

Vacuum-UV Two-photon ionization of Kr

Varvarezos Lazaros

Supervisors:

Prof John Costello

Dr Bill Brocklesby

Dr Lampros Nikolopoulos

EXTATIC Welcome Week 2017

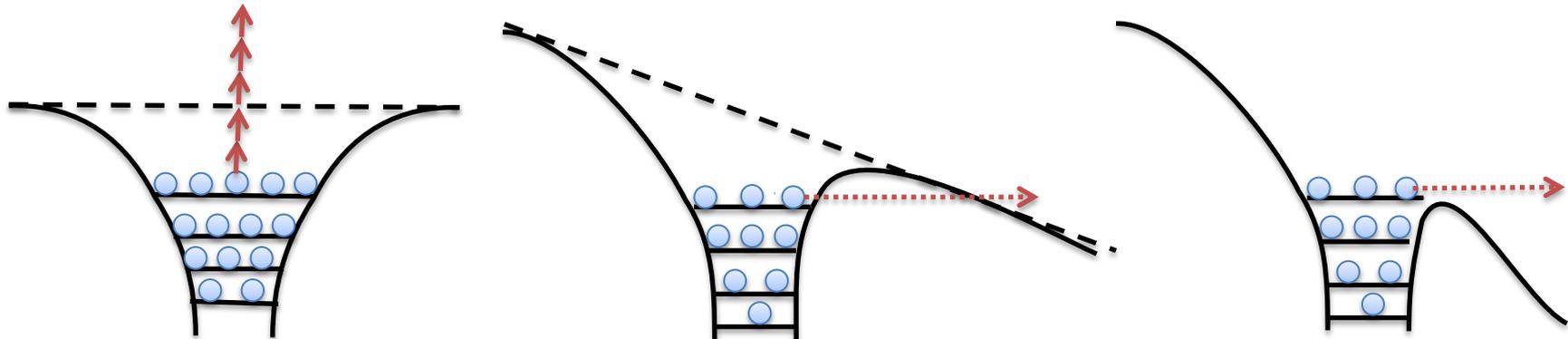
Czech Technical University in Prague, Prague

23rd Sep. 2017

Outline

- ✓ Ionization in the IR vs the VUV to X-ray region
- ✓ Atomic TPDI : Sequential vs Direct process
- ✓ The Kr ionization in the VUV region
- ✓ The experiment in FLASH
- ✓ Angular distribution and the extracted anisotropy parameters
- ✓ Conclusions and future work

Ionization of atoms in the IR spectral region



Multiphoton Ionization

Tunneling Ionization

Field Ionization

Laser Intensity



How can we distinguish between the different regimes ?

Keldysh parameter

Ionization of atoms in the IR spectral region

$$\sqrt{\frac{I_P}{2U_P}}$$

I_P : Ionization potential , U_P : Ponteromotive potential

$$U_P = 9.3 \cdot 10^{-14} I \left(\frac{W}{cm^2} \right) \lambda^2 (\mu m^2)$$

L. V. Keldysh, "Ionization in the field of a strong electromagnetic wave," *Sov. Phys. JETP*, vol. 20, no. 5, pp. 1307–1314, **1965**.

Multiphoton Ionization : $\gamma \gg 1$, Tunnel Ionization : $\gamma \approx 1$, Field Ionization : $\gamma \ll 1$

Example : Ionization of Argon

Ti:Sapphire laser $\lambda = 800 \text{ nm}$, $I_P = 15.76 \text{ eV}$

$$I = 10^{12} \left(\frac{W}{cm^2} \right) \Rightarrow \gamma = 12 \text{ MPI}$$

$$I = 10^{14} \left(\frac{W}{cm^2} \right) \Rightarrow \gamma = 1.2 \text{ TI}$$

$$I = 10^{16} \left(\frac{W}{cm^2} \right) \Rightarrow \gamma = 0.12 \text{ FI}$$

Atomic ionization in the FEL era

Generation of ultrashort pulses in the VUV up to X-ray region became possible with the advent of FELs

J. Andruszkow *et al.*, “First observation of self-amplified spontaneous emission in a free-electron laser at 109 nm wavelength,” *Phys. Rev. Lett.*, vol. 85, no. 18, pp. 3825–3829, **2000**.

- ✓ In VUV and X-ray radiation, the photon energy approaches or even exceeds the ionization potential !

- ✓ The Keldysh parameter becomes unsuitable for characterizing nonlinear interactions !

- ✓ In that case it's the comparison between the photon energy and the ponderomotive potential that defines whether the interaction is perturbative or not !

Atomic ionization in the FEL era

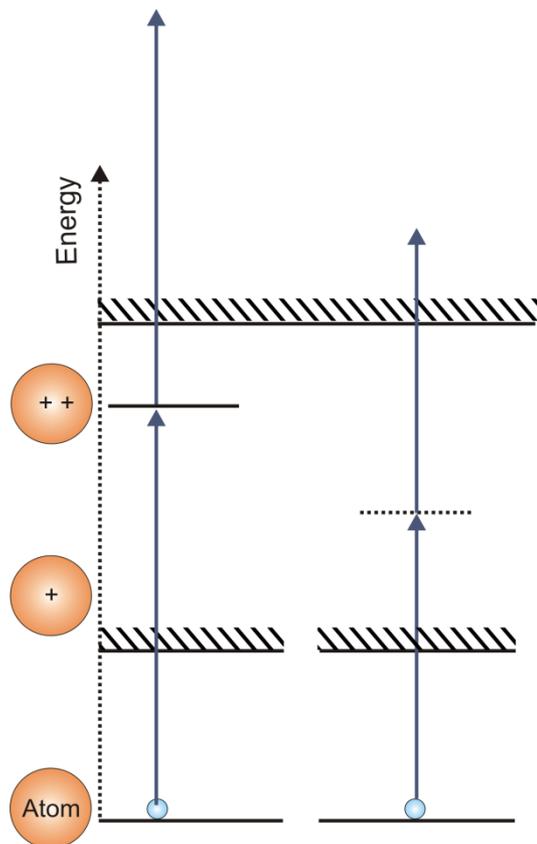
Non-perturbative behavior if $U_p \geq \omega \hbar$

Example :

λ (nm)	photon energy (eV)	Intensity where $U_p = \omega \hbar$ in (W/cm^2)
800	1.55	$2.6 \cdot 10^{13}$
40	31	$2.1 \cdot 10^{17}$

For FEL lasers, **multi-photon** ionization is the primary process and will involve *few photons*

TPDI Sequential vs direct ionization

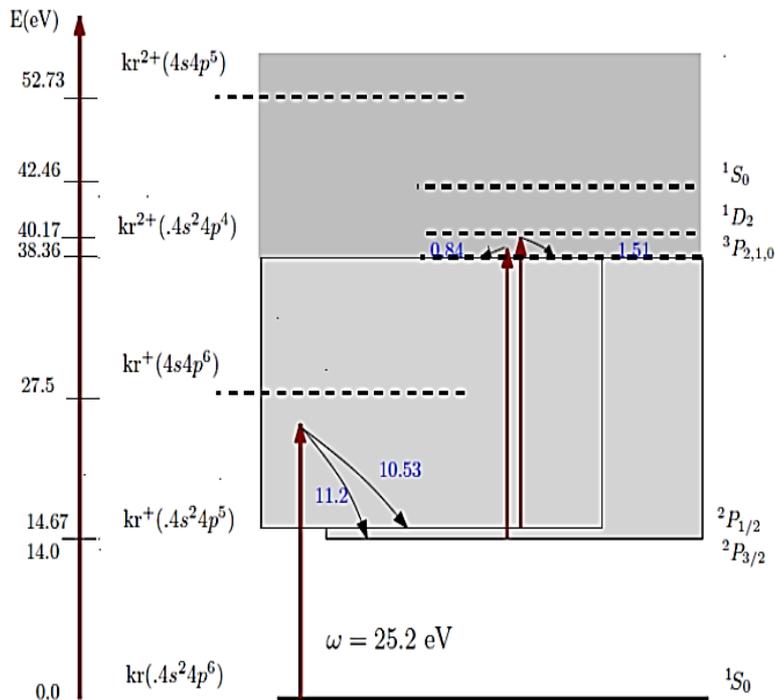


When the photon energy is **larger** than the ionization potential of the singly charged ion, **sequential TPDI** dominates

For Kr the **4p** ionization potential is **24.36 eV**, while the **3d** threshold lies at **96 eV**.

In our study, the photon energy was set at **25.2 eV**

The Kr case



Single-photon ionization of Kr atoms by synchrotrons

Becker U and Shirley D A 1996 *VUV and Soft X-Ray Photoionization and U Becker and D A Shirley (New York: Plenum) p 135*

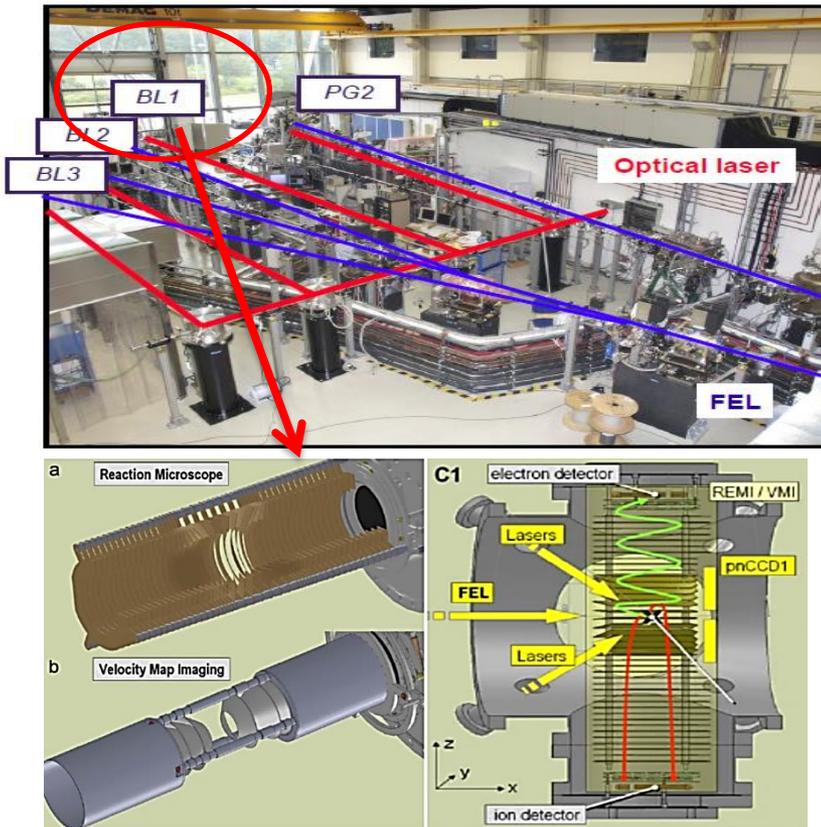
Only recently....

M. Braune *et al.* **2016** *J. Mod. Opt.* **63** 324

And theoretical treatment of the problem...

S. Fritzsche **2009** *J. Phys. B: At. Mol. Opt. Phys.* **42** 145602

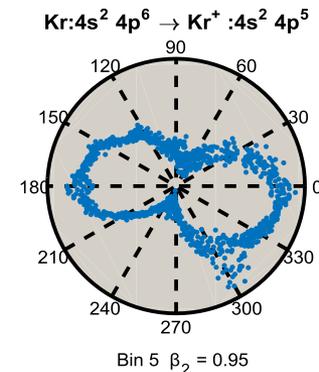
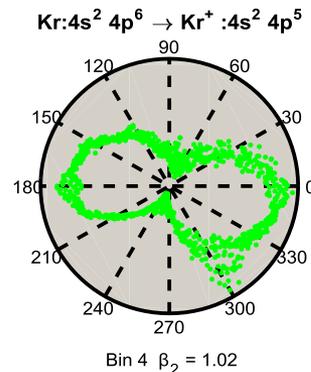
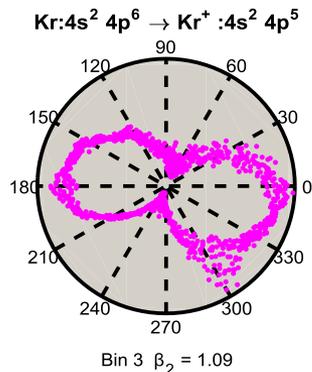
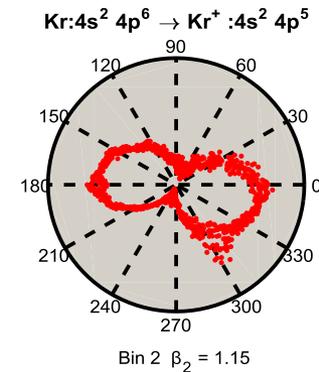
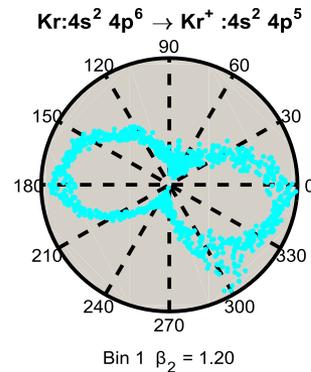
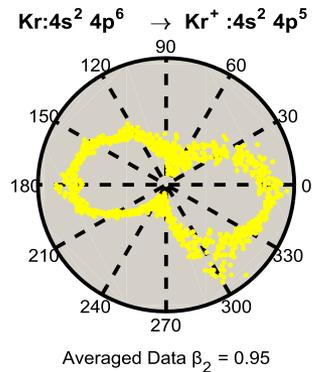
Experiment in FLASH



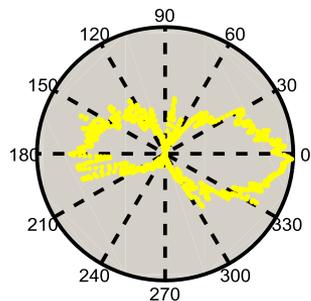
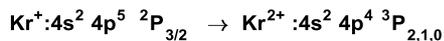
FEL parameters

- ✓ Pulse duration **80 fs** (pulse to pulse fluctuation between 60fs and 100 fs)
- ✓ Pulse energy at the focus : **3μJ to 10μJ**
- ✓ Focal spot : **50 μm**
- ✓ Peak intensities between **2 10¹² w/cm²** and **5 10¹² w/cm²**
- ✓ VMI resolution : **0.3 eV**

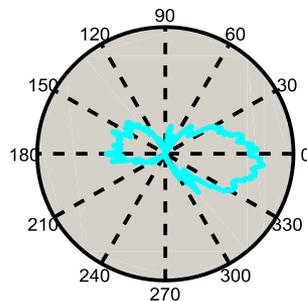
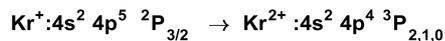
The first step: singly ionized Kr atoms



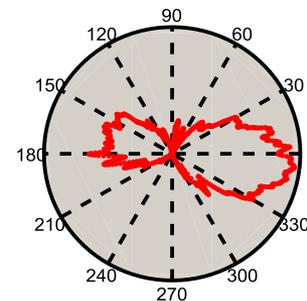
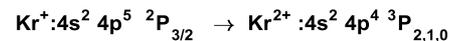
The second step : Ionization of the Kr⁺



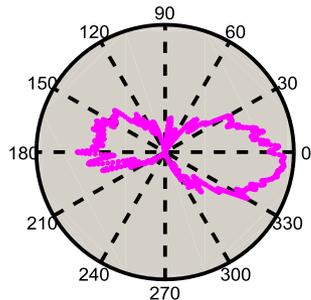
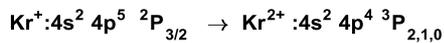
Averaged Data $\beta_2 = 1.55 \beta_4 = 0.24$



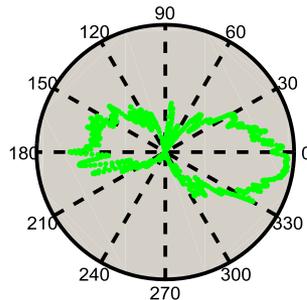
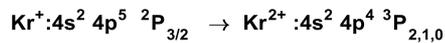
Bin 1 $\beta_2 = 2.11 \beta_4 = 0.24$



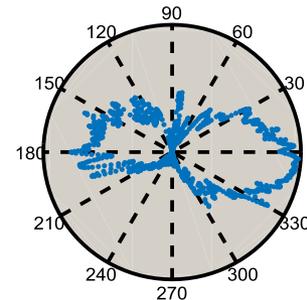
Bin 2 $\beta_2 = 2.10 \beta_4 = 0.33$



Bin 3 $\beta_2 = 1.95 \beta_4 = 0.34$

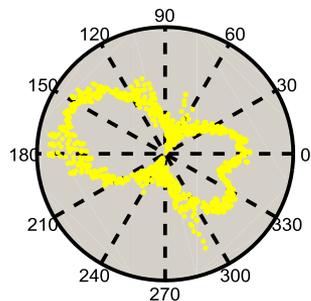
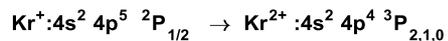


Bin 4 $\beta_2 = 1.70 \beta_4 = 0.28$

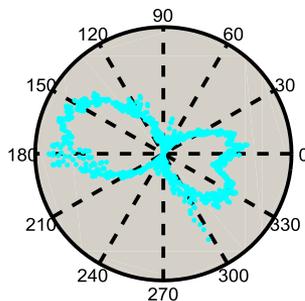
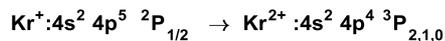


Bin 5 $\beta_2 = 1.57 \beta_4 = 0.22$

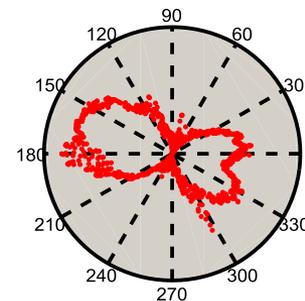
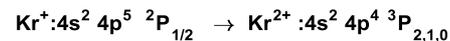
The second step : Ionization of the Kr^+



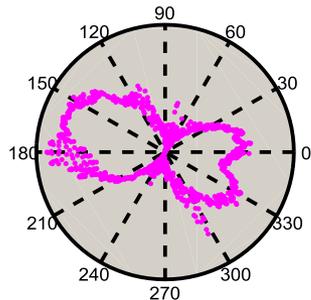
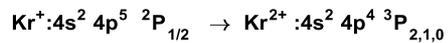
Averaged Data $\beta_2 = 0.82 \beta_4 = 0.08$



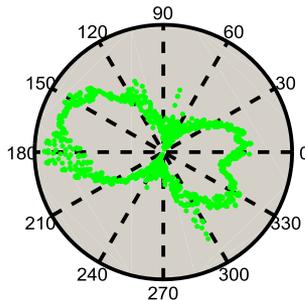
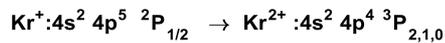
Bin 1 $\beta_2 = 1.55 \beta_4 = 0.21$



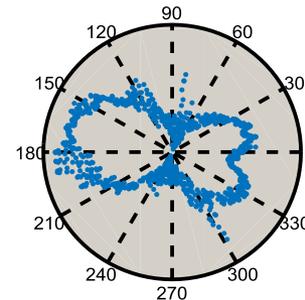
Bin 2 $\beta_2 = 1.38 \beta_4 = 0.15$



Bin 3 $\beta_2 = 1.22 \beta_4 = 0.11$



Bin 4 $\beta_2 = 1.07 \beta_4 = 0.10$



Bin 5 $\beta_2 = 1.08 \beta_4 = 0.08$

The β -parameters

Channel $\text{Kr}:4s^2 4p^6 \rightarrow \text{Kr}^+ :4s^2 4p^5$

Averaged data	Bin 1	Bin 2	Bin 3	Bin 4	Bin 5
$\beta_2 = 0.95$	$\beta_2 = 1.20$	$\beta_2 = 1.15$	$\beta_2 = 1.09$	$\beta_2 = 1.02$	$\beta_2 = 0.95$

Channel $\text{Kr}^+ :4s^2 4p^5 \ ^2P_{1/2} \rightarrow \text{Kr}^{2+} :4s^2 4p^4 \ ^3P_{2,1,0}$

$\beta_2 = 0.84$	$\beta_2 = 1.62$	$\beta_2 = 1.44$	$\beta_2 = 1.27$	$\beta_2 = 1.11$	$\beta_2 = 1.10$
------------------	------------------	------------------	------------------	------------------	------------------

Channel $\text{Kr}^+ :4s^2 4p^5 \ ^2P_{1/2} \rightarrow \text{Kr}^{2+} :4s^2 4p^4 \ ^3P_{2,1,0}$ (with β_4 included)

$\beta_2 = 0.82$	$\beta_2 = 1.55$	$\beta_2 = 1.38$	$\beta_2 = 1.22$	$\beta_2 = 1.07$	$\beta_2 = 1.08$
$\beta_4 = 0.08$	$\beta_4 = 0.21$	$\beta_4 = 0.15$	$\beta_4 = 0.11$	$\beta_4 = 0.10$	$\beta_4 = 0.08$

Channel $\text{Kr}^+ :4s^2 4p^5 \ ^2P_{3/2} \rightarrow \text{Kr}^{2+} :4s^2 4p^4 \ ^3P_{2,1,0}$

$\beta_2 = 1.55$	$\beta_2 = 2.11$	$\beta_2 = 2.10$	$\beta_2 = 1.95$	$\beta_2 = 1.70$	$\beta_2 = 1.57$
$\beta_4 = 0.24$	$\beta_4 = 0.24$	$\beta_4 = 0.33$	$\beta_4 = 0.34$	$\beta_4 = 0.28$	$\beta_4 = 0.22$

The first step electrons come from the un-polarized atom

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} (1 + \beta_2 P_2(\cos\theta))$$

Second step electrons come from the **aligned** $^2P_{3/2}$ intermediate state

An atom/ion with total angular momentum J is **aligned** when the population of the m -substates is non-statistical, but states with projections $m, -m$ are **equally** populated

$$\frac{d\sigma}{d\Omega} = \frac{\sigma}{4\pi} (1 + \beta_2 P_2(\cos\theta) + \beta_4 P_4(\cos\theta))$$

proportional to the alignment of the ionic core created by the ionization in the first step

Conclusions / Future work

- ✓ Intensity dependence of the β -parameters for all the channels involved
- ✓ Theoretical calculations to support our experimental findings

Acknowledgments

- Prof. John Costello, Dr. Lampros Nikolopoulos, Dr. Bill Brocklesby and Dr. Thomas Kelly
- NCPST and School of Physical Sciences
- All the group members
- Work supported by the Education, Audiovisual and Culture Executive Agency (EACEA)
Erasmus Mundus Joint Doctorate Programme
Project No. 2011 – 0033.

Thank you for your attention!