

MULTIPLE IONIZATION/EXCITATION OF ATOMS AND IONS IN XUV SHORT FIELDS

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High Order Harmonic Generation

Free Electron Laser

X-ray Lasers



-XUV domain
-High intensities
-Short pulses



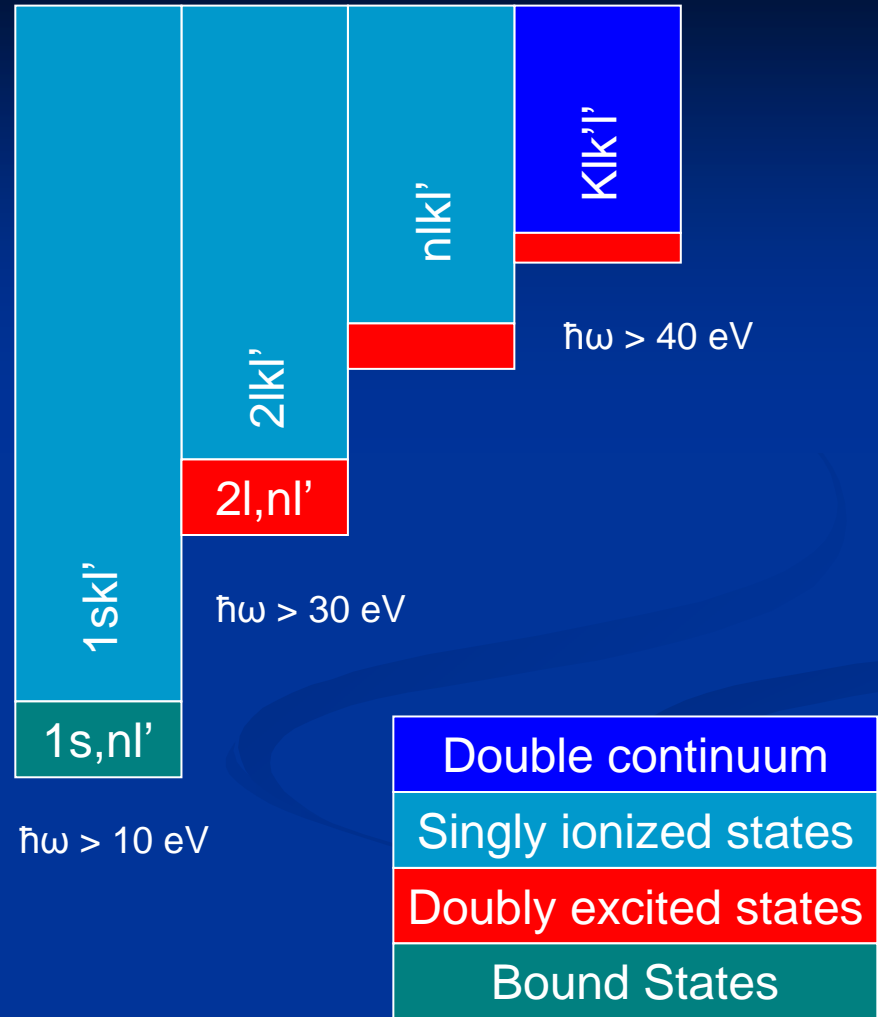
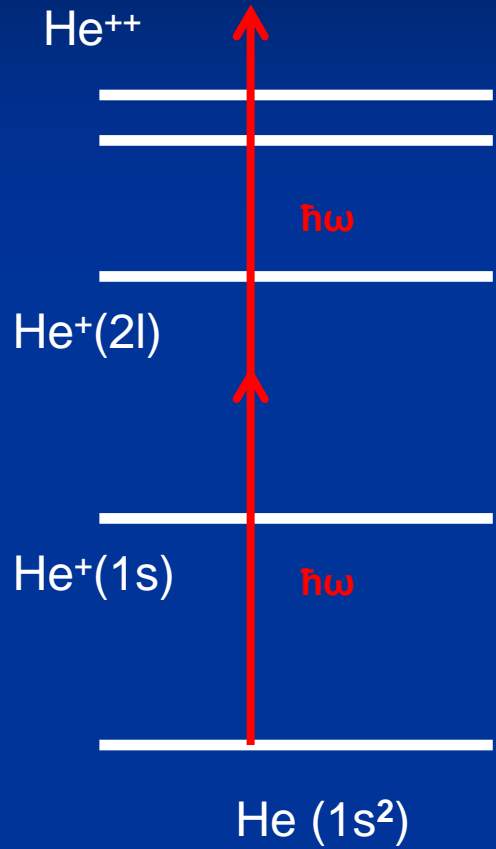
XUV laser-matter interaction in the non-linear domain

$I \sim 10^{12}-10^{13} \text{ W/cm}^2$ $\hbar\omega = 10 \text{ eV} - 80 \text{ eV}$

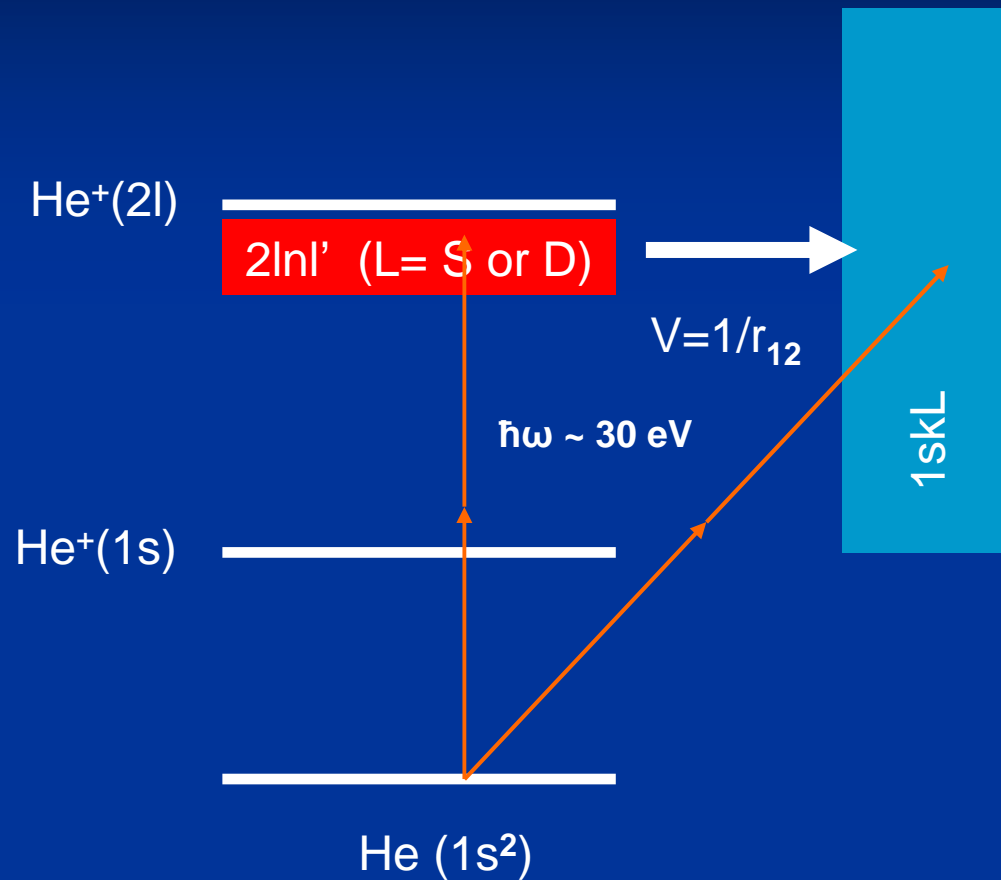
$K_{\text{Keldysh}} = (I_p / 2U_p)^{1/2}$ $K_{\text{Keldysh}} \gg 1$

multiphoton regime - absorption of two photons - helium

WAVELENGTH RANGE FOR TWO-PHOTON ABSORPTION IN HELIUM



TWO-PHOTON TWO-ELECTRON EXCITATION: He case



$T < \text{fs} ; T > \text{fs}$

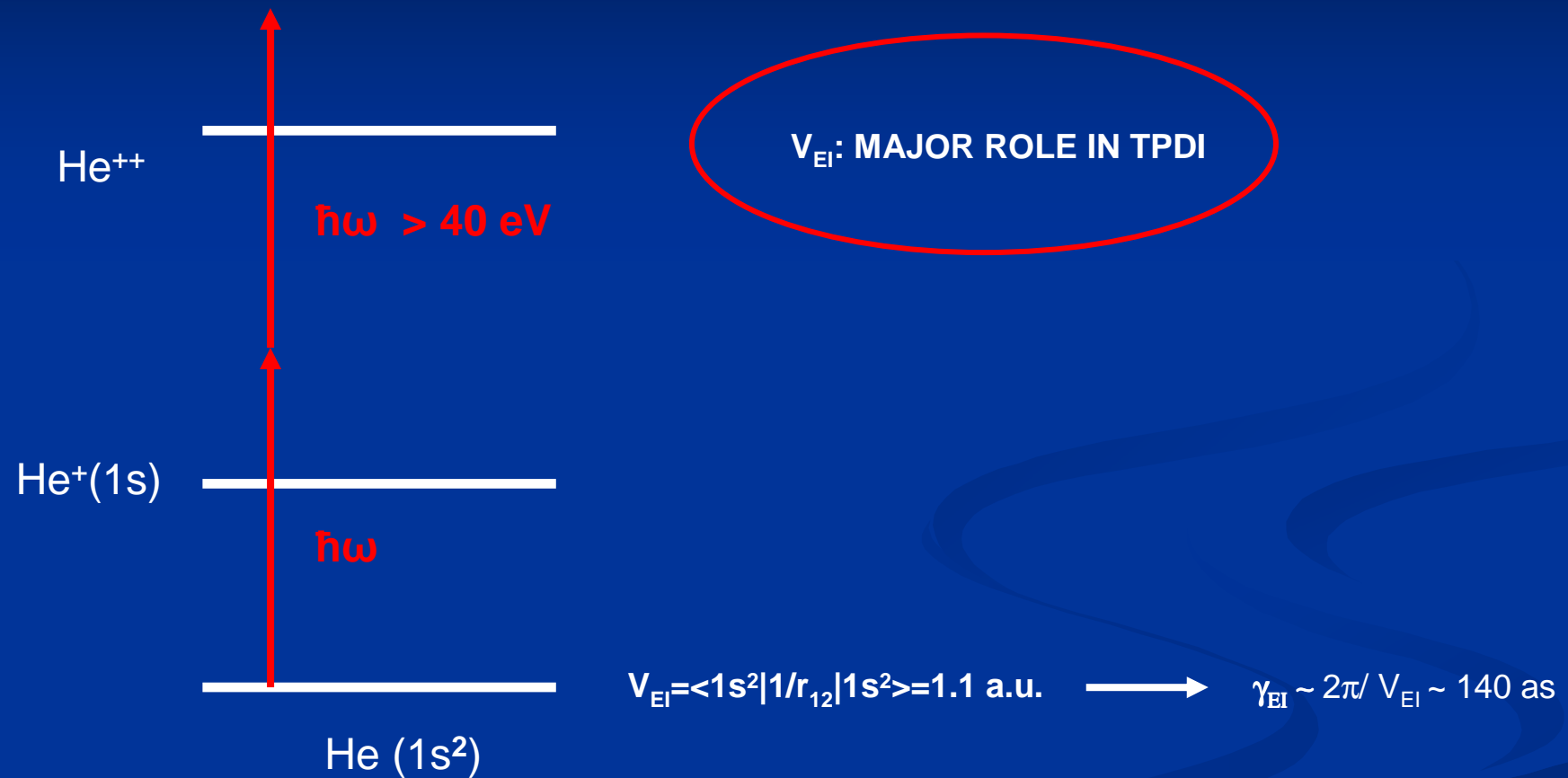
T : pulse duration

▪ $T \ll \gamma$

- deviation from Fano profile

Hasbani et al, J. Phys B **33** (2000)

TWO-PHOTON DOUBLE IONIZATION (TPDI): HELIUM CASE

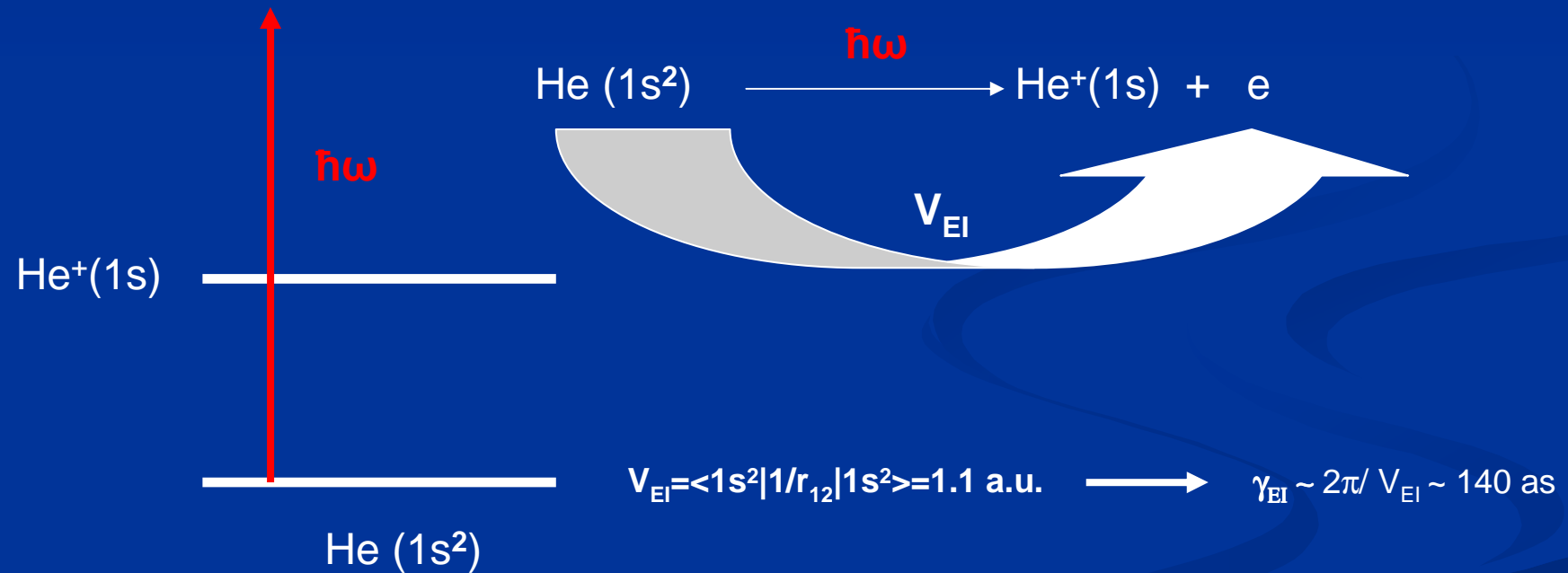


OUTLINE

- **TWO-PHOTON TWO-ELECTRON EXCITATION**
- **TWO-PHOTON DOUBLE IONIZATION (TPDI)**
 - **SEQUENTIAL REGIME**
 - **DIRECT REGIME**
- **CONCLUSIONS AND PERSPECTIVES**

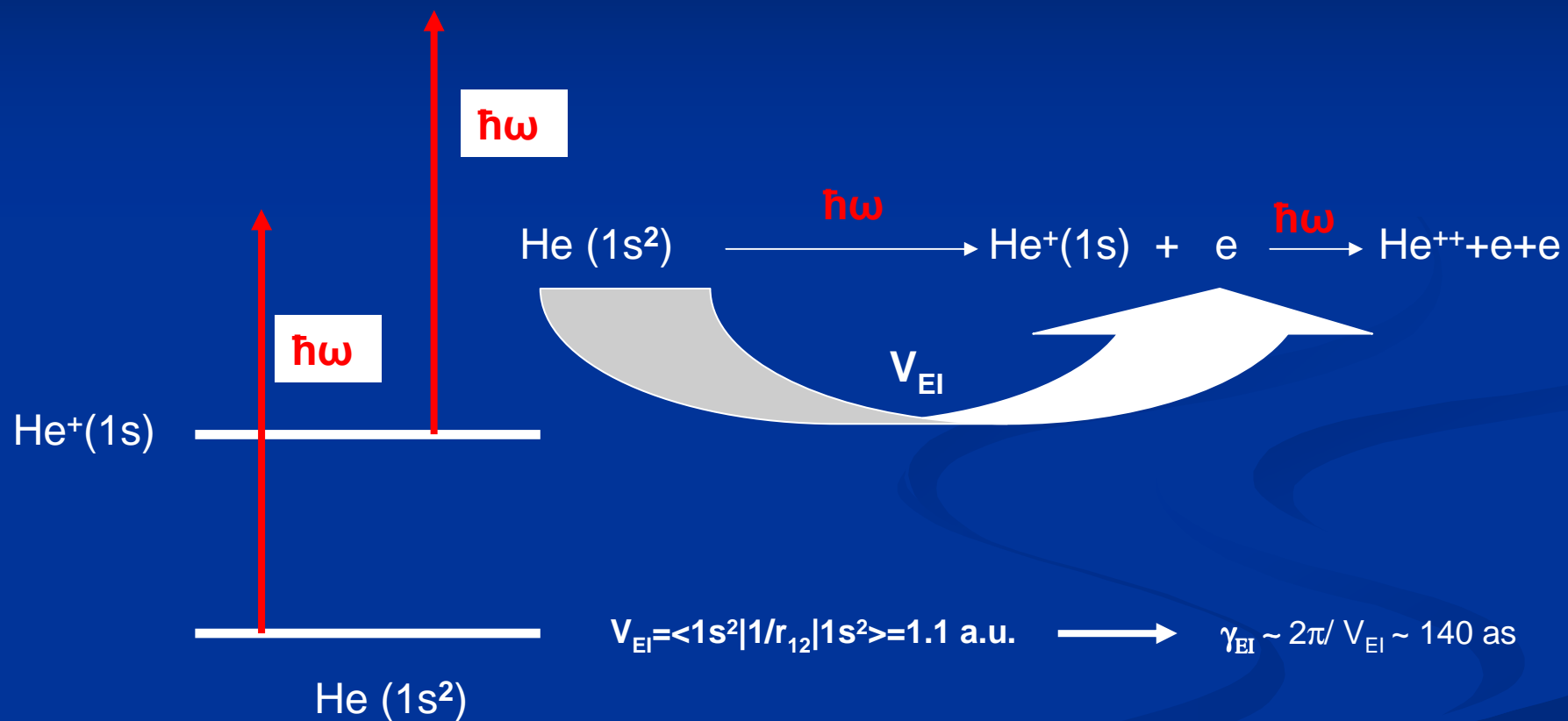
TWO-PHOTON DOUBLE IONIZATION (TPDI): HELIUM CASE

SINGLE IONIZATION



TWO-PHOTON DOUBLE IONIZATION: HELIUM CASE

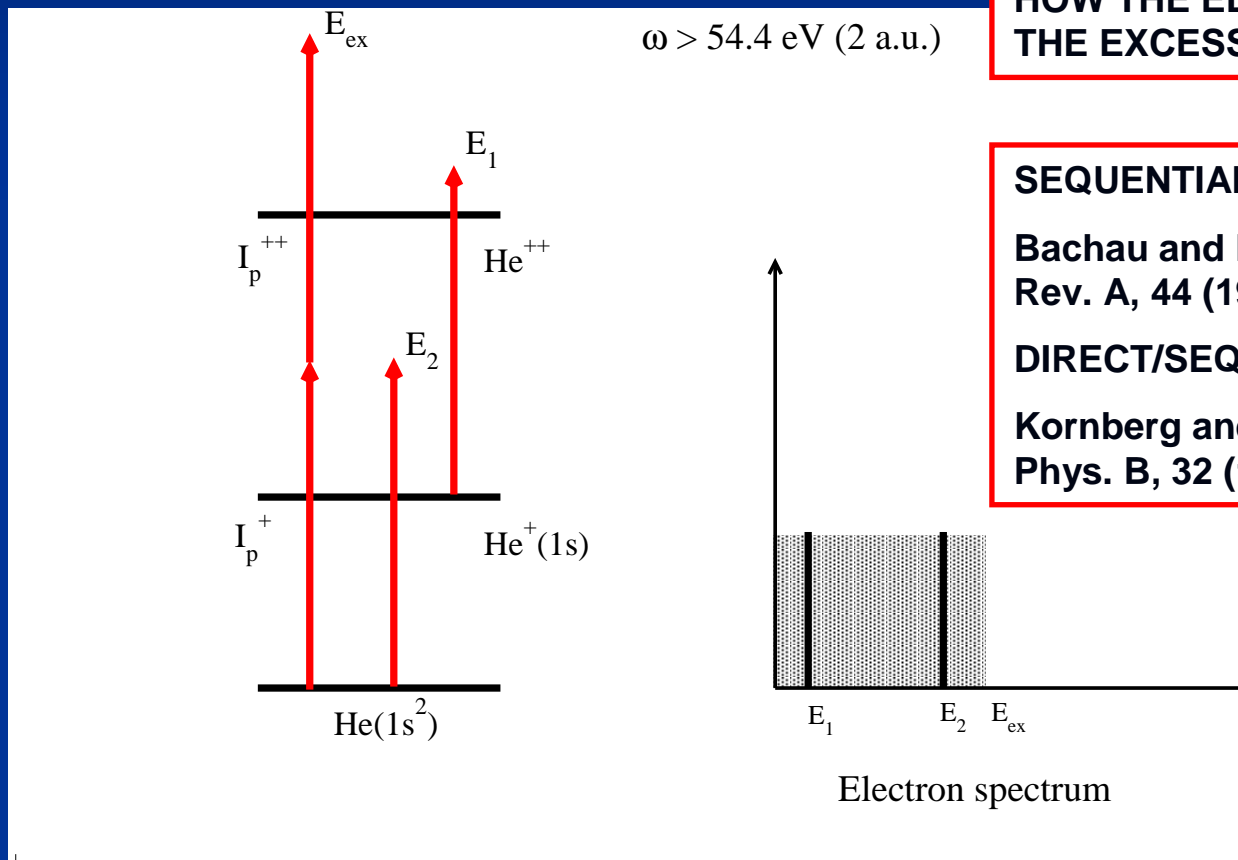
SEQUENTIAL REGIME ($\hbar\omega > 54.4$ eV or 2 a.u.)



TWO-PHOTON DOUBLE IONIZATION: HELIUM CASE

SEQUENTIAL REGIME ($\hbar\omega > 54.4$ eV)

- **DIRECT/SEQUENTIAL (TWO STEPS) REGIME**



**HOW THE ELECTRONS SHARE
THE EXCESS ENERGY ?**

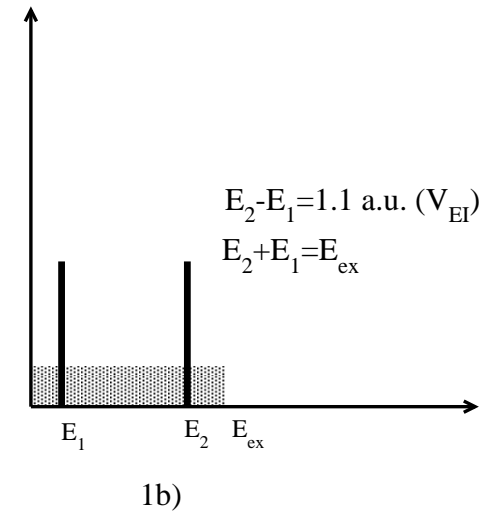
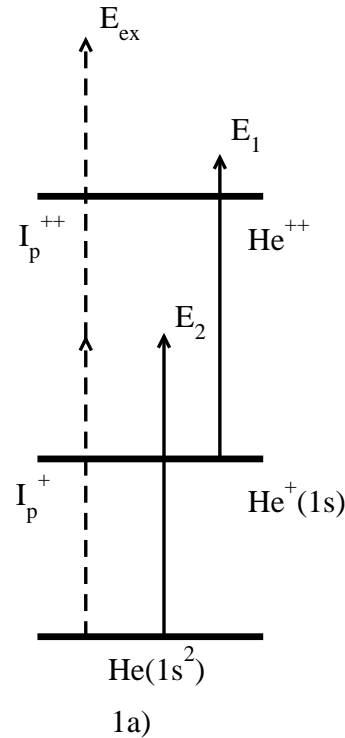
SEQUENTIAL TPDI:

**Bachau and Lambropoulos, Phys.
Rev. A, 44 (1991) R9**

DIRECT/SEQUENTIAL TPDI:

**Kornberg and Lambropoulos, J.
Phys. B, 32 (1999) L603**

- $T \gg \gamma_{EI} \sim 2\pi / V_{EI} \sim 140$ as
 - sequential DI dominates
 - $|E_1 - E_2| = V_{EI}$
 - rate $\sim T^2$ provided the intensity is not too large (10^{13} W/cm², P. Lambropoulos et al 2008)
 - simple formula (resolvent operator formalism-only including sequential double ionization channels) gives the peaks positions and widths

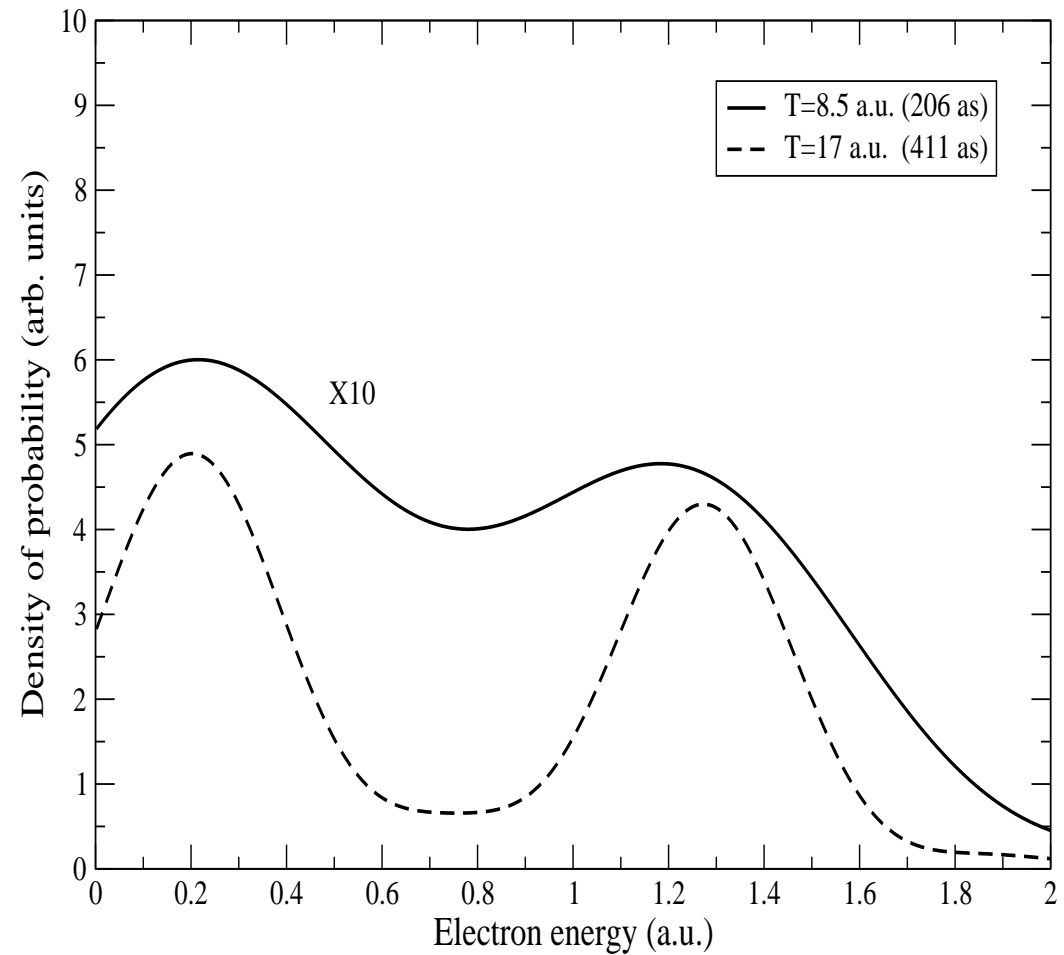
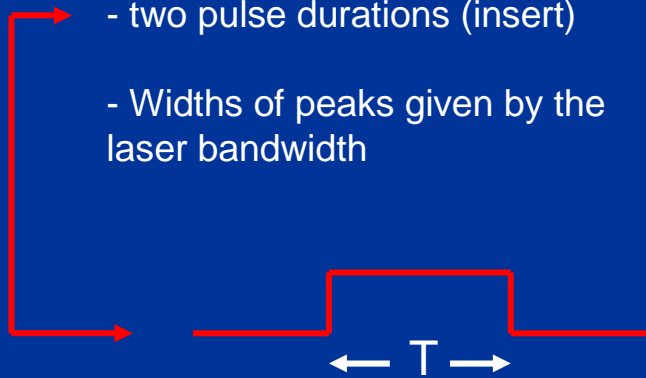


$$\frac{\gamma_{He^{++}} \gamma_{He^+}}{[(\epsilon_1 + \epsilon_2 - E_{ex})^2 + |\gamma_{He}|^2/4] [(\epsilon_1 - E_1)^2 + |\gamma_{He^+}|^2/4]}$$

Ionization rates of He and He⁺

- $T \rightarrow \gamma_{EI} \sim 2\pi / V_{EI} \sim 140$ as
- resolvent operator formalism-
only including sequential
ionization channels)
- $\hbar\omega = 60$ eV

- two pulse durations (insert)
- Widths of peaks given by the
laser bandwidth



▪ $T \longrightarrow \gamma_{EI} \sim 2\pi / V_{EI} \sim 140$ as

- We resolve the time-dependent Schrödinger equation (TDSE)

- Calculations involve direct and sequential channels

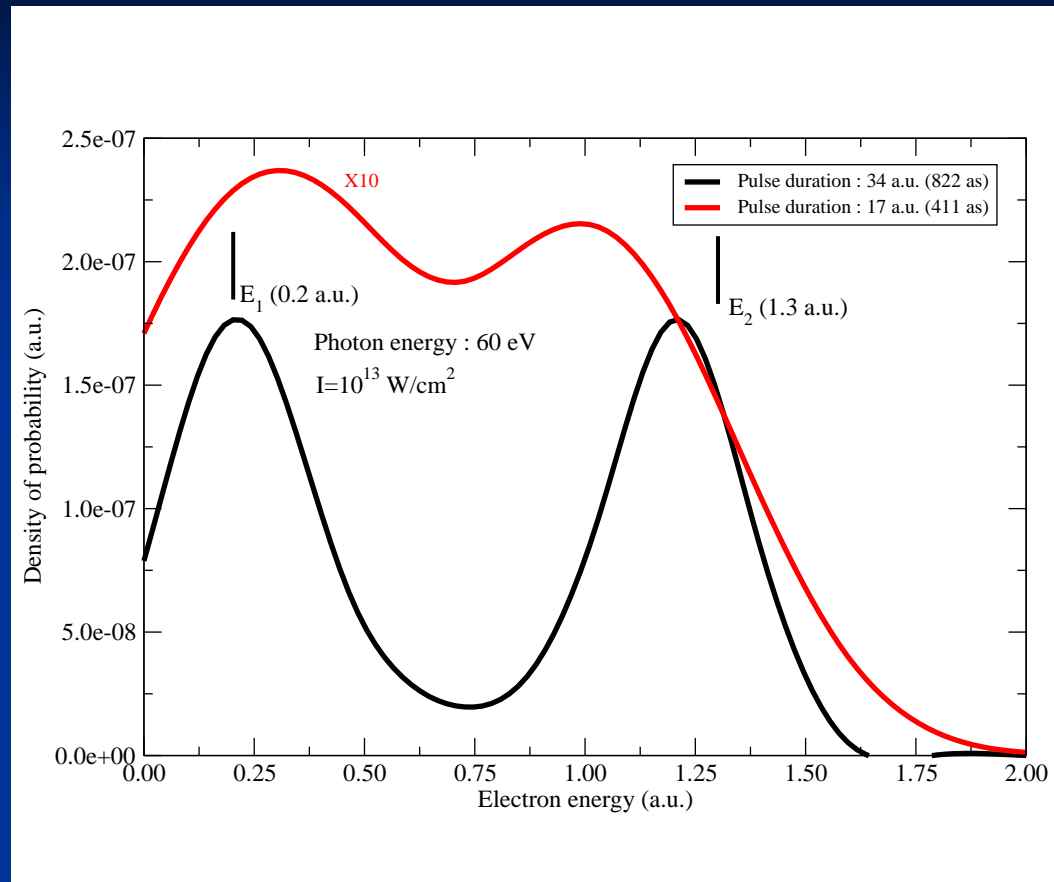
- The second photon “probes” the ionization of He

- The concept of direct and sequential processes are not meaningful for ultra short pulses

TPDI, sub-fs regime:

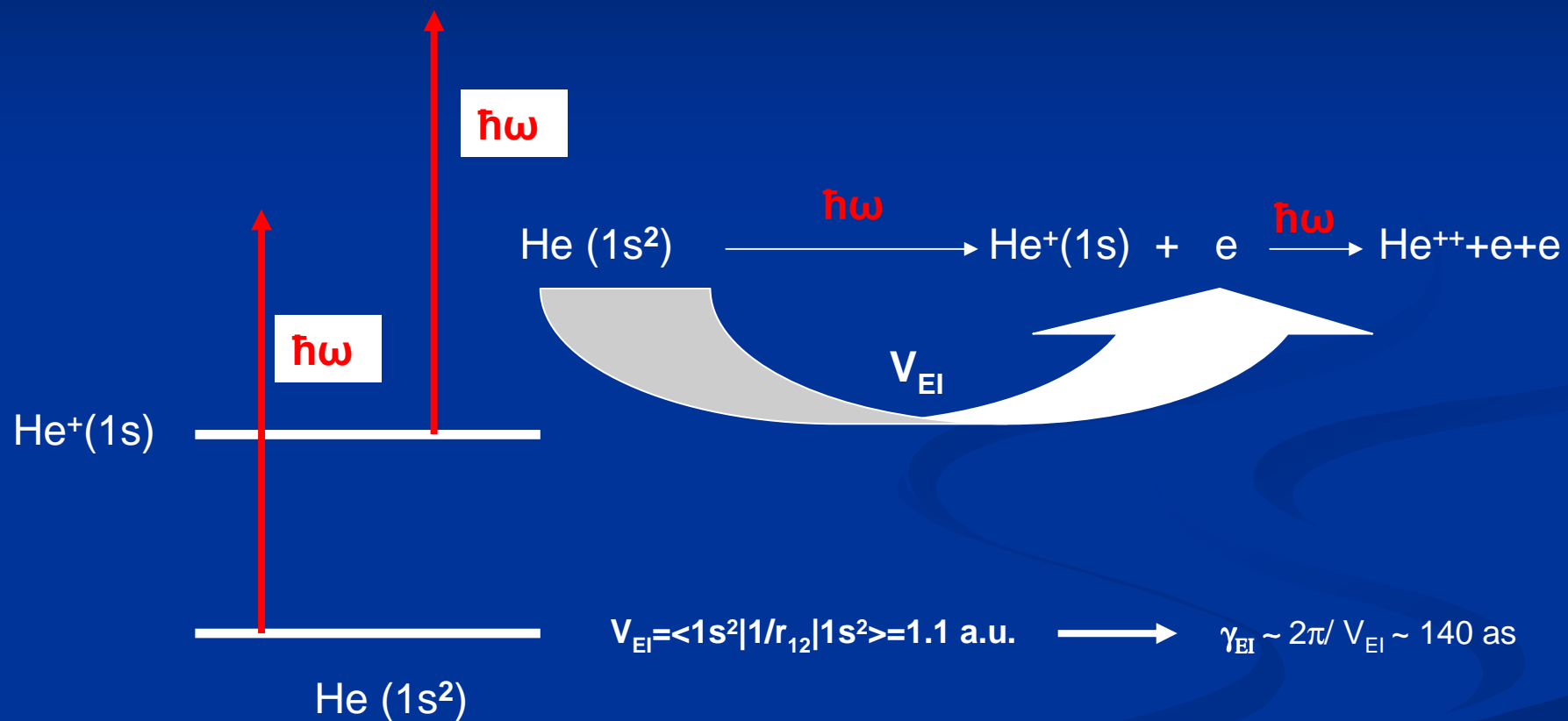
Foumouo et al J. Phys. B, 39 (2006) S427

Foumouo et al New J. Phys., 10 (2008) 025017



TWO-PHOTON DOUBLE IONIZATION: HELIUM CASE

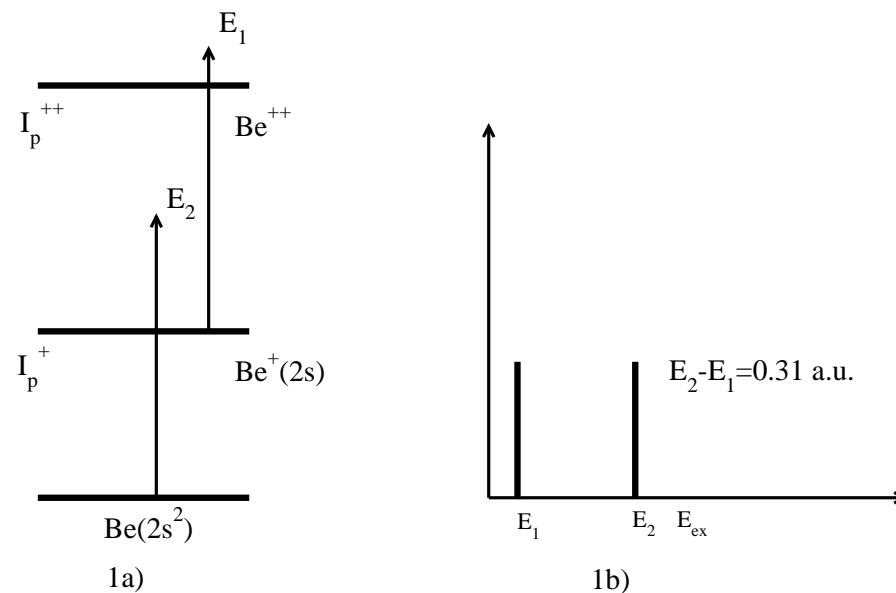
SEQUENTIAL REGIME ($\hbar\omega > 54.4$ eV)



• LIMIT OF 140 as UNREALISTIC ? THE CASE OF BERYLLIUM

- In Be $V_{EI} \sim 0.31$ a.u.
- $\gamma_{EI} \sim 2\pi / V_{EI} \sim 0.5$ fs
- $\hbar\omega > 18$ eV
- Effects expected for larger pulse durations

S. Laulan and H. Bachau,
Phys. Rev. A 69 (2004) 033408

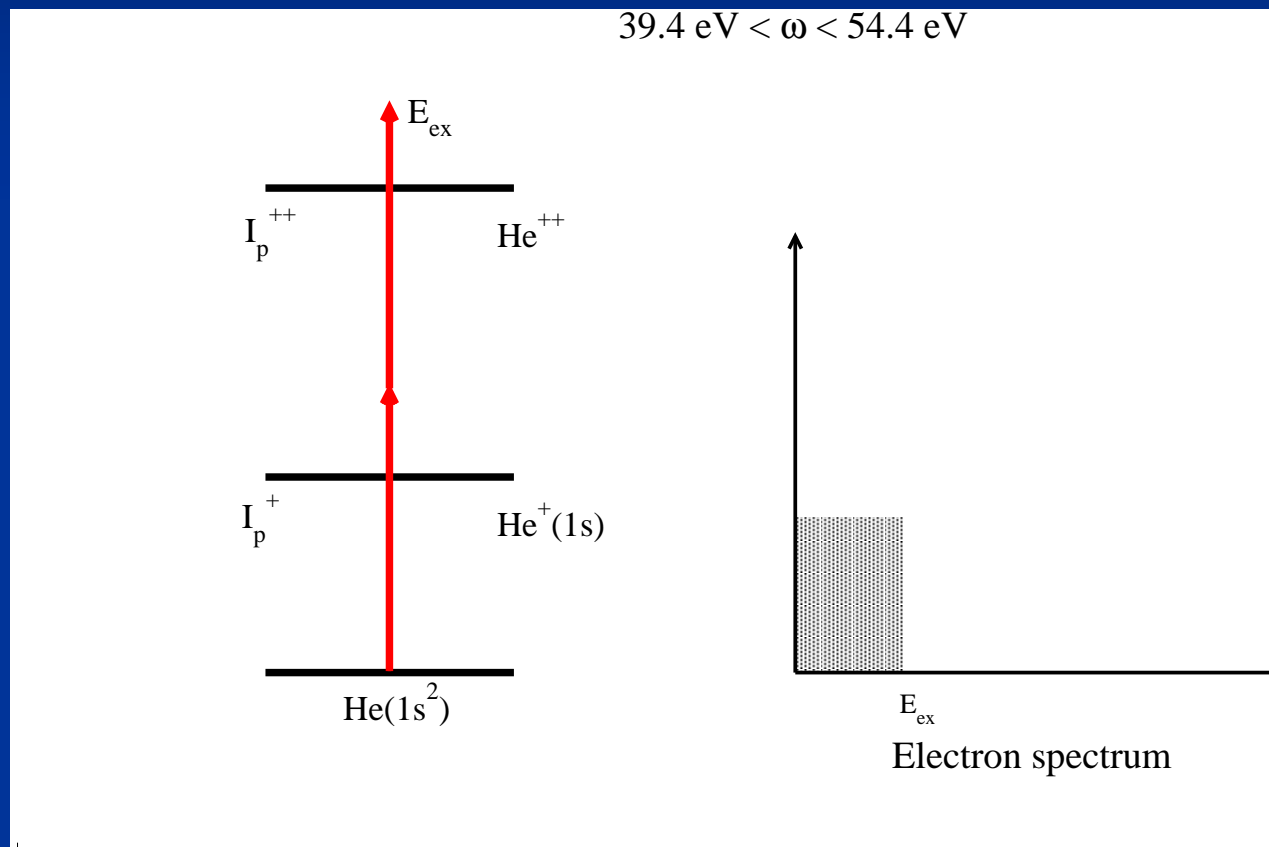


OUTLINE

- **TWO-PHOTON TWO-ELECTRON EXCITATION**
- **TWO-PHOTON DOUBLE IONIZATION (TPDI)**
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 - **DIRECT REGIME**
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TWO-PHOTON DOUBLE IONIZATION (TPDI): HELIUM CASE

DIRECT REGIME ($39.4 \text{ eV} < \hbar\omega < 54.4 \text{ eV}$)



• THEORETICAL ASPECTS

$$i \frac{\partial}{\partial t} \Psi(\mathbf{r}_1, \mathbf{r}_2, t) = [H + (\mathbf{p}_1 + \mathbf{p}_2) \cdot \mathbf{A}(t)] \Psi(\mathbf{r}_1, \mathbf{r}_2, t)$$

H : Hamiltonian for helium

Field potential (from $-T/2$ to $T/2$):

$$\mathbf{A}(t) = \mathbf{e}_z A_0 \left(\cos \frac{\pi t}{T} \right)^2 \cos \omega t$$

\mathbf{e}_z : polarization vector (linear pol.)

• SOLVE THE TDSE

- P. Lambropoulos et al, Phys. Rep., 305 (1998)
- H. Bachau et al, Rep. Prog. Phys., 64 (2001)

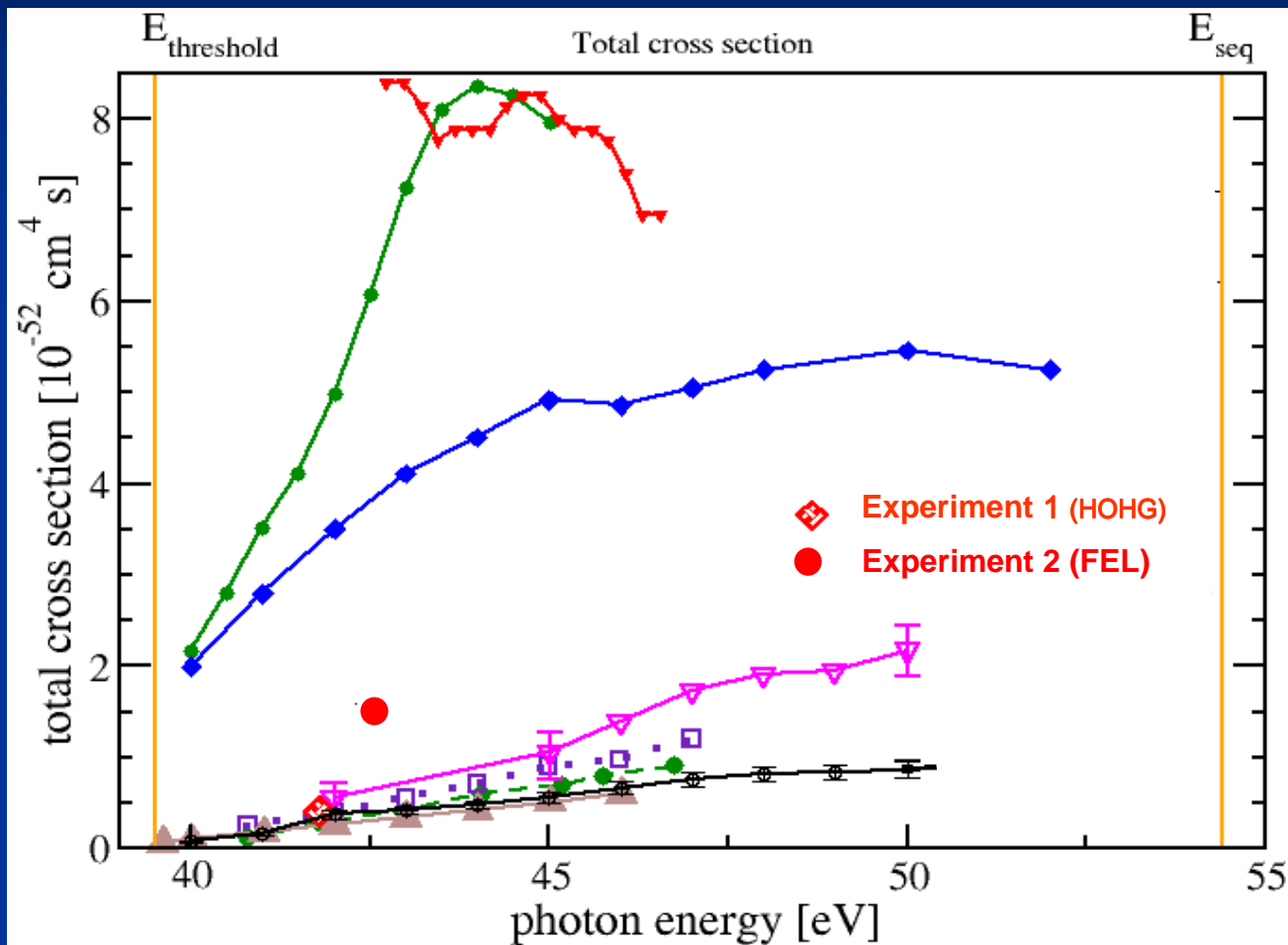
• DIFFICULTY: CALCULATING THE DOUBLE CONTINUUM (ROLE OF ELECTRON CORRELATIONS)

• INFORMATIONS:

- PROVIDED THE INTENSITY IS NOT TOO LARGE THE TPDI CROSS-SECTION EXIST (TPDI~T) AND CAN BE EXTRACTED FROM TDSE (OR DIRECTLY CALCULATED USING LOPT)
- ELECTRON ENERGY DISTRIBUTIONS
- ELECTRON ANGULAR DISTRIBUTIONS
- RECOIL ION MOMENTUM DIST.

E. Fomouo, Ph. Antoine, B. Piraux,
L. Malegat, H. Bachau and R.
Shakeshaft, J. Phys. B, 41 (2008)
051001 (Fast Track Communication)

● **TPDI CROSS-SECTION**



By decreasing order of magnitude

L. A. A. Nikolopoulos and P. Lambropoulos, *J. Phys. B* **40**, 1347 (2007)

L. A. A. Nikolopoulos and P. Lambropoulos, *J. Phys. B* **34**, 545 (2001)

E. Fomouo, G. L. Kamta, G. Edah, and B. Piraux, *Phys. Rev. A* **74**, 063409 (2006)

I. A. Ivanov and A. S. Kheifets, *Phys. Rev. A* **75**, 033411 (2007)

B. Piraux, J. Bauer, S. Laulan, and H. Bachau, *Eur. Phys. J. D* **26**, 7 (2003)

L. Feng and H. W. van der Hart, *J. Phys. B* **36**, L1 (2003)

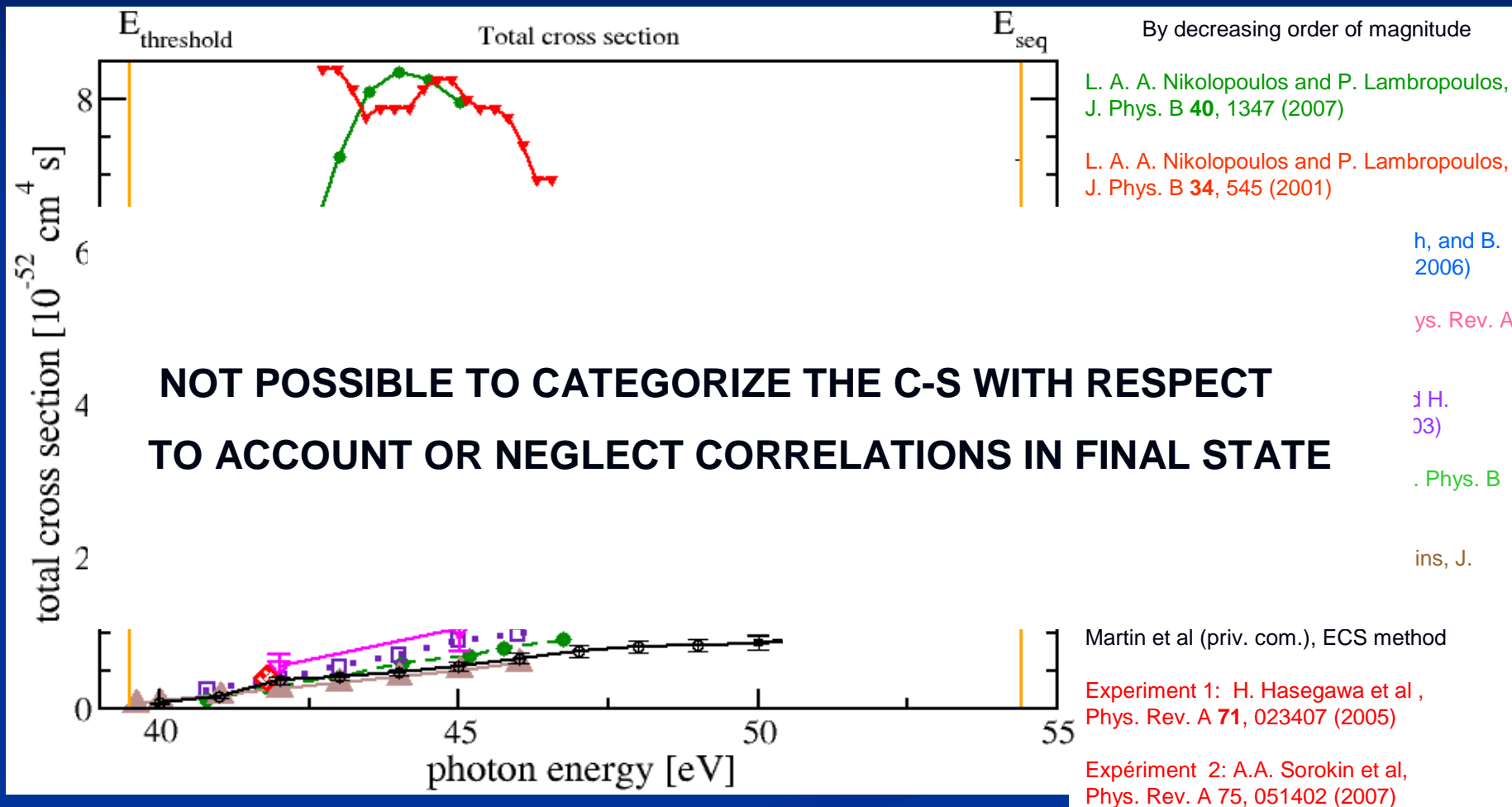
S. X. Hu, J. Colgan, and L. A. Collins, *J. Phys. B* **38**, L35 (2005)

Horner et al, *Phys. Rev. A* **76** (2007) 030701

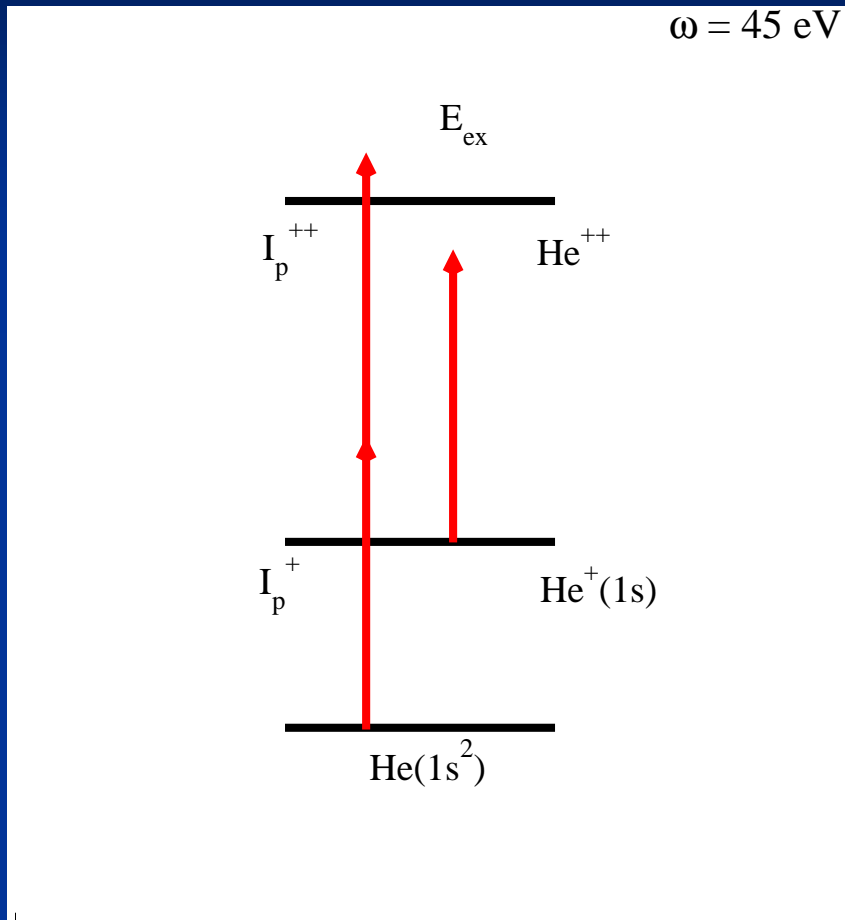
Experiment 1: H. Hasegawa et al , *Phys. Rev. A* **71**, 023407 (2005)

Expériment 2: A.A. Sorokin et al, *Phys. Rev. A* **75**, 051402 (2007)

● **TPDI CROSS-SECTION**

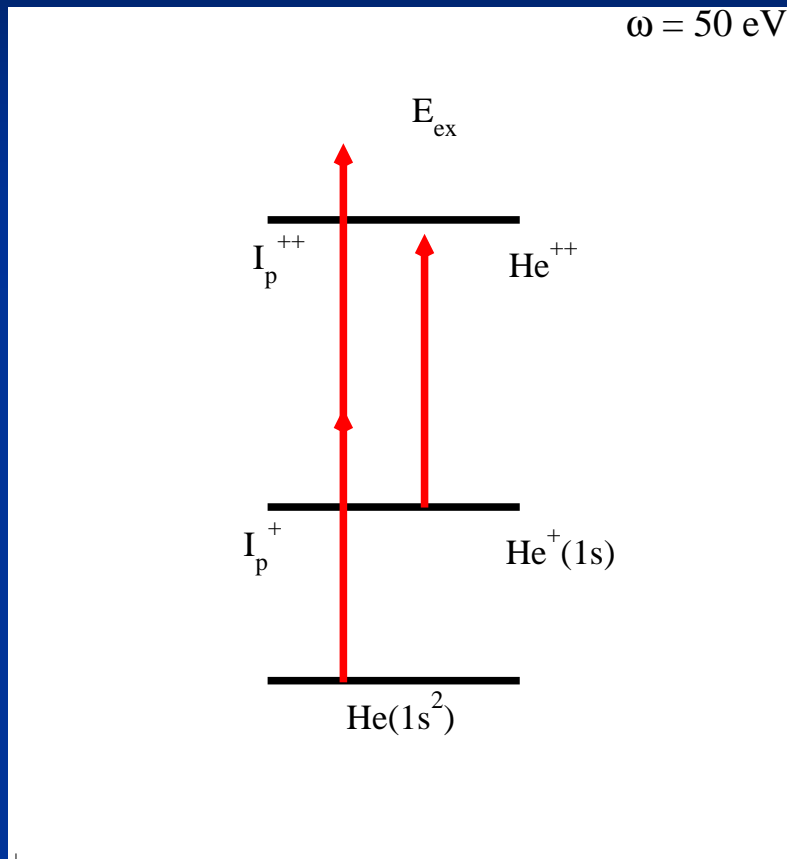


• TPDI ($\hbar\omega = 45$ eV)



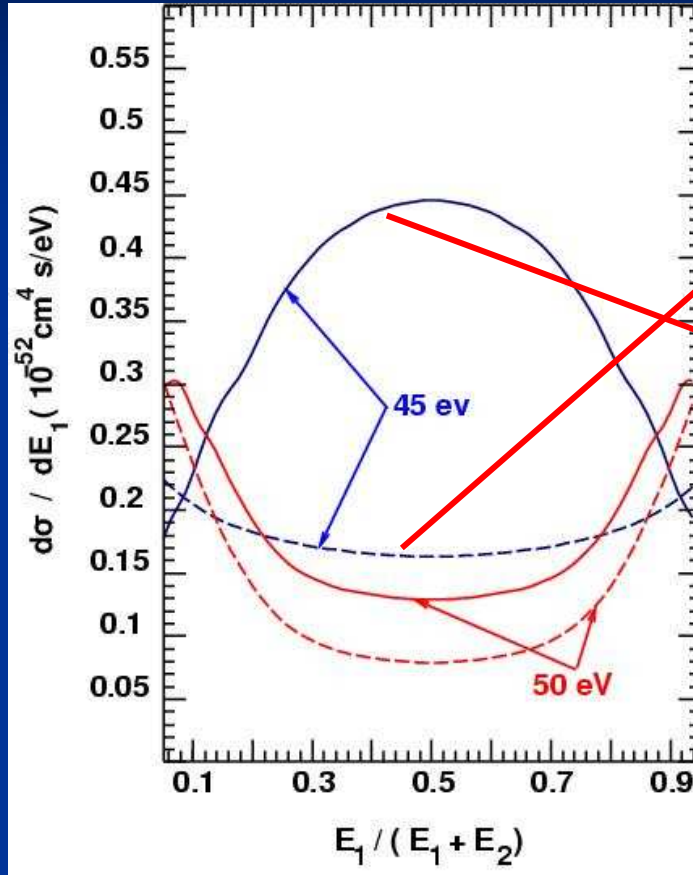
Excess energy : 0.4 a.u. (11 eV)

- TPDI ($\hbar\omega = 50$ eV), close to the threshold for sequential TPDI



Excess energy : 0.77 a.u. (21 eV)

● ENERGY DISTRIBUTION (HOW THE ELECTRONS SHARE E_{ex})



Integrated energy distribution (E_k) – Method 1:

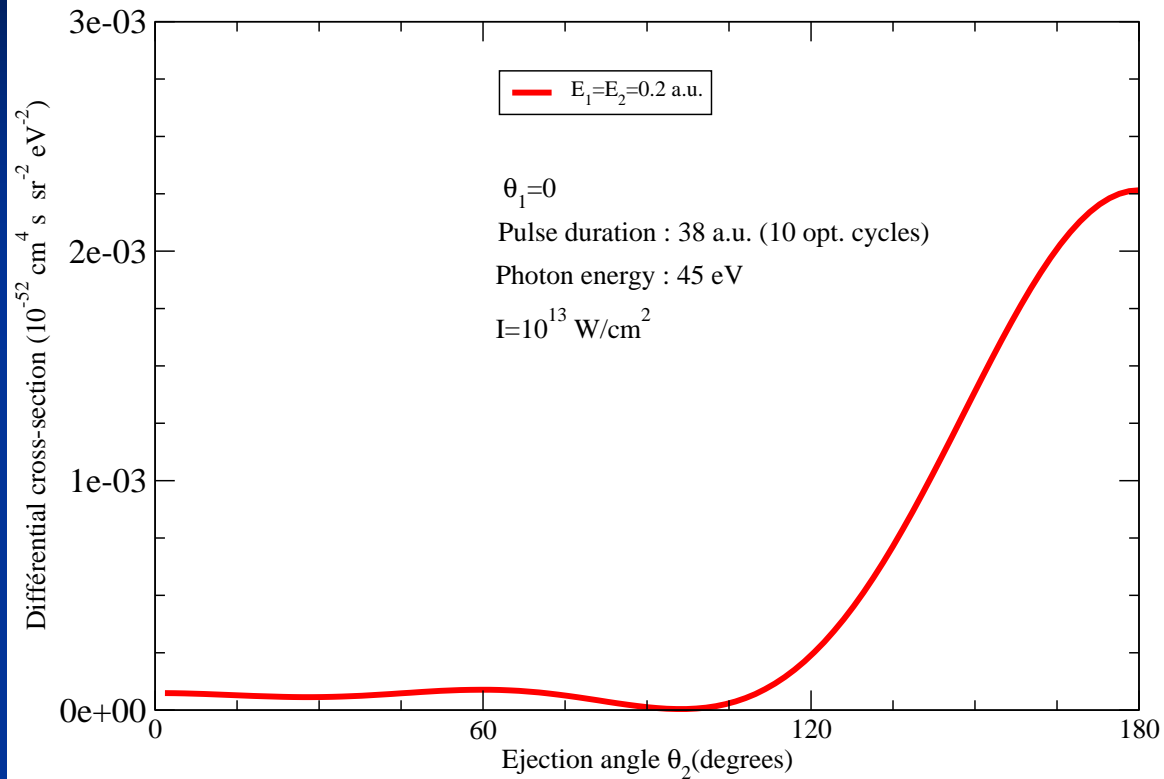
$$\frac{d P_{l_1, l_2}^{L, M}}{d E_k} = \int \left| \left\langle \psi_{E_k}^{l_1}(\mathbf{r}_1) \psi_{E_k}^{l_2}(\mathbf{r}_2) + 1 \right| \Psi(t = \frac{T}{2}) \right|^2 d E_k,$$

Integrated energy distribution (E_k) – Method 2 (J – matrix, Fomouo et al):

$$\frac{d P_{l_1, l_2}^{L, M}}{d E_k} = \int \left| \left\langle \psi_{E_k}^{l_1}(\mathbf{r}_1) \psi_{E_k}^{l_2}(\mathbf{r}_2) + 1 \right| \Psi(t = \frac{T}{2}) - \Psi_{SI} \right|^2 d E_k,$$

D.A. Horner et al, Phys. Rev. A 76 (2007),
ECS with electron correlations included in
final state : **U-shaped energy distribution**

• **TPDI ANGULAR DISTRIBUTION ($\hbar\omega = 45$ eV)**



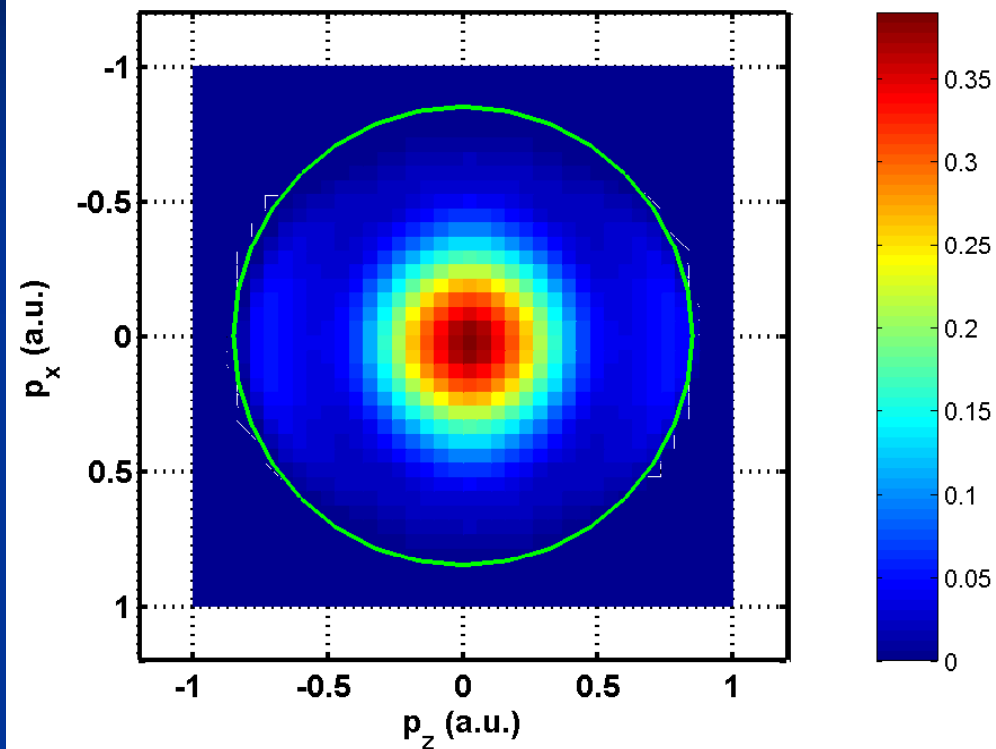
$\theta_1=0$

Excess energy : 0.4 a.u. (11 eV)

$E_1=0.2$ a.u.

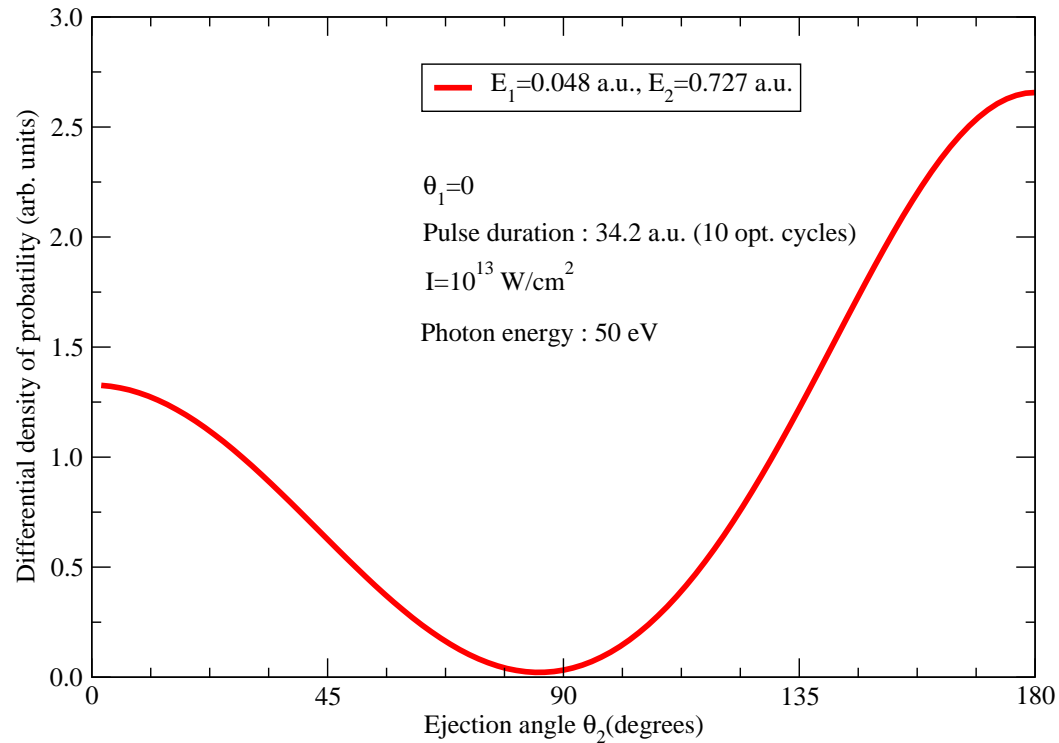
$E_2=0.2$ a.u.

● RECOIL ION MOMENTUM DISTRIBUTION ($\hbar\omega = 42$ eV)



E. Fomouo et al, J. Phys. B, 41 (2008) 051001 (Fast Track Communication)

• **TPDI ANGULAR DISTRIBUTION ($\hbar\omega = 50$ eV)**



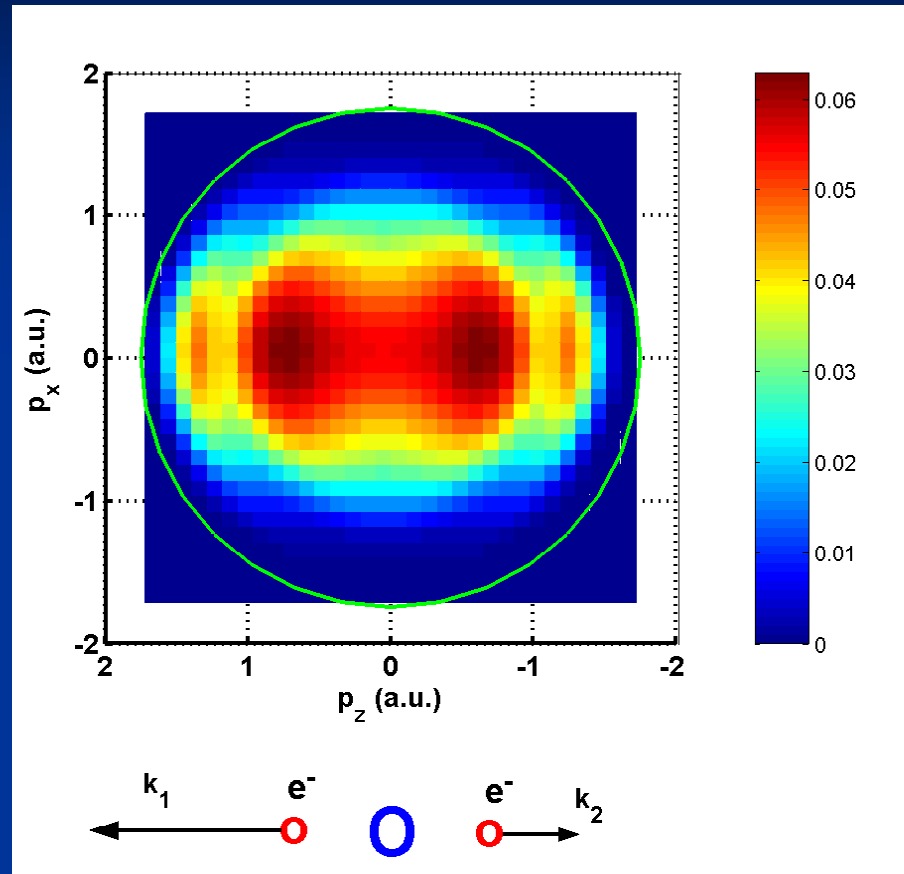
← ● $\theta_1=0$

Excess energy : 0.77 a.u. (21 eV)

$E_1=0.048$ a.u.

$E_2=0.727$ a.u.

● RECOIL ION MOMENTUM DISTRIBUTION ($\hbar\omega = 50$ eV)



● CONCLUSIONS AND PERSPECTIVES

- DEVELOPMENT OF SHORT AND INTENSE XUV LASER PULSES + SOPHISTICATED DETECTION TECHNIQUES OPENS NEW “AVENUES” IN A&M PHYSICS
- SPECTROSCOPY OF IONS AND ATOMS
- DYNAMICS OF FUNDAMENTAL PROCESSES LIKE DOUBLE IONIZATION
- Experiment are needed -recoil-ion momentum of crucial interest - already applied in Ne (FLASH) (R. Moshhammer et al, PRL 98 (2007) 203001)
- HIGHER ORDER PROCESSES
- APPLICATIONS ON MOLECULES (VIB. MOTION ~FS)

• THANK YOU

Workshop - 4th generation - 23-24
October 2008 - Paris