UNIVERSITY OF PÉCS

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Laser physics, nonlinear optics, and spectroscopy Program

Designs of optimized semiconductor contact grating terahertz sources

Ph.D. Thesis

Nelson Matheka Mbithi

Supervisor:

Dr. Gyula Polónyi

PTE TTK Institute of Physics



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1. Introduction

The advent of THz sources, which can generate pulses with high energies and peak electric and magnetic fields in the THz range (0 to 10 THz), has made new feasible applications such as nonlinear spectroscopy [1], manipulation of charged particles [2] and proposed for hadron therapy [3]. Terahertz pulses have been generated by optical rectification based on tilted-pulse-front excitation techniques [4]. The highest reported THz pulse energy in the low-frequency range to date utilizing the TPFP technique is more than 1.4 mJ in LN [5]. However, there are challenges in scaling the THz pulse energies and field strengths in lithium niobate; this includes: nonlinear interaction between the pump and THz beam leads to beam distortions [6, 7], large PFT angle leads to limited interaction length for THz pulse generation due to large angular dispersion [8]. In addition, the use of imaging optics in the case of conventional TPFP leads to imaging errors, which distorts the THz beam making it more difficult to focus the THz beam [9]. Several techniques to improve the conventional TPFP technique have been proposed and demonstrated [10-14]. On the other hand, organic materials have strong phonon absorption at low frequencies [15], are available in small sizes [16], and have a relatively low damage threshold [17]; this limits THz scalability.

Furthermore, THz pulse generation in semiconductors pumped in collinear geometry has been demonstrated [18, 19]. Unfortunately, semiconductors have small bandgap energies (E_g), which results in saturation effects when pumped at collinear geometry due to the presence of strong low order multiphoton absorption [20], limiting the useful pump intensity and, hence the terahertz energy. Pumping at long infrared pump wavelengths has been proposed and applied [21, 22] to eliminate low-order multiphoton absorption. THz pulses with energies as high as 14 µJ and conversion efficiency of 0.7% have been reported in ZnTe pumped at 1.7 µm infrared pump wavelength [20]. The contact grating (CG) based on TPFP was proposed [10] to achieve a collinear THz generation scheme at long pump wavelengths. Scalable THz pulses with up to 3.9 µJ and a conversion efficiency of 0.3% have been reported in the ZnTe CG source [22]. However, growing large size ZnTe crystals is difficult due to microbubbles [22]. Recently, GaP CG source has been reported [23]. GaP and GaAs are feasible alternatives as they are easier to grow and are less expensive [24]. This dissertation presents numerical simulations to design optimal GaP CG and GaAs CG THz sources and three ways of increasing the diffraction efficiencies to more than 91% in GaP and 89% GaAs for efficient THz pulse generation in the long infrared wavelengths are presented. In addition, numerical calculations were performed to determine the optimum pumping wavelength; in the calculations, the effect of the nonlinear refractive index and the wavelength dependence of the pumping OPA efficiency were taken into account.

2. Aims and Objectives

The main aim of this work was to numerically investigate THz generation efficiency and design optimized, scalable and highly efficient semiconductor contact grating terahertz sources pumped at a long infrared wavelength range.

By numerical simulations, the basic design aspects of semiconductors ZnTe and GaP, as well as their optimal pumping and phase matching conditions at infrared wavelengths below 2 µm, have been determined [25]. The first objective of the study was to extend by numerical calculations the determination of design aspects and the optimum pumping and phase matching conditions of GaAs and GaP pumped at long infrared wavelengths greater than 2 µm. I aimed to determine by numerical calculations the optimum pumping parameters such as pulse duration, pump intensity, crystal length, and phase-matching frequency that yield the highest terahertz conversion efficiency to give more insight into the semiconductor contact grating THz sources. In addition, It has been predicted and demonstrated that the elimination of low order multiphoton absorption (MPA) processes by using long pump wavelength can result in a significant increase in THz generation efficiency [20, 26-28] due to the possibility of using higher pump intensities without increasing the density of free carriers and their absorption in the THz range.

The second objective of the study was to numerically investigate the possibility of increasing terahertz generation efficiency in semiconductors by using longer wavelengths to suppress strong low-order multiphoton absorption and determine the optimum long pump wavelength limit where the suppression of multiphoton absorption in semiconductors GaP and GaAs is beneficial. I aimed to determine by numerical calculations the extent to which the suppression of higher order multiphoton absorption significantly benefits the terahertz generation efficiency by determining the optimum wavelength. Another aim was to investigate the effect of nonlinear refractive index on THz generation efficiency in semiconductors.

Scalable THz pulses with up to 3.9 μ J and a conversion efficiency of 0.3% have been reported in the ZnTe CG source [22]. However, growing large size ZnTe crystals is difficult due to microbubbles[22]. GaP and GaAs are feasible alternatives to ZnTe.

 The third objective was to extend the design of the scalable and efficient semiconductor contact grating to other semiconductors GaP and GaAs THz sources. I aimed to determine a set of design parameters by numerical calculations that yield the highest diffraction efficiencies based on rectangular and trapezoidal structures in GaP and GaAs contact gratings to give practical guidance for designing and fabrication of highly efficient THz pulse sources. Another aim was to investigate the effect of deviations from the perpendicular wall on diffraction efficiency.

The fourth objective was to investigate the possible ways of increasing the diffraction efficiencies of the GaAs and GaP as high as possible to even more than 90% at long infrared wavelengths. I aimed to enhance the diffraction efficiencies of the GaP and GaAs contact grating by adding an antireflective coating layer to the contact grating. Here, numerical investigation of three different ways of adding antireflective coatings were performed.

3. New Scientific Results

- According to the numerical calculations I performed for I. THz generation efficiency in GaP and GaAs crystals pumped in the wavelength range of 1 to 5 µm, I have shown that there is a strong benefit of suppression of strong multiphoton absorption in both GaP and GaAs. I have also shown a limit of long infrared wavelength pumping where the suppression of higher order multiphoton absorption (MPA) is beneficial. Beyond that limit, no benefit is associated with the suppression of MPA. In the case of GaP, such a limit is about 2 μm, corresponding to the cut-off wavelength of 5PA, whereas in GaAs, the limit is 3.85 µm, corresponding to the cut-off wavelength of 5PA. I have also shown that pumping at wavelengths that allows suppression of 2PA and higher order MPA, THz conversion efficiency of 0.9% and 1.14% can be achieved in GaAs and GaP, respectively [S1].
- II. According to my numerical calculations, using the modified model of THz generation, which took into account the nonlinear refractive index and its changes with the wavelength, besides other processes in the

semiconductors. For the first time, I have shown that the nonlinear refractive index causes a decrease in the THz generation efficiency and causes a shift of optimum pumping wavelengths to shorter values in semiconductors. The optimum wavelength was 2 μ m for GaP and 3 μ m for GaAs [S1].

- III. I have designed GaP CG and GaAs CG THz sources by numerical calculation that can be used to achieve a collinear THz generation scheme with which the THz energy can be scalable with pump spot size and available crystal sizes. The optimum CG design parameters, i.e., the groove spacing, groove depth and duty cycle, that yield the highest diffraction efficiency in the ±1 diffraction orders satisfying the velocity matching condition have been determined. Such GaP and GaAs CGs THz sources can be used to achieve single-cycle THz pulses with energies of μJ to mJ energy range [S1,S2].
- IV. Using numerical calculations, I have shown that the diffraction efficiencies of the GaP CG and GaAs CG can be increased from 69-75% to 88-90% by adding an appropriate antireflective coating on the CG profiles. The antireflective coatings minimize Fresnel losses due to reflection. In particular, I have shown that by adding three different antireflective coatings, namely, SiO₂ AR, adding

an AR of a material with a refractive index equal to the square root of the refractive index of the CG (GaP or GaAs) and adding a Norland optical adhesive 170 (NOA) AR, the diffraction efficiencies can significantly be enhanced [S2].

V. Due to the possible inaccuracy of the fabrication technique, the design parameters of the CGs may differ from the optimal values during fabrication. In order to achieve practical implementation, I have also shown that deviations of less than $\phi = 25^o$ (wall angle) from the perpendicular wall in the case of rectangular CG profiles, the diffraction efficiency loss is less than 5%. In contrast, beyond $\phi = 25^o$, the diffraction efficiency drops monotonically with an increase of wall angle. Therefore, during implementation, special attention should be taken not to exceed a wall angle of 25^o . I have also shown that the rectangular and trapezoidal profiles yield almost similar diffraction efficiencies [S2].

List of Publications

Publications related to the dissertation

- [S1] **N. M. Mbithi**, G. Tóth, Z. Tibai, I. Benabdelghani, L. Nasi, G. Krizsán, J. Hebling, and G. Polonyi, "Investigation of terahertz pulse generation in semiconductors pumped at long infrared wavelengths," J. Opt. Soc. Am. B, (2022).
- [S2] Z. Tibai, **N. M. Mbithi**, G. Almási, J. A. Fülöp and J. Hebling, "Design of semiconductor contact grating terahertz source with enhanced diffraction efficiency," Crystals, (2022).

Presentations

- [E1] **N. M. Mbithi**, G. Krizsán, G. Polónyi, Z. Tibai, J. Hebling, and J. A. Fülöp, "Highly efficient THz pulse generation in semiconductors utilizing contact-gratings technology," in *High Intensity Lasers and High Field Phenomena*, (Optical Society of America, 2020), JW1A. 13.
- [E2] **N. M. Mbithi**, G. Polónyi, Z. Tibai, G. Krizsán, L. Nasi, J. Hebling, J. A. Fülöp, Design of semiconductor contact-grating terahertz source for high energy terahertz pulse generation, MedPECS Medical Conference for PhD students and Experts of clinical Sciences, May 15 (2021).
- [E3] G. Krizsán, N. M. Mbithi, G. Polónyi, G. Tóth, M. Mechler, J. Hebling, and J. A. Fülöp, "Semiconductors as Highly Efficient Single and Multicycle Terahertz Sources," in *The European Conference on Lasers and Electro-Optics*, (Optical Society of America, 2019), cc p22.
- [E4] G. Krizsán, G. Polónyi, N. M. Mbithi, Z. Tibai, L. Pálfalvi, Gy. Tóth, L. Nasi, J. A. Fülöp and J. Hebling, Easily

- adaptable and scalable semiconductor THz pulse source, 46 th IRMMW-THz (2021).
- [E5] G. Polónyi, P. Nugraha, **N.M. Mbithi**, G. Krizsán, B. Monoszlai, M. Mechler, G. Tóth, J. Hebling, and J. Fülöp, "Highly Efficient Scalable Semiconductor Terahertz Sources," in 2019 44th International Conference on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz), (IEEE, 2019), 1-3.
- [E6] J. A. Fülöp, G. Polónyi, K. Gergö, N. M. Mbithi, P. S. Nugraha, G. Almási, L. Pálfalvi, Z. Tibai, G. Tóth, and J. Hebling, "Novel intense single-and multicycle THz sources," in 2020 45th International Conference on Infrared, Millimeter, and Terahertz Waves (IRMMW-THz), (IEEE, 2020), 1-2.
- [E7] J. A. Fülöp, G. Almási, G. Krizsán, N. M. Mbithi, M. I. Mechler, P. S. Nugraha, L. Pálfalvi, G. Polónyi, A. Sharma, and Z. Tibai, "Microstructured Intense THz Sources," in *Terahertz Science and Applications*, (Optica Publishing Group, 2019), TTh4D. 4.

Other publications

[S3] M. H. Abufadda, **N. M. Mbithi**, G. Polonyi, P. S. Nugraha, A. Buzady, J. Hebling, L. Molnar, and J. A. Fulop, "Absorption of Pulsed Terahertz and Optical Radiation in Earthworm Tissue and Its Heating Effect," J Infrared Millim Te **42**, 1065-1077 (2021).

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