INCREASED POWER, PULSE LENGTH, AND SPECTRAL PURITY
FREE-ELECTRON LASER FOR INVERSE-COMPTON X-RAY PRODUCTION
AND LASER INDUCED BREAKDOWN SPECTROSCOPY OF THIN FILM
PHOTOVOLTAICS

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To my Mom and Dad who have always supported me.
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ABSTRACT

The free-electron laser (FEL) system can be configured to produce X-ray or extreme ultraviolet (EUV) light via Compton backscattering and to perform many types of spectroscopy including laser induced breakdown spectroscopy (LIBS). In its most common incarnation, the FEL is limited by three major factors: average laser power, laser spectral purity, and laser pulse length. Some examples of the limitations that these shortcomings give rise to include limiting the range of remote spectroscopy, degrading spectroscopic precision, and lowering the attainable x-ray flux, respectively.

In this work, we explored three methods of improving the FEL. First, a beam expanding optic dubbed the TIRBBE was designed, built, and tested to prevent laser damage to the resonator mirrors and allow for higher average power. This optic had the added benefit of increasing the spectral purity. Second, an intra-cavity etalon filter dubbed the FROZEN FISH was designed, built, and tested to increase spectral purity and eliminate the frequency pulling (tendency of an FEL to pull towards longer wavelengths during a macropulse) all in a high damage threshold, fully wavelength adjustable package. Finally, a laser cooling scheme which allows for extension of the electron beam macropulse used to create the FEL light by counter-acting electron back-heating was explored. The first measurements of the back-heating temperature rise were taken, calculations of the required laser parameters were made, design of the full system was completed, and construction has begun.

Experimental work using LIBS to characterize thin film solar cells was also completed in anticipation of using the improved FEL to better characterize such materials. The frequency tunability and picosecond micropulse width of the FEL will allow for exploration of the frequency response of LIBS ablation and fine resolution of the make up of these materials with depth unattainable with a conventional fixed frequency nanosecond pulse laser.
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List of Acronyms

CIGS  Copper indium gallium diselenide $CuIn_{0.7}Ga_{0.3}Se_2$

DHS  Department of Homeland Security

EDX  Energy dispersive x-ray spectrometry

EUV  Extreme ultraviolet

FEL  Free-electron laser

FROZEN FISH  Filter realizing optimized and zealous enhancement of Near-IR FEL Intensity Spectrum

FTIR  Fourier transform infrared (spectroscopy)

HeNe  Helium-Neon laser

IR  Infrared

LIBS  Laser induced breakdown spectroscopy

LIDT  Laser induced damage threshold

ND filter  Neutral Density filter

NSF  National Science Foundation

PFN  Pulse forming network

RF  Radio frequency

TIR  Total internal reflection

TIRBBE  Total internal reflection Brewster beam expander
**TPI** threads per inch

**UH** University of Hawaii at Manoa

**XPS** X-ray photoelectron spectrometry

**XRD** X-ray diffraction
Introduction

The free-electron laser (FEL) group here at the University of Hawaii is currently under contract with the Department of Homeland Security to produce a compact high brightness x-ray source. This source is destined for use in detection of clandestine nuclear materials as a tightly focused source, one much more beam-like than the typical 'flashlight' mode of a electron beam colliding into a magnesium or aluminum plate like a typical dentist x-ray, is required. Our proposed source scatters IR photons produced from the FEL off of the electron beam being used to produce the FEL light. The scattering shifts the frequency of the IR photons into the x-ray regime. In order to produce any appreciable flux of x-rays, the IR laser and electron beam must have high luminosity at the interaction point. There are two critical requirements to this proposed source: a long, tightly focused electron beam and a commensurately long, tightly focused IR laser beam.

Our accelerator system at UH is already capable of the focusing needed for the electron beam, but the electron beam pulse length is limited by a phenomenon know as back-heating. To increase the pulse length and ultimately the x-ray flux, we are implementing a laser cooling scheme to combat the back-heating problem. This work is described in chapter 4.

The IR laser for our x-ray source must be very high brightness to take advantage of the electron beam parameters (i.e. there must be enough photons to collide with all of the available electrons). This level is not attainable in the FEL, but can be achieved by stacking pulses from the FEL on top of each other in an ultra low-loss storage cavity. However, even the highest quality mirrors cannot handle the power this this storage cavity. We propose a beam expanding optic to be placed in the resonator cavity near each mirror to alleviate this problem. In fact this same optic will be needed to avoid damage to the
mirrors in the FEL itself once we achieve longer electron beam pulse lengths available with the laser cooling scheme. This optic is described in chapter 2.

Two other challenges to producing high brightness IR in our storage cavity are the fact that the FEL inherently produces a spectrum with significant sidebands (power in wavelengths off the main peak) and which pulls toward longer wavelengths as a macropulse (a 4-10 µs duration pulse consisting of a train of 1 ps micropulses of light separated by 350 ps) progresses. These two challenges can be dealt with using an optical filter in the resonator cavity which we describe in chapter 3. This filter not only removes the undesirable sidebands and wavelength pulling, but due to the nature of the FEL amplification mechanism, actually allows for higher brightness to build up at the desired wavelength which we show theoretically.

With the x-ray source years from realization, we looked for some more immediately available applications of our FEL where its unique properties may be of use. While working at the Hawaii Natural Energy Institute (HNEI), the author explored using laser induced breakdown spectroscopy (LIBS) on photovoltaic devices being created there. This method employs a high power laser to create a plasma from a material to be investigated. The light coming off this plasma as it relaxes gives us information on the constituents of the ablated material. Our work there with a nanosecond pulse Nd-YAG laser pointed out deficiencies in ablation uniformity and mechanism that may be alleviated by using the picosecond pulses of the FEL. This work has been submitted for publication to the journal Thin Solid Films. Some perfunctory experiments on the ablation characteristics of the FEL on these materials are pending.

Finally, as part of the study of lasers and general optics, we have devised a method to create Bessel beams from a standard Gaussian mode. Bessel beams have the unique property of not diverging over large distances. The beams described in appendix 2 diverge 110 times more slowly than a Gaussian beam with the same central spot size. This type of beam could be very useful in remote spectroscopy applications where a high intensity beam collimated over long distances could produce strong return signals. A Bessel beam storage cavity could also be constructed and would have the advantage of tightly collimated beam over a longer distance (an equivalently focused Gaussian beam diverges very quickly) which would ease the strict focusing requirement of the electron beam and IR laser for x-ray production. This work is published in the American Journal of Physics.
Chapter 2

The TIRBBE: Total Internal Reflection Brewster Beam Expander

A high power laser is desirable for many applications but especially important for the inverse-Compton X-ray source we are building at UH. With higher power, more X-ray producing IR photon-electron collisions occur, leading to a brighter X-ray source. The peak power attainable in high power laser resonator cavities is limited by both electron beam macropulse length and optical damage thresholds of the resonator components: the gain medium, output coupling optics, and resonator mirrors. Two separate lines of research, laser-cooling of the cathode and the feed-forward phase stabilization system, will allow longer usable electron beam macropulses, but optical damage is still an issue.

In an FEL, the gain medium is actually the electron beam, which doesn’t suffer optical damage, and the output couplers are typically high damage threshold ZnSe, CaF$_2$, or Al$_2$O$_3$ (sapphire). This leaves only the resonator mirrors susceptible to optical damage. The 2.046 m cavity of the Mark V FEL results in a $1/e^2$ beam radius (the typical Gaussian beam parameter) of $w = 1.54$ mm at $\lambda = 3\mu m$ at the mirror which, due to fluence thresholds for Ag-coated Cu mirrors, limits the macropulse energy. These mirrors have a fluence threshold of 30 J/cm$^2$ at 3.0 $\mu m$ with 1 $\mu s$ pulses before damage according to measurements done with the Mark III FEL. Multiplying by $A = \pi w^2/2$ we calculate an energy limit of 1.12 J per macropulse.
To prevent damage to the mirrors, the beam is typically expanded to spread the energy over a larger area. There are three typically used beam expander designs: an inverted (Keplerian or Galilean) telescope, grazing incidence mirrors, and Brewster angle incidence cylindrical lenses. A telescope [2] is a simple solution, but suffers from reflection losses due to the normal incidence to the lenses. Grazing incidence mirrors are far from an optimal solution since they are expensive, bulky, susceptible to optical damage (especially if coated) and only expand the beam in one axis as shown in figure 2.1. Finally, using two cylindrical lenses at the Brewster angle, while not suffering from reflection losses, results in higher loss due to the extra surfaces from the two lenses.

Doctor Szarmes and I have come up with a better solution: the Total Internal Reflection Brewster Beam Expander (TIRBBE). The TIRBBE expands the area of the beam at the mirrors by approximately a factor of 10, resulting in commensurately lower fluence at the mirrors, the capability of higher output fluence and power levels, and does so in a compact, low loss, high damage threshold package. Additionally, this design allows a single surface to be used in output coupling, where a solution like two cylindrical lenses would require two surfaces, resulting in higher loss. The TIRBBE is illustrated in figure 2.2. The low loss with output coupling and high intra-cavity power achievable with this design is especially important for operation of an inverse-tapered FEL (a desired area of future research for our group) as low loss is critical for the turn-on and deep saturation of the laser [3].

2.1 Optical and Mechanical design

2.1.1 Overview

The TIRBBE is a prism with optically powered surfaces (i.e. curvature that focuses or defocuses a beam) that expand the beam sufficiently in a compact and damage resistant design. Doctor Szarmes’s initial design consisted of a CaF$_2$ or sapphire prism with two flat surfaces and one ellipsoidally curved surface as shown in the solid model in figure 2.3.

The beam would enter through the flat surface, be totally internally reflected (TIR) and expanded at the middle surface, and exit the other flat surface. While this design theoretically met the requirements for beam expansion, it was impossible to manufacture (approximately 20 different vendors were contacted) as the curved surface required different radii of curvature for the tangential and sagittal rays. Instead, we made the input face cylindrically curved to expand the beam in the tangential direction and the TIR face
Figure 2.1: Grazing incidence mirror for beam expansion. The size, cost, and difficulty of alignment make a grazing incidence mirror a sub-optimal solution to beam expansion at the mirrors.

Figure 2.2: TIRBBE concept showing beam expansion at the curved input surface (expansion in the plane of the page) and TIR surfaces (expansion is in/out of the page).
cylindrically curved in the sagittal direction as shown in solid model in figure 2.4 which enabled LightMachinery Inc. to manufacture the optic shown in figure 2.5 out of Corning CaF$_2$. We chose the input surface for tangential expansion and the top for sagittal as this combination gave the easiest to manufacture optic (due to the larger radii of curvature that could be used; a smaller radii would have been required if the output surface was used for either expansion since it is closer to the cavity mirror). A provisional United States patent has been applied for under E. B. Szarmes and J. M. D. Kowalczyk, "Beam Expander," Provisional Patent Application 61353160, June 9, 2010 [4].

2.1.2 Detailed design

1. The beam should be enter and exit the optic at the Brewster angle $\theta_B$ so there are zero reflection losses. The Brewster angle was calculated in CaF$_2$ to be $\theta_B = 59.41^\circ$ at 3$\mu m$
Figure 2.5: Calcium Fluoride TIRBBE fabricated by LightMachinery Inc. The optic is shown in a prism mount during bench testing. Note the curved input (in foreground) and TIR (top of optic in this image) surfaces.

Figure 2.6: Schematic of ABCD matrix analysis of the TIRBBE showing the matrices used for each aspect of the optic.
where the FEL is typically operated using equation 2.1.3. The index of refraction was calculated using the Sellmeier equation [5] shown in equation 2.1.1 with parameters shown in table 2.1.2 obtained from [6]. The Brewster angle changes less than 2.5 degrees across the 1-10 µm wavelength range of the FEL operation, so reflection loss is negligible.

2. The optic should have a low absorption coefficient \( \alpha \) at the wavelengths of interest. At 3 µm, \( \alpha = 2 \times 10^{-4} \text{ cm}^{-1} \) leading to an overall absorption in the optic of \( 1 - I/I_o = 1 - \exp[-\alpha \times (L_1 + L_2)] = 0.06\% \). Here \( L_1 \sim L_2 \sim 15 \text{ mm} \) are the two travel distances through the optic from the center of the resonator before \((L_1)\) and after \((L_2)\) the hitting the TIR surface (top surface in figure 2.6).

3. The optic must expand the beam by a factor of \( \sim 3 \) in both the sagittal and tangential planes. The radii of curvature \( R_{\text{tan}} \) (tangential) and \( R_{\text{TIR}} \) (sagittal) were chosen to yield an expansion of 3.3x and 3.1x in the sagittal and tangential planes respectively. These calculations are detailed in this section.

4. The sagittal and tangential beam curvatures must be equal at the mirror so that an inexpensive circular mirror can be used. A curvature of \( R_{\text{mirror}} = 166 \text{ mm} \) in both planes was achieved with the design described in this section.

5. The optic must be compact to fit in the existing space. The final optic fit into an area of only 40 mm x 13 mm x 12 mm easing integration in our existing optical system.

6. The optical damage threshold for CaF\(_2\) of \( 10^5 \) J/cm\(^2\) must be met. This is well beyond the damage threshold of the Ag-coated Cu mirrors of 30 J/cm\(^2\) calculated in the introduction to this section.

\[
n_{\text{CaF}_2} = n = \sqrt{1 + \frac{B_1 \lambda^2}{\lambda^2 - C_1} + \frac{B_2 \lambda^2}{\lambda^2 - C_2} + \frac{B_3 \lambda^2}{\lambda^2 - C_3}}; \quad (2.1.1)
\]

In order to determine the radii of curvature needed for sagittal and tangential expansion, ABCD matrix analysis (or ray-transfer matrix analysis) [8] was used to predict the behavior of the nearly TEM\(_{00}\) Gaussian parallel polarized FEL beam. This was accomplished by tracing the beam through the optic in figure 2.6 and building the appropriate
matrices. Note that we used the 'reduced slope' form of the ABCD matrices where the input is of the form \[ d \]

\[
A = \begin{bmatrix}
\frac{d}{n\theta}
\end{bmatrix}
= \begin{bmatrix}
\text{distance from the optic axis} \\
\text{index of refraction of the material * angle to the optic axis}
\end{bmatrix}
\] (2.1.2)

First, the Brewster angle \( \theta_B \), refracted angle \( \phi \), and incident angle at the TIR surface \( \psi \) were calculated using equations 2.1.3, 2.1.4, and 2.1.5. These angles along with the size constraint of roughly 40 mm in length constrained the propagation lengths \( L_1 \) and \( L_2 \) through the optic.

\[
\theta_B = \arctan(n)
\] (2.1.3)

\[
\phi = \arcsin(\frac{\sin(\theta)}{n});
\] (2.1.4)

\[
\psi = \frac{\pi}{2} - \theta + \phi;
\] (2.1.5)

Next the ABCD matrices could be determined. For rays in the tangential plane, the curvature of the input surface has a focusing effect described by 2.1.6.

\[
M_{\text{tangential vacuum to TIRBBE}} = \begin{bmatrix}
\cos(\phi)/\cos(\theta) & 0 \\
\frac{\Delta n_e}{R_{in}} & \cos(\theta)/\cos(\phi)
\end{bmatrix}
\] (2.1.6)

\[
\Delta n_e = \frac{n \cos(\phi) - \cos(\theta)}{\cos(\phi) \cos(\theta)};
\] (2.1.7)

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
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<td></td>
</tr>
<tr>
<td>(B_2)</td>
<td>0.4710914</td>
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</tr>
<tr>
<td>(B_3)</td>
<td>3.8484723</td>
<td></td>
</tr>
<tr>
<td>(C_1)</td>
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<td></td>
</tr>
<tr>
<td>(C_2)</td>
<td>0.01007833 (\mu m^2)</td>
<td></td>
</tr>
<tr>
<td>(C_3)</td>
<td>1200.5560 (\mu m^2)</td>
<td></td>
</tr>
</tbody>
</table>

Table 2.1: Sellmeier coefficients for vacuum ultra-violet (VUV) grade CaF2.
However, rays in the sagittal plane are not deflected from the optic axis so matrix \(2.1.8\) is the identity. 

\[
M_{\text{sagittal vacuum to TIRBBE}} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \tag{2.1.8}
\]

Matrix \(2.1.9\) was used with lengths \(L_1\) and \(L_2\) to determine the change in the beam as it propagates those distances through the optic. 

\[
M_L = \begin{bmatrix} 1 & L/n \\ 0 & 1 \end{bmatrix} \tag{2.1.9}
\]

At the TIR surface, the tangential rays are not deflected from the optic axis so matrix \(2.1.10\) is the identity. 

\[
M_{\text{TIR surface tangential}} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \tag{2.1.10}
\]

On the other hand, sagittal rays see a focusing effect described by matrix \(2.1.11\). 

\[
M_{\text{TIR surface sagittal}} = \begin{bmatrix} 1 & 0 \\ -2n \cos(\psi)/R_{\text{TIR}} & 1 \end{bmatrix} \tag{2.1.11}
\]

At the flat output surface, the matrices for tangential and sagittal rays are described in \(2.1.12\) and \(2.1.13\). 

\[
M_{\text{TIRBBE to vacuum tangential}} = \begin{bmatrix} \cos(\theta)/\cos(\phi) & 0 \\ 0 & \cos(\phi)/\cos(\theta) \end{bmatrix} \tag{2.1.12}
\]

\[
M_{\text{TIRBBE to vacuum sagittal}} = \begin{bmatrix} 1 & 0 \\ 0 & 1 \end{bmatrix} \tag{2.1.13}
\]

Finally, the beam propagation from the optic to the cavity mirror is described by matrix \(2.1.14\) with \(L_{\text{vacuum}} = 110\) mm. 

\[
M_{\text{vacuum}} = \begin{bmatrix} 1 & L_{\text{vacuum}} \\ 0 & 1 \end{bmatrix} \tag{2.1.14}
\]

To determine the total effect of the optic and propagation of the beam to the mirror, matrices \(2.1.6\) through \(2.1.14\) are multiplied together as shown in equation \(2.1.15\) for tangential rays and equation \(2.1.16\) for sagittal rays. 

\[
M_{\text{total tangential}} = M_{\text{vacuum}} \ast M_{\text{TIRBBE to vacuum tangential}} \ast M_L \ast M_{\text{TIR surface tangential}} \ast M_L \ast M_{\text{TIRBBE to vacuum tangential}} \tag{2.1.15}
\]

10
These total matrices can be used in the prescription described in equations 2.1.17 through 2.1.20 to treat Gaussian beams [8] and determine the beam width \( w \) and the beam radius \( R \) at the mirror in both the tangential and sagittal planes. Here \( n_1 \) and \( n_2 \) are both the index of refraction of vacuum.

\[
\hat{q}_{1,2} = (z + iz_R)/n_{1,2}
\]  

\[
M_{\text{total}} = \begin{bmatrix} A & B \\ C & D \end{bmatrix}
\]  

\[
\hat{q}_2 = \frac{A \ast \hat{q}_1 + B}{C \ast \hat{q}_1 + D}
\]

\[
\frac{1}{\hat{q}_{1,2}} = \frac{n_{1,2}}{R(z)} - i \frac{\lambda_o}{\pi w^2(z)}
\]

The input beam has a Rayleigh range of \( z_R = \pi w_o^2/\lambda = 532 \text{ mm} \). With the optic at \( z = 873 \text{ mm} \), the beam at the cavity mirror was made to have a radius of 166 mm in both the sagittal and tangential planes by choosing \( R_{in} = 85 \text{ mm} \) and \( R_{TIR} = -46 \text{ mm} \) (a positive radius indicates a concave radius of curvature from the perspective of the incoming beam, while a negative indicates convex).

### 2.2 Dispersion Theory

While the original intention of the TIRBBE was to expand the intra-cavity beam to avoid mirror damage, the dispersion arising from the multiple passes through the \( \sim 3 \text{ cm} \) optic is significant and causes pulse broadening. As a simple example, Saleh [5] shows how a Gaussian (in time) pulse will be broadened according to equation 2.2.1

\[
\tau = \tau_o \sqrt{1 + \left(\frac{z}{z_o}\right)^2}
\]  

(2.2.1)
where $\tau_o$ is the initial $1/e^2$ intensity pulse width, $z$ is the distance in the medium, $z_o = -\pi \tau_o^2 / D_v$, and $D_v$ is the dispersion coefficient defined by equation 2.2.2.

$$D_v = \frac{\lambda^3}{c^2} \frac{d^2 n(\lambda)}{d\lambda^2}$$

(2.2.2)

As an example calculation, at 3 $\mu$m the dispersion coefficient of Calcium fluoride is $D_v = -6.71 \times 10^{-25}$ $s^2/m$. Over 300 passes through the length of the TIRBBE (the approximate number of passes that an optical pulse makes during our typical operating electron beam macropulse length of 4 $\mu$s), a Gaussian optical pulse with $\tau_o = 1$ ps will broaden to $\tau = 2.17$ ps. Shorter pulses broaden even more significantly: a 0.1 ps pulse (representative of the spikes seen in figure 2.8) broadens to 19.2 ps. These simple calculations hint that the TIRBBE should smooth out the optical spiking (which refers to high frequency spikes in the optical pulse envelope, described in detail at the beginning of chapter 3) while only increasing the width of the picosecond optical pulses by about a factor of 2. However, the FEL gain curve imparts a quadratic phase to the optical field which is canceled by the TIRBBE dispersion on the scale of the entire micropulse as will be seen below.

Since the TIRBBE is in many ways like a prism, there are actually two types of dispersion for light going through it: the material dispersion described above and angular dispersion from the slightly different path that different wavelengths take through the optic. Figure 2.7 shows an exaggerated description of angular dispersion from which a simple geometric argument yields the dispersion in equation 2.2.3. Angular dispersion turns out to be 100 times smaller than the material dispersion and so it was neglected in these simulations.

$$\left[ \frac{d^2 \phi}{d\omega^2} \right]_{angular} = \frac{L \lambda^3}{2\pi^2} \frac{1}{n^3} \left( \frac{dn}{d\lambda} \right)^2$$

(2.2.3)

The dispersion due to the optic was integrated into the FEL simulation code originally generated by Dr. Szarmes by taking the Fourier transform of the optical pulse after each pass through the resonator, multiplying by the transfer function for the dispersive optic in equation 2.2.4 and then taking the inverse Fourier transform to restore the time-domain optical pulse before the next cycle of interaction with the electron beam.

$$H(\nu) = \exp \left[ i\pi D_v \nu^2 \right]$$

(2.2.4)
Figure 2.7: Diagram of how angular dispersion arises from the differing refraction angles for different wavelengths. The different path lengths in calcium fluoride and free space which different wavelengths travel through changes their relative phase (i.e. the optic has angular dispersion).

Note the group delay term has been ignored in the transfer function as it simply causes a delay in the pulse which we compensate for in the FEL by shortening the cavity. Example results of simulations with and without the dispersive effects due to the TIRBBE in the FEL cavity are shown in figure 2.8. The pulse is slightly broadened, but the optical spiking is completely eliminated. As noted above, the lack of significant broadening is expected, due to the nature of the FEL gain. The small signal FEL gain from reference [9] is shown in equation 2.2.5. Here j is a dimensionless current density which depends on parameters of the laser and $\nu_o$ is likewise dimensionless and proportional to minus the frequency. We have calculated that at the peak of the gain curve (which corresponds to the frequency that builds up in the resonator), the second derivative of the phase term is positive and imparts a phase $d^2\phi/d\omega^2 = 3.9 \times 10^{-26} 1/s^2$ to the amplified light. Comparing this to the value of $d^2\phi/d\omega^2 = D\nu z/(2\pi) = -1.28 \times 10^{-26} 1/s^2$ from the dispersion of the TIRBBE, we can see that these two phases act to cancel each other, leading to less broadening.

$$gain_{small\ signal} = j \left[ \frac{(2 - 2 \cos \nu_o - \nu_o \sin \nu_o)}{2\nu_o^3} + i \frac{(2 \sin \nu_o - \nu_o - \nu_o \cos \nu_o)}{2\nu_o^3} \right]$$ (2.2.5)

As an added benefit, the TIRBBE makes the FEL power output less sensitive to cavity detuning or slight shortening of the resonator cavity to maximize laser power. Detuning is necessary in order to compensate for the speed of light in the cavity being slightly less than c due to the slight index of refraction shift of the electron beam and Gouy
phase shift of the fundamental Gaussian mode supported by the resonator [5]. A simulation of a typical detuning curve without the TIRBBE is shown in figure 2.9. With TIRBBEs at either end of the resonator, a broader range of cavity lengths allow the laser to lase as evidenced by figure 2.10. This gives the FEL operator more control over the pulse output power as the pulse shape is very constant over the detuning curve and the power varies slowly with detuning.

While the average macropulse output power is lowered with the TIRBBEs in the cavity, it is much more constant over time as can be seen by comparing figures 2.11 and 2.12. This makes the output power within the macropulse and between macropulses much more constant and is important in applications such as LIBS where power fluctuations will lead to different amounts of ablated material. In fact, for LIBS the micropulse power spiking is another potential problem as the high power levels could lead to non-linear effects such as multi-electron ionization and multi-photon processes. Constant power levels are also highly desirable for spectroscopy where again high spikes can unintentionally lead to the same non-linear effects.

### 2.3 Bench Testing

The TIRBBE turned out to be difficult to manufacturer for a number of reasons. The two materials we wanted to construct the optic of, calcium fluoride and sapphire, are
Figure 2.9: Simulation of output energy vs. detuning and power vs. pulse time without the TIRBBE in the cavity. Note the lack of uniformity of the pulses as a function of detunings and the abrupt changes in output energy compared to the case with the TIRBBE. When comparing this figure to figure 2.10, note that the optical power is much higher (250 MW compared to 25 MW) at 0.00 mils detuning.
Figure 2.10: Simulation of output energy vs. detuning and power vs. pulse time with the TIRBBE in the cavity. Note the uniformity of the pulses over a wide range of detunings and the slow change in output energy compared to the case without the TIRBBE.

Figure 2.11: Simulation of average macropulse output power as a function of pass number (300 passes corresponds to a 4 \( \mu s \) macropulse) at \( \lambda = 3 \ \mu m \) without TIRBBEs in the resonator cavity. Note the fluctuating output power.
difficult to machine, mostly due to the hardness (163 Knoop, 4 Mohs for calcium fluoride, 2025 Knoop, 9 Mohs for sapphire, for comparison diamond is 7000 Knoop, 10 Mohs [10]). The small size of the optic made securing it for cutting the optic and polishing with the cylindrical polishers difficult according to LightMachinery Inc., the company that was eventually able to manufacture it.

Once the optic was in hand, we set out to verify that the optic was made to specifications. First, we measured the radii of curvature of the curved surfaces by impinging a collimated ~5 mm transverse diameter helium-neon (HeNe) laser approximately normal to the curved surface and finding the distance at which the laser is focused. The measured distance is the focal length $f = 2R$, which were approximately equal to the design values for both curved surfaces within the error of this crude measurement.

Next we set out to confirm that the optic operated as expected at the HeNe wavelength of $\lambda = 632.8$ nm. The setup is pictured in figure 2.13. The HeNe beam was collimated entering the TIRBBE so that the beam would not expand beyond the aperture of the Cohu CCD camera used to measure the width of the beam. The beam radius $w$ was measured as a function of longitudinal distance $z$, fit to equation 2.3.1 and plotted in figures 2.14 and 2.15 with and without the TIRBBE inserted into the beam line.

$$w(z) = \sqrt{\frac{2R\lambda}{\pi}} \sqrt{1 + \left(\frac{z}{z_R}\right)^2}$$  \hspace{1cm} (2.3.1)

Figure 2.12: Simulation of average macropulse output power as a function of pass number (300 passes corresponds to a 4 $\mu$s macropulse) at $\lambda = 3 \mu$m with two TIRBBEs in the resonator cavity. Note the constant output power once the laser turns on.
Figure 2.13: Optics table setup for bench test of TIRBBE. The components are (A) Melles Griot HeNe laser, (B) mirror, (C) focusing lens, (D) 35 µm pinhole, (E) collimating lens, (F) iris, (G) kinematic mount with TIRBBE, and (H) ND filters and CCD camera for capture and measurement of beam width. The ND filters were later removed and two polarizers were inserted between (E) and (F) to attenuate the beam as the ND filters introduced too much distortion.

Figure 2.14: Measured tangential beam width vs. z axis position of a HeNe laser. First the Rayleigh range \( z_R \) and location of the waist for the input beam were obtained by fitting measurements to equation 2.3.1 (green dashed line). Next the TIRBBE was inserted into the beam line and measurements of the beam width were taken to obtain the modified \( z_R \) and waist location (magenta dotted line).
As the figures indicate, the expander fulfills its basic function of expanding the beam in both the tangential and sagittal planes. However, when the input beam parameters derived from the fits to the input beam were used as inputs to the matrices in Equation 2.1.15 and 2.1.16, the calculated output didn’t match the measured output beam of the TIRBBE as shown in Figures 2.17 and 2.18. We changed to radii of curvature in the matrix calculations to better reflect the actual output of the TIRBBE and included those fits in the aforementioned figures.

There are multiple possible sources for the discrepancy effective radius of curvature. One possibility is that the actually cut tangential radius of curvature is in fact closer to the modified value of 107 mm than the design values. Another possibility is that the radius of curvature was not cut exactly in the correct location in the optic. As a point of interest, the first analysis we did of the tangential beam properties showed that the TIRBBE was actually very far off the design specification. In that initial experiment, glass neutral density (ND) filters were used to attenuate the beam before reading by the camera. These filters had inclusions in the glass that distorted the beam. In later characterizations of the TIRBBE, a set of two high quality quartz polarizers were used to attenuate the beam, leading to a much better beam image and fit to a Gaussian. These improved measurements showed
Figure 2.16: Transverse mode profile of the HeNe beam exiting the TIRBBE. Note that in the saggital plane (y-z, with z into the page), the profile is a good fit to a Gaussian, but in the tangential (x-z) plane the fit is much more poor. This is a likely cause of the large discrepancy between the design and as-built effective curvature in the tangential plane.

much better agreement between the design and as-built radii of curvature, though it was still not perfect.

While the HeNe mode was a very uniform circle in transvers profile, it was distorted by the TIRBBE as can be seen in figure 2.16, most likely due to the relaxed surface figure of the optic (the vendor was only willing to make the part on a best effort basis). This made the fits to a Gaussian profile inaccurate in figures 2.17 and 2.18 and also contributed to the discrepancy in design and as-built radius of curvature.

To compensate for the measured error and resulting effective radius in the as-built optic, its mount was designed to increase the distance from the optic to the mirror, allowing the beam to expand more, compensating for the reduced defocusing of the TIRBBE in the tangential plane. This error in effective radius required a resonator cavity mirror that was slightly torroidal instead of circular.
Figure 2.17: Comparison of tangential output beam of the actual, as-designed, and as-built TIRBBE. The actual output beam (red solid line) has a nearly identical waist to the beam predicted by ABCD analysis with $R_{in} = 85$ mm (green long dashed line), but is slightly less diverging. Changing the radius of curvature of the input surface in the ABCD analysis to $R_{in} = 107$ mm (blue short dashed line) yields a better fit to the actual.

Figure 2.18: Comparison of sagittal output beam of the actual, as-designed, and as-built TIRBBE. The actual output beam (red solid line) has a nearly identical waist and Rayleigh parameter as the beam predicted by ABCD analysis with $R_{TIR} = 46$ mm (green long dashed line).
2.4 Stable Mode Analysis

With the as-built TIRBBE different from the design, we were required to change the mirror at the TIRBBE end of the cavity to match the optic’s expanding properties. There was some room within the vacuum chamber that contains the mirror and output coupling optics (the optics box) to move the TIRBBE further from or closer to the mirror. This allowed us to find a placement of the optic and mirror curvature combination that produced a stable mode in the resonator described by Siegman [8] in equation 2.4.1:

\[
\frac{1}{\hat{q}} = \frac{D - A}{2B} \mp \frac{1}{B} \sqrt{\left(\frac{A + D}{2}\right)^2 - 1}
\]  

(2.4.1)

Here A, B, C, and D are the entries in the ABCD matrices that describe a round trip through the resonator described by matrix 2.4.2:

\[
M_{\text{resonator}} = M_{\text{TIRBBE mirror}} \ast M_{\text{cavity mirror}}
\]  

(2.4.2)

Matrix \( M_{\text{TIRBBE mirror}} \) consists of propagation from the resonator cavity center to the TIRBBE (distance \( L_T \)), the TIRBBE matrix [2.1.15] or [2.1.16] propagation from the TIRBBE to the mirror (distance \( L_{TM} \)), reflection off the TIRBBE mirror, and propagation back to the cavity center by swapping the A and D entries (denoted by a $ below) of the first three matrices and applying in reverse order as shown in equation 2.4.4. A similar procedure yields propagation to the far cavity mirror (without the TIRBBE) and back shown in equation 2.4.5:

\[
M_{\text{TIRBBE mirror}} = \begin{bmatrix} 1 & L_T \\ 0 & 1 \end{bmatrix} M_{\text{Stan, sag}} \begin{bmatrix} 1 & L_{TM} \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ -2/R_{\text{TIRBBE mirror}} & 1 \end{bmatrix} \times (2.4.3)
\]

\[
M_{\text{TIRBBE mirror}} = \begin{bmatrix} 1 & L_{\text{total}} \end{bmatrix} M_{\text{TIRBBE mirror}} \begin{bmatrix} 1 & L_T \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ -2/R_{\text{TIRBBE mirror}} & 1 \end{bmatrix} \times (2.4.4)
\]

\[
M_{\text{cavity mirror}} = \begin{bmatrix} 1 & L_{\text{cavity/2}} \\ 0 & 1 \end{bmatrix} \begin{bmatrix} 1 & 0 \\ -2/R_{\text{cavity mirror}} & 1 \end{bmatrix} \begin{bmatrix} 1 & L_{\text{cavity/2}} \\ 0 & 1 \end{bmatrix} \times (2.4.5)
\]

With the chamfered edge nearest the TIRBBE cavity mirror at 92.4 mm from the mirror, a stable resonator mode can be achieved with TIRBBE from 1 to 4.5 \( \mu m \). Outside
Figure 2.19: Stable fundamental Gaussian resonator modes in the tangential plane with one as-built TIRBBE. For $\lambda < 1.0 \mu m$ and $\lambda > 4.5 \mu m$, there is no stable mode within the resonator.

of this range, the mode is unstable and diffractive losses significantly degrade the power that can build up in the cavity. The beam widths as a function of distance from the center of the resonator cavity are shown in figures 2.19 and 2.20. In these figures, the TIRBBE is on the right (positive distance) side of the plot (near 900 mm from the center at 0 mm) and significantly expands the beam beyond what is shown (out to $\sim 3.5$ mm). It is interesting to note that for longer wavelengths, the stable mode has the beam expanded at the mirror farthest from the TIRBBE, actually leading to expansion of the beam at both mirrors with only one TIRBBE.

Movement of the TIRBBES along the axis of the resonator can have a dramatic impact on the stability. We found that if two TIRBBEs are used in a resonator cavity and kinematic mounts that allows translation of the TIRBBEs along the axis of the resonator are installed, stable mode can be found for wavelengths from $1 \mu m$ out to $5 \mu m$ with a $3 \text{ mm}$ displacement (the TIRBBEs move toward the mirrors in order to support longer wavelengths) of the optics. Modes for 1 and $5 \mu m$ are shown in figures 2.21 and 2.22 to illustrate that they are not too tightly focused (on the edge of stability) and the beam is still expanded to prevent optical damage.

2.5 Intra-cavity experiment

With the basic operation of the TIRBBE verified, we built an insertable mount shown in figure 2.23 to hold the optic in the beamline. The mount includes an outcoupling
Figure 2.20: Stable fundamental Gaussian resonator modes in the sagittal plane with one as-built TIRBBE. For $\lambda < 1.0\mu m$ and $\lambda > 4.5\mu m$, there is no stable mode within the resonator.

Figure 2.21: Tangential plane stable modes for the extreme wavelengths that can be supported in the UH FEL with two TIRBBEs on movable stages. Note the adequate expansion and relatively tight mode in the body of the resonator that leads to good coupling between the electron beam and optical mode.
Figure 2.22: Sagittal plane stable modes for the extreme wavelengths that can be supported in the UH FEL with two TIRBBEs on movable stages. Not the adequate expansion and relatively tight mode in the body of the resonator that leads to good coupling between the electron beam and optical mode.

plate off the Brewster angle to allow 10% output coupling from the cavity as illustrated in figure 2.24. The TIRBBE, output coupler, and $R_x=160$ mm, $R_y=149$ mm cavity mirror will be placed in the FEL resonator cavity operating at $3.00 \, \mu m$ to verify that this optic can operate as expected. The achieved power levels will be compared with those of the previous resonator setup to ensure that comparable power levels could be achieved.

Taking the average power from the simulation shown in figure 2.11 and calculating the fluence at the mirrors using $F = \frac{Power_{average} \times \tau}{A_{optical}}$ with $\tau$ the macropulse length and $A_{optical}$ the optical mode area we determined that with a $6\mu s$ usable pulse length and 10% output coupling, the mirrors will reach their damage threshold of $30 \, J/cm^2$. At that time it will be necessary to have a TIRBBE at both ends of the resonator, unless the laser is operated at the longer wavelengths where the beam is expanded at the cavity mirror opposite the TIRBBE.

2.6 Conclusion

The TIRBBE has been designed, manufactured, and a mount and mirror that allow a stable mode in the FEL resonator cavity have been constructed. In the future as the electron macropulse length is increased through the laser cooling (later in this dissertation) and feed-forward phase stabilization system [11], the TIRBBE will enable higher power to
Figure 2.23: Mount for the TIRBBE and output coupler in the FEL resonator cavity showing (A) TIRBBE, (B) CaF$_2$ output coupler, (C) output coupling mirror, (D) aluminium mount, and (E) kinematic mount to move the TIRBBE and output coupler combination up and down moving it in/out of the laser beam line.

Figure 2.24: Top view of TIRBBE and output coupler. The majority of the power in the cavity bounces between the near and far cavity mirrors as it is amplified each pass through the FEL. About 10% of the power is coupled out of the cavity by the plate slightly off the Brewster angle.
be achieved in the cavity, resulting in increased brightness for applications such as the inverse-Compton X-ray source we are currently constructing.
Chapter 3


As the optical field in an FEL grows during the turn on of the laser, the gain is reduced until it is equal to the cavity and output coupling losses [9]. The high optical fields give rise to two phenomenon that limit the utility of the laser: wavelength pulling and sideband instabilities. Wavelength pulling refers to the wavelength with peak intensity pulling toward longer wavelengths as the optical power builds up due to distortion in the gain curve at high power [9]. Sideband instabilities, or optical spiking, refers to high frequency spikes in the optical pulse envelope as shown in figure [3.1]. This is caused by the high field in the optical cavity conspiring with the undulator field to force the electrons to give up significant energy to the optical field (and consequently slowing down) before the electron beam has slipped behind the optical pulse. The change in velocity moves the electrons to a phase of the optical pulse where they are then poised to gain energy from the optical field, decreasing the field intensity in that temporal location of the pulse. This phenomenon feeds back on itself as once there is a small intensity modulation, the high and low intensity temporal parts of the pulse promote enhanced energy extraction and addition (respectively) to the optical pulse from the electrons. An intra-cavity optical narrow band-pass filter can eliminate both phenomenon resulting in a constant wavelength over the entire duration of
the electron macropulse (typically 4 to 10 $\mu$s with a 4 to 10 Hz repetition rate) and a much narrower and cleaner laser spectrum.

### 3.1 Review of previous intra-cavity filters

Intra-cavity filters have been proposed by a number of authors, most notably Cutolo et alii. They proposed electro-optic [11] and acousto-optic [12] filters where the sidebands experience high loss compared to the fundamental wavelength of light. The electro-optic filters have the disadvantages of requiring high voltage in the 1 to 10 kV range and a very narrow central peak that can hinder FEL startup. Acousto-optic filter could be a good choice, but require expensive specialized acousto-optic crystals that limit their bandwidth. For instance, germanium has the desired acousto-optic properties, but is only good below 4$\mu$m due to absorption and consequent optical damage at longer wavelengths. Other materials are required for operation in the visible and near IR. Additionally, matching the impedance of the power supply, transducer, and crystal opposes tunability. Both types of filters can generally not be used in a high power resonator due to damage thresholds of the materials required.

Multi-layer dielectric mirrors have also been proposed by Goldstein et al. [13], but these are complicated, expensive, not tunable, not producible from off-the-shelf parts.
Crisafulli et al. proposed a birefringent etalon filter specifically for controlling the chaotic frequency evolution in the inverse-tapered FEL [3]. It is that filter concept we designed and tested.

3.2 Theory

A properly spaced etalon can provide a means of filtering a laser beam, but in order for the etalon to be a narrow function of wavelength it must have high finesse [5] requiring the reflecting surfaces to have high reflectance at the desired pass wavelength according to equation [3.2.1]

$$F = \frac{\pi \sqrt{r}}{1 - r}$$

(3.2.1)

Here $r$ is the field reflection coefficient. A normal etalon would filter an incoming laser beam as any wavelength off the design wavelength would experience some degree of destructive interference and be attenuated. Typically the laser is normally incident leading to significant first surface reflection and high loss. However for an intra-cavity etalon filter, we need very low loss else the laser may not build to saturation as the gain will be less than the total loss of the cavity. This motivates us to put the etalon at the Brewster angle so that there is zero reflection at all surfaces at the design wavelength. The Brewster angle is a weak function of wavelength, so there is very little reflection even significantly off the design wavelength. Without reflection, the etalon effect doesn’t work as there are no multiple beams to constructively/destructively interfere. However the Brewster angle only exists for P-polarized light (the FEL is P-polarized by its nature). If off design wavelengths could somehow be changed from p to S-polarized by the two plates making up the etalon gap, they would experience significant reflection and the etalon effect would work as in the normal incidence case. Birefringent crystals such as sapphire (crystalline Al$_2$O$_3$) do precisely that: they rotate the polarization of the incoming light a particular amount per unit length of the crystal. If the crystal properties and length are chosen correctly, incident P-polarized light at the design wavelength can be rotated through S-polarized and back to P-polarized before hitting the etalon gap. Then the now P-polarized light is incident at the Brewster angle (both exiting and entering the crystal) and experiences zero reflection. However, any wavelength off the design wavelength will have some S-polarization when it hits the etalon gap, and experience reflection and the etalon filtering effect. This is the
basic principle behind the FROZEN FISH presented here. An example of the filter field transmission is shown in figure 3.2 and is described subsequently.

### 3.2.1 Mathematical Analysis

In order to calculate the polarization rotation of the sapphire plates, we used the following Sellmeier coefficients in 3.2.1 and 3.2.1 from [14] with equation 2.1.1 to determine the ordinary and extraordinary indices of refraction, $n_o$ and $n_e$.

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B_1$</td>
<td>1.431349</td>
<td></td>
</tr>
<tr>
<td>$B_2$</td>
<td>0.6505471</td>
<td></td>
</tr>
<tr>
<td>$B_3$</td>
<td>5.341402</td>
<td></td>
</tr>
<tr>
<td>$C_1$</td>
<td>5.279926e-3</td>
<td>$\mu$m$^2$</td>
</tr>
<tr>
<td>$C_2$</td>
<td>1.4238265e-2</td>
<td>$\mu$m$^2$</td>
</tr>
<tr>
<td>$C_3$</td>
<td>3.25017834e2</td>
<td>$\mu$m$^2$</td>
</tr>
</tbody>
</table>

Table 3.1: Sellmeier coefficients for ordinary rays in sapphire.

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>$B_1$</td>
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<td></td>
</tr>
<tr>
<td>$B_2$</td>
<td>0.5506914</td>
<td></td>
</tr>
<tr>
<td>$B_3$</td>
<td>6.592737</td>
<td></td>
</tr>
<tr>
<td>$C_1$</td>
<td>5.480413e-3</td>
<td>$\mu$m$^2$</td>
</tr>
<tr>
<td>$C_2$</td>
<td>1.479943e-2</td>
<td>$\mu$m$^2$</td>
</tr>
<tr>
<td>$C_3$</td>
<td>4.0289514e2</td>
<td>$\mu$m$^2$</td>
</tr>
</tbody>
</table>

Table 3.2: Sellmeier coefficients for extraordinary rays in sapphire.

The actual index of refraction experience by an incident laser depends on the angle $\theta_{kc}$ between its k-vector of the c-axis of the crystal. First we find the interior angle $\phi = \arcsin(\sin(\theta_B)/n_{ave})$ with $n_{ave} = (n_o + n_e)/2$ and then determine $\theta_{kc}$ using equations 3.2.2, 3.2.3, and 3.2.4. With $\gamma = 90^\circ$ and $\alpha = 29^\circ$ for the sapphire crystals, we can then calculate the angle and determine the extraordinary index using 3.2.5.

$$\hat{k} = \begin{bmatrix} -\sin(\phi) \\ \cos(\phi) \\ 0 \end{bmatrix} \quad (3.2.2)$$
\[
\hat{c} = \begin{bmatrix}
\cos(\gamma) \cos(\alpha) \\
\sin(\gamma) \cos(\alpha) \\
\sin(\alpha)
\end{bmatrix} 
\] (3.2.3)

\[
\theta_{kc} = \arccos(\hat{k} \cdot \hat{c}) 
\] (3.2.4)

\[
n_e(\theta_{kc}) = \left( \frac{\cos^2(\theta_{kc})}{n_o^2(\lambda)} + \frac{\sin^2(\theta_{kc})}{n_{re}^2(\lambda)} \right)^{-1/2} 
\] (3.2.5)

Next we determine the transmission polarization matrices at the entrance and exit of the crystals. We use the Fresnel equations [5] to determine the input and output reflectance and transmission in equations 3.2.6 through 3.2.11.

\[
r_{ip} = \left( n_{ave} \cos(\theta_B) - \cos(\phi) \right) / \left( n_{ave} \cos(\theta_B) + \cos(\phi) \right) 
\] (3.2.6)

\[
r_{is} = \left( \cos(\theta_B) - n_{ave} \cos(\phi) \right) / \left( \cos(\theta_B) + n_{ave} \cos(\phi) \right) 
\] (3.2.7)

\[
t_i = \begin{bmatrix}
t_{ip} & 0 \\
0 & t_{is}
\end{bmatrix} = \begin{bmatrix}
(1 + r_{ip}) / n_{ave} & 0 \\
0 & 1 + r_{is}
\end{bmatrix} 
\] (3.2.8)

\[
r_{op} = \left( \cos(\phi) - n_{ave} \cos(\theta_B) \right) / \left( \cos(\phi) + n_{ave} \cos(\theta_B) \right) 
\] (3.2.9)

\[
r_{os} = \left( n_{ave} \cos(\phi) - \cos(\theta_B) \right) / \left( n_{ave} \cos(\phi) + \cos(\theta_B) \right) 
\] (3.2.10)

\[
t_o = \begin{bmatrix}
t_{op} & 0 \\
0 & t_{os}
\end{bmatrix} = \begin{bmatrix}
n_{ave}(1 + r_{op}) & 0 \\
0 & 1 + r_{os}
\end{bmatrix} 
\] (3.2.11)

Next we determine the phase shift induced by the gap and crystal in equations 3.2.12 and 3.2.13 for use in calculating the polarization matrix for the etalon gap in equation 3.2.14.

\[
d_g = \frac{4\pi}{\lambda} g \cos(\theta_B) 
\] (3.2.12)

\[
d_c = \frac{2\pi L}{\lambda \cos(\phi)} \left( n_o(\lambda) - n_e(\lambda, \theta_{kc}) \right) 
\] (3.2.13)

32
\[ t_g = \begin{bmatrix} \frac{t_{tp} t_{op} e^{-id_g/2}}{(1 - r_{op}^2 e^{-id_g})} & 0 \\ 0 & \frac{t_{os} t_{os} e^{-id_g/2}}{(1 - r_{os}^2 e^{-id_g})} \end{bmatrix} \] (3.2.14)

The polarization retardation caused by the crystal in equation 3.2.18 is rotated into the P-S basis from the o-e basis using the rotation matrix \( R \) in equations 3.2.15, 3.2.16, and 3.2.17 before application in equation 3.2.19.

\[ \hat{p} = \begin{bmatrix} \cos(\phi) \\ \sin(\phi) \\ 0 \end{bmatrix} \] (3.2.15)

\[ \hat{o} = \frac{\hat{k} \times \hat{c}}{|\hat{k} \times \hat{c}|} \] (3.2.16)

\[ R = \begin{bmatrix} \hat{p} \cdot \hat{o} & \sqrt{1 - (\hat{p} \cdot \hat{o})^2} \\ -\sqrt{1 - (\hat{p} \cdot \hat{o})^2} & \hat{p} \cdot \hat{o} \end{bmatrix} \] (3.2.17)

\[ M = R^T \begin{bmatrix} 1 & 0 \\ 0 & e^{-id_c} \end{bmatrix} R \] (3.2.18)

\[ M_{total} = t_{co} M t_g M t_{ci} \] (3.2.19)

The top left of matrix 3.2.19 gives us the P-polarized transmission through the FROZEN FISH. While the thickness of the crystals \( L \) and incident angle \( \theta_B \) are fixed, the gap can be changed to alter the filter’s transmission properties. The fixed crystal thickness limits the wavelengths for which the filter has a narrow passband since it fixes the polarization at the gap. However, if the entire assembly is rotated about the optic axis, the amount of polarization rotation is altered since the c-axis, \( \hat{c} \), moves with the crystal while the input \( \hat{k} \) remains the same. This changes \( \theta_{kc} \), which changes the indices of refraction experienced by P- and S-polarized light, and consequently the amount of polarization rotation. Kinematics that allow this rotation will be a future enhancement to the current work.
Figure 3.2: Transfer function of the filter tuned for 2.226 µm. Here the gap phase change is $d_g = 46\pi$ for the 51.2 µm gap and the crystal phase change is $d_c = 11\pi$. Note the wider side lobes which are centered on wavelengths for which $d_c = 10\pi$ and $d_c = 12\pi$. The wider shape of the even-$\pi$ lobes was our motivation to choose the odd-$\pi$ configuration.

3.2.2 Filter Transmission

With this limitation on wavelengths that can pass through the filter in mind, we explored the best possible filter configurations and found that wavelengths with odd-$\pi$ phase changes through each crystal (i.e. $d_c = n\pi$ with $n$ odd) produced the narrowest filter shape with 100% transmission of the design wavelength. This was surprising since we were expecting less than 100% transmission at the etalon gap for S-polarized light since there is no Brewster angle. However it is the etalon effect and not the Brewster angle that produces zero reflection at the design wavelength for S-polarized light. For operation of an FEL however, the narrowest shape is not necessarily the best, since the wavelength of maximum gain changes as the optical power in the resonator cavity builds up [9]. We confirmed through simulation that the odd-$\pi$ configuration’s transfer functions are wide enough to allow the laser to turn on. An example odd-$\pi$ transfer function for the filter at 2.226 µm is shown in figure 3.2.

We were unable to achieve high power with so short a wavelength with the FEL at this time, so we looked at two adjacent wavelengths that the filter will pass: 3.035 and 3.420 µm the field transmission of which are shown in figures 3.3 and 3.4. Note that at 3.035 µm the filter pass band is wider than either the 3.420 or 2.226 µm cases. We chose to look at the filter transmission near 3.420 µm to verify our understanding of the filter.
Figure 3.3: Transfer function of the filter tuned for $3.035 \, \mu m$. Here the gap phase change is $d_g = 40\pi$ for the $60.7 \, \mu m$ gap and the crystal phase change is $d_c = 8\pi$.

Figure 3.4: Transfer function of the filter tuned for $3.420 \, \mu m$. Here the gap phase change is $d_g = 20\pi$ for the $34.2 \, \mu m$ gap and the crystal phase change is $d_c = 7\pi$. 
3.2.3 Wavelength Pulling

One of the challenges of using the FEL lies in the fact that the wavelength changes (or pulls) as the pulse power builds up. We ran simulations to see the effect of the filter on this phenomenon and plotted the results in figure 3.5. Without the filter, following some initial transients, there is a constant change in the wavelength over the length of an electron beam macropulse. With the 8 µs macropulses we are expecting from the feed-forward and laser cooling systems, we see a variation in wavelength of nearly 10% without the filter while with the filter installed in the cavity, we see a variation of less than 1% after the initial transients.

To confirm our simulations, we looked at a spectrogram of a 1 µs pulse from the FEL shown in figure 3.6 which shows approximately the same variation in peak wavelength per unit pulse time of 1.25% per µs as the simulation in figure 3.5.

3.2.4 Spectrum

The typical spectrum of an FEL is far from monochromatic, as seen in figure 3.6, but the chaotic time structure of the laser pulse can be smoothed with the FROZEN FISH. We ran simulations of the FEL with the filter and compared the spectra with and without the filter for the same two wavelengths we show the transmission functions for in figures 3.3 and 3.4. The results of those simulations are plotted in figures 3.7 and 3.8. As expected,
Figure 3.6: Measured spectrogram of the FEL tuned near 4.1 $\mu m$. Note the significant wavelength pulling of approximately 1.25% during the 1 $\mu s$ macropulse. An 8 $\mu s$ would lead to approximately 8 times the wavelength pulling, 10%, matching the simulation results in 3.5.
Figure 3.7: Simulated spectra at $\lambda = 3.035\mu m$ with and without the filter. The filter is in the even-$\pi$ (broader passband) mode illustrated in figure 3.3. Note the significantly higher peak brightness and elimination of sidebands in the filtered spectrum.

The filter significantly narrows the laser spectrum by only allowing power to build up at the design wavelength.

### 3.3 Optical and Mechanical design

We chose to use two 1.375 inch x 0.790 inch x 0.1260 inch sapphire plates we had in hand to construct the filter. These were more than large enough to accommodate the beam in the FEL. The rest of the filter was made from CNC machined aluminum according to the following requirements.

#### 3.3.1 Requirements

1. The beam should be enter and exit the filter at the Brewster angle $\theta_B$ so there are zero reflection losses. The Brewster angle was calculated in sapphire to be $\theta_B = 59.48^\circ$ at 3$\mu m$ where the FEL is typically operated using equation 2.1.3. The index of refraction was calculated using the Sellmeier equation [4] shown in equation 2.1.1 with parameters shown in table 3.2.1 and 3.2.1. The Brewster angle changes less than 1.2 degrees across the 1-4 $\mu m$ wavelength range of the FEL operation where sapphire is transmitting, so reflection loss is negligible.
Figure 3.8: Simulated spectra at $\lambda = 3.420\mu m$ with and without the filter. The filter is in the odd-$\pi$ (narrower passband) mode illustrated in figure 3.4. Note the significantly higher peak brightness and elimination of sidebands in the filtered spectrum. Also note comparable brightness achieved with narrow (this figure) and broader (figure 3.7) filter configurations.

2. The filter should have a low absorption coefficient $\alpha$ at the wavelengths of interest. At 3 $\mu m$, $\alpha = 2\times10^{-4} \text{ cm}^{-1}$ [15] leading to an overall absorption in the optic of $1 - I/I_o = 1 - \exp[-\alpha * 2L] = 0.013\%$ per pass. Here $L = 3.2 \text{ mm}$ is the travel distances through each sapphire plate.

3. The plates must be able to rotate the polarization through multiple $\pi$ phase shifts to allow for the filter to be used for multiple wavelengths without rotation about the optic axis. This requirements was met as seen by the multi-$\pi$ phase shifts in the previous section.

4. The two plates must be parallel. To accomplish this to the necessary precision, a two axis angular adjustment kinematic mount (similar to a standard mirror mount from a typical optics vendor such as Thorlabs Inc.) utilizing 100 thread per inch (TPI) screws was employed as shown in figure 3.11. The two plates had a slight wedge angle of 5 arc minutes and the two wedged sides of the plates made the etalon gap such that the wedge deflected the beam vertically. These two items enabled us to parallelize the plates by looking at the most intense deflected beams, which we knew came from the gap, and adjusting the two screws until there was a minimum number of interference
fringes across the beam. This design enabled us to achieve 1 interference fringe across the beam, as corroborated by our results below.

5. The initial gap must be on the order of 50 µm to allow for substantial overlap between the interfering beams in the etalon and a sharp filter shape. This was accomplished by placing a third adjustment screw on the sapphire plate mount that allowed for gross adjustment of the gap. The initial gap was set using a 50 µm thick piece of lens tissue.

6. The etalon gap must be adjustable in order to allow the correct gap for transmission at different wavelengths. For this a flexure stage was employed with a flexure arm of 1/2 inch that allowed for sub-micron adjustment of the gap when pushed with a 100 TPI screw. A schematic of the flexure concept is shown in figure 3.9. References [16] and [17] along with know-how from Germano Zerbini (former UH machinist) and Dr. Szarmes were used to design the mechanism that has zero angular deflection and could be adjusted with minimal force. The gap adjustments versus screw turns achieved is shown in table 3.3.1.

7. The optic must be compact to fit in existing space. The final optic fit into an area of only 64 mm x 50 mm x 44 mm easing integration in our existing optical system.

8. The optical damage threshold of $7 \times 10^6$ J/cm² for sapphire must not be exceeded. The typical intra-cavity laser power is four orders of magnitude below this.

<table>
<thead>
<tr>
<th># of turns</th>
<th>δ gap (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1/4</td>
<td>0.1588</td>
</tr>
<tr>
<td>1/2</td>
<td>0.6350</td>
</tr>
<tr>
<td>3/4</td>
<td>1.4288</td>
</tr>
<tr>
<td>1</td>
<td>2.5403</td>
</tr>
<tr>
<td>1 1/4</td>
<td>3.9694</td>
</tr>
<tr>
<td>1 1/2</td>
<td>5.7163</td>
</tr>
<tr>
<td>1 3/4</td>
<td>7.7811</td>
</tr>
<tr>
<td>2</td>
<td>10.1641</td>
</tr>
</tbody>
</table>

Table 3.3: Adjustment of gap distance versus number of turns on the 100 TPI screw

The complete filter which meets the above design goals is pictured in figures 3.10 and 3.11.
Figure 3.9: Illustration of the gap adjustment concept for the FROZEN FISH. The two sapphire plates are set at a nominal distance of 50 µm and adjusted to be parallel in a). In b), an actuator pushes sideways on one of the sapphire plate holders that is held in place by a thin flexible flexure made of aluminum. The plate moves closer to the other plate, changing the gap and the pass wavelength of the filter. Initially the actuator is just a screw, but an electronic actuator will be employed in the future to enable changing of the gap while the filter is installed in the laser vacuum system.
Figure 3.10: Top view of the FROZEN FISH illustrating the sapphire plate holders, flexures, etalon gap, and laser beam path through the etalon.

Figure 3.11: Angle view of the FROZEN FISH showing the parallelism adjuster screws for the rear sapphire plate holder and the adjuster screws for the etalon gap width. Each screw is a 100 thread per inch screw which was shown to be sufficient to obtain substantial overlap in the etalon and provide micron scale gap adjustment when coupled with the flexure stage.
3.4 Bench characterization

Before the filter is placed into the resonator cavity, the crystals were made parallel according to the preceding section, but the gap still needed to be set. In order to find a gap where the filter is effective, first a wavelength for FEL operation was chosen so that sapphire plates yield an odd-\(\pi\) phase retardation to give a narrow filter. We chose \(\lambda = 3.420\ \mu m\) as wavelengths in that region lased at sufficient power. The laser was activated and the gap (nominally set to \(\sim 50\ \mu m\)) was finely adjusted until the reflected signal from the filter was minimized. The setup for this procedure is shown in figure 3.12 where a detector was placed at J to capture the reflected signal. We were able to achieve a factor of 7 difference in the reflected signal as we changed the gap. While the exact gap was unknown, at the point of minimal reflection (maximum transmission) from the filter the gap must cause a multiple of \(2\pi\) phase shift at the design wavelength. Since the filter shape changes little with absolute gap (as long as the plates are sufficiently close so that the beams that reflect multiple times within the etalon overlap sufficiently), this procedure should set the filter in a useful configuration.

Once the filter was setup, we took measurements of the spectrum using the monochromator and setup in figure 3.12 with and without the filter in place at H over a range of wavelengths to map the transmission curve. The pick-off plate at E and detector at G were used to normalize the measurements of power taken at M. To calculate the power transmittance of the filter, we used equation 3.4.1.

\[
T_{\text{single pass}} = \left( \frac{\text{Power}_M/\text{Power}_G|_{\text{with filter}}}{\text{Power}_M/\text{Power}_G|_{\text{without filter}}} \right) \tag{3.4.1}
\]

The results of these measurements are shown in figure 3.13 along with the best fit theoretical curve. We had intended to look only at the P-polarized transmission, which shows the nice shapes in figures 3.3 and 3.4 but neglected to insert a polarizer to reject the S-polarized component. Nonetheless, the breakdown of the theoretical curve into the P and S-polarized components in figure 3.14 shows that we have achieved a good filter shape for P-polarized light. The transmitted P-polarized component is amplified in the FEL cavity; the S-polarized component is reflected out of the cavity on the next pass through the optical filter, so the filter as setup for this experiment passes 2.95 and 3.8 \(\mu m\) and predominantly rejects others. We had intended to setup the filter to pass 3.42 \(\mu m\) as well, however an angular misalignment or difference in the birefringent properties of the sapphire
crystals and the small jumps in the gap position adjustments made the filter shape less than ideal. The theoretical curves in figures 3.13 and 3.14 were generated assuming an incidence angle of 57.4° instead of 60° as designed. This essentially made for slightly less propagation distance in the crystal and less polarization rotation. Essentially the same curves could be generated assuming a 60° angle of incidence and slightly decreasing the birefrigence of the sapphire, leading to the same decrease in polarization rotation. Either stress caused by the small force holding the sapphire plates in place or frozen-in stress from manufacturing could have caused such a change in the birefringence. Additionally, the adjustment of the gap via the 100 TPI screws had small jumps (i.e. the screw did not turn smoothly) so continuous adjustment of the gap was not possible. This was most likely due to either the spring which keeps the flexure stage in good contact with the gap adjustment screw having too high of a spring constant or the flexure itself being too stiff and putting excessive force on the screw threads. Correction of both issues are being explored. Both will be eliminated in the next incarnation of the filter mount which has a mechanical actuator that will replace the screw assembly.

We have recently gained access to a Fourier transform infrared spectrometer (FTIR) which will use for more extensive characterization of the filter in the future. The FTIR
Figure 3.14: Theoretical power transmission of filter from 2.9 to 4.0 \( \mu \text{m} \). Note that the P-polarized transmission (which builds up in the FEL cavity) has strong minima and maxima, realizing a good filter. The S-polarized components that exit the filter are reflected out of the cavity on the next pass through the filter.

has the advantage of measuring the transmission of the filter much more quickly than stepping though wavelengths using FEL as was done here.

3.5 Conclusion

We have designed, simulated, constructed, and bench tested the FROZEN FISH and shown that it can greatly enhance the FEL in frequency and power output stability and spectral purity. Through our simulations we have shown that the frequency pulling effect is eliminated, the output power is constant after turn-on, and the spectral purity is improved by orders of magnitude. The as built FROZEN FISH has the mechanical precision required to obtain the parallelism and gap tuning needed to realize the properties evidenced by the simulation results presented.

During a post doctoral appointment at UH, we plan on performing experiments with the filter in the FEL resonator cavity to confirm the results of the simulations presented. We have already designed a mount and procured parts for a mechanical actuator that can change the gap of the filter in situ, allowing a broad number of wavelengths of light filtered with the FROZEN FISH to be produced on the fly.
Chapter 4

Laser cooling for extended pulse length operation of electron cathodes

As part of a Department of Homeland Security (DHS) project, we are currently trying to produce a compact high luminosity electron-photon colliding beam source (akin to an inverse compton X-ray source) using our IR FEL and electron beam. The system is pictured in figure 4.1. These types of X-ray sources are ideal for cargo inspection and detection of special nuclear materials (the main interest of the DHS), but have the ability to provide characterization information above and beyond what a typical IR LIBS system can. In fact, such as source has the potential to simultaneously provide both constituent and structural information from an IR LIBS and XRD hybrid system. To maximize the average radiated power and minimize the energy spread of X-rays produced by these sources, thereby maximizing their utility, the electron beam used both in the X-ray producing collisions and in the undulator to produce the laser light must be as long as possible with the minimum energy spread.

Currently, electron beam pulse lengths and energy spreads are limited by a phenomenon known as back-heating where the electrons emitted late in the RF accelerating field’s cycle are accelerated back into the cathode causing its temperature to increase during the pulse. This temperature increase causes an increase in the thermionic emission current from the cathode and subsequent beam loading (reduction of the RF accelerating field from the high current beam) [18] which then reduces the energy of electrons emitted later in
the pulse. Typically, back-heating and the induced energy slew are minimized by keeping pulse lengths short, $\sim 4\mu s$, leading to lower average power and a spread in wavelength. In this work, we aim to use surface cooling due to diffusion to cancel the back-heating effect. As shown qualitatively in figure 4.2 by maintaining the cathode slightly below its nominal temperature before the RF pulse, depositing enough thermal energy onto the surface of the cathode with a laser to bring it to nominal temperature, and relying on the cooling of the surface due to diffusion of heat from the laser pulse into the bulk of the cathode to be counteracted by back-heating, we attain a constant cathode temperature and the ability to increase the pulse length to $\sim 8\mu s$ while maintaining a constant energy electron beam.

To date, no publication of the time or temperature dependence of the back-heating phenomenon exists so this works constitutes the first known quantitative data available. The program of research for the laser cooling concept consists of the following 8 major milestones:

1. Measurement of the gross time resolved back-heating phenomenon with resolution on the order of 100 ns.

2. Measurement of the spatial distribution of back-heating over times scales on the order of milliseconds.

3. Calculation of the necessary laser pulse characteristics needed to cancel the back-heating effect using a simple first order model.

4. Refinement of the heat equation model to include the temperature dependent thermal diffusivity of the cathode.
Figure 4.2: Laser cooling concept showing how the diffusion of heat $Q$ deposited by the laser (green) cancels the heat added to the cathode by backheating (red) resulting in a constant temperature (blue).

5. Construction of the necessary electronics and optics to properly time and transport the laser pre-pulse.

6. Test of the laser cooling technique in-system.

7. Measurement of time and spatial dependence of the back-heating with fine (500 fs) resolution using a streak camera.

8. Testing of long term operation of the laser cooling technique.

To date the first four items have been completed. The remainder will be completed by the author during a post-doctoral appointment immediately following graduation, with items 5 and 6 set to be complete in the next 6 months.

4.1 Theory: cathode current and back-heating

The problem that this research addresses is that beam loading induced electron energy slew limits electron beam pulse length. In order to maximize x-ray flux, we need precise alignment of the laser e-beam interaction and micron spot sizes for each. If there is an energy slew in the electron beam (due to beam loading) then, due to the dispersive
e-beam and light focusing optics used to align the interaction, the beam will wander off the optimal alignment. This causes there to be less collisions and consequently less x-rays. If the pulse is longer, then the beam will wander so far off the optimal alignment, that we’ll produce almost no x-rays. So a longer e-beam pulse is of no utility if the energy changes dramatically over the pulse time.

Additionally, the energy slew will cause an energy spread in the electrons that do collide with IR photons, leading to an undesirable spread of energy in the produced X-rays.

The increase in beam loading that leads to a poor quality X-ray source is caused by a rise in cathode temperature during the course of the laser pulse. Figure 4.3 shows a measurement of the current out of the electron gun, which depends intimately on temperature as shown in equation 4.1.1. Here $A_G$ is the Richardson constant and $W$ is the work function for LaB$_6$, $T$ is the temperature, and $dW = \sqrt{e^3 E/(4\pi\epsilon_0)}$ depends on $E$, the applied electric field. A 4 $\mu$s pulse with a nearly flat top around 100 mA is ideal, but we see a 30% rise in current during the pulse due to back-heating.

$$I_{gun} = A_G T^2 e^{(W - dW)/(k_B T)}; \tag{4.1.1}$$

Figure 4.3: Measurement of electron gun current showing the increase in current over the course of the 4 $\mu$s macropulse due to back-heating.
The back-heating phenomenon is explained in figure 4.4. The gun cavity is on the left with the cathode inside. High power RF (1 MV) is fed into the cavity and depending on where in the RF cycle the electrons are emitted, they can take a variety of trajectories. Those emitted early experience a maximum acceleration before leaving the cavity. Those emitted later in the cycle experience some acceleration, but before they leave the cavity, the field turns around and partially decelerates the electrons, leading to a smaller final velocity. The skullduggery occurs if the electrons are emitted late in the positive part of the cycle; they experience some acceleration, but then decelerate and actually turn around before leaving the gun cavity, hit the cathode, and heat it (predominately through ionization at the energies available from the electron gun [19]). This is what we refer to as back-heating.
Figure 4.5: Spectral response of the Hamamatsu PV11 photodiode used to measure the temperature of the cathode (data from the manufacturer).

4.2 Measurement of back-heating

4.2.1 Theory

With an understanding of the phenomenon in hand, we set out to measure the temperature rise due to back heating. The cathode is a thermal source and we assume that the spectrum is approximately a blackbody with an emissivity which we determined empirically. With this assumption, we can use a photodiode to measure the light emitted from the source and relate the photodiode current to the temperature of the cathode. The total photodiode current can be written:

$$I_{PD} = K \int_0^\infty d\lambda S_{PD}(\lambda) \int \int dA d\Omega L_\lambda(\lambda)$$ (4.2.1)

where $S_{PD}$ is the spectral response of the photodiode shown in figure 4.5, $L_\lambda$ is the spectra irradiance from the cathode defined in equation 4.2.2, and $K$ is a geometry dependent factor determined empirically [20].

$$L_\lambda(\lambda) = \frac{2hc^2}{\lambda^5} \frac{1}{\exp(hc/(\lambda k_B T)) - 1}$$ (4.2.2)

The relation 4.2.1 supplies us with the relation between temperature and current needed for our measurement.
4.2.2 Implementation

We built a light blocking box and mount for the photodiode pictured in 4.6 and placed it on a window shown in 4.7 connected to the microwave gun pictured in figure 4.8. The cathode is at the center of the copper accelerator cavity and a small mirror reflects light in/out of the port.

The signal from the photodiode is in the tens of nano-amperes range, so must be amplified before transport from the microwave gun (which is located in a radiation shielded vault) to the measuring oscilloscope located in the control room. An amplifier circuit described in appendix A was used for this purpose.

The results of measurement of the photodiode amplifier output voltage and application of the analysis described in appendix A is shown in figure 4.9. This is the first result showing the temperature dependence of a thermionic gun due to back-heating and will be published once initial data on the laser cooling effect is collected.

4.3 Calculation of laser pulse characteristics for cooling

Figure 4.9 shows the cathode temperature increases approximately linearly from 1712K about 30 K over the 4 µs RF macropulse period, then cools slowly as the heat added by back-heating electrons diffuses away (the next macropulse is 1/10th of a second away
Figure 4.7: Laser input and observation port for the microwave electron gun used to feed the laser to the cathode and measure its temperature. A small mirror reflects light from the cathode up through this port.

Figure 4.8: Picture of the microwave electron gun showing the gun accelerator cavity, RF waveguide, and observation port.
so there is ample time for the cathode to return to its steady state temperature of 1712 K before the next pulse).

In order to use laser heat diffusion to cancel this heating, we need to understand how the temperature depends on time after the laser pulse. The laser pulse length of $\sim 5$ ns is much shorter than the time needed for appreciable heat diffusion, as we shall see below. Therefore, we assume a delta function pulse that deposits energy Q onto the cathode and use a 1 dimensional model to determine the temperature versus time relation. The heat conduction equation (4.3.1) must be solved for a quantity of heat Q located a $z=0$ at $t=0$.

$$\frac{\partial T}{\partial t} = D \nabla^2 T$$  \hspace{1cm} (4.3.1)

Here the thermal diffusivity, $D = \frac{K}{\hat{C}}$, $K$ is the thermal conductivity, and $\hat{C}$ is the volumetric heat capacity. Kittel [21] solves equation (4.3.1) for a spatial delta function pulse with energy Q (4.3.2).

$$\Delta T(z,t) = \frac{Q}{A\sqrt{\pi CK}} \frac{1}{\sqrt{t + t_o}} e^{-z^2/(4Dt)}$$  \hspace{1cm} (4.3.2)

Here $A$ is the cathode area making $Q/A$ the spatially averaged laser fluence and $t_o$ is the amount of time before the RF macropulse that the laser heat is deposited on the cathode. The $1/\sqrt{t}$ time dependence of the diffusive cooling makes an exact match to the linear back-heating rise impossible, but the goal of stabilizing the temperature can still be met, as the curves can be substantially matched over the region of interest.
The parameters $Q$ and $t_o$ in equation 4.3.2 can be used to tune the cooling curve to match the back-heating curve. We choose to equate the slopes of the two curves to match 2 $\mu$s into the 4 $\mu$s macropulse. For a given value of $Q$, this determined $t_o$. Next we swept values of $Q$ and looked at the resultant temperature of the cathode.

Figure 4.10 shows the cathode temperature for different values of $Q$. As $Q$ is increased (and subsequently the value of $t_o$ is also increased), the temperature of the cathode during the macropulse is flattened. There is of course a limit to $Q$ that can be applied to the cathode as eventually you will create a laser induced plasma (LIP) from the material on the surface of the cathode and permanently change its shape and emission characteristics. We expect to be well below this limit as most metals explored during our LIBS studies required energies of 50 mJ or more with a 300 $\mu$m beam radius to induce a LIP where we are dealing with less than 15 mJ energies in a beam with a radius on the order of 1.5 mm: a factor of 83 less fluence.

Figure 4.11 shows a comparison of usable pulse length for different laser pulse energies. Since the heat conduction equation is linear the superposition principle applies and we create the temperature curves by simply adding the back-heating curve to the cooling curve and shifting the baseline temperature. Note that changing the baseline temperature is accomplished by simply adjusting the heater for the thermionic cathode. Back-heating curves were extended to longer times by simple extension of the approximately 7.5 K/$\mu$s rise in temperature with macropulse time obtained from measurement. The true temperature should be slightly less than the results obtained in 4.11 as the decrease in temperature of the cathode due to laser cooling will actually decrease the total current output (since thermionic emission is governed by temperature according to equation 4.1.1) and subsequently decrease the back-heating current. McKee shows how the Bethe-Block equations for back-heating can be integrated into the one-dimensional heat equation. However, we know the effect must be less than a 30% decrease in current (due to the maximum rise in current of 30% and the linear relationship between back-heating and total current). Work to numerically solve the one-dimensional heat equation taking into account the aforementioned decrease in back-heating current is pending.

That said, the end result of these heating calculations is that a 15 mJ (or possibly less energy) laser pulse extends the macropulse length to 8.5 $\mu$s, achieving the goal of this line of research with a modest energy pulse.
Figure 4.10: Plot of heating due to laser pulse for different values of Q and $t_o$. As the magnitude of the laser energy Q is increased, the laser pulse must preempt the RF pulse by more time.

Figure 4.11: Plot of temperature for various laser pulse energies. As the pulse energy is increased, the RF pulse can be longer with the same variation in temperature. Note the key shows the laser pulse energy and the ‘usable’ RF macropulse duration for the laser energy. We see in the black dotted curve that a 15 mJ pulse can extend the usable macropulse length out to 8.5 $\mu$s, achieving the goal of this line of research.
4.4 Numerical solution to heating equation

The thermal diffusivity $D$ of the lanthanum hexaboride (LaB$_6$) electron gun cathode was assumed to be constant in 4.3.1 in the previous analysis, however Tanaka showed experimentally that the diffusivity depends on temperature [22]. According to Debye theory and assuming $T >> T_{Debye}$, the diffusivity of a metal should have a $1/T$ dependence [20], however this simple model turns out to not be accurate in the case of LaB$_6$. A best fit of Tanaka’s data shows a $1/T^{0.64}$ temperature dependence and is plotted along with a best fit to a $1/T$ dependence in figure 4.12. This dependence changes the differential equation 4.3.1 and its solutions. We numerically solved the new differential equation 4.4.1 (with $D = \tilde{D}/T^{0.64}$) with a finite difference method described by equation 4.4.2. Here $\tilde{D} = 12.7675 \text{ cm}^2 \text{K}^{0.64}/\text{s}$, $\Delta z = 3\mu\text{m}$, and $\Delta t = 0.02\mu\text{s}$. The initial laser pulse delta function set the temperature of a single spatial bin in the simulation to $T = 1715 + F/(C_v \Delta z)$ with $F$ being the laser fluence, $C_v = \sigma_{\text{thermal}}/D$ the heat capacity, and $\sigma_{\text{thermal}} = 488/T + 0.153 \text{ J/(cm s K)}$ from [22].

$$\frac{\partial T}{\partial t} = \frac{\tilde{D}}{T^{0.64}} \nabla^2 T \quad (4.4.1)$$

$$T(n + 1, j) = \frac{\tilde{D} \Delta t}{\Delta z^2 T(n, j)^{0.64}}(T(n, j + 1) - 2T(n, j) + T(n, j - 1)) + T(n, j); \quad (4.4.2)$$
The numerical solution taking into account the temperature dependence of the diffusivity showed results indistinguishable from those of the first order model in the previous section, confirming the validity of previous section.

### 4.5 Verification of heating spatial uniformity

Thus far, we have assumed a uniform distribution of heat in both the laser pulse and the back-heating. Due to non-idealities in the cavity and cathode and a deflection magnet, there is the possibility of non-uniform heating. The deflection magnet was installed to deflect a portion of the back-heating electrons from hitting the cathode, but only partially eliminates the backheating effect. The deflection of the electrons that exists the gun cavity for use in the rest of the system is compensated for by steering magnets.

To measure the effect of these non-idealities of the back-heating, we setup a vidicon camera with a telescope to record the illuminance of the cathode with different length RF macropulses. A vidicon camera is an older type of camera that is not susceptible to radiation damage like a CCD and can operate in the substantial radiation present near the electron gun. Figure 4.13 shows images obtained with and without back-heating from a 4 $\mu$s macropulse. There is a difference in overall brightness, but it is difficult to tell if there is any non-uniformity. Figure 4.14 shows the rescaled difference between these two files. In the area of the cathode the difference between the image with and without back-heating is roughly constant, indicating that the heat is evenly spread across the cathode surface. There is, however, a slight increase in intensity on the left part of the cathode. This is an expected result of the deflecting magnet near the cathode that is there to deflect some of the back-heating electrons away from the cathode before they can hit and heat it. If the laser cooling concept is successful, the deflecting magnet may no longer be necessary, leading to a more uniform electron beam exiting the gun cavity.

With this data in hand, we are ready to do a crude test of the laser cooling system. As a future refinement, we are in the process of taking measurements with a streak camera that has time resolution capability of 500 fs that will allow us to discern any heating non-uniformities on much finer time scale than is possible with the vidicon camera. If there are such non-uniformities, they can be compensated for by steering of the laser or shaping of its transverse spatial mode.
Figure 4.13: Images of the cathode with no backheating (left) and backheating (right) due to a 4 µs macropulse taken with a vidicon camera with a 17 ms frame time. The intensity of light from the cathode is uniform with and without back-heating.

Figure 4.14: Contour plot of the difference between the two images in figure 4.13. The difference is nearly flat in the area of the cathode showing that the heating is uniform on the time scale of the image acquisition of 17 ms.
4.6 Laser cooling experiment

4.6.1 System design

Due to the radiation present in the FEL test cell from the \( \sim 40 \) MeV electron beam colliding with parts of the beam line during start-up and with the beam dump during tuned operation, the YAG laser needs to be located outside the test cell and the laser must be transported to the cathode over an appreciable distance of 20 meters. We considered two options: an evacuated transport system with mirrors and a fiber optic transport system. The evacuated system has the advantage of being very low loss, but is very expensive, difficult to build, and requires a steering optic at each bend. The fiber system on the other hand, is higher loss, but is about \( 1/20 \)\(^{th}\) the cost and has the added advantage of converting the Gaussian mode output of the YAG laser to a uniform top-hat pulse at the output. Note the fiber is only higher loss due to the input and output coupling to/from the fiber; power transmission through 20 meters of the fiber chosen was better than 96%. The power loss was not a concern since the YAG could deliver 100 times the energy per pulse required according to the calculations in the last section. The biggest disadvantage of the fiber optic system is the possibility of optical damage to the input surface of the fiber due to the fundamental Gaussian spatial profile of the beam at the input which has twice the peak fluence as the output top-hat spatial profile.

We chose the Anhydroguide APCH 1 mm core multi-mode fiber from Thorlabs Inc. for laser transport. The fiber has a damage threshold of 25 MW for a 10 ns pulsed YAG fundamental (\( \lambda = 1064 \) nm) or 250 mJ per pulse, which is more than a factor of 10 greater than what our calculations predict we will need to extend the macropulse duration to 8 \( \mu s\).

The power transmission coefficient at the air/fiber interface was calculated using \( n_{fiber} = 1.534 \) \(^{23}\) and equation \(4.6.1\) from \(^5\) (assuming normal incidence) to be 96% leading to an overall transmission of the fiber (including both interfaces and attenuation) of 88%.

\[
T = 1 - R = 1 - r^2 = 1 - \left( \frac{n_{air} - n_{fiber}}{n_{air} + n_{fiber}} \right)^2 \quad (4.6.1)
\]

The major components of the laser transport system and described in figure \(4.15\). Procurements of these pieces are complete and construction of the transport system is in progress. To date we have tested the damage threshold of the fibers and have been able to
transport 23 mJ pulses through the fiber system without damage for 5 minutes. A bit of a learning curve was encountered as optical damage resulted to the fiber at energies much less than the damage threshold. Our first attempt to put high power through the fiber resulted in damage to the input surface due to the beam being too focused at the input. On our next attempt, a larger transverse area, but still converging beam was fed into the fiber. This resulted in damage within the fiber due to the beam coming to a focus approximately 10 cm within the fiber and causing serious internal damage to the fiber. Our third attempt with collimated beam (Rayleigh range of 20 meters) with a transverse area (made through the use of a telescope (1 converging and one diverging lens)) on the order of the fiber core was successful at carrying 23 mJ pulses. Long term testing of the transport of such a beam is pending.

4.6.2 Timing

In order to correctly time the laser pulse with respect to the electron macropulse and be able to vary its power simultaneously, we determined that it will be necessary to use two delay generators and a number of signals available from the FEL system. A timing diagram is presented in figure 4.16. The command charge signal from the FEL signals the charging of capacitors that will provide charge for the klystron that creates the high power RF used to accelerate electrons in both the gun cavity and the LINAC. There is then a delay as these capacitors charge before a thyratron (high power switch) is closed and charge flows to the pulse forming network (PFN). The PFN filters the pulse flowing to the klystron to make a smooth high voltage signal enabling the klystron to create stable, high power RF. This PFN pulse is 10 $\mu$s long and the operator can chose during what part of the pulse to feed an input signal to be amplified into the klystron by adjusting the width and position of the RF on pulse. The flashlamps of the YAG laser are triggered by a fixed delay 500 $\mu$s before the PFN trigger so that high power is built up in the YAG cavity in anticipation of firing of laser nearly coincident with the RF pulse that drives the electron beam. One delay generator (Berkeley Nucleonics 505-2C) will adjust the timing of the YAG Q-switch gate and RF on pulse (which have a fixed delay of 1 $\mu$s between them) relative to the PFN trigger (labeled variable 1 in the figure). Variable 1 controls the amount of time between the YAG flashlamps firing and the Q-switch which in turn controls the laser pulse energy and is a coarse adjustment on the order of tens of microseconds. The other delay generator will control the actual YAG Q-switch relative to the Q-switch gate. This Q-switch delay
Figure 4.15: Schematic of the laser cooling transport system. The YAG laser output is P-polarized and polarizer P1 is used for attenuation. Beamsplitter BS1 picks off part of the beam for power monitoring at detector D1. Lenses L1 and L2 focus and collimate the beam while the high power iris HP11 prevents any stray light from damaging the fiber cable holder. The fiber cable FC1 transports the beam to the radiation vault. Lens L3 recollimates the beam and lens L4 focuses it onto the cathode. Polarizer P2 polarizes the beam so that any stray reflections can be blocked by polarizer P3 before they enter the vidicon or streak camera. Beam splitter BS2 picks off part of the beam for power monitoring at detector D2. Finally, mirror M1 steers the beam onto the cathode through the window on the RF gun cavity.
Figure 4.16: Timing diagram of signals for laser cooling. See the text of this section for details.

is a fine time adjustment, on the order of one microsecond, that allows precise placement of the laser pulse relative to the RF pulse and subsequently the electron emission from the cathode that was shown to be necessary in figure 4.10.

### 4.7 Conclusion

With the back-heating data collected to date, we have shown that extension of the electron beam macropulse length by a factor of two out to an 8 µs pulse is possible taking into account all applicable phenomenon. The back-heating data collected shows a relatively uniform heating that can be appreciably compensated for by the cooling effect of
a relatively low power laser pulse. As part of a post-doctoral appointment at UH, we will be building and testing the laser cooling system in 2012.
Chapter 5

Analysis of CuIn$_{0.7}$Ga$_{0.3}$Se$_2$ solar cells with nanosecond pulse length laser induced breakdown spectroscopy (LIBS)

In this work, we show that laser induced breakdown spectroscopy (LIBS) with a nanosecond-pulse laser can be used to measure the constituents of CuIn$_{0.7}$Ga$_{0.3}$Se$_2$ thin film solar cells. This method has four significant advantages over methods currently being employed: the method is inexpensive, measurements can be taken in times on the order of one second, without high vacuum, and at distances up to 5 meters or more. The final two points allow for in-line monitoring of device fabrication in laboratory or industrial environments. Specifically, we report a reliable linear relationship between the copper and sodium spectral lines from LIBS and the atomic fraction of copper and sodium measured via secondary ion mass spectroscopy (SIMS), discuss the ablation process of this material with a nanosecond-pulse laser compared to shorter pulse duration lasers, and compare nanosecond-pulse LIBS and SIMS as depth profiling methods. Concurrently, we observed two opportunities where the FEL could contribute in this field of work: (1) the picosecond pulses cause a different ablation mechanism than nanosecond pulses and (2) a broad sweep of wavelength and its effect on LIBS has never been done and could yield detailed information on the plasma properties and its formation.
LIBS has long been used as a method for detection of trace elements [24] and has more recently been shown to be an effective method for measuring atomic fraction of constituents with depth [25, 26]. While these studies have looked at layers of one or two elements, to date few studies have been done on a thin film as complex as CuIn$_{0.7}$Ga$_{0.3}$Se$_2$ (CIGS) solar cells. We have shown previously that a reliable linear relationship between the sodium peak from LIBS and the mass of sodium deposited during deposition exists [27]; in this study, we build on that work and set out to use LIBS to measure the atomic fraction of copper and sodium in CIGS solar cells via calibration with secondary ion mass spectroscopy (SIMS) analysis. We specifically focused on sodium, the control of which in CIGS solar cells is critical to achieve high efficiency devices [28], but to date composition measurement techniques either cannot accurately quantify the sub-one atomic percent levels (for example, x-ray fluorescence (XRF) and energy dispersive X-ray spectroscopy (EDX)) or are expensive, time consuming, require high vacuum, and cannot be done at large distances (examples include EDX, SIMS, and x-ray photo-electron spectroscopy (XPS)). LIBS is an inexpensive and fast method that can be done at a large range of sample to instrument distances (from centimeters to meters) without the need for vacuum and is able to reliably detect and quantify sodium and copper atomic percentages.

5.1 Theory

5.1.1 LIBS spectra theory

LIBS employs a high power laser to create a plasma from the material to be investigated [29]. As the plasma cools and ions and electrons recombine, photons characteristic of the transition energies of the elements present are emitted, and these spectral lines are then matched to those of known lines of specific elements. In the most simple cases, the number of measured counts of an atom’s spectral line is proportional to the atomic percent as has been shown, for example, for chromium in steels [30]. This technique damages an area of the target on the order of the spot size of the laser (600µm in this study).

In general, the nature of the matrix of other materials in which a particular element resides will influence the ratio of the atomic percent of that element to the intensity of its corresponding lines (the so called 'matrix effect') [31]. Though it has been shown that in some cases this ratio can be constant over broad compositional ranges [32], in this study, the concentration range of interest of all constituents is narrow, so we assume and confirmed in
our results a constant linear relationship between the relative intensity of the atomic lines of a particular element and its atomic concentration.

5.1.2 LIBS plasma formation and ablation

In semiconductors like CIGS there are few conduction band electrons and the initial laser radiation for LIBS leads to either single or multi-photon band to band transitions instead of direct heating of the material [29]. After generation of conduction band electrons, the lattice heats due to coupling of the generated electron-hole pairs to the lattice. The photon absorption cross section is roughly 17 orders of magnitude greater for photons with energy above the band gap energy compared to below [33], leading to a higher density of deposited energy with photon energy above the band gap. The band gap of the CIGS materials created at the Hawai‘i Natural Energy Institute and used for this study has been measured to be \( \sim 1.2 \) eV, corroborated by the significant photon to electron-hole pair conversion evident from the quantum efficiency (QE) curves in figure 5.1 at wavelengths shorter than 1100 nm (energy greater than 1.13 eV). The laser wavelength used for LIBS in this study was 1064 nm (photon energy of 1.17 eV), enabling laser energy to be densely coupled into the CIGS material.

If the pulse duration is short compared to the lattice ion relaxation time (on the order of 100 fs to 1 ps), the initially excited electron-hole pairs do not have time to couple to the lattice before all of the pulse energy is deposited [34]. Since the interaction cross section of photons with the lattice is small compared to that of photons to the electron-hole excitation, this leads to a dense deposition of energy in the material that is typically greater than its heat vaporization, resulting in sublimation of the material, no mixing of the material at different depths, and an abrupt edge to the ablation crater.

Conversely, if the laser pulse duration is longer than the picosecond time scale, significant energy is coupled into the lattice phonon modes and diffuses into the material before the threshold energy density for vaporization can be reached. When this occurs, the material is first melted, then the melted material is vaporized. This sequence results in mixing of material at different depths and a rough edge to the ablation crater [34]. The nanosecond pulses used in this work are much longer than the lattice ion relaxation time and result in a rough ablation edge as will be discussed in the results section.
5.2 Experiment

5.2.1 CIGS PV cell fabrication

For this work a total of 18 PV cells were fabricated on 3 substrates in the following vertical stack: a 1”x1” thin titanium substrate, 1 µm of molybdenum deposited via magnetron sputtering, 10 mg of NaF deposited via evaporation, 1.8 µm of CIGS deposited via a 3-stage evaporation process [35], 100 nm of CdS n-type buffer layer deposited via chemical bath deposition, 80 nm of ZnO, 100 nm of indium tin oxide (ITO), and Ni/Ag grids for charge collection. The complete cells are pictured on the left in figure 5.2. During the CIGS deposition, the evaporation sources for copper and sodium were intentionally positioned to provide a significant gradient of evaporant across the substrates to be analyzed. This fabrication process resulted in functional solar cells with modest performance.

5.2.2 LIBS setup

Figure 5.3 shows a schematic of the LIBS measurement setup. The samples were interrogated in air with a 1064 nm Nd:YAG Continuum Surelite II laser at 100 mJ per pulse with a varying integration time between 1 and 3 seconds. This relatively high laser pulse energy was chosen to yield a signal to background ratio of 10:1 for all measurements and to create as uniform an ablation crater as possible (as higher power leads to more uniform ablation with nanosecond pulses [26]). In the far field, the laser spot diameter was 600 µm. A near-infrared (NIR) dichroic beam-splitter was used to assure collinearity between the laser and telescope optical axes. The resultant radiation was fiber coupled to an Ocean Optics LIBS 2500+ Spectrometer with a bandwidth of 500 nm to 900 nm. The results were digitally recorded. The current experiment is designed for detection at a distance of 5.2 m from the samples to emulate an in-line device fabrication setup.

An optically transparent fused silica window with a 1064 nm transmission of greater than 90 percent was used to allow transmission of the laser and resulting breakdown radiation. For purposes of system detection calibration, a calibrated tungsten filament thermal source was used to determine the system efficiency as a function of wavelength.

Five spots on each device were analyzed using LIBS (see figure 5.2) and the remaining area of the device was analyzed with SIMS. Twenty five 100 mJ pulses were used to collect spectra down to ~ 3.4µm (measured with a Tencor Alpha-Step profilometer) from the top of each device. The background in the LIBS spectra (due to Bremsstrahlung...
Figure 5.1: Typical quantum efficiency curve for the devices used in this study. This curves show the conversion efficiencies of photons at a particular wavelength to photocurrent production. Note the significant quantum efficiency for photon wavelengths less than 1100 nm (energy greater than 1.13 eV) which confirms that the band gap is below the YAG laser photon energy of 1.17 eV.

Figure 5.2: Image of one substrate with 6 devices (labeled d1 through d6) before LIBS (left) and after LIBS (right). Note the five craters formed from the five measurements taken on device 3 (d3). Five such devices spanning 3 substrates were used for this study.
Figure 5.3: Schematic of the LIBS measurement setup. Laser travels through the beam-splitter, is focused on the sample by a lens, and causes plasma formation of the sample. The sample emits photons which travel down to the beam splitter and are reflected into the telescope for collection by the spectrometer. A computer controls the spectrometer integration window and laser pulse timing.
radiation, stray light, and detector dark counts) was removed to improve the signal to noise ratio.

5.3 Results and Discussion

5.3.1 Analysis of LIBS craters

Profiles of the LIBS craters are presented in figure 5.4. These show a relatively flat floor to the craters in some cases, but a rough floor in others. The roughness is due to the long (nanosecond) pulses leading to melting, evaporation, and ultimate resolidification of the unevaporated material under the influence of chaotic forces from the plasma into irregular shapes. We attempted to use a higher energy pulse to keep the profile flatter and thereby have more confidence that a particular pulse corresponds to a particular depth in the material [26], but were only partially successful. The uneven ablation for each pulse leaves the utility of nanosecond LIBS in questions for fine depth analysis, but has little effect on the bulk analysis done here since the variations in the crater floors are small compared to the overall depth analyzed.
5.3.2 LIBS peak identification

In order to correlate the spectra obtained from LIBS to the presence of particular atoms, strong lines without overlap to other lines in the material that were within the bandwidth of the spectrometer were chosen from the NIST Atomic Spectra Database as shown in table 5.1. Note that with a spectrometer sensitive to the appropriate spectral range, strong lines for indium, gallium, selenium, and tin could be recorded allowing LIBS analysis of all constituents of the device.

Table 5.1: Strong spectral lines and their corresponding elements used for LIBS analysis.

<table>
<thead>
<tr>
<th>wavelength (nm)</th>
<th>element</th>
</tr>
</thead>
<tbody>
<tr>
<td>510.55</td>
<td>Cu</td>
</tr>
<tr>
<td>550.65</td>
<td>Mo</td>
</tr>
<tr>
<td>589.00</td>
<td>Na</td>
</tr>
<tr>
<td>625.81</td>
<td>Ti</td>
</tr>
<tr>
<td>636.23</td>
<td>Zn</td>
</tr>
<tr>
<td>643.85</td>
<td>Cd</td>
</tr>
</tbody>
</table>

5.3.3 Copper and Sodium calibration curves

As can be seen from figures 5.4 and 5.5 multiple pulses were required to obtain spectra for the entire device and therefore to use LIBS to measure its contents. However, due to saturation of the spectrometer, multiple spectra needed to be taken at each spot. In order to obtain an accurate calibration of the LIBS spectra to the atomic percent, all of the spectra originating from the ablated material (3.4 $\mu$m of device material from the 25 pulses) in a particular spot were added together and the sum was normalized to the total number of spectrometer counts. This procedure yields a normalized spectrum representative of the bulk of the device for a particular spot. LIBS signals taken from the 5 spots on each of 5 devices (see figure 5.2) were averaged and the standard deviation was calculated. SIMS analysis was done on each of the five devices and the atomic percent was averaged over the same 3.4 $\mu$m of ablated material as the LIBS analysis. The SIMS bulk average atomic percent (y coordinate) and LIBS bulk average peak height (x coordinate) data pairs for the copper and sodium are plotted along with a linear least squares fit in figures 5.6 and 5.7. The fits for both copper and sodium are satisfactory with values for $R^2 = 0.872$ and $R^2 = 0.935$, respectively, showing that indeed that LIBS produces a reliable signal proportional to the atomic fraction of copper and sodium in this concentration regime.
Figure 5.5: LIBS spectra for each pulse for the most copper rich device. One can see the intensity of copper lines at 510.55 nm, 515.32 nm, and 521.82 nm decrease to background levels approximately at pulse 18. Molybdenum peaks near 550.65 nm decrease just as the titanium peak at 625.81 nm appears. The sodium lines at 589.60 nm and 589.59 nm are roughly constant throughout the depth of the cells, save for the first 2 pulses which contain little sodium. Zinc and cadmium peaks only appear in the first three pulses, consistent with the device structure.
Figure 5.6: Linear fit of atomic fraction of copper from SIMS analysis vs. normalized photon counts from LIBS at 510.55 nm. Five LIBS measurements were taken and averaged for each data point. The error bars represent plus and minus one standard deviation. Note that the atomic percentage of copper in the CIGS layer is typically 25%, but here we have taken an average over the entire device through the molybdenum layer making the values on the y-axis lower.

\[ y(x) = 46.327x + 0.527, \quad R^2 = 0.872 \]

Figure 5.7: Linear fit of atomic fraction of sodium from SIMS analysis vs. normalized spectrometer counts from LIBS at 589.00 nm. Five LIBS measurements were taken and averaged for each data point. The error bars represent plus and minus one standard deviation.

\[ y(x) = 329.458x - 57.286, \quad R^2 = 0.935 \]
Figure 5.8: SIMS analysis of the median sodium and copper content device. One can clearly see the different device layers from the signal of their constituent elements. The ZnO is evident from the zinc signal near the surface. Below, the cadmium from the CdS layer is visible. Next the CIGS layer is indicated by the presence of copper. Finally the molybdenum and titanium substrates are indicated by increased signal for those elements.

5.3.4 Depth profile comparison between LIBS and SIMS

Recently there have been a number of studies [25, 26, 34, 37, 38] of LIBS as a depth profiling method. They have found that generally picosecond or femtosecond pulses are required to achieve sublimation and uniform ablation instead of the melting/evaporation and non-uniform ablation seen with the nanosecond pulses used in this study. Due to the mixing of layers with depth during melting and the subsequently non-uniform surface created for the next ablating pulse, the abrupt interfaces between layers seen in SIMS analysis in figure 5.8 are not observed in the corresponding LIBS analysis in figure 5.9.

However, the LIBS depth analysis does have the expected qualitative features. Spectra from only the first three pulses contained zinc and cadmium peaks, as expected from ZnO and CdS layers in the top 300 nm of the device. The copper peak slowly decreases until the entire CIGS layer has been ablated by pulse 25. Finally, the molybdenum and sodium peaks both decrease as the titanium signal is increasing at pulse 25 where SIMS analysis indicated the molybdenum/titanium interface to be.

There are some interesting features of the LIBS depth analysis in figure 5.9. The copper peak is greatest in the first couple of pulses and decreases linearly. This is most likely due to the ratio of CIGS to molybdenum in the melted material decreasing with
depth (corroborated by the increasing molybdenum signal in this range). Additionally, the molybdenum peak appears surprisingly early in the laser pulses (around pulse 3). This can be explained by the deep probing of the first few laser pulses around the edge of the crater evident in figure 5.4a. From this phenomenon, we theorize that the entire CIGS layer is melted during the laser pulse and resolidifies between pulses. This makes LIBS with nanosecond pulses impractical for analysis of all the constituents of CIGS materials as at the deposition temperature for the devices of 600 °C selenium evaporation is significant enough to require a constant vapor pressure of selenium in the deposition chamber in order to maintain stoichiometry. With the melting temperature of the copper, 1085 °C, a liquefied pool of CIGS will evaporate significant selenium, changing the composition of layers of the material below those being currently measured, and leading to a seemingly selenium poor device in one that is actually stoichiometrically correct.

5.4 LIBS and the FEL at UH

The knowledge gained as to the mechanism of ablation and the poor depth resolution of nanosecond-pulse LIBS point toward the use of picosecond or shorter laser pulse for fine depth resolution. The FEL at UH produces a train of 1 ps pulses with 350 ps between the pulses with a macropulse duration of 4 µs, resulting in a ~ 250 mJ macropulse energy at present time. Each picosecond pulse has 22 µJ of energy which can be focused to a spot ra-
radius of $r = 0.61\lambda/(n \sin \theta) \approx \lambda/2 = 1.5\mu m$ \[4\] assuming a numerical aperture, $n \sin \theta = 1.4$ (a reasonable value that assumes an index of refraction of the optic of 1.4 (typical of many glass lenses)). We’ve seen in this work that a fluence of $F_{YAG} = 100mJ/((\pi(600\mu m/2)^2) = 354mJ/mm^2$ gives a strong LIBS signal in these CIGS solar cells. The FEL is capable of producing a fluence of $F_{FEL} = 0.022mJ/((\pi(1.5\mu m)^2) = 3095mJ/mm^2$, which is more than sufficient to LIBS the CIGS solar cells studied here (ignoring any wavelength dependent effects).

With its fine frequency tunability and sufficient fluence (see calculation in this section) to create a laser induced plasma (LIP) the FEL has the potential to elucidate plasma formation in specific material and ultimately provide better characterization information. As a next step, we would like to use the FEL to perform a number of unprecedented experiments: depth profiling experiments on thin film solar cells and thin film photoelectrochemical devices and a wavelength sweep of LIBS results with the FEL spanning the 1 $\mu m$ to 10 $\mu m$ paying special attention to how the physical ablation and spectrum change near the bandgap wavelength for these materials.

5.5 Conclusion

We have shown that a bulk compositional analysis of the copper and sodium content in a complete CIGS solar cell is possible with LIBS; reliable curves were obtained showing that the atomic percentage is linearly proportional to the peak heights of the corresponding element. Additionally, while we have shown that depth analysis with nanosecond LIBS is not a viable method for precise depth profiling of this material, it can give qualitatively correct results with depth, and picosecond or femtosecond LIBS could produce results with depth on par with SIMS. The viability of LIBS for bulk analysis of CIGS solar cells and other thin film technology demonstrated here can lead to significant cost and time savings when compared to methods currently in use.
Chapter 6

Conclusion

To date significant progress has been made toward improving the possible average power, spectral purity, and pulse length of the FEL. We have designed, built, bench tested, and extensively simulated the TIRBBE which allows higher average power in the FEL resonator by avoiding mirror damage. Our simulations have shown that the dispersion of the TIRBBE also dramatically decreases the optical spiking which plagues the FEL. We have designed, built, bench tested, and extensively simulated the FROZEN FISH etalon filter and shown that it can eliminate the frequency pulling in the FEL and suppress sidebands. Through our bench testing, we have verified the FROZEN FISH design is sound in terms of its ability to attain the needed precision in both parallelism and gap control required for it to operate over the large range of wavelengths which the FEL is capable of producing. To enable longer pulse lengths in the FEL, we have made a first measurement of the backheating phenomenon, calculated the extension of electron beam (and subsequently laser beam) pulse lengths attainable with laser cooling using modest energy cooling pulses, explored the spatial distribution of backheating, and designed and began construction of the laser cooling system. Finally, we have shown that LIBS can be used to for bulk measurements of the constituents of photovoltaic materials. This work on LIBS has pointed out the shortcomings of a typical LIBS system and shown us opportunities where an FEL could produce highly desirable depth information and a first measurement of the effect of a frequency on the LIBS process across a vast frequency range.

As a post-doc, the author plans on completing tests of the TIRBBE and FROZEN FISH in the FEL cavity once the laser power can be increased to a level where the effect of these optics is pronounced. The laser cooling system will be constructed and first temperture
data should be available within the next year. A test of the FEL for LIBS will be completed as soon as funds become available for the required spectrometer.
Appendix A

System for measurement of back-heating of electron cathodes

The transimpedance amplifier circuit pictured in figure A.1 and represented schematically in A.2 was used to amplify the signal coming from the vacuum photodiode [39]. The operation amplifier (op-amp) U1 is setup for a transimpedance gain of one million with the 1 MΩ resistor R1 and connection to the photodiode (modeled by source I1) resulting in a signal of tens of millivolts at the measuring oscilloscope as described by equation A.0.1.

\[ v_{\text{scope}} = -1 \times (-I_1 \times R_1 + V_{U3, \text{wiper}}) \]  \hspace{1cm} (A.0.1)

The amplifier was powered by two 9 volt batteries V1 and V2 to eliminate any power supply noise and capacitors C4 and C5 were added to prevent any power rail oscillations. The zero level of the amplifier was adjusted with a potentiometer U3 to maximize its dynamic range. A 50 MΩ current limiting resistor R5 and an energy storage capacitor C3 were added to protect the photodiode from high current yet allow for fast switching. Capacitors C1 and C2 model parasitics of the photodiode and resistor, respectively. Finally, a buffer stage with op-amp U2 was added to drive the 130 feet of 50 Ω coaxial cable (modeled by T1 and capacitor C7) and oscilloscope (modeled by resistor R4 and capacitor C6). The installed device is pictured in figure A.3.

The bandwidth of the Linear Technologies LT1632 op-amp of 45 MHz with 45 V/ms slew rate was more than sufficient to detect the back-heating phenomenon which we assume causes a linear rise in voltage of the op-amp output of 40 mV during the 4 µs RF pulse (10 V/ms) as validated by experiment in the next section.
Figure A.1: Amplifier circuit for the vacuum photodiode.

Figure A.2: Schematic of amplifier circuit and model for transmission line and oscilloscope. See text for details.
Figure A.3: Photodiode and amplifier circuit installed in system.
A.0.1 Data and Analysis

To convert the measured amplifier circuit to a voltage the following procedure was used:

1. Generate photodiode current temperature pairs \((I_{PD}, T)\) using equation A.0.2 spanning a range of temperature values around those expected.

\[
I_{PD} = K \int_{0}^{\infty} d\lambda S_{PD}(\lambda) \int dA d\Omega L_{\lambda}(\lambda)
\]  

(A.0.2)

2. Fit an eighth order polynomial to the \((I_{PD}, T)\) pairs to get \(POLY_8(I)\). An eighth order polynomial was chosen so that there was a less than 1% asymptotic standard error in all fit variables to the data. There will be a simple proportionality constant \(K\) (the geometry factor in A.0.2) and offset \(I_o\) (due to the offset voltage of the transimpedance amplifier \(V_U3\text{wiper}\)) between this curve and the \(T\) versus photodiode amplifier voltage \((v_{scope})\) curve, i.e. \(T(v_{scope}) = K \ast POLY_6(v_{scope}/R1 - I_o)\).

3. Find \(K\) and \(I_o\) by increasing the heater voltage on the cathode, measuring the temperature with an optical pyrometer, and then measuring the output voltage of the amplifier and fitting these \((v_{scope}, T)\) pairs to \(T(v_{scope}) = K \ast POLY_6(v_{scope}/R1 - I_o)\), yielding temperature as a function of the photodiode amplifier output voltage at the oscilloscope. The value of \(K\) scaled the temperature values that the fit \(T(v_{scope})\) is valid for. This restricted the values of oscilloscope voltages that could be used to get a valid temperature. We checked that all of our sampled oscilloscope voltages fell in the valid range and when they did not (of course), expanded the range of \((I_{PD}, T)\) pairs generated in item 1 above. This process was iterated until the generated curve was valid for all collected data.

4. Use \(T(v_{scope})\) to convert the measured output voltage curves obtained from the oscilloscope to temperature.

The results of measurement of the photodiode amplifier output voltage and application of the above analysis is shown in figure A.4. This is the first result showing the temperature dependence of a thermionic gun due to back-heating and will be published once initial data on the laser cooling effect is collected.
Figure A.4: Plot of cathode temperature as a function of time during a 4µs macropulse. Note the difference in rise time due to backheating and fall time due to diffusion of heat.
Appendix B

Generation of Bessel Beams using a 4-f Spatial Filtering System

This work appears in the American Journal of Physics, Volume 77, Issue 3, pages 229 to 236 in 2009 and was co-authored by Jeremy M. D. Kowalczyk, Stephanie N. Smith, and Eric B. Szarmes all from the University of Hawaii at Manoa.

B.1 Abstract

We demonstrate a simple and straightforward method of producing Bessel beams using a 4-f spatial filtering system requiring no specialized optical components. The experiment employs the established technique of diffraction from a thin ring source, but the ring source itself is produced by the high-pass filtering of a uniformly illuminated circular aperture, yielding Bessel beams with a central spot radius of less than 35 $\mu$m that persist over a distance of 160 mm. The experiment unifies diffraction theory, Fourier optics, and the physical properties of Bessel beams in a single demonstration appropriate for an advanced undergraduate laboratory.

B.2 Introduction

Bessel beams [40] constitute the earliest example of “nondiffracting” optical waves exhibiting a narrow and persistent beam-like structure, and generated considerable interest when they were first demonstrated.[41] They have evolved over the years from items of curiosity to objects of considerable utility in research applications, and new methods of generating them continue to be explored.
The $m$th-order Bessel beam is a cylindrical optical beam of the form

$$E(r, \phi, z) = J_m(k_T r) e^{im\phi} e^{-i\beta z}$$

(B.2.1)

representing a nondiffracting wave $A(r, \phi)e^{-i\beta z}$ whose complex amplitude $A(r, \phi)$ is independent of $z$. While $E(r, \phi, z)$ is an exact solution to the Helmholtz equation $\nabla^2 E + k^2 E = 0$ in free space, the amplitude $A(r, \phi)$ satisfies the reduced equation $\nabla^2_T A + k^2_T A = 0$ where $\nabla^2_T$ is the transverse Laplacian and $k^2_T = k^2 - \beta^2$. The parameter $k_T$ is identified as a transverse $k$-vector and determines the width of the central lobe of the corresponding Bessel function.

It is the appearance of this narrow central lobe of the Bessel function that qualifies the wave to be described as a ‘beam’. However, it is also readily verified that the total beam power and the RMS beam width are infinite. Therefore, the Bessel beam falls into the same class of nonphysical approximations to real waves as the plane wave. The relationship between the Bessel beams and the plane waves was recognized by Durnin,[40] who showed that the zero$^\text{th}$ order Bessel beam is equivalent to a uniform superposition of plane waves whose $k$-vectors lie on the surface of a cone. This equivalence can be established by direct calculation of the angular spectrum of plane waves for the beam, i.e. the 2-dimensional spatial Fourier components $\tilde{A}(k_x, k_y)$ corresponding to the complex amplitudes of plane waves propagating at angles $(\vartheta_x, \vartheta_y) = (k_x/k, k_y/k)$ to the optical axis.[5] A calculation in cylindrical coordinates shows that $\tilde{A}(\kappa, \varphi) = \frac{2\pi}{k_T} \delta(\kappa - k_T)$, indicating that the spectrum of plane waves is distributed uniformly in $\varphi$, with $k$-vectors constrained to the value $k_T$ in the transverse plane corresponding to a fixed angle of $\vartheta = k_T/k$ in space.

This equivalence motivated the original technique of generating Bessel beams by collimating the waves emitted from a thin circular slit or ‘ring source’. If a thin circular ring of radius $a$ is uniformly illuminated, and a lens of focal length $f$ is placed at distance $f$ from the ring, then the spherical wave emanating from each ‘point source’ on the ring will be collimated into a plane wave propagating at angle $\vartheta = a/f$ to the optical axis, and a Bessel beam will form in the region behind the lens.

Many methods of generating Bessel beams exist, the simplest of which may be the annular slit described above. Holograms[43] and conical prisms (‘axicons’) have been used to produce high power Bessel beams for research applications, and axicons have been used to produce high-order Bessel beams.[45] The direct generation of Bessel beams using specially designed optical resonators has also been demonstrated.[46] From a pedagogic standpoint, McQueen et al[47] describe several experiments to generate Bessel beams using
Figure B.1: Superposition of wave fronts emitted from a ring source of radius $a$ yielding a Bessel beam on-axis. The beam radius increases with distance as the transverse $k$-vector $k_T$ decreases. For the doubled-lobed ring source in our experiments, the propagation delay $\Delta$ between the (+) and (−) lobes varies from $0.63\lambda$ to $0.24\lambda$ along the axial region where Bessel beam forms.

simple optical components, and show how a Bessel beam can recover its form after obstruction by an obstacle. Further examples of Bessel beams in the undergraduate laboratory are given by Basano and Ottonello, [48] including the use of thermal light in place of the more common laser. Rousseau et al.[49] describe a method to produce and characterize both diffracting and nondiffracting optical beams using a 4-$f$ system to spatially filter an Airy pattern.

Our method of generating Bessel beams also employs a 4-$f$ spatial filtering system, but our technique and its motivation both differ from those of Rousseau et al. We employ the established technique of diffraction from a thin ring source, but the ring source itself is produced by the high-pass spatial filtering of a uniformly illuminated circular aperture. In this manner we avoid the special construction of a narrow annular slit, and generate Bessel beams without the use of specialized optical components.

B.3 Theory

B.3.1 Direct generation of Bessel beams from a ring source

The generation of the lowest order (azimuthally symmetric) Bessel beam from a uniform ring source of radius $a$ can be obtained directly from the Fresnel diffraction integral in cylindrical coordinates, and bears a closer correspondence to the actual experiments
described here. For the diffraction of a complex field \( u_i(x', y') \) at \( z = 0 \) to a field \( u_d(x, y; z) \) at distance \( z \), the diffraction formula in rectangular coordinates,

\[
u_d(x, y; z) = i \lambda z e^{-ikz} \iint dx' dy' u_i(x', y') \times \exp\left[-i \frac{\pi}{\lambda z} ((x - x')^2 + (y - y')^2)\right], \tag{B.3.1}
\]
is converted into the symmetric Hankel transform\[8\]

\[
u_d(r; z) = i \lambda z e^{-ikz} e^{-i\frac{\pi r^2}{\lambda z}} 2\pi \times \int r' dr' u_i(r') e^{-i\frac{\pi r'^2}{\lambda z}} J_0\left(\frac{2\pi rr'}{\lambda z}\right), \tag{B.3.2}
\]
by substituting the coordinate transformation \( x = r \cos \phi, \ y = r \sin \phi, \ x' = r' \cos \phi', \ y' = r' \sin \phi' \), inserting the Bessel identity\[50\] \( e^{i\alpha \cos \psi} = \sum_{m=-\infty}^{\infty} i^m J_m(\alpha) e^{im\psi} \), and imposing azimuthal symmetry in the integration over \( \phi' \). This equation is valid in the Fresnel approximation for any azimuthally symmetric field \( u_i(r') \). Thus, if the input field is a ring source of radius \( a \) represented by a radial delta function \( u_i(r') = A \delta(r' - a) \), then the diffracted field is proportional to the zeroth-order Bessel function,

\[
u_d(r; z) = 2\pi i \lambda z e^{-ikz} e^{-i\frac{\pi a^2}{\lambda z}} aA J_0\left(\frac{2\pi ar}{\lambda z}\right). \tag{B.3.3}
\]

Strictly speaking, the Bessel beam proper is a nondiffracting beam with planar wavefronts and a constant spot size that remains undiminished in amplitude as it propagates, whereas the above beam falls off in amplitude as \( 1/z \) and also exhibits a \( z \)-dependent spot size and spherical wavefronts. However, the transverse amplitude profile displays the characteristic Bessel function \( J_0(k_T r) \), where \( k_T = k \sin \vartheta \approx k(a/z) \) is the transverse component of the \( k \)-vectors that propagate in a cone of half-angle \( \vartheta \) to form the Bessel beam. In the above beam this half-angle \( \vartheta \) varies with distance, as depicted in Fig. B.1, which yields a central lobe whose radius increases with \( z \). In contrast, the Bessel beam proper is composed of ideal plane waves that maintain a fixed \( \vartheta \) and a constant spot size as they propagate. In this context, it is interesting to note that strictly planar wavefronts are evidently not required to yield a beam amplitude whose radial dependence is confined to the argument of a Bessel function. The above beam can be converted to a standard Bessel beam with planar wavefronts and constant amplitude by collimating the diffracting waves with a thin lens of focal length \( f \) placed at distance \( z = f \) from the source plane, as in the original demonstration by Durnin et al.\[40\] If the complex transmittance \[5\] of the lens is written
\[ t_l(r) = \exp(+ikr^2/2f), \] then the transmitted field \( u_B(r) \) immediately after the lens is

\[
u_B(r) = u_d(r; f) t_l(r)
= \frac{2\pi i}{\lambda f} e^{-ikf} e^{-i\pi a^2/\lambda f} a A J_0 \left( \frac{2\pi ar}{\lambda f} \right), \tag{B.3.4}\]

which is precisely the amplitude of a nondiffracting Bessel beam of uniform phase and constant intensity whose plane wave components cross at a constant half-angle of \( \vartheta = a/f \). The experiments reported in this paper did not include a collimating lens to produce nondiffracting Bessel beams; instead, the \( z \)-dependence of the radial profile of the diffracted Bessel beam \( u_d(r; z) \) in Eq. [B.3.3] was measured directly.

### B.3.2 Generation of a ring source using a 4-\( f \) spatial filtering system

The ring source generated using the 4-\( f \) spatial filtering system was not a perfect radial delta function, and possessed both a finite width and a complex radial substructure. The theoretical form of this ring source can be calculated analytically from diffraction theory. The 4-\( f \) system is a two-lens imaging system\[5\] that performs a two-stage operation of successive spatial Fourier transforms, each of which transforms an input field \( u_1(x', y') \) in the input focal plane to an output field \( u_2(x, y) \) in the output focal plane according to the prescription \[5\]

\[
u_2(x, y) = \frac{i}{\lambda f} \exp^{-i\pi f/\lambda} \tilde{u}_1 \left( \frac{x}{\lambda f}, \frac{y}{\lambda f} \right), \tag{B.3.5}\]

where \( \tilde{u}_1(\nu_x, \nu_y) \) is the spatial Fourier transform of \( u_1(x', y') \) and is evaluated at the (cyclical) spatial frequencies \( \nu_x = x/\lambda f, \nu_y = y/\lambda f \). For an azimuthally symmetric field this transformation can be written in cylindrical coordinates, after employing the same manipulations leading to Eq. [B.3.2], in the form

\[
u_2(r) = \frac{i}{\lambda f} \int_0^\infty 2\pi r'dr' u_1(r') J_0 \left( \frac{2\pi rr'}{\lambda f} \right). \tag{B.3.6}\]

In the absence of spatial filtering, the 4-\( f \) system yields an output field which is simply a (phase-shifted) replica of the input field. The great utility of the 4-\( f \) system is the ability to spatially filter the image by manipulating the diffraction pattern appearing between the two stages; in our experiments, this filter was designed to transmit only those spatial frequencies that ‘composed’ the sharp edge of the uniformly illuminated aperture in the input plane.

Let us denote the input focal plane of the first stage as the object plane \( (x'', y'') \), the output focal plane of the first stage (serving as the input focal plane of the second stage) as the
Fourier plane \((x',y')\), and the output focal plane of the second stage as the image plane \((x,y)\). In our experiment, the object field \(u_o(r'')\) in the object plane is a uniform circular distribution of unity amplitude and radius \(a\) which yields a diffraction pattern \(u_F(r')\) in the Fourier plane. This diffraction pattern is multiplied by a transparent annular mask of inner radius \(p_1\) and outer radius \(p_2\) constituting a passband of spatial frequencies, and is transformed in the second stage of the 4-f system to the image plane. Thus, the two-stage transformation from the object plane to the image plane can be written as

\[
 u_i(r) = 4\pi^2 \frac{\lambda^2 f^2}{p_2^2} \int_{p_1}^{p_2} r' dr' J_0(\frac{2\pi r' r''}{\lambda f}) \times \int_0^a r'' dr'' J_0(\frac{2\pi r' r''}{\lambda f}) ,
\]

where an overall phase factor has been dropped. This image constitutes the ring source \(u_i(r)\) used to generate the Bessel beams in our experiments. Through a series of variable transformations this result can be simplified and expressed in a dimensionless form which is more convenient for numerical integration and experimental design,

\[
 u_i(\rho) = \int_{s_1}^{s_2} ds J_0(\rho s) J_1(s) ,
\]

where \(\rho = \frac{r}{a}\) is a dimensionless radius, \(s_{1,2} = \frac{2\pi}{\lambda f}(a p_{1,2})\) are dimensionless spatial frequencies, and the innermost integral was evaluated explicitly using the Bessel identity \(y J_0(y) = \frac{d}{dy}(y J_1(y))\). This exact result can be further approximated with high accuracy in terms of the Sine and Cosine integrals,

\[
 u_i(\rho) \approx \frac{1}{\pi \sqrt{\rho}} \left\{ Si[s_1(\rho - 1)] + Ci[s_1(\rho + 1)] \\
 - Si[s_2(\rho - 1)] - Ci[s_2(\rho + 1)] \right\} ,
\]

by substituting the asymptotic forms for the Bessel functions of large arguments. The effects of the upper and lower spatial frequency cutoffs \(s_2\) and \(s_1\) were investigated separately by direct numerical integration of Eq. (B.3.8). Figure [B.2] illustrates the effect of the high frequency cutoff \(s_2\) (with \(s_1 = 0\)), and indicates that the effect of \(s_2\) is to impose a Gibbs phenomenon on the sharp edge of the aperture. Figure [B.2] illustrates the effect of the low frequency cutoff \(s_1\) (with \(s_2 = 5000\)), and illustrates how the suppression of the lower frequencies yields a filtered image whose dominant features are concentrated about the sharp edge of the aperture. Numerical analysis suggests that the respective periods of oscillation observed in these graphs, \(\Lambda_2\) and \(\Lambda_1\), vary as \(\Lambda_2 = 2\pi/s_2\) and \(\Lambda_1 = 2\pi/s_1\), and these results
Figure B.2: Spatial filtering of a uniformly illuminated circular aperture: a) effect of high frequency cutoff $s_2$ (with $s_1 = 0$); b) effect of low frequency cutoff $s_1$ (with $s_2 = 5000$).

Figure B.3: Effect of high frequency cutoff $s_2$ on the shape of the filtered profile: a) amplitude, and b) intensity. i) $s_2 = 2184$; ii) $s_2 = 873$; iii) $s_2 = 437$ (all cases with $s_1 = 114$). Panels a,b(iii) correspond to the cutoffs used in our experiment.
also follow analytically from Eq. B.3.9. Clearly, the sharpest filtered edge (albeit with the lowest transmitted power) is obtained as both \( s_1 \) and \( s_2 \) approach infinity. However, in practical implementations of the 4-f system the high frequency cutoff is constrained by the sizes of lenses, mirrors, and other hardware. Moreover, while the overall width of the filtered edge is determined by the low frequency cutoff \( s_1 \), the shape evidently varies substantially with the high frequency cutoff \( s_2 \), as indicated in Fig. B.3. In order to achieve a practical ring source with a controlled, uniform profile, we specified the ratio \( s_2/s_1 = 3.83 \) as a design criterion, corresponding to roughly 1.2 periods \( \Lambda_2 \) of the \( s_2 \)-oscillations (i.e. the high frequency oscillations in Fig. B.3a) fitting into the region between the edge of the aperture and the nearest zero of the \( s_1 \)-oscillations (i.e. the low frequency oscillations in Fig. B.3b). The resulting ring source is displayed in Fig. B.3(iii) for the actual values of \( s_1 = 114 \) and \( s_2 = 437 \) used in our experiment. Because the \( s_1 \)- and \( s_2 \)-oscillations have the same inverse dependence on \( s_1, s_2 \), and the magnitudes of their overshoots are both independent of \( s_1, s_2 \), the shape of this profile is independent of the actual values of \( s_1, s_2 \) as long as their ratio remains the same. This result should prove useful in the design of experiments. Finally, we see that all of the ring sources produced by the 4-f filtering system yield an amplitude profile which is substantially anti-symmetric about the edge, in contrast to the even symmetry of an illuminated circular slit. Because of this odd symmetry, the diffracted field emanating from the ring source displays a null at a radius of \( \rho = 1 \) (\( r = a \)). However, the Bessel beam forms in a narrow region along the axis of the ring where the propagation angle from the normal is substantial. When combined with the spatial separation between the positive and negative lobes of the ring source based on the design criterion specified above, the resulting propagation delay between the two lobes is sufficient to impose a substantial degree of constructive interference along the axis. This propagation delay is identified by the parameter \( \Delta \) in Fig. B.1 and was a primary motivation for choosing a ring source whose lobes were as uniform in profile and as spatially separated as possible.

### B.4 Experimental Setup and Procedure

The experimental layout is illustrated in Fig. B.4. The 4-f system is comprised of lenses \( f_4 \) and \( f_5 \) (separated by \( 2f = 1000 \) mm) and spatial filter \( sf \), which transformed the uniformly illuminated circular aperture at \( obj \) to a sharply focused ring at \( img \). This ring source served as the generator of a freely propagating Bessel beam which was digitized...
and recorded using a CCD camera. The laser source was a 632.8 nm, 4 mW, Uniphase

1507P-0 polarized HeNe laser which was matched and focused through a 35 µm-diameter pinhole \( \text{ph} \) to improve the mode quality. The fixed polarizer \( p_2 \) was used to establish and maintain horizontal polarization in the transmitted beam, while the variable polarizer \( p_1 \) was adjusted between \( \sim 55^\circ - 70^\circ \) from the horizontal as needed to prevent saturation of the CCD camera. In order to compensate a slight beam deflection through the polarizer crystal, care was taken to re-steer mirror \( m_1 \) after every adjustment of \( p_1 \) to re-center the beam on the pinhole. The first null of the Airy pattern from the pinhole was allowed to expand to a diameter of \( \sim 20 \) mm, and the beam was then collimated by a 1''-diameter lens \( f_3 \) to provide uniform illumination of the object aperture. The circular aperture at \( \text{obj} \) consisted of a No. 5N steel washer (nominal ID 0.141''') mounted on a polished glass substrate with a dab of Bic® Wite-Out®. Aperture \( \text{ap} \) was an iris diaphragm adjusted to block stray light from leaking past the outer edge of the washer. In order to minimize aberrations and phase distortions in the 4-\( f \) spatial filtering system, lenses \( f_4 \) and \( f_5 \) were 2''-diameter compound lenses, each consisting of two 1000-mm plano-convex lenses mounted apex to apex. This compound configuration effectively minimizes longitudinal spherical aberration without the use of specialized achromats.\(^{52}\) The spatial filter \( \text{sf} \) itself consisted of a 1/4''-diameter steel washer with its center filled with Wite-Out®, suspended concentrically by a
Figure B.5: CCD image of the ring source used to generate the Bessel beams in our experiment, with $s_1 = 114$ and $s_2 = 437$. Note the well resolved double-lobed profile.

eine wire at the center of a 3″ iris diaphragm. The inner diameter of this iris was set to 24.3 mm corresponding to the design criterion of $s_2/s_1 = 3.83$ as discussed in Section II.B. In placing the spatial filter, we observed that the object beam came to a focus almost one inch downstream from the Fourier plane located halfway between lenses $f_4$ and $f_5$, indicating that the object beam was slightly diverging; this did not, however, affect the formation of Bessel beams. We also confirmed independently that variations as great as one inch in the longitudinal placement of the spatial filter did not affect the formation of Bessel beams. The filtered image revealed numerous bright features both within and outside of the ring source (see Fig. B.5), arising from phase objects such as dust particles and inclusions on the object substrate and from multiple reflections from the lenses in the beamline. Although these features were found to have a negligible effect on the subsequent formation of the Bessel beam, they were for most measurements blocked in the image plane by an iris diaphragm closed down to a diameter of 4.5 mm outside of the ring source (the “outer mask”), and by a 2.5 mm-diameter circular dab of Wite-Out® on a microscope slide within the ring source (the “inner mask”). Distinct Bessel beams formed at distances roughly 9 cm from the ring source and beyond. All images were recorded with a Santa Barbara Instrument Group CCDH-1 camera (640×480 pixels, 7.4 µm pixel size) and ST-237 CPU with 12-bit resolution connected to a PC. The exposure time was set to 10 ms for all images.
B.5 Experimental Results and Analysis

A CCD image of the ring source generated using the 4-f system is shown in Fig. B.5 and displays the expected double-lobed structure as well as lower-intensity ripples both inside and out. The image plane of the 4-f system was located empirically by translating the CCD camera along the beam in steps of 0.5 mm and identifying the single image which was distinctly the sharpest. The diameter of the null at \( \rho = 1 \), measured from the CCD image at several locations around the azimuth, was found to be \( 2a = 3.62 \pm 0.01 \) mm \((0.1425''\)) consistent with unity magnification of the No. 5N washer. Figure B.6 compares the radial dependence of the measured intensity profile with the theoretical result from Fig. B.3b(iii). The measured profile was obtained numerically from the CCD data by interpolating the radial profile (at fixed azimuth) in steps of 0.1 pixels using 2-dimensional linear interpolation between the four nearest pixels, and then averaging these radial profiles about the azimuth in steps of 0.1°; regions with visible inclusions near \( \rho = 1 \) were excluded. The simulated profile was obtained by convolving the theoretical curve from Fig. B.3b(iii) with rectangular bins 7.4 \( \mu m \) wide. The relative intensities of the lobes and ripples, and their absolute radial separation, show quite satisfactory agreement with theory. The slight discrepancy in the widths of the lobes may result from charge leakage between the pixels (i.e. “blooming”, visible as the downward streaks in Fig. B.5), from a possible limitation in the depth of focus due to the 0.032''-thickness of the No. 5N washer, or from diffraction of the lobes themselves (whose widths yield a Rayleigh parameter on the order of 0.5 mm). The straightforward generation of Bessel beams by spatial filtering, the essence of the current experiment, is illustrated in Fig. B.7 which was recorded at a distance of \( z = 22 \) cm from the image plane. In Fig. B.7a the spatial filter was removed from the 4-f system, so that the image plane contained an unfiltered replica of the object and yielded the characteristic diffraction pattern of a uniformly illuminated circular aperture with a Fresnel number of \( N_F \equiv a^2/\lambda z = 23.5 \), consistent with the \( \sim 24 \) maxima that can be seen to span the diameter. In Fig. B.7b the spatial filter was re-inserted, and the diffraction pattern was replaced with a Bessel beam. Both pictures were recorded with the outer mask in place and the inner mask removed at the image plane. Representative images of the Bessel beam at distances of \( z = 19 \) cm, 23 cm, and 29 cm are illustrated in Fig. B.8. The accompanying graphs show the intensity distribution in a horizontal section through each image together with a least squares fit of the lowest order Bessel function. Both the outer and inner masks were
Figure B.6: Radial profile of the ring source used to generate the Bessel beams in our experiment. The measured curve is an average profile extracted from the CCD data as described in the text. The analytical curve was calculated from the theoretical result in Fig. B.3b(iii).

inserted in the image plane at the ring source for these measurements. The fitting function was of the form

$$I(r) = A J_0^2 (k_t(r - r_0)) + B$$  \hspace{1cm} (B.5.1)$$

with the free parameters $A$, $B$, and $r_0$ determined by a nonlinear least-squares Marquardt-Levenberg algorithm. The measured intensity distributions show satisfactory agreement with the theoretical Bessel beam profiles with respect to the overall amplitudes and radii of the successive rings, but some irregularities remain due to aberrations. These irregularities were eliminated in the intensity distributions in Fig. B.9 which were generated numerically from the CCD data by performing a full azimuthal average of the linearly interpolated radial profiles in the manner of Fig. B.6. The resulting cross sections show excellent agreement with the theoretical fits (reduced $\chi^2 < 5 \times 10^{-4}$), and confirm the radial dependence of our Bessel beams out to beyond the 11th ring.

Figures B.8 and B.9 also clearly reveal the growth of the Bessel beam with distance, as suggested by Fig. B.1 and predicted by Eq. B.3.3. If we define the first zero of $J_0(k_tr)$ as $j_0 = k_tr_{cl}$, we may express the radius $r_{cl}$ of the central lobe as

$$r_{cl} = \frac{j_0}{k_T} = \frac{j_0 \lambda z}{2\pi a}$$  \hspace{1cm} (B.5.2)$$
where \( j_0 = 2.40483 \) and \( k_T = \frac{2\pi a}{\lambda z} \), showing that the beam radius increases in direct proportion to distance \( z \) from the source.

To quantify the \( z \)-dependence experimentally, CCD images were recorded in 1 cm increments at distances between 2 cm and 36 cm from the source. Both the outer and inner masks were inserted in the image plane at the ring source for these measurements. Distinct Bessel beams formed at distances \( z > 9 \) cm, and the corresponding radii of the first zeros were extracted by fitting Eq. B.5.1 to the 2-dimensional CCD data. The resulting beam radii are plotted as discrete points in Fig. B.10 together with the linear theoretical prediction from Eq. B.5.2, and demonstrate excellent agreement at distances between 10 cm and 26 cm from the source.

To investigate the nature of the deviations from linearity which are evident for \( z < 9 \) cm and \( z > 26 \) cm, we simulated the formation of Bessel beams by integrating Eq. B.3.2 using the theoretical ring source from Fig. B.3a(iii) as the source distribution \( u_i(r') \), and then fitting Eq. B.5.1 to the resulting 1-dimensional profiles. The experimentally observed deviations are fairly replicated by these simulations, and thus appear to be intrinsic to the diffraction of light in our experiment. The simulations reveal, in particular, that the light diffracting from the ring source does not substantially cross the optical axis to form a well-defined Bessel beam until \( z \approx 7.5 \) cm. They further reveal that the null between the (+) and (−) lobes of the ring source in Fig. B.3(iii) diffracts into a sharply defined "zone
of destructive interference” that diverges linearly and engulfs the optical axis at \( z \approx 27 \) cm. Both of these limits coincide well with the start and end of the linear region. The slight axial displacement of 1-2 cm between the linear regions in the experiment and simulation would be accounted for if the lobes of the ring source were 20% larger than predicted, which is not inconsistent with the measured profile in Fig. B.6.

Finally, it is interesting to compare the angular divergence of the Bessel beam in Fig. B.10 with the angular diffraction of a Gaussian beam. In particular, a TEM\(_{00}\) beam with a spot size of \( w_0 = 13 \) \( \mu \)m, equal to the smallest Bessel beam radius observed in the linear region, would have far-field angular divergence of 15.5 mrad, more than 110× greater than the divergence angle exhibited by the Bessel beam in Fig. B.10 and would expand to a radius of 2.5 mm over a distance of 160 mm, more than 70× greater than the 35 \( \mu \)m Bessel beam radius at the same distance.

## B.6 Conclusions

We have demonstrated a simple and straightforward method of creating Bessel beams using a 4-\( f \) spatial filtering system requiring no specialized optical components, and have described the robust design of a spatial filter capable of yielding Bessel beams that propagate over relatively large distances. The Bessel beams produced in the present experiments, while not entirely divergence-free, nevertheless diverged more than 110× more slowly than a diffracting Gaussian beam of the same spot size.

It is instructive to compare and contrast our Bessel beam with other types of beams such as the Gaussian beam above, nondiffracting Bessel beams, and diffracting beams of a more general nature. Previous authors\cite{42} have pointed out, for example, that nondiffracting Bessel beams are distinguished from the Airy pattern of an illuminated circular aperture by the fact that the latter involves a (1\(^{st}\)-order) Bessel function whose argument depends explicitly on distance and thus suffers diffraction. We would suggest that a more important distinction is the fact that the Airy pattern exhibits an overall 1/r-dependence that real Bessel beams (including ours) do not, and thus cannot be converted into a nondiffracting beam by a simple optical transformation. In contrast, while the \( z \)-dependent Bessel beams produced in our experiment are not entirely divergence-free, they nevertheless exhibit all of the other properties of nondiffracting Bessel beams, and differ from them only in their spherical wavefronts which can in principle be eliminated by a collimating lens.
In practice, collimation of the Bessel beams in our experiment would be complicated by the nature of the ring source, whose “zone of destructive interference” would largely nullify the field within the aperture of the lens. But from a pedagogic perspective, the experiment has many merits beyond its simple construction, perhaps the most important of which is a broadening of the concepts of diffraction-limited beam optics through a detailed introduction to the theoretical and physical properties of Bessel beams. Other benefits include the fundamental physical insight on the composition of beams as a superposition of plane waves, simple examples of diffraction in the Fresnel approximation, an introduction to the concepts of Fourier optics and spatial frequency analysis, and the opportunity for the student to employ numerical methods in both theoretical and experimental calculations. We thus anticipate that the greatest utility of these experiments will be found in the advanced undergraduate laboratory.

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Figure B.8: CCD images (left) and horizontal sections (right) of the Bessel beams at: (a) \( z = 19 \) cm; (b) \( z = 23 \) cm; (c) \( z = 29 \) cm. The numerical fits on the right are defined by Eq. [B.5.1]
Figure B.9: Azimuthally averaged radial profiles of the measured Bessel beams from Fig. B.8. The numerical fits are defined by Eq. B.5.1.
Figure B.10: Bessel beam radius vs. distance from the ring source. Measured points were obtained from a 2-D fit of Eq. B.5.1 to the CCD data. The linear theory is the prediction Eq. B.5.2. The simulation is the result of a numerical solution to the diffraction problem based on the theoretical ring source of Fig. B.3a(iii).
Bibliography


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