Programmable Pulse Bursts for Terahertz Spectroscopy by using Bolometric only Detection

Rupali Sharma[1], Matthias Schneller[1], Vinzenz Stummer[1], Audrius Pugzlys[1,2], Andrius Baltuska[1,2],
Jozsef A. Fülöp[3], Abhishek Gupta[3], Vineet Gupta[3], Ashutosh Sharma[3]
[1]TU Wien, Photonics Institute, A-1040 Vienna, Austria
[2]Center for Physical Sciences & Technology, LT-02300 Vilnius, Lithuania
[3]ELI-ALPS Research Institute, ELI-HU Nonprofit Ltd.Szeged, Hungary

***Abstract*— The generation of high-energy Terahertz (THz) pulses via optical rectification (OR) in Lithium-Niobate and organic crystals serves as an attractive route for applications such as nonlinear THz spectroscopy, single-shot THz imaging, and THz radar. Being generated by broadband near-infrared (NIR) driver pulses, OR-generated pulses typically feature sub-THz bandwidths, or higher, often substantially broader than narrow resonances of the sample. We report on our recent progress in utilizing the generation of microjoule-level THz pulses, by driving OR with a burst of closely spaced NIR pulses. By adjusting the pulse number and spacing in the burst we control over central THz frequency of equidistant narrowband spectral peaks. The spectral tunability in combination with a conventional bolometric detection allows selective detection of narrowband resonances of particular organic samples.**

I. INTRODUCTION

In recent decades, the generation of high-energy THz radiation using ultrashort laser pulses has advanced significantly, enabling THz pulse energies well beyond the microjoule level [1]. These developments allow to not only perform linear THz spectroscopy on a point sample, but also to drive its nonlinear response and to extend linear THz spectroscopy for THz imaging applications. In particular, Lithium Niobate (LN), when used in a tilted-pulse-front pumping (TPFP) configuration for efficient phase-matching, has demonstrated multiple percent THz conversion efficiency when driven by near-infrared (NIR) femtosecond pulses around 1 μm [2].

Applications in spectroscopy and strong-field control of matter demand shaped strong-field THz waveforms along with high field strengths. Despite many significant advancements in high-energy THz sources, current systems often lack capabilities in spectral shaping so far. Under phase-matched OR conditions, the temporal profile of the generated THz waveform is governed by the intensity envelope of the driving NIR pulse. A pulse train with uniform sub-picosecond spacing produces a frequency-domain spectrum with equidistant peaks, which are consequently transferred to the spectral profile of the THz signal generated from such a source. In this contribution, we present details of a tunable, narrowband THz generation system driven by burst-mode pump source and demonstrate its application in linear THz absorption spectroscopy using solely a frequency-insensitive detector.

II. RESULTS

Experiments were performed with a home built cryogenically cooled Yb:CaF_2 laser amplifier capable of generating up to 7 mJ, 250 fs pulses at 1 kHz repetition rate. The pump source employs a chirped-pulse amplification (CPA) scheme for generating bursts of NIR pulses with picosecond spacing. It is based on programmable amplitude and phase shaping at the low Megahertz oscillator repetition rate with a Vernier-effect burst formation in a regenerative amplifier [3]. The latter allows conversion of the initial nanosecond spacing of oscillator pulses to picoseconds, corresponding to THz intraburst frequencies. By active stabilization of intraburst spectral modes, interferometric stability and efficient energy extraction in the regenerative amplifier is ensured. The amplified NIR bursts are used to generate tunable THz pulses by OR using TPFP scheme in a LN crystal.

As shown in Fig.1, we generate a band of continuously tunable THz frequencies with narrow bandwidth. These spectra are obtained by taking Fourier transform of linear autocorrelation traces of the generated THz signal measured by a Michelson interferometer. The continuously-tunable intraburst repetition rate is determined by the inverse pulse spacing $1/\Delta t$, translating into the lowest-order THz frequency,

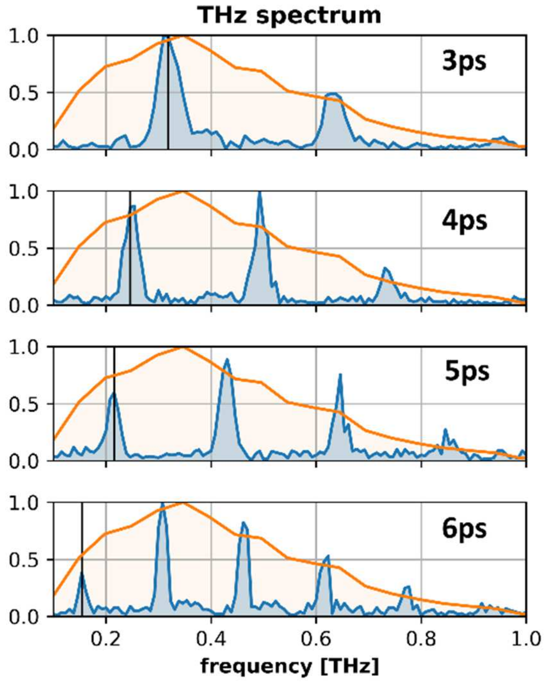

Fig. 1. Shaping of THz bursts: THz burst spectra generated from OR in LN in a TPFP scheme with 6 pulses and a varying pulse spacing Δt of 3, 4, 5 and 6 ps. The orange overlay indicates the THz spectrum of a single pulse. The black line indicates the tunable narrowband peaks at (top to bottom) 0.314 THz, 0.251 THz, 0.214 THz, 0.154 THz. The linewidth for Δt =3 ps case is 0.045 THz [3].

whereas the bandwidth Δf of the generated THz peak scales inversely to the product of pulse number and spacing $1/(N \cdot \Delta t)$.

Relying on the developed burst source, we aim to demonstrate linear THz absorption spectroscopy by using frequency-insensitive bolometer detectors. This is possible because the THz radiations generated from OR of the NIR burst source is narrow-band and tunable. Since bolometers integrate energy over all incident frequencies, we employ controlled modulation of burst modes to isolate specific frequency components. By systematically varying the burst pulse spacing, we record multiple N integrated measurements as outlined in Fig.2. We assume a given set point f_m of the fundamental THz peak, i.e. the THz spectral peak with the lowest center frequency.

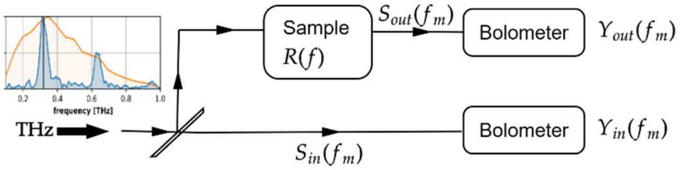

Fig. 2. Schematic representation of the bolometric only measurement of the absorption spectrum.

The spectrum of burst $S_{in}(f)$ interacts with the sample with the response function $R(f)$ and the output spectrum $S_{out}(f)$ after absorption from the sample is such that,

$$R(f) = \frac{S_{out}(f)}{S_{in}(f)}.$$

The measured signal Y_{out} from a frequency-insensitive detector can be expressed as,

$$Y_{out}(f) = \int S_{out}(f)df.$$

In order to get a spectroscopic response of the system with a required resolution Δf over a bandwidth BW, we take N measurements such that N $\geq \frac{BW}{\Delta f} = M$. We can obtain $S_{in}(f)$ and $S_{out}(f)$ for M unknown spectral components and obtain the response function.

For demonstrating controlled narrowband tunability, we choose α-lactose as a sample which features in the frequency range below 1 THz only one isolated absorption peak around $f_o = 0.55$ THz with a linewidth of about 20 GHz [4] which falls into the spectral bandwidth of THz generated in LN by TPFP as it is shown in the Fig.3. Due to the finite frequency-domain support of the LN-generated pulses (see Fig. 1), we can neglect any absorption at the sample of higher-order burst peaks and only consider absorption around the resonance frequency f_0 of α-lactose. In combination with the input reference measurement, we are able to exclude the higher-order peak contributions to the bolometric signal.

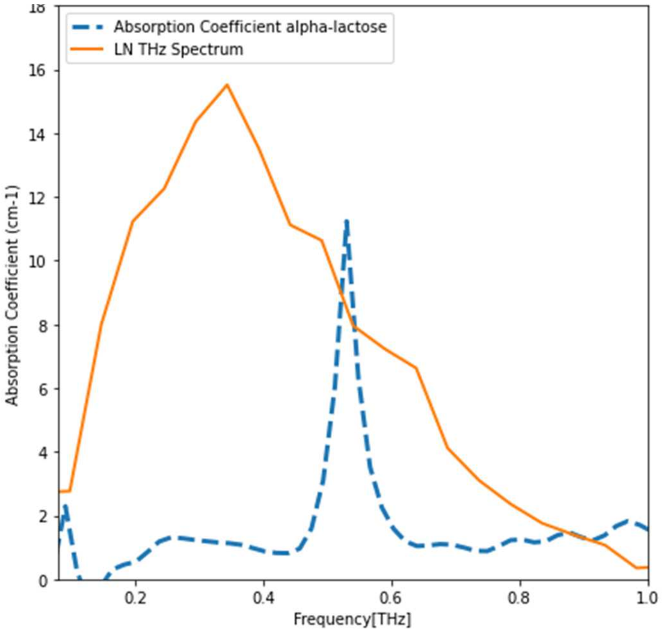

Fig. 3. Absorption spectrum of α-lactose measured by Elecro-Optic sampling. Absorption peaks at ~ 0.5 THz. By a solid line is shown the THz spectrum generated in LN by TPFP in the case of a single pulse pumping.

The linewidth of the fundamental peak is given by the inverse burst duration. Thus, for a frequency resolution sufficient to measure absorption features typical of THz samples, a scalable pulse number is required. Recently, we succeeded in the generation of frequency-scalable NIR driver bursts consisting of 40 pulses which results in 41.4 MHz linewidth [5] and is an indication of the spectral resolution. Thus, the burst-mode is a promising route for novel methods for spectral shaping of high-energy THz waveforms.

REFERENCES

[1] J. A. Fülöp, S. Tzortzakis, and T. Kampfrath, "Laser-Driven Strong-Field Terahertz Sources," *Advanced Optical Materials*, vol. 8, no. 3, p. 1900681, 2020, doi: 10.1002/adom.201900681.

[2] J. A. Fülöp, L. Pálfalvi, G. Almási, and J. Hebling, "Design of High-Energy Terahertz Sources Based on Optical Rectification," *Opt. Express, OE*, vol. 18, no. 12, pp. 12311–12327, 2010, doi: 10.1364/oe.18.012311.

[3] V. Stummer *et al.*, "Programmable Generation of Terahertz Bursts in Chirped-Pulse Laser Amplification," *Optica*, vol. 7, no. 12, p. 1758, 2020, doi: 10.1364/optica.403184.

[4] R. Damari *et al.*, "Strong Coupling of Collective Intermolecular Vibrations in Organic Materials at Terahertz Frequencies," *Nat Commun*, vol. 10, no. 1, p. 3248, 2019, doi: 10.1038/s41467-019-11130-y.

[5] V. Stummer *et al.*, "Frequency-mode-stable regenerative amplification at terahertz burst rates," *APL Photonics*, vol. 9, no. 3, p. 036116, 2024, doi: 10.1063/5.0167721.