

One- and two-electron systems in strong IR- and XUV-fields

Anatoli Kheifets and Igor Ivanov



Igor Bray



Centre for Antimatter-Matter Studies

KITP 2010

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Numerically accurate atomic physics
in strong field regime

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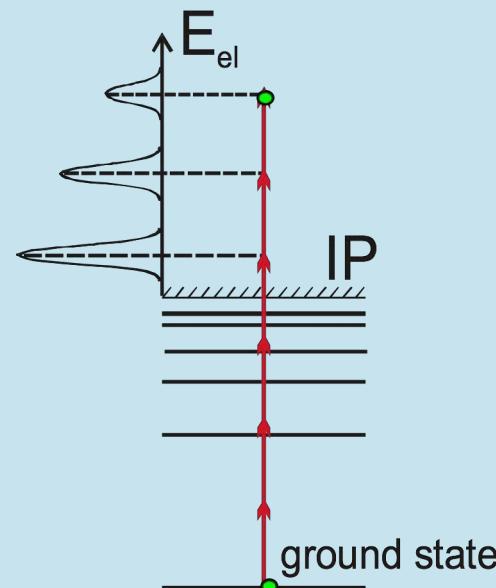


Regimes of strong field ionization

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Multiphoton-ionization

- low intensity,
- E-field of laser weak compared to atomic field
- perturbative regime



$$\gamma \gg 1$$

Regimes of strong field ionization

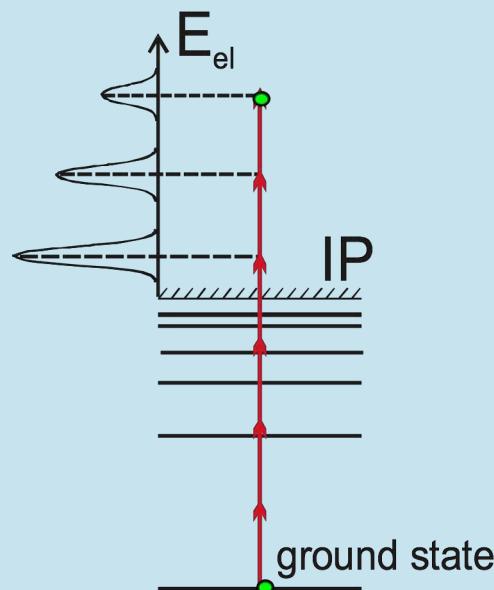
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Tunneling-ionization

- high intensity
- classical field picture
- field ionization in quasistatic laser field



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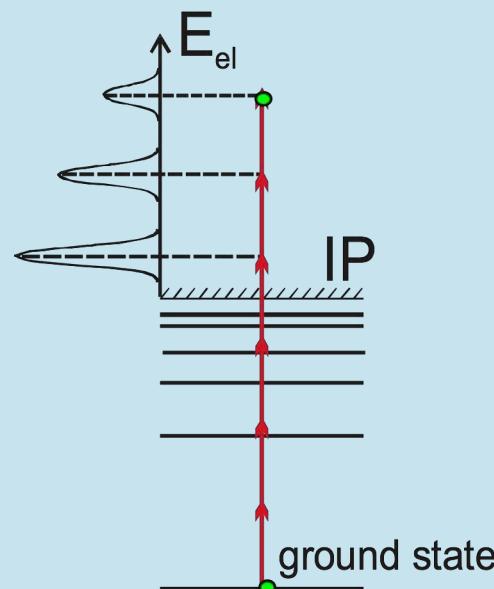
$$w(t) \propto \omega^2 \exp\left(-\frac{2(2\text{IP})^{3/2}}{3F(t)}\right)$$

$$\gamma = \sqrt{\frac{4\text{IP}}{I} \omega^2}$$

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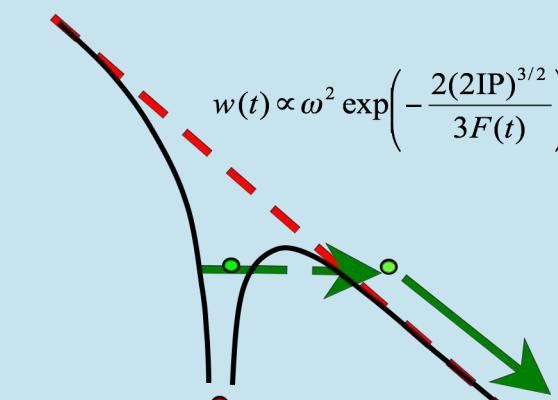
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$$U_p = I / 4\omega^2$$

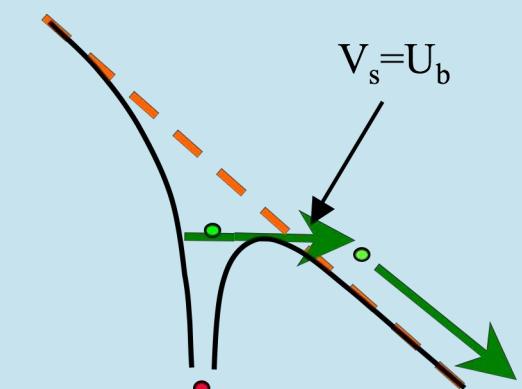
Tunneling-ionization

- high intensity
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Over-the-barrier-ionization

- highest intensities
- complete suppression of the Coulomb-barrier of bound state
- Atomic system dominated by external field



$$\gamma < 1$$

Case studies:

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- **Time-resolved atomic reactions**
 - *Electron correlation resolved in time*
 - *Double ionization of He*
 - *Single ionization of noble gases*

Outline

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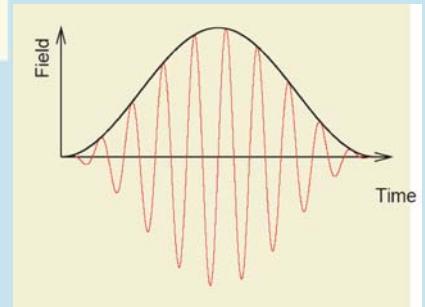
Outline

- Theoretical model
 - Time-dependent Schrödinger equation
 - *Basis based integration vs coordinate space integration*
 - Field-free atomic states
 - *One-electron HF states*
 - *Two-electron CCC states*
 - Discretization of continuum
 - *Pseudostates vs continuum states*

Theoretical Model

Theoretical Model

Field on:

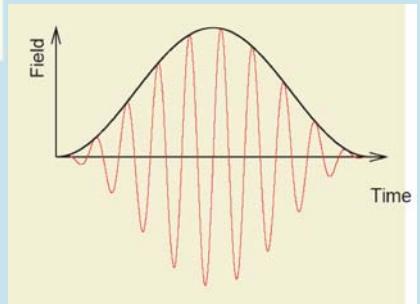


Theoretical Model

Field on:

Time-Dependent Schrödiger equation $i\frac{\partial\Psi}{\partial t} = \hat{H}\Psi$

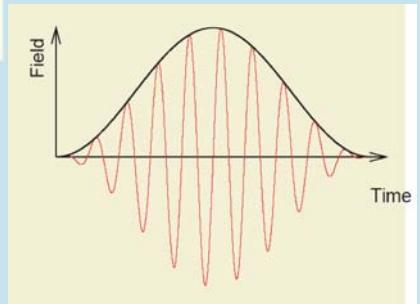
$$\hat{H} = \frac{\mathbf{p}_1^2}{2} + \frac{\mathbf{p}_2^2}{2} - \frac{2}{r_1} - \frac{2}{r_2} + \frac{1}{|\mathbf{r}_1 - \mathbf{r}_2|} + \mathbf{D} \cdot \mathbf{F}_{\text{AC}} \cos \omega t$$



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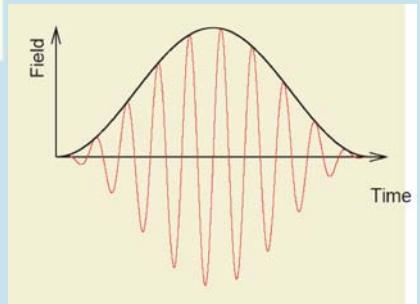
$$\Psi(\mathbf{r}_1, \mathbf{r}_2, t) = \sum_{j \in \left\{ \begin{smallmatrix} n_1 n_2 N \\ l_1 l_2 L \end{smallmatrix} \right\}} a_j(t) \phi_{n_1 l_1}^N(r_1) \phi_{n_2 l_2}^N(r_2) |l_1(1) l_2(2) L\rangle$$

Pseudostate basis: $\langle \phi_i^N | \hat{H} | \phi_j^N \rangle = E_i \delta_{ij}$

Theoretical Model

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Matrix notations:

$$i\mathbf{R} \cdot \dot{\mathbf{a}}(t) = \mathbf{H} \cdot \mathbf{a}(t), \quad \text{with initial condition } \Psi(\mathbf{r}_1, \mathbf{r}_2, t=0) = \Psi_0(\mathbf{r}_1, \mathbf{r}_2)$$

Theoretical Model

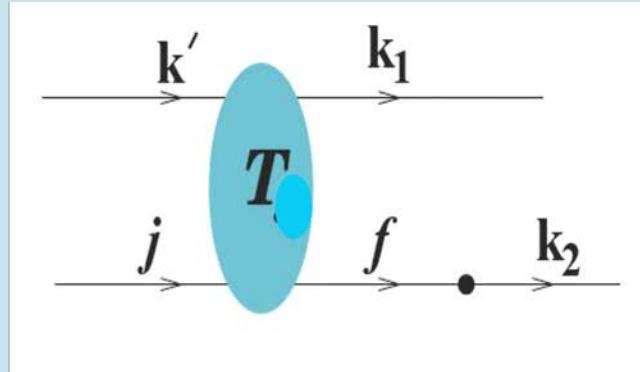
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Field off:

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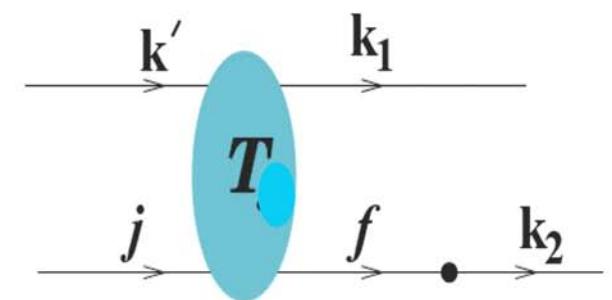
CCC expansion for two-electron continuum



Theoretical Model

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CCC expansion for two-electron continuum



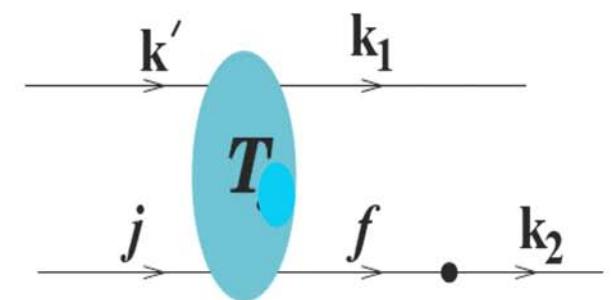
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Theoretical Model

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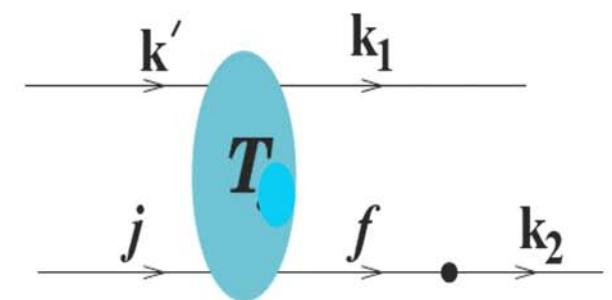
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Coulomb wave

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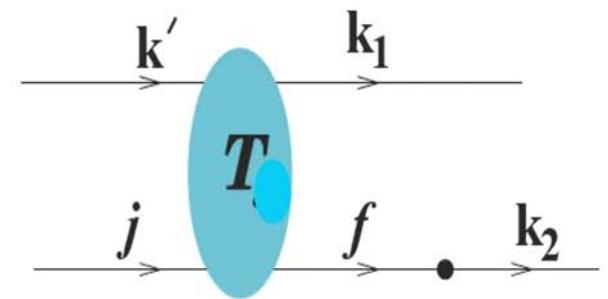
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Pseudostate

Theoretical Model

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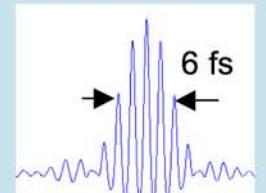
Pseudostate

Fully differential cross-section

$$\frac{d\sigma}{d\mathbf{k}_1 d\mathbf{k}_2} = |\langle \Psi(\mathbf{r}_1, \mathbf{r}_2, t = T) | \Psi_{\mathbf{k}_1 \mathbf{k}_2}(\mathbf{r}_1, \mathbf{r}_2) \rangle|^2 \quad T = N \frac{2\pi}{\omega} , \quad N \gg 1$$

Strong IR Field Ionization of Hydrogen

IR: 750 nm, $\omega = 1.5 \text{ eV}$, $T=2.5 \text{ fs}$, $FWHM=5.5 \text{ fs}$, $I = 10^{14-15} \text{ W/cm}^2$

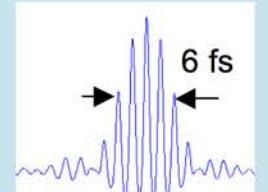


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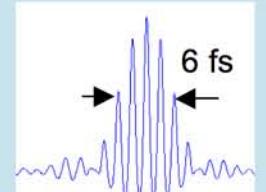
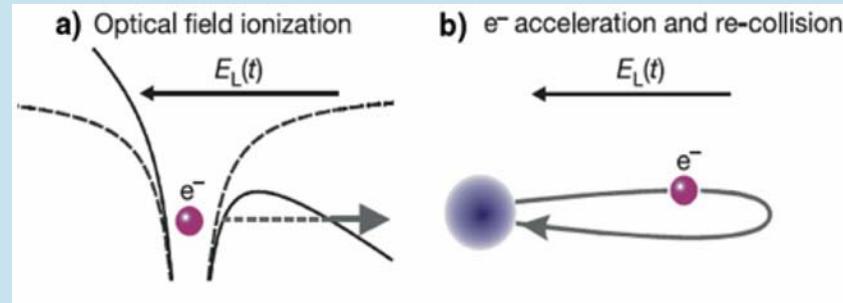
$$\gamma \approx 1, U_p = 0.25 \text{ au}, \xi = U_p / \omega \approx 5$$
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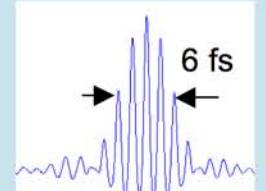
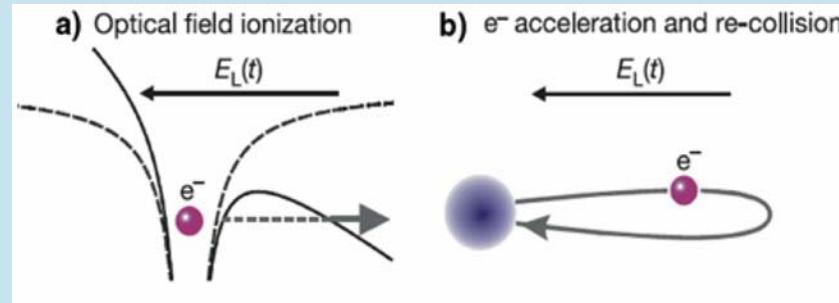


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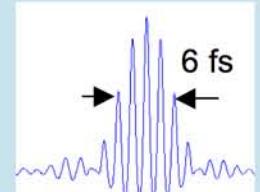
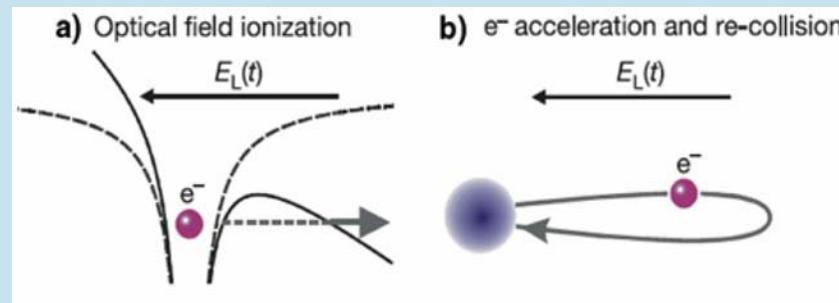
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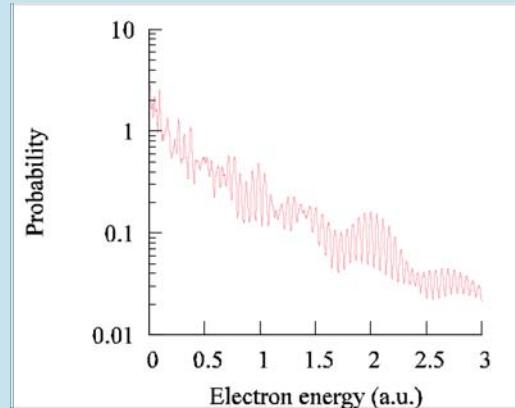
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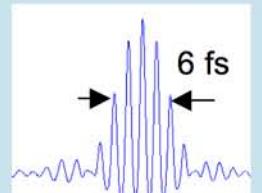
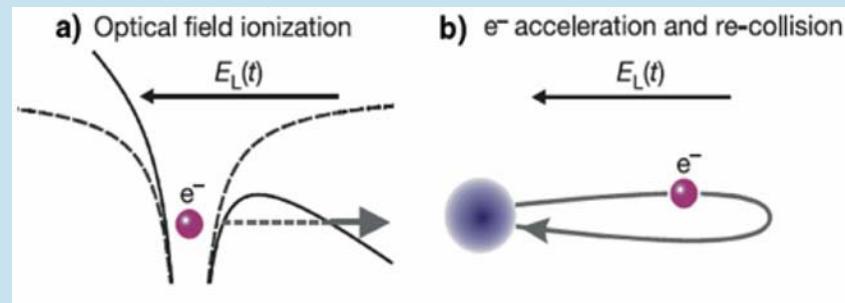
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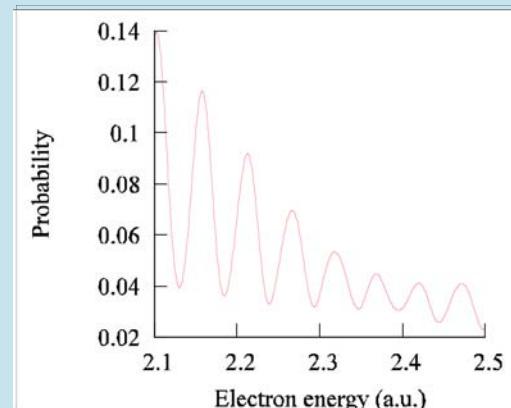
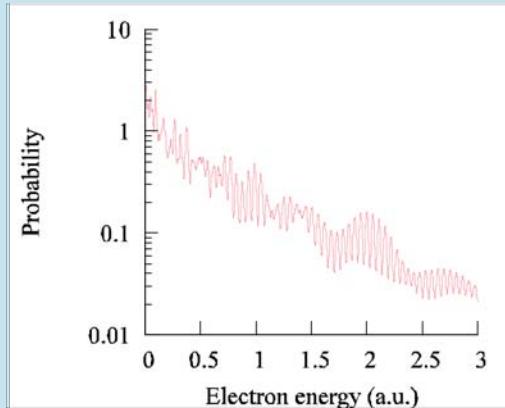
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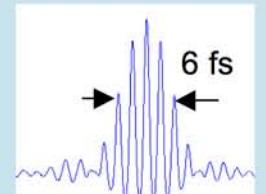
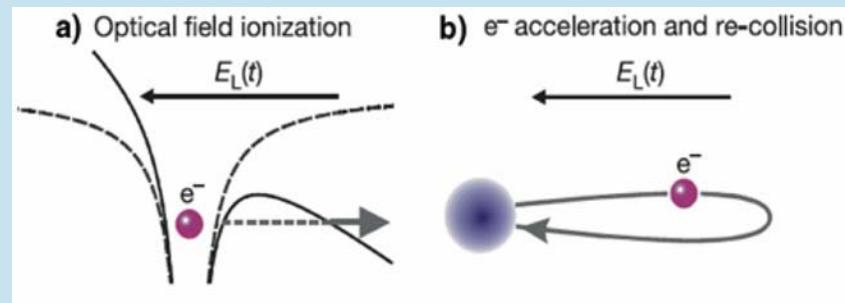
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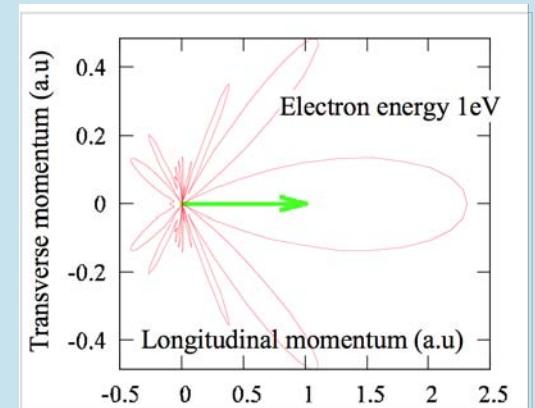
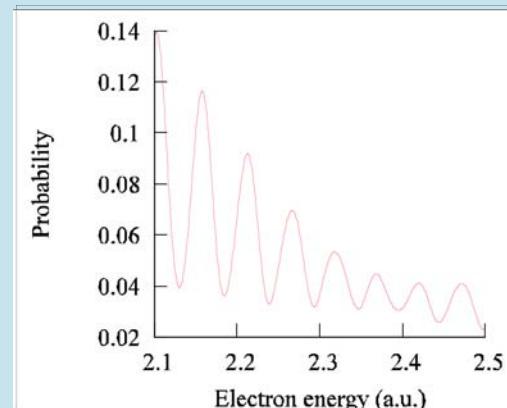
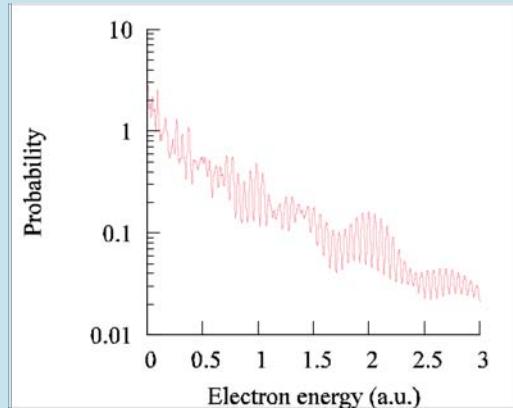
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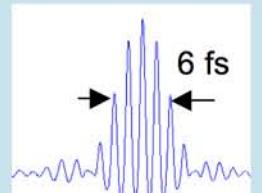
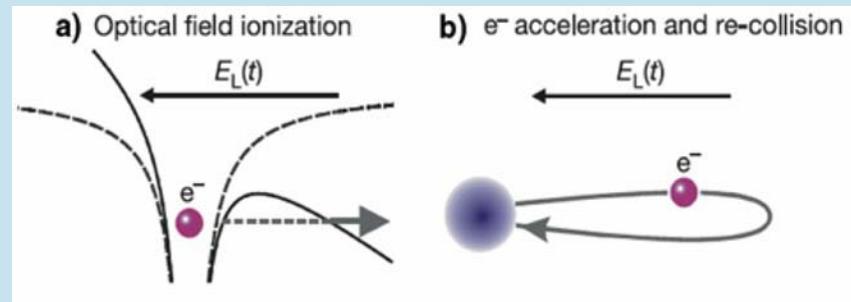
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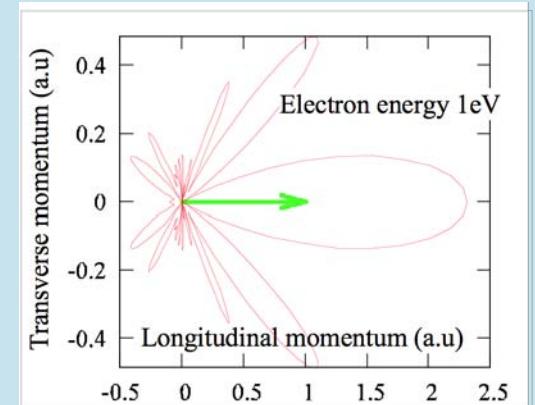
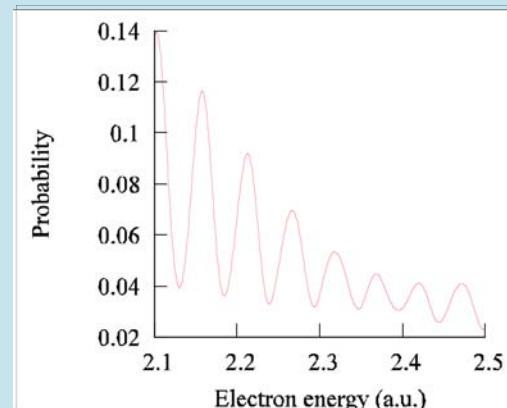
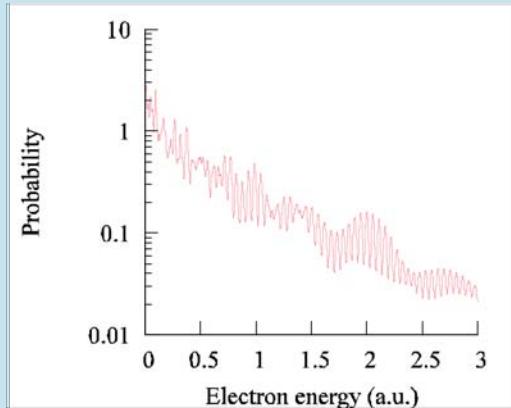


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TDSE



SFA / ADK

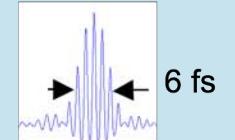
$$a_k = -i \int_0^{T_1} \langle \Psi_k(\tau) | \hat{p} A(\tau) | \Psi_g \rangle d\tau$$

Ψ_k - Volkov state

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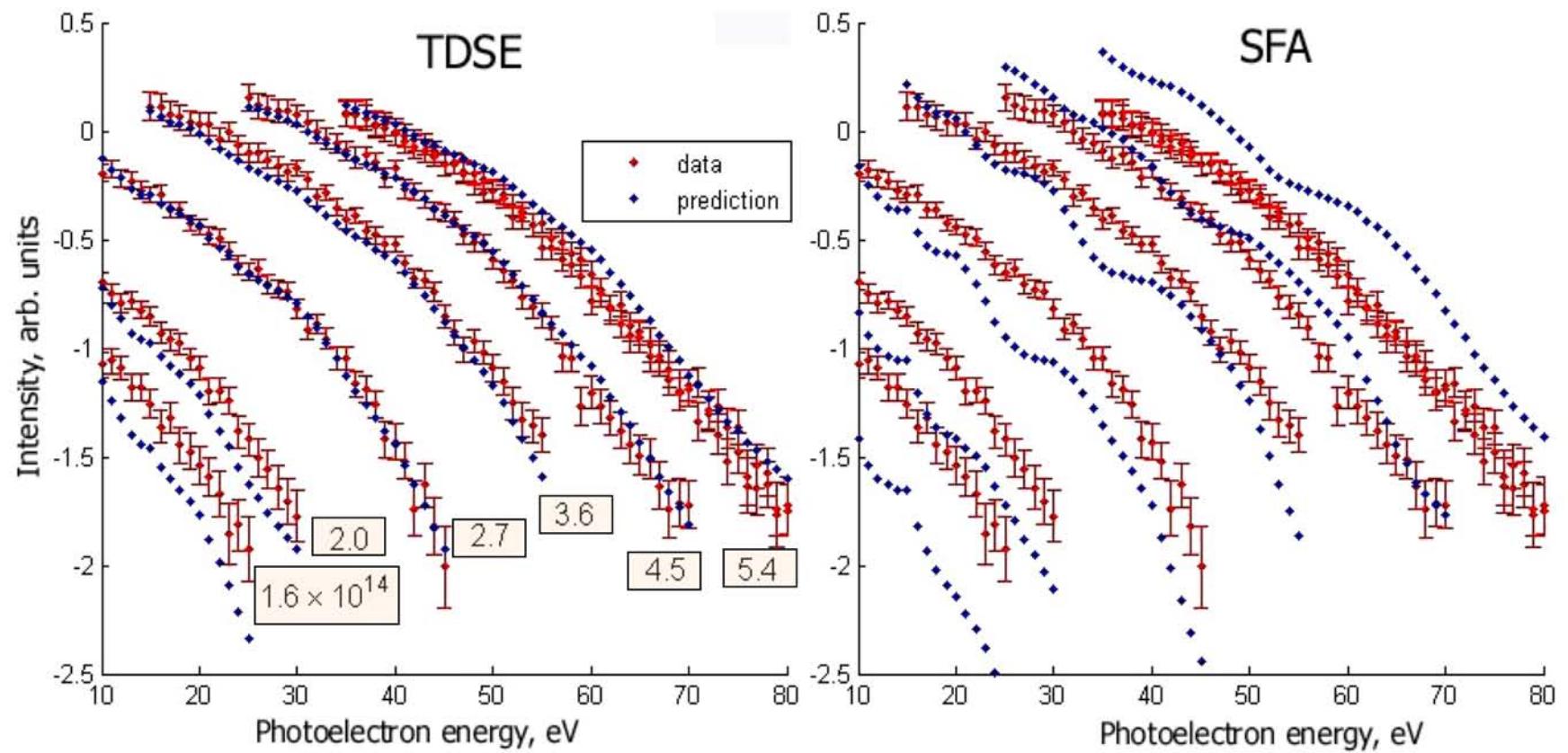
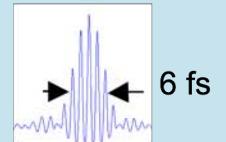


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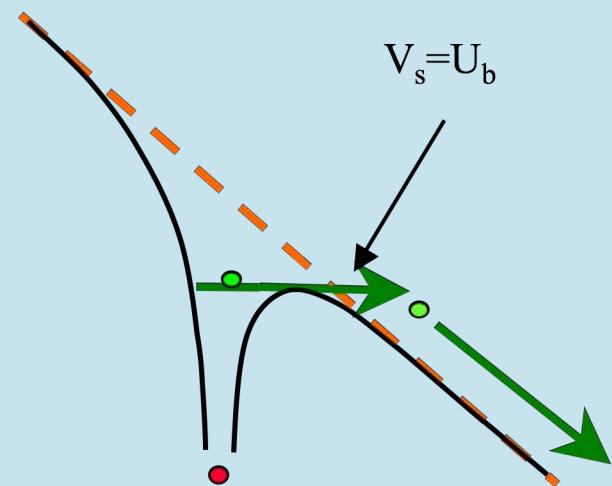
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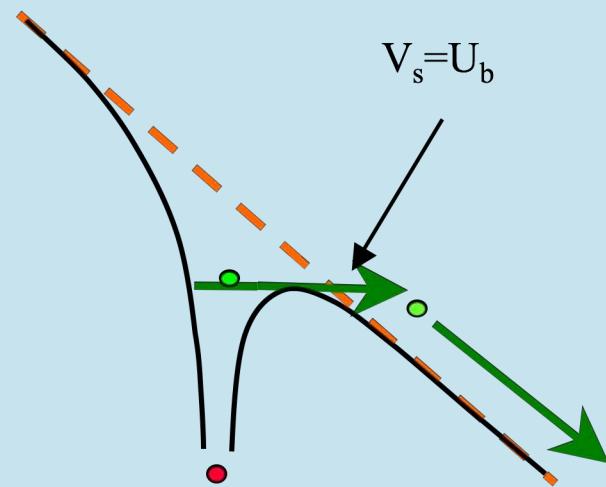
Over-the-barrier ionization

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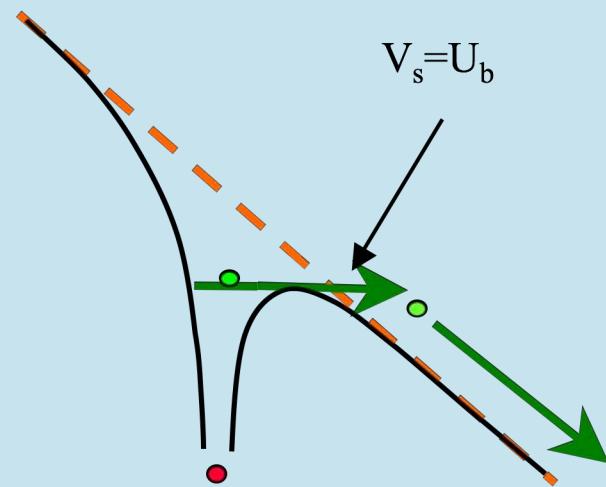
$$V_s = U_b$$

Over-the-barrier ionization



@795nm	IP[eV]	$I_{OBI}[\text{W/cm}^2]$	$\Gamma(I_{OBI})$
Li	5.4	$3.38*10^{12}$	5.2
He	24.6	$1.46*10^{15}$	0.5
Ne	20.2	$2.33*10^{14}$	1.2
Ar	15.8	$8.72*10^{13}$	1.8

Over-the-barrier ionization



@795nm	IP[eV]	$I_{\text{OBI}}[\text{W/cm}^2]$	$\Gamma(I_{\text{OBI}})$
Li	5.4	3.38×10^{12}	5.2
He	24.6	1.46×10^{15}	0.5
Ne	20.2	2.33×10^{14}	1.2
Ar	15.8	8.72×10^{13}	1.8

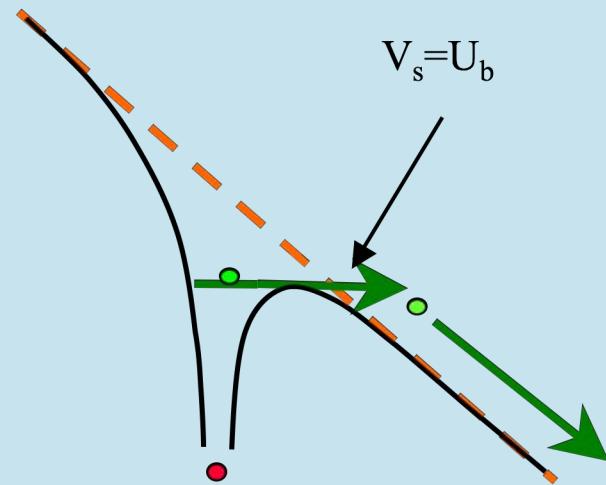
Over-the-barrier field strength (from 1D-model):

$$F_{\text{OBI}} = \frac{\text{IP}^2}{4Z}$$



Ionization probability approaches 1

Over-the-barrier ionization



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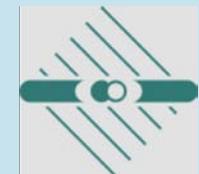
Over-the-barrier intensity:

$$I_{\text{OBI}}[\text{W/cm}^2] = \frac{\pi^2 c \epsilon_0^3 \text{IP}^4}{2e^6 Z^2} = 4 \times 10^9 (\text{IP[eV]})^4 Z^2$$

Strong Field Ionization of Lithium

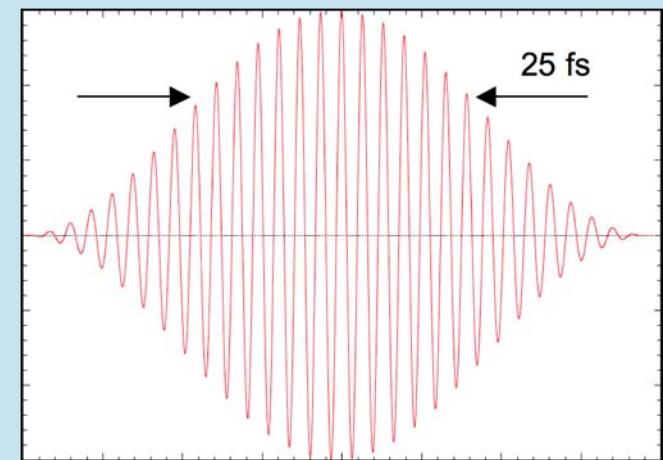
IR: 785 nm, $FWHM=25\text{ fs}$, $I = 2 \cdot 10^{11-14} \text{ W/cm}^2$

MPIK, Heidelberg

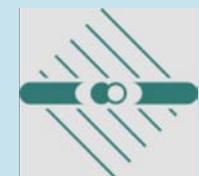


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IR: 785 nm, $FWHM=25 \text{ fs}$, $I = 2 \cdot 10^{11-14} \text{ W/cm}^2$

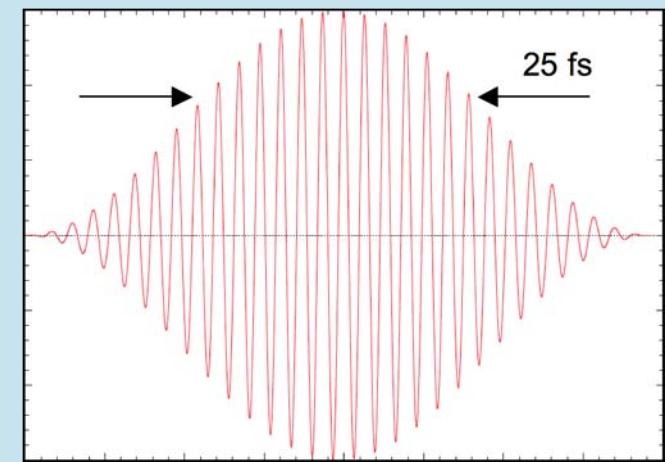
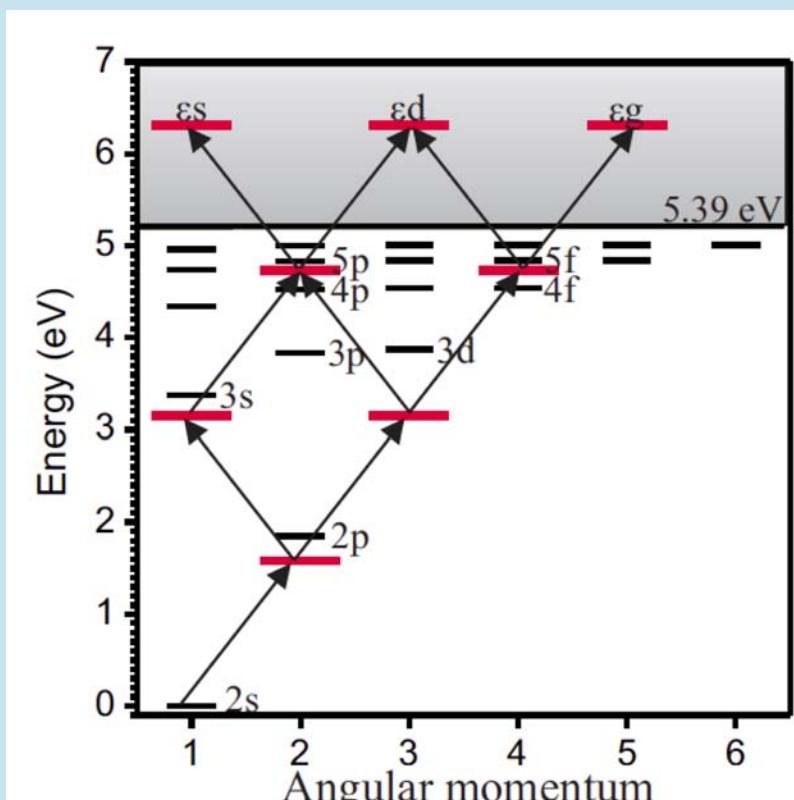


MPIK, Heidelberg

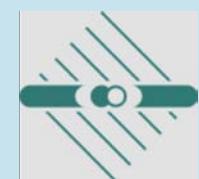


Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25 \text{ fs}$, $I = 2 \cdot 10^{11-14} \text{ W/cm}^2$



MPIK, Heidelberg



Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25$ fs, $I \leq 2 \cdot 10^{11} W/cm^2$

KITP 2010

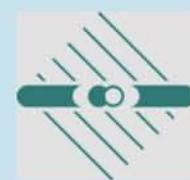
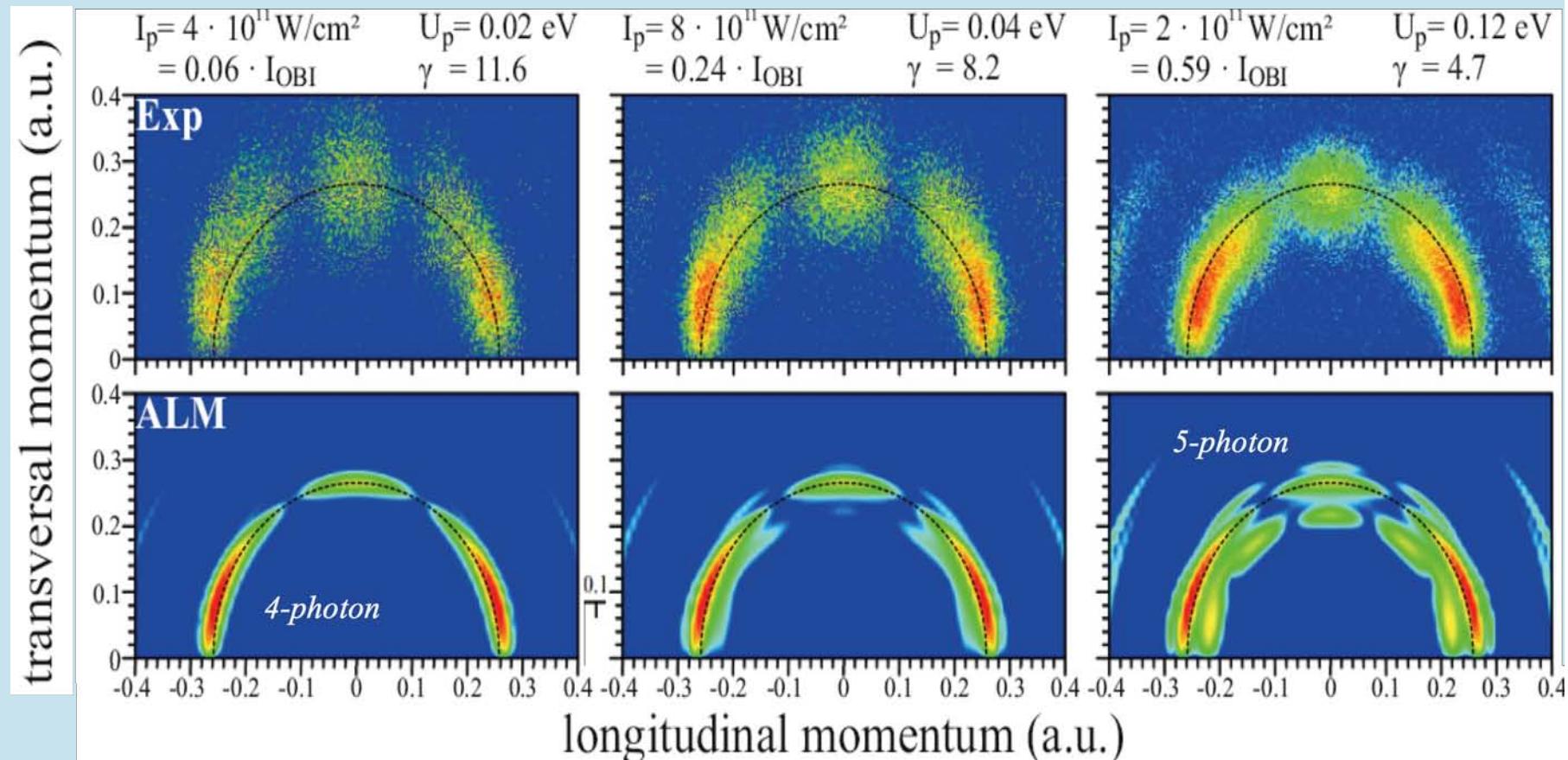
MPIK, Heidelberg



11

Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25$ fs, $I \leq 2 \cdot 10^{11} W/cm^2$

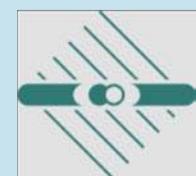


Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25\text{ fs}$, $I \leq 7 \cdot 10^{13} \text{ W/cm}^2$

KITP 2010

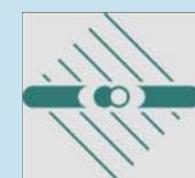
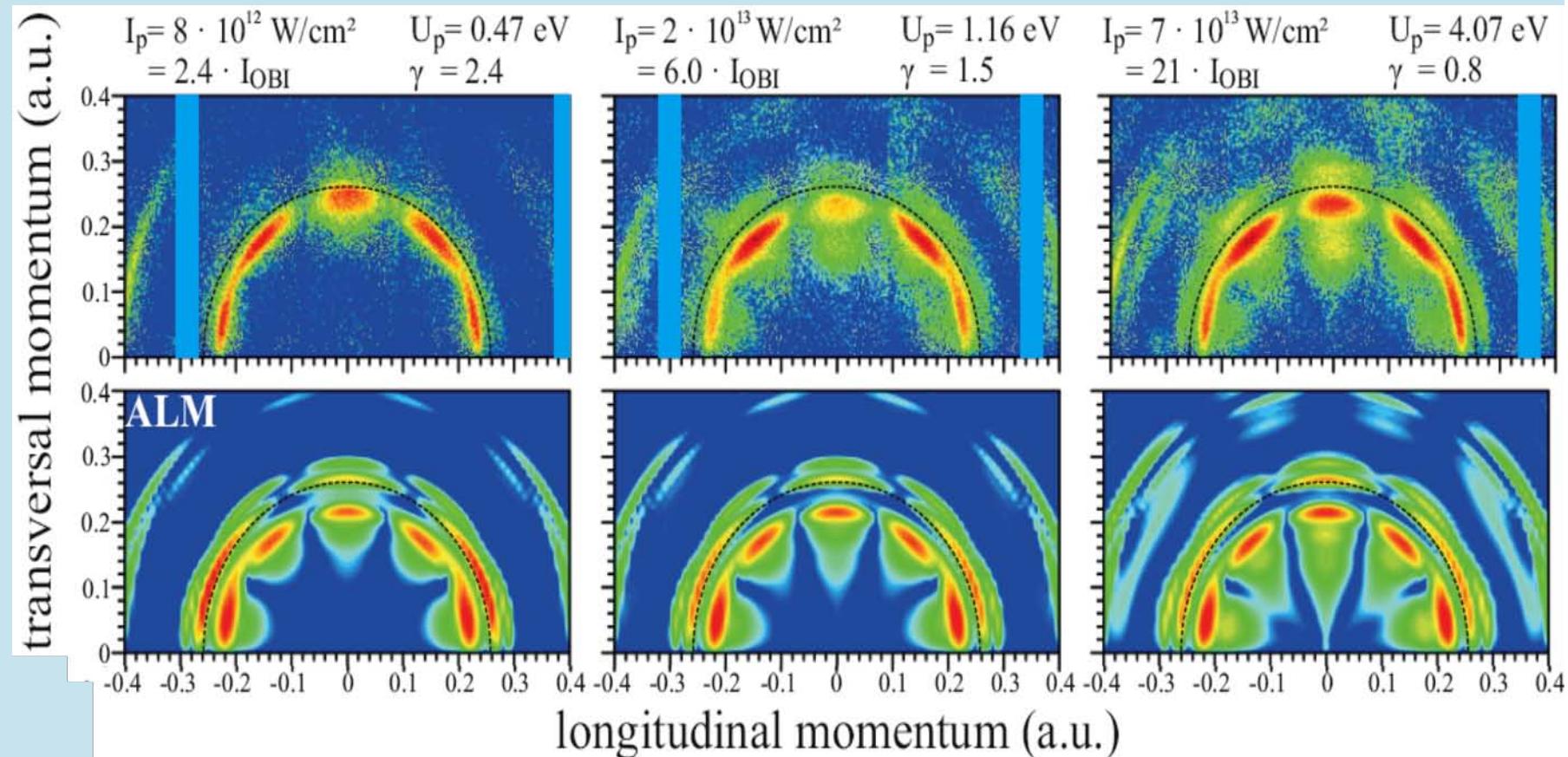
MPIK, Heidelberg



12

Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25\text{ fs}$, $I \leq 7 \cdot 10^{13} \text{ W/cm}^2$



Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25\text{ fs}$, $I = 2 \cdot 10^{13}\text{ W/cm}^2$

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13

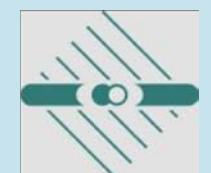
Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25\text{ fs}$, $I = 2 \cdot 10^{13}\text{ W/cm}^2$

Li Atom potentials

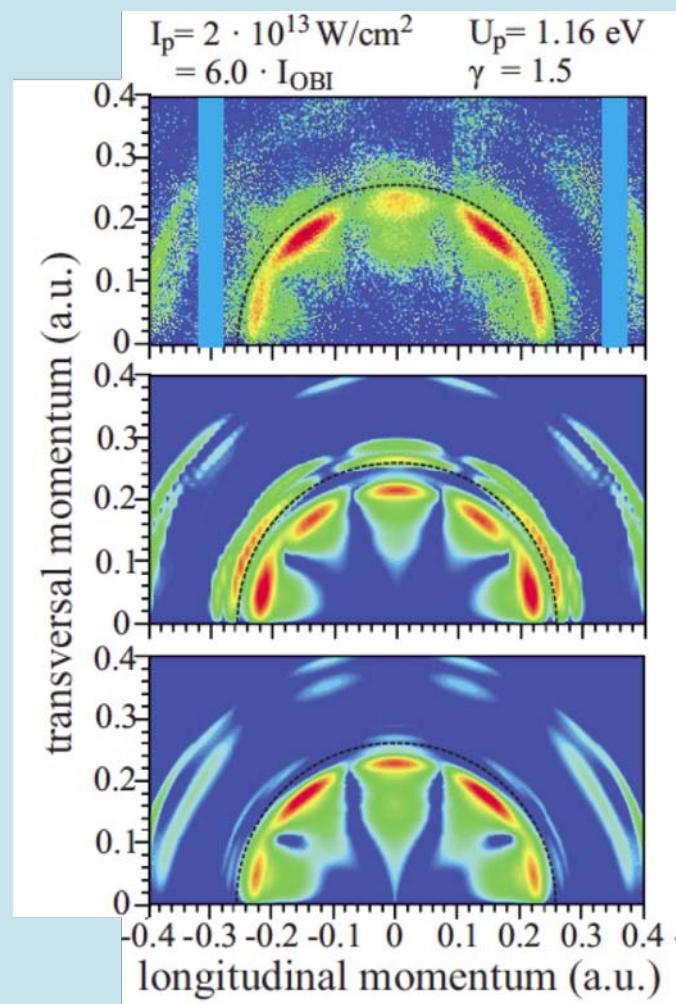
A. Sarsa, F. J. Galvez, and E. Buendia, At. Data Nucl. Data Tables **88**, 163 (2004).

K. Bartschat, *Computational Atomic Physics* (Springer-Verlag (Heidelberg, New York), 1996).



Strong Field Ionization of Lithium

IR: 785 nm, $FWHM=25$ fs, $I = 2 \cdot 10^{13}$ W/cm²



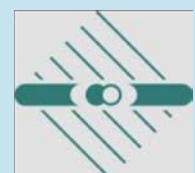
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Li Atom potentials

A. Sarsa, F. J. Galvez, and E. Buendia, At. Data Nucl. Data Tables **88**, 163 (2004).

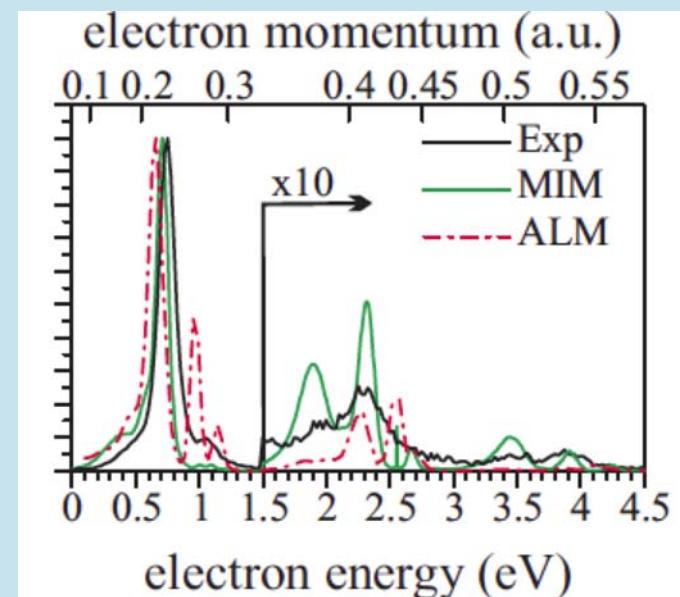
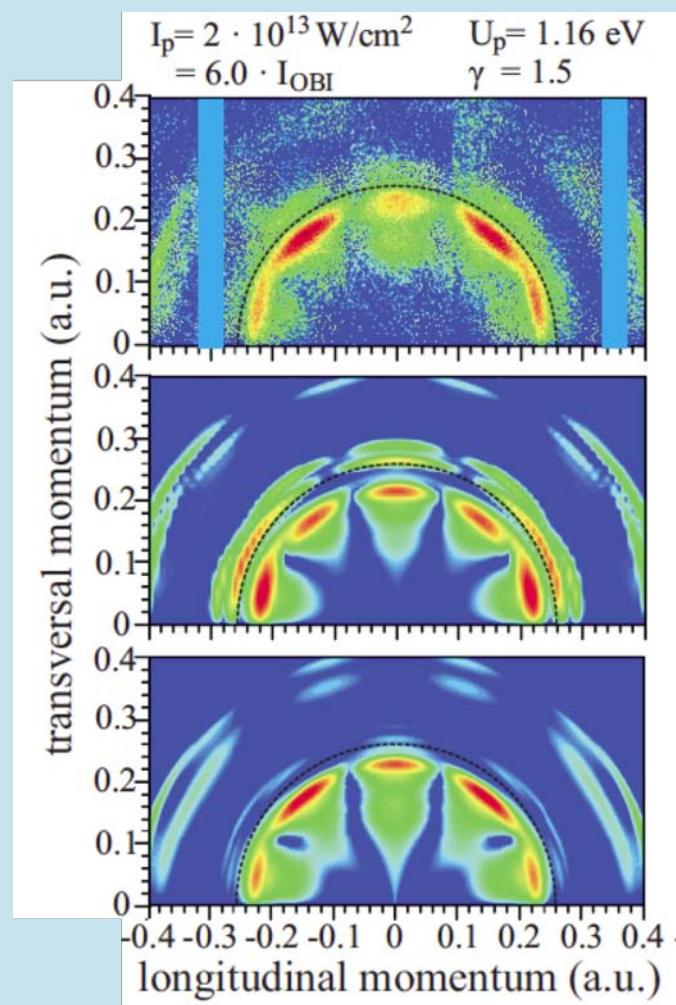
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MPIK, Heidelberg



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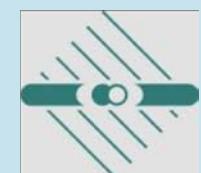
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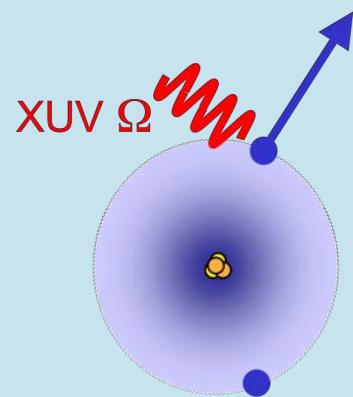
Mechanisms of double photo-ionization

Mechanisms of double photo-ionization

Shake-off

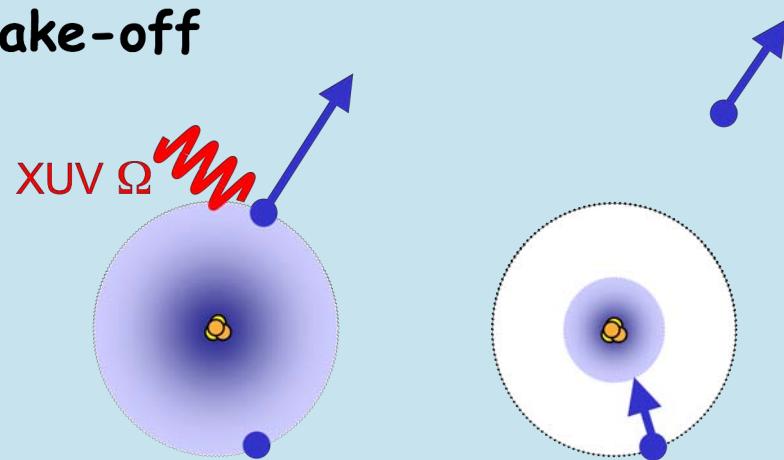
Mechanisms of double photo-ionization

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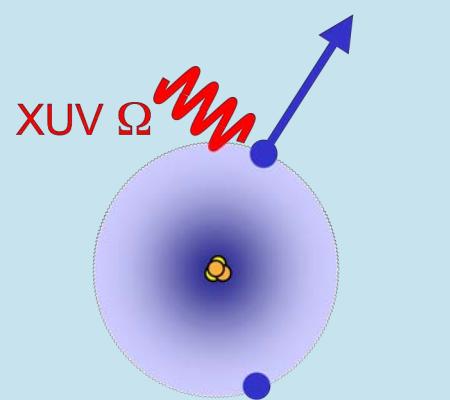
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Mechanisms of double photo-ionization

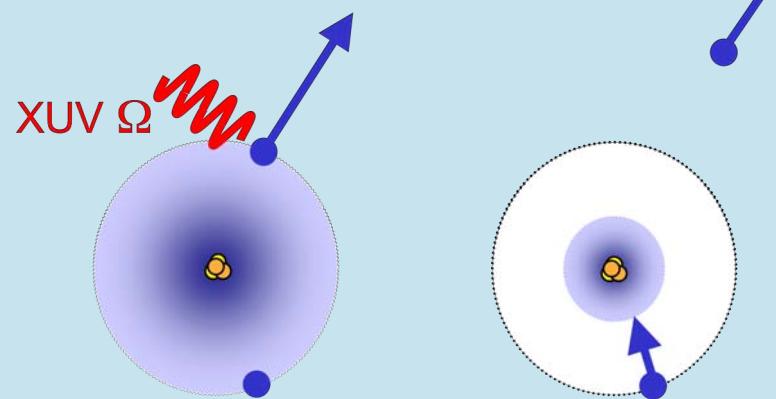
Shake-off



Fast, $\Omega \gg DIP$

Mechanisms of double photo-ionization

Shake-off

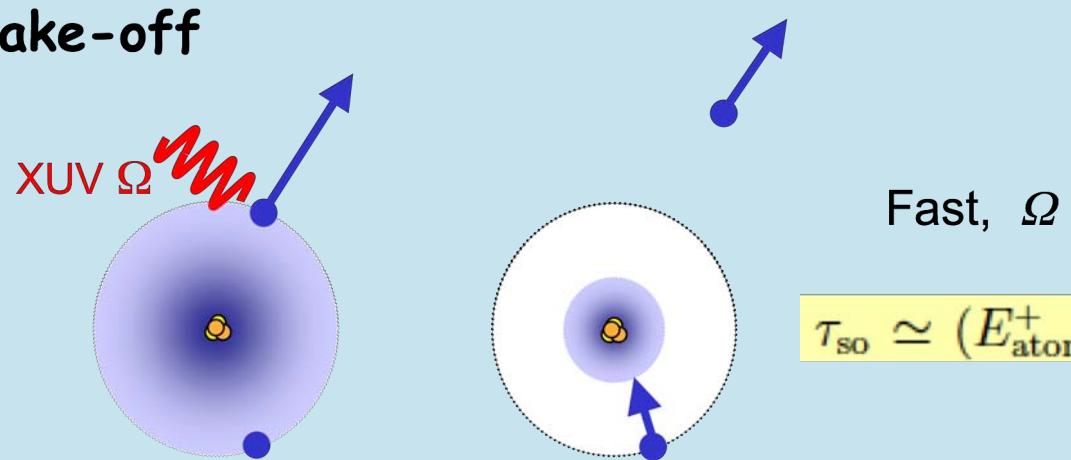


Fast, $\Omega \gg DIP$

$$\tau_{\text{so}} \simeq (E_{\text{atom}}^+ - E_{\text{ion}}^+)^{-1} \simeq 20 \text{ as}$$

Mechanisms of double photo-ionization

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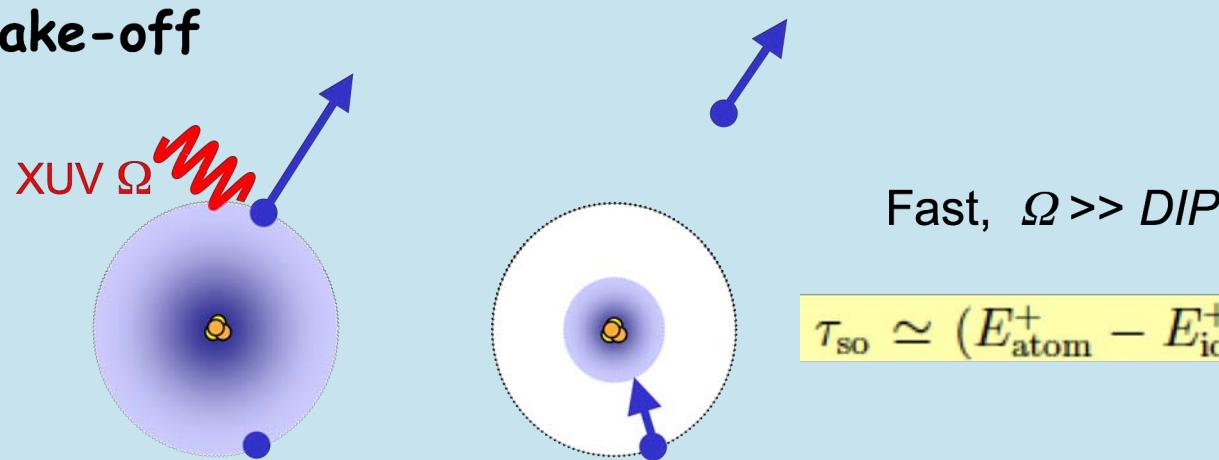
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Recollision, or
knock-out

Mechanisms of double photo-ionization

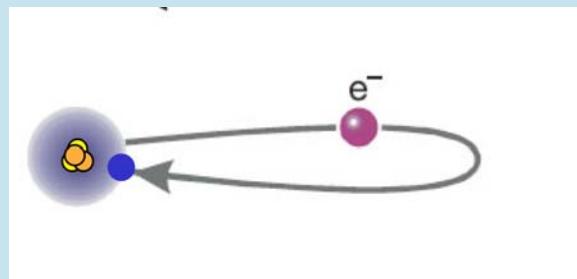
Shake-off



Fast, $\Omega \gg DIP$

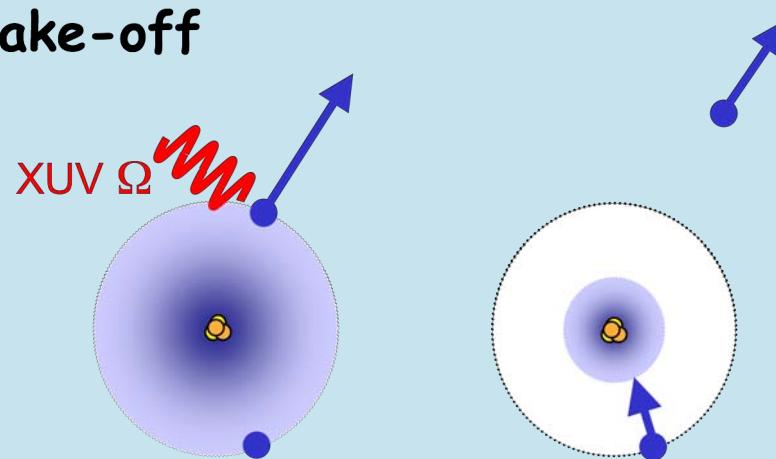
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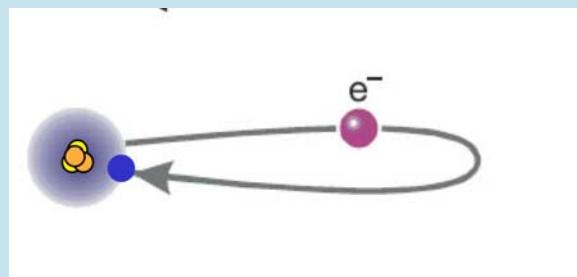
Shake-off



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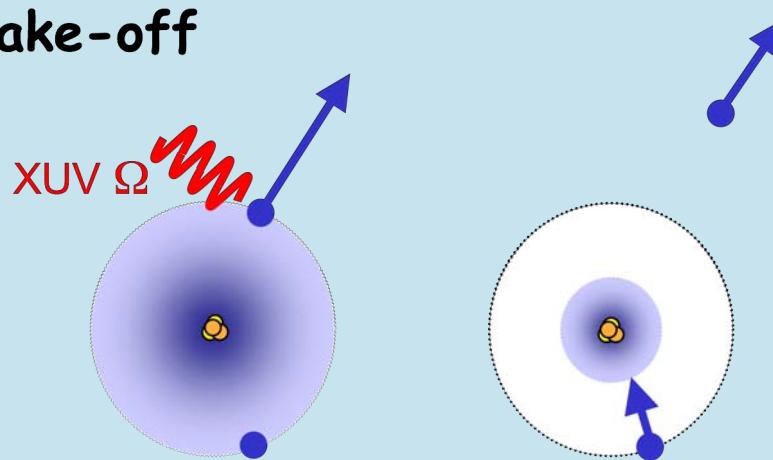
Recollision, or
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Slow, $\Omega - DIP \ll DIP$

Mechanisms of double photo-ionization

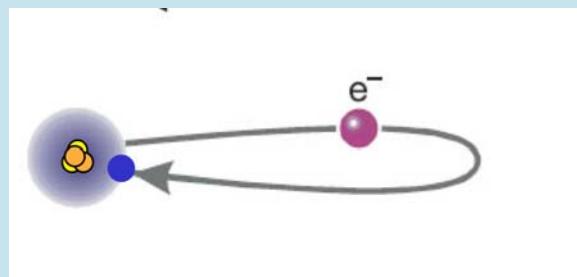
Shake-off



Fast, $\Omega \gg DIP$

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Recollision, or
knock-out



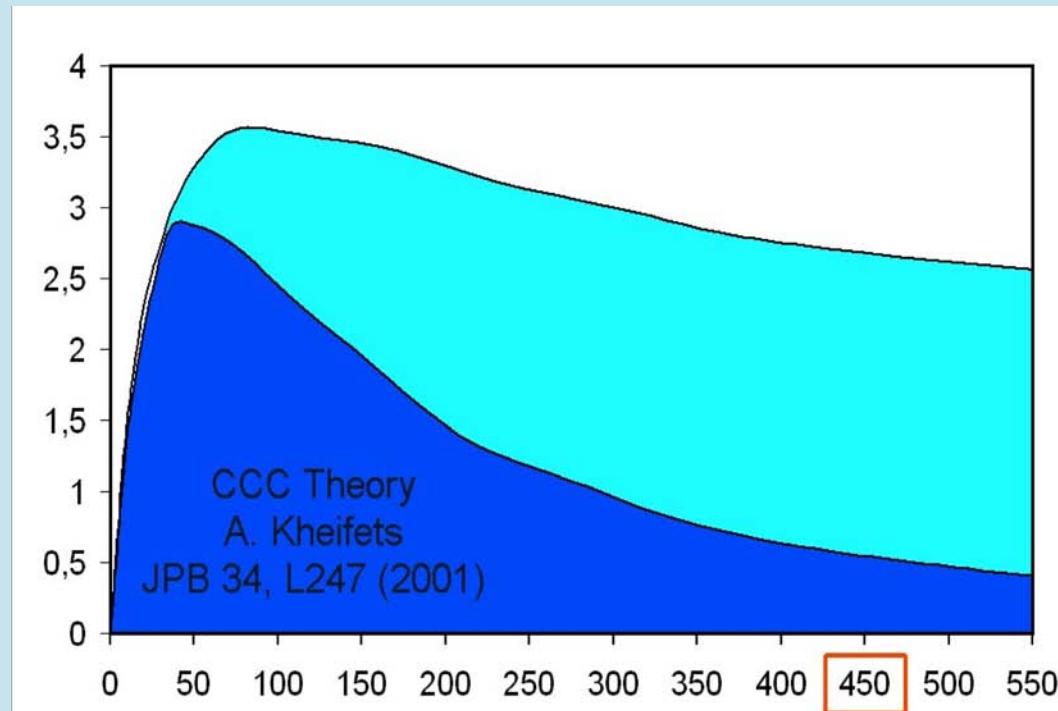
Slow, $\Omega - DIP \ll DIP$

$$\tau_{\text{ko}} \simeq (E_{Z^*} - E_{\text{exact}})^{-1} \simeq 200 \text{ as}$$

Ratio of double to single ionization in He

Ratio of double to single ionization in He

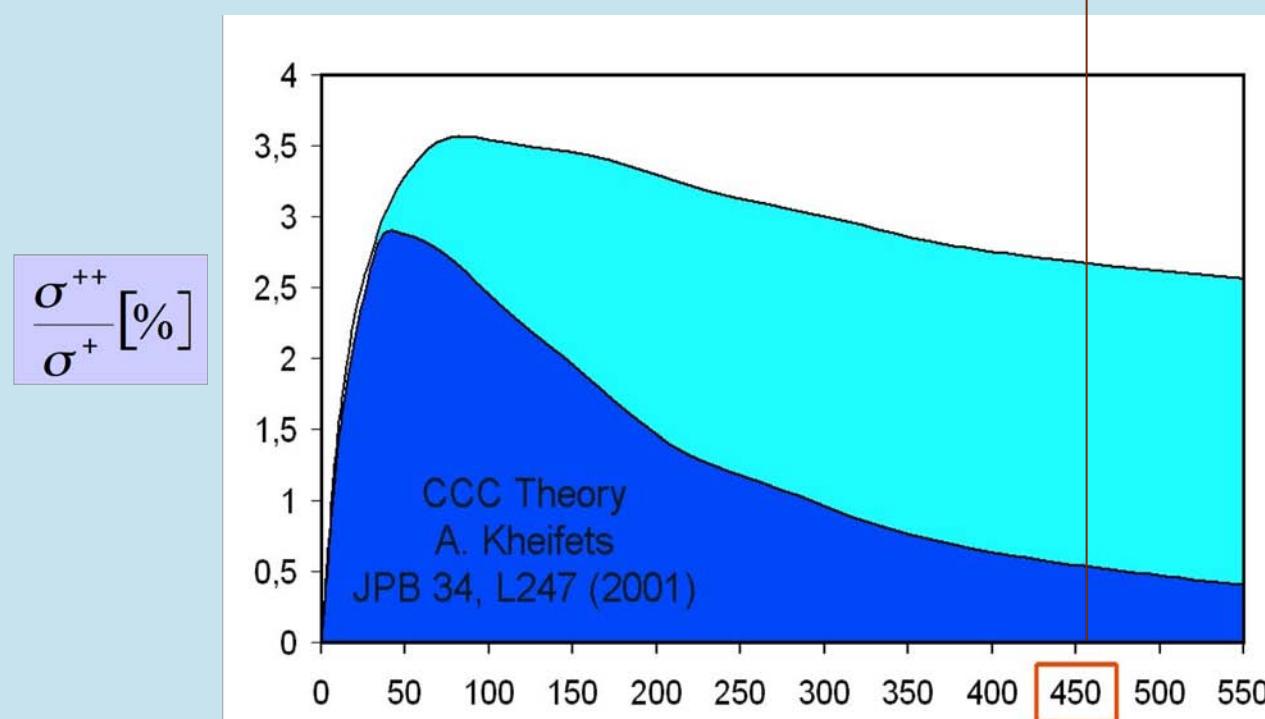
$$\frac{\sigma^{++}}{\sigma^+} [\%]$$



Photon energy above the threshold [eV]

Ratio of double to single ionization in He

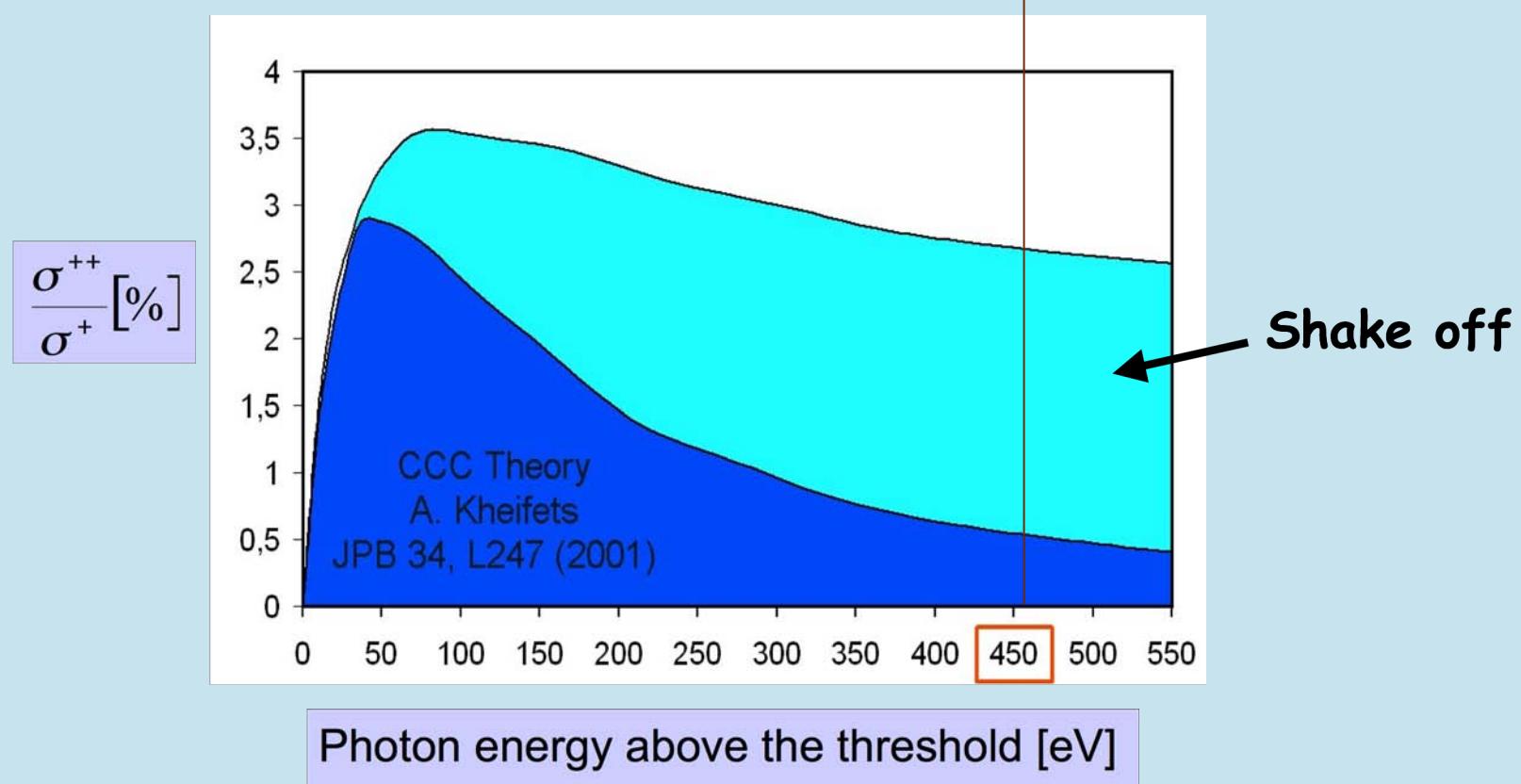
COLTRIMS experiment
Knapp et al PRL 89:033004, 2002



Photon energy above the threshold [eV]

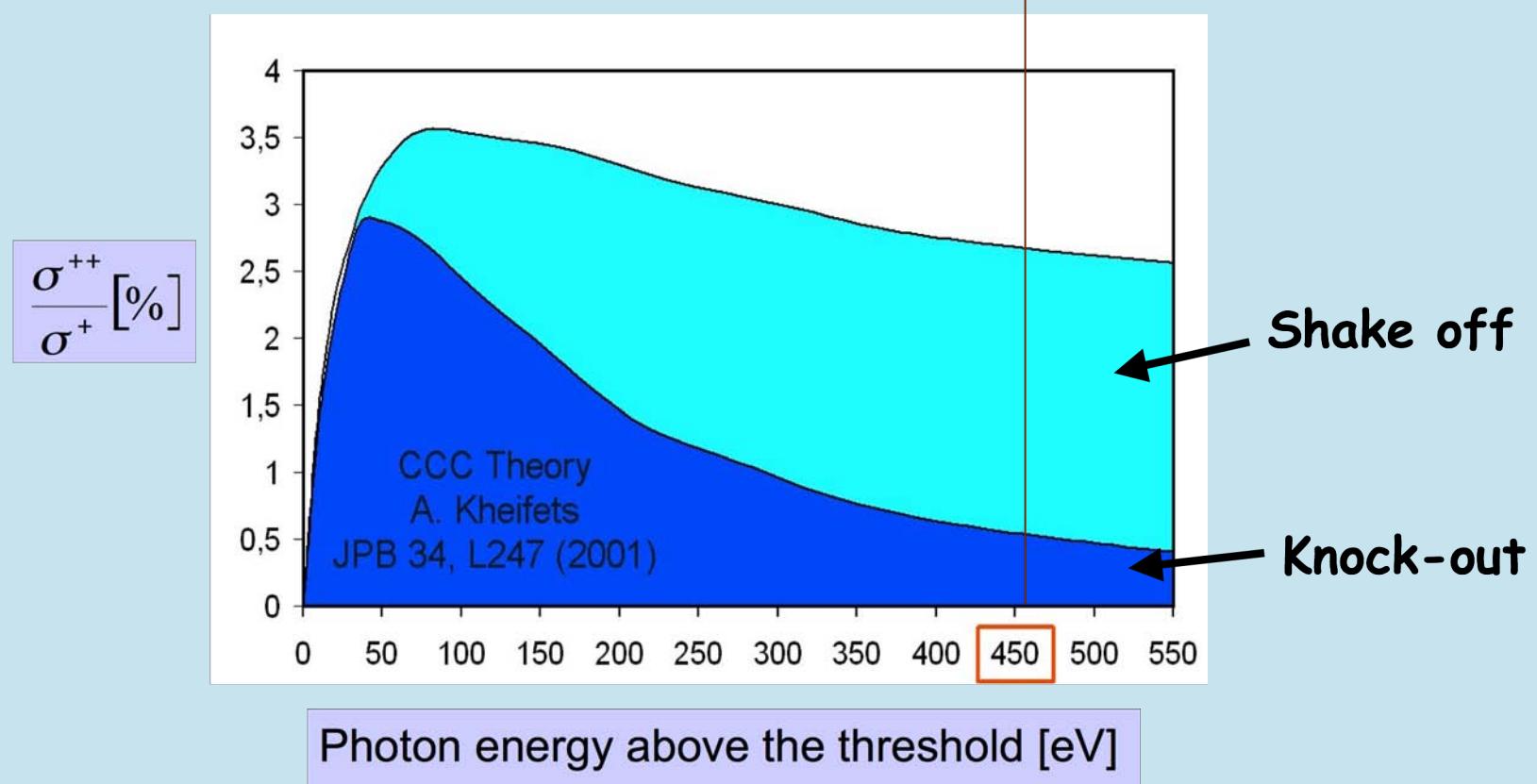
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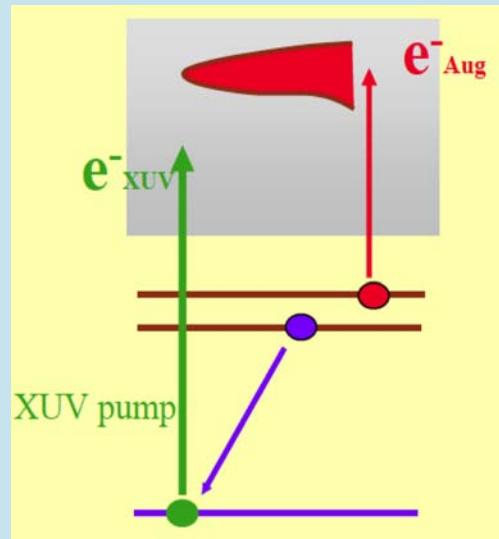
Use of Electron Correlation to Make Attosecond Measurements without Attosecond Pulses

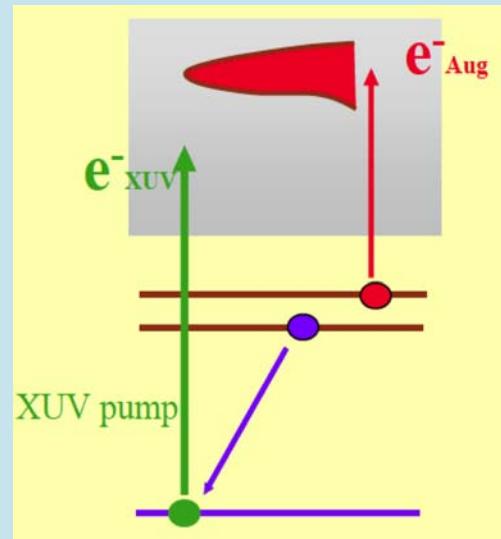
Olga Smirnova,¹ Vladislav S. Yakovlev,² and Misha Ivanov³

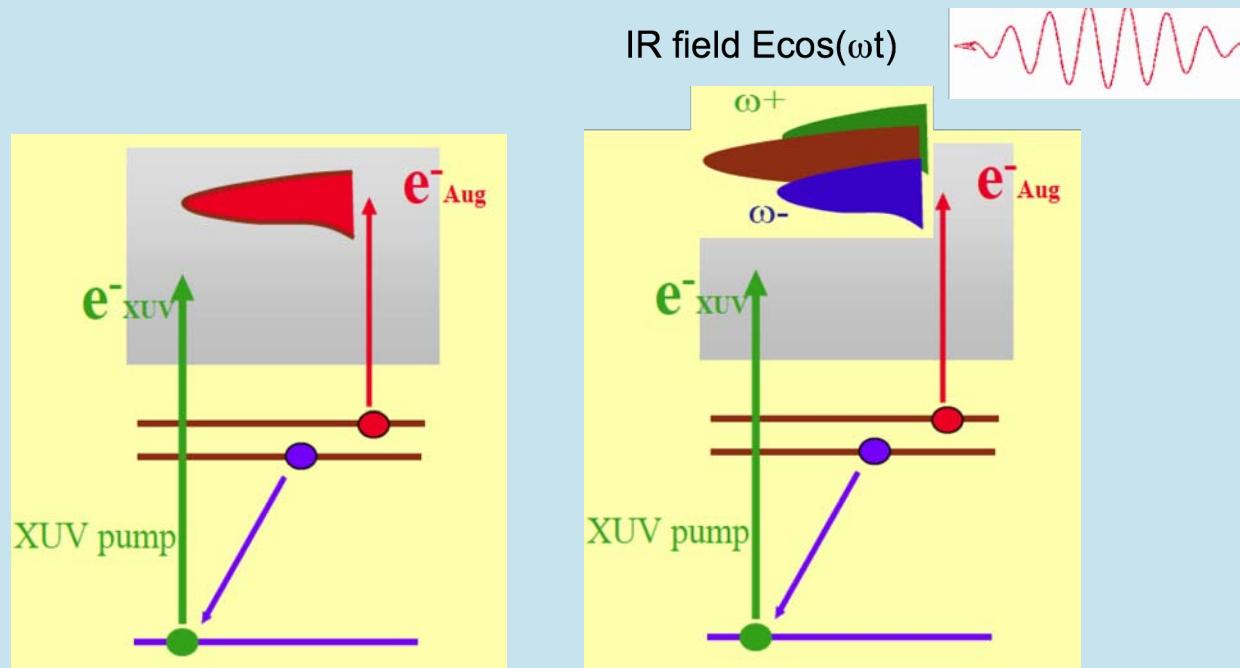
¹*Photonics Institute, Vienna University of Technology, Gusshausstrasse 27/387, A-1040 Vienna, Austria*

²*Department für Physik, Ludwig-Maximilians-Universität München, am Coulombwall 1, D-85748 Garching, Germany*

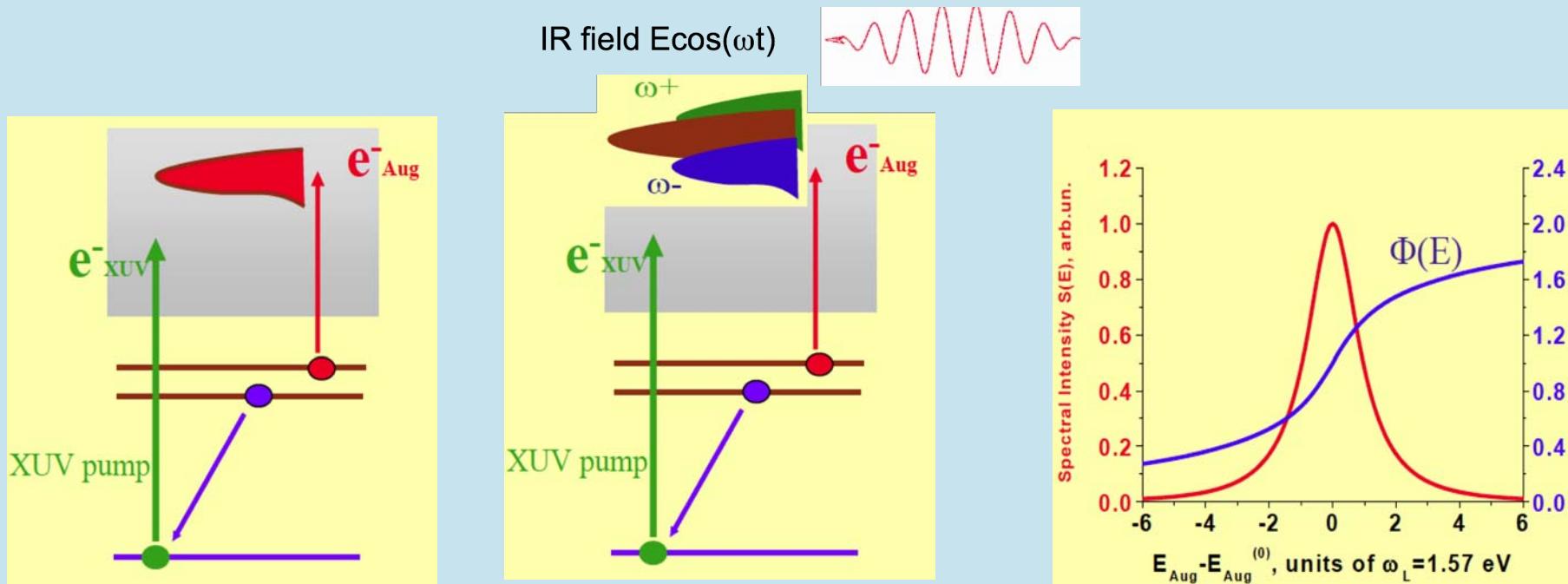
³*NRC Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6 Canada*

Use of Electron Correlation to Make Attosecond Measurements without Attosecond PulsesOlga Smirnova,¹ Vladislav S. Yakovlev,² and Misha Ivanov³¹*Photonics Institute, Vienna University of Technology, Gusshausstrasse 27/387, A-1040 Vienna, Austria*²*Department für Physik, Ludwig-Maximilians-Universität München, am Coulombwall 1, D-85748 Garching, Germany*³*NRC Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6 Canada*

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Use of Electron Correlation to Make Attosecond Measurements without Attosecond PulsesOlga Smirnova,¹ Vladislav S. Yakovlev,² and Misha Ivanov³¹*Photonics Institute, Vienna University of Technology, Gusshausstrasse 27/387, A-1040 Vienna, Austria*²*Department für Physik, Ludwig-Maximilians-Universität München, am Coulombwall 1, D-85748 Garching, Germany*³*NRC Canada, 100 Sussex Drive, Ottawa, Ontario K1A 0R6 Canada*

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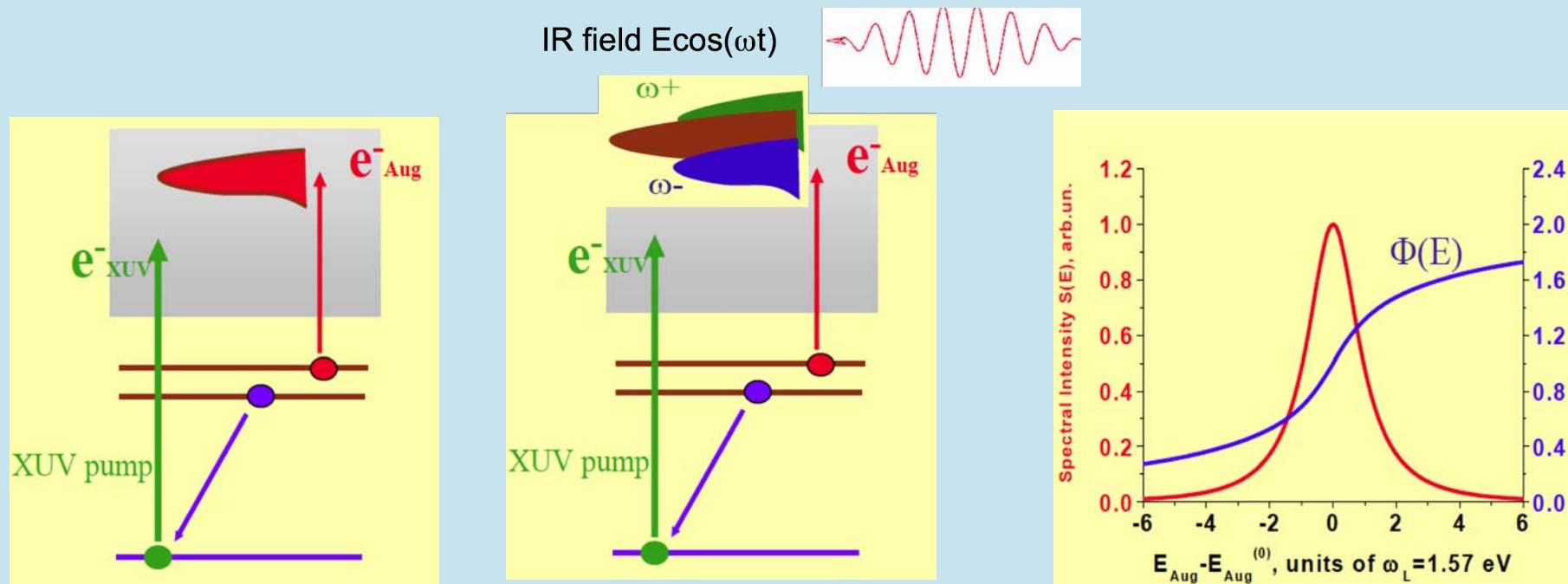
Use of Electron Correlation to Make Attosecond Measurements without Attosecond Pulses

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Dynamics of the decay: direct time domain measurement OR the spectral phase $\Phi(E)$
Correlated Atto-Second Two-electron Optical Reconstruction

SPIDER

**SPectral shearing Interferometry for Direct
Electric field Reconstruction**

SPIDER

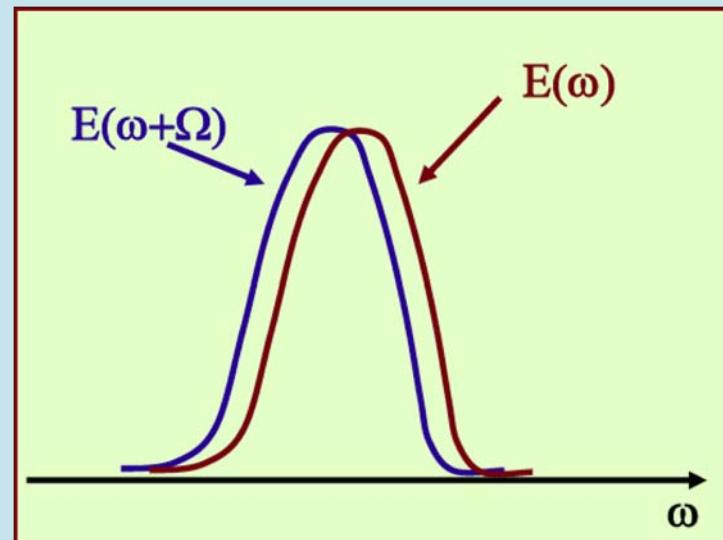
SPectral shearing Interferometry for Direct Electric field Reconstruction

$$\begin{aligned} E(\omega) &= |E(\omega)| e^{i\varphi(\omega)} \\ S(\omega, \tau) &= |E(\omega) + e^{i\omega\tau} E(\omega + \Omega)|^2 = \\ &= |E(\omega)|^2 + |E(\omega + \Omega)|^2 + |E(\omega)| |E(\omega + \Omega)| \cos(\underbrace{\omega\tau + \varphi(\omega + \Omega) - \varphi(\omega)}_{\theta(\omega)}) \end{aligned}$$

SPIDER

SPectral shearing Interferometry for Direct Electric field Reconstruction

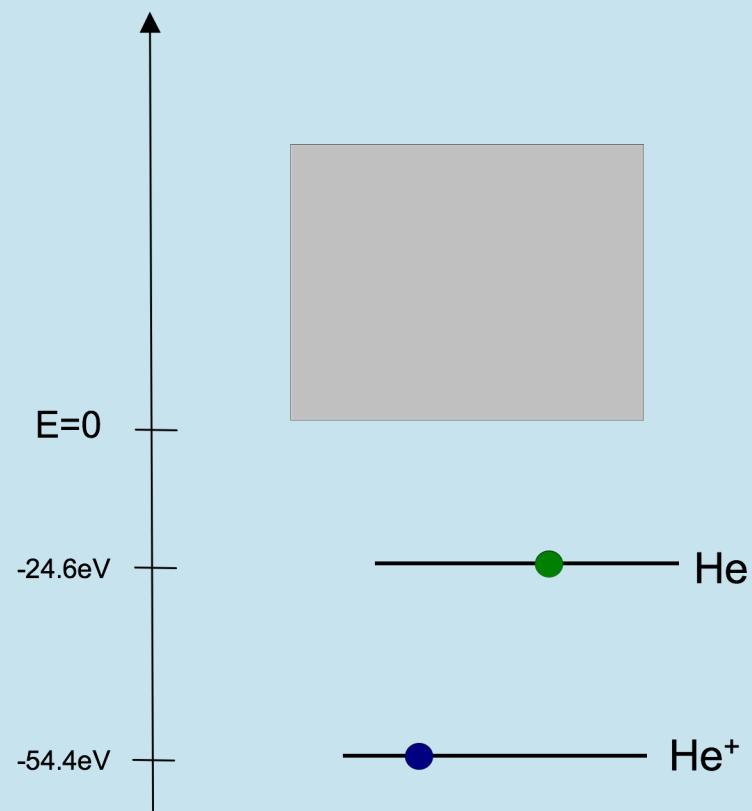
$$\begin{aligned}
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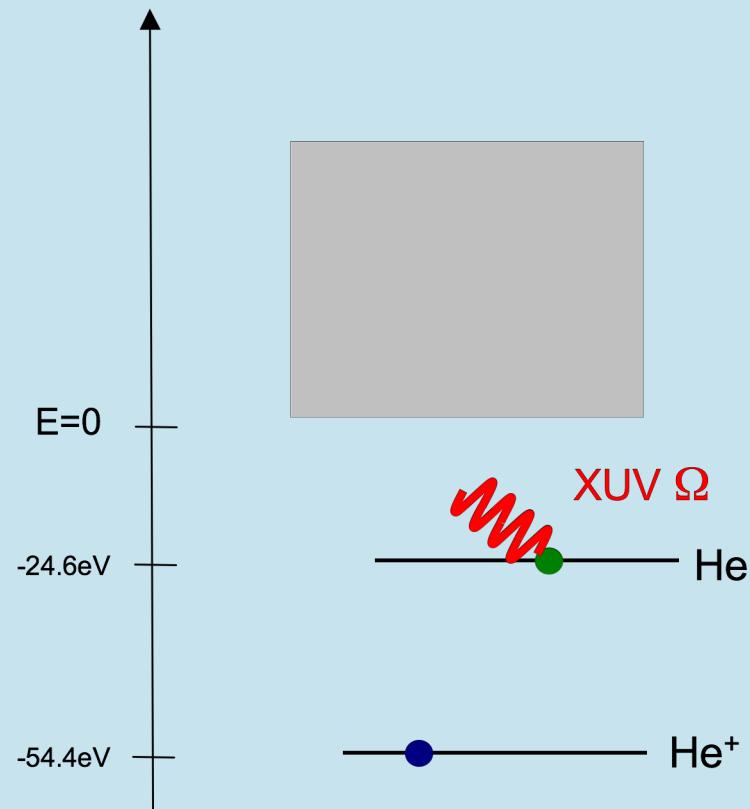
Photoelectron energy distribution in He

XUV only

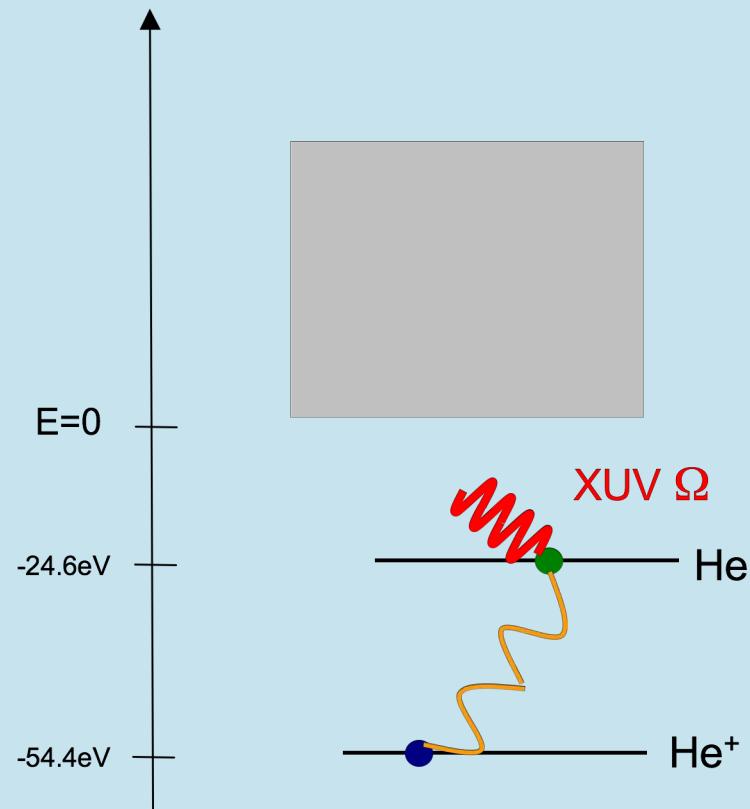
Photoelectron energy distribution in He XUV only



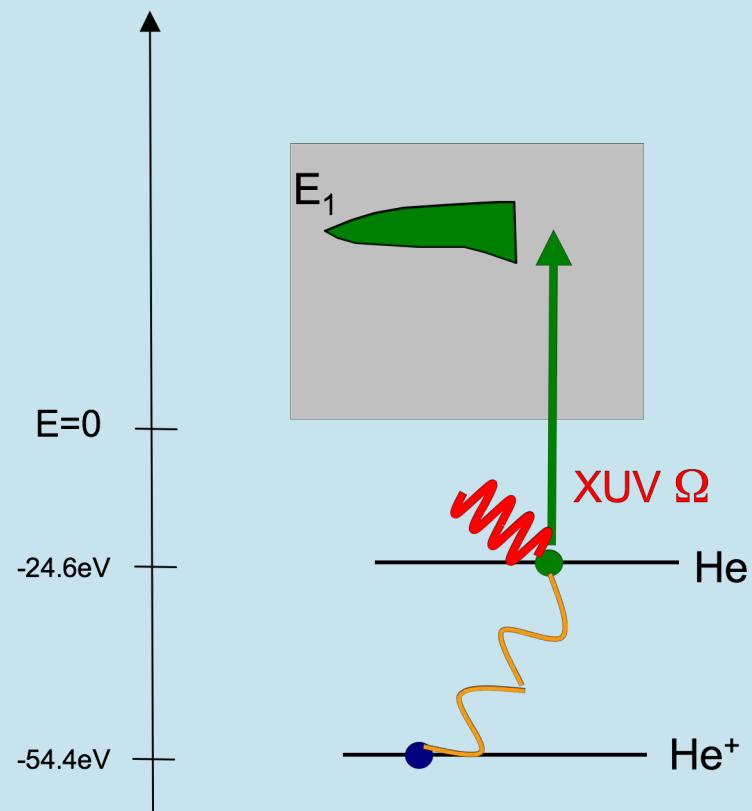
Photoelectron energy distribution in He XUV only



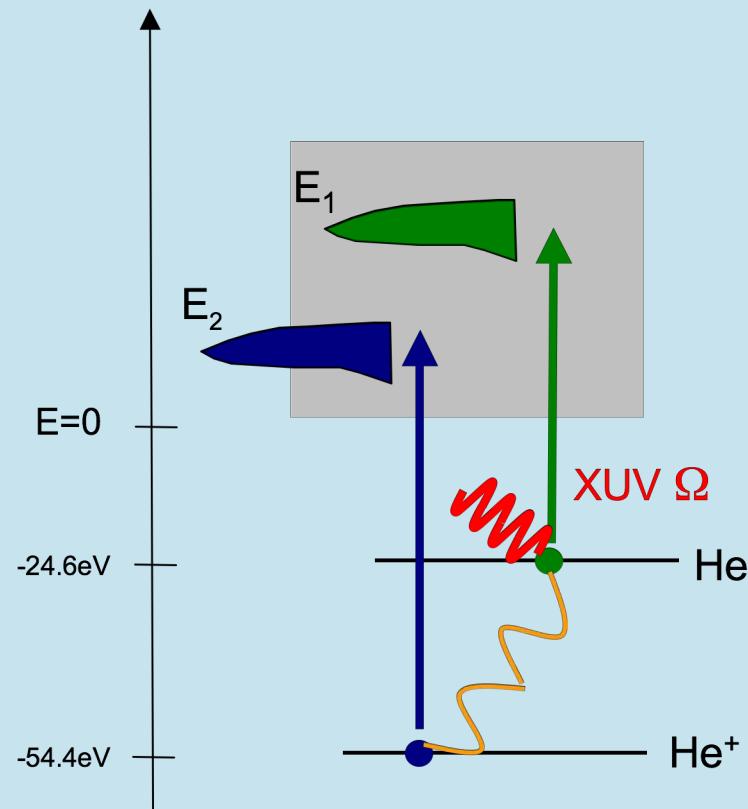
Photoelectron energy distribution in He XUV only



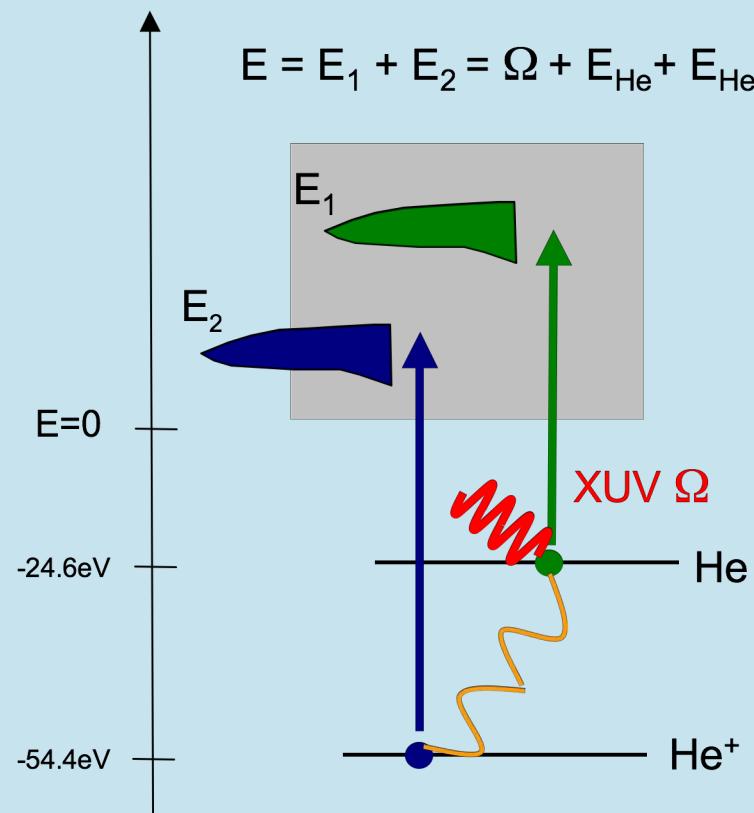
Photoelectron energy distribution in He XUV only



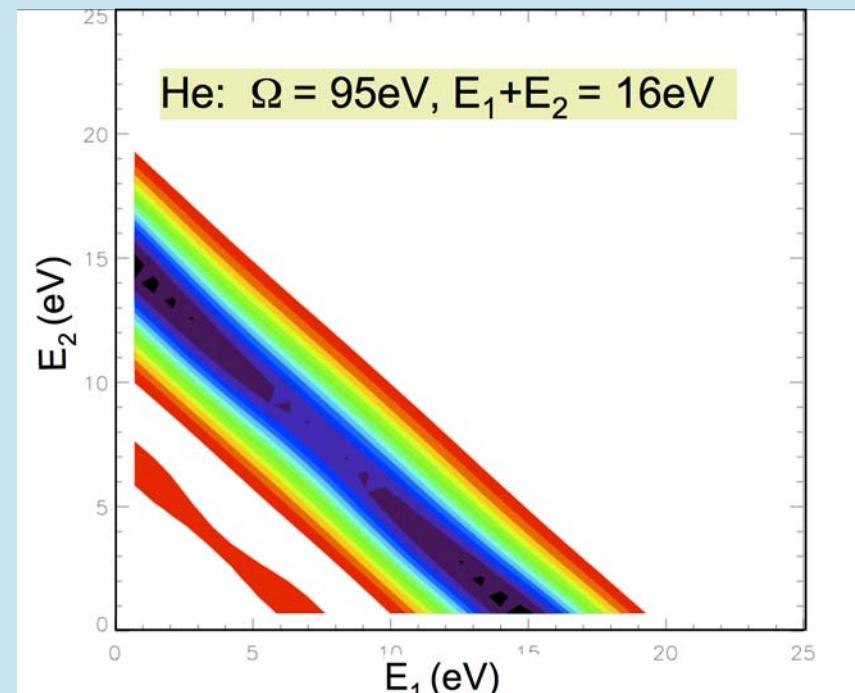
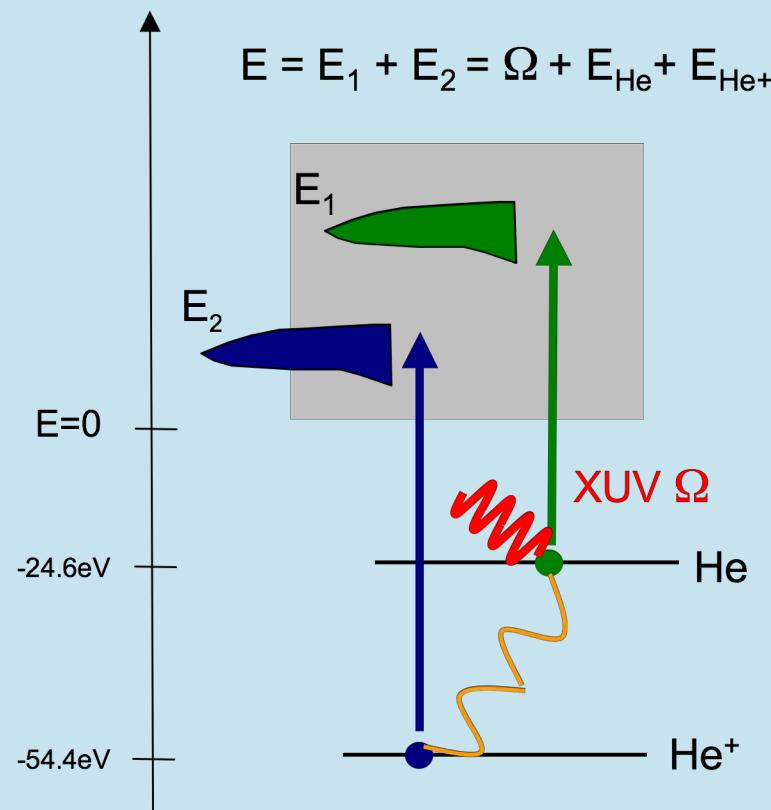
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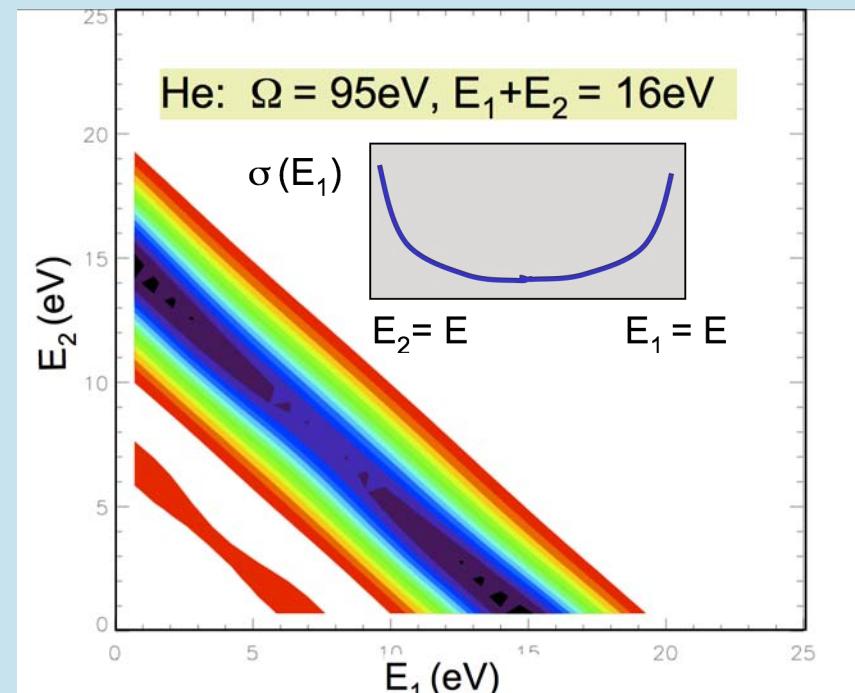
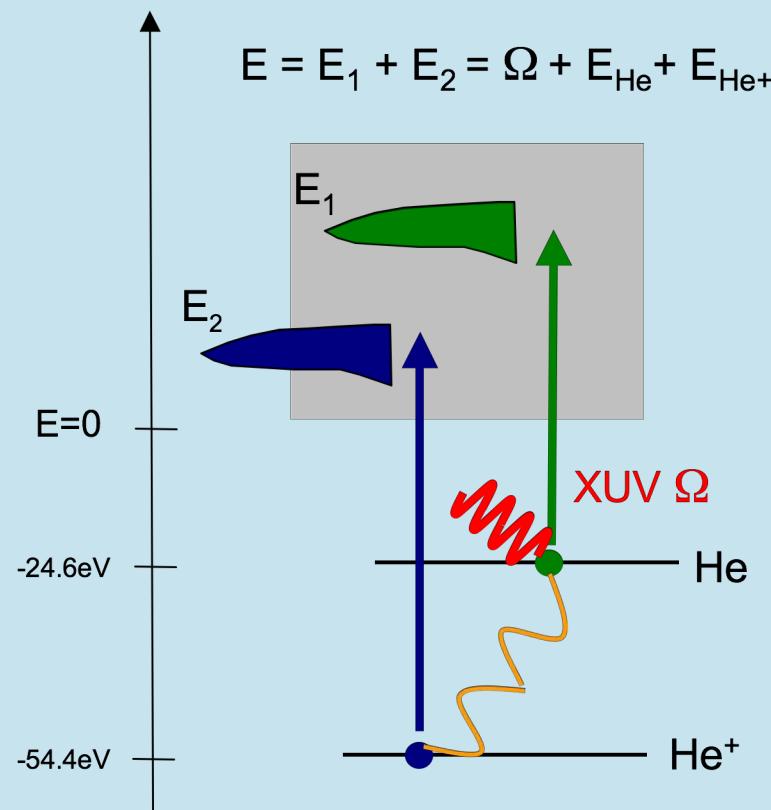
Photoelectron energy distribution in He XUV only



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Photoelectron energy distribution in He XUV only

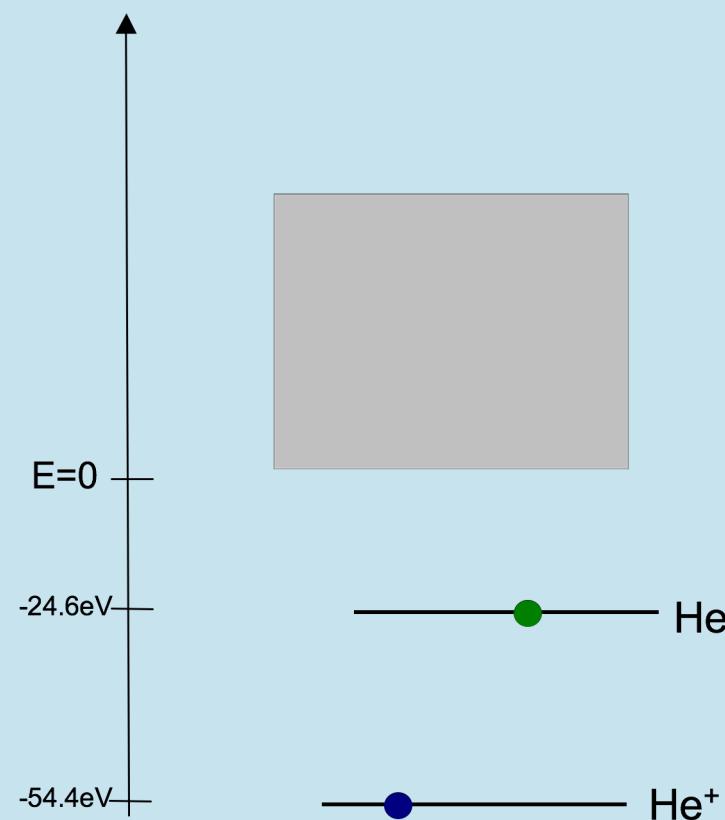


Photoelectron energy distribution in He

XUV + IR

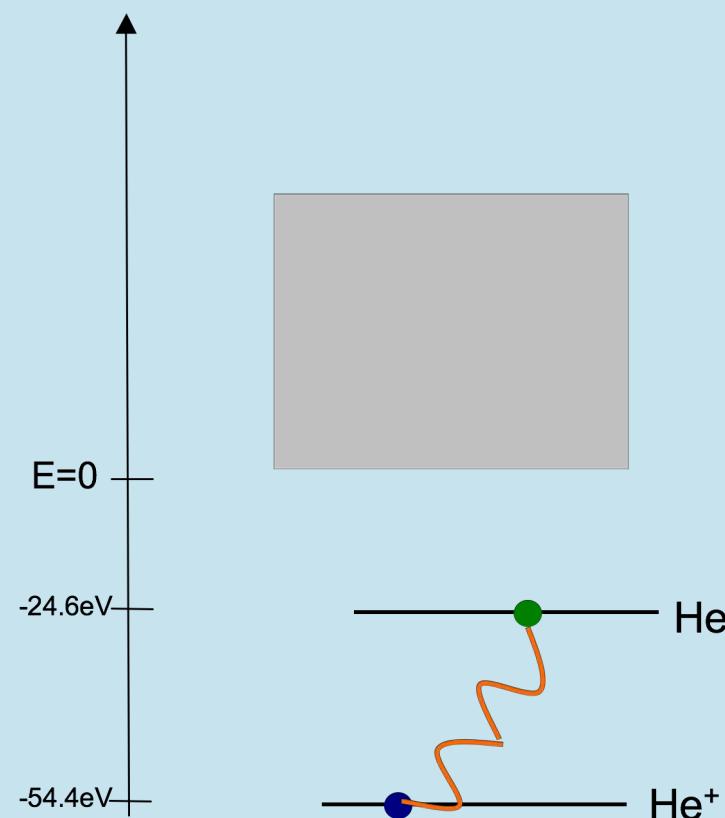
Photoelectron energy distribution in He

XUV + IR



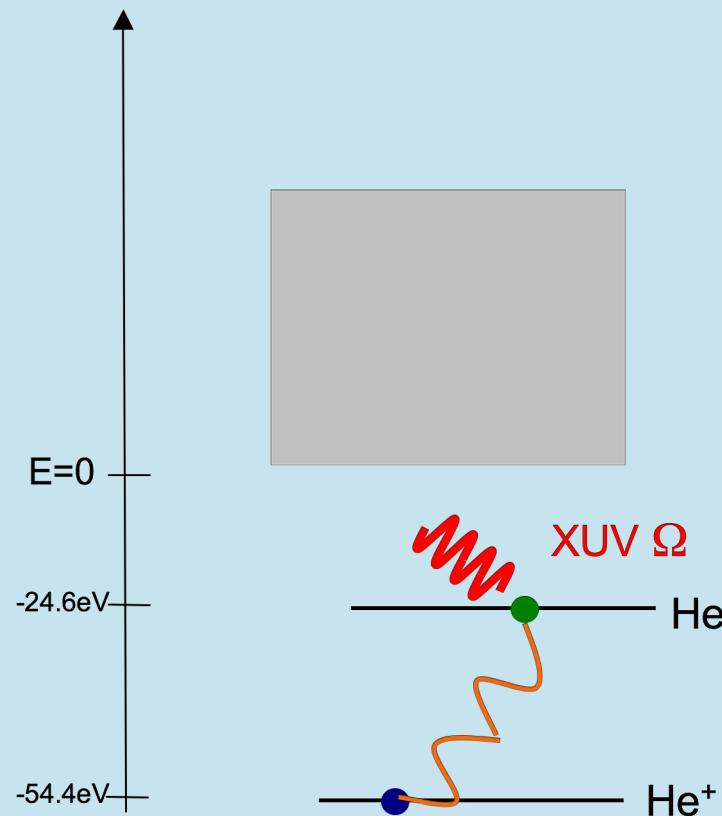
Photoelectron energy distribution in He

XUV + IR



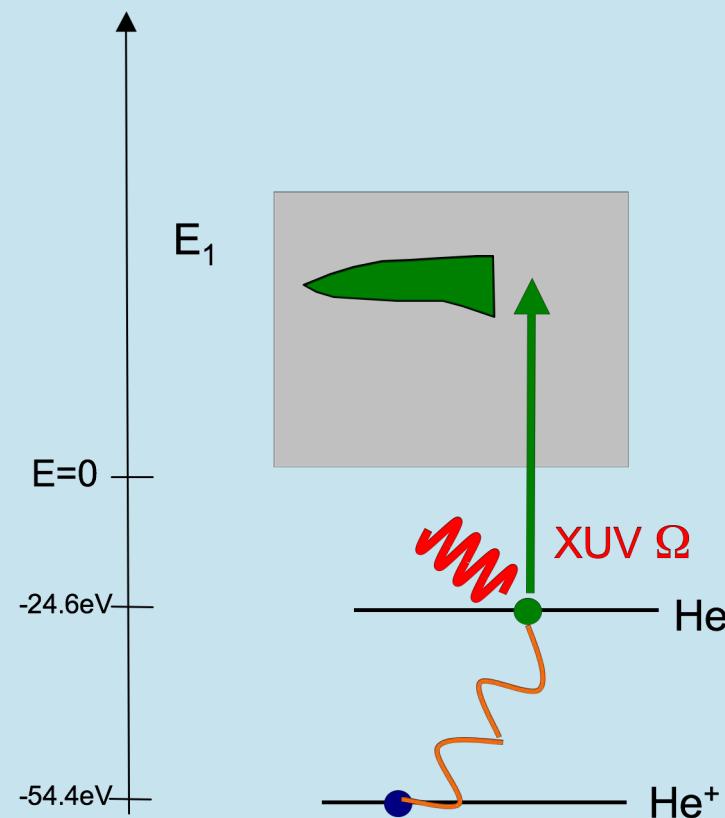
Photoelectron energy distribution in He

XUV + IR



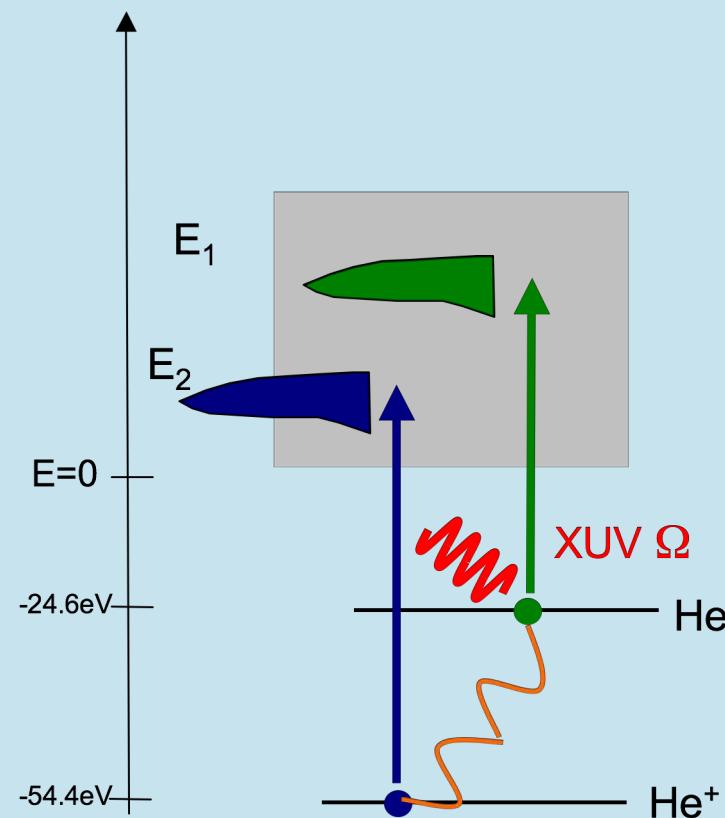
Photoelectron energy distribution in He

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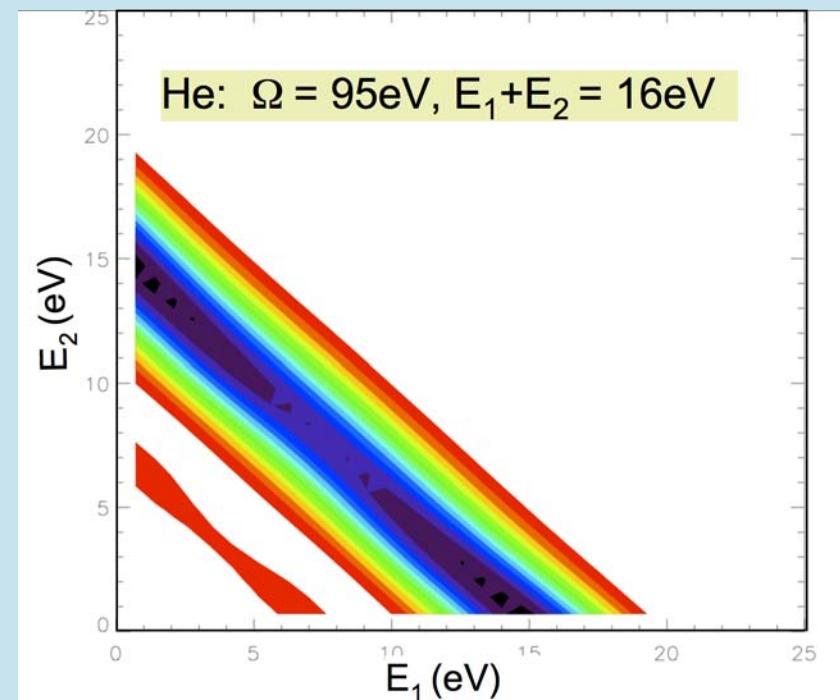
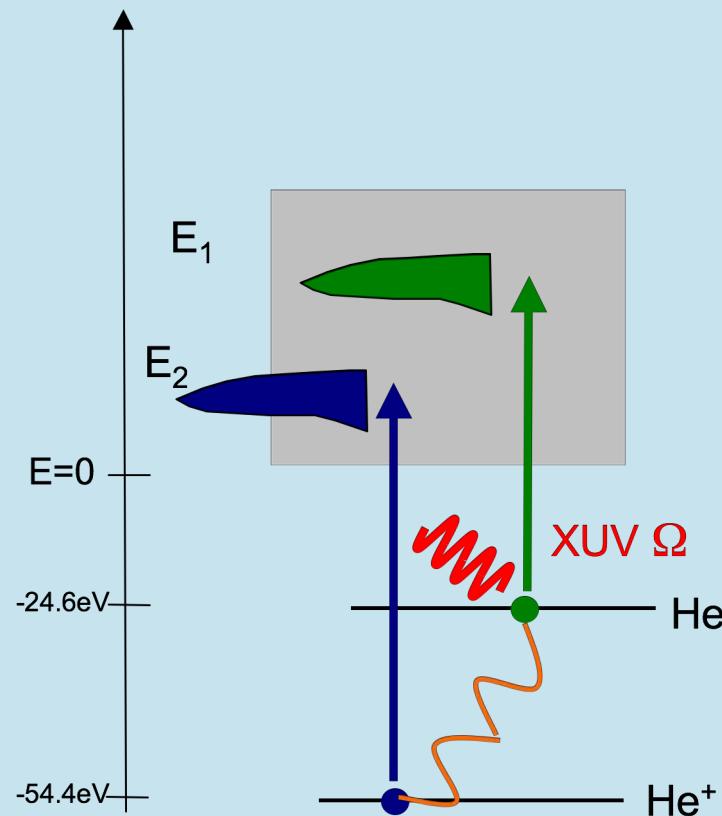


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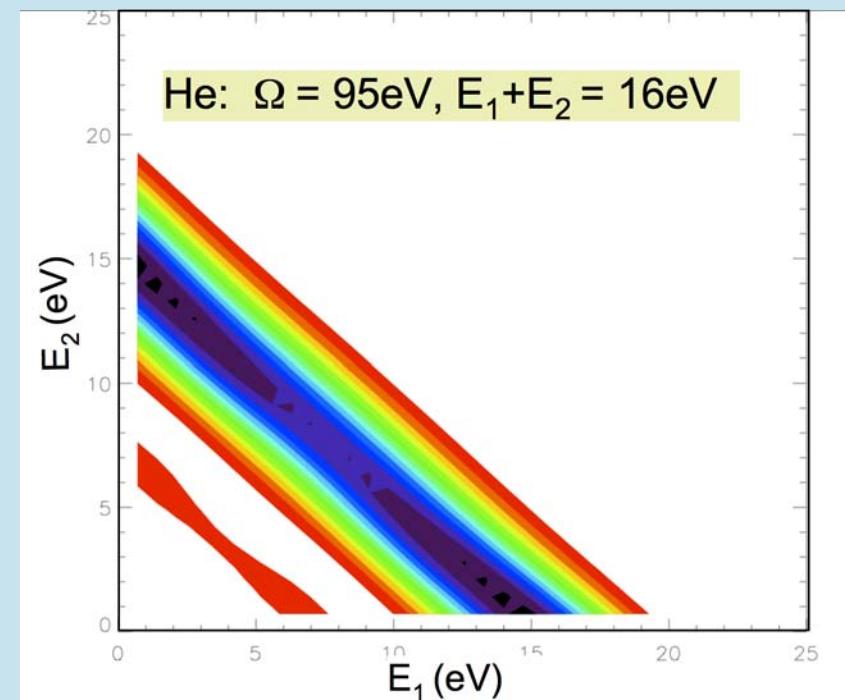
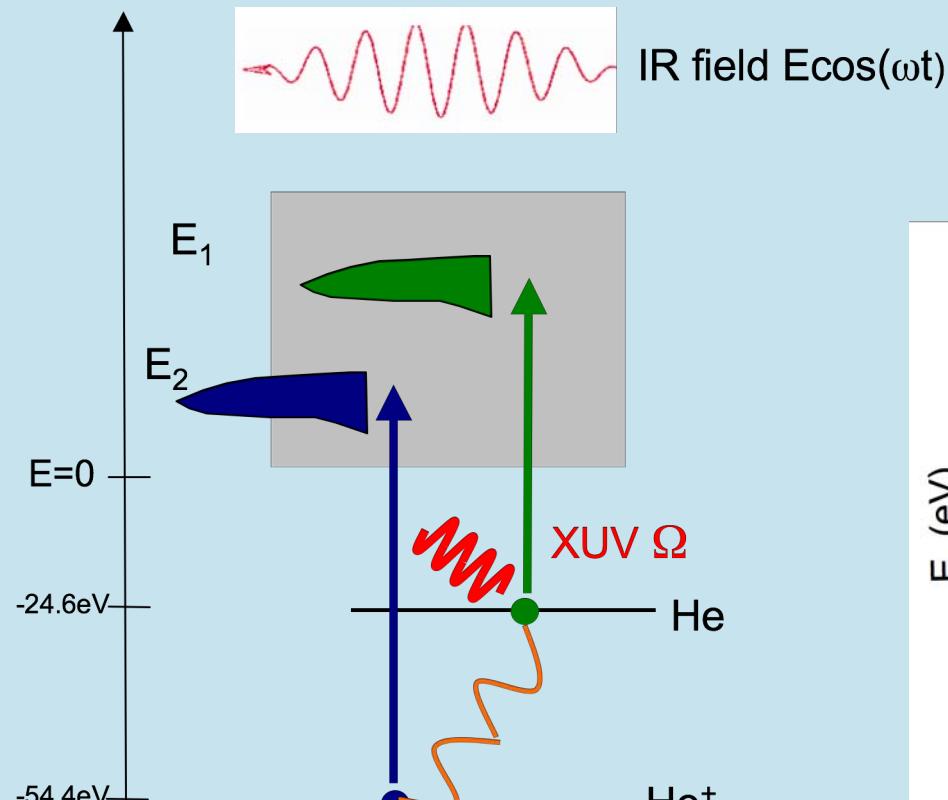


Photoelectron energy distribution in He XUV + IR

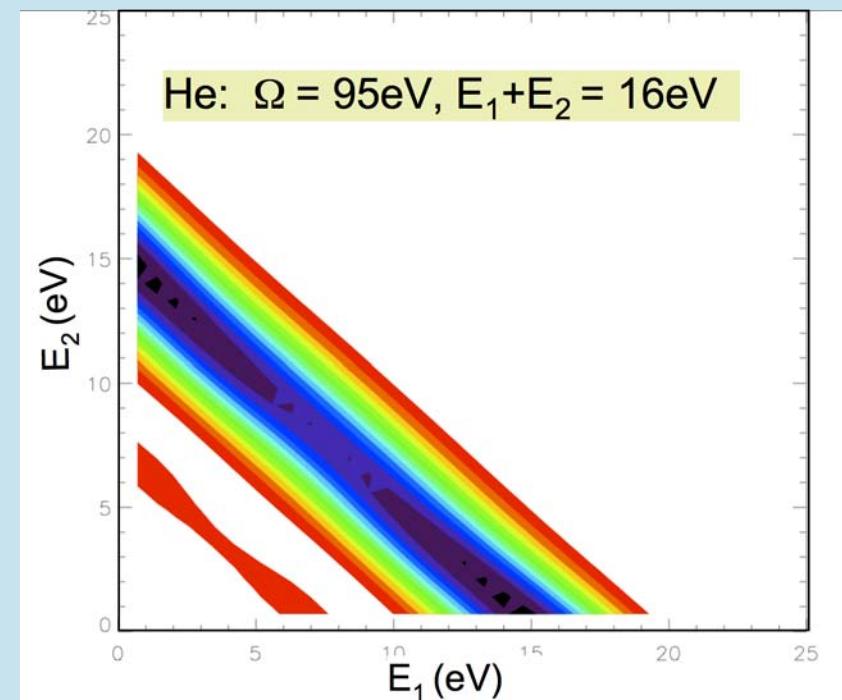
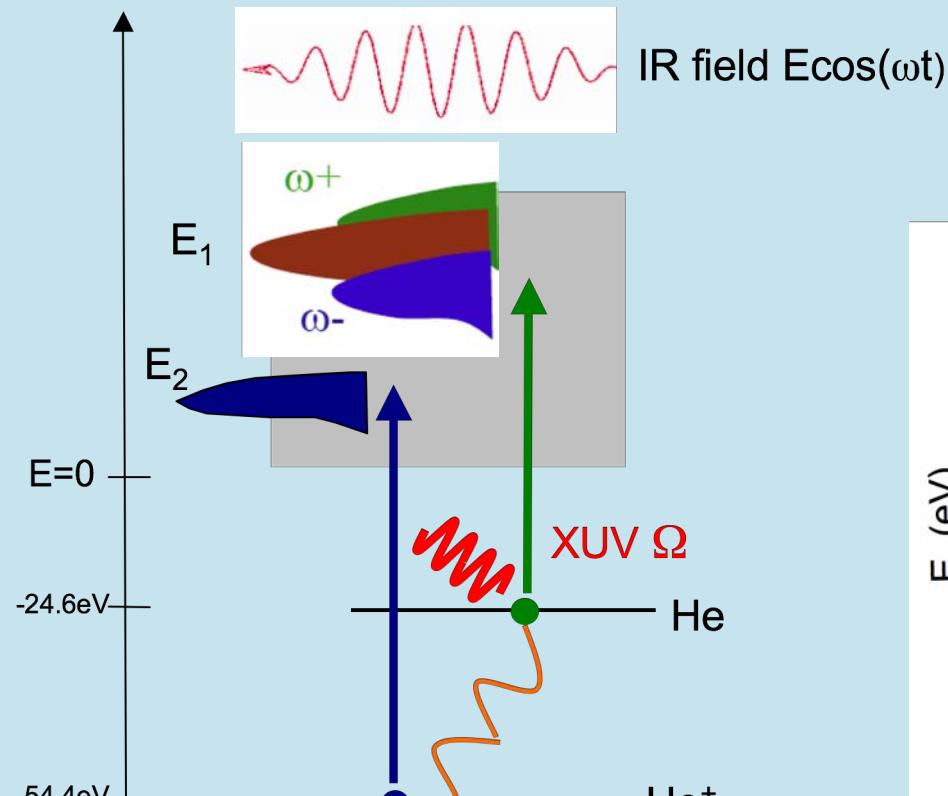


Photoelectron energy distribution in He

XUV + IR

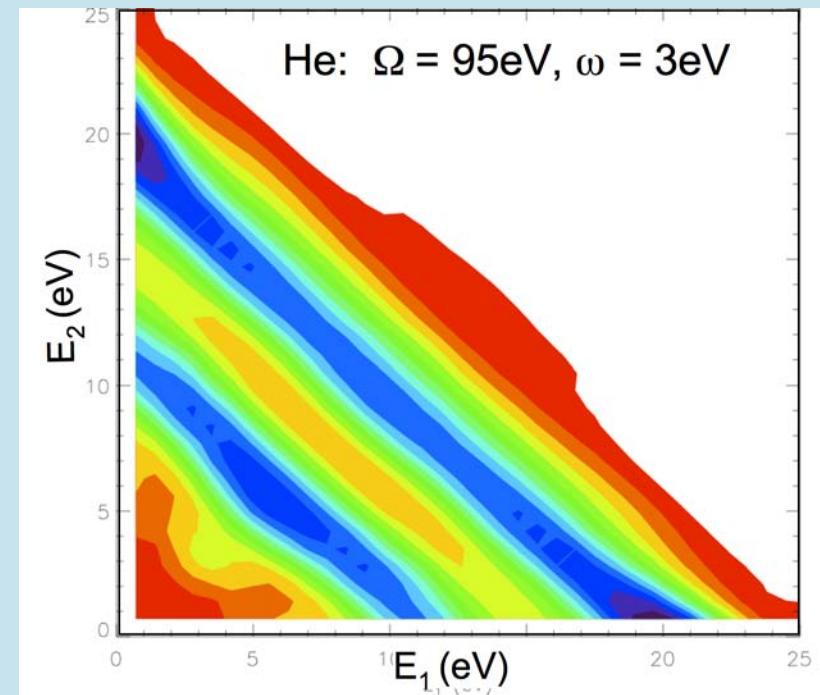
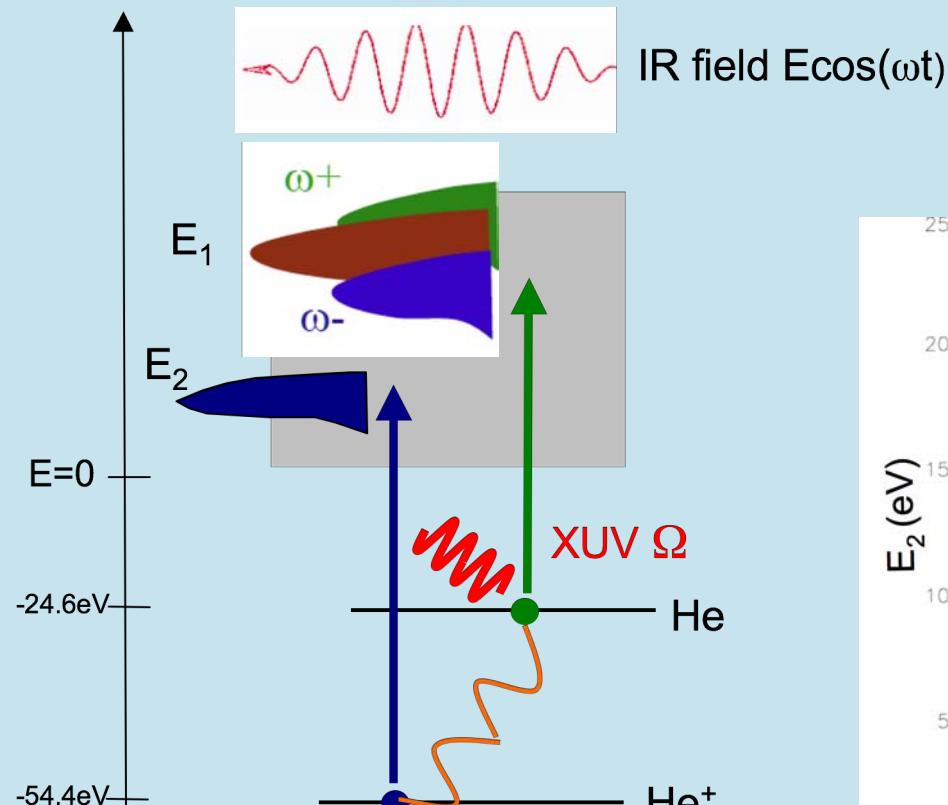


Photoelectron energy distribution in He XUV + IR



Photoelectron energy distribution in He

XUV + IR



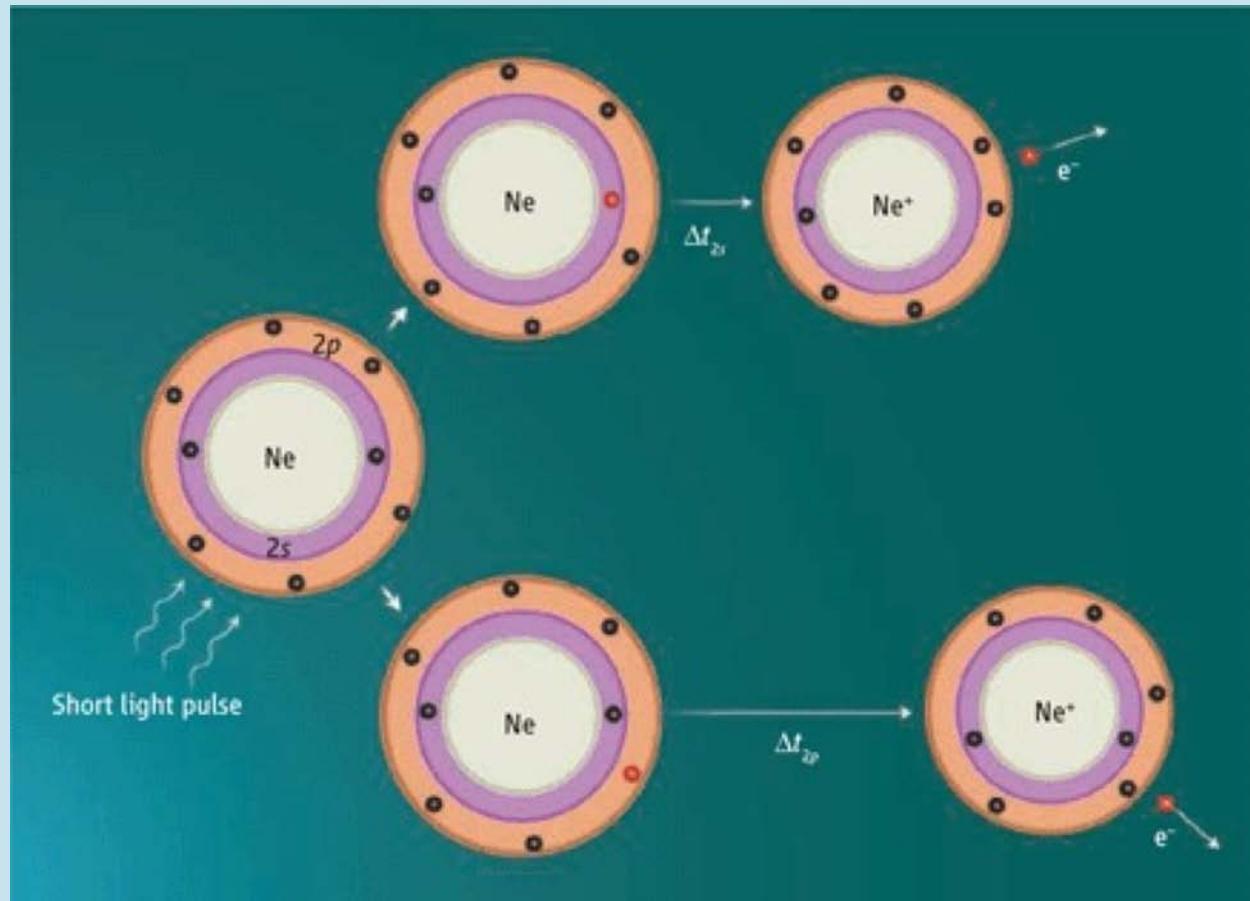
When Does Photoemission Begin?

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sub-10-as regime has been enabled by waveform-controlled near-single-cycle pulses (duration ~3.3 fs) of near-infrared (NIR, carrier wavelength ~750 nm) light (26). These pulses permit the generation of isolated sub-100-as extreme ultraviolet (XUV) pulses via high-order harmonic generation

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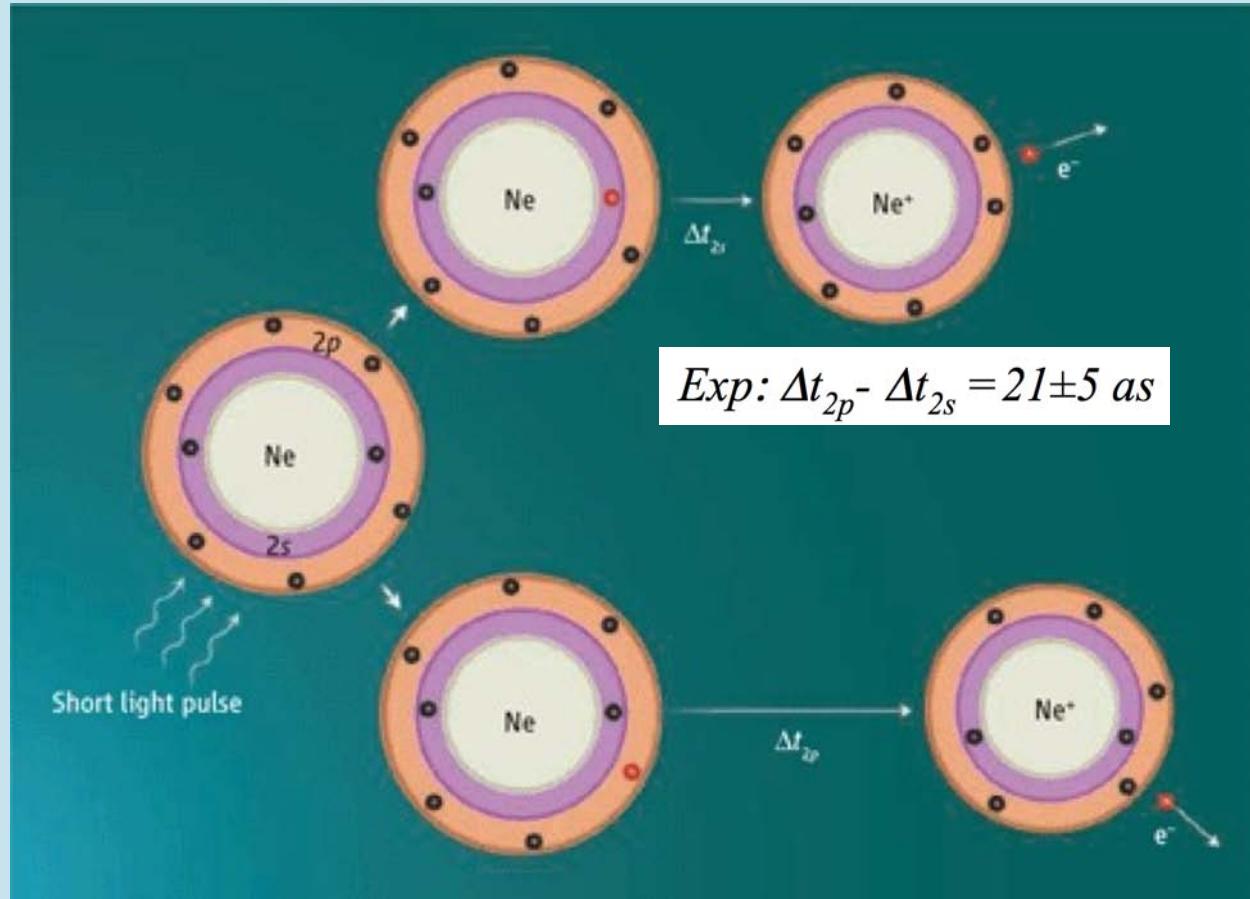


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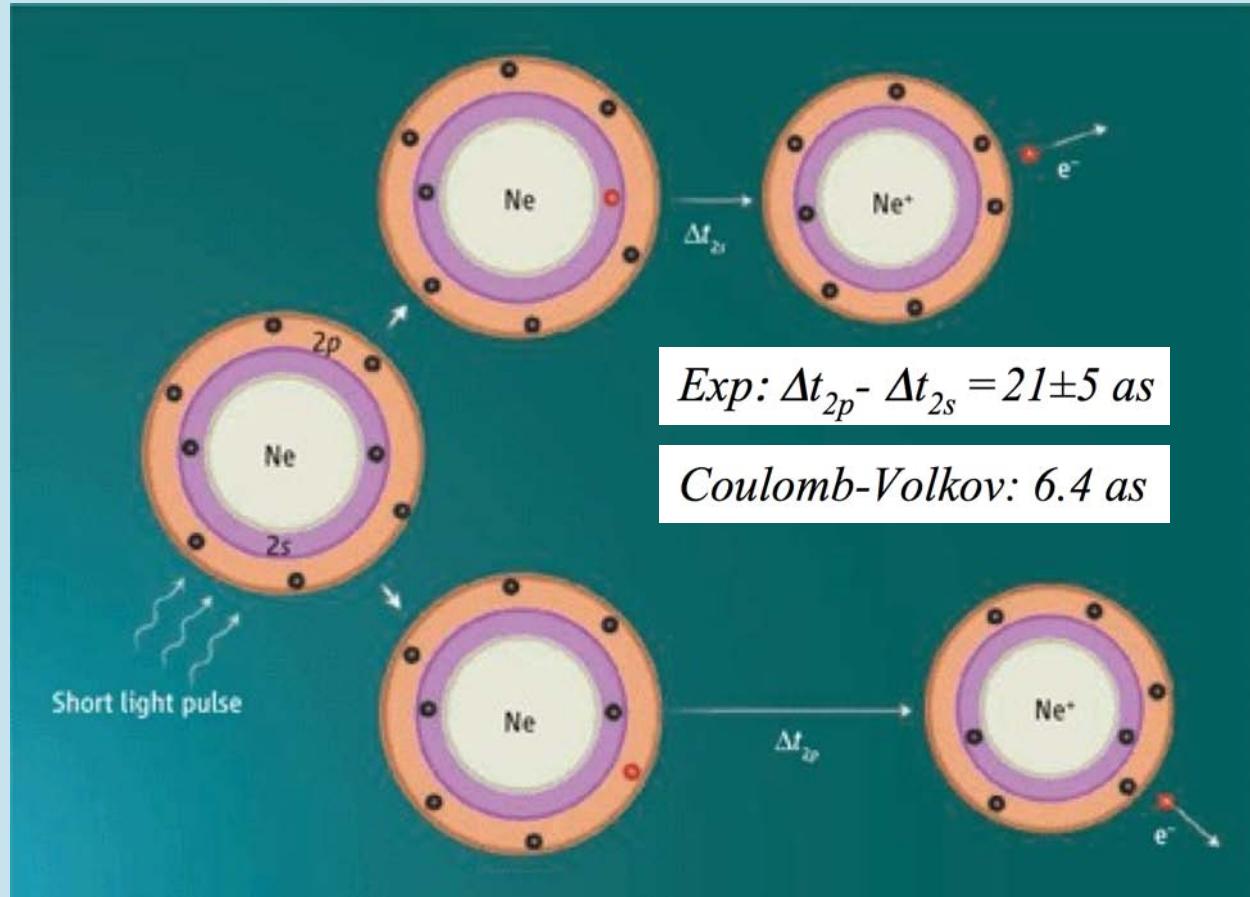


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 - $(N\gamma, e)$ on H
 - $(4\gamma, e)$ on Li
 - Electron correlation resolved in time
 - Double ionization of He
- Further directions
 - Single ionization of noble gases