$4f$ photoabsorption in Pt II to Pt V

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

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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

PhotoElectron Spectroscopy

Astro NIR Spectroscopy

Plasma Imaging

Soft X-ray Optics

VUV/EUV/Soft x-ray Emission & Absorption Spectroscopy

Padraig Dunne

Soft X-ray Sources

Emma Sokell

Soft X-ray Detectors

Laser/Matter Interactions

Fergal O'Reilly

Paddy Hayden

Soft X-ray Microscopy

Laser Induced Breakdown Spectroscopy

Colliding Plasmas

Gerry O'Sullivan

Tom McCormack

Atomic/Ionic Structure

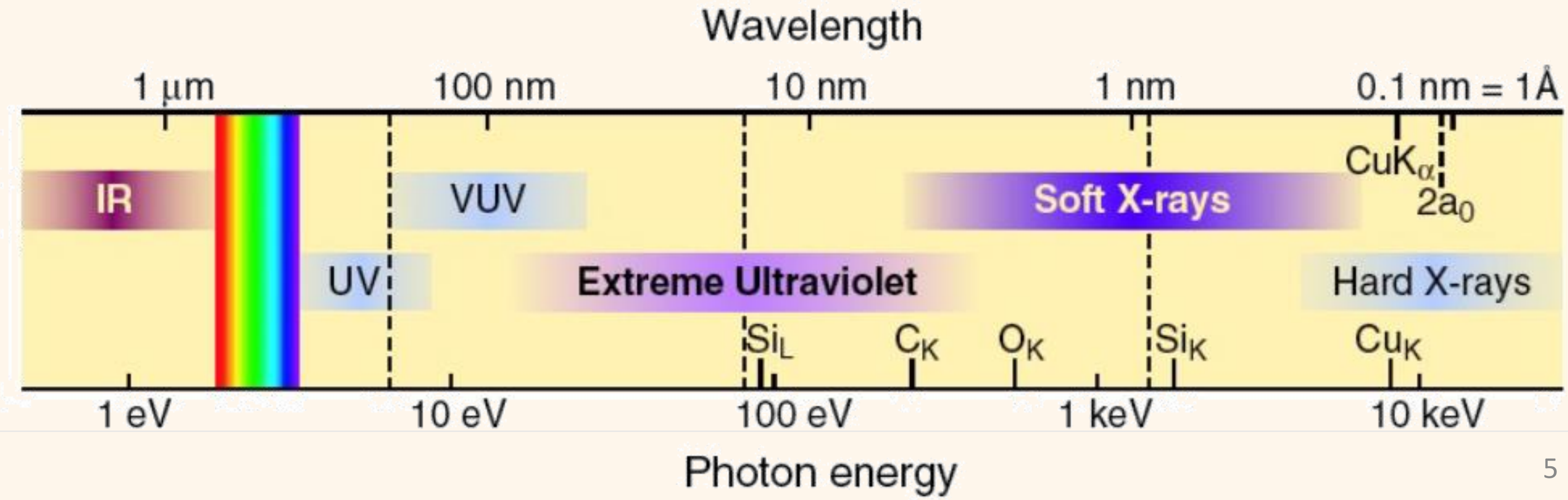
Soft X-ray Optical Modelling

Current Funded Projects

Researcher	Funder	Project Areas	
Dr Ben Delaney	Science Foundation Ireland	LPP Emission/Light Sources/Photoelec/ions	
Eric Doyle	Irish Research Council	Soft x-ray Absorption	Vis/IR spec
Xiongfei Bai	Chinese Scholarship Council/UCD	Colliding Plasmas	Plasma Modelling
Nicholas Wong	Science Foundation Ireland	Photoelectron/ion spec	Light Sources
Kevin Mongey	Science Foundation Ireland	LPP Emission	Light Sources
Ruairí Brady	Science Foundation Ireland	LPP Emission	Light Sources
Martina Donnellan	Irish Research Council/SiriusXT	Light Sources	LPP Emission
Ross Murray	Sustainable Energy Authority of Ireland (SEAI) and ESB	LIBS/Materials analysis	Vis/IR spec
Donnchadh O'Mahony	Irish Research Council/SiriusXT	Optical Modelling	Microscopy
Kirsten Dowd	Irish Research Council/Intel	LPP Absorption - Astro	Vis/IR spec
Eoin Fagan	UCD Physics	LPP Absorption	Vis/IR/Soft Xray
2023	Funding for 2 new PhD students and 2 new Post Docs	Spectroscopy for Astro	Vis/IR/Soft Xray

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?

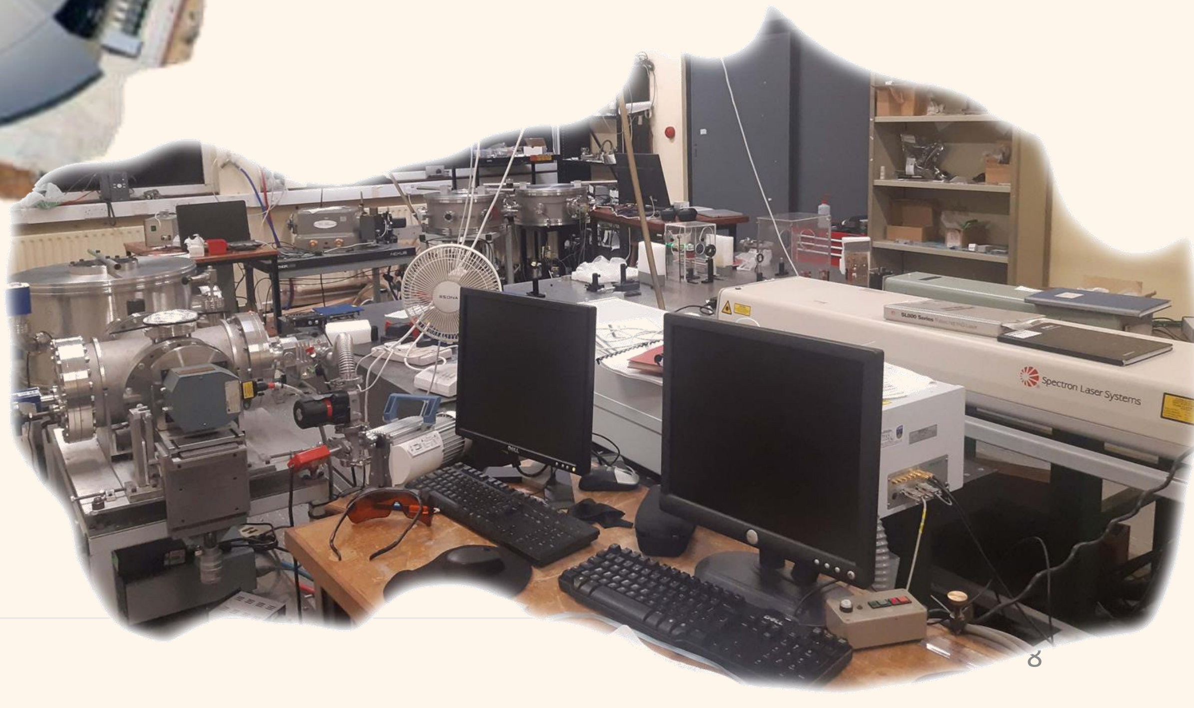
Why specifically at EUV photon energies?

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

Data relevant to:

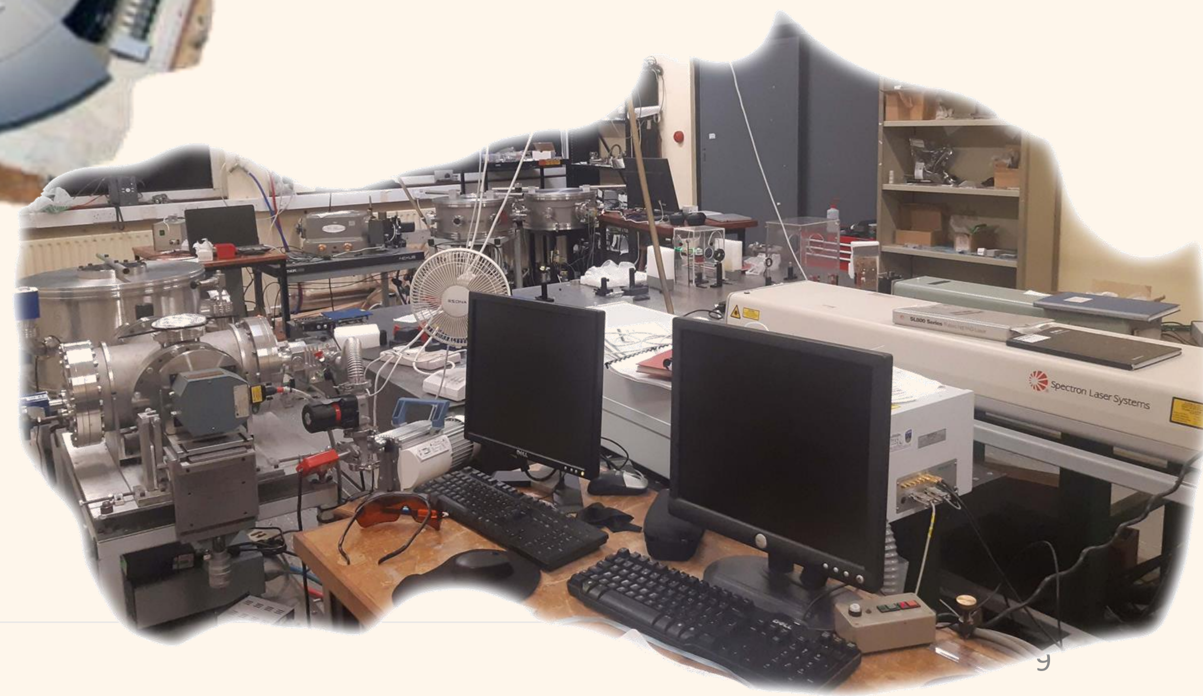
- Astrophysical spectra and models
- Laboratory plasma modelling & diagnostics
- Fundamental many-body theory
- Plasma/atomic X-ray laser schemes
- MCF & ICF
- DLP data guides large scale synchrotron expts

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 (ϕ)

  $T_e(eV) \approx bA^{1/5}(\lambda^2\phi)^{3/5}(\lambda^2\phi)$

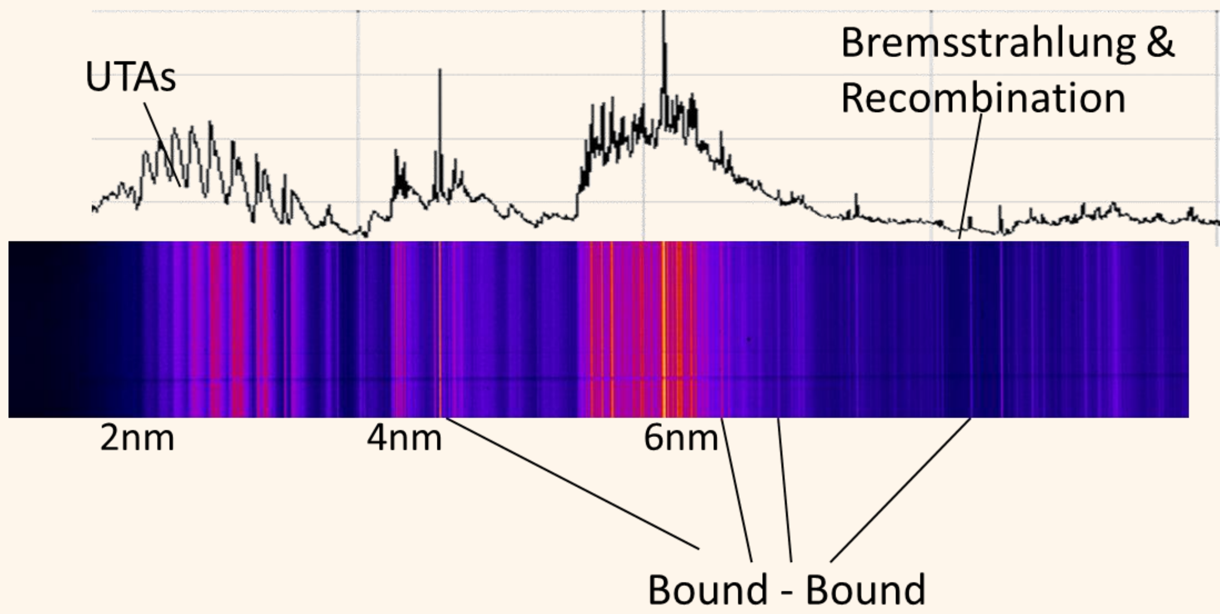
  Average charge $\approx 0.67(AT_e)^{1/3}$

  ϕ controls plasma temperature and ion distribution.
- Ions up to ~ 20 times ionised.
- Electron density $10^{19} – 10^{21}$ cm^{-3} depending on laser wavelength ($n_{ec} \propto \frac{10^{21}}{\lambda^2}$ cm^{-3})
- Hottest at centre, cooler margins - opacity issues
- ~ 100 μm size
- Duration ~ laser pulse Δτ (170ps-20ns)
- Expansion velocity ~ 10^6 - 10^7 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)



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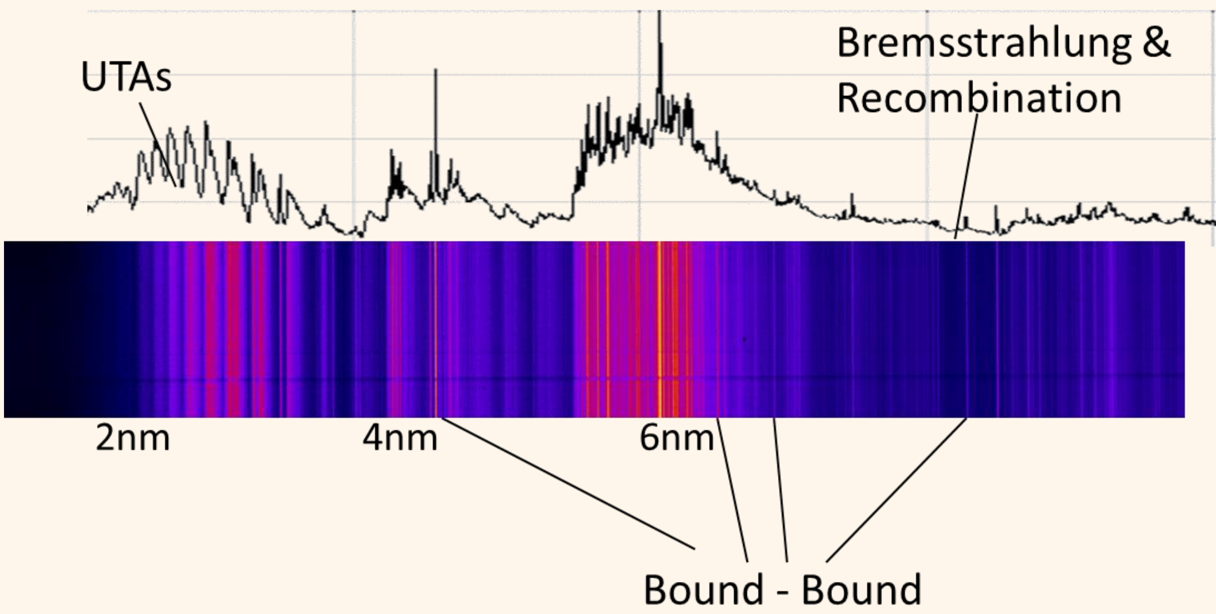
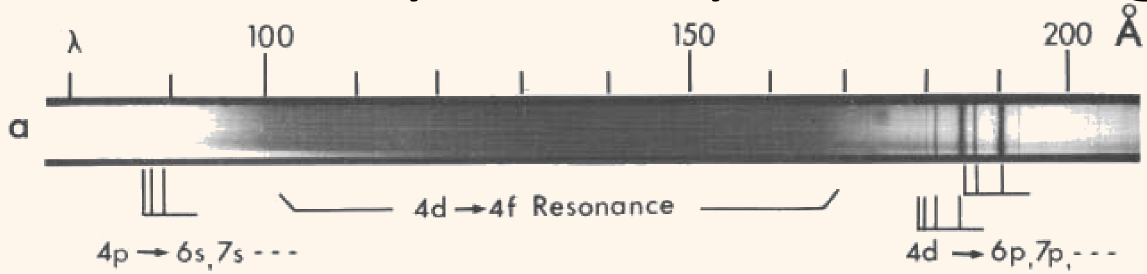


Free-Free* and *Free-Bound **processes yield continuum emission spectra suitable for application in absorption spectroscopy**

Laser Plasma Continua – History & Physical Origin



P K Carroll et al., *Optics Letters* **2**, pp72-74 (1978)

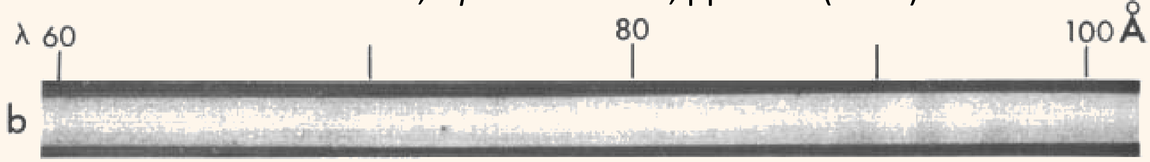


Fig. 1. (a) Absorption spectrum of xenon from 80 to 200 Å. The xenon pressure in the spectrograph was 0.05 Torr, and the number of laser pulses used was 30. For details of the xenon spectrum in this region see Madden and Codling.[5] The unmarked weak lines near 200 Å are due to O V. Oxygen present in 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.

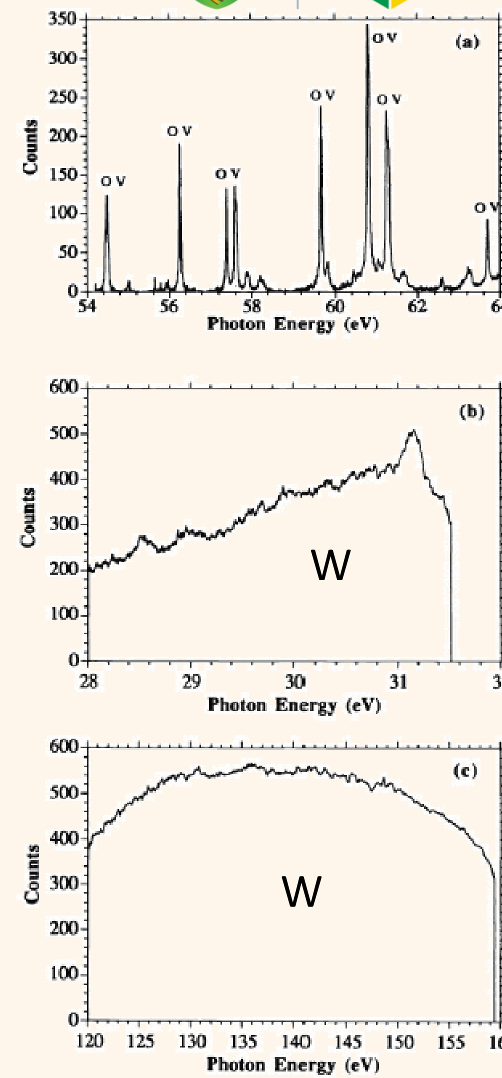


Fig. 2 (a) EUV emission spectrum of an aluminum oxide plasma showing the predominance of lines from O^{4+} in the 54→64 eV photon energy range. (b) and (c) Continuum emission from a tungsten plasma in the 30- and 140-eV spectral ranges.

E T Kennedy et al., *Optical Engineering* **33**, pp3894-3992 (1994)

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:

1. J T Costello et al., *Physica Scripta* **T34**, 77 (1991)
2. P Nicolosi et al., *J. Phys. IV* **1**, 89 (1991)

But Why is No Line Emission Observed?

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

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

- 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

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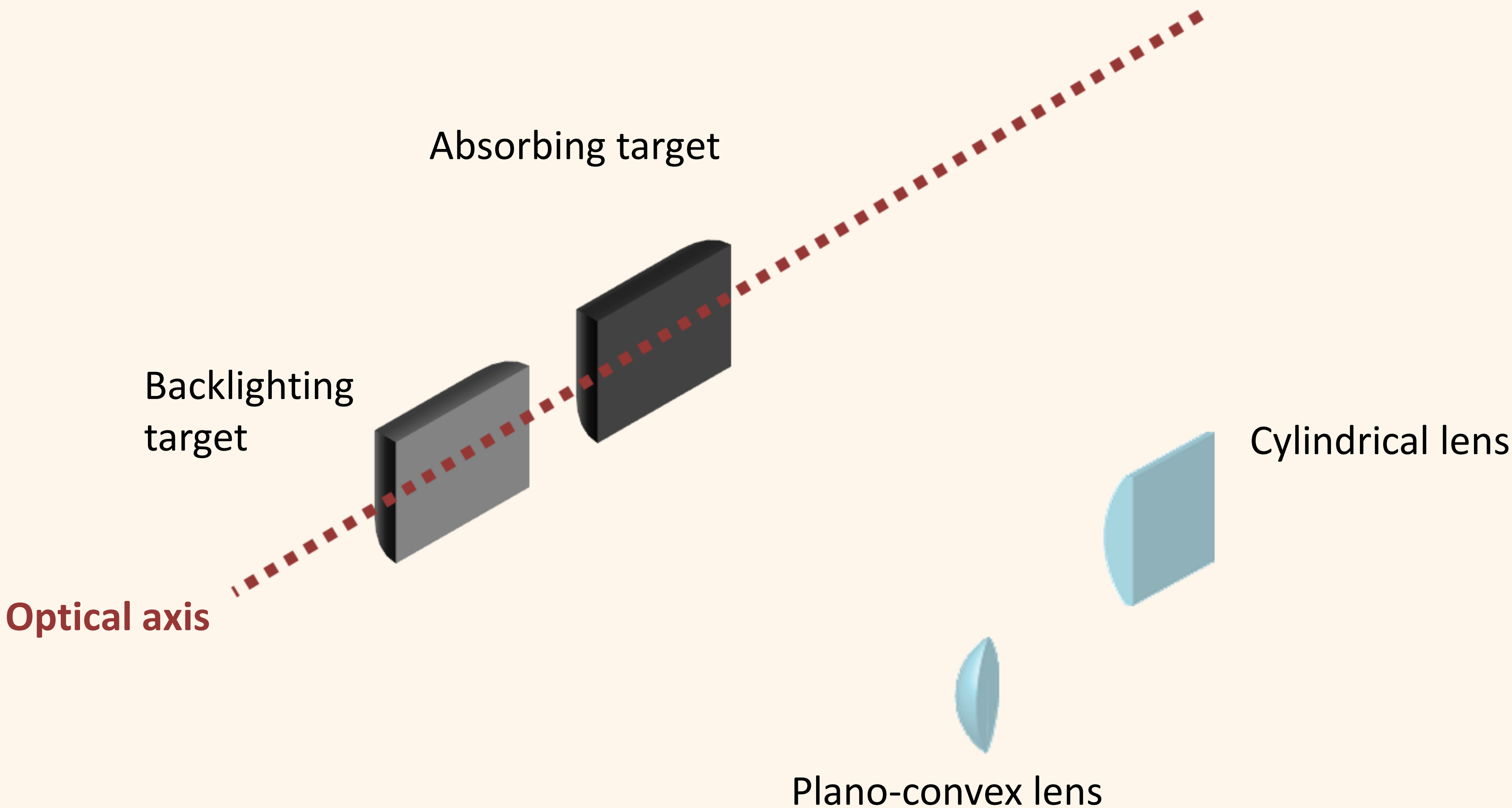
Economy, Ease of Use & Versatility

DLP Photoabsorption - The Set-up



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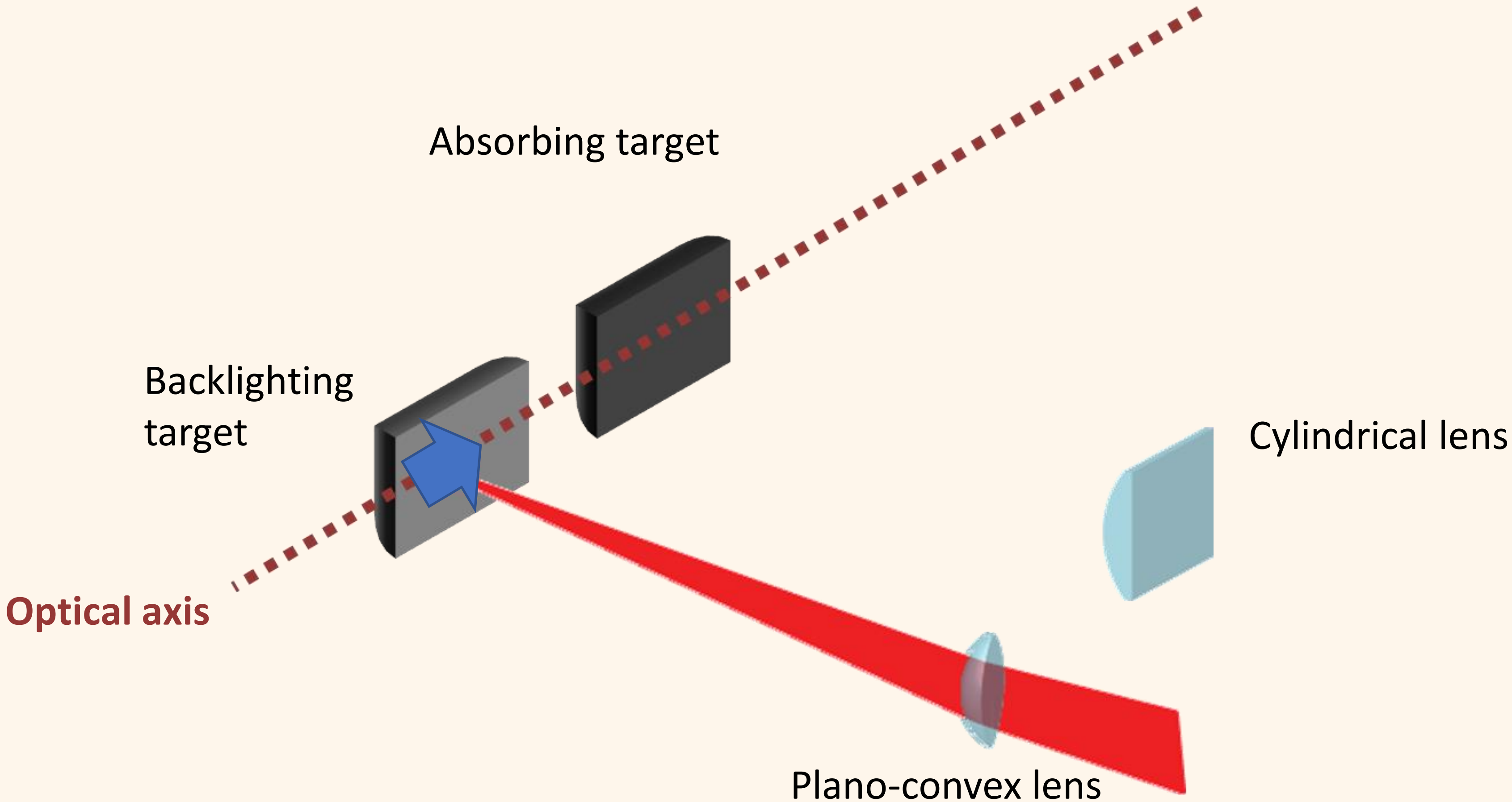

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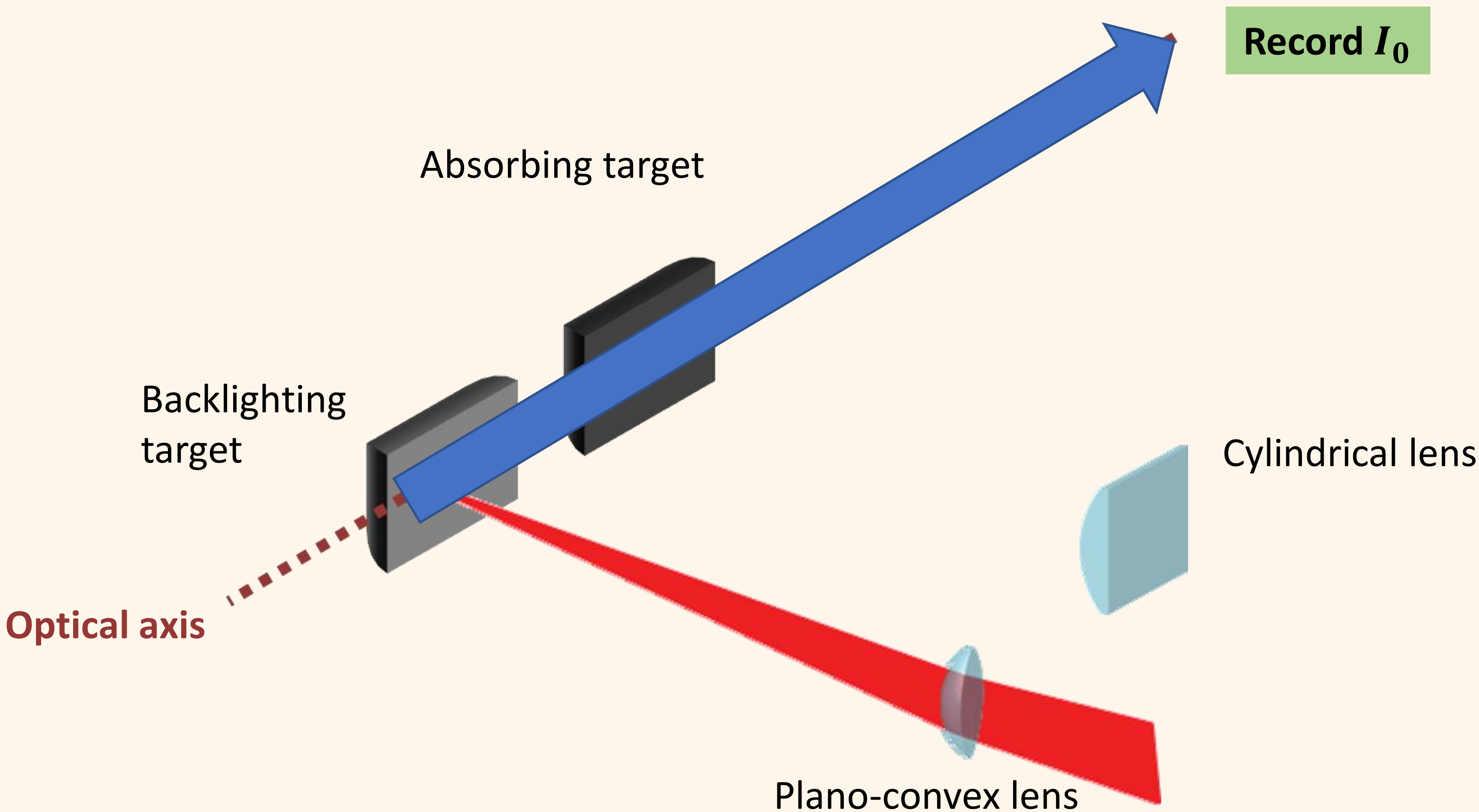

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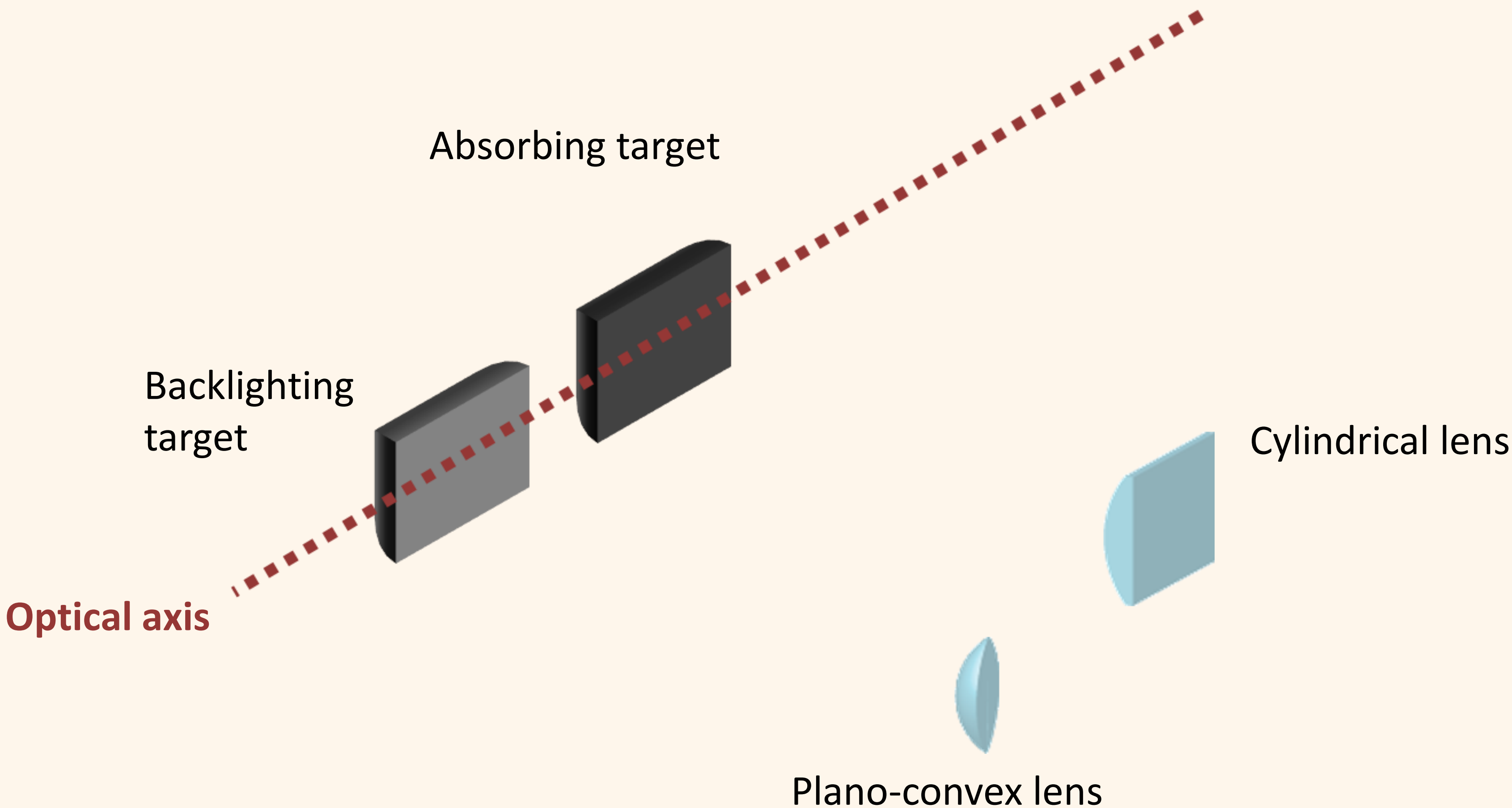

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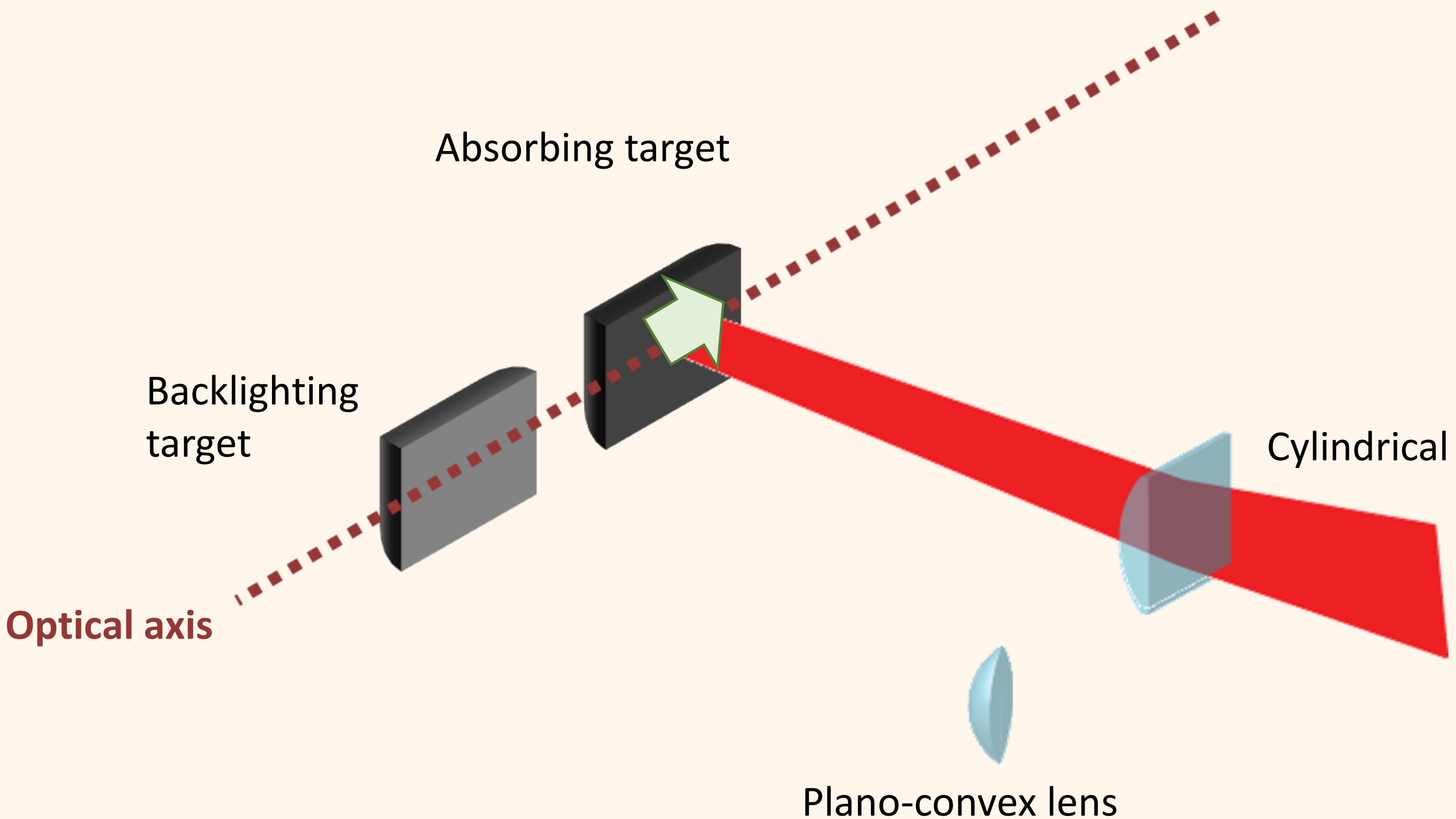

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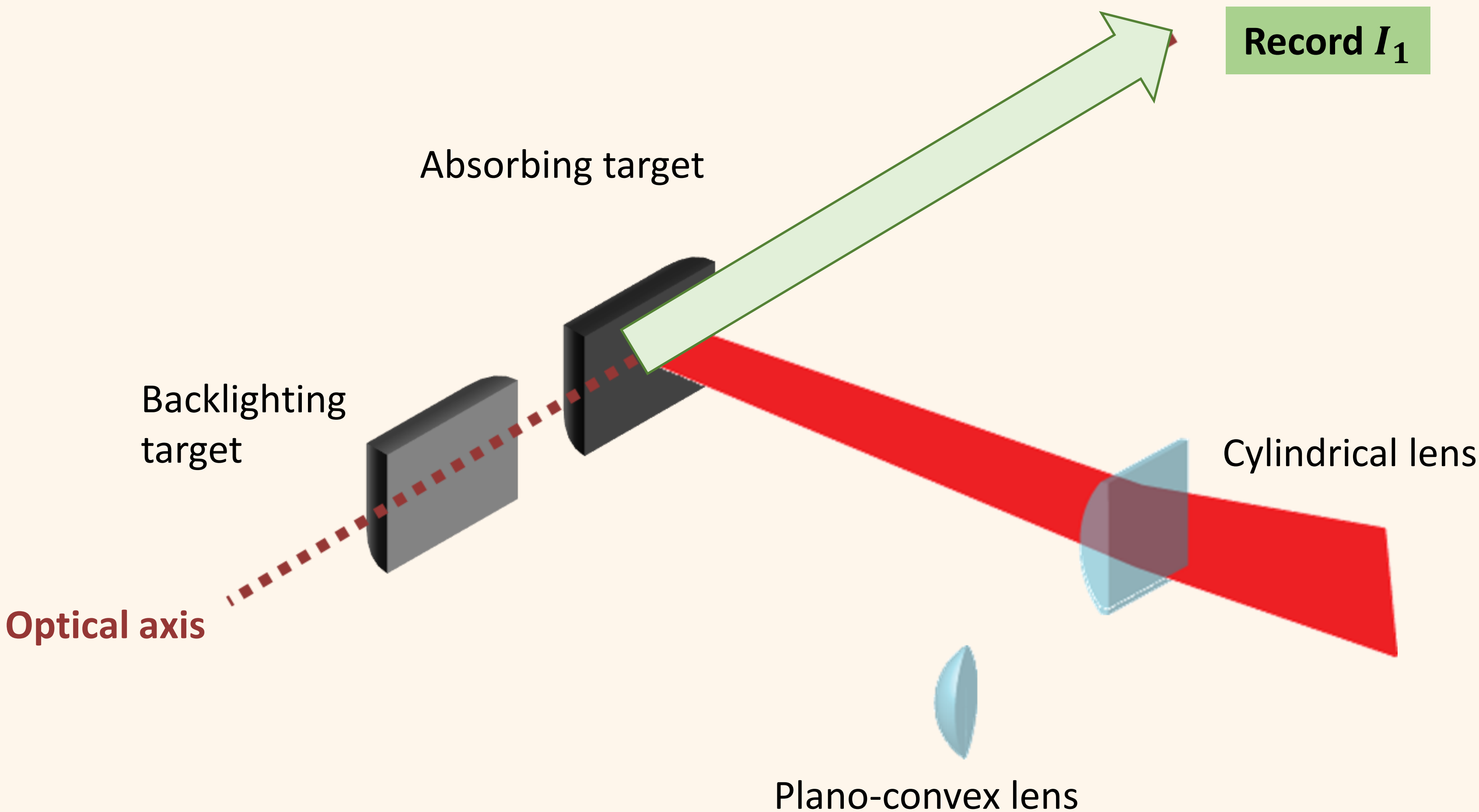

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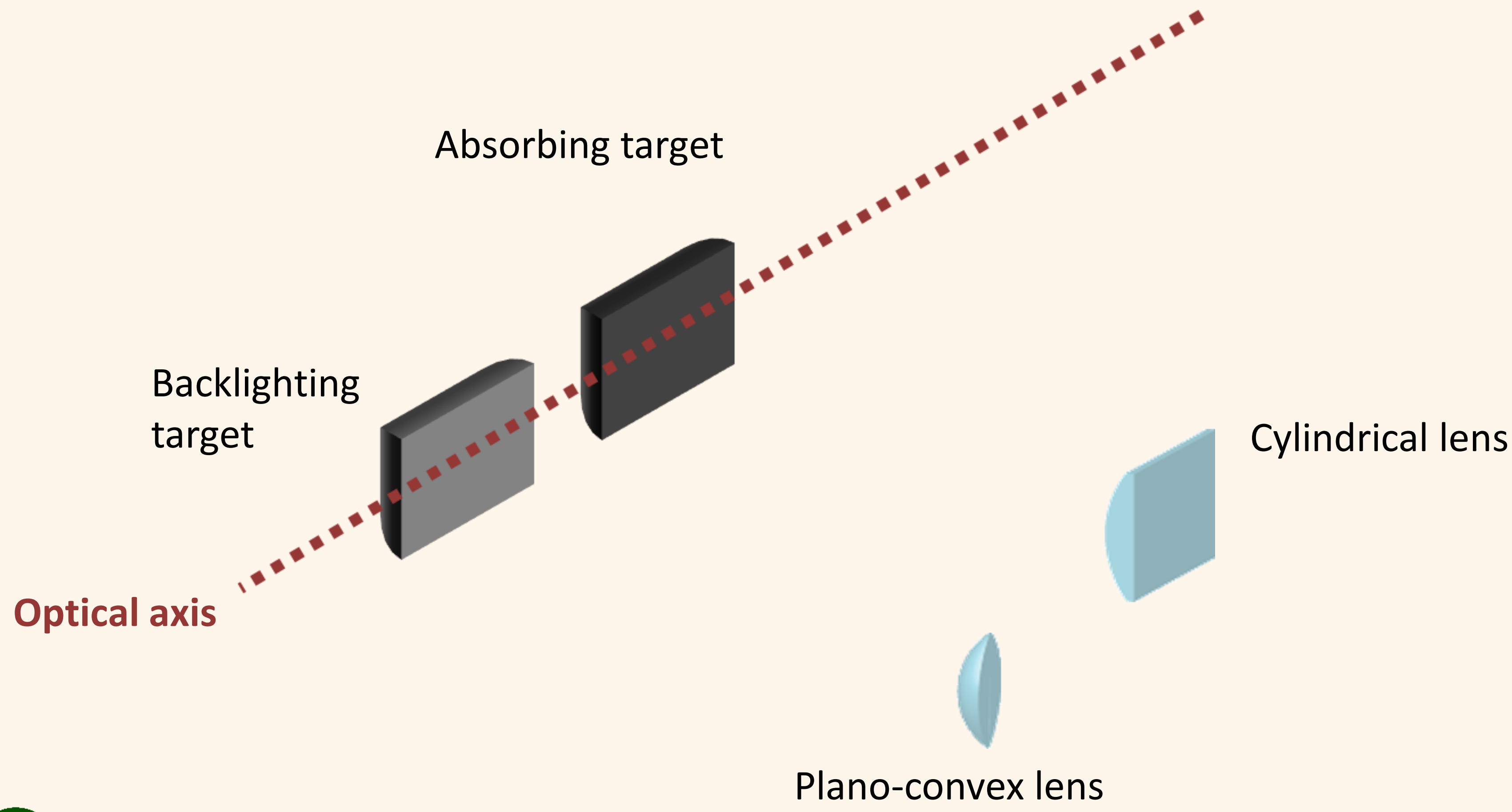

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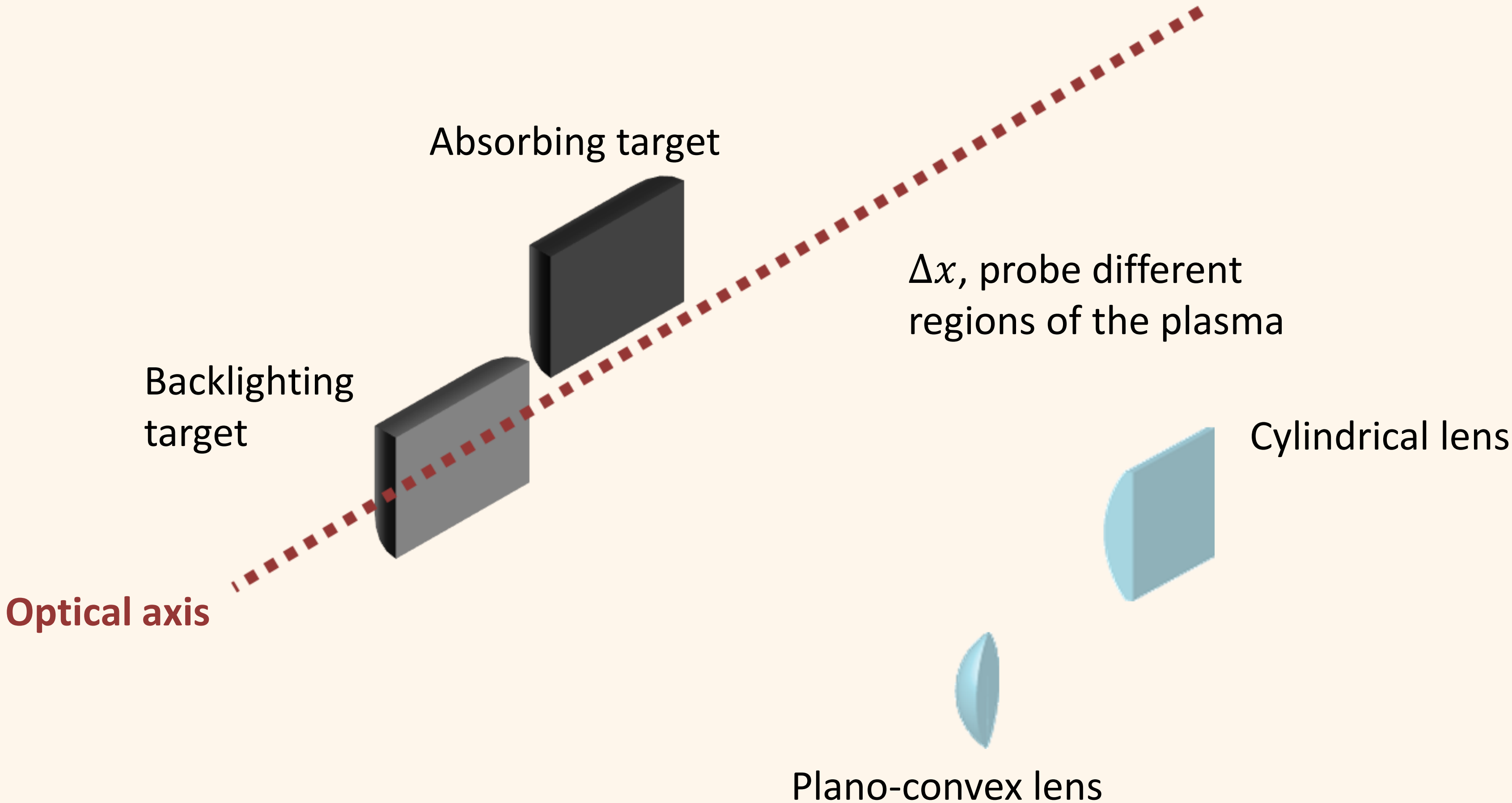

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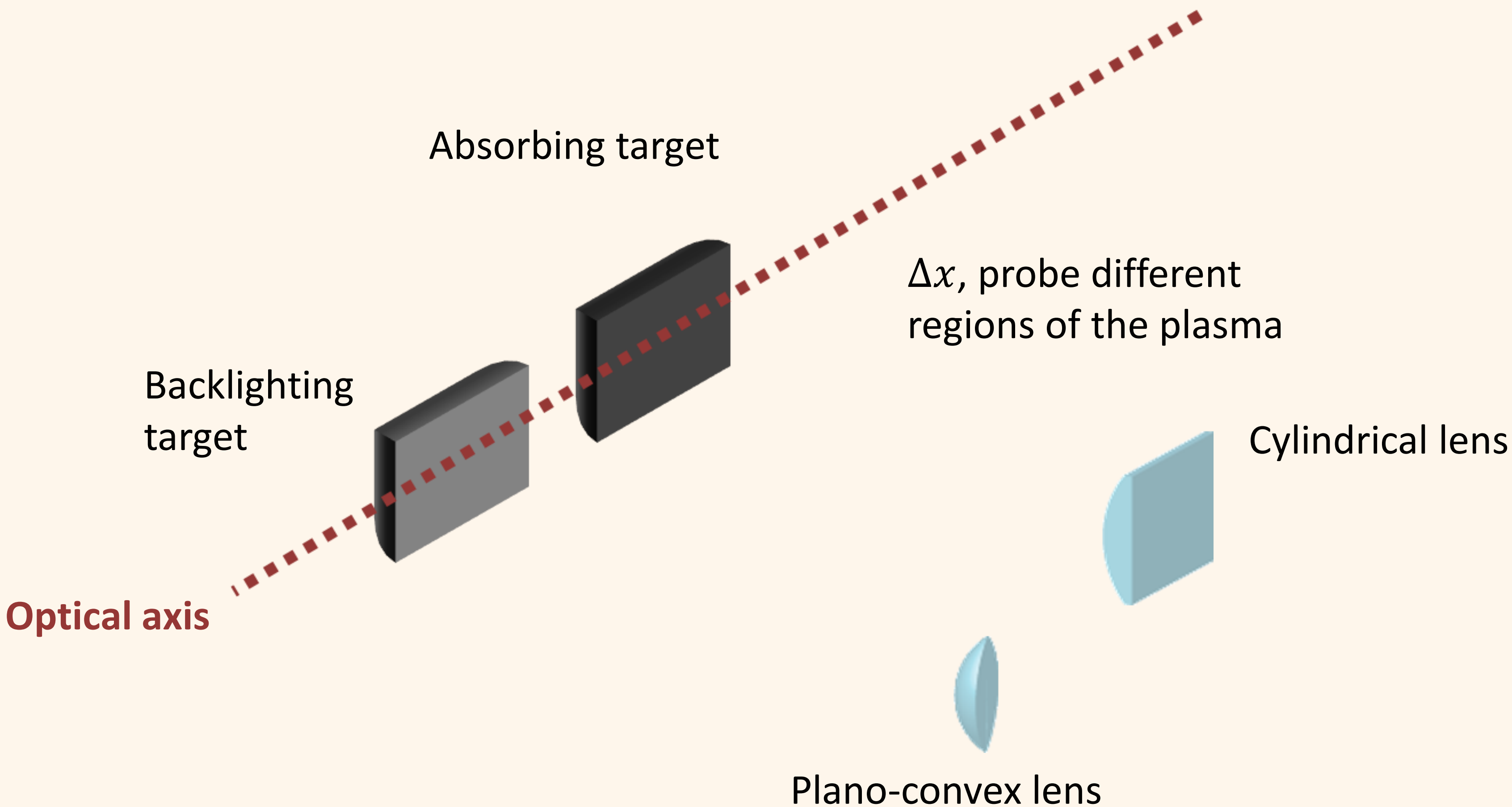

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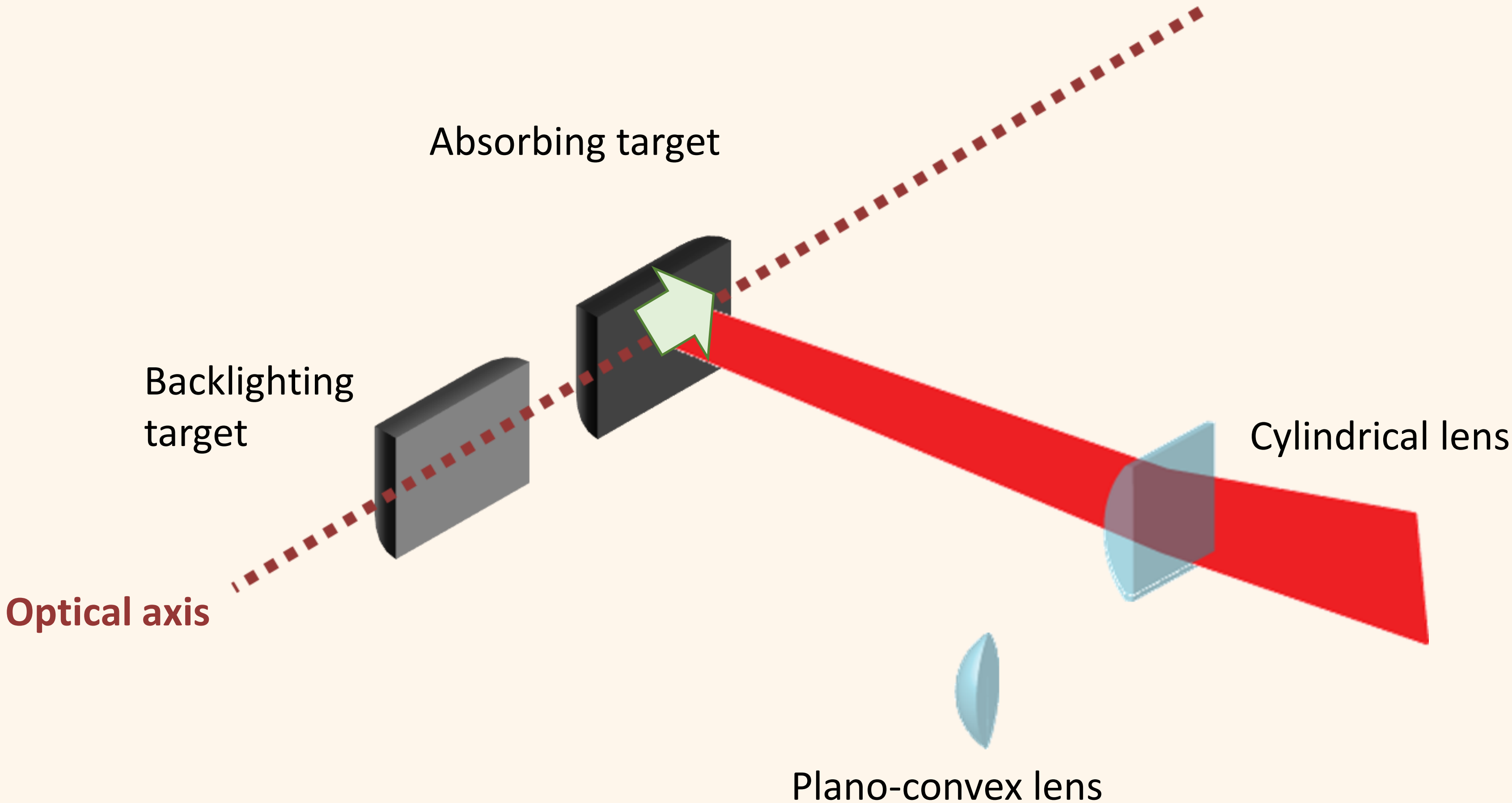

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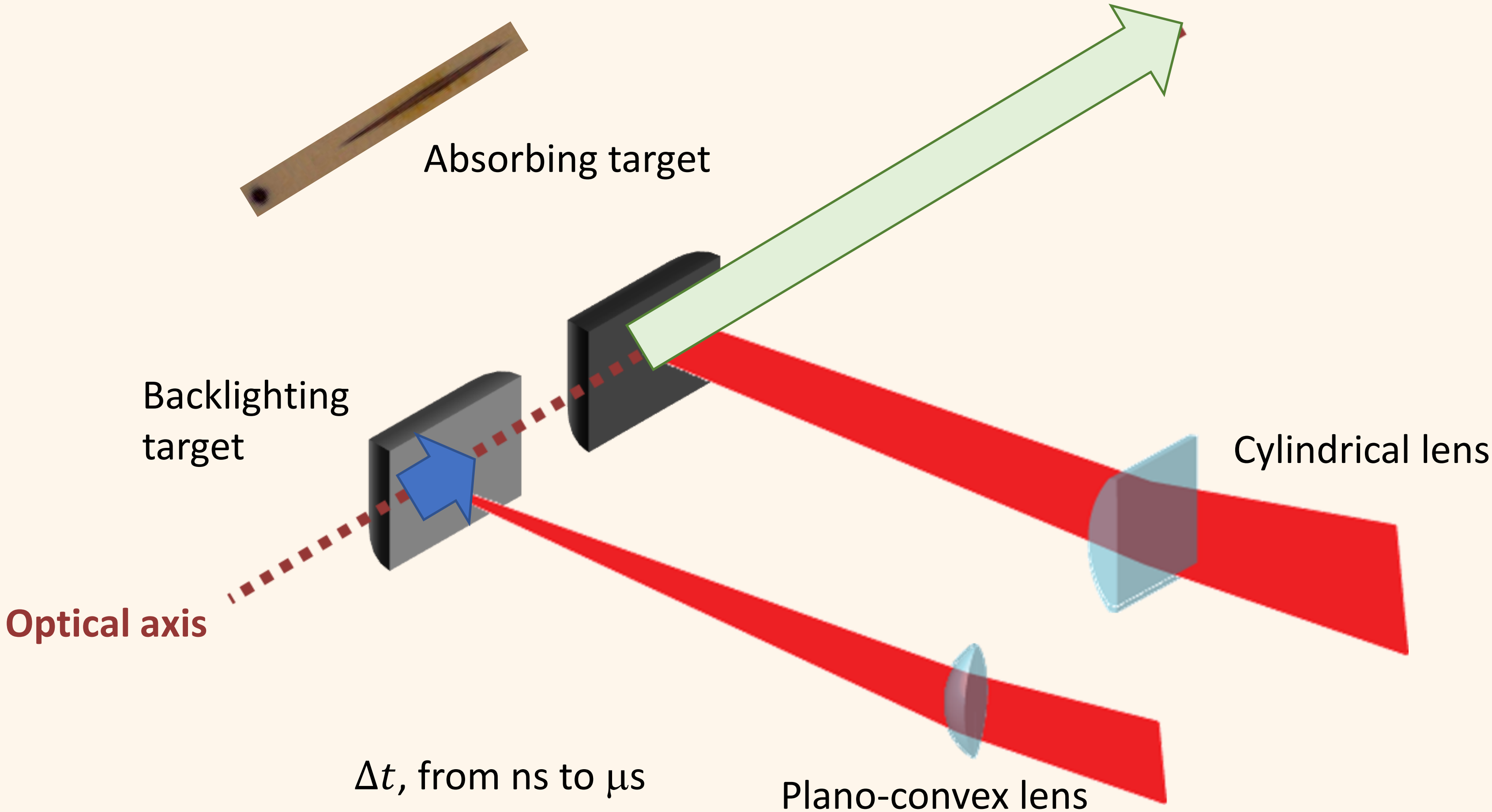

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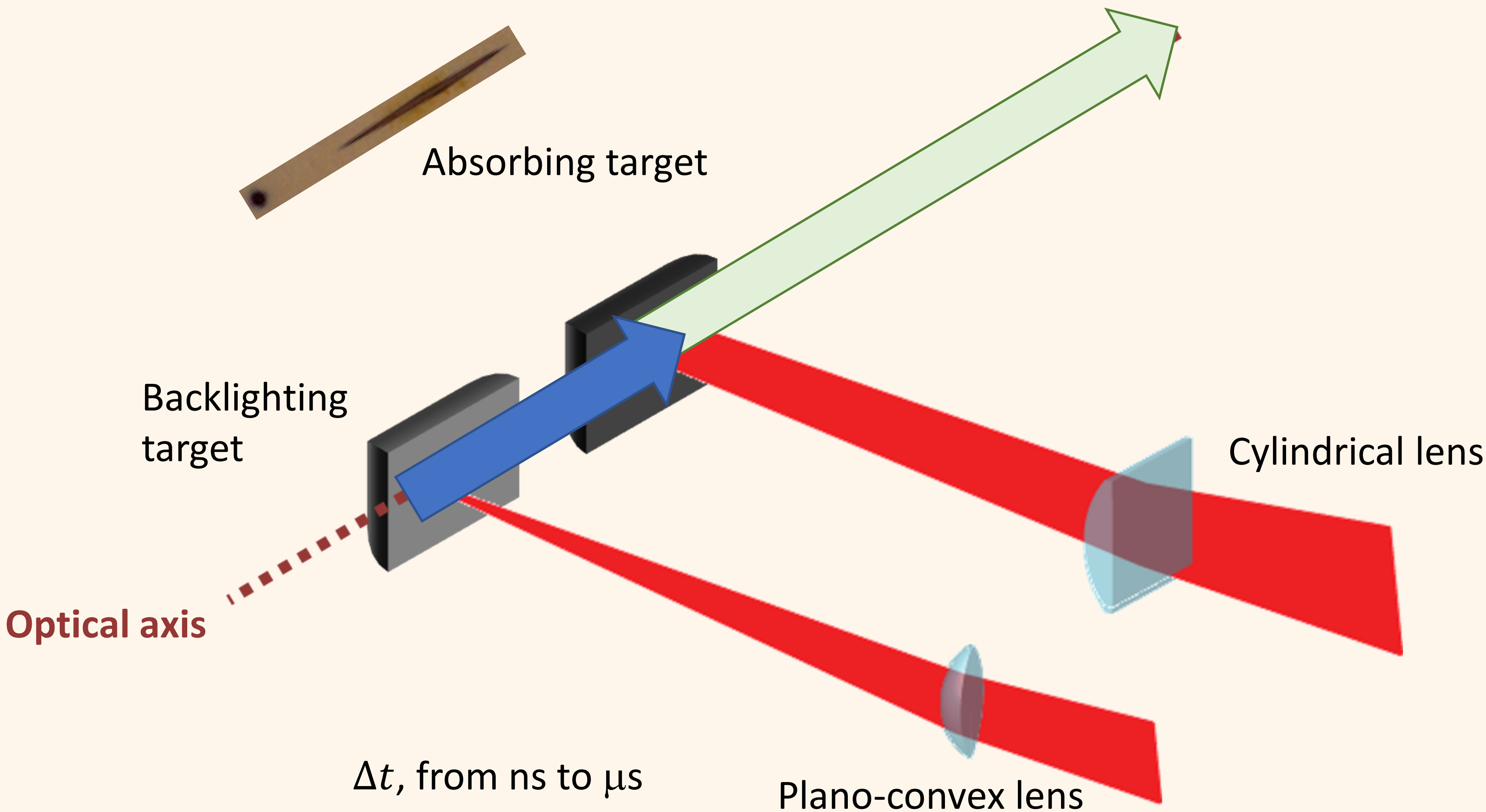

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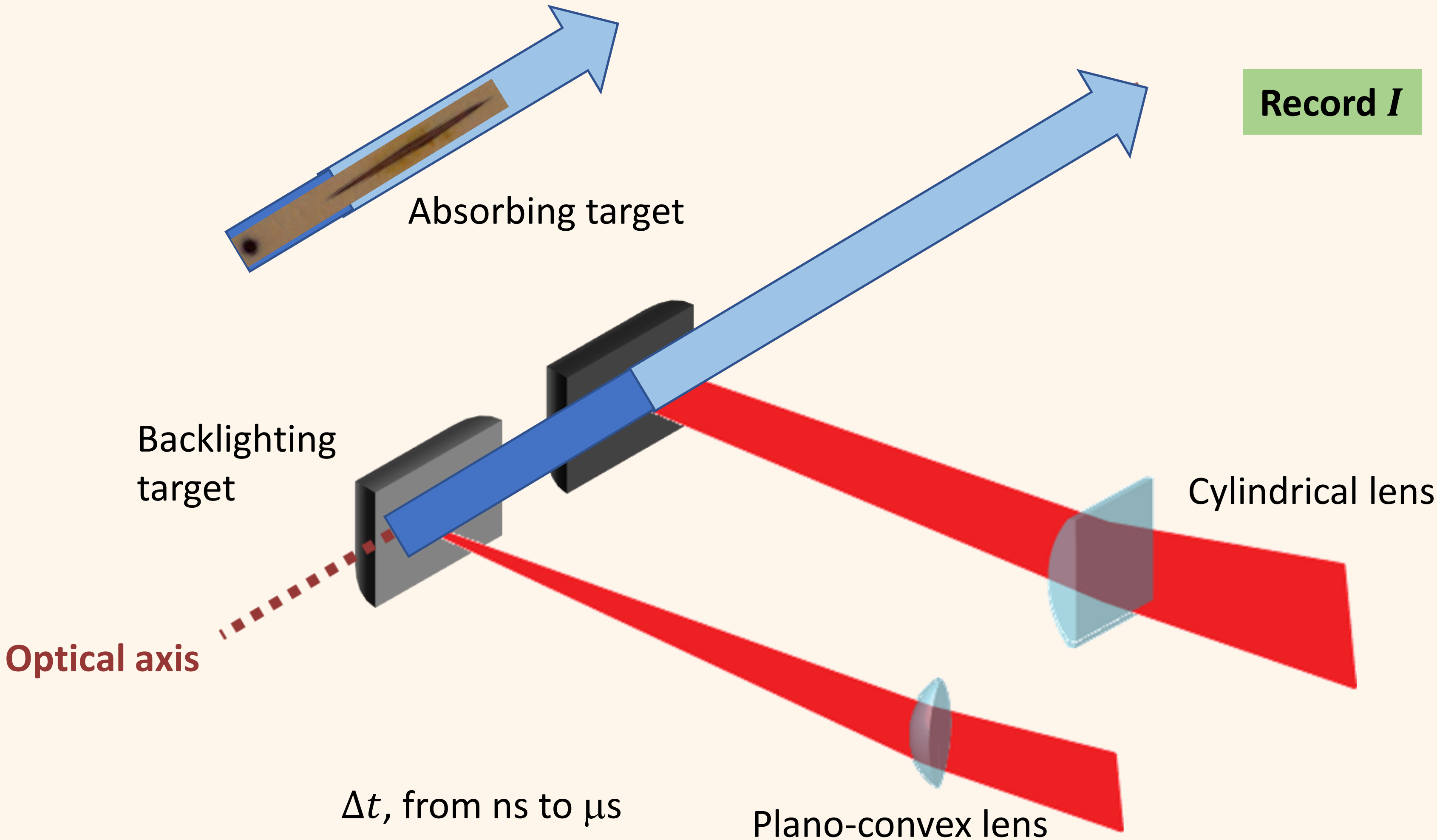

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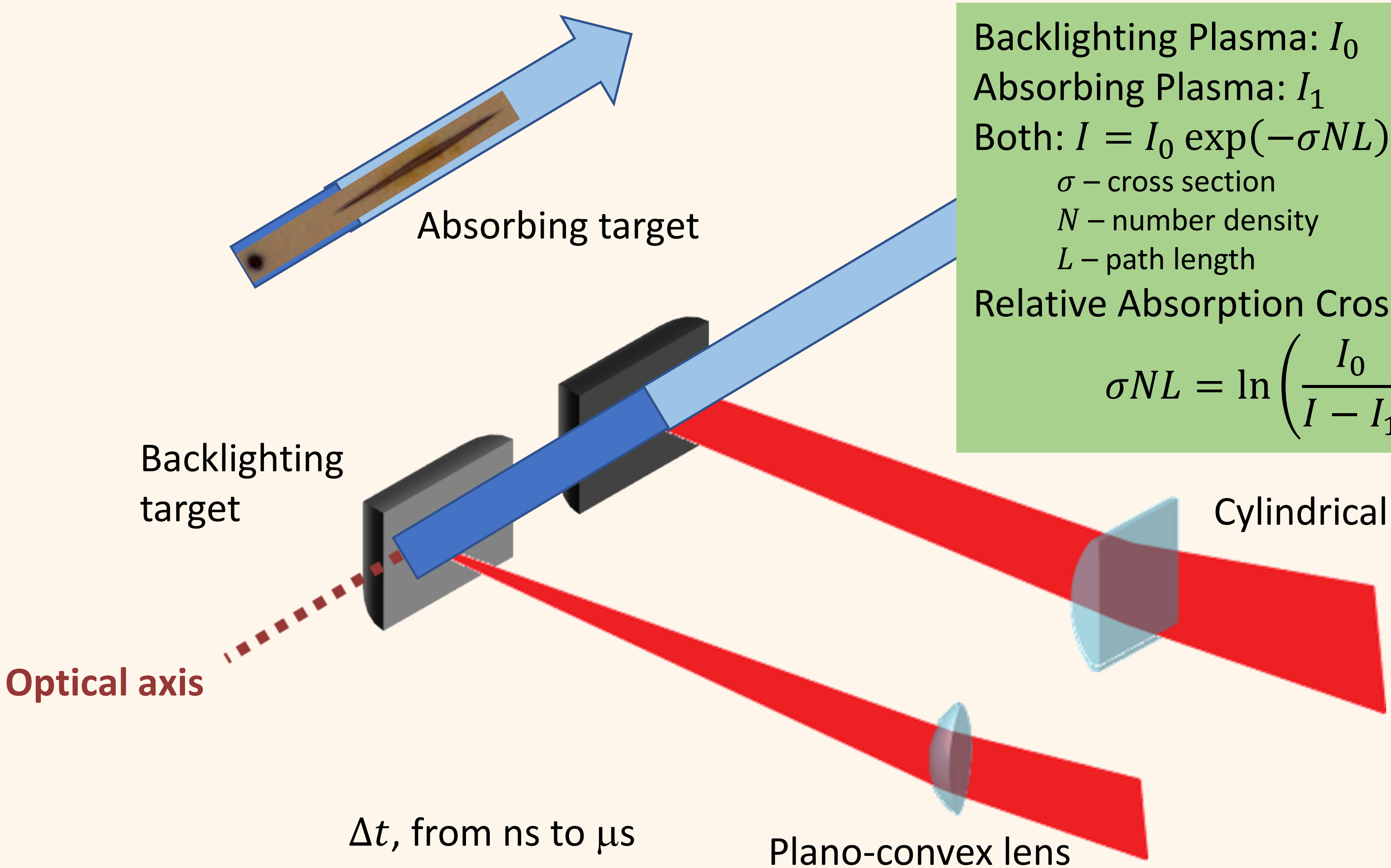


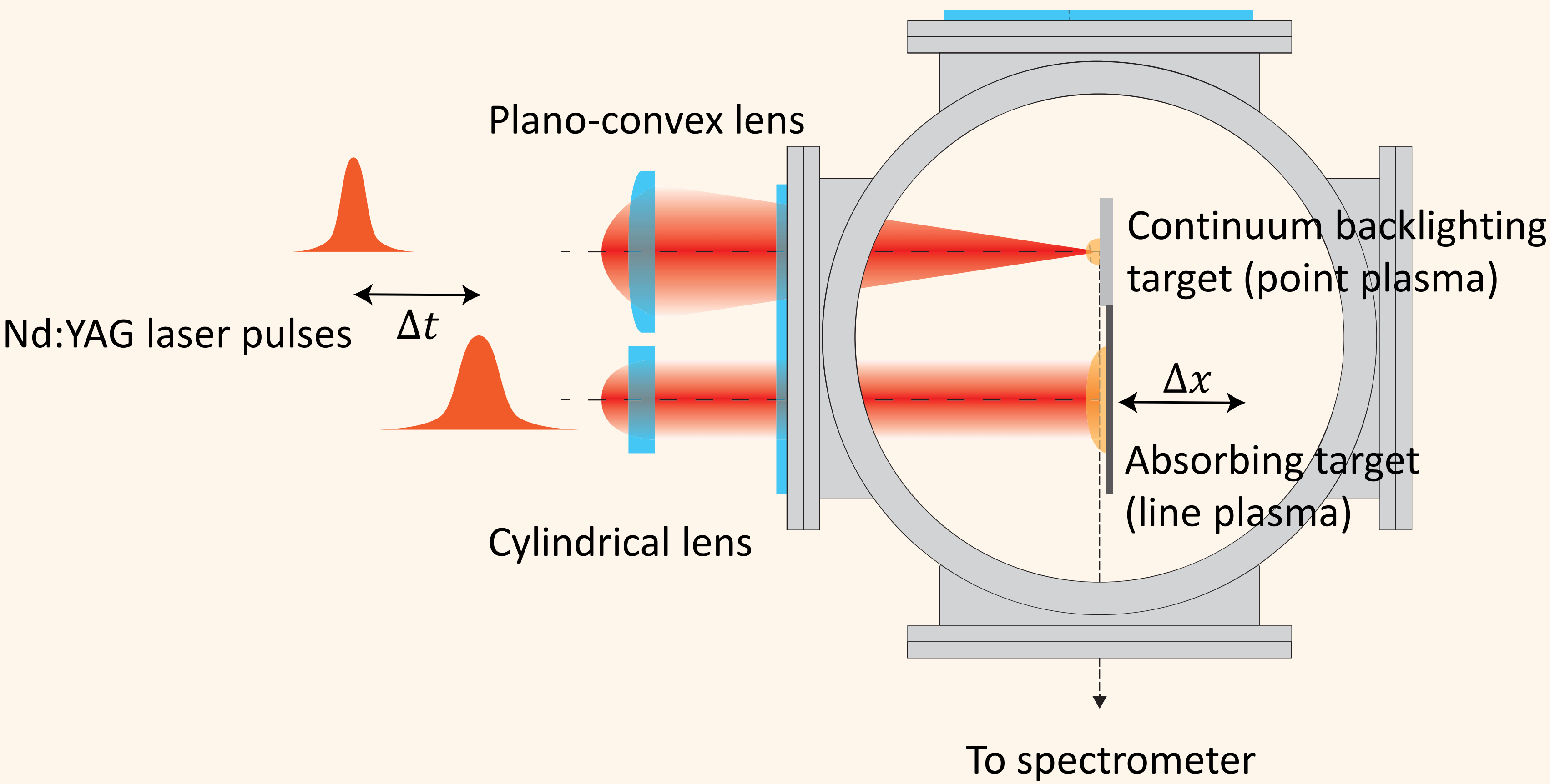
Backlighting Plasma: I_0
Absorbing Plasma: I_1
Both: $I = I_0 \exp(-\sigma N L) + I_1$

- σ – cross section
- N – number density
- L – path length

Relative Absorption Cross Section:

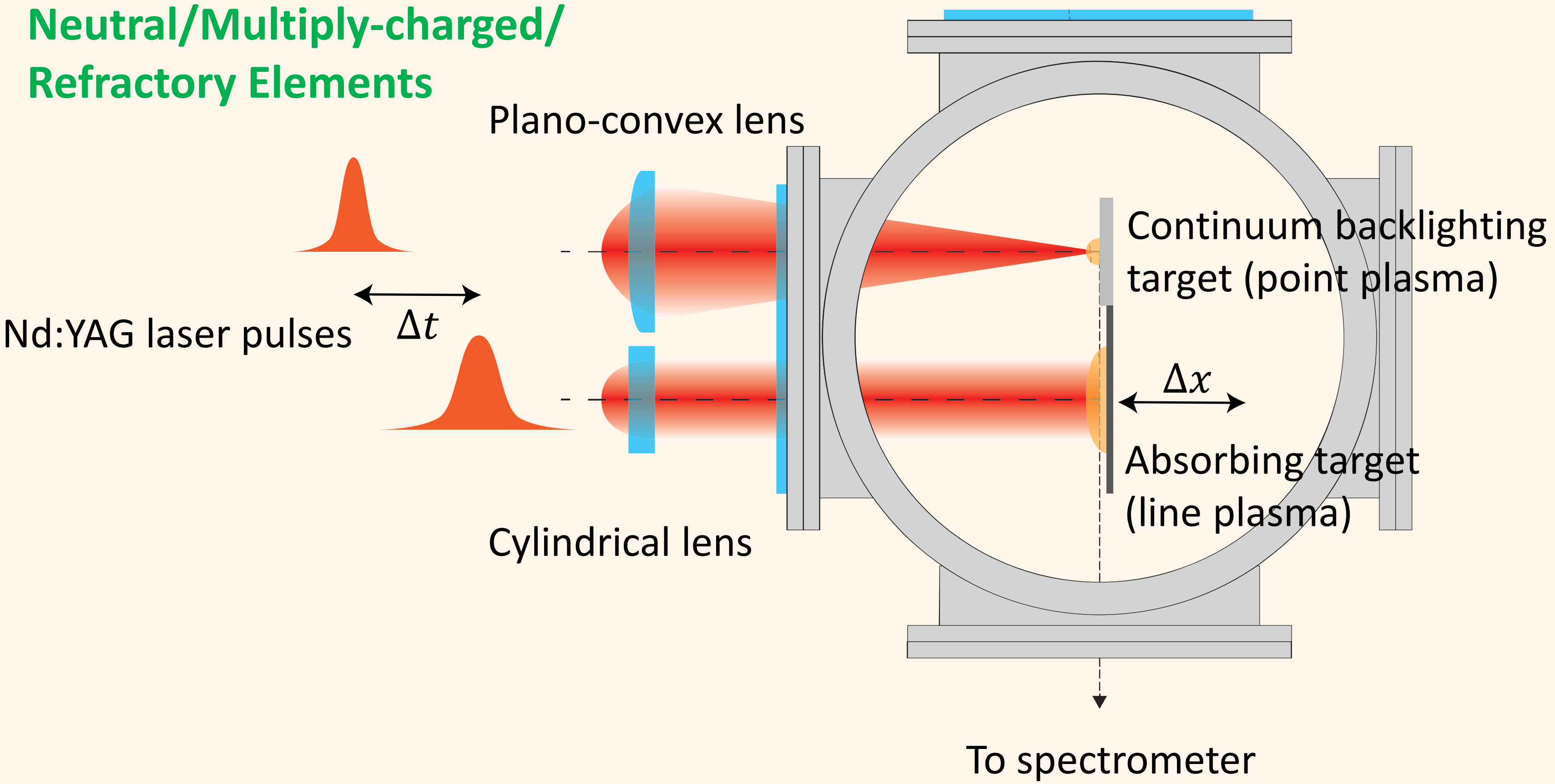
$$\sigma N L = \ln\left(\frac{I_0}{I - I_1}\right)$$

DLP Photoabsorption - The Set-up



DLP Photoabsorption - The Set-up

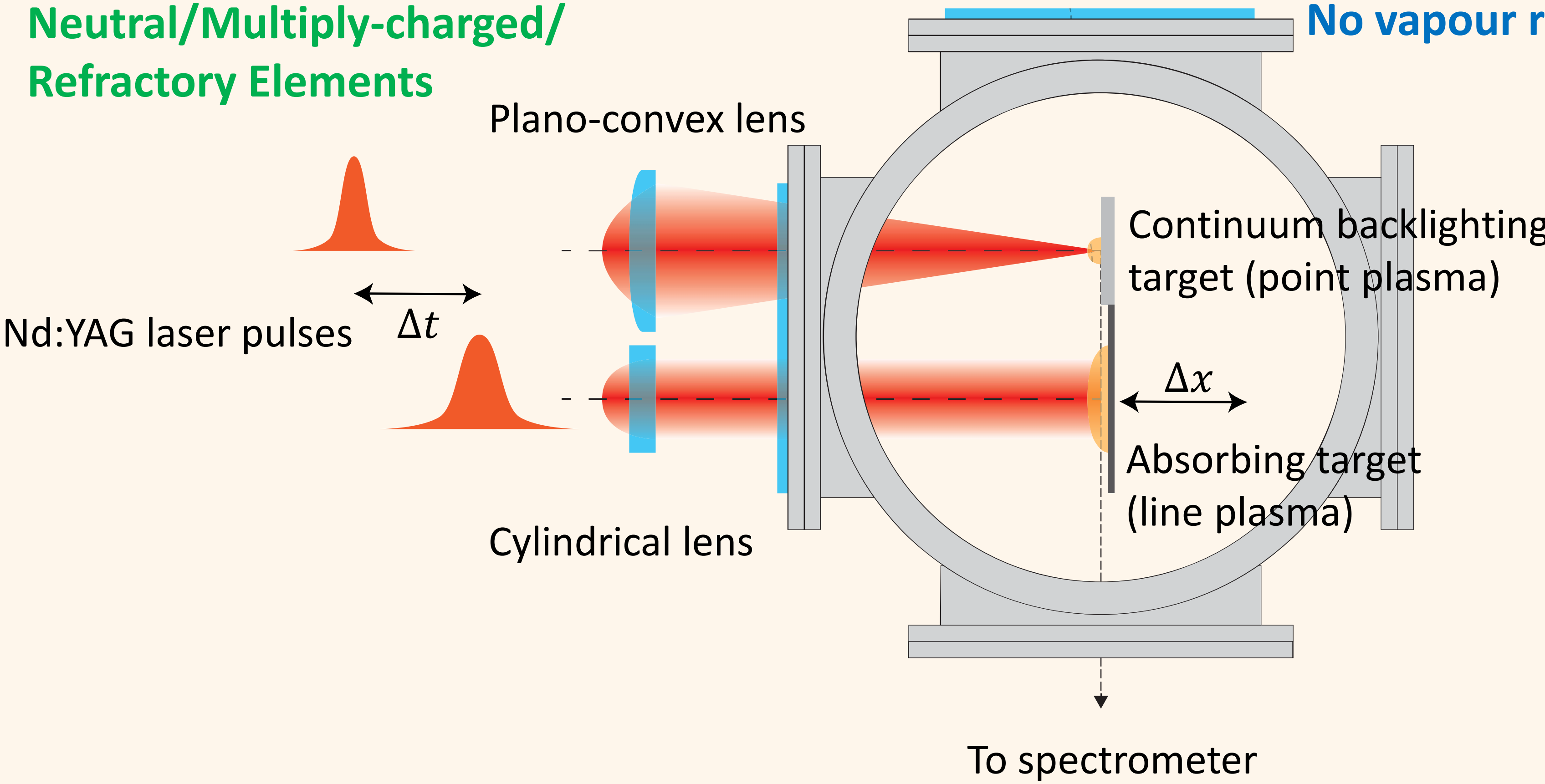
Species Flexibility:
Neutral/Multiply-charged/
Refractory Elements



DLP Photoabsorption - The Set-up

Species Flexibility:
Neutral/Multiply-charged/
Refractory Elements

No tuning required
No vapour required



DLP Photoabsorption - The Set-up

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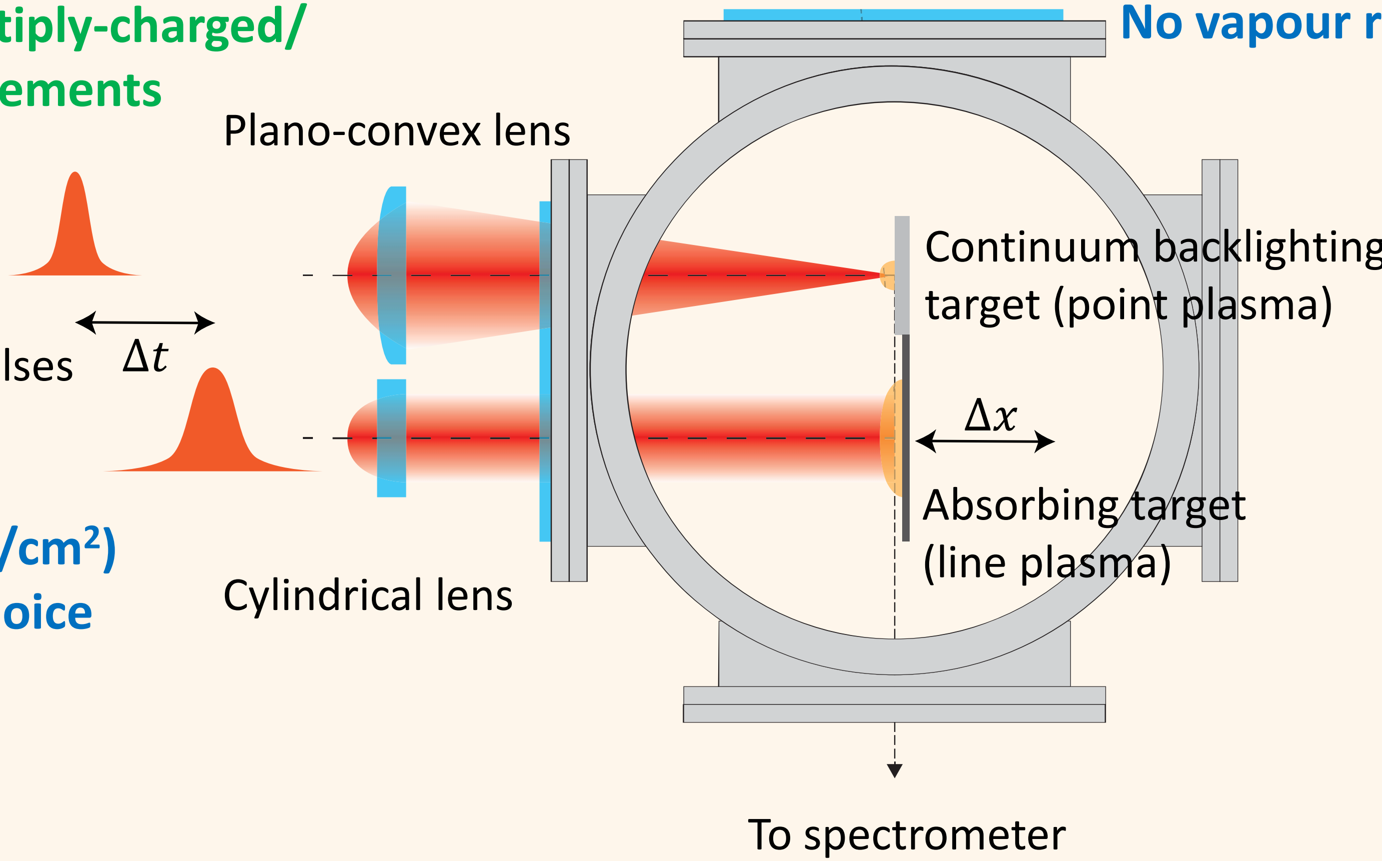
Nd:YAG laser pulses



Δt, Δx, ϕ (W/cm^2)
$\rightarrow$ Species choice

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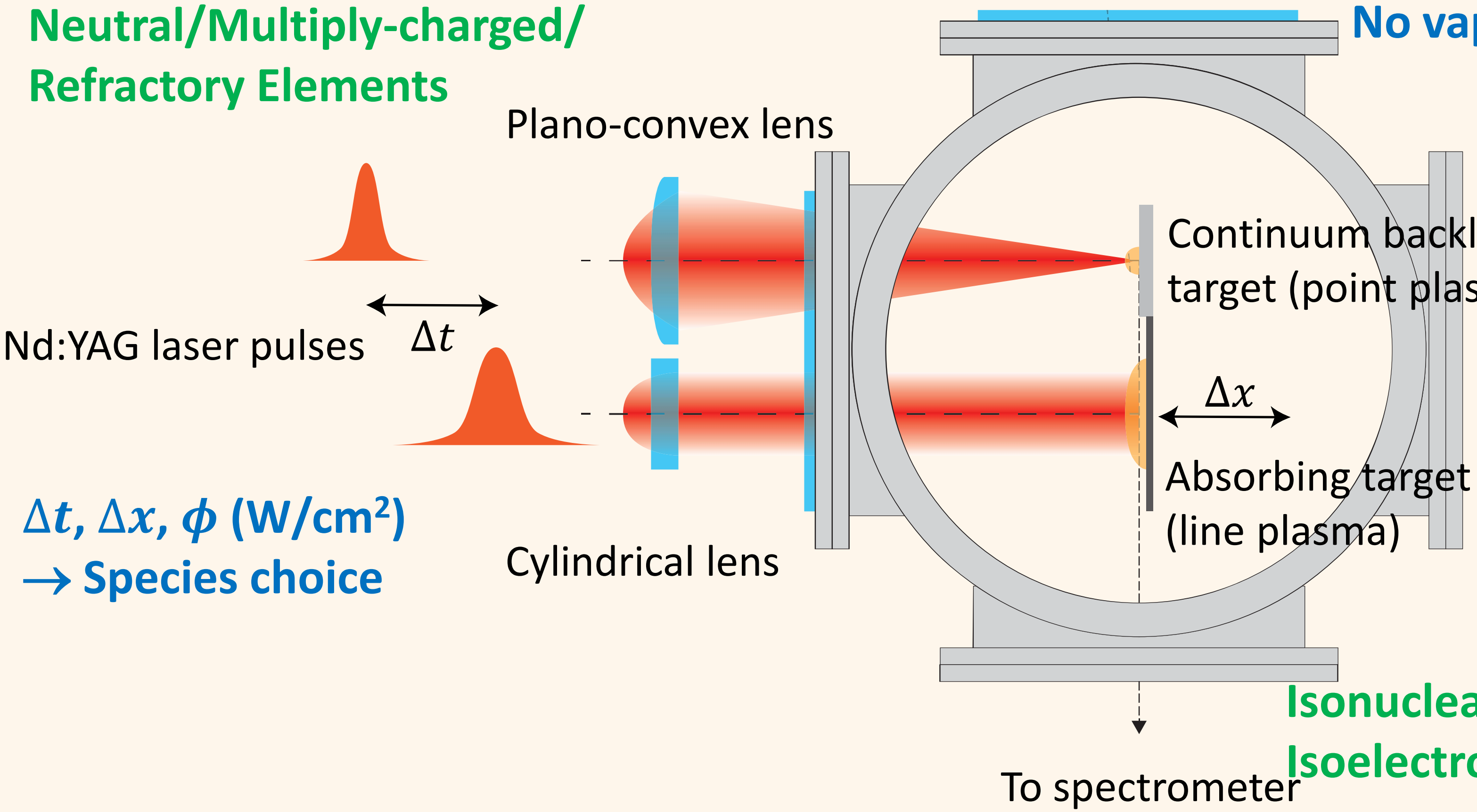

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$\rightarrow$ Species choice

Isonuclear Sequences
Isoelectronic Sequences

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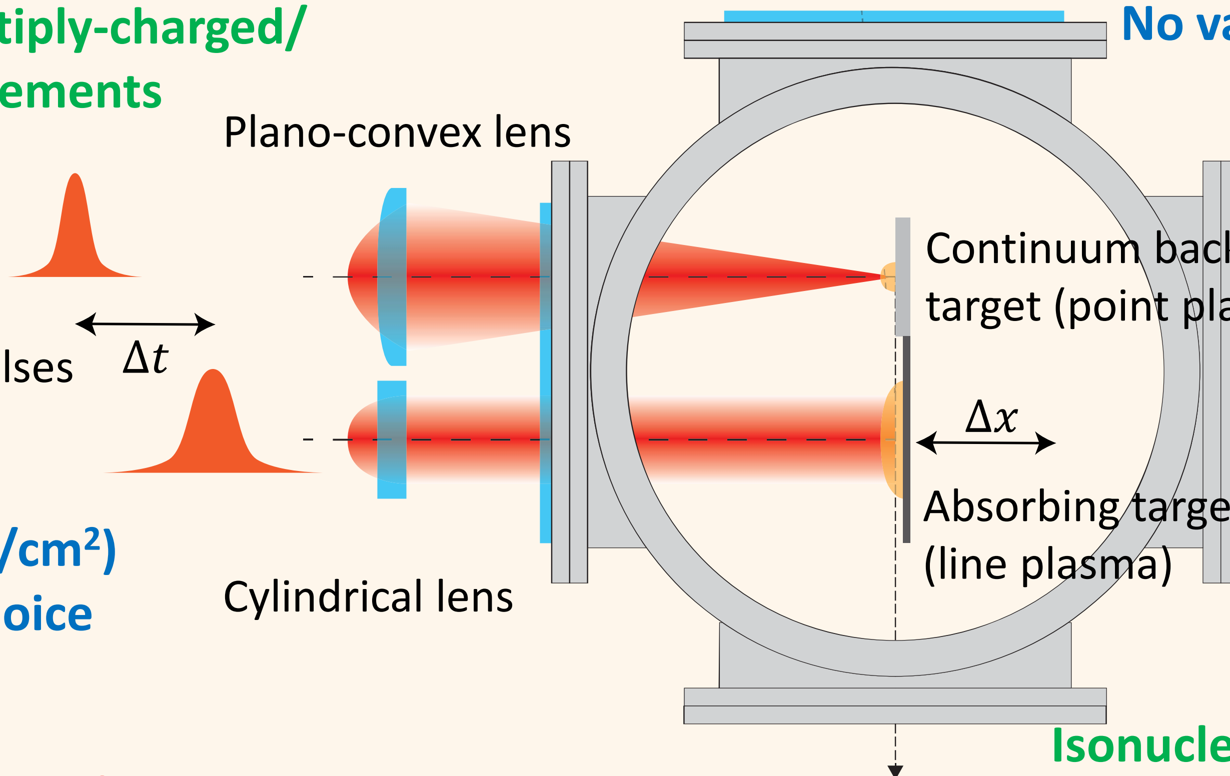

$\Delta t, \Delta x, \phi$ (W/cm²)
$\rightarrow$ Species choice

Isonuclear Sequences
Isoelectronic Sequences

Relative Absorption Cross Section:

$$\sigma N L = \ln\left(\frac{I_0}{I - I_1}\right)$$

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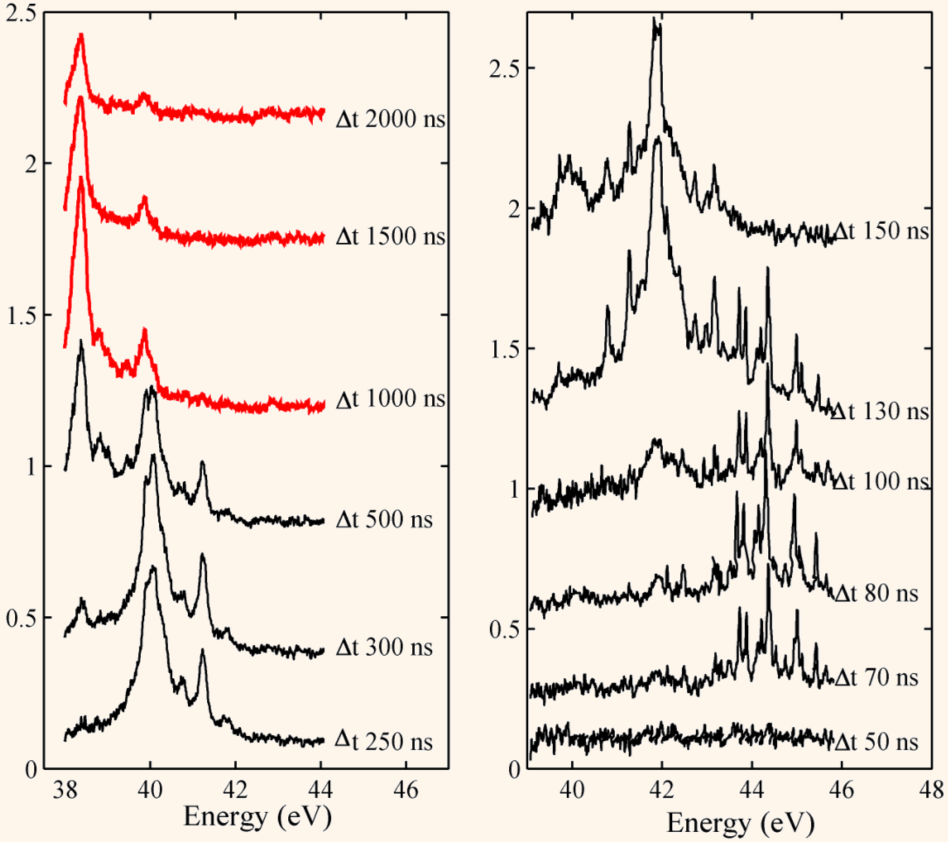
DLP Photoabsorption Example - Te



Δx = 2 mm

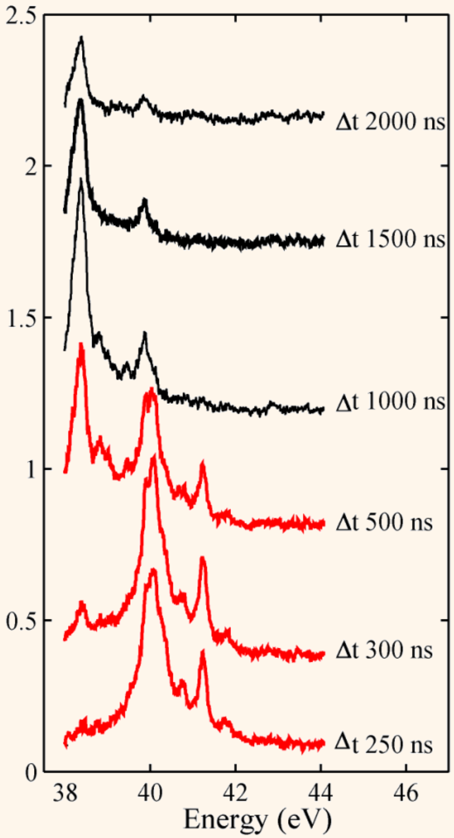
ϕ = 5 x 10^9 W/cm^2 (cylindrical lens)

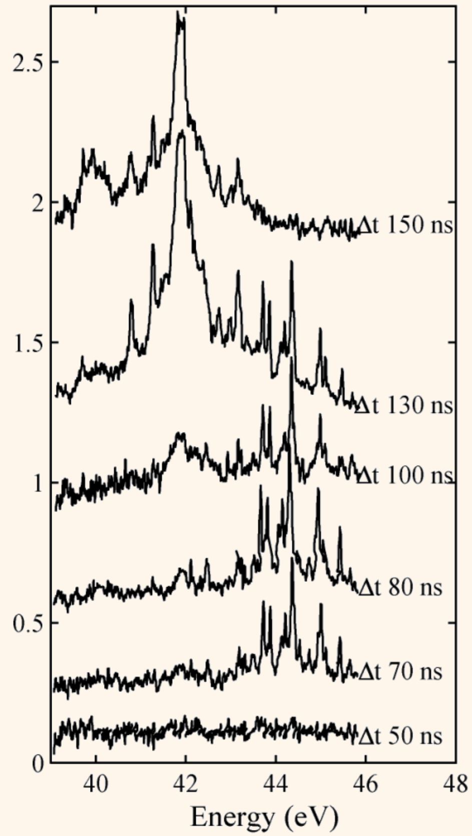
- Neutral Te dominates at the time delays (Δt) shown.
- Discrete structure arising from $4d - np$ transitions

Gaynor *et al.* (2005) *J. Phys. B: At. Mol. Opt. Phys.* **38** 2895

Murphy *et al.* (1999) *J. Phys. B: At. Mol. Opt. Phys.* **32** 3905

DLP Photoabsorption Example - Te





Δx = 2 mm

ϕ = 5 x 10^9 W/cm^2 (cylindrical lens)

- Te^+ dominates at Δt's shown.
- Discrete structure arising from $4d - np$ transitions

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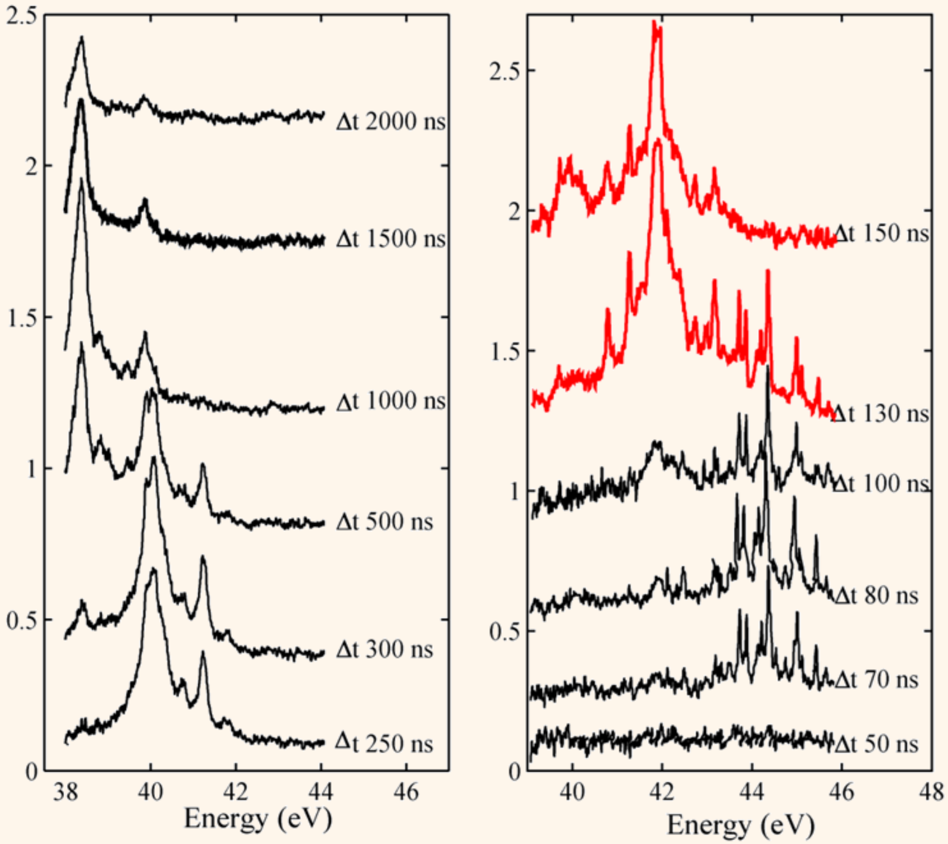

Δx = 0 mm

ϕ = 6 x 10^{11} W/cm^2 (spherical lens)

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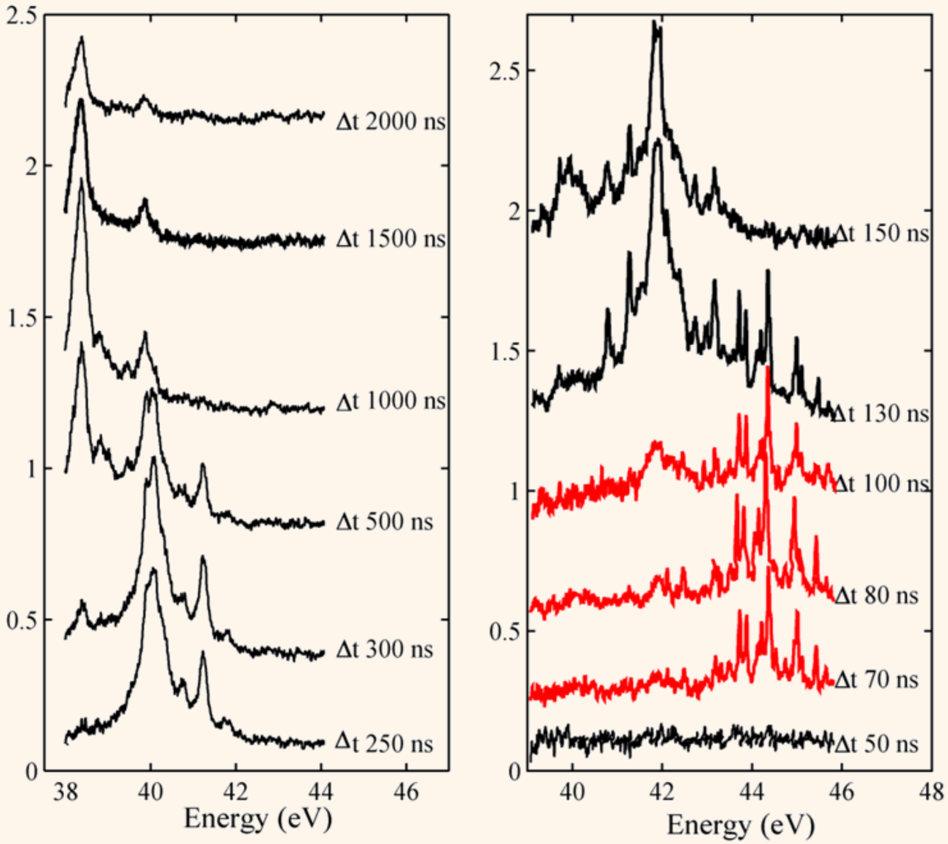

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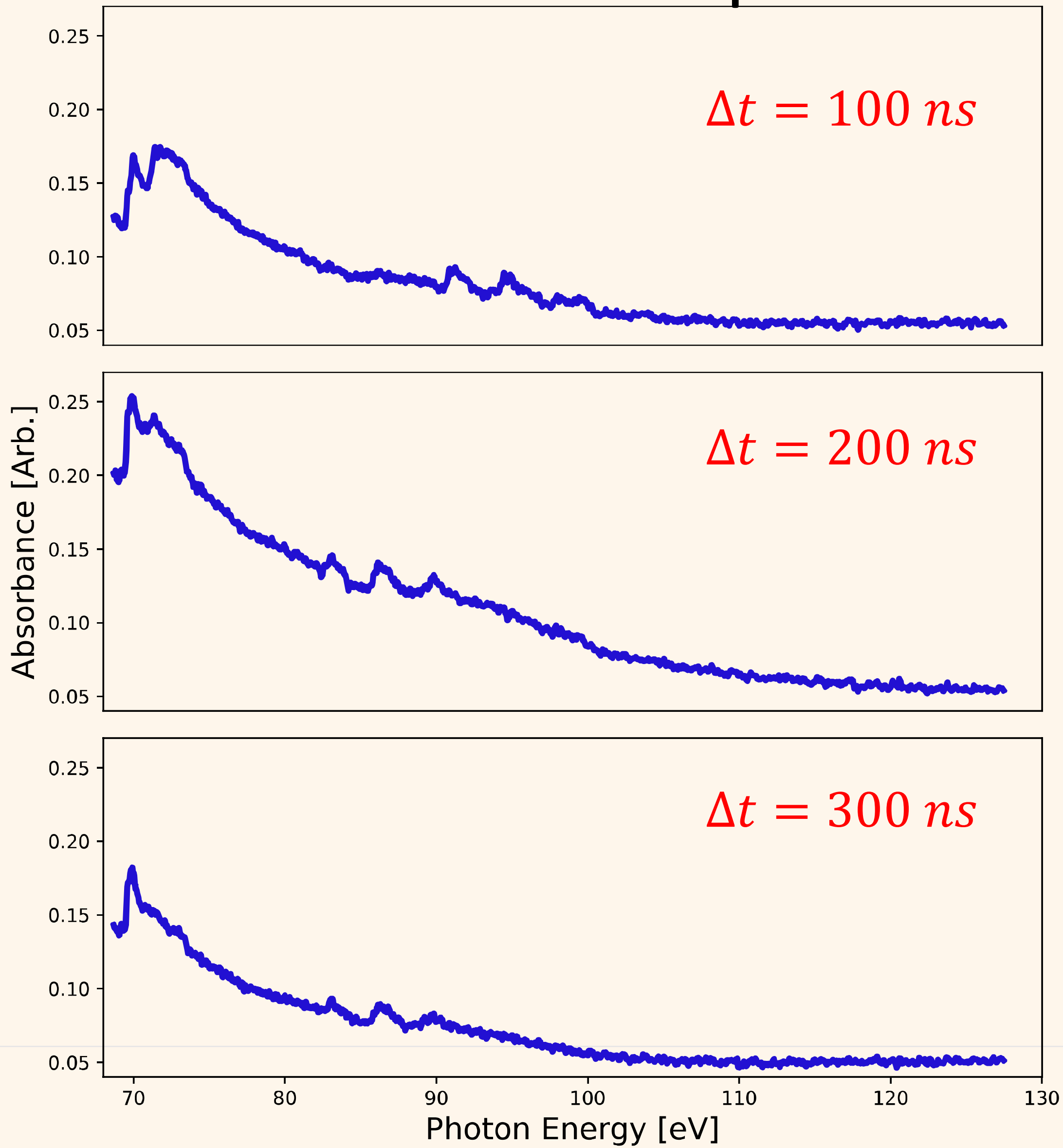
EUV Photoabsorption of Pt

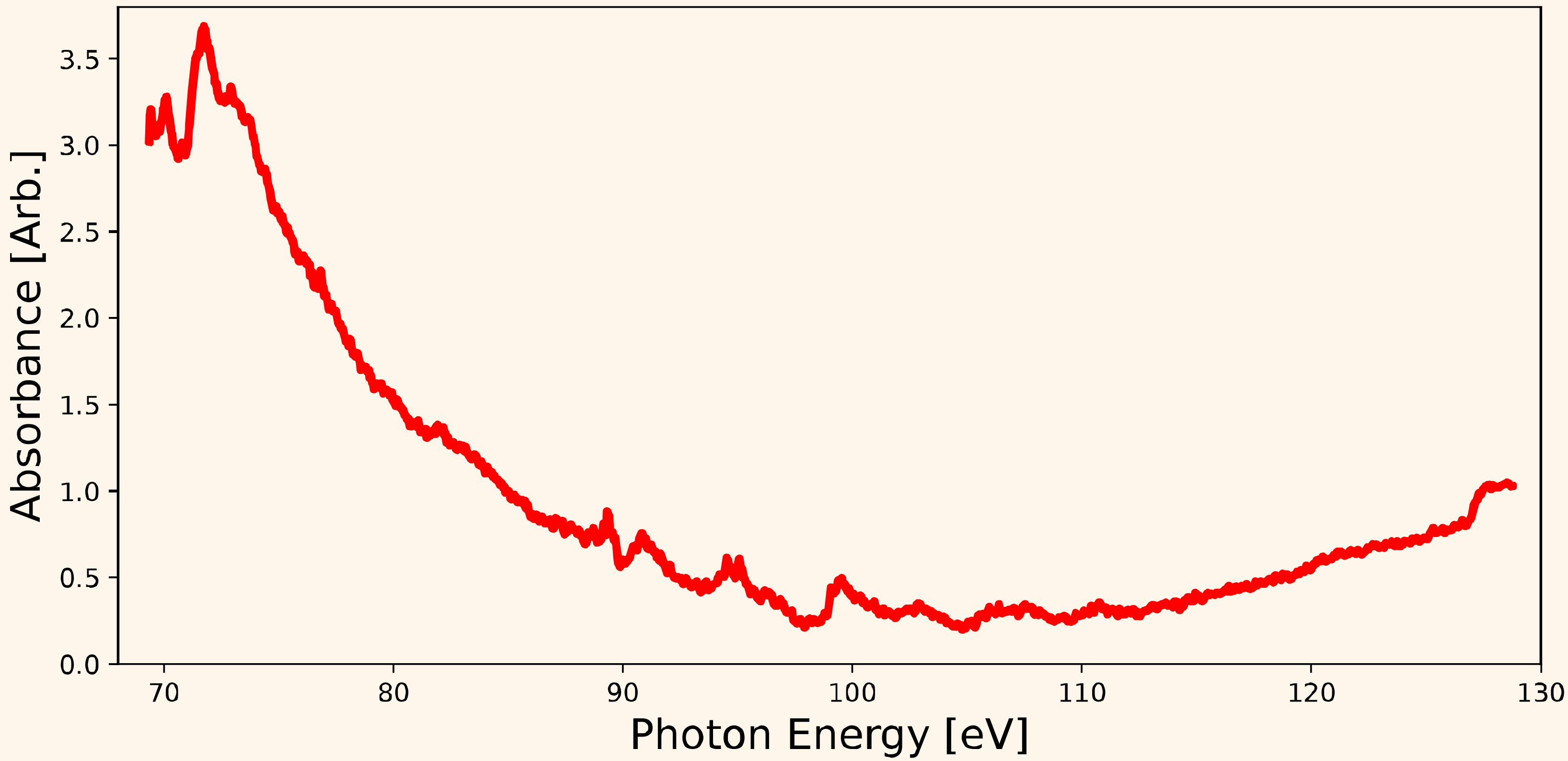


Δx = 2 mm

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

Eric Doyle *et al* 2023 *J. Phys. B: At. Mol. Opt. Phys.* **56** 135002

EUV Photoabsorption of Pt



Δx = 0 mm

ϕ = 5 x 10^{10} W/cm^2

Δt = 60 ns

Eric Doyle *et al* 2023 *J. Phys. B: At. Mol. Opt. Phys.* **56** 135002

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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
- Transition energies and oscillator strengths calculations using Cowan's RCN, RCN2 and RCG suite of codes Cowan R D 1981 The Theory of Atomic Structure and Spectra (Berkeley, CA: University of California Press)
- 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 $m = 9, 8, 7,$ and 6 in Pt^{+}, Pt^{2+}, Pt^{3+} and Pt^{4+} respectively. n is the principal quantum number with values of 5, 6, and 7, and δ is the ejection energy of a free electron of angular momentum $p, f, h,$ or k.

	Final	
Initial	Discrete	Continuous
$[Xe]\ 4f^{14}\ 5d^{m}$	$[Xe]\ 4f^{13}\ 5d^{m}\ nd$	$[Xe]\ 4f^{14}\ 5d^{m-1} + \delta(p,f,h,k)$
		$[Xe]\ 4f^{14}\ 5d^{m-2}\ 6d + \delta(p,f,h)$
$[Xe]\ 4f^{14}\ 5d^{m-1}\ 6s^{1}$	$[Xe]\ 4f^{13}\ 5d^{m-1}\ 6s\ nd$	$[Xe]\ 4f^{14}\ 5d^{m-1} + \delta(p,f,h,k)$
		$[Xe]\ 4f^{14}\ 5d^{m-2}\ 6d + \delta(p,f,h)$
		$[Xe]\ 4f^{14}\ 5d^{m-2}\ 6s + \delta(p,f)$
		$[Xe]\ 4f^{14}\ 5d^{m-3}\ 6s\ 6d + \delta(p)$
$[Xe]\ 4f^{14}\ 5d^{m-2}\ 6s^{2}$	$[Xe]\ 4f^{13}\ 5d^{m-2}\ 6s^{2}\ nd$	

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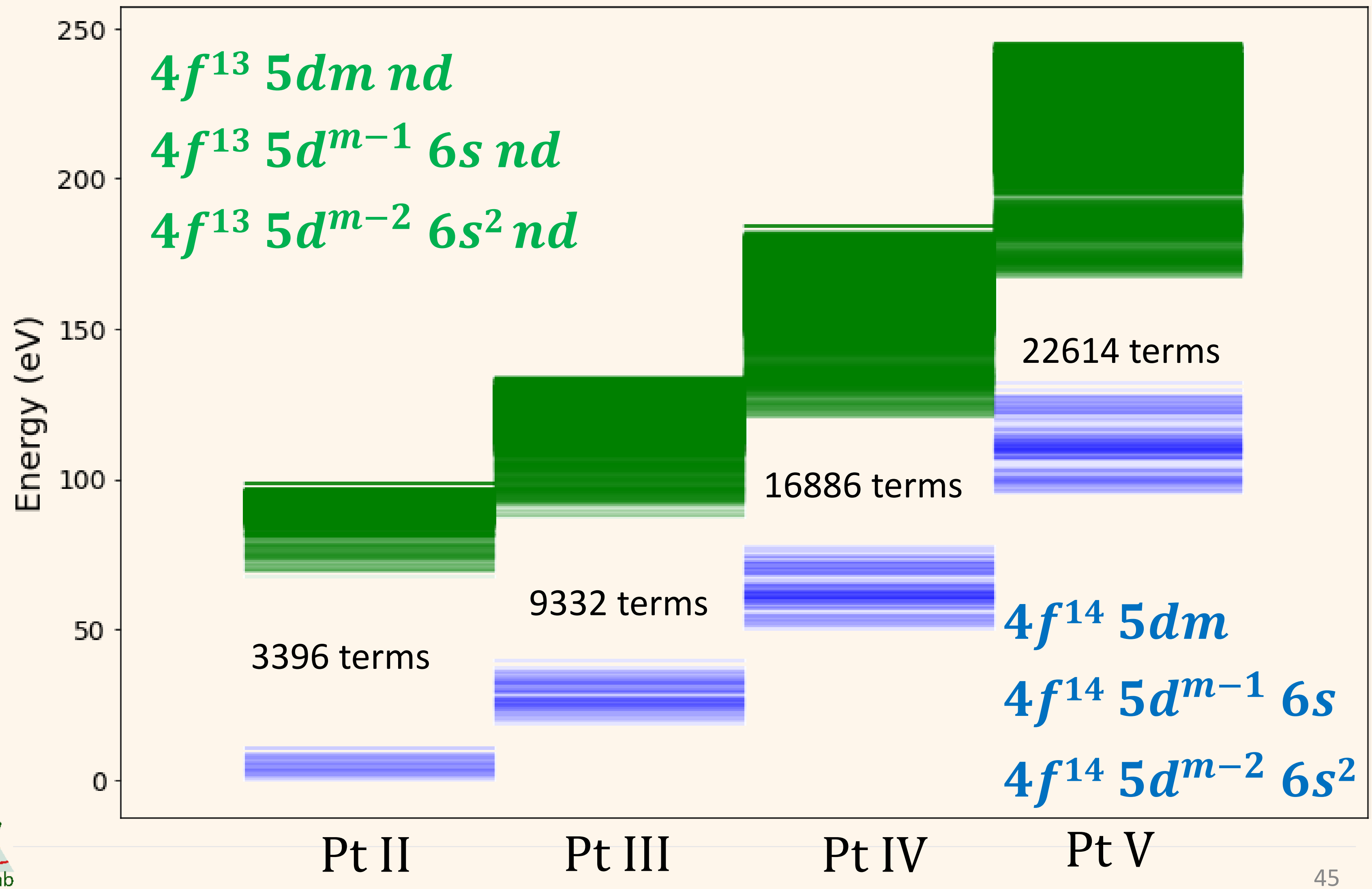
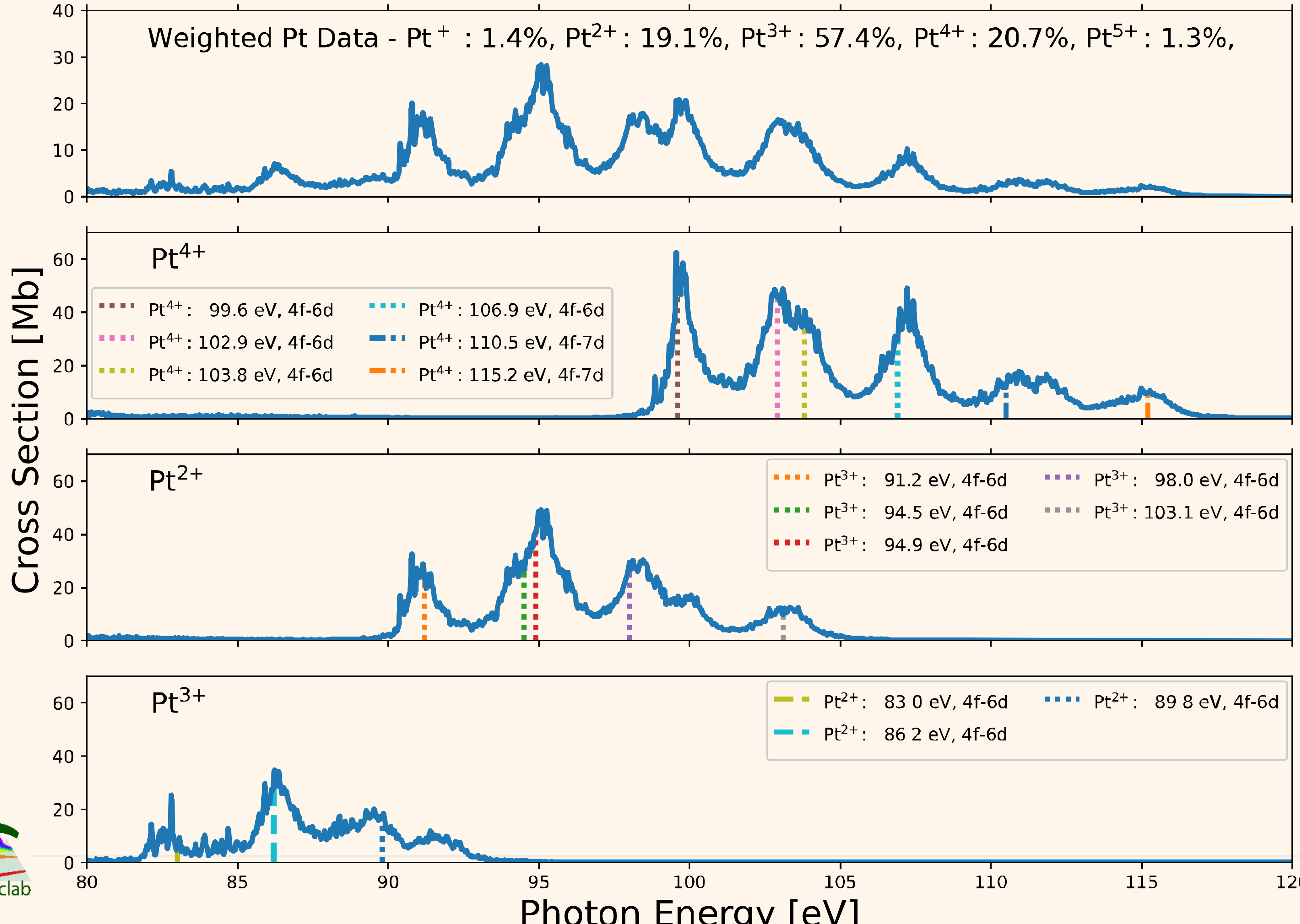
Cowan's Codes Calculations



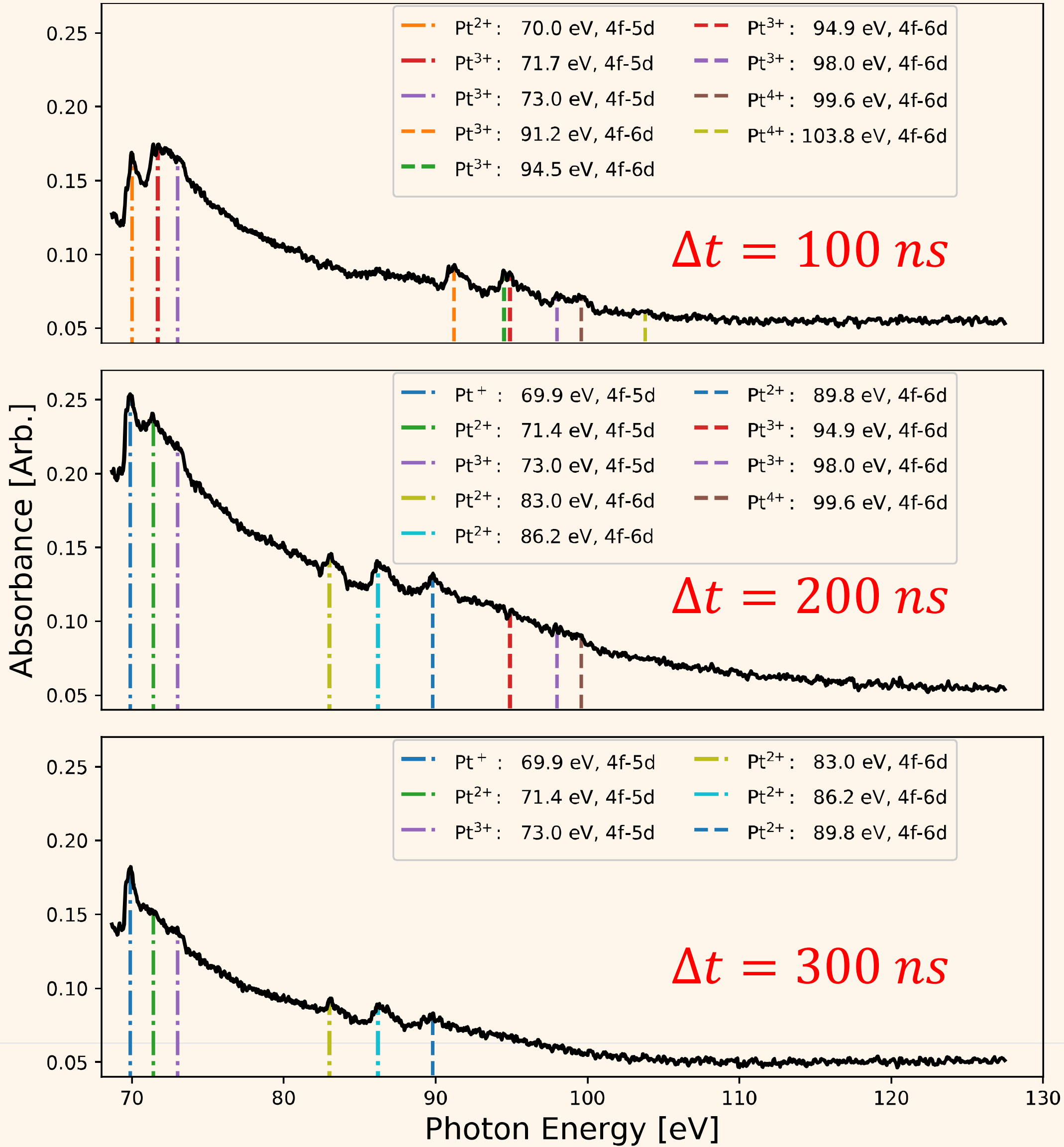
Identification of Absorption Features

- 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



Compare to Experimental Epectra



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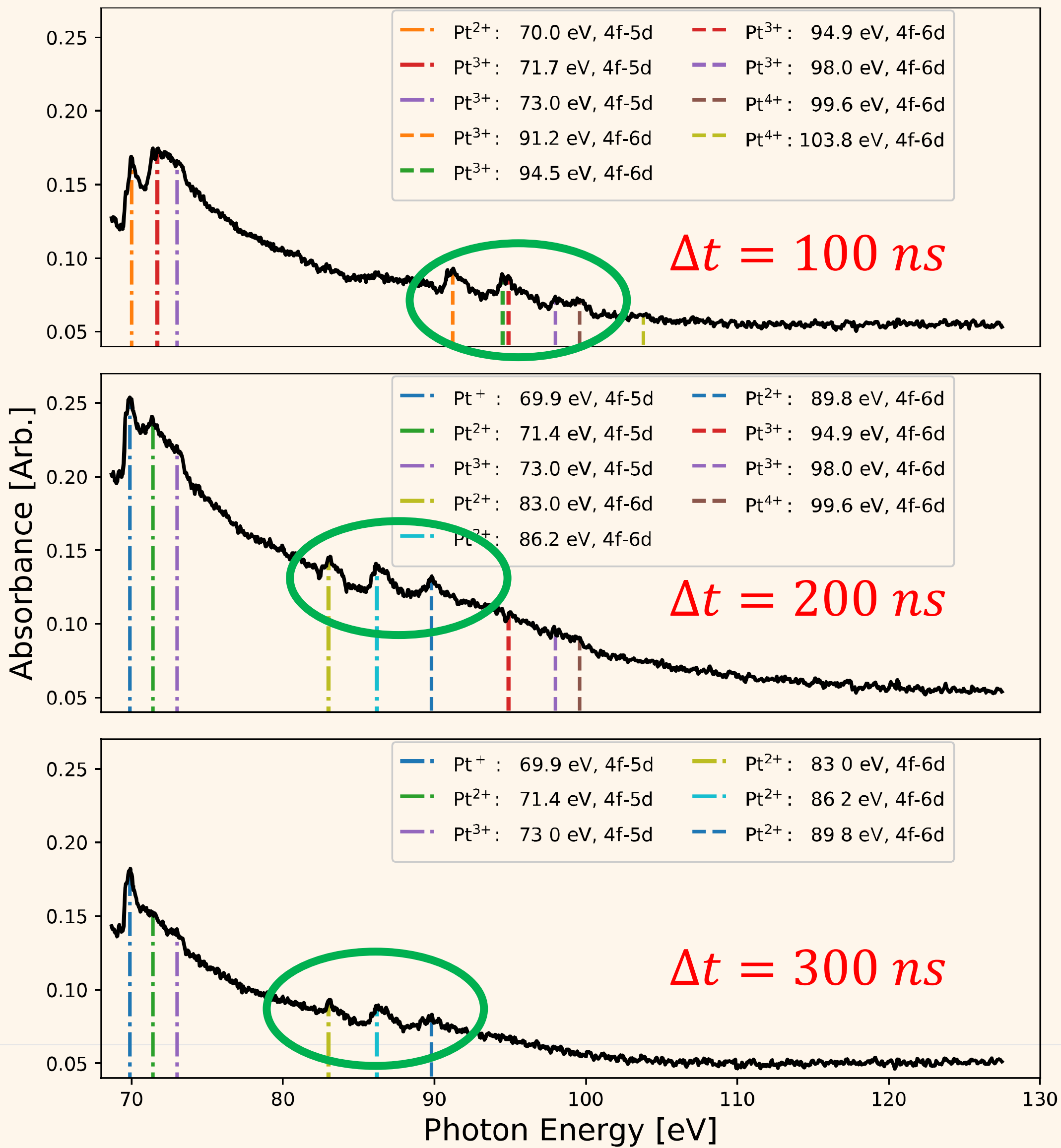
Δx = 2 mm

ϕ = 3 x 10^9 W/cm^2

$4f$ 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.

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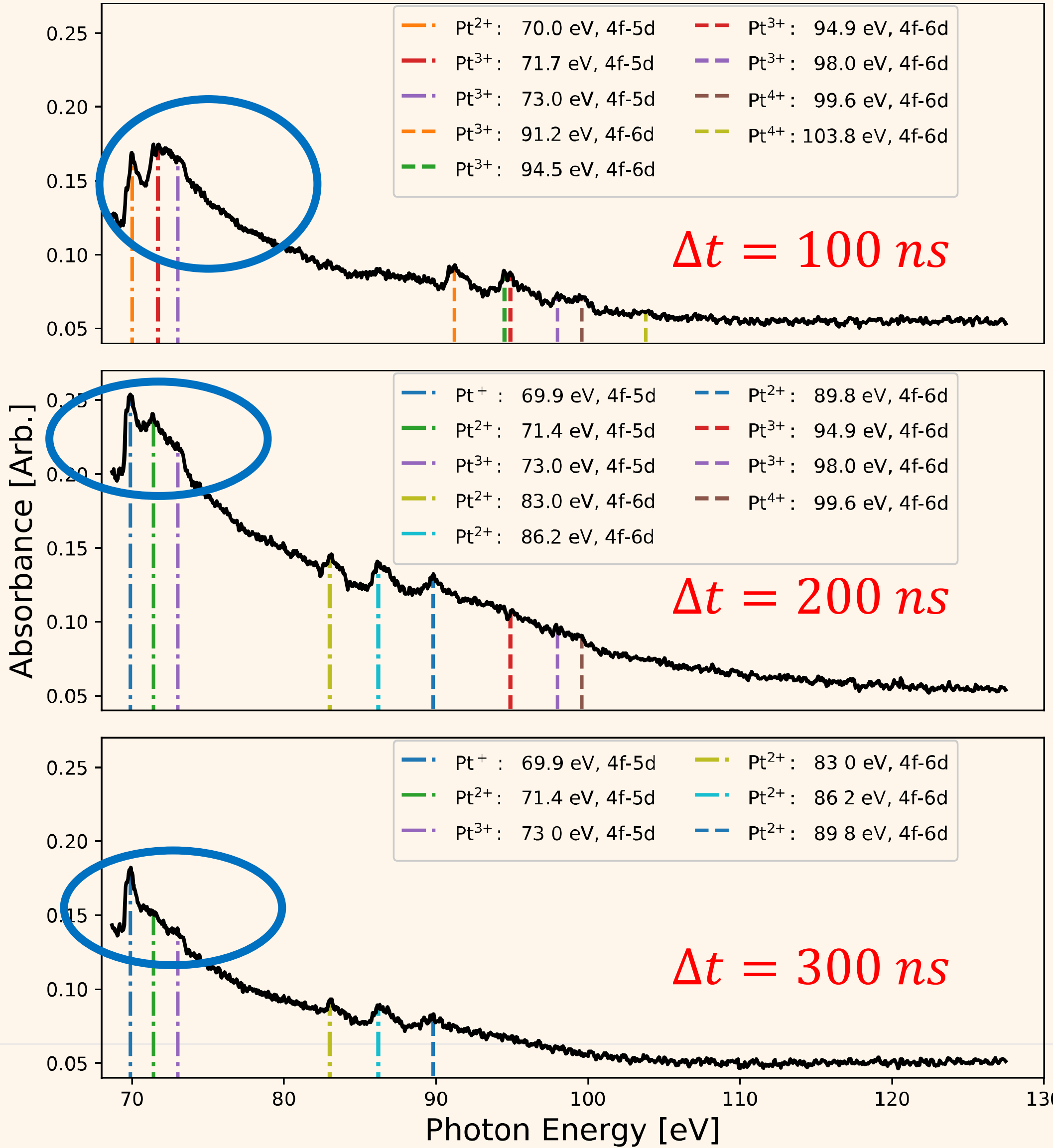
Δx = 2 mm

ϕ = 3 x 10^9 W/cm^2

$4f$ Photoabsorption in Pt II to Pt V



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

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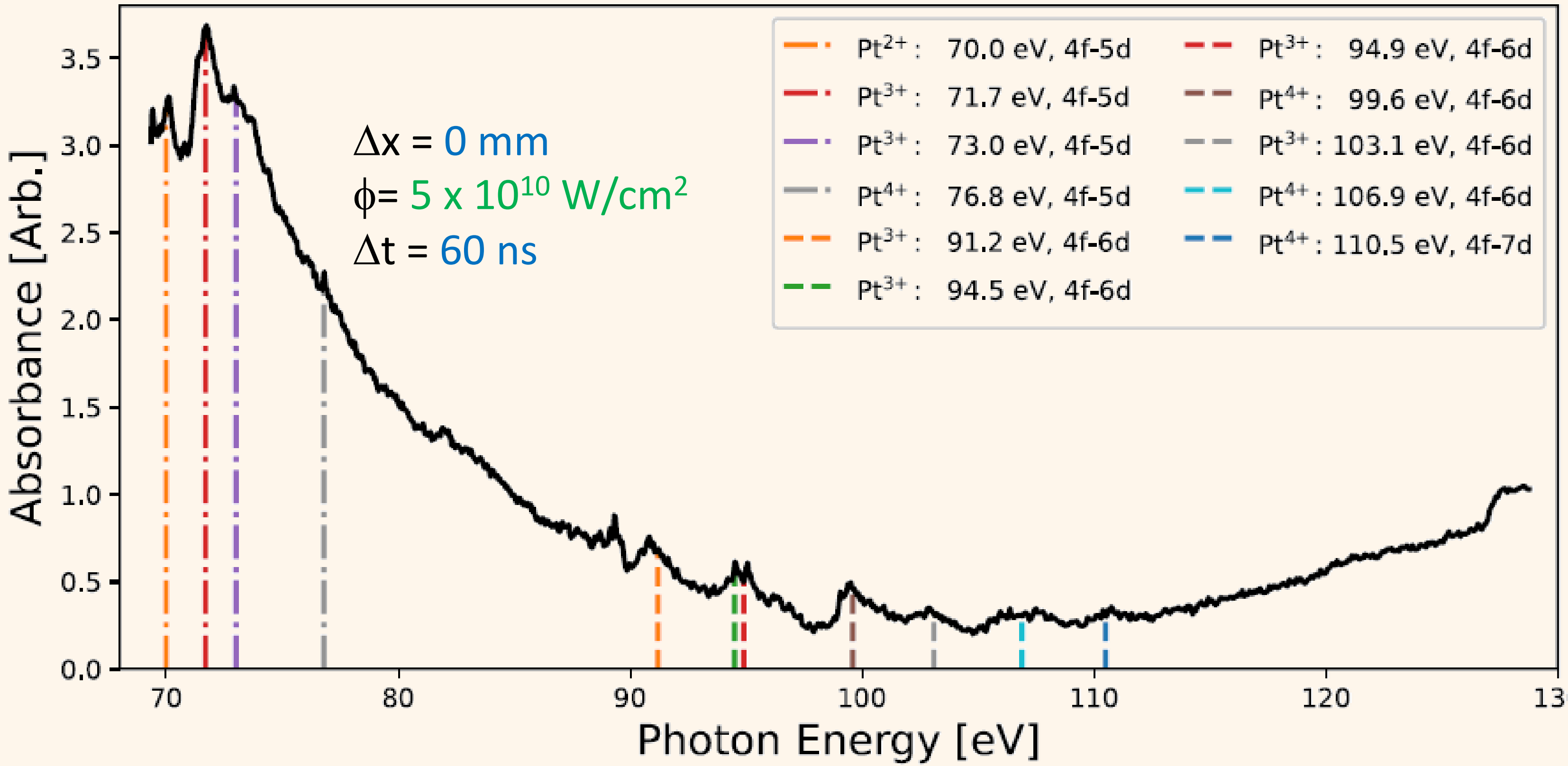
$\Delta x = 2$ mm

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

$4f$ Photoabsorption in Pt II to Pt V

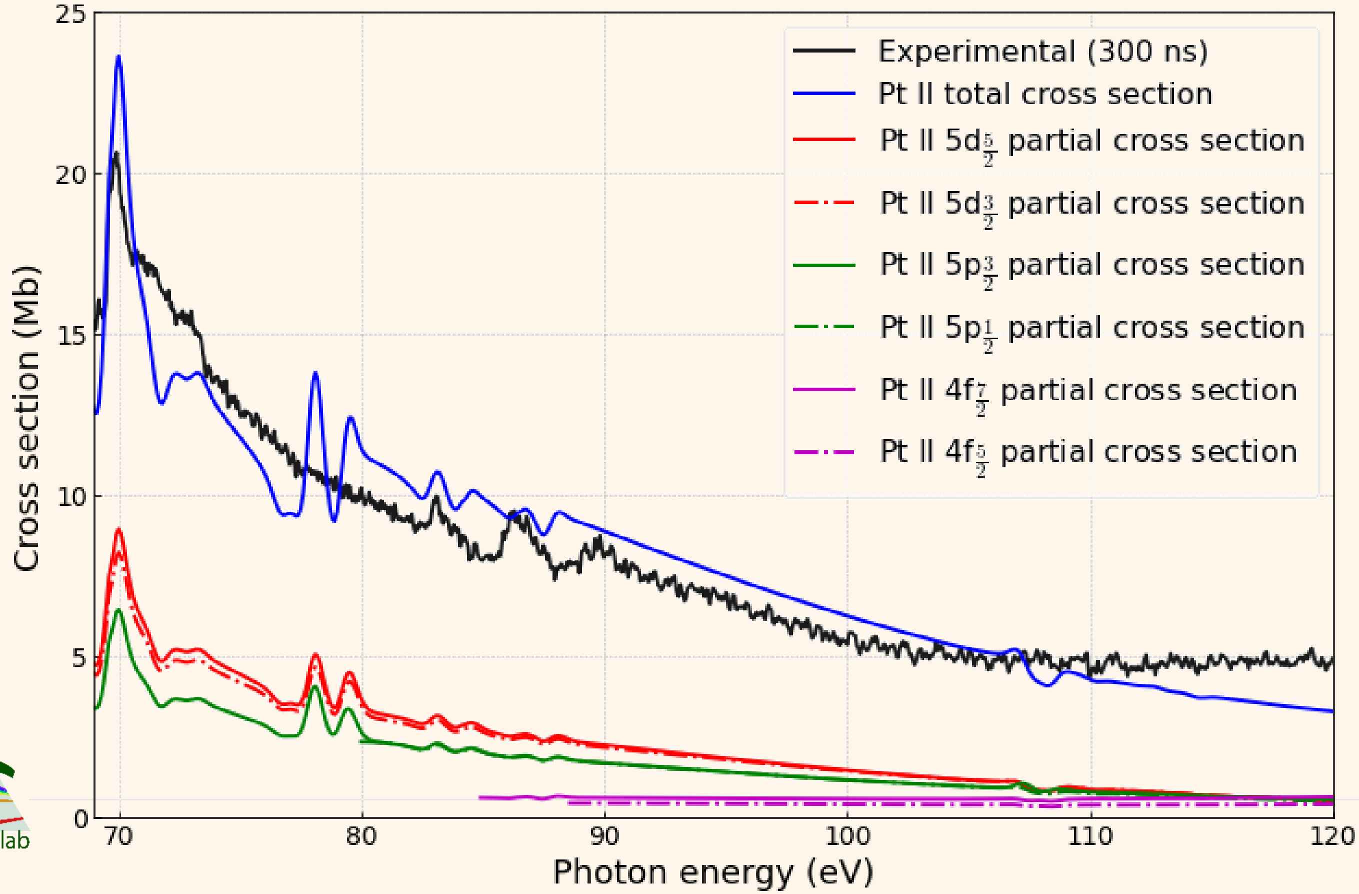


- The features identified sit on a continuum-like absorption feature
- This falls off with increasing energy between 70 and 110 eV.

RTDLDA Calculation

Libermann D A and Zangwill A 1984 *Comput. Phys. Commun.* **32** 75–82

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



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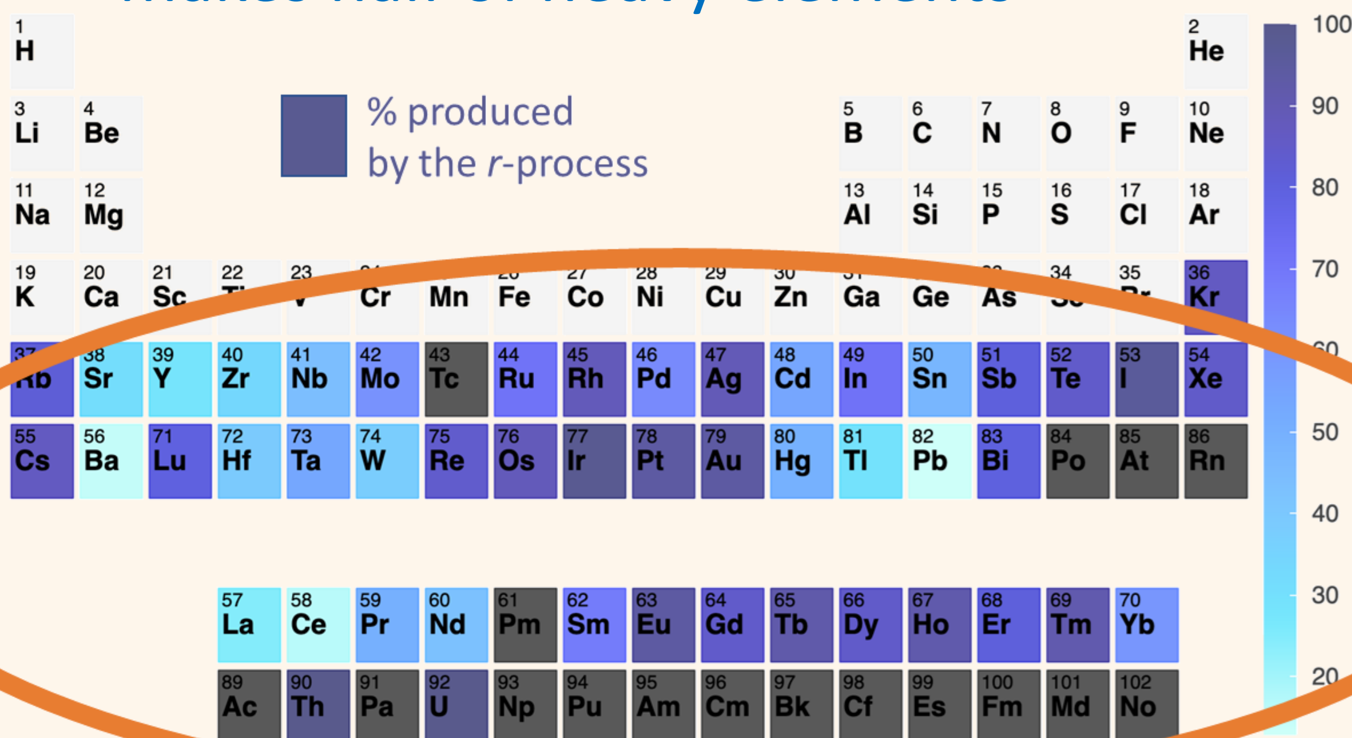
Speclab

Motivation

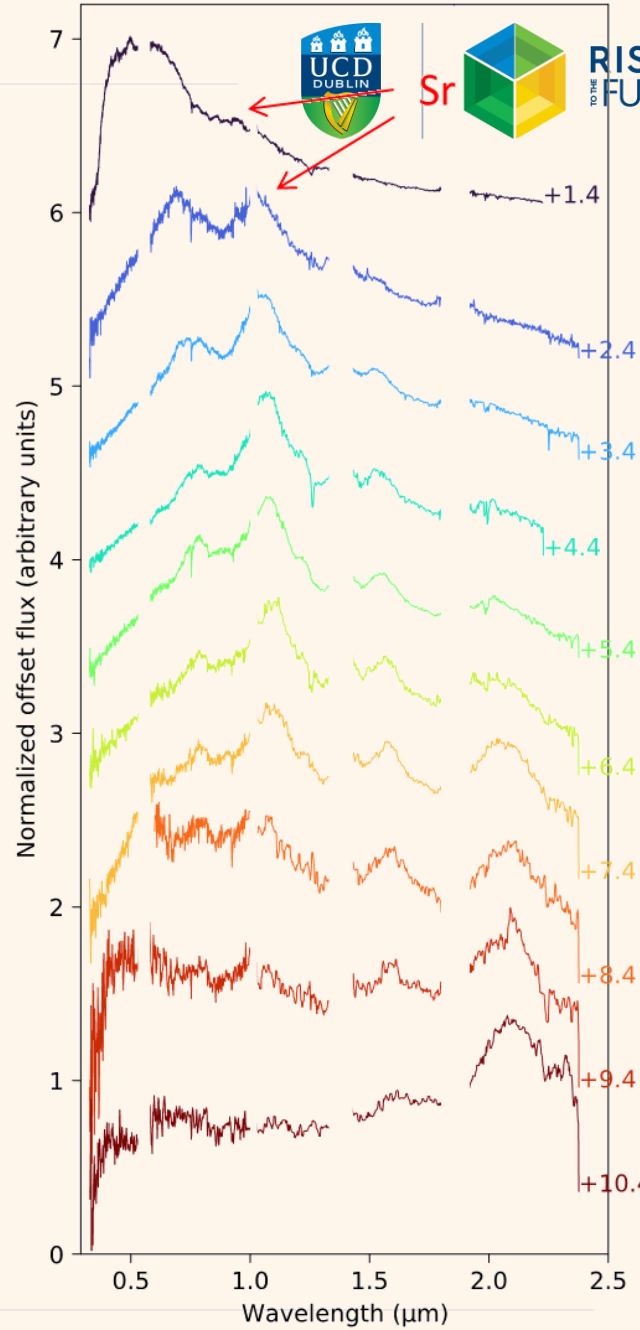
- The rapid neutron capture process(the r-process)
  - least understood element formation
  - makes half of heavy elements



Formation site unknown

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



Kilonova of GW170817

RISING TO THE FUTURE

Motivation

Funded by the European Union

European Research Council
Established by the European Commission

HEAVYMETAL

HOW NEUTRON STAR MERGERS MAKE HEAVY ELEMENTS

ERC Synergy

Speclab

Motivation

Node/PI	Copenhagen/ Darach Watson	Darmstadt/ Andreas Bauswein	Belfast/ Stuart Sim	Dublin/ Padraig Dunne
Team	Daniele Malesani & Johan Fynbo	Oliver Just & Gabriel Martínez-Pinedo	Connor Ballance & Cathy Ramsbottom	Paddy Hayden, Tom McCormack, Emma Sokell, Fergal O'Reilly
Expertise	Astronomical Observations	Merger simulations and nucleosynthesis	Radiative Transfer & Atomic Structure	Experimental Atomic Spectroscopy

Funded by the European Union

European Research Council
Established by the European Commission

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

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- **UCD School of Physics Mechanical and Electronic Workshops**

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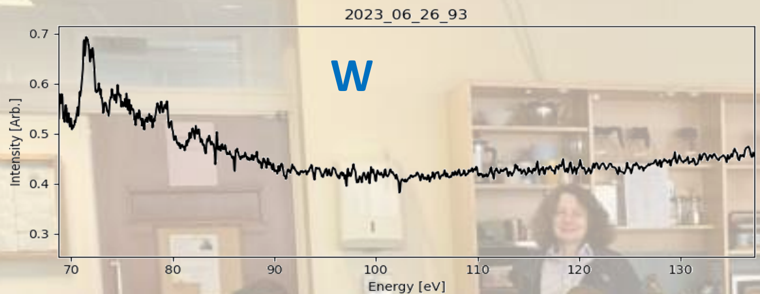
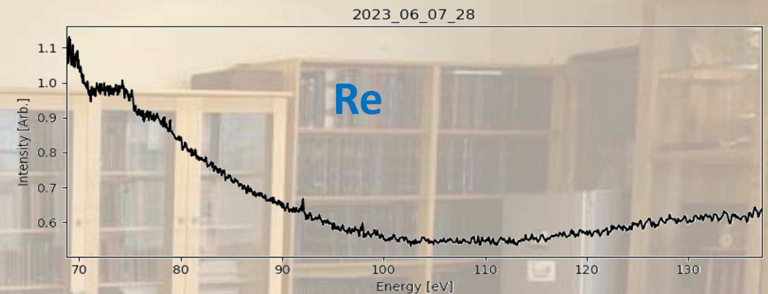
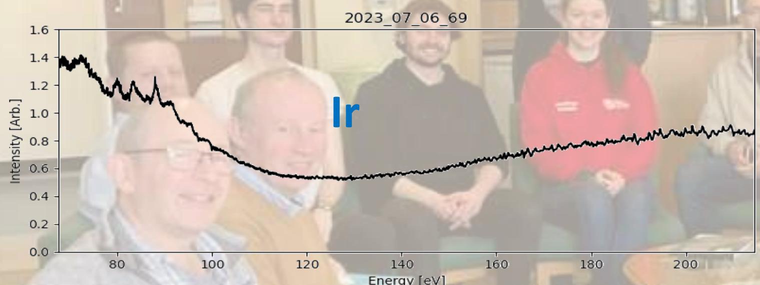
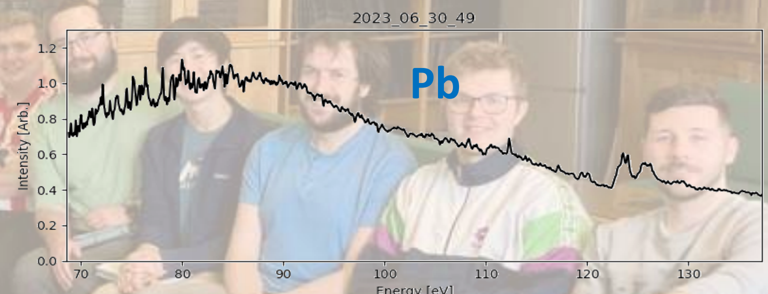
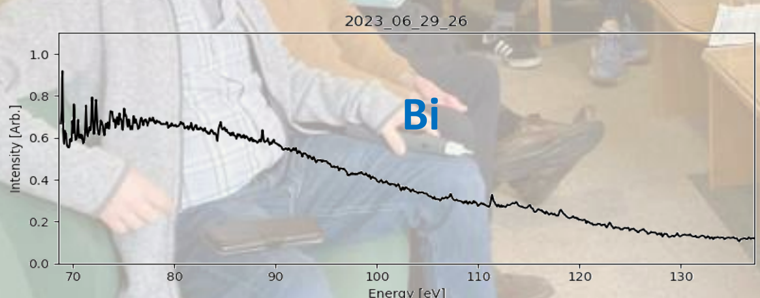
Future Work











Some 6th row transition metals