$4f$ photoabsorption in Pt II to Pt V

Paddy Hayden, Eric Doyle, Gerry O’Sullivan and Padraig Dunne

School of Physics, University College Dublin, Belfield, Dublin 4, Ireland
Talk Outline

• Speclab @UCD
• Dual Laser Plasma Technique - Intro
• Laser Plasma Continuum
• Dual Laser Plasma Technique – Details
• Photoabsorption of Pt II to Pt V
• Motivation
Atomic, Molecular & Plasma Physics Group Leaders

- **Plasma Physics**
  - Astro NIR Spectroscopy
  - VUV/EUV/Soft x-ray Emission & Absorption Spectroscopy
  - Laser/Matter Interactions
  - Laser Induced Breakdown Spectroscopy

- **PhotoElectron Spectroscopy**
  - Plasma Imaging
  - Soft X-ray Optics
  - Soft X-ray Sources
  - Soft X-ray Detectors
  - Soft X-ray Microscopy

- **Atomic/Ionic Structure**
  - Colliding Plasmas

- **Soft X-ray Optical Modelling**

- **Group Leaders**
  - Padraig Dunne
  - Emma Sokell
  - Paddy Hayden
  - Gerry O’Sullivan
  - Fergal O’Reilly
  - Tom McCormack
## Current Funded Projects

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<td>Spectroscopy for Astro</td>
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Dual Laser Plasma (DLP) Photoabsorption - The Idea

• Use **two** (normally) electronically **synchronised** LPPs
• One acts as the **sample** of atoms, ions or molecules
• The **other** acts as a **source** of continuum radiation
• The **continuum** emission is usually in the Vacuum-UV (VUV) or the Extreme UV (EUV) spectral region
Why Do Photoabsorption?

• Access to **ground state** (non-emitting) atomic and molecular **species** in the sample (vapour, plasma, etc.)
• You can then also **detect** (and potentially quantify) **metastable state species**
• **Photon excitation** => electric dipole excitation => less cluttered and **quite tractable** (from a theoretical perspective) **spectra**
Why Do (X)EUV Photoabsorption?

Why specifically at EUV photon energies?

• Access to more highly charged ions
• Photoionization continua
• Inner-shell/multi-electron excitations

Data relevant to:

• Astrophysical spectra and models
• Laboratory plasma modelling & diagnostics
• Fundamental many-body theory
• Plasma/atomic X-ray laser schemes
• MCF & ICF
• DLP data guides large scale synchrotron expts
Why Do DLP Photoabsorption?
Why Do DLP Photoabsorption?

Costs less than €500M!
Laser Plasma Continuum Source

- Temperature 10 – 100 eV depending on laser power density ($\phi$)
  
  \[ T_e (eV) \approx b A^{1/5} (\lambda^2 \phi)^{3/5} (\lambda^2 \phi) \]

  Average charge \( \approx 0.67 (AT_e)^{1/3} \)

  $\phi$ controls plasma temperature and ion distribution.

- Ions up to \( \sim 20 \) times ionised.

- Electron density \( 10^{19} – 10^{21} \text{ cm}^{-3} \) depending on laser wavelength ($n_{ec} \propto \frac{10^{21}}{\lambda^2} \text{ cm}^{-3}$)

- Hottest at centre, cooler margins - opacity issues

- \( \sim 100 \mu \text{m size} \)

- Duration \( \sim \) laser pulse $\Delta \tau$ (170ps-20ns)

- Expansion velocity \( \sim 10^6 - 10^7 \text{ cm s}^{-1} \)
Laser Plasma Continua – History & Physical Origin

Spectrum consists of:

- **lines** (bound-bound transitions), because of high density, lines from high n states are usually not seen
- **recombination radiation** (bound–free transitions)
- **bremsstrahlung** (free–free)
- In some cases lines cluster together to form an **UTA** (unresolved transition array)
Laser Plasma Continua – History & Physical Origin

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- In some cases lines cluster together to form an **UTA** (unresolved transition array)

*Free-Free and Free-Bound* processes yield continuum emission spectra suitable for application in absorption spectroscopy
Laser Plasma Continua – History & Physical Origin

Fig. 1. (a) Absorption spectrum of xenon from 80 to 200 Å. The xenon pressure in the spectrograph was 0.05 Torr, and the number of laser pulses used was 30. For details of the xenon spectrum in this region see Madden and Codling. The unmarked weak lines near 200 Å are due to 0 v. Oxygen present in the target gives rise to some emission lines as well. (b) The ytterbium continuum from 60 to 100 Å. The number of laser shots was 20. As in (a), the spectrum was obtained on a Kodak SC5 plate.

Short wavelength continua emitted from laser produced rare-earth (and neighbouring element) plasmas are predominantly line-free in origin

For a review of the early years including applications in photoabsorption spectroscopy see:


Fig. 2 (a) EUV emission spectrum of an aluminum oxide plasma showing the predominance of lines from O\(^+\) within the 54–64 eV photon energy range; (b) and (c) Continuum emission from a tungsten plasma in the 30- and 140-eV spectral ranges.

But Why is No Line Emission Observed?

• Line emission is due to complex $4d \rightarrow 4f$ transition arrays in (typically) 7 – 20 times ionized atoms:

$$4d^n 5s^q 5p^s 4f^m \rightarrow 4d^{n-1} 5s^q 5p^s 4f^{m+1}, q+s = r+t$$

• Furthermore $4f/5p$ and $4f/5s$ degeneracy and level crossing gives rise to overlapping bands of low-lying configurations, most of which are populated in the ca. 10 - 100 eV plasma

• Result – the summed oscillator strength for each $4d - 4f$ (XUV) array is spread out over a supercomplex of transitions producing bands of unresolved pseudo continua (the UTA’s) superimposed on the background continuum

• A UTA generally has too many lines to identify individual transitions and the linewidth > line separation. Both the energy level and spectral distributions can be parameterised statistically in terms of moments of the array (Bauche, and Bauche-Arnoult Phys Scr T40, 58, 1992)

• In addition, strong emission lines from simple $4d - 4f$ transition arrays, e.g., $4d^{10} - 4d^9 4f$ in Xe-like ions, are washed out by plasma opacity
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Spectra of elements with $Z > 62$ emit mostly continuum. The spectrum of Sm (the most extreme case) is essentially line free from 3-200 nm.
Benefits of Laser Plasma Continua

• Ease of production
• Ease of location
• Purity (spectral)
• Wide spectral coverage - (4 – 200 nm)
• Small emitting size (almost point-like, radiography & microscopy)
• Short pulse duration (< 100 ps - 50 ns)
• Easy synchronisation (Optical or Electro-optic)
• Shot to shot intensity reproducibility > 95%
• ~$10^{14}$ Soft X-ray Photons/sec/0.2% Bandwidth/2π sr

Economy, Ease of Use & Versatility
DLP Photoabsorption - The Set-up

Absorbing target

Backlighting target

Optical axis

Cylindrical lens

Plano-convex lens
DLP Photoabsorption - The Set-up

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Plano-convex lens
DLP Photoabsorption - The Set-up

- Absorbing target
- Backlighting target
- Optical axis
- Plano-convex lens
- Cylindrical lens
- Record $I_0$
DLP Photoabsorption - The Set-up

Absorbing target

Backlighting target

Optical axis

Cylindrical lens

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DLP Photoabsorption - The Set-up

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DLP Photoabsorption - The Set-up

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- Backlighting target
- Cylindrical lens
- Plano-convex lens
- Optical axis

Record $I_1$
DLP Photoabsorption - The Set-up

Absorbing target

Backlighting target

Optical axis

Cylindrical lens

Plano-convex lens
DLP Photoabsorption - The Set-up

Absorbing target

Δx, probe different regions of the plasma

Backlighting target

Optical axis

Cylindrical lens

Plano-convex lens
DLP Photoabsorption - The Set-up

- Absorbing target
- Backlighting target
- Optical axis
- Cylindrical lens
- Plano-convex lens

Δx, probe different regions of the plasma
DLP Photoabsorption - The Set-up

Absorbing target

Backlighting target

Optical axis

Plano-convex lens

Cylindrical lens
DLP Photoabsorption - The Set-up

Absorbing target

Backlighting target

Optical axis

$\Delta t$, from ns to $\mu$s

Plano-convex lens

Cylindrical lens
DLP Photoabsorption - The Set-up

- Absorbing target
- Backlighting target
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- $\Delta t$, from ns to $\mu$s
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DLP Photoabsorption - The Set-up

- Absorbing target
- Backlighting target
- Optical axis
- Cylindrical lens
- Plano-convex lens
- Record $I$

$\Delta t$, from ns to $\mu$s
DLP Photoabsorption - The Set-up

Backlighting Plasma: $I_0$
Absorbing Plasma: $I_1$
Both: $I = I_0 \exp(-\sigma NL) + I_1$

\[
\sigma -\text{cross section} \\
N - \text{number density} \\
L - \text{path length}
\]

Relative Absorption Cross Section:

\[
\sigma NL = \ln \left( \frac{I_0}{I - I_1} \right)
\]
DLP Photoabsorption - The Set-up

Nd:YAG laser pulses

Plano-convex lens

Cylindrical lens

Continuum backlighting target (point plasma)

Absorbing target (line plasma)

To spectrometer

$\Delta t$

$\Delta x$
DLP Photoabsorption - The Set-up

Species Flexibility:
Neutral/Multiply-charged/
Refractory Elements

Nd:YAG laser pulses \( \Delta t \)

Plano-convex lens

Cylindrical lens

Continuum backlighting target (point plasma) \( \Delta x \)

Absorbing target (line plasma)

To spectrometer
DLP Photoabsorption - The Set-up

Species Flexibility:
Neutral/Multiply-charged/
Refractory Elements

No tuning required
No vapour required

Nd:YAG laser pulses
Δt

Plano-convex lens

Cylindrical lens

Continuum backlighting
target (point plasma)

Absorbing target
(line plasma)

To spectrometer
DLP Photoabsorption - The Set-up

Species Flexibility:
Neutral/Multiply-charged/Refractory Elements

Nd:YAG laser pulses

$\Delta t$, $\Delta x$, $\phi$ (W/cm$^2$) → Species choice

No tuning required
No vapour required

Plano-convex lens

Cylindrical lens

Continuum backlighting target (point plasma)

Absorbing target (line plasma)

To spectrometer
DLP Photoabsorption - The Set-up

Species Flexibility: Neutral/Multiply-charged/Refractory Elements

No tuning required
No vapour required

\( \Delta t, \Delta x, \phi \text{ (W/cm}^2\text{)} \)
\( \rightarrow \) Species choice

Continuum backlighting target (point plasma)
Absorbing target (line plasma)
To spectrometer

Isonuclear Sequences
Isoelectronic Sequences
DLP Photoabsorption - The Set-up

Species Flexibility:
Neutral/Multiply-charged/
Refractory Elements

\( \Delta t, \Delta x, \phi \) (W/cm\(^2\))
\( \rightarrow \) Species choice

Relative Absorption Cross Section:
\[
\sigma_{NL} = \ln\left( \frac{I_0}{I - I_1} \right)
\]
DLP Photoabsorption Example - Te

$\Delta x = 2 \text{ mm}$

$\phi = 5 \times 10^9 \text{ W/cm}^2$ (cylindrical lens)

• Neutral Te dominates at the time delays ($\Delta t$) shown.

• Discrete structure arising from $4d - np$ transitions


DLP Photoabsorption Example - Te

\[ \Delta x = 2 \text{ mm} \]

\[ \phi = 5 \times 10^9 \text{ W/cm}^2 \text{ (cylindrical lens)} \]

- \( \text{Te}^+ \) dominates at \( \Delta t \)'s shown.

- Discrete structure arising from \( 4d - np \) transitions


DLP Photoabsorption Example - Te

\[ \Delta x = 0 \text{ mm} \]
\[ \phi = 6 \times 10^{11} \text{ W/cm}^2 \text{ (spherical lens)} \]

- Te\(^{2+}\) dominates at \(\Delta t\)'s shown.

- Discrete structure arising from \(4d - np\) transitions

DLP Photoabsorption Example - Te

$\Delta x = 0 \text{ mm}$

$\phi = 6 \times 10^{11} \text{ W/cm}^2$ (spherical lens)

- Te$^{2+}$ dominates at $\Delta t$'s shown.
- Discrete structure arising from $4d - np$ transitions

EUV Photoabsorption of Pt

$\Delta t = 100 \text{ ns}$

$\Delta x = 2 \text{ mm}$

$\phi = 3 \times 10^9 \text{ W/cm}^2$

EUV Photoabsorption of Pt

$\Delta x = 0 \text{ mm}$

$\phi = 5 \times 10^{10} \text{ W/cm}^2$

$\Delta t = 60 \text{ ns}$

Identification of Absorption Features

• Isoelectronic sequences in **Au previously studied** Su M G, Dong C Z, Murphy N and O’Sullivan G 2009 *Phys. Rev. A* **79** 042507


• **Focus on** $4f$ **transitions.**

• $5d$ and $6s$ orbitals are near-degenerate

• At EUV energies, many of the **upper levels** lie well above the ionisation potential. This facilitates autoionisation, which promotes **significant** transition line broadening by reducing the lifetime of the excited levels.
Cowan’s Codes Calculations

- **Initial and final configurations** where $m = 9, 8, 7, \text{ and } 6$ in $\text{Pt}^+, \text{Pt}^{2+}, \text{Pt}^{3+}$ and $\text{Pt}^{4+}$ respectively. $n$ is the principal quantum number with values of $5, 6, \text{ and } 7$, and $\delta$ is the ejection energy of a free electron of angular momentum $p, f, h, \text{ or } k$.

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<th>Initial</th>
<th>Discrete</th>
<th>Final</th>
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<td>[Xe] $4f^{14} 5d^m$</td>
<td>[Xe] $4f^{13} 5d^m \text{ nd}$</td>
<td>[Xe] $4f^{14} 5d^{m-1} + \delta(p,f,h,k)$</td>
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<tr>
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<td>[Xe] $4f^{13} 5d^{m-1} 6s \text{ nd}$</td>
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<td>[Xe] $4f^{13} 5d^{m-2} 6s^2 \text{ nd}$</td>
<td>[Xe] $4f^{14} 5d^{m-3} 6s 6d + \delta(p)$</td>
</tr>
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</table>
Cowan’s Codes Calculations

- $4f^{13} 5dm \text{ nd}$
- $4f^{13} 5d^{m-1} 6s \text{ nd}$
- $4f^{13} 5d^{m-2} 6s^2 \text{ nd}$

**Energy (eV)**

- Pt II
  - 3396 terms
  - $4f^{14} 5dm$
- Pt III
  - 9332 terms
  - $4f^{14} 5d^{m-1} 6s$
- Pt IV
  - 16886 terms
  - $4f^{14} 5d^{m-2} 6s^2$
- Pt V
  - 22614 terms
Identification of Absorption Features

- **Transitions** between each permutation of initial and final states were calculated, and subsequently broadened by decay to the continuum.

- Each transition **convolved** with a Lorentzian profile based on **transition widths** derived from the autoionisation lifetime, or else instrumental broadening.

- **Population** ratios of the ground states estimate by Boltzmann distribution.

- Contributions from **each ion stage were weighted** by a factor derived from a collisional-radiative **model** Colombant D and Tonon G F 1973 *J. Appl. Phys.* **44** 3524–37
Simulated Cross Section at $T_e = 9.0$ eV

Weighted Pt Data - Pt$^+$: 1.4%, Pt$^{2+}$: 19.1%, Pt$^{3+}$: 57.4%, Pt$^{4+}$: 20.7%, Pt$^{5+}$: 1.3%,

Cross Section [Mb]

Photon Energy [eV]
Compare to Experimental Epectra

\[ \Delta t = 100 \text{ ns} \]

\[ \Delta t = 200 \text{ ns} \]

\[ \Delta t = 300 \text{ ns} \]

56 135002

\[ \Delta x = 2 \text{ mm} \]

\[ \phi = 3 \times 10^9 \text{ W/cm}^2 \]
4\(f\) Photoabsorption in Pt II to Pt V

Broad peaks between 85 eV and 110 eV are \(4f \rightarrow 6d, 7d\) transition arrays, which move to higher energies with increasing ionisation.

\(\Delta t = 100 \text{ ns}\)

\(\Delta t = 200 \text{ ns}\)

\(\Delta t = 300 \text{ ns}\)


\(\Delta x = 2 \text{ mm}\)

\(\phi = 3 \times 10^9 \text{ W/cm}^2\)
4f Photoabsorption in Pt II to Pt V

Δt = 100 ns

Δt = 200 ns

Δt = 300 ns

Prominent features in the regions of 70 eV and 72 eV due to $4f \rightarrow 5d$ transitions


$\Delta x = 2$ mm

$\phi = 3 \times 10^9$ W/cm$^2$
The features identified sit on a continuum-like absorption feature. This falls off with increasing energy between 70 and 110 eV.
RTDLDA Calculation

- A many-body relativistic time dependent local density approximation (RTDLDA) calculation reproduces the form of the continuum absorption.


Motivation

• The rapid neutron capture process (the r-process)
  – least understood element formation
  – makes half of heavy elements
Motivation

- **Kilonova** neutron star mergers are a prime candidate
- First feature identified, **strontium** (Watson et al. 2019, Nature)
- Missing **atomic data** (line lists and collision strengths) for the **heavy elements**
Motivation

HEAVYMETAL
HOW NEUTRON STAR MERGERS MAKE HEAVY ELEMENTS

ERC Synergy
## Motivation

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<th>Node/PI</th>
<th>Copenhagen/Darach Watson</th>
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<th>Belfast/Stuart Sim</th>
<th>Dublin/Padraig Dunne</th>
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<td><strong>Team</strong></td>
<td>Daniele Malesani &amp; Johan Fynbo</td>
<td>Oliver Just &amp; Gabriel Martínez-Pinedo</td>
<td>Connor Ballance &amp; Cathy Ramsbottom</td>
<td>Paddy Hayden, Tom McCormack, Emma Sokell, Fergal O’Reilly</td>
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<tr>
<td><strong>Expertise</strong></td>
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<td>Merger simulations and nucleosynthesis</td>
<td>Radiative Transfer &amp; Atomic Structure</td>
<td>Experimental Atomic Spectroscopy</td>
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**Starting September 2023**

**Dublin**: 2 Post Doc + 2 PhD  
**Belfast**: 2 Post Docs + 3 PhD  
**Darmstadt**: 1 Post Doc + 4 PhD  
**Copenhagen**: 1 Postdoc + 2 PhD
Acknowledgements

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• **UCD School of Physics Mechanical and Electronic Workshops**
Future Work

Some 6\textsuperscript{th} row transition metals