UNIVERSITY OF PÉCS

Doctoral School of Physics

Nonlinear optics and spectroscopy program

Semiconductor-based terahertz sources with extreme high-efficiency

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1. Introduction and aims

The widespread availability of table-top laser sources triggered the development of various types of laser-driven pulsed terahertz (THz) sources. This development now enables to routinely provide THz pulses with unprecedented energies and peak electric and magnetic field strengths throughout the entire THz spectral range. Intense pulses at low terahertz frequencies of 0.1–2 THz are an enabling tool for nonlinear THz spectroscopy, for strong field control of matter [1], and for constructing compact particle accelerators, for enhancement of high-harmonic generation [2, 3], electron undulation [4], electron bunch acceleration [4, 5, 6] and for proton acceleration for hadron therapy [7, 8].

The low-frequency part of the THz spectrum (0.1 to 2 THz) is optimally fitting to such applications mentioned before. Up to now, optical rectification in lithium niobate (LiNbO3, LN) in combination with tilted pulse front pumping has been the most efficient source in this spectral range. However, the large pulse front tilt angle (63°) is disadvantageous for applications and makes the further increase of the THz energy challenging due to the limited interaction length [9], the imaging errors [10] and the nonlinear interaction between the pump and the THz [11], S4].

Contrary to LN, semiconductors, such as ZnTe or GaP, are widely used with collinear phase matching for optical rectification in the low-frequency THz range , although they were considered as less efficient for THz generation [57]. The highest THz energy reported from a semiconductor source was only 1.5 μJ [53]. The reason for the low efficiency was the smaller nonlinear coefficient and the strong two-photon absorption at the pump wavelength, associated with the free-carrier absorption at THz frequencies. Two-photon absorption can be avoided and free carrier absorption can be decreased if we use longer pump wavelengths. Therefore at longer pump wavelengths, typically requiring tilted pulse-front pumping, it is possible to suppress low-order multiphoton absorption. As a result, a higher pump intensity can be used and a higher THz generation efficiency can be expected.

My aim is to show that semiconductors pumped above the three-photon absorption edge are competitive with LN in efficiency and pulse energy. I would like to show that the realization of a new-type semiconductor-based THz source, called contact-grating, is possible with better properties for the applications. My aim is to show that the scaling-up of the THz generation efficiency is easier in semiconductors than in LN in the case of optimal pumping conditions.

2. Methods

In most of my simulations, the one-dimensional wave equation with the nonlinear polarization was solved in the spectral domain. This model takes into account the variation of the pump pulse duration with propagation distance due to material and angular dispersions, the absorption in the THz range due to phonon resonances and FCA, latter caused by MPA of the pump. For the pump, the projected propagation distance onto the THz propagation direction was used in order to account for the noncollinear phase matching in a TPFP scheme.

I have carried out a comparative study both numerically and experimentally in ZnTe pumping below and above the three-photon absorption edge and I could demonstrate the increase in efficiency as the low-order multi-photon absorption was suppressed [S1].

For pumping ZnTe at longer wavelength we need to tilt the pulse-front for phase-matching. Fortunately, the required pulse front tilt is less than half for semiconductors than for LN, therefore the angular dispersion is smaller and the reduced variation of pump pulse duration allows for longer effective length, with which the smaller nonlinear coefficient can be compensated.

I have carried out experiments with the first ZnTe based contact-grating THz source and demonstrated high efficiency and focusability [S3]. Here the grating structure is directly on the surface of the crystal with which it could substitute the entire tilted pulse front scheme. The incoming pump diffracts at the entry surface of the crystal, then it propagates with the right pulse front tilt to generate THz radiation. A symmetric arrangement, utilizing the two diffraction orders, can be used in a convenient collinear geometry with perpendicular incidence of the pump for excellent THz beam quality.

Based on the results of the experiments and simulations, I have given an estimation of the four-photon absorption coefficient of ZnTe, which was unknown in the literature [S1].

I have given practical guidelines through simulation data for the optimal pumping parameters of ZnTe and GaP THz sources, and finally I have shown that GaP is a versatile THz source which can be tailored both to single-cycle, and to multi-cycle THz applications [S2].

3. New Scientific Results

1. I have demonstrated both numerically and experimentally that ZnTe pumped at an infrared wavelength sufficiently long to suppress two-photon absorption (1450 nm) can have more than one order of magnitude higher efficiency ([26], 4•10-4). I have also shown that this efficiency can be further increased by 3.5 times (1.4•10-3) if we eliminate the three-photon absorption as well pumping at 1700 nm. [S1]

2. I have demonstrated the first ZnTe based, compact, monolithic, and alignment-free contact-grating THz source with high efficiency (0.3%) single-cycle THz pulses. Furthermore, I have shown that the THz beam can be focused near to the diffraction limit. This source is easy to up-scale to the mJ level THz pulse energies by increasing the pump spot size and energy. [S2]

3. With a high energy pumping source at 1700 nm, I was able to get 0.7% efficiency and 14 µJ THz pulse energy from a ZnTe prism in a conventional tilted-pulse-front pumping setup. The measured pulse energy is about ten times, and the efficiency is about 220 times higher than the previously reported results from ZnTe source. [S1]

4. With our experimental and numerical results I was able to give a (4±1)•10-5 cm5/GW3 estimation to the so far unknown four-photon absorption coefficient of ZnTe. [S1, S3]

5. In the case of ZnTe and GaP pumped at sufficiently long wavelengths to suppress two- and three-photon absorption I have shown the optimal pumping parameters for effective THz generation and maximal peak electric field. For maximum peak electric field in ZnTe 15 GW/cm2 pump intensity and 225 fs pump pulse duration are necessary, meanwhile in GaP these values are 20 GW/cm2 and 150 fs respectively. For maximum efficiency in ZnTe 15 GW/cm2 pump intensity and 150 fs pump pulse duration are necessary, and in GaP these values are 20 GW/cm2 and 100 fs respectively. These results give practical guidelines to prepare more effective semiconductor sources. [S3]

6. I have investigated the dependence of the waveform and the normalized spectra on the interaction length for fixed phase-matching frequencies of 2 THz, 3 THz and 4 THz in GaP for pumping at 1700 nm. I have demonstrated that a very broad spectrum can be generated in thinner (L < 5 mm) crystals, meanwhile in thicker (L > 5 mm) crystals with 3 – 4 THz phase-matching as the spectrum rapidly collapses the corresponding waveform evolves into a many-cycle pulse. As a consequence, GaP as a THz source can be tailored to the needs of the applications. [S3]

4. Related publications

[S1] Gy. Polónyi, B. Monoszlai, G. Gäumann, E. J. Rohwer, G. Andriukaitis, T. Balciunas, A. Pugzlys, A. Baltuska, T. Feurer, J. Hebling, and J. A. Fülöp, „High-energy terahertz pulses from semiconductors pumped beyond the three-photon absorption edge”Optics Express 24 (21), 23872-23882 (2016).

[S2] J. A. Fülöp, Gy. Polónyi, B. Monoszlai, G. Andriukaitis, T. Balciunas, A. Pugzlys, G. Arthur, A. Baltuska, and J. Hebling, “Highly efficient scalable monolithic semiconductor terahertz pulse source,” Optica, 3, 1075-1078 (2016).

[S3] Gy. Polónyi, M. I. Mechler, J. Hebling, and J. A. Fülöp,“Prospects of semiconductor terahertz pulse sources,” IEEE Journal of Selected Topics in Quantum Electronics, 23 (4), 1-8 (2017).

5. Other publications

[S4] Cs. Lombosi, Gy. Polónyi, M. Mechler, Z. Ollmann, J. Hebling, and J. A. Fülöp, „Nonlinear distortion of intense THz beams” New Journal of Physics 17, 083041 (2015).

6. References

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[7] A. Sharma, Z. Tibai, and J. Hebling, “Intense terahertz laser driven proton acceleration in plasmas,” Physics of Plasmas, vol. 23, no. 6, p. 063111, 2016.

[8] E. A. Nanni, W. R. Huang, K.-H. Hong, K. Ravi, A. Fallahi, G. Moriena, R. J. Dwayne Miller, and F. X. KÃ¤rtner, “Terahertz-driven linear electron acceleration,” vol. 6, p. 8486, Oct. 2015.

[9] J. A. Fülöp, L. Pálfalvi, M. C. Hoffmann, and J. Hebling, “Towards generation of mJ-level ultrashort THz pulses by optical rectification,” Opt. Express, vol. 19, pp. 15090–15097, Aug 2011.

[10] J. A. Fülöp and J. Hebling, Recent Optical and Photonic Technologies, ch. Applications of Tilted-Pulse-Front Excitation. 01 2010.

Available:https://www.intechopen.com/books/recent-optical-and-photonic-technologies/applications-of-tilted-pulse-front-excitation.

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