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**Laser-plasma interaction experiments using
high energy, high contrast OPCPA lasers**

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Declaration

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Abstract

A high-energy, high-contrast short-pulsed laser system using optical parametric chirped pulse amplification has been demonstrated as a platform for performing high intensity laser-matter interaction experiments. The existing Cerberus laser system at Imperial College was upgraded to include an improved stretcher system in its long-pulse OPCPA pump chain, and new optical parametric amplifier stages, including a high-energy single-shot stage, were designed, with the aim of achieving multi-terawatt power levels with optical parametric amplification alone once the upgrade is completed. A high-performance optical-limiter for protection of sensitive optical detectors was developed and characterized, and shown to provide orders of magnitude better performance than other systems reported in the literature. This setup was then modified for use as a supercontinuum light source, analyzed for its spectral stability properties, and shown to be stable in comparison to other supercontinuum sources reported in the literature. A laser pulse contrast diagnostic system incorporating the optical limiter into its architecture was designed, intended to allow measurement over high dynamic ranges and long timescales. An additional diagnostic for time-resolved absolute measurement of strong electromagnetic pulses from laser-plasma interactions using the linear electro-optic effect was demonstrated for the first time in the petawatt-regime. This system was tested at Vulcan Petawatt and shown to have favourable experimental noise characteristics in comparison to conventional EMP diagnostics. Furthermore, the electro-optic diagnostic was used to investigate the effects of target geometry on laser-driven EMP generation in the context of experiments with an ion-acceleration component.

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Chapter 1

Introduction

Powerful lasers can be used for a variety of purposes, ranging from widespread industrial applications such as cutting and welding, to fundamental physics investigations in areas including High Energy Density Physics and Laboratory Astrophysics [1], laser-driven Inertial Confinement Fusion (ICF) [2], relativistic laser-plasma physics [3] and sources of short-duration pulsed radiation, such as X-rays [4–6] and radio-frequency electromagnetic pulses (EMP), or compact accelerators for electrons [7, 8] and ions [9].

This PhD project is broadly focused on the use of high contrast laser systems (possessing a large ratio between the intensities of the main pulse and any pre-pulses) as a platform for performing high-intensity ($> 10^{18} \text{ W cm}^{-2}$) laser-matter interaction experiments. Alongside laser-plasma experiments using a range of novel targets (some very sensitive to pre-pulses), this has included some further development of the Cerberus laser system at Imperial College London through designs of high-contrast parametric amplification (Chapter 2). In addition to producing high-contrast laser pulses and applying these to experiments studying laser-driven electromagnetic pulse (EMP) generation and ion-acceleration (Chapter 7), this project has included the development and detailed characterization of a high-stability liquid-based supercontinuum source (Chapter 5) and demonstration of a liquid-based Optical Limiter (OL) system delivering orders of magnitude better performance than other OLs in the literature for the pulse parameters of interest (Chapter 4).

As part of the experiments studying laser-driven EMP generation, a novel diagnostic system for absolute measurement of strong pulsed electric fields was developed and tested, providing world-first low-noise electro-optic measurements of EMP from petawatt-regime laser-matter interactions (Chapter 6). In addition, a second diagnostic for measuring laser pulse contrast on a single-shot over long-timescales has been designed, utilizing the liquid-based Optical Limiter (OL) technology developed and described in (Chapter 4).

This thesis also contains plans for a substantial upgrade to the Cerberus laser system. The implementation of two additional planned optical parametric amplifier (OPA) stages will allow broadband amplification of pulses to energies of a few joules, and in theory shorter durations than possible with glass amplifiers due to some additional bandwidth generation, and the ability to support shaped bandwidth laser pulses. Replacing conventional laser amplifiers with OPAs is advantageous for production of the high-quality beams desired for laser-plasma research, particularly with regard to pulse contrast due to the lack of Amplified Spontaneous Emission (ASE) and highly intensity-dependent amplification. The very high-gain associated with OPAs allows for short amplifying-medium lengths compared to laser amplifiers, and thus more compact systems. Detailed designs for an upgraded Cerberus laser system, including a upgrades to both long and short pulse beamlines and a new capability for compression of laser pulses under vacuum conditions, are provided in Chapter 2. The experience and techniques required for this upgrade are intended to be applied at Orion (AWE) for the Orion Support Laser system.

1.1 Statement of Originality

The content of this thesis is the work of the author, unless specifically and explicitly referenced or stated otherwise.

1.2 Outputs by the author

1.2.1 Publications

The following publications involve work carried out by the author during the course of the PhD project:

1.2.1.1 Lead authored

T.S. Robinson et al., “Electro-optic analysis of the influence of target geometry on electromagnetic pulses generated by petawatt laser-matter interactions” in *EPJ Web of Conferences* **167**, 03007 (2018).

T.S. Robinson et al., “Spectral characterization of a supercontinuum source based on nonlinear broadening in an aqueous K_2ZnCl_4 salt solution”, *Applied Optics*, Vol. 56 No. 35 (2017).

T.S. Robinson et al., “Low-noise time-resolved optical sensing of electromagnetic pulses from petawatt laser-matter interactions”, *Scientific Reports* **7**, Article 983 (2017).

T.S. Robinson et al., “Ultrafast self-healing passive optical limiter based on metal salt solutions” (2017). **Completed and awaiting submission to *Optics Letters* pending resolution of IP ownership issues.**

1.2.1.2 Co-authored

F. Consoli, R. De Angelis, T. Robinson et al., “Generation of intense quasi-electrostatic fields due to deposition of particles accelerated by petawatt-range laser-matter interactions”, *Nature Communications* (submitted March 2018).

S. Patankar, E.T. Gumbrell, T.S. Robinson et al., “Absolute Calibration of Optical Streak Cameras on Picosecond Time-scales using Supercontinuum Generation”, *Applied Optics* **56**, 24 (2017).

N. Stuart, T. Robinson et al., “Comparative study on the temporal contrast of femtosecond mode-locked laser oscillators”, *Optics Letters* **41**, 14 (2016).

S.Patankar, E.T. Gumbrell, T.S. Robinson et al., “Multiwavelength interferometry system for the Orion laser facility”, *Applied Optics* **54**, 36 (2015).

N.H. Stuart, D. Bigourd, R.W. Hill, T.S. Robinson et al., “Direct fluorescence characterisation of a picosecond seeded optical parametric amplifier”, *Optics Communications* **336** (2015).

J. D. Hare, S. V. Lebedev, L. G. Suttle, N. F. Loureiro, A. Ciardi, G. C. Burdiak, J. P. Chittenden, T. Clayson, S. J. Eardley, C. Garcia, J. W. D. Halliday, N. Niasse, **T. Robinson**, R. A. Smith, N. Stuart, F. Suzuki-Vidal, G. F. Swadling, J. Ma, and J. Wu, “Formation and structure of a current sheet in pulsed-power driven magnetic reconnection experiments”, *Physics of Plasmas* **24**, 102703 (2017).

L.G. Suttle, J.D. Hare, S.V. Lebedev, G.F. Swadling, G.C. Burdiak, A. Ciardi, J.P. Chittenden, N.F. Loureiro, N. Niasse, F. Suzuki-Vidal, J. Wu, Q. Yang, T. Clayson, A. Frank, **T.S. Robinson**, R.A. Smith, and N. Stuart, “Structure of a Magnetic Flux Annihilation Layer Formed by the Collision of Supersonic, Magnetized Plasma Flows”, *Physical Review Letters* **116**, 225001 (2016)

J.D. Hare, L. Suttle, S.V. Lebedev, N.F. Loureiro, A. Ciardi, G.C. Burdiak, J.P. Chittenden, T. Clayson, C. Garcia, N. Niasse, **T. Robinson**, R.A. Smith, N. Stuart, F. Suzuki-Vidal, G.F. Swadling, J. Ma, J. Wu, and Q. Yang, “Anomalous Heating and Plasmoid Formation in a Driven Magnetic Reconnection Experiment”, *Physical Review Letters* **118**, 085001 (2017)

G.F. Swadling, S. V. Lebedev, G.N. Hall, F. Suzuki-Vidal, G.C. Burdiak, L. Pickworth, P. De Grouchy, J. Skidmore, E. Khoory, L. Suttle, M. Bennett, J.D. Hare, T. Clayson, S.N. Bland, R.A. Smith, N.H. Stuart, S. Patankar, **T.S. Robinson**, A.J. Harvey-Thompson, W. Rozmus, J. Yuan, and L. Sheng, “Experimental investigations of ablation stream interaction dynamics in tungsten wire arrays: Interpenetration, magnetic field advection, and ion deflection”, *Physics of Plasmas* **23**, 056309 (2016)

1.2.2 Conference talks

The following talks were given by the author at academic conferences or specialized EMP workshops:

- “Optical detection of electromagnetic pulses from petawatt solid and levitated microtarget interactions”, *Central Laser Facility High Power Laser Science Community Christmas Meeting*, Abingdon, December 2016.
- “Electro-optical measurement of electromagnetic pulses from petawatt laser-matter interactions”, *2nd EMP Working Day*, Warsaw, January 2017.
- “Electro-optical probing of strong electromagnetic pulses from petawatt laser-matter interactions”, *European Conference on Plasma Diagnostics*, Bordeaux, April 2017.
- “Electro-optical probing of strong electromagnetic pulses from intense laser-matter interactions”, *Plasma Physics by Laser and Applications Conference*, Messina, July 2017.

Chapter 2

High-Power Laser Systems

In this chapter, a high-level discussion of the most common types of high power laser architectures is presented, along with considerations of laser pulse contrast and how this can affect laser-plasma experiments. The benefits of optical parametric amplification, particularly in terms of contrast (the intensity ratio between the main pulse and ‘noise’ features), are discussed, followed by an introductory description of the Cerberus laser system at Imperial College London, the platform for most of the experiments described in this thesis. To conclude, a description of upgrades implemented or planned for Cerberus will be presented.

2.1 Master-Oscillator Power-Amplifier (MOPA)

The most commonly implemented architecture for high-power laser systems used in laser-plasma research is the Master-Oscillator Power-Amplifier (MOPA) configuration. This modular design separates the ‘laser’ (master oscillator), typically using mode-locking [10,11] to obtain high repetition rate trains of ultrashort pulses (as short as ~ 5 fs), from the additional amplification and beam-conditioning systems which make up the rest of the architecture. A simplified example of a MOPA architecture used for pump-probe type experiments is shown in Fig. 2.1, where the output of the master oscillator is split along two beamlines, which could be used for driving and diagnosing physical phenomena in a laser-plasma experiment.

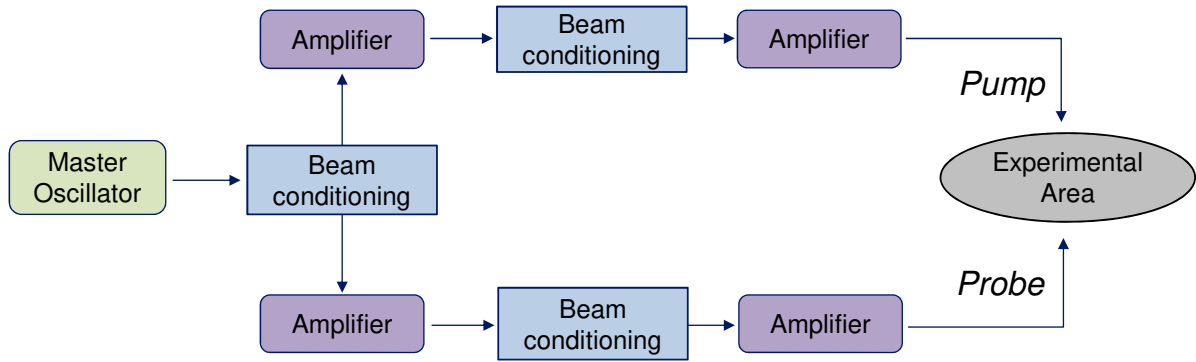


Figure 2.1: Simplified example of a Master-Oscillator Power-Amplifier (MOPA) architecture for high-powered laser systems, with a shared master oscillator acting as the source for two synchronized beamlines, in this case for a pump-probe experiment.

Using a shared master oscillator is advantageous for synchronization purposes, as this allows control systems to be locked to the oscillator pulse train. Consequently, the system is less sensitive to timing jitter associated with independent electronic control and triggering systems, while absolute synchronization and highly accurate time delays can be set using mechanical delay stages accurate to the few femtosecond level. For the example application of a pump-probe experiment, temporal synchronization is essential for reproducibility of experiments and reliable probing in time of the process being studied. Most high-power laser systems normally include Chirped Pulse Amplification setups (discussed in the next section) in order to reach the highest powers possible without catastrophic optically-induced damage to their constituent optical elements.

2.2 Nonlinear Optics

The study of nonlinear optics is essential for understanding and providing context related to the concepts discussed in this chapter, as well as Chapters 4 and 5, which deal respectively with nonlinear refractive index changes and supercontinuum generation. Nonlinear optics refers to the changes in properties of optical materials in the presence of strong light fields, and the understanding of physical phenomena associated with these modified behaviors.

The discussion in this section (with associated equations) follows that of Boyd [12], Chapter 1. The origin of nonlinear optical effects lies in the nonlinear polarization $\mathbf{P}(\mathbf{E})$ of dielectric

materials, given by

$$\mathbf{P}(\mathbf{E}) = \epsilon_0(\chi\mathbf{E} + \chi^{(2)}\mathbf{E}^2 + \chi^{(3)}\mathbf{E}^3 + \chi^{(4)}\mathbf{E}^4 + \dots) \quad (2.1)$$

where $\mathbf{E}$ is the electric field acting on the medium, and $\chi^{(i)}$ is the electric susceptibility of i^{th} order and ϵ_0 is the permittivity of free space. The wave equation for light in nonlinear optical media is

$$\nabla^2\mathbf{E} - \frac{n^2}{c^2} \frac{\partial^2\mathbf{E}}{\partial t^2} = \frac{1}{\epsilon_0 c^2} \frac{\partial^2\mathbf{P}}{\partial t^2}, \quad (2.2)$$

where n is the refractive index of the material and c is the speed of light in a vacuum. From this, it is apparent that additional terms in the nonlinear polarization result in additional terms in the wave equation, forming the basis of generation of new light waves in nonlinear optical media. Well-known nonlinear optical phenomena such as Second Harmonic Generation (SHG) [13], Third Harmonic Generation (THG) [14, 15], Sum and Difference Frequency Generation [16] (SFG and DFG) and the Optical Kerr Effect [17, 18] can all be predicted by inserting appropriate expressions for electric fields into Eq. (2.1). For example, consider an electric field containing two distinct frequencies, ω_1 and ω_2 , propagating through a nonlinear medium, described by

$$E(t) = E_1 e^{-i\omega_1 t} + E_2 e^{-i\omega_2 t} + \text{c.c.}, \quad (2.3)$$

where **c.c.** denotes the complex conjugate and for simplicity we consider a one-dimensional case and have neglected any possible wave-vector mismatch between the two fields. The square of this field is given by

$$\begin{aligned} [E(t)]^2 = & E_1^2 e^{-2i\omega_1 t} + E_2^2 e^{-2i\omega_2 t} + 2E_1 E_2 e^{-i(\omega_1 + \omega_2)t} \\ & + 2(E_1 E_2^* e^{-i(\omega_1 - \omega_2)t} + \text{c.c.} + E_1 E_1^* + E_2 E_2^*). \end{aligned} \quad (2.4)$$

By inserting this into Eq. (2.1), we can see that the second order polarization $\mathbf{P}^{(2)}$ contains

additional terms in $2\omega_1$ and $2\omega_2$, which correspond to SHG (the generation of a new wave at twice the driver frequency), and also terms in $(\omega_1 + \omega_2)$ and $(\omega_1 - \omega_2)$, which correspond to SFG and DFG respectively. Similar analysis can be carried out for the other terms in the Taylor expansion of the nonlinear polarization, which allows the existence a variety of other nonlinear optical processes to be predicted, including but certainly not limited to, Optical Rectification, Third Harmonic Generation (THG), the Optical Kerr Effect (leading to self-focusing and self-phase modulation, which are key to the discussion in Chapters 4 and 5) and Four Wave Mixing (FWM). An important consideration for the efficiency of nonlinear optical processes is *phase-matching*; this refers to maintaining a phase relationship between different frequency components involved in a mixing process (such as harmonic generation) along the propagation direction within the nonlinear medium (typically a crystal), such that contributions to the wave amplitude generated by the nonlinear interaction are in phase and constructively interfere at the crystal's output. If this phase relationship is not maintained, periodic energy transfer between the interacting waves occurs as the direction and conversion efficiencies remain low. Phase-matching must typically be actively ensured, and is commonly achieved by angle-tuning of birefringent crystals to remove the phase-mismatch between different frequencies (due to the frequency dependence of the crystal's refractive index), or alternatively by temperature tuning through the use of crystal ovens.

2.3 Chirped Pulse Amplification

The invention of mode-locking [10, 11, 19] resulted in the emergence of lasers capable of producing “ultrashort” pulses, ranging from hundreds of picoseconds down to tens of femtoseconds. Few-optical cycle pulses as short as 5.5 fs have been produced using Titanium-doped Sapphire (Ti:Sapphire) laser oscillators [20, 21]. However, the high peak powers associated with such short pulses can be problematic for systems involving additional amplification and transmissive optics due to intensity-dependent nonlinear phase (*B*-integral) accumulated by pulses, given by equation (2.5), where λ is the laser center-wavelength, n_2 is a given material's nonlinear

refractive index and $I(z)$ is the on-axis optical intensity.

$$B = \frac{2\pi}{\lambda} \int n_2 I(z) dz \quad (2.5)$$

Large B -integrals (>5) can rapidly lead to self-focusing and/or filamentation followed by beam breakup in cases where pulses have spatial intensity ripples, often resulting in catastrophic optical damage. Historically, the typical method for avoiding these issues has involved the use of larger beams and optical elements to reduce the incident intensity on the laser system itself. This can be undesirable for reasons of both cost and practicality, as solid state gain media and optical elements of sufficiently high surface quality (flatness, smoothness, damage threshold uniformity etc.) are usually difficult to fabricate on large scales. However, large-aperture optics are unavoidable when operating at high energies due to optical damage of surfaces and the limitations of saturation fluence in amplifiers.

Chirped Pulse Amplification (CPA) [22] is a method for producing extremely high peak power laser pulses without introducing the aforementioned damaging nonlinear effects. This involves amplification of a temporally stretched (chirped) pulse before re-compression to short pulse durations, shown pictorially in Figure 2.2. Stretching pulses reduces their peak powers, and hence the acquired B -integral. This allows smaller optical elements to be used in the laser system compared to conventional amplification schemes, or allows for extraction of higher peak powers from a given laser aperture where large on-target energies are also desirable. The stretching and compression of pulses in time is typically achieved using dispersive optical elements such as diffraction gratings [23, 24], prisms [25] or chirped mirrors [26], though high-dispersion glass blocks can be used for some applications.

2.4 Optical Parametric Chirped Pulse Amplification

Conceptually, Optical Parametric Chirped Pulse Amplification (OPCPA) is identical to CPA, except a different process is used for amplification. Instead of conventional gain-storage laser

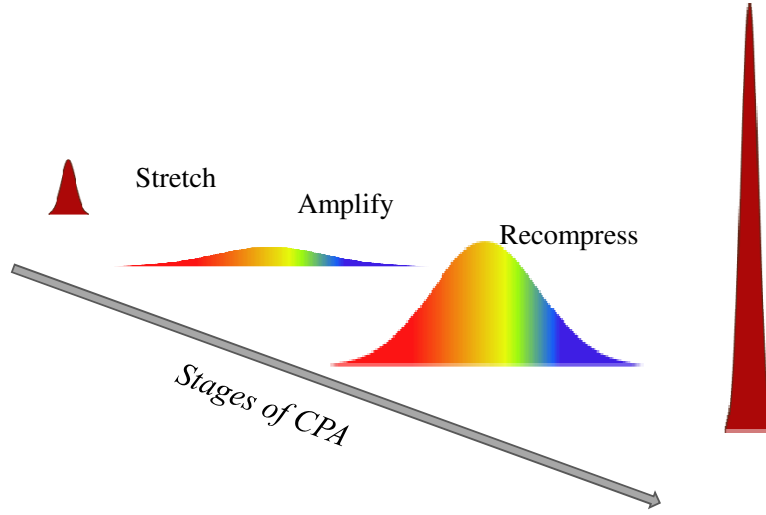


Figure 2.2: Visualization of the stages of Chirped Pulse Amplification (CPA). CPA involves amplification of a temporally-chirped (stretched) pulse before re-compression, yielding high peak-power pulses.

amplification, the nonlinear optical process of *Optical Parametric Amplification* (OPA) [27] is used.

Optical Parametric Amplifiers use Difference Frequency Generation (DFG), also known as stimulated down-conversion, to strongly amplify seed laser pulses. This involves mixing two pulses, typically termed the “pump” and “signal”, in a nonlinear crystal, resulting in conversion of pump photons with frequency ω_p to photons at the signal frequency ω_{sig} (therefore amplifying it), as well as at a frequency given by $\omega_{idl} = \omega_p - \omega_{sig}$, known as the “idler”. The DFG process is shown via a photon diagram in Fig. 2.3 and schematically in a laboratory setting in Fig. 2.4.

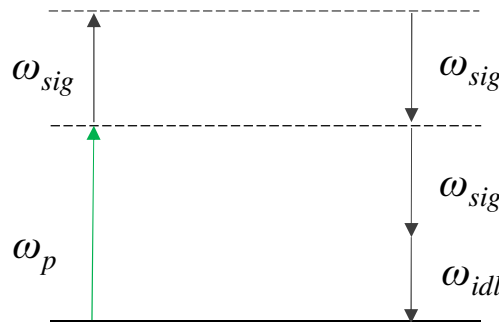


Figure 2.3: Photon picture of Difference Frequency Generation within a nonlinear crystal, where the dashed lines represent virtual photon states and arrows of different lengths represent photons of differing energies. The down-conversion of the pump photons is stimulated by the signal photons.

Idler photons are produced in order to satisfy photon momentum and energy conservation, hence the momentum relation $k_p = k_{sig} + k_{idl}$ must also be satisfied. This requirement is known as *phase-matching*, and is typically achieved by angle-tuning (critical phase-matching) or temperature tuning (non-critical phase matching). It is also worth noting that idler pulses are produced with the opposite chirp to the signal pulses [28], which has been shown to allow pulse re-compression using an identical system to the stretcher [29]. This could in principle be exploited for building particularly compact laser architectures and may also have some advantages in terms of pulse contrast, for example by using a non-imaging stretcher and compressor that is less prone to capturing scattered “off-angle” light.

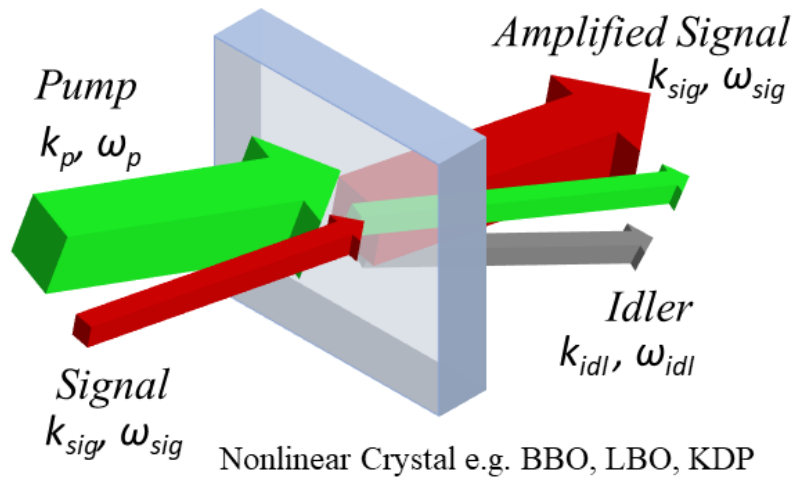


Figure 2.4: Diagram of the Optical Parametric Amplification process. A seed beam and high energy pump beam are propagated through a nonlinear crystal, leading to Difference Frequency Generation and hence amplification of the seed beam and generation of an idler component as the pump is depleted.

OPCPA systems possess several advantages over CPA systems; these include very high gains over broad bandwidths (potentially hundreds of nanometers for thin crystals), as well as the inherent nature of parametric processes. Parametric interactions do not involve energy storage in the host medium, so any heating is limited to parasitic absorption by impurities in the crystal, unlike the quantum defect in a “traditional” laser amplifier. Hence, OPAs do not suffer to the same extent from the thermal issues associated with conventional solid-state laser amplifiers, such as thermal lensing [30] and thermal stress-induced birefringence [31–33]. Thermal lensing is a result of a spatial temperature, and hence refractive index, variation in an amplifier rod,

which is effectively a Graded Refractive Index (GRIN) Lens. There are also some disadvantages, namely the requirement of a powerful and fairly high quality laser pulse for the pump, as well as greater operational difficulties due to the need to satisfy strict phase-matching as well as temporal and spatial overlap requirements for strong amplification.

2.5 Pulse Contrast in High-Power Laser Systems

The temporal *contrast* of a laser pulse refers to the ratio between its peak intensity and another part of the pulse at a particular point in time, hence the pulse contrast changes with time relative to the main peak. This is often quoted relative to the most intense ‘noise’ feature, and can sometimes be restricted to specific temporal windows, e.g. within 100 ps or 1 ns of the peak of the pulse. A visualization of the key noise features associated with ‘real’ laser pulses is given in Figure 2.5, not shown to scale to accentuate these possible features. These may include discrete pulses preceding or following the main pulse in time (pre- and post-pulses) or longer time-scale ‘pedestals’, both of which can have differing but significant impacts on the physics of a laser-matter interaction, for example by pre-ionizing targets.

Achieving high contrast is crucial for high-power laser experiments, as systems producing petawatt pulses with focused intensities exceeding $\sim 10^{20}$ W cm $^{-2}$ [34–36] are numerous, and a new generation of ultra-intense multi-petawatt systems (PETAL, Apollon, ELI-Beamlines 10 PW) are due to become operational soon [37–40], with feasible focused intensities exceeding 10^{23} W cm $^{-2}$. Typically, intensities of $\sim 10^{10}$ – 10^{12} W cm $^{-2}$ are sufficient to produce plasma in laser-solid interactions [41] and seriously affect experimental conditions. For example, a hydrodynamically expanding pre-plasma will result in deposition of the main pulse energy at low densities, changing the studied interaction (more detail on ionization and absorption processes is presented in the next Chapter). Alternatively, ablation of the target surface could result in shock waves coupling into the target, which limits how thin targets can be usefully fabricated and applied, as thinner targets are more easily destroyed by these shocks. At high powers, thermal waves transported by electrons or radiation can also destroy fragile targets. Based

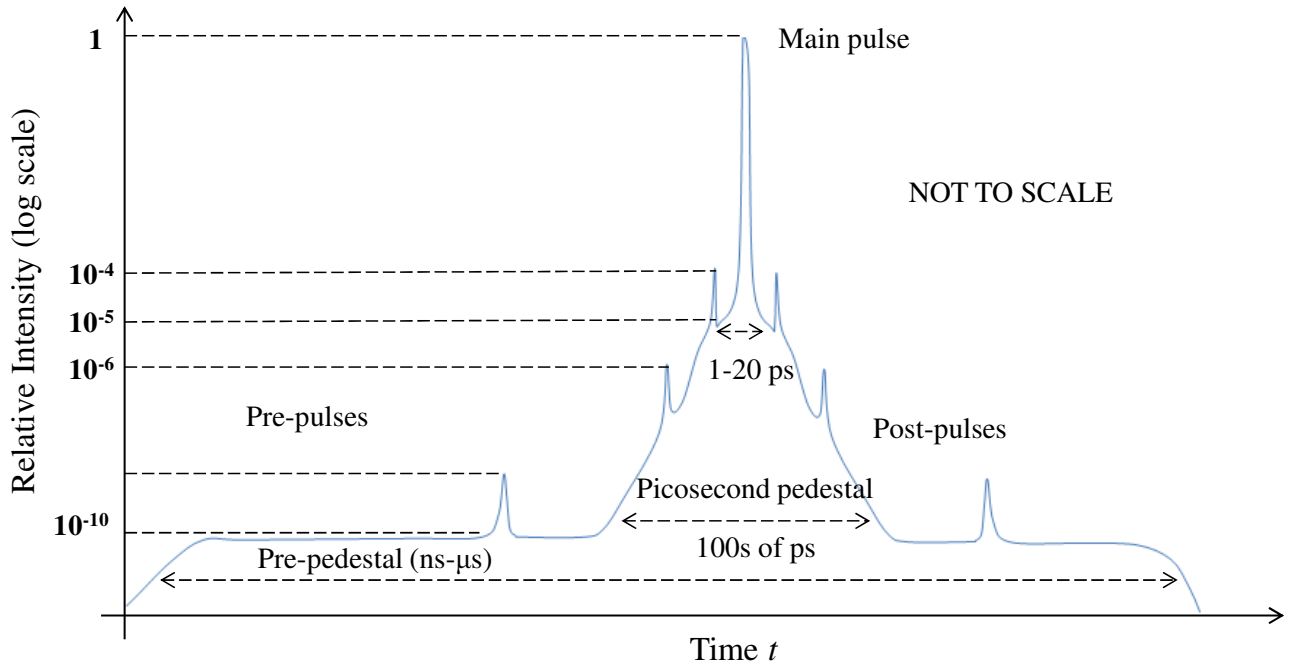


Figure 2.5: Representation of a real laser pulse with typical noise features around the main pulse, such as pre- and post-pulses and pedestals of short and long time-scales. Values on the relative intensity scale are approximate as noise features can vary from pulse to pulse. The temporal pulse profile is not shown to scale in order to accentuate the noise features.

on the aforementioned intensities, a contrast ratio of at least $10^{10} : 1$ would be required to prevent pre-plasma formation, with this value increasing for the newer generation of ultra-high power laser facilities. A system for measuring contrast over a large dynamic range is therefore critical to the success of high-intensity laser experiments and interpretation of measurements. Contrast information is also needed during data analysis to indicate the presence of detrimental pre-pulses or pre-pedestals (described in more detail in the next section), or to include their effect on the plasma density profile and energy absorption during simulations. Due to the low repetition rate of large-scale laser systems such as PETAL, Apollon, Vulcan, Orion or even Cerberus (limited by heating of the solid-state amplifiers), performing single-shot contrast measurements is also important.

2.5.1 Sources of Contrast Degradation

Contrast degradation can arise from multiple sources (see Figure 2.5); some of these are physics-limited, others amenable to ‘technical’ improvements. Pre- and post-pulses may be a result of

spectral clipping, leading to the formation of side-bands in the time-domain, though stray reflections in laser-oscillators or multi-passed amplifiers can also be responsible. The picosecond-timescale pedestals, or ‘shoulders’ (of triangular appearance on a log-scale), are typically attributed to small-angle optical scattering from the gratings [42] in stretcher systems [43, 44]; while scattering angles are sufficiently small for this light to still reach a target, these deviated rays are not fully compressed, resulting in a shoulder-like pedestal feature as shown in Fig. 2.5, distinct from the flatter nanosecond timescale pedestals.

Nanosecond timescale pre-pedestals in CPA systems are approximately uniform in intensity across the pulse’s temporal profile, and typically a result of Amplified Spontaneous Emission (ASE) [45] which can occur over timescales exceeding a microsecond, and does not re-compress to the same pulse durations as the main laser pulse. In OPCPA systems, an analogous process known as Amplified Parametric Fluorescence (APF) [46, 47] is responsible for formation of pre-pedestals. The timescales of this directly depend on the pump pulse duration, hence OPCPA systems with multiple OPA stages pumped by pulses of different durations, such as Cerberus, Orion or Vulcan, may have several noise pedestals on few-picosecond, nanosecond and hundreds of microseconds timescales.

Undesirable pre-pulses can also be generated from post-pulses in high-gain CPA amplifiers, according to a mechanism described by Didenko et al. [48] where interference of the main chirped pulse with a delayed replica within the gain medium, in the presence of the refractive index nonlinearity, results in phase modulation and hence a sinusoidally modulated intensity profile. For a chirped pulse, the spectrum is stretched out in time, hence any temporal intensity modulations are equivalent to spectral modulations. A CPA system’s compressor performs a Fourier transform of the incident light field, thus any sinusoidal modulations of the chirped pulse result in the generation of real satellite pulses upon compression, symmetrically spaced around the main pulse in time [48]. By necessity, the intensities used in optical parametric amplification systems (covered in more depth in Section 2.4) are sufficiently high for non-negligible B-integral (nonlinear phase) accumulation and nonlinear refractive index effects within the amplifier. Hence, the use of amplifiers (whether gain-storage or parametric) with plane-parallel end-surfaces, where back-reflections from the crystal-air interfaces are collinear with the main

propagating pulse, is accompanied by a risk of pre-pulse generation from the inevitably imperfect anti-reflection coatings on the crystal surfaces. Some of the aforementioned features are visible in contrast measurements from the front-end of the Cerberus laser system, shown in Fig. 2.6, such as the pedestal and pre-pulses. This measurement was taken using a third-order correlator diagnostic (built by N.H. Stuart [44]), which will be discussed in more detail in the next section (2.5.2).

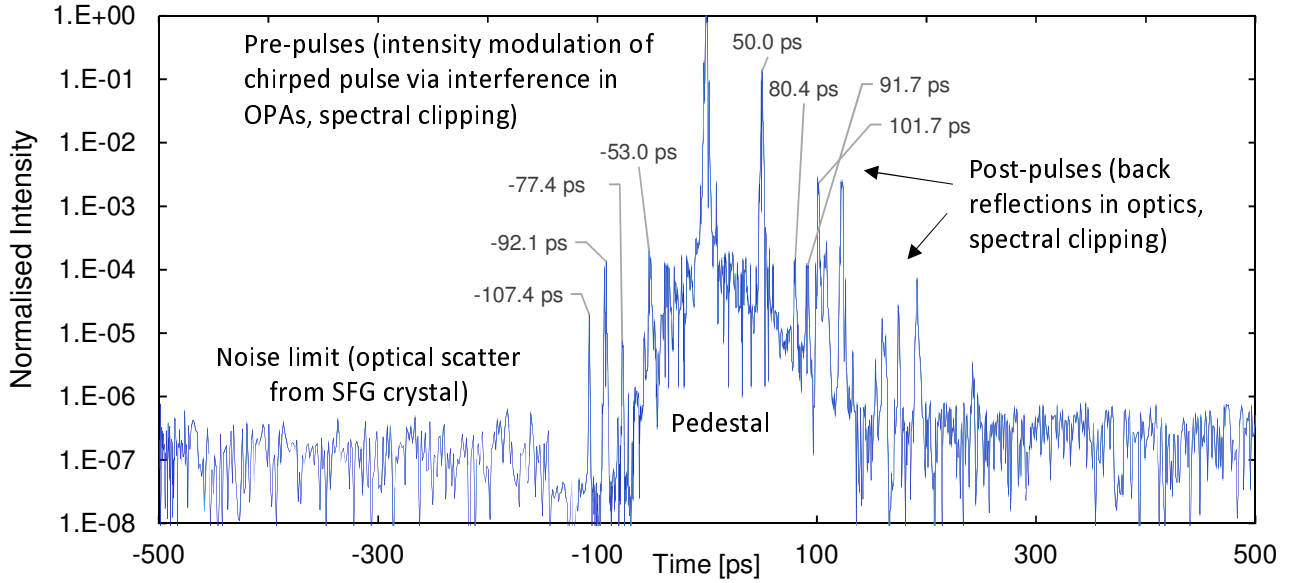


Figure 2.6: A pulse contrast measurement from the Cerberus laser system front-end (output of the ns-OPAs), demonstrating typical features that degrade contrast, such as the picosecond pedestal and some pre-pulses. The measurement was taken using a scanning third-order cross-correlator diagnostic, built by N.H. Stuart. The detection limit, which determines the dynamic range, here set by optical scatter within the nonlinear crystal, is a factor of 10^{-7} below the main peak.

Multiple pre-pulses can be seen in the measurement; some of these are symmetrically paired with a post-pulse in terms of temporal separation (those with similar absolute shifts from $t = 0$) from the main pulse and their relative intensities, which are attributed to the Didenko model of interference between a chirped pulse and post-pulse, while others are symmetrical in time, but differ by several orders of magnitude in intensity. These “ghost” pulses are most likely diagnostic artifacts (discussed further in [44]), resulting from the interaction between post-pulses on the 2nd harmonic probe and the fundamental main pulse inside the SFG crystal where the cross-correlation occurs, and can be reduced with more sophisticated diagnostics implementing probe pulse cleanup techniques [44].

2.5.2 Techniques for Improving Contrast

Several methods exist for enhancing the contrast of laser pulses onto target; these include frequency-doubling (or conversion to higher harmonics in general) before targets [49–51], the use of saturable absorbers in the beamline [52], plasma mirrors [53] as well as the use of Cross-Polarized Wave generation (XPW) [54–56]. XPW is an intensity-dependent rotation of light’s plane of polarization, hence can be used in conjunction with a polarizer to discriminate against lower intensity noise features, particularly within the front ends of laser systems. Energy losses from front-end contrast improvements are typically not an issue, as the energy can be recovered from the downstream amplifiers. Harmonic generation is a nonlinear intensity-dependent process (scaling with I^2 in the case of Second Harmonic Generation, for example), so the conversion from the fundamental to higher harmonics is much weaker for a pulse’s noise features, therefore increasing the ratio between the peak intensity and noise intensity. This can result in considerable contrast improvement. However, harmonic generation must be done post-compression, hence propagating these through nonlinear optical crystals can be problematic in terms of both dispersion compensation and B-integral accumulation. In practice, this severely limits the utility of harmonic conversion for contrast improvement, particularly for modern ultrashort, broadband, PW systems with sub-100 fs pulse durations. Instead, plasma mirrors are typically most successfully deployed for final-stage contrast enhancement [57–59] for applications requiring very high contrast ratios, although these have some limitations which are discussed below.

In a typical plasma mirror setup (an example is shown in Fig. 2.7a), the mirror element, typically rastered glass as a substrate material, is initially transparent, such that pre-pulses are transmitted through, however the leading edge of the main pulse ionizes the material once it reaches an intensity of $\sim 10^{10}$ - 10^{12} W cm⁻². The free electron density increases until the critical density is reached, upon which the plasma becomes highly reflective to the main pulse and any post-pulses, resulting in very high temporal contrast pulses reflected onto target. The time that this occurs relative to the peak (shown as t_{ref} in 2.7b) can be controlled by moving the mirror position relative to the focusing optic, providing some control of the incident intensity on the

material's surface. The reflectivity of the plasma mirror varies with intensity, and therefore changes with the intensity of the rising edge of the laser pulse. A small component of the pre-pulse will be reflected due to the finite reflectivity of the mirror substrate, however this can be minimized using anti-reflection coatings.

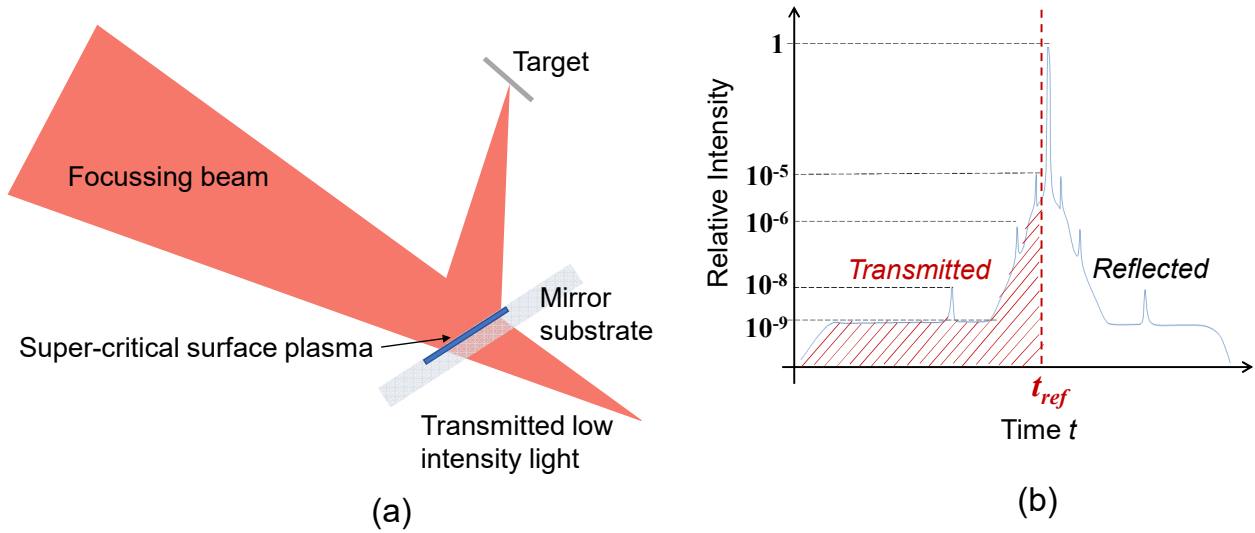


Figure 2.7: In (a), a diagram of a plasma mirror applied to high-intensity laser-matter interaction experiments is presented. An example temporal profile of a laser pulse is shown in (b), with the time at which a useful plasma mirror becomes highly-reflective shown as t_{ref} . The lower-intensity early-time section of the pulse indicated by the red diagonal lines is transmitted through the anti-reflection coated dielectric substrate, until the material becomes sufficiently ionized for the critical electron density to be reached.

Multiple plasma mirrors can be used for further improved contrast, at the expense of some energy loss required to ionize the substrate material. However, it could be argued that only the ‘useful’ energy on target is important, as with higher energy but poor contrast the desired experimental conditions are often unreachable. For example, the pulse contrast required for ion acceleration from ultra-thin foil targets, such as in the laser break-out afterburner regime where the target thicknesses of interest with near-IR drive lasers are often <100 nm (although this depends on the laser parameters so is not necessarily true in all cases), is typically estimated to be $>10^{12} : 1$ [60], achieved through use of plasma mirrors. Without these, fragile sub-100 nm targets are likely to be destroyed by shocks or thermal waves driven by the pre-pulses or ASE pedestal, such that the main pulse either interacts with a low-density plasma or even nothing at all.

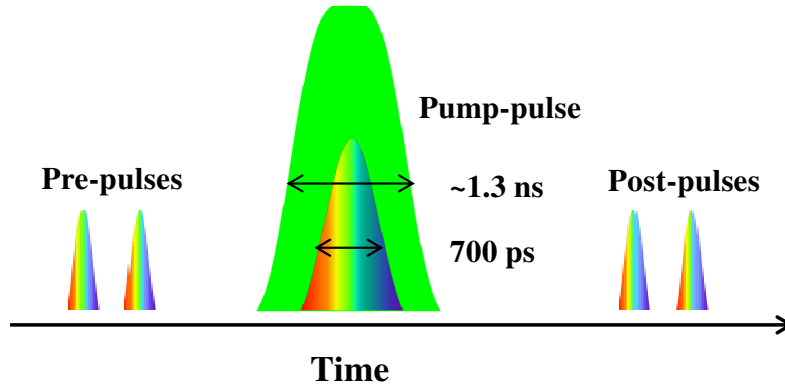


Figure 2.8: Diagram of the amplification in time using an OPCPA system. The gain window is limited to the pump pulse duration, such that pre- and post-pulses are not amplified, leading to higher contrast, high-intensity pulses upon re-compression.

Parametric Amplification is particularly suited to producing high contrast laser pulses, as the gain window is temporally limited to the pump pulse duration, as seen in Fig. 2.8. Consequently, the parametric fluorescence pedestals associated with OPAs are also limited in time to the duration of the pump pulse, and hence occur on significantly shorter time-scales than ASE pedestals from gain storage amplifiers. This is particularly the case if high-gain pre-amplifiers with picosecond pump pulse durations are used to provide most of the amplification [61], with OPAs pumped by nanosecond pulses and conventional gain-storage amplifiers acting as lower-gain, but higher energy, final stage amplifiers. Hence, the longer-timescale pedestals or any pre-pulses far from the main pulse see only a fraction of the total gain. Parametric fluorescence, arising from spontaneous down-conversion of pump photons seeded by virtual photons from vacuum fluctuations, while not entirely unavoidable, can be suppressed by increasing the intensity of the OPA seed pulses, as shown in a publication co-authored by the author of this thesis [62]. This observation was attributed to pump depletion; with increasing seed pulse intensity, the stimulated down-conversion of pump photons by the wings of the seed-pulse dominates the spontaneous conversion seeded by virtual photons, thus improving contrast.

However, OPA crystals with plane-parallel surfaces are also susceptible to generation of pre-pulses via interference of post-pulses and the main chirped pulse within the crystal if reflections occur within it, in accordance with the Didenko model [48] mentioned in the previous section. This is particularly prominent in situations where the signal pulse has been stretched to a

duration exceeding the round-trip propagation time of a pulse component back-reflected from the crystal-air interface, which is typically the case for OPAs amplifying nanosecond-duration pulses based on few millimeter to centimeter-scale crystals. Fortunately, a simple solution exists to mitigate this problem once identified: wedged crystals. When cut and polished with a small-angle wedge (of order of a few degrees), the end-surfaces are not plane parallel, ensuring that any back-reflected pulses are no longer collinear with the main propagating chirped pulse. This limits the interference between them, and hence prevents the formation of contrast-degrading pre-pulses from an otherwise high-contrast amplification technique. An experimental demonstration of this solution using the Cerberus OPCPA laser system, was given by N.H. Stuart, where the replacement of a high-gain OPA crystal with an alternative with an off-normal phase-matching angle resulted in the disappearance of several pre-pulses [44]. Even with a small wedge angle, it is necessary to compensate for the spatial phase difference introduced across the pulse profile, using either another identically wedged amplifier crystal rotated 180° about its propagation axis or an appropriate glass compensation block.

2.5.3 Contrast Measurement Techniques

Direct measurement of ultrashort pulses' temporal intensity profiles, for example using a photodiode or photomultiplier tube, is not generally possible. To characterize an extremely brief event like an ultrashort pulse, a physical process occurring on shorter time-scales than the light pulse is necessary to meet the time-resolution constraints; this is extremely limiting as ultrashort light pulses are some of the shortest known events. Rise times of photodiodes and photomultiplier tubes are limited by the transit times of photo-excited charge carriers and are generally on the order of nanoseconds (tens of picoseconds at best), far too slow for femtosecond or few-picosecond pulses, which must be characterized by indirect techniques. However, photodiodes can be used to obtain information about pre- and post-pulses and the ASE pre-pedestals occurring over nanosecond scales and longer (or tens to hundreds of picoseconds with the highest resolution detectors available), as demonstrated at Vulcan [47]. Although the main pulse durations are less than photodiode rise times, they can still be detected as intensity spikes

with durations set by the impulse response of the readout system, provided the diode is suitably protected. A water cell was used at Vulcan to limit the intensity incident on the photodiode from the main pulse when attempting to characterize lower intensity, long timescale pre-pulses and pedestals. Further discussion related to optical intensity limiting is presented in Chapter 4.

Indirect techniques used to measure short pulses, such as autocorrelation or cross-correlation, typically make use of nonlinear interactions between two pulses in crystals to produce signals that map out their temporal intensity profiles. Techniques such as Frequency Resolved Optical Gating (FROG) [63] or Spectral Interferometry for Direct Electric Field Reconstruction (SPIDER) [64] and their many variants are able to provide full information about the electric field and spectral phase, however they typically lack the dynamic range for useful pulse contrast measurements, with typical dynamic ranges ranging between 10^3 [65] and 10^6 (60 dB) for more advanced methods such as Self-Referencing Spectral Interferometry [66]. In this section, a few examples of techniques for contrast measurements will be reviewed.

2.5.3.1 Autocorrelators

Autocorrelation, one of the earlier contrast measurement techniques developed, was first demonstrated in 1966 by Maier and Kaiser to measure the length of nanosecond laser pulses [67]. This involved making a copy of a pulse using a beam-splitter in order to mix the two copies in a crystal with a $\chi^{(2)}$ nonlinearity, after undergoing a variable optical delay, as shown in Fig. 2.9. Early autocorrelations measured the crystal's SHG signal using photomultiplier tubes (PMTs). Methods to vary the time delay include mounting the autocorrelator reference arm's reflector (e.g. a corner-cube prism) on an electrically-driven loudspeaker [68] or shaker assembly [69].

Autocorrelators of this type are known as scanning second-order correlators, and have been used to measure time-dependent intensity profiles and contrast properties of laser pulses, achieving measurable contrast ratios of up to $10^8:1$ [68, 70]. However, these systems are often 'slow', requiring many pulses to be mixed before obtaining a useful measurement, with the acquisition time set by the time-resolution and temporal measurement range. Although measurement over

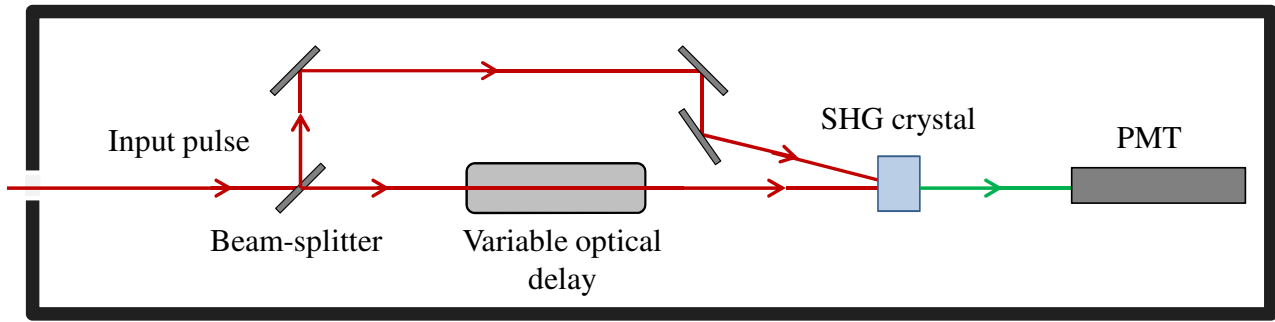


Figure 2.9: A simplified example schematic of a scanning autocorrelator used for measuring the temporal intensity profiles of short laser pulses.

relatively long time windows (up to ≈ 1 ns) is feasible, scanning autocorrelators are unsuitable for single-shot applications or real-time monitoring without suitable modifications [71,72]. Furthermore, the stability of the laser system over time becomes an important consideration when using a scanning system. Single-shot systems are thus preferable for measuring a laser pulse's temporal properties, with the key advantage that they can be used for real-time monitoring of a laser system. This is particularly desirable for high intensity petawatt systems (which typically have fairly low repetition rates e.g. one shot per 40 minutes at Vulcan PW), as it allows for on-shot contrast measurement. Single-shot architectures typically split a pulse, cross the copies at a particular angle in a nonlinear crystal and measure the 2^{nd} or 3^{rd} harmonic signal intensity (depending on the correlator design) using a CCD camera. This method allows temporal delays to be mapped into transverse displacements on the CCD. By increasing the crossing angle, larger time delays, and therefore temporal measurement ranges, can be obtained. However, this imposes a limit on the temporal ranges of single-shot systems due to practical limitations on pulse crossing-angles, although this can be mitigated somewhat via techniques using pulse-front tilt (typically added using diffraction gratings) [73] to increase the relative time delay. Furthermore, second order correlations are symmetric in time delay, so it is not possible to distinguish between pre- and post-pulses. As most laser-plasma experiments are particularly sensitive to pre-pulses rather than post-pulses, accurate measurement of these is critical for contrast characterization and to guide improvements to the laser systems used as experimental platforms. This problem may be overcome using a *third-order* or *cross-correlator*.

2.5.3.2 Cross-correlators

Third-order intensity correlation was demonstrated in 1969 for measurement of the temporal intensity profiles of laser pulses [74]. This technique has since been used to construct cross-correlators capable of single-shot duration and contrast measurements with a dynamic range of $10^8:1$ [75]. Cross-correlators involve mixing the original pulse with a frequency-doubled copy of itself in a nonlinear crystal, where a third harmonic signal is produced via SFG. This asymmetry between the fundamental and second harmonic pulses allows the direction of time to be resolved. The third harmonic is then typically measured using a PMT for a scanning system. A simplified example of a cross-correlator setup is shown in Fig. 2.10.

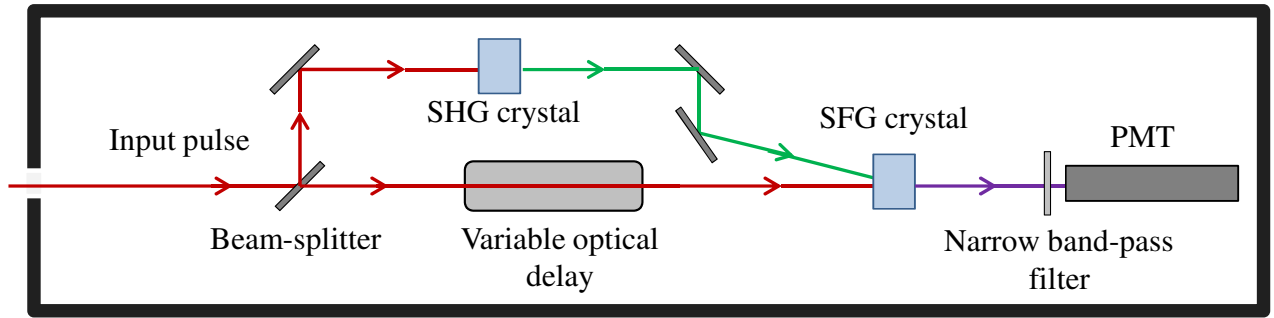


Figure 2.10: Simplified example schematic of a scanning third-order correlator used for measuring the temporal intensity profiles of short laser pulses. A pulse and a frequency-doubled copy are correlated in a crystal resulting in signal from a sum-frequency generation process. The use of a non-identical probe pulse allows for improved resolution and the ability to distinguish between pre- and post-pulses.

The second harmonic pulse has a shorter duration than the fundamental (a factor of $\sqrt{2}$ shorter for Gaussian pulse profiles), and thus can more finely probe its intensity profile. An ideal correlation yielding a perfect reconstruction of a pulse (with no need to apply an approximate deconvolution factor) requires a delta function probe, hence shorter probe pulses are desired. Scattered SHG light, from bulk or surface defects within the nonlinear crystal, a serious issue limiting the dynamic range of autocorrelators, can be reduced to low levels using narrow-bandwidth filters. Consequently, the measurement's dynamic range is limited mainly by the monitoring system's sensitivity and nonlinear crystal damage threshold [75].

2.5.3.3 Extending the capabilities of contrast diagnostics

The ultimate goal for a pulse contrast diagnostic system is high resolution, high dynamic range and single-shot measurement over a large temporal range, capturing few picosecond, ns and 100s of microsecond components of a ‘real’ laser pulse. Unfortunately, these characteristics do not typically come hand-in-hand; single-shot systems, while able to provide relatively high dynamic range measurements with good time resolution, are typically limited to relatively small time ranges. Some examples of these include the systems developed by Wang et al. and Oksenhendler et al.; Wang reported dynamic ranges of $>10^{10}$ over a 50 ps time window [76], while Oksenhendler reported a dynamic range of 10^8 over an 18 ps temporal range [77]. The aforementioned systems only provide measurements of temporal contrast, however it is also of interest to understand if there are any spatial contrast variations, which might map into longer or distorted pulses when focused onto targets. To address this, a Spatio-Temporal Cross-Correlator was developed by Ma et al., capable of measurement over a dynamic range of 10^8 within a 50 ps temporal window, across a 6 mrad angular range [78]. In addition, each of these systems employed Cross-Polarized Wave (XPW) generation to temporally clean the probe pulses [54], which can help prevent observation of ‘ghost’ diagnostic artifact pulses occurring as a result of mixing of the main pulse and post-pulses present on the probe within the SFG crystal. XPW involves an intensity-dependent polarization change as the light pulse propagates through a nonlinear crystal. When combined with a high-quality polarizer, this phenomenon can be used to effectively discriminate between the higher intensity main pulse and low-intensity satellite pulses.

In contrast to most single-shot designs, scanning cross-correlators are well-suited to perform reasonably high-dynamic range measurements on time-scales up to roughly 1-2 ns (close to the main pulse), however the acquisition time for such systems is long and unsuitable for real-time monitoring or on-shot contrast measurement. However, a single-shot cross-correlator was demonstrated by Kon et al. to have achieved a dynamic range of $\sim 10^{12}$ within a time-window of 1 ns via multi-channel acquisition, using 4 channels with independent optical delay lines [79]. Even 1-2 ns is still a relatively short time-window, as ASE pedestals can have >1 μ s

durations, and there is a possibility of pre-pulses occurring earlier in time than 1 ns prior to the main pulse. Extending the performance of scanning cross-correlators to longer time-scales typically increases the required procedure time [80]. Larger temporal ranges are desirable for characterizing the pulse and its noise features, as potentially detrimental ASE pre-pedestals may occur on time-scales of up to hundreds of microseconds.

An alternative technique for improving dynamic range was demonstrated using a plasma-shuttered streak camera to record both the main pulse and ASE background [80]. The plasma was produced by laser-induced breakdown of a rastered fused-silica plate [81]. The plasma shutter and a multi-shot accumulation method allowed the streak camera's dynamic range (typically 100 for a given gain setting) to be extended to $10^7:1$. Furthermore, multi-shot accumulation allowed the signal-to-noise ratio of the ASE background to be increased, while the plasma shutter attenuated the main pulse, allowing simultaneous measurement of both. The shuttering effect, demonstrated to suppress laser pulses [82–85], is a result of the plasma frequency exceeding the laser frequency (as previously discussed in the context of plasma mirrors), meaning that the plasma is highly reflective to the main pulse, greatly reducing the transmitted energy. Further improvements to the system, such as lowering the shutter's ionization threshold, allowed the system's dynamic range to be increased to 10^9 . The main disadvantage of this system is its unsuitability for single-shot applications.

A limitation of cross-correlators arises from the low sum-frequency (third harmonic) signal relative to detector noise levels. The development of an Optical Parametric Amplifier Correlator (OPAC) allowed this to be overcome [41]; by replacing the third-harmonic crystal with an OPA, the signal can be amplified, improving the signal-to-noise ratio and measurable contrast ratio to $>10^{11}:1$. The principle of the OPAC is the opposite of the plasma-shuttered streak camera; instead of suppressing the main pulse to improve the contrast measurement's dynamic range by protecting a high-sensitivity detector, lower-intensity pulse substructures are amplified allowing the entire pulse to be measured with a low-sensitivity detector. The OPA is operated at degeneracy (equal signal and idler frequencies) with the second harmonic light as the pump and the fundamental as the seed to amplify the lower-energy pulse sub-structures. Non-collinear geometry was used to spatially separate the signal and idler beams [41]. A single-shot variant

of the OPAC has also been demonstrated, [86] using a low-sensitivity CCD camera to measure contrast within a 50 ps window with a reported dynamic range of $>10^7$. This relatively large time window was achieved using a large pulse-front tilt. The same device was also tested using optical density filters to partition the CCD image and increase its dynamic range by $>10^3$. Although this improvement has raised the dynamic range to the $\sim 10^{10}$ level typically required to detect plasma-inducing pre-pulses when using laser systems producing intensities of 10^{20} - 10^{22} W cm $^{-2}$ on-target, measurement over a larger time window is required to fully characterize pulse sub-structures.

The contrast diagnostic systems summarized above do not individually fulfill all of the ideal requirements described at the beginning of this section. Hence, for properly characterizing pulse contrast, currently a suite of *complementary* diagnostics is needed, some with high temporal resolution within small time-windows, and others acting as survey instruments of lower resolution but capable of measuring over timescales exceeding 1 μ s. Appropriately filtered and calibrated photodiodes can be used for characterization of contrast over long timescales (ASE pedestals and pre-pulses) [87–89], although for short pulses, the time resolution of even fast photodiodes is insufficient for measuring the duration of typical pre-pulses. However, such instruments are able to provide measurement of the integrated pre-pulse energies, and are sufficient for locating pre-pulses relative to the main peak and characterizing features such as ASE pedestals which have much longer durations than the rise/fall times of fast photodiodes. A key disadvantage of photodiode-based contrast diagnostics however is their susceptibility to laser-induced damage while performing measurements, which can lead to operational inefficiencies and costs related to replacing diodes; a technique for mitigating this issue will be discussed in detail in Chapter 4.

2.6 The Cerberus Laser System

Cerberus is a Master-Oscillator Power-Amplifier (MOPA) type laser system based at Imperial College London, with both ‘long’ and ‘short’ pulse capabilities. It has provided the platform for carrying out the laser-matter interaction experiments described in this thesis, with the exception of the experimental campaign at the Vulcan Petawatt facility described in Chapters 6 and 7. The system has been built over several years by preceding researchers, including Nicholas Stuart, Siddharth Patankar and Damien Bigourd, led by Roland Smith. Cerberus is a hybrid CPA-OPCPA system, where the short-pulses undergo 4-stage high-contrast parametric amplification from ~ 1 nJ to ~ 10 mJ (a gain of $\sim 10^7$), such that the gain obtained from stimulated emission in the final-stage gain-storage amplifiers is limited to $\sim 10^3$. For higher energy single-shot operation of the short pulse beamline, a series of 7, 16 and 50 mm diameter Nd:phosphate glass rods can be used to reach energies > 1 J, at FWHM pulse durations of ≈ 500 fs. The long pulse beamline is a CPA system derived from the same master oscillator as the short pulse, where a diode-pumped Nd:YLF regenerative amplifier and 3 flashlamp-pumped Nd:YLF amplifiers provide the majority of the gain, with a repetition rate of 10 Hz and current pulse durations of ~ 1.3 ns. A series of 25 and 50 mm diameter Nd:phosphate glass rods are also available for single-shot operation with energies exceeding a joule, which can be used for driving shocks or for diagnostic purposes such as X-ray backlighting or Faraday Rotation Polarimetry for measurement of magnetic fields in plasmas [90, 91].

A simplified schematic of the Cerberus front end is shown in Figure 2.11. A mode-locked Nd:glass (phosphate) oscillator producing transform-limited 250 fs, 1054 nm pulses is used to seed both the long and short pulse beam-lines, which use CPA and OPCPA respectively to produce high power laser pulses. In the adjacent high intensity target area laboratory, H147, there is an additional 50 mm Nd:phosphate glass laser amplifier for the short pulse followed by a grating compressor (geometrically identical to H148, the Cerberus front-end laboratory) using dielectric gratings (Plymouth Grating Laboratories, 1740 lines/mm), prior to injection into the primary Cerberus interaction chamber.

At the beginning of this project, the long pulse beam-line was capable of producing 20 J pulses

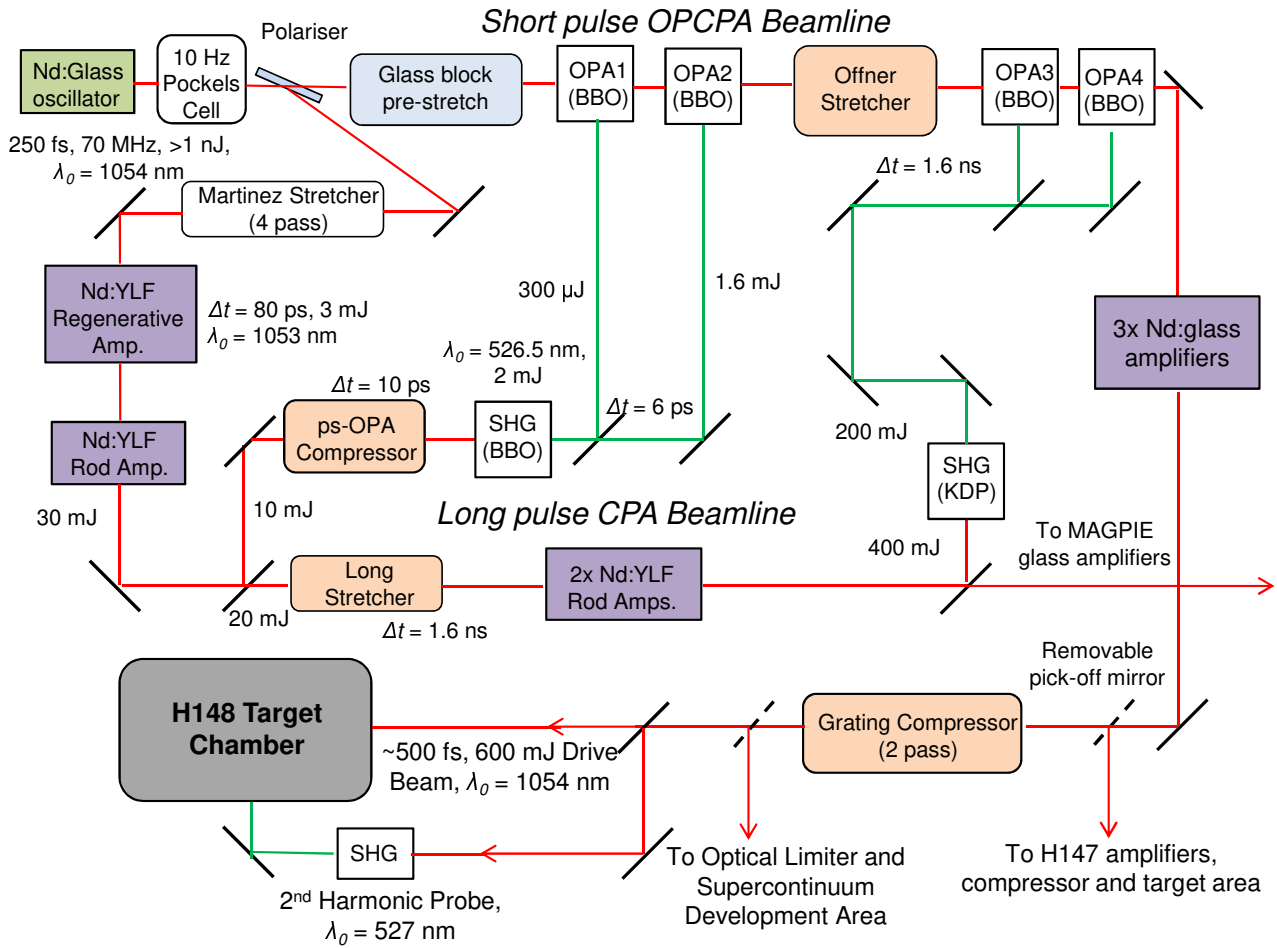


Figure 2.11: Schematic block diagram of the front-end of the Cerberus laser system in H148 at the beginning of the PhD project. The H147 target area now contains an additional 50 mm diameter Nd:phosphate glass laser amplifier followed by a grating compressor geometrically identical to that of H148, where the main laser-front end is located.

at 1.4 ns, with a short-pulse capability of ≈ 600 mJ at < 500 fs, exceeding a terawatt in peak power. These pulses can be delivered to multiple target areas for a range of experiments at terawatt or multi-terawatt peak powers; an experiment studying high-intensity laser-matter interactions with optically-levitated micro-targets [92], the first experiment using Cerberus' multi-terawatt capabilities, will be covered in Chapter 7, with the work reported in this thesis mainly concerning measurement of electromagnetic pulses (EMP) generated by laser-plasma interactions.

Particular attention was paid to pulse contrast within the Cerberus design. The glass power amplifiers on the short pulse line before re-compression are single-passed in order to limit the magnitude of any pre-pulses which might arise from back-scatter or discrete back-reflections in

a double pass geometry, and the multi-stage OPCPA is particularly suited to producing high quality, high contrast pulses in comparison to a conventional CPA system, which might use a regenerative amplifier to provide the majority of the gain. Similarly to the design at Vulcan [61], the picosecond OPA stages are intended to raise the seed intensity at the nanosecond OPAs, which helps to suppress nanosecond time-scale parametric fluorescence. Hence, any contrast-degrading pre-pedestals tend to be limited in time and intensity compared with ASE features of systems using gain-storage amplification exclusively.

2.6.1 Upgrades to Cerberus

One component of this PhD project involved designing and implementing a series of upgrades to the Cerberus laser system. These included some modifications to the spatial shapes of propagating beams using custom apodizers, and the construction and testing of a new Martinez-style stretcher for the long pulse beamline as part of a wider upgrade to the Cerberus long pulse capabilities. This long pulse upgrade is intended to also be used to upgrade the system's short pulse OPCPA capabilities, with designs completed for new OPA stages and ultimately a vacuum compressor system for increasing the maximum deliverable peak power to ~ 40 TW.

2.6.1.1 Beam shaping using serrated aperture apodizers

Spatial beam shaping is important for extracting optimal performance from a high-energy laser system such as Cerberus. Beams with high spatial uniformity, such as 'flat top' and supergaussian modes, are desirable for optimal energy extraction from amplifiers, nonlinear frequency conversion (to minimize reduction of the beam size due to gain-guiding) and for applications requiring uniform target irradiation; this, along with the use of phase-plates for spatially smoothing focal spots, is typically good practice for laser design and is critical for applications such as direct-drive Inertial Confinement Fusion to obtain the desired spherical implosion symmetry with minimal spatial perturbations [93], to name one example.

A beam's spatial intensity profile also affects its propagation properties. Gaussian (TEM₀₀)

beams have very low beam divergences and long Rayleigh lengths, and thus propagate particularly well. Conversely, a flat-top beam's intensity profile will change as it propagates due to diffraction, as it is not a free-space mode. Hence, supergaussian beams must be image-relayed to preserve their spatial intensity profiles if propagated over long distances. A popular technique for improving the spatial intensity uniformity of a laser beam involves apodization followed by spatial filtering.

Spatial apodization involves passing a beam through a modulated or serrated aperture transmission mask (the 'apodizer') which imprints a (typically) periodic high-spatial frequency pattern upon it, immediately before focusing into a spatial filter. After spatial filtering, the resultant beam has a smooth edge profile dependent on the edge profile of the apodizer serrations, but 'softened' by removal of high frequency components in the Fourier domain, as described by Auerbach and Karpenko [94]. In addition to a theoretical explanation of apodizers' effects, they also provide a set of design rules governing the physical characteristics of the 'engineered' periodic serrations. The first design rule sets a constraint on the spatial filter pinhole radius a required to filter out the unwanted high-frequency periodic component of the beam profile arising from the serration pattern, given by [94]

$$a \leq \frac{f\lambda}{2W}, \quad (2.6)$$

where f is the focal length of the optic used to focus into the pinhole, λ is the laser center wavelength and L is the base-width of a serration. The second design rule constrains the height-to-base width ratio of the serrations, where $H/W \geq 6$ must be satisfied for the edge profile of the spatially filtered beam to match the edge profile shape of the serrations, as desired.

Serrated apertures satisfying these design rules are difficult to fabricate mechanically, as a laser cutter with high cutting precision is required. Hence, the apodizers originally used in the Cerberus long pulse line had triangular serrations with a relatively low repeat frequency. However, as shown in [94], spatial intensity profiles with steeper edges can be obtained using Gaussian-tapered serrations, as the edge profiles should match those of the serrations if the

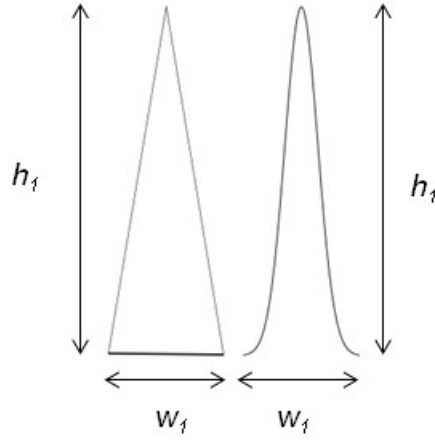


Figure 2.12: Outlines of triangular and Gaussian tapered serrations of equal height and base width, denoted in this case by h_1 and w_1 . The edge profile of the Gaussian serration is steeper.

design rules are followed. As Gaussian serrations have steeper sides than their triangular counterparts for equal serration periodicities, shown in Figure 2.12, they lead to steeper intensity profiles.

A selection of apodizers with 1" outer diameters and Gaussian-shaped serrations were designed using the open-source software *Inkscape* and fabricated by the Physics Mechanical Instrumentation Workshop. These were then inspected using an optical microscope and tested in the Cerberus long pulse beamline, replacing an existing apodizer with triangular serrations. The dimensions of the apodizers were given by the inner diameter D (from the center of the cut-out circle), the serration height H , and the serration base-width W , which was determined by choosing the FWHM of the Gaussian serrations to be 100 μm . Microscope images of the apodizer test pieces are shown in Figure 2.13. Varying the laser's cutting speed has shown that it influences the cut resolution; theoretically, maximizing the cut speed should improve resolution by reducing possible melt-damage. This can be seen from a comparison of Figures 2.13a and 2.13b.

The performance of the Gaussian-serrated apodizers was tested in the Cerberus long-pulse beamline. These were compared against the triangular-serrated apodizers that had previously been in use. Spatial profiles were measured with a CCD camera (Dataray WinCamD series beam-profiler) at various points in the beamline; immediately after the apodizer-spatial-filter combination in question, as well as periodically beyond that point, culminating in a final mea-

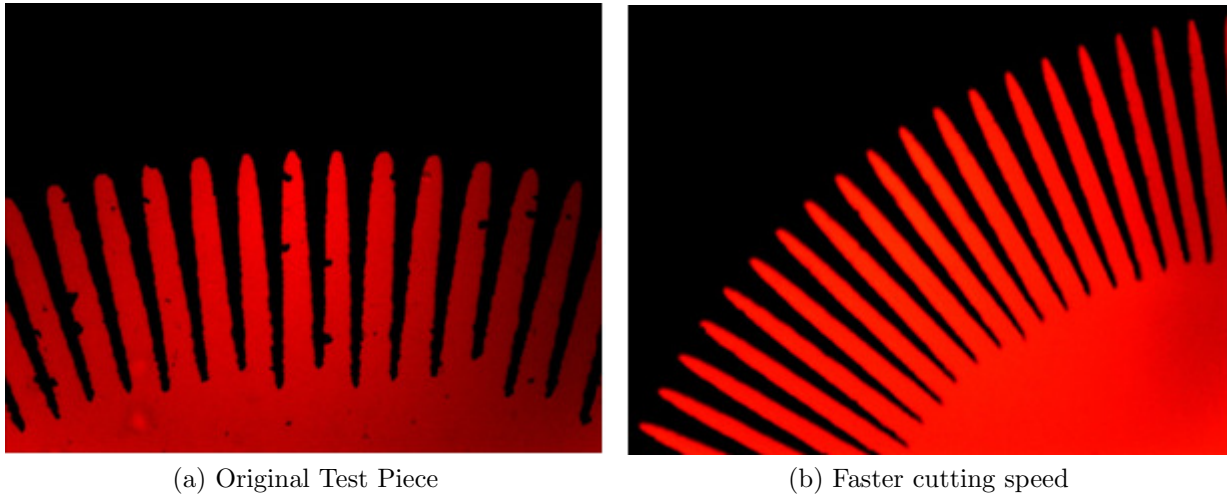


Figure 2.13: Optical microscope images of the Gaussian-serrated apodizers. The test pieces in (a) and (b) were cut at different speeds. The serrations are designed to have heights of 1.3 mm and base widths of 200 μm . The faster cut shown in (b) is more precise, as a faster cutting speed reduces melt-damage and some re-deposition of material, hence increasing the cut precision. I gratefully acknowledge David Williams' (IC Physics Main Workshop) for fabricating the apodizers.

surement before the beam-path to an adjacent laboratory (after ~ 10 m propagation), containing final amplification stages for an imaging line delivered to colleagues in MAGPIE. Figure 2.14 shows the beam profiles of the aforementioned investigation.

The Gaussian-serrated apodizer yielded clear improvements to spatial beam quality, with more uniform intensities towards the pulses' spatial centers and greater fill factors. Furthermore, the pulses' propagation properties were improved, with less ellipticity observed than with the other triangular-serrated apodizers. It is perhaps surprising that the pulses' propagation properties improved as well as the intensity uniformities, considering that flat-top beams are not free-space modes, and that the final ~ 3 m of path before the fourth beam profiling location was not image-relayed. This might be a consequence of the new edge profiles of the beams; the edge profiles from Gaussian apodizers should be also Gaussian, hence the pulses' intensity profiles are closer to a Gaussian mode compared to pulses undergoing beam shaping with the triangular serrated apodizers.

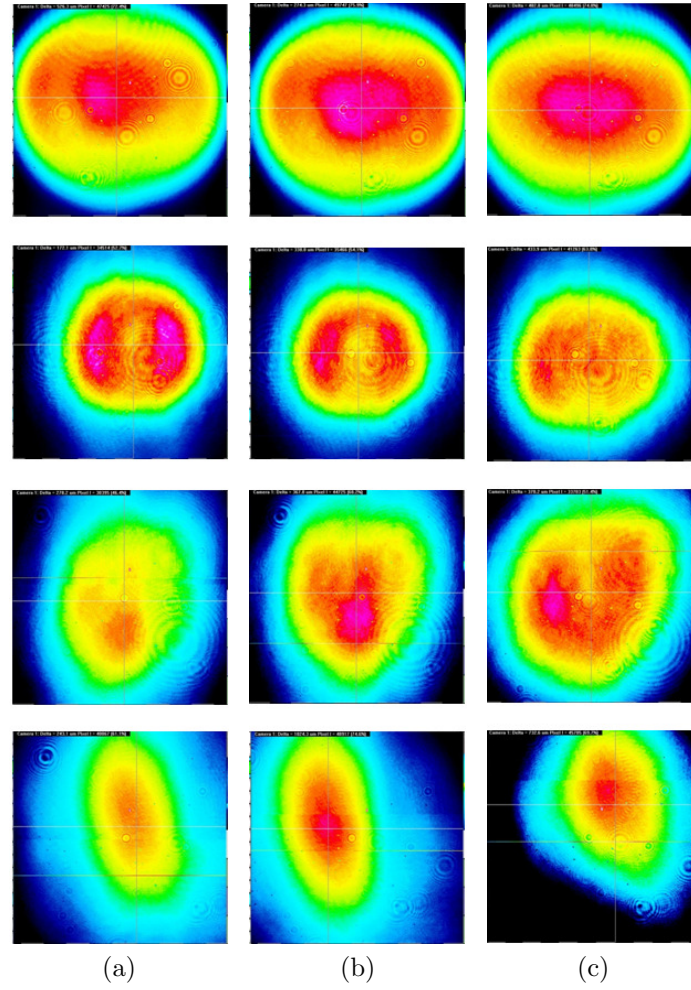


Figure 2.14: Beam profile comparisons at various points in the long-pulse line of laser system, where profiles in a given row are measured at the same location. Each column shows beam profiles obtained with a different type of apodizer before a vacuum spatial filter. Figures 2.14a and 2.14b are for triangular apodizers ($D = 8.5$ mm, $H = 0.5$ mm and $D = 8.0$ mm, $H = 1.0$ mm respectively), whereas Figure 2.14c depicts beam profiles obtained with the Gaussian serrations ($D = 8.0$ mm, $H = 1.3$ mm). The false-colour applied to the images by the Dataray Inc. software represents a linear scaling in intensity (although this is not calibrated to the “true” intensity), allowing easier viewing of the different regions of intensity, with mauve representing the highest intensities and black representing regions without incident light.

2.6.1.2 Long-pulse stretcher

One of the planned capability improvements to Cerberus is an upgrade from a non-imaging dual-grating ‘Z-configuration’ design in the stretcher for the long-pulse CPA line (used for pumping OPAs and long-pulse experiments) to a much more compact folded Martinez-style imaging stretcher design [95], shown schematically in Fig. 2.15b. This has already been shown to

produce ~ 2.6 ns chirped IR pulses via a prototyped stretcher constructed and tested separately from the main laser system.

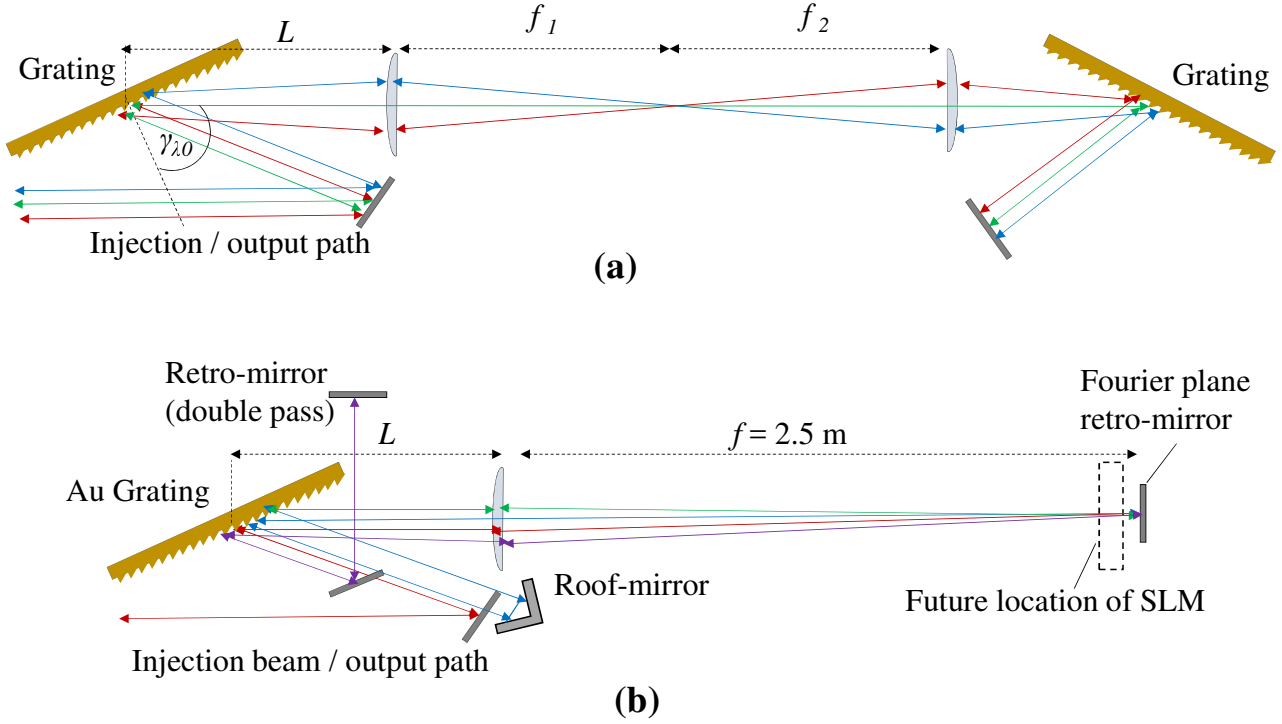


Figure 2.15: Schematics of (a) dual and (b) single grating designs for Martinez-style stretcher systems. The layout shown in (b) has been prototyped and tested alongside the main Cerberus laser system, with space left immediately prior to the Fourier plane for addition of a Spatial Light Modulator (SLM) allowing programmable temporal pulse shaping.

Longer pump pulses will allow the nanosecond time domain OPAs to be pumped more energetically before reaching saturation and yield improvements to bandwidth generation as the wings of chirped pulses are amplified, thus in theory allowing pulses to be compressed to shorter transform-limited durations while also increasing the maximum deliverable energies. As this new design is an imaging stretcher, there is also a true Fourier plane at the location of the retro-mirror. At the Fourier Plane, the spectral variation of the pulse is mapped onto a spatial axis, as the stretcher effectively Fourier transforms the spatial profile of the laser pulse. This allows the spectrum of a stretched pulse to be manipulated, for example by simply using a spectral mask, or a spatial light modulator (SLM) for phase manipulation. As the diffraction grating efficiency is polarization-dependent, phase manipulation can be used to shape a pulse's temporal intensity profile. A Spatial Light Modulator (SLM) has been purchased (Jenoptik SLM-S640 USB), with the intention of incorporating this into the Martinez stretcher, immedi-

ately prior to the Fourier plane in the position indicated in Fig. 2.15b. This capability could be applied to programmable fine-control of OPA pump pulse profiles, as well as to shaping the leading edges of pulses for potential use experimentally as shock drivers.

In a Martinez-style stretcher, the stretched pulse duration, ΔT , per pass through the system is given by [95]

$$\Delta T = \frac{G_{eff} \lambda_0 \Delta \lambda}{c(d \cos(\gamma_{\lambda_0}))^2}, \quad (2.7)$$

where λ_0 is the laser center wavelength, c is the vacuum speed of light, d is the grating groove separation, $\Delta \lambda$ is the bandwidth of the laser pulse, γ_{λ_0} is the diffraction angle (defined from the grating normal) of a ray at the center wavelength, and G_{eff} is the effective grating separation given for a general dual-grating telescoping Martinez stretcher by [95]

$$G_{eff} = [G - 2(f_1 + f_2)] \left(\frac{f_1}{f_2} \right)^2, \quad (2.8)$$

where G is twice the linear distance between the diffraction gratings, and f_1 and f_2 are the focal lengths of the telescope lenses. The Cerberus stretcher uses a single-grating folded design with unit magnification for compactness and to ensure ideal matching of the ‘two’ grating interactions, hence Eq. (2.8) reduces to

$$G_{eff} = 2(L - f), \quad (2.9)$$

where we have used $G = 2(L + f)$, where L is the distance between the grating and imaging lens of focal length f . In Fig. 2.16, a 3D-model of the stretcher prototype built for Cerberus is shown. In order to double-pass the stretcher, the propagation path requires some vertical tilt to obtain vertical separation. This is simply achieved by slightly raising the imaging lens, after aligning the first reflection from the Fourier Plane back onto the input beam path.

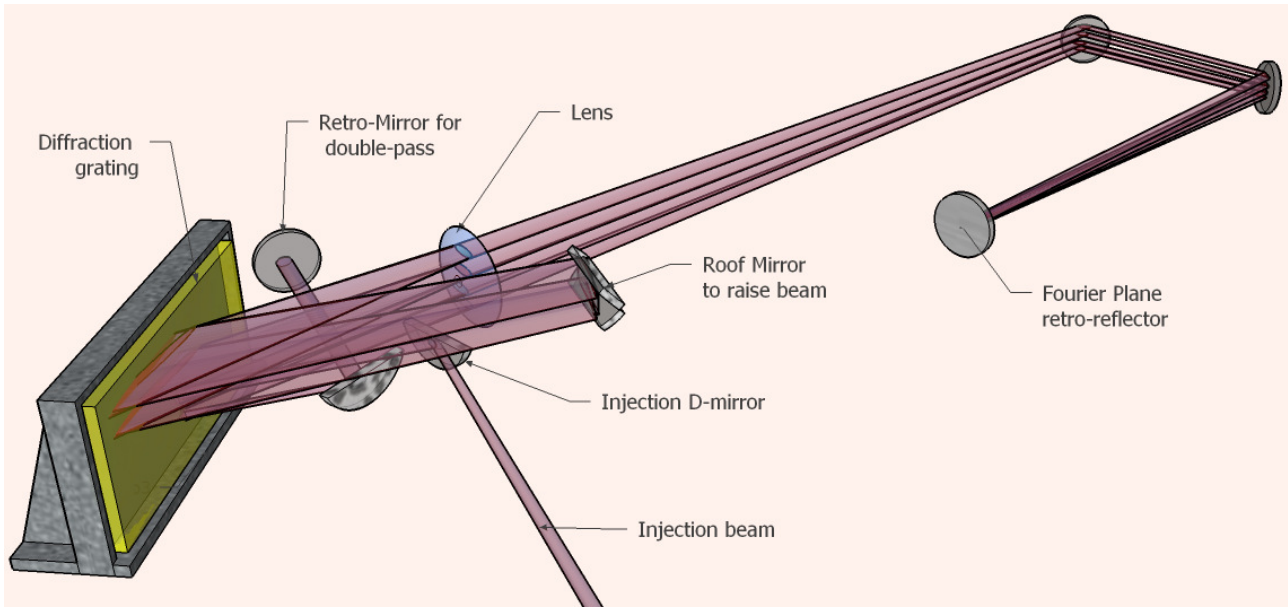


Figure 2.16: Model of the prototype Martinez-style stretcher built for the Cerberus long-pulse beamline, yielding stretched infrared pulses of ~ 2.6 ns FWHM durations. Vertical tilt is added to the propagating diffracted beams by slightly lifting the lens, in order to obtain multiple passes through the stretcher via the roof and retro-mirrors. The propagation path between the lens and the Fourier plane mirror is folded using a mirror pair for compactness.

2.6.1.3 Pump Chain Energy Upgrade

In order to effectively make use of the longer pump pulses made available by the new Martinez-style stretcher design, two additional Coherent Inc Nd:YLF amplifiers (7 and 9.5 mm rod diameters) will be incorporated into the OPCPA pump chain, increasing the available infrared long pulse energy from 0.4 J to ~ 1.2 J. The high-level layout for the upgraded long-pulse beamline is shown in Fig. 2.17, with the new Nd:YLF amplifiers shown in purple and the existing Nd:YLF amplifiers shown in light blue. With this increased energy available in the OPCPA pump chain, additional parametric amplification stages can be added into the system, also operating at 10 Hz and providing a high-repetition rate terawatt peak-power light source, with potential uses alongside liquid-jet and tape-drive targets being developed by colleagues at Imperial. As previously mentioned, the planned incorporation of an SLM into the new stretcher will allow temporal shaping of the OPA pump pulses, which can be used to optimize gain or bandwidth generation as desired via computer control. This proposed upgrade will be discussed further in the next section.

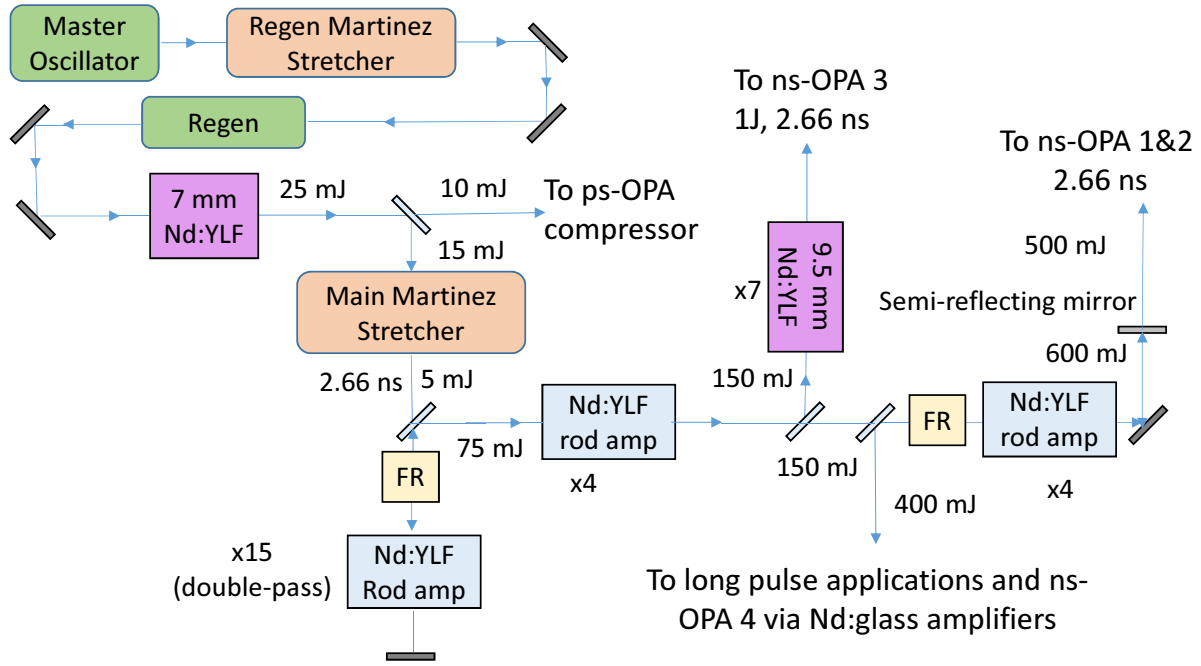


Figure 2.17: A simplified schematic of the upgraded Cerberus long-pulse arm for OPA pumping and long-pulse experiments. More detail about the new main Martinez-style imaging stretcher can be found in Figs. 2.15 and 2.16. Faraday rotators used for double-passing the existing flashlamp-pumped Nd:YLF rod amplifiers are shown as ‘FR’, and the regenerative amplifier has been shortened to ‘regen’. The new 7 and 9.5 mm flashlamp-pumped Coherent Nd:YLF amplifiers are shown in purple.

2.6.1.4 A High-Energy All-OPCPA System

The ultimate reduction of the ASE pedestals of laser pulses, leading to improved contrast over few hundreds of picosecond and longer timescales, is achieved by entirely removing any conventional gain-storage laser amplifiers from the system and replacing them with OPAs, including the final-stage amplifiers. ASE pedestals for relatively high contrast laser systems (such as those using picosecond and nanosecond pump-duration OPAs) typically have a quoted “long” timescale contrast of $\sim 10^6 - 10^{10}$ for the output pulses from their nanosecond OPAs; if final stage amplifiers with a gain of 10^3 are used, the intensity of this long timescale pedestal, *relative to the amplified main pulse*, will be approximately reduced by a factor of 10^3 , as only features temporally overlapping with the pump pulse will be amplified. Due to the high incident intensities, the final amplifiers are typically large aperture components, for example the 50 mm diameter Cerberus final-stage rod amplifiers, hence large nonlinear crystals are required if the rods are to be replaced by an OPA, which are difficult to fabricate. The OPA crystals should also

be sufficiently wedged to ensure minimal interference between any back-reflected component of the pulse and with the part propagating through the crystal, preventing contrast-degrading satellite pulse formation upon re-compression.

Initial work on the concept of large-aperture OPCPA was carried out at the Central Laser Facility (Rutherford Appleton Laboratory), demonstrating practically a high-energy single-shot OPA scheme. Recently, an all-OPCPA system reaching peak powers of 5 PW has been demonstrated [96]; this made use of very large LBO crystals, with final amplifier crystal dimensions of $160 \times 120 \times 10$ mm and $130 \times 130 \times 10$ mm. This is in principle, a similar design to the proposed upgrade to Cerberus, however on a much larger scale, both physically and financially. A high-level schematic of the upgraded Cerberus short pulse beamline, displaying the key components, amplification stages and additions to the pump pulse chain, is given in Fig. 2.18.

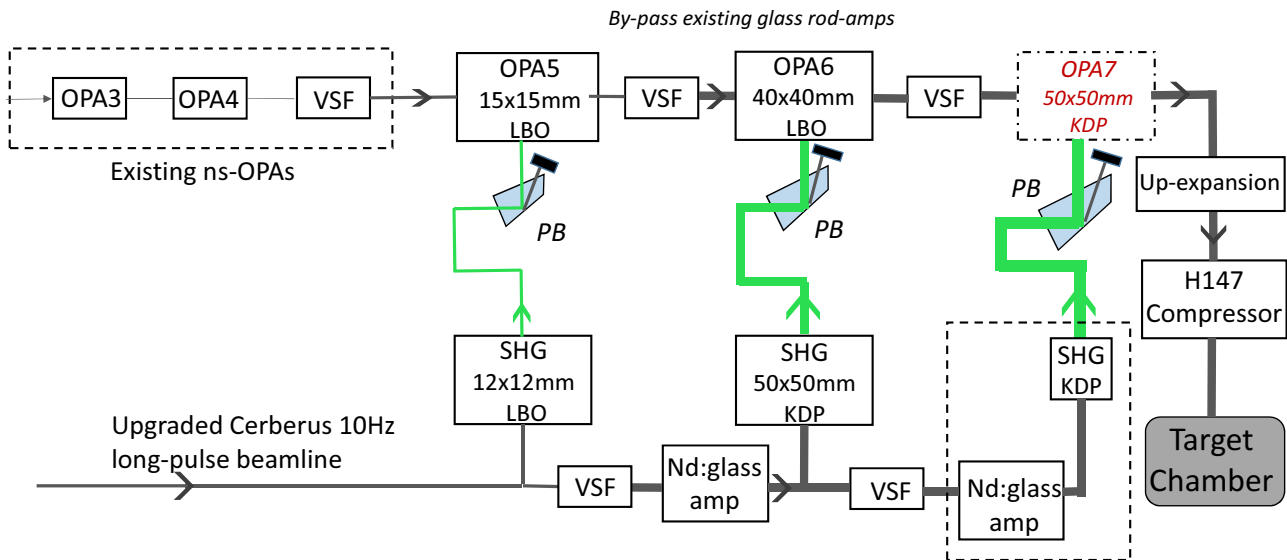


Figure 2.18: Block diagram of the planned upgrade to the Cerberus short-pulse capability. The output of the existing OPAs is up-expanded and amplified by a further two OPA stages, one operating at 10 Hz with a 15×15 mm LBO crystal, and one using a 40×40 mm LBO crystal with a high-energy single-shot pump pulse from the output of a 50 mm Nd:glass rod amplifier. Pellin-Broca (PB) prisms are used to spatially separate the residual infrared component from the OPA pump pulses from the second harmonic. The OPA 7 and associated pump beamline is enclosed within dashed lines, as although the 50×50 mm aperture KDP crystal is available, additional amplifiers and another SHG crystal would be required along with a re-engineered compressor in order to make full use of pulses from a hypothetical OPA 7.

The proposed high-energy OPCPA upgrade incorporates a trio of new LBO crystals and a single KDP into Cerberus; two relatively large LBO crystals (15×15 mm and 40×40 mm aperture)

acting as additional OPA stages, and the remaining LBO and KDP crystals used as frequency doublers for the pump pulses. The 15×15 mm crystal would be pumped at 10 Hz using the upgraded pump chain, producing near-terawatt short-pulse peak powers upon compression, while the 40×40 mm crystal would require sufficiently large pump energies that additional Nd:glass rod amplifiers would be required to provide the necessary gain. Hence, this final OPA stage would only be suitable for single-shot operation.

A set of Pellin-Broca prisms were designed by the author and fabricated by the Optics Workshop (John Murphy); these are suitable for use with large beam diameters (< 70 mm), and allow the residual infrared and frequency-doubled components of the OPA pump pulses to be spatially separated prior to delivery to the amplifier crystals, thus avoiding self seeding of the OPA from residual IR pump light. A pair of dichroic mirrors for harmonic separation and injection of the pump and signal pulses into the OPA crystals is required for each amplification stage. Even with non-collinear OPAs, the propagation distance required to separate residual pump and amplified signal pulses with large spatial extents can be impractical in relatively small-scale university laboratories. With potentially joule-level energies contained within the residual pump pulses, a robust method for safely dealing with these is necessary; dichroic mirrors can provide this capability, though the idler pulse must still be dealt with by steering it into a beam-stop once it is spatially separated from the signal pulse, as in a degenerate OPA scheme it cannot be removed using a harmonic separator.

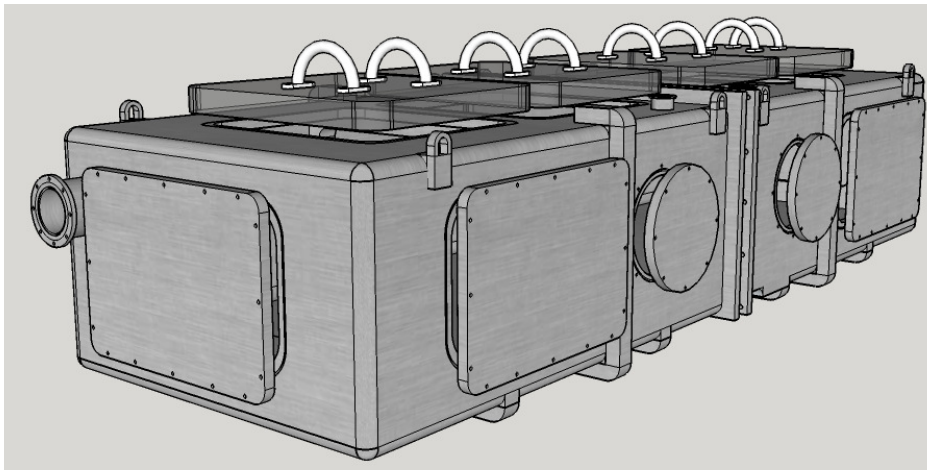
2.6.1.5 Vacuum Compressor Upgrade

Due to B-integral issues arising from pulse propagation through air and a MgF₂ window following compression, the amplified pulse energies available from the OPCPA upgrades are not useful for delivering to experiments in Cerberus' current form. A vacuum compressor system was designed to overcome this problem by removing the propagation path for compressed pulses through air or solid windows. Using this, along with a 6 cm diameter beam size, will enable energies of up to ~ 20 J to be compressed and delivered on target using only reflective optics, corresponding to peak powers of > 40 TW. This energy limit incorporates a factor of 2 safety

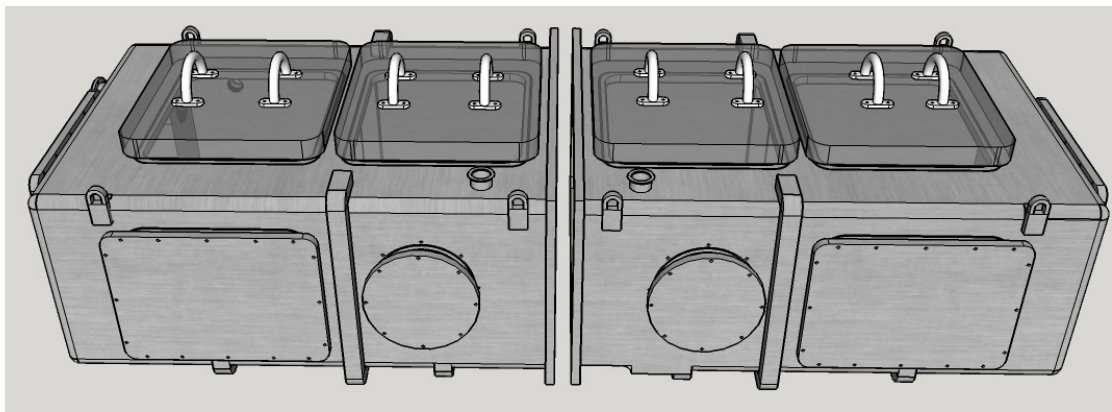
margin, and was determined via damage testing on dielectric grating test pieces with both compressed and stretched pulses.

The compressor chamber was designed using Google Sketchup (free CAD software); some sample images of the vessel and an optical layout are shown in Figures 2.19 and 2.20. The compressor vessel was designed to be constructed from aluminium and to have inner dimensions of $2.80 \times 0.93 \times 0.5$ m (length, width, height), with numerous ports allowing interior access.

This was designed to be a free-standing vacuum vessel, separate from the optical tables where the existing dielectric compressor system is located, and connected to the interaction chamber via a metal tube and vacuum bellows to isolate it from mechanical stresses induced by the frequent pressure changes occurring during an experimental campaign. A split-chamber design, where the two halves seal with a custom O-ring, was chosen to allow easier access into the H147 Cerberus high-intensity target area, with multiple gripping points allowing the chamber halves to be lifted with a crane. Numerous ports were included in the design to facilitate chamber access, both for maintenance and input/output of laser beams, connecting vacuum pumps, pressure gauges, vacuum feed-throughs for in-chamber electronics and any other required equipment.



(a) View 1



(b) View 2

Figure 2.19: Graphics from the Cerberus vacuum compressor CAD model. The vessel is intended to be manufactured from aluminium, split into 2 halves allowing it to be placed in the H147 target area, with 6 cm-thick perspex lids covering the roof access ports.

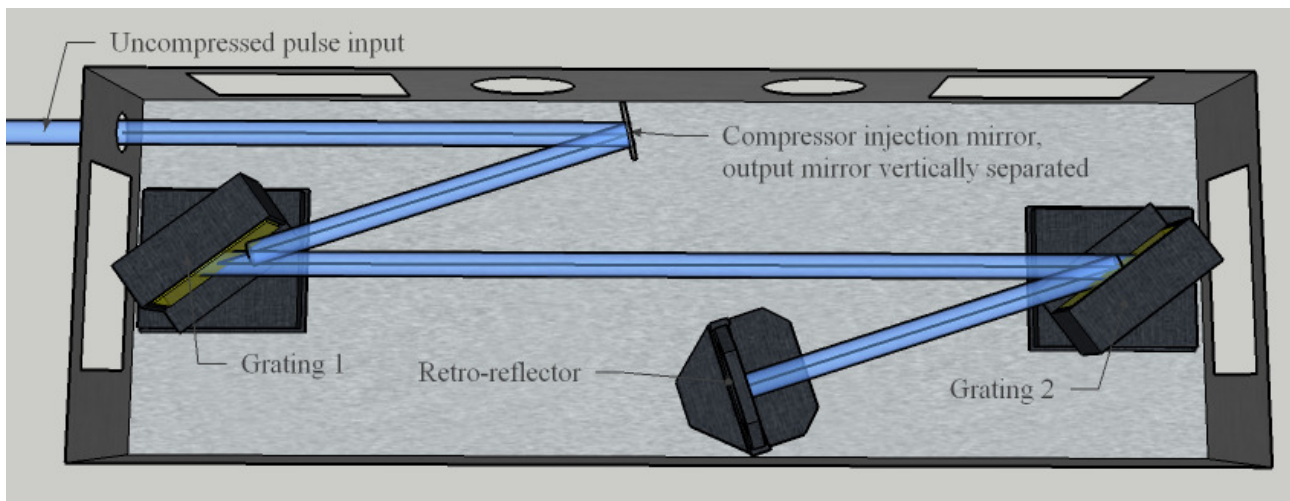


Figure 2.20: Optical layout of the vacuum compressor vessel designed for the Cerberus high-energy upgrade. Vertical tilt introduced by the injection mirror results in the compressed output beam emerging higher than the input, which can then be steered through a convenient port. The locations of the chamber inner walls are shown, along with openings for access ports.

Chapter 3

Laser-Plasma Interactions

All areas of experimental laser-plasma interaction studies are underpinned by theoretical frameworks of light interaction with matter, photon or field induced ionization and optical absorption mechanisms, along with laser-plasma instabilities (LPI) which can substantially affect the behavior of light interacting with plasmas. In this chapter, some theoretical background concerning the interaction of light and plasmas will be discussed to provide context for the experimental work described in the remainder of this thesis.

3.1 Light interaction with matter

In the high-field regime that is typical for laser-plasma interactions, with approximate laser intensities exceeding $10^{13} \text{ W cm}^{-2}$ and where electric field strengths are too high to be treated as perturbative to the atomic electric binding potentials, semi-classical models are usually appropriate to describe the light-matter interaction, rather than the Quantum Optics description, which is more relevant for few-photon interactions. Thus, light is most commonly treated as electromagnetic waves interacting with ionized matter. For dense, relatively low-temperature plasmas with $\Gamma \approx 1$, where Γ is the ratio of potential and thermal energies known as the coupling parameter, quantum effects must be taken into account when describing the plasma. For low values of Γ , the motion between charged particles is highly correlated.

3.1.1 Lorentz Force

The interaction between charged particles and electromagnetic fields is governed by the Lorentz Force $\mathbf{F}_L$, which is given in the non-relativistic case for a single particle by

$$\mathbf{F}_L = q(\mathbf{E} + \mathbf{v} \times \mathbf{B}), \quad (3.1)$$

where q is the particle charge, $\mathbf{E}$, $\mathbf{B}$ and $\mathbf{v}$ are the electric and magnetic field and velocity vectors respectively. For a continuous distribution of charge, the force density $\mathbf{f}$ (force per unit volume) can be written as

$$\mathbf{f} = \rho\mathbf{E} + \mathbf{J} \times \mathbf{B}, \quad (3.2)$$

where ρ and $\mathbf{J}$ are the respective charge and current densities. For a travelling plane EM wave in a vacuum, the electric and magnetic field strengths are related by $B = E/c$ for the non-relativistic case, hence for laser-matter interactions at moderate intensities ($< 10^{17} \text{ W cm}^{-2}$), the magnetic field term is often neglected.

Consider a free electron oscillating in the electric field of a plane wave laser field given by

$$\mathbf{E}_L = \mathbf{E}_0 \sin(\omega t - kz), \quad (3.3)$$

where $\mathbf{E}_0$ is a vector describing the electric field amplitude and direction, k is the wave-vector and z is a spatial coordinate. Neglecting the magnetic interaction term of the Lorentz force and integrating with respect to time gives the electron's oscillatory velocity, which has a maximum value of

$$v_e = \frac{e|\mathbf{E}_0|}{m_e\omega}, \quad (3.4)$$

where m_e is the electron mass. The classical peak kinetic energy, given by $U_k = \frac{1}{2}mv_e^2$, can thus be written as

$$U_k = \frac{e^2|\mathbf{E}_0|^2}{2m_e\omega^2}. \quad (3.5)$$

Alternatively, this can be expressed in terms of the laser intensity I and wavelength λ using the relations $\omega = 2\pi c/\lambda$ and $I = \epsilon_0|\mathbf{E}_0|^2/2$, yielding

$$U_k = \frac{e^2I\lambda^2}{4m_e\epsilon_0c^3}, \quad (3.6)$$

from which it is apparent that the peak kinetic energy gained by an electron in a laser field scales linearly with the laser intensity and quadratically with the wavelength. This has useful implications for the design of laser-plasma interaction experiments, as laser systems operating at longer wavelengths allow higher electron quiver energies to be reached at lower laser intensities. Furthermore, processes requiring overdense plasmas to occur, such as Radiation Pressure Acceleration of ions [97], can be accessed at lower electron densities with longer wavelength radiation. For this reason, there is currently growing interest within the community in high-power, short and ultra-short pulsed laser systems operating in the mid-IR.

3.1.2 Ponderomotive force

The Ponderomotive Force acts along the negative spatial gradients of electric fields - this typically results in electrons being forced into low $\mathbf{E}$ -field regions. A charged particle oscillating in an inhomogeneous electric field will experience different restoring forces within regions of stronger or weaker fields; during the half-cycle in the stronger field region, the force acting towards the weaker field region is stronger than the restoring force experienced during the half-cycle in the weak field region. Hence, there is a cycle-averaged net force acting towards the weak field region, against the gradient of the electric field. As the electric field strengths of laser pulses used in laser-plasma experiments vary spatially, the ponderomotive force is one of the

primary ‘global’ acceleration mechanisms of charged particles (typically electrons) in plasmas. This force is given by the equation

$$\mathbf{F}_P = -\frac{e^2}{4m\omega^2}\nabla(E^2), \quad (3.7)$$

where m is the mass of the oscillating particle, ω is the laser field angular frequency, e is the electron charge and $\mathbf{E}$ is the electric field strength.

The ponderomotive energy U_p describes the time-averaged kinetic energy acquired by a free electron oscillating in the sinusoidal electric field of a laser pulse, given by

$$U_p = \frac{e^2}{2m_e c \epsilon_0 \omega^2} I. \quad (3.8)$$

As ω is related to the laser wavelength by $\omega = 2\pi c/\lambda$, we recover the $I\lambda^2$ scaling for the energy gained by electrons oscillating in laser fields, as previously described in Section 3.1.1. This describes the same physics as in Eq. (3.6), but with a factor of 2 difference between the two expressions as Eq. (3.8) represents a time-averaged kinetic energy gain in sinusoidal fields, rather than the peak kinetic energy within the cycle.

3.2 Important plasma parameters

In this section, some key parameters for describing plasmas and understanding their behavior will be introduced. These include the Debye Length and the Plasma Frequency, which are related to the characteristic spatial and time scales of a plasma. This discussion of plasma parameters is then followed by a simple model treating a plasma as a system made up of distinct electron and ion fluids, within which phenomena such as collective charge oscillations and propagation of electromagnetic waves can be described.

3.2.1 Debye Length

A positively charged ion within a plasma will attract negative charges via electrostatic forces; this results in the accumulation of electrons spatially distributed around the ion, which acts to shield the individual positive charge, as for an observer at a ‘large’ distance, the shielded ion appears to have a net charge of zero. The length over which an individual charge is shielded by the response of the surrounding charged particles, or the length scale over which charge is effectively neutralized within a plasma, is known as the *Debye length* (λ_{De}).

An expression for the Debye length can be found using the following derivation, which follows that of Krueer [98]. An initially motionless charge is placed in a plasma with an initially uniform electron density n_0 , while the ions are treated as a fixed, neutralizing background. Poisson’s equation, $\nabla^2\phi = -\frac{\rho}{\epsilon}$ (where ρ and ϵ refer to the charge density and electric permittivity respectively), is used to determine the electrical potential ϕ , where $\mathbf{E} = -\nabla\phi$. We write Poisson’s equation as

$$\epsilon_0 \nabla^2 \phi = -q\delta(\mathbf{x}) + e(n_e - n_0), \quad (3.9)$$

where the charge is located at $r = 0$ for convenience and n_e is the density of the electron fluid. If we assume an isothermal equation of state, such that the electron pressure p_e is related to the electron temperature T_e by $p_e = n_e k_B T_e$, in the static limit the Lorentz force equation for the electron fluid can be written as

$$n_e e E = -k_B T_e \nabla n_e. \quad (3.10)$$

Since $\mathbf{E} = -\nabla\phi$, the electron density is

$$n_e = n_{e0} \exp\left(\frac{e\phi}{k_B T_e}\right). \quad (3.11)$$

Expanding Eq. 3.11 to first order, assuming $\frac{e\phi}{k_B T_e} \ll 1$, we obtain $n_e \approx n_{e0}(1 + \frac{e\phi}{k_B T_e})$. Inputting

this into Eq. (3.9) yields

$$\nabla^2 \phi - \frac{\phi}{\lambda_{De}^2} = -\frac{q}{\epsilon_0} \delta(\mathbf{x}), \quad (3.12)$$

where by dimensional analysis, we can define the quantity $\lambda_{De} = \sqrt{\epsilon_0 k_B T_e / n_{e0} e^2}$ to be the Debye length. By Fourier transforming and inverting, Eq. (3.12) can be solved for ϕ , given by

$$\phi = \frac{q}{r} \exp\left(\frac{-r}{\lambda_{De}}\right), \quad (3.13)$$

which demonstrates that the electrical potential resulting from a discrete charge exponentially decays with distance over a characteristic length λ_{De} .

3.2.2 Plasma frequency

The plasma frequency for electrons ω_{pe} , the characteristic (inverse) time scale on which collective electron motion occurs within a plasma [98], is given (in SI units) by

$$\omega_{pe} = \sqrt{\frac{e^2 n}{\epsilon_0 m_e}}, \quad (3.14)$$

where n is the electron density and m_e is the electron mass. This frequency determines the response time for oscillating electrons exposed to a particular applied field, and provides a time scale for charge neutralization in the plasma. The plasma frequency plays an important role in the light-plasma interaction, which will be discussed more in Section 3.3.1.

3.2.3 Two-fluid description of plasma

A basic model for describing plasmas is the *Two-Fluid description*, where the plasma is composed of electrons and a single ion species, where each type of charged particle is modelled as an individual fluid. The discussion in this section follows that of Kruer [98]. The fluids are

governed by Maxwell's equations as well as two fluid equations describing each charge species j , derived by taking velocity moments of the Vlasov equation (describing the evolution of collisionless plasmas). These are the continuity and force equations, respectively given by

$$\frac{\partial n_j}{\partial t} + \frac{\partial}{\partial \mathbf{x}} \cdot (n_j \mathbf{u}_j) = 0 \quad (3.15)$$

$$n_j \left(\frac{\partial \mathbf{u}_j}{\partial t} + \mathbf{u}_j \cdot \frac{\partial \mathbf{u}_j}{\partial \mathbf{x}} \right) = \frac{n_j q_j}{m_j} \left(\mathbf{E} + \frac{\mathbf{u}_j \times \mathbf{B}}{c} \right) - \frac{1}{m_j} \frac{\partial p_j}{\partial \mathbf{x}} \quad (3.16)$$

where p_j , n_j and $\mathbf{u}_j$ are the pressure, density and mean particle velocity of fluid j .

3.3 Waves in plasmas

The discussion in this section follows that given in Kruer, Section 1.5 [98]. The two-fluid model allows us to analytically describe a large number of plasma phenomena. An important feature of a two-fluid plasma is its ability to support waves or collective modes of interaction. These waves can appear in many different forms, ranging from the simple case of charge density fluctuations of the electron or ion fluids at a characteristic frequency (electron plasma or ion acoustic waves at high and low frequencies respectively) to hybrid oscillatory modes and magnetosonic waves in the presence of magnetic fields. Most importantly for this thesis, plasmas under the correct conditions can support the propagation of electromagnetic waves externally incident upon them.

3.3.1 Electromagnetic wave propagation in plasmas

The discussion in this section related to the propagation of electromagnetic waves in plasmas follows that of Kruer [98] Section 3.1, and assumes that the plasmas are not subject to large imposed or self-generated magnetic fields. Consider a high-frequency electric field of the form $\mathbf{E} = \mathbf{E}(\mathbf{x})e^{-i\omega t}$, where we assume $\omega > \omega_{pe}$ and the ions are a stationary, neutralizing background

with density $n_{0i}(\mathbf{x})$. Inserting this field into the force equation for the electron fluid and linearizing by neglecting the products of ‘small’ quantities, $\mathbf{u}_e \cdot \nabla \mathbf{u}_e$ and $\mathbf{u}_e \times \mathbf{B}$, gives

$$\frac{\partial \mathbf{u}_e}{\partial t} = -\frac{e}{m_e} \mathbf{E}(\mathbf{x}) \exp(-i\omega t). \quad (3.17)$$

Taking the time-derivative of the current density $\mathbf{J} = -n_0(\mathbf{x})e\mathbf{u}_e$ gives

$$\frac{\partial \mathbf{J}}{\partial t} = -n_0(\mathbf{x})e \frac{\partial \mathbf{u}_e}{\partial t} = \omega_{pe}^2(\mathbf{x})\epsilon_0 \mathbf{E}, \quad (3.18)$$

where $\omega_{pe}^2 = e^2 n_0 / \epsilon_0 m_e$ and the electron density $n_0 = Z^* n_{0i}$ due to quasi-neutrality. Here, Z^* represents the *average* ionization state of the plasma. From this, we integrate Eq. (3.18) and write

$$\mathbf{J} = \frac{i\omega_{pe}^2 \epsilon_0}{\omega} \mathbf{E} = \sigma \mathbf{E}, \quad (3.19)$$

where the coefficients have been written as the plasma’s high frequency conductivity σ . This, along with Maxwell’s equations can be used to derive wave equations for the electric and magnetic field components of an EM wave in a plasma. Assuming $\mathbf{B}$ has the same oscillatory form as $\mathbf{E}$, we write Faraday’s law as

$$\nabla \times \mathbf{E} = i\omega \mathbf{B}, \quad (3.20)$$

and Ampère’s law as

$$\nabla \times \mathbf{B} = \mu_0 \sigma \mathbf{E} - i\omega \mu_0 \epsilon_0 \mathbf{E}. \quad (3.21)$$

We then substitute the expression for σ given in Eq. (3.19) into Eq. (3.21) to obtain

$$\nabla \times \mathbf{B} = -\frac{i\omega}{c^2} \epsilon \mathbf{E}, \quad (3.22)$$

where $\epsilon = 1 - \omega_{pe}^2/\omega^2$ is the dielectric function of the plasma. Taking the curl of Eq. (3.20), substituting for $\nabla \times \mathbf{B}$ from Eq. (3.22) and using the standard vector identity $\nabla \times \nabla \times \mathbf{E} = \nabla^2 \mathbf{E} - \nabla(\nabla \cdot \mathbf{E})$ gives the wave equation for the electric field component of an EM wave as

$$\nabla^2 \mathbf{E} - \nabla(\nabla \cdot \mathbf{E}) + \frac{\omega^2}{c^2} \epsilon \mathbf{E} = 0. \quad (3.23)$$

A similar process, by taking the curl of Eq. (3.22) and using the relation $\nabla \times \epsilon \mathbf{E} = \epsilon \nabla \times \mathbf{E} + \nabla \epsilon \times \mathbf{E}$, yields the wave equation for $\mathbf{B}$

$$\nabla^2 \mathbf{B} + \frac{\omega^2}{c^2} \epsilon \mathbf{B} + \frac{1}{\epsilon} \nabla \epsilon \times (\nabla \times \mathbf{B}) = 0. \quad (3.24)$$

These wave equations can be used to find the dispersion relation for EM waves in plasmas with uniform density. Assuming a spatial dependence of $\exp(i\mathbf{k} \cdot \mathbf{x})$ and using the results $\nabla \epsilon = 0$ and $\nabla \cdot \mathbf{E} = 0$ to write the wave equations in the same form, we find the dispersion relation to be

$$\frac{\omega^2}{c^2} \epsilon = k^2. \quad (3.25)$$

Using the previously provided expression for the dielectric function of the plasma ϵ , the dispersion relation can be written as

$$k^2 c^2 = \omega^2 - \omega_{pe}^2. \quad (3.26)$$

This relation places a limit on the frequencies of light waves which can propagate within plasmas, as for $\omega < \omega_{pe}$, the wavenumber k becomes imaginary, such that light is reflected, with only an exponentially decaying component of the field (the *evanescent* wave) penetrating a short distance into the plasma. In this case, the characteristic response time of the electrons is sufficiently fast to shield out the field of the incident light wave. At the cut-off frequency

$\omega = \omega_{pe}$, the electron density value is the critical density n_{cr} for EM waves of frequency ω . Using the previously given equation for ω_{pe} , we can write an expression for the critical density as

$$n_{cr} = \frac{\epsilon_0 m_e \omega^2}{e^2}. \quad (3.27)$$

For relativistic interactions, the increase in the electron mass as energy is gained needs to be taken into account. Hence, the electron mass becomes γm_e , where γ is the usual relativistic gamma factor $\gamma = 1/\sqrt{1 - v^2/c^2}$, which leads to the following expressions for the relativistic plasma frequency and relativistic critical density (which will be revisited in Section 3.6.3 concerning Relativistic Transparency)

$$\omega_{pe*} = \gamma^{-1/2} \omega_{pe} \quad (3.28)$$

$$n_{cr*} = \gamma n_{cr} \quad (3.29)$$

The critical electron density n_{cr} of a plasma is an important quantity to consider for laser-plasma interactions. Plasmas satisfying $n_e < n_{cr}$ are referred to as *underdense* and can support EM wave propagation, whereas if $n_e > n_{cr}$ the plasma is overdense. The relationship between the critical density for a given ω and the free electron density determines which physical phenomena are likely to occur in a laser-plasma interaction. For example, absorption effects such as resonant absorption, Brunel heating and $\mathbf{j} \times \mathbf{B}$ heating, which are discussed later in the chapter, are all phenomena that occur near the critical surface of overdense plasmas, and can result in much stronger energy absorption of high intensity laser pulses than in the underdense case.

3.3.2 Electron Plasma Waves

The simplest type of wave supported in a plasma is the electron-plasma wave, where the more massive ions can be assumed to be immobile due to the high-frequency nature of the oscillation, with a uniform characteristic density n_{0i} that maintains the plasma quasi-neutrality. Following the discussion presented by Kruer [98], for longitudinal electrostatic waves, the electron motion

is collinear with the wave vector k , hence we can use a one-dimensional treatment. The electron fluid equations are given, in terms of the density n_e , mean velocity u_e and pressure p_e , by

$$\frac{\partial n_e}{\partial t} + \frac{\partial}{\partial x}(n_e u_e) = 0 \quad (3.30)$$

$$\frac{\partial}{\partial t}(n_e u_e) + \frac{\partial}{\partial x}(n_e u_e^2) = -\frac{n_e e E}{m_e} - \frac{1}{m_e} \frac{\partial p_e}{\partial x} \quad (3.31)$$

$$\frac{p_e}{n_e^3} = \text{constant}, \quad (3.32)$$

where we are assuming an adiabatic equation of state such that the wave has a phase velocity $v_{ph} = \frac{\omega}{k} \gg v_e$, where v_e is the electron thermal velocity. Taking a time-derivative of the continuity equation (Eq. (3.30)) and a spatial derivative of the force equation (Eq. (3.31)), then subtracting gives

$$\frac{\partial^2 n_e}{\partial t^2} - \frac{\partial^2}{\partial x^2}(n_e u_e^2) = \frac{e}{m} \frac{\partial}{\partial x}(n_e E) + \frac{1}{m_e} \frac{\partial^2 p_e}{\partial x^2}. \quad (3.33)$$

The electric field can be related to the density using Poisson's equation ($\nabla^2 \phi = -\frac{\rho}{\epsilon}$), such that

$$\frac{\partial E}{\partial x} = -\frac{e}{\epsilon_0}(n_e - Z^* n_{0i}), \quad (3.34)$$

where Z^* is the average degree of ionization of the ions. Next, we consider small-amplitude perturbations in density, velocity and electric field, and re-write these quantities as $n_e = n_0 + \tilde{n}$, $u_e = \tilde{u}$, $p_e = n_0 k_B T_e + \tilde{p}$, and $E = \tilde{E}$. Inserting these into Eqs. (3.32)-(3.34) and linearizing, such that terms containing products of perturbations are neglected, leads to the set of equations

$$\tilde{p} = 3m v_e^2 \tilde{n} \quad (3.35)$$

$$\frac{\partial \tilde{E}}{\partial x} = -\frac{e \tilde{n}}{\epsilon_0} \quad (3.36)$$

$$\frac{\partial^2 \tilde{n}}{\partial t^2} - \frac{n_0 e}{m} \frac{\partial \tilde{E}}{\partial x} - \frac{\partial^2 \tilde{p}}{\partial x^2} = 0. \quad (3.37)$$

The wave equation describing the small amplitude electron density fluctuations is then found by substitution of Eqs. (3.35) and (3.36) into Eq. (3.37):

$$\left(\frac{\partial^2}{\partial t^2} - 3v_e^2 \frac{\partial^2}{\partial x^2} + \omega_{pe}^2 \right) \tilde{n} = 0, \quad (3.38)$$

where ω_{pe} is the electron plasma frequency, given in the previous section, for a plasma with electron density $n_0 = Z^* n_{0i}$. Using a plane-wave trial solution $\tilde{n} \sim \exp(ikx - i\omega t)$, we can find the dispersion relation for electron-plasma oscillations:

$$\omega^2 = \omega_{pe}^2 + 3k^2 v_e^2. \quad (3.39)$$

The second term in Eq. (3.39) is a small wavenumber-dependent thermal correction, though if kinetic effects had been considered in the derivation, a small damping or growth term dependent on the nature of the electron velocity distribution function would also be included.

3.4 Ionization Mechanisms

In most laser-plasma interaction experiments, the target is not initially in a plasma state; it must be ionized by the laser pulse prior to any interaction between laser and plasma. Hence, it is essential to understand the underlying ionization mechanisms involved in a given experiment in order to properly understand the physics, and in the context of laser system design and characterization, relate this to potential pre-pulse mechanisms and timescales. The discussions of ionization mostly follow those presented in Chapter 2 of Mulser's book on high-power laser-matter interactions [99].

3.4.1 Multiphoton Ionization

The primary mechanism for initial ionization of ‘cold’ dielectric materials (with no free electrons) is via the absorption of laser photons by individual atoms and molecules. Typical ionization energies E_I range from 4 to 25 eV [99], with higher values of E_I associated with the inner electron orbitals, hence high-power laser systems which most commonly operate in the infrared (1.17 eV photon energies for 1054 nm wavelengths) cannot directly photo-ionize most materials due to their relatively low photon energies given by $E_{ph} = \hbar\omega$. X-ray, or in some cases ultraviolet, laser systems are necessary for applications involving direct or ‘bulk’ single-photon photo-ionization. However, multiphoton ionization, involving the “simultaneous” absorption of multiple photons, provides a means for producing the initial free electrons which may then act as a seed for further ionization and heating processes. This requires a high spatio-temporal photon coincidence, the likelihood of which is quantified by the instantaneous laser intensity. In the limit of the ionization energy being large compared to the laser photon energy, such that the number of photons N required for multiphoton ionization is relatively large, the dependence on the photon frequency is reduced and thus the process can be described as non-resonant. In this case, the ionization probability P_N reduces to

$$P_N \simeq \sigma_N I^N, \quad (3.40)$$

where σ_N is the generalized cross section for multiphoton ionization and I is the laser intensity. From Eq. (3.40), it is clear that the ionization probability scales both with the number of photons and the laser intensity, as logically expected. Once a ‘small’ number of free electrons are present following photo-ionization, these can act to seed other ionization processes such as collisional avalanche ionization. The threshold intensities for multiphoton ionization typically lie in the range 10^{11} - 10^{13} W cm⁻², depending on the laser wavelength and the target material, and are generally significantly lower for metals. However, breakdown is often observed to occur at lower intensities, which is commonly attributed to the presence of a few seed electrons before reaching the expected multiphoton ionization threshold, or small scale spatial non-uniformities

in the material that at to locally concentrate the electric field. There are a variety of possible reasons for the presence of these seed electrons, such as ionization by stray UV light, or the presence of impurities carrying weakly bound electrons.

3.4.2 Collisional Ionization

Energetic free electrons can collide with other electrons, ions or neutral atoms, imparting energies upon them in excess of the ionization energy, resulting in further ionization. Electrons freed by collisions can in turn result in further collisional ionization, in a process often referred to as avalanche or cascade ionization. This requires a certain free electron density, sufficiently high electron temperatures and a finite time after laser irradiation before becoming significant. Ignoring energy losses from the plasma (via radiation, recombination, thermal conduction via electron diffusion etc), the collisional ionization cross-section σ_I for a single electron oscillating in an electric field and impacting with an ion or atom is given by

$$\sigma_I = 2.8\pi a_0^2 \left(\frac{E_H}{E_I} \right)^2 \frac{\ln(u+1)}{u+1}, \quad (3.41)$$

where $u = (E_e - E_I)/E_I$, E_I is the ionization energy, a_0 is the Bohr radius, E_H is the ionization energy of atomic hydrogen and E_e is the average energy of an impacting electron [99,100]. Using σ_I and assuming a Maxwellian distribution of thermal electron velocities, the mean ionization rate for single electrons ν_I is given by

$$\nu_I = n_i \langle \sigma_I v_0 \rangle, \quad (3.42)$$

where n_i is the ion density the angle brackets denote time-averaging. As this expression is for a single electron, the total ionization rate for the plasma system will thus scale with the density of free electrons, enabling an avalanche process to occur.

3.4.3 Field ionization

Ionization can also occur via the interaction of atoms with strong electromagnetic fields. A theory of nonlinear ionization of atoms in the fields of strong electromagnetic waves was developed by Keldysh, who described general expressions for the ionization probability rates of atoms and solids, where the ionization is dependent on the frequency ω and amplitude F of the laser fields, and the atomic ionization potential I_0 [101]. The general formula for the ionization probability w of atoms by an electron transitioning from the ground state to freedom is

$$w = A\omega \left(\frac{I_0}{\hbar\omega} \right)^{3/2} \left(\frac{\gamma}{(1 + \gamma^2)^{1/2}} \right)^{5/2} S\left(\gamma, \frac{\tilde{I}_0}{\hbar\omega} \right) \times \exp \left(-\frac{2\tilde{I}_0}{\hbar\omega} \left[\sinh^{-1} \gamma - \gamma \frac{(1 + \gamma^2)^{1/2}}{1 + 2\gamma^2} \right] \right), \quad (3.43)$$

where $\gamma = \omega/\omega_t = \omega(2mI_0)^{1/2}/eF$ (ω_t is the frequency up until which electron tunneling is determined by the instantaneous field strength), $\tilde{I}_0$ is the effective ionization potential defined by

$$\tilde{I}_0 = I_0 + e^2 F^2 / 4m\omega^2 = I_0(1 + 1/2\gamma^2), \quad (3.44)$$

$S(\gamma, \tilde{I}_0/\hbar\omega)$ is a slowly varying (relative to the exponential in Eq. (3.43)) function of the frequency and field defined in [101], and A is a numerical coefficient of order 1. In the low frequency, high-field strength limit, where $\omega \ll \omega_t$ and $\gamma \ll 1$, Eq. (3.43) can be shown to reduce to an expression of the form [102]

$$w \propto \exp \left[-\frac{2(2I_0)^{3/2}}{3F} \right], \quad (3.45)$$

which is the well-known probability rate of tunneling ionization through a potential barrier in the presence of an electric field [103]. From Eq. (3.45), it is apparent that the ionization rate increases exponentially with the strength of the field perturbing the atomic potential. In the opposite limit, where the laser fields are high-frequency and of relatively low strengths, such

that $\gamma \gg 1$, the ionization rate can be written in the form

$$w \propto F^{2N}, \quad (3.46)$$

where N is the number of absorbed photons. This expression is clearly of the same form as Eq. (3.40), as laser intensity is proportional to the square of the electric field strength, hence we have recovered the multiphoton ionization rate from the more general Keldysh theory of nonlinear ionization.

3.5 Laser absorption in plasmas

A considerable amount of work has been dedicated to the study of laser absorption in plasmas, a non-trivial topic that can depend on a large number of experimental parameters, such that not all works in the literature are in agreement. Similarly to ionization, an understanding of absorption, and hence how laser energy is coupled into a target material, is essential to laser-plasma research, particularly to applications such as inertial fusion seeking routes towards practical and efficient ignition schemes. In this section, a discussion of the key mechanisms for optical absorption is presented.

3.5.1 Inverse Bremsstrahlung (Collisional absorption)

Following photo-ionization, inverse-Bremsstrahlung (IB) plays an important role in transferring energy from the laser fields to the sub-critical plasma [98]. This is a non-resonant process where energy is absorbed via de-phasing of oscillating electrons driven by the laser fields due to collisions with ions [104, 105], resulting in energy being lost from the oscillating E-fields and transferred to the thermal energy of the plasma. According to Ginzburg [106], the absorption due to IB in a plasma with a linear density profile, Maxwellian electron distribution and scale-

length L is given by

$$f_{IB} = 1 - \exp\left(-\frac{32}{15} \frac{\nu_{ei}}{c} L\right), \quad (3.47)$$

where ν_{ei} is the electron-ion collision rate given by

$$\nu_{ei} \propto \frac{n_{cr} Z^*}{T_e^{3/2}}, \quad (3.48)$$

where n_{cr} is the critical density, T_e is the electron temperature and Z^* is the average ionization state. The efficiency of IB decreases for hotter plasmas due to a strong reduction in ν_{ei} with rising temperatures. For ultra-intense short-pulse laser-plasma interactions, the electron distribution function will diverge from a Maxwellian, instead becoming largely determined by the electron quiver velocities in the laser fields; this modification of the electron distribution acts to reduce the absorption due to IB by a factor of ~ 2 , known as the Langdon factor [107]. Hence, for ultra-intense interactions, the relative importance of IB is decreased, and other mechanisms such as resonant absorption, Brunel heating and $\mathbf{j} \times \mathbf{B}$ heating become more prominent.

3.5.2 Resonant Absorption

Resonant absorption occurs at or close to the critical surface of overdense plasmas irradiated by p -polarized ($\mathbf{E}$ -field polarized in the horizontal plane) laser pulses at finite angles from normal incidence, where the oscillating electric field has a component directed into the plasma along the electron density gradient [108], as shown in Fig. 3.1. This field component drives electrons into the plasma, resulting in formation of electron-plasma waves in a collisionless regime as part of the incident light energy is transferred to electrostatic oscillations [98]. In the region around the critical surface, wave breaking results in damping of the electron oscillations and absorption of the wave energy by the plasma.

The absorption can be maximized for a given laser pulse and plasma density scale length by tuning the angle of incidence θ between the laser and plasma. For resonant excitation of

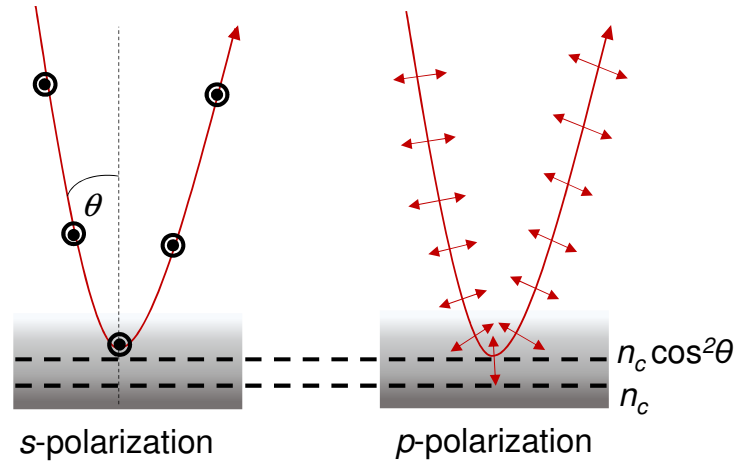


Figure 3.1: Diagram depicting resonant absorption of obliquely incident p -polarized light waves (at an angle θ from normal) at the critical surface with electron density n_c of a laser-plasma. For s -polarized light, the electric field direction is parallel to the planar surface, however for p -polarized light, there is a non-zero electric field component directed along the plasma density ramp, allowing excitation of electron-plasma waves and transfer of energy from the laser fields to the plasma. For resonant excitation of electrostatic oscillations, the fields must tunnel from the reflection surface (at density $n_c \cos^2 \theta$) to the critical surface, hence the incidence angle affecting the balance between the tunneling distance and magnitude of E-field component along the density ramp determines the level of absorption.

electron-plasma waves at the critical surface with electron density n_c , the obliquely incident fields must tunnel from the reflection surface at density $n_c \cos^2 \theta$. For large incidence angles, there is a large E-field component directed along the density gradient, however the tunneling distance is too large for resonant excitation. Conversely, for small angles, the tunneling distances are small but the E-field component directed along the density gradient will be minimal. Hence, an optimum value of θ can be found; this depends on the plasma's characteristic density scale length L (here the distance from the plasma's front edge to the critical surface) and the free-space wavenumber k_0 , and is given by [108]

$$\sin \theta \simeq (k_0 L)^{-1/3}. \quad (3.49)$$

For laser-matter interaction experiments using planar targets, this is achieved by varying the angle of incidence between the target and drive laser; for spherical micro-targets (as described in Chapter 7), this is not necessary as there will automatically be a component of the $\mathbf{E}$ -field at the resonance angle from the spherical surface normal that is optimally matched. Similarly, resonant

absorption can be strongly enhanced for initially planar targets by effects distorting the density profile of the critical surface, such as cratering [109] or rippling [110]. These can occur due to spatial variation in laser intensity profiles, such that p -polarized light is effectively incident over a larger range of angles, allowing the component directed along the density gradient to more efficiently drive electron-plasma waves.

This resonant absorption model is valid for relatively shallow density gradients in the non-relativistic regime, but this can be extended to better fit relativistic laser-plasma interactions by including the relativistic correction to the critical density. However, for ultra-intense interactions, the ion motion in the laser fields and associated changes to the plasma density profile should be accounted for as well as collective electron motion, which makes useful predictions for absorption using simple scaling laws difficult to make in the relativistic case. While beyond the scope of this work, a useful discussion of these effects is given by J.R. Davies in a review of laser absorption in overdense plasmas in the relativistic regime [111].

3.5.3 Brunel Heating

Intense electromagnetic waves obliquely incident on sharp boundaries between vacuum and overdense plasmas with steep electron density gradients can be strongly absorbed by a non-resonant mechanism, commonly referred to as Brunel or vacuum heating [112]. As described by Brunel, electrons are dragged out from the overdense plasma into the vacuum by the oscillating light fields, before being sent back into the plasma after the field direction reverses, with velocities $v \simeq v_{osc}$, where $v_{osc} = eE/m_e\omega$ is the electron quiver velocity under the influence of the EM fields and other symbols have their usual meanings. Since the electric field is shielded within the sharply-bounded overdense plasma, the kinetic energy of electrons re-entering the plasma is lost, resulting in damping of the oscillating EM fields as energy is transferred into the plasma in a relatively small number of energetic or "hot" electrons. In the absence of resonant effects and for a p -polarized wave with electric field E_L incident at angle θ , the absorption

coefficient f is given by

$$f = (\eta/2\pi) \frac{v_{osc}^3}{v_L^2 c \cos \theta} \quad (3.50)$$

where $v_L = eE_L/m\omega$ and v_{osc} is in this case the quiver velocity due to the electric field E_0 normal to the vacuum-plasma boundary, and η is a function of the ratio of the electron thermal velocity v_T and v_{osc} [112]. For conditions where $v_{osc}/\omega > L$, where L is the density gradient length, this absorption mechanism is more efficient than for resonant absorption. As the light intensities are increased, such that $v_L \rightarrow c$, relativistic corrections become necessary. The corrected absorption coefficient f_{rel} is given by Brunel as

$$f_{rel} = (\eta/\pi) c v_{osc} \frac{[(1 + v_{osc}^2/c^2)^{1/2} - 1]}{v_L^2 \cos \theta}. \quad (3.51)$$

3.5.3.1 Vacuum heating

The Brunel model was modified by Gibbon and Bell [113] to include three additional effects, observed in Particle-in-Cell (PIC) simulations, and named *vacuum heating*. The first effect is that of finite radiation pressure on the plasma surface electrons. Secondly, a small but finite underdense region in front of the overdense region from which the incident EM wave can drag out electrons which would have otherwise been shielded from the E-fields in a physically unlikely perfect step-profile plasma. Lastly, not all of the oscillating electrons return to the plasma after a single cycle, with some continuing to circulate in the vacuum region and gain more energy before returning to the plasma and depositing it.

3.6 Relativistic interactions

Comprehensive reviews of relativistic laser matter interactions are given by Umstadter [114] and Mourou et al. [115], and were used to inform the discussion presented in this section.

The relativistic regime for laser-plasma interactions describes a scenario where the velocities of electrons oscillating in intense laser fields become relativistic, or alternatively where their kinetic energy becomes comparable to or exceeds their rest mass-energy. This criterion can be written in terms of the electron quiver momentum p_0 as $p_0 > m_e c$, where m_e is the electron rest mass. The normalized vector potential a_0 , given in terms of the laser electric field amplitude E and angular frequency ω by

$$a_0 = \frac{p_0}{m_e c} = \frac{eE}{m_e \omega c}, \quad (3.52)$$

quantifies a laser-matter interaction's strength and can be used to determine if it is relativistic or not. It is also often useful to express a_0 in terms of the laser intensity I (in W cm^{-2}) and wavelength λ as $a_0 = 0.85 \times 10^{-9} \sqrt{I} \lambda$, particularly for initial estimation of which physical phenomena will be present for experiments with a particular laser system where deliverable intensities and λ are known in advance. For values of $a_0 > 1$, which occur above laser intensities of $\sim 10^{18} \text{ W cm}^{-2}$ at a wavelength of $1 \text{ }\mu\text{m}$, the interaction is considered relativistic.

A rich variety of phenomena accompany relativistic laser-plasma interactions, such as the creation of exotic states of matter at high densities and pressures and new laser-driven heating mechanisms such as $\mathbf{j} \times \mathbf{B}$ heating. The interaction of light with relativistic electrons also leads to relativistic transparency due to the increased mass of electrons as they gain energy, hole boring, and changes to the nature of light propagation with intensity in an analogous manner to conventional nonlinear optics through effects such as relativistic self-focusing. An introduction to these phenomena will be presented in this section.

3.6.1 $\mathbf{j} \times \mathbf{B}$ heating

In the relativistic regime, the $\mathbf{j} \times \mathbf{B}$ term of the Lorentz force becomes larger at high electron velocities and can no longer be neglected when describing the motion of charged particles. The $\mathbf{j} \times \mathbf{B}$ mechanism describes heating of a plasma caused by the electrostatic field driven by the oscillating component of the ponderomotive force of a very intense light wave [116], where

electrons oscillating in the laser electric field are longitudinally propelled by the magnetic field into the plasma. Following the summarized calculation of the ponderomotive potential by Wilks and Kruer [117], the equation of motion for an electron fluid element near the vacuum-plasma boundary is

$$\frac{\partial \mathbf{p}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{p} = -e \left[\mathbf{E} + \frac{\mathbf{v} \times \mathbf{B}}{c} \right], \quad (3.53)$$

where the momentum $\mathbf{p} = \gamma m_e \mathbf{v}$. Rewriting the right-hand side in terms of the vector potential $\mathbf{A}$ and scalar potential ϕ gives an equation for the longitudinal (in the direction of laser propagation) component of the force acting on the electron fluid

$$\frac{\partial \mathbf{p}_L}{\partial t} = e \nabla \phi - m_e c^2 \nabla (\gamma - 1) \quad (3.54)$$

where $e \nabla \phi$ is the electrostatic force and the second term is the relativistic ponderomotive force, where γ depends on the oscillatory momentum of the electrons. From this, the ponderomotive potential U_p is

$$U_p = (\gamma - 1) m_e c^2, \quad (3.55)$$

and can be used to provide an estimate of the energies of the accelerated energies, an effective temperature T_h , assuming the energies gained are due to U_p . An expression for T_h was given by Malka et al. as [118]

$$T_h \approx 511 \left(\sqrt{1 + \frac{I \lambda_\mu^2}{2.8 \times 10^{18}}} - 1 \right) \text{keV}, \quad (3.56)$$

where λ_μ is the laser wavelength in μm , I has units of W cm^{-2} , and the equation is valid for linear polarization states. Similarly to resonant absorption and Brunel or vacuum heating, the $\mathbf{j} \times \mathbf{B}$ heating mechanism involves oscillation of electrons driven by the laser fields near and

through the surface of an overdense plasma. Some of these electrons will be driven across the critical surface as they gain momentum from their $\mathbf{v} \times \mathbf{B}$ motion and thus freed from the laser fields once injected into the overdense region. Unlike ‘classical’ resonant absorption and Brunel heating, $\mathbf{j} \times \mathbf{B}$ heating is equally effective for either *s*- or *p*-polarized laser light. Consequently, the polarization of the incident pulses on a solid target is typically less of a concern at very high intensities due to the increased influence of $\mathbf{j} \times \mathbf{B}$ heating.

3.6.2 Hole boring

For a high-intensity linearly polarized light wave interacting with an overdense plasma (assumed here to be a perfectly reflecting surface), the light pressure P_L is given by $P_L = 2I/c$, where I is the laser intensity and c the vacuum speed of light [119]. For focused intensities in the region of $\approx 10^{21}$ W cm⁻², used during our Vulcan experiment for example (described in Chapters 6 and 7), this results in pressures of ≈ 670 Gbar, and hence a significant hole-boring effect in the target plasma. This leads to time-dependent absorption and generation of very energetic electrons and ions directed into the overdense plasma [119], and enables particles to be directly accelerated by radiation pressure [97, 120, 121].

3.6.3 Relativistic transparency and self-focusing

When electrons are accelerated to relativistic speeds, an increasing proportion of the energy gained contributes to raising the masses of electrons, which increase by the Lorentz factor $\gamma = \sqrt{1 - (v/c)^2}$. As the electron masses increase, their responsiveness to being driven by oscillating light fields is reduced, resulting in a lowering of the perceived electron density $n_{e*} = n_e/\gamma$. Physically, this means that the oscillating electrons do not move sufficiently quickly to shield the plasma from the laser fields. In addition, the surface layer of a laser-irradiated plasma will undergo some expansion, also lowering n_e . The combination of these behaviors results in a relativistic transparency effect when $n_{e*} < n_{cr}$, or if $\omega > \omega_{pe}/\sqrt{a_0}$, in what would typically be considered overdense plasmas [122–124]. Relativistic transparency plays an important role in

certain laser-driven ion acceleration schemes [60, 125, 126], one of which is considered in Section 3.7, generation of short electron bunches [127], and also temporal shaping of intense laser pulses on short timescales [128–132] for improved contrast.

Another important relativistic laser-plasma phenomenon is relativistic self-focusing, where the mass increase of electrons alters their dynamic behaviour in response to driving $\mathbf{E}$ -fields and locally decreases the plasma frequency ω_{pe} . Furthermore, the ponderomotive force will drive electrons away from regions of high light intensity, resulting in plasma electron density redistributions that alter the refractive properties of plasmas. For real laser pulses, there will be some finite variation in the spatial intensity profile, resulting in a spatially varying electron acceleration, meaning that the relativistic mass increases will be non-uniform within the plasma. As the effective mass of electrons governs how they interact with oscillating light fields, it must also therefore affect the refractive index of the medium; a spatially-varying refractive index profile dependent on the intensity of incident light is effectively a lens (which is often positive), and thus this is appropriately termed a self-focusing effect. The critical power for relativistic self-focusing is given by [133, 134]

$$P_{cr} = \frac{m_e c^5 \omega^2}{e^2 \omega_{pe}^2} \simeq 17 \left(\frac{\omega}{\omega_{pe}} \right)^2 \text{ GW}. \quad (3.57)$$

As is the case for conventional self-focusing via the Optical Kerr Effect, relativistic self-focusing of a pulse with a rippled spatial intensity profile will typically result in beam breakup via a filamentation instability [135]. Electrons accelerated within these filament structures produce electric currents which in turn generate strong magnetic fields [136, 137] leading to further redistribution of fast electrons [138] and potentially channeling of laser pulses over relatively long distances as the magnetic fields cause radiation filaments to merge [139, 140].

3.7 Ion acceleration in plasmas

Laser-driven ion acceleration is an extensively studied topic [9], offering the potential to accelerate ions to energies >94 MeV [141] over sub- μm distances, set by the size of the target. These sources also exhibit small source sizes, very low emittance and short burst durations. Due to the short acceleration distances compared to synchrotron or other RF accelerator facilities, laser-based accelerators have potential to be considerably more compact and cheaper. Although not yet a mature technology, there is great interest in development of compact and low-cost ion beam sources for applications such as hadron therapy, proton radiography of plasmas or alternatively in fast-ignition inertial confinement fusion schemes. Much of the research in this field aims to produce low-divergence, monoenergetic ion beams.

The Vulcan Petawatt experimental campaign, described in Chapters 6 and 7, involved an ion-acceleration component, hence it is important to provide some contextual information related to this. In this section, a brief background regarding the common acceleration scheme known as Target Normal Sheath Acceleration (TNSA) is described, along with some schemes relevant to ultra-thin foil targets used during the Vulcan PW experiment, namely relativistic transparency-enhanced ion acceleration and the enhanced TNSA described as the first stage in the Laser-Breakout Afterburner (BOA) scheme. Other ion acceleration schemes include radiation pressure acceleration [97] ('Light Sail'), dipole vortex acceleration [142] and collisionless shock acceleration [143, 144], though these are not discussed in this work.

3.7.1 Target Normal Sheath Acceleration (TNSA)

The most typically used mechanism for describing laser-driven ion acceleration is Target Normal Sheath Acceleration (TNSA) from solid thin-foil targets [145–147]. TNSA can be achieved using typical peak intensities on target exceeding $\sim 10^{18} \text{ W cm}^{-2}$ for laser systems operating at a center wavelength of $1 \mu\text{m}$. Ions are accelerated by a two-stage process; firstly, the electrons are accelerated by the laser and heated to high temperatures, resulting in their partial ejection from the target and the formation of a sheath of electrons at the rear of the target [148, 149].

Following the formation of the sheath, a strong space-charge electric field between the hot electrons and the ions is established, resulting in the acceleration of ions towards the electrons, and then out of the target.

3.7.2 Ion acceleration enhanced by relativistic transparency

Relativistic transparency, as well as the evanescent fields acting on the electron population lying within the skin depth of an overdense plasma, can result in greatly enhanced electron heating, and hence an enhanced TNSA process described in the next paragraph, as the energy transfer mechanisms acting on the plasma electrons shift from surface heating to volumetric heating [141, 150–152]. The Laser Break-Out Afterburner (BOA) acceleration regime, one of the earliest descriptions of TNSA enhanced by relativistic transparency and volumetric electron heating (although strictly speaking BOA refers to energy transfer to ions via a relativistic Buneman instability) [60], also occurs for ultra-thin targets irradiated by high-intensity laser pulses, where the thickness of the target is comparable to the electron skin depth of the overdense plasma at the laser frequency ω_L .

This enhanced TNSA process takes place via a multi-stage process: firstly, an enhanced electron heating effect occurs due to the finite evanescent laser field components still penetrating into the thin target. These evanescent fields are thus able to interact with the bulk of the electron population, resulting in conversion of the cold electron background into hot electrons, and their expulsion into the hot electron sheath at the rear of the target. This expulsion of electrons lowers the density of the target plasma and thus the electron plasma frequency ω_{pe} . Furthermore, the acceleration of electrons to highly relativistic speeds results in an additional lowering of ω_{pe} by a factor of $\langle\gamma\rangle^{-1/2}$, where $\langle\gamma\rangle$ is the average relativistic gamma parameter. This allows oscillating laser fields to propagate further into the plasma and results in the electrons experiencing an additional ponderomotive drive, leading to efficient volumetric heating. The laser then punches through the target plasma as it becomes relativistically transparent. In the specific case of BOA, this is in turn followed by the formation of a very strong longitudinal accelerating field structure via a Buneman-type instability in the plasma, which rapidly grows and acts to reduce

the relative drifts between ions and electrons [60, 126, 153, 154].

As relativistic transparency enhancement requires ultra-thin targets, of order 20-100 nm, it is only feasible with extremely high-contrast laser systems. With insufficient pulse contrast, the pre-ionization of the target will destroy it, rather than merely ablating part of the front surface, as is the case for thicker targets. Hence, it is typically necessary to use one or more plasma mirrors to improve the contrast, adding additional complexity to the drive laser setup. Alternatively, shorter wavelength laser systems could be used (by generating second or third harmonics, for example) such that the electron skin-depths are increased, allowing use of thicker targets.

Chapter 4

Optical Limiters for Sensor Protection

4.1 Introduction to Optical Limiters

The creation of increasingly powerful lasers has prompted considerable interest in the development of new techniques for protecting optical sensors. An optical limiter (OL) is a device which transmits low-intensity light while blocking potentially damaging high intensities, in an analogous manner to an electrical fuse’s blocking of large currents. The ideal case can be seen in Fig. 4.1, where below a threshold optical input energy (assuming the pulse duration and focal geometry are kept fixed) the pulses are perfectly transmitted with zero-losses, whereas light intensities exceeding the threshold are strongly attenuated such that the output energy is clamped to a safe and controllable level.

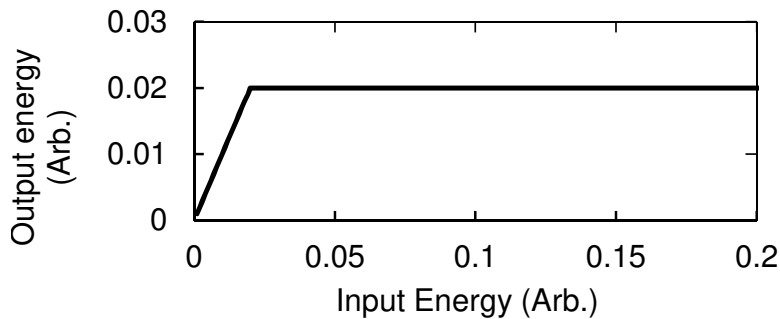


Figure 4.1: The relationship between input and output pulse energies for an “ideal” optical limiter. Below the cut-off energy, pulses are not attenuated, and the output is a linear function of input energy, however above this threshold, the output energy is clamped to a fixed value.

The OL described in this chapter uses the Optical Kerr Effect (self-focusing) in a highly non-linear liquid to provide an intensity-dependent loss upon subsequent spatial filtering. There are numerous OL designs reported in the literature using effects such as Reverse Saturable Absorption [155–158], the photorefractive effect [159], plasma shutters [80] and two-photon absorption [160–162], each with inherent advantages and disadvantages.

Reverse Saturable Absorbers are materials that are transparent at low optical intensities, but increasingly absorbing with increasing intensities due to larger absorption cross-sections from excited energy states. Various RSAs have exhibited suitability to optical limiting applications, including fullerene-doped glasses [163] and xerogel matrices [164] and graphene composite materials [165–167].

Soileau et al. developed an OL based on propagation through a nonlinear liquid (CS_2) and spatial filtering, but this was demonstrated using 40 ps (FWHM) pulses [168], with activation timescales set by the molecular reorientation of CS_2 [169]. The typical response time of the re-orientational Kerr nonlinearity of CS_2 is of the order of one picosecond [170], hence this technique would be ineffective for use with shorter pulses. A medium with a strong response from the femtosecond-timescale electronic hyperpolarizability is therefore preferable for applications with the current generation of high-intensity lasers operating well below 1 ps.

Water cells have been used at the Central Laser Facility (Rutherford Appleton Laboratory, UK) to protect photodiodes measuring long-timescale pulse contrast against high laser intensities [171]. The energy limiting mechanism of this setup was attributed to a partial plasma-mirroring or defocussing effect as the electron density at the laser focus in the liquid approached criticality. However, the energy-limiting mechanism was not further investigated or optimized and the maximum attenuation achieved was limited to a factor of ~ 10 [172]. A similar approach was reported by Sun et al., using a plasma shutter produced by focusing the laser onto a fused-silica plate in order to attenuate the main pulse by factors of 12–14 and protect photodetectors, and was tested using a 1 ns drive pulse [173].

To our knowledge, a damage-resistant system capable of attenuating high power pulses by *several orders of magnitude* on sub-picosecond timescales has not been previously demonstrated.

Using a liquid nonlinear medium ensures the system is ‘self-healing’ and not permanently damaged by focused pulses. This type of OL is desirable for a range of applications including long timescale, high dynamic range, laser pulse contrast measurements, where it can be used to protect sensitive optical detectors monitoring pulse features many orders of magnitude less intense than the main peak, which are typically highly relevant to intense laser-matter interaction experiments, and may require tight focusing of a laser pulse onto a small area photodiode or the entrance window of a photomultiplier tube.

The design described in this chapter draws inspiration from the aforementioned work of Soileau et al., who used CS₂ as the nonlinear liquid [168]. However, as previously mentioned, this system had a picosecond response time, too slow for Cerberus’ ~ 500 fs pulses or even shorter pulses produced by Ti:Sapphire systems, which are increasingly used within the community for driving laser-plasma interaction experiments. Furthermore, CS₂ is a particularly unpleasant chemical to handle, and thus was deemed highly undesirable for use in a routine contrast diagnostic, or mobile test system that could move between different laser laboratories for comparative studies [174].

4.2 The Optical Fuse

An optical limiting setup based on nonlinear effects in liquid media has been developed and characterized, and is shown schematically in Figure 4.2. Constructing an OL to be integrated into a high dynamic range, long time window contrast diagnostic for high-power laser systems poses a problem not encountered in previous work on OL development. In order to detect the low intensity pulse features, the main pulse must have a high peak-power (~ 4 GW). Hence, the optical limiting medium will undergo laser-induced breakdown, so many of the aforementioned OLs would be unsuitable as they would likely be permanently damaged with repeated irradiation. Consequently, a liquid medium was chosen over a solid as it would not be irreversibly damaged by ionization, and thus is said to be ‘self-healing’.

The device is intended for attenuating high intensity laser pulses of ≈ 500 fs durations, as

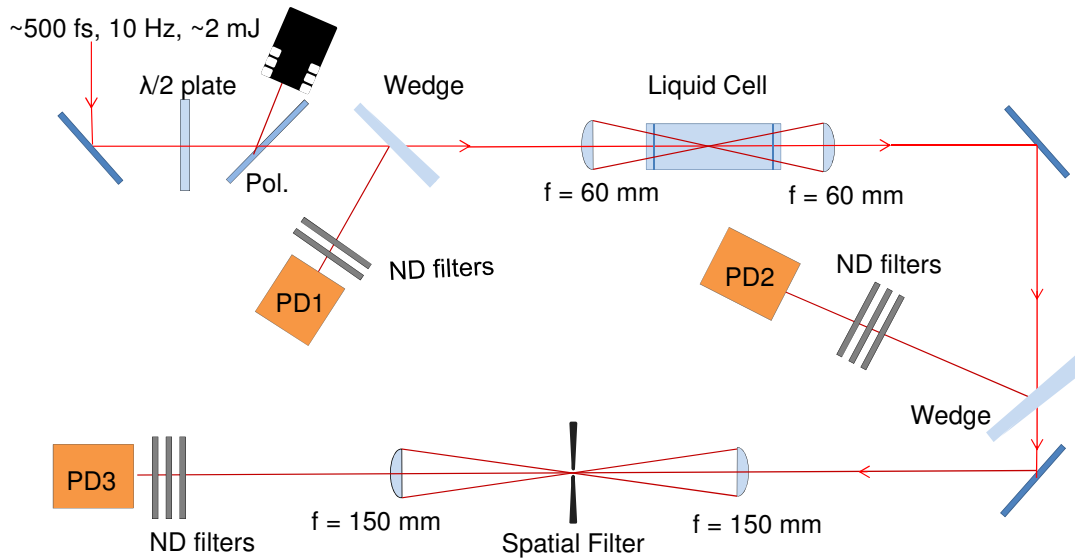


Figure 4.2: Schematic of a single liquid-cell, single spatial-filter optical limiting system for protection of sensitive photodetectors. This simple setup was later expanded upon to include a second liquid cell and spatial filter for improved performance.

provided by the Cerberus 10 Hz OPCPA front-end and similar systems such as the Orion laser at AWE, hence sub-picosecond activation times are required for it to function adequately. Highly concentrated ionic salt solutions are used to enhance the liquid's optical nonlinearities, inspired by the work of T. Jimbo et al., who demonstrated enhanced supercontinuum generation and self-focusing from aqueous salt solutions relative to pure water [175]. Zinc Chloride (ZnCl_2) and a Zinc-Potassium chloride double salt (K_2ZnCl_4) solutions were used at respective concentrations of (3810 ± 90) g/L and (652 ± 7) g/L, along with clean-room grade pure water (de-ionized and filtered) as a comparison. Relatively simple liquids such as the salt solutions are desirable for longevity purposes (compared to large organic molecules for example), as these should not degrade over time due to plasma formation in the liquid; ZnCl_2 or KCl molecules in solution are already dissociated, unlike CS_2 or hydrocarbon molecules that might be used for a similar application. These have been observed to dissociate following multi-photon ionization of inner-shell electrons by 200 fs IR pulses [176].

Zinc Chloride was chosen as the preferred solute, due to its very high solubility (3.95 kg/L in water at 20 °C) compared to ionic salts such as Sodium Chloride (0.36 kg/L at 20 °C) or Potassium Chloride (0.34 kg/L at 20 °C). This allows the liquid's nonlinear susceptibility, which increases with the density of metal ions and hence the salt concentration, to be raised

far above that of pure water or other aqueous salt solutions. In large-scale facilities such as Orion or Vulcan, it is possible to save considerable amounts of money, in both materials and technical staff time, if a diagnostic can be left in place shot-to-shot without need for frequent replacement, and so a long lifetime protection system utilizing relatively non-toxic materials would be distinctly advantageous.

The prototype OL consisted of a wave-plate and polarizer combination for input power control, an 80 mm-length liquid cell and 100 μm diameter spatial filter (SF) pinhole with associated lenses, as well as photodiodes (Thorlabs DET100A) detecting reflections from glass wedges in order to measure the pulse energies at each stage of the system. Multiple flip-mounted calibrated ND filters allowed the photodiodes to measure lower intensity pulses (when removed from the beam path) in order to determine the system's activation threshold and low-intensity response. The lens following the liquid cell was mounted on a translation stage for fine-tuning output collimation, and is necessary due to the refractive index differences between the various liquids investigated. Test pulses were derived from the Cerberus OPCPA front-end, operating at 1054 nm with energies < 2 mJ and ≈ 500 fs FWHM pulse durations.

The refined OL design is shown schematically in Fig. 4.3; this consisted of four positive-positive telescopes, two with liquid cells at focus and two with 50 μm SF pinholes in the focal planes.

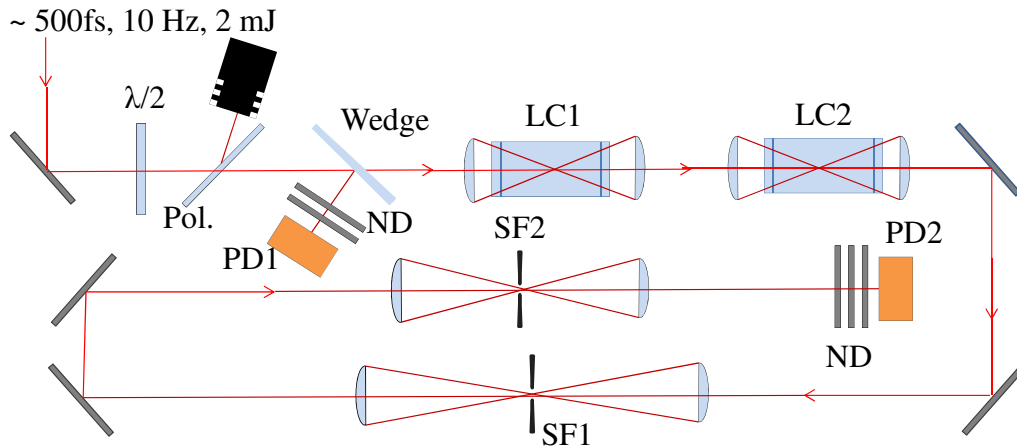


Figure 4.3: Schematic of the optical limiting system using two liquid cells and two spatial filters. The cell-cell, pinhole-pinhole configuration was found to provide higher attenuation than a sequential cell-pinhole arrangement.

For the final OL implementation, ZnCl_2 concentrations of (3810 ± 90) g/L (near saturation)

were used in each cell. Laser pulses (here with durations from ~ 1 ns to < 500 fs) were focused into the liquid cells (LC), where they experienced self-focusing via the Optical Kerr Effect (OKE) if their peak powers exceeded the critical power. Light pulses were then re-collimated and spatially filtered twice through $50\text{ }\mu\text{m}$ pinholes before detection by a photodiode (Thorlabs DET100A). An identical photodiode measuring the reflection from a glass wedge prior to the first LC monitored the input energy. Both detectors were calibrated using a Gentec-EO QE12 energy meter and independently calibrated ND filters.

The SFs were aligned at low power, using nJ-level pulses from the Cerberus 250 fs Nd:glass oscillator to avoid self-focusing. Hence, the liquid cells effectively act as additional lenses for the high power (500 ± 50) fs pulses, moving the focus away from the optimum position required for transmission through the SFs. Conversely, low power pulses can propagate through with minimal attenuation. It is worth noting that a setup such as this *aligned at high-power* could in principle be used for temporal cleaning of pulses; high-intensity pulses experiencing the OKE would have greater transmission through the SF, whereas the lower intensity, contrast-degrading pulse features would be attenuated by the spatial filter, thus increasing the contrast. In practice however, this would have limited utility due to the dispersion and B-integral acquired by short-pulses propagating through the liquid medium and other transmissive optics.

4.2.1 Optical Limiting Mechanisms

Multiple physical mechanisms contribute to the intensity-dependent attenuation provided by the OL. The primary mechanism for short pulses is self-focusing via the OKE in the liquid leading to large losses at the downstream spatial filters. Two SFs were used in series, as this increased both the attenuation and sharpness of the output energy cut-off, approaching near-ideal performance in terms of rapid energy damping above threshold, shown in detail in the next section. The OKE relates to an intensity-dependence of a medium's refractive index n , described by the well-known equation

$$n(\mathbf{r}, t) = n_0 + n_2 I(\mathbf{r}, t) \quad (4.1)$$

where $\mathbf{r}$ and t are spatial and temporal coordinates, n_0 and n_2 are the linear and nonlinear refractive indices, and $I(\mathbf{r}, t)$ describes a laser pulse's spatio-temporal intensity variation. The critical power for self-focusing P_{cr} is given by

$$P_{cr} = \alpha \frac{\lambda^2}{4\pi n_0 n_2} \quad (4.2)$$

where λ is the laser center-wavelength and α is a constant set by the pulse's spatial intensity profile. For a spatially varying laser intensity profile, a spatially-varying refractive index is induced in the material through which the laser is propagating; this is analogous to a Graded Refractive Index (GRIN) lens, hence typically results in focusing or defocussing of the laser pulse depending on the sign of a given material's n_2 coefficient.

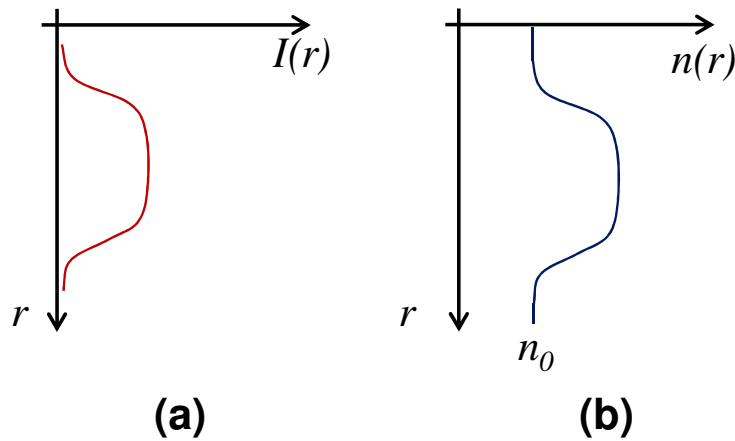


Figure 4.4: Illustration of intensity dependent self-focusing as a consequence of a spatially varying intensity profile of a laser pulse. The spatial intensity variation $I(r)$ of a supergaussian pulse is shown in (a), while (b) shows the resultant refractive index variation $n(r)$ according to Eq. (4.1). This is analogous to a Graded Refractive Index (GRIN) lens, hence results in focusing of the laser pulse.

Self-focusing also contributes to laser-induced breakdown in the focal region followed by plasma formation. The refractive index of an under-dense plasma, where the laser frequency is well above the plasma frequency (relevant conditions for the work reported here), is given by

$$n_{plas} = 1 - \frac{n_e}{2n_c} \quad (4.3)$$

where n_e and n_c are the plasmas free-electron and critical densities respectively [177]. As ionization of the liquid medium is intensity-dependent, a typical spatially varying laser intensity profile with a peak on-axis will result in a reduced refractive index in the central region relative to the edges of the beam, thus acting as a defocussing negative lens. Hence, plasma defocussing is expected to act *against* the OKE, except in media with negative n_2 coefficients. Along with diffraction, these opposing processes can lead to formation of plasma filaments, an active area of nonlinear optics research. Filaments were observed under some conditions, but were not the focus of this work.

Another loss mechanism is absorption by the plasma produced near the focus. The few mJ energies used in developing the prototype OL system we report on are low compared to many large-scale laser-plasma interaction experiments, hence the plasmas at focus are under-dense, allowing processes that typically occur at the critical density surface, such as resonant absorption, Brunel heating or $\mathbf{j} \times \mathbf{B}$ heating, to be neglected. Hence, the absorption was attributed to mechanisms such as inverse Bremsstrahlung. The relatively high transmission through the liquid cells, as will be shown in the upcoming sections, indicates that the plasmas are indeed sub-critical while the driver pulse is present. The presence of the different loss mechanisms was demonstrated using a setup comprising a single cell and spatial filter, with pulse energies monitored after both the LC and the SF; the relative contributions of these different attenuation mechanisms will be examined in more detail in the following section (4.2.2).

4.2.2 Optical Fuse Performance

4.2.2.1 Sub-picosecond pulses

The key characteristic of an optical limiter intended for applications with pulse contrast diagnostics is the ability to provide a strongly intensity-dependent loss to short pulses, on timescales of approximately a picosecond or less, with minimal effect on low-intensity light. In this section, performance results for different liquids in a simple single-cell, single-SF configuration as in Fig. 4.2, with a 100 μm pinhole diameter, are presented, followed by results for setups

modified to maximize attenuation by introducing additional liquid cells and SFs. Three liquids were investigated with sub-picosecond pulses: clean-room grade deionized and filtered water, a concentrated aqueous ZnCl_2 solution, and an aqueous K_2ZnCl_4 double salt solution (some other liquids had been considered in previous work, but these proved to be ineffective [178]). The full-system performances of these liquids within the simple OL setup, where attenuation has been defined here as the ratio of the input energy into the liquid cell and the energy output from the SF, are shown in Fig. 4.5, where it is clear that the ZnCl_2 solution provides the strongest intensity-dependent loss, with approximately $2\times$ better attenuation performance than the next best nonlinear liquid medium, the K_2ZnCl_4 double salt solution. The concentration of the double salt solution was lower than that of the ZnCl_2 solution, which suggests that the optical nonlinearity of the medium increases with salt concentration, as expected. The strong polarization of the metal cations in the salt solution results in a strong nonlinear optical response on the short-timescales required, compared to slower contributions to the nonlinear response such as molecular reorientation or electrostriction.

By monitoring the the pulse energy after the LC (but before the SF), the relative contributions of each stage of the OL to the total attenuation can be compared; this allows inferences about the dominant optical limiting mechanisms to be made. In Fig. 4.6, which shows the variation in attenuation factors with input energy following the LC and SF, the attenuation within the liquid is shown to be much smaller than at the SF, and can be seen to saturate above ≈ 0.1 mJ. Conversely, the SF attenuation appears to grow consistently with increasing drive energy as would be expected if driven by the OKE. This behavior can be understood by considering the timescales and mechanisms for ionization of the liquid contrasted against contribution of the OKE or other defocussing losses arising from spatially varying plasma electron densities.

Following Noack and Vogel's work on photo-ionization and laser absorption in liquid water [179], ionization is initiated by multi-photon absorption at the pulse's leading edge, followed by electron cascade impact ionization after n_e reaches a sufficient level. This rapidly increases n_e , and hence the absorption via inverse-Bremsstrahlung. However, for sub-picosecond pulses, n_e only reaches the level needed for cascade ionization very near the pulses temporal peak irradiance [179]. Hence, the absorption by the plasma only strongly affects the pulse's tail-end

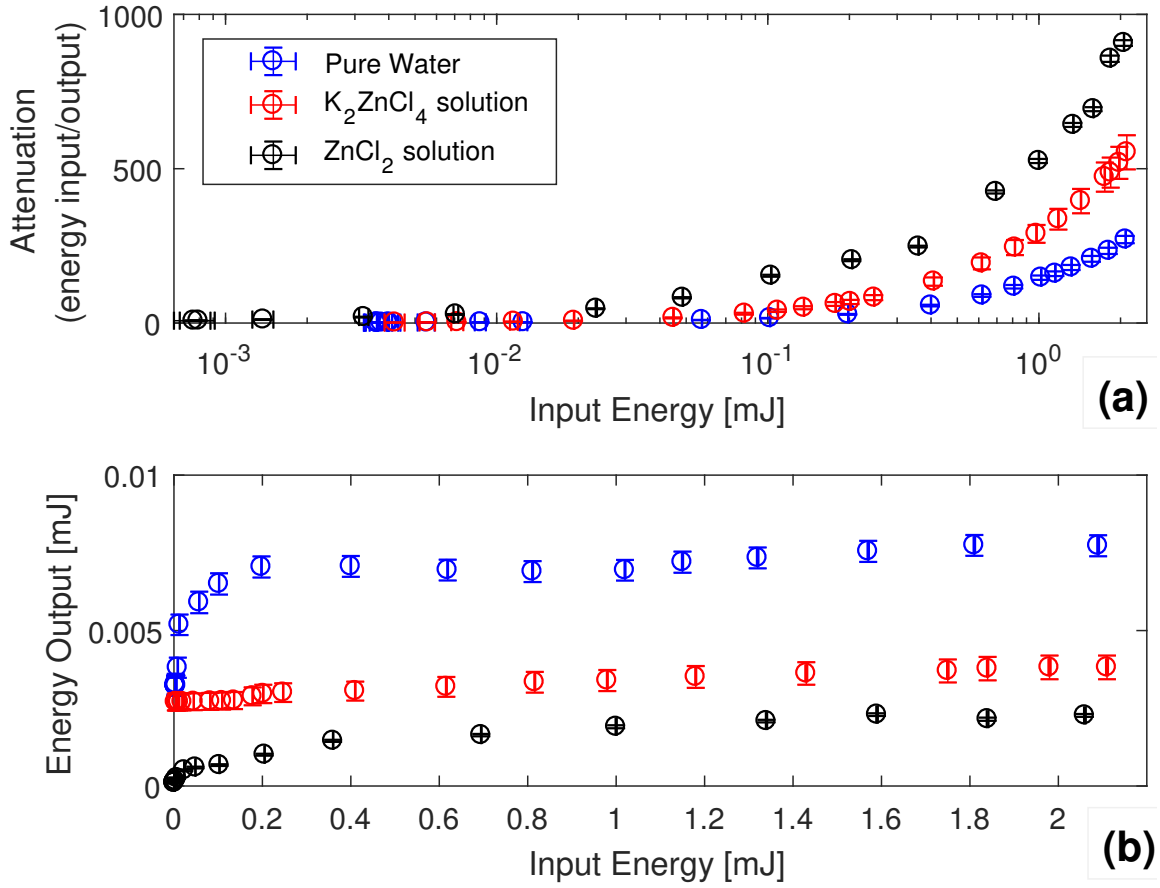


Figure 4.5: Full-system optical limiter performance for a single liquid cell, single SF design using pure water and a K_2ZnCl_4 double salt for comparison with the $ZnCl_2$ as a nonlinear medium. Parameters such as the focusing geometry into the liquid and SF pinhole size were fixed for all of the liquids. The performances are displayed in terms of both (a) the scaling of attenuation (energy input/energy output) with input energy and (b) the output energy for a given input energy, where energy readings presented are 100 shot averages for 10 Hz input pulses. The legend entry applies to both graphs. The displayed energy error bars are found from the shot-to-shot variation in measured pulse energies, while the uncertainty in attenuations is found by addition of the energy uncertainties in quadrature.

and thus does not provide sufficient attenuation for an effective sub-picosecond OL. In contrast to this, the much faster response timescales for the electronic Kerr effect allow short pulses with high peak powers to be efficiently attenuated at the spatial filter following self-focusing within the liquid cell. The observed attenuation saturation within the cell above 0.1 mJ input energies could also be explained by the inefficient increases in n_e on these timescales. Measurements with 1.3 ns pulses yielded much greater absorption and saturation occurring at a $\sim 10\times$ higher attenuation value, as will be shown in Fig. 4.8 in Section 4.2.2.2.

This agrees well with conclusions of [179], where ionization thresholds and absorption coef-

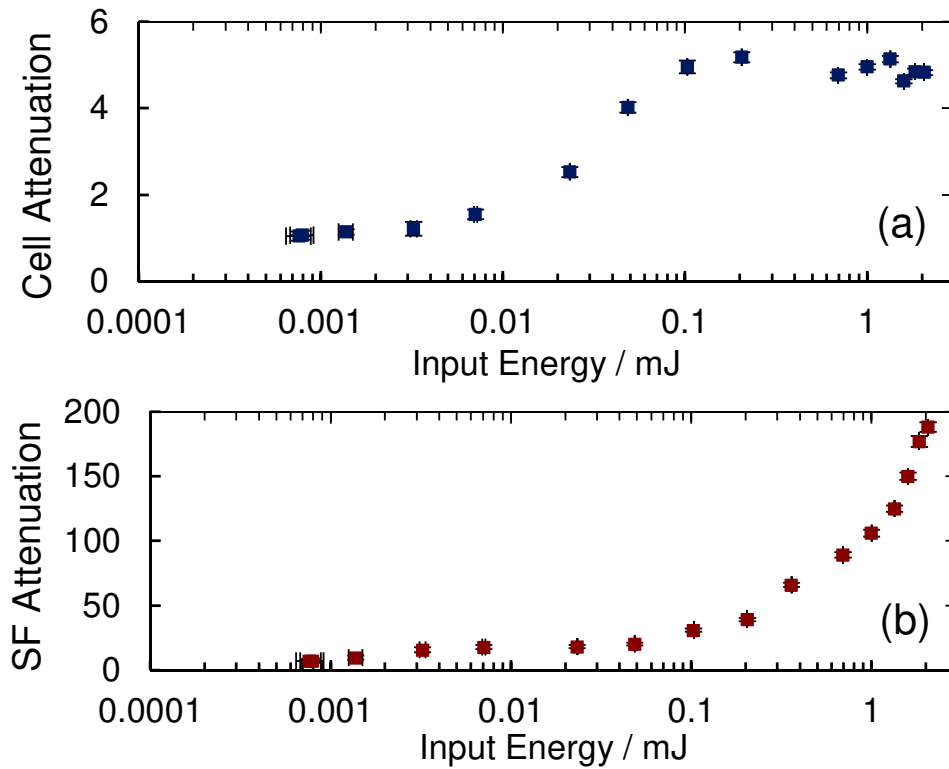


Figure 4.6: Attenuation (input $\div$ output energy) of (500 ± 50) fs 1054nm pulses (a) after a single cell alone and (b) at a single 100 μ m spatial filter. The SF attenuation is the energy ratio of the cell and SF outputs. Absorption in the cell saturates after 0.1 mJ, while losses at the SF continue to increase with input energy. Points shown here, and in subsequent graphs, are 100 shot (mean) averages, with error bars based on the standard deviations in energy added in quadrature to provide uncertainties in attenuation.

ficients were predicted to be lower and higher respectively for longer pulses. The temporal electron density evolution given by Noack [179] also provides an estimate for the time required for recombination processes to lower n_e below a level where post-pulses would be affected. This is of the order of a few nanoseconds, hence a contrast diagnostic using this type of OL would not be able to accurately measure pulse features for a few nanoseconds after the main peak. This is not a major drawback however, as it is the pre- rather than post-pulses that are most problematic in applications of high-intensity lasers, and therefore of most interest when configuring a diagnostic system and using it to assess and refine laser performance.

As a consequence of the main short pulse intensity-dependent loss mechanism of the OL being the Kerr defocussing loss at the SF, the design was modified accordingly to maximize this loss component. The addition of another liquid cell and SF, arranged in a cell-cell-SF-SF series configuration, was chosen such that further Kerr lensing could occur in the 2nd cell *before* the

main energy loss stages at the SFs. If instead a cell-SF-cell-SF configuration were to be used, the Kerr effect contribution in the second cell would be relatively weak, as its energy input would be approximately 2 orders of magnitude lower for ≈ 2 mJ input energies into ZnCl_2 due to the loss factor of ≈ 190 at the first SF. Both SFs contained $50\text{ }\mu\text{m}$ pinholes in the modified design to further enhance selection against a beam experiencing a strong Kerr effect.

The short-pulse attenuation from the refined setup exhibits an even stronger intensity dependence than in the simpler setup, as seen in Fig. 4.7. Low intensity light is slightly attenuated, at a level set by the linear absorption of the ZnCl_2 solution, and high intensities are reduced by up to 4 orders of magnitude in the range of input energies considered, comparing extremely favorably with other OL schemes reported in the literature.

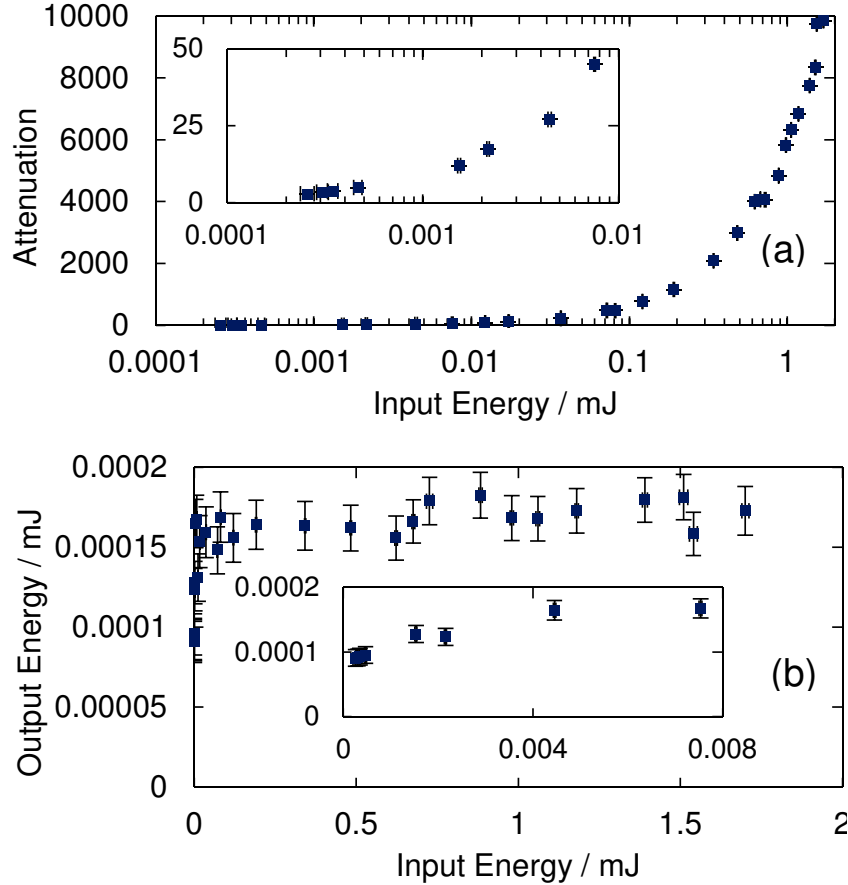


Figure 4.7: Variation of (a) attenuation and (b) energy output for 500 fs, 1054 nm pulses by a setup with two cells and two $50\text{ }\mu\text{m}$ spatial filters in series. All data points are 100-shot averages, with uncertainties in attenuation calculated by addition of the energy uncertainties in quadrature. Above an input threshold of $\sim 5\text{ }\mu\text{J}$, output energies are restricted to $< 180\text{ nJ}$, with the maximum observed attenuation at 1.7 mJ input reaching $\sim 10,000$. The low input energy behavior is shown as insets for clarity.

The OKE is thought to be the main attenuation mechanism in this case, as evidenced by the almost linear scaling of attenuation with energy (and therefore intensity), and the weak absorption by the focal plasma relative to the SF losses in the single cell, single SF case. From Fig. 4.7a, the critical power P_{cr} in the ZnCl_2 solution is estimated to be (0.9 ± 0.1) MW for 500 fs pulses. The energy output from the second SF is limited to < 180 nJ, which should be sufficient to prevent permanent damage to appropriately filtered photodiodes or photomultipliers which do not require a tight focusing geometry. The sharp cut-off in the energy throughput is close to that of an ideal OL, as can be seen by comparison with Fig. 4.1. The > 3 orders of magnitude difference in attenuation between low energy and 1.7 mJ pulses is expected to allow the dynamic range of a contrast diagnostic based on protected photodiodes to be increased by some orders of magnitude, a key aim of this work.

4.2.2.2 Nanosecond pulses

The OL incorporating ZnCl_2 solutions was also tested with nanosecond pulses, using both single-cell single-SF and dual-cell dual-SF (two liquid cells in series followed by two SFs) architectures. This allows the OL's performance to be quantified in a regime of longer pulses with higher energies, though at lower intensities than in the few-mJ short-pulse case.

Although the incident intensities were considerably lower than with short pulses, longer pulses were found to deposit energy more efficiently into under-dense plasmas, as shown in Fig. 4.8, where energy losses within the liquid are much higher compared to sub-picosecond pulses. The performance of the ZnCl_2 solution-based optical limiter is particularly good when compared to other liquids tested, such as deionized water or cyclohexane saturated with C-60 fullerenes [178], as can be seen from Fig. 4.9. The input intensity-dependent attenuation is higher both from absorption by the plasma formed in the focal volume within the liquid and the defocussing loss at the SF, despite a 100 μm pinhole used for the ZnCl_2 solution experiments instead of the 50 μm pinhole used for water and cyclohexane with C-60.

The performance of the dual- ZnCl_2 cell, dual-SF design far exceeds that of its simpler counterpart. The variation in attenuation and output energy with input energy is shown in Figs.

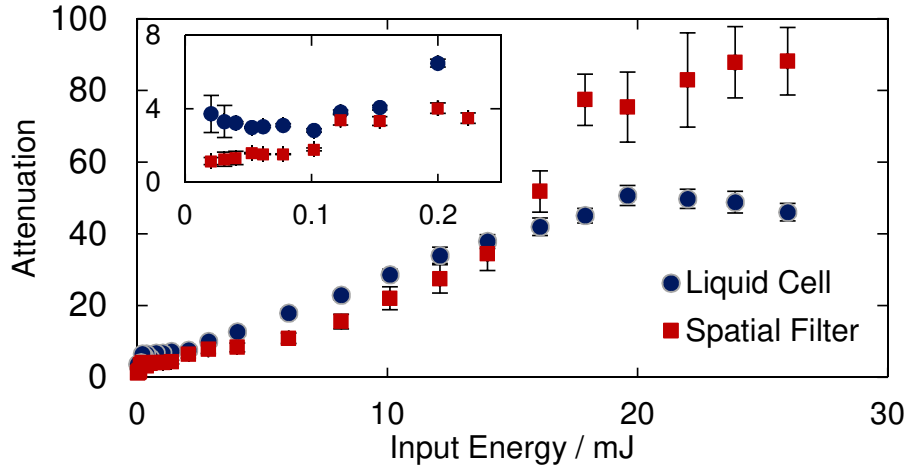


Figure 4.8: Attenuation factors (energy input/output) for a ZnCl_2 solution using 1.3 ns, 1053 nm input pulses at each stage of a single cell, single SF optical limiter. The input energy is defined as the energy input into the liquid cell. The SF attenuation is calculated from the energy incident onto the pinhole and the transmitted energy. Each data point is calculated from energies measured as 100 shot averages, with attenuation uncertainties calculated by addition of the energy uncertainties in quadrature. Attenuation factors at low input energies are shown inset for clarity.

4.10a and 4.10b, with near-total extinction of pulses achieved above a threshold input value of $\sim(110 \pm 40) \mu\text{J}$. The rapid drop in output energy coincides with photo-ionization of the focal volume, suggesting strong absorption and defocussing by the plasma resulting in minimal throughput from the SF pair. The measured uncertainties in output energy due to shot-to-shot energy variation increased substantially near-threshold (Fig. 4.10b), as expected close to the onset of a nonlinear process, which, along with visible self-emission from the focal region, led us to infer that this is the threshold for photo-ionization and the initiation of collisional avalanche ionization. If the OKE was dominant mechanism for both pulse durations, the SF attenuation for the considerably less intense 1.3 ns pulses would be expected to be much smaller than observed (Fig. 4.8). Hence, we conclude that plasma absorption and defocussing plays a greater role in the nanosecond regime, where inverse Bremsstrahlung is expected to be more efficient under conditions of larger free-electron densities; these are expected from longer duration pulses due to the increased prominence of collisional ionization on nanosecond timescales. This results in highly effective operation of the OL at both “long” and “short” timescales via these complementary attenuation mechanisms.

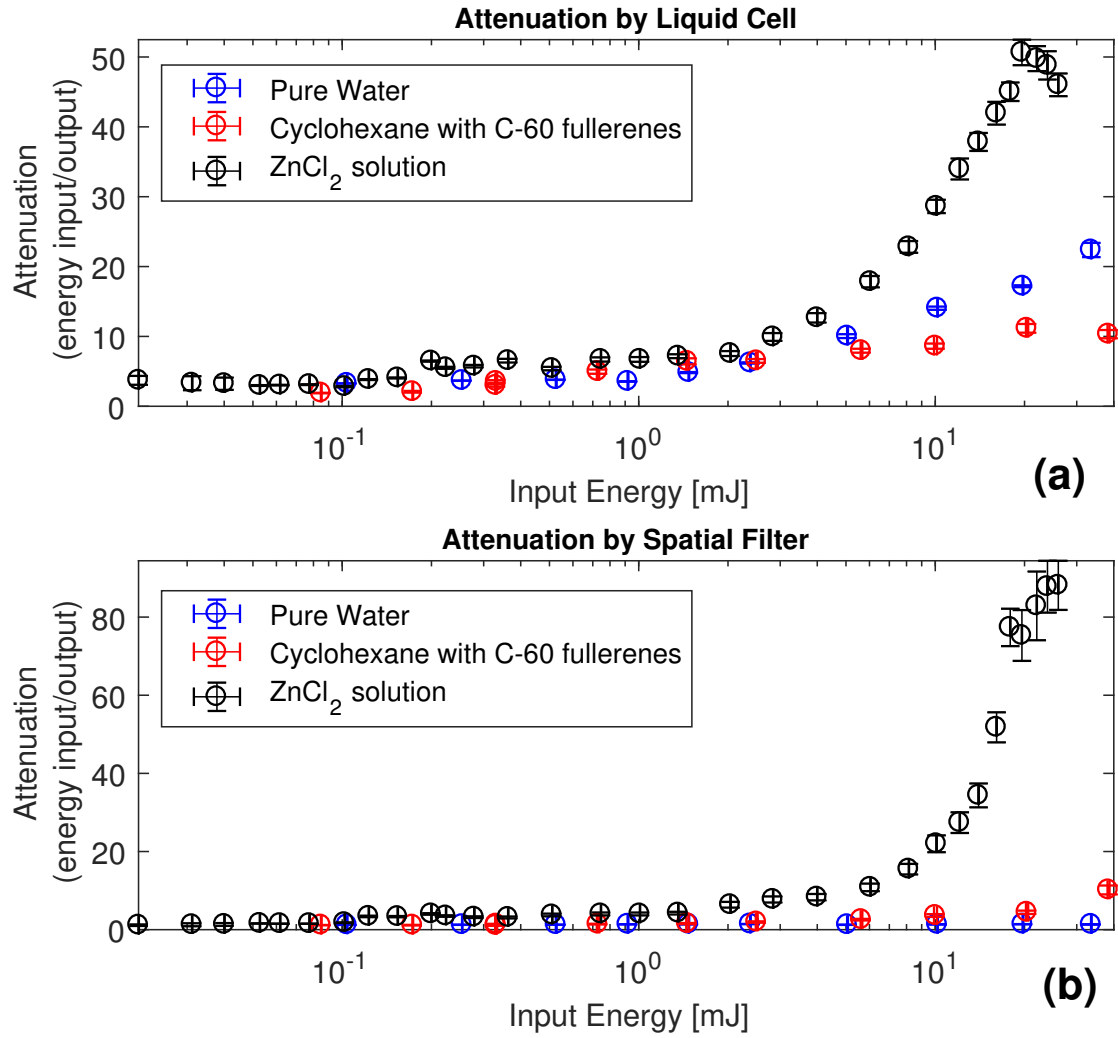


Figure 4.9: Single cell, single spatial filter optical limiter performance comparison between different liquids for 1.3 ns drive pulses. The attenuations at a given input energy are shown for (a) the liquid cell (energy input/output) and (b) the spatial filter for 3 different liquids. Each data point is calculated from energies measured as 100 shot averages, with attenuation uncertainties calculated by addition of the energy uncertainties in quadrature. Note that a 100 μm diameter SF pinhole was used for the ZnCl_2 solution instead of a 50 μm pinhole, which was the case for water and cyclohexane.

4.2.3 Summary of optical limiter development

This work has demonstrated an optical limiter based on liquid cells filled with a highly concentrated ZnCl_2 solution and a pair of spatial filters, capable of strongly attenuating high intensity sub-picosecond laser pulses by factors of up to 10^4 , while transmitting low intensities with low loss and linear attenuation behavior. The motivation for developing an optical limiter of this type arises from the need for protection of sensitive detectors in long-timescale photodiode-based pulse contrast diagnostics used in high-power laser facilities, such as Orion, Vulcan PW

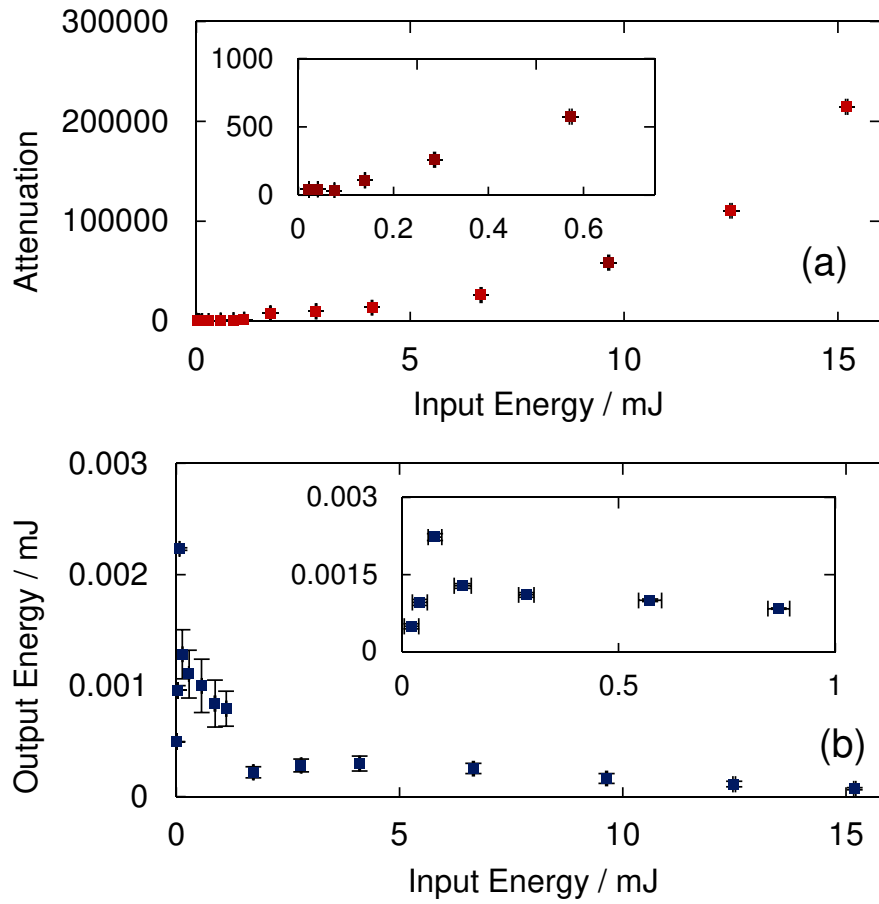


Figure 4.10: Variation of (a) attenuation and (b) energy output with input energy for an OL with two ZnCl_2 cells and two $50\ \mu\text{m}$ spatial filters in series driven by 1.3 ns, 1053 nm pulses. Each data point is calculated from energies measured as 100 shot averages, with attenuation uncertainties calculated by addition of the energy uncertainties in quadrature.

and others. The device's minimum activation time is set by the timescale of the electronic Optical Kerr Effect, on the order of femtoseconds and considerably faster than comparable devices. In addition, nanosecond pulses are attenuated extremely effectively, although primarily through plasma absorption and defocussing, and thus the optical limiter could also be applicable to sensor protection against long pulses. Furthermore, as the nonlinear medium is an ionic salt solution, it is not permanently damaged by ionization at focus. Hence, the system's damage threshold is limited only by damage to the supporting optics. These features make the setup highly suited to deployment as part of a long timescale, single-shot, contrast diagnostic for high power lasers.

4.3 A Long-Timescale Pulse Contrast Diagnostic

To overcome the time-window limitations of diagnostics using nonlinear wave mixing for characterizing pulse contrast over long-timescales, measurements are typically made using an array of differentially-filtered photodiodes (PD), each monitoring particular intensity ranges, connected to an oscilloscope. Setups such as these are capable of measuring on timescales of hundreds of picoseconds/nanoseconds up to seconds, depending on the chosen time-resolution and time-response of the photodiodes. Photodiodes with fast time responses require small active areas to minimize the junction capacitance and charge carrier transit time; hence, the laser must be focused onto the detector for sensitive measurement of low-intensity features. This, however, greatly increases the risk of damage from the pulse's high intensity peak; this may manifest as degradation in sensitivity over multiple shots, or as a catastrophic single shot failure.

Although replacing a diode can be relatively cheap, lost operational and technicians' time and their associated costs can rapidly grow, while instruments also typically require careful recalibration. As measurement of very low intensity features requires the main pulse peak to be of much greater intensity at the detector, this limits the useful dynamic range of a simple diode array. However, by incorporating a fast-response optical limiter into the diagnostic, detectors can be reliably protected from being damaged by the high-intensity peak. The OL described in the previous sections of this chapter can be used for this purpose.

4.3.1 Proposed Diagnostic Setup

A prototype design for a contrast diagnostic, protected from the intense main peak of the test pulses using the dual-cell dual-SF ZnCl_2 OL, is presented in this section. Above a threshold input pulse energy of $\sim 5 \mu\text{J}$ and peak power of $\sim 10 \text{ MW}$, the OL setup limits the energy throughput of the second SF to below 180 nJ . The layout of the device is shown in Fig. 4.11.

Multiple detectors (Thorlabs DET10A photodiodes) with differential neutral density filtering are placed downstream of the OL for measurement of the low-intensity leading edge of the laser pulse with minimal risk of damage. An InGaAs photodiode (Thorlabs DET10N) was also

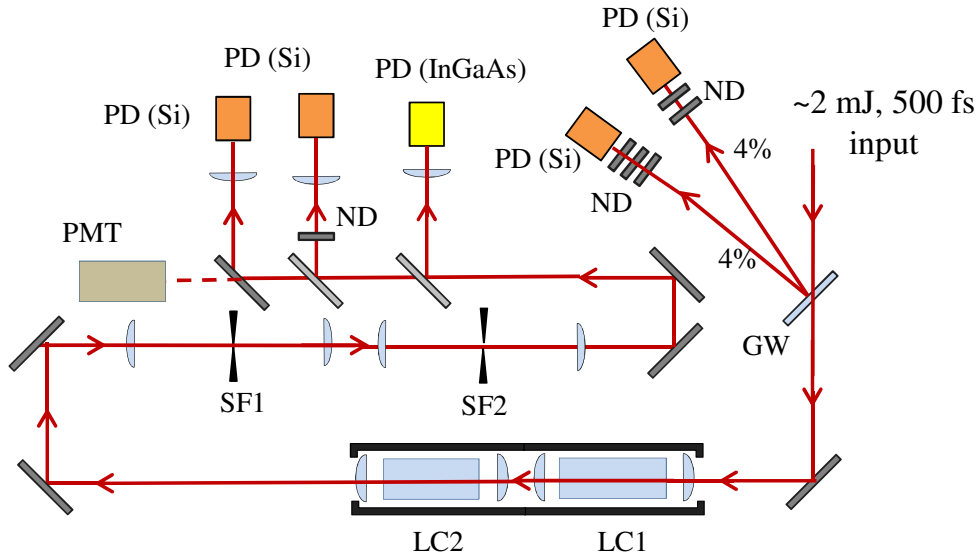


Figure 4.11: Schematic of the prototype long-timescale contrast diagnostic using the ZnCl_2 -based optical limiting system for sensor protection. Multiple photodiode channels are required for high dynamic range measurement, as each is individually capable of measuring over a dynamic range of $\sim 10^2$ limited by a “standard” 8 bit digital oscilloscope channel. An InGaAs detector with 5 ns rise-time is included for higher sensitivity measurement, at the expense of time resolution.

included in the setup design; although the ~ 5 ns rise-time of this diode is long compared to a DET10A (~ 1 ns), the loss of time resolution is offset by $\sim 45\times$ higher sensitivity to 1054 nm light, the operating wavelength for Nd:glass lasers, which are common for high-energy picosecond pulse duration systems. The diagnostic’s time-resolution can be most simply improved by using faster detectors (EOT InGaAS photodiodes with < 50 ps rise times are available, although relatively costly), although for time-resolved measurements of discrete pre-pulses, even an upgraded diagnostic would remain unsuitable. An uncoated glass wedge, with reflectivity of $\sim 4\%$ from each surface, allows the intensity of main peak of the pulse to also be monitored over a few orders of magnitude, and provides a reference for the input pulse energy.

Photodiodes attached to an 8-bit oscilloscope are capable of measuring over dynamic ranges of $\sim 10^2$, hence multiple detection channels are required for high dynamic range measurement; this could be achieved via placing varying levels of filtering on each diode, such that each monitors a particular intensity window. Of course, with a 12-bit oscilloscope, for example, the number of channels needed for a given dynamic range measurement is reduced; this is an important consideration for ultra-high dynamic range diagnostics measuring over a $> 10^{12}:1$ intensity

range, particularly for the next generation of ultra-intense laser systems, where long-timescale contrasts of 10^{12} are likely to be required to ensure ASE pedestal intensities remain below the ionization threshold of solid targets.

Finally, the addition of a photomultiplier tube (PMT) for increased dynamic range could be considered due to their high sensitivities, although the time resolution of PMTs is normally in excess of a few nanoseconds. Furthermore, a PMT typically uses a large detection area, meaning that laser pulses do not need to be so tightly focused onto the detector in order to capture the full energy in the pulse, as is the case with fast-photodiodes. Hence, when appropriately protected by an optical limiter and incorporated into a multi-channel contrast diagnostic, PMTs offer potential for ultra-high sensitivity measurement and thus increased dynamic ranges.

Chapter 5

A supercontinuum source based on laser-liquid interactions

The discussion in this chapter directly follows and builds upon the work published by the author in *Applied Optics* [180], concerning the development and spectral analysis of a supercontinuum laser source (with a ~ 40 μJ output spanning a ~ 600 nm spectral range with 3-4% RMS stability) based on propagation of intense laser pulses through a liquid medium with high optical nonlinearity. This includes a detailed characterization of the stability of the supercontinuum spectrum and how this changes with the pump pulse energy, as well as spectral correlations analysis for making inferences concerning the physical processes contributing to spectral broadening.

A supercontinuum laser is an ultra-broadband source of light with high spatial coherence generated by nonlinear optical methods [181]. Due to their large bandwidths (octave spanning) and coherence properties, white-light pulses are potentially compressible to extremely short transform-limited pulse durations [182–185], subject to appropriate management of spectral phase. Supercontinuum light sources may be used for a variety of purposes, including numerous biomedical applications [186] such as Optical Coherence Tomography [187], fiber-optic communications [188], chemical sensing (spectroscopy) and microscopy [189], remote sensing and atmospheric analysis [190] and white-light interferometry.

5.1 Supercontinuum generation mechanisms

Typically, spectral broadening is achieved via propagating short or ultrashort pulses through highly nonlinear media, such as crystalline solids and other bulk materials [191], photonic crystal fibers (PCF) [192–194], gas-filled [195, 196] or liquid-filled hollow-core PCFs [197, 198] or liquids [175, 199–203]. A combination of nonlinear processes is often responsible for broadening; these might include self-phase modulation, optical wave breaking [204, 205], modulation instabilities [206–208], four-wave mixing [209–211], soliton self-frequency shift effects [212–218] or stimulated Raman scattering.

Much of the reported work on supercontinuum sources and their stability focuses on fiber-based sources [219–224] or spectral broadening in solid bulk materials, such as YAG crystals [191, 225]. Preceding works have used heterodyne detection techniques to study supercontinuum noise [226, 227], while the dispersive time stretch technique, a powerful tool allowing real-time measurement, has been used to measure spectral fluctuations related to supercontinuum seeding [228] and spectra broadened by modulation instabilities [208, 220], which typically contain fine structure that can vary considerably pulse-to-pulse. The spectral stability of supercontinua from liquid CS₂-filled fibers was studied by Chemnitz et al. [198], where molecular re-orientational effects contributing to the optical nonlinearity associated with elongated liquid molecules were found to increase the pulse-to-pulse reproducibility of spectra compared to nonlinearity purely due to the electronic response.

A pulsed supercontinuum light source based on focusing ~ 500 fs infrared pulses (1054 nm center wavelength, (3.5 ± 0.2) nm FWHM bandwidth) into a concentrated K₂ZnCl₄ double-salt aqueous solution was developed and spectrally characterized for its stability at a range of input energies. The work reported in [180] and this thesis describes a broadband spectral stability characterization of a supercontinuum generated in a liquid medium spanning the visible to near-IR spectral regions. This investigation into supercontinuum generation grew out of the Optical Limiter work described in Chapter 4, where spectral broadening was observed as a by-product of the Optical Kerr gating technique used for optical intensity limiting.

Although other liquids or solvents, such as CS_2 , have better transmission properties with high optical nonlinearities, there are some key advantages to using aqueous salt solutions. Compared to CS_2 , which is teratogenic, highly toxic, volatile at room temperature and flammable, the salt solutions pose far fewer handling risks beyond mild corrosiveness and unlikely accidental ingestion. As CS_2 evaporates at room temperature, it is unwise to photo-ionize a flammable liquid-gas mixture. Unlike salt solutions, nonlinear liquids such as CS_2 or toluene chemically degrade many common sealants, potentially leading to containment issues in optical systems that require high quality entrance and exit windows. In addition, we do not expect lifetime issues such as photochemical degradation to affect salt solutions; ZnCl_2 and KCl molecules in solution are already dissociated, unlike CS_2 or hydrocarbon molecules, which been observed to dissociate following multiphoton ionization of inner-shell electrons by 200 fs IR pulses [176].

The linear optical absorption of aqueous salt solutions has been shown by Li et al. [229] to increase with salt concentration for wavelengths between 300-700 nm, although the change is less pronounced for near-IR wavelengths. The laser-induced bulk damage thresholds were measured for the K_2ZnCl_4 solution and pure water; these were found to be at pulse energies of (15 ± 4) μJ and (12 ± 3) μJ for the salt solution and pure-water respectively, suggesting that damage thresholds increase with salt concentration. A similar trend with salt solutions was found by Hammer et al. using 400 fs pulses [230]. As the light was focused into the center of a bulk liquid, the damage thresholds were not determined in terms of intensity or fluence, as the beam size at the breakdown point is not easily measurable, hence are reported in terms of input energy.

This short-pulse supercontinuum source was previously used for broadband interferometry applications [231] and calibration of the absolute photon response of optical streak cameras in a fast-sweep mode for optical pyrometry diagnostics at the AWE Orion laser facility, as this required high-brightness pulses spanning the full visible spectrum on a picosecond timescale [232]. Shot-to-shot stability is needed to ensure good reproducibility, essential for absolute streak camera calibration. Similarly, all multi-shot scanning applications benefit from spectrally and energetically stable pulses. Hence, the work reported in [180] and this chapter is necessary to demonstrate the source's stability.

5.1.1 Self-phase modulation

Self-phase modulation (SPM) arises as a consequence of the third-order nonlinear optical Kerr nonlinearity, where the refractive index of a material exhibits a dependence on the light intensity. The refractive index n of a material in this case is given by

$$n(\mathbf{r}, t) = n_0 + n_2 I(\mathbf{r}, t) \quad (5.1)$$

where $I(\mathbf{r}, t)$ describes the spatio-temporally varying intensity profile of a laser pulse, for example with a Gaussian temporal shape and neglecting the spatial intensity variation, $I(t)$ will have the form

$$I(t) = I_0 \exp\left(-\frac{t^2}{\tau^2}\right), \quad (5.2)$$

where the parameter τ is related to the FWHM of the pulse by

$$\tau = \frac{t_{FWHM}}{2\sqrt{\ln 2}}. \quad (5.3)$$

The spatial component of the refractive index variation can give rise to self-focusing, while the temporal variation results in SPM. For an initially unchirped pulse, SPM leads to a pulse acquiring chirp and subsequent spectral broadening, typically generating both ‘longer’ and ‘shorter’ wavelengths at the front and back of the pulse respectively.

Again neglecting any spatial intensity variation, the instantaneous phase of a plane-wave laser pulse, $\phi(t)$, is given by

$$\phi(t) = \omega_0 t - kz \quad (5.4)$$

where ω_0 is the center-frequency of the laser, k is the wavenumber and z is the spatial coordinate

along the propagation direction. Using $k = \frac{2\pi}{\lambda_0}n(I)$, and taking the time-derivative of $\phi(t)$, we obtain the instantaneous frequency $\omega(t)$ given by

$$\omega(t) = \omega_0 + \frac{4\pi L n_2 I_0}{\lambda_0 \tau^2} t \exp\left(-\frac{t^2}{\tau^2}\right), \quad (5.5)$$

where L is a propagation distance. For the central region of the pulse, Taylor-expanding the expression in Eq. 5.5 about $t = 0$ to first order gives

$$\omega(t) \approx \omega_0 + \alpha t, \quad (5.6)$$

where we have written $\alpha = 4\pi L n_2 I_0 / \lambda_0 \tau^2$ for convenience. From this expression, it is apparent that within its envelope, an intense laser pulse will undergo an instantaneous frequency shift across its temporal profile, resulting in an initially unchirped pulse acquiring a linear chirp and undergoing symmetric spectral broadening about the center-wavelength as a consequence [233–235].

5.1.2 Optical wave breaking

The combined effects of SPM and group velocity dispersion (GVD) were shown by Tomlinson et al. to lead to optical wave breaking, resulting in the formation of well-developed coherent spectral side-lobes [204]; this occurs where the frequency-shifted light overruns the pulse tails, by either traveling faster than the non-shifted light at the leading edge or slower at the trailing edge, such that the interference between the shifted and non-shifted light generates new frequencies. As SPM can be described as a Four-Wave Mixing phenomenon, for two widely separated frequencies, the interference between them will generate light shifted by their difference frequency [204]. Optical wave breaking has also been shown to occur for two co-propagating pulses in optical fibers, where cross-phase modulation replaces self-phase modulation as the initial spectral broadening mechanism [236].

5.2 Spectral stability characterization

5.2.1 Experimental setup

The Cerberus laser at Imperial College London, a Nd:glass system using Optical Parametric Chirped Pulse Amplification (OPCPA) capable of 10 Hz operation, was used to provide test pulses of up to 2.5 mJ with (500 ± 50) fs FWHM durations, $\sim 1.5\text{-}2\%$ RMS energy stabilities and (10.0 ± 0.5) mm $1/e^2$ beam widths. These were focused into cells containing the double-salt solution using a $f = 60$ mm plano-convex lens, in order to generate the supercontinuum within the focal volume under conditions of SPM and self-focusing, leading to filamentation. The focal spot size within the liquid was not possible to measure directly, and indeed varied with input energy due to self-focusing. The apparatus used to acquire spectra for characterizing the source's spectral stability is shown in Fig. 5.1.

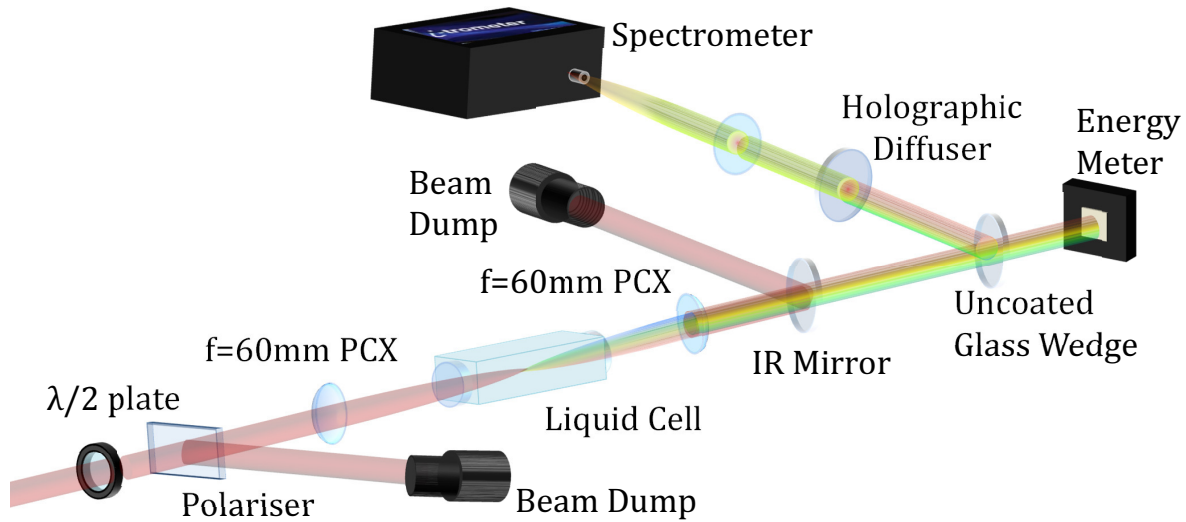


Figure 5.1: Layout of the setup used to acquire spectra for analysis of the supercontinuum source's spectral stability. A holographic diffuser was used to spatially smooth the pulses in order to isolate the overall pulse-to-pulse spectral stability.

Spectra were measured with a B&W Tek BRC624E spectrometer, a modular grating/linear-CCD device with a manufacturer-quoted resolution of 1.8 nm within a spectral range of 190-1100 nm and built-in nonlinearity correction. All spectra were acquired as single-pulse spectra; this was achieved by electronically triggering the spectrometer and a physical shutter to ensure each acquisition corresponded to a single light pulse, with a trigger source synchronized to

the laser system. The acquisition time was set to 100 ms to minimize any detection of any stray light. A spectrum was then saved for each single acquisition. A $\lambda/2$ plate and polarizer were used to control the input pulse energies focused into the liquid cell, containing an aqueous K_2ZnCl_4 solution of concentration (652 ± 7) g/L. The liquid cell was 80 mm in length, and sealed at each end with an 8 mm thick N-BK7 window, which was anti-reflection coated for 1054 nm on the outer surface and uncoated on the inner surface to better refractive index-match to the liquid. An infrared mirror was used to direct the residual fundamental 1054 nm component into a beam dump. To prevent the spectrometer saturating, an uncoated glass wedge was used to pick off $\sim 4\%$ of the white light, which was focused into the spectrometer. As the pulses in our case were relatively energetic, a substantial signal could be recorded even with a single pulse coinciding with acquisition time window. The transmitted component was directed onto a Gentec broadband surface-absorbing energy meter for simultaneous white-light energy monitoring capability. A holographic diffuser was used to spatially smooth the pulse's intensity profile prior to the spectrometer input to isolate the overall pulse-to-pulse spectral stability, in terms of generation of spectral content across the full beam, from any spatial inhomogeneity resulting in the spectrometer incompletely sampling the spectrum.

The energy content of the supercontinuum pulse (with the 1054 nm fundamental wavelength removed using an IR mirror) was found to scale weakly quadratically with the driver pulse energy within the range of energies considered, with a relatively large linear component (Fig. 5.2). This implies that the system efficiency of spectral broadening, defined here as the ratio of output supercontinuum energy (*after* removal of the fundamental wavelength) and input IR energy, weakly increases with input energy from $(0.99 \pm 0.05)\%$ to $(1.57 \pm 0.06)\%$ within the input energy range investigated. The trend-line has not been forced through (0,0), as with possibly several nonlinear broadening processes involved in generating the supercontinuum, it is not possible to confidently fit a model that can be used to extrapolate from the measured data points; hence, the closest fit to the available data was used.

The conversion efficiency is limited by the nonlinear medium's n_2 value and the interaction length over which the pulse is sufficiently intense for significant nonlinear effects to occur, which depends on factors such as the focal geometry and self-focusing in the liquid. For this

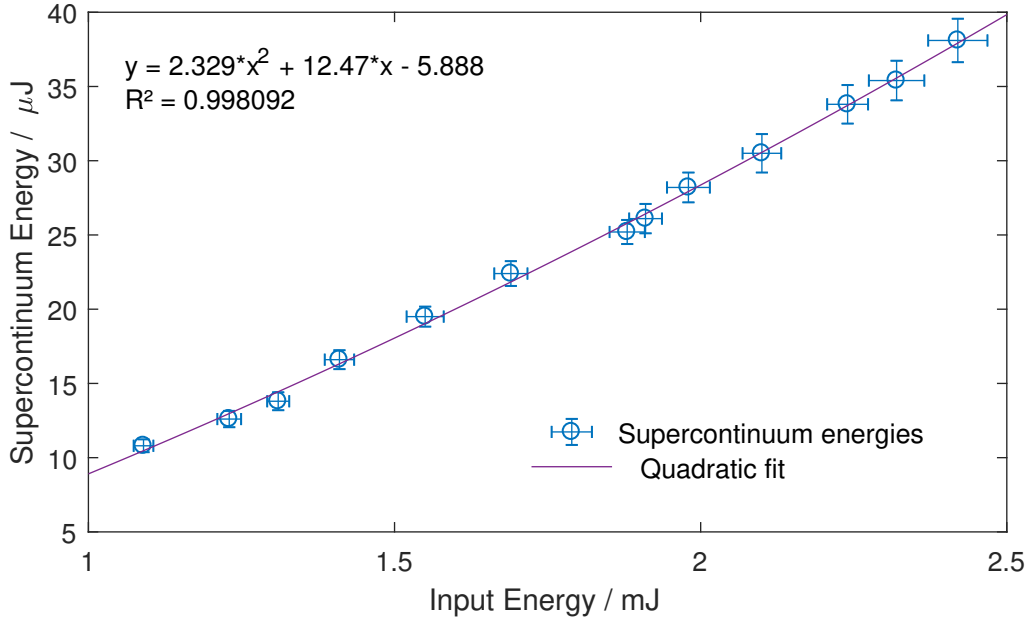


Figure 5.2: Variation of the supercontinuum pulse energy with the input driver pulse energy, measured after removing the fundamental 1054 nm component using an infrared mirror. Each data point is the mean value measured over 100 shots. A quadratic fit was found to match the data most closely, but is only valid for the data in the investigated input energy range, thus was not forced through (0,0). Error bars shown are the standard deviations of the mean input IR and supercontinuum energies.

reason, PCF supercontinuum sources typically exhibit higher conversion efficiencies due to their longer interaction lengths. However the usable pump energy in PCF is relatively low due to the risk of damaging the fiber’s internal structure (~ 400 nJ for 130 fs pulses delivered at 10 kHz repetition rate [237]), not an issue in our case. The supercontinuum pulse duration, broadened due to normal dispersion in the liquid, was measured to be ~ 5 ps using an optical streak camera with 1 ps time resolution (Axis Photonique #602). The source can be simply implemented and is resistant to damage from high light intensities breaking down the focal volume due to liquid media being ‘self-healing’. Salt solution-filled cells are low-cost and very easy to implement and align relative to PCF, although a relatively powerful laser is required to drive the supercontinuum. The salt solution used for this study was chosen based on work by T. Jimbo, who reported enhanced spectral broadening via SPM in concentrated salt solutions relative to water, due to increased nonlinearity by addition of metal cations [175].

5.2.2 Stability Results

A supercontinuum spectrum generated using (2.42 ± 0.04) mJ, (500 ± 50) fs, 1054 nm driver pulses and averaged over 50 shots is shown in Fig. 5.3. The standard deviation of the measured intensity counts at each wavelength point is shown as an error bar, with all 50 spectra displayed in gray to demonstrate the spatially-integrated shot-to-shot variability. These show that the spectra remain very consistent shot-to-shot. From Fig. 5.3, it is clear that the stability is not constant across the full spectrum, hence it is important to characterize the ‘local’ stability over the widest possible wavelength range. An absorption profile for water [238] is overlaid on the spectrum in Fig. 5.3, and a predicted spectrum before and after propagation through 5 cm of water is shown in Fig. 5.4.

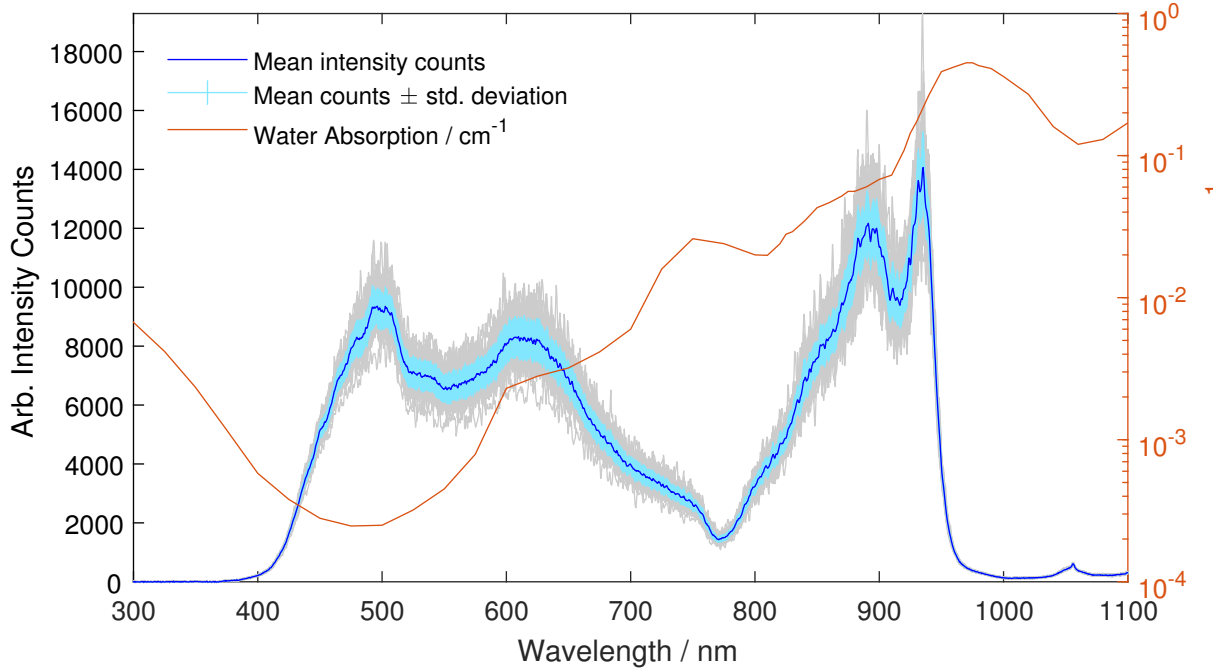


Figure 5.3: A supercontinuum spectrum generated from the K_2ZnCl_4 double-salt solution using 1054 nm drive pulses of (2.42 ± 0.04) mJ and (500 ± 50) fs. The spectrum was spatially smoothed using a holographic diffuser, and averaged over 50 shots (blue line), with the ensemble standard deviation at each wavelength point shown as an error bar. All 50 spectra measured at this input energy are also shown in gray for comparison. The spectrum is overlaid with the water absorption profile (orange line and right-hand side axis) [238].

From these, it is apparent that the structure in the supercontinuum is partly modulated by light absorption in the solvent (water), as strong absorption features coincide with some troughs in the supercontinuum spectrum. As a linear process, the absorption modulations on the spectral

shape are expected to be highly reproducible shot-to-shot. However, based on Fig. 5.4, the structure in the measured supercontinuum spectra can only partly be explained by absorption, hence is possibly a signature of the underlying nonlinear processes, such as the occurrence of optical wave breaking following SPM and GVD. Spectral content above ~ 1000 nm is also largely removed by the high-reflectivity infrared mirror used to remove the drive pulse, however a residual component of this is nonetheless detected by the spectrometer. In addition, water has strong absorption bands above $1.2\text{ }\mu\text{m}$, hence any generated longer wavelengths will be attenuated heavily within the liquid. For applications requiring broadband near- to mid-IR pulses generated in a liquid medium, the absorption on the spectral profile could be reduced by shortening the propagation path through the liquid, moving to an alternative solvent, or alternatively using a liquid jet [201], at the expense of increased engineering complexity.

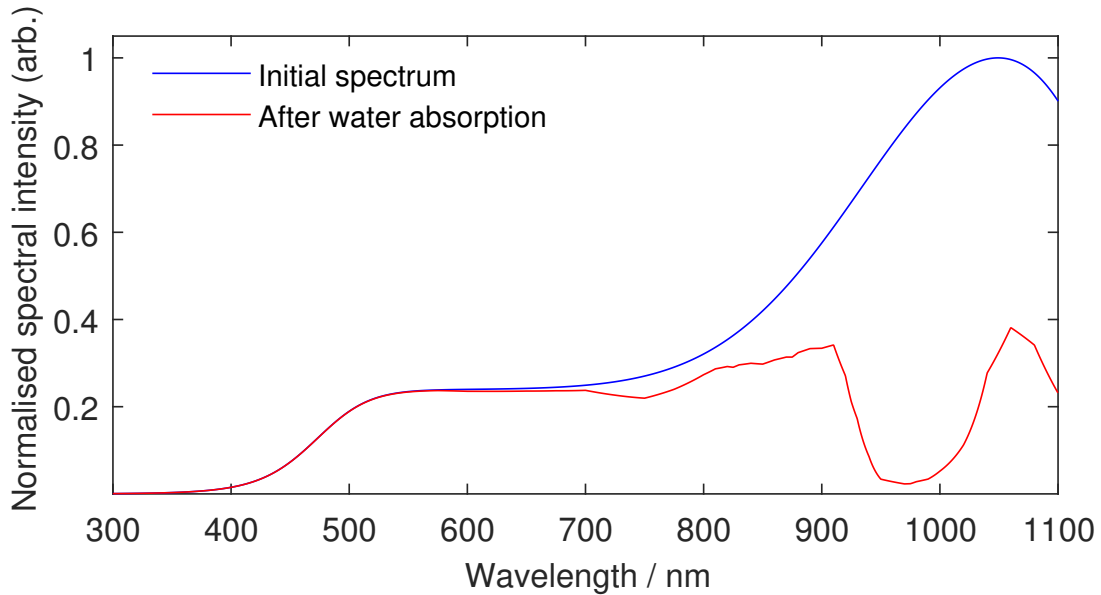


Figure 5.4: Calculated spectrum for propagation of a supercontinuum pulse through 5 cm of water under absorptive conditions. The absorption data used in this calculation was taken from [238]. Although there are some clear modulations bearing similarity to the spectral shape in Fig. 5.3, these are not sufficiently pronounced to explain the supercontinuum's spectral shape, which is likely also a signature of the underlying nonlinear spectral broadening processes.

For comparison to Fig. 5.3, several single-shot acquisition spectra are shown in Fig. 5.5, with the same axis limits for each plot; this demonstrates that the spectral profiles remain very consistent shot-to-shot.

The spectrometer used for these measurements had a sampling (linear CCD pixel read-out)

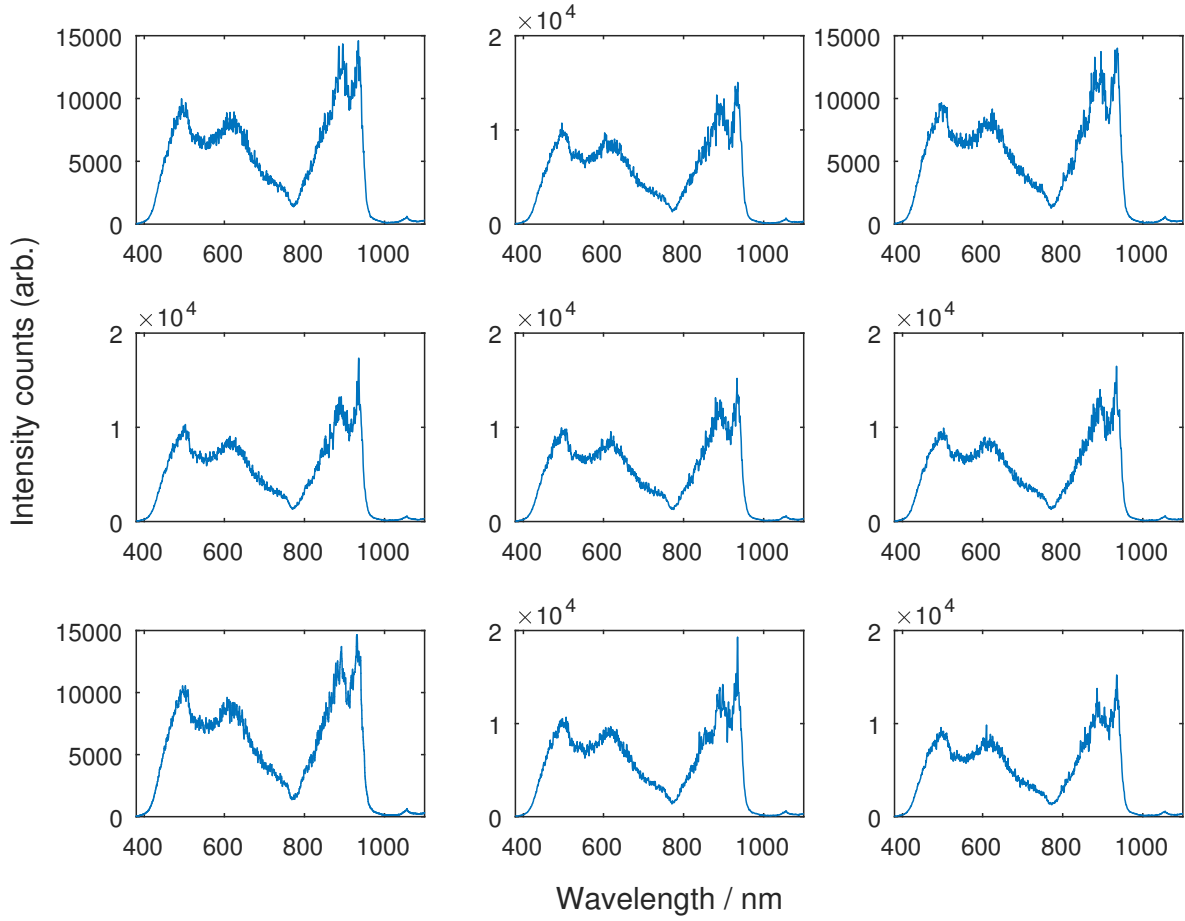


Figure 5.5: A selection of single-shot acquisition spectra with a mean input infrared pulse energy of (2.42 ± 0.04) mJ, demonstrating the good shot-to-shot spectral stability.

resolution (0.53 nm) higher than the manufacturer's quoted optical resolution (1.8 nm), and so there is additional uncertainty introduced to spectral measurements, on top of electrical and dark noise and shot-to-shot variation of the supercontinuum pulses, as a given pixel could detect photons with wavelengths outside of its 0.53 nm-wide bin. Hence, the spectral intensity data was sorted into wavelength bins of 1.8 nm widths to reduce the effect of this wavelength-resolution pixel uncertainty when quantifying the spectral stability in the following sections.

5.2.3 Scaling properties with input energy

A key advantage of using a fluid for the nonlinear medium is its inherent resistance to permanent damage via photo-ionization, color center formation or mechanical fracture. Ionic salt solutions are also already dissociated and therefore resistant to chemical degradation by bond-breaking

via irradiation over time. Hence, pulses with intensities exceeding the material breakdown threshold can be used to drive the supercontinuum, allowing larger broadband pulse energies to be obtained. Increasing the energy was found to be advantageous not only for supercontinuum generation efficiency, but also in terms of stability; if the nonlinear processes are driven further above the threshold intensities, small variations in input energy are expected to have less of an effect on output pulses properties than when operating very close to threshold. Alternatively, self-guiding effects in filaments [239, 240] could help to stabilize the supercontinuum by maintaining a sufficiently small beam-width in order to be above the threshold intensity for spectral broadening over longer interaction lengths. However, this requires a relatively smooth spatial pulse profile, as spatial intensity modulations are amplified by self-focusing, eventually resulting in multiple filaments or catastrophic beam breakup. The effect cannot be accounted for by only considering the input RMS energy stabilities (measured by the Gentec energy meter); as shown in Fig. 5.6, where each data point was taken at a different mean input energy, there is no clear correlation between input and output RMS stabilities when measuring over a range of mean input energies, suggesting the output pulse stabilities must also be dependent on another variable.

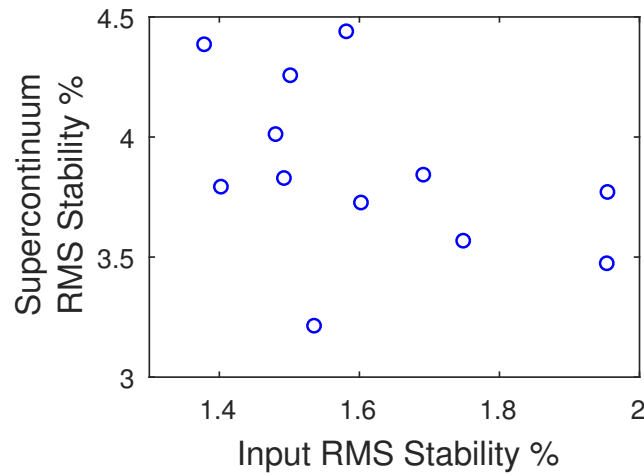


Figure 5.6: Variation of the output RMS energy stability against input RMS; no clear correlation was observed. Each data point reflects the RMS stabilities about a particular mean energy value, where the mean energies range between (1.09 ± 0.02) and (2.42 ± 0.04) mJ.

The stability improvement with increasing drive energy was tested in the case of the K_2ZnCl_4 source by considering two metrics: the energy stability of the white-light measured using an energy meter and the spectral stability across the full spectrum over 50 shots. Spectral stability

was quantified by considering the standard deviation (SD) of the measured intensity counts (as a percentage of the mean) at each wavelength value, or by considering standard deviations of spectral counts binned into variable wavelength bands. A *reduction in the SD* as percentage of the mean signifies an *increase in the spectral stability*. This is referred to as a ‘stability’, rather than an ‘instability’, for consistency with terminology commonly used within the field, as laser systems’ shot-to-shot energy stabilities (variations) are commonly quoted as RMS values, where increasing values reflect a more unstable system.

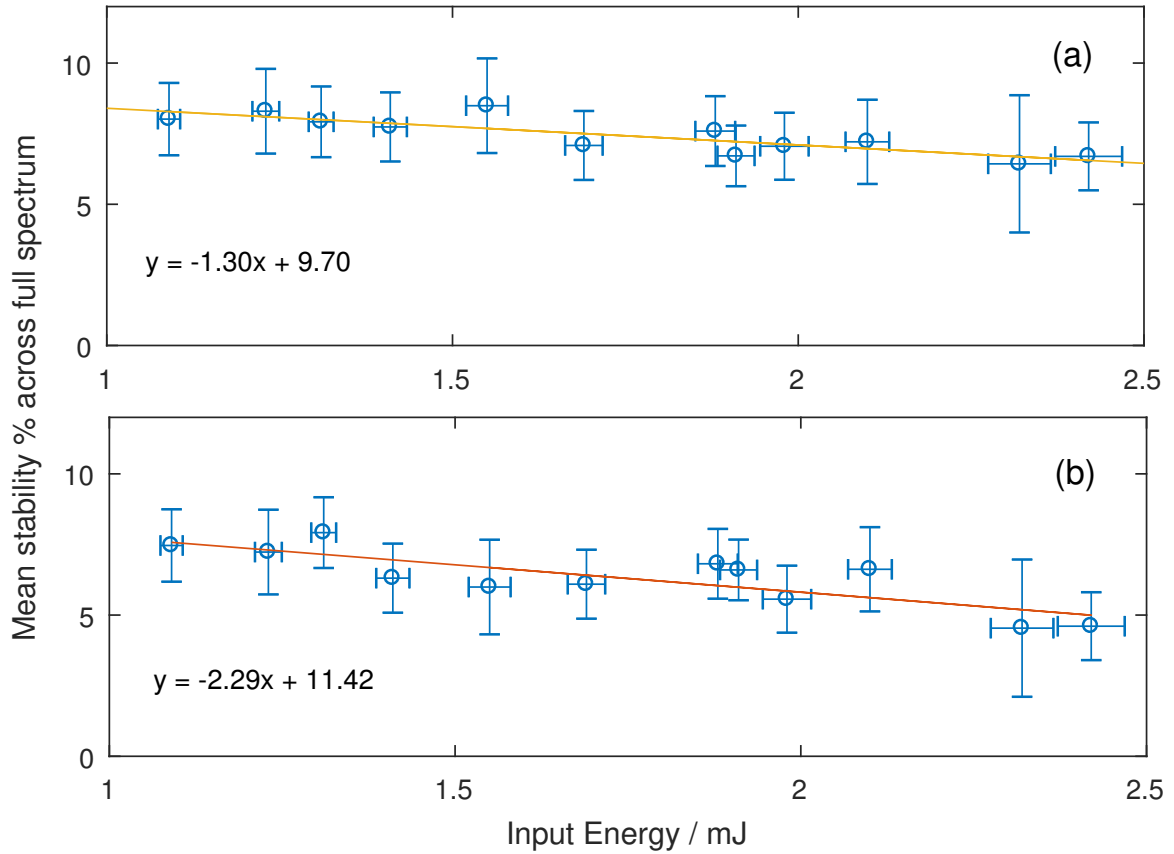


Figure 5.7: Energy scaling of the global shot-to-shot spectral stability σ_{glo} in the cases of (a) no scaling to input RMS energy stability and (b) with the data scaled to the input RMS stability values for each energy. The metric for stability at each energy is quantified by sorting spectral intensity data into 1.8 nm bins, and calculating σ_{glo} using Eqs. (5.7) and (5.8). Uncertainties shown in the error bars reflect the standard deviations of the mean stability percentages for each input energy value.

The variation of global spectral stability σ_{glo} with input energy is shown in Fig. 5.7, where the quoted stability values are calculated by sorting spectral intensity data into 1.8 nm width wavelength bins, determining the SD (σ_{λ_i}) of intensity counts (I_{λ_i}) in each bin (λ_i), as a percentage of the mean value of intensity counts ($\bar{I}_{\lambda_i}$). We then calculate the mean of these

SDs across the full spectrum to obtain an overall spectral stability value for each tested input energy. The spectral stability σ_{λ_i} of each wavelength bin as a percentage of $\bar{I}_{\lambda_i}$ is given by

$$\sigma_{\lambda_i} = \frac{100}{\bar{I}_{\lambda_i}} \sqrt{(\bar{I}_{\lambda_i}^2) - (\bar{I}_{\lambda_i})^2}, \quad (5.7)$$

where $\bar{X}$ denotes the mean of a variable X . For a given input energy, σ_{glo} is thus simply the mean of σ_{λ_i} , such that

$$\sigma_{glo} = \bar{\sigma}_{\lambda_i}. \quad (5.8)$$

1.8 nm bin widths were used to reduce the effect of the aforementioned pixel resolution uncertainty on the measured spectra, and were chosen based on the manufacturer's quoted optical resolution of 1.8 nm. A gradual increase in spectral stability, evidenced by a reduction in the SDs as percentages of the mean count values, was observed by increasing the input energy (Fig. 5.7a). This effect was found to be more pronounced if the RMS stabilities of the input pulse energies was taken into account, as shown in Fig 5.7b, where the stability dataset has been scaled according to the input RMS stabilities. For comparison with the spectral stability scaling with input energy, the scalings of the RMS stabilities of the energy contained within the supercontinuum with increasing input energy (Fig. 5.8) were also analyzed.

The energy stability is approximately constant across the range of energies considered, with some indication of a weak trend towards increased stability with increasing input energy. Similarly to the spectral stability behavior, the trend becomes more pronounced (steeper) after correcting for the variations in input RMS energy stabilities, however the uncertainties are relatively large compared to the differences between stability values. The scaled RMS supercontinuum energy stabilities range between $\sim 2.4\%$ and $\sim 4.5\%$ for input energy RMS stabilities ranging between $\sim 1.4\%$ and $\sim 2\%$, hence the K_2ZnCl_4 supercontinuum source has favorable energy stability properties considering its reliance on nonlinear optical spectral broadening processes.

Shape variations in the spectral stability plots (λ_i vs σ_{λ_i}) were observed as the driver pulse

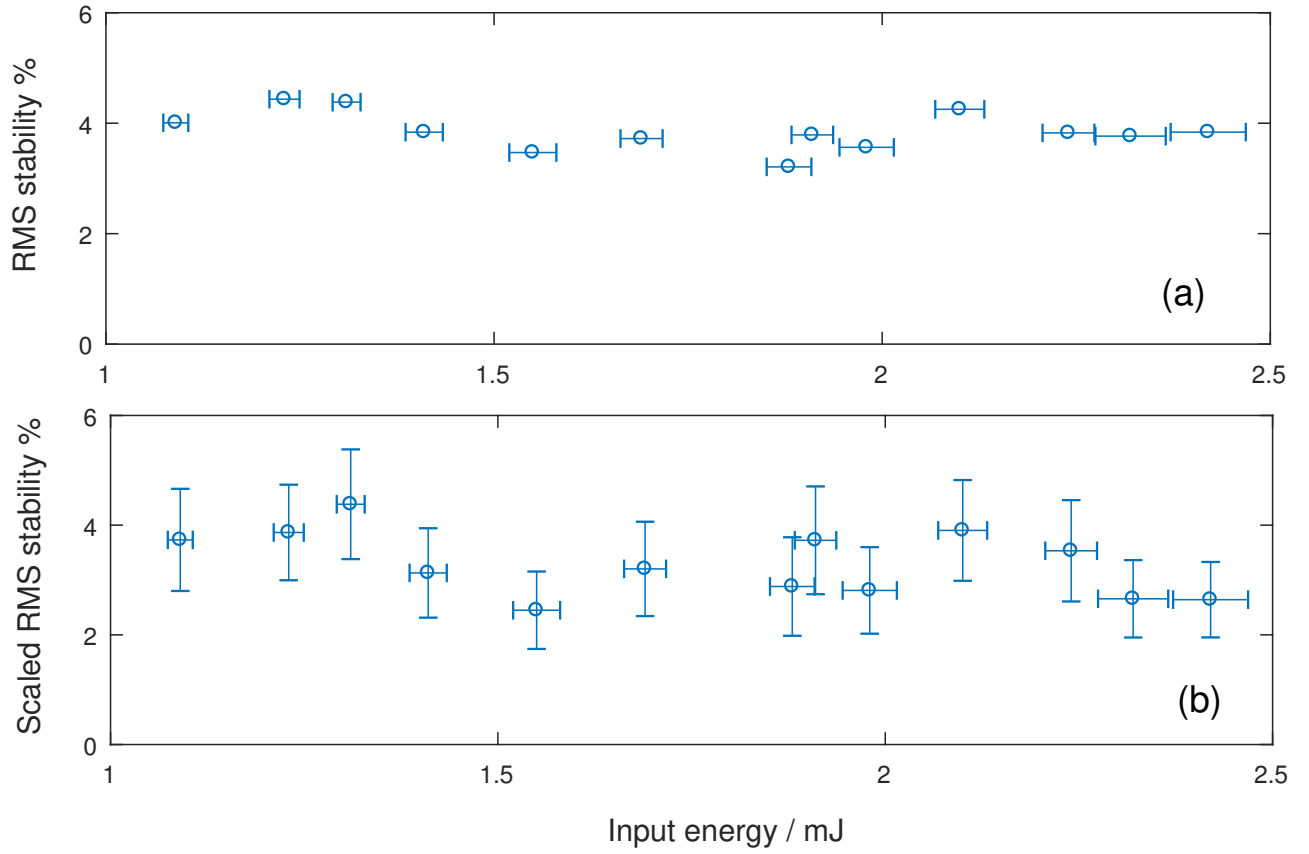


Figure 5.8: Variation in the RMS energy stability in the supercontinuum pulse, measured using a broadband surface-absorbing Gentec energy meter. The data is presented (a) without and (b) with scaling to the input pulse RMS energy stabilities. The scaling factor applied to (b) was the ratio of the smallest percentage RMS stability of the dataset and the input RMS stability for a given average input energy. The correction factors are displayed as error bars to indicate the additional uncertainty introduced by an increased input energy variability.

energies were increased, shown in Fig. 5.9. These manifested as both an overall decrease in the standard deviations in intensity counts measured in each bin, suggesting a global increase in spectral stability (reduction in unscaled σ_{glo} from 8.3% to 6.7%), and also apparent localized improvements in the stability of particular spectral regions, particularly around the edges of spectral peaks (400 nm, 970 nm). The effect is particularly pronounced close to 400 nm, the furthest generated wavelength from the drive wavelength, where σ_{λ_i} decreases from $\sim 15\%$ to $\sim 5\%$. This is likely due to there being larger numbers of photons in these wavelength bands as the supercontinuum is driven more strongly, progressively improving the signal-to-noise ratio, which has a much more pronounced effect in the regions with small photon counts. The spectral stability plots in Fig. 5.9 are only meaningful for wavelength bands in which there is clearly measurable signal, otherwise standard deviations of the counts as percentage of the mean value

in each band for each measured spectrum can become very large, as they only reflect the standard deviations in the mean counts due to instrument and shot noise.

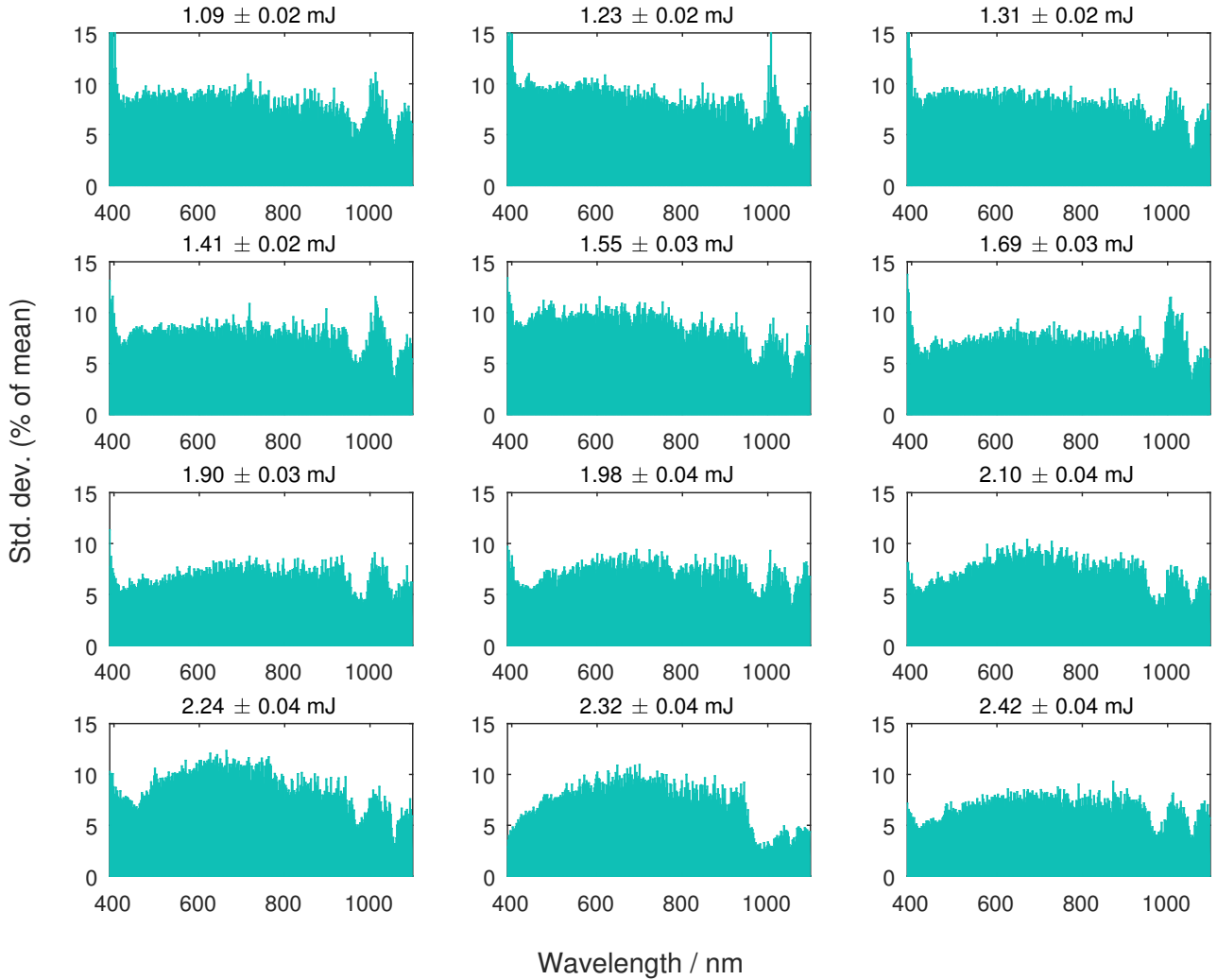


Figure 5.9: A sample of spectral stability plots depicting the standard deviation of the measured intensity counts in each wavelength bin as a percentage of the mean value. Each plot was obtained using ~ 50 spectra measured at the various indicated input driver pulse energies.

Away from the edges of spectral features, the detected counts are far beyond the instrument noise level (< 40 counts), with signal counts exceeding the noise level by $\times 100$ - 1000 , such that the global increase in spectral stability percentages is less pronounced. Hence, for spectral content between 410-970 nm, the spectral fluctuation inherent to the nonlinear broadening mechanisms dominates the detector noise. In regions where there are strong water absorption features, such as > 970 and < 1050 nm, the standard deviations are locally higher, reflecting the lower signal-to-noise ratios in the presence of absorption.

The growth of the supercontinuum spectrum with increasing input energy is also important to consider; this can yield information regarding the nonlinear processes that act to broaden the spectrum, for example if a new spectral feature appears above a certain threshold input energy. In Fig. 5.10, a series of spectra measured as 50 shot averages at each of the input energies considered is presented, ranging from ~ 1.1 mJ up to ~ 2.4 mJ.

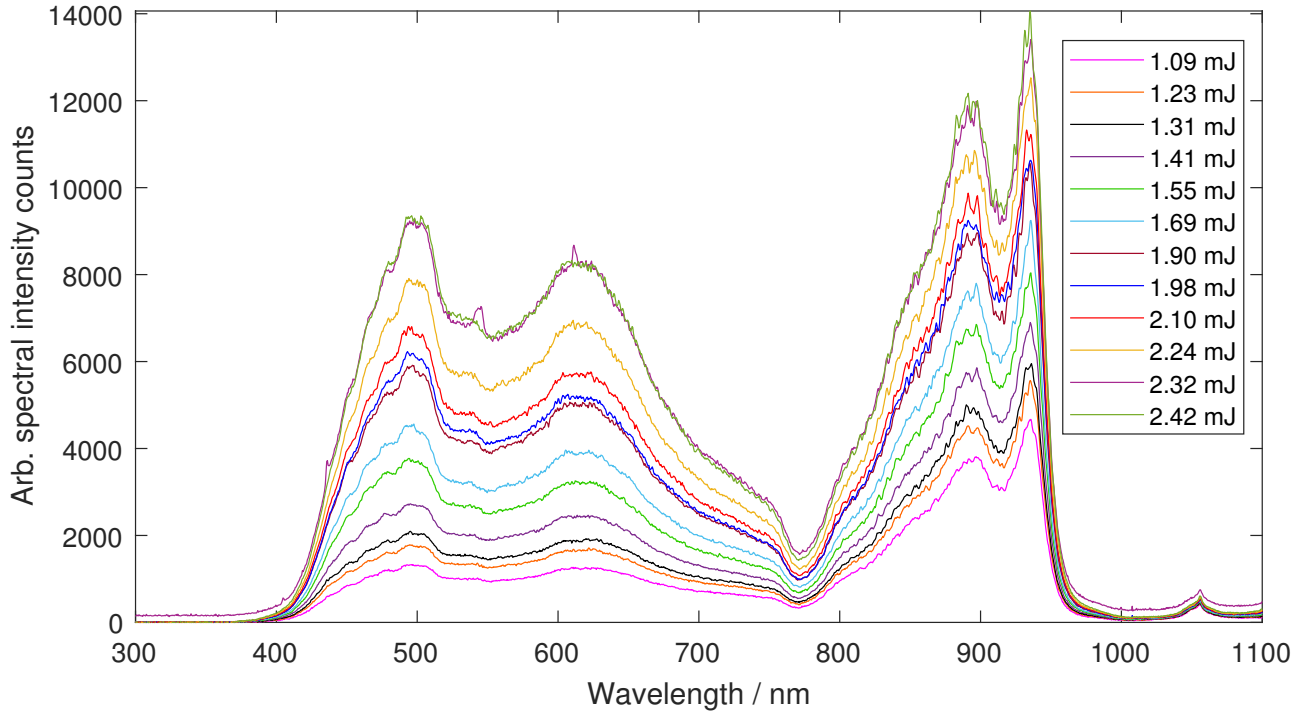


Figure 5.10: Spectra obtained from ~ 50 shot averages at different input laser pulse energies, demonstrating the spectral growth the supercontinuum. The peak/trough features in the visible sidebands become more prominent as the input energy is increased.

Within the considered energy range, there does not appear to be any substantial difference in the average spectral shapes, which appear to vary in measured intensity counts while preserving the overall form of the supercontinuum. However, the trough feature near to ~ 550 nm appears to become more prominent as the pump energy is increased, in contrast to the trough near ~ 910 nm, which remains approximately in proportion with the adjacent peaks. This is consistent with the idea that the sidebands and spectral shape are formed by a process other than SPM (symmetric about the pump wavelength) modulated by absorption, for example optical wave breaking. The spectra for ~ 2.3 and ~ 2.4 mJ pump pulses have very little difference between them in counts, although the energy content in the supercontinuum was measured to increase

between the two pump energy values from ~ 35 to $50 \mu\text{J}$, which suggests that this difference is more likely related to an apparatus issue rather than indicative of a physical mechanism suppressing the conversion of IR photons to other frequencies.

5.3 Spectral Correlations Analysis

Spectral correlation maps provide insight into the relationships between spectral intensities in different regions of the supercontinuum spectrum and can be used to quantify its dynamical evolution [220, 223, 225, 241]. The spectral correlation function $\rho(\lambda_1, \lambda_2)$ between wavelengths λ_1 and λ_2 is defined as

$$\rho(\lambda_1, \lambda_2) = \frac{\langle I(\lambda_1)I(\lambda_2) \rangle - \langle I(\lambda_1) \rangle \langle I(\lambda_2) \rangle}{\sqrt{(\langle I^2(\lambda_1) \rangle - \langle I(\lambda_1) \rangle^2)(\langle I^2(\lambda_2) \rangle - \langle I(\lambda_2) \rangle^2)}} \quad (5.9)$$

where quantities inside angle brackets are the ensemble averages across (in this case) the ~ 50 spectra taken at each input driver pulse energy. The correlations vary between $-1 < \rho \leq 1$, where $\rho = 1$ is by definition observed along the line $\lambda_1 = \lambda_2$. Positive correlations indicate that spectral intensities $I(\lambda_1)$ and $I(\lambda_2)$ increase or decrease together, whereas a negative correlation arises where a spectral intensity component increases at the expense of the other and vice-versa. This is informative about the transfer of energy between different spectral regions, and thus the spectral broadening mechanisms involved in the laser-liquid interaction by association. For example, the presence of a positive correlation between the relative intensities of two adjacent spectral regions suggests that they are generated by the same or directly linked processes, as might be seen with side-band generation associated with SPM or SPM followed by optical wave breaking. Strong correlations between particular separated and localized regions only could indicate the significance of anti-Stokes stimulated Raman scattering processes. Although not used in this work, the technique of *partial covariance* analysis can be applied to remove trivial correlations from the correlation maps, such as those from pulse-to-pulse intensity variations, which are typically not of interest [242].

A correlation map spanning the full measurable spectrum of the supercontinuum ($\Delta\lambda > 600$ nm) is shown in Fig. 5.11, with the region around the 1054 pump wavelength shown in greater detail in Fig. 5.12a. Yellow indicates perfect positive correlations, while cyan denotes negative correlation. Areas where there are no correlations are shown in black, such as the region below 400 nm where there is no measured spectral content above the noise level, hence no correlation would be expected with the rest of the spectrum.

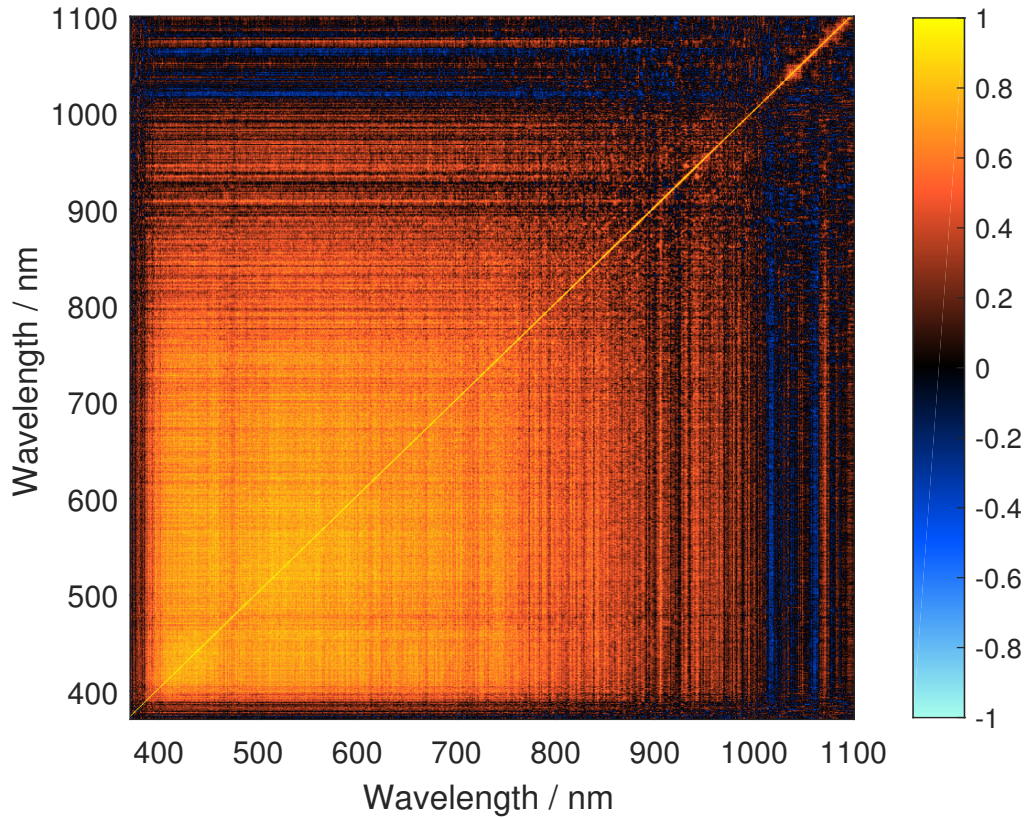


Figure 5.11: Spectral correlation map covering the full spectrum of the supercontinuum generated in the double-salt solution, measured for 1054 nm drive pulses of (2.10 ± 0.03) mJ energies and (500 ± 50) fs FWHM durations.

The spectral correlation maps indicate that self-phase modulation within a dispersive medium (with initially unchirped pulses), possibly followed by additional coherent sideband generation via another mechanism such as optical wave breaking, are likely broadening mechanisms; we observe regions of strong positive correlation, far from the 1054 nm fundamental wavelength, across the 400-800 nm region (Fig. 5.11), whereas there are negative correlations between the residual component of the 3 nm FWHM bandwidth near-infrared 1054 nm fundamental and the

entire spectral region spanning 400-960 nm (correlation values of ≈ -0.5) which is not removed by the IR mirror in the beamline (Fig. 5.12b). This suggests that energy is being shifted away from the fundamental into broad side-bands, as would be expected from a mixture of nonlinear processes such as SPM and GVD followed by optical wave breaking. There are many nonlinear optical processes that could be responsible for spectral broadening, hence it is likely that the observed spectra arise based on a complex interplay of multiple different nonlinear effects as well as some modulation contribution due to absorption in the liquid medium. It is, however, not possible to conclusively determine the dominant broadening mechanism within this work, as much of the spectral information is lost via absorption and the limited measurable spectral range, hence any possible Stokes sidebands are not analyzable. The negatively correlated regions were observed to have weaker correlation values than the positively correlated areas, mainly due to the much lower signal levels in the fundamental spectral region, which only consisted of the leaked IR through a high-reflectivity IR mirror. The correlation maps are consistent with those reported by Van de Walle et al. [225] arising from propagating short pulses through solid-core fibers or focusing them into bulk media, in their case YAG crystals, however the supercontinuum was primarily in the mid-IR rather than the visible and near-IR, as is the case with the aqueous K_2ZnCl_4 solution.

Localized weak negative correlation bands between the spectral peaks at ~ 894 and ~ 927 nm and the full visible region (Fig 5.12c) imply that there is some weak effect transferring energy between these features; this could be due to secondary self-phase modulation of light in these spectral regions, or alternatively another nonlinear effect resulting in spectral shifts. It is unlikely that this is an anti-Stokes Raman scattering effect, as the negative correlation is with the full band rather than a localized spectral region, which would be characteristic of photon energy gain from a particular water vibrational mode. In 24°C liquid-phase water, Raman modes are reported to be present at 3200 cm^{-1} and 3400 cm^{-1} [243], corresponding to photon energy shifts of 0.397 and 0.422 eV respectively. These would potentially result in shifts from 1054 nm to 789 and 776 nm, from 894 nm to 696 and 686 nm, and from 927 nm to 715 and 705 nm. A two stage anti-Stokes Raman shift would result in peaks at approximately 630, 615, 583, 569 and 557 nm from the aforementioned initial peaks. While there are indeed

slight negative correlations observed between the proposed fundamental wavelengths and the calculated Raman shifts, these do not stand out in terms of their correlation strengths from the large numbers of other wavelengths which are also negatively correlated with the fundamental wavelength and two main spectral peaks in the supercontinuum. Hence, we cannot conclude based on this evidence that Raman scattering contributes significantly towards the spectral broadening, in comparison to sideband-producing effects such as SPM followed by optical wave breaking, which are more consistent with the correlation maps.

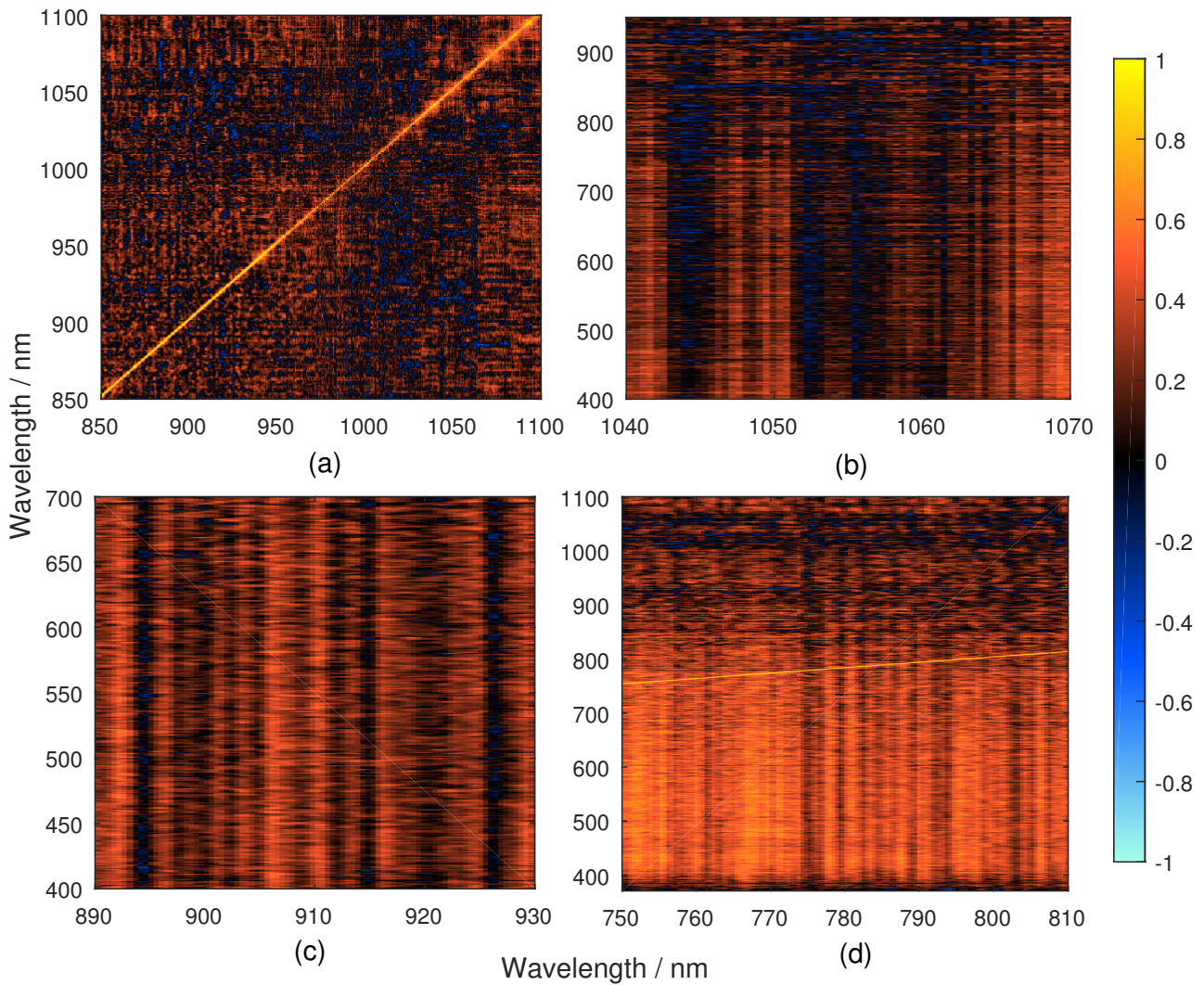


Figure 5.12: Spectral correlation maps from measurements using (2.42 ± 0.04) mJ driver pulses (a) in the region nearer to the fundamental drive wavelength in greater detail; (b) correlations between the fundamental drive wavelength region and the whole generated supercontinuum; (c) narrow bands of negative correlation at ~ 894 and ~ 926 nm, indicating that the supercontinuum generation is a multi-stage process as some energy is shifted from these secondary peaks into the visible region; (d) some weaker correlations are observed around the trough feature at ~ 780 nm, coinciding with a strong water absorption feature, possibly due to suppression of the signal relative to the instrument noise, although this effect is rather weak.

The presence of water absorption features coinciding with certain areas of the spectral profile, as shown in Fig. 5.3, can also be examined using spectral correlations; regions containing strong absorption features relative to the strength of the spectral broadening processes should exhibit weaker correlations, as these dominate the spectral intensity content to a greater extent and lower the signal-to-noise ratios. In Fig. 5.12d, around the spectral trough feature located at ~ 780 nm, some evidence of weaker correlations can be seen, along with a few bands where there is no correlation with the remainder of the spectrum. However, the effect is weak as there is sufficient spectral content in this region to be well above the spectrometer's noise level, such that the dominant behavior for this region is still negative correlation with the region around the drive wavelength and positive correlation with the remainder of the supercontinuum generated by nonlinear effects producing distinct sidebands. The weakness of this effect further suggests that absorption modulation is only partly responsible for the spectral shapes observed.

5.3.0.1 Input Energy Variation

The features present in correlation maps, as previously discussed, can be indicative of particular types of nonlinear broadening processes. Hence, it is instructive to consider if spectral correlations change with the input pump pulse energy, which might reveal the activation of another physical process above a certain threshold energy. This can be done by a simple comparison of the key features of the correlation maps at the various input energies.

Within the pump energy range considered, all of the correlation maps exhibit the same key characteristics, subject to some minor variation in correlation strengths; namely the two key regions of negative correlation around the pump wavelength, and positive correlation between ~ 390 - 1000 nm, which are indicative of sideband generation via energy shifting from the broadened fundamental (1030 - 1070 nm) into the 390 - 960 nm spectral region. This, combined with the insights from Fig. 5.10 concerning spectral growth with pump energy, suggests that a threshold for another nonlinear process does not lie within this pump energy range (~ 1 - 2.5 mJ).

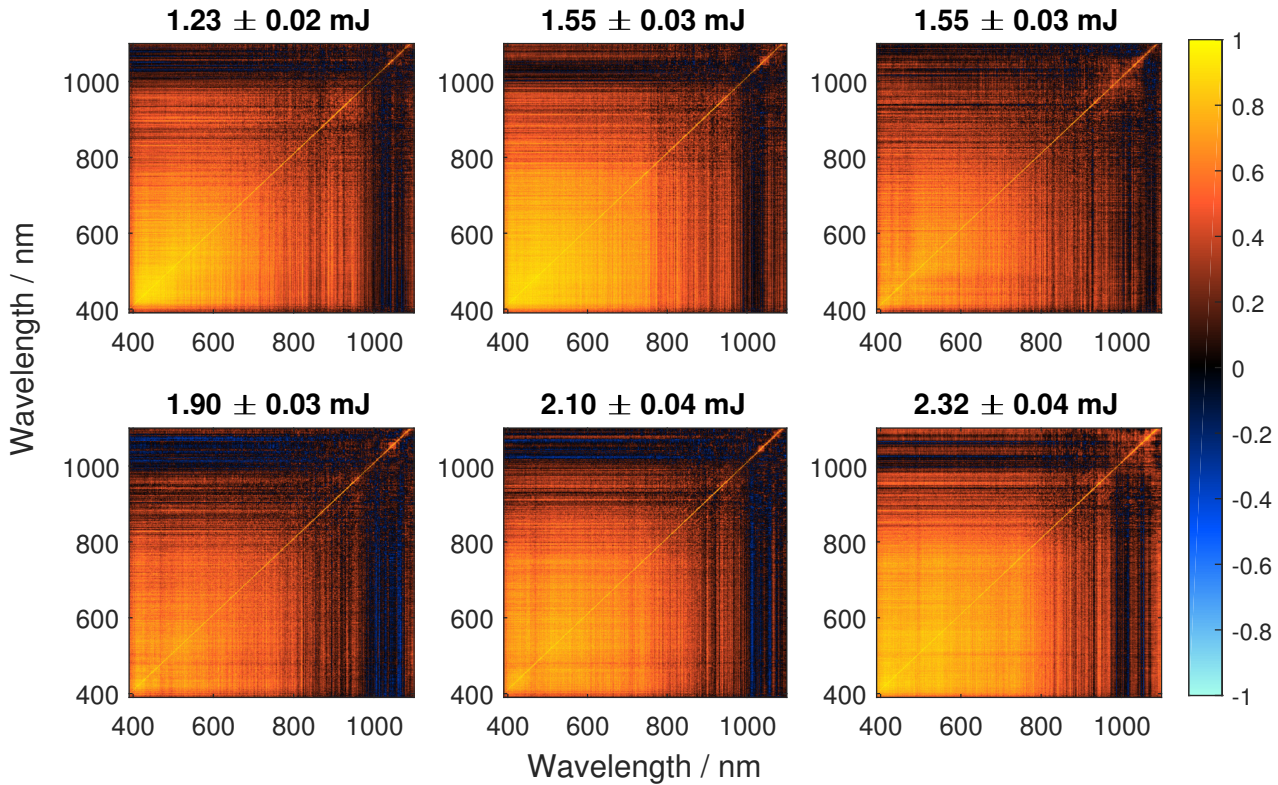


Figure 5.13: Spectral correlation maps for various average input energies, indicated for each map. The key features of a broad, positively correlated region and bands of negative correlation, representing shifting of energy from the broadened pump (1030-1070 nm) into sidebands (390-960 nm), are visible in each case. Some minor variation in correlation strengths are visible between the maps, particularly around the pump wavelength, most likely due to lower signal-to-noise ratio in this region following removal of the IR with a high-reflectivity mirror.

5.4 Conclusions

We have demonstrated that the spectral and energy stability of a simple-to-implement and spatially smoothed supercontinuum laser source based on spectral broadening in a concentrated K_2ZnCl_4 double salt solution is limited to variation of only a few % of the mean energy or spectral intensity values. This stability intrinsically depends on the energy stability of the infrared driver pulses, with white-light energy RMS stability values typically being approximately twice those of the driver pulses. Furthermore, a quadratic relationship between input and supercontinuum pulse energies was observed, while increasing the input energy has also been shown to improve the spectral stability and increase the efficiency of energy transfer from the IR into the supercontinuum. In addition, the use of a liquid nonlinear medium allows the source to be driven beyond its ionization threshold without causing permanent damage.

The shape of the supercontinuum spectrum appears to be determined partly by modulations due to absorption in the solvent (water), and an interplay between possibly several nonlinear processes such as SPM followed by optical wave breaking. The source's stability, relatively high single-shot energy and ~ 5 ps pulse durations (due to dispersive broadening in the liquid nonlinear medium) render it suitable for applications such as dynamic calibration of optical streak cameras in fast-sweep mode. The results represent a major improvement on previously published results concerning a supercontinuum source using K_2ZnCl_4 solutions [175], with an increase in spectral width of ~ 500 nm and supercontinuum output energies of 40 μJ , approaching the preceding work's drive pulse energy of 80 μJ .

Analysis of the correlations between the intensities of different spectral peaks shows positive correlations, indicative of sideband generation from a separate driver pulse that is characteristic of nonlinear effects such as SPM and GVD leading to optical wave breaking. Furthermore, filaments were observed within the liquid, suggesting that the source operated in a regime of strong self-focusing. From these factors, we conclude that the spectral broadening is most likely driven by a complex mixture of nonlinear processes such as SPM in a high n_2 and normally-dispersive medium followed by further sideband generation via optical wave breaking, rather than soliton effects or stimulated Raman Scattering.

The spectral phase properties of this supercontinuum source have not been measured; this would be a useful extension to this work in order to assess the compressibility of the broadband pulses. The transform-limited duration for pulses with these bandwidth properties is ~ 2 fs, hence if the higher order contributions to the spectral phase of the pulses are relatively small, a useful ultrashort light source could potentially be realized.

Chapter 6

Electro-Optic Electromagnetic Pulse Diagnostics

The discussion and figures presented in this chapter are directly based upon work published by the author in *Scientific Reports* [244] concerning the development of an electro-optic diagnostic system for measurement of electromagnetic pulses (EMP) generated by petawatt-regime laser-matter interactions, the first demonstration of such a diagnostic in this regime.

6.1 Introduction to laser-driven EMP

Electromagnetic pulses are strong, transient electromagnetic fields, typically observed in the radiofrequency (RF) to microwave spectral regions, which can be produced in the laboratory via intense pulsed laser-matter interactions in both the long (nanoseconds to 100s of picoseconds) [245–255] and short (picoseconds and shorter) [256–265] pulse duration regimes, where they can create fields exceeding 1 kV/m. The primary mechanisms responsible for generating such high electromagnetic fields in a laser-plasma interaction are typically attributed to ejection of a large flux of energetic ‘hot’ electrons from the plasma (as shown in Fig. 6.1) which strike the interaction chamber walls, followed by a transient ‘slow’ electron return current through the target mount to re-establish quasi-neutrality in the plasma [254,255,257,265]. This

transient discharge current causes the target mounting assembly to emit radiation analogously to an antenna. The ejected electrons striking an interaction chamber's metal walls can also stimulate resonances at frequencies determined by the electromagnetic cavity modes, which are determined by the geometry of the chamber and the hardware it encloses. This contributes a second component to the laser-driven EMP; Fig. 6.2 depicts these two EMP components acting concurrently within a metal interaction chamber, as typically found in most high-power laser facilities.

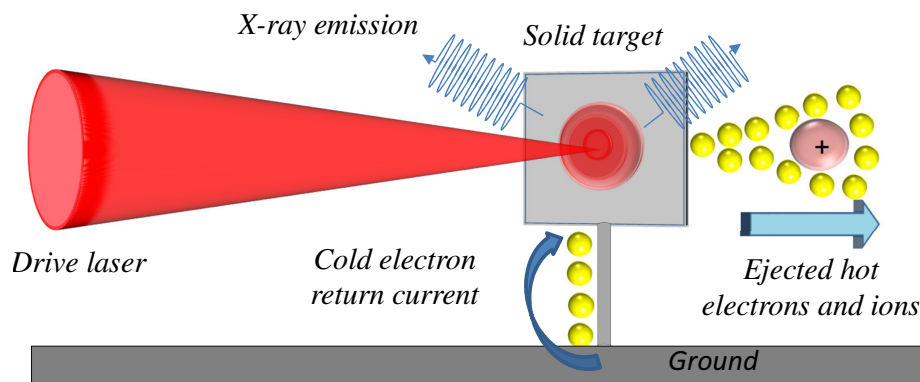


Figure 6.1: Visualization of the interaction between the drive laser and target leading to EMP generation in an interaction chamber. Electrons heated by the laser are ejected from the rear of the target, leaving the target in a net positive charge state. This results in a transient current of cold electrons drawn from the earth through the target assembly to maintain quasi-neutrality.

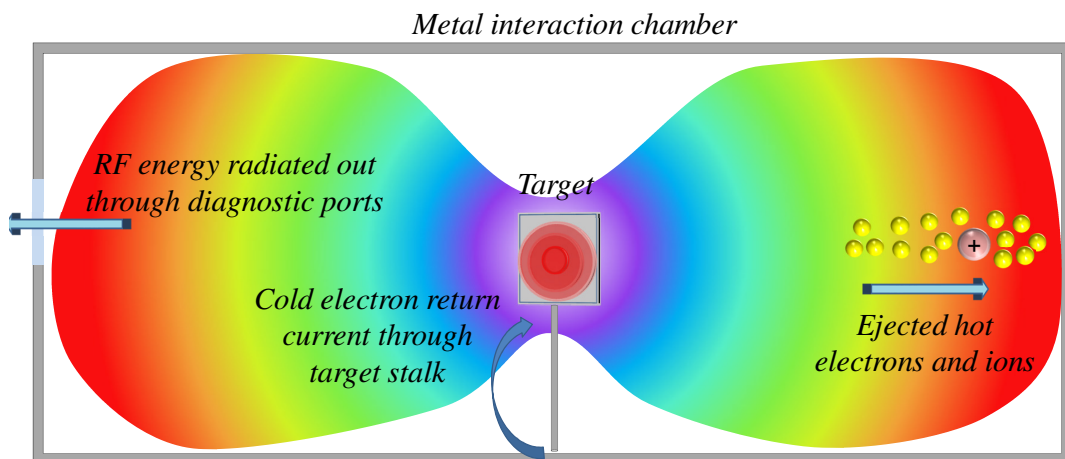


Figure 6.2: Visualization of the EMP generation mechanisms following a high-intensity laser-matter interaction. Electrons ejected from the target strike the conductive chamber wall; this impulse stimulus launches resonances governed by the cavity modes. The transient cold electron current drawn from the earth through the target holder results in antenna-like RF radiation emission from the target assembly at a range of frequencies distinct from the cavity modes.

EMP can be extremely problematic in high-power laser experiments, as EMP signals lasting

for hundreds of nanoseconds often adversely affect nearby electronic systems, possibly resulting in loss of data or even permanent damage to high-value equipment. However, RF emission can also be used to infer information about the interaction physics and complement other diagnostic tools. With the growing number of high-power lasers worldwide [266] (NIF [267], OMEGA-EP [268], Texas Petawatt [269], Orion [270], LMJ and PETAL [38], ELI [37, 271], Z-Petawatt [272], Apollon [39, 40] and more), it is important that we improve our understanding of EMP generation processes from laser-matter interactions for both protecting against its damaging effects (radiation hardening) and to explore its potential uses as a source of fast and intense pulsed electromagnetic fields. The target charging model for EMP generation developed by Poyé et al. [264, 265] has been experimentally validated for short laser pulses with relatively low energies (< 80 mJ), however the model's agreement with experiment is reduced towards the higher end of the investigated energy range, particularly for pulses longer than 0.5 ps, durations which are delivered by many petawatt-scale systems. With the advent of ultra-high intensity laser systems, improving theoretical understanding of EMP and our measurement capabilities in the high-energy petawatt-regime has become particularly important.

Conventionally, EMP is measured using a range of conductive antennas or electrical pickup probes connected to recording devices such as oscilloscopes, including but not limited to B-dot and D-dot probes (sensitive to the time derivative of the B- and D-fields and so are prone to 'noise' in signal recovery via numerical integration) and Möbius loops [273] (sensitive to magnetic fields). However, these diagnostics all require electrical signals to be transported from the point of measurement to oscilloscopes via conductive cables, and conductive probes can locally perturb the electric fields one is attempting to characterize. Even with shielding, conductive cables can pick up considerable amounts of additional EMP 'noise' signal - which might completely obscure the measurement of a particular field component by the probe - making the data difficult to interpret due to very low signal-to-noise ratios. This issue is particularly pronounced on experiments using high-energy picosecond laser pulses.

A solution to these problems is to employ a fundamentally different physical process to transform local electric fields into a useful low-noise signal that can be transported to a remote recording instrument with high fidelity. This chapter describes the use of the linear electro-

optic ‘Pockels’ effect [274] for probing EMP; this is suitable for operation in vacuum, uses dielectric crystalline sensors to minimize local disturbance to the field, and is inherently resistant to electrical noise as any ‘long’ distance signal transport can be done via optical fiber or free space propagation. Hence, in terms of electrical shielding, only an effective Faraday cage is required to protect a remote oscilloscope and optical-detectors from the general EMP background. This task can be simplified considerably by using sufficiently long fibers to completely remove the detection apparatus from the laser facility’s target area, with the caveat that any multi-mode fibers are short enough such that the modal dispersion is less than the time resolution of the optical detectors. An alternative approach to this would be to use single-mode fiber to remove the modal component from the fiber dispersion, however these require better and more expensive fiber-launch apparatus than was available during the diagnostic development process.

As petawatt laser-matter interaction experiments with the current generation of picosecond lasers are inherently single-shot, it is not feasible to significantly reduce noise via statistical analysis methods. Hence, low-noise *absolute* measurements with minimum disturbance to the original fields are of great interest. Absolute measurements of the EMP do not require numerical integration as part of the post-processing of data in order to obtain the field strengths, as is the case for B-Dot and D-Dot probes, which are sensitive to the time derivatives of the fields, rather than the field strengths themselves. Hence, such measurements are less able to reproduce low-frequency (slowly varying) or DC field components of EMP, and effectively amplify the high frequency components upon numerical integration of the voltages measured by conductive probes. This results in some unwanted distortion of signals, which can in principle be avoided when using electro- or magneto-optic probing systems.

6.2 Development of a prototype electro-optic EMP diagnostic

A diagnostic for measuring strong electromagnetic pulses produced by intense laser-matter interactions was developed and demonstrated for the first time in the petawatt regime. This diagnostic was electro-optic in nature, using polarization state modulation via the Pockels effect to obtain low-noise, time-resolved absolute measurements of pulsed electric fields. A multi-axis optical EMP diagnostic was built as part of an experimental campaign at the Vulcan Petawatt facility (Central Laser Facility, STFC Rutherford Appleton Laboratory) investigating the interaction of intense laser pulses with thin foil and optically levitated [92] targets. The results from the levitated, mass-limited targets and the full set of foil targets will be discussed in Chapter 7. The electric-field components of the EMP were detected on a single-shot basis via the Pockels effect in KDP crystals manufactured for longitudinal Pockels cell modulators, here re-purposed as probes. It is worth noting that the diagnostic could also be converted to measure magnetic fields by simply exchanging the electro-optic crystals with a suitable magneto-optic medium, such as a high Verdet constant glass.

6.2.1 Experimental Setup

The Pockels effect scales *linearly* with applied electric field, hence it is well-suited to absolute field measurements based on the use of sensors with well-known dielectric material properties. In our system, the field-induced time-varying polarization changes on a ~ 500 mW linearly polarized continuous-wave (CW) 532 nm laser beam, split between orthogonally orientated KDP crystals, were detected as intensity modulations via the inclusion of adjustable transmissive linear polarizers placed in the beam-line after each crystal. This technique is known as *polarization state modulation*, and has been previously applied to E-field measurement in many different contexts [217, 275–285]. It has also been used inversely to measure the electro-optic properties of materials by applying a known field [286]. Here, the use of an intra-cavity frequency-doubled Nd:YAG laser with Type I phase-matching ensured a stable, linearly polarized optical probe

source. After transmission through individual KDP crystal “field sensors” and downstream polarizers to modulate the optical intensity, the probe beams were focused and guided through 200 μm core diameter step-index multi-mode optical fibers to a Faraday cage housing a 500 MHz oscilloscope (Tektronix DPO4054) and photodetectors on the far side of the target area, approximately 9 m from the source plasma (~ 8 m of fiber). As the polarizers modulating the light intensity preceded the fiber injection stage, and the propagation path from the laser to the probe crystals was through free space, polarization-maintaining fibers were unnecessary for this design. The diagnostic architecture is shown in Fig. 6.3. The fiber outputs were directly attached to a set of 1 ns rise-time Thorlabs DET10A photodiodes (PDs) to maximize light collection by the active photosensor region and ensure robust optical alignment. A null channel with an identical photodiode and an optical path identical to that of the ‘signal’ channels was also attached to monitor any electrical noise pickup within the Faraday cage or fiber fluorescence.

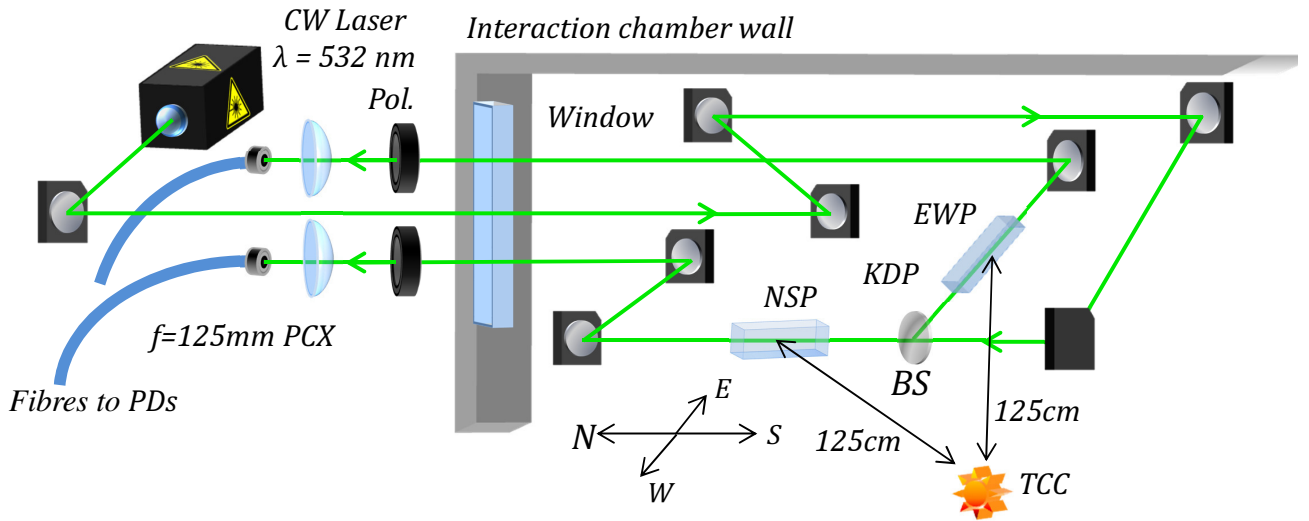


Figure 6.3: Layout of the optical EMP diagnostic in the Vulcan Petawatt interaction chamber, located near the North-East corner of the $2 \times 4.6 \times 2.2$ m steel vacuum vessel. Compass directions are shown for reference. The laser beam was split between the two directional probes using a 50/50 beam-splitter (BS). The prototype diagnostic used $f = 125$ mm plano-convex (PCX) lenses to focus into the fibres for simplicity. A vertical direction probe was also built, but did not provide useful data due to some movement of the chamber under vacuum, hence only the East-West and North-South probes (EWP and NSP) were used, with crystals 1.25 m from target chamber centre (TCC).

The KDP crystals used in the diagnostic were designed for use with longitudinal electric fields, as in KDP first-order relative retardations are only produced by fields along the optic axis [276].

This was tested off-line by applying transverse electric fields to the crystal via two parallel electrodes fed by a high-voltage pulse driver unit (Kentech PBG1) as shown in Fig. 6.4, from which the *vectorial selectivity* (unwanted sensitivity to transverse fields) of the crystals was determined to be > 35 dB, such that any errors on our measurements from Vulcan due to transverse field effects can be considered negligible. It is important to ensure that the electrodes are not ‘small’ relative to the size of the crystal faces to which they are attached, otherwise fringe field effects can result in the presence of an undesired longitudinal field component.

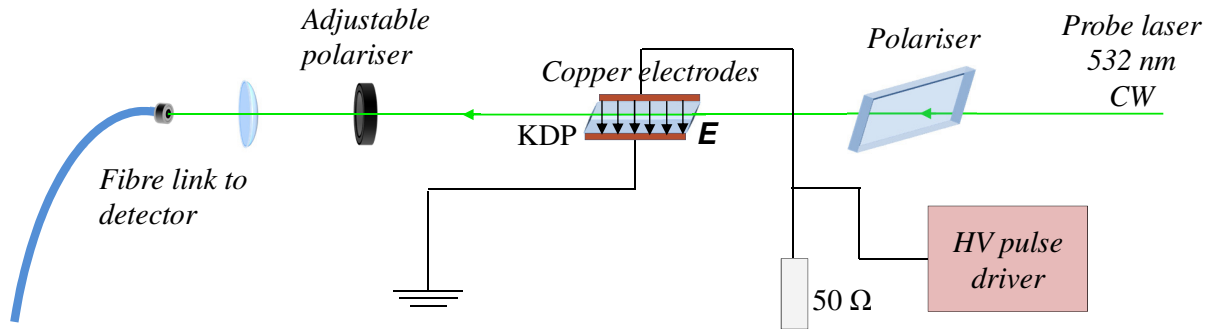


Figure 6.4: Setup for testing the vectorial selectivity of KDP crystals used for electro-optic EMP probing. Copper tape was used for the transverse electrodes above and below the crystal. The high-voltage test pulses were provided by a Kentech PBG1 driver unit driving into a 50 Ω terminator.

The crystals were rhomboidal with lengths, widths and heights of 25, 13 and 9 mm respectively, with end-faces cut at Brewster’s angle to eliminate “ghost” signals from internal reflections. The use of a longitudinal configuration with the laser beam directed along the optic axis is beneficial as it allows single field components to be measured with discrete crystals, and potential recovery of the full 3D field vector using a set of 3 orthogonal crystals. The phase retardation due to a longitudinal electric field E_l within a crystal of length L results in a polarization rotation (in radians) given by

$$\Delta\theta = \frac{2\pi}{\lambda_0} n_0^3 r_{63} L E_l \quad (6.1)$$

where n_0 is the crystal’s linear refractive index, r_{63} is the electro-optic permittivity and λ_0 is

the laser wavelength [276]. Hence, the electric field values are given by

$$E_l = \frac{\Delta\theta\lambda_0}{2\pi n_0^3 r_{63} L} \quad (6.2)$$

The experimental polarization rotation can be determined by comparison of the “null” voltage values with zero-field in the early time section of each voltage trace prior to high-intensity irradiation of a target. As the polarizers were set such that with zero-field, the voltage readings were half of their maximum value, the fractions of PD voltages V_i relative to the mean zero-field voltage V_0 were used to calculate rotation angles for a given voltage value using the relation

$$i = \frac{1}{2} \arcsin \left(\frac{V_i}{V_0} - 1 \right). \quad (6.3)$$

This system is thus sensitive to electric fields of both polarity along the measurement axis; fractions of $V_i/V_0 = 1, 0, 2$ correspond to respective angles of $i = 0^\circ, -45^\circ, +45^\circ$. The time resolution of the diagnostic was limited on our initial experiment by the 1 ns rise time of the photodiodes, which set an upper bound on the resolvable signal frequencies; the quarter-period of the highest measurable frequency is equal to the rise time, yielding an upper frequency limit of 250 MHz. This measurement bandwidth is considerably lower than the bandwidths of state-of-the-art oscilloscopes and photodetectors (~ 100 GHz), however the issue may easily be resolved by using faster but more expensive optical sensors and readout systems matched to a suitably low temporal dispersion fiber transport system.

To the best of our knowledge, this is the first time such a diagnostic has been used to measure EMP from petawatt-regime laser-matter interactions. Other work has attempted to optically sense magnetic fields generated by laser-plasma interaction [253]. A similar, independently developed, system to that described in this thesis was used by Consoli et al. for EMP detection from long-pulse (~ 3 ns FWHM, focused intensity of $\sim 4 \times 10^{14}$ W cm $^{-2}$) laser-plasma interactions [245]; this diagnostic used a different electro-optic crystal (BSO), laser wavelength and setup, with the sensor crystal placed 85 mm from the target. In the work reported here,

the crystals were both placed 1.25 m from the target, with no direct line of sight to mitigate against unwanted detection of X-rays or direct charged particles (for example by scintillation), positioned with their optic axes along orthogonal axes of the interaction chamber. The ability to detect EMP so far away from the target is useful, as diagnostic access near to the target is typically very limited in high-intensity interaction experiments.

While the work of Consoli et al. demonstrated the efficient use of electro-optic EMP measurement techniques in the nanosecond regime [245], the work described here is the first demonstration of absolute measurement of EMP fields by electro-optic methods in a petawatt-class laser-plasma experiment, using a laser providing hundreds of joules of energy. These are the type of experiments where EMP is observed to be most intense [264], leading to well-known difficulties in obtaining reliable measurements of EMP fields within the vacuum chamber, and also creating greater risk to equipment. The region of parameter space accessed during this experiment is significantly different to that of earlier work with nanosecond lasers; with intensities over a million times higher, the interaction physics is considerably altered, with relativistic effects resulting in additional heating mechanisms (resonant absorption [108, 287], $\mathbf{j} \times \mathbf{B}$ heating [116, 288] and Brunel heating [112]) and influencing electron transport or blow-off processes. Due to these factors, it is very important to prove that electro-optic techniques can be successfully applied in the petawatt regime, which is most applicable to experiments driven by relativistically intense, highly-energetic, laser systems such as PETAL, Apollon or the ELI facilities.

Defining the bottom North-East corner of the chamber as the origin of a Cartesian coordinate system (with units in centimeters), the North-South and East-West probe crystals were located at coordinates (56, 142, 181) and (37, 153, 181) respectively, as shown in Fig. 6.5a. The crystal mounts consisted of three aluminium plates of 8, 5 and 10 mm thicknesses enabling fine alignment adjustments, shown in Fig. 6.5b. An additional 8.5 mm-thick plastic layer immediately below each crystal ensured there was no direct contact between crystals and conductive materials. The plate assemblies were attached to a 10 mm-thick stainless steel optical breadboard; these all obscured the direct line of sight between crystals and target, providing significant shielding from direct X-rays and fast charged particles.

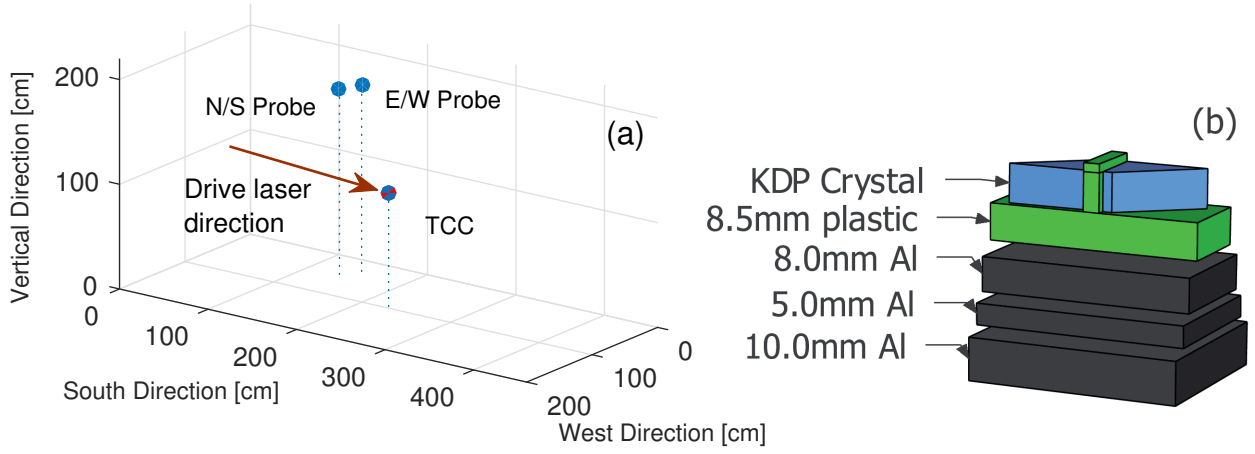


Figure 6.5: (a) Cartesian coordinate plot depicting the location of the KDP crystals within the chamber. The origin is defined here as the bottom North-East corner of the Vulcan Petawatt interaction chamber. Target Chamber Center (TCC), where targets were located, is also shown for comparison. (b) Simplified schematic of the crystal mounts, where the middle two aluminium layers enable fine adjustment and the plastic insulates the crystals from surrounding metals. The aluminium plates were in direct contact with each other, but are shown to be slightly separated here for easier viewing.

The 10 mm breadboard was attached via steel posts to another steel breadboard, with a separation of ~ 12 cm. This helps to reduce the X-ray flux incident onto the crystals (which should follow an inverse-square law), as electrons freed by X-ray photons on the surfaces of electro-optic crystal are thought to contribute to the total E-field across it [245]. Without shielding, this potential noise contribution could appear to be a low-frequency EMP component, despite not actually coming from the RF emission. However, while providing shielding from X-rays and charged particles, the presence of nearby conductive material results in some local perturbation of the field [245, 251].

KDP is a good choice of crystal for this application, as it possesses a reasonably high electro-optic coefficient of $r_{63} = 9.7$ pm/V [289], and does not scintillate strongly in the absence of impurity ions [290], so we can neglect X-ray-induced scintillation as a source of unwanted non-electric field driven intensity changes. Furthermore, by using a free-space setup within the interaction chamber and placing the fiber-injection optics outside, the coupling of optical self-emission from the target and any scintillation from the crystals into the fibers is minimized, and can be resolved as a prompt event coinciding with the arrival of the drive laser, distinct from the longer timescale chamber ring-down and EMP emission.

Isomorphs of dihydrogen phosphates, which include KDP, have been shown to possess clamped electro-optic r_{63} coefficients that are practically independent of laser wavelength across their transparency ranges [291–293] and flat frequency responses in the high-frequency (zero strain) limit up to driver field frequencies of 500 MHz [291], and later ~ 10 GHz [294]. These are therefore ideal for the diagnostic’s broad frequency range, which could resolve signals between 500 kHz to 250 MHz, determined by the optical detectors’ time-resolution and the chosen temporal measurement window. This is related to the piezo-electric and photo-elastic vibrational behavior of the crystal; at high frequencies (above ~ 100 kHz), the crystal motion is inhibited by inertia, yielding the r_{63} value at zero strain [291]. Hence, in the high-frequency limit of the Pockels effect, KDP has the desirable property of a near-ideal flat frequency response, essential for accurate measurement of EMPs that typically contain a broad frequency spectrum and multiple temporal structures. Even in the low frequency (zero-stress) limit (flat below 20 kHz [291], $r_{63} = 10.7$ pm/V [289], hence the change in electro-optic coefficient will only result in a small error in determination of the applied E-field. The transition region between constant stress and constant strain limits lies between ~ 20 -100 kHz, in which there is larger variation in r_{63} , which must be considered if measuring signals at high precision in this low frequency range.

However, there are also disadvantages to this diagnostic architecture: it cannot be easily moved within the chamber without realignment due to the free-space propagation paths, and small movements of the chamber between atmospheric pressures and vacuum could result in some loss of alignment, as the fiber coupling optics were not physically attached to the chamber. As a consequence, the vertical probe beamline lost alignment upon pumping down the chamber in our proof of concept system, hence only field components in the floor plane were measured, the so-called North-South and East-West chamber modes. This problem could be mitigated via all-fiber delivery into and out of the target chamber, with only the laser and detection system placed externally. Preferably, this modified design would use polarization maintaining fibers to allow the in-chamber probing setup to be as compact as possible, in addition to being mobile; further discussion of refinements to the diagnostic can be found in Section 6.5.

6.3 Time-resolved Field Measurements

The experimental results demonstrating the functionality of the prototype EMP diagnostic in this chapter are from a 269 nm thick Parylene-N plastic foil target (1.1 g/cm^3 mass density), with a laser pulse of duration 1.7 ps, center wavelength 1054 nm and 386 J delivered to target (227 TW) and focused to a $4.7 \times 5.5 \text{ }\mu\text{m}$ FWHM spot, with 32.8% of the energy in the FWHM, yielding a peak intensity of $4.8 \times 10^{20} \text{ W cm}^{-2}$. Due to space constraints we were unable to remove the detectors and oscilloscope entirely from the Vulcan PW target area, but we provided shielding of them by using a Faraday cage; however, the few-millivolt EMP-induced ‘noise’ signals on the null photodiode channel were considerably smaller (by ~ 3 orders of magnitude) than those detected from an end-terminated cable inside the interaction chamber attached to another oscilloscope housed in a Faraday cage close to the experiment ($\sim 1 \text{ m}$ from the chamber), which could exceed a volt in amplitude. The cable was identical in length to those connected to other (conductive) EMP probes, and was used to provide information on the ‘noise’ contribution of fields coupling directly to the cable and oscilloscope, for comparison to the noise pickup by the electro-optic setup.

This information demonstrates the high relative noise-immunity of the electro-optic setup in comparison to more conventional methods. Furthermore, much of this unwanted electrical noise was at higher frequencies beyond the diagnostic’s measurement range (as limited by the oscilloscope and photodetectors, which had respective 3dB points of 500 MHz and 250 MHz), approximately between 0.8-1.2 GHz, and therefore most likely an artifact of the oscilloscope electronics coupling directly with EMP. This allowed the vast majority of the noise to be removed by application of a numerical low-pass filter (a finite impulse response 70th order Blackman-Harris window filter with a 250 MHz cut-off frequency), as shown in Fig. 6.6, such that the noise level before and after the arrival of the main laser pulse is approximately the same.

The optical diagnostic successfully measured the EMP’s electric field components within the interaction chamber in both North-South (N/S) and East-West (E/W) directions; the temporal electric field evolutions are shown with and without a numerical low-pass filter applied in Figs.

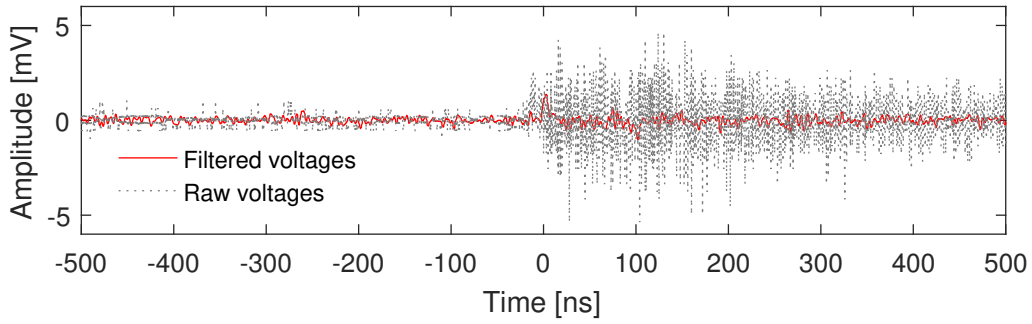


Figure 6.6: The electrical noise measured by the reference channel both with and without a 70th order Blackman-Harris numerical low-pass filter with 250 MHz cut-off frequency applied. The detected noise has frequency content which mostly exceeds the resolvable frequency range of the diagnostic, and hence can be effectively reduced with a low-pass filter.

6.7b and 6.7a respectively. In both probing axes, the EMP signals consisted of rapidly rising peaks of (4.3 ± 0.2) ns (N/S) and (4.0 ± 0.2) ns (E/W) FWHM durations, followed by a decaying oscillation, with reversal of the N/S field direction after ~ 50 ns. The observed decay in field amplitudes is thought to be due to energy radiated out of the chamber through various diagnostic ports and dielectric windows [251, 252]. The finite impulse response 70th order Blackman-Harris window filter with a 250 MHz cut-off frequency (corresponding to the detector bandwidth) was chosen in order to minimize numerical side-lobes, and hence undesirable time-domain Fourier artifacts arising from frequency domain filtering.

A maximum field in the crystal of 10.9 kV/m was measured with the N/S probe within the resolvable frequency range of the sensor system, with a smaller field component of 5.5 kV/m measured along the E/W axis. Both measurements were made at a distance of (1.25 ± 0.01) m from the plasma. The fields decayed to the observable background noise level after ~ 500 ns. This is consistent with predicted electric field strengths within the Vulcan Petawatt target chamber calculated by Mead et al. to be 7.2 kV/m and 16.5 kV/m in the E/W and N/S directions respectively, based on the idea that the main contribution to EMP is driven by electrons ejected from the plasma striking the interaction chamber wall, causing it to resonate with a dominant N/S mode [257]. The measured values are for fields within the dielectric crystals, hence one expects the vacuum field strengths to be higher and thus closer to Mead's predicted field strengths. Otherwise, weaker field strengths might be due to a smaller hot electron flux striking the rear chamber wall during the experiment. It should be noted that

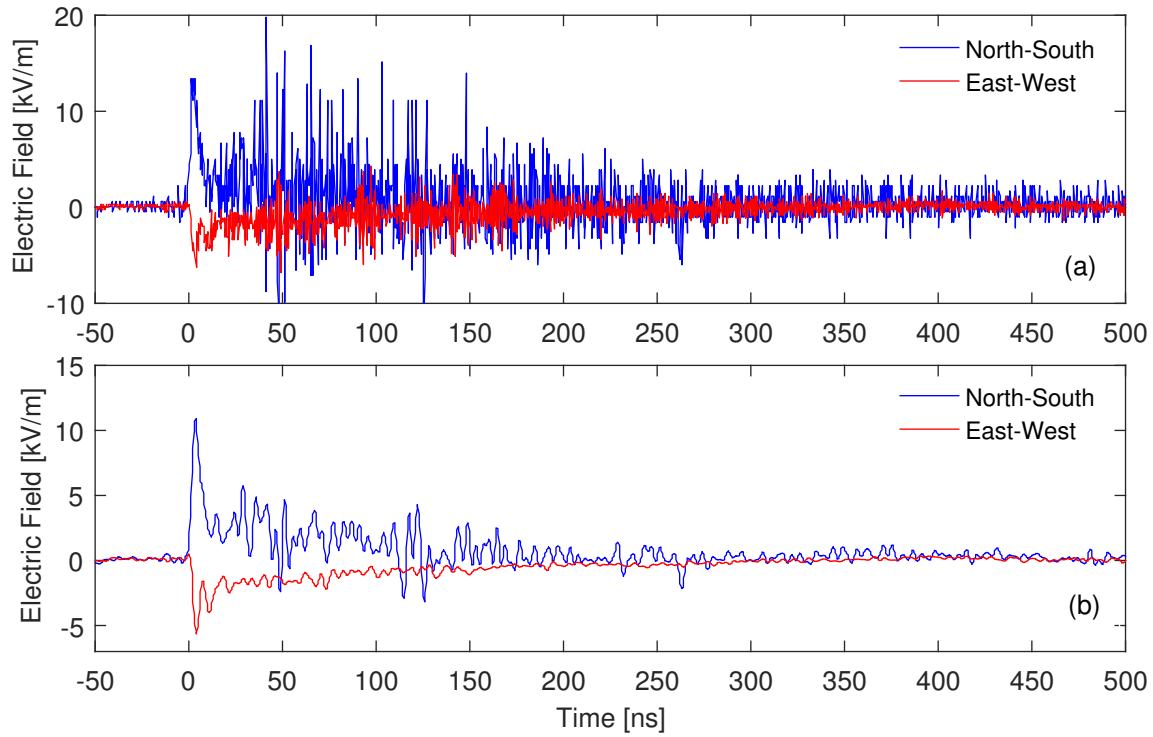


Figure 6.7: Temporal electric field behavior (a) calculated using the raw voltage data and (b) with a low-pass frequency filter applied to remove high-frequency electrical noise, and the contribution to the initial peak by optical self-emission coupled into the optical fibers subtracted. The time axes have been shifted such that $t=0$ corresponds to the arrival of the 227 TW drive laser pulse on target. A 70th order Blackman-Harris window filter with a cut-off frequency of 250 MHz was chosen to minimize side-lobes and hence time-domain artifacts.

the maximum field in the time-domain may be larger, as EMP has been reported previously to contain components up to several GHz, which are not detectable by the photodetector or oscilloscope used here, but would be visible to a higher time resolution system employing the Pockels effect. However, the difference is likely to be small, as can be seen from the very minor change in maximum electric field between unfiltered and filtered Figs 6.7a and 6.7b, but it is important to note that this measurement does not provide reliable information regarding the content of the high frequency signal components.

From background test shots with the probe laser inactive, the small peaks on each channel (including the ‘null’ noise reference) arising from coupling of self-emission from the plasma into the optical fibers are consistently the same size on a given shot, however there is some small variation between shots due to differences in drive laser pulse energies. Hence, by processing the data from the electrical noise reference channel on each shot exactly as if it were a ‘real’

electric field measurement, the apparent contribution of the self-emission to the initial peaks can be determined and accounted for. This method gives apparent E-fields of +1.9 kV/m and +0.5 kV/m on the N/S and E/W signals respectively, which have been subtracted from the initial peaks in Fig. 6.7b.

Due to the single-shot nature of high-energy petawatt laser-plasma experiments, it is generally not possible to gather extensive statistics for a given measurement to reduce uncertainty. The driver-field frequency response of KDP is flat in the range of resolvable frequencies, hence the uncertainty on the measured field is predominantly due to the instrument noise on each measurement including oscilloscope white and sampling noise and residual EMP coupling to the electronics. This can be determined by again examining the apparent E-fields on the noise reference channel to obtain the instrument noise within the time-window of interest, thus incorporating all of the aforementioned noise contributions; with maximum noise amplitudes of ± 0.9 (N/S) and ± 0.3 kV/m (E/W), this corresponds to $\pm 8.2\%$ and $\pm 5.4\%$ uncertainties on the N/S and E/W mode peaks respectively. For weaker signals, the percentage error correspondingly increases, with a value of $\pm 50\%$ on the minimum measurable field values based on a minimum acceptable signal-to-noise ratio of 2. Temperature variation is another potential source of uncertainty in electro-optic measurement techniques, with phase retardation varying by -0.18% /K for KDP at room temperature [276]. In our case however, the measurement was robust as the temperature within the Vulcan chamber is stable within $< 1^\circ\text{C}$.

The minimum intra-crystal electric field observable with this new diagnostic is set by the minimum polarization rotation-induced intensity change resolvable above the experimental noise; this was determined to be 1.8 kV/m for the N/S channel and 0.6 kV/m for the E/W, based on a minimum acceptable signal-to-noise ratio of 2, as analysis of the noise reference channel yielded maximum noise features of 0.9 and 0.3 kV/m for the N/S and E/W probing channels. This could be effectively improved by increasing the laser power reaching the photodiodes, either by optimizing coupling efficiency into the optical fibers, or by simply using a more powerful probe laser. Hence, a given polarization rotation will result in a larger measured voltage change relative to the noise, allowing smaller fields to be measured, thus increasing the diagnostic's dynamic range. As the polarizers in the diagnostic were set such that half of the CW laser power

was transmitted with no electric field applied to the crystals, the maximum measurable field $|E_{max}|$ is set by the field required to rotate the laser polarization by $\pm 45^\circ$ and can be calculated using Eq. 6.2, yielding $|E_{max}| = 79.2 \text{ kV/m}$ using $r_{63} = 9.7 \times 10^{-12} \text{ m/V}$ [289], $\lambda_0 = 532 \text{ nm}$, $L = 25 \text{ mm}$ and $n_0 = 1.514$ at 532 nm . Hence, the useful dynamic range is currently ~ 45 , limited by electrical noise pickup by the detection setup. This could be improved by a factor of ~ 5 before necessitating use of a more advanced recording system with a greater bit-depth than is typical for fast 8-bit oscilloscopes operating in single shot mode.

It is important to note that the above values represent the electric fields inside the crystals rather than the vacuum fields within the chamber. It is possible to calibrate the crystals to external fields using a TEM cell setup [295, 296], however doing this for relatively large longitudinal probe crystals is not trivial, requiring custom apparatus. Typically, we are most interested in relative EMP measurements between shots while varying a parameter such as target thickness or pulse energy, however an approximation for the radiated vacuum fields can be calculated using the method described by Massey et al. [276]; this analysis approximates rectangular crystals as ellipsoids of revolution about the optic axis and only requires knowledge of the crystal aspect ratio a (the ratio of the longest and shortest axes) and dielectric constant ϵ along the optic axis.

The analysis uses the Poisson equation to estimate E-fields under the electro-quasistatic approximation, where times of interest t_1 (here the period of a frequency component) must be much longer than the time t_2 required for an electromagnetic wave to propagate at the velocity c/n (c is the speed of light, n is the refractive index of the propagation medium) over the largest length of the system [297], in our case the crystal length L . Since the largest frequency that the electro-optic diagnostic can measure well is 250 MHz , the smallest t_1 is 4 ns , therefore the condition is satisfied as $t_2 = 382 \text{ ps}$ for a propagating 250 MHz E-field. The vacuum field strength E_0 is related to the intra-crystal field E_{crys} by

$$E_0 \approx \frac{E_{crys}(D_3^{-1} + \epsilon - 1)}{D_3^{-1}} \quad (6.4)$$

where D_3 is the depolarization factor [298] for the optic axis, the major axis of the ellipsoid of revolution given by

$$D_3 = b^{-1} \left(\frac{1}{2} ab^{-1/2} \ln \frac{a + b^{1/2}}{a - b^{1/2}} - 1 \right) \quad (6.5)$$

where we have defined $b = (a^2 - 1)$ for convenience. For the crystals used here, $a = 2.78$ and $\epsilon = 21$, [299] yielding $D_3 = 0.11924$ and hence an approximate conversion relation between vacuum and intra-crystal fields of $E_0 \approx 3.38 E_{crys}$, such that we can thus estimate the maximum radiated field strengths to be approximately 37 and 19 kV/m in the N/S and E/W axes. This can be compared to an estimated free-space to crystal coupling parameter of ~ 2.84 based on measurements using a pulsed open-capacitor setup (Fig. 6.8), where the crystals placed between the two parallel capacitor plates were subject to a ~ 9 kV pulse-amplitude longitudinal electric field. This coupling parameter gives corresponding vacuum fields of approximately 31 and 16 kV/m, in reasonably good agreement with the values predicted from electro-quasistatic theory. The high voltage pulses were produced using a Kentech PBG1 Pockels cell driver, with Indium Tin Oxide (ITO) coated PET sheets, separated by 10 cm, acting as transparent planar electrodes. The electrode dimensions were $250 \times 200 \times 0.2$ mm, such that the parallel plates were ‘large’ compared to the crystal dimensions to avoid any fringe-field effects and maximize the uniformity of the longitudinal calibration field.

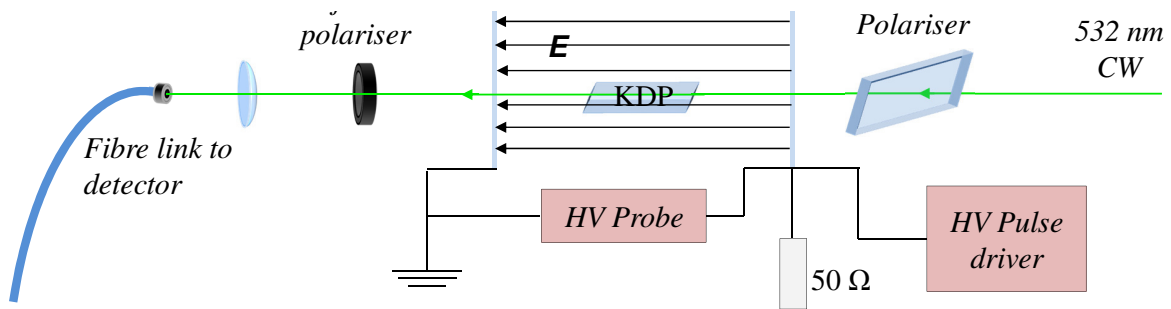


Figure 6.8: Setup for testing the free-space to intra-crystal field coupling of KDP used for electro-optic EMP probing. ITO-coated PET sheets were used as transparent electrodes, allowing a longitudinal calibration setup to be implemented relatively easily.

It should be noted that if the fields are sufficiently strong to either extinguish the laser or double the voltage measured by the photodiodes, there is a potential ambiguity in the measurements

as fields greater than $|E_{max}|$ will induce polarization rotations greater than $\pm 45^\circ$, which will result in a change in laser intensity with the opposite sign to that which is expected. Hence, it is essential that users are aware of this ambiguity if the change in photodiode voltage approaches the magnitude of the zero-field voltage ($V_i \rightarrow 2V_0$ or 0) while running the diagnostic. This problem can be compensated for by changing the electro-optic crystal to obtain different r_{63} and n_0 values, or alternatively by changing the laser wavelength; by selecting a lower r_{63} crystal or a longer wavelength, the polarization rotation for a given electric field strength can be reduced, allowing the maximum measurable field value to be increased before the aforementioned ambiguity becomes significant. The crystal length is also a key parameter for selecting the range of measurable fields, with increasing length leading to higher linear sensitivities at the expense of a loss of ultimate time resolution and precise spatial localization of the detector ‘position’.

6.4 EMP Spectral Analysis

The Vulcan interaction chamber is a conductive cuboid, with 2, 2.2 and 4.6 m separations between steel walls in the East-West, vertical, and North-South dimensions respectively; treating the chamber as a simple parallel cavity resonator supporting Transverse Electric (TE) modes, the fundamental resonance frequencies are set by the chamber geometry. These correspond to resonances of 76, 82 and 101 MHz in the E/W, vertical and N/S dimensions respectively [257]. It is important to note that these frequencies are expected based on an empty interaction chamber, and thus are likely subject to some considerable modification by in-chamber hardware including metallic optical tables and optomechanics. The optical diagnostic detected the fundamental resonances of the interaction chamber at (76 ± 4) and (101 ± 2) MHz, although not on the expected probe channels. Frequency content at (149 ± 5) and (225 ± 6) MHz was also measured, matching the predicted 2nd and 3rd harmonics of the E/W mode (152 and 228 MHz), albeit in the N/S probing direction; these are summarized in Table 6.1. A spectrum is shown in Fig. 6.9, obtained by Fast-Fourier Transform of the low-pass filtered time-domain E-field data.

Expected Frequencies [MHz]	Measured Frequencies [MHz]		
	<i>EO-CH1 (N-S)</i>	<i>EO-CH2 (E-W)</i>	<i>D-Dot</i>
76	76 ± 4	Not detected	Not detected
101	102.7 ± 0.6	101 ± 2	Not detected
152	149 ± 5	Not detected	148.5 ± 4.5
202	Not detected	Not detected	Not a sharp peak
228	225 ± 6	Not detected	Not a sharp peak

Table 6.1: Frequencies of the expected harmonics and detected spectral peaks

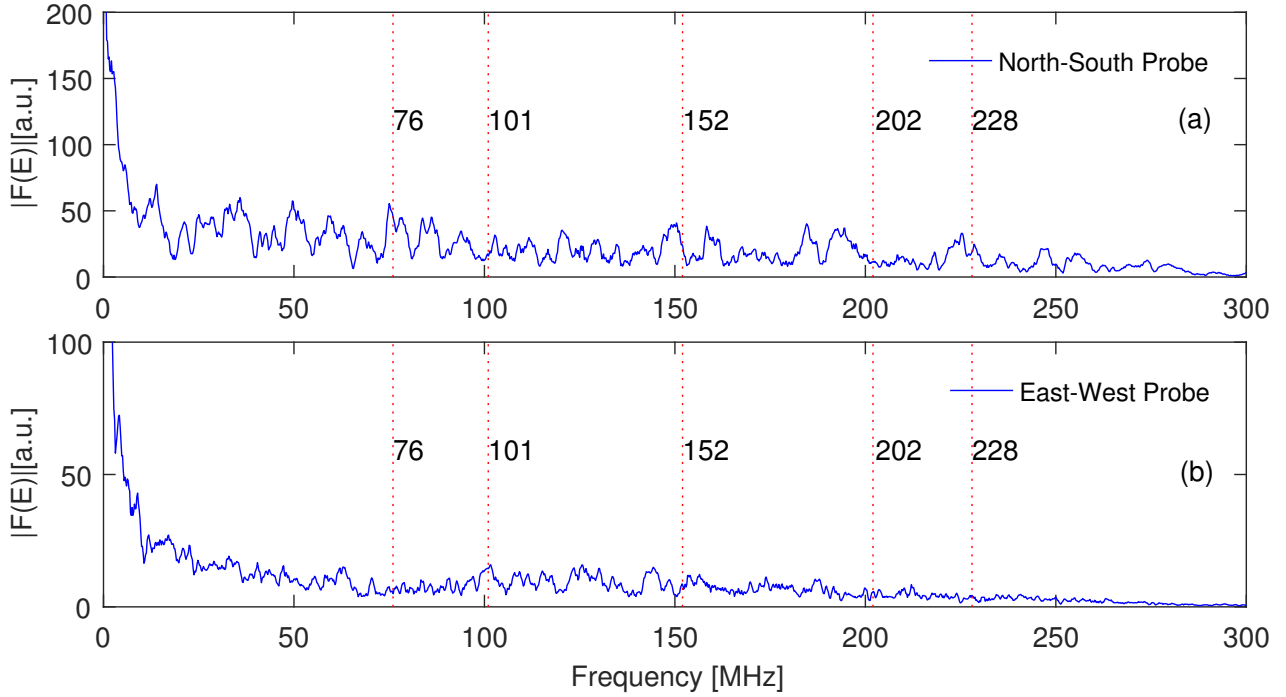


Figure 6.9: Frequency spectra of the EMP with the 250 MHz cut-off low-pass filter applied from (a) the North-South and (b) East-West probes. Several peaks are visible in the spectra, with some corresponding extremely well with the theoretical fundamental empty chamber resonances and their harmonics (marked on the plot). The large quantity of conductive materials within the interaction chamber is expected to allow several other resonances to be supported, to which we attribute the presence of the other spectral peaks. A 1st degree Savitzky-Golay filter was applied in the frequency domain for noise-reduction.

Interestingly, the expected E/W fundamental mode frequencies were detected more strongly by the N/S probe, which also seems to have a narrow peak at (102.7 ± 0.6) MHz, near the N/S fundamental of 101 MHz. This will be discussed further in Chapter 7. The peak field magnitude detected by this probe was approximately twice that of the orthogonal field component, and some of the frequency content in this dimension is likely to have been up- or down-shifted to other frequencies by the presence of numerous metal objects throughout the

chamber (such as the silver main focusing parabola optics mounts, breadboards and a range of other diagnostics), which allow multiple additional resonances and their harmonics to be supported [251], potentially with similar spectral amplitudes to the fundamental cavity modes as observed in our case. Furthermore, the rapid transient discharge current through the target holder, as described by the Poyé model [264, 265], will result in electromagnetic field emission at a specific range of frequencies not necessarily corresponding to the chamber fundamentals, which in turn may also be frequency-shifted.

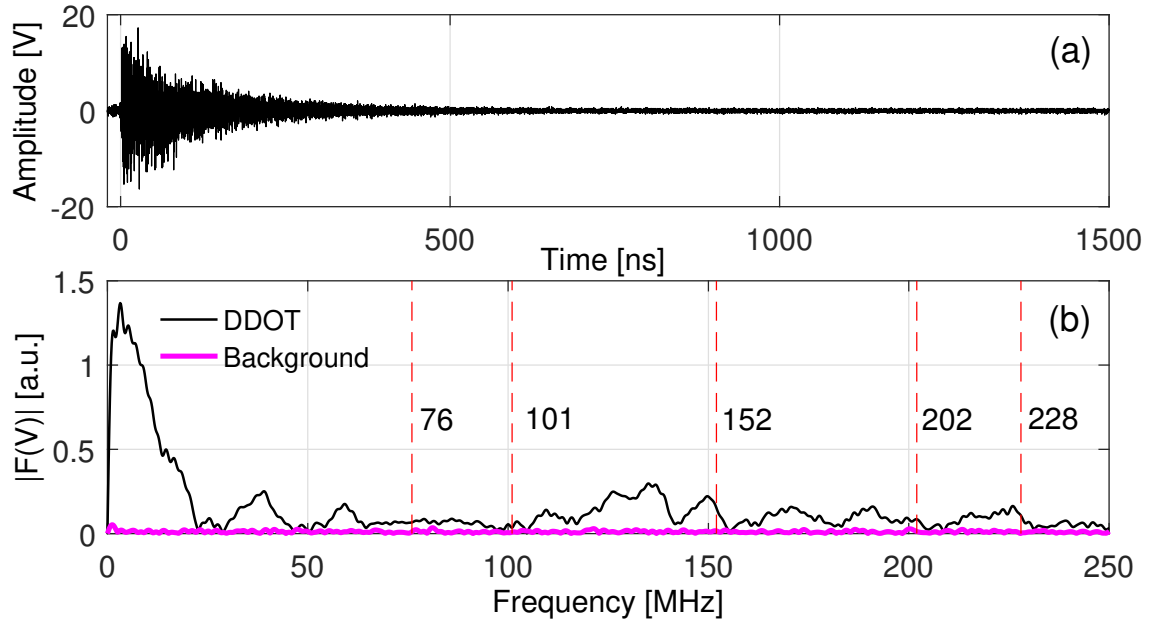


Figure 6.10: (a) Time domain signal measured by the D-Dot probe. (b) Spectrum in the 0-250 MHz frequency range for associated EMP and background noise signals. Harmonics corresponding to those theoretically expected in chamber and listed in Table 1 are outlined here by dashed red vertical lines. The D-DOT probe was installed and operated by F. Consoli and R. De Angelis.

In Fig.6.10 we show the signal acquired with a 12.5 GHz Tektronix DPO71254C oscilloscope, from a customized commercial D-Dot conductive probe (installed and operated by F. Consoli and R. De Angelis), placed within the experimental chamber at the coordinates West: 72 cm, South: 4 cm, Vertical: 160 cm, of the reference system in Fig.6.5, behind a ~ 10 cm thick silver-coated glass mirror with respect to the target. This protected it from direct line of sight to the majority of X-rays and particles emitted from the target. The probe was a customized version of the Prodyn AD-80(R), with 5.5 GHz of 3 dB bandwidth. It was equipped with a balun (model BIB-170G), having 150 kHz-10 GHz of 3 dB bandwidth and leading to common

mode rejection > 20 dB. The probe had a dual-dipole structure with high rejection to common mode disturbances. It was set in a vertical plane (edge on to the target) such that both dipoles were exposed to residual hard X-rays coming from the laser-plasma that would, to first order, generate a common mode effect on the probe, thus canceling or at least reducing the common mode signal.

The spectrum was obtained by applying the Fast Fourier Transform (FFT) to the first 600 ns of the signal of Fig. 6.10a and it is shown in Fig. 6.10b in the 0 – 250 MHz range, to compare these results with those already discussed for the electro-optical measurements. In Fig. 6.10b an estimation of the background noise spectrum is also shown. This was achieved by applying the FFT to the signal in Fig. 6a on the (600 ns, 1200 ns) interval, where the EMP contribution had already ceased. The spectrum of the EMP signal detected by the D-DOT is well beyond the background level for most of the observed frequency range. Harmonics corresponding to those theoretically expected within the chamber and listed in Table 6.1 are outlined with dashed red lines. Information on measurements associated with these resonances is reported within the same Table. Good correspondence with both the expected theoretical values and electro-optic measurements was found for the 148.5 MHz contribution; some agreement can be observed also for both the 202 MHz and the 228 MHz harmonics.

Time-frequency analysis was also carried out on the electro-optic data using the short-time Fourier transform technique spectrogram MATLAB function; this provides insight into the temporal behavior of the various frequency components and energy transfer between different chamber modes. Spectrograms for each measurement channel are shown in Fig. 6.11. The different chamber fundamental modes are apparent in the early time regions of the spectrograms from the bright regions separated in frequency space, however these quickly decay below the minimum measurable field level of the E/W probe (Fig 6.11)b, consistent with the much weaker signals measured along that axis. The time-frequency analysis will be considered in more detail in the next chapter, in Section 7.2.2.

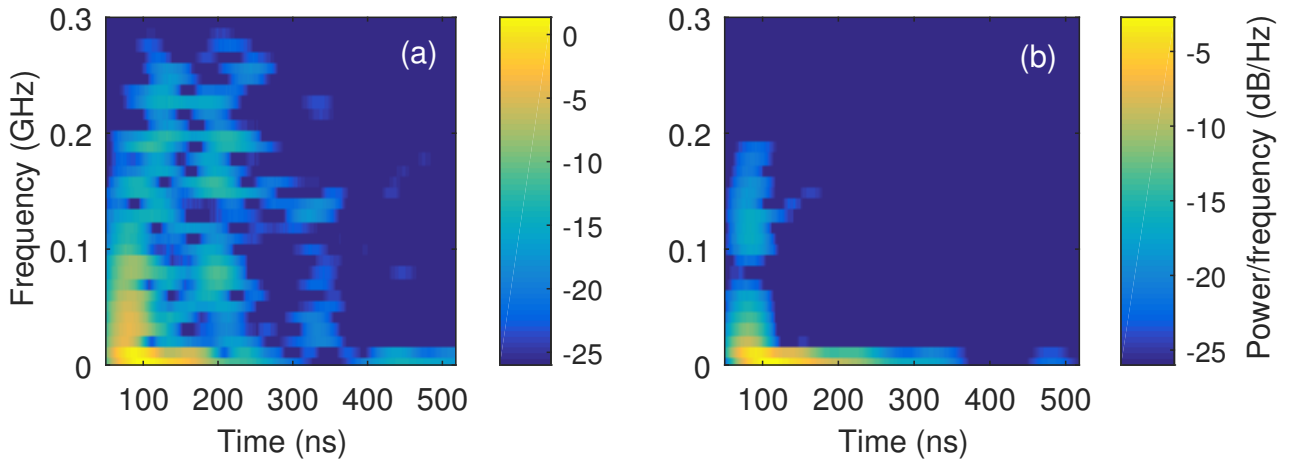


Figure 6.11: Spectrograms depicting the temporal changes in the spectral behaviour of the low-pass filtered electric field traces from shot #29 for (a) the N/S probe and (b) the E/W probe. As the channels measure orthogonal fields, different frequencies are strongly detected in the early time region, some of which match the chamber fundamental modes. More detail on the time-frequency analysis will be provided in the next section.

6.4.1 Comparability of different EMP diagnostics

It is generally difficult to make reliable comparisons between different EMP diagnostics unless they are in identical locations; the presence of numerous metal objects within an interaction chamber results in complex spatially-varying EMP field topologies [251], such that field strengths and relative strengths of the spectral components can vary greatly between different positions within and outside of the chamber. Moreover, diagnostics of different nature often have different detection efficiencies, in terms of bandwidth and polarization. These issues are further compounded by inherent shot-to-shot energy variation and movement of diagnostics and other metallic objects between shots, such that recreating the same conditions if placing probes in identical positions on different shots is non-trivial in practice.

The physical mechanism used to measure fields will also impact on the time-domain electric fields obtained from a diagnostic; an example of this is conductive probes, which are typically sensitive to the time-derivative of the fields, compared to electro-optic probes, which provide absolute measurements. Sensitivity to the time-derivative implies that DC or static fields are not well-reproduced, with the high-frequency components effectively amplified by the measurement. In contrast to this, an electro-optic measurement will capture both the low and high frequency behavior of a field, assuming the optical detectors have sufficiently high time-

resolution, hence $\mathbf{E}$ -field measurements from the two different types of diagnostic are liable exhibit different features more prominently.

6.5 Diagnostic Modifications and Upgrades

There are some diagnostic design improvements that could be implemented relatively easily on the proof-of-principle device. Firstly, delivery of the laser light into and out of the chamber via optical fiber would allow the device to be more mobile within the interaction chamber, thus minimizing any space constraints with simultaneous use of other diagnostics. The dynamic range of the diagnostic can be significantly improved via two methods: reducing electrical noise, and increasing the incident laser power on the diodes through both optimized fiber injection and higher initial laser power by a factor of ~ 5 before necessitating the use of a > 8 bit detection system. For optimum noise reduction, complete removal of detectors and oscilloscopes from the target area into a Faraday cage in a well-shielded room is recommended; as there can be no electrical noise pickup from the optical fibers, this should allow significant signal-to-noise ratio improvements compared to conventional “all electrical” EMP diagnostics. Note that the optical fiber modal dispersion should be considered if using multimode fibers over ‘long’ distances, such that the maximum possible time-delay between modes is smaller than photodiode rise times.

The diagnostic’s time-resolution and bandwidth could be improved by simply using faster photodiodes and oscilloscopes, though this is primarily a matter of cost rather than technical capability. However, it should be noted that the propagation time through the crystals can also limit the ultimate time resolution, as the beam effectively sees the ‘average’ field over this time frame, hence it is unnecessary to use photodiodes with shorter rise times than the crystal transit time. For the 2.5 cm long KDP crystals, the transit time for 532 nm light is ~ 126 ps (3.5 GHz bandwidth), hence the time resolution could be increased by almost an order of magnitude by only replacing the photodiodes and oscilloscope. The fastest commercially available photodiodes for visible light have rise times of ~ 9 ps and 45 GHz bandwidths (Newport Model 1014), therefore a crystal length of 1.8 mm would be necessary to match ~ 9 ps time resolution,

at the expense of reduced sensitivity. If the crystal type was changed for operation in the mid infrared (1550 nm), just outside of KDP's transparency range, 100 GHz photodetectors could be purchased (Finisar XPDV412xR).

Deuterated KD*P crystals are transparent between 200-2100 nm and, as dihydrogen phosphate isomorphs, offer similarly flat frequency-response properties as KDP but with higher electro-optic coefficients. It should be noted that the fastest oscilloscopes are limited to bandwidths of ~ 100 GHz, hence a hypothetical diagnostic using such a setup would be approaching the limit of available direct electronic measurement technology. The quarter-period of a 100 GHz photodiode is 2.5 ps, so assuming the rise-time matches the quarter period of the maximum resolvable frequency component, a KD*P crystal thickness of 520 μm (commonly manufactured) would be necessary to maintain time resolution. However, the sensitivity of such a short crystal to electric fields would be considerably reduced (by 98% compared to a 25 mm crystal), and hence would only be suited to measurement of very strong electric fields, which might be found closer to the target chamber center. Furthermore, increasing the bandwidth of oscilloscopes also typically increases the intrinsic noise, resulting in poor signal-to-noise ratios, though time-frequency analysis as described in this chapter can allow some discrimination between real features and artifacts. An electro-optic probing system for EMP using two complementary diagnostics can be considered; one very fast but with low sensitivity, and another with far higher sensitivity but correspondingly reduced bandwidth. Alternatively, a non-oscilloscope based readout system could be considered for very high time resolution applications, for example through use of a streak camera, at the expense of a reduced temporal measurement window.

Finally, a useful design change would be to replace all metal components in the diagnostic architecture with dielectric counterparts. This would minimize local electric field perturbations from the optomechanical setup for optimal measurement of the original fields at the diagnostics location. Implementation of all the aforementioned upgrades would become costly when attempting to operate at frequencies approaching 100 GHz, and hence is only realistic for large national facilities if such high frequency measurement was essential. However, the system described here demonstrates the intrinsic robustness and promise of the electro-optic EMP measurement principle.

A refined EO-diagnostic architecture, built for use in a laser-levitated target experiment with Cerberus, is shown in Fig. 6.12. In this model, the laser beam is delivered to the crystal and removed from the interaction chamber via optical fiber, while the majority of the metal components, such as the optical breadboard, have been replaced with custom 3D printed dielectric counterparts. These modifications increase diagnostic mobility, robustness of alignment, and help to reduce local field perturbations by the diagnostic setup. A polarizer is placed after the input fiber collimator, such that the CW beam passing through the KDP crystal is linearly polarized. The angular orientation of this polarizer is set to maximize transmission. This could be further improved if polarization maintaining fiber was used on the input. Upon passing through the crystal, any polarization changes are mapped onto intensity modulations via the second polarizer, removing the need for polarization-maintaining fiber to carry signals to the photodetectors.

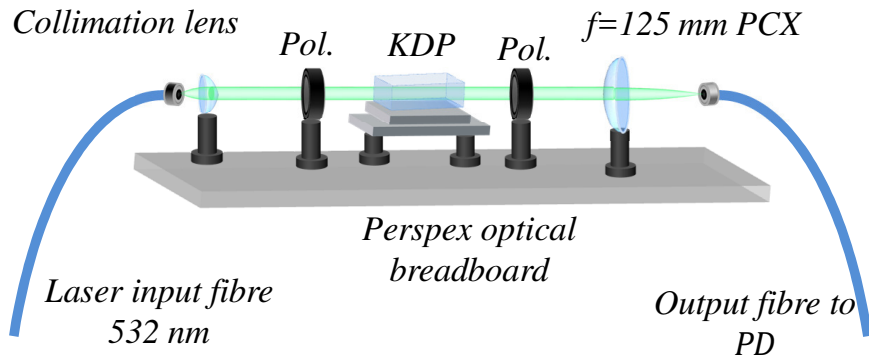


Figure 6.12: Second generation version of the electro-optic EMP diagnostic used for experiments using the Cerberus laser at Imperial College. The laser is delivered into and out of the interaction chamber via optical fiber, allowing the diagnostic to be moved around without loss of alignment. Where possible, optomechanics have been replaced with custom 3D-printed dielectric counterparts for minimal perturbation to the local E-field.

6.6 Conclusion

This chapter describes the first electro-optic measurement of EMP from petawatt-regime short-pulse laser-plasma interactions with low electrical noise background and no requirement for direct-view or close proximity to the target. In light of new ultra-high intensity laser systems,

such as PETAL, Apollon and the various ELI facilities, coming into operation in the near future, it is important that we can demonstrate the applicability of devices with high inherent noise-immunity capable of measuring EMP in this specific regime of extreme laser-plasma physics. Not only is the EMP particularly problematic in experiments using such high-intensities (such as ion acceleration or fast-ignition inertial fusion schemes), this area is where interest is at a maximum from within this research community. Maximum field components of (10.9 ± 0.9) and (5.5 ± 0.3) kV/m were measured within the crystals at a distance of 1.25 m from the plasma (with external fields estimated to be ~ 31 -37 and ~ 16 -19 kV/m, depending on whether the experimental or theoretical conversion parameter is used), and the fundamental chamber resonances at 76 and 101 MHz and some of their harmonics were detected, in good agreement with previous EMP studies at Vulcan using conductive probes and theoretical models.

The optical diagnostic is inherently resistant to the electrical noise that plagues conventional EMP measurement techniques using conductive probes and cables, and hence provides an attractive alternative for monitoring EMP at high-power laser facilities. Relatively easily attainable performance increases in resolution, bandwidth and noise reduction are possible by increasing the CW probe laser power, using faster photodetectors, changing the electro-optic crystal lengths and improved electrical shielding or remote operation of the detection apparatus. Furthermore, the diagnostic setup could be adapted for magnetic field detection by replacing the electro-optic KDP crystals with a suitable magneto-optic medium such as a high-Verdet constant glass.

The diagnostic could also be made more mobile and compact by introducing fiber transport into and out of the interaction chamber, a desirable property as diagnostic access around target-chamber center is often very limited. Some of these modifications were implemented in order to use the updated diagnostic for a Cerberus experiment, and will be further refined to produce EMP monitoring systems for use at Rutherford Appleton Laboratory.

Chapter 7

EMP generation from novel targets

This chapter presents an analysis of the laser-plasma experiments carried out at Vulcan Petawatt and using the Cerberus laser system, based at Imperial College London. Both systems have relatively similar OPCPA front-end architectures, with comparable temporal contrast properties at each facility [174]. The effects of varying the target geometry on EMP from high-intensity laser-plasma interactions will be considered, including results from shots using mass-limited, optically-levitated micro-targets [92]. The discussion in this chapter uses material from work published by the author in the conference proceedings for *Plasma Physics by Laser and Applications (PPLA)* 2017, in the *European Physical Journal: Web of Conferences* [300], and directly follows on from Chapter 6 and [244], which describe in detail the experimental setup and methodology for EMP measurement used to obtain the Vulcan PW results presented in this chapter. Another article describing results from the Vulcan PW experiment, co-authored with F. Consoli and colleagues from ENEA-Frascati, has been recently submitted to *Nature Communications*.

The role of target geometry in EMP generation has not been extensively studied, with few published results on this topic. Poyé describes a dependence on the surface area of planar targets due to lowering of the potential barrier experienced by electrons as charge redistributes across the surface [264, 265], but there are no published results using unconventional targets such as ultra-thin plastic foils or mass-limited isolated spheres. The EMP from a laser-target

interaction is expected to depend on the electron temperatures reached within the plasma, as hotter electrons are more likely to overcome the barrier potential and escape from the ionized targets [264, 265]. In this work carried out at Vulcan Petawatt and using Cerberus at Imperial College, we consider the effects on EMP of varying the thickness of foil targets, and using levitated micro-targets trapped in-vacuo using the optical levitation trap developed by Price et al. [92]. A summary of the key experimental shot parameters from the Vulcan experiment, for shots on which EMP signals were measurable, is presented in Table 7.1.

Shot #	Target Material	Thickness [nm]	Pulse Length [ps]	Energy on-target [J]	Peak Intensity [$10^{20} \text{ W cm}^{-2}$]
14	Parylene-N	550	1.0	372	14
18	Parylene-N	269	1.1	369	12
19	Formvar	20	1.3	342	9.7
20	Parylene-N	550	0.98	359	14
25	Parylene-N	550	1.0	319	11
29	Parylene-N	269	1.7	386	5

Table 7.1: Shot information for shots with measurable EMP data from the Vulcan PW experiment (April/May 2016). A $\pm 10\%$ uncertainty is assumed in the pulse duration measurements provided by the Vulcan laser team. Peak intensities are calculated using the focal spot images taken prior to each shot, as well as pulse energies and durations.

The refined EO-probe design, previously shown in Fig. 6.12 (Chapter 6), was deployed for EMP measurement during the Cerberus experiment studying laser-interactions with levitated mass-limited targets and more conventional thin metal foils and wires.

7.1 Time-domain electric field analysis

7.1.1 Solid Targets

Electro-optic EMP measurements were obtained for 6 shots using plastic thin-foil targets of a range of thicknesses. These shots were all acquired at Vulcan PW. In the time-domain, the traces in each case look relatively similar, with a sharp initial rise in electric-field, followed by an oscillating decay over approximately 300 ns. The measured intra-crystal electric fields are shown for comparison in Fig. 7.1, where the peaks have been corrected for the contribution

from optical self-emission based on the reference channel traces using the process described in Section 6.3.

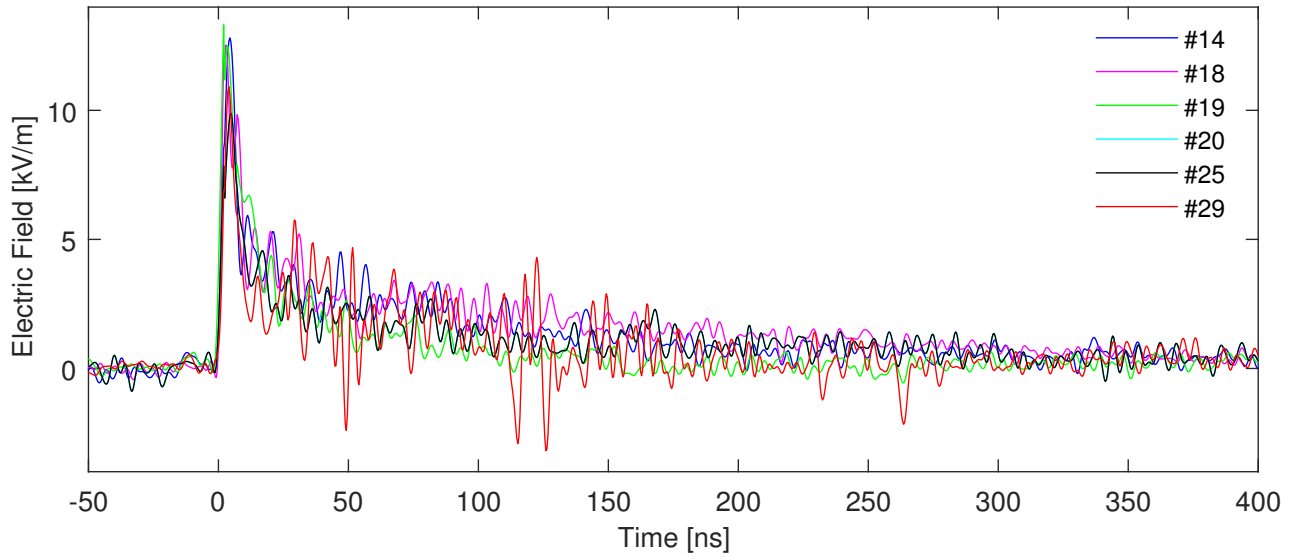


Figure 7.1: Time-domain intra-crystal electric fields measured electro-optically at Vulcan Petawatt. The data from all shots with detectable EMP are shown for comparison. There is a fairly high degree of consistency between the shots, with the main difference being slightly varying peak intra-crystal E-field values.

While in general the measured EMP signals are similar, the results indicate that the peak EMP fields generated during the interaction might scale with the target thickness, potentially suggesting a weak trend towards peak fields increasing with decreasing thicknesses, as seen in Fig. 7.2. However, due to the small number of shots with EMP data from this experimental campaign (particularly for the most thin targets), this result is not conclusive, and more data from the electro-optic diagnostic is necessary to confirm this interpretation. It is interesting to note however that easurements of EMP from the same experimental campaign using a D-DOT probe, obtained by F. Consoli and presented in an upcoming paper to be submitted to *Nature Communications* [301], also show an increase in EMP with decreasing target thickness. Conversely, the drive pulse energies on target do not seem to strongly affect the peak E-fields detected by the EO-EMP diagnostic, at least within the small energy range and target thicknesses considered, shown in Fig. 7.3.

This observation might be explained by a limited version of the enhanced electron heating that occurs in the relativistic regime, where the free electron skin-depth (as seen by the drive

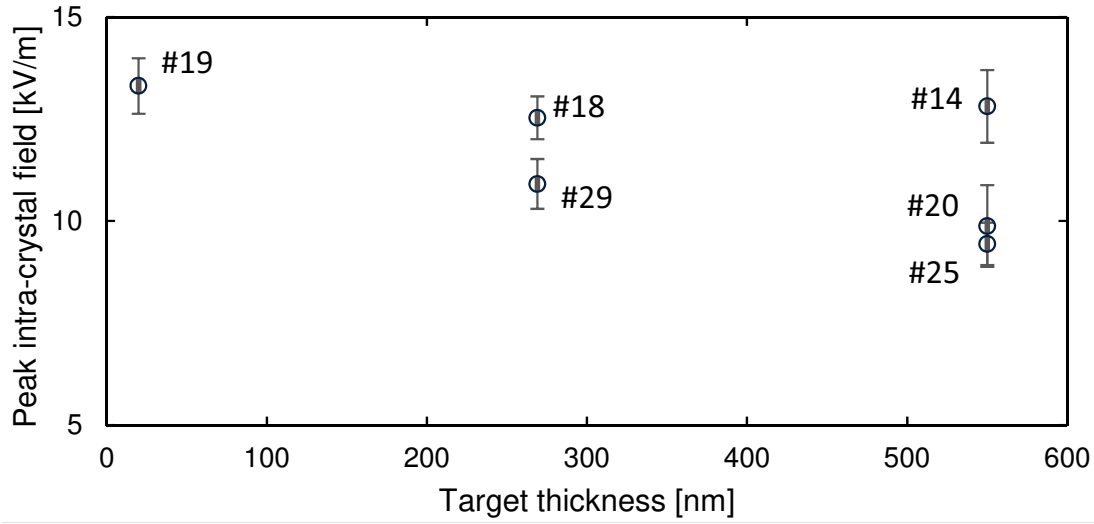


Figure 7.2: Variation of the peak-intra crystal electric field detected along the Vulcan N/S direction with the target thicknesses. The target material was Parylene-N for all shots except for 20 nm, which used Formvar - shot numbers are given as annotations. The error bars are determined by the noise level on a given shot, where we have defined the length of error bars to be the apparent E-field obtained by processing the reference noise channel data as a ‘real’ field measurement.

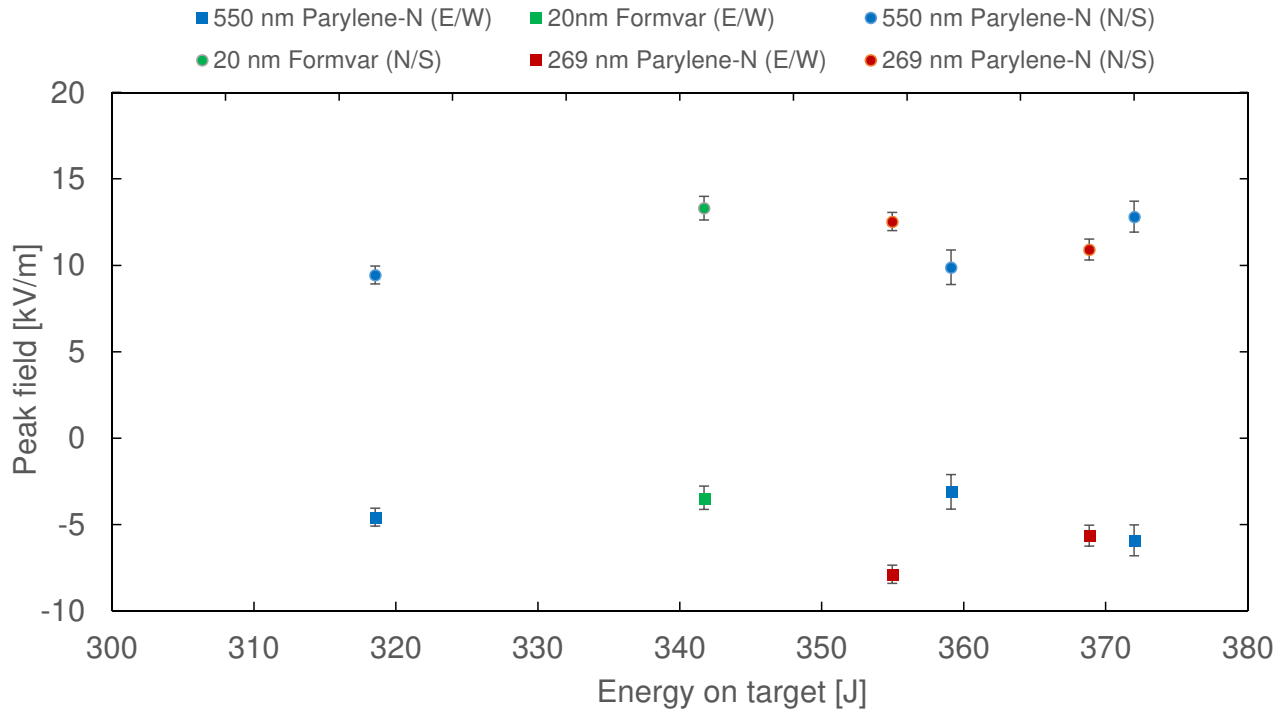


Figure 7.3: Variation of the peak-intra crystal electric field detected along the Vulcan N/S direction with the drive energy on target. The target material was Parylene-N for all shots except for 20 nm, which used a Formvar target. The error bars are found in the same way as in Fig. 7.2.

laser) is comparable to the target thickness. Even in an over-dense plasma, where fields do not propagate beyond a very short range evanescent wave of order the skin depth, if the target is sufficiently thin, the laser field still exerts a ponderomotive force on the electrons within the plasma, resulting in rapid and efficient heating of these electrons. As the electrons gain energy, the plasma can become relativistically transparent, allowing the laser fields to continue propagating into the target and allowing the electrons to be driven further. These effects have been observed for ultra-thin targets in both simulations and experiment, during investigations of ion acceleration in the relativistic transparency [141, 151, 152] or laser break-out afterburner regimes and is typically referred to as ‘enhanced TNSA’ [60, 126, 147, 154, 302]. However, these regimes require very high temporal pulse contrasts to become properly accessible; experiments by Fuchs et al. on ultra-thin foils required typical contrast ratios of $\sim 10^8$ - 10^{12} [60, 302]. With poor contrast, the likelihood of an ultra-thin target surviving the shock driven by the pre-pulse and maintaining sufficiently high electron densities is reduced.

As no plasma-mirrors were used for contrast-enhancement during the Vulcan experiment described here, it is highly unlikely that the contrast would be sufficiently high to reach the levels of enhanced TNSA described in the literature, which is consistent with the weak enhancement of EMP peak fields with decreasing target thickness. Hence, it is important to examine any available contrast data from Vulcan and Cerberus. A pulse contrast scan from the Cerberus OPCPA front-end, measured immediately before the experimental campaign investigating laser interactions with levitated micro-targets and solid foils, is given in Fig. 7.4, obtained using the cross-correlator diagnostic developed by N. Stuart [44, 174]. This shows the contrast ratios to be $\sim 10^6$ - 10^7 over timescales of hundreds of picoseconds, and $10^5 \approx 83$ ps prior to the main pulse.

The pulse located ≈ 48 ps prior to the main-pulse is likely to be a diagnostic artifact, generated by post-pulses on the second harmonic probe, which could be confirmed using a 3rd order correlator using Cross-Polarized Wave Generation (XPW) for probe pulse cleanup, as demonstrated by N. Stuart for a previous Cerberus contrast scan [44], leading to the disappearance of a ‘ghost’ pre-pulse measured at ~ 50 ps when using an XPW-enhanced cross-correlator. Such ‘ghost’ pre-pulses typically exhibit close temporal symmetry around the main pulse, except that the apparent pre-pulse feature will have a few orders of magnitude lower intensity due to

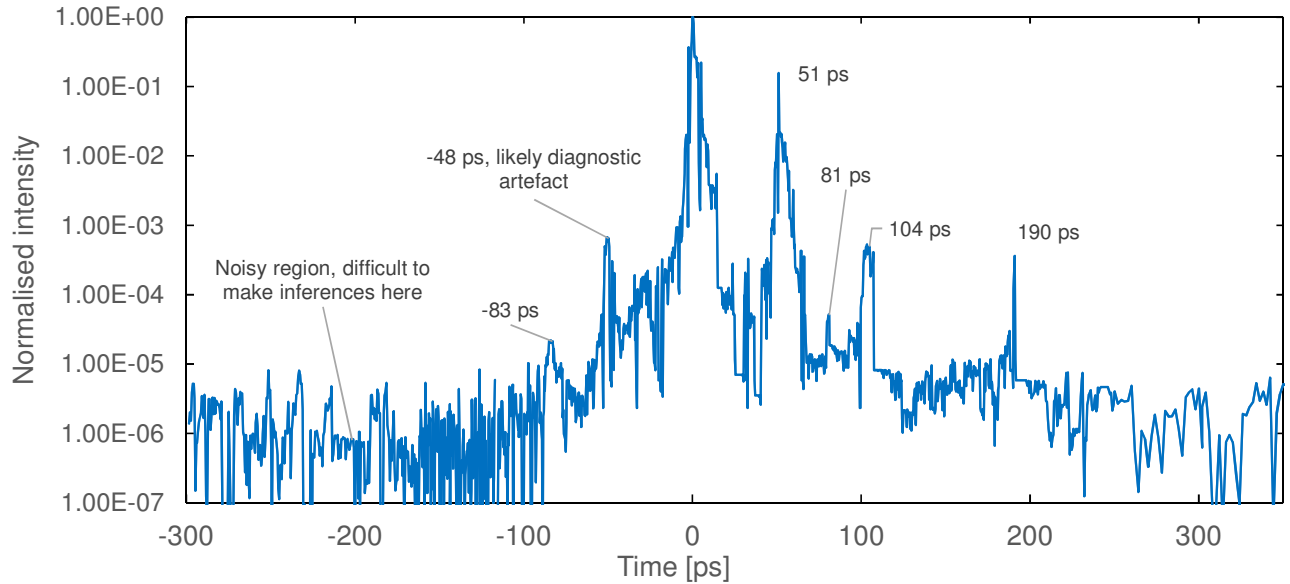


Figure 7.4: Pulse contrast scan of the Cerberus front-end using a third-order correlator diagnostic (developed by N. Stuart and used in [44, 174]), taken prior the Cerberus experimental campaign. Each data point is an average of 4 measurements. The time axis is set such that $t = 0$ corresponds to the location of the main pulse, and pre- or post-pulse features are annotated. The noise floor of this measurement was around 10^{-7} below the main pulse intensity, which means that although the ASE pedestal cannot be measured, the problematic pre-pulse features corresponding to previous contrast characterizations can be resolved. Sharp steps in the pulse profile are attributed to filter calibration errors when changing between different intensity ranges.

the nonlinear energy scaling of the second harmonic generation and sum-frequency generation processes that respectively produce the probe pulse and 3^{rd} harmonic signal.

The Vulcan front-end has similar contrast properties to Cerberus (which is consistent with their similar architectures incorporating ps and ns time domain OPAs), with contrast ratios of $\sim 10^6$ on timescales of ≈ 200 ps and $\sim 10^5$ approximately 50 ps prior to the main pulse. These values are taken from measurements by N.H. Stuart four months prior to the Vulcan campaign, presented in [44]. Hence, for both the Cerberus and Vulcan PW experimental campaigns, we would not expect to see any drastic enhancement in either ion energies or EMP without the use of plasma-mirrors based on the contrast measurement. However, the marginal increases in peak EMP electric fields with decreasing target thickness at Vulcan could indicate the presence of some minor enhancement in electron heating according to the conditions described by Yin and Albright [60].

During the Cerberus experiment, an EO-EMP diagnostic with both fiber input and output (Fig

6.12) was used. The probe setup was placed at a horizontal distance of 20 cm from TCC, in the two positions shown in Fig. 7.5, measuring in the radial direction and angled away from this for comparison. In each position, the distance between TCC and the center of the probe crystal was kept fixed. A higher CW probe laser power was used in this experiment compared to at Vulcan, such that the average zero-field signals on the photodiodes were $\sim 50\text{--}70$ mV, instead of ~ 20 mV. This, combined with very little ambient electromagnetic noise, helped to increase the sensitivity of the setup to electric fields. Despite this, no measurable EMP was detected from solid targets using Cerberus in either diagnostic orientation, even with tungsten wire targets - only optical scatter or self-emission was measured. The solid targets were mounted on dielectric glass stalks, which could have slowed down the discharge through the target holder. This however was not the case for tungsten wire targets, which were mounted with a direct conduction path to earth.

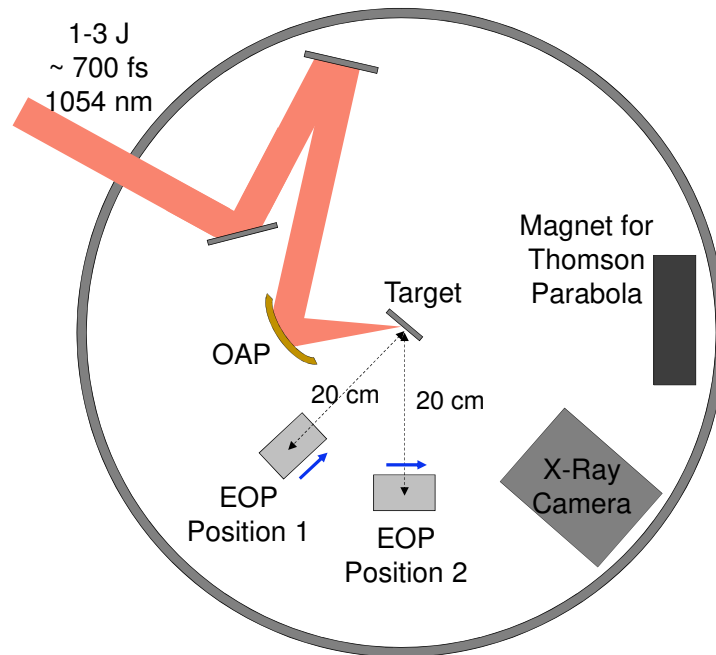


Figure 7.5: Schematic of the experimental setup used for electro-optic EMP measurement, using the Cerberus laser system at Imperial College. A gold-coated F/3 off-axis parabola (OAP) is used to focus onto target. The spherical interaction chamber also contained an X-ray camera and magnet for a Thomson parabola ion spectrometer. The two EO-EMP probe (EOP) positions are shown, with the blue arrows indicating the probing direction and longitudinal crystal axis.

As the probe is mobile, measurements were attempted along the interaction chamber's radial

axis as well as perpendicularly to this, and also at 180° to determine if peaks were self-emission or real fields. This means that the radiated electric field strengths were less than ≈ 500 V/m, which is still a relatively large value. As an additional check, the Faraday cage containing the oscilloscope was left open for some shots to look for ambient electrical noise; none was detected on the oscilloscope. This suggests that there is very little EMP emission at a level measurable by the EO-diagnostic from the relatively low drive-energy (1-3 J) laser-matter interactions achievable with Cerberus during this experiment.

The E-field traces for a variety of targets were all very similar; only self-emission/optical scatter peaks were observed, which were similar sizes in all cases. This is consistent with the source of the peaks being stray light, as attempts were made to keep the drive laser pulse energy fixed. For this experiment, the electric field detection threshold of the EO-EMP diagnostic was in the range of ~ 400 - 700 V/m, depending on the base signal level on a particular shot. This threshold is approximately half that of the Vulcan experiment, due to the mean zero-field signal on the photodiodes being ~ 50 mV instead of ~ 20 mV, and the lower ambient noise level. Hence, all the electro-optic data from Cerberus can tell us is that the electric field component of the EMP from our levitated target or solid target shots is less than ~ 500 V/m, despite the probe being placed ~ 20 cm from the radiating target plasma, compared to 1.25 m while at Vulcan PW. This demonstrates clearly one of the limitations of EO diagnostics compared to conductive probes, which typically are more sensitive, though much more prone to noise pickup, both directly, and via their conductive cables.

7.1.2 Optically levitated micro-targets

According to the Poyé model, the return current drawn through the target mounting assembly is a key contributor to EMP generated in a laser-plasma interaction. Hence, a target geometry which is completely electrically isolated from the interaction chamber and earth should remove the antenna-like RF radiation component arising from the target holder, and consequently reduce the EMP. Optically levitating targets is a practical method for achieving these conditions, in our case using the in-vacuo levitation trap developed by C.J. Price et al [92]. Further devel-

opment of the optical trapping technology by S. Giltrap et al. enabled hollow spherical shell targets to be trapped, alongside silicone oil droplets and other solid transparent targets; these were shot during the Cerberus campaign.

The EO diagnostic was unable to detect any EMP, above the minimum resolvable field-strength limit set by experimental noise, from interactions with levitated micro-targets using either Cerberus (oil droplets, glass spheres and few-nm silver-coated glass shells), or more significantly, Vulcan PW (Fig. 7.6), where the drive pulses contained orders of magnitude greater energies, and accelerated protons of energies >30 MeV were observed from droplets with no accompanying EMP; the ion acceleration results will be discussed separately in an upcoming publication led by S. Giltrap. Hence, any generated EMP fields were below the experimental electrical noise level, meaning field strengths were less than ≈ 1 kV/m and ≈ 500 V/m within the interaction chamber during the Vulcan and Cerberus campaigns respectively, and are thus not a concern for electrical interference with laboratory equipment.

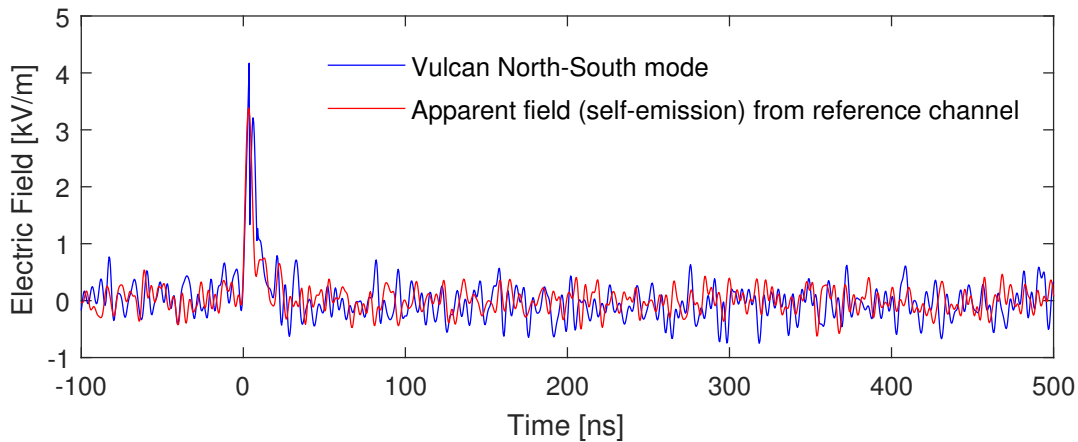


Figure 7.6: EMP trace measured from shooting an optically-levitated $10\ \mu\text{m}$ silicone oil droplet with a peak intensity of $1.6 \times 10^{18}\ \text{W cm}^{-2}$ (a defocused spot of $50\ \mu\text{m}$ was used to ensure a good hit). No EMP is measurable above the experimental noise level, despite observation of > 30 MeV protons, with only optical self-emission peaks visible on the processed EMP trace, as can be inferred from the reference channel (no laser).

In Fig. 7.6, the reference channel allows us to infer that the detected peak is due to optical self-emission from the target plasma being coupled to the fiber readout system. The reference channel consists of an optical fiber-launching setup immediately adjacent to the probe channel and an identical photodiode within the Faraday cage, such that scattered light from within

the Vulcan interaction chamber is coupled in to a similar degree, and that contribution from ambient electrical noise from the EMP detected by the EO-probes can be separated from the measurement.

Without EMP fields above the detection threshold from the laser-droplet interaction, it is not possible to comment on how isolated spherical geometry affects the frequency spectrum of any generated EMP. However, as there is no return current-driven component to the EMP, the source of any RF fields must be the interaction chamber cavity modes. Hence, if a sensitive enough diagnostic could be built, we predict a relative reduction in the frequency content of the droplet EMP spectrum which is distinct from the chamber harmonic modes.

7.2 Spectral analysis of EMP

Using the spectral analysis procedures described in Chapter 6, a comparison can be made between the shots taken at Vulcan PW. The resonant cavity modes predicted by Mead et al. [257] to be present in the Vulcan PW interaction chamber (without in-chamber metallic hardware) and lying within the measurement bandwidth of the EO-diagnostic are shown for reference in Table 7.2. The field strengths detected along the East/West measurement axis of the Vulcan interaction chamber are generally $\approx 2\times$ lower than those measured along the North/South direction. Despite measures taken during the experiment to shield the photodetectors and oscilloscope from ambient electromagnetic noise, inevitably this was not 100% effective. Hence, the spectral analysis presented in this section primarily considers spectra measured along the N/S axis, where signal-to-noise ratios are higher in all cases.

Mode and E-field Direction	Frequency [MHz]
E/W fundamental	76
N/S fundamental	101
Vertical fundamental	82
E/W 2 nd harmonic	152
N/S 2 nd harmonic	202

Table 7.2: Table showing the predicted cavity resonances for the interaction chamber at Vulcan Petawatt. The information contained in this table was taken from [257].

The typical forms of the acquired spectra can be seen in samples from shots #18 and #29 are shown in Fig. 7.7, with the ambient electrical noise background spectrum shown in red in each case. The spectral noise levels were determined by identically processing the measured voltage data from the identical noise-reference photodiode, which was connected to the same oscilloscope. The reference data is essential for determining which spectral components are actually measured by the probe, and which are a consequence of imperfect shielding by the Faraday Cage. From this, it is clear that the measured peaks are detected by the probe, with the exception of the predicted vertical fundamental mode at 82 MHz, which is observed with approximately equal spectral powers in data from both the EO probes measuring in the horizontal plane and noise-reference channels. This can therefore be discounted from the probe data as a noise contribution, and is consistent with the expectation that the probing setup should be insensitive to E-fields in the vertical direction.

In Fig. 7.8, the spectra from all shots are overlaid onto each other to allow a rough comparison between shots. From this, it is apparent that all the spectra have a similar form, with the relative spectral power varying somewhat between shots.

7.2.1 Reproducibility of spectral features

The reproducibility of the Vulcan EMP measurements shot-to-shot is important to quantify when deploying novel diagnostics. While there are indeed some differences in the spectra, numerous common spectral features, summarized in Table 7.3, are observable on all shots, irrespective of target or drive laser pulse parameters. Spectral components up to 200 MHz are shown, as above this frequency the signal-to-noise ratios were too small for meaningful conclusions to be made. Certain components, such as the chamber fundamental modes, are expected to be measured on every shot as the bulk in-chamber architecture (optical tables etc) remains fixed. As previously discussed, other variations in the EMP's spectral content can be attributed to the movement of conductive material such as optical mounts in the chamber between shots, which was not specifically controlled during the experiment, and to other frequencies emitted as a result of the discharge current drawn through the target assembly. Within the diagnostic's

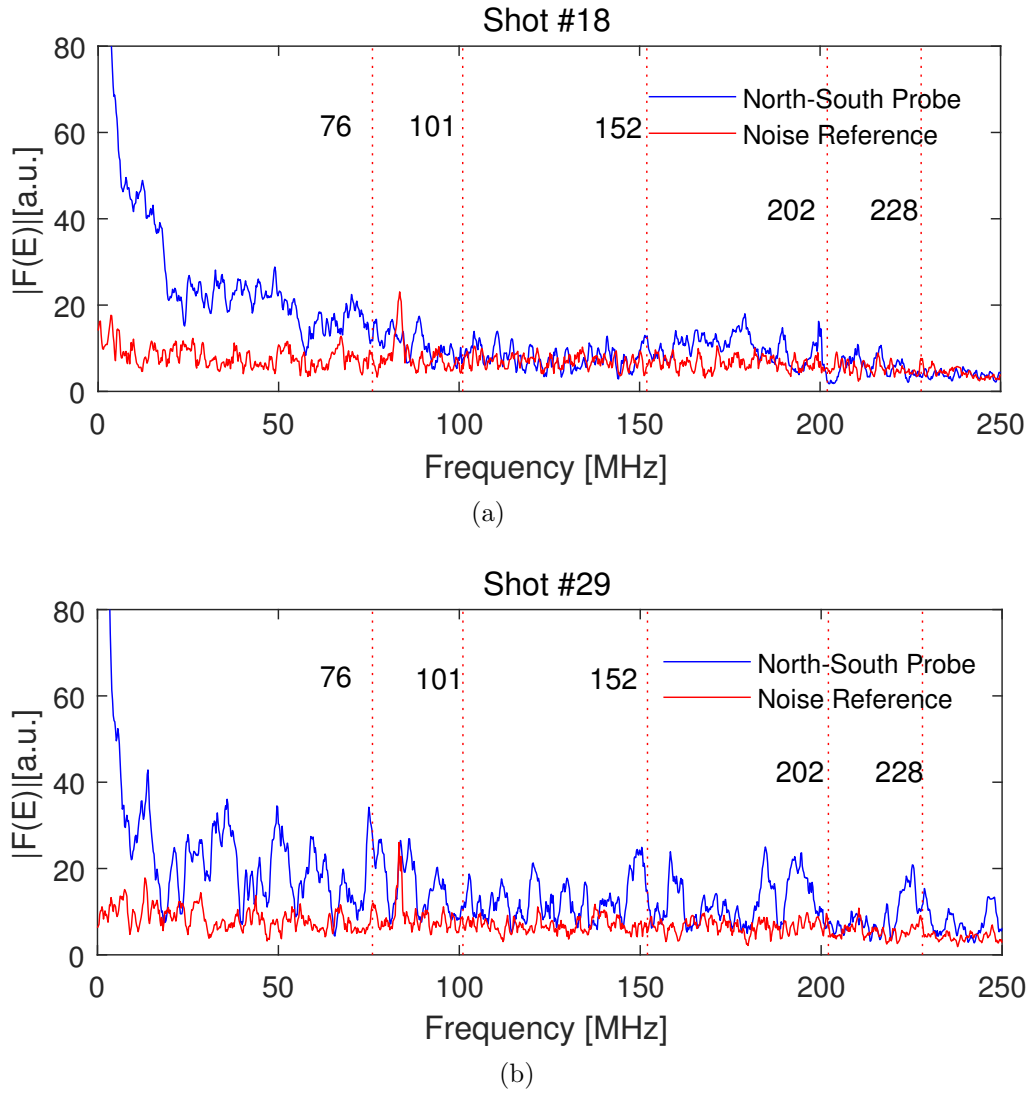


Figure 7.7: EMP spectra for sample shots #18 and #29, with background noise data shown for comparison. From both spectra, it is apparent that any detection of the 83 MHz cavity mode (vertical) is a result of ambient EMP noise coupling to the detection apparatus.

bandwidth range (0-250 MHz), the spectral resolution is limited to ~ 0.21 MHz, in this case by the oscilloscope time-base settings and sampling frequency.

A higher degree of reproducibility between shots taken on the same day of the experiment can be seen in the spectra from shots #18, #19 and #20 of the Vulcan campaign. The other 3 shots, (#14, #25 and #29) all took place on different days during the experiment, and exhibit less close agreement between themselves and the shot data acquired on the same day. The reproducibility is quantified by considering the standard deviation (SD) in the mean center-frequency, $\sigma/\sqrt{n}$, of features measured for a given identified frequency value (Column 1 of Table 7.3), where we have divided by $\sqrt{n}$ to account for differing sample sizes. The SD for the set of

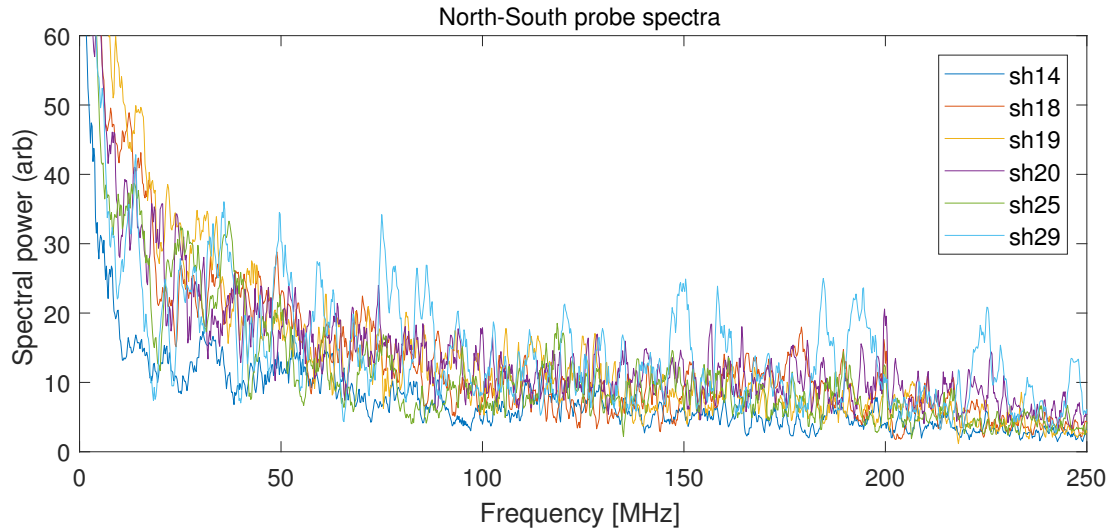


Figure 7.8: All EMP spectra measured along the Vulcan North-South probing axis. Although the spectral power varies somewhat between shots, the spectra are generally similar in form.

shots restricted to the single day S_{rs} , was then subtracted from the SD of all the shots, S_{all} , and the differences $S_{all} - S_{rs}$ plotted in Fig. 7.9. A positive difference value indicates that the SD in the mean peak position for an identified spectral feature is smaller for the restricted dataset; a majority (8 of the 12) features, exhibit a value ≥ 0 , suggesting a higher degree of spectral reproducibility between shots taken on the same day.

This chronological link, with no clear trend linked to target or shot parameters, suggests that minor spectrum modifications are more likely linked to movement of conductive objects within the interaction chamber between shots on different days, which can affect the frequency modes supported within the chamber [252]. Although it was not monitored and thus cannot be properly confirmed, there is a greater likelihood that the positions of metal objects within the chamber were more similar for shots taken on the same day. While the dataset, and hence sample size, is small such that is difficult to draw reliable conclusions, this evidence suggests the internal chamber architecture and contents are the dominating factors in the spectral make-up of the EMP, rather than the thickness of the plastic foil targets used.

This does not imply that target geometry is unimportant, as the plastic foils mainly differed in thickness, and no EMP was observed above the instrument noise floor with levitated targets. Furthermore, the target holder assembly was identical for these shots; as this forms part of the radiating element according to the Poyé model, it follows that the frequency content of

Spectral Feature [MHz]	Observed frequency [MHz] at shot #					
	#14	#18	#19	#20	#25	#29
30	30.2	32.5	31.0	30.8	29.4	32.3
40	39.8	39.4	39.0	41.0	40.0	41.0
50	50.0	49.0	50.0	51.9	49.8	49.6
60	60.0	61.3	61.3	61.3	60.6	59.6
71	72.0	70.2	71.6	70.8	71.3	69.6
74	74.0	74.6	74.6	74.2	74.6	75.0
76	75.4	76.7	78.0	76.7	75.8	75.0
94	94.2	95.8	93.3	94.0	95.0	94.0
102	102.9	100.4	102.3	102.5	104.6	102.7
120	120.2	120.2	121.9	119.4	120.2	120.2
152	151.5	152.1	152.7	152.3	152.3	150.4
200	199.6	199.6	199.6	199.8	199.8	199.6

Table 7.3: Sample of common spectral features along the Vulcan North-South interaction chamber axis identified between shots (within a small $\approx \pm 1$ MHz tolerance). The minimum uncertainty is set by the diagnostic spectral resolution of 0.21 MHz. A generally closer agreement was observed for the 3 shots taken on the same day, shown in italics.

radiated EMP will remain consistent. This lack of spectral dependence on the target thicknesses is in contrast to the peak electric fields measured, which depend on the ejection of hot electrons and hence the plasma electron temperatures; these have been demonstrated to vary with target thickness in the context of the BOA and relativistic transparency ion-acceleration regimes [60, 126, 154]. An interesting feature of the electro-optic EMP data is that some of the fundamental chamber resonant modes were observed in the measurement direction perpendicular to their expected direction; for example, the electric field directions of 76 and 152 MHz modes were predicted to be along the E/W chamber direction by Mead et al. [257], rather than N/S. In an electromagnetic parallelepiped-shaped hollow cavity, TE and TM cavity modes are allowed. Some of these modes are degenerate, so share resonance frequencies. Moreover, the presence of objects in the chamber can locally change the modal field distribution without changing the resonance frequency remarkably [252], as previously discussed in Section 6.4.

7.2.2 Time-frequency analysis

As introduced in the previous chapter, short-time Fourier analysis techniques can be used to infer information about the dynamical evolution of spectral modes emitted following the

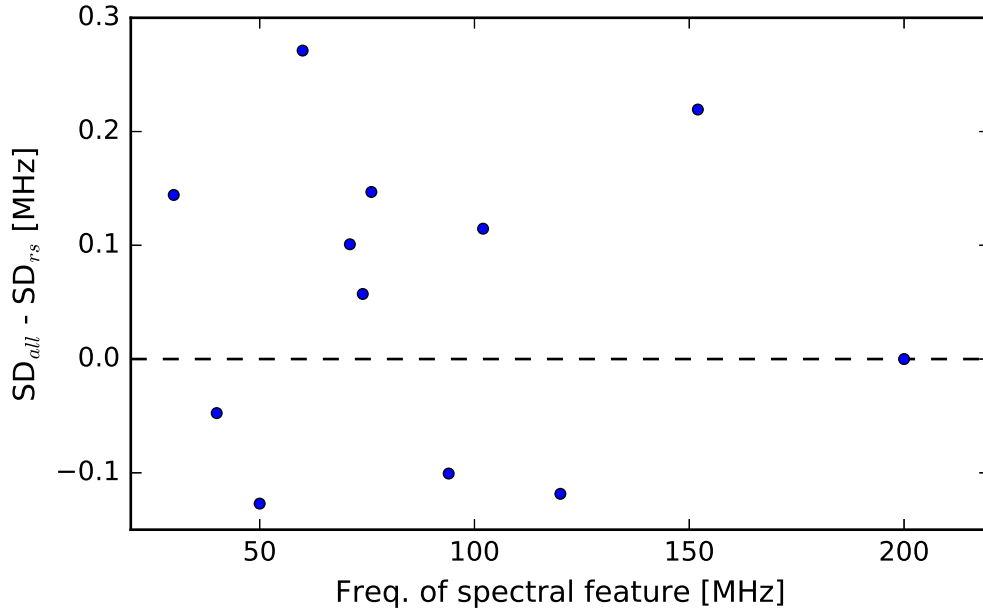


Figure 7.9: Plot of the difference between the standard deviations (SD) S_{all} of the mean position of spectral features between all shots and the SD S_{rs} calculated from a restricted population of shots taken on the same day (#18, #19, #20). The SD of the mean $\sigma/\sqrt{n}$, where n is the number of samples, has been used to account for the different sample sizes.

laser-plasma interaction. A spectrogram for electric fields measured along the N/S direction is shown in Fig. 7.10. From this, a late-time, secondary detection of EMP fields with frequencies matching the interaction chamber cavity modes was observed.

A likely explanation for this behaviour is that the two previously discussed mechanisms contributing to EMP generation are temporally offset; an electron bunch ejected from the target plasma will have a finite transit time to the interaction chamber walls, whereas a positively charged target can immediately draw a neutralization current through its supporting structure.

This explanation can be tested against time-of-flight (TOF) measurements, sensitive to charged particles and X-rays. Although we were not able to obtain TOF diagnostic data for the shots with EO-EMP data, the TOF data from one of the other shots in the series (#21) is still useful for providing some insight into the delayed activation of the interaction chamber cavity modes. A synthetic monocrystalline CVD diamond detector was used as a TOF diagnostic. This was fabricated by Tor Vergata University laboratories with superficial interdigitalized electrodes [303,304] and operated by colleagues from ENEA-Frascati (F. Consoli, R. De Angelis, F. Ingenito). It was placed in the Vulcan chamber's western wall, with a direct view of the

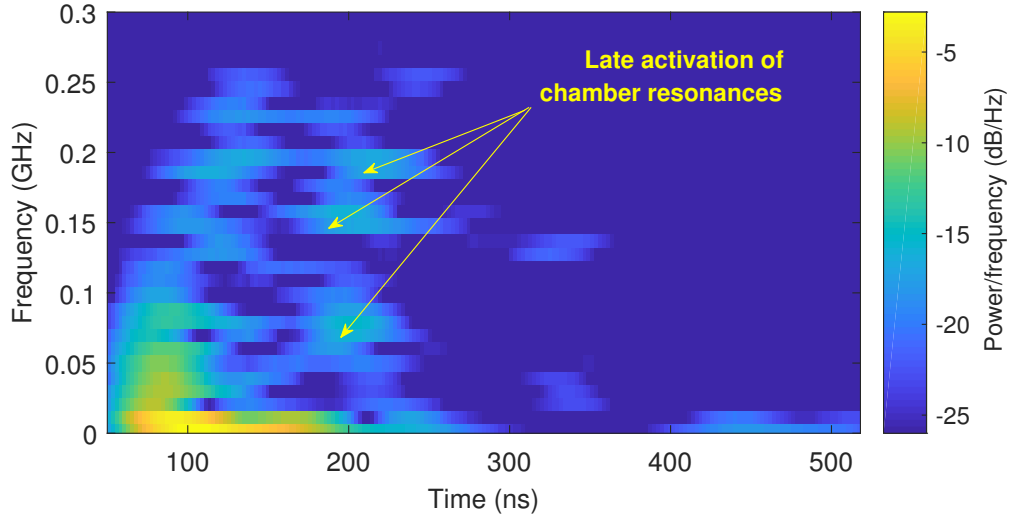


Figure 7.10: Spectrogram for EMP fields measured along the Vulcan N/S direction taken on shot #29 (see Table 7.1), which had the highest energy incident onto target. There appears to be some late-time activation of the interaction chamber cavity modes, likely due to ejected electrons striking the chamber walls after a finite transit time, whereas EMP radiated due to the discharge current through the target holder can occur as soon as the target becomes positively charged.

target and 100 V biasing, at distance of ≈ 1 m from TCC. A TOF trace for shot #21 is shown in Fig. 7.11; from this, we can observe the separate arrival of X-rays (at 390 ns) as well as “fast” and “slow” particles (above 490 ns) at the diamond detector. As the ambient EMP noise coupling proved problematic to shield, a 60 m length of RG58 cable was introduced between the detector and the oscilloscope, adding a delay of ≈ 311 ns and temporally separating the EMP background from the ‘true’ diamond signal.

The X-ray photo-peak provides a reference for T_0 , the time of the initial laser strike, relative to other features, as the time taken for photons emitted from TCC can be calculated to be ≈ 3.3 ns. The arrival of fast particles (few MeV fast ions, few keV electrons) peaked ≈ 100 ns after the photo-peak, followed by a broad peak representing slower charged particles. These timescales are approximately consistent with the N/S mode EMP spectrogram for shot #29 (Fig. 7.10), where secondary activation of the fundamental chamber modes can be seen ≈ 110 ns after the initial EMP emission. Although we expect some shot-to-shot variation in the speeds to which ejected fast particles are accelerated, there should be some finite time between fast electrons striking the chamber walls and the excitation of RF modes. This provides some evidence for

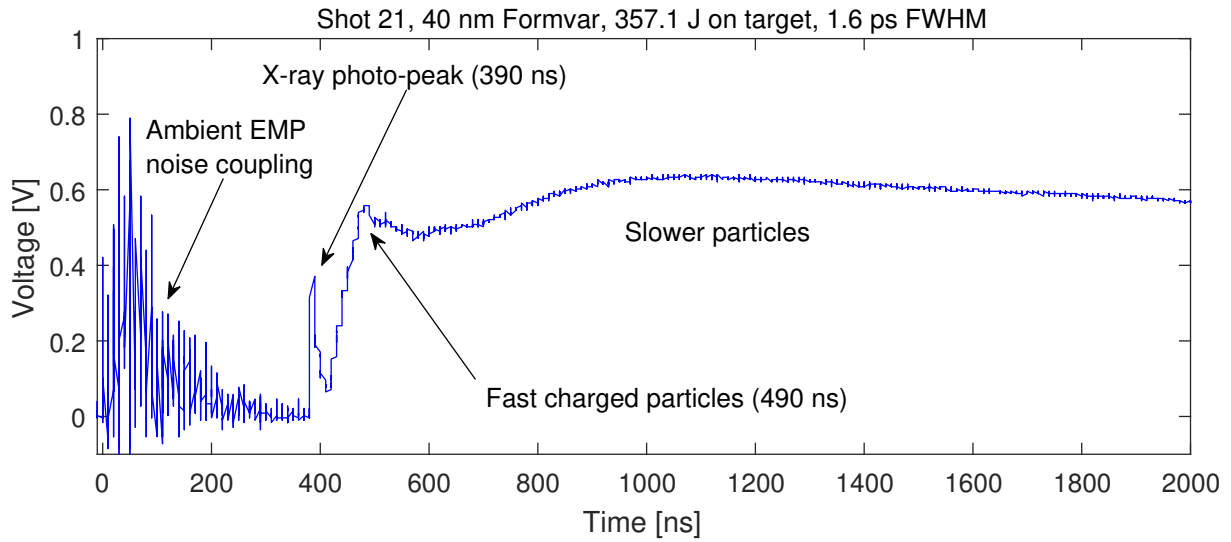


Figure 7.11: A time-of-flight trace recorded using a biased CVD diamond detector. A 40 nm-thick Formvar target was used here, irradiated by a 357 J, 1.6 ps pulse, with a peak intensity on target of $5.6 \times 10^{20} \text{ W cm}^{-2}$. A 60 m length of RG58 cable was used to temporally separate the diamond signal from the ambient EMP noise coupling to the oscilloscope. The temporal resolution of this measurement was 1 ns. The time axis has been shifted such that the initial laser strike $T_0 = 0$. The key contributions to the measured signal are labelled on the plot.

the cavity modes being excited later in time by ejected particles striking the chamber walls, while the radiated fields emitted from the target assembly due to the neutralization current occur earlier in time, closer to T_0 .

If true, this implies that the RF-emission from the target assembly is the larger contribution to EMP in experiments of this type, which could explain the lack of measurable EMP from shots with optically levitated micro-droplets, which lack a physical discharge current path but still result in ejection of charged particles.

7.3 Conclusions

Although the electro-optic EMP dataset obtained from Vulcan PW is limited to only 6 shots, there appear to be certain trends identifiable from the time- and frequency-domain data. Firstly, the peak EMP E-fields from ultra-thin plastic foil targets appear to correlate negatively with the target thickness; this cannot be explained by the laser energies on target being higher for certain shots, as there is no clear correlation between the input energy and the measured peak

fields. Hence, at least within the restricted ranges of energy and target thicknesses considered, we suggest that EMP produced by a laser-plasma interaction scales with the target geometry, potentially due to enhanced electron heating when the thickness of targets is comparable to the electron skin depth. The lack of measurable EMP from electrically-isolated, optically-levitated, targets also indicates that removing the discharge current component of EMP contributes substantially to mitigating the EMP problem.

In contrast to the peak intra-crystal field values, the obtained EMP spectra do not appear to show any strong dependence on target thickness, with spectral features largely being reproduced in approximately the same positions on every shot. There appear to be minor shot-to-shot variations in the position of several identified spectral features, quantified by the standard deviations of the mean center frequency, which appear to be reduced for shots taken on the same day. This suggests that minor modifications to spectral features are more likely to be driven by movement of conductive objects within the chamber, as opposed to variation in target thicknesses. Time-frequency analysis combined with time-of-flight measurements, which show a late secondary appearance of frequencies matching the resonant cavity modes ≈ 10 ns after the arrival of fast electrons at the chamber walls, also tentatively suggest that the cavity modes activate later in time than the initial EMP emission (which may include frequency components matching the cavity modes). We thus consider the early-time EMP contribution to be RF-emission from the target assembly, as in the Poyé discharge current model.

Additional shots at petawatt facilities are required to confirm these observations, however they provide a starting point which can be used to tailor experiments for further studies into laser-driven EMP generation, in terms of experimental parameters such as target type and target holder design, along with other factors including the placement or movement of conductive objects in the target chamber, particularly as ultra-high energy petawatt systems such as PETAL and Apollon 10 PW become active.

Chapter 8

Conclusion

8.1 Summary of Thesis Achievements

A self-healing optical limiter device using optically nonlinear liquids has been developed, capable of functioning across a broad range of pulse durations and intensities, with performance orders of magnitude better than the previous state of the art and with functionality at both nanosecond and sub-picosecond pulse durations. Multiple liquids were investigated, with concentrated aqueous ZnCl_2 solutions found to be the best choice for the liquid medium among those considered. A design for a contrast diagnostic incorporating this optical limiter for protecting the sensitive optical detectors has been proposed, enabling single-shot contrast measurements over long timescales with low-risk of damage to the detectors, allowing dynamic range to be increased.

A very simply implemented supercontinuum light source was constructed and characterized in terms of energy conversion, its spectral profile and spectral stability, with an article on this topic published in OSA *Applied Optics* and used to support applications including broadband interferometry [231] and absolute calibration of optical streak cameras. Quantifying the spectral stability was necessary to complement work concerning the dynamic calibration of optical streak cameras in fast-sweep mode; the standard deviation in measured spectral intensities for each wavelength value across the full spectrum was found to have a mean value of $\approx 6\text{-}7\%$. The source

stability can be increased by increasing the input driver pulse energy; this was attributed to driving a nonlinear process further above threshold into a more stable regime, hence small fluctuations in input energy have a smaller effect on the output white-light stability compared to pulses with intensities close to the spectral broadening threshold. The energy conversion efficiency from the driver pulses to the supercontinuum was also found to increase with input energy, with broadband pulse energies of ~ 40 μJ observed after removal of the driver pulse component (initially 2.42 mJ) via an infrared mirror.

An electro-optic diagnostic capable of measuring strong electromagnetic pulses from petawatt-regime laser-matter interactions was developed, yielding the first measurements of their kind, with potentially a first observation of an increase in peak EMP fields with ultra-thin targets. This work forms the basis of publications by the author in *Scientific Reports* and *EPJ: Web of Conferences*, as well as a co-authored article submitted to *Nature Communications*. Absolute measurements with very low inherent instrument noise are obtained from EO systems, as the polarization rotation involved in the Pockels effect scales linearly with the intra-crystal electric field; this removes the need for numerical integration to extract the electric fields from the acquired voltage traces, a source of significant signal distortion in D-Dot probes and similar schemes. This diagnostic was then further optimized for experiments at Imperial College on the Cerberus laser system, and was deployed to characterize the EMP from a range of different target types and geometries. Despite increasing the sensitivity of the electro-optic diagnostic by a factor of ≈ 2 , the EMP fields from Cerberus were still too small to be detected, due to its low sensitivity relative to conventional conductive probes.

A substantial upgrade on the Cerberus long-pulse chain and short-pulse beamlines has been designed, which would convert it to an all-OPCPA system, with benefits in terms of contrast as there is no possibility for amplified spontaneous emission (ASE) after the master oscillator due to a lack of gain-storage amplifiers. Indeed, the only source of longer-timescale contrast degradation would be parametric fluorescence within the OPAs [62]. However, as OPAs do not store energy within the crystals, any parametric fluorescence would be limited to the pump pulse duration, ≈ 1.5 ns at 527 nm following the Cerberus pump-chain upgrade, compared to possible ASE durations in excess of a microsecond. To properly make use of the upgraded max-

imum laser energies, a vacuum compressor system was also designed; this removes the energy constraint imposed by B-integral acquired from propagation through air and an interaction chamber entrance window, such that the maximum usable peak power is ~ 40 TW. This is limited by the laser-induced damage threshold of the dielectric diffraction gratings in the H147 high-intensity target area.

8.2 Applications

The optical limiter system described in Chapter 4 has potential applications beyond protecting photodiodes in a long-timescale laser pulse contrast diagnostic. As high-energy pulsed lasers become increasingly common in military contexts, there will likely be continued interest in protecting sensor systems against laser-based attacks; incorporating an effective self-healing OL into military imaging systems provides a means to protect against both intense long (nanosecond) and short (sub-picosecond) pulses. For this reason, we are pursuing intellectual property protection on the OL system, which has delayed submission of the journal article prepared on this topic.

Supercontinuum sources, such as the simply-implemented and stable source described in Chapter 5, have numerous potential applications (particularly if the broadband pulses are compressed), including white-light interferometry, reflectometry, biomedical applications such as Optical Coherence Tomography, and ultrafast spectroscopy, as well as high precision absolute optical streak camera calibration as described by Patankar et al. [232].

As electro-optic EMP diagnostics have successfully demonstrated their capabilities in providing low-noise, absolute field measurements in both the nanosecond and picosecond (high intensity petawatt) drive laser pulse duration regimes [244, 245], they are likely to increasingly feature within the diagnostic suites of those interested in EMP characterization. EO-EMP diagnostics are not limited to use in laser-facilities; any application requiring measurement of strong electric fields in a high-noise environment would be well-served by such systems due to their inherent noise-resistance and potential for zero conductive components in the probe setup.

8.3 Future Work

While the optical limiter system has been demonstrated to perform favorably against comparable setups in the literature, there remains much optimization that could be done before potential release as a marketable product; parameters such as the liquid cell length, the focusing geometry into the liquid and the structure of the liquid cell could all be considered in order to improve the system. Understanding the behavior of the OL at ultrashort pulse durations (<100 fs) is also important for a system capable of effectively attenuating laser pulses at a variety of pulse durations. There is also currently no information on how the OL attenuation scales at higher pulse energies, though the maximum usable incident pulse energy will be limited by the damage thresholds (at a given pulse duration) of the the solid state optical elements forming the system.

Similarly, the optimal focal geometry for maximizing the energies of supercontinuum pulses generated in K_2ZnCl_4 double-salt solutions has not been investigated. Increasing the focal lengths of lenses could help to increase the interaction length over which self-phase modulation (SPM) and other processes such as optical wave-breaking take place, thus improving the conversion efficiency of the spectral broadening. Although the conversion efficiency and spectral stability have been shown to increase with the input pulse energy, it is unclear where the upper limit to this effect occurs due to the onset of another physical process, such as increased laser absorption into the plasma formed in the focal region with increasing free electron densities. Hence, studying the supercontinuum behavior with drive energies in the $\approx 5\text{-}15$ mJ range would be worthwhile. There will be an upper limit on the usable energies with the liquid-cell based setup, in practice set by the damage threshold of optical elements, which will occur at energies far below the B-integral or compressor grating damage limits of the laser system or the energies required to reach critical plasma electron densities. Measurement of the spectral phase properties of the output supercontinuum would also be interesting as an assessment of the compressibility of the broadband pulses. However, this is difficult to do in practice, as a custom diagnostic for spectral phase measurement over such a broad spectral range (>600 nm) and relatively long pulse duration (~ 5 ps) would need to be built, such as an X-FROG setup [73, 305, 306].

The electric fields generated by Cerberus with foil and levitated targets were insufficiently large to exceed the electro-optic EMP diagnostic's detection threshold, hence there remain experiments to be carried out investigating the effects of target geometry on laser-driven EMP generation. Completely different targets, such as monomer or cluster gas jets, should also be investigated due to their growing presence in laser-plasma research. The diagnostic architectures developed during this PhD project were designed for use at Vulcan Petawatt (a relatively high-energy facility compared to the emergent generation of Ti:Sapphire PW-lasers), hence a comparable facility would be recommended for carrying out follow-up experiments, allowing direct comparisons to be made.

There is also potential for using the fields produced by laser-plasma interactions as a means of studying pulsed high-field physics. For example, an electrically isolated conductive plate placed in the path of an electron bunch ejected from the target can be rapidly charged, allowing a region of rapidly rising strong-electric field to be produced between it and a nearby second appropriately placed earthed plate; this topic is discussed in an upcoming paper by F. Consoli and co-authored by the author of this thesis. If a relatively uniform electric field is required in this strong-field region, a minimally perturbative field diagnostic, such as an electro-optic system only containing dielectric components, would be highly desirable.

In general, the motivation for studying laser-generated EMP is growing, driven by the increasing number of high energy, ultra-high intensity laser facilities being constructed worldwide, such as Apollon 10 PW, the ELI 1 & 10 PW laser systems, as well as LMJ and PETAL. As the energies and peak-powers provided by these lasers increase, the EMP problem will become more pronounced. High repetition rate petawatt systems currently in development, such as ELI-beamlines L3 (Prague, Czech Republic) or CLPU Vega-PW (Salamanca, Spain) will provide new capabilities to study EMP, particularly for radiation hardening applications; currently the effects of high-repetition exposure of electronic systems to EMP are unknown. In such an experimental environment, low-noise EMP diagnostic capabilities providing absolute measurements will be essential for high-quality EMP characterization.

There is considerable work to be done on the implementation of the laser upgrade designs

outlined in Chapter 2, including transferring the prototyped Martinez-style stretcher into the long-pulse beamline and incorporating a Spatial Light Modulator, adding in the additional flashlamp-pumped Nd:YLF amplifiers, procurement and commissioning of the vacuum compressor system and commissioning of the new OPA stages, with the potential of operating the Cerberus short-pulse beamline as an all-OPCPA system for optimal contrast enhancement.

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