All-Optical Electron Acceleration with Ultrafast THz Pulses

by

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Submitted to the Department of Electrical Engineering and Computer Science
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Abstract

This thesis discusses a series of advances toward—and resulting in—the demonstration of the first ultrafast THz-driven electron gun, a technology with the potential to deliver unprecedented electron beam quality to scientists studying matter at the ultrafast and ultrasmall scale via electron diffraction or x-ray imaging.

In Part 1, we discuss various advances in generation of high energy pulsed THz radiation, a spectral regime uniquely effective at accelerating electrons but historically lacking in efficient sources. In particular, through various improvements to the grating-based tilted pulse front (TPF) technique, we demonstrate a record conversion efficiency of 1%. We also implement echelon-based TPF, achieving 3.5x higher efficiency than grating-based TPF for short (∼100 fs) pulses. Finally, we reuse the residual pump to obtain a recycled efficiency around half to a quarter that of the original. This reduced efficiency can be linked to spatio-spectral distortions in the residual pump, and we characterize these distortions to better understand the asymmetric dynamics of the THz generation process.

In Part 2, we discuss the design, testing, and commissioning of an electron gun driven exclusively by THz radiation. The accelerating structure, capable of broadband, dispersionless THz propagation and sub-wavelength confinement, is analyzed through electromagnetic simulations and experimental tests. We also characterize the accelerated electrons in absolute charge and spectrum as a function of emission phase and THz energy, while showing that the behavior matches well with theory and simulation. Our first-version THz gun delivers near 1 keV electrons accelerated by field strengths surpassing that of the best operational RF guns. The gun also delivers narrowband electron spectra which can already be used for low-energy electron diffraction.

Thesis Supervisor: Franz X. Kärtner
Title: Professor of Electrical Engineering

Thesis Supervisor: Erich P. Ippen
Title: Elihu Thomson Professor of Electrical Engineering
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Chapter 1

Overview

1.1 Quest for an optically-driven x-ray free electron laser

Just as the invention of the x-ray tube a century ago unleashed a revolution in medicine, breakthrough science often occurs at the heels of breakthrough scientific tools. Some of nature’s best kept secrets lie in the ultra-small (1 nm) and ultra-fast (1 fs), which can only be revealed through x-ray free electron lasers built on state-of-the-art electron accelerators. Scientific discovery in this ultra-small and ultra-fast regime is certain to create vital opportunities and inventions, including better data storage, computer chips, solar cells, and fusion reactors, but the tool to make those discoveries must first become ubiquitous.

Unfortunately there are currently only two x-ray free electron lasers in the world because they require kilometer-long underground tunnels and billions of dollars to build. Much of the size and cost is attributed to bulky, rather inefficient vacuum electronics (klystrons, magnets, undulators) producing the electromagnetic forces needed to accelerate or manipulate the electrons. One audacious project in our group, called AXSIS (attosecond x-ray science, imaging, and spectroscopy) aims to build a tabletop free electron laser without being constrained by conventional accelerator orthodoxy consisting of long tunnels of vacuum waveguides and heavy RF electronics [59]. An
Figure 1-1: Overview of the AXSIS (attosecond x-ray science, imaging, and spectroscopy) project. The proposed AXSIS machine is a THz-driven attosecond coherent x-ray source based on inverse Compton scattering, with the goal of obtaining atomically-resolved movies of ultrafast processes in important biological macromolecules. Adapted from [59]. Chapter annotations denote the realm of contributions this thesis work has made to the effort.

<table>
<thead>
<tr>
<th></th>
<th>RF</th>
<th>THz</th>
<th>Optical</th>
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<tr>
<td>Wavelength</td>
<td>few cm</td>
<td>0.1-1 mm</td>
<td>~ μm</td>
</tr>
<tr>
<td>Bunch charge</td>
<td>~1-10 nC</td>
<td>&lt;100 pC</td>
<td>~10 fC</td>
</tr>
<tr>
<td>Accelerating field</td>
<td>~100 MV/m</td>
<td>~1-10 GV/m</td>
<td>&gt;100 GV/m</td>
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Table 1.1: Comparison of RF, THz, and optical frequencies as candidates for driving an accelerator. Performance in key requirements (bunch charge and accelerating field) is compared.

An overview of the AXSIS machine is shown in Figure 1-1.

Distinctively, the AXSIS machine uses laser-based THz radiation rather than klystron-based RF radiation to power the accelerator. While this may seem like simply a shift in the operating frequency, its technological implications are enormous. Laser-based THz radiation has significantly higher damage threshold, energy-efficiency, compactness, and it enjoys perfect time synchronization with the UV photomitter which emits the electron bunch. The wavelength of THz radiation is also long enough to accommodate electron bunches of up to 100 pC within an accelerating half-cycle, whereas a purely optical-wavelength accelerator would accommodate prohibitively small electron bunches (Table 1.1).
To generate x-rays from the THz-accelerated electrons, the AXSIS machine uses a back-propagating laser to undulate the traveling electrons via inverse Compton scattering; the undulating electrons then radiate back light at x-ray frequencies.

1.2 Outline of thesis

In this thesis, we discuss the author's two major contributions made toward the overall AXSIS effort: (1) generation of high energy THz pulses and (2) demonstration of a THz-driven electron gun. These two subsystems drive the rest of the machine. The THz source imparts the force needed to drive the electrons to relativistic speeds as quickly as possible, while the THz electron gun produces and tailors the electron bunches with high spatiotemporal quality down the accelerating beamline and such that they efficiently interact with the laser during the inverse Compton scattering stage. These two subsystems are placed in context via the annotations in Figure 1-1.

In Chapter 2, we begin with an introduction to ultrafast optics and accelerator science. We provide a theoretical and experimental discussion of essential background topics such as mode-locked lasers, nonlinear optics, and electron beam characterization.

Chapter 3 dives into the vast subject of THz pulse generation. We first justify that tilted pulse front optical rectification is the best method for energy-scalable THz pulse generation while providing the necessary theoretical and numerical backing. Next we detail various techniques that we have found to improve the generation efficiency. Then we report two experiments that were performed: one on record-high efficiency THz generation and one on residual pump pulse recycling.

In Chapter 4, we investigate the physics behind THz-electron interaction using ‘streaking experiments’ and analyzing them via simulations. We find that electrons gain energy when they are emitted in the presence of the THz field, and the energy gained depends on the phase of the THz in which they were emitted, following THz vector potential. Further we see that the electron spectrum is significantly broadened by space charge, and that this effect weakens with increasing THz field.
Chapter 5 reports on the THz electron gun, the main feature of this thesis. We first introduce the concept of a single-cycle electron gun, which is in contrast to conventional electron guns which are highly multi-cycle. Next we discuss the methodology used to design, fabricate, and characterize the gun along with the surrounding apparatus. Then we present the results of the gun’s performance in charge, spectrum, spatial quality, scalability, and reliability.

Finally, in Chapter 6, we conclude with a summary of our work and future outlook.
Chapter 2

Background

In this chapter, we introduce the theoretical framework and experimental know-how pertinent to the work in this thesis. The first two sections, ultrafast lasers and optical parametric processes, provide information on how laser radiation is produced, manipulated, and characterized. The last section, electron beams, takes a quick dive the vast field of accelerator physics. Since we approached the field of accelerators from a laser background (casting the accelerator as a application of the laser), we aimed to glean as much inspiration and knowledge of electron beams as was helpful for the design of a THz-driven gun, without delving so deeply as to be bound by the conventions of the accelerator discipline.

2.1 Ultrafast lasers

2.1.1 Mode-locked oscillators

Every femtosecond amplified laser system starts with a mode-locked oscillator. An oscillator that is not mode-locked lases at a single frequency (longitudinal mode), or at most a few frequencies, due to mode competition and the non-uniformity of gain. The mode with highest gain eats up all the available population inversion. Mode-locked oscillators have stable multimode operation and the modes have a fixed (typically linear) phase relationship with one another. In the time domain, this will be
a pulse train of ultrashort pulses with a repetition rate equal to the cavity fundamental frequency (e.g. \( f = \frac{c}{2L} \) for a linear cavity).

Mode-locking refers to a group of methods which share a common motif: insert an element into or configure the laser cavity in such a way that pulsed, high intensity portions (spatially and temporally) of the beam experience low loss, while low intensity portions of the beam experience high loss. These methods may employ a variety of nonlinear phenomena, including Kerr-lensing and saturable absorption used in free space oscillators, or nonlinear phase shifting (such as in Figure-8 lasers) and nonlinear polarization evolution (NPE) used in fiber oscillators.

Today, the Kerr-lens Ti-Sapphire oscillator is the predominant laser used in high field femtosecond laser applications due to its wide bandwidth. Some of our experiments utilized Ti-Sapphire oscillators and amplifiers which were commercially built.

**Yb-doped fiber oscillator**

Ytterbium-doped fiber lasers lasing around 1 micron are also very popular and tend to be cheaper. Some of our experiments utilized Yb-doped lasers as well. The Yb-doped fiber oscillator in our experiments was a homebuilt NPE laser as depicted in Figure 2-1. The oscillating beam in the cavity circulates clockwise in the figure. It experiences gain when propagating through the Yb-doped fiber (YDF). The stretching that occurs inside the long single-mode fiber (SMF) is overcompensated by the grating pair compressor. In this way, the beam exits the grating compressor with an anomalous dispersion and exits the SMF with a normal dispersion. Somewhere during propagation through the SMF, the pulse achieves transform-limited maximal intensity. At this point NPE causes the higher intensity portions of the beam to undergo more polarization rotation than the lower intensity portions of the beam, as illustrated in Figure 2-2. Consequently the beam exiting the fiber will have different polarization states for different portions (of higher or lower intensity) of the beam. The following waveplate pair and polarizing beam splitter (PBS) are then adjusted to minimize cavity loss for the polarization state favoring the high intensity (pulsed) signal.
CHAPTER 2. BACKGROUND

Figure 2-1: Fiber oscillator schematic. SMF: single mode fiber. YDF, Yb-doped fiber. PBS, polarization beam splitter. WDM, wavelength division multiplexer. LD, laser diode. Adapted from [64].

Figure 2-2: Nonlinear polarization evolution (NPE) illustration. Adapted from [86].
2.1.2 Damage and nonlinear effects at high intensity

Although the average power of most amplified femtosecond lasers is relatively low (e.g. 2 W), the peak power of the femtosecond pulses can be in the tens of gigawatts. Further, when the pulses are focused, they can reach intensities in the range of many TW/cm². In these regimes, laser-induced breakdown in many materials is common. As optical intensity increases, multiphoton absorption increases, creating free carriers which leads to further absorption. This is why shorter pulses tend to be more susceptible to damage, specifically having a damage threshold that scales with $\sqrt{\tau}$ where $\tau$ is the pulse duration. The absorption of energy leads to catastrophic chemical changes in the material, hence permanently damaging the material. Microscopic faults and impurities generally produce more free carriers. Since optical surfaces tend to have more defects than does the bulk and since Fresnel reflections off surfaces tend to increase the local optical intensity, damage occurs more often on surfaces. For example, in lithium niobate-based THz generation, damage occurs around a fluence of 50 mJ/cm² for 800 nm pulses on the surface where pump undergoes total internal reflection¹ as can be seen in Figure 2-3. Contrarily, bulk lithium niobate damage does not set in even at fluences of 186 mJ/cm² for sub-ps 1 micron pulses [31].

Another effect that occurs at high intensity is self-focusing due to the Kerr nonlinearity. In ultrafast amplifiers, the amount of accumulated nonlinearity is tracked by the B integral, which is defined as

$$B = \frac{2\pi}{\lambda} \int n_2 I(z) dz$$

(2.1)

This equation is simply the total nonlinear phase shift accumulated while propagating through the amplifier. Values of $B$ above 3 imply a risk of unwanted nonlinear lensing and spectral broadening. Excessive self-focusing can also lead to damage. The B integral is budgeted beforehand in the design of ultrafast amplifiers by calculating the integral through the cavity elements in all of the expected round-trips.

¹Based on personal experience
2.1.3 Chirped pulse amplification

The invention of chirped pulse amplification [106] started a new era in high energy solid state femtosecond lasers. It allowed for a way to bypass the damage and nonlinear issues in amplifying pulses well beyond their intensity thresholds in the amplifying materials.

Stretcher and compressor

The key to reducing the B integral and risk of damage in an amplifier is stretching out the pulse duration and compressing it after it has been amplified. Typically the stretcher induces a positive chirp and the compressor induces a negative chirp. An ideal CPA system has a zero total phase shift:

\[
\phi_{\text{stretcher}}(\omega) + \phi_{\text{amplifier}}(\omega) + \phi_{\text{compressor}}(\omega)
\]  

Practically, the stretcher and compressor can be designed to have equal and opposite dispersion \((\phi_{\text{stretcher}}(\omega) + \phi_{\text{compressor}}(\omega)) = 0\). However the amplifier dispersion, though small compared to the stretcher/compressor dispersions, can often be impossible to compensate perfectly. Instead, using the “knobs” on the compressor \((G\) and \(\theta\), see definitions below), the dispersion can be eliminated for the second and third order dispersion terms.

A basic compressor consisting of a grating pair [108] is shown in Figure 2-4a. A pulse enters from the top left, diffracts off of the first and second gratings, and is retroreflected back via a roof mirror. A pickoff mirror routes the compressed beam
onward. As evident pictorially from the illustration, the long wavelength part of
the pulse (denoted red) travels a longer path length than does the short wavelength
part (denoted blue). Consequently the longer wavelengths lag behind the shorter
wavelengths. The phase imparted by the compressor is

\[ \phi_{\text{compressor}} = 2 \frac{\omega}{c} \frac{G}{\cos \theta_i} \cos(\theta_i - \theta(\omega)) \]  (2.3)

where \( G \) is the slant distance between the gratings, \( \theta_i \) is the incidence angle, and
\( \theta \) is the diffraction angle off of the first grating. The second derivative of this phase
gives the group delay dispersion:

\[ \frac{d^2 \phi_{\text{compressor}}}{d\omega^2} = \frac{-4\pi^2 Gc}{\omega^3 d^2 \cos^3(\theta(\omega))} \]  (2.4)

where \( d \) is the grating groove separation. Note that the GDD is not a constant
function with respect to frequency, so third and higher order dispersion terms are also
present. Also note that Eq. 2.4 is always negative, so the compressor can only impart
negative \( \left( \frac{d^2 \phi}{d\omega^2} < 0 \right) \) dispersion onto its pulse regardless of the parameters. The term
‘compressor’ arises from the fact that most pulses stretched by propagation through
dielectric materials are normally (positively) chirped.

It is possible to make a stretcher that introduces the same phase as the compressor
but with opposite sign. This is done using a -1 magnification telescope along with
the grating, as shown in Figure 2-4b. This scheme is a practical implementation of
the more general stretcher-compressor duality, which is illustrated via a ray tracing
diagram in Figure 2-5 and derived in [124]. The rays approaching Grating 2 from
the bottom and diffracting toward plane S represent the compressor, while the rays
approaching Grating 2 from the top (after having reflected off the spherical mirror)
and diffracting toward plane Q represent the stretcher. In this pictoral demonstration
it can already be seen that the delay difference between \( \lambda_l \) and \( \lambda_s \) are equal and
opposite.
Figure 2-5: Ray tracing diagram to show stretcher and compressor duality. Adapted from [124].
Figure 2-6: Regenerative amplifier schematic. Adapted from [90].

Regenerative amplifier

The regenerative amplifier is a laser oscillator similar to the oscillators described in Section 2.1.1 but with an additional mechanism for switching pulses into and out of the cavity. Regenerative amplifiers typically have more than 6 orders of magnitude of gain with excellent TEM$_{00}$ output beam quality. The switching mechanism consists of an intracavity pockels cell and an external cavity Faraday rotator along with several polarization optics, as shown in Figure 2-6.

The mechanism for trapping a pulse inside the cavity is as follows. The verbs below describe the progression of a sample pulse.

1. Enters the schematic as a p-polarized pulse
2. Transmits through the external cavity thin film polarizer (TFP)
3. Converts to 45° polarization at the half wave plate ($\lambda/2$)
4. Converts to s-polarization at the Faraday rotator
5. Reflects off the intracavity TFP
6. Converts to left-circular polarization at the quarter wave plate ($\lambda/4$)
7. Transmits unchanged through the pockels cell (switched off)
8. Converts to p-polarization at the second pass of the $\lambda/4$
9. Transmits through the intracavity TFP

42
10. Gets amplified upon propagation through the amplifier crystal

11. Transmits through the intracavity TFP

12. Repeats Steps 6-11 indefinitely

The mechanism for switching an intracavity pulse to the output port is as follows.

We begin at Step 11 from the previous list and switch on the pockels cell birefringence such that it behaves as a half wave plate.

1. Converts to left-circular polarization at the $\lambda/4$

2. Converts to right-circular polarization at the pockels cell (switched on)

3. Converts to s-polarization at the second pass of the $\lambda/4$

4. Reflects off the intracavity TFP

5. Converts to $-45^\circ$ at the Faraday rotator

6. Converts to s-polarization at the $\lambda/2$

7. Reflects off the external cavity TFP toward the output port

Pulses are typically trapped inside the regenerative amplifier for hundreds of round-trips, as controlled by the pockels cell driver and a electronic delay generator. For example, in the Amplitude Systems HP2 laser that we used, it was trapped for 1800 ns with a round-trip time of 20 ns, making for about 90 round-trips.

2.1.4 Pulse characterization

Here we describe three basic methods used to characterize ultrashort pulses. In general, in order to measure an event in time, you need a shorter event. Femtosecond pulses are the shortest events in time that humans have been able to create. Therefore, the only way to measure the duration of a femtosecond pulse is to use another femtosecond pulse.
CHAPTER 2. BACKGROUND

Figure 2-7: Setup for intensity autocorrelation or FROG. Adapted from [109].

Intensity autocorrelation

Since the electric field of optical light is oscillating extremely fast (hundreds of THz), there are no devices which have a response time fast enough to directly measure the field, with the exception of a few nano-devices utilizing free electrons [94]. There are, however, several methods measure the intensity envelope or even the relative phase profile of a pulse. The idea behind intensity autocorrelation is to infer the pulse intensity envelope based on a cross-correlation of the pulse with itself, or the pulse’s autocorrelation. The autocorrelation of a pulse with intensity envelope \( I(t) \) is

\[
I_{ac} = \int_{-\infty}^{\infty} |I(t)I(t-\tau)|^2 \, dt
\]  

(2.5)

Unfortunately an infinite set of functions can have the same autocorrelation. Therefore measuring the autocorrelation itself does not give a definite pulse shape. If the pulse shape is known in advance, and a FWHM pulse duration is needed, then the intensity autocorrelation is a simple and suitable method of pulse characterization. Commonly, a Gaussian or Sech\(^2\) pulse shape is assumed. These have deconvolution factors of 0.7071 and 0.6482, respectively.

In practice, the autocorrelation can be obtained by second harmonic generation through a thin crystal, yielding a second harmonic that is proportional to the product between the two pulses integrated over time. A delay stage on one of the pulses controls \( \tau \). A typical setup of an intensity autocorrelation is shown in Figure 2-7.

It is also possible to obtain the intensity autocorrelation in a single shot, obfus-
Figure 2-8: Single shot intensity autocorrelation illustration. Adapted from [109].

Figure 2-9: Single shot intensity autocorrelation image. Adapted from .

cating the need for a motorized delay stage. One can replace the lens in Figure 2-7 with two mirrors, such that the beams are deflected toward the same direction but are not focused onto the SHG crystal. Using Figure 2-8 as a visual guide, one can see that the temporal delay between the two pulses is mapped to space (the vertical axis in Figure 2-8) when the pulses propagate through the nonlinear medium. The photons that are produced as the result of SHG between two pulses propagate along the bisector of the angle between the two pulses. A camera is then used to image the spatial profile of the SHG, which represents the intensity autocorrelation. Because the two input pulses travel at different angles, their individual SHG beams can be prevented from entering the camera, making the measurement background-free.

Single-shot autocorrelation requires knowledge of the temporal delay per pixel. To calibrate this value, the autocorrelation at two arbitrary, but known temporal delays is collected. The temporal delay is determined by dividing the spatial delay (which
can be measured by the Vernier scale on most translation stages) by the speed of light. Then the temporal delay spacing is divided by the pixel spacing between the peaks of the two autocorrelations to obtain the delay-per-pixel value.

\[
\frac{\text{Temporal delay}}{\text{Pixel spacing}} = \frac{2}{c} \times \frac{\text{Spatial delay}}{\text{Pixel spacing}}
\]

Note that the factor of two accounts for the round-trip time of a typical retroreflector-based delay stage as in Figure 2-7.

**FROG**

The FROG (Frequency Resolve Optical Gating) is the simplest and most widely-used method to characterize the full temporal shape of an ultrafast pulse including phase information. In practice, it is only a slight change to the intensity autocorrelation, but the implications are large. Instead of using a photodetector to measure the SHG intensity, a spectrometer is used to measure the SHG spectrum. The obtained spectra can be plotted as a function of the delay \(\tau\) to yield a 2D spectrogram. This spectrogram contains all the information to determine the temporal shape of the pulse without ambiguity, except for the absolute phase and temporal direction. Computing temporal shape from the FROG trace involves an interative phase retrieval algorithm, but software packages abound for use by the nonspecialist, such as Femtosoft [61]. Figure 2-7 shows the FROG setup and Figure 2-10(b) shows an example of a FROG trace.

In order to verify the accuracy of the phase retrieval, the retrieved spectrum is compared with the measured spectrum. For complex pulses, it is often difficult to obtain a good FROG trace. Several practical tips are described below.

- The the two beams arriving on the SHG crystal must overlap well at their foci.

To do this, one should place a camera near the location of the SHG crystal
and adjust the pointing of the beams as well as the longitudinal position of the camera such that the beam foci overlap. Next the SHG crystal’s longitudinal position can be adjusted to maximize the SHG signal.

- The generated SHG light must *all* be collected. This can sometimes be more difficult than expected because of the high divergence of the SHG signal. A large NA lens is recommended for imaging the SHG crystal into the FROG spectrometer.

- The beam must not have coupled spatiotemporal features such as angular dispersion or spatial chirp. If so the foci of the two beams on the SHG crystal will overlap in such a way that the autocorrelation function will be corrupted. A good indication of this happening is when the FROG trace is horizontally asymmetric as shown in Figure 2-10. An example of this is illustrated in Figure 2-11 where angularly dispersed beams are focused onto the SHG crystal (angular dispersion becomes spatial chirp at the focus). The higher optical frequency portions of the two beams ($\omega_0 + \delta\omega$) will overlap at a certain delay ($\tau_0 - \delta\tau$) producing SHG at $2(\omega_0 + \delta\omega)$, while the lower optical frequency portions ($\omega_0 - \delta\omega$) will overlap at another delay ($\tau_0 + \delta\tau$) producing SHG at $2(\omega_0 - \delta\omega)$. As shown in Figure 2-11, this leads to an asymmetric, and thus non-retrievable, FROG trace.

- Pre-processing of the FROG trace prior to phase retrieval is very important. It is important to crop parts of the FROG trace that contain no information and diligently apply noise reduction filters. Most of these functions are built into FROG software such as Femtosoft [61].

**Electro-optic sampling**

Electro-optic (EO) sampling is a method to directly measure the electric field of long wavelength radiation, such as mid-infrared, far-infrared, or THz beams, using a short
wavelength ultrashort pulse (\(\sim \)100 fs). EO sampling relies on the electro-optic effect, which is utilized in Pockels cells and electro-optic modulators (EOM).

In the electro-optic effect, a static electric field induces birefringence in an optical medium proportional to the applied field amplitude. Inversely, that applied field amplitude can be measured by probing the field-induced birefringence. In EO sampling, the THz electric field is treated as a static field since its period is much longer than the optical probe pulse duration. Hence, the THz and optical fields must propagate with similar velocities inside the EO crystal in order for this approximation to be valid. To measure the full THz waveform, THz field measurements are made at multiple delays between the optical probe and the THz pulse to reveal the THz waveform.

Figure 2-12 illustrates the typical EO sampling setup and operation. A THz pulse and IR probe pulse overlapped in time are impinging an EO crystal (typically ZnTe or GaP), both with linear polarization. Inside the crystal, the THz induces birefringence which causes the IR field to experience a difference in phase between the polarization...
components parallel to the crystal’s a-axis and c-axis. This phase retardation results in an elliptical IR polarization at the output of the crystal.

The phase retardation is linearly proportional to the THz electric field by the following equation [70].

$$\Delta \phi = \frac{\omega L}{c} n^3_0 r_{41} E_{THz}$$ \hspace{1cm} (2.8)

Here, $L$ is the length of the crystal, $r_{41}$ is the electro-optic coefficient of the crystal along the axis parallel to the THz polarization. In the case of ZnTe, $r_{41} = 4 \text{ pm/V}$.

To measure the phase retardation, the IR pulse is then transmitted through a $\lambda/4$ plate which transforms its polarization to nearly circular (perfectly circular without the THz). Finally, a Wollaston prism or polarizing beam splitter (does not matter) splits s- and p-polarization components of the beam and their intensity difference is measured by a balanced photodetector. The need for the $\lambda/4$ plate is not immediately obvious, since the THz-induced birefringence already causes slight elliptical polarization in the probe beam. However, the intensity in the s- and p-polarization induced by the phase retardation is proportional to $\cos \Delta \phi$, which is a nonlinear function. With the $\lambda/4$ plate, the intensities are proportional to $\sin \Delta \phi \approx \Delta \phi$ for small angles.

The intensities on the balanced photodetector of the s- and p-polarizations of a
beam with total intensity of $I_0$ are

\begin{align}
I_x &= \frac{I_0}{2} (1 - \sin \Delta \phi) \approx \frac{I_0}{2} (1 - \Delta \phi) \quad \text{(2.9)} \\
I_y &= \frac{I_0}{2} (1 + \sin \Delta \phi) \approx \frac{I_0}{2} (1 + \Delta \phi) \quad \text{(2.10)}
\end{align}

For small angles where $\Delta \phi \ll 1$, the signal of the balance photodetector is proportional to the THz amplitude.

\[ I_s = I_y - I_x = I_0 \Delta \phi = \frac{I_0 \omega L}{c} n_0^3 r_{41} E_{THz} \quad \text{(2.11)} \]

Here we discuss a few experimental tips in the implementation of an EO sampling setup.

- **Polarization orientation:** The THz field should be oriented parallel to either the a- or c-axis of the EO crystal, whichever has a larger nonlinear coefficient. The probe field should be oriented at a 45° angle to the THz field to make it most sensitive to the THz-induced birefringence.

- **Back reflections:** Often in the EO sampling waveform, it will appear as if there is a second, attenuated THz pulse following several picoseconds behind the first. This second pulse is not real, but is rather caused by the second pass of the probe pulse through the EO crystal. For example, suppose the crystal thickness is $L = 200 \ \mu m$ with an index of $n = 2.78$. A probe pulse undergoing a round trip through the crystal (after undergoing Fresnel reflections at both facets) will take $2nL/c = 3.7 \ \text{ps}$. Thus, the ‘ghost’ THz pulse will appear at 3.7 ps in delay behind the main one. This issue can be solved by using an electro-optic crystal which is bonded to an electro-optically inactive wedge which disallows second passes of the probe pulse through the crystal at the same angle as the first.

- **Water absorption:** Water vapor has several absorption lines in the THz frequency range, with one particularly absorptive one at 0.5 THz. Thus, the THz
spectrum will often have dips in it, which in the time domain are represented by small oscillations following the main single-cycle pulse. This issue can be solved by nitrogen purging the entire path of THz propagation.
2.2 Optical parametric processes

Optical parametric processes are a class of light-matter interactions in which the quantum state of the material is unchanged. In other words, the process is lossless; there is no energy or momentum transferred from the photons to the material. Optical parametric processes tend to occur nearly instantaneously. In linear optics, parametric processes include refraction, diffraction, and elastic scattering, whereas non-parametric processes include absorption, Raman scattering, and fluorescence. In nonlinear optics, parametric processes include three-wave-mixing, four-wave-mixing, or higher order harmonic generation and self-phase modulation, whereas non-parametric processes include Raman amplification, multiphoton absorption, or saturable absorption. A sampling of second-order parametric nonlinear optical processes is shown in Figure 2-13. In the figure, various sets of wavelengths $\omega_1$, $\omega_2$, and $\omega_3$ are used to generate other sets. The relation $\omega_3 = \omega_2 + \omega_1$, however, must always hold. By convention, the subscripts of $\omega$ are in order of increasing frequency, but we will ignore this convention briefly in the next section.
2.2.1 Difference frequency generation

We now analyze the nonlinear parametric process of difference frequency generation (DFG). DFG occurs when a strong pump at frequency $\omega_3$ and a weak seed laser at $\omega_1$ generate a field at $\omega_2$ which originally was non-existent, $A_2(0) = 0$. See Figure 2-13. For simplicity of analysis, suppose $\omega_3$ is undepleted by the nonlinear interaction, so that the amplitude $A_3$ is constant. This is valid in the case when there is a strong pump, weak seed, and low conversion efficiency. In the example of THz generation, $\omega_3$ represents the pump laser’s center frequency, $\omega_1$ represents a weak laser with slightly longer wavelength than the pump (this can be a separate laser or simply one frequency component within the pump laser’s bandwidth), and $\omega_2 = \omega_3 - \omega_1$ represents the generated THz frequency.

First, note that any arbitrary plane wave laser pulse propagating along $z$ can be expressed as the product of an envelope $A(t, z)$ and a carrier $e^{j(\omega_0 t - k_0 z)}$.

$$E(t, z) = A(t, z)e^{j(\omega_0 t - k_0 z)} \quad (2.12)$$

According to Fourier theory, this equation in turn be expressed alternatively as the sum or integral of monochromatic waves:

$$E(t, z) = \int_{-\infty}^{\infty} E(\omega, z)e^{j\omega t}d\omega \quad (2.13)$$

Where the amplitude of each monochromatic wave is

$$E(\omega, z) = A(\omega, z)e^{-jk(\omega)z}. \quad (2.14)$$

Maxwell’s equation with nonlinear polarization and the slowly varying amplitude approximation yield the following set of coupled differential equations [7]. See Section
2.2.4 for the derivation of these nonlinear equations.

\[
\frac{dA_1}{dz} = \frac{2i\omega_1^2 d_{\text{eff}}}{k_1c^2} A_3^* A_2 e^{i\Delta k z} \quad (2.15)
\]

\[
\frac{dA_2}{dz} = \frac{2i\omega_2^2 d_{\text{eff}}}{k_2c^2} A_3^* A_1 e^{i\Delta k z} \quad (2.16)
\]

where \(k_i = n_i\omega_i/c\) are the amplitudes and wavevectors of the various frequencies and \(\Delta k\) is the phase mismatch, \(\Delta k = k_1 + k_2 - k_3\). In the case of perfect phase matching (\(\Delta k = 0\)), the solution to Eq. 2.16 is

\[
A_1(z) = A_1(0) \cosh \kappa z \quad (2.17)
\]

\[
A_2(z) = i \sqrt{\frac{n_1\omega_2}{n_2\omega_1}} \frac{A_3}{|A_3|} A_1^*(0) \sinh \kappa z \quad (2.18)
\]

where

\[
\kappa^2 = \frac{4d_{\text{eff}}^2 \omega_1^2 \omega_2^2}{k_1k_2c^4 |A_3|^2} \quad (2.19)
\]

We can see from this result that both \(|A_1|\) and \(|A_2|\) grow monotonically at first, and eventually, exponentially with distance. The generated field \(A_2\) has a phase that is proportional to the difference between \(A_1\) and \(A_3\): \(\angle A_2 = \pi + \angle A_3 - \angle A_1\). Finally, the rate of growth is proportional to the pump intensity \(|A_3|^2\), nonlinear coefficient \(d_{\text{eff}}\), and the product \(\omega_1\omega_2\).
2.2.2 Manley-Rowe relations

The Manley-Rowe relations state that in any second order process, the following three quantities remain invariant.

\[ M_1 = \frac{I_2}{\omega_2} + \frac{I_3}{\omega_3}, \quad M_2 = \frac{I_1}{\omega_1} + \frac{I_3}{\omega_3}, \quad M_3 = \frac{I_1}{\omega_1} - \frac{I_2}{\omega_2} \] (2.20)

Since \( \frac{I_i}{\omega_i} \) is proportional to the photon count of component \( i \), the Manley-Rowe relations can be interpreted to be the principle of photon conservation. For example, if the pump photons, \( I_3/\omega_3 \), are fully depleted, the number of photons at \( \omega_1 \) and \( \omega_2 \) would be equal to \( I_1/\omega_1 + I_2/\omega_2 + I_3/\omega_3 \), respectively. The Manley-Rowe places an upper limit to the efficiency of a DFG process. In the best case scenario, all pump photons are converted into difference-frequency photons, yielding a power conversion efficiency of

\[ \frac{I_{out}^2}{I_{in}^3} = \frac{\omega_2}{\omega_3} \quad \text{or} \quad \frac{I_{out}^1}{I_{in}^3} = \frac{\omega_1}{\omega_3} \] (2.21)

If the difference in the pump and generated frequency is large, even this optimal efficiency can be small. In Chapter 3, we will introduce cascading, a phenomenon which allows the efficiency to surpass that prescribed by the Manley-Rowe relation.

2.2.3 Simple depleted calculation of optical rectification

In addition to the undepleted calculation in Section 2.2.1, we can also make a depleted calculation of the nonlinear process via a numerical ODE solver. The ODE solves the general nonlinear wave propagation equation, derived later in Eq. 2.39. Figure 2-15 shows the evolution of five different frequency components as intrapulse three-wave-mixing effects occur between all of them simultaneously.

Note that as the wave propagates, the two components which initially start with all the power (375 and 375.5 THz) start transferring their power to the difference frequency at 0.5 THz. At small signals (short propagation lengths), this 0.5 THz component grows exponentially as we saw in the undepleted calculation. However,
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Figure 2-15: Phase-matched three-wave-mixing evolution of five different frequencies. Initially, power is split between two adjacent infrared frequencies (375 and 375.5 THz). As the wave propagates, the THz idler component (0.5 THz), DFG IR component (374.5 THz), SFG IR component (376 THz) are generated and amplified. The DFG IR component. Meanwhile the higher frequency fundamental component (375.5 THz) is depleted while the lower frequency fundamental component (375 THz) is temporarily amplified and then depleted.

as its signal grows larger, its growth starts to slow down to linear pace. This slowdown occurs not only because the pump (375.5 THz) and signal (375 THz) fields are depleted, but also because the 0.5 THz component itself begins to donate power to the 376 THz component via sum frequency generation (SFG) with the pump (375.5 THz) component. As the number of spectral components grows, the dynamics can become very complex and must be solved numerically.

2.2.4 Ab-initio formulation of three-wave-mixing physics

Although an exact analytical solution can be obtained for difference frequency generation in the simple conditions of monochromatic, collinear plane waves, in general, solutions involving ultrafast pulses, which have many frequency components, will re-
quire computer simulations. Here we attempt to derive a mathematical formulation for any two-dimensional three-wave-mixing parametric process in a form that can be computed by a computer’s ODE solver.

**Nonlinear polarization**

The first step in analyzing a nonlinear process is to derive the nonlinear polarization. In Section 2.2.1, the nonlinear polarization was a single term proportional to the product of the two frequency components. In general, there will be more than two frequency components present, so the nonlinear polarization can be expressed as on the integral of all the possible products of frequency components whose frequency sum or difference is the frequency of interest \( \omega \).

To show this, we start with the general form of the nonlinear polarization \( P \) and derive it as two integrals containing either all sum frequency generation (SFG) or all difference frequency generation (DFG) processes.

\[
P(\omega) = 4\varepsilon_0 d_{\text{eff}} \int_{-\infty}^{\infty} E(\omega - \Omega)E(\Omega)d\Omega \tag{2.22}
\]

\[
= 4\varepsilon_0 d_{\text{eff}} \int_{-\infty}^{0} E(\omega - \Omega)E(\Omega)d\Omega + 4\varepsilon_0 d_{\text{eff}} \int_{0}^{\infty} E(\omega - \Omega)E(\Omega)d\Omega \tag{2.23}
\]

\[
= -4\varepsilon_0 d_{\text{eff}} \int_{0}^{\infty} E(\omega + \Omega')E(-\Omega')d\Omega' + 4\varepsilon_0 d_{\text{eff}} \int_{0}^{\infty} E(\omega - \Omega)E(\Omega)d\Omega \tag{2.24}
\]

\[
= 4\varepsilon_0 d_{\text{eff}} \int_{0}^{\infty} E(\omega + \Omega')E^*(\Omega')d\Omega' + 4\varepsilon_0 d_{\text{eff}} \int_{0}^{\infty} E(\omega - \Omega)E(\Omega)d\Omega \tag{2.25}
\]

The first term represents all DFG processes and the second represents all SFG processes.

Because THz pulses are the subject of this thesis, we will now apply this general form of the nonlinear polarization toward THz generation. We will use ‘THz’ and ‘idler’ interchangeably. However, the idler need not be applied to THz frequencies only. It can also be applied to, e.g., mid-IR frequencies or the lower frequency component in any three-wave-mixing process.

The nonlinear polarization for the idler at THz frequencies (denoted here as \( \Omega \)) is
\[ P(\Omega) = 4\epsilon_0 d_{\text{eff}} \int_0^\infty E(\omega + \Omega) E^*(\omega) d\omega \quad (2.26) \]

\[ = 4\epsilon_0 d_{\text{eff}} \int_0^\infty [A(\omega + \Omega)e^{-jk_z(\omega+\Omega)}][A^*(\omega)e^{jk_z(\omega)}]d\omega \quad (2.27) \]

\[ = 4\epsilon_0 d_{\text{eff}} \int_0^\infty A(\omega + \Omega) A^*(\omega) e^{j[-k_z(\omega+\Omega) + k_z(\omega)]}d\omega \quad (2.28) \]

The nonlinear polarization at optical frequencies (denoted here as \( \omega \)) in Eq. 2.29 contains two terms because of the ambiguity of whether the generated frequency corresponds to the ‘pump’ or ‘signal’ in the traditional sense. The first term is difference frequency generation between a ‘pump’ optical photon and ‘idler’ THz photon to generate a ‘signal’ photon. The second term is sum frequency generation between a ‘signal’ optical photon and ‘idler’ THz photon to generate a ‘pump’ photon.

\[ P(\omega) = 4\epsilon_0 d_{\text{eff}} \int_0^\infty E(\omega + \Omega) E^*(\Omega) d\Omega \quad (2.29) \]

\[ + 4\epsilon_0 d_{\text{eff}} \int_0^\infty E(\omega - \Omega) E(\Omega) d\Omega \quad (2.30) \]

\[ = 4\epsilon_0 d_{\text{eff}} \int_0^\infty A(\omega + \Omega) A^*(\Omega) e^{j[-k_z(\omega+\Omega) + k_z(\Omega)]}d\Omega \quad (2.31) \]

\[ + 4\epsilon_0 d_{\text{eff}} \int_0^\infty A(\omega - \Omega) A(\Omega) e^{j[-k_z(\omega-\Omega) - k_z(\Omega)]}d\Omega \quad (2.32) \]

**2D wave equation for nonlinear media**

Armed with the expression for the nonlinear polarization, we now proceed to deriving the wave equation for nonlinear media. To make it general for noncollinear geometries, we derive it in 2D rather than 1D. Here, for simplicity, we consider only the scalar wave equation and assume a fixed polarization direction for all the waves. This assumption is valid for the tilted pulse front method of THz generation (described in Chapter 3), where only the nonlinear polarization along the c-axis (\( d_{\text{eff}} = d_{33} \) nonlinear susceptibility) is activated.

In the most general form, nonlinear optical phenomena can be described as solu-
tions to the wave equation. A simplifying approximation for the case of plane waves or waves with a slowly varying temporal envelope is used to make $\nabla \cdot E = 0$. Assuming monochromatic waves with angular frequency $\omega$ traveling predominantly in the $z$ direction, the 2D wave equation can be expressed as the Helmholtz equation driven by nonlinear polarization $P$.

$$\left[ \nabla^2 + k(\omega)^2 \right] E(\omega, x, z) = -\frac{\omega^2}{\epsilon_0 c^2} P(\omega, x, z) \quad (2.33)$$

Extending this derivation to 3D can be done straightforwardly by treating the additional transverse dimension, $y$, the same way as $x$ is treated here.

This is a second order partial differential equation. The polarization term makes the equation nonlinear and coupled, leading to difficult numerical problem. Ravi et al. [98] introduced the method of spatial Fourier decomposition which reduces this problem to a system of ordinary differential equations.

The solution of Eq. 2.33 can be decomposed into its Fourier spatial frequencies in the $x$ direction. By definition of the inverse Fourier transform, $E(\omega, x, z)$ can be expressed as the sum of its Fourier components in $x$.

$$E(\omega, x, z) = \int_{-\infty}^{\infty} E(\omega, k_x, z) e^{jk_x x} dk_x \quad (2.34)$$

Differentiating this twice with respect to $x$ gives

$$\frac{\partial^2 E(\omega, x, z)}{\partial x^2} = \int_{-\infty}^{\infty} -k_x^2 E(\omega, x, z) dk_x \quad (2.35)$$

Using the relation $k^2 = k_z^2 + k_x^2$, the Helmholtz equation (Eq. 2.33) now can be rewritten in the Fourier domain as

$$\left[ \frac{\partial^2}{\partial z^2} + k_z(\omega)^2 \right] E(\omega, k_x, z) = -\frac{\omega^2}{\epsilon_0 c^2} P(\omega, k_x, z) \quad (2.36)$$

We assume solutions of the form of a pulse with envelope $A(\omega, k_x, z)$ propagating in the $z$ direction.


\[ E(\omega, k_x, z) = A(\omega, k_x, z)e^{k_zz} \]  

(2.37)

The second derivative of this solution can be written as

\[ \frac{\partial^2 E}{\partial z^2} = \frac{\partial^2 A}{\partial z^2}e^{-jk_zz} - k_z^2 A e^{-jk_zz} - 2jk_z \frac{\partial A}{\partial z}e^{-jk_zz} \]  

(2.38)

We now plug Eqs. 2.37 and 2.38 into Eq. 2.36. The second term in the right hand side of Eq. 2.38 will cancel out with the second term in the left hand side of Eq. 2.36. The first term in the right hand side of Eq. 2.38 will be neglected because of the slowly varying envelope approximation, \(|k_z A| \gg \left| \frac{d^2 A}{dz^2} \right|\). This approximation says that the envelope varies slowly compared to the rate of field oscillations (third term).

The resulting equation is

\[ \frac{\partial A(\omega, k_x, z)}{\partial z} = -j \frac{\omega^2}{2k_z(\omega)\varepsilon_0 c^2} P(\omega, k_x, z)e^{jk_z(\omega)z} \]  

(2.39)

This is the general nonlinear wave propagation equation.

**System of coupled nonlinear equations**

This derivation of the nonlinear wave propagation equation (Eq. 2.39) could apply for any nonlinear process by plugging in the polarization (Eq. 2.22) into the nonlinear wave propagation equation (Eq. 2.39).

\[ \frac{\partial A(\omega, k_x, z)}{\partial z} = -j \frac{\omega^2}{2k_z(\omega)\varepsilon_0 c^2} \int_{-\infty}^{\infty} E(\omega - \Omega)E(\Omega)d\Omega \]  

(2.40)

Here, we apply for the case of THz generation by optical rectification.

\[ \frac{\partial A_{\text{THz}}(\Omega, k_x, z)}{\partial z} = -j \frac{\Omega^2}{2k_z(\omega)\varepsilon_0 c^2} P_{\text{THz}}(\Omega, k_x, z)e^{jk_z(\Omega)z} \]

\[ \frac{\partial A_{\text{IR}}(\omega, k_x, z)}{\partial z} = -j \frac{\omega^2}{2k_z(\omega)\varepsilon_0 c^2} P_{\text{IR}}(\omega, k_x, z)e^{jk_z(\omega)z} \]
Here the nonlinear polarizations are those derived in Eqs. 2.26 and 2.29. Plugging them in gives

\[
\frac{\partial A_{\text{THz}}(\Omega, k_x, z)}{\partial z} = -j \frac{2\Omega^2}{k_z(\Omega)c^2} d_{\text{eff}} \int_0^\infty A_{\text{IR}}(\omega + \Omega, k_x, z) A^*_\text{IR}(\omega, k_x, z) e^{j[-k_z(\omega+\Omega)+k_z(\Omega)+k_z(\Omega)]z} d\omega
\]

(2.41)

\[
\frac{\partial A_{\text{IR}}(\omega, k_x, z)}{\partial z} = -j \frac{2\omega^2}{k_z(\omega)c^2} d_{\text{eff}} \int_0^\infty A_{\text{IR}}(\omega + \Omega, k_x, z) A^*_\text{THz}(\Omega, k_x, z) e^{j[-k_z(\omega+\Omega)+k_z(\Omega)+k_z(\Omega)]z} d\Omega
\]

(2.42)

\[
- j \frac{2\omega^2}{k_z(\omega)c^2} d_{\text{eff}} \int_0^\infty A_{\text{IR}}(\omega - \Omega, k_x, z) A_{\text{THz}}(\Omega, k_x, z) e^{j[-k_z(\omega-\Omega)-k_z(\Omega)+k_z(\omega)]z} d\Omega
\]

(2.43)

This system of equations is general and can be applied to any arbitrary 2D geometry since the beams can be decomposed to \(\omega\) and \(k_x\) components. The second equation in Eq. 2.43 explains the generation of IR photons via both sum and difference frequency generation. The first equation, on the other hand, only includes DFG components since it is reasonable to assume that any SFG between two THz photons producing a higher frequency THz photon would be both phase-mismatched and too weak to be considered. The subscripts (THz, IR) are only for clarification and play no essential role in differentiating the components, since that is already accomplished by the frequency argument. They do matter, however, when their values are encoded into separate numerical arrays in a simulation.

While this system of equations fully describes the three-wave-mixing physics, other important terms affecting the evolution of the generated wave (such as absorption or noncollinear propagation) have been left out for simplicity. These will be added in when we study the particular case of THz generation by optical rectification in Chapter 3.
2.3 Electron beams

Optical THz pulses possess an unique advantage for electron acceleration because of their high achievable electric fields, which serve to accelerate an electron beam as quickly as possible to semirelativistic velocities to avoid the beam-degrading effects of space charge at low velocities. We start our section on discussing the measure of electron beam quality, also known as emittance.

2.3.1 Emittance

In an electron beam or bunch, each particle is fully represented by a point in six-dimensional phase space with coordinates \((x, p_x, y, p_y, z, p_z)\). Instead of momentum \(p\), typically the variable \(x'\) is substituted for \(p_x\)

\[
x' = \frac{dp_x}{dp_z}.
\]  

(2.44)

Typically, motion of particles in the three dimensions is decoupled, so the phase space along one of the dimensions can be visualized as in Figure 2-16.

In most accelerator systems, the primary objective is to maximize the brightness of the delivered beam. The brightness, defined below, determines how densely a beam of electrons can be focused on a particular spot or how well-collimated the beam can be. Both properties are of utmost importance to accelerator applications such as ultrafast electron diffraction or x-ray generation.
\[ B = \frac{N_e}{\epsilon_x \epsilon_y \epsilon_z}. \]  

(2.45)

Since the number of electrons \( N_e \) in the bunch is fixed by the cathode material and photoemission UV laser pulse, the challenge of an accelerator is minimizing the emittances \( \epsilon \) in each of the axes \( x, y, z \). Emittance is defined as the phase space area occupied by the electron bunch, divided by \( \pi \)

\[ \epsilon_x = \frac{A x x'}{\pi}. \]  

(2.46)

Liouville’s theorem [115] states that the density of particles in phase space doesn’t change along a beam transport line so long as it experiences only conservative fields (drift spaces, magnetic fields). The normalized emittance, \( \beta \gamma \epsilon_x \) is conserved during acceleration and cannot be decreased by electromagnetic fields, only decreased.

While it is true that the upper limit for brightness is set by the normalized emittance of the electron bunch at the time of its production, in the real world the emittance that is typically measured and used is the RMS emittance. RMS emittance is a statistical quantity of a collection of particles defined as

\[ \epsilon_{n,x} = \frac{1}{mc} \sqrt{\langle x^2 \rangle \langle x'^2 \rangle - \langle xx' \rangle^2}. \]  

(2.47)

At the beam waist, this reduces to

\[ \epsilon_{n,x} = \frac{1}{mc} \sigma_x \sigma_{p_x}. \]  

(2.48)

This definition of emittance measures how closely the phase space of the bunch represents an ellipse as well as how much volume that ellipse occupies. This can be seen by the ellipse equation

\[ \langle x'^2 \rangle x^2 + \langle x^2 \rangle x'^2 - 2 \langle xx' \rangle xx' = \epsilon_{rms}^2. \]  

(2.49)

In this definition, the emittance can be decreased by acceleration but not by conservative forces. The area of the ellipse can be preserved but not decreased by
beamline elements and the shape and orientation of the ellipse can be modified while preserving the area. Figure 2-17 shows the transformation of the phase space distribution over a lens and drift region.

**Role of emittance in applications**

In electron microscopes, lower emittances allow for higher resolutions, as implied by Eq. 2.48. In ultrafast electron diffraction, molecules are probed with an electron bunch and their structure can be interpreted from the resulting interference pattern. The quality of the interference pattern however depends on the transverse spatial coherence length of the bunch. At the beam waist, the coherence length is inversely proportional to the transverse emittance:

\[
L_c = \frac{\hbar \sigma_r}{mc \epsilon_r}
\]  

(2.50)

where \( \sigma_r \) is the transverse position spread and \( \epsilon_r \) is the transverse emittance.

Likewise, in free-electron lasers, smaller emittances would mean smaller facilities and lower costs. The power gain \( \frac{P(z)}{P_0} \) is set by the FEL gain length, which is proportional to the cubic root of the emittance:

\[
L_g \propto \gamma \lambda_u^{1/3} \epsilon_x^{1/3}.
\]  

(2.51)
2.3.2 Space charge

Space charge is a collective and statistical effect in which the electron bunch generates a “self-field” which acts upon itself. Over time, this self-field will act to expand and spoil the beam. Further, this self-field can also act to limit photoemission at the photocathode. The dependencies and properties of the self-field are discussed next.

Transverse self-field

For a beam with Gaussian linear charge density $\lambda$, it can be shown that the transverse space charge fields at the beam core ($x \ll \sigma_x$ and $y \ll \sigma_y$) are

\begin{align*}
E_x &= \frac{1}{2\pi \epsilon_0} \frac{\lambda}{\sigma_x (\sigma_x + \sigma_y)} x \quad (2.52) \\
E_y &= \frac{1}{2\pi \epsilon_0} \frac{\lambda}{\sigma_y (\sigma_x + \sigma_y)} y \\
B_x &= -\frac{\mu_0}{2\pi} \frac{\lambda \beta c}{\sigma_y (\sigma_x + \sigma_y)} y \\
B_y &= \frac{\mu_0}{2\pi} \frac{\lambda \beta c}{\sigma_y (\sigma_x + \sigma_y)} x. \quad (2.55)
\end{align*}

Combining Equations 2.52, we derive the radial fields for a beam with radius $r_0$:

\begin{align*}
E_r(z) &= \frac{1}{4\pi \epsilon_0} \frac{2\lambda(z) r}{r_0^2} \\
B_\phi(z) &= -\frac{\mu_0}{4\pi} \frac{2\lambda(z) \beta r}{r_0^2}. \quad (2.57)
\end{align*}

Plugging these fields into the Lorentz force equation: $F = q(E + v \times B)$, we obtain the following, remembering that $1 - \beta^2 = \frac{1}{\gamma^2}$.

\begin{align*}
F_x &= qE_x(1 - \beta^2) \propto \frac{\lambda x}{\gamma^2} \quad (2.58) \\
F_y &= qE_y(1 - \beta^2) \propto \frac{\lambda y}{\gamma^2}. \quad (2.59)
\end{align*}
Thus the space charge force is most intense at nonrelativistic energies and drops quickly as the particles are accelerated. Further, the space charge force exerted on an electron increases linearly with its distance from the beam center.

**Longitudinal self-field**

Likewise the longitudinal field can be derived by solving Stokes law about a small section of the beam traveling inside a metal cylindrical vacuum chamber as shown in Figure 2-18.

\[
\int E \, ds = -\frac{\partial}{\partial t} \int B \, dA \quad (2.60)
\]

This expression can be solved by using the radial and azimuthal expressions for the electric and magnetic field from Equation 2.52. To a first order Taylor expansion for the linear charge density \( \lambda(z) \), the longitudinal electric field at the axis of the beam is

\[
E_{z0} = E_{zw} - \frac{q}{4\pi \epsilon_0 \gamma^2} \left( 1 + 2 \ln \frac{r_w}{r_0} \right) \frac{\partial \lambda}{\partial z} \quad (2.61)
\]

\[
\propto \frac{1}{\gamma^2} \frac{\partial \lambda}{\partial z} \quad (2.62)
\]

where \( E_{zw} \) and \( r_w \) are the electric field and the radius of the wall, respectively. Again the longitudinal self field drops with the beam energy \( \gamma \) squared.
Photocurrent suppression

Space charge can also limit the charge per bunch that can be emitted inside the photoinjector. When a certain amount of electrons have been emitted from the surface of a flat photocathode, the field created by that bunch can suppress further emission of electrons, even if there are still photoemitting (e.g. UV) photons impinging the surface.

Consider that a uniform pancake shaped electron bunch has been emitted from a flat cathode, as shown in Figure 2-19, and is subject to an accelerating field $E_{acc}$. That bunch itself generates two fields that act on the cathode surface: the space-charge field $E_{sc}$, which comes directly from the Coulomb repulsion of the bunch, and the image charge field $E_{im}$, which is due to charges on the metal surface redistributing themselves to minimize electric gradients inside the conductor. By symmetry, the image charge field is equal to the space-charge field. Now consider a test electron sitting at the cathode surface. it will not escape the surface if the space charge force from electron bunch exceeds the accelerating force from the RF. This *space-charge-limited* regime occurs when

$$E_{sc} + E_{im} = 2E_{sc} = E_{acc}. \quad (2.63)$$

Suppose we have an accelerating field $E_{rf}$ and a uniform UV beam with radius $R$ illuminating the photocathode surface. Then the total charge we would expect to emit is

$$Q = CV = \left(\epsilon_0 \frac{A}{d}\right) V = \epsilon_0 \pi R^2 E_{sc} = \epsilon_0 \pi R^2 \frac{E_{acc}}{2}, \quad (2.64)$$

with a density of

$$\sigma = \epsilon_0 \frac{E_{acc}}{2} \quad (2.65)$$

Therefore, for a fixed spot size of the photoemitter, the total charge produced from a photocathode is proportional to the electric field at the surface of the emitter,
during operation in the space-charge-limited regime. If the photoemitter energy is small and Eq. 2.63 is not met \((2E_{sc} \text{ never exceeds } E_{acc})\), then operation is not in the space-charge-limited regime; all electrons will be emitted regardless of the field strength–there would be no dependence of \(Q\) on \(E_{acc}\). Chapter 4 presents experimental evidence of this phenomenon.

Having considered the space charge forces on photoemission as well as propagating electron beams, it’s clear that in all stages of the photoinjector, higher accelerating field promotes higher charge density either through enhanced emission yield or space charge reduction.

### 2.3.3 Linear accelerator

Electrons are accelerated by interaction with time-varying electromagnetic fields. In order for acceleration to occur, such fields must have an electric field component in the direction of electron propagation, so that the electron can “ride along” with its accelerating wave. Plane waves in vacuum have only transverse field components. We must therefore use boundary conditions to configure the field distributions such as to create a longitudinal component.

One configuration is the cylindrical waveguide shown in Figure 2-20. When electromagnetic fields impinge the waveguide from one end (possibly with the help of an adiabatic taper section), they excite one or more of the waveguide spatial modes. Free-space linearly polarized THz beams, such as those generated by optical rectifica-
tion, tend to couple into the TE_{11}, TE_{12}, and TM_{11} modes which when superimposed do not have a suitable longitudinal accelerating component [32]. Instead, the beam must be converted to a free-space TM_{01} mode via, e.g., a segmented waveplate [82] before being coupled into the waveguide.

Without proof (consult, e.g., [115]), the longitudinal component of the axially-symmetric modes (TM_{0m}) takes the form

\[ E_z = E_0 J_m(k_{cr}r) e^{i(\omega t - m\phi - k_z z)}, \]

(2.66)

where \( m \) is the mode number corresponding to the number of nodes in the \( r \) direction. The lowest order mode (\( m = 0 \)) has the highest on-axis longitudinal field and is the most useful mode for acceleration.

Here \( k_z \) is the propagation constant, given by

\[ k_z = \sqrt{k^2 - k_{cr}^2}, \quad \text{where} \quad k = \frac{\omega_0}{c} n \]

(2.67)

The refractive index is \( n = 1 \) in this case because the waveguide is hollow. It’s easy to see that when \( k = k_{cr}, \) \( k_z \) goes to zero and propagation ceases, giving way to an imaginary \( k_z \) which is synonymous with attenuation. Thus, \( k_{cr} \) corresponds to the \textit{cutoff frequency}, given by

\[ \omega_c = k_{cr} \frac{c}{n} \]

(2.68)

In order to satisfy the boundary condition that the longitudinal field vanishes at
the radius, \( a \), of the waveguide, we set \( k_c a = 2.405 \) because that is the location of the first node of the Bessel function \( J_0(k_c a) \). This implies

\[
a = \frac{2.405}{k_c}
\]  

(2.69)

For example, a waveguide designed for 0.5 THz at cutoff would have a radius \( a = 229.5 \) \( \mu \text{m} \).

The purpose for explaining this hollow cylindrical metal waveguide is to illustrate how longitudinal fields can be obtained using a waveguide configuration and how the cutoff frequency arises. In practice, hollow cylindrical waveguides cannot effectively accelerate electrons because the phase velocity necessarily exceeds the speed of light:

\[
v_{ph} = \frac{\omega_0}{k_z} > \frac{\omega_0}{k} = c, \quad \text{for } n = 1
\]  

(2.70)

The phase would roll over the electrons and the net acceleration would be zero. This phase velocity can be engineered to have any value by partially filling the inner wall with a dielectric material, with a cross section shown in Figure 2-21.

The analytical solution to the dielectrically loaded cylindrical waveguide can be found in [122]. To demonstrate the ability to engineer phase velocity, we simply plot here a particular waveguide having a phase velocity approximately equal to half the speed of light at 0.5 THz for the TM\(_{01}\) mode (Figure 2-22).

Finally, we note that linear accelerators (linacs) employed at large facilities such
as SLAC tend to use disk-loaded standing wave accelerating structures, whereas the cylindrical waveguide approach just described is a traveling wave structure. Thus far in our experiments, traveling wave structures have been used for THz acceleration because of the higher bandwidth and ease of fabrication. In the future, standing wave structures would be worth investigating.

### 2.3.4 Electron injector

The electron injector is the first stage of an accelerator chain which generates the electrons and conditions them to match the requirements of the following main accelerator. Electron injectors start with the cathode, where electrons are generated, and end after one or multiple initial acceleration stages.

Space charge forces present the biggest challenge when it comes to injectors, as they play a dominant role in limiting and spoiling the quality of the electron beam. Since space charge forces scale inversely with the square of the beam energy, typically the injector accelerates the electron beam up to semirelativistic energies (100’s of keV to few MeV) where space charge forces become small or negligible.

The electron gun is the first stage of acceleration within the injector. Its accelerating field acts directly upon the cathode where the electrons are born. Because space charge forces are the largest at the gun, the ultimate beam quality in a linac-based accelerator is set by the gun. It is thus of critical importance to increase the
accelerating field in the electron gun, as that shortens the length of time the electrons spend at low space-charge-dominated energies.

**Parallel plate waveguide**

In conventional facilities, the electron gun consists of the cathode inside a pillbox shaped cavity. A standing wave $\text{TM}_{01}$ mode is excited in the cavity and the accelerated electrons escape through a small iris aperture on one end of the cell.

Because high Q factors require a narrow bandwidth, such resonant structures are incompatible with broadband THz pulse drivers. There is thus a need for a new type of gun geometry that employs a high field traveling wave. Such a design was conceived recently [23] consisting of a parallel-plate waveguide instead of pillbox cavity. The $\text{TM}_0$ mode of the parallel-plate waveguide, shown in 2-23, has several advantages over cylindrical structures: (1) it has zero cutoff frequency, (2) zero frequency dispersion, (3) sub-$\lambda/2$ field confinement.

To see this intuitively, recall the boundary condition for electromagnetic fields at a perfect conductor surface:

\[
\hat{n} \times \mathbf{E} = 0, \quad \text{Tangential component of } \mathbf{E} = 0 \tag{2.71}
\]

\[
\hat{n} \times \mathbf{H} = \mathbf{j}_s, \quad \text{Tangential component of } \mathbf{H} = \text{surface current density} \tag{2.72}
\]

\[
\hat{n} \cdot \mathbf{D} = \sigma, \quad \text{Normal component of } \mathbf{D} = \text{surface charge density} \tag{2.73}
\]

\[
\hat{n} \cdot \mathbf{B} = 0, \quad \text{Normal component of } \mathbf{B} = 0 \tag{2.74}
\]

In effect, no tangential component of the E-field can exist on the waveguide wall.
but *any* value of the normal component can exist. Therefore there are no boundary constraints on modes with a polarization orthogonal to the walls, since the surface charge $\sigma$ adjusts to satisfy Eq. 2.73. Likewise, the orthogonal B-field on the wall must be zero, while the tangential B-field can take on any value, since the surface current $j_s$ adjusts to satisfy Eq. 2.72. The field expressions for the TM0 mode is

$$E = \hat{x} \frac{k_z}{\omega \epsilon_0} H_0 e^{-j k_z z}$$  \hspace{1cm} (2.75)\\
$$H = \hat{y} H_0 e^{-j k_z z}.$$  \hspace{1cm} (2.76)

Exciting the PPWG with a $x$-polarized plane or fundamental Gaussian wave is unlikely to excite higher order TM modes because those modes have longitudinal E-field components which are nonexistent in the excitation beam. The general expression for the TM$_m$ mode takes the form

$$E = -\frac{j H_0}{\omega \epsilon_0} \left[ -\hat{z} \frac{m \pi}{d} \sin \left( \frac{m \pi}{d} x \right) + \hat{x} j k_z \cos \left( \frac{m \pi}{d} x \right) \right] e^{-j k_z z}$$  \hspace{1cm} (2.77)\\
$$H = \hat{y} H_0 \cos \left( \frac{m \pi}{d} x \right) e^{-j k_z z}.$$  \hspace{1cm} (2.78)

with

$$k_z = \sqrt{\left( \frac{\omega}{c} \right)^2 - \left( \frac{m \pi}{d} \right)^2}$$  \hspace{1cm} (2.80)

For the TM$_0$ mode, the phase and group velocities simplify to $c$ and there is no cutoff wavelength. In fact, at a frequency of zero, the TM$_0$ mode of the waveguide is simply a parallel plate capacitor. Figure 2-24 shows the dispersion relation plot for the first three modes.
2.3.5 Electron beam characterization

When characterizing an electron beam, typically the properties of interest are charge per bunch, electron energy spectrum, transverse emittance, and electron pulse duration. The charge per bunch and energy spectrum can be directly measured, and with a bit of effort, the pulse duration as well through the use of a streak camera. Emittance takes a bit more effort to measure. The spatial profile of the beam must be measured over several longitudinal distances, or a scanning slit must be placed in the beam [25]. In this section, we will discuss two simple characterization methods for measuring the spectrum and charge of relatively low energy electrons (< 100 keV).

Faraday cup

The simplest method for detecting the electron beam current is to put a piece of metal in the path of the beam and measure the current flowing from that metal to ground. Faraday cups operate in this way, but they are shaped in various ways that resemble a cup to minimize the escape of secondary electrons produced when an incident electron strikes the surface at high velocity. Typically Faraday cups are connected to ground via an ultrasensitive ammeter which measures currents down to the few femtoampere regime. The advantage to using an ammeter is that the measured current gives an absolute measure of the electron beam current. Ammeters such as Keithley 6514 and 6517 can achieve 1 fA sensitivity in electrically quiet conditions (even the movement
Figure 2-25: Illustration of an electron multiplier. The incident electron hits the channel’s inner wall, spurring the generation of secondary electrons, which go on to do the same, until at the output the number of electrons is greatly enhanced.

of a hand can create parasitic currents on the order of pA if the Faraday cup is not well-shielded) after about 30 seconds of averaging.

Another way to measure the beam current amplitude is via a channel electron multiplier (CEM). The advantage of CEMs is that they provide up to $10^8$ gain when the number of incident electrons is low. CEMs also have an aperture shaped to minimize the escape of secondary electrons like the Faraday cup. However, in order to provide gain, incident electrons are sent through a long spiraled channel designed to maximize the number of wall collisions and thus the generation of secondary electrons traveling down the channel. Typically electron-detection CEMs are grounded at the entrance and biased at high positive voltages at the end of the channel to facilitate the generation and flow of secondary electrons. Figure 2-25 illustrates this concept. In a good CEM, the electron flux escaping the back of the channel and hitting the anode is linearly proportional to the incident electron flux. Typical CEM output currents can be directly read by an oscilloscope. CEMs are thus much more sensitive, faster (no averaging required to get sensitivity), and immune to environmental disturbances compared to ammeters (no need for electrical shielding). However, the disadvantage is that they provide only a relative measure of the current.

**Retarding field analyzer**

The retarding field analyzer (RFA) is perhaps the simplest and cheapest device for measuring electron energy spectra. RFAs assume that electrons traveling into the device are highly directional (or linear) along the input axis, and they measure only the electron kinetic energy along that axis. Some RFAs are shaped as a sphere and thus measure the radial kinetic energy of electrons (or other particles) originating from the
center. Figure 2-26(a) shows a conceptual drawing of a linear RFA. Electrons travel into the RFA by crossing through a grounded mesh (Mesh 1) and enter the retarding field region between Mesh 1 and 2. There they experience an opposing voltage barrier equal to $V$. Electrons which have energy higher than $eV$ will cross Mesh 2 and enter the Collector. Those which have energy lower than $eV$ will be turned back. Sweeping $V$ across a range of voltages results in a bias sweep, or cumulative electron distribution function, shown in Figure 2-26(b). Differentiation of this function results in the desired electron spectra, as shown in 2-26(c). Because derivatives are sensitive to high frequency noise, it is important to reduce the shot-to-shot noise of the electron bunch. Often the bias sweep is smoothed before differentiation, even though it decreases the effective spectral resolution of the instrument.

Ideally the electric field in the retarding region is uniform so that there are no trajectory effects (imagine an electron being deflected transversely away by a nonuniform field). For this reason it is important to have flat meshes and perform electrostatic simulations solving Poisson’s equation to ensure that the fields are uniform. High resolution RFAs sometimes utilize electrostatic lenses to image the electron beam from its origination point to the collector [45].
Figure 2-26: (a) Concept drawing of a mesh-based retarding field analyzer (RFA). (b) Raw current detected by the collector as a function of voltage sweep across mesh 2. (c) Derivative of the current with respect to voltage yields the measured electron spectra.
Chapter 3

Ultrafast THz Pulse Generation

We established in the previous chapter the benefits to using THz pulses as a driver for electron acceleration, but the way to obtain substantial amounts of radiation in a little-explored region of the electromagnetic spectrum is a nontrivial task. The generation of THz radiation with meaningful efficiency has historically been a challenging task. In this chapter we discuss methods of generating ultrafast THz pulses with strong electric fields. In particular, we focus on the tilted pulse front pumping (TPFP) method in lithium niobate which, to date, has proven most efficient and scalable to high energies. Note that from there on, we will refer to efficiency as the optical-to-THz conversion efficiency, which is obtained by dividing the measured impinging laser pump energy by the measured output THz energy.

3.1 Summary of techniques: the case for optical rectification

Table 3.1 shows a summary of all the possible techniques to generate ultrashort pulsed THz radiation in the few THz regime. Photoconductive antennas are routinely used to analyze material linear properties (via THz time-domain spectroscopy) but, due to voltage saturation, cannot produce the high energy THz pulses needed to induce material nonlinear effects or accelerate electrons. Free electron lasers and air-plasma
Table 3.1: Intense THz pulse generation schemes in the few THz regime. OR: optical rectification.

four-wave-mixing enables generation of frequencies across the entire THz spectrum due to the absence of solid-state materials, but the efficiency tends to be low. Optical rectification (OR) is a $\chi^{(2)}$ nonlinear process that yields large THz energies. Originally done in ZnTe with efficiencies of $10^{-4}$, the two most promising materials, lithium niobate and organic crystals, have boosted their efficiencies from 0.25% and 2%, respectively, in 2012, to 1% and 3%, respectively, in 2017, thanks to the work from the groups of Hebling [31], Hauri [113], and Kärtner (which includes the work in this thesis).

In addition to efficiency, the generating material must also be scalable to large pump energies. This is a realm where lithium niobate shines over competing organic crystals. Organic crystals are expensive (only one commercial company, Rainbow Photonics, produces them), fragile, difficult to fabricate consistently, and have limited aperture sizes. Researchers try to get around the limited aperture size through the tedious art of tiling separate crystals [113]. On the other hand, lithium niobate crystals are inexpensive thanks to the telecommunications industry which uses lithium niobate for electro-optic modulators. Further lithium niobate crystals have apertures exceeding 25 mm, damage threshold exceeding 100 mJ/cm$^2$, and a bandgap large enough to prevent two- and three-photon absorption in the case of 1030 nm pumping. In Section 3.3.3, we will quantify further reasons why lithium niobate is the best choice of material for high energy THz pulse generation.
CHAPTER 3. ULTRAFAST THZ PULSE GENERATION

3.2 Optical rectification: intrapulse difference frequency generation

Optical rectification is very similar to difference frequency generation (DFG). In DFG, a single photon at a IR frequency $\omega$ mixes with a photon at a lower IR frequency $\omega - \Omega$ to generate a photon at the difference frequency of $\Omega$, which lies in the THz spectral range, and another photon at $\omega - \Omega$ for energy conservation. Figure 3-1(a) depicts an energy level diagram of this process. Assuming the initial infrared power is predominantly at $\omega$, the maximum power efficiency achievable by DFG is $\Omega/\omega$, the case in which 100% of the photons at $\omega$ are converted to $\Omega$ (100% photon efficiency). This value is typically $< 1\%$ for THz frequencies.

Optical rectification can be seen as an intrapulse DFG process, where the DFG occurs between different frequency components of the same broadband pulse. Figure 3-1(b)-(c) illustrates this process in the frequency domain. The high frequency roll-off of the generated spectrum is determined by the bandwidth of the pump IR pulse, and the low frequency roll-off is due to the dipole antenna radiation effect ($E \propto \Omega$). The key difference in optical rectification is that a single IR photon at $\omega$ can be
Figure 3-2: (a) Energy level diagram of cascading. An IR photon at $\omega$ generates a THz photon at $\Omega$ plus a downshifted IR photon at $\omega - \Omega$. The downshifted IR photon can be recycled to generate another THz photon at $\Omega$ plus a further downshifted photon at $\omega - 2\Omega$. This process is repeated as long as the IR and THz frequencies are phase-matched. (b)-(f) Spectral picture of cascading. (b) Two frequency components in the broadband IR spectrum generate a difference frequency that lies in the THz regime. (c) The higher frequency component is downshifted. (d) The downshifted frequency component mixes with an even lower frequency component to generate another THz component. (e) The higher frequency IR component is downshifted once more. (f) In the end, the output IR spectrum is redshifted and broadened.
downconverted more than once (e.g. $\omega \rightarrow \omega - \Omega \rightarrow \omega - 2\Omega \rightarrow \cdots$) to generate multiple THz photons. This repeated downconversion phenomenon is called cascading and is made possible because the frequency difference between the IR photons is so small that phase-matching is satisfied for multiple cascading cycles. Figure 3-2(a) depicts an energy level diagram of cascaded optical rectification, and Figure 3-2(b)-(f) depicts a spectral diagram of the process. A readily observable indication of cascading is an output IR pulse spectrum that is significantly redshifted and broadened (Figure 3-2(f)). Photon efficiencies of $> 100\%$ are routinely observed in highly cascaded THz generation experiments.

### 3.3 Mathematical description of optical rectification

#### 3.3.1 Undepleted pump derivation

Note also that in this geometry the incident infrared and generated THz are polarized in the same direction, so the following equations will be scalar.

For a THz pulse centered at frequency $\Omega$, the one-dimensional frequency domain wave equation in an arbitrary medium can be deduced from Maxwell’s equations as

$$\frac{\partial^2 E(\Omega, z)}{\partial z^2} + k^2(\Omega)E(\Omega, z) = \frac{\Omega^2}{\epsilon_0 c^2} P_{NL}(\Omega, z)$$  \hspace{1cm} (3.1)

where $k(\Omega) = \frac{2\pi}{\lambda} n(\Omega)$ is the wavenumber inside the medium. $P_{NL}$ is the nonlinear polarization given by

$$P_{NL}(\Omega, z) = \epsilon_0 \chi^{(2)}_{eff} \int_0^\infty A(\omega + \Omega)A^*(\omega)e^{i\Delta kz} d\omega$$  \hspace{1cm} (3.2)

where $\chi^{(2)}_{eff}$ is the 2nd order nonlinear coefficient, $A(\omega)$ is the complex amplitude of the IR electric field in the frequency domain, and $\Delta k$ is the phase mismatch. Equation 3.2 basically states that the driving nonlinear polarization is an aggregate of all possible DFG processes between any two components $A(\omega + \Omega)$ and $A(\omega)$ of
The equation governing the evolution of the THz field \( A(\Omega, z) \) can be obtained from the Maxwell’s equations under the slowly varying envelope approximation as

\[
\frac{\partial A(\Omega, z)}{\partial z} = -\frac{1}{2} \alpha(\Omega) A(\Omega, z) - j\mu_0 \frac{\Omega c}{2n(\Omega)} P_{NL}(\Omega, z).
\]  

(3.3)

The first term on the right hand side of Equation 3.3 corresponds to the loss from the linear material absorption coefficient \( \alpha(\Omega) \). The second term corresponds to the gain from the nonlinear polarization. A good starting point for determining the efficiency trends is to solve this equation for the case of an infinite plane wave, perfect phase match, and negligible pump depletion. The efficiency in this case is equal to \[43\]

\[
\eta = \frac{2\Omega^2 d_{eff}^2 L^2 I}{\varepsilon_0 n_g(\omega)^2 n(\Omega)c^3} \exp \left[ \frac{-\alpha(\Omega) L}{2} \right] \frac{\sinh^2[\alpha(\Omega)L/4]}{[\alpha(\Omega)L/4]^2}.
\]  

(3.4)

Here, \( d_{eff} \) is the effective nonlinear coefficient, \( L \) is the length of the nonlinear interaction, \( n_g(\omega) \) is the group index of the IR, and \( I \) is the intensity of the pump beam. One can observe that there is a sweet spot for the interaction length \( L \) that maximizes the efficiency. This occurs near the point where \( \frac{\alpha(\Omega)L}{2} = 1 \). In this case, the efficiency can be written as

\[
\eta \propto \frac{8\Omega^2 d_{eff}^2 I}{\varepsilon_0 n_g(\omega)^2 n(\Omega)c^3 \alpha(\Omega)^2}.
\]  

(3.5)

From this equation, we see that the efficiency of optical rectification is, in general, proportional to the intensity of the pump pulse \( I \), the square of the nonlinear coefficient \( d_{eff} \), the square of the THz frequency \( \Omega \), the inverse square of the THz absorption coefficient \( \alpha(\Omega) \), the inverse square of the IR group index \( n_g(\omega) \), and the inverse of the THz index \( n(\Omega) \). This relation is useful in the determination of a suitable nonlinear material, to be discussed in Section 3.3.3.
3.3.2 Cascaded optical rectification (depleted pump)

The derivation for efficiency in Section 3.3.1 assumed that the pump pulse is unchanged (non-depleted pump approximation). As described in Section 3.2, cascading involves the depletion of higher IR frequency components in the pump pulse and creation of downshifted frequency components. Thus, to include the effects of cascading, the evolution of the pump pulse must be incorporated mathematically.

The evolution of both the THz field $A(\Omega, z)$ and the IR field $A(\omega, z/\cos \gamma)$ can be written as a system of equations, using again the slowly varying envelope approximation. Note that the change of coordinate in the IR field ($z \rightarrow z/\cos \gamma$) is due to the noncollinear geometry. In other words, the IR travels a factor of $1/\cos \gamma$ farther than the THz within an arbitrary period of time.

\[
\frac{\partial A(\Omega, z)}{\partial z} = -\frac{1}{2} \alpha(\Omega) A(\Omega, z) - \frac{j \mu_0 c}{2n(\Omega)} P_{NL}(\Omega, z) \tag{3.6}
\]

\[
\frac{\partial A(\omega, z/\cos \gamma)}{\partial z/\cos \gamma} = -\frac{1}{2} \alpha(\omega) A(\omega, z/\cos \gamma) - \frac{j \mu_0 \omega \cos \gamma}{2n(\omega)} P_{NL}(\omega, z) \tag{3.7}
\]

The nonlinear polarizations are given as

\[
P_{NL}(\Omega, z) = \epsilon_0 \chi^{(2)}_{eff} \int_0^\infty A(\omega + \Omega, z/\cos \gamma) A^*(\omega, z/\cos \gamma) e^{j\Delta k z} d\omega \tag{3.8}
\]

\[
P_{NL}(\omega, z) = \epsilon_0 \chi^{(2)}_{eff} \int_0^\infty A(\omega + \Omega, z/\cos \gamma) A^*(\Omega, z) e^{j\Delta k z} d\Omega \tag{3.9}
\]

One can write a simulation to solve this system of ordinary differential equations to obtain a variety of results. In the remainder of this thesis, simulations will be performed by the 2D network flow code described in the next section or by more ab-initio code (THOR2D) by Ravi et al. [98].
Simplified 2D model

A simple model can be implemented by using a nonorthogonal coordinate system aligned along the directions of propagation for IR and THz, as shown in Figure 3-3. The idea behind the model is that the IR field predominately propagates in the z direction while the THz field predominately propagates in the z’ direction. With each time step, the fields make one move forward in their respective directions. As discussed in the previous paragraph, the IR advances a factor of $1/\cos \gamma$ more than the THz in each time step, and this fact is used to derive the spacing of the grid points as well as the angle between the axes in the nonorthogonal coordinate system. Furthermore, since the THz and IR fields propagate along their respective axes and flow into grid nodes from different directions, the situation begins to resemble a network flow problem, where the flux into the next node is determined by the field amplitudes in the nodes preceding it according to Eqs. 3.6 and 3.7. As illustrated in Figure 3-3, the flow occurs along the arrows annotated ‘THz’ and ‘IR’ and the grid nodes are the intersections between the grid lines. This network flow problem can be formulated numerically by the forward Euler method of integration. At each step along the flow of THz or IR, the fields determined by the following.

\[
A_{i+1,j+1}^{THz} = A_{i,j}^{THz} + \Delta z' \frac{\partial A_{i,j}^{THz}}{\partial z'} \\
A_{i,j+1}^{IR} = A_{i,j}^{IR} + \Delta z \frac{\partial A_{i,j}^{IR}}{\partial z}
\]

This model assumes the following: (1) IR and THz fields propagate along the axes without any divergence and that angular walk-off effects are small. (2) No dispersion and all frequencies are perfectly phase matched. If the interaction length is too short for any substantial divergence or walk-off to occur, Assumption (1) is valid. Assumption (2) is plausible when the fluence is small, since phase mismatch is only becomes a dominant issue when cascading causes extreme spectral broadening.

\[1\text{Idea inspired by Koustuban Ravi [98]}\]
Figure 3-3: Illustration of the nonorthogonal 2D grid used to simulate tilted pulse front THz generation. The evolution of the THz and IR fields represent a network flow problem.

Here we keep the fluences small to compare with our data.
Material & $d_{eff}$ (pm/V) & $\alpha(\Omega)$ (cm$^{-1}$) & $n_g(\omega)$ & $n(\Omega)$ & FOM (pm$^2$cm$^2$V$^{-2}$) \\
\hline
ZnTe & 68.5 & 1.3 & 2.81 & 3.17 & 7.27 \\
CdTe & 81.8 & 4.8 & 2.81 & 3.24 & 11.0 \\
GaAs & 65.6 & 0.5 & 3.56 & 3.59 & 4.21 \\
DAST & 675 & 50 & 2.25 & 2.58 & 41.5 \\
sLiNbO$_3$ 293 K & 168 & 17 & 2.18 & 4.96 & 18.2 \\
sLiNbO$_3$ 100 K & - & 4.8 & - & - & 48.6 \\
\hline

Table 3.2: Properties and figure-of-merit of several candidate materials for THz generation. $n_g(\omega)$ is given at 1.55 $\mu$m and $n(\Omega)$ is given at 1 THz. Data adapted from [43].

### 3.3.3 Choice of nonlinear material: lithium niobate

Having derived a simple, albeit very approximate equation for the efficiency in Eq. 3.5, one can derive a figure-of-merit (FOM) parameter for a given crystal’s ability to generate THz efficiently. Keeping only material parameters in Eq. 3.5, we obtain a FOM equal to

$$FOM = \frac{4d_{eff}^2}{n(\omega)^2n(\Omega)\alpha(\Omega)^2}$$  \hspace{1cm} (3.12)

Table 3.2 gives the properties and FOM for several candidate nonlinear materials for THz generation. The best two materials are DAST$^2$ and stoichiometric lithium niobate (sLiNbO$_3$, or sLN). DAST is a rare and expensive organic material that has limited aperture sizes due to fabrication difficulties, whereas lithium niobate is a common material used in the telecommunications industry and can readily be grown to large apertures. Furthermore, lithium niobate has a large bandgap and high optical damage threshold, reducing two-photon absorption effects and making it scalable to high intensity pumping. Because of these benefits, lithium niobate is our material of choice for THz generation by optical rectification.

$^2$4-N,N-dimethylamino-4’-N’-methyl stilbazolium tosylate, an organic salt nonlinear crystal
3.4 Phase matching via tilted pulse front pumping

A key challenge to producing high nonlinear conversion is phase matching. In lithium niobate, phase matching is an especially critical problem because the pump and THz velocities are severely mismatched. This section discusses the use of a clever phase matching technique called tilted pulse front pumping (TPFP) to solve this problem.

The driving nonlinear polarization is largest when the phase mismatch $\Delta k$ is minimized. We can write $\Delta k$ as

$$\Delta k(\Omega) = k(\Omega) + k(\omega) - k(\omega + \Omega) \approx k(\Omega) - \omega \frac{dk}{d\omega} \bigg|_{\omega_0} = \frac{\Omega}{c} [n(\Omega) - n_g(\omega_0)]$$ (3.13)

Here, the approximation is made under the assumption that $\Omega \ll \omega_0$, where $\omega_0$ is the center frequency of the IR pulse. Equation 3.13 states that the phase mismatch is proportional to the difference between the phase index of THz and group index of IR. In lithium niobate however, the difference between these two parameters is large, causing prohibitively short coherence length. For example, for 0.5 THz radiation generated by a pump pulse centered at 1030 nm, $n(\Omega) = 4.95$ and $n_g(\omega_0) = 2.25$, leading to a coherence length of $L_{coh} = \pi/\Delta k = 0.7$ mm.

To minimize the phase mismatch, a noncollinear phase-matching geometry must be employed. One remarkably effective noncollinear geometry is the tilted pulse front pumping (TPFP) geometry, shown in Figure 3-4(a). In TPFP, an incident IR beam propagating in the $z'$-direction has a pulse front that is tilted with respect to its propagation direction by an angle of $\gamma$, given by

$$\cos \gamma = \frac{n_g(\omega_0)}{n(\Omega)}$$ (3.14)

The generated THz propagates in the $z$-direction, normal to the tilted pulse front.
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Figure 3-4: (a) Tilted pulse front pumping scheme. (b) Angular dispersion picture of phase-matching.

The coordinate transform between the \( z' \)- and \( z \)-direction is

\[
z' = z / \cos \gamma
\]  

(3.15)

A physical explanation of this is that the IR pulse propagation distance is \( 1 / \cos \gamma \) larger than the THz propagation distance. With this modification, the phase mismatch becomes

\[
\Delta k \approx \frac{\Omega}{c} \left[ n(\Omega) - \frac{n_g(\omega_0)}{\cos \gamma} \right] = 0
\]  

(3.16)

or in other words, the process is phase matched. A physical explanation of this scheme is that the velocity of the pump \textit{projected in the direction of its pulse front} is matched to the velocity of the THz wave.

### 3.4.1 Angular dispersion picture of TPFP

A tilted pulse front is created by inducing angular dispersion into the IR beam. Hebling [42] showed that the pulse front tilt angle \( \gamma \) induced by an angular dispersion \( \frac{d \theta}{d \lambda} \) is given by

\[
\tan \gamma = \frac{n(\omega)}{n_g(\omega) \frac{d \theta}{d \lambda}}
\]  

(3.17)

Equation 3.17 infers that a pulse front tilt is synonymous with angular dispersion, which begs for an alternative picture of the TPFP phase-matching scheme based
on angular dispersion. Figure 3-4(b) shows the wavevectors \( \mathbf{k}(\omega_0) \), \( \mathbf{k}(\omega_0 + \Omega) \) of two incident IR frequency components and the wavevector \( \mathbf{k}(\Omega) \) of the generated THz. The two IR wavevectors are angularly separated by an angle of \( \frac{d\Omega}{d\omega} \) (\( \frac{d\Omega}{d\omega} \) is the angular dispersion), and the THz wavevector \( \mathbf{k}(\Omega) \) is angularly separated from the IR wavevector \( \omega_0 \) by the pulse front tilt \( \gamma \). It can be intuitively seen by vector addition that \( \Delta \mathbf{k}(\Omega) = \mathbf{k}(\Omega) + \mathbf{k}(\omega_0) - \mathbf{k}(\omega_0 + \Omega) \) is zero. Hence, the angular dispersion picture of phase-matching is consistent with the tilted pulse front picture given in Section 3.4.

### 3.4.2 Phase-matching is limited by dispersive effects

There are two factors that limit the interaction length of the phase-matching: material dispersion and angular dispersion. Both have the effect of causing the IR pulse to accumulate chirp as it propagates. For THz generation in lithium niobate, the chirp caused by angular dispersion is the predominant limitation of interaction length, because the angular dispersion must be significantly large in order to meet the phase matching condition.

We recall the expression for the nonlinear polarization from Equation 3.2:

\[
P_{NL}(\Omega, z) = \varepsilon_0 \chi^{(2)}_{eff} \int_0^\infty A(\omega + \Omega) A^*(\omega) e^{j(\Delta k z)} d\omega
\]

In the case of perfect phase match \( \Delta k = 0 \), the integrand in Equation 3.2 can be rewritten as

\[
|A(\omega + \Omega)||A^*(\omega)| e^{j[k(\omega+\Omega)-k(\omega)]} z = |A(\omega + \Omega)||A^*(\omega)| e^{j\left[\frac{d\mathbf{k}}{d\omega} |\omega + \Omega| - \frac{d\mathbf{k}}{d\omega} |\omega| \right]} z
\]

\[
= |A(\omega + \Omega)||A^*(\omega)| e^{j\left[\frac{d\mathbf{k}}{d\omega} |\omega + \Omega|^2 + \frac{\partial^2}{\partial \omega^2} |\omega|^3 + \cdots \right]} z
\]

\[
= |A(\omega + \Omega)||A^*(\omega)| e^{j\Delta \mathbf{k}_c(\omega, \Omega) z} \tag{3.18}
\]

where \( \Delta \mathbf{k}_c(\omega, \Omega) \) is the phase mismatch between the two IR components \( \omega + \Omega \) and \( \omega \) due to chirp.
The red arrows denote the integrand of Equation 3.2 and the blue arrow denotes the total integral of Equation 3.2, or the sum of the red arrows, in the case of (a) zero chirp (perfect phase match), (b) moderate chirp, (c) severe chirp (perfect phase mismatch).

\[ \Delta k_c(\omega, \Omega) = \frac{d^2 k}{d\omega^2} \bigg|_{\omega} \Omega^2 + \frac{d^3 k}{d\omega^3} \bigg|_{\omega} \Omega^3 + \cdots \quad (3.19) \]

A physical picture of the effect of dispersion on the THz generation efficiency can be seen in the phasor diagram of Figure 3-5. The red phasors represent the normalized integrand (Equation 3.18) at various frequencies \( \omega \) within the integral, and the blue phasor represents the sum of the red phasors, or the nonlinear polarization (Equation 3.2). Figure 3-5(a) shows the phasor diagram without any dispersive effects. The red phasors are lined up leading to a long blue phasor (large nonlinear polarization). On the contrary, Figure 3-5(b) shows the phasor diagram in the event of dispersive effects. The red phasors are misaligned because there is a frequency dependent phase \( \Delta k_c(\omega, \Omega) \). As a result, the blue phasor is shortened and the nonlinear polarization is reduced. As the propagation distance \( z \) increases, the frequency dependent phase mismatch is accentuated, causing the angle between the red arrows to increase. At some length \( L_D \), the blue arrow will go to zero as shown in Figure 3-5(c). For pulses dominated by second order dispersion, \( L_D \propto \frac{\tau}{\frac{d^2 k}{d\omega^2} \big|_{\omega_0}}, \) where \( \tau \) is the transform-limited pump pulse duration. This is the physical picture of the limitation in coherence length caused by dispersive effects.
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Figure 3-6: Schematic of pulse front tilting imaging setup. PFT, pulse front tilt; \( \theta_i \), incidence angle; \( \theta_d \), diffraction angle; \( f \), focal length; \( n \), refractive index; \( n_g \), group index; \( \lambda_1, \lambda_2, \lambda_3 \) illustrate angular dispersion induced by grating.

3.5 System implementation of TPFP

In the previous section, we gave a theoretical framework of optical rectification and the TPFP method. In this chapter, we will utilize this knowledge to explore several techniques for efficiency optimization. These will include (1) optimization of the pulse front tilting setup, (2) optimization of the pump pulse parameters, (3) cryogenic cooling of the lithium niobate crystal, and (4) implementation of a THz antireflection coating for the lithium-niobate-to-air interface.

3.5.1 Pulse front tilt is induced by diffraction from grating

To experimentally realize a pulse front tilt with angle \( \gamma \), the IR beam must have angular dispersion. In [42], Hebling derived the relation between pulse front tilt \( \gamma \) and angular dispersion \( \frac{d\theta}{d\lambda} \) to be

\[
\tan \gamma = \frac{n}{n_g} \frac{\lambda}{\lambda} \frac{d\theta}{d\lambda} \tag{3.20}
\]

An angular dispersion \( \frac{d\theta_d}{d\lambda} \) in the pump beam can be realized by \( m \)th-order diffraction from a grating with groove density \( g \). Using the grating equation \( \sin \theta_i + \sin \theta_d = mg\lambda \), we derive the angular dispersion to be

\[
\frac{d\theta_d}{d\lambda} = \frac{mg}{\cos \theta_d} \tag{3.21}
\]
Putting Equations 3.20 and 3.21 together, we arrive at the expression for the pulse front tilt in from a grating

$$\tan \gamma = \frac{n \lambda g}{n_g \cos \theta_d}.$$ \hspace{1cm} (3.22)

### 3.5.2 Relay-imaging of grating

The diffracted beam experiences undesired chirp and spatial walkoff as it moves away from the grating. Even if the THz generation crystal were placed directly behind the grating at some small distance, the beam would have already experienced significant group velocity mismatch, leading to poor conversion efficiency. Ideally, the crystal would be superimposed at the location of the impinged beam on the grating; however, this is physically impossible. One solution is to image relay the beam diffracted from the grating onto the crystal, placed some distance away, such that the beam on the crystal is an image of the diffracted beam from the grating. A single-lens image relay scheme is shown in Figure 3-6.

Proper imaging of the grating requires that the tilted pulse front coincide with the image of the grating, or in other words, that the angle of the grating image $\theta$ must match the angle of the pulse front tilt $\gamma$. If these two angles are mismatched, only the center (the point intersecting the optical axis) of the pulse front will be properly imaged and generate THz efficiently. Points in the pulse front away from the center will be offset from the image plane and become ‘blurry’. From the phase matching point of view, points that are not on the image plane will have group delay mismatch leading to poor conversion efficiency. A more detailed analysis of these effects can be found in [87].

To derive the angle of the grating image, we first observe in Figure 3-6 that the angle of the grating object is $\theta_d$. The grating image has the shape of the original grating scaled transversely by magnification factor $M_t$ (from the imaging setup) and by index $n$ (from refraction into the crystal). Consequently, the angle of the grating image $\theta$ inside the crystal is
\[ \tan \theta = nM_t \tan \theta_d. \] (3.23)

To derive the angle of the pulse front inside the crystal, we begin with the expression for pulse front tilt angle before image relay given by Equation 3.20. A smaller transverse magnification factor causes a larger angular dispersion, and refraction into a crystal reduces the angular dispersion. Therefore, Equation 3.20 should be inversely scaled by \( M_t \) and \( n \). The expression for pulse front tilt inside the crystal becomes

\[ \tan \gamma = \frac{\lambda g}{nM_t \cos \theta_d} \] (3.24)

Note that we have now set the diffraction order \( m \) to 1 since a majority of commercial gratings are designed for this order.

### 3.5.3 Design of a TPFP setup

We are now presented with a design problem. In order to image properly, we must match \( \theta \) and \( \gamma \). Furthermore, in order to achieve phase matching, we must set \( \gamma = \cos^{-1} \left( \frac{n(\omega_0)}{n(\Omega)} \right) = \cos^{-1} \frac{\lambda}{2g} \approx 63^\circ \), as discussed in Section 3.4.

\[ \gamma = \theta = 63^\circ. \] (3.25)

A final constraint is that the diffraction angle \( \theta_d \) should be close to the Littrow angle \( \theta_{\text{lit}} = \sin^{-1} \frac{\lambda}{2} \) in order to maintain high diffraction efficiency from the grating:

\[ \theta_d \approx \theta_{\text{lit}}. \] (3.26)

To achieve these objectives, we have control over three free variables: grating groove density \( g \), transverse magnification \( M_t \), and diffraction angle \( \theta_d \). Table 3.3 summarizes the free parameters and constraints. One approach to this design problem is to first decide on a groove density \( g \) and then back-calculate \( M_t \) and \( \theta_d \). Manipulating Equations 3.24 and 3.23 and setting \( \tan \gamma = \tan \theta \) gives
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<table>
<thead>
<tr>
<th>Free parameters</th>
<th>Constraints</th>
</tr>
</thead>
<tbody>
<tr>
<td>$g$, groove density</td>
<td>$\gamma = \theta = 63^\circ$</td>
</tr>
<tr>
<td>$M_t$, transverse magnification</td>
<td>$\theta_d$ should be close to littrow angle $\sin \theta_{litt} = \frac{g\lambda}{2}$</td>
</tr>
<tr>
<td>$\theta_d$, diffraction angle</td>
<td></td>
</tr>
</tbody>
</table>

Table 3.3: Free parameters and constraints for design of the tilted pulse front pumping setup.

Figure 3-7: Plot of required incidence angle as a function of groove density for optimal phase matching. A groove density of 1500 l/mm is chosen because it has a required incidence angle close to the Littrow angle.

\[
g = \frac{n_g \cos \theta_d \tan^2 \gamma}{n\lambda \tan \theta_d}.
\]

(3.27)

Since incidence angle is experimentally easier to measure, we can apply $\sin \theta_i = g\lambda - \sin \theta_d$ to convert from diffraction angle to incidence angle. Figure 3-7 shows a plot of the required incidence angle as a function of groove density based on Equation 3.27. It is seen that a groove density of 1500 l/mm is near optimal because its required incidence angle closely matches the Littrow angle. Having fixed a groove density $g$, the incidence angle and transverse magnification can be back-calculated from Equations 3.24 and 3.23 to be $46^\circ$ and 0.6218, respectively.

Finally, a transverse magnification $M_t$ is realized physically by setting the object and image distances, $s_1$ and $s_2$, such that

\[
M_t = -\frac{s_2}{s_1}.
\]

(3.28)
where \( s_2 \) and \( s_1 \) are subject to the lensmaker equation \( \frac{1}{s_1} + \frac{1}{s_2} = \frac{1}{f} \). It is beneficial to use long focal lengths \( (f > 200 \text{ mm}) \) in order to reduce imaging aberrations.

### 3.5.4 How sensitive is the imaging condition?

We now take a moment to consider the sensitivity of the imaging condition given in Eq. 3.23, which states that the angle of the grating image must match the required pulse front tilt angle of 63°. This condition was first stated in [29] and corroborated in [48]. We analyze this condition through the simulation code THOR2D (THz Optical Rectification 2D) written by K. Ravi [98] which models the full experimental system including the imaging setup. The code has been verified by experiment within our range of interest (see next two paragraphs). The example system that we study via simulations and experiment in the remainder of this section is depicted in Figure 3-8.

By varying the angle of grating image and recording the THz generation efficiency we can observe the sensitivity grating image angle condition. Eq. 3.23 shows that we can vary the grating image angle by changing either the incidence/diffraction angle off the grating or the magnification of the beam. We scan both of these parameters and record the resultant efficiency in Figure 3-9. The locus of highly efficient operating points centers around the line corresponding to a fixed pulse front tilt \( (\theta_{PFT} = 63^\circ) \) and not significantly overlapped with the line corresponding to a fixed grating image tilt \( (\theta_{\text{grating image}} = 63^\circ) \).

The THOR2D code has been verified by experiments as shown in the white circles in Figure 3-9. The efficiency corresponding to those points are plotted in Figure 3-10. The experiments were performed using a zoom lens telescope and congruent lithium niobate at room temperature. All dimensions and parameters entered into the code were closely matched to the experimental values. The \( 1/e^2 \) spot size used in these experiments and simulations was 3 mm.

Going a little further, we plot in Figure 3-11 the efficiency as a function of diffraction angle over a large range, with the magnification optimized at each point to achieve maximum efficiency. This is essentially a plot of the efficiency along the solid line in Figure 3-9 and it shows that the efficiency is slowly-varying with respect to the
Figure 3-8: (a) Schematic of the setup used in our simulations and in the experiment used to verify those simulations. The imaging is done by a zoom lens. (b) Parameter values for the setup.
Figure 3-9: Efficiency as a function of magnification and diffraction angle. Simulated using the THOR2D code. Experimental data points were performed using a zoom lens telescope and congruent lithium niobate at room temperature.

Figure 3-10: Experimentally-obtained efficiency as a function of diffraction angle, with the magnification optimized at each point.
diffraction angle so long as the pulse front tilt angle is fixed at the optimum value.

This result—that the optimal efficiency depends only on an optimal pulse front tilt angle ($\theta_{PFT} = 63^\circ$) and not on the imaging condition—is surprising given that imaging conditions typically have a strong effect on phase-matching dynamics. One possible explanation is that the spot size of 3 mm is not large enough to be in a range where the imaging condition has a strong effect. As spot sizes get larger, the potential for efficiency degradation due to imaging errors should increase [87]. Nonetheless we learn from these results that for modest spot sizes $\sim 3 \text{mm}$, there is no need to fulfill the imaging condition given in Eq. 3.23. With one condition removed, the design of the TPFP setup becomes less constrained. Practically, this means that one can experimentally optimize the efficiency with only one parameter rather than two coupled parameters. The choice of whether to adjust the grating angle or magnification is up to the user, but adjusting only one of them is necessary.

Figure 3-11: Efficiency as a function of diffraction angle given that the pulse front tilt angle is fixed at the optimum value of $63^\circ$ (via adjusting magnification).
3.6 Tricks to improve efficiency

3.6.1 Optimization of pump pulse duration

The pump pulse that impinges onto the crystal can be characterized spatially and temporally. A full investigation of the spatial properties optimal for THz generation is out of the scope of this thesis. However, because the theoretical analysis in Section 3.3.1 assumed plane waves, spatial properties are of secondary importance so long as the pump pulse is of large enough spatial extent and good enough beam quality that the plane wave approximation can be applied.

The dependence of the THz generation efficiency as a function of temporal properties is of greater interest. In Section 3.2, optical rectification was treated as an intrapulse DFG process in the frequency domain. Thus, the optimization of the pump pulse should be treated in the frequency domain as well. The pump pulse can be characterized by two parameters: bandwidth and chirp. We know intuitively from phasor diagrams in Figure 3-5 that the optimal chirp condition is to have no chirp at all, or in other words, to have a transform-limited (TL) pulse.

We proceed to determination of the optimal pump bandwidth. Having fixed the chirp condition to transform-limited pulses, we can directly correlate the pump bandwidth to its time-domain pulse duration (assuming a Gaussian profile). Figure 3-12 shows a simulation of the THz generation efficiency as a function of transform-limited pump pulse duration at full-width-half-maximum (FWHM). It is seen that there exists a maximum at around 500 fs. The reason for having a sweet spot in the pump pulse duration (assumed to be transform-limited from here on) can be seen by looking at the two extremes. When the pulse duration is short, its bandwidth is large, causing dispersive effects to dominate. This in turn reduces the interaction length of the nonlinear process which ultimately limits the conversion efficiency. On the other hand, when the pulse duration is long, its intensity is low, reducing the nonlinear yield (see Equation 3.4). It turns out that a transform-limited pump pulse with duration around 0.5 ps balances the trade-off between these two effects. Therefore, the optimal pump is a transform-limited pulse with pulse duration of 0.5 ps.
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Figure 3-12: Simulation of the THz generation efficiency as a function of pump pulse duration and temperature. The plot shows that the optimal efficiency is achieved at a pump pulse duration of around 0.5 ps and at cryogenic temperatures. Annotations show the results achieved by us (MIT) and by Fulop [30]. sLN, stoichiometric lithium niobate; cLN, congruent lithium niobate. Simulation adapted from [30].
3.6.2 Cryogenic cooling of lithium niobate crystal

A major obstacle of THz generation in lithium niobate is the large absorption coefficient of room-temperature lithium niobate. The absorption can be dramatically reduced at cryogenic temperatures. Figure 3-13 shows that the absorption coefficient at 1 THz in lithium niobate drops dramatically from $\sim 20 \text{ cm}^{-1}$ at room temperature to $\sim 4 \text{ cm}^{-1}$ at cryogenic temperatures. Based on the figure of merit described in 3.12 and the simulation in Figure 3-12, the THz generation efficiency can be increased by a factor of 3 through cooling to 100 K.

It is important to re-optimize the experimental setup after cryogenically cooling the lithium niobate. When the crystal temperature is changed, its refractive index changes as a well, leading to a suboptimal phase-matching of the optical beam’s TPFP configuration. More specifically, both $n_g(\omega_0)$ and $n(\Omega)$ are changed by temperature, so the required pulse front tilt angle (Eq. 3.14), actual pulse front tilt angle (Eq. 3.24), and grating image angle (Eq. 3.23) will experience discrepancy from one another. Seen more clearly through THOR2D simulations, Figure 3-14 shows that the most efficient configuration parameters (magnification and incidence angle) shifts when the temperature is lowered from room to cryogenic temperatures. The simulation results overlap well with the theoretical curves confirming that the shift predictably results
We now turn our attention to the efficiency enhancement afforded by cryogenic cooling through experiment and simulation of the system depicted in Figure 3-8. Since the optimal efficiencies at room temperature (300 K) and cryogenic temperature (77 K) require different setup configurations, we plot the efficiency as a function of temperature for a setup optimally configured for 300 K in Figures 3-15(a)-(b) and for 77 K in Figures 3-15(c)-(d). There is excellent agreement between simulation and experiment in these curves. These curves show the temperature dependent efficiency for a fixed configuration, and they illustrate the importance of reconfiguring the setup whenever the crystal temperature is changed. For example, for a crystal optimized at room temperature, there may seem at first to be very little enhancement (≈25%) in the efficiency until the setup is realigned. On the other hand, if a setup optimized at cryogenic temperature suddenly runs out of coolant, the warming of the crystal can lead to a seven-fold decrease in the efficiency. By looking at the highest efficiency at each temperature’s optimal configuration, we see experimentally that there is a factor

![Efficiency versus two experimental configuration parameters](image)

Figure 3-14: Efficiency versus two experimental configuration parameters (magnification and incidence angle) for room and cryogenic temperatures. Note the shift in the locus of points of highest efficiency. Simulations performed using THOR2D with undepleted pump. See Figure 3-10 for the parameters used in this simulation. For definition of “Fixed PFT” and “Fixed Image tilt” in the legend, see Section 3.5.4 as well.

from temperature-induced changes in the refractive indices.
Figure 3-15: Efficiency as a function of temperature for a setup optimized at (a)-(b) room temperature and (c)-(d) cryogenic temperature. Simulation and experiment are in agreement.

of about 2.30 enhancement (2.38 in simulations) in going from room to cryogenic temperature.

3.6.3 THz output coupling

The large THz index mismatch between lithium niobate and air causes a 44% loss of efficiency in the output coupling due to Fresnel reflection. This problem can be solved by using an antireflection (AR) coating optimized for THz or by using a silicon-kapton structure.

THz antireflection coating

While AR coatings for optical wavelengths have well-established materials and fabrication processes, AR coatings for THz wavelengths are still in their infant stages. In this section, we present our investigation of several candidate THz AR coatings for lithium niobate.
Table 3.4: Parameters for design of AR coating optimized for 0.5 THz based on several candidate materials, along with computed and experimental results. $n$, refractive index at 0.5 THz; $\alpha$, absorption coefficient at 0.5 THz; $t$, optimal thickness; $R$ computed reflection at 0.5 THz; $\frac{T_{\text{computed}}}{T_0}$, computed transmission divided by transmission without coating (efficiency enhancement); $\frac{T_{\text{experimental}}}{T_0}$, experimental transmission divided by transmission without coating (efficiency enhancement).

The optimal thickness $L_{AR}$ for an AR coating is

$$L_{AR} = \frac{\lambda}{4n_{AR}} \quad (3.29)$$

where $n_{AR}$ is the index of the coating material. Under this condition the normal incidence reflection coefficient is (from [44])

$$R = \left( \frac{n_0n_1 - n_{AR}^2}{n_0n_1 + n_{AR}^2} \right)^2 \quad (3.30)$$

Equation 3.30 goes to zero when

$$n_{AR} = \sqrt{n_0n_1} \quad (3.31)$$

For the lithium-niobate-to-air interface (where $n_1 = 4.96$ and $n_0 = 1$), the optimal AR coating index is $\sqrt{4.96} = 2.23$. Table 3.4 shows the parameters and for several candidate THz AR coating materials. The efficiency enhancement was computed and experimentally tested for each material with our broadband THz source (Section 3.7.2). Z-cut crystal quartz showed the best performance with an efficiency enhancement of 1.5, compared to a computed enhancement of 1.78. The discrepancy is mainly due to the broad bandwidth of the THz covering regions away from the design frequency. Since this AR coating is single layer, the reflection increases quickly
at frequencies away from the design frequency. This is shown in the power reflection spectra in Figure 3-16(a). Our measurement of 1.5 enhancement was corroborated measurements from another group [11].

There are two final considerations for these single-layer THz AR coatings. First note that the single-layer AR spectra in Figure 3-16(a) are rather narrow and would shrink the bandwidth of single-cycle, broadband THz pulses. This in turn has the undesirable effect of reducing the peak THz field, limiting applications in nonlinear spectroscopy and charged particle acceleration.

The second consideration is that of optical damage to the coating. Since surfaces have a lower threshold for optical damage (see Section 2.1.2) and since polymers such as Kapton and Parylene have lower damage threshold, it is likely that the effectiveness
of a polymer-based THz AR coating will be stymied by damage occurring when the AR coating makes contact with the pump beam. In the next section, we discuss a possible alternative to the THz AR coating which may be more broadband and work at high optical fluences.

**Silicon-kapton output coupler**

It is also possible to out-couple the THz in a broadband way without fabricating difficult, damage-resistant AR coatings by using a silicon-kapton structure. The silicon-kapton structure, depicted in Figure 3-17 is simply a thick high resistivity float zone (HRFZ) silicon window with the output facet of the lithium niobate crystal pressed against one side of the silicon and with thin kapton taped to the other side. The THz index of silicon is 3.4, making for more adiabatic transmission of THz radiation from the lithium niobate (index of 4.9). The Fresnel reflection from lithium niobate to silicon is

$$ R_{\text{fresnel}} = \left( \frac{n_{\text{LN}} - n_{\text{Si}}}{n_{\text{LN}} + n_{\text{Si}}} \right)^2 \approx 3\% $$  \hspace{1cm} (3.32)

compared to 44% for lithium niobate to air. Propagation of THz through a thick undoped silicon window is lossless ($\alpha < 0.04 \text{ cm}^{-1}$) and dispersionless [35] thanks to the lack of free carriers. For the same reason, the damage threshold of silicon is high,
enabling it to withstand higher optical fluences than polymer-based AR coatings. The transmission from silicon to air is facilitated by a kapton AR coating. According to Eq. 3.31, the optimal AR coating index for the silicon-to-air interface in the 0.1-2.0 THz range is 1.84, close to the kapton index of 1.86 [16]. For a frequency of 0.5 THz ($\lambda = 600 \mu m$), the optimal AR coating thickness is 81 $\mu m$, close to the thickness of 75 $\mu m$ kapton tape (kapton tape is typically sold in thickness denominations of 25 $\mu m$, 50 $\mu m$, 75 $\mu m$, 100 $\mu m$, and so on).

The silicon-kapton structure has a slightly larger bandwidth than single-layer AR coatings, as shown in Figure 3-16(a). When a candidate single-cycle THz pulse is passed through the structure as shown in Figure 3-16(b), it has a transmission of 90%, in contrast to 88% for the ideal single-layer AR coating.
3.7 Experiment: High efficiency THz generation pumped at 1030 nm

In this section, we discuss in detail the experiment leading to THz generation with a record 1% conversion efficiency, or about a factor of 4 higher than the second highest reported efficiency of 0.22% [30] as of the time of the experiment.

3.7.1 Experimental setup

The experimental setup consists of a sub-ps pump source and a THz generation component, as depicted as Figure 1. The pump source is a Yb:KYW chirped pulse regenerative amplifier (RGA) producing 2 mJ pulses with 1 kHz repetition rate at a center wavelength of 1030 nm and bandwidth of 2.6 nm. The dielectric grating compressor following the RGA compresses the pulses to 680 fs, which is \( \sim 15\% \) longer than the transform-limited pulse duration assuming a Gaussian profile. The seed for the RGA was a mode-locked Yb-doped fiber oscillator emitting 70 fs, 0.2 nJ pulses at 80 MHz [50] amplified to 1.6 nJ by a Yb-doped fiber amplifier. After losses through the optical elements in the setup, the impinging pump energy into the LN crystal was 1.2 mJ.

The TPFP scheme was achieved using a grating and a lens for image relay, as shown in Figure 3-18. The laser beam is incident at 46 degrees to the normal of a 1500 line/mm gold grating and is then imaged onto the LN crystal by a single 230 mm bestform lens (L1). The grating-to-lens distance was 584 mm and the lens-to-crystal distance was 379 mm, implying a demagnification of 1.54. A half-wave plate was used to rotate the polarization of the pump to be parallel to the optic axis of the crystal. A cylindrical lens was used to reshape the \( 1/e^2 \) pump diameter to 3.0 mm in the horizontal and 3.0 mm in the vertical directions, which corresponds to a pump fluence of 17 mJ/cm\(^2\).

The congruent lithium niobate (cLN) crystal is doped with MgO at 6% in to reduce photorefractive losses [68]. The crystal was z-cut and shaped into an isosceles
prism with a vertex angle of 56 degrees and base angles of 62 degrees to make THz propagation normal to its output face. This geometry is also necessary to circumvent the small THz critical angle of $\sim 11$ degrees due to large THz refractive index in LN. We also applied a 75 $\mu$m kapton tape THz antireflective coating which improved the collected energy by about 20-30%. The infrared (IR) beam experiences total internal reflection at the THz output face and is transmitted through the adjacent surface. The entry and exit surfaces for the IR beam are anti-reflection (AR) coated for 1030 nm.

The predominant motivation for cryogenic cooling is to result in high conversion efficiencies which can then be exploited to scale to large THz energies by scaling the pump energy. Since it is relatively difficult to grow stoichiometric LN (sLN) to large dimensions beyond 1 cm, we opted to use cryo-cooled cLN despite its relatively higher absorption [89] and lower effective second order nonlinear coefficient $d_{eff}$ [28]. The crystal was indium soldered to a nickel heat sink whose thermal expansion is
well-matched to that of LN, the heat sink was screw-mounted to a commercial liquid nitrogen dewar, and a silicon diode temperature monitor was adhered to the bottom face of the crystal by thermally-conductive glue.

The THz power was measured by focusing the output onto a pyroelectric detector (Microtech Instruments) by a single off-axis parabolic mirror with an effective focal length of 25.4 mm. In order to match to the voltage relaxation time of the pyroelectric crystal, the repetition rate of the laser was reduced to 10 Hz. The THz pulse energy was then calculated from the voltage modulation using the factory-calibrated responsivity of 3.4 ± 0.4 V/mW.

The THz waveform was measured by means of a conventional electro-optic (EO) sampling technique [118], shown in Figure 3-19. An 80 MHz, 70 fs IR pulse train from the mode-locked fiber oscillator, which is the seed laser for the RGA (ensuring optical synchronization), were spatio-temporally overlapped with the THz pulses and focused onto a 200 µm thick, 110-cut ZnTe crystal. The IR pulses sample the THz field-induced birefringence as a function of delay, which is swept at a constant rate (1.2 mm/sec) using a calibrated motorized translation stage. A quarter-wave plate followed by a polarizer converts the field-induced birefringence to an intensity modulation, and the intensity modulation is recorded by a fast photodiode. Because of the much higher repetition rate of the IR pulse train, a boxcar integrator (Stanford Research SR250) is used to electronically gate out all other pulses except the one whose intensity is modulated by the THz. The integrated signal is then recorded by an oscilloscope.

Spatial characterization of the THz beam was performed using a microbolometer imaging camera (NEC IRVT0831) at 30 Hz with continuous averaging of 8 frames. The camera had 23.5 µm pixels and a spectral response that rolls off below 1 THz [84]. Thus the beam characteristic measured is heavily weighted towards the high frequency components.
3.7.2 Results and discussion

Figure 3-20 depicts the THz energy as a function of the pump energy. The THz energy increases with a power dependence of 1.56 without much sign of roll-off from free-carrier absorption. The maximum THz energy achieved with the cryo-cLN crystal was 12.0 µJ at 1.2 mJ of pump energy, corresponding to an efficiency of 1%. To further verify the high efficiency conversion, the spectrum of the IR beam after THz generation was measured as shown in Figure 3-21. The large red shift and broadening of the IR spectrum is a consequence of the repeated frequency down-conversion of the IR pulse leading to repeated THz generation. This ‘cascading’ has been suggested as the reason for large conversion efficiencies [114, 51, 52]. A calculation of the center-of-mass optical frequency shift of the IR spectrum at full efficiency divided by the THz center frequency (0.45 THz) reveals roughly 5.5 cascading cycles, or 550% photon conversion efficiency. To verify that the broadening and the red shift of the output IR spectrum can indeed be explained by the concept of cascading, one can use an effective 1-D system of coupled equations similar to [40, 56] which predict an extent of broadening qualitatively consistent with experiments for similar parameters.
Figure 3-20: Pump energy sweep of THz generated from cryogenic temperature cLN. The power dependence was 1.56.
Figure 3-21: Power spectrum of IR beam after THz generation in cLN for various conversion efficiencies.
Temporal characterization of the THz pulse generated at room temperature was performed by EO sampling. Figure 3-22(a) depicts the measured single-cycle THz waveform with a cycle period of \(\sim 2.2\) ps. The theoretical calculation of the temporal electric field waveform is overlaid with the experimental measurement in Figure 3-22(a) and resembles the basic feature of the experimental result. The temporal waveform is sensitive to the tilt angle of the pulse front, the input amplitude, as well as phase spectrum of the IR pulse which can explain the discrepancy between theory and experiment. The corresponding experimental and calculated spectra are presented in Figure 3-22(b). It is seen that the THz pulse has a center frequency of 0.45 THz and a full-width-half-maximum (FWHM) bandwidth of 0.4 THz with a tail extending beyond 1 THz, showing good match with the simulations based on the model in [29]. The echo pulse at \(\sim 3.5\) ps is a common artifact caused by multiple reflections of the IR probe pulses off the EO crystal and can be disregarded. Due to the echo pulse, a time window was applied prior to the Fourier transform; therefore, the typical absorption lines of water are not observed because of the limited frequency resolution. The temporal shape of the THz pulses from cryogenic LN is expected to be similar to that at room temperature except for a slightly shifted spectrum towards high frequency due to reduced losses.

Figure 3-23 depicts the beam at the focus of an off-axis parabola with 50 mm of effective focal length. The actual focused beam diameter is believed to be wider by
a factor of 2 or 3 considering the high-pass spectral response of the camera with a
cutoff at 1 THz and our pulse center frequency of 0.45 THz. Nevertheless, the focused
beam diameter of 1 mm shows a near diffraction-limited beam quality at the $\sim 1$ THz
portion of our pulse.

Discrepancy with published results

Our initial publications [51, 52] reported an efficiency of 1.15% at room temperature
and implied that 3.7% efficiency is achieved because of the 3.2 enhancement factor
observed when cryogenic cooling was applied. Since those publications, we have
consistently observed and reported a max of 1% efficiency at cryogenic temperature.
The reason for this discrepancy will be clarified here.

When trying to detect the THz signal for the first time, we had used a voltage
preamplifier to boost the signal from the pyroelectric detector. The amplification
factor from this preamplifier was mistakenly not taken into account when retrieving
the THz energy from the final measured voltage. Based on the verified efficiency
measurements we now have, it is plausible that the preamplifier had been set to a
gain factor of 5, which would translate to an efficiency of 0.23% (cLN) and 0.34%
(sLN) at room temperature.

In the initial cryogenic temperature measurements, we never performed or re-
ported a direct measurement of the efficiency, because of the difficulty of coupling all the THz energy out of the cryogenic dewar and into the pyroelectric detector. Instead we measured the relative change in THz energy as the temperature was increased from cryogenic to room temperature (the setup was optimized at cryogenic temperature) and assumed an efficiency of 3.7% based on the enhancement factor (3.2) multiplied by the maximum efficiency obtained at room temperature (1.15%). Having since developed better models of the THz generation process, we now better understand why this is not a valid assumption: (1) Due to the noncollinear geometry, optimal alignment of the setup strongly depends on the crystal temperature, so the enhancement factor must compare the efficiency obtained at the optimal setup at one temperature to the optimal setup at another. (2) The pointing of the THz beam as well as its intensity distribution across the beam changes as a function of the crystal temperature. Therefore we must capture all the THz energy to have a reliable measure of the efficiency.
3.8 Experiment: Echelon-based tilted pulse front

THz generation

In the last section, we discussed an experiment achieving 1% THz generation efficiencies from a 1030 nm pump laser. To phase match the IR pump pulse and THz wave, the IR beam is angularly dispersed via a grating, resulting in a continuous tilted pulse front. The THz generated travels orthogonal to the tilted pulse front, thereby keeping pace with the faster-propagating IR pulse. The efficiency was achieved because the pulse duration of 690 fs was located on a sweet spot which minimizes the detrimental effects of the two extremes in pulse duration. On one hand, toward longer pulses, the peak intensity is too low, thus reducing the nonlinear conversion. On the other hand, toward shorter pulses, there is increased degradation of the pulse duration away from the image plane due to angular dispersion [78], which also reduces the total nonlinear conversion. The latter effect is a byproduct of the angular dispersion introduced to phase match the IR and THz beams, but angular dispersion in the IR beam is not a fundamental property needed to generate THz. In fact, if we can somehow bypass the latter effect, then shorter pulses (< 100 fs) from commonly available Ti:Sapphire amplifiers can be used to generate THz radiation with comparable, or even higher, levels of efficiency. In this chapter, we show results from a method that does exactly that.

3.8.1 Coherent addition of Cherenkov THz radiation

Consider an IR pulse traveling through a nonlinear medium with no pulse front tilt. A small amount of THz radiation will be generated before it is outrun by the IR pulse. This THz radiation propagates symmetrically on both sides of the IR beam resembling a Cherenkov cone [2], at an angle $\gamma$ determined by Eq. 3.20, which is rewritten here.

$$\cos \gamma = \frac{n_g(\omega_0)}{n(\Omega)}$$

(3.33)
Figure 3-24: Producing discrete tilted pulse fronts using an echelon. (a) Mechanical drawing of the echelon used in the experiment. (b) Illustration of the discrete tilted pulse front created from reflection off of the echelon. (c) Simulation of the THz wave generated via Cherenkov radiation from a single IR pulse traveling through a nonlinear medium.

Figure 3-24(c) shows the Cherenkov cone of THz generated from a single IR beam. Now consider an array of time-delayed “beamlets” traveling side-by-side. The Cherenkov cones of THz radiation superpose coherently on one side to produce a THz beam with intensity proportional to $N^2 I_0$ where $N$ is the number of beamlets and $I_0$ is the intensity of the THz generated from a single beamlet.

Such an array of beamlets can be created from reflective echelon consisting of multiple steps. When a single collimated input beam is reflected off the echelon, it is spatiotemporally transformed into a discrete tilted pulse front consisting of beamlets reflected off of each echelon step. The spot size of each beamlet is equal to the step height ($H$) and the longitudinal delay of adjacent beamlets is equal to twice the echelon step width ($W$).

Because there is no angular dispersion in this setup, the length over which the tilted pulse front retains its pulse duration is limited by the Rayleigh range of the beamlets, or $z_R = \frac{\pi w_0^2}{\lambda}$, which is independent of the spectral bandwidth and is typically much larger than the interaction length limited by angular dispersion. This
allows for the advantage of shorter pulses, i.e. higher intensity, to be leveraged.

In this experiment, we demonstrate THz generation using a discrete tilted pulse front pump beam produced by reflection from a gold-coated echelon. Both 1-lens and varifocal zoomlens imaging systems were used to transport the reflected beam to both room temperature and cryogenic temperature crystals. We achieve efficiencies significantly higher than those obtained by grating-based tilted pulse front and observe different transmitted IR spectrum.

3.8.2 Experimental setup

Our echelon (schematic shown in Figure 3-24(a)-(b)) was fabricated by machining a stair-step pattern onto a steel substrate followed by coating a layer of gold. The step width (W) was $150 \pm 0.2 \mu m$ and the step height (H) was $69.00 \pm 0.02 \mu m$, forming a discrete tilted pulse front angle of $42.6^\circ$. The beam incident on the echelon had a width of approximately 9 mm, so the number of beamlets was approximately 60. For a given imaging demagnification factor of $M \approx 5$, the tilt angle inside the crystal is

$$\gamma = \arctan \left( \frac{2HM}{Wn_{LN}^2} \right)$$

(3.34)

Figure 3-25 shows the experimental setup. The pump laser was a 800 nm, 1 kHz Ti:Sapphire amplifier (Coherent Legend) with pulse energy of 1.5 mJ, pulse duration of 70 fs, and FWHM bandwidth of 24 nm. The laser pulse is split 90%/10% to the THz generation / electro-optic sampling beamlines. In the THz generation beamline, the pump is reflected at normal incidence with respect to the echelon steps. The echelon is slightly tilted downward to allow the beam to pass the mirror that originally reflected it toward the echelon. The maximum delivered energy to the crystal was about 1 mJ. Two imaging configurations were used: (1) the 1-lens imaging system consisted of a 8 cm achromatic lens with distances set according to the lensmaker equation, while (2) the varifocal zoomlens was a homebuilt 3-lens system consisting of focal lengths $f_1 = 30 \text{ cm}$, $f_2 = -7.5 \text{ cm}$, $f_3 = 7.5 \text{ cm}$, with a magnification range of 0.16 to 0.29. Cryogenic cooling was done using a commercial liquid nitrogen dewar (Cryo
CHAPTER 3. ULTRAFAST THZ PULSE GENERATION

Figure 3-25: Experimental setup used for echelon-based tilted pulse front THz generation. Note that the 1-lens and 3-lens (zoom lens) imaging systems are substituted interchangeably into the setup.

Industries) with a custom cold plate for mounting the crystal and a custom vacuum tail for accommodating the input and output windows at the abnormal THz exit angle of 63°. The crystal (1 % stoichiometric lithium niobate) was attached to the cold plate by mechanical pressure with a 1 mm sheet of indium foil in between for cushioning and thermal contact. The generated THz output was collected by a 90° off axis parabolic mirror (3 inch diameter, 3 inch effective focal length) and refocused by a similar mirror but with 2 inch focal length. For EO sampling, a GaP crystal was placed at the focus of the second mirror, and the probe beam was coupled in through a hole in the second mirror. The probe beam, like in most EO sampling setups, was passed through a quarter wave plate and polarizing beam splitter and detected using a balanced photodetector.

While we characterized the THz energy using the conventional methods of pyroelectric detection (Microtech Instruments) and thermal power metrology (TK Instru-
ments), we also used the new method of directly characterizing the THz energy using spatial EO sampling. Since EO sampling provides a direct measure of the absolute electric field (or intensity according to $I = \frac{c \varepsilon_0 n}{2} |E|^2$), we only need to collect the electric field over all space to determine the energy. To do this, instead of focusing the probe beam onto the electro-optic crystal, the probe beam remains collimated, overfilling the crystal aperture and sampling the birefringence on all parts of it. The beam then passes through a quarter wave plate and Wollaston prism, just as in conventional EO sampling. The two split beams from the Wollaston prism are then incident onto a camera, whose focal plane images the surface of the EO crystal. The balancing is then performed post-facto once all the images are collected. This results in a transverse image of the THz field at a particular time step.

### 3.8.3 Results and discussion

Figure 3-26 shows the results of THz generation as a function of pump energy, which was controlled by an upstream half wave plate and polarizer combination. Four datasets are shown, corresponding to the four combinations of room/cryogenic temperature crystal and 1-lens/zoom-lens imaging system. The THz energy plotted here was obtained by the spatial EO sampling method as described above. Pyroelectric and thermal power detection were also used, and their results were consistently a factor of 2 higher than those obtained with the spatial EO sampling method. Hence the results plotted here are conservative numbers for the efficiency.

Note that the efficiency saturates at higher pump energies when using the 1-lens imaging system, which can be explained by the fact that imaging imperfections increase farther away from the optical axis. In contrast, with afocal imaging (i.e. zoom lens) the imaging is perfect everywhere within the paraxial approximation [87]. It is possible that at low pump pulse energies, most of the THz generation occurs at the center of the beam where intensity is greatest, leading to comparable performance in both 1-lens and zoom lens configurations; whereas at high pump energies, the efficiency at the center of the beam saturates due to spectral distortion [97], and further growth in efficiency depends on whether the edges of the beam are well-
The main results are summarized in Table 3.5. In terms of maximum efficiency, the zoom lens imaging performed better than 1-lens imaging in both room and cryogenic operating temperatures, consistent with the hypothesis that better imaging leads to more parts of the beam participating in efficient generation. The efficiency enhancement due to cryogenic cooling was also slightly higher for the zoom lens. This can be explained potentially by the fact that the 1-lens imaging setup may have limited interaction length because of beam divergence, whereas the zoom lens retains a collimated beam within a Rayleigh range on both sides of the crystal.

The maximum achieved efficiency of 0.33% is comparable as the record efficiency achieved via grating-based tilted pulse front on Ti:Sapphire lasers of 0.35% [6], a setup which used > 200 fs pulses. If our efficiency based on pyroelectric detection was used, it would be a factor of 2 higher than the current record.

EO sampling traces and spectra of the generated THz in Figure 3-27(a)-(b) show that the pulses are single-cycle with a slight positive chirp. Spectra from cryogenic operation always have more high frequency content than those from room temperature operation, as expected, since absorption favors the higher frequencies. Interestingly, the spectra for 1-lens imaging have larger bandwidth than those for zoom lens imaging. This may be explained by the fact that the pump beam divergence from 1-lens imaging
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<table>
<thead>
<tr>
<th>Setup</th>
<th>Max THz energy (µJ)</th>
<th>Max efficiency %</th>
<th>Cooling enhancement</th>
</tr>
</thead>
<tbody>
<tr>
<td>1L RT</td>
<td>1.6</td>
<td>0.19</td>
<td></td>
</tr>
<tr>
<td>1L CT</td>
<td>2.4</td>
<td>0.27</td>
<td>1.4</td>
</tr>
<tr>
<td>ZL RT</td>
<td>2.1</td>
<td>0.21</td>
<td></td>
</tr>
<tr>
<td>ZL CT</td>
<td>3.1</td>
<td>0.33</td>
<td>1.6</td>
</tr>
</tbody>
</table>

Table 3.5: Summary of results from echelon-based THz generation. 1L, 1-lens imaging system. ZL, zoom lens imaging system. RT, room temperature. CT, cryogenic temperature.

Figure 3-27: Characterization of the echelon-based THz beam. (a) Electro-optic sampling trace of the THz waveform. (b) Corresponding THz spectra. (c) Beam profile of the THz at the focus of the second off axis parabola, obtained using the spatial EO sampling method described in the text. 1L, 1-lens imaging system. ZL, zoom lens imaging system. RT, room temperature. CT, cryogenic temperature.

leads to more phase matching of the higher frequency THz components. Although it has overall less efficiency, it appears to promote higher efficiency near the high frequency regions of the spectrum (∼2 THz).

3.8.4 Conclusion

The echelon is an optical element that can convert a collimated IR beam into a collection of time-delayed “beamlets” which looks like a discrete tilted pulse front. When the discrete tilted pulse front is transported into a lithium niobate crystal via either 1-lens or zoom lens imaging, it generates THz efficiency due to the coherent addition of the Cherenkov THz waves generated from a single beamlet. Our results show that with zoom lens imaging and cryogenic lithium niobate crystals, the echelon-based method can generate THz with efficiencies comparable to the conventional
grating-based TPF technique.
3.9 Experiment: Recycling and characterization of residual pump beam

3.9.1 Abstract

Tilted-pulse-front optical rectification, a predominant method for generating high field THz, yields THz pulses with efficiencies up to 1%, while the residual 99% of the energy remaining in IR pump is often beam-dumped and unused. Here, we demonstrate recycling and recompression of the residual IR for additional THz generation, electro-optic sampling, or other downstream applications. Limitations to the recycling efficacy are linked to spatiotemporal distortions of the residual pump beam through careful spectrally-resolved beam profile characterization.

3.9.2 Introduction

Tilted-pulse-front THz generation was first proposed in 2002 [42] and has gained mainstream adoption, along with organic crystals [113], as a predominant method of laser-based THz generation over the past decade [121]. Numerous improvements have been added to improve the peak field [48], energy [30, 31, 125], efficiency [51, 52] of the generated THz. Some efforts include expanding the THz frequency range [119] and bandwidth [15], while others consider the impact of pump beam format on the THz generation efficiency [67, 87]. Still others investigate the spatiotemporal distortions of the generated THz beam [76].

While much work has been dedicated to understanding the generated THz beam, no experimental analysis or utilization of the residual IR beam has been published to our knowledge, despite the fact that nearly 99% of the total laser energy remains in that beam. The difficulty lies lack of understanding of the spatiotemporal distortions induced on the IR beam due to the nonlinear process. These distortions have been studied numerically in [97, 98] and are predicted to be the main factor limiting conversion efficiency [97].

In this work, we use the residual pump beam to generate additional THz radiation
from a second crystal. We observe that the recycled efficiency decreases when the original efficiency increases, due to the distortions on residual pump induced by the THz beam. Experimentally, we visualize these distortions spatially and spectrally to better understand the tilted pulse front nonlinear conversion process.

Furthermore, this work compares the relative performance of continuous and discrete tilted pulse fronts for THz generation at 800 nm. The residual pump recycling measurements are performed in both grating- and echelon-based tilted pulse front setups having similar input spectra and spot sizes.

### 3.9.3 Experimental setup

Figure 3-28 shows setup for both grating-based and echelon-based THz generation. The pump laser, a 800 nm Ti:Sapphire regenerative amplifier (Coherent), delivered transform-limited 70 fs pulses at 1 kHz repetition rate. A flip mirror is used to route the beam, which has a waist size of 3 mm, to either the echelon-based or grating-based tilted pulse front setups. Each setup starts with a Galilean beam expanding/reducing telescope to ensure that the final spot sizes on the lithium niobate crystals are close to equal, as shown in Figure 3-28(c1) and (c2). However, the spot size in the grating setup is nonetheless smaller than that of the echelon setup by about 20%. Therefore, for the pump fluence dependence data described later in Figure 3-29, the impinging energy ranges are scaled accordingly, with max energies of 1.0 mJ for the echelon setup and 0.63 mJ for the grating setup.

In the echelon-based setup, a discrete tilted pulse front is induced by reflecting at normal incidence off the reflective echelon as described in 3.8. The echelon is tilted downward such that the beam passes under the previous mirror and impinges a zoom lens with magnification close to $M = 1/5$ which images the echelon discrete tilted pulse front onto the first lithium niobate crystal (LN1). Fine-tuning of the pulse front tilt angle was achieved by slightly adjusting the zoom lens magnification.

In the grating-based setup, a continuous tilted pulse front is induced by diffracting the pump beam off a 1800 l/mm grating ($\theta_d = 56.9^\circ$). Fine-tuning of the pulse front tilt angle was achieved by slightly adjusting the grating incidence angle. The tilted
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Figure 3-28: Schematic of both echelon- and grating-based tilted pulse front THz generation, along with their respective residual pump recycling setup. (a) Lithium niobate crystal pumping and transmission geometry. (b) Pump power transmission through the lithium niobate crystal as a function of fluence. (c1) Profile of beam impinging the first lithium niobate crystal (LN1) for the echelon setup. Note that the beam contains strong spatial modulations due to the grooves on the echelon. (c2) Profile of beam impinging LN1 for grating setup. (d1,d2) Residual pump recycling configuration, where the second lithium niobate crystal (LN2) is placed near the image plane of LN1. (e1,e2) Residual pump characterization configuration, where a camera is placed in the image plane of the LN1. E, reflective echelon with 150 μm step width and 69 μm step height. G, 1800 l/mm holographic grating; L1: 25 cm spherical lens; L2: 15 cm spherical lens; LN1: first lithium niobate crystal; LN2: second lithium niobate crystal (identical to LN1); PD: THz pyroelectric detector; KE: Knife edge; Slit: 180 μm slit at Fourier plane; Cam: Beam profiling camera; HWP: Half wave plate; TFP: 45°; Thin film polarizer.
pulse fronts are demagnified and relayed by a $M = 3/5$ confocal telescope with $f_1 = 25$ mm and $f_2 = 15$ mm onto a 5% MgO-doped congruent lithium niobate at room temperature. Optionally, the 70 fs pulses can be elongated to roughly 200 fs by using a knife edge at the Fourier plane of the confocal telescope to narrow the spectrum. The resulting spectral width (5.5 nm) and pulse duration (200 fs) are then close to optimal for grating-TPF THz generation efficiency at 800 nm, according to [6, 120]. This knife edge was used for the residual pump characterization measurements discussed in Section 3.9.5.

The LN crystal is shaped like an isosceles prism ensuring that a normally-incident beam entering through facet A exits the crystal at facet C at a normal angle, after it experiences total internal reflection (TIR) off facet B (Figure 3-28(a)). The total propagation length within the crystal is 11.5 mm regardless of the distance $h$ between the incident beam and the nearest vertex (see Figure 3-28(a)). It was verified that the residual pump spectrum remains the same despite changes to $h$ or decreases in crystal temperature (down to 100 K). A pyroelectric detector (Microtech) was used to measure the generated THz energy, which exits the crystal at normal incidence to facet B. The residual pump beam exits through facet C.

### 3.9.4 Residual pump recycling

Under the paraxial approximation, a 1:1 confocal telescope image-relays each ray from the object plane to the image plane with perfect preservation of its position and angle. Thus, the TPF impinging the first LN crystal (LN1) can, in principle, be reconstructed at a second LN crystal (LN2) by simply placing a 1:1 confocal telescope after LN1 as shown in Figure 3-28. The transmission of the IR beam from the input of LN1 to the input of LN2, shown in Figure 3-28(b) is about 82% owing primarily to Fresnel losses at LN1 and does not depend on the pump intensity, implying minimal multiphoton absorption.

The THz energy and efficiency in both setups as a function of input pump fluence are shown in 3-29(a)-(b). The echelon performs better than the grating over all fluences. At maximum fluence of 25 mJ/cm² the efficiency of the echelon is 3.5 times
Figure 3-29: Recycling performance of residual pump pulse as a function of pump fluence. (a) Absolute THz energy generated from LN1 and LN2. (b) Conversion efficiency at LN1 and LN2. (c) Ratio of the LN2 efficiency to LN1 efficiency. (d) and (e) Residual pump spectrum for various pump fluences. All measurements are done for both the grating (GRT) setup and echelon (ECH) setup. (f) Redshift of the centroid wavelength of the transmitted spectrum as compared to the input spectrum.
higher than that of the grating, illustrating the advantage of using discrete tilted pulse fronts to circumvent the limitations of angular dispersion for THz generation.

Figure 3-29(c) plots the ratio of the LN2 to LN1 efficiencies. The ratio, henceforth called recycled efficiency ratio, is reduced as the pump fluence is increased, going from 60% to 17% for the echelon and 31% to 10% for the grating. Note the transport loss from LN1 to LN2 of ~18% (Figure 3-28(b)) is accounted for in the LN2 efficiency calculation. This reduction in recycled efficiency ratio is due to the nonlinear spatiotemporal distortion of the residual pump beam, whose effect strengthens with pump fluence. The fact that the recycled efficiency ratio is higher for the echelon may be attributed to the fact that spatial-spectral distortions are not as severe as those of the grating (see Section 3.9.5). Spectrally, the pump beam redshifts dramatically to the point where the wavelengths at the extremes are no longer well phase matched for THz generation [97]. These redshifted spectra are shown in Figure 3-29(d) and (e).

We quantify the redshifting via computing the difference in the wavelength centroid between the transmitted spectrum and the input spectrum, or

$$\lambda_{\text{redshift}} = \lambda_c^{\text{transmitted}} - \lambda_c^{\text{input}}$$

(3.35)

where the wavelength centroid of a spectrum $S(\lambda)$ is defined as

$$\lambda_c = \frac{\int_0^\infty S(\lambda) \lambda d\lambda}{\int_0^\infty S(\lambda) d\lambda}.$$  

(3.36)

The redshift increases with input fluence, as shown in Figure 3-29(f), reflecting the production of THz photons via intrapulse difference frequency generation. We can approximate the intrinsically generated efficiency (THz that is generated but is not detected due to absorption, reflection, or other loss) via the redshift by

$$\eta_{\text{redshift}} = \frac{\lambda_{\text{redshift}}}{\lambda_c^{\text{input}}}.$$  

(3.37)

This efficiency calculated from the redshift, shown in the right vertical axis in
Figure 3-29(f), shows that over 2% (1.4%) of the IR energy is converted to THz in the grating (echelon) setup (assuming no other loss mechanisms for IR). Surprisingly the echelon experiences less intrinsic conversion compared to the grating, even though its collected efficiency is much higher.

The material GDD induced by propagation through the first crystal (4235 fs$^2$) is not compensated automatically by the confocal telescope, the way that the GDD due to angular dispersion (GDD-AD) [1] (-57857 fs$^2$ over length of crystal) is. Thus, the material GDD elongates the transform-limited 70 fs pulse at the image plane to 182 fs. However, the effect on recycled efficiency is likely small, since the second crystal, which is aligned for maximum efficiency, can simply be moved downstream by 3.97 mm to offset the material dispersion through the negative valued GVD-AD (1067 fs$^2$/m in air). Over this distance, the spatial walkoff due to angular dispersion is negligible and experiments have predicted that the efficiency varies by less than 20% when using GVD-AD to compensate for residual GDDs of up to $3\times10^5$ fs$^2$ [14].

Finally, we show the EO sampling waveforms and spectra in Figure 3-30. The echelon produces a broader THz spectrum than the grating, since the generation bandwidth is not as limited by GDD due to angular dispersion. The recycled spectra are much narrower for both setups, indicating that the transmitted pump has significantly more GDD and other spectral distortions which limit the generation of higher THz frequencies. We explore the distortion of the residual pump pulse further in the next section.

### 3.9.5 Residual pump characterization

By replacing the second crystal (LN2) with a camera as shown in Figure 3-28(e1), we can also study the residual pump’s spatial profile. The camera longitudinal position was precisely positioned to minimize the beam width on the camera, so that we image the plane of overlap (henceforth called object plane) between the angularly dispersed rays. We also observed that this position best resolved the small scratches on the surface of the grating, which only verifies that we are imaging the object plane correctly. The measurements performed here were only for the grating-based setup.
Characterization of the residual pump beam emerging from the echelon-based setup is the subject a future study.

The captured beam profiles are spectrally resolved by placing a scanning slit in the Fourier plane (between the two L3’s in Figure 3-31(a). The best spectral resolution of 0.5 nm was achieved using a slit width of 180 $\mu$m. We found that the spectral broadening dynamics can be more clearly seen when the input beam has a narrow bandwidth, so we clip the input spectrum using a knife edge (KE) in the Fourier plane of the telescope between the grating and LN1 to have a 5.5 nm FWHM bandwidth. The resulting input spectrum can be seen Figure 3-31(a).

The distortion to the residual pump caused by the nonlinear process in the LN crystal has spatial features in addition to spectral features since the THz and IR propagate noncollinearly. To illustrate this, Figure 3-31(a) shows the amplification of a single THz ray as it propagates from its generation point (henceforth called “bottom”) to the region where it loses spatial overlap with the pump beam (henceforth called “top”). Throughout this propagation, the THz pulse walks across the transverse extent of the IR pulse front, inducing nonlinear effects of different proportions in each section.
Figure 3-31: Residual pump characterization of grating TPF. (a) Illustration of the amplification of a single THz ray. Note that the ray moves across the transverse extent of the IR pulse front. In experiences different amounts of difference frequency and sum frequency generation in different transverse regions of the IR beam, and these differences account for the spatially-dependent spectral distortions of the IR beam. (b) Spectrally-resolved beam profiles of the residual pump at different fluences.
The amplification of a three wave process depends on the strength of the two beating fields. Thus, the amplification of a redshifted optical wave $A_{\text{IR}}(\omega - \Omega)$ (plus THz wave) via difference frequency generation (DFG) between two optical fields is proportional to those two field strengths $A_{\text{IR}}(\omega)A_{\text{IR}}(\omega - \Omega)$. Similarly, the amplification of a blueshifted optical wave $A_{\text{IR}}(\omega + \Omega)$ via sum frequency generation (SFG) between an optical and a THz field is also proportional to those two constituent field strengths $A_{\text{IR}}(\omega - \Omega)A_{\text{THz}}(\Omega)$.

We analyze three different points along the THz path. At point A, no THz field is yet generated and the IR field is weak (being at the edge of a Gaussian spatial profile), leading to little three-wave-mixing. At point B, some THz field is present and the IR field is strong, leading to large difference frequency generation (DFG) between the IR components and some sum frequency generation (SFG) between the THz and IR components. At point C, the THz field is relatively high and the IR field is relatively weak, leading to a plateau in DFG and strong SFG. This behavior is verified by the spectrally-resolved 1D beam profiles shown in Figure 3-31(a).

The asymmetric dynamics can be more comprehensively visualized by plotting, in Figure 3-31(b), the spectrally-resolved beam profiles for various pump fluences.

For fluences below 6 mJ/cm$^2$, we observe that the amount of redshift increases monotonically with decreasing x position, or in other words, the redshift tends toward the top of the beam. Beyond a fluence of 6 mJ/cm$^2$, however, the redshift position moves toward the bottom of the beam. This is due to the rapid redshifting occurring right when the pulse enters LN1, before the it arrives at the object plane. Due to noncollinear phase matching, each redshifted spectral component is associated with a k-vector whose x-component is proportional to its wavelength and directed toward the bottom of the beam. According to calculations, during the 0.7 mm propagation distance from the input face of the crystal to the object plane, the most redshifted k-vector at 850 nm can travel an x distance of over 1 beam width away from center of the original beam. Indeed, Figure 3-32 shows experimental evidence of this spatial walkoff effect using spectrally-resolved z-scan measurements. Since the centroid of these redshifted components does not intersect with the input spectral components,
they can be effectively treated as chromatic aberration.

Another feature in Figure 3-31 is the spatially-invariant spectral modulation occurring at wavelengths beyond $\sim 810 \, \mu m$. This modulation period is in the range of 1 THz and decreases with increasing fluence, as shown in Figure 3-33. These modulations arise from the beating between the optical and THz waves to generate new redshifted optical wavelengths. The modulation frequency should be comparable to the THz center frequency before any significant absorption. Other studies have shown that the intrinsically generated THz does indeed have a center frequency in the 1 THz range. As the fluence increases, the generated THz frequency decreases from 1.2 THz to 0.85 THz due to the phase mismatch between higher THz frequencies and longer redshifted wavelengths.
3.9.6 Conclusion

We have shown experimentally that the residual pump pulse can be recycled with an efficacy decreasing with the input pump pulse fluence. The recycled efficiency ratio is higher for discrete tilted pulse front pumping, which may be attributed to the different pump distortions resulting from continuous vs. discrete TPF nonlinear dynamics. To visualize the pump distortions in the case of the grating-based setup, we characterized the residual pump beam using spatially-resolved beam profiles. Our data show that the amount of redshifting and blueshifting are very different across different parts of the beam, and can be traced to the relative amplitudes of the THz and IR beams as they travel noncollinearly in the nonlinear medium. Finally, our experimental data support the claim that the discrete tilted pulse front scheme from the echelon performs significantly better than the continuous tilted pulse front scheme from the grating for short $\sim 100$ fs pulses.

While the current recycled efficiency ratio may not be high enough to warrant the construction of a second stage, several possibilities exist for future efforts. For example, a linear spatial light modulator may be placed in the Fourier plane to correct the distorted phase of the various frequencies before the second crystal. With the large amount of power remaining in the residual pump beam, efforts in this area are well worth pursuing. Figure 3-34 offers an application for additional THz sources pumped by recycled beams in the form of a multi-staged THz accelerator.
Figure 3-34: Conceptual drawing for a multistage THz accelerator powered by a cascade of recycled residual pump beams.
Chapter 4

THz streaking

As discussed in the thesis overview (Chapter 1), femtosecond electron bunches with keV-MeV energies and eV-keV energy spread are used to seed x-ray FELs [79]. However, the ultrashort electron bunches are also used directly by condensed matter physicists as a tool for ultrafast electron diffraction (UED). UED has been used to resolve state transitions in carbon nanotubes [112], molecular structures [53], organic salts [33], and charge density wave materials [20]. Such semirelativistic electron sources are not only of interest for ultrafast electron diffraction [4, 80], but also for electron energy-loss spectroscopy [112, 19].

Thus far, the output energy spread (hence pulse duration) of ultrafast electron guns has been limited by the achievable electric field at the surface of the emitter [80], which is 10 MV/m for DC guns [74] and 200 MV/m for RF guns [126]. Single-cycle THz fields provide a unique opportunity to not only achieve GV/m surface electric fields (e.g. as high as 27 GV/m at 0.47 THz [107]), but also with relatively low THz pulse energies, since a single-cycle transform-limited waveform is the most efficient way to achieve intense electric fields.

In this chapter, we study how single-cycle THz fields interact with electrons near the point of emission. In this regard, Li et al. and Wimmer et al. have demonstrated THz streaking from excited atoms [72] and an isolated nanotip [116], respectively. While the basic physics of their work, like ours, demonstrates strong THz field-induced electron manipulation from rest, it is geared toward understanding low-charge (<0.1
fC) electron emission from atoms and nanostructures. Unlike these earlier experiments, we study this interaction for the first time in the extreme conditions of GV/m THz fields and strong space charge on a bulk metal cathode platform, with the goal of building a single-cycle THz electron gun.

As a proof-of-concept, we demonstrate a mean acceleration of 18 eV on 50 fC electron bunches using a 6 $\mu$J, single-cycle THz field with peak field of 72 MV/m (ponderomotive energy of 28.5 eV). The experimentally obtained acceleration results are in good agreement with particle tracking simulations. To demonstrate the scalability of this approach to the keV energies required of an electron gun, we show via simulation that a 2 GV/m THz pulse can achieve a mean energy of $\sim$100 keV with a RMS energy spread of 1.3%.
4.1 Physics of THz streaking

For a non-relativistic electron to acquire a net energy gain from an electromagnetic wave, i.e. streaking, at least one of two conditions must be met: (1) the spatial extent of the field has micro- to nano-scale features such that the electron enters or exits the field ‘abruptly’ or (2) The electron is emitted or ‘born’ in the field. Condition (1) is easily achieved in conventional electron guns and has been satisfied by introducing physical boundaries which reflect [63] or absorb [93] the electromagnetic wave but allow passage of the electron, or by using nanotips, which enhance the field by ten- to hundred-fold within a nano-localized volume [46, 116]. In contrast, Condition (2) can be met by simply emitting an electron in the presence of an accelerating field (emit-and-accelerate scheme) on a flat surface of copper (which acts as the material boundary).

In our streaking experiment, Condition (2) was met by emitting the electrons from a flat copper surface in the presence of the THz field. Bulk copper can withstand $>1$ GV/m of field strength at THz frequencies [107] and a diffraction-limited beam at 0.45 THz focused on it has a beamwaist of 0.6 mm, allowing long acceleration volumes in comparison to the localized regions on nanotips. Thus, a flat copper photo-cathode is a good platform for THz-based acceleration because it is scalable by orders of magnitude in bunch charge and field strength without risk of optical damage.

To provide basic understanding of the electron dynamics before describing experimental results, Figure 4-1 shows an animation of emission and acceleration of a multi-electron bunch by a single-cycle THz field for our experimental conditions. The
electron momentum gain $p$ is described by the THz electric vector potential:

$$ p = -eA = -e \int_{t_{emit}}^{t} E_{\text{thz}}(t') dt' $$

and the kinetic energy gain at nonrelativistic energies is described by

$$ KE = m_0 c^2 \sqrt{1 + \left( \frac{p}{m_0 c} \right)^2} - m_0 c^2 \approx \frac{p^2}{2m_0} $$

Figure 4-2(top) shows the THz field at the photocathode surface as a function of time. The time-energy evolution of a single electron exposed to the THz field is shown in Figure 4-2(bottom) for two field strengths. Photoemission occurs at -0.47 ps, the first node, or zero-crossing, of the THz electric waveform where the field switches from positive to negative. Shortly after emission, for a THz field of 72 MV/m (blue curve), the electron is accelerated to 120 eV by the main negative half-cycle of the THz waveform (-0.47 to 0.47 ps) and thereafter decelerated to 26 eV by the final positive half-cycle (0.47 1.43 ps).

Figure 4-3 shows the time evolution of the energy spectrum for an electron bunch of 50 fC. Due to the long 525 fs green pulse (over which the electrons are emitted),
some electrons are emitted during a phase of the THz which does not maximize the vector potential and therefore receive less acceleration. As a result, the spectrum has a mean energy below that given by the vector potential, unlike [26]. The evolution of the electron bunch is determined in two stages. At short time scales (<3 ps), the strong THz field dominates the energy of the bunch and the bunch behaves largely as that of the single electron.

At long time scales (>3 ps, after the THz has passed), space charge forces cause leading/trailing electrons to gain/lose energy, resulting in a chirped energy spectrum. We obtain a mean electron energy of 20 eV and a peak energy of 90 eV (Figure 4-3(d)). Given that the energy gain scales quadratically with the accelerating field in the nonrelativistic regime, the red curve in Figure 4-3(b) and plots in Figs. 2(e) and 2(f) show that a 2 GV/m field will achieve a mean energy of 27 keV. Additionally, because the electrons are quickly accelerated out of the low-energy regime, the RMS energy spread is reduced to 3.5%. By modifying the anode and using a shorter green pulse duration, a mean energy of 99 keV and spread of 1.3% can be achieved (see Section 4.4).

4.1.1 Particle tracking method

The particle tracking simulation implemented to model the evolution of an electron bunch in the presence of the THz field is described here briefly.

The trajectories were modeled by integrating the kinematic equations for every particle \( i \) using a 4th order Runge-Kutta solver.

\[
m \frac{dv_i}{dt} = F_{field} + F_{bias} + \sum_j (F_{im,ij} + F_{sc,ij}) \tag{4.3}
\]

\[
\frac{dr_i}{dt} = v_i \tag{4.4}
\]

Here, \( m \) is the relativistic mass, \( F_{field} \) is the electric force due to the THz pulse, \( F_{bias} \) is the electric force due to the DC bias, \( F_{im,ij} \) is the image charge force acting
Figure 4-3: Electron acceleration in a single-cycle terahertz field. (a) Electro-optic sampling trace of terahertz pulse. (b) Single-electron energy evolution in time. After emission (indicated by the green dashed line), the electron is accelerated by the negative half-cycle of the THz electric field and thereafter decelerated by the final positive half-cycle. Energies gained from a 72 MV/m (blue) and 2 GV/m (red) field are 26 eV and 27 keV, respectively. (c) 50 fC electron bunch energy evolution under a 72 MV/m THz field showing the effect of finite electron bunch duration and space charge. At long time scales, space charge induces an energy chirp on the spectrum. (d) The final energy spectrum (gray line, simulation; black dots with one sigma error, experiment) shows an mean energy of 20 eV (18 eV in experiment) with a peak energy of 90 eV (92 eV in experiment).
on the $i$th particle from the image charge of the $j$th image particle in the bulk metal cathode, and $F_{sc,ij}$ is the particle-particle Coulomb (space charge) force. The THz beam was modeled as a fundamental Gaussian beam with numerical aperture of 1/2. The THz pulse spectrum was directly adapted from that obtained by EO sampling with a flat spectral phase profile.

Parameter values matching those in the experiment were used routinely in the simulations (Section 4.2 describes these experimental parameters in more detail). The initial electron bunch had a Gaussian spatial profile with beamwaist of 50 $\mu$m and a Gaussian temporal profile with FWHM of 375 fs. The initial kinetic energy was 0.18 eV (green two-photon energy subtracted by the copper work function) and the initial velocity vectors were uniformly distributed over a hemisphere. The particles were released 0.1 nm away from the surface. 6000 macroparticles were used to represent a total bunch charge of 50 fC, corresponding to a charge of 52e- per macroparticle, where e- is the unit charge.
Figure 4-4: Experimental setup. (a) Electrons are emitted from a flat copper cathode by a 525 fs green (515 nm) pulse and accelerated by a p-polarized single-cycle terahertz pulse. Electron spectrum information is obtained by applying a retarding bias and measuring the photocurrent. (b) Photograph of apparatus inside a vacuum chamber. (c) Terahertz beam intensity profile at the focus.

4.2 Experimental setup

In the experiment, schematic and photograph shown in Figures 4-4(a)-(b), an emit-and-accelerate scheme is employed on a flat copper photocathode, akin to the cathodes used in conventional accelerators (e.g. LCLS [21]) and well suited to high-electron-brightness applications due to its simplicity and robustness. Electrons are emitted by two-photon ionization at the photocathode using a 525 fs pulse at 515 nm (green) at 1 kHz repetition rate with a total charge per bunch of 50 fC. The photocurrent was measured at the photocathode. The emitted electrons were exposed to a p-polarized, SC THz pulse with a carrier frequency of 0.45 THz (Figure 4-4(a)). The reflection from the photocathode increases the THz field twofold. The THz pulse delivered onto the copper is focused to a beam waist of 1.1 mm (Figure 1(c)) and has a final energy of 6 µJ with a calculated peak electric field on the surface of 72 MV/m.

The performance of the THz gun was determined by the electron energy spectra of the emitted electron bunches. The spectra were measured by taking the derivative
of the photocurrent as a function of bias voltage. At reverse biases, the bias acts as a highpass filter for the photocurrent and therefore allows the measurement of the yield of electrons above an energy equal to the reverse bias times the unit charge. This setup essentially functions like a retarding field analyzer without a grid [103].

4.2.1 THz beam delivery

The THz pulses are generated by optical rectification (OR) of tilted-pulse-front near-infrared (NIR) pulses in cryogenic lithium niobate (LN). Up to 12 $\mu$J THz pulse energy is generated with a conversion efficiency of about 1.0% at the crystal. The diverging THz beam is collimated by a 50.8 mm high-density polyethylene lens, converted from s- to p-polarization by a periscope, coupled into the vacuum chamber, and focused onto the photocathode by a parabolic mirror with an effective focal length of 25.4 mm. The THz beam is incident onto the photocathode at an angle of 67$^\circ$ (largest angle by which a f/1 focused beam can be incident on a flat surface without clipping). The large angle of incidence maximizes the electric field component normal to the photocathode surface.

The THz pulse is temporally characterized by a standard electro-optic (EO) sampling setup employing a 200 $\mu$m thick, 110-cut ZnTe crystal. 70 fs NIR pulses from the mode-locked fiber oscillator (see Section 4.2.2) sample the THz field-induced birefringence as a function of delay. A quarter-wave plate followed by a polarizer converts the field-induced birefringence to an intensity modulation, and the intensity modulation is recorded by a photodiode. The measured pulse has a carrier frequency of 0.45 THz and a FWHM bandwidth of 0.4 THz. The THz beam was characterized spatially by a pyroelectric detector array (Spiricon Pyrocam III).

4.2.2 Pump laser

The pump source for THz generation is a Yb:KYW chirped pulse regenerative amplifier (RGA) producing 1.5 mJ pulses with 1 kHz repetition rate at a near-infrared center wavelength of 1030 nm and bandwidth of 2.1 nm. The dielectric grating com-
pressor following the RGA compresses the pulses to a transform-limited pulse duration of 750 fs. The seed for the RGA was a mode-locked Yb-doped fiber oscillator emitting 70 fs, 0.2 nJ pulses at 80 MHz amplified to 1.6 nJ by a Yb-doped fiber amplifier [50]. After losses through the optical elements in the setup, the impinging pump energy into the (LN) crystal was 1.2 mJ.

### 4.2.3 Photoemitter laser

About 2\% of the available NIR pump energy was used to generate the green photoemitter pulses by second harmonic generation in a 0.75 cm long BBO crystal. A BG-39 bandpass filter was used to remove the fundamental NIR component. The green pulses have a FWHM duration of 525 fs and are focused at an angle of 67° onto the photocathode with a beam waist of 25 μm. Since the emitted charge scales as the green pulse’s intensity squared due to two-photon ionization, the effective duration of the electron bunch at emission is estimated to be lower than the duration of the green pulse approximately by a factor of $\sqrt{2}$, corresponding to 375 fs. Since both the THz and green pulses are produced from the same 750 fs NIR laser, the timing jitter between them is negligible (see Section 5.3.4 for a full characterization).

### Anode and cathode

A Poisson equation solver (Superfish [57]) was used to model the static DC electric potential between the anode and cathode and it confirmed that the bias field is uniform over the emission area. Varying the DC bias between the cathode and anode does not impact the electron dynamics during the exposure to the THz field for two reasons. First, the bias was swept over +/- 110 V, corresponding to a DC field of +/- 52 kV/m, which is more than three orders of magnitude weaker than the THz field. Second, during the transient interaction with the THz pulse on the picosecond timescale, the electron dynamics are dominantly governed by the THz field.
4.3 Results

4.3.1 Streaking and acceleration

In Figure 4-6(a), the experimentally measured spectrogram is plotted as a function of energy and delay (between the green pulse and THz pulse) for 0, 36, and 72 MV/m THz field strengths. Note, a positive delay corresponds to the situation where the THz pulse arrives after the green pulse and vice versa. At a delay of 0.25 ps at 72 MV/m, we observe photocurrent up to 92 eV while the mean energy was measured to be 18 eV. This is in good agreement with the simulations shown in Figure 4-3(c) that predict a peak energy of 90 eV and a mean energy of 20 eV.

The pulsewidth of emission is 17% of the THz carrier period and is therefore
capable of revealing THz electric field phase-dependent effects. Figure 4-6(b) plots the photocurrent as a function of delay at a fixed bias voltage of -0.1 V. A similarity between the photocurrent trace and the integrated THz field is observed. Given the fact that the photocurrent is proportional to the mean electron energy in this bias regime, the data reflects that the electron momentum gain is governed by the vector potential,

\[ p = -e \int_{t_{\text{emit}}}^{\infty} E(t')dt'. \]  

(4.5)

In attosecond streaking experiments [66], electrons emitted by an attosecond x-ray pulse sample the waveform of an visible streaking pulse. Analogously, here in THz streaking, the electrons emitted by a femtosecond visible pulse sample the waveform of a THz streaking pulse. Thus, this method provides an alternative to EO sampling as a way to characterize the THz temporal profile.

In Figure 4-6(c), we compare the bias sweep with THz OFF and THz ON (36 MV/m) at a delay of 0.25 ps. We observe a significant rise of photocurrent at reverse biases up to \( \sim -30 \) V in the THz ON case, indicating clear evidence of THz acceleration since an electron traversing the reverse bias and registering current on the anode must have had energy greater than \( eV_{\text{bias}} \).

It is also interesting to note that at positive biases, the THz OFF case continues to show a gradual increase of photocurrent as a function of bias, while the THz ON case shows a flattening of the curve. This behavior, as confirmed by simulation, is due to space charge. In the case of THz OFF, the space charge effect is so great that even small increases in bias voltage lead to more photoelectrons escaping the surface. In the case of THz ON, most of the emitted charge is pulled from the surface so space charge effects are diminished as is the dependence on bias voltage.

In Figure 4-6(d), the energy spectra are in good agreement with the main features (mean/peak energy and spectral shape) of the simulation results. The broad, monotonically-decaying shape of the energy spectra is due to the low energy and high charge of the bunch, placing it in a regime where space charge effects, rather than
Figure 4-6: Experimental evidence of terahertz-driven electron acceleration. (a) Spectrograms showing electron acceleration at various THz field intensities. At 72 MV/m, we observe an increase of photocurrent at reverse biases up to -92 V, indicating that electrons achieved a peak energy of 92 eV to overcome the potential barrier. (b) Experimental correlation between the photocurrent and integrated terahertz field reflects that the acceleration is governed by the vector potential. The bias here is set to a regime where the photocurrent is proportional to the electron energy. (c) Bias sweeps of photocurrent (dots with one sigma error bar) show a significant rise in photocurrent at strong reverse biases indicating clear evidence of THz acceleration. (d) Electron energy spectra comparing THz OFF and THz ON (36 MV/m). Spectra from (a) and (d) are obtained by differentiating the bias sweeps.
the THz field, plays a dominant role in shaping the spectra. In contrast, narrow, unimodal spectra are featured in most other streaking experiments (e.g. [116]) where the electron bunches are either high energy or low charge.

Finally, the increase in total photocurrent at Bias>0 for THz ON in Figure 4-6(c) is due to the strong THz field acting in conjuction with the static field to increase the space-charge limited charge density [100], allowing more electrons to escape the cathode.

### 4.3.2 Space charge phenomena

The interplay between space charge and the THz field can be further investigated by operating in the strong forward bias regime. At a forward bias of 900 V, or 410 kV/m DC electric field, emitted electrons experience a strong pull toward the anode even in the absence of THz. Instead of the second-harmonic green photoemitter used previously, we use an NIR photoemitter with pulsewidth of 750 fs to achieve greater current densities (more IR energy was available for the experiment). The level of current impinging the anode increases with the strength of the photoemitter, as can be seen in the baseline current of the three plots in Figures 4-7(b)-(d) and plotted as a function of photoemitter energy in Figure 4-7(a)(Inset). However, not all emitted electrons may be collected at the anode. At higher current densities, some later-emitted electrons are repelled by space charge forces from the “pancake” of electrons before it. These later-emitted electrons do not travel very far from the photocathode before being pushed back and recombining with the cathode (see Section 2.3.2).

The NIR emits electrons via 4-photon ionization, so theoretically a photocurrent yield proportional to the fourth power of impinging energy would apply, in the absence of space charge. Instead we measure a reduced power scaling of 1.75 of the baseline photocurrent with photoemitter energy in Figure 4-7(a)(Inset), evidence of transitioning into the space-charge-limited regime.

We recall from Section 2.3.2 that the space-charge-limited regime occurs when $2E_{sc} \geq E_{acc}$, and that the emitted charge is $Q \propto E_{acc}$ for a constant emission area. Why then is the photocurrent still increasing with photoemitter energy when $E_{acc}$
Figure 4-7: Space charge effects in terahertz acceleration. (a) The stronger the space charge force, the more the terahertz pulse enhances the current yield. (b) Terahertz field induces an increase in the current yield even at delays of several picoseconds due to surface dynamics of electrons on copper.
is constant and there is no dependence on photoemitter energy in Eq. 2.64? One possible reason is that the photoemitter spot profile is Gaussian rather than the uniform profile assumed in the Eq. 2.64. While the center of the Gaussian profile, where the charge density is highest, may be space-charge-limited, the edges of the profile may not. Thus, as photoemitter energy is increased, there is still room for photocurrent to increase at the edges of the emission area where charge density is still low. More and more portions of the beam will become space-charge-limited as the photoemitter energy is increased, making for a gradual transition from no space charge to fully space-charge-limited. Hence instead of a power scaling of 4 (no space charge) or 0 (full space charge), we measure an intermediate power scaling of 1.75. This idea is illustrated in Figure 4-8.

Looking further, we observe the effect of THz on photoemission for varying current densities. Figures 4-7(b)-(d) shows the photocurrent as a function of delay for several photoemitter energies at a constant THz field. For an energy of 0.7 µJ, the charge emitted is so small that the DC bias alone is capable of extracting all the electrons, and the THz field does not extract additional yield above the base level. This is evident in Figure 4-7(d), where the peak photocurrent yield with THz (τ_{delay} = 0 ps) is no different from the baseline yield (τ_{delay} = −3 ps). Since the charge is not limited by the field strength (E_{acc}), a photoemitter energy of 0.7 µJ does not yet bring operation into space-charge-limited regime.

As the photoemitter energy is increased to 3.5 µJ, however, the THz field plays a role in increasing the yield, as is reflected in the increased enhancement factor shown.
in Figure 4-7(a). The enhancement factor is defined at the collected photocurrent with THz divided by the baseline photocurrent. Operation at this point is in the space-charge-limited regime. The enhancement factor is further amplified at 7 \( \mu \)J, as the operation goes further into the space-charge-limited regime.

We observe another surprising phenomenon at long delays, shown in Fig. 4-7(e). For THz pulse arrivals before the photoemitter (negative delays), there is no photocurrent modulation. At positive delays (THz pulse arrives after the photoemitter), however, there is a gradual decay of photocurrent as a function of delay even at points where there is no overlap between THz and IR. Furthermore, the decay length is greater for weaker reverse biases. A possible explanation of this asymmetric behavior is the fact that surface charges are generated by multiple scattering within copper and linger in their excited state for several picoseconds. The number of these surface electrons is accentuated with greater photoemitter energy, while the number of electrons emitted at zero delay remains the same due to space charge limitations. Therefore, the effect is greater for higher biases due to the fact that higher biases allow more charge to be emitted. The experimental setup was checked for no back-reflections.
Figure 4-9: 2 GV/m THz gun. (a) Electron bunch spectrogram as a function of distance from cathode in the presence of a 2 GV/m THz field. The bunch is accelerated by the THz negative half-cycle to 99 keV over 83 m. At 83 m (gray dashed line), it enters a hole in the anode and escapes the decelerating half-cycle. (b) A schematic of the modified anode with a hole. (c) Energy spectrum at 125 m from cathode. The RMS energy spread is 1.3%. For this simulation, a green pulse duration of 20 fs and spot size of 12 µm were used. Other parameters such as the THz waveform and bunch charge of 50 fC were preserved.

4.4 Design of 2 GV/m gun

Here we show how the knowledge gained from this THz streaking platform can be leveraged to design a high field (2 GV/m) gun.

With increased accelerating field strength, one can consider a way to prevent the electrons from being decelerated by the final positive half-cycle of the THz pulse. If the anode-cathode spacing is reduced to the distance that the electrons travel over the accelerating half-cycle (83 µm for a 2 GV/m field), then the electrons would be recombined on the anode before the onset of the decelerating half-cycle. Equivalently, a small sub-THz wavelength hole can be drilled into the anode to allow the electrons to escape the THz field region and be used for downstream applications, as illustrated in Figure 4-9(b).

To reduce the energy spread, one can reduce the duration of the photoemitter pulse, such that the electrons are subject to the same vector potential. Figure 4(a) shows the evolution over distance of an electron bunch with an initial duration of 20 fs and transverse size of 12 µm. An energy of 99 keV is achieved over a 83 µm distance.
At 83 \( \mu \text{m} \) (gray dashed line), the bunch enters a hole in the anode and escapes the decelerating THz half-cycle. The RMS spread at 125 \( \mu \text{m} \) is 1.3\%, as shown in Figure 4-9(c).

### 4.5 Conclusion

In conclusion we have demonstrated THz-driven streaking from a bulk surface. From a simple flat copper photocathode, we demonstrated acceleration of a 50 fC electron bunch in agreement with particle tracking simulations. We also showed the dominant effect of Coulomb repulsion on electron spectra under low accelerating fields (72 MV/m). Finally, we showed the dependence of the space-charge-limited regime on photoemitter energy (charge density).

While the proof-of-concept does not yet compare to the performance of state-of-the-art DC guns [111] or RF guns [81], THz guns holds promise due to their orders-of-magnitude higher field breakdown threshold [107, 39]. Further, we argued that with upgrades to readily-available GV/m THz sources and shorter photoemission laser pulses (20 fs), this scheme can yield monoenergetic electrons with energies up to 100 keV, comparable to the performance of current guns used for ultrafast electron diffraction. In the next chapter, we will demonstrate a quasi-monoenergetic gun with near keV energy that was designed with the lessons learned from these streaking experiments.
Chapter 5

THz-driven, all-optical electron gun

As discussed in the previous chapter, the electron accelerator is an important tool for scientific research. The ultrashort electron beams it produces are used to probe atomic dynamics with electron diffraction and injected into accelerators for x-ray light sources (such as free electron lasers, or FELs). We begin the chapter by discussing some common requirements necessary for these applications.

5.1 Requirements of an electron gun

These applications require that the e-beam have the properties shown in Figure 5-1.

- **Narrow energy spread**: Beam emittance is proportional to the product of energy spread and pulse duration. For a compact electron gun without a buncher, the short pulse duration goes hand-in-hand with narrow energy spread. In FELs, low energy spread is needed to satisfy the matching condition. Finally, fairly low energy spread is needed in electron diffraction to resolve individual diffraction fringes. However, spreads of a few percent are tolerable.

- **High bunch charge**: In some ultrafast diffraction experiments, only the total current matters, but in many applications, the charge per bunch is essential.
Instances include diffract-and-destroy imaging of non-repetitive behavior mechanisms \[3\] (e.g. ablation) and coherent x-ray generation where the x-ray flux scales as the square of the charge per bunch.

- **Ease-of-implementation**: Electron guns using klystrons or high voltage electronics to generate the accelerating field are complex.

- **Low timing jitter**: Timing jitter is the RMS variation of the relative delay between the UV photoemitter and the accelerating wave on a shot-by-shot basis, as illustrated in Figure 5-1. Large timing jitter causes experimental data to be either uncertain or ‘smeared out’.

Narrow energy spread and high bunch charge can be achieved by having a **high accelerating field** and a **long accelerating wavelength**. High accelerating field shortens the time it takes for electrons to reach relativistic velocities, which reduces the deleterious effects of space charge. Long accelerating wavelength increases the half-cycle ‘bucket’ over which the bunch is accelerated. By having a larger ‘bucket’, or longitudinal tolerance, more electrons can be accelerated in a single bunch. **Ease-of-implementation** and **low timing jitter** can be both achieved by using a single laser to power both the UV photoemitter and the accelerating wave instead of generating them with separate systems. As shown in Figure 5-1, using the same laser also enables intrinsic stabilization.
5.2 Existing technologies

The realization of a compact electron gun requires very intense electric fields in order to achieve electron bunches with high charge density, good beam quality, and large kinetic energy.

To achieve high fields on the surface of the photoemitter, there are currently two types of electron guns: DC and RF guns. DC guns utilize bulky high voltage electronics, while RF guns utilize high power RF fields, which involve large klystrons, pulsed heating issues [17, 69], and elaborate synchronization schemes [12, 37] with best jitter performance thus far of 96 fs [12]. Furthermore, DC and RF guns have field limitations of around 10 MV/m [74] and 200 MV/m [126], respectively, due to breakdown mechanisms on common accelerator materials such as plasma breakdown [105] or Joule heating [17, 69]. The field and timing stability limitations of DC or RF guns hinders the achievable resolution of time-resolved electron diffraction or x-ray experiments. The first two columns of Table 5.1 summarize these limitations.

The plasma breakdown threshold scales as the square root of the frequency [75] and Joule heating reduces with pulse energy [69]. As a general rule of thumb,

$$E_{thres} \propto \frac{f^{1/2}}{\tau^{1/4}}.$$  \hspace{1cm} (5.1)

Because of this, there has been great interest in the development of compact, short-pulse-driven accelerators operating in higher-frequency regions of the electromagnetic spectrum [8, 10] driven by intrinsically-synchronized sources such as lasers.

Near-infrared (NIR) laser pulses are one option because of their high fields (multi-GV/m), accessibility, and relatively low cost sources. However, due to the short NIR wavelength, phase matching between the electromagnetic field and electron, space charge effects, limited charge density, and fabrication of accelerating structures present challenging hurdles. Laser plasma wakefield acceleration has achieved GeV electrons [24, 71, 83] using 100 TW - PW laser facilities at low repetition rates with percent-level energy spread and jitter.

Nevertheless, the need for higher extraction field and timing stability in a compact
### Table 5.1: Comparison of electron gun technologies

<table>
<thead>
<tr>
<th>DC Gun</th>
<th>RF Gun</th>
<th>THz Gun</th>
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<tr>
<td><img src="DCGun.png" alt="Image of DC Gun" /></td>
<td><img src="RFGun.png" alt="Image of RF Gun" /></td>
<td><img src="THzGun.png" alt="Image of THz Gun" /></td>
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<table>
<thead>
<tr>
<th>Parameter</th>
<th>DC Gun</th>
<th>RF Gun</th>
<th>THz Gun</th>
</tr>
</thead>
<tbody>
<tr>
<td>$E_{\text{acc}}$</td>
<td>$&lt; 10$ MV/m [74]</td>
<td>$&lt; 200$ MV/m [126]</td>
<td>$&lt; 27$ GV/m [107]</td>
</tr>
<tr>
<td>$t_{\text{jitter}}$</td>
<td>none</td>
<td>$&gt; 96$ fs [12]</td>
<td>none</td>
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<table>
<thead>
<tr>
<th>Requirements</th>
<th>Requirements</th>
<th>Requirements</th>
</tr>
</thead>
<tbody>
<tr>
<td>• Photoinjection laser</td>
<td>• Photoinjection laser</td>
<td>• Photoinjection laser</td>
</tr>
<tr>
<td>• HV power supply</td>
<td>• Megawatt klystron</td>
<td>• Single-pass amplifier (if more power needed)</td>
</tr>
<tr>
<td>• HV feedthrough</td>
<td>• External synchronization</td>
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</tr>
</tbody>
</table>

electron source, that may ultimately lead to lower emittance electron bunches [22], has propelled the development of photonic (IR- or THz-driven) linear accelerators (linacs) with promising results [91, 9, 82, 88]. However, the potential advantages of photonic linacs have not extended to photonic guns, the initial acceleration stage that is quintessential to determining the final electron beam quality and can benefit most from higher accelerating fields. The difficulty, which lies primarily in phase matching the electromagnetic wave with nonrelativistic electrons, is greater for short IR wavelengths [123] than for THz radiation.

The advent of efficient THz generation techniques [51, 52, 6] with routine generation of GV/m fields [113, 102] from few-mJ lasers has made THz based acceleration a realistic possibility and has motivated studies for using THz radiation in improving electron energy and beam properties in high-brightness linacs [41, 117] and proton post-accelerators [88].

A laser-based, THz-driven electron gun has the potential to achieve GeV/m accelerating gradients for common accelerator materials [107] with intrinsic timing stability. Such a gun would be ultracompact and easy-to-implement because it would
be pumped by an amplified arm of the already-present photoinjector laser, without the need for any high power electronics. The third column of Table 5.1 summarizes these potential advantages.

Thus, leveraging the streaking experiments from the previous chapter to exploit the GV/m fields possible with optically-generated THz sources, here we implement the first THz gun. Designing it with simple geometries and flexible machining requirements in mind, we integrate the THz gun in a practical, compact machine that is powered by a 1 kHz, few-mJ laser and operates without external synchronization. Our first results demonstrate high field (350 MV/m) acceleration up to 0.8 keV, as well as percent-level energy spread in sub-keV, multi-10 fC bunches. These results, which are already suitable for time-resolved LEED experiments, confirm the performance of a THz-driven gun technology that is scalable to relativistic energies [23].
CHAPTER 5. THZ-DRIVEN, ALL-OPTICAL ELECTRON GUN

Figure 5-2: THz gun concept. (a) Photograph of the THz gun. (b) A single-cycle THz pulse, generated via optical rectification in lithium niobate (LN), is coupled into the THz gun, which takes the form of a parallel-plate waveguide (PPWG) for field confinement. A UV-back-illuminated photocathode emits an electron bunch, which is accelerated by the THz field. The bunch exits through a slit on the top plate and a retarding field analyzer (RFA) measures its energy spectrum. (c) Cross-section of the gun, showing the UV-photoemitted electrons accelerated by the THz field and exiting through the slit.

5.3 Methods

5.3.1 Summary of experimental setup

The THz gun (Figure 5-2(a)-(c)), takes the form of a copper parallel-plate waveguide (PPWG) with a subwavelength spacing of 75 µm. We exploit this structure’s TEM mode for unchirped, uniform enhancement of the THz field [54]. A free-space vertically-polarized THz beam is coupled into the PPWG by a taper. EM simulations (Figure 5-3(a)) [23] were utilized to optimize the taper and calculate the coupling efficiency. Inside, a copper film photocathode serves as the bottom plate of the PPWG. There, a UV pulse back-illuminates the film, producing electrons inside the PPWG by photoemission. Concurrently, the THz field accelerates the electrons vertically across the PPWG. The electrons exit the gun through a slit on the top plate (anode)
and are spectrally characterized by a retarding field analyzer (RFA) or counted by a Faraday cup. Both UV and THz pulses are generated from the same 1030 nm pump laser, ensuring absolute timing synchronization.

The THz pulse, generated in lithium niobate by the tilted pulse front (TPF) method, is focused into the gun with a maximum impinging energy of 35.7 $\mu$J. EO sampling at PPWG-center (location of the center of the gun with the gun removed) and PPWG-thru (focus of an image-relay following propagation through the PPWG) reveals single-cycle durations of $\tau_{THz} = 1.2$ ps (Figure 5-3(d)), confirming that the PPWG induces minimal dispersion. Figure 5-3(b) shows the THz beam profile at the free-space focus. Inside the waveguide, the horizontal (x) beam profile remains unaltered while the vertical (z) profile is distributed uniformly across the 75 $\mu$m spacing. Based on this fact, Figure 5-3(c) shows the calculated beam profile inside the gun. Taking into account the energy, waveform, beam profile, and coupling efficiency, the THz pulse has a calculated peak field of 153 MV/m in free-space and 350 MV/m in the gun.

The UV emitter pulse, generated by frequency-quadrupling of the pump laser, has a wavelength of 258 nm, an estimated duration of $\tau_{uv} = 275$ fs (roughly 12.5% of the THz period), and a focused beamwaist of 60 $\mu$m (x) and 20 $\mu$m (y) on the photocathode.

5.3.2 THz source

For the pump laser, we used a 1 kHz, 1030 nm Yb:KYW regenerative amplifier [13] seeded with a 42.5 MHz Yb:KYW oscillator from Amplitude Systems. The pulses are compressed to the transform limit of 550 fs (sech$^2$) FWHM with shot-to-shot energy fluctuations of 0.5%. The available 4.2 mJ compressed pulses are split: 99% for THz generation and 1% for UV generation. For THz generation, the beam impinging onto the lithium niobate crystal has an energy of 3.4 mJ and a $1/e^2$ diameter of 2.0 mm (sagittal) and 3.4 mm (tangential).

To generate the THz pulses, we employed the TPF pumping technique in a 5.6% MgO-doped congruent lithium niobate (LN) crystal [42] cooled to 100 K in a liquid
Figure 5-3: Design and characterization of the gun. (a) Several snapshots of the single-cycle THz wave coupling into the PPWG, based on EM simulations. (b) THz beam intensity at the free-space focus (measured). (c) THz beam intensity in the gun (calculated). The colorbars of the two beam profiles show a 5.3x intensity enhancement in the PPWG. (d) Temporal profiles measured via EO sampling of the THz electric field at PPWG-center and PPWG-thru (scaled). (See main text for definitions). (Inset) Power spectrum of the THz beam.
nitrogen cryostat. The IR pump beam is diffracted off a 1500 l/mm grating to acquire a TPF [42], which is then subsequently imaged—in the tangential direction—onto the LN using a 150 mm cylindrical lens. Another cylindrical lens with a sagittal focal length of 100 mm was used to shape the impinging pump beam for highest efficiency. The extracted optical-to-THz energy conversion efficiency was near 1.0% with 35.7 µJ of THz energy. We used a Gentec SDX-1152 calibrated pyroelectric THz joulemeter to measure the THz energy. Shot-to-shot energy fluctuation was 2%. Concurrently, a thermal power meter (Ophir Optronics) measured 18 mW at 1 kHz. A Spiricon Pyrocam IV camera was used to image the THz beam.

For temporal characterization, we employed an oscillator-based EO sampling setup since the pulses from the amplifier were too long to effectively probe the THz waveform. Oscillator probe pulses were overlapped with THz pulses on a 200 µm, 110-cut ZnTe crystal. The probe pulses sample the THz-induced birefringence as a function of delay and are subsequently interrogated by a quarter-wave plate, polarizer, and photodiode combination, as typical of EO sampling setups [118]. Because of the much higher repetition rate of the oscillator pulse train, a boxcar averager (SRS SR250) was used to electronically gate out the pulse that overlapped with the THz. The EO crystal mount was custom fabricated such that the crystal is in the same position as the center of the gun for the PPWG-center measurement in Figure 2(d). For the PPWG-thru measurement, the gun was placed in its operating position and the transmitted THz beam was image-relayed via two additional parabolic mirrors onto a second focus, where the EO crystal was then placed. The difference in carrier envelope phase offset of roughly $\pi/2$ between the PPWG-center and PPWG-thru measurements is due to the Guoy phase shift in the waveguide.

**Characterization**

In Figure 5-12, the THz energy was varied by changing the IR pump energy and measured using the pyroelectric detector. Since the acceleration process depends on the spatiotemporal properties of the THz beam, it is important to verify that there are no significant distortions in the THz temporal and spatial profiles as the
Figure 5-4: THz spatiotemporal properties as a function of energy. (a) EO sampling waveforms for various THz energies. (b) The relative field strength as a function of THz energy at Peak 1 (black dots) and Peak 2 (gray dots) of the waveform, as labelled in (a). The measured peak field strengths scale as the square root of the THz energy according to the fits (dashed lines). The error bar radius is determined by calculating the RMS noise of the EOS signal in the absence of THz. (c) The normalized horizontal and (d) vertical lineout THz beam profiles at the free-space focus for various THz energies. Profiles are Gaussian with minimal variations in shape as a function of THz energy. (e) The horizontal (gray squares) and vertical (black circles) 1/e² beam diameters as a function of THz energy, shown here to all reside near their respective collective mean value (dotted lines). (Inset) 2D beam profiles at several sample THz energies, showing minimal variation.
energy is changed. It is also important to verify that the THz field strength scales proportionally as the square root of the THz energy. We first measure the temporal profile via EO sampling for various THz energies in Figure 5-4(a). Aside from scaling in field strength, the shape of the temporal profile has little variation as a function of THz energy, with the carrier-envelope phase and pulse duration remaining roughly constant. In Figure 5-4(b), we sample the THz field as a function of THz energy at two peaks of the waveform as labelled in Figure 5-4(a): Peak 1 (black dots) and Peak 2 (gray dots). The field strengths at these two peaks correspond to the maximum accelerating fields experienced by the electron in the gun. The data fit well to a square root function (dashed lines), thus verifying that the peak accelerating field scales with the square root of the THz energy.

Next we measure the spatial profile of the THz beam at the free-space focus using a THz camera for various energies in Figure 5-4(c)-(e). The normalized horizontal and vertical lineout profiles are overlapped in Figure 5-4(c)-(d), revealing negligible variation in their Gaussian-like shapes. Further, the $1/e^2$ beam diameters are plotted in Figure 5-4(e) as a function of THz energy for the vertical (black circles) and horizontal (gray squares) profiles. The diameters vary by an average of only 3.7% (horizontal) and 2.9% (vertical) with respect to the mean (dotted lines) over the range of THz energies. The 2D beam profiles for several THz energies are shown in the inset, revealing negligible variation.

### 5.3.3 UV source

The UV phototrigger pulse was obtained by frequency-quadrupling the fundamental 1030 nm pump as shown in Figure 5-5. The first second-harmonic generation (SHG) stage consisted of a 0.5 mm thick type I BBO crystal with $\phi=23.7^\circ$, generating approximately 3 $\mu$J of 515 nm pulses. Two dichroic mirrors reflecting 515 nm and transmitting 1030 nm were used to remove the strong pump beam. The second SHG stage consisted of 0.5 mm thick type I BBO crystal cut for $\phi=44.6^\circ$ but tilted to $\sim50^\circ$, generating 600 nJ of 258 nm pulses. The conversion efficiency from fundamental to UV was about 7%. A CaF$_2$ prism was used to spatially separate the various
wavelengths. The prism-induced dispersion over the subsequent 0.5 m propagation was determined through calculations to cause negligible increase of the pulse duration. The UV energy impinging the copper photocathode was 270 nJ. Both nonlinear conversions were in the unsaturated regime and the phase-matching bandwidths of the two BBO crystals are broader than the spectral bandwidths of both the 1030 nm and 515 nm pulses. Therefore, we estimate of the UV pulse duration as roughly half that of the fundamental. The focused UV beamwaists on the photocathode were 20 µm (x) and 60 µm (y).

### 5.3.4 Timing jitter

Low timing jitter is one of the key advantages of the laser-based THz-driven electron gun over conventional RF guns, which have timing jitter typically on the order of hundreds of femtoseconds. Only via additional advanced electronic synchronization schemes can the jitter of RF guns be lowered. Here we provide measurements of the timing jitter of our all-optical gun using EO sampling. Since our original EO sampling measurements were performed with oscillator pulses (see Section 5.3.2), they cannot provide a comparable estimate of the timing jitter between the UV—which was generated by the amplified IR pump—and the THz pulse. Further, the amplified
IR pulses were too long (550 fs) for adequate EO sampling. Therefore, we present here an estimate of the timing jitter using EO sampling data by 50 fs probe pulses on a comparable setup pumped by a 4 mJ Ti:Sapphire amplified laser system (Coherent Legend).

Figure 5-6(a) shows the THz waveform. If the probe delay is positioned to measure the THz field at the zero-crossing over many shots, any signal fluctuation that occurs should be primarily due to timing jitter of the THz and probe pulse, rather than to amplitude noise. On the other hand, if the probe is positioned to measure the THz field at the first crest, any signal fluctuation that occurs should be primarily due to amplitude noise.

Figure 5-6(b) shows the EO signal over 2500 shots with the probe delay positioned at the THz zero-crossing. After notch-filtering out two environmental noise sources (electrical interference at 60/120 Hz and acoustic noise from the air handling system at 110/220 Hz), the measured EO signal RMS fluctuation was 0.0023 rad. Dividing this value by the slope of the THz field at the zero-crossing (0.0048 rad/ps) gives a RMS timing jitter of 4.8 fs.

To further verify our timing jitter measurement, the probe was positioned at a delay that was far from temporal overlap with the THz pulse. The RMS fluctuation was 0.0011 rad, originating only from the shot-to-shot intensity variations of the laser. This measurement indicates that zero-crossing fluctuation measurement (0.0023 rad) was above this intensity noise limit. Furthermore, the probe was positioned at the first crest of the THz waveform (Figure 5-6(a)) and the measured RMS fluctuation was 0.0011, indicating that the THz amplitude noise was equal or lower than the limit set by laser intensity variation, and that the zero-crossing fluctuation was dominated by timing jitter.

5.4 THz gun design

A variety of THz gun structures were proposed, including rectangular waveguides, pillbox structures, and multi-cell standing wave structures. For the first demostr-
Figure 5-6: Timing jitter measurement using EO sampling. (a) THz waveform, with annotations indicating the first crest and zero-crossing of the field. (b) EO signal measured over 2500 shots with the probe parked at the THz zero-crossing. The original signal (blue) is notch-filtered (red) at the 60/120 Hz electrical interference lines and at the 110/220 Hz acoustic noise lines. The filtered RMS fluctuation divided by the slope at the zero-crossing gives a timing jitter (between THz and probe pulses) of 4.8 fs. (Inset) Power spectrum of the signal.
tion, we opted to use a simple parallel plate waveguide (PPWG) structure because of its simplicity and compatibility with broadband THz pulses. The PPWG, having a subwavelength spacing of \( d = 75 \, \mu\text{m} \), guides only the TEM (TM0) mode. This mode has zero cutoff frequency and a propagation constant given by \( k_z = \omega/c \) for all frequencies regardless of the spacing between the plates [65], and we leverage this property for broadband, unchirped enhancement of the THz field [54].

Two parallel plates, fashioned with 18\(^\circ\) tapers, were fabricated separately and afterwards sandwiched together with high-precision Kapton shims in-between to set the spacing and enforce parallellicity. We ultimately operated with a shim thickness of \( 75 \pm 15 \, \mu\text{m} \) after optimization (see Section 5.4). Plastic (PEEK) screws were used to clamp the plates together to maintain electrical isolation between the plates. EM simulations were performed in HFSS to obtain the optimal taper angle for efficient coupling. Fabrication of the THz waveguide was performed in-house using conventional machining tools. A flatness tolerance of 5\( \mu\text{m} \) over a 1 in\(^2\) area was specified for the parallel plate sections. A 9V reverse bias was applied across the plates to help with electron extraction.

A 25 nm copper film coated on a UV-grade quartz substrate was used as the photocathode. The coating of the photocathode was performed in-house by evaporative physical vapor deposition. A chromium adhesion layer of a few nm was first deposited onto the substrate. In addition to functioning as a photocathode, the copper film functions as one of the PPWG plates. To minimize interface losses, the surface of the photocathode is placed flush with the surface of the parallel-plate structure to a tolerance of a few microns using an optical-flat mirror surface. To minimize THz diffraction losses through the thin film, the film is thickened to 125 nm (equal to the copper skin depth at 0.5 THz) along the THz propagation path from the PPWG input until \( \sim 0.25 \, \text{mm} \) before the 25 nm thick photoemission region. To ensure electrical connectivity between the photocathode and the PPWG, the copper film extended around to the edges of the quartz substrate and the edges were in contact with the PPWG.

The exit slit anode was cut out of a slab of 100 \( \mu\text{m} \) polished stainless steel shim
CHAPTER 5. THZ-DRIVEN, ALL-OPTICAL ELECTRON GUN

Figure 5-7: Cross-section of the center of the gun shows that the field amplitude inside the gun is highly uniform and unperturbed by the slit aperture.

Figure 5-8: Measurement of the PPWG power transmission, $T$ (blue), and the corresponding power coupling efficiency into the gun, $\eta_{\text{gun}}$ (light blue), for various PPWG spacings. Based on these values, the normalized field inside the gun, $E_{\text{ppwg}}$ (green), can be determined. A spacing of 75 $\mu$m optimizes the normalized field strength.

stock. The precise slit width of 20 $\mu$m over a 2 mm length was achieved by picosecond laser micromachining. An optical microscope was used to verify the dimensions to within a tolerance of 2 $\mu$m. EM simulations in Figure 5-7 confirm that, with a width of 20 $\mu$m ($\lambda_{\text{THz}}/33$), the slit causes minimal distortion to the THz field distribution.

Characterization

A THz network analyzer setup was used to characterize the power transmission, $T$, through the PPWG for various spacings, as shown in Figure 5-8. Our vector network analyzer (VNA) consisted of an Agilent E8363B and millimeter wave extender V03VNA2-T/R with 70 dB of dynamic range at 0.220-0.325 THz. The transmitter and receiver were connected to corrugated horns designed for coupling the VNA waveguide mode to a free-space Gaussian mode with a waist of 6 mm. We placed
two THz polyethylene lenses with focal lengths of 25 mm a distance of $2f$ apart between the transmitter and receiver and set the background level. Given the waist of 6 mm (diameter of 12 mm) and focal length of 25 mm, the f-number is about 2, which is well-matched to the optimal f-number of our PPWGs taper section. For the VNA measurements we replaced the photocathode with a polished aluminium block to eliminate diffraction losses in the PPWG. The PPWG was placed in the center between the two THz lenses and the PPWG transmission was characterized. The PPWG spacing was varied by changing the thickness of the Kapton shims between the two plates.

In Figure 5-8, EM and analytical calculations (see Section ) helped to determine the power coupling efficiency into the gun, $\eta_{\text{gun}}$ (light blue), as a function of $T$ (blue). Although $\eta_{\text{gun}}$ increases with spacing $d$, the field strength inside the gun, $E_{\text{THz}}$, is a trade-off between coupling efficiency on one hand, and field confinement on the other, as expressed by $E_{\text{THz}} \approx \sqrt{\eta_{\text{gun}}/d}$. It is desirable to have the highest field strength possible inside the gun. By plotting the normalized field strength, $E_{\text{THz}}$ (green), as a function of spacing, we found the optimal value at a spacing of 75 $\mu$m and a coupling efficiency of 0.3.

**Calculation of coupling efficiency, $\eta_{\text{gun}}$**

Here, we show how the THz coupling efficiency into the gun, $\eta_{\text{gun}}$, can be calculated from the measured power transmission data, $T$, shown in Figure 5-8 (blue line).

First, we make the assumption that the out-coupled free-space mode, denoted by $E_{\text{ppwg}}(x,y,z)$ and shown in Figure 5-9(b), varies minimally for different PPWG spacings, $d$. This has been validated by EM simulations for values of $d$ within our region of interest: $0 < d < 200 \, \mu$m. This mode is the beam which couples most efficiently from free-space into the TEM mode of the PPWG, with a coupling efficiency denoted by $\eta$. Our THz beam in-coupled into the PPWG can be approximated as a fundamental Gaussian beam, denoted by $E_{\text{gaussian}}(x,y,z)$ and also shown in Figure 5-9(b). The amount power in the $E_{\text{ppwg}}(x,y,z)$ component of the $E_{\text{gaussian}}(x,y,z)$ mode, as a fraction of the total power, is denoted by $F$. Using EM simulations, we
determine $F$ by computing the overlap integral over a chosen plane normal to $y$:

$$F = \frac{|\int E_{gaussian}^* E_{ppwg} dx dz|^2}{\int |E_{gaussian}|^2 dx dz \int |E_{ppwg}|^2 dx dz} \quad (5.2)$$

Note the integrand is scalar because the modes have only one and the same polarization. We obtained a result of $F = 0.8$.

Second, we assume that the large majority of transmission losses come from wall ohmic losses inside the PPWG (region $2 \leftrightarrow 3$ in Figure 5-9(a)) and from reflections at the interfaces between the PPWG and taper sections (regions $1 \leftrightarrow 2$ or $3 \leftrightarrow 4$ in Figure 5-9(a)). We proceed to express the power transmission and reflection at the interfaces as follows.

**Transmission** $1 \rightarrow 2$: $F \eta$

**Transmission** $3 \rightarrow 4$: $\eta$

**Reflection** $3 \rightarrow 4$: $1 - \eta$

A reflected wave at the $3 \rightarrow 4$ interface propagates backward toward the $2 \rightarrow 1$ interface. There it experiences a second reflection.

**Reflection** $2 \rightarrow 1$: $1 - \eta$

Also, the propagation along $2 \leftrightarrow 3$ induces ohmic losses. The propagation efficiency of one pass is denoted by

**Propagation** $2 \leftrightarrow 3$: $\beta$

Using EM simulations with finite conductivity surfaces, we determined $\beta = 0.83$. We can now calculate the total transmission through the structure:

**Transmission** $1 \rightarrow 4$: $T = F \eta [\eta + \beta^2(1 - \eta)^2 \eta + \beta^4(1 - \eta)^4 \eta + \cdots]$

After some algebraic simplification, we obtain

$$T = \frac{F \eta^2}{1 - \beta^2(1 - \eta)^2} \quad (5.3)$$

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Figure 5-9: Calculation of THz coupling efficiency into the PPWG gun. (a) Schematic of the PPWG gun denoting the interfaces at which reflections occur. (b) EM simulations showing the in-coupled free-space Gaussian mode ($E_{\text{gaussian}}$) and the out-coupled free-space mode of the TEM waveguide ($E_{\text{ppwg}}$).
This equation gives the power transmission through the waveguide, $T$, as a function of the TEM mode coupling efficiency, $\eta$. Since we have measurements of $T$ and wish to know $\eta$, we reverse the equation:

$$\eta = \frac{\beta^2 T + \sqrt{[F - \beta^2(F + T)]T}}{F + \beta^2 T}$$  (5.4)

Finally, the coupling efficiency of our THz beam from free-space into the center of the gun, $\eta_{\text{gun}}$, as a function of $\eta$ is simply

$$\eta_{\text{gun}} = F \sqrt{\beta \eta}$$  (5.5)

5.4.1 Electron detection

Following its exit from the gun, the electron bunch drifts into a retarding field analyzer (RFA) [11], consisting of a channel electron multiplier (CEM) (Photonis, Inc.) collector and two static, uniform field regions formed by two biased mesh electrodes. The first region (between the gun and first electrode) boosts the electron energy by 300 eV to enhance the detection efficiency of the CEM. The second region (between first and second electrodes) acts as a highpass filter for the electron energy by retarding the electron trajectory using a variable bias $-V_{\text{bias}}$. Electrons having energy less than $eV_{\text{bias}}$ are repelled by the electrode while those with more energy pass through, being subsequently detected by the CEM. Each spectrum was collected by taking the derivative of the measured current with respect to $V_{\text{bias}}$. The intrinsic energy resolution of the analyzer is about 2 eV. With post-process smoothing, the effective resolution is about 16 eV.

The meshes were TEM grids (Ted Pella, Inc.) with a thickness of 13 $\mu$m and a pitch of 12.5 $\mu$m. Each mesh had a transmission of 36%, so the total bunch charge was determined from dividing the detected charge by $0.36^2 = 0.13$. Each mesh was placed on 100 $\mu$m thick stainless steel soldering plates and sandwiched by 500 $\mu$m PEEK shims which enforce their spacing and parallelicity as well as providing electrical isolation. The soldering plates contained “fingers” on which high voltage
biasing wires were soldered. The RFA was placed 1.5 mm from the exit anode of the electron gun, measured by the distance between their nearest planes.

Absolute charge measurements were obtained by rewiring the grounded input terminal of the CEM to a Keithley 6514 picoammeter and turning off the CEM bias. In this configuration the CEM essentially acts as a Faraday cup. Secondary electron emission on the CEM is not taken into account, but it would only increase the total charge count if it were. The picoammeter had a RMS noise level of 300 fA.

5.4.2 Particle tracking simulations

3D particle tracking simulations incorporating space charge were used to model the electron bunch evolution in the presence of the THz field. The emitted electron bunch had a Gaussian spatial profile with beam waists of 20 $\mu$m (x) and 60 $\mu$m (y) and a Gaussian temporal profile with FWHM of 275 fs. The initial kinetic energy was 0.18 eV (equal to the excess energy) with a uniform momentum distribution over a half-sphere [18]. 5000 macroparticles were used to represent a total bunch charge of 32 fC, corresponding to a charge of -40e per macroparticle. The trajectories were modeled by integrating the kinematic equations for every particle $i$ using a 4th order Runge-Kutta solver:

\[
\begin{align*}
    m\frac{dv_i}{dt} &= F_{field} + F_{bias} + \sum_j (F_{image,ij} + F_{coulomb,ij}) \\
    \frac{dr_i}{dt} &= v_i
\end{align*}
\]

Here, $m$ is the relativistic mass, $F_{field}$ is the electric force due to the THz pulse, $F_{bias}$ is the electric force due to the 9V reverse DC bias, $F_{image,ij}$ is the force on the $i$th particle due to the $j$th image particle, and $F_{coulomb,ij}$ is the particle-particle Coulomb force (space charge). The THz beam was modeled as a plane wave with a Gaussian distribution in amplitude in the x direction. The THz waveform in $F_{field}$ was directly imported from the EO sampling trace.
5.5 Results and discussion

5.5.1 Spatial overlap and emission profile

We found that when the photoemitting UV beam is not spatially coinciding with the slit, no electrons are detected at the outside of the gun, since they are blocked by the opposite plate. When the UV beam coincides with the slit in the y direction and is moved along the x direction (see Figure 5-2 for coordinate system reference), we observe a 1D emission profile as shown in Figure 5-10. The profile has a 2 mm width corresponding to the 2 mm slit length. It also has a peak at about $x_{UV}=0.6$ mm, due to maximum spatial overlap with the peak of the THz beam.

5.5.2 Temporal overlap and energy spectrogram

An electron’s final momentum gain, $p_e$, depends on emission time and can be expressed as $p_e = q \int_{t_{emit}}^{t_{exit}} E_{THz}(t) dt$, where $t_{emit}$ is the emission time and $t_{exit}$ is the time the electron exits the PPWG. In cases where $t_{exit} \gg \tau_{THz}$ (the electron exits long after passage of the THz pulse), $p_e$ can be simplified to $p_e = q \int_{t_{emit}}^{\infty} E_{THz}(t) dt = qA(t_{emit})$, where $A(t_{emit})$ is the THz vector potential at the time of emission. To determine the optimum emission time for acceleration, we record the electron energy gain ($U_e$) spectra and bunch charge ($Q$) versus delay in Figure 5-11(a)-(b). The UV emitter can precede (<-2 ps), overlap (-2 to 2 ps), or succeed (>2 ps) the THz pulse.
the overlap region (-2 to 2 ps), $U_e$ maps out the phase and amplitude of $A_{THz}(t_{emit})$, similar to THz streaking in gases [26]. One exception is that between -0.25 and 0.4 ps, emission occurs in the positive half-cycle of the THz field (opposing Lorentz force), causing a suppression of charge and energy gain. Two delays are selected to be the operating points of the gun. The first delay, $\tau_1 = -2$ ps, produced the highest peak acceleration while the second delay, $\tau_2 = 0.9$ ps, produced the most monoenergetic spectra. The total bunch charge was 40 fC at $\tau_1$ and 32 fC at $\tau_2$.

When the photoemission precedes the THz pulse (<-2 ps), a large energy spread centered about $\sim0.45$ keV is observed. The origin of these broadened spectra, enduring for long decay times, is attributed to multiple complex mechanisms encompassing thermal [47] or time-of-flight effects. Further discussion is provided in Section 5.5.6. When the emission succeeds the THz pulse (>2 ps), there is no net acceleration from that pulse. The constituency of electrons slightly elevated to 50 eV is attributed to the aforementioned decay effects probed by a back-reflected THz pulse arriving at 18 ps.
CHAPTER 5. THZ-DRIVEN, ALL-OPTICAL ELECTRON GUN

5.5.3 Electron energy spectra

In Figure 5-11(c)-(d), we take a closer look at the energy spectra from the two operating points, $\tau_1$ and $\tau_2$, for three different THz energies, $W_{THz}$. Each spectrum exhibits a unimodal distribution with an average energy gain increasing with $W_{THz}$. Except for the $W_{THz} = 35.7 \mu J$ spectrum at $\tau_1$, the spectral shapes are asymmetric with a pedestal toward lower energies and a maximum yield toward higher energies, followed by a sharp cutoff. The high yield near the cutoff indicates that most electrons are emitted at the optimal THz phase and concurrently experience the same acceleration. The pedestal can be attributed to electrons emitted away from the optimal phase, resulting in a lower energy gain. Such dependence of energy gain on emission phase is also evident in RF guns [62].

5.5.4 THz energy scaling

We continue investigations at $\tau_1$ and $\tau_2$ by plotting $U_e$ versus $W_{THz}$ on a spectrogram, as shown in Figure 4(a)-(b). At both delays, $U_e$ scales mostly linearly with $W_{THz}$ or, equivalently, with $E_{THz}^2$. This scaling law can be explained by $U_e = p_e^2/2m \approx E_{THz}^2$, which is valid when $t_{exit} \gg \tau_{THz}$. Alternatively, if $t_{exit} \ll \tau_{THz}$, the energy gain would be dominated by $U_e = q \int_{z_{emit}}^{z_{exit}} E_{THz}(z)dz$, leading to a $U_e \approx E_{THz}$ scaling law, as is typical in RF guns [38] and would be the case in this study for larger field, reduced PPWG spacing, or relativistic electrons.

At $\tau_1$, increasing the THz energy results in an increase of absolute energy spread (Figure 5-12(a)). Consequently, the relative energy spread, $\sigma_U$, remains roughly constant at around 20-30% (Figure 5-12(c)). The bunch charge increases monotonically with THz energy (Figure 5-12(e)). We obtain a peak energy gain of 0.8 keV at $W_{THz} = 35.7 \mu J$ (Figure 5-12(a)).

At $\tau_2$, the absolute energy spread remains constant with THz energy (Figure 5-12(b)). Correspondingly, the relative energy spread monotonically decreases with THz energy, to a minimum of 5.8% centered near 0.4 keV (Figure 5-12(d)). The pedestal regions are neglected in the energy spread calculations, since over time those
Figure 5-12: THz scaling at $\tau_1$ and $\tau_2$; delay positions defined earlier in Figure 5-11(a).
(a)-(b) Energy gain plotted on a spectrogram to highlight its scaling as a function of accelerating THz energy or THz field. Error bar radius is equal to the absolute RMS energy spread. (c)-(d) Relative RMS energy spread, $\sigma_U$, of the accelerated bunch. (e)-(f) Total detected bunch charge exiting the gun, $Q$. Error bar radius is equal to the RMS instrument noise.
electrons separate from the main bunch. Half of this spread comes from THz shot-to-shot fluctuations (2%), while another large contribution comes from the spread in electron emission time: $\Delta t_{\text{emit}} = \tau_{uv} = 275 \text{ fs} = \frac{T_{THz}}{8}$. By stabilizing the laser and shortening $\tau_{uv}$ via an OPA [127], the energy spread can be further reduced. In Figure 5-12(f), the bunch charge increases with THz energy below $7 \mu J$, indicating that the emission is space-charge-limited [22]. Above $7 \mu J$, the bunch charge plateaus, indicating that the THz field overcomes the space charge force and extracts all the emitted electrons.

### 5.5.5 Numerical verification and insights

In Figure 5-13(b), we show the calculated single-electron energy gain versus delay, utilizing the shape of the measured THz waveform with a fitted field strength (Figure 5-13(a)). We compare it with the measured peak energy gain from Figure 3(a). Since the experimentally-measured peak energy gain represents the gain of the electron emitted at the optimal delay and spatial position, comparing it with our analytical expression for single-electron energy gain is justified. Several experimental features are represented in this simple analytical model: (1) suppression region around 0 ps, (2) relative energy gain levels and (3) delay between the two peaks. This model provides an alternate method for quantifying the THz field strength inside the gun. Our fitted peak field was 480 MV/m.

To better understand the bunch dynamics under the influence of self-fields and the THz field, multi-electron particle tracking simulation (Supplementary Materials) results in Figure 5-13(d) show the evolution of the energy spectrum of the 32 fC bunch emitted at $\tau_2$ as it propagates along $z$. Immediately following emission, the bunch experiences a strong accelerating field, growing in energy to 350 eV over the first 3 $\mu m$. During the time that the THz pulse interacts with the bunch (corresponding $z$ distance: 0 to 25 $\mu m$), the energy undergoes four acceleration/deceleration cycles, caused by the four oscillation cycles in the THz field following $\tau_2$. The THz pulse is passed by the time the bunch reaches 25 $\mu m$, verifying that $t_{exit} \gg \tau_{THz}$, and the bunch drifts to the exit while continuing to experience energy spreading due to
Figure 5-13: Numerical analysis of THz gun. (a) THz electric field measured by EO sampling with a fitted field strength. (b) The single-electron energy gain, calculated analytically, is overlaid with the peak energy gain obtained from experiment in Figure 3(a). (c) Simulated energy spectrum (blue line) of the bunch at the gun exit for emission at $\tau_2$, showing excellent agreement with experiment (black line). (Inset) Temporal profile of electron bunch at the gun exit, showing a FWHM pulse duration of 321 fs, elongated by space charge. (d) Simulated evolution of the energy spectrum along $z$. The THz pulse is passed by the time the electrons reach 25 $\mu$m.
space charge forces. At the gun exit \((z_{\text{exit}} = 75 \, \mu\text{m})\), the simulated energy spectrum has excellent overlap with the experimental spectrum (Figure 5-13(c)). The sharp cutoff, pedestal height, pedestal length, and central lobe width are all reproduced flawlessly by the model. The simulated temporal profile at the gun exit (Figure 5-13(c) inset) exhibits a pulse duration of 321 fs, longer than the initial 275 fs due to space charge. All the numerical analyses incorporated space charge, imitated the experimental conditions, and used the THz field profile shown in Figure 5-13(a).

In conclusion, we demonstrated high field \((>300 \, \text{MV/m})\), quasimonoeenergetic (few percent spread) THz acceleration of multi-10 fC electron bunches to sub-keV energies in an ultracompact, robust device. No degradation in performance was observed over 1 billion shots. While the operating pressure was 40 \(\mu\text{Torr}\), no change in performance was observable up to 10 mTorr. This first result of a jitter-free, all-optical THz gun, powered by a few-mJ laser, performs in accordance with underlying simulations and is encouraging for future developments. In its current state, it can be used for time-resolved LEED [36]. Further improvements on the gun structure and THz field promise relativistic electrons [23].

**Energy spread and emittance**

This particle tracking simulation tool can be used to analyze the effect of UV pulse duration, \(\tau_{\text{UV}}\), and space charge on the final energy spread. In Figure 5-14(a), the energy spectrum at delay \(\tau_2\) is plotted for three different \(\tau_{\text{UV}}\). Narrower energy spectra are achieved by reducing \(\tau_{\text{UV}}\). The reason is because smaller \(\tau_{\text{UV}}\) leads to lower variation in the phase of the THz cycle over which electrons are emitted. Consequently, there is lower variation in the final energy after acceleration of the electrons, hence lower energy spread. We observe that the spectrum at \(\tau_{\text{UV}}=275\) fs has an energy spread of 15.3\% (5.9\% without pedestal) and the spectrum at \(\tau_{\text{UV}} = 91\) fs has an energy spread of 5.9\% (3.6\% without pedestal).

In Figure 5-14(b), the same energy spectrum at \(\tau_{\text{UV}} = 275\) fs is plotted alongside an all-else-equal simulation with space charge effects turned off. With space charge turned off, the energy spread is reduced to 12.9\% (2.9\% without pedestal).
Figure 5-14: Numerical simulation of the effect of (a) UV pulse duration and (b) space charge on the energy gain spectrum and energy spread for emission at delay $\tau_2$.

Figure 5-15: Numerical simulation of the evolution of x and y bunch emittance ($\epsilon_x = \sqrt{\langle x^2 \rangle \langle x'^2 \rangle - \langle xx' \rangle^2}$) as a function of time for emission at delay $\tau_2$. 
Further, the simulation tool can be used to estimate the emittance of the generated e-beam. In Figure 5-15, the evolution of the bunch emittance in both x and y are plotted as a function of time. From 0 to 4 ps, the rapidly-varying THz field strongly modulates the emittance. After its passage, the bunch drifts freely and the emittance grows slowly under the influence of space charge. The emittance values at the output of the gun (approx. 8.6 ps) are 0.8 mm·mrad in x and 0.16 mm·mrad in y. These values are comparable to the emittances obtained with 30 keV [110] and 100 keV [111] DC guns with 10-100 fC bunch charge. Lower emittance values are expected with stronger fields and more acceleration. Our future version of the THz gun, as described theoretically in [23] achieves 2 MeV electron bunches normalized emittances of 0.02 (x) and 0.06 (y) mm·mrad using 0.5 GV/m fields, opening up promising applications for such THz guns.

5.5.6 Long delay scan

In order to have a fuller understanding of the electron dynamics induced inside the gun by the THz and UV pulses, we acquire a spectrogram over a wide range of delays in Figure 5-16(a). Between -2 and 2 ps, the electron spectra change rapidly with respect to delay due to the temporal overlap with the main THz pulse. Here, the spectra are narrowband and the momentum gain follows the vector potential of the THz field, as described in the main text.

When the UV pulse precedes the THz pulse (<= -2 ps), we observe broad, elevated electron spectra enduring over a long delay window to nearly 50 ps. A number of physical processes may contribute to this behavior. Detailed investigations will be the topic of a forthcoming article.

One possibility is thermally-assisted THz field emission, a process investigated in [47] and more generally in [27, 49]. As UV photons are absorbed, electrons are promoted in energy. The increase in kinetic energy causes an elevated electron temperature distribution (i.e., hot electrons) over a period of tens of fs. The hot electrons then collide with phonons to dissipate heat to the lattice via electron-phonon collisions, resulting in an elevated lattice temperature, or a smeared-out Fermi-Dirac
Figure 5-16: Delay scan. (a) Spectrogram showing the energy gain spectra over a wide range of delays. Broad electron spectra is exhibited for UV pulse delays of up to 50 ps preceding the THz pulse. The presence of weak, back-reflected THz pulses is evident at 18 ps. (Inset) This back-reflection occurs at the interface between the PPWG and the output taper, which is 2.75 mm away from the exit anode. (b) Normalized current as a function of delay, showing exponential decay behavior preceding the main (0 ps) and back-reflected (18 ps) THz pulses. Exponential curve fitting determines the decay time to be 16.7 ps.
distribution with a high energy tail. This elevated lattice temperature decays over a multi-ps time scale, as is known from experiments on similar thin films [49]. When the THz field impinges the surface during this time and lowers the Schottky barrier, electrons in the higher tail of the distribution have an increased tunnelling probability. Once emitted, the electrons are subject to free-space THz acceleration. Unlike UV photoemission, which creates an electron bunch of defined duration, the field-emitted electrons here can be emitted over a wide range of THz phase, so long as the field can sufficiently lower the Schottky barrier to enable tunnelling. Consequently, as the THz field increases, the spectra grow broader (Fig. 4(a)) and the emitted charge increases (Fig. 4(e)).

Figure 5-16(b) shows the normalized current as a function of delay. We observe exponential decay behavior preceding the main (<0 ps) and back-reflected (2 to 18 ps) THz pulses. The base level of current is about a factor of 0.48 times the maximum current. A decay time can be determined by fitting an exponential function (offset by the base level) to the normalized current, shown as a red/blue curve for the decay preceding the main/back-reflected THz pulse. We find that the exponential decay time is 16.7 ps for both curves, suggesting that there is an underlying thermal relaxation constant that is independent of THz field strength. This decay time is comparable to that measured from transient reflectivity measurements on similar thin metal films in [49], which was, e.g., ∼10 ps for a 20 nm Au film at 1 mJ/cm² pump fluence.

Another possibility for the decay behavior is time-of-flight effects. The bias voltage of 9 V in the 75 µm gap between the two plates of the PPWG implies that an electron released on the cathode takes 84 ps to reach the anode ($t_{exit} = 84$ ps). During this time, the bunch can be manipulated by the arriving THz pulse.

When the UV pulse succeeds the THz pulse (>2 ps), we observe a scaled-down (in energy) replica of the aforementioned decay effects. This can be attributed to the presence of a weak, back-reflected THz pulse arriving later at 18 ps (by definition, the envelope of the main THz pulse arrives at 0 ps). Physically, the back-reflection occurs in the gun at the interface between the end of the PPWG and the output.
taper, which is 2.75 mm of propagation away from the exit anode (see inset in Figure 5-16(a)). The roundtrip propagation of 5.5 mm matches well with the arrival of the back-reflected THz at 18 ps.
Chapter 6

Conclusion

6.1 Summary of work

This work consisted primarily of two developments: high energy THz pulse generation and THz-driven electron acceleration. Together, they form the front-end of an all-optical electron accelerator (and eventually, x-ray free electron laser vis-a-vis Chapter 1). Individually, these contributions are meaningful results in their own right.

Indeed, high energy THz pulses alone are an enabling tool for nonlinear spectroscopy [58] since, unlike optical frequencies which manipulate mainly valence electrons, THz radiation can control molecular rotations, lattice vibrations, and spin waves. They are also useful for remote sensing [73], polaritonic microscopy [104], tomographic imaging [55], electron metrology [60], and even research topics such as high harmonic generation in solids [101]. Meanwhile, high field, well-synchronized accelerators are needed primarily in ultrafast electron diffraction and x-ray free electron lasers for probing processes occurring on timescales of 1 fs and spatial dimensions of 1 nm.

To improve the source used so often in the aforementioned THz applications, several methods have been explored in this thesis. Whereas previous experiments of the tilted pulse front pumping scheme primarily used short (∼100 fs) pulses or very long ∼1 ps pulses, we tried pumping with pulse durations near the sweet spot (a balance between high peak intensity and long interaction length) of 500-700 fs to achieve one
of the highest conversion efficiencies to date. Furthermore we cryogenically cooled the lithium niobate crystal to reduce the THz absorption by a factor of >5 and enhance the efficiency by 2-3 fold. We also studied and demonstrated various types of antireflective coatings for the THz output facet, with the best result of obtaining 50% more efficiency through using a crystal quartz plate as a single layer coating.

In addition to improving the efficiency of the tilted pulse front technique, we also demonstrated recycling of the pump pulse in a second nonlinear crystal. Unlike the first stage, the second generation stage has lower efficiency due to spatial and spectral distortions in the pulse. These distortions were characterized experimentally to provide insight into the asymmetric nonlinear dynamics resulting from tilted pulse front phase matching.

This thesis also ventured into the still-nascent field of laser-driven particle acceleration. First we leveraged our high energy THz source to perform streaking experiments on a flat copper photocathode, the type of photocathode most suitable for producing short, highly charge bursts of electrons. From the numerical tools developed to model the streaking experiment, we designed a high field electron gun. To achieve optimal coupling of THz radiation into the gun, we conceived of a tapered parallel plate design with subwavelength dimensions, modeled it using electromagnetic software, and fabricated it using several iterations of CAD modeling and precision machining. The necessary apparatus for characterizing the gun, including the vacuum chamber and electron spectrometer, was also custom-designed for accuracy, speed, and flexibility. Finally, the commissioned electron gun achieved results matching well with simulations, achieving a peak energy near 1 keV with 40 fC bunch charge in one operation point, and monochromatic 400 eV electron bunches with 5.8% energy spread in another. Characterization of the THz phasing and THz scaling were in line with expected theoretical behavior. The peak accelerating field on the gun was characterized to be higher than that of the best RF guns by a factor of 1.7 without causing any damage and operating for over 1 billion shots.
6.2 Future outlook

Despite the important gains made in the past several years to high field accelerators using THz radiation, some of which are described in this thesis, much more work needs to be done to realize the technology’s full potential.

One significant hurdle is achieving the necessary THz energy levels. Initial simulations predict THz energies on the order of 1-10 mJ are needed to achieve MeV electrons from a gun [23] and 20 mJ to achieve 10 MeV acceleration from a linac [117]. Given that experimental imperfections are often overlooked in initial simulations, these numbers likely represent the required minimum THz energy in the best case scenario.

Of course, to scale up the THz energy, one does not need to make innovative feats to achieve higher efficiency. One can simply apply the brute force method of building more powerful pump lasers. A 1 J laser could potentially produce 10 mJ THz pulses if the efficiency remained constant during scaling. However, it is likely the efficiency would decrease due to the asymmetric nature of the tilted pulse front pumping scheme. Additionally, a 1 J laser operating at a useful repetition rate (∼1 kHz) is likely prone to frequent maintenance difficulties and would no longer be compact. Finally, ensuring that the beam quality of the laser remains in good condition is an ongoing research endeavor [5].

Advances are being made to improve the THz efficiency on several fronts, such as echelon-based tilted pulse front generation in lithium niobate. Indeed, first experiments with the echelon already yielded efficiencies of 0.66% at cryogenic temperatures (based on pyroelectric detection) [85], and almost 3x higher efficiency when compared side-by-side with grating-based generation under the same conditions.

Organic crystals have also yielded auspicious results, albeit only from a small number of research groups thus far. The highest optically-generated THz pulse energy produced thus far, 0.9 mJ, was achieved by impinging a tiled organic crystal mosaic with a rare high energy 1.25 μm laser.

Yet another possibility is that of THz beam combining. With improvements to
pump pulse recycling, it is conceivable that similar conversion efficiencies can be achieved in the second, third, or even further stages using the same pump laser. To leverage all these ‘re-pumped’ THz pulses, one would need to conceive methods to coherently combine them into a single high energy pulse. Alternatively, one can design a gun which accepts THz pulses incident from multiple angles and combines them at the acceleration point.

Finally, much theoretical work has been done by Ravi et al. on the efficient generation of multi-cycle THz pulses [99, 96]. Such pulses can be coupled into a more conventional type of electron gun (although dimensioned for THz wavelengths) which has multiple resonant cavities that store up energy from the many cycles of incoming THz radiation. Future efficiency results from the experimental demonstration of these techniques will bring to light whether one should focus on designing multi-cycle or single-cycle THz accelerator structures.

As we have observed from this short discussion, in order to advance THz-driven accelerators, future efforts on THz generation/manipulation and THz-driven accelerator design must be made in close synchronization.
Appendix A

List of Publications


Appendix B

List of Conference Proceedings


[100] J. Rosenzweig, N. Barov, S. Hartman, M. Hogan, S. Park, C. Pellegrini, G. Trav- 
ish, R. Zhang, P. Davis, G. Hairapetian, and C. Joshi. Initial measurements of 
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D. Golde, T. Meier, M. Kira, S. W. Koch, and R. Huber. Sub-cycle control 
of terahertz high-harmonic generation by dynamical Bloch oscillations. *Nature 

[102] Mostafa Shalaby and Christoph P Hauri. Demonstration of a low-frequency 
three-dimensional terahertz bullet with extreme brightness. *Nature communica-


[104] Prasahnt Sivarajah, Benjamin K. Ofori-Okai, Stephanie M. Teo, Christopher A. 
Werley, and Keith A. Nelson. The homogenization limit and waveguide gradient 
index devices demonstrated through direct visualization of THz fields. *New 

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121, nov 2011.

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