Robust optimization of single-cycle THz setups based on phase-matching via tilted pulse fronts using an incident-fluence metric

Tobias Kroh^{*,a,b,c}, Timm Rohwer^a, Lu Wang^{a,b}, Umit Demirbas^{a,d}, Huseyin Cankaya^a, Mikhail Pergament^a, Franz X. Kärtner^{a,b,c} and Nicholas H. Matlis^a

^aCenter for Free Electron Laser Science (CFEL), Deutsches Elektronen-Synchrotron (DESY),
Notkestraße 85, 22607 Hamburg, Germany;

^bUniversität Hamburg, Department of Physics, Luruper Chaussee 149, 22761 Hamburg, Germany;

^cCenter for Ultrafast Imaging, Luruper Chaussee 149, 22761 Hamburg, Germany;

^dLaser Technology Laboratory, Antalya Bilim University, 07190 Dosemealti, Antalya, Turkey

ABSTRACT

Despite the popularity and ubiquitousness of the tilted-pulse-front technique for single-cycle terahertz (THz) generation, optimization of the experimental setup remains complex and difficult due to the sensitive dependence on and coupling between the optical pulse parameters, including fluence, beam size, angular dispersion and temporal compression. Here we present a systematic and robust method to tune the tilted pulse-front setup, based on use of selected multi-dimensional scans, which enables a straight-forward and accurate determination of optimum parameter values. Our methodology not only allows us to determine parameter sensitivities and achieve a robust optimum in the performance, but also enables a verification of certain physical properties of the lithium niobate prism, including the THz refractive index. The detailed step-by-step procedure is discussed and applied to a tilted-pulse-front THz setup at both room temperature and cryogenic temperatures. The procedure can be applied to any setup based on the tilted-pulse-front geometry and is important for the construction of high energy THz sources required for strong field terahertz applications such as novel particle acceleration schemes or beam manipulators.

Keywords: Terahertz, Nonlinear Optics, Tilted-pulse-front technique, Single-cycle THz pulses

* tobias.kroh@desy.de; Tel. +49 (0)40 8998-6359; <https://ufox.cfel.de/>

1. INTRODUCTION

Intense single-cycle terahertz (THz) pulses have not only helped to advance fields of spectroscopy, where they can be used to probe or control many intrinsic resonances such as lattice vibrations in solids^{1,2}, all-optical molecular alignment^{3,4}, but have also enabled study of strong-field phenomena in the THz range unraveling both perturbative⁵⁻⁷ and nonperturbative extreme nonlinearities of matter⁸⁻¹⁰. Recently, the availability of strong THz transients has enabled the field of compact THz driven charged particle accelerators and manipulators to emerge which holds great promises of providing table-top accelerators and their application in science¹¹.

Alongside flourishing fields of application, recent years have seen rapid development of table-top THz sources, including broadband THz pulses generated from plasmas¹²⁻¹⁵, low frequency pulses in the range of $\sim 10 \mu\text{J}$ from large area semiconductor switches¹⁶⁻¹⁸, or spin emitter heterostructures^{19,20}. Among the table-top sources the most common method is intra-pulse difference frequency generation (DFG) of intense ultrashort laser pulses in nonlinear materials due to both its versatility and scalability. Depending on the THz frequency range of interest, there is a wide range of nonlinear materials available such as semiconductors, organic crystals or ferroelectric crystals such as lithium niobate (LN)²¹. The latter has become a popular choice due to its availability at large cm-sizes, high optical damage threshold, low absorption in the optical range as well as high effective nonlinear coefficient²². However, low frequency phonon contributions to the dielectric function lead to a significant difference in refractive indices in the THz and optical range. Thus, collinear phase-matching via birefringence or temperature tuning is not possible. Consequently, methods for non-collinear phase matching were developed. Initial work in near single-cycle THz generation from LN relied on Cherenkov shockwave radiation^{23,24}, which is problematic to scale to high energies since the pump beam needs to be focused to a narrow line below the size of

the THz wavelength. This motivated discrete tilted intensity fronts to be used for improved THz generation first ^{1, 2, 25}. Later, Hebling and coworkers pioneered non-collinear phase-matching utilizing a pulse with a continuous intensity front tilted such that the projected group velocity of the laser pulse matched the THz wave velocity ²⁶. This allows the THz to build up coherently as it slides along the pump laser front.

In the years thereafter, the achieved THz pulse-energies were rapidly improved by several orders of magnitude towards the multi μJ -level ^{22, 26-29}. First, most experiments utilized broadband Ti:Sa pump systems while later, supported by theoretical studies ^{30, 31}, the focus shifted towards using more narrowband pulses with center wavelengths $> 1 \mu\text{m}$ associated with increased interaction lengths and reduced free-carrier generation via multi-photon absorption respectively ³²⁻³⁵. Further improvements were enabled via cryogenic cooling of the LN crystals yielding decreased THz absorption ³⁶ and enabling the highest conversion efficiencies reported to date ^{37, 38}. Others investigated the effects of chirped pump pulses ^{39, 40} or concentrated on finding optimal imaging configurations for reduced pump pulse duration variations across the tilted-pulse-front impacting the homogeneity of the generated THz beam ^{31, 41-44}. With increasing THz energies generated the reported conversion efficiencies reached beyond the so called Manley-Rowe limit (THz photon number exceeds pump pulse photon number), which is possible due to cascaded DFG processes, where redshifted pump photons are effectively reused after generating a THz photon ⁴⁵⁻⁴⁹. As phase-mismatch is less pronounced for redshifted pump photons, the DFG process is dominant compared to the opposite process where repeated sum-frequency-generation of pump-THz photons would annihilate THz photons instead, thus leading to overall enhanced THz conversion. The broadening of the pump pulse via this cascaded nonlinear interaction in combination with the group-velocity dispersion (GVD) associated with angular dispersion necessary to tilt the pulse imposes the main limitation for the maximum efficiency of the process ⁵⁰. Via development of increasingly advanced simulation tools to study the complex spatio-temporal effects in tilted-pulse-front setups ^{22, 51-53}, further limiting effects such as THz absorption by free carriers ⁵⁴, self-phase modulation leading to phase-mismatch ³⁴ or stimulated Raman scattering enhancing the breakup of the pump pulse ⁵⁵ were identified. In order to circumvent some of these limitations, several modified setup schemes were investigated, promising to allow scaling to even higher THz energies ^{43, 56-58}. With reported pulse energies up to $> 400 \mu\text{J}$ ³⁸ the conventional tilted-pulse front setup remains unbeaten and promises to enable single-cycle THz pulses with energies of mJ-level.

Despite the popularity and ubiquitousness of the tilted-pulse-front technique, optimization of the setup remains complex and difficult due to the sensitive dependence on and coupling between the optical pulse parameters, including fluence, beam size, angular dispersion and temporal compression of the pump pulse across the tilted intensity front. To the best of our knowledge, the literature lacks systematic optimization studies focused on tilted pulse front setups. In addition, there is a lack of experimental characterizations of important parameters such as imaging magnification and optimized pulse-front-tilt. Thus, it remains challenging to compare different schemes reported in the literature. In this work, the setup was designed and built to decouple most of the important parameters and determine an optimum configuration for a given laser pulse duration and energy via selected multi-dimensional parameter scans. In addition, characterization techniques in order to improve the comparability of different tilted-pulse-front setups are proposed and conducted. The obtained data allows to determine sensitivities of parameters on both the efficiency and the THz beam profile. Based on the findings, a step-by-step procedure of how to perform the setup optimization is given.

The manuscript is organized as follows. In the first section, the design strategy and the experimental setup are introduced. In the second part, the individual parameter scans and measurements are motivated alongside the discussion of the step-by-step optimization procedure proposed. The section concludes with a discussion of the results and parameter sensitivities.

2. EXPERIMENTAL SETUP

In order to allow a systematic optimization of a setup relying on a tilted-pulse-front based non-collinear phase-matching, the first step is to ensure the setup design allows for such optimization to be carried out and that the tuning parameters are decoupled to a reasonable extend. First, the optical design is discussed, before describing the implemented setup as well as important characterization techniques that will improve comparability with other setups. Moreover, a set of selected multi-dimensional parameter scans for optimizing the setup is motivated and discussed.

Figure 1. Schematic of the experimental setup. The pulse front is tilted via a gold-coated holographic diffraction grating (G), while a cylindrical telescope (L_1 , L_2) was employed to recompress the pulse inside the 5% MgO doped congruent lithium niobate (cLN) crystal. Two half-wave-plates allowed to adjust the pump laser polarization such that both diffraction efficiency of the grating and effective nonlinear coefficient of the intra-pulse DFG process were maximized. In order to optimize the performance of the setup, the pump beam waist σ_x^i , the grating angle (θ_i , θ_d) and thus the pulse front tilt angle γ , the pump energy E_p as well as the crystal position along x and z -axis were systematically scanned.

2.1 Setup design: A practical view on choosing a proper imaging setup for the optimization procedure

Two important parameters for the design of a tilted pulse front setup are the tilt-angle γ of the intensity front (see Fig. 1) as well as the angle of the grating image inside the crystal γ_{im} . While the correct pulse front tilt ensures noncollinear phase-matching of the intra-pulse DFG process, the image location of the grating defines the region across which the pump pulse duration is re-constructed after its spatio-temporal broadening due to the angular dispersion imposed on the beam by the grating. Both angles sensitively depend on the choice of grating parameters such as the groove density ρ , angle of diffraction θ_d as well as the magnification factor M of the imaging setup^{31, 41, 42, 59}:

$$\tan \gamma = \frac{m \lambda_0 \rho}{M n_g \cos \theta_d} \quad (1)$$

$$\tan \gamma_{im} = M n_g \tan \theta_d \quad (2)$$

Here, m denotes the order of diffraction picked from the grating (usually $m = \pm 1$), λ_0 the center wavelength of pump laser and n_g the optical group index of LN. In order to optimize the symmetry and quality of the generated THz pulses, γ should equal γ_{im} . As the pulse-front tilt has to be chosen according to $\cos \gamma \approx n_g/n_{THz}$ in order to fulfill the non-collinear phase-matching condition²⁶, Eqs. (1) and (2) recommend selected pairs of ρ and M for optical setups.

Among the frequently used configurations for tilted-pulse front setups for THz generation, there are two versions of imaging setups, which are widely used. Most setups utilize a single lens for imaging the tilted-pulse front into the crystal whereas others rely on a telescope for imaging. One advantage of the single-lens setup is, that the magnification and thus both the pulse-front-tilt angle and the angle of the image can be tuned via adjusting the grating-lens and lens-crystal distances, often used as optimization procedure in experiments^{34, 35, 38, 58, 60}. However, it is harder to decouple effects due to a change in pump fluence from effects caused by the altered angles of pulse-front tilt and image plane during this optimization procedure and determine a global optimum of the THz efficiency since all three parameters are simultaneously changed. Imaging telescopes yield the advantage of a fixed magnification factor M independent of the distance of the position of the object and that they can be characterized with better precision. Thus, once constructed and characterized, it is easier to keep track of the fluence on the crystal and this way decouple effects caused by the pulse-front tilt from a

change in pump fluence. Additionally, single-lens setups have been reported to suffer more severely from imaging errors degrading the pump pulse duration across the intensity front and thus degrading the homogeneity and quality of the generated THz beam^{43,44}. Thus, in order to achieve reduced imaging errors, a more accurately characterized setup as well as easier decoupling of scanning parameters, a two-lens telescope was chosen for the setup shown in Fig. 1. Note that in such a setup, the pulse-front tilt angle required for phase-matching is then optimized via the grating angle, which has the slight disadvantage that it can be more tedious to scan.

As the non-collinear phase-matching takes place in the x-z plane, there is an optimal pump beam size σ_x^f (see Fig. 1) associated with an optimum interaction length between pump and THz beam. The beam width along the y-axis however is not directly affecting the phase-matching process and thus allows adjustment of the pump fluence to its optimal level. Additionally, given a large crystal, this beam size σ_x^f can be increased for scaling tilted-pulse-front setups to higher pump energies. Thus, cylindrical optics for individual adjustment of both σ_x^f and σ_y^f were chosen for the optimization procedure in this work.

The imaging setup and diffraction grating for the setup were selected following the suggestions given in Refs.^{31, 41, 42, 59}. A holographic diffraction grating was implemented as it yields the advantage of an almost flat diffraction efficiency across a wide range of angles. This is particularly helpful for scans of the grating angle in order to tune the pulse front tilt without affecting the pump energy incident onto the crystal significantly. To approximately achieve a well compressed pulse duration across the tilted intensity front inside the crystal, combinations of $M = f_2/f_1$ (note, in some literature the angular magnification $1/M$ is used instead) and grating period ρ should be chosen, such that $\gamma = \gamma_{im}$. Table 1 summarizes the design chosen for the setup presented in this work.

Table 1. Parameters used for the design and the ones found for the experimentally optimized tilted-pulse-front setup used in this work for both cryogenic temperatures (80K) as well as room temperature (RT).

Parameter	Design	Experimental results
Groove density ρ	1500 lines/mm	1500 lines/mm
Center wavelength λ_0	1020 nm	1020 nm
Telescope magnification M	0.6666	0.6721 ± 0.0007
Incident angle θ_i	42.8° (80K), 41.0° (RT)	41.52° (80K), 39.96° (RT)
Diffraction angle θ_d	58.2° (80K), 60.9° (RT)	59.7° (80K), 62.3° (RT)
n_{gr} (1020 nm)	2.2158* (80K), 2.2316 (RT) ⁶¹	
n_{THz}	4.89** (80K), 5.21** (RT)	~5.0 (80K), ~5.4 (RT)***
Grating magnification M_{gr}	0.48 (80K), (RT)	0.67 (80K), 0.61 (RT)
Pulse-front-tilt angle γ	63.0° (80K), 64.8° (RT)	63.7° (80K), 65.4° (RT)
Crystal cut angle α	63.0° (80K), 64.8° (RT)	62.4°

* obtained via extrapolation of measured values given in⁶².

** obtained from THz time-domain spectroscopy measurements at ~300 GHz

*** Estimated based on inserting the obtained experimental results into Eq. (1).

2.2 Experimental setup & measurement devices

The setup geometry implemented is shown in Fig. 1. A 2-stage cryogenic Yb:YLF amplifier system delivering high energy pulses centered at ~1020 nm with a pulse duration of ~1.35 ps at 10 Hz repetition rate was used as pump source⁶³. A multi-kHz repetition rate version of this amplifier, potentially with sub-250-fs pulse duration is under development for future experiments⁶⁴. For the experiments, up to around ~25 mJ were utilized and sent onto a gold-coated holographic diffraction grating (Spectrogon AB, 1500 l/mm, 50x50 mm, > 90 % efficiency for 1020 - 1032 nm, deviation 13°) mounted on a hybrid rotation stage with integrated translation capability (Newport, DGM-1). The diffracted beam of -1st order ($m = -1$) was sent through a Keplerian cylindrical telescope ($f_1 = 300$ mm, $f_2 = 200$ mm, Thorlabs, 1" diameter, B-coated) that images the grating into the 5% MgO doped congruent lithium niobate (LN) crystal (27x27x25 mm, height 25 mm, cut

angle $\alpha \sim 62.4^\circ$), if the distance d_2 (second lens-crystal) is set correctly depending on d_1 (grating-first lens). The LN crystal was AR-coated on the two surfaces where the pump beam enters and exits the crystal, while the THz output surface remained uncoated. The positioning of the crystal within the pump beam was performed using a motorized stage (Thorlabs, LTS150) for movement along the z-axis and a manual translation stage for positioning along the x-axis. Two half-waveplates before and after the grating ensured optimal polarization for highest diffraction efficiency (TM polarization) and maximized effective nonlinear coefficient d_{eff} of the intra-pulse-DFG process, which is approximately proportional to the 4th power of the electric field projected onto the optical axis of the LN crystal and thus shows a $\cos^4$ -dependence on the polarization angle (see Fig. 3b).

A motorized $\lambda/2$ waveplate (Thorlabs, PRM1Z8) in combination with a thin-film-polarizer (TFP) were implemented to automate pump energy scans. The incident IR pump energy was measured using a large area energy meter (Coherent Inc., LabMax TOP with sensor EnergyMax-USB J-50MB-LE) while the extracted THz pulse energies were measured using two different pyro-electric detectors: A sensitive detector with a 9 mm active area (Gentec-EO, SDX-1152 with MT coating) without traceable calibration in the frequency range < 4 THz, and a less sensitive but larger area detector (SLT Sensor- und Lasertechnik GmbH, THz 20, 20 mm active area) with flat frequency response across the whole THz range, crucial for characterizing and optimizing broadband THz-pulses, with traceable calibration at 693 GHz performed at the PTB Berlin, Germany (calibration certificate 73426 PTB 18). A cross-calibration of both detectors with broadband THz pulses centered around ~ 200 GHz yielded a calibration factor of (74 ± 3) mV/ μ J for the Gentec detector, from which the signal was acquired using a digital storage oscilloscope (Tektronix, TDS 2024C). In both cases, the THz energy was acquired automatically via a desktop computer and home-built software. To avoid distortions of the measurement, residual infrared and second harmonic light exiting the LN was blocked upstream of the detectors by a sheet of black polyethylene (PE, 1.9mm thickness) with a measured average transmission of $\sim 50\%$ in the 150 - 450 GHz range by THz time-domain spectroscopy, which agrees well with the transmission of $\sim 54\%$ determined throughout the detector cross-calibration. The transmission factor of 54% was used in order to obtain the THz energy in front of the PE covering the detector.

Experiments were conducted both at room- and cryogenic temperatures, for each of which there were slight modifications: At room temperature, a three-layered Kapton-tape (Thorlabs, KAP22-075) coating was applied to the THz output surface of the crystal in order to reduce transmission losses. In addition, the crystal was indium bonded to a copper cold-finger inside a vacuum chamber attached to a liquid nitrogen Dewar in order to be cooled to ~ 80 K. At cryogenic temperature, no AR-coating for THz was applied and the usable THz energy was measured outside the vacuum chamber after transmission through a 3 mm thick uncoated TPX window (TYDEX).

As there is still a lack of reliable THz cameras for the characterization of broadband pulses in the spectral range < 1 THz with sufficient resolution, a motorized x'-y'-z' scanner (x', y': Thorlabs, NRT 100-M, z': Thorlabs, LTS300) was constructed in order to scan a sensitive small-area pyroelectric detector (WiredSense GmbH, MPY-01-XX and MPY-LG-XX, 2x2 mm active area) across the THz beam profile and reconstruct images of the THz beam. The detector signal was acquired using a fast-readout digital oscilloscope (Pico Technology, PicoScope 2408B) via home-built software, with integration and background subtraction implemented for reduced SNR. The same sensitive detector was used in a Michelson type THz interferometer utilizing a pellicle beam splitter in order to measure the THz spectrum via Fourier transformation of the recorded interferogram.

As mentioned in section 2.1, the pump width σ_x^i in the phase-matching plane (x-z-plane) was adjusted via different Galilean cylindrical telescopes that were set up before the tilted-pulse-front setup. The pump beam sizes along x (σ_x^i) and y-axis (σ_y^i) were monitored using a large area CCD camera (DataRay Inc., WinCamD-LCM-C), while the telescope characterization described in the next section was performed using a smaller CCD camera (Ophir, Spiricon USB-SP 926) and a CW alignment laser (CrystalLaser Inc., 1030 nm wavelength).

3. STEP-BY-STEP OPTIMIZATION PROCEDURE

Given fixed laser parameters, the nonlinear process underlying the THz generation depends heavily on phase-matching required for efficient buildup of the THz and on the effective interaction length between pump pulse and THz pulse. In addition, the temporal compression of the pump pulse in the interaction volume and the peak fluence impact the THz generation. Consequently, optimization involves tuning the pulse-front tilt angle γ (phase-matching), the transverse size of the beam in the pulse-front-tilt plane σ_x^f alongside with setting the transverse position, d_a , of the beam relative to the crystal apex (which determines the effective interaction length), setting the longitudinal position of the crystal relative to the image

plane (temporal compression) and finally adjusting the transverse size of the beam in the vertical, non-tilt plane σ_y^f (optimal fluence). Fig 2(a) provides an overview over these steps needed for the optimization.

Figure 2. (a) Schematic of the experimental optimization procedure performed with in-depth schematics showing the multi-parameter scans necessary to optimize the pulse-front tilt angle γ (b) and the transverse pump beam size σ_x^f and the crystal position (c). Frequently needed scans, where automation is useful, are visualized via the computer icon.

Some steps are rather simple, such as maximizing the effective nonlinear coefficient for the DFG process via alignment of the pump polarization along the optical crystal axis, as shown in Fig. 3(b). Nonetheless, this step is important as the THz energy is sensitively dependent on the pump polarization and only a few degrees of mistuning lead to significant degradation of the efficiency.

3.1 Optimization of the pulse front tilt

While some steps are simple, other steps, however, require up to several parameters to be tuned at once, as some affect one another. For example, adjusting the pulse-front can be performed via turning the diffraction grating. In this process however, both diffraction efficiency and magnification factor of the grating change, thus affecting the pump fluence incident on the crystal. Therefore, the pump energy must be adjusted to keep the pump fluence at a constant level in order to compare results obtained from different grating angles. Alternatively, it is possible to scan the energy for each new grating angle and compare the measurements at the same fluence level later, which is the procedure chosen here (see. Fig. 2(b)). As the image location of the grating may be slightly affected during the scan, the crystal-lens distance d_2 was additionally scanned, which can be considered optional in the overall optimization strategy, though. Note, that optimizing the pulse front tilt ideally requires a sufficiently large pump size σ_x^f on the order of the effective interaction length (> 1 mm), as for small beam sizes phase-matching matters less and the dependence on the pulse-front tilt is less pronounced^{53, 65}. On the other hand, the absolute changes in the beam size via the varying grating magnification M_{gr} can distort the optimization, which becomes more severe for larger beams. As such, large changes in σ_x^f could distort the optimization of gamma via the affected interaction length and in addition, the apex-distance d_a would have to be readjusted for each new grating angle, increasing the complexity of the procedure.

In preparation for the grating angle optimization, it is crucial to calibrate the grating mount angle to the actual set of (θ_i, θ_d) and to characterize the imaging setup in place. Firstly, this allows comfortable pump fluence determination at the crystal position from measured beam profiles before the setup and secondly, to determine the pulse-front tilt found optimal (or angular dispersion respectively). The latter can be used to compare one tilted-pulse-front setup with another. The rotation angle θ_{mount} can be calibrated via the angle at which the zero-order beam is diffracted back onto itself (θ_{mount}^0). The calibrated angle of incidence is obtained via $\theta_i^{cal} = \theta_{mount} - \theta_{mount}^0$, while θ_d is obtained via the grating equation. If the grating is now rotated by an angle $\Delta\theta_{mount}$, then θ_d is rotated by the same amount, while this time θ_i is obtained via the grating equation. Note, that since $\theta_d \neq \theta_i$, the grating (de)magnifies the beam along the x-axis via $M_{gr} = \cos \theta_d / \cos \theta_i$.

The telescope on the other hand can be effectively characterized by its magnification M , on which both the pulse front tilt and image-plane angle depend (see Eqs. (1) and (2)). Therefore, it is crucial to measure M with high precision. A simple yet accurate measurement technique is to image an object placed on a micrometer stage at distance d_1 before the telescope onto a CCD camera mounted on a stage at distance d_2 after the second lens. Using the ray-tracing matrix formalism it can easily be shown that $\Delta d_2 = M^2 \Delta d_1$. Thus, recording the positions d_2 where the image is sharp vs various positions d_1 of the

object, allows measurement of M^2 via the slope of a linear fit to the data with orders of magnitude higher precision as compared to using a simple resolution test target. For the setup discussed here, $M = 0.6721 \pm 0.0007$ as shown in Fig. 3(a).

Figure 3. (a) Determination of the imaging setup magnification M via recording the shift Δd_2 of the image position of a resolution test target (see insert) for known longitudinal displacements of the object before the telescope Δd_1 . The magnification factor M can then be determined via a linear fit (orange) to the data. (b) Shows the optimization procedure of the pump polarization via the angle of the half-wave-plate after the grating. In agreement with expectations the recorded THz energy follows a $\sim \cos^4$ law with the angle of the pump electric field polarization relative to the optical axis of the crystal. Fitting to the data allows effective and precise determination of the optimum waveplate angle.

With M and M_{gr} known, the pump peak fluence F_p incident on the crystal can then be obtained using the characteristics of the pulse before the grating:

$$F_p = \frac{T_{setup} E_p^i}{\pi M_{gr} M M_x \sigma_x^i M_y \sigma_y^i} \quad (3)$$

Where E_p^i is the incident pulse energy onto the grating, σ_x^i and σ_y^i denote the 1/e radii of the intensity profile, T_{setup} is the combined transmission through the setup including transmission through the telescope as well as the diffraction efficiency of the grating. T_{setup} was measured to be surprisingly high for the presented setup with around 96% and constant over the range of angles scanned. M_x and M_y are the magnification factors of cylindrical telescopes used to shape the beam before the tilted-pulse-front setup. So far $M_x = M_y = 1$. Fig 4 shows the sensitive dependence of the measured THz energy on both pump fluence and θ_i for operation at both room and cryogenic temperature (80K). The optimal angle differs, since the refractive index in the THz range changes as the crystal is cooled to lower temperatures, thus requiring a change in pulse-front tilt for phase-matching. Note, that even though at cryogenic temperatures the pump fluence was kept low to avoid optical damage during the scans, the achieved energies are comparable to the room-temperature setup pumped at almost twice the fluence. Both scans show how even a small deviation from the optimal pulse-front tilt affects the THz conversion efficiency. The optima found also allow for a rough estimate of the THz refractive indices. Using Eqs. (1) and (2), assuming an optimal setup configuration and inserting all known parameters except the THz indices, it is possible to obtain an estimate for their values in the setup (see Tab. 1). The estimated refractive indices are within a range of approximately ± 0.2 compared to published values^{66, 67}.

Figure 4. Optimization of the pulse front tilt angle via tuning of the grating angle at room-temperature (a) and with a cryogenically cooled LN crystal (b). The THz energies plotted were measured outside the crystal or Dewar respectively.

3.2 Optimizing the interaction volume of the nonlinear process

After the grating angle is set to the optimum found, the interaction length is to be optimized next. As can be seen from Fig. 2(c), optimizing σ_x^f requires the crystal position within the pump beam to be optimized each time. Energy scans are required in addition, as the fluence would otherwise be different for each σ_x^f . In order to test different pump sizes, 5 different cylindrical telescopes with different M_x were constructed in front of the tilted-pulse-front setup. For scans of the crystal position it proved helpful to use a large area THz detector, as otherwise the detector position had to be constantly adjusted due to the THz output location changing with crystal position. For each σ_x^f ($1/e^2$ intensity radius) the maximal observed THz energy at $F_p = 90 \text{ mJ/cm}^2$ is shown in Fig. 5(a) for the setup operated at room temperature. The corresponding optimum distance d_a at each tested σ_x^f was found to lie within the range of 3 – 4 mm, while the accuracy of the measurement was $\sim 0.5 \text{ mm}$. The global optimum is observed at $\sigma_x^f \approx 2.5 \text{ mm}$ ($1/e^2$ intensity radius) and $d_a = 4 \pm 0.5 \text{ mm}$. Fig 5(b) shows the recorded THz energy and conversion efficiency as a function of the pump fluence for the setup operated at room temperature. THz pulses with usable energies up to $\sim 30 \mu\text{J}$ at a conversion efficiency $\sim 0.12\%$ were generated while pumping with a fluence up to 160 mJ/cm^2 .

The impact of the crystal position onto the THz energy is shown in Fig. 5(c) for $\sigma_x^f \approx 1.4 \text{ mm}$. Though d_a and d_2 are shown not to be independent of each other, in principle they can be optimized individually because the range within which the THz energy would degrade significantly is on the order of one mm. Thus, a detailed 2D scan of d_a vs d_2 , as shown in Fig. 5(c), is not necessary for a rough optimization. As can be seen from Fig. 5(d), the distance d_a affects the THz energy more severely at smaller values. If the beam is set completely onto the crystal, the impact onto the THz energy is less pronounced. We note that recent numerical studies have revealed the temporal THz waveform to degrade from a near single-cycle pulse for pump beams offset far from the crystal apex, even if the energy may not be affected much⁶⁸. As such it is recommended to stay as close to the crystal apex as possible while maintaining reasonable efficiency levels.

Note, that in addition it is important to perform these scans at reasonable fluence levels, since optimization at low fluences may yield different optimal configurations in comparison to high fluences due to the different effective interaction lengths⁵³. The trend seen in Fig 5(a) verifies the existence of an optimal pump beam width so far only predicted by simulations. Small beams do not suffer as much from imperfect imaging conditions, but the THz may not build up effectively if the interaction length is larger. Larger pump beams however suffer from more severe temporal distortions due to imaging errors and parts of the beam going through more material degrade due to breakup of the pulse, thus not contributing to the THz generation effectively anymore^{53,69}.

Figure 5. (a) The maximum THz energy (determined at room temperature) strongly increases with pump beam radius σ_x^f and saturates for $\sigma_x^f \approx 2.5 \text{ mm}$ ($1/e^2$ intensity). The measurements were compared for constant fluence of $F_p = 90 \text{ mJ/cm}^2$. (b) THz pulses with energies up to $\sim 30 \mu\text{J}$ at a conversion efficiency $\sim 0.12\%$ were generated pumping with $\sigma_x^f = 2.5 \text{ mm}$ and a fluence up to 160 mJ/cm^2 . (c) The THz energy (here $F_p = 90 \text{ mJ/cm}^2$ and $\sigma_x^f = 1.24 \text{ mm}$) is shown to depend both on the longitudinal (d_2) and transverse position of the LN crystal (d_a). (d) Shows the asymmetric decrease in THz energy if the crystal position is offset from its transverse optimum which is found to be $d_a \approx 3 - 4 \text{ mm}$ independent of the pump beam size.

3.3 Finalization of the setup

The last step of the optimization procedure allows adjustment of the vertical pump size σ_y^f via cylindrical optics before the setup (M_y) such that the incident pulse energy illuminates the crystal with the fluence found to yield highest efficiency. With this final step, the total efficiency of the setup is finally maximized.

Fig 6(a) shows the spectrum of the THz pulses to be centered around 0.22 THz for the room temperature setup. As the pump beam is elliptically shaped, it is not surprising that the recorded THz beam profile is slightly elliptical as well (see Fig. 6(b)). However, the measured THz beam sizes ($1/e$ intensity radii) measured at different distances along z' are well-fitted by Gaussian beams with waists σ_x^{THz} and σ_y^{THz} and their waist set to the crystal output surface. Note, that the THz was propagating at an angle of $\sim 12^\circ$ with respect to the z' axis, which matches the expected angle calculated by Snell's law based on the difference between pulse-front tilt angle (65.4°) and crystal cut α (62.4°).

Figure 6. (a) Recorded spectrum of the finalized setup via a Michelson type interferometer. The interferogram is shown in the inset. (b) The measured THz beam sizes σ_x^{THz} and σ_y^{THz} ($1/e$ intensity radii) along propagation z' were fitted assuming Gaussian beam propagation (dashed lines). The THz beam profile recorded at closest distance to the crystal surface is depicted in the inset.

3.4 Sensitivity of the conversion efficiency on the setup parameters

The pulse front tilt angle γ was observed to be a crucial parameter to be set correctly, as a small deviation of the grating angle θ_i by only 1° can lead to a drop of the efficiency by $\sim 60\%$. As γ is not observed to depend on the pump fluence significantly it thus can be adjusted at low pump fluence. This is advantageous in order to not optically damage the crystal accidentally during the procedure. As discussed, it is important to rather start with a larger beam size σ_x^f and the full beam incident on the crystal, in order to ensure γ has the largest impact onto the efficiency. The pump polarization affects the efficiency just as severely but is experimentally more straightforward to optimize via a half-wave-plate. Furthermore, the beam size in the phase-matching plane sensitively affects the efficiency. While the efficiency first grows with increasing beam pump width, there is an optimum size after which the efficiency decreases slightly. Unfortunately, the beam size could not be expanded further to study this behavior in more detail as the size of our optics prevented even larger $1/e^2$ beam radii beyond 3 mm. As expected, the crystal position along x and z should be changed simultaneously in order to optimize the interaction volume. However, if the whole pump beam is incident onto the crystal, the x and z positions impact the conversion efficiency less than the other parameters. In this case, a rough optimization can be performed by maximizing the THz energy via independent scans of d_2 and d_a . That optimization should be performed at high fluence, as the fluence level affects the effective interaction length of the nonlinear process.

4. CONCLUSION AND OUTLOOK

To conclude, a robust and systematic method to tune the tilted pulse-front setup, based on selected multi-dimensional parameter scans was introduced. The optimization strategy enables a straightforward determination of ideal parameters such as angular dispersion, temporal compression, beam size and pump fluence. From the data taken, the sensitivities on parameters were derived and discussed. Additionally, the characterization techniques discussed allow better comparability of different tilted pulse-front setups.

Such systematic optimization is important for the construction of high energy THz setups and can be applied for any nonlinear material used in such non-collinear phase-matching geometry. Thus, it is of special relevance for high field THz applications such as strong-field THz spectroscopy or charged particle acceleration. For example, the presented technique was used to construct and optimize two high energy setups in order to power a novel prototype of a THz electron gun, with first experiments yielding electron bunches with energies in the keV regime. Furthermore we note, that recently Bodrov and coworkers suggested improved performance of tilted-pulse front setups via a modified optimal imaging design in which image and crystal cut are at larger angle than the pulse front tilt, however requiring an outcoupling prism for the generated THz ⁴². A systematic experimental study of their proposed design with comparison to our results may be of interest for future work.

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