Efficient Acceleration of Electrons by an Intense Laser and its Reflection

Dissertation

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By

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Abstract

Here I present an experimental, theoretical, and computational exploration of an extremely efficient scheme for laser-based acceleration of electrons. A series of experiments were performed at the Air Force Research Laboratory in Dayton, OH, to show that a high-repetition-rate short-pulse laser (3 mJ, 40 fs, 1 kHz) normally incident on a continuous water stream can accelerate electrons in the back-reflection spray with >1% laser-to-electron efficiency for electrons >120 keV, and with >MeV electron energies present in large number. Characterization of the accelerated electrons was followed by explorations of appropriate focal conditions, pre-plasma conditions, and laser-intensity parameters. These experiments show clear signatures of plasma instabilities, with substantial $\frac{3}{2}\omega$ and $\omega/2$ optical harmonics detected concurrently with efficient electron acceleration. Particle-in-cell (PIC) simulations of high-intensity laser interactions are able to reproduce the electron energies and acceleration efficiencies, as well as plasma instabilities. Analysis of the simulations suggest that electrons are accelerated by a standing wave established between incident and reflected light, coupled with direct laser acceleration by reflected light. Using hydrodynamic simulations of the laser pre-pulse interaction as initial conditions for PIC simulations of the main-pulse interaction clarifies mechanisms by which experimental manipulation of pre-pulse has effectively determined electron-acceleration efficiency in the laboratory.
Dedicated to my brilliant wife, Megan, and to both of our families.
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This once-classified American flying saucer now sits on exhibit next door to our laboratory, in the National Museum of the U.S. Air Force. Its display plaque states that while this aircraft was designed to fly at Mach speeds, dangerous aerodynamic instabilities prevented it from ever breaking 40 mph. As a graduate student, I have occasionally walked by it and pondered: sometimes about nature’s opinion of our big ambitions, and sometimes about insufficient communication of scientific research. Image credit: Wikimedia Commons.
Chapter 1: Introduction

1.1 Strong X-ray radiation in the “wrong” direction

As I will describe throughout this dissertation, what we have observed in our experiment was unexpected: 2 MeV electron radiation where we ought have seen 80 keV radiation, hundreds of times more electrons than anyone thought was reasonable, strongest radiation with our laser at an angle that should have made radiation weakest, and a high-energy electron spray in the wrong direction. Two years ago, however, these details were not apparent; rather, what was known was that a surprisingly efficient source of laser-produced X-rays in the laboratory had resulted in nearly 1 Rem/hr radiation dose at the back of the chamber, higher than had we had ever heard of from such a laser. It was at this point that I was brought into the research group to study of the source of unexpectedly high laser-produced radiation in the laboratory. The dissection of this physical phenomenon is the subject of this dissertation.

I learned how to perform high intensity laser experiments on the Scarlet laser at Ohio State. As part of this group, I performed ultra-high intensity experiments of electron and ion acceleration, and developed a knack for computational data analysis and experimental diagnostics development. The Scarlet laser can fire its highest-intensity pulse once-per-minute, but, in practice, experiments are limited to about ten shots per day. Thanks to our continued efforts at Ohio State in high-acquisition

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· *A novel femtosecond-gated, high-resolution, frequency-shifted shearing interferometry technique for probing pre-plasma expansion in ultra-intense laser experiments*, Feister et al.. Copyright 2014, AIP Publishing LLC.
rate diagnostics and regenerating targets, Scarlet’s experimental repetition rate is steadily advancing towards its once-per-minute limit.

In the context of other large laser facilities, a once-per-minute repetition rate is actually quite high; some lasers require an hour or more of cool-down time between shots. For my dissertation, however, I decided to focus on a much higher-repetition rate laser system, where the laser can fire one thousand times per second and the data acquisition rate is limited only by its computer control. I did this experimental dissertation project on the 1 kHz Red Dragon laser system at the Air Force Research Laboratory (AFRL) at Wright Patterson Air Force Base (Fig. 1.1). This laboratory is located in Dayton, OH. While repetition rate at AFRL is sixty thousand times higher than at Scarlet, energy per pulse is one thousand times lower. This puts frontier high intensity laser research out of reach, but enables a range of possibilities for deeper understanding of plasma dynamics at high intensities that may have passed unnoticed in the global scientific race to study physics at the highest laser intensities. The AFRL laser facility is physically smaller than Scarlet (e.g. one room vs. four rooms). This can be a benefit in that there are fewer laser-related components and they are quick to replace or repair, leaving more time to perform experiments. Furthermore, a smaller laser is quicker to align, leaving yet more time-per-day for experiments. Because the repetition rate is 1 kHz rather than a few times per day, one can (and we now do) obtain a flood of experimental data in minutes (with the right choice of experimental diagnostics and the automation of data acquisition). The AFRL Red Dragon laser is the second-most-intense laser in Ohio (second to Scarlet, of course).
Figure 1.1: **AFRL Red Dragon laser system.** The modified Red Dragon system with which I did my dissertation experiment takes up one room. Inside a vacuum-sealed interaction chamber, the laser comes to small focus on a stream of water, creating plasma. Labeled are parts of the experimental apparatus; if the laser-system terms are not familiar, revisit this figure after reading Chs. 2 and 3.

The incredibly efficient radiation source was discovered at AFRL before I arrived, and partly by accident. While the AFRL experiment was set up to measure X-rays emitted in the direction of the laser, what was actually found was strong X-ray signal in the opposite direction: in the laser reflection, or specular, direction. The research team did not begin investigating this effect until a radiation dosimeter (i.e. a Geiger counter) was placed, for safety reasons and through curiosity, outside the target chamber behind the focusing optic. At that point, they measured radiation levels of nearly 1 Rem/hr! This is a high rate of radiation; the standard workplace limit for one year is 5 Rem, total. Fortunately, nobody was unsafely exposed; it is standard laboratory procedure to stay clear of the laser-plasma interaction area at all times during operation. Still, the research team immediately shut down the experiment and notified the Air Force radiation officers, and the laboratory safety was improved through lead shielding appropriate for this strength of radiation.

Once safety was addressed, the group was able to turn to the fact that this radiation source is scientifically surprising; radiation dose of this magnitude, in this direction, was unexpected. As physical scientists, we seek to understand such surprises. A first experimental study coarsely characterized the radiation at various locations
around the interaction chamber, using a radiation dosimeter. A second experimental study aimed to understand the energy of X-ray emissions that were detectable at a large distance from the interaction. It was at this stage that I was invited to join the research team, with a specific goal: build a toolkit to better characterize this radiation source, and then determine fundamentally what drives it. I gladly took on this task, because it was very appealing; the problem was rife with the potential for interesting plasma dynamics. I would need to bring to bear computational and laboratory skills I had honed over the past four years at the Scarlet laser, and I could do this in an environment where a thousand interactions per second between laser and plasma were available for study. Creative computational data analysis, design of novel high acquisition rate diagnostics, and analytic thinking were required to solve this problem; I thought: “I can solve this problem. I can figure out what’s going on here.” And so, with that naïveté, I plunged in headfirst; and I must say, it has been a very rewarding experience.

Typically, when one thinks of the radiation emitted from short-pulse laser-matter interaction, one thinks of two broad categories. The distinction between these regimes occurs when the laser intensity can cause an electron to become relativistic in a single laser cycle. This relativistic threshold occurs at an intensity of $10^{18}$ W cm$^{-2}$ for an 800 nm laser. At sub-relativistic intensities, e.g. $10^{15}$ W cm$^{-2}$ or $10^{16}$ W cm$^{-2}$, electron acceleration is dominated by effects such as resonance absorption [1] which cause keV-energy isotropic radiation and/or radiation at angles other than the laser incidence or reflection axis. At higher intensities, say $10^{20}$ W cm$^{-2}$ or $10^{21}$ W cm$^{-2}$, relativistic interaction effects dominate and cause very high-energy radiation (tens, hundreds, or even thousands of MeV) which is biased forward along the laser axis. The AFRL laser system lies at the relativistic threshold, so one expects to see some combination of low-energy isotropic and higher-energy laser-direction radiation.

A substantial literature of simulation/modeling papers exists that seeks to understand laser-direction electron acceleration from laser interaction with solid targets [e.g. 2, 3, 4, 5]. A number of other papers investigate electron acceleration along other axes from laser light obliquely interacting with solids [6, 7], studying electrons preferentially emitted at large angles relative to the incident laser. Significantly less attention has been devoted to speculatively directed electron acceleration mechanisms.

2When I write of a “relativistic electron” or an electron with “relativistic energy”, I mean that the electron’s kinetic energy (e.g. 1 MeV) exceeds its $E = mc^2$ rest mass energy (511 keV).

3See also [8, 9, 10, 11, 12, 13, 14, 15, 16, 17].
near normal laser incidence. Normal laser incidence means that the target plane is perpendicular to the direction of laser propagation. In the case of normal incidence, the laser reflects back on itself (hence, the reflection angle is $0^\circ$). I call a measurement of electrons emitted in that backward direction a $0^\circ$ measurement.

The normal incidence, $0^\circ$ electron measurement configuration has been generally avoided in relativistic-threshold-intensity physics experiments because it is seen as both difficult and uninteresting. First, a laser reflected from the target with $0^\circ$ reflection angle will retrace its original optical path, risking damage to optical components as it travels backwards through the laser amplification system. Second, the measurement is difficult because at $0^\circ$ (normal incidence), the detector apparatus gets in the way of the laser. That is, measurement of electrons accelerated towards the laser mirrors would suggest placing electron detectors directly into the laser beam path (preventing the laser-target interaction from occurring). Third, and most problematically, the measurement is seen as uninteresting because well-understood mechanisms for electron acceleration in this intensity regime work best with large angles between the laser and target. These mechanisms lose efficiency as the laser-target geometry approaches normal incidence from wide angles, so why go through the trouble of such a difficult measurement?

We found evidence that electron acceleration could be actually be enhanced rather than reduced in a normal incidence configuration, and we were in a unique position to make the $0^\circ$ measurement. First of all, our laser is small, and although we did damage a laser amplifier while learning protective techniques, our system is flexible enough that it could be quickly repaired. Second, the laser repetition rate is 1 kHz (rather than a few times per day), so an uncharted parameter space could be rapidly explored. Third, our final laser mirror is fairly inexpensive, and we were willing to “sacrifice” the optic (and its reflection quality) by drilling a hole in it to make a $0^\circ$ electron energy measurement. This hole was necessary to allow electrons, accelerated at $0^\circ$, to exit the incident laser beam path and be characterized. Our goal in implementing this difficult setup was to characterize the AFRL radiation source, to understand the mechanisms involved with specularly accelerated electrons in a qualitative way, and to fill a gap in the literature. Insights into this particularly efficient electron acceleration will ultimately create a foundation for further optimization of the electron beam parameters (energies, total charge, emittance) in this and related experiments.
Prior experiments in laser-based electron acceleration to $\lesssim$MeV final energies have noted that acceleration mechanisms can be enhanced through the presence of a laser pre-pulse [18]. The importance of pre-plasma scale length is well established [19, 20], in the emission of electrons and X-rays. The laser pre-pulse, which is purposeful or accidental laser energy deposited prior to the ultra-intense laser interaction, is the usual source of pre-plasma. The pre-pulse serves to create an expanding plasma in front of the target and the presence of this plasma allows for a rich variety of laser-plasma interactions that would not occur with a sharp vacuum-solid interface [21]. Where experiments with laser and target conditions similar to ours have noted the importance of pre-plasma, they have also highlighted the importance of oblique incidence and $p$-polarization.\(^4\) It has been noted that electron acceleration, to energies exceeding the laser’s characteristic ponderomotive energy [22], is possible at relativistic threshold intensity with oblique incidence, $p$-polarized interactions. The laser-to-electron acceleration efficiency is typically much less than 1%; Mordovanakis et al. [18] used a 3 mJ laser to accelerate 7 pC of charge with an energy spectrum with highest spectral density at 0.8 MeV ($\sim$0.2% laser-to-electron conversion efficiency), and Mao et al. [23] used a 240 mJ laser to accelerate 100 pC of electrons per shot with similar energy ($\sim$0.04% efficiency). These results (which, relative to the conical electron spray presented in this study, show well-collimated electron beams) depend sensitively on the $p$-polarization of the incoming laser pulse and work best at large angles of incidence; in these experimental cases, $\geq 45^\circ$.

\(^4\)In a $p$-polarization interaction, the laser’s electric field oscillations are (by definition) not parallel to the target plane. The result is that the electric fields push and pull electrons into and out of the target, and electrons traverse the density variations in the pre-plasma. In an idealized normal-incidence interaction, laser fields are completely parallel to the target (and so a normal incidence interaction is not $p$-polarized).
Figure 1.2: **Side view diagram of laser and water target.** In this simplified view of our experiment, an infrared (IR) laser is focused onto a flowing stream of water (column-shaped stream, as would come out of a hose). Under high pressure, water exits a 30 μm diameter nozzle and flows laminarly; the laser interaction occurs 1 mm downstream from the nozzle. The pre-pulse of the laser creates a plasma and vapor cloud that extends in front of the water column; the main pulse interacts with this expanding plasma and is reflected. A side view of the pre-plasma and the laser interaction is provided by hundred-fs-resolved probe laser shadowgraphy (Appendix A).

Unlike the aforementioned studies, we describe an experimental measurement of several hundred pC of charge (>120 keV) accelerated anti-parallel to the laser, generated from a 3 mJ laser focused onto a water jet (Fig. 1.2). This represents a vast improvement in total accelerated charge over previously reported electron acceleration measurements with mJ-class lasers interacting with high density targets. Since the electrons of the total-charge measurement were filtered to exclude those <120 keV, this conservatively represents >1% laser-to-electron conversion efficiency. The presence of numerous >MeV electrons is confirmed through experimental energy spectrum measurements. This energy is well in excess of the predicted 80 keV characteristic energy [22] for this laser intensity. As might be expected given the significant improvement in total charge accelerated, previously identified mechanisms relying on p-polarization and oblique laser incidence do not explain our results, which are obtained at normal incidence.
Over the past two years, our AFRL research group, which consists of a variety of AFRL physicist-engineers, Ohio State professors, postdoctoral fellows, graduate students, and at times undergraduates, has endeavored to solve this mystery: What is this radiation source, and what is driving it? After many experimental studies, we can confidently say we have well-characterized the radiation source. The AFRL radiation source is primarily energetic electrons, with energy spectral components well in excess an MeV, and with hundreds of pC of >120 keV electrons per laser pulse. To obtain this characterization, the experiment needed to be adapted in a variety of ways, which will be discussed in this dissertation, and it was necessary to design and implement novel high-acquisition-rate electron diagnostics.

The answer to what is driving this source remains still an active topic of research, but we have made tremendous progress in computational modeling of the interaction and development of theoretical models. One thing we can say now with near-absolute confidence is that a large pre-pulse is essential to this acceleration mechanism. We have experimentally begun to narrow in on the exact pre-pulse requirements. Strong optical harmonics present in reflected light tell us about this pre-pulse condition. The evolution of pre-plasma is monitored using a custom-designed probe laser which casts a shadow of the target (and more). Diagnostics have been developed to characterize the nanosecond pre-pulse and shown how fluctuations of laser pre-pulse are correlated with modified electron-acceleration. We have developed creative techniques for multivariate manipulation of the laser pre-pulse. In the most extreme case, we are able to deliberately control the amount of electron acceleration, changing dose outside the experimental chamber from 1 Rem/hr to 0.005 Rem/hr within seconds, by modifying nothing in the experiment except the laser pre-pulse.

Beyond laser pre-pulse setting the stage for electron acceleration, we must ask: What else do we know? Or, what do we think we know? One of the most convincing mechanistic explanations, identified through prior literature and through our simulations, are those of standing-wave acceleration, direct laser acceleration, and electrostatic acceleration. An electromagnetic standing-wave occurs when the laser creates interference with itself as it reflects from the target. When this interference leads to electron acceleration, this is called standing wave electron acceleration. Building from prior work on standing wave electron acceleration, we have delved into how a laser and its reflection can accelerate large numbers of electrons in the reflection direction. We have developed a theoretical model for this involving electrostatic fields.
and direct acceleration by the laser reflection.\(^5\) The role of plasma wave instabilities, suggested through experimental data, has been explored through simulations, and explanations of the concurrence of these plasma waves with electron acceleration has been developed. Ultimately, we have analyzed more than ten consecutive experimental campaigns\(^6\) and well over a hundred individual simulations to develop our best possible understanding. This understanding is synthesized in this dissertation, and we can speak to the radiation source in a much more clear way than when we started.

Finally, having brought together our best efforts in the laboratory, simulation, and theory, I have attempted to create our most realistic simulations to-date. I did this by extending our project’s simulation of laser pre-pulse and combining this with a simulation of the main pulse interaction, and then replicating our laboratory alignment procedure within the simulation. The results have been promising, improving the match between simulation and experiment. As with our other simulations presented in this dissertation, high numbers of electrons are emitted, with high energies, and in the direction of laser reflection. The dynamics of these most-realistic simulations are quite complicated, more so than our idealized theories and simulations. While a full analysis of these most-realistic simulations is outside the scope of this dissertation, I will present a few experimental insights already obtained from these simulations.

We build our understanding on the foundation of significant prior research, and there is still much research to be done within our group. I present here the work we have done and the state of our understanding on the efficient acceleration of electrons from the interaction of a laser and its reflection.

### 1.2 Further motivation for study

High-intensity lasers (>10\(^{17}\) W/cm\(^2\)) have enabled particle acceleration to high energies (keV to GeV) through a variety of methods and mechanisms, all of which exploit the large laser and plasma electric fields present during intense laser interactions with matter. Such laser sources are capable of accelerating electrons to GeV energies through direct laser-field and/or laser wakefield acceleration \([24, 25, 26]\). Direct laser-field and laser wakefield acceleration are two ways a laser can provide strong

\(^5\)“Direct laser acceleration”, or direct laser-field acceleration, refers to the ability of an above-relativistic-threshold intensity laser to push electrons in its propagation direction.

\(^6\)In this time, we have enabled literally billions of high-intensity laser pulse interactions with water. The overwhelming majority of interactions were not recorded at all. Many others were recorded as part of an average, and some were recorded individually.
electron acceleration over a short distance, and direct laser acceleration will be discussed in the context of this dissertation study in Ch. 4. In addition, ultra-intense laser-plasma interaction can create bright X-ray sources from high harmonic generation, Bremsstrahlung collisions of electrons, betatron oscillations [27], and other techniques. These techniques could be applied to our research in the future. However, with the exception of Bremsstrahlung, they were not employed in this dissertation work and will not be discussed.

Laser-based acceleration of electrons to a relativistic energy of order 1 MeV with mJ-class tabletop short-pulse lasers could enable a high repetition rate (up to 1 kHz), compact, ultra-short (<100 fs) relativistic electron source. Important applications for laser-accelerated electron beams include radiotherapy with electron energies well above what can be achieved with conventional linear accelerators [28, 29].

The creation of highly pulsed X-ray and UV radiation through Compton scattering of another pulse of light with the electron beam presents another unique opportunity. While there has been much success in Compton scattering off electron beams accelerated from underdense targets, e.g., from laser wakefield acceleration [30], Naumova et al. [31] highlight the possibilities that sub-fs bunches of electrons from laser-solid interactions offer for creating Compton light sources with extremely short timescale pulsing. Using ultra-intense, ultra-short lasers to create these electron beams can significantly shorten the temporal duration of the resulting electron pulses, even to timescales shorter than what can be achieved with photocathode technology [32]. Thus laser-accelerated electron bunches may provide some advantage to efforts to create free electron lasers [33]. A number of groups are investigating laser-accelerated electron sources with these goals in mind.

1.3 Outline

The remainder of this dissertation is organized in the following way: experiment, theory, simulation, and then conclusion. As the primary experimental apparatus is a high-intensity laser, techniques for high-intensity laser manipulation and diagnosis are shared in Chapter 2. Experimental studies to characterize and understand the unique AFRL radiation source are described in Chapter 3; each experimental study motivates the next. A theoretical framework is developed in Chapter 4 with which to understand our experiment, and simulations are proposed as a tool for further exploration. Laboratory conditions are recreated through high-performance-computing
simulation in Chapter 5. These simulations lead to improved understanding of the
electron acceleration which underlies our experimental observations; the chapter ends
by summarizing our best physical interpretation of results. Chapter 6 concludes
the dissertation with a discussion tying together our experimental, theoretical, and
computational scientific research. Four appendices provide additional details of shad-
owgraphy, interferometry, X-ray diagnostics, laser considerations, and the design and
calibration of a high-acquisition-rate electron spectrometer.
Chapter 2: High-intensity laser as an experimental instrument

Before I could begin to tackle the challenge of understanding the unique qualities of the laser-produced radiation at AFRL, I needed to develop a strong understanding of the experimental instrument. A well-rounded experimental high energy density (HED) plasma physicist is competent both in the plasmas which are studied and the instrument that drives them. The scientific instrument needs to be understood even in the extreme case that you never once touch it (completely unheard of at Scarlet or AFRL, but a real possibility at large government science facilities), because you still must recognize its quirks and limitations when designing and interpreting your experiment. Even in the middle of an experiment, for example, a laser-plasma physicist may recognize a slight smear in a laser focus image as a symptom of broader laser misalignment and halt everything to prevent an invalid experimental result.

Ultra-high-intensity lasers are the plasma-creating instrument of which I have become expert. Other sources for HED plasma include self-imploding electrical wires called Z-pinch[es] [34], high-energy/long-pulse lasers [35, 36], and high-energy ion beams [37], all of which I have become familiar with through research and conferences but which are outside the scope of this dissertation. The basic schema of every high-intensity laser experiment is as follows:

\[
\text{Generate short pulse} \rightarrow \text{Amplify to high energy} \rightarrow \text{Focus onto target} \quad (2.1)
\]

The experimental latitude comes in the details, of course. How short do we make the pulse? How many pulses per second do we amplify, and to what energy? How tightly do we focus the laser, and onto what target; what are we trying to study? While I cannot go into all the details of our experimental setup, nor do justice to the history

\[7\]

I worked four years with Ohio State’s Scarlet laser and two years with AFRL’s Red Dragon laser; Table 2.1 compares these two systems.
and continued development of the ultra-high intensity short-pulse laser, I will touch on the most important elements to my core dissertation narrative.

In this chapter, I will walk you through selected techniques relevant to creating and manipulating a high-intensity laser, and how these have specially adapted to suit our experimental goals. While I am doing this, I will describe a handful of ways to characterize high-intensity lasers. We would not be able to reproduce, theorize about, or computationally model our experiment without these characterizations. I will tie each of these laser instrument techniques, tools and characterizations directly into the subject of upcoming chapters. This chapter will speak to the laser instrument and deliberately exclude the tools and techniques used in observing the interaction between the laser and target (our “experimental diagnostics”); these will be covered in the context of the experiment throughout Ch. 3.

2.1 Short-pulse lasers begin with short-pulse oscillators

Every laser begins with a laser oscillator. An oscillator is a device in which light bounces back and forth between two mirrors. In between these mirrors is a “gain medium,” e.g. a crystal or CO\textsubscript{2} gas, which gives up energy (via atomic transitions) to the light as it passes through. By design, the first mirror is not 100\% reflective, so some light always leaks out while the majority of light is reflected to the gain medium for re-amplification. The little bit of light that leaks through the mirror is the laser output.

In the off-the-shelf short-pulse laser oscillator design used in the laboratories at Ohio State and AFRL, the choice and positioning of the Ti:Sapphire crystal and the mirrors of the laser oscillator are rigged in such a way that nature finds it energetically favorable to bounce a single very short pulse, rather than a continuous beam of light, back and forth between the mirrors [38]. The short pulse makes endless round trips back and forth between two mirrors at the speed of light; it tallies twelve feet of travel per round trip, which takes 12 ns at the speed of light. Every time the short pulse reflects off the first mirror, it splits and a laser pulse leaks out; hence, the oscillator.

---

\textsuperscript{8}Actually, no mirrors are 100\% reflective. High-quality silver mirrors are still only about 96\% reflective, and even specially designed, layered-dielectric mirrors are in the 99.9\% range. Household mirrors are usually mounted on opaque materials, hence our common intuition that silver mirrors are purely reflective, and 100\% opaque. With dielectric mirrors, ultra-high reflectivity for certain wavelengths is often traded for low reflectivity at others. Because the partially reflecting dielectric surface sits on transparent glass, you can see through most of the mirrors in our laser lab.
leaks out about 80 million short pulses per second. Each exiting pulse can be thought of as a few-micron-thick packet of light (it lasts less than 30 fs) with an energy in the range of picojoules.

There are two reasons I wrote about the laser oscillator here. First, at a basic level, I think it is important to understand where our laser starts. Second, we take advantage of the precise timing between each of the 80 million pulses per second exiting the laser oscillator to probe the plasma conditions with huge temporal latitude and precision. This trick of generating a secondary, diagnostic laser from the oscillator pulses is further described in Appendix A.

2.2 Pockels cells pick out pulses

While the laser oscillator emits \( \sim \)80 million low-energy laser pulses per second, it is impossible with current technology to amplify all 80 million to high energy for use in our experiment.\(^9\) Much more reasonable is to amplify a small subset of these pulses, and this is what is done. Only one in 80,000 pulses is amplified in the stage subsequent to the oscillator; the 80 MHz pulse train is reduced to a 1 kHz pulse train. This decimation is achieved through a crystal called a Pockels cell.

A Pockels cell is a crystal which, upon application of high voltage, slightly changes its optical index of refraction properties [39]. Specifically, light passing through a Pockels cell will have its polarization rotated (e.g. changed from vertical to horizontal polarization) when a specific level of high voltage is discharged through a capacitor, across the bulk crystal. The Pockels cell pulse-picking system consists of a pulsed high voltage supply (Fig. 2.1a), a Pockels cell (Fig. 2.1b), and two polarizers. If you input a series of laser pulses, and quickly turn on and off the high voltage just once, you can rotate the polarization of just one pulse from the series. The voltage on/off rate must be very fast to make this work; as mentioned earlier, there is only about 12 ns time delay between laser pulses coming out of the oscillator. Polarizers complete the pulse selection process. A polarizer before the Pockels cell blocks all but a single polarization of input light. A polarizer after the Pockels cell reflects one polarization and transmits another, ensuring that only pulses with rotated polarization pass to

\(^9\)If you wanted to amplify every pulse coming out of the laser oscillator to Scarlet’s output energy of 15 Joules, you would be making 1.2 GJ of laser energy every second, immediately destroying the laser itself with all the waste heat – not to mention, after adding in wall-plug-to-laser inefficiencies, needing tens to hundreds of coal plants to power the thing.
the next amplifier. By flipping the voltage for just one pulse every ms, you get an output pulse train at 1 kHz.

Figure 2.1: **Damaged Pockels cell high voltage driver and crystal.** Pockels cells systems operate through pulsed high voltage, which can go wrong in a rather dramatic way. a) This high voltage Pockels cell driver started smoking in the laser system due to a flimsy resistor and insufficient cooling. b) The failure was so dramatic that it shattered the Pockels cell crystal, which is transparent to the laser regardless of voltage state (the pulse rejection happens at the polarizer).

The reason I am describing this device in particular is that we use the Pockels cell system in three unique ways in our experiment at AFRL. First, we control the laser repetition rate by changing how many times per second we flip the voltage; we can easily convert our 1 kHz system to a 10 Hz or 0.1 Hz system. Second, we selectively amplify the polarizer-rejected laser pulses (the >99%) to probe the laser-plasma interaction at arbitrary time delay; see Appendix A for a detailed description of this nuanced diagnostic. Third, we use our four Pockels cells, positioned at various points in the main laser chain, to temporally sculpt the laser pulse energy [40]. Our detail to temporal sculpting is unconventional at a high-intensity laser facility and requires more nuanced control of the Pockels cell system than would be needed to just pick out the main pulse and suppress the pre-pulse (the conventional application). As an example of the temporal sculpting technique, we can let through three consecutive
pulses with a first Pockels cell, then cut down the pre-pulses to a greater or lesser extent by how well we align the polarizer and/or crystal angle of a second. Through careful pre-pulse sculpting alone we can make the difference in our experiment between producing an undetectably small number of electrons and flooding our detectors with signal.

2.3 Amplification to high intensity

After Pockels cells pick out pulses, these pulses are optically routed through laser amplifiers that boost the energy of these pulses. In our laboratory, for each amplification stage, a Ti:Sapphire laser crystal is energized into an atomically excited state by dedicated long-pulse lasers. The long-pulse lasers deposit energy into the crystal over many nanoseconds, a time period overlapped and followed by the main short pulse laser which passes through the crystal and extracts the energy in femtoseconds. So as to extract as much energy as possible, multiple passes of the main pulse through the crystal are typical. While the crystal is waiting for its energy to be fully extracted, it continuously emits some light; this nanoseconds-long “Amplified Spontaneous Emission” (ASE) is one source of laser pre-pulse. Laser pre-pulse is a defining element in this dissertation work, and this topic will be picked up again in Section 2.7.

Before I can discuss further details of the amplification process, I need to introduce the concept of laser intensity. Laser intensity is a central characterization in our experiments. Intensity is a quantification of the localized electromagnetic field strength; given the same total energy of light, high-intensity light will rip material apart in a drastically different way than low-intensity light. Therefore, laser intensity is unique from laser energy. An analogy from chemistry is: a large block of ice may have same total thermal energy, but neither the temperature nor properties, of a smaller cap of steam\textsuperscript{10}. Laser intensity is defined as:

\[ I = \frac{E}{At} \]  

(2.2)

where E is the laser energy, A is the area of the laser beam, and t is the time duration of the laser pulse. Intensity for a given laser is not unique; for example, focusing the laser decreases the area of the beam, increasing the intensity. A laser system will typically have a maximum intensity accessible without major re-configuration;

\textsuperscript{10}Another analogy I like to use: A long, gentle push feels quite a bit different than a lightning-fast karate chop.
at the Scarlet laser system, this number is $\sim 10^{22}$ W/cm$^2$; at AFRL, this number is $\sim 10^{18}$ W/cm$^2$.

A high-intensity laser will damage materials (and optical components) that a lower-intensity laser (of the same or greater energy) may not. To avoid optical component damage without sacrificing laser energy, laser intensity can be lowered by increasing beam area and/or pulse duration.

A technique called chirped pulse amplification (CPA) [41] is the current best-and-only method of reaching extreme laser intensities with university-scale laser facilities. As an example, the laser amplification chain at Scarlet is shown in Fig. 2.2. The laser is temporally stretched (duration increased, intensity lowered) before amplification and then temporally re-compressed after amplification (restored to high intensity).\textsuperscript{11} Temporal stretching (increasing pulse duration) lowers intensity without requiring beam expansion; this keeps the size of crystals and mirrors reasonable to manufacture (the largest crystal at Scarlet is three inches in diameter). After the amplification stages, the final beam at Scarlet is stretched to 6-inch diameter to avoid damaging the final optics, while it is expanded to 1-inch diameter at AFRL.

I won’t go into details beyond the basic concept for the chirped-amplification technique, as my dissertation topic is not specifically tied into the laser amplification process. The topic of laser amplification is of interest to this work in three ways: 1) the slowly varying component of laser pre-pulse originates in the amplifiers, 2) terms such as pulse stretching, compressing, etc. are common in discussions of high-intensity laser experiments such as this one, and 3) understanding the multi-step amplification process and limitations helps explain why the highest-intensity lasers are a frontier, and shows why a laser might need a long beam-path and large diameter (hence, taking various amounts of money and floor space).

\textsuperscript{11}Temporal stretching can be achieved in a recoverable (re-compressible) way by through a clever technique called “chirping.” Through geometrical arrangement of optical gratings, optical frequencies of the laser bandwidth are re-arranged such that the “bluer” light lags behind the “redder” light in the laser beam (or vice versa). In the case of sound rather than light waves, this frequency variation through time sounds like a bird chirp. This citation should help an interested reader learn more: [42].
Figure 2.2: **Scarlet laser amplification diagram.** The Scarlet laser chain relies on chirped pulse amplification to reach ultra-high intensities. Intensity is deliberately reduced during the amplification process; the pulse is temporally stretched from 30 fs to hundreds of picoseconds. As it is amplified to greater energy, it is more and more spatially stretched (laser is telescoped to larger beam size) to avoid damaging optics. After amplification, it is even further spatially stretched so it can be temporally compressed back down to 30 fs without damaging optics. At this point it is ultra-high power (400 TW). The pulse is then extremely spatially re-compressed (by focusing the laser) and it becomes ultra-high-intensity \( (>10^{21} \text{ W/cm}^2) \). Graphic: Scarlet laser team.

### 2.4 Vacuum isolation: Ultra-high intensities without that burning smell

At high intensities, air is no longer a passive medium in which the laser propagates; the laser starts strongly interacting with the air. High-intensity lasers will ionize air as they propagate. On the lower intensity end, the laser beam changes the air index of refraction and self-distorts, causing issues later on for laser focusing (or damaging optics with particularly hot spots). At high enough intensities, the laser deposits most of its energy in air and you get your “experiment” happening mid-way through the laser system. Intensity surges as the laser is focused, so focusing the amplified laser in air will always result in ionization. This ionization will steal laser energy,
bend the laser light, and severely modify the laser focus. In other words, air can become a barrier to high laser intensities.

For these reasons, vacuum pressures are maintained at all points in the laser system where the laser is expected to reach high intensity. At Scarlet, the vacuum levels are about $10^{-6}$ Torr. In that system, everything leading up to the pulse compressor and after must be kept under vacuum. A vacuum-tight laser-plasma interaction chamber from Scarlet is shown in Fig. 2.3. At AFRL, since the final laser intensity is a thousand times lower, intensity effects in air are negligible everywhere except near the laser focus. For this reason, only the target chamber at AFRL is held under vacuum; vacuum levels are maintained at 1/40th room pressure, or 20 Torr. This number is chosen as a compromise in that it is low enough to prevent this laser from ionizing the background air but high enough to prevent freezing the flowing water target.

I have detailed the importance of vacuum isolation because of a possible critique of our setup: there are still substantial numbers of molecules (air and water vapor) present in our chamber held at 20 Torr pressure. Is 20 Torr pressure low enough to prevent ionization effects from distorting the laser focus? We checked this empirically by letting the laser focus in our vacuum chamber, with no target blocking the beam path. We used a camera to look at a piece of paper on which the laser was imaged. If significant distortions were present, the laser would look spatially non-uniform after focus. However, the quality was not seen to deteriorate, so we concluded that the 20 Torr pressure is sufficiently low for this experiment and laser intensity.
Figure 2.3: **Original experimental vacuum chamber at Scarlet.** The entire experiment is housed in an aluminum vacuum chamber from which all the air is removed; otherwise the high-intensity laser would ionize air molecules and self-distort. I was involved with building and using many of the laser and plasma diagnostics annotated in this figure. Few of these are used in this dissertation experiment, but if you are interested, please ask sometime for a tour of Scarlet!

### 2.5 Measuring and optimizing laser intensity

As defined in Eq. 2.2, laser intensity is equal to energy per area per time (W/cm\(^2\)). A direct measurement of intensity has not yet been reliably developed for application to ultra-high-intensity lasers. As a result, experimental laser intensities quoted here and in literature are derived from measurements of pulse energy, focal spot area, and pulse duration. However, as will be discussed in this section, these values also are somewhat indirectly measured due to the impossibility of placing standard scientific instruments at the focus of an extreme light source. Because of this, experimental laser intensities typically have at least a factor of two error, although this error is not generally stated. In this section, I will outline how energy, focal spot area, and pulse duration are measured and how each can be experimentally controlled to enhance laser intensity.\(^{12}\)

\(^{12}\)Further measurement details along with the actual intensity calculation for the AFRL laser system can be found in Appendix C.
Energy is measured using a calorimetric energy meter. This detector captures the entire laser in a ceramic material and measures how much heat is generated. Laser energy can be increased through the addition of laser amplifier stages (a long and expensive process) and reducing the energy losses through the system (cleaning mirrors, replacing damaged crystals, better timing of the pump lasers, etc.). An added difficulty for the energy measurement is that the full-energy measurement is performed prior to the final laser mirrors (to avoid the focusing the laser). In this experiment, energy losses due to these mirrors were separately measured and corrections to energy were applied post-hoc. Another experimental difficulty with regards to the intensity calculation is that a realistic laser pulse will not contain all of its energy in the short pulse; pre-pulse and post-pulse on the femtosecond, picosecond and nanosecond scale contributes to the measured energy, but not the focused intensity.

Focal spot area is measured by making an image of the focal spot with a microscope objective and camera, and measuring the area containing the majority of the energy. The focused, full-energy laser would destroy the objective and camera, so filters are added well-before the laser comes to focus. Lowering laser energy through reduced amplification is common practice in the field, but is not preferred as laser amplification can change the focal spot area.

Focal spot area is reduced through use of an off-axis parabolic (OAP) mirror and by reducing optical aberrations. The off-axis paraboloid (OAP) mirror is the final optic in the laser chain before the laser hits the target. Its goal is to take a big laser and focus it down to a very tiny point. One uses a mirror rather than a lens in part because the lens would be damaged (and beam distorted) during high-intensity laser transmission through glass. One uses a OAP mirror rather than a concave spherical mirror (which would be cheaper) because a paraboloid geometrically creates a perfect point for all incoming rays, which means a significantly better focus in practice. The focal spot area can be further decreased by systematically avoiding optical distortions from the laser system at the engineering stage (thermal aberrations, mirror surface defects, etc.), by careful alignment through all optics in the laser system, and by choosing an OAP with a large beam size to focal length ratio (this ratio is called the f/#). A significant modification to the OAP is central to this dissertation. We drilled a hole in the OAP in order to let electrons pass through its center. Adjusting the experiment to maintain small focal spot area despite this egregious deliberate damage was no small feat and involved adaptive optics (Section 2.6).
Pulse duration is measured with a single-shot autocorrelator [43] ("single-shot" as the measurement is complete after a single firing of the laser pulse). The single-shot autocorrelator works by splitting the laser into two arms and overlapping them in a non-linear crystal. Through nonlinear optical effects and clever geometry, the spatial profile of second-harmonic light emitted from the crystal corresponds to the duration of the pulse. Pulse duration can be decreased through adjustment of the alignment and separation of compression gratings. However, there is a limit to pulse duration: this limit is set by the bandwidth of the laser spectrum and by the original pulse duration. As the laser amplifies, its bandwidth narrows, meaning the shortest pulse it can sustain is actually longer than that which exited your oscillator. Autocorrelators are a routine tool of the short-pulse laser physicist; I have built these devices a few times at Scarlet, and autocorrelation measurements contributed to our laser intensity calculation ($1.5 \cdot 10^{18}$ W cm$^{-2}$) for this dissertation.

![Image](image.png)

**Figure 2.4: Scarlet on-shot laser diagnostics table.** At Scarlet, I designed the laser diagnostics table for looking at the properties of the laser in parallel with its ablation of a target. Much like you might take a tissue sample into the lab and perform all sorts of tests to get a sense of your overall tissue health, we take a small section of the laser and do a lot of tests to diagnose beam properties. A small fraction of light passes through a mostly reflective mirror, and we split that bit again many times and perform many diagnostic tests. Graphic: Chris Willis and Scott Feister.
2.6 Adaptive optics: Eyeglasses for the laser

The experimental work of this dissertation would not be possible without a high-intensity laser focus. Aberrations to the laser wavefront as the laser propagates through the imperfect optical system can distort the focus, increasing focal spot area and thereby decreasing the focused laser intensity. Adaptive optics can correct the problem to a limited extent (see diagram of Fig. 2.5). Adaptive optics are mirrors or lenses that can be formed into shapes more complex, custom, and arbitrary than “curved” or “flat.” This allows for corrections to the laser wavefront, which can result in control of the laser focus.

In high-intensity laser experiments, one typically uses adaptive optics to correct for the small aberrations in the wavefront that accumulate from the amplification, stretching, and compressing processes. One can also use adaptive optics to adapt to an arbitrary goal, such as optimizing the amount of radiation output from your experiment. However, installation of adaptive optics comes at the price of relying on them in your system, and the correction is quite limited in range. Correcting wavefronts through better alignment is preferred whenever that is the main source of aberration.

Figure 2.5: Wavefront correction by adaptive optics. Adaptive optics are often used to correct the laser wavefront, in this case for best focus. Graphic: Franki Aymond and Scott Feister.

A deformable mirror has a surface membrane pushed and tugged on by actuators, and can programmed to warp into a desired shape. This adaptive optic has prevalent use in high-intensity laser physics for its potential to flatten wavefronts and therefore
The AFRL deformable mirror has 37 points of actuation, a 1.5 inch membrane diameter, and sits in air; the deformable mirror at Ohio State has 33 actuators, a 4-inch membrane diameter, and sits in vacuum. While the deformable mirror can correct some issues with the wavefront, the limited actuators in the deformable mirror also imprint new errors in the wavefront. For this reason, neither the mirrors at Ohio State nor AFRL are in the beamline for upcoming experiments (the beam quality is good enough without). However, the deformable mirror at AFRL was essential to certain of these dissertation experimental studies (which purposefully involved a very distorted final optic). The deformable mirror was used in two modes: decreasing the vacuum focal spot size, and enhancing an experimental output: an optical harmonic of the laser. I will write about our experimental use of the deformable mirror in Ch. 3.

2.7 Laser contrast: Pre-pulse to main pulse ratio

We might prefer our high intensity pulses to strike the target without any forewarning, but typically some energy is deposited on the target before the main pulse has arrived. This “pre-pulse” energy has a variety of sources. The oscillator creates short pulses every 12 ns, and some of these will become significant pre-pulses if the Pockels-cell system does not sufficiently suppress them. Pre-pulse could also come in the form of a small-percentage reflection of the main pulse anywhere in the laser chain, if this light finds a path to the target that is quicker than that followed by the main pulse. As was discussed in Sec. 2.3, long pre-pulses (nanosecond-long rather than femtosecond-long pulses) are generated by the amplifiers as they naturally emit light from their gain medium between the time when they are pumped and the time the main pulse extracts their energy. As a general metric pre-pulse intensity relative to the main pulse, laser contrast encompasses time-varying short-pulse and long-pulse sources. For this reason, the laser contrast is ambiguous unless cited at a certain point in time relative to the main pulse, (e.g., $1:10^{-6}$ at -3 ps.).

To measure pre-pulse arriving 1 ns to 60 ns before the main pulse, creative misuse of a commonplace laboratory tool is surprisingly effective. The measurement begins by scattering laser light onto a filtered photodiode. An photodiode is a detector that makes a signal proportional to light, which, for example, used to measure pre-pulse in [46]. This photodiode is optically filtered such that the main pulse does not saturate
the photodiode. Diode signal vs. time is visualized on an oscilloscope, and is proportional to pulse energy vs. time. The photodiode and oscilloscope together have $\sim 1$ ns temporal resolution$^{13}$, which means that the 30 fs short pulse is represented as a 1 ns pulse of equal energy. A typical photodiode + oscilloscope combination can show three orders of magnitude between the main pulse and pre-pulses, which is insufficient to identify low-energy pre-pulse features. The next step of this measurement is that optical filters are removed from the photodiode such that the detector saturates. Importantly, the detector has not yet saturated when the pre-pulse hits the detector. Therefore, features of the pre-pulse previously below the detection threshold become visible. This filter removal process is repeated to several degrees of saturation. These photodiodes can be quite robust to saturation and allow for you to see down many orders of magnitude without degradation to non-saturated detector linearity. For nanosecond timescale pre-pulse measurement, this trick effectively increases the dynamic range of the photodiode diagnostic from three orders of magnitude to nine orders of magnitude.

To measure pre-pulse intensity 500 fs to 2 ns prior to the main pulse, a third-order cross-correlator [47] is the usual tool. While I won’t go into great detail of this diagnostic (for more information, see Luan et al. [47]), we used a custom implementation by Kyle Frische at AFRL to establish the picosecond laser pre-pulse for this dissertation work.$^{14}$ The third-order cross-correlator works by splitting the laser into two arms, doubling the frequency of one arm in one non-linear crystal, then interacting the two arms in another non-linear crystal. The amount of resulting ultra-violet light corresponds to the amount of energy at the delay time between the two arms. A pre-pulse will make a different signal than a post-pulse of equal energy in the third-order cross-correlator. This distinction between post-pulse and pre-pulse is important and

$^{13}$While higher temporal resolutions are possible, these devices cost more and are more fragile, negating the effectiveness of the next step of the measurement.

$^{14}$With a separate, custom-built third-order cross-correlator at Scarlet (Fig. 2.6), I diagnosed a major issue with the laser pre-pulse. This was of the larger successful moments in my graduate career, in that it was essential to what followed: tens-of-MeV ion acceleration using novel liquid crystal thin films at Scarlet. For more information on these thin-film experiments, read Poole et al. [48].
the motivation for using third-order cross-correlators. Post-pulses are usually disregarded in the interpretation of HEDP experiments (the high-intensity interaction has already occurred when they arrive on target), whereas pre-pulses are considered significant (they diffuse the target prior to the arrival of the main pulse).

Figure 2.6: **Scarlet’s in-house third-order cross-correlator.** I did major modifications to the Scarlet cross-correlator, which resulted in finding and correcting sources of pre-pulse and increasing Scarlet’s contrast.

I write about the pre-pulse diagnostics because we discovered that the pre-pulse temporal characteristics were the strongest controlling factor in the electron acceleration studied in this dissertation work. Energy of the pre-pulse severely modifies the target with which the main pulse interacts, a reality that has been well-established by prior experimental work. Essentially, we found that our target required precise, last-minute modification by the pre-pulse in order for the main pulse interaction to accelerate electrons. Experimental characterization of the pre-pulse allows both experimental reproducibility and accurate computational modeling of the laser interaction. As I will explain in Ch. 5, I adapt the experimental pre-pulse measurements

15In a second-order autocorrelator, a pre-pulse and post-pulse of equal energy make the exact same signal, which is why building the third-order version is worth the effort when diagnosing pre-pulse conditions.
as an input for simulations dedicated to modeling the target modification; these simulations are distinct from the main pulse interaction simulations. Accurate pre-pulse modeling led to re-interpretation of certain experimental data and changed the result of the main pulse interaction simulations. Experimental manipulation of pre-pulse characteristics and results of such manipulation are shown in Ch. 3.

2.8 Comparison of Scarlet and Red Dragon systems

I learned how to do HEDP research on the Scarlet laser, and I did my dissertation work using the Red Dragon laser. The Scarlet (Ohio State) and Red Dragon (AFRL) laser systems are complementary in a variety of ways; the Red Dragon trades off intensity for repetition-rate and compactness. Table 2.1 compares these two systems using many of the metrics I described in this chapter.

<table>
<thead>
<tr>
<th></th>
<th>Scarlet</th>
<th>Red Dragon</th>
</tr>
</thead>
<tbody>
<tr>
<td>Laboratory location</td>
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<td>AFRL, Dayton</td>
</tr>
<tr>
<td>Approx. cost to build</td>
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<td>$1 mil</td>
</tr>
<tr>
<td>Wavelength</td>
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<td>780 nm ±40 nm</td>
</tr>
<tr>
<td>Pulse rate</td>
<td>1/ min</td>
<td>1 kHz</td>
</tr>
<tr>
<td>Pulse energy</td>
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<td>8 mJ</td>
</tr>
<tr>
<td>Pulse duration</td>
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<td>40 fs</td>
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<tr>
<td>Beam diameter (post-compression)</td>
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<td>1 in.</td>
</tr>
<tr>
<td>Focal spot size</td>
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<td>2 μm</td>
</tr>
<tr>
<td>Peak intensity</td>
<td>$5 \cdot 10^{21}$ W/cm$^2$</td>
<td>$5 \cdot 10^{18}$ W/cm$^2$</td>
</tr>
<tr>
<td>Instantaneous power (averaged over 30 fs)</td>
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<td>0.4 TW</td>
</tr>
<tr>
<td>Average power (averaged over one minute)</td>
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<td>8 W</td>
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<td>Main:pre-pulse contrast (at -5 ps)</td>
<td>1:10$^{-9}$</td>
<td>1:10$^{-6}$</td>
</tr>
</tbody>
</table>

Table 2.1: Comparison of the Scarlet and Red Dragon laser systems.
Chapter 3: Experimental characterization of the radiation

3.1 Initial experimental setup

The experiment utilized the modified Red Dragon laser (KM Labs) at the Air Force Research Laboratory (AFRL) at Wright-Patterson Air Force Base. The Red Dragon is a Ti: sapphire based 1 kHz system producing up to 8 mJ, 42 fs Gaussian full width at half maximum (FWHM) pulses at 780 nm ± 20 nm. The final focusing optic is an aluminum-substrate, gold-coated 30° OAP (effective f/1.3) producing a measured 2.2 μm FWHM focal spot and a peak laser-intensity of $1.5 \times 10^{18}$ W cm$^{-2}$ in vacuum. The target was a 30 μm diameter vertical laminar distilled water column flowing from a 30 μm glass capillary nozzle. The water column flows at 14 m/s and remains laminar for several mm after exiting the nozzle, and its position remains stable to within 2 μm. A background vapor pressure of 20 Torr was maintained to prevent freezing of the flow. The experimental interaction (illustrated in Fig. 1.2) is housed in a 3-inch-thick steel vacuum chamber.

3.2 Dosimetric explorations show directionality

An interesting question is to what extent is the radiation outside the target chamber backwards-going. To answer this question, radiation through the chamber walls was monitored by a radiation survey meter sensitive to X-rays above 25 keV. We placed the dosimeter at various locations about the target chamber and recorded the radiation levels. It was discovered that the radiation dose is unusually high outside the chamber behind the OAP compared to the forward propagation direction (directly opposite side), and all other angular positions, as shown in a polar plot in Fig. 3.1. This was not expected based on prior experience with ultra-intense laser plasma interactions, where the $\vec{J} \times \vec{B}$ force dominates to push electrons forward, resulting in a
Bremsstrahlung radiation peaked in the forward direction. We hypothesized that the back-directed radiation dose could be explained if a significant number of energetic electrons generated during the laser plasma interaction propagated backward, generating Bremsstrahlung radiation in the aluminum OAP, its mount, and the chamber wall. However, the presence of these electrons was at that point still conjecture.

Figure 3.1: **Survey meter dose measurement diagram.** A radiation survey meter (dosimeter) was placed at various positions around the outside of the experimental vacuum chamber; these positions are marked with a gray square and gray circles. At each position, dose was measured. A lead brick within the chamber shields radiation in the leftward direction, which is where experimental data stations sit. Further lead shielding, not shown, exists further from the chamber than the dosimeter was placed, which protects researchers in the building. Despite general symmetry of objects within the chamber between the forward (top) and backward (bottom, gray square) direction, dose is more than four times greater in the backward direction. Since a peak in radiation in this direction was unexpected, this measurement served as a motivation for further study of the source of the radiation source.

The water nozzle can be rastered in X and Y to achieve a map of backward-going dose as a function of water nozzle position (Fig. 3.2). This allows for a variety of target alignments to be explored with a complete overview of the area. Furthermore, it removes error from a prior method where the laser alignment would be centered in
the water column, and it shows that a laser centered on the water column (normal incidence) does indeed create a large amount of backward radiation.

Figure 3.2: **Dose response to varied focal depth and transverse position.** A focal depth and transverse scan of the water column relative to the laser shows features of strong backward X-ray emission both at normal incidence (water jet transverse position = 0 μm) and for glancing incidence on the water column (water jet transverse position = ± 13 μm). In this plot, the X and Z axes are as shown with arrows in Fig. 3.1.
3.3 X-ray characterization indicates high energies

Figure 3.3: X-ray measurement experimental setup. A single-hit X-ray spectrometer was used to characterize the electrons emitted in the direction of back-reflected light. The X-ray collimator and X-ray detector (both described in Appendix B) sit 8.6 m and 10.3 m behind the target.

X-rays initially were expected to be the dominant type of radiation emitted from the water column. While we did not have the diagnostics to detect electrons, we did have advanced abilities to characterize the X-rays, which we eventually hypothesized to be created by an electron beam interacting with the chamber walls. One of these advanced capabilities was spectral characterization of the X-rays, which would tell us indirectly about the energy of the electron radiation source. A single-hit x-ray spectrometer was aligned specifically to look at the target through the back of the OAP. The experimental setup for this study is shown in Fig. 3.3. A full description of the single-hit X-ray diagnostic is found in Appendix B. The measured X-ray spectrum showed no peak; the spectrum at the energy limit of the detector, 800 keV, was still rising. This confirms the presence of high-energy X-rays created by the interaction. The X-ray energy is well in excess of what might be expected from a laser of this energy. In the case of a Bremsstrahlung source, high-energy X-rays indicate high-energy primary electrons.
Looking at the isotropic emissions, one decides that the high energy x-rays are not generated in all directions, but preferentially backwards. This conclusion is in agreement with the dosimetry.

A natural question was: to what extent is this X-ray radiation emitted in all directions, compared with localized to the direction of the back-reflected light? The question is answered by analyzing the X-ray spectrum from a different angle. As might be expected from the dosimetric explorations, this analysis showed that the emitted X-rays strength is much lower in the non-backward direction (Fig. 3.4).

3.4 Pre-pulse discovered to be essential

The laser for the X-ray spectral study had a fairly large nanosecond-timescale pre-pulse; we hypothesized that cleaning up the pre-pulse would result in an even stronger backward electron radiation. The opposite was ultimately true, in that pre-pulse was found to be essential to the radiation source. The laser’s nanosecond pre-pulse was controlled by manipulation of four Pockels cells at successive points in the laser amplification chain. The timing and physical angles of the Pockels cells were manipulated to achieve independent pre-pulse extinction ratios for each of the Pockels cells. For example, a good pre-pulse extinction after the 80 MHz laser oscillator would eliminate inherent femtosecond pre-pulse replicas arriving 12.5 ns, 25 ns, and 37.5 ns prior to the main pulse. A deliberately poor pre-pulse extinction after the main amplifier would allow these short pulses as well as significant long-pulse pre-pulse to
arrive on target. A calculation of the pre-pulse profile required the synthesis of three independent laser diagnostics. These diagnostics are introduced in Ch. 2 and the analysis is described in Appendix C.

Figure 3.5: Specular X-ray spectrum and pre-pulse effect. The X-ray spectrum shows >800 keV high energy X-rays are being generated at some point during or after the laser-plasma interaction. High-energy X-rays are greatly suppressed when pre-pulse is reduced, while low-energy X-rays are enhanced.

When the pre-pulse was cleaned to < 1 : 10^{-10} at 10 ns, the radiation was drastically suppressed. With otherwise the same laser, radiation behind the chamber port dropped from 750 mRem/hr to 150 mRem/hr with the cleaner pre-pulse. Additionally, low-energy components of the X-ray spectrum were increased, and high-energy components of the X-ray spectrum were suppressed. The importance of pre-pulse was mirrored in our first simulations of the laser-matter interaction. These simulations modeled only the main pulse interaction; the target was modeled either with or without a pre-plasma. Neglecting to include a pre-plasma in the simulation resulted in a complete loss of electron acceleration. I will go into much more detail of simulations of this electron acceleration process in Ch. 5.

The importance of pre-pulse led to the desire for new pre-plasma diagnostics. The shadowgraphy/interferometry diagnostic allows for observation of pre-plasma spatial distribution anywhere from tens-of-nanoseconds before the main pulse arrival to 200 fs before it arrives. The diagnostic can also be used to watch the target explosion both before and after the main pulse has interacted (Fig. 3.6). For example, the target is seen to recover fast enough to deal with the high-repetition rate of our experiment.
Probe beam shadowgraphy indicates that material disturbed by interaction with the 1 kHz laser fully exits the interaction region within $\sim 30 \mu s$ [49], and so the experiment can be run continuously at 1 kHz. The details of this diagnostic are quite extensive, and I have given this diagnostic its own section in Appendix A.

Figure 3.6: **Shadowgraphic target evolution.** When the laser strikes a column of water, the water column releases a burst of electrons and then explodes. The evolution of the pre-plasma, the target explosion, and the target recovery can be visualized using shadowgraphy. The above frames show the target explosion and recovery at various times after the main pulse arrival ($t = +0 \text{ fs}$). Due to optical refraction through the cylinder, the right and left edges of the single water column appear as two dark vertical bands. The time step is labeled upper left in each frame. (For more details of shadowgraphy and interferometry, see Appendix A.)

### 3.5 Primary source characterized as electrons

While simulations performed concurrently to the X-ray characterization (see Ch. 5) showed electron beam-like emission from $10^{18} \text{ W cm}^{-2}$ laser/water jet interaction, this distinction had not yet been shown experimentally. The laser interaction was clearly producing large numbers of energetic X-rays, but these X-rays could either be 1) primarily emitted from the target itself or 2) emitted secondarily from the vacuum chamber walls and optical elements. If primarily energetic electrons were being emitted from the target, this would indicate an efficient electron acceleration mechanism with normal laser incidence. This turned out to be the best description; I will describe how we came to this conclusion.
We tested the question of whether electrons were a primary radiation source by putting a magnetic deflection diagnostic into the chamber and imaging radiation coming off the target. A 1 inch diameter Lanex screen [50] was positioned behind the OAP to directly observe the backward-propagating electrons. This setup is shown in Fig. 3.7. A 25 μm thick aluminum foil placed in front of the Lanex renders the setup light tight and also acts as low energy electron filter (blocks most $\lesssim 50$ keV electrons). When energetic electrons hit the Lanex screen, light is emitted in the visible spectrum. This light was imaged onto a 12-bit CCD camera outside the vacuum chamber. Fig. 3.7 shows phosphor images of backward propagating electrons partially obscured by the OAP hitting the Lanex detector, and also shows the same view when a single rare earth magnet (0.16 Tesla surface field) was placed just above the OAP. The direction of deflection in the presence of this magnetic field confirms that the fluorescence is due to negatively charged electrons. Electrons are deflected and dispersed on the Lanex screen, with the OAP shadow moving $\sim 12$ mm. Note that the distance between the OAP and the water jet target is only $\sim 27$ mm, making it difficult to place a pair of magnets with a yoke in this space.

Since the backward-directed radiation source is deflected in a manner consistent with electrons and transmits through an aluminum filter blocking $\gtrsim 50$ keV electrons, we concluded the presence of backward-propagating energetic electrons originating from the laser-plasma interaction.
Figure 3.7: Magnet deflection test for electron radiation. The insertion of a magnet confirms that these particles are in fact electrons. Left: Energetic electrons ejected from the water jet are incident on a fluorescent “Lanex” screen [50]. A camera in a light-tight housing images the optical light produced by the screen. Measurements were made with and without a rare earth magnet (\( \sim 0.16 \) T surface field). Right: False-color visible-light images of fluorescent emission from a Lanex screen due to energetic electrons arriving from the target region. The edge of the Lanex screen is shown by a dash-dotted white line. Light recorded outside of this is due to scatter from the beam tube. Upper right panel presents results without a rare earth magnet, showing a beam-like feature on the screen and a distinct shadow created by the off-axis parabola (OAP). Lower right panel presents results when a rare earth magnet is added. In this panel the OAP shadow moves from its original position (dashed white line) to significantly further to the right (dotted white line), validating the hypothesis that the fluorescence is due to energetic electrons.

3.6 High laser-to-electron energy efficiency demonstrated

To measure the number of backward-propagating electrons, the gold-coated, aluminum-substrate OAP was electrically isolated and used as a Faraday cup to collect electric charge (Fig. 3.8). The collected charge was measured as an average current on a Keithley Instruments 610C solid-state electrometer, and this measurement was interpreted as charge per pulse after dividing by the number of laser-target interactions per second (1000). This method of obtaining charge per pulse was corroborated on a single-shot basis by analysis of the fast voltage trace observed using
a 100 MHz oscilloscope in place of the electrometer. This measurement showed that 600 pC of electrons per laser pulse were measured to be accelerated into the OAP. This result includes electrons of all energies.

Figure 3.8: Faraday cup diagram. We measured the current flowing out of the electrically isolated OAP as a metric for the number of electrons accelerated. By inserting glass between the interaction and the Faraday cup, electrons accelerated from the interaction will be blocked or scattered. Low-energy electrons will be entirely blocked or deflected by the glass, and the current still flowing through the OAP is due to higher-energy electrons that pass through the glass. This test, along with others described in this chapter, showed that there were hundreds of pC of high energy particles being accelerated.

To determine the contribution of highly energetic electrons to the measured charge, a 100 μm glass cover slip can be inserted into the laser beam path to filter out the majority of backward-going <120 keV electrons and <3.4 MeV protons. The cover slip attenuates the laser by ~15% and slightly degrades the focal spot quality, reducing the laser intensity to ~ 7 × 10^{17} W/cm². 300 pC of electrons per laser pulse, >120 keV, were measured to be accelerated into the OAP.

Since the result is surprising (and one basis, supported by other evidence, for efficiency claims presented throughout this dissertation), our interpretation of electron number per pulse was verified in two ways. First, the effect of the cover slip, 20 Torr
vacuum, and electron scattering out of the the OAP was modeled in MCNP particle-scattering software. The cover slip and OAP were modeled in MCNP with realistic geometry and materials, encased in 20 Torr vacuum, and an electron beam of various energies was allowed to propagate from the target to the OAP. It was observed that the effect of electron scattering in 20 Torr vacuum, scattering within the cover slip, and scattering out of the OAP changed the 300 pC of electrons per pulse measurement by less than 15%, and that electrons below 120 keV were effectively blocked as expected. Second, the current measurement was checked using independent electrometers and oscilloscopes. For the electrometer, the current was measured over a few seconds, and divided by the number of laser pulses in that interval. For the oscilloscope, the voltage spike for single shots was measured and the current calculated by integrating the voltage and accounting for the terminating resistance. In both cases, the number of 300 pC of electrons per pulse was reached.

A lower bound on conversion efficiency can be calculated by assuming that all electrons above 120 keV are equal to 120 keV. This calculation results in a 1.2% conversion efficiency of laser energy into high-energy electrons, a number which will be quoted throughout the text. To be clear, this is a lower bound; if the average electron energy \( >120 \) keV is 500 keV, the conversion efficiency into high-energy electrons would be 5%. However, the experimental evidence provided in this section assures a lower bound of 1.2%, and so we can confidently say that this acceleration mechanism has >1% efficiency into \( >120 \) keV electrons in the specular direction.

### 3.7 Experimental adaptations for measurement of electron energy spectrum

Since there was evidence of an novel and efficient means of accelerating electrons at normal laser incidence, it was important to characterize the result. One important element of this was answering the question of the energy of the backward-emitted electrons. It was necessary to design and characterize an electron spectrometer for this experiment. We chose to design this spectrometer for high-acquisition rate operation; it can measure 100 spectra per second. This is in contrast with typical electron spectrometers in our field, which are designed for few-times-per-day measurements. The detector used a magnet to deflect electrons according to their energy, and a Lanex scintillating screen coupled to a linear CCD for high sensitivity. There were several creative elements to this electron spectrometer; while they are quite interesting, they
are not essential to the experimental narrative. I discuss further details of the electron spectrometer design as well as its absolute charge calibration at a linear accelerator at the University of Notre Dame in Appendix D. The electron spectrometer was eventually fielded in the experiment and showed greater-than-MeV electrons produced, which is in line with our predictions from X-ray measurements but exceeding expectations from prior literature. However, this measurement could only be made after several significant modifications to the laser system, which I will describe next.

![Diagram of experimental setup](image)

Figure 3.9: **Experimental setup for electron spectrum measurement.** Drilling a hole in the OAP was necessary to conduct this experiment. To measure the electron spectrum in the direction anti-parallel to the laser axis (back-reflection direction), a 3 mm hole was drilled into the 25.4 mm diameter 30° gold-coated f/1.1 OAP. Electrons produced by intense laser interactions with a flowing water target pass through the hole in a 6.3° full-angle cone. Electron energies are characterized by a magnetic spectrometer located 127 mm behind the target. A wire is connected to the back of the electrically isolated aluminum-substrate OAP to use it as a Faraday cup, allowing for a measurement of electron number per pulse. The laser is polarized in the plane of the diagram.

We wanted to measure a spectrum directly behind the off-axis paraboloid (OAP) mirror, but even fairly energetic electrons will be scattered within the thick Aluminum substrate of the OAP. This caused a serious problem: a measurement off to the side
of the OAP would not speak to the novel nature of this mechanism, which succeeded with a normal-incidence laser interaction (producing electrons that may have had particularly high energies in the backward-going direction). To take the measurement at 0°, then, we decided to drill a hole in the center of the OAP. Electrons emitted directly backward towards the OAP could pass through the hole and be characterized by an electron spectrometer, which was installed behind the OAP. (See Fig. 3.9.) Although this hole in the OAP was necessary in order to highlight the interesting features in the experiment, the hole in the OAP not only reduces the laser energy, but reduces the focusability of the beam (through the physics of diffraction). Worse, the hole-drilling process warps the OAP and makes it a less-refined mirror surface. To mitigate these issues, we used adaptive optics (see Ch. 2); software was developed to guide a deformable mirror towards mitigating some of the newly introduced issues.

A deformable mirror moves its motors to change its membrane and adapt the beam wavefront. We have written custom algorithms to use the deformable mirror to enhance a variety of output conditions, such as laser intensity. To maintain flexibility in accommodating multiple devices as inputs and outputs to the adaptive routine, we implemented a genetic algorithm ourselves in LabVIEW. A genetic algorithm [51] is one in which a wide parameter space is explored (in this case, the myriad deformations possible with the deformable mirror) by randomly changing in a way that mimics nature. The quality of a given mirror deformation is characterized through a “fitness function.” In our first implementation, the fitness function is a quantifier of how intense the laser spot has become with the given “genes” (specific deformable mirror positions). Adjustments that lead to better outcomes are kept, while those that lead to worse outcomes are discarded. A new generation of deformations is made by making random perturbations to the best deformable mirror positions of the current generation. By repeating this process over several generations, a genetic algorithm enables evolution towards an outcome (e.g. a very intense laser spot). To optimize the focal spot in our specific genetic algorithm implementation, the following process is executed. First, the focal spot camera and parabola are aligned to make the most intense focus when the deformable mirror set to its flattest position. This is done coarsely by assessing the focal spot spark in air, and then finely by small iterations while looking at the focal spot camera feedback. Second, the deformable mirror explores a variety of random fluctuations to its deformation, and for each fluctuation the following fitness function is assigned using the camera’s image of the focal spot:
In the above fitness function, $I_{\text{spot}}$ is the integral of pixel values within the ‘spot’ radius (defined by the FWHM of a Gaussian fit to the image), $A_{\text{spot}}$ is the area of the spot, and $I_{\text{image}}$ is the integral of pixel values in the image. The operation of the genetic algorithm is illustrated in Fig. 3.10 has been shown to converge to a small focal spot.

Figure 3.10: Genetic algorithm feedback loop. Adaptive optics allowed restoration of the laser focal spot energy and permitted this experiment. To left: Diagram of the focal spot measurement apparatus. The deformable mirror is located two turning mirrors prior to the OAP. To right: The focal spot measured after the ‘best focus’ deformable mirror optimization is quite small, enabling high intensity and indicating the genetic algorithm was successful.

3.8 Natural fluctuations of electron number trend with $3\frac{3}{5}\omega$ harmonic

Optical harmonics backscattered from the incident laser can give an insight into the fast timescale plasma conditions. The laser wavelength is centered at $\lambda = 780$ nm
(near-infrared), and its angular frequency is calculated as $\omega = 2\pi/\lambda$. The laser half-harmonic is defined as $\omega/2$ (mid-infrared, outside the visible range), the doubled harmonic is $2\omega$ (blue), and the three-halves harmonic is $\frac{3}{2}\omega$ (green). The visible backscatter optical spectrum is measured using an Ocean Optics USB4000 spectrometer. The light reaching the spectrometer passes through the dielectric coating (TLMB) of the final mirror prior to the OAP. Because of the TLMB coating, wavelengths in the $>700$ nm range are greatly attenuated in transmission. This attenuation is advantageous for blocking the $\omega$ light while passing the $\frac{3}{2}\omega$ and $2\omega$ light. This allows one to look specifically at the optical harmonics, without saturating the detector with the $\omega$ laser reflection. With this spectrometer diagnostic, optical harmonics of the laser (especially the $\frac{3}{2}\omega$ harmonic) are clearly seen concurrent to high-efficiency electron acceleration. The harmonics detected with the optical spectrometer are highlighted in Fig. 3.11a.

\[16\] The optical spectrometer is triggered such that 3 or 4 pulses will be caught per trigger when operating the laser at 1 kHz, and 1 pulse will be caught per trigger when the laser is operating at 250 Hz. Based on conversations with the manufacturer, this is an unfortunate limitation of the USB4000, which requires 3.8 ms of additional delay before opening the electronic shutter if one tries to reduce exposure time below 3.8 ms. The result is that, for single-shot acquisition studies of optical harmonics, the laser was operated at 250 Hz rather than 1 kHz.
Figure 3.11: **Backscattered optical harmonics.** a) Backscattered light shows a strong green harmonic ($\frac{3}{2}\omega$) signal when observed with an optical spectrometer. b) A color image of the OAP mirror surface, which has a gold coating that reflects visible light including the blue (2$\omega$) and green ($\frac{3}{2}\omega$) harmonics. One sees spatial features in the harmonics, specifically, a patchiness in the green light (likely due to plasma inhomogeneities). c) For reference when viewing panel b, this is a wide-view photo of the OAP in its translation mount. The hole in the center of the OAP is bright rather than dark due to incident laser scattering at its edge.

A qualitative backscattered light image is created with a color CCD camera. The camera “looks” at the surface of the parabola, through the back of a TLMB-coated mirror. To achieve this imaging, the parabola is brought into focus prior to any interaction. Then, an ND3 filter is added and the shutter is reduced. This allows one to see the imaged OAP surface while excluding room lights and the majority of infrared laser light (which is reflected by the TLMB-coated mirror). Fig. 3.11b shows an example backscattered light image, with Fig. 3.11c as a reference for the chamber setup.

By analyzing the single-shot data of 37 seconds of data acquisition (370 single shot data points), one can see that the backscattered green light is correlated with the integral of the electron spectrum (Fig. 3.12). This corroborates first-hand the experimenters’ intuition about these two values, and also the success in enhanced electron generation when the ‘optimize green backscatter’ fitness function (Eq. 3.2) was used to tune the deformable mirror.
Figure 3.12: **Statistical correlations between $\frac{3}{2}\omega$ and electron number.** Unintended shot-to-shot variations in the experiment can provide additional insight into the backward-going-electron-acceleration phenomenon. For example, the experimenters note that the backscattered optical spectrum and the electron spectrum fluctuate on a shot-to-shot basis. One may wonder if the fluctuations are correlated. The graph above plots elements of these two experimental outputs: the integral of the electron spectrometer signal against the integral of backscattered $\frac{3}{2}\omega$ light (green, 500 to 550 nm). From this graph of 370 laser shots (37 seconds of data acquisition), one sees that the number of electrons at the electron spectrometer coarsely increases with the quantity of green backscattered light produced.

### 3.9 Deliberate optimization of $\frac{3}{2}\omega$ boosts electron number

Since analysis of correlations due to natural fluctuations in the laser-plasma interaction showed green light trended with electron number, we decided to try and force green light and see if electron number would increase. The genetic algorithm can be configured to optimize the backscattered green light rather than the laser focal spot. In this case, a Semrock green-pass filter is added to the backscatter color camera, and the following fitness is assessed on the green channel:

$$Fit_{\text{backscat}} = \frac{I_{\text{spot}}}{A_{\text{spot}}}$$  \hspace{1cm} (3.2)

$I_{\text{spot}}$ and $A_{\text{spot}}$ are assessed identically to in the case of the focal spot camera optimization, and the normalization factor $I_{\text{image}}$ is removed because the total amount of
backscattered light in an image is expected to increase rather than remain fixed as the algorithm progresses.

We found that optimizing the green light using this algorithm resulted in a marked increase in electron radiation. The data, however, was taken on a day for which the “pre-optimized” laser focus was not good. An example of the color image which was operated upon by the genetic algorithm is shown in Fig. 3.11b.

3.10 Stimulated Raman Scattering suspected, $\omega/2$ light measured

Since $\frac{3}{2}\omega$ was established to be important to the experiment, it was important to verify the presence of the $\omega/2$ plasma waves from which it is created. The near-infrared (NIR) spectrum of backscattered light is measured using an Ocean Optics NIR spectrometer. Integration is set to record a small number of shots per data point. This data is recorded on a separate computer, and therefore a single spectrum cannot be created between the two spectrometers for a single shot while operating the triggering at 1 kHz.

Half-omega light is seen to be present (Fig. 3.13). Blue-shifting of light as it exits the plasma is strongly indicated. Large shot-to-shot fluctuations in blue-shifting are seen in the data (Fig. 3.13); however, statistical correlations could not be completed in this case due to the equipment limitations. Together with the presence of $\frac{3}{2}\omega$, this suggests Stimulated Raman Scattering (SRS) is at play in the experiment. The strong connection of $\frac{3}{2}\omega$ with electron charge either tells us that similar plasma conditions (pre-plasma extent) are required for electron acceleration and SRS, or that the SRS is intimately tied into the electron acceleration mechanism.
Figure 3.13: **Backscattered ω/2 optical harmonics.** a) A strong, blue-shifted back-reflected ω/2 signal is measured in the experiment. b) Each of the 100 shots averaged together for Fig. 3.13a is plotted individually. While individual traces are completely obscured in this plot, one can see that broad fluctuations in blue-shifting exist between these nominally identical experimental interactions.

### 3.11 Electron energy spectrum measured

In this section, I present direct experimental evidence of MeV electron generation, as obtained through our implementation of the electron spectrometer. The characterization of energy spectrum of electrons, through the hole in the OAP, is one of the principal scientific results of this dissertation. Raw data from 194 electron spectra measurements is shown in Fig. 3.14; a complete analysis of the data is shown in Fig. 3.16. The experimental electron-spectra of this section are compared with electron spectra from simulations later in this paper, in Fig. 5.5.
Figure 3.14: **194 raw electron-spectra.** Raw data of 194 consecutive spectral acquisitions are displayed. Experimental electron spectra measurements are made through the hole in the off-axis paraboloid. Each horizontal streak represents the spectrum from a single CCD exposure (10 ms for 2.9 mJ and 2.3 mJ laser energies, 100 ms for 1.4 mJ). On-target laser energy was reduced from 2.9 mJ to 1.4 mJ in three steps by rotating a variable waveplate which is followed by a polarizer. This reduces the energy of both the main pulse and pre-pulse (which are contained in the same beam). Large shot-to-shot variations in the electron spectra are evident. These traces are statistically analyzed in Fig. 3.16.

A first correction to the raw electron-spectrum is conversion to electron number at the detector. The raw electron-spectra data (CCD counts) is converted into a proper electron-spectrum (pC/MeV/s.r.) by applying an absolute energy calibration. Appendix D outlines the calibration of the high-acquisition rate electron spectrometer designed and built for this experiment. This absolute calibration of detector sensitivity was obtained through a combination of energy deposition modeling (MCNP) and experimental calibration at an MeV linear accelerator at the University of Notre Dame.
Figure 3.15: **Geant4 modeling of electron scattering in 20 Torr.** Low-energy electrons will be scattered by background air and water vapor molecules as they travel from the target to our detector. This effect will skew the measured electron spectrum towards higher energies. A Geant4 model of our experimental setup calculates, as a function of electron energy, the percentage of electrons that will miss the electron-spectrometer slit due to scattering. This correction is applied to experimentally measured spectra to enable a better estimate of the electrons leaving the target. (Graphic adapted from Abraham Handler, who performed these calculations.)

A second correction to the electron spectrum is obtained by considering the effect of electron scattering between the target and detector. The electrons may scatter in the pre-plasma or in the background 20 Torr water vapor. We consider scattering within the pre-plasma to be part of the interaction under study, and so we apply a numerical correction only for the effect of scattering in 20 Torr. The calculation was performed in the particle-scattering code Geant4, and showed that low-energy electrons will tend to be preferentially scattered into a wider beam, increasing the number of these particles originally ejected within the slit’s solid-angle but then missing the slit (Fig. 3.15).
Figure 3.16: Experimental electron spectra at three intensities. a) A statistical analysis (mean and standard deviation) of experimental electron-spectra measurements of Fig. 3.14. For each laser energy, a mean electron spectrum (solid line) is bracketed with the standard deviation (1σ range, filled area). To show the spectrum closest to that which leaves the plasma, each spectrum is adjusted for detector sensitivity and electron scattering in 20 Torr. b) The portion of the electron beam that passes through the hole of the OAP is imaged with a Lanex phosphor camera [52]. The black rectangle denotes the part of the beam (1 mm x 3.2 mm) that enters the magnetic spectrometer’s slit and is energy-resolved. In this image, a diagnostic 250 μm thick stainless steel mesh sits in front of the camera; both camera and mesh are removed before electron spectra measurements.

There are several important features of this result that require explanation. Wide variations between consecutive experimental electron-spectra of Fig. 3.14 indicate fluctuations in the laser-plasma interaction occurring while the experimental apparatus is nominally unchanged. This is in part due to small, sometimes unmeasurable changes in the experimental inputs (such as pre-pulse energy). These natural fluctuations can be recorded, and correlations between the fluctuations of experimental outputs can lead to new insights of the underlying plasma dynamics (recall Sec. 3.8). It is important to highlight that the characteristics of these variations would not be apparent at a low-repetition-rate laser facility. The number of acquisitions shown in Fig. 3.14 would represent several months of dedicated laser time for a facility operating at a few shots per day. In this experiment, the data acquisition (including reducing
the laser energy in three steps) took only a few minutes. Only a high-repetition-rate experiment such as ours can enable statistical analysis of the data such as that shown in Fig. 3.16b.

We have seen no spatial variations in Lanex images of the electrons passing through the hole in the OAP, except those due to the radially symmetric electron dispersion in 20 Torr vacuum. The uniformity of the portion of the beam passing through the hole in the OAP (Fig. 3.16b) suggests a uniform backward-electron-spray at the scale of the hole in the OAP. If we make an educated guess that spatial variations are limited throughout the full electron beam and assume a uniform backward spray covering the solid angle of the OAP, the electron counts per steradian for laser energy of 3 mJ is consistent with the 300 pC/pulse above 120 keV measured in Sec. 3.6.

Fig. 3.16 shows that as laser energy is decreased in three steps, electron number is reduced for all energies. The reduction in electron number is apparent for high electron energies (∼MeV). When the laser energy is reduced to below 1.4 mJ, the electron spectrum is confined to low energies; when the intensity increases to above 2.3 mJ, the spectrum shows much higher energy electrons. Further, as indicated by the change in signal, the total number of electrons emitted increases dramatically. This increase in total yield is consistent with the total charge-emitted measurements discussed above. A large part of the variation in electron spectra between main-pulse laser-energies may be attributable to the proportional reduction in pre-pulse energy, as will be discussed in Chs. 5 and 6.

3.12 Summary of experimental results

The measured spectra of Fig. 3.16 clearly shows electron acceleration to >1 MeV for a laser with an equivalent ponderomotive energy scale [22] of only ∼90 keV. In the spectrum of Fig. 3.16, electrons were measured backward along the laser axis with an apparent peak number density between 500 keV and 1.0 MeV. To our knowledge this kind of super-ponderomotive electron-acceleration in the back-reflection direction has not been previously reported. We note that these spectra could only be obtained by the adjusting of nanosecond contrast parameters to give a pre-plasma of appropriate extent.

The electron number per pulse measured on the OAP/Faraday cup and associated with these high energy spectra is very large: 600 pC/pulse. This Faraday cup measurement is integrated over all energies, and over a 26° emission angle. However,
we have previously shown [53] through the insertion of a glass slide filter that at least half of the charge per pulse exceeds $\sim 120$ keV. Given the 2.9 mJ of on-target laser energy, we have produced $\geq 100$ pC/mJ in association with a MeV-peaked electron spectrum; a surprisingly large charge per laser mJ when compared with measurements of electrons at oblique angles (e.g., 2.3 pC/mJ reported in [18], 0.4 pC/mJ in [23], and 0.15 pC/mJ in [54], albeit all with significantly smaller solid-angle electron beams). The LSP simulation predicts 100 pC of $\gtrsim 150$ keV electrons to be measured in the solid angle of our Faraday cup, which is within a factor of three of the experimental value. When the laser energy is reduced, the electron spectrum peak is significantly lower and the total signal is drastically reduced. This shift is highlighted in Figs. 3.16 and 3.14. Given an energy reduction from 2.9 mJ on-target to 1.4 mJ on-target, the peak of the electron energy spectrum shifted down towards 500 keV.

Leveraging the kHz repetition rate of the experiment, analysis of shot-to-shot laser fluctuations shows positive correlation between the $3\frac{3}{2}\omega$ plasma harmonic and electron acceleration. The correlation is confirmed in that deliberate optimization of the $3\frac{3}{2}\omega$ using a deformable mirror and genetic algorithm also increases electron number. The presence of blue-shifted $\omega/2$ light is verified in the experiment concurrently with electron acceleration. Experimentally, this correlation could be due to pre-pulse conditions, or it could be that the energetic electrons originate in an SRS process. I attempt to tease out this distinction through simulations in Ch. 5.
Chapter 4: Physical mechanisms of electron acceleration

Executing an experiment is only half of the story; the other half is physical interpretation. In this chapter, I give a background in theory and literature helpful in developing a physical interpretation of our experimental electron acceleration. I begin by describing prior experiments in HEDP that show features resembling ours. Above all else, these experiments demonstrate that there is not a unique, obvious way in which electrons might be accelerated under our experimental conditions; there are several ways. The chapter continues with targeted points of physics theory regarding electron acceleration in laser-plasma interactions. The theory topics move from physically simple (one electron in a uniform electric field) to physically complex (the interplay of a laser and collective plasma-dynamics). Each element of theory presents another way in which electrons can be accelerated in the sometimes chaotic dynamics of laser-plasma interactions. I follow this with a brief discussion motivating the use of computational simulations to gain complementary insight into how each theoretically sound mechanism may or may not contribute to the overall electron acceleration in our experiment.

4.1 Related literature and experiments suggest multiple interpretations

Electromagnetic standing-waves are one possible source of low-energy electrons in our experiment, which could be further accelerated by the back-reflected laser pulse. Yu et al. [55] hypothesizes that very high-energy electrons can be created in the backward going direction at normal incidence through a standing wave mechanism. This paper posits however, with 1D PIC as backup, that the propagating wave trumps the standing wave as an acceleration mechanism in the very-low-density region of the pre-plasma. It attributes the high-energy electrons to large initial momentum
through return current, coupled with the back-reflected light’s relativistic ponderomotive acceleration. Orban et al. [52] shows that under conditions matching the AFRL experiment, the standing wave mechanism coupled with direct laser acceleration from reflected light can explain high energy electron acceleration and depends on the presence of a pre-pulse. Kemp et al. [56] explores how the standing wave mechanism can play a role in super-thermal electron distributions observed in forward-going electron acceleration, and shows how this mechanism becomes less uniform and more stochastic in interactions with structured targets. While our targets are not deliberately structured, this suggests that perturbations in the water and inhomogeneities imprinted by the pre-pulse might be significant to the experimental process.

Wang et al. [57] connects the generation of super-thermal, MeV electrons in high-intensity laser interaction (in the forward direction) with the presence of plasma filaments. For this experiment, the target is a gas jet, and electrons are accelerated using the filaments in the forward direction. The paper claims that filaments are the primary source of high energy acceleration. We have not performed any experimental studies of whether filament formation occurs on the surface of our target, so we cannot rule out experimentally that filaments could play some role in efficient electron acceleration. For example, we have casually observed filament-like streaks occasionally in experimental shadowgraphy. Zhavoronkov et al. [58] explains hard X-ray generation through electron emission in bunches; a forward-backward motion of the critical surface sends out jets of electrons perpendicular to the laser incidence direction. Their setup is quite similar to ours: 5 mJ, 30 fs, 795 nm, 1 kHz laser focused to 6 microns onto a 20 μm water column. They do operate under significantly higher vacuum (10^{-2} Torr compared with our 20 Torr). Thin jets of electrons are formed in sub-femtosecond bunches. These electron jets collide with a secondary metal target to create Bremsstrahlung and characteristic X-rays.

Kmetec et al. [59] is a significant paper in understanding our results with respect to observed backscattered harmonics, in that it matches several features of the current work. Especially interesting is the following quote:

The scattered light from the plasma qualitatively correlates with the x-ray production. Under poor focusing, the visible scattered light is mostly blue, measured to be the second harmonic of the incident laser. Improved focusing creates green, corresponding to the 3/2 harmonic of the laser...
The hard x rays become detectable approximately when the green scattered light is visible, and continue to increase as the focusing improves. (Article page 2 / Journal page 1528)

The experiment matches our experience in the laboratory quite well:

1. Hard x-ray signal correlates to the $\frac{3}{2}\omega$ harmonic
2. MeV X-rays are produced by a mJ-class laser
3. Very high conversion-efficiency of laser energy into energetic X-rays (0.3% of laser energy into >20 keV X-rays)
4. Similar experimental setup: chamber held at 20 Torr, 10 mJ laser Ti:sapphire laser (120 fs), focused to $10^{18}$ W cm$^{-2}$; diamond-turned (5 cm) OAP, 3 micron diameter focus.
5. Necessity of pre-pulse for efficient X-ray production
6. High shot-to-shot fluctuations in X-ray yield (40% root-mean-squared fluctuation)

However, it is distinguished from the current work in the following ways:

1. They do not attempt to measure electrons, and they believe their x-ray generation occurs isotropically within the target. We have shown electrons to be the primary radiation source, and the X-rays are not emitted isotropically but rather backward-directed.
2. While our experimental laser operates at normal incidence, and theirs is at 30-degree incidence and $p$-polarized.
3. Our experience is that the X-ray signal is very sensitive to pre-pulse condition; their experience is the opposite.
4. They do not see the X-ray spectrum change strongly as they change laser energy from 10 mJ to 40 mJ; we see a strong variation in the electron spectrum when we change from 1.5 mJ to 3 mJ.
As the laser is aligned for optimal focus in their experiment (by translating the laser focus relative to target), so too does the pre-plasma scale length. This effect is not controlled for in their experiment. Since they do see their hard x-ray signal drastically increase when the laser focal depth is adjusted, the confluence of the increased main-pulse and pre-pulse intensity may be working together to enhance X-ray hits. This calls into their question of their claim of low sensitivity to pre-pulse.

Baffigi et al. [60] supports the effect of a correlation between X-ray production and $\frac{3}{2}\omega$ dependence, with intensity and pre-plasma scale length as the varied parameters (Fig. 2 of that paper). Tarasevitch et al. [61] varies pre-pulse and shows a steep dependence of the $\frac{3}{2}\omega$ harmonic; they find the $\frac{3}{2}\omega$ signal turns on abruptly with a pre-pulse delay of 25 ps, and flattens for longer delays out to 60 ps (Fig. 3 of that paper). This is mirrored by Veisz et al. [62], which attempts to explain the “blue, then green, then white” observation of Kmetec et al. [59]. The paper describes theoretically, giving caveats and context, how $\frac{3}{2}\omega$ light can be generated in short-pulse, moderate pre-plasma scale length interactions. Fig. 4 from that paper shows a “sweet spot” for pre-plasma scale length (at $\frac{L}{\lambda} = 2.5$ at $\frac{n_c}{4}$) with regard to creating the $\frac{3}{2}\omega$ harmonic. In our experiment, many pre-plasma conditions have been explored through pre-pulse shaping with the Pockels cells. The pre-pulse “sweet spot” that creates the highest number of accelerated electrons for us also produces the most harmonic.

As is clear from the literature, there are several means for electrons to be accelerated in a plasma under similar conditions to our experiment. A consideration of how each of these mechanisms plays a role in our experiment would be incomplete without going into some physical detail on the various known electron-acceleration mechanisms; this is what I will do now.

4.2 Electrons ripped from the target and accelerated

Before going into the details of electron acceleration, I’d like to answer a frequent question: Where do we get the electrons that are accelerated in this experiment? Is the laser making them? In our experiments, we are not actually creating electrons, we are merely accelerating them.\textsuperscript{17} The source for our electrons is the water-column target, which, like all of us, is filled with electrons. The target’s electrons are liberated from their atoms in the ultra-intense laser interaction, during which molecular H\textsubscript{2}O

\textsuperscript{17}We aren’t creating them, yet, that is. Lasers will soon reach the in-vacuum intensity necessary to create electron-positron pairs directly from light energy (through $E = mc^2$) [63].
falls apart, leaving hydrogen ions, oxygen ions, and free electrons. At this point, the local volume of liquid becomes a plasma. If the pre-pulse is intense enough (in our case, it is) [64], this ionization will happen during the pre-pulse interaction rather than the main pulse interaction.

Take a pause, and let’s imagine: A giant asteroid is heading directly towards Earth. The radio informs us that this asteroid is, fortunately, several thousand years away from impact. That is really good, because it takes time to move heavy things. It takes a much smaller rocket to deflect the asteroid over a course of a thousand years than, say, over several months. In the world of laser-matter interactions, ions are really heavy things. Photons are massless, molecules are ancient history (as per the previous paragraph), and electrons are two thousand times lighter than even a single-proton ion.

If the ion is high-intensity-laser physics’ asteroid, the laser pre-pulse is its puny rocket. The pre-pulse actually manages to change the entire experiment because it has a lot of time (nanoseconds) to move ions and re-arrange the target. The laser heats the electrons, which would expand if they were not trapped by the electrostatic attraction of the ions. Given enough time (picoseconds), the electrons thermalize with the ions, which expand hydrodynamically [65]. When the main pulse finally arrives, the surface of the target has already been diffused to low density (and has been pre-heated). In the context of this dissertation work, this is not a bad thing; the electrons in the diffused pre-plasma can be effectively accelerated by the main pulse. This is because the laser can propagate through the low-density region of pre-plasma, which provides a source of electrons, but it could not have propagated into the non-diffused liquid.

4.3 Electron acceleration by quasi-static electric-fields

Conceptually, the simplest electron-acceleration scheme is to use a simple electrostatic-field. Let’s say we’ve got an electric field with no magnetic fields. How does the electric field affect the electron?

\[ \vec{F} = q\vec{E} \]  

(4.1)

An electron in a constant electric-field will gain more and more energy as time goes on.

The theory of electrostatic electron-acceleration is widely known, but it’s important to remember that this basic acceleration mechanism can be present in a complex
system. Although lasers are dynamic interchanges of electric and magnetic fields, electrostatic fields can be found in plasmas. For example, the laser may drive electrons out of one region and into another through the ponderomotive force [22]. An electron will try to get away from regions of high electron-concentration. If one area has a surplus of electrons and another has a deficit, there will be a strong electric field from the depleted area to the surplus area. Electrostatic fields, or quasi-static electric-fields (not oscillating as fast as the light waves, but not exactly lasting forever), are established by having areas of unequal charge.

4.4 Direct laser acceleration

One way to accelerate electrons using the back-reflected laser is through direct laser acceleration: to sweep them up in the actual laser light waves. To start understanding direct laser acceleration, consider the electrostatic field scenario of the previous section, but now add in a uniform magnetic field. The force equation from the electrostatic case is modified to include the effect of this magnetic field. By standard electromagnetism theory, the force equation is:

$$\vec{F} = q(\vec{E} + \vec{v} \times \vec{B})$$  \hspace{1cm} (4.2)

The Lorentz force equation says that electric fields accelerate charged particles, while magnetic fields can only deflect them. However, deflection can be the key to an acceleration scheme. Let’s say I’m sparring in martial arts. I am tempted to charge at the judo master, but I know he/she will deftly redirect my momentum straight into the ground. The judo master doesn’t actually impart energy, yet I somehow end up on my face. This same sort of idea comes into play regarding direct laser acceleration of electrons. A laser is composed of transverse oscillating electric and magnetic fields, yet it can accelerate electrons along its propagation axis. It does this by combining transverse acceleration of the electric field with deflection by the magnetic field into the propagation direction.

Before we worry about even an idealized laser (the “plane-wave approximation”’), I’d like to build a basis for how an electron, starting at rest, will be accelerated by static, orthogonal electric and magnetic fields into the third orthogonal dimension. Let’s imagine an electron starting at rest in a uniform electric and magnetic field. The electric and magnetic fields are of infinite extent and in orthogonal directions. Let’s say that the electric field is in the +X direction, and the magnetic field is
in the +Y direction. What happens to an electron that finds itself, at rest, in this volume? Thanks to the electric field, it accelerates in the -X direction (it has negative charge.) Now that it’s moving, the magnetic field slightly deflects the electron in the +Z direction.

Now we can imagine a different volume, where the electric field is in the -X direction and the magnetic field is in the -Y direction. What happens to the electron that starts at rest? First, it accelerates in the +X direction. Once moving, the magnetic field slightly deflects the electron in the +Z direction, as before. In fact, the game is rigged: As long as the cross products of the fields are in the +Z direction, a particle of any sign charge, starting from rest, will be deflected into the +Z direction.

In a nutshell, the simplified electric and magnetic fields tell us how direct laser acceleration works to accelerate electrons in the direction of the laser propagation, at least in the most simple case of an electron starting from rest. A laser consists of orthogonal electric and magnetic fields that oscillate, and the cross product of the fields points in the same direction at all times. At any brief moment (less than half a cycle) in the laser oscillation, no matter which way the stationary electron is initially pulled by the electric fields, it also is deflected forward in the direction of the laser by the associated magnetic field.

Consider the plane wave approximation, in which uniform, orthogonal electric and magnetic fields of infinite extent oscillate sinusoidally with a single frequency, propagating forward via Maxwell’s equations. Imagine an electron popping into existence, at rest, at a point in space and time where the electric and magnetic fields are zero. If the first half of the next oscillation cycle gives the electron forward momentum, the second half takes it back. The first half of the oscillation deflects the electron motion somewhat into the plane wave’s propagation direction, but the second half of the oscillation deflects the electron motion back out of the propagation direction. At the end of one full cycle, the electron has translated in the +Z direction but is again at rest. If one desires to accelerate an electron to some non-zero final energy, the solution is that the electron must enter and exit the wave at the right moment, and under the right conditions. For the plane wave, this is impossible; it is uniform and infinite in all extents. For a real laser, entry into and escape from the wave is possible; it is geometrically and temporally featured, especially during interaction with a plasma. The permanent imparting of laser energy to an electron, through a
process similar to that described above (but with additional injection and ejection conditions), is called direct laser acceleration.

When describing direct laser acceleration and plasma-wave acceleration, physicists like to make an analogy to ocean-wave surfing. A surfer must paddle in the direction of the wave, gaining speed to match the wave, before standing up in its crest and getting the additional speed boost. Similarly, surfing a plasma or light wave requires some initial conditions: Electrons that do not have the appropriate velocity at the right moment of the wave cycle don’t effectively gain energy. Electrons that start too slow (or are moving in the wrong direction) oscillate in the wave as it passes, like a man bobbing in a raft (or swimming parallel to the shore) as waves pass under. If the electrons start too fast, they surpass the wave crests and valleys, gaining and then giving energy back to the wave each time, like a speed boat crashing past the waves on a lake. If the electrons start with just the right momentum at just the right position to catch the wave at its crest but not overtake it, they can get a sustained push from the wave and be accelerated to high energy. If the electron exits the wave before oscillation resolves (such as through a critical density reflection of the wave, or field dissipation through laser divergence), the electron can permanently retain its gained energy.

The mechanism of direct laser acceleration within a plasma is a major topic in its own right, the details of which are beyond the scope of this dissertation (see Krygier [66] and Ngirmang et al. [67] for detailed discussions). Direct laser acceleration and/or quasi-electrostatic/plasma-wave acceleration could explain the high energies of backward-going acceleration seen in the experiment. Our hypotheses is that the laser, reflected from the front surface, performs direct laser acceleration in the reflection. In this case, the standing-wave and/or SRS mechanisms, which will be described in the next two sections, would be important for giving the electrons the initial kick needed to catch the backward wave and surf it to high energies.

4.5 Standing-wave acceleration

Light reflected from a surface will interfere with the not-yet-reflected light and make a standing-wave pattern. The result is electromagnetic fields, located in the pre-plasma, that oscillate but do not appear to propagate [68]. The more equal the incident and reflected pulses, the more pronounced the standing wave pattern. The standing wave, which lasts as long as the pulses overlap, can boost electrons from low
energies to higher energies, pushing them into the reflected light for further direct laser acceleration. This mechanism for acceleration, which will be briefly described, is present for the moderately intense laser case \(a_0 \sim 0.5\) and the highly relativistic case \(a_0 \geq 1\). In the moderate-intensity case, acceleration is less efficient, operates over a shorter distance, and operates over a larger fraction of the laser period.

Kemp et al. [3] identified standing-wave acceleration as an important mechanism for accelerating electrons in the forward direction for normal incidence and at intensities \(\sim 10^{20} \text{ W cm}^{-2}\); Orban et al. [69] extended this understanding for accelerating in the backward direction for normal incidence and at intensities \(\sim 10^{18} \text{ W cm}^{-2}\). This mechanism is an efficient accelerator of electrons when the overlap of the forward-going and reflected laser pulse gives rise to a standing-wave pattern, i.e.,

\[
E_y(y = 0, z, t) = 2 E_{y0} \sin\left(\frac{2\pi}{\lambda} (z - z_c)\right) \sin(\omega t) \quad (4.3)
\]
\[
B_x(y = 0, z, t) = 2 B_{x0} \cos\left(\frac{2\pi}{\lambda} (z - z_c)\right) \cos(\omega t) \quad (4.4)
\]

where \(z_c\) is the position of a sharp interface where the reflection occurs and \(\omega\) is the angular frequency of the laser. The above standing-wave equations are found by adding together two oppositely traveling electromagnetic waves of form \(\vec{E} = E_{y0} \sin(\omega t \pm k z + \theta)\), where \(k = \frac{2\pi}{\lambda}\). \(\vec{B}\) is determined for each traveling wave such that it satisfies the relation \(\hat{k} = \hat{E} \times \hat{B}\), and choice of \(\theta\) is simplified with the condition that the combined electric field at \(z_c\) will be zero. Kemp et al. [3] consider electron acceleration in these time and space-varying electric and magnetic fields for highly relativistic electrons where \(v \sim c\). Fig. 4.1 illustrates the acceleration of electrons in both the forward-going (dashed lines) and specular directions (solid lines). As in other figures, the \(-z\) direction is the direction away from the target. Note that because of the constructive and destructive interference, there are moments in every laser cycle where \(E_y\) is zero and \(|B_x|\) is peaked, and moments where \(B_x\) is zero and \(|E_y|\) is peaked (Eqs. 4.3 & 4.4). Fig. 4.1 highlights these times, which are crucial for understanding the acceleration of the electrons.

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Figure 4.1: **Standing-wave acceleration mechanism for** $a_0 > 1$. An illustration of standing-wave acceleration at relativistic intensities ($a_0 \gtrsim 1$) in its four different stages. The target is located at some $+z$ value above the area shown. Thick solid lines indicate electrons that are ultimately accelerated away from the target while dashed lines indicate electron trajectories accelerated into the target. Panel a. illustrates the “push” phase, Panel b. illustrates the “rotate” phase, and Panel c. illustrates the “drift” phase. Finally, in Panel d., if the magnetic fields are substantially weaker than during the “rotate” phase the electron will experience a mild deflection and continue its overall motion. Graphic: Chris Orban.

In their paper, Kemp et al. [3] numerically integrated the trajectories of electrons in a simple standing wave (Eqs. 4.3 & 4.4). [3] concluded that the maximum momenta attainable by electrons is given by

$$p_{\text{max}} = 1.45 a_0$$

(4.5)

where $p_{\text{max}}$ is a normalized to $mc$. Kemp et al. [70] presented evidence from 1D(3v) simulations (their Fig. 4) that $p_{\text{max}}$ is a reasonably accurate estimate for a cutoff feature in their forward-going energy distribution. As illustrated in our Fig. 4.1, Eq. 4.5 should apply equally well to electrons accelerated away from the target. However, in this case, once outside of the standing wave, electrons should gain additional momenta from interacting with the reflected pulse.

A simple estimate for this additional momentum comes from [71] by applying a plane-wave approximation [72] to electrons moving with non-zero momenta away
from the target. These considerations yield

\[ p_{\text{zf}} = p_{\text{z0}} + \frac{-a_0^2}{2(p_{\text{z0}} + \sqrt{1 + p_{\text{z0}}^2})} \]  \hspace{1cm} (4.6)

for the “final” momentum, \( p_{\text{zf}} \), of the electron from interacting with the reflected laser light where \( p_{\text{z0}} \) is the initial momentum (e.g. provided by the standing-wave mechanism), and \( a_0 \) is the \( a \)-value of the laser field. Note that since motion away from the target implies \( p_{\text{z0}} < 0 \), as \( -p_{\text{z0}} \) becomes large the denominator of the second term in Eq. 4.6 tends to zero and the additional momentum provided by this second term becomes significant. Fig. 4.2 uses Eq. 4.6 to show how the cutoff momentum, and corresponding cutoff energy, scale with intensity by assuming \( p_{\text{z0}} = -1.45a_0 \).

Figure 4.2: Cutoff momentum vs. intensity for standing-wave acceleration. Simple estimates for the cutoff momentum (and corresponding kinetic energy). The solid black line shows the 1.45 \( a_0 \) cutoff from Kemp et al. [3], who considered standing-wave acceleration in the forward direction. The gray dashed line shows the prediction of Eq. 4.6 which is a simple plane-wave estimate for the momentum boost from electrons launched by the standing wave into the reflected laser pulse. The boost from reflected light significantly increases the final momentum of electrons accelerated via standing-wave acceleration. Graphic: Chris Orban.

Kemp et al. [3] treat standing-wave acceleration at relativistic intensities \( (a_0 \gtrsim 1) \), and conclude that \( p_{\text{max}} = 1.45a_0 \) provides a useful rule of thumb when electrons are
highly relativistic \((v \sim c)\). Electrons that are only moderately relativistic \((v \sim 0.5c, a_0 \sim 0.5)\) cannot be accelerated by this mechanism, but there is another pathway outlined in Orban et al. [52].

Figure 4.3: **Standing-wave acceleration mechanism for** \(a_0 \sim 0.5\). A slightly different acceleration pathway is taken by non-relativistic electrons; above is an illustration of standing-wave acceleration at moderately relativistic intensities \((a_0 \sim 0.5)\). Panel a. shows the “push” phase, exactly as before. Panel b. illustrates the “rotate” phase, which starts the movement away from the target. Panel c. shows how, a quarter cycle later, the standing-wave electric-fields accelerate the electron to the left. Finally, in Panel d., the electron trajectory can be deflected by the standing wave magnetic fields to become roughly parallel with the laser axis. Graphic: Chris Orban.

As before, electrons receive a “push” from the laser electric fields in the first step (Panel a), but their velocity is not fast enough to reach the node of the E-field by the end of the “rotate” step (Panel b). However, by this point the electron has an appreciable momentum in the \(-z\) direction and it will continue to move away from the target during the next step (Panel c) during which the laser electric field accelerates the particle to the left. In the last step (Panel d), the magnetic fields bend the electron trajectory to be nearly parallel with the laser axis, and the electron may continue moving away from the target. Note that this sequence works analogously a
half-cycle later for electrons near $\Delta z = 0.125\lambda$ that are pushed instead to the left by the standing-wave electric-fields followed by an analogous deflection away from the target by the magnetic and electric fields.

Notice that the electron has only moved $\lambda/8$ away from the target via this mechanism during the course of the laser cycle, whereas the electrons in Fig. 4.1 have moved $\lambda/4$ away over this same interval. This moderately relativistic standing-wave acceleration is less energetic and less efficient than the relativistic case illustrated in Fig. 4.1. However, the acceleration depicted in Fig. 4.3 occurs on the timescale of the laser cycle and electrons are ejected in sub-fs bunches, which is desirable for some applications, and these electrons can be launched into the reflected laser pulse and accelerated to substantial energies. To our knowledge, this mechanism had not been described in the literature prior to Orban et al. [52].

### 4.6 Stimulated Raman Scattering (SRS)

In addition to the standing-wave mechanism, Stimulated Raman Scattering (SRS) provides a way to produce large numbers of moderate-energy electrons. As in the standing-wave case, these electrons could then be further accelerated by the reflected laser light.

SRS is an effect in plasmas which creates large numbers of electrons through light conversion into plasma waves of half the frequency [21]. This effect is especially intriguing in the context of our experimental observations of three-halves omega light and half-omega light emitted from the target.

A plasma is defined as a system of charges which are coupled to one another via their self-consistent electric and magnetic fields. The electron plasma wave is a longitudinal electrostatic wave, which distinguishes it from light, a transverse electromagnetic wave. Both light and electron plasma waves can exist in a plasma, and both involve the oscillation of electrons in the plasma. However, propagating light and electron plasma waves of the same frequency are sustained differently depending on the plasma electron density.

The natural frequency of oscillation for electrons in a plasma depends on electron mass, charge, and number density:

$$\omega_{pe} = \sqrt{\frac{n_e e^2}{m_e \epsilon_0}}$$  \hspace{1cm} (4.7)
where $\omega_{pe}$ is the electron plasma frequency (the natural frequency of oscillation) and $n_e$ is the electron number density. The standard physical constants $m_e$, $e$, $\epsilon_0$ are respectively the electron mass, electron charge, and permittivity of vacuum.

The dispersion relation (function relating the wavenumber/wavelength to the wave frequency) for electron plasma waves is:

$$\omega^2 = \omega_{pe}^2 + 3k^2v_e^2$$  \hspace{1cm} (electron plasma wave)  \hspace{1cm} (4.8)

where $\omega$ is the frequency of the electron plasma wave, $\omega_{pe}$ is the electron plasma frequency, and the entire term $3k^2v_e^2$ is a small thermal correction dependent on wavenumber. $k$ is the wavenumber (radians per distance; $k = \frac{2\pi}{\lambda}$), and $v_e$ is the electron thermal velocity. In other words, the plasma best sustains electron plasma waves at the plasma frequency; all other waves will rapidly damp out.

This can be contrasted to the dispersion relation for light in the plasma, which is permissive to propagation of many frequencies:

$$\omega^2 = \omega_{pe}^2 + k^2c^2$$  \hspace{1cm} (light wave in plasma)  \hspace{1cm} (4.9)

where $c$ is the speed of light. The plasma can sustain all light wave frequencies higher than the plasma frequency, but lower-frequency light oscillations than the plasma frequency cannot propagate. Given the electron plasma frequency’s dependence on electron number density from Eq. 4.7, we can solve Eq. 4.9 for a “critical density” at which light of a certain wavelength (given in micron units) can no longer propagate:

$$n_{crit} = 1.1 \times 10^{21} \frac{\lambda_\mu^2}{\lambda_\mu^2} \text{ electrons} \cdot cm^{-3}$$  \hspace{1cm} (4.10)

Hence, the critical density is lower for low-frequency oscillations. The critical density is also the electron density at which the light becomes resonant with the plasma, and so plasma waves are well-sustained for the frequency $\lambda_\mu$ at density $n_{crit}$. In the case of an electron density gradient increasing monotonically from vacuum density to solid density, light can propagate past the point of half-harmonic resonance. It is at this resonance point (the quarter-critical density for $\omega$, equaling the critical density for $\omega/2$) that half-harmonic SRS is strong [21].

The $\omega/2$ plasma waves will resonate and continue to grow until the waves reach densities where they no longer resonate. In other words, one would expect strong $\omega/2$ plasma waves when there is a large physical extent of plasma near the quarter-critical density. The longer the scale length of pre-plasma, then, the larger the space
for SRS instability growth. $\omega/2$ plasmons and photons can join $\omega$ laser photons or plasmons to create $\frac{3}{2}\omega$ plasmons or $\frac{3}{2}\omega$ photons. If one experimentally observes large $\frac{3}{2}\omega$ optical light in conjunction with $\omega/2$ optical emissions, and the pre-plasma is extended, it is likely that strong $\omega/2$ plasma waves and SRS processes are at play.

I have only scratched the surface of SRS, and I would like to give interested readers a place to learn more. A book entitled “The Physics of Laser Plasma Interactions” [21] is an excellent resource. It covers many issues that are relevant including detailed descriptions of light waves and electron/ion waves in plasma, momentum and energy matching conditions which must be met for conversion between photons and plasma waves, and derivation of the growth rates for instabilities such as SRS under various plasma conditions.

4.7 Motivation for computational studies

The experimental results do not cleanly tell us how each of these electron acceleration mechanisms come into play in our experiment. It does ground us, however, and assures us that certain mechanisms are present in some form; for example, we do know that backscattered $\omega/2$ and $\frac{3}{2}\omega$ light is seen in conjunction with our electron acceleration, and that this can be an indicator of SRS plasma instabilities. We also know that an extended pre-plasma is essential to efficient electron acceleration. In our experiment, we have shown that the pre-pulse and intensity characteristics necessary for efficient $\frac{3}{2}\omega$ light generation are similar to those required for efficient electron acceleration. It is well-known that $\frac{3}{2}\omega$ light requires an extended region of near quarter-critical density plasma. It is also the case that $\frac{3}{2}\omega$ light is more efficiently produced in the simulations when the laser is made to focus before or after the critical density, not in it.

In one interpretation, the $\frac{3}{2}\omega$ light is connected directly to the electron acceleration. That is, perhaps the $\frac{3}{2}\omega$ light and electron acceleration are seeded from a common source: SRS. This interaction creates $\omega/2$ plasma waves and moderate-energy electrons. A $\omega/2$ plasma wave or photon can scatter with the fundamental light to make $\frac{3}{2}\omega$ waves and light. By dephasing of the plasma wave, the same SRS process creates moderate-energy electrons, and these electrons could be accelerated by the back-reflected laser. In another interpretation, the $\frac{3}{2}\omega$ light is only indirectly linked to the electron acceleration. That is, both require similar pre-plasma and intensity characteristics. For example, Short et al. [73] show evidence of $\frac{3}{2}\omega$ generation
from two-plasmon decay in filaments. Or, perhaps the electrons would be accelerated in the standing wave, while the $\frac{3}{2}\omega$ is created through $\omega/2$ scattering by SRS. Another possibility is that the chaotic nature of the laser-plasma interaction drives density fluctuations that results in strong quasi-static electric fields, and these fields to the heavy lifting of electron acceleration. The experiment record leaves open the possibility for some combination of all of these mechanisms.

While we do not have the experimental evidence to clearly distinguish the role of each of these mechanisms, one way to dig deeper into the problem is through computational modeling. In simulations, one can observe the very short timescales involved with the laser-plasma interaction. Our best experimental diagnostic still integrates over the entire laser interaction timescale, so this is a major gain. Another quality of simulations is access to electric field, particles, densities, etc. at all points in space in the simulation. This allows a precise tracking of the mechanisms at play in the simulations that cannot be matched with experiments. The big downside of a simulation, of course, is that it is not reality, so everything that is learned there must be verified to be numerically consistent and validated with real observations. We modeled the interaction through simulations in Ch. 5 to gain more insight into the mechanisms, taking care to assess the simulation based on its underlying physical soundness and several experimentally known outputs.
Chapter 5: Computational studies and physical interpretation

As described in the previous chapter, theory and prior experimental work leave open several electron-acceleration mechanisms which cannot be ruled out by the experimental results. The electrons could be accelerated backwards from rest through standing-wave acceleration and/or Stimulated Raman Scattering. The electrons could have their energies boosted through quasi-electrostatic fields, plasma waves, or direct laser acceleration.

In this chapter, I describe computational studies performed to assess the significance of each of these mechanisms and inform further experimental study. First, I will go over the basics of Particle-in-cell (PIC) simulation, and how the technique might be useful in interpreting this experiment. Next, I will show how an idealized version of our experiment exhibited standing-wave acceleration, but did not show the large amounts of charge accelerated. A more realistic simulation campaign was undertaken, in which the pre-plasma was modeled as an exponential decay. In these simulations, efficient high-energy electron-acceleration in the specular direction was observed (as in experiment), with an efficiency of the simulation highest under conditions of focusing near the critical density rather than near the solid density surface. Varying intensity inputs to the simulations revealed the importance of reflectivity, supporting the standing-wave-and-direct-laser-acceleration theory. To move to a higher level of analysis, the pre-pulse effect was modeled in FLASH hydrodynamic code. This hydrodynamic simulation showed new features such as a two-scale length exponential density decay and a 3-dimensional depression, which had not been previously modeled. Importantly, the pre-pulse was not modeled at a single focal-depth; rather, this hydrodynamic simulation was integrated with the PIC code and assessed at several laser focal-depths. These parameter scans were performed to mimic our experimental laboratory procedures for alignment, and the resulting simulation is shown to better
match experimental parameters such as total charge accelerated. The most realistic simulation exhibits new and complex plasma wave dynamics, which are a topic for study beyond this dissertation. I conclude this section by assessing our best theoretical understanding, which has been formed from analysis of the experiment and simulations.

5.1 Particle-in-cell (PIC) simulations of main pulse interaction

Particle-in-cell simulation works by solving Maxwell’s equations for fields on a discretized grid, with particles existing within that grid but independent of the grid points (i.e. the particles exist in the grid cells). Two simplifying physical concessions are made in these simulations: fields are calculated on a discrete grid, and particles are represented as “macroparticles.” Rather than modeling individual electrons, protons, ions, etc., charge density is created through “macroparticles” in the grid. This choice is to sidestep the computational impracticality/impossibility of modeling the large number of real particles present in even a micron-scale’s volume of material. Macroparticles are oversized versions of real particles. For example, an electron macroparticle has the same charge-to-mass ratio as a real electron but much larger charge and mass; the macroscopic motions of an electron macroparticle in an electromagnetic field will match those of electrons. The macroparticles can directly interact with each other through statistical local collisions (if this effect is turned on in the software), or indirectly interact with one another through electric and magnetic fields interpolated from grid points. The macroparticles move under the influence of the grid fields, then update the grid fields according to their new positions.

Macroparticles in a PIC code are sometimes described as clouds of charge. This is true in the sense that the electric field is calculated on a grid, which means that (as long as collisions are turned off), one does not have strong \( \frac{1}{r^2} \) interactions wherever \( r \) is smaller than the grid spacing. In effect, then, macroparticles are clouds of charge that are about as large as the grid spacing. Because a subset of Maxwell’s equations are solved at each time step across the grid points, light will propagate in the absence of macroparticles (up to a frequency limited by grid spacing and time step). A laser can be added to a PIC simulation by appropriately oscillating the electromagnetic fields at the boundary of the simulation.
We performed PIC simulations using the code LSP [74] in a 2D(3v) Cartesian geometry. Since the laser-target interaction is simulated using only two spatial dimensions, symmetry must be assumed along some physical axis. We take advantage of the translational symmetry along the length of the water jet and make the natural choice to set the symmetry direction parallel to the direction of the jet. The simulated laser pulse strikes the water target with a tangential polarization vector as it does in the experiment.

For simplicity, and in an effort to minimize the computational expense of the simulations, we do not simulate the entire 30 μm diameter of the water jet. Instead, only a section of the water jet is simulated. This section is further simplified to a flat-slab geometry (instead of including the natural curvature of the water jet). More realistic initial conditions for the simulations will be discussed in later sections of this chapter.

5.2 Idealistic, diffuse pre-plasma PIC simulations

A first series of PIC simulations with diffuse, perfectly uniform pre-plasma high-light dynamics of standing-wave acceleration. The idealized aspect of these first simulations stems from using an extremely low pre-plasma density (10^{10} \text{cm}^{-3}) with a flat density profile\footnote{The pre-plasma density is set low enough that charge separation effects should be minimal. As such the electron macroparticles should respond to the laser electric and magnetic fields as tracer particles. The ions in the simulation were immobile and fixed in ionization state in order to create a neutralizing background. The results we present in this section are insensitive to the exact value of this extremely low density, as one would expect.} and by using an ideal conductor, which acts as a perfect mirror, to reflect the laser pulse. With this choice the electric and magnetic fields along the laser axis and where the forward and reflected pulses overlap are well described with by simple standing wave (Eqs. 4.3 & 4.4).
Figure 5.1: **Idealized, diffuse-plasma simulation electron trajectories.** *Left panel:* Electron trajectories from idealized simulations where the same ultra-intense laser pulses are incident on a perfectly reflecting interface with an extremely diffuse pre-plasma. Trajectories have been color coded according to their $p_z$ momenta towards (gray or white) or away (dark gray or black) from the target. Although the trajectories are highly chaotic in each case, this shading helps to highlight the moment when the macroparticle is flung away from the target. *Right panel:* An analysis of angles and kinetic energies of electrons that are accelerated away from the target. The dotted circle represents the kinetic energy cutoff predicted by Eq. 4.6.

Fig. 5.1 presents the main result from these idealized simulations. The left panel shows the electron trajectories while the right panel shows an analysis of the energies and angles of the escaping electrons. The electron trajectories in the left panel have been color coded according to their momenta towards or away from the target. Although the trajectories are highly chaotic in this case, the shading helps to highlight the moment when an electron is flung away from the target. Once “launched” in the $-z$ direction, an electron may be deflected and accelerated by the reflected laser pulse but it will generally continue moving away from the target until it exits the simulation. The $5 \cdot 10^{18}$ W cm$^{-2}$ result in Fig. 5.1 naturally provides a good illustration for standing-wave acceleration with $a_0 \gtrsim 1$. A close look at the trajectories near the laser axis and near $z \sim -17 \mu$m do exhibit the quarter-circle turn away from the target as described in Fig. 4.1. Other trajectories show motion in the $x$-direction followed by a somewhat-more-than-90-degree turn away from the target that still produces electron motion away from the target with significant $p_z$ momentum.
Eq. 4.6 does a reasonable job of predicting the cutoff energy in the idealized simulation, as seen in the right-hand panel of Fig. 5.1. The dotted circle shows the kinetic energy corresponding to the momentum cutoff of Eq. 4.6. This prediction matches well the result where the dotted circle appears at energies just beyond the red-shaded zone that indicates the energies and angles where most of the electrons exit the simulation.

5.3 Exponential pre-plasma PIC simulations

The idealized simulations modeled the pre-plasma as a low-density plasma, and the target as a high-density conductor. The transition between low density and high density was a discontinuous step. To better match the experiment, an effort was needed to mimic the pre-plasma that is observed through shadowgraphy and interferometry in the experiment. Rather than a discontinuous step to low density, the pre-plasma is modeled as a decaying exponential density ramp from liquid density out to vacuum. By changing the laser intensity and pre-plasma scale parameters while observing the electron acceleration, these simulations highlight features that may be important to the experiment. I will describe these insights in the next several subsections.

To understand the interplay of basic mechanisms driving electron acceleration, for simplicity we assumed an exponential-scale-length pre-plasma density profile instead of using a more realistic pre-plasma profile. To be consistent with the overall extent of the observed pre-plasma (through interferometry), this exponential scale length must be $\ll 10-20 \, \mu m$. The exponential scale length is set to be $1.5 \, \mu m$ except where otherwise indicated. The qualitative results are unchanged for scale lengths as small as $\sim 0.75 \, \mu m$ and as large as $\sim 4 \, \mu m$. The simulations were run between 400 fs and 500 fs in order to adequately model the laser propagation, target interaction and the propagation of the electron beam. Electron trajectories were tracked in these simulations for later analysis.

5.3.1 Role of high reflectivity

After experiencing standing-wave acceleration or SRS, an electron has the chance to be further accelerated by the reflected laser. This acceleration in the back-reflected
light may be the key to understanding the high-efficiency, high-charge electron acceleration under study. In this case, we need two things: 1) Source of numerous moderate-energy electrons 2) High reflectivity for an intense pulse in back-reflection.

When intensity is varied in simulation, efficiency is seen to track with reflectivity. The high reflectivity requirement for electron acceleration likely explains why the laser experiment works best when the target is not at best focus. Water, and plasma in general, is more reflective at moderate intensities (Fig. 5.2). The numerous-moderate-energy-electrons condition also explains the balancing act between lots of pre-plasma and a little; too little pre-plasma, and there is no source for moderate-energy electrons. Too much pre-plasma, and the pulse does not reflect.

Figure 5.2: **Reflectivity scaling with intensity and pre-plasma scale.** Reflectivity scales depending on the presence of pre-plasma and the extent to which the material is a conductor. Graphic: Chris Orban.

Absorption increases at high intensity because of relativistic effects. Since \( a_0 > 1 \), the inertia of the electrons is larger than it normally is. This causes an increase in the energy absorbed by the laser. At much lower intensities (i.e. pre-pulse intensities), the dominant mechanism is collisional absorption (i.e. inverse Bremsstrahlung). But the hotter the plasma gets, the less collisional it becomes. Near the regime of \( 10^{18} \) W cm\(^{-2} \), the plasma is very hot and collisional effects are minimal; this has been
checked this by turning collisions off in LSP and noticing that we get almost exactly the same ejected electron results. However, the intensity is still not yet so high that relativistic absorption becomes important. $10^{18}$ W cm$^{-2}$ appears to be a peak in the reflectivity versus intensity.

### 5.3.2 Femtosecond bunching of emitted electrons

**Figure 5.3:** Femtosecond bunching of accelerated electrons. Femtosecond bunching is seen in 2D PIC simulations, but has not been experimentally confirmed. A plot of the electron charge leaving the PIC simulations through the specular boundary ($z = -30 \mu m$) as a function of time (0.2 fs bins). For each intensity, electron macroparticles that escaped with kinetic energies $> 100$ keV were recorded. Results with different kinetic energy thresholds are qualitatively similar. Graphic: Chris Orban.

An empirical observation from the simulations worthy of note is that many of the escaping electrons leave the target in sub-fs bunches. Fig. 5.3 shows the amount of charge per time leaving the edge of the simulation at $z = -30 \mu m$ binned in increments of 0.2 fs (i.e. 4× the timestep). Particularly apparent at $10^{18}$ W cm$^{-2}$ and $5 \cdot 10^{17}$ W cm$^{-2}$ intensities are moments where a substantial amount of charge leaves the edge of the simulation in under a femtosecond. There may be sub-fs bunching in the $5 \cdot 10^{17}$ W cm$^{-2}$ case as well. Similarly short bunches of electrons have been
observed in simulations of solid density target irradiation by Naumova et al. [31] who emphasize the novelty of using these bunches to create secondary light sources with ultra-short or attosecond features. From the standpoint of the AFRL experiment, confirmation of the bunched nature of the escaping electrons (e.g. through detection of coherent transition radiation [75]) remains an important goal for future work.

5.3.3 Spatial distortions exist in reflection, standing wave enhances electric fields

Figure 5.4: Simulation fields and trajectories in reflection. Reflected light is distorted by the target, being non-flat and a non-uniform conductor. This is seen in 2D PIC simulations to actually collimate the reflected light and provide a better acceleration mechanism. Left panel: Transverse electric fields and electron trajectories from a $10^{18}$ W cm$^{-2}$ simulation with a realistic 1.5$\mu$m scale length pre-plasma. Right panel: Transverse electric fields and electron trajectories from an “idealized” $10^{18}$ W cm$^{-2}$ simulation described in Orban et al. [52]. Both panels show results at $t = 60$ fs after the front of the laser pulse has arrived at either the critical density or the perfectly reflecting surface. At this time the standing wave is no longer present and the laser pulse is entirely in reflection, moving towards the bottom of the page.

Comparing the reflected laser fields in Fig. 5.4 shows that in the realistic case the laser field is highly modified from interacting with the target, whereas in the idealized case the reflected laser pulse is still a simple function of time and space. This essential difference between the reflected laser fields provides a good explanation for
why Eq. 4.6, which is derived with a simple plane-wave assumption, gives reasonable results for the idealized case but not for the realistic case.

Comparing the electron trajectories plotted in Fig. 5.4 for the realistic case and the idealized case we can see how the complex structure of the realistic reflected pulse works to keep escaping electrons closer to the laser axis for significantly longer. Because of the longer interaction time with the laser fields, these electrons ultimately reach higher kinetic energies than in the ideal case. The confinement of these electrons along the laser axis also explains why the realistic simulations feature significant numbers of electrons escaping parallel to the laser axis, whereas the idealized simulations almost exclusively show electrons moving at two preferred angles away from the target, depending on where the electrons originate.

Compared to other studies investigating the reflection of laser pulses from solid targets, Ruhl et al. [8] attribute the collimation of an electron beam emerging from a solid target at oblique incidence to quasi-static magnetic fields that build up in the pre-plasma. Pegoraro et al. [2] also emphasize the importance of quasi-static magnetic fields for electron acceleration in underdense plasmas. While we see persistent magnetic fields in these PIC simulations after the laser pulse has reflected from the target, careful analysis of escaping electron trajectories indicate strong deflections that are only from the reflected laser pulse.

5.3.4 Match to experimental electron spectra

3D PIC simulations with a single exponential pre-plasma scale were performed with the same LSP configuration as the 2D simulations.\textsuperscript{19} Fig. 5.5 shows a comparison of 3D PIC simulation electron spectra to experimental electron spectra. The 3D PIC spectra are calculated using only those electrons ejected in the laser specular direction ($0^\circ \pm 6.3^\circ$, which mimics the size of the hole in the OAP). The plot demonstrates very good quantitative and qualitative agreement between experiment and simulation above 1 MeV, with the two PIC simulations bracketing the experiment both in laser intensity and electron spectral density.

\textsuperscript{19}Further details of the 3D PIC simulations and results are found in [67].
Figure 5.5: Agreement of electron spectra between experiment and simulation. Plotted here is an averaged experimental electron spectrum of Fig. 3.16 (with its standard deviation, or 1σ range) alongside two 3D PIC simulations with laser intensities bracketing the experimental intensity. Important to note is that the y-axis (charge density) is absolute and the same for both experimental and simulation plots. For the PIC simulations, only electrons emitted backward (toward the OAP) within a 6.3° half-angle cone (size of the hole in OAP) are included. As in Fig. 3.16, the experimental spectrum is adjusted for detector sensitivity and electron scattering losses in 20 Torr. The overall quantitative agreement fully supports our discussion of high-efficiency electron acceleration, and the shared qualitative features above >1 MeV are evidence of a super-ponderomotive-acceleration mechanism.

Below 800 keV, PIC simulations have consistently predicted more numerous electrons than were measured in experiment, both in the simulations of Fig. 5.5 and in numerous other simulation spectra not shown. One possible explanation for this discrepancy is that effects such as self-repulsion of electron bunches and long-distance deflection from a charged target cannot be fully expressed in the femtosecond-dynamic PIC simulations. In addition to good quantitative agreement, certain qualitative features are reproduced between the experimental and electron spectra. Two important such features are the spectra “leveling out” at electron energies well exceeding the ponderomotive energy scale (which is between 50 and 150 keV in all cases), and the lack of a single exponential decay. This agreement between PIC simulations
and experiment, seen across simulations with numerous configurations and numerical resolutions, fully supports our discussion of high-efficiency electron-acceleration. The shared qualitative features support our decision to study PIC simulations for insights into the experimentally identified super-ponderomotive-acceleration mechanism. Although I have chosen to plot the 3D, $\lambda/8$ cell-size-resolution simulations, good qualitative and quantitative agreement has also been seen consistently in the higher-resolution ($\lambda/16$ and $\lambda/32$) 2D PIC simulations.

5.4 Role of plasma scale length, focal depth, geometry, and back-scattered light

The strongest experimental signature of the electron acceleration experimentally was a strong dependence on the presence of a pre-plasma. This strong dependence is also seen in 2D PIC simulations, as is clear from Fig. 5.6.

![Figure 5.6: Conversion efficiency with and without pre-plasma. Conversion efficiency from laser energy to backward-escaping electron energy as a function of intensity, with and without a pre-plasma. Effectively, in this range of intensities, a target unperturbed by laser pre-pulse does not exhibit backward electron acceleration. This has been validated in our experiment.](image-url)
I did a pre-plasma and focal depth scan in the 2D PIC simulations with a laser at $3 \cdot 10^{18}$ W cm$^{-2}$, mimicking how the laser pre-pulse and focal depth would be explored in the laboratory during a typical alignment. In the simulation, the pre-plasma exponential scale length was varied between 1.5 $\mu$m and 8.0 $\mu$m, while the laser focal depth was varied between 30 $\mu$m before and after the critical density surface. The harmonics emission increased as the laser was focused in front of or behind the critical surface, while the peak electron energy increased when the laser is focused at the critical surface. The reflected light is seen to come off in filaments, as would be expected for such high intensities. The harmonics in 2D LSP simulations are seen to increase as the laser is focused before or after the critical density. Since greatest electron acceleration in our experiment is correlated both with high optical harmonic generation and with the laser being focused tens of microns in front of or behind the target, this is self-consistent.

In addition to studying electron ejection, I analyzed the frequency of the back-reflected light in Particle-in-cell simulations to determine the significance of harmonics to electron acceleration. This was done to pursue any possible correlations between harmonic generation and electron acceleration during the intense laser-matter interaction. I extracted the fields as a function of time and space, and then did a Fourier transform on the time dimension to extract power as a function of frequency. Looking only at the part of the simulation in vacuum allowed me to analyze the reflected light power spectrum. Half-harmonic generation was reproduced at between the critical and quarter-critical densities (see Fig. 5.7). Two-harmonic emissions were seen near the critical density. These results are well in line with theory. Numerically, harmonics emission was seen to be sensitive to simulation resolution; increasing from $\lambda/16$ to $\lambda/32$ changes the power spectrum. Plasma scale length parameter scans and focal depth scans show how backscattered light trends with these parameters; slightly out of focus (10 $\mu$m to 40 $\mu$m in either direction) increases backscattered light. Unlike the experiment, electron acceleration does not fall off strongly as a function of focal depth for this set of simulations.
Figure 5.7: **Fourier transform analysis of PIC simulations.** Left panel: Frequencies present in the simulation were analyzed via a temporal Fourier transform through all points in simulation space. This transform shows evidence of $\omega/2$, $3\omega/2$, and $2\omega$ plasma waves in the simulation. In this simulation, the laser focal depth is set 30 microns past the original target surface. It is in these defocused cases that the strongest harmonics are seen in our simulations, matching well with experimental experience. Right panel: The $3\omega/2$ plasma waves and/or light are seen to occur between the quarter-critical (green curve) and critical densities (white curve), as expected from SRS theory.

I also explored some different target geometries: a plume and a cup. The density profiles were created “by hand” in python. While back-reflected light changed, neither showed significant deviation from the slab-target geometry. However, stitching a cup into the target showed a marked increase in back-reflected intensity. The cup acts as a miniature optic, focusing the light, although not as would be expected from simple ray optics (due to self-filamentation at these intensities).
Figure 5.8: **Slab and cup target comparison.** The cup target in LSP simulations shows greater focusing in reflection than the flat target. Areas of greater electromagnetic energy are whiter; arrows show the Poynting vector of light, highlighting the divergent reflection from the slab target and more-collimated reflection from the cup target.

5.5 Hydrodynamic simulations of pre-pulse irradiation

While the LSP simulations have been able to reproduce many experimental features, other features are notably absent. There was experimental evidence to suggest more complex pre-plasma than a single scale length, but it is difficult to extract precise densities from the interferometry. For example, I tried to stitch together the pre-plasma profile based on what I knew from the experimental interferometry, but there was too much latitude in absolute electron number density for reliable transfer to LSP. In addition, interferometry could not see within the densest regions of the plasma, as the probe pulse, at 420 nm wavelength, cannot penetrate deeply into regions that are most important for the ultra-intense laser interaction. We could not directly see what is happening to the pre-plasma scale near the critical density. Geometrical features such as a conical imprint from the laser were suspected, but it was unclear whether a plume-like bulge or a cone-like cavity would dominate the geometry of the pre-pulse. LSP simulations assumed an exponential density profile and with a flat geometry for simplicity and in spite of clear indications that the pre-plasma contains more features than this. Finally, focal depth of the laser is, in our experiment, the same as the focal depth of the pre-pulse. This means that whatever pre-plasma the pre-pulse makes, the main pulse can only focus at one depth within
that target; it was unknown at what density in the pre-plasma the main-pulse laser was focusing.

The hydrodynamic code FLASH [76] was used to model the pre-pulse interaction with the target, giving a 3-dimensional idea of the target prior to the arrival of the main pulse. Our goal was to enhance our understanding of the V-like feature we see in shadowgraphy, and get an absolute number density for electrons as a function of depth. The pre-pulse from diode measurements was synthesized into a longer-pulse pattern (with the same energy) and simulated in FLASH.

FLASH is considered a hydrodynamic code because it models electrons and ions as coupled fluids, rather than as interacting particles. In a hydrodynamic code, each cell carries a density, temperature, and pressure. In FLASH, a cell contains both an electron temperature and an ion temperature. Deposited laser energy increases the electron temperature. Ion temperatures increase in response to the elevated electron temperature because the electrons and ions are treated as coupled fluids. One shortcoming of FLASH is that the physics of room-temperature fluids are not modeled. However, the code is completely open-source, well-documented, well-benchmarked for short-timescale/hot plasmas, and has a strong user support structure.

I inherited the FLASH simulations within our group from Mark Schillaci. Mark performed modeling of the pre-pulse as his Master’s thesis project in our group [77]. While the simulations had been performed in 2D cylindrical coordinates with a laser impinging on a water droplet, we had not yet attempted to compare these results with our experimental interferometry or integrate these results into the LSP simulation. I did analysis on Mark’s final simulations by performing an Abel transform in density on the FLASH outputs. The result qualitatively matched our index shifts map from interferometry, which implied that this was an avenue worth pursuing. With extensive help from Petros Tzeferacos of the FLASH center (Univ. of Chicago), I added a water column as a target to FLASH (its 3D geometry) and modified the simulation to prevent the expansion of the target into vacuum due to the hydrodynamically incorrect treatment of the liquid as a gas. I also modified the laser pre-pulse to more accurately reflect ours. Specifically, I matched the energy and temporal features of our photodiode diagnostic to the FLASH-simulation-modeled laser. This measured diode response showed a replica of the main, 30 fs short pulse. Since FLASH is not designed to simulate high-intensity, short-pulse phenomena, this “spike” in intensity was temporally broadened to 1 ns with the same energy. This will miss
the details of the short-timescale interaction, but deposit the appropriate amount of energy to drive hydrodynamic expansion.

5.6 Passing the baton: FLASH as initial conditions to LSP

I designed a handoff between FLASH and LSP so that the conditions at the end of a 10 ns FLASH pre-plasma simulation become the beginning of a 400 fs PIC main interaction simulation. FLASH models the pre-pulse interaction with the water column, and the distortions to the water column and diffusing of it as plasma. LSP models the interaction of the main pulse with this much-distorted plasma.

I will now describe the specific transition from FLASH to LSP, which is scripted to enable multiple iterations of the simulations. If water is flowing in -Y direction, and the laser is incident along +X. FLASH does not model the laser field propagation or its polarization; its laser modeling involves only geometric ray-tracing and energy deposition. After the pre-pulse interaction simulation is completed in 3D FLASH, I extract a density profile at Y = 0. I input this density profile into LSP, which has the laser incident along +X and polarization along +Z. The FLASH coordinates are transformed into LSP coordinates such that the critical surface always sits near X = -5 \( \mu \)m, leaving 30 \( \mu \)m before and 20 \( \mu \)m after the critical surface, plenty of space in which to model the interaction. The laser focal depth is handed off from FLASH to LSP, such that if the laser is focusing near the original target surface in FLASH, it is focusing at that same point in space (no matter what has happened to the target surface since the FLASH simulation started) in LSP.

The entire LSP simulation is initialized with eight free electrons for every two \( \text{H}^+ \) and one \( \text{O}^{6+} \) ion. We believe this is valid for two reasons. First, the FLASH simulation will underestimate the ionization state of the target because we end the simulation while the laser intensity is still below 10\(^{13}\) W cm\(^{-2}\), which is an approximate rule of thumb for the largest intensity where the fluid approximations in FLASH are valid. In the real experiment, the last pico-second of the pre-pulse will include intensities that exceed this value. To minimize the computational expense of the LSP simulations, we do not model this final picosecond before the main, 30 fs pulse reaches the target. Second, pre-ionization was verified to make an insignificant difference to ejected-electron spectrum and angular outputs. This was shown by initializing the LSP simulation in a lower ionization state (three free electrons for every two \( \text{H}^+ \) and one \( \text{O}^+ \) ion). The front of the main pulse quickly ionizes the entire laser area to
O\textsuperscript{6+}. LSP does not model ionization energy loss from the laser, so laser energy is not lost to this ionization in the simulation. The ions are ionized to O\textsuperscript{6+} instead of O\textsuperscript{8+} because of the much higher ionization potential of the two innermost-shell electrons. Finally, performing these simulations with a higher ionization state has computational advantages, because the number of electron macroparticles in the simulation grows much less rapidly than in simulations where the oxygen ions are singly ionized. It also enables better analysis of the electron acceleration pathways, as electrons created through ionization during rather than before the simulation cannot be tracked in LSP.

5.7 New features found in preliminary analysis of the handoff simulation

The modeled pre-plasma PIC simulations brought the best match to experiment, and with it, complex plasma dynamics. We have just begun to analyze the details of the plasma dynamics in these new simulations, and these electron dynamics will remain outside the scope of this dissertation work. However, I am explaining these handoff simulations and including some of their results here because we have already gained several clear insights from these simulations, both from the pre-pulse FLASH stage and the main pulse LSP stage.

Rather than assume one focal depth, the target was “aligned” in the handoff simulations exactly as we would do in our laboratory experiment: by scanning the focal depth and looking for the highest number of backward-ejected electrons. The handoff simulation was performed for nine laser focal-depths. Remarkably, the laser-to-electron energy conversion efficiency of this simulation at its best focal depth (laser intensity $1.5 \cdot 10^{18}$ W cm\textsuperscript{-2}, laser focus 15 μm within the target) reaches 4.5%, exceeding that found with previous parameter scans of a single exponential scale length by about a factor of three.
Figure 5.9: **Laser-to-electron conversion efficiency at varied laser focal depth.** Laser-to-electron conversion efficiency is plotted against laser focal depth (relative to the unperturbed water-column-target surface) in ten 3D FLASH + 2D LSP handoff simulations. The focal depths are chosen for high focal-depth resolution surrounding $+15 \, \mu m$. For this plot, an LSP peak laser-intensity of $3.0 \cdot 10^{18} \, \text{W cm}^{-2}$ is used; when $1.5 \cdot 10^{18} \, \text{W cm}^{-2}$ intensity is used (better matching experimental laser parameters, not shown), $>120 \, \text{keV}$ conversion efficiency is 4.5% at $+15 \, \mu m$. The plotted simulation results support experimental dosimetry focal scans (c.f. Fig. 3.2) showing that slight laser defocus relative to the unperturbed target surface provides best acceleration conditions.

A dip in electron acceleration efficiency is seen as moving through focus (Fig. 5.9), which is a signature reminiscent of the experiment. Perhaps counter-intuitively, aligning the laser to focus directly on the water column surface is less efficient than focusing fifteen microns before or after the surface both in the experiment and simulation. The explanation for this partly lies in that the pre-pulse modifies the target in such a way that the critical surface location moves away from the target surface when the pre-pulse is focused on the target surface. This is seen in the FLASH stage: placing the laser focus precisely at the liquid target surface can result in a poor main pulse focus at the final critical surface.
Figure 5.10: **Double-exponential fit to FLASH pre-plasma density.** FLASH simulations of pre-pulse interaction with the water target show that a double-exponential fit to the final pre-plasma density profile is more appropriate than a single-exponential fit.

The FLASH simulations also indicate that a single exponential scale length is inadequate to describe the experimental density profile. The density profile seen in the modeled case fits a two-exponential scale length (see Fig. 5.10). The two exponentials can be joined at a “knee”\(^{20}\); while this is seen in the FLASH simulations, it was not modeled in prior LSP simulations. The long exponential part of the pre-plasma seems to serve two primary roles. First, it shields the backward-emitted particles from the strong charge-up of the main pulse interaction region, allowing them to escape in large numbers. Second, it is in this region in which the \(\omega/2\) and \(3/2\omega\) optical harmonics occur, which is important for understanding our experimental measurements.

\(^{20}\)It would appear that this dynamically heated hydrodynamic expansion, modeled with a single electron temperature per space and time coordinate, gives a qualitatively similar feature to the well-studied two-temperature expansion (c.f. Fig. 3 of [78].) Exploration of this relationship is outside the scope of this dissertation.
Figure 5.11: Depression in water column, FLASH simulations. 3D FLASH simulations of the pre-pulse interaction predict that a sizeable depression of the liquid density surface (plotted blue) is established prior to the main pulse interaction. This plot represents the state of the water column 1 ps prior to the main pulse interaction. The critical density surface (not plotted) also exhibits a concave shape. This geometrical depression suppresses wide angles of backward emission of electrons (due to collisions, at these angles, with the surrounding dense plasma).

Another prominent feature of the FLASH simulations is that the pre-pulse causes a large depression in the water column surface (see Fig. 5.11), and this depression is seen also in the critical surface. We originally expected that the pre-pulse would make more of a pre-plasma plume than a pre-plasma depression, so this result required modifications in our mental models. Upon observing the outputs of LSP simulations, it becomes clear that one role this depression serves is to suppress wide angles of backward emission of electrons. The density details of such a depression would be unobservable via interferometry, as it exists within the water column. However, looking back at experimental interferometry data, we have observed large disturbances toward the back of the target (e.g. Fig. 5.12) prior to the arrival of the main pulse that lend credibility to this effect.
Figure 5.12: **Experimental shadowgraph showing possible cavitation due to pre-pulse.** This is a contrast-enhanced experimental shadowgraph showing the state of the water column 1 ps prior to the arrival of the main pulse. The image is made by the shadow of a 420 nm probe beam which is perpendicular to the main laser and water column, as described in Appendix A. The photodiode-measured pre-pulse from this experiment became the template for the FLASH simulation. The shadowgraph has been annotated to show how the FLASH hydrodynamic simulation result of a cavitation is supported by experiment.

We have seen violent electron density waves connected with high efficiency electron acceleration in our most precise attempt to match experiment and simulation to-date. What we do not know is how those density fluctuations actually drive the plasma waves. Acceleration occurs at and behind the critical density ($1.7 \times 10^{21}$ electrons/cc), so my SRS hypothesis as the source of acceleration is not supported (although 3/2-omega plasma waves are generated in another location of the simulation). These simulations involved a focal scan of 3D FLASH + 2D LSP, and showed much higher conversion efficiency (up to 4.5%) when compared with our previous simulations (capped around 1.5%).
Figure 5.13: Violent plasma dynamics of FLASH + LSP handoff simulations. Violent plasma dynamics play out in the PIC stage of the 3D FLASH + 2D LSP handoff simulations. The laser enters from the left side of the simulation; at this timestep (147 fs into the 400 fs LSP simulation), the main pulse has already mostly reflected. The colormap shows electron density, where $1.7 \cdot 10^{21}/cm^3$ is the critical density. The dots represent a selection of electron macroparticles, each of which has been or is destined to be accelerated to the left. The black particles have already been accelerated to beyond 1 MeV energies; the white particles have yet to be accelerated. We have recently begun analyzing these simulations, and look forward to learning many new things about the electron acceleration dynamics. The bulk of this analysis is beyond the scope of this dissertation.

5.8 Summary and discussion of simulation results

As realism was added, more and more experimental features were matched, but at the cost of simplicity. The simplest simulations were valuable in that they enabled us to test ideas about the basic roles of each process. The more complex simulations allowed us to benchmark simulation against experiment and to study the processes in a more realistic environment. As has been demonstrated in the past several chapters, isolating these features and processes through simulation is has shown to be effective.
We have come to the following conclusions regarding the mechanisms of electron acceleration in the simulations:

1. Electrons are launched both towards and away from the target through interactions with the standing wave created by the overlap of forward and reflected light. Electrons can be launched if they are positioned near the half-way point between a node and an anti-node of the electric field. These locations experience both strong electric and magnetic fields due to the standing wave. Electrons may be affected by strong electrostatic fields from charge separation in the pre-plasma, especially near the critical density where the density is large.

2. Electrons that are launched from in the specular direction find themselves in a diffuse plasma, and have an opportunity to get an extra “boost” in kinetic energy by interacting with the reflected laser pulse through direct laser acceleration. This boost does not occur for electrons launched into the target; those electrons are instead slowed by interactions with the dense plasma.

3. This kinetic energy boost from the reflected laser pulse is more effective than expected from simple assumptions.

4. Quasi-static electric fields arising from the modification of the electron density profile by the standing wave can also play a role in increasing the electron kinetic energies.

The simulations did not identify SRS as the primary mechanism of electron acceleration; most of the electrons were seen to originate beyond the critical surface (in regions denser than the $\omega/2$ plasma waves). While I will look forward to the possibility of future work finding unique connections between our experimental electron-acceleration mechanism and SRS, I propose the following possible explanation for the correlation between high-number electron acceleration and $\frac{3}{2}\omega$ that has been seen in the experiment:

1. In the case of severe wavefront deformation (due to the hole in the off-axis parabola), optimization of intensity in artificial “no-target” vacuum conditions (as is routine, with a focal spot camera) is distinct from optimization of the focal intensity within the pre-plasma of the experiment.
2. The strength of $\frac{3}{2}\omega$ optical emissions provides a metric of the intensity of laser focus within the plasma, between the quarter-critical and critical surfaces, as per SRS theory. As the best focus is obtained between the quarter-critical and critical surfaces within the plasma, the $\frac{3}{2}\omega$ emissions increase and intensity near the critical surface is increased which enhances the standing-wave and direct laser acceleration mechanisms we have observed in simulation.

3. The presence of $\omega/2$ and $\frac{3}{2}\omega$ light verifies the presence of an extended sub-critical pre-plasma. In simulations, the return current from extended sub-critical pre-plasma is seen to effectively shield backward-accelerated electrons from the strong attractive electric fields of the negatively charged interaction region, thus allowing electrons to escape in larger number than would otherwise be possible.
Chapter 6: Conclusion and future work

We have measured energetic electron acceleration in the back-reflection direction from a normal incidence, 3 mJ laser interaction with water. The spectrum averaged over many interactions shows a peak spectral-density between 500 keV and 1 MeV, with individual spectra having peak spectral-densities exceeding 1.3 MeV. These high energies cannot be explained through laser-ponderomotive scaling. The spatially integrated backward accelerated (>120 keV) charge is 300 pC/pulse, a number obtained by averaging over many interactions. The accelerated charge is measured with a Faraday cup, and is consistent with the electron spectrometer measurement if a wide spray of equal spectral quality is assumed. This total charge accelerated for the input laser energy exceeds every case documented in the literature. This work represents a major advance for the field in two ways: 1) we explored experiments with normally reflected ultra-intense laser light and 2) the process of electron acceleration appears to be significantly enhanced at normal incidence. We have delved into the mechanism using experimental and computational techniques. The experimental investigation involved an analysis of connections between pre-pulse, backscattered optical harmonics through SRS, and accelerated electron number (along with energy spectrum). The computational investigation required modeling of the pre-pulse in hydrodynamic code, then a simulation of the experiment at a variety of focal conditions to mimic our experiment. Optical harmonics were investigated and found to be a good probe of pre-plasma characteristics as well as a tool for experimental optimization. Analysis of experimental probe beam interferometry led to even greater insights as to the sensitivity of electron acceleration to pre-pulse conditions.
6.1 Discussion

I am confident in our characterization of the accelerated electrons. The electron number and energy spectrum have been carefully and directly measured in the laboratory. I am comfortable with our current theoretical understanding (summarized in Sec. 5.8), as developed through prior literature and analysis of particle trajectories in our simulations. This explanation is that a standing wave is established by the overlap of the laser and its reflection, launching electrons in both the forward and backward directions. The electrons, which may be affected by quasi-static electrostatic fields in the plasma, can be efficiently accelerated to high energy by direct laser acceleration in the reflected light. However, we have not performed direct experimental observations of the standing wave, direct laser acceleration, or quasi-static field acceleration dynamics.\(^{21}\) Rather, as in the parable of the six blind men and an elephant [79], we have experimentally probed the underlying acceleration mechanism in a variety of indirect ways. Without direct experimental evidence, why do I stand behind this theoretical explanation? Many of the salient experimental features have been successfully reproduced through simulations and matched with theoretical explanations. This convergence of experiment, simulation and theory for a previously unexplained phenomenon is a great success of the dissertation.

Well-understood experimental features, by simulation and theory:

**Normal-incidence interaction with strong backward radiation**: Matched well in all simulations with an extended pre-plasma. Normal laser incidence results in consistent overlap between incident pulse and its reflection, enabling a standing wave with oscillating electromagnetic fields exceeding the peak laser-field.

**Electrons primarily emitted in the laser specular direction**: Matched well in simulations. The forward-going radiation is suppressed by the target, and the backward radiation is enhanced through back-reflected direct laser acceleration.

\(^{21}\)The femtosecond acceleration effects clearly observable in particle trajectories of the LSP simulations would be incredibly difficult or impossible to directly observe experimentally. Experimental analysis of reflected light phase shifts such as that outlined in [56] may provide one future avenue for further detailed study.
300 pC of electrons, >300 keV, per pulse: Matched to a factor of five in single-scale length simulations, and to within factor of two after modeling the pre-pulse hydrodynamically. High reflectivity, extended pre-plasma enables standing-wave patterns and direct laser acceleration within the pre-plasma.

Electron spectral peak density near 1 MeV: In simulations, electrons are accelerated efficiently both below and above 1 MeV; spectral peak density is well below 1 MeV. The missing low-energy electrons have been understood by their preferential scattering as they leave the target and arrive at the experimental detector. Direct laser acceleration and electrostatic fields enable the high energy electron acceleration.

Emission of significant $\frac{3}{2}\omega$ and $\omega/2$ light: Matched well in simulations; evidence of plasma waves at these frequencies in the simulations. Strong SRS signal is evidence of an extended near-quarter-critical density pre-plasma.

Highest radiation when laser slightly out of focus: Matched well in the FLASH + LSP handoff simulations. Because of pre-plasma effects, focusing at the liquid surface results in a pre-plasma profile less suited for electron acceleration and a main pulse focus further from critical density.

Experimental features with partial explanations:

Multiple-scale length density profile and geometric shape: This observation of shadowgraphy and interferometry is matched by the FLASH simulations of the pre-pulse interaction. A knee feature seen in FLASH simulation as the transition from 3 micron to 8 micron scale length. The extended pre-plasma serves as a source of electrons.

High radiation dose for non-normal incidence interactions: The experimental parameter scan of Fig. 3.2 shows strong radiation dose at glancing incidence to the target. We have performed neither simulations nor experimental characterization in the glancing-incidence configuration. We speculate that this glancing-incidence radiation is due to the well studied mechanism of resonance absorption [80].

Shot-to-shot variations in blue-shift of half-omega light Fig. 3.13 shows strong shot-to-shot variation in the blue shift of the $\omega/2$ light emitted from the
plasma, with spectral peaks between 1300 nm and 1600 nm, while the average peak is at 1600 nm ($\omega/2$). The analysis of simulations does not sufficiently resolve the $\omega/2$ light to make a reliable assessment of the spectral blue-shift. While we have done no specific studies regarding the implication for pre-plasma conditions, we speculate that the shot-to-shot variation in blue-shift is due to the chaotic plasma dynamics of high-intensity laser interactions.

**Different efficiencies between 1 kHz and 10 Hz:** When changing the laser pulse repetition rate between 1 kHz and 10 Hz, similar experimental results can be obtained; however, both pre-pulse and focal depth must be adjusted. Observations within the chamber show, even with 20 Torr, an extended liquid water vapor spray extends from the target at 1 kHz; this spray likely changes the laser focal properties, modifying electron acceleration.

**Poorly understood experimental observations:**

**Similar backward radiation dose for 40 fs and 80 fs nominal pulse durations:** Experimentally increasing the distance between pulse compression gratings reduces the pulse duration (measured by the autocorrelator). Yet, the overall radiated dose is not strongly affected until pulse duration is reduced beyond $\sim$100 fs. This was not matched in simulations, which showed that increasing the pulse duration in this manner would result in reduced-efficiency electron acceleration. The experimental observation was made with the radiation dosimeter. Based on the simulations, I hypothesize that a measurement with the electron spectrometer would show reduced peak spectral density.

Some features we have better theoretical explanations for than others; and while several features are matched in simulation, some are not. I recognize these experimentally observed but yet-unexplained features as avenues for future research for deeper understanding of the electron acceleration and its experimental context. In addition to those above, I will now discuss at length simulation and experimental choices and observations through their successes and shortcomings.

Maintaining the interaction chamber at 20 Torr rather than high vacuum was experimentally necessary to prevent freezing of the flowing water-column target. Since the experiment is conducted at 20 Torr and not high vacuum, the effect of laser focus distortion (self-focusing) due to the intensity dependence of the index of refraction
was assessed. Analytically, this effect can be quantified through computing the “B-integral” for the laser pulse [81]. Using the experimental parameters, the B-integral is estimated to be less than 0.2 for the laser propagating between the OAP up to 25 μm away from the water jet target, which is approximately where the ultra-intense pulse may encounter significant pre-plasma. As a rule of thumb, self-focusing only becomes important for B-integral values of 3-5 or more [81]. Thus the effect is calculated to be small. This conclusion is supported by empirical evidence in the laboratory. The laser was allowed to propagate through various levels of background pressure, and the spectrum and beam mode after going through focus is observed. No spectral shift is apparent below 40 Torr, and no beam mode degradation is observed below 80 Torr. Self-focusing would result in beam mode degradation after focus, and so self-focusing due to background pressure can be ruled out as a relevant effect for this laser and experiment at 20 Torr.

Is there still self-focusing within the pre-plasma? Possibly, but this effect is distinct from one due to the background pressure. Rather, this effect would be due to the extended pre-plasma seen in the FLASH modeling and experimental interferometry. The LSP simulations suggest that, while apparent self-focusing occurs, the electric field enhancements near the critical density are primarily due to the standing wave between transmitted and reflected light. Our typical experimental characterizations of intensity are of in-vacuum intensity. Measurements of intensity within the pre-plasma are not possible, so we cannot easily check if this is specifically the case in our experiment. Judging on-target intensity based on in-vacuum intensity is correct if the pre-pulse is very small (and pre-plasma is negligible), and the laser is very well-focused onto the target. This first condition is not met in many HEDP experiments to-date; a laser with peak in-vacuum intensity of $10^{21}$ W cm$^{-2}$ and $10^{-6}$ : 1 contrast will make a sizeable pre-plasma, modifying the ultimate laser intensity. In this case, judging intensity on-target based on the in-vacuum intensity is wrong, as is clearly demonstrated even with the perfect laser modes simulated. The effect of pre-plasma expansion will be mitigated with high-Z materials, which expand more slowly. In any case, I believe one should verify the negligible extent of pre-plasma in an experiment through means such as interferometry before believing that a vacuum intensity is the on-target intensity. The difference between in-vacuum and in-plasma intensity is surely even worse with imperfect laser modes, and especially if a deformable mirror has been used to create the “best focus” in vacuum. This best focus in vacuum
will never be reached in our experiment; the laser will be modified in the plasma well-beforehand.

$\frac{3}{2}\omega$ plasma waves were observable within the PIC simulation space ($\frac{3}{2}\omega$ light at the boundary of the simulation space was generally not observed). Unlike with the simulations, we cannot look within the experimental plasma, but $\frac{3}{2}\omega$ light exiting the plasma is a signature of the plasma waves within. If SRS is not the primary source of electron acceleration, why is it well correlated with $\frac{3}{2}\omega$ light at the boundary of our experimental space? The answer to this question is neither theoretically nor experimentally resolved. However, I have the following hypothesis based on what I have observed in the laboratory and simulations. Making a high-quality focus at the focal plane in vacuum does not guarantee a good focus within the plasma. When a high-quality, high-intensity focus is made within the plasma, more SRS emissions will occur. Hence, making a good focus between the quarter-critical and critical density in the plasma would lead to a strong $\frac{3}{2}\omega$ signal. It would also lead to effective electron acceleration through the standing-wave and direct laser acceleration mechanisms within the plasma. Running the deformable mirror to optimize for a small, intense focal spot in vacuum is not the worst thing for electron efficiency, and we saw that high in-vacuum intensity consistently helped us accelerate large numbers of electrons. Even better, however, is running the deformable mirror to optimize the green harmonic generation, during which we were enhancing the pre-plasma scale length and making a high-quality focus within the plasma. I propose that this is why optimizing the deformable mirror for the green harmonic resulted in higher numbers of electrons accelerated.

The PIC simulations are incredibly valuable to this experimental interpretation, in that they allow us to follow the dynamics of individual electrons. One of the positive features of simulations are that they are reproducible. The simulation can be repeated as many times as desired with precisely the same energy, focal parameters, beam shape, etc. However, experiments are a muddier affair, with limits to the control and characterization of each of these parameters. Simulations have several limitations, even beyond the physical accuracy of various abstractions and computational compromises to the physics. In order to be realistic (to avoid “theoretically possible, but practically impossible” results) simulations must adapt to the experimental muddiness; they also must characterize how the plasma reacts to realistic variations in the laser input. In the experiment, for example, we “tune up” the focal
depth and pre-pulse characteristics before achieving a high efficiency electron acceleration result; could we re-create this process in the simulation? My approach to this problem involved repeating the simulation with a variety of inputs, reflecting deliberate and uncontrolled variations in the laboratory.

Within the LSP simulations of the main pulse interaction, I varied many more parameters than I had time to discuss in this dissertation. These explorations led to a confidence that I was reproducing not only the single-shot results, but the multi-shot “feel” of adjustments performed each day in the high-repetition-rate experiment. One parameter I was not able to vary is the laser mode quality; to my knowledge, there is no method to modify the laser wavefront within LSP. Laser mode quality is known to be an issue in any real experiment, because of aberrations in the laser. However, we do not simulate the mode quality of the laser in either the pre-pulse or main pulse simulations. The features as the laser comes to focus will likely be important, in that local “hot spots” can seed instabilities in the plasma. This has the potential to modify the plasma dynamics and local intensities, and modify the electron acceleration. We have seen this in the laboratory, in that a poorly focusing laser may not accelerate electrons efficiently. While the effect may be important in the laboratory (and may inhibit electron acceleration if uncontrolled), our simulations can at least demonstrate that a non-uniform laser mode is not required for efficient electron acceleration.

Particles are deflected and turned around due to the strong charge-up of the target; this effect is real, but overestimated by 2D PIC. This discrepancy was noted when analyzing the trajectories of particles in the 2D PIC simulations. Charging up of the target causes problems for interpreting these simulations in that the electrons that would be accelerated from the target are drawn back to the simulation, reducing the overall efficiency and skewing the electron spectrum in energy and angle. When compared with their 3D counterparts, electrostatic fields fall off less rapidly as a function of distance. While important to understand, we were able to work around these issues. 3D simulations of the target confirmed that the effect does not change any of our essential conclusions. For details of these 3D simulations, which are used to produce the simulation electron spectrum of Fig. 5.5, see Ngirmang et al. [67]. An additional artifact of 2D PIC is that the laser comes to focus more slowly in 2D than it would in 3D. This difference will tend to reduce the dependence of the simulation to focal depth parameter scans. The 2D LSP simulations showed strong electron
dependence to focal depth of the laser, but not as strong as in our experiment; this may be one reason.

This is the first simulation our group has done where a hydrodynamic code (FLASH) is used to model the pre-pulse interaction as an initial condition for a particle-in-cell code (LSP) simulation of the main interaction. This technique is very powerful, as both the long timescale dynamics of plasma expansion and the short timescale dynamics of high-intensity laser-plasma interaction can be modeled. While handoff simulations in general are not a novel concept, using FLASH outputs as LSP inputs is not conventional. The handoff between FLASH and LSP transferred density values; an improved handoff would transfer ionization, temperature, and other relevant parameters as well.

Moving to 3D has pros and cons, and the balance between these factors played out with my FLASH simulations. Previous simulations in our group of FLASH have been in 2D; this is the first work to endeavor to represent the 3D water-column target and show its evolution as the pre-pulse interacts with it. The simulation resolution needed to be made more coarse (0.9 \( \mu \text{m}/\text{cell} \)) in 3D than 2D in order to run on moderate-sized computer clusters. Discontinuous jumps in our equation of state tables for water tended to arrest the simulation more frequently in 3D than in 2D; this should be corrected by revisiting the resolution of these tables. Smaller scale features, such as unstable fluid flows, could not be represented in my 3D FLASH simulations. On the other hand, 3D simulations could model the water column and laser geometry accurately. While the laser and water column are each cylindrically symmetric, their axes of symmetry are different. Complete modeling of the hydrodynamic evolution of the target therefore requires 3D geometry.

A better understanding of pre-plasma conditions, provided by the FLASH simulations, has implications for future target choice. For example, a depression in the surface of a \( \mu \text{m} \)-thick flat target would evolve differently than this 30 \( \mu \text{m} \) thick water target (e.g. the surface depression might become a hole), and hence the pre-plasma and therefore the electron acceleration would be substantially modified.

### 6.2 Future work

We believe we have recreated the essential mechanisms of the laboratory in our simulations, and have a sound theoretical basis for electron acceleration through the standing-wave and direct laser acceleration mechanisms. Future analysis of FLASH
LSP handoff simulations are likely to give us even more insight into the complex plasma dynamics occurring in the laboratory; however, already we now have enough knowledge to propose to the simulation and new experimental campaigns. One idea for future experimental work will involve the controlled injection of a nanosecond timescale pre-pulse, statistical analyses of electron-beam fluctuations using single-shot acquisitions, and spatial optimization of the electron beam with adaptive optics. Another idea is that the technology of the femtosecond-probe-pulse delay could be combined with the electron source to make an electron or X-ray video of the plasma dynamics using two targets; one to create electrons/X-rays, and one to be observed.

Now that we better understand the role of pre-pulse in electron acceleration under the experimental conditions, we could improve our understanding further by making a distinction between responses of the electron spectrum to changes in laser energy vs. changes in pre-pulse energy. Figs. 3.16 and 3.14 inadvertently convolve these two effects; as laser energy is decreased from 2.9 mJ to 1.4 mJ, the pre-pulse energy is decreased by the same ratio. In the future, energy scans of the main pulse energy separately from the pre-pulse energy could be enabled by splitting the laser beam to create an artificial pre-pulse (Fig. 6.1.) The two arms of the laser could then be independently attenuated.
Figure 6.1: **Proposed artificial pre-pulse experiment.** Artificial pre-pulse injection will be achieved by splitting the main pulse in two unequal parts using a variable waveplate followed by a polarizer. The higher-energy part (main pulse) will be the polarizer transmission, and will travel a longer path than the low-energy part (pre-pulse), the polarizer reflection. The delay between the two pulses will be controlled, giving the experimenters control of the time during which the pre-plasma hydrodynamically expands. Also controlled will be the beam area, pulse duration, and energy of the pre-pulse. Based on the work in this dissertation, we expect this added control will enable high-efficiency electron acceleration without the day-to-day irreproducibility of manipulating the laser’s natural pre-pulses.

The experimental and simulation electron spectra of Figs. 3.16 and 5.5 provide an opportunity to re-examine the X-ray spectra of Figs. 3.4 and 3.5. The presence of MeV electrons at the electron spectrometer provides evidence independent to the X-ray spectra that there are > 800 keV electrons in the experiment. The presence of these electrons well outside the target region on the one hand supports the veracity of the X-ray data, and on the other hand, reinforces the electron Bremsstrahlung interpretation. However, the X-ray spectrum of Fig. 3.5 does not look like a typical Bremsstrahlung spectrum; rather than falling off at high energies, the spectrum appears to be rising up. This spectral shape may be explained by modification of the X-ray spectrum through filtering of X-rays between the source and detector. For example, the low-energy electrons and X-rays will be preferentially blocked and scattered by the metals between the X-ray detector and the target. Given the extremely narrow acceptance-angle of the X-ray detector, scattered X-rays are preferentially rejected. As a point of future work, it would be interesting to understand how well the X-ray spectrum fits quantitatively with the measured electron spectrum data. This
calculation could be performed by modeling the experimental chamber and single-hit X-ray detector geometry in *Geant4* or MCNP, while also using the measured or simulation electron spectrum as an initial condition.

It would be helpful to perform further quantitative and qualitative comparisons of the FLASH pre-pulse simulations and experimental pre-plasma observations. Unlike the main pulse interaction, which occurs on the femtosecond scale, the pre-pulse interaction can be well-characterized in our laboratory. It would be good to clamp down the FLASH simulations to our observed pre-plasma characteristics. One thing that makes this difficult is that we cannot observe the pre-plasma characteristics within the target. Also, it has been difficult to get absolute measurements of the neutral-material density from projections of disturbances to the wavefront, especially because our reference is 20 Torr rather than vacuum. Nominally, these problems in the interferometry analysis can and should be overcome.

The pre-pulse was modeled in FLASH with temporal coarseness, to accommodate limitations of a hydrodynamic code in short-pulse laser-matter interactions. In addition to a long-pulse component (as measured with our photodiode diagnostic), a high-intensity 30 fs pre-pulse arrives on target 6.2 ns before the main pulse. This short-pulse component was modeled as a 1 ns long pulse. The total energy contained in the pre-pulse was correct, but the instantaneous intensity of the pre-pulse was incorrect. This approximation results in less-steep spikes in density as a result of the pre-pulse interaction. Also, absorption can be a sensitive function of intensity. Future work should aim to make more realistic pre-pulse conditions. I even would suggest modeling the “picket” interaction in LSP. This simulation would then be a four-part simulation: FLASH for initial pre-pulse (4 ns), LSP for femtosecond pre-pulse interaction (400 fs), FLASH for more pre-pulse expansion (6.2 ns), LSP for main pulse interaction (400 fs).

### 6.3 Final thoughts

Mechanisms to accelerate ions and electrons with lasers to the highest possible energies are the subject of the most prominent research in laser-based particle acceleration (currently GeV-energy electrons are possible [24]); however, these physics experiments do not have compactness, repetition rate or ∼MeV electron acceleration efficiency as a primary concern. They utilize short-pulse lasers at their technological limits, meaning amplification stages sometimes spanning several rooms, up-to
quarter-million dollar optics, lasers that pulse from 10 Hz down to just a few times per day, and energy-per-pulse of order Joules. In configurations where electrons have been accelerated to \(\sim\text{MeV}\) with low-energy (<10 mJ) lasers, the number of electrons generated has been small (<10 pC total charge). Through this research, we show that a previously unexplored configuration enables acceleration of numerous electrons to \(>\text{MeV}\) energy and \(>1\%\) laser-to-energetic-electron conversion efficiency for electrons \(>120\text{ keV}\) (300 pC total charge, one hundred times more efficient than previously measured). This was discovered only through a measurement of X-ray and electron production in a direction that is generally not performed: directly counter-propagating to the laser as it interacts with the target. The acceleration mechanism has been explored both in the laboratory and through supercomputer simulations of the laser-plasma interaction.
Appendix A: Probe beam shadowgraphy and interferometry

The pre-pulse was essential to acceleration mechanisms, and was determined in a variety of ways. One important pre-pulse diagnostic was a laser beam that was split off of the oscillator, separately amplified, and imaged the interaction. Pre-plasma characteristics could be recovered as a function of time. One part of this analysis required significant computer programming (and collaboration with the program developers, researchers of HEDP in France) to extract pre-plasma scale length information from interferometric images. Although I did not go into great detail of the use of this diagnostic in my dissertation, it was invaluable in the day-to-day evaluation of pre-pulse conditions. I will describe its design and use in this appendix.

A.1 Shadowgraphy: Imaging the interaction

We can take a laser beam through the side of our target and look at what’s going on moments before the target interactions. For example, if the laser contrast is not very high, we will see bits of the target have started blowing off in the form of “pre-plasma.”
Figure A.1: **Steps for splitting, amplifying and delaying probe beam.** The beam path of the probe pulse, leading up to the target chamber. The steps are annotated, and each step’s motivation is described in the text.

Figure A.2: **Probe beam path within experimental chamber.** An experimental layout of the probe beam after it enters the experimental vacuum chamber. The probe beam diverges through the pre-plasma region, is re-collimated by a microscope objective, then is split and analyzed shadowgraphically and interferometrically.
Ultra-intense laser-matter interaction experiments (>10^{18} \text{ W/cm}^2) with dense targets are highly sensitive to the effect of laser “noise” (in the form of pre-pulses) preceding the main ultra-intense pulse. These system-dependent pre-pulses in the nanosecond and/or picosecond regimes are often intense enough to modify the target significantly by ionizing and forming a plasma layer in front of the target before the arrival of the main pulse. Time resolved interferometry offers a robust way to characterize the expanding plasma during this period. We have developed a novel pump-probe interferometry system for an ultra-intense laser experiment that uses two short-pulse amplifiers synchronized by one ultra-fast seed oscillator to achieve 40-femtosecond time resolution over hundreds of nanoseconds, using a variable delay line and other techniques. The first of these amplifiers acts as the pump and delivers maximal energy to the interaction region. The second amplifier is frequency shifted and then frequency doubled to generate the femtosecond probe pulse. After passing through the laser-target interaction region, the probe pulse is split and recombined in a laterally sheared Michelson interferometer. Importantly, the frequency shift in the probe allows strong plasma self-emission at the second harmonic of the pump to be filtered out, allowing plasma expansion near the critical surface and elsewhere to be clearly visible in the interferograms.

A.2 Interferometry: Probing shifts in index of refraction

To aid in the reconstruction of phase dependent imagery from fringe shifts, three separate 120° phase-shifted (temporally sheared) interferograms are acquired for each probe delay. Three-phase reconstructions of the electron densities are then inferred by Abel inversion. This interferometric system delivers precise measurements of pre-plasma expansion that can identify the condition of the target at the moment that the ultra-intense pulse arrives. Such measurements are indispensable for correlating laser pre-pulse measurements with instantaneous plasma profiles and for enabling realistic Particle-in-Cell simulations of the ultra-intense laser-matter interaction [49].
A.3 Computational analysis of interferometric images

Neutrino\textsuperscript{22} and IDEA\textsuperscript{23} were both used for interferometric analysis. IDEA is a closed-source analysis software that our group had used previously; it works only in post-analysis mode. Neutrino, on the other hand, is open-source; I identified that it could be adapted to give real-time feedback of pre-plasma parameters in the laboratory. I put in significant work to adapt the open-source code Neutrino work well at our interferometric analysis in a real-time setting.

![Phase shift reconstruction from interferometry](image)

Figure A.3: Phase shift reconstruction from interferometry. Reconstructing phase shifts from fringe shifts involves tracking the curvature of fringes. This is done through Fourier wavelet analysis in software such as IDEA and Neutrino.

Neutrino is open-source, allowing me to separate the functions from the program. I interfaced quite a bit with the developers of Neutrino, and we were able to work through some of the kinks of python scripting (and eventually the C++ source code). My first attempt at on-the-fly analysis was rather clunky and relied solely on the included python-scripting interface. Because of this, it ran outside of LabVIEW on a separate computer. The computers communicated through a shared network folder; one would take the image, the other would analyze it. Because of this complexity, it was not actually used much in the laboratory. Seeing this failure, I repackaged the interferometry analysis functions into a DLL which could be called in Windows from LabVIEW, enabling on-the-fly interferometry analysis. This is an easier-to-use implementation and allows us to assess the pre-plasma characteristic scale lengths on a day-to-day basis.

\textsuperscript{22}https://github.com/aflux/neutrino

\textsuperscript{23}http://www.optics.tugraz.at/idea/idea.html
Figure A.4: **Abel inversion of interferometry.** Shadowgraphy only gives a 2D projection of the phase interference pattern. Abel inversion is a mathematical transform that allows one to extract 3D information (the density at the center of the laser-plasma interaction) from a 2D projection, provided that the original geometry has cylindrical symmetry. In our case, we think it is a reasonable assumption of cylindrical symmetry about the laser axis, at least prior to the main pulse interaction. The pre-plasma is analyzed by performing an Abel inversion on the interferometry data, in this case with the IDEA software.

Figure A.5: **First pass at real-time interferometric analysis (shared folder).** My first attempt on-the-fly phase analysis was this dual-computer scheme. Neutrino is scripted through its python interface on a Linux-based computer, while a Microsoft Windows computer runs LabVIEW and acquires images. The two computers communicate raw and analyzed images through a shared network folder. This scheme turned out to be too burdensome to run on a daily basis.
Figure A.6: **Second pass at real-time interferometric analysis (DLL).** My second attempt at on-the-fly analysis was harder to program, but ultimately simpler. The essential source code elements were extracted from Neutrino and re-compiled into a DLL executable, called from LabVIEW in Windows. Because of its direct implementation into LabVIEW and single-computer operation, it was actually usable for day-to-day operations (the original goal).
Appendix B: X-ray diagnostics

Although the primary source of backward-going radiation in this experiment is electrons, these electrons collide with the aluminum parabola and steel chamber walls to create secondary x-rays. These x-rays were quantified in a variety of ways. This quantification is important to this dissertation in that it preceded all of the electron studies; high X-ray energies and X-ray doses led to the desire to the discovery of the primary electron radiation source. In this appendix, I will describe the X-ray diagnostics used to make these measurements.

B.1 Single-hit x-ray spectrometer

A single-hit x-ray spectrometer with a custom collimator isolated and measured the energies of backward-propagating x-rays and allowed an indirect assessment of primary backward-going electron energies. The apparatus was aligned along a line-of-sight to the target which ‘looks’ through an aluminum vacuum flange, thinned to 1.7 mm at center, and the back of the OAP. Line-of-sight x-rays therefore have possible origins within the target itself, the OAP, or the aluminum flange. The x-ray spectrometer, an Amptek X-123 configured with a stack of three 3 mm x 3 mm CdTe detectors and a digitizer [82], saturates at 800 keV. A custom collimator was designed and implemented to reduce the effective size of the detector and to shield the detector from non-line-of-sight photons. This 102 mm thick collimator was machined from 936 bearing bronze (12% Pb, 7% Sn, and 81% Cu). Placement of the collimator and detector within a steel vacuum tube provided additional shielding and permitted the collimator (itself a potential source of non-line-of-sight photons) to be positioned 1.7 m in front of the detector. In order to estimate the measurement errors due to the detection of secondaries, the laser interaction chamber and single-hit spectrometer apparatus were modeled in MCNP [83]. For 1.5 MeV mono-energetic, isotropic photon sources, MCNP predicts 1 scattered photon incident on the detector.
per 10 signal photons, a ratio which improves for lower energies. The placement of the collimator and the detector at 8.6 m and 10.3 m from the interaction region, respectively, results in a detection event rate less than one tenth of the laser repetition rate, and therefore a probability of double counts below 1%. For each x-ray spectrum, attenuation due to material in the line-of-sight was taken into account.

Figure B.1: **Single-hit X-ray detector and collimator**. A bronze X-ray collimator was designed by John Morrison to keep a strict angular tolerance on the backward-going X-rays. It is mounted in front of a single-hit X-ray detector.

**B.2 Survey meter dosimeter**

Radiation dose was measured using a Fluke Biomedical, Model 451P, Ion Chamber Survey Meter with a 25 keV low-energy detection threshold.

**B.3 GAMMA-RAD5 dosimeter**

The Amptek GAMMA-RAD5 detector is typically designed for counting single x-rays, but can also be used as a fast-response dosimeter. This was achieved by setting the bias on the PMT very low, so that a burst of many x-rays would register as one large x-rays. The more dose, the 'more energetic' the burst of x-rays. In this way, the under-biased Amptek GAMMA-RAD5 detector allows for dose measurements at
1 kHz, albeit with a rather large dose low-energy cutoff. That is, the measurement registers zero up until one makes tens of mRem/hr at 1 kHz. This diagnostic is inferior to the survey meter in that it is uncalibrated, but superior in that it can give single-shot feedback at 1 kHz (rather than once per second).
Appendix C: Laser protection and characterization

C.1 Laser alignment to normal incidence

The laser is polarized perpendicular to the water flow direction, and comes to a focus 10 to 20 μm in front of the liquid water surface as was seen by examining air breakdown at 200 Torr in shadowgraphy. All other data presented in this paper were taken at normal incidence. To ensure normal incidence, the laser was vertically squared with the water column (verified in shadowgraphy) and horizontally centered on the water column curvature (verified using a microscope objective to within 3 μm).

C.2 Pockels cells and back-reflection protection

Ultra-intense laser experiments with near-solid density targets at normal incidence present a challenge: optics in the laser can be damaged by the reflected light from the plasma critical density surface that can be amplified as it travels backward through the laser amplifier chain. To avoid damage due to normal incidence reflections, the laser system was modified by optically isolating the amplifiers using polarizers and a large-aperture Pockels cell.

A 20-mm aperture Pockels cell (FastPulse Lasermetrics 5406SC / CF1043) sandwiched between crossed polarizers was added to the laser chain, after the final amplifier and before the compressor. This actively isolates the amplifiers from back-reflected seed pulses. The sub-10 nanosecond temporal gate window of the cell ensures that the forward going pulses pass through the system with minimal loss by 90-degree polarization rotation, while the back-reflected beams arriving after >60 ns go through the cell in its ‘off’ state. This results in rejection of their unchanged polarization state by the crossed polarizer at the entrance. To further protect the laser chain from any depolarized component of back-reflected pulse leaking through
this system, all low extinction polarizers at the entrances and exits of the three manufacturer-implemented Pockels cells (two between the first and second amplifier, one between the pulse stretcher and first amplifier) were replaced with custom, high extinction, low group-velocity dispersion causing polarizers (Alpine Research Optics).

### C.3 Intensity calculation

The laser intensity was determined through a series of measurements at full amplification, with reflective ND filters in place to attenuate the beam. A 42 fs pulse duration (FWHM) was measured using a single-shot second-order autocorrelator [43] (Coherent), assuming a Gaussian temporal profile. A 2.0 μm FWHM focal spot was measured in-situ using a 20x Edmund Optics 59-878 microscope objective and a 2D Gaussian fit to the image. To calculate the energy in the laser focus, an energy meter (Coherent LabMAX-TOP / PM150-50C) was used to measure pulse energy prior the final focusing OAP, and then this number was reduced according to two sources of energy loss. First, about 45% of the energy is lost due to angular scattering from imperfections in the machine-grooved, gold-coated OAP. This loss was determined by using the laser focus to bore a ~300 μm hole in aluminum foil (a hole area much larger than the actual laser focus), then reducing the energy to avoid air breakdown and comparing energy meter values before the OAP and after the laser focus. Second, only 70% of the energy reaching the imaging apparatus appears in the Gaussian focus. This second ratio was determined by assessing the image pixel-value integration of the 2D Gaussian fit and comparing it to the integration of the entire background-reduced focal spot image. From these calculations we have found that when 5 mJ is incident on the OAP, only about 2.8 mJ reaches the focal area and 2 mJ is contained in the 2.0 μm Gaussian focus. From these numbers we obtain a peak focal intensity of $1.5 \times 10^{18}$ W/cm$^2$.

### C.4 Pulse duration and pre-pulse profile

One of the experimental variables that has been found to strongly influence results from high-intensity laser-plasma interaction experiments is the plasma scale length in front of the intended target caused by emission from the laser system preceding the main laser pulse, commonly referred to as ‘pre-pulse’ [84, 85]. To determine the
effect of pre-plasma in this experiment, the laser’s temporal profile was measured on the nanosecond, picosecond, and femtosecond scales.

**Femtosecond pulse duration**

A 42 fs pulse duration (FWHM) was measured using a single-shot second-order autocorrelator [43] (Coherent), assuming a Gaussian temporal profile.

**Nanosecond and picosecond pre-pulse measurement**

A Thorlabs DET10A photodiode and 2.5 GHz Tektronix oscilloscope, a single-shot autocorrelator [43], and a scanning third-order cross correlator [47] were all used to characterize the Red Dragon’s degree of pre-pulse. Similar to the nanosecond diagnostic outlined by [46], the photodiode was saturated to reveal low-signal nanosecond pre-pulse features. To increase dynamic range, multiple calibrated-neutral-density-filters were successively removed, with measurements made at each level of saturation. The measurements were stitched together to create a single high-dynamic-range temporal trace. The femtosecond-scale main pulse and its replicas were re-scaled according to the impulse response of the photodiode/oscilloscope system, a scaling confirmed by measurement of contrast of the amplified spontaneous emission (ASE) 600 ps prior to the main pulse with the third-order cross correlator.

**C.5 Energy measurement**

Energy is measured prior to each experiment using a standard energy meter, and is monitored during the experiment using stray light reflected from a window onto a ThorLabs DET10A photodiode.
Appendix D: Design and calibration of a high-acquisition-rate spectrometer

The electron spectrum is measured with a custom, high-acquisition-rate electron spectrometer aligned to measure electrons through the center of the OAP hole. The electron spectrometer is located 127 mm behind the target (100 mm behind the OAP).

D.1 Lanex and linear CCD detector

The spectrometer uses a slit followed by two yoked magnets to deflect electrons according to their energy onto a phosphor screen (Lanex regular) that is pasted directly upon a bare linear CCD chip (MighTex CCD camera, Toshiba 1304DG linear CCD) and made light-tight with a 25 μm thick covering of aluminum. Before measurements, a separate permanent magnet is swung into place between the OAP and electron slit (magnetically but not physically blocking the electrons’ paths) to check that recorded signal drops to zero. This ensures the measured signal is from electrons and not due to a background x-ray or optical signal. The spectrometer is triggered and acquires electron traces at ~100 Hz when not limited by exposure time.
Figure D.1: **Lanex scintillator pasting on linear CCD array.** The electron spectrometer’s detector consists of a strip of Lanex scintillator pasted onto a linear CCD array. Graphic: Drake Austin

### D.2 Relative calibration with Monte Carlo particle-scattering codes

Electron energy positions at the detector are calibrated using Hall probe magnetic field measurements combined with relativistic particle deflection calculations. The detector stack’s energy sensitivity is calibrated in two steps. First, the energy deposition efficiency into Lanex and the CCD depletion region is obtained as a function of electron input energy through MCNP [83] particle-scattering calculations incorporating the aluminum cover, Lanex and CCD. Second, the amount of light that reaches the CCD is determined from diffusion calculations performed on the light generated at different Lanex depths. The contribution from the photon and electron signals are then combined. The resultant detection efficiency curve, which is consistent with previous Lanex calibration efforts at and beyond 2 MeV [86], is presented in Fig. D.4 with a low energy cutoff at approximately 250 keV.
Figure D.2: **Lanex detector layers.** Our Lanex CCD detector has multiple layers which were modeled in MCNP. Electrons are scattered and lose energy in each layer, and the energy lost depends on the angle of incidence and electron energy. The response of the detector to an electron of a single energy is calculated by the fraction of energy absorbed by the scintillating layers. This calculation is repeated for several electron energies and angles to build a full sensitivity curve. Graphic: Drake Austin
Figure D.3: **Electrical tape test for CCD electron sensitivity.** A test was done where sections of the Lanex were taped over to completely block photons. This allowed us to verify that the relative contribution of electrons directly exciting the CCD was small, and that the use of a scintillator was therefore justified. Graphic: Drake Austin

Figure D.4: **Plot of Lanex + CCD sensitivity to electrons.** The final spectral response calculation indicates the relative influence of photons and electrons. Graphic: Drake Austin
D.3 Absolute calibration at the University of Notre Dame

We needed a robust, energy-tunable electron source as a benchmark for the new electron spectrometer. At the invitation of my undergraduate advisor (Dr. Philippe Collon) and his colleague in the Radiation Laboratory (Dr. Jay LaVerne), Drake Austin and I traveled to the University of Notre Dame to calibrate our electron spectrometer using an on-campus van de Graaff particle accelerator. This accelerator is an electrostatic van de Graaff accelerator with an acceleration energy between 1 MeV and 2.8 MeV. It can continuously put out tens of microamps of beam current at these energies and is normally used to study corrosion of materials under intense electron radiation (useful in the engineering of nuclear reactors).

![Notre Dame Radiation Laboratory MeV electron accelerator](image)

Figure D.5: **Notre Dame Radiation Laboratory MeV electron accelerator.** The experimental electron spectrometer calibration used known-energy electrons from the University of Notre Dame’s Radiation Laboratory van de Graaff accelerator. Upper left in the image are Scott Feister (left) and Drake Austin.

The experimental setup for this calibration was as follows. Electrons are electrostatically accelerated under vacuum ($10^{-7}$ Torr). The beam is collimated and steered using electromagnets to exit a duraluminum window, 3 mm thick. The beam then diverges through air 13.5 cm before entering the front slit of our electron spectrometer box, at which point a sliver of the beam is detected on either a Faraday cup or
our Lanex + CCD detector. A cross-calibration is then established between charge through the slit and counts on the CCD. After dividing counts by exposure time, and accounting for the area of the slit, we can then establish how many nC/area of charge creates one count on the Lanex + CCD detector. This we refer to as our absolute calibration of the Lanex + CCD detector.

The box is 1 inch of steel on the front and 0.5 inch of steel on the sides, top, and bottom. This slit is a 1 mm wide, 3.2 mm tall, and 1 inch deep hole in the front cover of the electron spectrometer box; the material front cover of the electron spectrometer is steel. The back of the box is left open for easy access. Directly behind the slit is placed one of two things: a simple Faraday cup to measure charge, or the line CCD + Lanex.

A 25.4 mm diameter, 6.58 mm thick bronze disc (our “Faraday cup”) is set on 16 mm-thick teflon insulator and placed ~6 mm behind the slit. Current from the disc to ground is measured using a Keithley Instruments electrometer. To ensure that our measured current is due only to electrons entering the slit (rather than those arriving through other paths), we subtract any current that remains when the entrance slit is closed with a piece of steel. This current is under 4% at 1 MeV. Since the current is measured with a flat disc rather than a cup, it loses a percentage of electrons by scattering out of its surface. The ratio of incident electrons at a given energy to lost electrons is estimated in the few-to-ten percent range and will be modeled in MCNP, as future work.

In the case of our Lanex + CCD detector, the CCD signal (counts per pixel, at all pixels) is recorded along with the CCD exposure time. Our desired signal is from the optical phosphor emissions from electrons; however, the linear CCD is also sensitive to X-rays. To see the effect of X-rays, a 1100 Gauss surface-field magnet is placed adjacent to the slit, deflecting electrons away from the slit but not affecting X-rays generated outside the enclosure. With magnet, the signal in the CCD region behind the slit matches the region behind the steel the wall, implying the signal localized to the region behind the slit is due to electrons only. (Without a magnet, the signal in the CCD region behind the slit is much higher than the region behind the steel.) X-ray signal appears as a “spiky” background and is present across all regions of the CCD (both where the slit is and where it is not). This X-ray background is averaged for each accelerator energy and subtracted from the CCD data. To avoid extra signal from background room light, the lights were turned off, and any remaining

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background was recorded and subtracted. The sensitivity of the CCD to electrons directly (rather than through optical phosphor emissions) was characterized to be very small by adding electrical tape to the phosphor (which will block light but not significantly attenuate electrons) and seeing the signal drop to nearly zero.

The slit is placed 13.5 cm from the vacuum exit port, the minimum distance possible without disturbing other experiments in the laboratory. The divergence of the electron beam due to scattering through the duraluminum exit port and 13.5 cm of air before the slit is substantial, but not relevant to this calibration due to careful setup. Specifically, the total charge and Lanex + CCD detector response are both characterized for the same sliver of charge that passes through the slit, and so angular spread before the slit does not affect our calibration. Electron energy attenuation and energy spread due to scattering air might be relevant to the calibration, but is found to be insignificant for our purposes.

The just-described absolute calibration of the Lanex + CCD detector at normal incidence is one step short of a complete calibration of the electron spectrometer, as angles of incidence onto the detector change as the electrons bend in the spectrometers magnetic field. This final step, accounting for signal change as a function of angle of incidence onto the phosphor, is calculated in MCNP. This calculation is checked at > MeV by assuming that energy will scale with the distance traveled by the electron through the phosphor layer (more phosphor interaction at greater angles).
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