

Intense Terahertz Sources for Nonlinear Interactions

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Abstract

The present status and the prospects of efficient pulsed terahertz (THz) sources based on optical rectification in lithium niobate and semiconductors, pumped by femtosecond pulses with tilted intensity front, will be discussed. Applications range from nonlinear THz spectroscopy and multispectral imaging to electron and proton acceleration.

1. Introduction

THz pulses with high energy and field strength in different parts of the THz spectrum are needed for resonant and nonresonant control over ionic motion, spin dynamics, bound and free electrons [1]. The low-frequency part of the THz spectrum (0.1 to 2 THz) is particularly suitable for applications involving charged particle beams, because the longer wavelength of THz pulses compares well with typical sizes of particle beams and bunches. Compact THz-driven particle accelerators with unprecedented flexibility can become important for free-electron lasers, materials science, and could revolutionize medical therapy with x-ray, electron, or proton beams [2, 3].

Table-top femtosecond laser sources are now routinely used to generate THz pulses with unprecedented energy and peak electric and magnetic fields. Different parts of the low- (~0.1–2 THz) and mid-frequency (~2–20 THz) bands of the THz range can be accessed by optical rectification (OR) in LiNbO₃ (LN) [4], semiconductors [5–7], or organic materials [8]. Here we present cutting-edge THz sources based on LN and semiconductors, and their prospects.

2. High energy and high average power LiNbO₃ THz sources

In the low-frequency THz range OR in LN, using tilted-pulse-front pumping (TPFP) for phase matching [9], has been providing the highest THz pulse energies [4] and field strengths [10]. To achieve >0.4 mJ THz pulse energy it was essential to use longer (≥ 0.5 ps) pump pulses [4], which enabled to increase significantly the interaction length for THz generation. While it is expected that mJ or even multi-mJ pulse energies will be available with this

technology, also its limitations became apparent. The main reason is the large pulse-front tilt angle ($\sim 63^\circ$) required for phase matching. This requires sophisticated approaches for further optimization of TFPF in LN [11].

THz nonlinear spectroscopy requires μJ THz pulse energies, which can typically be provided by mJ pump pulse energies at kHz repetition rates. However, in many cases intense or moderately intense THz pulses at MHz repetition rates pumped by a compact fiber laser source would be desired. Besides spectroscopic applications, multispectral imaging and security could also benefit from such a high average power THz source. However, TFPF THz sources with μJ or sub- μJ pump energies suffer from very low efficiencies because THz absorption of LN and a small pump spot size limit the interaction length.

To circumvent this limitation, an absorption-reduced waveguide (ARWG) THz source was introduced [12], where both the optical pump as well as the generated THz radiation are guided such that velocity matching in a TFPF scheme is fulfilled (Fig. 1). Efficient THz generation is enabled by the THz waveguide cladding with orders of magnitude smaller absorption coefficient in the THz range than that of the LN core. More than one order of magnitude increase in the conversion efficiency is predicted.

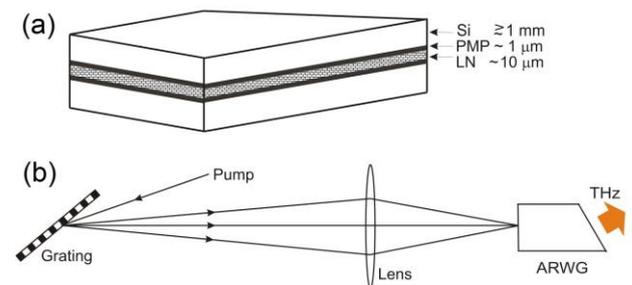


Figure 1. (a) The ARWG structure. (b) The ARWG THz source with TFPF (top view).

3. Efficient semiconductor THz sources

The potential of semiconductor nonlinear optical materials for high-energy high-field THz pulse generation by OR has been recently recognized [5–7]. Whereas

pumping OR in ZnTe at 0.8 μm , near its collinear phase-matching wavelength, resulted in maximum 1.5 μJ THz pulse energy at 3.1×10^{-5} efficiency [13], pumping at 1.7 μm wavelength recently resulted in more than two orders of magnitude higher efficiency, as high as 0.7%, and 14 μJ THz pulse energy [7]. The reason for the enormous increase in efficiency was the elimination of lower-order (2nd- and 3rd-order) multiphoton pump absorption at the longer pump wavelength and the associated free-carrier absorption in the THz range.

At such longer infrared pump wavelengths TFPF has to be used for phase matching in semiconductors. An important advantage is the much smaller required pulse-front tilt angle in semiconductors (typically below 30°) than that in LN. A smaller tilt angle enables a larger effective length for THz generation. This can help to compensate for the smaller nonlinear coefficient of semiconductors. Secondly, a smaller tilt angle significantly reduces the spatial nonuniformity of the interaction length for THz generation, and consequently that of the THz beam. The potentially much better spatial homogeneity enables an easier increase of the pumped area and the THz energy.

The small tilt angle is also advantageous for the realization of a contact-grating (CG) THz source (Fig. 2). Such a CG source has recently been demonstrated in ZnTe [6], resulting in the generation of 3.9 μJ THz pulses with up to 0.3% efficiency. Importantly, the CG technology enables to practically eliminate spatial nonuniformity of the interaction length, which leads to a straightforward scalability of the THz energy and excellent THz beam profile and focusability. CG fabrication technology enables grating sizes on the 5-cm scale. It is expected that THz pulses with >1 mJ energy and >20 MV/cm electric field strength will be achievable with less than 200 mJ pump energy from a CG source. Such pump pulses in the wavelength range of 1.7 μm to 2.5 μm , though not available presently, are definitely feasible in the near future from optical parametric amplifiers or infrared laser technology. Efficient monolithic semiconductor THz sources can become a key technology for compact THz particle accelerators and other THz high-field applications.

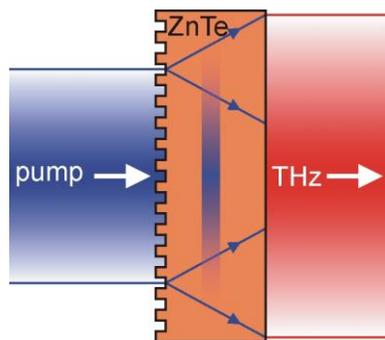


Figure 2. Monolithic contact-grating THz source with a collinear pumping geometry in ZnTe [6].

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7. References

1. T. Kampfrath, K. Tanaka, and K. A. Nelson, "Resonant and nonresonant control over matter and light by intense terahertz transients," *Nat. Photonics*, **7**, Aug. 2013, pp. 680–690.
2. L. Pálfalvi, J. A. Fülöp, Gy. Tóth, and J. Hebling, *Phys. Rev. ST Accel. Beams* **17**, Mar. 2014, Art. no. 031301, doi: 10.1103/PhysRevSTAB.17.031301.
3. E. A. Nanni, W. R. Huang, K.-H. Hong, K. Ravi, A. Fallahi, G. Moriema, R. J. D. Miller, and F. X. Kärtner, *Nat. Commun.* **6**, Oct. 2015, pp. 8486, doi: 10.1038/ncomms9486.
4. J. A. Fülöp, Z. Ollmann, Cs. Lombosi, C. Skroboł, S. Klingebiel, L. Pálfalvi, F. Krausz, S. Karsch, and J. Hebling, "Efficient generation of THz pulses with 0.4 mJ energy," *Opt. Express*, **22**, Aug. 2014, pp. 20155–20163, doi: 10.1364/OE.22.020155.
5. F. Blanchard et al., "Terahertz pulse generation from bulk GaAs by a tilted-pulse-front excitation at 1.8 μm ," *Appl. Phys. Lett.*, **105**, Dec. 2014, Art. no. 241106.
6. J. A. Fülöp et al., "Highly efficient scalable monolithic semiconductor terahertz pulse source," *Optica*, **3**, Sep. 2016, pp. 1075–1078.
7. 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," *Opt. Express*, **24**, Oct. 2016, pp. 23872–23882, 10.1364/OE.24.023872.
8. C. Vicario et al., "Generation of 0.9-mJ THz pulses in DSTMS pumped by a Cr:Mg₂SiO₄ laser," *Opt. Lett.*, **39**, Dec. 2014, pp. 6632–6635.
9. J. Hebling, G. Almási, I. Z. Kozma, and J. Kuhl, "Velocity matching by pulse front tilting for large-area THz-pulse generation," *Opt. Lett.*, **10**, Oct. 2002, pp. 1161–1166.
10. M. Sajadi, M. Wolf, and T. Kampfrath, "Terahertz-field-induced optical birefringence in common window

and substrate materials,” *Opt. Express*, **23**, Nov. 2015, pp. 28985–28992.

11. L. Pálfalvi, Z. Ollmann, L. Tokodi, J. Hebling, “Hybrid tilted-pulse-front excitation scheme for efficient generation of high-energy terahertz pulses,” *Opt. Express* **24**, April 2016, pp. 8156–8169, doi: 10.1364/OE.24.008156.

12. L. Pálfalvi, J. A. Fülöp, and J. Hebling, “Absorption-reduced waveguide structure for efficient terahertz generation,” *Appl. Phys. Lett.*, **107**, Dec. 2015, Art. no. 233507, doi: 10.1063/1.4937347.

13. F. Blanchard et al., “Generation of 1.5 μ J single-cycle terahertz pulses by optical rectification from a large aperture ZnTe crystal,” *Opt. Express*, **15**, Oct. 2007, pp. 13212–13220.