UNIVERSITY OF PÉCS
Doctoral School of Physics
Nonlinear optics and spectroscopy program

Investigations into high energy THz and ultra-broadband IR radiation from organic salt crystal emitters

PhD thesis

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1. Preliminaries

The importance of THz radiation has been significantly increased over the recent decades due to the developing laser- and THz-science. There are plenty of planned and realized applications of THz radiation used in several fields including industry [1], security and defense [2], medicine [3] and scientific research such as control over matter [4] or electron acceleration [5]. THz source development is the base of these studies and aims to gain the ability to create simple, easily available sources with high level of control over their parameters. Source development is still an ongoing, intensely developing part of THz science.

Organic salt crystals (OSC)s is group of crystals consist of organic compounds and some of them own extremely high nonlinear properties (e.g. DAST 490±90 pm/V at 1535 nm [6]). They can be used for both THz generation via optical rectification and THz detection via electro-optic effect. The most well-known OSC types are the DAST, DSTMS, OH1, HMQ-TMS and BNA crystals. Although some of their properties are similar, they possess different characteristics. Their typical size is a couple of mm with few hundred µm thickness. They are strongly birefringent crystals, thus collinear phase matching for THz generation is possible over different IR wavelengths. They have several strong THz absorption peaks due to their complex molecule structure [7]. The generated THz spectrum usually covers the range starting from 0.1 THz up to 3 THz or 5 THz but 15 THz has been also demonstrated [8] [9] [10]. The generated THz pulse energies are usually in the tens of µJ range as well an outstanding 0.9 mJ [11] has been also reported. The typical conversion efficiencies are around 1-3% at certain pump wavelengths and pump intensities [9] [11].

2. Objects and methods

Just like any other source, OSCs are also suffer from different handicaps. The primary goal was to experimentally test alternative approaches which can solve their bottlenecks or exploit their unused capabilities. The generated THz and influenced IR radiation from DAST and DSTMS OSCs have been investigated and characterized in the following scenarios:

Unconventional pumping wavelengths: The complexity and difficult accessibility of the usual high-energy pump sources around 1500 nm has resulted in the use of a common, Ti:sapphire pumping source around 800 nm. The results are compared with other THz sources pumped by this easily accessible pumping source. This is complemented with calculations about the expected THz spectrum close to the optimal pumping wavelength

Expanded crystal surface: The comparison of a partitioned DSTMS, a mosaic set of smaller oriented crystals with a single crystal. This is inspired by the bottlenecks in production and manufacturing procedures of single crystals.

Recycling of pump beam: The examination of the IR pump passed through a DAST crystal as a potential IR source with extreme spectral bandwidth.

3. Thesis statements

1) It was demonstrated that the conversion efficiency of DAST ($4x10^{-5}$) and DSTMS ($6x10^{-5}$) organic salt crystals are comparable to other optical rectification based THz sources such as ZnTe ($3.1x10^{-5}$) or $LiNbO_3$ ($11.6x10^{-5}$) crystals when they are pumped at Ti:sapphire wavelengths. [12]
2) It was shown that the spectra of the generated THz radiation extends towards the higher frequencies when the DAST or DSTMS organic salt crystals are pumped around the optimal 680-740 nm wavelengths instead of Ti:sapphire wavelengths due to the spectrally broader phase-matching properties. [12]

3) It was experimentally proved that individual THz beamlets from a partitioned crystal — mosaic arrangement of smaller oriented crystal segments — are capable of a high level of constructive interference, which was manifested in a near single-cycle THz pulse waveform and a Gaussian like smooth focal spot with beam quality factor of $M^2 \approx 1.8$. Therefore, a THz beam generated in a partitioned crystal and a THz beam generated in a single crystal have technically identical spatio-temporal beam quality around their focal planes. [13]
4) It was experimentally shown that the passing infrared pump beam in a DAST organic salt crystal can undergo extreme spectral broadening and achieve ~1.5-octave broad spectrum covering the range between 1.2 µm and 3.5 µm. [14]

4. Publication list

1. Publications in the topic of thesis

B. Monoszlai, C. Vicario, M. Jazbinsek, C.P. Hauri: “High energy terahertz pulses from organic crystals: DAST and DSTMS pumped at Ti:sapphire wavelength”, *Optics Letters, Vol. 38, Issue 23, pp. 5106-5109 (2013)*

Carlo Vicario, **B. Monoszlai**, and Christoph P. Hauri: “GV/m Single-Cycle Terahertz Fields from a Laser-Driven Large-Size Partitioned Organic Crystal”, *Phys. Rev. Lett. 112, 213901, (2014)*

C. Vicario, **B. Monoszlai**, G. Arisholm, and C.P. Hauri: “Generation of 1.5-octave intense infrared pulses by nonlinear interactions in DAST crystal”, *Journal of Optics, Vol. 17, 094005, (2015)*

2. Non-refereed conference abstracts in the topic of thesis

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3. Publications in other

C. Vicario, **B. Monoszlai**, Cs. Lombosi, A. Mareczko, A. Courjaud, J. A. Fülöp, C. P. Hauri: "Pump pulse width and temperature effects in lithium niobate for efficient THz generation", *Optics Letters, Vol. 38, Issue 24, pp. 5373-5376 (2013)*

P. N. Juranic, A. Stepanov, P. Peier, C. P. Hauri, R. Ischebeck, V. Schlott, M. Radovic, C. Erny, F. Ardana-Lamas, **B. Monoszlai**, I. Gorgisyan, L. Patthey, R. Abela: "A scheme for a shot-to-shot, femtosecond-resolved pulse length and arrival time measurement of free electron laser x-ray pulses that overcomes the time jitter problem between the FEL and the laser", *JINST, Vol. 9, pp. P03006 (2014)*

A. Trisorio and M. Divall and **B. Monoszlai** and C. Vicario and C. P. Hauri: "Intense sub-two-cycle infrared pulse generation via phase-mismatched cascaded nonlinear interaction in DAST crystal", *Optics Letters, Vol. 39, Issue 9, pp. 2660-2663 (2014)*

P. N. Juranić, A. Stepanov, R. Ischebeck, V. Schlott, C. Pradervand, L. Patthey, M. Radović, I. Gorgisyan, L. Rivkin, C. P. Hauri, **B. Monoszlai**, R. Ivanov, P. Peier, J. Liu, T. Togashi, S. Owada, K. Ogawa, T. Katayama, M. Yabashi, and R. Abela: "High-precision x-ray FEL pulse arrival time measurements at SACLA by a THz streak camera with Xe clusters", *Optics Express, Vol. 22, Issue 24, pp. 30004-30012 (2014)*

C. Vicario**, B. Monoszlai**, M. Jazbinsek, S.-H. Lee, O-P. Kwon and C. P. Hauri: "Intense, carrier frequency and bandwidth tunable quasi single-cycle pulses from an organic emitter covering the Terahertz frequency gap", *Scientific Reports 5, Article number: 14394 (2015)*

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 Vol. 3, Issue 10, pp. 1075-1078 (2016)

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 Vol. 24, Issue 21, pp. 23872-23882 (2016)

4. Non-refereed conference abstracts in other topics

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5. References

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