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Laser Produced Plasmas as a Source of Ions, Protons and X-rays

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The thesis is submitted to University College Dublin in fulfilment of the requirements for the degree of Doctor of Philosophy in Physics

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May/2014
To my parents
Abstract

The work presented in this thesis is primarily focused on the use of a laser produced plasma as a source of protons, ions and X-rays. It explores high impact applications of both high power ultrafast lasers and nanosecond lasers.

Section 1 gives a general introduction to the physics governing the experiments and the lasers in the following sections. The physics of plasmas is described in Section 1.1. The physics of ultrafast lasers is described in detail in Section 1.2 from the theory of mode locking of oscillators to the compression of high power ultrafast systems. The physics and mechanisms of X-ray lasers are then explained in Section 1.3. Both the collisional excitation mechanism and the Linford gain equation are described along with a summery of the challenges in pumping X-ray lasers. Next in Section 1.4 laser accelerated protons are described in detail. Acceleration mechanisms are described and the interaction mechanism with insulators is introduced. Finally, a number of tools for photoabsorption studies are described in Section 1.5. In this Section the dual laser plasma technique for obtaining photoabsorption spectra is described along with the atomic structure codes (the Cowan and RTDLDA codes) used to simulate the spectra.

Section 2 describes all the laser systems used in this thesis. The various components of the Quantronix ultrafast laser system from UCD including the oscillator, the Odin II amplifier and the Thor amplifier are described in Section 2.2, Section 2.3 and Section 2.4. The operation of the Surelite III and the Spectron SL805 Q-switched nanosecond lasers are described in Section 2.5 and Section 2.6. Finally, the Taranis multi-terawatt system and its different components are described in Section 2.7.

Section 3 is covers two different experiments involving the generation of X-rays from a laser produced plasma. The first technique described in Section 3.3 is the generation of coherent X-rays by pumping a preformed laser plasma with the Taranis laser system. A Ni-like Mo laser was successfully pumped yielding energies of $2 \times 10^{-7}$ J with a gain length product of 3000.

The second technique described in Section 3.5, involved inner shell X-ray emission from an indium target irradiated by the Quantronix laser system. The indium K series emission was observed using a lithium-drifted silicon detector. Seven counts of $K_\alpha$ emission was detected under the following conditions (12000 shots through a 0.5 mm pin hole filtered with a 60$\mu$m Al foil and a laser energy of 30 mJ). Due to the laser reliability this experiment is only a preliminary one which is intended to be continued at a later stage.

Section 4 describes a laser accelerated proton experiment conducted in the Center for Plasma Research in Queens University Belfast, using the Taranis laser system. In this experiment 13 MeV protons were accelerated from
10 µm gold foil targets into a sample of BK-7 glass. The interaction of the protons with the glass were observed by taking spatially resolved images of the transient opacity induced by the protons interacting with the BK-7 Glass and an optical probe beam. These spatially resolved images are presented in Section 4.3.

In Section 4.4.2 an optical streaking technique experiment is described to calculate the lifetime of the transient opacity induced by the proton interaction. This process showed that the opacity in the glass began to occur 62 ps after the TARANIS main pulse was fired and reaches 30% transmission 88 ps after the firing of the TARANIS main pulse. Due to the limited time window available, the exact lifetime of the opacity is not known, however glass is seen to de-excite after 138 ps and has returned to 50% transmission by 175 ps.

Section 5 describes the refurbishment of the 1-m normal incidence VUV spectrometer. It describes the replacement of a photographic plate based detection system with a linear CCD array. The CCD array can detect the VUV radiation through a sodium salicylate phosphor coating which emits at 410 nm on interacting with VUV radiation. Different phosphors are compared in terms of sensitivity and ease of coating and the grounds for choosing sodium salicylate are explained. The adaptations to the spectrometer to use the linear CCD array are described and the details on calibrating the spectrometer are explained.

Finally, Section 6 describes a set of spectroscopic experiments which use the refurbished 1-m normal incidence spectrometer. First Section 6.2 describes a repeat of the photoabsorption of indium and indium plus which was conducted previously on the spectrometer. This was designed as a proof of principle of the working of the new phosphor based linear CCD array system.

Next, Section 6.3, describes the photoabsorption of thulium in the 23 eV to 40 eV region. The experimental spectrum is compared to the RTDLDA calculations and to the autoionized Cowan code calculations. The simulations successfully describe the observed absorption structure and \( 5p \rightarrow nd \) and \( 5p \rightarrow ms \) transitions are identified.

In Section 6.4 an indium laser produced plasma is successfully reheated using the Odin-II first amplifier of the Quantronix laser system. An experiment is described which shows emission from an indium plasma from transitions which are normally observed in absorption. This reheating was optimized with a specific set of target parameters (a delay of \( \Delta \tau = 500 \text{ ns} \) between the lasers, the Odin was focused into the center of the absorbing line plasma, which was set at a height of \( \Delta z = -0.2 \text{ mm} \) from the optical axis of the spectrometer).
# Contents

1 Introduction 15

1.1 Plasmas 15

1.1.1 Introduction to Plasmas 15
1.1.2 Equilibrium in Plasmas 16
1.1.3 Local Thermodynamic Equilibrium (LTE) 16
1.1.4 Coronal Equilibrium (CE) 17
1.1.5 Collisional Radiative Equilibrium 17
1.1.6 Atomic Processes in Plasmas 19

1.2 Ultrafast Lasers 20

1.2.1 Introduction 20
1.2.2 Ti: Sapphire as a Lasing Medium 21
1.2.3 Mode Locking 23
1.2.4 Mode Locking Techniques 24
1.2.5 Chirp Pulse Amplification (CPA) 28
1.2.6 Frequency Resolved Optical Grating (FROG) Technique 30

1.3 X-ray Lasers 35

1.3.1 Introduction 35
1.3.2 Collisional Electron Excitation 37

1.4 Proton Acceleration 39

1.4.1 Introduction 39
1.4.2 Target Normal Sheath Acceleration (TNSA) 40
1.4.3 Proton Interactions with Insulators 45

1.5 Photoabsorption of Thulium (Tm) 46

1.5.1 Dual Laser Plasma (DLP) Technique 46
1.5.2 Atomic Structure Codes 47

2 Lasers 55

2.1 Introduction 55

2.2 Ti:Lite Oscillator 56

2.2.1 Introduction 56
2.2.2 Colibri Pump Laser 56
2.2.3 Ti-Light Layout and Operation 56

2.3 Odin 2 Amplifier System 60

2.3.1 Introduction 60
2.3.2 Darwin Pump Laser 61
2.3.3 Odin-II Stretcher Layout and Operation 62
2.3.4 Odin-II Amplifier layout and operation 63
2.3.5 Darwin Pumping Line of the Odin-II Amplifier 66
2.3.6 Odin-II Compression System 67
3 Laser Generated X-ray Sources

3.1 Introduction ................................................. 88
3.2 Review of Target Geometries for X-ray Lasers .......... 89
  3.2.1 Exploding Targets ...................................... 92
  3.2.2 Prepulse Treatment ..................................... 92
  3.2.3 Transient Pumping Scheme .............................. 94
  3.2.4 Traveling Wave Scheme .................................. 94
  3.2.5 Grazing Incidence Pumped Configuration (GRIP) .... 95
3.3 Ni-Like Mo X-ray Laser ..................................... 95
  3.3.1 Introduction ............................................. 95
  3.3.2 Experimental Set Up ..................................... 96
  3.3.3 Energy Measurements using the Flat Field Spectrometer 97
3.4 Results and Conclusions ................................... 99
  3.4.1 Ni-Like Mo X-Ray Laser ................................. 99
  3.4.2 Conclusions .............................................. 103
3.5 Inner shell X-ray Emission of Indium .................. 104
  3.5.1 Introduction ............................................. 104
  3.5.2 Experimental Set Up ..................................... 105
  3.5.3 Lithium Drifted Silicon [Si(Li)] Detectors .......... 106
  3.5.4 Calibration and Optimization of the Si(Li) Detector 106
  3.5.5 Pile Up Phenomenon ..................................... 111
  3.5.6 Optimization of X-ray Counts ......................... 112
  3.5.7 Analysis of Inner Shell Emission Lines from In Ablation 113
4 Ultrafast evolution of Transient Opacity in Borosilicate Glass
Induced by Laser Driven Ions
4.1 Introduction .......................................................... 122
4.2 Experimental Set Up ............................................... 123
  4.2.1 Introduction ...................................................... 123
  4.2.2 Proton Beam Experimental Set Up ........................... 123
  4.2.3 Proton Energy Measurement ................................... 125
4.3 Spatially Resolved Images of the Transient Opacity Induced by the Protons Interacting with the BK-7 Glass .............. 126
  4.3.1 Introduction ...................................................... 126
  4.3.2 Demonstration and Filtering of Proton Beams ............... 126
  4.3.3 Depth Calibration of Proton Beams .......................... 131
  4.3.4 Proton Scans at Different Time Delays ..................... 138
4.4 Single Shot Direct Mapping of the Temporal Evolution of the Transient Opacity via the Optical Streaking Technique ........ 140
  4.4.1 Introduction ...................................................... 140
  4.4.2 Optical Streaking .............................................. 140
  4.4.3 Optical Streaked Images of Proton Beams .................. 144
4.5 Conclusions .......................................................... 149

5 Refurbishment of the 1-m Normal Incidence UV Spectrometer
5.1 Introduction .......................................................... 152
5.2 The 1-m Normal Incidence Spectrometer Detector ............... 153
  5.2.1 Phosphor Evaluation .......................................... 154
  5.2.2 Laser Generated Plasma Spectra ............................. 158
  5.2.3 Comparison of Phosphors ..................................... 160
  5.2.4 Sodium Salicylate Deposition onto the Array ................ 162
5.3 Focusing the Spectrometer ......................................... 162
5.4 The 1-m Normal Incidence Spectrometer Chamber ................ 163
5.5 Calibration of 1-m Normal Incidence Spectrometer .............. 167

6 Photoabsorption and fs experiments using the 1-m Normal Incidence Spectrometer
6.1 Introduction .......................................................... 182
6.2 Photoabsorption of Neutral In and In+ .......................... 182
  6.2.1 Introduction ...................................................... 182
  6.2.2 DLP Set Up ...................................................... 183
6.2.3 Photoabsorption of In and In$^+$ .......................... 188
6.3 Photoabsorption Studies of Thulium (Tm) Ion States .... 190
   6.3.1 Introduction .............................................. 190
   6.3.2 Previous Photoabsorption Studies of Tm ................. 190
   6.3.3 4d Photoabsorption in Thulium .......................... 191
   6.3.4 Results for 5p Photoabsorption .......................... 195
   6.3.5 Results for the Photoabsorption of Neutral Tm .......... 198
   6.3.6 Results for the Photoabsorption of Singly Ionized Thulium 212
   6.3.7 Results for the Photoabsorption of Doubly Ionized Thulium 220
   6.3.8 Conclusions .............................................. 229
6.4 fs Reheating of an In Laser Produced Plasma ................. 231
   6.4.1 Introduction .............................................. 231
   6.4.2 Experimental Set Up for the fs Reheating of an In Laser Produced Plasma .............................. 231
   6.4.3 Results for fs Reheating of an In Laser Produced Plasma 233
   6.4.4 Conclusions .............................................. 238
List of Figures

1.1 Absorption cross Section of $\text{Ti} : \text{Al}_2\text{O}_3$ as a function of wavelength [12] .................................................. 22
1.2 The emission of $\text{Ti} : \text{Al}_2\text{O}_3$ as a function of wavelength [12] .................................................. 23
1.3 Position in the Ti:Sapphire crystal of minimum spot size, minimum pulse width and maximum intensity [18] ......................... 27
1.4 Diminishing oscillatory behaviour of spectral band width and temporal pulse width to a steady state with multiple passes through the crystal [18] .................................................. 28
1.5 SHG autocorrelator geometry [27] ........................................... 30
1.6 Autocorrelation ambiguities [27] ........................................... 31
1.7 Schematic of an (SHG) FROG geometry [27] ............................ 32
1.8 Graphic illustrating 3 pulses with different linear chirps and their corresponding spectrograms underneath ......................... 33
1.9 Graphic representation of the FROG general projections algorithm [27]. ................................................................. 34
1.10 Schematic energy level diagram for relevant transitions of Ne-like, $\text{Se}^{+24}$ [30] .................................................. 37
1.11 Schematic energy level diagram for relevant transitions of Ni-like $\text{Eu}^{+35}$ [31] .................................................. 38
1.12 Depth dose comparison of different medical ionizing sources [38] ........................................................................ 40
1.13 Proton acceleration from an electrostatic sheath at the rear of a target [40]. .................................................. 43
1.14 Images of electron circulation inside both a thick and thin target [41] .................................................. 44
1.15 Plot of peak proton energy as a function of target thickness [42] ........................................................................ 45
1.16 Schematic of the DLP set up for Photoabsorption [47] .......... 47
2.1 Schematic of the Ti-Light oscillator [3] ........................................... 57
2.2 CW spectral output of the Ti-Light ........................................... 58
2.3 Output of the photodiodes while the Ti-Light is in ML operation ................................................................. 59
2.4 ML spectral output of the Ti-light ........................................... 60
2.5 The Darwin pump laser cavity [4] ........................................... 61
2.6 Odin II stretcher system [5] ........................................... 62
2.7 Visible beam pattern on the stretcher grating [5] ...................... 63
2.8 Odin-II amplifier system [5] ........................................... 64
2.9 Output of the PC ................................................................. 65
2.10 Schematic of the Odin-II compressor system .......................... 67
2.11 The Thor Relay Imaging chamber layout [6] ...................... 68
2.12 Schematic of the Thor amplification system [6] ...................... 69
2.13 Spectral output of the Thor amplifier system .......................... 71
2.14 Measured FROG trace of the Thor ................................. 72
2.15 Pulse duration and beam profile of the Surelite III pump laser 73
2.16 The Surelite III cavity with it’s shutter closed [8] ............... 74
2.17 The Surelite III cavity with the shutter open [8] .................. 75
2.18 Spectron SL805 beam structure and pulse duration ............ 77
2.19 Schematic of the Spectron laser ........................................ 77
2.20 Schematic of the optical layout of the TARANIS laser system [12] ................................................................. 80
2.21 Mira 900 F layout [13] .......................................................... 81
2.22 stretcher of the 1 Micron Legend F [14] ............................... 82
2.23 The beam pattern seen on the stretcher grating [14] .......... 83
2.24 The regenerative amplifier [14] ........................................... 83
2.25 TARANIS compressor system .............................................. 85
3.1 LASNEX simulation of (a) electron density and (b) the gain of the 326 Å Ne-like Ti line vs the distance from the hydrodynamic expansion from the target. [6] The solid lines illustrate the 2 factors with a prepulse and the dashed lines illustrate the 2 factors without a prepulse ........................................ 93
3.2 Schematic of the TARANIS 2 chamber [1] ......................... 96
3.3 Difference between Ni-Like Mo X-ray Lasing and a Mo Long pulse alone ................................................................. 99
3.4 Optimizing the time delay (\(\Delta \tau\)) for X-ray lasing ........ 100
3.5 Mo growth curve showing a standard Linford fit [1] ............ 101
3.6 Delay scan for the Ni-Like Mo laser at a target length of 5 mm [1] ................................................................. 102
3.7 Final optimized and filtered Ni-Like Mo at 1.0 ns X-ray laser output [1] ................................................................. 103
3.8 Chamber layout for the X-ray emission experiment ............ 105
3.9 Calibration of the Si(Li) detector ........................................... 107
3.10 The counts from \(^{241}Am\) with different filter thickness ....... 108
3.11 Pinhole size impact on \(^{241}Am\) photon counts .................. 109
3.12 Ratio of pinholed counts to non pinholed counts ............. 110
3.13 Measured laser plasma spectrum with different pinhole aperture sizes. Both measurements involved 20 min exposures for a total of 12000 shots each. The detector was 21 cm from the plasma ................................................................. 111
3.14 X-ray counts accumulated at three different lens focal positions ................................................................. 112
3.15 \(K_\alpha\) Counts over a range of different focal positions ....... 113
3.16 X-ray spectrum showing \(K_\alpha\) and \(K_\beta\) of In ................... 114
3.17 $K_{\alpha}$ spectrum from an indium laser produced plasma with counts corrected for the Al filter ........................................ 115
3.18 bremsstrahlung fit equation 3.5 for a series of different temperatures ................................................................. 117
4.1 Schematic of the ultrafast evolution of opacity general set up
(Drawn by B. Dromey) .................................................... 123
4.2 Target holder (front view) for proton interaction studies 124
4.3 Target holder (side view) for proton interaction studies 125
4.4 First image of Proton effects on a camera ........................................ 127
4.5 Probe beam image of the glass ........................................... 128
4.6 Proton penetration through the $40 \mu m$ portion of the filter 129
4.7 Probe beam only after the transient absorption has disappeared. 130
4.8 Probe beam only for Shot 1 with the Al mask 131
4.9 Probe and the TARANIS main pulse for Shot 1 with the pin hole mask blocking the entrance of the glass 132
4.10 Transmission plot of Shot 1 with the mask 133
4.11 Vertical Cross section of Fig. 4.10 134
4.12 Horizontal cross section of the lower proton signature of Shot 1 135
4.13 Transmission plot of Shot 2 with the mask 136
4.14 Vertical cross section of Shot 2 137
4.15 Horizontal cross section of the proton signature of Shot 2 138
4.16 Proton penetration depths vs delays for $\approx7$ J shots 139
4.17 Optical spectrometer for time resolved spectroscopy 141
4.18 Spectrum of the probe beam for optical streaking 142
4.19 Calibrated spectrum of the probe beam 143
4.20 Optical streaked image of the probe beam alone 143
4.21 Probe optical streaking of glass spanning the time window of
25 ps before the TARANIS main pulse and 175 ps after the
TARANIS main pulse 145
4.22 Absorbance equation applied to the probe and the probe and
the TARANIS main pulse 146
4.23 Transmission Vs Calibrated time axis of Fig. 4.22 147
4.24 Transmission vs calibrated time axis of 5 ps to 205 ps after
the TARANIS main pulse window 148
5.1 Schematic of the $1$-m normal incidence spectrometer showing
the paths of 22 nm and 200 nm light 153
5.2 Sensitivity curve of the ILX511 linear array 154
5.3 Sodium salicylate phosphorescence following a 500 ms expo-
sure to the deuterium source with no slit 155
5.4 Comparison of fluorescent spectra of sodium salicylate and p-terphenyl [4] ................................. 155
5.5 Phosphorescence of p-terphenyl to the deuterium source with no slit .................................................. 156
5.6 Effect on counts of adding layers of p-terphenyl .............................................................. 157
5.7 Phosphorescence of P43 to the Deuterium source with no slit ....................................................... 158
5.8 Plate holder adapted with a motor coupled to a pulley system with a 10:1 gearbox ............................. 159
5.9 First spectra of Al on 1-m normal incidence spectrometer, using sodium salicylate and a 1 mm slit .... 159
5.10 First complete spectrum of Al laser plasma, using the camera with the sodium Salicylate on the microscope slide and a 1 mm slit .......................................................... 160
5.11 Comparison of P43, sodium Salicylate and no phosphor .............................................................. 161
5.12 Focusing of the detector arc using the Hg II 253 nm line .............................................................. 163
5.13 Illustration of the Aluminium/Indium disk absorption target .......................................................... 164
5.14 Mount for the continuum source target ......................................................................................... 165
5.15 Front view of the fitted 1-m normal incidence spectrometer chamber ............................................. 166
5.16 Alignment camera magnified view of the slit ................................................................................. 167
5.17 Schumann-Runge absorption of deuterium continuum over the approximate range of 192 nm to 187 nm ................................. 168
5.18 Schumann-Runge absorption of the emission from an Aluminium laser plasma ............................. 169
5.19 Aluminium emission lines obtained from a laser plasma in the region where Schumann-Runge absorption occurs ......................................................... 169
5.20 (a) Four Al lines viewed from three different detector positions illustrated by the colours red, green and blue and brought to overlap well on the middle Al I 193.645 nm line (b) a zoomed in view of the Al II 199.053 nm line under the conditions of Fig. 5.20 (c) a zoomed in view of the Al II 193.645 nm line under the conditions of Fig. 5.20 (d) a zoomed in view of the Al III 186.279 nm line and the Al III 185.471 nm line under the conditions of Fig. 5.20 (a) ................................................................. 170
5.21 (a) Plot of NIST Angle vs Pixel for the 5 long wavelength lines shown in Table 5 ......................................... 171
5.22 Plot of the improved NIST angle vs Pixel after shifting the pixels to optimize the residuals ................ 173
5.23 Figure showing the agreement of the residuals with the spectrum .................................................... 175
5.24 Overlapping of the first two camera positions to agree with
the detector function of Fig. 5.22 ........................................ 176
5.25 First collated segment of the master Al spectrum .................. 177
5.26 Complete uncalibrated Al master spectrum .......................... 177
5.27 Complete pixel vs angle curve for all 159 Al lines identified
from the NIST database. ..................................................... 178
5.28 Complete calibrated Al master spectrum ............................... 179
5.29 Calibrated Al spectrum from 15 nm to 50 nm ....................... 179
5.30 Calibrated Al spectrum from 50 nm to 100 nm ...................... 180
5.31 Calibrated Al spectrum from 100 nm to 150 nm .................... 180
5.32 Calibrated Al spectrum from 150 nm to 200 nm .................... 181
6.1 Top view photograph of the target chamber set up for the DLP
system for the 1-m normal incidence spectrometer .................... 183
6.2 The continuum spot plasma burn pattern .............................. 184
6.3 The absorbing plasma column burn pattern .......................... 185
6.4 Block diagram of the timings required to run both lasers ......... 186
6.5 Delay of the Spectron and Surelite lasers with a flash lamp
trigger delay of C=T=220 ns .............................................. 187
6.6 Plot showing the measurement of \( \Delta z \) by plotting the contin-
uum counts as a function mechanical feedthrough position ... 188
6.7 Time evolution of the Photoabsorption of In and In\(^+\) at a
range of delays from 50 ns to 400 ns .................................... 189
6.8 RTDLDA comparison with the experimental observed Spec-
trum [6] ........................................................................ 191
6.9 Synthetic spectra based on RTDLDA calculations of 4d trans-
sitions of the first four Tm ion states .................................. 192
6.10 Synthetic spectra based on Cowan code calculations of 4d trans-
sitions in Tm to Tm\(^{3+}\) .................................................. 193
6.11 Time evolution of the Photoabsorption of a thulium laser plasma
at a range of time delays from 150 ns to 1900 ns .................... 197
6.12 Comparison of Tm Photoabsorption on the 1-m normal inci-
dence spectrometer to the work of Tracey [4] ....................... 199
6.13 Comparison of Tracey’s spectrum of neutral thulium [4] with
the photoabsorption of the thulium plasma at a time delay of
\( \Delta \tau = 1900 \) ns. .......................................................... 200
6.14 RTDLDA comparison with photoabsorption of the thulium
plasma at a time delay of \( \Delta \tau = 1900 \) ns. ............................... 201
6.15 Comparison of the Cowan code autoionized output and the
photoabsorption spectrum recorded by Tracey [4] .................. 203
6.16 Comparison of the non autoionized transitions calculated by
Cowan and Tracey’s spectrum [4] ....................................... 205
6.17 Comparison between using 5d decay channels and using 5d + 7s decay channels ........................... 206
6.18 Comparison of the Cowan code autoionized output and the recorded spectrum with laser delay $\Delta \tau = 1900$ ns ........... 209
6.19 Comparison of synthetic autoionized spectra to an experimental spectrum obtained at a laser time delay of $\Delta \tau = 1900$ ns 211
6.20 Thulium photoabsorption spectra at delay times which demonstrate absorption due to Tm$^+$ ....................... 212
6.21 RTDLDA comparison with photoabsorption of the thulium plasma at a time delay of $\Delta \tau = 1900$ ns ............... 213
6.22 Comparison of different scaling factors of singly ionized thulium to the photoabsorption measurement at $\Delta \tau = 400$ ns .... 215
6.23 Comparison of synthetic spectra to experimental spectra obtained at a laser time delay of $\Delta \tau = 400$ ns .................. 217
6.24 Comparison of synthetic spectra and experimental spectra at $\Delta \tau = 375$ ns ............................................. 219
6.25 Photoabsorption spectra recorded at delay times which demonstrate the presence of Tm$^{2+}$ ions in the plasma ........ 221
6.26 Comparison between the synthetic RTDLDA spectrum and the experimental spectrum recorded at a delay of $\Delta \tau = 225$ ns. 222
6.27 Comparison of synthetic spectra generated with different scaling factors for autoionized doubly ionized thulium to the photoabsorption measurement at $\Delta \tau = 225$ ns .............. 224
6.28 Comparison of synthetic spectra with experimental spectrum obtained at a laser time delay of $\Delta \tau = 225$ ns ............... 226
6.29 Spectrum of the plasma recorded at a time delay of $\Delta \tau = 150$ ns, compared with the synthetic spectra of Tm$^{2+}$, Tm$^{3+}$ and Tm$^{4+}$ 228
6.30 Photograph of the line focus with a ruler to estimate the focal area for the fs reheating experiment ............... 232
6.31 Block timing diagram of the Darwin triggering the Spectron 233
6.32 A plot of the photoabsorption of an indium spectrum marking the known lines as a check for the calibration .......... 234
6.33 Spectra showing the effect of reheating of an indium plasma where $\Delta \tau = 500$ ns, $\Delta z = -0.2$ mm with the ultrafast laser focused at the centre of the indium plasma .............. 235
6.34 Spectra showing the effect of reheating of an indium plasma where $\Delta \tau = 1100$ ns, $\Delta z = -0.2$ mm with the ultrafast laser focused 1 mm before the indium plasma ..................... 237
List of Tables

1 Summery of progress in X-ray lasers from 1984 - 2005 90
2 Comparison of outputs of X-ray lasers with different geometries 91
3 Output characteristics of the Ni-like Mo laser pumped using the TARANIS multi TW system while employing the GRIP configuration 104
4 Table comparing the measured energy values of the X-ray K series for In to the published energy values 115
5 Diffraeted angle $\beta$ (NIST Angle) for individual Pixels in Fig. 5.21 172
6 Table showing the calculation of the residuals from the function obtained in Fig. 5.21 172
7 Pixel and NIST angle data used in Fig. 5.22 174
8 Table showing the calculation of the residuals from the function obtained in Fig. 5.22 174
9 Calculated $4d \rightarrow 4f$ transitions for Tm (Cowan code) 194
10 Calculated $4d \rightarrow 4f$ transitions for Tm$^+$ (Cowan code) 194
11 Calculated $4d \rightarrow 4f$ transitions for Tm$^{2+}$ (Cowan code) 194
12 Calculated $4d \rightarrow 4f$ transitions for Tm$^{3+}$ (Cowan code) 195
13 Comparison of the calculated widths and the widths published in Whitfield et al. for two selected lines 207
14 Table showing the transitions identified in Fig. 6.33. All the transitions published by Connerade [1] involve $4d^{10}5p^1 \rightarrow 4d^{10}5p^2$ and transitions published by Duffy [2] involve $4d^{10} \rightarrow 4d^{10}5p^1$ 236


1 Introduction

1.1 Plasmas

1.1.1 Introduction to Plasmas

Plasmas are often described as the fourth state of matter. In everyday life plasmas are rare, although if one considers the form of matter throughout the universe, plasmas are the most abundant form of matter. Plasmas appear throughout the universe as interstellar gas and in the corona and atmosphere of stars. Here on earth plasmas appear in natural strikes of lightning, in man made sparks of gas-filled electric discharge tubes and in Tokamak fusion reactors, for example.

Plasmas can be fully ionized, where electrons and ions are the only constituents or they can also contain neutral unionized constituents. A plasma is defined as any quasineutral gas that exhibits collective behaviour among its charged constituents.

The term quasineutral originates from the approximation that the density of the free electrons in a plasma is approximately equal to the density of the ionic charge in the plasma. Collective behaviour occurs due to the long range electrostatic forces that exist between the plasma’s charged constituents. This means that the particles within a plasma will respond collectively to disturbances such as external electric fields acting on the plasma. This collective behaviour is observed in a plasma beyond a certain range which is known as the Debye length, as defined by equation 1.1.

\[
\lambda_d = \sqrt{\frac{\varepsilon_0 k T_e}{n e^2}} \quad (1.1)
\]

Where:

- \( \lambda_d \) - Debye length
- \( \varepsilon_0 \) - permittivity of free space
- \( k \) - Boltzmann constant
- \( T_e \) - electron temperature
- \( n \) - density of the plasma
- \( e \) - charge of the electron.

Over distances smaller than the Debye length the individual interactions between particles dominate interactions and collective effects are unimportant.
1.1.2 Equilibrium in Plasmas

The ratio of the ion density of one ionized species of a plasma to that of another is an important parameter when defining the state of a plasma. If this were a perfect thermodynamic enclosure there are certain assumptions that could be made about the plasma [1].

1. All ions, electrons and neutral species follow a Maxwell-Boltzmann distribution.
2. The population distribution of states in a given atom follow a Boltzmann distribution.
3. The ratio of the number of ions in a given ion state \( z \) to the number of ions in the state \( z-1 \) obeys the Saha equation.
4. The intensity distribution with respect to frequency due to the temperature of the plasma follows the Planck black-body equation.

A plasma that satisfies the above four conditions is said to be in complete thermodynamic equilibrium. This kind of plasma is unrealistic due to energy losses from the system through the emission of light or expansion. As such there are three other less complete equilibrium conditions which are better at describing plasmas found in nature which only require the plasma to be in a steady state i.e. not rapidly ionizing or undergoing rapid recombination. These three equilibrium states are as follows:

1. Local Thermodynamic Equilibrium (LTE)
2. Coronal Equilibrium (CE)
3. Collisional Radiative Equilibrium (CRE)

These three equilibrium states allow us to define the ratio of ion densities in a plasma.

1.1.3 Local Thermodynamic Equilibrium (LTE)

In LTE the excitation and de-excitation mechanisms are assumed to be governed by collisions. LTE supports assumptions 1 \( \rightarrow \) 3 of the four conditions required for a plasma in complete thermodynamic equilibrium (see Section 1.1.2). In this model however, one cannot assume Planck’s radiation law due to spontaneous emission from the atoms and ions disturbing the thermodynamic equilibrium. Since the LTE condition relies so heavily on collisions
and the temperature fitting of a Saha-Boltzmann distribution, the density of the plasma must be high to support this. A necessary but not sufficient requirement for LTE is that the minimal electron density is given by equation 1.2 [1]:

\[ n_e \geq 1.6 \times 10^{12} \sqrt{T_e(\chi)^3} \]  

(1.2)

Where

- \( T_e \) - electron temperature.
- \( \chi \) - excitation potential (in eV) of the highest energy transition found in the plasma.

The main advantage of this model is that the relative populations of ion states can be found without any knowledge of transition probabilities and are determined solely by the laws of statistical mechanics.

1.1.4 Coronal Equilibrium (CE)

Coronal equilibrium (CE) is typically applied to systems of low density and high temperature such as in the solar corona. In this steady state model excitation and ionization occurs due to collisions of electrons with ions and neutrals and de-excitation occurs due to emission of radiation. The plasma is assumed to be optically thin so very little of the light emitted due to de-excitation is reabsorbed by the plasma. The population densities of the plasma are too small to be able to assume a Boltzmann distribution for temperature measurement. Coronal plasma densities are so low that three body recombination is assumed to be very unlikely, whereas radiative recombination and de-excitation mechanisms are dominant. One can still use the Saha equation to calculate the ionization ratio [2].

1.1.5 Collisional Radiative Equilibrium

The first demonstration of the laser in 1960 [4] marked a new era of plasma research. It was quickly realized that if the laser light was focused onto a small spot by a lens or a spherical mirror that the power density was sufficient to ionize matter into a plasma. Today laser produced plasmas are used in many fields of research including spectroscopy, light sources, imaging, holography and many more.

Collisional Radiative (CR) equilibrium is an intermediate steady state model which is applicable to laser produced plasmas. The CR equilibrium model was developed by Colombant and Tonon [3] and provides equations
that calculate the ion population density ratio between an excited state $Z+1$ and the neighbouring lower ion state $Z$ at a given temperature. The ratio is expressed in equation 1.3, neglecting dielectric recombination.

$$\frac{n_{z+1}}{n_z} = \frac{S(Z, T_e)}{\alpha_r(Z + 1, T_e) + n_e \alpha_3b(Z + 1, T_e)}$$ \hspace{1cm} (1.3)

Where:

- $T_e$ - electron temperature
- $S(Z, T_e)$ - collisional ionization rate
- $n_e$ - electron density
- $\alpha_r(Z, T_e)$ - radiative recombination rate
- $\alpha_3b(Z, T_e)$ - three-body recombination rate

The electron temperature can be estimated from equation 1.4.

$$T_e = 5.2 \times 10^{-6} Z^{\frac{1}{5}} (\lambda^2 \phi)^{\frac{2}{5}}$$ \hspace{1cm} (1.4)

Where:

- $Z$ - the ionization of the atomic species.
- $\lambda$ - the wavelength of the laser (in $\mu m$)
- $\phi$ - the power density of the laser ($W cm^{-2}$)

With the electron temperature calculated from equation 1.4 it is now possible to calculate the collisional ionization rate using the equation 1.5.

$$S(Z, T_e) = \left( \frac{9 \times 10^{-6} \xi_z \left( \frac{T_e}{\chi_z^2} \right)^{\frac{3}{2}}}{\chi_z^2 \left( 4.88 + \frac{T_e}{\chi_z^2} \right)} \right) e^{-\frac{T_e}{\chi_z^2}}$$ \hspace{1cm} (1.5)

Where:

- $\chi_z$ - the ionization potential of charge state $Z$
- $\xi_z$ - the number of electrons in the outermost subshell of charge state $Z$. 
The electron temperature can also be used to calculate the radiative recombination rate and the three-body recombination rate by using equation 1.6 and equation 1.7, respectively.

\[
\alpha_r(Z, T_e) = 5.2 \times 10^{-14} \left( \frac{\chi_z}{T_e} \right)^{\frac{1}{2}} Z \left[ 0.429 + \frac{1}{2} \log \left( \frac{\chi_z}{T_e} \right) + 0.469 \left( \frac{T_e}{\chi_z} \right)^{\frac{1}{2}} \right] \quad (1.6)
\]

\[
\alpha_{3b}(Z, T_e) = 2.97 \times 10^{-27} \frac{\xi_z}{T_e \lambda_e^2 \left( 4.88 + \frac{T_e}{\chi_z} \right)} \quad (1.7)
\]

Where all the variables in equation 1.6 and equation 1.7 have their usual meanings.

Three conditions must be satisfied in order for the CR model to be valid according to Colombant and Tonon.

1. The electron velocity distribution must be described by a Maxwell Boltzmann distribution. This occurs when the time for the electrons to reach equilibrium is smaller than the electron heating time.

2. The density of the ions in stage \((Z+1)\) must not significantly change in the time it takes for the population distribution of charge \(Z\) to be established.

3. The plasma must be optically thin to its own emission.

The CR model can be applied to any laser produced plasma in which collisional excitation is the principal excitation mechanism. These are lasers such as the Surelite III and the Spectron SL805 Nd:YAG described in Section 2.5 and Section 2.6 respectively. This model was applied to calculate the ion populations of Thulium (Tm), Tm\(^+\) and Tm\(^{+2}\) by applying equation 1.3.

### 1.1.6 Atomic Processes in Plasmas

Energy exchange occurs in plasmas through a variety of atomic processes which depend on the plasma density. The atomic processes that are of most interest from the point of view of this work are listed and briefly described:

- **Collisional Excitation**
  
  Collisional excitation occurs when a free electron collides with a neutral or ionic species. In this case kinetic energy is transferred to the atom or ion which results in an electronic transition from the ionic ground state to an excited state. The inverse process
involves a collision, which results in a de-excitation of an excited ion species to that ion ground state, with the transfer of kinetic energy to the free electron involved in the collision.

- Collisional ionization

  Collisional ionization occurs when a free electron collides with a neutral or ionic species, again resulting in kinetic energy being transferred to the atom or ion. In this case the kinetic energy transferred is higher than the ionization potential of the atom or ion. The ionized electron will then function as a free electron in the plasma. In the inverse of collisional recombination two electrons encounter an ion, with the recombination of one of those electrons with the ion species, increasing the kinetic energy of the ion or atomic species and of the remaining free electron.

- Photo-excitation

  Photo-excitation or photoabsorption occurs when an atomic or ionic species absorbs a photon resulting in the excitation of an electron from the atom or ion ground state to an excited state. The inverse of photoabsorption is photo-de-excitation also referred to as photoemission in which an excited electron relaxes to the atom or ion ground state with the emission of the excess energy as a photon, which has energy equal to the energy separation between the excited and ground state.

- Photoionization

  Photoionization occurs when an atomic or ionic species absorbs a photon, which has energy higher than the ionization potential of the atomic or ionic species. The electron is then ejected into the continuum. Photo-recombination is a process where an electron recombines with an ionic species with the release of a photon.

1.2 Ultrafast Lasers

1.2.1 Introduction

Ultrafast lasers are highly versatile devices which have many applications in physics, engineering and in medicine. The high power densities associated with these devices have opened up many new fields including High Harmonic
Generation [5], [6], Proton Acceleration [7], [8], [9] and X-ray Lasers [10]. In addition to this, lower energy ultrafast lasers also have applications used as ultrafast probes.

Ultrafast lasers have a fundamental property defined from the Heisenberg uncertainty principle that states that the limiting factor of how short a pulse can be made is related to the spectral bandwidth of the ultrafast laser, since the spectral bandwidth is proportional to the energy. This is referred to as the time bandwidth product and has been instrumental in the design of ultrafast oscillators and amplifiers. The requirements of the time bandwidth product are summarized in equation 1.8 [11].

$$\Delta E \Delta \tau = 1.82 \cdot eV \cdot fs$$  \hspace{1cm} (1.8)

Where:

- $\Delta E$ - FWHM of the energy of the laser photons
- $\Delta \tau$ - FWHM of the temporal pulse width.

1.2.2 Ti: Sapphire as a Lasing Medium

The $Ti:Al_2O_3$ laser (also known as a Ti:Sapphire laser or titanium-sapphire laser) is the most popular ultrafast laser in use throughout the world. $Ti:Al_2O_3$ is the gain medium used in the both the UCD SpecLab Group’s TW laser discussed in Section 2.2, Section 2.3, and Section 2.4 and in the TARA-NIS multi TW laser system in the The Centre For Plasma Physics, Queens University, Belfast discussed in Section 2.7.

$Ti:Al_2O_3$ has a broad optical absorption band as shown in Fig. 1.1. The solid line in Fig. 1.1 demonstrates absorption for $p(\pi)$-polarized radiation and the dashed line demonstrates the absorption of $s(\sigma)$-polarized radiation.
Figure 1.1: Absorption cross Section of Ti : Al₂O₃ as a function of wavelength [12]

The peak of the absorption cross Section lies very close to 500 nm as shown in Fig. 1.1. The pump lasers used by the mentioned ultrafast lasers namely the frequency doubled Colibri discussed in Section 2.2.2, Darwin discussed in Section 2.3.2 and the Surelite-III discussed in Section 2.5 all operate at 532 nm which lies close to the centre of the frequency dependent absorption cross Section shown in Fig. 1.1. Scientific literature contains examples of Ti:Sapphire being pumped at different wavelengths that span the wavelength range shown in Fig. 1.1 and at all of these wavelengths Ti:Sapphire demonstrates almost quantum limited conversion of pump energy into useful emission. [12]

The emission wavelength range of the Ti : Al₂O₃ crystal is demonstrated in Fig. 1.2. The $\pi$ and $\sigma$ polarized emissions are labelled in Fig. 1.2. The oscillator dependent gain line shape is displayed as the dashed line, which is dependent on the oscillator length.
The wide emission bandwidth shown in Fig. 1.2 makes the Ti : Al$_2$O$_3$ excellent in terms of tunability and in terms of spectral bandwidth limits for short pulse generation. A birefringent optic incorporated into a suitable cavity configuration allows tuning of the wavelength mode which will be amplified in a cavity. This technique is employed in the Mira-900-F, the oscillator of the TARANIS laser system described in Section 2.7.1.

The broad bandwidth emission from Ti:Sapphire also makes it excellent for supporting short pulses as the time bandwidth-product requires the potential for a broad spectral output in order to support short pulses.

1.2.3 Mode Locking

Mode Locking (ML) is the first step in any ultrafast laser system. Mode Locking spectrally broadens the pulse of an oscillator which defines the minimum temporal pulse duration achievable not only from the mode locking oscillator but from the subsequent amplification stages that may follow.

Lasers typically operate through an inherent resonance between the system's frequency dependent gain line shape function and the modes defined by the length of the cavity. Once gain saturation begins to occur in a homogeneously broadened gain medium such a Ti : Al$_2$O$_3$, the gain line shape function will only overlap with one mode which typically lies close to the centre frequency of the gain line shape function. When a laser settles into

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{figure1_2.png}
\caption{The emission of Ti : Al$_2$O$_3$ as a function of wavelength [12]}\end{figure}
only amplifying a single mode in this manner it is said to be operating in Continuous Wave (CW) mode.

A laser can also be made to operate with multiple modes which traverse the system together as a train of regularly temporally spaced pulses. These pulses can be made very short in terms of temporal pulse width. A laser operating under such conditions is said to be ML. A ML train of pulses have the following properties:

- The spacing between respective ML pulses can be expressed as:

\[ \Delta T = \frac{2l}{c} \]  

(1.9)

Where:

- \( l \) - length of the of the cavity
- \( c \) - speed of light
- \( \Delta T \) - pulse spacing

- The temporal pulse widths become narrower as the maximum number of modes \( (N) \) being amplified increases. As \( N \) increases the temporal pulse width \( (\tau) \) of the pulses approaches:

\[ \tau = \frac{\Delta T}{N} \]  

(1.10)

The separation between ML pulses governed by equation 1.9 is simply a distance = speed \( \times \) time relationship as each ML pulse in the train will be separated by a round trip time of the pulses traversing the oscillator. The pulse width property explained by equation 1.10 is a consequence of the time-bandwidth product explained in Section 1.2.1. In simple terms, a larger number of modes means the spectral bandwidth of the pulses will be wider, which results in an ability to create shorter pulses.

1.2.4 Mode Locking Techniques

The properties of ML pulses and how they differ from CW operation were described in Section 1.2.3. A number of different techniques for achieving ML are described below, followed by a more detailed description on Kerr-lens ML. Kerr-lens ML is the method employed in the Ti-Light oscillator in use at UCD.

- Dye Jet ML
ML is achieved by placing a saturable absorber in the Ti:Sapphire cavity [13]. The usual saturable absorber used in this wavelength region is 1,1',3,3',3'-hexamethylindotricarbocyanine iodide (HITCI) which is positively charged, indocarbocyanine dye used on the near infrared (NIR). ML occurs as the leading temporal wings of the pulse are absorbed but as the centre, higher energy part of the pulse saturates the dye, leaving the more energetic part of the pulse unaffected. As the beam passes through the dye multiple times, the dead time of the absorber will result in absorption of the trailing temporal wings of the pulse until eventually all that is left is a high energy saturating short pulse. The advantage of this kind of ML is the self-start nature of it without any triggering by the user. Also, the concentration of the HITCI is shown to have little impact on the final temporal pulse width, which makes the specifications of the saturable absorber flexible.

- Coloured Glass Filter

- Moving Mirror
Additive-Pulse Mode Locking

- ML is achieved by attaching an external cavity to the oscillator. A component of the mode locked output is reflected off a beam splitter where it is focused into a non-linear fiber via a lens. The output of the fiber is then reflected back into the fiber and back to the beam splitter by a mirror on the other side of the fiber. The length of the external cavity and the fiber is matched to the length of the cavity. The fiber will induce an intensity dependent phase shift to the incoming beam which will interact interferometrically with the exiting beam of the oscillator. If the path length of the external cavity is tuned correctly, the intense part of the beam from the external cavity will constructively interfere with the peak of the cavity output, while the temporal wings will interact destructively, resulting in pulse shortening [16].

The method of ML employed by the lasers in this thesis is the self focusing Kerr-lens ML. Since the first demonstration of self-focusing Kerr-lens ML [17] it has attracted considerable attention due to ease of use and the reliability. The system has also generated temporal pulse widths close to the limits imposed by the spectral bandwidth of the Ti:Sapphire crystal.

A standard Kerr-lens system layout consists of a (high reflectivity) high reflectivity (HR) mirror and an output coupler (OC) mirror along with a pair of spherical mirrors, which focus the spontaneous emission of the crystal back into the crystal. The oscillator uses a pump beam, focused onto the crystal through one of the spherical mirrors. The oscillator also contains an intra-cavity prism pair the function of which is to compensate for linear and higher order dispersion.

Kerr-lensing or self-focusing is a process in which the refractive index of the crystal becomes a function of the laser intensity. The Gaussian beam self-focuses because the radial intensity distribution of the Gaussian beam leads to the refractive index varying as a function of beam radius, forming a positive lens shape. [24]. When the beam is focused into the crystal using the spherical mirrors, this beam focusing occurs in both space and time [18]. This is evident in Fig. 1.3 which shows the results of numerical modelling. The spot size and the temporal pulse width as a function of distance inside the crystal are shown on the respective left and right axis of Fig. 1.3 (a). The intensity as a function of distance is shown in Fig. 1.3 (b).
It has also been shown that the initial cycles of the nonlinear passes through the crystal have an associated transient oscillatory behaviour in both spectral bandwidth and in the pulse length which after a number of passes through the crystal both stabilize to a steady state as shown in Fig. 1.4. The spectral bandwidth oscillations are shown in Fig. 1.4 (a) with increased beam passes through the crystal and the temporal pulse width oscillations are shown in Fig. 1.4 (b) with increased beam passes through the crystal.
The same calculations show that on each pass through the crystal the beam emerges positively chirped, which needs to be compensated for in order to obtain the shortest pulses. The intra-cavity prisms serve this function. The linear chirp (second order spatio-temporal distortions) can be completely compensated for by choosing prisms of any material and appropriately separating them to compensate for the path difference of the various wavelengths of the beam. Investigation has also shown that shorter temporal pulse widths can also be achieved if the prism pair also compensate for third and fourth order spatio-temporal distortions. This can only be achieved by using prisms made of special non-dispersing materials such as LaKL21 glass [19].

Compensating for these higher order spatio-temporal distortions allows for ML operation with temporal pulse durations as small as 17 fs.

1.2.5 Chirp Pulse Amplification (CPA)

ML oscillators can generate an optimum temporal pulse width but generally only with an energy of the order of 5 nJ. Such a small energy results in a maximum focused power density of the order of $10^{10}$ W cm$^{-2}$ which is not enough for applications such as X-ray lasers or HHG which require power densities of the order of $10^{16}$ W cm$^{-2}$. For such applications there is a need to be able to add considerable energy while still preserving the short temporal
pulse duration. The difficulty with such short pulse durations is that the peak power of the beam is very high and that optics can be easily damaged during amplification.

The discovery of Chirped Pulse Amplification (CPA) provided a solution to this problem [20]. CPA involves stretching the output of the oscillator using a dispersive medium, generally a diffraction grating, which induces Group Delay Dispersion (GDD) as different wavelengths can travel different path lengths due to the wavelength dependence on the diffraction angle from a grating or the wavelength dependence on the refractive index of a prism. The dispersed beam will now have lower peak power due to the pulse duration being broadened by GDD. The stretching process normally involves multiple reflections of the beam off the diffraction grating as demonstrated on the Odin-II stretcher grating in Fig. 2.7 and in the TARANIS Micron Legend stretcher grating in Fig. 2.23. The purpose of the multiple reflections is to spatially reconstruct the beam before further amplification.

The pulse can then be amplified through multiple passes through a gain medium without the concern of damage due to the dispersed nature of the beam from the stretcher. There are two configurations of amplifier:

- Multi pass amplifier
- Regenerative amplifier

Multi pass amplifier systems involve a fixed, small number of passes through the crystal amplifier. The multi pass amplifier uses an array of mirrors to reflect the pulse repeatedly through the crystal amplifier, with the pulse taking a slightly different path at each pass [21]. Examples of this kind of configuration are illustrated in the 8-pass and 2-pass amplification stages of the Odin-II described in Section 2.3.4 and in the Thor amplification described in Section 2.4.3.

A regenerative amplifier is one in which an optical cavity is formed. The beam passes through the amplifying crystal a large number of times which can be varied by electro optic elements. A regenerative amplifier provides diffraction limited amplification which accumulates to high gain over a large number of passes [22]. An example of a regenerative amplifier configuration is the 1 Micron Legend F as part of the TARANIS laser system described in Section 2.7.2.

The amplified beam is then compressed by a pair of dispersive media, usually diffraction gratings whose separation and angular tilt compensate for both the GDD caused by the stretcher component of CPA, and the GDD obtained from other dispersive optics through the amplification process. The first grating induces angular dispersion on the incoming beam, which gives
rise to GDD. The GDD dispersion then increases as the pulse propagates away from the first grating. After the required dispersion is achieved, a second inverted grating is used to re-collimate the beam by compensating for the angular dispersion [23]. The result is a GDD caused by the first grating and frozen by the second grating [22].

1.2.6 Frequency Resolved Optical Grating (FROG) Technique

For many years the main tool for measuring the temporal pulse width of a laser pulse was an autocorrelator. A schematic of a second harmonic generation (SHG) autocorrelator is shown in Fig. 1.5. The autocorrelator works by using the pulse to measure itself. The pulse is passed through a beam splitter and a variable delay to one of the pulses labelled \( E(t - \tau) \) in Fig. 1.5. The two split beams \( E(t - \tau) \) and \( E(t) \) are focused onto the Second-harmonic-generation crystal where the second harmonic output is observed by a detector when the two pulses arrive close together (as shown by the dotted blue arrow). If the pulse arrival time is mismatched the two beams just pass through the crystal as illustrated by the two green lines either side of the blue arrow in Fig. 1.5.

![Figure 1.5: SHG autocorrelator geometry [27]](image)

This detected intensity observed by the detector can be used to reconstruct partial information about the input pulse using equation 1.11.
\[ A(\tau) = \int_{-\infty}^{\infty} I(t)I(t - \tau)dt \]  

(1.11)

Where:

- \( A(\tau) \) - The intensity autocorrelation
- \( I(t) \) - intensity of the un-deviated component of the beam
- \( I(t - \tau) \) - intensity of the variably delayed beam.

The autocorrelation function does yield some measure of the temporal pulse width because no second harmonic intensity will be produced if the pulses do not partially overlap in time. The problem is that the intensity autocorrelation is only a measure of the beam’s magnitude, but we do not have information about the temporal phase, which adds many ambiguities as to the type of pulse being measured. An example of these ambiguities is illustrated in Fig. 1.6. Two pulses, with different temporal phases, are demonstrated in red and blue in Fig. 1.6 (a). The corresponding autocorrelation of the same two pulses is shown in Fig. 1.6 (b) along with a Gaussian pulse in green.

![Figure 1.6: Autocorrelation ambiguities](image)

It is clear from Fig. 1.6 that very different pulses can yield the same autocorrelation trace. This is the reason why it is impossible to extract the temporal phase information from the amplitude, as clearly illustrated in Fig. 1.6. Many different temporal phases can share the same magnitudes, which leads to the well-known 1-D phase retrieval problem [25].

The solution to this problem was found by Trebino et al [26], who used a device called a Frequency Resolved Optical Grating (FROG). An example of a second harmonic generation (SHG) FROG geometry is illustrated in Fig. 1.7. The FROG works by splitting the incoming beam just like in the autocorrelator and applying a variable delay to one of the pulses labelled...
\( E(t - \tau) \) in Fig. 1.7. The two split beams \((E(t - \tau)\) and \(E(t)\)) are focused onto the second-harmonic-generation crystal. The second harmonic output passes to the spectrometer when the two pulses arrive close together temporally (as shown by the dotted blue arrow). If the pulse arrival time is mismatched, the two beams just pass through the crystal as illustrated by the two green lines either side of the arrow in Fig. 1.7. At near arrival times of \(E(t - \tau)\) and \(E(t)\) the SHG signal enters a spectrometer where it is spectrally dispersed onto a camera.

![Figure 1.7: Schematic of an (SHG) FROG geometry [27]](image)

The camera of the FROG detects what is known as a spectrogram. Graphically, the spectrogram is a plot of the frequency of the input beam against the delay introduced by the variable delay path. The spectrogram also contains information about the intensity of the pulse at a given frequency and delay time. A typical spectrogram observed for a set of three pulses, each containing a different degree of linear chirp, is illustrated in the bottom row of Fig. 1.8. The top row of all three pulses illustrates how the frequency of the pulse changes with time. The spectrogram of three different chirped pulses, a negatively chirped pulse, an un-chirped pulse and a positively chirped pulse are illustrated underneath each respective pulse of Fig. 1.8. It is clear from Fig. 1.8 that the tilt of the spectrogram is always in the same direction as the direction of the linear chirp of the input pulse. The spectrograms also illustrate in false colour, the intensity of the respective pulses, where purple represents the highest intensity and red illustrates the lowest.
In addition to the graphical advantages of the spectrogram, it can be expressed mathematically as shown in equation 1.12:

\[
S_g^E(\omega, \tau) \equiv I_{FROG}(\omega, \tau) = | \int_{-\infty}^{\infty} E_{\text{sig}}(t, \tau) \exp(-i\omega t) \, dt |^2 \tag{1.12}
\]

Where:

- \( E_{\text{sig}}(t, \tau) \propto E(t)|E(t - \tau)|^2 \) the exact form depends on the nonlinear optical interaction employed by the FROG geometry
- \( w \) - the frequency of the input beam

An algorithm called general projections is then applied to extract \( E(t) \) from the spectrogram \( S_g^E(\omega, \tau) \). The algorithm is basically a deconvolution and the general form is graphically depicted in Fig. 1.9. The algorithm involves knowledge of two sets of constraints, one of which is the set of \( E_{\text{sig}}(t, \tau) \) values which satisfy the nonlinear-optical constraint (dependent on the FROG geometry) which is illustrated by the green oval in Fig. 1.9. The second constraint consists of the set of \( E_{\text{sig}}(t, \tau) \) that satisfy the data constraints of the measured spectrogram (according to equation 1.12), illustrated by the brown oval in Fig. 1.9.
The algorithm solves a two-dimensional phase retrieval problem in which the single value solution is $E_{\text{sig}}(t, \tau)$ which is the only overlapping value between these two constraints (illustrated as the solution point at the centre of the two constraint sets) [26]. The algorithm begins with a random set of complex numbers which is labelled (1.9) as an initial guess for $E_{\text{sig}}(t, \tau)$. These initial guesses are then used as possible values that satisfy the nonlinear constraints (as shown by the orange arrow connecting the initial guess and the green oval as seen in Fig. 1.9). The results of this projection are then proposed for the set of possible values that satisfy the data constraints (as shown by the far right orange arrow connecting the green oval to the brown oval in Fig. 1.9). The first surviving values of the nonlinear-optical constraints and data constraints are then projected back onto the nonlinear-optical constraints as shown by the orange arrow returning from the brown oval to the green oval in Fig. 1.9. This process is iteratively repeated, each time reducing the number of $E_{\text{sig}}(t, \tau)$ possibilities until only one unique value for $E_{\text{sig}}(t, \tau)$ remains.

Since the general projections algorithm has obtained a single value of $E_{\text{sig}}(t, \tau)$, the final step of the 2-D phase retrieval problem involves solving for $E(t)$. This is done by simply assuming that $E_{\text{sig}}(t, \tau)$ is the one-dimensional Fourier transform, with respect to a dummy variable $\Omega$, of a new signal field $E_{\text{sig}}(t, \Omega)$. The $E(t)$ phase extraction is then completed by setting $\Omega$ to zero, illustrated by equation 1.13:

$$E(t) \propto E_{\text{sig}}(t, 0) \quad (1.13)$$
In equation 1.13 there is a complex multiplicative constant but its description lies outside the scope of this thesis.

These spectrograms are then used as a set of constraints in conjunction with geometrical constraints in an iterative algorithm called the general projection which yields a value for $E_{\text{sig}}(t, \tau)$. The 2-D phase retrieval technique is a useful method that can yield a value for the temporal phase $E(t)$, which is effectively full knowledge of the pulse. To conclude, the FROG is a device that generates a spectrogram which measures the pulse duration, frequency components and intensity simultaneously.

1.3 X-ray Lasers

1.3.1 Introduction

X-ray lasers are sources of light with a great number of applications owing to their short wavelength and therefore potentially very high spatial resolution in imaging. Ever since the first demonstration of the X-ray laser by D.L Matthews [28] there has been considerable interest in the development of the field in terms of the theoretical description of the mechanism and experimental diagnostics and characterization of the source. X-ray lasers are of considerable interest in fields such as characterization of materials, high resolution metrology, atomic physics, photochemistry, biological imaging, holography and the diagnostics of very high density plasmas.

Currently there are four principal methods of generating coherent an X-ray radiation.

1. High Harmonic Generation (HHG) of high power optical or infrared lasers
2. Synchrotron Sources
3. Free Electron Lasers (FELs)
4. Amplified Spontaneous Emission (ASE) in plasmas

ASE is the mechanism relevant to this thesis and is used in Section 3.3 in the amplification of an X-ray laser.

Typical optical and infrared laser systems utilize a positive feedback system which consists of a pair of mirrors which reflect spontaneous emission back into the system, which themselves force amplifying transitions in the medium through a process called stimulated emission. The gain realized by the system is then proportional to the length of the gain medium, where the length of the medium can be increased with multiple passes.
There are many challenges in making X-ray lasers that are not present in optical or infrared resonators.

1. Mirrors are considerably less reflective at X-ray laser and EUV wavelengths, which mean that positive feedback, will not be able to amplify to the same high levels.

2. Population inversion has a very transient lifetime at X-ray wavelengths and is usually shorter than the time taken for light to traverse a cavity a useful number of times.

3. Population inversion requires a pumping power which is proportional to $\lambda^{-3}$ [24], which means that very high intensity lasers are required to achieve the necessary population inversion. (The first pumping of an X-ray laser was achieved with a power density of $5 \times 10^{13}$ W cm$^{-2}$ focused to a $0.02 \times 1.12$ cm line [28].

To this end ASE is typically conducted in a single pass geometry with no mirrors or attempt at feedback. The pump laser arrives at the gain medium usually focused to a line with a cylindrical lens and pumps the medium into a state of population inversion. The spontaneous emission will occur over $4\pi$ and as this sphere of light expands it will result in stimulated emission amplification along the line plasma during the transient lifetime of population inversion (10’s of ps). During this period gain occurs according the Linford equation shown in Equation 1.14 [29].

$$I = \left( \frac{E}{g} \right) (e^{gl} - 1)^{\frac{3}{2}} (gl)^{-\frac{1}{2}}$$

(1.14)

Where:

- $g$ - small signal gain coefficient.
- $E$ - constant proportional to the emissivity.
- $l$ - length of the plasma column.

The gain length product $(gl)$ is normally quoted as a comparison benchmark of the X-ray output. This factor is only physically meaningful when the X-ray laser does not operate in a saturated regime as when a laser saturates it quickly deviates from the Linford behaviour.
1.3.2 Collisional Electron Excitation

Collisional electron excitation was the first mechanism experimentally observed for X-ray lasing and the first one to be theoretically understood. In this mechanism the ground state electron is excited to the population inversion upper level, predominantly by direct electron impact collisions. The population inversion is assisted by having both a dipole forbidden direct transition to the ground state and the lower levels being depopulated quickly by strong dipole-allowed transitions to the ground state.

The first observation of an X-ray laser utilized a Ne-like collisional electron mechanism of Se [28]. The Ne ion has a full outer shell which serves as a bottleneck ion resulting in an abundance of this lasing ion in the plasma. Ne-like ions always lase through a 3p-3s transition. A simplified diagram of the laser transitions of interest for the Se X-ray laser is illustrated in Fig. 1.10. The ground state 2p⁶ is collisionally excited up to one of the five 3p excited states. The return decay is dipole forbidden. Lasing occurs through the five 3p → 3s transitions the transitions which are amplified. The population inversion is maintained by the rapid decay of the 3s back down to the ground 2p state.

![Figure 1.10: Schematic energy level diagram for relevant transitions of Ne-like Se^{+24}[30]](image)

37
The disadvantage of using a Ne-like collisional electron scheme is the very high pump power required to achieve the degree of ionization. An alternate scheme and the one of interest for this thesis is the Ni-like collisional excitation scheme. The Ni-like excitation scheme is very similar to the Ne-like collisional electron scheme except the lasing transitions involve $4d \rightarrow 4p$ transitions. The Ni-like collisional excitation scheme was first demonstrated with $Eu^{+35}$ as a lasing medium. A simplified diagram of the laser transitions of interest for the $Eu^{+35}$ X-ray laser is illustrated in Fig. 1.11. Electrons are occasionally excited from the $3d^{10}$ ground state up to one of the three $4d$ excited states. The direct return transition is once again dipole forbidden and so lasing occurs between the $4d$ upper states and one of the $4p$ lower levels. Once again the population inversion is supported by a rapid decay of $4p$ electrons down to the ground $3d^{10}$.

![Figure 1.11: Schematic energy level diagram for relevant transitions of Ni-like Eu$^{+35}$ [31]](image)

The Ni-like lasing ions produce amplification at shorter wavelengths than the Ne-like scheme for a given state of ionization. The Ni-like collisional scheme was applied to Ag using the Centre for Plasma Physics group’s TARANIS laser system as described in Section 3.3.
1.4 Proton Acceleration

1.4.1 Introduction

Proton acceleration is another field that has resulted from the development of ultrafast lasers over the last number of years. Modern laser based proton acceleration can be first traced to three experiments conducted at the turn of the millennium [7], [8], [9]. These experiments reported high intensity multi-MeV proton acceleration from irradiating thin metallic or plastic targets. The protons were found to originate from surface hydrocarbon or water contaminants which are present in typical lab conditions [32].

There has however, been much debate about which side of the target, the protons originate. One theory is that the protons emerge from the rear of the target in a process called Target Normal Sheath Acceleration (TNSA), which is discussed in Section 1.4.2 and is the direction taken for the purpose of this work. Another leading theory is that the protons originate from the front of the target in a process named Hole Boring. In this, the radiation pressure pushes the over dense ionized target inwards, resulting in a steepened density profile. The radiation pressure and the hot electron temperature results in a number of collision-less shock waves, which are consistent with the reflection of ions from the shock front [33].

Even with the indecision concerning their origin there is confidence for the potential for applications for high energy collimated proton beams. Applications include proton radiography [34], fast ignition fusion [35], proton heating [36] and hadron therapy [37]. The use of proton beams for hadron therapy is the main focus of the present work.

Proton beams are of particular interest for hadron therapy of cancer due to the way that protons deposit their energy compared to the more conventional X-ray or electron beams used. The depth at which a proton deposits its energy is directly dependent on the energy of the proton which reduces the ionizing damage induced to shallow and deep tissue relative to the target tumour. In addition to protecting the healthy tissue the well localized Bragg peak associated with proton energy deposition ensures higher irradiation at the stopping point which is chosen to be the location of the tumour. A comparison of the different ionizing sources is demonstrated in in Fig. 1.12.
Laser produced proton beams are not yet at the point where 200 MeV protons can be generated. Work is continuing into new ways of increasing the laser to proton energy conversion efficiency which is currently at about 12% [8].

### 1.4.2 Target Normal Sheath Acceleration (TNSA)

Target Normal Sheath Acceleration (TNSA) is believed to be the main mechanism through which proton beams can be accelerated using high intensity lasers. There are two main processes, which lead to the acceleration of protons from a thin target surface, the ponderomotive force and the sheath acceleration.

If a plasma is below the critical density, where the frequency of the laser pulse is not the same as the oscillation frequency of the electrons ($\omega_p$), the plasma is said to be under dense. When a laser pulse passes through such a plasma it will impart energy to the electrons. This force is derived from the
full Lorenz force, which has the following form:

$$F = q(E + v \times B)$$  \hspace{1cm} (1.15)

Where:

- \(E\) - electric field of the laser
- \(v\) - velocity of the electrons in the atom
- \(B\) - magnetic component of the laser

The force can be evaluated using second order perturbation theory. The force works by considering an equilibrium state for the electrons and a first order perturbed state, where the electrons oscillate due to the electric field of the laser only. The electrons will oscillate up and down between the first order perturbed state and the equilibrium state.

A second order perturbed state occurs when the magnetic component of the electromagnetic wave distorts the oscillation of the electrons. When a charged particle comes into contact with a magnetic field, the charged particle will begin to orbit the magnetic field line. This magnetic field, which depends on the \(v \times B\) term in equation 1.15, disappears when the electron returns to the equilibrium state. The net effect is a type of helical drift of the electrons in the direction of the laser pulse, which speeds up whenever the electron is accelerated towards the first perturbed state by the electric field, and drops off as the electron is accelerated back towards the equilibrium state by the electric field. The force that acts on the electrons in this manner is what is known as the ponderomotive force [39].

The equation of the ponderomotive force on a plasma is given as follows:

$$F = -\frac{\omega_p^2}{\omega^2} \nabla \left( \frac{\varepsilon_0 E^2}{2} \right)$$ \hspace{1cm} (1.16)

Where:

- \(E\) - electric field of the laser
- \(\omega_p\) - plasma frequency
- \(\omega\) - frequency of the laser.

When a short pulse is incident on the surface of a metal, there is no time for thermodynamic equilibrium to be reached (see Section 1.1.2) and therefore, the energy distribution of the accelerated electrons is not a simple Maxwellian distribution. The distribution contains a quantity of electrons that have low
energy (cold electron temperature), which defines the bulk of the electrons. There is also a quantity of high energy electrons (hot electron temperature), directly accelerated by the laser. The cold electrons do not have sufficient energy to penetrate the target material, however hot electrons do and in doing so, they set up a Debye sheath (see equation 1.17) at the rear of the target.

This Debye sheath, which results from a combination of the hot electron density and a relativistic temperature, will establish a very strong electric field according to the equation 1.17 [40]:

\[
E \approx \frac{kT_{\text{Hot}} \text{ Megavolts}}{e\lambda_d \text{ micron}}
\]

(1.17)

Where:
- \( k \) - Boltzmann Constant
- \( T_{\text{Hot}} \) - Temperature of the hot electrons
- \( e \) - charge of the electron
- \( \lambda_d \) - Debye length

The proton acceleration from the sheath is demonstrated in Fig. 1.13. The hot electron density moves quickly through the target. The hot electrons (labelled with the black dashed line in Fig. 1.13) penetrate far out of the rear of the target (labelled with the red line in Fig. 1.13). The hot electron density will fall with the ion density to maintain quasi-neutrality but will escape slightly further than the ion scale length from the rear of the target. The region, from where the ion scale length ends to the point where the hot electron scale length finishes forms a strong Debye sheath as shown in Fig. 1.13. The cold electrons (labelled by the blue dashed line in Fig. 1.13) do not have the energy to penetrate as far out as the ions or the hot electrons. An ion charge sheet is formed, from a point where the cold electrons penetrate, to where the Debye sheath forms.
Figure 1.13: Proton acceleration from an electrostatic sheath at the rear of a target [40].

A strong potential difference forms between the ion charge sheet and the Debye sheath which has a magnitude governed by equation 1.17. This electric field is responsible for accelerating protons from the ion charge sheet. This process continues until the hot electrons are energetically depleted by accelerating ions.

The hot electrons formed by the ultrafast laser travel in bunches, where the bunch length is on the order of one laser pulse length. This occurs due to the fact that the hot electrons are highly relativistic and are traveling at almost the speed of light. These hot electrons tend to reflect off the Debye sheath at the rear of the target causing circulation back into the target. This circulation is sensitive to the target thickness which is demonstrated in Fig. 1.14. A thick target, defined as one in which the thickness of the target L is greater than half the pulse length ($L_p$) is shown in Fig. 1.14 (a). A thin target is one in which L, as shown in Fig. 1.14 (b), is smaller than half of the pulse length ($L_p$).
If a 100 fs pulse is considered, $L_p$ can be obtained by multiplying the temporal pulse width by the speed of light. We obtain:

$$L_p = (3 \times 10^8 \text{m/s}) \times (100 \times 10^{-15} \text{s}) = 30 \mu\text{m}$$

If the target length $L$ is over 15 $\mu\text{m}$ the circulation of the electrons is similar to that in Fig. 1.14 (a), where on reflecting off the sheath the electrons circulate back into the target very locally, contributing very little to the effective electron density. On the other hand, if the target length is lower than 15 $\mu\text{m}$ the electrons recirculate as shown in Fig. 1.14 (b), which has shown in simulation to increase the density of hot electrons without significantly changing the temperature of the system [41].

The plot illustrated in Fig. 1.15 reflects the measurements and calculations of the peak proton energy as a function of different target thickness. It can be observed in the thin target situation described in Fig. 1.14, that the peak proton energy falls in a steep quasi-linear trend. This trend was maintained in the region lower than 15 $\mu\text{m}$ ($\frac{L_p}{2}$ for a 100 fs pulse) in Fig. 1.15.
For the thick target situation described in Fig. 1.14 (over 15 µm) the peak proton energy also dropped but with a much flatter slope as illustrated in Fig. 1.15.

![Figure 1.15: Plot of peak proton energy as a function of target thickness][42]

This decrease in the energy with target thickness is connected to the recirculation of the electrons and the change in the electron density already discussed. The increased hot electron density with the smaller thickness target has the effect of lowering the Debye length of the sheath according to equation 1.1. A much smaller Debye length results in an increase in the electric field according to equation 1.17 which results in a higher acceleration of the protons and therefore a higher final energy.

1.4.3 Proton Interactions with Insulators

The protons produced by the interaction of the TARANIS laser with thin foils are very useful as probes of the interaction of the protons with various materials. Studies up until recently on proton-matter interactions have primarily focused on microsecond (µs) [43] and nanosecond (ns) [44] proton pulses which to date have been created through chemical scavenging techniques. The laser produced protons are formed with initial picosecond (ps)
bunch duration, high particle numbers and reasonable beam spatial quality. These features of laser accelerated proton beams provide an excellent framework for the study of time resolved proton-matter interaction dynamics, on ultrafast time scales in aqueous solutions and crystalline solids.

The crystalline solid chosen is SiB$_2$O$_3$ borosilicate (BK-7) glass which has a band gap of $\sim 3.5$ eV. Previous work has observed both intense photon and ps electron pulses which induce strong polarization in SiO$_2$ that form a high density of electrons and holes [45], [46]. This resulting electron hole plasma has been previously documented to have given rise to a transient opacity to optical probe radiation following the initial irradiation of intense photon or ps electron pulses. After the transient period, assuming the damage threshold for the material had not been exceeded, the material returned to the original levels of transparency. The electrons in the electron hole plasma are thought to reside in the band gap just beneath the conduction band, allowing a probe pulse of (1.5 eV) to provide sufficient energy to promote electrons into the conduction band [45].

The work presented in this thesis aims to build on this previous work, by forming the electron hole plasma with a burst of protons, accelerated with an ultrafast laser. The probe pulse allows for the observation of the same transient opacity formed with protons, while demonstrating a proof of principle of the experiment with the intent of extending the experiment to water phantoms for biomedical applications and other more complicated materials by dosimetry and imaging of opacity.

1.5 Photoabsorption of Thulium (Tm)

1.5.1 Dual Laser Plasma (DLP) Technique

A schematic of the DLP set up for photoabsorption is shown in Fig. 1.16. A continuum back lighter is formed by focusing a ns temporal pulse duration laser onto a tungsten (W) or a lanthanide target. The result is a spot plasma which emits broad band continuum radiation formed by recombination radiation, bremsstrahlung and super transition arrays due to the large number of open shells in the ions involved. A component of this light travels along the optical axis as shown in Fig. 1.16 and is recorded (as $I_0$) in a spectrometer. Next a ns laser is focused to a line plasma via a cylindrical lens onto the absorbing target (Tm in this case), while at some controlled delay later the continuum plasma is formed as before on the W target (as shown in fig 1.16). The result is light from the continuum source, interacting with the absorbing plasma, which absorbs specific wavelengths from the broad continuum source. The remaining light of the continuum is then detected by
the spectrometer, and is recorded (as \( I_1 \)).

\[ \frac{I_0}{I_1} = e^A \]  

(1.18)

Where

- \( A \) - Absorbance

The absorbance is now available as a function of wavelength to be compared with the absorption features predicted for the absorbing plasma based on a combination of plasma and atomic structure codes.

### 1.5.2 Atomic Structure Codes

The majority of the theoretical calculations conducted in this work were computed using the Hartree-Fock with configuration interaction code due to Cowan [48]. Since the majority of the work presented here is experimental, the code was used as a tool to facilitate the comparison of the experimentally obtained photoabsorption spectra to theoretically predicted spectra.

The code numerically calculates radial wave functions in order to determine the transitions set by the user in the input to the code. It then solves the Schrödinger equation with the calculated radial wavefunctions in order
to produce a transition oscillator strength as a function of wavelength. Thus
the code provides a mechanism for the user to easily calculate what discrete
transitions may be observed in the experimental spectra. The term energies,
electrostatic, spin orbit and exchange parameters may be scaled from their
\emph{ab initio} values as an aid to interpreting the experimental spectrum.

Another code used for this work was the Relativistic Time Dependent
Local Density Approximation (RTDLDA) code [49]. The code replaces the
many electron Schrödinger equation with a total energy function. The system
is then disrupted by an external electric field which results in a frequency
dependent atomic polarizability. The photoabsorption cross-section can then
be calculated from the atomic polarizability with the simple relation shown
in equation 1.19.

\[
\sigma(w) = 4\pi \frac{w}{c} \text{Im} (\alpha(w))
\]  

(1.19)

Where:

* $\alpha$ - atomic polarizability
* $w$ - input frequency (Rydbergs)
* $\sigma$ - photoabsorption cross section (Mbars)
* $\text{Im}$ - imaginary component

This work uses the David Relativistic Code to calculate bound free continuum
transitions.

**References**


49


2 Lasers

2.1 Introduction

The UCD Spectroscopy group has worked with lasers since 1970 when it purchased a ruby laser. Currently there are eleven lasers in the group ranging from basic Nd:YAG ns lasers to a Ti:sapphire 1-Tw 10 Hz system. This chapter will describe all the lasers used in the experiments which form the basis for this thesis, starting with the Ti:sapphire 1-TW laser which consists of the Ti:Lite Oscillator, the Odin-II amplifier and the Thor amplifier. The Ti:Lite described in detail in Section 2.1 outputs between 215 mW and 235 mW of ML power at 810 nm at 778 MHz. The Odin-II then amplifies this through the CPA technique described in Section 2.3 and can output either a compressed 3.2 mJ at 10 Hz or 2.9 mJ at 1 kHz in 30 fs or an uncompressed 5.8 mJ at 10Hz to be further amplified by the Thor amplifier. The Thor amplifier discussed in Section 2.4 continues the amplification of the uncompressed Odin-II output, resulting in a final output of 30 mJ at 10 Hz at 30 fs.

Two ns lasers are also discussed in this chapter. The first is the Surelite-III discussed in Section 2.5, which is a Q-switched laser system with an output 800 mJ at 1064 nm at 7 ns pulse duration. About 470 mJ of this energy is frequency doubled and reflected into the Thor amplifier as a pump laser. The remaining fundamental wavelength can be used for experiments. The second ns laser discussed in this chapter is the Spectron-SL805 described in Section 2.6, which again is a Q-switched Nd:YAG laser which outputs 1.1 J in 18 ns.

Finally the TARANIS laser system in the Center of Plasma Physics in Queens University, Belfast, is also described in detail. The oscillator of the TARANIS system is the Mira 900 F described in Section 2.7.1 and typically outputs 300 mW of ML power at 1053 nm at 76 MHz. This ML train enters the first amplifier known as The 1 Micron Legend F, discussed in Section 2.7.2, which amplifies the beam through CPA and outputs 0.4 mJ to be amplified by the Nd:Glass amplifiers. The Nd:Glass amplifiers discussed in Section 2.7.3 can amplify the beam significantly. The beam is then compressed by a pair of gratings, which output a final energy of up to 40 J in 800 fs. The repetition rate of the final output is 1 shot every 10 mins. The repetition rate of TARANIS is limited by the time needed to completely discharge the flash lamps of the Nd: Glass amplifiers. This also gives the Bd:Glass an opportunity to cool which reduces the possibility of thermal lensing.
2.2 Ti:Lite Oscillator

2.2.1 Introduction

The Ti-light is the oscillator stage for the UCD Speclab Group’s TW laser. The purpose of this system is to generate a broad spectral output described in Section 1.2.1 which can then be amplified in the subsequent amplification stages of the laser. The broad spectral output is generated using the Kerr-lens mode-locking technique (see Section 1.2.4) which generates a train of temporally compressed pulses. This train of pulses can then be used both to trigger other systems of the laser and to create the spectrally broadened output required for CPA amplification (see Section 1.2.5).

2.2.2 Colibri Pump Laser

The Colibri is a diode pumped frequency doubled Nd:YVO$_4$ laser, which is yttrium orthovanadate doped with neodymium. Nd:YVO$_4$ crystals are known to have a high and smooth absorption cross-section at near infrared wavelengths. This makes it a better choice than the more common Nd:YAG crystal for use in small cavity lasers where diode pumping is required and space is at a premium [1]. Investigations have also revealed that a diode pumped Nd:YVO$_4$ is more favourable from the point of view of both the power output and the beam-waist than a Nd:YAG laser of the same design [2]. This enhanced output is also favourable for intra-cavity second harmonic generation (SHG) such as that used in the Colibri.

The Colibri is pumped with a tuneable diode pump and is temperature regulated at 26.4 °C. The diode has a center wavelength between 750 and 850 nm. The 1064 nm output is frequency doubled by a lithium triborate (LiB$_3$O$_5$) (or LBO) crystal that is temperature regulated at 41.9°C. LBO is used because of its good conversion efficiency for SHG and high damage threshold. The heat is applied to the LBO crystal to both improve the crystal phase matching for SHG and to prevent condensation from forming on the crystal. The laser outputs a 532 nm beam at a repetition rate of 778 MHz.

2.2.3 Ti-Light Layout and Operation

The schematic and layout of the Ti-Light Ti:sapphire oscillator is illustrated in Fig. 2.1.
The Colibri beam is guided to the Ti: Sapphire crystal by two plane mirrors (shown but not labelled in Fig. 2.1). The beam is then focused on to the Ti: Sapphire crystal via lens L1, the pump beam focusing lens. The output of the crystal reflects between HR (the high reflector mirror) and OC (the output coupler). These two mirrors are what define the oscillator. There are two paths that lead from the crystal to one of these mirrors. In one of these paths the crystal output reflects off C1 (the first curved mirror), this beam then reflects off M1 (mirror 1) and passes through PR1 (Prism 1). This beam then reflects off mirrors M2 and M3 before being reflected into PR2 (Prism 2). This spectrally stretched beam then reflects off the HR mirror and back along the same beam path until the beam reflects off spherical mirror C1.

The beam is then focused back into the Ti: Sapphire crystal where amplification occurs and reflects off spherical mirror C2 where it reflects off mirror M4 into the OC. The OC reflects a component of the beam back into the oscillator, where it will follow the same path already described, amplifying with every pass through the Ti: Sapphire crystal.

The operation of the laser, discussed so far, describes continuous wave (CW) operation of the oscillator where the amplified wavelength is only related to the length of the cavity. The spectral output of the laser operating
in CW mode is demonstrated in Fig. 2.2. The center wavelength is measured to be 784 nm. The spectrum in Fig. 2.2 was measured using an Ocean Optics USB-2000+ spectrometer.

![Figure 2.2: CW spectral output of the Ti-Light.](image)

The CW output of the laser can vary between 195 and 230 mW.

As explained in Section 1.2 a large spectral bandwidth is a fundamental requirement in achieving ultra-short pulses. The spectral bandwidth shown in Fig. 2.2 is too narrow for compression to fs pulses. The solution is for the oscillating beam in the Ti-Light to be ML. This gives a reduced temporal pulse width output with a larger spectral bandwidth. Mode Locking is achieved by momentarily moving PR2 (Fig. 2.1) into the beam path between OC and M4 with a motorized actuator. The beam becomes dispersed and also corrected for spatio-temporal distortions by PR1. The mode locking mechanism is discussed in Section 1.2. The output of the beam also passes through a beam splitter (BS1) before exiting the Ti-Light. A small component of this will be reflected to BS2 and M5 where the output of the beam is
monitored by two photodiodes. While in CW mode, the photodiodes observe a pulse of light every time the 784 nm pulse traverses the cavity (HR to OC). In ML operation, the photodiodes will observe a train of pulses consisting of other neighbouring modes which are being amplified. A typical ML train, as seen by the photodiodes, is illustrated in Fig. 2.3. The red arrow between the peaks in Fig. 2.3 shows the separation between peaks to be 12.85 ns. The reciprocal of this value gives a repetition rate of 778 MHz.

Figure 2.3: Output of the photodiodes while the Ti-Light is in ML operation.

The train of pulses shown in Fig. 2.3 is used as the mode locked trigger signal for subsequent amplification stages. The passing of the beam between Prism 1 and Prism 2 (in Fig. 2.2.3) defines the spectral bandwidth in ML mode. Spectrally, the ML output is of the form of Fig. 2.4. The red line at Full Width and Half Maximum (FWHM) gives a spectral bandwidth of 81 nm. The spectrum was again measured using the Ocean Optics USB-2000+ spectrometer.
The output depicted in Fig. 2.4 is the seed input to the Odin-II amplifier stage. Typically the ML output of the Ti-light is between 215 mW and 235 mW.

2.3 Odin 2 Amplifier System

2.3.1 Introduction

The layout and operation of the Odin-II amplifier system will be considered in detail. First to be considered is the Darwin in Section 2.3.2 which is the main pump laser for the Odin-II. In Section 2.3.3 the stretcher system of the Odin-II is examined. This is where the output of the Ti-light is stretched using multiple passes of a diffraction grating. The stretched pulse will then be reflected into the amplification system as explained in Section 2.3.4. The amplified pulse will then be optionally reflected into the Thor amplifier for further amplification or will continue into the compressor system where the
output is compressed by a grating pair as discussed in Section 2.3.6)

2.3.2 Darwin Pump Laser

The Darwin pump laser is a diode pumped Neodymium doped Yttrium Lithium Fluoride (Nd:YLF) laser. A Nd:YLF laser is used because it has higher energy per pulse compared to the more commonly used Nd:YAG laser. This higher energy comes about from an upper state lifetime of 480 µs, which is a considerably longer lifetime compared to the Nd:YAG upper state lifetime of 240 µs [7]. The Nd:YLF also has the advantage of a lower thermal lensing than Nd:YAG which results in lower optical distortions under high power operation. A schematic diagram of the Darwin cavity is illustrated in Fig. 2.5.

![Figure 2.5: The Darwin pump laser cavity [4]](image)

The pump chamber houses both the diode pump and the Nd:YLF crystal. The lens is required to compensate for the inherent astigmatism associated with Nd:YLF crystal emission. The Q-switch consists of an acousto-optic modulator. A polarizer (shown in Fig. 2.5) is used to purify the polarization of the amplifying beam. The second harmonic generation (SHG) crystal is LBO, the same crystal as is used for the Colibri in the Ti-Light (see Section 2.2.2). The Darwin can operate with a repetition rate of 10 Hz or 1 kHz. The trigger signal is generated by the photodiodes of the Mode Locked Ti-Light (shown in Fig. 2.3) which is then put through a divider which outputs a 10 Hz or a 1 kHz trigger signal used by the Darwin and by the Odin-II Pockel Cell (discussed in Section 2.3.4). The operating energy of the Darwin is \( \approx 3 \ J \).
2.3.3 Odin-II Stretcher Layout and Operation

As discussed in Section 1.2.5, CPA requires a spectrally broad pulse to be temporally stretched before amplification takes place. The Odin stretcher system takes the ML output of the Ti-Light (discussed in Section 2.2.3) and uses a grating to stretch the pulse before sending it into the amplifier. A schematic of the Odin stretcher system is illustrated in Fig. 2.6.

![Fig. 2.6: Odin II stretcher system [5]](image)

The beam enters the stretcher chamber through the output window labelled 1 in Fig. 2.6 where it passes through lenses 2 and 3, a beam collimating telescope. The collimated beam then passes through 4, a Faraday isolator before being steered into the stretcher by steering mirrors 5 and 6. The beam is aligned vertically central and to the left side of the grating (beam pattern 1 in Fig. 2.7), labelled 7 in Fig. 2.6. The stretching beam is diffracted vertically to spherical mirror 8 which focuses the diffracted beam on to flip mirror 9, which lies at the focal plane of spherical mirror 8. Flip mirror 9 then reflects the beam back to spherical mirror 8 which in turn arrives back at grating 7 both spread vertically and with a small shift to the right (beam pattern 2 in Fig. 2.7).

After the beam returns to grating 7 the beam is diffracted towards mirror 10. Mirror 10 then reflects the beam back to the grating, shifted slightly to the left and in between the previous two arrival points (beam pattern 3 in Fig. 2.7). The beam then diffracts back to mirror 8 and mirror 9 as before, followed by one more return journey to grating 7 (beam pattern 4 in Fig. 2.7) and is reflected out to mirror 11 and 12 where it enters the amplifier by way of iris 60. The final beam pattern visible on the grating is illustrated in Fig. 2.7.

62
After stretching, the output power from the Odin-II Stretcher should be approximately 60 mW. The beam is then amplified by the Odin-II amplifier discussed next.

2.3.4 Odin-II Amplifier layout and operation

After the beam has been appropriately stretched as discussed in Section 2.3.3 the beam is then amplified. The layout of the amplifier compartment of the Odin-II is illustrated in Fig. 2.8.
The stretched train of pulses of about 60 mW arrive at mirror 13 where they are aligned into the 8-pass amplifier with mirrors 14 and 15. The beam makes four passes through the Ti-sapphire crystal (labeled 18 in Fig. 2.8) by following the optical path of mirror 17, mirror 19 and mirror 20 where each pass results in a small displacement on all the involved optics. After the completion of the first four passes the beam is reflected by mirror 21 and 22 to the Pockel Cell (PC) (labeled 24 in Fig. 2.8) just missing mirror 29. On entering the PC the train of pulses will pass through a horizontal polarizer which ensures that all pulses entering the PC are horizontally polarized. When the PC receives a short high voltage trigger (discussed in Section 2.3.2) the polarization of any pulse passing through the PC at that time will have its polarization changed to vertical. Any vertically polarized pulse exiting the PC will be allowed to pass polarizer 26 and will be allowed to proceed to periscope 27 and continue amplification. All horizontal pulses will be rejected.

This is illustrated by Fig. 2.9 where a half-wave plate (labeled 25 in Fig. 2.8) is placed before PC output polarizer 26. The waveform displayed in Fig. 2.9 (a) shows the output of the PC with the half-wave plate placed between the output of the PC and polarizer 26. The black plot shows a zoomed image of the missing pulse from the pulse train where the PC has changed the polarization. The diagram in Fig. 2.9 (b) shows the output of the PC with the half-wave plate removed from the system.
In Fig. 2.9 (a) the half-wave plate switched the polarizations output by the PC so outputs normally horizontal are now vertical and visa-versa. The result is that the whole pulse train is transmitted through polarizer 26 and only the pulse picked by the PC is rejected. By removing the half-wave plate, as shown in Fig. 2.9 (b) the plot illustrates the single pulse effected by the PC, which will then be further amplified in the system. This pulse occurs at the same time as the missing pulse shown in Fig. 2.9 (a) as the half wave plate effectively inverts the rejected and transmitted outputs of the PC.
The pulse selected in the PC from the pulse train (of the form of 2.9 (b)) re-enters the multi-pass amplifier at mirror 28 for another four passes of crystal 18 by reflecting between mirrors 17, 19 and 20 where it undergoes another four passes of amplification. Just as before, the beam leaves the multi-pass amplifier through mirror 21 but this time the beam is reflected by mirror 29 with an energy per pulse of the order of 0.3 mJ.

The pulse diameter is then reduced with the combination of lens 30 and spherical mirror 31. The smaller diameter beam then reflects off mirror 32 where it is focused by lens 33. The beam is focused into crystal 35 after being steered mid focus to that point by mirror 34. After the first pass the beam arrives at mirror 36. The beam then reflects to mirror 38 via mirror 37. Mirror 38 then steers the beam through crystal 35 for its second and final pass.

After the beam exits the two pass amplifier stage it passes through lens 39 which is the first optic of a beam expanding telescope. During the beam expanding process the beam reflects off mirror 40 and the final diameter of the beam depends on the state of flip mirror 64. If the flip mirror is disengaged from the beam path then the beam travels towards lens 41 which in conjunction with lens 39 establishes the beam diameter of the compression system. This beam then enters the Odin-II compressor system (as explained in Section 2.3.6). If flip mirror 64 is in the beam path the uncompressed beam is reflected through lens 65 which, in conjunction with lens 39, establishes a beam diameter of 7 mm. The beam exits the Odin-II and enters the Thor amplifier system. The Thor amplifier system is discussed in Section 2.4. The Odin-II uncompressed output is 5.8 mJ at 10 Hz and 4.2 mJ at 1 kHz.

2.3.5 Darwin Pumping Line of the Odin-II Amplifier

The Darwin (discussed in Section 2.3.2) is a pump beam of the Odin-II system. The Darwin pump laser (shown at the bottom of Fig. 2.8) fires directly into a beam expanding telescope (labelled as 50) and is then split by a beam splitter 51. The reflected / lower energy component reflects off mirror 52 where it is focused by lens 53 into the 8-pass amplification crystal (labelled 18 in Fig. 2.3.4). The pump energy remaining after the first pass through the crystal is focused back into the crystal again by spherical mirror 54 passing straight through mirror 17.

The higher energy component that was transmitted by beam splitter 51 is steered to the double-pass amplifying crystal 35 by mirrors 55 and 57 and is focused by lens 56 just before mirror 57. The remaining beam after the first pass of crystal 35 is then refocused into crystal 35 by spherical mirror 58.
2.3.6 Odin-II Compression System

The Odin-II compressor system is designed to recompress the beam after amplification. This is the final stage of CPA (see Section 1.2.5). A schematic of the Odin-II compressor system is shown in Fig. 2.10. The red beam in Fig. 2.10 illustrates the uncompressed beam from the amplifier, the orange beam illustrates a beam vertically spread out after diffracting from a diffraction grating and the blue beam illustrated the compressed beam exiting the system.

![Schematic of the Odin-II compressor system.](image)

The beam enters the compressor (as shown by the red beam of Fig. 2.10). The beam enters periscope 43 which reflects the beam underneath grating 45 to grating 44. The beam diffracts off grating 44 up to grating 45 as shown by the orange beam in Fig. 2.10. The beam then diffracts off grating 45 again following the orange beam path over grating 44 to the roof mirrors which consists of two rectangular mirrors (labelled 46 in Fig. 2.10). The beam reflects back to grating 45 after reflecting off the roof mirrors 46 as shown by the orange beam path. The beam then diffracts off grating 45 following the blue beam path back to grating 44. The beam then diffracts off grating 44 underneath grating 45 and parallel with the incoming uncompressed red beam towards the compressed output aperture. The compressed output of the Odin-II is measured to be $3.27 \pm 1.77 \text{ mJ}$ at 10 Hz and $2.9 \text{ mJ}$ at 1 kHz and was measured during a service.
2.4 Thor Amplifier System

2.4.1 Introduction

The Thor system is the final amplification stage of the UCD Speclab Group’s TW laser system. The Thor system consists of a Surelite III pump laser which is described in Section 2.5, a Pump Relay Imaging Chamber, the Thor Amplifier Section and the Thor Compression System. The output of the Surelite lasers enters the pump relay imaging section of the chamber, the Surelite beam is split using a beam splitter and both components are relayed to the Thor amplifying crystal. This process will be explained in Section 2.4.2. Section 2.4 will describe the amplification of the output from the Odin-II uncompressed output (discussed in Section 2.4.3) which consists of spatial filtering the Odin beam followed by 4-pass amplification. Finally Section 2.4.3 will describe the Thor compression, which is identical to the Odin-II compression system discussed in Section 2.3.6.

2.4.2 Thor Pump Relay Imaging Chamber

A relay imaging system is used in order to split the beam in two and preserve the beam characteristics of both components so they may pump the Thor crystal effectively from both directions. A schematic of the Thor Relay Imaging Chamber is shown in Fig. 2.11.

![Figure 2.11: The Thor Relay Imaging chamber layout [6]](image)

The Surelite III beam (discussed in Section 2.5) enters the system through an iris P1. The beam gets split into two components at beam splitter P4. The reflected component reflects off mirror P5 where it passes through lens P6. While focusing, the beam is reflected through mirror P7 and P8 into
the imaging chamber P9. The chamber P9 is kept under vacuum to stop the focused beam from ionizing the atmosphere thus breaking down the wavefront of the beam. The beam exits the imaging chamber where it reflects off mirror P10 and is reflected out of the Relay Imaging chamber by mirror P11 into the amplification compartment discussed in Section 2.4.

The transmitted component from the beam splitter P4 is similarly focused by lens P12, through the iris P13, where mirrors P14 and P15 steer the focusing beam into imaging chamber P16, which is identical to P9. The output of P16 is then reflected off P17 where it is also reflected into the amplification compartment by mirror P18.

2.4.3 Thor Amplification Section.

A schematic of the Thor amplification system is shown in Fig. 2.12.

![Figure 2.12: Schematic of the Thor amplification system [6]](image)

The output of the Odin-II enters the system from the top left of the system at window 1 in Fig. 2.12. The beam is steered to spatial filter 13 with mirrors 4, 5, 10, 11 and 12. The spatial filter is a pin hole, which removes the higher order diffraction fringes when a beam is focused onto it. These diffraction fringes arise from scattering at the edges of optics and from deformities in the amplifying media. The removal of these higher order diffraction fringes
The spatially filtered beam enters the 4-pass amplification stage through mirrors 14, 15 and 16. The beam reflects off mirror 17, makes its first pass through the crystal labelled C in Fig. 2.12 where upon it reaches mirror 18. The beam reflects from 18 to mirror 19, where after reflection off mirror 19, it makes the second pass through crystal C. The beam then arrives at mirror 20 where after reflecting to mirror 21 it makes the third pass through crystal C to arrive at mirror 22. The beam then makes one final pass through crystal C after reflecting off mirror 23. The fully amplified beam is collected by mirror 24, where after reflecting through mirror 25, 26 and 27 it passes through a beam expanding telescope consisting of lenses 28 and 29 where the pulse is expanded to a 12 mm diameter. The beam is then steered into the compressor system labelled 31 in Fig. 2.12 by mirror 30.

2.4.4 Thor Compression System

The compression system of the Thor is identical to the one in the Odin-II. A schematic of the layout is shown in Fig. 2.10 and the operation is discussed in Section 2.3.6. The output of the Thor compressor is 1.55 mJ when the Thor is not pumped with the Surelite III and 31 mJ when the system is pumped. The spectral output of the Thor compression system is displayed in Fig. 2.13. The spectrum was obtained using the Ocean optics USB-2000+ spectrometer.
The temporal pulse width of the Thor was also measured using a Swamp Optics USB-8.9 FROG, which uses the FROG technique discussed in Section 1.2.6. The FROG measurement was obtained using the Odin-II un-pumped throughput, which was then reflected into the FROG using a 10% reflectivity wedge. The FROG measurement obtained is illustrated in Fig. 2.14. The measured FROG trace shown on the left of Fig. 2.14 illustrates the spectrogram measurement made by the time camera at the end of the spectrometer of the FROG as discussed in Section 1.2.6. The retrieved Frog trace shown on the right is the reconstructed spectrogram calculated from the temporal phase calculated from the FROG algorithm explained in Section 1.2.6.
A successful frog measurement calls for the measured and the retrieved FROG traces look similar such as the traces obtained in Fig. 2.14. The important measurement from the FROG is the temporal FWHM, which is measured to be 39.3 fs.

2.5 Surelite III Pump laser

2.5.1 Introduction

The Surelite III pump laser is the pump laser for the second stage amplifier of the TW laser (see Section 2.4). This laser is a Nd:YAG (Neodymium doped with Yttrium Aluminium Garnet) laser which operates at a wavelength of 1064 nm. The laser typically delivers 800 mJ with a pulse duration of 7 ns. This energy is measured with the Litron LPM251-3 photo diode detector [9]. The Litron is an InGaAs photo diode with a diffuser and a filter which transmits at 1064 nm. The Gaussian temporal profile of the Surelite III laser is illustrated in Fig. 2.15. The pulse duration was measured at full width and half maximum (FWHM) using the temporal oscilloscope output of the Litron Photo detector.
The Surelite output is passed through a second harmonic crystal which has the effect of frequency doubling the output of the Surelite to 532 nm radiation, which is reflected through a high reflectivity mirror into the Thor amplifier. The operation of the Thor amplifier is explained in Section 2.4.2. The second harmonic output is approximately 470 mJ at 532 nm. The second harmonic crystal is not 100% efficient and there is a sufficient amount of fundamental 1064 nm leakage radiation remaining for use in an experiment. The remaining available output at 1064 nm is 370 mJ.

2.5.2 Surelite III Operation with the Cavity Closed

The Surelite III has a simple optical design as illustrated in Fig. 2.16. The diagram depicts the system with the Q-switch closed.
The laser cavity of the Surelite III depicted in Fig. 2.16 consists of a pair of mirrors, one of high reflectivity (HR) and the output coupler (OC) which is partially reflecting. The other main components are the laser head, a pockels cell (PC), a polarizer and a quarter wave-plate. The laser head is a gain medium based on a Nd: YAG crystal rod with two flash lamps and a polarizer is an optical filter that becomes highly reflective when vertically polarized light impinges on it. A quarter wave plate changes the polarization state of the light wave traveling through it and is used to convert linearly polarized light to circularly polarized light and vice versa. The pockels cell is basically a voltage controlled wave plate.

When the laser is turned on, the flash lamps will discharge periodically, and there will be no voltage applied to the pockels cell. Only light that is emitted along the axis of the red arrow in Fig. 2.16 will remain in the cavity. Any light that is not horizontally polarized will be reflected out of the cavity by the polarizer and be lost.

Any horizontally polarized light will follow the lower path shown in Fig. 2.16. The horizontal light will reflect from the OC, pass through the polarizer unaffected and will pass through the quarter wave-plate, which will change the light from horizontally polarized to circularly polarized. With no voltage applied to the PC, the shutter is closed, and this keeps the light circularly polarized as it passes through the PC and reflects off the HR mirror.

Following the upper path shown in Fig. 2.17, the light again passes through the PC unaffected and passes into the quarter wave-plate which
again changes the polarization of the light, this time to vertical. Since the polarizer is highly reflective to vertically polarized radiation it is reflected out of the cavity before it has any chance to be significantly amplified.

### 2.5.3 Surelite III Operation with the Cavity Open

The Surelite III diagram illustrated in Fig. 2.17 depicts the system with the shutter open.

![Diagram](image)

**Figure 2.17: The Surelite III cavity with the shutter open [8]**

The open shutter cavity layout of the open shutter of the Surelite III, shown in Fig. 2.17 is identical to that of the closed shutter discussed in Section 2.5.2. The only difference is that the PC now has 3600 Volts applied to it. The light emitted from the laser head along the axis of the red arrow in the lower path shown in Fig. 2.17 behaves the same way until after the first pass of the 1/4 wave-plate (see Section 2.5.2 for description up to this point). The circularly polarized wave then passes through the PC, but because it is now open, it has the effect of phase retarding the extraordinary component of the light by 90°, making the emerging light vertically polarized.

This light then reflects of the HR mirror and then passes through the PC again which phase retards the extraordinary component of the light by 90° again so it emerges circularly polarized. The beam then passes through the quarter wave-plate and emerges horizontally polarized. The horizontally polarized light passes through the polarizer unaffected and into the laser head where it is amplified by another pass through the laser head. A component
of the light passes though the output coupler as a pulsed beam and the remainder gets reflected back through the cavity for an extra pass and further amplification.

2.5.4 Q-switching in the Surelite III

Q-switching is a technique which allows for the production of pulses of high power because of their high energy in a short pulse duration.

This technique involves discharging the flash lamps while the cavity is closed (see Section 2.5.2). The flash lamp discharge time is approximately $375 \mu s$ but to ensure optimum population inversion, which allows for a high gain coefficient, the PC is triggered on after $235 \mu s$ after the flash discharge trigger [8].

With the cavity opened, (see Section 2.5.3) the light oscillating in the cavity will be greatly amplified and unobstructed, lasing begins with a high-intensity energy release. The PC is triggered off, 7 ns after it was switched on resulting in a very high power pulse. This Q-switching results in an 800 mJ output per pulse and the short pulse duration of 7 ns.

2.6 Spectron SL805 Laser

The Spectron SL805 is the other laser used for photo absorption experiments. Like the Surelite III the Spectron laser is a Nd:YAG which operates at 1064 nm. The maximum pulse energy of the laser is 1.1J. This laser has a number of longitudinal modes generated by the cavity geometry so the system is a compromise between beam structure and extra energy. The typical beam structure and the pulse duration of the Spectron laser are displayed in Fig. 2.18.
The FWHM displayed in Fig. 2.18 is 18 ns with six longitudinal modes.

2.6.1 Spectron Layout

A schematic overview of the layout of the Spectron SL805 laser is presented in Fig. 2.19. This schematic is derived from the description and photographs in the SL800 Pulsed Nd : YAG manual [10].

The Spectron laser (shown in Fig. 2.19) consists of a laser cavity, a pair of steering mirrors, a beam expanding telescope and an amplifier pump chamber. The following components are located in the laser cavity, a curved high
reflectivity (HR) mirror, flat (OC) an inter cavity telescope (ICT), polarizer, oscillator pump chamber and the pockels cell (PC).

The inter cavity telescope is used to compensate for thermal lensing in the rod and reduce the divergence of the beam. Thermal lensing is the inducing of an inhomogeneous refractive index change in the crystal due to thermally induced mechanical stress [11]. This can manifest physically as a change in the radius of curvature of the crystal side mirror of the cavity. The telescope maintains the beam spot size as close to the diameter of the crystal as possible while also compensating for the thermal lensing effects that may compromise the stability of the cavity. The cavity stability is expressed as follows:

\[
0 \leq (1 - \frac{L}{R_1})(1 + \frac{L}{R_2}) \leq 1
\]  

(2.1)

where

- \( L \) - length of the cavity
- \( R_1 \) - radius of curvature of mirror 1
- \( R_2 \) - radius of curvature of mirror 2

As previously mentioned, the telescope has the effect of reducing the divergence of the beam, which allows to obtain a smaller spot size when the laser output is focused in experiments.

The remaining components in the laser cavity relate the oscillator pump chamber, the pockels cell and polarization. The oscillator pump chamber is a gain medium based on a Nd: YAG crystal rod with a flash lamp. The two upper red lines in the schematic illustrated in Fig. 2.19 define the light paths in the laser cavity.

The principle of Q-switching in the Spectron laser is similar to the Q-switching in the Surelite III (see Section 2.5) except that the impact of the pockels cell (PC) is reversed. When the laser is turned on, the flash lamp will also be on, and there will be a voltage applied to the pockels cell. The polarizer plate will only allow horizontally polarized light to oscillate in the cavity.

Only light, exiting the PC from the oscillator pump chamber, that has a horizontal polarization component will not be rejected by the polarizer. This horizontal component then reflects off the HR mirror, passing through the polarizer unaffected and the polarization is further rotated by the PC to circular. After passing through the oscillator pump chamber, reflecting off the OC and back through the oscillator pump chamber, the light polarization is further rotated to vertical by the PC and subsequently rejected by the
polarizer. With the PC switched on, light cannot make more than two passes through the oscillator pump chamber.

If the PC is switched off there will not be any change in the light polarization and horizontally polarized light will remain that way for the complete loop of the laser allowing continuous oscillations.

The optimized output performance of the Spectron laser is related to the switching time between the flash lamp and the pockels cell. When the flash lamp activates, it triggers a quarter wave-voltage ($90^\circ$) to be applied to the PC. This voltage is applied across the PC for between 120 and 150 µs which is the optimum time after the lamp trigger for maximum population inversion. The voltage is then removed from the PC, which stops the beam from being rejected by the polarizer, allowing amplification between the HR and the OC resulting in a high gain in the laser rod. The PC is only switched off for approximately 18 ns. This results in approximately a 100 mJ energy pulse in a short pulse duration.

After the laser cavity, the beam passes through two steering mirrors after which it passes through a beam expanding telescope which expands the beam to fill the entire amplification rod in the amplification pump chamber. The second stage is a single pass rod amplifier consisting of a rod of Nd:YAG surrounded by flash lamps. The final output of the Spectron laser is approximately 1.1 J with a pulse duration of typically 18 ns.

2.7 TARANIS Laser System

The TARANIS Laser system layout is as shown in Fig. 2.20.
It can be seen from Fig. 2.20 that the TARANIS laser system consists of three amplification stages.

- The Mira 900 F (oscillator)
- The 1 Micron Legend F (regenerative amplifier)
- Nd: Glass single pass amplifiers

After the amplification stages the beam is compressed by a grating compressor system. Each of these components is discussed in detail in what follows.
2.7.1 The Mira 900 F

The TARANIS laser system uses $TiAl_2O_3$ (titanium-sapphire) as a gain medium. The $TiAl_2O_3$ is pumped by an all-solid state, single frequency, CW laser which operates at 532nm, the Verdi-18W. This pumping laser can produce up to 14 W. The schematic of the layout of the Mira 900 F is presented in Fig. 2.21.

The Verdi-18W pump laser enters the system as shown in Fig. 2.21 and is focused by lens L. The radiation is then emitted along two beam paths. The first is mirrors M4, M3, M2 and the output coupler (OC) (also labelled M1) and the second beam path is through mirrors M5, M8 and M9, where M9 is the high reflectivity mirror in the system. This setup defines the system in CW operation.

When mode-locking is activated, prism P1 changes the path of the beam from going to M8 to going to M6, where the beam passes through prism P2 and to the other high reflectivity mirror M7. The changing of prism P1 has the effect of changing the length of the cavity allowing neighbouring non resonant modes to undergo gain, leading to a train of pulses, which are in phase.

The laser also has a birefringent filter labelled BRF in Fig. 2.21. This allows for wavelength tuning by changing the polarization of a narrow wavelength range. The components of the beam that are of a different wavelength to the one altered by the crystal are removed by an additional polarizer not shown in Fig. 2.21.

The birefringent filter in the oscillator is set up to only reflect the 1053 nm light. This allows amplification to occur at only 1053 nm. After this, the mode-locked pulses have a pulse duration of 200 fs and a bandwidth of 13
nm. The oscillator has an average power of 300 mW and a repetition rate of 76 MHz.

2.7.2 The 1 Micron Legend F

After the oscillator, the regenerative amplifier or “Regen” is the next step in amplification of the beam. This amplifier takes advantage of the chirped pulse amplification process (CPA) in which a beam is stretched and then amplified. CPA is used to prevent the beam from damaging the optics, as the power of the beam is decreased by stretching of the pulse. The pulse can then later be compressed resulting in an overall major increase in intensity. The stretcher of the 1 Micron Legend F is shown in Fig. 2.22.

![Stretcher Diagram](image)

**Figure 2.22: Stretcher of the 1 Micron Legend F [14]**

The output from the Mira 900 F enters the stretcher as shown at the top of Fig. 2.22, reflects off mirror 1 and passes through the Faraday isolator and then reflects off mirrors 3 and 4 to the vertical retro mirror. The beam is then reflected through the alignment apertures 6 and 7 onto the grating 8. The grating is orientated so that the first order of the diffraction input beam is reflected to a curved mirror labelled 9. This beam is then reflected to the centre of the flat mirror labelled 10. The flat mirror then reflects the beam back to the centre of the curved mirror and then back on to the grating.

This dispersion introduces a path difference in the beam. This dispersed beam is then reflected back along the same path three more times in order to spatially reconstruct the beam. After the four passes of the beam onto the grating there will be a pattern as shown in Fig. 2.23.
The beam enters the stretcher system as a circular beam second from the top in Fig. 2.23. When the beam has completed the four passes of the stretcher it leaves as the circular beam, illustrated in the bottom trace of Fig. 2.23. The beam is then reflected off the pick off mirror labelled 11 in Fig. 2.22 and then passes through the polarization rotator labelled 12 in Fig. 2.22 where it enters the regenerative amplifier part of the system. The schematic of the regenerative amplifier is illustrated in Fig. 2.24.

The pump beam enters the regenerative amplifier part of the 1 Micron
Legend F at the bottom left of Fig. 2.24 (the green beam). This pump beam is generated by an Evolution-15 Nd:YLF pump laser with a frequency doubled central wavelength of 527 nm with a power of 12 W at a repetition rate of 1 KHz. The pump beam is split by beam splitter BS1. Half of the beam is focused onto the Ti:sapphire crystal by the lens L1 and the other half of the pump beam reflects off pump mirror PM2 and PM3 before being focused on to the Ti:sapphire crystal by the lens L2 via pump mirror PM2 as shown by the green beam path in Fig. 2.24.

The vertically polarized stretched seed pulse arrives from the stretcher as shown in Fig. 2.24, where it reflects off the pumped Ti:sapphire crystal in the direction of the mirror M3 towards the mirror M4. Between the mirrors M3 and M4 the stretched beam encounters a quarter wave plate, which rotates the polarization of the stretched pulse by 90°. If pockels cells PC1 and PC2 are not active, then the beam will just reflect off mirror M4, where it will pass through the quarter wave plate again making the beam horizontally polarized. The beam will then reflect off mirror M3, M1 and M2, where the beam will reflect off mirror M2 and make a second pass through the Ti:sapphire crystal, where it will make a second double pass through the quarter wave plate returning to mirror M2 vertically polarized. After reflecting off mirror M2 the beam will be reflected out of the amplifier by the polarizer beam splitter P1, which only reflects vertically polarized light. The stretched beam in this case undergoes little or no amplification from this triple pass through the Ti:sapphire crystal.

For amplification of the beam a voltage is applied to PC1 making it active, after the first pass of the beam through the crystal, which effectively nullifies the effect of the quarter wave-plate thereafter. Since the polarity of the beam remains horizontal, the beam becomes effectively trapped in a loop, reflecting between mirrors M2 and M4, being amplified with every pass through the Ti:sapphire crystal. The polarizer beam splitter P1 will pass the horizontally polarized beam indefinitely. After about 80 passes through the Ti:sapphire crystal, a quarter wave voltage is applied to PC2, making it active, which will permit the pockels cell PC2 to advance the polarization on the pass from P1 to M2 and back again. The vertically polarized light will be reflected out of the amplifier by the polarizing beam splitter P1. While trapped in the amplifier the stretched beam is amplified by about $10^5$ times in energy.

After the amplification stage the beam is passed through a pulse slicer (another pockels cell) where the repetition rate of the output is defined. In normal operation, the pulse slicer is triggered with the capacitors of the ND: glass amplifiers used to further amplify the beam after the regenerative amplifier. The output of the laser can be set to 500 Hz by the pulse picker and then be selected by a flip mirror, allowing the output to pass through
the Micron compressor.

2.7.3 Nd: Glass Single Pass Amplifiers and Spatial Filters

The hole smooths the radial intensity by removing the higher order Bessel terms formed by the diffraction of the beam from the lens. The lens generates the Fourier transform of the spatial profile of the beam, changing it from a top hat function in space, to a sinc function in k space. The oscillating wings of the sinc functions are interpreted as the higher order Bessel terms formed by diffraction of the lens. These higher order Bessel functions form at distances larger than the circumference of the pinhole and do not get through. The output of the pinhole then passes through the second lens which performs an inverse Fourier transform and re-collimates the beam. The remaining portion of the beam then passes through the next single pass Nd: glass amplifier, after which it passes through another spatial filter. This is then repeated one more time before the beam enters the compressor chamber.

2.7.4 TARANIS Compressor System

The compressors system of TARANIS consists of two 21cm × 42cm gratings, a vertical retro reflector made of two 7 inch ×10 inch mirrors and two 5 inch ×7 inch mirrors for guiding the beam into the gratings and for steering the compressed beam into a mirror box. A schematic of the beam path is shown in Fig. 2.25.

![Figure 2.25: TARANIS compressor system](image)

The input beam reflects off the input mirror, and the first order is diffracted off grating G1 to grating G2 and to the vertical reflector. The beam after changing height is then reflected back to grating G2, where it is diffracted back through grating G1 to the output mirror. The output from these compressors goes into the mirror box. Inside the mirror box (not shown in Fig.
2.25), two 5 inch × 7 inch mirrors are used to turn the beam towards the target chamber. The grating assembly is under vacuum. The final output of the TARANIS laser system is approximately 40 J in 800 fs with 15 min between shots.

References


3 Laser Generated X-ray Sources

3.1 Introduction

This chapter addresses the generation of X-ray sources using a laser produced plasma as the source medium. The chapter explores two types of X-ray sources:

1. A nickle-like molybdenum (Ni-like Mo) X-ray laser
2. Inner shell emission from laser produced plasmas

The first experiment was conducted at the Center For Plasma Physics, Queens University Belfast, and was published in the 2009 in the International Society of Photonics and Optics (SPIE) conference proceedings [1]. This chapter references components of the work presented in that paper related to my involvement with the generation of the Ni-like Mo X-ray laser. The novelty of the Ni-like X-ray laser is in the GRazing Incidence Pump (GRIP) target geometry. To this end the chapter begins with a survey of target geometries starting with a demonstration of amplified spontaneous emission (ASE), (discussed in Section 1.3) on exploding foil targets and ending with the most recent target geometries, including GRIP, with the objective of tracing the advantages of different target geometry configurations. The chapter will discuss the experimental set up for making the X-ray laser focusing on the methods of converting X-ray counts into energy.

The chapter will highlight the optimization of the Ni-like Mo X-ray laser including both the effects of the time delay of the prepulse with respect to the main pump pulses and the length of the amplifying target. The different X-ray laser configurations are discussed in terms of power output and efficiency.

The second experiment discussed is inner shell emission from laser produced plasmas. The experiment was conducted in UCD with the UCD Speclab Group’s Thor TW laser (see Section 2.4) and with the Group’s lithium drifted silicon Si(Li) detector. The Thor system’s reliability permitted only preliminary work to be carried out but it may serve to speed up the continuation of this experiment at future dates.

The chapter outlines the experimental setup and a brief general discussion of the operation of Si(Li) detectors, with a detailed emphasis on the calibration and filtering optimization. Some work on X-ray K series generation is highlighted by comparing the analyzed data with the published K series information. The chapter concludes with a brief demonstration of the effect of different focusing conditions on the $K_{\alpha}$ counts along with some conclusions and suggestions for future research.
3.2 Review of Target Geometries for X-ray Lasers

To compare the different target geometries, a literature review was carried out. Data from different X-ray laser publications were compared and tabulated in terms of gain, efficiency and output characteristics. Table 1 summarizes the relevant characteristics of different target geometries, the gain achieved and efficiencies involved in achieving that gain which was reported or inferred from the respective papers.

The criteria used in the comparisons in Table 1 are as follows:

- Reference - reference number of the experiment in this thesis.
- Date - date on which the referred experiment was conducted.
- Novelty - experimental novelty of the target geometry.
- Species - ionic species used in the X-ray amplifier.
- Max Gain - maximum gain coefficient measured in the referred experiment.
- Gain Length - product of the “Max Gain” and the length over which the gain occurred.
- Pump Energy - Pumping energy used to excite the population inversion.
- Prepulse Energy - Energy of the prepulse fired at the optimum time delay before the main pulse.
- Eff Parameter - This efficiency is the ratio of the “gain length” row entry and the “Pump Energy” row entry which was introduced in [7] and applied to the rest of the data in Table 1.
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<td>[6]</td>
<td>1993</td>
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<td>14.4</td>
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<tr>
<td>[8]</td>
<td>1998</td>
<td>Target Smoothing</td>
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<td>2000</td>
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<td>62</td>
<td>18</td>
<td>4.8</td>
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<tr>
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<td>2001</td>
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<td>Ge^{+22}</td>
<td>19</td>
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<td>36</td>
<td>33.6</td>
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<td>2002</td>
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<td>Zn^{+20}</td>
<td>7</td>
<td>21</td>
<td>500</td>
<td>1.6</td>
<td>42</td>
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<td>GRIP</td>
<td>Mo^{+14}</td>
<td>58</td>
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<td>1</td>
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<td>2005</td>
<td>GRIP</td>
<td>Mo^{+14}</td>
<td>55</td>
<td>11</td>
<td>0.15</td>
<td>0.07</td>
<td>73333.33</td>
</tr>
</tbody>
</table>

Table 1: Summary of progress in X-ray lasers from 1984 - 2005

The gain length product does not provide any information on the output of the X-ray laser in terms of energy, divergence, aperture size or pulse duration. It is interesting that it is the only quoted quantity common to all the experiments. It should also be noted that there is no apparent convention for expressing output parameters and therefore it is difficult to benchmark the X-ray output.

The Eff Parameter values shown in Table 1 was only published by Nickles et al. [7] and has been applied to all the other papers cited in this thesis. It should be mentioned that both the gain length and therefore the Eff Parameter have limited meaning if the X-ray laser achieves saturation. The use of gain length in saturated systems is generally accepted in the literature and so is listed here.
A comparison of all the different target geometry novelties presented in Table 1 is illustrated in Table 2. It can be seen that the table is not complete and there are many gaps relating to inconsistencies in the presentations of various papers.

Information could be taken from the individual publications to extract other values implicit in the definition of those quantities. In the case of brightness there is a relationship to all the other quantities depending on what other information is in the publication. Brightness is expressed as shown in equation 3.1:

$$\text{Brightness} = \frac{E}{t\theta A} \text{Wcm}^{-2}\text{sr}^{-1} \quad (3.1)$$

Where:
- $E$ - Energy
- $t$ - Pulse Length
- $\theta$ - Divergence
- $A$ - Emitting Area

<table>
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<tr>
<th>Reference</th>
<th>Wavelength (nm)</th>
<th>Energy (J)</th>
<th>Pulse Length (ps)</th>
<th>Divergence ($\text{sr}$)</th>
<th>Emitting Area ($\text{cm}^2$)</th>
<th>Energy/Solid Angle ($\text{Jsr}^{-1}$)</th>
<th>Brightness ($\text{Wcm}^{-2}\text{sr}^{-1}$)</th>
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</tr>
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</tr>
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<td>[9]</td>
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<tr>
<td>[13]</td>
<td>18.9</td>
<td>1.5 x 10^{-7}</td>
<td>50</td>
<td>N/A</td>
<td>3.8 x 10^{-5}</td>
<td>N/A</td>
<td>N/A</td>
</tr>
</tbody>
</table>

Table 2: Comparison of outputs of X-ray lasers with different geometries

The criteria used in the comparisons in Table 2 are listed as follows:

- Reference - Reference number of the experiment referred to in this document (as per Table 1).
- Wavelength - Wavelength of the obtained outputs.
- Energy - Energy measured at the X-ray line wavelength.
- Pulse Length - The lifetime of the X-ray line.
- Divergence - Expanding solid angle of the beam as it emerges from the X-ray emitting area.
- Emitting Area - The area of the plasma over which the X-ray radiation is emitted.
- Energy / Solid Angle - Is energy that would be detected over 1 steradian.
- Brightness - The power detected per steradian over the emitting area.

As stated earlier, the information from both Table 1 and Table 2 is required to understand the improvements obtained in X-ray laser output and efficiency. For example, one may look at the brightness of reference [12] and conclude that by instead adding a half cavity mirror such as the one used in [5] will yield around the same output brightness. On the other hand, if one considers that the pump laser driving [12] is 1 J and the one used in [5] is 154 J, then it can be reasoned that using the GRIP geometry is superior in terms of the Eff Parameter to simply inserting a half cavity mirror. We will now give brief descriptions of the different geometries.

3.2.1 Exploding Targets

The first demonstration of the X-ray laser was in 1985 [2]. Exploding foil targets, which consisted of a 750-Å layer of Se evaporated onto a 1500-Å substrate of Fomvar (C\textsubscript{11}H\textsubscript{18}O\textsubscript{5}) which was irradiated by a driving laser pulse, focused to a line plasma with a cylindrical lens. The Ne-like Se result involving \(3p \rightarrow 3s\) transitions were then extrapolated to higher Z materials. The Exploding foil targets were used, as slab targets were thought to introduce density gradients in the plasma which would refract the X-ray beam out of the plasma prematurely. Lee et al. were the first to successfully show that the slab thickness had little or no effect on lasing [3].

3.2.2 Prepulse Treatment

A major advance in the amplified spontaneous emission (ASE) (see Section 1.3) technique came with the prepulse technique, where the target is irradiated with a low power (long pulse) pulse a few ns before the main pulse (short
pulse) irradiates the target. This prepulse technique was first demonstrated by Nilsen et al. [6]. The prepulse was found to create a larger region with optimum gain density which also had less steep density gradients as shown in Fig. 3.1.

![Figure 3.1: LASNEX simulation of (a) electron density and (b) the gain of the 326 Å Ne-like Ti line vs the distance from the hydrodynamic expansion from the target. [6] The solid lines illustrate the 2 factors with a prepulse and the dashed lines illustrate the 2 factors without a prepulse.](image)

Fig. 3.1 shows a favorable difference that results from the addition of the prepulse. The larger gain region and the lower density gradient allow the laser to be amplified over the entire length of the plasma column instead of being refracted out by steep density gradients. The time delays between pulses ranged from 4 to 7 ns.
3.2.3 Transient Pumping Scheme

In addition to the prepulse scheme, the transient pumping scheme better utilizes the transient (short lived) nature of the gain region. Collisional excited population inversion works in a quasisteady state in that the population inversion lifetime is much shorter than the lifetime of the plasma. The idea behind the transient pumping scheme is that extremely high gain coefficients can be obtained if the laser pumping time (laser pulse length) occurs at times faster than the inter-atomic relaxation times. Inspection of Table 1 shows the pump energy requirements for lasing are reduced when transient pumping schemes are used [7], [8], [10], [11]. The gain coefficients for the mentioned references are also very high compared to the experiments before the adoption of the transient scheme however, the gain length from the gain length products suggest that the length over which these gains occur is much smaller.

The limited gain region is partly due to the refraction of the X-rays out of the plasma but that is not the only restriction. The short lived high gain regions do not provide sufficient lifetime for the beam to traverse centimeter length plasma columns previously observed on exploding foils and slab target configurations. This limits the high gain region to an order of millimeters of plasma column length.

3.2.4 Traveling Wave Scheme

The traveling wave scheme is a concept used to increase the length of the plasma column to maintain the transient pumping scheme. The plasma is formed as the beam traverses the plasma, meaning that when the amplifying X-ray beam arrives at a point in the plasma column, that point in the plasma has formed later than the previous point and so the new region still has the required density and the population inversion to continue amplifying. In any CPA beam, the pulse is passed through a compressor which consists of a pair of gratings with a carefully chosen tilt and separation to correct spatio temporal distortions such as spatial chirp, temporal chirp and pulse front tilt (see Section 1.2). The traveling wave setup intentionally misaligns the second grating to retain pulse front tilt. The pulse front tilt can then be tailored to compensate for the group velocity lead required to form the amplifying plasma later along the edges of the plasma column. This will preserve the transient conditions all along the column [15]. Table 1 and Table 2 list the experiments where the traveling wave geometry was used [10], [11]. The data in Table 1 does indicate some improvement of gain length products while the gain coefficient is at least as good or better, which suggests that the wave is
amplifying in the plasma for longer.

3.2.5 Grazing Incidence Pumped Configuration (GRIP)

Grazing incidence pumped configuration is the main focus of this chapter in terms of comparing X-ray geometries. The geometry has the same excitation pulse focused to a line as with earlier configurations but the excitation pulse arrives in at grazing incidence as opposed to the standard normal incidence. This novel pumping geometry was first demonstrated by Keenan et al. [13]. The grazing angle is chosen to refract the laser pulse in the plasma. This occurs according to the equation:

$$\theta_r = \sqrt{\frac{n_{eo}}{n_{ec}}}$$  \hspace{1cm} (3.2)

Where:
- $\theta_r$ is the refracting angle.
- $n_{eo}$ is the maximum density within the gain region.
- $n_{ec}$ is the pump laser critical density.

The refraction angle experienced in the gain region permits the beam to turn back into the gain region [13]. This second pass of the pump beam effectively doubles the gain length. The energy output of a GRIP laser is sensitive to the grazing angle and to the delay between the prepulse and the main pump beam [16]. The GRIP geometry is the most efficient geometry as the pump energy requirements are reduced to levels as low as 150 mJ [13]. When compared to the other geometries, the GRIP configuration has some of the highest maximum gain (gain coefficients) accompanied with reasonably good gain length products, however the gain length products are not as high as those of of the traveling wave.

3.3 Ni-Like Mo X-ray Laser

3.3.1 Introduction

In Sections 3.3 and 3.4 we discuss the pumping of a Ni-Like X-ray laser source using the TARANIS multi-Terawatt system at Queens University Belfast. The experiment utilizes the GRIP target geometry explained in Section 3.2.5 to produce X-ray output that can be compared to other X-ray lasers published in the literature in terms of energy, gain length and the Eff parameter.
3.3.2 Experimental Set Up

This experiment was conducted in the TARANIS 2 vacuum chamber. A schematic of the target chamber used for X-ray lasing is shown in Fig. 3.2.

![Schematic of the TARANIS 2 chamber](image)

*Figure 3.2: Schematic of the TARANIS 2 chamber [1]*

The primary detector used is a flat field spectrometer, which is comprised of a 1200 groves/mm Hitachi grating followed by a back thinned Andor high
resolution camera is shown in the bottom left of Fig. 3.2. The function of this camera is to observe the axial output of the X-ray laser and define the axis to which the target lies and to which the lasers are aligned.

The two pulses shown in Fig. 3.2 come from the TARANIS laser described in Section 2.7. The long pulse (LP) (shown in blue) is the reflection from a beam splitter which is taken from the Nd:Glass amplification phase of the beamline, before the compressor. This beam usually has between 5 and 10 J and a pulse duration of the order of 1.2 ns. The beam is focused to a line of length 9 mm and width 80 \( \mu \)m at near normal incidence to the target using a combination of a 300 mm focal length lens and a 304.8 mm radius of curvature spherical mirror. The power densities obtained from focusing the LP vary between \( 5.8 \times 10^{11} \) and \( 1.2 \times 10^{12} \) W/cm\(^2\).

The short pulse (shown in red) is the component of the beam which has continued through the full amplification and compression of the beamline. The energy of the short pulse was between 2 and 5 J and the pulse duration was of the order of 800 fs. The beam enters from the right of Fig. 3.2. This beam is focused to a line 7 mm long and 40 \( \mu \)m in width using a spherical mirror tilted at 10 degrees to the incoming beam in accordance with other reported GRIP schemes [12], [13], [16]. The short pulse is focused to a power density of \( 7.1 \times 10^{14} \) and \( 1.8 \times 10^{15} \) W/cm\(^2\).

Before entering the chamber the long pulse passes through a variable optical delay, which allows the long pulse to arrive at any time between 4 ns before the short pulse and 1 ns after the short pulse. The LP requires realignment to the optical axis every time the time delay is altered.

### 3.3.3 Energy Measurements using the Flat Field Spectrometer

In order to obtain a measurement of the output energy of the X-ray laser using the spectrometer it was necessary to convert the counts obtained from the X-ray laser into an energy measurement. The first step is to convert the counts into an energy incident on the CCD, which is computed using equation 3.3.

\[
Energy = (hv)(\text{Photon\#}) = (hv) \left( \frac{(counts - B_{offset}) E_{eff}}{(EM_{gain})(QE)} \right) \tag{3.3}
\]

- **Photon\#** - number of photons detected by the camera
- **Counts** - number of counts detected after the camera’s signal processing.
- $QE$ - quantum efficiency of the camera in converting photons of a given wavelength into electrons.
- $EM_{gain}$ - an alterable amplification of the initial electrons produced by the photons.
- $EC_{eff}$ - proportion of the produced electrons which register as a count.
- $h$ - Planks consent.
- $v$ - frequency of the incident photons.
- $B_{offset}$ - offset in the software resulting from background subtraction.

All of the quantities of equation 3.3 are obtained from either the CCD camera documentation or from the software that interfaces with the camera. With the energy of the photons known, the energy of the X-ray laser can be determined by calculating the throughput of the spectrometer. The total throughput of this spectrometer was calculated using the following equation:

$$T_{tot}(\lambda) = T_g(\lambda) T_s(\lambda) T_f(\lambda)$$  \hspace{1cm} (3.4)

- $T_{tot}(\lambda)$ - total throughput of the spectrometer.
- $T_s(\lambda)$ - fraction of the beam accepted by the effective slit.
- $T_g(\lambda)$ - is the grating efficiency at the wavelengths involved.
- $T_f(\lambda)$ - transmission function of the Al filter in front of the camera.

A value of 0.2 was used for the grating efficiency $T_g$[1]. This was an old measurement taken from the synchrotron at Daresbury and the value is not likely to be accurate due to additional surface contamination on the grating. $T_s$ the fractional beam acceptance was calculated to be 0.33 [1]. This value was an estimate of the amount of the beam likely to have made it both through the slit and without being clipped by the grating. The value of the transmission function $T_f$ was obtained from the tabulated values of the transmission of Al [19].
3.4 Results and Conclusions

3.4.1 Ni-Like Mo X-Ray Laser

Once the flat field spectrometer was aligned to the focus of the LP and the SP, the experiment involved finding the optimum timing $\Delta \tau$ (discussed in Section 3.3.2) between the arrival of the LP and the SP. Strong X-ray lasing was observed when $\Delta \tau = 1$ ns and is demonstrated in Fig. 3.3. In Fig. 3.3 (a) the observed spectrum displays the camera counts when the LP is followed by the SP with a delay of $\Delta \tau = 1$ ns. The lasing occurs on the 18.9 nm $4d_{3/2} \rightarrow 4p_{1/2}$ and the 22.6 nm $4d_{3/2} \rightarrow 4p_{3/2}$ laser transitions. In Fig. 3.3 (b) the spectrum displays the camera counts when the LP is fired alone.

![Figure 3.3: Difference between Ni-Like Mo X-ray Lasing and a Mo Long pulse alone](image)

The spectrum Fig. 3.3 (a) demonstrates strong amplification of the 18.9 nm $4d_{3/2} \rightarrow 4p_{1/2}$ and 22.6 nm $4d_{3/2} \rightarrow 4p_{3/2}$ laser transitions. The camera in Fig. 3.3 (a) is under filtered hence the over-saturation of the transitions and the
additional presence of Mo lines. In order to establish optimum time delay \((\Delta \tau)\), the spectrum was scanned for values of \(\Delta \tau = 0.5\) ns and 1.5 ns and the results are displayed in Fig. 3.4. In Fig. 3.4 (a) and Fig. 3.4 (b) the observed spectrum displays camera counts when the LP is followed by the SP with delays of \(\Delta \tau = 1.5\) ns and \(\Delta \tau = 0.5\) ns respectively. In Fig. 3.4 (c) the spectrum displays the camera counts when LP is fired alone. The blue dotted line at 18.9 nm in all sub figures illustrates where amplification is taking place for one of the 2 lasing transitions.

**Figure 3.4: Optimizing the time delay \((\Delta \tau)\) for X-ray lasing**

Fig. 3.4 (a) and Fig. 3.4 (b) illustrate weak amplification of the 18.9 nm \(4d_{3/2} \rightarrow 4p_{1/2}\) transition line for \(\Delta \tau = 1.5\) ns and \(\Delta \tau = 0.5\) ns respectively. When contrasted with Fig. 3.4 (c) it is clear that some amplification is taking place at 18.9 nm. Fig. 3.4 illustrates the sensitivity in the plasma conditions necessary for X-ray lasing to occur. At the un-optimized time delays shown in Fig. 3.4 there is no evidence of amplification of the 22.6 nm \(4d_{3/2} \rightarrow 4p_{3/2}\) transition. The saturation of the camera at the \(\Delta \tau = 1\) ns time
delay required that the camera be filtered with Al so that the optimized time delay and the near optimized time delays can be distinguished.

The gain length is then calculated by measuring the output energy (see Section 3.3.3 on counts to energy conversion) of the Ni-like X-ray laser as a function of target length and this is shown in Fig. 3.5. The target length was changed by varying the lens position with respect to the LP focusing mirror.

![Graph showing XRL energy output versus target length]

**Figure 3.5: Mo growth curve showing a standard Linford fit [1]**

From the energy plot in Fig. 3.5 the gain coefficient is found using the standard Linford fit [17]. The gain coefficient was found to be 60 cm$^{-1}$ with a gain length product of 3000. The Linford fit does not agree with the data points for a very long distance. This is due to the laser quickly deviating from small signal gain behavior and beginning to saturate from target lengths of 1.75 mm and longer.

In order to more accurately determine the optimum delay between the LP and the SP, the variable optical delay before the input to the chamber was used at a target length of 5 mm and the output is presented in Fig. 3.6.
Fig. 3.6: Delay scan for the Ni-Like Mo laser at a target length of 5 mm

Fig. 3.6 illustrates that the amplification characteristic is better at delays before the optimum $\Delta \tau = 1$ ns than after the optimum $\Delta \tau = 1$ ns. The drop in signal at earlier delays is due to the plasma still having too steep density gradients formed by the initial prepulse. This results in a reduction in the size of the effective gain region (as discussed in Section 3.2.2). The drop in signal at later delays is due to the cooling of the plasma to below the Ni-like stage which results in reduced gain due to lower achievable population inversion. A 5 mm plasma column length was chosen as Fig. 3.5 illustrates saturated behavior at a 5 mm target length. The final optimized and suitably filtered Ni-like Mo X-ray laser output is shown in Fig. 3.7.
Figure 3.7: Final optimized and filtered Ni-Like Mo at 1.0 ns X-ray laser output [1].

The information in Fig. 3.7 represents two views of the same data. On the left, the figure shows the raw output of the camera. On the right, the binning and the calibration of the raw camera image results in a spectral representation of the observed counts at the calibrated wavelengths.

3.4.2 Conclusions

A Ni-like Mo laser was successfully pumped using the TARANIS multi TW system while employing the GRIP configuration. The laser achieved saturation-like behavior with target lengths of 1.75 mm or more (shown in Fig. 3.5) resulting in a gain coefficient calculation of 60 cm$^{-1}$ from the Linford plot and a corresponding gain length product of 3000. The laser was amplified to approximately 200 nJ once the optimum delay between the Long Pulse and the Short Pulse was used. In order to compare the Ni-like X-ray laser described here to the other similar GRIP configurations discussed in Table 1 and Table 2 a single row of data is provided in Table 3 for comparison and summary purposes.
Comparing the results shown in Table 3 to the other published work in Table 1 it can be seen that the second highest gain efficiency parameter was achieved. Other parameters could not be directly compared to the work presented in Table 2 since the energy was the only output directly measured, it can be said however that $2 \times 10^{-7}$ J was the highest energy of any Ni-like Mo laser discussed in this report or any other reference material. This energy number will have uncertainties with it associated with the assumptions made on the X-ray spectrometer throughput values used in its calculation (see Section 3.3.3).

There are additions that could be made to the experiments to expand the knowledge of the X-ray laser operating characteristics. Further measurements are needed to obtain sufficient information on parameters directly comparable to that presented in Table 2, including the lifetime of the lasing lines and the divergence of the laser light. Also, additional studies of different GRIP angles and observing the effect these have on the energy and on the other measured parameters listed in Table 2 could be performed.

### 3.5 Inner shell X-ray Emission of Indium

#### 3.5.1 Introduction

In Section 3.5 we discuss the X-ray experiment conducted, involving inner shell X-ray emission using the Thor terrawatt system. The experiment focuses the TW laser to an estimated power density of $1.2 \times 10^{18}$ W cm$^{-2}$ to produce K-series X-ray emission which is detected by the lithium drifted silicon detector. The plasma temperature formed by the indium target under these high intensity conditions is estimated to be $6.1906 \times 10^5$ eV according to the CR model discussed in Section 1.1.5 however this model is not applicable for a laser with a temporal FWHM lower than 100 ps. The indium is expected to contain predominantly hydrogen-like and helium-like ions at these temperatures. The mechanism of inner shell X-ray emission involves the laser directly photoionizing an inner shell electron. A K shell transition occurs when a 2p electron de-excites to take its place with the emission an

<table>
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<th>Prepulse Energy</th>
<th>Eff Parameter</th>
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<td>J</td>
<td></td>
<td>KJ$^{-1}$</td>
</tr>
<tr>
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<td>3000</td>
<td>4.5</td>
<td>7.7</td>
<td>$6.667 \times 10^5$</td>
</tr>
</tbody>
</table>

*Table 3: Output characteristics of the Ni-like Mo laser pumped using the TARANIS multi TW system while employing the GRIP configuration.*
X-ray photon with energy in the keV range. The experiment was intended as a preliminary one to guide future research in this area.

### 3.5.2 Experimental Set Up

The chamber for the X-ray emission experiment was conducted in the 50.8 cm diameter feed-through collar chamber. A schematic of the experimental set up is shown in Fig. 3.8.

![Figure 3.8: Chamber layout for the X-ray emission experiment](image)

The Thor TW laser pulse (discussed in Section 2.4) enters the chamber through one of the side ports after passing through a fused-silica anti-reflection coated window (\(\lambda = 800\) nm) and a 20 cm nipple (not shown in Fig. 3.8). The fused silica was required to reduce the chirp induced on the pulse from group velocity dispersion and the nipple was required to reduce the time the Thor TW laser spent traveling in air. The beam will start to self-focus by the Kerr effect if it is allowed to travel in air for more that 20 cm. The laser is then focused on to the In target by a plano-convex lens, which has a focal position that can be controlled with a computer controlled Zaber actuator. The In target is mounted at 45 degrees with respect to the incident beam and is connected to three computer controlled Zaber actuator stages which allows three dimensional movement to the incoming beam. This allows a constant refreshing of the target if desired for the experiment. When evacuated, the vacuum pressure in the chamber was approximately \(1 \times 10^4\) mbar.
3.5.3 Lithium Drifted Silicon [Si(Li)] Detectors

Lithium drifted silicon or Si(Li) detectors are made up of a p-type silicon semiconductor crystal. The detector system is attached to the chamber port (shown in Fig. 3.8). The entrance window contains beryllium and is attached to an arm going inside the chamber. The photons that enter the beryllium window interact with the crystal to produce electron hole pairs, the number of which is in proportion to the energy of the incoming photon. When the electron hole pairs recombine, it results in a deposition of charge on the crystal electrode of the detector. This electrode is electronically connected to a field effect transistor (FET) which is then connected to the preamplifier for the first stage of spectral processing. The FET system operates under cryostatic conditions using liquid nitrogen, as a coolant, and is able to function with very low noise at these temperatures [14]. The Si(Li) detector is a single channel detector that has the advantage of no dispersion.

3.5.4 Calibration and Optimization of the Si(Li) Detector

Before the detector can be used to detect X-ray emission from a laser produced plasma it is necessary to calibrate the channels to an energy axis. This is carried out by using a pellet of the radioactive isotope of Americium $^{241}$Am as a calibration reference. The calibration is shown in Fig. 3.9. Part (a) of Fig. 3.9 shows the three $^{241}$Am lines used to calibrate the Si(Li) detector on a calibrated axis while Fig. 3.9 (b) shows the linear function used for converting a channel number into photon energy (keV).
With the calibration shown in Fig. 3.9 any counts from any channel number can be converted into counts of a given photon energy. The calibration energies were obtained from nuclear tables [18]. The width of the 17.75 keV line was measured at half maximum to have a value of $\Delta E = 0.246$ keV giving the resolution of the instrument to be 70.12.

Since the detector saturates easily with laser plasmas as a source and bremsstrahlung generates much higher counts than $K_\alpha$ and $K_\beta$ emission, it is important to filter the Si(Li) detector with a pin hole aperture in a thick disk of Al, whose aperture is further filtered with thin sheets of Al. Since high counts can be obtained from the $^{241}$Am pellet, it is beneficial to use the pellet to experiment with different filter thicknesses and aperture sizes. The variation of counts at different energies for different filter thicknesses are shown in Fig. 3.10. The information in Fig. 3.10 describes the counts from $^{241}$Am as follows:

- (a) - a 100 $\mu$m thick Al filter
(b) - a 40 µm thick Al filter
(c) with no filter

Figure 3.10: The counts from $^{241}$Am with different filter thickness

All plots in Fig. 3.10 are conducted with a 1 mm pinhole for a period of 10 minutes and at a distance of 13 cm from the source. The red plots in graphs (a) and (b) represent the transmission of the Al as a function of energy up to 30 keV with the transmission expressed on the respective right axis [19]. The blue dotted lines in (a) and (b), that extend the red plots, represent an extrapolation of the transmission up to 60 keV assuming a minimum absorption by the aluminium.

The plots in Fig. 3.10 show that the Al filter does not significantly affect the counts at 12 keV and above, which will prove useful for filtering the bremsstrahlung but not the $K_\alpha$ or the $K_\beta$ emission. The other variable used to limit the number of photons saturating the detector is the aperture diameter. The impact of different aperture sizes on the counts is demonstrated in
Fig. 3.11. Fig. 3.11 (a) and 3.11 (b) shows counts from $^{241}\text{Am}$ with a 0.5 mm pinhole aperture and a 1 mm pinhole aperture respectively. Fig. 3.11 (c) shows the spectrum recorded with no pinhole.

![Graph showing photon counts from different pinhole sizes](image)

*Figure 3.11: Pinhole size impact on $^{241}\text{Am}$ photon counts*

The counts in Fig. 3.11 (c) are much higher than those with a pinhole resulting in a scale that is a factor of two higher.

The data in Fig. 3.11 demonstrate the counter-intuitive energy dependence on the count reduction with decreasing pinhole size. In Fig. 3.12, the data from Fig. 3.11 is expressed as an energy ratio of a pin hole of a given size to no pin hole.
In Fig. 3.12 there are two energy ratio graphs, one for a pinhole size of 0.5 mm and one for 1.0 mm as follows:

- (a) For a 0.5 mm pinhole aperture individual ratios were obtained by dividing counts of Fig. 3.11 (a) by the corresponding no pinhole counts in Fig. 3.11 (c)

- (b) for a 1.0 mm pinhole aperture individual ratios were obtained by dividing counts of Fig. 3.11 (a) by the corresponding no pinhole counts in Fig. 3.11 (c)

The reason for the energy dependence on the pinhole aperture size is not clear. It may be some kind of scattering at lower energies. Further investigation of the detector with different radiation sources and different energies particularly, in the 30 to 60 keV region may give further insight into this characteristic.
3.5.5 Pile Up Phenomenon

A pinhole aperture diameter of 0.5 mm is required in all X-ray experiments involving the laser produced plasma source of X-rays. The 1 mm pinhole gave a pile-up phenomenon which suppressed data sets. Fig. 3.13 demonstrates the pile-up phenomenon. Fig. 3.13 (a) shows the laser produced X-ray counts with a 0.5 mm pinhole and Fig. 3.13 (b) shows the laser produced X-ray counts for a 1 mm pinhole.

![Graph showing measured laser plasma spectrum with different pinhole aperture sizes](image)

*Figure 3.13: Measured laser plasma spectrum with different pinhole aperture sizes. Both measurements involved 20 min exposures for a total of 12000 shots each. The detector was 21 cm from the plasma.*

The effect of pile-up illustrated in Fig. 3.13 (b) results from having too wide a pin hole. It most likely comes about from a high photon flux that arrives at times that are much shorter than the dead time of the detector. The effect of pile-up can be recognized by an accelerated counts in the energy range normally expected for bremsstrahlung. By only changing the pin-hole
to the 0.5 mm aperture as shown in Fig. 3.13 (a) a clear bremsstrahlung signal is visible to 15 keV. There is no $K_\alpha$ and $K_\beta$ lines visible in Fig. 3.13 (a) due to the laser being out of focus by about 0.7 mm.

### 3.5.6 Optimization of X-ray Counts

Preliminary optimization of the focusing conditions for the highest conversion efficiency into X-rays was also completed. Fig. 3.14 shows the counts obtained from a set of three different relative focal positions. Fig. 3.14 (a), 3.14 (b) and 3.14 (c) were measured at relative focal positions $+30 \ \mu\text{m}$, $0 \ \mu\text{m}$ and $-20 \ \mu\text{m}$. All were performed over 120000 shots with a 160 $\mu\text{m}$ filter, with a pin hole of 0.5 mm and with a laser energy of 31 mJ.

![Figure 3.14](image)

*Figure 3.14: X-ray counts accumulated at three different lens focal positions.*

Fig. 3.14 shows the count variations at 3 different focal positions that were varied by the order of 10’s of microns. There is no absolute focus measurement. The $+30 \ \mu\text{m}$ relative focal position shown in Fig. 3.14 (a) are the counts formed from the lens being closest to the plasma, while the $-20 \ \mu\text{m}$ position is the lens furthest away.

112
The $K_\alpha$ count over a range of different focal positions is displayed in Fig. 3.15. All focal positions shown in Fig. 3.15 were performed over 120000 shots with a 160 $\mu$m filter, a pin hole of 0.5 mm and with a laser energy of 31 mJ. Error bars represent the standard deviation (square root of the counts).

$\text{Figure 3.15: } K_\alpha \text{ Counts over a range of different focal positions.}$

It is clear from Fig. 3.15 and Fig. 3.14 that there is no significant difference in the counts over the range that the focal position is moved. All of the counts are within one standard deviation of one another except for the count at relative position $+50 \, \mu$m for which the count was zero.

3.5.7 Analysis of Inner Shell Emission Lines from In Ablation

A typical optimized X-ray spectrum obtained from the experimental set up discussed in Section 3.5.2 is illustrated in Fig. 3.16. Fig. 3.16 depicts the accumulation 12000 shots through a 0.5 mm pin hole filtered with a 60 $\mu$m Al foil and a laser energy of 30 mJ. The second (black) plot in Fig. 3.16 with the blue outline is an expanded image in the 22 keV - 28 keV energy range highlighting the fine features of the $K_\alpha$ and $K_\beta$ emission. The magnified area also shows the measured values of the $K_\alpha$ and $K_\beta$ series.
The red spectrum in Fig. 3.16 shows a typical X-ray spectrum obtained from laser produced plasmas. The accumulation of the counts in the 5 - 12 keV region is bremsstrahlung originating from the high density of free electrons in the plasma, which originate from the laser ejecting inner shell electrons. The energy of the bremsstrahlung is related to the temperature of the plasma. The discrete states shown in the 23 - 28 keV region is the \(K_\alpha\) and \(K_\beta\) which are transitions from 2p and 3p states respectively. It should be noted that the reason the bremsstrahlung counts are lower than the \(K_\alpha\) and \(K_\beta\) discrete lines is due to the Al 60 \(\mu\)m filter transmittance (discussed in Section 3.9) at the bremsstrahlung energies. The plot in Fig. 3.17 (b) demonstrates what the data in Fig. 3.16 would depict if there was no Al filter. A fourth order polynomial fit of the transmission data for Al [19] is presented in Fig. 3.17 (a). This filter transmission is divided by 100 and applied to the data from Fig. 3.16 to obtain the unfiltered counts representation shown by Fig. 3.17 (b).
Figure 3.17: $K_{\alpha}$ spectrum from an indium laser produced plasma with counts corrected for the Al filter

In the magnified area of Fig. 3.16 the measured energies of the $K_{\alpha}$ and $K_{\beta}$ transition series are identified. These measured values are compared with the published energy values of the transition [20] and presented in Table 4.

<table>
<thead>
<tr>
<th>Transition</th>
<th>Measured (keV)</th>
<th>Published (keV)</th>
<th>Residual (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$K_{\alpha 2}$</td>
<td>24.02</td>
<td>24.002</td>
<td>0.018</td>
</tr>
<tr>
<td>$K_{\alpha 1}$</td>
<td>24.232</td>
<td>24.21</td>
<td>0.022</td>
</tr>
<tr>
<td>$K_{\beta 3}$</td>
<td>27.227</td>
<td>27.238</td>
<td>0.011</td>
</tr>
<tr>
<td>$K_{\beta 2}$</td>
<td>not resolved</td>
<td>27.276</td>
<td>not resolved</td>
</tr>
<tr>
<td>$K_{\beta 1}$</td>
<td>27.876</td>
<td>27.86</td>
<td>0.016</td>
</tr>
</tbody>
</table>

Table 4: Table comparing the measured energy values of the X-ray K series for In to the published energy values.
Table 4 illustrates that differences between the measured and published values are of the order of the resolution of the instrument calculated in Section 3.9. The differences in the energies that are within the resolution of the spectrometer are likely deviations due to using only three radiation lines to calibrate the system.

The CR model was used to estimate the electron temperature of the plasma. The temperature was estimated to be $6.19 \times 10^5$ eV. The shape of the intensity profile $I$ of bremsstrahlung is related to the temperature and the to the energy according to equation 3.5 [21]:

$$I(E) = \exp \left( \frac{-E}{kT} \right)$$

Where

- $k$ - Boltzmann constant
- $T$ - electron temperature
- $E$ - photon energy

A fit of the equation 3.5 is demonstrated for the unfiltered X-ray spectrum shown in Fig. 3.17 and a number of different temperatures:
Figure 3.18: bremsstrahlung fit equation 3.5 for a series of different temperatures

The temperature calculated by the CR model is shown in the purple fit in Fig. 3.18. It can be seen that it does not fit the bremsstrahlung shape very well. The red and the blue plots represent a factor of 10 respective increases and decreases in temperature. The green represents half of the CR temperature $3.09 \times 10^5$ eV which has the best fit of the bremsstrahlung intensity profile. This suggests that the CR model overestimates the temperature of the plasma by at least a factor or 2, which further verifies that the CR model only works for steady-state 100 ps FWHM and longer laser pulses.

3.5.8 Conclusions and Future Work for Inner Shell X-Ray Emission

As mentioned in the introduction, this was a preliminary analysis of inner shell X-ray emission for In. An approximate calibration was successfully obtained for the energy range of the Si(Li) using only the strong radioactive $^{241}$Am, which consisted of the 13.95 keV, 17.75 keV, and 59.54 keV lines. This calibration provided a coarse linear calibration function which produced reasonable residuals.

The filtering characteristics of the detector were also investigated with the radioactive $^{241}$Am lines where the decrease in the counts on adding a filter was found to be consistent with the transmission of Al to a given of X-ray energy [19]. This study provided a means to correct the X-ray spectrum to
show the true counts of bremsstrahlung, which is normally suppressed by the Al filter, to become realistically scaled (as shown in Fig. 3.17).

The observation of $K_{\alpha}$ and the $K_{\beta}$ lines was successfully achieved and the calibration was adequate to observe evidence of all the K series lines which are within the resolution of the instrument. The Thor TW laser reliability limited the time available to fully explore the optimum filtering and find the best pinhole aperture size for the detector. In addition, too narrow a scan was conducted on the focusing conditions for optimum X-ray emission and so should be expanded and repeated a number of times to check the consistency. It should also be noted that at the time of the running of this experiment the FROG device for measuring the pulse duration of the laser was not operational and there is no certainty of what the pulse duration was in producing these X-rays. Additional investigative work on the pulse duration could be carried out to obtain the optimum pulse length for X-ray generation.

Finally, the literature reviewed refers to higher conversion efficiencies being obtained by ablating a rough target as opposed to a smooth target [22]. A survey of fresh target ablation vs used target ablation could yield an interesting comparison if this research was continued.

References


4 Ultrafast evolution of Transient Opacity in Borosilicate Glass Induced by Laser Driven Ions

4.1 Introduction

The aim of this chapter is to explain a novel detection technique which has been applied to detecting laser driven proton beams in a microscope slide of borosilicate (BK-7) glass. This experiment was conducted at The Centre for Plasma Physics, Queen’s University Belfast using the TARANIS laser system (explained in Section 2.7. The experiment was designed to better understand the behavior of protons in various materials specifically BK-7 glass (as introduced in Section 1.4.3) on picoseconds time scales.

This chapter outlines my involvement in a greater collaboration of scientists involved in this experiment namely the early experimental measurements of the transient opacity in BK-7 glass. The collaboration involves Queens University Belfast, Lawrence Livermore National Laboratory, Livermore, California, 94551, USA and University College Dublin, Ireland.

The experimental apparatus used for the different experiments is described in Section 4.2.2. An explanation of the use of radiochromic film (RCF), a dosimetry tool which calculates the energy of the laser produced proton beams impacting the glass, begins Section 4.2.3. The chapter then continues to describe the experimental setup and the results specific to two experiments, which provide different information about the proton interaction with the BK-7 glass. The two experiments give:

1. Spatially resolved snapshot images of the transient opacity induced by the protons interacting with the BK-7 glass all taken at different time delays with respect to the chirp pulse amplified (CPA) TARANIS laser system.

2. Single shot direct mapping of the temporal evolution of the transient opacity via the optical streaking technique.

The spatially resolved snapshot experiment begins in Section 4.3.2 by demonstrating the need for aluminium (Al) filtering to block emission, from the proton source target, which saturates the transient opacity imaging camera. Different thicknesses of Al are used for this purpose. The spatial calibration of the transient opacity imaging system is outlined in Section 4.3.3. With a calibration system, the images could then reveal the depth of penetration of the protons in the glass. The proton penetration depths were recorded as
a function of delay time between the main TARANIS pulse and the probe pulse. This was done to understand the depth of penetration as a function of time and the relaxation process of the glass-proton interaction.

An optical streaking technique was employed because of variations in the shot to shot variation in the TARANIS laser system’s energy. The optical streaking concept and set up are explained in Section 4.4.2. The optical streaking permits the transient opacity behavior to be observed within a 200 ps time window. Different optical streaking time windows are shown in Section 4.4.3 and consistent behavior of the protons was observed. The chapter closes with a conclusion which summarizes the results of the experiment.

4.2 Experimental Set Up

4.2.1 Introduction

In this Section is a description of the general set up common to both configurations of the ultrafast evolution of transient opacity in BK-7 glass induced by laser driven ions experiment. The emphasis will be on the geometric layout of the experiment which is designed to contextualize the set up for the Sections that follow.

4.2.2 Proton Beam Experimental Set Up

A schematic of the general set up of the target system used to accelerate proton beams is shown in Fig. 4.1

![Figure 4.1: Schematic of the ultrafast evolution of opacity general set up (Drawn by B. Drome)](Image)

The Chirped Pulse Amplified beam enters the chamber and is focused by a parabolic mirror onto the rear of the Au-foil target as shown by the red beam
on the left in 4.1). The power density was estimated to be $2 \times 10^{19}$ W cm$^{-2}$. The proton beams emerge from the rear of the Au foil target as illustrated by the green line labelled ions in Fig. 4.1. The interaction region of the protons and the BK-7 glass was transversely probed by a synchronized infrared probe (1053 nm, 400 fs, $\leq 10$ µJ cm$^{-2}$ which originated in the Micron Legend (see Section 2.7.2).

The probe output was then imaged to a CCD camera system allowing images of the transient opacity to be recorded and analyzed. The Au target and the BK-7 glass are connected as a single unit or set to a magnetic base, which is attached to an x/y/z computer controlled actuator, allowing movement of the pair under vacuum. Pictures of the Au target and the BK-7 unit (front view) and (side view) are shown in Fig. 4.2 and Fig. 4.3 respectively.

![Figure 4.2: Target holder (front view) for proton interaction studies](image)
The gold targets are racked in an aluminium frame as shown in Fig. 4.2 and provide the target area for the TARANIS main pulse. This view corresponds to the left facing side of Fig. 4.3. The L-shaped clamps labelled on the right of Fig. 4.3 are used to hold the glass slide (labelled on the top of Fig. 4.3) in position for the proton beam interaction. The L-shaped clamps are attached to a small stage which allows the glass to be moved forward and back (left and right in Fig. 4.3) to change the distance traversed by the protons. The glass slide has an Al mask with either a pin hole or a slit drilled into it on the target facing side (labelled Slit/ Pin Hole in Fig. 4.3). This was used to calibrate the images of the transient opacity (explained in Section 4.3.3). The magnetic base is labelled on the bottom of Fig. 4.3 and, as mentioned already, it allows the set to be moved in an x-y-z direction with computer controlled actuators. The system is generally moved in the up and down direction, (as viewed in Fig. 4.2) when aligning the TARANIS main pulse from one gold target to the next.

4.2.3 Proton Energy Measurement

In order to get an indication of the energy of the protons, radiochromic film (RCF) detectors were used [1]. The radiochromic effect involves the direct colouration of a material by the absorption of energetic radiation without the need for latent chemical, optical or thermal development. The RCF
changes colour due to the process of transverse solid state polymerization, which is a chemical process in which the ions induce a lengthening of the side (or transverse) carbon chains, which make up the material. The RCF is transparent prior to irradiation and turns blue in the irradiated areas. This opaquing process takes about $10^{-3}$ s and will remain thereafter. The polymerization will also occur if it is exposed to UV light. The energy of the ionizing radiation is then related to the absorbency of a narrow wavelength of light in the effected regions which can be quantified using equation 4.1:

$$A = \ln\left(\frac{I}{I_0}\right)$$ (4.1)

Where $I$, in equation 4.1, is the intensity of the light after passing through the irradiated RCF and $I_0$, is the intensity of the light without getting absorbed by the irradiated RCF. Since a proton requires 1 MeV of energy to penetrate the RCF film, a multi-frame temporal picture of the interaction can be observed where the depth the protons penetrated through the stack is proportional to the energy of the protons. The protons were found to have a maximum energy of approximately 12 MeV [3].

4.3 Spatially Resolved Images of the Transient Opacity Induced by the Protons Interacting with the BK-7 Glass

4.3.1 Introduction

The first of the two experimental studies of the ultrafast evolution of transient opacity in BK-7 Glass is examined in Section 4.3. The experiment begins with the demonstration and filtering of the proton beams described in Section 4.3.2. This concludes in Section 4.3.4 with an investigation of how the depth penetration of the protons in the glass changes as a function of probe time with respect to the TARANIS main pulse.

4.3.2 Demonstration and Filtering of Proton Beams

The initial proton experiments recorded spatially resolved snapshot images of the protons which involved observing their transient effects (introduced in Section 1.4.3) on the valence electrons of glass which is an insulator. The glass is placed on the target mount, as shown in 4.3. The glass is illuminated by the ultrafast probe beam as shown by the red arrowed line in 4.1. When the probe beam is fired on its own, the camera will image the glass and the mask covering it. Fig. 4.4 shows the first shot taken of a proton beam using a low resolution camera protected with a neutral density 3 filter. In this
experiment Fig. 4.4 the TARANIS main pulse and the probe beam arrived at the same time.

In Fig. 4.4 the camera is imaging the interaction of the protons coming from the left. The vertical line down the centre of the image in Fig. 4.4 is the edge of the glass slide. Slightly to the left of the vertical central line there is a bright region which is the plasma-emission from the TARANIS main pulse interacting with the foil. This plasma emission consists of X-rays, vacuum ultraviolet (VUV) light, visible and infrared which is emitted from the Au plasma forming the protons. To the right of the vertical line down the center there is darkening which is the region of the glass slide affected by the combination of the protons and the probe beam combination.

Steps needed to be taken to reduce the plasma-emission as it caused the camera to saturate. To minimize the saturating effects on the camera, Al filters were placed between the target and the glass slide. The filtering was done by offsetting a 100 µm sheet of Al foil on top of a 60 µm foil which were both on top of a 40 µm foil such that all three foil thicknesses were exposed. The result was a multilayer stacked filter which would permit the effects of each filter to be viewed at the same time. An optical delay was also employed between the probe beam and the TARANIS main pulse. The optical delay was comprised of two mirrors on a micrometre screw system for which a movement of 0.03 mm corresponded to a delay of 1 ps. The image of glass shown in Fig. 4.5 represents the photon interaction with the glass when only the probe beam is fired.

Figure 4.4: First image of Proton effects on a camera.
The black interface vertical outline, at the right, in Fig. 4.5 represents the glass/filter slide interface to the clamp on the aluminium frame and the black blobs that appear beside the interface is the glue used to secure it. The proton signature obtained when the probe beam was fired with a delay of $\Delta \tau = 200$ ps with respect to the TARANIS main pulse is demonstrated in Fig. 4.6. The impact of the multilayer stacked filter is evident in Fig. 4.6. The multilayer Al filter was set up in front of the slide to determine the optimum thickness to reduce emission from the plasma but still allow sufficient protons to interact with the glass.
There is a clear difference between Fig. 4.5 and Fig. 4.6. First the image position has displaced 0.41 mm in the vertical direction with respect to Fig. 4.5 from the firing of the main pulse. From the exposure in Fig. 4.6, the only part of the filter where the protons managed to penetrate was the 40 \( \mu \text{m} \) part of the filter which is located in the top right region. The proton source in the set up that Fig. 4.6 represents was 50 mm from the glass slide. There were 15 sheets of RCF placed 33 mm from the target in order to get an indication of the energy of the proton beams with the multilayer filter, located 4 mm from the target. The TARANIS main pulse energy was 7.583 J and the probe beam was delayed by 200 ps after the TARANIS main pulse. The image in Fig. 4.7 displays the effects of the probe beam on its own after the effects seen in Fig. 4.6 have disappeared.
The exposure in Fig. 4.7 demonstrates that the glass is not permanently altered by the proton interaction. The protons beam interacts with the valence electrons in the glass slide. After 200 ps the probe beam transmits through the glass as depicted by the exposure in Fig. 4.6.

In the regions where the protons have interacted with the glass, valence electrons gain energy. That energy, when combined with further energy from interaction of the probe beam at $\Delta \tau = 200$ ps, is sufficient for the valence electrons to cross the band gap into the conduction band, making the glass opaque to the probe light in these regions (as described in Section 1.4.3). The probe light has no effect on the regions of the glass not treated with the protons as the band gap of borosilicate glass ($\approx 4$ eV) [2] is of too high an energy for the probe beam to overcome alone. At some time later (evaluated later in this chapter), after the electrons have fallen back into their ground valence state, another probe beam is fired at the glass and it is apparent from the exposure in Fig. 4.7, that no electrons are promoted into the conduction band. In addition, there are no obvious residual effects to the glass from the excitation.
4.3.3 Depth Calibration of Proton Beams

In order to obtain spatial information as to the depth of penetration of the proton beams, a 1.5 mm thick Al mask was used with four 1.05 mm diameter drilled holes, separated by 1.45 mm centre to centre. The purpose of these holes was to be able to obtain a pixel to distance calibration. Each mask hole was covered by the 40 µm filter used in Section 4.3.2. The photon pattern displayed in Fig. 4.8 is a result of the probe beam for these conditions. The mask is illustrated by the darkened area on the right of the image of Fig. 4.8.

![Image](image.png)

*Figure 4.8: Probe beam only for Shot 1 with the Al mask*

Staying with the same configuration and conditions as described above, the probe beam was fired at a delay $\Delta \tau = 225$ ps after the TARANIS main pulse which had an energy of 7.67 J. The image in Fig. 4.9 displays the result.
The two dark regions, depicted in Fig. 4.9 represent the 1.05 mm pin hole gaps, filtered with the 40 µm filter where the protons can pass unaffected by the mask and interact with the glass.

To improve the contrast of the proton signature to the imperfections of the glass slide, the Beer Lambert equation (see equation 1.18) is applied to this data, where the counts with the probe beam only condition are expressed as $I_0$ and the counts with the probe beam delayed after the TARANIS main pulse configuration are expressed as $I$. The resulting plot is presented in Fig. 4.10.
The dark regions displayed in Fig. 4.10 clearly show the absorption of the probe beam in the glass. It was decided arbitrarily that a transmission of approximately 0.75 or lower be considered a proton signature as this was lower than the general oscillation of the imperfections in the glass. The transmission is obtained by exponentiating the element values of Fig. 4.10. The transmission of the average of a 38 vertical cross sectional slice of 4.10 is plotted in Fig. 4.11. All 38 vertical cross sections lie in the black absorbed region.
The x axis in Fig. 4.11 is a clockwise 90° rotation of the y axis in Fig. 4.10. This rotation allows the reduction in transmission between pixel 58 and 499 of Fig. 4.11 to be interpreted as the lower proton signature of Fig. 4.10, which is labelled by the left red arrow in Fig. 4.11. This gives the diameter of the lower proton signature as 441 pixels. Using the same process, the reduction in transmission from pixel 666 to the very right pixel is the upper proton signature in the image Fig. 4.10.

The mid-point of the lower region in the plot Fig. 4.11 is pixel 279. The upper absorption signature is cut off at pixel 1024 but as pin holes are of equal diameter and the radius of the lower proton signature is 221 pixels then the mid-point can be estimated to be at pixel 887. This is assuming the absorption of the upper proton signature begins at pixel 666. As stated earlier, the centre to centre distance between pin holes is 1.45 mm and therefore the difference between the upper proton signature and the lower proton signature mid-points should correspond. From this:

\[
608 \text{ pixels} = 1.45 \text{ mm}
\]

\[
1 \text{ pixel} = 0.00238 \text{ mm} = 2.38 \mu \text{m}
\]
This value can be checked by recalling that the lower proton signature has a diameter of 441 pixels which when converted into mm results in a diameter of:

\[ 441 \text{ pixels} = 1.05 \text{ mm} \]

\[ 1 \text{ pixel} = 0.00238 \text{ mm} = 2.38 \mu\text{m} \]

This pair of measurements shows very good correlation between pin hole diameter and the separation between pin holes. This scaling can now be applied to a horizontal cross section in the image Fig. 4.10 allowing the calculation of the depth of penetration of the protons into the glass if we assume that the protons have only reached depths where the glass is opaque to the probe beam.

The plot in Fig. 4.12 illustrates the horizontal cross section of the lower proton signature presented in Fig. 4.10.

*Figure 4.12: Horizontal cross section of the lower proton signature of Shot 1*

From the plot in Fig. 4.12 it is evident that there is a steep drop in transmission which begins where the mask ends and the transient opacity begins.
The transmission continues to drop until the maximum opacity of the proton signature is reached. After the maximum opacity (114 µm) transmission increases to over 0.556 at 673 µm.

The target stage was moved vertically and a second shot was taken with the same conditions as above, including the Al mask, except with a $\Delta\tau = 250$ ps delay between the probe beam and the TARANIS main pulse. The absorbance, is presented in Fig. 4.13. The laser energy for this shot was 7.31 J.

![Figure 4.13: Transmission plot of Shot 2 with the mask](image)

The proton opacity region is located in the vertical centre region of Fig. 4.13. The glass also has more imperfection regions leading to a more noisy transmission signal than that taken in shot 1 (Fig. 4.10). For that reason, an average transmission of 53 vertical pixel cross-sections of the opaque region of Fig. 4.13 was used to plot Fig. 4.14.
Figure 4.14: Vertical cross section of Shot 2

The red arrows illustrating pixel numbers in Fig. 4.14 depict the extremities of the 1.05 mm pin hole, namely pixel 269 and pixel 721. The red arrow above the dip in transmission illustrates a width of 452 pixels corresponding to the 1.05 mm pin hole diameter. When we apply the same calibration of pixel to distance, as carried out in shot 1 with the plot in Fig. 4.11 the scale becomes:

1 pixel = 2.32 µm

Just as with shot 1, the depth of penetration of the protons can be obtained, assuming that the protons have only reached depths which are opaque to the probe beam. The plot in Fig. 4.15 illustrates the corresponding average transmission of 49 horizontal cross-sections of the opaque region presented in Fig. 4.13.
The right side of the glass surface, from the plot in Fig. 4.15 illustrates an interpretation of the proton interaction region. The absorption is described with a sharp drop in transmission which is discussed in Shot 1. The transmission then increases quadratically as a function of glass depth until pixel 693 \(\mu m\) where the transmission increases linearly. The quadratic region is assumed to be the proton interaction region following the known Bragg shape of proton interactions (see Section 1.4.1). This is what defines the 693 \(\mu m\) distance describing opacity from the protons.

4.3.4 Proton Scans at Different Time Delays

It is evident from the measured data used to generate the plots Fig. 4.11 and Fig. 4.14 that the pixel calibration varied very little as the target mount was moved vertically to shoot all the gold targets on the multi-shot rack (shown in Fig. 4.2). The shot to shot variation of the TARANIS laser output and, as a consequence, the proton flux system made it difficult to compare one
shot to another. The laser shots using the pin hole mask that were produced by similar energy are grouped together in Fig. 4.16.

![Graph showing proton penetration depths vs delays for ≈7 J shots](image)

*Figure 4.16: Proton penetration depths vs delays for ≈7 J shots*

From the plot in Fig. 4.16, there is a definite trend from $\Delta \tau = 200\ ps$ of increasing proton penetration with delay time. There was only one shot below $\Delta \tau = 200\ ps$ and even though there was no significant difference in the energy of the laser, the penetration depth does not fit the trend. This pattern could not be extended to other delay times due to a high fluctuation in energy from the TARANIS laser output.
4.4 Single Shot Direct Mapping of the Temporal Evolution of the Transient Opacity via the Optical Streaking Technique

4.4.1 Introduction

To overcome the problems of shot to shot variation in energy (discussed in Section 4.3.4) a decision was taken to alter the experiment to observe the transient opacity on a single shot basis. This was achieved through a single shot mapping of the temporal evolution via the process of optical streaking [4], [5]. The concept and experimental implementation of optical streaking are explained in Section 4.4.2 and measured data are demonstrated along with a number of optical streaked images in Section 4.4.3.

4.4.2 Optical Streaking

Optical streaking involves stretching the existing probe beam to approximately 200 ps with a linear frequency sweep and then passing the probe beam through the part of the glass which the protons have interacted with. This means that the probe beam would take approximately 200 ps to pass through the glass and the protons would begin to interact during this probe traversing period. The different wavelengths in the linearly chirped probe beam permit direct mapping of the temporal evolution of the transient opacity onto the spectrum of the probe pulse. The probe beam emerging from the glass is then passed through a spectrometer, which disperses the different frequency components spatially onto the pixels of a CCD camera. The spectrometer layout is shown in Fig. 4.17.

140
The stretched beam passes through the glass target and enters the spectrometer, illustrated in Fig. 4.17, through the entry slit. The beam reflects off mirror 1, and diffracts off the diffraction grating. These diffracted wavelengths have the evolution of the transient opacity encoded within them. These different wavelengths then reflect off mirror 2, where they arrive at the exit aperture where a camera is mounted. The full range of wavelengths corresponds to the temporal range of the stretched pulse. The longer wavelengths correspond to earlier times in the transient opacity evolution and will arrive earlier than the shorter wavelengths. The purpose is to be able to detect the protons arriving in the time it takes for the shorter wavelength radiation to arrive.

The probe beam was stretched to 200 ps and a spectral bandwidth of 10 nm. The spectrometer was calibrated by aligning the centre wavelength (1053 nm) onto the centre of the camera. This defined the centre time of the probing time window which could then be altered by changing the optical delay (discussed in Section 4.2.2). The full 10 nm spectrum dispersed on the CCD camera from the output aperture of the spectrometer is exhibited in Fig. 4.18.
The horizontal axis of Fig. 4.18 represents the flow of time between -25 ns and +175 ps relative to when the TARANIS main pulse would have arrived. The vertical axis of Fig. 4.18 shows a space axis where the sharp cut off of light represents a slit through which the protons can enter the glass (just like the pin hole mask discussed in Section 4.3.3). The spectrum shown in Fig. 4.18 is calibrated and the result is depicted in Fig. 4.19.
Given that the spectral output of the probe beam is contained in the 200 ps temporal output, the spectrum shown in Fig. 4.19 can be expressed as a time axis as shown in Fig. 4.20.
From the time delays shown in Fig. 4.20 any time delay can be obtained by increasing the optical delay discussed in Section 4.2.2.

4.4.3 Optical Streaked Images of Proton Beams

The next step involved observing the transient opacity seen in Section 4.3.3 and Section 4.3.4 using the optical streaking method. The first evidence of proton transient opacity was observed in a time window between -25 ps before and +175 ps after the TARANIS main pulse. The probe beam alone through the glass target is shown in Fig. 4.18. The probe beam is then fired and arrived at the glass slide 25 ps before TARANIS was fired. Due to the temporal pulse duration of the probe beam, the probe beam does not completely clear the glass slide until 175 ps after the TARANIS main pulse and the optical streaked output is demonstrated in Fig. 4.21. The energy of the TARANIS main pulse was 8.883 J.
Figure 4.21: Probe optical streaking of glass spanning the time window of 25 ps before the TARANIS main pulse and 175 ps after the TARANIS main pulse.

There is a visible opacity on the right hand side of Fig. 4.21 where the transient opacity is evident. The contrast can be enhanced by applying the Beer Lambert equation (see equation 1.18) between the matrix elements that make up Fig. 4.21 and that of Fig. 4.18 (as discussed in Section 4.3.3). The resulting plot is shown in Fig. 4.22.
Figure 4.22: Absorbance equation applied to the probe and the TARANIS main pulse

The start of the proton signal is quite clear in absorbance as shown in Fig. 4.22. A pixel to time calibration shown in the plot of Fig. 4.20 is applied to the matrix element data making up Fig. 4.22 to obtain a delay. A strip of 20 horizontal cross sections were averaged at the slit region, highlighted in Fig. 4.18 from the data in 4.22. The averaged absorbance measurements from Fig. 4.22 where exponentiated to obtain the transmission as a function of probe delay as shown in Fig. 4.23.
The proton interaction time is very clear in Fig. 4.23. It is observed by a steep 70% dip in transmission starting at 62 ps after the TARANIS main pulse, which only begins to recover at 137 ps after the TARANIS main pulse. This 70% dip in transmission is the same proton signature observed in Section 4.3.3.

The consistency of this experiment was tested by choosing a slightly different time window and testing whether the proton opacity occurs around the same time. The optical delay was moved by 30 ps to cover the range of 5 ps after the arrival of the TARANIS main pulse to 205 ps after the TARANIS main pulse and the same procedure used to arrive at Fig. 4.23 was repeated as shown in Fig. 4.24. The TARANIS main pulse had 9 J of energy.
Figure 4.24: Transmission vs calibrated time axis of 5 ps to 205 ps after the TARANIS main pulse window

There are quite a few similarities between the transmission variation shown in Fig. 4.24 and that shown in 4.23. The proton beam begins to reduce the transmission of the probe beam at approximately 50 ps after the firing of the TARANIS main pulse.

None of the time windows scanned were sufficient to see how long it takes for the transient opacity to disappear. All that can be surmised is that the transmission seems to increase gradually as the energy imparted to the valance electrons dissipates.
4.5 Conclusions

In this experiment protons were successfully accelerated from foil targets using the main pulse of the TARANIS laser system in Queens University, Belfast. The protons traversed a 5 mm gap and interacted with the valance electrons of the glass in a BK-7 microscope slide. The interaction region was observed with an ultrafast probe beam which transversely probed the glass at different time delays (176, 200, 225 and 250 ps) with respect to the firing of the TARANIS main pulse and imaged a transient opacity in the regions where the protons had interacted with the glass onto a CCD camera.

The proton beams passed through a pin hole mask with holes of a known diameter and separation drilled in it. The probe beams interacted with the glass only in the regions where the holes allowed the protons into the glass as demonstrated in Fig. 4.9. The imaging allowed for a spatial calibration of:

\[ 1 \text{ pixel} \approx 2.3 \mu m \] (4.2)

With this calibration, the depth penetration of the proton beams with different time delays could be observed. There was evidence of increased penetration depth as a function of time delay (as illustrated by Fig. 4.16). The investigation of this trend was limited by the TARANIS laser system’s stability which sometimes fluctuated by up to 1.5 J between shots. This level of instability changes the energy imparted to the protons on a shot to shot basis meaning that the delay the probe beam arrived was not the only variable that needed to be considered.

This problem was solved by altering the set up to one that involved optical streaking. Optical streaking involves stretching in the probe pulse by changing the spacing between the gratings in the probe compressor while preserving an almost linear wavelength sweep through the pulse. When this temporally chirped probe pulse is used to probe the glass opacity it will encode the opacity information into the spectrum of the pulse. The information is then dispersed by the diffraction grating of a spectrometer on to the pixels of a CCD camera. The resulting CCD images have spatial information on the vertical axis and temporal information on the horizontal axis.

Since the set up was changed, the 2.3 \( \mu m \) per pixel calibration was assumed to no longer be valid as there was uncertainty on whether the imaging lens had been moved during the set up of the optical streaking. Since no other spatial calibration was taken, the attention focus of the experiment was directed towards the temporal information encoded into the horizontal component of the optically streaked images.

The two calibrated optically streaked images shown in Fig. 4.23 and Fig. 4.24 demonstrate the time window -25 ps to +175 ps with respect to
the TARANIS main pulse and window +5 ps to 205 ps with respect to the TARANIS main pulse respectively. There is a strong consistency of 80 ps in the onset time of the transient opacity in both optical streaks.

This suggests that 80 ps is approximately the transit time of the fastest protons. If one assumes a 5 mm gap between the target and the glass, and an 80 ps transit time, the energy of the protons are calculated to be 20.4 MeV which is of the order of energies that are detected by the RCF (discussed in Section 4.2.3).

This work has set the framework as a proof of principle experiment of the absorption of protons in different materials. The obvious extension of this experiment is to repeat it in different materials, such as water phantoms to understand the transient absorption of protons in hadron therapy, for example.

References


5 Refurbishment of the 1-m Normal Incidence UV Spectrometer

5.1 Introduction

The 1-m Normal incidence spectrometer is an existing spectrometer in the UCD Speclab Group with an effective spectral sensitivity in the 200-2000 Å (62-6 eV) spectral range. The spectrometer, originally built in house in UCD and, was intended for use as a quick survey instrument to study outer shell photoabsorption of ionized 5th row elements [1] and was later used to study 4d → 5p photoabsorption of In⁺ [2]. The spectrometer works under vacuum (approx. 1 × 10⁻⁴ mbar) due to the complete absorption of UV radiation by the oxygen molecule between ≈ 185 nm and 150 nm. The spectrometer vessel was built in a conical shape to keep the volume of air to a minimum and reduce pump down time. At either end of the cone is a hemi-spherical dome one is 32 cm in diameter by 5 cm high and the other is 60 cm in diameter and 10 cm high. The spectrometer is linked to the rectangular target chamber by an optical axis channel. The channel consists of a ball valve to allow the targets to be changed without the need to break the vacuum in the spectrometer. The target chamber optical axis port consisted of a 15 µm slit with additional side holes to allow faster evacuation of the rectangular chamber’s 16.5×15×15 cm volume.

The diffraction grating of the system has 1200 grooves per mm. The platinum coated grating has a blaze angle (angle of maximum reflection) of 5.5 degrees resulting in a blaze angle of 77.75° and a 1-m curvature radius. Platinum has a work function of 6.35 eV which makes it unaffected by incoming radiation down to 195 nm. The original detector for the system was a photographic plate which worked by imaging a 15µm slit onto the concave grating, and obtaining a 1:1 image onto the photographic plates. The 15µm slit was maintained as it allowed the resolution of the spectrometer to achieve the theoretical maximum of the grating.

A schematic of the light path inside the the 1-m spectrometer is illustrated in Fig. 5.1.
5.2 The 1-m Normal Incidence Spectrometer Detector

The use of photographic plates has declined significantly over the years due to availability and a high processing costs. For this reason the 1-m normal incidence VUV spectrometer detector was upgraded to a CCD. Due to budgetary and space limitations, the utilization of an optical detector alongside a phosphor wavelength shifter to view UV radiation was preferred over UV back thinned silicon devices. The phosphor wavelength shifter absorbs the UV radiation and then after a non-radiative relaxation process, re-emits the radiation as an optical wavelength visible to the detector. The Sony ILX511, 2048 pixel CCD linear image sensor combined with the Ames Photonics Larry series linear CCD array controller were found to be well suited as a cost effective optical detector. The Sony ILX511 image sensor is a $1 \times 2048$ pixel array with a pixel size of 14 by 100 $\mu$m with a pitch of 14 $\mu$m. The sensitivity of the Sony ILX511 CCD linear image sensor is presented in Fig. 5.2. A sample detector along with p43 phosphor and an Ames Photonics Larry controller was generously donated by Prof Larissa Juschkin.
From information provided by the sensitivity curve in Fig. 5.2 the best phosphors are those that emit between 410 nm and 535 nm. Three phosphors were chosen to this effect.

1. Sodium salicylate
2. P-terphenyl
3. P43 (Gd$_2$O$_2$S:Tb)

5.2.1 Phosphor Evaluation

Sodium salicylate is known to have an emission spectra peaking at around 416 nm and a conversion efficiency of approximately 65% [3]. This lies well within the ILX511’s image sensor’s 85-90% sensitivity region. The first test of sodium salicylate was made by spraying a solution of sodium salicylate dissolved in methanol onto a 1 mm thick preheated glass slide. The glass slide was preheated to 80-100 °C to boil off the methanol leaving a uniform film on the slide. The phosphorescence from a deuterium lamp was detected by an uncoated Sony ILX511 linear array sensor with the sodium salicylate treated glass slide covering it. The 15 µm slit was removed from the optical axis to maximize the light getting to the diffraction grating. The response
of a single sprayed and evaporated coating of sodium salicylate is illustrated in Fig. 5.3.

![Graph of sodium salicylate phosphorescence](image-url)

**Figure 5.3:** Sodium salicylate phosphorescence following a 500 ms exposure to the deuterium source with no slit.

It has been shown that p-terphenyl had a higher conversion efficiency than sodium salicylate[4], this is illustrated in Fig. 5.4.

![Comparison of fluorescent spectra](image-url)

**Figure 5.4:** Comparison of fluorescent spectra of sodium salicylate and p-terphenyl [4]

A layer of p-terphenyl was vaporized onto a quartz screen. A quartz
material was required because of the absorption characteristics of BK-7 glass below 390 nm. A vaporization technique was employed due to the difficulty of finding a solvent to dissolve p-terphenyl. There were two coatings of p-terphenyl applied onto the quartz screen because a single coating appeared to have no visible phosphorescence. The measured phosphorescence observed from the two thin layers of p-terphenyl vaporized onto a 1.5 mm piece of quartz screen is illustrated in Fig. 5.5.

Figure 5.5: Phosphorescence of p-terphenyl to the deuterium source with no slit.

Even with the higher conversion efficiency of p-terphenyl, indicated in Fig. 5.4, the data from measurements represented in Fig. 5.5 and Fig. 5.3 clearly demonstrate that a higher proportion of the incoming photons was detectable when a sodium salicylate phosphor was used. One possible reason for this is that the thickness of the sodium salicylate was closer to the optimum thickness that balances conversion efficiency with self-absorption [4]. Additional layers of p-terphenyl were added to the already coated quartz screen and the effects of increasing the thickness of the p-terphenyl is illustrated in Fig. 5.6.
The data making up the plots in Fig. 5.6 demonstrates the effect of thickness of the phosphor on the number of counts observed. The results were obtained without any significant refinement of the focusing or height adjustment of the detector with respect to the optical axis of the grating. A difficulty with the p-terphenyl is that the vacuum evaporation is a long process, requiring over an hour and a half per layer. In addition, limitations of the evaporation system available provided only a limited control of the homogeneity of the coating, evident from layer 16 and beyond in Fig. 5.6. Also, there was an inherent risk of damaging the detector with frequent removal and replacement of the Sony ILX511 sensor from the controller (two Sony ILX511 were damaged during this data collection process).

The third phosphor option was to use a sample of P43, which has a chemical composition Gd$_2$O$_2$S:Tb and a peak emission in the green at 543 nm. The Sony ILX511 linear sensor array is still quite sensitive at this wavelength according to the information in Fig. 5.2. There was no P43 powder available to carry out an appropriate investigation to experiment with coating techniques or thicknesses, however, a P43 commercial sample which had been bought for other purposes was obtained for testing. There was not sufficient P43 to cover the whole Sony ILX511 sensor so only half of the sensor window was covered and the phosphorescent response of P43 is evident on the right side (from about pixel 1000) of the plot in Fig. 5.7.
The left hand side of the plot in fig 5.7, below the 1000 pixel mark, has no P43 phosphor and the bump at the 250 pixel mark represents the weak sensitivity the detector has to these (below 200 nm) wavelengths.

On the right hand side of the 1000 pixel mark in Fig. 5.7, the P43 phosphor is evident but it did not seem particularly responsive in the longer wavelengths.

5.2.2 Laser Generated Plasma Spectra

Since recording the spectrum of a laser produced plasma is the ultimate application of the phosphor it was decided to use the 1-m normal incidence UV spectrometer to observe a laser produced plasma. To do this, the same feed-through collar described in 3.8 was mounted on the spectrometer. The Al target was placed at the center of the collar and the target was aligned to graze the optical axis of the spectrometer. The photographic plate holder was upgraded to a detector system. A stepper motor complete with chassis, gearbox belt, and pulleys was constructed by the School of Physics Mechanical Workshop. The system was optimized to reduce friction and increase the torque so that the detector could be navigated freely from one side of the Rowland circle to the other. The stepper motor was controlled by a USB Stepper B controller. The adapted photographic plate holder with the stepper motor and pulley system coupled through a 10:1 gear case is depicted in Fig. 5.8
A single layer of sodium salicylate was evaporated onto a 1.5 mm thick glass slide and secured in a mounting approximately 1 mm in front of the detector. The 15 micron slit at the entrance to the spectrometer was removed and replaced with a 1 mm slit to maximize the light entering the spectrometer. Three scans were taken at different spectral positions with laser produced plasmas and then joined together using the Origin Lab software package. The first spectrum recorded from an aluminium laser plasma is presented in Fig. 5.9.
The spectrum recorded from an aluminium laser plasma in Fig. 5.9 shows a series of very broad lines. The broadness of the spectrum is due to the large 1 mm width of the entrance slit, the fact that the phosphor is approximately 2 mm from the detector array and the 1 meter distance from the Rowland circle to the grating had not been optimized so the spectrum is not focused. These spherical wave emissions over the distance between the phosphor and the Sony ILX511 sensor array, expand to the point where lines spread into the neighbouring pixels, resulting in a further broadening of the lines. The experiment was repeated over the full range of the spectrometer and the spectrum is presented in Fig. 5.10.

![Figure 5.10: First complete spectrum of Al laser plasma, using the camera with the sodium Salicylate on the microscope slide and a 1 mm slit](image)

The target used to produce this spectrum was approximately 40 cm away from the slit, which resulted in a poor solid angle of light getting into the system. The feed-through collar was then replaced with the original rectangular chamber described in Section 5.1 which improved the solid angle and compensated somewhat for the grating’s poor reflectivity at short wavelengths. The detector was then optimized to the proper height.

5.2.3 Comparison of Phosphors

In order to improve the number of counts, the next step was to build on the results obtained for Fig. 5.9 and to apply the phosphors directly to the detector sensor. There were three options available, to coat the array detector with sodium salicylate, use the donated detector with the P43 phosphor already applied, or to coat a detector with p-terphenyl. Due to the long and
unreliable phosphor preparation required, p-terphenyl was rejected. A semi-log comparison of sodium salicylate, P43 and no phosphor using the 253 nm Hg II line from a calibration Hg lamp is illustrated in Fig. 5.11. For the comparison a 150 µm slit was created by obstructing the 1 mm slit with two commercial razor blades.

![Figure 5.11: Comparison of P43, sodium Salicylate and no phosphor](image)

It is clear from Fig. 5.11 that P43 has almost no phosphorescent effect, when compared to the “no phosphor” trace. The reason is the Hg II line has insufficient energy to induce phosphorescence. The obvious effect of P43 is the induced scattering of the light that would have reached the detector without the P43 coating. Sodium salicylate is clearly the best choice of phosphor in this wavelength region, which makes it a logical choice overall even though there is broadening of the emission that may impact on resolution. The following summarizes the reasons for choosing sodium salicylate as a phosphor for the detector:

1. Sodium salicylate has strong phosphorescence through the whole spectral range of the spectrometer.

2. It can be easily applied (see Section 5.2.4) onto the detector meaning that if there is a sensor failure there will be minimal time lost fitting the replacement.

3. The phosphorescent wavelength lies in the peak of the sensitivity curve of the Sony ILX511 linear sensor array.
5.2.4 Sodium Salicylate Deposition onto the Array

Since sodium salicylate had demonstrated good conversion efficiency (see Fig. 5.11) an equally workable method for coating the phosphor directly onto the sensor array was required. The procedure used to prepare the slide, in the sodium salicylate evaluation (Section 5.2.1), would be unsuitable given the heating process employed and the sensor specification (max storage temperature: -30 to +80°C). A coating method for sodium salicylate was reported by Knapp [5] who used a nebulizer to spray very fine misty droplets onto a desired surface [5] and taking this discriminative stimulus an ultrasonic wave nebulizer was purchased. This device uses an oscillator to generate mechanical vibrations in a piezoelectric element [6]. This piezoelectric element supports the reservoir that contains approximately 30ml of the methanol/sodium salicylate solution, which in turn is also made to oscillate at the same frequency. The oscillation forms cavities on the solution/air boundary, which evolve into capillary waves or ripples, from which fine mist droplets, of the order of 0.001 - 0.005 mm in diameter, (known as aerosols) are formed.

A scalpel was used to remove the Sony ILX511 sensor array window to allow the phosphor to be applied directly to the array pixels removing the spectral broadening introduced by the gap between the protective glass and the pixel array. The mist containing methanol/p-terphenyl salicylate solution is sprayed repeatedly back and forth onto the pixel array. The air flow rate in the fume cupboard was too high and had the effect of sucking the mist away from the sensor before it had a chance to settle on the surface so the process was moved to just outside of the fume cupboard entrance. The mist is sprayed back and forth for about 5-7 minutes, until the solution in the nebulizer reservoir has been consumed. The sensor array is then placed inside the fume cupboard for a few minutes to allow the methanol to evaporate and the sodium salicylate time to crystallize on the surface. Once this process is finished, which can be recognized by the lack of damp reflections on the sensor, the array should be inspected for regions that do not have a uniform coating. These regions can be recognized where the reflectance of the silicon array surface is still high. The process of using the nebulizer is then repeated, concentrating on regions of uneven coating. This process should be repeated until there appears to be an even uniform coating across the array.

5.3 Focusing the Spectrometer

The spectrometer setup was originally focused for the photographic plates and the pulley system (discussed in Section 5.2.1) was set up so that the
detector would be approximately positioned at the film plane where the plate was located. The arc segment, on which the photographic plate/detector was positioned was located on the stage by a pivot post that permitted limited rotation about this point. Assuming that the location of the pivot point was correctly at the zero order, the focusing of the system could be fine-tuned by rotating the long wavelength end of the arc segment in and out with respect to that pivot point. With the output, it was found that further improvement could be obtained by adjusting the position of the grating. The improvement obtained from altering the angle of the detection arc with respect to the zero order pivot point is represented by Fig. 5.12.

![Graph](image)

Figure 5.12: Focusing of the detector arc using the Hg II 253 nm line

It can be seen from Fig. 5.12 that improving the focusing not only increased the peak intensity of the line but also has the effect if improving the resolution. It should be noted that the maximized output was obtained with a maximum focus setting and that a better performance may have been achievable had more space been available in the chamber.

5.4 The 1-m Normal Incidence Spectrometer Chamber

All the early work on the phosphor optimization, using laser produced plasmas (see Section 5.2.1), was done using a large (52 cm diameter) cylindrical feed through collar chamber coupled with a custom made bellows to the 1-m normal incidence UV spectrometer. The large distance from the target (centre of the chamber) to the slit, which was located at the radius plus 18cm of vacuum coupling adapters away, led to a very poor solid angle of light getting
into the spectrometer. The result (depicted in Fig. 5.10) demonstrates reasonable signal in the longer wavelength part of the spectrum (region where the grating is blazed for) followed by a complete loss of signal, from about halfway across, as the grating becomes too inefficient to reflect the low light levels incident from the slit at the small solid angle.

For this reason, it was decided to change the chamber to the rectangular box chamber and introduce a few more mechanization features as outlined in Section 5.2.2. The mechanical feed-through screw type linear positioner was kept and used as a linear height adjuster for the absorber target used for photo absorption. The objective is to be able to perform spatially resolved spectroscopy of the absorbing plasma by controlling the part of the plasma plume that the continuum emission passes through. A DC motor was attached to the feed-through screw type linear positioner and used to rotate a disk with the target attached to it. The absorbing target for the indium photo absorption experiment (to be discussed in Section 6) used an aluminium ring, which had indium compressed into it. The photograph shown in in Fig. 5.13 demonstrates an example of the aluminium/indium disk used as a target for photoabsorption studies.

![Figure 5.13: Illustration of the Aluminium/Indium disk absorption target](image)

The aluminium ring was required for calibration purposes (see Section 5.5) and the lens could then be easily shifted to shoot the inner part of the disk (the indium). The continuum emission for the spectrometer is W (tungsten) or Sm (samarium) depending on the region of interest. Sm serves as a good continuum emission from 200 nm down to about 65 nm and W is useful below 65 nm and improves beyond the spectral region of the spectrometer. Both elements were obtained in the form of 7.5 mm rods and were fitted on to a Zaber stepper motor as required.
The continuum emission source was positioned to be as close to the absorber and the slit as possible, in order to maximize the signal entering the spectrometer. This layout configuration had the laser focal point between 20 and 30 mm from the slit which was a considerable improvement over that with the feed through collar chamber where the laser focal point was on the order of 430 and 450 mm away.

In order to assist with the alignment of the emission of the laser produced plasma to the optical axis of the spectrometer, an optical camera was mounted on the outside of the chamber looking in through the front with a magnifying lens. With the camera, the overlap of the lasers along the optical axis can be more easily and accurately observed and finely adjusted with a resolution of within 10's of microns. An optical camera views is illustrated in Fig. 5.15 A front view of the target chamber, without the magnifying lens, displays the absorption target and the continuum source target with the slit in the background, in Fig. 5.15 A magnified image of the slit with the continuum source target in the foreground is viewed in 5.16.
Figure 5.15: Front view of the fitted 1-m normal incidence spectrometer chamber
5.5 Calibration of 1-m Normal Incidence Spectrometer

Since the vacuum ultraviolet (VUV) spectrometer is to be used in the detection region that includes radiation from 200 nm to 25 nm, the first step is to establish a wavelength calibration. A good reference point is to observe the absorption of UV radiation in molecular oxygen (The Schumann-Runge bands), which are in the 192 nm and 176 nm region. The absorption of UV radiation in molecular oxygen was measured using the deuterium lamp and is illustrated in Fig. 5.17.
Figure 5.17: Schumann-Runge absorption of deuterium continuum over the approximate range of 192 nm to 187 nm

Since the detector pulley system is not absolute and does not give reproducible position along the spectrum (due to backlash in the pulley belt when changing direction and the detector occasionally catching when traversing the arc) it was decided that a master calibration spectrum should be taken. This calibration spectrum would always be obtained prior to that of the material of interest during an experiment and compared to the master spectrum and used to identify the wavelength position of the detector. Aluminium was chosen to be the calibration species.

With aluminium chosen as the calibration source the aluminium target was shot in the same wavelength range as the Schumann-Runge bands presented in Fig. 5.17. The lines surrounding the absorption band were used to identify the surrounding lines from the known lines of the National Institute of Standards and Technology (NIST) database. The absorption of the aluminium emission lines due to the Schumann-Runge bands are illustrated in Fig. 5.18.
Using the NIST database it was possible to identify all the nearby lines and begin the process of calibration of the spectrometer. In Fig. 5.18 it is clear that there is only one strong line before the Schumann-Runge absorption bands and this was Al I at 193.645 nm. Similarly, further left of the 193.645 nm line (not shown in Fig. 5.18) there was another line at 199.053 nm. The next step was to shoot the same region under vacuum and to calibrate the line that is beginning to form at pixel 1125 in Fig. 5.18. A plot of the same region under vacuum is shown in Fig. 5.19.

A challenge with calibrating the system is that the system uses a flat
detector which detects radiation from a circular focus. As a consequence, lines even within one frame are seen to shift in such a way that prevents spectra from different detection positions viewing the same lines from being able to successfully overlap. This shifting effect is illustrated in Fig. 5.20.

![Figure 5.20:](image)

**Figure 5.20:** (a) Four Al lines viewed from three different detector positions illustrated by the colours red, green and blue and brought to overlap well on the middle Al I 193.645 nm line (b) a zoomed in view of the Al II 199.053 nm line under the conditions of Fig. 5.20 (c) a zoomed in view of the Al II 193.645 nm line under the conditions of Fig. 5.20 (d) a zoomed in view of the Al III 186.279 nm line and the Al III 185.471 nm line under the conditions of Fig. 5.20 (a).

In Fig. 5.20 three different detector positions are viewing the same three lines (all shown in Fig. 5.20 (a)). The lines from the three detector positions are overlapped so that when zoomed in, the 193.645 nm line is perfectly lined up in all three detector positions (shown in Fig. 5.20 (c)). It is evident from inspecting the other lines on both sides of the 193.645 nm line, that the surrounding lines can not be made to overlap properly in the three positions (shown in Fig. 5.20 (b) and (d)). The above overlapping makes it evident, that simply joining all the different detector positions together and identifying a set of lines, is not going to work.

In order to calibrate this spectrum a function is calculated which linearly relates the pixel number to the angle at which the radiation is diffracted through. The pixel number can be related to the angle because it has a linear relationship. It can be seen that the relationship between pixel number and wavelength is non linear from the grating equation.
\[ m \lambda = d ( \sin \alpha + \sin \beta ) \] (5.1)

Where:

- \( m \) - integer representing the order of diffraction
- \( \lambda \) - wavelength of the radiation being diffracted
- \( d \) - groove spacing
- \( \alpha \) - incidence angle
- \( \beta \) - diffracted angle.

The \( \alpha \) is the incidence angle with respect to the grating normal which by convention is a negative number.

The first step in creating the calibration function that relates the pixel number to the diffracted angle, is to calculate the incident angle \( \alpha \) of the spectrometer. This angle was measured geometrically using an alignment laser to be \( \alpha = 7.75 \) degrees. It is possible to convert the published wavelengths into a diffracted angle \( \beta \), using equation 5.1 which would have been observed had the wavelength been measured on the 1-m normal incidence spectrometer Fig. 5.19.

The wavelength information from Fig. 5.19 is used in equation 5.1 to calculate the diffracted angle \( \beta \) (NIST angle) and the result is displayed in Table 5.

Figure 5.21: (a) Plot of NIST Angle vs Pixel for the 5 long wavelength lines shown in Table 5
Table 5: Direct angle $\beta$ (NIST Angle) for individual Pixels in Fig. 5.21

In Table 5 the pixel column represents the pixel number taken to be the centre of a line, and the NIST wavelength column is the closest corresponding observed wavelength from the NIST database for that pixel. The NIST angle column is the angle $\beta$ that the corresponding NIST wavelength value is diffracted at in equation 5.1. The plot depicted in Fig. 5.21 demonstrates the linear function of the NIST angle $\beta$ as a function of pixel. The plot in Fig. 5.21 (a) can be described by equation 5.2.

$$y = -8.06461 \times 10^{-4}x + 22.69311 \quad (5.2)$$

The lines displayed in Fig. 5.19 are viewed from a different detector position in Fig. 5.21 (b). The peaks of the lines in Fig. 5.21 (b) are represented by black dots on the line in Fig. 5.21 (a).

Using the angles calculated by the linear function obtained from 5.21 (a), the wavelength can be calculated from equation 5.1. This wavelength is then subtracted from the wavelength obtained from the NIST database and the difference is expressed as a residual. The calculation of the residuals for the points in Fig. 5.21 is shown in Table 6.

<table>
<thead>
<tr>
<th>Function angle (deg)</th>
<th>Function wavelength (nm)</th>
<th>Residuals (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>21.9431</td>
<td>199.02814</td>
<td>0.02486</td>
</tr>
<tr>
<td>21.54471</td>
<td>193.64607</td>
<td>-0.00107</td>
</tr>
<tr>
<td>21.14229</td>
<td>188.41052</td>
<td>-0.13252</td>
</tr>
<tr>
<td>20.95196</td>
<td>185.61104</td>
<td>0.19096</td>
</tr>
</tbody>
</table>

Table 6: Table showing the calculation of the residuals from the function obtained in Fig. 5.21

In Table 7, the function angle is the calculated angle according to the
equation 5.2 and the function wavelength is the wavelength calculated from the function angle according to the equation 5.1 where $\beta$ is now the function angle. The residuals is the difference between the function wavelength in Table 6 and the NIST wavelength in Table 5.21 for the same pixel.

The residuals in Table 6 have a considerable variation and need to be improved by moving a selected pixel, in turn, forward or backwards and then putting the new pixel positions back into the function obtained from in equation 5.2.

This procedure is repeated until increasing or decreasing any pixel by 1 results in an increase in the residuals. Once this point is reached a new optimized function be plotted in the same way as Fig. 5.21 and then the residuals can be recalculated. Equation 3.3 describes and the optimized pixel and NIST angle plot represented in Table 7 and in Fig. 5.22.

$$y = -8.11623 \times 10^{-4} x + 22.69964$$

(5.3)
Table 7: Pixel and NIST angle data used in Fig. 5.22

It is evident from the R-squared and the standard deviation (SD) values shown in Fig. 5.22 that the points used in Table 7 have a good fit. When these new values are used and the pixel numbers are entered into the function calculated in Fig. 7 (equation 5.3) the residuals are calculated and displayed in Table 8.

Table 8: Table showing the calculation of the residuals from the function obtained in Fig. 5.22

The resolution of the spectrometer is defined as $\frac{\lambda}{2\Delta\lambda}$. For example if $\lambda = 193.653$ nm then the FWHM at that wavelength was measured to be 0.198 nm by subtracting the shorter wavelength at FWHM from the longer wavelength at FWHM. The upper limit of the resolution of the spectrometer is then calculated to be 978. This represents the upper limit on the resolution of the spectrometer. When an experiment is being conducted this calculation will need to be repeated in the region of interest.

Optimizing the residuals reveals one more challenge to overcome before this calibration can be applied to the whole spectrometer. This challenge is illustrated in Fig. 5.23.
Figure 5.23: Figure showing the agreement of the residuals with the spectrum

The same spectral lines discussed in Fig. 5.21 and Table 7 are depicted in Fig. 5.23. Fig. 5.23 (b), (c) and (d) show the four strongest lines in Fig. 5.23 (a) are expanded and presented in 5.23 (b), (c) and (d) where the pixels for the optimum residual shown in Tables 7 and 8 are pinpointed on the spectrum. It is evident from Fig. 5.23 (b) and Fig. 5.23 (c) that the optimized residuals match with the spectrum but in Fig. 5.23 (d) the optimized residuals have placed the best pixel unacceptably far from the centre of both lines. The shift of the optimum residual position to the apparent position from the spectrum is the same phenomenon that was observed in Fig. 5.20. This demonstrates that only a certain amount of the spectrum recorded at a single detector position can be used before a new spectrum must be imported and collated in such a way that the peaks of the lines shown in Fig. 5.23 (d) overlap with the points of the arrows. The spectrum from the new detector position should be imported and joined at some point before the line begins, perhaps before pixel 2040. The best compromise between putting the peaks in the correct position to agree with the function and making the master spectrum free of phosphor defects should be attempted. An example of this collation process is shown in Fig. 5.24.
The two spectra in Fig. 5.24 are in final positions and agree with the function calculated in Fig. 5.24 to the nearest pixel. The black spectrum is arbitrarily taken as being correct for all pixels before pixel 1800 and the red spectrum is taken to be correct for all pixels after pixel 1800. (Any pixel, between 1600 and 1875, could clearly have been used to separate the beginning and the end of the two spectra). The spectrum in red is then assumed to be correct until the lines in it begin to disagree with the function obtained in Table 8. All the points from the red detector position in Fig. 5.24 prior to pixel 1800 are not considered and all the pixels after 1800 from the black detector position are equally ignored. This results in the final joined spectrum of the two detector positions shown in Fig. 5.25.
The procedure should then be continued for the next line. It was found that the line at the extreme right of Fig. 5.25 would not completely agree with the function obtained in Fig. 5.22. To that end, another detector position was imported and collated the same way as in Fig. 5.24 and Fig. 5.25 so that the line and those that follow agree with the function obtained in Fig. 5.22. This process was repeated for 57 different detector positions, which covered the detection area of the spectrometer. The final Al spectrum obtained is shown in Fig. 5.26.

The complete Al spectrum, displayed in Fig. 5.26 was obtained by col-
lating different detector positions over the spectral range range of the spectrometer. The spectrum progresses from the longer wavelength on the left to the shorter wavelength on the right. The three colours of the spectrum in Fig. 5.26, black, red and blue represent the number of shots required to obtain the counts shown in Fig. 5.26. The difference in the number of shots illustrates the reduced reflectivity of the platinum coated grating in the red and the blue region. Just as in Fig. 5.22, there was a function obtained by plotting the pixels of the identified lines of Fig. 5.26 against the angles of the corresponding identified lines in the NIST database. A total of 159 Al lines were identified from the spectrum displayed in Fig. 5.26. There were many additional lines due to impurities in the grade of aluminium used from the mechanical workshop. The total function obtained is shown in Fig. 5.27 and can be described by equation 5.4.

\[ y = -8.11362 \times 10^{-4}x + 22.69927 \]  

(5.4)

Figure 5.27: Complete pixel vs angle curve for all 159 Al lines identified from the NIST database.

The linear fitting of 159 lines of Al that correspond to known lines in the NIST database is illustrated in Fig. 5.27. The standard deviation of the points from the best fit of the line is 2.3437 \times 10^{-4}. This is a very good result and it appears to confirm a high degree of confidence in the calibration. The mean of the absolute value of all the wavelength residuals is 0.003 nm. The final step, is to use the calibration function obtained in Fig. 5.27 (equation 5.4) to convert all the pixels, shown in Fig. 5.26, into angles and then using equation 5.1, convert all the angles into wavelength.
The final result is plotted in Fig. 5.28. The segments of expanded spectra, divided into near 50 nm windows, are presented in Fig. 5.31, Fig. 5.32, Fig. 5.33 and Fig. 5.34.

**Figure 5.28:** Complete calibrated Al master spectrum.

**Figure 5.29:** Calibrated Al spectrum from 15 nm to 50 nm
Figure 5.30: Calibrated Al spectrum from 50 nm to 100 nm

Figure 5.31: Calibrated Al spectrum from 100 nm to 150 nm
Figure 5.32: Calibrated Al spectrum from 150 nm to 200 nm

References


6 Photoabsorption and fs experiments using the 1-m Normal Incidence Spectrometer

6.1 Introduction

This chapter presents data obtained using laser produced plasmas as a source of ions for photoabsorption in the case of indium and thulium and for fs-laser reheating in the case of indium. Spectra was recorded on the refurbished 1-m normal incidence spectrometer introduced in Section 5. The results of three experiments are discussed in this chapter:

1. $4d \rightarrow np$ and $4d \rightarrow mf$ photoabsorption of neutral In and In$^+$

2. $5p \rightarrow nd$ and $5p \rightarrow ms$ photoabsorption of neutral Tm, Tm$^+$ and Tm$^{2+}$

3. Emission studies of the fs reheating of an indium laser produced plasma.

In order to verify the refurbishment setup of the 1-m spectrometer, and in particular, the performance of the new detector, photoabsorption of In and In$^+$ were recorded as a repeat of the photoabsorption studies performed by Gráinne Duffy [2]. The experiment involved probing the indium plasma at different delay times with the continuum emitted by a tungsten laser produced plasma in the DLP configuration explained in Section 1.5.1.

With the quality of the photoabsorption setup confirmed by the photoabsorption of In, the key experiment involved replacing the absorbing species with Tm (Section 6.3), to perform photoabsorption studies of a laser produced plasma. New identifications were made in this work with suggestions for further studies at shorter wavelengths.

Finally the results of a fs laser reheating of an indium plasma will be outlined in Section 6.4.

6.2 Photoabsorption of Neutral In and In$^+$

6.2.1 Introduction

Section 6.2 describes an In photoabsorption experiment conducted on the 1-m normal incidence spectrometer. The aim of this experiment is to repeat, using the new phosphor coated linear CCD array detector, the photoabsorption studies of neutral [1] and singly ionized [2] indium. In Section 6.2.2 the DLP photoabsorption system is explained in detail including the time delay system for the lasers and the height control for the absorbing plasma. Photoabsorption spectra are presented and described in Section 6.2.3 and compared to the observations of the previous work mentioned.
6.2.2 DLP Set Up

The general dual laser plasma (DLP) set up is described in Section 1.5.1. In order to isolate different ion states in a plasma, and observe them in photoabsorption, it is necessary to be able to control the arrival times of the two Nd:YAG laser pulses with respect to one another. A photograph of the DLP set up in the target chamber of the 1-m normal incidence spectrograph is shown in Fig. 6.1. A yellow line connecting the continuum spot plasma, the absorbing line plasma and the entrance slit, represents the optical axis of the spectrometer.

![Figure 6.1: Top view photograph of the target chamber set up for the DLP system for the 1-m normal incidence spectrometer](image)

A piece of laser burn paper was placed at the focus of the Spectron laser to allow for an estimate of the spot size of the laser at focus. The burn paper was then photographed in the vicinity of a ruler as shown in Fig. 6.2. The lines of the ruler are clearly visible and yield:

\[ 196 \text{ pixels} \approx 1 \text{ mm} \]

The focus of the laser depicted by the burn mark on the brown paper near the top of Fig. 6.2 yielded a spot width of 0.8 mm
When this value for the spot is combined with the laser energy of 643 mJ at the lens and a temporal FWHM of 18 ns as measured from the FWHM of the beam in Fig. 2.18 the power density at the target can be estimated using equation equation 6.1:

\[
\phi = \frac{E}{\tau\pi \left(\frac{\omega}{2}\right)^2}
\]

(6.1)

Where:

- \( E \) - energy of the pulse
- \( \tau \) - laser temporal FWHM
- \( \omega \) - width of the spot

The power density focused onto the continuum target by the Spectron laser is estimated to be \( 6 \times 10^9 \text{ W cm}^{-2} \). A measurement of the area of the line plasma created by the Surelite is also shown in Fig. 6.3. Again the burn paper was photographed in the vicinity of a ruler as shown in Fig. 6.3. The lines of the ruler are clearly visible and yield:

115 pixels \( \approx 1 \text{ mm} \)

The focus of the laser depicted by the burn mark on the brown paper near the top of Fig. 6.3 yields a width of 1.4 mm and a length of 17 mm.
When this value for the spot is combined with the laser energy of 268 mJ at the lens and a temporal FWHM of 7 ns as measured in the FWHM of Fig. 2.15 the power density at the target can be estimated using equation 6.2:

\[
\phi = \frac{E}{\tau \omega L}
\]  
(6.2)

Where:

- \(E\) - energy of the pulse
- \(\tau\) - laser temporal FWHM
- \(\omega\) - width of the column
- \(L\) - Length of the column

The estimated power density on the target is \(1.6 \times 10^8\) Wcm\(^{-2}\).

Both lasers require a trigger pulse for their respective flash lamps and both lasers require some delay (laser dependent) before a second trigger pulse triggers a Q-switch, which results in the laser firing. To trigger the lasers a Quantum Composer 9520 pulse and delay generator was used. This device has four output ports, which were used to control the Q-switches and flash lamps of both lasers. Fig. 6.4 shows a block diagram of the timings required to synchronize both lasers.
The timings shown in Fig. 6.4 will give optimized output and cause both laser pulses to arrive at the target chamber at the same time as measured by a fast photodiode and an oscilloscope.

The timing is controlled by changing the trigger time of the Surelite flash lamp with respect to the Spectron flash lamp trigger time (port A). This is monitored using the Litron energy monitor which measures the time delay between pulses. The Litron measurement for the Spectron and the Surelite, where the Surelite flash lamp trigger (port C) is sent 220 ns before the Spectron flash lamp trigger is illustrated in Fig. 6.5.
The plot in Fig. 6.5 shows the delay between the pulses when the Surelite flashlamp is triggered \(-220\) ns with respect to the Spectron flashlamp trigger. The result is a 64 ns delay between the arrival of the 2 pulses. In practice, the Spectron laser has a jitter of about 10-15 ns [3]. When the above measurement was repeated five times, it was found to have a mean delay of 51.2 ns with a standard error of 4.7 ns. This process was carried out for a number of different flash lamp trigger delays and each trigger delay was measured five times so that a mean and standard error could be obtained for the separation.

To change the delay between the pulses, all that is needed is to, add or subtract time to the Surelite flash lamp trigger to move the arrival time between the pulses without effecting the internal timings of either laser. Note: the choice to only change the Surelite delay is arbitrary and the experiment could just as easily have been done with changes to the Spectron delay.

To achieve photabsorption of the highest ion states the absorbing plasma needs to be formed on the optical axis or higher, so that the continuum is absorbed by the hottest and densest part of the plasma. The height of the absorbing target can be adjusted using a mechanical feed-through positioner to the chamber. The best absorption (good contrast and high ion stages) was found to occur when the absorbing target is beginning to block the continuum from getting into the spectrometer.

To quantify the the height \(\Delta z\) of the absorbing target relative to the height of the continuum target the counts of the W continuum are measured as a function of absorber height which is adjusted by means of the mechanical
feed-through positioner. A typical height measurement plot is shown in Fig. 6.6.

\begin{center}
\includegraphics[width=0.5\textwidth]{figure6.6.png}
\end{center}

\textit{Figure 6.6: Plot showing the measurement of $\Delta z$ by plotting the continuum counts as a function mechanical feedthrough position.}

The vertical line labelled optical axis in Fig. 6.6 shows the height where $\Delta z = 0$ (where the absorber and the continuum are at the same level). All counts shown in Fig. 6.6, at all micrometer positions lower than the optical axis (4.7 mm), illustrate the absorbing target blocking the continuum light resulting in the lower counts observed at these micrometer positions. At points to the right of the optical axis (> 4.7 mm), the continuum is unaffected by the absorbing target due to the absorbing target being lower than the optical axis and therefore not blocking the light from entering the spectrometer.

\section*{6.2.3 Photoabsorption of In and In$^+$}

The successful recording of the photoabsorption spectra of an indium plasma illustrated that the multiple components of the DLP setup were fully functional. The time evolution of the photoabsorption spectra of In and In$^+$ is highlighted in Fig. 6.7. The offset spectra are colour coded according to the delay time at which they were recorded.
At the longer time delays of $\Delta \tau = 300$ and $\Delta \tau = 400$ ns, described by the green and orange photoabsorption spectra in Fig. 6.7, there is strong evidence of neutral In from the $4d^{10}5s^25p^1 \rightarrow 4d^{10}5s^25p^2$ transitions between 17 and 19 eV as described by Connerade [1]. Also observed in the same photoabsorption spectra is the strong In$^+$ resonance at 20.11 eV resulting from the $4d^{10}5s^2 \rightarrow 4d^{10}5s^2np^1$ and $4d^{10}5s^2 \rightarrow 4d^{10}5s^2nf^1$ transitions previously observed by Duffy [2]. Features due to neutral In are seen to disappear between the times $\Delta \tau = 200$ and $\Delta \tau = 150$ ns as the plasma is hotter at shorter delays and In$^+$ begins to dominate the spectrum. There is an observed broadening of the 20.1 eV resonance between delays $\Delta \tau = 150$ and $\Delta \tau = 200$ ns which is clear evidence of the excited In$^+$ $4d^{10}5s^15p^1 \rightarrow 4d^{10}5s^15p^2$ transitions [2]. This experiment successfully demonstrates the time-saving advantages of a digital CCD detection system over that of the slow and cumbersome photographic plate technique.
6.3 Photoabsorption Studies of Thulium (Tm) Ion States

6.3.1 Introduction

In Section 6.3 the discrete and continuum structure in the 5p photoabsorption and photoionization spectrum of thulium in the 25 eV - 40 eV region are presented. Spectra of neutral Tm, Tm$^+$ and Tm$^{2+}$ were recorded. The experimental set up for photoabsorption of Tm is identical to the one described in Section 6.2.2. The Section begins with a review of the previous photoabsorption work on Tm in Section 6.3.2. Preliminary Cowan code and RTDLDA calculations (introduced in Section 1.5.2) are shown in Section 6.3.3 and used to describe 4d discrete and continuum transitions, which are then compared to the published observed 4d photoabsorption spectrum of neutral Tm.

A complete time evolution of the measured 5p photoabsorption is shown in Section 6.3.4 and the individually identified ion states are addressed in turn, selected by an appropriate time delay between the lasers and interpreted by performing Cowan code and RTDLDA calculations. The spectrum of neutral Tm is discussed in Section 6.3.5, Tm$^+$ in Section 6.3.6 and Tm$^{2+}$ in Section 6.3.7. The conclusions of the Tm experiment will then be highlighted in Section 6.3.8.

6.3.2 Previous Photoabsorption Studies of Tm

Early photoabsorption studies of Tm were conducted by Tracey [4], who studied neutral 5p transitions from of all the lanthanides by heating the respective elements in a windowless furnace. The vapors were then exposed to synchrotron radiation where the respective absorption intensities were recorded using a spectrometer in the 20 to 40 eV region. None of the individual transitions were identified in Tm, however detailed calculations on neutral Yb showed that 5p → nd and 5p → ms transitions were present in this experiment. The use of vapors meant only photoabsorption of neutral Tm could be observed.

More recently, photoionization studies of Tm were carried out by Whitfield et al. [5] using photoelectron spectroscopy. Tm was studied again in vapor form which limited the studies to the neutral spices, however the absorption transitions were analyzed with the aid of Hartree Fock Cowan code calculations. This work established the Cowan code scaling parameters, the resonance transitions and some of the continuum states which could be replicated for the neutral thulium laser produced plasma and extrapolated to the higher ion states.

In addition to 5p photoabsorption, 4d photoabsorption was also observed
by Radtke [6]. Photoabsorption measurements of Tm vapors were again measured and probed using a synchrotron and a photographic plate based spectrometer. A significant resonance was observed peaking at approximately 7.15 nm and Hartree-Fock calculations confirmed that the resonance was due to $4d^{10}4f^{13}^2F_{2} \rightarrow 4d^{9}4f^{14}^2D_{2}$.

6.3.3 4d Photoabsorption in Thulium

Initial calculations were performed with the Cowan code and the RTDLDA calculations to compare the synthetic spectra with the observed spectrum from Radtke [6]. The RTDLDA calculates a very good representation of the giant resonance shape as illustrated in Fig. 6.8. The measured resonance [6] is shown in Fig. 6.8 (a), while the RTDLDA calculation, shifted by -0.198 nm so as to agree with the wavelength of the resonance, is demonstrated in Fig. 6.8 (b).

![Figure 6.8: RTDLDA comparison with the experimental observed Spectrum [6]](image)

No measured photoabsorption spectra exist for Tm$^+$, Tm$^{2+}$ or Tm$^{3+}$ so the evolution of the resonance with increasing ionization can only be eval-
uated using calculations. The RTDLDA calculations of the different Tm ion states are compared to one another in Fig. 6.9. A shift of -0.198 nm was placed on the calculated RTDLDA output for the neutral spectrum to make it overlap with the experimental spectrum recorded by Radtké. The calculated spectra of the other ion states of Tm are shifted to get agreement with the predicted $4d \rightarrow 4f$ transition wavelengths calculated by the Cowan code. A shift of -0.198 nm is applied to Tm$^+$ and -0.1988 nm was applied to Tm$^{2+}$. The RTDLDA calculation for Tm$^{3+}$ is not shifted as there is no clear line from the Cowan code (Fig. 6.10) to which the RTDLDA can be offset.

![Figure 6.9: Synthetic spectra based on RTDLDA calculations of 4d transitions of the first four Tm ion states](image)

The smaller features in Fig. 6.9, at lower wavelengths than the main resonance, represent discrete transitions which the RTDLDA code is less reliable at describing.

Values for the $4d \rightarrow 4f$ transition obtained from the Cowan code calculation are scaled against the value of the $4d \rightarrow 4f$ transition measured at
7.15 nm in neutral thulium [6]. This shift was also applied to the RTDLDA calculation shown in Fig. 6.9. The synthetic spectrum based on the Cowan code is shown in Fig. 6.10. All of the scaling factors were set to 80% as used by Whitfield et al. [5] except for the spin orbit interaction which was left unchanged.

The main transitions depicted in Fig. 6.10, according to the Cowan code for the mentioned ion states of thulium are illustrated in Tables 9, 10, 11 and 12. A shift equal to +0.9767 nm was applied to all the wavelengths in all the tables. This shift was equal to the difference between the calculated Cowan wavelength for $4d^{10}6s^24f^{13}[^2F_7^2] \rightarrow 4d^96s^24f^{14}[^2D_5^2]$ in Tm and the wavelength measured by Radtke [6].

**Figure 6.10: Synthetic spectrum based on Cowan code calculations of 4d transitions in Tm to Tm$^{3+}$**
<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (nm)</th>
<th>Oscillator Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4d^{10}6s^24f^{13} \quad 2F_{7/2} \rightarrow 4d^{9}6s^{2}4f^{14} \quad 2D_{5/2}$</td>
<td>7.151</td>
<td>4.91</td>
</tr>
<tr>
<td>$4d^{10}6s^24f^{13} \quad 2F_{5/2} \rightarrow 4d^{9}6s^{2}4f^{14} \quad 2D_{3/2}$</td>
<td>6.88493</td>
<td>3.61</td>
</tr>
</tbody>
</table>

Table 9: Calculated $4d \rightarrow 4f$ transitions for Tm (Cowan code)

<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (nm)</th>
<th>Oscillator Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4d^{10}6s^14f^{13} \quad 3F_{4} \rightarrow 4d^{9}6s^{1}4f^{14} \quad 3D_{3}$</td>
<td>7.15</td>
<td>5.57</td>
</tr>
<tr>
<td>$4d^{10}6s^14f^{13} \quad 1F_{3} \rightarrow 4d^{9}6s^{1}4f^{14} \quad 1D_{2}$</td>
<td>7.15</td>
<td>4.10</td>
</tr>
<tr>
<td>$4d^{10}6s^14f^{13} \quad 3F_{3} \rightarrow 4d^{9}6s^{1}4f^{14} \quad 3D_{2}$</td>
<td>6.88</td>
<td>4.17</td>
</tr>
<tr>
<td>$4d^{10}6s^14f^{13} \quad 3F_{2} \rightarrow 4d^{9}6s^{1}4f^{14} \quad 3D_{1}$</td>
<td>6.88</td>
<td>2.68</td>
</tr>
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</table>

Table 10: Calculated $4d \rightarrow 4f$ transitions for Tm$^{2+}$ (Cowan code)

<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (nm)</th>
<th>Oscillator Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4d^{10}4f^{13} \quad 2F_{7/2} \rightarrow 4d^{9}4f^{14} \quad 2D_{5/2}$</td>
<td>7.15</td>
<td>4.97</td>
</tr>
<tr>
<td>$4d^{10}4f^{13} \quad 2F_{5/2} \rightarrow 4d^{9}4f^{14} \quad 2D_{3/2}$</td>
<td>6.88</td>
<td>3.59</td>
</tr>
</tbody>
</table>

Table 11: Calculated $4d \rightarrow 4f$ transitions for Tm$^{2+}$ (Cowan code)
<table>
<thead>
<tr>
<th>Transition</th>
<th>Wavelength (nm)</th>
<th>Oscillator Strength</th>
</tr>
</thead>
<tbody>
<tr>
<td>$4d^{10}4f^{12}[^4H_6] \rightarrow 4d^{10}4f^{15}[^3H_6]$</td>
<td>7.28</td>
<td>3.04</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^4F_4] \rightarrow 4d^{10}4f^{15}[^3F_4]$</td>
<td>7.27</td>
<td>4.14</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^3H_5] \rightarrow 4d^{10}4f^{15}[^3H_5]$</td>
<td>7.18</td>
<td>1.78</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^3H_4] \rightarrow 4d^{10}4f^{15}[^3F_4]$</td>
<td>7.18</td>
<td>2.28</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^3F_3] \rightarrow 4d^{10}4f^{15}[^3F_3]$</td>
<td>7.18</td>
<td>2.45</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^3H_4] \rightarrow 4d^{10}4f^{15}[^3F_3]$</td>
<td>7.17</td>
<td>1.52</td>
</tr>
<tr>
<td>$4d^{10}4f^{12}[^3H_5] \rightarrow 4d^{10}4f^{15}[^3F_3]$</td>
<td>7.16</td>
<td>2.08</td>
</tr>
<tr>
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<td>1.95</td>
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</tr>
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<td>6.69</td>
<td>15.73</td>
</tr>
</tbody>
</table>

*Table 12: Calculated 4d → 4f transitions for Tm$^{3+}$ (Cowan code)*

These two sets of calculations set a good foundation for a DLP experiment to measure both the broad resonance and any discrete transitions that might occur.

### 6.3.4 Results for 5p Photoabsorption

The 5p photoabsorption is known to create structure in the spectrum of neutral thulium in the region of 25 to 35 eV. Calculations using the Cowan code predict that 5p photoabsorption in Tm$^{+}$ and Tm$^{2+}$ will also be observable in this region. The time evolution of the photoabsorption structure of thulium...
laser produced plasmas are shown in Fig. 6.11. The different delay times $\Delta \tau$ between the formation of the absorbing plasma and the probing continuum plasma are shown by the different coloured photoabsorption spectra starting from the shortest $\Delta \tau$ at the bottom to the longest $\Delta \tau$ at the top which have been vertically offset for clarity. All the spectra have undergone a 5-point moving average smoothing in order to reduce the influence of noise on the spectra.
Figure 6.11: Time evolution of the Photoabsorption of a thulium laser plasma at a range of time delays from 150 ns to 1900 ns

The observed ion states start with Tm$^{2+}$ at $\Delta\tau = 150$ ns $\rightarrow$ $\Delta\tau = 275$ ns, which can be identified from a characteristic spike at 34.46 eV along with discrete transitions between 29 eV and 29.5 eV. The ion state Tm$^+$ begins to manifest strongly from approximately $\Delta\tau = 400$ ns with a pair of tran-
sitions between 28 and 29 eV, which seems to be still present at 1100 ns mixed with onset of neutral ions. The Tm$^+$ spectrum also contains a broad group of transitions spanning from 32.2 eV to approximately 35 eV which peaks in intensity at about 34.2 eV. Finally, neutral Tm begins to appear at $\Delta \tau = 1100$ ns but is not isolated from Tm$^+$ until $\Delta \tau = 1900$ ns. The presence of the neutral Tm at $\Delta \tau = 1900$ ns ion can be inferred from the discrete pair of lines at 27.3 eV and 27.64 eV along with broad quasi continuum states spanning from approximately 32.1 eV to 35.6 eV and peaking at 32.44 eV.

6.3.5 Results for the Photoabsorption of Neutral Tm

As the spectrum of neutral thulium has been analyzed by Tracy [4] using a furnace and synchrotron, it is the ideal place to start the analysis of the time-resolved study of the thulium laser produced plasma. The first step was to run the Cowan code to reproduce the assignments by Whitfield et al. [5]. Similarities between the spectra recorded in this work at time delays $> 1100$ ns and the spectrum recorded by Tracey [4] can be seen in Fig. 6.12. The calibration of the Tm spectrum is confirmed by comparing it to the results of the photoabsorption conducted by Tracey [4]. This comparison is made with all of the later ion states which include neutral Tm as shown in Fig. 6.12. The different photoabsorption spectra delay times $\Delta \tau$, between $\Delta \tau = 1100$ ns and $\Delta \tau = 1900$ ns, are identified by different colours beginning with the shortest $\Delta \tau$ time at the bottom to the longest $\Delta \tau$ time at the top and are offset for clarity. All the spectra have undergone a 5-point moving average smoothing in order to reduce the influence of noise on the spectra.
Figure 6.12: Comparison of Tm Photoabsorption on the 1-m normal incidence spectrometer to the work of Tracey [4]

The important distinguishing peaks at 27.37 eV and 27.66 eV are present in all of the delays presented in Fig. 6.12 and are within 0.035 eV of the published energy [4]. Even without calculations, it is clear, by comparing Tracey’s absorption trace (shown as the top purple trace in Fig. 6.12) to all the others, that neutral Tm is not completely isolated from Tm$^+$ until $\Delta \tau = 1900$ ns.

The resolution of the 1-m normal incidence spectrometer is higher than that of the spectrometer used by Tracey [4]. The line at 27.66 eV in Tracey’s data has been resolved into two lines as demonstrated in Fig. 6.13. The spectrum obtained by Tracey is illustrated in Fig. 6.13 (a) and the spectrum obtained on the 1-m normal incidence spectrometer is illustrated in Fig. 6.13 (b).
Figure 6.13: Comparison of Tracey’s spectrum of neutral thulium [4] with the photoabsorption of the thulium plasma at a time delay of $\Delta\tau = 1900$ ns.

With the calibration confirmed, the next step is to identify features using the RTDLDA and Cowan codes. The RTDLDA code is expected to accurately describe the quasi-continuum observed between 32.1 eV and 35.6 eV. The RTDLDA output is compared with the Tm spectrum at $\Delta\tau = 1900$ ns in Fig. 6.14. The comparison between the RTDLDA cross section shifted by 0.4 eV and the experimental neutral thulium spectrum is shown in Fig. 6.14 (a) and Fig. 6.14 (b).
When a shift of $+0.4$ eV is applied to the RTDLDA calculations the quasi-continuum absorption is well described by RTDLDA output in terms of shape. The lower energy discrete behaviour is not well described by the RTDLDA calculations but this is as expected from the way the code approximates the many electron Schrödinger equation with a total energy function. The resonance occurs due to photoexcitation of the 5p electron at 31.42 eV (as calculated by Cowan), which results in a combination of absorption due to direct photoionization and to discrete auto-ionized transitions.

The next step is to compare the discrete photoabsorption at $\Delta \tau = 1900$ ns observed at approximately 27.3 eV and 27.6 eV to that of the Cowan code output.

The ground state of neutral thulium is $5p^64f^{13}6s^{2}$ and the dominant photoabsorption transition in the specified spectral region is to $5p^54f^{13}6s^25d^1$. The Cowan calculations included $5p \rightarrow ns$ and $5p \rightarrow md$ (where $7 \leq n \leq 10$) and (where $5 \leq m \leq 10$). The higher m and n values are much weaker transitions than $5p \rightarrow 5d$ but need to be included to correct for the interchannel configuration interaction effects. Since the $5p^54f^{13}6s^25d^1$ excited state has a binding energy which is estimated by the Cowan code to be 21.29 eV above the ionization potential of neutral thulium, autoionization decay is likely to occur. In this process the $5p^54f^{13}6s^25d^1$ will undergo a radiationless decay through a number of decay channels which will also result in the ionization
of an electron. For neutral thulium the calculated 5d decay channels are as follows:

1. \(5p^5f^{13}6s^25d^1 \rightarrow 5p^6f^{13}6s^1 + \epsilon l(l = 1, 3)\)
2. \(\rightarrow 5p^6f^{12}6s^2 + \epsilon l(l = 0, 2, ...\))
3. \(\rightarrow 5p^6f^{13}5d^1 + \epsilon l(l = 1)\)
4. \(\rightarrow 5p^6f^{12}6s^15d^1 + \epsilon l(l = 0, 2, ...\))
5. \(\rightarrow 5p^6f^{11}6s^25d^1 + \epsilon l(l = 1, 3, ...\))
6. \(\rightarrow 5p^5f^{14}6s^1 + \epsilon l(l = 2)\)

Also considered in the calculation were the 7s decay channels which are subsequently shown in Fig.6.17 to influence the decay profile. The 7s decay channels calculated were as follows:

1. \(5p^5f^{13}6s^27s^1 \rightarrow 5p^6f^{13}6s^1 + \epsilon l(l = 1)\)
2. \(\rightarrow 5p^6f^{12}6s^2 + \epsilon l(l = 0, 2, ..\))
3. \(\rightarrow 5p^6f^{13}7s^1 + \epsilon l(l = 1)\)
4. \(\rightarrow 5p^6f^{12}6s^17s^1 + \epsilon l(l = 0, 2, ..\))
5. \(\rightarrow 5p^6f^{11}6s^27s^1 + \epsilon l(l = 1, 3, ..\))
6. \(\rightarrow 5p^5f^{14}6s^1 + \epsilon l(l = 3)\)

Where:

- \(\epsilon\) - the kinetic energy of the ionized electron.
- \(l\) - the angular momentum of the ionized electron.

The kinetic energy of the free electron is calculated by subtracting the binding energy of each final state of the decay channel from the binding energy of the \(5p \rightarrow 5d\) or the \(5p \rightarrow 7s\) excited state. Autoionization calculations for higher \(5p \rightarrow md\) and \(5p \rightarrow ns\) are physically possible, however the Cowan code dimensions would not permit the processing of these additional decay channels.

The Cowan output consists of a file which contains a column of transition energies and a column of line widths based on all the decay channels possible for that transition. The Cowan code autoionization calculation output, where only transitions originating from the
The synthetic autoionized transitions presented in Fig. 6.15 have been convolved with Lorenzians with a width equal to the calculated autoionized widths. The shifts applied to all the synthetic spectra were based on lining up
the higher energy peak of the 27 eV to 28 eV transitions with the transitions observed in Tracey’s spectrum at 27.6 eV. There is very little difference between the spectra created with different scaling factors and so an 80% scaling was chosen to best fit the data to agree with the scaling chosen by Whitfield et al. [5]. There is also reasonable agreement between the synthetic spectra and the structure observed by Tracey between 35 eV and 37 eV.

There is a disproportionate strength and shape to the higher energy wide resonance described by the Cowan code. It is clear that the Cowan code is overestimating oscillator strength of the wide absorption resonance between 32 eV and 35 eV. For this reason the peak of the wide resonance was not used to determine the shifts of the synthetic spectra when compared to the experimental data.

There is structure in the 26 eV to 27 eV region which is not well described by the addition of autoionization broadening, however it is predicted by the Cowan code calculation. A comparison is made between a stick plot of the non autoionized photoabsorption transitions and Tracey’s spectrum in Fig. 6.16.
Figure 6.16: Comparison of the non autoionized transitions calculated by Cowan and Tracey’s spectrum [4]

It is clear from Fig. 6.16 that the Cowan code does predict transitions between 26 eV and 27 eV but that the combination of the weak oscillator strengths and the larger energy spacing between the lines means that after convolution these transitions will not have significant magnitude.

The shape of the resonance is also sensitive to the number of decay channels included in the calculation.

A difference can be observed by calculating the 5d decay channel spectrum and then comparing the spectrum with a combination of 5d and 7s decay channels included in a single calculation as illustrated in Fig.6.17. The calculated spectrum, obtained by including both 5d and 7s decay channels, is plotted in Fig. 6.17 (a) and the calculated spectrum from the 5d decay
channels is plotted in Fig. 6.17 (b).

Figure 6.17: Comparison between using 5d decay channels and using 5d + 7s decay channels

The wide absorption resonance is clearly made broader with the inclusion of the 7s decay channels. Based on the improvements observed by including the 7s decay channels it stands to reason that further improvements in the
shape of the resonance could be obtained with the inclusion of additional
decay channels from other excited states. This could not be explored further
even though the Cowan code dimensions were not exceeded with the addition
of further decay channels the limitation lies with the shrink program, that
processes the output, as it appears unable to process all the continuum states.

As a final comparison with previous work, widths of two specific lines are
compared with the published widths from the paper of Whitfield *et al.* in
Table 13.

Where:

- Upper term - the LS term of the upper energy level in the transition.
- Energy X% - The energy difference between the upper level described
  by the upper term column and the ground state - \((5p^64f^{13}6s^2\left[2F_\frac{7}{2}\right])\),
as calculated with X% scaling.
- Width X% - The width the line is broadened due to the inclusion of
decay channels in the Cowan calculation.
- Energy (Published) - The energy experimentally observed difference
  between the ground state \((5p^64f^{13}6s^2\left[2F_\frac{7}{2}\right])\) and the upper level
described by the Upper term column, as published in Whitfield *et al.* [5].
- Width (Published) - The experimentally observed width the line is
  broadened due to autoionization decay as published in Whitfield *et al.* [5].

<table>
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<th>Upper term</th>
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<th>Width 80%</th>
<th>Energy 85%</th>
<th>Width 85%</th>
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<th>Width 90%</th>
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<th>Width Published</th>
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<td>27.14</td>
<td>0.322</td>
<td>27.19</td>
<td>0.327</td>
<td>27.12</td>
<td>0.223</td>
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</tbody>
</table>

*Table 13: Comparison of the calculated widths and the widths published in Whitfield *et al.* for two selected lines.*

It is clear that energy of a transition varies proportionally with the scaling
parameters used in the Cowan input, so replicating the energy of the
transition for both upper terms would require scaling between 80% and 85%. Whitfield et al. conducted their calculations using a custom version of the Cowan code with a scaling factor of 80%. Their calculations appeared to underestimate the widths when compared to their own measured values. The widths calculated using the standard Cowan code appeared to agree more closely with the Whitfield experimental data than that of the custom version of the Cowan code. The widths calculated, in this work, by the standard Cowan code were within a factor of 2 of the experimental Whitfield data.

The next step was to compare the Cowan code calculations with the experimentally observed thulium neutral spectrum observed with a laser delay of $\Delta \tau = 1900$ ns. In this comparison, the absorbing species is a laser produced plasma and so contributions arising from absorption from the $5p^{6}4f^{13}6s^{2}\left[2F_{\frac{3}{2}}\right]$ state were also possible and included in these calculations. The experimental spectrum observed with a laser delay of $\Delta \tau = 1900$ ns is shown in Fig. 6.19 (a) and the spectra from the Cowan calculations for scaling of 90%, 85% and 80% are plotted in Fig. 6.18 (b), Fig. 6.18 (c) and Fig. 6.18 (d).
In Fig. 6.18 the synthetic autoionized transitions have been convolved with Lorenzians with a width equal to the calculated autoionized widths. Again, there is not a huge variation in the shape of the spectrum from the scaling parameters and so 80% is chosen as it both agrees with Whitfield et al calculations and there is more evidence of the lower energy peak at 27.3 eV than with any of the other scaling parameters. Once again, the wide resonance in the 32 eV to 35 eV region is not well described by the Cowan calculations for the same reasons as for the comparison with Tracey’s data.
Cowan calculations were used to simulate the spectrum of the $4f^{14}6s^1$ excited.

For the excited $4f^{14}6s^1$ of thulium the calculated 5d decay channel are as follows:

1. $5p^54f^{14}6s^15d^1 \rightarrow 5p^64f^{14}6s^0 + \epsilon l (l = 1, 3)$
2. \[ \rightarrow 5p^64f^{13}6s^1 + \epsilon l (l = 1, 3, ..) \]

Where:

- $\epsilon$ - the kinetic energy of the ionized electron.
- $l$ - the angular momentum of the ionized electron.

The comparison of the neutral thulium spectrum to both the autoionized excited $4f^{14}6s^1$ transitions and the 80% scaled autoionization spectra is shown in Fig. 6.19. The photoabsorption spectrum obtained at a delay $\Delta \tau = 1900$ ns, is plotted in Fig. 6.19 (a), the synthetic autoionized spectrum from the $4f^{14}6s^1$ excited state is plotted in Fig. 6.19 (b) and the autoionized photoabsorption spectrum with an 80% scaling parameter is illustrated in Fig. 6.19(c).
Figure 6.19: Comparison of synthetic autoionized spectra to an experimental spectrum obtained at a laser time delay of $\Delta \tau = 1900\,\text{ns}$

The autoionized spectrum shown in Fig. 6.19 (c) does illustrate agreement with the experimentally obtained spectrum shown in Fig. 6.19 (a) in the 27 eV to 28 eV region and again in the 35 eV to 36 eV region. It is unclear why the transitions around 33 eV seem so much stronger than the group around 27 eV, when this is not reflected in the experimental spectrum. The calculated excited state shown in Fig. 6.19 (b) requires an overly large shift of 1.123 eV for the transition to overlap with the 30.5 eV absorption line and this with an 85% scaling. It should also be noted that the transition overlapping with the 30.5 eV line is the highest energy transition calculated in the $4f^{14}6s^1$ excited state and the Cowan code is known to dump oscillator strength into the final transition of a series.[7] This is therefore a tentative assignment.
6.3.6 Results for the Photoabsorption of Singly Ionized Thulium

In the case of Tm$^+$ there are no previously published spectra for comparison. Spectra for two delay times, namely $\Delta \tau = 400$ ns and $\Delta \tau = 1100$ ns shown in black and red respectively in Fig. 6.20 indicate features that could arise from the Tm$^+$ ion. All the spectra have undergone a 5-point moving average smoothing in order to reduce the influence of noise on the of the spectra.

![Thulium photoabsorption spectra at delay times which demonstrate absorption due to Tm$^+$](image)

**Figure 6.20:** Thulium photoabsorption spectra at delay times which demonstrate absorption due to Tm$^+$

Although the spectrum recorded at $\Delta \tau = 1100$ ns does have structure due to absorption by Tm$^+$, the features at $\Delta \tau = 400$ ns are much stronger and there is very little neutral thulium present in the plasma. The results of the RTDLDA calculations are illustrated in Fig. 6.21. The calculated photoabsorption cross section is shown in Fig. 6.21 (a) and for comparison, the experimental spectrum which was recorded at a time delay of $\Delta \tau = 400$ ns
is presented in Fig. 6.21 (b). The RTDLDA-generated spectrum is shifted by $+0.3 \text{ eV}$ to align it with the experimental data.

Figure 6.21: RTDLDA comparison with photoabsorption of the thulium plasma at a time delay of $\Delta \tau = 1900 \text{ ns}$.

The computed cross section shown in Fig. 6.21 (a) suggests that the absorption rising edge occurs between 33.75 eV and 34 eV, whereas in Fig. 6.21 (b) in the spectrum observed for $\Delta \tau = 400 \text{ ns}$, the rising absorption edge is seen between 32.25 eV and 34 eV. The RTDLDA calculation output in 6.21 (a) gives the falling edge of the quasi-continuum absorption resonance between 34 eV and approximately 35.75 eV. The RTDLDA calculation does however suggest that the absorption should continue to fall after 35.75 eV, whereas some absorption is observed in the experimental spectrum (6.21
(b)) at higher energies. This may be attributable to the presence of other absorbers in the plasma such as oxygen and nitrogen which both have transitions in this energy region.

Just as with neutral autoionization, calculations were performed for singly excited thulium. The ground configuration of singly excited thulium is $5p^64f^{13}6s^1$ and all significant absorption transitions are again observed through inner shell transitions to a combination of $5p^64f^{13}6s^1nd^1$ and $5p^64f^{13}6s^1ms^1$ excited states.

Since the $5p^54f^{13}6s^15d^1$, $5p^54f^{13}6s^2$, $5p^54f^{13}6s^16d^1$, $5p^54f^{13}6s^17s^1$ and $5p^54f^{13}6s^17d$ excited states have binding energies which are estimated by the Cowan code to be 3.42 eV, 5.49 eV, 6.43 eV, 12.33 eV and 13.49 eV above the ionization potential of singly ionized Tm, autoionization decay is likely to occur. The autoionization mechanism is as described in Section 6.3.6 for neutral thulium except there are fewer possible decay channels because there are less valence electrons. For singly ionized thulium the possible nd decay channels are as follows where the kinetic energy is calculated as in neutral thulium:

1. $5p^54f^{13}6s^1nd^1 \rightarrow 5p^64f^{13}6s^0 + \epsilon l (l = 1, 3)$
2. \quad $\rightarrow 5p^64f^{12}nd^1 + \epsilon l (l = 0, 2, ..)$
3. \quad $\rightarrow 5p^64f^{12}6s^1 + \epsilon l (l = 0, 2, ..)$

Where:

- $(n = \{5, 6, 7\})$ were included in the calculations.

In addition the following ms decay channels were also included.

1. $5p^54f^{13}6s^2ms^1 \rightarrow 5p^64f^{13}6s^0 + \epsilon l (l = 1)$
2. \quad $\rightarrow 5p^64f^{12}6s^0ms^1 + \epsilon l (l = 0, 2, ..)$
3. \quad $\rightarrow 5p^64f^{12}6s^1ms^0 + \epsilon l (l = 0, 2, ..)$

Where:

- $(m = \{6, 7\})$ were included in the calculations.
- $\epsilon$ - the kinetic energy of the ionized electron.
- $l$ - the angular momentum of the ionized electron.
Just as with neutral thulium, all of the above decay channels were generated with different scaling parameters and compared to the photoabsorption measured from the laser produced plasma. Again, due to the absorber being a laser produced plasma, transitions from the states $4f^{13}6s^1[^3F_4]$, $4f^{13}6s^1[^3F_3]$, $4f^{13}6s^1[^3F_2]$ and $4f^{13}6s^1[^1F_3]$ are all possible and were included in all calculations. The comparison of the measured photoabsorption spectrum at $\Delta \tau = 400$ ns for the different scaling parameters is shown in Fig. 6.22. The experimental spectra observed with a laser delay of $\Delta \tau = 400$ ns is presented in Fig. 6.22 (a) along with the Cowan calculations with scalings of 90%, 85% and 80% plotted in Fig. 6.22 (b), Fig. 6.22 (c) and Fig. 6.22 (d) respectively. The synthetic autoionized transitions have been convolved with Lorenzians with a width equal to the calculated autoionization widths.

![Figure 6.22: Comparison of different scaling factors of singly ionized thulium to the photoabsorption measurement at $\Delta \tau = 400$ ns](image)

The synthetic spectra reasonably describe the experimental spectrum.
shown in Fig. 6.22 (a). The absorption region observed between 28 eV and 29 eV is described by $5p \rightarrow 5d$ and $5p \rightarrow 6s$ transitions and the absorption region observed between 33 eV and 37 eV is described by a complex mixture of ms and nd transitions. Based on the higher energy region (34 eV and over) 80% scaling is seen to fit the data best as the peak of the resonance occurred around 34 eV, and then falls in strength till 35 eV and levels out towards 37 eV. The 80% scaling generates the plot that best reproduces this structure and overlaps well when a 0.415 eV shift is applied. Although the Cowan code reproduces the spectrum well at high energy, the width of the 28 eV to 29 eV transition series is a better fit at higher scaling factors.

Next it is shown that singly ionized thulium is the dominant ion at $\Delta \tau = 400$ ns. To this end two excited states of singly ionized thulium were calculated along with doubly ionized thulium and compared to the spectra obtained at $\Delta \tau = 400$ ns. The two excited states considered were $4f^{14}$ and $4f^{12}6s^2$, however the $4f^{12}6s^2$ showed no evidence of being excited at $\Delta \tau = 400$ ns. Due to the high number of continuum states only 5d decays could be considered for the $4f^{14}$ excited state:

1. $5p^{5}4f^{14}5d^{1} \rightarrow 5p^{6}4f^{13} + \epsilon l (l = 0, 2)$
2. $\rightarrow 5p^{6}4f^{12}6s^{1} + \epsilon l (l = 1, 3, ..)$
3. $\rightarrow 5p^{6}4f^{11}6s^{1}5d^{1} + \epsilon l (l = 0, 2)$

The experimental spectrum observed with a laser delay of $\Delta \tau = 400$ ns is shown in Fig. 6.23 (a). The synthetic autoionized singly ionized spectrum with an 80% scaling is plotted in Fig. 6.23 (b). The autoionized excited states based on $4f^{14}$ are shown in Fig. 6.23 (c). Finally the doubly ionized autoionization spectrum (whose decay channels are described in Section 6.3.7) is plotted in Fig. 6.23 (d).
Figure 6.23: Comparison of synthetic spectra to experimental spectra obtained at a laser time delay of $\Delta \tau = 400$ ns

As already discussed, there is good overlap between the observed spectrum taken at $\Delta \tau = 400$ ns shown in Fig. 6.23 (a) and the autoionized synthetic spectra plotted in Fig. 6.23 (b). The synthetic spectrum representing the $4f^{14}$ excited state, plotted in Fig. 6.23 (c), suggests evidence for absorption at 24.24 eV. There is also a possibility that the 28.7 eV line from the
$4f^{14}$ excited state is contributing some of the remaining width to the features between 28 eV to 29 eV in the observed spectrum of Fig. 6.23 (a) that are not described by the singly ionized calculations plotted in Fig. 6.23 (b). A final indication of the presence of the $4f^{14}$ excited state at this delay time is at 32.93 eV where it contributes to the total absorption of the spectrum in that region.

When comparing the doubly ionized autoionization spectrum in Fig. 6.23 (d) with the experimental spectrum observed in Fig. 6.23 (a), there is no clear evidence of thulium doubly ionized at $\Delta \tau = 400 \text{ns}$.

As the delay decreases there is an increased dominance of the $4f^{14}$ excited state which is illustrated in the comparison of the $\Delta \tau = 375 \text{ns}$ delay and the Cowan simulations as illustrated in Fig. 6.24. The experimental spectrum observed with a laser delay of $\Delta \tau = 375 \text{ns}$ is shown in Fig. 6.24 (a). The synthetic autoionized singly ionized spectrum with 80\% scaling is plotted in Fig. 6.24 (b). The spectrum for the autoionized excited state $4f^{14}$ is shown in Fig. 6.24 (c). Finally, the doubly ionized autoionization spectrum (with decay channels described in Section 6.3.7) is plotted in Fig. 6.24 (d).
Figure 6.24: Comparison of synthetic spectra and experimental spectra at $\Delta \tau = 375$ ns

As with the $\Delta \tau = 400$ ns delay time, the spectrum obtained at time delay $\Delta \tau = 375$ ns, plotted in Fig. 6.24 (a), is well described by the synthetic spectrum of singly ionized thulium as plotted in Fig. 6.24 (b). The main difference between the $\Delta \tau = 375$ ns and $\Delta \tau = 400$ ns is the evidence of stronger $4f^{14}$ excited state features predicted in Fig. 6.24 (c). The features from 28.7
eV and 30.92 eV in Fig. 6.24 (c) have become visible and have good overlap with the observed spectrum features in the same region of Fig. 6.24 (a). The features at 32.93 eV in Fig. 6.24 (c) are also still present at this delay time but are not significantly stronger. There is no indication that the excited higher energy transitions observed in Fig. 6.24 (c) are significantly contributing to the absorption resonance between 33 eV and 36 eV in Fig. 6.24 (a). This region remains best described by the autoionized singly ionized thulium as plotted in Fig. 6.24 (b).

When comparing the doubly ionized autoionization spectrum in Fig. 6.24 (d) with the experimental spectrum observed in Fig. 6.24 (a), there is no clear evidence of thulium doubly ionized at $\Delta \tau = 375$ ns.

### 6.3.7 Results for the Photoabsorption of Doubly Ionized Thulium

As in the case of Tm$^+$, there are no published experimental spectra for Tm$^{2+}$ to compare with calculations. Spectra recorded at two delay times displayed in Fig. 6.25 demonstrate absorption due to the Tm$^{2+}$ ion, namely $\Delta \tau = 150$ ns and $\Delta \tau = 225$ ns shown in black and red, respectively, in Fig. 6.25. Both spectra have undergone a 5-point moving average smoothing in order to reduce the influence of noise on the spectra.
Figure 6.25: Photoabsorption spectra recorded at delay times which demonstrate the presence of Tm$^{2+}$ ions in the plasma.

In both spectra, Tm$^{2+}$ absorption is present in Fig. 6.25. Analysis will be performed on the $\Delta \tau = 225$ ns data as the absorption features are stronger than those at $\Delta \tau = 150$ ns. The RTDLDA cross section, with a shift of $+0.425$ eV, is shown in Fig. 6.26 (a) and the same energy region of photoabsorption at $\Delta \tau = 225$ ns is shown in Fig. 6.26 (b). Blue grid lines are included for ease of comparison.
When a shift of +0.425 eV is applied to the RTDLDA calculations demonstrated in Fig. 6.26 (a), the absorption observed at $\Delta \tau = 225$ ns is well described by the RTDLDA output in terms of shape. The RTDLDA calculations show in Fig. 6.26 (a) that the rising edge of the first absorption resonance begins at 33.98 eV, peaks at 34.38 eV and immediately forms a falling edge at 35.08 eV. The RTDLDA calculations then reveal a shoulder between 35.08 eV and 35.28 eV followed by a plateau region; and finally another resonance starting at 36.38, rising up to a peak at 36.68 eV and then returning close to the zero Mb level at 36.88 eV. All of this behavior is consistent with what is observed in Fig. 6.26 (b) except the final absorption peak between 36.68 eV and 36.88 eV is not as pronounced as the RTDLDA
calculations would imply.

As with the previous ion states the Cowan code was used to perform autoionization calculations of the doubly ionized thulium ion. The ground state of doubly ionized thulium is $5p^64f^{13}$ and all significant absorption transitions are again observed through inner shell transitions to a combination of $5p^54f^{13}nd^1$ and $5p^54f^{13}ms^1$ excited states. Since the $5p^54f^{13}d^1$ excited state has a binding energy which is estimated by the Cowan code to be 6.123 eV and above the ionization potential of triply ionized thulium, an autoionization decay is likely to occur. The autoionization mechanism is as described for neutral thulium in Section 6.3.5 except there are a lower number of possible decay channels due to there being less valence electrons. For doubly ionized thulium the only possible 5d decay channel is as follows where the kinetic energy is calculated as in neutral thulium:

$$5p^54f^{13}d^1 \rightarrow 5p^64f^{12} + \epsilon l (l = 0, 2, ..)$$

Where:

- $\epsilon$ - the kinetic energy of the ionized electron.
- $l$ - the angular momentum of the ionized electron.

Just as with neutral and singly ionized thulium all of the above decay channels were generated with different scaling parameters and compared to the photoabsorption measured from the laser produced plasma. Again due to the absorber being a laser produced plasma, transitions from the states $4f^{13} \left[ ^2F_7 \right]$ and $4f^{13} \left[ ^2F_5 \right]$ are both possible and are included in all calculations. The comparison of the measured photoabsorption spectrum at $\Delta \tau = 225$ ns against those calculated using the different scaling parameters is presented in Fig. 6.27.
Figure 6.27: Comparison of synthetic spectra generated with different scaling factors for autoionized doubly ionized thulium to the photoabsorption measurement at $\Delta \tau = 225$ ns

There is very little difference structurally between the spectra with different scaling parameters. The individual shifts are based on lining up the large synthetic peak with the peak observed at 34.5 eV in Fig. 6.27 (a). Other than this there are no clear features to compare with. For this situation a
scaling of 85% is chosen so as to keep the required shift below 0.5 eV.

The absorption spectrum obtained at $\Delta \tau = 225$ ns was compared to the synthetic spectrum of singly ionized thulium and different excited states in order to identify the features which are not due to photoabsorption by Tm$^{2+}$. The $4f^{12}6s^2$ excited state of singly ionized thulium was found to be excited at this time delay. Due to the high number of continuum states only autoionizing 5d decay channels could be considered for the $4f^{12}6s^2$ excited state:

1. $5p^54f^{12}6s^25d^1 \rightarrow 5p^64f^{12}6s^3 + \epsilon l (l = 1, 3)$
2. $5p^64f^{12}5d^1 + \epsilon l (l = 1, 3)$

Where:

- $\epsilon$ - the kinetic energy of the ionized electron.
- $l$ - the angular momentum of the ionized electron.

The experimental spectrum observed with a laser delay of $\Delta \tau = 225$ ns is shown in Fig. 6.28 (a). The synthetic autoionized singly ionized spectrum with 80% scaling is plotted in Fig. 6.28 (b) and the synthetic autoionized doubly ionized spectrum with 85% scaling is plotted in Fig. 6.28 (c). The calculated spectrum for singly ionized thulium based on the autoionized excited state $4f^{12}6s^2$ is presented in Fig. 6.28 (d). Finally the calculated spectrum for singly ionized thulium based on the autoionized excited state $4f^{14}$, is shown in Fig. 6.28 (e).
Figure 6.28: Comparison of synthetic spectra with experimental spectrum obtained at a laser time delay of $\Delta \tau = 225$ ns

As already discussed, the high energy peak at 34.5 eV in Fig. 6.28 (a) is well described by the Cowan calculation for the autoionized $\text{Tm}^{2+}$ which is plotted in Fig. 6.28 (c). This calculation for Fig. 6.28 (c) also accounts for a 29 eV to 29.5 eV component of the 28 eV to 32 eV group of transitions in Fig. 6.28 (a). The autoionized $\text{Tm}^+$ calculations plotted in Fig. 6.28 (b) has a significant presence in the 28 eV to 28.75 eV region, which appears to
contribute to the 28 eV to 32 eV group of transitions in Fig. 6.28 (a).

The \(4f^{14}\) excited state of \(\text{Tm}^+\) plotted in Fig. 6.28 (e) also contains structure between 28.9 eV and 31.2 eV that appears in the measured spectrum in Fig. 6.28 (a). Also in Fig. 6.28 (e) a low energy line at 24.5 eV and higher energy lines at 33.2 eV and above do not clearly be seen to impact on the measured spectrum in Fig. 6.28 (a). Finally the excited state \(4f^{12}6s^2\) plotted in Fig. 6.28 (d) appears to have features predominantly between 30.5 eV and 32 eV group of transitions in Fig. 6.28 (a). The strong high energy feature between 36.5 eV and 37.5 eV does not appear in the measured spectrum but this may be due to reflection limitations of the grating that severely restrict the sensitivity of the spectrometer in this region.

Most of the structure observed at the time delay \(\Delta \tau = 225\,\text{ns}\) has been identified, however some of the finer structure between 28 eV and 30 eV is unsatisfactorily described.

A final task is to demonstrate that there is no evidence of triply ionized thulium. Calculations for triply ionized thulium using the cowan code showed that it does not have any excited states that lie above the ionization potential and so autoionization is not possible. A comparison between the spectrum obtained at the earliest time delay observed \(\Delta \tau = 150\,\text{ns}\) is compared with autoionized doubly ionized thulium and instrumentally broadened triply ionized thulium, along with the \(4f^{12}6s^2\) excited state. The experimental spectrum observed with a laser delay of \(\Delta \tau = 150\,\text{ns}\) is shown in Fig. 6.29 (a) and the autoionized doubly ionized thulium spectrum is plotted in Fig. 6.29 (b). The instrumentally broadened calculated triply ionized thulium spectrum is plotted in Fig. 6.29 (c) and the \(4f^{12}6s^2\) excited state spectrum is plotted in Fig. 6.29 (d).
Figure 6.29: Spectrum of the plasma recorded at a time delay of $\Delta \tau = 150$ ns, compared with the synthetic spectra of Tm$^{2+}$, Tm$^{3+}$ and Tm$^{+*}$.

The spectrum observed with a laser delay of $\Delta \tau = 150$ ns shows no definitive evidence of triply ionized thulium as can be seen by comparing Fig. 6.29 (a) and Fig. 6.29 (c). From the peak at 34.5 eV it is clear that there is still doubly ionized thulium present as seen by comparing Fig. 6.29 (a) and Fig. 6.29 (b). With the $4f^{12}6s^2$ excited state in Fig. 6.29 (d), there is some evidence of contribution to some of the structure between 30.5 eV and 32 eV.
in Fig. 6.29 (a).

6.3.8 Conclusions

Photoabsorption of Tm ions was successfully achieved using the DLP set up described in Section 6.2.2 and with the refurbished 1-m normal incidence spectrometer described in Section 5.

Cowan code calculations and RTDLDA calculations were conducted for 4d photoabsorption and the RTDLDA calculations fit very well with the only published absorption spectrum available.

Photoabsorption of neutral Tm has been described before both experimentally and theoretically. In this thesis, the observation of neutral Tm at the time delays $\Delta \tau = 1100 \rightarrow 1900$ ns served to confirm the calibration of the energy region by direct comparison with the published result as completed in Fig. 6.12.

The RTDLDA calculations describe the high energy absorption resonance behavior very well in terms of shape, provided a shift of + 0.4 eV was applied. Autoionization calculations were conducted to simulate the shape of the transitions observed between 27 eV and 27.8 eV in Fig. 6.18. The first group of calculations only considered the ground state $5p^64f^{13}6s^2 [2F_7]$ which is consistent with the conditions in a furnace, which allows direct comparison between the Cowan calculations and the spectrum recorded by Tracey [4] and the Cowan code calculations recorded Whitfield et al. [5].

The synthetic spectra calculated with different scaling parameters were compared with Tracey’s spectrum in Fig. 6.15. At low energy there was no clear variation between the different scaling parameters. The only differences in varying the scaling parameters were global shifts of the spectrum to higher energy by increasing % scaling and some differences in the absorption structure between 35 eV and 36 eV. The 80 % scaling from the paper was retained as there was no reason to choose a different scaling.

The effects of autoionization on line width were then accounted for in the calculations and the results were then compared to the measured photoabsorption spectrum taken at $\Delta \tau = 1900$ ns. This comparison was completed in Fig. 6.18 and this time all ground configuration J values were included in the calculation. As with the initial ground state calculations, there was very little difference for calculations using different scaling parameters. When 80% scaling was used the 27 eV to the 28 eV region seemed to be divided into virtually two symmetrical peaks. These peaks were almost equal in size, which somewhat reflects the absorption characteristics in the measured $\Delta \tau = 1900$ ns spectrum and for this reason the 80% scaling was chosen.
Absorption of Tm$^+$ was observed in the time range $\Delta \tau = 400 \rightarrow 1100$ ns peaking at $\Delta \tau = 400$ ns as in Fig. 6.20. The spectrum at $\Delta \tau = 400$ ns was compared to theory as the transitions were much stronger than those of other time delays. The RTDLDAG calculations were applied to singly ionized thulium and were found to best fit with a shift of 0.3 eV as shown in Fig. 6.21. The rising edge of the RTDLDAG calculations is very steep compared to the experimentally observed spectra. The falling edge described by the calculations overshoots 35.75 eV which is the point at which regular noise continues on the experimentally observed spectrum.

Autoionization calculations using the Cowan code were also used to simulate the spectrum at $\Delta \tau = 400$ ns in Section 6.3.6. Singly ionized thulium required $5p \rightarrow md$ and $5p \rightarrow ns$ excitation channels (where $5 \leq m \leq 6$ and $6 \leq n \leq 7$) to best describe the absorption transitions. The absorption spectrum at $\Delta \tau = 400$ ns is compared for different scaling factors in Fig. 6.22. Based on the shape, 80% scaling was chosen to best describe the $\Delta \tau = 400$ ns data. The choice is based on the shape of the 34 eV to 36 eV region.

It was also shown in Fig. 6.23 that there is no evidence of doubly ionized thulium at $\Delta \tau = 400$ ns. Autoionization calculations were also performed for the $4f^{14}$ excited state of Tm$^+$, where due to the high number of continuum states only 5d decays could be included in the calculation. There was evidence of absorption from the $4f^{14}$ excited state at $\Delta \tau = 400$ ns. Reducing the delay time to $\Delta \tau = 375$ ns the evidence of the $4f^{14}$ excited state became stronger as shown in Fig. 6.24. It is also seen from Fig. 6.24 that there is still no evidence of doubly ionized thulium at $\Delta \tau = 375$ ns and that singly ionized thulium still has a strong presence.

Doubly ionized thulium was observed in the time range $\Delta \tau = 150 \rightarrow 225$ ns. The spectrum recorded at 225 ns was compared to simulations due to the transitions again being the strongest. The RTDLDAG calculations described the high energy absorption resonance behavior very well in terms of shape, provided a shift of -0.425 eV was applied. With the shift, the RTDLDAG describes the shape of three resonances well in terms of shape and peak value as shown in Fig. 6.26.

In Section 6.3.7 autoionization calculations were also conducted on doubly ionized thulium but only with 5d decay channels. The inclusion of 6s decay channels in addition to the 5d channels caused the spectra to broaden more than is observed in the experimental spectra at $\Delta \tau = 225$ ns, therefore only 5d decay channels were used. The autoionization calculations were once again performed with different scaling parameters and each of these were compared to the observed spectra at $\Delta \tau = 225$ ns in Fig. 6.27. Each of the spectra was shifted so that the tall peak overlapped with the 34.5 eV peak. There was no clear difference in shape caused by the different scaling
parameters. A scaling of 85% was chosen in order for the synthetic transition to align with the experimentally observed absorption at 34.5 eV in Fig. 6.27 with a maximum shift of 0.5 eV.

The spectrum at $\Delta \tau = 225 \text{ ns}$ seems to be made up a combination of singly and doubly ionized thulium ion states, along with components of $4f^{14}$ and $4f^{12}6s^2$ excited states as shown in Fig. 6.28.

Finally it is clearly evident from Fig. 6.29 that triply ionized thulium was not present in the plasma. The shortest delay in which absorption occurred was $\Delta \tau = 150 \text{ ns}$. The potential of seeing triply ionized thulium could be improved by focusing the the line plasma to a sharper line and therefore a higher power density. The disadvantage to increasing the power density on the target is that thulium tends to emit at these wavelengths at higher power densities. This problem could be fixed by accounting for the emission of thulium when using the Beer-Lambert equation.

### 6.4 fs Reheating of an In Laser Produced Plasma

#### 6.4.1 Introduction

Section 6.4 describes a short experiment involving the reheating of an In laser produced plasma with output of the Odin amplifier of the Quantronix ultrafast laser discussed in Section 2.3. The interaction of the fs laser was observed by detecting In emission at energies that were observed in absorption by Connerade [1]. The experimental set up is described in Section 6.4.2. The results of the fs reheating of the In plasma will be illustrated in Section 6.4.3.

#### 6.4.2 Experimental Set Up for the fs Reheating of an In Laser Produced Plasma

The experimental set up for the fs reheating of an In laser produced plasma is very similar to the DLP set up described in Section 6.2.2. The main difference is that the continuum emitting W rod has been removed. In addition, a new flange system was designed and mounted on the optical axis of the spectrometer which allowed for a CF nipple to be extended out with a fused silica window attached to it. This nipple was used to minimize the time the laser spent traveling in air as described in Section 3.5.2.

The 1-mJ output of the Odin (described in Section 2.3) passed through the Thor and entered the chamber through the nipple window and was brought into focus by a 10 cm lens along the optical axis where the absorbing plasma is formed in Fig. 6.1. The power density of the Odin at the focus was
The In plasma was formed using the Spectron laser described in Section 2.6. The laser was focused to a line plasma using a 10 cm cylindrical lens. A piece of laser burn paper was placed at the focus of the Spectron to allow for an estimate of the spot size of the laser at focus. The burn paper was then photographed in the vicinity of a ruler as shown in Fig. 6.30. The lines of the ruler are clearly visible and yield:

\[
78 \text{ pixels} = 1 \text{ mm}
\]

The focus of the laser depicted by the burn mark on the brown paper near the top of Fig. 6.30 yields a width of 0.3 mm and a length of 16 mm. The Spectron laser had a measured energy of 643 mJ and a temporal FWHM of 18 ns.

\[4 \times 10^8 \text{ Wcm}^{-2}\]

A value for the power density of \(4 \times 10^8 \text{ Wcm}^{-2}\) was obtained using equation 6.1.

The electronic delay system also had to be altered to accommodate the ultrafast laser. The requirement was for the ultrafast laser to arrive at a set delay after the Spectron laser had formed the line plasma. This was achieved by using the trigger for the Darwin pump laser of the Odin to trigger the Quantum Composer box discussed in Section 6.2.2. A Block diagram of the timing system is shown in Fig. 6.31. With the Darwin triggering the Quantum Composer all that remained is to add some delay X to the flashlamp to get the laser pulses to arrive at the desired time with respect to the ultrafast laser, while preserving the required 120 µs delay required for optimum Q-switching. An X is included in Fig. 6.31 for delay A for the Spectron flashlamp because this value was varied. The trigger from
the Darwin seemed to drift with respect to the Odin output, meaning that the delay between the Spectron and the ultrafast laser had to be constantly monitored through the course of the experiment.

![Block timing diagram of the Darwin triggering the Spectron](image)

Figure 6.31: Block timing diagram of the Darwin triggering the Spectron

6.4.3 Results for fs Reheating of an In Laser Produced Plasma.

The linear CCD detector was placed in the same position on the Rowland circle to obtain the same spectrum obtained in Fig. 6.7. The calibration was confirmed by comparing all the neutral absorption lines in the measured spectrum with the published energies [1]. The published lines that agreed with the calibration of the 1-m spectrometer to within 0.035 eV (approximately the resolution of the 1-m spectrometer) are highlighted in Fig. 6.32 with black lines.
With the calibration optimized the next step was to demonstrate the reheating that occurred with the fs interaction with the indium plasma. The effect of reheating on the plasma was observed by recording three measurements.

1. The emission spectrum of the In line plasma only
2. The scattering of the Odin in the spectrometer only
3. The emission of the In line plasma and the Odin together

The sum of measurements 1 and 2 were compared to measurement 3 and where there were differences it was concluded that a reheating interaction had occurred. An altered reheated spectrum was observed with a delay of $\Delta \tau = 500 \text{ ns}$ between the lasers. The Odin was focused into the center of the absorbing line plasma, which was set at a height of $\Delta z = -0.2 \text{ mm}$ from the
optical axis. The normalized spectrum from the scattering of the Odin alone is shown in Fig. 6.33 (a). The normalized sum of both the Odin scattering and the In spectral emission alone are shown in Fig. 6.33 (b). The normalized difference between emission observed when both the Spectron and the Odin were fired at the same time and the sum spectrum illustrated in Fig. 6.33 (b) are illustrated in Fig. 6.33 (c). The red lines labelled with letters illustrate the energies of absorption transitions as observed by either Connerade [1] or Duffy et al. [2].

![Figure 6.33: Spectra showing the effect of reheating of an indium plasma where \( \Delta \tau = 500 \text{ ns}, \Delta z = -0.2 \text{ mm} \) with the ultrafast laser focused at the centre of the indium plasma.](image)

There are very few differences between Fig. 6.33 (a) and Fig. 6.33 (b) in terms of different features except that a number of the features in the Odin scattering in Fig. 6.33 (a) are enhanced in Fig. 6.33 (b) due to In emission lines lying in the same energy region.

There are, however, peaks in Fig. 6.33 (c) that are not visible in either of the the other two spectra. These are believed to be a result of fs-reheating of the In plasma. The letters in the transition labels correspond to a key which can be converted to an observed transition using Table 14. The different
Table 14 will now be explained.

- Key: alphabetical label allocated to a given transition in Fig. 6.33.
- Energy: energy of the published transition (In the case of ref [1] the energies were converted from the published wavelength)
- Lower Term: published ground term for a given transition.
- Excited Term: published excited term of the system (The term in brackets represents the outer 5p term and the term at the end indicates the total term of the system) with the exception of terms from ref [2] where the bracketed term represents the 4d core, followed by the total term of the system.
- Reference: reference number in this chapter representing the author from whom this data was obtained.

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<th>Key</th>
<th>Energy (eV)</th>
<th>Lower Term</th>
<th>Excited Term</th>
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</table>

Table 14: Table showing the transitions identified in Fig. 6.33. All the transitions published by Connerade [1] involve 4d¹⁰5p¹ → 4d⁶5p² and transitions published by Duffy [2] involve 4d¹⁰ → 4d⁶5p¹.
The most clearly enhanced transitions shown in Fig. 6.33 are the following transitions:

\[ \{A, B, D, F, H, N, O, L\} \]

The remaining labelled lines show no effect of enhancement. As yet, no obvious pattern or trend in the enhancement/ non enhancement of the lines in Fig. 6.33 (c) has been discerned. This is obviously a topic for further study.

Having observed the enhancement shown in 6.33 (c) a range of delay times (150, 200, 500, 550, 1000, 1100, 1200 ns ), at two plasma heights (0.1 and 0.2 mm) below the optical axis and two focal spot sizes of the Odin were investigated to see if further enhancement was possible. However further enhancement was not observed.

Figure 6.34: Spectra showing the effect of reheating of an indium plasma where \( \Delta \tau = 1100 \text{ ns}, \Delta z = -0.2 \text{ mm} \) with the ultrafast laser focused 1 mm before the indium plasma.

It is clear from Fig. 6.34 that there is little or no difference between summing the separately taken Odin and the Spectron spectra illustrated in
Fig. 6.34 (b) and the difference between shooting both lasers at the same time and separately, as illustrated in Fig. 6.34 (c).

6.4.4 Conclusions

In Section 6.4.3 two experimental parameters, both delay time between the lasers $\Delta \tau$ and the focal position of the ultrafast laser with respect to the absorbing plasma column, demonstrated a completely different interaction of the fs laser with the plasma. In the case of $\Delta \tau = 500 \text{ns}$ and the laser being focused at the centre of the plasma, there was a clear difference between the summed spectra from both lasers being fired individually and the reheated spectrum. There were a number of observed emission enhancements at energies involving transitions that had previously been published.

The transitions that experienced enhancement, listed in Table 14, were almost all from neutral In, with one from In$^+$ labelled L. Other than these transitions involving $4d \rightarrow 5p$ transitions, there is currently no basis identified as to why these transitions were enhanced and others were not. Clues to this could perhaps be obtained by shooting more neighbouring target elements with published photoabsorption transitions and investigating whether enhancement occurs with similar terms. A variety of different laser conditions, including full energy from the Thor, or different pulse durations could change which states are enhanced. Clearly a lot remains to be done in this area.

The mechanism for this response is speculated to be the fs laser coupling to the 4d electron and ionizing it, followed by a $5p \rightarrow 4d$ emission transition resulting in emission at energies that would normally only be observed in absorption.

The fact that the other experimental conditions yielded no or reduced enhancement may imply that this effect can only be seen either under a specific set of power density conditions or at specific delay times in the lifetime of the neutral In ion.

References


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