VUV & XUV Photoabsorption Spectroscopy (and Imaging) with Laser-Plasma Continua

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Outline of Talk

Part I
- ‘Table Top’ Laser Generated Plasma Basics
- The ‘Centre for Laser Plasma Research’ (CLPR)/NCPST
- Laser Plasma Continua - Origin, Brief History & Update

Part II
- The ‘Atomic Photoelectric Effect’
- Case studies in VUV Photoabsorption Spectroscopy
  - ‘Hollow Lithium’ & ‘Krypton-Like’ Ions - Rb$^+$ & Sr$^{2+}$
- VUV Photoabsorption Imaging:
  - Tracking Ba$^+$ ions in an Expanding Laser Plasma Plume
- Photoabsorption/Photoionization of Atoms in Intense Laser Fields - ‘Pump Probe’ Experiments with VUV FELs
Part I -
Table Top Laser-Plasma Light Source (LPLS) Basics
How do you make a laser plasma?

Target

- Emitted - Atoms, Ions, Electrons, Clusters, IR - X-ray Radiation

Vacuum or Background Gas

Laser Pulse
- 1 J/10 ns

Lens

Spot Size = 100 μm (typ. Diam.)
- Φ > 10^{11} W.cm^{-2}
- T_e = 100 eV (~10^{6} K)
- N_e = 10^{21} cm^{-3}
- V_{expansion} ≥ 10^{6} cm.s^{-1}

Plasma Assisted Chemistry
What does a Laser Plasma look like?

- **Target**
- **Expanding Plasma Plume**
- **Incident Laser beam**
- **Substrate**

**PLASMA GENERATION**

**PLASMA EXPANSION**

**FILM GROWTH**
Vacuum and eXtreme-UV Spectral Ranges

VUV - Normal Incidence Region

XUV - Grazing Incidence Region

\[ E(eV) = \frac{hc}{\lambda} = \frac{1239.8}{\lambda}(nm) \]
NCPST/CLPR

Who are we?
What do we do?
NCPST/ CLPR - Who are we?

- Consortium of new and existing laboratories in plasma physics, chemistry and engineering
- Fundamental and Applied Scientific Goals
- CLPR node is divided into 4(6) laboratories focussed on PLD and photoabsorption spectroscopy/ imaging

Staff: John T. Costello, Eugene T. Kennedy, Jean-Paul Mosnier and Paul van Kampen
PDs: Dr. Amit Neogi, Dr. Mohamed Khater - 1 new EU and 1 new SFI Fellow
PGs: John Hirsch, Kevin Kavanangh, Adrian Murphy (JC) Jonathan Mullen (PVK), Alan McKiernan, Mark Stapleton (JPM), Eoin O’Leary & Pat Yeates (ETK)
MCFs: Jaoine Burghexta (Navarra) and Nely Paravanova (Sofia)
NCPST/ CLPR - What do we do?

DCU
*Pico*/Nanosecond Laser Plasma Light Sources
VUV, XUV & X-ray Photoabsorption Spectroscopy
VUV Photoabsorption Imaging
VUV LIPS for Analytical Purposes
ICCD Imaging and Spectroscopy of PLD Plumes

Orsay/Berkeley Synchrotrons
Photoion and Photoelectron Spectroscopy

Hamburg - FEL
Femtosecond IR+XUV Facility Development
V-UV Photoabsorption Imaging Laboratory

Project Title: **VUV Photoabsorption Imaging***
Project Aim: *To develop VUV photoabsorption imaging as a diagnostic of expanding plasma plumes (and other dynamic samples)*

DCU Personnel: John Hirsch, Kevin Kavanagh & John T. Costello
Collaborators: Giorgio Nicolosi and Luca Poletto (Univ. Padua)
First Experiment: RAL-CLF X-ray Laboratory 1999

Project Title: **VUV Thin Film Imaging (DCU - AC Fellowship)**
Project Aim: *Use the VPIF to perform transmission and reflection imaging and spectroscopy of thin films.*

DCU Personnel: Jonathan Mullen and Paul van Kampen

Key Words: Plasma, Laser, Spectroscopy, Imaging, VUV
**Project Title:** Controlled Generation of Laser Plasmas  
**Project Aim:** Optical diagnostics of plasmas formed on non-planar target configurations (e.g., confined in capillaries) in magnetic fields  

**CLPR Personnel:** Pat Yeates and Eugene T Kennedy  
**Collaborators:** J Lunney (TCD)  

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**Project Title:** Dual Laser Plasma Absorption  
**Aim:** Study of fundamentals of photoabsorption by ‘simple’ ions e.g., recently Kr-like ions (Rb$^+$ and Sr$^{2+}$)  

**Key Reference:** A Neogi et al., Phys.Rev.A (Submitted Dec. 02)  

**CLPR Personnel:** J T Costello, E Kennedy, J-P Mosnier and P van Kampen  
**Collaborators:** Gerry O’Sullivan (UCD) and Victor Sukhorukov (State Univ. Rostov)  

**Key Words:** Plasma, Laser, Probe, Spectroscopy, XUV
Project Title: VUV LIPS for Analytical Science

Project Aim: VUV Laser Induced Plasma Spectroscopy Technique (VUV-LIPS)
Record detection limits for C in Steel (2ppm !!)


CLPR Personnel: E O’Leary, M Khater, J T Costello and E T Kennedy

Key Words: Plasma, Laser, Analytical Spectroscopy, LIPS, VUV
Soft X-ray (SXR) Spectroscopy Laboratory (new)

Project Title: SXR Absorption Spectroscopy***

Project Aim: To extend the current Dual Laser Plasma Photoabsorption technique (XUV/ 20 ns) into the X-ray (0.2 - 2nm) and picosecond time domains.

DCU Personnel: Adrian Murphy, Eugene Kennedy and John T. Costello

Collaborators: Padraig Dunne/Gerry O’Sullivan (UCD) and Ciaran Lewis (QUB)

First Experiment: RAL-CLF ASTRA 2002

Key Words: Plasma, Laser, Point Projection Imaging, X-ray,
Project Title: Pulsed Laser Deposition

Project Aim: \textit{PLD of GaN and ZnO}.

Laser Plasma Spectroscopy and Imaging is carried out at DCU to characterize the ablation mechanisms. Films are grown in TCD (soon DCU) and tested in DCU using 10K laser induced photoluminescence technique.


CLPR Personnel: Mark Stapleton, Alan McKiernan and Jean-Paul Mosnier

Collaborators: D O’Mahony & J Lunney (TCD) / E McGlynn & M Henry (DCU)

Key Words: PLD, Laser Ablation, Spectroscopy, Imaging
Who’s yer man talking with Lar?
Laser Produced ‘Rare Earth’ Continua - Physical Origin, History & Update
Firstly - a quick reminder!
Radiative Processes in Laser Plasmas

1. Bound - Bound Transitions - Line Emission/Absorption

\[ A^{n+} + h\nu \cancel{\rightarrow} A^{n++} \text{ or } A^{n+} + e \cancel{\rightarrow} A^{n++} \cancel{\rightarrow} A^{n+} + h\nu' \]

2. Bound - Free Transitions - Recombination/Photoionization*

\[ A^{(n+1)+} + e \cancel{\rightarrow} A^{n+} + h\nu \]

3. Free - Free Transitions - Bremsstrahlung/Inv. Bremsstrahlung

\[ A^{n+} + e(T_1) \cancel{\rightarrow} A^{n+} + e(T_2), T_1 > T_2 \]
Free-Free and Free-Bound processes will give us continuum
Laser Plasma *Rare Earth* XUV Continua

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 O. 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.


What is the Origin of the Continuum?

Continua emitted from laser produced rare-earth (and neighbouring element) plasmas are predominantly free-bound in origin.
But why is no line emission observed?

Line emission is due to complex 4d-4f arrays in (typically) 7 - 20 times ionized atoms

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

Furthermore 4f/5p and 4f/5s level crossing gives rise to overlapping bands of low lying configurations, most of which are populated in the \( \sim 100 \) eV plasma

Result - the summed oscillator strength for each 4d - 4f (XUV) and 5p - 5d (VUV) array is spread out over a supercomplex of transitions producing bands of unresolved pseudo continua (so called ‘UTA’) superimposed on the background continuum

Even expectedly strong lines from simple 4f - 4f arrays are washed out by plasma opacity
There are up to 0.5 million allowed transitions in LS coupling over the ~10 eV bandwidth of a UTA.

In fact this is a lower bound since many additional LS forbidden transitions are ‘switched on’ by the breakdown in LS coupling here - G O’Sullivan et al., J.Phys.B 32, 1893 (1999)
Features of Laser Plasma Continuum Light Sources (LPLS)

- Ease of Production
- Ease of Location
- Purity (Spectral)
- Wide Spectral Coverage - (4 ≤ 200 nm)
- Small Emitting Size (almost point-like, radiography etc.)
- Short Pulse Duration (< 100 ps - 50 ns)*
- Easy Synchronisation (Optical or Electro-Optic)
- **Insensitivity to ambient pressure**
- Shot to shot intensity reproducibility ≤ 5%
- \( \sim 10^{14} \text{ Photons/pulse/sr/nm}^{**} \)

*Depending on exciting laser pulse
Brief History/ Highlights of Laser Plasma Rare-Earth’ Continua

7. First Industrial Application - DuPont - Insulator Band Structure
   VUV Reflectance Spectroscopy - R H French, Physica.Scripta 41, 404 (1990) -
   System subsequently made available commercially from ARC

For a review of the early years including applications in
photoabsorption spectroscopy see:
Recent Developments in LP Continua I

Picosecond LPLS (RAL, UK)

Recent Developments in LP Continua -II

Streak Camera Trace from a tungsten plasma

Time Resolved XUV Photoabsorption Spectra of a Th Plasma (5d - 5ε,f)

XUV Pulse Width Dependence on picosecond laser pulse energy

Recent Developments in LP Continua -III

MBI Source - 2 trains per second/ 25 - 400 Micro-Pulses per train
15 mJ - 0.5 mJ per micropulse & 25 ps pulse duration
XUV Pulse Duration (44 ps - Cu and 73 ps - PET)

Experimental Setups

All Solid State !

Summary - LP Continuum Light Sources

1. Table-top continuum light source now well established
2. Covers Deep-UV to soft X-ray spectral range
3. Pulse duration can be < 100 ps !
4. Continuum flux ~ $10^{14}$ photons/pulse/sr/nm
5. Low cost laboratory source
6. Next step - Working on (100 ps) Laser- (6ns) Pre-plasma source - we already see a flux gain of up to X4 with Cu- (Sm ?)


Problem of plasma debris for work in clean environments - proposals to solve, Michette, O’Sullivan, Attwood,…
Part II - What to do with LPLS?

Photoabsorption/Photoionization Spectroscopy & Imaging of Laser Plasma ‘Sample’ Plumes
The ‘Atomic’ Photoelectric Effect

Simple in principle

\[ A^{n+} + h\nu \leftrightarrow A^{(n+1)+} + e(T), \ T = h\nu - \text{IP} \]

But only for one-electron systems!
(Hydrogen and H-like ions)

**Problem** -
Electron correlation (Coulombic crosstalk) makes possible one photon - many electron excitations!
Why Do (VUV) Atomic (and Ionic) Photoionization?

Why Photoabsorption?
- Access to ground/metastable state (Dark) species
- Electric dipole excitation yields tractable spectra

Why specifically at VUV/XUV photon energies?
- Photoionization continua
- Inner-shell/multi-electron excitations

Data relevant to-
- Astrophysical spectra and models
- Laboratory plasma modelling
- Fundamental many-body theory
- X-ray laser schemes
- ICF
# Photoionization Processes

<table>
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<td></td>
<td></td>
<td>Photoions- Ground/Excited States</td>
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</table>

- **Intense**
- **Monochromatic**
- **Polarised**
- **Pulsed/CW**
- **Tunable**

- **.synchrotron/ Insertion Dev., Upconverted Lasers, LPLS, FEL**
The Dual Laser Plasma Photoabsorption Set-up

No tuning required
No vapour required

Flexible
Neutral/Multiplycharged/Refractory Elements

\(\Delta x, \Delta T, I(W/cm^2)\) ➞ Species choice

Relative Absorption Cross Section
\(\sigma_{NL} = \ln(I_0/I)\)

XUV - Dual Laser Plasma Pump-Probe (DCU)

Wide Energy Capture (20 - 200 eV)  $E/\Delta E \sim 1500, \Delta T \sim 20$ ns
Relative Cross Sections  Resonances Positions/strengths

More efficient use of large facility beamtime !!
Photoionization: Beyond the single-electron approximation

‘Hollow Lithium’
Why Lithium?

Simplest - many electron atom

Valence (Direct) ionization (L-shell ionization)

Direct double and even triple ionization (K and/or L shell)

Inner shell single and double electron excitation possible - K, KL and K² (Hollow Atom)
First consider (VUV) photoionization of He

\[
\text{He: } 1s^2(^1S) + h\nu_{\text{VUV}} \rightarrow \text{He}^+ (1s) + \epsilon_1, \text{ IP} = 24.58 \text{ eV}
\]

1-electron photoionization just like the H case, ergo no problem here!

However it is also possible to induce 1-photon, 2-electron excitations -

**Resonant** 1 electron ionization pathway (channel)

\[
\text{He: } 1s^2(^1S) + h\nu_{\text{VUV}} \uparrow \text{He}^{**}2s2p (^1P) \downarrow \text{He}^+ (1s) + \epsilon'1
\]

**Direct** 1 electron ionization pathway (channel)

Also holds for He-like ions, e.g., Li^+. 
Simplified Energy Level Diagram for He

- $1s^2 (1S)$
- $1s^2p (1P)$
- $2s2p (1P)$
- $1s\infty p (1P)$
- He Continuum - $He^+$

Autoionization
Two pathways to a final (quantum) state ⇒ We must observe *interference fringes* in photoionization.

Profile parameters, $q$ & $\Gamma$

$$f(\varepsilon) = \frac{(q + \varepsilon)^2}{1 + \varepsilon^2}, \varepsilon = \frac{2(E - E_0)}{\Gamma}$$

$q \propto$ ratio of probability amplitudes for resonant/direct ionization.
What About Many-Electron Atoms & Ions?

Take the simplest case - Li or Li-like (3e) Ions

1. Direct (valence) ionization \(-1s^22s(2S) + \hbar\nu_{XUV} \uparrow 1s^2(1S) + \epsilon_1\)

2. K\(^{-1}\) (K shell) \(-1s^22s(2S) + \hbar\nu_{XUV} \uparrow 1s2snp (2P) \uparrow 1s2s(1S) + \epsilon_1\)

3. K\(^{-1}\)L\(^{-1}\) (K/L sh.) \(-1s^22s(2S) + \hbar\nu_{XUV} \uparrow 1s2lnp (2P) \uparrow 1s2l(1S) + \epsilon_1\)

4. K\(^{-2}\) (K shell) \(-1s^22s(2S) + \hbar\nu_{XUV} \uparrow 2s^22p (2P) \uparrow 1s^2(1S) + \epsilon_1\)


First optimise the atomic (Li) or ionic (Li\(^+\)) fraction in the plasma!

First try Li\(^+\)

\[ 1s2s(2S) + h\nu_{XUV} \rightarrow 1snp (2P) \]  
(He-like Li) Rydberg series

Variation in Li\(^+\) absorption with time after Li plasma breakdown & region of plasma plume probed
Ditto for Li Optimisation

\[ 1s^22s(2S) + h\nu_{\text{XUV}} \uparrow 1s2snp (2P) \]

\[ \uparrow 1s2s(1S) + e^- \]

Variation in Li absorption with time after Li plasma breakdown & region of plasma plume probed

Fig. 7 (a) K-shell absorption spectrum of atomic lithium plasma. Indicated on the plot are the resonance lines of valence excited Li\(^+\) and Li\(^+\). Their weakness is a measure of the purity of the spectrum with respect to the ionization stage and state of excitation. (b) Temporal and spatial evolution of ground-state lithium atoms in a dye-laser-produced lithium plasma; \(\Delta x\) is the spatial position (in millimeters) of the target with respect to the optical axis of the system and \(\Delta z\) is the interlaser time delay in nanoseconds.
Once optimised we looked for the 2-electron resonances!

First member of famous Madden-Codling Series - but in He-like Li!

Li$: 1s^2(1S) + h\nu_{XUV} \rightarrow 2s2p (1P) \rightarrow \text{Li}^{2+}(1s) + \epsilon'$

Actually first observed in 1977 by P K Carroll and E T Kennedy, PRL 38, 1068 (1977)

$1s^22s(2S) + h\nu_{XUV} \rightarrow 2s^22p (2P) \rightarrow 1s^2(1S) + \epsilon$

*Satellite to 1st member of the famous Madden & Codling 2e series in He: $1s^2(1S) + (weak) h\nu_{XUV} \rightarrow 2s2p (1P)^

“First Observation of a Photon Induced Triply Excited State in Atomic Lithium”

Then we moved to a synchrotron - Hasylab, DESY - Beamline BW3
Since our first report 1994/95 there have been over 100 experimental/theoretical studies published on Hollow Li.
High resolution photoion measurement of the $1s^22s - 2s^22p$ resonance of Li
NEXT - Example of a DLP Experiment on Ions

XUV Photoabsorption along an Isoelectronic Sequence - Kr-like Ions
Why Photoionization of Atomic Ions?

Not a lot known so far - see J. West, JPB 34, R45 (2001)

Require

♣ Sufficiently high density of ions
♣ Appropriate high (VUV/XUV) photon flux sources

Ideally like to employ

♣ Photoabsorption, Photoion/electron spectrometry (PIS/PES)
♣ Fluorescence, Co-incidence & Angle Resolved PES etc.

Laser techniques-
RLDI: Lucatorto & McIlrath, NIST
Dual Laser Plasma: Dublin, Padua, Belfast, Tokyo,...

Synchrotron + ion beam techniques-
Photoion: Aarhus/Daresbury, Orsay, Berkeley,...
Photoelectron: Orsay, Berkeley,...
Why Specifically Kr-like Ions?

Electronic Configuration

$4s^24p^6$

Prototypical high-Z closed shell atom - beyond simple Fano theory

30+ years of research in both single and multiphoton ionization

Will the photoionization dynamics ($q/\Gamma$) change (a little or a lot?)

How will current many electron photoionization theory stand up?
XUV Photoabsorption along an Isoelectronic (Kr-like) Sequence - I

\[ 4s^24p^6 + h\nu_{\text{VUV}} \rightarrow 4s4p^6np + 4s^64p^4nl' \rightarrow \text{Kr}^+(4s^24p^5) + \epsilon' l \]


NB: Resonance profile quite different
XUV Photoabsorption along an Isoelectronic Sequence - II

\[ \sigma_{4s}(\omega) = \text{Re}^2 \left\langle 4s^14p^6n p j J \mid D \mid 0 \right\rangle + \text{Im}^2 \left\langle 4s^14p^6n p j J \mid D \mid 0 \right\rangle \]

\[ q = -\frac{\text{Re} \left\langle 4s^14p^6n p j J \mid D \mid 0 \right\rangle}{\text{Im} \left\langle 4s^14p^6n p j J \mid D \mid 0 \right\rangle} \]
Photoelectron Spectroscopy of Ions

Merged photon and ion beams

LURE (Orsay) \( \text{Ca}^+ \)

Difficulties

- Electron spectrometer efficiency low
- Short interaction length
- Low densities (<<atom experiments)

SuperACO, Aarhus, Photon Factory, Advanced Light Source.
- Photoion spectrometry
- Absolute cross sections

So far!! Only PES for Resonant Photoionisation of \( \text{Ca}^+ \)
- \( 3p \rightarrow 3d \): Very high cross section process

J. B. West: Topical Review

Photoelectron Spectroscopy of Ions – FEL??
Photoionization with LPLS - A Plasma Plume Diagnostic ?
Photoionization as a plasma diagnostic

**VUV Photoabsorption Imaging**

Pass a *collimated* VUV beam through the plasma sample and measure the spatial distribution of the absorption.

\[ I = I_0 e^{-\sigma \int n(l) \, dl} \]

VUV Photoabsorption Imaging Facility – ‘V-P-I-F’

Experimental set-up
The obligatory picture!!
VPIF Specifications

Time resolution: ~20 ns (200 ps with new laser)
Inter-plasma delay range: 0 - 10 µsec
Delay time jitter: ± 1 ns
Monochromator: Acton™ VM510 (f/12, f=1.0 m)
VUV photon energy range: 10 - 35 eV
VUV bandwidth: 0.03 eV [@30 eV]
Detector: Andor™ CCD, 1024 x 2048/13 µm x 13 µm pixel
Spatial resolution: ~150 µm (H) x 150 µm (V)
Why use a laser plasma light source?
The VUV continuum pulse duration is ~ 20 nsecs; hence we have automatic temporal resolution of this order (< 250 ps possible).

Since the laser plasma source has a coherence length not exceeding a few µm’s image analysis is not complicated by interference patterns on the image.

Why VUV light?
VUV light can access resonance lines of all atoms and moderately charged ions.

VUV light can access electron density regimes that are inaccessible to visible light photoabsorption/ shadowgraphy techniques.

VUV beams suffer from significantly reduced refraction compared with more conventional visible beams which simplifies image processing and analysis.
In Addition!

VUV light can be used to measure photoionization continua of atoms and moderately charged ions. In particular it can be used to measure resonant photoionization, e.g.,

\[ \text{Ba}^+(5p^66s\,^2S) + \nu \rightarrow \text{Ba}^+(5p^56s\,5d\,^2P) \rightarrow \text{Ba}^{2+} (5p^6\,1S) + e^- \]

The advantage of this process over conventional optical absorption measurements on laser ablated/plasma plumes is that almost all photons are converted directly into photoelectrons - no radiation transfer to worry about!

So we can use Beer's law:

\[ I(E) = I_0(E) e^{-\sigma(E) \int n(l) dl} \]

The \textbf{ABSOLUTE} photoionization cross-section for \( \text{Ba}^+ \) has been measured:

[\text{Lyon et al., J.Phys.B 19, 4137 (1986)}]

\textbf{Ergo!}

We should be able to extract maps of column density 'NL' = \( \int n(l) dl \)
Quantities extracted from the I and Io images

Absorbance: \[ A = \log_{10}\left(\frac{\int I_0(x, y, t, \lambda) d\lambda}{\int I(x, y, t, \lambda) d\lambda}\right) \]

Equivalent Width:

\[ W_E = \Delta \lambda \left(\frac{\int [I_0 - I] d\lambda}{\int I_0 d\lambda}\right) \]

\[ W_E = \int [1 - e^{-\sigma(\lambda)NL}] \]
Time Resolved Absorbance Maps - Ba\(^+\), 5p - 6d Resonance @ 467 Å
Equivalent Width \([W_E]\) Distributions
Convert from $W_E$ to NL

Compute $W_E$ for a range of NL and fit a function $f(NL)$ to a plot of NL vs. $W_E$

Apply pixel by pixel

$$W_E = \int \left[ 1 - e^{-\sigma(\lambda)NL} \right]$$
Result - Column Density [NL] Maps

(a) 100 ns - (f) 500 ns
One can also extract the plume 'dark front' velocity!

Figure 4.22. Plot of the plasma plume dimension along the direction of expansion (perpendicular to the target) with time. (see text for details)
And compare it with simple models of plasma expansion.
Next steps in fundamental photoionization studies?

Atoms and Molecules in Laser Fields

1. Attosecond pulse generation/ HHG

2. Photoionization of ‘state prepared’ species
   (a) Weak Optical + Weak X-VUV
   (b) Intense Optical + Weak (Intense) X-VUV

3. Atoms, Molecules, Cluster & Ions in Intense Fields
   *(Multiple-Photo and Optical Field-Ionization)*
Experiment Type I

(a) Weak Optical + Weak X-VUV
Photoionization of Laser Excited Species (Weak Optical + Weak X-VUV)


Experimental Setup

Level Scheme
CIS (expt. & theor.)

Angle Resolved PES

Figure 5. Experimental (a) and calculated (b)–(d) spectra of the autoionizing resonances Na 2p\(^5\)3s3p excited from Na 2p\(^6\)3p \(^3\)P\(_{1/2}\).

Figure 6. Two \(\eta\)-scans (\(\Theta = 0^\circ\) and \(90^\circ\)) and one \(\Theta\)-scan (\(\eta = 0^\circ\)) for the resonance 2p\(^2\)(3s3p \(^3\)P) \(^5\)S\(_{1/2}\). Full curves according to formulae (1) for \(J = \frac{1}{2}\), \(\rho_1 = 0.38\) and \(\rho_2 = 0.95\).
So type I (weak laser excitation) experiments appear straightforward with current laser technology.
Experiment Type II

Intense Optical + Weak (Intense) XUV
Two subsets of experiments

II-A.
Direct photoionization in a non-resonant laser field

II-B
Resonant photoionization in a resonant laser field
Let's look at II-A - ‘Direct photoionization in a non-resonant laser field’ first*

First Laboratory demonstrations of (non-resonant) photoionization of atoms in intense fields

*Slides provided by Patrick O'Keefe and Michael Meyer, LURE, ORSAY!
Ponderomotive streaking of the ionization potential as a method for measuring pulse durations in the XUV domain with fs resolution

presence of IR:
- shift of IP
- broadening of PES peaks
- sidebands

Test-experiments at LLC: M. Meyer, P.O’Keefe (LURE), A.L’Huillier (LLC)
fs-laser system: Ti:Saph. 800 nm, 50 fs, 1 kHz
VUV --> HHG, $\Delta T \approx 30$ fs, 1 kHz,
IR --> up to 0.5 mJ --> 1-10 TW/cm$^2$
PES: magnetic bottle spectrometer
- high angular acceptance
- high energy resolution for $E_{\text{kin}} < 10$ eV
Cross correlation experiments using high order harmonics

$\Delta T$ [fs]

$E_{\text{kin}}$ (eV)

$E = 15.8$ eV

$VUV$

$e^-$

IR

$\lambda$ (laser) = 800nm

$H_{11} = 17$ eV

$H_{13} = 20$ eV

$H_{15} = 23$ eV

$H_{17}$

$H_{19}$

$H_{21}$

$\text{Generation (HHG)}$

$\text{Ar}^+ 3p^5$

$\text{Ar} 3p^6$

$E_{\text{kin}} (eV)$

$50$

$0$

$-50$

$5$

$10$

$15$

$20$
Cross correlation experiments using high order harmonics

\[ E_{\text{kin}} = h \nu - IP - U_p \]
\[ U_p [\text{eV}] = \alpha I [\text{W/cm}^2] \]

\[ I_{\text{eff}} \sim \Delta T \quad \text{------} \quad \Delta E(\Delta T) \]

\[ I_{\text{SB}}(\Delta T) \sim \int_{-\infty}^{+\infty} f[I_{\text{IR}}(t - \Delta T)] I_{\text{VUV}}(t) \, dt \]

Test Experiment (HHG):

VUV \(\approx\) 30 fs \quad IR \approx 50 fs \quad I (VUV) \approx 10^6 - 10^7 \text{ photons / pulse}

I (IR) \approx 5 \text{ TW/cm}^2 \quad \text{------} \quad \Delta E(\text{max}) = 180 \text{ meV}
\[ \Delta E / \Delta T \sim 30 \text{ meV} / 10 \text{ fs} \]

Single pulse spectra: \(\sim 3 \text{ electrons / pulse}\)
\[ \text{---} \quad \Sigma 30\,000 \text{ shots (30 sec / } \Delta T) \]

I (FEL) \approx 10^{13} \text{ photons / pulse}
But far more interesting would be:

Type IIB-Experiments
Resonant photoionization in an intense/resonant fields

=> Study intensity controlled (tuned) autoionization !!
Ideally we would like a VUV/ XUV source with lots of photons to do type IIA & IIB experiments !!
It’s gotta be a VUV/XUV FEL!
X-VUV FELs + Femtosecond OPAs - The Ultimate Photoionization Expt?

- Tuneable: NOW! 80 - 110 nm (20 - 60 nm in 2004)
- Ultrafast: 100 fs pulse duration
- High PRF: 1 - 10 bunch trains/sec with up to 11315 pulses/bunch
- Energy: Up to 1 mJ/bunch
- Intense: 100 $\mu$J (single pulse) / 100 fs / 1 $\mu$m => $10^{17}$ W.cm$^{-2}$

• Moving to XUV (2005) and X-ray (2010):
  • Need a Linac + insertion devices => Fraction of a GigaEuro!!

Project Title: ‘Pump-Probe’ with DESY-VUV-FEL (EU-RTD)

Aim: FEL + OPA synchronisation with sub ps jitter

Key Reference: http://tesla.desy.de/new_pages/TDR_CD/start.html

Personnel: MBI, DESY, CLPR-DCU, LURE, LLC, BESSY
First Experiments at DESY Phase 1 FEL
Nature 420 (2002) 482
Pump-probe experiments in the gas phase (project: II-02-037-FEL)

M. Meyer, LU.R.E., Orsay, France

IIA - characterization of fs - FEL pulses
- cross correlation experiments (Ar, Xe, He)

IIB - application of fs - FEL pulses
- interference in molecular dissociation (H₂)
- coupling of autoionization states (Xe, He, Li, Ca, Ca⁺, Ba, Ba⁺)

Participating groups:
L.U.R.E., Orsay, France
M. Meyer, L. Nahon, P. O'Keeffe
Lund Laser Center / MAX-Lab, Lund, Sweden
J. Larsson, A. L'Huillier, S. Sorenson
Max-Born Institut, Berlin, Germany
I. Will, H. Redlin

HASYLAB, Hamburg, Germany
J. Feldhaus, E. Ploenjes, K. Tiedke
Dublin City University, Dublin, Ireland
J. Costello & E. Kennedy
Type IIA-Experiments
Non resonant photoionization in an intense laser field

Just repeat Meyer et al. experiments with High Harmonic Source replaced by a VUV FEL

=> synchonise IR + FEL pulses on a fs timescale

But far more interesting would be:

Type IIB-Experiments
Resonant photoionization in an intense/resonant fields

=> Study intensity controlled (tuned) autoionization !!
Proposed experiment at the FEL

**Exp.:** Two-photon double-resonant excitation  
**FEL:** $h\nu = 65.1$ eV (tunable)  
**Laser:** $\lambda = 750 - 800$ nm (tunable)

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**Coupling of He Doubly Excited States**

- $2s2p$ $^1P - 2s3d$ $^1D$
- Intense Laser
- $20$ fs (34 meV)
- $VUV$

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A. I. Magunov, I. Rotter and S. I. Strakhova  
H. Bachau, Lambropoulos and Shakeshaft  
*PRA 34, 4785 (1986)*
Laser-Induced Transitions between Doubly Excited States of Helium

Gas target
Longer wavelength (60 eV)
Déjà vu !!!

What about more complex atoms in intense laser fields?

‘Simplest’ complex atom is Li!
Laser-Excited Hollow Li

$1s^22s^2(2S) + (\text{weak})h\nu_{\text{Laser}} \rightarrow 1s^22p^2(2P) + (\text{weak})h\nu_{\text{XUV}} \rightarrow 2s2p^2(2D)$

Experiment - D Cubannes et al. PRL 77, 2194, (1996)

Hollow Li in an intense laser field

$1s^22s^2(2S) + h\nu_{\text{XUV}} \rightarrow \{2s^22p^2(2P) + (\text{intense})h\nu_{\text{Laser}} \leftrightarrow 2s^23d(2D)\}$

Reminder - Hollow Li 2e Resonance

First member of famous Madden-Codling Series - but in He-like Li!

$$\text{Li}^+: 1s^2(1S) + h\nu_{\text{XUV}} \uparrow 2s2p (1P) \downarrow \text{Li}^{2+}(1s) + \varepsilon' l$$

Actually first observed in 1977 by P K Carroll and E T Kennedy, PRL 38, 1068 (1977)

$$1s^22s(2S) + h\nu_{\text{XUV}} \uparrow 2s^22p (2P) \downarrow 1s^2(1S) + \varepsilon l$$

Satellite to 1st member of the famous Madden & Codling 2e series in He:
$$1s^2(1S) + \text{weak} h\nu_{\text{XUV}} \rightarrow 2s2p (1P)$$

“First Observation of a Photon Induced Triply Excited State in Atomic Lithium”
Photoionization of Atoms in Intense Fields - Predictions

Could this be done with a laser plasma VUV/XUV source?
YES!

In principle you just cross the sample with intense laser and weak XUV beams BUT!

The ‘Sample’ must be dilute - otherwise RLDI will dominate and drive it into an ionized state.

Sample dilute => DLP photoabsorption experiment is unsuitable.

Can still use a laser plasma X-VUV source BUT

Need wavelength selection and high (average) X-VUV intensity.

More sophisticated Photoion (or electron) spectroscopy needed.

Count rate low - ~ 1 ion/laser shot for He with $V_{\text{int}} \sim 10^{-3}$ cm$^{-3}$!
But - the Ca & Ca$^+$ 3p-3d resonances have:

1. Cross sections up to 3000 MB vs. < 1MB for Li
2. An excitation widths up to 100s meV
3. A VUV (normal incidence) excitation energy (31 eV)

Scheme- Ca$^+$:

\[ 3p^64s (^2S) + h\nu_{\text{XUV}} \rightarrow \{3p^53d4s (^2P) + h\nu_{\text{Laser}} \leftrightarrow 3p^53d4p (^2D)\} \]

Possible collaboration with QUB on ion beam/TOF?

Many other possibilities - Ca, Ba, Sr, Na, K and their ions
Collaborators
Photoionization Summary
Conclusions

- Need VUV/XUV/X-ray source with shortest possible pulse width commensurate with providing high flux/pulse and high average power (continuously tunable) radiation
- Must be flexible on pulse repetition rate x energy per pulse product!
- Combining with a high peak power tunable source opens up possibilities to study photoionization of a whole range of atoms, ions, etc. in weak and intense resonant fields for the first time
- Good laser, laser plasma source and photoion diagnostics highly desirable