NONLINEAR SYSTEMS FOR FREQUENCY CONVERSION FROM IR TO RF

Dissertation
Submitted to
The School of Engineering of the
UNIVERSITY OF DAYTON

In Partial Fulfillment of the Requirements for
The Degree of
Doctor of Philosophy in Electro-Optics

By
Brian D. Dolasinski, M.S.

UNIVERSITY OF DAYTON
Dayton, OH
December 2014
NONLINEAR SYSTEMS FOR
FREQUENCY CONVERSION FROM IR TO RF

Name: Dolasinski, Brian David

APPROVED BY:

___________________                                        _______________
Joseph W. Haus, Ph.D.                                        Partha Banerjee, Ph.D.
Advisory Committee                                           Committee Member
Chairman                                                     Director
Associate Professor                                         Electro-Optics
Electro-Optics Program                                      Program

___________________                                        __________________
Imad Agha, Ph.D.                                             Adam Cooney, Ph.D.
Committee Member                                            Committee Member
Assistant Professor                                          Director
Physics Program                                             Electro-Optics

___________________                                        __________________
John G. Weber, Ph.D.                                         Eddy M. Rojas, Ph.D., M.A., P.E.
Associate Dean                                              Dean
School of Engineering                                        School of Engineering
ABSTRACT

NONLINEAR SYSTEMS FOR
CONVERSION FROM IR TO RF

Name: Dolasinski, Brian David
University of Dayton

Advisor: Dr. Joseph W. Haus

The objective of this dissertation is to evaluate and develop novel sources for tunable narrowband IR generation, tunable narrowband THz generation, and ultra-wideband RF generation to be used in possible non-destructive evaluation systems.

Initially a periodically poled Lithium Niobate (PPLN) based optical parametric amplifier (OPA) is designed using a double-pass configuration where a small part of the pump is used on the first pass to generate a signal, which is reflected and filtered by an off-axis etalon. The portion of the pump that is not phase matched on the first pass is retro-reflected back into the PPLN crystal and is co-aligned with the narrow bandwidth filtered signal and amplified. We demonstrate that the system is tunable in the 1.4 μm -1.6 μm signal range with a linewidth of 5.4 GHz.

Next the outputs of seeded, dual periodically poled lithium niobate (PPLN) optical parametric amplifiers (OPA) are combined in the nonlinear crystal 4-dimethylamino-N-methyl-4-stilbazolium-tosylate (DAST) to produce a widely tunable narrowband THz source via difference frequency generation (DFG). We have demonstrated that this novel...
configuration enables the system to be seamlessly tuned, without mode-hops, from 1.2 THz to 26.3 THz with a minimum bandwidth of 3.1 GHz. The bandwidth of the source was measured by using the THz transmission spectrum of water vapor lines over a 3-meter path length. By selecting of the DFG pump wavelength to be at 1380 nm and the signal wavelength to tune over a range from 1380 nm to 1570 nm, we produced several maxima in the output THz spectrum that was dependent on the phase matching ability of the DAST crystal and the efficiency of our pyro-electric detector. Due to the effects of dispersive phase matching, filter absorption of the THz waves, and two-photon absorption multiple band gaps in the overall spectrum occur and are discussed. Employing the dual generator scheme, we have obtained THz images at several locations in the spectrum using an infrared camera that runs at a rate of 35 frames per second. We have demonstrated the ability to image 2 THz to 26 THz both in static and in real time conditions. We will present images of carbon fibers illuminated at different THz frequencies.

Lastly, microwave generation was demonstrated by ultrafast photo-excitation experiments to induce non-equilibrium quasi-particle relaxation. Using a laser with a pulse energy of 1 mJ and a pulse duration greater than 120 fs (808 nm wavelength) incident on a charged, superconducting YBa$_2$Cu$_2$O$_{7-\delta}$ (YBCO) thin film ring, the photo-response was measured with a series of microwave antennas. From the observed nanosecond response time of the transient pulse, we extracted the frequency spectrum in the GHz regime that was dependent on the incident beam diameter, pulse duration, power, and the physical structure of the YBCO thin film.
ACKNOWLEDGMENTS

I would like to acknowledge my former advisor, Professor Peter Powers. I will be forever thankful for his guidance, encouragement, and positive attitude during my study in the University of Dayton. He was one of my best role models for a scientist, mentor, and friend. Peter was the reason I have decided to pursue a career in nonlinear optics. I am also very grateful to my other advisor Professor Joseph Haus for his support and guidance over the course of my research with LOCI. I also wish to thank Dr. Banerjee, Dr. Agha, and Dr. Cooney for serving on my doctoral committee and their helpful insights.

I would like to thank Dr. Jason Deibal and Doug Petkie at Wright State University for their assistance in THz imaging. Also a special thanks to Dr. Tim Haugen and Dr. Tom Bullard of AFRL for the opportunity to study microwave generation. I would also like to thank Tom Bullard, John Bulmer, and Jay Patel for their group effort contribution during several superconductor experiments.

Most importantly I would like to thank my wife Heather, my Parents Dave and Mary, and my sister Lise who supported my educational pursuits.
TABLE OF CONTENTS

ABSTRACT .............................................................................................................. iii

ACKNOWLEDGMENTS .............................................................................................. v

LIST OF FIGURES ........................................................................................................ viii

LIST OF TABLES ......................................................................................................... x

CHAPTER 1: INTRODUCTION AND OBJECTIVES ......................................................... 1
  1.1 Introduction ........................................................................................................ 1
  1.2 Problem Statement ........................................................................................... 2
  1.3 Review of Related Techniques ........................................................................ 5
    1.3.1 Overview of Infrared Generation ............................................................... 5
    1.3.2 Current THz wave techniques ................................................................. 7
    1.3.3 Description of Wideband Microwave Generation .................................... 9
  1.4 Motivation .......................................................................................................... 11
  1.5 Experimental Approach .................................................................................. 12
  1.6 Dissertation Outline ......................................................................................... 13

CHAPTER 2: PERIODICALLY POLED LITHIUM NIOBATE SEEDED OPTICAL PARAMETRIC AMPLIFIER ................................................................. 15
  2.1 Introduction ........................................................................................................ 15
  2.2 Nonlinear Interactions ..................................................................................... 16
  2.3 Overview of PPLN Injection Seed OPG/OPA .................................................. 17
    2.3.1 OPA Wave Equation ................................................................................. 20
  2.4 Quasi-Phase Matching (QPM) ........................................................................ 24
  2.5 Periodically Poled Lithium Niobate (PPLN) OPA ........................................... 24
  2.6 Etalon Seeded PPLN OPA ................................................................................ 29
    2.6.1 Overview of off-axis Fabry-Perot etalon feedback seed ......................... 29

CHAPTER 3: NARROW BANDWIDTH TUNABLE OPA .................................................... 36
  3.1 Introduction ........................................................................................................ 36
  3.2 Narrow Bandwidth OPA ................................................................................... 37
    3.2.1 Dual Pass OPG method ............................................................................ 37
    3.2.2 Experimental Setup ................................................................................... 41
  3.3 Results ................................................................................................................ 44
  3.4 Application ......................................................................................................... 48
  3.5 Conclusion .......................................................................................................... 51
LIST OF FIGURES

Figure 1.1. Electromagnetic Spectrum. ................................................................. 1

Figure 1.2. Contour plot of the parametric gain for the idler and signal wavelengths versus the pump wavelength. The top has a QPM period of 30.68 microns and the bottom has a period of 34.03 microns. Figure taken from [15]. ........................................ 5

Figure 1.3. Experimental setup and generated spectrum of the generated THz waves. The resolution of such a system is 100GHz. Figure taken from reference [29]. ................. 9

Figure 1.4. Gain vs Frequency characteristics of L shaped slot antenna. Antenna radius $r = 80$ mm. Figure taken from reference [34]. ......................................................... 10

Figure 2.1. Optical parametric generation (OPG) is where the single pump photon splits into a signal photon and idler photon. .......................................................... 18

Figure 2.2. Seeded OPG resulting in OPA of signal photon ................................... 19

Figure 2.3. The normalized intensity output phase matched bandwidth of OPG and OPA in PPLN. ........................................................................................................... 23

Figure 2.4 PPLN with length $L$ and single crystal grating $\Lambda$. .......................... 25

Figure 2.5. MgO:PPLN OPA with a multi-grating crystal. ....................................... 26

Figure 2.6. The ordinary and extraordinary refractive indices for MgO:PPLN. ........ 27

Figure 2.7. QPM MgO:PPLN phase matching bandwidth for a single period. ........ 28

Figure 2.8. An off axis Fabry-Perot etalon that gives rise to multiple beam interference in both the transmission and reflection orders. ........................................ 29

Figure 2.9. Fabry-Perot transmission as a function of wavelength and reflection coefficient. .................................................................................................................. 32

Fig. 2.10. An off-axis etalon where the frequencies satisfy the peak transmittance condition of Eq. 2.47. ................................................................. 34

Figure 3.1. Reflection polarization rotator with the fast axis of a quarter-wave plate rotated to 45 degrees. .................................................................................... 38
Figure 3.2. First pass and second pass scheme for the narrowband generation of an OPA. .................................................................39

Figure 3.3. QPM bandwidth for MgO:PPLN at different periodicities and temperatures. .................................................................40

Figure 3.4. The configuration of the off-axis etalon injection seeded PPLN OPA. ........41

Figure 3.5. Measured transmission of a doubled-pass etalon at the maximum FSR and minimum FWHM. .........................................................43

Figure 3.6. Narrow Bandwidth OPG Source operating at 1550 nm (a.) Input energy loss (b.) Output energy ..........................................................45

Figure 3.7. (a.) Mirror Seeding (b.) Etalon-Mirror Seeding (c.) Etalon Seeding ........45

Figure 3.8. Narrowband OPG injection seeding with a mirror, etalon-mirror combination, and with an etalon alone. ..........................................................46

Figure 3.9. Spectrum measurements of the narrowband OPG laser scan, centered at 1549 nm, and tuned over the whole signal band. ..............................47

Figure 3.10. Illustration and measurement of (a) double-pass etalon tuning and (b) OPA injection seeding centered on multiple OPG bandwidths. ..................48

Figure 3.11. The experimental setup to measure the absorption features of CO₂. ..........49

Figure 3.12. The absorption of pure CO₂ in the IR spectrum simulated with HITRAN. The data was simulated with a laser path length of 10 m, a temperature of 296 K, and at a pressure of 70 Torr. ..........................................................49

Figure 3.13. Carbon dioxide absorption features measured with etalon injection seeded OPA.................................................................50

Figure 4.1. Diagram of Difference Frequency Generation (DFG) in χ(2)material. The second frequency is amplified similar to OPA while the THz frequencies are generated. ......................................................................54

Figure 4.2. Molecular diagram of active nonlinear chromophore cation stilbazolium and anion tosylate. .............................................................56

Figure 4.3. Refractive indices of DAST. ..........................................................58

Figure 4.4. Absorption coefficients for DAST in SWIR regime. ..........................59

Figure 4.5. Index of refraction frequency dependence along the a-axis of the DAST crystal. ..................................................................................59
Figure 4.6. Logarithmic absorption along the a-axis of DAST as a function of frequency. .................................................................60

Figure 4.7. DAST DFG tuning bandwidth. The region that is red signifies where phase matching is most probable. .................................................................61

Figure 4.8. Output map of DAST DFG as a function of combined wavelength polarization angle and frequency. .................................................................62

Figure 5.1. Fan out grating period and quasi-phase matched signal range for PPLN crystal with temperature variations. .................................................................65

Figure 5.2. Experimental setup used to generate a narrowband THz wave. The dual injection seeded OPG is mixed in the DFG crystal DAST. .................................................................66

Figure 5.3. The multi-grating OPA and the fan out grating OPA. The dual outputs are both narrowband and tunable to provide the necessary frequency separation for DFG to generate THz waves. .................................................................68

Figure 5.4. FTIR transmission measurements of materials to be used as filters for SWIR signal wavelengths. .................................................................68

Figure 5.5. Comparison of measured DFG system transmission with the known THz transmission spectrum in air at a 3 m path length at STP. No background corrections were made to the measured signal data. .................................................................70

Figure 5.6. Generated THz transmission spectrum from 1 THz to 27 THz. The scan is presented without background correction and plotted using five different scans with partially overlapping scanning ranges and different filter combinations. The data was continuously recorded as the fan out MgO:PPLN crystal was tuned and scanned across the desired spectrum at a 3.1 GHz linewidth. .................................................................72

Figure 5.7. Measured DAST DFG signal as a function of combined OPA polarization angle and frequency. No background correction was performed and the data was recorded at increments of $\Delta \lambda = 0.15$ nm. .................................................................73

Figure 5.8. Output mapping of DAST DFG signal from 3.8 THz to 12 THz. (a) Modeled DFG power as a function of polarization angle and frequency. (b) Measured DFG signal as a function of polarization angle and frequency. No background correction was performed and the data was recorded at increments of $\Delta \lambda = 0.15$ nm. .................................................................74

Figure 5.9. Output mapping of DAST DFG signal from 12 THz to 27 THz. (a) Modeled DFG power as a function of polarization angle and frequency. (b) Measured DFG signal as a function of polarization angle and frequency. No background correction was performed and the data was recorded at increments of $\Delta \lambda = 0.15$ nm. .................................................................74
Figure 5.10. Output power from DAST DFG. (a) Modeled output power as a function of OPG power and frequency. (b) Measured output power as a function of OPG power and frequency with no background corrections made and the data was taken at increments of $\Delta \lambda =0.15$ nm.

Figure 5.11. Transmission spectrum using a single IR filter placed after the DAST crystal without a background correction. The data was recorded in increments of $\Delta \lambda =0.35$ nm.

Figure 5.12. Imaged THz waves from a 3 meter path length from the DAST Crystal.

Figure 6.1. Measurement of surface roughness of different heat treated SiNC samples.

Figure 6.2. The configuration depicts the method in which the reflection spectrum was recorded via single point detection or imaging with a camera.

Figure 6.3. THz reflection measurement of six scans for the three SINC heat-treated samples with the developed source. No background correction was made to the measured samples. The gaps in the spectrum are due to the phase miss matching characteristic inherent to DAST.

Figure 6.4. Averaged reflection data for 6 scans at different locations on each SINC sample.

Figure 6.5. Gaussian oscillator fit of a CMC THz sample scan after a thermal treatment of 10 hours.

Figure 6.6. Permittivity extraction from reflection data for different treated samples.

Figure 6.7. Imaged reflection of the THz beam off the three samples at a frequency of 14.7 THz. The labeled region of interest (ROI) marks where the THz beam is incident on the samples. The images were taken at a standoff distance of 20 cm from the three samples.

Figure 6.8. Imaged reflection of the THz beam off the three samples at a frequency of 19.1 THz. The labeled region of interest (ROI) marks where the THz beam is incident on the samples. The images were taken at a standoff distance of 20 cm from the three samples.

Figure 7.1. An ultrafast laser pulse is incident on YBCO thin film which results in a decrease in Cooper pairs.

Figure 7.2. The transient optical response of YBCO at superconducting temperatures due to different pulse durations.
Figure 7.3. Resistance due to the change in quasi-particle number density. On the time scale of a ns the resistance is that of a delta function. .......................... 107

Figure 7.4. An ultrafast laser pulse is incident on YBCO thin film ring which results in a decrease in current. ................................................................. 108

Figure 7.5. Frequency response of 4rth order RL model with a switch time of $\tau = 1.3 \times 10^{-11}$s and a ring resistance of $R=3.6k\Omega$. ...................................................... 113

Figure 8.1. YBCO ring atop a solenoid wrapped sapphire rod with attached hall probe and thermocouple inside a Styrofoam container placed inside a plastic Dewar. ............................................................... 117

Figure 8.2. The single shot YBCO ring antenna experimental setup. ................................ 117

Figure 8.3. (a.) Linearized horn antenna gain. (b.) Linearized antenna insertion loss. .... 119

Figure 8.4. Illuminated uncharged YBCO ring. (a.) Several measured signals of generated microwave radiation. (b.) Averaged received power spectrum. .................. 121

Figure 8.5. Illumination of a charged YBCO ring. (a.) Generated microwave radiation of several signals averaged together. (b.) Averaged received power spectrum. ................................................................. 121

Figure 8.6. Different frequency bands generated simultaneously in a single transition. ................................................................................................................. 122

Figure 8.7. Microwave response due to different pulse durations. (a) Measured voltage signal due to different laser pulse durations. (b) Frequency response due to different laser pulse durations. ...................................................... 123

Figure 8.8. RF power measured as a function of laser pulse duration. ...................... 124

Figure 8.9 (a.) Measured magnetic field before and after laser illumination as a function of position over the ring. (b.) Radiated magnetic field as a function of current measured from the center of the ring. ......................................................... 125

Figure 8.10. Microwave response due to different induced currents. (a) Measured voltage signal due to different currents. (b) Frequency response due to currents. .... 126

Figure 8.11. Measured peak power as a function of induced current in the ring. ........ 126

Figure 8.12. The measured peak power frequency response of three different rings as a function of average laser power. ......................................................... 127

Figure 9.1. Possible THz source configuration. .......................................................... 132
LIST OF TABLES

Table 2.1 Parameters for the index of refraction in MgO:PPLN. ........................................ 27
Table 4.1. Parameters for DAST SWIR indices of refraction. ............................................. 58
Table 8.1. YBCO Ring Geometry. ......................................................................................... 116
CHAPTER 1
INTRODUCTION AND OBJECTIVES

1.1 Introduction

Terahertz (THz) radiation is defined as the region of the electromagnetic spectrum with frequencies between 100 GHz and 20 THz. A THz gap occurs between the infrared and the microwave spectrum. Infrared devices (IR) have dimensions that are typically much larger than the emitted wavelength and support many modes. In contrast, microwave (RF) electronic devices are comparable in size to the emitted radiation and typically support one mode or lower order modes. Thus, when describing the engineering technology behind the different spectrums we are describing different fields of research. The merging of optical technology with RF devices provides a path of tunability across the gap and the merging of the fields. Therefore, merging of the fields is accomplished by utilizing nonlinear systems to tune across the gap from IR to THz and IR to RF as seen in Fig. 1.1.

Fig. 1.1. Electromagnetic Spectrum
The work in this dissertation is to evaluate and develop novel methods for tunable narrowband IR generation, tunable narrowband THz generation, and ultra-wideband RF generation with IR frequency conversion. These tasks are achieved through modeling and experimentation to predict and analyze the generated spectrums. An additional objective is to show that THz imaging is possible through the use of DFG from a nonlinear crystal when multiple CMC samples are illuminated with a laser that has a high average power, high repetition rate, and a fast wavelength tuning source.

1.2 Problem Statement

Nonlinear optics and optoelectronic techniques have been developed for the production and measurement of broad band and narrow band frequencies that range from infrared (IR) to microwave radiation (RF). Significant progress has been made over the years in the development of sources for IR spectroscopy, terahertz (THz) imaging, and RF sensing. These methods are a primary concern in the nondestructive evaluation (NDE) of ceramic matrix composites (CMC) [1].

Composite materials such as nicalon and carbon fiber are increasingly being used as structural and engine components in high performance military aircraft [2]. Ceramic composites improve a variety of features such as aerodynamic performance, they impart strength to the airframe of the craft, and they can resist the decomposition of the airframe when operating in a corrosive environment [3]. However, due to the complex fabrication process ceramic material can have defects that compromise the strength of the CMC material. The configuration of such material in an aircraft is based on the requirement that the energy from aerodynamic heating is either rejected or absorbed into the material
resulting in fatigue [4]. In addition, during the overall lifecycle of the aircraft operating in the field the materials can show signs of fatigue that can lead to catastrophic failure.

During the manufacturing process, as each layer of the CMC is built up, foreign materials can infiltrate the slurry melt and be embedded within the layers [5]. As the material is tempered the foreign debris can cause a void to open up thus creating gaps between the interwoven fibers. The process serves to decrease the overall strength of the composite by decreasing the layer adhesion. Current methods in quality control are either inadequate or incapable of detecting defects in the field. The result is that the component is removed from the system prior to failing and tested in a lab environment or the material will simply fail in the operating system. In either case the result will be increased costs of part production and downtime of the aircraft. In addition, the maintenance of such materials requires complex inspection and repair methods, which increases the overall downtime of an aircraft.

New methods are being explored to improve and simply the detection of defects on aircraft operating in the field. An aspirational key goal of these explorations is to overcome the shortcomings of traditional NDE methods that involve techniques such as x-ray analysis, ultrasonic evaluation, eddy current detection, and thermography. New NDE technologies that involve noninvasive techniques, noncontact measurements, and non-ionizing energies are being evaluated to accomplish the detection of defects in CMCs in the field. The methods under currently evaluation involve Fourier transform infrared spectroscopy (FTIR) [6], narrowband THz wave sensing [7], and microwave spectroscopy (MS).
Technologies that use the three mentioned spectrums are being used in a variety of applications that span from communications; medical diagnosis, security, computing, etc. However, the aforementioned technologies still need development before they can be deployed in NDE field applications; the current technologies used for NDE are mature [8]. CMC NDE can be performed with all spectra in the lab environment. However, for fielded applications, techniques such as microwave spectroscopy and THz imaging requires a high average power for desirable signal to noise ratios. For instance, infrared spectroscopy requires high power, narrow bandwidth, and wide wavelength tunability. In addition these techniques require a compact source that can easily be maneuvered around the systems which have embedded CMC materials. Thus, this research effort was an attempt to develop key technologies in each spectrum that could effectively be used in equipment to characterize and image ceramic composites.

Quality control solutions for the maintenance of CMC materials are needed by the Air Force to reduce the expenditure of both money and personnel time. By developing powerful sources to overcoming known limitations and supplementing the system design with innovative solutions we can begin to create key technologies to solve the needs of the industry. By using nonlinear conversion techniques to bridge the gap between the incompatible technologies in a given spectrum, new methods and new products can be generated. In this dissertation techniques are developed that generate: tunable narrowband infrared frequencies using self-seeded optical parametric amplification (OPA), widely tunable THz wave generation through optical difference frequency generation (DFG) schemes, and ultra-wideband microwave generation through the interaction of an ultrafast laser pulse with a high temperature superconductive (HTS) loop antenna.
1.3 Review of Related Techniques

1.3.1 Overview of Infrared Generation

Multiple approaches exist to generate synchronized narrowband pump laser pulses for spectroscopy. One common method uses a solid state laser to generate frequency-shifted Stokes beams [9]. When combined with a fiber amplifier system, the laser pulse duration from a master oscillator is spectrally broadened in a nonlinear crystal and amplified in a synchronously pumped optical parametric oscillator (OPO) [10, 11]. With the addition of a seed laser injected into the OPO at low repetition rates, a narrow band OPA is generated [12, 13]. The result is a mid-IR parametric source that spans wavelengths from 1 to 10 μm that is of value in material fingerprinting [14]. The bandwidth of such a system using quasi-phase matched (QPM) periodically poled lithium niobate (PPLN) as an OPA source is given in Fig. 1.2.

![Figure 1.2](image)

Figure 1.2. Contour plot of the parametric gain for the idler and signal wavelengths versus the pump wavelength. The top has a QPM period of 30.68 microns and the bottom has a period of 34.03 microns. Figure taken from [15].

The main advantage of having a parametric source is that the emission results in an ultra-broad tunability that spans several microns [16]. Comparable mid-IR laser sources
have a gain bandwidth that is normally limited to much less than micron [17]. The main disadvantage of an OPA-OPO system is that the configuration requires a cavity to maintain synchronous operation and to stabilize both the power and the wavelength [18]. Another drawback with these nonlinear systems is that they require an additional seed laser introduced into the complex cavity.

A fundamental problem with the OPA-OPO nonlinear optical process is that waves propagating through the material interact travel at different phase velocities. This causes a phase mismatch that prohibits a sustained coherent buildup of frequency converted energies. To bypass the OPA-OPO drawbacks a different method known as quasi-phase matching (QPM) can be used in some materials. QPM techniques were developed to ensure phase matching was optimized by making periodic adjustments to the phase of the transmitted waves. QPM periodically re-orientates regions of the lithium niobate crystal in order to reset the phase of the nonlinear polarization and maintain a coherent buildup of the signal and idler wave conversion [19]. One of the most promising avenues for the application of QPM in with the second-order nonlinear susceptibility ($\chi^2$) optical parametric generation (OPG) process.

The ability of nonlinear OPG to provide broadly tunable coherent radiation made it a key candidate for the development of a tunable IR source. The OPG has a simple configuration since it is a single-pass process and requires no optical cavity. However, the free-running bandwidth is typically too large for many applications in NDE [20]. The bandwidth may be controlled by injection-seeding with a narrow bandwidth signal. Such narrowing has been demonstrated with diode lasers and by using the output of a filtered OPG [21]. Using multiple diodes to cover the spectra is an expensive alternative. The
etalon filtered OPG approach has the advantage of being able to generate a seed over the operating range of the OPG crystal which is comparable to the OPA-OPO. Previous filtered OPG setups consist of a two crystal scheme, where the output of the first OPG is filtered and sent into the second crystal. This approach works well, but the addition of a second crystal also increases the cost of the system.

The double-pass injection seeded OPG was invented to reduce the overall complexity of such an OPO-OPA system and extend the overall wavelength range beyond that which is capable with a standard Mid-IR laser. To the best of our knowledge a double-pass etalon reflected injection seeded optical parametric amplification (OPA) in PPLN crystal has not been configured to generate narrowband frequencies with wide tunability.

1.3.2 Current THz wave techniques

In many studies, broad-bandwidth mode locked femtosecond lasers with high peak power pulses have been utilized with nonlinear crystals to generate broadband THz radiation. Linear filtering techniques for converting single femto-second (fs) light pulses into terahertz repetition-rate emissions have been developed [22]. Using phase filtering pulse rates up to 5 THz has been demonstrated. Pulse shaping is achieved by the spatial filtering of the frequencies components that are dispersed in a lens singlet and a grating. The energy density profile of the shaped pulse was measured by the cross correlation with a 70 fs probe pulse. Due to the cost of such a system and because most of the generated THz waves were too weak to be used, alternative techniques were explored.

A method involving electro-optic materials and fs pulses was discovered and became the next generation technique for the emission of THz waves. A femto-second laser pulse was passed through the electro-optic material LiTaO$_3$ which generated sub-
picosecond pulses [23]. The radiated time resolved picosecond pulses indicated that the
THz bandwidth was generated from the surface of the electro-optic material from a region
within one coherence length. This method demonstrated that THz waves could be
efficiently generated through optical means without the use of a semi-conductive gap
antenna.

In 1994 a new nonlinear technique emerged for generating THz waves by the
optical rectification of femtosecond pulses in electro-optic materials such as 4-
dimethylamino-N-methyl-4-stilbazolium-tosylate (DAST) [24]. One of the added benefits
of such a scheme was the scalability of the rectified electric field strength of the THz waves
as a function of incident optical laser fluence, crystal thickness, and crystal damage
threshold.

Other promising schemes for the generation of high power, narrow-band, sub-
millimeter waves employ nonlinear difference frequency generation (DFG) between two
laser sources combined in a crystal with a high nonlinear coefficient [25]. Generation of
widely tunable THz radiation sources was achieved using nonlinear parametric oscillation
(OPO) [26], and optical parametric generation (OPG) [27]. Multiple advantages are
associated with the DFG method that facilitate wide band tunability with narrow bandwidth
THz waves. However, several limitations are found when the DFG process is used in an
OPO configuration. OPO schemes result in drawbacks such as mode hopping and broad
bandwidth emission on the order of 100 GHz [28]. Such a system is given in Fig. 1.3 where
water line absorption is not present due to the large bandwidth of the generated spectrum.
The drawbacks limit the applications of OPO-DFG schemes. Many of the applications require that such a system possess the inherent ability for wide tunability with a narrow bandwidth to optimize the object transparency or material fingerprint. We present a method that consists of dual tunable OPAs to generate narrow bandwidth to serve as DFG mixing frequencies for the emission of THz waves over a broad spectrum.

1.3.3 Description of Wideband Microwave Generation

Wideband (WB) microwave generation has the potential to revolutionize communications and microwave spectroscopy in that it transmits and receives pulsed waveforms temporally compressed. In contrast, the traditional microwave method transmits and receives pulses that are compressed in frequency, i.e. they are narrowband. The WB method allows the transmission over a wide range of frequencies and a low power density can be used for material sampling. A great deal of interest in this field has been recently sparked by the FCC’s permission to use the unlicensed 3-10 GHz range [30]. A number of multiband antennas has been evaluated for WB microwave applications such as monopole antennas, slot antennas, and inverted antennas. There are a multitude of antennas that can serve in the WB regime that are not limited to G-shape radiators [31], E-shaped
slot ground plane [32], ring monopole [33], etc. However, while most of these antennas can cover the R through Ku band in discrete sections, they require large areas in the order of the wavelength $50 \times 50 \text{mm}^2$ used to transmit or detect. Such antenna characteristics are given in Figure 1.4.

![Gain vs Frequency characteristics of L shaped slot antenna](image)

Figure 1.4. Gain vs Frequency characteristics of L shaped slot antenna. Antenna radius $r=80$ mm. Figure taken from reference [34].

Traditionally, reducing the size of an antenna decreases its performance characteristics such as the bandwidth, gain, and efficiency [35]. Due to the known limitations of traditional antennas we explored new techniques involving high temperature superconductors, which may be explored and developed to overcome the undesired radiator characteristics.

An option for wideband microwave generation is a method that involves the interaction of an ultrafast laser and a thin film high temperature superconductor (HTS). A femtosecond laser pulse incident on a semiconductor photoconductive switch is generally used for the generation of THz waves and as a detector for THz waves [36]. In addition, thin film HTS material has been evaluated as a THz generation source acting like an ultrafast photoconductive switch [37]. THz radiation has been generated by the ultrafast modulation of the super-current in the HTS thin film excited by an IR femtosecond laser pulse. However, these interactions have been limited to the THz domain.
We extend the transient photo-response to simultaneously generate frequencies on the scale of GHz and THz. Our technique uses a fixed IR wavelength of 808 nm with different pulse durations. To the best of our knowledge, the interaction between a HTS thin film with an ultrafast laser has not been used to generate such low frequencies.

In our experiments a non-traditional antenna design is evaluated. The means in which we measure the generated microwave radiation is performed by pulsed RF S-parameter measurement method, which utilizes several antennas to record the ultra-wideband signal. The HTS device was fabricated on a sapphire substrate to ensure the transparency of our high energy femtosecond laser pulses (Ti:sapphire regenerative amplifier system). The selected substrate material also preserves the bandwidth of the transient signal with minimal material interaction. In the experiment we utilized a high bandwidth (65 GHz) digital sampling oscilloscope in combination with two types of receiving antennas which included a 18-45 GHz horn antenna and a 1-18 GHz horn antenna.

1.4 Motivation

From the discussion in the prior sections, most of the work that was performed to generate the IR, THz, and GHz frequencies (sources) covered the desired spectra of interest. However, many of the drawbacks such as the footprint of the source, complexity, and bandwidth limitations are obstacles that prevent the transition of the technologies to the field. The NDE applications of interest in the IR and THz, especially spectroscopic ones, require seamless tunability with a narrow bandwidth that can be developed into a compact source. While for microwave generation an ultra-wideband compact source is desired for signal sampling techniques.
The objective of this dissertation is to evaluate and develop novel methods for tunable narrowband IR generation, tunable narrowband THz generation, and ultra-wideband RF generation by IR frequency conversion. These tasks are achieved through modeling and experimentation to predict and analyze the generated spectrums. In an additional objective, we applied the developed THz source as a spectroscopic tool to detect chemical degradation in several CMC samples.

1.5 Experimental Approach

The first step of this research was to set up an injection seeded OPG system using a parallel domain periodically poled lithium niobate crystal (PPLN). Using this configuration, measurements were taken to estimate the wavelength tunability by injection seeding with the reflection from an off-axis double-pass etalon. With the tunability limitations noted, dual injection seeding with individual laser diodes and crystals were explored by using two different poling domains. Based on the frequency separation of the two individual seeded OPG stages, difference frequency generation (DFG) was demonstrated with the combination of the two signals. DFG was performed in the nonlinear crystal 4-dimethylamino-N-methyl-4-stilbazolium-tosylate (DAST), in which we explored narrowband seamless tunability across 28 THz. Using this setup, data was collected to make estimates of the electrical and optical properties of CMC materials. The composite material parameters were then used, along with scattering theory and optical medium transfer theory, to analyze the reflection data. Various single point detection scans were collected from the samples with various forms of thermal representative damage.

The next goal was to use ultrafast IR laser pulses to generate microwave radiation. The incident laser was initially collimated to match the outer diameter of an HTS thin film YBa$_2$Cu$_2$O$_{7-\delta}$ (YBCO) ring and in other explorations the beam diameter was collimated to
match the track width of the film. A current was induced on the YBCO samples by inducing a change of magnetic flux using a solenoid. A series of broad band microwave antennas were used to detect and measure the emitted frequencies due to the photo-response of the material. With this setup, the multiple studies were performed to measure the material parameters including, but not limited to, laser power, laser polarization, current on the ring, ring diameter, laser beam diameter, laser position on the ring, etc.

1.6 Dissertation Outline

In Chapter 2 nonlinear interactions of interest will be discussed. These interactions focused on optical parametric amplification (OPA) in PPLN, quasi-phase matching (QPM) techniques are introduced using periodically poled lithium niobate (PPLN), and PPLN techniques are discussed. In this chapter we discuss how PPLN-OPA is used to frequency convert a pump source to the desired spectrum. Chapter 3 is a complete setup and description of the developed tunable nonlinear IR source. In Chapter 4 THz difference frequency generation (DFG), DAST properties, and phase matching calculations are discussed. Then in Chapter 5, the developed narrowband DFG-THz source is discussed. In Chapter 6 measurements of CMC materials and protective coatings with the DFG-THz source are discussed. Chapter 7 is an introduction to the physics describing the interaction between an ultrafast laser pulse and a high temperature superconductor (HTS) ring. The purpose of this chapter is to describe how the interaction induces a photo-transient response. Lastly, Chapter 8 is a study on the generation of wideband microwaves from the illuminated HTS source.
CHAPTER 2

PERIODICALLY POLED LITHIUM NIOBATE SEEDED OPTICAL
PARAMETRIC AMPLIFIER

2.1 Introduction

Nonlinear optics is a richly diverse research field in which the nonlinear response of some materials is used to induce optical mixing (cross-talk) between several incident optical radiation fields. When the incident electric fields are strong, which is the case with pump lasers, the properties in some crystals can be affected by the amount of excess energy. Classically, these types of processes can be describe in terms of the wave equation. The vehicle by which all nonlinear effects occur is the polarization. The polarization is then describe by a Taylor series expansion in the electric field term given as

\[ P(t) = \chi^{(1)} E(t) + \chi^{(2)} E^2(t) + \chi^{(3)} E^3(t) + \chi^{(4)} E^4(t) + \ldots \]  

(2.1)

In Eq. 2.1 the linear susceptibility is given by \( \chi^{(1)} \) and all others represent the higher order harmonics that give rise to nonlinear response in the material. A number of processes can be inferred from these higher order mixing terms when more than one laser is present at more than one wavelength. The second-order susceptibility is responsible for the primary interactions of interest. These interactions include but are not limited to second harmonic generation, optical parametric generation, optical parametric amplification, difference frequency generation, and sum frequency generation.
This chapter focuses on the key topics in nonlinear optics that were used to develop sources in the infrared to be used as possible candidates for nondestructive evaluation tools. In this chapter OPG and OPA process will be covered as well as its relevance in generating tunable narrowband spectrums in the IR. Difference frequency generation will be covered in Chapter 4 due to its key role in this dissertation as a THz generation method.

2.2 Nonlinear Interactions

The nonlinear interactions of interest are associated with the $\chi^{(2)}$ term of the nonlinear susceptibility. We can consider two incident plane waves on a nonlinear material. Initially the two plane waves are independent of one and other and are represented as

$$E(z, t) = E_2 e^{i(k_2 z - \omega_2 t + \varphi_2)} + E_3 e^{i(k_3 z - \omega_3 t + \varphi_3)} + \text{c.c.} .$$  \hspace{1cm} (2.2)

The electric field can be rewritten in terms of its complex amplitude

$$E(t) = A_2 e^{-i\omega_2 t} + A_3 e^{-i\omega_3 t} + \text{c.c.} .$$  \hspace{1cm} (2.3)

When considering the polarization effect of the $\chi^{(2)}$ term between the two fields in the nonlinear medium, the result becomes the square of the total electric field

$$P^{(2)}(t) = \chi^{(2)}(E(t))^2 .$$  \hspace{1cm} (2.4)

By only evaluating the temporal component of the incident fields and absorbing the phase into the complex amplitude, the total polarizability can be written as

$$P^{(2)}(t) = \chi^{(2)}[A_2^2 e^{-i2\omega_2 t} + A_3^2 e^{-i2\omega_3 t} + 2A_2A_3 e^{-(i\omega_2 + i\omega_3)t} + 2A_2A_3^* e^{-(i\omega_2 - i\omega_3)t} + 2A_2^*A_3 + 2A_2A_3^* + \text{c.c.}] .$$  \hspace{1cm} (2.5)

A close inspection of the polarization equation reveals that new frequency mixing terms have arisen. Due to the varied frequencies from mixing it is common to rewrite the polarization in terms of the processes. Therefore we can sum over both the negative and positive frequency components in the polarization as
\[ P^{(2)}(t) = \sum_{i=1}^{n} P(\omega_i) e^{-i\omega t} \quad \text{where} \quad i = 1, 2, 3, \ldots \] (2.6)

The second order nonlinear interactions can now be identified in terms of their independent complex amplitudes which are

\[
\begin{align*}
P(2\omega_2) &= \chi^{(2)} A_2^2 & \text{SHG} \rightarrow \omega_2 \\
P(2\omega_3) &= \chi^{(2)} A_3^2 & \text{SHG} \rightarrow \omega_3 \\
P(\omega_2 + \omega_3) &= 2\chi^{(2)} A_2 A_3 & \text{SFG} \rightarrow \omega_2 + \omega_3 \\
P(\omega_2 - \omega_3) &= 2\chi^{(2)} A_2 A_3^* & \text{DFG} \rightarrow \omega_2 - \omega_3 \\
P(0) &= 2\chi^{(2)} [A_2 A_2^* + A_3 A_3^*] & \text{Rectification}
\end{align*}
\]

In Eq. 2.7 we note that four out of the five second order processes generate frequency components that can be utilized to generate wavelengths different than the input components. However, due to a variety of nonlinear material limitations generally only one process tends to be efficient enough to be used in applications. The phase matching condition normally can’t efficiently be satisfied for more than one process at a time.

2.3 Overview of PPLN Injection Seed OPG/OPA

The key to having a laser with a large tuning spectral range is the development of optical parametric generation (OPG). The method of OPG is quite simple; in a nonlinear crystal such as PPLN, a high intensity/power, high frequency beam at \( \omega_p \) (pump beam), is converted into a low power, low frequency signal \( \omega_s \). In the process a third beam or idler beam is generated at a frequency \( \omega_i \). A graphical representation is given in Fig. 2.1.
Figure 2.1. Optical parametric generation (OPG) is where the single pump photon splits into a signal photon and idler photon.

The frequency ordering of the three emissions are $\omega_i < \omega_s < \omega_p$. The signal and idler frequencies are tunable but are limited by the conservation of energy and QPM requirements of the crystal. QPM will be covered in more detail in later in the chapter where it plays a key role in PPLN nonlinear techniques. In order for such an interaction to occur, energy conservation must take place,

$$
\hbar \omega_p = \hbar \omega_s + \hbar \omega_i. \tag{2.9}
$$

For an interaction to be efficient, conservation of momentum must be satisfied as well, and is expressed in terms of the three wave vectors as

$$
\hbar k_p = \hbar k_s + \hbar k_i. \tag{2.10}
$$

OPG is a process to transfer a fixed frequency, high energy pump beam to a low power, variable frequency signal beam. OPG can be exploited to achieve frequency tunability. The primary method is done by amplifying a weak seed signal beam while generation is being done in the nonlinear crystal and is commonly known was optical parametric amplification (OPA). In this method we can use the principles of a seeded QPM-OPG to generate a narrow line width OPA and can be viewed in Fig. 2.2.
A semi-classical method to understanding narrowband OPA is to view the QPM-OPG as a process between the pump photon and a quantum fluctuation. The quantum fluctuation can be thought of as $\frac{1}{2}$ photon per mode in a region in the nonlinear crystal where the pump beam resides. Another method is to consider a situation where there is 1 photon per mode in the signal beam and no photons in the idler branch. The photons in the signal branch get amplified in the conversion process of the pump beam in which they have an efficiency response that can be modeled as a sinc squared function. Now, to narrow the phased matched bandwidth of the QPM-OPG a narrow band laser diode can be used as a weak injection seed. The process is equivalent to injecting an excess number of photons into a single mode of the crystal. These excess photons are then amplified higher than the OPG background. The result is that the seeded OPG can be used as a narrowband and tunable OPA source. The only drawback is that the OPA requires high pump intensities that operate at low repetition rates which are typically 1 to 100 kHz. However, it provides high output energies, broad frequency tunability, and are easy to operate due to the lack of an oscillator cavity.
2.3.1 OPA Wave Equation

In order to understand the injection seeded OPA we can derive the wave equations to estimate the coupling effect between the fields. In this model, for the time being, absorption and diffraction will be neglected. However, dispersion is included in the model due to phase matching. From Maxwell’s equations

\[ \nabla^2 \vec{E} = \varepsilon_0 \frac{\partial^2 \vec{D}}{\partial t^2} \quad \text{Wave Equation} \]

\[ \vec{D} = \varepsilon_0 \vec{E} + \vec{P}^L + \vec{P}^{NL} \quad \text{Constitutive Equation} \] \hspace{1cm} (2.11)

\[ \nabla^2 \vec{E} = \mu_0 \varepsilon_0 \frac{\partial^2 \vec{E}}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}^L}{\partial t^2} + \mu_0 \frac{\partial^2 \vec{P}^{NL}}{\partial t^2} \quad \text{Nonlinear Wave Equation} \]

We can develop the wave equation and implement the constitutive condition to re-derive the wave equation in terms of the nonlinear polarization which is given in Eq. 2.11. The optical parametric amplification process can be interpreted from the Manley-Rowe coupled nonlinear equations derived from the nonlinear wave equation. A linearly polarized, monochromatic plane wave at a frequency \( \omega \), propagating in the \( z \) direction is:

\[ \mathbf{E}(z, t) = \text{Re}\{A(z)e^{j(\omega t - kz)}\}. \] \hspace{1cm} (2.12)

The plain wave travels through a medium of nonlinear polarization at the same frequency.

\[ \mathbf{P}^{nl}(z, t) = \text{Re}\{A(z)e^{j(\omega t - k_p z)}\}. \] \hspace{1cm} (2.13)

By implementing the slowly varying amplitude approximation:

\[ \frac{d^2 A}{dz^2} \ll 2k \left| \frac{dA}{dz} \right|, \] \hspace{1cm} (2.14)

we can derive the propagation equation of the field,

\[ \frac{dA}{dz} = -j \frac{\mu_0 c_0 \omega}{2n} \mathbf{P}^{nl} e^{[-j(k_p - k)z]}. \] \hspace{1cm} (2.15)
In Eq. 2.15, $c_0$ is the speed of light in vacuum, $n$ is the refractive index at frequency $\omega$, and $k$ is the wave vector in the material. The propagation equation depicts the nonlinear polarization, which acts as the source driving term for the variations in the amplitude of the propagation wave.

The interaction between three waves in a nonlinear medium, at frequency $\omega_p$, $\omega_s$, and $\omega_i$ can now be explored using the propagation equation. The frequencies selected are such that $\omega_i < \omega_s < \omega_p$ and $\omega_p = \omega_s + \omega_i$. The component of the nonlinear polarization vector along the $x$ direction is expressed as

$$\mathbf{P}_i^{nl} = \varepsilon_0 \chi^{(2)}_{ijk} E_j E_k,$$

(2.16)

In Eq. 2.16 $\chi^{(2)}$ is a third-rank second order nonlinear susceptibility tensor where the Einstein summation convention is used. The three waves are assumed collinear, all wave vectors are parallel, with the same polarization component. Thus, the coupled wave equations for all three frequencies can be written as

$$\frac{dA_i}{dz} = -j \frac{\omega_i d_{\text{eff}}}{n_i c_0} A_i^* A_p e^{(-j\Delta k z)}$$
$$\frac{dA_s}{dz} = -j \frac{\omega_s d_{\text{eff}}}{n_s c_0} A_s^* A_p e^{(-j\Delta k z)}.$$ 
$$\frac{dA_p}{dz} = -j \frac{\omega_p d_{\text{eff}}}{n_p c_0} A_p A_i e^{(-j\Delta k z)}.$$

(2.17)

In the equations above $d_{\text{eff}}$ is the effective nonlinear coefficient, this is dependent on the propagation direction and the polarization of the three waves which is determined by quasi-phase matching. This is related to the second-order susceptibility by

$$\chi^{(2)} = \frac{1}{2} d_{\text{eff}} = \frac{1}{2} d.$$

(2.18)
After some algebraic manipulations, the three coupled equations can be written in the form of the Manley-Rowe (M-R)

\[
\frac{1}{\omega_i} \frac{dI_i}{dz} = \frac{1}{\omega_s} \frac{dI_s}{dz} = \frac{1}{\omega_p} \frac{dI_p}{dz}.
\] (2.19)

In Eq. 2.19 the intensity is written as a function of the field squared

\[
I_j = \frac{1}{2} \frac{n_j}{\varepsilon_0 c_0} |A_j|^2,
\] (2.20)

For OPA the initial condition is that the signal beam is weaker than the pump beam and gets significantly amplified in the process, while at the same time the idler beam is generated.

If we neglect the pump depletion \((A_p = \text{constant})\), assume there is an initial signal intensity \((A_{s0} = \text{seed beam})\), and no idler beam at the beginning \((A_{i0} = 0)\), the Manley-Rowe coupled equation is solved to get the signal and idler intensities. After a length L of the nonlinear crystal, assuming a large gain \((\Gamma L >> 1)\), the solution is:

\[
I_s = I_{s0} \left[1 + \frac{\Gamma^2}{g^2} \text{sinc}^2 (gL) \right] \rightarrow (\Gamma L >> 1) \rightarrow I_s = \frac{I_{s0}}{4} e^{2\Gamma L}
\]

\[
I_i = I_{i0} \left[1 + \frac{\Gamma^2}{g^2} \text{sinc}^2 (gL) \right] \rightarrow (\Gamma L >> 1) \rightarrow I_i = \frac{\omega_1 I_{i0}}{4\omega_s} e^{2\Gamma L}
\] (2.21)

\[
\Gamma^2 = \frac{\omega_s^2 \omega_{d eff}^2 |A_p|^2}{n_s n_p c_0^2} = \frac{2\omega_s \omega_{d eff}^2 I_p}{n_s n_p c_0^3 \varepsilon_0} = \frac{8\pi^2 d_{eff}^2 I_p}{n_s n_p c_0^3 \varepsilon_0 \lambda_s^\lambda_s}
\]

From the equation above, the parametric gain of the signal is defined as

\[
G = \frac{I_s(L)}{I_{s0}} = \frac{1}{4} e^{2\Gamma L}.
\] (2.22)
By using the intensity solution in Eq. 2.21, we can demonstrate the difference between OPG and OPA. QPM is used in PPLN as the nonlinear crystal choice and will be discussed later in the chapter.

![QPM-PPLN OPG/OPA](image)

Figure 2.3. The normalized intensity output phase matched bandwidth of OPG and OPA in PPLN.

Initially, collinear quasi-phase matching is considered in PPLN at a temperature of 92° C with a periodic grating constant of $\Lambda = 29.5 \, \mu m$. The OPA and OPG normalized output bandwidths are given in Fig. 2.3. Thus, when a pump laser at a wavelength of 1.064 $\mu m$ is co-aligned with an injected seed laser centered at a wavelength of 1.52 $\mu m$ with a 5 GHz bandwidth, the seed beam is amplified and generally retains the 5 GHz bandwidth. Now OPG is a situation where the pump spontaneously generates a broadband signal and idler frequencies that are dependent on phase matching.
2.4 Quasi-Phase Matching (QPM)

We can define the phase matching condition in a nonlinear crystal as

\[ \Delta k = k_1 - k_2 - k_3 . \]  

(2.23)

In the equation, the three wave vectors represent the pump (1), idler (2), and signal (3) respectively. In the case of QPM, a new vector is used to play a part in the phase matching process in a crystal. The implemented vector represents a grating period and is given as \( k_g \). The new phase matching vector is given as

\[ \Delta k_{\text{QPM}} = k_1 - k_2 - k_3 - k_g . \]  

(2.24)

The grating vector \( k_g \) is commonly represented with a grating period \( \Lambda \)

\[ k_g = \frac{2\pi}{\Lambda} . \]  

(2.25)

With the grating vector the frequency output of the nonlinear process can be controlled by adjusting the value of the periodicity. The QPM process was considered when calculating the intensity in Fig. 2.3. In the calculation a pump wavelength of 1.064 μm, an idler of 2.734 μm, and a grating period of 29.5 μm was used. We found that our signal wavelength phase matched approximately at 1.52 μm. Thus, we are able to control the frequency content by adjusting the grating period of the crystal used.

2.5 Periodically Poled Lithium Niobate (PPLN) OPA

PPLN has one of the highest nonlinear coefficients of the available nonlinear materials. It has a nonlinearity of \( d_{33} = 27 \text{ pm/V} \) [38]. A PPLN QPM grating contains several reversed domains that secure the phase matching condition. The reversal of the domains can be seen in Fig. 2.4.
QPM periodically flips the optical axis of the lithium niobate crystal in order to reset the phase of the nonlinear polarization and maintain a coherent buildup of the signal and idler wave conversion. For a wave propagating in the crystal, dispersion causes a phase mismatch. By using the domain structure, we can invert the energy flow for every instance when there is a phase change by $\pi$. The effect restricts the crystal interaction to one coherence length

$$L_c = \frac{\pi}{\Delta k}.$$  \hspace{1cm} (2.26)

If we were to attempt pump conversion without QPM the energy would be converted back to the original pump frequency after one coherence length. Thus, without QPM in PPLN the nonlinear interaction would be very inefficient.

In 1998, P. E. Powers et al. introduced a tunable seeded QPM OPG in PPLN [39]. Tunability was achieved by temperature tuning and translating a PPLN crystal with a single period. Similarly, we desire a tunable MgO:PPLN QPM for OPA as seen in Fig 2.5.
Figure 2.5. MgO:PPLN OPA with a multi-grating crystal.

We define the wave vectors for the ordinary and extraordinary wave vectors for our OPA process

\[ k_i = \frac{2\pi n_i}{\lambda_i}. \]  

(2.27)

When a pump source and injected seed signal wavelength is present, we can determine the QPM period from the phase mismatch

\[ \Delta k = k_p - k_s - k_i = 2\pi \left( \frac{n_x(\lambda_p, \theta, T)}{\lambda_p} - \frac{n_x(\lambda_s, \theta, T)}{\lambda_s} - \frac{n_x(\lambda_i, \theta, T)}{\lambda_i} - \frac{1}{\Lambda} \right). \]  

(2.28)

We can compensate for phase mismatching by choosing the desired grating period for the desired frequencies. For MgO:PPLN we use a type 0 phase matching technique. The Sellmeier equation for MgO:PPLN.

\[ n^2 = a_1 + b_1 f + \frac{a_2 + b_2 f}{\lambda^2 - (a_3 + b_3 f)^2} + \frac{a_4 + b_4 f}{\lambda^2 - a_5^2} - a_6 \lambda^2. \]  

(2.29)

In addition, we note that the temperature of the crystals plays a crucial in the phased matched output frequency due to the change in refractive index. The temperature effect is taken into account in Eq. 2.29 using
The desired parameters needed to calculate the indices of refraction in MgO:PPLN are given in Table 2.1 [40].

<table>
<thead>
<tr>
<th>Refractive Index</th>
<th>5% MgO: Doped PPLN</th>
<th>Parameters</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>nₑ</td>
<td>nₒ</td>
</tr>
<tr>
<td>A₁</td>
<td>5.756</td>
<td>5.653</td>
</tr>
<tr>
<td>A₂</td>
<td>0.0983</td>
<td>0.1185</td>
</tr>
<tr>
<td>A₃</td>
<td>0.2020</td>
<td>0.2091</td>
</tr>
<tr>
<td>A₄</td>
<td>189.32</td>
<td>89.61</td>
</tr>
<tr>
<td>A₅</td>
<td>12.52</td>
<td>10.85</td>
</tr>
<tr>
<td>A₆</td>
<td>1.32×10⁻²</td>
<td>1.97×10⁻²</td>
</tr>
<tr>
<td>B₁</td>
<td>2.86×10⁻⁶</td>
<td>7.941×10⁻⁷</td>
</tr>
<tr>
<td>B₂</td>
<td>4.7×10⁻⁸</td>
<td>3.134×10⁻⁸</td>
</tr>
<tr>
<td>B₃</td>
<td>6.113×10⁻⁸</td>
<td>-4.641×10⁻⁸</td>
</tr>
<tr>
<td>B₄</td>
<td>1.516×10⁻⁴</td>
<td>-2.188×10⁻⁶</td>
</tr>
</tbody>
</table>

Table 2.1 Parameters for the index of refraction in MgO:PPLN.

From the desired parameters, we can calculate the indices as a function of temperature and wavelength. The extraordinary and ordinary indices of refraction are given in Fig.2.6.

![Figure 2.6](image)

Figure. 2.6 The ordinary and extraordinary refractive indices for MgO:PPLN.

The desired tunable OPA with MgO:PPLN can now be calculated in terms of input wavelength, temperature, and periodicity. A single grating period of A=29.5 μm for type 0
quasi-phase matching was calculated as a function of temperature and wavelength for an OPA process. The result is given in Fig. 2.7.

The figure demonstrates that wide wavelength tunability can be easily achieved with temperature and overlapping ranges can be made by fabricating samples with different grating periods. We can consider the crystal to be z-cut, the axis of the highest nonlinearity $d_{33}$ aligned with the polarization of the incident pump source and seed signal. Due to the polarization orientation, the incident waves will couple to the most efficient nonlinearity. The result is a high efficiency for the single pass in the crystal [41]. The benefit of this type of system is that we can achieve the same level of efficiency as a complicated OPO scheme but with greater tunability than the OPO can produce. Therefore, we can negate the use of a complex cavity and use a simple single-pass or double-pass system to achieve our goal.
2.6 Etalon Seeded PPLN OPA

Even though the output of an OPG source is as efficient as the OPO method the high bandwidth is not acceptable for a narrowband IR source as an NDE evaluation tool. The only choice we are left with is OPA. Narrow-bandwidth OPA can be achieved by two different injection-seeding methods. The first is the well-documented OPG in band diode injection seeding. The seeding beam will be amplified by stimulating the different and amplify only one mode of operation. That mode will have a higher energy and will be narrow band because the seed source is narrowband. While this method has many benefits, the tunable range is limited to the range of the diode laser. Thus, in some situations seeding across the entire tuning range of the selected nonlinear crystal is not possible. In order to achieve a self-seeded effect an etalon filtered OPG can be used to seed a pump source to generate and OPA.

2.6.1 Overview of off-axis Fabry-Perot etalon feedback seed

The reduction in the overall linewidth of the converted OPG signal can be accomplished with an off-axis etalon. We can consider an etalon as two parallel reflecting surfaces separated by a distance d with an index between the two plates given as that of air (n=1) as seen in Fig. 2.8.

![Figure 2.8. An off axis Fabry-Perot etalon that gives rise to multiple beam interference in both the transmission and reflection orders.](image)
The phase of each transmitted wave differs from that of the proceeding wave by the
difference in their paths. Thus, the difference between the optical paths of \( E_1 \) and \( E_2 \) is
\[
\frac{n B_1 A_2 B_2 - B_1 C_1}{2 d} = \frac{2 \text{nd} \sin \theta}{\cos \phi} - \frac{2 \text{nd} \sin \phi \sin \theta}{\cos \phi} = \frac{2 \text{nd} \cos \phi}{\cos \phi} - \frac{2 \text{nd} \sin \phi (n \sin \phi)}{\cos \phi}.
\]
\[\text{(2.31)}\]

The total path difference between each successive transmitted pair corresponds to
\[
\delta = \left( \frac{\pi}{\lambda} \right) 2 \text{nd} \cos \theta.
\]
\[\text{(2.32)}\]

Assuming that the reflecting surfaces are identical, then the intensity of the light will reflect
with at a fraction \( R \). The relative amplitudes of the transmitted waves are
\[
\frac{I_R}{I_{\text{inc}}} = R.
\]
\[\text{(2.33)}\]

Therefore, the ratios of the reflected and transmitted electric field amplitudes are
\[
\frac{E_R}{E_{\text{inc}}} = -\sqrt{R}, \quad \frac{E_T}{E_{\text{inc}}} = \sqrt{1-R}.
\]
\[\text{(2.34)}\]

Now the transmission of the etalon can be calculated in terms of the electric fields at the
same instant of time \( t \). The procedure can be accomplished by initially starting with a plane
wave incident on the surface of the etalon.
\[
E(t) = E_0 e^{-i \omega t + \frac{2 \pi n}{\lambda} z}.
\]
\[\text{(2.35)}\]

The propagation direction is \( z \) for this case, thus at \( z=0 \) at the point \( A_1 \) of the input face, the
incident wave can be written as
\[
E(t) = E_0 e^{-i \omega t}.
\]
\[\text{(2.36)}\]
Using the fraction of the transmitted field in Eq. 2.34, we can determine the transmitted field amplitude after crossing the second surface

\[ E_1 = (1 - R)E_0 e^{-i(\omega t - k d/\cos \theta)} = (1 - R)E_0 e^{-i(\omega t - \delta_0)}. \]  

(2.37)

In the next state, the field is transmitted across the first surface and is reflected twice before being transmitted through the second surface

\[ E_2 = (1 - R)RE_0 e^{-i(\omega t - \delta_0)} e^{i\delta}. \]  

(2.38)

Thus, following the same approach the other orders can be found as

\[ E_3 = (1 - R)R^2E_0 e^{-i(\omega t - \delta_0)} e^{i4\delta} \]
\[ E_4 = (1 - R)R^3E_0 e^{-i(\omega t - \delta_0)} e^{i6\delta} \]
\[ \quad \vdots \]
\[ E_N = (1 - R)R^{N-1}E_0 e^{-i(\omega t - \delta_0)} e^{i(N-1)\delta} \]

(2.39)

The total transmitted field due to an incident laser beam is the total sum of all fields which can be assumed as an infinitely large number of reflections.

\[ E_T = E_1 + E_2 + E_3 + \cdots \]
\[ = E_0(1 - R)e^{-i(\omega t - \delta_0)}(1 + Re^{i2\delta} + Re^{i4\delta} + \cdots) \]
\[ = \frac{E_0(1 - R)e^{-i(\omega t - \delta_0)}}{1 - Re^{i2\delta}} \]

(2.40)

The time averaged intensity is proportional to the modulus squared of the field amplitude, therefore the transmitted intensity is given in terms of the incident wave intensity as

\[ I_T = \frac{I_0(1 - R)^2}{1 + R^2 - 2R \cos(2\delta)} = \frac{I_0(1 - R)^2}{1 + R^2 - 2R + 2R(1 - \cos(2\delta))} = \frac{I_0(1 - R)^2}{(1 + R)^2 - 4R \sin^2(\delta)}. \]  

(2.41)

Now we define that the coefficient of finesse \( F \)

\[ F = \frac{4R}{(1 - R)^2}. \]  

(2.42)
Thus, we can write the transmittance function of the etalon due to an off axis beam as
\[ T = \frac{1}{1 + F \sin^2(\delta)}. \] (2.43)

The transmitted intensity becomes a periodic function of \( \delta \) that varies between a maximum and a minimum as \( \delta \) changes. The transmission is given as a function of wavelength given in Fig. 2.9.

The finesse \( F \) describes the resolution of the etalon. The finesse determines the Full Width at Half Maximum (FWHM) of the transmitted peak, and is dependent on the reflectance of the mirrors. Thus, the narrower the linewidth of the transmitted intensity means that the cavity mirrors have a high reflection coefficient. The distance between mirrors determines the Free Spectral Range (FSR), which is usually expressed as a frequency.

![Etalon Transmission of a Laser](image)

Figure 2.9. Fabry-Perot transmission as a function of wavelength and reflection coefficient.
The wavelength separation between transmission peaks is determined from the FSR frequency of the etalon, which is

\[ v_{FSR} = \frac{\lambda_0^2}{2nd \cos \theta} \tag{2.44} \]

The finesse \( \xi \) of the cavity defined as

\[ \xi = \frac{v_{FSR}}{\delta_c} = \frac{\pi \sqrt{R}}{1 - R} = \frac{\pi \sqrt{F}}{2} \tag{2.45} \]

In Fig. 2.9 the transmission as a function of wavelength is given. The widths of the peaks get narrower as the mirror reflectivity increases, i.e. as the finesse increases. When these peaks get very narrow, light can only be transmitted if the plate separation \( d \), refractive index \( n \), and wavelength \( \lambda \) satisfy an integer multiple of \( p\pi \). Thus, this is the property that allows the etalon to act as a very narrow band-pass filter which is

\[ \delta = \frac{2\pi nd \cos \theta}{\lambda} = p\pi \tag{2.46} \]

For waves traveling at an angle \( \Theta \) with respect to the axis of the etalon, the transmission is

\[ T = \frac{1}{1 + F \sin^2 \left( \frac{2\pi nd \cos(\theta)}{\lambda} \right)} = \frac{1}{1 + F \sin^2 (\pi \cos(\theta) \frac{v}{v_{FSR}})} \tag{2.47a} \]

Maximum transmittance occurs at frequencies given by integer values: \( p=1, 2,... \)

\[ v_p = p \frac{c}{2d \cos(\theta)} = pv_{FSR} \sec(\theta) \tag{2.47b} \]

A plot of the relation given in Eq. 2.47b is given in Fig. 2.10.
Fig. 2.10. An off-axis etalon where the frequencies satisfy the peak transmittance condition of Eq. 2.47.

From Fig. 2.10, for incident light with a spectral width smaller than the free spectral range, each frequency component corresponds to one angle. Thus, the emission from the etalon can be used as a filter for a broad band OPG in which a single frequency can be guided from the etalon to injection seed a PPLN OPA. The bandwidth from a PPLN OPG is normally around 300 GHz. In the next chapter we will show that we can reduce the bandwidth to that of a diode laser bandwidth. In addition we will demonstrate that by tuning the angle of the etalon we can seed the OPA across the bandwidth of the OPG.
CHAPTER 3
NARROW BANDWIDTH TUNABLE OPA

3.1 Introduction

A narrowband optical parametric amplifier (OPA) enables a seed laser to be amplified in a nonlinear crystal. The non-seeded amplifier has a broadband gain spectrum and the process is called optical parametric generation (OPG). The narrow band OPA output can be tuned across the OPG bandwidth. An OPA at near infrared wavelengths has been used extensively for time resolved optical spectroscopy, multi-photon fluorescence microscopy and telecommunications [42]. The generation of coherent multi-wavelength, narrowband mid-infrared laser radiation is an essential component in the near future for spectroscopic applications. For example, anti-Stokes and stimulated Raman scattering spectroscopic measurements require tunable low noise IR laser sources [43].

The ability of nonlinear OPG to provide broadly tunable coherent radiation made it a key candidate for the development of a tunable IR source. The OPG has a simple configuration since it is a single-pass process and requires no optical cavity. However, the free-running bandwidth is typically too large for many applications in NDE [44]. The bandwidth may be controlled by injection seeding with a narrow bandwidth signal. Such narrowing has been demonstrated with diode lasers and by using the output of a filtered OPG [45]. Using a diode laser as the seed source is expensive and severely limited in wavelength tunability. The etalon filtered OPG approach has the advantage of being able
to generate a seed over the operating range of the OPG crystal. Previous filtered OPG setups consist of a two-crystal scheme, where the output of the first OPG is filtered and sent into the second crystal. This approach works well, but the addition of a second crystal also increases the cost of the system.

The proposed method presents an approach that allows us to use a single crystal that performs the same function as the two-stage system in the near to mid-wave infrared. By reducing the number of costly elements, this approach can become practical and viable for many applications, such as gas detection, medical diagnostics, and remote sensing [46].

The developed double-pass injection seeded OPA method is to reduce the overall complexity of such a system and extend the overall wavelength range beyond that which is capable with an OPO system. To the best of our knowledge a double-pass reflected etalon injection seeded OPA in PPLN crystal has not been configured to generate narrowband frequencies with wide tunability.

3.2 Narrow Bandwidth OPA

3.2.1. Dual Pass OPG method

Broad infrared bandwidths can be generated using MgO:PPLN OPG. Thus, the proposed method uses a double-pass pump laser scheme. On the first pass, a half-wave plate rotates the pump laser’s polarization such that a small component contributes to a phase matched OPG process. The first-pass OPG output is separated from the pump source beam path by a dichroic mirror to be used for recombining the injection seed with the pump later. On the first pass, the unconverted pump source is incident on a quarter-wave plate in order to change the polarization state of the beam to a circular polarization. The pump beam is reflected at the mirror and passes through the wave plate a second time. The result of such an interaction is similar to that of an electro-optic Q-switch; the pump polarization is
aligned with the axis with a large nonlinear coefficient. We can represent the interaction in terms of Jones vectors. The polarization of the a horizontal linearly polarized input beam can be switched to a vertical polarization state on the reflected output using a quarter wave plate with its fast axis rotated to 45 degrees as seen in Fig. 3.1.

![Figure 3.1. Reflection polarization rotator with the fast axis of a quarter-wave plate rotated to 45 degrees.](image)

Initially the input beam is incident on the rotated quarter wave plate and is given as

$$
\frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 & i \\ i & 1 \end{bmatrix} \begin{bmatrix} 1 \\ 1 \end{bmatrix} = \frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}.
$$

(3.1)

The beam becomes circularly polarized upon which it is then incident on a mirror. The difference between right or left circular polarization is only the based on the view point of the observer. Thus, if we unfold the cavity we can view the interaction between the circularly polarized beam and the mirror by the solution

$$
\frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 \\ i \end{bmatrix} = \frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}.
$$

(3.2)

The reflected circular polarized beam then is incident on the rotated quarter wave plate and the output is a linearly polarized beam in the vertical position

The beam becomes circularly polarized upon which it is then incident on a mirror. The difference between right or left circular polarization is only based on the viewpoint of the observer. Thus, if we unfold the cavity we can view the interaction between the circularly polarized beam and the mirror by the solution

$$
\frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 \\ i \end{bmatrix} = \frac{e^{-i\pi/4}}{\sqrt{2}} \begin{bmatrix} 1 \\ i \end{bmatrix}.
$$

(3.2)

The reflected circular polarized beam then is incident on the rotated quarter wave plate and the output is a linearly polarized beam in the vertical position
The overall effect of using such a scheme serves to convert the pump beam into a linear state that is vertically polarized on the second pass to ensure a maximum interaction with the component of PPLN with the highest nonlinearity of $d_{33}$. The configuration of such a system is given in Fig. 3.2.

Figure 3.2. First pass and second pass scheme for the narrowband generation of an OPA.

However, the OPG process is limited to the emission bandwidth due to the phase-matching condition for the crystal and the natural line width due to the interaction of the nonlinear material and beam. Generally, this has a resolution of approximately 300 GHz. In addition, the OPG bandwidth increases as the signal and idler are tuned towards degeneracy. Thus, by injecting a number of excess photons into the signal branch they are amplified to a degree much higher than the OPG background [47]. Injection of a small number of photons within a narrow bandwidth beam produces a narrowband OPA output as illustrated in Fig. 3.2.
We can achieve frequency tuning by using quasi-phase matching (QPM) techniques and the temperature tuning of the PPLN crystal with five different grating periods. With the implementation of a variable grating vector, the frequency output of the nonlinear process can be controlled by adjusting the value of the grating period ($\Lambda$) in the nonlinear crystal. The QPM equation for the pump, signal, and idler is written in terms of the extraordinary index $n_e$ for the given wave and is

$$\Delta k = k_p - k_s - k_i - \frac{2\pi}{\Lambda} = 2\pi \left( \frac{n_e(\lambda_p, T)}{\lambda_p} - \frac{n_e(\lambda_s, T)}{\lambda_s} - \frac{n_e(\lambda_i, T)}{\lambda_i} - \frac{1}{\Lambda} \right).$$

(3.4)

The correlation of the signal and idler occurred at different locations on the same phase matching efficiency bandwidth. The quasi-phase matched bandwidth for the multi-grating MgO:PPLN crystal is given in Fig. 3.3.

![MgO:PPLN Quasi-Phase Matched Bandwidth](image)

**Figure 3.3.** QPM bandwidth for MgO:PPLN at different periodicities and temperatures.
3.2.2 Experimental Setup

In order to injection seed to generate an OPA without using a costly tunable laser diode, an etalon was used. The first-pass OPG output can be filtered with a narrow-bandwidth Fabry-Perot etalon. This filtered signal is sent back through the same PPLN crystal, which is co-aligned along with the retro-reflected pump laser. This approach provides narrow line width and broad tuning. The configuration for the second PPLN pass is given in Fig. 3.4.

![Second Pass Diagram](image)

**Figure 3.4.** The configuration of the off-axis etalon injection seeded PPLN OPA.

The pump source for the experiment is a single frequency Q-switched ND:YAG laser operating at 1064 nm, delivering up to 740 μJ of energy per pulse, with pulses of 10 ns duration at a 30 Hz repetition rate. The pump laser was focused to a spot size (1/e field radius) of 0.4 mm. Prior to the focal region the pump laser passes through an optical isolator, half-wave plate, and a dichroic mirror that is highly transmissive at 1064 nm and highly reflective in the 1550 nm region. The half-wave plate was oriented such that the pump laser passing through the PPLN crystal is close to horizontally polarized. Note that
the PPLN z-axis is vertically oriented and hence the pump nominally has the wrong polarization for the PPLN’s designed quasi-phase matching. By rotating the waveplate, it is possible to control how much of the pump beam contributes to a quasi-phase matched interaction. Moreover, the laser polarization purity is not 100%, so that even when the half-wave plate rotates the pump to be horizontally polarized, a small portion of the laser is vertically polarized. The dichroic mirror was placed in the setup to redirect the return narrowband OPG beam. The PPLN is 5% MgO doped, 10 cm in length, 2 cm wide, 0.5 mm thick, and was placed so that the pump focus occurs in the middle of the crystal. The initial crystal-grating period used was \( \Lambda = 29.5 \mu m \) and the polished faces were AR-coated for all of the interacting wavelengths in the PPLN bandwidth. A 10 cm focal length lens placed 10 cm (1f) from the output of the PPLN crystal and served to collimate both the pump and OPG output. Following the lens is a second dichroic mirror that reflects the signal and transmits the pump. The transmitted pump passes through a quarter-wave plate such that its polarization state becomes circular. It is then retro-reflected and as a result, the sense of rotation is reversed. After passing through the quarter wave plate a second time, the beam is again linearly polarized, but orthogonal to the input. Hence, when the incident pump is horizontally polarized on the first pass, which does not quasi-phase match. The retro-reflected pump beam is vertically polarized, which will amplify the quasi-phase matched signal. The lens and mirror placement are roughly in a 4f configuration so that the returning pump laser has the same spot size at the same location as on the first pass. The reflected signal is transmitted through an off-axis etalon and then retro-reflected with a mirror. Hence, on the second pass through the crystal, the pump is properly oriented for quasi-phase matching and the filtered seed has a narrow bandwidth. We also note in the
results section that the retro-reflecting mirror placed behind the etalon is not necessary, and that we obtain narrow bandwidth feedback from the etalon when it is oriented slightly off-axis. The line width and FSR of the etalon were directly measured from the transmission of a narrow line-width diode laser operating at 1550 nm see Fig. 3.5. The etalon consists of two mirrors with a piezo-electrically controlled air-gap. The piezo material placed between the Fabry-Perot etalon mirrors expanded slowly with increasing voltage and changed the air-gap spacing [48]. By changing the air-gap spacing, we are able to tune the transmission of the etalon. The double-pass etalon FSR was measured to be approximately 1493 GHz with an FWHM of 1.41 GHz.

![Double Passed Etalon Transmission](image)

Figure 3.5. Measured transmission of a doubled pass etalon at the maximum FSR and minimum FWHM.

In order to demonstrate the tunability of the system over a broad range of wavelengths, the crystal temperature was tuned over the range of 20 ºC to 220 ºC by using a generic oven and an Omega temperature controller with a sensitivity of ±0.2 ºC.

The etalon filtered injection beam was recombined with the pump beam at the dichroic mirror for a second pass through the nonlinear medium. The output bandwidth
tunability was accomplished by adjusting the temperature of the crystal [49] and the bias control for the piezoelectric driven etalon. The added benefit was that the physical placement of the etalon did not have to be adjusted. The fine controlled bias auto aligned the injection seeder over the OPG signal bandwidth. Hence, manual tuning was not needed to produce the narrow bandwidth output signal. The final narrowband OPA laser beam was measured using a CVI monochromator with a line scan camera. The monochromator and camera combination allowed us to acquire a large spectral range in a single shot.

3.3 Results

We determined the energy conversion efficiency by comparing the phase-matched narrow band OPA output signal at a wavelength of 1550 nm with those of the input energy of the pump beam. The energy measurements were taken after the beam passed through multiple optical components at various stages for the forward pulse and the return signal. The measurements are provided in Fig. 3.6. The input energy from the pump beam in Fig. 3.6 (A) was measured to be 0.62 mJ prior to entering the PPLN crystal. After the crystal, 0.433 mJ emerged to interact with the optical devices. The losses incurred were due to the horizontal polarization of the beam and scattering from the crystal. The Fresnel losses incurred due to a single pass through the PPLN were measured to be approximately 30%. Roughly, 1% of the beam interacted with the crystal and produced a broadband OPG output. Fractional beam conversion occurred due to residual orthogonal polarization of the pump. The pump beam then traversed the optical devices and combined with the filtered narrow band reflection for a second pass through the crystal. The combined pump and narrowband beam energy was found to be 0.36 mJ. A loss of 0.073 mJ was a result due to the pump beam transmission through the second dichroic mirror. For successful PPLN
OPG injection seeding, the energy threshold for pulses in the nanosecond range is less than 10 pJ [50]. In Fig. 3.6 (B), the etalon filtered injection beam had sufficient energy to combine with the 0.36 mJ pump beam and injection seed the OPG. The final output energy of the narrowband OPA was measured to be .09 mJ. The final signal conversion efficiency was calculated to be approximately 14.52%.

Figure 3.6. Narrow Bandwidth OPG Source operating at 1550 nm (a.) Input energy Loss (b.) Output Energy.

Figure 3.7. (a.) Mirror Seeding (b.) Etalon-Mirror Seeding (c.) Etalon Seeding.
We note that the mirror placed behind the etalon was not needed to effectively injection seed and generate the narrowband OPA. This was due to the fact that the back mirror in the etalon was partially coated for SWIR.

In Figure 3.7 three schemes are depicted. Injection seeding with a mirror, etalon-mirror combination, and an etalon without an external mirror. To demonstrate the injection seeding of the OPG source, a signal band centered at 1542 nm was selected. The three seeded outputs from the configurations depicted in Fig 3.7 were measured and are given in Figure 3.8. In Fig. 3.7 (A), when the mirror was used without the etalon, a broadband output was generated. In Fig. 3.7 (B), a narrowband OPG beam was generated when we used the mirror-etalon combination and the same narrow bandwidth was obtained when the etalon was used without the retro-reflecting mirror (Fig. 3.7 (C)). The etalon injection seeder was tunable over the entire signal bandwidth of approximately 350 GHz.

![OPA Etalon Injection Seeding Comparison](image)

Figure 3.8. Narrowband OPG injection seeding with a mirror, etalon-mirror combination, and with an etalon alone.

To demonstrate the effective tunability of the OPA source a signal band centered at 1549 nm was selected. Multi-wavelength narrowband OPA was obtained by controlling
the seeding periodically at discrete wavelengths. An etalon scanning rate of 50 ms was used, and the output analysis was performed simultaneously using a monochromator. Fig. 3.9 depicts narrowband images with the injection seeder tuning over the PPLN signal bandwidth.

![Image of OPA Injection Seeded Etalon Scan](image)

**Figure 3.9.** Spectrum measurements of the narrowband OPG laser scan, centered at 1549 nm, and tuned over the whole signal band.

Fig. 3.9 shows that optimal seeding and intensity occurs when the injection beam is aligned with the center of the pump beam. Out of band seeding generated a narrowband OPA plus a broadband background. We note that it is possible to synchronize the etalon tuning with temperature tuning or grating tuning. Multiple signal bandwidths were then selected to exhibit the broad tunability of our narrowband OPA source. The crystal temperature was periodically increased in increments of approximately 40°C over a range starting from 20°C to 220°C. For incident waves traveling at an angle Θ with respect to the axis of the etalon, the double-pass transmission can be calculated in the form of an integer multiple π. In which the etalon acts as an interferometer where single wavelengths can be
selected and is illustrated in Fig. 3.10 (a). The result is that the orientation of the etalon can be tuned to sweep across the bandwidth of the OPG resulting in a narrowband OPA with a bandwidth on the order of the FWHM of the tuned peak from the etalon which is given in Fig. 3.10 (b).

![Figure 3.10](image)

Figure 3.10. Illustration and measurement of (a) double-pass etalon tuning and (b) OPA injection seeding centered on multiple OPG bandwidths.

### 3.4 Application

The narrowband OPA source key performance features were evaluated using the known absorption features of carbon dioxide. The key features entailed the seeded OPG line width and the effective tunable range of our source. The experimental setup for our static cell CO₂ measurements is given in Fig. 3.11. The output of the seeded OPG was directed to a multi-pass gas cell, which provides a path length of 10 m. The transmitted output was measured with an extended response Germanium detector. The narrowband generator was operated over a narrow range from 1582 nm to 1584 nm.
We tuned the seeded OPG over four CO\textsubscript{2} absorption features by holding the PPLN crystal temperature fixed at 150°C and ramp modulated the etalon to scan at a 10 s rate.

Figure 3.11. The experimental setup to measure the absorption features of CO\textsubscript{2}.

In Fig. 3.12 we graphically depict a known mid-IR transmission band of carbon dioxide which was generated with HITRAN [51]. The line strengths were plotted using a 10 m path length at a pressure of 70 Torr.

Figure 3.12. The absorption of pure CO\textsubscript{2} in the IR spectrum simulated with HITRAN. The data was simulated with a laser path length of 10 m, a temperature of 296 K, and at a pressure of 70 Torr.
The line shape was simulated using a Voigt profile. In order to match the simulated conditions our static gas cell in the experimental setup was evacuated to 70 Torr with pure CO$_2$. A path length of 10 m was used at a temperature of 296 K. The absorption bands centered on 1582 nm were conveniently located in the tunable range of our narrowband generator and thus were used for our measurements of a subset of the CO$_2$ features.

Figure 3.13 represents the absorption spectrum (about 2 nm wide) of the seeded OPG source. The static cell absorption spectrum was overlain with the calculated Hitran profile. In the figure, the measured absorption lines matched the known features for CO$_2$. From our measured CO$_2$ data we determined that the measured line width of the CO$_2$ lines were $\Delta \lambda = 0.044$ nm (5.4 GHz). The seeded OPG linewidth is less than 5.4 GHz since the Doppler-limited linewidth of the CO$_2$ lines is 3.35 GHz. The measured features are a convolution of the seeded OPG linewidth and the CO$_2$ line shape. The effective tunable range of our source for a single signal bandwidth was measured to be 287 GHz.

![CO$_2$ Absorption Peak Measurement](image)

Figure 3.13. Carbon dioxide absorption features measured with etalon injection seeded OPA.
3.5 Conclusion

A narrowband OPG source has been developed and demonstrated. In the experiment, a conventional diode laser injection seeder was replaced by an off axis Fabry-Perot etalon. With an off-axis etalon, we were able to injection seed an OPG resulting in an energy of 90 μJ in the signal with conversion efficiency of 14% from the pump to the signal. We have measured a small range of the CO₂ absorption spectrum, compared the transmission spectrum with the known absorption features, and found the result to be in excellent agreement. The measured linewidth of the narrowband OPG laser was found to be less than 5.4 GHz with an effective tunable range of 287 GHz at a fixed temperature. Much broader tunability is possible by changing temperature or QPM periodicity. The use of an off axis etalon serves as a viable option as a replacement for laser diode injection seeding in order to make a tunable narrowband OPG laser. The narrow bandwidth of the system makes it a suitable option for NDE applications that require a high resolution source.
CHAPTER 4
DIFFERENCE FREQUENCY GENERATION IN DAST

4.1 Introduction

The material 4-dimethylamino-N-methyl-4-stilbazolium-tosylate (DAST) is a crystalline structure first reported by Marder et al [52]. DAST has attracted a lot of attention in the nonlinear optics community because it possesses one of the highest known nonlinear optical coefficients, e.g. $d_{111} = \frac{1}{2} \chi_{111} = 260 \text{ pm/V}$ [53]. In this chapter a brief summary is given that will cover important linear and nonlinear material properties of DAST and show its applicability for the efficient difference frequency generation of THz radiation.

4.2 Overview of THz Generation by DFG

In Chapter 2 we introduced a few of the nonlinear interactions of interest. DFG is represented by the nonlinear polarization component

$$P(\omega_s - \omega_s^*) = 2\chi^{(2)}A_s A_s^*.$$  (4.1)

In this type of interaction a new frequency component arises from the difference of two input frequencies. We can use this process to generate tunable far infrared radiation by holding one frequency constant while increasing or decreasing the other. Another added benefit is that the generated THz waves will have a linewidth that is comparable to the sources used. From Eq. 4.1 we note that the DFG process is nearly the same as the OPA process. However, in this situation a weak seed is not used. The two signal wavelengths
that are mixed in the nonlinear crystal that have the same magnitude where one frequency is not in the OPG bandwidth. The DFG interaction can be viewed in Fig. 4.1.

![Diagram of Difference Frequency Generation (DFG) in $\chi^{(2)}$ material. The second frequency is amplified similar to OPA while the THz frequencies are generated.](image)

In the diagram, for every amplified photon $\omega_{s_2}$ a higher frequency photon $\omega_{s_1}$ is destroyed. Due to conservation of energy between the three frequencies, the THz frequency is generated as the difference between the two signal inputs.

**4.3 DFG Wave Equation Overview**

In the onset, two wavelengths (two signal beams) are present and we are to determine the difference frequency between the two beams. The model becomes complex due to the large attenuation associated with THz generation. Thus, loss due to the type of crystal used must be taken into consideration. Initially, we substitute a nonlinear component of the polarization at the difference frequency $\omega_3$

$$
P^{\text{nl}}(\omega_3 = \omega_1 - \omega_2, z, t) = \varepsilon_0 (2d_{\text{eff}}) E_1(z) E_2(z) \cos[(\omega_1 - \omega_2)t - (k_1 - k_2)z] . \tag{4.2}
$$

Similar to OPA we can solve the M-R equations and determine the electric field amplitude just inside the exit face of the nonlinear crystal for the difference frequency
\[ E_3(L) = \frac{E_0 \Omega_3}{\varepsilon_3} (\frac{\mu_0}{\varepsilon_3})^{1/2} (2d_{\text{eff}}) E_1 E_2 e^{-i(\Delta kL)} - 1 \Delta k. \]  
(4.3)

The output intensity of \( \omega_3 \) is related to the square of the field

\[ I_{\omega_3} = \frac{1}{2} \left( \frac{\varepsilon_3}{\mu_0} \right)^{1/2} E_3 E_3^*. \]  
(4.4)

By substituting the field equation into the intensity equation the output intensity is found as

\[ I_{\omega_3} = \frac{1}{2} \left( \frac{\mu_0}{\varepsilon_0} \right)^{1/2} \frac{\omega_3^2 (2d_{\text{eff}})^2}{n_1 n_2 n_3 c^2} I_{\omega_1} I_{\omega_2} \frac{\sin^2 \left( \frac{1}{2} \Delta kL \right)}{(\frac{1}{2} \Delta kL)^2}. \]  
(4.5)

The output intensity can be rewritten as the output power in terms of the input power

\[ P_{\omega_3} = \frac{1}{2} \left( \frac{\mu_0}{\varepsilon_0} \right)^{1/2} \frac{\omega_3^2 (2d_{\text{eff}})^2 L^2}{A n_1 n_2 n_3 c^2} P_{\omega_1} P_{\omega_2} T_1 T_2 T_3 \frac{\sin^2 \left( \frac{1}{2} \Delta kL \right)}{(\frac{1}{2} \Delta kL)^2}. \]  
(4.6)

In Eq. 4.6, \( T_1, T_2, \) and \( T_3 \) are the surface power transmission coefficients at the frequencies \( \omega_1, \omega_2, \) and \( \omega_3. \) In the equation the beam cross section is assumed to interact with the area \( A \) of the nonlinear crystal. The output power can be put into its final form by considering the effects of phase matching \( \Delta k \) (4.7), the losses due to absorption \( \alpha \) (4.8), and the transmission coefficients \( T \) related to each specific index of the nonlinear crystal (4.9).

\[ \Delta k = k_{s1} - k_{s2} - k_{\text{THz}} = 2\pi \left( \frac{n_e(\lambda_{s1}, \theta)}{\lambda_{s1}} - \frac{n_e(\lambda_{s2}, \theta)}{\lambda_{s2}} - \frac{n_{\text{THz}}(\lambda_{\text{THz}}, \theta)}{\lambda_{\text{THz}}} \right), \]  
(4.7)

\[ \Delta \alpha = \alpha_1 - \alpha_2 - \alpha_{\text{THz}}. \]  
(4.8)

\[ T = \frac{4n}{(n + 1)^2}. \]  
(4.9)
Thus the final form of the estimated output power due to DFG in a nonlinear crystal can be written as

\[ P_3 = S e^{-\alpha_m L} \left( 1 + e^{(-\Delta\alpha L)} - 2e^{(-\frac{1}{2}\Delta\alpha L)} \cos(\Delta k L) \right) \frac{(\Delta k L)^2 + (\frac{1}{2}\Delta\alpha L)^2}{(\Delta k L)^2 + (\frac{1}{2}\Delta\alpha L)^2}. \] (4.10)

\[ S = 32 \left( \frac{\mu_0}{\varepsilon_0} \right)^\frac{1}{2} \frac{2\omega_3 d_{\text{eff}} L}{(n_e(\lambda_1, \theta) + 1)(n_e(\lambda_2, \theta) + 1)(n_e(\lambda_3, \theta) + 1)} \left( \frac{1}{c^2} \right) \left( \frac{P_{\omega_3} P_{\omega_2}}{A} \right)^2. \]

4.4 DAST Properties

DAST is an organic salt made of a cation stilbazolium and the anion tosylate in which its structure is given in Fig. 4.2. The crystal orientation is achieved by Coulomb attraction between the cation and anion. Stilbazolium is a very active nonlinear chromophore and the tosylate acts as a medium for non-centrosymmetric crystal orientation [53].

![Molecular diagram of active nonlinear chromophore cation stilbazolium and anion tosylate.](image)

Figure 4.2. Molecular diagram of active nonlinear chromophore cation stilbazolium and anion tosylate.

The crystalline form of DAST is monoclinic crystal and belongs to the point group m. This type of point group is written in reduced form

\[
\begin{bmatrix}
0 & d_{12} & d_{13} & 0 & d_{15} & 0 \\
0 & 0 & 0 & d_{24} & 0 & d_{26} \\
0 & d_{31} & d_{32} & d_{33} & 0 & d_{35} \\
0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix}
\] (4.11)

The matrix for DAST can be simplified because the polarization of the incident mixing fields is in the extra-ordinary index. Therefore only along the x axis or a combination of
the x and y axis will be considered. Thus, the z axis interaction can be neglected. The matrix representing the nonlinear interaction can be written in a tensor format of the polarization

\[
\begin{bmatrix}
    d_{11} & d_{12} & d_{13} & 0 & d_{15} & 0 \\
    0 & 0 & 0 & d_{24} & 0 & d_{26} \\
    d_{31} & d_{32} & d_{33} & 0 & d_{35} & 0
\end{bmatrix}
\]

In Eq. 4.12 all interactions that contain \( A_z \) is zero. The polarization of both frequencies is taken as identical. The \( d_{\text{eff}}(\theta, \phi = 0) \) coefficient for the DFG polarization in DAST becomes a function of angle. All frequencies are \( \hat{e} \) polarized (extraordinary), where \( \theta \) is the angle between the \( k \)-vector and the propagation z axis of the incident fields

\[
A_{01=02} \hat{e} = A_{01=02} \left[ -\cos(\theta + \phi)\hat{x} + \sin(\theta + \phi)\hat{y} \right].
\] (4.13)

The d coefficients of interest are given for \( d_{111} = 260 \text{ pm/V}, \ d_{122} = 40 \text{ pm/V}, \ d_{266} = 39 \text{ pm/V}, \) and \( d_{311} = d_{322} = 2 \text{ pm/V} \) [54, 55, 56, 57]. In order to estimate the nonlinear optical conversion of DAST, the refractive indices need to be evaluated. The dispersion of the refractive indices is given by the Sellmeier equation [54].

\[
n^2(\lambda) = n_0^2 + \frac{q\lambda_0^2}{\lambda^2 - \lambda_0^2}.
\] (4.14)

In Eq. 4.14 \( \lambda_0 \) is the resonance wavelength and \( q \) is the amplitude of the oscillator. Other oscillators are taken into account that add or subtract from the index generated by the main oscillator at resonance. These oscillators are taken into account by the parameter \( n_o \). The parameters that described the index of refraction for 3 different axes are given in table 4.1 [58]. DAST is an anisotropic material with a large difference in refractive index that is dependent on the orientation of the incident wave. In order to estimate the phase matching
efficiency and power conversion in DAST the indices and absorption spectrums in the SWIR and THz regions are needed.

<table>
<thead>
<tr>
<th>DAST INDEX</th>
<th>$n_1$</th>
<th>$n_2$</th>
<th>$n_3$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Q</td>
<td>1.645</td>
<td>0.469</td>
<td>0.234</td>
</tr>
<tr>
<td>$n_0$</td>
<td>2.078</td>
<td>1.585</td>
<td>1.565</td>
</tr>
<tr>
<td>$\lambda_0$</td>
<td>533.3</td>
<td>503.5</td>
<td>501</td>
</tr>
</tbody>
</table>

Table 4.1. Parameters for DAST SWIR indices of refraction.

The indices of refraction and the absorption coefficients for SWIR are given in Fig. 4.3 and Fig. 4.4. Strong absorption bands can be seen for the three axes approximately centered at a wavelength of 1650 nm. One of the features that make DAST a key nonlinear material is that in the wavelength range we are to use for DFG THz wave generation is below 1600 nm, and the absorption is less than $2 \text{cm}^{-1}$.

![Index of Organic Crystal DAST](image)

Figure 4.3. Refractive indices of DAST.
Figure 4.4. Absorption coefficients for DAST in SWIR regime.

The indices of refraction and absorption for the DAST crystal b-axis in the FIR was first reported by Walther et al in 2000 [59]. The other index of refraction that we are interested in is along the a-axis of the crystal which is given in Fig. 4.5 and the absorption is given in Fig. 4.6 [60].

Figure 4.5. Index of refraction frequency dependence along the a-axis of the DAST crystal.
4.5 DAST THz Model

In normal DFG calculation different types of phase matching processes are generally used. Usually, two sources are mixed in the DFG crystal that are a combination of extraordinary and ordinary waves. However, the THz generation method through DFG in DAST can be accomplished using type 0. Type 0 phase matching is where the polarization of both input waves are the same and will phase match. This occurs because of the large resonance in the refractive between the mid-IR and the FIR which can be seen in Fig. 4.5. Using the components of the dispersion and refraction in the DAST crystal we can calculate the phase matching efficiency and power generation as a function of polarization angle and frequency. The phase matching efficiency is given in Fig. 4.7. The figure depicts where the DAST phase matches efficiently and provides insight as to where we should expect our signal as a function of polarization angle and frequency. In Fig. 4.7 one of the signal wavelengths was held constant at $\omega_1=1560.8$ nm and the other signal wavelength was tuned from $\omega_2=1380$ nm to 1559 nm.
In the model $\lambda_{\text{THz}}$ is estimated from energy conservation for each of the angular phase matching frequencies $\omega_{s1} = \omega_{s2} + \omega_{\text{THz}}$ using Eq. 4.10. Therefore, one can estimate the DFG phase matched output power because the indices and absorption parameters are known over the desired spectral range. In the simulations we considered the nonlinear process where the three wavelengths are coupled via the $d_{111}$ nonlinear coefficient in the nonlinear tensor of DAST and when the linear polarization of the beam is rotated off this component. The simulated DFG output power as a function of frequency and polarization is given in Fig. 4.8.

![Figure 4.7. DAST DFG tuning bandwidth. The region that is red signifies where phase matching is most probable.](image_url)
Using the same wavelength parameters that were given for phase matching, assuming that each of the signal wavelengths are at an average power of 0.5 W, both with the same polarization, and with an identical beam waist of 0.15 mm. We can achieve a maximum power of 0.2 mW at key frequencies. It is of note that at lower frequencies at least 0.02 mW is still theoretically achievable. In addition, the DAST crystal should provide a large tuning band of up to 28 THz. Experimentally the linewidth of the generated THz waves should closely follow linewidth of the combined signal waves. Therefore, if we use two narrow linewidth signals for DFG in DAST, we can generate a tunable narrowband THz source for NDE inspection.

![Figure 4.8. Output map of DAST DFG as a function of combined wavelength polarization angle and frequency.](image)
CHAPTER 5

NARROWBAND THZ DFG VIA DUAL SEEDED OPA

5.1 Introduction

In the pursuit of developing new THz technology several schemes are available to generate and to detect THz radiation. THz research has been a fruitful field for both theoretical and experimental explorations [61]. One of the primary techniques involves terahertz time-domain spectroscopy (THz TDS), a method which relies on electromagnetic transients using picosecond laser pulses. Several novel methods for the measurement of such waves have become practical. These methods include THz pump probe measurements, THz spectroscopy, and THz correlation spectroscopy. These sub-millimeter waves have been harnessed for applications in imaging, sensing, and molecular spectroscopy. Its unique position in the electromagnetic spectrum makes it a subject of studies exploring uses in the fields of security, medicine, and nondestructive inspection. In the pursuit of developing new THz technology several schemes are available to generate and to detect THz radiation.

The developed method utilized dual OPA seeded MgO:PPLN crystals in order to perform difference frequency generation to extend the overall THz spectrum tunability. The primary goal was to develop a narrowband THz source than can scan from 1-28 THz while maintaining adequate power for spectral imaging. In this chapter we will describe our setup to generate THz waves using the techniques covered in the previous chapters. In
addition, we will present measurements in this frequency regime and compare them to our generated models of THz DAST DFG.

5.2 THz DAST DFG

5.2.1 Tandem Seeded MgO:PPLN OPAs

In our DFG method two detuned signal beams are co-aligned and passed through the nonlinear crystal DAST of thickness L. The index of refraction for the crystal in the optical and THz region are very similar [62]. Type-0 collinear phase matching can occur when all waves are vertically polarized with respect to the z cut of the crystal which results in the down conversion from the optical regime to THz frequencies. In order to generate the two widely tunable signal beams a single pump frequency is equally divided and passed through two different types of MgO:PPLN crystals. In each PPLN crystal the pump frequency is spontaneously converted to a signal and idler pair due to the inherent nonlinearity at the selected index [40]. The generated signal idler pair is a result of quasi-phase matching which is dependent on factors such as crystal temperature, PPLN poling, and pump frequency. QPM was selected over traditional birefringent phase-matching (BPM) because the output beams are naturally co-aligned. QPM periodically re-orientates regions of the lithium niobate crystal in order to reset the phase of the nonlinear polarization and maintain a coherent buildup of the signal and idler wave conversion. QPM with high conversion efficiency can be maintained over a large spectral range [63]. Here, we consider the case for two independent MgO:PPLN crystal-grating patterns. The first PPLN type is that of a multi-grating periodicity discussed in Chapter 2. Due to the constraints on the available injection diode seeding lasers covering the range of 1380 nm to 1570 nm and our available Nd:YAG pump laser, the choice of grating period spanned the range from about
27 to 31 μm. Thus, OPA wavelength tuning in the multi-grating would be a result of grating period and temperature tuning.

Properly chosen multi-grating PPLN samples permit the correct phase matching parameters necessary for injection seeding. However, certain drawbacks occur if both OPG stages were to implement a crystal with a single period; both signal wavelengths would be constrained by the availability of a few grating periods and require large temperature changes. This approach would prevent continuous narrowband tunability. In order to achieve seamless tunability a PPLN crystal with a continuous grating periodicity (fan out) was used [64]. A single temperature was selected and the crystal was translated to achieve wavelength tunability. The QPM results for the fan out grating period versus signal wavelength is given in Fig. 5.1.

Figure 5.1. Fan out grating period and quasi-phase matched signal range for PPLN crystal with temperature variations.
5.2.2 Experimental Setup

A schematic of the experimental configuration is given in Fig. 5.2. In the diagram, two MgO:PPLN OPG crystals are used in the DFG mixing process. Each crystal is 5 cm long and is 1 mm thick. One MgO:PPLN crystal was grated with a multi-grating pattern. Temperature tuning of the generated spectral range was matched to the seed laser wavelength. The secondary stage was an MgO:PPLN with a fan out grating pattern of 25° which allowed continuous bandwidth tuning by position without temperature tuning. The temperature of the multi-grating PPLN was varied for a variety of wavelengths while the fan out crystal was maintained at a temperature of 120°C.

![Figure 5.2. Experimental setup used to generate a narrowband THz wave. The dual injection seeded OPG is mixed in the DFG crystal DAST.](image)

The pump source of each stage was an Nd:YAG of single wavelength at 1.064 μm. The source was Q-switched at 10 kHz with a pulse width of 1 ns. The pump beam was vertically polarized using a half-wave plate with respect to the z-cut MgO:PPLN crystals. The pump source was collimated using a 16 cm focal length lens and equally divided with a beam-splitting cube. Each beam was projected through each MgO:PPLN crystal and focused to
a beam waist \( w \) of 0.2 mm in the center of the crystals (\( 1/e \) intensity diameters). The fan out crystal was advanced to maintain an identical path length and phase. Similar performance was measured from each stage. The generated signal wavelength power from each stage was 0.8 W and an average energy per pulse of 80 \( \mu \)J.

Diode laser injection seeding was used to narrow the bandwidth of the broadband signal wavelengths. Injection seeding a nanosecond pulsed OPG in MgO:PPLN requires less than 1 mW \[^{[6]}\^5\]. The diode lasers were operated at a power of 4 mW and achieved sufficient seeding power to bandwidth convert each OPG. All diode lasers were vertically polarized with respect to the z-cut MgO:PPLN crystals. Three different diode lasers were used as injection seeders in the experiment. One laser was able to tune over a range of 1.47 \( \mu \)m to 1.54 \( \mu \)m. Another was able to tune from 1.52 \( \mu \)m to 1.57 \( \mu \)m. The last was able to tune from 1.38 \( \mu \)m to 1.47 \( \mu \)m. The multi-grating MgO:PPLN was maintained at a single desired frequency while the fan crystal was scanned over the full range of each of the diode lasers. The injection seeded output bandwidth from each stage was 1.8 GHz, which was comparable to each other and is given in Fig. 5.3.

The THz frequencies of interest were generated by mixing the output signal waves from the OPA’s in the DAST crystal. The output of each OPA stage was collimated and co-aligned. During the overlap process, the unconverted pump and idler wavelengths were removed from the system using frequency selective mirrors. The co-aligned beams were then passed through a half-wave plate in order to control the polarization angle of the combined linearly polarized beams.
Figure 5.3. The multi-grating OPA and the fan out grating OPA. The dual outputs are both narrowband and tunable to provide the necessary frequency separation to generate THz waves.

Figure 5.4. FTIR transmission measurements of materials to be used as filters for SWIR signal wavelengths.
To achieve optimal mixing in DAST, the combined narrowband OPA beams were focused in the crystal with a beam waist of 0.15 mm (1/e intensity diameters). The emitted THz beam was then filtered from the unconverted signal beams. The spectral variety of materials were examined using an FTIR Bruker and evaluated as possible filters for the separation. The measured FIR transmission for various materials are given in Fig. 5.4.

The filtering process was performed using either a high band pass germanium filter (HBP) or a low band pass (LBP) germanium filter in combination with a silicon coated germanium crystal (see Fig. 5.2). These filters were chosen because they permitted the minimal THz absorption while providing maximum absorption of the unconverted signal wavelengths. The generated THz beam was then collimated using an off-axis parabolic mirror and focused on a single point pyro-electric detector. The pyro-electric detector sensitivity was measured to be 8.1×10^4 V/W at 632 nm. The sensitivity of the detector was not measured at THz frequencies due to the limited access to a tunable THz source with constant amplitude. We assume that the sensitivity will be no greater in the FIR. The pyro-electric detector was then synchronized with a chopper at a 7 Hz frequency using a lock-in amplifier. The chopper was placed directly after the output of MgO:PPLN multi-grating crystal. In addition, a power meter was used in the final examination of the system in place of the pyro-electric detector. In the final configuration three frequencies were selected and measured at peak output levels in which just one filter was used (HBP or LBP).

The verification and the bandwidth of the generated THz waves were confirmed by the propagation in the atmosphere. The bandwidth of the THz DFG is dependent on the combined bandwidth of the two OPA signals. However, we estimate the THz bandwidth over a 3 m path length in air. The measurement was performed by comparing the known
water-absorption features that are known to be as narrow as the linewidth of the generated THz beam.

5.3 Measurements

The method used to identify that THz waves were generated was done through material finger printing. The THz waves were compared with the known water vapor absorption in the atmosphere at a given temperature and pressure. The calculated frequency due to the interaction of the mixed OPA signals resulted in less than 2 cm\(^{-1}\) difference from the known THz absorption features. The dual OPA source wavelengths were tuned to generate a frequency bandwidth between 1.5 THz and 27 THz. The DAST DFG THz wave was evaluated in three ways. Initially the generated THz waves were passed and filtered using a two filter process.

![THz Propagation Absorption](image)

Figure 5.5. Comparison of measured DFG system transmission with the known THz transmission spectrum in air at a 3 m path length at STP. No background corrections were made to the measured signal data. Combinations of LBP Ge, HBP Ge, and stock germanium filters were used. The configuration served to reduce the unconverted signal power on the pyro-electric detector.
to a minimum. However, the method described also increased the overall absorption of the
THz beam incident on the detector. The aforementioned method was implemented to
partially sample a small range of the developed system. A comparison of the measured
transmission was made with the known THz transmission spectrum in a range of 3 THz to
8.5 THz. The known transmission spectrum was generated at STP for a 3 m path length
using HITRAN data [51]. The comparison between our experimental signal and HITRAN
data is illustrated in Fig. 5.5.

For the measurements the seed laser wavelengths were recorded. During the progression
of the scan the frequency was calculated and the measured THz signal was plotted as a
function of frequency. When compared to the model calculations a good correspondence
was noted between the expected peaks and valleys in the transmission spectrum. Thus, the
generated narrowband THz spectrum by means of the difference frequency process in
DAST was validated. In addition, by measuring the narrowband water absorption we were
able to estimate the linewidth of our system. The linewidth of the injection seeded OPG
signal was approximately 1.8 GHz. Therefore, the expected linewidth of the generated THz
beam should be on the same order. The measured features of the recorded spectrum is a
convolution between the generated THz transmission beam and the water vapor line shape.
The measured linewidth of our system was calculated to be 3.1 GHz. Thus, such as system
has a narrow bandwidth with a large dynamic range, which has distinct advantage over the
conventional broadband OPO systems [66]. A full spectrum scan for our system was
recorded and is displayed in Fig. 5.6. Several OPA tuning frequency and temperature
ranges were selected.
To generate the desired wavelength range the MgO:PPLN multi-grating was held at a single output frequency while the fan crystal was swept across the IR spectrum to cover the desired tuning range. In addition, the spectral bandwidth remained around 3 GHz throughout the entire tuning wavelength range. The measured transmission spectra was generated using five overlapping spectral ranges.

![Measured DAST DFG Spectrum](image)

Figure 5.6. Generated THz transmission spectrum from 1 THz to 27 THz. The scan is presented without background correction and plotted using five different scans with partially overlapping scanning ranges and different filter combinations. The data was continuously recorded as the fan out MgO:PPLN crystal was tuned and scanned across the desired spectrum at a 3 GHz linewidth.

The variations of the power in the transmission spectrum that are observed in Fig. 5.6 are due to the phase miss-matching and absorption that is inherent to the DAST crystal. In addition, multiple peaks and valleys occur in the spectrum, which can be attributed to the water vapor absorption due to the propagation of THz in air. The observed large signal dips such as those recorded at 20 THz are due to strong phonon absorption in DAST [67]. This strong phonon absorption could not be compensated for due to the intrinsic phase matching.
limitations of the DAST crystal which can be gauged by comparing the measured signal to our developed model. We can control the output of generated signal by change the polarization orientation of the combined OPA signals. By keeping the polarization state of both pump sources identical, rotating them onto the a-b axis of the DAST crystal, we can estimate the signal strength and overall waveform of the DFG as a function of polarization angle. In Fig. 5.7 we tuned our system to generate the desired frequency content in 0.15 nm increments and rotated the polarization angle of the combined pump in 5° steps. The result was that a threshold for the THz signal generation occurred around 35°, which was to be expected.

![DAST THz DFG OPA Polarization Angle](image)

Figure 5.7. Measured DAST DFG signal as a function of combined OPA polarization angle and frequency. No background correction was performed and the data was recorded at increments of \( \Delta \lambda = 0.15 \) nm.

Since we were able to detect variations in the output signal level as a function of polarization angle two separate criteria was made for our model comparison. A range of 3.8-12 THz and 12-26 THz was selected for the model comparison. Comparable power for
both wavelengths were selected for mixing in the DAST crystal and power was 0.4 W for each.

The measured signal and the modeled signal over the desired frequency range as a function of combined OPA polarization angle are given in Fig. 5.8 and Fig. 5.9.

Figure 5.8. Output mapping of DAST DFG signal from 3.8 THz to 12 THz. (a) Modeled DFG power as a function of polarization angle and frequency. (b) Measured DFG signal as a function of polarization angle and frequency. No background correction was performed and was recorded at increments of $\Delta \lambda = 0.15$ nm.

Figure 5.9. Output mapping of DAST DFG signal from 12 THz to 27 THz. (a) Modeled DFG power as a function of polarization angle and frequency. (b) Measured DFG signal as a function of polarization angle and frequency. No background correction was performed and was recorded at increments of $\Delta \lambda = 0.15$ nm.
The overall shape of the measured DFG signal follows the same waveform structure as that of the modeled parameters. However, slight variations in the structure occur due to the sampling that was performed when the indices were measured for the known crystal structure. The acquired indices and transmission of the DAST in the THz domain were first taken with a tunable system at a larger bandwidth, with a different recording method, and with a different filtering technique [68]. In addition the generated spectrum is influenced by the type of IR filter used. The transmission coefficients of the filter will dictate the strength of each frequency in the measured spectrum. Therefore minor differences in the generated THz spectrum and the modeled simulation were expected.

![DGF DAST Output Power Model](image1)

![Measured Signal DFG DAST](image2)

Figure 5.10. Output power from DAST DFG. (a) Modeled output power as a function of OPG power and frequency. (b) Measured output power as a function of OPG power and frequency with no background corrections made and the data was taken at increments of $\Delta \lambda = 0.15$ nm.

In the last evaluation of the generated THz signal, a power to signal strength trend comparison was performed with the combined narrowband OPG sources polarized at an angle of $\theta = 10^\circ$. The selected frequency range of the signal was generated from 3.5 THz to 12 THz. In this comparison the dual OPG stages were equally reduced in power. The modeled power and the measured power are given as a function of frequency and combined OPG power in Fig. 5.10.
The measured signal waveform structure closely follows the modeled power parameters. Minimal variations occur in the generated THz signal which are attributed to the germanium filters used and atmospheric absorption. When the power of the combined OPA fell below 0.3 W, the detector response used was negligible. The results for the polarization and power comparison indicate that the largest THz signal gain occurs around a polarization angle of 17°.

The THz results are very promising for NDE imaging applications. Therefore, a high power scan was performed by filtering with a single filter only in order to measure the power of the three largest peaks with a Gentec detector in place of the Pyro-electric detector.

![High Power DAST DFG Spectrum](image)

Figure 5.11. Transmission spectrum using a single IR filter placed after the DAST crystal without a background correction. The data was recorded in increments of $\Delta \lambda = 0.35 \text{ nm}$.

In this measurement, four scans were overlapped to reach the full range of the system. The polarization angle of the combined beams was set at an angle of $\theta = 17^\circ$. The
power of the individual OPG injection seeded stages were set to 0.4 W each. The measured spectrum as a function of frequency is shown in Fig. 5.11.

From the figure, it was noted that a power mismatch occurred. The expected features due to the water vapor absorption and the phase mismatching were present. However, due to the selected polarization angle and reduced filtering the higher frequencies were able to generate large signal gains when compared to the lower frequencies. The limitations can be attributed to the available filters for that specific range. The signal dips caused by phonon absorption could not be compensated for. The THz signal power was measured at three selected frequencies of 16.1 THz, 18.9 THz, and 26.5 THz. With the limitations noted, the system attained THz signal strengths at select frequencies exceeding 0.09 mW.

Figure 5.12. Imaged THz waves from a 3 meter path length from the DAST Crystal
Signal levels found at approximately 1 V provide a large dynamic range necessary for imaging applications. The THz beam was imaged at these three frequency locations. A BAE FIR room temperature camera was placed in the beam path 3 m from the DAST crystal. We were able to successfully image the THz waves at these three locations. The images of these frequencies are given in Fig. 5.12.

5.4 Conclusion

To create a coherent THz-wave source with continuous narrowband frequency tunability based on the DFG DAST scheme, dual injection seeded OPG sources pumped with a 1.064 μm wavelength was developed. The tunability of the two OPG stages was successfully achieved by injection seeding with two different MgO:PPLN crystals. When the tandem OPGs were combined as a source for the DFG method, a tunable narrowband THz source was demonstrated with DAST. We demonstrated a wide tunability with the injection seeded dual OPG stages and a high spectral resolution of 3.1 GHz for the generated THz spectrum between 1.5 THz and 27 THz. We measured the power and signal strength as a function of DFG frequency and as a function of combined OPG polarization angle. We demonstrated the dependence of the output THz wave power at a given frequency to the linear polarization angle of the combined mixing beams in the DAST. These measurements validate this method as suitable for many spectroscopic applications due to the resolution of the system and tunable power that can be produced. The simplicity and size of the THz source is an essential requirement for the development of future applications. When compared with available sources, the aforementioned nonlinear system presents distinct advantages in linewidth resolution, broad tunability, and ease of use. In
addition, we have demonstrated that such a source is powerful enough for NDE imaging applications.
CHAPTER 6
THZ NONDESTRUCTIVE EVALUATION OF CERAMIC MATRIX COMPOSITES

6.1 Introduction

NDE techniques have been used to characterize defects such as voids, cracks, etc. in CMC materials due to thermal breakdown. THz radiation has been used as a novel detection tool for imaging these defects [70]. The ceramic materials are a subset of composites. Ceramic matrix composites have found a wide range of applications including jet engine components, structural components, and thermal protection systems (TPS). They have become an ideal choice for aircraft due to their high strength and thermal resistance. Silicon carbide (SiC) is a ceramic material which is strong, light weight, and is found in a whole host of applications that require reinforcement. The THz spectrum can be used to measure the photo-elastic effect in these materials when they are stressed. Applications have been developed to characterize the defects in SiC wafers in the far infrared (FIR) [71]. However, most techniques are not able to be fielded. Thus the developed DFG THz source was evaluated as a possible candidate for the detection of CMC defects in SiNC.

The developed THz system is intended to perform two tasks for the evaluation as a possible candidate for the detection of CMC material fatigue. The tasks encompass single point reflection spectroscopy and THz imaging. The added benefit is that the system does
not possess inherent limitations that would prevent the system from being configured into a compact fiber package.

6.2 THz CMC Study

A series of SiNC ceramic matrix composite samples were made to evaluate the source as a feasible detection tool. Three CMC samples of the same material were each individually tempered to a specific value of $1200^\circ C$ at a different temporal value. These materials were heated to these values to simulate aerodynamic heating. Distinct chemical changes were reported in these samples as a result. Due to the initial porosity of such composites results in the chemical degradation as a function of temperature. New molecular chemicals are formed and released from the composite which results in a buildup on the surface of the sample. Thus a change in surface roughness is a direct result due to the oxidation of these samples. A change in surface roughness can be directly measured using white light interferometry and is given in Fig. 6.1.

![Figure 6.1. Measurement of surface roughness of different heat treated SiNC samples.](image-url)
The molecular film buildup is on the order of hundreds of microns with a variable roughness. Thus, the surface of these materials have a roughness on the same order as the generated THz wavelengths which result in rough surface scattering of the incident THz waves [72]. The average roughness of an untreated SiNC sample was measured to be at 1 μm, and after the heat treatment the average roughness was measured to be 9.1 μm.

For the material of interest, the reflected THz signal provides a spectroscopic signature of the damage similar to that of the absorption without the need of reflected phase. The SiNC CMC’s are comprised of polar dielectric materials made up of molecules with vibrational and rotational oscillations. The Gaussian oscillator model may be used to describe the resonance in the permittivity. To develop a general metric by which the samples could be compared, the reflection data was converted to permittivity by fitting a series of Gaussian oscillators to the data. The Gaussian oscillator describes the real ($\varepsilon_1$) and imaginary ($\varepsilon_2$) components of the complex dielectric function $\tilde{\varepsilon}(\kappa_i)=\varepsilon_1 + i\varepsilon_2$, where $\kappa_i$ is the wavenumber which is related to the wavelength and frequency of the wave. The imaginary component can be expressed as the oscillator (Eq. 6.1) and the real component can be expressed as the principal part of an integral from -$\infty$ to $\infty$ over $\kappa_i$ along the real axis (Eq. 6.2)

$$\varepsilon_2(\kappa_i) = A e^{-\frac{(\kappa_i - \kappa_0)^2}{2\Delta \kappa^2}},$$  \hspace{1cm} (6.1) \\
$$\varepsilon_1(\kappa_i) = \frac{2}{\pi} \text{P} \int_{\kappa_i - \kappa_0}^{\infty} \frac{\kappa_i \varepsilon_2(\kappa_i)}{\kappa_i^2 - \kappa_0^2} d\kappa_i.$$  \hspace{1cm} (6.2) \\
The oscillator’s parameters are selected as optimization variables of the measured reflection spectrum and a fit is performed. Due to the multitude of peaks in the reflection spectrum, several oscillators are needed to perform an accurate reflection fit. The
reflectivity is a function of permittivity and the overall summation of the oscillators. Thus, the reflection fit is

\[
R = \frac{(\sqrt{\varepsilon} - 1)^2}{(\sqrt{\varepsilon} + 1)^2}.
\]

(6.3)

Many CMC related compounds have a strong dispersion relative to absorption [73]. For CMC materials a negative derivative of the reflection coefficients reveal spectral peaks at frequencies near the extinction coefficient. Since the reflectivity from the surface of such materials is simply the square of the magnitude of the reflection coefficient, it is possible to qualitatively compare the reflectivity of each sample absorption feature.

6.3 CMC Reflection Measurements

6.3.1 Experimental Setup

Each sample was individually evaluated by placing them in the path of the collimated generated THz beam. The THz beam was filtered using a single IR filter placed after the DAST. The CMC sample under assessment was rotated to an angle of 45 degrees with respect to the normal incidence of the beam. The reflectance spectrum from each sample was recorded by two different methods. The reflection spectrum was recorded by placing a pyro-electric detector 2 cm away from the rotated sample. The detector was synchronized with a lock-in amplifier and a 7 Hz chopper was placed after the parallel grated PPLN OPA. The second method consisted of imaging the samples at a specific frequency. The selected frequencies were 14.7 THz and 19.1 THz. The samples were imaged by placing a BAE FIR camera 20 cm from the rotated sample. The camera frame rate was 35 frames per second. The schematic for the pyro-electric detector and camera configuration is given in Fig. 6.2.
The CMC SiNC Hi-Nicalon was selected as the evaluation material. When tempered distinct chemical changes occur in SiC fibers and coatings. Thus, the reflection and absorption features of the SiNC material are known to exhibit spectral changes when tempered [74]. For the composite, chemical frequency peaks in the FIR spectrum associated with SiO$_2$ and B$_2$O$_3$ shift in the range between 5 THz to 20 THz. Thus, three samples of SiNC Hi-Nicalon were annealed at high temperature three different times. One sample was left untreated, another was heated to 1200$^\circ$C for 10 hours, and the last was heated to 1200$^\circ$C for 100 hours.

Each sample was constructed with a length of 15.5 cm, a width of 1.3 cm, and a nominal thickness of 0.2 cm. The term nominal thickness was used to indicate that the exact surface thickness of a localized area varied due to the weave pattern. The CMC samples were constructed to be dimensionally identical; however, the initial porosity of the samples differed.

![Figure 6.2.](image)

Figure 6.2. The configuration depicts the method in which the reflection spectrum was recorded via single point detection or imaging with a camera.

### 6.3.2 THz Reflection Measurements

The configuration for the single point detector was used to perform a scan of the samples. Multiple scans were performed on each sample and were averaged together for
each individual sample. The reflection from the three samples were measured in a range between 0.1 THz and 28 THz and is given in Fig. 6.3.

![THz measurement of SINC-Hi-Nicalon at different treatments](image)

Figure 6.3. THz reflection measurement of six scans for the three SINC heat-treated samples with the developed source. No background correction was made to the measured samples. The gaps in the spectrum are due to the phase miss matching characteristic inherent to DAST.

The reflectance bands affiliated with the formation of B$_2$O$_3$ and SiO$_2$ were apparent in all heat treated (oxidized) samples in the figure. However, we are only able to analyze the strength of the signal peaks where phase matching in the DAST is optimal. The measured reflectance data was in agreement with the known absorption response of the tempered samples. At the frequency of 14.7 THz the sample with the longest heat treatment of 100 hours provided the strongest reflectance signal followed by the sample heat treated at 10 hours and then the untreated sample. The reflectance band located approximately at 14.7 THz depict the vibrational modes of SiO$_2$ [75]. Another vibrational mode occurred at
approximately 19.1 THz but is masked by the SiC vibrational mode [76]. In addition, the reflectance band B$_2$O$_3$ resulted in a contribution to the SiO$_2$ band at 14.7 THz [77].

From the data we were able to determine which samples had undergone chemical degradation. In order to verify that the reflection spectrum from each sample did not coincide at one standard deviation. A difference in error was estimated for the three spectrums and is given in Fig. 6.4. The location was selected because it depicts where the reflected signal from the three samples were most closely located to one and other. From Fig. 6.4 we note that the sample signal measured for each was not divergent between scans. This signifies that the resolution of the system would permit the estimation of the permittivity for each sample.

![Figure 6.4](image)

Figure 6.4. Averaged reflection data for 6 scans at different locations on each SINC sample.

The permittivity of each absorption feature in the bandwidth of each sample can be estimated using the Gaussian oscillator model parameters. These parameters are the offset values of each oscillator and their complex poles, which is used to describe features in the absorption spectrum. A sample of the oscillator fit used is given in Fig. 6.5.
From the fitted data we can then extract the necessary information about the permittivity in which we can estimate the real and imaginary components. A permittivity comparison can be made between the untreated and treated samples. Thus, the comparison can aid in the overall analysis of these materials and validate the source as an NDE tool. The estimated permittivity for the three samples is given in Fig. 6.6.

Figure 6.5. Gaussian oscillator fit of a CMC THz sample scan after a thermal treatment of 10 hours.

Figure 6.6. Permittivity extraction from reflection data for different treated samples.
6.3.3 THz Sample Imaging

The THz reflection spectrum was imaged using a BAE camera with the aforementioned method. Imaging was performed without the use of a beam chopper or lock-in amplifier. Images were taken of each sample and no background corrections were made. Two frequencies at 14.7 THz and 19.1 THz was selected for imaging due to the dramatic difference in the reflected signal strengths of our source. The images of the three heat treated samples are given in Fig. 6.7 and Fig. 6.8. In the figure, the imaged samples are labeled and are marked with a rectangular grid. In addition, the region of interest (ROI) is circled. The ROI is the location where the incident THz beam is focused onto the different samples. The order of the imaged signal strength of the three samples corresponded to the data collected using the chopper pyro-electric detection method. Thus, the SiNC treated at 100 hours depicts the strongest reflected signal followed by the 10 hours and then the untreated sample.

![Image of THz Sample Imaging](image)

Figure 6.7. Imaged reflection of the THz beam off the three samples at a frequency of 14.7 THz. The labeled region of interest (ROI) marks where the THz beam is incident on the samples. The images were taken at a standoff distance of 20 cm from the three samples.
Figure 6.8. Imaged reflection of the THz beam off the three samples at a frequency of 19.1 THz. The labeled region of interest (ROI) marks where the THz beam is incident on the samples. The images were taken at a standoff distance of 20 cm from the three samples.

6.4 Conclusion

The results presented in this chapter validate the claim that the developed THz system is a viable candidate as an NDE inspection tool. Due to the system’s narrow bandwidth and broad frequency tunability, we were able to differentiate chemical degradation as a result of thermal loading between three SINC different CMC samples. We were able to determine the difference from two different detection methods. Initially, the thermally treated samples were identified by point detection and the permittivity was calculated. A clear difference in the permittivity was found. In the second method, we were able to determine the chemically degraded samples by simple THz wave illumination. In which the spectral response from the samples were imaged and correlated with the point detection data.
CHAPTER 7
PHOTORESPONSE OF THIN FILM HIGH TEMPERATURE SUPERCONDUCTORS

7.1 Introduction

There are two types of transient photo-response mechanisms due to the interaction of a high temperature superconductor and an ultrafast laser pulse. These interactions are normally regarded as the bolometric and the non-equilibrium transient response. The ultrafast interaction refers to the voltage transient behavior that is a direct result of the Cooper pair breaking process. Once these pairs are broken the electrons pair and relax into their equilibrium state. During relaxation the pairs are capable of emitting microwave and THz frequencies. The microwave and terahertz regimes are some of the most technically transformative regions of the electromagnetic spectrum. One of the most interesting fields of study for this region is the conversion from the optical spectrum into the microwave and terahertz region. In this process where two different engineering communities are united one can expect new and exciting technologies to be developed in order to bridge the gap.

The discovery of high temperature superconductors (HTS) has been a point of interest in the optics community because of their potential as a means to detect and generate THz wave radiation. The new concept under investigation is the use of the HTS material as a microwave generator. The material discussed in this experiment utilizes the only
known HTS material that can be fabricated into a thin film with complex geometries. In this chapter, the physics of the mechanism in which a photo-excited superconductor radiates is described.

7.2 Theoretical Background

The Cooper pairs in a superconductor are broken with ultrafast pulsed laser illumination [78]. Various non-equilibrium phenomena have been observed from laser irradiation. As a consequence of photon and Cooper pair interaction a multitude of quasi-particles are emitted. These quasi-particles are excited electrons that cascade through the material as they relax back toward equilibrium. A voltage transient is a direct result from the propagation of the particles in the material. In this process only the paired electrons are affected by the photon interaction. The propagating phonons initially remain in their equilibrium state, which is in equilibrium with the sapphire substrate [78]. Therefore, the photon thermal interaction in the material can be used to control the resistance of the material. Thus, the radiation from a current transient emitted by a low frequency circuit can be amplified by fabricating its structure out of HTS and illuminated with an ultrafast laser. Thus, two components need to be taken into account in order to model the material as a microwave radiator.

7.2.1 YBCO Optical Response

The theoretical study of transient non-equilibrium phenomena in superconductors involve the understanding of a probable four component system. The system consists of understanding the interaction between quasi-particles, Cooper pairs, and phonons. The process of interaction of these particles can be evaluated with their associated time constants as they relax towards equilibrium after the injection of photons. Relaxation of
the HTS involves several important time constants: (I) inelastic scattering of quasi-particles by electron-electron pairs \( \tau_{e-e} \) with electron-phonon interactions \( \tau_{e-ph} \); (II) quasi-particle recombination rate into Cooper pairs \( \tau_{ph-e} \); (III) quasi-particle generation by phonon pair breaking \( \tau_{ph-e} \); (IV) phonon decay by diffusion into the environment \( \tau_{\text{diff}} \), and phonon diffusion time across a thermal boundary \( \tau_{\gamma} \), and the phonon diffusion time into the sapphire \( \tau_{\text{sap}} \).

The scattering process for the electron-electron interaction results in an avalanche process in the material where quasi-particles are emitted in the material and cascade through the material which results in the multiplication of non-equilibrium particles. For YBCO, the time constant equation was estimated to be [79]

\[
\tau_{e-e} = (2 \times 10^{-17}) E^2 \text{ (eV)} \text{ (sec)}. \quad (7.1)
\]

In equation 7.1 \( E \) is the energy of an electron in this state above the Fermi energy. Thus, for an electron with a low energy of \( E = 1.0 \text{ eV} \) the corresponding time constant is \( \tau_{e-e} = 0.05 \text{ fs} \).

The electron phonon interaction time constant \( \tau_{e-ph} \) describes the time in which the electron transfers energy to the phonons propagating in the medium. For YBCO, the time constant equation as a function of frequency is [80].

\[
\tau_{e-ph} = \frac{3.3 \times 10^{-19}}{\left(\frac{h \omega}{2 \pi}\right)^3 \text{ (eV)}} \text{ (sec)} . \quad (7.2)
\]

In the equation above the phonon energy is represented by \( \frac{h \omega}{2 \pi} \). Equation 7.2 can be used to represent the electron-phonon and phonon electron interaction relaxation times. However, the two are linked to the heat capacities of the electrons and phonons.
\[
\frac{C_e}{\tau_{e-ph}} = \frac{C_{ph}}{\tau_{ph-e}}.
\]  
(7.3)

Thus, for phonons in the low energy regime of the superconductor with an energy of 2 meV [81], the transition time is approximately \( \tau_{e-ph} = 40 \text{ ps} \).

The phonon diffusion process within the HTS YBCO thin film is characterized by \( \tau_{\text{diff}} \). The equation for the diffusion process is estimated in Eq. 7.4 [80].

\[
\tau_{\text{diff}} = \frac{d^2C}{\kappa}.
\]  
(7.4)

In Eq. 7.4 \( d \) is the diffusion distance that is the thickness of the superconductor film, \( C \) is the specific heat capacity of the film, and \( \kappa \) is the thermal conductivity of YBCO. For HTS YBCO at a temperature of 80 K has a thermal conductivity of \( \kappa = 0.026 \text{ W/cm}, \) a specific heat capacity of \( C = 1 \text{ J/cm}^3 \), and a diffusion length of 100 nm. Thus the transition time for a phonon to diffuse along the C axis can be estimated to be \( \tau_{\text{diff}} = 3.4 \text{ ns} \).

The phonon diffusion process continues until the penetration depth reaches the substrate region. Once it reaches this area the phonon experiences a thermal boundary resistance due to the new thermal characteristic properties of the sapphire substrate. The time constant in which the phonons need to cross the boundary can be calculated from Eq. 7.5 [82].

\[
\tau_\gamma = CR_{bd}d. 
\]  
(7.5)

In Eq. 7.5 \( R_{bd} \) is the thermal boundary resistance between the YBCO thin film and the sapphire substrate. For our YBCO sample the specific heat capacity was \( C = 1 \text{ J/cm}^3 \), \( R_{bd} = 1 \times 10^{-3} \text{ Kcm}^2/\text{W,} \) and at a thickness of \( d = 300 \text{ nm} \). Thus the phonon diffusion time across a thermal boundary can be estimated to be \( \tau_\gamma = 3 \text{ ns} \).
The final diffusion process that needs to be considered is the time necessary for the phonon to diffuse through the sapphire substrate.

$$\tau_{\text{sap}} = \frac{d_{\text{sap}}^2 C_{\text{sap}}}{\kappa_{\text{sap}}}.$$  \hspace{1cm} (7.6)

In Eq. 7.6 $C_{\text{sap}}$ is the heat capacity, $\kappa_{\text{sap}}$ is the thermal conductivity, and $d_{\text{sap}}$ is the thickness of the sapphire substrate. For sapphire $C_{\text{sap}} = 1.66 \text{ Jcm}^3/\text{K}$, $\kappa_{\text{sap}} = 0.35 \text{ W/ (cmK)}$, and $d_{\text{sap}} = 0.5 \text{ mm}$ [83]. Thus, the time for thermal conduction through the sapphire substrate is approximately $\tau_{\text{sap}} = 12 \text{ ms}$. The YBCO material and its associated time constants are of interest in the photo-response process thus the phonon diffusion time can be neglected in the substrate. When considering the photo-response process, we can neglect the substrate diffusion, however in the relaxation and recombination process substrate diffusion needs to be considered because the substrate acts as a reservoir in which the phonon energy can escape.

The process that governs the optical response of HTS YBCO is derived from the aforementioned time constants. Initially the Cooper pairs are broken into single electrons (quasi-particles) by the incident photons. For an incident wavelength of 808 nm the energy of a single photon can be estimated to be $E = 1.53 \text{ eV}$. Each photon will break a single Cooper pair and create two unbounded electrons (quasi-particles) each of energy $2\Delta$ and $E-2\Delta$, where $2\Delta$ is the superconducting gap energy. The Cooper pair binding energy between the two electrons is 30 meV [82]. Once the Cooper pairs are broken due to a photon, the quasi-particles will relax to the normal state. The normal electrons will cascade through the material to lower energy, break other Cooper pairs, and cause an avalanche process. In general a laser pulse will induce this process. The interaction time is much less
than a femtosecond, thus a laser pulse on the order of 120 fs will trigger the interaction. The interaction will continue as long as the incident laser pulse is present within the recombination time of the Cooper pairs in the superconductor. The nature in which the electrons undergo relaxation and recombine into Cooper pairs is energy dependent. For high energy electrons the process of relaxation at an energy of 1 eV is $\tau_{e-e} = 0.05$ fs. The electron-electron relaxation is a fast process when compared to the electron-phonon process at a time of $\tau_{e-ph} = \tau_{ph-e} = 40$ ps. Thus, quasi-particles with high energy electrons will relax through an interaction with other Cooper pairs.

The interaction time between electron-electron quasi-particle generation and recombination will decrease as the electron energies deplete. Thus, once a threshold is reached relaxation is transferred to the phonon-electron process at approximately $\tau_{e-ph} = 33$ ps. The phonon process in which the energy is transferred between the electron and phonon respectively dominates the quasi-particle reaction. The three phonon processes dominate the interaction which include the breaking of Cooper pairs by phonons, the recombination of Cooper pairs by releasing a phonon, and phonons propagating into the substrate.

Once the pulse has passed, the electron and phonon energy will reach equilibrium at the critical temperature $T_c$. The cooling of the electrons and phonons occur over a nanosecond period, in which this time constant is determined from the time necessary for the high energy phonons to escape into the substrate and time the time necessary for the quasi-particle lifetime after a photo-transition to reach thermal equilibrium. Thus, total temporal dependence can be taken in quadrature

$$\tau = \sqrt{\tau_{e-e}^2 + \tau_{e-ph}^2 + \tau_{ph-e}^2 + \tau_{\text{diff}}^2 + \tau_{\gamma}^2 + \tau_{\text{sup}}^2}.$$  

(7.7)
In order to validate the prior process the interaction time should agree with the macroscopic quasi-particle lifetime constant for a material to reach thermal equilibrium developed in the Bardeen Cooper Schrieffer (BCS) theory. The quasi-particle lifetime recombination rate for a superconductor is \[ \tau_0 = \frac{\Delta_0}{k_B T_C} e^{\frac{\Delta_0}{k_B T}}. \] (7.8)

In Eq. 7.8, \( \tau_0 \) is a constant dependent on the material that reflects the strength of the electrons and phonons interacting due to a bolometric process. Transient photo-impedance measurements have measured the bolometric process in YBCO to be approximately \( \tau_0 = 10 \text{ ns} \) \[ 85 \], for YBCO the energy gap is inferred as \( \Delta_0 = 2.5k_B T_C \) \[ 86 \], and the critical temperature for thin film YBCO is \( T_C = 86.6 \text{ K} \) \[ 87 \]. To estimate the macroscopic quasi-particle lifetime we estimate the amount of heat delivered to the sample from photons in a temporal envelope of a 120 fs. The temperature increase \( T \) of the sample is due to the incident laser heating, which can be written as a function of the incident energy absorbed in a thin film \[ 80 \].

\[ T = \frac{F}{C \delta} \left( \frac{E_{\text{ABS}}}{E_0} \right). \] (7.9)

In Eq. 7.9 \( F \) is the incident laser fluence, \( \delta \) is the penetration depth of the light, and \( \frac{E_{\text{ABS}}}{E_0} \) is the fraction of the total energy absorbed into the thin film. The fraction of the incident energy absorbed can be written as a function of sample reflectivity \( R \), penetration depth \( \delta \), and sample thickness \( d \) as

\[ \frac{E_{\text{abs}}}{E_0} = (1-R)(1-e^{-\frac{d}{\delta}}). \] (7.10)
For YBCO the penetration depth at a wavelength of 808 nm, at an average power of 1 W, for a 1 mJ pulse is approximately $\delta = 1/\alpha = 100$ nm [88]. The reflectivity $R = 0.1$ [89], specific heat capacity was $C = 1$ J/cm$^3$, the sample thickness $d = 300$ nm, and the laser fluence $F = 5.09 \cdot 10^{-3}$ J/cm$^2$ (laser average power 1 W, pulse duration 120 fs, and 5 mm spot size of $1/e^2$). Thus, from the parameters given one can estimate that for 120 fs approximately a temperature increase is 4.35 K. The temperature increase can then be used in Eq. 7.8 to calculate the quasi-particle lifetime which results in a time constant on the order of half a nanosecond. Thus, the switching mechanism due to the photo-transient YBCO material is limited to nanosecond pulses.

The transient photo-response Cooper pair and quasi-particle density varies as a function of time. Initially a laser pulse much shorter than a nanosecond is incident onto superconducting YBCO at time $t_0$. The absorbed photons break multiple Cooper pairs and generate quasi-particles. At the time $t_1$ quasi-particles cascade through the material and further break Cooper pairs through the e-e process until the phonon energy becomes the primary transfer process at time $t_2$. The phonon process begins the process of recombination on the order of picoseconds. In this period the Cooper pair density begins a steady increase from the maximum number of quasi-particles $n_2$ until $t_3$ is reached. At $t_3$ the phonons and electrons reach equilibrium in which the energy in the phonons needs to escape in the form of heat diffusion which is on the order of nanoseconds. At the time $t_4$ the maximum density of Cooper pairs has been reached and all excess energy of the system has diffused into the substrate and the YBCO is again a perfect superconductor.
7.3 Kinetic Inductance Model

The kinetic inductance is an expression that is related to the inertial mass of the mobile current charge carriers in an electric field and is an equivalent to a series inductance in a circuit. The kinetic inductance is a physical expression that is contained in the total energy of a closed system that contains a multitude of particles of mass \( m \), number density \( n \), carrying an electric current \( I \). The total energy of the system can be written as \([90]\).

\[
E = \int_{\text{all space}} \frac{1}{2} \mu H^2 \, d\tau + \int_{\text{conductor}} \frac{1}{2} n m v^2 \, d\tau .
\]  

(7.11)

In Eq. 7.11 \( \mu \) is the permeability, \( H \) is the magnetic field, and \( v \) is the particle velocity. In the model we assume that the conductor is homogenous and has a uniform cross section. The parameters of \( m \), \( n \), \( e \), and \( \mu \) are assumed independent of \( H \) and \( I \). With the assumptions included we can re-write the above Eq. 7.11 as

\[
E = \frac{1}{2} L_m I^2 + \frac{1}{2} \left[ \left( \frac{m}{2 n e^2} \right) \left( \frac{1}{\sigma} \right) \right] I^2 .
\]  

(7.12)

In Eq. 7.12 \( l \) is the length, \( \sigma \) is the cross-sectional area of the material, the current \( I = n e v \sigma \), and \( L_m \) is the magnetic inductance. The magnetic inductance is not effected by the presence of incident photons on the material and is only affected by the material geometry. Thus, the kinetic inductance and the resistance of the material is the only parameters under evaluation in the photo-response process. The kinetic inductance is \([90]\)

\[
L_k = \frac{m}{2 n e^2} \frac{1}{\sigma} .
\]  

(7.13)

In addition the resistance of a wire conductor is given by

\[
R = \left[ \left( \frac{m}{2 n e^2} \right) \left( \frac{1}{\sigma} \right) \right] \frac{1}{\tau} .
\]  

(7.14)
In Eq. 7.14, $\tau$ is the electron collision time. The kinetic inductive reactance is written as a function of frequency $\omega$

$$\omega L_k = \left(\frac{m}{2ne^2}\right)(\frac{1}{\sigma})\omega. \quad (7.15)$$

By comparing Eq. 7.14 and 7.15 we note that for a superconductor $\tau \to \infty$ and the resistance is $R=0$. This means at any frequency the reactance $\omega L_k$ will dominate the resistance at any frequency. The kinetic reactance is the key component in the initial fast response of the superconductor thin film. Thus, the signal response is on the order of a few picoseconds which is known. However, by increasing the electron collision time and making $\omega < \frac{1}{\tau}$ we can generate signals into the GHz spectrum. Thus, by changing the geometry of the thin film the resistance becomes the dominant feature. Thus, in the initial onset of incident photon the kinetic inductance is dominant, but when the material is patterned in a specific manner the resistance becomes dominant.

The kinetic inductance for a superconducting thin film strip can be written in terms of length $l$, width $w$, thickness $d$, effective mass $m$, Cooper pair density $n_s$, total density of carriers $n_0$, quasi-particle density $n_{qp}$, temperature dependent London penetration depth, and the plasma frequency of YBCO $\omega_p$ [89],

$$L_k = \frac{m\ell}{2ne^2wd} = \mu_0\lambda_L^2 \frac{1}{wd} = \frac{\ell}{2\varepsilon_0\omega_p^2} \left[\frac{n_0}{n_0 - n_{qp}}\right] \left[\frac{1}{wd}\right]. \quad (7.16)$$

In Eq. 7.16 the Cooper pair density is related to the quasi-particle density by $n_s = n_0 - n_{qp}$.

When a laser pulse is incident on a thin film superconductor a decrease in the Cooper pair density occurs in response, incidentally the kinetic inductance changes due to the decrease in the number of super carriers due to the change in current density (reactance) as seen in Fig. 7.1.
The current density is given as a function of Cooper pair density $n_s$, the charge they are carrying $e$, and the velocity of the Cooper pairs $v_{sc}$.

$$J_s = 2n_s e v_s.$$  \hfill (7.17)

When a current bias is applied to the YBCO thin film strip, the velocity of the Cooper pairs will have to increase to maintain a constant current density due to the decrease in the super carriers in the location of the laser pulse illumination. The acceleration of the remaining carriers will cause a momentary voltage across the strip. The effect can be viewed by time differentiating the current density

$$\frac{\partial J_s}{\partial t} = 2n_s e \frac{\partial v_s}{\partial t} + 2e v_s \frac{\partial n_s}{\partial t}.$$  \hfill (7.18)

If we treat the system with a constant current bias of $I$, the equation becomes

---

Figure 7.1. An ultrafast laser pulse is incident on YBCO thin film which results in a decrease in Cooper pairs.
\[ 0 = \frac{2n_x e^2}{m} E + J_s \frac{2}{n_x} \frac{\partial n_x}{\partial t}. \] (7.19)

In Eq. 7.19 E is the electric field generated across the thin film strip due to the acceleration of the carriers. The voltage across the strip can be estimated by setting \( E = \frac{V}{l} \) and \( J_s = \frac{1}{wd} \) in Eq. 7.19

\[ V = -I \frac{m \ell}{2e^2 wd} \frac{1}{n_x^2} \frac{\partial n_x}{\partial t} = I \frac{d}{dt} \left[ -\frac{m \ell}{2n_x e^2 wd} \right] = I \frac{d}{dt} L_k. \] (7.20)

The optical response \( V \), is generated due to the change in the kinetic inductance. Thus under a current bias \( I \) the optical response is found by substituting Eq. 7.16 into Eq. 7.20

\[ V = I \frac{1}{2\epsilon_0 \alpha_p^2} \frac{\ell}{wd} \frac{n_0}{(n_0 - n_{qp})^2} \frac{dn_{qp}}{dt}. \] (7.21)

In Eq. 7.21 the change in the optical response is due to the change in the kinetic inductance. The optical response changes as a function of current \( I \) and the rate at which Cooper pairs are either broken or recombined. In the prior description the process of pair breaking and recombination was described and the over optical response will follow that pattern. The optical response varies as the derivative of the quasi-particle density, therefore the response will have a bi-stable signal response.

In order to predict the optical response, the quasi-particle generation rate needs to be evaluated. In order to find a solution we can use the Ruthwarf and Taylor (R-T) rate equations [91].

\[ \frac{dn_{qp}(t)}{dt} = q_{qp}(t) + 2\beta n_{so} - Rn_{qp}^2, \] (7.22)

\[ \frac{dn_{so}(t)}{dt} = p(t) + \frac{Rn_{qp}^2}{2} - \frac{Rn_{so}^2}{2} - \frac{(n_{so} - n_{soT})}{\tau_y}. \] (7.23)
The R-T rate equation describe superconductor system that are temporarily not in equilibrium. For our situation, we are generating a number of quasi-particles due to a photo-transition. In Eq. 7.22-23 the external quasi-particle generation rate \( q_{qp}(t) \) from an optical pulse is given as the number of particles per unit volume, the phonon generation rate \( p(t) \) from the external excitation of an optical pulse is given as the number of phonons per unit volume, \( n_{qp} \) is the quasi-particle density, \( n_{\omega} \) is the density of phonons generated due to the active quasi-particles, and \( n_{\omega} \) is the density of phonons is found by solving Eq. 7.23 for the a steady state condition (i.e. no photon injection and phonon generation term \( p(t)=0) \)

\[
n_{\omega T} = \frac{\tau_b}{\tau_r} n_{qp T}, \tag{7.24}
\]

The probability of breaking Cooper pairs by a phonon absorption is given by \( \beta \), in which \( \beta = 1/\tau_b \) which is the reciprocal of the Cooper pair breaking time and is on the order of a picosecond [92]. The recombination rate of the constituent quasi-particles into Cooper pairs is related to the recombination lifetime \( \tau_r \) and the quasi-particle equilibrium density \( n_{qp T} \) given by Eq. 5.25 [92, 93].

\[
n_{\omega T} = n_0 \left( \frac{T}{T_c} \right)^2. \tag{7.25}
\]

Thus, the recombination rate is written as the reciprocal of the recombination lifetime and the quasi-particle equilibrium density

\[
R = \frac{1}{\tau_r n_{qp T}}. \tag{7.26}
\]

In order to numerically solve the R-T differential equation, the number of quasi-particles generated due to an optical pulse has to be evaluated. The intensity and fluence of
Laser radiation are two very important characteristics when dealing with laser-material interactions. For the present work, intensity is characterized first. The intensity of an ideal Gaussian laser pulse at a time \( t \) and at a distance from the beam axis \( r \) is

\[
I(r, t) = I_0 e^{-2\frac{r^2}{w^2}} e^{-4\ln2 \frac{t^2}{\tau^2}}.
\]  

(7.27)

In Eq. 7.27 \( I_0 \) is the pulse peak intensity, \( \tau \) is the temporal full width half max, and \( w \) is the beam waist (the beam waist is where the intensity decreases to \( 1/e^2 \) with respect to the max value). The beam waist is

\[
w_1 = \frac{\sqrt{2}}{2 \ln 2}.
\]  

(7.28)

The total energy can then written as

\[
E = \int I(r, \theta, t)dA dt = \int_0^\infty \int_0^{2\pi} dr d\theta \int_{-\infty}^{\infty} e^{-4\ln2 \frac{t^2}{\tau^2}} dt.
\]  

(7.29)

Each individual integral can be evaluated as the respective integrals given as

\[
\int_0^\infty re^{-ax^2} dr = -\frac{1}{2a} e^{-ax^2} |_0^\infty = \frac{1}{2a},
\]  

(7.30)

\[
\int_{-\infty}^{\infty} e^{-ax^2} dx = \sqrt{\frac{\pi}{a}}.
\]  

(7.31)

The energy delivered from a single Gaussian pulse can then be estimated as

\[
E = I_0 \frac{w^2}{4} \frac{2\pi \tau}{\sqrt{4\ln 2}}.
\]  

(7.32)

When the repetition rate of the laser is \( f \) and the average power is \( P \), the energy in each pulse can be represented by

\[
E = \frac{P}{f}. 
\]  

(7.33)
Therefore we can determine $I_0$ by the substitution 7.33 into 7.32:

$$I_0 = \frac{4}{w^2 2\pi \tau} \sqrt{\frac{4 \ln 2}{\pi}} \left(\frac{P_{\text{ave}}}{f}\right). \quad (7.34)$$

Now that we have written the initial intensity as a function of power beam waist and temporal pulse, we want an intensity of each pulse as a function of time incident on the YBCO thin film. For the thin film the absorption coefficient $\alpha$, with a reflection coefficient $R$, a pulse width $\tau_0$, we can write the absorbed intensity as a function of time

$$I = I_0 (1 - R)(1 - e^{\alpha d})e^{-\frac{t-t_0}{\tau_0}}. \quad (7.33)$$

The energy from each photon in the pulse envelope at a wavelength $\lambda$ can be written as

$$E = \frac{1.24(\text{eV})(\mu\text{m})}{\lambda(\mu\text{m})}. \quad (7.34)$$

In order to calculate the density as a function of photon density per unit volume per unit time, the number density of photons has to be estimated for a single temporal pulse. For a laser at a wavelength of 808 nm would results in a photon energy of 1.53 eV or $2.45 \times 10^{-19} \text{J}$. The single photon energy can be used to estimate the number density $n_\gamma$ as a function intensity over a given thickness depth $d$:

$$n_\gamma = \frac{I}{d(1.53)} = \frac{I_0}{d(1.53)} (1 - R)(1 - e^{-\alpha d})e^{-\frac{t-t_0}{\tau_0}} = \frac{2}{w^2 \pi \tau} \sqrt{\frac{4 \ln 2}{\pi}} \frac{P_{\text{ave}}}{f} \frac{(1 - R)(1 - e^{-\alpha d})e^{-\frac{t-t_0}{\tau_0}}}{d(2.45 \times 10^{-19} \text{Joules})}. \quad (7.35)$$

From the number density of the generated photons a relationship can be made with the quasi-particle density. In this we can assume that the number of quasi-particles generated by a single photon interaction is given by the efficiency $\eta$. The quasi-particle density given as the number of quasi-particles per unit volume per unit time can be calculated as
\[ q_{qp} = \eta \left( \frac{2}{w^2 \pi \tau} \sqrt{\frac{4 \ln 2}{\pi}} P_{\text{ave}} \frac{(1-R)(1-e^{-\omega d})e^{-\frac{t}{\tau_0}}}{f d(2.45 \times 10^{-19} \text{ Joules})} \right). \]  

(7.36)

From the derived equations the proper substitutions can be made in order to calculate the quasi-particle response in order to calculate the optical response given in the following equations

\[ \frac{dn_{qp}}{dt} = \eta \left( \frac{2}{w^2 \pi \tau_0} \sqrt{\frac{4 \ln 2}{\pi}} P_{\text{ave}} \frac{(1-R)(1-e^{-\omega d})e^{-\frac{t-t_0}{\tau_0}}}{f d(2.45 \times 10^{-19} \text{ Joules})} \right) + \frac{2}{\tau_b} n_{oa} - \eta R n_{qp}^2, \]  

(7.37)

\[ \frac{dn_{oa}}{dt} = \frac{R n_{qp}^2}{2} - \frac{1}{\tau_b} n_{oa} - \frac{1}{\tau_f} (n_{oa} - n_{oaT}). \]  

(7.38)

The ordinary differential equation can be solved using the Runge-Kutta (R-K) numerical algorithm. In order to estimate the optical response Eq. 7.37 and its derivative was incorporated in Eq. 7.21.

Figure 7.2. The transient optical response of YBCO at superconducting temperatures due to different pulse durations.

The Cooper pair breaking time constant \( \tau_b \) was 1 ps, the recombination time was 0.5 ps, and the phonon escape time was 3 ns. The optical response model due to different incident
pulse durations of a 5 mm beam diameter (the beam waist is where the intensity decreases to \(1/e^2\)), average power of 1 W, on a 5 mm piece of YBCO is given in Fig. 7.2.

The calculated optical transients in Fig. 7.2 show a strong dependence on the width of the excitation pulse. In order to generate a large number of quasi-particles one only has to illuminate the HTS with a pulse width far less than the phonon escape time \(\tau_\gamma\). In general the generation rate determined by the pulse width has to be faster than the phonon escape time in which the energy can escape through other means than by breaking Cooper pairs. The effect can be inferred by Fig. 7.2 where when the pulse duration reaches a nanosecond the quasi-particle generation becomes negligible, thus the optical response becomes negligible. The presence of a fast peak followed by a negative dip determines the oscillatory kinetic inductive response due to Cooper pairs breaking and repairing. Therefore the effect can be used to modulate the resistance in a current carrying superconductor by means of laser beam parameters, geometry, and current to make an amplifying microwave antenna.

**7.4 Ultrafast photo-response for microwave generation**

The resistance of superconducting YBCO can be modulated using the ultrafast photo response phenomena. The resistance of a superconductor is dependent on the number of Cooper pairs present in the sample. The resistance is dependent on the number of quasi-particles combined to form Cooper pairs. Therefore, the ultrafast response of a femtosecond laser pulse incident on the HTS can generate an ultrafast resistance spike. For example a thin film YBCO ring known as a Corbino disc with an outer diameter of \(b=20\) mm, an inner diameter of \(a=15\) mm, a thickness \(d\), has a resistance that is dependent on the geometry of the ring and the resistivity of the material [79]. The resistivity \(\rho\) of the material
is dependent on the quasi-particle density $n_{qp}$, the free electron mass $m$, the electron scattering time $\tau$, and the electronic charge $e$:

$$\rho = \frac{m}{n_{qp} \tau e^2}.$$  \hspace{1cm} (7.39)

The overall resistance of the thin film ring can then be written as a function of the resistivity and is

$$R = \left(\frac{\rho}{2\pi d}\right) \ln\left(\frac{b}{a}\right).$$  \hspace{1cm} (7.40)

The quasi-particle density rate due to an ultrafast pulse is on the order of several picosecond depending on the temporal pulse width. Thus, the resistance is on the same temporal scale and is given in Fig. 7.3. The resistance change can be viewed as an ultrafast modulation/switch to current when passed in the thin film loop. In a general loop antenna an analog signal is conducted around the ring. If the same approach is taken and the YBCO thin film ring is treated as a loop antenna, the process can be viewed as a dc antenna signal because when compared to the emitted frequencies the switching mechanism time can be viewed as a delta function. In addition, the emitted frequencies can then be amplified on a scale proportional to the current on the ring.

![Figure 7.3. Resistance due to the change in quasi-particle number density. On the time scale of a ns the resistance is that of a delta function.](image)
The effect is proportional to the current because the excess current carrying electrons pair up in the same manner as the prior electrons in the system. Therefore, there is a larger number of Cooper pairs that can interact with the photons in the ultrafast pulse. The result is that the excess energy can be radiated away in the form of an ultra-wideband pulse.

The excess energy in a superconductive thin film loop is radiated in two ways after the interaction with an ultrafast laser pulse. One way that was mentioned before is that due to the reactance the charge carriers accelerate, in which an accelerating charge radiates. The primary method for evaluation is the radiation due to a current transient in a superconductor caused by a fast superconductive to normal transition in a loop antenna as seen in Fig. 7.4.

![Diagram](image_url)

Figure 7.4. An ultrafast laser pulse is incident on YBCO thin film ring which results in a decrease in current.

In order to assess the interaction, the system can be evaluated as a superconducting dc circuit with a fast transient. One method to begin the evaluation is to examine the ideal duel capacitor model developed by Boykin [94]. If one charged capacitor is attached to a
discharged capacitor by an ideal wire the energy before in the one capacitor is greater than
the energy after in both. The effect can’t be explained as Joule heating because the system
is in an ideal superconductive state. A conventional circuit possesses a small resistance,
however in the ideal superconductive state there is no resistance. Boykin’s explanation of
the paradigm contained radiation effects. To represent the emitted radiation a non-linear
resistor was used in an RL circuit to account for the missing energy. The power radiated
from the non-linear resistor can be modeled as magnetic dipole radiation from a circular
loop of radius a with a current I(t). The power can be calculated from the loop through the
current. Once the current is known, the E and H fields can be found, the radiated power
can be obtained from the pointing vector. Thus for the loop the radiated power through a
spherical shell centered a distance r away is given by

\[ P_{rad}(t) = \frac{\pi a^4}{6\varepsilon_0 c^3} [\tilde{I}(t - r/c)]^2 = \beta [\tilde{I}(t - r/c)]^2. \] (7.41)

The thin film YBCO loop can be modeled using the radiating equivalent circuit equation
(ECE) where the current in now modulated by the quasi-particle resistance change as a
function of laser pulse width. Thus, the current in the loop operates as a function of the
resistance change due to the picosecond response found in the RT model from an ultrafast
laser. The current is dependent on the integration of the RL circuit is [97]

\[ I(t) = I_0 e^{\int_{\tau_0}^{\tau} R(t') dt'}. \] (7.42)

The resistance in the power transition given in Eq. 7.22 becomes a function of
quasi-particle density due to laser pulse interaction.

\[ P_{rad}(t) = \beta I_0^2 \left( \frac{R^2(t)}{L} - \frac{\dot{R}(t)}{L} \right)^2 e^{-\int_{\tau_0}^{\tau} \frac{R(t') dt'}{L^2}}. \] (7.43)
In Eq. 7.43 the resistance can then be modeled as a resistor that is dependent on the quasi-particle generation and destruction.

### 7.4.1 RL Fourth Order Approximation

In Eq. 7.12, we stated that the kinetic inductance was the key component in photo-response of the superconductor. However, by forming the HTS into a ring the macroscopic inductance must be taken into account as well. In order to estimate this effect, an approximation for the current must be made to calculate the frequency response in Eq. 7.43. Essentially, the traditional RL circuit fails to take into account the total inductive energy of the system or the radiation component of the system. The transient response of the RL model will respond in a different manner than the normal exponential decay curve. It will respond in a similar manner to that of an RLC system where the circuit behaves like an overdamped and underdamped oscillator [97]. The reason for this is due to the change in resistance. The underdamped oscillations are due to the decrease in resistance of the quasi-particles recombination and the over damped oscillations are due to the increase in resistance due to the Cooper pair separation. We can model the system as an RL circuit and approximate the change in current using a Taylor expansion [97]. We can consider a thin film YBCO circular loop with an outer radius $a$. We can take a point $s$ to be on the surface of the ring. A circular superconducting thin film loop is illuminated by a laser and causes the system to reduce to its normal state. As a result, we can model this change as a voltage transient in the thin film ring over the circuit $A$. This radiating circuit equation is

\[ \oint_A E_t(s) ds = -\frac{\mu_0}{4\pi} \frac{\partial}{\partial t} \oint_A \frac{I(t - \frac{r}{c})}{r} d\vec{s} d\vec{s}'. \]  

(7.44)
The voltage transient of the circuit can be modeled as the sum of voltage drops in the circuit (lhs) and the rate of change of the magnetic flux in the ring (rhs). In Eq. 7.44, \( E_i \) is the electric field inside the conductor, and \( r \) is

\[
r = |\vec{s} - \vec{s}'|.
\]  

(7.45)

Now if we assume that a voltage transient exists due to the laser interaction Eq. 7.44 can be rewritten for the RL circuit as

\[
RI(t) - V(t) = -\frac{\mu_0}{4\pi} \frac{\partial}{\partial t} \oint_{A} \frac{I(t - \frac{r}{c})}{r} d\vec{s} d\vec{s}'.
\]  

(7.46)

Once the transition occurs, Eq. 7.46 will lose the voltage term and reduce to the normal RL circuit equation. The inductance of the circuit is considered to be the normal ring inductance with an outer diameter \( b \), inner diameter \( a \), and thickness \( d \):

\[
L = \frac{\mu_0}{4\pi} \oint_{A} \frac{d\vec{s} d\vec{s}'}{r} = \frac{\mu_0 d}{2\pi} \ln\left(\frac{b}{a}\right).
\]  

(7.47)

In Eq. 7.46 the right hand side of the equation can be expanded with a Taylor series into the fourth order [97]. The fourth order approximation of the current in a transient RL circuit is

\[
-\frac{\mu_0}{4\pi} \frac{\partial}{\partial t} \oint_{A} \frac{I(t - \frac{r}{c})}{r} d\vec{s} d\vec{s}' \approx L\dot{I}(t) - \alpha \ddot{I}(t) + \beta \dddot{I}.
\]  

(7.48)

In Eq. 7.48, \( \alpha \) and \( \beta \) are constants and defined by

\[
\alpha = -\frac{\mu_0}{8\pi c^2} \oint_{A} r d\vec{s} d\vec{s}' = \frac{2a^3 \mu_0}{3c^2},
\]

\[
\beta = -\frac{\mu_0}{24\pi c^3} \oint_{A} r^2 d\vec{s} d\vec{s}' = \frac{\pi a^4 \mu_0}{6c^3}.
\]  

(7.49)
Now when a transient voltage occurs due to the laser interaction we can rewrite Eq. 7.46 in terms of the fourth order approximation

$$\beta \ddot{I} - \alpha \dot{I}(t) + L \dot{I}(t) + R I(t) - V(t) = 0.$$  \hfill (7.50)

Now, we can treat the voltage change as a function of characteristic time of the photoresponse transient

$$\beta \ddot{I} - \alpha \dot{I}(t) + L \dot{I}(t) + R I(t) - V_0 e^{-t/\tau_c} = \beta \ddot{I} - \alpha \dot{I}(t) + L \dot{I}(t) + R I(t) - V_0 e^{-t/2a} = 0.$$  \hfill (7.51)

In the equation above the characteristic time of the transient current decay is on the order of the loop constant time of flight for the current. For the current we can numerically find the Eigen solutions in Eq. 7.51 for the fourth order homogeneous ODE in the form

$$\beta \lambda^4 - \alpha \lambda^3 + L \lambda + R = 0.$$  \hfill (7.52)

Three different currents can be found from the given Eigen solutions that pertain to the dampening and are given below. The three currents result in different radiation trends. As the resistance increases or decreases as a function of time the type of dampening will change.

$$I(t) = \begin{cases} Ac^{\lambda_1 t} + Be^{\lambda_2 t} + Ce^{-t/\tau} & \text{O.Damped} \quad \text{Re al } = \lambda_1, \lambda_2 \\ (A+Bt)e^{\lambda t} + Ce^{-t/\tau} & \text{C.Damped} \quad \lambda = \lambda_1 = \lambda_2 \\ e^{\delta}(A \cos(\omega t) + B \sin(\omega t)) + Ce^{-t/\tau} & \text{Underdamped} \quad \lambda_{1,2} = \delta \pm \imath \omega \end{cases}.$$  \hfill (7.53)

The radiated power frequency response of such a system is given in Fig. 7.5. In the power calculation we assumed that the characteristic time was on the order of the time of flight around the ring \(\tau = 1.3 \times 10^{-11}\) s. From the figure we note that if the interaction time is on the order of one period around the ring, GHz radiation can be generated.
Latypov et al. showed that for certain resistance levels, three types of dampening will occur [98]. The Eigen solutions of the current are strongly dependent on the resistance and dictate how the system can behave in a similar manner to an RLC circuit solution. Due to Cooper pair interaction we know that the resistance is changing on the order of the relaxation period. However, in order to generate microwave radiation the resistance change has to be on the order of the time need for the current to complete one period around the ring. In the next chapter we will discuss the measured microwave signal response from a number of YBCO HTS thin film rings, where the measured response displays similarities to both the kinetic inductance model and the RL model.
CHAPTER 8

HTS MICROWAVE GENERATION

8.1 Introduction

In this experiment, we study the microwave response of YBCO thin film rings due to an ultrafast laser interaction. We have developed an RF amplifier that is dependent on the amount of current present which operates as a DC antenna. In our approach a non-traditional laser-pulsed antenna is developed. In this study the dependence of the transient response of the YBCO was systematically evaluated based on current, magnetic field strength, average laser power, and laser pulse duration. In addition, it is shown that the microwave spectral waveform changes as a function of ring geometry.

8.2 Single Pulse Ring Resonator Experimental Setup

In the experiment, the interaction between a thin film ring at superconductive temperatures and an ultrafast laser pulse less than a picosecond was studied. The rings for the experiment consisted of YBaCuO thin film deposited by evaporation methods with 100-300nm layer thickness onto sapphire substrates. Different geometries of YBCO were fabricated as shown in Table 8.1.

The YBCO rings were slowly cooled and submerged into a Styrofoam container in a plastic Dewar filled with liquid nitrogen. Once in its superconducting state, a persistent current was induced in the material. In order to put a current on the rings, they were inductively charged using a 100 turn solenoid. The thin film had a critical temperature $T_c$
=88K and a transport current density $J_c=4 \text{ MA/cm}^2$. For some of the measurements the temperature and magnetic field strength were monitored using a cryogenic thermocouple and hall probe shown in Fig. 8.1. Experiments with an isolated YBCO ring were also performed. In this method we eliminated the nearby metal housing and wire. They were eliminated to reduce the possibility of extraneous signals due to electromagnetic coupling.

<table>
<thead>
<tr>
<th>Ring</th>
<th>Outer Diameter</th>
<th>Track Width</th>
<th>Thickness</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>20</td>
<td>5</td>
<td>300</td>
</tr>
<tr>
<td>2</td>
<td>20</td>
<td>1</td>
<td>300</td>
</tr>
<tr>
<td>3</td>
<td>10</td>
<td>1</td>
<td>300</td>
</tr>
<tr>
<td>4</td>
<td>10</td>
<td>5</td>
<td>300</td>
</tr>
<tr>
<td>5</td>
<td>5</td>
<td>1</td>
<td>300</td>
</tr>
<tr>
<td>6</td>
<td>20</td>
<td>10</td>
<td>200</td>
</tr>
<tr>
<td>7</td>
<td>20</td>
<td>5</td>
<td>200</td>
</tr>
<tr>
<td>8</td>
<td>20</td>
<td>1</td>
<td>200</td>
</tr>
</tbody>
</table>

Table 8.1. YBCO Ring Geometry.

When this type of experiment was done, the rings were energized to the critical current $I_c$ by means of a removable hand held solenoid. Current in the ring was calculated by measuring the magnetic field at the center of the ring. An added benefit was that these rings are the only type of antenna that can remain powered when electromagnetically isolated from its inductive solenoid.

With this method the rings could be completely isolated. This was an important point because secondary resonances were observed due to the close interaction with adjacent wires. To create an ultrafast superconducting to normal transition, a femtosecond
laser with a 1 mJ pulse energy was used. We measured the generated microwave radiation by pulsed RF S-parameter method.

Figure 8.1. YBCO ring atop a solenoid wrapped sapphire rod with attached hall probe and thermocouple inside a Styrofoam container placed inside a plastic Dewar.

We utilized a high bandwidth (65 GHz) digital sampling oscilloscope in combination with the antenna. The oscilloscope was synchronized and triggered by the lasers’ q-switch. The antenna were centered in the plane of the YBCO ring at the phase center of the radiator. They were placed at a distance of 170 cm from the ring. The setup is given in Fig. 8.2.

Figure 8.2. The single shot YBCO ring antenna experimental setup
We fabricated the HTS devices on a sapphire substrate which is transparent to our high energy femtosecond laser pulses (Ti:sapphire regenerative amplifier system). In addition the material was selected in order to preserve the bandwidth of the transient signal with minimal material interaction. The photo-response of the superconducting thin film is thought to break Cooper pairs resulting in microwave radiation. To measure the radiated power from 0.2-40 GHz two microwave antenna were utilized in which the spectral range of each horn was 1-18 GHz and 18-45 GHz.

8.3 Microwave Response of YBCO

In this section, the microwave response of the HTS YBCO thin film rings are studied systematically. Eight different geometries are used to study laser pulse duration, current dependence, magnetic field, and laser power. These permutation result in a varied frequency response dependent on the variable used.

8.3.1. Receiving Antenna

In order to estimate the real generated waveform and the transmitted spectrum from the ring, the gain and loss of the receiving antenna must be taken into account. In order to calculate the power at the radiator, the gain and loss of the receiving antenna is linearized. The linearized gain and loss for the 1-18 GHz is given in Fig. 8.3.

From the gain and loss of each antenna receiver, we are then able to calculate the power from each YBCO ring at the point of transmission. This can be accomplished with the use of the Friis equation

$$P_{nk} = \frac{G_{nk}G_{rk}LP_{nk} \lambda^2}{(4\pi R)^2}.$$  \hspace{1cm} (8.1)
In Eq. 8.1 we can treat the YBCO rings as a radiator that radiates in all space with the same intensity for a given frequency from a single point. Thus, each YBCO ring is treated as an isotropic radiator. In Eq. 8.1 $G_{tk}$ is isotropic=1, $G_{rk}$ is the gain of the receiving antenna, $L$ is the insertion loss, $P_{tk}$ is the power of the transmission, $P_{rk}$ is the power received, $R$ is the distance of the receiving antenna, and $\lambda$ is the wavelength of the wave. With the given parameters we can then calculate the power radiated from each ring given as

$$P_{tk} = \frac{P_{t} (4\pi R)^2}{G_{rk} L_{t} \lambda^2}.$$  

(8.2)

8.3.2 Microwave Response Waveform YBCO

Initially, an isolated YBCO ring with no induced current was illuminated and is given in Fig. 8.4. The ring for the measurement had a 20 mm outer diameter, a 5 mm track width, a 300 nm in thickness, and the measured temperature was at 78 K. The ring was illuminated with a 120 fs pulse, a pulse energy of 1 mJ, and a beam diameter of 5 mm (1/e...
intensity). Illumination was repeated several times and was averaged. From Fig. 8.4 we see that the measured microwave signal response did not vary over the course of the multiple laser illuminations. At a high average laser power of 1 W we note that from the calculated power, frequency content much higher than expected was encountered. The majority of the signal power was observed to reside in a range between 15-35 GHz at a maximum power of 1 mW. With such a short pulse time interaction with the material, the thermal equilibrium temperature of the quasi-particles and phonons do not have enough time to increase above the critical temperature range. For the thin film thickness 300 nm, which was much greater than the optical penetration depth, normal electrons were generated near the top of the material. The relaxation of the normal electrons when the bolometric process occurs is due to the heat diffusion in the material and the thermal boundary resistance. This type of relaxation is on the order of one ns if the material is brought above the critical temperature of the superconductor. The transition time for the measured response is much longer than the normal bolometric recovery process. Thus, we were able to generate microwave radiation beyond the bolometric limit for a superconductor held below its critical temperature [99].

An interesting effect occurred when a current of 1 A was induced on the same ring. The recorded power spectrum is given in Fig. 8.5. In the figure the recorded frequency spectrum shifted to a lower frequency regime. When a current was present an amplitude range 2-18 GHz became the dominant feature. The maximum transmitted power of the generated frequency was measured to be approximately 60 mW. When compared to the prior interaction, the original power level becomes indistinguishable from the elevated noise floor of the new signal.
Figure 8.4. Illuminated uncharged YBCO ring. (a.) Several measured signals of generated microwave radiation. (b.) Averaged received power spectrum.

Figure 8.5. Illumination of a charged YBCO ring. (a.) Generated microwave radiation of several signals averaged together. (b.) Averaged received power spectrum.
Figure 8.6. Different frequency bands generated simultaneously in a single transition.

The relevant content resides in the 2-18 GHz regime. Hence, the recorded signal has become a result of the modulated current induced in the superconductor similar to the relaxation of an RL circuit with an ultrafast switch. In addition, we note that significant power is generated in multiple microwave bands that range from R through the Ku band. The generated band structure is given in Fig. 8.6.

In the experiments, we observed that the shape of the waveforms for several microwave responses varied with laser power, current, pulse duration, and polarization angle. Moreover, the generated bandwidth exhibit different features for thin vs thick films and ring diameter. In the next sections we will show that the key to generating different frequency content in the transmitted power from the YBCO generator was strictly due to the geometry alone.
8.3.3 Laser Pulse Duration Study

For comparison, we measured the waveforms due to the interaction of different laser pulse durations with a single YBCO ring at a current of 5 A. Figure 8.7 depicts the microwave power response dependence on the duration of the optical pulse. The frequency content of the measured radiation is not dependent on the duration of the optical pulse, only the amplitude is dependent on the pulse duration. For this situation the optical beam diameter and power was held constant at 5mm (1/e intensity) and 1.02 W respectively.

In Fig. 8.8 we plot the maximum power of the signal as a function of pulse duration. As the duration of the pulse is increased to a nanosecond, the generated microwave power becomes unmeasurable. It is generally believed that the kinetic inductive response can only occur at temperatures far below the critical temperature of the super conductor [89]. We note that the response of the YBCO ring antenna agrees with the pulse duration limit of the developed kinetic inductance model given in Chapter 7. The oscillation in the bipolar...
feature in the measured response is similar in waveform to the kinetic inductance model, but has a frequency response similar to the RCE model.

8.3.4 Current Dependence of the RF Response

In this experiment, a single ring was selected, and all variables were held constant except for the current. The film thickness was 300nm, the ring diameter was 10 mm, and the track thickness was 1 mm. The measured substrate temperature was 79 K. The laser average power was measured to be 1.02 W. Magnetic field measurements of the ring were made as a function of position across the ring and as a function of current. In Fig. 8.9 (a), a current of 1.5 A was induced and the field was measured. The ring was then illuminated with an ultrafast pulse, and the field measurements were taken again. The magnetic field strength was depleted by the laser interaction and reduced down to the residual magnetic field level. We note that under the isolated ring experiment the entire current in the ring can be depleted by a single laser pulse. In Fig. 8.9 (b) we measured the maximum magnetic

Figure 8.8. RF power measured as a function of laser pulse duration.

124
field from the center of the ring as a function of current. The magnetic field measurements depict a situation where the thin film ring becomes saturated.

In the next phase of the experiment the ring was inductively charged a number of times at several different current values. After each charge the ring was transitioned with same laser parameters for each illumination.

![Graphs showing magnetic field measurements](image)

Figure 8.9 (a.) Measured magnetic field before and after laser illumination as a function of position over the ring. (b.) Radiated magnetic field as a function of current measured from the center of the ring.

The typical waveform response was similar for all of the rings. The response for the 10 mm outer diameter ring is given in Fig. 8.10. The frequency response between the different currents did not change. Therefore, the generated frequency spectrum did not depend on the amount of current present. However, the amplitude of the generated RF radiation did change as a function of current. The radiated peak power was tracked as a function of current and is given in Fig. 8.11.
Figure 8.10. Microwave response due to different induced currents. (a) Measured voltage signal due to different currents. (b) Frequency response due to currents.

The radiated power as a function of current closely follows the trend of the magnetic field as a function of current. At 4 A the radiated power begins to decline with the magnetic field due to the saturation of the material.

Figure 8.11. Measured peak power as a function of induced current in the ring.

The thin film ring radiation is closely correlated to the current which is modulated by the Cooper pair separation. The ring response is similar to a DC amplifier where an excess number electrons are paired to form Cooper pairs due to the induced current.
However, limitations exist that limit the amount of current that can be placed on the thin film before saturation is reached and damage occurs. The other option in which the amplitude can be adjusted is possibly through the laser power.

8.3.5 Power Dependence of the Measured Response

For comparison, experimentally obtained waveforms at different track widths for rings with an identical thickness of 200 nm were evaluated. The track width for the three was selected to be 10 mm, 5 mm, and 1 mm. We noted once again that the laser power only played a role in the amplitude of the response. The laser power was varied while the laser pulse duration was held at a constant for all three rings. The evolution of the amplitude in the frequency response is illustrated for the three rings in Fig. 8.12.

![Laser Power Study](image)

**Figure 8.12.** The measured peak power frequency response of three different rings as a function of average laser power.

The peak power amplitude of the rings with a track width of 5 mm and 10 mm initially decreases at a very slow rate at the lower power range of the applied laser. Once
the average laser power exceeded 0.7 W, the measured RF power begins to increase very rapidly.

The physical mechanism behind this observation is a result of the dynamical change in Cooper pair density. At a low power and larger track width compared to the laser spot size the number density of Cooper pairs are more broadly spaced in the material as they are conducted around the ring. Due to the diameter of the laser illumination, only a fraction of the Cooper pairs are affected because the interaction cross-section is small when compared to the full width of the thin film. We observed that the ring with the 1 mm track width had the opposite trend. Initially, at an average laser power below 0.1 W the RF generated power significantly reduced the penetration depth of the laser. However, once the average laser power exceeded 0.1 W the measured RF response grew faster than the wider rings. This was because the Cooper pairs were confined into a smaller interaction cross section where the maxim number of pairs could be broken by the laser pulse. During the laser power study a saturated curve did not occur in the RF response of the chosen rings. This indicates that a laser with a greater energy per pulse could be used in future experiments.

8.4 Conclusion

The results presented in this chapter show that HTS YBCO thin film rings are capable of generating broadband microwave radiation in a range that covered the R band through the Ku band. The frequency response of the system can only be change by the geometry of the rings. We demonstrated that the amplitude of the radiated power can by controlled through the average laser power, induced current, or pulse duration of the laser. The maximum radiate power was found to be on the order of hundreds of milli-watts. The
combination of broad bandwidth and high radiated power would make such a method a possible candidate for a microwave spectroscopy source in NDE.
CHAPTER 9
CONCLUSIONS

In this dissertation three different sources associated with three different spectrums were developed and tested. The tunable IR source proved to be narrow enough to perform chemical sensing. The developed narrowband THz system was used as a spectroscopic source and provided enough power to identify chemically degrade CMC samples. In the final system we were able to generate broad band microwave radiation with the transient HTS thin film YBCO ring method. With the developed method we were able to provide quantitative characterization of the interaction of YBCO with ultrafast illumination.

Chapter 1 provided an overview of the current sources under evaluation as possible candidates in the IR, THz, and microwave regimes. The chapter discussed the current needs for fielding new NDE technologies to examine structural properties of aerospace materials.

Chapter 2 was an overview of the nonlinear techniques associated with PPLN OPA and QPM. PPLN OPA served as the key method for narrowband tunability in both the IR source and the THz source. In the chapter I discussed several different features that were used in our experimental setup in Chapter 3.

Chapter 3 is where the developed IR source and measurements were discussed. In this chapter the relevant measurements were discussed that pertained to frequency tuning and the bandwidth of our source. With an off axis etalon, we were able to injection seed an OPA and generate signal wavelengths with 90 μJ of energy with a conversion efficiency
of 14% from the pump to the signal wavelength. We measured a small fraction of the CO$_2$ absorption spectrum with the source. The measured line width of the narrowband OPA laser was found to be less than 5.4 GHz with an effective tunable range of 287 GHz at a fixed temperature.

In Chapter 4 we discussed DFG THz generation using a DAST crystal. In addition we discussed the phase matching ability of DAST and the theoretical projection of power as a function of polarization angle. In this chapter the reader was introduced to techniques for evaluating the linear and nonlinear optical properties of crystals such as DAST.

In Chapter 5 THz wave radiation was generated by mixing two signal wavelengths from tandem seeded MgO:PPLN OPA stages in a DAST crystal. I demonstrated a wide tunability with the system across a range of 1.5 THz to 27 THz at a linewidth of 3.1 GHz. In addition the THz power from the system was large enough to perform imaging in the FIR. The THz source was then used to detect the chemical degradation in several CMC samples by spectroscopic reflection measurements and imaging which was given in Chapter 6. With the large dynamic range of our developed source we added to the field of narrowband THz generation. By successfully detecting chemical changes in materials we have validated it as a candidate for further development.

In order to increase the overall power of the system, a multi-pass approach with the DAST should be attempted with an off axis cavity. In this process non-collinear phase matching could yield a power increase by a factor of 2 due to the multiple passes. Another method to increase the average power would be to couple both seed lasers into one PM fiber and simultaneously pass them through a single PPLN fan crystal. The pump beam could be split and passed through the cascaded PPLN crystals as seen in Fig. 9.1.
method would serve to eliminate the final dichroic mirror in our developed system in which half of the combined OPA power is lost. The only real drawback with such a source is that the frequency separation between the two seed lasers should not decrease below 1 THz. If the two seed lasers are decreased below this limit then possible interference effects could occur or third-harmonic generation effects could take place.

![Figure 9.1. Possible THz source configuration.](image)

However, the foot print of the system is far too large to be used in the field. In order to reduce the size of the current system, the source should be redesigned into a complete rugged fiber package. Developing the system into a fiber package would not be without many challenges. Due to the effects of dispersion, multi-order fiber bundles would be required and crystal fiber packages would have to be developed.

In Chapter 7 an overview of RF and THz generation due to the interaction of an ultrafast laser pulse on YBCO was explored. We gave a detailed explanation of how THz waves were generated due to the change in the kinetic inductance as a function of quasiparticle density. In order to theoretically estimate the possible generation of microwave radiation, the RL RCE model was explored. In the RCE model, we discussed that the
relaxation period would have to be on the order of the time of flight of the quasi-particles around the ring. In order for microwaves to be generated the resistance change would have to be on the same order of magnitude. In future investigations, models should be developed that take the effects of vortex interaction into consideration. The materials that were investigated were high temperature superconductors, therefore vortex pinning may be a key feature in a future model. Initially we assumed that the process of ultrafast switching was due to the phonon and Cooper pair interaction alone. However, the vibrational motion from a number pined vortices being trapped in the incident laser cross section may account for the loss of current in the charged ring on the order of the nanosecond time scale.

Finally, in Chapter 8 we generated broadband GHz radiation using an ultrafast laser and multiple HTS thin film YBCO rings. We were able to characterize multiple parameters that dictated the frequency response and power generation amplitude from the rings. A variety of theoretical problems arise from the mechanism of how this thin film radiates in the microwave regime. Further investigation of this method would require exact resistance measurements of these rings similar to how a Josephson junction is measured. Such measurements may prove difficult due to the ultrafast relaxation of the resistance. However, through the resistance the source of the current dissipation may identify a key feature in the radiative process. In addition, electro-optic sampling should be investigated for the situation where the radiated electric field from the HTS ring modulates the refractive index of an EO active crystal. The generated THz waves could be analyzed through EO sampling and could provide a precise picture of the relaxation process at the location of the incident laser.
REFERENCES


pumped periodically poled lithium niobate,” in Advanced Solid-State Photonics, J. Zayhowski, ed., Vol. 83 of OSA Trends in Optics and Photonics (Optical Society of America, 2003), paper 339.


