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A simple optical spectral calibration technique for pulsed THz sources

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Abstract: We have quantified the sensitivity of a simple method to measure the frequency spectrum of pulsed terahertz (THz) radiation. The THz pulses are upconverted to the optical regime by sideband generation in a zinc telluride (ZnTe) crystal using a *continuous wave* (cw) narrow-bandwidth near-infrared laser. A single-shot spectral measurement of sideband pulses with a high resolution spectrometer directly provides the spectral information of the THz pulses without the need of adjustable elements in the detection setup. This method has been applied at the free electron laser FELIX, where, for a wavelength of 150 μm (2 THz), pulse trains of 5 μs duration with an integrated energy of 800 nJ, as well as single pulses with an energy as low as 13 nJ could be characterized on a single-shot basis.

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1. Introduction

Spectral characterization of pulsed terahertz (THz) sources is not straightforward. A grating spectrometer with a suitable detector (e.g. a pyroelectric detector or a helium-cooled bolometer) can be employed, but is not very practical for wavelengths longer than 500 μm (frequencies smaller than 0.6 THz) due to the increasing grating size and corresponding increase in size of the spectrometer. Alternatively, Fourier-transform spectrometers can be used where the spectrum is retrieved from an interferogram, although single-shot detection of pulsed sources is not straight-forward.

Ultrashort, broad bandwidth pulses are often characterized in THz time-domain spectroscopy (THz-TDS) setups, which use femtosecond optical laser pulses to sample the temporal profile of the THz electric field in a photoconductive antenna or an electro-optic crystal [1, 2]. Single-shot versions of this technique have been developed for applications where the THz pulses are fluctuating (in intensity, spectral width or temporal profile), see e.g. Ref. [3] and references therein. Although very powerful, these techniques require ultrafast (sub 100 fs) probe laser pulses, and are not suited for the characterization of long THz pulses. The clear advantage of the THz-TDS detection techniques, however, is that there is only a single component, the photoconductive antenna or electro optical crystal, which has to operate in the THz domain; all other components of the "THz detector" (such as lenses, photo-diodes, CCD arrays) are standard components operating in the visible and near-infrared spectral region.

The same advantage holds for techniques which upconvert the THz radiation to the optical or near-infrared range. Parametric upconversion or difference-frequency generation (DFG) of continuous wave (cw) and pulsed THz radiation has been demonstrated with high power pulsed lasers as pump sources. A variety of crystals has been used such as GaSe [4, 5], GaAs [6], and ZnGeP₂ [7], which all need angle tuning to optimize phase matching in order to obtain a sufficiently high conversion efficiency for a broad THz wavelength range. These crystals are therefore not suited for a THz spectrometer since, in general, the frequency of the THz radiation is not known *a priori*.

The electro-optic crystals GaP and ZnTe, which are frequently used for broadband electro-optic THz detection in THz-TDS experiments, do not require angle-tuning [8–11] and are therefore well suited for application in a broadband THz upconversion spectrometer. Sidebands are generated at frequencies $\nu_{\text{carrier}} \pm N \cdot \nu_{\text{THz}}$, with $N = 0, 1, 2, \dots$ and ν_{carrier} the frequency of the optical or near-infrared (NIR) laser. Simultaneous measurement of the carrier and sideband frequency with a grating spectrometer and CCD array directly provides the spectrum of the THz pulse. With a pulsed Ti:Sapphire laser (wavelength 780 nm, pulse duration 3 ps) and a ZnTe crystal, Jamison et al. [10] measured 2 THz radiation (wavelength 150 μm , bandwidth 2.4 μm) generated by a Free Electron Laser (FEL). Additionally, they demonstrated the broad-

band capabilities of the upconversion spectrometer by single-shot measurements of unipolar sub-picosecond THz pulses where the THz pulse is the Coulomb field of a single-relativistic electron bunch [10].

In this article we quantify the performance of a simple, high-resolution THz spectrometer based on upconversion in a ZnTe crystal. Instead of a high power pulsed optical pump laser, we use a narrow bandwidth cw laser operating at 780 nm. A similar approach, although using a GaP crystal, has recently been reported by Zaks et al. [9] for spectral calibration of the long pulsed (approximately 5 μ s duration) mm-wave FEL at UCSB. Here, we demonstrate the capabilities of such an upconversion spectrometer for short pulsed THz radiation. Measurements in the 100–220 μ m (1.3–3 THz) spectral range have been performed at the Free-Electron Laser for Infra-red eXperiments (FELIX) [12], where the performance of the upconversion spectrometer can be compared with direct detection using a far-infrared grating spectrometer. As we show that the upconversion scheme is extremely sensitive when applied for pulsed THz FELs, we are convinced that it can be applied for other pulsed THz sources as well.

2. Experimental setup

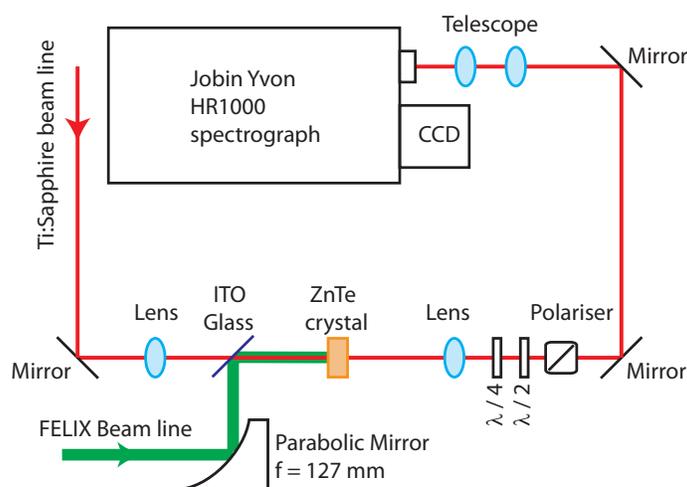


Fig. 1. Schematic representation of the experimental setup.

The setup employed for the experiments is schematically indicated in Fig. 1. The THz source for the test of the spectral calibration scheme is FELIX, operating in its longest wavelength region between 100 and 220 μ m (1.3–3 THz). The pulse structure of FELIX consists, as for most Radio Frequency (RF) accelerator based light sources, of macro-pulses and micro-pulses. The macro-pulses have a repetition rate of 10 Hz and a pulse duration of 5 μ s. Each macro-pulse consists of a 1 GHz train of Fourier-transform limited micro-pulses of 10–50 ps duration. We used macro-pulse energies of up to 5 mJ just before the ZnTe crystal, corresponding to a micro-pulse energy on the order of 1 μ J. Spectral calibration of FELIX is performed by coupling a fraction of the output into a 390 mm focal length grating-spectrometer (VM-504, Acton Research Corporation) equipped with an array of 48 pyro-electric detectors, yielding direct detection of the spectral profile.

For upconversion, the THz beam is focused by a parabolic mirror ($f=127$ mm) and reflected by an Indium Tin Oxide (ITO) coated quartz plate towards a 4 mm thick (110) oriented ZnTe crystal. The NIR radiation is produced by a cw single-mode Ti:Sapphire laser (Sirah Matisse-

TX) providing typically 200 mW of output at 780 nm with an approximately 1 MHz linewidth. The output of the Ti:Sapphire laser is frequency calibrated to 3 GHz absolute accuracy using a wavemeter (HighFinesse WS-5). The output of the Ti:Sapphire laser is focused by a 150 mm focal length lens in the ZnTe crystal. The ITO coated plate acts as a dichroic mirror, transmitting the NIR light without significant losses. The output power of both THz and NIR laser can be controlled by calibrated attenuators.

The polarization direction of the NIR and the THz beams are parallel, maximizing the sideband intensity [10]. As the polarization of the generated sidebands is perpendicular to the polarization of the NIR beam, the carrier can easily be suppressed by several orders of magnitude with a Glan-Laser polarizer while preserving the sideband intensity. A half-wave plate minimizes the transmission of the NIR carrier beam through the polarizer. The carrier laser intensity is further reduced by a quarter-wave plate located after the ZnTe crystal, removing the residual birefringence of the crystal [3, 13].

The transmitted light is dispersed in a grating spectrometer (Jobin Yvon HR1000) and imaged on a gated, image-intensified 512x512 pixel CCD array (Princeton Instrument PI-MAX) at the exit of the spectrometer. The gate pulse of the CCD detector exactly coincides with start and end of the FELIX macro-pulse. The output of the well-calibrated Ti:Sapphire laser is used to calibrate the dispersive axis of the CCD images. The spectral calibration of the spectrometer setup is performed by stepping the frequency of the Ti:Sapphire laser while keeping the CCD camera position and grating angle fixed. Prior to the THz upconversion measurements the Ti:Sapphire laser is again fixed to the original frequency around 780 nm.

The sideband intensity is maximized for a THz wavelength of 151 μm (66 cm^{-1}) by optimizing the ZnTe crystal orientation (rotation around the propagation direction of the co-linear laser beams), the overlap of the two foci, and the position of the crystal along the foci.

3. Experimental results and analysis

Figure 2 shows spectra obtained using the upconversion scheme for three different wavelengths of FELIX. It is explicitly mentioned that these spectra are recorded without any change to crystal orientation or spectrometer settings demonstrating the simplicity and flexibility of the detection scheme. The peak at the origin of the frequency axis is the remaining NIR laser signal. The width of this peak determines the instrument resolution of the upconversion spectrometer to be about 0.2 cm^{-1} , limited by the dispersion of the grating spectrometer and CCD pixel size to almost equal extent.

The upconversion spectral calibration scheme is compared with the FELIX calibration scheme using direct THz detection with the far-infrared grating spectrometer. The inset to Fig. 2, which includes the data of the three spectra presented in the main panel, shows the near-perfect match between the spectral calibration using the upconversion spectrometer and the far-infrared grating spectrometer. The solid red line indicates the perfect calibration curve.

The use of the rather thick (4 mm) ZnTe crystal leads to very efficient sideband generation for frequencies up to 3 THz (see Fig. 2). On first thought, a thicker crystal will enhance the efficiency of sideband generation by increasing the interaction length of the THz and optical waves. However, increasing the thickness also leads to higher absorption losses and increased phase mismatch between optical and THz waves, thereby limiting the interaction length to less than the crystal thickness [14, 15]. For the three THz frequencies displayed in Fig. 2, the interaction length is approximately 4 mm (for 1.4 THz / 46 cm^{-1}), 2 mm (2 THz / 66 cm^{-1}), and 0.5 mm (2.9 THz / 95 cm^{-1}) [14, 15].

The sideband signal is sensitively detected with the spectrometer / CCD combination. With all attenuators (38 dB, or an attenuation factor of 6310) in the THz beam - resulting in 800 nJ / macro-pulse (or 160 pJ / micro-pulse) at 151 μm - and using only 20 mW of the Ti:Sapphire

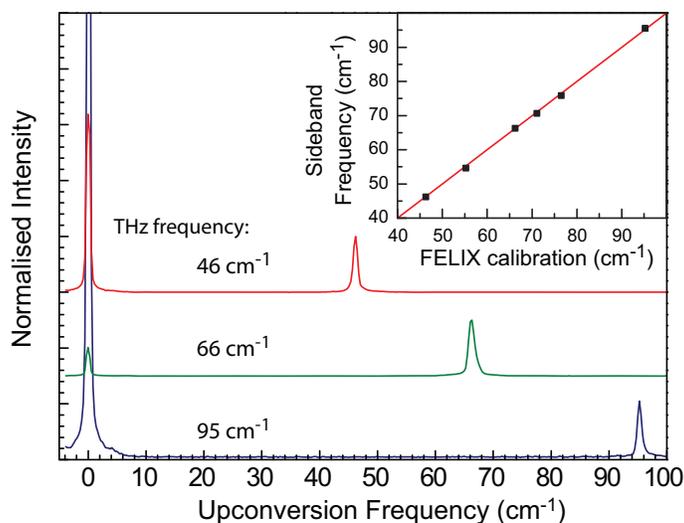


Fig. 2. Upconversion spectra recorded for three THz frequencies. The frequency of the near-infrared laser is defined as zero, such that the THz frequency can be directly read. The individual spectra are normalized to the maximum of the sideband signal and are vertically offset for readability. The FEL intensity for the upper and lower spectrum was smaller, thus leading to the apparent increase of the carrier laser signal. The spectra are all recorded at a NIR laser intensity of 20 mW and full THz pulse energy. The inset shows the central frequency obtained using upconversion against direct calibration with a Far-Infrared grating spectrometer.

laser, the generated difference frequency band could at maximum detector gain still be detected on a single-shot basis with a signal-to-noise ratio (the peak-to-peak noise level taken from the non-illuminated CCD pixels) exceeding 5, yielding a single-shot detection limit of 160 nJ per macropulse. With the demonstrated sensitivity, it is apparent that even the output of a much simpler diode laser system would be sufficient to perform the spectral calibration of FELs, and a more cost efficient set-up could be realized.

A maximum sideband energy of 150 pJ was achieved using the maximum Ti:Sapphire laser power (200 mW) and FELIX operating at full power of 5 mJ per macro-pulse. The order of magnitude of the sideband power is compliant with model calculations for this crystal. Under these conditions up to the fifth ($N=5$) sum- and difference-frequency sidebands could be detected at maximum gain of the image intensified CCD array.

Figure 3 shows the power dependence of the sideband energy recorded with the THz source operating at 151 μm . The left panel shows the THz laser energy dependence recorded while using 20 mW of the NIR laser. The right panel shows the NIR laser power dependence recorded for a THz energy of 50 μJ / macro-pulse. Clearly, both curves are perfectly linear, as expected. The noise-limited detection limit can thus be directly improved by increasing the NIR laser intensity.

Table 1 summarizes the signal-to-noise ratio and noise-limited detection limit for different benchmarking conditions. The first row shows the numbers discussed above for verification of the achievable detection limit, while the second row show the numbers for maximum sideband energy conditions. The last row shows the numbers obtained for measurement of a single micro-pulse. For these measurements, the micro-pulse repetition rate was reduced to 25 MHz and the detector gate was reduced to 10 ns duration. The micro-pulse energy is 13 nJ. The

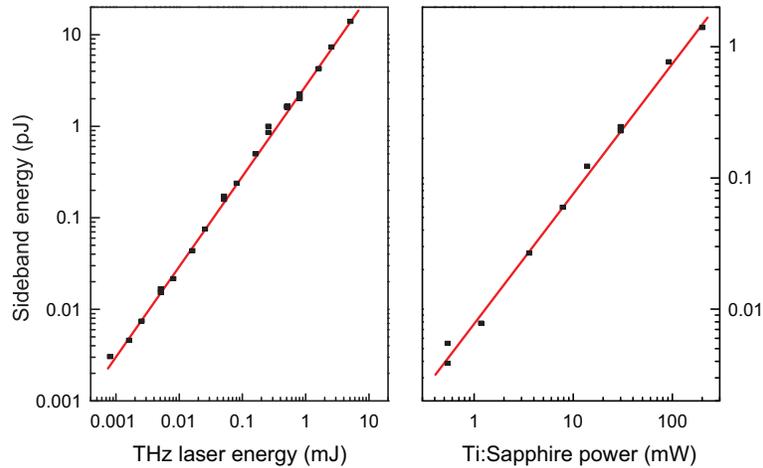


Fig. 3. THz laser energy and NIR laser power dependence of the sideband energy recorded at $151 \mu\text{m}$. For the left panel, a NIR laser power of 20 mW was used, for the right panel a THz macro-pulse energy of $50 \mu\text{J}$ was used.

deduced noise-limited detection limit of 13 nJ at maximum gain of the image-intensifier is in reasonable agreement with the limit obtained for minimum signal conditions for full macro-pulse detection considering the increased NIR laser power and the near-perfect linear scaling of the sideband energy with NIR laser power. It implies that the noise level on the CCD pixels is nearly integration-time independent, and thus dominated by read-noise. It should be noted that the detection limit is of course dependent on the spectral profile of the THz pulse as it increases with the number of detector pixels over which the light is distributed.

Table 1. Signal-to-noise ratio (SNR) and noise-limited detection limit for several benchmarking experimental conditions

Description	NIR power	THz energy	Gain	SNR	Noise limit
Min. sideband energy	20 mW	800 nJ	Maximal	5	160 nJ
Max. sideband energy	200 mW	5 mJ	Minimal	120	$40 \mu\text{J}$
Single micro-pulse	200 mW	13 nJ	Maximal	1	13 nJ

The spectral bandwidth of the THz light can be varied by slightly changing the length of the FEL cavity [16]. Figure 4 shows the spectral profile obtained using both direct detection (panel a) and upconversion (panel b, black lines) calibration schemes for three different cavity lengths. The spectra obtained by direct detection immediately reflect the spectral profile of FELIX as the instrumental resolution is small compared to the width of the FELIX spectrum. It is clearly demonstrated in Fig. 4(b) that the spectral profile of the THz light is also directly reflected in the spectral profile of the upconversion spectra, although the width of the upconversion spectral profile is clearly limited by the instrument resolution of the upconversion spectrometer. The dashed red lines in Fig. 4(b) plot the direct detection spectra after convolution with the instrument function of the upconversion grating spectrometer, showing good agreement with the experimental upconversion spectra.

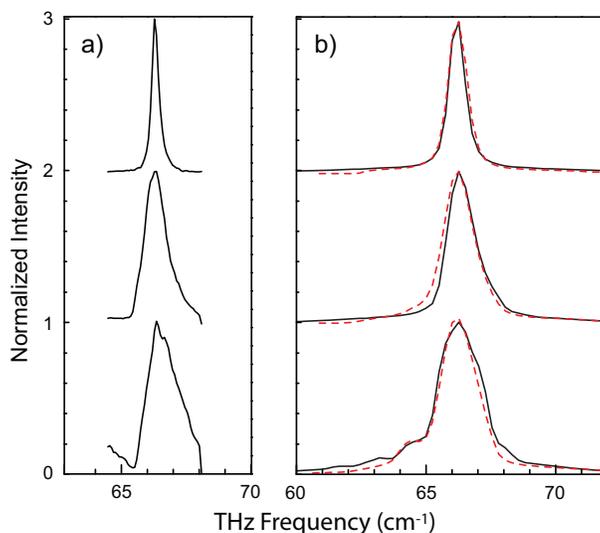


Fig. 4. Single-shot spectral THz profiles for different FEL cavity lengths: (a) using direct detection, (b) Black lines: using upconversion. Dashed red lines: direct detection spectra convoluted with the upconversion spectrometer instrument function (approximately 0.2 cm^{-1} full width at half-maximum causing noticeable broadening). We note that the broadest direct detection spectrum (lower curve, panel b) could not be completely measured using direct detection, resulting in a reduced width of the convoluted spectrum.

4. Conclusion

The upconversion spectrometer setup provides a very sensitive and versatile detection scheme for THz radiation by upconversion to the NIR range allowing the application of highly efficient optical detection techniques. The detection scheme does not require adjustment of any element in the setup when the THz frequency or spectral profile is modified. The results presented above indicate that a noise-limited detection limit of about 160 nJ per macro-pulse using 20 mW of the NIR laser, and 13 nJ per micro-pulse using 200 mW of the NIR laser, could be achieved on a single-shot basis. The complexity of the system can be significantly reduced by replacing the Ti:Sapphire laser by a narrow-band, frequency-stabilized diode-laser system; increased power, up to the Watt-level, can be obtained from systems equipped with a tapered amplifier.

The upconversion spectrometer is readily extended to longer wavelengths as well, and will be used as on-line spectrometer for FLARE, the Free electron Laser for Advanced spectroscopy and high-Resolution Experiments, currently under construction at the Radboud University in Nijmegen. FLARE, will combine a short-pulse pump-probe mode (1% resolution) with a high-resolution spectroscopic mode (spectral resolution in the 10^{-5} – 10^{-6} range) and will operate in the 100–1500 μm (0.2–3 THz) range [17]. The here presented scheme is directly applicable for the pump-probe mode of FLARE but also holds great promise for application for the spectroscopic mode as well, owing to the possibility to extend the upconversion spectrometer by adding a sensitive wavemeter, allowing very accurate frequency determination of the sideband.

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