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Adapting High Brightness Relativistic Electron Beams for Ultrafast Science

A dissertation submitted in partial satisfaction of the requirements for the degree Doctor of Philosophy in Physics

by

Cheyne Matthew Scoby

2013
This thesis explores the use of ultrashort bunches generated by a radiofrequency electron photoinjector driven by a femtosecond laser. Rf photoinjector technology has been developed to generate ultra high brightness beams for advanced accelerators and to drive advanced light source applications. The extremely good quality of the beams generated by this source has played a key role in the development of 4th generation light sources such as the Linac Coherent Light Source, thus opening the way to studies of materials science and biological systems with high temporal and spatial resolution. At the Pegasus Photoinjector Lab, we have developed the application of a BNL/SLAC/UCLA 1.6-cell rf photoinjector as a tool for ultrafast science in its own right. It is the aim of this work to explore the generation of ultrashort electron bunches, give descriptions of the novel ultrafast diagnostics developed to be able to characterize the electron bunch and synchronize it with a pump laser, and share some of the scientific results that were obtained with this technology at the UCLA Pegasus laboratory.

This dissertation explains the requirements of the drive laser source and de-
scribes the principles of rf photoinjector design and operation necessary to produce electron bunches with an rms longitudinal length < 100 femtoseconds containing $10^7 - 10^8$ electrons per bunch. In this condition, when the laser intensity is sufficiently high, multiphoton photoemission is demonstrated to be more efficient in terms of charge yield than single photon photoemission.

When a short laser pulse hits the cathode the resulting beam dynamics are dominated by a strong space charge driven longitudinal expansion which leads to the creation of a nearly ideal uniformly filled ellipsoidal distribution. These beam distributions are characterized by linear space charge forces and hence by high peak brightness and small transverse emittances. This regime of operation of the RF photoinjector is also termed the “blow-out regime.” When the beam charge is maintained low, ultrashort electron bunches can be obtained enabling novel applications such as single shot Femtosecond Relativistic Electron Diffraction (FRED).

High precision temporal diagnostic and synchronization techniques are integral to the use of femtosecond electron bunches for ultrafast science. An x-band rf streak camera provides measurements of the longitudinal profiles of sub-ps electron bunches. Spatial encoded electro-optic timestamping is developed to overcome the inherent rf-laser synchronization errors in rf photoinjectors.

The ultrafast electron beams generated with the RF photoenjector are employed in pump-probe experiments wherein a target is illuminated with an intense pump laser to induce a transient behavior in the sample. FRED is used to study the melting of gold after heating with an intense femtosecond laser pulse. In a first experiment we study the process by taking different single-shot diffraction patterns at varying delays between the pump an probe beams. In a second experiment a variation of the technique is employed using the rf streak camera to
time-stretch the beam after it has diffraction from the sample in order to capture the full melting dynamics in a single shot.

Finally, relativistic ultrashort electron bunches are used as a probe of plasma dynamics in electron radiography/shadowgraphy experiments. This technique is used to study photoemission with intense laser pulses and the evolution of electromagnetic fields in a photoinduced dense plasma. This experiment is also performed in two different modes: one where different pictures are acquired at different time delays, and the other where a single streak image is used to obtain visualization of the propagation electromagnetic fields with an unprecedented 35 femtosecond resolution.
The dissertation of Cheyne Matthew Scoby is approved.

Benjamin Joel Schwartz

James Rosenzweig

Jianwei Miao

Pietro Musumeci, Committee Chair

University of California, Los Angeles
2013
To Erika,

for her friendship, patience, strong editing skills, and unconditional support.
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In elementary school, I already wanted to be a scientist. My three favorite books were the Red Cross First Aid Manual, a tattered old Schaum’s Outline of General Chemistry, and a copy of the Space Shuttle Operator’s Handbook. I was inspired by watching rocket launches from Vandenberg AFB with my grandfather, who was an electrical engineer and worked on avionics systems (guidance and control) for a number of space launch vehicles, including the Titan IV. Years later, my stepfather Mark (who has engineered mechanical designs for many space systems) taught me to fly R/C gliders and even design a balsawood framed plane for my high school physics class. I hope that I have done them proud.

Graduate school has been an amazing experience and a time of great personal and intellectual growth. I had the honor of being Prof. Pietro Musumeci’s first Ph.D. student - I thank him for his patience, and for teaching me the value of “getting drunk every night.” When I first walked into his office at the end of my first year of graduate school, I felt like a New York banker stepping into the Wild West. I learned how to ride well enough to keep up with the rest of the cowboys, and I’ve come to discover that science is full of them.

I appreciate Dr. Ren-Kai Li for being an ever-gracious sounding board for my ideas. He is a great scientist, and I am lucky to have worked with him. His presence in the lab always cheered me up when I knew a late night of experiments lay ahead.

I am grateful to my comrade in arms, Josh Moody, who taught me to solder to MIL-SPEC standards. More importantly, he convinced me that I was one of the good ones (though I was never quite sure what that meant). His help has done more for my intact mental health post-dottorato-di-ricerca than he probably
realizes, so I may owe him a drink or two at the Cove Bar in Ariel’s Grotto.

The UCLA Particle Beam Physics Lab is a strange mix: part-fraternity and part-world class research laboratory. From the Starcraft II tournaments to the frequent group barbecues interspersed with inspiring journal club meetings and conferences, I was always glad to spend time with the PBPL Marauders.

Thanks to Dr. Gabe Marcus who was never too busy to offer encouragement during what seemed like the bleakest hours. I somehow lost our bet by several months despite my best attempts to delay his thesis work.

I owe David “Bronze-like Lustre” Schiller much for sharing his expertise on sailing, climbing, and computers. We’ll always have 800 Degrees. I have great memories sailing with Oliver Williams who can always feel welcome to file a float plan with me before his trips to Catalina. Thanks to the O’Sheas for their O’Shenanigans and to Andrey Knyazik for being a kind friend.

Pegasus lab has transcended its infancy, and is now host to the next generation of students: Joe Duris was my gym, gym, gym partner - I am glad we didn’t die on our first rock climbing trip. Hoss To and Evan Threlkeld were my SC2 coaches. Thanks to them, and to David, Kevin, and Emma for their help finishing up the final experiments. I’m happy to be leaving Pegasus lab in their capable hands.

Finally, I thank my entire family, whom I have seen far too infrequently during this past year. (Sorry, Mom!) Above all, I thank Erika, my wife, whom I met in graduate school my first year. I’m glad that I was able to convince her to go out with me. Erika has been completely selfless throughout the writing of this thesis and has been there for me as I’ve gone through both triumph and defeat, both joy and depression in completing my doctorate. Thank you for bearing all the lonely nights and husbandless weekends - I’ll make it up to you, I swear!

Endeavor to persevere.
Vita

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Publications


C.M. Scoby, R.K. Li, and P. Musumeci. “Effect of an ultrafast laser induced
plasma on a relativistic electron beam to determine temporal overlap in pump-probe experiments,” Accepted to Ultramicroscopy (2012)


CHAPTER 1

Introduction

1.1 The reasons to study ultrafast science

There are two established scale frontiers in science - that of time and of space. It is a continuing challenge to study systems that are ever smaller and which exhibit dynamics that occur at shorter timescales. The term “ultrafast” has come to mean any process that involves time intervals less than 1 nanosecond. There is typically a correlation in the hierarchy of structural dynamics processes between structure size and the timescale [1]. For example, a size/speed hierarchy might be: (1) the motion of electrons around an atom, (2) rotational and vibrational modes of free diatomic molecules, (3) coherent vibrational motion of atoms in crystalline solids (phonons), and (4) folding and unfolding of large organic molecules, like proteins. As the system under study becomes smaller, so does the relevant timescale of that system’s dynamics.

In general, ultrafast science can encompass very broad topics such as materials science (e.g., melting [2], phonon-driven dynamics like martensitic phase transitions in iron [3] or visualizing direct lattice motion [4]), biology [5, 6], chemistry [7], and plasma physics [8]. The timescales of such dynamical systems can vary widely depending on the mechanism for energy transfer and the response of the system. For example, a typical thermal phase transition such as melting in gold can occur on a timescale less than 10 picoseconds (1 ps = 10^{-12} s). In contrast,
a non-thermal phase transition mechanism like melting of a semiconductor can take place in a few hundred femtoseconds (1 fs = 10^{-15} s) [9, 10, 11]. Excitation of material surfaces with intense sub-100-fs optical pulses creates a highly non-equilibrium state in the electronic system of a material, bridging the study of plasma and condensed matter physics.

Understanding these dynamics at the ultrafast, ultrasmall frontier helps one understand the interactions that are fundamental to nature, even at the macroscopic scale. The current focus in the ultrafast science community is on processes that occur on the sub-picosecond timescale. Technology continues to be developed to create femtosecond optical (laser) pulses, pulsed x-ray beams, and
bunched electron beams which can act as probes to study sub-picosecond dynamics. Continuing to develop widely accessible ultrafast methods is the key to furthering ultrafast science.

1.2 Femtosecond pump-probe techniques

The pump-probe approach to ultrafast study is as follows; we induce some change in the system under study (pump), and make an observation (probe). The difficult part is doing it fast. The temporal length of the transient probe determines the “shutter speed” in these types of experiments. If we want to capture the motion of molecules, atoms, or rapidly evolving plasmas, the probe must occur at sub-picosecond time scales. To put the relevant time scales in perspective, 100 femtoseconds is approximately the time it takes for an atom to move 1 Angstrom, a water molecule to vibrate about 10 times, or half the time required for the photoisomerization of a rhodopsin molecule to occur (the fastest photochemical reaction that has been directly studied [12]).

Electronic triggering and fast electronic detectors have made nanosecond measurements mainstream, but purely electronic methods are not fast enough to access the ultrafast regime. The most important technology that has expanded the ultrafast timescale boundary has been the development of the ultrafast laser. For a historical review, see e.g. the papers by Brabec et al., 2000 [13] and Corkum et al., 2007 [14]. In the last two decades, state-of-the-art laser systems advanced from producing several nanosecond pulses to < 20 fs pulses.

Laser mode locking triggered the development of ultrashort laser pulses. The mode locking process requires a laser gain medium with very broadband laser transitions (high gain bandwidth), as well as a method for loss modulation in
Chirped pulse amplification (CPA) of an ultrafast laser pulse

Color represents wavelength

Figure 1.2: Chirped Pulse Amplification. To avoid damage to optical components by a very intense laser pulse, the pulse is temporally *chirped* (intentional dispersion of frequency components in time). After amplification the pulse is compressed by applying a negative dispersion.

The cavity that preferentially amplifies the shortest most intense pulses. It is possible to obtain modelocking using both active (high frequency acousto-optic modulation) and passive (saturable absorbers or Kerr lensing based) modulators. In modern titanium:sapphire (Ti:Sa) lasers, Kerr lensing is a popular choice and ideal for stable cavity operation [15].

Non-linear effects from ultrashort laser pulses limit the peak intensity that a laser amplifier can achieve without deteriorating the laser pulse quality or damaging intra-cavity optics. Chirped pulse amplification was the key breakthrough that enabled overcoming these intensity-driven problems [16]. By first chirping the laser pulse with a diffraction grating stretcher, different frequency components of the pulse become temporally spread out, and the instantaneous optical intensity drops dramatically. The result is that each spectral component receives
energy from the broadband gain medium, but the peak intensity stays below the cavity damage threshold. After amplification, a second grating compresses the chirped pulse, generating a very high intensity and once again ultrashort pulse.

Today < 30 fs laser systems are commercially available with sufficient energy per pulse to incite ultrafast dynamics in a variety of samples [17]. As a result, the femtosecond laser has become the vanguard pump tool in ultrafast pump-probe experiments. Hard x-rays and terahertz pulses have been proposed as more exotic pumps that could be used to drive specific transitions.

While the femtosecond laser has pretty much cornered the market on ultrafast pumps, three distinct types of probes have produced interesting material science results and driven research into ultrafast phenomena:

1. the laser-probe technique [18, 19, 20, 21, 22, 23, 24]
2. the x-ray-probe technique [25, 26]
3. the electron-probe technique [2, 27, 28].

The information obtained by these different types of probes is, in most cases, complimentary – no single technique can completely replace the others. In this thesis we will focus on the electron probes, but it is useful to initially include optical and x-ray probes in the discussion.

Due to the wide availability of commercial ultrafast laser systems the first and most common pump-probe method to investigate structural dynamics has been optical ultrafast spectroscopy, to wit the wavelength resolved measurement of reflectivity or transmissivity changes in a sample as a function of pump-probe delay [29]. This method can provide indirect information about structural dynamics, since laser photons can probe the electronic band structure of the material which
depends on the underlying crystal structures. The disadvantage is that the reconstruction of the structural dynamics is model-dependent. Another method is femtosecond laser interferometry which can provide direct insight into surface structural dynamics, like the evolution of surface plasmons, with single wavelength spatial resolution [30], but with the caveat that the probe is limited to measuring surface interactions only.

On the other hand, using lasers as probes has a few drawbacks which limit the applicability of laser-only techniques. For example visible or infrared light sources have wavelengths that are hundreds of nanometers and cannot offer atomic scale spatial resolution. To truly resolve the motion of individual atoms, one must turn to sub-angstrom wavelength probes.

The x-ray source seems to provide a solution to the requirement of high spatial resolution, as with Angstrom-size wavelength they can resolve the individual positions of the atoms in a sample. Femtosecond x-ray sources do exist (e.g., x-ray free electron lasers), they are still considered a fairly exotic source requiring large scale facilities and limited accessibility versus conventional x-ray sources. Furthermore, the interaction of light with matter has a very small cross section, so x-ray experiments require a very high intensity (photon flux) and sensitive detection to compensate for this effect.

Electrons with energy in the 30−100 keV range (corresponding to a De Broglie wavelength of 7.0−3.7 picometers) have been used in what is commonly known as ultrafast electron diffraction technique (UED) wherein an ultrashort laser pulse deposits energy in a sample and the electrons strobe at variable delays the relaxation pathways of the system. Because of the higher scattering cross-section and the relatively more compact size versus x-ray diffraction sources, UED offers complementary information to x-ray based structure analysis techniques.
and has enabled a number of breakthrough studies in the last ten years. In the
next section we offer a quantitative comparison of x-ray versus electron diffraction
techniques for structural dynamics applications.

1.3 Comparison of x-ray and electron sources for structural dynamics

In this section we compare three primary classes of probes: Sub-relativistic elec-
trons (here defined as $E < 1$ MeV), relativistic electrons ($E \geq 1$ MeV), and
X-FEL generated x-rays. A more detailed review is offered in Ref. [31].

With the advent of the x-ray FEL in the last 5 years these high-brightness
x-ray sources have received much attention for the quality and large scope of
ultrafast science performed. Notable examples of XFELs include SLAC LCLS,
DESY FLASH, FERMI@Elettra in Trieste, Italy, and SACLA in Japan. Indeed,
there are many aspects of the x-ray probe scheme that make it a desirable probe
including the very short pulse length, high spatial coherence enabling single-shot
diffusive imaging, and Angstrom-size wavelength.

It is undeniable that ultrashort, high-brightness x-ray pulses are an invalu-
able tool for ultrafast science but they have the disadvantage that the technology
of producing the probe x-ray beam is extremely complex [32, 33]. The stand-
out example is the Stanford Linear Accelerator Center’s Linac Coherent Light
Source (SLAC LCLS) which is a more than 1 km long facility featuring dozens
of electromagnets, magnetic undulators, bunchers, rf linacs, x-ray optics, and
laser synchronization systems [34]. The required footprint for such a machine
is seen in Figure 1.3. In every case, the world’s high brightness x-ray sources
are national laboratory-scale user facilities; access under these conditions is very
Figure 1.3: Schematic showing the expansive facility required to produce an X-FEL pulse. The scientific results produced by such a machine are irreplaceable, but even as the number of next generation light source facilities blossoms worldwide, their complexity and scale restricts the availability.

limited. Accordingly, there is a need for easily accessible small laboratory (or table-top) schemes for generating ultrafast probes.

There are several labs around the world developing and using compact electron sources for ultrafast science [35]. DC guns produce a laser-generated electron beam with an energy in the 10–100 keV range, and will be grouped in the following discussion as sub-relativistic sources. Rf guns employ stronger accelerating fields and produce electron bunches with an energy of a few MeV. The rf structure adds complexity and cost to the probe source but the higher energy offers a few advantages.

In general, photoinjector sources have orders of magnitude lower cost and complexity compared to ultrafast x-ray sources. Laser-generated electron probe beams offer the approach most amenable to ultrafast studies in a small univer-
Figure 1.4: Not quite table-top. This is the Pegasus accelerator system, capable of producing, manipulating, and characterizing sub-100 femtosecond high brightness electron bunches.

Since femtosecond pulsed lasers are the preferred method for ultrafast pumping, any ultrafast research lab will already have access to this type of system; the laser can serve double-duty and generate ultrashort electron beams from compact photo-electron sources.

Laser-pump/electron-probe experiments have produced significant ultrafast science measurements in complement to x-ray probe techniques as is clear for many types of studies, for example ultrafast electron diffraction (UED) [2, 36] and electron-plasma radiography [8, 37]. In Table 1.1 we summarize the main differences between these various probes.

**Particles per pulse and minimum pulse duration:** X-FEL technology has vastly shortened the minimum pulse duration achievable by x-ray probes
Figure 1.5: The applications of the various ultrafast sources discussed in this chapter are complementary; there is overlap, but no single technique can be used for all types of measurements. The arrows show trends in attributes between each source. See Table 1.1 for details.
Table 1.1: A comparison of important parameters in high-brightness ultrafast probes. X-ray free electron lasers such as the SLAC LCLS are the current front runners in minimum pulse duration and coherence. In this table, “particle” refers to either electrons or photons. Adapted from Ref. [31].

without compromising the source brightness, providing up to $10^{13}$ photons in a few femtosecond pulse. Once the x-ray pulse is generated it can be propagated for great distances without pulse lengthening.

Conventional UED setups employ non-relativistic 30-100 keV photogun electron sources. Because electrons have electric charge they tend to repel one another. As more charge is added the self fields of the beam increase; this induces bunch lengthening, emittance growth, and degraded brightness [38].

Non-relativistic electron pulses suffer from severe bunch lengthening in the propagation region after the cathode due to the strong space charge repulsion at these low energies [39, 40]. This effect seriously degrades the temporal resolution available at the sample and significant efforts have been recently directed towards improving the temporal resolution to the sub-ps level [28, 41]. Ultimately, researchers have been able to achieve sub-ps resolutions only by significantly reducing the number of electrons (down
to $10^4$ particles per pulse if compression schemes are not used) with the compromise of integrating over multiple shots to collect a single diffraction image (see information on the *stroboscopic pump-probe mode* in the next section.

Due to relativistic effects, the space charge forces are strongly suppressed for an MeV electron beam. Figure 1.6 shows the large range of bunch charge that can be accessed in an rf gun while maintaining a sub-ps bunch length. This allows the generation of electron pulses intense enough to capture in a single shot the structural information of a sample. This ability of the rf gun opens the way to the study of irreversible ultrafast processes [28, 35, 42]. Even in this case, one diffraction pattern yields only one data point in the time-series and it is still necessary to scan the time delay between the pump and the probe beams in order to reconstruct the full time-history of the process.

The maximum charge that can be achieved from conventional DC guns while maintaining sub-ps length is $10^4 e^-/pulse$. By using an rf accelerating cavity to rf compress the beam after the gun, this figure has recently been increased to more than $10^6$ electrons per bunch. These results generated much interest in the ultrafast electron diffraction community since they enable single shot mode data acquisition [43, 44]. Sub-ps length beams with up to $10^8 e^-/pulse$ have been demonstrated from rf guns. A redesigned rf gun has been proposed that would accelerate as well as compress the photo-beam [45].

**Interaction cross section:** The interaction of x-ray beams with matter is set by classical Thomson scattering from the electrons in the material. For a single incident x-ray, $\sigma_X = (8\pi/3) r_e^2 \approx 6.6 \times 10^{-25}$ cm$^2$ where $r_e =$
Figure 1.6: Measured bunch length vs. bunch charge for an MeV electron bunch generated with the Pegasus rf gun.

\[ \frac{1}{4\pi\varepsilon_0} \frac{e^2}{m_e c^2} = 2.81 \times 10^{-15} \text{ m} \] is the classical electron radius. Generally this describes the interaction of an electromagnetic wave scattering with an electron, which does not depend on the wavelength of the incident wave. On the other hand, an electron beam interacting with matter will experience Coulomb forces and undergo classical Rutherford scattering, for which the differential cross section has the form 

\[ \frac{d\sigma}{d\Omega} = \left( \frac{Ze^2}{pv} \right)^2 / \sin^4 \left( \frac{\theta}{2} \right). \]

After inclusions of the proper screening factor, this cross section integrates to be \( > 10^{-20} \text{ cm}^2 \).

The enormous five orders of magnitude difference in cross-section for the two physical processes is what makes electron probe studies attractive in

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many cases, like the study of surface phenomena, of the bulk structure of thin foils or of the molecular structure in gas phase where the low number of potential scattering sites along the probe particle path (gas particle density is much less than solid state densities) would not provide a sufficient signal to noise ratio if x-rays were used as the probe. Probing with $10^7$ electrons per pulse is, in terms of the expected signal-to-noise, equivalent to probing with $10^{12}$ x-ray photons per shot, a peak flux accessible to only a handful of x-ray users at one of the few existing 4th generation light sources.

**Penetration depth:** An immediate consequence of the increased interaction cross section is a decrease in the mean free path of the probe electrons versus x-rays. Electrons are ideal for probing thin films; this is useful if thick bulk sample cannot be made (e.g. it is difficult to crystallize certain proteins more than a few molecular layers thick). However, the low penetration depth inhibits using electrons for in situ measurements of liquid solutions.

On a positive note, there is generally an overlap between the penetration depth of energetic electrons and the absorption depth of energy from a femtosecond optical pulse. This leads to uniformity of the pumped region of the sample and therefore higher sensitivity to small structural changes. With x-rays there is always the problem to match the pumped thickness with the probed thickness [46].

**Particles needed per image:** The beam charge is set by the requirement of having enough scattered electrons to be able to detect the Bragg peaks. Using DC electron guns for UED studies on a 20 nm thick sample, it typically takes between 100 – 1000 shots to scatter enough electrons to form a resolvable diffraction pattern. The milestone time-resolved non-relativistic
UED results on Al melting have been obtained using 150 shots of 6000 electrons each, that is a total of 1 million electrons per image [3]. This is the minimum charge per bunch that is required to obtain a high contrast diffraction pattern. For an x-ray source this number is usually $10^{11}$ photons per pulse or larger due to the lower cross-section.

**Relative spatial coherence:** If we expect to resolve diffraction angles $\theta_B$, then the rms spread of angles internal to the beam distribution $\sigma_\theta$ must satisfy the relation $\sigma_\theta << \theta_B$. The spatial coherence length is defined in terms of the uncorrelated transverse angular spread of the beam\(^1\) ($\sigma_\theta$) and the probe wavelength $\lambda$ as

$$L_c = \frac{\lambda}{2\pi \sigma_\theta}.$$  \hspace{1cm} (1.1)

The primary achievements of FELs were flux and most importantly coherence (both spatial and temporal). Indeed, the new X-ray FEL sources will provide almost full coherence over the beam diameter at X-rays energies, resulting in huge coherence lengths that can be microns for micron sized beams. The coherence length generally refers to the size of the area that can be probed and preserve information about the sample. The application dictates the required coherence length; diffraction from crystal structures requires only coherence on the order of the lattice spacing (0.1 - 10 nm) while to perform coherent diffractive image reconstruction [47] of a large object (e.g. a virus [5]) may require a few microns coherence length. The coherence length of x-ray sources is still vastly superior to that obtained

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\(^1\)This is due to the photoemission mechanism which introduces an intrinsic spread in the distribution of angles of the beam electrons at the cathode, the so-called thermal emittance.
from any ultrafast electron source, and is in excess of 50% of the x-ray beam spot size [48].

This is a major achievement of such a technology which will be difficult to match with other tools. This being said, it is important to discuss what level of coherence is actually needed and for which scientific problem. Low energy pulsed electron guns have been estimated to have beams with a coherence length in the order of 3 – 8 angstroms. This value is enough for crystallographic studies, as typical interatomic distances are in that order of magnitude; however, even in systems with much longer superstructures low energy electron diffraction delivers good quality patterns.

1.4 Ultrafast science with femtosecond relativistic electron beams

Pump-probe experiments can be performed in a variety of modes:

- The *stroboscopic mode* entails sending many pulses (up to several thousand) through the sample for a given pump-probe delay. A single data frame is built up from many shots (e.g., see Figure 3 in Ref. [49]) because the brightness is not high enough to resolve the interaction in only a single pulse. This type of data collection relies on repeatability of the dynamic process over a large number of shots.

- The *single shot mode* is utilized when the probe beam is bright enough to record the desired information with a single pulse. Each subsequent shot is taken at a slightly different pump probe delay. These “frames” are analyzed in the proper order to reconstruct the full time history of the dynamic process under study. Since multiple shots of the same delay are
unnecessary, one may study irreversible processes. This mode requires high brightness with a short beam which presents the problem of creating and maintaining a very high current beam against the effects of space charge expansion [50].

• Finally, the streaked mode enables the researcher to capture the entire time history in a single pump-probe event. Here, a streak camera or other instrument is required to time-resolve the probe after it interacts with the sample. This method has the advantage that the temporal resolution no longer depends on the probe temporal length, but rather on the resolving power of the streaking element.

There are significant challenges that must be addressed to expand the scope of successful pump-probe experiments with electron probes. This thesis will address them, as listed below.

1. *Rf photogun injection encompasses a very large and complex parameter space.* At Pegasus laboratory we have adapted a high brightness rf gun to produce ultrashort relativistic electron beams. Strong coupling between different effects, viz. rf fields and space charge, make it challenging to obtain an analytical formula for the beam bunch length as a function of all the input parameters of the photoinjector; these parameters include rf phase and amplitude, solenoid settings, laser pulse duration, spot size and intensity. Instead one relies on computational methods to track the evolution of electrons through the accelerator.

   In practice, there is a trade between brightness and bunch length. An ongoing goal has been to understand and optimize drive laser and rf gun injection parameters to adapt the beam to a variety of ultrafast studies,
but the input parameters are not entirely unique distributions. The earliest ultrafast beams were generated operating the gun in the “blowout regime,” so as to produce dynamically optimized beams that exhibit only linear self-forces [51] and prevent space charge induced emittance growth. Since then we have discovered other modes of rf gun operation appropriate to a wider range of femtoseconds studies, including single-shot experiments enabled by an rf deflector as a streaking element.

2. **Single-shot and streak modes require high detection efficiency.** In DC gun setups micro-channel plate (MCP) detectors provide high sensitivity and good spatial resolution for imaging keV electrons. Though there are less scattering events per shot versus rf gun sources, the use of an optimized MCP setup approaching every-electron detection allows DC guns to obtain individual frames in single-shot [52]. Our group tested the MCP detection scheme with a relativistic electron beam. In a later series of studies we found that an optimized fluorescent screen coupled to an intensified CCD camera through a high speed (that is, large aperture) lens can provide every-electron detection for MeV beams better than the MCP which suffered from smearing effects [53].

3. **Large temporal jitter between rf and laser systems prevent synchronization of the pump and probe beams.** Inherently, this degrades the temporal resolution of stroboscopic and single-shot mode studies (but, does not affect streaked mode studies). DC gun sources generally do not have the timing jitter problem\(^2\) since the stability of the DC power supplies used to accelerate the particles can be better than one part in \(10^5\), but with the addition of rf compressor elements jitter must once again be taken into

\(^2\)Although they must still solve the pump-probe time zero problem [54].
account [44]. Jitter between the electron and the laser beams at the Pegasus photoinjector has been measured to be around 1 ps [55]. A solution to the jitter problem is presented by the adaptation of electro-optic sampling based timestamping to achieve non-destructive electron beam timing. In Chapter 4 we present results making a proof of principle pump-probe timestamped measurement with 200 femtosecond resolution.

In this thesis ultrashort relativistic electron beams are applied to study two different problems: First we illustrate a materials science application of this machine by exploring the ultrafast melting of single crystal gold through femtosecond relativistic electron diffraction (FRED) [56, 57]. Second, we study the creation and evolution of a laser-induced plasma using electron shadowgraphy, following high-fluence laser photoemission and subsequent ablation of a metal target with temporal resolution better than 35 femtoseconds [58].

1.5 Organization of this dissertation

Each of the following chapters constitutes a different experiment. While the physics of the experiments varies widely, they are unified in that they all enrich, enable, and/or demonstrate the study of ultrafast phenomena in an rf photoinjector laboratory.

Chapter 1 gives background on the motivation for studying ultrafast systems in this thesis.

Chapter 2 presents the rf photoinjector as a tool to study materials through Femtosecond Relativistic Electron Diffraction (FRED). The rf deflecting cavity is introduced to compare single-shot scanning versus single-shot complete history acquisition for FRED.
Chapter 3 makes a slight diversion to address the electron-laser timing problem. We introduce electro-optic sampling as a method to timestamp pump-probe events and lower timing uncertainty.

Chapter 4 discusses a robust plasma shadowgraphy based method for determining time zero in pump-probe experiments.

Chapter 5 extends the utility of plasma shadowgraphy to dense plasma radiography - we achieve a sub-35-fs measurement of the ultrafast evolution of a plasma. Here, the plasma deflects beam electrons, forming a shadow in the beam profile. We compare streak mode acquisition with single shot scanning, using electro-optic timestamping to correlate pump and probe to better than 200 fs rms.

Chapter 6 concludes the thesis, and looks forward to the next steps planned to expand the Pegasus photoinjector laboratory’s ultrafast pump-probe capabilities.
CHAPTER 2

Femtosecond Relativistic Electron Diffraction

In this chapter, we describe two experiments using ultrafast electron diffraction to study the femtosecond laser induced melting of gold films.

Firstly, we report the experimental demonstration of time-resolved relativistic electron diffraction. Single shot diffraction patterns from a single crystal gold sample were recorded using ultrashort 3.5 MeV electron bunches from a radio frequency photoinjector. By scanning the pump pulse time-delay, we studied the Bragg peaks amplitude change due to the laser-induced melting of the sample. The observed time scale matches the one predicted using a simple two temperature model of the heating of the thin foil. Time-resolved relativistic electron diffraction using mega-electron-volt (MeV) electron beams with $10^7$ particles in 100 fs bunch length opens exciting possibilities in ultrafast structural dynamics [57].

Secondly, we report on the experimental demonstration of using relativistic electron diffraction and an rf deflecting cavity to capture in a single shot the entire time-history of the ultrafast laser-induced heating and melting of a single crystal gold sample. By recording the time variation in the Bragg peaks on the streak image of a 16 ps long electron beam it is possible to reconstruct with 400 fs temporal resolution the evolution of the sample structure induced by a 35 mJ/cm$^2$, 400 nm laser pump pulse [56].
2.1 Background

2.1.1 A UED case study

Ultrafast electron diffraction (UED) of laser-induced dynamics was first performed in 1984 by Williamson, Mourou, and Li [59]. In this experiment they studied the laser-induced melting of polycrystalline aluminum. The pump laser pulse was provided by a modelocked Nd:YAG laser (center wavelength 1064 nm), with a maximum fluence of 13 mJ/cm$^2$. The laser pulse length of 20 ps limited the temporal resolution of the experiment as the timescale of melting is 10–15 ps.
The advancements of femtosecond laser technology, specifically the proliferation of modelocked titanium sapphire (Ti:Sa) laser systems, allowed researchers to probe structural phase transitions (for example, melting in aluminum) with higher temporal sensitivity. Results from a laser-pump/laser-probe type (i.e., purely optical) study of the laser-induced change of reflectivity in polycrystalline aluminum with femtosecond time resolution suggested a non-thermal time-scale in the evolution of this transition [19] and prompted further investigations which led to a modern reprisal of the classic UED experiment in 2004 by Siwick, et al. [49].

In Siwick’s UED experiment, electrons directly probed atomic positions with a breakthrough sub-picosecond resolution. Therefore, by delaying the laser-pump with respect to the electron-probe, the researchers measured the delay-dependent diffraction pattern, and from it calculated a temporal history of the structure factor of the material. The result without doubt demonstrated that the melting of aluminum takes place when the lattice temperature increases due to the electron-phonon coupling - a process that occurs on thermal time-scales.

This pair of studies clearly demonstrates the need for complementary ultrafast techniques. Both the laser-probe and electron-probe experiments reveal multiple aspects of the same underlying material dynamics (i.e., the ultrafast onset of the aluminum liquid phase). Without structural information, the electronic disordering probed by the laser-probe experiment would indicate that the melting occurs faster than can be explained by thermal heat transfer for the photoexcited electrons to the atomic lattice (electron-phonon coupling). By studying the atomic structure through UED, it is evident that the lattice melting occurs on a longer timescale than indicated by the optical reflectivity experiment. In fact, the Siwick experiment matches the prediction by the thermal two-temperature model
In the last 5 years femtosecond relativistic electron diffraction (FRED, a.k.a. MeV UED) has developed into a valid alternative to conventional non-relativistic electron diffraction for ultrafast structural dynamics, enabling single-shot studies using more electrons per pulse than the sub-relativistic UED sources. Worldwide MeV UED efforts have been increasing with two labs in the United States at UCLA and Brookhaven National Laboratory, one in China at Tsinghua University, and one at Osaka University in Japan [60, 61, 62].

One of the great drives with all UED efforts, MeV UED included, is to push the temporal resolution to the < 10 fs limits achievable by next generation light sources. The typical pump-probe style experiments suffer from a variety of contri-
butions to timing [63] which need to be overcome to have successful time-resolved diffraction studies. One of the major problems is the synchronization between the pump and the probe. To this end we have developed electro-optic sampling as a timestamping method and demonstrated data resorting with better than 200 fs precision. The subject of timing drives the discussion in Chapters 3 and 4.

2.1.2 Streaked MeV UED

Even before Williamson and Mourou published their seminal 1984 paper on UED [59], they recognized the possibility of an alternative setup in which the diffracted electron beam is streaked using deflecting plates to record the time evolution of the changing pattern [64]. The advantage in such configuration is that a short electron bunch is no longer required and the temporal resolution is determined by the sweeping speed of the plates or more generally by the resolution of the streak camera system.

Recently, there has been renewed interest in streak camera based UED in the context of rf photoinjector based MeV UED [65, 66]. Using an rf resonant cavity for the deflection allows for higher deflecting voltages which together with a relatively high oscillation frequency offer faster sweeping speeds than what is possible with traditional ramped voltage sweeping plates.

In the second part of this chapter we demonstrate the reconstruction of the entire time-history of an ultrafast structural process using a single continuously time-resolved diffraction pattern. The test process we study is the ultrafast laser induced heating and melting of a single crystal 20 nm thick Au foil.
2.2 Theory

We begin this section with a general introduction to electron diffraction. We then discuss the structure factor and how it can be calculated and used to determine atomic positions in solid state samples. Finally, we discuss the physics of ultrafast melting in gold, introducing the two-temperature model to quantify phonon-electron coupling as a mechanism for heat dissipation following pumping by an intense femtosecond laser pulse.

2.2.1 History and theoretical overview of electron diffraction

Inspired by the work of Einstein on the apparent duality of light waves and photons (particles), De Broglie first expounded the concept of a matter wave in his 1924 Ph.D. thesis\(^1\). Here he described the motion of particles in a way analogous to the wave mechanics of light propagation. An important result was the postulation of a fixed relationship between the particle’s momentum \( p = \gamma mc\beta \), and its De Broglie wavelength \( \lambda \), satisfied by

\[
\lambda = \frac{h}{p} = \frac{h}{mc\beta} \sqrt{1 - \beta^2}, \tag{2.1}
\]

where \( h \) is the Planck constant. With this definition of wavelength for a matter wave, researchers could confirm the hypothesis of particle-wave duality through experiments with the properties of wave motion, for example, interference.

The interference of x-rays in a crystalline material, i.e. x-ray diffraction, had already been observed in 1912 by Max von Laue and his colleagues\(^2\). Following

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1\(^{\text{Nobel Prize in Physics, 1929.}}\)
2\(^{\text{Nobel Prize in Physics, 1914.}}\)
this discovery, x-ray crystallography was pioneered by W.H.\(^3\) and W.L. Bragg (father and son!) who were able to quantify the crystal lattice of the specimen under study from the observed diffraction pattern\(^4\).

In terms of the spacing of the crystal lattice planes, Bragg’s law is deceptively simple

\[
n\lambda = 2d\sin\theta.
\] (2.2)

The interplane distance \(d\) in general depends on the crystal structure of the target material. For example, the familiar spacing for a cubic lattice (examples of which include bulk aluminum and gold metals) in terms of the nearest neighbor distance \(a\) is

\[
d = \frac{a}{\sqrt{h^2 + k^2 + l^2}}, \quad \{h, k, l\} \in \mathbb{Z} \geq 0
\] (2.3)

Davisson and Germer (Bell Labs employees) confirmed the wave nature of electrons when they observed diffraction from an electron beam striking a nickel target\(^5\). In fact, the electron diffraction pattern was found to be equivalent to that obtained from x-ray diffraction, obeying the Bragg law with \(\lambda\) given by Eqn. (2.1).

By going from keV to MeV electrons, one must pay the price that the scattering angles \(\theta\) associated with the same measured scattering vector \(s\) becomes small as the probe wavelength decreases per Equation 2.1. Increasing the energy of the beam, we observe the scattering angle for a given Bragg peak decrease toward the unscattered (direct) beam as shown in Figure 2.3. As our group has

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\(^3\)W.H. Bragg’s Ph.D. advisor was J.J. Thomson. Thomson won the Nobel Prize in Physics, 1906 for discovering the electron.

\(^4\)Father and son shared the Nobel Prize in Physics, 1915.

\(^5\)Nobel Prize in Physics, 1937.
Figure 2.3: Bragg peak dependence on beam energy (wavelength). Here we scan the beam energy by changing the rf gun gradient. This is the change in the (111) peak of a polycrystalline aluminum foil studied in an early UED experiment at the Pegasus lab [60].

Experimentally verified, this small angle scatter makes it practically impossible to study thick samples with reflection mode diffraction. We must instead rely on keeping the sample thin enough (as required by the multiple scattering discussion of Section 2.2.2) to perform transmission mode diffraction. Even so, the scattering angles are small enough that we require a large distance (a few meters) to allow the diffracted particle to separate from the un-diffracted (direct) beam. This is essential to ensure proper spatial resolution and separation of peaks of the diffraction pattern.
2.2.2 Scattering cross sections

In Chapter 1 we have already generally discussed the cross section in terms of the Rutherford scattering cross section. While the basic electromagnetic interaction is the same, the relativistic beam experiences a different effective cross section when interacting with an atom due to the relativistic screening function.

For relativistic charged particles interacting with atomic potentials, it is necessary to calculate the Born cross section for elastic scattering $\sigma_{el}$ based on density $n_0$ and atomic number $Z$ [67]. Once calculated the relativistic elastic cross section allows one to estimate the elastic mean free path for a given material, $\Lambda = (n_0 \sigma_{el})^{-1}$. Incidentally for MeV electrons the inelastic scattering contribution is negligible compared to the effect of multiple elastic scattering events [68].

For example, using the analytic screening function developed by Salvat, et al. to calculate the elastic mean free path in solid atomic gold for 3.5 MeV electrons, one finds $\Lambda_{Au} = 40$ nm. If the thickness of the material is very large compared to the mean free path, multiple scattering tends to randomize and blur the diffraction angles. This effect becomes more pronounced as $Z$, $n_0$, and the sample thickness increase. Figure 2.4 shows the difference in multiple scattering probabilities for 200 nm gold versus 50 nm gold. The majority of the electrons for the thinner sample experience single scattering, while the thick foil signal is dominated by multiple scattering. By contrast, even a 200 nm carbon film has the largest probability for zero scattering events. Typically for samples much thicker than $5 - 10$ times elastic mean free path, the scattering angles becomes so large that the resulting pattern is too diffuse to measure and all structural information gathered by the probe beam is lost.
Figure 2.4: The number of scattering events for MeV electrons through solid materials is well explained by Poisson statistics. As the material thickness increases, the probability for multiple scattering increases and the diffraction pattern degrades.

2.2.3 Detection Efficiency

Before work done at Pegasus lab in the past few years, very little was known about 100% efficient detection of MeV electrons. A supporting technology that was required for all the subsequent measurements is an optimized imaging detector. The need for high sensitivity to a small number of electrons led us to explore active detectors.

In conventional non-relativistic electron diffraction, micro-channel plates (MCP) are used to directly image keV electrons or amplify the flux of low energy electrons inside an image-intensifier. The amplified electron flux is then converted by
a scintillator to visible photons which are subsequently fiber-optically coupled to a high efficiency charge-coupled device (CCD) camera. It is relatively straightforward to achieve single-electron detection capability due to the large gain of the MCP and the high light collection efficiency of the fiber-optics coupling.

We have tested an MCP for MeV electrons and we obtained high quality single-shot diffraction patterns. Blurring of the patterns was observed as a result of the large penetration depth of MeV electrons and the resulting excitation of secondary electrons in many surrounding micro-channels. It was also found that due to the active amplification process the signal from the MCP has larger fluctuations which may be a concern in single-shot measurements where very small changes in the pattern are to be detected [69].

An effective alternative for the detection of MeV electrons is the use of optimized passive scintillator screens which are low cost and provide high electron-to-photon conversion efficiency and improved spatial resolution. We decided to use a high yield phosphor screen coupled to an intensified CCD with a large numerical aperture lens to increase collection efficiency. A phosphor screen yields as many as a few thousand photons for each MeV electron due to the large penetration depth of MeV electrons. Considering an energy loss rate of \(1.2 - 1.5 \text{ MeV cm}^2/\text{g}\) for \(1 - 4 \text{ MeV}\) electrons and a screen density of \(34 \text{ mg/cm}^2\), the total energy deposition by each electron is approximately \(E_{\text{loss}} = 50 \text{ keV}\). For an optimal choice of phosphor material and screen composition, the efficiency in conversion of this energy into output visible photons is on the order of \(\eta = 15\% - 25\%\). Approximately half of these photons will exit from the screen side facing the CCD camera while roughly the other half exits from the back side. Since the photon spectrum is narrowly peaked at \(h\nu = 2.27 \text{ eV (545 nm)}\), we have \(n_{\text{scr}} = (1/2) E_{\text{loss}} \eta/h\nu = 1.7 - 2.8 \times 10^3\) as an estimate of the number of photons.
emitted from each side of the screen per incident MeV electron. With a properly
designed lens coupling system whose collection efficiency is higher than 1% and a
state-of-the-art CCD camera capable of single-photon detection, single-electron
imaging is possible. This was demonstrated in a recent paper where up to the
(800) diffraction spot was detected in a single shot [53].

2.2.4 Lattice dynamics

A significant advantage of electron diffraction experiments over x-ray probe ex-
periments is that it is possible to measure directly nuclear positions (recall that
electrons undergo Rutherford scattering) and quantify the collective transient be-
behavior of a material in response to optical excitation. Specifically, it is possible to
track the loss of an ordered state in a crystalline material with the collective (rms)
motion of its constituent atoms and the diffraction pattern can be interpreted to
extract lattice structure motion.

In response to energy being deposited in a gold single crystal lattice by a
laser pulse, for example, long range ordering in the crystal breaks down due to
complex electron-lattice coupling dynamics. During this strongly driven phase
transition in which the lattice vibrates itself apart, a liquid structure arises with
only local correlation between atoms. Experimentally, this transition is known
to be on the order of a few picoseconds for lattice excitation with femtosecond
laser pulses [49].

2.2.5 The Two Temperature Model

In order to understand the time scales associated with the heating and melting of
a metallic crystal, it is important to follow the path of the energy deposited in the
sample by the laser. Most of the laser energy is absorbed within an optical skin
depth from the illuminated surface. A distribution of fast electrons is created and quickly moves across the thin sample at Fermi velocities. At the same time, due to the strong electron-electron scattering, the electron distribution thermalizes quickly so that < 100 fs after illumination it is possible to define an electron temperature which is homogeneous across the sample.

At this point, the energy in the electron system relaxes to the lattice modes. Due to the isotropy of the cubic crystal structure, one can assume that the energy deposited by the laser on a gold sample excites a thermal distribution of the lattice vibrations allowing the definition of a lattice temperature and the description of the relaxation process with a classical two temperature model (TTM) [70]. We note that in principle the electron subsystem could be coupled strongly with a subset of the phonon excitation modes. This approximation is valid as long as the probed area is within a homogeneously pumped region, and the crystal lattice motion is not strongly anisotropic. This coupling is described simply with a set of coupled equations (in 1-D, neglecting spatial dependencies):

\[ C_e(T_e) \frac{dT_e}{dt} = -\gamma_e (T_e - T_l) + S(t) \]

\[ C_l(T_e) \frac{dT_l}{dt} = \gamma_e (T_e - T_l) . \]

According to Eqn. 2.4, \( C_e \) and \( C_l \) are the electron and lattice sub-system heat capacities, respectively; \( \gamma_e \) is the electron-phonon coupling constant; and \( S(t) \) is the driving (source) term describing how the electron sub-system absorbs energy from the pump-laser. In extreme cases both \( C_e \) and \( \gamma_e \) can be functions of \( T_e \) [71]. For this reason, the linear TTM as presented here is valid only for lower laser fluence values used in this experiment. As seen in Figure 2.5, a laser pulse
Figure 2.5: Two-temperature model for a copper surface illuminated by a 100 fs, 10 mJ/cm² titanium:sapphire laser pulse. With 10 mJ/cm² fluence can heat electrons to $T_e \sim 1700$ K before re-equilibrating with the atomic lattice.

The source term $S(t)$ is highly dependent on the optical penetration depth (<10 nm from the surface) as well as the range of the ballistic electrons. As long as the sample is thinner than the ballistic electron range (estimated to be $>100$ nm after 20 fs) the pump laser pulse has uniformly pumped the entire sample thickness with respect to the timescale of the interaction of the probe beam. In this context, a uniform (not dependent on $z$, i.e. the axis normal to the crystal surface) TTM holds, and one can apply Equations 2.4. For thicker samples, one needs to consider energy transport by adding $z$- and radial dependences to the TTM and solving the associated system of partial differential equations.
2.3 Experiment

An example of a typical pump-probe UED setup that we perform at Pegasus is as follows. We use a 20 nm thick gold single crystal, oriented with the (001) crystal axis parallel to the beam axis as the diffraction target. A Ti:sapphire laser system produces a 2 mJ 800 nm 40 fs FWHM laser pulse which is split in a pump and a photocathode driver line. By exploiting the large yield from multiphoton photoemission\(^6\) \cite{72}, only < 25% of the initial laser power is needed to generate the electron beam at the cathode. The > 75 MV/m accelerating gradient in the rf gun quickly accelerates the beam to 3.5 MeV, where the effect of the beam self-fields is strongly reduced. After the gun, the solenoid is used to minimize the beam divergence. Then, the particle distribution is emittance-filtered by a collimating pinhole of 1 mm diameter placed just in front of the sample at 0.8 m from the cathode.

A smaller propagation distance from the cathode would allow reaching shorter bunch lengths at the sample by providing less drift space for space charge to expand the beam, but the need for various beamline elements (gun solenoid, gate valve, injection laser mirror box, detection screens, EOS crystal and time-of-arrival screen) prevent placing the sample closer than 60-70 cm from the cathode.

The collimating pinhole aperture just before the sample plane significantly improves the spatial resolution and signal-to-noise ratio in the diffraction pattern. The dimensions of the hole are optimized in order to let \(10^7\) particles through thus preserving the ability to record the diffraction pattern in a single shot. The pinhole provides a few important functions.

Without the collimator, the dimensions of the probe beam depend on the beam dynamics and are sensitive to many operating parameters. By using a

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\(^6\)See Appendix A
fixed aperture the probe area has a well-defined size easing matching to the pump beam and alignment. During operation, collimation has the significant advantage of removing the dark current background that constitutes a large source of noise in UED patterns.

Most importantly, the collimating aperture effectively removes the high-emittance particles from the beam, thereby cleaning up the transverse phase space. This provides a significant improvement in the spatial resolution. The aperture selects the central part of the beam with the lowest divergence and also to control the size of the probed region [73]. Just before the hole, the beam normalized rms emittance is 0.7 mm-mrad (mostly due to cathode thermal emittance) and the rms size is 2 mm, from which we obtain an rms beam divergence < 50 µrad. The transmitted beam has ∼ 1 pC of charge, an rms transverse size of 0.25 mm and an rms normalized emittance < 0.075 mm-mrad. The electron bunch length is measured with an X-band rf deflector located at 1.8 m from the cathode to be < 200 fs. Particle tracking simulations using the General Particle Tracer (GPT) show in Figure 2.7 that the beam is still slightly expanding as it propagates so the measurement is only an upper limit on the bunch length at earlier points along the beamline.

In order to couple better to the interband absorption edge in gold, we double the frequency of the pump laser pulse (to give a pump wavelength of 400 nm and a photon energy of 3.1 eV) [74]. Accordingly, we can pump with less laser fluence, and mitigate the risk of optical damage of the sample. The spatial overlap between the pump and the probe is obtained using a fluorescent screen sensitive to both laser and electron beam that can be inserted in the beamline at the sample plane. The pump laser beam is fired with the same 1 Hz repetition rate

7http://www.pulsar.nl/gpt/
Figure 2.6: The undeflected UED setup scheme.
Figure 2.7: RMS bunch length as a function of longitudinal position. Increasing the bunch charge significantly lengthens the bunch. Increasing the rf gun gradient can more quickly accelerate the beam and somewhat mitigate this space charge induced broadening.

of the rf gun and focused on the sample to a spot 1.5 mm in diameter to ensure uniform excitation over the 1 mm diameter region probed by the electrons. A slightly off-axis mirror allows nearly normal (< 5 degree) incidence for the pump beam. The maximum fluence on the sample is 35 mJ/cm$^2$. This can be reduced with variable attenuators. A small portion of the pump laser beam is picked up by a 1% beam splitter and sent to a virtual target screen monitored by a feedback loop. This maintains the alignment at the sample within a 100 µm tolerance.

While a single pump event is non-destructive to the sample, we chose to use a motorized $x - y$ translation stage to move the sample to a fresh spot between laser shots. We recorded the diffraction pattern (DP) using a phosphor screen located at 1.1 m from the sample.
Figure 2.8: The experimental diffraction pattern from a (001) oriented single crystal gold film. Up to 20 diffraction peaks are visible in a single shot, limited only by the size of the beamline aperture. (Lower left quadrant) When the laser is turned on, the peaks fade due to the Debye-Waller effect, and a liquid diffraction ring appears as the sample melts.

2.4 Results and Analysis

2.4.1 Single-Shot mode result

Up to twenty Bragg peaks can be identified in a typical DP image (see Fig. 2.8) taken in a single shot on our beamline. The cubic crystal is oriented with a crystal axis along the incident beam axis so that the projection of the pattern onto the plane is a square lattice, indexed by \((nm0)\). For each diffraction spot,
we use a two-dimensional Gaussian fit to calculate its amplitude, the center position and the width. By translating the sample in the absence of the pump laser, we observe some variation (mostly in the relative amplitude) of the Bragg peaks which is due to the imperfections in the single crystal sample. To take into account such non-uniformity of the sample, each laser-on shot is compared with a laser-off diffraction pattern taken immediately prior to the laser illumination. For a given sample position, the variation in the normalized amplitude for each Bragg peak is less than 1%. This can be seen by the stability of the trace in Fig. 2.9 for the time delays when the laser arrives after the electron beam.

As the pump delay arm is scanned, the peak intensity drops suddenly when there is an overlap between the laser pump and the electron probe beams. The diminishing of the Bragg peak intensity in this case indicates a decrease in the structure factor indicative of the disordering of the crystal lattice. As the atoms gain energy, the disorder increases until a liquid phase forms. As the liquid state atoms spread out, they tend to space at a mean interatomic distance that is sampled by the probe beam. The result is the onset of an intensity ring in the diffraction pattern as shown in the lower left quadrant of Fig. 2.8.

2.4.2 Melting metals with femtosecond laser pulses

The increase in the lattice temperature (described by the TTM of Section 2.2.5) is the explanation for the time-dependent intensity variation in the diffraction peaks. The amplitude of the Bragg peaks is in fact very sensitive to the lattice temperature through the Debye-Waller factor which depends on the rms amplitude of the atomic motion. In Figures 2.9 and 2.14 one can notice that the (220) peak streak disappears faster than the (200) peak. This is consistent with the higher order diffraction spots being more sensitive to the lattice temperature.
Figure 2.9: Temporal scan of single shot dynamic UED of single crystal gold pumped with a femtosecond laser. (top) As the temperature of the lattice increases, the scattered intensity drops per the Debye-Waller effect. The lattice temperature is calculated with the two temperature model. (middle) Increasing the fluence drives the optical energy absorption to saturation. (bottom) The onset of a liquid peak in the pattern coincides with the lattice losing long range order.
increase than the lower order ones in agreement with the scattering vector dependence in the Debye-Waller factor. In general, this factor multiplies the peak intensity, and has the form

\[ I_{\text{peak}} = I_0 e^{-2M(T)} \]  

(2.5)

with \( M(T) \propto \bar{u}(T) s^2 \). The temperature dependence is captured in \( \bar{u}(T) \), which is the rms vibrational amplitude of the lattice planes. This amplitude can be determined for a specific crystal geometry using the Debye model. The strong \( s \) (scattering vector magnitude) dependence tends to make this effect more noticeable for the higher order (large \( s \)) peaks [75].

A prediction of the Bragg peaks amplitude using the TTM and the Debye-Waller factor is also shown. The experimentally extracted electron-phonon coupling constant \( \gamma_e = 0.15 \) is consistent with the value found in the literature [76]. As the temperature goes above the melting threshold we observe a departure from the Debye-Waller prediction as long range order disappears. The solid begins to melt and the crystalline structure is lost. The Bragg peak structure in the diffraction patterns completely vanishes 10 – 15 ps after the laser hits the sample.

2.5 Streaked Mode UED

2.5.1 Experimental modifications for streaked mode UED

The experimental setup for streaked UED is very similar to the one already described. However in this adaptation, we make a long drive electron beam. After interacting with the sample, this long beam is sent into an rf cavity before
impinging on the detector, positioned 1 m downstream of the diffraction target. In the rf cavity, a mode similar to the TM110 of a cylindrical pillbox imparts a transverse momentum kick to the electron beam. The arrival time of the center of the electron bunch in the cavity is synchronized with the time when the rf oscillating electric field changes sign (zero crossing). For bunch lengths shorter than 1/4 period of the rf sine wave, the momentum transferred by the field to the particles varies monotonically with time, and the leading front of the electron pulse is deflected in the opposite direction than the beam tail. Consequently in the following drift space, the beam gradually elongates along the coordinate corresponding to the deflection axis of the cavity. The temporal structure of the beam thus becomes correlated with one of its transverse coordinates and can be imaged on a simple fluorescent screen inserted in the beam path. Considering as an example a vertically deflecting cavity positioned after a diffraction target and phased at the 0° (180°) zero crossing, particles that probe the sample at an earlier time will hit the top (bottom) of the screen. The image formed on the detector will contain the temporal evolution along one transverse coordinate with the diffraction information encoded in the other dimension. In other words, the streak camera technique enables the deflecting mode yielding the entire history of the ultrafast process under study in a single time resolved DP.

The electron bunch length defines the width of the temporal window probed by the electrons and should in principle be maximized. On the other hand, if the electron bunch is longer than \( \sim 40° \) of rf phase, the nonlinearities associated with the curvature of the rf wave in the deflecting cavity have to be taken into account and the conversion from the observed vertical beam image on the screen to the temporal dimension is less straightforward. The maximum allowable temporal width then depends on the rf frequency used to drive the deflecting cavity. As an example, for an X-band structure resonating at 9.6 GHz, the bunch length
should be maintained less than 15 ps.

The temporal resolution depends on the sweeping speed of the deflecting voltage and not on the bunch length as long as this one remains smaller than the maximum allowable temporal width defined above. In the thin lens model, the proportionality factor (transverse kick term) between $t$ and $y$ on the screen located at a distance $L$ from the cavity center is given by \cite{77}

$$K = \frac{eV_0}{m_0c\gamma} \quad (2.6)$$

where $c$ is the speed of light, $e_0$ and $m_0$ are the electron charge and mass, and $\gamma$ is its Lorentz factor. Inputting the Pegasus parameter for a screen distance of $L = 15$ cm, and a 4 MeV beam energy, with deflector settings of a 500 kV deflecting voltage and a 9.6 GHz rf frequency, $K \sim 1$ $\mu$m/fs and a 2 ps long beam stretches to 2 mm in its vertical dimension. The vertical rms beam size observed on a downstream screen can be approximately (exactly for Gaussian distributions) written as the quadrature sum of the contribution due to the deflection and the minimum rms spot size ($\sigma_0$) achieved when the cavity voltage is off,

$$\sigma_y = \sqrt{\sigma_0^2 + (K\sigma_z/c)^2} \quad (2.7)$$

taking $\sigma_z$ to be the rms bunch length at the deflector plane. In order to resolve the dynamics of the structural change imprinted on the bunch temporal profile from the vertical streak of the beam as seen at the screen, we require that the contribution to the vertical spot size due to the $t$-correlated deflection largely exceeds $\sigma_0$. Minimization of $\sigma_0$ can be achieved by collimation or by using a vertically focusing quadrupole before the deflecting cavity. As was the case here, a 180 $\mu$m spot size prior to the deflection corresponds to a $< 400$ fs temporal
resolution. With more rf power available (larger $K$) and a better focusing system (smaller $\sigma_0$), the temporal resolution of this technique could theoretically be developed to $< 40$ fs.

Since generating an ultrashort bunch is no longer a requirement of the electron gun, the number of particles in the beam can be increased by orders of magnitude with respect to the present configuration. By increasing the laser energy illuminating the cathode, rf photoinjectors can produce up to $10^{10}$ particles in 15 ps bunch length providing sufficient signal to characterize an ultrafast process with a single pulse.

With respect to the first experimental setup, the electron beam is made much longer by making a long laser pulse. The temporal laser pulse shaping is achieved using five a-BBO crystals of thicknesses 1 mm, 2 mm, 4 mm, 8 mm, and 16 mm, respectively. When a short linearly polarized laser pulse is sent in such birefringent crystal with principal axis oriented at 45 with respect of the laser polarization vector, the two polarization components travel with different velocities and the output pulse is split with a temporal separation proportional to the crystal length [78]. For a 1 mm long crystal, the output temporal spacing is 0.5 ps. By repeating this process using $n$ crystals of increasing thickness it is possible to generate a long train of $2n$ equally spaced laser pulses with alternating polarization. If such laser pulse train is sent on a cathode, each of the electron beamlets generated will expand under the action of its own space charge force and eventually a relatively flat top electron beam distribution (see Fig. 2.10) is obtained [79]. This is a compact scheme to directly control and shape the longitudinal profile of the electron beam.

Note that in Figure 2.10 that the electron beamlets generated by a varying number of a-BBOs are not equidistant. By choosing the injection phase on the
Figure 2.10: The stacked BBO technique yields a beam that approximates a long flattop distribution. The beamlets can be smoothed by adjusting the rf gun injection phase, and thus changing the bunching factor.

cathode to be near 45 degrees, the bunch train compression in the rf photogun is minimized but a residual phase-dependent bunching effect still occurs and causes a deformation of the train longitudinal profile. Also the intensity in each beamlet is not constant along the macropulse due to the time-varying electric field on the cathode surface inducing an rf phase-dependent lowering of the work-function for the photoemission process (Schottky effect) resulting in a not perfectly flat-top beam distribution.

2.5.2 Streaked mode result

The DP image shown in the right half of Fig. 2.12 is obtained by turning on the streaking voltage in the rf cavity. Due to the kick imparted by the rf fields,
the different Bragg peaks stretch in the vertical dimension. Depending on the
transverse dimension of the Bragg peaks and on the crystal axes orientation it
might be possible that different spots overlap when vertically streaked. The
measurements described here were taken on samples where the angle of the (200)
peak was between 15° and 30° above the horizontal. This orientation makes it
possible to clearly distinguish at least one of the (200) and (220) peaks time-
streaks.

The phase of the rf cavity is set so that particles that arrive at the front (tail)
of the bunch are deflected downward (upward), i.e., the direction of the time axis
is pointing up in the picture. The measured length of the beam streak on the
screen is 10 mm, in agreement with our expectation for a 16 ps long electron
beam.

Time-resolving the diffraction pattern in the absence of a pump laser is use-
ful to demonstrate that the electron beam (or the beam generated radiation)
does not cause any significant change or damage to the crystal structure. Any
beam-induced structural change would appear in this picture as a time-dependent
diffraction signal.

It is important to compare the dimensions of the Bragg spots on the rf
deflector-off image with the length of the streak with the deflector-on. The finite
vertical dimension of the unstreaked spot in fact spoils the perfect correlation
between vertical position and time induced by the rf cavity, limiting the tempo-
ral resolution achievable with the technique. This is an important point as by
improving the beam quality (lowering the beam emittance), it would be possible
to obtain a narrower diffraction spot and significantly better temporal resolution
even without any increase in the sweeping speed. In the left half of Fig. 2.12
the rms vertical size of the unstreaked Bragg peaks is ∼ 250 µm which yields a
Figure 2.11: The deflected UED setup scheme.
Figure 2.12: A static (unpumped state) diffraction pattern of (001) oriented single crystal gold. In the center of the figure we compare the line profile of the unstreaked (red) and streaked (blue) Bragg peaks.

In order to test the capability of the technique to resolve the structural change in the sample, we perturb the system by illuminating the crystal with an intense (35 mJ/cm$^2$) ultrashort 400 nm pump laser pulse. In Fig. 2.13 we show the single shot streak images of the diffraction pattern with the laser off (right) and with the laser on (left). In the case where no pump laser is used, the length of the Bragg peaks streaks is comparable to the length of the un-diffracted electron beam. It is immediately clear that when the pump laser is turned on, the streaks of the Bragg peaks are significantly shortened.

A more quantitative picture is obtained by horizontally slicing the diffraction pattern. The temporal length of each slice is set to 400 fs which is the resolution limit set by the dimensions of the unstreaked beam. Using the unstreaked
diffraction image as a reference for the relative position of the spots, we subdivide each Bragg peak streak into $\sim 40$ slices 250 $\mu$m wide and 400 fs long. We then fit the amplitude, position, and width of the Bragg peak in each slice (i.e., for each time sample point) with a Gaussian function.

We normalize each Bragg peak slice amplitude with the corresponding intensity of the direct (un-diffracted) peak. We then compare the laser-on data sets with the laser-off to obtain the time-evolution of the sample after the laser excitation. The observation time-window is limited by the length of the electron pulse to 16 ps. The determination of the time zero, which is the instant when the
Figure 2.14: Deflected mode UED on a single crystal gold film. The intensity of the Bragg peaks is normalized to the diffraction pattern obtained from an un-pumped sample. The decay is fit well by the TTM, but for the used fluence of 35 mJ/cm$^2$, the sample is clearly in a saturation state.

laser hits the sample and the ultrafast process starts, is obtained by observing at what point the streak begins to change from the un-pumped case. This is an example of a process-dependent time zero determination. Chapter 4 describes a sample-independent synchronization method that can determine time zero with 150 fs precision.

2.5.3 Conclusions on streaked mode diffraction

While the temporal resolution of the streaked mode technique in this first demonstration is comparable to the one that can be achieved with conventional UED
(due to lack of detail at finer timescales), the significant novelty is in the possibility of obtaining the entire history of the ultrafast structural evolution in one continuously time-resolved electron beam shot. Furthermore, there is a clear path toward improving the temporal resolution of continuously time-resolved MeV UED technique. The resolution depends critically on the unstreaked transverse beam size and hence minimizing the beam emittance could further improve the smallest temporal features that can be resolved in the streak. This can be achieved for example by replacing the 1-mm-diameter collimator currently used with a smaller one to select only the particles with the lowest possible divergence and emittance. One only needs to maintain the transmission through the hole high enough to allow the recording of the entire time-history of the process in a single shot.

The extension of this technique to non-relativistic electron sources is an interesting possibility. In this regard, it is important to notice that if one is looking for single shot data acquisition, the number of particles per interval of temporal resolution has to be relatively large ($> 10^6$) to be able to resolve each diffraction peak for different time slices. Since we have removed the requirement of having an ultrashort bunch length, a much greater charge can be used in a single pulse allowing a significant increase in the peak current (i.e., the number of particles per unit time). On the other hand, a limitation comes for larger peak currents, as the transverse beam quality (i.e., normalized emittance) especially for lower beam energy, starts to degrade quickly. A different approach in non-relativistic electron diffraction systems, would be to use the rf deflector, and minimize the synchronization jitter in order to be able to integrate over multiple shots. The advantages in this case would be a faster acquisition time (no time-delay scans are needed) and a better resolution (limited by the jitter of the beam with respect to the rf phase).
We demonstrated for the first time the feasibility of observing the time evolution of an ultrafast process in a single shot using an rf deflecting cavity in conjunction with an intense relativistic electron beam. As an example, the application of this technique to the ultrafast heating of a single crystal gold sample allows a single shot determination of the electron-phonon coupling constant. With further optimization including a more efficient detector, a stronger deflecting field, and increased electron beam brightness, rf deflector based MeV UED has the potential to yield sub-10 fs temporal resolution with single shot acquisition capabilities.
CHAPTER 3

Electro-Optic Sampling for Ultrafast Timing

3.1 Background and Motivation

3.1.1 Timing uncertainty in rf photoinjectors

Ultrafast science in modern accelerator facilities relies on femtosecond (fs) laser technology. The laser typically serves a dual purpose of generating an electron beam from a photocathode as well as optically pumping energy into the system under study. Such pump-probe experiments often require precise synchronization between the ultrashort bunched electron beam (bunch) and a laser pump pulse.

Radiofrequency (rf) photoinjectors hold an impressive precedent for producing ultrashort bunches, due in part to an accelerating electric field that is typically an order of magnitude stronger than attainable from DC sources. When driven with an ultrashort laser pulse, these electron beam sources can generate bunches shorter than 30 fs with exceptional brightness and beam quality [80]. Bunch lengths of less than 5 fs rms are theoretically possible with magnetic or rf bunch compression [81].

This ultrashort bunch length comes at a cost - induced synchronization errors between the photoelectron bunch and the laser. By comparison fluctuations in a DC photoinjector arise from instability of the high voltage power supply, and 30 – 100 keV DC guns tend to be stable to 1 part in $10^{-4} - 10^{-5}$ (so that the
beam time-of-arrival would jitter by only 30 femtoseconds after a 1 meter drift). In rf gradient guns and rf compressors, random fluctuations in rf signal phase and amplitude fed into the gun will transform into time-of-flight jitter for the photoelectron bunch, and increase uncertainty in the relative timing in bunch-laser interactions up to a few hundred femtoseconds.

This rf-laser jitter problem manifests itself in all ultrafast electron beam applications, even when used as a drive beam in 4th generation light sources; the electron beam timing jitter impresses itself on the time of arrival of the x-ray laser pulse [82]. If FEL users hope to correlate the results of pump-probe experiments to pump delay, then it is important to either control/reduce the rf jitter or determine the relative timing between the bunch (and hence the FEL radiation) and the pump laser. Our approach to the jitter problem consists of measuring as accurately as possible the timing difference, then using this information to post-process the data, rather than trying to actively control and minimize the jitter contributions. Our derived figure for the total laser-beam jitter is relatively high compared, for example, to the LCLS jitter figure [83], however the amplitude of the jitter is unimportant in the post-processing scheme described above.

Electro-optic sampling (EOS) techniques provide an ideal solution to the problem of determining bunch-laser timing jitter. In EOS schemes the electron bunch electric field is sensed by an electro-optically active medium and is imprinted on an ultrashort probe laser pulse as a cross-correlation with the laser pulse. The probe pulse length is analogous to a shutter speed in detecting the electric fields.

### 3.1.2 A review of successful EO timestamping experiments

Ultrafast EOS was first performed by Auston and Valdmanis to sense optically induced fields in electro optic materials [84, 85]. Later, Wu and Zhang revived
the idea to characterize free space THz pulses produced by optical rectification [86, 87].

The first demonstration that EOS could be applied to non-destructively sensing and profiling relativistic ultrashort electron bunches at FELIX FEL [88]. The next year, EOS was used to directly measure the time profile of beam-induced wakefields in beamline walls [89]. Since then, EOS methods have been applied to directly measure the bunch longitudinal electric near-field profile [90], as well as temporally characterize terahertz coherent transition radiation in a laser-plasma accelerator [91].

Cavalieri, et al. [92] made the first demonstration of EO time-stamping at the Short Pulse Photon Source (SPPS) beamline at the Stanford Linear Accelerator Center. The EOS timing measurements were corroborated with simultaneous measurements from a fast x-ray diagnostic. After this benchmarking study, the group successfully applied the technique to time-stamp multi-shot pump-probe x-ray diffraction data tracking the ultrafast bond softening in bismuth crystals [93], and later applied it to diffuse x-ray scattering measurements of nucleation dynamics in indium antimonide [94]. In both cases, the time-stamping technique achieved temporal resolution better than 100 fs, close to the limit set by the x-ray pulse length. Thus, EO time-stamping evolved from proof of concept to a useful temporal diagnostic for ultrafast science [32].

3.1.3 Extending EO timestamping to low-charge beams

The high charge beams measured with EOS at these facilities were obtained from post-acceleration of the initial few MeV beams produced at the beginning of the accelerator chain. At Pegasus we had to solve the issues with electro-optic sampling stemming from the very low energy of the electron beam (3 – 4 MeV),
and from the fact that in order to generate very short electron pulses at this energy, the charge needs to be maintained less than 10 pC.

<table>
<thead>
<tr>
<th>Source</th>
<th>Charge, $Q_{\text{beam}}$</th>
<th>Energy, $\gamma mc^2$</th>
<th>Rel. EOS modulation, $\Gamma$</th>
</tr>
</thead>
<tbody>
<tr>
<td>UCLA Pegasus</td>
<td>$&lt; 10$ pC</td>
<td>3 MeV</td>
<td>$3 \times 10^{-3}$</td>
</tr>
<tr>
<td>FELIX FEL</td>
<td>200 pC</td>
<td>45 MeV</td>
<td>$5 \times 10^{-2}$</td>
</tr>
<tr>
<td>SLAC SPPS</td>
<td>3.8 nC</td>
<td>28.5 GeV</td>
<td>$\equiv 1$</td>
</tr>
</tbody>
</table>

Table 3.1: Experimental beam parameters for successful EOS time-stamping experiments. The EOS modulation scales as $\Gamma \propto Q_{\text{beam}}$.

By comparison, for the experiment by Berden et al. at the FELIX FEL facility, the charge was 200 pC with an energy of 45 MeV [90]. The SPPS x-ray source used a bunch with a total charge of 3.8 nC and a highly relativistic energy of 28.5 GeV [92]. The peak electric field of the bunch scales proportionately with charge, and so in both experiments it is orders of magnitude above the peak field of a Pegasus bunch (Table 3.1). To compensate the relatively weak peak field, we took considerations in the design of the electro-optic diagnostics including choice of EO crystal medium, EO crystal thickness, and its positioning relative to the beam axis. These parameters were compared to the anticipated minimum detectable modulation of the probe laser.

### 3.2 Theory

#### 3.2.1 The electro-optic effect

The electro-optic (EO) effect is a change in the birefringence of a crystalline material in response to an externally applied electric field. In practice, EOS requires four processes to occur: (1) A transient external electric field induces
time-dependent birefringence. (2) Light with a coherent polarization state (e.g., a laser pulse) interacts with the modified EO medium. (3) The transiently birefringent EO medium encodes information about the external E-field as a phase modulation on the polarization state. (4) The phase modulation is decoded into a detectable signal.

Birefringence is a property of certain crystals in which light will experience a different index of refraction, \( n \), depending on the direction of propagation and the polarization with respect to the optical axis\(^1\) of the crystal, the ordinary and extraordinary axes parameterized by \( n_o \) and \( n_e \), respectively.

If we consider light propagating only along the optical axis, the ordinary and extraordinary rays co-propagate, and polarization state changes from linear to elliptical or circular. The effect can be parameterized in the addition of a phase retardation term \( \Gamma \) to one of the polarization components, such that

\[
\Gamma = \frac{2\pi L}{\lambda} \Delta n
\]  

(3.1)

with \( \Delta n = n_e - n_o \), \( \lambda \) is the vacuum wavelength, and the birefringent material has a thickness \( L \). This phenomenon is the basis for the design of phase retarding optics, such as half- and quarter-wave plates.

The richness of non-linear optics (NLO) arises because strong electromagnetic fields can distort the electronic structure of a material, and change its interaction with light. For example, an electro-optic material will gain birefringence in response to an applied electric field, gaining a transient \( \Delta n \) term that can act on the polarization state of light according to Eqn. (3.1) [95]. These effects manifest themselves as a change in the non-linear susceptibility \( \varepsilon \) within Maxwell’s equations [15]. In a homogeneous medium, one expresses the Maxwell equations in

\(^1\)we consider only uniaxially birefringent materials.
terms of the electric displacement field vector given by $D = \varepsilon_0 \varepsilon E$.

In anisotropic media, $\varepsilon$ is not a scalar quantity but rather a second rank tensor. This implies that the resultant displacement field in the crystal does not have to be along the same direction as the externally applied field. Furthermore, generally the electric permittivity depends on the electric field, that is, $\varepsilon(E)$. This dependence is typically represented as a power series expansion. This representation encompasses various NLO effects, including those we will explain below.

Again, in anisotropic media there can be mixing between different field vector components, that is a term that goes like $E_z = O(E^2)$ may be $E_z \propto E_x \cdot E_y$.

A solution of the eigenmodes to the wave equation (with the tensor susceptibility) is necessary to determine how light will propagate in the crystal. An equivalent and much simpler approach is to use the index ellipsoid to determine the index of refraction along the principal axes of the crystal [96]. In this formalism, we use $D \cdot E = \text{constant}$ to represent a uniform energy surface in the crystal. The ellipsoid equation can be written after diagonalization along the eigenvectors of $\varepsilon^{-1}$ as

$$\sum_{i,j} \varepsilon^{-1}_{ij} x_i x_j = \frac{x^2}{n_x^2} + \frac{y^2}{n_y^2} + \frac{z^2}{n_z^2} = 1 \quad (3.2)$$

for $i, j \in \{x, y, z\}$

The linear electro-optic effect (also known as the Pockels effect) introduces an electro-optic coefficient tensor $r$ satisfying

$$\sum_{i,j} \left(\varepsilon_0^{-1} \delta_{ij} + \sum_k r_{ij}^k E_k\right) x^i x^j = 1 \quad (3.3)$$

Equation (3.3) implies that $r_{ij}^k = r_{ji}^k$ for any material. The form of $r$ depends
Table 3.2: We compare the (electro-)optical properties of zinc telluride (ZnTe) and gallium phosphide (GaP). Note that the visible and terahertz refractive indices are similar, indicating approximate phase matching for terahertz and probe laser pulses in the medium. Values calculated from Ref. [98].

strongly on the crystal structure of the EO material and its associated symmetry group [95]. In these experiments, we tried gallium phosphide (GaP) and zinc telluride (ZnTe) crystals as our electro-optic medium. We made this choice for three reasons: (1) These materials are both transparent to 800 nm light, (2) they have relatively high electro-optic responses of the materials readily available, and (3) they exhibit a fairly weak optical dispersion for 800-nm light [97]. Ultimately, we decided on ZnTe because of the larger EO coefficient compared to GaP.

Due to this symmetry crystals that have the zincblende structure are nominally optically isotropic, that is \( n_x = n_y = n_z \equiv n_0 \). The electro-optic tensor greatly simplifies, and the only non-zero terms are \( r^{(2)}_{13} = r^{(3)}_{12} = r^{(1)}_{23} \equiv n_{41} \) (it is conventional to drop an index per Table I in Ref. [98]). The Pockels effect for zincblende crystals is described in terms of only a single \( r \) component \( (r_{41}) \) and Equation (3.3) becomes
\[
\frac{1}{n_0^2} \left( x^2 + y^2 + z^2 \right) + 2 r_{41} \left( E_x y z + E_y z x + E_z x y \right) = 1 \tag{3.4}
\]

The principle axes define a cubic structure, such that \( x = [1, 0, 0], y = [0, 1, 0], \) and \( z = [0, 0, 1] \). Considering the beam electric field \( \mathbf{E}_b \) propagating perpendicular to the (110) crystal plane \( (\mathbf{k}_b \parallel [1, 1, 0]) \), the electric field will lie in this (110) plane. We follow the calculation by Casalbuoni, et al. and define the principle (orthogonal) axes as \( \mathbf{X} = [-1, 1, 0] \) and \( \mathbf{Y} = [0, 0, 1] \). If the beam polarization vector makes an angle \( \varphi \) with the X-axis, then the electric field can be written in the crystal coordinated system \( x, y, z \) as \( \mathbf{E}_b = |\mathbf{E}_b| \left( -\cos \varphi / \sqrt{2}, \cos \varphi / \sqrt{2}, \sin \varphi \right) \).

The cross-terms in Eqn. 3.4 prevent us yet from finding the change to the nominal index of refraction, \( n = n_0 + \Delta n \). It is apparent that a simple rotation in the (110) plane by an angle \( \Psi \) will provide a principle axis transformation from \( \{\mathbf{X}, \mathbf{Y}\} \) to \( \{\mathbf{U}_1, \mathbf{U}_2\} \) and diagonalize the susceptibility tensor if we satisfy

\[
\cos (2\Psi) = \frac{\sin \varphi}{\sqrt{1 + 3 \cos^2 \varphi}}. \tag{3.5}
\]

In the new coordinate system, the induced birefringence (for small \( E_b \)) is [98]

\[
\Delta n = n_0^3 r_{41} E_b \sqrt{1 + 3 \sin^2 \varphi} \tag{3.6}
\]

where \( \varphi \) is the angle that the applied electric field vector \( \mathbf{E}_b \) makes with the \( X \) axis of the zincblende EO crystal. \( n_0 \) is the nominal index of refraction of the probe laser in the crystal medium. The combination of Eqn. (3.1) with Eqn. (3.6) allows us to calculate the relative phase shift of the two components of the probe laser electric field projected along the induced principle axes, \( \mathbf{U}_1 \) and \( \mathbf{U}_2 \) so that
Figure 3.1: (a) The 110-cut of a zincblende crystal in the cubic coordinate system. (b) The new crystal axes in the cubic coordinate system are $U_1 = [-1, 1, 0]$, $U_2 = z = [0, 0, 1]$, and $U_3 = [1, 1, 0]$. 
\( \Gamma (\varphi) = \frac{\pi L}{\lambda} n_0^3 r_{41} E_b \sqrt{1 + 3 \cos^2 \varphi} \). \quad (3.7)

### 3.2.2 EO signal detection

The EO-induced phase modulation on the probe laser can be decoded into an intensity modulation in either the crossed polarizer or balanced detection setup [99, 86]. In the crossed polarizer setup the electro-optic crystal is sandwiched between two linear polarizers whose fast axes are mutually orthogonal. When there is no external electric field there is zero laser intensity transmitted through the crossed polarizers. The electro-optic effect changes the laser polarization to elliptical, and some component is projected onto the second polarization axis, and leaks through the second polarizer to illuminate the detector.

The probe laser pulse propagates normal to the crystal face. The laser E-field can be broken down into its orthogonal components,

\[
E_l = \begin{pmatrix}
E_x \\
E_y
\end{pmatrix} = \begin{pmatrix}
E_{0x} e^{i\phi_x} \\
E_{0y} e^{i\phi_y}
\end{pmatrix}.
\]

The difference \( \Delta \varphi = \phi_x - \phi_y \), as well as the relative amplitudes \( E_{0x} \) and \( E_{0y} \) determine the polarization state of the laser. For example, if \( \Delta \varphi = i = \exp(i\pi/4) \), corresponding to a quarter wave retardation, and \( E_{0x} = E_{0y} \), the polarization vector will appear to rotate in the \( x - y \) plane as the laser propagates, forming a circularly polarized state. If \( \Delta \varphi = 0 \) but the ratio of \( E_{0x} \) to \( E_{0y} \) changes, then the laser will be linearly polarized, rotated from the \( x \)-axis by an angle \( \arctan(E_{0y}/E_{0x}) \). To simplify the following calculations, we use the Jones calculus [100] to propagate the polarization state through the electro-optic system. In this formalism, the electric field vector is normalized, and an overall phase term (which
does not affect the norm) is added so that the x component is always real.

Starting with a linearly polarized probe laser, we want to find a set of Jones matrices to model the crossed-polarizer EOS set up. If the polarization vector is rotated azimuthally so that it makes an angle $\theta$ with the X-axis, then the initial laser E-field is $E_l = E_l (\cos \theta, \sin \theta)$. In practice, this can be done with the combination of a half wave plate and linear polarizer before the EO crystal. We define various Jones matrices in the $\{X, Y\}$ coordinate system:
\[ R(\Psi) = \begin{pmatrix} \cos \Psi & -\sin \Psi \\ -\sin \Psi & \cos \Psi \end{pmatrix} \]  

(3.8)

\[ \text{EO} (\varphi) = \begin{pmatrix} \exp(-i\Gamma(\varphi)/2) & 0 \\ 0 & \exp(+i\Gamma(\varphi)/2) \end{pmatrix}. \]  

(3.9)

\( R(\Psi) \) is a simple 2-d rotation by angle \( \Psi \), and \( \text{EO} (\varphi) \) is the phase modulation from the EO effect. The entire crossed-polarizer EO interaction gives an output electric field

\[ E_{\text{det}} = \begin{pmatrix} -\sin \theta & \cos \theta \end{pmatrix} R(-\Psi) \text{EO} (\varphi) R(\Psi) \begin{pmatrix} \cos \theta \\ \sin \theta \end{pmatrix} E_I \]  

(3.10)

Note that the analyzer term has also been rotated by an angle \( \theta \), and is still crossed (at 90 degrees) with respect to the incoming polarization. The detected intensity is

\[ I_{\text{det}} = \frac{1}{2} \varepsilon_0 \left| E_{\text{det}} \right|^2. \]

Performing the matrix multiplication, we find the intensity detected on the CCD is

\[ I_{\text{det}} = I_0 \sin^2 \left( \frac{\Gamma(\varphi)}{2} \right) \sin^2 (2\theta + 2\Psi(\varphi)). \]  

(3.11)

This equation, combined with the expression for \( \Psi \) in terms of \( \varphi \) from Eqn. 3.5, makes it obvious that for a given orientation of the external [beam] electric field (\( \varphi \)), that there is an azimuthal angle setting of the polarizer and analyzer (\( \varphi \)) that maximizes the transmitted intensity, \( I_{\text{det}}(\theta, \varphi) \). A contour plot mapping this effect is shown in Fig. 3.3

Eqn. 3.11 implies that (a) the transmitted signal is strongest for the external [beam] field applied parallel to the X axis and (b) The transmitted intensity can
Figure 3.3: Rotating the polarization of the probe laser pulse allows maximization of the transmitted intensity. However, the direction of the externally applied electric field will diminish as it rotates from the X axis.

always be maximized with the proper choice of rotation for the polarizer/ana
er system. For a given angle $\varphi$ for the external field, the angle $\theta_{\text{max}}$ that maximizes the EO intensity is found as

$$\theta_{\text{max}} = \frac{\pi}{4} - \Psi (\varphi) \quad (3.12)$$

and, assuming that we rotate the polarization of the probe beam to maximize the signal, we simplify the detected signal (for small $\Gamma$) as
\[ I_{\text{det, max}} = I_0 \sin^2 \left( \frac{\Gamma}{2} \right) \approx \frac{I_0}{4} \Gamma^2 \equiv AE_b^2. \]  

(3.13)

### 3.2.3 Estimate of the EO modulation

Equation 3.13 provides a relation between the detected intensity and the bunch electric field. From an experimental standpoint, we are interested in calculating the relationship between the total charge in the electron bunch and the detected EO signal and to do so we require a model of the bunch electric field.

The electron beam is approximately a long uniformly dense cylinder in the beam rest frame. In the lab frame, the spatial beam length (longitudinal) is \( \sigma_z \), the beam radius (transverse) is \( \sigma_r \), and the beam carries a total charge \( Q \). We analytically calculate the field for such a cylindrically symmetric distribution, and find that far from the ends of the beam, the electric field is radial with

\[ E'_\perp = E'_r = \frac{1}{2\pi \varepsilon_0 r \gamma \sigma_z} \frac{Q}{r} , \quad r > \sigma_r \]  

(3.14)

Because the beam is relativistic, we have adjusted the charge density by a factor of \( 1/\gamma \) due to lengthening of the bunch when we boost into the beam rest frame. Boosting back to the lab frame the transverse field components gain an additional scaling by the Lorentz factor while the longitudinal fields are unchanged, that is \( E_r = \gamma E'_r \) and \( E_z = E'_z \) and the factors of \( \gamma \) cancel. Substituting this field into Equation 3.7 we find that

\[ \Gamma (\varphi) = \frac{\pi L}{\lambda} n_0^3 r_{41} \left( \frac{1}{2\pi \varepsilon_0 r \sigma_z} \frac{Q}{r} \right) \sqrt{1 + 3 \cos^2 \varphi} . \]  

(3.15)
Assuming, \( Q \sim 1 \text{ pC} \) in a 2.5 MeV 1 ps bunch at a distance of 500 \( \mu \text{m} \), the peak electric field will be on the order of 2 kV/m in the lab frame. According to Eqn. (3.15), the peak EO retardation will be 1.4 mrad.

3.3 Experimental design and setup

The experimental EOS timestamping design at Pegasus lab required careful consideration of our low bunch charge (1 - 5 pC) and modest beam energy (< 3 MeV). According to Equations 3.13 and 3.15 the peak electro-optic modulation is \( I_{\text{det}} \propto Q_{\text{tot}}^2 \). This results in a \( \sim 10^3 \) reduction versus the previous highest sensitivity EOS result at Felix FEL (see Table 3.1).

In designing the EO detector it was necessary to get the EO crystal as close as possible to the traveling electron bunch (practically, one can only go as close as one bunch radius to the edge), choose a sensitive enough EO medium, and to select a sufficiently thick crystal so that the effective interaction region of electron beam and probe laser is long enough.

The crystal chosen was \( 0.5 \times 10 \times 10 \text{ mm} \) ZnTe, sold by Del Mar Photonics. It was cut so that the (110) plane (i.e., the plane normal to the [1, 1, 0] crystal axis) was coincident with the large face if the crystal. In this cut, the \([-1, 1, 0]\) and \([0, 0, 1]\) crystal axes lie in this plane. In this configuration, the ZnTe is optically isotropic and thus the extraordinary and ordinary polarization modes have an equal electro-optic coefficient.

ZnTe has the major disadvantage in that it strongly absorbs EM radiation at 1 and 5 THz [101]. This leads to difficulty in measuring terahertz pulses induced by bunches with length less than 100 fs, as clipping the pulse spectrum tends to broaden the signal in the time domain. The degradation of pulse temporal profile
by terahertz resonances motivated us to try EOS with a GaP crystal instead of ZnTe. For few-pC beams, the nearly eight times smaller EO coefficient made GaP very difficult to use in our accelerator. Furthermore, we determined that despite the smearing of the EO signal, the centroid is unchanged permitting the ZnTe material’s use in time-stamping even for sub-100 fs bunches.

The EO crystal mount is designed to interact minimally with the electron beam. It is a stainless steel (non-magnetic) block with a slit cut in it. In an early design, a metal shield was installed upstream of the crystal to avoid directly exposing the crystal to beam electrons. The shield acted as a pickup antenna/resonator and the EO crystal became swamped with beam-induced wakefields, akin to those measured in Ref. [89]. This signal had very large amplitude, but it was impossible to extract timing information from the wake signal. After removing the metal shield, the wakefield signature vanished, and we easily resolved the bunch direct near field. The EO crystal was not blinded by the beam halo when the shield was removed.

The crystal is inserted into the slit and secured with two nylon-tipped set screws. The block is bolted to a rod that mates with a Huntington vacuum translation stage. After suffering several broken crystals, it is advisable to use a vacuum-tolerant epoxy to secure the crystal in lieu of the set screw. We include a drawing of the entire mount system.

To minimize the setup footprint area and the number of optics required, we opted for a spatial encoding scheme. In its most simple incarnation this scheme requires only two polarizers, a half wave plate, an electro-optic medium, and a camera with imaging system (Figure 3.5).

To make the temporal (en-/de-)coding of the EO signal more intuitive, we use an orthogonal crossing geometry between the electron beam and the probe laser.
Figure 3.4: (a) $0.5 \times 10 \times 10$ mm ZnTe crystal. (b) Broken crystal after over-tightening of the set screw. (c) The original design for the ZnTe holder, showing the crystal shield block that was later removed.
The [1, 1, 0] axis (optical axis) is oriented parallel to the propagation of the probe laser. In this geometry, there is an immediate 1-to-1 correspondence between the probe laser transverse coordinate and the centroid timing. The orthogonal geometry achieves the maximum temporal resolution in mapping temporal information onto the space coordinate.

The EO device is lowered along the vertical axis toward the beam (longitudinal) axis with a Huntington linear motion stage. To attain the highest possible EO modulation, we position the edge of the crystal within one to two beam radii (400 − 800 µm) of the beam axis. The smaller the beam size, the closer the beam can be to the crystal, hence increasing the modulation. The solenoid is typically set to minimize the beam size at the EO sensor’s longitudinal position. Collimating the electron beam with the solenoid allows us to see the shadow of the EO crystal to check horizontal alignment.

Using EOS as a time-stamp requires exactly synchronizing the EOS probe laser pulse with the target pump laser pulse. This is simply done by dividing the EOS probe pulse from the pump pulse with an ultrafast laser beam splitter (see Figure 3.6).

The pulse exits the regenerative amplifier with 2 mJ of energy. 1 mJ is sent to generate UV on the cathod, with the rest going to the pump line. 10 µJ of energy is diverted into the EOS probe pulse, or around 0.5% of the total available laser pulse energy. This yields a peak intensity for the EO probe pulse of \( I_0 = 0.1 \text{ GW/cm}^2 \), which is much larger than the minimum detectable illuminance of the CCD detector.

In fact, the intensity is so large, we require neutral density filters to sufficiently attenuate the probe laser and avoid overwhelming the CCD. The total effective attenuation is optical density (OD) 3.
Figure 3.5: The near zero crossing electro optic sampling setup. (a) The EO probe is a linearly polarized femtosecond laser pulse. (b) A passing electron bunch induces birefringence in the EO crystal, here zinc telluride. (c) The EO profile is encoded as a phase modulation on the transverse profile of the probe laser pulse. (d) A polarization analyzer with its axis perpendicular to the initial laser polarization extinguishes all except the modulated part of the laser. The EO signal is detected directly on a CCD.

In the spatial encoding scheme with crossed polarizers, the sensitivity depends on the purity of the polarization state of the probe laser, and hence on the quality of the polarizers. The polarizers are CVI Melles-Griot Glan Laser Polarizers. They have an extinction ratio of $10^5$. Ideally, there would be zero laser background with no electron beam (no electric field). However, even with these polarizers there is significant laser leak through. This forms the background signal which must be subtracted.

The detector is a lens coupled CCD camera. A TV macro lens (focal length 50-mm) and spaces to provide appropriate magnification images the crystal plane.
Figure 3.6: The EOS setup as viewed from above. The key concept is the splitting of the EOS probe pulse from the pump, ensuring no extra timing jitter is added between the two pulses.

The initial setup of this experiment did not include the camera lens, and the intensity modulated EO probe pulse illuminated the detector directly. For the lens-less setup, the background and signal both suffered from structure caused by diffraction from the crystal edge and defects. Scattering caused the signal to diffuse, lowering the peak modulated intensity. Adding the imaging system solved both of these problems and increased the overall sensitivity of the EO sensor. The magnification of the lens system is M.

The CCD has 1360 × 1024 active pixels and has a diagonal length of 8 mm.²

²Although this is a “1/2-inch format” CCD, digital sensor formats use a non-standard “inch” for historical reasons. The 8 mm actual diagonal length is listed in the manual for the sensor.
Each pixel is a square with side 4.7 µm. Each pixel in the 90-degrees interaction has a calculated temporal calibration of 4.7 µm/c × M. M here is the magnification factor from the lens system.

The pump beam path length is carefully measured and adjusted to match the total cathode drive laser path (including the path length taken by the electron beam to the target). In the accelerator bunker, a fast photodiode verifies coarse synchronization between pump and probe to 500 ps.

### 3.4 Results and Discussion

In this section we share the main results of the EOS timestamping experiments performed at Pegasus lab. To begin, we interpret the basic geometry of the spatially encoded signal, comparing the experimentally observed fields to those calculated in particle-in-cell (PIC) simulations. We include discussion of the retarded electric potential and draw an analogy to the Cherenkov effect for radiation from charges moving in a dielectric material [102]. Considering the Cherenkov-like effects, we optimize the EO probe laser polarization state to be maximally sensitive to the actual direction of electric field in the EO crystal. In this optimized condition we find that the minimum detectable charge is < 10 pC. We discuss further optimizations, including crystal imaging, to extend the detection efficacy to sub-pC bunches.

After calculating the basic timing parameters of the EOS signal, we obtain time-of-arrival information. We monitor the time-of-arrival for a large number of shots (with drift correction on the rf parameters), and find experimentally a normal distribution with δτ = 700 fs rms, consistent with the rf-induced photoinjector time-of-flight error calculation from Section 3.4.4.
A first demonstration of timestamping is achieved to show correlation between rf phase and time-of-flight in the photoinjector. These timings are confirmed with particle tracking solutions of single-particle equations of motion in the photoinjector.

Finally, electro-optic sampling is used to timestamp data taken from an actual ultrafast plasma shadowgraphy pump-probe experiment, providing pump-probe relative timing information to better than 200 fs rms, close to the bunch length timing limit.

3.4.1 Basic spatially-encoded 2-D EOS signal interpretation

We have established that EOS is sensitive to the electric field magnitude and polarization inside the EO crystal. We now describe the fields due to the ultrashort MeV electron beam. The true EM field must be derived from a full consideration of the fields generated by a collection of charges that comprise the beam, Lorentz boosted along the beam axis. In the simplest model, we can treat the field in the lab frame as quasi-static, and model the electric field of the relativistic bunch in free space as a planar terahertz half-wave pulse, with \( \omega_b = \frac{2\pi c}{\sigma_z} \). However, this planar half-wave approximation does not hold when we attempt to calculate the EM fields inside the EO crystal.

As the beam travels underneath the EO crystal, the front edge of the EO crystal becomes a boundary for coherent transition radiation (CTR) [103]. From such a boundary, one expects a train of vertical wavefronts to form (parallel to the front edge). This CTR field is the result of the planar half wave colliding with the vertical vacuum-dielectric boundary and “breaking off.” Once this transition happens, the dielectric’s front edge is the source of this wave, and the wave propagation optics are determined by the dielectric properties of the medium.
Figure 3.7: (a) EOS experimental image and (b) 2-D slab PIC simulation showing the electric field induced by the electron beam. The crystal used here is ZnTe. The signal in the experimental image displays a smaller $\theta$, due to leakage of the field around the sides of the crystal; this effect is not present in the 2-D simulation.
and the emission is not guaranteed to be coherent (denying the recovery of bunch timing information). In addition to the CTR field, there is a Cherenkov-like field is “attached” to the position of the beam at the lower edge of the crystal, and therefore the horizontal position of the Cherenkov wake is determined causally by the longitudinal position of the beam. Ideally, it is the Cherenkov signal alone we wish to measure since it is guaranteed to have a temporal structure analogous to the relative beam timing. In reality, the total field induced in the crystal is the superposition of the CTR zone and Cherenkov zone radiation [104].

3.4.1.1 Advanced modelling - PIC and TurboWAVE

We used the turboWAVE particle-in-cell code [105] to model the transient electric field pulse in the EO crystal (including dispersive effects) with the correct 3-d geometry. Both the CTR- and Cherenkov like zone fronts are clearly visible in the simulation. As the probe delay is increased and the beam moves further from left to right, the Cherenkov field outdistances the CTR signal and can be easily (and separately) measured from the CTR. Furthermore, the wavefront of the Cherenkov has a characteristic tilt with respect to the horizontal plane. The E field is polarized parallel to this front, and so in practice it can be separated from the CTR by rotating the probe laser polarization.

Once we have established the spatio-temporal connection between the Cherenkov wake, extracting the beam-laser delay becomes nearly trivial. The horizontal position of the signal is related to the TOA of the electron bunch with respect to the laser pulse. Using Fig. 3.7 as a reference, later (earlier) arrival of the laser shifts the signal horizontally to the left (right). A CCD camera is used to read

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3Special thanks to Dr. Dan Gordon of the U.S. Naval Research Lab for setting up the turboWAVE simulation and working with us to interpret the results following fruitful discussions at the 2010 Advanced Accelerator Concepts Workshop held in Annapolis, Maryland.
The coherent transition radiation (CTR), Cherenkov, and bunch direct field zones are readily identifiable.

out the time information of the bunch electric field, which is mapped onto the horizontal profile of the transmitted laser pulse [102].

Despite using high extinction ratio polarizers, in practice a small fraction of the laser pulse leaks onto the CCD even without the beam field. Contributions to the total background are due to the finite extinction ratio of the polarizers and to the scattering and residual birefringence in the electro-optic crystal itself. Background images are acquired without the beam and subtracted from the images taken with the beam on. While a small optical bias has been found to maximize the EOS signal-to-noise ratio [106, 107], in these first measurements
we have chosen to work as closely as possible to the zero-optical bias to minimize the effect of amplitude and shape fluctuations of the laser.

A typical image of the 2D field profile is shown in Fig. 3.7. Here the laser timing is such that the crystal is illuminated when the electron beam has traveled nearly to the end of it. By looking at the figure, it is immediately apparent that the sampled electric field shows a clear angle with respect to the beam direction. In vacuum, the field of a nearly relativistic electron beam points perpendicularly to the beam direction.

In our case, since the electron bunch length is sub-ps, the frequencies associated with the beam field will be in the THz range. Because of the relatively large index of refraction of ZnTe \((n \approx 3)\) at these frequencies, the electromagnetic field travels more slowly in the crystal than the beam does in vacuum. In an infinitely wide slab geometry, the angle of the field lines with respect to the beam direction would be exactly the one given by the Cherenkov condition \(\varphi_c = \arcsin(1/\beta n_{THz} \approx 19^\circ)\).

In absence of the dielectric, the electric field of the quasirelativistic beam would have a direction mostly perpendicular to the beam path. In our three-dimensional geometry, the field can be seen as a superposition of the two cases, with matching boundary conditions at the crystal surfaces. In other words, the field leaks around the thin crystal, and appears to move slightly faster. For this reason, the angle is larger than the Cherenkov prediction \([108]\). It should also be noted that the laser useful region, taking into account the CCD and polarizer apertures, was 2 to 3 times smaller than the 10 mm crystal width. In order to avoid transient effects from the crystal boundary, where the field angle could be not well defined, the laser was aligned to illuminate the downstream side of the crystal (away from the cathode). Because of the large frequency content
Figure 3.9: Determination of the beam field polarization by scanning input probe laser polarization.

In the beam spectrum, both spatial and temporal dispersion contribute to the broadening of the horizontal profile and to the angular spread of the field lines as the electromagnetic pulse travels into the crystal. Experimentally, we also found that the angle of the field lines strongly depends on the beam steering both horizontally and vertically.

3.4.1.2 Balancing the detection, dealing with background

By rotating the two crossed polarizers by the same angle and adjusting the input half-wave plate to maintain constant laser illumination of the crystal, we can control the input laser polarization angle $\theta$ (see Fig. 3.9) and study the amplitude...
of the electro-optic signal as a function of this quantity [109]. The result is shown in Fig. 3.9. The data shows a maximum of the signal for \( \theta = -17^\circ \). This indicates that the induced optical axes in the crystal are rotated by an angle \( \Psi = \theta - 45^\circ \) from the horizontal. Using Eq. (3) we obtain \( \varphi = 36^\circ \) for the direction of \( \vec{E}_b \). This result is in agreement with the result inferred in the analysis of the 2D image when taking into account experimental errors. Optimization of the incoming laser polarization allows us to measure the electro-optic signal with very low charge beams.

3.4.2 Charge dependence of the EO signal

We show in Fig. 3.10 the amplitude of the EOS signal as a function of charge. This confirms that the implemented setup is able to monitor beams of very low charges (i.e. \(< 10 \text{ pC}\)) as required by our ultrafast science applications. As expected, the amplitude of the signal is quadratic as a function of charge. This is consistent with Equations 3.13 and 3.15 which predict that the peak electro-optic modulation is \( I_{\text{det}} \propto Q^2 \). For zero optical biasing and a small induced birefringence, the intensity transmitted between the two crossed polarizers is \( I \propto \sin^2 \Gamma \approx \Gamma^2 \), which is verified by the data in Figure 3.10.

Using the total incoming infrared laser intensity (measured without the analyzer) as a normalization for the electro-optic signal, we calculate for the 10 pC case \( \Gamma \approx 10 \text{ mrad} \). This corresponds to an electric field \( E_b \approx 100 \text{ kV/m} \) which matches well the field expected for a 3.5 MeV pancake beam at 1 mm from the beam axis. Such small modulation is at the lower limit of what can be detected using the 8-bit CCD employed in the experiment. Using a better CCD array, and decreasing the shot-to-shot fluctuations on the laser intensity, it could be possible to push this limit further down. Moreover, by improving the beam pointing
Figure 3.10: In the crossed-polarizer encoding, the EOS signal goes as $Q$ squared.

jitter, one could use a larger part of the 2D image for TOA determination and obtain a signal even for lower charges. Assuming such improvements, since the ZnTe crystal thickness contributes more to the spreading of the peak than to its amplitude, a thinner crystal could be employed without suffering too much signal loss.

3.4.3 Extracting time-of-arrival from spatially encoded EOS

Various effects need to be taken into account when estimating the temporal resolution of our EOS technique for longitudinal bunch profile measurements. Since a horizontal lineout of the CCD image is a projection of the bunch field profile
due to the bending of the field lines inside the dielectric, this must be accounted for in order to retrieve the exact bunch length. The low energy of the beam implies a relatively large opening angle of the field, which will spatially broaden the EOS signal as read on the CCD camera [106]. For a 1 mm distance between the beam axis and the crystal, the smear of the signal can be estimated as $2 \times 1 \text{ mm}/c\gamma \approx 800 \text{ fs}$, where $\gamma \approx 8$ is the beam Lorentz relativistic factor. The thickness of the crystal, 0.5 mm in this experiment, which in simple terms has an effect equivalent to the shutter speed of the camera we are using to observe the beam, also limits the minimum bunch length that can be detected with this geometry to $0.5 \text{ mm} \cdot (n/c) \approx 3 \text{ ps}$, where $n$ is the index of refraction of the crystal at 800 nm. Finally, ZnTe has an absorption band at 3 THz. The resulting dispersion of the THz components of the beam field in the crystal (both temporally and spatially) naturally limits the resolution to a few hundred femtoseconds.

In spite of the smearing introduced by these effects which are responsible for broadening the width of the signal from the ultrashort beam to $> 2 \text{ ps}$, this technique works very well as a TOA diagnostic because the position of the bunch centroid is unaffected. For given settings of the rf photoinjector, the spreading in the EOS signal is fixed so that the position of the peak is a faithful indicator of the relative TOA of the laser with respect to the electron beam. In practice the TOA information is extracted by taking a horizontal lineout of the 2D image and fitting a peak to determine the beam position.

In order to obtain a calibration from pixel number to time, we recorded the position of the peak in the EOS signal as a function of the position of the infrared laser delay line. A simple linear fit yields a calibration of 28 fs/pixel, which matches the estimate obtained using the CCD camera pixel size and the relativistic beam velocity [110]. We measure the single-shot temporal resolution
Figure 3.11: Lineout from an EOS 2D image. The horizontal centroid of the image yields the beam-laser time-of-arrival to better than 50 fs.

(limited by spatial/pointing jitter) by comparing EOS centroids in adjacent pixels in the vertical dimension. The total single-shot centroid resolution is better than 50 fs rms.

To further verify the beam centroid timing measurement, we used an $\alpha$-BBO crystal to make a pair of UV pulses separated by 0.5 ps. This yielded two identical electron bunches with 0.5 ps separation (as verified with the rf deflector). Using EOS, we were able to clearly identify both beams. The measured inter-beam separation varied by $< 50$ fs.
3.4.4 Rf-induced time of arrival jitter

The total timing stability error between the pump laser and photoelectron beam will have components due to (1) low level rf-to-laser oscillator pulse train phase locking, (2) time-of-flight differences in the rf photoinjector due to (i) injection phase jitter from (1) and (ii) klystron thermal and electrical instabilities causing rf field strength fluctuations. In general, the total timing error $\delta\tau$ can be estimated as

$$\delta\tau_{total}^2 = \delta\tau_{lock}^2 + \delta\tau_{phase}^2 + \delta\tau_{amplitude}^2$$  \hspace{1cm} (3.16)

For the Pegasus system, the total pump laser to electron bunch jitter has been measured with EOS to be $\delta\tau_{total} \approx 700$ fs rms. In the following subsections, the sources of these contributions to the total timing jitter are discussed.

The first contribution to the jitter, $\delta\tau_{lock}$ originates from the active synchronization electronics between the low level master rf oscillator and the modelocked pulse train produced by the Ti:S laser oscillator. The rf master oscillator is a piezoelectric crystal oscillator - it is no more than a quartz crystal that vibrates when a voltage is applied to it.

Recall that a modelocked laser will produce not a single pulse, but a pulse
train with repetition frequency set by the length of the cavity (the inverse of the fundamental round-trip time in the cavity), \( F = \frac{c}{L_{\text{roundtrip}}} \). The pulse train phase and frequency can be modulated by changing the cavity length in a time dependent way.

Our laser oscillator, an ultrafast Titanium:Sapphire system (Micra, by Coherent, Inc.) has an integrated system (Synchrolock AP) that measures an external reference rf signal (in this case a 78.33 MHz low level signal) [master]. A fiber-coupled fast photodiode simultaneously measures the Micra pulse train [slave]. The Synchrolock is completely integrated with the Micra oscillator, and it is an easy to use turn-key system. Its electronics detect errors between the rf and laser signals. The error signal is derived by frequency mixing the signal generated by the oscillator pulse train photodiode signal with a stable clock signal.

In response to the error signal, the Synchrolock can change the length of the cavity with three different controls: a stepper motor micrometer on the output coupler mirror (for slow, gross adjustment), a low frequency galvanometer, and a high frequency PZT “tweeter” mirror to provide the finest adjustments to the slave laser pulse train. In practice, this method can provide as good as 200-fs synchronization between low level rf and the laser pulse. With only fundamental locking, the lock error upper bound is \( \delta_{\text{lock}} = 500 \text{ fs rms} \) according to the Synchrolock specifications.

The remaining jitter contributions arise during the acceleration of the beam, and can be considered a time of flight jitter. These contributions can be divided into rf injection phase and electric field strength errors. The rf injection phase error is a consequence of the synchrolock error between rf and laser pulse. On top of the previously discussed Synchrolock errors, each rf amplifier can introduce a phase jitter. For a 2.856 GHz photoinjector drive frequency, this timing error
Figure 3.13: A schematic of the rf and laser system timing. Not shown is the feedback of the phase and gun power measurements to the amplifier.
yields an injection phase error of 0.5 degrees rms. The larger rf contribution is from klystron output fluctuations. We monitor the rf power in the rf photoinjector with a loop pickup antenna. With rf feedback on, we measure a random jitter in the loop power of $\delta P_{rf}/P_{rf} = 0.25\%$ rms. This means that the fluctuations in electric field strength are $\delta E_{rf}/E_{rf} = (1/2) \cdot \delta P_{rf}/P_{rf} \approx 0.1\%$ rms on the typical peak field of 80 MV/m.

To estimate the contribution from rf parameter errors to time of flight fluctuations, we solve the longitudinal equations of motion. Since an analytical solution does not exist for a 1.6-cell rf cavity, we solve the equations using a Monte Carlo method; The initial conditions (injection phase $\varphi_0$ and peak field $E_{rf}$) are chosen randomly from Gaussian distributions with rms values set by the estimated values above, centered at the nominal set values, $\varphi_0 = 25^\circ$ and $E_{rf} = 80$ MV/m.

To understand the source of the fluctuations in cathode to target time-of-flight, we explore the expected longitudinal dynamics particles in the rf photoinjector. We limit ourselves to a 1-d single particle solution to the longitudinal equations of motion [111].

\[ \frac{d\phi}{dz} = k \left( \frac{\gamma}{\sqrt{\gamma^2 - 1}} - 1 \right) \]  \hspace{1cm} (3.17)

\[ \frac{d\gamma}{dz} = \frac{eE(z)}{2mc^2} \sin(\phi + kz) \]  \hspace{1cm} (3.18)

To solve the equations, we use a simulated electric field map $M(z)$ which we scale so that $E(z) = E_0 M(z)$. We choose $E_0$ so that the solution for $\gamma$ agrees with the measured average beam energy.

We want to determine how the random fluctuations affect the time required for the electrons to travel from the cathode to the diffraction target. This time is

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the rf gun time-of-flight (TOF), $\tau_{rf}$. Based on the solution for $\phi$ and $\gamma$ from the initial conditions ($\phi_0, \gamma_0 = 1$), we can calculate the time it takes for a particle to traverse the gun as the first term of Eqn. 3.19. The addition of a drift term allows us to calculate the total TOF as the sum of the phase advance (the particle is assumed to stay in the same bucket, that is $(\phi - \phi_0) < 2\pi$) term and the drift after leaving the gun with velocity $\beta c$:

$$\tau_{rf} (\phi_0) = \frac{\phi - \phi_0}{\omega} + \frac{L}{c} \frac{\gamma}{\sqrt{\gamma^2 - 1}}.$$ (3.19)

We will return to this formalism in Section 3.4.6 of this chapter when we verify the longitudinal dynamics with EO timestamping data. For now, we only want to calculate $\delta \tau_{rf}$, given by the rms value of Equation (3.19) resulting from the initial phase and electric field distributions. With typical operational settings for the rf system, we measure a power in the rf gun corresponding to a peak electric field of $80 \text{ MV/m} \pm 0.1\% \text{ rms}$. This value is obtained with a PID feedback on the kW rf amplifier based on the gun power. The fluctuation of the phase (no feedback) is around 2 degrees rms, centered around 25 degrees. We solve the longitudinal equations of motion numerically using a Monte Carlo generated set of initial conditions, $(E_0, \phi_0)$, sampled from gaussian distributions with their centroid and rms width values equal to those from the measured distributions. For 1000 simulated shots of the rf photoinjector with this distribution of initial conditions, we obtain the TOA jitter, with an rms of 700 fs.

Considering all these contributions, we now reconsider Equation (3.16) for the total pump-probe timing jitter:
Figure 3.14: The randomly sampled initial distribution in $(E, \phi)$ parameter space. These parameters are used to determine the distribution of time-of-flight through the rf photoinjector.

\[ \delta \tau_{total}^2 = \delta \tau_{lock}^2 + (\delta \tau_{phase}^2 + \delta \tau_{klystron}^2) \]
\[ = (500 \text{ fs rms})^2 + (500 \text{ fs rms})^2 \]
\[ \delta \tau_{total} \approx 700 \text{ fs rms} . \]

We arrive at a predicted value consistent with the EOS temporal jitter measurement. This jitter is well within the 10 ps time window monitored by the EO sensor.
Figure 3.15: A histogram generated from a distribution of relative bunch time of arrival obtained by solving the longitudinal equations of motion of a single electron in the rf gun.

3.4.5 EOS measured time-of-flight

The accuracy of the single-shot time stamp from the EOS-based measurement is determined by how well the peak of the signal is measured. The 2D image profile is very reproducible, fluctuating mostly in the horizontal direction due to the TOA time jitter. The most important systematic error to the TOA determination comes from the beam pointing jitter which alters the spatial configuration of the field in the crystal thus appearing as a false timing error. In order to give an estimate for this effect, we have tried to quantify the shot-to-shot differences in the 2D images which contain the information of the field distribution in the crystal. By taking lineouts at different rows of the 2D image (and hence different...
Figure 3.16: Histogram of EOS-measured time of arrival for 1000 consecutive shots.

heights from the beam axis in the crystal), one finds different peak positions due to the field angle. If the 2D images were perfectly reproducible, these values should correlate perfectly. By restricting the analysis to a region of the 2D image < 0.5 mm from the crystal edge, the mean rms deviation from the perfect correlation observed over 100 consecutive images is 50 fs, which can be taken as a self-consistent value for the accuracy of the TOA measurement from a single-shot EOS image.

The relative TOA jitter was measured by acquiring the EOS signal for 1000 consecutive shots. The measurement is taken with the feedback loops on the rf phase and rf amplitude closed in order to correct slow drifts over time scales longer
than a few minutes. The TOA distribution is displayed in Fig. 7, which shows a standard deviation of 700 fs. This number is high compared to that of other state-of-the-art accelerator laboratories, and an upgrade to the shot-to-shot phase and amplitude stability of the rf system is planned at Pegasus to improve this parameter. Without the implementation of a nondestructive single-shot TOA measurement technique like the one described in this chapter, the resolution of ultrafast pump-probe studies using an rf photoinjector would be limited by such jitter to $> 700$ fs rms. In our system, thanks to the described online TOA determination, the temporal resolution of an ultrafast electron diffraction study can be calculated as the quadrature sum of the electron bunch length (which for low beam charge is $< 150$ fs rms), the laser pulse length ($< 40$ fs rms), and the accuracy of the EOS-based measurement and is better than 200 fs.

3.4.6 Gun dynamics: Time of Flight vs. Energy

As a proof of principle timestamping experiment, and to validate the longitudinal dynamics model of the Section 3.4.4, we make measurement of final beam energy using a dipole spectrometer. Simultaneously the EOS system extracts the beam’s time of flight through the gun minus an offset - we call this the relative time of arrival of the beam.

Rather than let the rf jitter map out a set of small variations in initial phase as before, we use the analog phase shifter to change the injection phase $\phi_0$ and therefore change (in a correlated way) the time-of-flight and final energy of the beam. The curves in Figure 3.17 are compared to the EOM per the 1-d rf gun model. Each point represents the mean value of a few dozen shots, with the error bars extracted from the standard deviation from the mean. As the injection phase becomes larger, the error bars grow. The rf curvature is larger at this phase, and
Figure 3.17: Photoinjector longitudinal dynamics, extended phase scan.
so the jitter in the rf parameters samples a less stable set of initial conditions.

The true single-shot timestamp of the spectrometer measurement is demonstrated in Figure 3.18. Here, each data point is a single shot. The data points are spread around the model line because of fluctuations in the rf amplitude around the average value for the acquisition period. The points that lie on the line are spread due to noise in injection phase.

3.5 Conclusions

We have implemented a novel single-shot nondestructive time-of-arrival measurement for ultrashort high brightness beams suitable for ultrafast relativistic
electron diffraction or other low-charge pump-probe measurements at Pegasus laboratory [55]. The measurement technique is based on a unique electro-optic sampling scheme using a crossing geometry where the laser and the electron beam are traveling in perpendicular directions. Because of the relatively large index of refraction in the dielectric medium, the field lines in the crystal are bent. This requires a particular angle for the incoming laser polarization to maximize the signal and sensitivity to $1-5$ pC electron bunches. Finally, we have made a proof of principle demonstration of single-shot as well as stroboscopic EO timestamping of phase and energy measurements, in good agreement with numerical solutions of the rf gun single particle equations of motion.
CHAPTER 4

Pump-probe Synchronization

In this chapter we report on a simple and robust method to measure the absolute temporal overlap of the laser and the electron beam at the sample based on the effect of a laser induced plasma on the electron beam transverse distribution, successfully extending a similar method from keV to MeV electron beams. By pumping a standard copper TEM grid to form the plasma, we gain timing information independent of the sample under study. In experiments discussed here the optical delay to achieve temporal overlap between the pump electron beam and probe laser can be determined with 150 fs precision.

4.1 Motivation and background

The laser pulse pump and electron bunch probe must be synchronized if a meaningful time-dependent signal is to be extracted from an experiment. With electron photoinjector sources, the pump laser is typically split from the main laser pulse that is used to drive the gun cathode. After this split, the path of the drive laser pulse from the splitter to the cathode plus the electron beam from the cathode to the target must be exactly equalized with the path of the pump pulse from the splitter to to the target. While this seems like a simple task, one must remember that to overlap the pump and probe within 100 fs (the typical temporal resolution) the path length difference must be less than 30 µm.
The approach to synchronization is as follows: (1) To first order, the path lengths can be measured with a tape measure. A correction must be applied if the electrons travel at speed \( v < c \), since the pump pulse travels at the speed-of-light. This step should achieve mm-level equalization. (2) If the experimental setup dictates that the laser pulse must be split a large distance upstream, one can use a fast photodiode to overlap the pulses and ensure that the pathlengths are within \( 1 \text{ ns} \sim 30 \text{ cm} \), but usually this is not necessary if step 1 was made carefully. These methods do not get very close to the required synchronization.

The final step to achieve sub-ps synchronization is to insert a target, pump it, and look for laser-induced changes whilst scanning the relative length of the pump and probe lines. There are several problems with this approach. First, the pump can destroy the sample if the laser fluence is too high, or if the studied process is irreversible. This approach wastes shots, and requires replacing the sample sooner. Second, the effect of pumping the sample may make only a small change in the probe beam. Because of the low bunch charge of some DC guns, a stroboscopic scan must be made in which thousands of shots for each delay point may be necessary, requiring a fairly long time to achieve. Third, depending on the sample and process under study, the observable dynamics may not be prompt with the pump event. An example is the femtosecond laser heating of metals. Here the conduction band electrons absorb the laser photons and heat up. Over a timescale of ps, the energy is transferred to the atomic lattice which begins to disorder. For example in our electron diffraction experiments, we measure changes in the diffraction pattern based on transient atomic lattice dynamics. That is, we do not observe a signal until a pump-probe delay has passed that depends on the specific material electron-lattice coupling. For these reasons, we seek a laser pump based synchronization method that is clear, reproducible, does not destroy a sample, and is sample independent.
Photoemission from a metal is known to be a prompt process, and as a parallel to the process occurring at the photocathode. This type of emission can yield nanocoulombs of charge for 100 µJ UV laser energies in the photogun (assisted by the Schottky effect due to the large electric field on the cathode). The IR pump laser\(^1\) used here is on the order of 1 millijoule and, unassisted by an external extraction field, the total charge yield is limited to < 100 pC. The probe electron beam will scatter from the emitted charge, and provide time-zero determination. There are several examples in the literature of this deflectometry technique being applied to synchronize keV (non-relativistic) electron bunches from DC photoguns with a pump laser pulse [8, 37, 112, 113, 114, 115, 116, 117].

When a metal surface is illuminated by an intense femtosecond laser pulse a variety of processes take place including non linear laser absorption, photoemission [118, 119], plasma formation, thermal sublimation, and ablation [120]. The time scales of these processes are all different and depend on many parameters including laser fluence, surface conditions, etc. A detailed description of the phenomena is complicated [121]. Nevertheless to the purpose of this synchronization technique what is important is that transient electric fields associated with electron cloud expansion and plasma formation develop on ultrafast (< 1 ps) time scales. Photoemission is the fastest such process that can occur.

By passing the electron bunch through the laser induced plasma, it is possible to detect the induced transient fields that act on the beam as an electron lens, transversely deflecting a portion of the bunch. If we observe the transverse beam distribution on a downstream screen, there will be a change in beam density in the area of the bunch that interacted with the plasma. Studying the evolution of this shadow as a function of the pump-probe delay can provide an accurate

\(^1\)See Appendix A for a description of multiphoton photemission.
method to set the zero crossing delay for a particular experiment, independent of the details of the setup or the samples used in that experiment.

This method of determining temporal overlap has not been reported for electron bunches with relativistic energies. In this chapter, we describe the first time zero determination for femtosecond relativistic electron diffraction using plasma shadowgraphy with plasma induced by ultrashort laser pulses on copper TEM grids.

### 4.2 Experiment

In this time-zero determination experiment, the photoinjection system was not optimized to produce a beam with minimum bunch length, but rather to improve the beam transverse emittance. The laser was focused on the cathode to a spot size $< 50 \, \mu m$ rms. The UV intensity was adjusted to produce a total bunch charge of 0.3 pC to reduce the bunch length expansion due to space charge. The rf gun peak electric field was set to 45 MV/m, giving an electron bunch energy of 2.4 MeV. In this configuration beam dynamics simulations indicate a bunch length of 700 fs rms at the sample located 80 cm from the cathode.

The target used in these experiments is a standard 3-mm diameter copper TEM square grid (uncoated PELCO and Gilder brand TEM grids from Ted Pella, Inc.) Experiments were performed with 300-, 500-, and 2000-mesh grids. Similar behavior was noted for all mesh sizes. For ease of analysis, the results presented in this paper were obtained with the finest grids. The 2000-mesh grids have a single pitch size of 12.5 $\mu m$ defined by a 7.5 $\mu m$ wide hole and a 5 $\mu m$ wide bar. The thickness of the copper is nominally $6 \pm 2 \, \mu m$, which is thick enough to cause multiple wide angle scatter events for the electrons that strike the bar.
Figure 4.1: Schematic drawing of pump-probe geometry (not to scale). The electron bunch probes from the front of the TEM grid, while the laser pump illuminates the grid from the back. The shadowgraph shown is at a pump-probe delay for which the shadow from the plasma is most well-defined. The target in this case is a 500-mesh copper TEM grid.
These scattered electrons do not propagate to the detection screen at the end of the beamline and are effectively lost. In the square grid geometry these pitch dimensions give a total electron transmission of 36%.

75% of the available laser energy is split off and used to pump the sample target under study. The pump pulse travels through an optical delay line that provides a delay between the pump and cathode drive laser pulses. With a micro-stepping motor, the delay is controllable in steps of 16 fs.

The pump pulse passes through a lens, enters the beamline, and is reflected by an in-vacuum mirror to back-illuminate the TEM grid. At the grid the focused pump laser spot size is 40 \( \mu \text{m} \) rms. The detailed layout geometry is shown in Figure 4.1. The average laser power is measured using a joulemeter to be 1 mJ just before entering the beam line. Taking into account all transmission efficiencies, we estimate the maximum fluence on target is 7 J/cm\(^2\).

It should be noted that we use the 800 nm pulse to pump the grid, eliminating non linear frequency multiplication stages. It has been demonstrated [72] that for a laser pulse with high enough intensity as used in this experiment, multi-photon photoemission is more efficient in extracting charge from a metal surface than single-photon photoemission after a higher harmonic conversion. The formation of the plasma is initiated by multi-photon photoemission in which three 800 nm photons with a combined energy of 1.55 eV \( \times 3 = 4.65 \text{ eV} \) are absorbed by the copper. A very high charge density is expected to be produced on the grid from the photoemission process alone, on the order of 2.5 pC over a 40 \( \mu \text{m} \) spot, i.e. 900 pC/cm\(^2\).

The operating mode of the Pegasus photoinjector at the time of these experiments was a “parabolic-profile cigar” aspect ratio beam, obtained by having a very small, intense laser spot on the cathode (here < 50 \( \mu \text{m} \) rms). In this regime,
Figure 4.2: Simulated and experimental shadowgraph images for two solenoid focusing conditions. (left column) Focusing before the pinhole and the TEM grid. (right column) Focusing after the TEM grid. The diameter of the beam area interacting with the plasma is 2 mm on the phosphor screen. Taking into account the magnification factor, $M \sim 1.8$, the extent of the deflecting electric field on the grid is approximately 1 mm.
the beam undergoes a self expansion driven by transverse space charge forces with the final result being a long collimated electron bunch having low emittance and high compressibility [122]. This configuration is not optimized to provide minimum bunch length, but rather to have high transverse uniformity and high beam quality (i.e., low emittance).

To improve beam quality, we steer the beam through a 1 mm diameter pinhole drilled in a non-magnetic steel plate. The pinhole also reduces the bunch charge from 0.3 pC to 0.16 pC. The pinhole is mounted 3 cm upstream of the TEM grid and serves to remove dark current, select a smaller subset of the beam, and improve the uniformity of the transverse beam profile and of the shadow image. A phosphor screen is placed 2 m downstream of the grid.

The only focusing element on the beamline is the emittance compensation solenoid located immediately after the gun exit. We have compared two focusing schemes for this experiment. The beam is focused (before/after) the TEM grid to obtain a (real/virtual) point source illumination of the grid. \( M \sim 1.8 \) is the magnification chosen for these experiments. The typical data shadowgraph images for either focusing scheme is shown in the bottom row of Figure 4.2.

In both schemes, the beam drifts after interacting with the plasma to strike a high yield phosphor screen positioned 2 meters downstream of the TEM grid. The screen produces a photon intensity pattern \( I_{x,y} \) that is proportional to the transverse charge density (integrated over the longitudinal profile). A large aperture lens is used to efficiently couple the screen to a PI-MAX2 intensified charge coupled device (ICCD) [53]. The screen-lens-ICCD system has been calibrated to measure bunch charge that would otherwise be too small to detect with a Faraday cup or integrating current transformer (ICT). The sum of the counts in the shadow images indicate a total bunch charge after the pinhole of \( \sim 160 \, \text{fC} \pm 10\% \).
if we include shot-to-shot charge fluctuations.

4.3 Results and Discussion

In the first focusing mode, the beam is focused to a minimum transverse size (a waist) at a point before the pinhole and TEM grid. When the beam passes through the the grid, the plasma electric field deflects beam electrons away from the center of the plasma distribution. Since the beam electrons are diverging from the focus point, the resultant shadowgraph depicts a transverse charge distribution with a circular depletion region (hole), as shown in the first column of Figure 4.2.

In the second focusing mode, the solenoid current is reduced so that the beam achieves a waist at a point after the pinhole and TEM grid. In this scheme, the beam electrons are convergent as they pass through the TEM grid. The plasma on the grid still deflects electrons away from the center of the beam, but since the electron trajectories go on to cross in the focus, the grid plasma now effectively causes the deflected electrons to form a region of higher charge density in the transverse profile of the beam as recorded at the detector plane. The result is a bright spot in the image rather than a depletion region, shown in the second column of Figure 4.2.

We simulate the propagation of the electron beam with the General Particle Tracer (GPT) code (by Pulsar Physics). While an analytical model of the plasma evolution in time combined with electron beam dynamics using GPT has been published [114], in order to understand the behavior of the beam in the different focusing conditions we utilize a much simpler model for the plasma and assume it as a static and uniform electron distribution.
Figure 4.3: A sequence of frames showing the temporal evolution of the shadow cast by the charge distribution on the grid. In this case, the solenoid is set to achieve a focus upstream of the TEM grid. The delay between subsequent frames is 500 fs.
The total simulated plasma charge was 2.5 pC, and the charge disk size was 30 \( \mu \text{m} \) rms. Figure 4.2 compares shadowgraphs from data (taken from the maximum signal delay) and simulation for both focusing configurations, showing good agreement with the experimental images in the two different solenoid conditions.

Moving the translation stage to vary the time delay between the pump and the probe beam at the grid we obtain the sequence of images shown in Figure 4.3. In this case the solenoid is set in the first focusing mode, i.e. with the beam focus located before the TEM grid.

To evaluate the signal from each intensity map image, \( I_{x,y} \), we define the 2d correlation coefficient \( \Gamma(t';t) \) of an image at delay \( t \) with a reference image at delay \( t' \) taken well before the zero crossing:

\[
\Gamma(t';t) = \frac{\sum_{x,y} C_{x,y}(t)C_{x,y}(t')}{{\sqrt{\sum_{x,y} C_{x,y}^2(t)}}{\sqrt{\sum_{x,y} C_{x,y}^2(t')}}}
\]

where \( C_{x,y}(t) = [I_{x,y}(t) - \bar{I}(t)]/\bar{I}(t) \) and \( \bar{I} \) is the mean of all pixels in \( I_{x,y} \). The 2-D correlation coefficient effectively compares the pattern of two images at different delays \( t \) and \( t' \) while ignoring absolute variations in intensity [123]. Since each image is normalized by its own mean pixel value, shot to shot fluctuations in charge or energy (due to rf amplitude and phase jitter) have little effect on the correlation.

Since the reference image is taken at a time \( t' \) well before the measured zero crossing, each image is compared to a near uniform background. As the hole appears as a function of delay, the correlation signal decreases until it reaches a minimum value corresponding to the delay for which the hole pattern is most well-defined.
Figure 4.4: The 2-D correlation signal as a function of pump-probe delay. The beam is focused before the hole in this analysis. In the main graph, each data point represents the correlation coefficient obtained from a single shot image. For the inset graph, the data has been collected in 700 fs bins and the scale changed to highlight the fast onset of the signal. The fit is a convolution of an exponential decay with a gaussian recovery and is used to guide the eye. A dashed line has been introduced to indicate the separate timescale for recovery.
We choose to use the 2-D correlation coefficient to quantify the effect of the plasma on the beam in lieu of the size or intensity of the depleted region in the electron transverse distribution. Here the goal is to demonstrate the determination of timing information using a simple analysis algorithm.

Figure 4.4 shows the evolution of the autocorrelation signal $\Gamma(t'; t)$ as the pump laser is made to come earlier relative to the electron bunch. The beam is focused before the hole in this analysis. $\Gamma(t'; t)$ takes a value of 1 for shots in which the pump laser comes much later than the electron bunch ($t < 0$). As the pump laser delay is reduced, the plasma is formed in time to interact with the electron bunch, part of the bunch is deflected, as illustrated by a fast reduction in $\Gamma(t'; t)$ for $t > 0$. For $t >> 0$ the autocorrelation signal should once again tend toward zero as the plasma recombines or dissipates. For the scan shown in Figure 4.4, the plasma recombination occurs over $> 100$ ps, a much longer timescale than its formation.

In addition to free electrons from photoemission (having a thermal velocity $v_{th} \sim 10^6$ m/s due to the excess kinetic energy imparted during photoemission), the plasma has components from ions and electrons ejected from the bulk material by other ultrafast laser-solid interaction mechanisms [121]. The presence of positive Cu$^+$ ions in the plasma may explain the long tail ($> 100$ ps) of the signal in Figure 4.4 due to the much higher mass (and therefore slower dynamics) of ions. The presence of positive ions also explains the different focusing behavior in the hole center (apparent in the later frames of Figure 4.3). This is one piece of evidence in the ongoing discussion of the ablation of metals by femtosecond laser pulses [124].

The time zero is easily obtained from the data in Figure 4.4. A linear fit is made to the falling edge of the data and the intercept with the $\Gamma = 0$ line
determines very precisely the zero delay between probe electron beam and pump laser pulse. The uncertainty in the slope translates to a $< 150$ fs uncertainty in determination of the zero-crossing.

4.4 **Proof of principle EOS time-stamping**

The plasma shadowgraphy experiment provides an excellent test setup to apply electro-optic sampling based time-stamping. The basic experimental setup is as described in Chapter 3. The pump-laser delay stage was set roughly in the the center of the sharp falling edge of the curve it Fig. 4.4. We made a synchronized acquisition of the EOS and shadowgraph data. In post-processing, the shadow
Figure 4.6: A successful time-stamping of the shadowgraph data. Both the fitted hole size and average intensity (indicating charge) around the hole region deviate from the fitted line with 200 fs rms, giving an upper bound of the EOS time-stamping resolution.

“hole” size and average intensity of the region as measured in the shadowgraphs were plotted against the centroid time delay extracted from the EOS data.

The jitter of the rf system sampled pump-probe delays with an rms value of 1 ps in this experiment (Fig. 4.6). We make a linear fit to this data, and find the correlation to be better than 200 fs rms. This sets an upper bound on the EO time-stamping resolution.
4.5 Conclusions

Laser-induced plasma shadowgraphy by an ultrashort MeV electron beam has been demonstrated as a reproducible method to determine the time-zero delay on the order of 150 femtoseconds for pump-probe experiments. Single-shot electro-optic sampling based time-stamping has been applied to the shadowgraph data as a proof-of-principal study. We have shown the ability to time stamp pump-probe events to better than 200 fs.
CHAPTER 5

Shadowgraphy of Ultrafast Laser Ablation of Copper

In the experiment described here we use an intense ultrashort laser pulse to create an optically-induced plasma by thermally-assisted multi-photon photoemission and laser ablation at a copper surface. A relativistic electron beam travels through the plasma and interacts with the plasma’s electromagnetic fields. Part of the beam deflects, and a shadow forms in the beam profile. In Chapter 4, we already touched on the basic phenomenology of the interaction of an ultra-short, high intensity laser pulse pumping a metal surface. We used a multi-shot approach to study this problem with an ultrashort electron beam wherein a mechanical path length change varies the temporal delay between a laser pump and short probe electron beam.

In this chapter we describe an experiment designed to explore the laser-induced plasma evolution. Utilizing a long (4 ps) electron probe beam with a short laser pump pulse, we make a measurement with the use of an rf streak camera (echoing the approach used in the streak-based ultrafast electron diffraction of gold in Chapter 2) with a temporal resolution of 35 fs rms. We are able to visualize the entire history of the initial plasma formation in a single shot. A detailed analysis shows that the initial growth of the shadow occurs because of the expansion of the plasma EM field event horizon at the speed of light. Modeling
the Coulomb scattering of the beam electrons from the retarded EM potential allows us to extract the total charge and time-scale of the plasma formation. We find that when the pump laser fluence increases, there is a transition from instantaneous photoemission to a thermally-assisted emission process consistent with the electron-phonon coupling time scale. With these two results we demonstrate the use of an electron rf photoinjector source to probe the ultrafast evolution of optically-induced plasmas.

5.1 Background and Motivation

In previous chapters of this thesis we have described experiments using electrons as direct probes of ultrafast laser-induced processes, specifically the ultrafast laser-induced melting of gold. In the investigation of solid state structural dynamics, time resolved electron diffractometry is used to obtain accurate reciprocal space observation of the motion of the atomic nuclei with sub-ps temporal resolution [125, 126]. Ultrafast electron microscopy, both in single shot and stroboscopic modes, yields atomic spatial resolution snapshots from the fs to the ns time scales depending on the electron bunch length [127]. Similar techniques can also be applied to study the ultrafast dynamics of non-neutral plasmas.

Recently, there has been a drive to develop diagnostics of ultrafast dynamics in dense plasmas. Motivations range from plasma accelerators, to inertial confinement fusion, to fundamental plasma physics. Changes in plasma density can be measured with higher time resolution from a variety of optical probe techniques, including Raman scattering [128], Thomson scattering [129, 130], interferometry [131], longitudinal frequency domain holography [132], and supercontinuum spectral interferometry [133].
Despite the success of these optical-only techniques to measure density perturbations in very dense plasmas, they become ineffective in probing lower density plasmas, such as those produced in ultrashort laser ablation and photoemission studies [113]. The direct plasma-photon coupling experiments (Thomson and Raman scattering) require a very high probe laser intensity to compensate for the very small optical scattering cross sections. Furthermore, laser probes are entirely insensitive to the electromagnetic (EM) fields produced by the plasma distributions. Indeed, the EM fields themselves may exhibit dynamic effects at a shorter timescale than the plasma can evolve. Since charged particles couple strongly to EM fields, they can be a more sensitive tool to study low density ultrafast plasma evolution.

The use of electrons as direct probes of ultrafast laser-induced processes has progressed tremendously in the last ten years. In the investigation of solid state as well as gas phase structural dynamics, time resolved electron diffractometry is used to obtain accurate reciprocal space observations of the motion of the atomic nuclei with sub-ps temporal resolution [125, 126]. Ultrafast electron microscopy, both in single shot [134] and stroboscopic [127] modes, yields atomic spatial resolution snapshots from the fs to the ns time scales, depending on the electron bunch length. For studies of electromagnetic field dynamics, such as the very complex evolution of laser driven high density plasmas, proton radiography has been the preferred technique [135]. Attempts have been made to diagnose optically-induced plasmas using < 100 keV electrons [113, 115], but the main issue in widening the range of these studies is the relatively short penetration depth of low energy electrons. Relativistic electrons are an excellent probe of electromagnetic fields; they can penetrate thicker and denser plasmas than their non-relativistic counterparts and, compared to protons, offer higher temporal resolution and increased sensitivity to small electromagnetic perturbations.
All of these electron based probing techniques share the common issue that their temporal resolution is limited by the ability to generate a short electron beam [136]. This has been one of the main motivations for the introduction of high power RF guns and bunching cavities to shorten the beam bunch length to less than 100 fs [42, 43, 57]. However, the noise in the RF-laser synchronization electronics generates jitter in the time of arrival between the laser and the electron beam at the target, thus limiting the temporal resolution to a few hundred fs.

RF-deflector based streaking offers a solution to this problem. In 1982, Mourou and Williamson first proposed the concept of time-streaking the electron beam after it has recorded information from a sample [64]. Recently, this idea has been revisited and a proof-of-principle demonstration using an RF deflecting cavity was obtained with a single shot diffraction study of the ultrafast laser induced melting of gold [56]. This approach has the main advantages that it relaxes the requirements on both the electron bunch length and the time of arrival jitter, since temporal resolution is instead set by the resolving power of the RF streaking system.

In this chapter we demonstrate the use of an RF deflecting cavity and a relativistic electron beam to capture, in a single shot and with 35 fs temporal resolution, the evolution of electromagnetic fields generated by intense laser illumination of a metal sample.

The improvement in temporal resolution in our setup allows for the first time the direct observation of the expansion and propagation of the field light cone during the first few hundred fs after the photoemission. The data analysis allows to infer the amount and the time constants of the emitted charge, showing the potential of this technique to provide information for ultrafast laser ablation studies.
5.2 Theory

5.2.1 Illumination of metal with intense femtosecond laser pulses

The physics of short pulse high intensity laser illumination of metal targets is rich and depending on the laser intensity regime, many physical processes can take place [121] such as superfast melting [137], ablation [138, 139], and electron [140] and ion [141] emission.

Because of the relative timescales of energy transfer mechanisms in metals, the initial response of the material after femtosecond laser interaction is limited to the emission of electrons into the vacuum. In the first few hundred fs, the laser energy is deposited only in the electron subsystem. The electron distribution gains a non-equilibrium excitation [74], and ballistic (viz. “hot”, with \( v_{th} \sim 10^6 \text{ m/s} \)) electrons rapidly diffuse on the order of one hundred nanometers into the bulk material [142]. The electron sub-system begins to thermalize via electron-electron interactions and at the same time the higher energy tail of the electron distribution can be emitted at the vacuum interface [143]. The dynamics of the energy relaxation from the excited electron distribution become complicated, as some of the energy of the excited electrons is given to the atomic lattice through electron-phonon coupling [71]. A further complication of the photoemission process from the classical Spicer photoemission model [144, 145] is the space-charge induced modification of the material work function [146].

A typical phenomenological approach used to describe these effects is the classical two-temperature model (TTM), which first arose in the early 1970s [70] to describe material dynamics following the absorption of ps laser pulses. With shorter (fs) pulses, and higher laser intensity, the basic TTM has been modified to include fast diffusion (through the bulk material by ballistic electron trans-
port) [147] as well as electron emission and space charge (Schottky) saturation mechanisms. While the validity of this model in the initial times after excitation is debatable, it has been shown to provide good description, even for fluences near the ablation limit [148].

The generalized Fowler-Dubridge theory states that the $n$–photon photomission current is proportional to $I^n$, where $n$ is an integer (See Appendix A). For very high laser intensity $I$, there is evidence that there is a non-linear enhancement to the photocurrent, and that $n$ should be replaced by an intensity dependent non-integer [149]. A recent paper [150] determines the thermal and field effects for extreme fluence photoemission, and gives a recipe for the calculation of the total emitted charge as a function of laser fluence for pump pulses less than 100 fs.

5.2.2 Femtosecond probing of transient electromagnetic fields with MeV electrons

Particle deflection by transient laser-induced electromagnetic fields is a well known effect which has been used in the past to determine time-zero in pump-probe experiments and to study the effect of plasma evolution and charge cloud expansion [8, 37, 112, 115, 151, 152].

In the first few hundred fs, the laser energy is deposited only in the electron subsystem, and the dominant process is the emission of the excited tail of the electron energy distribution. Free charge photoemission results in an electromagnetic wave being launched in free space. Assuming that the charge is emitted at rest, the main effect will be a Coulomb-like electric field propagating at the speed of light from the point of emission. A probing particle’s response will be delayed by a time interval related to the in-vacuum propagation of this electromagnetic wave.
field from the point of emission.

In our investigation of the femtosecond laser induced photoemission, we particularly want to extract information about the total charge emitted, as well as the time scale of the emission, with the end goal being an estimate of $Q(t)$. For that reason, it is necessary to have a very high temporal resolution to study the scattering of the beam electrons.

Our setup’s improved temporal resolution allows, for the first time, the direct observation of the expansion and propagation of the electromagnetic field light cone during the first few hundred fs after the photoemission event. By modeling the electromagnetic scattering of the probe electrons, including the speed-of-light propagation of the fields’ light cone, we infer the amount of charge and the timescale of the emission process, thus showing the potential of this setup to study ultrafast high intensity laser-matter interactions.

5.3 Experiment

The basic plasma shadowgraph experiment is very similar to the other pump-probe setups already described in this thesis. Typically with ultrafast studies in a photoinjector we have made the electron beam as short as possible to maximize the temporal resolution. However, in this experiment we approach the opposite extreme and make a very long electron bunch (many picoseconds long) and rely on the deflector to give good time resolution. The advantage of a long beam in this case is that an extended history of the process under study can be obtained in a single shot. This technique has been demonstrated previously in the diffraction study of the ultrafast laser induced melting of gold [56].

To achieve a long electron bunch, we illuminate the cathode in the RF gun
with a quasi-flattop 4 ps long UV laser pulse obtained using the stacked $\alpha$–BBO crystals technique [78]. A single 266 nm laser pulse (split from the main infrared laser pulse which is also used to pump the sample) passes through a series of $\alpha$–BBO crystals. If a short linearly polarized laser pulse is sent into the crystal with the crystal’s principal optical axis rotated by some angle with respect to the laser’s polarization vector, then the two components of the polarization will experience different group velocity delay and will split into two laser pulses with a separation proportional to the thickness of the crystal. The angle of rotation determines the relative intensity of the two pulses. For these experiments, the crystals are set so that all the pulses share equal intensity.

The long laser profile has some gaps between the beamlets. However, the electron bunch longitudinal distribution is not as simply proportional to the instantaneous intensity profile of the laser pulse train. Though, at first approximation, the cathode photoemission process can be assumed to be nearly instantaneous at the timescale of the beam pulse train, the dynamics of the rf photoinjector significantly influence the final macrobunch longitudinal profile [79].

Furthermore, in this experiment we injected less than 1 pC of charge to avoid significant beam evolution and obtain a uniform 4 ps long bunch longitudinal profile. The spot size at the cathode is <30 $\mu$m rms to improve the beam emittance [153]. The beam energy at the gun exit is 3 MeV. With these careful considerations of the RF phase and the longitudinal space charge induced dynamics [154, 155] we limit the temporal evolution of the beam distribution and maintain a near-uniform longitudinal and transverse beam profiles.

Inserting a metallic mesh into the beam path scatters the electrons that hit the bars of the mesh into a wide angular distribution, effectively blocking them. The result is that the mesh casts a shadow as shown in Figure 5.1(b). Two im-
Figure 5.1: Transverse electron beam profile. (a) No grid. (b) 300 mesh copper grid inserted.

Important parameters can be inferred from this shadowgraph. Firstly, the angular magnification due to the divergence of the electron beam from a focus upstream of the target can be calculated since the spatial dimensions of the grid are known, as well as the size of a pixel as imaged onto the detector. Secondly, we can glean information about the transverse emittance from the grid image. Just as a collimated light source produces a shadow with high contrast edges while an angularly diffuse light source blurs the edges, a beam with lower emittance will produce a shadowgraph with higher sharpness. This fact is the basis for high brightness photoinjector nanoemittance measurements that utilize TEM grids to measure transverse emittance too low to measure by conventional techniques [153]. The requirement of maintaining low emittance limits the phase-induced stretching that we can impose by increasing the injection phase, as the emittance degrades as the phase increases due to a combined effect of compression (increased energy spread) and a higher Schottky effect (increased thermal emittance component) as the injection phase approaches peak field.
Figure 5.2: Cartoon scheme of the undeflected shadowgraphy experiment.

5.3.1 Forming the shadowgraph

The metal targets under study in this experiment are copper and gold metal transmission electron microscopy (TEM) grids. The grids are available in various rulings, or mesh sizes, given in the number of openings per inch. Two mesh sizes were used, 300- and 2000-mesh. The coarser mesh was used to determine the overall magnification of the electron beam optics and spatial calibration of the shadow images since it was possible to resolve individual bars in the shadowgraph. The finer 2000-mesh grids were used for pumping since they better approximate a uniform metal surface with effective transmission of 40%. In this case, it is not possible to resolve the individual bars.

The total laser pulse output from the regenerative amplifier is split 75 − 25%. The pump is the larger portion of the main 800 nm laser pulse. Prior to sending the pump pulse into the accelerator bunker, the pulse energy is 1.2 mJ. A half
wave plate and cube polarizer combination are used as a broadband variable attenuator and then the beam is transported to the accelerator bunker (with a 60% efficiency) where it passes through a 30 cm lens and is focused to a minimum spot (25 \( \mu \text{m} \) rms) on the target sample in the vacuum chamber located 90 cm from the cathode. A trigger enabled fast mechanical shutter is installed before the target to ensure that only one pulse can reach the sample and that it is synchronized with the ICCD image acquisition. This is an essential detail if the pump fluence is high enough to be destructive to the sample.

We split off 1% of the pump pulse energy after the lens, and focus it onto a Yttrium-Aluminum-Garnet (YAG) screen. Placed at the same distance from the lens as the cathode, this serves as a virtual pump screen where we can continuously monitor pump intensity, alignment, and spot size. From the intensity profile, we measure a minimum size at the focus of \( \sigma = 20 \mu \text{m} \) rms.

Spatial alignment is fairly straightforward. The sample holder has a piece of DRZ phosphor mounted on the edge. This acts as a screen for both the electron and pump laser beams. After all steering and focusing parameters for the electron bunch have been set, the screen is inserted and an image of the transverse beam profile is saved. The focused and highly attenuated pump laser is then steered to achieve spatial overlap with the electron bunch. The spatial alignment can be verified (independently of temporal alignment) by removing all attenuation from the pump laser pulse and opening the shutter. If the spatial alignment is good and a metal mesh target is inserted, a bright spot will appear on the beam profile where the mesh has been completely ablated.

As previously mentioned, the pump laser pulse is split from the same beam that drives the photoinjector cathode. As a result the pump pulse and photoelectron bunch are synchronized to within the inherent laser-rf jitter of \( \sim 1 \text{ ps} \). The
Figure 5.3: Cartoon scheme of the deflected shadowgraphy experiment.

path lengths from the beamsplitter to the sample target is on the order of several meters. To ensure the pump and probe overlap, the two path lengths must be equidistant to about 30 microns. Accordingly there is a mechanical delay stage installed on the pump line with a retro-reflecting mirror pair on top. This serves as the fine adjustment to achieve temporal overlap between the electron bunch and the pump laser pulse.

It can be tedious to find the correct temporal alignment. It is necessary to first have good spatial alignment between pump and probe beams as described in the previous section. Coarse temporal alignment is achieved through careful measurement (with a tape measure!) of both path lengths. In the end, however, one must pump the grid and look for the shadow signal while patiently scanning the pump delay stage. Attenuating the pump laser to just below the single-shot damage threshold will produce a detectable signal while preserving the grid for more shots. There is the saving grace that the grid plasma, once formed, is very long lived. Some sort of signal is apparent even several hundred picoseconds after the pump laser pulse hits the sample. It is therefore more productive to err on the side of pumping too early than too late.

We can treat the solenoid focusing with a geometrical ray optics formalism.
Figure 5.4: The steps for making a deflected shadowgraph. (a) Full transverse beam profile. (b) Quadrupole focused beam. (c) Quadrupole focus with the slit inserted. A 2000 mesh copper grid is inserted, and the dark spots are damaged areas of the grid that transmit 100% of the beam. (d) The 300 mesh grid is inserted to enable calculation of the angular magnification from the solenoid and quadrupole magnets. (e) The 2000 mesh grid is swapped back in and the deflector is turned on. The phase is set to provide a uniform longitudinal distribution, but in this shot the rf phase and amplitude jittered such that the bunching factor changed and the beamlets overlapped, forming the dark horizontal bands.

The gun solenoid is used to focus the electron beam to a small spot 20 cm before the target position. The spot size at the target is 400 $\mu$m rms and a magnification ratio of 10 is obtained at the detection screen, located 2 m downstream. After careful spatial alignment and rough synchronization of the pump arm, we fine tune the pump-probe delay while monitoring the beam transverse profile for the scattering of the central area of the relativistic probe beam by the Coulomb electric field of the photoemitted charge. This forms the pre-streaked signal, as demonstrated in Figure 5.2.
Once a clear signal is established, we insert a 100 µm wide slit in the beam axis and select a horizontal slice of the beam aligned with the pump laser. A 9.6 GHz 500 kV 9 cell RF deflector is then used to temporally streak the beam in the vertical direction and obtain a time-dependent shadowgraph image. Figure 5.3 shows the setup, modified from the previous undeflected scheme, to include the slit and streak camera. Quadrupole lenses (not shown in Fig. 5.3) are used to maximize the streaking resolution of the system which is set by the beam size on the screen when no deflection applied.

From an analysis of the beam image through the slit with the pump laser and the RF deflector off (inset of Fig. 5.5) we infer a 35 fs temporal resolution for this system.

5.4 Results and Discussion

When the pump laser is turned on, we obtain streaks such as those shown in Fig. 5.5. An immediate observation is the very fast transverse expansion of the scattered region (shadow); in the first few hundred fs it grows from a small region, comparable in size to the laser-illuminated area, to a large hole of mm-size. By comparing two subsequent shots, it is also apparent that the technique gives a result that is insensitive to time-of-arrival (TOA) jitter. In fact, in these images the pump-probe TOA jitter causes the onset of the scattering (the so-called time-zero of the laser-beam interaction) to jump vertically as the laser strikes the sample at different times relative to the electron beam as explained in Chapter 3. However, as long as the laser pulse falls within the relatively long (4 ps) time window covered by the electrons, the detector still collects the entire time-history of the ultrafast process with unmodified temporal resolution.
Figure 5.5: Rf induced jitter can be measured from these shadowgraphs from consecutive shots, but the jitter does not affect the resolution or interpretation of the shadowgraph. (inset) The unstreaked transverse profile after the beam passes through a collimating slit demonstrates the limit of the deflector temporal resolution.

The interpretation of the time-dependent shadowgraphs requires a model of the electromagnetic fields of the photoemitted charge distribution and their evolution, along with a solution of the MeV electrons’ equations of motion in these fields [37]. Initially to simplify the model, we start by assuming that the electrons are emitted instantaneously when the laser strikes the sample, and then remain in proximity to the small area illuminated by the pump. In reality, the photoelectrons are emitted with a small initial velocity and then experience laser ponderomotive and self space charge forces which cause them to expand. At the moderate laser intensities employed in this experiment, the maximum kinetic energy of the electrons is much less than 10 keV, corresponding to a speed $< 0.1c$. At this speed the emitted electrons can only move 10 $\mu$m in the first 300 fs. Hence, it is a good approximation (at least initially) to neglect the spatial
Figure 5.6: Ray tracing for shadowgraphy with a solenoid-induced beam waist upstream of the target grid.

evolution of the charge distribution. With these assumptions, the photoemitted charge can be modeled as a point charge $Q$ at the origin; the associated field is a simple Coulomb field. A more realistic model would also include the presence of the grid, which locally modifies the Coulomb electric field.

We calculate the motion of the probe electrons scattering from the Coulomb-like potential using the small angle kick approximation. A diagram of the scattering geometry is given in Figure 5.6. Assuming that that the total momentum of the relativistic electrons $p = \gamma m \beta c$ is unchanged during the interaction, the change in the transverse component of the momentum is $\Delta p_\perp = \int F_\perp dt$. For a probe MeV electron with transverse offset $b$ which crosses the $z = 0$ plane at $t = t_0$, the scattering angle from a static point charge $Q$ at the origin can be written as

\[
\chi \approx \frac{\Delta p_\perp}{p} = \frac{1}{\gamma m \beta c} \int_{-\infty}^{\infty} \frac{e Q b dt}{(b^2 + \beta^2 c^2(t - t_0)^2)^{3/2}}
= \frac{2eQ}{\gamma m \beta^2 c^2 b}
\]

which is equivalent to the Rutherford formula for small scattering angles.
Figure 5.7: The event horizon of the Coulomb field expands spherically from the instantaneously photoemitted charge.
This scattering angle assumes a static field for all \( t \). In our case, though, the charge is emitted at \( t = 0 \), i.e. \( Q(t) = Q \cdot H(t) \), where \( H \) is the Heaviside function. The Lienard-Wiechert formula must be used to compute the electric field with a time dependent charge. The field at position \( \mathbf{R} \) and time \( t \) depends on the charge at the retarded time \( t' \) satisfying \( t = t' + |\mathbf{R}|/c \) [156]. This results in the replacement of the lower integration limit of the integral in Eq. (5.1) with the time when the trajectory of a particle of transverse offset \( x = b \) and longitudinal coordinate \( z = \beta c(t - t_0) \) intersects the expanding light cone (at the event horizon as shown in Figure 5.7). For ultra-relativistic particles, \( \beta \approx 1 \) and the solution to the intersection is immediate: \( t' = (b^2/c^2 + t_0^2)/2t_0 \). For \( \beta < 1 \), a quadratic equation must be solved to find the retarded time \( t' \). The scattering angle as a function of the transverse and longitudinal position of the particle within the beam becomes

\[
\chi(b, t_0) \approx \frac{\Delta p}{p} = \frac{1}{\gamma m \beta c} \int_{v(b, t_0)}^{\infty} \frac{eQb dt}{(b^2 + \beta^2 c^2(t - t_0)^2)^{3/2}}
\]

\[
= A Q/b \cdot f(b, t_0)
\]  

(5.2)

where \( A = 2e/\gamma m \beta^2 c^2 \) and \( f(b, t_0) \to 1 \) for large \( t_0 \), recovering Eqn. (5.1) in the steady state.

In order to reconstruct the image on the detector screen, we need to apply the photoemitted charge induced kick \( \chi(b, t_0) \) on the electron trajectories. Due to the gun solenoid focusing geometry, all electron trajectories effectively come from a point source located at a distance \( L_0 \) upstream of the grid. In our model, we substitute out the transverse offset \( b \approx \alpha L_0 \) where \( \alpha \) is the electron angle coming out of the focus. Adding the small angle kick from the photoemitted charge, the
particle radial position \( r_{sc} \) on the detector screen at a distance \( L \) from the focus can be written as

\[
r_{sc}(\alpha, t_0) = \alpha L + \chi(L - L_0) = \alpha L + \frac{AQf(\alpha L_0, t_0)(L - L_0)}{\alpha L_0}
\]

It is instructive to examine the behavior of \( r_{sc} \) for different values of the initial angle \( \alpha \). If \( \alpha \) is very large, we expect \( r_{sc} \) to be very large. If \( \alpha \) is very small, then the second term in Eq. (5.3) dominates, and \( r_{sc} \to \infty \). Therefore, at each time \( t_0 \), there must be a local minimum for \( r_{sc} \), meaning that there will be no beam electrons within some \( r_{min} \) of the beam axis. This implies the creation of a “hole” shaped shadow in the transverse beam profile. We can calculate the minimum radius by solving \( dr_{sc}/d\alpha = 0 \), and the steady state result (for large \( t_0 \)) is

\[
r_{min} = 2\sqrt{\frac{L}{L_0} AQ(L - L_0)}
\]

The characteristic behavior of the hole size as a function of charge and position along the beam has been verified using particle tracking simulations to follow the trajectories of the relativistic electrons in the retarded field associated with photoemission. The simulation code reproduces the experimental shadowgraph well (see Fig. 5.10).

The model can be easily generalized from a point charge source to an extended charge distribution. The time-dependent scattering in this case is simulated considering the beam scattering from a large ensemble of macroparticles sampled from a photoemitted charge distribution that is assumed to have the same rms size as the laser spot. The main result in this case is that not all of the MeV beam electrons are expelled from the hole region. The probe electrons closer to the beam axis effectively interact with a smaller total charge, and so scatter to a smaller \( r_{min} \), causing some filling of the hole (Fig. 3a). We experimentally
Figure 5.8: Deflected mode shadowgraph simulations with variations of the photoemission model. The total charge is the same for all simulations. (top row) Instantaneous emission of charge comparing a point charge spatial distribution (top left) to a transverse gaussian distribution \( \sigma = 25 \) micron rms (top right). (bottom row) Point charge source comparing instantaneous emission (bottom left) with thermal emission, \( \tau = 1.4 \) ps (bottom right).
confirmed that the contrast of the hole region decreases when the laser spot size is increased on the target to 50 µm rms.

In Fig. 5.9 we show the measured data and respective simulations for various pump laser fluences. The fluences used are all above the ablation threshold so that the sample must be moved to a fresh spot for each data point. The hole radius is extracted from the images by fitting two step functions with finite rise and fall time to a 35 fs long temporal slice taken at the end of the probed temporal window where the hole radius is nearly constant. The relationship between the hole radius and the charge (Eq. 5.4) is used to infer the emitted charge as function of laser fluence, which is then used to generate the simulation images.

The results of the analysis are summarized in Fig. 5.11 where we plot the square of the hole size as a function of incident pump fluence (F) and calculate the corresponding charge predicted by the model (Eq. 5.4). Photoemission from a copper surface using 800 nm laser is a third order process, but the data shows
Figure 5.10: The hole edge as a function of time has been extracted from a shadowgraph. For this relatively high pump fluence, the data is well-modeled using a thermal emission process. The curve for instantaneous emission is shown for comparison.
Figure 5.11: The Rutherford scattering formula predicts the hole radius should increase as the total charge squared. With the model we can infer the total charge. In a multi-photon photoemission dominated pump fluence range, the total charge should increase as fluence cubed.

behavior significantly different from the expected $Q \propto F^3$. This is not surprising due to the large charge density at emission ($10^{12} \text{ e}^-/\text{cm}^2$) which suggests strong space charge induced saturation of the emission process.

Furthermore, it has been shown that for copper, multi-photon photoemission should be prompt [72] for laser pulses with peak intensities $< 20 \text{ GW/cm}^2$. This implies that the timescale driven to first order by the temporal profile of the incident laser pulse, and that the electron sub-system does not have time to fully thermalize before the photo-excited electrons are emitted. As observed in our experiment described in this chapter, at larger laser fluences there are some
differences between the model and the data in the shape of the hole size as a function of time (rightmost column in Fig. 5.9). An important issue known from early studies on multi-photon photoemission is the role taken by the non-thermal electron distribution generated by the ultrashort laser pulse excitation [157]. For certain materials (for example tungsten) it was verified that the charge yield follows a different power law due to the fast heating of the electrons, and that the emission occurs on time scales longer than the laser pulse, thus departing the prompt emission regime. As the pump laser intensity increases, it is clear that there is thermally-driven coupling that dominates the amount of charge emitted from the metal surface. We can refine the model to include a contribution due to a slower thermal component of the emission (Fig. 3b). By assuming $Q(t) = Q \cdot (1 - \exp(-t/\tau)) \cdot H(t)$, we obtain a good fit to the data for an emission time-constant $\tau=1.4$ ps (see Fig. 5.10) in agreement with electron-lattice coupling time-constants.

## 5.5 Conclusions

In conclusion, we have demonstrated the use of relativistic electrons to probe the electromagnetic field evolution originating from charge emission at ultrafast time scales. By using a high streaking speed RF deflecting cavity, we avoid the limitations on temporal resolution set by the electron beam bunch length or by the time-of-arrival jitter that are typical of single-shot electron probing schemes.

The record 35 fs temporal resolution achieved in our experiment allows direct observation of the speed-of-light propagation of the Coulomb field generated when a charge is emitted from a metal surface illuminated by an intense fs laser pulse. By applying a simple model to the analysis of the time-dependent shadowgraphs in the first few hundred fs, we extract the total amount of charge and the
time scale of the emission process. These findings can be applied to diagnosing ultrafast laser-induced plasma dynamics.
CHAPTER 6

Conclusions

In this thesis, we have discussed a number of interesting and unique experiments in ultrafast science that can be undertaken with ultrashort relativistic electron bunches produced from an rf photoinjector. We have showed single-shot diffraction results with an effective beam brightness that equals that of free electron laser x-ray sources. We have discussed a viable solution for relative laser-electron beam timing in spatially encoded electro-optic sampling, and verified a method for finding the absolute zero-crossing for pump-probe experiments with MeV electrons. We have demonstrated a powerful dense plasma radiography technique using an rf streak camera to observe the evolution of a photo-induced plasma with 35-femtosecond resolution.

6.1 Looking Ahead

It is undeniable that our group has completed much work in the few short years since the commissioning of the Pegasus Photoinjector Laboratory at UCLA, but there is still much work that can be done to improve the facility, and enable many more classes of ultrafast research. An S-band dual slot resonance linac (DSRL) is being constructed and will be installed at the Pegasus Laboratory in collaboration with Fartech Inc. The DSRL consists of 10 full cells plus the entrance and exit half cells, and its total length is 60 cm. It will be placed 1.1 m from the gun.
cathode, and the collimation hole and diffraction sample will be moved to 1.9 m position. The maximum energy gain of DSRL is 10 MeV. Instead of running the DSRL on the crest of the rf phase for high energy gain, it is possible to adjust its phase and amplitude to impart a negative energy-position correlation on the beam longitudinal phase space. By letting the beam propagate through a short drift section after the DSRL, its phase space rotates in the upright position and maximum compression is achieved. According to particle tracking simulations, the minimum pulse length achieved at the sample will be as short as 4 fs rms.

An ambitious goal of the community is to push the limit of current technology and take advantage of the high brightness of the intense MeV electron beam from the rf photoinjector source for imaging applications. There are two major limitations in expanding the capabilities of the MeV diffraction camera towards high spatial resolution imaging mode: i) the limited beam coherence from the metal photocathode in the RF gun and ii) the technical difficulties associated with building strong lenses for relativistic energy beams.

The first issue has been addressed by exploring a new regime of rf photoinjector operation [153]. First measurements of the beam emittance with a novel pepper-pot technique utilizing TEM grids indicate normalized emittance values lower than 50 nm. When the beam is focused to a spot size of 0.2 mm this corresponds to a transverse coherence length of 4 nm. By further reducing the spot size on the cathode to < 20 µm rms, taking advantage of progress in cathode research to reduce the thermal energy spread below 0.1 eV, particle tracking simulations predict MeV beams with < 10 nm-rad normalized emittance. The second issue can be addressed with the use of extremely strong permanent quadrupole magnets that are currently being developed by the UCLA Particle Beam Physics Laboratory.
The installation of a specially designed chamber with a jet nozzle will permit injection of gas or liquid samples into the beamline. Upgrades to the current laser system are planned to provide more energy for sample pumping. These are among the upgrades that have the potential to bolster the Pegasus accelerator’s status as a capable material science laboratory. The community of ultrafast scientists is rapidly growing, and with the techniques and diagnostics described in this thesis, rf photoinjectors will continue to gain popularity as an accessible and irreplaceable compact source producing profound ultrafast science in laboratories around the world.
Multi-Photon Photoemission

Photoemission is one of the fundamental processes in the physics of the generation of charged particle beams. In rf photoinjector sources, widely used for free-electron lasers and advanced accelerator applications, a cathode in a high field rf cavity is illuminated with photons with energy higher than the cathode work-function. Under these conditions, when a bound electron in the cathode material absorbs a single photon, it can be emitted by the surface, resulting in a linear relationship (with proportionality constant called quantum efficiency or QE) between the number of emitted electrons and the incident number of photons [158, 159].

It has long been known that for higher illumination intensities, charge will be emitted even for sub-work-function energy photons. The condition in this case is that $n$ photons have to be absorbed at the same time in order to promote an electron to an unbound state with enough kinetic energy to escape the material. The signature of the n-photon photoemission is the scaling of the emitted charge as the $n$-th power of the laser intensity [160]. On a log-log scale, the charge yields (number of electrons per number of photons) for single and multi-photon emission are lines with different slopes. Even though the $n$—photon process is statistically more rare than the single photon one, for large enough illuminating intensity, it will be advantageous to use multi-photon photoemission (MPP) to extract the maximum amount of charge from a cathode.
A limit to reaching this crossover intensity could be set by the damage threshold of the surface. Using relatively low energy but ultrashort laser pulses allows one to stay below the damage threshold (a few hundred GW/cm$^2$ for sub-ps pulses), yet access very high intensities, and hence achieve relatively large charged particle yields. In doing this, one must be careful to avoid space charge effects from the emitted electrons near the surface. The accumulation of a surface charge layer in the region close to the cathode eventually leads to the creation of a virtual cathode and cuts off further emission [161]. This can be avoided by providing a large extracting field on the cathode to quickly accelerate away the emitted charge.

In the low field regime, many papers have explored the subject of MPP from metal cathodes using short laser pulses [162, 163]. The focus of this early work was on the significant role played by thermal decoupling of the electron and the lattice after the short pulse excitation. The total charge yields found in these early experiments were not impressive as space charge and virtual cathode effects in the low accelerating field environment set a tight upper limit to the extractable surface charge density. Further, up until a few years ago, virtually all rf photoinjectors were, at least in the design stage, conceived to run with a relatively long laser pulse 510 ps on the cathode in order to limit the space charge forces and generate high brightness 1 nC class beams [164].

Given these premises, it is not surprising that the common paradigm to generate high brightness electron beams with photocathodes has been to frequency up-convert the available drive laser power to the ultraviolet (UV) to overcome the work function of the material and emit sizable beam charge. Depending on the beam applications, metals (having relatively low QE, typically between $10^{-5}$ and $10^{-4}$) have often been preferred to semiconductors in rf photoinjectors for
For MPP, it is improper to define a QE as the number of electrons is not linearly dependent on the number of photons. If we would have up-converted the photons to UV instead of sending the IR directly onto the cathode, assuming a 10% conversion efficiency, the QE required to obtain the same amount of charge their fast response, durability, and vacuum compatibility.

In this Appendix we study the properties of an electron beam generated by MPP from a very short infrared (IR) laser pulse on a Cu cathode in a rf photoinjector. We have recorded up to 50 pC of beam charge when using a 80 fs long 2 µJ of 800 nm pulse. The efficacy of this technique increases dramatically as the laser pulse length falls below a few hundred femtoseconds as is apparent from Figure A.1.

![Figure A.1: Total emitted charge vs. laser pulse length.](image_url)
would be > 0.1%, an order of magnitude higher than any metal cathode ever tested in a working rf photoinjector. The photoemission has been measured to be prompt and the thermal emittance levels are comparable to the values measured for the UV case [51].

According to the generalized Fowler-Dubridge theory for MPP, the emitted current density when photons of energy $h\nu$ illuminate the surface can be written [165] as a sum of partial currents $J_n$ where

$$J_n(h\nu) = a_n A \left( \frac{e}{h\nu} \right)^n (1 - R_\nu)^n I^n T_e^2 F \left( \frac{nh\nu - e\Phi}{k_BT_e} \right)$$  \hspace{1cm} (A.1)

in which $n$ is the order of the $n-$photon process, $a_n$ is a constant representing the probability of the multi-photon process to happen (both photon absorption, and escape from the surface), $T_e$ the electronic temperature, $A = 120$ A/cm$^2$ K$^2$ is the Richardson constant, $e$ is the electron charge, $k_B$ and $h$ are the Boltzmann and the Planck constant, and $R_\nu$ is the reflectivity coefficient at the frequency $\nu$ and $\Phi$ is the cathode work function modified by the Schottky effect. The Fowler function, represented in terms of a di-logarithmic function: $F(x) = Li_2(x) = \int_0^\infty \ln[1 + \exp(-x - y)] \, dy$, strongly suppresses the terms for which its argument is negative. Since the $a_n$, representing the likelihood of a $n-$photon process, quickly decrease with increasing $n$, the dominant term in the sum is given by the lowest $n$ such that $n \times h\nu > e\Phi$.

For laser intensities larger than a critical value $I_{cr} \geq \left( \frac{e}{h\nu} \right) \sqrt{\frac{a_1(1-R_\nu)}{3a_3(1-R_\nu)^3}}$ for which $J_3(h\nu) \geq J_1(3h\nu)$, the 3-photon process yields more electrons than the single photon emission process. (See Figure A.2 for the crossover at $I_{cr}$). The $a_n$ are material dependent and difficult to calculate theoretically. Using order of magnitude estimates for $a_1 \sim 10^{-14}$ cm$^2$A$^{-1}$ and $a_3 \sim 10^{-34}$ cm$^6$A$^{-3}$ [166, 167] and assuming similar reflectivities at IR and UV wavelengths, $I_{cr} \approx 10$ GW/cm$^2$. 

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Figure A.2: For a high enough laser intensity, the multi-photon charge yield (here represented in terms of charge density) surpasses the single-photon emission.

Generally, solid state laser media have gain bandwidth centered in the near IR region (and so below the work function of most metal cathodes) so that the higher energy photons are usually obtained by frequency multiplying the IR laser pulses. Since the harmonic conversion (usually taking place in non linear crystals) is lossy, the intensity threshold over which employing a $n-$photon emission process starts to be convenient is practically even lower than $I_{cr}$.

The use of MPP for efficiently generating charge was experimentally explored at Pegasus lab, using the photoinjector setup as described elsewhere in this thesis. In this study, the gradient in the rf gun was be varied between 55 MV/m and 90 MV/m by changing the input rf power. This was to establish a clear dependence
The laser spot size on the cathode is 900 $\mu$m rms. The roll-off toward saturation is apparent in the lowest extraction field plot. The total charge yield as a function of laser pulse energy and gun extraction field are visible in Figure A.3.

As a final practical note, one might argue that the MPP process is more sensitive to energy fluctuations (jitter) and spatial inhomogeneity of the laser pulse than conventional photoemission. In practice, the UV laser pulse used to overcome the cathode work function in conventional rf photoinjectors is created by non-linear processes starting from an IR pulse and its characteristics (energy...
stability and spatial inhomogeneity) also depend on the third power of the initial IR laser pulse intensity.

In conclusion, significant charge yield is possible through MPP using modest levels of laser energy incident on metal cathodes. Because of the tremendous progress in ultrashort laser systems and the new developments in photoinjector beam dynamics, it becomes feasible to drive a photoinjector with sub-work-function laser photons. For this operating mode, the favorable scaling with laser intensity is even more important than the measured emission efficiency, which is influenced by many factors including the surface quality.

The explanation of the larger yields measured compared to early papers on this subject are to be sought in the very high electric field on the cathode and the ultrashort pulse length which enables us to reach high intensities without incurring surface damage. These results have the potential to revolutionize the field of electron beam generation in rf photoinjectors by greatly simplifying the requirements (and the cost) of the photoinjector driver laser system which typically starts from IR photons and uses non-linear optics to generate photons of enough energy to overcome the material work function. Going to MPP eliminates some level of complexity involved with converting the IR photons to UV photons, while still producing a high brightness ultrashort electron beam.
References


[99] Lionel Duvillaret, Stéphane Rialland, and Jean-Louis Coutaz. Electro-optic sensors for electric field measurements. i. theoretical comparison among


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