively address different lattice, electronic, and spin degrees of freedom [11].

It has been recently demonstrated that semiconductor nonlinear materials offer a promising alternative with scalability to highest THz pulse energies and field strengths [12-16], enabling also the construction of monolithic contact-grating sources [14]. Lee et al.

showed the generation of multicyle narrow-bandwidth THz pulses in periodically inverted GaAs structures using OR at 2 μm wavelength [17].

Semiconductor materials have been playing an important role in many photonics-based technologies since decades. Novel high-power ultrashort-pulse laser and parametric sources, operating at infrared wavelengths, have recently enabled new application areas in nonlinear optics [18, 19]. To design and optimize setups and devices utilizing intense optical driving of semiconductors, the knowledge of their nonlinear optical parameters, such as the nonlinear refractive index or multiphoton absorption coefficients, is very important.

Theoretical scaling laws for MPA coefficients have been given for direct-bandgap semiconductors based on a two-band model [20]. An early summary on theoretical models and experimental values of two- (2PA) and three-photon absorption (3PA) coefficients for a few selected materials can be found in Ref. [21]. The most commonly used method to measure 2PA and 3PA coefficients is the z-scan technique [22]. This has been applied to measure the tensor properties (anisotropy) [23], by using the three nonlinear eigenpolarizations [24], and the dispersion of third-order nonlinearities (2PA) [23, 25] in various semiconductors. The dispersion and the anisotropy of 2PA and 3PA in GaAs has been measured in the 1.3–2.5 μm wavelength range by 100-fs pulses [26]. A maximum in the 3PA coefficient has been found both in theory and experiment near the 3PA cut-off wavelength. In contrast, little is known about the four-photon absorption (4PA) and nonlinearities of even higher order in semiconductors and other important optical materials.

This knowledge can be crucial for applications driven by infrared pulses. For example, in the newly developed very perspectivic semiconductor THz generators, 4PA can be a major limitation and hence should be considered in device design [16, 27]. A first attempt to estimate the 4PA coefficient in ZnTe has been made based on THz generation results [16], but this approach is very indirect and therefore subject to uncertainties. GaP is another semiconductor nonlinear material of high interest for efficient THz generation [16, 27], but no experimental data on its 4PA coefficient are available. Thus, there is a clearly perceived lack of knowledge on important material data.

2. METHODS

The investigated hybrid-type setup is a combination of the conventional scheme, containing a diffraction grating and imaging optics, and a nonlinear material with an echelon profile on its entrance surface (nonlinear echelon slab, NLES, Figure 1). Pump pulses of 200 fs pulse duration and 1030 nm central wavelength were delivered by a cryogenically cooled Yb:CaF_2 regenerative amplifier operating at 1 kHz repetition rate. Up to about 2.5 mJ pump pulse energy was used in the experiment. At the pump wavelength used and at room temperature, a pulse front tilt of about 63° is required for phase matching in LN. In case of the NLES, this pulse front tilt angle needs to be produced in air, at the entrance of the crystal. Here, $n_{\mathrm{g}} = 2.215$ is the group index of LN at the 1030 nm pump wavelength.

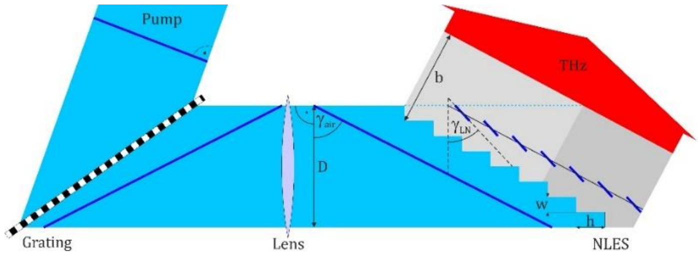

Figure 1. Scheme of the experimental setup.

For the simulations multicycle THz pulses, the one-dimensional wave equation with the nonlinear polarization has been solved in the spectral domain (see Eq. (1)-(5) in [6]). The simulation model took into account the variation of the pump pulse duration with propagation distance due to material and angular dispersions, the frequency-dependent absorption and refractive index in the THz range due to phonon resonances and FCA, latter caused by MPA of the pump. In case of calculations involving phase-matching frequencies higher than 1 THz, the dispersion of the effective second-order nonlinear susceptibility, $\chi^{(2)}$, was also taken into account [28].

The semiconductor nonlinear material selected for the simulations was GaP. GaP is preferred here because of its relatively large direct (indirect) bandgap of 2.78 eV (2.27 eV), its availability in larger sizes and better quality than e.g. ZnTe, and the relatively small absorption and dispersion in the THz range. Furthermore, a small pulse-front tilt angle of about 20°-30° is needed for phase matching in OR, resulting in only a minor variation of the duration of (sub-)ps-long pump pulses with propagation distance due to angular dispersion.

Nonlinear transmission measurements with fs pulses have been carried out by using the z-scan technique. The experimental setup is shown in Figure 2. Pump pulses of $\lambda_0 = 1.76$ μm central wavelength were delivered at 1 kHz repetition rate by a tuneable OPA, driven by a Ti:sapphire laser system. The full width at half maximum of the nearly Gaussian spectral intensity distribution of the pump pulses was about 50 nm. A set of dichroic long-pass filters have been used to suppress possible parasitic spectral components below about 1.65 μm. A pulse duration of 95 fs was measured by autocorrelation.

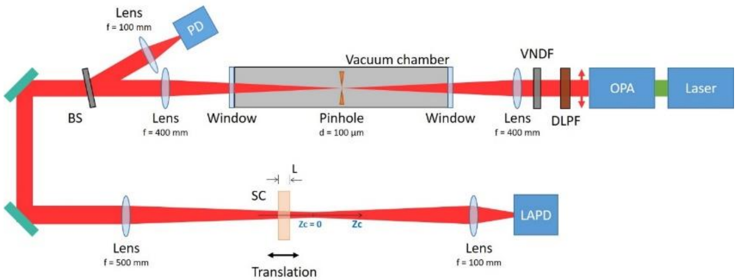

Figure 2. Scheme of the experimental setup.

A spatial filter was used to ensure a pump beam of good quality, with nearly Gaussian intensity profile and cylindrical symmetry. A lens with 500 mm focal length was used to focus the beam for the z-scan measurement. The horizontal and vertical beam profiles have been carefully measured by the knife-edge technique at several positions along the propagation direction of the focused beam. The waist radius of the focused pump beam was $w_0 = 39$ µm (at $1/e^2$ of the peak intensity). The Rayleigh range was $2z_{\mathrm{R}} = 5.5$ mm, significantly larger than the crystal length L. The sample crystal was mounted on a motorized linear stage to move it along the beam propagation direction (z-axis) around the focus. A large-area Ge photodiode was used to measure the power transmitted through the sample.

3. NEW SCIENTIFIC ACHIEVEMENTS

I. A new type of tilted-pulse-front pumped THz source based on a $LiNbO_3$ plane-parallel slab with an echelon structure on its input surface has been demonstrated [29]. Single-cycle pulses of 1.0 µJ energy and 0.30 THz central frequency have been generated with 5.1×10^{-4} efficiency. One order-of-magnitude increase in efficiency is expected by pumping a cryogenically cooled echelon of increased size and tichkness with a TiLsapphire laser. This new scheme enables straightforward scaling to high THz pulse energies by increasing the lateral size and the pump pulse energy and focusing to extremely high field strengths. [S1].

II. Multicycle THz pulse generation by OR in the semiconductor nonlinear material GaP was investigated with the help of numerical simulations. In the range of 0.1–7 THz frequency, pump-to-THz energy conversion efficiencies up to 3% were predicted for 5 mm crystal length. For an optimal combination of THz frequency, pump intensity, and crystal length, efficiencies as high as

8% of multicycle THz pulses can be expected [27] [S2, S4].

III. Intensity-dependent 4PA coefficients in GaP and ZnTe semiconductors have been measured by the z-scan method using pump pulses of 1.76 μm wavelength and 95 fs duration. The 4PA coefficients vary from 2.6×10^{-4} to 65×10^{-4} cm^5 GW^{-3} in GaP, and from 3.83×10^{-4} to 9.7×10^{-4} cm^5 GW^{-3} in ZnTe. The anisotropy in 4PA has been shown in GaP. The knowledge of the intensity-dependent 4PA coefficients is important for the design of practical devices, such as efficient semiconductor THz sources [S3, S5].

4. ARTICLES RELATED TO THE TOPIC OF THIS THESIS

[S1] **P. S. Nugraha**, G. Krizsán, C. Lombosi, L. Pálfalvi, G. Tóth, G. Almási, J. A. Fülöp, and J. Hebling, "Demonstration of a tilted-pulse-front pumped plane-parallel slab terahertz source," *Opt Lett*, vol. 44, pp. 1023-1026, 2019.

[S2] **P. S. Nugraha**, G. Krizsan, G. Polonyi, M. I. Mechler, J. Hebling, G. Toth, and J. A. Fulop, "Efficient semiconductor multicycle terahertz pulse source," J Phys B-at Mol Opt, vol. 51, 094007, 2018.

[S3] B. Monoszlai, **P. S. Nugraha**, Gy. Polonyi, Gy. Tóth, L. Pálfalvi, L. Nasi, Z. Ollmann, E.J. Rohwer, G. Gaumann, T. Feurer, J. Hebling, and J. A. Fülöp, "Measurement of Four-Photon Absorption in Semiconductors," submitted to *Opt Express*, 2019.

[S4**]** G. Tóth, **P. S. Nugraha**, G. Krizsán, M. I. Mechler, G. Polónyi, J. Hebling, and J. A. Fülöp, "Efficient semiconductor source of multicycle terahertz pulses using intensity-modulated pump," in *Conference on Lasers and Electro-Optics*, OSA Technical Digest (online) (Optical Society of America, 2018).

[S5] G. Polónyi, B. Monoszlai, **P. S. Nugraha,** G. Tóth, L. Pálfalvi, L. Nasi, Z. Ollmann, E. J. Rohwer, G. Gäumann, T. Feurer, J. Hebling, and J. A. Fülöp, "Measurement of Effective Four-Photon Absorption in Semiconductors," in Nonlinear Optics (NLO), OSA Technical Digest (Optical Society of America, 2019).

[S6] **P. S. Nugraha**, G. Krizsan, C. Lombosi, G. Toth, L. Palfalvi, G. Almasi, and J. Hebling, and J. A. Fulop, "Tilted-Pulse-Front Pumped Plane-Parallel $LiNbO_3$ Slab THz Source," *2019 44th International Conference on Infrared, Millimeter, and Terahertz Waves (Irmmw-THz),* 2019

5. Other articles

[S7] S. Li, **P. S. Nugraha**, X. Su, X. Chen, Q. Yang, M. Unferdorben, F. Kovács, S. Kunsági-Máté, M. Liu, X. Zhang, C. Ouyang, Y. Li, J. A. Fülöp, J. Han, and W. Zhang, "Terahertz electric field modulated mode coupling in graphene-metal hybrid metamaterials," *Opt Express*, vol. 27, pp. 2317-2326, 2019.

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