

Attoseconds and the exascale: on laser plasma surface interactions



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Abstract

The commissioning of multi-petawatt class laser facilities around the world is gathering pace. One of the primary motivations for these investments is the acceleration of high-quality, low-emittance electron bunches. Here we explore the interaction of a high-intensity femtosecond laser pulse with a mass-limited dense target to produce MeV attosecond electron bunches in transmission and confirm with three-dimensional simulation that such bunches have low emittance and nano-Coulomb charge. We then perform a large parameter scan from non-relativistic laser intensities to the laser-QED regime and from the critical plasma density to beyond solid density to demonstrate that the electron bunch energies and the laser pulse energy absorption into the plasma can be quantitatively described via the Zero Vector Potential mechanism. These results have wide-ranging implications for future particle accelerator science and associated technologies.

Contents

List of publications	xi
List of Figures	xiii
List of Symbols and Abbreviations	xvii
1 Introduction	1
1.1 High-energy-density physics	2
1.2 Electromagnetism fundamentals	4
1.3 The definition of a classical plasma	5
1.3.1 The Debye length	6
1.3.2 The plasma parameter	7
1.3.3 Collisionality and the plasma frequency	8
1.4 The Lawson-Woodward theorem	10
1.5 Laser-solid density plasma linear interaction	10
1.6 Relativity	12
1.6.1 Ultra-relativistic similarity theory	13
1.6.2 Relativistic lasers and plasmas	15
1.6.3 Conservation of generalised transverse momentum	16
1.7 QED effects	17
1.7.1 High-energy photon emission and radiation reaction	17
1.7.2 Multi-photon Breit-Wheeler pair production	19
1.8 Simulating the interaction	19
1.8.1 Supercomputing resources	19
1.8.2 Particle-In-Cell codes	20
1.8.3 Hydrodynamic codes	27
1.9 Generating the interaction	29
1.10 Summary	32

2 The Zero Vector Potential Absorption Mechanism	33
2.1 Motivations and an overview	33
2.2 Introduction	35
2.2.1 ZVP electron bunch energies	42
2.2.2 ZVP bunches oblique incidence scaling	45
2.2.3 Defining characteristics of the ZVP mechanism	47
2.3 Numerical simulations of the ZVP mechanism	48
2.3.1 The ZVP mechanism in 3D3V	48
2.4 The ZVP electron bunch	55
2.4.1 Energy absorption in the ZVP regime	66
2.4.2 Unpacking the QED effects of Figure 2.12	68
2.4.3 Errors	70
2.5 Conclusions	71
3 Attosecond X-ray harmonics on the ORION laser facility	73
3.1 Introduction	74
3.2 Theory	75
3.2.1 The ROM model	75
3.2.2 The normalisation factor	80
3.2.3 Hole boring	81
3.2.4 Harmonic source size	85
3.2.5 The SHHG optimal parameter space	86
3.3 Simulations	88
3.3.1 1D PIC simulations of SHHG	88
3.3.2 Hydrodynamic simulations of preplasma formation	93
3.4 The experiment	99
3.4.1 Target chamber geometry and polarisation	99
3.4.2 Targets	102
3.4.3 Contrast and plasma mirrors	103
3.5 Experimental data processing	104
3.5.1 Image plate calibration	104
3.5.2 OHREX calibration	106
3.5.3 Extracting the data	107
3.5.4 KBRXM	110
3.6 Experimental results	111
3.7 Summary and discussion	115

4 Attosecond electron bunches and X-ray pulses on GEMINI PW	117
4.1 Overview	117
4.2 X-ray harmonics on GEMINI PW	119
4.3 Observation of ZVP electron bunches	121
4.4 Application of XHHG to white light Laue diffraction	127
4.5 Contrast	128
4.5.1 Contrast on GEMINI PW	128
4.5.2 Kahaly mirror	132
4.6 Summary	133
5 Summary and future work	135
5.1 Summary	135
5.2 Beyond GEMINI PW and future work	136

Appendices

A General plasma physics	141
A.1 Lorentz transformations of electromagnetic fields	141
A.2 The headlight effect	142
A.3 Geometric transverse emittance	142
A.4 The Bourdier method	145
B PIC simulations	149
B.1 Convergence of 3D PIC simulations	149
References	151

x

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List of Figures

1.1	Laser systems across the globe, both commissioned, planned and theorised.	3
1.2	A diagram to illustrate the derivation of plasma frequency.	9
1.3	A schematic of the PIC code loop and the algorithms performed.	21
1.4	A representation of the staggered Yee grid.	22
1.5	Smilei simulation box decomposition into cells, patches and MPI patch collections.	24
1.6	Representation of the interaction of the ARCHER2 hardware and software components when running Smilei.	25
1.7	Image of the ORION SP1 double beamlet structure in the near-field.	30
2.1	Diagram of a p-polarised laser pulse incident at angle θ specularly reflected from a solid density plasma.	36
2.2	Diagram of a <i>p</i> -polarised laser pulse incident on an ablating overdense plasma.	39
2.3	The numerical simulation using a 3D Particle-In-Cell (PIC) code of the Zero Vector Potential (ZVP) mechanism.	49
2.4	Propagation of a vector potential zero through the ablating ZVP electron bunch.	53
2.5	Electron dynamics in 3D PIC simulation for both linear and circularly polarised relativistic laser pulses.	54
2.6	The Fourier transform of the reflected laser pulse in 3D PIC simulations.	55
2.7	2D PIC simulation results qualitatively describing typical mass-limited ZVP electron bunch structure.	57
2.8	Energy spectra for mass-limited electron bunches formed via linearly and circularly polarised laser pulses.	59
2.9	Mean mass-limited ZVP electron bunch normalised kinetic energies extracted from 2D PIC simulations.	61
2.10	Mass-limited electron bunch train length as a function of laser intensity and plasma density.	62
2.11	The relative errors for each mean energy data point compared to Figure 2.9.	65

2.12	Peak instantaneous bulk electron bunch total energy escaping to the plasma bulk rear.	66
2.13	An exploration of ZVP 2D PIC simulation stability.	71
3.1	2D PIC simulation of SHHG beaming effect via hole boring.	82
3.2	Typical 1D SHHG Smilei simulation results	90
3.3	Comparison between simulation and analytical predictions of hole boring and SHHG normalisation factor.	91
3.4	The effect of the PIC code particle shape function on numerical heating.	92
3.5	Reflected beam harmonic content up to the keV range in a high-resolution 1D PIC simulation.	93
3.6	Attosecond pulse train from the filtered reflected laser pulse.	94
3.7	Silicon dioxide plasma mirror switch on from irradiation by the SP2 main pulse.	95
3.8	The attenuation of the SP2 laser pulse as it propagates through a switched-on PM.	96
3.9	Typical preplasma formation from the incidence of the ORION SP2 laser prepulse on a plastic target.	96
3.10	Preplasma scale length parameter scan.	97
3.11	Typical electron temperature of a plastic target after irradiation by a petawatt class laser prepulse.	98
3.12	ORION XHHG experiment target chamber set up.	100
3.13	ORION target chamber geometry	101
3.14	ORION XHHG experiment targets	103
3.15	Unprocessed IP from ORION experiment	108
3.16	Typical ORION experiment uncalibrated IP response quartz ($10\bar{1}0$) crystal and Fourier transform	109
3.17	Typical ORION experiment calibrated IP response for the quartz ($10\bar{1}1$) crystal.	111
3.18	Dependence of the harmonic beam spectral intensity on laser spot size at 2.405 keV.	112
3.19	X-ray harmonic intensities measured on the ORION experiment compared to theory and simulation.	113
3.20	The ORION result in context.	116
4.1	XHHG on the GEMINI PW laser system.	121
4.2	Planned GEMINI-PW experimental setup for the measurement of ZVP electron bunches.	122
4.3	Attosecond ZVP electron bunch targets	124
4.4	Native contrast of the GEMINI PW laser system.	128

A.1	Emittance calculation for an ideal Gaussian distribution in phase space.	144
A.2	145
B.1	Comparison between the initial 3D simulation and a lower resolution version.	150

List of Symbols and Abbreviations

Note All quantities are defined in SI units unless otherwise specified.

α	Twiss parameter = $-\frac{\langle x_i x'_i \rangle}{\epsilon_{\text{rms}}^i}$
a_0	Normalised vector potential = $\frac{e \mathbf{E}_L }{m_e c \omega_L}$
A	Atomic mass number
\mathbf{A}	Three-vector potential
\mathbf{A}^μ	Four-vector potential = $(\phi/c, \mathbf{A})$
β	Normalised speed = v/c , or Twiss parameter = $\frac{\langle x_i \rangle}{\epsilon_{\text{rms}}^i}$
β	Normalised velocity = \mathbf{v}/c
\mathbf{B}	Magnetic field
\mathbf{B}_L	Magnetic field of a laser pulse
c	Speed of light = $3.00 \times 10^8 \text{ m s}^{-1}$
δ	Skin depth
$\delta(\mathbf{x})$	Dirac-delta function
$d\mathbf{s}$	Vector line element
Δ	Change in a variable
D	Number of dimensions
∇	Partial derivative = $\hat{\mathbf{x}}_\mu \frac{\partial}{\partial x_\mu}$, $\mu = x, y, z$
ϵ_0	Permittivity of free space = $8.854 \times 10^{-12} \text{ F m}^{-1}$
ϵ_{rms}	Transverse geometric emittance
$\epsilon_{n,\text{rms}}$	Transverse normalised emittance
e	Absolute charge of an electron = $1.602 \times 10^{-19} \text{ C}$
e^\pm	Electron (−) or positron (+)
\mathbf{E}	Electric field
E_S	Schwinger electric field = $1.3 \times 10^{18} \text{ V m}^{-1}$
\mathbf{E}_L	Electric field of a laser pulse

f	Distribution function
\mathbf{F}_L	Lorentz force = $q(\mathbf{E} + \mathbf{v} \times \mathbf{B})$
γ	Lorentz/gamma factor = $\frac{1}{\sqrt{1-\beta^2}}$, or Twiss parameter = $\frac{\langle x_i'^2 \rangle}{\epsilon_{rms}^i}$, or high-energy photon
η	Efficiency
I	Electromagnetic field intensity
\mathcal{H}	Hamiltonian
θ	Angle of incidence of a laser pulse
\mathbf{J}	Current density
\mathbf{k}	Electromagnetic three-wave vector
\mathbf{K}^μ	Electromagnetic four-wave vector = $(\omega/c\mathbf{k})$
K	Boltzmann constant = $1.38 \times 10^{-23} \text{ J K}^{-1}$
λ_D	Debye length $\equiv \sqrt{\frac{\epsilon_0 K T_e}{n_e e^2}}$
λ_L	Laser pulse wavelength
Λ_ν^μ	Lorentz transformation matrix
L	Length
\mathcal{L}	Lagrangian
μ_0	Vacuum permeability = $1.257 \times 10^{-6} \text{ N A}^{-2}$
m_e	Mass of an electron = $9.11 \times 10^{-31} \text{ kg}$
n	Harmonic order
n, N	Number
n_c	Plasma critical density = $\frac{\omega_L^2 m_e \epsilon_0}{e^2}$
n_e	Plasma electron number density
\bar{n}_e	Normalised plasma electron number density = $\frac{n_e}{n_c}$
n_i	Plasma ion number density
\mathbf{n}	Vector normal to a surface
N_A	Avogadro's number
N_D	Number of particles within the Debye sphere = $\frac{4}{3}\pi \lambda_D^3 n$
ω	Angular frequency of an oscillation or low energy photon
ω_L	Laser pulse angular frequency
ω_p	Plasma frequency = $\sqrt{\frac{e^2 n_e}{m_e \epsilon_0}}$

ϕ	Angle of laser polarisation out of the plane of interaction, or scalar electromagnetic potential
Φ, Ψ	Phase
\mathbf{p}	Three-momentum
\mathbf{P}	Pressure
\mathbf{P}^μ	Four-momentum = $(U/c, \mathbf{p})$
Q	Charge
ρ	Density
r_L	Relativistic Larmor radius = $\frac{\gamma m_e v}{e \mathbf{B} }$
\mathbf{r}	Radial vector
$\hat{\mathbf{r}}$	Radial unit vector
R	Rate
σ	Surface area
S	Relativistic similarity parameter = \bar{n}_e/a_0
$S(\mathbf{x})$	Shape function
τ	Average time or standard deviation of time
t	Time
T	Kinetic energy
T_e	Plasma electron temperature
u, v	Speed
\mathbf{u}, \mathbf{v}	Velocity
U	Total energy
v_ϕ	Phase velocity
V	Potential or volume
w_L	Beam waist
w_p	Quasi-particle weight
χ_γ	Photon quantum parameter
x, y, z	Standard Cartesian coordinates in 3D
$\mathbf{x}, \mathbf{y}, \mathbf{z}$	Vectors along each axis of the Cartesian coordinate system
$\hat{\mathbf{x}}, \hat{\mathbf{y}}, \hat{\mathbf{z}}$	Unit vectors along each axis of the Cartesian coordinate system
Z	Ion charge state in units of e

Subscripts . . .

C	Pseudocapacitor
e	Electron
γ	Photon
L	Laser pulse or Longitudinal
T	Transvere to a laser pulse
pol	Along the polarisation vector of a laser pulse
\perp	Perpendicular
\parallel	Parallel
i, j	Indices
s	Species
x, y, z	Along the specific Cartesian axes
1D, 2D, 3D . . .	One-, two- or three-dimension(al)
AOI	Angle of Incidence
AR	Anti-Reflective
ARP	Apparent Reflection Point
BBXRD	BroadBand X-Ray Diffraction
BW	Breit-Wheeler
CLF	Central Laser Facility
CPA	Chirped Pulse Amplification
CSE	Coherent Synchrotron Emission
DPM	Double Plasma Mirror
FDTD	Finite Difference Time Domain
HB	Hole Boring
HED	High-Energy-Density
HHG	High Harmonic Generation
HPC	High Performance Computing
OHREX	ORION High Resolution X-ray
OMM	Oscillating Mirror Model
PIC	Particle-In-Cell
PM	Plasma Mirror

QED	Quantum Electro-Dynamics
ROM	Relativistic Oscillating Mirror
RPM	Relativistic Plasma Mirror
RR	Radiation Reaction
SF-QED	Strong-Field Quantum Electro-Dynamics
SHHG	Surface High Harmonic Generation
TCC	Target Chamber Centre
XHHG	X-ray High Harmonic Generation
VLA	Vacuum Laser Acceleration
ZVP	Zero Vector Potential

1

Introduction

Contents

1.1	High-energy-density physics	2
1.2	Electromagnetism fundamentals	4
1.3	The definition of a classical plasma	5
1.3.1	The Debye length	6
1.3.2	The plasma parameter	7
1.3.3	Collisionality and the plasma frequency	8
1.4	The Lawson-Woodward theorem	10
1.5	Laser-solid density plasma linear interaction	10
1.6	Relativity	12
1.6.1	Ultra-relativistic similarity theory	13
1.6.2	Relativistic lasers and plasmas	15
1.6.3	Conservation of generalised transverse momentum	16
1.7	QED effects	17
1.7.1	High-energy photon emission and radiation reaction . .	17
1.7.2	Multi-photon Breit-Wheeler pair production	19
1.8	Simulating the interaction	19
1.8.1	Supercomputing resources	19
1.8.2	Particle-In-Cell codes	20
1.8.3	Hydrodynamic codes	27
1.9	Generating the interaction	29
1.10	Summary	32

1.1 High-energy-density physics

We sit on the verge of the exascale revolution, in both computing resources [1] and laser power. With rapid adoption of the pioneering advancements of Chirped Pulse Amplification (CPA) by Strickland and Mourou [2] and of Optical Parametric Chirped Pulse Amplification [3], multi-petawatt facilities have been commissioned and built across the globe and plans for sub-exawatt facilities are now underway. Figure 1.1 details the parameter space accessible by current and proposed facilities [2, 4–68], clearly, there is no indication of wavering interest or support for the field of High-Energy-Density (HED) physics.

HED physics is the laboratory study of the behaviour of matter with a pressure above 10 GPa, approximately one million atmospheres, and containing free electrons not confined to a solid state [72], typically in the plasma state of matter. HED conditions are found for a vast range of densities and temperatures (from zero to a million million Kelvin) and operating in both the quantum and relativistic realms. The applications are equally diverse, including but not limited to inertial confinement fusion, particle acceleration for scientific or medical purposes and light sources as diagnostic tools. Ubiquitous in the natural universe, beyond our solar system, all that can be observed in the sky is radiation emitted from HED plasmas [73].

Independently, the field of attosecond physics has flourished [74]. By irradiating gas targets with mildly ionising laser pulse intensities, pulses of attosecond radiation, composed of high-order harmonics of the laser, have been generated. Recognised by the 2023 Nobel Prize for Physics, the research of Agostini, Krausz and Huillier have utilised this High Harmonic Generation (HHG) to unveil the inner workings of atoms and molecules, tracking fundamental electronic dynamics [75].

At the intersection of HED and attosecond physics, there is another course for the generation of harmonics with the potential to produce brighter and shorter pulses than those of the laser-gas interaction. This thesis concerns itself with that interaction, that of a high-power short-pulse laser incident on a flat solid target. Through this Surface High Harmonic Generation (SHHG), state-of-the-art 10 PW laser facilities such as ELI-np [19] can produce electron bunches and light pulses of

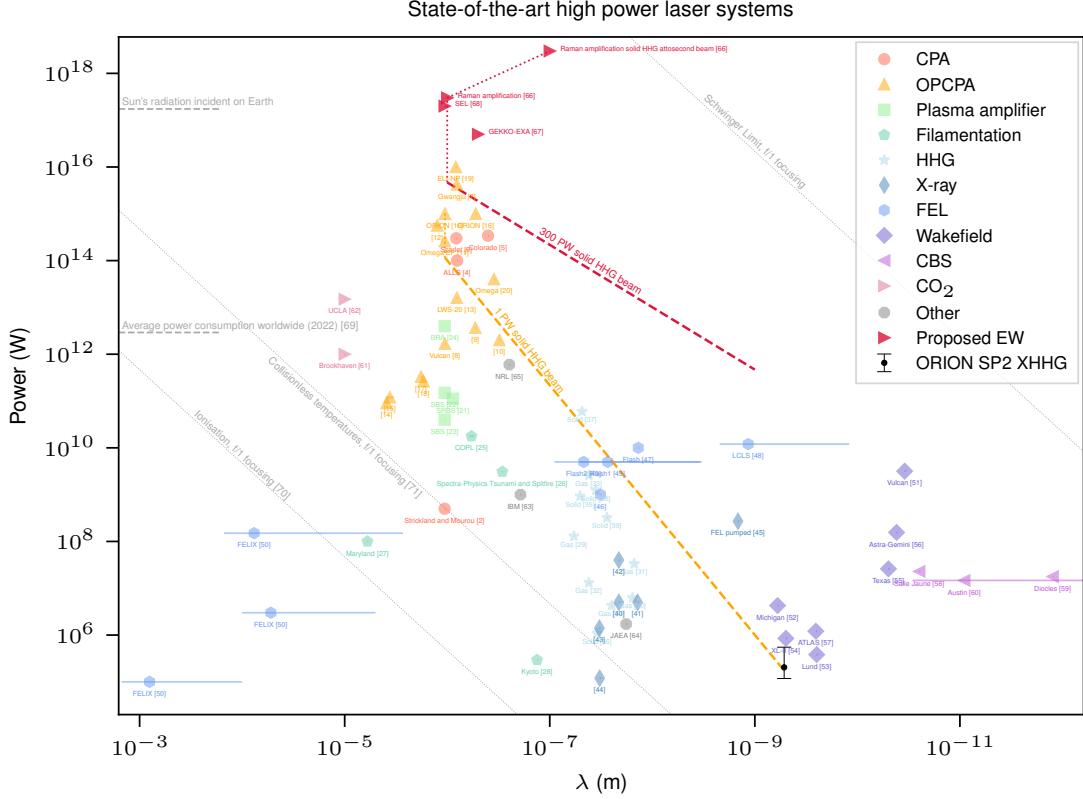


Figure 1.1: A by no means exhaustive plot of high power light producing facilities from across the globe, both commissioned, planned and theorised [2, 4–71] to provide an overview of the parameter space currently and soon to be accessible. The experimental result obtained at the ORION laser facility [16] is included, and will be discussed in detail in chapter 3. The dashed orange line is from the relevant theoretical model. The red dashed line marks the parameter space attainable via the methods discussed in this thesis when coupled with a sub-ettawatt laser pulse. Such a beamline would provide intensities in the water window many orders of magnitude beyond that currently available from state-of-the-art facilities.

exceptional charge and brightness, and of attosecond duration, thus uncovering new avenues for attosecond resolution diagnostics and for probing vacuum non-linearities.

Seemingly counter-intuitively, as the laser power increases, via relativistic effects and for particular initial conditions, greater coherency in electron dynamics can be observed and signals are amplified. Before delving into this fascinating phenomenon, the remainder of this chapter provides some of the relevant background information. Chapter 2 introduces the ZVP model of attosecond absorption to describe this laser-solid interaction. Chapter 3 focuses instead on the reflected radiation and its high harmonic content, specifically with regard to the measurement of X-

ray harmonics on the March 2023 ORION experiment. This chapter accessed a moderately different regime to that of ZVP. Chapter 4 brings together both absorption and reflection in the ZVP regime, detailing the upcoming GEMINI PW experiment. Finally, Chapter 5 summarises the body of work covered by this thesis and discusses potential future work.

1.2 Electromagnetism fundamentals

The spatiotemporal propagation of electric $\mathbf{E}(t, \mathbf{x})$ and magnetic $\mathbf{B}(t, \mathbf{x})$ fields must satisfy Maxwell's equations [73]

$$\nabla \cdot \mathbf{B} = 0, \quad (1.1a)$$

$$\nabla \cdot \mathbf{E} = \frac{\rho}{\epsilon_0}, \quad (1.1b)$$

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \mu_0 \epsilon_0 \partial_t \mathbf{E}, \quad (1.1c)$$

$$\nabla \times \mathbf{E} = -\partial_t \mathbf{B}. \quad (1.1d)$$

Here, $\epsilon_0 = 8.85 \times 10^{-12} \text{ F m}^{-1}$ and $\mu_0 = 1.26 \times 10^{-6} \text{ N A}^{-2}$ are the vacuum permittivity and permeability respectively and $\rho(t, \mathbf{x})$ and $\mathbf{J}(t, \mathbf{x})$ the total charge and current densities of the charged particles present in the system.

A particle with charge q and velocity \mathbf{v} in the presence of electromagnetic fields experiences the Lorentz force,

$$\mathbf{F}_L = q(\mathbf{E} + \mathbf{v} \times \mathbf{B}). \quad (1.2)$$

The electromagnetic fields can be obtained from the scalar, ϕ , and vector, \mathbf{A} , potentials as [76]

$$\mathbf{E} = -\nabla\phi - \partial_t \mathbf{A}, \quad (1.3)$$

$$\mathbf{B} = \nabla \times \mathbf{A}. \quad (1.4)$$

The Vlasov-Maxwell system of equations

A collisionless and fully ionised plasma is fully described in the kinetic description by the Vlasov-Maxwell system of equations [77]. Each plasma species, s , of particles with mass m_s and charge q_s is represented by its distribution function $f_s(t, \mathbf{x}, \mathbf{p})$ at time t , position \mathbf{x} and momentum $\mathbf{p} = m_s\gamma\mathbf{v}$. The distribution satisfies the Vlasov equation, that is,

$$\left(\partial_t + \frac{\mathbf{p}}{m_s\gamma} \cdot \nabla + \mathbf{F}_L \cdot \nabla_{\mathbf{p}} \right) f_s = 0, \quad (1.5)$$

where \mathbf{F}_L is the Lorentz force given in equation 1.2. The electric $\mathbf{E}(t, \mathbf{x})$ and magnetic $\mathbf{B}(t, \mathbf{x})$ fields that generate the force must satisfy Maxwell's Equations (Equations 1.1).

This self-consistent system of equations describes the dynamics of plasma particles within electromagnetic fields. The particles modify the fields via their charge and current densities,

$$\rho(t, \mathbf{x}) = \sum_s q_s \int d^3 p f_s(t, \mathbf{x}, \mathbf{p}), \quad (1.6)$$

and

$$\mathbf{J}(t, \mathbf{x}) = \sum_s q_s \int d^3 p \mathbf{v} f_s(t, \mathbf{x}, \mathbf{p}), \quad (1.7)$$

respectively.

1.3 The definition of a classical plasma

As outlined in F. Chen's definitive textbook 'Introduction to Plasma Physics and Controlled Fusion' [73], a plasma must fulfil three criteria, namely,

1. Ionisation: a plasma must consist of both charged and neutral particles. Of course, this alone cannot define a plasma, any gas will contain some degree of ionisation. Note, with reference to figure 1.1 [70], that upon incidence a modern high-power laser system will instantaneously fully ionise a target.

2. Quasineutrality: while locally there can be (often extreme) electromagnetic forces and charge concentrations at work, over the length scales of the plasma, such forces are screened out and the plasma bulk remains net neutral in charge.
3. Collective behaviour: unlike in a gas, where collisions dominate the dynamics, the particles in a plasma generate electromagnetic fields that interact at a distance. Thus a particle's motion depends not only on its immediate vicinity but on the surrounding plasma conditions. Indeed, it is often the so-called *collisionless* plasmas, where collisions can be safely neglected, that are of most interest, as is the focus of this thesis.

These conditions can be quantitatively defined by the Debye length, the plasma parameter and the plasma frequency as laid out in the following sections.

1.3.1 The Debye length

The Debye length describes the extent to which a plasma can shield electromagnetic fields within and so remain quasineutral. Consider an infinitely extended plasma with a test charge placed at some point. What then is the scalar potential $\phi(\mathbf{x})$ around it? If the plasma had no kinetic energy, the charged particles would arrange themselves immediately adjacent to the test charge and once this equilibrium state was reached there would be no electromagnetic fields present. More realistically, the plasma will have some temperature, likely a very large temperature, and so some particles will be able to escape the potential of the test charge and thus leak electromagnetic fields into the plasma bulk. Poisson's equation (equation 1.1b in the static case) reads

$$\epsilon_0 \nabla^2 \phi = -e(Zn_i - n_e), \quad (1.8)$$

where $\epsilon_0 = 8.854 \times 10^{-12} \text{ F m}^{-1}$ is the permittivity of free space, $e = 1.602 \times 10^{-19} \text{ C}$ is the charge of an electron, Z is the plasma ion charge in units of e and n_i and n_e are the number densities of plasma ions and electrons respectively.

Since the electrons are significantly more mobile than the ions due to their lower mass, it is in general the electrons and not the ions that respond to the test

charge and the ions can be assumed to provide a constant background of positive charge density. If the number density of electrons follows a Boltzmann temperature distribution in the presence of a potential energy $-e\phi$, then

$$n_e = n_{e,0} e^{e\phi/KT_e}, \quad (1.9)$$

where $n_{e,0}$ is the electron number density far from the test charge, $n_i = n_{e,0}/Z$, $K = 1.38 \times 10^{-23} \text{ J K}^{-1}$ is the Boltzmann constant and T_e is the electron temperature in Kelvin. Note that in plasmas it is very common for different species to have differing temperatures depending on the mechanism for energy absorption and the timescales for collisions compared to the timescale of the study.

Substituting equation 1.9 into equation 1.8 and Taylor expanding the exponential term in the limit that the plasma is weakly coupled ($e\phi \ll KT_e$),

$$\nabla^2 \phi = \frac{\phi}{\lambda_D^2}, \quad (1.10)$$

where

$$\lambda_D \equiv \sqrt{\frac{\epsilon_0 K T_e}{n_e e^2}}, \quad (1.11)$$

is the *Debye length* and describes the thickness of the charge sheath surrounding the test charge. For quasineutrality to hold for the plasma bulk, its spatial dimensions, L , must extend beyond a few Debye lengths, *i.e.*

$$L \gg \lambda_D. \quad (1.12)$$

1.3.2 The plasma parameter

In order for the derivation of section 1.3.1 to be statistically valid, there must be a large number of charged particles within the shielding sheath. The number of particles within the *Debye sphere* is

$$N_D = \frac{4}{3} \pi \lambda_D^3 n, \quad (1.13)$$

where, N_D is the *plasma parameter*. Note that, as discussed already, in most cases it is most suitable to choose the number density n to be the number density of

electrons, n_e . To ensure the plasma is suitably ionised (criterion 1) and that the plasma engages in collective behaviour (criterion 3),

$$N_D \ggg 1. \quad (1.14)$$

Quite the reverse is necessary for the creation of a quantum plasma.

1.3.3 Collisionality and the plasma frequency

Collective behaviour depends not only on the ability for large numbers of particles to interact via electromagnetic forces but also that these forces dominate over collisions in describing particle trajectories. Taking ω as the typical frequency of plasma oscillations and τ as the average time between collisions, for a plasma (as opposed to a gas)

$$\omega\tau > 1 \quad (1.15)$$

is required. It now remains to determine what is the typical frequency of collisions in a given plasma. While the types of plasma waves and their associated frequencies of oscillation are multitudinous, the characteristic frequency, the *plasma frequency*, ω_p , naturally arises from the most straightforward. It describes the response of electrons to charge imbalances within an infinite uniform plasma at rest in the absence of magnetic fields or temperature fluctuations. As in Section 1.3.1, the ions provide a constant background of positive charge.

Consider a semi-infinite plasma existing for $x > 0$, with electron density n_e and ion density n_e/Z of charge state Z^1 . Suppose the electron fluid is displaced by some perfectly isotropic force into the plasma bulk a distance $(\Delta x)\hat{x}$ as in Figure 1.2. The total charge of displaced electrons within a surface area of σ is

$$Q = -en_e\sigma\Delta x. \quad (1.16)$$

¹This description has direct relevance to the Zero Vector Potential mechanism which will be made clear in Chapter 2.

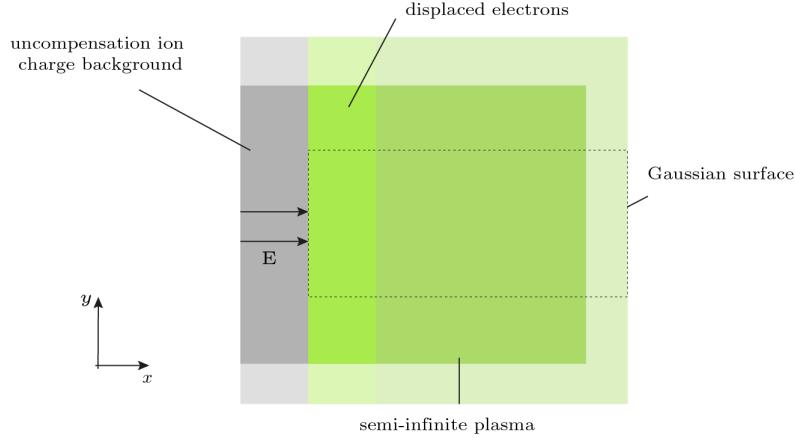


Figure 1.2: A diagram to illustrate the derivation of plasma frequency. The electrons of a semi-infinite plasma are displaced inwards by some external force leaving in their wake an uncompensated space charge of ‘immobile’ positive ions. By constructing a Gaussian surface along the dashed line, the electric field associated with the positive space charge can be calculated from Gauss’ Law.

Applying the integral form of Gauss’ law (from equation 1.1b) to the surface described in Figure 1.2, the uncompensated charge leads to

$$-\sigma E \hat{\mathbf{x}} = \frac{Q}{\epsilon_0} \hat{\mathbf{x}} = -\frac{en_e \sigma \Delta x}{\epsilon_0} \hat{\mathbf{x}} \quad (1.17)$$

at the plasma surface. By the Lorentz force, Equation 1.2, the displaced electrons will experience a restoring force, $-eE\hat{\mathbf{x}}$, perpendicular to the surface due to the electron-ion charge imbalance. The equation of motion for electrons on that surface is therefore

$$m_e \frac{d^2 \Delta x}{dt^2} = -eE = -\frac{e^2 n_e}{\epsilon_0} \Delta x. \quad (1.18)$$

Equation 1.18 clearly describes a simple harmonic oscillator with a characteristic frequency given by the plasma frequency,

$$\omega_p = \sqrt{\frac{e^2 n_e}{m_e \epsilon_0}}. \quad (1.19)$$

Interestingly, the collisionality condition is always satisfied for a classical Maxwellian plasma.

1.4 The Lawson-Woodward theorem

The Lawson-Woodward theorem states that there can be no net electron energy gain from laser fields [78], quite at odds with one of the primary aspects of this thesis, that is, the acceleration of electrons. There are, however, several conditions that are frequently met, namely,

1. The interaction region is infinite.
2. The interaction occurs in a vacuum.
3. The electron is ultra-relativistic ($v \approx c$) along the acceleration gradient.
4. No electro- or magnetostatic fields are present.
5. Non-linear effects are neglected.

Several of these apply to the various accelerations of electrons considered here. It is this final condition that is most damning to the application of the theorem. Throughout this thesis, the ultra-relativistic laser pulses under consideration ensure non-linear effects cannot be neglected. It is indeed such non-linearities that are of most interest.

1.5 Laser-solid density plasma linear interaction

Consider small transverse electromagnetic waves propagating through a plasma. Linearising equation 1.2 for a single plasma electron by assuming only small field variations and thus small velocity variation,

$$m_e \dot{\mathbf{v}}_e = -e \mathbf{E}. \quad (1.20)$$

Combining the time derivative of Equation 1.1c and the curl of Equation 1.1d,

$$\nabla \times \nabla \times \mathbf{E} = -\mu_0 \dot{\mathbf{J}} - \mu_0 \epsilon_0 \ddot{\mathbf{E}}. \quad (1.21)$$

Considering only fast oscillations, such that ions are effectively immobile,

$$\mathbf{J} = -n_e e \mathbf{v}_e \quad (1.22)$$

and using the identity $\nabla \times \nabla \times \mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E}$,

$$\nabla(\nabla \cdot \mathbf{E}) - \nabla^2 \mathbf{E} = -\frac{\mu_0 n_e e^2}{m_e} \mathbf{E} - \mu_0 \epsilon_0 \ddot{\mathbf{E}}. \quad (1.23)$$

Assuming plane wave solutions of the form

$$\mathbf{E} = \mathbf{E}_0 e^{i(\mathbf{k} \cdot \mathbf{x} - \omega t)}, \quad (1.24)$$

where \mathbf{k} is the wave-vector and ω the frequency of oscillations and noting the waves are transverse $\mathbf{k} \cdot \mathbf{E} = 0$,

$$|\mathbf{k}|^2 \mathbf{E} = -\frac{\mu_0 n_e e^2}{m_e} \mathbf{E} + \mu_0 \epsilon_0 \omega^2 \mathbf{E} \quad (1.25)$$

and hence the dispersion relation for electromagnetic waves propagating in a plasma is

$$\omega^2 = c^2 |\mathbf{k}|^2 + \omega_p^2. \quad (1.26)$$

Equation 1.26 exhibits a *cutoff* dependent on the plasma density via ω_p . The *critical density*, n_c , is defined as the density above which a laser pulse of frequency ω_L cannot propagate through a plasma. This occurs for $\omega_L = \omega_p$, thus,

$$n_c = \frac{m_e \epsilon_0 \omega_L^2}{e^2}. \quad (1.27)$$

As the plane wave has a spatial dependence $\sim \exp(\mathbf{k} \cdot \mathbf{x})$, if $n_e > n_c$, \mathbf{k} is imaginary and the wave no longer propagates through the plasma and instead exponentially attenuates over a skin depth,

$$\delta = \frac{1}{|\mathbf{k}|} = \frac{c}{\sqrt{\omega_p^2 - \omega_L^2}} \quad (1.28)$$

and is reflected. For typical high-power lasers with wavelengths in the visible or near-infrared, fully ionised solids tend to have densities well above the critical density and thus produce plasma mirrors.

1.6 Relativity

Modern high-power lasers operate in the domain of relativistic mechanics and in general interactions are highly non-linear. It is useful to introduce some of the basic principles of relativity. There has been growing interest in the curvature of spacetime from relativistic lasers [79], however, this effect remains undetectable at present. Thus, throughout this thesis, the inner product of 4-tensors is defined using the Minkowski Metric [76].

Many useful quantities can be arranged into contravariant four-vectors that undergo a Lorentz transformation for a change of frame of reference [76], specifically,

$$\mathbf{A}'^\mu = \Lambda_\nu^\mu \mathbf{A}^\mu, \quad (1.29)$$

where Λ_ν^μ is the appropriate Lorentz transformation, and primed symbols typically denote boosted frames of reference. Without loss of generality, the coordinate system can be defined such that the boosted frame travels along the **x**-axis with respect to the initial frame. Thus, the Lorentz transformation is defined as

$$\Lambda_\mu^\nu = \begin{pmatrix} \gamma & -\beta\gamma & 0 & 0 \\ -\beta\gamma & \gamma & 0 & 0 \\ 0 & 0 & 1 & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}. \quad (1.30)$$

Generally, a *beta factor* is a normalised speed or velocity of an object,

$$\beta = \frac{v}{c}, \quad (1.31)$$

here it refers to the frame velocity and its associated Lorentz or *gamma factor* is

$$\gamma = \frac{1}{\sqrt{1 - \beta^2}}. \quad (1.32)$$

Four-vectors relevant to this thesis are listed in Table 1.1. Transformations of electromagnetic fields under reference frame boosts are given in Appendix A.1. Maxwell's equations are Lorentz covariant.

Focusing now on the 4-potential \mathbf{A}^μ and choosing the Lorenz gauge,

$$\partial_\mu \mathbf{A}^\mu = \nabla \cdot \mathbf{A} + \frac{1}{c^2} \partial_t \phi = 0, \quad (1.33)$$

Symbol	Name	Components	Invariant
\mathbf{X}^μ	4-displacement	(ct, \mathbf{x})	$-c^2\tau^2$
\mathbf{A}^μ	4-potential	$(\phi/c, \mathbf{A})$	
\mathbf{J}^μ	4-current density	$(c\rho, \mathbf{J})$	$-c^2\rho_0^2$
\mathbf{K}^μ	4-wave vector	$(\omega/c, \mathbf{K})$	
\mathbf{P}^μ	4-momentum	$(U/c, \mathbf{p})$	$-m^2c^2$

Table 1.1: Four-vectors of relevance to this thesis. New parameters are the proper time, τ , the proper charge density, ρ_0 , energy, $U = \gamma mc^2$, three-momentum, $p = \gamma m\mathbf{v}$.

then Maxwell's equations can be written

$$\partial_\nu \partial^\nu \mathbf{A}^\mu = -\frac{1}{c^2\epsilon_0} \mathbf{J}^\mu. \quad (1.34)$$

Equation 1.34 can be solved to yield

$$\mathbf{A}(\mathbf{x}, t) = \frac{\mu_0}{4\pi} \int \frac{\mathbf{J}(\mathbf{x}', t_a)}{|\mathbf{x} - \mathbf{x}'|} d^3\mathbf{x}', \quad (1.35)$$

where $t_a = t - |\mathbf{x} - \mathbf{x}'|/c$ is the advanced time. Hence, electromagnetic fields radiated from charged particles in motion can be calculated².

1.6.1 Ultra-relativistic similarity theory

Consider a relativistically intense laser pulse normally incident on a collisionless plasma as in Figure 1.2. Again neglect ion motion. The electron distribution is fully described by the Vlasov equation (Equation 1.5) with the self-consistent electric and magnetic fields satisfying Maxwell's equations (Equations 1.1). Suppose the incident laser pulse has an initial vector potential

$$\mathbf{A}(t = 0) = \mathbf{a}((y^2 + z^2)/R^2, x/c\tau) \cos k_L x. \quad (1.36)$$

This envelope form for the potential, $\mathbf{a}((y^2 + z^2)/R^2, x/c\tau)$, is sensible provided $k_L R \gg 1$ and $\omega_L \tau \gg 1$, where R is the focal spot radius and τ the pulse duration. For fixed laser envelope, the laser-plasma dynamics depend on just four dimensionless

²Or even, as is performed in the full technical derivation relevant for the work of Chapter 3, electromagnetic fields can be described in terms of the future charge motions they will incite [80].

variables: the normalised focal spot size, $k_L R$, the normalised pulse duration, $\omega_L \tau$, the normalised laser vector potential amplitude

$$a = \max \left| \frac{e\mathbf{A}}{m_e c^2} \right|, \quad (1.37)$$

in terms of the peak laser electric field amplitude \mathbf{E}_L ,

$$a_0 = \frac{e|\mathbf{E}_L|}{m_e c \omega_L}, \quad (1.38)$$

and the normalised plasma density

$$\bar{n}_e = \frac{n_e}{n_c}. \quad (1.39)$$

By normalising the system of equations and combining these last two expressions into the *relativistic similarity parameter*,

$$S = \frac{\bar{n}_e}{a_0}, \quad (1.40)$$

it is possible to show that in the ultra-relativistic limit ($a_0 \gg 1$), the dynamics of the system are similar for constant S [81] with plasma electrons following similar trajectories where

$$\mathbf{p} \sim a_0. \quad (1.41)$$

There is also a more physical meaning to the S parameter. Consider again Section 1.5 on the propagation of linear electromagnetic waves through a plasma but now for the case of an ultra-relativistic laser pulse. For an electron rotating in an electromagnetic field,

$$\mathbf{F}_\perp = \gamma m_e \mathbf{a}_\perp, \quad (1.42)$$

where \mathbf{a}_\perp is the acceleration perpendicular to the motion and thus the response of the electrons is reduced by a factor of γ . While some find the *relativistic mass* correction to be somewhat unhelpful nomenclature for the phenomenon [76], it has nevertheless become commonplace within the literature of relativistic plasma physics [70]. Turning the handle, one finds that the relativistic plasma frequency is

$$\omega_p^{\text{rel}} = \sqrt{\frac{e^2 n_e}{\gamma m_e \epsilon_0}}. \quad (1.43)$$

Using equation 1.41, and taking $v \approx c$, then $\gamma \approx a_0$ and the normalised relativistic cutoff density is simply S . Thus, the ultra-relativistic similarity parameter is simply a measure of the overdensity of a plasma once relativistic corrections have been applied, *i.e.* for $S > 1$, a laser pulse will be reflected, however for $\bar{n}_e > 1$ and $S < 1$, one enters the regime of relativistically self-induced transparency [82]. It is now possible to define the parameter space of interest in this thesis: relativistic laser-plasma surface interactions occur for $a_0 \gg 1$ and $S > 1$.

1.6.2 Relativistic lasers and plasmas

The descriptor *relativistic* is applied liberally in this thesis. When applied to electromagnetic fields or laser pulses it refers to

$$a_0 \geq 1. \quad (1.44)$$

When applied to particles, their Lorentz factors are

$$\gamma = \frac{1}{\sqrt{1 - \beta^2}} \geq 2, \quad (1.45)$$

corresponding to a speed, $u \geq 0.87c$. *Ultra-relativistic* implies these quantities are much larger than the conditions provided. A relativistic laser pulse will accelerate electrons to relativistic velocities in a fraction of a laser pulse cycle. Consider an electron in the presence of a uniform electric field of magnitude $a_0 = 100$, an intensity accessible by current state-of-the-art laser facilities. The work done on that particle by the field is

$$T = (\gamma - 1)m_e c^2 = \int \mathbf{E} \cdot d\mathbf{x}, \quad (1.46)$$

The field will accelerate an electron to relativistic velocities in a distance less than 1 % of a corresponding laser pulse wavelength. It is reasonable therefore to assume the interaction is instantaneously relativistic.

Upon entrance to the relativistic regime, the motion of an electron fundamentally changes. Consider Equation 1.2. For non-relativistic laser pulses, the magnetic field component can be neglected and the electron simply oscillates along the electric

field vector direction. Once electron velocities approach c , this approximation is no longer valid. Electrons are rotated in the magnetic field and accelerated along the laser propagation direction. In a plasma, this enables inwards compression of the surface, leading to laser-induced hole boring [83].

1.6.3 Conservation of generalised transverse momentum

Consider a holonomic system of N relativistic particles under the influence of electromagnetic forces. A particle j with charge e_j and mass m_j experiences a scalar potential,

$$V_j = e_j(\phi - \mathbf{A} \cdot \mathbf{v}_j) \quad (1.47)$$

and hence the system is described by the Lagrangian [84]

$$\mathcal{L} = \sum_{j=1}^N \left(-m_j c^2 \sqrt{1 - \beta_j^2} - e_j(\phi - \mathbf{A} \cdot \mathbf{v}_j) \right), \quad (1.48)$$

The generalised momentum corresponding to coordinate x_j is

$$p_{j,x} = \frac{\partial \mathcal{L}}{\partial \dot{x}_j} = \frac{m_j \dot{x}_j}{\gamma_j} + e_j A_x, \quad (1.49)$$

describing both the linear mechanical momentum and the momentum of the electromagnetic field. Via Noether's theorem, if L is independent of x_j , *i.e.* spatially homogeneous along x for particle j , then

$$\dot{p}_{j,x} = 0 \quad (1.50)$$

since

$$\frac{d}{dt} \left(\frac{\partial \mathcal{L}}{\partial \dot{x}_j} \right) = \frac{\partial L}{\partial x_j}. \quad (1.51)$$

Taking the Lorenz gauge, consider a linearly polarised Gaussian laser pulse, with an axis of polarisation along x incident on a solid target at rest. Then A_x is approximately constant along x near the beam centre³. Integrating Equation 1.50, fully constraining the gauge by setting the potential to zero initially, and

³Constant relative to the scale of typical electron trajectories in such an interaction.

noting that there is no linear momentum at the target, the generalised transverse momentum conservation equation for an electron in the laser field is

$$p_T = eA, \quad (1.52)$$

where p_T is the electron momentum along the polarisation axis of the laser pulse and A is the laser pulse 3-vector potential amplitude. As a sanity check, this expression complies with the ultra-relativistic similarity result of Equation 1.41.

Note that this is only valid provided the electron does not radiate along the direction of polarisation as discussed by Sokolov *et al* [85]. The implications of *Radiation Reaction* are discussed in the following section.

1.7 QED effects

Next-generation laser facilities will enable the testing of decades-old theoretical predictions of Strong-Field Quantum Electro-Dynamics (SF-QED). Already Fedeli *et al* have shown in simulations that current PW-class laser facilities can access this regime using an all-optical set-up based on laser-solid surface interactions [86]. The Schwinger Limit $E_S = 1.32 \times 10^{18} \text{ V m}^{-1}$ is the field intensity at which the vacuum non-linearity can produce real particles. If by some means an electron can be directed towards a plane electromagnetic wave, by consideration of the Lorentz transformations of electromagnetic fields (equations A.1), it is possible that provided the electron is sufficiently relativistic, in its own reference frame it will ‘see’ electromagnetic fields intense enough to access such vacuum non-linearities. The first two frontiers of SF-QED that will be accessed are Radiation Reaction and multi-photon Breit-Wheeler electron pair production. Brief introductions to these phenomena are now presented.

1.7.1 High-energy photon emission and radiation reaction

When a charged particle undergoes an acceleration, it emits electromagnetic radiation. If the electromagnetic field is sufficiently strong, *i.e.* approaching the Schwinger Limit in the rest frame of the particle, then a non-negligible fraction

of the particle momentum can be transferred to the emitted high energy photon, substantially impacting the dynamics of the accelerated particle. This back reaction is known as Radiation Reaction (RR).

Smilei (detailed in the following section) implements the process of high-energy photon emission as Inverse Compton Scattering on the basis of several assumptions [87], namely,

1. Radiating particles are ultra-relativistic and therefore radiation is emitted in the direction of travel of the particle.
2. The field varies slowly over the timescale of photon emission, this is the *locally-constant field approximation* and requires ultra-relativistic field strengths;
3. but they are small with respect to the Schwinger Limit, specifically requiring the invariants $\sqrt{c^2\mathbf{B}^2 + \mathbf{E}^2}$ and $\sqrt{c\mathbf{B} \cdot \mathbf{E}} < E_S$.
4. Real particles radiate independently of their neighbours, this requires the emitted wavelength to be shorter than the typical inter-particle spacing.

Provided such conditions hold, the rate of photon emission depends on two invariants [88], the electron quantum parameter

$$\chi = \frac{\gamma}{E_S} \sqrt{(\mathbf{E} + \mathbf{v} \times \mathbf{B})^2 - (\mathbf{v} \cdot \mathbf{E})^2/c^2}, \quad (1.53)$$

where \mathbf{v} is the electron velocity and the emitted photon quantum parameter

$$\chi_\gamma = \frac{\gamma_\gamma}{E_S} \sqrt{(\mathbf{E}_\perp + \mathbf{c} \times \mathbf{B})^2 - (\mathbf{c} \cdot \mathbf{E})^2/c^2}, \quad (1.54)$$

where γ_γ is the normalised photon energy $= \hbar\omega_\gamma/m_e c^2$. The exact relationship is complex and in the fully quantum domain ($\chi > 1$), is it not practical to solve the integrations required for all particles. Instead, values are extracted from precalculated tables and supplied to a Monte Carlo algorithm.

1.7.2 Multi-photon Breit-Wheeler pair production

Multi-photon Breit-Wheeler pair production, also known as non-linear Breit-Wheeler is the decay of a high energy photon, typically produced via RR, into an electron-positron pair in the presence of a strong electromagnetic field, explicitly,

$$\gamma + n\omega \rightarrow e^- + e^+. \quad (1.55)$$

The strength of the effect is dependent on the Lorentz invariant photon quantum parameter, Equation 1.54. In a constant electric field, the rate of pair production increases rapidly up to $\chi_\gamma \approx 10$ at which point it saturates and slowly reduces.

1.8 Simulating the interaction

Modelling laser-plasma interactions is a notoriously challenging endeavour. Due to the complexity of the many-bodied systems involved (a fully ionised centimetre cubed block of plastic contains on the order of 10^{23} particles) and the stochasticity of particle motion, it is frequently impossible to construct models *ab initio*. Instead, hydrodynamic simulation codes such as HYADES [89] and FLASH [90] and Particle-In-Cell simulation codes such as Smilei [77], Osiris [91] and EPOCH [92] are used to construct phenomenological models and to direct experimentation.

1.8.1 Supercomputing resources

Modern High Performance Computing (HPC) systems are poised to enter the exascale regime ($> 10^{18}$ Floating Point Operations Per Second). With limited improvements in microprocessor technologies, such power is achieved through massive parallelisation across processing units. Able to study the dynamics of billions of macroparticles, PIC codes test the limits of modern supercomputing architectures. ARCHER2, the UK's national supercomputer, came online in November 2021, with it delivering over ten times the resources of its predecessor (ARCHER) [93]. An HPE Cray EX supercomputing system with a peak performance estimated at 28 Pflops s^{-1} across 5860 nodes each with dual AMD EPYCTM 7742 64-core processor for a total of 750,080 cores, ARCHER2 was able to supply

the resources required to run the costly PIC simulations for this research. The substantially cheaper HYADES simulations were performed on the Rutherford Appleton Laboratory's SCARF HPC cluster [94].

1.8.2 Particle-In-Cell codes

Discretisation of the Vlasov-Maxwell equations

Finding numerical solutions to the Vlasov-Maxwell equations is no straightforward. While codes exist that are capable, such as Valis [95], the requirement of high resolution in both position and momentum is exceedingly costly and the use of such codes is limited with respect to their size, duration and number of spatial dimensions. A more tractable approach is to discretise the distribution function into N_s *quasi-particles*⁴. These are often referred to as *macro-particles* in practice and typically represent a large number of real particles, such that

$$f_s(t, \mathbf{x}, \mathbf{p}) = \sum_{p=1}^{N_s} w_p S(\mathbf{x} - \mathbf{x}_p(t)) \delta(\mathbf{p} - \mathbf{p}_p(t)), \quad (1.56)$$

where w_p is the quasi-particle's weight, \mathbf{x}_p and \mathbf{p}_p are its position and momentum respectively, δ is the Dirac-delta distribution and $S(\mathbf{x})$ the shape-function chosen to represent the spatial extent of the quasi-particle. The Vlasov equation is then integrated along the continuous trajectories of quasi-particles while Maxwell's equations are solved on a discrete spatial grid of *cells*. Such a code is aptly named a *Particle-In-Cell* (PIC) code. A schematic of the standard PIC code algorithm is presented in Figure 1.3. After particle and field initialisation, fields are interpolated at particle positions. The well-established momentum-conserving *Boris pusher* algorithm computes the new macro-particle velocities and positions [97]. Particles are advanced in time using a *leap-frog* scheme, where positions are defined at integer, n , time steps and momenta at half-integer, $n + 1/2$. The charge conserving Esirkepov algorithm [98] projects the new charge and current densities onto the grid to then solve Maxwell's equations using the Finite-Difference-Time-Domain approach [99]. To ensure space and time centring of the electromagnetic field derivatives in

⁴Originally introduced by Langdon and Birdsall as *clouds* [96].

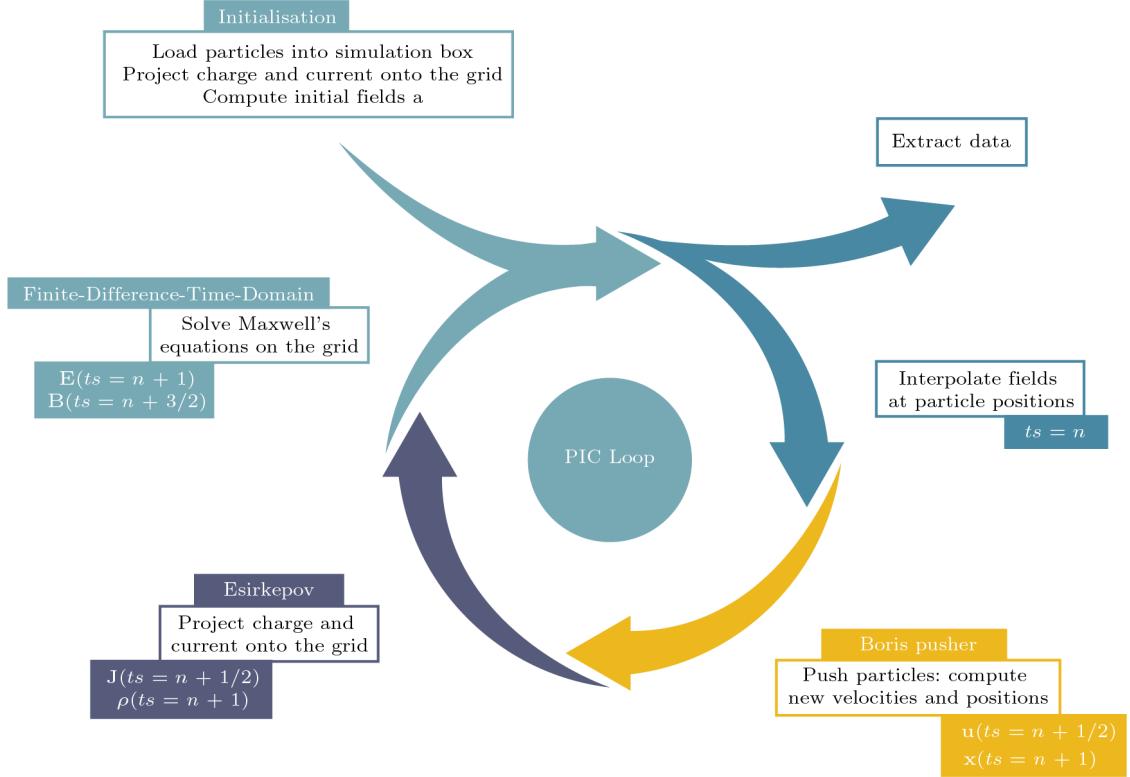


Figure 1.3: A schematic of the PIC code loop and the algorithms performed from time step, ts , from n to $n + 1$.

Maxwell's equations, electric and magnetic fields are discretised on the staggered *Yee grid* as represented in Figure 1.4. with electric fields defined at integer time steps and magnetic fields at half-integer time steps. PIC codes can function in one-, two- and three-dimension spatially but always with three-dimensions in velocity.

Smilei

Smilei (for Simulating Matter Irradiated by Light at Extreme Intensities) is a modern, collaborative, massively parallel, fully relativistic and open source plasma physics PIC code and the major workhorse for this thesis⁵. Produced at École Polytechnique, Paris [77], its recent development was motivated by the recent advancements of multi-petawatt facilities both globally and locally with the completion in 2020 of the 10 PW Apollon laser facility and by the availability of supercomputing power which has ‘skyrocketed’ in recent years [77]. The 3D PIC simulations examined in

⁵At points benchmarks against the EPOCH and Osiris PIC codes were performed.

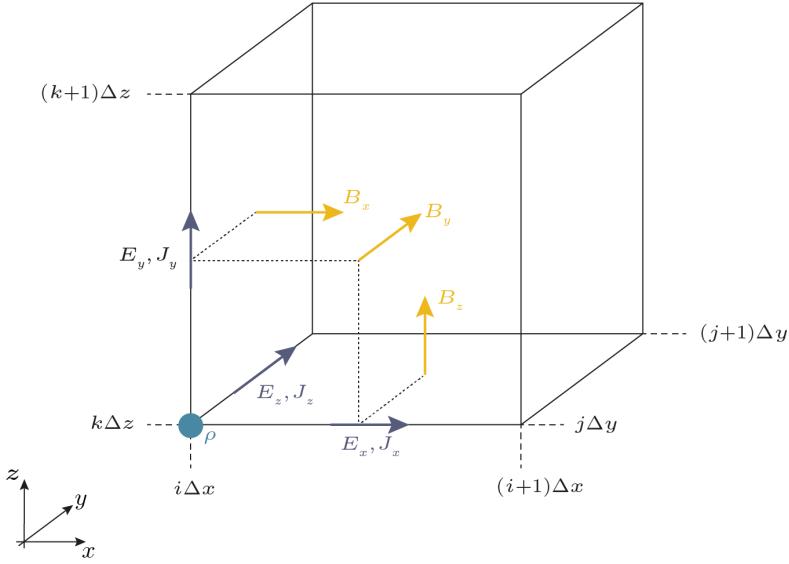


Figure 1.4: A representation of the staggered 3D Yee grid. For the cell at $(i\Delta x, j\Delta y, k\Delta z)$ for spatial centring of the curl operations, including the locations where all system properties are defined.

this thesis demanded the parallelisation of almost 20 % of the computing resources available on the ARCHER2 supercomputer.

Reference units

Given the broad range of magnitudes linked to multi-petawatt and femtosecond laser pulses, solid density plasmas, micrometre wavelengths, and attosecond electron bunches, it becomes significantly more convenient to transform them into dimensionless and normalised values. Smilei operates in such units. This normalisation is not chosen *a priori*, instead results can be scaled by an arbitrary reference angular frequency. This is extremely useful when working with boosted frames of reference. As this thesis focuses on the interaction of a laser pulse with plasma, the laser pulse angular frequency, ω_L is set as the frequency of reference. A list of the most common normalisations is provided in Table 1.2.

Simulation parameters

Silver-Müller boundary conditions, chosen for the simulation box edges [100], absorb and inject electromagnetic waves and particles. There can be non-physical reflection of electromagnetic waves at such boundaries leading to some error.

Units of	SI units	Normalisation
velocity	m s^{-1}	c
charge	C	e
mass	kg	m_e
momentum	kg m s^{-1}	$m_e c$
energy/temperature	J	$m_e c^2$
time	s	ω_L^{-1}
length	m	c/ω_L
number density	m^{-3}	n_c
electric field	V m^{-1}	$m_e c \omega_L / e$

Table 1.2: Smilei normalisations for common quantities. The laser angular frequency ω_L is set at the reference angular frequency.

The quasi-particle shape function $S(\mathbf{x})$ determines the projection of particle charge onto the grid. It is symmetric in all dimensions with respect to \mathbf{x} and extends over n cells of width Δx in each direction where n is the interpolation order. It can be written as a product across D dimensions,

$$S(\mathbf{x}) = \prod_{\mu=1}^D s^{(n)}(x^\mu). \quad (1.57)$$

Smilei implements orders 2, 3 and 4, the explicit shape functions are

$$s^2(n) = \begin{cases} \frac{1}{\Delta x} \left(1 - \left|\frac{x}{\Delta x}\right|\right) & \text{if } |x| \leq \Delta x, \\ 0 & \text{otherwise,} \end{cases} \quad (1.58a)$$

$$s^3(n) = \begin{cases} \frac{3}{4\Delta x} \left(1 - \frac{4}{3} \left(\frac{x}{\Delta x}\right)^2\right) & \text{if } |x| \leq \frac{1}{2}\Delta x, \\ \frac{9}{8\Delta x} \left(1 - \frac{2}{3} \left|\frac{x}{\Delta x}\right|\right)^2 & \text{if } \frac{1}{2}\Delta x < |x| \leq \frac{3}{2}\Delta x, \\ 0 & \text{otherwise,} \end{cases} \quad (1.58b)$$

$$s^4(n) = \begin{cases} \frac{2}{3\Delta x} \left(1 - \frac{3}{2} \left(\frac{x}{\Delta x}\right)^2 + \frac{3}{4} \left|\frac{x}{\Delta x}\right|^3\right) & \text{if } |x| \leq \Delta x, \\ \frac{4}{3\Delta x} \left(1 - \frac{1}{2} \left|\frac{x}{\Delta x}\right|^3\right) & \text{if } \Delta x < |x| \leq 2\Delta x, \\ 0 & \text{otherwise.} \end{cases} \quad (1.58c)$$

While the correct implementation of collisions in PIC codes remains an open problem [101], Smilei has implemented relativistic binary collisions between macroparticles using a Monte-Carlo-based scheme [102]. The aforementioned QED processes

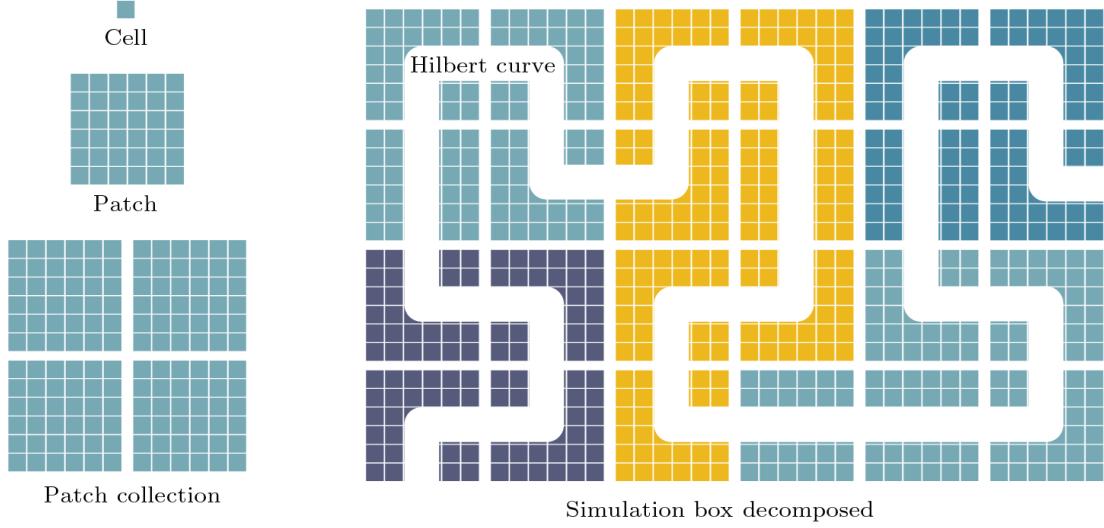


Figure 1.5: Smilei simulation box decomposition into cells, patches and MPI patch collections. Cells are grouped into patches, patches are grouped into MPI patch collections. These collections are assigned patches contiguously along the Hilbert curve.

of Inverse Compton scattering and non-linear Breit-Wheeler pair production are included using the built-in Smilei packages [77]. These processes can lead to cascades of many particles being added to the simulations. Macroparticle merging can increase simulation efficiency and reduce the memory footprint. Smilei implements such a scheme, inspired by that designed by Vranic *et al* [103], that is computationally efficient and conserves energy, momentum and charge within a cell. While Smilei contains modules to handle ionisation, these are deemed unnecessary for the laser intensities considered in this thesis, as highlighted by Figure 1.1.

Parallelisation in practice

In the following discussion, where differences in language occur, objects are given their ARCHER2 (Smilei) names. The Smilei simulation box consists of a grid of cells as in Figure 1.5. The box is decomposed into *patches* consisting of many cells. Patches are arranged into *MPI patch collections* assigned contiguously along a Hilbert curve.

Archer consists of many *CPUs (cores)* that can each perform computational tasks. CPUs are grouped into *nodes*. Memory is shared within a node such that all CPUs (cores) in a node can operate on the same data. When optimised, ARCHER2's

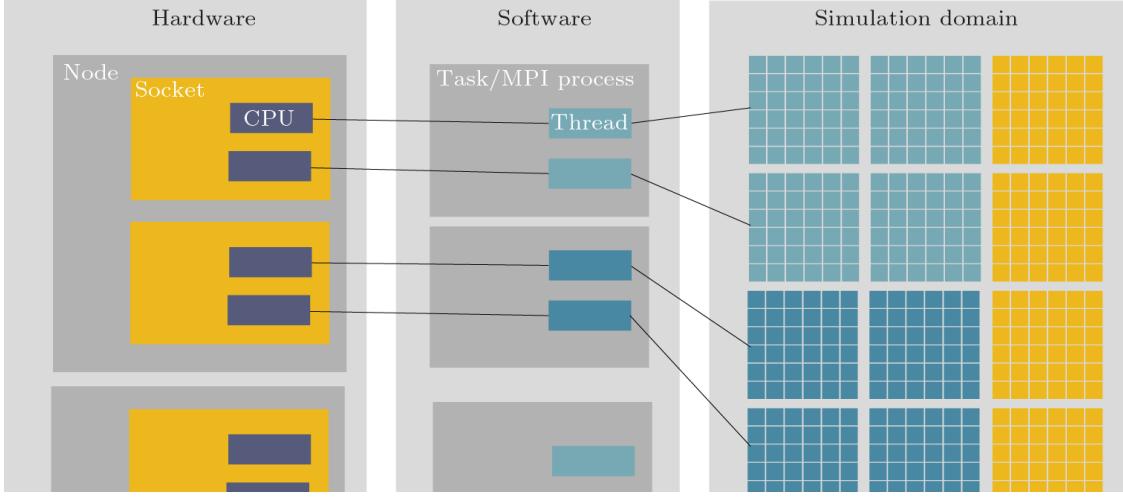


Figure 1.6: Representation of the interaction of the ARCHER2 hardware and software components when running Smilei. Each CPU is assigned a thread, each task is carried out by one socket. A task tackles one MPI patch collection with threads working through patches.

memory in each node is split into 8 *sockets*. These 8 sockets each perform a *task* (*MPI process*). Each task (*MPI process*) has 16 CPUs (cores) assigned that each perform a *thread*. A thread is a sequence of instructions from the program.

Each task (*MPI process*) handles one *MPI patch collection*. Threads work through patches. Figure 1.6 represents this division of labour. Threads do not need to wait for other threads in their process to tackle new patches in their MPI patch collection. This is a form of local *dynamic load balancing*. If an MPI patch collection is overloaded, a patch is offloaded to a contiguous MPI patch collection.

There are several considerations for simulation optimisation. Tasks (*MPI processes*) should always be assigned more patches than threads. To apply the Hilbert curve, the number of patches in each dimension must be a power of 2. The fewer cells in a patch, the more efficient the load balancing, however, the cost of synchronisation between patches increases, although generally the gain in efficiency from load balancing by increasing patch number will win out at later times with increasing efficiency for increased frequency of load balancing [77]. Note that smaller patches are preferable when there are small regions with large numbers of particles, as in laser-solid surface interactions, however, the minimum patch size is dependent on the shape function of the macro-particles.

Sources of error inherent to PIC codes

Despite their relatively intuitive interpretation, PIC codes are famously finicky and prone to errors, most notably that of numerical self heating⁶.

To ensure stability, or at least to minimise instability, there are several conditions which must be met. Naturally, the time step, Δt , and cell size ($\Delta x \Delta y \Delta z$) must adequately capture all interesting features of a given simulation. Typically such features are plasma wave oscillations,

$$\Delta t \omega_p \ll 1, \quad (1.59)$$

and laser pulse electromagnetic field oscillations or higher order harmonics of the laser pulse if that is of interest, for the n th harmonic

$$\Delta t \omega_L n \ll 1. \quad (1.60)$$

Note this also ensures that macroparticles are smaller than the wavelengths of the system, a requirement to ensure they mimic real particles [104]. Relativistic PIC codes must satisfy the much-acclaimed [105] Courant-Friedrichs-Lowy condition,

$$\frac{1}{c\Delta t} > \sqrt{\frac{1}{(\Delta x)^2} + \frac{1}{(\Delta y)^2} + \frac{1}{(\Delta z)^2}}, \quad (1.61)$$

thus preventing light and relativistic particles from crossing a cell in one timestep and generating numerical Cerenkov radiation [106]. As with real plasmas and real particles, to avoid numerical charge fluctuation and ensure the collective behaviour of macroparticles,

$$\frac{4}{3}\pi\lambda_D^3 n_{\text{macro}} = N_{D,\text{macro}} \gg 1, \quad (1.62)$$

where n_{macro} is the macroparticle density [106]. To avoid numerical heating, the cell size must resolve the Debye length,

$$\frac{\lambda_D}{\Delta x} \geq 1, \quad (1.63)$$

⁶Standard PIC code algorithms are charge and momentum conserving but not energy conserving [77].

failure to do so may cause plasma self-heating until the Debye length matches the cell size [106]. Interestingly, Brackbill *et al* [107] observed in their simulations that setting $\lambda_D/\Delta x = 1$ was most effective at reducing spurious heating. Arber *et al* [108] performed extensive simulations exploring this instability. If the Debye length is not resolved, after an initial period of rapid self-heating, the temperature increases linearly and can be modelled as

$$\frac{dT_{\text{eV}}}{dt_{\text{ps}}} = \alpha_H \frac{n_{23}^{3/2} \Delta x_{\text{nm}}^2}{N_{\text{ppc}}}, \quad (1.64)$$

where T_{eV} is the temperature in electron volts, t_{ps} is the time in picoseconds, n is the plasma electron number density in units of 10^{23} cm^{-3} , Δx_{nm} is the cell size in nanometres and N_{ppc} is the number of macroparticles per cell with α_H , the heating coefficient, determined from simulation. For a top-hat macroparticle shape function, they observed $\alpha_H = 3000$ with an order of magnitude reduction for every increase in order of the shape function. Further improvements were also noted from the use of current smoothing techniques. Note that the heating curves are roughly self-similar at all points and thus while Equation 1.64 was established in the linear regime only, its scalings remain useful for simulation comparison at all times.

The final instability that shall be discussed is the *finite grid instability*. This is the aliasing error associated with particle properties being deposited at grid points. Is most catastrophic for cold drifting plasmas and depends on the *beam Debye length*,

$$B = \frac{u}{\omega_p \Delta x}, \quad (1.65)$$

where u is the beam speed. While their theory predicts stability for $B > 0.25$, Brackbill *et al* [107] observed instability growth for all beam temperatures in their simulations, although they found the percentage error is a small fraction for $B > 10$.

1.8.3 Hydrodynamic codes

A hydrodynamic code approximates the plasma distribution as a fluid. This is achieved by taking the velocity moments of the distribution function, f_s for fluid

species s . Similar to the charge and current density calculations of Section 1.2, the first three return the number density of particles

$$n_s = \int f_s d\mathbf{v}, \quad (1.66)$$

the fluid velocity, \mathbf{u}_s and momentum,

$$\mathbf{p}_s = m_s n_s \mathbf{u}_s = \int f_s \mathbf{v} d\mathbf{v}, \quad (1.67)$$

and the pressure tensor

$$P_s = m_s \int f_s (\mathbf{v} - \mathbf{u}_s)(\mathbf{v} - \mathbf{u}_s) d\mathbf{v}. \quad (1.68)$$

The fluid equations that govern the evolution of these quantities can be extracted from the Vlasov equation, Equation 1.5 by taking the appropriate moments ⁷. Likewise producing the continuity equation

$$\frac{\partial n_s}{\partial t} + \nabla \cdot (n_s \mathbf{u}_s) = 0, \quad (1.69)$$

for conservation of particle number and the equation of motion

$$m_s n_s \left(\frac{\partial \mathbf{u}_s}{\partial t} + (\mathbf{u}_s \cdot \nabla) \mathbf{u}_s \right) = q_s n_s (\mathbf{E} + \mathbf{u}_s \times \mathbf{B}) - \nabla \cdot P_s \quad (1.70)$$

for conservation of momentum. This equation is analogous to the Navier-Stokes equation for standard fluids except for the addition of the electromagnetic fields. The viscosity term is collected within the pressure tensor [73].

Here lies a problem. The presence of the pressure tensor in Equation 1.70 prevents the closure of the system of equations. Taking the second moment of the Vlasov equation to obtain the energy transport equation would then contain the third moment of the distribution function, presenting the same issue. Instead, a *closure relation* is introduced, enabling the construction of a self-contained theory using a finite number of moments.

HYADES is a 1D three fluid (electrons, ions and radiation) radiative hydrodynamic code [89] and is the code of choice in this thesis. The equations of mass

⁷Of course, these equations can also be derived by first principles by considering the dynamics of a fluid element [73]

Parameter	ORION	GEMINI	
Beamlines	SP1	SP2	N & S
Power (PW)	0.5	1	0.5
Energy (J)	200	500	12
Wavelength (nm)	527	1053	800
Parabola, $f/\#$	$f/3$	$f/3$	$f/2$
Focal spot FWHM (μm)	< 20	< 10	2
Duration (fs)	500	500	40
Shot rate	5/day	5/day	3/min
Peak a_0 (approx)	10	30	24

Table 1.3: The ORION and GEMINI petawatt class short pulse beamlines for comparison. The GEMINI North (N) and South (S) beamlines are equivalent.

and energy transport are solved in a Lagrangian coordinate system, *i.e.*, unlike strictly Eulerian PIC codes, the mesh defining regions of the simulation moves with the plasma it describes. To complete the system up to the second moment, the *equation of state*, describing pressure in terms of the temperature and density of the system, and the heat transfer equation are added.

The reduction of dimensionality achieved through this approximation dramatically reduces the computational cost of the simulation. However, much of this thesis relies on the precise velocity distribution of particles. Naturally, such a code cannot model these phenomena and the primary tool is the PIC code.

1.9 Generating the interaction

Two experiments are discussed in this thesis, one on ORION, and one planned on GEMINI. Their typical beam parameters are presented in Table 1.3. The GEMINI-PW laser facility of the Central Laser Facility (CLF) at Harwell Campus, Oxfordshire is a petawatt class facility consisting of two 30 fs beams, North and South, each delivering a maximum focused intensity of $2 \times 10^{21} \text{ W cm}^{-2}$ at a repetition rate of 0.05 Hz. The commissioning of such high-frequency facilities

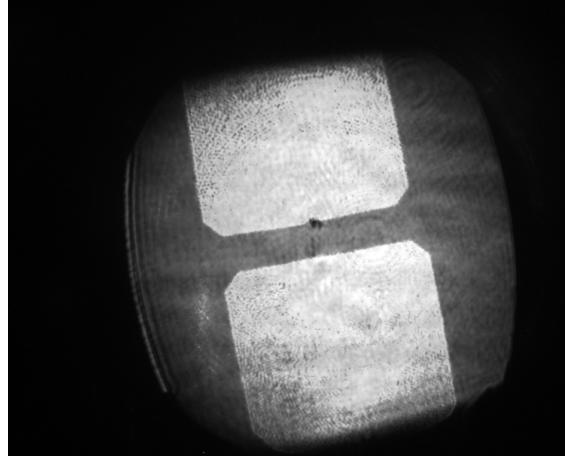


Figure 1.7: Image of the ORION SP1 double beamlet structure in the near-field. The two beamlets are then superimposed on the target.

has ushered in a paradigm shift in high-power laser physics experimentation with the entry of statistically significant results.

The ORION laser facility of AWE, Aldermaston is the UK's most powerful laser system with two short pulse beamlines, SP1 and SP2 and ten 'long pulse' beams. The ORION SP1 beamline is produced by passing the SP2 beamline through two frequency doubling crystals creating two equivalent beamlets that are then superimposed at the target. The double beamlet structure at the near-field is imaged in Figure 1.7.

Laser contrast is a major concern for the interaction discussed in this thesis. There is always some spurious signal arriving before the main pulse of interest. In high-power laser systems, this *prepulse* can be itself of ionising intensities, dramatically altering the initial conditions of the system. Generally, the prepulse preheats the plasma leading to expansion. This is catastrophic if one's aim is to interact a laser with a solid density plasma.

The ORION SP2 beamline temporal intensity envelope can be modelled as

$$I_{\text{SP2}} \sim (I_{0 \text{ p}} \operatorname{sech}(t/t_p)^2 + I_{0 \text{ pp}} e^{-(t/t_{\text{pp}})^2} + I_{0 \text{ ps}} e^{-\operatorname{abs}(t)/t_{\text{ps}}} + I_{0 \text{ ns}} e^{-(t/t_{\text{ns}})^8}), \quad (1.71)$$

where the constants are detailed in Table 1.4 [109]. The picosecond pump residual arises from parametric fluorescence in the ps OPA, the picosecond pedestal from scatter in the stretchers and noise on the OPA pump laser and the nanosecond

Pedestal, i	$I_{0,i}$	t_i
Main pulse, p	1	0.2 ps
Picosecond pump residual, pp	10^{-3}	3 ps
Picosecond pedestal, ps	5×10^{-5}	8 ps
Nanosecond pedestal, ns	10^{-11}	3 ns

Table 1.4: The native ORION SP2 pulse and prepulse pedestal constants as defined in Equation 1.71.

pedestal from parametric fluorescence from the nanosecond OPAs. The ‘long’ duration of the prepulse precludes the use of PIC codes. It is here that the hydrodynamic codes can be more appropriate.

The SP1 temporal intensity profile is

$$I_{\text{SP1}} \sim (I_{\text{SP2}}^2 + I_{\text{SP2}} \times 10^{-8}). \quad (1.72)$$

The second term arises from limitations in the harmonic separation system. The frequency doubling mechanism is not 100 % efficient, *i.e.* some of SP2 beamline remains and must be filtered out. The main pulse of the GEMINI beamlines is more appropriately modelled as a Gaussian.

A Plasma Mirror (PM) is an effective and now standardised tool for the improvement of laser contrast. These single-use optics have initially an Anti-Reflective (AR) coating of reflectivity R_1 . As the incident laser fluence passes the damage threshold of the optic, reflecting plasma forms on the front surface. The optic is ‘switched on’ with a reflectivity R_2 . The PM therefore improves the contrast by a factor R_2/R_1 . There is a balancing act in choosing the peak intensity on the PM. Too early and the PM will switch on before the arrival of the main pulse. Too low and there is not enough plasma at the surface at the arrival of the main pulse and R_2 is lowered. A peak intensity around $1 \times 10^{16} \text{ W cm}^{-2}$ is best [110]. PMs also act as a low-pass spatial filter, improving the quality of the focal spot on target [111]. The use of PMs before the main target ensure the required conditions for a relativistic laser pulse to produce a Relativistic Plasma Mirror (RPM), the main focus of this thesis.

1.10 Summary

This chapter has introduced the field of HED physics. The basic equations that govern the system have been supplied and some simple derivations performed to describe the dynamics of particles in this regime. Details have been given of both suitable simulation codes and laser systems that enable the modelling and generation of the interaction of interest in this thesis. It is now time to present that interaction.

2

The Zero Vector Potential Absorption Mechanism

Contents

2.1	Motivations and an overview	33
2.2	Introduction	35
2.2.1	ZVP electron bunch energies	42
2.2.2	ZVP bunches oblique incidence scaling	45
2.2.3	Defining characteristics of the ZVP mechanism	47
2.3	Numerical simulations of the ZVP mechanism	48
2.3.1	The ZVP mechanism in 3D3V	48
2.4	The ZVP electron bunch	55
2.4.1	Energy absorption in the ZVP regime	66
2.4.2	Unpacking the QED effects of Figure 2.12	68
2.4.3	Errors	70
2.5	Conclusions	71

2.1 Motivations and an overview

Throughout the history of experimental science, light has always been the primary tool of investigation and discovery. Through the creation of synchrotron radiation sources and more recently XFELs, electron bunches have been employed to create increasingly specialised light sources for the study of matter of all kinds. At SLAC, the United States' forefront electron accelerator, applications hail from many

disciplines: science, medicine, industry and homeland security. At Diamond Light Source, the UK's national synchrotron science facility, studies range from novel drugs to ancient paintings. Unsurprisingly, extensive research efforts have been enlisted for the production of electron bunches of ever greater charge, energy and coherency.

Multi-petawatt laser facilities are now available across the globe for the study of laser-plasma interactions in the ultra-relativistic regime $a_0 \gg 1$. Here, there is a novel method for high-charge electron bunch creation. Via relativistic effects, a laser pulse organises the electrons at the surface of a solid-density plasma into coherent bunches that can be ejected at high speeds. Those discussed in this thesis have properties comparable to those of forefront accelerators but on ultra-short timescales. The quality, charge and duration of the attosecond electron bunches described here would enable the study of the most fundamental interactions of our universe.

Electron bunch formation from solid targets has received much interest in recent years [112–119] with some experimental evidence for attosecond electron bunches from intense laser-solid interactions [120–122]. This interest stems partially from their ability to produce higher charge bunches at lower intensities compared to gas targets [120]. This chapter proposes a new mass-limited target setup to generate electron bunches of extreme charge density. These electron bunches are fully characterised in 3D PIC simulations to compare their quality to those of existing electron bunch production methods. They are described quantitatively via an extended version of the ZVP Mechanism and the corresponding implications for laser to plasma energy absorption are also considered including the junction of ZVP with SF-QED effects that will be accessed by next-generation laser facilities. The ZVP theory has direct relevance to HHG due to the known intrinsic link between electron bunches and the reflection mechanism [123, 124].

These results have excited the community to perform experiments to realise these electron bunches for the creation of ultra-bright X-ray pulses: after the recent successful campaign at the ORION laser facility, detailed in Section 3, details are given in Chapter 4 for the upcoming experiment to observe the electron bunches directly at the GEMINI PW laser at the Central Laser Facility, UK.

This Chapter is organised as follows. Section 2.2 outlines the ZVP mechanism including quantitative calculations of system properties. In Section 2.3.1 the first 3D PIC simulations to observe the ZVP mechanism are presented. Further PIC simulations are presented in Section 2.4 focusing now on the hot electron bunches produced via the mechanism, their properties, energy scalings and implications for SF-QED. Finally, concluding remarks are given in Section 2.5.

2.2 Introduction

Here is presented one model for the interaction between a relativistic laser pulse and a solid density plasma: the ZVP mechanism as proposed by *Baeva et al* [125] and later developed by *Savin et al* [124, 126]. Alongside the theories of the Relativistic Electron Spring [127] and Coherent Synchrotron Emission [128], ZVP is a post-ponderomotive model of attosecond absorption and reflection. The model considers a quasi-static surface equilibrium state analogously to ion acceleration in the hole boring [129] and light sail regimes [130].

An explanation for the absorption of laser energy into dense plasmas was first suggested by Wilks and Kruer [83], a ponderomotive mechanism where plasma electrons are heated directly by the laser pulse via the $\mathbf{J} \times \mathbf{B}$ force. This thesis is interested in the so-called ‘post-ponderomotive’ regime where the frequency of relativistic plasma oscillations ($\omega_p \sim \sqrt{S}$) are greater than the $\mathbf{J} \times \mathbf{B}$ induced plasma electron oscillations at $2\omega_L$. The plasma electrons’ response is then sufficiently rapid to compensate for the ponderomotive pressure of the laser pulse with the formation of electrostatic fields between electrons and ions and so respond adiabatically to the applied $\mathbf{J} \times \mathbf{B}$ force. Hence, plasma electrons cannot be heated directly by the laser pulse. Interestingly, this condition suggests a criterion for the ZVP regime, $S > 4$, slightly more constraining than $S > 1$ as is typically stated for the ZVP regime [126]. The bulk plasma must have $S > 1$ to prevent relativistic transparency and so ensure a laser-surface interaction. Then the ponderomotive pressure of the laser will typically compress the front surface to densities such that the overdensity condition is satisfied, provided the target is of sufficient thickness. Note that neglecting the

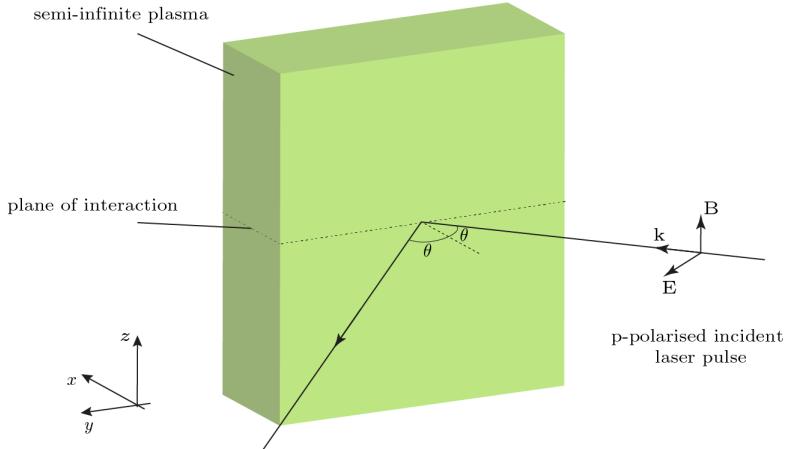


Figure 2.1: Diagram of a p-polarised laser pulse incident at angle θ specularly reflected from a solid density plasma. From consideration of the Lorentz force equation, it is clear that all forces and therefore all plasma particle dynamics are confined to a plane.

pre-adiabatic formation phase requires a sufficiently steep density gradient around the relativistic critical density surface (where $S = 1$) to shift the main interaction to a region where this condition on the overdensity is satisfied. Preplasma formation and scale length will be discussed in great detail in the later chapters. For now, assume the technology exists to sufficiently control this phenomenon.

Provided all conditions are met, the ponderomotive pressure of the laser pulse compresses electrons at the front surface of the plasma and so shifts the laser-plasma surface interaction to plasma densities well beyond the relativistic critical density, leaving in its wake a positive space charge of ions. This electron-ion charge separation leads to the formation of a *pseudocapacitor* electrostatic field. Having entered a regime of adiabaticity, the plasma skin layer is confined within a potential well consisting of the ponderomotive pressure of the laser pulse and the Coulomb potential of the pseudocapacitor field. Thus is formed a high-density and longitudinally thin electron bunch (sometimes referred to as an electron sheath in the literature [127]) at the plasma surface.

To understand this system, consider now a relativistic linearly polarised laser pulse obliquely incident, at an angle of incidence θ , on a semi-infinite plasma, existing for $x \geq 0$ as in Figure 2.1.

The Hamiltonian of a single electron confined within the potential well [84] is

$$\mathcal{H} = c\sqrt{m_e^2c^2 + |\mathbf{p}|^2} - e\Phi. \quad (2.1)$$

Here, the second term of Equation 2.1 describes the contribution to the electron's energy from the electrostatic potential, Φ , of the pseudocapacitor. The first term is the electron energy, U , extracted from the invariant of the relativistic 4-momentum of the electron, $\mathbf{P}^\mu = (U/c, \mathbf{p})$,

$$\mathbf{P}_\mu \mathbf{P}^\mu = \frac{U^2}{c^2} - |\mathbf{p}|^2 = m_e^2c^2. \quad (2.2)$$

Decomposing the electron's 3-momentum into orthogonal components: p_{prop} , along the laser propagation direction, p_{pol} , along the polarisation axis of the laser pulse and p_\perp , perpendicular to both, there are two simplifications to be made. Firstly, by canonical conservation of transverse momentum, $p_{\text{pol}} = eA$, where A is the laser vector potential amplitude. Secondly, in the case of a p-polarised laser pulse (the known optimum for ZVP electron bunch generation [124] and HHG [131]), with reference to Figure 2.1 and the Lorentz force law, the forces at play confine the electron trajectory to the $p_{\text{prop}}-p_{\text{pol}}$ plane and the essential interaction geometry is two-dimensional. This is provided one considers length scales smaller than the focal spot of the laser pulse on the target, such that variation of the ponderomotive pressure with the transverse shape of the laser pulse can be neglected.

Explicitly, the Hamiltonian can be written as

$$\mathcal{H} = c\sqrt{m_e^2c^2 + p_{\text{prop}}^2 + e^2A^2} - e\Phi. \quad (2.3)$$

Equation 2.3 describes a potential well that constrains the electrons: on one side is a longitudinal space charge $-e\Phi$ that prevents further propagation into the target while on the other, a term associated with the vector potential of the laser pulse, e^2A^2 . It is clear that should the vector potential pass through zero¹, one of the potential well walls is totally suppressed, enabling electrons in the skin layer to escape the plasma, breaking adiabaticity. The necessity of vector potential zeroes

¹Note that in writing $p_{\text{pol}} = eA$ one has determined the gauge in which the zero becomes a defined quantity

for this violent reconstruction of the plasma surface led Baeva to coin the term ‘Zero Vector Potential’ mechanism to describe this process [125]. Indeed, while elementary electromagnetism tells us a laser pulse will exponentially decay within a skin layer of a plasma without passing through zero, Baeva *et al* [125] demonstrated in PIC simulations that in this non-linear regime, zeroes do exist and do propagate through the skin layer. The explanation relies on a Doppler shift in the laser field due to the relativistic motion of the ablating plasma electrons, and a mathematical formalism of this process proceeds as follows.

As the ZVP mechanism is a relativistic phenomenon, it is absolutely essential to perform a relativistic analysis. Since all accelerated electrons travel at approximately speed c , surface electrons undergo similar trajectories. Acting collectively they oscillate in the laser pulse field. Consider first a transformation to the frame of reference where the laser pulse is normally incident to the plasma surface, this frame travels at velocity $\mathbf{v} = (c \sin \theta) \hat{\mathbf{y}}$ with electrons streaming at $-\mathbf{v}$. This is the Bourdier frame of reference, detailed in Appendix A. Using Equation 2.2, $U = \gamma m_e c^2$ and integrating Equation 1.50 in the boosted frame noting $p_T(t = 0) = \gamma_{\mathbf{v}} m_e v = m_e c \sin \theta \cos \theta$,

$$\gamma^2 = 1 + (a_0 + \sin \theta \cos \theta)^2 + \left(\frac{p_{\text{prop}}}{m_e c} \right)^2, \quad (2.4)$$

where all parameters are in the boosted frame. Using $\mathbf{p} = \gamma m_e \mathbf{v}$, the longitudinal velocity is

$$v_{\text{prop}} = \frac{\tilde{p}_{\text{prop}} c}{\sqrt{1 + (a_0 + \sin \theta \cos \theta)^2 + \tilde{p}_{\text{prop}}^2}}, \quad (2.5)$$

where $\tilde{p}_{\text{prop}} = p_{\text{prop}}/m_e c$. Thus, should the transverse vector potential pass through $-\sin \theta \cos \theta$, zero for normal incidence, the surface can propagate towards the laser pulse at very close to speed c . Transforming back to the laboratory frame, at the peak of ablation ($\mathbf{u} \approx -c \hat{\mathbf{x}}$) and using the equations for relativistic velocity addition,

$$\mathbf{u}'_{||} = \frac{\mathbf{u} - \mathbf{v}}{1 - \mathbf{u} \cdot \mathbf{v}/c^2}, \quad (2.6)$$

$$\mathbf{u}'_{\perp} = \frac{\mathbf{u}_{\perp}}{\gamma_v (1 - \mathbf{u} \cdot \mathbf{v}/c^2)}, \quad (2.7)$$

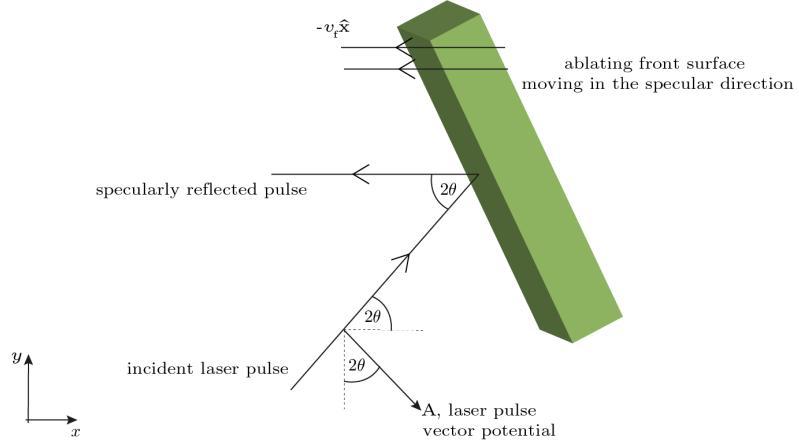


Figure 2.2: Diagram of a p -polarised laser pulse incident on an ablating overdense plasma. The laser is incident obliquely at an angle of θ and is reflected specularly. The plasma ablates specularly also. Note that this process occurs on sub-laser pulse cycle timescales and it is therefore only the electrons that are ablating in this process. The interaction geometry is confined to a 2D plane.

where $\gamma_v = 1/\sqrt{1 - |\mathbf{v}|^2/c^2}$ [76], one finds that this peak ablation at speed $\approx c$ occurs now in the specular reflection direction. Simultaneity is broken and ripples co-move along the surface with the incident laser pulse wavefronts.

Transform now to the rest frame of the ablating front. Beyond the relativistic critical density surface, the vector potential of the laser pulse decays evanescently. At the spatial centre of the laser pulse, it can be described simply by

$$\mathbf{A}'_{\text{L}}(t', r') = A'_0 \cos(\omega'_{\text{L}} t') \exp(-r'/\delta') \hat{\mathbf{r}}'_{\text{pol}} = A'_0 \hat{\mathbf{r}}'_{\text{pol}}, \quad (2.8)$$

where the primed symbols indicate that these quantities are measured in the rest frame of the expanding front. A'_0 is the vector potential amplitude and ω'_{L} is the frequency of the laser pulse, r' is the propagation distance of the laser into the plasma, δ' is the skin depth and $\hat{\mathbf{r}}'_{\text{pol}}$ a unit vector defining the polarisation direction of the laser pulse. Un-primed coordinates will indicate the lab frame measurements.

While previous demonstrations of the existence of vector potential zeroes assumed that the ablation occurs normally to the plasma surface, it is necessary to confirm that zeroes are still predicted for specular ablation. Consider a p -polarised laser pulse confined to the x - y plane incident with an angle of incidence θ on an ablating overdense plasma expanding with velocity $-v_f \hat{x}$ in the lab frame, as in Figure 2.2.

The direction of polarisation is

$$\hat{\mathbf{r}}_{\text{pol}} = \hat{\mathbf{x}} \sin 2\theta - \hat{\mathbf{y}} \cos 2\theta \quad (2.9)$$

and the velocity of the rest frame of the ablating front relative to the lab frame is $-v_f \hat{\mathbf{x}}$.

Applying the Lorentz transformation to the electromagnetic 4-potential, \mathbf{A}^μ , where Λ_μ^ν is given by Equation 1.30, immediately from the y -coordinate transformation,

$$A'_L \cos 2\theta' = A_L \cos 2\theta. \quad (2.10)$$

Applying the headlight effect for a source moving at an angle 2θ to the boosted frame (a full derivation is given in Appendix A.2),

$$\cos(2\theta') = \frac{\cos(2\theta) - \beta}{1 - \beta \cos(2\theta)} \quad (2.11)$$

and rearranging Equation 2.10, the vector potential in the lab frame is

$$A_L = \frac{1 - \beta \sec(2\theta)}{1 - \beta \cos(2\theta)} A'_0 \cos(\omega'_L t') \exp(-r'/\delta'). \quad (2.12)$$

Writing the boosted frame space-time coordinates in terms of the lab frame coordinates,

$$ct' = \gamma(ct - \beta x), \quad (2.13)$$

$$x' = \gamma(x - \beta ct), \quad (2.14)$$

yields

$$A_L = A_0 \cos(\omega_L t - kx) \exp\left(-\frac{\sqrt{(x - \beta ct)^2 + (y/\gamma)^2}}{\delta}\right), \quad (2.15)$$

where

$$A_0 = \frac{1 - \beta \sec(2\theta)}{1 - \beta \cos(2\theta)} A'_0, \quad (2.16)$$

$$\omega_L = \gamma \omega'_L, \quad (2.17)$$

$$k = \frac{\beta \gamma \omega'_L}{c}, \quad (2.18)$$

$$\delta = \frac{\delta'}{\gamma}. \quad (2.19)$$

The oscillatory term in Equation 2.15 demonstrates the propagation of vector potential zeroes within the plasma target. From the structure of this term, it would appear that these zeroes are expelled from the plasma along the specular direction at a speed

$$v_\phi = \frac{\omega_L}{k} = \frac{c}{\beta} = -\frac{c^2}{v_f}. \quad (2.20)$$

In their original ZVP paper, Baeva laid some doubt on their version of this calculation, instead suggesting similarity theory predicts zeroes propagate at speed c [125]. This is important since it directly affects the properties of the reflected radiation. It is well known that the emission of radiation occurs primarily at the point where the transverse momentum goes to zero, corresponding to the passage of the zero of the vector potential [123] but equally that the width of the radiated pulse depends on the advance time emission point [132]. If the zeroes moved at speed c , then the observed emitted pulse would be infinitely thin, producing radiation with perfect coherence in all cases. This is so in the regime detailed in Chapter 3 where the pulse width is instead determined by the extent of radiating capability around the zero.

In the ZVP regime, where distinct bunches are formed at the plasma surface and the analysis above is valid, relevant to this chapter and Chapter 4, simulations have suggested finite advanced time bunch widths decreasing rapidly with increasing laser intensity [132]. This is precisely what is predicted by Equation 2.20. As the laser intensity increases, v_f naturally increases. Thus, the zeroes propagate closer to the speed of light and reduce the advanced time bunch width. The burst of radiation can be described by the Coherent Synchrotron Emission (CSE) theory of SHHG [128] or at least its modified form [132], thus CSE and ZVP are intrinsically linked, however, while CSE focuses on reflection and the SHHG spectrum, ZVP is concerned with laser pulse energy absorption.

To summarise, for a sufficiently intense laser pulse, electrons at the surface of an irradiated solid target are accelerated by the laser to relativistic velocities at a fraction of a laser pulse cycle and therefore electrons both follow similar trajectories

and respond adiabatically to the $\mathbf{J} \times \mathbf{B}$ force of the laser pulse. They form into a high charge density spatially thin coherent electron bunch on the front surface of the plasma but are displaced inwards from the approximately immobile ions via the ponderomotive pressure of the laser. This charge separation generates a longitudinal electrostatic pseudocapacitor field that confines electrons to a potential well on the front surface of the plasma, preventing further propagation of the electron bunch into the plasma bulk. When the zero of the vector potential passes through the electron bunch, the ponderomotive pressure instantaneously vanishes and electrons are ejected specularly from the target, co-propagating with the zeroes of the vector potential and emitting radiation.

After expulsion from the plasma, the pseudocapacitor is discharged as the electron bunch accelerates across it. Upon encountering the subsequent laser pulse peak amplitude, the bunch is then rotated back towards the plasma and launched into the bulk at high energy along the laser propagation axis (by conservation of transverse momentum in the plasma bulk), as it does so emitting coherent synchrotron radiation in transmission.

2.2.1 ZVP electron bunch energies

In [125], Baeva *et al* propose energy scalings in one-dimension (1D) for an electron bunch produced in the ZVP regime as a function of the incident laser pulse intensity and plasma density. This was extended to three-dimensions (3D) by Savin *et al* [124]. What follows is that discussion with closer consideration of both the constants of proportionality and their consequences. Note that throughout the electron bunch is treated as infinitesimally thin, as proved to be a reasonable assumption in previous work on ZVP [124, 131, 133]. Sub-bunch dynamics have been explored in more detail by Gonoskov *et al* [134].

Consider again the semi-infinite block of plasma presented in Figure 1.2, normally irradiated by a laser pulse with wavelength λ_L and peak electric field, E_L . It is now the ponderomotive pressure of the laser that displaces the electron fluid in

this picture. The electron surface moves inwards until the pressure exerted by the peak instantaneous ponderomotive pressure of the laser pulse cycle,

$$\mathbf{P}_L = \epsilon_0 E_L^2 \hat{\mathbf{x}} = \epsilon_0 \left(\frac{a_0 \omega_L m_e c}{e} \right)^2 \hat{\mathbf{x}} \quad (2.21)$$

is equal and opposite to the pressure exerted by the pseudocapacitor field,

$$\mathbf{P}_C = \frac{QE_C}{\sigma} \hat{\mathbf{x}} = - \frac{(en_e \Delta x)^2}{\epsilon_0} \hat{\mathbf{x}} \quad (2.22)$$

from Equations 1.16 and 1.17. Equating the magnitudes of \mathbf{P}_L and \mathbf{P}_C , the maximum displacement inwards of electrons is

$$\Delta x \hat{\mathbf{x}} = \frac{c}{\omega_L} \frac{a_0}{\bar{n}_e} \hat{\mathbf{x}} = \frac{1}{kS} \hat{\mathbf{x}}, \quad (2.23)$$

where k is the wavevector of the laser pulse. Correspondingly,

$$E_C = \frac{en_e}{\epsilon_0} \Delta x = \frac{\omega_L c m_e a_0}{e} = E_L. \quad (2.24)$$

Applying the results of Equations 2.23 and 2.24, when the ponderomotive pressure vanishes and the electron bunch is launched across the pseudocapacitor, the relativistic kinetic energy gained by a single electron is

$$T = \int \mathbf{F} \cdot d\mathbf{s} = \int_{\Delta x}^0 -eE_C dx = \int_{\Delta x}^0 -\frac{en_e x}{\epsilon_0} dx = \frac{1}{2} m_e c^2 \frac{a_0^2}{\bar{n}_e} \quad (2.25)$$

and an electron gamma factor,

$$\gamma = \frac{1}{\sqrt{1 - \beta^2}} = 1 + \frac{a_0^2}{2\bar{n}_e}. \quad (2.26)$$

Assuming all displaced electrons are captured by the potential well and launched as a coherent bunch, the total number of electrons in the bunch is

$$N_e = n_e \sigma \Delta x = \frac{\sigma a_0 n_c}{k} = \sigma \epsilon_0 E_L \quad (2.27)$$

and hence, the total kinetic energy of the electron bunch is

$$U_{ZVP} = N_e T = \frac{\sigma n_c}{k} \times \frac{1}{2} m_e c^2 \frac{a_0^3}{\bar{n}_e}. \quad (2.28)$$

It is now interesting to compare Equation 2.28 to the laser energy deposited upon the plasma surface and therefore consider what fraction of the laser energy

can be absorbed via the ZVP mechanism. Using $E_C = E_L$, Equation 2.28 can be rewritten as

$$U_{ZVP} = \frac{1}{2\omega_L S} \sigma c \epsilon_0 E_L^2. \quad (2.29)$$

The expressions for energies in Equations 2.25 and 2.28 require the electron bunch to fully discharge the pseudocapacitor before interaction with the subsequent laser pulse peak. Since the electron bunch travels at speed $\approx c$, the peak displacement (and thus the pseudocapacitor width) must satisfy

$$\Delta x \leq \frac{\lambda}{8}. \quad (2.30)$$

Using Equation 2.23, it is clear Equation 2.30 is satisfied for $S \geq 1.3$ at normal incidence.

For the case of normal incidence, bunches are produced at a frequency of $2\omega_L$, naturally following the frequency of the $\mathbf{J} \times \mathbf{B}$ force. Assuming a sinusoidal plane wave incident with surface area σ , the energy available during the pushing phase (a quarter-cycle) is

$$U_{L,1/4} = \sigma \frac{T}{4} \langle I_L \rangle = \frac{2\pi}{8\omega_L} \sigma c \epsilon_0 E_L^2. \quad (2.31)$$

Hence,

$$\eta_{ZVP} = \frac{U_{ZVP}}{U_{L,1/4}} = \frac{2}{\pi S}. \quad (2.32)$$

Interestingly, this analytical result predicts the trend observed by A. Savin [126] in PIC simulations both in magnitude and in scaling. Indeed, A. Savin demonstrated in numerical simulation

$$\eta_{ZVP} \sim S^{-1.000(3)}, \quad (2.33)$$

however, this result led A. Savin to conclude that increasing S reduces absorption, increasing the energy in the reflected SHHG beam thus increasing high harmonic efficiency, seemingly in tension with the results of other works [127, 132] that suggest $S = 1$ is optima. The resolution arises from awareness of two distinct conversion efficiencies that describe the reflected harmonic spectrum: the total conversion

efficiency into the full reflected beam and the conversion efficiency for individual harmonics. While the overall conversion into the reflected beam decreases for decreasing S , the slope of the harmonic spectrum becomes shallower and SHHG efficiency increases. Indeed, high harmonic efficiency necessitates high reflection inefficiencies due to the production of ZVP electron bunches as higher energy bunches produce more coherent reflected radiation [132].

Unfortunately, it is not possible to link the energy scaling derived in this Section to CSE since the harmonic emission occurs before the ZVP acceleration phase. In Dromey *et al*, the CSE regime was demonstrated for the first time in transmission through thin foils, however, laser pulses are now of sufficient intensity to access CSE in reflection from solid targets. This has been identified as a more efficient approach if it can be reached due to the production of higher density and shorter duration electron bunches and therefore brighter and more coherent CSE [135].

2.2.2 ZVP bunches oblique incidence scaling

This section is inspired by ideas from the work of Gonoskov *et al* [127] and Vincenti *et al* [129] to extend the theory of the ZVP mechanism for energy absorption to the more practical² case of oblique incidence.

Consider again the laser pulse incident on a solid density plasma existing for $x > 0$ at angle θ . Transforming to the frame of reference in which the laser is normally incident (quantities in this frame are indicated by the primed symbol), the electron and ion bulk plasma species stream at velocity $\mathbf{v}_d = -c \sin \theta \hat{\mathbf{y}}$. Applying the Lorentz force law along the longitudinal direction ($\hat{\mathbf{x}}$), for a displacement of the electron fluid x'_e (one assumes that the expression for a single electron at the surface describes the surface since all electrons follow similar trajectories), travelling at speed \mathbf{v}' ,

$$-e(\mathbf{v}'(x'_e) \times (\mathbf{B}'_L(x'_e) + \mathbf{B}'_i(x'_e)) \cdot \hat{\mathbf{x}} + E'_C(x'_e)) = 0, \quad (2.34)$$

where the laser magnetic field,

$$\mathbf{B}'_L = \frac{m_e \omega'_L a_0 \sin(\omega'_L t' - k' x'_e)}{e} \hat{\mathbf{z}} \quad (2.35)$$

²Not only is this more feasible in experiment but has been shown to optimise SHHG [127].

and B_i is the magnetic field generated by the uncompensated ion current, $\mathbf{J}_i = Zn'_i(x'_e)\mathbf{v}_d$, where the electron fluid has been displaced. As before, from Equation 1.17,

$$E'_C = \frac{en'_e x'_e}{\epsilon_0}. \quad (2.36)$$

Applying Maxwell-Ampère's Law, Equation 1.1c, and noting that by symmetry there can be no variation in the magnetic field with y' or z' , it becomes clear that

$$-\frac{d(\mathbf{B}'_i)_{z'}}{dx'} = \mu_0(\mathbf{J}_i)_{y'}. \quad (2.37)$$

Integrating Equation 2.37 from $-\infty$ to x'_e , noting that $\mathbf{B}_i(|\mathbf{x}| = \infty) = 0$ and assuming a constant density profile n'_i for $x > 0$ with $Zn_i = n_e$,

$$\mathbf{B}'_i(x'_e) = \mu_0 en'_e x'_e c \sin(\theta) \hat{\mathbf{z}}. \quad (2.38)$$

Using equations 2.35, 2.36 and 2.38 and making the reasonable approximation that the relativistic electrons on the surface move at speed $v'_y \approx \pm c$ at peak displacement ($x'_e = x'_p$), 2.34 can be written as

$$-e \left(\pm c \left(\pm \frac{m_e \omega'_L a_0}{e} + \mu_0 en'_e x'_p c \sin \theta \right) + \frac{en'_e x'_p}{\epsilon_0} \right) = 0. \quad (2.39)$$

For the laser to be in the pushing phase, the first term must be negative, corresponding to \mathbf{v}' and \mathbf{B}'_L having the opposite sign, hence,

$$c \left(-\frac{m_e \omega'_L a_0}{e} \pm \mu_0 en'_e x'_p c \sin \theta \right) + \frac{en'_e x'_p}{\epsilon_0} = 0, \quad (2.40)$$

where here the \pm tracks the sign of \mathbf{v}' . After some manipulation, one arrives at

$$x'_p = \frac{1}{k' S' (1 \pm \sin \theta)}. \quad (2.41)$$

Transforming back to the lab frame,

$$x_p = \frac{\cos^2 \theta}{k S (1 \pm \sin \theta)}. \quad (2.42)$$

Convincingly, this reduces to Equation 2.23 for $\theta = 0$ and predicts the suppression and enhancement of the two surface oscillations per laser pulse cycle. Explicitly, for a

laser pulse propagating at $y = x \tan \theta$, the peak displacement of the electron surface is enhanced for \mathbf{B}_L in the $+\hat{\mathbf{z}}$ -direction and suppressed for \mathbf{B}_L in the $-\hat{\mathbf{z}}$ -direction.

Again, integrating to find the work done as the electron bunch accelerates across the pseudocapacitor,

$$T(\theta) = \int \mathbf{F} \cdot d\mathbf{s} = \frac{1}{2} m_e c^2 \frac{a_0^2}{\bar{n}_e} \frac{\cos^4 \theta}{(1 \pm \sin \theta)^2}. \quad (2.43)$$

While it was to be expected that a component of the electric field acting into or out of the plasma would change the magnitude of the displacement, this result suggests that increasing the angle of incidence can increase the electron energy gain in the enhanced cycle more than the decrease in the suppressed cycle. Note that this is to be expected as it is known that SHHG efficiency is improved for non-zero angle of incidence [127, 132]. It would be interesting to explore whether the presence of an external magnetic field could be applied to mimic the effect of oblique incidence by replacing the magnetic field due to the uncompensated ion current.

While this model would suggest an optimal angle for electron energy and therefore of SHHG of $\pi/2$, if $\theta > \pi/4$, then, if the relativistic electron bunch is travelling at c along the specular reflection direction, the subsequent laser peak amplitude will never ‘catch up’ with the electron bunch, and electrons can escape, generating high charge electron bunches in reflection as observed in experiment by Lin *et al* [120], but reducing the efficiency of SHHG.

Finally, the total bunch energy as a function of θ ,

$$U_{ZVP}(\theta) = \frac{\sigma n_c}{k} \times \frac{1}{2} m_e c^2 \frac{a_0^3}{\bar{n}_e} \frac{\cos^6 \theta}{(1 \pm \sin \theta)^3}. \quad (2.44)$$

As anticipated, oblique incidence can increase laser absorption efficiency for higher S plasmas which are currently more easily accessed compared to the more optimal low S plasmas.

2.2.3 Defining characteristics of the ZVP mechanism

In her original paper on the ZVP mechanism, T. Baeva *et al* [125] outlined 6 defining characteristics of the ZVP mechanism, namely,

1. The existence of vector potential zeroes moving through the skin layer in the laboratory frame;
2. The existence of zeroes in the incident laser pulse vector potential required for the formation of fast electron bunches;
3. The generation of fast electron bunches with ultra-short temporal duration;
4. That such fast electron bunches follow the energy scalings of equations 2.25 and total energy 2.28;
5. Injection of the fast electron bunches is along the propagation axis of the laser pulse;
6. There must be an intrinsic link between the fast electron bunches and coherent SHHG;

with the moving zeroes within the skin layer being the key delineator between this post-ponderomotive regime of laser pulse energy absorption and other proposed mechanisms. While such observational requirements are far beyond the reaches of current experimental capacities, numerical simulations in both 1- [125] and 2-dimensions [124] have confirmed the above points. Now is presented the first 3D simulations attempting to demonstrate these criteria.

2.3 Numerical simulations of the ZVP mechanism

2.3.1 The ZVP mechanism in 3D3V

3D simulation results are presented in Figure 2.3 alongside comparison to an equivalent 2D simulation. Simulation parameters are given in Table 2.1, such parameters are compatible with the 10 PW ELL-NP state-of-the-art short pulse laser facility [136] incident on foam targets. Figure 2.3c) clearly reveals the existence of high energy density electron bunches propagating through the plasma bulk in the direction of the laser pulse. Note that this ZVP criterion is a direct consequence of conservation of transverse momentum inside the plasma bulk where the laser

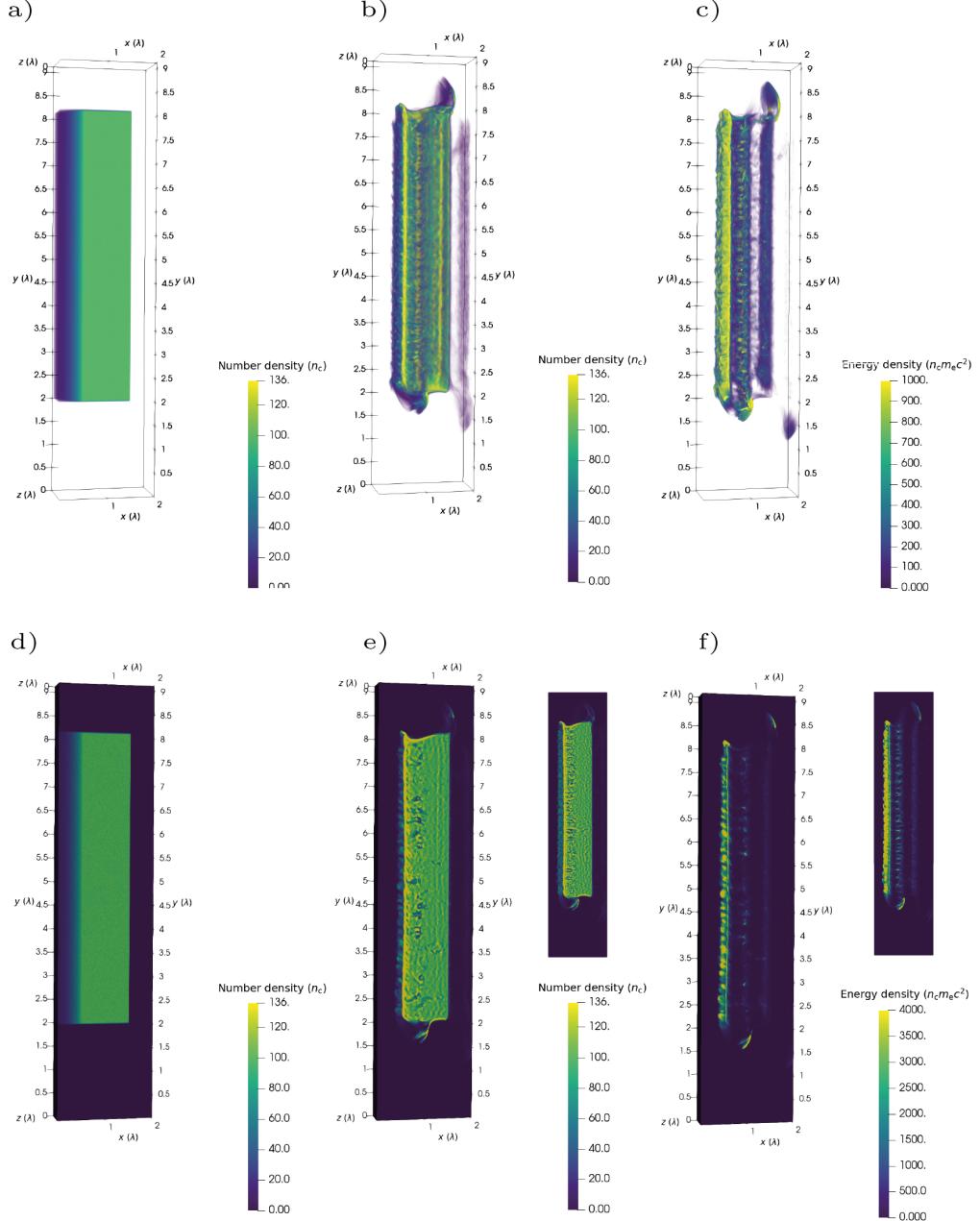


Figure 2.3: Simulation results from a 3D PIC simulation of the ZVP mechanism. a) The initialised electron number density. b) The electron number density several cycles later, the plasma bulk is intact, however, there is evidence of instabilities and electron bunches propagating through and around the plasma. c) The electron kinetic energy density at the same timestep. Note that the scale has been clipped to enable observation of both electron bunches propagating through and around the plasma bulk. Significantly higher energy density, corresponding to a higher charge density and attosecond duration is observed for the electron bunches propagating around the bulk. d-f) Plots clipped through $z = \lambda_L/2$ for a-c) respectively to access the internal structure of the plasma bulk. The accompanying plots for figures e) and f) are equivalent 2D PIC simulations demonstrating excellent agreement.

Laser (3D, normal incidence)		
Parameters	Real	Smilei
Wavelength, λ (nm)	1060	2π
Angular frequency, ω_L (fs $^{-1}$)	1.8	1
Beam waist, w_L (nm)	6λ	12π
Focal point, (f_x, f_y, f_z) (nm)	$(0.5\lambda, 5\lambda, 0.5\lambda)$	$(\pi, 10\pi, \pi)$
Spatial envelope, E_i , $i = y, z$	$E_i \sim e^{-(i-i_f)^2/w_L^2}$	
Temporal envelope, E_t	$E_t \sim e^{-(t-4\lambda/c)^2/((4\lambda/3c)^2 \ln 2)}$	
Simulation box		
Size, $x \times y \times z$ (nm)	$2\lambda \times 9\lambda \times \lambda$	$4\pi \times 18\pi \times 2\pi$
Sim length (fs)	35.22	20π
Spatial resolution, Δx (nm)	$\lambda/128 = 8.28$	0.0491
Temporal resolution, Δt (as)	$\Delta x/11c = 2.51$	0.00446
Collisionless, pre-ionised randomly-initialised aluminium plasma		
Electron x profile, $n(x)$	$\begin{cases} n_e & \text{for } 2\lambda \leq x \leq 3\lambda, \\ n_e e^{(x-2\lambda)/0.2\lambda} & \text{for } x \leq 2\lambda. \end{cases}$	
Electron y profile, $n(y)$	$\begin{cases} 1 & \text{for } 2\lambda \leq y \leq 8\lambda, \\ 0 & \text{otherwise.} \end{cases}$	
Electron z profile, $n(z)$	$\begin{cases} 1 & \text{for } 0.125\lambda \leq z \leq 0.875\lambda, \\ 0 & \text{otherwise.} \end{cases}$	
Ion profile, $n_i(x, y, z)$	$n_i = n(x)n(y)n(z)/13$	
Macro-electrons per cell	729	
Macro-ions per cell	8	
Ion temperature, T_i (keV)	0	0
Electron temperature, T_e (keV)	10	0.02
Stability criteria		
$\lambda_D/\Delta x$	0.288	
$1/\Delta t \omega_p$	24.4	
$\Delta x/c\Delta t$	11	
Macro-particles in the Debye sphere	210	

Table 2.1: Simulation parameters in both real and normalised Smilei simulation units for the 3D3V simulations.

fields cannot propagate. Figure 2.3b) shows these bunches escape to the rear of the bulk but lose energy as they do so. Looking now at Figure 2.3e) and the internal structure of the plasma bulk, these bunches drive two-stream and filamentation instabilities [137]. The bulk propagating electron bunches are accompanied by higher density electron bunches to either side of the plasma block with the side of emission alternating every half laser pulse cycle with the $\mathbf{J} \times \mathbf{B}$ force.

Considerations and convergence of 3D simulations

The plasma specifications were chosen to minimise computational load while ensuring numerical convergence, requiring over 100 billion macroparticles. The electron temperature is raised significantly higher than that which would be expected in such a laser-plasma system so as to resolve the Debye length. Possible plasma temperatures are calculated using 1D HYADES simulations in the following Chapter. While this temperature is unphysical and will lead to some small plasma expansion over the course of the simulation (negligible on these timescales), the temperature remains small compared to the energy imparted to the electron bunches by the laser pulse. The striking similarity between the 2 and 3D simulation results is a natural consequence of the 2D nature of the interaction geometry. It is, however, still reassuring to show that previous work withstands the test of reality's geometry. The simple elegance of the ZVP mechanism is not lost in the chaos.

For the 3D simulations only, particles were initialised randomly not regularly to avoid numerical error. Regularly initialised plasma blocks in 3D simulations rapidly blow apart due to spurious large amplitude fields generated on the large plasma surfaces due to macroscopic electron-ion charge separation at initialisation. 2D simulations with randomly initialised particles tend to produce nano-structures reminiscent of the cosmic web. The absence of these structures in 3D is telling. Unsurprisingly this error can be reduced by increasing the plasma temperature.

The longitudinal thickness of the target does not impact the interaction and is chosen for computational efficiency, indeed it is standard to consider such targets of thickness $\geq \lambda_L$ as bulk targets [71], however, for sufficiently long pulse durations,

the effect of hole boring necessitates thicker targets to make this approximation as is necessary in the following Chapter.

The initial 3D simulation parameters were chosen to be consistent with previous work on the ZVP mechanism, however, such simulations are cumbersome. In order to query the defining characteristics outlined by Baeva, a lower-resolution simulation was performed with similar parameters to the initial simulation. Comparisons between the simulation outputs are made in Figure B.1 in Appendix B.1. Good convergence is qualitatively demonstrated by the presence of characteristic features of the ZVP mechanism. While the instabilities are similar in structure, the change in seeding changes their exact positions. As instabilities are not the focus of this thesis this variation is of no cause for concern.

Confirmation of ZVP in 3D

Figure 2.4 tracks the transverse momentum distribution along the polarisation axis of the laser pulse of an electron bunch during its ablative journey, clearly demonstrating the existence of a singular zero of the vector potential propagating through the electron bunch and away from the plasma bulk. The zero propagates at a speed of $1.4c$, corresponding to a surface velocity of $0.71c$. In this simulation, the zero propagates through the bunch over a few nanometres and before crossing the pseudocapacitor. Since Gonoskov *et al* have determined a scaling for electron bunch width [127], calculation of the advance time bunch width, and correspondingly the duration of the attosecond pulse, only requires the calculation of the surface velocity. It is unfortunate that this emission occurs before the ZVP phase.

Further simulation results are presented in Figure 2.5. In Figures 2.5a)-d), comparisons are made of the electron energy distributions for simulations with zeroes present in the vector potential (linearly polarised) and without (circularly polarised). Evidently, zeroes are required for the formation of high energy, short duration attosecond bunches. From Figure 2.5a) it is clear that electron bunches are injected, propagating through the plasma bulk in the laser propagation direction. Figure 2.5c) demonstrates the quasi-monoenergeticity of the high-energy electron

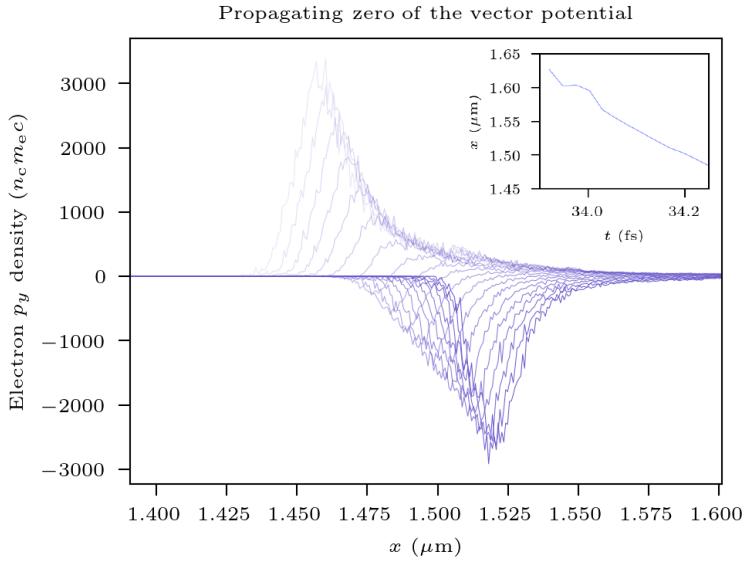


Figure 2.4: Propagation of a vector potential zero through the ablating ZVP electron bunch via the proxy of transverse momentum conservation. The zero of the vector potential exists where the transverse momentum is macroscopically zero. Each line represents a timestep with time increasing with decreasing colour. With increasing time the bunch and the zero move in the $-\hat{\mathbf{x}}$ -direction with the zero overtaking the electron bunch. The inset tracks the position of the zero of the vector potential with time.

bunches as initially identified in 1D by Baeva. Although the shape in the phase space is more complex in 3D, the attosecond duration at a given energy is retained. Figures 2.5e) and f) describe the surface dynamics in the linearly polarised case. One can observe the high-density bunches on the front surface with the peak in energy density occurring after acceleration across the pseudocapacitor.

Finally, Figure 2.6 compares the spectra of the reflected light in the presence and absence of zeroes of the vector potential. Individual harmonics are not resolved due to blueshifts between successive pulse cycles from considerable hole boring, a phenomenon discussed in great detail in the following Chapter. The spectrum from the circularly polarised light is typically over two orders of magnitude below that of linearly polarised light. Thus, all defining characteristics of the ZVP mechanism have been identified in 3D simulations with the notable exception of the energy scalings. Next-generation supercomputers will be required to perform such large scale parameter scans.

2.3. Numerical simulations of the ZVP mechanism

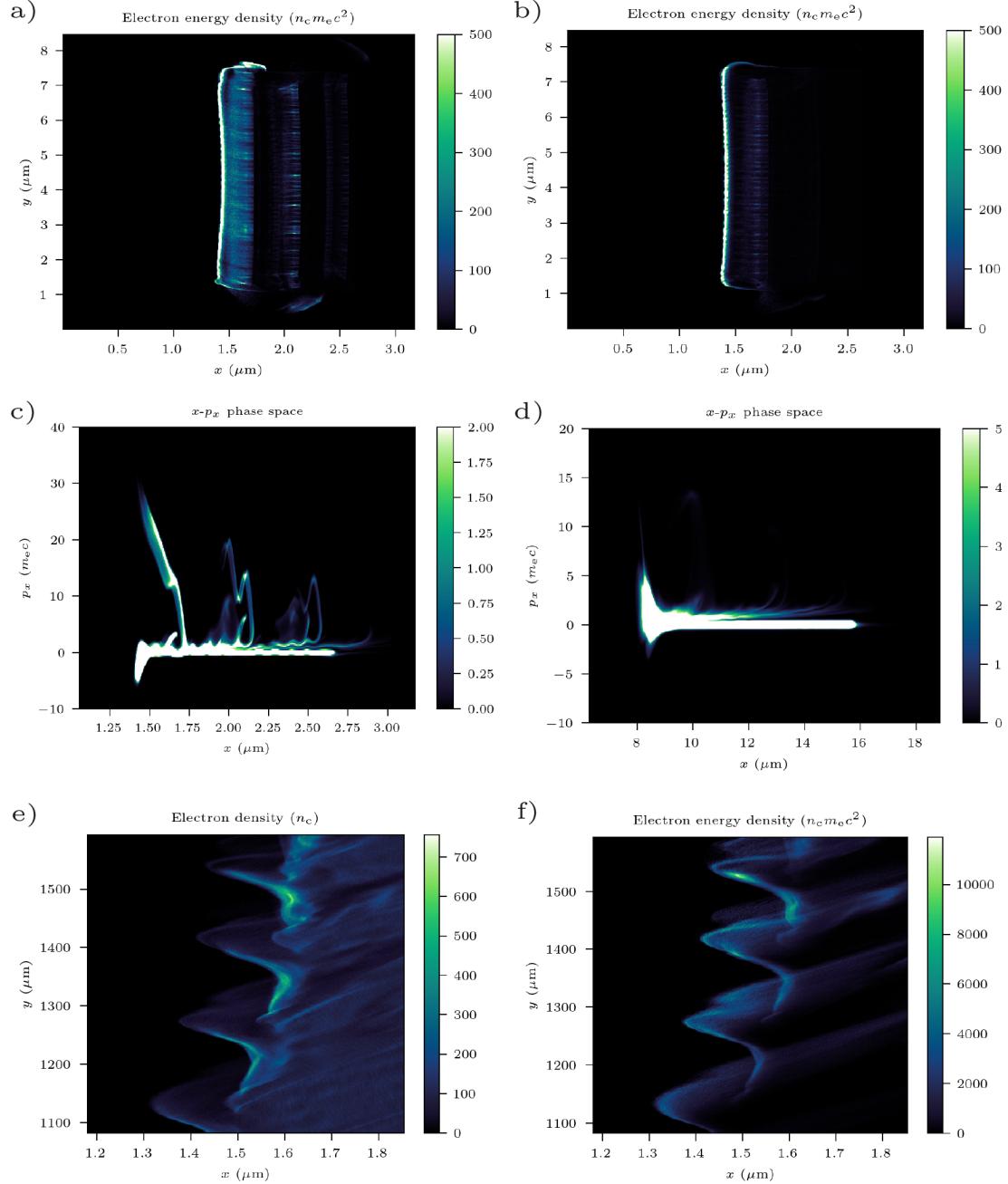


Figure 2.5: Electron dynamics in a 3D PIC simulation. a) and b) Electron energy density for linearly and circularly polarised laser pulses respectively. c) and d) Electron longitudinal momentum for linearly and circularly polarised laser pulses respectively. e) Electron density at the plasma surface streaked in time. f) Electron energy density at the plasma surface streaked in time.

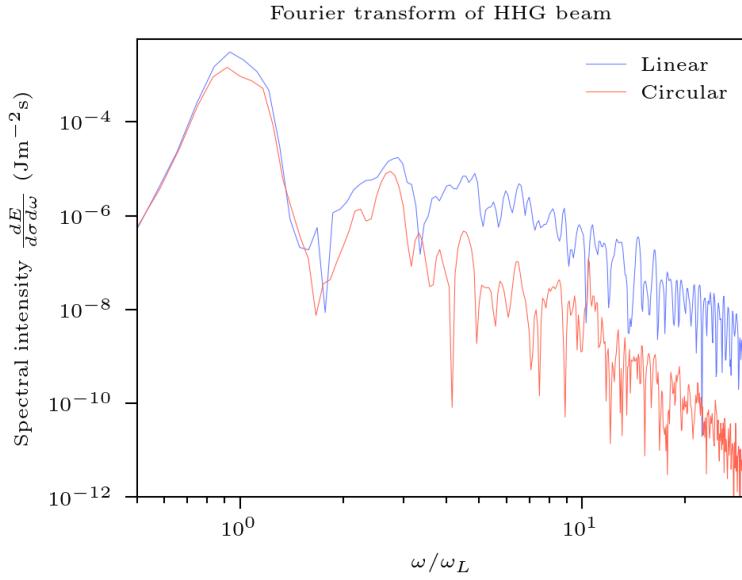


Figure 2.6: The Fourier transform of the reflected laser pulse in 3D PIC simulations both with and without zeroes in the vector potential.

2.4 The ZVP electron bunch

Following the excellent agreement demonstrated between 2 and 3D PIC simulations, the remainder of this Chapter explores ZVP in 2D. Previous work on the ZVP mechanism has highlighted the high energy and short duration of electron bunches, what follows is a full characterisation of their properties. A ZVP electron bunch is an electron bunch produced via the ZVP mechanism. Once produced and accelerated across the pseudocapacitor field, it is launched back in the laser propagation direction. While the bunch has no spatial separation between energies when propagating with the zero of the vector potential, the turning point of the electrons is longitudinal momentum dependent due to the Coulomb attraction of the ions after overshooting the pseudocapacitor field. Baeva *et al* showed that the electron bunch has a quasi-monoenergetic spectrum: the electron bunch is now of attosecond duration in the spectro-temporal domain and there is a one-to-one relationship between energy and position with the higher energies trailing the lower energies. The full bunch is confined to 130 as while a single energy is confined to 5 as. If, however, the plasma bulk is transversely mass-limited relative to the laser spot size, when rotated back

towards the plasma block, some of the electron bunch will overshoot and escape the potential well without significant chirping of the bunch as can be seen in Figure 2.3. Such electron bunches retain their high charge density and ultra-short duration with the tradeoff of increased divergence. Note that electron bunch properties are imprinted in the transmitted harmonics: high divergence but attosecond duration.

ZVP electron bunches can therefore be placed into two categories: ultra-high charge, ultra-short duration electron bunches from mass-limited targets, hereafter labelled mass-limited electron bunches, of interest due to their unique properties, and bulk propagating bunches, hereafter labelled bulk bunches, which have lower charge densities, are imprinted with instabilities and are instead of interest due to their connection to energy absorption and reflection in this post-ponderomotive regime. To investigate these two bunch types further, 2D PIC simulations were performed.

Attosecond nano-Coulomb mass-limited electron bunches

The plots of Figure 2.7 describe a typical mass-limited ZVP electron bunch qualitatively. The electron bunch under interrogation is ultra-relativistic with a mean energy of 51(11) MeV and a duration of 35 as. It propagates at an angle of -0.393 rad relative to the laser propagation direction, *i.e.* the x -axis, and has a transverse geometric emittance in the simulation plane (the x - y plane) of 35(7) nm rad. The calculation of the transverse geometric emittance, a measure of the quality of the electron beam, is given in Appendix A.3. Note that while the bunch does not propagate in the laser propagation direction, this does not mean it must be rejected under consideration of the ZVP bunch conditions. We must expand the definition of a ZVP electron bunch. In the mass-limited case, a bunch must propagate at some angle to the laser due to conservation of canonical momentum while it remains in the thrall of the laser pulse. For an equivalent bunch in a corresponding 3D simulation, the transverse geometric emittance in the z plane is 7.4(10) nm rad. This electron bunch has a total charge of 0.35 nC for a slab of plasma of thickness 0.75λ in the z -direction. Note again the two-dimensional nature of the interaction geometry. Electrons a distance less than

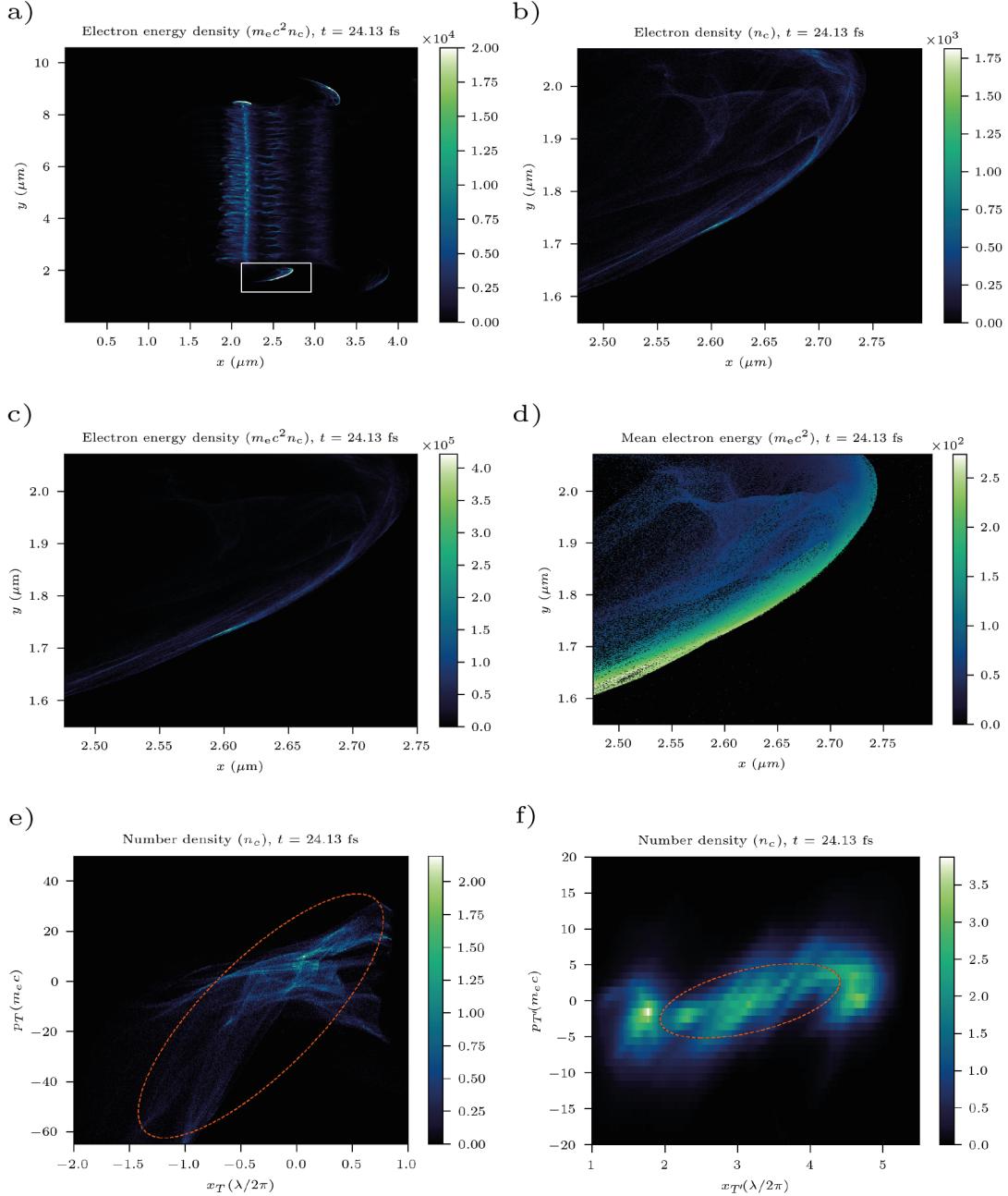


Figure 2.7: 2D PIC simulation results qualitatively describing typical mass-limited ZVP electron bunch structure. a) Electron energy density for the full simulation window, corresponds to Figure 2.3f). The box highlights the bunch presented in the following plots. b) Electron number density of the electron bunch. c) Electron energy density of the electron bunch, the colour bar scale has been increased compared to Figure a) to observe the internal structure. d) The mean electron energy across the electron bunch, suggesting a position-dependent energy or quasi-monoenergetic nature to the electron bunch [125]. Cells with no macroparticles are black. e) The transverse phase space in the 2D simulation plane. The ellipse describes the calculated emittance. The skew of the ellipse is a consequence of a low-density tail on the phase space beyond the bottom left corner. f) This plot was extracted from the equivalent 3D simulation and describes the transverse emittance in the z -direction. Again the ellipse marks the emittance. The relatively well-defined border to the phase space and the mild tilt (indicating only mild divergence) are direct consequences of the 2D nature of the interaction.

twice the relativistic Larmor radius,

$$r_L = \frac{\gamma m_e v}{e|\mathbf{B}|} \quad (2.45)$$

will escape to the side of the target when rotated back towards the plasma. Here, γ and v correspond to the electron velocity. The total number of electrons in the mass-limited bunch is

$$N = 2n_e r_L L_z \Delta x, \quad (2.46)$$

where L_z is the width of the plasma in the z -direction. Combining Equations 2.45 and 2.46 with 2.23 and 2.26 and taking $v \approx c$ for the ultra-relativistic electron bunch,

$$N \approx 2\gamma n_c \frac{L_z}{k^2}. \quad (2.47)$$

Equation 2.47 can be rewritten in terms of fundamental constants as

$$N \approx 2(1 + 0.5 \frac{a_0^2}{n_e}) L_z \frac{m_e \epsilon_0 c^2}{e^2}. \quad (2.48)$$

For these simulation parameters, this corresponds to a total bunch charge, $Q = eN$, of 0.37 nC, a remarkably successful prediction of the ZVP model. Perhaps counter-intuitively, it would appear the total charge scales inversely with the plasma density. Instead, charge can be increased either by increasing the laser pulse intensity or L_z . Indeed, provided the laser pulse intensity remains relativistic, the focal spot can be increased indefinitely and there is no limit to the mass-limited electron bunch total charge. For a realistic laser pulse with beam width 10λ incident on a larger plasma block, Equation 2.48 predicts a charge of 9.3 nC.

Caution is necessary when the standard definition of emittance is applied to a non-Gaussian beam profile, risking over- or underestimating the emittance. When applied to an ideal Gaussian distribution, the elliptical contour that defines the emittance contains 39.3 % of the beam particles [138], however, Figures 2.7 e) and f) contain 75.5 % and 38.6 % of the beam respectively. The large overestimate of the former is a consequence of the long low-density tail on the distribution to the bottom left of the plot.

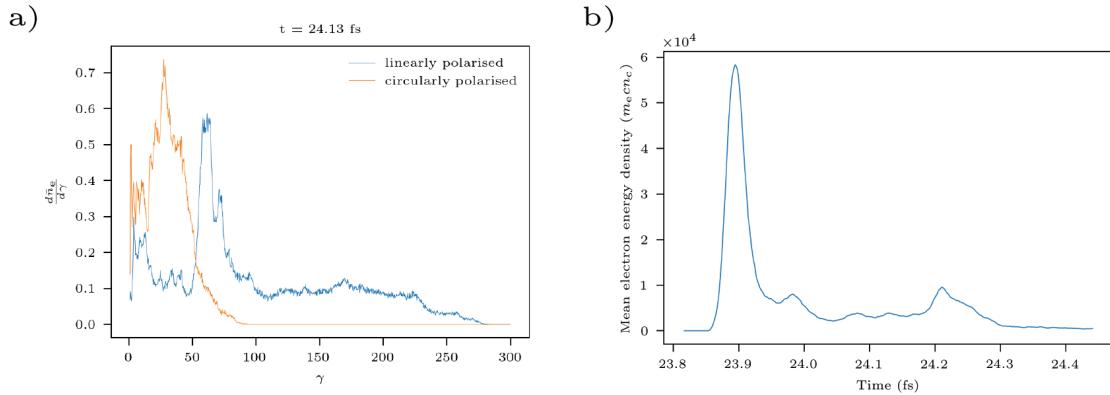


Figure 2.8: Energy spectra for mass-limited electron bunches formed via linearly and circularly polarised laser pulses. a) Energy spectra for mass-limited electron bunches formed via linearly and circularly polarised laser pulses. b) Mean energy density streaked in time through the centre of a mass-limited ZVP electron bunch demonstrating the attosecond duration.

Figure 2.8a compares the electron bunch energy to an equivalent bunch produced by a circularly polarised laser pulse³. The mean electron bunch energy is over three times lower as there is no ZVP acceleration phase and there is no quasi-monochromatic structure [125]. Figure 2.8 b) plots the mean electron bunch energy in time at a point as the bunch passes, clearly demonstrating the attosecond duration of the electron bunch.

Applications: from electron bunches to attosecond light

There is a plethora of applications for high charge, attosecond electron bunches, primarily for the purpose of resolving attosecond scale phenomena [74]. Already, femtosecond pump-attosecond probe experiments are underway [139, 140] but the higher intensities and charge densities accessible in the laser-solid regime compared to in laser-gas interactions [120, 132] would represent a step-change in the field by enabling atto-pump atto-probe experiments [116]. Potential applications include: electron microscopy and atomic diffraction to temporally resolve photoelectric processes such as Bragg diffraction [141], ultra-fast electron radiography to probe the evolution of the formation of magnetic fields in dynamical systems [142] or

³A circularly polarised laser pulse will expel electrons from a mass-limited target in a corkscrew shape, the bunch is therefore only loosely equivalent.

for XFEL seeding [121]: the record XFEL X-ray pulse duration stands at 280 attoseconds [143], substantially longer than the durations accessible using this technique. Electron bunches are also a promising alternative for radiotherapy due to their superior penetration depth in biotissues compared to X-rays [144].

Analogously to the SHHG process, rapid acceleration of an electron bunch generates a burst of radiation whose properties (brightness, coherency, duration, spectrum) are determined by the corresponding properties of the electron bunch (charge, emittance, duration and energy). Thus globally electron bunches are used as a diagnostic tool in synchrotrons and XFELs. Other acceleration mechanisms for X-ray generation include: bremmstrahlung radiation from firing the electron bunch at a secondary high- Z target [145], interaction with a counter-propagating laser pulse [146, 147] or injection into a laser or plasma wakefield accelerator, even with charge densities suitable for accession to the solid density plasma wakefield regime [120]. The mass-limited electron bunches produced via the ZVP mechanism have transverse emittances comparable in all planes to those conditioned in state-of-the-art nano-Coulomb electron bunch accelerators [148, 149]. Such facilities typically produce electron bunches with geometric emittances of \sim mm rad prior to damping ring injection [150] and \sim nm rad post-injection [148]. Thus, the mass-limited ZVP electron bunches are ideal candidates for the production of bright X-rays of unprecedentedly short duration and are suitable for study in the new attosecond regime with applications to physical, chemical and biological systems.

Parameter scan of electron bunch mean energy

It is important to confirm that mass-limited electron bunches follow the same scaling relations as has previously been confirmed for bulk ZVP electrons in both 1D [125] and 2D [124] PIC simulations. The mean mass-limited electron bunch kinetic energies were extracted from 120 2D PIC simulations and are plotted in Figure 2.9. The dependence of the energy on both a_0 and \bar{n}_e is immediately apparent. Therefore, these electron bunches must be accelerated by a non-ponderomotive mechanism.

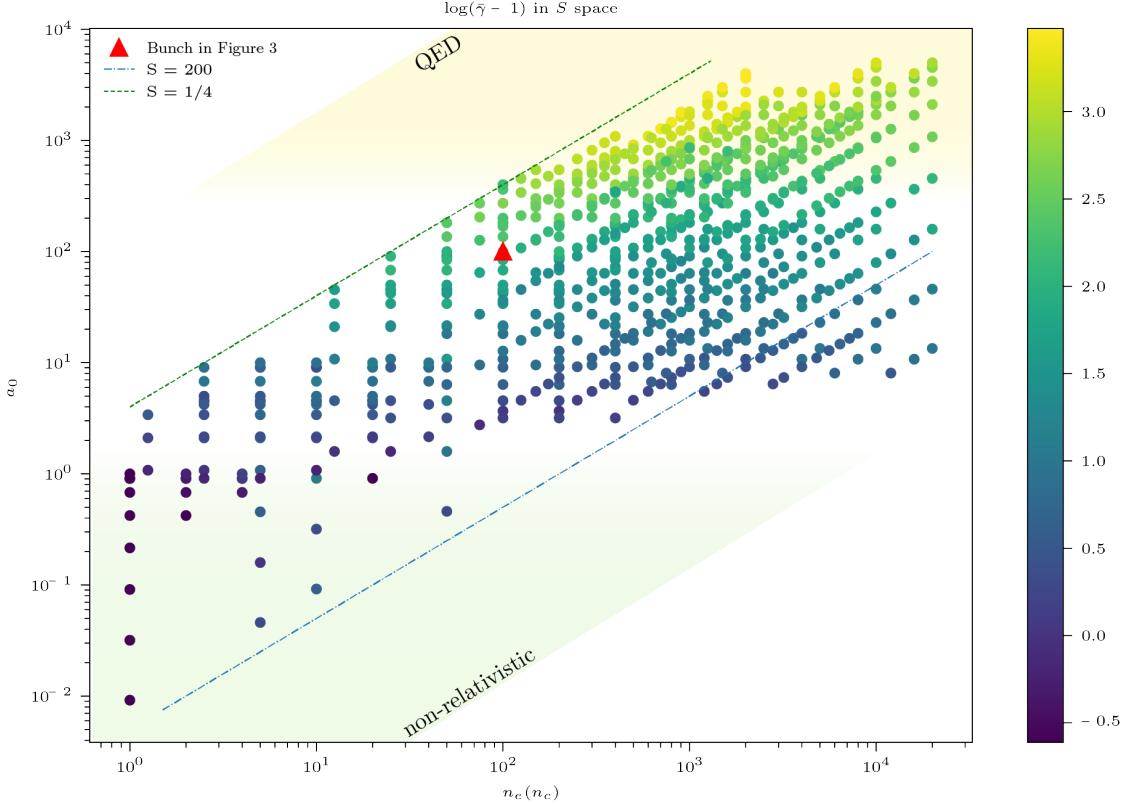


Figure 2.9: Mean mass-limited ZVP electron bunch normalised kinetic energies extracted from 2D PIC simulations. The bunch detailed in Figure 2.7 is highlighted.

The parameter scan took plasma block densities ranging from the critical plasma density to well-beyond solid density for the aluminium target and with laser pulse peak intensities ranging from non-relativistic ($a_0 < 1$) through to the Quantum Electro-Dynamics (QED) plasma regime ($a_0 > 300$) up to a peak $a_0 = 5000$ to investigate the change in scaling observed by Savin *et al* [133] at the onset of QED effects. This study is also the first to extract specific bunch energies as opposed to total simulation box energy gain, representing a far more precise test of ZVP theory. Particle merging was turned on for macro-photons and macro-electrons for $a_0 \geq 1800$ to prevent overloading of the available supercomputer memory due to the vast number of particles produced in this SF-QED regime.

A total of 856 data points for mass-limited electron bunch mean energies were extracted from the simulations. Bunches are arranged in trains with an average length of 4, however, isolated bunches are more useful for attosecond diagnostics. The number of bunches in a given train as a function of simulation peak intensity

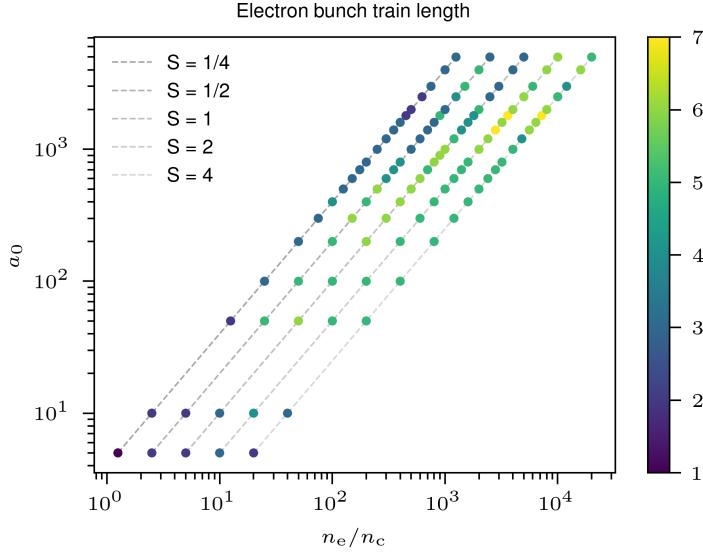


Figure 2.10: Mass-limited electron bunch train length as a function of laser intensity and plasma density. Lines of constant peak S parameter are plotted for reference. Clearly, decreasing S reduces the bunch length.

and plasma density is plotted in Figure 2.10. It is evident that reducing the peak S reduces the train length. Where $S < 1$, the transition to relativistic self-induced transparency occurs in the rising edge of the laser pulse corresponding to a breakdown of ZVP and an early cutoff to the bunch train, suggesting one route to the more useful isolated attosecond electron bunches. Other routes to isolate electron bunches can be likely borrowed from CSE theory given the close relation between the two. The interest in isolated attosecond pulse of radiation has led to the proliferation of ideas, proposed techniques include the attosecond lighthouse technique using laser pulse wavefront rotation [151], few-cycle laser pulses [152], non-collinear laser pulse gating [153], soon to be performed in experiment, and circular polarisation gating [154].

Return now to Figure 2.9. The ZVP energy relations must be modified before comparison to the data since these electron bunches experience a further direct laser acceleration phase before reaching the measurement point. Thévenet *et al* [122] suggested that attosecond electron bunches produced in reflection exhibit precisely the phase and energy properties required to ‘surf’ the reflected laser pulse and experience vast acceleration gradients over the Rayleigh length of the laser

pulse. This process is known as Vacuum Laser Acceleration and could provide a route to a fully optical scheme to create GeV nano-Coulomb electron bunches. Certainly, some of the bunch will have the necessary injection close to the laser propagation axis, however much of the bunch propagates at an angle to the laser and this must be accounted for. Interestingly, in Figure 4.2 modelling oblique incidence, more of the bunch appears to propagate along the axis. A parameter scan is necessary to find the optimal parameters.

Electrons are ejected from the plasma in phase with the subsequent laser pulse cycle peak and diverge outwards from that point. Near to the laser focus, the electrons experience a field

$$\mathbf{E}(x, y, t) = E_0 e^{-(y-f_y)^2/w_L^2} \cos(k_L(x-f_x) - \omega_L t) \hat{\mathbf{y}} = E_0 f(x, y, t) \hat{\mathbf{y}}. \quad (2.49)$$

The work done by this field is then

$$\Delta T = \int e \mathbf{E} \cdot d\mathbf{x}. \quad (2.50)$$

Note that for this process, the laser pulse electric field and electron bunch direction of travel will always be aligned no matter to which side of the plasma bulk the electrons are accelerated and therefore ΔT will always increase the energy of the electron bunch. The electron trajectories can be approximated as linear from the ejection point (y_e, x_e) to the observation point (y', x') , correspondingly,

$$x = y \frac{(x' - x_e)}{(y' - y_e)}, \quad t = \sqrt{x^2 + y^2}/c. \quad (2.51)$$

Equation 2.50 can now be integrated along a given electron's path and therefore the gamma factor increases by

$$\Delta\gamma = \int \frac{e E_y(y) dy}{m_e c^2} = a'_0(y) F, \quad (2.52)$$

where a'_0 is the vector potential of the subsequent laser pulse cycle peak and

$$F = \frac{2\pi}{\lambda} \int f(y) dy. \quad (2.53)$$

The gamma factor gain from the ZVP acceleration phase is determined by the electric field at the corner of the plasma block,

$$\Delta\gamma_{\text{ZVP}} = \frac{(a_0 e^{-(2\lambda - y_f)^2/L_0^2})^2}{2\bar{n}_e} = 0.31 \frac{a_0^2}{\bar{n}_e}. \quad (2.54)$$

Thus,

$$\gamma = 1 + (0.31) \times \frac{a_0^2}{\bar{n}_e} + F \times a'_0. \quad (2.55)$$

The final term could be neglected or at least reduced somewhat once super-Gaussian spatial laser pulses become common in this intensity regime or by use of a suitable plasma separator [155], as applied in [156]. Both acceleration phases fail to meet the criteria of the Lawson-Woodward theorem. The ZVP phase is dependent on the existence of electrostatic forces, while the secondary phase occurs for a finite interaction region.

The data of Figure 2.9 is taken from two measurement points, enabling two F values to be investigated. Fits to Equation 2.55 were made for each data set using the Ordinary Least Squares regression model of the statsmodels Python module [157], allowing the constants to vary. The non-linear ZVP energy can be applied to the linear model via construction of the necessary composite parameter. Both fits were successful with r^2 -values of 0.81 and 0.84 and found the same ZVP pre-factor $F_{\text{ZVP}} = 0.47 \pm 0.02$, slightly above that anticipated from the model. The model is sensitive to the laser pulse intensity, likely much of the bunch originated from closer to the laser pulse focus than the target corner explaining the higher value.

The first data set measurement point was $(0.41 \mu\text{m}, 0.25 \mu\text{m})$ from the target edge. This gives $F = 0.31$ compared to 0.22 ± 0.02 predicted by the fit. The second, at $(0.41 \mu\text{m}, 0.31 \mu\text{m})$ from the target edge giving $F = 0.39$ compared to 0.34 ± 0.02 from the fit.

This is the first demonstration of ZVP theory to calculate absolute values and not only the scaling relationship. Such order of magnitude calculation is essential for the comparison to other absorption models and the determination of dominant modes. It is certainly remarkable that such a simple theory for energy absorption

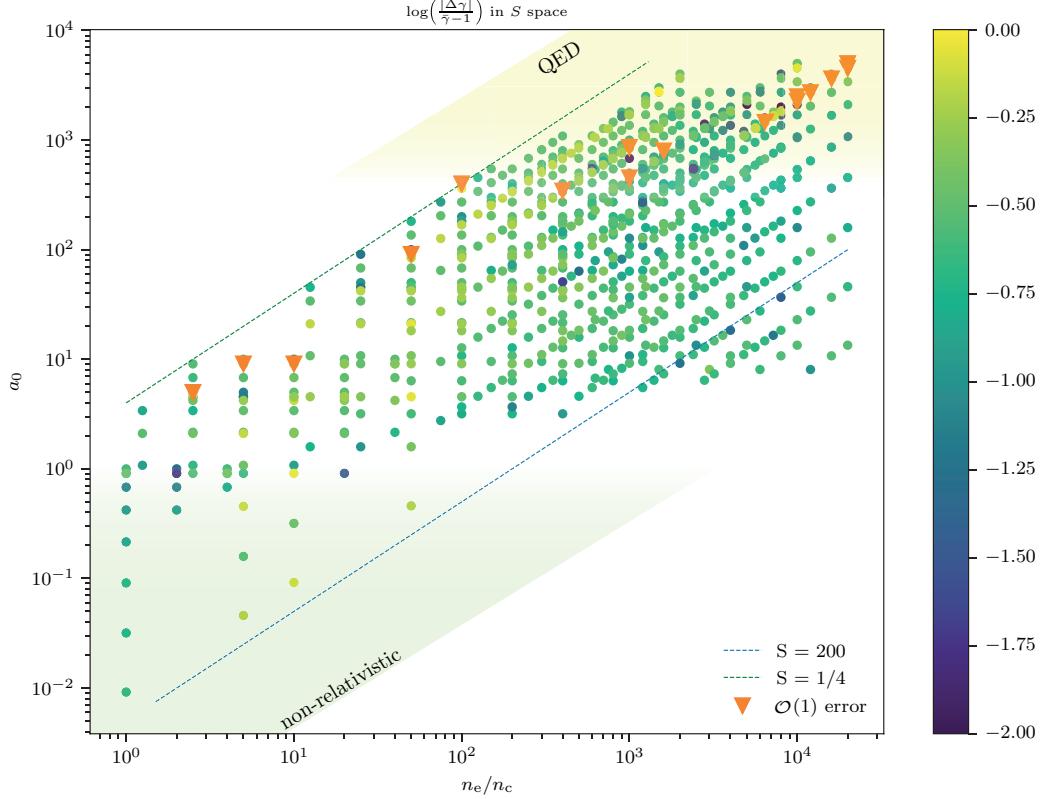


Figure 2.11: The relative errors for each mean energy data point compared to Figure 2.9. The orange triangles indicate data points for which the model fails to predict the mean energy.

has such predictive power in this highly non-linear and seemingly chaotic many-particle system. It is interesting that increasing laser intensity to such extremes will, at least for a short time, cause relativistic effects that simplify the dynamics before the total annihilation of a target.

The relative error between data and theory is plotted in Figure 2.11. Those points marked by an orange triangle have associated errors of over an order of magnitude. Reassuringly, such points occur only after the onset of QED effects, known to impact the ZVP mechanism [133], and for $S < 1$, that is, where the plasma becomes relativistically transparent to the laser pulse, a fundamentally different regime. The model is inconsistent in the non-relativistic domain as can be expected. To summarise, it would appear the ZVP model is valid for $10 \leq a_0 \leq 300$ and $S \geq 1$.

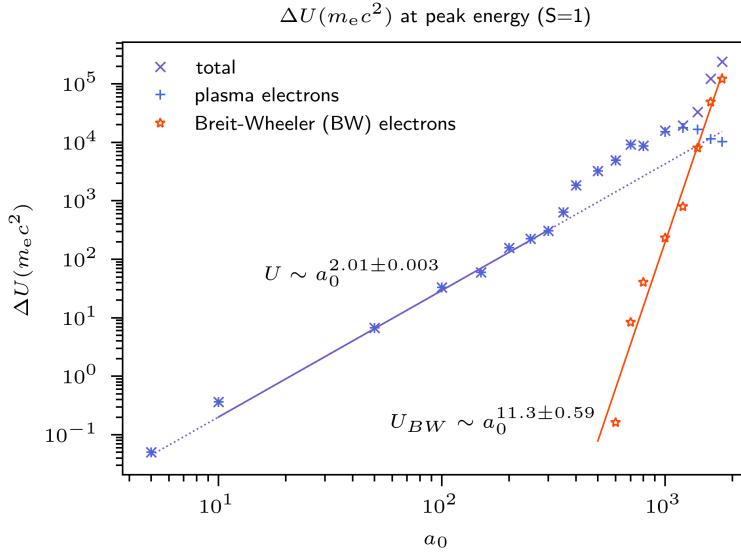


Figure 2.12: Peak instantaneous bulk electron bunch total energies after escaping to the plasma bulk rear. Energies begin to deviate from the anticipated scaling at approximately $a_0 = 300$. Above $a_0 = 1000$, BW pair-produced electrons begin to dominate and the peak energies rise rapidly.

2.4.1 Energy absorption in the ZVP regime

As stated previously the laser-plasma coupling exists in a state of adiabaticity with the exception of the ZVP acceleration phase and hence Equation 2.28 describes the absorption of laser pulse energy. As two bunches are produced per laser pulse cycle, the rate of energy transfer is, therefore,

$$R = \frac{U\omega_L}{\pi}, \quad (2.56)$$

for normal incidence. To observe the scaling for U in 2D PIC simulations, peak instantaneous electron bunch energies escaping to the rear of the bulk were extracted from those PIC simulations with $S = 1$. For constant S ,

$$U \sim a_0^2. \quad (2.57)$$

Energies are plotted in Figure 2.12. Fitting the total energy within the range of validity established for the ZVP model finds

$$U \sim a_0^{2.01\pm 0.003}, \quad (2.58)$$

reproducing with great success the anticipated scaling within the ZVP regime. It is satisfying that the number of electrons and bunch mean energies both follow their anticipated ZVP scalings. It was not possible to reproduce the constants of Equation 2.28 as the neutralising return current in the plasma bulk generates an electrostatic field on the rear side of the plasma block, decelerating bulk electron bunches as they escape the plasma. It should be possible to calculate the deceleration by considering the number of electrons expelled by the plasma. It is, however, clear from the simulations that at least some electrons in the escaping bunch are trapped by this rear-side potential well and thus reduce its ability to slow electrons.

While Equation 2.28 describes energy absorption into hot electron bunches, the coupling of such hot collisionless electrons to the bulk plasma, given the lack of collisionality, must be indirect. There are two key mechanisms [158]. Firstly, via a cooler resistive return current of electrons that neutralises the current of the injected hot electrons that escape the potential well of the front surface. Since all hot electrons travel at approximately speed c , the magnitude of the return current depends not on the total energy absorbed but instead on the total number of electrons injected, as given by Equation 2.27. The current depends thus linearly on laser spot area and the electric field magnitude and, unexpectedly, not on the plasma density⁴. Secondly, dissipation of energy occurs via the formation of large amplitude bulk plasma waves induced in the wake of the hot electron bunches. Sherlock *et al* [158] calculate the magnitude of the induced wakefield to be

$$E_W = \frac{eN_e c}{\omega_p \epsilon_0} = \sigma \sqrt{\frac{m_e \epsilon_0}{n_e}} E_L, \quad (2.59)$$

where here the bunch velocity has been set to c , bulk electrons will be accelerated by E_W and their kinetic energy converted to heat via collisions. Interestingly, this reproduces the mid-temperature electron scaling with density that was observed by Chrisman *et al* [159] in their study of hot electron energy coupling in cone-guided fast

⁴Note that for a sufficiently thin target, the return current induces an electrostatic field on the back surface of the target which can then reflect hot electron bunches and decelerate them to the point of a return to collisionality. This is a reality for the PIC simulations explored in this thesis, however, since realistic targets are much thicker this shall be neglected.

ignition of inertial fusion targets. This is a different possible explanation to their self-declared ‘hand waving argument’. Excluding this study, such formulations for heat transfer to the plasma bulk within the ZVP regime remain untested in simulations.

Note also that as the laser pulse intensity rises, the fraction of energy absorbed by the ion species via hole boring increases. Savin [126] determined for $S = 1/2$, $a_0 = 100$, that this would be almost 20%.

2.4.2 Unpacking the QED effects of Figure 2.12

In Savin’s acclaimed paper [133], they determined theoretically and demonstrated in simulation that at $a_0 = 300$, $n_e = 50n_c$, there is a transition from standard ZVP scalings to an enhanced QED scaling associated with Breit-Wheeler (BW) electrons increasing the pseudocapacitor plate charge. Explicitly,

$$T \sim \frac{a_0^5}{\bar{n}_e}. \quad (2.60)$$

At first glance of Figure 2.12, Savin’s results are inconsistent with this parameter scan. Perhaps the measurement method can explain this via the well-known effect of radiation trapping from RR [160], also observed in these PIC simulations. After acceleration across the pseudocapacitor, the electron bunch encounters the subsequent laser peak. If the electron bunch gamma factor and laser intensity are both large enough, electrons radiate a significant fraction of their energy and are thus stopped in their tracks. Unable now escape the potential well at the plasma surface they are trapped and are not observed to escape the plasma until the laser pulse intensity reduces. Such an effect would not impact Savin’s scalings but would of course inhibit the observation of the scaling for electrons to the rear of the plasma block. Note that should this be the case and the collisionless electron bunches remain within the plasma bulk, Savin’s ZVP QED model truly applies directly to energy absorption by the plasma bulk.

There is another interpretation. Returning now to Figure 2.12, there are two interesting aspects. Firstly, there is a sudden jump in total energy at $a_0 \approx 300$. This cannot be explained by ZVP theory nor QED theory since the jump is observed in

the absence of Smilei's QED modules. Secondly, there is an even stronger jump in total electron bunch energy above $a_0 = 1000$. Decomposing the total energy into bulk electrons and those produced via the BW process, this is clearly a QED effect. Energy in BW produced electrons scales at a staggering

$$U_{\text{BW}} \sim a_0^{11.3 \pm 0.8}, \quad (2.61)$$

while the energy of bulk plasma electrons decreases. Perhaps this is a signal of Savin's QED ZVP electron bunches only at a higher energy due to the substantially greater plasma density of these simulations. The reduction in bulk electron energy can be attributed to an oversaturation of the front surface with BW electrons.

Combining equations 1.54 and 2.28, and assuming the electron radiates all its energy to the photon, for the ZVP mechanism at the point of emission one finds

$$\chi_\gamma = \frac{\sqrt{2}|\mathbf{E}|}{E_S} \frac{a_0^2}{\bar{n}_e}. \quad (2.62)$$

The probability of BW pair production begins to rapidly increase around $\chi = 1$, therefore, the transition to QED will occur at

$$a_0 \approx \left(\frac{E_S}{\sqrt{2}} \frac{e\bar{n}_e}{m_e c \omega_L} \right)^{1/3} = \left(\frac{a_s \bar{n}_e}{\sqrt{2}} \right)^{1/3}, \quad (2.63)$$

where $a_s = 7.73 \times 10^5$ is the normalised vector potential associated with the Schwinger Field. Repeating the calculation instead as a function of S , one finds instead

$$a_0 \approx \left(\frac{a_s S}{\sqrt{2}} \right)^{1/2}. \quad (2.64)$$

This corresponds to a transition to rapid pair production with Savin's parameters of $a_0 \approx 301$ versus $a_0 \approx 739$, consistent with both studies. This is interesting, ZVP with Savin parameters predicts a significantly lower transition to QED than could be expected from the Wilks scaling for $\mathbf{J} \times \mathbf{B}$ heating and could be tested at ELI-np.

Following Savin's QED ZVP theory, one can predict

$$U_{\text{QED}} \sim \frac{a_0^7}{S}, \quad (2.65)$$

therefore much work remains to understand the scaling of Figure 2.12 and to unify these results, starting with the application of the methods of this analysis to the parameter space explored by Savin. Undoubtedly, the advent of next-generation exa-watt scale lasers will be exceedingly interesting if such scalings in bunch energy can be maintained.

Linear Breit-Wheeler can safely be neglected in these simulations. There is simply not enough energy in the system. For two interacting photons of energy E_1 , E_2 , by consideration of four-momenta, the threshold condition for pair production is

$$E_1 E_2 \geq (m_e c^2)^2. \quad (2.66)$$

For a near-infrared laser photon of energy $\hbar\omega_L$, the interacting photon must have an energy in excess of 200 GeV. Despite the extreme acceleration gradients considered in this thesis, the ZVP mechanism predicts photons of energies no greater than 10 GeV and thus linear Breit-Wheeler is suppressed.

He *et al* [161] identified an alternative mechanism for linear BW using solid density targets, where forwards- and back-scattered high energy radiation reaction-produced photons interact within a hollowed-out plasma channel. In the geometry of interest in this thesis, while photons are produced in both directions, their production is localised to the plasma surface and at no points do their paths cross preventing the occurrence of this mechanism.

2.4.3 Errors

A set of simulations was undertaken to explore the stability of some of the PIC simulation assumptions. The effect of including collisions, increasing the temperature to resolve the Debye length and particle merging were explored. The results are presented in Figure 2.13. In general, including collisions had little to no effect on the results and changing the Debye length had a small but not systematic impact on the results. Note that while numerical heating is not directly dependent on laser intensity, increases to a_0 would increase the electron bunch density. Thus, resolving the Debye length of the ultra-high density (orders of magnitude greater

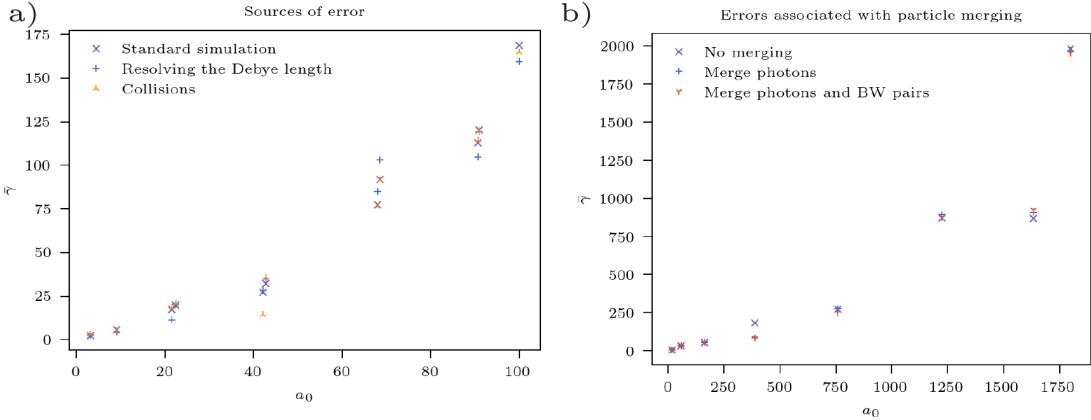


Figure 2.13: An exploration of ZVP 2D PIC simulation stability and assumptions. a) Electron bunch mean energies extracted from simulations with a_0 , $\bar{n}_e = 100$. Here, the x -axis is the normalised vector potential of the laser pulse cycle that made them, this is why there appears to be results stacked. b) Electron bunch mean energies extracted from simulations with a_0 , $\bar{n}_e = 1800$ both with and without particle merging.

than solid density) electron bunches produced by the higher laser pulse intensities considered here is doubtful, although the high energy of such particles may mitigate this issue. Particle merging was essential for simulations with $a_0 > 1800$ due to the proliferation of high energy photons and electron-positron pairs. At least at $a_0 = 1800$, it would appear particle merging had negligible impact, however, it is likely the error would increase with a_0 .

2.5 Conclusions

The Zero Vector Potential mechanism describes the post-ponderomotive rapid absorption of ultra-relativistic laser energy by a solid density overdense, collisionless and fully ionised plasma on the timescales of ion immobility. The defining characteristics of the mechanism have been identified in 3D PIC simulations including the observation of a zero of the vector potential propagating at speed $\approx 1.4c$ through a high-density ZVP electron bunch at the front surface of the plasma early in the ablative journey of the electron bunch. Simulations have suggested that from currently operational 10 PW short pulse laser facilities and foam targets, the ZVP mechanism can produce a train of attosecond duration, nano-Coulomb

electron bunches, each with a transverse emittance of a few nm rad. Such charge and quality are comparable to state-of-the-art electron bunch accelerators but on paradigm-shifting timescales. Such timescales being those on which atomic processes occur, these electron bunches could be manipulated to literally ‘shed light’ onto fundamental biological and chemical processes. Via a massive 2D PIC parameter scan the energies of such mass-limited electron bunches have been compared to those predicted by the ZVP model, identifying a range of validity for the model, specifically $a_0 > 10$, $S > 1$. These simulations were also used to confirm the energy absorption scaling in 2D up to and into the QED regime, posing new ideas and questions for the theoretical understanding of ZVP QED. Some discussion was given on the modes of energy absorption and on simulations exploring various of the assumptions in the model.

Chapter 4 details the upcoming GEMINI PW experiment to observe ZVP electron bunches for the first time. On the theme of experimentation, the following Chapter switches gears from absorption to reflection for the discussion of recent results on the ORION laser facility.

3

Attosecond X-ray harmonics on the ORION laser facility

Contents

3.1	Introduction	74
3.2	Theory	75
3.2.1	The ROM model	75
3.2.2	The normalisation factor	80
3.2.3	Hole boring	81
3.2.4	Harmonic source size	85
3.2.5	The SHHG optimal parameter space	86
3.3	Simulations	88
3.3.1	1D PIC simulations of SHHG	88
3.3.2	Hydrodynamic simulations of preplasma formation	93
3.4	The experiment	99
3.4.1	Target chamber geometry and polarisation	99
3.4.2	Targets	102
3.4.3	Contrast and plasma mirrors	103
3.5	Experimental data processing	104
3.5.1	Image plate calibration	104
3.5.2	OHREX calibration	106
3.5.3	Extracting the data	107
3.5.4	KBRXM	110
3.6	Experimental results	111
3.7	Summary and discussion	115

3.1 Introduction

The simulations of Chapter 2 are a relatively efficient and cost-effective mode of study. In comparison, high-power laser experiments are a cumbersome and technical challenge. However, the proliferation of errors in simulation codes leads to their inevitable deviation from reality, while reality itself diverges from the idealised conditions of the codes. For now, at least, experiments remain an essential component of the scientific method. This chapter reports on the March 2023 experiment at the ORION laser facility, AWE, Aldermaston, the UK’s most powerful laser system. The pulse duration is too long to create the attosecond electron bunches of the previous chapter. Instead, this chapter focuses on the ROM model of SHHG. This model describes the properties of radiation reflected from a relativistic laser pulse interaction with a solid-density plasma, another related route to bright attosecond light sources.

A fundamentally different mechanism to its Nobel prize-winning counterpart, SHHG from solids has demonstrated significantly higher conversion efficiencies and, relying on plasma oscillations not ionisation, does not suffer from the same limits to applied intensity [162]. SHHG has thus become a field of considerable interest for the production of bright coherent attosecond harmonics. First observed from the interaction of a CO₂ laser-solid interaction in 1981 [163] and followed shortly by a theoretical justification [164], interest in SHHG was revived by the arrival of CPA technology enabling access to unprecedented laser pulse intensities. To describe this new relativistic interaction, 1994 saw the arrival of a new theory, the Oscillating Mirror Model (OMM) [165]. Harmonic of the incident laser pulse were seen to be formed by the Doppler effect induced by a thin reflecting charge sheet at the plasma surface oscillating periodically with the laser pulse. This theory predicts a cutoff harmonic number of $n_c = 4\gamma_s^2$, where γ_s is the maximum gamma factor of the charge sheet. This relatively low cutoff is at odds with experimental results, the current record standing at the 3200thharmonic observed on the Vulcan PW laser system

in 2007 [166]. It was only after the development of relativistic similarity theory [81], which laid the groundwork for Baeva’s ROM model, that the experimentally observed cutoff could now be explained [131]. The similarity between the ORION and Vulcan parameter spaces indicates the most appropriate description of SHHG is the ROM model. It is described in more detail in Section 3.2.

There are many fascinating applications for SHHG beyond that of attosecond physics. The high frequency photon content of the harmonic beam has excellent penetration into dense materials with preliminary studies indicating notable improvement in energy absorption, suitable for auxiliary heating of ICF targets [167] or warm dense matter studies. This high energy content also enables the focusing of the harmonic beam to significantly smaller spot sizes than the incident diffraction-limited laser pulse. An all-optical coherent focusing of high harmonics with realistic laser parameters has been shown to produce over 1000 times intensity gain [168], thus providing a realistic route to the Schwinger limit [169]. Such a beam could have potential for enhancing the signal of photon-photon scattering [170] or of gravitational waves from twisted light [79]. Simulations have shown how a harmonic beam can probe SF-QED if a solid target is placed at the new focal position [86].

This chapter reports on the March 2023 experimental campaign at ORION to observe and measure the absolute intensity of X-ray High Harmonic Generation (XHHG) via the ROM mechanism. Section 3.2 covers the theory of the ROM model of SHHG from relativistic laser solid interactions. Section 3.3 presents simulation results that guided both experimental design and data analysis. The experimental design is covered in Section 3.4, in Section 3.5 the data analysis. Results are presented in Section 3.6 that are then summarised in Section 3.7.

3.2 Theory

3.2.1 The ROM model

For the moderately relativistic sub-picosecond ORION SP1 and SP2 beamlines, the most appropriate description for their non-linear interaction with a solid density target is the Relativistic Oscillating Mirror (ROM) model. The full technical

formulation constitutes the main body of Baeva's 2008 thesis [80], a notably more rigorous derivation than the original paper on the same topic [131]. This is achieved via application of the highly generalised Apparent Reflection Point (ARP) formalism to the case of $S > 1$ in relativistic similarity theory, showing that the ARP coincides with the relativistic critical density surface. This section provides a more qualitative, physically motivated description of this theory, the salient point of Baeva's thesis being that, while the surface motion is generally complex and parameter space dependent, at the short moment in time when most high harmonics are generated, the surface motion is always parabolic. By application of the Bourdier method, detailed in Appendix A.4 [171], Baeva demonstrated that the envelope of the spectral content of the reflected beam does not deviate from the normal incidence linearly polarised case for either s- or p-polarised incidence. Hence, the following discussion is restricted to normal incidence. It begins with relativistic similarity theory and Equation 1.41, $\mathbf{p} \sim a_0$. As discussed in Section 2.2, microscopically electrons follow similar relativistic trajectories. However, from Equation 2.5, v_{prop} , the velocity of electrons along the propagation axis of the laser pulse, is not necessarily relativistic except at the passing of the zero of the vector potential where $v_{\text{prop}} \rightarrow c$. Macroscopically, the well-defined critical density surface within the skin depth (located at $S = 1$), has a gamma factor that tracks the electrons' velocity along the propagation direction (x -direction) at that point,

$$\gamma_s = \frac{1}{\sqrt{1 - v_{\text{prop}}^2/c^2}} = \sqrt{\frac{1 + a(t, x)^2 + p_{\text{prop}}^2/(m_e c)^2}{1 + a(t, x)^2}}. \quad (3.1)$$

Equation 3.1 indicates surface gamma factors of order unity except at zeroes in the vector potential where $\gamma_s \sim a_0$. In the vicinity of the zero, $a \sim a_0 \sin(\omega_L t) \approx a_0 \omega_L t$. Thus, while the velocity of the surface is a smoothly varying function around the maximum, $v_{s,\max} \sim 1 - \mathcal{O}(a_0^{-2})$, the *gamma factor spikes* as the vector potential passes through zero. At this point, the surface emits high-frequency photons. The surface radiates such photons only in the vicinity of the gamma spike. Thus, the pulse width can be approximated. In ZVP theory, zeroes overtake the emitted photons. Radiation from the extended electron bunch that is emitted first arrives

at the observer last. In the ROM regime, Baeva demonstrates zeroes move at speed c . Consequently, all points in the skin depth radiate coherently. Since the radiation overtakes the sub-light speed surface, photons emitted from the surface first are observed first. The radiation emitted therefore has a duration of

$$\Delta l \approx (c - |\mathbf{v}_{s,\max}|) \Delta t \approx \frac{\Delta t}{\gamma_{s,\max}^2}, \quad (3.2)$$

where Δt is the temporal duration of the gamma spike.

Writing the surface velocity as a smoothly varying function around its maximum,

$$\mathbf{v}_s(t) = -(v_{s,\max} - c\alpha(\omega_L t)^2) \hat{\mathbf{x}}. \quad (3.3)$$

Without loss of generality, the peak of the gamma spike is set to $t = 0$ for cleaner notation. Around the gamma spike, the surface gamma factor is thus

$$\gamma_s(t) \approx \frac{1}{\sqrt{1 - (\mathbf{v}_{s,\max}/c)^2 - 2\alpha(\omega_L t)^2}}. \quad (3.4)$$

Since $1 - (\mathbf{v}_{s,\max}/c)^2 = 1/\gamma_{s,\max}^2$, the duration of the gamma spike scales as

$$\Delta t \sim \frac{1}{\sqrt{\alpha}\omega_L \gamma_{s,\max}}. \quad (3.5)$$

Hence, $\Delta t \sim a_0^{-1}$ and $\Delta l \sim 1/a_0^{-3}$ and the cutoff frequency corresponding to the highest frequency component of the pulse is

$$\frac{\omega_c}{\omega_L} \sim \sqrt{\alpha}\gamma_{s,\max}^3 \sim a_0^3. \quad (3.6)$$

Equation 3.6 reproduces the cutoff of the full derivation. Within the ROM model, this sharp pulse of radiation is masked by the low-order harmonics. Filtration is necessary to realise the attosecond duration of the high harmonic pulse.

The harmonic spectrum envelope for photon energies below the cutoff can be calculated from examination of the fields at the surface. Consider the Poynting vector

$$\mathbf{S} = \frac{1}{\mu_0} \mathbf{E} \times \mathbf{B}. \quad (3.7)$$

Regardless of the complex internal dynamics of the skin layer, an external observer will identify based on the fields observed in vacuum that the point of reflection

(the apparent reflection point, x_s) occurs where $\hat{\mathbf{n}} \cdot \mathbf{S}(x_s, t(x_s)) = 0$, here $\hat{\mathbf{n}}$ is normal to the plasma surface. This point must exist since the plasma is totally reflecting in the ROM regime. Correspondingly,

$$\hat{\mathbf{n}} \times \mathbf{E}(x_s, t(x_s)) = 0. \quad (3.8)$$

This represents a balance of the electric fields at the ARP. Decomposing into the incoming and reflected waves at the boundary,

$$E_i(x_s(t) - ct) = -E_r(x_s(t) + ct) \quad (3.9)$$

for all t . Applying the approximation of a plane wave, the phase structure at the reflection point is maintained as the reflected wave propagates away from the interaction region to the observation point ($E_r(x_s(t) - ct) = E_r(x' - ct')$ for an observation point (x', t') near an electric field peak.). Thus, determining the modulation of the reflected beam is akin to solving Equation 3.9.

Writing $\Phi = x_s(t) - ct$, $\Psi = x_s(t) + ct$ and taking the time derivative of Equation 3.9,

$$\frac{dE_r(\Psi)}{d\Psi} = \frac{c - \mathbf{v}_s(t)}{c + \mathbf{v}_s(t)} \frac{dE_i(\Phi)}{d\Phi} \quad (3.10)$$

where $v_s(t)$ is given in Equation 3.3. Integrating Equation 3.3, the reflected wave phase is

$$\Psi(t) = \Psi(0) + \frac{ct}{\gamma_{s,\max}^2} + \frac{1}{3}c\alpha\omega_L t^3. \quad (3.11)$$

Very close to the gamma spike peak the linear term dominates the interaction, corresponding to the highest order harmonics as previously discussed. The non-linear term dominates for

$$\delta\Psi = |\Psi(t) - \Psi(0)| \gg \left(\frac{3}{\alpha}\right)^{1/2} \frac{c}{\omega_L \gamma_{s,\max}^3}. \quad (3.12)$$

Note the correspondence between Equation 3.6 and the right-hand side of Equation 3.12. In this regime,

$$t = \left(\frac{3\delta\Psi}{c\alpha\omega_L}\right)^{1/3}. \quad (3.13)$$

At such intermediate times around the gamma spike, the linear term can be neglected, this is equivalent to writing $v_{s,\max} = c$. Thus,

$$\frac{dE_r(\Psi)}{d\Psi} = \left(\frac{2c - c\alpha(\omega_L t)^2}{c\alpha(\omega_L t)^2} \right) \frac{dE_i(\Phi)}{d\Phi}. \quad (3.14)$$

Assuming the incoming radiation is slowly varying around the short duration of the gamma spike, $dE_i(\Phi)/d\Phi \approx dE_i(\Phi)/d\Phi|_{\Phi=\Phi(0)}$ produces something easily integrated,

$$\frac{dE_r(\Psi)}{d(\delta\Psi)} = \frac{2}{\alpha^{1/3}} \left(\frac{c}{3\omega_L^2 \delta\Psi} \right)^{2/3} \frac{dE_i(\Phi)}{d\Phi}|_{\Phi=\Phi(0)}. \quad (3.15)$$

Hence,

$$E_r(\Psi) = -E_i(\Phi(0)) - \left(\frac{3c^2}{\alpha\omega_L^2} \right)^{1/3} \frac{dE_i(\Phi)}{d\Phi}|_{\Phi=\Phi(0)} \times (\delta\Psi)^{1/3}. \quad (3.16)$$

From simple phase modulation, the reflected radiation gains the quasi-singularity

$$E_r(x, t) = \text{const}_1 - \text{const}_2 \times (ct - x)^{1/3}. \quad (3.17)$$

Accordingly, the harmonic content near the gamma spike follows the envelope obtained by the Fourier transform,

$$|E(\omega)|^2 \sim \left| \int (ct - x)^{1/3} e^{-i\omega t} dt \right|^2 \sim \frac{1}{\omega^{8/3}}. \quad (3.18)$$

From the full derivation, Baeva finds that each gamma spike contributes a spectral intensity

$$I_n \sim \frac{1}{n^{8/3}} \text{Ai}^2 \left(\frac{n^{2/3}}{N} \right), \quad (3.19)$$

where $N = \alpha^{1/3} n_{\text{cr}}/2$, $n_{\text{cr}} = 2/(1 - v_s)$. For $a_0 \gg 1$, $n_{\text{cr}} \approx 2/(1 - (1 - 1/2\gamma_s^2)) = 4\gamma_s^2$.

This is the roll-off from OMM. The Airy-function is

$$\text{Ai}(x) = \frac{1}{\sqrt{\pi}} \int_0^\infty \cos \left(ux + \frac{1}{3} u^3 \right) du. \quad (3.20)$$

Equation 3.19 has two distinct regions. For $n < N^{3/2} = \sqrt{8\alpha}\gamma_s^3 = n_c$ and

$$I_n \sim n^{-8/3}. \quad (3.21)$$

Above the cutoff, the spectrum decays exponentially.

Note that Equation 3.19 only accounts for the contribution from a single gamma spike. In general, the slow envelope of the spectrum is unchanged [80] but there can be destructive interference between spikes leading to modulations in the spectrum for specific harmonics. This can cause certain orders to vanish such as is the case for even order harmonics for s-polarised incident light [172]. This results from symmetries in the equations of motion [173].

3.2.2 The normalisation factor

Baeva's theory provides the relative intensity of harmonics. However, for comparison with the absolute spectral intensity of harmonics in this experiment, the normalisation factor is required. This can be calculated from conservation of energy. For arbitrary harmonic order scaling, n^{-p} , the spectral intensity of the harmonic beam is

$$I_\omega(\omega) = \frac{dE_\omega(J)}{dAd\omega} = I_0 \sum_{n=1, \text{ odd}}^{n_C} n^{-p} S_n \left(\frac{\omega}{\omega_L} - n \right) \quad (3.22)$$

up to the cutoff, n_C . Here, $S_n(\omega/\omega_L - n)$ is the spectral shape function of the n^{th} harmonic in reciprocal space and I_0 is the normalisation factor of interest.

From conservation of energy,

$$\int I_\omega(\omega) d\omega dA = ER, \quad (3.23)$$

where E the total energy of the laser pulse and R is the reflectivity of the RPM. Intuition suggests the spectral shape function for the n^{th} harmonic retains the spectral shape of the incident laser pulse, *i.e.* for a laser pulse with a Gaussian temporal profile, this corresponds to a Gaussian centred at $n\omega_L$ of width $\sigma_L = 1/t_L$, where t_L is the laser pulse width. Simulations show this is a reasonable approximation for ORION parameters ¹.

Simulations also show that for the SP1 laser, the incident laser pulse significantly suppresses the even harmonics, hence,

$$ER = \int_0^\infty I_0 \sum_{n=1, \text{ odd}}^{n_C} n^{-p} e^{-(\omega/\omega_L - n)^2/\sigma_L^2} d\omega. \quad (3.24)$$

¹Hole boring discussed in the following section can lead to a Doppler shift in harmonic energy across the laser pulse corresponding to a Doppler broadening of the spectral shape function in reciprocal space, however, unlike the ZVP simulations of the previous chapter, this is negligible for ORION parameters.

The integral and summation order can be reversed. Since $\sigma_L \ll \omega_L$, all integrals in the summation are $\approx \int_{-\infty}^{\infty}$ and therefore,

$$ER \approx I_0 \sum_{m=0}^{n_C/2-1} (1+2m)^{-p} \sqrt{\pi} \sigma_L \omega_L, \quad (3.25)$$

thus,

$$ER \approx I_0 \sqrt{\pi} \sigma_L \omega_L ((1 - 2^{-p}) \zeta(p) - 2^{-p} \zeta(p, \frac{n_C + 1}{2})), \quad (3.26)$$

where $\zeta(p)$ and $\zeta(p, (n_C + 1)/2)$ are the Riemann Zeta and Hurwitz Zeta functions respectively. The final term can be neglected for a petawatt class laser pulse where $n_C \gg 1$. In the case of an ideal p -polarised laser pulse,

$$ER \approx I_0 \sqrt{\pi} \sigma_L \omega_L \zeta(p). \quad (3.27)$$

For the Relativistic Oscillating Mirror (ROM) regime, $p = 8/3$ and the RPM is extremely efficient, $R \approx 1$. Thus, I_0 can be estimated from the system parameters.

3.2.3 Hole boring

On long timescales relative to a laser pulse cycle, via the ponderomotive pressure of the laser, the plasma front moves inwards. This is hole boring [83]. The velocity of hole boring can be derived by consideration of conservation of momentum in this quasi-static state. Since the hole boring velocity is laser pulse intensity dependent, the spatial profile of the laser will be imprinted on the surface. Typically Gaussian in shape, for high-power laser systems, this can to first order generate a focusing RPM and a beaming of the specularly reflected signal, as in Figure 3.1. To access the highest possible electromagnetic field intensities, the laser pulse is focused on target to the diffraction limit. However, since the diffraction limit scales linearly with the wavelength, higher-order harmonics can be refocused to a smaller spot via this mechanism allowing access to unprecedented peak intensities as previously noted. Regardless of any blue-skied pursuits, it is evident that any prediction of SHHG beam intensity must account for hole boring.

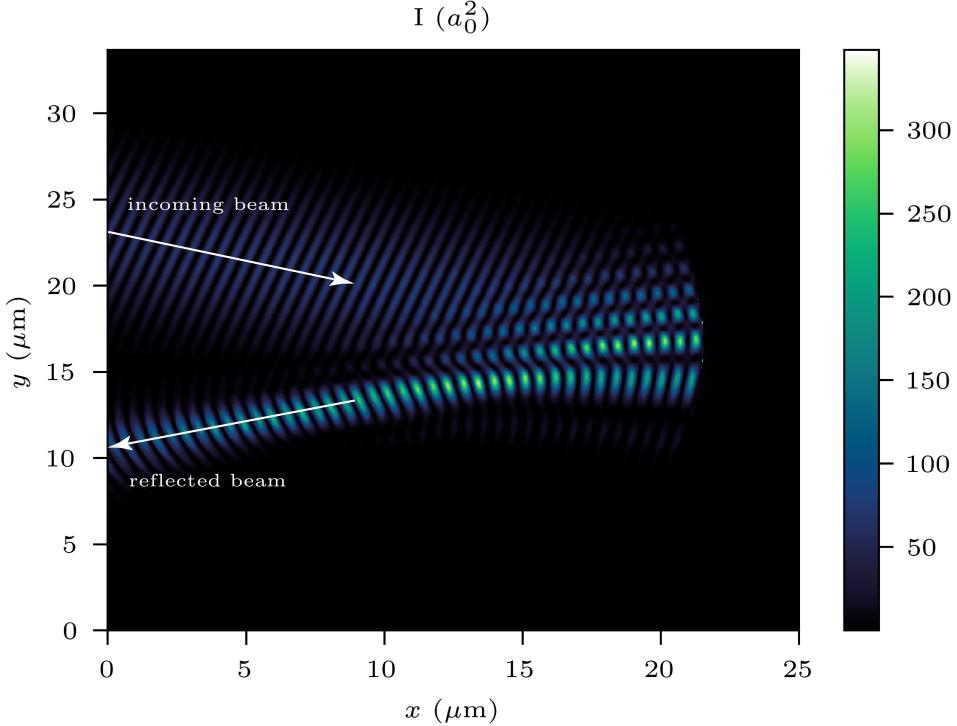


Figure 3.1: Electromagnetic field intensity in a 2D PIC simulation of a relativistic ($a_0 = 30$) laser pulse incident on a solid density plasma. The incoming beam is specularly reflected off the target which is curved by the radiation pressure leading to beaming in the reflected harmonic beam.

Applying momentum balance between the laser pulse and particles in the rest frame of the RPM surface, the hole boring velocity is

$$\frac{v_{\text{HB}}}{c} = \sqrt{\frac{R \cos \theta}{2} \frac{Z m_e}{A m_p} \frac{n_c}{n_e(x_i(t, y))}} a_L(t, y) = \Pi a_L(t, y), \quad (3.28)$$

where R is the RPM reflectivity, θ is the angle of incidence, 16° in the ORION experiment, Z and A are the atomic and atomic mass numbers respectively for the plasma ions, n_c is the plasma critical density, $n_e(x_i(t, y))$ the electron number density and $x_i(t, y)$ the depth of hole boring, from the Supplementary Material [129]. For the ORION laser pulse parameter space $\Pi \ll a_L \forall t$, *i.e.* relativistic electrons and non-relativistic ions. Hence, the relativistic correction derived by Robinson *et al* [174] to Equation 3.28 can be neglected. Due to the high contrast and long duration of the ORION beamlines, there is minimal preplasma formation on the front surface and therefore the number density is simply the number density of

the material in solid form and n_e is independent of $x_i(t, y)$. Robinson *et al* [175] generalised momentum conservation to multiple species. Replacing the mass density with the composite mass density $\rho = \sum_j m_{ij} n_{ij}$, then

$$\frac{An_e}{Z} \rightarrow \sum_j \frac{A_j n_{ej}}{Z_j}, \quad (3.29)$$

where n_{ej} is the number density of electrons that originated from the j ion.

The spatiotemporal envelope of the normalised vector potential of the laser pulse incident on the target surface can be modelled as

$$a_L(t, y) = a_0 e^{-\frac{y^2}{2w_L^2}} g(t - t_0) \quad (3.30)$$

where w_L is the beam waist on target and $g(t)$ the temporal envelope, typically a Gaussian or sech profile and t_0 the main pulse peak time.

Integrating Equation 3.28,

$$x_i(y) = \int v_{HB} dt = \Pi \int_{-\infty}^t a_L(t, y) c dt. \quad (3.31)$$

At the peak of the main pulse,

$$x_i(y) = \Pi a_0 c e^{-\frac{y^2}{2w_L^2}} G, \quad (3.32)$$

where $G = \int_{-\infty}^{t_0} g(t - t_0) dt \sim t_L$ and t_L is the laser pulse temporal width.

The total denting is a combination of the peak electron-ion charge separation, x_e (which leads to the intrinsic phase of the SHHG beam [176]) and Equation 3.31. Note that for the long pulse duration of the ORION laser, $x_i \gg x_e$ and therefore x_e can be neglected.

Applying a Taylor expansion to the spatial profile of Equation 3.31 around the laser spot centre,

$$x_i = \text{constant} - \frac{y^2}{4f_p} + \mathcal{O}(y^4), \quad (3.33)$$

where, to first order, this is the equation of a parabolic mirror with focal length

$$f_p(t) = \frac{w_L^2}{4\Pi a_0 c G}. \quad (3.34)$$

Following the Vincenti *et al* derivation [129], the denting parameter is defined as,

$$\delta_T = x_i|_{(y=0)} - x_i|_{(y=\sqrt{2}\omega_L)}. \quad (3.35)$$

Hence,

$$\delta_T = \frac{w_L^2}{2f_p} = 2\Pi a_0 c G, \quad (3.36)$$

and is independent of laser focal spot size.

If the spatial profile of the n^{th} harmonic beam can be adequately described by a Gaussian at the plasma mirror plane, with a beam width described by the harmonic source size, w_n ,

$$h_n \sim e^{-r^2/w_n^2}, \quad (3.37)$$

then the beam profile is known at all distances, z from the target. Its divergence, defined as

$$\theta_n = \lim_{z \rightarrow \infty} \frac{w_n(z)}{z}, \quad (3.38)$$

is therefore

$$\theta_n = \theta_n^0 \sqrt{1 + \Psi_n^2}, \quad (3.39)$$

where $\theta_n^0 = \lambda_n/\pi w_n$ is the harmonic divergence in the absence of RPM denting and

$$\Psi_n = \frac{2\pi}{\cos \theta} \left(\frac{w_n(0)}{w_L} \right)^2 \frac{\delta_T}{\lambda_n} \quad (3.40)$$

is the dimensionless focusing parameter. If $\Psi_n \gg 1$, as is true for the short wavelength X-ray harmonics of interest,

$$\theta_n \approx \frac{w_n(0)}{f_p \cos \theta} \quad (3.41)$$

and the divergence is dominated by RPM curvature.

Far from focus, at the detector plane a distance z from the target,

$$w_n \approx z \tan \theta_n. \quad (3.42)$$

The corresponding magnification factor at detection is thus

$$\gamma_n(z) = \frac{w_n(z)}{w_n(0)}, \quad (3.43)$$

and the laser intensity at detection is reduced by a factor $\gamma_n(z)^{-2}$.

At large distances,

$$\gamma_n \approx \frac{z \tan(w_n(0)/(f_p \cos \theta))}{w_n(0)} \quad (3.44)$$

Taking the Taylor expansion of the tangent, one sees that the magnification factor is only weakly dependent on the harmonic source size (f_p is independent of the source size), whereas the magnification is strongly dependent on the laser spot size ($\sim w_L^4$).

At the new RPM focal point, $z = z_f$, the demagnification factor is [129]

$$\gamma_n(z_f) = \frac{1}{\sqrt{1 + \Psi_n^2}}. \quad (3.45)$$

This determines the new peak intensity to be yielded by hole boring alone. A pre-curved target can achieve the same effect.

3.2.4 Harmonic source size

There is some harmonic source size shrinkage inherent to the interaction. Perhaps surprisingly, at photon energies below the harmonic cutoff, Vincenti *et al* demonstrated a weak dependence on harmonic order [129], finding $w_n \approx 0.5w_L$ from their 1D PIC simulation parameter scan. Approaching the cutoff, the harmonic source size shrinkage is enhanced by the roll-off. While it is unfeasible to perform a similar parameter scan to confirm this dependence, at least the magnification factor is only weakly dependent on w_n and hence this uncertainty does not pose a huge issue for calculation of the signal at OHREX, nor for calculation of the peak intensity at the new focus, since the wings of the spectrum do not contribute. Applying the theory to ORION beamline parameters and fitting a Gaussian to the new spot size, there is no additional shrinkage for the SP2 spot and at worst case for the SP1 laser, $w_{1000} = 0.21w_L$.

Incidentally, source size shrinkage reduces the error on the parabolic mirror approximation. The fractional error between the approximation and the true surface as a function of the distance from the centre of the mirror is

$$\text{err} = e^{y^2/w_L^2} \left(1 - \frac{y^2}{w_L^2} \right) - 1. \quad (3.46)$$

At $y = 0.5w_L$ is 4%, compared to 104% at $y = w_L$. This suggests minimal leakage of X-ray harmonics around the main beam.

3.2.5 The SHHG optimal parameter space

Interest in SHHG has prompted many numerical and experimental studies for the determination of its optimal parameters. The interaction efficiency increases with laser pulse intensity [132, 177] with a relativistic similarity parameter close to unity being preferable [127, 132]. The effective similarity parameter can be reduced with a finite preplasma scale length, as is hoped to be achieved in the GEMINI PW experiment (the ORION pulse duration produces too much hole boring over the long pulse duration to fashion a preplasma). The transverse coherence of the incident beam should be as good as possible to mitigate phase errors, this is best achieved with a diffraction-limited beam [178]. While SHHG in transmission from thin foils is the optimum for currently accessible laser pulse intensities, SHHG in reflection is more efficient for next-generation systems [135].

One might assume that target surface variation, root-mean-square roughness, Δs , need be less than the highest harmonic order that one wished to generate. However, Dromey *et al* demonstrated categorically in both experiment and simulation that surface roughness need only be less than the excursion amplitude of the oscillating electrons at the target front surface. Thus specular reflection is assured and scattering into the wings of the harmonic beam minimised [179]. This is generally satisfied by $\Delta s \ll \lambda_L$. In their experiment, harmonics were visible provided $\Delta s < \lambda_L/16$. Note that preplasma expansion will somewhat improve target smoothness before the main pulse arrival.

There is some debate regarding the angle of incidence with simulation parameter scans finding optima ranging from 45–62° [127, 132, 180]. However, all can agree that normal is not preferable. For ideal normal incidence odd harmonics are suppressed [172]. Even harmonics are suppressed for s-polarised incident light due to the symmetry of the interaction [71] while circularly polarised light significantly suppresses the entire mechanism [124]. This led to the suggestion of polarisation gating for the generation of isolated attosecond pulses [154].

Waveform tailoring with the inclusion of multiple laser wavelengths can improve SHHG efficiency [181]. Simulations have suggested the production of denser electron bunches and stronger acceleration for two colour pulses [182]. Improved efficiency has been confirmed in experiment [38]. Interestingly, the two colours must have identical polarisation, this is the inverse of two colour SHHG from gases.

High-power laser systems have inherent prepulses of ionising intensities. They will preheat a target leading to thermal expansion prior to the arrival of the main pulse, producing an exponential preplasma, $n_e \sim e^{x/L}$. SHHG is exceedingly sensitive to this preplasma formation, evoking particular interest in the field [168, 183–186]. While there is some variation in quoted optimal values, they are generally small relative to the laser wavelength, with scale lengths, $L \approx 0.1\lambda_L$. The difficulty in measuring such small lengths perhaps goes some of the way to explain the variety of suggested optima.

For $L \leq 0.02\lambda_L$, Coherent Wake Emission (CWE) dominates over SHHG. Kahaly *et al* demonstrated experimentally that harmonic emission varied smoothly from the CWE regime to the SHHG regime to SHHG suppression as the laser contrast was reduced [186]. Increasing the preplasma scale length increases the amplitude of surface oscillations, increasing SHHG production. However, if the scale length is too large, parametric instabilities in the underdense region begin to dominate and SHHG is suppressed [185]. The use of 1D hydrodynamic codes to measure prepulses from laser contrast trace measurements has become standard practice [183, 185, 187]. Although, this shall be called into question in the following chapter.

Fortunately, there are many approaches to control the various steps of preplasma generation. The heating effect of the prepulse can be reduced by reducing the angle of incidence or the prepulse itself can be reduced by reducing the intensity of the beam through apodisation or use of a frequency doubling crystal. As discussed in Chapter 1, a PM or chain of PMs are a crude tool to dramatically improve the laser contrast. A secondary beamline can generate a more precise controllable prepulse [186], or the surface can be recompressed using a circularly polarised prepulse that will cause minimal heating [187].

3.3 Simulations

3.3.1 1D PIC simulations of SHHG

Numerical simulations of the SP1 and SP2 interactions with CVD and PMMA were performed using the Smilei PIC code to confirm the analytical results of Section 3.2 for ORION beamline parameters. Application of the Bourdier method (plasma initialised streaming at $\mathbf{v} = c \sin \theta \hat{\mathbf{y}}$) enabled the computationally efficient simulation of the interactions in 1D, a necessity for the sub-ps ORION laser pulse duration. Even so, resolution, numerical heating and noise remain a serious problem for the accurate reconstruction of the physical phenomena despite the optimised supercomputer architecture. At least 10 cells per wavelength are necessary for the highest resolved harmonic [132]. Incidentally, for X-ray harmonics, this is on the order of the Debye length for solid-density plasmas ($T_e = 115$ eV, $n_e(\text{CVD}) = 1.05 \times 10^{30} \text{ m}^{-3}$, $\lambda_D = 7.8 \times 10^{-11} \text{ m}$, $E_{n,\max} = 1.6 \text{ keV}$). For the simulation of only a single laser pulse cycle, Edwards and Mikhailova required up to 5000 particles per cell [132] to resolve X-ray harmonics above error from noise and numerical heating. Unfortunately, numerical heating scales linearly with time further increasing the computational cost for ORION timescales [108].

Thankfully, to make comparisons to the derivations of Section 3.2 only a low resolution is necessary. Simulation parameters are detailed in Table 3.1. Parameters are given in real units in the laboratory frame and normalised Smilei units in the boosted Bourdier frame. Note that polarisation angles and normalised vector

SP1 Laser (1D, 16° angle of incidence)		
Parameters	Real	Smilei
Wavelength, λ (nm)	527	2π
Angular frequency, ω_L (fs $^{-1}$)	3.58	1
Polarisation, ϕ (°)	-42.2	-42.2
Normalised vector potential, a_0	7 - 10	7 - 10
Temporal FWHM, t_{FW} (fs)	470	1690
Temporal envelope, E_t	$E_t \sim e^{-(t-t_{FW})^2 \ln 2/t_{FW}^2}$	
SP2 Laser (1D, 16° angle of incidence)		
Parameters	Real	Smilei
Wavelength, λ (nm)	1053	2π
Angular frequency, ω_L (fs $^{-1}$)	1.79	1
Polarisation, ϕ (°)	19.6	19.6
Normalised vector potential, a_0	20 - 30	20 - 30
Temporal FWHM, t_{FW} (fs)	470	843
Temporal envelope, E_t	$E_t \sim e^{-(t-t_{FW})^2 \ln 2/t_{FW}^2}$	
Simulation box		
Parameters	Real	Smilei
Size, x (nm)	10λ	20π
Sim length, T_{sim} (fs)	$2t_{FW} + 5\lambda/c$	
Spatial resolution, Δx (nm)	$\lambda/128$	
Temporal resolution, Δt (as)	$\Delta x/2c$	
Macro-electrons per cell	1000	
Macro-ions per cell	1000	
Ion temperature, T_i (keV)	0	0
Electron temperature, T_e (keV)	0.115	2.25×10^{-4}
Collisionless, pre-ionised, regularly-initialised plasma		
Parameters	Real	Smilei
CVD density, n_e (m $^{-3}$)	1.05×10^{24}	$\begin{cases} 294 \text{ for SP1} \\ 1175 \text{ for SP2} \end{cases}$
PMMA density, n_e (m $^{-3}$)	3.90×10^{23}	$\begin{cases} 109 \text{ for SP1} \\ 435 \text{ for SP2} \end{cases}$
Electron x profile, $n(x)$	n_e for $\lambda \leq x \leq 9\lambda$	

Table 3.1: Smilei parameters to compare Section 3.2 analytical results to numerical simulation for ORION parameter space. Real parameters are taken in the laboratory frame with the given units, while Smilei parameters are in the boosted frame with normalised units. The CVD ion distribution consists of carbon ions with number density $n/6$ while the PMMA ion distribution consists of carbon, oxygen and hydrogen ions with number densities $1/6, 1/12, 1/12$ respectively, each with 1000 macroparticles per cell.

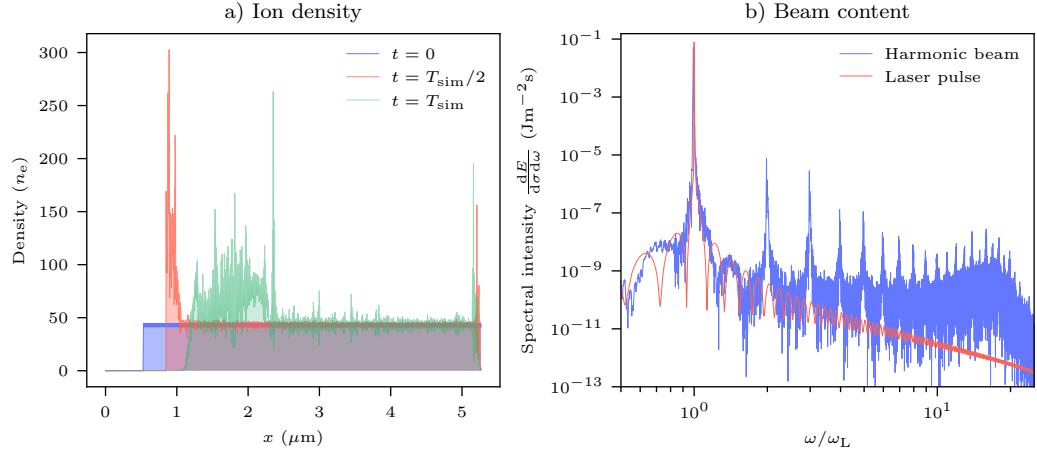


Figure 3.2: 1D PIC simulation of the SP1 ORION beamline geometry at $a_0 = 7$ interacting with a pre-ionised CVD target. a) Ion density at initialisation ($t = 0$), halfway ($t = T_{\text{sim}}/2$) and at the simulation end ($t = T_{\text{sim}}$) demonstrating hole boring. b) Absolute spectral intensity of the laser pulse and reflected harmonic beam obtained via Fourier transform. The harmonic beam retains the laser pulse spectral width. Even harmonics are significantly suppressed at this polarisation. The large bump in the distribution around the 18th harmonic is spuriously generated via numerical heating.

potentials are unchanged by the frame transformation. The fourth interpolation order for the particle shape function was found to be an efficient choice for the minimisation of numerical heating for such low-resolution non-Debye length-resolving simulations.

Figure 3.2 presents typical simulation results for the SP1 laser geometry, SP2 parameters produced similar results. Figure 3.2a depicts ion density profile evolution via hole boring. While the density increases to several times its initial level near the plasma surface, this need not be accounted for in the hole boring calculation. Additional ions accumulating near the surface have already acquired the necessary momentum and are simply co-propagating inwards with the surface. Figure 3.2b exhibits the harmonic content of the specularly reflected beam with harmonics retaining the spectral shape of the incident beam. Even harmonics are significantly suppressed relative to odd harmonics. The bump in the distribution around the 18th harmonic is entirely spurious, a consequence of numerical heating. Increasing the simulation resolution shifts the bump to the right. Allowing for 10 cells per maximum harmonic wavelength, this distribution can only be trusted up to the 12th

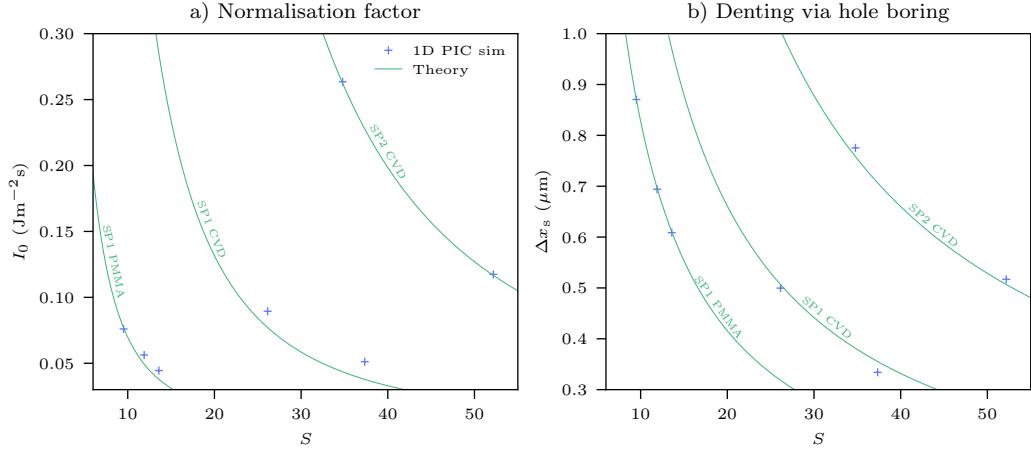


Figure 3.3: Comparison between simulation and analytical predictions of hole boring and SHHG normalisation factor in the ORION laser facility parameter space for CVD and PMMA targets. a) The normalisation factor, I_0 , where the spectral intensity, $I = I_0 n^{-p}$ for low order harmonics. b) The depth of hole boring compared to the initialised plasma surface. Note that the steep density gradient as visible in Figure 3.2a ensures the surface position is well-defined. Both parameters are expressed in the laboratory frame.

harmonic. It is here the signal begins to deviate. Undoubtedly, energy conservation does not hold for this simulation. However, it is still possible to extract I_0 . Making the reasonable assumption that Debye heating does not affect the SHHG process and that the spectrum is simply the sum of the two contributions (justified by the observation of the continuation of harmonics over the bump in the distribution), I_0 can simply be extracted as the peak intensity of the distribution, corresponding to the intensity at $\omega/\omega_L = n = 1$).

Figure 3.3 compares the analytically calculated parameters with those extracted from the simulations. There is remarkable agreement for the parameter space accessible with the ORION laser facility and the assumptions made in the analysis. The effect of numerical heating on the hole boring extent is illustrated by Figure 3.4 where the change in the surface position is plotted over time for the analytical calculation and PIC simulations with 2nd and 4th order shape function. The deviation from the theoretical prediction increases with time. Numerical heating leads to higher electron temperatures at the plasma surface and correspondingly a spurious preplasma expansion analogous to prepulse preplasma expansion. Artificially

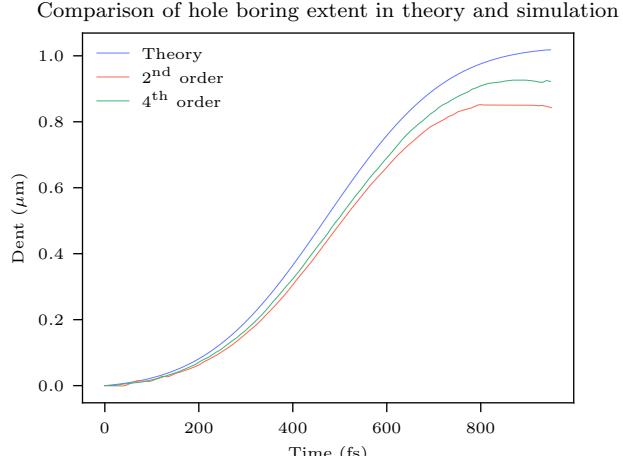


Figure 3.4: The effect of the PIC code particle shape function on numerical heating demonstrated via the extent of hole boring. The deviation from the theoretical prediction increases with time, and the error decreases for increasing order of the shape function.

increasing the plasma temperature to increase the Debye length and reduce numerical heating, while improving SHHG accuracy² reduces hole boring accuracy.

The X-ray harmonic spectrum produced from a CVD target irradiated by an SP1-type laser pulse is resolved in Figure 3.5. The simulation has 16384 cells per laser wavelength, the minimum multiple of 2 required to resolve the harmonics accessed with the OHREX quartz crystals. It was necessary to use a short test laser pulse with a $5\lambda_L$ FWHM. The simulation time before the peak of the laser pulse was increased to reduce the error in the spectrum from unphysical hard cutoffs to the laser pulse profile. A steep preplasma scale length was applied to the plasma distribution of $0.04\lambda_L$ to model the peak of the ORION pulse and suppress CWE.

At the X-ray intensities of Figure 3.5b, no harmonic structure is visible, indeed for the non-optimal ORION target chamber geometry, merging begins around the 20thharmonic. This must be accounted for in the calculation of the signal intensity. Note the s-polarised part of the reflected beam is over 50 times weaker than the p-polarised beam at the 1000th harmonic.

It would be interesting to extract the reflected beam temporal profile after filtering sub-X-ray harmonics. This can be achieved by applying the inverse

²Albeit up to a point, since changes to the preplasma scale length can also affect SHHG efficiency.

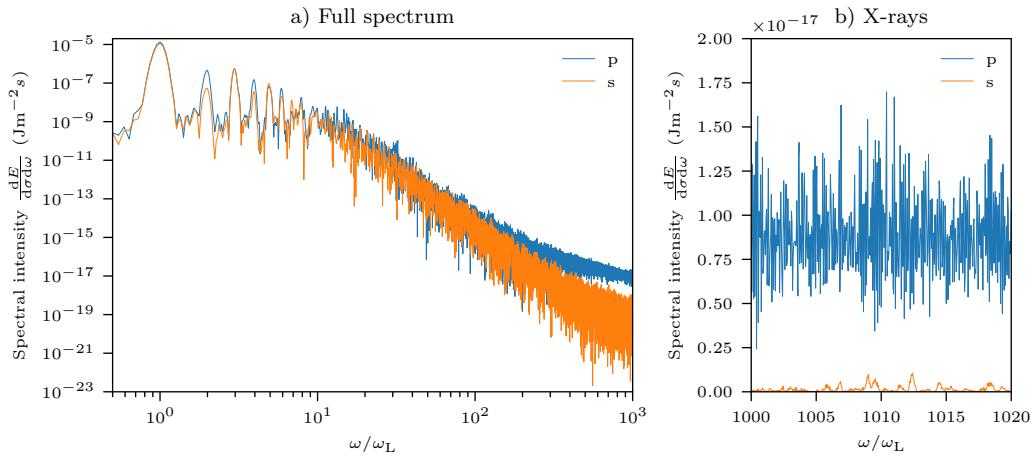


Figure 3.5: Reflected beam harmonic content up to the keV range in a high-resolution 1D PIC simulation of a test pulse with SP1 ORION beam geometry incident on a CVD target. The absolute spectral intensity is calculated from the Fourier transform of the reflected beam electric field. The s- and p-polarised components of the beam are analysed separately for both a) the full spectrum and b) the X-ray harmonics accessible with the OHREX quartz ($10\bar{1}1$) crystal.

Fourier transform to the Fourier-transformed temporal profile with the lower-order harmonics removed. Unfortunately, beyond $n = 50$, noise overwhelms the signal of interest (from ROM theory, the X-ray harmonic intensity is at least 8 orders of magnitude below the laser pulse intensity). A substantially larger simulation would be required to produce the filtered signal. Figure 3.6 compares the reflected pulse structure to the reflected pulse structure with harmonics below $n = 50$ removed. The filtered beam consists of a series of attosecond pulses of radiation, the brightest pulse is of duration 30.3 as. This is an order of magnitude above the duration of the X-ray pulse as predicted from the cutoff expression. Note that the intensity of the first few laser cycles is insufficient for the generation of harmonics above $n = 50$ and there is therefore no signal here, effectively reducing the pulse train length.

3.3.2 Hydrodynamic simulations of preplasma formation

Following Dollar *et al* [185], hydrodynamic simulations of the ORION laser systems were performed using HYADES to confirm the suitability of the contrast conditions.

Simulations were performed in 1D planar geometry with a mesh of 511 evenly spaced points covering 20 μm of the target (SiO₂, PMMA, CVD, etc). Equation of

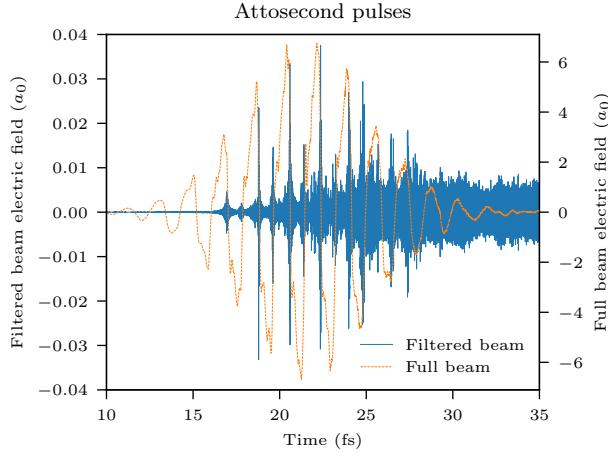


Figure 3.6: Attosecond pulse train from the filtered reflected laser pulse. The filtered beam contains only harmonics above $n = 50$.

state data was taken from the in-built HYADES data tables. For low temperatures and pressures, the data for the minimum values in the tables are used, for temperatures and pressures above the highest values recorded in the tables, the values are linearly extrapolated from this point. Ionisation was calculated using the local thermal equilibrium average atom model. Low temperature ($T < 0.15\text{eV}$) values for the absorption and refractive indices of the target materials were input [188]. The threshold for laser energy absorption is set to $1 \times 10^6 \text{ ergs cm}^{-2} = 0.1 \text{ W cm}^{-2}$. Electrons and ions were initialised at a temperature $1.551 \times 10^{-2} \text{ eV}$. The real polarisation of the interaction cannot be treated using a 1D code since the electric field vector is rotated during propagation [189]. Instead, simulations were performed in both s- and p-polarisation to provide a sense of the scale of the interaction.

As expected, no pre-ionisation or preplasma formation was observed in the simulations of the high-contrast SP1 laser pulse with any target material. This is in stark contrast with the SP2 laser. In the absence of a PM, the preplasma expansion of the targets is many orders larger than the laser pulse wavelength, necessitating the use of a PM and further HYADES simulations.

The distance between the PM and the main target is 15 mm, thus the beam diameter at the PM is $\approx 15/(f/3) = 5 \text{ mm}$. Hence, the intensity at the PM can

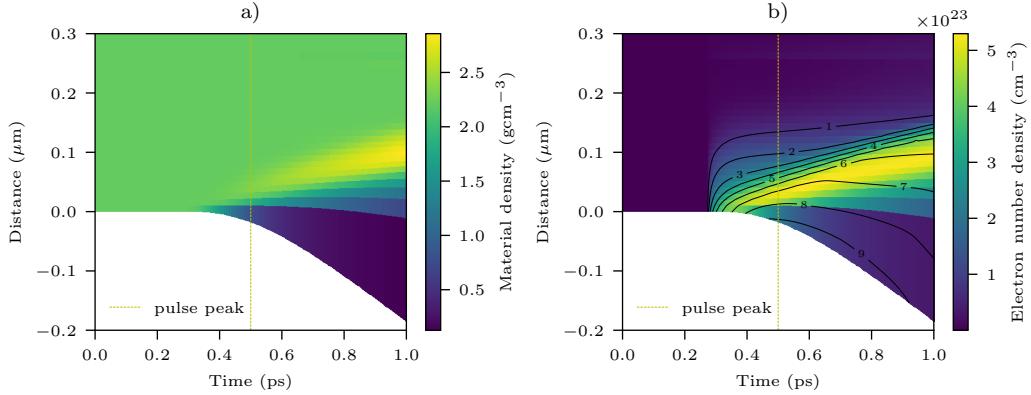


Figure 3.7: Silicon dioxide plasma mirror switch on from irradiation by the SP2 main pulse at 45° angle of incidence and p-polarisation. The laser pulse travels in the $+\hat{x}$ -direction. The main pulse peak intensity is marked at 500 ps in the simulation. a) Material density as a function of time, a short preplasma is formed alongside hole boring. b) Electron number density as a function of time. The contours map the ionisation level.

be calculated. To account for variation around this approximation, simulations were performed with plausible high and low intensities.

First, the interaction between the SP2 beamline and a SiO_2 PM was simulated. Note that it is not possible to model the AR coating in hydrodynamic codes. Instead, it was assumed that the thin coating has minimal impact on the interaction. No ionisation was observed from the high-intensity laser prepulse. Figure 3.7 plots the interaction between a low-intensity main pulse and the plasma mirror. Similar results were obtained for s- and p-polarisation. These simulations suggest ionisation begins at the start of the main pulse. There is some preplasma expansion and hole boring. The focusing effect of this hole boring is minimal for the large spot size on PM and can thus be ignored. The collisionless skin depth and plasma frequency can be calculated at each mesh point in the plasma from the electron density. The corresponding laser attenuation at each point is plotted in Figure 3.8 at the peak of the main pulse. The laser is rapidly attenuated and therefore switched on despite not attaining total ionisation at the front surface (at a cost to its reflectivity).

Satisfied that the PM operates as anticipated, a parameter scan of the targets was then conducted. Figure 3.9a describes the typical evolution of target density with the application of the SP2 prepulse. In this case, the prepulse has been

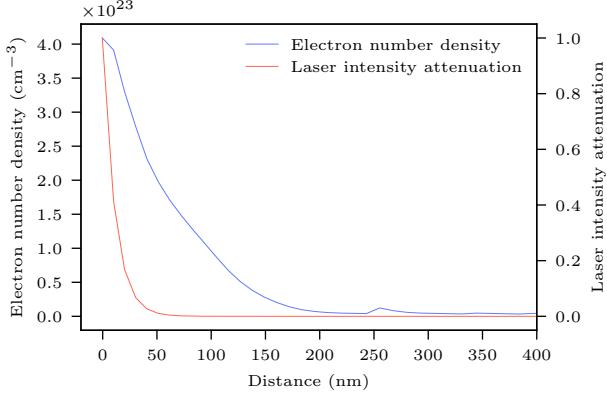


Figure 3.8: The attenuation of the SP2 laser pulse is calculated as it propagates through a switched-on PM.

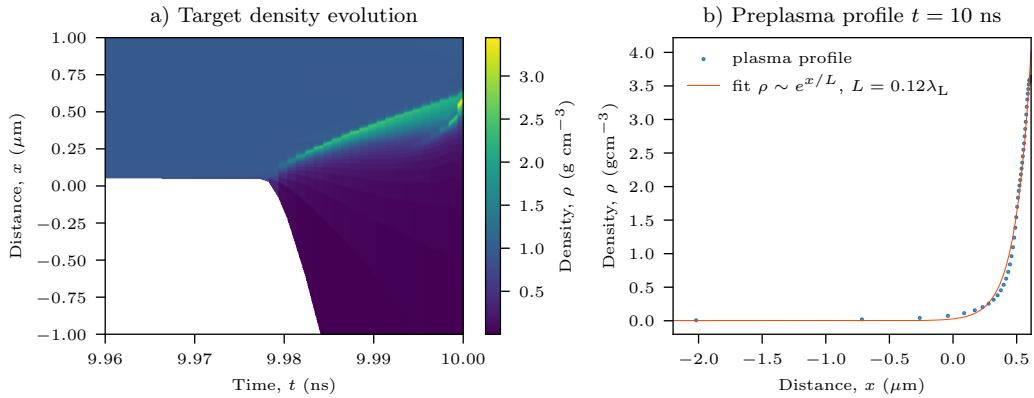


Figure 3.9: Typical preplasma formation from the p-polarised 16° angle of incidence of the ORION SP2 laser prepulse on an HDPE target. In this simulation the laser is modelled from 10 ns before the main pulse with $a_0 = 30$ and attenuated by a PM of reflectivity 0.004. a) Typical preplasma expansion elucidated via target density. By the time of the main pulse, hole boring is dominant over plasma expansion. b) An exponential fit ($\rho \sim e^{x/L}$) for the preplasma scale length, L , at the time of arrival of the main pulse. For this simulation the optimum fit is $L = 0.12\lambda_L$.

attenuated by a plasma mirror with a reflectivity of 0.004. By the time of the main pulse arrival in this simulation, hole boring dominates over preplasma expansion. Indeed, the laser pulse intensity has already reached relativistic intensities a couple of picoseconds out from the main interaction. Figure 3.9b includes an exponential fit to obtain the corresponding close to optimal preplasma scale length of $L = 0.12\lambda_L$ at the time of arrival of the SP2 main pulse.

Figure 3.10 repeats the analysis of 3.9 for a variety of target materials, reflectivities, polarisations and laser intensities. The s-polarised light simulations

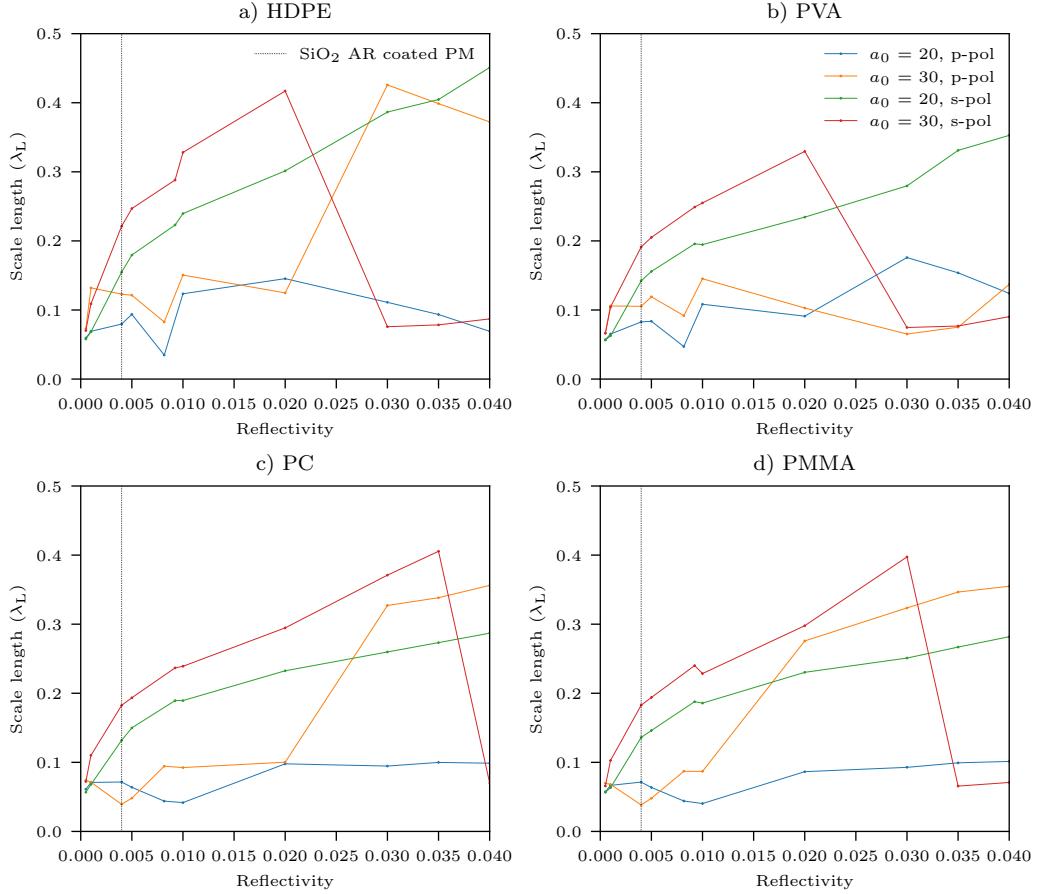


Figure 3.10: Preplasma scale length parameter scan for a variety of plastic targets. Target materials are a) HDPE, b) PVA, c) PC and d) PMMA. The dashed line highlights the reflectivity of the SiO_2 AR-coated PM for the SP2 wavelength. The preplasma scale length remains small for the range of parameters explored.

follow a clear trend: increasing incident laser energy leads to an increased scale length up to a transition point that will be determined by the point at which hole boring overcomes plasma expansion. This will depend on the interplay of target ion density, ionisation potential and heating. Interestingly, the trend is much less consistent for p-polarised light. It is known that heating increases for p-polarised light as there is now a component of the laser pulse electric field acting into the plasma, however, the crossing of the high and low intensities suggests a dynamic balance between hole boring and plasma heating. Regardless, the change in scale length is not significant at these PM reflectivities. Unfortunately, it was not possible to conduct a similar parameter scan for the CVD targets, it would

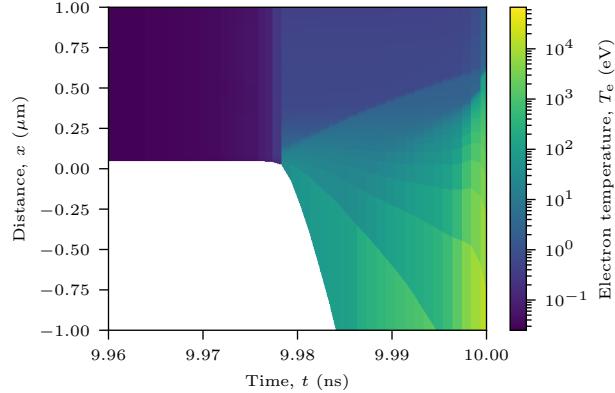


Figure 3.11: The typical electron temperature of a plastic target after irradiation by a petawatt class laser prepulse. This is the same simulation as presented in Figure 3.9. Namely, an $a_0 = 30$ p-polarised laser pulse attenuated by a plasma mirror of reflectivity 0.004 incident on an HDPE target at 16° . The bulk and preplasma temperatures differ by multiple orders of magnitude.

appear that the EOS tables provided for such a target material are not suited for this interaction type. However, one can assume that the preplasma scale lengths of irradiated CVD targets would be lower than those established in Figure 3.10 due to the high damage threshold of the material.

The question of plasma temperature, raised with regards to the initialisation of PIC code main pulse simulations as presented in Chapter 2, can now be redressed. Figure 3.11 plots the plasma electron temperature from 40 ps before the main pulse arrives. Once plasma expansion initiates, the extent of heating rapidly diverges across the preplasma and bulk. Evidently, it does not make sense to define a temperature for a system not in equilibrium. Indeed, there is no standardised practice within the field and yet temperature can have a significant effect, such as has already been remarked from simulations on hole boring. There is also some experimental evidence to say SHHG efficiency scales inversely with plasma temperature [186]. Plasma temperature is highly sensitive to the precise experimental conditions. At the time of the main pulse arrival at the plasma surface (as defined by the peak of the electron density distribution), the temperature is 112 eV. This is at least consistent with the value chosen for PIC simulations but should be treated with wariness.

Note hydrodynamic codes would be unsuitable for the prediction of the plasma temperature increases induced by the main pulse interaction given the high non-linearity of the dynamics in this regime that could not be adequately captured by such a code.

3.4 The experiment

The two experimental goals were to resolve and measure the absolute intensity of X-ray harmonics produced by the interaction of the ORION SP2 beamline with a range of flat solid targets. After debris from the plasma mirror optic damaged the parabola it was necessary to switch to the higher contrast but lower energy SP1 beamline. Only after the experiment was it established from simulations that it is not possible to resolve X-ray harmonics with the ORION target chamber geometry. However, absolute intensities were measured for both the SP1 and SP2 beamlines and for CVD and PMMA targets. ORION short pulse beamline parameters are provided in Figure 1.3.

The experimental design is relatively straightforward. CAD drawings of the experimental setup are presented in Figure 3.12. The laser was focused onto the target at Target Chamber Centre (TCC) with the ORION High Resolution X-ray (OHREX) spectrometer positioned along the specular reflection direction. Additional X-ray pinhole diagnostics monitored the spot size. The SP2 beamline reflects off an additional PM optic at 45° Angle of Incidence (AOI) before the main target to improve the laser prepulse contrast. Then both the SP1 and SP2 beamlines arrive at TCC along the same axis.

3.4.1 Target chamber geometry and polarisation

Both the non-linear SHHG interaction and the linear OHREX crystal spectrometer are sensitive to the polarisation of incident light. It is therefore important to track the polarisations of the beamlines as they propagate through the system.

The ORION target chamber has its own defined geometry with the target located at the origin (TCC), sketched in Figure 3.13. The polarisation angles of

3.4. The experiment

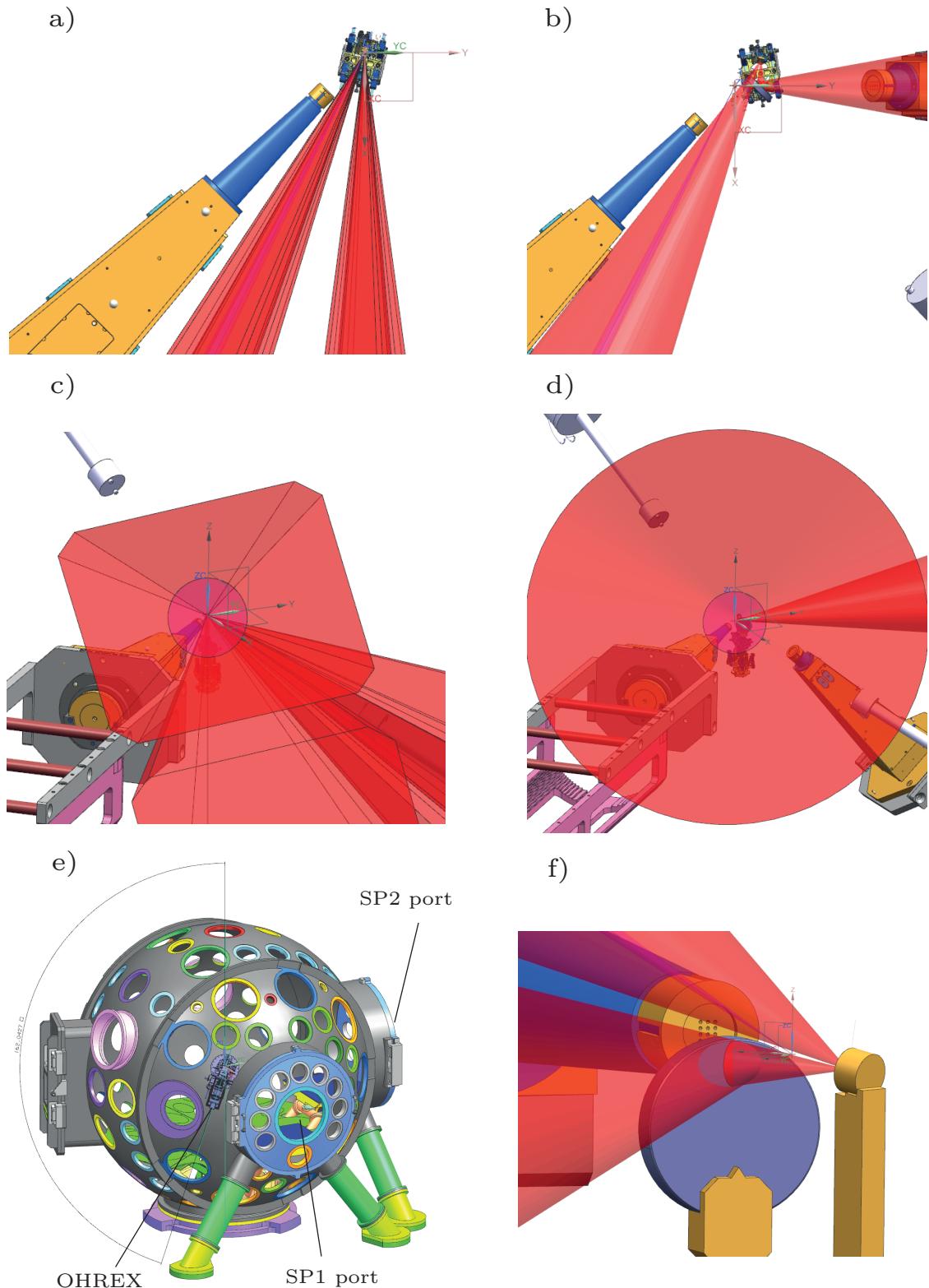


Figure 3.12: CAD images of the ORION target chamber set up for this experiment. a) and b) SP1 and SP2 beamlines respectively from parabola to TCC to OHREX spectrometer. c) and d) SP1 and SP2 beamlines respectively looking from OHREX to TCC. e) Annotated ORION target chamber model. f) SP2 beamline reflecting off plasma mirror before main target incidence.

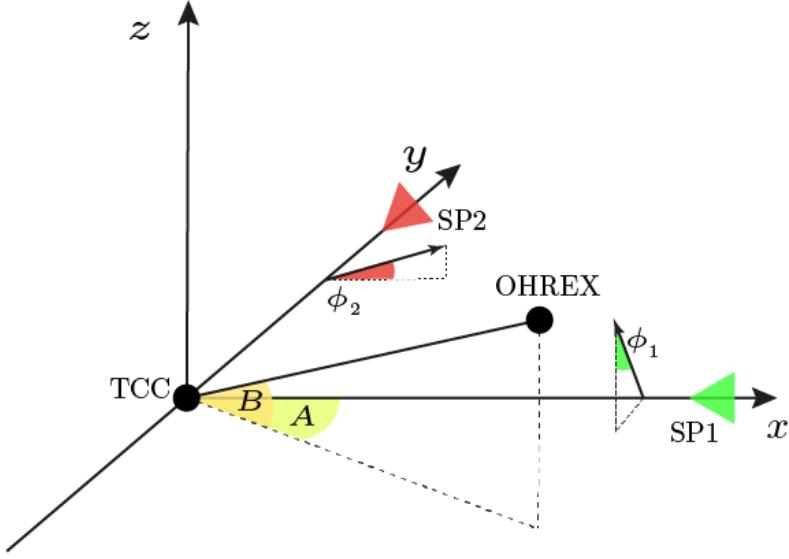


Figure 3.13: ORION target chamber geometry with the location of the target, OHREX spectrometer and the SP1 (green) and SP2 (infra-red) beamlines and their corresponding polarisations, $\phi_1 = 11.8^\circ$ and $\phi_2 = 16.4^\circ$.

the two beamlines are $\phi_1 = 11.8^\circ$ and $\phi_2 = 16.4^\circ$. Following the reflection of the SP2 beam off the plasma mirror, both the SP1 and SP2 beamlines propagate in the $-\hat{x}$ -direction towards the origin. The OHREX crystal is located at

$$\mathbf{r}_{\text{OHREX}} = r_0(\cos B \cos A, -\cos B \sin A, \sin B), \quad (3.47)$$

where $r_0 = 2.4\text{ m}$, $A = 26.82^\circ$ and $B = 18.15^\circ$, setting the rotation angle of the target. As in Figure 3.12e, the OHREX spectrometer sits on the outside of the target chamber tilted an angle 18° to the vertical.

The interaction plane is defined by the vector

$$\mathbf{n} = \frac{\mathbf{r}_{\text{OHREX}}}{r_0} \times \hat{\mathbf{x}} = (0, \sin B, \cos B \sin A). \quad (3.48)$$

The cosine rule can be applied to determine the polarisation of the laser pulses in the interaction plane, for polarisation vector $\hat{\mathbf{E}}$,

$$\frac{\mathbf{n}}{|\mathbf{n}|} \cdot \hat{\mathbf{E}} = \cos \theta, \quad (3.49)$$

where θ defines the angle between the polarisation vector and the vector normal to the interaction plane. This corresponds to angles out of the interaction plane of 42.2° for the SP1 beam (rotating anticlockwise out of the interaction plane when

looking from TCC to parabola) and 19.6° for the SP2 beam (rotating clockwise out of the interaction plane when looking from TCC to parabola). Again applying the cosine rule, the angle of incidence is 16° .

The polarisation vector can then be split into s- and p-polarised components. The s-polarised component remains unchanged by the interaction but the p-polarised component must be rotated, its direction along $\mathbf{n} \times \mathbf{r}_{\text{OHREX}}$. Figure 3.5 illustrates the suppression of the s-polarised component at the X-ray photon energies of interest. This component is therefore neglected.

The OHREX spectrometer sits flat on the target chamber wall but rotated an angle $C = 18^\circ$ to the vertical. The OHREX interaction plane is correspondingly rotated. Again, applying the cross-product, the OHREX interaction plane is

$$\mathbf{n}_O = [-\cos(A)\sin(B)\sin(C) - \sin(A)\cos(B)\cos(C), -\cos(A)\cos(B)\cos(C) + \sin(A)\sin(B)\sin(C), \cos(A)\sin(B)\cos(C)]$$
(3.50)

Applying the cosine rule between the p-polarised vector and \mathbf{n}_O , the angle of the polarisation vector of the harmonic beam out of the plane of the OHREX interaction is 77.23° for both SP1 and SP2.

3.4.2 Targets

Specialised target holders were designed and built by the AWE and CLF target fabrication teams. These hold the targets at the required angle out of the horizontal plane with an additional fiducial wire for alignment. Targets were mounted on the ORION Multi-Target-Mount and alignment was performed offline by Dr Ed Gumbrell.

There are multiple considerations when it comes to target density. In practice accessing the $S \approx 1$ regime is not possible using a petawatt class laser system with even the lowest density solid plastic targets. However, the 2007 Vulcan experiment successfully reproduced the $8/3$ scaling up to the X-ray regime for $S \approx 50$ [166] despite simulations repeatedly emphasising the improvement of harmonic efficiency for reduced similarity parameter. At the same time, as discussed in Section 3.2, target hole boring leads to an increased harmonic beam divergence. High electron

Figure 3.14: ORION experiment 3D scans of target roughness. a) CVD with roughness $\Delta s = 4\text{ nm}$ and $S \approx 26, 35$ for SP1 and SP2 beamlines respectively. b) PMMA with roughness $\Delta s = 11.2\text{ nm}$ and $S \approx 10, 13$ for SP1 and SP2 beamlines respectively.

density targets are perhaps therefore more suited to the petawatt class laser system thereby maintaining the beaming of harmonics.

The surface perturbation of a selection of low-density plastic targets was analysed by the CLF target fabrication team leading to the choice of PMMA plastic targets. CVD targets were also selected, having the highest ionisation potential (and therefore lowest susceptibility to laser prepulse) of solids that can be polished optically flat albeit at a higher S parameter. Target surface scans performed by AWE and CLF are given in Figure 3.14 [190].

3.4.3 Contrast and plasma mirrors

As illustrated in Figure 3.12, a plasma mirror optic is included before the main target to improve the contrast of the SP2 beamline [111]. HYADES simulations of the effect of this optic are detailed in Section 3.3.2. Provided the harmonic beam divergence is less than or equal to the $f/3$ cone of the laser pulse, as is anticipated, the harmonic beam should not be clipped by the PM in this setup.

The intention was to use AR-coated fused silica optimised for 45° AOI and the SP2 pulse wavelength (BCP45R) [191]. This has a reflectance of 0.398 % at $\lambda_L = 1053\text{ nm}$ for unpolarised light. Unfortunately, the debris from the PM caused damage to the parabola. For the second SP2 shot, the high reflectivity fused silica PM was replaced by a thin foil of silicon nitride. The reduction in available material of the thin foil lowers the risk of damaging debris. At 45° AOI, $\lambda = 1.053\mu\text{m}$ and $\phi_2 = 16.4^\circ$, the silicon nitride PM pre-switch-on has a reflectivity of 5.55 % [188]. It is safe to assume consistent behaviour after switch-on for the two PMs.

3.5 Experimental data processing

3.5.1 Image plate calibration

Image plates (IPs) are reusable recording media that detect ionising radiation and are particularly suitable for the detection of X-rays produced in laser-plasma interactions. Their response is well understood and their sensitivities to a wide spectrum of photon energies have been absolutely calibrated on the ORION facility [192], albeit for the FLA3000 scanner, not the FLA7000 used in this experiment. However, the deviation in response is negligible for the photon energies measured. In this experiment, the Fuji Biological Analysis System (BAS) TR-type IPs were used. They have a phosphor layer composed of $\text{BaFBr}_{0.085}\text{I}_{0.15}$ with density 2.61 g cm^{-3} and thickness $60 \mu\text{m}$ but no mylar layer. This makes them suitable for low-energy X-ray detection. When scanned, the IP releases blue photons via photostimulated luminescence (PSL), which are then collected by a photomultiplier tube. The PSL value is generalised across scanner types from the measured ‘Grey’ (G) value by

$$\text{PSL} = (0.23284G^2 \times 10^{-9}) \left(\frac{\Delta x}{100} \right)^2 W \times 10^{-L/2}, \quad (3.51)$$

where Δx is the scanner resolution ($= 25 \mu\text{m}$ in this experiment), L is the latitude parameter, and

$$W = 0.092906 + 1370.8e^{-0.014874V} + 654.24e^{-0.011026}, \quad (3.52)$$

where V is the scanner voltage [193].

IP photon sensitivity, ψ , the number of PSLs per incident photon, is dependent on photon energy. Meadowcroft *et al* modelled this as,

$$\psi_j = \eta(m_j h\nu + c_j), \quad (3.53)$$

where $h\nu$ is the photon energy and m_j and c_j are linear fit parameters valid for specific energy ranges, j . For the Fuji BAS TR-type IP and X-rays in the range 0-6.0 keV, $m_j = 0.54(5) \text{ mPSL keV}^{-1}$ and $c_j = 0.020(2) \text{ mPSL}$. The IP absorption efficiency in mPSL per photon is

$$\eta(h\nu, T_i, T_s) = \exp(-n_i \Phi_i(h\nu) T_i) [1 - \exp(-n_s \Phi_s(h\nu) T_s)], \quad (3.54)$$

where n is the layer density, $\Phi(h\nu)$ is the total cross-section of the layer, T the effective layer thickness, s and i correspond to the sensitive (phosphor) and insensitive (mylar) layers of the IP respectively [194]. The first term is neglected in the absence of an insensitive (mylar) layer in TR-type IP. Below 50 keV, the dominant mode for X-ray absorption into the IP is the photo-electric effect, where

$$\Phi_{\text{ph}} \approx 3 \times 10^{12} \frac{Z^4}{(h\nu)^{3.5}} \quad (3.55)$$

and Z is the atomic number [195] and Φ_{ph} is given in units of Barn per atom. At 2.4 keV, that corresponds to a sensitivity of 1.32 mPSL per incident photon.

It is generally unavoidable that some time will elapse between the laser shot and the IP scan. For this experiment, 30 minutes was typical, in which time some fading of the IP occurs that must be accounted for. IP fading can be modelled as an attenuation factor,

$$F(t) = A \exp(-t/\tau) + B, \quad (3.56)$$

where t is the time between shot and scan and A , τ and B are found from fits to experimental data. A key aspect of the exponential decay is that the attenuation depends only on the signal at that moment in time and not the initial conditions. This has been shown to be true in experiment [192].

At 20 °C at the ORION facility, Meadowcroft *et al* [192] determined that for the Fuji BAS TR-type IP, the optimum fit for the parameters of Equation 3.56 is $A = 0.347(22)$, $B = 0.693(11)$ and $\tau = 35.5(53)$ minutes. Therefore at 30 minutes, $F(t) = 0.84$.

In summary, the number of PSL measured on an IP can be converted to an incident number of photons via

$$N(h\nu) = \frac{\text{PSL}}{F(t)} \frac{10^3}{\psi(h\nu)} = P(h\nu)\text{PSL}. \quad (3.57)$$

3.5.2 OHREX calibration

The Orion High REsolution X-ray (OHREX) spectrometer, housed on the ORION laser target chamber outer wall, utilises a spherically bent crystal geometry to spatially focus and spectrally analyse photons from the target chamber [196] with a high signal-to-noise ratio. The measured signal has been absolutely calibrated for a range of X-ray photon energies using a variety of crystals [197]. The OHREX spectrometer can hold two crystals at a time. At each crystal's spatial focal plane, a two-dimensional image is formed, one dimension is spatial, the other spectral. The energy range accessed by a given crystal is determined by the crystal rotation but all OHREX crystals are designed for operation at a nominal central Bragg angle of $\theta_B = 51.3^\circ$ with the corresponding wavelength determined from Bragg's Law, $n\lambda = 2d \sin \theta_B$, for the appropriate crystal plane. The range around that central photon energy is determined by the crystal width in the spectral dimension.

MacDonald *et al* determined a quadratic fit for each crystal's dispersion relation to connect position along the crystal image to photon energy [197]. Unfortunately in this experiment, the image lengths varied from those in the previous experiment, a likely consequence of slight defocusing of the optic. The OHREX geometry is designed such that precise focus is not necessary to achieve good results and optimisation is not routinely performed [196].

Instead, a simple linear dispersion relation based on the known maximum and minimum energies accessed by the crystal was applied across the crystal images, a reasonable approximation to the dispersion relation determined by MacDonald *et al* [197] (the quadratic correction is small). The energy ranges for the three lowest energy OHREX crystals are given in Table 3.2.

Provided full illumination of the 6 cm \times 4 cm crystal, as can be assured for harmonic beams at 2.4 m from the target, the spatial dimension can be safely integrated over to calculate the measured signal, $M(h\nu)$ in J mm $^{-1}$ and remove uncertainty from the IP drifting from the ideal focal plane. This corresponds to

Crystal	Range, $n = 1$ (eV)	Range, $n = 2$ (eV)
KAP (100)	585-625	1170-1245
Quartz (10 $\bar{1}$ 0)	1830-1950	3660-3900
Quartz (10 $\bar{1}$ 1)	2330-2480	4660-4960

Table 3.2: Photon energy ranges captured by the three lowest energy OHREX crystals when operating at their nominal central Bragg angle of 51.3° or first and second diffraction orders, n .

a source spectral intensity incident on the crystal $S(h\nu)$ measured in $\text{J keV}^{-1} \text{sr}^{-1}$ via the spectrometer response, $G(h\nu)$, explicitly,

$$M(h\nu) = S(h\nu)G(h\nu). \quad (3.58)$$

The absolute throughput of the crystals was measured by MacDonald *et al* in a previous ORION experiment and fit parameters for

$$G(h\nu) = A(h\nu)^2 + B(h\nu) + C, \quad (3.59)$$

where $(h\nu)$ is the photon energy measured in eV, were determined for both p- and s-polarised incident light and for first and second diffraction orders [197]. The parameters for the lowest few energy crystals are presented in Table 3.3. There is unfortunately no spectrometer response data for the 10 $\bar{1}$ 0 crystal to first order due to the Si K edge within that energy range and the dramatic effect this has on absorption in its vicinity [198].

The OHREX is equipped with a 50 μm Beryllium filter to protect the crystals. The corresponding signal attenuation can be calculated using X-ray transmission data [199].

3.5.3 Extracting the data

The quartz OHREX crystals 10 $\bar{1}$ 0 and 10 $\bar{1}$ 1 were fielded on the experiment. Crystal images were recorded with BasTR2040 Fuji Image Plate. A typical shot image scanned with the FLA7000 scanner and converted to photostimulated luminescence units (PSLs) is presented in Figure 3.15. The average background signal was

Crystal	Order	Polarisation	<i>A</i>	<i>B</i>	<i>C</i>
KAP (100)	1	s	1.72×10^{-15}	-4.69×10^{-12}	2.89×10^{-9}
		p	1.40×10^{-14}	-1.74×10^{-11}	5.42×10^{-9}
	2	s	3.64×10^{-16}	-9.64×10^{-13}	6.95×10^{-10}
		p	5.03×10^{-10}	8.09×10^{-13}	5.03×10^{-10}
Quartz (10̄10)	1	s
		p
	2	s	4.50×10^{-15}	-3.40×10^{-11}	6.52×10^{-8}
		p	1.13×10^{-15}	-8.86×10^{-12}	1.73×10^{-8}
Quartz (10̄11)	1	s	1.00×10^{-16}	-1.74×10^{-12}	4.93×10^{-9}
		p	2.78×10^{-15}	-1.41×10^{-11}	1.79×10^{-8}
	2	s	4.70×10^{-16}	-4.50×10^{-12}	1.11×10^{-8}
		p	2.10×10^{-16}	2.11×10^{-12}	5.30×10^{-9}

Table 3.3: Sensitivity fit parameters as a function of photon energy, $h\nu$ in electron-volts ($G(h\nu) = A(h\nu)^2 + B(h\nu) + C$) for the three lowest energy OHREX crystals for p- and s-polarised incident photons and first and second diffraction orders [197]. Note that no data is available for the first order of the quartz (10̄10) crystal.

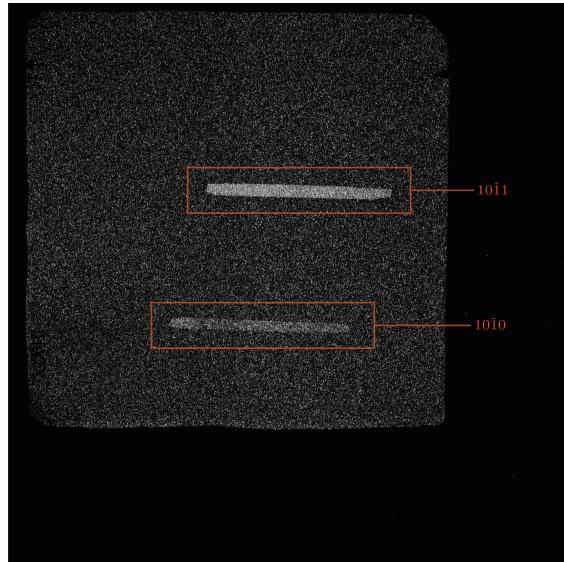


Figure 3.15: Unprocessed shot data from a FLA7000 scanned image plate converted to PSLs. The image plate and two crystal images are clearly visible.

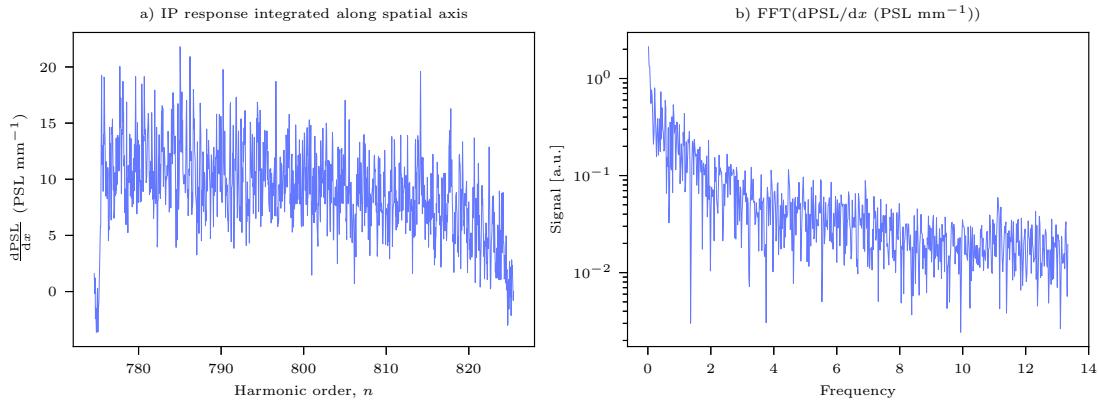


Figure 3.16: Typical (SP1, PMMA) uncalibrated shot data for the quartz (10\bar{1}0) image a) IP spatial axis integrated signal with dispersion axis. b) Fourier transform of a) with no evidence of harmonics.

subtracted. The x - and y -axes were converted from pixels to mm using the scanner resolution, ($25 \mu\text{m px}^{-1}$), and then to photon energies using the appropriate dispersion relations. The data was integrated over y to obtain the intensity in units of PSL mm^{-1} across each crystal image. The corresponding source signal is thus

$$S(h\nu)[\text{J keV}^{-1} \text{sr}^{-1}] = \frac{d\text{PSL}}{dx} \frac{P(h\nu)}{G(h\nu)} h\nu, \quad (3.60)$$

which can then be converted to a measured spectral intensity per harmonic per steradian, ready to be directly compared to the theory,

$$I_n^{\text{meas}}|_{(r=1 \text{ m})} = S(h\nu) \frac{dh\nu(\text{keV})}{dn}. \quad (3.61)$$

No sensitivity data is available for the $10\bar{1}0$ quartz crystal, instead, this lower energy crystal was fielded to attempt resolving of the X-ray harmonics. Figure 3.16 is a typical integrated signal in PSL mm and the corresponding Fourier transform for the quartz ($10\bar{1}0$) crystal image. As anticipated harmonics cannot be resolved.

Calibration and polarisation

The choice of the spectrometer response function, $G(h\nu)$, is non-trivial. One must first be assured that the second-order contribution is small relative to the first, true for this source spectrum. And also think carefully about the anticipated polarisation in the OHREX interaction plane. The polarisation of the XHHG beam relative

to the OHREX plane of incidence and reflection is 10.5° out of the plane. Unlike the RPM interaction, the OHREX crystal reflection is an entirely linear process and it is therefore acceptable to decompose the laser pulse into its constituents, explicitly, the field incident on the crystal is

$$\mathbf{E}_O = \mathbf{E}_{O,s} + \mathbf{E}_{O,p}. \quad (3.62)$$

After interaction with the crystal, the field is

$$\mathbf{E}_{\text{detector}} = \alpha_s(h\nu)\mathbf{E}_{O,s} + \alpha_p(h\nu)\mathbf{E}_{O,p}, \quad (3.63)$$

where $\alpha_i(h\nu)$ is the energy-dependent ($h\nu$) amplitude sensitivity of the reflection for s- and p-polarised respectively. Since the two polarisations are orthogonal, the intensity is

$$I = \alpha_s^2(h\nu)|\mathbf{E}_{O,s}|^2 + \alpha_p^2(h\nu)|\mathbf{E}_{O,p}|^2. \quad (3.64)$$

Noting that $\alpha_i^2(h\nu)$ are the calibration factors, $G_i(h\nu)$, and that

$$|\mathbf{E}_{O,s}| = |\mathbf{E}_O| \sin \phi \quad (3.65)$$

and

$$|\mathbf{E}_{O,p}| = |\mathbf{E}_O| \cos \phi, \quad (3.66)$$

where ϕ is the angle out of the interaction plane,

$$I_{\text{detector}} = (G_s(h\nu) \sin^2 \phi + G_p(h\nu) \cos^2 \phi) |\mathbf{E}_O|^2 = F(h\nu) |\mathbf{E}_O|^2, \quad (3.67)$$

where $F(h\nu) = (G_s(h\nu) \sin^2 \phi + G_p(h\nu) \cos^2 \phi)$ is the energy-dependent calibration factor for this OHREX orientation.

3.5.4 KBRXM

The ORION SP1 and SP2 spot size was routinely monitored on shot by the AWE laser team via two X-ray pinhole cameras and the time-integrating Kirkpatrick-Baez X-ray microscope (KBXRM) diagnostic, sat on the target chamber Port 21 and pointed at the notional TCC. This reflective X-ray microscope optic has a ten times

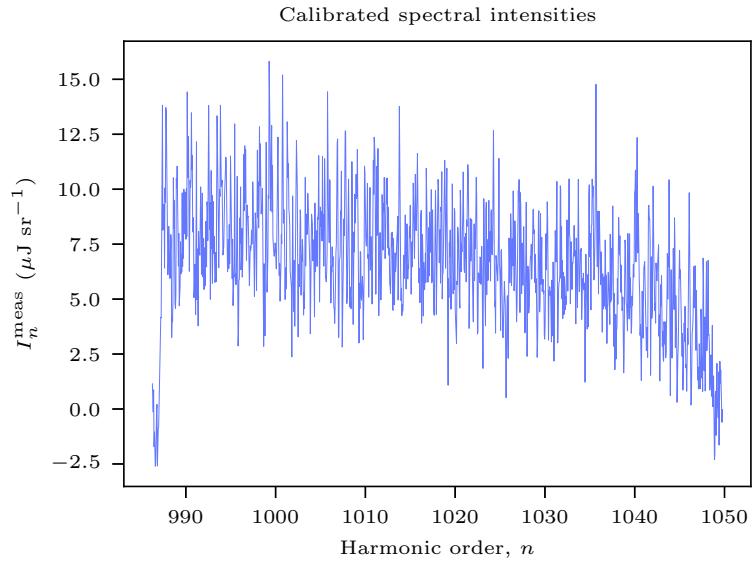


Figure 3.17: Typical (SP1, PMMA) ORION experiment calibrated IP response for the quartz (10 $\bar{1}$ 1) crystal.

magnification and spatial resolution of $15 \mu\text{m}$, comfortably resolving the laser spot size. Images were again recorded with TR-type IP. Direct mapping between the recorded images and real laser irradiance profiles is a highly non-trivial problem with any proposed model depending strongly on the assumptions made. However, by making the very reasonable assumption of spatial and temporal blurring of the spot size in the X-ray image due to electron transport away from the laser spot, the measured spot size will be an upper bound on the real spot size. It follows that the upper bound on optical spot widths corresponds to a lower limit of irradiance (since the on-shot energy is well-defined). These diagnostics determine sensible upper bounds for the laser spot size are $20 \mu\text{m}$ and $10 \mu\text{m}$ for the SP1 and SP2 beamlines respectively.

3.6 Experimental results

Figure 3.17 is a typical calibrated signal from the quartz (10 $\bar{1}$ 1) crystal, ready for comparison to the theoretical prediction. For each shot the mean signal across the dispersion axis was calculated and the errors from statistics, the fade time

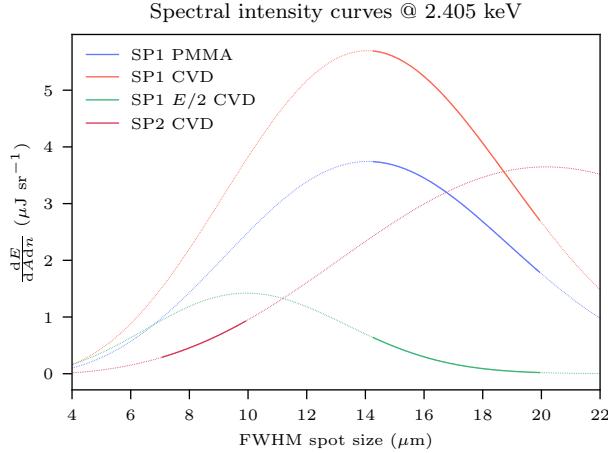


Figure 3.18: Dependence of the harmonic beam spectral intensity on laser spot size at 2.405 keV. Targets and beamlines relevant to the experiment are plotted. The half-energy single beamlet SP1 laser is demarcated by $E/2$. The thick lines indicate likely values for the laser spot sizes in this experiment. The SP1 harmonic beams are strongly affected by the exponential roll-off, unlike the SP2 beamline.

(11 %), the IP sensitivity (1.5 %), the dispersion axis (2.9 %) and the OHREX crystal reflectivity (11 %) accounted for.

As demonstrated in Section 3.2, the intensity of X-ray harmonics is highly sensitive to the intensity of individual pulse cycles due to the proximity of the measurement to the exponential roll-off in the spectrum. However, even neglecting the shot-to-shot variation, it is not possible to know the precise sub-cycle spatiotemporal structure of a petawatt-class laser pulse. Indeed, the simple measurement of the peak intensity of such a pulse remains an open problem [200, 201]. Approximations and assumptions must be made, that is, that the SP1 and SP2 beamlines can be adequately described by spatial Gaussians and temporal sech^2 and sech profiles respectively, and that the duration and energy are well-defined.

The upper limit on laser pulse spot size obtained from the KBRXM diagnostic corresponds to a lower limit on laser pulse intensity and therefore also of XHHG efficiency near roll-off. Figure 3.18 plots XHHG spectral intensity curves for the quartz (10̄11) crystal central energy as a function of laser spot size for the targets and laser pulse configurations explored in the experiment. The thick lines range from the upper limits of spot size as measured by the KBRXM diagnostic to a

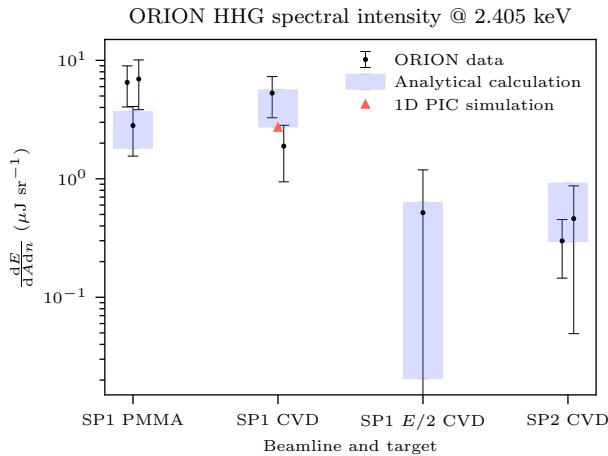


Figure 3.19: X-ray harmonic spectral intensities at 2.405 keV measured on the ORION experiment compared to theory and simulation. The analytical calculation corresponds to the thick lines marked in Figure 3.18. The 1D PIC simulation result is derived from Figure 3.5 and scaled to the ORION laser pulse duration.

spot area half that for the upper bound, a reasonable lower bound on the spot size. The spectral intensity of the harmonic beam from the SP1 beamline sits in a delicate balance between beam divergence via hole boring and the exponential roll-off. Note that only the roll-off leads to a reduction in the total energy contained within the harmonic beam. The strong non-linearity of the interaction leads to the single beamlet SP1 beamline producing a harmonic beam over 6 times weaker than the double beamlet configuration. Interestingly, the higher intensity of the SP2 beamline causes greater divergence and a lower spectral intensity of the harmonic beam than for the SP1 laser pulse.

Figure 3.19 plots the experimental results with comparison to theory and the simulation of Figure 3.5. Experimental data points are consistent within their errors with one another, the theory and the simulation, albeit with large uncertainties with such a low shot number. Notably, for the CVD targets, the SP1 beamline is on average almost 10 times brighter than the SP2 beamline despite the greater energy content and greater component of the laser pulse electric field directed into the plasma for the SP2 beamline. At the same time, the single beamlet shot is over 5 times weaker in intensity compared to the mean intensity of the full SP2

beamline. These highly non-linear scalings are consistent with XHHG theory but cannot be understood to be from bremsstrahlung emission.

Bremsstrahlung emission from steep density profile low-Z solid density targets irradiated by relativistic laser pulses remains an area of active research with experiments to explore this in the planning stages at the University of York. However, it is possible to derive approximate best-case scenario scalings of spectral intensity with incident energy. A plasma in equilibrium has a kinetic energy $E = 3/2k_B T$ [73]. In the ROM regime, energy absorption does not depend on incident energy, thus the plasma temperature increases linearly with incident energy. Now, assuming the target can be treated as a radiating black body, the spectral radiated intensity increases most rapidly with temperature when $h\nu \ll k_B T$ at which point the spectral radiance scales linearly with temperature [202]. Thus, in the best case, one could expect a linear increase in the spectral intensity measured from the double beamlet compared to the single if that signal originated from bremsstrahlung emission. Note further that this scaling depends not on the incident intensity but only on the incident energy, a well-constrained quantity on ORION.

The above discussion relied on the assumptions of equilibrium and a black-body radiation spectrum. While this is valid for the long radiating period after the main pulse interaction, in much of this thesis, emphasis has been placed on the non-equilibrium state of the interaction itself. There are two species of relevance during this time: the hot electrons around the front surface directly heated by the laser pulse and prepulse, and the hot electrons that are reflected specularly and are responsible for SHHG. In [203], bremsstrahlung of these species is measured experimentally for a 30 fs laser pulse and $a_0 = 2.8$. As the populations of each of these species are so small, the peak contribution to the photon spectrum is many orders of magnitude smaller than that of the experimental signal and can therefore be neglected.

Applying hole boring theory, the measured intensity of the SP2 beamline implies an energy $0.102_{-0.044}^{+0.1733} \mu\text{J}$ per harmonic at 2.405 keV, corresponding to a total efficiency of $2.04_{-0.86}^{+3.47} \times 10^{-10}$ into each harmonic at these photon energies. Due to harmonic source size shrinkage and the roll-off, the efficiency of the SP1 beamline

will be significantly lower. Integrating over the full spectrum given the SP2 X-ray efficiency [204], at the new focal position, the peak intensity is increased by a factor 1040^{+740}_{-290} , corresponding to peak powers higher than any other light source globally at this time. This dramatic magnification at focus is not unrealistic and is indeed consistent with the literature. Vincenti *et al* demonstrated a 3 order of magnitude gain for a 3 PW 20 fs system [168]. The lower power here is compensated by the longer duration. It is interesting that from Equation 3.45, for the ROM regime, the integration over the spectrum at the new focal spot contains a harmonic series that is only terminated by the roll-off. Thus, while simulations have demonstrated a gain of 1000 for a 3D simulation, it is unlikely that it had the necessary resolution to resolve the full peak in intensity and it could be notably higher.

Flying focus discussion here.

3.7 Summary and discussion

This chapter has described in full the experimental methods and results of the recent ORION XHHG experiment. The X-ray harmonic intensities were measured using the OHREX spectrometer. To describe the results, the ROM model of SHHG was applied, combining all pieces of the puzzle to reconstruct the measured intensities analytically. The experimental campaign was supported by PIC and hydrodynamic simulations of the main pulse and prepulse interactions respectively. PIC simulations agree with the analytical results for the ORION parameter space. No preplasma expansion was expected for the SP1 beamline and reasonable preplasma scale lengths for the SP2 beamline provided the addition of a single PM optic. Some discussion of plasma temperature was given.

The SP2 result is plotted in context with other laser systems in Figure 3.20. The XHHG beam sits in the vicinity of betatron radiation from laser wakefield experiments. Betatron radiation, however, is typically on the order of a few femtoseconds in duration. The stated power of the XHHG beam is averaged over the entire laser pulse to enable comparison to the dashed line of ROM scaling. In reality, the duration of the X-ray radiation burst, from Figure 3.6, is at most

3.7. Summary and discussion

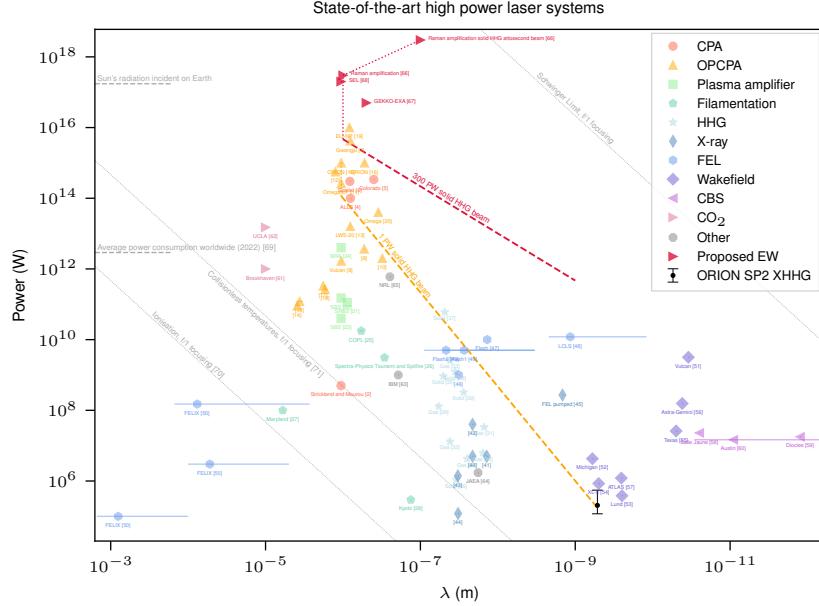


Figure 3.20: The ORION SP2 result in context with other high power laser systems. This is the only measurement to date of the absolute intensity of XHHG. Dashed lines indicate the power of the ORION SHHG beam and the SHHG beam that could be anticipated from the use of a sub-exawatt scale facility as will soon become available.

30 as and the beam is broadband, corresponding to a peak power of at least $2 \times 10^{10} \text{ W keV}^{-1}$. This is comparable to the LCLS, the most power XFEL. Figure 3.20 also highlights the power spectrum that could be obtained from a next-generation sub-ettawatt facility as are being commisioned currently or could be produced via Raman Amplification [66]. Simulations have suggested the Coherent Synchrotron Emission regime can be accessed for these laser pulse intensities up to the X-ray regime [132], hence the shallower scaling. Thus an entirely new parameter space becomes available both from the individual harmonics and for the conservative estimate of the intensity at the new focus. The figure is somewhat misleading with regard to the Schwinger Limit. This is for f/1 focusing. The RPM from an exawatt laser would have a must tighter focus and would access the Schwinger limit at the new focus. These exciting opportunities have contributed to the success of the recent proposal to study XHHG at GEMINI PW.

4

Attosecond electron bunches and X-ray pulses on GEMINI PW

Contents

4.1	Overview	117
4.2	X-ray harmonics on GEMINI PW	119
4.3	Observation of ZVP electron bunches	121
4.4	Application of XHHG to white light Laue diffraction	127
4.5	Contrast	128
4.5.1	Contrast on GEMINI PW	128
4.5.2	Kahaly mirror	132
4.6	Summary	133

4.1 Overview

The intensity of the attosecond duration X-ray harmonics on the ORION SP1 and SP2 beamlines has been measured. Even so, the uncertainties remained large, the geometry sub-optimal and the pulse train exceedingly far from an isolated attosecond pulse. Indeed, none of the proposed mechanisms for isolation (polarisation gating, lighthouse technique, etc.) could have any hope of success. The high shot rate, few femtosecond, highly customisable GEMINI PW facility at the CLF can address all the points raised while also providing conditions suitable for the production

of attosecond ZVP electron bunches discussed in Chapter 2. Naturally, GEMINI PW is attended by its own suite of technical challenges. The prior chapters of this thesis formed the body of a proposal for five weeks of beam time at the facility with shot dates planned for June 2024. This chapter outlines the extensive planning process for this upcoming experiment.

There are three primary experimental goals. First, to repeat the ORION experiment on GEMINI PW with an optimised geometry and thus resolve and measure the absolute intensity of X-ray harmonics, detailed in Section 4.2. Second, to observe ZVP electron bunches in transmission while simultaneously measuring the XHHG signal they generate in reflection, detailed in Section 4.3. Finally, the application of the X-ray harmonic beam in a proof of principle white light Laue diffraction setting is described in Section 4.4. Section 4.5 addresses the subject of laser contrast, a much thornier issue for the few femtosecond GEMINI beamline compared to ORION and covers the parallel experiment proposed by collaborators at Queen’s University Belfast to provide some illumination to the problem at hand. Note that no petawatt class sub-100 femtosecond laser facility has observed harmonics at oblique incidence. This section will also detail the strategy of contingency should the contrast prove to be too poor for the production of harmonics. Not discussed in this chapter is the work undertaken by Elliot Denis to temporally resolve chirped harmonic beams using hyper-spectral imaging.

The experimental geometry is relatively simple. Post Double Plasma Mirror (DPM) contrast enhancement¹, the GEMINI PW South (S) beam is focused by an $f/2$ parabola onto the solid density target at 45° AOI and p-polarisation, directing the specularly reflected harmonic beam to the West side of the chamber and the XUV and X-ray spectrometers.

¹A previous experiment measured the GEMINI DPM setup produced a reduction in peak pulse intensity of 50 % but improved the contrast by 5 orders of magnitude. More details on the DPM system are given in Section 4.5.

4.2 X-ray harmonics on GEMINI PW

The first action is to repeat the ORION experiment at GEMINI PW. For this purpose, the Oxford Engineering Department is designing and building a replica OHREX spectrometer to resolve and measure the absolute intensity of X-ray harmonics with OHREX crystals on loan from ORION. The lower energy KAP (100) OHREX crystal listed in Table 3.2 is more suited to GEMINI PW due to the lower energy of the beamline compared to ORION. The KAP crystal and replica OHREX will be calibrated on GEMINI PW using boron nitride targets. Regardless, the quartz crystals will also be fielded.

A parameter scan of XHHG as a function of S can be performed by varying the laser intensity, via beam apodisation, and the target material. The majority of shots will use target wheels of the relatively high damage threshold fused silica with the plan to advance to a Kapton tape. The fused silica targets are chemically etched in one corner to aid alignment.

Since SHHG is suppressed for circularly polarised light, bremsstrahlung emission and other X-ray production mechanisms can somewhat be ruled out by comparing the XHHG signal observed for p- and circularly- polarised light, produced via a quarter wave plate. At 45° AOI, however, this suppression is not total. And, the efficiency of heating is also reduced, reducing the signal from bremsstrahlung.

The OHREX spectrometer design is such that its technical complexity is entirely bound to the spherically bent OHREX crystals, thus reducing the challenges of alignment [196] and replica building. It is relatively insensitive to the distances between source and crystal (2.4 m) and from crystal to image plane (0.524 m). Variation in the angle of incidence from 38.7° shifts the spectral image away from the nominal energy range. The alignment of the spherically bent crystal planes to the crystal surface enables the relative convenience of optical alignment. A light source placed at the target location produces a line focus at the focal plane.

Filtering is necessary to reject the high-intensity low-order harmonics, however, for photon energies in the 600 eV range accessed by the KAP crystal, the standard beryllium filter transmission is too low. Instead, 400 nm of aluminium flash-coated

onto 1 micron of mylar is more suitable, corresponding to a transmission of 16 % at 600 eV [199]. Light leakage and alignment will be checked with IP before switching to the Raptor Photonics Eagle XV in vacuum X-ray CCD camera (EA4240XV-BN-CL) [205] to utilise the high shot rate on GEMINI PW. Note that with 2048×2048 active pixels of size $13.5\text{ }\mu\text{m} \times 13.5\text{ }\mu\text{m}$, not all of the 4 cm crystal image can be captured simultaneously by the camera.

The CCD camera must be liquid-cooled. For the prevention of damaging condensation on the camera window, a gate valve is required to isolate the OHREX when pumping the main chamber (waiting times for the camera to reach acceptable temperatures while under vacuum would be prohibitively long).

It is essential to check there is the required resolution in both the spectrometer and CCD camera for harmonic observation. For the highest energy crystal, nominal photon energy of 2.405 keV, 96 harmonics sit in the range accessed by the crystal, corresponding to a fractional energy $\Delta E/E = 6.45 \times 10^{-4}$ between harmonics. At $\Delta E/E = 1 \times 10^{-4}$, the OHREX crystal resolution is sufficient. Characterising the spectral shape of the harmonics at such energies is probably beyond the capability of such a detector. Those 96 harmonics are spread over the 4 cm crystal image. At a resolution of $13.5\text{ }\mu\text{m}$, the CCD camera has 31 pixels between harmonics and the harmonic width is about a pixel.

A 1D PIC simulation of the GEMINI PW beamline for the planned geometry predicts harmonics will be resolved for the KAP energy range, detailed in Figure 4.1. While the spectrum is similar to that for ORION parameters, the structure of the electric field is markedly different, illustrated in Figure X Since the preplasma scale length is $0.2\lambda_L$, large fractions of the laser pulse energy are stored in the pseudocapacitor field on the front surface of the plasma that can then be radiated simultaneously [127]. The ROM condition of no energy flux at the boundary is no longer valid. The reflected radiation is temporally compressed, producing attosecond pulses without the need for filtration and of greater peak intensity than that of the laser pulse. This regime is best described theoretically by the modified CSE model [132] but the ROM model does not deviate from this significantly.

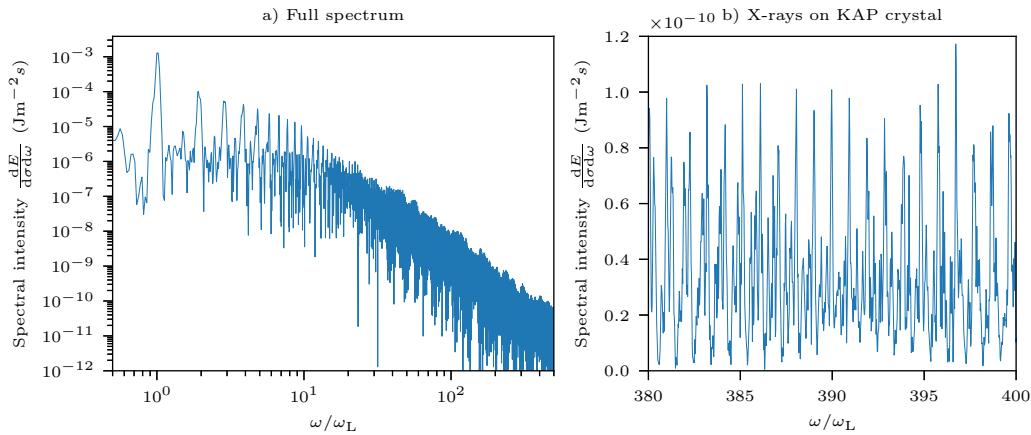


Figure 4.1: 1D PIC simulation of XHHG on the GEMINI PW laser system. The beamline is incident on a SiO_2 target under optimised conditions. a) The full harmonic spectrum. b) Window of the spectrum recorded by the KAP OHREX crystal. The harmonics are clearly visible.

Signals at the detector can be predicted using the theory of Chapter 3 using GEMINI PW parameters. Applying ROM and hole boring theory to a fused silica target and accounting for filtering and assuming no merging of harmonics, one can anticipate signals at the OHREX crystal position of $19.6 \mu\text{J sr}^{-1}$ per harmonic at 0.6 keV for the KAP crystal and $1.89 \mu\text{J sr}^{-1}$ per harmonic at 2.405 keV for the quartz (10̄11) crystal. Orienting the OHREX for s-polarisation, to maximise the signal at the image plane, corresponds to an average of 3.4 photons per pixel and 0.20 photons per pixel respectively. The quantum efficiency of the detector is close to 100 %. These photon numbers are no less than those measured in the ORION experiment while the background will be lower due to the order of magnitude lower energy on target. Implying adequate statistics for the observation of such signals.

4.3 Observation of ZVP electron bunches

The second stage of the GEMINI PW experiment is the observation of ZVP electron bunches, both mass-limited and bulk-produced. There has been some experimental evidence of attosecond and nano-Coulomb electron bunch generation from nano-targets [117, 121] and in reflection from solids [120, 122] with the latter demonstrating satisfaction of the specific conditions for Vacuum Laser Acceleration (VLA) to 10

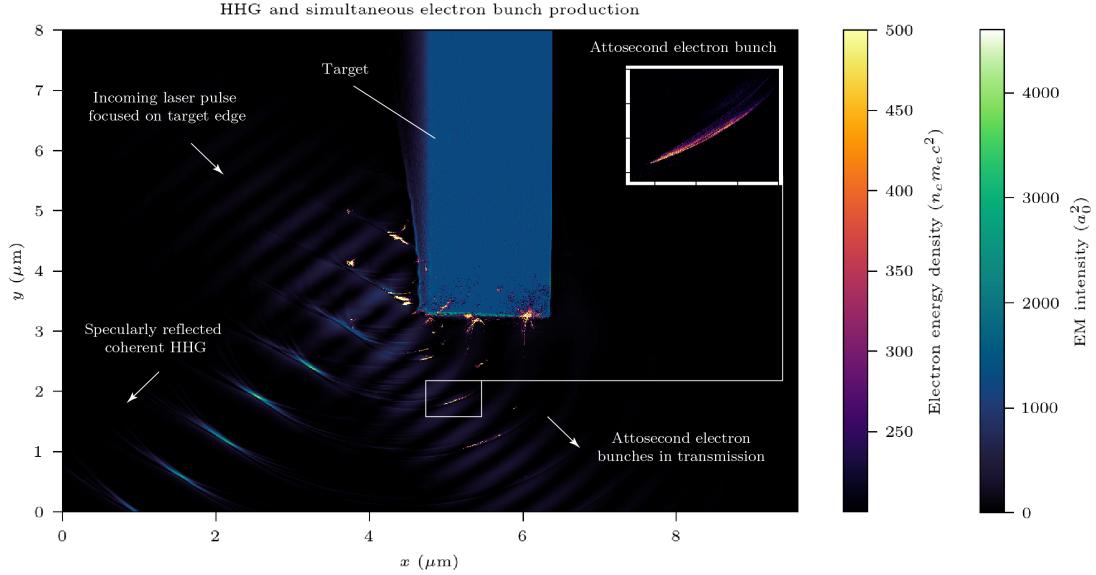


Figure 4.2: A simulation of the planned GEMINI-PW experimental setup for the measurement of ZVP electron bunches. This novel setup enables the simultaneous measurement of attosecond ZVP electron bunches and their coherent emission of X-ray light. The GEMINI-PW laser pulse is incident at 45° on the low-density polyethylene target with a preplasma scale length of $0.2\lambda_L$. For this angle of incidence, transmitted bunches and specularly reflected X-ray harmonics are produced at a frequency of ω_L .

MeV for a mildly relativistic laser pulse ($a_0 \approx 3.1$). Such experiments have guided the planning process for this experiment stage.

This experiment aspires to produce mass-limited ZVP electron bunches in the configuration simulated in Figure 4.2. Emitted electron bunch energies are maximised by focusing the 2 micron laser spot onto the edge of a transversely mass-limited target. The simulations suggest it is possible to simultaneously measure the specularly reflected XHHG and the attosecond ZVP electron bunches that produce it. Note via equations 2.47 and 2.25, the low-density target of Figure 4.2 is a more practical choice for the experiment relative to the aluminium targets of predominant use in Chapter 2, producing larger and more energetic bunches. Both bulk and mass-limited ZVP electron bunches will be generated. Via conservation of transverse momentum, the two species will be spatially separated at the observation point with bulk bunches at lower energies, experiencing no further acceleration phases post-ZVP.

While normal incidence was most convenient for the initial ZVP simulations, oblique incidence is better for SHHG [127, 132] and, as can be understood from

Equations 2.43 and 2.44, the new energy scaling expressions, for the ZVP mechanism. Not only is oblique preferable but it is essential to mitigate damage to the laser optics via back-reflection. The SHHG beam intensity at the focus can be over 1000 times that of the incident laser pulse [169]. It is necessary to conduct PIC code parameter scans for this new geometry.

High target edge precision is essential for the generation of attosecond duration ZVP electron bunches. The CLF Target Fabrication team has produced $200\ \mu\text{m}$ silicon wafer targets with well-defined edges of sub-micron precision via the Bosch process. The target design is presented in Figure 4.3a. The tapered finger structure adds a failure point to mitigate damage to adjacent targets from shock propagation. The cone of the array holder is of sufficient size for the full laser pulse to access the target edge. Scans, detailed in Figure 4.3, found an average surface roughness of 74.4 nm. The etching technique leads to scalloping of the surface parallel to the target surface and perpendicular striations towards the target rear. Neither pose any concern with regard to interference with the laser pulse in transmission. The target design and scans were performed by Sam Astbury [206]. Before firing target edges, SHHG production will be optimised for unstructured silicon wafer targets, providing information on the bulk propagating ZVP electron bunches for comparison with the mass-limited case. Again, a quarter-wave plate can be implemented to suppress the ZVP mechanism in the absence of vector potential zeroes.

Some relaxation can be anticipated in the requirement for sub-micron precision. There will be some target smoothing via preplasma expansion and the smoothing effect of the main pulse interaction as noted by Dromey *et al* [179]. Furthermore, transmission direction is dictated by laser vector potential and not surface angle, unlike reflection. At worst, ZVP energy scalings will be mildly affected by the change in angle. Thus tape drives will also be explored as an option. However, the $50\ \mu\text{m}$ jitter of such devices predicts a low success rate of $\sim 4\ \%$. Regardless, the gathering of sufficient statistics is not necessarily unreasonable with the high shot rate of GEMINI PW. Shot-to-shot variations in laser pulse focal spot position are

4.3. Observation of ZVP electron bunches

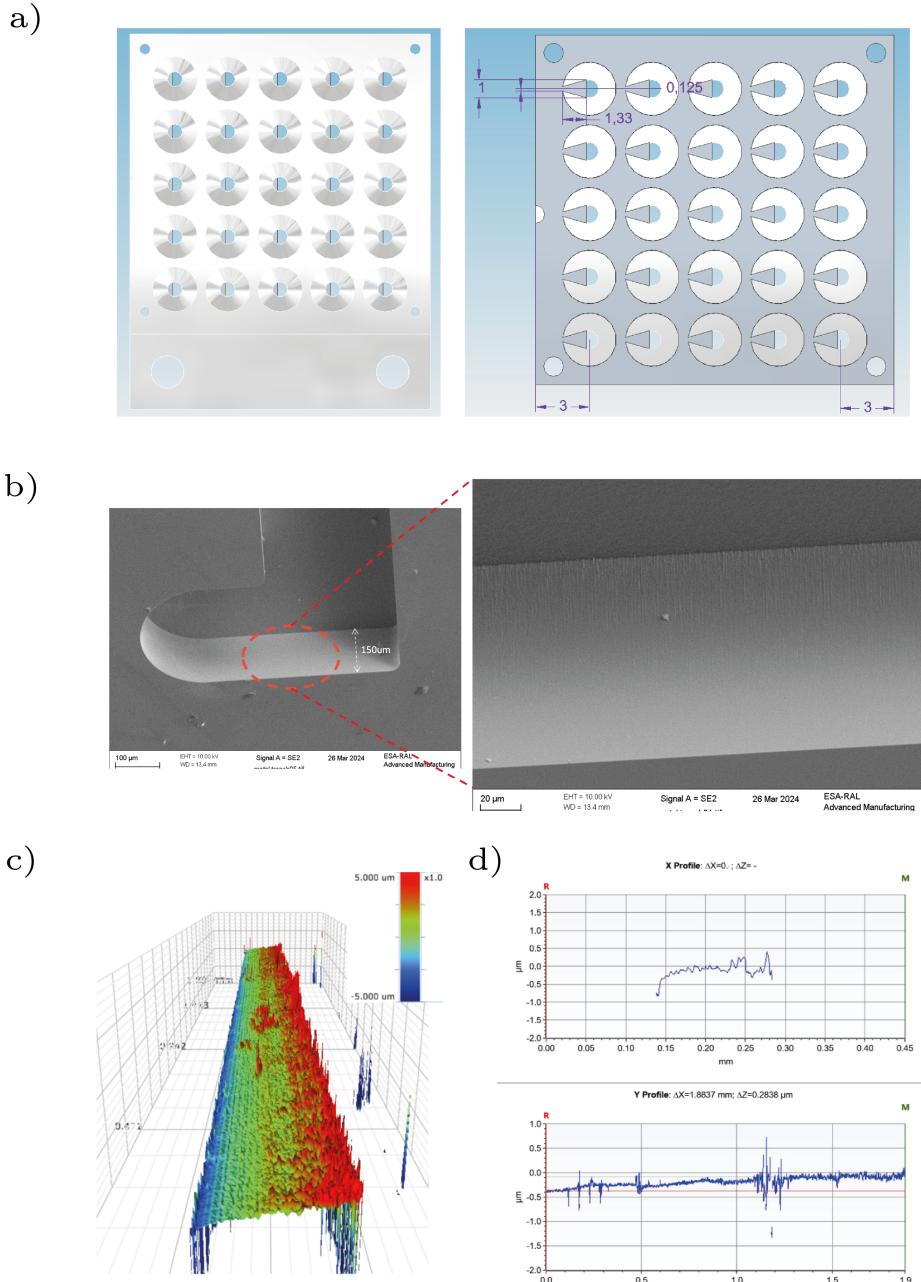


Figure 4.3: Targets designed for observation of ZVP electron bunches. a) Target array design with conical array holder and tapered finger, lengths in mm. b) Scan of the etched wafer, striations develop towards target rear due to etching technique. c) A 3D plot of surface variation. Scalloping and striations are visible but too small to interfere with laser pulse transmission at 45°. d) Lineouts of c), the y-profile is along the target edge of interest.

typically on the order of the focal spot itself. It is likely therefore that a third of silicon wafer target shots will hit an adequate portion of the target edge.

Successful shots will be identified using a third harmonic imaging line [207]. This diagnostic will also reduce the uncertainty of the hole boring calculations. The non-linearity of the third harmonic production mechanism reduces the possibility of background noise, producing a clear signal and a closer match to the true spot size than the X-ray emission diagnostics of the ORION experiment. For high resolution, the collimating reflective optic must be placed roughly the distance of the $f/2$ parabola focal length from the main interaction. Near the main interaction region, the B-integral is large thus precluding the use of ND filters and lenses. Instead, a wedge, typically double-passed, acts as an attenuator to extract the lower order highly divergent harmonics. A hole allows the less divergent higher orders through, enabling on-shot accurate spot size measurement. Far from the interaction, the collimated third harmonic signal can be extracted, filtered, refocused and imaged. Some light is reflected to the target via the wedge on the second pass, however this is not a concern. The focal length of the new optic is larger than that of the $f/2$ parabola and path lengths between the wedge and the target are on the order of a nanosecond. After such timescales, the target does not reflect specularly.

Resolving the $16\text{ }\mu\text{m}$ GEMINI PW spot is significantly more challenging than the $6\text{ }\mu\text{m}$ spot of the 1 J Astra-GEMINI laser system. This imaging line has been modelled by the ORION laser scientists using Zemax [208] accounting for all diffraction and spherical aberration. They have identified a set-up using two spherical mirrors for collimation and imaging that provides $10\text{ }\mu\text{m}$ resolution with 20 times magnification, reasonably resolving the spot. While the high spatial orders associated with the clipping of the reflected beam on target edge shots cannot be resolved, this should significantly alter the image structure, enough to identify their presence. Over time the wedge will be coated by material ablated from the main target interaction and will need to be replaced to prevent absorption increases and corresponding heat damage.

The size of the wedge hole must be established. Sitting somewhere between the near and far field, to first order, rays from all parts of the spot are lost equally through the hole in the wedge. After passing through the optical system, therefore, the wedge hole should simply reduce the signal at the image plane and not affect the resolution. Nonetheless, this should be modelled. One could argue that the wedge can reduce the spherical aberration by the removal of paraxial rays. The larger the hole the less likely the clipping of the potentially highly divergent high harmonic beam. Eventually, this will impact the third harmonic image resolution. At a minimum, the OHREX crystal must be fully illuminated requiring a half angle of 0.71° .

Applying the extended ZVP theory to the oblique incidence case for GEMINI PW parameters predicts electron bunches with a charge of ≈ 10 nC and energy of ≈ 30 MeV. Thus, a $15\ \mu\text{m}$ aluminium foil filter will reject the laser light without significantly affecting the electron bunches. Firstly, mass-limited and bulk electron bunch trajectories and beam divergences will be imaged on a LANEX screen. An MeV electron spectrometer aligned to the target edge can then measure the time-integrated energy spectrum. The spatially separated bunch types can be independently measured by adjusting the spectrometer position. Mordovanakis *et al* used Image Plate stacks to obtain the electron bunch structure and emission angle. This may prove necessary if the bunch types overlap due to high divergence [209].

While resolving attosecond durations remains a serious technical challenge of experimental science, it is possible to measure the production of Coherent Transition Radiation from a secondary target and therefore infer the presence of an attosecond bunch train. There is not time in this experiment to investigate this but provided success on this experimental campain, should form part of a future proposal. When electrons pass from one medium to another they emit transition radiation, in this case, the aluminium filter. If they are arranged into temporally regular spaced bunches that radiation becomes coherent and harmonics of the temporal spacing of the bunches can be observed. Both bulk propagating and mass-limited ZVP electron bunches will produce COTR, however, the attosecond duration should

mean the mass-limited bunches produce significantly brighter COTR that extends to XUV energies but that can be distinguished even at the optical range. [120].

4.4 Application of XHHG to white light Laue diffraction

The eventual but still somewhat distant goal is the creation of an attosecond X-ray diagnostic to illuminate crystal dynamics on attosecond timescales. Inspired by Laue diffraction measurements demonstrating dislocation microstructure of shocked crystals [210], this will be a proof of principle test of Laue diffraction from an X-ray harmonic beam of a single unshocked crystal.

It was Laue diffraction that led to the discovery of Bragg's law,

$$n\lambda = 2d \sin \theta \quad (4.1)$$

for diffraction order, n , incident wavelength, λ , the distance between crystal planes, d , and angle between the crystal plane and the incident photon, θ [211]. Irradiation of a crystal target with a 'white light', *i.e.* a broadband X-ray source, Equation 4.1 is satisfied for a range of crystal planes and corresponding angles simultaneously, producing *Laue spots* accordingly. The quasi-broadband X-ray harmonic beam is well suited to such an experiment provided sufficient photon numbers can be obtained.

This will be attempted using the BroadBand X-Ray Diffraction (BBXRD) diagnostic placed in the path of the specularly reflected beam and roughly 50 cm from the target. The lead-shielded and IP-lined BBXRD contains the crystal sample with a hole to allow the harmonic beam to enter. The sample is placed at an angle on a rotatable washer enabling a range of angles and corresponding energies to be tested: starting with a quartz target to investigate photon energies of 1.7 - 2.5 keV and moving to silicon to explore energies of 2.4 - 3.3 keV. The IP must be shielded to reject laser light, beryllium is most suitable. It may be necessary to integrate over multiple shots to produce the Laue spot.

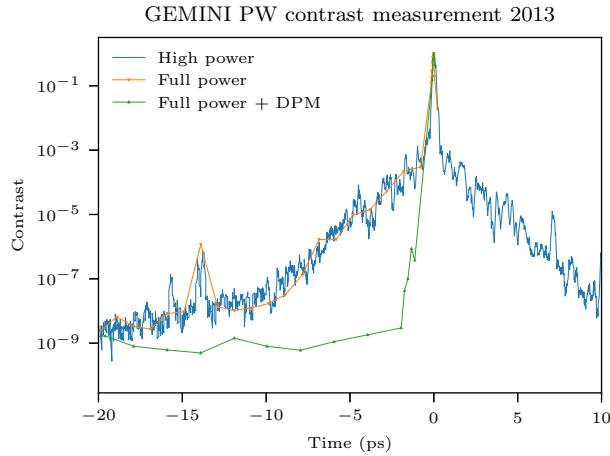


Figure 4.4: Native contrast of the GEMINI PW laser system measured using the Sequoia third order autocorrelator scans. The noise floor of the detector is $\approx 1 \times 10^{-9}$. The resolution is 100 fs, thus the main pulse is not resolved by the detector.

4.5 Contrast

4.5.1 Contrast on GEMINI PW

An underlying current of this thesis has been the subject of prepulse and corresponding preplasma expansion. While bright X-ray harmonics have been observed on both the Vulcan [166] and ORION sub-ps laser systems, few cycles laser pulse systems routinely underperform versus expectations. To date only at normal incidence (thus minimising heating) have harmonics been observed on GEMINI PW [212]. Evidence points to preplasma formation. The only parameter that can suppress SHHG from a p-polarised relativistic laser pulse is an overlarge preplasma [185] and indeed the transition to suppression is rapid [186]. While there exists established theories on the operation of plasma mirrors, these are based on fairly large assumptions that remain untested. Given the notoriety of the challenge of observing SHHG despite laser contrast being a relatively well-understood concept, and the general successes attributed to hydrodynamic codes, it seems quite possible that these large assumptions are the critical concern. They could also be responsible for the range of optimal scale lengths observed in experiments.

The most recently available contrast measurements of the GEMINI PW laser are presented in Figure 4.4. The DPM setup improves the contrast by at least 10^5 . More

recent measurements of the GEMINI PW DPM system have found a reflectivity of $1.23 \times 10^{-4}\%$ and 60 % before and after switch on, corresponding to a contrast enhancement of 4.88×10^5 . The DPM setup includes an acetate debris shield, without which shot quality would quickly degrade from plasma ablation between the mirrors.

Note in Figure 4.4 the early switch-on of the plasma mirror system. For a peak intensity on PM of $1 \times 10^{16} \text{ W cm}^{-2}$, this switch occurs an order of magnitude below typical ionising intensities [70]. Early PM switch-ons is nothing new [110], however, it points to a serious issue. Such early switch-ons cannot be described by the basic theory nor hydrodynamic simulation. Perhaps all can be explained by the complex atomic scale interaction of the laser pulse with the dielectric AR coating. Indeed, perhaps the haste to maximise the contrast at the nanosecond pedestal has been the downfall at the picosecond scale. And yet, providing the measured GEMINI PW contrast data to HYADES simulations predicts acceptable preplasmas, inconsistent with the experiment results.

Recent work has shown evidence of the propagation of an opacity front into a target due to laser prepulse before ionisation and plasma formation. Such a transition from transparency will dramatically affect laser pulse energy deposition into the target leading to significantly earlier plasma mirror switch-on times. The interaction appears to occur over a few picoseconds, a timescale to which preplasma scale length is highly sensitive. Note that such an interaction would be impossible to simulate given the multiple scales upon which preplasma formation occurs given the complexity of the transition to opacity. The GEMINI experiment, however, provides an excellent opportunity to probe this process in parallel to the main experimental goals.

A pure fused silica target consists of valence and conduction bands only. Electrons are excited to the conduction band by the laser and then simply drop back down again. As impurities are added to the target, the band structure becomes more complex and electrons can drop to intermediate states from which they cannot then return easily to the ground state. Thus, more energy can be absorbed and the transition to opacity occurs at earlier times. Hence, the more pure a silica

target, the later the transition to opacity and the smaller the preplasma scale length. This also suggests switching out the first plasma mirror for a pure uncoated silica target could improve contrast at the timescales where it matters. In this case, the first PM deals with the ps pedestal with its later switch on and the second PM tackles the ns pedestal with its traditional AR coating.

Presumably early plasma formation also occurs for Vulcan or ORION targets, however, once relativistic intensities are reached, recompression will occur. For such long pulse durations, there is sufficient time for steepening before the main interaction.

Contrast reduction will only become more critical with the arrival of sub-eta watt class next-generation laser facilities where native picosecond pedestals will already be relativistic. Steep preplasma scale lengths are not only relevant to the matters presented in this thesis but are essential for any thin foil interaction including the popular use case of ion acceleration via TNSA.

To investigate early plasma switch on, a 50 ps chirped probe beam will be focused on the target. Optical spectrometers in transmission and reflection will identify the opacity and plasma generation times respectively. This measurement could categorically demonstrate the failure of hydrodynamic codes for any pre-ionisation laser pulse intensities. One may wonder why such a failure is not identified in other branches of HED physics, such as the case of Inertial Confinement Fusion. It is simply a matter of scales. These effects are only relevant for the few femtosecond petawatt laser systems in this case of high sensitivity to preplasma scale length. To distinguish the probe beam from the bright non-linear target interaction, it will be focused on the interaction area out of the main interaction plane and with opposite polarisation.

Strategy

A series of contingencies has been developed to ensure the observation of harmonics under the assumption that preplasma scale length is the main issue. Phases increase in cost to peak laser intensity and therefore to the theoretical harmonic limit. During optimisation, the OHREX will be replaced with an XUV angularly-resolved

spectrometer. As of 2019, no experiment to confirm HB theory had been performed on a PW system [168]. Such XUV spectrometer measurements could be very interesting both to confirm the HB theory in the PW regime and to infer the intensity increase at PM focus. The first step will be to scan through the focal spot of the laser. Wavefront curvature at the target from slight defocussing can reduce divergence from hole boring thus increasing intensity levels at the detector while also reducing pre-heating.

The next action will be to apodise the beam, again this reduces the intensity on target and the harmonic beam divergence and has been shown to tip preplasma scale length into the optimal regime for SHHG [186]. This will reduce the intrinsic prepulse intensity by the same factor as the main pulse. To ensure the SHHG roll-off is above the 2.4 keV harmonics (such that the signal on OHREX is non-negligible), requires $a_0 > 8$. Such apodisation would reduce the prepulse by ≈ 10 times and corresponds to a reduction in the beam radius by a factor of 2. While harmonics were observed on the 1 J precursor to GEMINI PW, Astra-Gemini, apodising back to Astra-Gemini parameters does not guarantee success, the new front end produces a unique prepulse profile.

The intensity on DPM can be reduced leading to later switch-on times but a lower peak reflectivity. Failing that, there are more drastic measures available. First, a second target chamber layout has been designed that will reduce the angle of incidence on target to 30° to reduce target heating and second, frequency doubling is known to significantly enhance contrast. In fact, as with ORION this will likely overshoot, producing effectively zero preplasma.

Testing frequency doubling would be very useful for demonstrating the validity of the new contrast theory. The frequency doubling crystal available at GEMINI PW will reduce the intensity on target both from the generation efficiency (25 %) and apodisation (9 %) (the crystal has a diameter of 60 mm), therefore if there is an improvement in signal, it must be from the improvement to contrast. While Kahaly *et al* were able to observe harmonics for a beam apodised to $a_0 = 0.7$, provided the scale length was optimised, resorting to this method to produce harmonics

would preclude the observation of X-ray harmonics. However, it would justify the investment into a larger frequency-doubling crystal for future XHHG experiments at GEMINI PW. Such a setup would not require a DPM and the diffraction-limited spot is smaller. Hence, despite the inevitability of efficiency cost due to the frequency doubling process, similar intensities on target could be realised provided a 25 % efficiency non-apodising frequency-doubling crystal.

Once harmonics are visible, aspects of the system can be changed and the opacity and early switch-on hypotheses can be tested by looking for quantitative improvements to the signal. Replacing the pure fused silica target with impure BK7 or AR-coated targets should reduce the harmonic signal. Equally, replacing the first PM in the DPM chain with an uncoated PM should delay the PM switch on and improve the signal. Note that the CWE signal is distinguishable from the SHHG signal: it is more divergence and experiences a sharp cutoff at the plasma frequency [186].

4.5.2 Kahaly mirror

At some point, the signal will overshoot and enter the CWE regime, preferably early in the list of contingencies presented. At this point, the preplasma scale length can now be optimised. Kahaly *et al* [186] designed a simple but elegant method to do this. Simply partially cover a mirror in the laser chain with a small mirror. The difference in path length of light on this mirror is determined by the mirror thickness and distance from the main laser mirror surface. Thus the arrival time at the target of this added prepulse can be controlled. Since this has effectively apodised the beam, the parabola will then focus the prepulse spot to a larger area compared to the main pulse spot ensuring a uniform preplasma across the main spot while the diffraction-limited main spot will experience minimal aberration from the subtracted prepulse. The target must be routinely monitored to ensure the prepulse spot coincides with the main pulse spot. The mirror should have a mirrored back surface and an AR-coated front surface with a slight angle between the two faces to prevent any parasitic prepulse.

Kahaly *et al* show a linear relationship between main pulse delay relative to prepulse and scale length (due to more time for the preplasma to expand) but also note that for the same scale length, a larger delay (accompanied by smaller energy in the prepulse) produces higher quality harmonics. This could be understood as a natural consequence of the lower plasma temperatures, which improve the ability of the system to perform coherent SHHG behaviour. If harmonics are accessed using the Kahaly method, the signal could be improved by moving the prepulse mirror further from the main pulse mirror while also reducing the fraction of overlap between the two mirrors to reduce the energy in the prepulse. The Kahaly mirror should be placed before the final mirror before the parabola. The further back in the chain it is placed, the greater the danger of diffraction from the mount causing damage to the optical system. The linear relationship between main pulse delay and scale length coupled with the XUV SHHG signal measurements will quantify contrast improvements for discrete changes to the system.

4.6 Summary

This chapter has detailed the strategy, goals and hypotheses surrounding the upcoming GEMINI PW experiment, including supporting PIC and hydrodynamic simulations. It may prove that the contrast of GEMINI PW is insufficient to enable access to the highly interesting XHHG. However, should this be the case, a strong plan of action has been laid out that should both enable some level of success in harmonic generation and provide invaluable insights that could change the approach to SHHG experiments in the future. If harmonics can be resolved, there are many exciting avenues for new physics. Namely, the main experimental goals are the resolution of X-ray harmonics, the observation of ZVP electron bunches, the application of the harmonic beam as a diagnostic tool and the measurement of attosecond chirp on optical and XUV harmonics.

5

Summary and future work

Contents

5.1	Summary	135
5.2	Beyond GEMINI PW and future work	136

5.1 Summary

This thesis explored the post-ponderomotive interaction between high-power laser systems and solid-density targets. Chapter 2 explored the ZVP mechanism developed by Baeva and extended by Savin. It is instructive to refer to the future avenues suggested by Savin [126]. Almost all have been addressed. An experimental campaign to observe ZVP is underway. An analytical model for the observed inverse scaling of absorption with similarity theory for the ZVP regime has been developed and discussed and has been related to CSE and an extensive QED parameter scan was performed. The results were somewhat unexpected but inspired an extension of the theory to demonstrate the transition to QED is dependent on system parameters via the ZVP mechanism.

Beyond the scope suggested by Savin, this chapter extended the theory of ZVP to account for oblique incidence and mass-limited attosecond electron bunches, whose

properties were thoroughly interrogated. The first 3D PIC simulations identifying ZVP and the first 2D parameter scan for the confirmation of energy absorption were performed. The results of previous studies were validated using a more direct methodology where individual bunch energies were extracted.

Chapter 3 switched from theory to experiment and from ZVP to ROM, reporting on the results of the 2023 experimental campaign at ORION, supported by theoretical predictions and simulations. The theories of Baeva on the ROM model and Vincenti on hole boring were combined and applied to the calculation of absolute intensity through consideration of conservation of energy and extended to account for all gamma spikes for the sub-ps laser pulse. These results were compared to simulations before application to the ORION data. By consideration of the main sources of error, realistic limits on the predictions were made, finding reasonable agreement with the experimental results, suggesting the production of bright coherent attosecond X-ray harmonics and peak radiation field intensities beyond currently operational facility limits.

Chapter 4 brought both previous chapters together, discussing the upcoming GEMINI PW experiment and all its goals and perceived technical challenges that will pave the way for future work in this field.

5.2 Beyond GEMINI PW and future work

While all of Savin's suggestions have been explored, as is the nature of scientific research, new results are a chain reaction, producing an exponential expansion of ideas. Many of the theoretical ideas of this thesis will not be tested in the upcoming experiment but could form part of future beamtime proposals. It would be particularly interesting to demonstrate in experiment the heating effect of the XHHG beam or to attempt a more direct inference of the new peak intensity of the magnified harmonic beam. X-ray measurement on GEMINI could be combined with one of the isolating attosecond pulse ideas, for example, polarisation gating, to generate and measure the intensity of an isolated attosecond X-ray pulse. To fully probe ZVP theory would require a full parameter scan in target density. Foam

targets would gain access to the optimal low S regime [213] without the need for preplasma formation. To truly demonstrate the attosecond duration of the ZVP electron bunches would require imaging of their XUV CTR harmonics.

On the side of the simulations, it is indispensable, to test the new predictions for oblique incidence energy scalings and total electron bunch charge along with the angle of bunch ejection, the non-zero transverse vector potential of the laser will prevent the bunch from propagating directly along the transmission axis. It would also be useful to perform a parameter scan of the preplasma scale length. In this work, it was assumed that the optima for electron bunch production are simply those for SHHG, as was shown to be true for preplasma scale length in the Supplementary Information of [122] for their electron bunches in reflection.

Now that attosecond ZVP electron bunches can be produced, options must be explored for their guidance and application. The QED ZVP produced several interesting results. It must be redressed to understand the rapid rise in BW energy and justify the theory for the transition to QED and look further for the transition to SF-QED. Computational limits precluded the full reconstruction of the ORION X-ray harmonic beam even in 1D. These limits could be stretched by the arrival of GPU supercomputing architectures, currently being tested on ARCHER2.

The return of SHHG study to the CLF after more than a decade hiatus is certainly an indicator of the wider renewed interest in the field. This is in part spurred on by the recent Nobel Prize for attosecond physics and the rapid adoption of multi-petawatt class facilities globally. Vulcan 20-20 is only around the corner. Set to be the world's most powerful laser, this ambitious laser facility will provide 200 J in 20 fs. With the capability to access triple digits of peak normalised vector potentials, it is ideally suited to the study of XHHG and ZVP. The GEMINI PW experiment will build expertise providing proof of principle results with a bright future in mind.

Appendices

A

General plasma physics

Contents

A.1	Lorentz transformations of electromagnetic fields	141
A.2	The headlight effect	142
A.3	Geometric transverse emittance	142
A.4	The Bourdier method	145

A.1 Lorentz transformations of electromagnetic fields

The Lorentz transformations for electromagnetic field components parallel, \parallel , and perpendicular, \perp , to a frame of reference boost of velocity \mathbf{v} are [76]

$$\mathbf{E}'_{\parallel} = \mathbf{E}_{\parallel}, \quad (\text{A.1})$$

$$\mathbf{B}'_{\parallel} = \mathbf{B}_{\parallel}, \quad (\text{A.2})$$

$$\mathbf{E}'_{\perp} = \gamma_{\mathbf{v}}(\mathbf{E}_{\perp} + \mathbf{v} \times \mathbf{B}), \quad (\text{A.3})$$

$$\mathbf{B}'_{\perp} = \gamma_{\mathbf{v}}(\mathbf{B}_{\perp} - \mathbf{v} \times \mathbf{E}/c^2). \quad (\text{A.4})$$

A.2 The headlight effect

The headlight effect describes the beaming of an isotropically emitting source travelling at some velocity relative to an observer. Consider the geometry of figure 2.2 with the source (the laser pulse) travelling at an angle 2θ to the observer (in this case, the ablating front). A photon with energy E emitted from the rest frame of the source (the laboratory frame in this case) has a 4-momentum

$$\mathbf{P}_\mu = \left(\frac{E}{c}, \frac{E}{c} \cos 2\theta, \frac{E}{c} \sin 2\theta \right). \quad (\text{A.5})$$

As the interaction geometry is confined to a 2D plane, the z -component can be safely neglected. Applying the lorentz boost of equation 1.30,

$$\begin{aligned} \frac{E'}{c} &= \gamma \left(\frac{E}{c} - \beta \frac{E}{c} \cos 2\theta \right) \\ \frac{E'}{c} \cos 2\theta' &= \gamma \left(\frac{E}{c} \cos 2\theta - \beta \frac{E}{c} \right). \end{aligned} \quad (\text{A.6})$$

Solving these equations for the angle in the boosted frame,

$$\cos 2\theta' = \frac{\cos 2\theta - \beta}{1 - \beta \cos 2\theta}. \quad (\text{A.7})$$

A.3 Geometric transverse emittance

A beam¹ of particles is fully described by its six-dimensional particle phase space distribution

$$\rho(\mathbf{x}, \mathbf{p}) = \rho(x, p_x, y, p_y, z, p_z), \quad (\text{A.8})$$

where $\mathbf{p} = p_x \hat{\mathbf{x}} + p_y \hat{\mathbf{y}} + p_z \hat{\mathbf{z}}$ is the canonical momentum [138]. Under the Hamilton formalism, for ideal conditions, the six-dimensional volume of the beam in this space, termed the *emittance*, arises as a conserved quantity and is therefore a useful quantity to describe the beam quality. (something to do with it affecting the ability to focus the beam?? check the papers) It is useful to rotate the coordinate system so as to align with the beam's propagation. The distribution can be written as

$$\rho(\mathbf{x}', \mathbf{p}') = \rho(x_L, p_L, x_T, p_T, x_{T'}, p_{T'}), \quad (\text{A.9})$$

¹In this section it is electron beams and not bunches that are referred to to demonstrate the generality of these concepts.

where L is longitudinal to the beam's propagation direction, and T and T' are two orthogonal directions transverse to the beam's propagation. Where discussed in this thesis, T' will unanimously refer to the z -direction, that is, the additional direction in 3D simulations, all such simulations are designed such that the z -direction is transverse to the beam propagation direction.

Recording a six-dimensional phase space in experiment is impossible while in simulations it is almost prohibitively costly in terms of data storage. Hence, it is common practice to project the distribution onto three orthogonal sub-spaces corresponding to each spatial axis, L, T and T' and compute the area on each. Note that since the electron beam is ultra-relativistic, all electrons propagate at approximately c and therefore it is the transverse and not the longitudinal emittance that describes the beam's quality. As a particle beam does not typically exist with well-defined borders, the area used to describe the emittance is restricted to an ellipse containing only the high-density core of the distribution. For a subspace i , where $i = T$ or T' , Floettmann *et al* [214] derive the *transverse normalised emittance* as

$$\epsilon_{n,rms}^i = \frac{1}{m_e c} \sqrt{\langle x_i^2 \rangle \langle p_i^2 \rangle - \langle x_i p_i \rangle^2}, \quad (\text{A.10})$$

where $\langle \rangle$ is the second central moment of the particle distribution,

$$\langle ab \rangle = \frac{\int ab \rho(\mathbf{x}', \mathbf{p}') dV}{\int \rho(\mathbf{x}', \mathbf{p}') dV} - \frac{\int a \rho(\mathbf{x}', \mathbf{p}') dV \int b \rho(\mathbf{x}', \mathbf{p}') dV}{(\int \rho(\mathbf{x}', \mathbf{p}') dV)^2}, \quad (\text{A.11})$$

here $dV = \Pi_j dx_j dp_j$ for $j = L, T, T'$.

When working with emittances, most frequently in the literature it is the *transverse geometric emittance*, ϵ_{rms}^i that is discussed. This is a natural consequence of it being more readily accessible in experiments [138]. The geometric and normalised emittances are related via

$$\epsilon_{rms}^i = \frac{\epsilon_{rms}^i}{\gamma \beta_L}, \quad (\text{A.12})$$

where $\gamma = 1/\sqrt{1-\beta^2}$ refers to the beam's mean energy and $\beta_L \approx c$ is the ultrarelativistic beam's longitudinal speed.

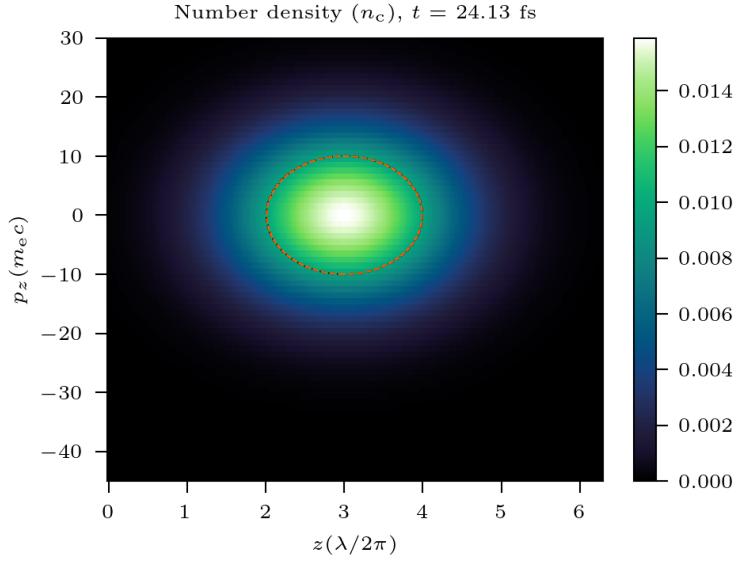


Figure A.1: Emittance calculation for an ideal Gaussian distribution in phase space, $f_n(z, p_z) = 1/(2\pi s_z s_{pz}) \exp[-((z - m_z)^2/(2s_z^2) + (p_z - m_{pz})^2/(2s_{pz}^2))]$, centred at $(m_z, m_{pz}) = (3, 0)$ with standard deviations $(s_z, s_{pz}) = (1, 10)$.

The Courant-Snyder invariant which describes the ellipse that corresponds to the emittance is²

$$\epsilon_{\text{rms}}^i = \gamma x_i^2 + 2\alpha x x' \beta x_i'^2, \quad (\text{A.13})$$

here the coordinates are x_i and $x'_i = p_i/p_L$ [215]. The Twiss parameters are

$$\alpha = -\frac{\langle x_i x'_i \rangle}{\epsilon_{\text{rms}}^i}, \quad (\text{A.14})$$

$$\beta = \frac{\langle x_i \rangle}{\epsilon_{\text{rms}}^i} \quad (\text{A.15})$$

and

$$\gamma = \frac{\langle x_i'^2 \rangle}{\epsilon_{\text{rms}}^i}. \quad (\text{A.16})$$

Thus the shape of the ellipse and the divergence of the beam can be determined. The elliptical contour defining the emittance of an ideal Gaussian phase-space distribution is given in Figure A.1. The contour is numerically calculated to contain 39.4(1) % of the particles, compared to 39.3 % from theory. Here the discrepancy arises from the finite grid on which the distribution is defined.

²Regrettably β and γ are the standard notations for the Twiss parameters, at all other locations in this Thesis, these parameters will refer to the standard relativistic beta and gamma factors of objects respectively.

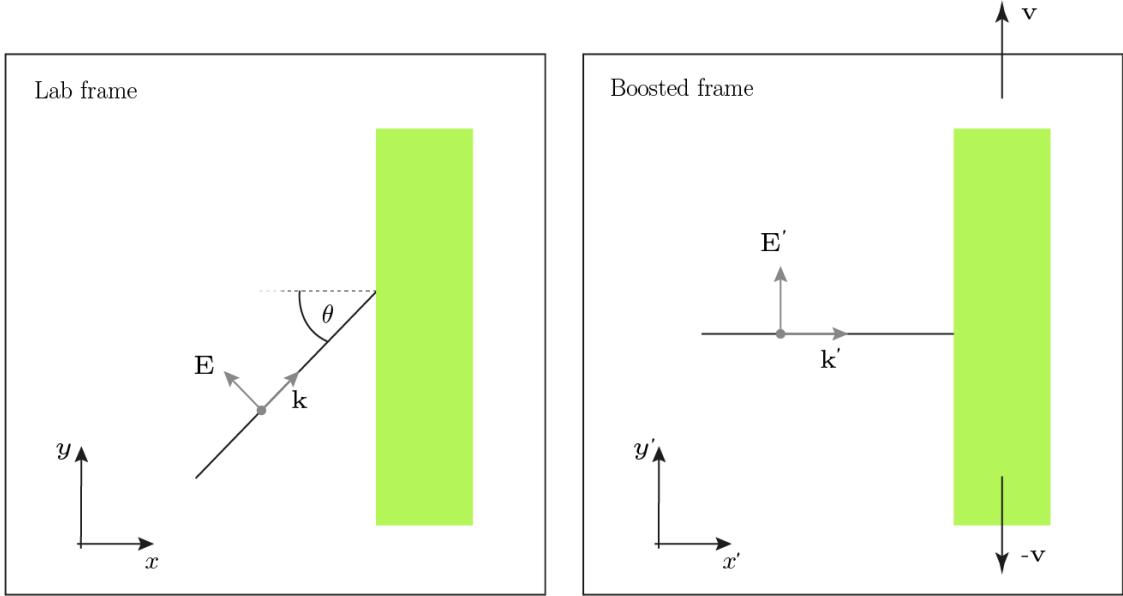


Figure A.2

A.4 The Bourdier method

The Bourdier method enables the modelling of an obliquely incident laser pulse in 1D geometry [171]. While some of this section may seem trivial, it is frequently misquoted in the literature. It therefore seems of great importance to provide a full derivation.

Consider a photon incident on a plasma block at angle θ as in Figure A.2. A boost is applied with velocity \mathbf{v} to the laboratory frame such that the photon is normally incident on the now streaming plasma at velocity $-\mathbf{v}$. The velocity transformation for the photon's velocity, \mathbf{u} , parallel to the boost is

$$\mathbf{u}'_{\parallel} = \frac{\mathbf{u}_{\parallel} - \mathbf{v}}{1 - \mathbf{u} \cdot \mathbf{v}/c^2}. \quad (\text{A.17})$$

Setting $\mathbf{u}'_{\parallel} = 0$, it is clear that

$$\mathbf{v} = \mathbf{u}_{\parallel} = c \sin \theta \hat{\mathbf{y}} \quad (\text{A.18})$$

in this geometry and

$$\gamma_v = \frac{1}{\sqrt{1 - \mathbf{v}^2/c^2}} = \sec \theta. \quad (\text{A.19})$$

Since Snell's law is frame invariant, the photon remains normal as it propagates into the skin depth of the plasma, a frame in which the interaction reduces to a 1D problem has been successfully found for all $\theta < \pi/2$. Those familiar with the topic may wonder how this is possible considering the 'ripples' that can be observed on the plasma surface for oblique incidence. The explanation for this is of course the relativity of simultaneity.

It remains to determine how do all the relevant quantities transform as such a boost is applied. Starting with a simple one: the photon's wave four-vector is

$$\mathbf{K}^\mu = \left(\frac{\omega}{c}, \mathbf{k} \right) \quad (\text{A.20})$$

and thus the frequency transforms as

$$\frac{\omega}{c} = \gamma_v \left(\frac{\omega'}{c} - \frac{\mathbf{v} \cdot \mathbf{k}'}{c} \right). \quad (\text{A.21})$$

Since $\mathbf{v} \cdot \mathbf{k}' = 0$,

$$\omega' = \omega \cos \theta. \quad (\text{A.22})$$

As

$$n'_c = \frac{m_e(\omega')^2}{4\pi e^2}, \quad (\text{A.23})$$

$$n'_c = n_c \cos^2 \theta, \quad (\text{A.24})$$

while the plasma block will be Lorentz contracted along $\hat{\mathbf{y}}$. Hence, the number density of electrons will increase as

$$n'_e = \frac{n'_e}{\cos \theta} \quad (\text{A.25})$$

leading to the perhaps unexpected

$$\bar{n}'_e = \frac{\bar{n}_e}{\cos^3 \theta}. \quad (\text{A.26})$$

Time is dilated

$$t' = \frac{t}{\cos \theta}. \quad (\text{A.27})$$

Consider now the more general case where the photon's electric field is rotated out of the x - y plane, *i.e.*

$$\mathbf{E} = E_0(-\cos \phi \sin \theta, \cos \phi \cos \theta, \sin \phi) \quad (\text{A.28})$$

and correspondingly

$$\mathbf{B} = \frac{\hat{\mathbf{k}} \times \mathbf{E}}{c} = \frac{E_0}{c}(\sin \phi \sin \theta, -\sin \phi \cos \theta, \cos \phi). \quad (\text{A.29})$$

The Lorentz transformations for electro-magnetic fields are

$$\mathbf{E}'_{\parallel} = \mathbf{E}_{\parallel}, \quad (\text{A.30})$$

$$\mathbf{B}'_{\parallel} = \mathbf{B}_{\parallel}, \quad (\text{A.31})$$

$$\mathbf{E}'_{\perp} = \gamma_{\mathbf{v}}(\mathbf{E}_{\perp} + \mathbf{v} \times \mathbf{B}), \quad (\text{A.32})$$

$$\mathbf{B}'_{\perp} = \gamma_{\mathbf{v}}(\mathbf{B}_{\perp} - \mathbf{v} \times \mathbf{E}/c^2). \quad (\text{A.33})$$

Using the above expressions for \mathbf{E}_{\perp} and \mathbf{E}_{\parallel} and transforming to the boosted frame,

$$\mathbf{E}' = E_0 \cos \theta(0, \cos \phi, \sin \phi). \quad (\text{A.34})$$

As anticipated for normal incidence there is no component of the E-field normal to the surface. Conveniently, the polarisation of the incident photon is unchanged despite having components both parallel and perpendicular to the transformation and

$$|\mathbf{E}'| = |\mathbf{E}| \cos \theta. \quad (\text{A.35})$$

The picture can now be completed. Since

$$a'_0 = \frac{e|\mathbf{E}'|}{m_e e \omega'} \quad (\text{A.36})$$

it follows that [216]

$$a'_0 = a_0, \quad (\text{A.37})$$

$$S' = \frac{S}{\cos^3 \theta}. \quad (\text{A.38})$$

Normalised Smilei units now come into their own in this new framework. In Smilei units, distances, times and vector potentials are unchanged by the interaction and can simply be extracted from the simulation in the required frame by multiplying by the frame of interest's reference quantity. Care must still be taken with densities.

B

PIC simulations

Contents

B.1 Convergence of 3D PIC simulations	149
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B.1 Convergence of 3D PIC simulations

To reduce the computation cost of the large 3D simulations, a lower resolution version of the original 3D simulation was performed. Results are presented in figure B.1, demonstrating reasonable convergence.

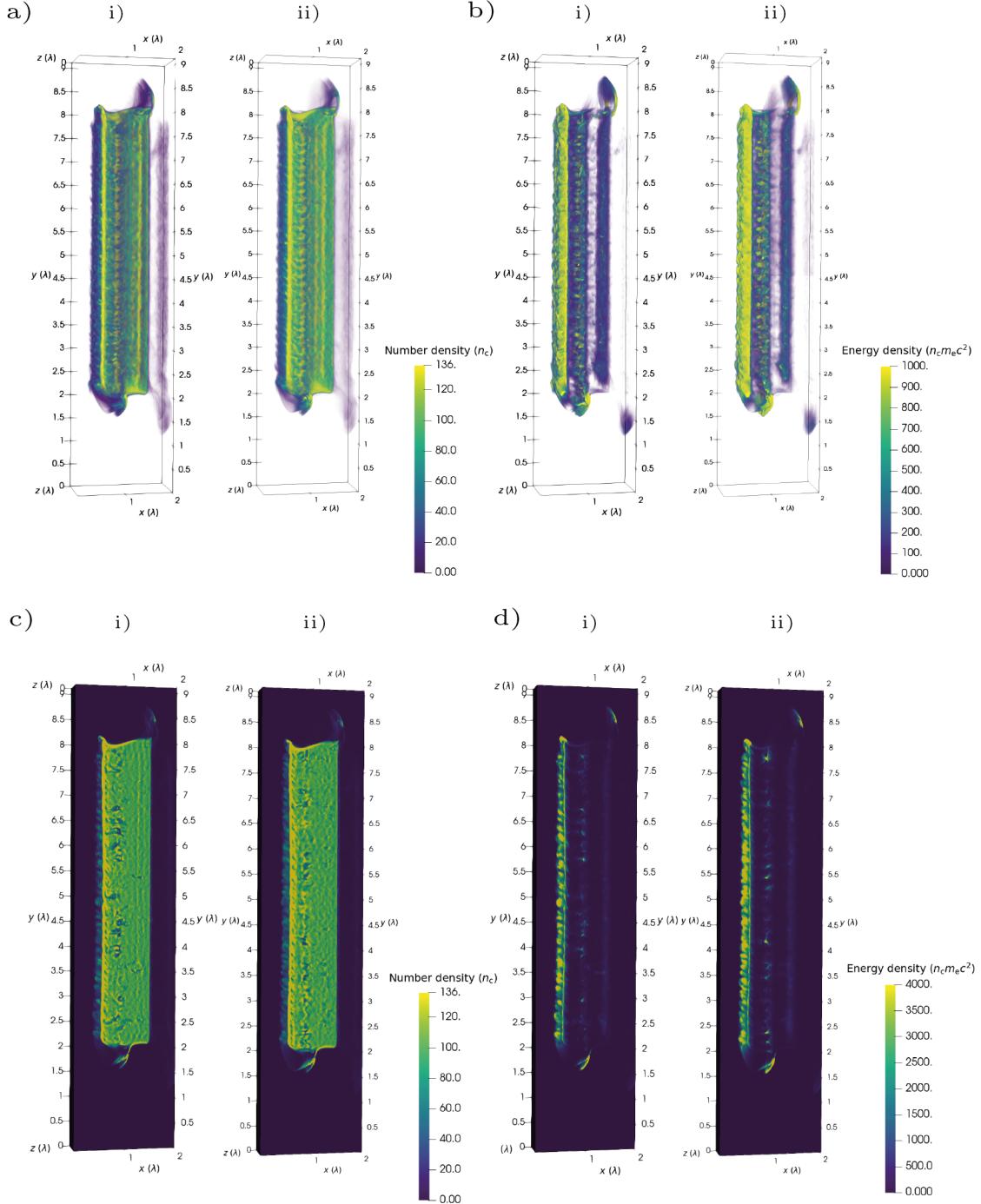


Figure B.1: Comparison between the initial 3D simulation and a lower resolution version. a) Electron number density. b) Kinetic energy density. c) and d) are slices of a) and b) respectively. i) Initial simulation. ii) Lower resolution simulation. Good convergence is demonstrated.

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